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Oscillatory Hall effect in high-mobility two-dimensional electron gases

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We report an unexpected anomaly in the zero-field Hall coefficient of two-dimensional electron systems in high-mobility GaAs/AlGaAs heterostructures. Our device layout allows the investigation of mesoscopic systems with variable Fermi energy as well as with tunable Hall probes. At very low temperature, both positive and negative deviations from the noninteracting Hall coefficient γ_H^0 are observed, which can be twice as large as γ_H^0 itself. A distinct regularity in the deviations and their temperature dependence are interpreted as the spontaneous formation of localized spins and their indirect exchange interaction.

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The possibility of spontaneous spin polarization (SSP) in low-dimensional electron systems is widely discussed for experiments with low Fermi energies and at low temperature. SSP has been claimed, for example, in open quantum dots,[1] in carbon nanotubes,[2] in quantum point contacts,[3] and in mesoscopic two-dimensional electron systems (2DES) in GaAs/AlGaAs heterostructures.[4,5] However, despite much work, both nature and origin of the exchange interaction that drives the spin polarization remains widely debated. Two mechanisms are generally cited in literature: many-body exchange arising from Coulomb interaction at low carrier densities,[6–9] and the possibility of a Kondo effect by virtual bound states or by localized spins in confined mesoscopic systems.[10–12] For the latter case it was suggested that a many-body spin polarization arises from an indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange,[5] which can be tuned with gate voltage in an oscillatory manner. To resolve the uncertainty in the subject, one needs to investigate the nature of spin polarization in these systems themselves, which can then be associated with the respective exchange interaction. The experimental study discussed in this Rapid Communication addresses this issue.

Most early experimental reports on SSP are based on equilibrium[3] or nonequilibrium transport spectroscopy in 1D or 2D,[4] which act only as an indirect probe of spin effects. Susceptibility measurements with tilted-field Shubnikov–de Haas oscillations[13] or direct measurements of the magnetization with a modulated magnetic field[14] also indicate nontrivial spin dynamics at low densities, but the situation remains unclear as the data have been interpreted both as the many-body Bloch-Stoner ferromagnetic instability, and as the formation of interacting or noninteracting localized moments.[14]

An alternative tool to investigate spin phenomena is the measurement of the zero-field Hall coefficient. In the presence of localized spins, electron scattering is spin selective and leads to an anomalous contribution to the Hall voltage. For years, this has been demonstrated in numerous systems, for example, in Kondo lattice compounds[15] or in magnetic semiconductors.[16,17] In nonmagnetic 2DES, Hall measurements focused mostly on the physics of the quantum Hall effect, or on electron-electron interactions in macroscopic devices.[18] However, the sensitivity of these techniques to localized spins remain unexploited so far, although it might be crucial for the understanding of the origins of SSP. We report here results from a systematic study of the quasi-zero-field Hall coefficient (γ_H) within mesoscopic regions of high-mobility nonmagnetic 2DES embedded in GaAs/AlGaAs heterostructures. At temperatures $T \gtrsim 0.8$ K, the T-dependence of γ_H indicates a Curie-type dc susceptibility ($\propto 1/T$), while at low T ($\lesssim 0.3$ K) strong deviations from the noninteracting Hall coefficient γ_H^0 lead to an oscillating γ_H as the Fermi energy E_F is varied. These results are interpreted as the formation of localized spins within the 2DES and a SSP arising from RKKY-type indirect exchange interaction.

The measurement of γ_H is carried out in mesoscopic silicon modulation-doped GaAs/AlGaAs-based high electron mobility transistors (HEMT) with an as-grown mobility of 3×10^6 cm^2/Vs and an 80 nm-thick spacer layer. For detailed material properties we refer to earlier investigations.[4,5] A purely electrostatic method for Hall measurements at mesoscopic length scales is adopted, consisting of two split-gated Hall probes on two sides of a 4×4 μm^2 or 5×5 μm^2 full gate [see Fig. 1(a)]. The coupling of the Hall probes to the central region is tunable with split-gate voltages V_G^{QPC1} and V_G^{QPC2}. During operation, both V_G^{QPC1} and V_G^{QPC2} are driven to a negative voltage such that the central mesoscopic region is connected to the Hall probes through quasi-1D channels. Both channels operate in the ballistic regime with two-probe conductance $\sim 6-10e^2/h$ at $V_G^{QPC1/2} = -0.8$ V, thereby reducing the scattering of electrons within the Hall probes themselves.

