



Nano-scale Noise Spectroscopy for Materials Applications

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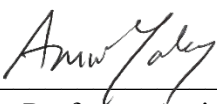
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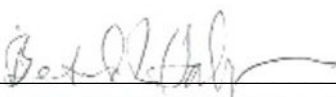
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Nano-scale Noise Spectroscopy for Materials Applications

A DISSERTATION PRESENTED

BY

RUOLAN XUE

TO

JOHN A. PAULSON SCHOOL OF ENGINEERING AND APPLIED SCIENCES

IN PARTIAL FULFILLMENT OF THE REQUIREMENTS

FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY

IN THE SUBJECT OF

APPLIED PHYSICS

HARVARD UNIVERSITY

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Nano-scale Noise Spectroscopy for Materials Applications

ABSTRACT

The rise of two-dimensional (2D) materials has opened new frontiers for exploring low-dimensional physics. These systems exhibit distinct electronic and spin behaviors compared to their 3D counterparts, and offer tunability through stacking, twisting, and thicknesses control. Yet, their micrometer-scale lateral size and atomic-scale thickness pose a major challenge for conventional condensed matter probes, calling for highly sensitive, local measurement techniques.

In parallel, quantum information science has also made exciting technological advancements in the past decades. Quantum sensing, built on the precise control of coherent quantum systems, has become a powerful tool in precision metrology, biosensing, and condensed matter physics. Among various platforms, nitrogen-vacancy (NV) centers in diamond, originally developed for quantum networks, have emerged as a highly sensitive, non-invasive nanoscale probe for studying spin and charge dynamics.

NV centers have already proven their value through pioneering studies of magnetic and electronic properties in quantum materials, often cross-validated by optical spectroscopy and electronic transport. These early efforts paved the foundation for using NV magnetometry as a standalone probe to uncover new physics. With a mature understanding of device fabrication, spin-sample coupling, and coherent control techniques, NV-based sensing now accesses previously unreachable regimes of spatial, spectral, and temporal resolution in condensed matter systems.

This thesis is driven by the question: *What makes NV centers uniquely powerful as magnetometers for condensed matter research?* Specifically, how do their quantum coherence and sensitivity in momentum and frequency space enable access to new physical phenomena?

The thesis presents work that spans both validation and discovery. First, we used NV AC magnetometry to measure the magnetic penetration depth in the 2D cuprate superconductor BSCCO, obtaining values and temperature scaling consistent with established results. This part is discussed in Chapter 5. More significantly, we uncovered a new regime of spin transport in atomically thin Heisenberg ferromagnets: *magnon hydrodynamic transport*. Through the coherent spin response of NV centers, we directly observed magnon second sound—an analog of collective density wave in fluid. This work also expands the experimental toolkit for studying fluctuation-dissipation phenomena in low-dimensional magnets with spin decoherence properties.

This thesis begins with an overview of quantum sensing with NV centers and recent advances in understanding 2D magnetic materials. I then introduce NV noise spectroscopy—the central technique enabling the discovery of magnon hydrodynamics. Chapters 3 and 4 present experimental observations and theoretical insights into magnon hydrodynamic transport. Finally, Chapter 5

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summarizes our independent BSCCO measurements and ongoing efforts toward wide-field NV methodologies, providing a complete record of our contributions to this emerging field.

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Citations to Previously Published Work

Some of the figures and a portion of the text in Chapter 1 to 4, and Appendix 7.2 have appeared in the following paper, with co-authors listed below:

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Introduction

We are in an exciting era where rapid developments in quantum technology may profoundly impact computing, communication, and sensing technologies. Among the various areas of quantum science and engineering, quantum sensing is one of the earliest that find achieving cross-field applications and deliver unique values with the probes' quantum nature. In my PhD thesis, I focus on combining quantum sensing methodologies with novel material systems to explore electronic and spin properties at the microscopic level. I hope my PhD work has contributed to advancing both

our understanding of spin properties in novel materials and the development of quantum sensing methodologies.

0.1 A NATURAL SENSOR IN DIAMOND

Various quantum systems have been developed and extensively studied as qubits, including well-established superconducting qubits; atoms, ions, and molecules trapped in optical lattices; spin defects in wide-bandgap materials; semiconductor spin qubits and emerging topological qubits which are still in their infancy. Among these platforms, solid-state defects are a rapidly evolving branch, particularly promising for quantum networks and sensing applications.

The negatively charged Nitrogen-vacancy (NV^-) centers in a diamond lattice are one such example of naturally occurring defects. These centers form when a nitrogen atom substitutes for a carbon atom in the diamond lattice and pairs with a nearby ionized vacancy. The resulting defect has two electrons to form spin triplet ground state, that shows remarkable optical and quantum properties, are usually referred as NV centers for the simplicity.

The NV^- center electronic state can be optically excited. Upon relaxation, this electron has spin state dependent possibility to emit red fluorescent photons, a signature property of NV centers⁷⁴. This phenomenon also explains why diamonds sometimes exhibit a pink hue—when the concentration of NV centers dominates over other color-defects.

Spin structure and 2-level system Negatively charged NV centers (NV^-) have a spin triplet ground state, with a zero-field splitting (ZFS) of approximately 2.87 GHz between the $m_s = 0$ and $m_s = \pm 1$ states⁷⁶. This splitting is slightly sensitive to temperature and lattice strain⁴⁰. Applying a bias magnetic field further separates the $m_s = \pm 1$ states by $\pm \gamma B$, where the gyromagnetic ratio is $\gamma = 28 \text{ GHz/T}$. Microwave fields resonant with the energy difference between m_s states enable selective transitions. In the rotating frame approximation one can effectively approximate the system

of $|m_s = 0\rangle$ and $|m_s = (\pm)1\rangle$ states as a two-level quantum system on a Bloch sphere. This framework provides a convenient way to visualize spin dynamics under specific pulse sequences and will be used in the later discussions.

Optical readout of the spin states The optical properties of the NV center provide crucial access to its spin quantum state, forming the basis for optically resolved quantum sensing and imaging. Due to the existence of a wide side phonon band⁸¹, the NV^- electron can be excited from the ground triplet state (^3A) to the excited triplet state (^3E) using continuous laser light in the 450–637 nm range. This off-resonant optical excitation also emits phonon and preserves the electron spin state.

Upon relaxation from ^3E to ^3A , two decay channels are possible: (1) direct radiative decay, which emits a fluorescence photon, and (2) non-radiative decay via intermediate spin singlet states, which also leads to spin initialization. For electrons in the $m_s = \pm 1$ states, there is a $\sim 30\%$ chance of transitioning to the singlet state ^1A , whereas this transition is largely forbidden for $m_s = 0$. As a result, the likelihood of red photon emission reflects the spin state, providing up to 30% optical contrast. Additionally, the decay through the singlet pathway tends to return the spin to the $m_s = 0$ state, enabling optical spin initialization.

This unique spin-dependent relaxation mechanism allows for optical readout of the qubit spin state with exceptional resolution, including single-spin detection. Compared to traditional electron paramagnetic resonance (EPR), which measures ensembles of unpaired electron spins in materials-ensemble using long-wavelength microwaves, NV centers offer vastly improved spatial resolution and sensitivity, making them indispensable for high-precision quantum technologies.

Coupling to nuclear spins The electron spin naturally couples to the nuclear spin of the nitrogen atom in the NV center through hyperfine interaction. Depending on whether the nitrogen isotope is ^{14}N or ^{15}N , each electron spin state is split into 2 or 3 energy levels, corresponding to the nuclear spin state⁴². The orientation of the nitrogen nuclear spin is fixed by its quadrupole axis,

which does not co-evolve with the NV center’s electron spin. The hyperfine coupling strength is between 3 and 5 MHz, which is very small compared to the NV center’s zero-field energy level splitting (2.87 GHz). Although we can select a specific nuclear spin state to work with, in practice, for driving Rabi and pulsed control of NV centers, we use strong microwave power to address the 2 or 3 states simultaneously. As a result, we expect to observe nitrogen nuclear spin hyperfine-induced oscillations in Ramsey and other measurements.

Good coherence of NV The qubit state in NV centers exhibits excellent coherence over a wide temperature range, making them highly effective for quantum sensing applications. The spin coherence times (T_2^* and T_2) of a single NV center typically range from hundreds of nanoseconds to milliseconds, while the spin relaxation time (T_1) can extend from milliseconds to several seconds⁵. This exceptional coherence and long lifetime allow the NV spin state to interact with and accumulate information from external magnetic signals over an extended period, which can then be optically read out.

The magnetic field sensitivity of NV centers, fundamentally limited by their coherence time, is discussed in detail in a review paper⁵. For shallow-implanted NV centers (depth < 5 nm) used in this thesis, the DC magnetic field sensitivity is typically on the order of several $\mu\text{T}/\sqrt{\text{Hz}}$, while AC magnetometry using dynamical decoupling sequences can achieve sensitivities in the tens of $\text{nT}/\sqrt{\text{Hz}}$ range⁵. NV centers also operate effectively over a broad temperature range—from millikelvin to room temperature. Compared to other quantum systems such as superconducting qubits, NV centers offer the unique advantage of coherent operation at ambient conditions.

The formation of NV Although NV centers naturally occur in diamond, controlling their quality, depth, and density is critical for experimental purposes. To achieve this, lab-grown electronic-grade diamonds and ion-implantation techniques are commonly employed³⁶. During implantation, high energy nitrogen atoms replace carbon atoms and create vacancies in the diamond lattice. After implantation, the formation of NV centers relies on thermodynamic processes, particularly at ele-

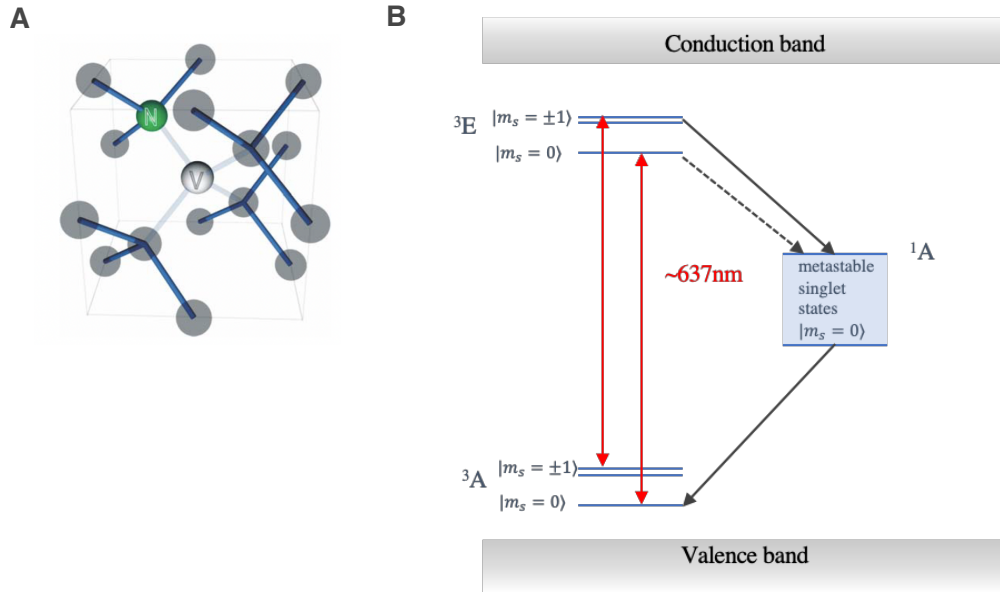


Figure 1: NV center in diamond lattice and its energy structure. The energy levels of an NV center lie deep within the diamond bandgap (~ 5.5 eV). The optical transition between the electronic states 3A and 3E corresponds to a wavelength of approximately 637 nm. These direct excitation and decay processes are spin-conserving. In contrast, a non-radiative relaxation pathway exists via metastable singlet states, which predominantly affects the $|m_s = \pm 1\rangle$ sublevels of 3E and ultimately relaxes the spin into the $|m_s = 0\rangle$ ground state.

vated temperatures. Heating the diamond lattice above 1000°C in vacuum enhances atomic mobility, promoting the pairing of nitrogen atoms with vacancies. This pairing is energetically favorable compared to the separated nitrogen and vacancy states, ensuring the stability of NV centers under such conditions³⁶. The common NV formation probability is 5-10% of the implanted N density.

The orientations of NV centers align with the direction of the carbon-carbon bonds in the diamond lattice. Therefore, the orientations of the formed NV centers relative to the surface of the diamond chip depend on the cut of the diamond. In this thesis, as well as in most cases, diamonds are cut along the $\langle 100 \rangle$ direction, where NV centers exhibit four different orientations, all pointing at a 54.5° angle from the $\langle 100 \rangle$ surface. A less commonly used cut is along the $\langle 111 \rangle$ direction, where one of the NV orientations is perpendicular to the diamond surface, making experimentally aligning magnetic field to the NV orientation easier.

0.2 SENSING PROTOCOL AND SENSITIVITIES

The NV center, a qubit with an $S = 1$ spin state, couples to the environmental magnetic field via the Hamiltonian:

$$\hat{H}_{\text{NV}} = D_{\text{zfs}} \hat{S}_z^2 - \gamma_{\text{NV}} \mathbf{B}(t) \cdot \hat{\mathbf{S}}, \quad (1)$$

where D_{zfs} is the zero-field splitting of the NV center, and γ_{NV} is the electron spin gyromagnetic ratio, approximately 28 GHz/T. The second term describes the coupling of the NV spin to any magnetic signal, including fields generated by currents or materials. By comparing the spin's final state to its initial state, one can extract information about the time-varying magnetic field, which will be detailed in the following sections.

DC MAGNETOMETRY

In the presence of a magnetic field, energy level of spin states undergo Zeeman splitting. The simplest method to measure the magnetic field is by determining the transition frequency using optically detected magnetic resonance (ODMR), also known as electron spin resonance (ESR), through sweeping the drive field frequency. Alternatively, the splitting can be inferred from the phase accumulated during spin precession in a superposition state, a technique known as Ramsey spectroscopy. The fundamental sensitivity limit for both methods is set by the spin dephasing time, T_2^* , which corresponds to the intrinsic linewidth of ESR spectrum. In ESR measurements, additional factors such as laser and microwave power broadening further influence sensitivity.

AC FIELD AND NOISE SPECTROSCOPY

AC magnetic field Ramsey spectroscopy can measure a magnetic field by tracking the phase evolution of a superposition state $\phi = \gamma B_{\parallel} \tau$. AC magnetometry has a similar idea as measuring phase accumulated at the superposition under time-varying magnetic field. By inserting pi-pulses periodically into a Ramsey sequence, one can dynamically decouple the NV spin from static magnetic fields. Only magnetic fields which change sign over the dynamical decoupling sequence produce a phase evolution^{14,8}.

This spin-locking technique leverages the extended precession time, also called spin coherence time, T_2 , which can be extended by tens to a hundred times compared to T_2^* . If the signal is modulated at the same frequency as the π -pulse sequence, akin to a lock-in measurement in transport experiments, it becomes possible to extract low-frequency (typically tens of kHz to tens of MHz) modulated fields with the pulse sequence called spin-echo and its higher-order sequences¹⁹.

NV centers can also measure coherent AC fields, using techniques such as Rabi oscillation and AC Stark effect⁹⁴. While this type of signal is not the primary focus of my graduate study, these methods are powerful tools for sensing and material characterization.

Noise spectroscopy In contrast to coherent fields, magnetic fields that vary in amplitude or phase between repeated dynamical decoupling measurements are usually treated as noise that would need to be minimized and separated from the signal. However, magnetic noise that originates from spin or current excitations, gives information about the spin or current spectral function $S_m(\mathbf{k}, \omega)$ of the material. A spin system's response function can be probed by measuring the magnetic field noise in the vicinity of the surface of the sample. When NV is placed into local environment with magnetic noise, the randomness of the field will bring variation $\langle \delta\phi^2 \rangle$ to the phase of NV state per shot, leading to decoherence of the state over many realization of the measurement. So by monitoring the coherence (T_2, T_2^*) or lifetime (T_1) of our qubit, one can extract information of magnetic

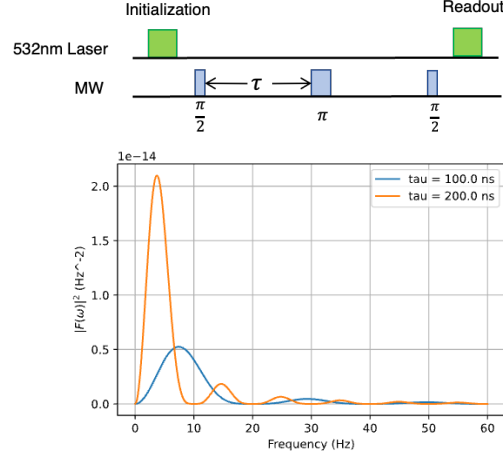


Figure 2: Spin echo sequence and filter functions. The spin echo pulse sequence is shown alongside examples of the corresponding filter functions for precession times $\tau = 100$ ns and $\tau = 200$ ns.

noise of NV's local environment. The frequency bandwidth of the noise probed by the NV center is set by the measurement sequence used.

The spin-echo filter function is centered at frequencies given by the precession time τ (Fig. 2). The peak in the frequency filter indicates the most sensitive frequency segment to which the spin echo sequence is subject. The filter function is the Fourier transform of precession window, that filters out magnetic field that is varying too fast or too slow. The optimum response frequency being $1/2N\tau$ for a sequence containing N π -pulses (higher order variation of spin echo, Carr-Purcell-Meiboom-Gill (CPMG) and XY-N) and the higher N gives sharper frequency selection. More commonly spin-echo and CPMG are used for the information in phase – to extract the amplitude of a varying magnetic field that has been locked-in to the pulse sequence. Here I note that the loss of phase coherence also has its own value in terms of discover new physics¹¹⁵ that will be elaborated in my graduate thesis.

The nature of NV-based noise sensing provides a unique probe for studying the low-frequency dissipation of materials through fluctuation-dissipation theorem. The typical frequency filter for echo-type sensing is in the kHz–MHz regime, while relaxation-type sensing extends to several GHz.

The corresponding energy scale, on the order of microelectron-volts (e.g., 1 GHz corresponds to 4 μeV), is too small to be resolved by traditional techniques such as neutron scattering. Nuclear magnetic resonance (NMR) serves as a bulk counterpart to NV measurements; however, the momentum filter function can differentiate these two methods. This distinction will be discussed further in the next section (0.2.1).

0.2.1 WAVELENGTH SELECTIVITY

In addition to frequency selectivity that is present in all types of NV measurements, NV centers exhibit selectivity in momentum space due to the standoff distance of the sensor. In a dipolar-coupled spin and sensor system, the distance d introduces a momentum filter factor ke^{-kd} (k is the wave-vector of the magnetic field), which peaks at approximately $1/d$. This implies that waves with much larger spatial frequencies than $1/d$ are effectively "averaged out" from the perspective of a sensor, rendering them undetectable. By adjusting d , different regions of momentum space can be probed^{92,16}. This unique capability of NV magnetometry distinguishes it from other techniques used to investigate the spectral density of materials, such as neutron scattering, nuclear magnetic resonance (NMR), and muon spectroscopy, which lack momentum selectivity and tunability.

It is then natural to expect that the distance dependence of the magnetic signal would vary for spin systems of different natures. This variation arises not only because the NV center probes different momentum slices at various distances, but also because the transport behavior being measured can change. For example, by comparing the scattering length scale to the standoff distance d , NV magnetometry can probe ballistic, diffusive, or hydrodynamic behaviors of electrons or spins on a material¹. This aspect has been extensively studied in theory^{79,17,1,24}, but there have been very few experimental demonstrations⁴⁹. Experimentally, controlling and precisely adjusting the NV-sample distance remains a significant challenge. However, this capability holds immense potential for uncovering new insights into material properties.

0.3 PROGRESS IN NV-BASED QUANTUM SENSING IN CONDENSED MATTER

The advancement of local probe techniques and the exploration of mesoscale physics are mutually reinforcing. As nano-materials emerge, they demand experimental tools that can resolve signals at similar or smaller length scales. In turn, these local probes enable new discoveries in the physics at such length scales. Among them, the NV center—whether deployed as a scanning probe or optical magnetic microscope—offers nanoscale access to magnetic textures and dynamics^{25,26}, superconducting order parameters, and current distributions^{50,93,16}.

The use of NV centers as magnetic probes has been explored for over a decade, offering several advantages over other magnetometers such as SQUID, muon spectroscopy, and NMR in the study of material physics. **First**, it provides a nanoscale microscope of magnetic signals. The intrinsic spatial resolution of NV centers is sub-nanometer due to their atomic-defect nature. Although current fabrication and instrumentation limit the practical spatial resolution of scanning NV microscopy to tens of nanometers⁶², NV centers remain superior for local studies, particularly in 2D materials where rich local physics arises from micron-scale sample sizes and widespread inhomogeneities in devices. **Second**, NV centers operate under remarkably broad conditions, from high vacuum to extreme pressures exceeding gigapascals²³, and from millikelvin to several hundred kelvin. Even under extreme conditions, NV sensors achieve sensitivities reaching $\text{sub-}\mu\text{T}/\sqrt{\text{Hz}}$ ⁷⁹, making them exceptional for diverse experimental setups. **Third**, as quantum sensors, NV centers enable multiple sensing modalities. Unlike classical spins, which measure magnetic fields via Zeeman splitting, quantum superpositions of $|0\rangle$ and $|1\rangle$ states allow for advanced sensing sequences and algorithms to suppress noise and extend measurable signal types. For example, spin echo can eliminate DC noise (spatial inhomogeneity), while techniques such as CPMG-N, XY-N, and T_1 -rho measurements extend the sensing frequency range from DC to several GHz, providing unmatched versatility. **Fourth**, NV centers can be used as a in-situ temperature, pressure, electric field sensor aside from being used as a

magnetometer.

0.3.1 MAPPING CURRENT PROFILES

Below I describe how the DC and AC sensing properties of both coherent and noise signals have previously been used to study the electron and spin transport processes in materials.

Viscous current One example to demonstrate the power of spatial imaging is the visualization of viscous current in micrometer-wide 2D channels^{50,93}, relying on the Biot-Savart law that convert current signal to magnetic field that can be measured by NV. The local probe with high spatial resolution can map out current profile that distinguishes viscous flow from ohmic flow^{50,93}. Beyond imaging current profile, NV centers can also probe Johnson noise of electron systems, revealing different electron transport regimes of ballistic, ohmic and even non-equilibrium transport regimes through the relaxometry^{49,111}.

Superconducting current Another major area of research focuses on imaging current profiles in superconductors (SCs). For example, NV magnetometry can detect Meissner screening currents in thin-film SCs under an external magnetic field, providing information about the London penetration depth λ_L across different temperatures⁹⁸ (see Chapter 5). NV sensors have also been instrumental in uncovering subtle effects in superconducting Josephson junctions, such as the diode effect and competing ground states that remain featureless in standard transport measurements. In these cases, the nanoscale spatial resolution of NV magnetometry offers insight into the local magnetic behavior, potentially guiding the design and optimization of superconductor-based quantum devices¹⁹. Additionally, NV imaging has enabled the direct visualization of vortices in type-II SCs, revealing detailed vortex structures and allowing for comparisons and confirmation with theoretical hypothesis⁹⁰.

0.3.2 MAGNETIC SYSTEMS

The static and dynamical properties of a magnet contain valuable information about the parameters entering spin Hamiltonian, and nature of its spin excitations. In-depth study of the magnetic properties can lead to advances of both fundamental knowledge and device application based on spin orientation and excitations.

Experimental tools for probing static magnetic properties include muon spin rotation (μ^+ SR), Superconducting Quantum Interference Devices (SQUID), Hall measurements, and torque magnetometry. To investigate magnetic excitations, such as spin waves (magnons) in ferromagnets and antiferromagnets or topological excitations like skyrmions and merons, well-established techniques such as nuclear magnetic resonance (NMR), neutron scattering, and microwave transmission/absorption are widely used. These excitations play a central role in advancing magnet-based computing²¹ and next-generation information storage devices⁵⁹, making their characterization a key area of interest in condensed matter physics.

However, the emergence of two-dimensional (2D) magnets presents challenges for the conventional experimental methods. The techniques mentioned above typically require substantial sample volumes to generate detectable signals, often necessitating single crystals or powdered samples. However, exfoliated 2D magnets are usually atomically thin with lateral dimensions of only a few micrometers, making signal detection challenging. As a result, new approaches, such as optical dichroic imaging and spectroscopy^{44,108}, as well as nanoscale magnetic probes¹⁰⁰, are needed for these studies. NV centers has been demonstrated as a great probe for the spin physics happening in both 3D and lower dimensions.

Static Magnetization. NV centers excel in imaging static magnetic structures with spatial resolution down to tens of nanometers. This capability enables quantitative determination of the magnetization in atomically thin ferromagnets CrI_3 by mapping the weak local stray fields generated by the

sample, detected through scanning single-NV magnetometry⁹¹. Notably, such quantitative magnetization measurements can be challenging using traditional magneto-optical dichroism techniques due to limited understanding of light–matter interaction mechanisms.

More intriguingly, by fully utilizing the atomic-scale spatial resolution of NV centers, one can resolve fine magnetic textures at the nanometer scale. For example, NV magnetometry has been used to image static skyrmions formed in ferromagnetic dots, enabling the reconstruction of their full spin structure and extraction of the topological order and chirality²⁵. Another notable study visualized nanoscale FM/AFM domain patterns modulated by moiré potentials in twisted CrI₃ bilayers, providing direct evidence for stacking-dependent interlayer coupling⁸⁴.

Magnons, also known as spin waves, are the elementary excitations in magnetically ordered systems. They are bosonic quasiparticles that can be thermally populated or driven by resonant microwave fields. When a nitrogen-vacancy (NV) center is placed in close proximity to a magnetic sample, its spin couples to the dynamics of local magnetic moments via dipolar interactions. For example, in yttrium iron garnet (YIG), low-frequency ferromagnetic resonance (FMR) modes can overlap with the NV spin transition, leading to enhanced spin relaxation of the NV qubit or coherent Rabi oscillations of the NV center^{26,64,92}.

Magnon dispersion is subject to the change of magnetic field. Generally speaking, applied magnetic field increases the Zeeman energy of spins $\delta E_{\text{Zeeman}} = \gamma \mathbf{B} \cdot \mathbf{S}$ that will increase the size of the magnon gap. Kittel model gives the predicted FMR of an isotropic ferromagnet to be $\omega_{\text{FMR}} = \gamma \sqrt{H(H + 4\pi M_s)}$. H is the applied bias field and M_s is the saturation magnetization of the material. NV energy level also changes with the bias field $\omega_{\pm} = 2.87\text{GHz} \pm \gamma B_{\parallel}$ linearly respond to the parallel field and higher order dependence for the perpendicular field component. The NV lower transition intersects with YIG FMR at a few hundred gauss and introduce enhanced relaxation to NV centers. The rich features in NV-YIG magnon hybride spectrum could contribute to parametric pumping, NV excited state relaxation and more physics to be studied and understood. The NV-YIG

magnonics is still a fast evolving field to date^{26,96}.

The thermal magnon gas in YIG can be populated by coherently driving magnons at FMR and scatters-off into the incoherent thermal bath. The increased magnon population is characterized by the chemical potential μ , which determines the magnon density according to the high-temperature approximation of the Bose-Einstein distribution:

$$n(\omega, \mu) = \frac{k_B T}{\hbar\omega - \mu}. \quad (2)$$

By measuring the change in the relaxation rate of NV transitions—induced by the increased density of thermal magnons—as a function of the coherent drive, one can extract the magnon chemical potential μ . The experimentally extracted saturation value of μ shows precise agreement with the theoretically predicted magnon band minimum²⁶. These results demonstrate that NV centers serve as local, non-perturbative probes for thermal magnon populations and chemical potential.

To advance these studies in the field of two-dimensional (2D) magnets, one promising direction is to probe magnon excitations in soft 2D magnetic materials, where the magnon gap typically lies in the gigahertz regime. This motivation led to our initial exploration of CrCl_3 .

Sub-gap spin fluctuations In a magnetic material, not only transverse spin fluctuations (magnons) generate magnetic signal, but longitudinal spin fluctuations (e.g., magnon scattering associated with momentum and energy transfer) can also induce additional relaxation or decoherence of NV centers. NV centers have been used to probe sub-gap spin transport properties in ferro- and antiferromagnetic materials^{95,29} across different transport regimes, including the diffusive and ballistic limits. Other regimes, such as the hydrodynamic spin transport regime, remain to be explored experimentally. This endeavor is one of the main subjects of this thesis

Fluctuations in Superconductor Another topic of interest is low-energy dynamics in unconventional superconductors. NV noise spectroscopy measurements on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (BSCCO)

capture magnetic fluctuations generated by quasi-particle excitations deep in the superconducting phase, as well as super-current fluctuations close to phase transition that deviate from predictions of BCS mean-field theory. Under small bias field, these measurements suggest the presence of vortex liquid-solid transition dynamics, a characteristic feature of type-II superconductors⁵⁷. Moving forward, NV centers are expected to be increasingly utilized for investigating the dynamics of low-energy, sub-gap spin dynamics and quasi-particle excitations.

Overall, NV centers offer powerful capabilities for material characterization due to their unique broadband frequency sensitivity and momentum selectivity. Numerous studies have demonstrated the versatility of NV magnetometry in probing both static and dynamic properties of spins and electrons. Moving forward, we anticipate further discoveries of novel physical phenomena using this quantum probe, as well as similar quantum defects in two-dimensional materials (e.g., hBN, SiC) and even molecular systems.

"Now I have a lab with expensive toys to play with."

Amir Yacoby

1

A 2D Heisenberg Ferromagnet

A PHYSICIST'S INTUITION MIGHT SUGGEST THAT A 2D HEISENBERG FERROMAGNET CANNOT EXIST IN PRACTICE. This was also my impression from Mermin-Wagner theorem before beginning my research on the two-dimensional magnetic material CrCl_3 during my PhD. In this chapter, I will review the relevant literature and present our experimental calibration and theoretical modeling on the static magnetization of CrCl_3 , shedding light on our understanding of this intriguing and

controversial magnetic material.

1.1 BACKGROUND: 2D MAGNETS AND CrCl_3

Two-dimensional (2D) magnets are exfoliated from magnetic materials with layered lattice structure in which the interlayer coupling is of van der Waals nature^{38,83}. The measurement of spin polarization in these atomically thin samples has sparked interest in the physics and applications of 2D magnets, making it a popular topic of research. Compared to bulk magnetic materials, 2D magnets offer several advantages, including easier integration with other materials to form heterostructures and the potential to enable highly efficient spintronic devices⁸³.

The Mermin-Wagner theorem states that continuous symmetry cannot be spontaneously broken at non-zero temperatures in systems with dimensionality $d \leq 2$. However, in practice, there has been experimental observations of long-range order in atomically thin 2D ferromagnets. The existence of long range order has been explained by the presence of anisotropy in the crystal, arising from various mechanisms, including spin-orbit coupling (SOC), which leads to magnetocrystalline anisotropy, dipole-dipole interactions associated with shape anisotropy, or anisotropic exchange coupling.

$$H = -J \sum_{\langle ij \rangle} (S_i^x S_j^x + S_i^y S_j^y + \alpha S_i^z S_j^z) - g\mu_B \sum_i (B_z S_i^z + B_x S_i^x) + H^{\text{dip}}. \quad (1.1)$$

An example spin Hamiltonian is shown in Eq. 1.1, describing a monolayer chromium trihalide. The exchange coupling J is ferromagnetic for all three compounds CrI_3 , CrBr_3 , CrCl_3 (see table 1.1). Due to the hexagonal symmetry of the lattice, the spin exchange couplings are effectively isotropic within the plane, rather than imposing C_3/C_6 symmetry⁴⁵. The anisotropy in the exchange interaction is captured by the parameter α in front of the $S_z S_z$ term, which primarily orig-

inates from the ligand spin-orbit coupling (SOC)⁸⁵. In the cases of CrI₃ and CrBr₃, the relatively large ligand SOC stabilizes spins out-of-plane (easy-axis anisotropy), which accounts for the ferromagnetism observed in the monolayer forms of these materials^{38,112}.

CrX ₃	J(meV)	α	anisotropy type
Cl	1.7	-0.02	easy plane(xy)
Br	2.4	0.07	easy axis(z)
I	2.7	0.30	easy axis(z)

Table 1.1: Exchange Energy(J) and magnetic anisotropy(α) theoretical values for CrX₃(X=I, Br, Cl)^{99,106}

CrCl₃ exhibits a more complex magnetization behavior. The existence of long-range order in its monolayer form is less straightforward due to the negligible out-of-plane anisotropy ($\alpha \approx 0$). The spin Hamiltonian of CrCl₃ closely resembles that of a Heisenberg ferromagnet. From the bulk down to the bilayers, experimental evidence indicates in-plane ferromagnetic intralayer order and antiferromagnetic interlayer coupling^{65,15}. Monolayer CrCl₃ was first stabilized on a Graphene/6H-SiC(0001) substrate, where it was reported to exhibit a Berezinskii-Kosterlitz-Thouless (BKT) phase transition at temperature range very close to $T_c = 13$ K. Below phase transition temperature, hysteresis and finite X-ray magnetic circular dichroism(XMCD) signal at zero bias field is always observed, that suggests at least local order exists in the measurement region⁷. At the same period, due to the in-plane magnetic nature and low anisotropy, several studies^{85,58,3} suggest that CrCl₃ behaves as a 2D-XY ferromagnet, supporting vortex-like topological spin excitations and enabling two-dimensional superfluid spin transport below T_c . The theoretical work appears to contradict the existing experimental data.

