Pseudospin Quantum Hall Ferromagnetism Probed by Electron Spin Resonance

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We study the effect of the pseudospin ferromagnetism with the aid of an electrically detected electron spin resonance in a wide AlAs quantum well containing a high quality two-dimensional electron system. Here, pseudospin emerges as a two-component degree of freedom, that labels degenerate energy minima in momentum space populated by electrons. The built-in mechanical strain in the sample studied imposes a finite "Zeeman" splitting between the pseudospin "up" and "down" states. Because of the anisotropy of the electron spin splitting we were able to independently measure the electron spin resonances originating from the two in-plane valleys. By analyzing the relative resonance amplitudes, we were able to investigate the ferromagnetic phase transitions taking place at integer filling factors of the quantum Hall effect when the magnetic field is tilted. The pseudospin nature of these transitions is demonstrated.

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At low temperatures, the multicomponent two-dimensional electron systems (2DES) possess a rich physics with a set of unconventional ground states and a nontrivial spectrum of various charge and spin excitations [1–12]. The generally accepted approach to describe the complex behavior of such structures implies the introduction of an additional internal degree of freedom of an electron, which is in many respects similar to spin and is therefore often called pseudospin by analogy. The pseudospin naturally emerges in multivalley systems, where it marks degenerate energy minima in momentum space populated by electrons [13–15]. Mechanical stress typically lifts this degeneracy and introduces a finite energy splitting between valley minima, i.e., a "Zeeman" energy between the pseudospin "up" and "down" states. Other instances of 2DES with a pseudospin degree of freedom include semiconductor heterostructures with several degenerate electron layers separated by a tunnel barrier [16] and wide quantum wells with two or more size quantization subbands occupied [2,17].

The close analogy between spin and pseudospin degrees of freedom suggests the existence of such phenomena as pseudospin ferromagnetism stabilized by electron-electron interactions [18,19] and the corresponding quantum phase transitions. For example, in the case of multivalley 2DES these transitions manifest themselves as a macroscopic redistribution of electrons between the valleys. In a zero magnetic field, the Stoner-like pseudospin phase transitions have been recently observed in the high-quality AlAs quantum wells with ultralow 2DES sheet densities of the order of $\sim 10^{10}$ cm^{-2} in the temperature range of 100 mK [20,21]. Such low densities and temperatures ensure that the kinetic energy of the electrons is negligibly small if compared to the characteristic energy of Coulomb repulsion. Another way to "freeze" kinetic motion of electrons is to apply an external magnetic field that turns their energy spectrum into a set of discrete Landau levels. Thus, high magnetic fields considerably relax the aforementioned strict constraints on the electron density and sample temperature allowing one to observe the wealth of ferromagnetic phase transitions associated with the change in spin and pseudospin polarization, even in samples with a rather average quality [22,23]. The investigation of pseudospin ferromagnetism remains of increasing interest since many of the novel atomically thin semiconductors possess such a degree of freedom either in the form of a valley or a layer "label" [7,24,25].

The most common approach to study the pseudospin ferromagnetism is to capture the evolution of the sample magnetoresistance while gradually varying several parameters, including the "pseudospin" energy splitting [26], the 2DES sheet density [2,27], or the tilt angle of the magnetic field with respect to the structure surface [28]. In large magnetic fields the phase transition then reveals itself as a hysteretic spike in the 2DES magnetoresistance, as the formation of ferromagnetic domains with different pseudospin polarization allows for the enhanced dissipation in their boundaries. Again, a close analogy may be drawn with

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the spin ferromagnetism observed in pseudospinless 2DES [22,23]. However, the transport measurements have several significant drawbacks. For example, the spike typically emerges only when the characteristic size of the nucleating domains is comparable to the 2D channel width, i.e., the electrons cannot percolate from one side to the other without crossing the domain boundaries, thus, the transport approach works well only in the close vicinity of the transition. Furthermore, it may be difficult to distinguish the spin and pseudospin ferromagnetic transitions from each other judging by the sample resistance alone, especially at large filling factors, as they both lead to the formation of similar spikes. At low temperatures ≤ 100 mK the resistance spikes typically vanish [29] complicating the studies of the pseudospin ferromagnetism even further. Other techniques are optical experiments, including photoluminescence [30] and Raman scattering [31], magnetocapacitance measurements [32], and nuclear magnetic resonance studies [4].

