

LETTERS

Observation of spin Coulomb drag in a two-dimensional electron gas

C. P. Weber¹, N. Gedik^{1,2}, J. E. Moore¹, J. Orenstein¹, J. Stephens³ & D. D. Awschalom³

An electron propagating through a solid carries spin angular momentum in addition to its mass and charge. Of late there has been considerable interest in developing electronic devices based on the transport of spin that offer potential advantages in dissipation, size and speed over charge-based devices¹. However, these advantages bring with them additional complexity. Because each electron carries a single, fixed value ($-e$) of charge, the electrical current carried by a gas of electrons is simply proportional to its total momentum. A fundamental consequence is that the charge current is not affected by interactions that conserve total momentum, notably collisions among the electrons themselves². In contrast, the electron's spin along a given spatial direction can take on two values, $\pm\hbar/2$ (conventionally \uparrow, \downarrow), so that the spin current and momentum need not be proportional. Although the transport of spin polarization is not protected by momentum conservation, it has been widely assumed that, like the charge current, spin current is unaffected by electron-electron ($e-e$) interactions. Here we demonstrate experimentally not only that this assumption is invalid, but also that over a broad range of temperature and electron density, the flow of spin polarization in a two-dimensional gas of electrons is controlled by the rate of $e-e$ collisions.

In this work, spin diffusion is characterized by the transient spin grating technique³, which is based on the optical injection of spin-polarized electrons. The two-dimensional electron gas (2DEG) resides in a GaAs quantum well, in which the carriers are donated by Si impurities doped into the GaAlAs barrier layers. Near-bandgap illumination of the GaAs excites electrons whose initial spin is determined by the helicity of the light⁴. If the GaAs is excited by two non-collinear, coherent beams of light with orthogonal linear polarization, then in the region where the beams interfere the helicity varies sinusoidally from plus one to minus one. The optical-helicity wave generates a wave of electron-spin polarization with the same spatial frequency, which in turn generates a sinusoidal variation (grating) in the index of refraction through the Kerr effect. The wavevector of the injected spin-density wave is in the plane of the 2DEG, and the spin polarization is oriented perpendicular to this plane.

The time evolution of the transient spin grating directly reveals the nature of spin transport and relaxation in the electronic system, functioning like a time-domain version of neutron scattering. We measure the spin polarization by detecting the diffraction of a probe beam off the grating. A sensitive coherent detection scheme (Methods) enabled acquisition of the ~ 150 grating decays required to characterize the spin dynamics for each sample throughout the temperature-wavevector ($T-q$) parameter space.

In this Letter we present results for three quantum well samples, with electron concentrations of 7.8×10^{11} , 4.3×10^{11} and

$1.9 \times 10^{11} \text{ cm}^{-2}$, corresponding to Fermi temperatures of 400, 220 and 100 K, respectively. Figure 1 shows the initial decay rate of the spin grating as function of T in the most heavily doped sample, for several grating wavevectors from 0.4×10^4 to $2.5 \times 10^4 \text{ cm}^{-1}$. The dependence on T can be described in terms of three regions. For $100 \text{ K} < T < 300 \text{ K}$, the decay rate varies slowly. For $50 \text{ K} < T < 100 \text{ K}$, the decay rate increases rapidly with decreasing T , and for $T < 50 \text{ K}$, it reaches a slowly varying plateau.

We begin by discussing the decay rate where it varies slowly, that is, below 50 K and above 100 K. In the high- T region, the spin dynamics

