Magnetization Measurements of Magnetic Two-Dimensional Electron Gases

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We directly measure the magnetization of both the conduction electrons and Mn^{2+} ions in (Zn,Cd,Mn)Se two-dimensional electron gases (2DEGs) by integrating them into ultrasensitive micromechanical magnetometers. The interplay between spin and orbital energy in these magnetic 2DEGs causes Landau level degeneracies at the Fermi energy. These Landau level crossings result in novel features in the de Haas–van Alphen oscillations, which are quantitatively reproduced by a simple model.

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The quantum Hall effect in two-dimensional electron gases (2DEGs) arises in large part from the interplay between delocalized, current-carrying states near the center of each Landau level and localized states which lie between the Landau levels (LLs). Transport measurements probe the localized states near the Fermi energy (E_f), but provide very little direct information about the total density of states (DOS) of a 2DEG. In contrast, measurements of magnetization (or any other thermodynamic quantity) are a powerful tool for exploring the total DOS of a degenerate Fermi gas. Recent advances in high-sensitivity magnetometry have allowed magnetization studies of 2DEGs in the quantum Hall regime, and have provided new insights into the total DOS of 2DEGs formed in GaAs [1–4].

In this Letter, we employ newly developed micromechanical cantilever magnetometers [5] to study the magnetization of 2DEGs in a radically different physical regime from the previous work. Here, the 2D electrons in a (Zn,Cd,Mn)Se quantum well interact strongly with the local magnetic moments of the Mn^{2+} ions via *s*-*d* exchange [6]. This produces a large conduction band spin splitting that dominates over the cyclotron energy and creates repeated crossings of spin-split LLs with increasing magnetic field. By measuring the 2DEGs' magnetization, we map out in detail its DOS under these circumstances. This is of particular interest because the mixing of nearly degenerate LLs is believed to play a crucial role in the still unresolved question of the localization of 2DEGs in small magnetic fields [7], as well as in the topology of the quantum Hall phase diagram in multisubband systems [8]. We observe unusual features in the magnetization resulting from the LL crossings. The data agree with a simple model based upon the disorder-broadened, single-particle DOS and supported by complementary transport and optical measurements on the same samples.

The samples are grown by molecular beam epitaxy (MBE) in two stages. The first stage, which ultimately forms the micromechanical cantilever, is an insulating $GaAs/Al_{0.7}Ga_{0.3}As$ heterostructure grown in a dedicated III-V MBE chamber. It is capped and transferred in an inert atmosphere to a dedicated II-VI MBE chamber for the growth of a symmetrically modulation-doped, 10.5 nm wide, $Zn_{1-x-y}Cd_xMn_ySe$ quantum well, wherein 1/16 monolayer MnSe is deposited digitally for every seven monolayers of $Zn_{0.87}Cd_{0.13}Se$ (giving an average Mn composition $y \sim 0.009$). Low temperature transport measurements confirm the formation of a 2DEG in these samples [9]. Mesas of the II-VI heterostructure are patterned by optical lithography and a BCl_3 reactive ion etch. The exposed III-V heterostructure is then processed into free-standing cantilevers, each of which supports a II-VI mesa at its end [Fig. 1(a)] [4,5]. The levers are typically $\sim$350 μm long, 50 μm wide, and 100 nm thick, with a calculated torsional spring constant $\gamma \sim 3 \times 10^{-12}$ Nm. The II-VI mesas are $40 \times 100\ \mu m^2$ and dominate the moment of inertia of the cantilever, lowering its resonant frequency ν_0 to $\sim$400 Hz. At low temperatures ($T < 10$ K) and in zero applied field, the cantilevers have a mechanical quality factor $Q \sim 2 \times 10^4$.

The cantilever is mounted in the vacuum space of a ^{3}He cryostat with an 8 T superconducting solenoid whose field (B) is normal to the 2DEG to within a few degrees. The cantilever's displacement is detected by a fiber-optic interferometer using $\sim$30 nW from a 1310 nm laser diode. Pointing the fiber near the base of the cantilever minimizes unintentional illumination of the 2DEG. Since the cantilever is not metallized and the light is well below the band gap of GaAs, very little power is coupled into the cantilever/2DEG structure. We measure ν_0 by driving the cantilever at its lowest flexural resonance with a phase-locked loop, the output of which is read by an oven-stabilized frequency counter.