We suggest an alternative way to probe the evolution of the pseudospin degree of freedom in a multivalley 2DES in the regime of the quantum Hall effect. Our approach relies on the possibility to observe the resistively detected electron spin resonances (ESR) originating from different valleys independently, as it will be discussed in further details later in the manuscript. Analysis of the relative amplitude of the resonances allows us to directly assess the occupancy and the spin state of each of the valleys and thus provides unique opportunities to measure the modification of the pseudospin polarization near the phase transition in details. The possibility to apply this method to other materials is discussed in Supplemental materials [33].

Here, we focus on the pseudospin ferromagnetism at the even filling factor $\nu = 2$ of the 2D electron system confined at a 15 nm wide AlAs quantum well grown in the [001] direction with the aid of MBE. The large cyclotron mass in such a structure (0.47 of the free electron mass) ensures the reduced values of energy splittings between Landau levels. As a result, the characteristic energy of electron-electron interactions dominates the energy scale of the quantum Hall effect leading to the pronounced role of many-body physics if compared to the conventional GaAs-based structures. Note that in GaAs the effective mass has the value of $0.067m_0$ and is an order of magnitude smaller than in AlAs. Studying an AlAs-based 2DES with a valley pseudospin offers another advantage if compared to structures with multiple electron layers, as the valley pseudospin physics is not obscured with the electrostatic effects of charge redistribution between layers [30].

The ESR detection technique was based on high sensitivity of the 2DES resistance to the absorption of electromagnetic radiation in the quantum Hall effect regime [51]. The variation of the 2D channel resistance ΔR_{xx} was measured with the aid of a double lock-in amplifier technique. As the detailed description of this approach may be found in our previous works [52], we will present

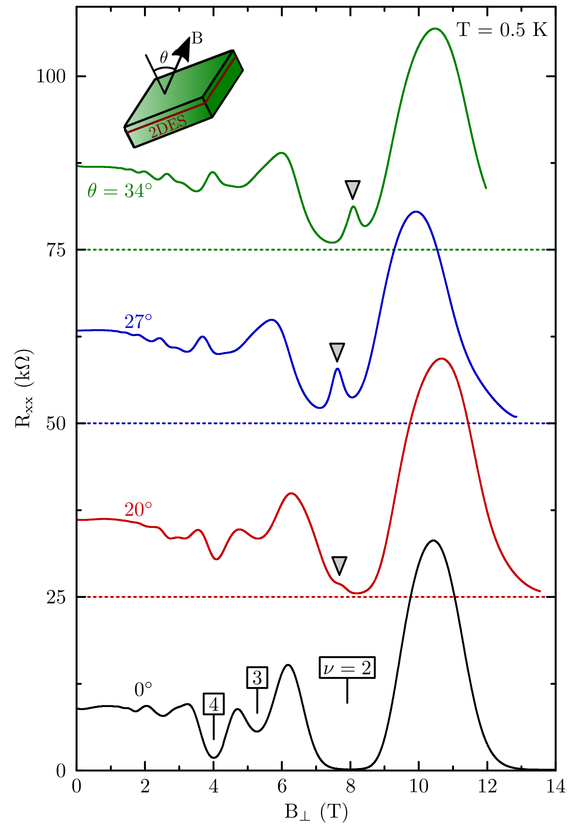

FIG. 1. Magnetic field dependencies of the sample resistance in the quantum Hall effect regime for $\theta = 0°$, 20°, 27°, and 34°. Here, θ is the angle between the magnetic field and the normal to the sample surface. The sample orientation with respect to the field is illustrated in the inset. The positions of the first several fillings ν are indicated. The curves are shifted upward for clarity. The location of the spike in sample resistance associated with the ferromagnetic phase transition is marked.

only a brief summary in Supplemental Material [33]. The experiments were performed at the temperature of 0.5 K.

Typical dependencies of the sample resistance R_{xx} on the perpendicular magnetic field are plotted for several tilt angles θ between the magnetic field and the normal to the 2DES plane in Fig. 1. Additional transport characteristics may be found in Supplemental Material [33]. The values of θ are indicated near each curve. The in-plane component was aligned with the crystallographic direction [100]. Increasing θ brought about a pronounced resistance spike located at the $\nu = 2$ quantum Hall minimum. The perpendicular magnetic field position of the spike depends rather weakly on the value of θ, in contrast to the spike at the spin ferromagnetic transition, highlighting the different origin of this feature. For comparison, in the ZnO/MgZnO heterojunction with similar density, the change in θ of less than 5° leads to the complete disappearance of the spike [23,53].

