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Nondiffusive Spin Dynamics in a Two-Dimensional Electron Gas

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We describe measurements of spin dynamics in the two-dimensional electron gas in GaAs/GaAlAs quantum wells. Optical techniques, including transient spin-grating spectroscopy, are used to probe the relaxation rates of spin polarization waves in the wave vector range from zero to 6×10^4 cm^{-1}. We find that the spin polarization lifetime is maximal at a nonzero wave vector, in contrast with expectations based on ordinary spin diffusion, but in quantitative agreement with recent theories that treat diffusion in the presence of spin-orbit coupling.

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Electronic systems with strong spin-orbit interaction (SOI) exhibit exotic effects that arise from the coupling of spin polarization and charge current, such as spin Hall currents [1–7], current-induced spin polarization [8], and spin-galvanic effects [9,10]. These effects involve manipulation of the electron spin via electric, rather than magnetic, fields, creating the potential for applications in areas from spintronics to quantum computing [11]. However, SOI is a double-edged sword, as it also has the undesired effect of causing decay of spin polarization, reflecting the nonconservation of the total spin operator, $\vec{S}$, i.e., $[\vec{S}, \mathcal{H}] \neq 0$, where $\mathcal{H}$ is any Hamiltonian that contains spin-orbit coupling.

In the consideration of spin-orbit coupled systems, a great deal of attention has been focused on quantum wells or heterostructures fabricated in III-V semiconductors, where breaking of inversion symmetry allows coupling that is linear in momentum. For electrons propagating in [001] planes, the most general form of the linear coupling includes both Rashba [12] and Dresselhaus [13] terms:

$$\mathcal{H}_{\rm so} = \alpha(k_y\sigma_x - k_x\sigma_y) + \beta(k_x\sigma_x - k_y\sigma_y), \quad (1)$$

where $k_{x,y}$ is the electron wave vector along the [1 0], [0 1] directions in the plane, and α and β are the strengths of the Rashba and Dresselhaus couplings. The spin-orbit terms generate an effective in-plane magnetic field $\vec{b}_{\rm so}$ whose direction depends on $\vec{k}$. Spin nonconservation takes the form of precession, at a rate governed by $\vec{b}_{\rm so}(\vec{k})$, during an electron's free flight between collisions. In the Dyakonov-Perel (DP) regime [14], where the precession angle during the free-flight time τ is small, a spatially uniform spin polarization will relax exponentially to zero at the rate $1/\tau_s \propto |\vec{b}_{\rm so}|^2\tau$. Although this process relaxes the initial polarization state, spin memory is not entirely lost, even after the electron undergoes many collisions. The relationship between real-space trajectory and spin precession leads to a correlation between the electron's position and its spin. Such correlations are predicted to enhance the lifetime of certain spatially inhomogeneous spin polarization states beyond what would be expected for conventional spin diffusion [15].

Burkov *et al.* [15] considered $\mathcal{H}_{\rm so}$ with only Rashba coupling ($\beta = 0$), and showed that a helical spin density wave with wave vector $\sqrt{15}m\alpha/2$ decays more slowly than a uniform (or $q = 0$) spin polarization, by a factor 32/7. This contrasts with ordinary spin diffusion, where the decay rate of a polarization wave increases monotonically ($\propto q^2$) with increasing wave vector. More recently, Bernevig *et al.* [16] showed that the lifetime of the spin helix is enhanced further in the presence of both Rashba and Dresselhaus coupling, and diverges as $\alpha \rightarrow \beta$. The infinite lifetime, or "persistent spin helix" (PSH) state, is a manifestation of an exact $SU(2)$ symmetry of $\mathcal{H}_{\rm so}$ at the point $\alpha = \beta$. As a consequence of the $SU(2)$ symmetry, a spiral spin polarization in the z, x_+ plane (where $\hat{z}$ and $\hat{x}_+$ are the normal and [1 1] directions, respectively) with wave vector $4m\alpha\hat{x}_+$ is a conserved quantity. These predictions suggest the possibility of manipulating spin polarization through SOI, without necessarily compromising spin memory, by controlling α/β with externally applied electric fields.

