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Anomalous spin resonance around even fillings in the quantum Hall regime

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The electron spin resonance (ESR) of two-dimensional electrons with large effective mass was studied experimentally near even filling factors of the integer quantum Hall effect. Surprisingly, the electron spin resonance did not vanish at the exact even fillings even in the case of large electron densities, where the ground state of the system was previously reported to be paramagnetic. Furthermore, the ESR amplitude was comparable between even and odd fillings. Such anomalous behavior suggests a substantial degree of spin polarization of the even fillings and was observed in two different material systems, namely AlAs quantum wells and ZnO/MgZnO heterojunctions. In AlAs quantum wells, spin resonance tended to split into two well-resolved lines near even fillings whereas it remained a single peak near odd ones. The evolution of ESR with varying filling factor around even fillings is studied in detail. The reported nontrivial findings suggest the modification of the ground state around even filling factors with the aid of strong *e-e* interaction and may be viewed as a precursor for the Stoner instability.

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I. INTRODUCTION

The discovery of the quantum Hall effect [1] has had a vast impact on the evolution of semiconductor physics, and it resulted in the creation of a whole new branch of condensed-matter research. Several decades later the field is still active [2–10]. One of the main reasons for this is that the physical picture underlying the even integer quantum Hall effect (IQHE) is far from complete, and, in fact, this phenomenon is well understood only in a rather limited case of weakly interacting electrons. This paper aims to extend the existing experimental efforts by probing the IQHE by means of electron spin resonance (ESR) in the regime, where the *e-e* interaction dominates any other characteristic energy of the two-dimensional electron system.

To evaluate the characteristic *e-e* interaction energy under the conditions of the IQHE, it is convenient to treat the IQHE first in terms of a simple single-particle model. In this case, the energy spectrum of a two-dimensional electron system (2DES) exposed to a large magnetic field B represents a set of spin-split Landau levels separated either by a cyclotron energy, $\hbar\omega_c = eB/m^*$, or by a Zeeman splitting, $\hbar\omega_z = g\mu_B B$. Here e, m^*, and g stand for electron charge, effective mass, and Landé factor, and μ_B denotes the Bohr magneton. The Coulomb interaction energy may be estimated as $E_c = e^2/\epsilon l_B$, where the magnetic length $l_B = \sqrt{\hbar/eB}$ sets the characteristic distance between the electrons, and ϵ is the dielectric constant of the medium. In typical material systems, the Zeeman splitting does not exceed the cyclotron energy, hence the ratio of Coulomb energy with respect to the cyclotron frequency $r = E_c/\hbar\omega_c$ defines the strength of the *e-e* interactions in the IQHE regime. As the parameter r is proportional to the effective electron mass, the weakly interaction regime is established in the 2DES confined in semiconductor heterostructures with small m^*, for example, in GaAs/AlGaAs quantum wells with a typical value of $m* = 0.067m_0$ [11,12].

In contrast, ZnO/MgZnO heterojunctions and AlAs/AlGaAs quantum wells boast of effective mass that is several times heavier [13,14] and reaches values higher than $0.3m_0$, ensuring the dominance of the Coulomb energy. In such structures, the vast modification of both the ground state and the excitation spectrum was reported not only for the integer [15–27], but also for the fractional quantum Hall effects [28–33]. Furthermore, at electron densities smaller than some critical value $n_c \sim 2 \times 10^{11}$ cm^{-2}, a phase transition near small even filling factors was observed, as the ground state of the system became fully spin-polarized [16,19]. To a certain degree, such behavior is reminiscent of the Stoner ferromagnetism at zero external magnetic field [34,35]. Even at large densities, where even fillings demonstrated a more conventional behavior by retaining paramagnetic ordering, the spin physics turned out to be far more complex than in the weakly interacting regime, as will be demonstrated later in the manuscript. Thus, these material systems are ideal candidates to probe the many-body physics in the IQHE regime.

