Observation of a two-dimensional spin-lattice in non-magnetic semiconductor heterostructures

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(Dated: October 10, 2018)

Tunable magnetic interactions in high-mobility nonmagnetic semiconductor heterostructures are centrally important to spin-based quantum technologies. Conventionally, this requires incorporation of “magnetic impurities” within the two-dimensional (2D) electron layer of the heterostructures, which is achieved either by doping with ferromagnetic atoms [1], or by electrostatically printing artificial atoms or quantum dots [2, 3, 4, 5]. Here we report experimental evidence of a third, and intrinsic, source of localized spins in high-mobility GaAs/AlGaAs heterostructures, which are clearly observed in the limit of large setback distance (≈80 nm) in modulation doping. Local nonequilibrium transport spectroscopy in these systems reveal existence of multiple spins, which are located in a quasiregular manner in the 2D Fermi sea, and mutually interact at temperatures below 100 milliKelvin via the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange. The presence of such a spin-array, whose microscopic origin appears to be disorder-bound, simulates a 2D lattice-Kondo system with gate-tunable energy scales.

Unintentional magnetic impurities are expected to be absent in high-quality nonmagnetic semiconductors, such as molecular beam epitaxy-grown GaAs/AlGaAs heterostructures. Contrary to this general belief, recent observation of a Kondo-like resonance in low-energy density-states of one dimensional (1D) quantum wires indicated existence of localized spin in mesoscopic GaAs/AlGaAs-based devices [6]. Subsequently, evidence of localized spins was also reported in unconfined quasi-ballistic 2D systems [7, 8], instigating the question whether localized spins are intrinsic to GaAs/AlGaAs-based nonmagnetic semiconductors, and if so, what is the microscopic origin of these spins. A fascinating aspect of this problem is the possibility of a layer of mutually interacting spins: a system of considerable importance in studying various forms of magnetic ordering, quantum phase transitions and non-Fermi liquid effects (see Ref. [9] for a review).

In the past experiments with ballistic 1D or 2D systems [3, 4, 8], localized spins were detected with nonequilibrium transport spectroscopy. Pronounced structures, commonly known as zero-bias anomaly (ZBA), in differential conductance ($dI/dV$) of mesoscopic devices close to zero source-to-drain electric potential ($V_{SD} = 0$) were interpreted to be a consequence of spin-spin or spin-conduction electron exchange interaction. Here, we have augmented the nonequilibrium transport spectroscopy with perpendicular-field magnetoresistance measurements, which not only confirm the existence of multiple localized spins within high-mobility GaAs/AlGaAs heterostructures, but also reveals a striking order in the spatial distribution of these spins that becomes visible over a narrow range of $n_{2D}$.

Mesoscopic devices fabricated from Si monolayer-doped GaAs/AlGaAs heterostructures were used, where the 2D electron layer was formed 300 nm below the surface. A thick (≈80 nm) spacer layer of undoped AlGaAs between the dopants and the electrons provided a heavily compensated dopant layer with a filling factor $f \approx 0.9$. The resulting high electron mobility ($\sim 1-3 \times 10^6 \text{cm}^2/\text{Vs}$) provides a long as-grown elastic mean free path $\sim 6-8 \mu\text{m}$, which acts as an upper limit to the device dimensions, ensuring quasi-ballistic transport. (Micrograph of a typical device is shown in Fig. 1a.)

At low electron temperatures ($T \lesssim 100 \text{ mK}$) and zero magnetic field, both equilibrium and nonequilibrium transport display rich structures well up to linear conductance $G \sim 10 - 15 \times (e^2/h)$, as the voltage $V_G$ on the gate is increased. For most devices the structures are strongest at carrier density $n_{2D} \sim 1 - 3 \times 10^{10} \text{cm}^{-2}$ (Fig. 1b), and consist of a repetitive sequence of two types of resonances at the Fermi energy ($E_F$). This is illustrated in the surface plot of $dI/dV$ in Figs. 1c for device 1 of Fig. 1a. We denote the strong single-peak resonance at $V_{SD} = 0$ as ZBA-I, which splits intermittently to form a double-peaked ZBA with a gap at $E_F$, henceforth referred to as ZBA-II. The illustrations of ZBA-I and ZBA-II in the inset of Fig. 1c were recorded at points I and II in Fig 1b, respectively. We define $\Delta$ as the half-width at half-depth of ZBA-II. While similar nonequilibrium characteristics was observed in over 50 mesoscopic devices from 5 different wafers, reducing the setback distance below $\sim 60 - 80 \text{ nm}$ was generally found to have detrimental effect on the clarity of the resonance structures (for both ZBA-I and ZBA-II), often leading to broadening or complete suppression.

