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Al. L. Efros,[1] E. I. Rashba,[2,*] and M. Rosen[1]

[1]*Naval Research Laboratory, Nanostructure Optics Section, Washington, D.C. 20375*

[2]*Department of Physics, MIT, Cambridge, Massachusetts 02139*

(Received 24 April 2001; published 29 October 2001)

A theory of spin injection from a ferromagnetic source into a semiconductor through a paramagnetic ion-doped nanocrystal is developed. Spin-polarized current from the source polarizes the ion; the polarized ion, in turn, controls the spin polarization of the current flowing through the nanocrystal. Depending on voltage, the ion can either enhance the injection coefficient by several times or suppress it. Large ion spins produce stronger enhancement of spin injection.

DOI: 10.1103/PhysRevLett.87.206601 PACS numbers: 72.25.Hg, 73.63.–b, 85.75.Mm

The concept of a spin field effect transistor (spin FET), initiated by Datta and Das [1] and based on the electric field controlled precession of electron spins driven by the spin-orbit interaction [2], created a wide and active interest in spin injection from spin selective contacts into semiconductor microstructures. A growing body of work has resulted in the new field of spintronics [3], based on the spin properties of electrons and holes rather than on their properties as charge carriers, as is the case in traditional semiconductor electronics. The recent discovery of long spin relaxation times [4,5] (greater than a hundred nanoseconds) has stimulated additional interest in this field. However, developing effective room temperature (T_R) spin sources remains a critical requirement for spin-polarized transport. The T_R spin injection coefficient from a ferromagnetic metal (FM) to a semiconductor (SC) is currently only about 1% [6,7]. Impressively large spin injection coefficients of the order of 50%–90% have been achieved from a semimagnetic SC, but only for temperatures $T \sim 4$ K and in an external magnetic field [8–10]. Mechanisms controlling spin injection and the spin valve effect in the ballistic and diffusion regimes have been discussed in a number of papers [11–14]. It has been argued that a tunnel contact at the FM-SC interface should increase spin injection, at least in the diffusive regime [13], and available experimental data seem to support this conclusion [15–17]. Several types of spin filtering contacts have been proposed [18]. Since a spin FET is an interference device and electron spin precession driven by spin-orbit coupling is momentum dependent [1], monochromaticity of the injected electron beam is another critical requirement for a spin emitter.

In this Letter, we propose using a semiconductor nanocrystal (or a small quantum dot) doped by a paramagnetic ion as a connector between the FM and the SC, and develop a theory of spin injection through such a device. This system (i) controls the spin injection in a way quite similar to that of a tunnel contact, (ii) is completely controlled by the voltage across (or current through) the dot, and does not require an external magnetic field, (iii) results in an enhancement of the spin injection coefficient with increasing current, and (iv) is a nearly monochromatic source of spin-polarized electrons. Having in mind the strong dependence of the ion polarization on the current, we also envisage using such nanocrystals as nonlinear elements (mixers) working at the ultimate limit of miniaturization of a single paramagnetic ion [19].

The underlying physical picture is as follows. A tunnel current via the nanocrystal goes through only one of the two electron-ion spin levels that are formed by the electron-ion exchange interaction. Spin-polarized electrons, injected from the FM into the nanocrystal, align the spin $\hat{\boldsymbol{S}}$ of the ion [20]. When the ion spin is aligned with the electron spin, the dot acts as a filter either enhancing or suppressing the spin-polarized current. The effect is most pronounced for large S.

High quality semiconductor nanocrystals of controlled diameter d (down to 20 Å) are fabricated by the colloidal chemistry technique [21,22]. Strong carrier confinement results (i) in level spacings and Coulomb blockade energies that can exceed T_R by an order of magnitude (as revealed in tunneling experiments with CdSe [23,24] and InAs [25] nanocrystals) and (ii) in the dramatic enhancement of the electron-ion exchange interaction, which causes a splitting of the electron (and hole) spin sublevels in doped dots [26]. The doping of nanocrystals is a challenging task [26,27], and only recently have the first high-quality ZnSe:Mn^{2+} nanocrystals with $d \approx 57$ Å been fabricated [28]. They show an average exchange splitting of 28 meV, which is comparable to T_R.

