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Giant spin rotation under quasiparticle-photoelectron conversion: Joint effect of sublattice interference and spin-orbit coupling

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Spin- and angular-resolved photoemission spectroscopy is a basic experimental tool for unveiling spin polarization of electron eigenstates in crystals. We prove, by using spin-orbit coupled graphene as a model, that photoconversion of a quasiparticle inside a crystal into a photoelectron can be accompanied with a dramatic change in its spin polarization, up to a total spin flip. This phenomenon is typical of quasiparticles residing away from the Brillouin-zone center and described by higher rank spinors and results in exotic patterns in the angular distribution of photoelectrons.

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Establishing spin polarization of quasiparticles in crystals is of crucial importance for semiconductor spintronics, physics of metallic surfaces, and the emerging field of topological insulators. The latter typically comprises narrow-gap systems with strong spin-orbit (SO) coupling. Graphene, a zero-gap conductor, attracts attention due to its linear Dirac-Weyl energy spectrum and prospects for applications. Its quasirelativistic spectrum, manifesting itself in unconventional quantum Hall effect\textsuperscript{1,2} and Klein tunneling,\textsuperscript{3} is essentially a nonrelativistic phenomenon originating from two equivalent sublattices A and B; their effect is conveniently accounted for by pseudospin. However, the relativistic effects that entangle their spin and pseudospin degrees of freedom\textsuperscript{4} lift the spin degeneracy of the energy spectrum. Intrinsic spin-orbit interaction in graphene\textsuperscript{5} is weak, not exceeding tens of $\mu$eV,\textsuperscript{6–9} but breaking the up-down symmetry by a substrate can result in a substantial extrinsic spin-orbit coupling. Such a coupling of the scale of 10 meV was discovered by Varykhalov et al.\textsuperscript{10} by spin- and angular-resolved photoemission spectroscopy (SARPES) techniques. Despite its modest magnitude, this coupling modifies essentially the zero-gap nonrelativistic spectrum. This makes spin-orbit coupled graphene an excellent platform for unveiling nontrivial effects of spin-orbit coupling on SARPES spectra.

In graphene, the zeros of the gap are in the achieved in two nonequivalent corners of the Brillouin zone (BZ) K and K’ [Fig. 1(c)]. In the vicinity of these points, the nonrelativistic quasiparticles (electrons and holes) possess a quasi-relativistic energy spectrum $E(\mathbf{k}) = \pm \gamma k$, $\mathbf{k}$ being a quasimomentum counted relative to the K(K’) point.\textsuperscript{11} Near the K point, the nonrelativistic Hamiltonian is $\mathcal{H}_0 = \gamma (\mathbf{s} \cdot \mathbf{k})$, where the pseudospin $\mathbf{s} = (s_1, s_2)$ is a vector of Pauli matrices acting on ($A, B$) sublattices [Fig. 1(a)]. The leading term in the extrinsic spin-orbit coupling is $\mathcal{H}_{\text{soc}} = \frac{2}{\hbar} \lambda (\mathbf{s} \times \mathbf{s})$, where $s$ are Pauli matrices of the real spin and $\lambda$ is the spin-orbit coupling constant;\textsuperscript{4} it couples spin to the pseudospin. We disregard the breaking of the ($A, B$) symmetry by the staggering potential of the substrate; for some substrates it is weak and graphene behaves as quasi-free standing.\textsuperscript{10,12} The two-sublattice structure of graphene is known to result in an interference effect in the nonrelativistic ARPES spectrum that produces strong photoemission anisotropy but does not distort the shape of the isoenergy surfaces.\textsuperscript{13} In relativistic spectra, the joint effect of interference and the spin-pseudospin entanglement produces giant rotations of electron spins during photoemission, resulting in drastic differences in the spin polarization of quasiparticles inside the crystal and photoelectrons in vacuum. This phenomenon is the focus of this Rapid Communication.

The Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{soc}}$ acts in the space of four-spinors defined in the basis $\{|A\uparrow\rangle, |A\downarrow\rangle, |B\uparrow\rangle, |B\downarrow\rangle\}$, the products of the twofold-degenerate Bloch functions of the K point at A(B) sublattices and spin-up(down) spinors. It is convenient to change to a basis $\{|A\uparrow\rangle, |B\downarrow\rangle, |B\uparrow\rangle, |A\downarrow\rangle\}$ and express each bispinor $\Psi_{\nu \mu}$ in terms of two spinors $\varphi_{\nu \mu}$ and $\phi_{\nu \mu}$, as $\Psi_{\nu \mu} = \{\varphi_{\nu \mu}, \phi_{\nu \mu}\}$ (for details, see Ref. 14). The eigenvalues are