As the cantilever oscillates, the sample rotates relative to B, and any anisotropic component of the sample's magnetic moment results in an additional restoring torque on the cantilever. This produces a shift in ν_0 proportional to $\partial^2 F/\partial\theta^2$, where F is the free energy of the sample and θ is the sample rotation relative to B. This torsional oscillator approach offers a number of advantages: it provides maximum sensitivity for B applied normal to the 2DEG plane;

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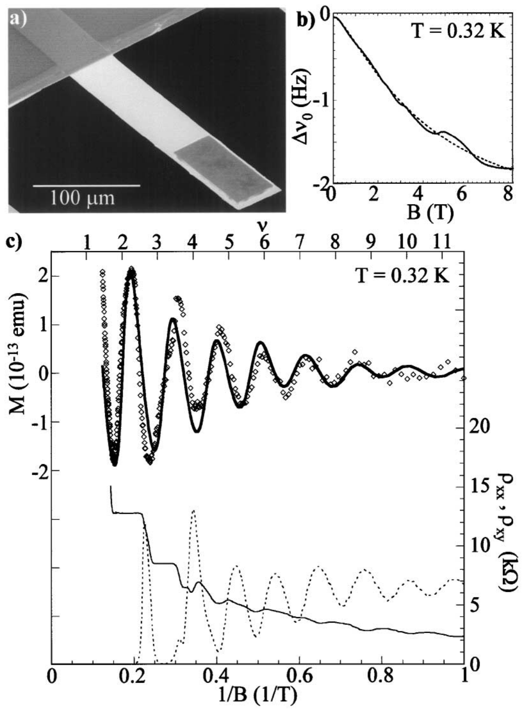

FIG. 1. (a) Scanning electron microscope image of 100 nm thick GaAs cantilever supporting $40 \times 100\ \mu m^2$ magnetic 2DEG mesa. (b) Solid line: change in resonance frequency of a cantilever containing a magnetic 2DEG vs applied field B. Dashed line: fit to the paramagnetic Mn contribution. (c) Open points: magnetization of the magnetic 2DEG, after subtracting the smooth background. These aperiodic oscillations are reproduced by the simulation (thick line) described in the text. The top axis shows the positions of integers filling factor $\nu = nh/Be$ assuming $n = 2.8 \times 10^{11}$ cm^{-2}. Thin (dashed) line: longitudinal (Hall) resistivity ρ_{xx} (ρ_{xy}) of a sample from nearby on the same wafer. The ρ_{xx} data has been multiplied by a factor of 2 for clarity.

it provides excellent rejection of isotropic magnetizations, including the majority of background signals; and it allows us to measure samples $\sim 10^4$ times smaller than in previous studies [1–3,10], diminishing the effect of nonequilibrium eddy currents and gross sample inhomogeneities.

Figure 1(b) shows the change in resonance frequency $\Delta\nu_0$ as a function of B of a cantilever containing a single magnetic 2DEG at $T = 0.32$ K. Small oscillations corresponding to the de Haas–van Alphen (dHvA) signal of the 2DEG are superposed upon a smooth background due to the paramagnetic Mn ions. This background (which is not present in samples without Mn) [4] can be fit by calculating [11] $\partial^2 F/\partial\theta^2$ for a spin-$\frac{5}{2}$ ion with very weak single-ion and g-factor anisotropy (the Mn^{2+} ions are in a $^6S_{5/2}$ state, which is highly isotropic) as well as shape anisotropy. We note that there are roughly 10^3 spin-$\frac{5}{2}$ Mn ions for each 2DEG electron, but the ions produce a relatively small torsional signal because of their weak anisotropy.

Figure 1(c) shows this data (open symbols) with the smooth background subtracted and $\Delta\nu_0$ converted into a magnetic moment $M = \Delta\nu_0(B)/2B\gamma\nu_0(0) = (A/B)\partial^2 F/\partial\theta^2$, where A is the sample area. Note that, for a magnetic moment of fixed orientation relative to the sample, this is equivalent to the more familiar form $M = -A\partial F/\partial B$. In contrast to the sawtooth shape observed in high-mobility GaAs 2DEGs [3,4], the smoothness of the oscillations implies that the disorder broadening of the LLs is comparable to their separation. Also plotted is the longitudinal resistivity ρ_{xx} (dashed line) and Hall resistivity ρ_{xy} (thin line) measured on a sample taken from nearby on the same wafer. Low-field ($B < 0.5$ T) ρ_{xy} data from this sample show $n \sim 2.8 \times 10^{11}$ cm^{-2} (and mobility $\sim 8 \times 10^3$ cm^2/V s). The positions of the integer filling factors ν assuming $n = 2.8 \times 10^{11}$ cm^{-2} are indicated in Fig. 1(c). The data show roughly one oscillation per LL, indicating that they are spin resolved, consistent with the large spin splittings known to occur in these materials. However, the dHvA oscillations deviate from the expected $1/B$ periodicity, particularly at lower ν. The observation of identical behavior in ρ_{xx} shows that the aperiodicity is not an artifact of the background subtraction. This aperiodicity also does not result from n changing with B, since the high-B (up to 8 T) ρ_{xy} data show the same slope as at low B (with quantum Hall plateaux superposed), indicating that n remains constant [12]. In addition, the precise quantization of ρ_{xy} at $\nu = 2$ and $\nu = 3$ attests to the absence of parallel conductance at low temperatures.