My PhD research may also shed light on the understanding of the magnetism of the low temperature phase of CrCl₃. This will be discussed in details in Section 1.5.

1.2 SPIN DYNAMICS

Recent studies have shown that in the CrX_3 family, where spins reside on a honeycomb lattice, the magnon band structure can exhibit Dirac-like dispersion⁷¹. Introducing Dzyaloshinskii-Moriya (DM) interaction on top of the Dirac magnon band can induce nontrivial topology in the magnon spectrum. This interaction opens a gap, creating topologically nontrivial magnon bands that support magnon edge states, enabling topological spin transport^{18,107}. However, unlike electrons, which are fermions and obey the Pauli exclusion principle, magnons are bosons and can occupy the same state within a band. As a result, it is impossible to tune the magnon chemical potential to the Dirac point. Direct measurement of the gap topology around the Dirac point is therefore very challenging.

On the other hand, the low-energy spin dynamics remains interesting and has potential for spintronic applications. When carefully reviewing the energy of magnons in Chromium Tri-halides, CrI_3 and CrBr_3 exhibit a low-energy magnon gap in the tens to hundreds of GHz range¹⁰⁸. In contrast, CrCl_3 , due to its low magnetic anisotropy, can even hosts magnons in the GHz regime. These magnon modes in CrCl_3 are easily accessible through microwave excitation and manipulation⁶¹.

The spin dynamics of exfoliated few-layer CrCl_3 has not been extensively studied, primarily due to the limited sample size and weaker signal intensity compared to its bulk counterpart. However, few-layer samples may exhibit different magnetic behaviors due to changes in anisotropy and interlayer-stacking-induced interactions⁴⁸. NV centers can serve as local and sensitive probes for exploring magnetism in exfoliated 2D magnetic samples.

Beyond magnonic excitations, CrCl_3 being the two-dimensional magnets with in-plane spin orientation and approximately isotropic spin interaction is also a promising platforms for studying spin superfluidity⁶⁶.

1.3 SAMPLE FABRICATION

With the material of interest being exfoliated few-layer CrCl_3 down to the monolayer limit, this section describes how we integrated the sample with the NV magneto-microscope in our experiment.

To integrate layered materials with NV centers in diamond, we adopted the shallow-ensemble sensing method and transferred 2D magnets directly on diamond surface. We used 2mm*2mm E6 electronic-grade diamond and implanted NV centers at a depth of approximately 50 nm using 25 keV implantation energy (NV depth and density characterization is provided in Appendix 7.2). A Ti/Au coplanar waveguide was then fabricated on the diamond surface for on-chip microwave delivery (fabrication details in Appendix 7.1). The coplanar waveguide gap is 30/50 μm . Compared to a single stripline, the coplanar waveguide provides a relatively uniform driving field within the gap region, ensuring that the spatial scan of the measurement spot is not significantly affected by variations in the Rabi rate.

CrCl_3 samples are encapsulated on both sides with freshly exfoliated, unannealed hBN. The choice of hBN as the encapsulation material is based on the fact that graphite would quench photons injected into and emitted from the diamond substrate, effectively reducing the detected photon counts. Additionally, the hBN used for encapsulation is not annealed or heat-cleaned, as annealing can introduce fluorescent defects in hBN (e.g., carbon-related defects, boron vacancies), leading to increased background photon counts.

hBN- CrCl_3 -hBN stacks were assembled inside a glovebox using freshly exfoliated (< 2 hours) CrCl_3 flakes. Since CrCl_3 is highly sensitive to moisture. Monolayers, in particular, can degrade within a few hours after exfoliation (being hard to pickup) and disappear for overnight, even when stored in a glovebox. Fabrication details are provided in Appendix 7.1.2.

After encapsulation, the stack was transferred onto the diamond surface using polycarbonate

(PC), which was subsequently removed by immersing the entire chip in chloroform in atmosphere. The sample was then loaded into the measurement setup. No significant degradation was observed in the hBN double-sided encapsulated sample after 30 minutes to 1 hour of ambient exposure, although longer exposure effects remain to be investigated. The samples were measured at cryogenic temperatures (4–30 K) under high vacuum (3×10^{-7} Torr) for months to years without noticeable change in their behavior, suggesting long-term stability under these conditions.

1.4 STATIC FIELD MEASUREMENT

1.4.1 MEASUREMENT CONDITIONS

Experimental Setup Optical pulses are provided by a 532 nm laser modulated by an acousto-optic modulator (AOM) and focused onto the diamond surface through a home-built confocal system. (P.S. Partway through this experiment, we upgraded the 532nm source to a Cobolt diode laser that can be pulse-controlled at nanosecond scale and by-passed the AOM). The time-averaged power of the focused laser is maintained between 100 and 150 μ W. A static bias magnetic field is applied using a permanent magnet mounted on a piezoelectric positioning stack (Attocube). NV fluorescence is detected using an avalanche photodiode with a 550nm longpass filter. Microwave pulses for NV spin control are generated by a Rohde & Schwarz SGS100A microwave source with built-in IQ(in-phase and quadrature) mixing, amplified by a 25 W microwave amplifier (Amplifier Research 25S1G6), and delivered to the coplanar waveguide. Pulse timing for all instruments, including IQ channels, is controlled by a Quantum Machines OPX1.

NV measurements require an external bias field to lift the degeneracy of the $|m_s = \pm 1\rangle$ energy levels. The minimum DC field that can be applied using an in-chamber permanent magnet (K&J D36-N52) is 2 mT, when the magnet is retracted to its furthest position. The applied field is oriented along the sample normal and at 54° to all NV orientations. The perpendicular field can

be continuously tuned up to 8 mT; beyond this, the NV contrast decreases below 2%. Since the measurement time scales quadratically with the NV contrast, this contrast reduction significantly impacts data acquisition efficiency. The drop in contrast is likely due to off-axis bias field-induced level mixing^{89,35}.

The maximum field achievable with the permanent magnet is 140 mT. To optimize NV contrast and coherence time (T_2), the field must be aligned with one of the four NV orientations⁸⁶. The magnet position is constrained by the range of attocube stages, allowing field alignment with NV orientations only within the 30–140 mT range. Unless stated otherwise, in the following discussion, applied fields above 30 mT are assumed to be at 54° to the material surface normal, while low fields (2–10 mT) correspond to an out-of-plane configuration.

We also spent time and effort to replace the permanent magnet with a home-machined and wired superconducting coil with a ferrite core, which would allow more flexible field sweeping and bidirectional field control. However, the coil failed to maintain superconductivity at the base temperature of the cryostat. This issue is suspected to arise from insufficient cooling due to limited thermal links and excessive wiring layers.

DC field measurement The static magnetic field at the sensing spot is measured via optically detected magnetic resonance (ODMR) or Ramsey spectroscopy. Both methods are sensitive to the magnetic field component projected along the NV axis, which determines the level splitting between the NV spin states $|0\rangle$ and $|-1\rangle$:

$$\omega_{0 \rightarrow -1} = \gamma(\mathbf{B}_{\text{NV}}^{\text{bias}} + \mathbf{B}_{\text{NV}}^{\text{sample}}), \quad (1.2)$$

where $\gamma = 28 \text{ MHz/mT}$ is the NV gyromagnetic ratio, and the total field consists of contributions from both the sample and the applied bias field. By spatially mapping $\omega_{0 \rightarrow -1}$, the stray field from sample along the NV axis can be isolated from the background applied field.

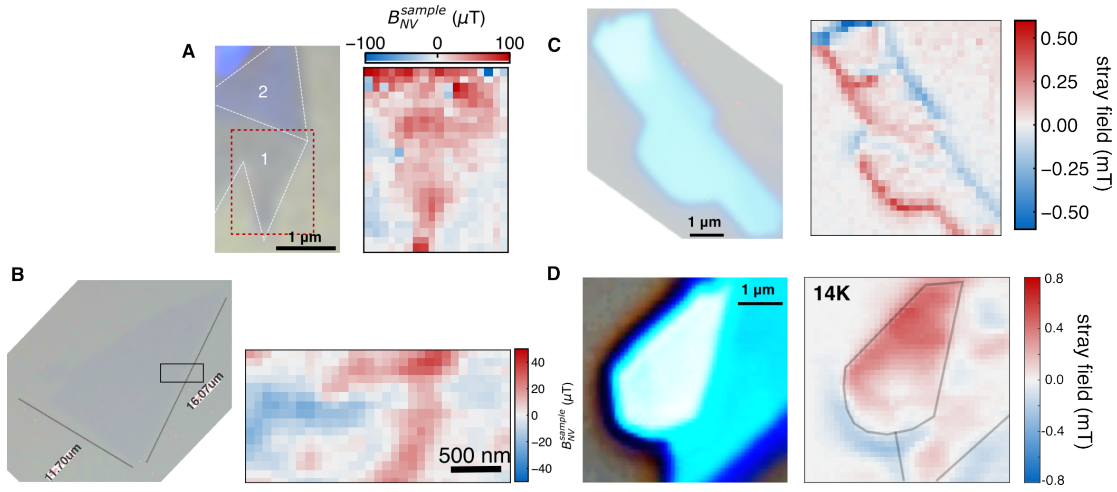


Figure 1.1: Optical and ESR maps of various samples measured in 131mT bias field. (A–B) show monolayer samples, "Pizza" and "Titanic." (C–D) correspond to thicker samples: (C) a 25/29 nm-thick sample ("Multilayer") and (D) a 26 nm-thick sample ("Teardrop"). Data is pure sample contribution of magnetic field along the NV direction.

The typical ESR contrast in our measurements varies under different magnetic field conditions. For a 2–4 mT out-of-plane field, which aligns the four NV orientations to the same resonance frequency, we observe a Rabi contrast of approximately 10%, representing the intrinsic NV contrast without power limitations. When the magnetic field is aligned along one NV axis at 64 mT, the Rabi contrast drops to 3–5%. For fields above 130 mT, we typically observe a contrast of 6–7%.

The theoretical maximum optical contrast for a single NV center is $\sim 30\%$. For an ensemble of NVs, when the magnetic field is aligned along one of the four NV axes, the expected maximum contrast reduces to 7.5%. In practice, the observed contrast is often lower due to factors such as strain and surface charge effects. Notably, under magnetic fields above 100 mT, the contrast generally improves because the NV centers become less sensitive to local strain variations³⁵.

1.4.2 RESULTS

Static field measurements (ESR and Ramsey spectroscopy, as mentioned above) were performed to characterize the magnetization and static magnetic structure of each sample. A confocal laser spot

was raster-scanned across the sample region using galvo mirrors. The spatial resolution of the scan is not limited by the accuracy of the galvo mirrors but rather by the diffraction-limited optical spot size, which is approximately 600 nm. The typical sample size is a few microns.

Figure 1.1 presents ESR maps at 131 mT of several samples alongside their corresponding optical images. For clarity, samples are named based on their shape, facilitating reference in later discussions. The measurements are not limited to the four samples shown here.

To obtain the field maps, background subtraction was performed to remove the contribution from the permanent magnet applied bias field. This was achieved by linearly fitting the field in a nearby sample-free region and subtracting the fitted field profile generated by the permanent magnet from total signal.

As shown in Fig. 1.1, thick samples generate a stray field approximately ten times larger than that of monolayer samples, despite both thick samples having a thickness of 45–50 layers. This behavior arises because CrCl_3 is a bulk antiferromagnet. Within each layer, the spins are ferromagnetically ordered, while across layers, they are antiferromagnetically aligned^{65,61,15}.

Under the applied 130 mT bias field, a spin-flop transition occurs, causing the initially anti-aligned spins to cant toward the field direction. This canting can explain the observed stray field in bulk samples, with its magnitude corresponding to the spin canting angle.

With 130 mT applied bias field, we still see spatial inhomogeneity of sample magnetic field in almost all samples. This could come from the existence of domains in the material that is introduced by strain/bubble/substrate-inhomogeneity. Spatial variation of the signal is very common in 2D materials, especially when measured with local probes.

1.4.3 RECONSTRUCTION OF THE MAGNETIZATION

In our ESR measurements, the NV center detects the external magnetic field projected onto the NV axis. Without performing vector magnetometry, our field maps provide information about only

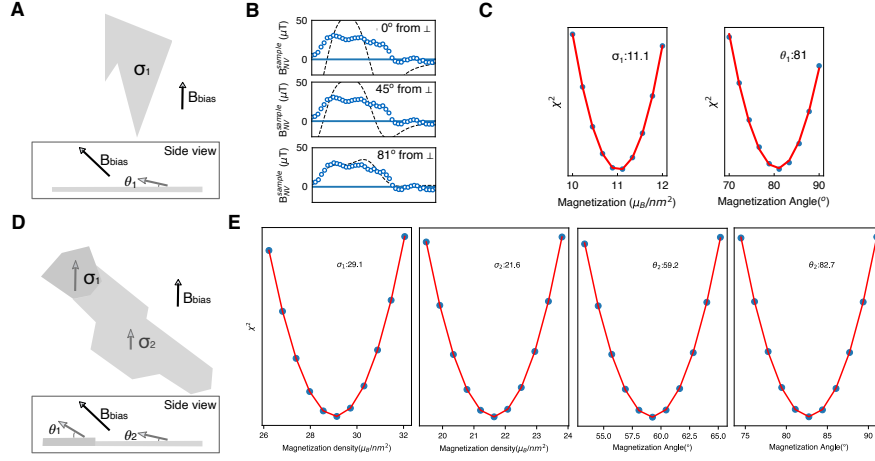


Figure 1.2: A Diagram of the monolayer region depicted in Fig. 2A. B Experimental linecut stray field data in comparison to simulations of the stray field for a magnetization value of $11 \mu_B/\text{nm}^2$ with different magnetization angles. C Best fit results for both magnetization angle and orientation minimizing the χ^2 residual with respect to the linecut data. D Multilayer CrCl_3 diagram for sample shown in Fig. 4A. The sample has two main regions with thicknesses of 29 nm (σ_1 region), and 25 nm (σ_2 region). E Best fit magnetization and value for the two different regions based on minimizing the residual χ^2 between the simulation and experiment for the stray field image.

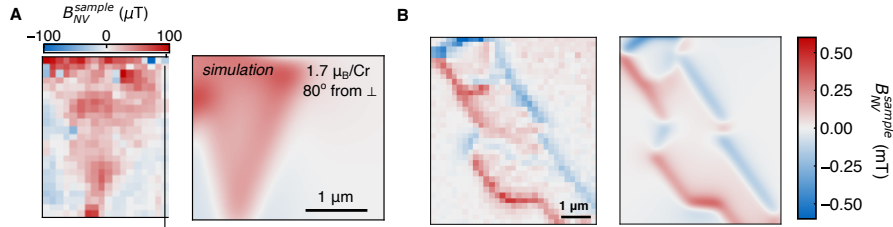


Figure 1.3: Comparison between the measured stray field maps and the corresponding optimized simulated stray fields for both the Pizza monolayer and multilayer samples. The simulations are plotted using the same color scale as their respective experimental measurements. Surface magnetization density and angle for the simulation of B is denoted in Fig. 1.2

one component of the magnetic field, while the full field vector is $\mathbf{B} = (B_x, B_y, B_z)$. Despite this incomplete information, it is still possible to infer the sample magnetization.

One widely used approach is Fourier-space reconstruction¹⁰, which has been demonstrated in various studies⁹¹. This method assumes that the magnetization is two-dimensional, with spins confined along a specific axis or within a known plane. However, the method depends on definitive knowledge of the direction of magnetization, and its accuracy is highly dependent on the quality of the field maps, including the number of measurement points and noise levels. We initially attempted this approach but found it ineffective for our measurements.

Recently, researchers have explored using machine learning models to reconstruct magnetization and current distributions from measured magnetic fields⁷⁷. However, the results of this method are also sensitive to the choice of training dataset and can be unstable due to limitations in our map resolution, noise, and the presence of magnetic domains in the 2D ESR data.

To infer magnetization from our magnetic field maps, we adopted a minimum χ^2 -optimization method:

$$\chi^2 = \sum_{(x,y)} \frac{(B_{\text{meas}} - B_{\text{sim}})^2}{B_{\text{sim}}}, \quad (1.3)$$

where B_{meas} is the measured magnetic field component projected along the NV axis, and B_{sim} is the simulated NV-projected field generated by a 2D magnet with shape S , extracted from the corresponding flake's optical image. The two fitting parameters are the uniform 2D magnetization density $\sigma(x, y)$, defined as constant σ if $(x, y) \in S$ and 0 otherwise, and the angle θ between the applied field and the surface normal.

We first perform a forward simulation of B_{sim} generated by a given magnetization (M, θ) and optimize these parameters to minimize χ^2 in Eq. 1.3. The local minimum indicates the most likely bulk magnetization and tilt angle.

This is a computationally intensive process, as it requires calculating the spatial distribution of the magnetic field from a grid of spins in each iteration. Direct calculations using the Biot-Savart law would be too complex and computationally demanding. Instead, we implemented the Forward Fourier (FF) method³⁷ for field simulations to improve efficiency.

The Forward Fourier method involves calculating the magnetic field generated by a given magnet configuration. It is performed in Fourier space based on Fourier-space(k-space) coordinates. In k-space, Maxwell's equations are simplified to algebraic relations between the stray field $B(\mathbf{k})$ and 2D magnetization $\sigma(\mathbf{k})$:

$$B_{xy}(\mathbf{k}, z) = -i\mu_0 k_{xy} \frac{e^{-kz}}{2} \sigma(\mathbf{k}, 0)$$

$$B_z(\mathbf{k}, z) = \mu_0 k_{xy} \frac{e^{-kz}}{2} \sigma(\mathbf{k}, 0)$$

$\mathbf{B}$ is calculated under the assumption that magnetization is a uniform 2D density σ across the whole sample flake (or regions of uniform thickness as determined by AFM). The shape of 2D magnetic regions are extracted from optical microscope and AFM images of CrCl_3 flakes. Further, we assume the magnetization's azimuthal angle aligns with the applied bias field, yet the polar angle and magnitude can vary due to competing effects among bias field, spin in-plane anisotropy, and shape-related dipolar field, etc. We extract the 2D magnetization σ and the polar angle θ by least-square optimization of the simulation compared to the stray field measured in the experiment (Fig. 1.2).

Results for χ^2 extraction of the magnetization density and angles are shown in Fig. 1.2C,E. The best-fit magnetization angle and value are presented in the figure 1.2C,E and figure 1.3 along with simulated stray fields for these best-fit parameters. An analogous procedure is used on data for multilayer sample.

We note that for the monolayer case(Fig. 1.2A-C), the best-fit magnetization from the stray field profile is $1.6\mu_B/\text{Cr}$, corresponding to approximately 30% lower than the expected value from the

dipolar model: According to the linearized spin-wave model(see the coming section)1.4.4, the magnetization at 6 K is 80% of the saturation magnetization. Also according to a previous study⁷, CrCl_3 has a saturation magnetization of $2.9 \mu_B/\text{Cr}$, yielding an expected magnetization of $2.2 \mu_B/\text{Cr}$ at 6K. This discrepancy may arise from material differences between the study by the Parkin group and ours, but most likely from imperfections in the simulation-optimization process. In particular, in cases where both magnetization angle and value are unknown, magnetization reconstruction is prone to systematic errors¹⁰. The results of the χ^2 optimization in our simulation are sensitive to slight mismatches between the model geometry and the true sample geometry, as well as the orientation of applied bias field relative to the sample geometry.

I.4.4 TEMPERATURE DEPENDENCE OF THE MAGNETIZATION

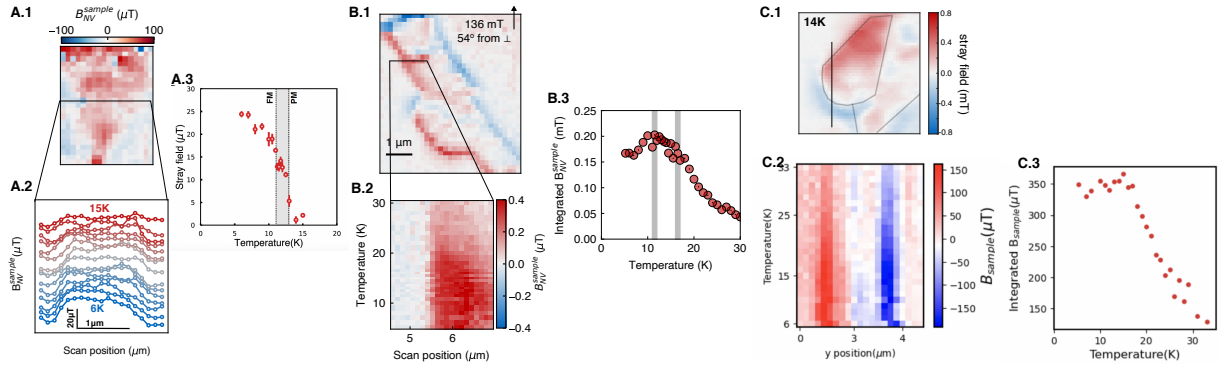


Figure 1.4: X.1 Stray field maps of three samples under a 131 mT bias field (for C, the bias field is 64 mT along one NV orientation): "Pizza" (A, monolayer), "Multilayer" (B, 29/25 nm), and "Teardrop" (C, 26 nm). Unless otherwise specified in the plot, measurements were performed at 6 K. X.2 Temperature dependence of a selected linecut (solid black line) across each sample. The magnetic field generated by the sample decreases with increasing temperature, suggesting the melting down of magnetic order. X.3 Integrated magnetic field from the linecuts. For A and B, the integrated field is obtained by averaging over sample pixels, while for C, it is extracted by taking the difference between the peak and dip values in the linecut. The integrated field should be linearly linked to the magnetization value by a geometry factor. All linecuts clearly show diminishing magnetization with increasing temperature. The remaining signal above T_C may originate from polarized spins in the paramagnetic phase under the applied bias field. For B, the magnetization exhibits distinct features at both the ferromagnetic (FM) and antiferromagnetic (AFM) transitions. In particular, the downturn below the AFM transition suggests the development of antiferromagnetic coupling.

Magnetization data were taken at various temperatures from base(5K) to above the transition temperature($T_c = 12\text{K}$ for monolayer sample and $T_c = 17\text{K}$, $T_N = 14\text{K}$ for bulk sample) with bias field.

We first employ stray field measurements on the monolayer sample(Fig. 1.4 A) to estimate the ferromagnetic transition temperature and exchange constant of a monolayer CrCl_3 film. The temperature-dependence of the integrated stray field (Fig. 1.4 A.2) reveals a continuous decrease in the magnetization with increasing temperature and a ferromagnetic-to-paramagnetic phase transition at $T_c = 12.5\text{K}$ (Fig. 1.4), a value consistent with previous measurements on epitaxially-grown monolayer CrCl_3 ⁷.

The measured static field of the CrCl_3 *multilayer* and *teardrop*. By monitoring the stray field across a single linecut as temperature increases (Fig. 1.4B-C), we observe two features in the temperature-dependent magnetization indicative of magnetic phase transitions(Fig. 1.4B.3), above which the signal decays with increasing temperature characteristic of a paramagnetic response. Based on a comparison to existing literature on this material^{65,15,13}, we conclude that the material undergoes two phase transitions – the first at $\sim 16.5\text{K}$ associated with ferromagnetic ordering within the layers, and the second at $\sim 12\text{K}$ associated with antiferromagnetic alignment between layers. Measurements on *Teardrop* exhibit similar qualitative behavior with slightly different transition temperatures. However, the expected feature at the AFM transition is not as pronounced in this sample. This may be due to the lower applied bias field in this case, as well as the requirement for a higher signal-to-noise ratio to resolve subtle features across the AFM transition.

SELF-CONSTANT SPIN-WAVE THEORY FOR A MONOLAYER SAMPLE

In linearized spin-wave regime, one assumes no correction to the magnon band dispersion from the spin-wave interactions. However upon increasing temperature, the gas of thermal magnon would modify the amplitude of average magnetization M , and this effect is taken into account of

the model, make it self-consistent spin-wave model. Here we compute the temperature dependence of the magnetization of a monolayer sample:

$$m_z(T) = g\mu_B \langle S_1^z \rangle = m_0 - \frac{g\mu_B}{2N} \sum_{\mathbf{q}, s=\pm} n_B(\omega_{\mathbf{q},s}), \quad (1.4)$$

where $m_0 = g\mu_B S$ is the expected saturation magnetization, $n_B(\omega) = [e^{\beta\omega} - 1]^{-1}$ is the Bose-Einstein distribution function, and $\mathbf{q}$ runs over $\mathbf{q} = \mathbf{b}_1 n_1/n + \mathbf{b}_2 n_2/n$ with $n_1, n_2 = 0, 1, \dots, (n-1)$ and $n^2 = 2N$.

As temperature approaches T_c from below, the higher-order terms in the spin operator expansion: $\hat{S}_z, S^+$ and S^- become important so that the linearized spin-wave theory can become insufficient. To partially account for those terms, one can take into account the renormalization of magnon dispersion due to finite magnon density and magnon interactions: $\omega_{\mathbf{q},s} \rightarrow \epsilon_{\mathbf{q}}$. Specifically, the renormalized dispersion $\epsilon_{\mathbf{q}}$ can be obtained by self-consistently solving^{32,63}:

$$\epsilon_{\mathbf{q}} = \omega_{\mathbf{q}} - \frac{1}{2NS} \sum_{\mathbf{q}'} [\omega_{\mathbf{q}} + \omega_{\mathbf{q}'} - \omega_{\mathbf{q}-\mathbf{q}'} - \omega_{\mathbf{0}}] n_B(\epsilon_{\mathbf{q}'}). \quad (1.5)$$

Here we neglected the small contribution due to the upper magnon branch.

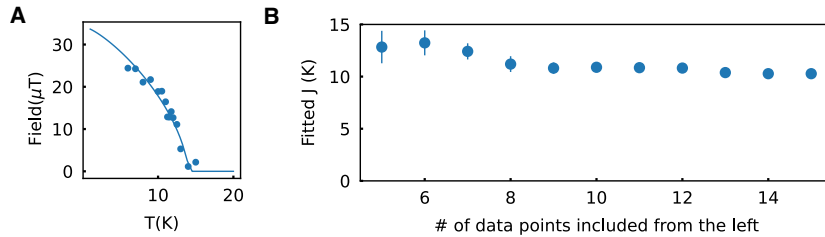


Figure 1.5: **A** Stray field measured on monolayer CrCl_3 fitted to self-consistent linearized Spin Wave model. J is $10.3\text{K} \pm 0.2\text{K}$ from the fitting including all data points. **B** J dependence on fitting range. x-axis represents the number of data points from the low temperature end included in the fitting

This approach provides a good approximation for numerically capturing the temperature-dependent magnetization, as long as the selected temperature points are sufficiently far from the transition temperature. However, performing the fit relies on one key assumption: the domain structures and the azimuthal magnetization angle remain unchanged across the temperature range of the fit. To validate this assumption, we first conduct the following analysis:

We extract the magnetization azimuthal angle in the previous section. For monolayer CrCl_3 , the best-fit magnetization is almost in-plane for all samples studied. Dipolar interaction, single-ion anisotropy and external bias field determine this magnetization angle, which is expected to be constant in the ferromagnetic ordered state until very close to the transition temperature. Indeed, the temperature-dependent stray field profile as seen in Fig. 1.4 A-C.2 and Fig. 3.4 is similar between temperatures, indicating that primarily the magnetization value is changing. Thus, the stray field measured by the NV at the same location across different temperatures should be linearly proportional to the magnetization density σ , $B_{\text{sample}}^{\text{NV}} = \mathbf{g} \cdot \sigma$, where $\mathbf{g}$ is the factor containing geometric information.

Self-consistent linearized spin-wave (SWT) analysis can well capture the magnetization as a function of the changing temperature, except in the critical region near the transition temperature T_c ^{32,63}. We fit the stray field data to a SWT model that has taken the bias field and lattice symmetry into account with two free parameters: $\mathbf{g}$ and exchange interaction J . We see that the data is well captured by SWT (Fig. 1.5A). The fitted J is not sensitive to the range of fitting, giving confidence in the robustness of this method (Fig. 1.5B). In conclusion, we report the J that fits data points between 6 and 11 K to be $10.9\text{K} \pm 0.4\text{K}$.

1.5 UNDERSTANDING THE CrCl_3 SPIN HAMILTONIAN

To better observe the DC magnetization measured via NV magnetometry in monolayer and multilayer CrCl_3 samples, and to understand their static and finite-frequency magnetic properties, it is essential to compare the relevant energy scales in the spin Hamiltonian of both monolayer and multilayer CrCl_3 lattices.

1.5.1 SPIN HAMILTONIAN OF MONOLAYER CrCl_3

In this section, I want to cover the spin Hamiltonian of CrCl_3 . The Hamiltonian is the foundation of modeling magnon dispersion, thus to understanding static magnetization, phase transition and even spin dynamics of CrCl_3 .

$$H = -J \sum_{\langle ij \rangle} (S_i^x S_j^x + S_i^y S_j^y + \alpha S_i^z S_j^z) - g\mu_B \sum_i (B_z S_i^z + B_x S_i^x) + H^{\text{dip}}. \quad (1.6)$$

Here $\alpha \approx 0.9930$ is the single-ion anisotropy parameter⁴⁷; since $\alpha < 1$, CrCl_3 is an easy-plane ferromagnet; J is the nearest neighbour exchange coupling. The crystal structure of CrCl_3 corresponds to a honeycomb lattice with the lattice constant $a = 5.942 \text{ \AA}$; the indices i and j in the first term of Eq. (1.6), thus, encode the two nonequivalent sublattices. The second term describes the Zeeman coupling associated with the applied magnetic field $\mathbf{B}$; $g \approx 2$ is the g -factor; $S = 3/2$. For $B_x = 0$, the system remains rotationally invariant, whereas a nonzero $B_x \neq 0$ explicitly breaks in-plane $U(1)$ -symmetry. The third term describes the dipole-dipole interactions between the spins, depending on the shape of the exfoliated material:

$$H^{\text{dip}} = -\frac{1}{2} \sum_{i \neq j} S_i^\alpha D_{\mathbf{r}_i - \mathbf{r}_j}^{\alpha\beta} S_j^\beta, \quad D_{\mathbf{r}}^{\alpha\beta} = \frac{\mu^2}{|\mathbf{r}|^3} \left(3 \frac{r^\alpha r^\beta}{|\mathbf{r}|^2} - \delta^{\alpha\beta} \right), \quad (1.7)$$

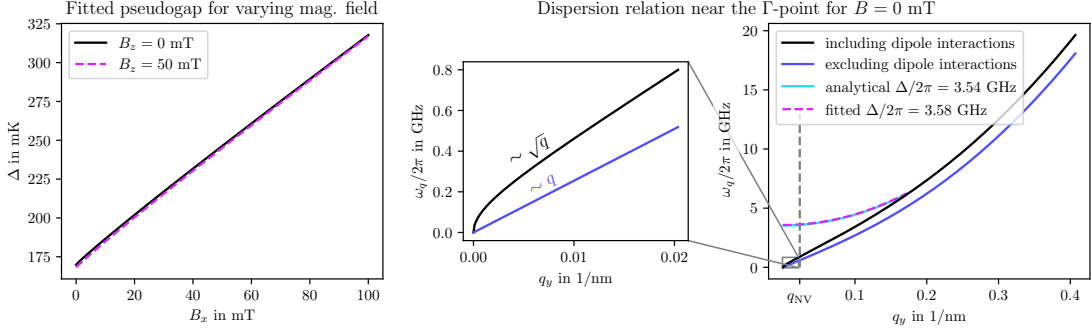


Figure 1.6: Linearized spin-wave theory. Left: Pseudogap Δ , defined as the effective gap when fitting high energy magnon to a parabolic format, as a function of the applied field. We find that i) $\Delta \approx 175$ mK at $B = 0$, ii) it increases by less than a factor of two when B_x is increased from 0 to 100 mT, and iii) Δ is approximately B_z -independent. Right: Magnon dispersion for $B = 0$ near the Γ -point. We observe that the large-momenta magnons are barely affected by the dipolar interactions and their dispersion is fairly quadratic – this can be seen from a parabolic fit to the magnon dispersion for large q (dashed pink line). The fitted value of Δ agrees well with the analytical result (corresponding to the cyan line). Middle: zoom-in into the magnon dispersion in the immediate vicinity of the Γ -point shows that the dipolar interactions can substantially modify the low-momenta magnons.

where $\mu^2 \approx (g\mu_B)^2$ ($\mu^2 \rightarrow \mu_0(g\mu_B)^2/(4\pi)$ in the SI units) and $\mathbf{r} = (\rho, z)$ is a three-dimensional vector. These interactions, similar to the single-ion anisotropy, also favor in-plane magnetization. For future reference, we note that $D_{\mathbf{r}}^{\alpha\beta} = D_{-\mathbf{r}}^{\alpha\beta} = D_{\mathbf{r}}^{\beta\alpha}$ and $D_{\mathbf{r}}^{xz} = D_{\mathbf{r}}^{yz} = 0$ – this latter property follows from the fact that we consider an atomically-thin sample positioned in the xy -plane with $z = 0$.

For CrCl_3 , we have the following set of energy scales:

$$T \simeq (5 - 20) \text{ K}, \quad J \approx 10.9 \text{ K}, \quad (1 - \alpha)J \approx 76 \text{ mK}, \quad (1.8)$$

$$g\mu_B B \Big|_{B=100 \text{ mT}} \approx 134 \text{ mK}, \quad \mu^2/(\mathcal{A}_0 a) \approx 5 \text{ mK}, \quad (1.9)$$

where $\mathcal{A}_0 = 3\sqrt{3}a^2/2$ is the unit cell area. These estimates show that i) the exchange coupling J is several orders of magnitude larger than the anisotropy terms and magnetic field, ii) the single-ion anisotropy and applied field might be comparable, and iii) the dipolar interactions appear as the least relevant.