A number of ESR experiments have been performed on similar semiconductor heterostructures containing high-quality 2DES with large electron mass. In Ref. [36], the spin resonance of the conduction electrons hosted at the Si/SiGe quantum well was studied in the QHE regime near the ferromagnetic even fillings. In wide AlAs wells, ESR was used to measure the electron g-factor tensor [37,38] and the effects of nuclear spin relaxation in the regime of both the integer and fractional quantum Hall effect [39,40]. The evolution of the ESR amplitude around the ferromagnetic phase transition in

ZnO-based 2DES was revealed in Ref. [41]. The renormalization of the ESR resonant frequency brought about by the spin-orbit interaction at small magnetic fields was reported in the ZnO/MgZnO heterojunction [42]. Large effective electron mass and, as a result, small splitting between the Landau levels allowed for the entanglement between the spin degree of freedom and the orbital motion of the electrons in the QHE regime in a narrow AlAs quantum well, and this coupling was resolved with the aid of ESR [43]. Note that experiments reported in this publication [43] were focused on the modification of the electron g-factor due to the spin-orbit interaction around the odd filling factors, while almost no analysis of the ESR amplitudes and widths near even fillings was performed. In contrast to the above-mentioned publications, the present paper focuses on the study of the spin resonance near nominally spin-unpolarized small even filling factors, i.e., away from the ferromagnetic phase transition and on the paramagnetic side of it, in the ZnO/MgZnO heterojunction and narrow AlAs quantum well, simultaneously.

The spin resonance of the conduction electrons in the QHE regime implies the resonant absorption of electromagnetic radiation accompanied by the intra-Landau-level spin flip. The excited state created during this process consists of a coupled electron on the upper spin branch of the Landau level and a remnant hole on the lower one and is traditionally referred to as a spin exciton. The dispersion of this excitation has been calculated for the exact odd fillings in the weakly interacting regime in Ref. [44], and it has been studied extensively with the aid of inelastic light scattering (ILS) [45–47].

In the strongly interacting limit, spin excitons have also been investigated experimentally by techniques other than ESR. The e-e interaction induced renormalization of the spin exciton dispersion has been revealed in ZnO/MgZnO heterostructures [23]. It was demonstrated that even in the presence of strong e-e interactions, their contribution to the spin splitting energy equals zero at the long-wavelength limit, and thus the Larmor theorem [48] still holds true. The evolution of the spin exciton intensity measured by means of ILS was utilized to detect the ferromagnetic phase transition around even fillings at tilted magnetic fields [19] and to investigate the depolarization of a quantum Hall ferromagnet [49], i.e., the almost fully spin polarized ground state of the system around odd fillings. Note that although ILS is an extremely powerful method to study the excitations of the 2DES ground state, it has several limitations. For example, the ILS implies resonant scattering, and thus the intensity of scattered light may depend on the wavelength of the exciting laser [see Fig. 2(a) in Ref. [20]]. Furthermore, the application of such a method to AlAs-based 2DES is complicated by the indirect band gap of the structure, thus the electron spin resonance technique turns out to be the optimal choice. Another essential difference between ESR and ILS is the wave vector k of the electromagnetic radiation used. Typical transferred k in ILS is around $0.1 l_B$ and thus the electron system is probed on the length scale of several $l_B \sim 0.1\ \mu$m under typical conditions, while in the ESR experiments the value kl_B is around 10^{-4}-10^{-5} and the properties of the system are probed on a much larger scale. Despite the above-mentioned differences, the ESR and ILS experiments yield similar results when the spin properties of the strongly correlated electron system are probed. For example, the evolution of both the ILS [19] and ESR [41] signal corresponding to the spin exciton was qualitatively the same near the ferromagnetic phase transition around even fillings, namely an intense spin exciton line was observed in the ferromagnetic phase, while the amplitude of this line became almost completely damped on the paramagnetic side of the transition.

II. EXPERIMENTAL DETAILS

The experiments reported in the present manuscript were carried on a [001] AlAs quantum well with a width of 4.5 nm and on a MgZnO/ZnO heterojunction with similar electron densities of 4.0 and 3.4×10^{11} cm^{-2}, respectively. The samples were fabricated in a Hall bar using standard optical lithographic techniques. Soldering and annealing of indium was used to form low-temperature Ohmic contacts to the 2DES. The samples were mounted inside the He^3-refrigerator of the cryostat with a superconducting magnet. The experiments were carried out at a temperature of 0.5 K and in a magnetic field up to 15 T. The magnetic field is oriented perpendicular to the plane of the 2D electron system.

The exact experimental procedure used to detect the spin resonance of two-dimensional electrons was described in detail in our previous publications [50,51]. Here we will present only a brief summary. The detection of ESR was based on the extreme sensitivity of the longitudinal resistance of the 2D channel to the microwave radiation absorption [52]. To increase the signal-to-noise ratio, the standard double lock-in technique was utilized, so that the first lock-in amplifier monitored the resistance of the channel, while the second one was tuned to the frequency of the amplitude modulation of the microwave radiation and measured the variation of the 2DES resistance due to the absorption of the radiation. Spin resonance was then observed as a peak in this variation δR_{xx} when the magnetic field was slowly swept and the radiation frequency was kept constant. The hyperfine interaction of electron and nuclear spins is known to cause dynamic polarization of nuclear spins in GaAs/AlGaAs heterostructure [53] and complicate the precise measurements of the ESR width by distorting the ESR line shape. However, under typical experimental conditions, no signs of this effect have been registered in the samples under study.