In addition to the anomalous magnetic-field dependence, the dHvA oscillations in these magnetic 2DEGs also exhibit an unusual variation with temperature. Magnetization data from the same sample over the temperature range $0.32 < T < 6.0$ K shows a striking shift of the extrema to lower B with increasing T [Fig. 2(a)]. This trend is strongest at higher ν [Fig. 2(c)]; for example, the maximum at $B = 2.5$ T in 0.36 K data is shifted by a full period in the 4.3 K data. The ρ_{xx} oscillations (not shown) have the same behavior, again ruling out artifacts due to data reduction, while the ρ_{xy} data show that the shifting of the oscillations is not due to a change of n with either B or T [9].

The B and T dependence of the dHvA oscillations can be understood by considering the unusual spectrum of LLs in these magnetic 2DEGs, which results from the strong s-d exchange interaction between the local moments of the Mn ions and the spin degree of freedom of the conduction electrons. This interaction produces a spin splitting of the 2DEG (ΔE_s) which is proportional to the Mn polarization: $\Delta E_s = E_{sat} B_{5/2}\{5\mu_B B/[k_B(T + T_0)]\}$ where E_{sat} is the saturated conduction band spin splitting, $B_{5/2}(x)$ is the spin-$\frac{5}{2}$ Brillouin function, and T_0 is an empirical correction reflecting the weak d-d exchange between Mn ions [13]. Since the s-d exchange acts only on the spin degree of freedom of the 2DEG electrons, we simply add its effect to the orbital quantization and assume the absence of LL mixing. Figure 3 shows the magnetic-field dependence of the spin-split LL centers for both a

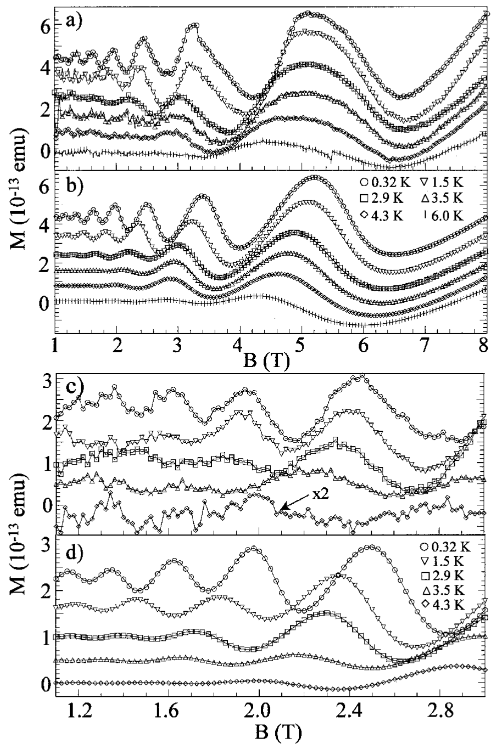

FIG. 2. (a) dHvA oscillations from the same sample as in Figs. 1(b) and 1(c) over a range of temperatures. Lower field data is shown on a magnified scale in (c). (b), (d) Calculated magnetization for each of the temperatures in (a) and (c) using the parameters from the fit in Fig. 1(c).

nonmagnetic GaAs 2DEG ($\Delta E_s = g^* \mu_B B$) and a magnetic 2DEG, where the energy for spin-split LLs is given by $E_{m,\pm} = \hbar\omega_c(m + \frac{1}{2}) \pm \Delta E_s$ (with $m = 0, 1, 2 \ldots$). Also shown is the chemical potential (thick line) $\mu \approx E_f$ in the absence of disorder broadening. The principal difference in the magnetic 2DEG is that ΔE_s is larger than the cyclotron splitting $\hbar\omega_c$ and is nonlinear in B; this leads to crossings of LLs. Figure 3(c) shows a more realistic calculation of the DOS and μ of a magnetic 2DEG, taking into account the broadening of the LLs. When the LLs cross near E_f, their overlap enhances the DOS. This forestalls the passage of E_f from one LL to the next, producing the observed aperiodicity in the dHvA oscillations. Since the crossings are created by a temperature-dependent ΔE_s, their positions shift with temperature, producing the unusual behavior in Figs. 2(a) and 2(c) [14].