To gain a complete comprehension of the origin of the aforementioned spike at $\nu = 2$, we will now analyze the results obtained from the ESR experiments. Typical ESR

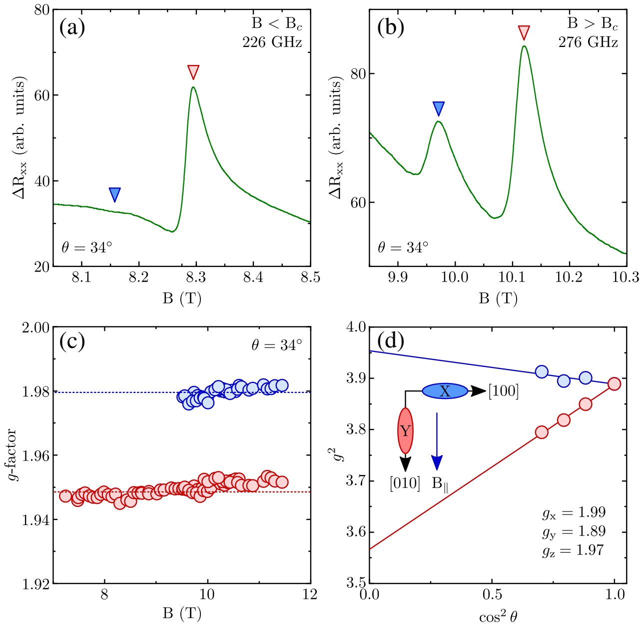

FIG. 2. Typical ESR peaks observed at $\nu = 2$ and $\theta = 34°$ below the spike [panel (a)] and above it [panel (b)]. The temperature was 0.5 K. (c) The magnetic field dependence of the electron g factors for both ESR peaks. The tilt angle was 34°. (d) The squared g factors for both ESR peaks vs the $\cos^2\theta$. Solid lines represent fits to the data using Eq. (1). The extracted eigenvalues of the g tensor are indicated in the same panel. The inset illustrates the orientation of the in-plane component of the magnetic field.

lines observed below and above the spike location B_c are shown in Figs. 2(a) and 2(b) for the case of $\theta = 34°$. The frequency of the exciting radiation is denoted near each curve. While at lower fields, one strong resonance line and one with a very small amplitude were detected, at B higher than B_c, two resonances with comparable amplitudes were present. The positions of the resonance lines are marked with arrows. This observation emphasizes the essential modification of the $\nu = 2$ ground state at B_c. For angles 20° and 27° the ESR behavior is similar, as can be seen in Sec. 3 of Supplemental Material [33].

The nature of the two-peak ESR response becomes clear if we consider the anisotropy of the spin splitting and the electron g factor in AlAs-based 2DES that has been extensively studied in Ref. [54]. The symmetry of the heterostructure dictates that each of the electron valleys is characterized by its own anisotropic g-factor tensor with three eigenvalues g_x, g_y, and g_z. Here, g_z stands for the out-of-plane g factor, while g_x and g_y define the Landé factor in the plane of the 2DES. The principal axes are [100], [010], and [001] for both tensors. Since the electron valleys translate into each other under a 90° rotation [see the inset to 2(d)], their g-factor tensors should retain that symmetry. When the magnetic field is aligned with the [100] crystallographic direction, one of the valleys has the g factor equal to g_x while the value of g for the other valley is $g_y \neq g_x$. As the resonant magnetic field B of each peak at a given frequency f is defined by the g factor according to $hf = g\mu_B B$, this nonzero difference $\delta g = g_y - g_x$ splits the peaks of the spin resonances originating from different valleys. Here, h stands for the Planck constant and μ_B denotes the Bohr magneton.

If we fix the tilt angle θ and align the in-plane component of the magnetic field with one of the principal axes, the g-factor will be given by Eq. (1). Note that exactly this orientation of the field was used in the experiment.

$$g = \sqrt{g_z^2 \cos^2\theta + g_\parallel^2 \sin^2\theta}. \quad (1)$$

Here, $g_\parallel$ stands for the electron g factor in the 2DES plane and takes the value of g_x for one valley and g_y for the other one. As a result, the g-factor difference $\delta g \sim (g_x - g_y)\sin^2\theta$ decreases for smaller angles θ and becomes exactly zero at $\theta = 0°$.