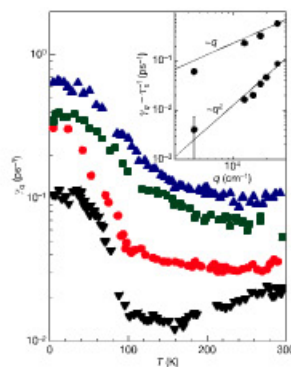


Figure 1 Spin-grating decay at various wavevectors (q) and temperatures (T) for the sample with Fermi temperature 400 K. Main panel, the initial decay rate, γ_0 of the spin grating as a function of T for (bottom to top) $q = 0.45 \times 10^4, 1.3 \times 10^4, 1.8 \times 10^4$ and $2.5 \times 10^4 \text{ cm}^{-1}$. Inset, the initial decay rate of the spin grating as a function of q . Points are $\gamma_0 - \tau^{-1}$; τ is obtained from decay of homogeneous ($q = 0$) spin excitation. Error bars (s.d.) are the size of the points except as shown. Lower points and line, room temperature. The line is a fit of the data to $\gamma_0 = \tau^{-1} + D_s q^2$, giving a spin diffusion length $l_s = (D_s \tau)^{1/2} = 0.81 \mu\text{m}$ and a 'spin mean-free-path' $l = 2D_s / v_s = 60 \text{ nm}$. The observation of diffusive motion is internally consistent, as l is much smaller than both l_s and the smallest grating wavelength, $2.5 \mu\text{m}$. Upper points and line, 5 K. The line has slope = 1, corresponding to ballistic, rather than diffusive, spin-motion with a velocity of $2.3 \times 10^7 \text{ cm s}^{-1}$.

¹Physics Department, University of California, Berkeley, and Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA. ²Laboratory for Molecular Sciences, Arthur Amos Noyes Laboratory of Chemical Physics, California Institute of Technology, Pasadena, California 91125, USA. ³Center for Spintronics and Quantum Computation, University of California, Santa Barbara, California 93106, USA.

can be accurately described in terms of independent processes of spin diffusion and spin relaxation. In this description, the decay rate varies with q quadratically, as $\gamma_0 = \tau^{-1} + D_s q^2$, where D_s is the spin diffusion coefficient and τ is the spin relaxation time⁵. In the inset to Fig. 1 we plot $\gamma_0 - \tau^{-1}$ versus q at 295 K (lower points) and 5 K (upper points), on logarithmic axes. Here $1/\tau$ is independently determined from the decay rate of the circular dichroism induced by a circularly polarized pump beam⁶ (see Supplementary Information). A comparison of the 295 K data with a line of slope two shows that the decay of the grating is well described by diffusive dynamics, with $D_s = 130 \text{ cm}^2 \text{ s}^{-1}$ and $\tau_s = 50 \text{ ps}$.

Next, we examine the spin-grating dynamics at $T < 50 \text{ K}$. As shown in Fig. 1 inset, the initial decay rates at 5 K are linear in q at the higher wavevectors. The change in power law exponent from two to one indicates that a crossover from diffusive to ballistic dynamics takes place as the sample temperature is lowered. In the ballistic regime, electrons propagate a distance comparable to the grating wavelength, λ , without scattering and the initial decay rate is $v_s q$, the reciprocal of the time required for an electron moving with the Fermi velocity (v_s) to traverse a distance $\lambda/2\pi$.

Although the grating's initial decay rate saturates near $v_s q$ when T reaches $\sim 50 \text{ K}$, its time dependence continues to change as T is lowered further. Figure 2 shows the grating amplitude as function of time for several temperatures between 5 K and 100 K, measured with a grating wavevector of $2.5 \times 10^4 \text{ cm}^{-1}$ (the T indicated is the lattice temperature, which is below the electron temperature, as will be discussed later). An oscillatory structure appears in the decay curves, becoming increasingly pronounced as T decreases. The growth of these oscillations is a consequence of the increase of the mean-free-path, l , in the regime where $q l \geq 1$.

To determine D_s from data such as those in Fig. 2, we use an expression for the time dependence of a spin fluctuation that is applicable throughout the diffusive-ballistic crossover regime. If a spin polarization wave is introduced at $t = 0$, its subsequent time dependence is the Fourier transform of $S(q, \omega) \propto [i\omega - D_s q^2 + \gamma_0]^{-1}$, where $D_s(q, \omega)$ is the dynamic spin diffusivity. In the limit $q \ll k_F$,

$$D_s(q, \omega) = \frac{v_s/2}{\sqrt{(i\omega/v_s - 1/l)^2 + q^2}} \quad (1)$$

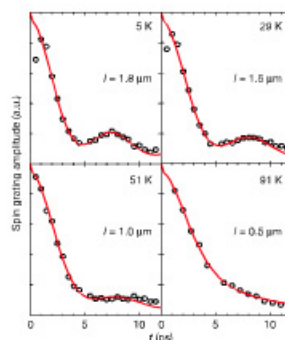


Figure 2 Time dependence of the spin grating's amplitude. The lines are fits of the data to $S(q, \omega)$. The values of l determined from these fits are indicated in each panel. Owing to laser heating, the temperature T_e of the electron gas is higher than the lattice temperature indicated.