Regardless of the energy scale comparison, the dipolar interaction plays a crucial role in determining both the static properties and low-energy spin dynamics of this magnet. On one hand, dipolar interactions are expected to stabilize the ferromagnetic phase in the absence of an applied magnetic field¹¹, in contrast to having the Berezinskii-Kosterlitz-Thouless (BKT) physics, where a thermal vortex gas proliferates within the magnet. On the other hand, dipolar interactions modify the low-energy magnon dispersion (see Fig. 1.6), effectively opening an effective "pseudogap" for high-energy magnons dispersion. The pseudogap, approximately being 3.5 GHz (corresponding to 50 mK), is comparable to the Zeeman energy under our experimental conditions. The detailed definition and discussion on pseudogap is in the supplementary material of Ref¹⁰². We will also have more discussion about the significance of pseudogap later (Section. 3.2.5).

Overall, the spins in CrCl_3 follow a Heisenberg spin model on a two-dimensional honeycomb lattice, with subtle modifications due to ion-anisotropy, dipolar interactions, and the external Zeeman field.

1.5.2 INTERLAYER COUPLING IN CrCl_3 MULTI-LAYERS

In multilayer CrCl_3 , spins in adjacent layers are antiferromagnetically coupled. The interlayer exchange coupling energy is on the order of $J \sim 0.1$ K for CrCl_3 single crystals. There is evidence that the exfoliation process may alter the stacking configuration, thereby affecting the interlayer coupling strength. An increase in the coercive field by a factor of 4–5, from 0.2–0.25 T in bulk CrCl_3 to 0.9–1 T in exfoliated thin flakes, suggests enhanced interlayer AFM coupling^{48,97}. However, even with the enhancement, compared to the intralayer exchange coupling ($J_{\text{FM}} \sim 10$ K), the interlayer coupling in exfoliated thin flakes remains only 0.5–1% of the intralayer interaction.

The relative strength of AFM interlayer coupling is further supported by our observation of spin canting in thin flakes. Under a 131 mT applied bias field, the spin canting is 4–5% of the saturation magnetization, which is consistent with a coercive field on the order of 1–2 T for the exfoliated

flakes⁴⁸. Furthermore, in the magnetization-temperature data (Fig. 1.4), two distinct features are observed, corresponding to the ferromagnetic (FM) (~ 17 K) and antiferromagnetic (AFM) (~ 14 K) phase transitions. A possible point of confusion is why the AFM transition temperature is comparable to the Curie temperature, despite the AFM exchange energy being two orders of magnitude smaller. An intuitive explanation is that the AFM phase may be stabilized by the formation of ferromagnetically ordered layers. Once formed, each layer carries an effective magnetic moment $\mathbf{M}$, which is coupled to adjacent layers through J_{AFM} . Since $\mathbf{M}$ represents the collective effect of all spins within the dipolar interaction range, the AFM phase transition occurs at an enhanced temperature, nearly 100 times higher than the corresponding exchange energy J_{AFM} .

Given the weak interlayer interactions in CrCl_3 , multilayer samples can still be treated as a collection of weakly coupled 2D ferromagnetic spin sheets when discussing spin dynamics. They exhibit similar behavior in terms of high-energy spin dynamics.

I can fly a few students up.

Amir, before group retreats

2

Noise Spectroscopy with NV Centers

WHEN I FIRST THOUGHT ABOUT NOISE, MY INSTINCT WAS TO ELIMINATE IT. HOWEVER, MY PHD RESEARCH TAUGHT ME TO APPRECIATE ITS POTENTIAL AND EXPLORE HOW TO MAKE USE OF IT. This journey revealed that noise is not merely a disruption but can store valuable information. In certain contexts, what we perceive as "noise" can be interpreted as a signal, offering new opportunities for analysis and understanding.

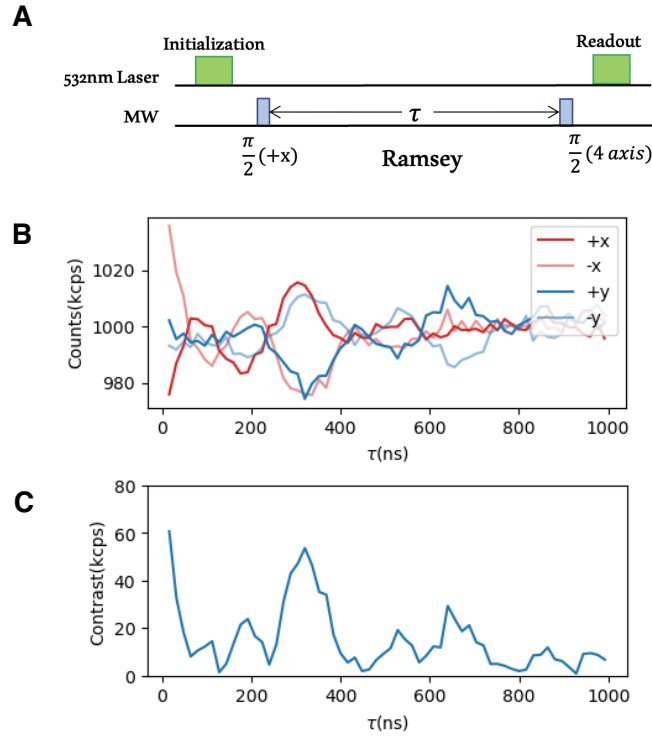


Figure 2.1: Ramsey Spectroscopy. **A** Pulse sequence for four-axis Ramsey spectroscopy. The spin is initialized in a superposition of $|0\rangle$ and $|1\rangle$ with phase $\varphi = 0$. After a free-precession time τ , the spin state is read out using $\pi/2$ pulses along four axes (+x, -x, +y, -y) to extract spin contrast and reconstruct the phase while accounting for variations in contrast. **B** Results of the four-axis readout. **C** Absolute contrast of Ramsey spectroscopy. Due to the presence of nitrogen nuclear spin hyperfine interactions and a surrounding spin bath, NV coherence exhibits collapses and revivals rather than a simple exponential decay^{20,55}.

The fluctuation-dissipation theorem, which I learned in Statistical Physics, states that thermodynamic fluctuations in a system at detailed balance are intrinsically linked to the response function of the same physical quantity, and vice versa. This implies that thermal fluctuations, also referred to as thermal noise, in a spin bath can naturally reveal the susceptibility of the desired system.

2.1 MOTIVATION: SURPRISING OBSERVATION FROM STATIC MEASUREMENTS

I presented and discussed key results from static field measurements in Chapter 1, primarily using ESR. However, during the initial exploration stage, I investigated alternative measurement schemes for probing static magnetic fields, including the Ramsey sequence.

The Ramsey sequence is illustrated in Fig. 2.1A. A four-axis readout is implemented to extract the spin phase while accounting for variations in spin contrast, which arise from hyperfine and dipolar coupling within the surrounding spin bath^{20,55} (Fig. 2.1B,C). These interactions introduce fluctuations that affect the coherence of the NV spin, making it crucial to optimize measurement parameters for reliable field sensing.

For DC field measurements, the free-precession time τ is chosen to maximize the Ramsey sensitivity, which follows $\eta \sim \frac{1}{C\tau}$, where C is the optical contrast. Since the sensitivity depends on a trade-off between coherence time and signal contrast, τ is selected based on the point where contrast reaches a local maximum. In our case, we found that $\tau \sim 300$ ns provided the optimal balance between contrast and measurement sensitivity⁵.

I used a Ramsey sequence with a fixed precession time of $\tau = 300$ ns, which corresponds to a peak in contrast and the optimal sensitivity point. The measurement spot was raster-scanned across a trilayer CrCl_3 sample, generating a 2D map of Ramsey contrast, as shown in Fig. 2.2.

Significant spin decoherence is spatially correlated with the shape of the trilayer CrCl_3 sample. The decoherence observed at the sample edges arises from the stray field of the magnet, which changes drastically at these boundaries due to discontinuities in magnetization. As a result, each NV center within the measurement ensemble experiences a large field gradient, leading to accumulated phase variations. This spatial inhomogeneity in the magnetic field induces dephasing in our measurements.

Surprisingly, I observed significant decoherence not only at the edges but also in the bulk region

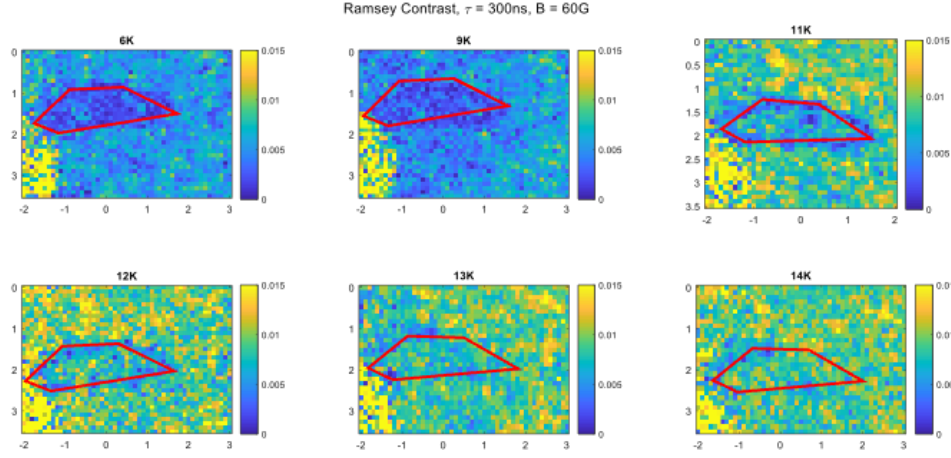


Figure 2.2: 2D Map of Ramsey Contrast. A spatial scan of the trilayer CrCl_3 as a function of temperature. The NV centers beneath the sample region exhibit increased decoherence, indicated by lower contrast at a fixed τ compared to the surrounding "background" area. This excess decoherence gradually diminishes as the temperature approaches T_c and disappears above it.

of the sample. Compared to NV centers positioned away from the sample (background), the NVs within the bulk exhibit excess decoherence, despite the absence of spatially varying stray fields — except in the case of domain walls.

Furthermore, the decoherence exhibits an inverse dependence on temperature: as temperature increases, the noise decreases. One possible explanation for the fading decoherence at the edges upon warming above the Curie temperature is the reduction of magnetization. However, the persistent decoherence observed in the bulk remains puzzling.

To investigate this effect further, I examined multiple samples under various applied fields. In all cases, bulk decoherence was consistently present and exhibited the same trend in temperature dependence, suggesting an intrinsic mechanism rather than an experimental artifact.

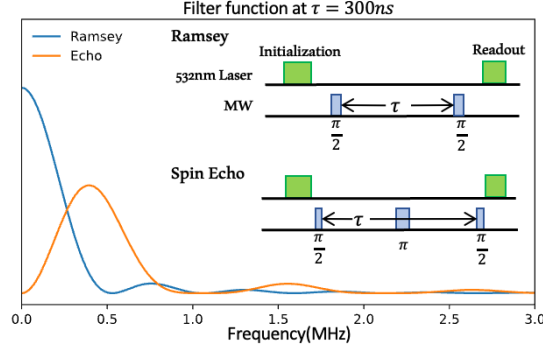


Figure 2.3: Filter functions Comparison of the Filter Functions and Pulse Sequences of Ramsey and Spin Echo.

2.2 DEVELOPMENT OF MEASUREMENT SCHEMES

To better extract the finite-frequency components of the sample's spectrum, we extended our measurement schemes to include dynamical decoupling techniques²⁰. The first challenge we encountered was understanding the complex dynamics introduced by the spin environment and the alignment of the applied bias field within an ensemble NV measurement spot.

2.2.1 DYNAMICAL DECOUPLING

A more effective approach to resolving the spectrum of magnetic noise is to use dynamical decoupling instead of the Ramsey measurement. By inserting π pulses during the evolution time, the sequence acts as a filter that suppresses DC and low-frequency components of the signal (Fig. 2.3). High-frequency signals, relative to N/τ (where N is the number of π pulses), average out during the precession time, resulting in optimal sensitivity at a frequency of approximately N/τ ¹⁰³. We leverage this property of dynamical decoupling to isolate finite-frequency magnetic signals from the spatial inhomogeneity of the stray field, allowing us to further investigate the decoherence effects observed in Ramsey measurements.

The preliminary measurement of T_2 on the *Teardrop* sample (Fig. 2.4) is both encouraging and

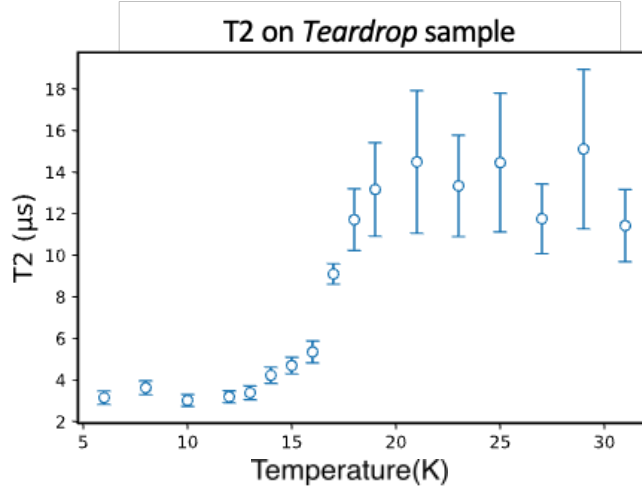


Figure 2.4: T_2 on *Teardrop* Spin echo coherence time (T_2) measured on *Teardrop* as the sample is warmed up. Coherence is suppressed between 5–15 K and recovers to its intrinsic level across the phase transition. Error bars represent the fitting uncertainty. The relatively large error bars for T_2 points above 20 K arise from T_2 exceeding the longest measurement τ , preventing full characterization of the decay.

puzzling. The increase in low-frequency noise upon cooling is also observed in spin echo measurements, which effectively rules out local static field inhomogeneity as the primary cause. The anomalous behavior of low-frequency (kHz–MHz) magnetic noise remains an open question, especially since thermal magnon fluctuations, which should increase with temperature, can be easily excluded as an explanation.

Meanwhile, extracting meaningful physical quantities directly from T_2 measurements requires further refinement. In the following paragraphs, I introduce developments in the experimental protocol and data analysis aimed at addressing these challenges.

2.2.2 MEASUREMENT OF HAHN-ECHO TIME TRACE

NV centers are initialized into the $m_s = 0$ state using a 532 nm laser pulse of 3.2 μ s duration. Transitions between spin states are driven by microwave pulses at the spin resonance frequency, which, in all of our experiments, corresponds to the splitting between the 0 and -1 states.

A microwave pulse resonant with the $m_s = 0 \leftrightarrow m_s = -1$ transition, applied for a duration equal to one-quarter of the Rabi cycle, rotates the NV spin into a superposition state, such as $(|0\rangle + |1\rangle)/\sqrt{2}$. Typical Rabi rates in our setup are 15 MHz.

The Hahn-echo sequence begins with an initialization pulse from a green laser, followed by a $\pi/2$ pulse that prepares the NV spin into a superposition state (Eq. 2.1). During the free evolution period τ , the spin accumulates a phase φ (Eq. 2.2) due to the environmental magnetic field along the NV quantization axis. A π pulse then flips the spin by 180° (Eq. 2.3), causing it to accumulate an equal phase in the opposite direction over the same duration τ . The sequence concludes with a final $\pi/2$ pulse applied along two orthogonal axes (+x and -x), projecting the spin into two distinct states (Eq. 2.5 and Eq. 2.6). The difference between the two optical readouts is determined by φ_1 , as shown in Eq. 2.7.

$$|0\rangle : \frac{\pi}{2} + x \rightarrow (|0\rangle + i|1\rangle)/\sqrt{2} \quad (2.1)$$

$$\text{evolve } \tau/2 \rightarrow (|0\rangle + ie^{i\varphi}|1\rangle)/\sqrt{2} \quad (2.2)$$

$$\pi + y \rightarrow (|0\rangle + ie^{-i\varphi}|1\rangle)/\sqrt{2} \quad (2.3)$$

$$\text{evolve } \tau/2 \rightarrow (|0\rangle + ie^{-i\varphi_1}|1\rangle)/\sqrt{2} \quad (2.4)$$

$$\frac{\pi}{2} + x \rightarrow [(1 - e^{-i\varphi_1})|0\rangle + i(1 + e^{-i\varphi_1})|-1\rangle]/2 \quad (2.5)$$

$$\text{or } \frac{\pi}{2} - x \rightarrow [(1 + e^{-i\varphi_1})|0\rangle - i(1 - e^{-i\varphi_1})|-1\rangle]/2 \quad (2.6)$$

$$C = \cos\varphi_1 \quad (2.7)$$

Throughout this work, the Hahn-echo “contrast” is obtained in the following way. At a particular evolution time, we record the difference in red photon counts following two different Hahn-echo sequences with the same evolution time, where in the two sequences, the last $\pi/2$ pulse is along the $+x$ and $-x$ axis, respectively. The counts difference for a given evolution time is then divided by the counts difference obtained for the minimum evolution time allowed by our instruments — 16 ns — which serves as a reference. The above 2-axis readout is used in the equilibrium noise measurements presented in this work. We periodically used a full 4-axis readout, including projections along $+y$ and $-y$, to ensure that the above procedure reliably measures the spin contrast.

2.2.3 HAHN-ECHO DYNAMICS IN THE PRESENCE OF MAGNETIC FIELD NOISE AND HYPER-FINE COUPLING WITH ^{13}C AND ^{14}N NUCLEAR SPINS

The interaction between the electronic wavefunction of the NV center, initialized and subsequently manipulated by a $\pi/2$ pulse into the state $(0 + -1)/\sqrt{2}$, and its surrounding environment can be complex. The Hahn-echo contrast, as discussed in Section 2.2.2, represents an ensemble-averaged signal from all NVs within the sensing spot.

We distinguish two primary contributions to NV decoherence. The first arises from dipolar in-

teractions with incoherent magnetic fields (magnetic noise), predominantly generated by P1 centers in the diamond⁶, surface spins⁹ and the nearby magnetic sample. The second contribution comes from hyperfine interactions with coherent magnetic field sources intrinsic to the diamond—namely, ¹³C and nitrogen nuclei—whose spins precess in the applied magnetic field and periodically entangle with the NV center.

The following sections detail this spin evolution and describe how T_2 is extracted in the presence of hyperfine coupling.

MAGNETIC NOISE

The first effect arises from dipolar interactions with stray, incoherent magnetic fields originating from distant sources, which are particularly relevant for noise magnetometry measurements. In this context, "distant" refers to field sources whose spatial overlap with the NV electron wavefunction is negligible, ensuring the absence of contact interaction—a valid assumption when the field source is more than a few nanometers away from the NV spin⁷³.

In the Hamiltonian, these interactions are described by

$$H^{\text{stray field}} = B_{\text{external}}(t) \cdot \mathbf{S}, \quad (2.8)$$

where $\mathbf{S} = \frac{\gamma}{2}\sigma$ represents the NV electron spin. Here, $B_{\text{external}}(t)$ is assumed to be independent of $\mathbf{S}$.

If we evolve $H^{\text{stray field}}$ over a Hahn-echo sequence of length t , we find that the NV wavefunction acquires a phase

$$\varphi(t) = \gamma \left(\int_0^{t/2} dt' B(t') - \int_{t/2}^t dt' B(t') \right). \quad (2.9)$$

The assumption here is that the π pulse duration is much shorter than t , a condition that is well satisfied in these experiments—typical π pulse times range from 20–50 ns, while T_2 values always

exceed several μs .

Over many repeated measurements, and under the assumption of Gaussian magnetic field fluctuations, the NV density matrix exhibits a loss of state purity given by $\exp(-2\langle\varphi(t)^2\rangle)$, which arises from the incoherent nature of $B(t)$. Consequently, the interaction of the NV with time-dependent magnetic fields leads to an exponential decay envelope in the Hahn-echo signal:

$$C(t) = \exp(-2\langle\varphi(t)^2\rangle). \quad (2.10)$$

The decay characteristics, captured by the dephasing function $\varphi(t)$, are determined by the frequency distribution and intensity of stochastically varying magnetic fields.

Intrinsic magnetic field fluctuations originate from nearby P1 centers, surface spin, nitrogen nuclei, and ^{13}C nuclei in the bulk of the diamond. The contribution of the magnetic sample on the surface of the diamond, to first order, is simply an added time-dependent magnetic field $B(t) = B_{\text{intrinsic}}(t) + B_{\text{sample}}(t)$, which enters the Hahn-echo signal as a product. Here it is assumed that there is no non-linear interaction between the magnetic field induced by the sample and that induced by the spin bath in diamond.

$$C^{\text{noise}}(t) = \exp(-2\langle\varphi_{\text{intrinsic}}(t)^2\rangle) \times \exp(-2\langle\varphi_{\text{sample}}(t)^2\rangle) \quad (2.11)$$

This means that the sample contribution to dephasing is separable from the complex hyperfine interactions intrinsic to the diamond, as well as the intrinsic dephasing from the spin bath in the diamond.

HYPERFINE COUPLING

In addition to incoherent magnetic fields, the NV center experiences hyperfine interactions with nearby spins, specifically ^{13}C and nitrogen nuclei. These interactions are described by the Hamiltonian:

$$H^{\text{hyperfine}} = \sum_{i,j=x,y,z} A_{i,j} S_i I_j. \quad (2.12)$$

The electron spin of the NV center, $\mathbf{S}$, interacts with the nuclear spin, $\mathbf{I}$, through the hyperfine coupling tensor $\mathbf{A}$. In the context of spin echo measurements, which primarily couple to the S_z component at first order, this interaction can be simplified to include terms of the form $I_z S_z$, $I_y S_z$, and $I_x S_z$.

Under unitary evolution, these Hamiltonian terms give rise to quantum-coherent oscillations in the Hahn-echo signal, as the nuclear spins periodically entangle with the NV wavefunction. Importantly, to zeroth order, these coherent effects are independent of the distant magnetic field noise sources discussed in the previous section. As a result, the total Hamiltonian can be expressed as two components:

$$H = H_{\text{hyperfine}} + H_{\text{stray field}}. \quad (2.13)$$

Under unitary evolution, the Hahn-echo signal $C(t)$ is described as a product of exponential and oscillatory terms:

$$C(t) = \exp(-2\langle\varphi(t)^2\rangle) \times \text{oscillatory terms}. \quad (2.14)$$

The exponential decay arises from dipolar interactions with the magnetic environment, while the oscillatory terms originate from hyperfine interactions with nearby spins. In noise magnetometry experiments, our primary interest lies in characterizing the exponential decay component of the Hahn-echo signal.

To quantitatively extract the exponential dephasing function from our experimental data, it is es-

essential to develop a method for isolating the oscillatory terms. Here, we describe how this is achieved by fitting the full Hahn-echo signal using a physical model that accounts for hyperfine interactions. Later, we demonstrate that key parameters of the dephasing function can also be estimated through a simpler analysis. This serves to validate that the central results remain independent of the specific analysis approach.

The two primary nuclear spin species in the diamond substrate are ^{13}C and ^{14}N . The orientation of ^{14}N nuclear spins is pinned by quadrupole interactions, effectively acting as a static ("DC") field for the NV spin²⁰. This results in oscillations observed in Ramsey measurements (as seen in Fig. 2.1). However, since the field-induced phase shift is static, it is canceled out by a spin-echo sequence.

The hyperfine interaction between an NV center with spin $\mathbf{S}$ at a vector $\mathbf{r}$ from a ^{13}C with spin $\mathbf{I}$ was modeled in a study by Childress et al.²⁰. Here, we restate the main points for the sake of completeness. The Hamiltonian that describes the hyperfine interaction between the NV and the ^{13}C nucleus is given by:

$$H^{\text{hyperfine}} = -\mu_e\mu_n \frac{8\pi|\psi_{\text{electron}}(\mathbf{r})|^2}{3} \mathbf{S} \cdot \mathbf{I} + \left\langle \frac{\mu_e\mu_n}{r^3} (\mathbf{S} \cdot \mathbf{I} - 3[\hat{\mathbf{r}} \cdot \mathbf{I}][\hat{\mathbf{r}} \cdot \mathbf{S}]) \right\rangle, \quad (2.15)$$

where ψ_{electron} is the electron wavefunction of the NV, and μ_e and μ_n are the dipole moments of the NV and ^{13}C , respectively. $\mathbf{r}$ is the relative location of the ^{13}C nuclear spin to the NV center. The first term is the contact interaction — only relevant for ^{13}C nuclei within a few lattice sites. The second term is the dipolar interaction with the ^{13}C . Evolving this Hamiltonian yields an expression for an oscillatory component of the Hahn-echo signal:

$$C^{\text{oscillatory terms}}(t) = \left(1 - \frac{|\mathbf{B}_0 \times \mathbf{B}_{-1}|^2}{|\mathbf{B}_0|^2|\mathbf{B}_{-1}|^2} \sin^2(\omega_{-1}t) \sin^2(\omega_0t) \right). \quad (2.16)$$

In the equation of NV contrast, $\mathbf{B}_0$ and $\mathbf{B}_{-1}$ are the effective dipole field generated by electron

spin sensed at the location of ^{13}C nuclear spin. The two frequencies ω_0 , ω_{-1} can also be associated to the electron spin state $|m_s = 0, -1\rangle$, since $|m_s = -1\rangle$ has larger magnetic moment, the fast frequency ω_{-1} should correspond to it.

As stated earlier, when considering only first-order effects, the contributions from S_x and S_y can be neglected. ω_0 represents the precession of the electron spin state at the Larmor frequency of the ^{13}C nuclear spin in an applied magnetic field, through coupling term $I_z S_z$. On the other hand, ω_{-1} is determined by the splitting of the NV electron spin $|1\rangle$ state induced by the dipolar interaction term from nuclear spin, through $I_x S_z$ and $I_y S_z$. It is also the entanglement strength of the electron spin $|1\rangle$ and the nuclear spin that depends on the distance between the ^{13}C and the NV center.

Fig. 2.5 shows a simulated Hahn-echo signal in the presence of this hyperfine coupling to a nuclear spin and a weak bias field of 4 mT. The fast oscillations are associated with ω_{-1} , while the slow oscillation is ω_0 .

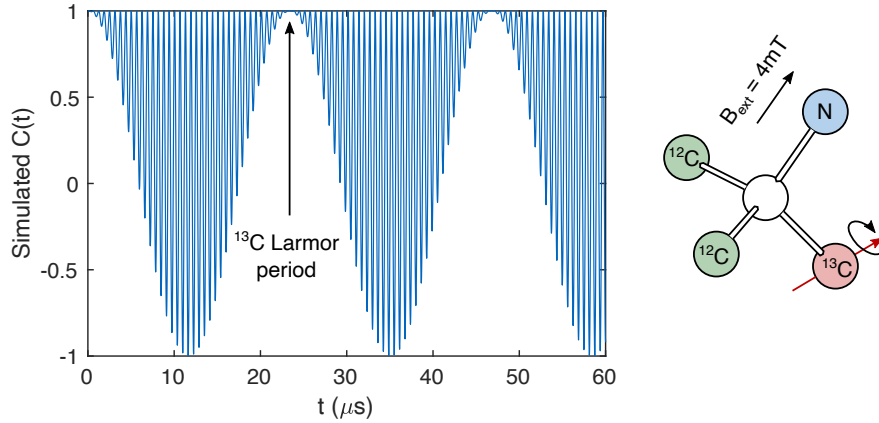


Figure 2.5: Simulation of Hahn-echo signal in the presence of hyperfine coupling to a single ^{13}C spin with an applied bias field of 4 mT. Fast oscillations are associated with the hyperfine splitting of the $| - 1 \rangle$ state, while slow oscillations are associated with the precession of the $|0\rangle$ state at the bare Larmor period of ^{13}C in a magnetic field.

When multiple ^{13}C centers exist at slightly different distances from their respective NV centers as is the case in our ensemble measurements, we must sum over all hyperfine interactions $V_{\text{tot}} = \sum_j V_j$. This summation corresponds to a product in the unitary evolution of the Hahn-echo signal

$C_{\text{tot}} = \prod_j C_j$. To a first approximation, ω_0 is the same for all ^{13}C nuclei, again representing the bare Larmor precession frequency, while ω_{-1} depends strongly on the position of the ^{13}C relative to the NV center as well as the position relative to the NV axis. By taking a product over a range of ω_{-1} values, an approximate expression for the Hahn-echo signal is obtained.

$$C(t) = \exp(-\langle\phi(t)^2\rangle) \times \left[\exp\left(-\left(t - \frac{2\pi n}{\omega_0}\right) \omega_C\right)^m (1 - b \sin^2(\omega_0 t)) \right], \quad (2.17)$$

where ω_C and b are fit parameters which encode the distribution of ω_{-1} values. $n = 0, 1, 2, \dots$, which describes how the initial exponential decay due to ^{13}C spins later revives after a multiple of the bare Larmor period when 1. all the ^{13}C nuclear spins returns to the initial orientation and 2. all of the different ω_{-1} contributions cancel. m being the phenomenological exponent to capture the decay due to distribution of ω_{-1} , and the value of $m = 3$ can best capture the experimental curve, $m=4$ can best capture the simulation. A simulated example of such a scenario is shown in Fig. 2.6⁸⁶.

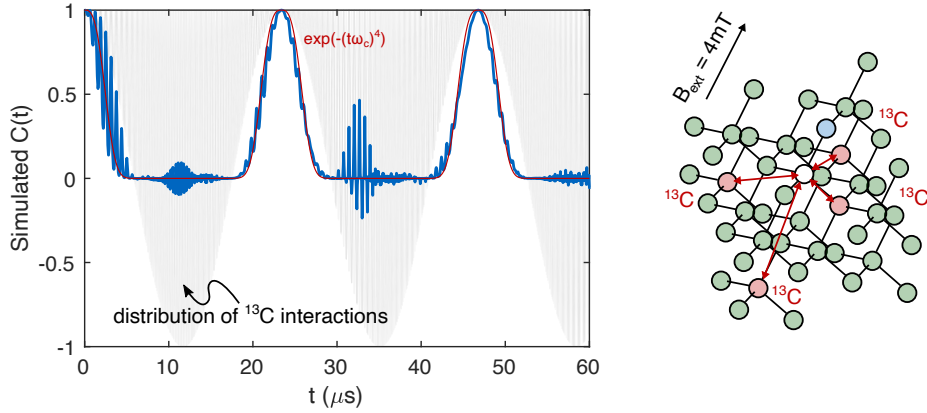


Figure 2.6: Simulation of Hahn-echo signal in the presence of hyperfine coupling to multiple ^{13}C spins with different coupling strengths at an applied bias field of 4 mT. Fast oscillations interfere with each other to produce an exponential decay with periodic revivals at the bare ^{13}C Larmor frequency. The decay is best captured by a $e^{-(t\omega_c)^m}$, where $m=4$ for the specific simulation parameters used. The timescale associated with this exponential decay, ω_C , depends on the distribution of ^{13}C hyperfine interaction strengths.

Finally, a fast oscillation $\sin^2(\omega_N t)$, likely originating from hyperfine coupling to nitrogen nuclei, needs to be included. The Nitrogen nuclei has fixed location relative to the NV center electron wavefunction, so that the oscillation rate is a constant, independent of applied magnetic field. The full Hahn-echo signal is therefore described by:

$$C(t) = \exp(-2\langle\varphi(t)^2\rangle) \times \left[\exp\left(-\left(t - \frac{2\pi n}{\omega_0}\right)\omega_C\right)^m\right] (1 - b \sin^2(\omega_0 t) \sin^2(\omega_N t)) \quad (2.18)$$

We obtain the hyperfine coupling parameters ω_C , ω_N and b based on Hahn-echo measurements of NV centers away from the sample (Fig. 2.7). At 4mT field, the ^{13}C revival period is 47 μs . The fast nitrogen hyperfine oscillation is 3.3 MHz, and the initial dephasing due to distribution of ^{13}C occurs with a time constant of 5.6 μs . For this fitting, we assume that the dephasing function $2\langle\varphi^2\rangle = (t/T_2)$, so that the decoherence follows a simple exponential form. This is shown to be a good approximation in section 2.2.2. We found that the initial dephasing and oscillations, both due to hyperfine interactions, are unaffected by increasing temperature over this temperature range (Fig. 2.8). The only changes to these dynamics are changes to T_2 , either due to the presence of the sample or due to changing temperature.

In cases where T_2 is long enough that the ^{13}C revivals are present, the most effective method to extract T_2 is by measuring the contrast at the peak of the 1st ^{13}C revival, $\tau_{1\text{st revival}}$, and dividing the measured contrast by the reference at zero evolution time. This quantity is converted to T_2 by assuming a simple exponential dephasing $1/T_2 = -\log(C(1\text{st revival}))/\tau_{1\text{st revival}}$. For the data shown in Fig. 2.7, the above method gives a value of $T_2 = 42 \mu\text{s}$ which is in good agreement with the T_2 obtained from fitting the entire curve (40.2 μs). Because the complex hyperfine dynamics are essentially independent of temperature, each measurement on the samples consists of calibrating the specific time of the ^{13}C revival, and measuring changes to the contrast at this evolution time as a function of temperature or spatial position relative to nearby points off the sample.

The above procedure is used when changes to T_2 induced by the sample or increasing temperature are small enough that the first revival is still present. In cases where the sample signal is large enough to eliminate the first revival (i.e. the bulk sample), the initial Hahn-echo decay, including the temperature-independent hyperfine oscillation parameters, is fit to extract T_2 . In this case, the only free parameter of the fit is T_2 (Fig. 2.9). The hyperfine parameters are determined on the background and held fixed as a function of temperature.