$$E_{\nu \mu}(k) = \frac{\nu \mu}{2} (\sqrt{\lambda^2 + 4 \gamma^2 k^2} - \mu \lambda),$$

where $\nu, \mu = \pm 1$. The spectrum consists of two ungapped and two gapped hyperbolas shifted by $\lambda$ that are shown in Fig. 1(b) with their quantum numbers. The spectrum is similar to unbiased bilayer graphene\textsuperscript{11} but with different nature of eigenstates and narrower gap. In the new basis, matrices of the quasiparticle spin are $\mathbf{S} = \sigma_3 \mathbf{s}$, and their mean values in the $(\nu, \mu)$ eigenstates are

$$\langle \mathbf{S} \rangle_{\nu \mu}(k) = \frac{2 \mu \gamma \mathbf{k} \times \mathbf{z}}{\sqrt{\lambda^2 + 4 \gamma^2 k^2}},$$

$\mathbf{z}$ being a unit vector perpendicular to the substrate. We note in passing that $\langle \mathbf{S} \rangle$ is proportional to the group velocity $\partial E_{\nu \mu}/\partial \mathbf{k}$. The sign of the chirality of eigenspinors $(\varphi_{\nu \mu}, \phi_{\nu \mu})$ is determined by $\nu$, the sign of spin polarization by $\mu$, and the sign of the energy by the product $\nu \mu$. The polarization of all branches is depicted in Fig. 1(b). Such a spin-polarized energy with $|\lambda| \approx 13$ meV was deduced from SARPES data taken from graphene on a Au/Ni(111) substrate\textsuperscript{10} for $|E_{\nu \mu}(k)| \approx |\lambda|$; data for lesser energies are not available.

According to Eq. (2), quasiparticles are in-plane polarized perpendicular to the momentum $\mathbf{k}$, and the magnitudes of their spins depend on $k$ and can even vanish. This is well

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compatible with the fact that $S^2 = 3$ because spin-orbit coupling results in large nondiagonal components of $S$ in the $2 \times 2$ spin-pseudospin space; hence, restrictions on its diagonal components (“crystal” spin) are relaxed. The scale of the anticipated effect follows from the ratio of $\mathcal{H}_0$ and $\mathcal{H}_0'$, that is about $k/k_s$, where $k_s = |\lambda|/2\gamma$ is a spin-orbit momentum. The term $\mathcal{H}_0$ Zeeman energy of the pseudospin in an effective magnetic field $\mathbf{k}$, describes the dependence of the interference of Bloch waves by two equivalent sublattices, and this mechanism affects the spin sector through the term $\mathcal{H}_{\text{int}}$. This spin renormalization is strongest for $k/k_s \leq 1$ [Fig. 1(b)] and leads to vanishing spin at the symmetry points $K$ and $K'$ as a result of lattice interference. We emphasize that this is impossible for simple lattices, where $(S^2 - 3$ is maintained only by in-plane polarization or by out-of-plane spin rotation, as recently reported.

While the effect of sublattice interference on the angular dependence of ARPES, unveiled by Shirley et al., is well documented for monolayer and bilayer graphene and for graphite, the recent progress in studying quasiparticles by SARPES techniques, the novelty of results, and the large magnitude of spin-orbit coupling found in a number of systems, all call for proper understanding of interference-generated spin patterns in photoemission. They are specific for the single-step transformation of Bloch spinors of quasiparticles into photoelectron plane waves with large in-plane (Brillouin) momentum and manifest themselves in singular $k$ dependencies in the $k \rightarrow 0$ limit. The Fano effect in atomic photoionization and polarization-dependent interband transitions never show such singularities. They also are absent in the single-step normal photoemission showing remarkable polarization properties, depending on the incidence conditions.

For calculating photoemission, one needs to employ detailed wave functions that are products of the components of envelope spinors $\Psi_{\mu}(\mathbf{k})$ and the Bloch basis functions. At the $K$ point of the BZ [Fig. 1(c)], the $|A\rangle$ functions differ from the symmetrically equivalent to them $|B\rangle$ functions by the phase factor $(-\omega)$, with $\omega = \exp(i2\pi/3)$. The $\omega$ factor reflects the effect of sublattice interference on photoemission in terms of the envelope functions, while the specific form of $|A\rangle$ is of no importance as long as small spin-orbit corrections, depending on atomic form factors, factorize out and can be disregarded. For nonrelativistic electrons $|A\rangle$ the interference factor in the photoemission intensity reduces to $|k/k||E/|E|| - \omega$ near the $K$ point $k \ll K$; here $k_s = k_y - ik_x$. The first term in the brackets originates from $\mathcal{H}_0$, and the second from the interference of outgoing photoelectrons ($\exp[i(k - R_k - R_{\mu})] = \omega$, with $R_{\lambda A,B}$ for sublattice coordinates). The interference factor is related to the quasiparticle pseudospin, whose mean value equals $\langle \sigma_x - i\sigma_y \rangle = -\omega|E|/|E|$ for a quasiparticle with a moment $\mathbf{k}$ along $\mathbf{K}$. The resulting photoemission is highly anisotropic, as described by the large $k$ limit of Eq. (5) and displayed in Fig. 2(b).