The charge density n_{2D} and the Fermi energy E_F in the central area (the active region of the device) are tuned by the voltage V_G^{FG}. The longitudinal linear conductivity (σ_{xx}) of the HEMT at 75 mK is shown in Fig. 1(b) for the full range of V_G^{FG}. The shaded region indicates the gate voltages where the T-dependence of the Hall effect is measured and corresponds to $n_{2D} = 3-5.5 \times 10^{14}$ m^{-2}. High σ_{xx} shows that the HEMT operates far away from the strongly localized regime. The Hall resistance ρ_{xy} of a device is shown in Fig. 1(c) as a function of $B_\perp$ for various E_F. To avoid any contribution from possible Hall probe misalignment, only the odd component of the Hall resistance $\frac{1}{2}(\rho_{xy}(B_\perp) - \rho_{xy}(-B_\perp))$ is considered for the evaluation of $\gamma_H = \frac{1}{2}(\rho_{xy}(B_\perp) - \rho_{xy}(-B_\perp))/B_\perp$.[19] Furthermore, the slope of the Hall resis-

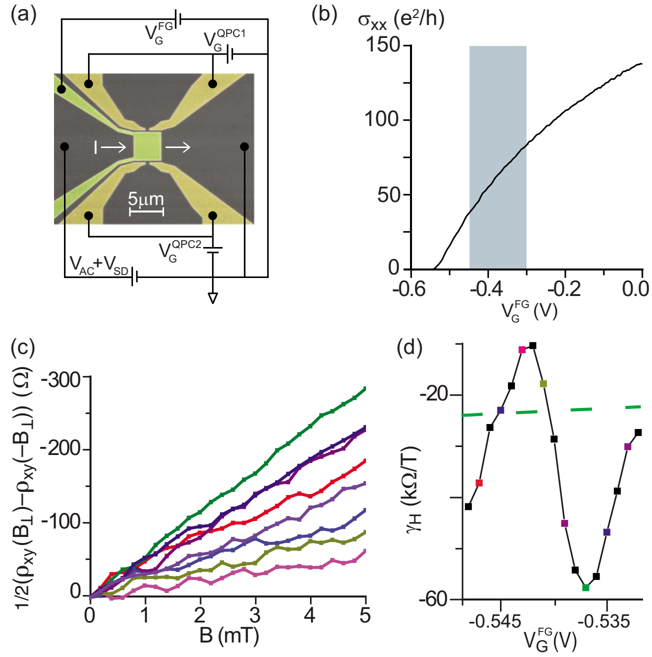

FIG. 1. (Color online) Mesoscopic Hall device: (a) Scanning electron microscopy image of a device with electrical setup. Negative V_G^{QPC1} and V_G^{QPC2} confine a 4–5 μm wide mesa. V_G^{FG} varies the Fermi energy in the central region. (b) σ_{xx} as a function of V_G^{FG}. The gray region indicates the voltage range investigated in the measurements. (c) Asymmetric Hall resistance up to 5 mT at several Fermi energies. (d) The resulting γ_H and the theoretical Hall coefficient γ_H^0 shown as dashed green/gray line. The colored (grayscale) dots correspond to the colored (grayscale) lines in Fig. 1(c).

tance is only analyzed in the low-field regime between ± 5 mT. An example of γ_H is shown in Fig. 1(d). The Hall voltage at such low fields is only a few tens of nanovolts, but this field range prevents influence from commensurability effects and quantum interference.[5] Furthermore, at such low magnetic fields the Landau level spacing is much smaller than the base temperature, making quantum Hall physics irrelevant for this experiment.