To make the basic physics clear, we choose a simple model of the strong Coulomb blockade and sequential tunneling in the master equation approximation [29] and assume that a single paramagnetic ion with spin S resides at the center of a spherical dot. Electrons can populate the $1s$ state of the dot; because of the large Coulomb blockade energy, the population number can take on only the values $n = 0$ and 1. For $n = 1$, the exchange interaction $\hat{H}_{\rm ex} = -\beta_{\rm ex}(\hat{\boldsymbol{s}}_e \cdot \hat{\boldsymbol{S}})$ between the electron and ion forms two sublevels ($\lambda = \pm 1$) with total spin $J = S + \lambda/2$, where $\hat{\boldsymbol{s}}_e$ and $\hat{\boldsymbol{S}}$ are the electron and ion spins. The sublevels are separated by an energy $E_{\rm ex} = |\beta_{\rm ex}|(S + 1/2)$. Two ions, Mn^{2+} and Gd^{3+}, with $L = 0$ electron configurations and spin $S = 5/2$ and $7/2$, respectively, can be

considered as candidate ions. For Mn^{2+} doped dots, the $J = 3$ level is the lowest one (in accordance with Hund's first rule). The strength of the exchange interaction, β_{ex}, is proportional to the square of the electron wave function at the paramagnetic ion, and, for a small dot, increases as d^{-3} [26]. For CdSe:Mn^{2+} dots with $d \approx 25$ Å, an estimate based on the data of Ref. [28] results in $E_{ex} \approx 250$ meV.

Therefore, we consider tunneling through states belonging only to a single component of the $J = S \pm 1/2$ doublet with total spin projections $\tau = -J, \ldots, J$. Electron populations n_τ of these states are related to the mean population $\bar{n}$ of the dot as $\bar{n} = \sum_\tau n_\tau$. In the absence of the electron, spin projections are $\sigma = -S, \ldots, S$ and population probabilities of these states are ν_σ. The normalization condition is

$$\bar{n} + \bar{\nu} = 1, \qquad \bar{n} = \sum_\tau n_\tau, \qquad \bar{\nu} = \sum_\sigma \nu_\sigma . \tag{1}$$

In our model, the exchange interaction between the electron residing in the dot and the FM contacts is neglected. In the absence of a current, all population numbers n_τ, as well as ν_σ, are equal, and the magnetization of the dot is zero both in the populated and the unpopulated states, $M_p \propto \sum_\tau \tau n_\tau = 0$, $M_{np} \propto \sum_\sigma \sigma \nu_\sigma = 0$. Therefore, the magnetization of the dot found in what follows arises entirely from the current I passing through it.

Spin-polarized currents I_α passing through the dot are controlled by the conductivities γ_α^μ of the left and right contacts, $\mu = L, R$. They depend on the electron spin projections $\alpha = \pm 1/2$. We consider the γ_α^μ as phenomenological parameters which characterize the spin selectivity of each contact [30]. Spin-polarized current to (or from) a specific quantum state of the dot can be found by multiplying γ_α^μ by the square of the Clebsch-Gordan (CG) coefficient $\langle \alpha\sigma | J\tau \rangle$ and by the Fermi functions $f_\mu(E) \equiv f(E - \zeta_\mu)$ [or $1 - f_\mu(E)$], where E is the energy of the electron level in the dot and the ζ_μ are the Fermi levels in the respective leads. The coefficients $P_\alpha = \sum_\mu \gamma_\alpha^\mu f_\mu(E)$ and $Q_\alpha = \sum_\mu \gamma_\alpha^\mu [1 - f_\mu(E)]$ characterize the total in- and out-going spin-polarized current.