For relativistic electrons $|A\rangle = 0$, the envelope spinor $\Phi_{\mu}(\mathbf{k})$, describing the flux of spin-polarized photoelectrons from the $(\nu, \mu)$ eigenstate, can be found by adding the components of $\Psi_{\mu}(\mathbf{k})$ corresponding to the same spin polarizations of the basis spinors supplied with the proper phase factors originating from $|A\rangle$ and $|B\rangle$ functions

$$
\Phi_{\mu}(\mathbf{k}) \propto \left( \begin{array}{c} \varphi_{\nu\mu}^{(1)}(\mathbf{k}) - \omega \varphi_{\nu\mu}^{(1)*}(\mathbf{k}) \\ \varphi_{\nu\mu}^{(2)}(\mathbf{k}) - \omega \varphi_{\nu\mu}^{(2)*}(\mathbf{k}) \end{array} \right),
$$

where superscripts indicate the upper and lower components of the spinors $\varphi$ and $\phi$. This spinor, defined with the accuracy to a factor depending on the atomic matrix element, intensity of the source, etc., describes the dependence of the sign flux on the azimuth $\theta$ [Fig. 1(c)].

The final form of $\Phi_{\mu}(\mathbf{k})$ is determined by the explicit form of the quasiparticle spinors

$$
\varphi_{\nu\mu}(\mathbf{k}) = \frac{\gamma k}{\sqrt{2|E_{\text{parity}}|/\lambda^2 + 4\gamma^2 k^2}} \left( \begin{array}{c} i\nu k^2 / k^2 \\ 1 \end{array} \right),
$$

where $\gamma$ is the crystal spin-orbit parameter.
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essential only within a narrow vicinity of the degeneracy point controlled by intrinsic SO coupling [about 10 μeV \(\sim 10^{-3}\lambda\) (Refs. 6–9)] and trigonal SO corrections [for \(k \leq \alpha k^*_x \approx \alpha k^*\), \(\alpha\) being a lattice constant, \(\alpha k^*_x \sim 10^{-3}\) (Ref. 35)]; both for the parameter values of Ref. 16.

It is seen from comparing Figs. 1(b) and 3(a)–3(d) that the difference between the spins of quasiparticles and photoelectrons \((\hat{S}_\nu^x(k) - \hat{s}_\nu^x(k, \theta))\) differs between three \(\pi/3\) sectors around the \(K\) point; for panels (a) and (b) of Fig. 3, it is minimal inside the internal sector that is bright. These data allowed reconstructing the general pattern of spin polarization in the \(k \approx k^*_x\) range.\(^7\) Unveiling the spin polarization in the \(k \approx k^*_x\) range is more challenging. Actually, it is an open question whether reconstructing \((\hat{S}_\nu^x(k) - \hat{s}_\nu^x(k, \theta))\), related to four-component spinors \(\Psi_\nu^x(k)\), from \(\hat{s}_\nu^x(k, \theta)\), related to two-component spinors \(\Phi_\nu^x(k)\), is a well-posed mathematical problem and how much spin information is lost during photoemission. In any case, reconstruction of the \(k \approx k^*_x\) region should essentially include the data from external sectors.

All calculations were performed for spin-orbit coupled graphene. However, the qualitative conclusions are general and are based on (i) a narrow-gap spectrum of quasiparticles with strong spin-orbit coupling described by four-component spinors, and the presence of the \(\omega\) phase factors that requires (ii) existence of (nearly) equivalent sublattices and (iii) residence of quasiparticles away from the BZ center. These requirements are fulfilled for the Bi\(_1\)-Sb\(_2\) (Refs. 25 and 28) and Pb\(_{1-x}\)Sn\(_x\)Te (Ref. 36) compounds. Therefore, we expect that their SARPES spectra of bulk electrons and the electrons from their Tamm-Shockley surface bands should manifest similar anomalies.

In conclusion, we demonstrated a dramatic effect of the spin-pseudospin entanglement on the SARPES spectra of spin-orbit coupled graphene. Similar effects are predicted for different narrow-gap spin-orbit coupled materials.

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