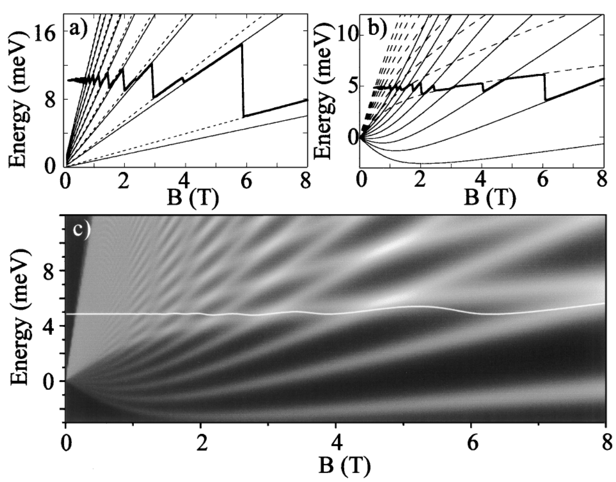

FIG. 3. Landau levels (dashed: spin down; solid: spin up) and the chemical potential (thick line) for 2DEGs in (a) GaAs and (b) a (Zn,Cd,Mn)Se magnetic quantum well in which $E_{\rm sat} = 7.7$ meV, $n = 2.94 \times 10^{11}$ cm^{-2}, $T = 0.32$ K, and $T_0 = 1.55$ K. The effect of broadening is shown in (c).

A quantitative comparison between this model and the data is provided by calculating the free energy,

$$F = n\mu - k_B T \int_{-\infty}^{\infty} g(E) \ln\left[1 + \exp\left(\frac{(E - \mu)}{k_B T}\right)\right] dE, \tag{1}$$

of the 2DEG, and from this $M = (A/B)\partial^2 F/\partial\theta^2$. We assume the DOS $g(E)$ to be a series of Gaussians, each of width $\Gamma \propto B^{1/2}$ centered at each LL [1] and normalized so that the total degeneracy of each LL is Be/h. μ is calculated at each B by requiring that n remains fixed. The few parameters in this calculation are tightly constrained by independent measurements: n is determined from low-field ρ_{xy} data ($\sim 2.8 \times 10^{11}$ cm^{-2}), while T_0 and $E_{\rm sat}$ are determined independently by fitting magnetophotoluminescence (PL) shifts to the form of ΔE_s. Magneto-PL reflects the spin splitting of the exciton, which in bulk undoped (Zn,Mn)Se is 5.7 times the conduction band spin splitting [15]. In doped quantum wells, however, this factor can range from 4 to 7.5 [16]. The saturation magneto-PL splitting in our samples is measured to be 31 meV, implying $4 < E_{\rm sat} < 8$ meV. These values are consistent with time-resolved Kerr rotation measurements that directly measure the conduction band spin splitting on the same samples. We determine $T_0 \sim 1$ K from magneto-PL data taken at $T > 6$ K in order to minimize the effect of heating of the Mn ions.

Figure 1(c) shows a comparison of the lowest temperature (0.32 K) dHvA data with a calculation of M using $E_{\rm sat} = 7.7$ meV, $T_0 = 1.55$ K, and $n = 2.94 \times 10^{11}$ cm^{-2}. The value of $\Gamma = 0.36$ meV$/T^{1/2}$ is chosen to reproduce the relative amplitudes of the oscillations. These parameters [identical to those used in Figs. 3(b) and 3(c)] give a faithful reproduction of the positions of the oscillations at $T = 0.32$ K. The agreement between the simulation and data (including the independent measurements of $E_{\rm sat}$, n, and T_0) is remarkable considering the unusual aperiodicity of the dHvA oscillations. By keeping the values of $\Delta E_{\rm sat}$, T_0, n and Γ fixed at the values given above, we produce the temperature-dependent simulations shown in Figs. 2(b) and 2(d). While the detailed shape of the oscillations is not perfectly reproduced by the calculations, the positions of all the extrema as well as their temperature dependence are in striking agreement with the data.

This suggests that—despite the unusual behavior of the dHvA oscillations—the total DOS of the 2DEG is accurately described by our model that ignores LL mixing: In other words, the positions of the LLs are not strongly affected by the crossings. While small deviations of the LLs from this model cannot be ruled out by the magnetization measurements, we have found that the same LL model also reproduces the extensive transport data from the same samples [9]. Since the extended states probed by transport are much narrower in energy than the total DOS, the modeling of transport data would be expected to be very sensitive to deviations from the model.

In summary, we have used an ultrasensitive micromechanical magnetometer to gain new insights into the total DOS of a magnetic two-dimensional electronic system, complemented by transport and optical measurements on the same sample. Striking features in the data are explained by a simple, noninteracting model for the DOS which includes the crossing of disorder-broadened LLs.

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