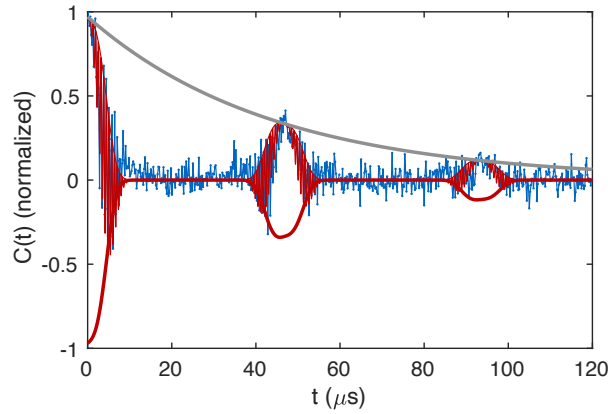


Figure 2.7: Hahn-echo signal taken on bare diamond at 6 Kelvin with 4 mT out of plane field. The grey line shows a simple exponential dephasing component. The red envelope is a ^{13}C oscillation, and the fast oscillations are due to hyperfine coupling to nitrogen nuclei. Here the fitted $T_2 = 40.2 \mu\text{s}$.

At higher magnetic fields, the external field aligns more closely with the NV axis, modifying the hyperfine interaction. In this regime, the faster oscillatory component, ω_{-1} , originating from transverse hyperfine coupling, is suppressed. Meanwhile, the slower oscillation, corresponding to the ^{13}C Larmor precession, becomes more rapid as its frequency scales proportionally with the applied field strength. This behavior allows for a more straightforward extraction of the dephasing time. Specifically, by measuring the Hahn-echo signal at discrete time points coinciding with ^{13}C Larmor revivals, we can isolate the decay envelope and fit it to an exponential function, thereby obtaining an accurate estimate of the coherence time. Example measurement curves illustrating this approach are shown in Fig. 2.10.

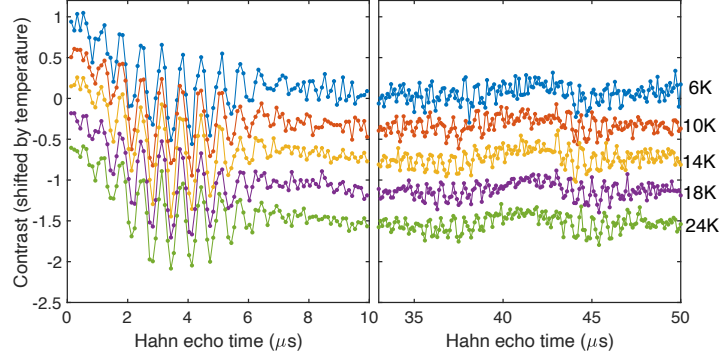


Figure 2.8: Temperature-dependent Hahn-echo decay dynamics at 4mT out of plane field taken on a bare region of diamond with only hBN coverage. Because the oscillations and ^{13}C revival period are independent of temperature, T_2 associated with “low-signal” samples like monolayer CrCl_3 can be obtained by measuring at the times corresponding to ^{13}C revivals, as described in the text.

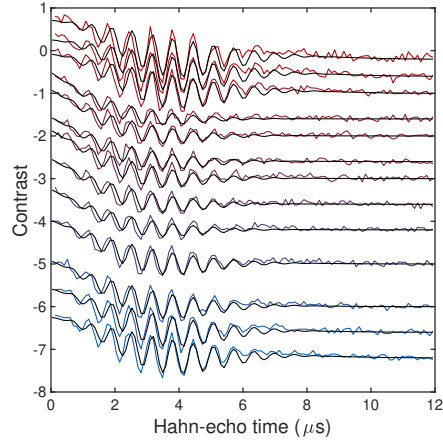


Figure 2.9: Example of fits (black) to first ^{13}C decay on bulk CrCl_3 sample. The only free parameter is T_2 . Curves are taken at different temperatures and offset vertically for clarity from 24 K (red) to 6 K (blue). In this case, the dephasing induced by the sample is large enough to fully suppress the first ^{13}C revival, so that T_2 can be fully captured in the first 10 μs .

2.2.4 ISOLATION OF THE SIGNAL

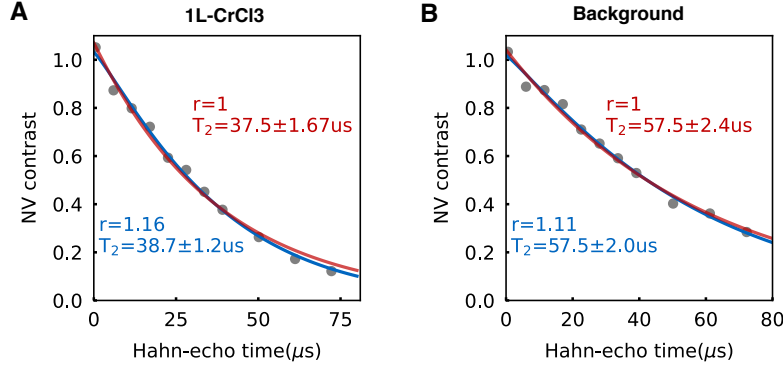


Figure 2.10: Hahn-echo measurement curve at $T = 5.2$ K A on a monolayer CrCl_3 and B off-sample. Blue curves are fitting the data points to the exponential function with a stretched exponent r . Red curves are fitted assume $r=1$, the simple exponent. Difference between fitted decoherence times from the two methods are the same within the error bars

To isolate the dephasing contribution induced by the sample, a background-subtraction procedure is necessary. Here, we employ the simplest approach: first, we measure the dephasing rate in a reference region away from the sample, then subtract this background contribution from the rate measured on the sample. The dephasing rate in both cases is extracted by fitting the Hahn-echo decay to a simple exponential function. This method provides a straightforward way to quantify the additional dephasing introduced by the sample while minimizing the influence of intrinsic NV decoherence mechanisms.

$$\frac{1}{T_2} = \frac{1}{T_2^{\text{sample}}} - \frac{1}{T_2^{\text{background}}} \quad (2.19)$$

The most rigorous approach to analyzing the dephasing would involve first normalizing the Hahn-echo signal measured on the sample by the corresponding background signal. This normalized trace could then be fitted using either a stretched exponential function (Section 2.2.3) or a crossover model²⁷ as a function of the Hahn-echo time (see also Section 2.2.3). However, in our case, the signal-to-noise ratio, combined with the presence of hyperfine interactions with nuclear spins in

the diamond, makes this level of analysis impractical. Instead, we demonstrate that a simple exponential fit reliably captures the essential behavior of T_2 (Fig. 2.10). To validate this approach, we also performed a stretched exponential fit and found no significant difference in the extracted T_2 values (Fig. 2.11). While the stretched exponent itself varies from sample to sample and with temperature, the extracted T_2 remains consistent, confirming the robustness of the simpler fitting method.

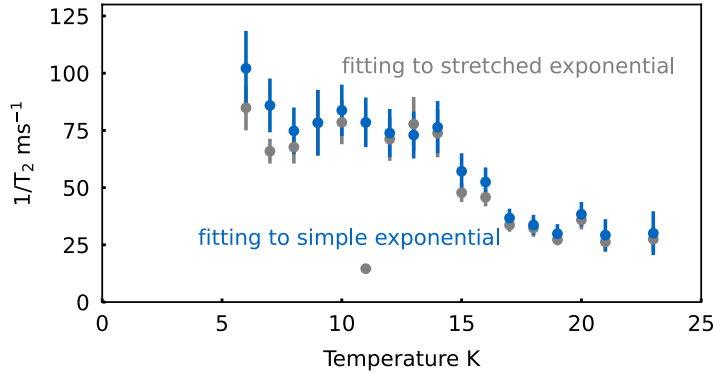


Figure 2.11: Temperature-dependent T_2 extracted from stretched exponent and a simple exponent.

At 4 mT, the accessible portion of the Hahn-echo decay curve is limited due to the slow Larmor precession of ^{13}C spins (see Section 2.2.3). In this case, we assume the dephasing follows a simple exponential decay for analysis. The validity of this assumption at higher fields, where the Larmor oscillations become more rapid and the full decay curve is more accessible, reinforces our confidence in the extracted T_2 values, even in the presence of hyperfine interactions with ^{13}C nuclei.

Similarly, we obtain a comparable T_2 by numerically identifying the point in the Hahn-echo decay curve where the contrast reaches $1/e$. This consistency across different extraction methods ensures that our main observations remain robust and independent of the specific analysis approach used to determine $1/T_2$. It also justifies our choice of fitting the decay to a simple exponential and applying a straightforward background subtraction — where the dephasing rate measured off the sample is subtracted from that measured on the sample.

Most spatial and temperature scans of the local decoherence rate are performed using a fixed evolution time τ chosen such that the Hahn-echo contrast decays to approximately $1/e$. For normalization, the shortest evolution time is set to $\tau_0 = 16, \mu\text{s}$. By monitoring the contrast change between these two time points, we can efficiently extract $1/T_2$, significantly reducing measurement time. However, this approach introduces limitations: when T_2 varies drastically due to sample or temperature changes, the measurement error increases as the chosen τ deviates from the actual T_2 . An improved approach could involve adaptively selecting τ based on the contrast ratio at τ and τ_0 , allowing for more accurate measurements in cases where T_2 changes significantly. Implementing such an adaptive scheme may be particularly useful for tracking rapidly evolving signals.

The extraction of magnetic noise from NV decoherence follows a straightforward method based on the assumption of a simple exponential decay. The spin contrast decay is directly linked to the variance of the superposition phase,

$$\frac{C(\tau)}{C(\tau_0)} = e^{-\tau/T_2} = e^{-\langle\varphi^2\rangle}. \quad (2.20)$$

Here, the phase accumulation in the superposition state is governed by the spin precession under an external magnetic field,

$$\varphi = \gamma B_{\parallel} \tau. \quad (2.21)$$

As a result, the variance of the accumulated phase directly corresponds to the variance of the time-dependent magnetic field,

$$\langle\delta B^2\rangle = \frac{\langle\delta\varphi^2\rangle}{\gamma^2\tau^2}. \quad (2.22)$$

This provides a direct way to quantify the local magnetic noise from the NV decoherence measurement. For reference, the equilibrium magnetic noise in a monolayer system is approximately $1 \mu\text{T}^2$ (Fig. 3.1).

2.2.5 MEASUREMENTS ON THE BACKGROUND

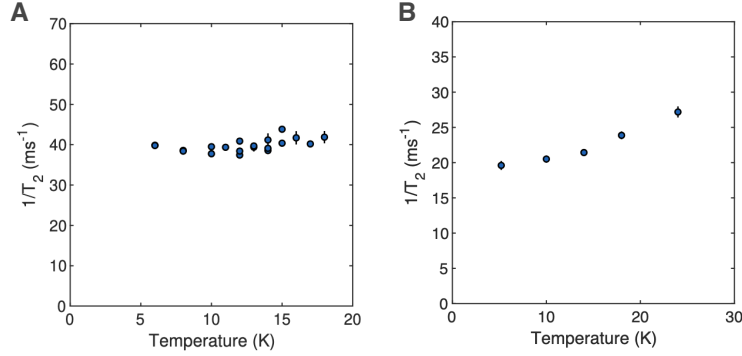


Figure 2.12: Measurements on bare diamond with only hBN coverage at 4 and 131 mT. **A.** Measurements on bare diamond with only hBN coverage at 4 mT out of plane field. T_2 is obtained by measuring the contrast at the first ^{13}C revival and dividing it by the reference $1/T_2 = -\log(C(\text{first revival}))/\tau_{\text{first revival}}$. **B.** The spin echo curve is sampled only at C13 revival time, from which we extract T_2 as a function of temperature.

Here, we present measurements on a background region of the diamond, covered only by hBN. The signal shows that the intrinsic dephasing is only weakly temperature-dependent over the 0–20 K range. This behavior holds for both a 136 mT field aligned with the NV axis and a 4 mT field applied out of plane (Figs. 2.12). In practice, background measurements were taken on a nearby region of the diamond for each sample location studied. Across all regions, the same qualitative behavior was observed, with only minor variations ($\sim 5\%$) in the background dephasing rate. Because the background measurements had fewer temperature points than the sample measurements, the temperature-dependent background $1/T_2$ rate was interpolated and subtracted from the sample data to isolate the sample-specific contribution to the signal.

2.3 LASER EFFECT ON CrCl_3

At a certain stage of our research, we realized that the green laser used for NV center initialization induced a transient excitation in the bulk CrCl_3 sample. Given that CrCl_3 is a semiconductor with

a band gap of approximately 2.4 eV⁵⁶, it can be optically excited by the 532 nm green laser, which has sufficient energy to promote carriers across the band gap. Density functional theory (DFT) calculations suggest that the conduction and valence bands near the band edges possess opposite spin orientations, meaning that optical excitation not only generates electron-hole pairs but also perturbs the material’s spin structure¹⁰⁹.

Additionally, the focused laser delivers approximately 200 μ W of power within a 500 nm Gaussian spot, raising concerns about local heating effects that can not be effectively captured by our thermometer below the sample holder. While we did not systematically study the underlying mechanisms of laser-induced perturbations in CrCl_3 , we report our observations of these effects in this section.

It is worth noting that the data shown in Figure 3.5 were acquired without accounting for the laser excitation effect and thus include contributions from laser-induced signals. All other data were taken after this effect was identified and properly mitigated.

2.3.1 EFFECT OF TIME DELAY BETWEEN LASER INITIALIZATION AND HAHN-ECHO SEQUENCE

An example of this laser-induced effect is shown in Fig. 2.13A, where introducing a delay between the NV initialization laser and the first $\pi/2$ pulse of the measurement sequence alters the measured $1/T_2$. Notably, no such effect was observed off the sample.

The relaxation timescales observed in our samples, on the order of $\sim 10\mu\text{s}$ at most, appear to be faster than those reported in previous studies. To equilibrate the sample before start taking measurement, we introduced a sufficiently long delay between the initialization and the Hahn-echo sequence to allow T_2 to reach a steady state. This transient laser-induced effect appears to be specific to the bulk sample, where, at both low and high fields (Fig. 2.13B and C), a delay exceeding 10 μs was required to reach equilibrium.

In the monolayer samples studied, we observed that adding a delay never significantly affected the

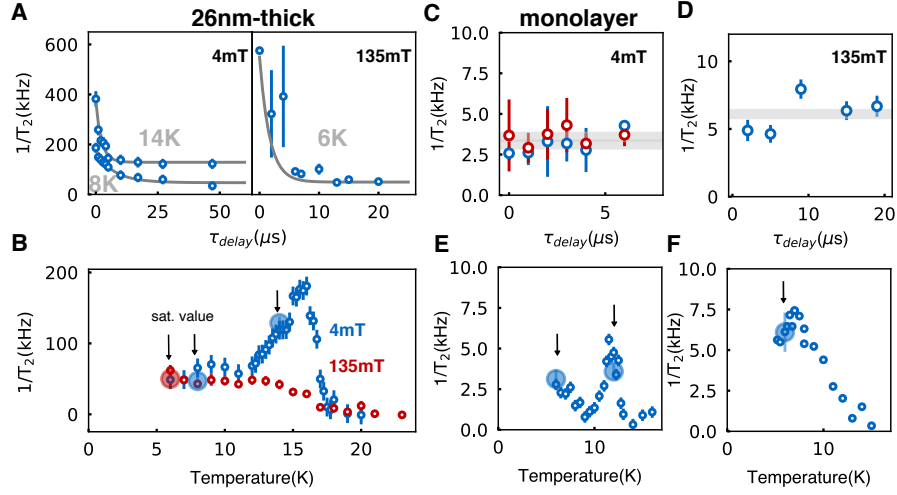


Figure 2.13: Magnetic noise signal saturates with delay time between initialization and laser readout. **A** Magnetic noise signal on *teardrop* sample in various fields and temperatures. $1/T_2$ decreases first with increasing delay time and later reach saturation. We extract the saturation value from fitting to the biased exponential form: $Ae^{-\tau_{\text{delay}}/T} + B$. **B** Saturation value from **A** plotted on temperature-dependence trace. The extracted saturation value agrees with temperature scan taken at a fixed delay time. **C-D** Delay test on monolayer flakes. Blue and red traces are taken at 6K and 12K correspondingly. Grey line indicates the averaged value of different delay times (from **C-D**). All data sets on monolayer are insensitive to delay-time within our sensitivity. **E-F** Temperature traces taken with $2\mu\text{s}$ delay time overlapping with the averaged values. Data in **C** and **E** are taken on monolayer sample 3. Data in **D** and **F** are taken on monolayer sample 2.

measured value of T_2 (see for example Fig. 2.13D and F) at either 4 mT or 136 mT. A more in-depth study of the transient T_2 signal of bulk CrCl_3 will be the subject of future investigation.

2.3.2 BEHAVIOR OF THE LASER EXCITATIONS

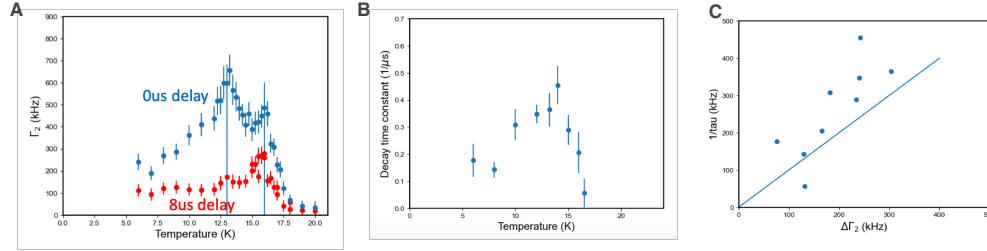


Figure 2.14: Temperature dependence of the laser induced dynamics. **A** Another temperature-dependent decoherence rate ($\Gamma_2 = 1/T_2$) trace measured on the *teardrop* sample, at 4 mT along surface normal, showing two distinct transitions at 13 K and 16 K. Two curves are recorded with a delay of 0 and 8 μs between the end of the initialization laser pulse and the start of the dynamical decoupling sequence. **B** The decay time of the transient excitation induced by the laser is extracted by measuring the signal decay as a function of the delay time and fitting it to a simple exponential curve. **C** The decay rate versus the amplitude of the laser induced magnetic noise.

Given time constraints, we did not explore the physics of laser-matter interactions beyond the scope of our main research. However, here we document the observed phenomena associated with the laser effect, which may suggest interesting directions for future studies.

We observe that the equilibrated noise (after a long delay) exhibits a prominent peak only at the temperature corresponding to the ferromagnetic transition. Across the AFM transition, we either observe a discrete step (Fig. 2.14A) or a gradual decay of magnetic noise from its peak at the ferromagnetic transition to a lower baseline below the AFM transition (Fig. 2.13).

In the equilibrium measurements, the absence of enhanced noise at the AFM transition is expected for a reason. In an antiferromagnetic (AFM) order, the order parameter alternates on the scale of a single unit cell, corresponding to a high spatial frequency of approximately 1 nm^{-1} . Given that our NV-to-sample distance is on the order of $\sim 50 \text{ nm}$, the mismatch between the order parameter wavelength and the effective momentum filter set by the NV naturally suppresses the detection

of magnetic fluctuations occurring at AFM transitions.

In contrast, for the 0 μ s delay case, we observe an enhancement of magnetic noise not only at the ferromagnetic transition but also at the AFM transition. This suggests that the laser interacts with CrCl_3 spins and selectively amplifies magnetic noise at the AFM transition (moderate-level noise enhancement can be seen in all temperature points). Our hypothesis to explain the laser induced noise peak at the AFM transition is that, in the presence of laser excitation, intralayer magnetic excitations (magnons) are populated by the laser and may couple to interlayer critical spin fluctuations. This coupling could amplify the fluctuations to a level detectable by the NV measurement. However, the precise mechanism underlying this effect remains an open question for both experimental and theoretical investigation.

Another observation is that the laser-induced excitation exhibits a characteristic decay time that varies with temperature (Fig. 2.14B). The decay rate of the laser-induced noise appears to peak at temperatures near both the AFM and FM phase transitions, which are also the temperatures at which the induced noise is high. This suggests that proximity to a phase transition, where intrinsic magnetic fluctuations are naturally enhanced, facilitates the relaxation of the laser-stimulated excitation. Indeed, when we plot the relaxation rate against the amplitude of the laser-induced noise, a positive correlation is observed.

We also examined the power dependence of the laser-stimulated noise. As expected, stronger laser excitation induces a higher magnetic noise amplitude, which eventually saturates at a specific power. Notably, the saturation value also exhibits temperature dependence. While the baseline noise level at zero power remains similar between 6 and 12 K, the material's response to laser excitation becomes more pronounced at higher temperatures.

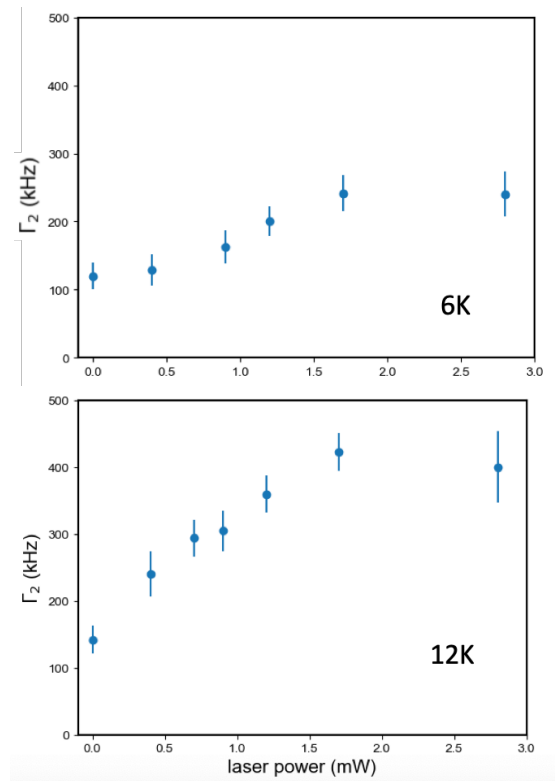


Figure 2.15: Amplitude of the laser-stimulated noise as a function of laser power. The noise amplitude increases with laser power and saturates between 1.5 and 2 mW. The saturated value is influenced by the sample temperature. The 0 mW point is the value of equilibrium noise taken after long enough delay time between the laser the spin echo sequence.

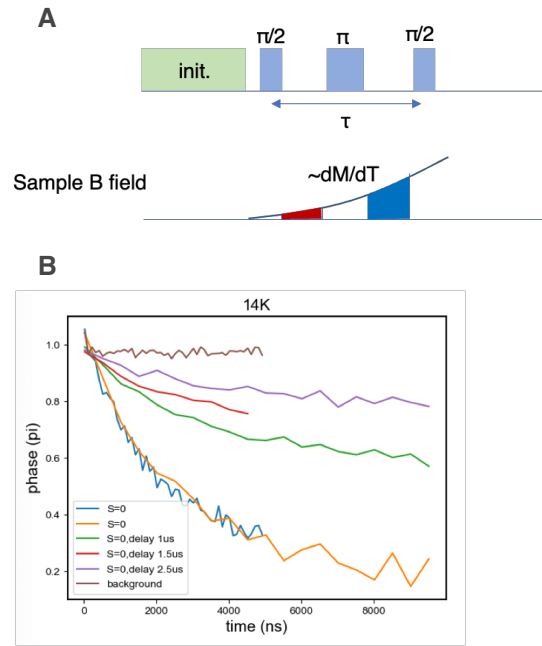


Figure 2.16: Laser-Induced Magnetization in the *Teardrop* Sample **A** Pulse sequence of the spin echo measurement, along with the inferred stray field from the sample based on our data. **B** Evolution of the spin superposition phase as a function of spin echo precession time. No phase is measured for bare diamond region, noted as background.

2.3.3 MAGNETIZATION INDUCED BY LASER EFFECT

As discussed in section 2.2.2, by reading out the spin projection along four axes (+x, -x, +y, -y), one can determine the phase of the spin superposition: θ (in the state $|0\rangle + e^{i\theta}|1\rangle$). At equilibrium of the sample, the phase of the spin echo measurement is expected to be zero, as any static magnetic stray field from the sample or external magnet is canceled out due to the equal-time but opposite-direction spin precession. However, we consistently observe a nonzero accumulated phase when no or short delay time is introduced between laser pulse and the spin echo sequence. This suggests that the laser stimulates a change in the sample's magnetic field (ΔB), which evolves on a timescale of microseconds (Fig. 2.16A).

The phase accumulated by NV centers arises from the temporal variation of the induced magnetic field, $\Delta B(t)$, during the NV sensing sequence. Upon arriving, the laser locally heats or excites the sample, leading to transient change of magnetization and a corresponding local modification in the stray field. As the sample cools and recovers its magnetization, the stray field gradually changes back to the original B_0 over time. By analyzing the phase accumulated during the spin echo sequence, we can infer the evolution of this magnetic recovery process.

For longer spin echo times τ , the accumulated phase difference increases and eventually saturates at a maximum value. The phase picked up by the NV centers is given by

$$\gamma \left(\int_0^\tau B(t) dt - \int_\tau^{2\tau} B(t) dt \right), \quad (2.23)$$

where $B(t)$ is the time-dependent magnetic field. A simple model for this relaxation process assumes an exponential recovery of the stray field,

$$B(t) = B_0 - \Delta B e^{-t/T_{\text{relaxation}}}, \quad (2.24)$$

as illustrated in Fig. 2.16.

The measured phase with an added delay during the initial period of magnetization evolution aligns with our proposed picture (Fig. 2.16B). The introduction of a delay causes the echo sequence to miss the initial rapid change in the magnetic field, leading to an overall lower accumulated phase. Furthermore, in regions where the sample is not in close proximity to the NV centers, we observe no phase accumulation, reinforcing our interpretation that the detected phase shift originates from changes in the sample's magnetization, rather than the imperfection of the pulse sequence or experimental settings.

Similar experiments were independently performed by Brian Zhou's group at Boston College¹⁰⁹. Their study, which carefully modeled the effects of laser excitation on magnetization, arrived at a similar conclusion: optical photon with the energy above band gap could induce extra sample's magnetization.

To completely eliminate the laser effect on CrCl_3 , one possible approach is to utilize total internal reflection at the diamond surface to read out the NV signal from the side of the diamond¹². This would ensure that the material stacked on the diamond remains free from direct laser excitation. Another experimental scheme involves reading the NV signal from the back of the diamond while introducing a reflective metal thin film between the diamond surface and the material. Although this setup would prevent direct optical imaging of the material under a confocal microscope, the weak magnetic field screening effect of thin metal films suggests that NV-based magnetometry should still be feasible. However, it is important to consider that the thickness of the metal film will contribute to the NV-to-sample distance, which may affect sensitivity.

*We need to convince ourselves first—and that’s usually the
hardest part, even harder than convincing the referee.*

Amir Yacoby

3

Discovery of Magnon Hydrodynamics

THIS CHAPTER PRESENTS AN EXCITING DISCOVERY: THE REALIZATION OF A HYDRODYNAMIC REGIME IN A DENSE MAGNON GAS. In solid-state physics, hydrodynamic transport refers to a strongly scattering regime where momentum is conserved in particle transport, leading to distinctive collective dynamics and temperature-dependent behavior. Hydrodynamic transport has emerged as a promising area of research, with growing experimental evidence in electron systems^{87,52,4,22,68}.

Due to the universality of hydrodynamic principles across physical systems, this topic is also of broad interest to the general physics community.

3.1 MAIN RESULTS FROM NOISE MEASUREMENTS

In this section, we present the main results observed when placing NV centers in close proximity to the CrCl_3 sample, the coherence properties of the NV centers are influenced by the presence of the magnetic sample and its spin dynamics. By interpreting the magnetic signals detected by our sensors, we can infer the underlying physical phenomena occurring within the sample. This provides a non-invasive method to study the intrinsic properties of a material system, particularly after carefully characterizing and mitigating the laser-induced effects through optimized measurement sequences.

As suggested by the preliminary Ramsey and T_2 measurements in Section 2.1, we expect to observe anomalous magnetic noise scaling with temperature. Indeed, this effect has been consistently reproduced across 8–9 different samples, and various thicknesses.

3.1.1 MONOLAYER

Monolayer CrCl_3 measurements under a small bias field (4 mT) are shown in Fig. 3.1. The transition temperature of monolayer CrCl_3 has been reported to be approximately 12 K⁷, a value independently confirmed by our static magnetometry measurements (Fig. 1.4A).

In all three measurements, the sample-induced decoherence increases as the temperature decreases below the phase transition temperature, continuing this trend down to the lowest measurement temperature. In addition to this unexpected low-temperature behavior, pronounced peaks appear near the transition temperature, between 11–14 K. The fine double-peak structure in the middle plot, as well as the broader features in regions 1 and 3, may arise from inhomogeneity in tran-

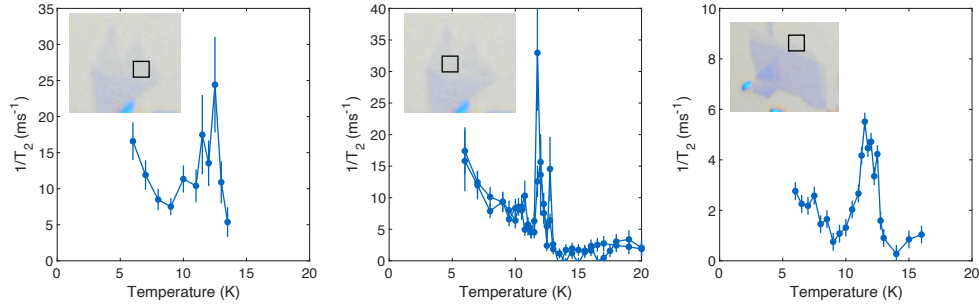


Figure 3.1: Spin decoherence measured on Monolayer CrCl_3 flakes Dephasing measurements with 4mT out of plane field on various regions and samples after background subtraction. Region 3(right most plot) shows the same phenomenology as region 2(middle) and 1(left), but a much lower signal. This is likely due to the fact that the sample of region 3 is so narrow that part of the confocal averaging area lies off the sample.

sition temperatures of a few percent on length scales of the sensing spot.

The peak in noise near the phase transition is likely a result of critical fluctuations⁶⁰. For the 4 mT applied field, the magnetic field direction is aligned along the sample surface normal, preserving in-plane spin rotational symmetry. Across the phase transition—whose exact nature remains debated—this preserved symmetry corresponds to a gapless excitation mode at the critical point. Such a mode can exhibit strong fluctuations at low energies (hundreds of kHz to MHz), which are detectable by NV spin-echo measurements.

This feature is suppressed under a stronger applied magnetic field(Fig. 3.2), which may result from the explicit symmetry-breaking effect of the planar component of the field(136mT along NV orientation, 54° from surface normal). The energy gap in spin excitation spectrum induced by around 100 mT in-plane field component corresponds to approximately 2.7 GHz. Although this gap remains small compared to the thermal energy at 12 K (250 GHz), the thermally induced magnetic fluctuations become off-resonant with our spin-echo measurement, leading to reduced sensitivity to these fluctuations.

Away from the phase transition, magnetic noise persists and even increases as the sample is cooled

to the lowest measurement temperature. This phenomenon is consistently observed in all monolayer samples at both low field (4 mT, Fig. 3.1) and at high field (136 mT, Fig. 3.2). Surprisingly, the amplitude of magnetic noise, quantified by $1/T_2$, is reduced by only 20–30% at high field compared to the low-field measurement at the same temperature. In contrast, as discussed earlier, a 30-fold increase in the bias magnetic field changes the magnon gap from approximately 100 MHz to 2.7 GHz, shifting the transverse spin fluctuations from the signal magnon band much further off-resonance with the spin echo filter frequency (100 kHz–10 MHz). This indicates that the observed magnetic noise must originate from mechanisms that are largely insensitive to changes in the magnetic field.

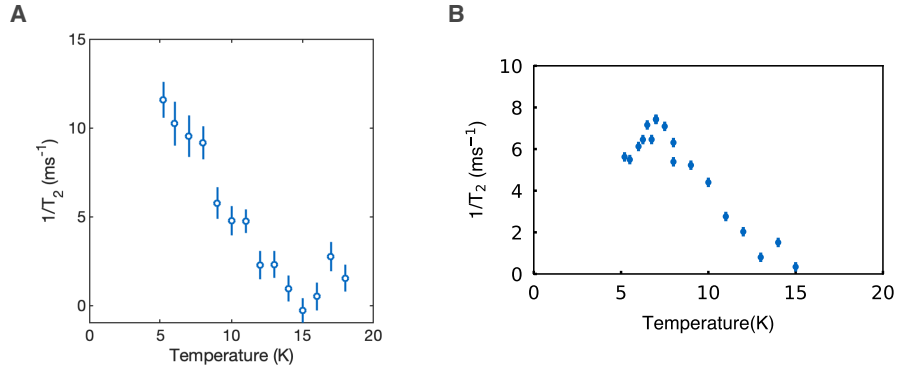
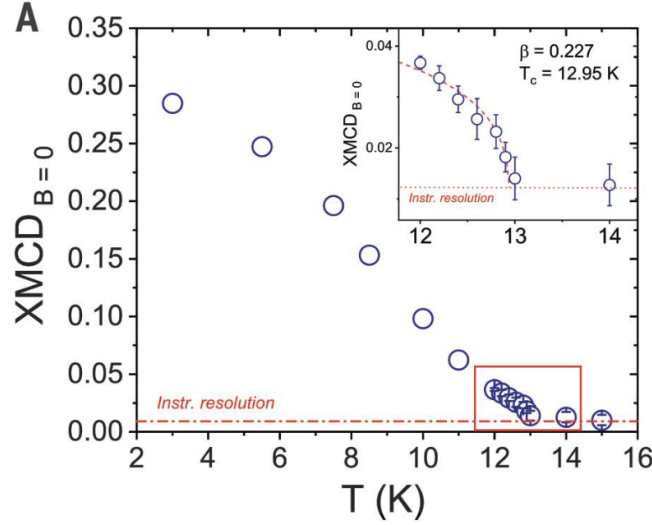


Figure 3.2: Spin decoherence induced by Monolayer CrCl_3 at 136mT Dephasing measurements with 136mT field aligned to NV direction measured on the same regions corresponding to region 1 and 2 in Fig. 3.1 A-B.