Transient spin-grating (TSG) experiments [17,18] are particularly well suited to testing the theoretical predictions of Refs. [15,16], as they directly probe the decay rate of nonuniform spin distributions. In the TSG technique, a spatially periodic polarization of the out-of-plane spin S^z, is created by a pair of interfering ultrashort laser pulses. By varying the relative angle of the two pump beams, we are able to vary the magnitude of the grating wave vector $\vec{q}$ in the range 0.44–5.3×10^4 cm^{-1}, corresponding to wavelengths in the range from ≈1–15 μm. The subsequent time evolution of the spin polarization wave amplitude, $S^z_q(t)$, is measured by coherent detection of a time-delayed probe beam that diffracts from the photoinjected "spin grating."

In this Letter we report detailed TSG measurements on two samples, each consisting of 30 GaAs quantum wells of thickness 12 nm, separated by 48 nm barriers of

$Ga_{0.7}Al_{0.3}As$. One sample was doped with eight atomic layers of Si in the central 14 nm of each barrier to create a high-mobility ($\mu \simeq 150\,000\ \mathrm{cm^2/V\,s}$ at 5 K) two-dimensional electron gas (2DEG) with carrier density $7.8 \times 10^{11}\ \mathrm{cm^{-2}}$. The second sample is identical except that the mobility of the electrons was reduced to $\simeq 3500\ \mathrm{cm^2/V\,s}$ by placing 83% of the dopant atoms in the wells, rather than in the barriers.

The TSG measurements show that the lifetime of S_q^z is maximal at nonzero $|\vec{q}|$ in both samples, in contrast to expectations for simple diffusion but in agreement with the prediction of Burkov *et al.* [15]. The magnitude of the lifetime enhancement and its dependence on the direction of $\vec{q}$ point to the presence of both Rashba and Dresselhaus interactions in our samples. Quantitative agreement between the theory of Bernevig *et al.* [16] and our experimental results suggests the feasibility of realizing and detecting a PSH in samples engineered to achieve $\alpha \approx \beta$.

Panels (a) and (b) of Fig. 1 show the decay of $S_q^z(t)$ for the high-μ and low-μ samples, respectively, measured at the wave vectors where the decay rate is smallest. In both samples the decay of the grating is clearly not a single exponential at low T and crosses over to nearly single exponential as room temperature is approached. All the decay curves can be fitted to a double-exponential form, $a_1 \exp(-t/\tau_1) + a_2 \exp(-t/\tau_2)$, with equal weighting factors ($a_1 = a_2$) over almost the entire range of T. The only exception is the $T < 25$ K regime of the high-μ sample, where the momentum relaxation rate $1/\tau$ becomes comparable to the spin-precession frequency Ω. In this ($\Omega\tau \geq 1$) regime the initial decay is damped oscillatory rather than exponential [18]. The solid symbols in panels (c) and (d) are the time constants τ_1 and τ_2 extracted from the double-exponential fit, plotted as a function of T. For both samples the ratio between the fast and slow rates is at least a factor of 10 at low T and gradually diminishes as T approaches room temperature. The solid lines are the lifetimes of the uniform spin polarization $\tau_s(T)$, as obtained from decay of transient Faraday rotation induced by a single, circularly polarized pump beam. For all temperatures, the $q = 0$ polarization decays as a *single* exponential, whose lifetime lies *between* the two lifetimes observed at nonzero q.

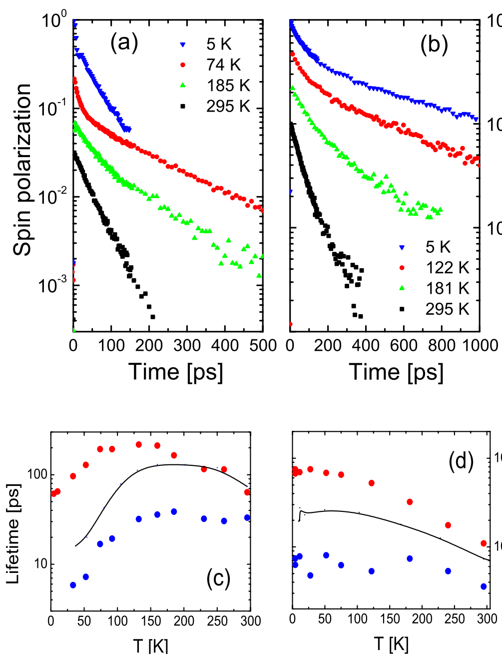

FIG. 1 (color online). Decay of the out-of-plane component of spin polarization wave with wave vector in the [1 1] direction, for (a) the high-mobility sample with $|\vec{q}| = 0.58 \times 10^4\ \mathrm{cm^{-1}}$ and (b) the low-mobility sample with $|\vec{q}| = 0.69 \times 10^4\ \mathrm{cm^{-1}}$, for several temperatures. The initial value of spin polarization is estimated at a few percent for all measurements. Decay curves are offset vertically for clarity. Solid symbols in panels (c) and (d) indicate the temperature dependence of the two time constants obtained from a double-exponential fit to the decay curves. Solid lines are the lifetime of a spatially uniform spin polarization.