The low-temperature mobility was equal to 4×10^4 cm^2/V s for an AlAs well and 4×10^5 cm^2/V s for a ZnO-based heterostructure. The quality of the structures was good enough to observe well-developed Shubnikov–de Haas oscillations with the resistance of the sample reaching zero at high fillings, as is illustrated in Figs. 1(a) and 2(a), where the longitudinal magnetoresistance of the 2D channel in the MgZnO/ZnO heterojunction and the AlAs quantum well is demonstrated. Previously, the analysis of the magnetoplasma excitation spectrum proved that in an AlAs well of such width, electrons occupy the single valley located at the X points of the Brillouin zone along the growth direction [14]. The single-particle effective masses determined with the aid of cyclotron resonance spectroscopy turned out to be close for both material samples: $0.26m_0$ for the AlAs well [14] and $0.32m_0$ for ZnO [13].

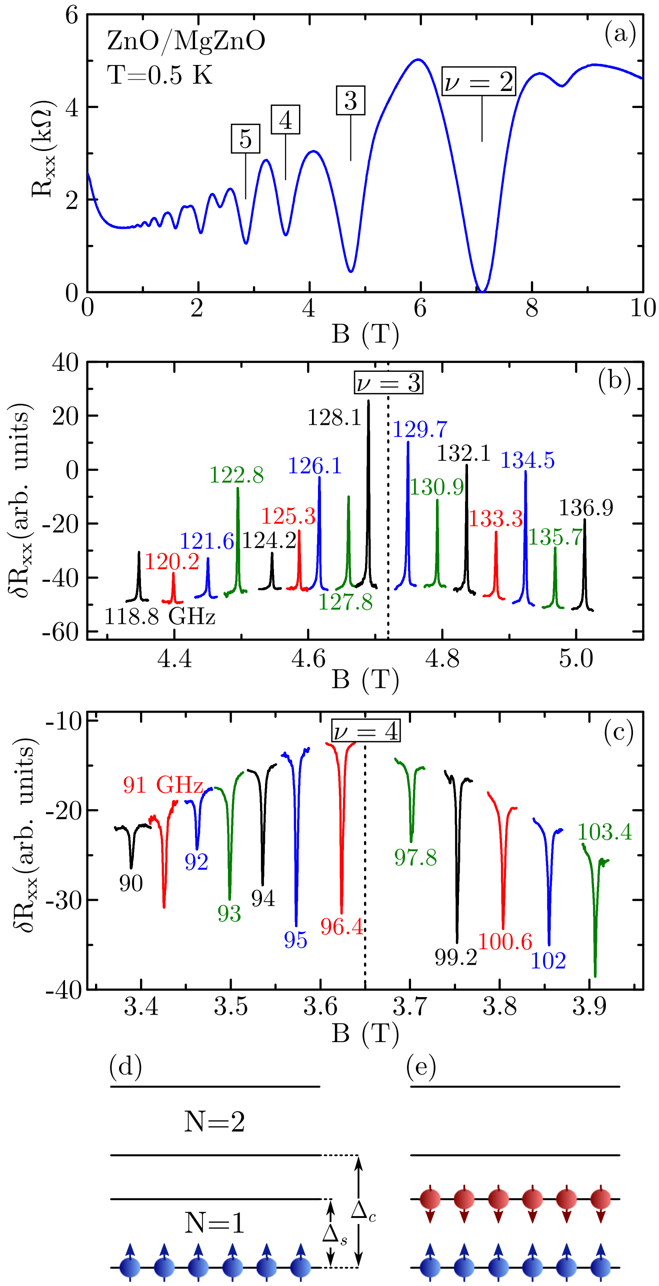

FIG. 1. (a) Typical magnetoresistance of a 2D electron channel confined at a ZnO/MgZnO heterojunction ($T = 0.5$ K). The position of the first several fillings is indicated. (b),(c) Typical ESR lines measured in the same sample in the vicinity of $\nu = 3$ and 4, respectively. The corresponding microwave frequency is indicated near each ESR peak. (d),(e) The Landau level ordering for the samples under study near the filling factor of $\nu = 3$ and 4 and the expected distribution population based on Ref. [19].