3.1.2 DISCUSSION ON MONOLAYER PHASE TRANSITION

The nature of the phase transition in monolayer CrCl_3 remains elusive to us. Experiments conducted by Stuart Parkin’s group using x-ray magnetic circular dichroism (XMCD)⁷ suggest that, upon extracting the remaining local magnetization from hysteresis measurements, the magnetization signal at $B = 0$ follows a 2D XY spin model’s scaling with a fitted exponent $\beta = 0.227$. This value contrasts with the critical exponent expected for a 2D Ising model, $\beta = 0.125$. CrCl_3 suggests an exciting platform for studying the spin XY model and the associated topological excitations^{58,3}.



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Figure 3.3: Critical behavior of magnetization. Remaining XMCD signal at $B = 0$ of CrCl_3 monolayer on graphene/6H-SiC(0001), extracted from hysteresis XMCD measurements. The magnetization near the tricritical point can be fitted with a critical exponent $\beta = 0.227$, consistent with the 2D XY model. Copyright to Science 374, 6567 (2021): 616–620.

Notably, this scaling exponent aligns with the experimental data only within a narrow temperature range, approximately between 12–13 K. At lower temperatures, the magnetization behavior deviates from the 2D XY model predictions, indicating that additional factors may influence the magnetic properties outside this temperature window. It is plausible that the system initially exhibits XY model characteristics just below the critical temperature (T_c). However, upon further cooling, shape-related dipolar anisotropy or material strain may become significant, altering the ground state of CrCl_3 from a state lacking long-range order to an ordered state. This transition changes the nature of the elementary excitations from vortex-type topological excitations to conventional spin waves (magnons).

We observe ferromagnetic order in CrCl_3 samples ranging from multilayers to monolayers, in the temperature range of 5–12 K. As discussed in Section 1.4.4, static magnetization maps obtained via NV ensemble measurements show no clear signatures of static vortices in the system; instead,

the maps are consistent with uniform magnetization (Fig. 1.3). Furthermore, the temperature dependence of magnetization between 5–12 K agrees well with predictions from spin wave theory under a 130 mT bias field (Section 1.4.4). Notably, even in the absence of an external in-plane field, shape-induced dipolar anisotropy can break the in-plane symmetry, leading to an ordered state in monolayer CrCl_3 .

To more accurately determine the ground state of CrCl_3 under zero bias field, it is essential to employ probes with higher sensitivity and spatial resolution to resolve finer magnetic structures. Conducting unperturbed magnetization measurements with fine temperature resolution would also be beneficial. For instance, scanning SQUID susceptometers or scanning single-NV magnetometry have demonstrated the capability to quantitatively probe magnetization, and magnetic domains. These methodologies could provide more precise insights into the intrinsic magnetic properties of CrCl_3 without introducing external perturbations.

Although the system may exhibit hints of XY physics, as indicated by data collected by the parkin group, this physics is extremely susceptible to both small external bias fields and shape anisotropies of the sample under study. These extrinsic factors very easily force the system into a conventional ferromagnetic state, as we observe in our measurements.

3.1.3 MULTILAYER CrCl_3

We also performed magnetic noise measurements ($1/T_2$) on thicker CrCl_3 samples, ranging from bilayers to 50-layer stacks. The key difference compared to monolayer flakes is the presence of interlayer antiferromagnetic (AFM) coupling between layers of ferromagnetically ordered spins. As a result, CrCl_3 exhibits two distinct magnetic phase transitions: an interlayer AFM transition at $T_N \approx 14$ K and an intralayer ferromagnetic (FM) transition at $T_C \approx 17$ K^{15,65}. Given this richer phase structure, we expect to observe additional features in the temperature-dependent measurements of magnetic noise.

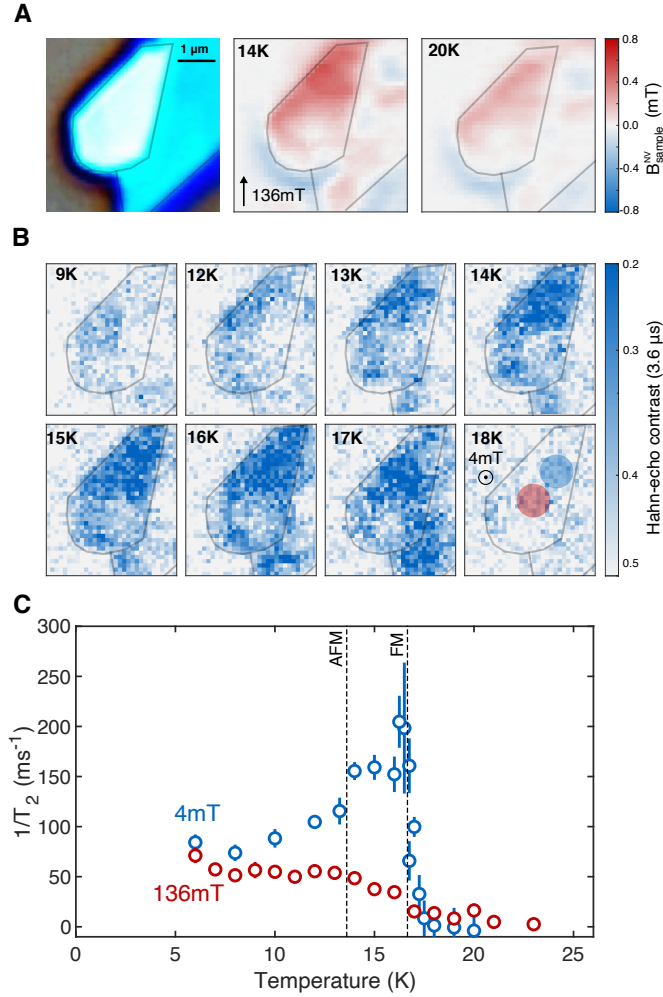


Figure 3.4: Magnetic noise measured on *teardrop* sample. **A.** Optical image of a thin bulk CrCl_3 flake alongside stray field maps measured at 14 K and 20 K under a 136 mT magnetic field applied 54° from the sample normal. The grey contour outlines the sample edge. **B.** Spatial map of dephasing spectroscopy under a 4 mT out-of-plane field. The color scale represents Hahn-echo NV contrast with a total evolution time of 3.6 μs . **C.** Temperature dependence of the NV decoherence rate ($1/T_2$) measured at selected regions indicated in panel B. These measurements reflect magnetic noise on the *Teardrop* (45-L) CrCl_3 flake. Intrinsic decoherence from the diamond spin environment has been calibrated and subtracted. Red markers denote data taken at 136 mT, while blue markers correspond to 4 mT.

Among the wide range of samples studied, the most representative data set is shown in Fig. 2.4 (additional examples are discussed later). In the temperature-dependent 2D maps of decoherence (Fig. 2.4B), a clear loss of NV center coherence is observed beneath the *Teardrop* sample. This is evident from the strong spatial correlation between the noise distribution and the sample boundary, indicated by the light gray contour.

As the temperature increases, the decoherence progressively intensifies, until the signal abruptly disappears above the Curie temperature ($T_C = 17$ K), marking the transition to the paramagnetic phase.

To better resolve the temperature dependence of the magnetic noise, we performed local $1/T_2$ measurements at selected points on the sample (Fig. 2.4B, 18K plot) while sweeping the temperature (Fig. 2.4C), under both 4 mT and 136 mT bias fields. Similar to the monolayer case, as the sample is cooled from 20 K—well above T_C —there is little excess noise within the frequency range of the NV echo filter function. A sharp increase in noise is observed near the $T_C = 17$ K, particularly in the low-field (4 mT) case. This upturn is attributed to critical spin fluctuations associated with the ferromagnetic phase transition. In contrast, under the higher field of 136 mT, the in-plane component of the bias field explicitly breaks the spin rotational symmetry, opens up a spin excitation gap, thereby suppressing the ‘peak’ in magnetic noise.

For the 4 mT curve, we observe a plateau in $1/T_2$ between T_N and T_C . This relatively high noise level may again be attributed to a gapless goldstone mode associated with in-plane spin rotations. The antiferromagnetic (AFM) transition is marked by a downward step in the noise level, without any signature of critical fluctuations. The AFM locking of the spin in each layer may gap out the goldstone mode existing between 14 and 17 K, causing the sudden drop of the noise. One possible explanation for not seeing critical fluctuation in the noise spectrum is that the AFM order has a high spatial frequency of approximately $1/c \sim 1 \text{ nm}^{-1}$, where c is the lattice constant along the z -direction. Such short-wavelength modes are invisible to NV centers, whose sensitivity is peaked

around momentum $1/z_{\text{NV}} \sim 1/(50 \text{ nm})$. As the temperature is further reduced, $1/T_2$ gradually decreases until reaching a low-temperature plateau, with the trend hinting at an eventual reversal at the lowest temperature point. In contrast, under a 136 mT bias field, the magnetic noise associated with the phase transitions is significantly suppressed, while the low-temperature behavior remains largely unaffected.

The signal from multilayer samples exhibits more complex dynamics compared to the monolayer case, suggesting that interlayer antiferromagnetic (AFM) coupling contributes to the observed magnetic noise. Nevertheless, the persistent trend of increasing—or plateaued—magnetic noise upon cooling remains puzzling and not fully understood.

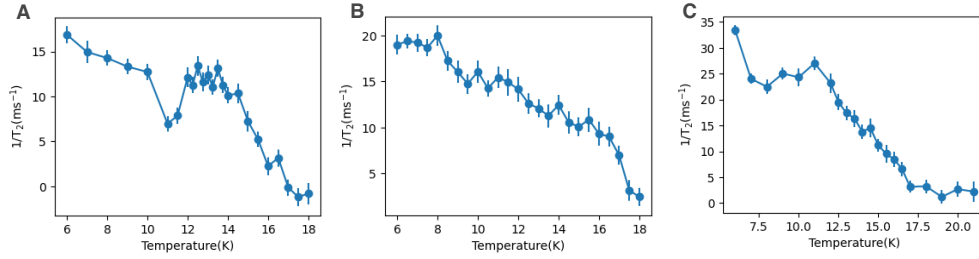


Figure 3.5: Other noise measured on multi-layer samples at 65mT. A. A bi-layer sample. B. A tri-layer sample. C. A 7-layer sample. All measured under a 64mT field aligned to the NV direction and no delay between the laser pulse and the spin echo sequence.

Additional data from samples of various thicknesses are shown in Fig. 3.5. Similar to the previous high-field measurements, the applied magnetic field suppresses critical fluctuations near the magnetic transition temperatures by opening a magnon gap. The overall rising trend in magnetic noise with decreasing temperature remains consistent with earlier observations. From monolayer measurements, we know that significant magnetic fluctuations can arise even in a single ferromagnetic layer. In multilayer samples, the noise amplitude does not scale linearly with thickness. This suggests that the interlayer antiferromagnetic (AFM) coupling leads to partial cancellation of magnetic noise contributions from adjacent layers, thereby reducing the overall observed noise.

We now turn to the development of a theoretical framework, in collaboration with our theorist colleagues, to explain the anomalous behavior consistently observed in CrCl_3 samples. This framework is based on the concept of hydrodynamic magnon transport, which we propose as a unifying explanation for the observed magnetic noise trends.

3.2 HYDRODYNAMICS IN A 2D HEISENBERG MAGNET

In this section, we discuss the development of magnon hydrodynamics theory, which provides a qualitative understanding of the experimental observations and, ultimately, an intuitive picture of the spin transport regime.

3.2.1 NON-INTERACTING MAGNON GAS

Given the good agreement between the spin-wave model and our temperature-dependent static magnetization measurements, and the extracted exchange coupling $J = 10.9 \text{ K}$ closely matching values obtained from other experimental methods (see Section 1.4.4), suggesting magnon (spin-wave) should be relevant particle when we try to address the puzzle of finite-frequency magnetic noise. Specifically, we proceed under the assumption that low-frequency magnetic fluctuations originate from magnons – how?

We calculated the magnon band dispersion using material parameters $J = 10.9 \text{ K}$, $\alpha = 1-0.7\%$, and a magnetic moment of $2.8 \mu_B$ per Cr atom at saturation magnetization⁷, along with the lattice constants of CrCl_3 . The resulting magnon dispersion follows a parabolic form (Fig. 3.6), with a gap determined by the applied external field: $\Delta_{\text{zeeman}} = \gamma_s B$, giving approximately 80 MHz for 4 mT and 2.7 GHz for 136 mT. This field-induced magnon gap exceeds the bandwidth of spin-echo sensitivity (inset of Fig. 3.6). To facilitate comparison between magnon energy and thermal energy, we also present the dispersion using temperature as a second axis, based on the relation $\hbar\omega = k_B T$.

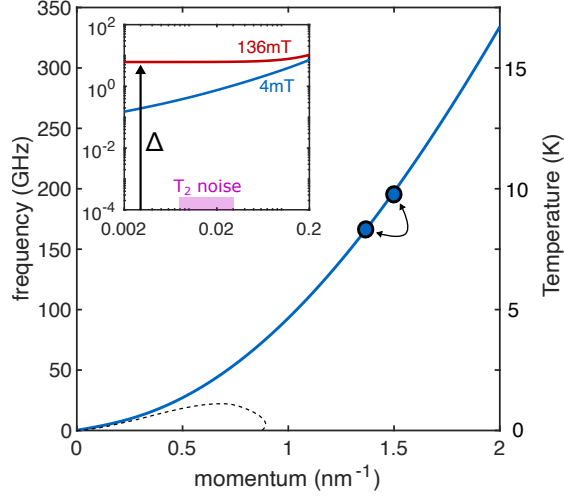


Figure 3.6: Magnon dispersion of monolayer CrCl₃ Calculated magnon dispersion for monolayer CrCl₃, including dipolar interactions. The magnon energy is labeled in both frequency (GHz) and temperature (K), related via $\hbar\omega = k_B T$. The inset highlights the T_2 sensing window in pink and shows the magnon band gaps under two different bias field conditions.

At our typical measurement temperatures (5–11 K), thermal magnons occupy frequencies in the range of 100–200 GHz, which are even further off-resonant from the T_2 sensing band. The noise contribution from damping of such off-resonant magnons can be estimated analytically.

Noise from magnons—Following the details in Xue *et al.*¹⁰², the magnetic noise contribution from single-magnon processes can be estimated (up to geometric prefactors of order one) as:

$$\mathcal{N}(\omega) \simeq \frac{(\mu_0 \mu_B g_s)^2 T}{\mathcal{A}_0 \omega} \int_0^\infty \frac{dq}{2\pi} e^{-2qz_{NV}} q^3 \frac{2S\gamma_q \omega \omega_q}{(\omega^2 - \omega_q^2)^2 + 4\omega^2 \gamma_q^2}. \quad (3.1)$$

Here, γ_q denotes the finite damping rate of magnons, and $\mathcal{A}_0$ is the unit cell area. The exponential factor accounts for the momentum filter imposed by the NV depth z_{NV} , favoring contributions from magnons with wavelength comparable to z_{NV} .

We focus on the low-frequency limit $\omega \rightarrow 0$, for which the noise expression simplifies to:

$$\mathcal{N} \approx \frac{2S(\mu_0\mu_B g_s)^2}{\mathcal{A}_0} \frac{T\gamma_{\text{qNV}}}{\omega_{\text{qNV}}^3} \int_0^\infty \frac{dq}{2\pi} e^{-2qz_{\text{NV}}} q^3, \quad (3.2)$$

where we assume the magnon dispersion ω_q varies slowly near q_{NV} (see Fig. 3.6). Using typical parameters: $T = 5$ K, $z_{\text{NV}} = 50$ nm, and $\gamma = 300$ MHz^{46,61}, we estimate $\mathcal{N} \simeq 0.06$ nT²/Hz for $B = 4$ mT and $\mathcal{N} \simeq 0.5 \times 10^{-4}$ nT²/Hz for $B = 135$ mT. Both are significantly smaller than the experimentally measured noise. Moreover, the experimental data shows only a factor of two difference between the data taken at 4 mT and 135 mT, while the single-magnon model predicts nearly two orders of magnitude difference.

Noise from longitudinal fluctuations—The longitudinal fluctuation of a magnon gas is characterized by the magnon density response function, $\chi_{zz}(\omega, q)$. Unlike transverse fluctuations, which correspond to oscillations in the direction of local magnetization due to the presence of magnons (captured by the terms $\chi_{+-}(\omega, q) + \chi_{-+}(\omega, q)$), longitudinal fluctuations describe changes in the *amplitude* of the local magnetization caused by variations in magnon density. Importantly, the characteristic frequency of longitudinal fluctuations is not tied to the single-magnon dispersion. To analyze this, we must evaluate the full response function.

The response function for longitudinal spin fluctuations of a non-interacting magnon gas is evaluated analytically in our paper¹⁰². In the regime of interest—where the frequency ω is smallest and the temperature is the largest energy scale—the resulting magnetic noise scales as:

$$\mathcal{N}(T) \propto \frac{T^2}{z_{\text{NV}}^3 \Delta^{3/2}}, \quad (3.3)$$

indicating that the magnetic noise from longitudinal fluctuation of non-interacting magnon gas increases quadratically with temperature. This can not simply explain our experiment.

3.2.2 STRONGLY INTERACTING MAGNON GAS

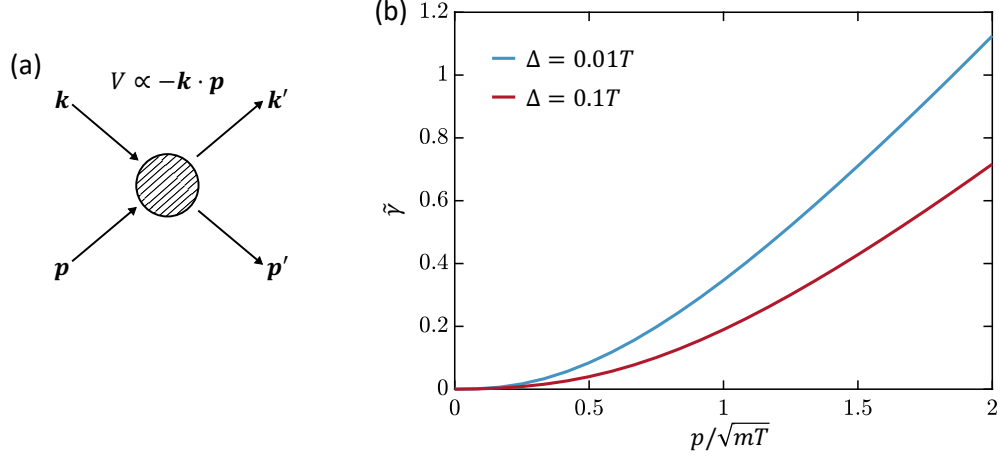


Figure 3.7: Exchange mediated magnon-magnon interaction. (a) The interaction vertex of a two-magnon scattering process due to the exchange coupling J . (b) The corresponding dimensionless magnon decay rate $\tilde{\gamma}$ as a function of momentum p . We find a strong momentum dependence highlighting that the low-momenta magnons barely-scatter off each other ($\tilde{\gamma}(p) \sim p^2$ for $p \rightarrow 0$), in contrast to the large-momenta ones.

When evaluating magnon–magnon interactions mediated by exchange coupling, we find that the magnon decay rate is momentum-dependent, as shown in Fig. 3.7(b). This behavior originates from the momentum-dependent interaction vertex, $V_{\text{exc}} \simeq -Ja^2 \mathbf{k} \cdot \mathbf{p}$, where J is the exchange constant and a is the lattice constant. This form of the interaction indicates that the coupling strength becomes significant at high momenta, implying that high-energy magnons experience stronger interactions and thus enhanced decay.

Including magnon–magnon interactions in the evaluation of magnon dynamics is essential for understanding this system. When these interactions are significant, the system may naturally enter the hydrodynamic transport regime. In this regime, the magnon gas is dominated by momentum-conserving scatterings, exhibiting fluid-like transport behavior and potentially supporting emergent excitations such as sound modes^{78,67}.

Before analyzing the magnetic noise in the magnon hydrodynamic regime, we first estimate the

scattering lengths associated with various mechanisms to validate the applicability of the hydrodynamic assumption in the dense magnon gas present in our system.

3.2.3 MEAN-FREE-PATH OF VARIOUS SCATTERING MECHANISMS

A hydrodynamic description requires approximate conservation of magnon number and that momentum-conserving collisions dominate over the relevant length and frequency scales. To evaluate this, we first estimate the single-magnon lifetime using the linewidth of ferromagnetic resonance (FMR)⁶¹. The inferred magnon damping rate is on the order of a few hundred MHz. Combined with the thermal magnon velocity, this yields a magnon propagation length of several tens of micrometers in the temperature range of 5–15 K. This propagation length is the longest among the relevant magnon scattering length scales, supporting the assumption of magnon number conservation within the measurement volume—a key prerequisite for applying hydrodynamic theory.

Furthermore, we estimate the relative importance of momentum non-conserving processes by comparing their mean free paths to that of momentum-conserving magnon-magnon (normal) scattering, which gives rise to hydrodynamic behavior. Specifically, we evaluate the temperature dependence of the mean free paths associated with thermal magnons undergoing magnon-magnon normal scattering, magnon-phonon scattering, and magnon-magnon Umklapp processes. Magnon-impurity scattering is difficult to estimate, as the impurity density is a microscopic, sample-dependent parameter that is hard to quantify. However, a favorable aspect is that the impurity density remains constant with temperature. Therefore, the corresponding scattering rate is also expected to be constant.

According to Ref.²⁸, the momentum-conserving magnon-magnon collision mean free path in a Heisenberg magnet can be crudely estimated as $l_{\text{mfp}} \simeq (J/T)^{3/2}/(na)$, where J is the exchange energy, T is temperature, n is the magnon density, and a is the lattice constant. Using this estimate, we find $l_{\text{mfp}} \simeq 18$ nm at $T = 5$ K and $l_{\text{mfp}} \simeq 3$ nm at $T = 10$ K. Since the magnon disper-

sion relation is subject to temperature renormalization effect, above 10K the mean-free-path value we acquire should also be renormalized. However, we want to argue that in order to see qualitative comparison between all mean-free-paths evolving as a function of temperature, we assume the existence of ferromagnetic order and the bare magnon dispersion and do the estimation up to 40K. Based on the magnon-magnon scattering mean-free-path, below we estimate the magnon-phonon and magnon-Umklapp mean free paths and compare all relevant length scales.

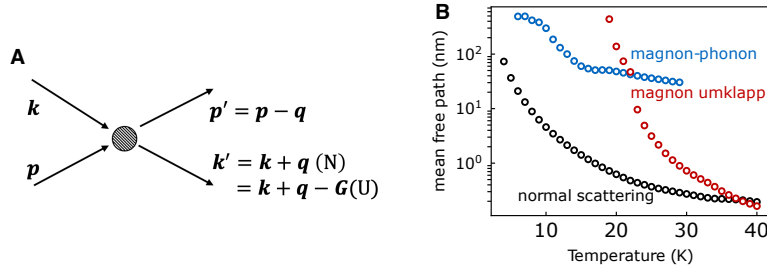


Figure 3.8: Mean-free-path of various scattering processes. **A** Depiction of the 4-magnon processes, including normal (N) and Umklapp (U) scatterings. **B** Magnon-phonon and magnon-magnon mean-free-paths estimated from non-renormalized magnon dispersion. Momentum-conserving magnon-magnon normal scattering (black) is the leading scattering process over a broad range of temperatures.

MAGNON-PHONON MEAN-FREE-PATH

To estimate the magnon-phonon mean free path, we utilize existing thermal conductivity data of bulk CrCl_3 from the literature^{75,53} and apply the Drude model of thermal transport. We assume that magnons in the bulk predominantly propagate within the two-dimensional layers, such that the magnon-phonon scattering rates in bulk are representative of those in a monolayer. This assumption is consistent with the conclusions of Ref.⁷⁵.

Refs.^{75,53} report measurements of thermal conductivity, κ , in bulk CrCl_3 under zero magnetic field and under an applied magnetic field of 18 T. A temperature-dependent enhancement of κ with increasing field strength is attributed to the depopulation of intralayer magnons. This reduction in

magnon population leads to a decrease in the phonon scattering rate, τ_{eff}^{-1} . The thermal conductivity is related to the phonon properties through the Drude model:

$$\kappa = \frac{1}{3} c_{\text{ph}}(T) v_s^2 \tau_{\text{eff}}(T), \quad (3.4)$$

where $c_{\text{ph}}(T)$ is the phonon specific heat and v_s is the speed of sound. From the difference in κ between 0 T and 18 T, we extract the change in phonon scattering rate, $\tau_{\text{eff},0\text{T}}^{-1} - \tau_{\text{eff},18\text{T}}^{-1}$, which we attribute primarily to the suppression of magnon-phonon scattering by the magnetic field. This yields an estimate of the magnon-phonon scattering rate in bulk CrCl_3 . Using the estimated magnon thermal velocity v_{th} and the measured $c_{\text{ph}}(T)$ from Ref.⁷⁵, we obtain a lower bound on the magnon-phonon mean free path—see Fig. 3.8.

MAGNON-MAGNON UMKLAPP MEAN-FREE-PATH

Magnon-magnon Umklapp scattering is a process that involves large momentum transfers by reciprocal lattice vectors $\mathbf{G}$, and typically occurs for high-energy magnons near the edge of the Brillouin zone (BZ). This scattering mechanism becomes relevant when magnons near the zone boundary are significantly populated, which happens when the thermal energy becomes comparable to the magnon bandwidth, J . In practice, studies on other magnetic systems^{80,30} have shown that Umklapp processes can dominate over normal magnon-magnon scattering above a crossover temperature, $T_{\text{crossover}} \sim 2J$. The precise ratio of $T_{\text{crossover}}$ to J can vary by factors of order unity, depending on lattice geometry, dimensionality, and the magnon dispersion relation.

Because the magnon-magnon Umklapp scattering occurs via the same mechanism as the normal magnon-magnon scattering, we can estimate the relative strength of Umklapp scattering by numerically evaluating the ratio of possible Umklapp and normal scattering configurations, see Fig. 3.8A. Both processes share the same interaction vertex, and thus the mean-free-path of Umklapp scatter-

ing can be obtained from the normal magnon-magnon mean-free-path multiplied by this ratio of allowed states:

$$r(\epsilon) = \frac{\# \text{Umklapp Scattering Config}}{\# \text{Normal Scattering Config}} \Big|_{\text{at energy } \epsilon}. \quad (3.5)$$

Numerical evaluation of r involves discretizing the BZ into finite momentum grids and traversing all possible momentum combinations $\mathbf{p}, \mathbf{k}, \mathbf{q}$. Each process that satisfies the energy conservation law will be classified into either Umklapp or normal scattering, depending on whether $\mathbf{k}'$ falls outside the first BZ or within it, respectively. The numerical simulations were run on an 11×11 grid within a single BZ, while the magnon band structure was taken from the self-consistent spin-wave theory.

The Umklapp mean-free-path is thus estimated as $l_U(T) = l_N(T)/r(\epsilon_{th})$, where $l_N(T)$ refers to the normal exchange mean-free-path. Figure 3.8B shows that for the experimentally relevant temperatures (5-15 K), the Umklapp scattering is sub-leading. In this calculation, we assume that the system remains an ordered ferromagnet at all temperatures shown in Fig. 3.8B. This assumption is made to explicitly demonstrate the crossover expected to occur between Umklapp and normal scatterings at higher temperatures.

Having established that exchange magnon-magnon interactions are the dominant scattering mechanism, we now proceed to evaluate whether a hydrodynamic description is consistent with the experimental observations.

3.2.4 HYDRODYNAMIC RESPONSE

We are increasingly confident in taking the hydrodynamic limit when evaluating the strongly interacting magnons, after establishing the estimation of mean-free-paths. The NV spin echo sequence, which detects magnetic noise as a dephasing effect, essentially captures the magnetic fluctuation of the hydrodynamic magnon bath.

According to the fluctuation-dissipation theorem, the fluctuation is related to the imaginary part of the spin system's susceptibility. We argue that transverse fluctuations can be neglected due to the large detuning between the NV sensing window and the magnon frequency. The noise can thus be expressed as (detailed theory derivation could refer to the supplementary material of Ref. ¹⁰²):

$$\mathcal{N}_{zz}(\omega) = \frac{(\mu_0 \mu_B g_s)^2}{16 \mathcal{A}_0} \coth\left(\frac{\omega}{2T}\right) \sum_{ab} \int_0^\infty \frac{dq}{2\pi} e^{-2qz_{NV}} q^3 \text{Im} \left[\tilde{\chi}_{zz}^{ab}(q, \omega) \right]. \quad (3.6)$$

where $\mathcal{A}_0$ is the unit cell area, a, b are sub-lattice indexes, z_{NV} is the distance from the NV center to the sample, and $\tilde{\chi}_{zz}(q, \omega)$ is the density-density correlation function of the hydrodynamic magnon gas.

Our theory collaborators carefully established the response function in the hydrodynamic limit:

$$\chi(\omega, q) = -\frac{q^2 n^2}{\omega^2 m n + i\mu\omega q^2 - q^2 n \frac{2\epsilon\omega n + icT\kappa q^2}{\omega n + ia\kappa q^2}}, \quad (3.7)$$

where m is the magnon mass, n is the magnon density, and

$$a \approx -\frac{3}{\pi^2} \ln(1-z), \quad b \approx \frac{1}{2}, \quad c \approx \frac{3(1-z)}{\pi^2} [\ln(1-z)]^2, \quad (3.8)$$

$$\epsilon \approx -\frac{\pi^2}{6 \ln(1-z)} T \quad (3.9)$$

where z is the partition function which is approximately 1 at the experimental temperature.

The newly introduced transport coefficient κ represents the thermal conductivity, while μ is the viscosity of the magnon gas. Estimations of κ suggest that it is sufficiently large that should tend to

infinity. Equation 3.7 becomes

$$\text{Im}[\chi(\omega, q)] = \frac{\omega q^4 \mu / m^2}{(\omega^2 - \tilde{v}_s^2 q^2)^2 + \omega^2 (\mu q^2 / mn)^2}, \quad (3.10)$$

This response function exhibits a resonance mode at $\omega^2 = \tilde{v}_s^2 q^2$, which is identified as the sound mode. The sound velocity is given by

$$\tilde{v}_s^2 \simeq -\frac{T}{m}(1-z) \ln(1-z) \approx \frac{\Delta}{m} |\ln(\Delta/T)|, \quad (3.11)$$

in which, Δ is the pseudogap of the thermal magnon (discussed in the following section 3.2.5). The sound velocity depends on the size of the pseudogap, the magnon mass, and weakly on temperature.

Since the frequency is still sufficiently small, we can take the limit $\omega \rightarrow 0$. In this limit, the imaginary part of the response function becomes

$$\text{Im}[\chi(\omega \rightarrow 0, q)] \approx \frac{\omega \mu}{m^2 \tilde{v}_s^4} \approx \frac{\omega \mu}{\Delta^2 \ln^2(\Delta/T)}, \quad \mathcal{N} \propto \frac{T \mu}{\Delta^2 \ln^2(\Delta/T)}. \quad (3.12)$$

Here, the viscosity μ determines the damping of the resonance mode.

We note that in the high-density regime of interest, where $z \approx 1$, the sound velocity scales as $\tilde{v}_s^4 \sim \ln^2(\Delta/T)$, which still grows with increasing temperature T (since $\Delta \ll T$). However, this logarithmic growth is weak. As a result, for the magnetic noise to decrease with increasing T , the product μT is allowed to increase with T no faster than $\ln^2(\Delta/T)$.

Notably, the exchange-driven magnon-magnon interactions yield a viscosity $\mu_{\text{exc}} \sim 1/T$, which is consistent with the experimental observations¹⁰². In contrast, if the interactions were of a contact type, the resulting viscosity would scale as $\mu_c \sim T$, leading to magnetic noise that *increases* with temperature—contradicting experimental results.