To confirm the proposed origin of the two-peak ESR we have checked the g factors extracted from the magnetic field positions of each peak at a number of radiation frequencies and tilt angles. Figure 2(c) illustrates the magnetic field dependence of the g factors for $\theta = 34°$. In full agreement with our previous findings [54], the g factor remains almost constant across the whole range of the magnetic fields studied. Based on the Eq. (1), which suggests a linear relationship between the squared g factor and $\cos^2\theta$, we have plotted the relevant experimental data with circles in Fig. 2(d) using exactly these coordinates. The solid lines represent the fits according to Eq. (1). The data fit well with that simple formula and the eigenvalues of the g tensor extracted from fitting [see inset to the Fig. 2(d)] are in good agreement with previously reported values. This finding highlights that the observed separate peaks originate from the two in-plane valleys and their relative amplitude is defined by the spin state and occupancy of each valley.

The evolution of the relative ESR amplitude $\gamma = A_1/(A_1 + A_2)$ is plotted in Figs. 3(c) and 3(d) for the case of $\theta = 27°$ and 34°, respectively. Here, A_1 denotes the amplitude of ESR with the smaller g factor and A_2—the resonance with the greater one. For each experimental point the magnetic field was calculated as an average between the magnetic field positions of two ESR peaks at a fixed radiation frequency. If only one resonance was present, the magnetic field of the second one was calculated using the corresponding g factor. For comparison we show the sample resistance in the vicinity of the spike in the panels (a) and (b) of the same figure. Both demonstrated dependencies share the same key features. In the range of magnetic fields below the spike the value of $A_1/(A_1 + A_2)$ is close to 1 suggesting that the majority of electrons resides in only one of the valleys implying that the ground state is pseudospin polarized. Furthermore, the strong ESR response indicates the substantial spin polarization of the

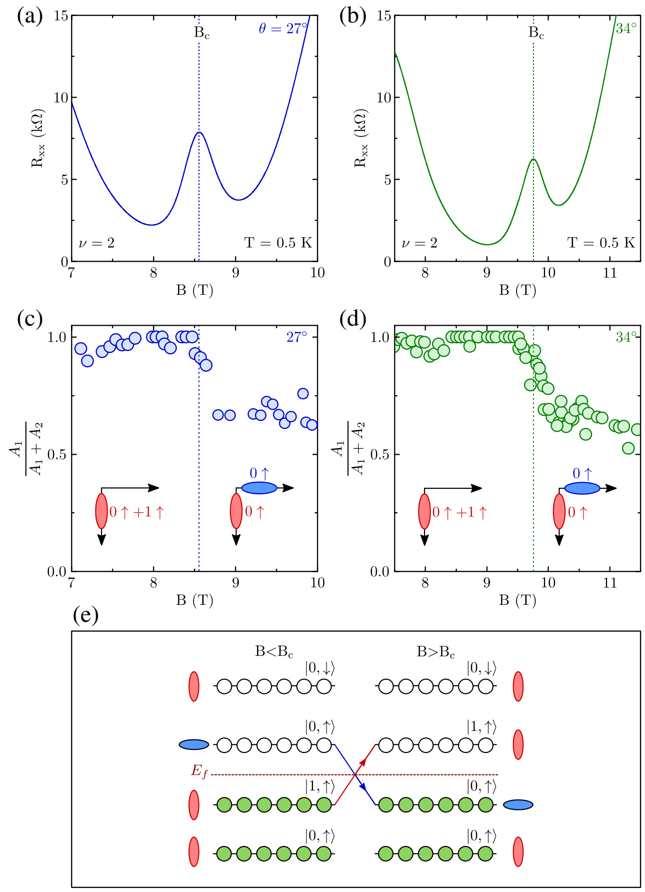

FIG. 3. (a),(b) The spike observed in the 2D channel resistance at $\nu = 2$ for $\theta = 27°$ and 34°, respectively. (c),(d) The evolution of the relative amplitude of the ESR peaks around the phase transition for the same tilt angles. The magnetic field location of the spike is indicated with a dotted line. In the inset the suggested filling of the Landau levels on both sides of the phase transition is presented. (e) The schematic representation of the energy levels crossing near the pseudospin ferromagnetic phase transition.

electron system, as well. Taking these two observations into account, we conclude that the electrons populate the two lowest Landau levels with the same spin polarization but with two different indices $N = 0$ and 1 in this range of magnetic fields. Such a level arrangement implies the formation of a spin ferromagnet at $\nu = 2$ and is in fact quite common in two-dimensional electron systems with a large cyclotron mass, as has been demonstrated in rather extensive studies of spin polarization at even fillings of the quantum Hall effect in single valley AlAs and ZnO structures [23,29,53,55]. The origin of such ordering is discussed in Supplemental Material in further detail [33].