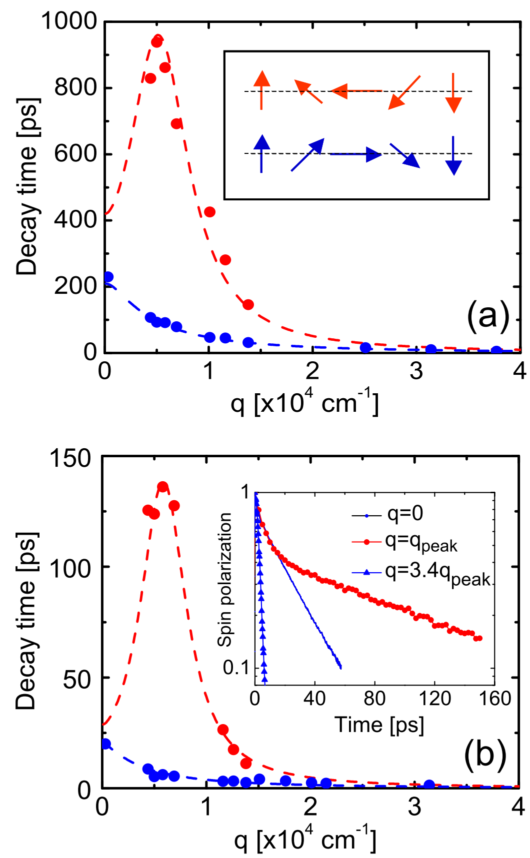

FIG. 2 (color online). Decay lifetimes obtained from best fit of a double-exponential to experimentally measured decay of spin polarization, as a function of wave vector $\vec{q}$ parallel to the [1 1] crystal axis. Data are shown for the (a) low-μ and (b) high-μ samples, at 50 K. The dashed lines are fits to a theory (described in the text) in which the two lifetimes correspond to the positive and negative spin helices illustrated in (a) inset. (b) inset: comparison of spin polarization decay at $q = 0$, $0.58 \times 10^4\ \mathrm{cm^{-1}}$, and $2.01 \times 10^4\ \mathrm{cm^{-1}}$.

The dispersion of the decay rates with $\vec{q} \parallel [11]$ is shown in Figs. 2(a) and 2(b), for the high and low μ samples, respectively. We present results for $T = 50$ K, a temperature at which both samples are in the $\Omega\tau < 1$ regime, and yet the ratio τ_2/τ_1 remains large (in a certain range of q). For both samples, the larger of the two time constants peaks sharply at $|\vec{q}| \approx 0.6 \times 10^4$ cm^{-1}. The lifetime of the more rapidly decaying component decreases monotonically with increasing $|\vec{q}|$, consistent with simple diffusion.

The spin-dynamical effects illustrated in Figs. 1 and 2, biexponential decay and nonmonotonic dispersion, are in quantitative agreement with the theory of coupled charge and spin dynamics in the presence of $\mathcal{H}_{so}$, which we describe briefly below. Assuming a single, isotropic τ, Burkov *et al.* [15] and Bernevig *et al.* [16] derived a set of four equations that describes the coupling of electron density $n_q(t)$ and the three components of $\vec{S}_q(t)$ brought about by the SOI. Along the [1 1] and $[1\,\bar{1}]$ directions, the four equations separate into two coupled pairs. For $\vec{q} \parallel [1\,1]$, spin precesses in the z-x_+ plane, leading to coupling of S^{x+} and S^z.

Solving the pair of equations that couple S^z and S^{x+} yields two eigenmodes and corresponding frequencies $i\omega_{1,2}(q) \equiv 1/\tau_{1,2}(q)$. The $\vec{q} \parallel [11]$ eigenfrequencies are

$$\frac{1}{\gamma_0\tau(q)} = \frac{q^2}{2q_0^2} + 3 + \sin 2\phi \pm \sqrt{(1 - \sin 2\phi)^2 + \frac{4q^2}{q_0^2}(1 + \sin 2\phi)}, \quad (2)$$

where $q_0^{-1} \equiv \hbar/m^* v_{so}$ and $\gamma_0 \equiv v_{so}^2 k_F^2 \tau$ are characteristic length and frequency scales for DP spin relaxation. The parameters $\phi \equiv \tan^{-1}(\alpha/\beta)$ and $v_{so} \equiv \sqrt{\alpha^2 + \beta^2}$ reflect the relative and combined coupling strengths of the Rashba and Dresselhaus interactions, respectively. The spin diffusion coefficient $D_s \equiv v_F^2\tau/2$ is given by $\gamma_0/2q_0^2$.