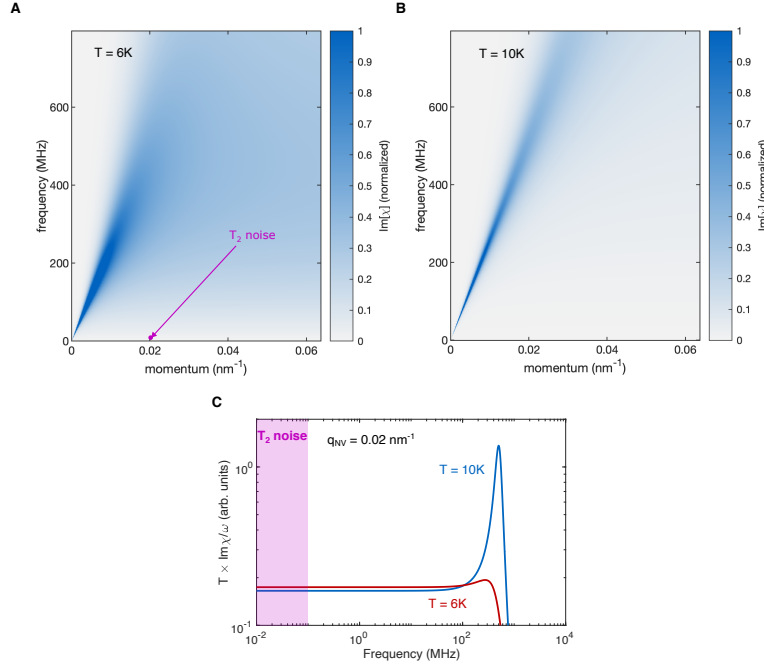


Figure 3.9: Hydrodynamic sound mode. Imaginary part of the density-density response function, Eq. (3.10), at $T = 6\text{ K}$ (A) and $T = 10\text{ K}$ (B). As temperature increases, both the sound velocity and the lifetime of the sound mode grow. Notably, the NV center's sensing window is significantly detuned from the sound mode resonance at q_{NV} . (C) The resulting probed magnetic noise can decrease with increasing T , as long as $\mu T \sim \text{const.}$

3.2.5 INTUITIVE UNDERSTANDING OF THE PHYSICAL PICTURE

Theoretical calculations suggest that in the hydrodynamic regime of a strongly interacting magnon gas, the low-frequency magnetic fluctuations exhibit an anomalous temperature dependence, consistent with our experimental observations. Specifically, we probe the low-frequency tail of the damping of the sound mode associated with magnon hydrodynamics. The damping is governed by the viscosity of the magnon fluid: in a denser and hence more viscous medium, the sound mode becomes more coherent and experiences less dissipation. As temperature increases, the system approaches a cleaner hydrodynamic regime, where reduced damping leads to a decrease in the observed magnetic noise.

The observation of viscous damping relies on several key conditions: (1) The measurement temperature is the highest energy scale in the system. In our case, $T \sim 200$ GHz, whereas the magnon gap is only a few GHz, and the NV probe frequency is in the MHz range. This relatively high temperature thermally populates magnon states to the high-momentum states near the Brillouin zone boundary, where momentum-dependent magnon-magnon scattering amplitudes are strongest (see Fig. 3.7). (2) Momentum-conserving magnon-magnon interactions dominate over momentum non-conserving processes, such as Umklapp and magnon-phonon scattering. In particular, Umklapp scattering remains subdominant even near T_c in CrCl_3 (see Fig. 3.8), preserving the hydrodynamic regime. This behavior contrasts with many other magnetic materials, where Umklapp scattering becomes the dominant process around $T \sim 2J^{80,30}$, thereby overwhelming hydrodynamic behavior. (Theoretically, $T_c = 4.5J$ for a 3D Ising model; in CrCl_3 , T_c is close to J due to its lower dimensionality and exchange strength.) (3) The NV-to-sample distance must lie within a specific window: it must be larger than the magnon-magnon scattering mean free path to ensure that transport within the sensing volume is hydrodynamic rather than ballistic. At the same time, it should be shorter than the magnon propagation length $l = \tau_m v_{th}$ —where τ_m is the thermal magnon lifetime and v_{th} is the average thermal magnon velocity—in order to avoid probing the decay of magnons.

The magnetic field dependence of the signal initially came as a surprise to us. Comparing the decoherence signals measured on a monolayer sample at 4 mT and 135 mT, we found that the signal strength changed by only about 20-30%, despite a 30-fold increase in magnetic field (Fig. 3.1 and Fig. 3.2). This observation motivated the introduction of the concept of a pseudogap, Δ , which had appeared in earlier theoretical discussions but lacked a clear definition.

We define Δ as an effective gap for high-energy thermal magnons, which is the most relevant part of the magnon excitations for hydrodynamics. The pseudogap primarily arises from the intrinsic magnon gap and the modification of the low-momentum magnon dispersion due to dipolar interactions. Fig. 3.10 illustrates this idea: the middle and right panels demonstrate how dipolar in-

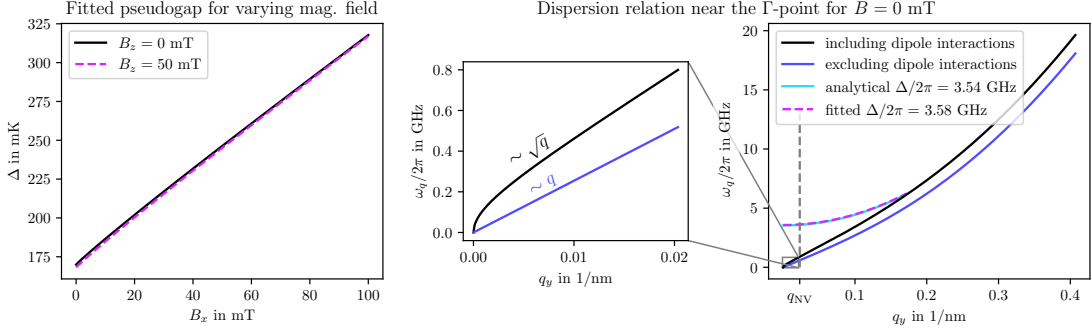


Figure 3.10: Linearized spin-wave theory. **Left:** Pseudogap Δ as a function of the applied field. We find that (i) $\Delta \approx 175$ mK at $B = 0$, (ii) it increases by less than a factor of two as B_x is increased from 0 to 100 mT, and (iii) Δ remains approximately independent of B_z . **Middle:** Zoom-in of the magnon dispersion near the Γ -point reveals that dipolar interactions significantly modify the low-momentum magnons. **Right:** Magnon dispersion for $B = 0$ near the Γ -point. At large momenta, the magnons are minimally affected by dipolar interactions, and their dispersion is well approximated by a quadratic form, as shown by the parabolic fit (dashed pink line). The extracted value of Δ agrees well with the analytical prediction (cyan line).

teractions significantly alter the low-momentum magnon spectrum, leading to an effective gap that persists even for high-momentum magnons. And the left panel shows a numerical evaluation of Δ , which changes by less than a factor of two as the in-plane field B_x is increased from 0 to 100 mT.

This effective gap, Δ , is particularly important because high-momentum magnons dominate the hydrodynamic regime and thus Δ being one of the physical quantity that directly influences hydrodynamic properties such as the sound velocity.

3.2.6 FITTING TO EXTRACT MAGNON VISCOSITY AND PSEUDOGAP

We fit the $1/T_2$ data within the ferromagnetic state to Eq. 3.12, assuming a temperature-dependent viscosity of the form $\mu = \mu_k/T$, where μ_k is a constant. This yields the fitting expression:

$$\frac{1}{TT_2} = \frac{A}{T\Delta^2 \log^2(\Delta/T)}, \quad (3.13)$$

with two free parameters: the pseudogap Δ and the prefactor A . We express A in terms of the NV-to-sample distance z_{NV} and μ_k as $A = C\mu T/z_{\text{NV}}^4$, where the coefficient C captures geometric and material constants. Specifically,

$$A = \frac{1}{k_B} \gamma_{\text{NV}}^2 \frac{3(3 + \cos^2 \theta_{\text{NV}})}{128\pi z_{\text{NV}}^4} (\mu_0 \mu_B g_s)^2 \mu_k. \quad (3.14)$$

Here, $\theta_{\text{NV}} = 54^\circ$, $z_{\text{NV}} = 65 \text{ nm}$ is the NV-to-sample distance, μ_0 is the vacuum permeability, μ_B is the Bohr magneton, $g_s \approx 2.1$ is the Landé g -factor in CrCl_3 , γ_{NV} is the NV spin gyromagnetic ratio, and k_B is the Boltzmann constant. In the case of 4 mT bias field, the signals from all four NV orientations are averaged, but since each NV experiences the same angle with respect to the magnetization in CrCl_3 , we use a fixed $\theta_{\text{NV}} = 54^\circ$ for the analysis. The viscosity at each temperature T is then extracted from the fitted A by converting to μ_k via Eq. 3.14, and computing $\mu = \mu_k/T$.

We note that, while the theoretical expression fits the experimental data well, the extracted value of μ is highly sensitive to uncertainties in the NV-to-sample distance z_{NV} and other microscopic parameters of the magnon system. In particular, the precise dependence of the sound mode velocity v_s on microscopic parameters (see Eq. 3.11) significantly affects the extracted μ , as the noise scales to the 4th power with v_s . The fitted pseudogap Δ primarily captures the temperature dependence of the signal rise and agrees within an order of magnitude with the theoretical prediction.

3.3 COMMENT: NV AS A PROBE FOR SPIN SUSCEPTIBILITY

The magnetic noise — equivalent to the phase fluctuations that lead to NV decoherence — is directly proportional to the imaginary part of the spin susceptibility, $\text{Im}[\chi(q, \omega)]$, of the system. This susceptibility is probed through a momentum filter determined by the spatial location of the NV center relative to the sample, and a frequency filter set by the specific measurement sequence. Our work thus demonstrates how NV center coherence can be used to probe magnetic fluctuations

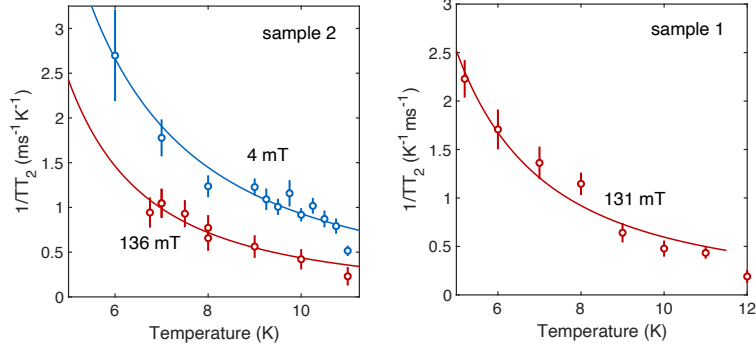


Figure 3.11: Fit to monolayer $1/TT_2$ using viscosity model. Sample 2 has fit parameters $A = 55 \pm 10 \text{ K}^2/\text{ms}$ and $\Delta = 1.2 \pm 0.2 \text{ K}$ at 4mT and $A = 40 \pm 10 \text{ K}^2/\text{ms}$ and $\Delta = 1.7 \pm 0.7 \text{ K}$. Sample 1 has fit parameters $A = 35 \pm 10 \text{ K}^2/\text{ms}$ and $\Delta = 1 \pm 0.4 \text{ K}$.

governed by $\text{Im}[\chi(\mathbf{q}, \omega)]$, enabling inference of magnon transport properties:

$$\mathcal{N}(\omega) \sim \frac{T}{\omega} \int d\mathbf{q} e^{-2qz_{\text{NV}}} q^3 \text{Im}[\chi(\mathbf{q}, \omega)]. \quad (3.15)$$

Beyond magnetic noise sensing, NV centers can also probe the spin susceptibility through an alternative technique first introduced by the Zhou group¹¹⁰. The method relies on synchronizing a modulated magnetic signal with a dynamical decoupling sequence applied to the NV center. Under this lock-in scheme, the NV spin phase accumulation reflects the response of the sample to the modulated drive. By varying the relative phase between the modulation and the pulse sequence, one can extract the real and imaginary components of the susceptibility. In Zhou *et al.*, this technique was demonstrated on exfoliated CrBr_3 to measure its ac magnetic susceptibility—a signal typically too weak to be detected using conventional coil-based measurements.

NV centers have emerged as a versatile platform not only for probing the magnetization properties of two-dimensional magnets^{91,84}, and characterizing spin excitations^{114,26}, but also for studying the spin susceptibility and subgap dynamics in magnetic systems^{102,95}. In the next chapter, I will describe a more direct observation of the magnon sound mode in CrCl_3 , enabled by a newly devel-

oped NV-based experimental modality.

The whole floor is 5 Gauss.

- Amir

4

Direct Measurement of Magnon Second Sound

A KEY HALLMARK OF MAGNON HYDRODYNAMICS IS THE EMERGENCE OF COLLECTIVE EXCITATION MODES— THE MAGNON SOUND MODE. In conventional transport regimes, spin excitations behave either diffusively or ballistically, depending on scattering length scales. However, in

the hydrodynamic regime, strong momentum-conserving magnon-magnon interactions lead to the formation of long-lived collective modes, analogous to sound waves in fluids. One such collective excitation is the magnon sound mode, which manifests as a resonance in the longitudinal spin current response. Detecting this mode is a crucial step in experimentally establishing the hydrodynamic nature of magnon transport.

In this chapter, we report the direct spectroscopic observation of the magnon sound mode in the layered van der Waals magnet CrCl_3 . This observation was both remarkable and unexpected. By driving the system with a tunable-frequency AC magnetic field and probing the NV spin response, we identified a resonance feature near the theoretically predicted sound frequency. What makes this result surprising is that the sound mode lies far outside the typical frequency range of the NV sensor’s filter function. Despite this, the resonance was still visible in our measurement. This result provides a strong evidence for the existence of a hydrodynamic regime in CrCl_3 and opens the door to new experimental approaches to study spin hydrodynamics in low-dimensional magnets.

In the sections that follow, I describe the experimental setup used to detect this mode, the theoretical framework for understanding the signal, and the rising new puzzles to our future research.

4.1 HAHN-ECHO MEASUREMENTS WITH RF DRIVE APPLIED TO THE SAMPLE

In this section, we present the development of a new experimental method designed to directly probe the spectroscopic signature of magnon second sound predicted by theory.

4.1.1 HAHN-ECHO SEQUENCE WITH AND WITHOUT RF DRIVE

We use the Hahn-echo sequence to detect magnetic noise stimulated by an applied RF drive on the sample. The measurement consists of two interleaved sequences: one with the RF drive turned on and one with the drive turned off (Fig. 4.1). The total measured noise includes both thermal and

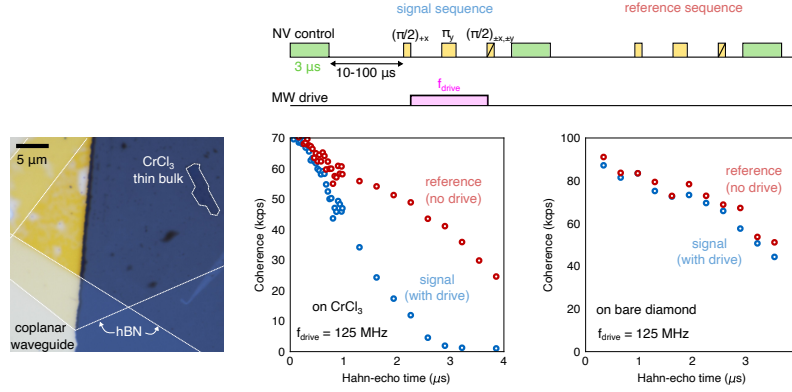


Figure 4.1: RF-driven spin echo measurement scheme. Optical image of the diamond surface, showing the CrCl₃ stack that we perform driven measurement, and with coplanar waveguide patterned on diamond. Driven measurements are performed using a sequence of signal (Hahn-echo with RF drive applied) and reference (Hahn-echo with drive off) experiments. Coherence is the remaining contrast of the NV spin, extracted by repeating the measurement four times, with the final $\pi/2$ pulse projecting along $\pm x$ and $\pm y$, followed by quadrature summation of the outcomes, reported in kilocounts per second (kcps). Example data are shown for a drive frequency $f_{\text{drive}} = 125$ MHz, drive field amplitude of 0.4 mT, and an out-of-plane bias field of 1 mT. On the CrCl₃ sample, the RF drive leads to enhanced NV decoherence, while no difference is observed on bare diamond. This contrast indicates that the drive induces additional magnetic noise in the sample, which is isolated by comparing the signal and reference coherence

drive-induced components: $N_{\text{total}} = N_{\text{thermal}} + N_{\text{stimulated}}$. The stimulated contribution, $N_{\text{stimulated}}$, is extracted by comparing NV spin coherence between the two sequences at a fixed Hahn-echo evolution time τ :

$$N_{\text{stimulated}} = -\frac{1}{2\gamma_{\text{NV}}^2 \tau^2} \log \left(\frac{C_{\text{drive on}}(\tau)}{C_{\text{drive off}}(\tau)} \right), \quad (4.1)$$

where $C_{\text{drive on}}(\tau)$ and $C_{\text{drive off}}(\tau)$ are the measured NV coherences with and without the RF drive, respectively. This relationship connects NV spin contrast to magnetic noise intensity. Equation 4.1 is equivalent to measuring the total noise with the drive on and subtracting the thermal background.

In all of our measurements, we observe that the stimulated noise scales linearly with the RF drive power (Fig. 4.2). Notably, the drive frequencies used (10–500 MHz) are far detuned from both the NV spin resonance (2.8 GHz) and the detection frequency of the Hahn-echo sequence (223 kHz). As a result, applying the RF drive to NV centers in bare diamond does not affect the measured co-

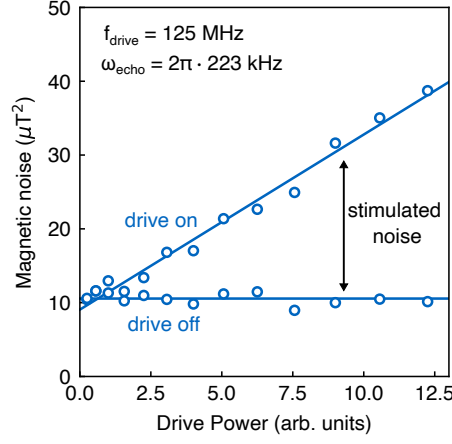


Figure 4.2: Stimulated magnetic noise scales linearly with RF drive power. Data shown were taken at a fixed drive frequency of 125 MHz and a fixed Hahn-echo precession time of 4.3 μ s.

herence (Fig. 4.1, on bare diamond). Additionally, our use of four-axis readout (by applying the last $\pi/2$ -pulse along four projection axes $\pm x, \pm y$) confirms that the RF drive induces only incoherent magnetic noise and does not cause a deterministic spin phase shift. This indicates that the observed decoherence stems from interactions between the RF drive and the NV environment—specifically, the response of the sample. Measurements on bare diamond regions reveal no significant response, except near the P1 center resonance, where the induced signal is more than an order of magnitude weaker than that from the sample.

4.1.2 CHARACTERIZING DIAMOND/SAMPLE JOULE HEATING DUE TO RF DRIVE

In these measurements, an RF drive current is applied to the coplanar waveguide for a few microseconds, which causes heating of the diamond surface. This heating must be carefully accounted for to (1) ensure accurate temperature values when reporting temperature-dependent data and (2) properly calibrate the noise signal induced by temperature changes. To address this, we use the intrinsic temperature dependence of the spin contrast of NV centers in bare diamond as an internal thermometer.

Specifically, the NV spin contrast exhibits a dip between 30–50 K and recovers at higher temperatures. This behavior, previously observed and attributed to thermal population of phonon modes that couple the NV ground and excited states³⁴, is linked to the photophysics of the NV center. It affects the spin contrast uniformly across different types of measurements (e.g., Rabi, echo, etc.), independent of the evolution time. Importantly, this temperature-dependent contrast is distinct from the temperature dependence of the coherence time T_2 , yet it influences all Hahn-echo measurements and must be taken into account when analyzing temperature-sensitive signals.

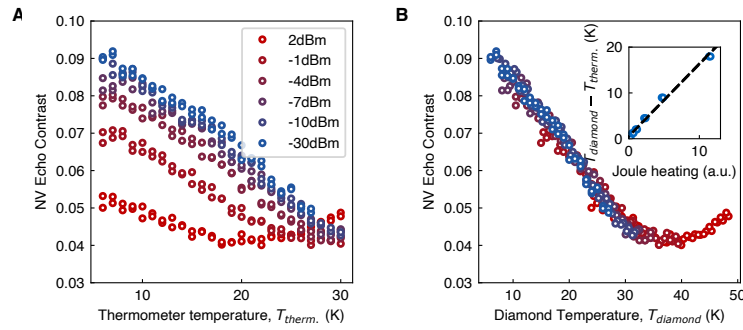


Figure 4.3: Characterizing heating of diamond substrate due to Joule heating of coplanar waveguide used to deliver RF drive. **A** Temperature-dependent measurements of NV spin-echo contrast on bare diamond as a function of the stage thermometer temperature. Drive on/off echo measurements (total evolution time 4.56 μ s, drive frequency of 125 MHz) are performed in succession. The Hahn-echo contrast for both measurements are plotted together for each value of RF drive power, and give essentially the same result. Points are labeled by the output drive power of the microwave source during drive on period. The power-dependence of the curve arises due to Joule heating generated by the coplanar waveguide, which causes the diamond to be at a higher temperature than that of the thermometer. **B** Local heating of the diamond can be accounted for with a shift to the thermometer temperature by an amount which is directly proportional to the drive-induced Joule heating generated by the coplanar waveguide as measured by a thermometer/heater PID loop.

A systematic dependence on RF drive power is observed in temperature-dependent Hahn-echo measurements on the bare diamond, where the drive stimulates no magnetic noise (Fig. 4.3A). The same effect is observed in sequential signal(drive-on) and reference(drive-off) measurements spaced by 20 μ s, indicating that this effect is due to heating of the diamond induced by Joule heating of the coplanar waveguide from the RF current. This dependence can be accounted for with a simple

thermal model of our diamond, heater, and thermometer system. Our sample platform contains a local thermometer/heater with a PID feedback loop; for a set temperature, at each drive power additional Joule heating is applied to the diamond, and the PID feedback loop quickly stabilizes the temperature of the thermometer to the set temperature by reducing the heater output. However, the temperature of the diamond surface and that of our thermometer can be different because the Joule heating of the coplanar waveguide induced by the drive is applied locally to the surface of the diamond, whereas the thermometer is placed on the metallic cryogenic sample platform, which has a finite thermal conductivity. Generically, $Q \propto \kappa(T_{\text{diamond}} - T_{\text{thermometer}})$, where Q is the average Joule heat applied to the diamond surface via the coplanar waveguide, κ is the thermal conductivity of the diamond and the gold-plated cryogenic sample platform separating the diamond surface from the thermometer. Q can be measured by monitoring the relative drop in the PID heater power (in units of percentage of our heater controller output) required to maintain the thermometer temperature when the drive is turned on – the value of Q for a given drive power was found to be temperature-independent over the measured temperature range. κ is assumed to be constant over the low temperature range of interest. With this in mind, the dependence seen in Fig. 4.3A can be systematically accounted for with a straightforward equation $T_{\text{diamond}} = T_{\text{thermometer}} + Q * A$, where A is a constant of proportionality. Indeed, temperature traces of the Echo contrast taken at different drive powers collapse together by adding a correction to the thermometer temperature, which is constant for each value of drive power (Fig. 4.3B). This relative temperature shift was found by least squares minimization between the curve with -30 dBm of drive power (essentially zero drive) and temperature-shifted data at each increasing value of drive power.

In further agreement with our thermal model, the temperature shift that best collapses the data is found to scale linearly with the measured Joule heating, Q , of the coplanar waveguide induced by the RF drive (with a proportionality constant 1.585 K/Q). Furthermore, a similar temperature-dependence is observed in successive drive on/off measurements (which are spaced within 20 μ s of

each other). Transient thermal effects can thus be neglected on these relatively short time scales, likely due to the long thermal timescale, ~ 1 s, of the diamond substrate. The data in Fig. 4.3B serves as a calibration to determine the temperature of the diamond for a given measurement of thermometer temperature and Joule heating. In all other RF drive data presented later, temperature refers to the diamond temperature as determined by this procedure.

To further check that this procedure is reliable in obtaining the diamond/sample temperature, we measure the transition temperature of the sample using driven Hahn-echo noise measurements with different RF drive duty cycles (Fig. 4.4), where the transition manifests as a peak in stimulated noise at a specific temperature. The duty cycle was changed via the time delay between laser initialization of NV centers and Hahn-echo measurement. After correcting for average Joule heating, Q for each RF drive duty cycle used, and using Fig. 4.3B as a calibration, we determine the diamond temperature for both traces. The transition temperature of the material obtained via the diamond temperature agrees well between these two measurements, with only a slight systematic difference in stimulated noise intensity. The transition temperature obtained using stimulated noise measurements with this duty cycle are in good agreement with the transition temperature obtained via static field measurements presented in the main text.

4.1.3 RF MAGNETIC FIELD CALIBRATION

At relatively low frequencies, the magnetic field measured at the diamond surface is expected to be proportional to the current flowing through the coplanar waveguide, and thus to the square root of the Joule heating. We verified this by measuring the AC magnetic field as a function of input power using NV centers in bare diamond—both at 10 MHz using an AC magnetometry sequence and at 780 MHz using the Rabi rate at a 130 mT bias field (Fig. 4.5).

To quantify the corresponding heating, we measured the heat delivered to the diamond, Q , as a function of input power at these two frequencies. This was done by monitoring the drop in heater

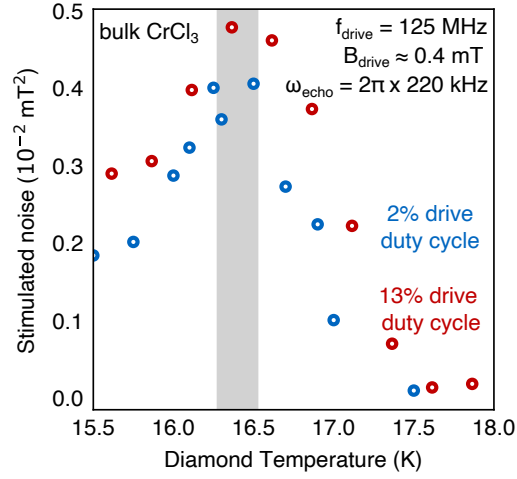


Figure 4.4: Measurement of bulk CrCl_3 ferromagnetic transition temperature using stimulated noise with two separate measurements with different duty cycles. The duty cycle was varied by changing the delay between the laser initialization and the Hahn-echo measurement. The Hahn-echo sequence and drive pulse are both $4.56\mu\text{s}$ long. The temperature axis represents the diamond temperature after measuring the average Joule heating and correcting the thermometer temperature using the procedure described in the text. The measured transition temperature here, as indicated by the grey bar centered where the signal is peaked, agrees well between the two measurements and with that obtained via static field measurements presented in Chapter 3.

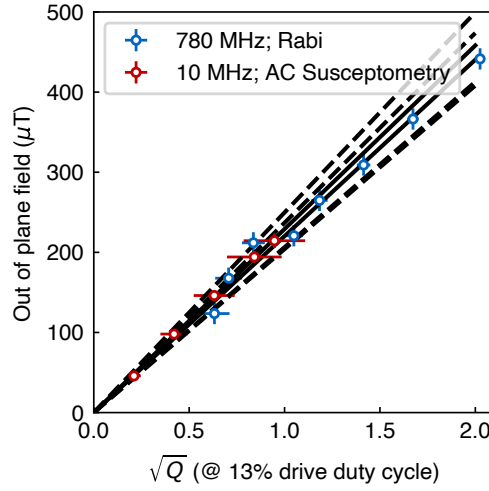


Figure 4.5: Calibrate the applied AC magnetic field. The out of plane AC magnetic drive field on the surface of diamond was measured as a function of input power using the NV centers via AC susceptometry at 10 MHz drive field, and at 780 MHz drive field via the Rabi rate at 130 mT bias field. The Joule heating generated by the coplanar waveguide on the surface of the diamond, Q , for a given RF frequency and drive power was measured by switching on the drive with 13% duty cycle and measuring the relative drop in PID heater power required to maintain the stage temperature at 12K. The magnetic field is found to scale with $\sqrt{Q}$ at both frequencies with a similar constant of proportionality ($220 \pm 20 \mu\text{T}/\sqrt{Q}$ at 10 MHz and $225 \pm 25 \mu\text{T}/\sqrt{Q}$ at 780 MHz), indicating that Joule heating of the coplanar waveguide on the diamond surface is the only relevant heating process. Using this calibration, measurement of Q (at 13% duty cycle) at a fixed frequency and power gives an indirect measurement of the RF magnetic field. The laser used in the NV magnetometry measurements was found to impart a negligible amount of heat on the sample stage. Black solid lines are best fits at each frequency with dashed lines being the standard deviation taking in to account error bars in the data.

power required to maintain the stage temperature at 12 K when the RF drive was turned on (13% duty cycle). We found that the proportionality between the measured magnetic field and $\sqrt{Q}$ was nearly identical at both frequencies within error bars. This confirms that Joule heating of the coplanar waveguide is the dominant heating mechanism at these frequencies.

This calibration allows us to estimate the local AC magnetic field generated by the waveguide at any drive power and frequency. Specifically, by measuring Q under a 13% duty cycle and using the empirically determined proportionality constant from Fig. 4.5 ($220 \pm 20 \mu\text{T}/\sqrt{Q}$), we can correct for variations in RF transmission. In practice, we adjust the microwave source output to keep Q constant across the range of drive frequencies, ensuring a consistent local magnetic field at the NV center.

4.2 SPECTROSCOPIC MEASUREMENT OF THE MAGNON SOUND

The drive-based measurement introduced in the previous section (sec 4.1) provides a method for probing the spectrum of low-frequency excitations. We apply this technique to bulk CrCl_3 in Fig. 4.1, which consists of approximately 45–50 anti-ferromagnetically stacked monolayers. Due to the significant frequency mismatch between the NV sensing sequence and the drive signal, a strong material response is required to produce a measurable signal. In contrast, monolayer or few-layer samples do not produce signals above the noise floor using the current method.

Nonetheless, we expect hydrodynamic magnon behavior to persist even in multi-layer samples when interlayer coupling is present. This is because the interlayer antiferromagnetic coupling is only about 1% of the energy scale of intralayer ferromagnetic exchange. As a result, the basic magnon dynamics should still resemble those of a monolayer, with additional complexity arising from interlayer interactions.

4.2.1 SPATIAL RESPONSE OF THE STIMULUS

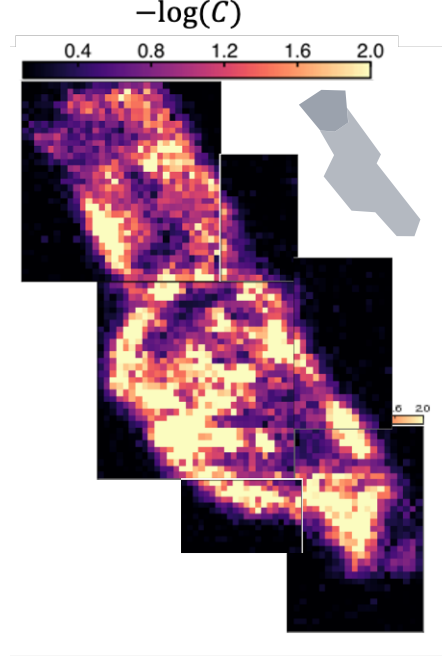


Figure 4.6: Spatial map of drive-induced noise measured on the 45–50 layer CrCl_3 sample at a drive frequency of $f_{\text{drive}} = 125$ MHz and Hahn-echo detection frequency $\omega_{\text{echo}} = 2\pi \cdot 223$ kHz. The upper-right inset shows the sample geometry. The color scale represents the drive-induced dephasing, quantified as $\langle \varphi^2 \rangle$, extracted from the normalized NV spin contrast C . Here, C is defined as the contrast measured at echo time $\tau = 4.3$ μs with the drive on, normalized by the contrast from the same sequence with the drive off.

This spatial map, taken at a fixed drive frequency $f_{\text{drive}} = 125$ MHz, shows the spatial dependence of the stimulated magnetic noise. As also observed in Fig. 4.1, regions with only hBN-covered diamond exhibit no response to the drive at 125 MHz, consistent with the insensitivity of the diamond-intrinsic spin noise to this drive frequency. Spatially, this appears as zero excess noise outside the sample boundary.

Within the CrCl_3 flake, we observe strong spatial variation in the stimulated noise, with the signal concentrated primarily near the sample edges. This spatial pattern may be related to how the RF drive couples momentum into low-frequency excited modes of the system. The sound mode at the

drive frequency is theoretically estimated to have a finite momentum $k \sim 0.01 \text{ nm}^{-1}$ (Fig. 3.9), whereas the microwave field generated by the coplanar waveguide—with a gap size of $30 \text{ }\mu\text{m}$ —carries nearly zero momentum. While the microscopic mechanism enabling this momentum coupling remains unclear, it could be influenced by the finite size of the sample and the presence of edges, whose Fourier Transform contains high spatial frequency components, may facilitate coupling between the uniform RF drive and finite-momentum modes. This could explain why the stimulated noise appears most prominently at the sample boundaries.

4.2.2 SPECTROSCOPIC RESPONSE OF THE STIMULUS

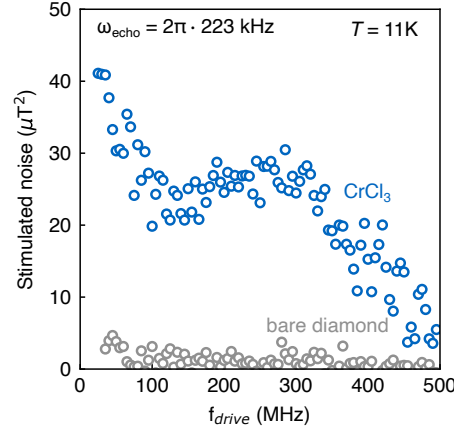


Figure 4.7: Spectroscopic response of the stimulating field on and off the CrCl_3 sample. The blue curve shows the measured response at a location on the CrCl_3 flake with relatively strong drive-induced noise. The NV center filter frequency is fixed at $\omega_{\text{echo}} = 2\pi \cdot 233 \text{ kHz}$, while the drive frequency is swept between 20 MHz and 500 MHz at a temperature of 11 K. Grey dots represent a reference measurement taken under identical conditions on bare diamond covered only with hBN. The stimulated noise is extracted from the contrast ratio between successive drive-on and drive-off sequences.