The nature of suppression of such low-energy resonances with increasing $T$ and at finite in-plane magnetic
field ($B_1$) indicate Kondo-like exchange (Figs. 1d-1g) \cite{8}, and hence presence of localized moments. A complete description can be obtained with the so-called “two impurity” Kondo model, which embodies the interaction of an ensemble of localized spins within the sea of conduction electrons \cite{10, 11, 12, 13, 14, 15}. (See Ref. \cite{8} and Supplemental Information for arguments against alternative explanation of the ZBA.) In the presence of antiferromagnetic coupling of individual spins to surrounding conduction electrons, the zero-field splitting of the Kondo-resonance (ZBA-II) arises due to a $V_G$-dependent, oscillatory inter-impurity exchange $J_{12}$, leading to the gap $\Delta \sim |J_{12}|$ at $E_F$. At certain intermediate values of $V_G$ one obtains ZBA-I when $|J_{12}| < k_B T$. For ZBA-II, nonzero $J_{12}$ results in a nonmonotonic suppression of $dI/dV$ at low bias ($|V_{SD}| \lesssim \Delta$, as indeed observed experimentally (Figs. 1e and 1g), while for ZBA-I this decrease is monotonic, and reflects suppression of single-impurity Kondo-resonance at individual noninteracting spins (see Figs. 1d and 1f) \cite{8}.

Mesoscopic devices showing clear resonances in the nonequilibrium transport also display a characteristic linear magnetoresistance (MR) over the same range of $n_{2D}$, when a small magnetic field ($B_1$) is applied perpendicular to the plane of 2D electron layer. As shown for a different device, at high $T$ ($\approx 1.4$ K), the MR consists of small peak-like structures at specific values of $B_1$, superposed on a parabolically increasing background (Fig. 2d), while at low $T$ ($\approx 30$ mK), the magnetotransport breaks into quasi-periodic oscillations irrespective of the structure of the ZBA (Fig. 2a and 2b). We note that both these evidences indicate electron transport in a mesoscopic, quasi-regular array of antidots, which has been studied extensively in artificially fabricated antidot-lattices \cite{16, 17, 18}. In the classical regime (high $T$), the peaks in MR correspond to commensurable cyclotron orbits enclosing fixed number of antidots \cite{16}, while at low $T$, quantum interference leads to the phase coherent oscillations in magnetococonductance, arising from transport along multiple connected Aharonov-Bohm rings as the inelastic scattering length exceeds sample dimensions \cite{17}.

The magnetotransport data shown in Fig. 2 allows an estimation of the inter-antidot distance $R$. In Fig. 2d, on subtracting the background, signature of commensurable orbits at $B_1 \approx 0.08$, 0.05 and 0.03 T corresponding to cyclotron radius of the electron encircling one, two and four antidots respectively (see inset). While this gives $R \approx 500$ nm (using the $n_{2D} \approx 1.31 \times 10^{10}$ cm$^{-2}$), a more accurate estimate of $R$ was obtained by Fourier transforming the low-$T$ phase-coherent oscillations of Fig. 2a and 2b. In Fig. 2c, the power spectra calculated over the range 0 to 0.065 T at both values of $V_G$ show a strong peak at the frequency $f_{AB} \approx \epsilon R^2/h \approx 105 \pm 10$ T$^{-1}$, corresponding to one flux quantum through unit cell of the antidot lattice (orbit b). This gives $R \approx 670 \pm 30$ nm, which is consistent with the estimate from commensurability effect. Satellite peaks often appears in the power spectra, for example those at $f_{AB} \approx 190$ T$^{-1}$ (orbit c) and 50 T$^{-1}$ (orbit a), which can be associated to specific stable orbits as indicated in the inset of Fig. 2a. $R$ was found to be weakly device-dependent, varying between $600 - 800$ nm, but insensitive to the lithographic dimensions of the devices.