The solutions for $\vec{q} \parallel [1\,1]$ are especially interesting when $\alpha = \beta$. In this case the eigenvectors are forward and backward spin spirals in the z, x_+ plane. At the wave vector $q_0\sqrt{8}$, the corresponding eigenfrequencies are 0 and $16\gamma_0$. Zero decay rate for spin polarization at a "resonant" q is a manifestation of the exact $SU(2)$ symmetry identified by Bernevig *et al.* [16], enlarging the $U(1)$ symmetry identified previously [19,20]. The $U(1)$ symmetry arises when $\alpha = \beta$ because $\vec{b}_{so} \parallel \hat{x}_-$ for all k. The physical basis for the $SU(2)$ symmetry is that $b_{so} \propto \cos\theta$, where θ is the angle between $\vec{k}$ and $\hat{x}_-$. In this case, the net rotation of the electron's spin in the $\hat{z}$-$\hat{x}_+$ plane depends only its net displacement along $\hat{x}_+$, and equals 2π for $\Delta\hat{x}_+ = \pi/q_0\sqrt{2} \equiv \Lambda$. A helical spin wave, with wavelength Λ and sense of rotation matching that of the precessing electrons, will not decay when $\alpha = \beta$.

A TSG experiment creates a sinusoidal modulation of S^z, an equally weighted linear combination of the forward and backward spin helices. The subsequent time evolution of these two eigenmodes has the form of a double-exponential decay with equal weighting factors. After decay of the unstable eigenmode, the PSH remains stable, despite the rapid electron scattering and spin precession that are occurring. The eigenfrequencies for $\vec{q} \parallel [1\,\bar{1}]$ can be obtained from Eq. (2) as well, if we replace ϕ by $-\phi$. At the same $SU(2)$ point where the PSH exists for $\vec{q} \parallel [1\,1]$, the spin dynamics for $\vec{q} \parallel [1\,\bar{1}]$ obey simple diffusion.

Several features of the physics at the $SU(2)$ point remain when $\alpha \neq \beta$, but both are nonzero. The lifetime at the resonant wave vector is enhanced relative to the case when only one of two interactions is present, although it no longer diverges. The enhancement is expected to be stronger for $\vec{q} \parallel [1\,1]$ than $\vec{q} \parallel [1\,\bar{1}]$. The eigenvectors are still admixtures of S^z and S^{x+} and the photoinjected wave of pure S^z still decays as a double exponential. However, the weighting factors are q dependent and the long-lived state will be an elliptical, rather than circular, spin helix.

With this overview of the theory, we return to the experimental data. The dotted lines in Fig. 2 show the best fits obtained by varying parameters in Eq. (2). The fits yield parameter values $v_{so} = 460$ m/s, $\phi = 0.2$, and $D_s = 1000$ cm^2/s for the higher-μ sample and $v_{so} = 480$ m/s, $\phi = 0.08$, and $D_s = 105$ cm^2/s for the intentionally disordered sample. The above value of v_{so} for the high-μ sample predicts a spin-precession frequency for ballistic electrons, $\Omega = v_{so}k_F = 0.10$ THz, that is within 10% of the experimental value [21]. The theory applies equally well to both samples, despite a difference of about ten in their scattering rates. Note that smaller τ leads to smaller spin relaxation rates, as the entire dispersion scales with the DP relaxation rate, $4\gamma_0$.

A surprising feature of the theory [16] is the sensitivity of the dispersion curves to small admixtures of Rashba coupling into a pure Dresselhaus system, or vice versa. Our quantum wells are designed to be perfectly symmetric and therefore no Rashba term is expected. However, the maximum lifetime enhancement, ~7, for the higher μ sample is significantly larger than the factor 32/7 predicted for $\alpha = 0$. This discrepancy is accounted for by a relatively small admixture $\alpha \simeq 0.2\beta$. It is possible that the presence of a small Rashba contribution can be traced to differences in the interface on either side of the well [22,23], or to the tendency for δ dopants to diffuse preferentially towards the growing crystal surface [24], both known features of the GaAs/GaAlAs system.