The measured spectrum is encouraging, particularly in the shape of the response curve on CrCl_3 sample. The signal from the sample region shows clear contrast compared to the hBN-covered bare diamond, indicating that the microwave drive stimulates spin excitations in the magnetic material. Closer inspection reveals two spectral components: a resonance-like feature around 300 MHz that

transitions smoothly into a low-frequency response below 100 MHz.

To better understand the measured spectrum, we performed a phenomenological fit using two components. The first is a resonance term inspired by the theoretical response function in Eq. 3.7. Since the dephasing signal is proportional to the variance of the magnetic field fluctuations, and those in turn are proportional to the square of the response function, we expect $\langle \phi^2 \rangle \propto \langle B^2 \rangle \propto \chi^2$. This leads to a resonance contribution from the sound mode that takes the form below

$$\frac{A_{\text{sound}}}{(f^2 - f_0^2)^2 + f^2 \gamma_{\text{sound}}^2}, \quad (4.2)$$

where A_{sound} captures the amplitude of the resonance peak, f_0 is the resonance frequency determined by the sound mode dispersion, and γ_{sound} is the associated damping rate.

The spectral feature observed below 100 MHz can be captured by adding a low-frequency Lorentzian component:

$$\frac{A_{\text{Lorentzian}}}{f^2 + D^2}, \quad (4.3)$$

where $A_{\text{Lorentzian}}$ represents the amplitude of this contribution, and D is a diffusion-related constant that sets the linewidth of the low-frequency mode.

Our spectrum (Fig. 4.7) can be decomposed to the two components with a 5-parameter fitting using $(A_{\text{Lorentzian}}, A_{\text{Sound}}, \gamma_{\text{sound}}, D, \omega_0)$. We took the same measurement at various temperatures and fitted with the same method to see the temperature evolution of the spectrum.

We observe that across a wide temperature range (6–17.5 K), the two-component fit captures the experimental spectra well at each temperature point (Fig. 4.8). Visually, the resonance mode becomes more pronounced from 6 K to 13 K, consistent with the expectation that the sound mode sharpens and becomes better defined as the system enters a deeper hydrodynamic regime at higher temperatures. Notably, above the transition temperature ($T_c = 17$ K for this and generally bulk

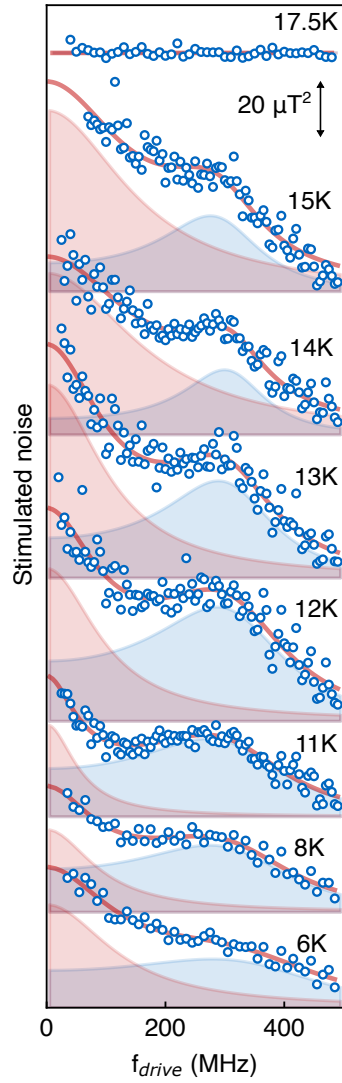


Figure 4.8: Spectroscopic response of the stimulating field on the CrCl_3 sample at various temperatures. The red shaded area represents the Lorentzian component, and the blue shaded area corresponds to the resonance component, both extracted from a five-parameter phenomenological fit to the measured spectrum. The red solid line shows the combined fitting result that best captures the data at each temperature.

CrCl_3), the stimulated noise drops to the noise floor and becomes featureless in its frequency dependence. This suggests that the spin excitations coupled to the microwave drive are strongly associated with the magnetic phase of the material.

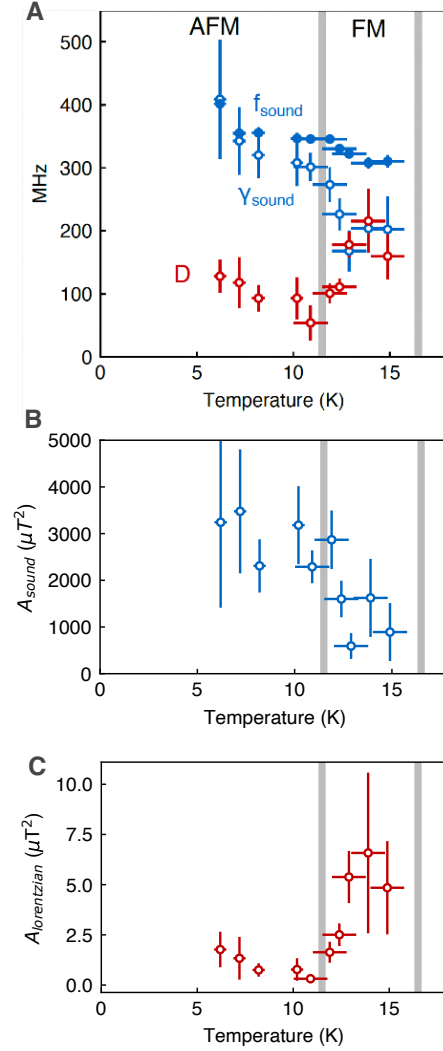


Figure 4.9: Fitting parameters of the temperature-dependent spectrum. **A.** Temperature dependence of the resonance frequency f_{sound} (solid blue line), the damping rate γ_{sound} (blue circles), and the width D of the Lorentzian component (red circles). **B.** Fitted amplitude of the resonance component, A_{sound} . **C.** Fitted amplitude of the low-frequency Lorentzian component, $A_{\text{lorentzian}}$. In all three panels, the grey vertical lines mark the antiferromagnetic (AFM) and ferromagnetic (FM) phase transition temperatures. The x-axis error bars indicate uncertainty in temperature, as determined by the method described in Section 4.1.2.

The fitting parameters shown in Fig. 4.9 exhibit distinct temperature dependencies for the resonance and Lorentzian components, suggesting they may originate from different sources of magnetic fluctuations. In particular, both the width D and amplitude $A_{\text{Lorentzian}}$ of the Lorentzian component change abruptly across the antiferromagnetic (AFM) transition. The increase in amplitude and broadening of the mode suggest that interlayer AFM coupling may suppress this source of low-frequency noise.

We emphasize that the parameters extracted for the resonance component— γ_{sound} , A_{sound} , and f_{sound} —do not directly correspond to microscopic quantities of the sound mode. In the following section, we develop a quantitative description based on hydrodynamic theory to interpret the observed spectrum. In particular, the high-frequency drive is effectively down-converted to the echo detection window, allowing us to probe the mode off-resonantly. Furthermore, we will show that the fitting parameters obtained from the experimental spectra—such as A_{sound} , f_{sound} , and γ_{sound} —should not be used as a direct probe of the underlying physical properties of the magnon sound mode as described by hydrodynamic theory.

4.2.3 THEORETICAL MODELING OF THE NV SIGNAL

To relate the theoretical susceptibility of the magnon hydrodynamic regime [Eq. 3.7], particularly the sound mode that was first predicted in theory, to the signal measured by the NV centers, we must evaluate the full filter function that the NV sequence imposes on the environmental magnetic noise.

We begin by assuming that the microwave drive introduces a small perturbation to the magnon density:

$$\delta n(\mathbf{q}, \omega) = -\chi(\mathbf{q}, \omega)V(\mathbf{q}, \omega), \quad (4.4)$$

where $V(\mathbf{q}, \omega)$ represents how the drive couples to the sample spin. For simplicity, we assume $V(\mathbf{q}, \omega)$ to be constant with respect to $\mathbf{q}$, although in reality it could vary depending on the microscopic details of the coupling mechanism.

Second, we are applying a finite-length microwave pulse for the drive, which can be written as

$$f(t) = \sin(\omega_d t + \phi - \mathbf{q}_d \cdot \mathbf{r}_{\text{NV}}) \Theta(T_d/2 - t) \Theta(T_d/2 + t). \quad (4.5)$$

In the function of the drive, we want to address that there is phase randomness in the term $\phi - \mathbf{q}_d \cdot \mathbf{r}_{\text{NV}}$. The phase randomness comes from the fact that we spatially averaged over an ensemble of NVs, as well as the microwave phase drive can start with random phase ϕ . The Fourier transform of the finite length drive impose a frequency modulation on top of the drive frequency ω_d , the linewidth of the modulation is given by the echo sequence (length of the drive) $1/T_d$.

The stimulation-induced decoherence is then computed as an integral over momentum and frequency, reflecting both the driven noise generated by the sample and the filter functions imposed by the NV sensing protocol:

$$\begin{aligned} \langle \phi^2 \rangle = & 4Y^2 \int \frac{d^2 \mathbf{q}_1}{(2\pi)^2} \int \frac{d^2 \mathbf{q}_2}{(2\pi)^2} W_q^{\text{ZNV}}(\mathbf{q}_1, \mathbf{q}_2) V_{\mathbf{q}_1} V_{\mathbf{q}_2} \\ & \times \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} \chi(\mathbf{q}_1, \omega_1) \chi(\mathbf{q}_2, \omega_2) W_d(\omega_1, \omega_2) W_{\text{echo}}(\omega_1, \omega_2), \end{aligned} \quad (4.6)$$

where W_q^{ZNV} is the NV center's momentum filter function derived from dipolar coupling to the sample, and takes the form $W_q^{\text{ZNV}} = q^2 e^{-2qz_{\text{NV}}}$ ¹⁶. The function W_d represents the spectral broadening due to the finite duration of the microwave drive, while W_{echo} is the frequency-domain filter

function of the NV spin echo sequence¹⁴, given by:

$$W_{\text{echo}}(\omega) = \frac{\sin^4(\omega\tau/4)}{\omega^2}. \quad (4.7)$$

This formulation gives the honest evaluation of how the sample response couples to our sensor (may subject to order 1 geometry factors).

To build physical intuition behind the integral expression in Eq. 4.6, we examine the behavior of the key filter functions and susceptibility:

First, the presence of χ^2 rather than just the imaginary part of the susceptibility agrees with our phenomenological fitting, where the NV signal is modeled as proportional to the square of the response function. This reflects the second-order response to the driven field and is valid in the linear driving regime (Fig. 4.2). This is in contrast to the equilibrium noise measurement we discussed in Chapter 3, where we are probing the only imaginary part of the susceptibility.

The momentum filter function $W_q^{z_{\text{NV}}} = q^3 e^{-2qz_{\text{NV}}}$ arises from dipolar coupling and, after integrating over angles in 2D momentum space, peaks at a characteristic momentum scale $q^* \sim 1/z_{\text{NV}}$, where z_{NV} is the distance of the NV center to the sample. However, this is a broad function in momentum, meaning the NV does not probe a sharp momentum slice but rather averages over a range of q around q^* .

The frequency-domain filter function from the NV echo protocol,

$$W_{\text{echo}}(\omega) = \frac{\sin^4(\omega\tau/4)}{\omega^2}, \quad (4.8)$$

is a rapidly oscillating function that decays as $1/\omega^2$ for large ω . In our experiment, the microwave drive frequency ω_d lies in the hundreds of MHz, while the NV echo integration time τ is on the order of a few microseconds. This means the filter oscillates very rapidly at ω_d , and we can approxi-

mate its contribution by the envelope $\sim 1/\omega_d^2$. As a result, the NV measurement effectively samples the magnetic noise power at ω_{echo} induced by high-frequency driving, with $1/\omega_d^2$ suppression at higher drive frequencies due to the filter function.

By taking the $1/\omega_d^2$ envelope approximation for the frequency-domain filter $W_{\text{echo}}(\omega)$ at high driving frequency, the NV-observed signal simplifies to

$$\langle \phi^2 \rangle \propto \frac{1}{\omega_d^2} \int_0^\infty dq q^3 e^{-2qz_{\text{NV}}} \chi(q, \omega_d) \chi(q, -\omega_d), \quad (4.9)$$

where the integrand reflects the momentum filter and the squared susceptibility at the drive frequency ω_d .

However to fully capture the low frequency (the signal at $< 100\text{MHz}$) response, we still need to add a diffusive component described by

$$\chi(\omega, q) = \frac{iDq^2}{\omega + iDq^2}, \quad (4.10)$$

to get better alignment with the temperature-dependence data, in which the two components has different behavior as changing temperature.

4.2.4 DISCUSSION

We also consider the possibility that the observed resonance arises from antiferromagnetic resonance (AFMR), especially given that the driven measurements were performed under a small bias field of 1 mT. In bulk CrCl_3 , two types of AFMR modes exist—optical and acoustic—analogue to phonon classifications. The optical mode lies at approximately 10 GHz⁶¹, which is well beyond the frequency range of our measurement and therefore irrelevant in this context. The acoustic AFMR mode, on the other hand, has a frequency that increases linearly with the applied field. At the exper-

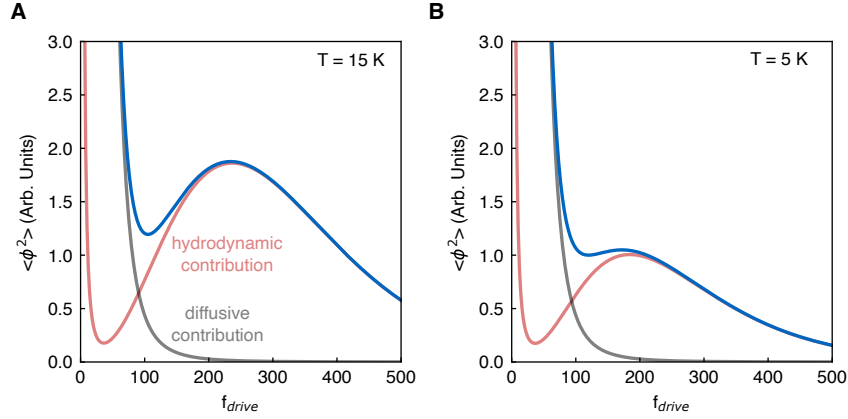


Figure 4.10: Theory for the driven experiment. Orange curves represent the evaluation of the momentum integral in Eq. (4.9) as a function of the drive frequency, for the hydrodynamic susceptibility in Eq. (3.10). We observe a hydrodynamic bump around $\omega_d \simeq v_s/z_{\text{NV}}$ and an enhancement of the signal for small ω_d . As temperature increases, this hydrodynamic contribution gets enhanced. The experimental data are better captured when a diffusive mode (grey curves) contribution is included, with the susceptibility given by Eq. (4.10).

imental field strength of 1 mT, the acoustic AFMR is expected to occur around 28 MHz⁶¹, roughly an order of magnitude lower than the resonance we observe. Furthermore, AFMR corresponds to the $k = 0$ spin wave mode, which is unlikely to couple efficiently to NV centers due to the momentum filtering effect imposed by the NV-sample distance, as described by Eq. 4.9.

Specifically, the momentum filter function favors modes around $q^* \sim 1/z_{\text{NV}}$, corresponding to excitation frequencies near 5 GHz (see Fig. 3.10). These modes are both far off-resonance from the applied microwave drive and strongly suppressed by the NV echo sequence’s frequency filter.

Another evidence that further excludes the direct detection of single-magnon bands comes from our field-dependent measurements (Fig. 4.11).

The driven measurements performed at various out-of-plane fields (1.1 mT to 7.2 mT) all exhibit nearly the same resonance frequency of 360 ± 20 MHz. This result contradicts the expected field-dependent behavior of the acoustic AFMR mode. Additionally, we measured the stimulated noise at 131 mT, a field strength at which magnons are expected to be gapped out, yet a signal is still

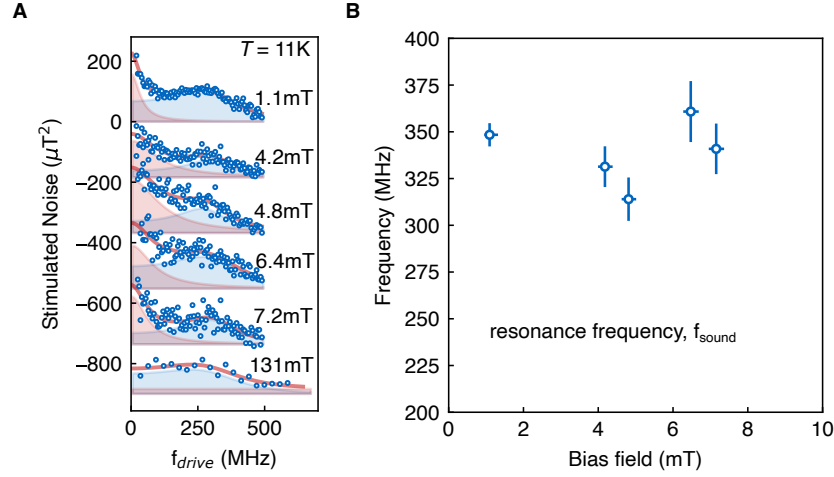


Figure 4.11: Magnetic field dependence of stimulated noise. **A** Stimulated noise spectra measured under different applied magnetic fields. The 131 mT dataset was acquired with the field applied along the NV axis (54° from the surface normal) and has been scaled by a factor of 100 for visibility. **B** Fitted resonance frequency as a function of out-of-plane bias field. The fitted resonance frequency at 131 mT (not shown in the plot) is 360 ± 20 MHz.

observed. Due to a different circuit configuration, the drive power in this measurement was approximately ten times lower. Nevertheless, a clear emergence of stimulated noise remains visible below 500 MHz, while the low-frequency component is significantly suppressed.

Figure 4.11 is plotted using a linewidth parameter of less than 1 MHz, whereas the calculated resonance detected by the NV centers appears significantly broader, on the order of ~ 200 MHz. This discrepancy is attributed to the broadening effect introduced by finite momentum filtering. Another important observation is that the $1/\omega_d^2$ prefactor for the diffusive component diverges more rapidly at low frequency than what is observed experimentally for the low-frequency component.

This theoretical calculation implies that the peak position of the resonance can be used to roughly estimate the velocity of the sound mode via $v_s \approx 2\pi f_{sound}/k_{NV} \sim 150$ m/s. This value, while representing a crude estimate, is within a factor of two of the theoretically predicted value for monolayer $CrCl_3$ (~ 300 m/s). The deviation could originate from the pseudogap difference between a multilayer sample (experiment) and the monolayer model that we evaluated (theory). Besides, the

theoretical estimation is based on the magnon properties of a bare magnon band. In contrast, the experiment is conducted in the magnon hydrodynamic regime, where strong magnon interactions are expected to renormalize the band dispersion³³, enhancing the effective magnon mass and slowing down the magnon sound mode. The extraction of viscosity μ and pseudogap Δ strongly relies on the precise knowledge of the effective magnon mass under strong renormalization, thus making it challenging to extract from the current measurements.

5

Other Unpublished Works

The work in this Chapter was conducted in collaboration with Dr. Chunhui Du (BSCCO project), Dr. Bart Machielse (diamond nanobeam), and Dr. Mark Ku (wide field imaging).

5.1 BEYOND MAGNETISM: LOCAL MICROWAVE RESPONSE OF SUPERCONDUCTORS

We also explored the possibility of characterizing the superfluid properties of a 2D unconventional superconductor using NV centers during the period of 2018–2019. The key idea was to

utilize the NV center as a nanoscale, local magnetic field sensor to directly image the current distribution within the superconducting sample. By mapping out the spatial profile of the magnetic fields generated by screening currents, one can reconstruct the underlying current flow patterns. This information enables the extraction of the Pearl penetration depth, a fundamental parameter that characterizes the superfluid density. Such a measurement offers a non-invasive and contact-free method to probe the superfluid density, which is particularly valuable for studying 2D materials where conventional bulk techniques are less effective. Similar work has been published around the same time of our investigation¹⁰¹.

5.1.1 EXPERIMENTAL METHODOLOGY

$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ (BSCCO) is a layered cuprate superconductor that retains its superconducting properties down to the atomically thin limit¹⁰⁵. Remarkably, the superconducting transition temperature (T_c) of exfoliated monolayer BSCCO remains comparable to that of the optimally doped bulk crystal, suggesting the robust 2D superconductivity of BSCCO and, more broadly, other layered cuprates with similar lattice structures.

In this investigation, our collaborator Dr. S. Y. Frank Zhao from Prof. Philip Kim's group exfoliated a 55-nm-thick BSCCO flake and transferred it onto a diamond substrate. The sample is a 20 μm wide, over 150 μm long strip placed in parallel to the microwave stripline pre-patterned on the diamond surface to deliver microwave for NV spin manipulation (Fig. 5.1A). This setup enabled local probing of superconducting properties through NV magnetometry.

The diamond surface contains shallow-implanted single NV centers, spaced approximately 1 μm apart. The green confocal laser addresses and readout the NV centers from above. Remarkably, the fluorescence signal from individual NV centers remains visible even beneath the BSCCO stack (Fig. 5.1), enabling spatially resolved mapping of electromagnetic fields within the superconductor.

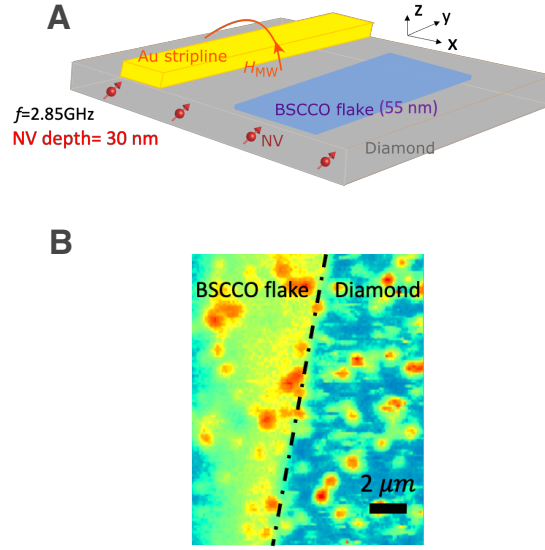


Figure 5.1: Experimental Configuration. **A** Schematic of the sample geometry. A gold (Au) stripline was fabricated on the diamond surface prior to the transfer of the BSCCO flake. **B** Confocal fluorescence image of a region on the diamond containing the edge of the BSCCO flake. Red spots correspond to individual NV centers. The black dashed line indicates the boundary of the BSCCO sample.

Below the superconducting transition temperature, the BSCCO flake exhibits the Meissner effect, expelling magnetic fields by generating screening currents that create an opposing magnetic field. This results in a suppression of the internal static magnetic field, which can be detected using electron spin resonance (ESR) of NV centers, where shifts in the NV resonance reflect changes in the local static field profile¹⁰¹.

In this work, we extended the capability of NV centers to probe finite-frequency electromagnetic responses by developing an AC measurement scheme. Under a resonant microwave drive, the NV spin undergoes coherent transitions between spin states—known as Rabi oscillations⁴³. Rather than detecting static magnetic fields, we use the Rabi frequency as a direct measure of the local oscillating magnetic field amplitude. The Rabi frequency is linearly related to the microwave field strength via $f_{\text{Rabi}} = \gamma B_{\text{AC}}$, where γ is the NV spin's gyromagnetic ratio. This method enables us to characterize the superfluid's electromagnetic response at finite frequencies.

5.1.2 RESULTS AND MODELING

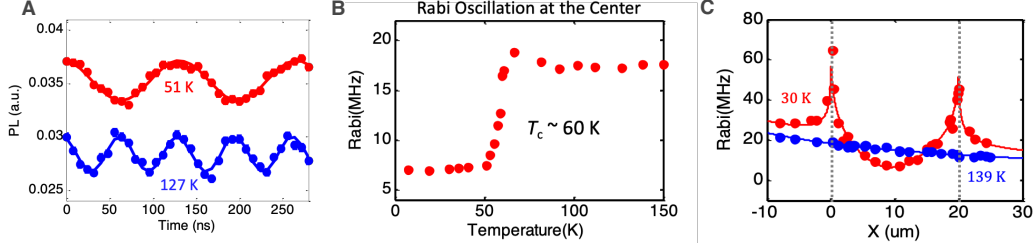


Figure 5.2: Rabi oscillations measured on and off BSCCO **A** Rabi oscillations measured at an NV center located beneath the center of the BSCCO flake at 51 K (below T_C) and 127 K (above T_C). **B** Rabi frequency measured at the same NV center as a function of temperature. The Rabi rate increases sharply as the temperature rises above the superconducting transition, indicating $T_C \approx 60$ K for this sample. **C** Spatial mapping of the Rabi frequency across the sample, measured above and below T_C . The gray dashed line indicates the boundary of the BSCCO flake. The x-axis represents the vertical distance between each NV center and the long edge of the sample. Although this is not a strictly one-dimensional scan, the flake's large aspect ratio allows us to assume quasi-translational symmetry along its long edge. Each dot represents the Rabi frequency measured at a single NV center, and the solid line shows our theoretical model fitted to the data.

By measuring the Rabi oscillations of an NV center located at the center of the BSCCO flake, both below and above the superconducting transition temperature T_C , we observe a factor-of-two difference in the Rabi frequency (Fig. 5.2). This observation supports the picture that, in the superconducting state, screening currents are induced in response to the applied microwave magnetic field B_{AC} (at $f = 2$ GHz), which reduces the field amplitude reaching the NV center and consequently lowers the Rabi rate.

Because the BSCCO flake is much thinner than the bulk magnetic penetration depth, the field screening is better described by the Pearl penetration depth⁷⁰, given by $\lambda_P = 2\lambda_L^2/t$, where λ_L is the London penetration depth and t is the film thickness. Since λ_P can exceed both the flake thickness and its lateral dimensions, the screening is incomplete, allowing a finite microwave magnetic field to reach the NV center even at the center of the flake. This residual field accounts for the slowed down but still measurable Rabi oscillation observed in the superconducting state.

By measuring the Rabi oscillation rate at the center of the BSCCO flake, we can extract the su-

perconducting phase transition temperature. As the temperature approaches T_C from below, the screening effect rapidly diminishes, and the Rabi rate recovers to the same level observed at NV centers far from the BSCCO sample (Fig. 5.2B). The transition temperature T_C , determined by the midpoint of the sharp increase in the Rabi frequency, is estimated to be approximately 60 K for this sample.

Spatially, the screening currents suppress the microwave magnetic field amplitude in the center of the flake, while enhancing it near the edges due to field concentration generated by the induced meissner currents. This spatial distribution is captured by measuring Rabi frequencies at multiple NV centers positioned at varying distances from the flake edge. As shown in Fig. 5.2C, the Rabi rate varies with distance, clearly reflecting the screening profile. A natural next step is to ask whether we can extract a characteristic decay length from this spatial profile in order to determine the Pearl penetration depth.

We developed a model based on an infinitely long 1D stripe with finite width. This is a good approximation in this case, as all measurements were taken near the center region of the BSCCO flake, where edge effects from the two ends of the ribbon are negligible due to the sample length being much greater than the Pearl penetration depth. Additionally, we adopted the DC limit in our simulation, as the microwave excitation frequency of 2 GHz corresponds to an energy of approximately 8 μeV , which is orders of magnitude smaller than the superconducting gap of BSCCO^{105,69}.

Under these assumptions, we constructed an equation that relates the surface current density distribution $K_y(x)$ in a 1D superconducting ribbon to the Pearl penetration depth λ_p . This is achieved by expressing the z-component of the magnetic field using both the Biot–Savart law and London’s second equation:

$$\frac{B_z(x)}{\mu_0} = H_e(x) + \frac{1}{2\pi} \int_{-a}^a \frac{K_y(x')}{x' - x} dx' = -\lambda_p \frac{dK_y(x)}{dx} \quad (5.1)$$

Here, all quantities are functions of the x -coordinate only. μ_0 is the vacuum permeability, $H_e(x)$ is the externally applied magnetic field whose spatial profile is determined by the geometry and position of the stripline relative to the sample, and a is half the width of the superconducting ribbon. λ_p is the Pearl penetration depth.

Given the drive field profile and a value of λ_p , this equation allows us to solve for the induced surface current distribution $K_y(x)$. Once $K_y(x)$ is obtained, the induced magnetic field and the total magnetic field can be calculated using the Biot–Savart law.

The solid line in Figure 5.2C represents the fit of this model to the experimental data. The only fitting parameter is the Pearl penetration depth λ_p . The drive field profile $H_e(x)$ is extracted from the Rabi frequency measured on the same species of NV centers at temperatures above T_C (blue dots in Figure 5.2C).

5.1.3 PENETRATION DEPTH

Through the fitting between the model and the data, we extract the Pearl penetration depth from spatially-resolved measurements at each temperature. The extracted values are $\lambda_p = 3 \mu\text{m}$ at 6 K and $\lambda_p = 5 \mu\text{m}$ at 40 K. Both values are significantly smaller than the length of the sample, validating the infinite 1D channel approximation across the full temperature range. To facilitate comparison with bulk measurements, we convert the Pearl penetration depth to the London penetration depth using $\lambda_p = \lambda_L^2/t$, where t is the thickness of the superconducting film (Fig. 5.2A). The resulting London penetration depth follows a T^2 dependence, consistent with the dirty-limit behavior of a d-wave superconductor, described by $\lambda(T) - \lambda(0) \propto T^2$ ⁸². The extrapolated zero-temperature

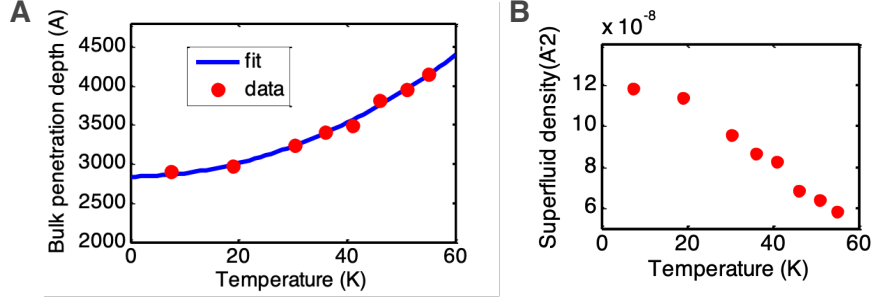


Figure 5.3: Extracted penetration depth and superfluid density **A** London penetration depth, converted from the fitted Pearl penetration depth, plotted as a function of temperature. The blue solid curve shows a fit to the expected T^2 dependence, consistent with the dirty-limit behavior of a d-wave superconductor. **B** Extracted superfluid density versus temperature, derived from the London penetration depth.

value of $\lambda(0) \approx 2800$ Å also agrees well with previous reports, such as $\lambda(0) = 2600$ Å from T.

Jacobs *et al.*⁴¹.

The superfluid density can also be obtained from the penetration depth using the relation $\rho_s(T) \equiv \lambda^{-2}(T)$. The extracted temperature dependence of ρ_s shows good agreement with previously reported results⁸⁸, when normalized by the superconducting transition temperature of this sample.

The preliminary characterization demonstrates the powerful capability of NV centers for locally probing superconductivity in systems with limited sample volume. Further studies of vortex states and phase transitions in BSCCO could be particularly interesting. Recent work has reported evidence of vortex liquid and solid phases in BSCCO⁵⁷.

5.2 WIDE-FIELD IMAGING OF MAGNETIZATION

I explored ways to expand the measurement techniques listed in the rest of this thesis beyond just scanning confocal, and into potentially higher throughput widefield modalities. The project was motivated by Fourier imaging², a method of applying large magnetic field gradients to achieve sub-diffraction spatial resolution. We hope to achieve sub-diffraction resolution in the wide-field to simultaneously image fine magnetic and current structures.

To integrate a wide-field imaging path into the existing confocal microscope, we introduced a flipper mirror before the objective to switch between the confocal and wide-field configurations. A 100 mW, 532 nm laser (Cobolt Samba 100 532nm) can be switched on/off using a single-pass acousto-optic modulator (AOM, ISOMET 1250C-848), driven by an RF signal from the AOM driver (ISOMET 535C-2-B). The laser beam then passes through a Thorlabs BE02-05-A adjustable telescope to adjust the illuminated area. A focusing lens (focal length: 400mm) directs the beam onto the back aperture of the objective, resulting in a collimated illumination of the diamond surface.

A dichroic mirror is used to separate the reflected green laser from the red photoluminescence (PL) emitted by the NV centers. The PL is then collected, focused, and imaged onto a Basler camera for wide-field readout.

The result is a 10 μ m gaussian-diameter luminescent spot on the diamond surface that can be readout simultaneously.

We demonstrated the characterization capability of wide-field imaging (Fig. 5.4). However, since the scientific goals of this work required maximizing sensitivity rather than imaging area, we opted to use confocal scanning for the hydrodynamic project.

First, the measurement sensitivity is fundamentally power-limited. Sensitivity scales as $\sqrt{N}$, where N is the number of detected photons per second⁵. In wide-field imaging, the excitation beam is expanded to a $\sim 10 \mu$ m diameter, increasing the illuminated area by 100~400 times compared to a confocal spot. Saturating NV centers over such a large area requires tens of milliwatts of laser power—exceeding the objective’s power rating and inducing considerable sample heating. Operating in the non-saturated regime requires proportionally longer averaging times to reach similar sensitivity. Additionally, the camera pixels (Basler) have lower detection efficiency compared to avalanche photodiodes (APDs) used in confocal setups, further reducing overall sensitivity.