The key observation is that γ_H develops regular oscillations at low temperatures over a wide range of V_G^{FG}. An increase in n_{2D}, the field range to calculate γ_H, or application of source-drain bias V_{SD} decreases the amplitude of the oscillations.[20] The amplitude of these oscillations also decreases with increasing T (see Fig. 2). Above $T \sim 1000$ mK no oscillations are observed and the measured γ_H is consistent with the expected noninteracting value $\gamma_H^0 = -1/n_{2D}|e|$ (the orange/gray dashed line in Fig. 2). The sensitivity of the oscillations is also illustrated in the inset where γ_H is shown over the entire range of V_G^{FG}: The blue/dark gray squares are recorded at very high n_{2D} at 75 mK with a 50 mT range and the red/gray dots are recorded in the active region of n_{2D} at $T=1.7$ K with a 5 mT range; both agree well with γ_H^0.

First we focus on the overall features in the T-dependence of γ_H. Both the minima (M1, M2, etc. in Fig. 2) and the maxima (P1, P2, etc. in Fig. 2) indicate a T-dependent anomalous Hall effect (AHE), as shown in Fig. 3(a) (M points) and Fig. 4(a) (P points). Two regimes shall be emphasized: (1) For $T \gtrsim 0.8$ K and for all gate voltages, γ_H

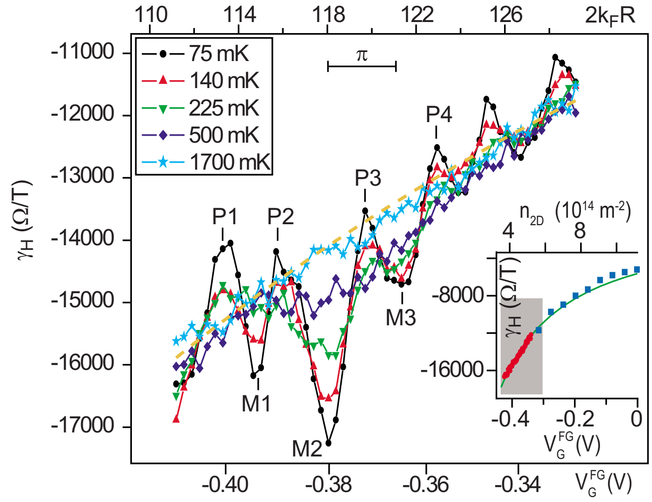

FIG. 2. (Color online) Hall coefficient at different temperatures: Asymmetric zero-field γ_H (range 5 mT) over V_G^{FG} and $2k_F R$ shows periodic fluctuations that vanish with increasing T (orange/gray dashed line corresponds to γ_H^0). Inset: γ_H over V_G^{FG} and n_{2D}. The blue/dark gray squares were recorded at $T=75$ mK with 50 mT range and the red/gray dots were recorded at $T=1.7$ K in the active region of n_{2D} (gray box) with 5 mT range. Both sets of data combine to a smooth function of n_{2D} over the full range.

decreases (becomes more negative than γ_H^0) as T is decreased. There is no oscillation in γ_H as a function of V_G^{FG} in this regime. (2) For $T \lesssim 0.3$ K, the T-dependence of γ_H at M and P points is the complete opposite [see Figs. 3(a) and 4(a)], resulting in the oscillation of γ_H as a function of n_{2D} at low T. The opposite signs of the anomalous Hall contributions contradict an explanation based on the freezing of carriers. Moreover, the regularity and the amplitude (which can be $\gg e^2/h$) of the oscillation rules out mesoscopic fluctuation effects arising from quantum interference of backscattered electrons.[21] A correction of γ_H arising from Coulomb interaction is contradicted by the oscillatory behavior and the

