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Measurement of spin diffusion in semi-insulating GaAs

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We use optical transient-grating spectroscopy to measure the spin diffusion of optically oriented electrons in bulk, semi-insulating GaAs(100). Trapping and recombination do not quickly deplete the photoexcited population. The spin diffusion coefficient of 88±12 cm^2/s is roughly constant at temperatures from 15 K to 150 K, and the spin diffusion length is at least 450 nm. We show that it is possible to use spin diffusion to estimate the electron diffusion coefficient. Due to electron-electron interactions, the electron diffusion is 1.4 times larger than the spin diffusion. © *2011 American Institute of Physics*. [doi:10.1063/1.3592272]

The burgeoning field of semiconductor spintronics relies on moving spin-polarized electrons through distances comparable to the dimensions of an electronic device. The importance of spin transport has led to several studies of spin diffusion in GaAs quantum wells. Spin transport in quantum wells can differ markedly from that in the bulk material due to different scattering rates and, especially, to the different spin-orbit coupling.[1] Nonetheless, there have been relatively few measurements[2–4] of spin diffusion in bulk GaAs. In n-doped samples with $n = 1 \times 10^{16}$ and 2×10^{16} cm^{-3}, the spin diffusion coefficient D_s ranged from 10 to 200 cm^2/s.

Spin diffusion in semi-insulating GaAs (SI-GaAs) has not been reported. SI-GaAs has been proposed as a platform for nuclear spintronics[5] due to its low carrier density. Moreover, Kikkawa *et al.* showed that electrons could be optically oriented in SI-GaAs and would subsequently diffuse into an adjacent ZnSe film, maintaining their spin polarization.[6] Given that SI-GaAs is a ubiquitous substrate material for thin film growth and for spintronic devices, such spin diffusion is of practical consequence, whether intentional or not. In this work, we find that SI-GaAs has a large, temperature-independent spin diffusion coefficient.

We measured spin diffusion with an ultrafast transient spin grating,[7] which measures the decay rate γ_s of a spin-density wave (the "grating") with wavelength Λ and wavevector $q = 2\pi/\Lambda$. The grating amplitude decays—through spin relaxation, electron-hole recombination, and diffusion—at a rate of

$$\gamma_s(q) = D_s q^2 + 1/\tau_0. \quad (1)$$

Here, D_s is the spin diffusion coefficient, and τ_0 is the lifetime for trapping, recombination, and spin relaxation. Measurement at several q determines D_s. We measure in a reflection geometry, and improve the detection efficiency by heterodyne detection.[8] Noise is further suppressed by a 95 Hz modulation of the grating phase and lock-in detection.[9]

The SI-GaAs sample was grown by Wafer Technology. It was undoped, oriented (100), and had a room temperature resistivity $\rho \geq 10^7$ Ω-cm and a Hall mobility $\mu_H \geq 5000$ cm^2/V-s.

The pump and probe pulses came from a mode-locked Ti:Sapphire laser with a wavelength near 800 nm and a repetition rate of 80 MHz. The two pump pulses were focused on a spot 65 μm in diameter with a total fluence of 3.0 μJ/cm^2 except as indicated; probe pulses were always a factor of 2.5 weaker. Assuming one photoexcited electron per absorbed photon in a 1 μm absorption length,[10] we photoexcite $\sim 8.5 \times 10^{16}$ cm^{-3} carriers, greater than the typical concentration of deep traps in SI-GaAs.[11] In this way we are able to measure the motion of free carriers at times longer than the trapping time, and to use the density of photoexcited electrons as an estimate of the free-carrier density.

Figure 1 shows typical results of spin-grating measurements. After dropping rapidly for 0.5 ps, the diffracted signal—and the spin grating amplitude—decays exponentially at a rate of γ_s. Higher-q gratings decay more quickly, as expected for diffusive motion. The solid lines show fits of the data to the form $A + B\exp[-\gamma_s(q)t]$. The size of the constant offset A averages 2.5% of the exponential decay (and never exceeds 7%), so it does not significantly influence the values of γ_s. We speculate that the offset might arise from a small fraction of localized carriers.