Second, sensitivity is non-uniform across the field of view due to the Gaussian profile of the ex-

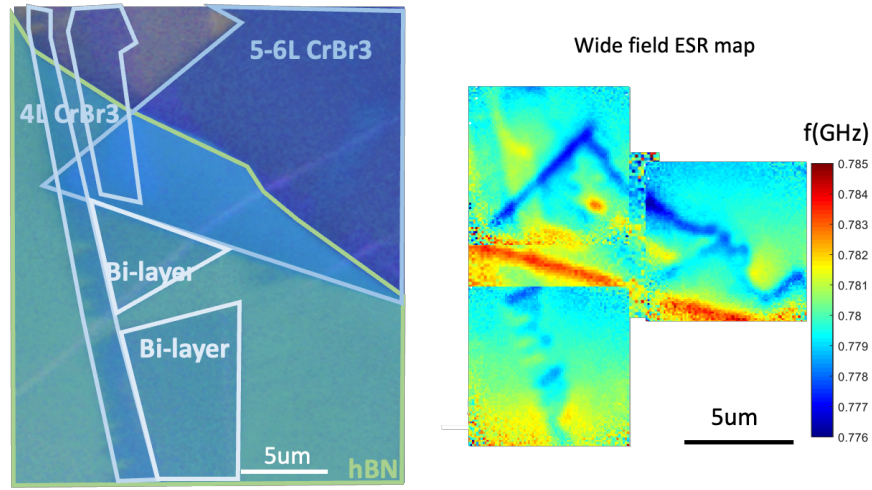


Figure 5.4: Wide-field imaging of stray fields from CrBr_3 flakes. In the left panel, exfoliated CrBr_3 flakes naturally contain regions of varying thickness, ranging from bilayer to 5–6 layers. CrBr_3 exhibits both interlayer and intralayer ferromagnetism, resulting in increasing magnetization with thickness. The right panel shows the magnetic field map acquired using wide-field NV imaging. Each image was averaged over 4–5 hours to achieve the presented signal-to-noise ratio. Noisy regions near the corners of each frame arise from reduced laser density and corresponding lower magnetic sensitivity.

citation beam, leading to weaker illumination—and hence reduced sensitivity—at the edges (see Fig. 5.4). In contrast, confocal scanning ensures uniform sensitivity by adjusting laser power point-by-point.

Third, implementing pulsed protocols with wide-field imaging is challenging due to the difficulty of achieving spatially homogeneous microwave driving over $\sim 10 \mu\text{m}$, which poses a significant microwave engineering problem (required rabi rate to be uniform within 5% in the field of view). Additionally, the camera shutter rate (typically 1–100 kHz) is much slower than the duty cycle of pulsed sequences, which greatly extends the total measurement time when synchronization with the camera is required.

Lastly, although wide-field images appear smoother with higher pixel density, the spatial resolution remains diffraction-limited, just like confocal imaging.

Nonetheless, future work based on wide-field imaging is a promising direction, and the results

shown here represent initial steps highlighting its potential value. All the comparisons suggest that future development of wide-field techniques would benefit from engineering higher laser power and achieving a more uniform spatial power distribution. In addition, developing spatially uniform microwave delivery is essential to fully utilize wide-field imaging for advanced sensing protocols.

5.3 FABRICATION OF DIAMOND NANOBEAM STRUCTURES

Diamond nanobeams containing NV centers have been used in non-scanning experiments to sense materials that are difficult to integrate with bulk diamond substrates, such as YIG²⁶. These nanobeams are typically less than 1 μm wide and a few micrometers long, making them suitable for pick-up by a sharp metal tip via static electromagnetic forces and transfer using standard 2D material stacking stages. Alternatively, the nanobeams can be picked up by a 2D material stack (e.g., hBN) and embedded into the device. This approach enables local NV magnetometry but lacks scanning capability.

The goal of this project is to fabricate diamond nanobeams with a built-in orientation marker, ensuring that the implanted surface NV centers are controlledly land facing the sample surface during transfer. To achieve this, we designed and developed fabrication recipes specifically tailored for creating such orientation-specific nanobeams.

The nanobeams are designed with a triangular cross-section for the convenience of the etching process. A bent structure is incorporated at the end of each beam to indicate its orientation during the transfer process. The nanobeam is suspended on a base composed of larger triangular supports, connected by narrow “weak links” that are intentionally designed to break easily during transfer onto the target material.

In addition, since triangular cross-section nanobeams can serve as effective waveguides, notches are introduced along the beam at 3 μm intervals. These notches help isolate single NV centers

within individual optical cavities, allowing the photons emitted by NV fluorescence to couple out through the notches.

The general flow of the nanobeam fabrication recipe includes the following steps for defining the etch mask and releasing the structure(the full recipe seen in Appendix 7.1.3):

1. E-beam lithography to define the shape of the etch masks. Both HSQ and metal hard masks were attempted, each requiring slightly different processing.
2. Top-down etching using O_2 plasma to open deep vertical trenches.
3. Angled etching using a focused Ar ion beam (FIB) to undercut and release the nanobeams from the substrate.

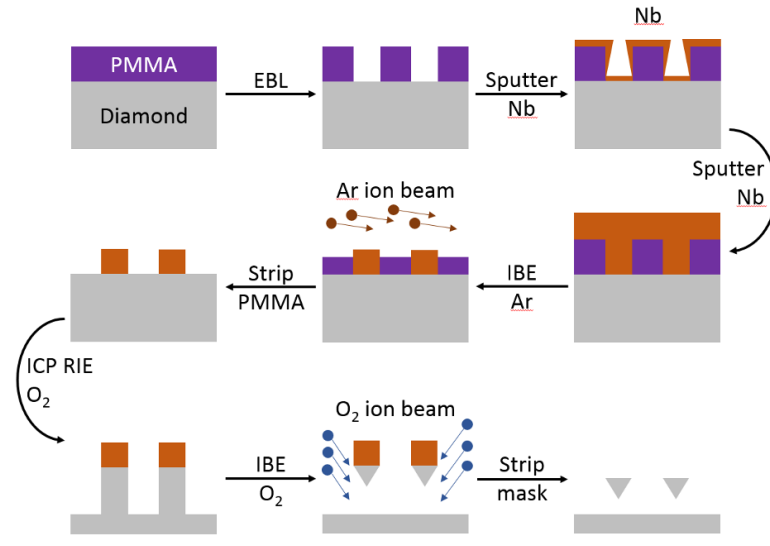


Figure 5.5: Fabrication process of diamond nanobeams using Nb metal mask. Schematic of the nanobeam fabrication process, including electron beam lithography, metal mask deposition, O_2 plasma deep etching, and a final angled etch using a focused ion beam (FIB).

For the etching masks, we developed two approaches. The first uses hydrogen silsesquioxane (HSQ) as an electron beam lithography (EBL) resist, which maintains sufficient thickness during

O₂ plasma etching, although it has low selectivity during the angled ion beam (with Ar) etch. To improve selectivity for both etching steps, we further developed a metal mask technique. In this method, a metal layer (thermally evaporated Ti or sputtered Nb) is deposited onto a developed PMMA (positive EBL resist) layer. This metal mask provides better etch resistance for both the top-down and angled etching processes.

Scanning electron microscope(SEM) image and measurement was taken at the steps after deposited mask, O₂ etch and after ion beam etch to evaluate the wear-level of mask, lateral dimensions and check whether it has been etched through to release nanobeams(Fig. 5.6).

We also tested the transfer process of nanobeams onto a quartz substrate. By identifying the orientation of the bent end, we could determine that the top surface of the nanobeam was correctly positioned on the substrate.

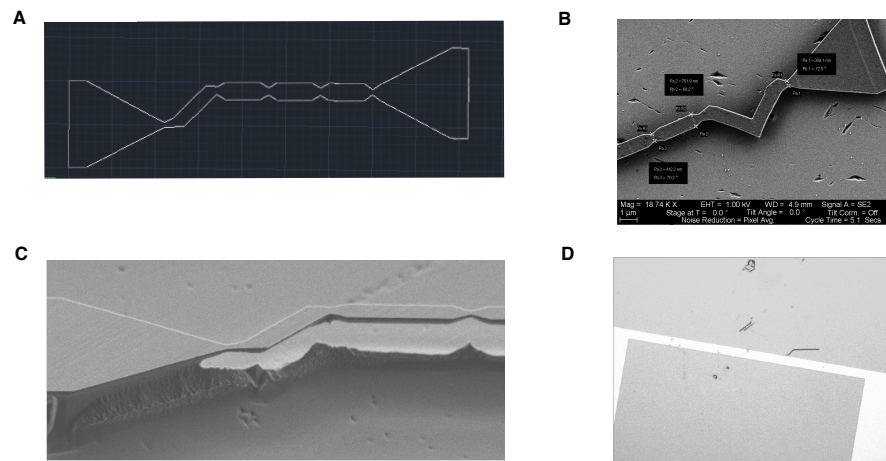


Figure 5.6: Images of diamond nanobeams. A CAD layout of the etch mask for a single nanobeam. B–C Top-down and side views of diamond nanobeams after angled etching (B shows nanobeam with metal mask still on; C shows nanobeam after tri-acid cleaning, showing a smooth diamond surface). D Optical microscope image of a nanobeam transferred onto the target substrate.

In collaboration with Ziwei Qiu, we used a transfer stage to pick up nanobeams with hBN stacks and utilized the NV centers in the diamond nanobeams for electric field sensing.

Compared to the previously used diamond nanobeams in²⁶, our design offers improved control

over the NV-to-sample surface distance. Owing to their small contact area, nanobeams enable better contact between the NV center and the sample surface than other diamond nanostructures, such as chiplets. However, because the surface NV centers are located far from the waveguide center, they exhibit lower brightness than naturally occurring NV centers in nanobeams, due to reduced coupling efficiency. A promising approach to address this issue is to combine implanted ensemble diamonds with this geometry, especially for sensing bulk material systems that are otherwise difficult to contact with diamond (except through scanning probes).

6

Conclusion

The main result of the thesis is the discovery of magnon hydrodynamic transport and magnon second sound in a Heisenberg ferromagnet, revealed through both DC and noise magnetometry using ensemble NV centers in a diamond substrate. Our investigation began with the observation of an anomalous temperature scaling of thermal magnetic noise, which we attribute to viscous damping of magnon second sound. This interpretation further inspire the stimulated noise magnetometry, where by directly driving the mode we observed a resonant spectroscopic feature consistent with

magnon sound propagation.

To our knowledge, this work presents the first direct experimental observation of magnon hydrodynamic transport, supported by a well-developed theoretical model that quantitatively captures both the spectroscopic feature, temperature and magnetic field dependence of the data. Hydrodynamic transport has previously been observed in electronic systems^{50,93}, plasmons in graphene¹¹³, and phonons^{51,104}, with theoretical proposals also made for excitons⁵⁴ and other quasiparticles. Such transport properties open the door to novel device functionalities, such as designing unidirectional particle flow using Tesla valve geometries^{31,39}.

In later part of thesis, we also demonstrate how NV center magnetometry can be applied to study superconductivity in mesoscopic BSCCO flakes. By leveraging both temperature- and spatial-dependent Rabi measurements, we extract the Pearl penetration depth and derive superfluid density, showing good agreement with known scaling laws and previous bulk measurements. Our model, based on a 1D ribbon geometry, provides a reasonable approximation given the sample's aspect ratio and the experimental configuration.

We explored ways to expand these measurement capabilities to widefield imaging to parallelize imaging measurements. While this technique shows promise, especially for visualizing large areas with varying magnetization, we find its sensitivity, uniformity, and compatibility with pulse protocols to be limiting compared to confocal scanning. These comparisons highlight key engineering challenges—particularly in laser power handling and microwave delivery—that must be addressed to fully realize the potential of wide-field NV magnetometry.

The development of diamond nanobeam fabrication expands the range of configurations for interfacing NV centers with materials and devices fabricated on non-diamond substrates. Beyond spin hydrodynamic measurements in 2D magnetic materials, these nanobeams can potentially be applied to other promising systems that grow in bulk crystal form.

Overall, this thesis discussed several experimental developments and scientific discoveries that

underscore both the power and current limitations of NV-based quantum sensing for condensed matter systems. These results lay the groundwork for both technique and device improvement in future studies.

7

Appendix

7.1 SAMPLE FABRICATION AND RECIPES

This section describes three parts of the fabrication process: (1) patterning Ti/Au striplines on a $2\text{ mm} \times 2\text{ mm}$ diamond substrate, (2) preparation of CrCl_3 stacks, and (3) fabrication of orientation-specified diamond nanobeams.

7.1.1 PATTERN WAVEGUIDE ON DIAMOND SURFACE

To efficiently deliver microwave signals to NV centers on the surface of a diamond substrate, we fabricated Ti/Au microwave striplines directly on the diamond. Figure 7.1 shows the two stripline designs used in our experiments. The coplanar waveguide design in **A** is more commonly used, as it provides a relatively uniform microwave field when the sample is positioned at the center of the gap. The design shown in **B** was specifically used for BSCCO experiments, where we accounted for spatial variations in Rabi frequency caused by the distance-dependent microwave field strength.

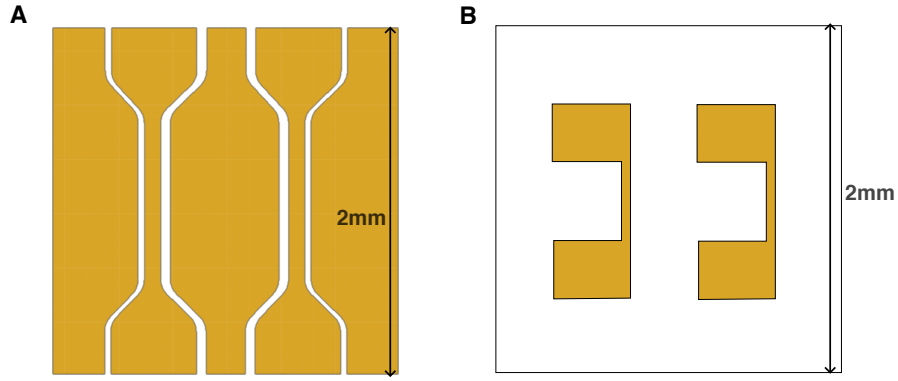


Figure 7.1: Two microwave stripline designs used in the experiments. **A** Two pairs of coplanar waveguides patterned on a $2\text{ mm} \times 2\text{ mm}$ diamond substrate. The trenches on both sides of each stripline are asymmetrically designed, with one side $30\text{ }\mu\text{m}$ wide and the other $50\text{ }\mu\text{m}$. Yellow regions indicate Ti/Au evaporation, while white regions are windows opened to the diamond substrate. The wider ends serve as bond pads. **B** A second design with a single pair of striplines on the same diamond substrate.

The fabrication processes for the two stripline designs differ. For designs like Fig. 7.1**B**, standard electron-beam or photolithography using a positive resist is typically employed, followed by metal deposition and lift-off. However, for the design shown in Fig. 7.1**A**, which has large areas covered by Ti/Au, using electron-beam lithography to define all the deposition regions is time-consuming and prone to significant charging effects on the diamond substrate. To address this, we developed a fabrication method based on an etching process, which combines pre-deposition, photolithography with an etching step to define the metal pattern. The detailed recipe is provided below.

Step 1: Tri-acid cleaning. Boil the diamond substrate in a 1:1:1 mixture of nitric, perchloric, and sulfuric acids for at least 20 minutes. Rinse thoroughly with distilled water afterward.

Step 2: Mounting. Fix the 2 mm × 2 mm diamond substrate onto a 4/6-inch carrier wafer using Kapton tape. Place the diamond slightly off-center to minimize edge bead formation on all sides.

Step 3: Thermal evaporation of Ti/Au.

- **Titanium (Ti):** 10 nm thickness, 2 pellets, power ~38%, current ~180 mA, deposition rate ~0.2 nm/s.
- **Gold (Au):** 300 nm thickness, 4 pellets, power ~22%, current ~120 mA, deposition rate ~0.2 nm/s.

Ensure that the chamber pressure reaches 2×10^{-7} Torr before starting the deposition to guarantee good adhesion between metal and diamond.

Step 4: Photolithography.

- Spin-coat SI813 at 5000 rpm.
- Soft bake at 110 °C for 2 minutes.
- Expose using a SUSS MA6 Mask Aligner at 405 nm (h-line), with an exposure dose of 80 mJ/cm².
- Detach the sample from the carrier wafer.
- Develop in a 1:3 mixture of MF-351:H₂O for 1 minute 15 seconds (stir the solution well before use).

Step 5: Metal etching.

- Au etch: Use 28A/S etchant for 107 seconds.
- Rinse by dipping in four sequential DI water baths and blow dry.

- Ti etch: Etch for 10 seconds to ensure clean removal.
- Rinse in four water baths.

Step 6: Resist removal. Sonicate the diamond in acetone to remove photoresist, then rinse with isopropanol (IPA) and blow dry.

Then the diamond substrate can be wiredbonded to a PCB board ready for flake transfer.

7.1.2 HBN ENCAPSULATED CrCl_3 STACKS

Bulk crystals of CrCl_3 were grown by chemical vapor transport, following the method described in Ref. ⁴⁸. CrCl_3 flakes were exfoliated onto 95 nm SiO_2 on Si substrates. The sample thicknesses were initially identified by their red color contrast on the SiO_2/Si chip. Flakes were then double-encapsulated with thin hBN layers (~ 10 nm) using standard polycarbonate/PDMS transfer techniques inside a glovebox with O_2 and H_2O levels maintained below 0.1 ppm.

The stacking process had to be completed within 2–4 hours after exfoliation, particularly for monolayer flakes, due to their high sensitivity to degradation even in glovebox conditions. During stacking, the temperature used to heat the chip was carefully kept below 80°C to avoid damaging the flakes. After double-side encapsulated, the full heterostructure was then transferred onto the diamond substrate between the ground and center conductors of the coplanar waveguide, all within the glovebox environment. After transfer, the sample was taken out of the glovebox and sequentially cleaned by immersion in chloroform, acetone, and isopropanol to remove any residual polycarbonate from the surface.

The thickness of the monolayer samples were confirmed by optical color contrast of the sample on 95 nm SiO_2/Si . Monolayers of this material exhibit a 3% color contrast in the red channel on 95 nm SiO_2 on Si. Multi-layer samples thickness were measured with Atomic Force Microscope (AFM).

7.1.3 DIAMOND NANOBEAMS

The fabrication of the diamond nanobeam involves six major steps: tri-acid cleaning of the diamond substrate, electron-beam lithography to write the etch mask, metal deposition to form the etch mask, oxygen plasma etching for top-down etching, angled ion-beam etching to create the triangular shape and release the nanobeam from the diamond substrate, and finally, tri-acid cleaning to remove the metal masks. The process was developed based on the fabrication process of diamond nano-phonic structure that was run by Bart Machiels.

1. **Clean diamonds** (Tri-acid cleaning, HF, piranha, and solvent)
2. **Spin PMMA**
 - (a) Choose appropriate PMMA for desired thickness, which should be larger than final mask thickness as ion beam planarization step will etch down some thickness of mask
 - (b) Example: If target Nb thickness is 300 nm → use 950 PMMA C4, which spins 400 nm at 3000 rpm. Check MicroChem PMMA spin curves
3. **Put conductive layer** (Au or spacer or aquasave)
 - (a) If using Au: EE-4, 5 nm
4. **EBL** (EL-4/EL-5)
 - (a) If using 950 PMMA C4: can write with 1 nA, dose is around 1000-1400 $\mu\text{C}/\text{cm}^2$
(other settings: write field 500 μm , 200,000 dots)
5. **Develop**
 - (a) If using Au: dip in Au etchant for 3 s, rinse with DI water, then IPA

- (b) Developing PMMA: 60 s in MIBK/IPA 1:3, then rinse with IPA, then dry with N₂ gun

6. **Sputter Nb** (Yacoby group sputter tool)

- (a) Determining what thickness to deposit:
 - i. Determine critical feature: e.g. for waveguides with support pad, waveguide is the critical feature
 - ii. Determine width of critical feature (e.g. 800 nm)
 - iii. Thickness to deposit = half of width of critical feature, plus 20% (e.g. for 800 nm width, deposit 400 nm + 20% = 500 nm of Nb)
- (b) Use gun 3 (rate is 0.127 nm/s)
- (c) Rotate stage such that it is normal to the target (i.e., 90 or 270 degrees, depending on which side you measure from)

7. **Ion beam planarization** (IBE-01)

- (a) To mill off Nb on top of PMMA. Since this is a milling process with Ar ion beam, and etch time is long, sample heats up easily. This will overbake the PMMA and make it harder to remove afterwards. To mitigate this, we run the etch at lower substrate temperature and break up the etching into cycles of etching and cooling (instead of running a single etch)
- (b) Recipe: Ar 300 BV, 20stage angle, chiller temperature at 5
- (c) Number of duty cycles depends on thickness of Nb deposited (etch rate is 4 – 4.1 nm/min)
- (d) Set cycle run time to 10 minutes, cycle cool time to 5 minutes

- (e) Run sufficient etch such that all Nb on top of PMMA is removed, and PMMA surface is exposed for subsequent stripping
- (f) How to check if sample is fully planarized: The sample starts off shiny if there is Nb on top. Once Nb is removed, you will be able to see the underlying PMMA. To be sure, do an SEM image, as you can see the underlying PMMA even if there is a thin layer of Nb on top.

8. Strip PMMA – 2 steps

- (a) Remover PG, 80, 24 h
- (b) Matrix plasma stripper (RIE-12), 90, 5-10 minutes

9. Top-down etch (RIE-11)

10. Angled etch (IBE-01)

Alternative to Nb mask, one can use thermally evaporated Ti mask as etching masks. The edge smooth level is comparably worse than Nb mask, but process could be simplified. For the application of our nanobeam, the roughness from Ti mask is also acceptable. The step 6 and 7 will then be replaced by the following:

1. Thermal evaporate Ti (Yacoby group thermal evaporator)

- (a) load 3 Ti pellets.
- (b) Thermal evaporation starts when the vacuum reached $5\text{e-}7$ torr and evaporate 300nm Ti. Slower the deposition rate may produce a smoother Ti mask.

2. Lift off

- (a) Lift off with soaking the diamond chip in Aceton overnight

7.2 NV DEPTH AND DENSITY AND PROPERTIES OF SENSING SPOT

Having knowledge of the implanted NV depth and density is crucial for our understanding of the magnon hydrodynamic measurements. This section describes how we characterize these properties of the diamond in experiment.

7.2.1 SENSING SPOT GAUSSIAN PROFILE

The sensing spot has a spatial profile given by that of the focused green laser, which spatially convolutes all measurements. A photoluminescence scan over the edge of gold wire pattern can be used to determine this spatial profile (Fig. 7.2). The dark region in the PL map shown in Fig. 7.2A is the gold wire, and the bright region is the surface of the diamond with shallow NV centers. We expect the edge of the metal to be much sharper than the spatial profile of the beam, so that it can be realistically modeled as a step edge for this purpose. The PL map then represents a convolution of a step function with the laser beam's spatial profile. As a result, a derivative of the spatial profile of PL counts reveals the underlying spatial profile of the laser beam, which is very well-modeled as a gaussian with 1σ width of 158 ± 3 nm (Fig. 7.2B), corresponding to a beam waist of $4\sigma = 632 \pm 12$ nm. A perfectly focused gaussian beam for this wavelength of light would have a 532 nm waist.

7.2.2 NV DEPTH

The diamond used in this work was implanted with 25 keV nitrogen ions. SRIM simulation suggest that this implantation energy results in a mean stopping distance of 25 nm from the surface⁷². Given that SRIM simulations underestimate the actual NV depth by a factor of 2⁷³, we expect an average NV depth closer to 50 nm in the experiment.

To measure the NV depth, we measure the stray field from a current-carrying wire — in this case, the central line of the coplanar waveguide. For distances not too far from the central line we

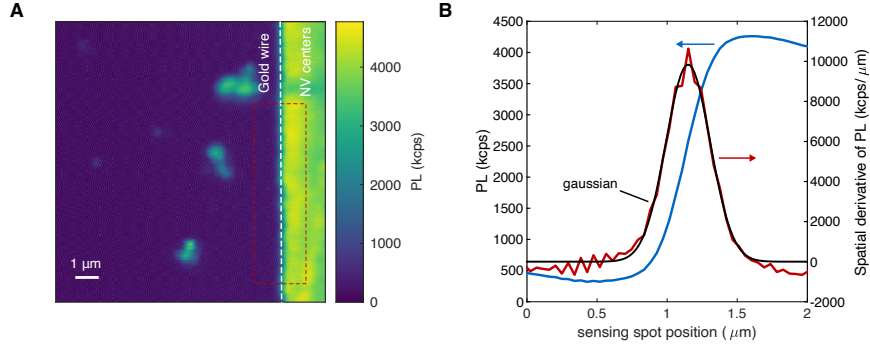


Figure 7.2: **A** Spatial photoluminescence (PL) raster scan over a gold wire. **B** Spatial average of the PL and spatial derivative of PL in the red box indicated in **A**. The spatial derivative of PL can be fit to a gaussian profile with a standard deviation of $\sigma = 158 \pm 3 \text{ nm}$. This corresponds to a beam waist of $4\sigma = 632 \pm 12 \text{ nm}$.

can neglect the stray field effects of the ground plane. A fixed field of 135 mT was applied parallel to the direction of the wire tilted at a 54° angle from the plane of the surface in order to select the quantization axis of NV centers closest to the direction of current (Fig. 7.3A). For a given position of the sensing spot relative to the wire, a positive/negative voltage bias is applied to the coplanar waveguide during the first/second leg of a Hahn-echo sequence (Fig. 7.3B), and then the phase (ϕ_{NV}) induced by the stray field from the current is recorded through a 4-axis projective measurement. The Hahn-echo evolution time was fixed at $4 \mu\text{s}$ per leg of the sequence ($8 \mu\text{s}$ total), so that the magnetic field generated by the drive voltage, projected along the NV axis, is given by $B_{\text{NV}} = \pi \phi_{\text{NV}} / (2 * 4 \mu\text{s} * 2.8 \text{ MHz/G})$, where ϕ_{NV} is in units of radians. We measure the induced NV phase over a range of bias voltages, and the resulting curve is fit to a line to extract the generated magnetic field per volt. Fig. 7.3C shows two such measurements for different positions of the sensing spot relative to the wire edge. This procedure is done for various positions of the sensing spot relative to the wire within a distance of $2 \mu\text{m}$ from the edge of the wire. Special care was taken to recalibrate the Rabi rate at every spatial point because, while varying the position of the sensing spot relative to the wire, the microwave drive field also strongly varies.

The stray field from a current-carrying wire can be numerically simulated, taking into account

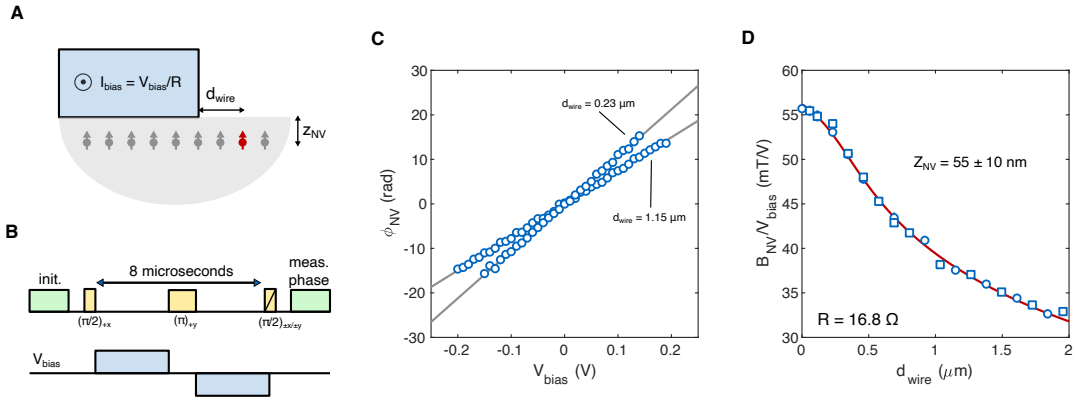


Figure 7.3: **A** Schematic of experiment for measuring the NV depth (z_{NV}) via the stray field from a conducting gold wire. The edge of the gold wire is determined via PL measurements, so that the distance between the sensing spot indicated by the red NV center, and the edge of the wire, is known (d_{wire}). A bias voltage, V_{bias} , drives a current through the wire. The NVs in this case face along the direction of the wire and 54° tilted from the surface normal. A field of 136mT is applied along this axis. **B** Differential measurement of the stray field induced by the voltage applied to the wire. A positive and negative voltage bias are applied in the first and second legs of the Hahn-echo sequence, respectively. The differential phase induced by this bias voltage is projectively read out. **C** Example of a scan of NV phase, ϕ_{NV} , versus bias voltage at two spatial positions relative to the wire. The data follow a linear trend, from which the generated magnetic field per volt can be determined. **D** Measurement of the magnetic field per volt as a function of distance from the wire edge. Squares and circles represent two different runs at different positions along the wire. The red line is a fit to the data based on a simulation from which we extract the wire resistance, R , and the NV depth, z_{NV} . Error bars on fit parameters are determined by a χ^2 test.

the known thickness of the coplanar waveguide (300 nm), a spatial convolution from the finite gaussian width of the laser beam (beam $1\sigma = 158\text{nm}$), and the position of the edge of the stripline as determined from PL measurements (e.g. Fig. 7.2B). Here, skin effects in the gold metal are neglected, and the NV centers are assumed to be all at a constant depth. As shown in Fig. 7.3D), the data as a function of spatial position can be fit to the model with two free parameters — the resistance of the coplanar waveguide (whose inverse gives the current per volt), and the NV depth. The stray magnetic field signal is found to be independent of NV depth for points more than about $0.5\text{ }\mu\text{m}$ from the stripline, so that the two fit parameters are completely uncorrelated. Repeating this procedure twice for two different linecuts across the microwave line yields a similar trend. By fitting the model to the results of these two trials, we determine the NV depth to be $55\text{ nm} \pm 10\text{ nm}$ in good agreement with the value expected based on the 25 keV implantation energy. It is important to point out that implantation straggle results in a distribution of NV center depths, where what we refer to as depth here should be understood as the mean of the depth distribution. With an additional bottom hBN thickness of 10nm, the NV-to- CrCl_3 distance in this experiment is, on average, $65\text{ nm} \pm 10\text{ nm}$.

7.2.3 NV DENSITY

The diamond was irradiated with a nitrogen atom fluence of $2 \times 10^{12}/\text{cm}^2$. At the implantation energy used here (25 keV), the conversion efficiency of nitrogen to nitrogen-vacancies after annealing is about 5-10%⁷². This results in a nitrogen-vacancy center density of $1 \times 10^{11}/\text{cm}^2$. Each sensing spot has roughly 100 NVs of all crystallographic species (using the FWHM of the gaussian beam). For a single field orientation, there are about 25 addressable NVs ($1/4$ of the total NVs) within the sensing spot.

7.3 NOISE MEASUREMENTS ON THE BACKGROUND

Measurements on a background region of diamond with only hBN coverage for two fields show that the intrinsic dephasing is only weakly temperature-dependent over the 0-20 K temperature range for both 136 mT along the NV axis, and 4 mT out of plane fields (Figs. 7.4 and 7.5). In practice, background measurements were taken on a nearby region of diamond for every sample region measured; the same qualitative behavior was always observed with only weak $\sim 5\%$ variations in the observed background rate. The temperature point density in the background measurements is lower than that taken on the sample, so that the background $1/T_2$ rate as a function of temperature was interpolated and then subtracted from the temperature-dependent rate measured on the sample.

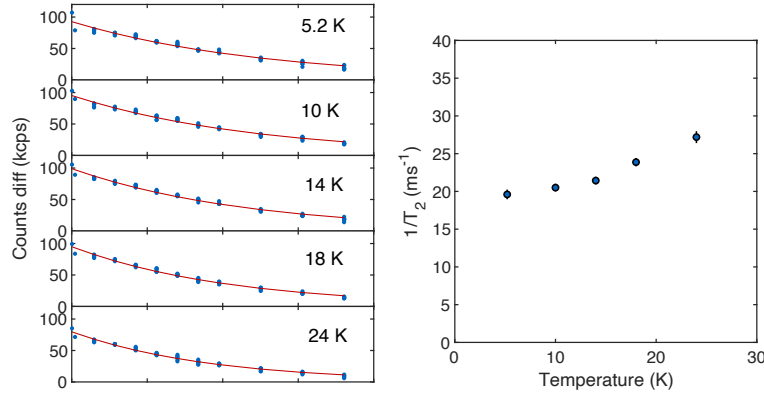


Figure 7.4: Measurements on bare diamond with only hBN coverage at 131 mT. Counts diff refers to the difference in counts taken over two Hahn-echo sequences where the last $\pi/2$ pulse is along $\pm x$, as explained in 2.2.2. The red line is a fit to a simple exponential dephasing, from which we extract T_2 which is then plotted in the right panel as a function of temperature.

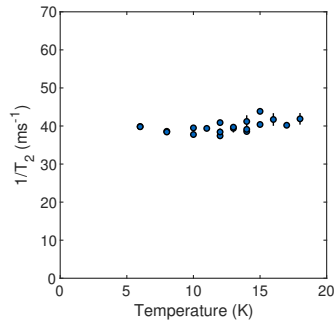


Figure 7.5: Measurements on bare diamond with only hBN coverage at 4 mT out of plane field. T_2 is obtained by measuring the contrast at the first ^{13}C revival and dividing it by the reference $1/T_2 = -\log(C(\text{first revival}))/\tau_{\text{first revival}}$.

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