where equation (1) extrapolates from the small- q limit⁶ to the ballistic regime. In attempting to fit the grating decay curves in the plateau regime, we found that equation (1) is not quite sufficient to describe the data. It is necessary to add to the Fourier transform of $S(q, \omega)$ a small, slowly decaying exponential with relative initial amplitude ~ 0.1 and characteristic time $\sim 25 \text{ ps}$. We speculate that this slow exponential may originate from a small fraction of localized electrons. The solid lines through the data points in Fig. 2 show the results of the fitting procedure, with fitting parameters l , v_s and the amplitude and time constant of the slow exponential. Despite the complicating presence of the slow exponential, we believe that the fits give an accurate indication of l , as this is the only parameter that determines the rate at which the oscillations are damped. Finally, the spin diffusion coefficient is determined from the relation $D_s = v_s l/2$.

The temperature dependence of D_s obtained from our analysis of the spin-grating dynamics is shown in Fig. 3 for quantum wells of different electron density. For the two lower-density samples (middle and lower panels), the dynamics were diffusive at all T , consistent with their lower mobility. To characterize charge transport in the same set of samples, we performed 4-probe measurements of the 2D charge conductance, σ_{xx} , carrier density, n , and mobility, μ , on chips from the same set of wafers. Together, these measurements allow us to test the assumption that the scattering processes that control spin diffusion and charge conduction are the same. The link between conductance and diffusion coefficient is the Einstein relation, $D_s = \sigma_{xx}/e^2 \chi_s$, where σ_{xx} and χ_s are the spin conductance and

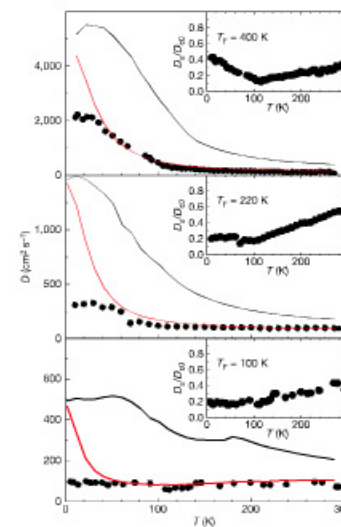


Figure 3 Comparison of motion of spin and charge, for samples with the Fermi temperatures (T_F) shown. Dots (main panels), spin-diffusion coefficients D_s determined from optical measurements. Black lines, quasiparticle diffusion coefficients D_0 determined from transport data. Insets, D_s/D_0 . Red lines, D_s predicted from spin Coulomb drag theory, taking $\chi_s = \chi_0$.

susceptibility, respectively. If the spin and charge scattering rates were the same (that is, $\sigma_s = \sigma_c$), then D_s would equal $(\chi_0/\chi_c)D_{s0}$ (ref. 7), where $D_{s0} = \sigma_c/\rho_s$ and $\chi_0 = N_F(1 - e^{-T_0/k_B T})$ is the non-interacting susceptibility (see Supplementary Information; E_F is the Fermi energy, N_F is the density of states at E_F , and k_B is Boltzmann's constant). Physically, D_{s0} is the quasiparticle diffusion coefficient¹¹, approaching $\mu E_s/e$ and $\mu k_B T/e$ in the degenerate and non-degenerate regimes, respectively. D_{s0} calculated from the 4-probe transport data and plotted in Fig. 3 is considerably larger than D_s at all T and for each of the samples. The ratio is far greater than can be accounted for by many-body enhancement of the spin susceptibility, as the factor χ_0/χ_c is less than 1.4 in this range of electron density¹⁰.