We determined the spin diffusion coefficients by fitting $\gamma_s(q)$ to Eq. (1), as shown in Fig. 2(a). The measured D_s represents electron spin diffusion with no appreciable contribution from the holes. The near-bandgap absorption of circularly polarized light by GaAs excites spin-polarized electrons and holes, but the hole spins rapidly randomize, leaving behind spin-aligned electrons.[12] By fitting to times after the rapid initial decay, we obtain γ_s for electrons only. Moreover, the photoexcited electron and hole populations are spatially uniform, so the electrons' motion is not hindered by electron-hole Coulomb attraction, and spin diffusion may occur more quickly than ambipolar diffusion.[7]

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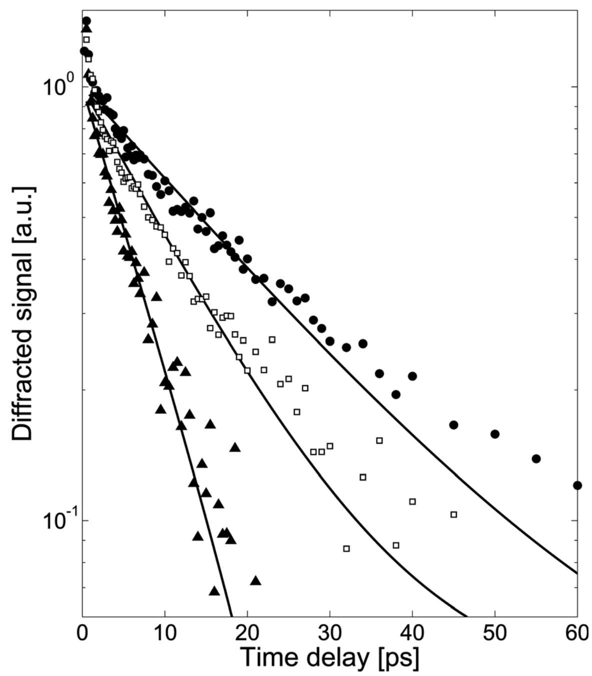

FIG. 1. Decay of the transient spin grating at 15 K (semilog scale). Curves correspond to wavevectors $q = 2.01\times10^4$, 3.14×10^4, and 4.53×10^4 cm^{-1} (slowest to fastest). Solid lines are a least-squares fit to an exponential decay plus a constant offset.

Uniform excitation also precludes any photorefractive grating.

The 15 K data in Figs. 1 and 2(a) were all taken with a pump fluence of 3.0 μJ/cm^2. The signal size decreases at higher temperatures, so data were taken with fluences of up to 9.0 μJ/cm^2. We found that the fluence had a small effect on the grating decay rate: as compared to 3.0 μJ/cm^2, decay rates measured at 6.0 to 9.0 μJ/cm^2 were typically slower by 5% to 20%, whereas attenuating to 1.5 μJ/cm^2 did not change γ_s at all. The origin of this fluence-dependent grating decay is a topic of further research. However, the measured changes in γ_s are sufficiently small that they do not add much uncertainty to the measured D_s.

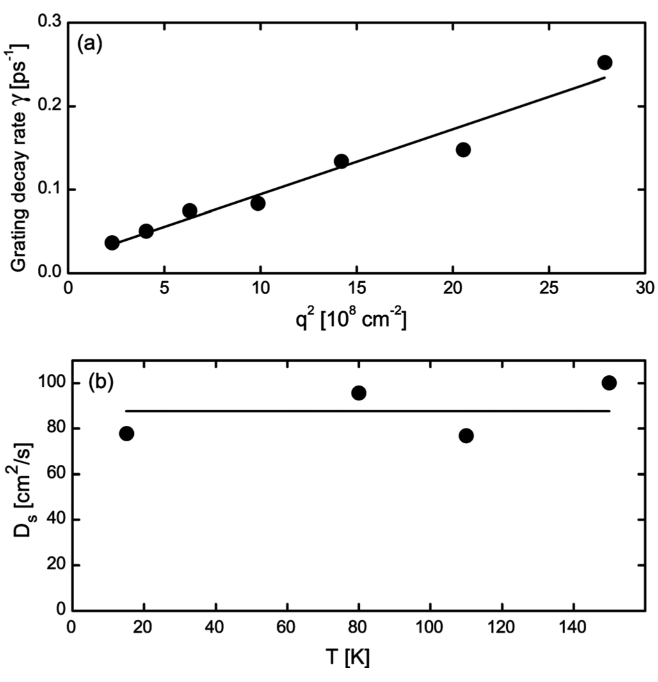

FIG. 2. (a) Decay rate of the spin grating vs q^2 at 15 K. The line is a least-squares fit to the form of Eq. (1), indicating diffusive behavior with $D_s = 78$ cm^2/s. (b) Spin diffusion coefficient vs temperature. The line is the mean value of 88 cm^2/s.