Collectively, the observed nonequilibrium characteristics and low-field MR results indicate formation of a quasi-regular 2D spin-lattice embedded within the Fermi sea, where apart from the Kondo-coupling, the conduction electrons would also undergo potential scattering at the lattice sites. The nature of such potential scattering can be cotunneling \cite{12}, or scattering off the tunnel barrier at the localized sites \cite{19}. In this framework, the RKKY exchange between the spins naturally leads to an oscillatory behavior of $J_{12}$, where range function $\Psi(2k_F R)$ in the interaction magnitude reverses its sign with a periodicity of $\pi$ in $2k_F R$, $k_F = \sqrt{2\pi n_{2D}}$ being the Fermi wave vector. Analytically \cite{20},

$$\Delta \sim |J| \sim E_F (J \rho_{2D})^2 |\Psi(2k_F R)|$$  \hspace{1cm} (1)$$

where $\rho_{2D}$ is the 2D density of states and $J$ is the exchange coupling between an impurity spin and local conduction electron. Fig. 3a shows the direct confirmation of this, where we have plotted $\Delta$ as a function of $2k_F R$ for the device in Fig. 1c. The clear periodicity of $\approx \pi$ (within $\pm 5\%$) in $2k_F R$ can be immediately recognized as the so-called “$2k_F R$-oscillations” in the RKKY interaction, establishing the spin-lattice picture.

The absolute magnitude of $J_{12}$, and hence $\Delta$, for a 2D distribution of spins may differ widely from the simple two-impurity RKKY interaction, and would be affected by frustrated magnetic ordering or spin glass freezing \cite{21}, as well as deviation from perfect periodicity in the spin arrangements \cite{22}. Nevertheless, a framework for relative comparison of $\Delta$ in different samples can be obtained from Eq. (1) by normalizing $\Delta$ with $E_F$. As shown in Fig. 3b, adjusting for the experimental uncertainty in $k_F$ and $R$, $\Delta/E_F$ for four different devices with various lithographic dimensions can be made to collapse on the solid line proportional to modulus of pairwise RKKY range function over a wide range of $2k_F R$ \cite{20}.

With known inter-spin distance, we shall now discuss two outstanding issues of this paper: (1) the microscopic origin of the uniform array of antidots, and subsequently, (2) emergence of the localized spins. A structurally intrinsic origin of antidots in 2D Fermi sea of modulation-doped high-mobility GaAs/AlGaAs heterostructures can arise from long-range potential fluctuations in the conduction band. Direct experimental imaging of disorder in similar systems reported typical distance between fluctuation to be $\approx 0.5 - 1$ $\mu$m, in excellent agreement to the magnitude of $R$ in our devices \cite{23}. In presence of
strong correlation in the dopant layer at large $f$, theoretical investigations have also indicated a well-defined length scale in the spatial distribution of the potential fluctuations \[ \xi \].

A disorder-templated localized moment formation can then be envisaged through local depletion of electrons, analogous to moment formation in metal-semiconductor Schottky barriers \[ 10 \]. We discuss this on the basis of three common representations of disorder and screening in high-mobility GaAs/AlGaAs systems, as schematized in Fig. 4a-c \[ 23 \]. At high $n_{2D}$, the background disorder is linearly screened at all points, with local charge fluctuations $|\delta n_{2D}| \ll \langle n_{2D} \rangle$ (Fig. 4a). With decreasing $n_{2D}$, linear screening will break down locally at the maxima of slow potential fluctuations, where $n_{2D}$ becomes smaller than the local (rapid) density fluctuations of the dopants, resulting in the formation of single-particle localized states. Assuming a random distribution of the dopants, this is expected to occur at $n_{2D}^* \sim [1 - f] n_\delta / \pi^{1/2} / \xi$, where $n_\delta = 2.5 \times 10^{12}$ cm$^{-2}$ is the bare dopant density in our devices, and $\xi$ is the localization length. From $\Delta_B$ of orbit a in Fig. 2a we estimate $\xi \approx 150$ nm, which gives $n_{2D}^* \approx 1.9 \times 10^{10}$ cm$^{-2}$, which indeed marks the onset of strong ZBA (see Fig. 1b).

On further lowering of $n_{2D}$, the system crosses over to the strongly localized regime ($G \ll e^2/h$), and the 2D electron system disintegrates into puddles, which are often interconnected through quantum point contacts (Fig. 4c). While local spins can form at the point contacts \[ 4, 23 \], the commensurability effect and phase-coherent magnetoconductance oscillations shown in Fig. 2 cannot be explained in such a picture, as they require extended and uninterrupted electron orbits.