The contrast in the diffusion coefficients of charge and spin is surprising, as the assumption $D_s = D_{s0}$ is widely used in modelling spin transport in semiconductors. However, this assumption fails to take into account e - e collisions, whose rate can be much faster than those of impurity or phonon scattering. The e - e scattering events can be ignored in the description of charge transport because they conserve total momentum. However, they can have a profound effect on spin transport, as illustrated in Fig. 4. For the collision depicted between electrons with opposite spin, the charge current is conserved while the spin current reverses direction.

D'Amico and Vignale have proposed that the microscopic process shown in Fig. 4 can change the nature of macroscopic spin transport. Seen macroscopically, e - e collisions transfer momentum between the spin-up and spin-down populations, creating a force damping their relative motion that these authors term 'spin Coulomb drag' (SCD)¹⁰. Spin diffusion, which requires a counterflow of the spin populations, is damped by SCD, while charge diffusion is not. (The recently observed^{11,12} spin Hall effect also involves the counterflow of spin populations, and so should be damped by SCD.) According to D'Amico and Vignale¹⁰, the reduction of D_s relative to D_{s0} is:

$$\frac{D_s}{D_{s0}} = \left(\frac{\chi_0}{\chi_c}\right) \frac{1}{1 + |\rho_1|/\rho} \quad (2)$$

where $\rho = 1/\sigma_c$ is the charge resistance and ρ_1 is the spin drag resistance, parameterizing the rate of momentum exchange between spin \uparrow and \downarrow electrons. D'Amico and Vignale, and Hensberg and Jensen¹³, have calculated $\rho_1(T)$ for a 2DEG using the random phase approximation, obtaining results that depend only on the electron density of the quantum well.

Equation (2) predicts that despite the complex T dependences of the individual diffusion coefficients, their ratio depends primarily on the single factor, $|\rho_1|/\rho$. We test this prediction in Fig. 5, without invoking any assumptions or adjustable parameters, by plotting D_{s0}/D_s (the inverse of equation (2)) versus $|\rho_1|/\rho$ for each of the three samples measured in this study. The transport coefficients are taken directly from our measurements, while ρ_1 was calculated using equation (2) of ref. 14. The resulting graph reveals the simple linear dependence of D_{s0}/D_s on $|\rho_1|/\rho$ predicted by equation (2) over a

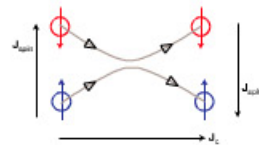


Figure 4 | A representation of e - e scattering that does not conserve spin current. Before the collision the spin current, $I_{\text{spin}}^{\text{dir}}$, is positive; after, it is negative. The charge current, I_C , does not change. Colours correspond to spin states.

large range of $|\rho_1|/\rho$, implying that SCD is indeed the origin of the large suppression of D_s relative to D_{s0} . The fact that the slope is slightly greater than unity is consistent with the expectation that the many-body enhancement of χ_c relative to χ_0 is small in this density regime¹⁰. Finally, the fact that D_{s0}/D_s extrapolates to near unity as $|\rho_1|/\rho \rightarrow 0$ indicates that the spin and charge diffusion coefficients approach each other in the limit that the spin drag resistance becomes smaller than the ordinary resistance. This result provides independent evidence that the spin grating and four-probe techniques used in this work accurately measure equilibrium spin and charge transport coefficients, respectively.

Returning to the T dependence shown in Fig. 3, the solid lines show the prediction of equation (2) for D_s , with the factor χ_0/χ_c set equal to unity. As could be anticipated from the discussion of Fig. 5, SCD quantitatively accounts for the suppression of D_s relative to D_{s0} over a broad range of temperature and electron density. It is clear, however, that the measured D_s consistently departs from theory below 40 K. We believe that this discrepancy indicates that at low T the photoexcited electron gas does not cool to the lattice temperature. If the electron gas retains the heat, Q , deposited by the excitation, its temperature T_e will rise to approximately $(T^2 + 2Q/\beta)^{1/2}$, where $\beta = 5.3 \times 10^5 \text{ eV cm}^{-2} \text{ K}^{-2}$ is the temperature coefficient of the electronic specific heat. We estimate $Q = 4.8 \times 10^6 \text{ eV cm}^{-2}$, assuming that each absorbed photon deposits approximately 10 meV (the energy width of the laser pulse) into the Fermi sea. The resulting estimate for the minimum T_e is indeed $\sim 35 \text{ K}$.