Under steady state conditions, the balance equation,

$$P_\alpha \nu_\sigma = Q_\alpha n_\tau, \qquad \tau = \alpha + \sigma , \tag{2}$$

ensures equal rates of the formation of the populated τ state from the unpopulated σ state plus an α electron, and for the decay of the τ state through the same channel. CG coefficients do not enter into this set of equations. The solution of this system of equations is

$$n_\tau / n_0 = \kappa_{1/2}^\tau, \qquad \nu_\sigma / n_0 = (Q_\alpha / P_\alpha) \kappa_\alpha^{2\alpha(\sigma+\alpha)}. \tag{3}$$

This satisfies the equation $n_\tau n_{-\tau} = n_0^2$. Here,

$$\kappa_\alpha = P_\alpha Q_{-\alpha} / P_{-\alpha} Q_\alpha, \qquad \kappa_{-\alpha} = \kappa_\alpha^{-1}. \tag{4}$$

Equation (3) allows one to find the magnetization M_J of the populated state [31] in terms of the Brillouin function $B_J(x)$, $M_J = \sum_\tau \tau n_\tau / \sum_\tau n_\tau = B_J(\ln \kappa_{1/2})$, where

$$B_J(x) = (J + 1/2) \coth[(J + 1/2)x] - \coth(x/2)/2 .$$

For $Jx \ll 1$, $B_J(x) \approx J(J + 1)x/3$, and it saturates to $B_J(x) \approx J$ for $x \gtrsim 2$. One sees that M_J is completely determined by $\kappa_{1/2}$, has the sign of $(\kappa_{1/2} - 1)$, and does not depend on the CG coefficients. In the theory of paramagnetic ions, the argument of the Brillouin function is $x = \mu^* H / k_B T$, where μ^* is the magnetic moment of an ion and H is an external magnetic field. Therefore, $\ln \kappa_{1/2}$ plays the role of the magnetic energy measured in units of $k_B T$. The ion is not polarized if $\kappa_{1/2} = 1$. This is the case for $I = 0$, and also, for arbitrary I, if both leads have equal spin selectivities, $\gamma_{1/2}^L / \gamma_{-1/2}^L = \gamma_{1/2}^R / \gamma_{-1/2}^R$. M_J reaches its maximum for the case of opposite magnetization of the two leads.

Spin-polarized currents I_α are given by

$$I_\alpha = \frac{1}{2} \sum_{\sigma\tau} [(\gamma_\alpha^L f_L - \gamma_\alpha^R f_R)(\nu_\sigma + n_\tau) - (\gamma_\alpha^L - \gamma_\alpha^R) n_\tau] \langle \alpha\sigma | J\tau \rangle^2 . \tag{5}$$

Equation (5) is symmetric in L and R and is valid if spin relaxation in the dot is neglected. Using Eq. (2), the relation $P_\alpha + Q_\alpha = \gamma_\alpha^L + \gamma_\alpha^R$, and a similar relation for P_α, one can prove the identity,

$$(\gamma_\alpha^L f_L - \gamma_\alpha^R f_R)(\nu_\sigma + n_{\sigma+\alpha}) - (\gamma_\alpha^L - \gamma_\alpha^R) n_{\sigma+\alpha} = 2\gamma_\alpha^L \gamma_\alpha^R \nu_\sigma (f_L - f_R)/Q_\alpha . \tag{6}$$

Substituting Eqs. (3) and (6) into Eq. (5) results in

$$I_\alpha = n_0 \frac{\gamma_\alpha^L \gamma_\alpha^R}{P_\alpha} (f_L - f_R) \sum_{\sigma\tau} \kappa_\alpha^{2\alpha(\sigma+\alpha)} \langle \alpha\sigma \,|\, J\tau \rangle^2 . \tag{7}$$

Hence, the currents I_α increase with the electrochemical potential drop between the leads, and the spin polarization of the current depends on the total spin J of the state participating in the injection. Using explicit expressions for the CG coefficients, $\langle \alpha, \sigma \,|\, J\tau \rangle^2 = 1/2 + \lambda\alpha\tau/(S + 1/2)$, we find that the α polarized current passing through the λ component of the exchange split level is

$$I_\alpha^{(\lambda)} = n_0 (f_L - f_R) \frac{\gamma_\alpha^L \gamma_\alpha^R}{2P_\alpha} \sum_{m=-J}^{J} \kappa_\alpha^m \left(1 + \frac{\lambda m}{S + 1/2}\right), \tag{8}$$

where $m = 2\alpha\tau$ is a new summation index, $-J \le m \le J$.