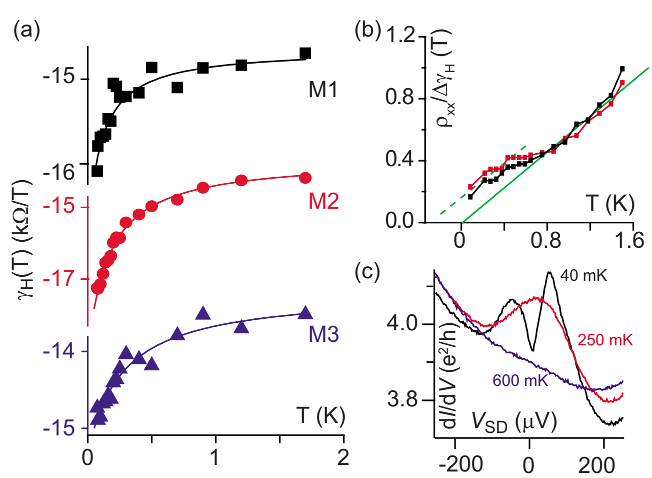

FIG. 3. (Color online) (a) The temperature dependence of M points is monotonic. The solid lines are a guide to the eyes. (b) T-dependence of $\rho_{xx}/(\gamma_H - \gamma_H^0)$ shows three distinct regimes over T. (c) Nonequilibrium transport spectroscopy shows three distinct T regimes as well.

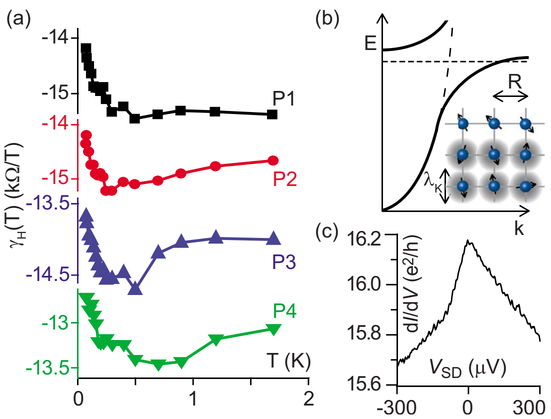

FIG. 4. (Color online) (a) The temperature dependence of P points is nonmonotonic. The upturn exceeds the noninteracting high-T value (see also Fig. 2). (b) An enlargement of the Fermi surface can lead to a change in sign in the effective mass and thus the nature of the quasiparticles. (c) Nonequilibrium transport spectroscopy shows a resonance in dI/dV at E_F, recorded at a P point.

small interaction parameter $r_s \approx 3$ for the operating n_{2D}.

However, in the presence of localized spins, a spin-dependent scattering of conduction electrons may lead to the AHE. In general, the Hall coefficient with anomalous contribution is a sum of two components:[19,22] $\gamma_H = \rho_{xy}(B_\perp)/B_\perp = \gamma_H^0 + R_S\chi$ with the dc susceptibility χ. Both sign and magnitude of the parameter $R_S = A/\mu_0\rho_{xx} + B/\mu_0\rho_{xx}^2$ depend on the spin scattering mechanism, either skew scattering ($\propto \rho_{xx}$) or side-jump processes ($\propto \rho_{xx}^2$), with numerical factors A and B and the vacuum permeability μ_0. In the presence of repulsive scatterers, both processes lead to negative corrections to ρ_{xy} at low T.[16] Here, the contribution from side-jump processes is neglected, as these were shown to be weak compared to skew scattering in GaAs systems:[23]

$$\gamma_H = \gamma_H^0 + \frac{A\rho_{xx}}{\mu_0}\chi. \quad (1)$$

Hence the magnetic nature of the system can be extracted by plotting $\chi^{-1} \propto \rho_{xx}/\Delta\gamma_H = \rho_{xx}/(\gamma_H - \gamma_H^0)$ as a function of T.