In the vicinity of the spike the γ ratio experiences an abrupt reduction down to a value close to 0.5. Such behavior indicates the macroscopic change in the valley occupation, i.e., a rather sharp phase transition from a pseudospin ferromagnetic ordering to a paramagnetic one. In terms of Landau levels this means that the majority of electrons reside in the lowest spin-polarized $N = 0$ levels of the two valleys. Note that the detected phase transition is not accompanied with the change in spin polarization and is thus purely pseudospin in nature. At the magnetic field above the spike the γ ratio changes only slightly indicating that the paramagnetic state remains stable. The same behavior is observed for the case of $\theta = 20°$ as well, further strengthening our conclusions. The range of magnetic fields, where the value of γ undergoes a drastic change, is rather narrow ∼0.5 T and agrees well with the width of the resistance spike. This highlights that the magnetic field region where the two phases coexist is correctly measured and indicates the many-body nature of the observed pseudospin ferromagnetism, as in a single-electron picture the amplitude ratio should decay rather slowly over a very wide range of fields.

The suggested level arrangement before and after the phase transition is schematically presented in Fig. 3(e) and is discussed in further detail in Supplemental Material [33]. Such ordering implies that for a given valley splitting the system is driven through a transition by increasing the perpendicular component of the magnetic field. The energy scale of the quantum Hall state is defined by exchange interaction, cyclotron and valley splittings, and spin Zeeman energy. Both the exchange and cyclotron energies are defined by the perpendicular magnetic field and do not change in tilted fields for a fixed ν. The valley splitting is determined by the mechanical strain and is not altered at nonzero θ, as well. The only splitting that is directly affected by tilting the sample is the Zeeman energy, however, the spin state of the system at both sides of the transition is the same. This leads to a paradox that changing θ should not result in any change of the 2DES ground state, yet the perpendicular component of the critical magnetic field is clearly shifted with θ.

The possible solution to this puzzle is to consider the g-factor anisotropy. As the g factors are different for both valleys for the magnetic field orientation used in our experiments, changing θ increases the splitting of the Landau levels with the same spin and index but with different pseudospin orientation. To put it simply, tilting the magnetic field increases the valley splitting and acts, in some sense, similar to external mechanical stress. This analogy may be extended if we consider the sample resistance for a fixed perpendicular magnetic field and several θ. The resistance demonstrates a spikelike behavior (see Fig. 5 of Supplemental Material [33]): as θ is increased, resistance first rises, reaches peak value, and then drops similar to the phase transitions induced by the strain variation [19]. Increasing θ doesn't introduce any additional nonidealities of the electron system due to the inhomogeneous strain distribution. Note that similar smooth defects are believed to significantly distort the physics of the pseudospin ferromagnetism [56–58]. The valley splitting brought about by $\theta \neq 0$ is small, so that for increasing θ to drive the system through a phase transition, the system should already be close to it even for $\theta = 0°$, for instance due to the built-in strain. Crude single-particle estimates yield the magnetic field shifts of the spike ~0.2 T for $\theta = 34°$. The actual shifts may be even larger due to the *e*-*e* renormalization of the g factor observed in similar structures [59]. The suggested role of nonzero θ is further emphasized by the fact that the spike around $\nu = 2$ is observed in an anomalously wide range of θ from 20° to 34° and above indicating that changing θ shifts the relative position of energy levels rather slowly in full agreement with the proposed mechanism.

The data depicted in Fig. 3 clearly show that the ESR amplitude ratio γ deviates from being strictly 0.5 or 1 on either side of the transition. In the ideal case, these exact values would be present if electrons were equally distributed in both valleys or if only one of them was occupied. However, in real 2DES density and strain inhomogeneities may cause the observed deviations. Note that in full analogy with spin ferromagnetism [18,27] the exchange interaction stabilizes the pseudospin ferromagnetic ordering [58] and thus renders the pseudospin polarized state less susceptible to these nonidealities.

In conclusion, we have investigated the effect of the pseudospin ferromagnetism with the aid of an electrically detected electron spin resonance in a wide AlAs quantum well with a high quality two-dimensional electron system. The pseudospin degree of freedom labels the in-plane electron valleys in such a structure. The finite Zeeman splitting of the pseudospin states are introduced by the internal mechanical stress. The anisotropy of the electron spin splitting allowed us to independently measure the electron spin resonances originating from these two valleys. The analysis of the relative resonance amplitudes allowed us to investigate in detail the phase transitions occurring at integer filling factor $\nu = 2$ of the quantum Hall effect in tilted magnetic fields and to demonstrate their pseudospin nature.

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