It is a remarkable property of Eq. (8) that it includes exactly the same sums which enter in the definition of the magnetization M_J. Because of this fact, the currents $I_\alpha^{(\lambda)}$ can be expressed in terms of M_J:

$$I_\alpha^{(\lambda)} = \bar{n} (f_L - f_R) \frac{\gamma_\alpha^L \gamma_\alpha^R}{2P_\alpha} \left[1 + \frac{2\alpha\lambda M_J}{S + 1/2}\right]. \tag{9}$$

This shows that the effect of the paramagnetic ion on the spin-polarized currents $I_\alpha^{(\lambda)}$ through the dot is completely described in terms of the magnetization of the dot in the populated state, M_J.

Equation (9) permits one to find the dependence of the spin polarization ratio, $\Gamma^{(\lambda)} = (I_{1/2}^{(\lambda)} - I_{-1/2}^{(\lambda)})/(I_{1/2}^{(\lambda)} + I_{-1/2}^{(\lambda)})$, on M_J. Having in mind applications to spin injection from

FM to SC, we take the right lead to be spin nonpolarized, and introduce the notation $\gamma^L_{\pm 1/2} = \gamma^L(1 \pm \Delta)/2$, $\gamma^R_\alpha = \gamma^R/2$, where Δ describes the spin selectivity of the contact between the FM and the dot. Using the definition $P_\alpha = \gamma^L_\alpha f_L + \gamma^R_\alpha f_R$, one finds, after some algebra,

$$\Gamma^{(\lambda)}(M_J) = \Gamma_0 \frac{1 + \lambda\Gamma_0^{-1} M_J/(S + 1/2)}{1 + \lambda\Gamma_0 M_J/(S + 1/2)}, \qquad (10)$$

$$\Gamma_0 = \Delta/[1 + (1 - \Delta^2)\gamma^L f_L/\gamma^R f_R]. \qquad (11)$$

Equations (10) and (11) relate $\Gamma^{(\lambda)}$ to Δ. Parameter Γ_0 does not depend on the paramagnetic center. It describes the spin selectivity of the dot for small currents when $M_J \ll J$ (also for tunneling through the $J = 0$ level of a dot with $S = 1/2$).

Equations (10) and (11) are the principal result of the paper. Below, we discuss the insight they provide on the spin transport for the case that the voltages on the source, gate, and drain can be varied independently, i.e., f_L and f_R are independent variables controlling M_J, while Δ does not depend on the voltage.

We see from Eqs. (10) and (11) that, for small voltage, when $f_L \to f_R$ and $M_J/J \to 0$, the polarization ratio equals Γ_0 for both values of λ, and Γ_0 is always less than Δ. Therefore, an unpolarized ion suppresses spin injection. The ratio Γ_0/Δ depends on the ratio of the conductivities of the contacts, γ^L/γ^R, and, when the conductivity of the FM contact is much larger than that of the SC, $\gamma^L/\gamma^R \gg 1$, the suppression is strong. However, M_J grows quickly with increasing voltage (see Fig. 1a), while Γ_0 decreases slowly. When M_J becomes larger than $\Gamma_0(S + 1/2)$, the spin injection blockade breaks down and the polarization ratio acquires its normal magnitude $\Gamma^{(\lambda)} \sim \Delta$. The stronger the suppression of Γ_0, the narrower is the range of voltages in which the "spin injection blockade" occurs. In this region of the parameters, the dot behaves as a highly nonlinear spin emitter.