Figure 2(b) shows the values of D_s at several temperatures as determined from the fits to Eq. (1). The spin diffusion is roughly constant in temperature, and it is fast—comparable to that previously seen[9] at low temperature in a quantum well with $\mu = 69000$ cm^2/V-s. This supports our conclusion that most photoexcited electrons remain mobile for times of at least τ_0. (The values of τ_0 were 60, 100, 28, and 23 ps at 15, 80, 110, and 150 K, respectively). Noting that the spin lifetime $\tau_s \geq \tau_0$, the spin diffusion length $L_s = \sqrt{D_s\tau_s}$ is at least 450 nm. This length, comparable to the optical absorption length, suggests the possibility of efficient spin injection from SI-GaAs into thin epilayers.[6]

Finally, we infer electron diffusion from the measured spin diffusion. One must consider spin Coulomb drag,[13] the effect of electron-electron collisions that transfer momentum between counter-diffusing spin-up and spin-down populations. This effect has been observed to suppress spin diffusion, relative to electron diffusion, by factors of up to 8 in high-mobility quantum wells.[9] The electron diffusion is[14]

$$D_e = \frac{D_s}{\chi_0/\chi_s - \chi_0 e^2 \rho_{\uparrow\downarrow} D_s}. \tag{2}$$

Here, χ_s is the spin susceptibility, $\chi_0 = \partial n/\partial\mu$ is the electronic susceptibility, and $\rho_{\uparrow\downarrow}$ is the spin transresistivity. Our inferred value of D_e thus should be regarded as an estimate, because it depends—through χ_s, χ_0, and $\rho_{\uparrow\downarrow}$—on the density n and heating ΔT of the photoexcited electrons, which are known only approximately.[15]

We estimate $\rho_{\uparrow\downarrow}(T)$ from Fig. 1 of Ref. 14 and calculate $\chi_0(T)$ numerically[16] for a noninteracting electron gas of density $n = 8.5\times10^{16}$ cm^{-3}, assuming spherical, parabolic bands. We use the Perdew-Wang parametrization[17] for the exchange-correlation energy to obtain $\chi_0/\chi_s \approx 0.82$. Equation (2) gives values of D_e ranging from 110 to 140 cm^2/s. D_e is consistently about 40% higher than D_s, showing the importance of electron-electron interactions even in this high-resistivity material.

As a check on D_e, we convert diffusion to mobility using the Einstein relation, $\mu_e = e\chi_0 D_e/n$. We find that μ_e increases from 7500 cm^2/V-s at the highest measured temperature to 13000 cm^2/V-s at the lowest. We are not aware of any measurement of electron mobility in SI-GaAs at low temperature. However, our mobility agrees reasonably with that of *undoped* n-GaAs at low temperature,[18] while the room-temperature mobilities of SI-GaAs and of undoped n-GaAs are comparable.[11,18,19] This rough agreement shows that transient spin gratings can complement transport in measuring D_e. The technique might be particularly useful in ferromagnets with a large anomalous Hall effect.

Our observation of roughly temperature-independent spin diffusion contrasts with the apparent strong temperature dependence in n-doped GaAs: a sample with $n = 2\times10^{16}$ cm^{-3}

had a $D_s = 10$ cm^2/s at 4 K (Ref. 3), and one with $n = 1 \times 10^{16}$ cm^{-3} had a $D_s = 200$ cm^2/s at room temperature.[4] In the absence of electron-electron interactions, the spin diffusion would equal the charge diffusion, $n\mu/e\chi_0$. Lowering the temperature decreases n/χ_0, but it generally increases the mobility, two effects that partly cancel. At low T, n/χ_0 approaches a nonzero minimum value due to degeneracy. Because our photoexcited density exceeds the densities studied in n-doped samples, degeneracy sets in at a higher temperature. Thus for the same $\mu(T)$ we would expect diffusion in our samples to equal that of n-doped samples at 150 K, while exceeding it by a factor of 4 at low temperature. Differing mobilities among our semi-insulating sample and the two n-doped samples reported likely account for the remaining differences in D_s.

Semi-insulating GaAs is an important substrate for spintronic materials and structures, and it might also be useful for nuclear spintronics. We have measured its electron spin diffusion coefficient, D_s, under conditions in which most photoexcited carriers are not trapped. This coefficient, which is difficult to determine from transport measurements, measures the motion of electron spins. D_s has a high value of ~ 88 cm^2/s, independent of temperature.

Our result suggests that SI-GaAs could be useful for injecting spin into adjacent layers. It also holds a lesson for optical-orientation experiments in which a film is measured on top of a SI-GaAs substrate: unless a barrier-layer is grown, spins in the substrate will likely diffuse into the material being measured. Finally, we have shown that one can use spin diffusion to estimate electron diffusion, if one takes proper account of electron degeneracy (through χ_0) and of electron-electron interactions through the spin susceptibility and spin Coulomb drag.

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