Our experiments thus outline a new microscopic mechanism of local moment formation in high-mobility GaAs/AlGaAs-based semiconductors with remote modulation doping. This has serious implications on the possibility of a gate-tunable static spontaneous spin polarization in mesoscopic devices at low temperatures, and hinged on the Kondo-coupling of the localized moments to the surrounding conduction electrons. To verify such a coupling, we have estimated the ratio $\epsilon / \Gamma$ for localized states using experimentally observed Kondo temperature $T_K$, and that $T_K \sim (E_F / k_B) \exp(\pi \epsilon / 2 \Gamma)$ is the $U \to \infty$ limit, where $\epsilon$ and $\Gamma$ are the energy of single-electron state (with respect to $E_F$), and level broadening respectively (Fig. 4b), and $U \sim e^2 / \xi \gg E_F$, is the on-site Coulomb repulsion. Taking the measured $T_K \approx 265$ mK at ZBA-I at point I in Fig. 1c as an example, we find $\epsilon / \Gamma \approx -2.1$, which confirms the Kondo regime.

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**Acknowledgement:** We acknowledge discussions with C. J. B. Ford, G. Gumbs, M. Stopa, P. B. Littlewood, H. R. Krishnamurthy, B. D. Simons, C. M. Marcus, D. Goldhaber-Gordon and K. F. Berggren. The work was supported by an EPSRC funded project. C.S. acknowledges financial support from Gottlieb Daimler- and Karl Benz-Foundation.
FIG. 1: Nonequilibrium characteristics: a, Picture of a set of two typical devices and electrical connections. The active area of a device is defined by the gate-covered region of the etched mesa. The data shown here was obtained from device 1, with device 2 and other side-gates kept grounded. b, Typical linear conductance, $G$, vs. gate voltage $V_G$ (and electron density $n_{2D}$) for device 1 at 30 mK and zero external magnetic field. c, Surface plot of differential conductance $dI/dV$ of device 1 in $V_{SD} - V_G$ plane. Each $dI/dV - V_{SD}$ trace at a particular $V_G$ was vertically shifted for leveling. The insets illustrate ZBA-I and ZBA-II-type resonances at points I and II in Fig. 1b, respectively. d, Surface plot of ZBA-I in in-plane magnetic field ($B ||$). The dashed line shows a linear monotonic splitting with effective $g$-factor $|g^*| \approx 0.5$, which confirms the role of spin. e, Surface plot of ZBA-II in $B ||$. The single-impurity Kondo-behavior dominates above $B || \sim |\Delta|/g^* \mu_B$ (the horizontal line), where $\Delta$ is the half-gap defined in text. f, Monotonic temperature ($T$) suppression of ZBA-I. g, $T$-dependence of $dI/dV$ at ZBA-II. $dI/dV$ is nonmonotonic in $T$ for $|V_{SD}| \lesssim \Delta$.

FIG. 2: Quantum and classical magnetotransport in perpendicular magnetic field ($B_\perp$). Typical linear magnetconductance oscillation at a, a single-peak resonance (ZBA-I), and b, a double-peak resonance (ZBA-II). c, Power spectra of the magnetconductance oscillations. The filled markers (blue: ZBA-I and red: ZBA-II) represent spectra obtained from the range $|B_\perp| \leq 0.065$ T, while the empty (blue) marker represents the spectrum (vertically scaled for clarity) from $0.065$ T $< B_\perp < 0.15$ T. The orbits corresponding to the peaks are indicated in the schematic. d, Four-probe linear magnetoresistance at 1.4 K for five electron densities from 1.22 (topmost trace) to $1.39 \times 10^{10}$ cm$^{-2}$ (bottom trace), where well-defined ZBA’s appear at low temperatures. Inset: Magnetoresistance after subtracting the parabolic background. The dashed lines, which denote various commensurate orbits, are computed using the average density of $1.31 \times 10^{10}$ cm$^{-2}$, and $R \approx 500$ nm.
FIG. 3: RKKY indirect exchange and "2$k_F$R-oscillations": a, $2\Delta$ from Fig. 1c as a function $2k_F R$, where $k_F$ is the Fermi wave vector, and $R$ is the inter-spin distance obtained from magnetoconductance oscillations. b, $2\Delta / E_F$ as a function of $2k_F R$ for four different devices. Solid line is proportional to the pairwise RKKY range function (see text). Local disorder governs the experimentally attainable range of $2k_F R$ within a given mesoscopic device.
FIG. 4: Schematic of background disorder and local moment formation. While the rapid fluctuations arise from dopant density fluctuations, the slow quasi-regular fluctuations indicates effect of strong correlation in the donor layer at large filling. a, large, b, intermediate, and c, low, carrier density regimes are shown. Our experimental results conform to the island-in-sea scenario of Fig. 4b.