In Fig. 1 the dependence of the magnetization and $\Gamma^{(\lambda)}$ on voltage is shown for a Mn^{2+} doped dot. With increasing voltage, $\Gamma^{(+)}$ and $\Gamma^{(-)}$ change in opposite ways. If $\kappa_{1/2} > 1$, the magnetization M_J and $\Gamma^{(+)}$ increase with voltage, while $\Gamma^{(-)}$ decreases; $\Gamma^{(-)}$ can even change sign (see Fig. 1b). The region of enhanced spin injection, $\Gamma^{(+)} > \Delta$, lies to the right of the bold solid line described by

$$M_J/(S + 1/2) = \Delta/(1 + \gamma^R f_R/\gamma^L f_L). \qquad (12)$$

The parameter range $\gamma^L \approx \gamma^R$ and $f^L \gg f^R$ is optimal for spin injection enhancement. Under these conditions $\Gamma_0 \ll 1$, and Eq. (10) takes the form

$$\Gamma^{(\lambda)} \approx \lambda M_J/(S + 1/2). \qquad (13)$$

It is remarkable that Γ_0 drops out from this equation and, therefore, $\Gamma^{(\lambda)}$ depends on Δ only through M_J. For $\Delta \ll 1/2J$, expansion of $B_J(x)$ for small x yields $\Gamma^{(\lambda)} \approx 2\lambda J(J + 1)\Delta/3(S + 1/2)$. Thus, for small Δ, the enhancement factor is $\Gamma^{(+)}/\Delta \approx 2(J + 1)/3$, which is 8/3 for Mn^{2+}. For larger Δ, $\Delta \gtrsim 1/2J$, the magnetization saturates; $\Gamma^{(\lambda)} \approx 1$ and is independent of Δ. In both cases, *large J enhances the polarization ratio*. The dependence of $\Gamma^{(\lambda)}$ on Δ, for a Mn^{2+} doped dot, is shown in Fig. 2.

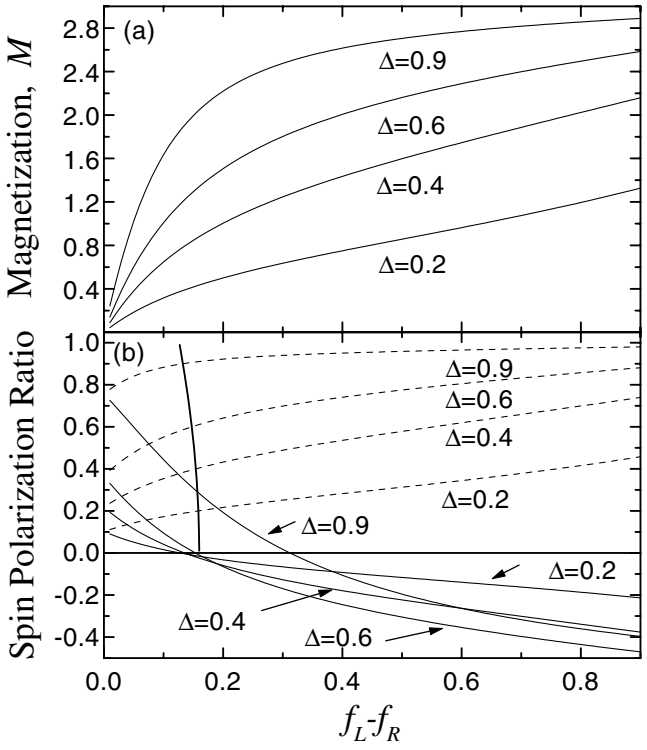

FIG. 1. Dependence of the magnetization M_3 (a) and the spin polarization ratio $\Gamma^{(\pm)}$ (b) on $f_L - f_R$, monitoring the voltage drop between the leads, for different values of Δ. Calculations were done for a Mn^{2+} doped nanocrystal. Dashed and solid lines in (b) describe $\Gamma^{(+)}$ and $\Gamma^{(-)}$ for $J = 3$ and $J = 2$ states, respectively. The solid bold line in (b), $\Gamma^{(+)} = \Delta$, separates the regions of enhanced (to the right) and suppressed (to the left) spin injection. Note the similarity in shape of the M_J and $\Gamma^{(+)}$ curves for $f_L - f_R \gtrsim 0.3$, in agreement with Eq. (13).