Finally, we note that SCD can be highly advantageous for spintronic applications, as it increases the distance that a spin packet can be dragged by an electric field, E , before it spreads owing to diffusion¹⁵. The length L_D that a packet of width w will drift before it broadens by a factor of two is $w^2 \mu / D_s$. In the absence of SCD, the ratio μ / D_s equals $e/k_B T_F$ (where T_F is Fermi temperature) or $e/k_B T$ in the degenerate or non-degenerate regimes, respectively, and L_D/w is independent of the underlying scattering rates. In the degenerate regime, for example, $L_D/w = eE w / k_B T_F$; drifting a spin packet further than w is only possible in a strong E limit, where the potential drop across the packet exceeds the Fermi energy. Introducing SCD slows the counterflow of spin \uparrow and \downarrow electrons without affecting their co-propagation, amplifying L_D/w by the factor $1 + |\rho_1(T)|/\rho$. Clean materials with strong e - e scattering will have the largest values

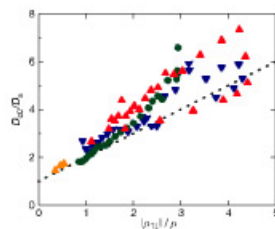


Figure 5 | Relation between suppression of spin diffusion and spin drag resistance. x-axis, ratio of $|\rho_1(T)|$ (where ρ_1 is the spin drag resistance), determined from SCD theory¹⁰, to measured resistivity, ρ . y-axis, ratio of quasiparticle diffusion coefficient, D_{s0} , to spin diffusion coefficient, D_s . Temperature is an implicit parameter. Points are for samples with $T_F = 400 \text{ K}$ (red), 220 K (green) and 100 K (blue). Points for $T < 40 \text{ K}$ are not shown because the electrons do not cool below 40 K . Orange points for a sample with $T_F = 400 \text{ K}$, with μ increased by depositing a portion of the Si dopants into the well. Line has unity slope and intercept, indicating the prediction of equation (2) for $\chi_s = \chi_0$. For points above the line, $\chi_s > \chi_0$.

of $|\rho_1(T)|/\rho$, and hence be the best media for propagation of spin information.

METHODS

Quantum well characteristics. The GaAs/Ga_{0.3}Al_{0.3}As samples were grown in the (100) direction by molecular beam epitaxy, and each consist of ten quantum wells of thickness 12 nm, separated by 48 nm barriers. The Si dopants were deposited in eight single atomic layers at the centre 14 nm of each barrier to maximize their distance from the 2DEG. The carrier concentration, n , mobility, μ , and electrical resistivity, ρ , were measured using 4-probe transport techniques without illumination. For the samples with n of 7.8×10^{11} , 4.3×10^{11} and $1.9 \times 10^{11} \text{ cm}^{-3}$ per quantum well, at low temperature μ reached 240,000, 92,000 and $69,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively.

Optical methods. The two interfering beams that generate the optical-helicity wave derive from a Ti:sapphire laser, which produces a train of optical pulses with duration 100 fs, interpulse separation 11 ns and centre wavelength 820 nm. The incident power density for most measurements was $\sim 500 \text{ W cm}^{-2}$, corresponding to $\sim 6 \text{ W cm}^{-2}$ absorbed per quantum well. For $T > 35 \text{ K}$, grating decay rates did not change when measured at incident powers down to 100 W cm^{-2} , suggesting that photoconductive holes do not play a significant role in the electron spin transport (typical electron hole recombination times were $\sim 750 \text{ ps}$). At low T the grating decay rate increased slowly with decreasing power, consistent with the electron heating model described in the main text.

The grating wavevector was directed along the GaAs (001) direction. To detect the induced spin grating, we mixed diffracted and transmitted probes to produce a photodetector current linear in the diffracted field^{16,17}. The decisive advantage in this scheme is realized by modulating the relative phase of these two beams sinusoidally at 1.2 kHz. Synchronous detection with a lock-in amplifier at the modulation frequency leads to considerable rejection of laser noise and stray light.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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