To investigate whether a model based on localized spins yields a quantitative understanding of the observations, we have measured γ_H around M points at closely spaced temperatures, and converted it to $\rho_{xx}/\Delta\gamma_H$ ($\propto \chi^{-1}$). In Fig. 3(b) we find: (1) When $T \gtrsim 0.8$ K, the solid green/gray line indicates $\rho_{xx}/\Delta\gamma_H \propto T$, which resembles the Curie susceptibility of independent spins in a paramagnetic system. This will henceforth be referred to as the free spin regime. (2) For $0.3\ \text{K} \lesssim T \lesssim 0.8$ K, $\rho_{xx}/\Delta\gamma_H$ deviates from the green/gray line and saturates at a finite value. This can be understood as Kondo screening of localized spins where the conduction electrons form a singlet with the localized spins, leading to the T-independent Pauli susceptibility. The Kondo temperature T_K, which is estimated independently from transport measurements to ≈ 300–500 mK,[5,10] fits well to the observed behavior of χ^{-1}. (3) At lower T ($\lesssim 300$ mK), the dashed green/gray line indicates $\rho_{xx}/\Delta\gamma_H \propto (T+T_0)$, implying a modified Curie-Weiss law that may arise from short-range coupling of neighboring spins.[16] The sign of the parameter T_0 indicates the dominant mode of coupling and is positive for antiferromagnetic coupling and negative for ferromagnetic coupling. In our HEMT, the limitations on the temperature range and the accuracy of the Hall voltage measurement prevent an unambiguous determination of T_0 at every M point. T_0 varies for M points but is predominantly positive ($T_0^{M1} \approx 280 \pm 150$ mK, $T_0^{M2} \approx 350 \pm 150$ mK, and $T_0^{M3} \approx 280 \pm 150$ mK).

To connect the AHE at an M point to the low-energy density of states (DOS), the differential conductance dI/dV is analyzed over the same temperature range.[5] In the free spin regime, the localized spins act only as additional scattering mechanism for conduction electrons and hence do not affect the DOS [blue/dark gray trace of Fig. 3(c)]. In the Kondo-screened regime, the antiferromagnetic coupling of conduction electrons to individual localized spins adds one virtual resonant state at each localized spin, enhancing the DOS at E_F. The corresponding zero-bias peak in dI/dV [red/gray trace of Fig. 3(c)] shows characteristic features of the Kondo effect.[5] Below $\sim$300 mK the short-range interaction between the spins dominates and suppresses the Kondo effect for $e|V_{SD}| < k_B T_0$, resulting in a double peaked DOS resonance [black trace of Fig. 3(c)]. The existence of RKKY indirect spin interaction is confirmed by an independent evaluation of the interspin distance R (≈ 1.1 μm) from magnetotransport measurements:[5] The RKKY interaction energy oscillates in 2D over the Fermi wave vector k_F as $|J(k_F)| \propto |\cos(2k_F R)|/(k_F R)^2$, leading to a periodicity of π over $2k_F R$ as observed in the separation between successive P points and M points (see upper x axis of Fig. 2).

The π-periodicity of the oscillations implies that $|J|$ is nearly zero at P points, enabling the Kondo effect to persist down to lowest T. The resulting single resonance in the DOS at E_F is shown in Fig. 4(c). A possible mechanism for the upturn in γ_H at these points may involve the Kondo screening cloud of dimension $\lambda_K = (\hbar v_F)/(k_B T_K)$,[24] around each localized spin, where v_F is the Fermi velocity. When the screening clouds of neighboring localized spins overlap, the conduction electrons can be scattered resonantly at the sites of the localized spins.[25,26] In such a scenario, a many-body phase with enlarged Fermi surface is formed [see schematic of Fig. 4(b)], quenching quasiparticle scattering and suppressing a part of the Hall voltage.[15] The upturn in γ_H is then expected below a coherence temperature, that can be $\lesssim T_K$, depending on the number of spins and screening electrons.[26–28] The plausibility of this scenario is supported by the similarity of the interspin separation R and $\lambda_K \approx 1$ μm for $n_{2D} = 3 \times 10^{14}$ m^{-2} and $T_K \approx 500$ mK. The observations thus indicate that coherent scattering might set in, but only an experiment at far lower temperature will be able to verify this scenario, by investigating whether γ_H changes its sign.

In summary, we have measured the zero-field Hall coefficient γ_H in mesoscopic 2DES and report unanticipated oscillations of γ_H over E_F at low temperature. The periodicity and temperature dependence of the oscillations confirm experimentally the existence of localized spins and their mutual interaction in GaAs/AlGaAs-based mesoscopic systems. As most low-dimensional systems are based on 2D electrons as a host, these results from mesoscopic systems could also shed new light on low-temperature transport properties and spontaneous spin polarization in nanoscopic systems, like quantum wires or dots.

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