The spin polarization ratios for currents flowing through the two respective levels of the exchange doublet exactly coincide as $f_L - f_R \to 0$; they are equal in magnitude but opposite in sign as $f_L \to 1$, $f_R \to 0$.

Heretofore, we have neglected spin relaxation, which suppresses the spin alignment of the ion. There are two basic spin relaxation times, T_n and T_ν, for a populated and unpopulated dot, respectively. Equations that include relaxation are more complex than Eq. (2). However, in most cases the criteria, $\gamma_{\rm p} T_n \gg 1$, $\gamma_{\rm np} T_\nu \gg 1$, ensure that spin relaxation has only a minor effect on the magnetization of the dot. Here, $\gamma_{\rm p} \sim Q_\alpha^{-1}$ and $\gamma_{\rm np} \sim P_\alpha^{-1}$ are decay rates of the populated and unpopulated states, respectively. For time dependent voltages, the dynamics of M_J and $\Gamma^{(\lambda)}$ are controlled by the same relaxation times. Typically, $10^9\ {\rm s}^{-1} \lesssim \gamma^{L,R} \lesssim 10^{14}\ {\rm s}^{-1}$ [25]. The room temperature spin relaxation time T_ν of Mn^{2+} in bulk ZnSe is 100–1000 ns [32], and should be much longer in small dots. For undoped CdSe dots, the spin relaxation time, τ_s, measured at $T_{\rm R}$ was ≈0.4 ns [33].

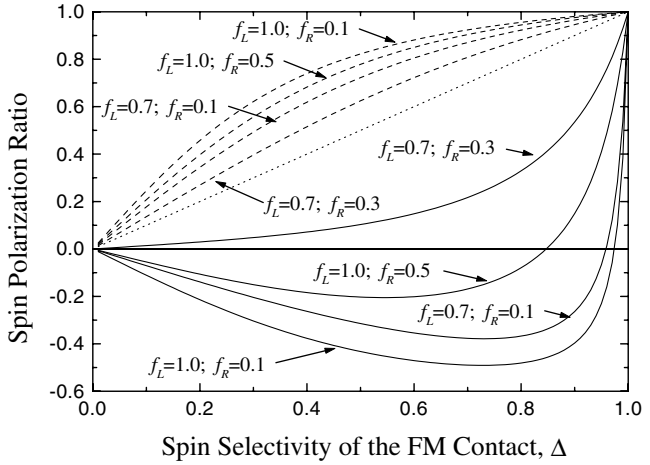

FIG. 2. Dependence of the spin polarization ratio $\Gamma^{(\pm)}$ on Δ for several values of f_L and f_R calculated for a Mn^{2+} doped nanocrystal and $\gamma_L = \gamma_R$. Dashed and solid lines describe $\Gamma^{(+)}$ and $\Gamma^{(-)}$ for $J = 3$ and $J = 2$ states, respectively. The dotted line, $\Gamma^{(+)} = \Delta$, separates the regions of enhanced (above the line) and suppressed (below the line) spin injection.

Extrapolation to T_R of the data of Ref. [34], taken for excitons in InAs/GaAs dots, results in $\tau_s \approx 1$ ns. Therefore, spin relaxation can usually be neglected.

In conclusion, we have shown that spin injection from a FM electrode into a nanocrystal aligns the spin of a paramagnetic ion and that this strongly influences the spin polarization of the current. For large ion spin S, the ion can enhance spin injection by a factor of about 3, and yields nearly 100% polarized current even when the FM source is not 100% polarized. We expect this effect to be observable at room temperature for small nanocrystals and quantum dots with a large exchange splitting.

Al. L. E. and M. R. thank the ONR for financial support and D. J. Norris for a stimulating discussion. E. I. R. acknowledges support from DARPA by the ONR Grant No. N000140010819 during the final stage of this work.

*Current affiliation: Department of Physics, SUNY at Buffalo, Buffalo, NY 14260.
Email address: erashba@mailaps.org

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