A further prediction of the theory is that even small α/β generates a large anisotropy in the dispersion curves [25]. To test this prediction we measured S_q^z at 50 K in the higher-μ sample, for $\vec{q}$ oriented along the [1 1], $[1\,\bar{1}]$, and [1 0] directions. The results shown in Fig. 3 demonstrate that the lifetime enhancement is strong for $\vec{q} \parallel [1\,1]$ and weak for $\vec{q} \parallel [1\,\bar{1}]$. The line through the [1 1] curve is calculated using Eq. (2) for the lifetimes and Eq. 22 of Ref. [16] for the weighting factors of the double-exponential decay. The parameters are the same as used

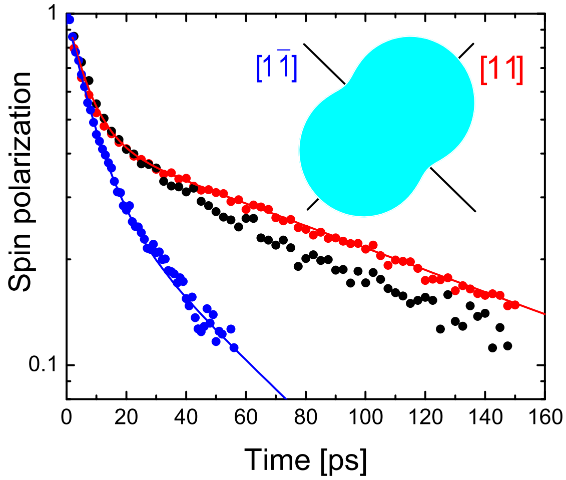

FIG. 3 (color online). Decay of spin polarization in the high-μ sample as a function of time for three orientations of the grating wave vector (top-to-bottom: [1 1], [1 0], and [1 $\bar{1}$]). The lines through the data are predictions of the theory described in the text, using the parameters that were obtained from fitting the dispersion curves. The anisotropy is smaller in the low-μ sample, which is consistent with the conclusion based on the dispersion data. Inset: Fermi contour for a system of free electrons subject to the SOI described by Eq. (1), showing the origin of the extremal orientations. Rashba coupling set to one-half of Dresselhaus (larger than in the systems under study here) for the purpose of illustration.

to fit the dispersion along [1 1]. The weighting factors and decay rates for $\vec{q} \parallel [1\,\bar{1}]$ are calculated using the *same parameters*, provided we replace ϕ by $-\phi$. The theory is clearly quite successful in providing a quantitative description of the spin relaxation dynamics seen in our experiment.

The only feature of the data not captured by the theory described above is the gradual decrease in the τ_2/τ_1 ratio with increasing T (shown in Fig. 1) that takes place for both samples. The characteristic scale of T clearly is not the spin-orbit splitting, which is $\sim v_F q_0$ or about 1 K for our samples. We speculate that the T dependence of τ_2/τ_1 is a consequence of the cubic (in k) Dresselhaus coupling, $\mathcal{H}_{cD} \propto k_x k_y^2 \sigma_x - k_y k_x^2 \sigma_y$, which does not appear in Eq. (1). $\mathcal{H}_{cD}$ is non-negligible when $\langle k_z^2 \rangle$, the expectation value of k_z^2 in the lowest subband, becomes comparable to k_F^2. The cubic Dresselhaus coupling destroys the proportionality $\Omega \propto k$, which ensures that the precession angle in a free flight between collisions depends only on the electron's displacement, and not on its velocity. In the presence of $\mathcal{H}_{cD}$ the precession angle will depend on k, and the fractional spread in precession angle for a given Δk is $\sim k_F \Delta k / \langle k_z^2 \rangle$. Assuming a thermally induced momentum distribution $\Delta k \sim T/v_F$, the relative variation in precession angle is $\sim T/E_1$, the ratio of the temperature to the energy of the first excited subband. Our results are consistent with E_1 (which is $\approx$500 K for our samples) as the characteristic energy scale above which the correlations that generate the PSH are lost.

In summary, we observe that spin polarization waves in GaAs QW's survive substantially longer than diffusive dynamics predict. Theory and experiment suggest that $SU(2)$ spin symmetry can be recovered, despite the presence of SOI, if the condition $\alpha = \beta$ can be met. Balancing the Rashba and Dresselhaus terms may enable modulation of the spin correlation length, with large dynamic range, through the application of external electric fields.

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