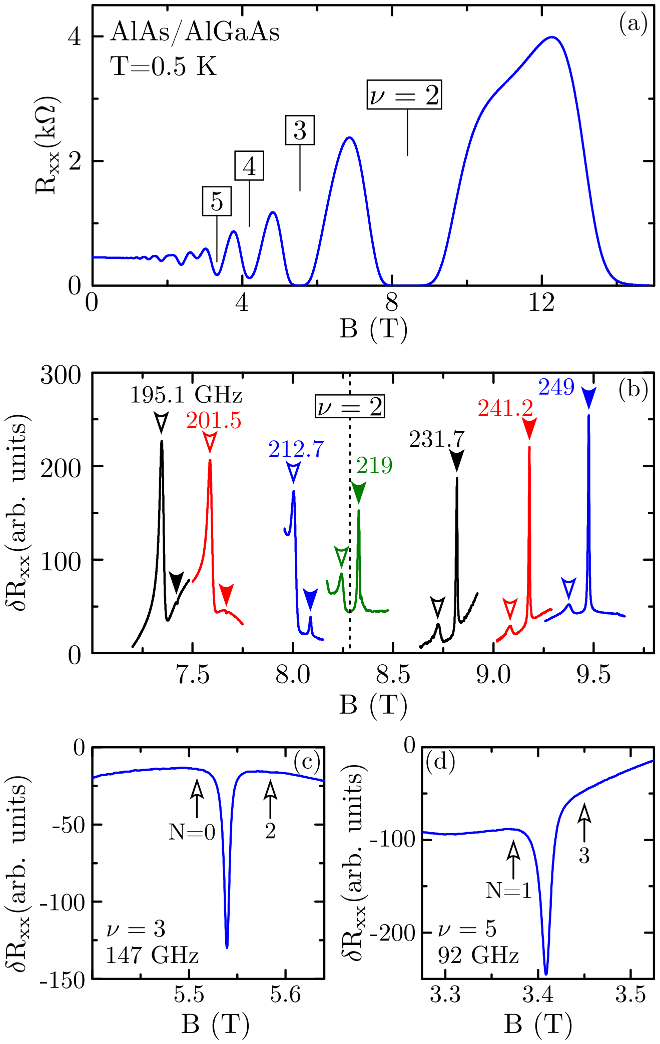

FIG. 2. (a) Typical magnetoresistance of a 2D electron channel confined at a 4.5 nm [001] AlAs quantum well ($T = 0.5$ K). The position of the first several fillings is indicated. (b) Typical ESR lines measured in the vicinity of $\nu = 2$ in the same sample, respectively. The corresponding microwave frequency is indicated near each ESR peak. (c) and (d) Typical ESR peaks observed at $\nu = 3$ and 5 in the AlAs-based 2DES. The arrows mark the expected position of spin resonances corresponding to the intra-Landau-level spin flips for the neighboring levels. The index of Landau levels is denoted.

The indicated values of m^* ensure that the selected electron concentrations are well above the critical density of the ferromagnetic transition, hence small even fillings are expected to be spin-unpolarized. The corresponding Landau level ordering near the fillings of 3 and 4 is schematically depicted in Figs. 1(d) and 1(c), respectively. In this case, the electron-spin resonance should not be observed, as the intra-Landau-level spin flips are prevented by the absence of empty states on the upper spin-split sublevel. Exactly this behavior was revealed for weakly interacting 2DES [52,54], where even fillings have paramagnetic ordering. Yet our experimental findings challenge this straightforward picture, as in both material systems studied ESR could be detected not only near odd fillings, but also at even ones with comparable amplitude. Such anomalous behavior indicates a more complex spin ordering of the system in the case of strong *e-e* interaction even far from the ferromagnetic phase transition.

III. RESULTS AND DISCUSSION

Figures 1(b) and 1(c) display ESR measured in the MgZnO/ZnO heterojunction around two neighboring filling factors $\nu = 3$ and 4, respectively. The corresponding microwave frequency is indicated near each resonance line. The ESR signal is quite intense, and the ESR linewidth is typically smaller than 2 mT, indicating slow relaxation rates and the high quality of the structure. Furthermore, the amplitude of the ESR is of the same order around even filling factor $\nu = 4$ and odd $\nu = 3$ highlighting the substantial degree of spin polarization of the even fillings.

The key to understanding the origin of this anomalous ESR around even fillings was acquired when spin resonance was studied in the AlAs quantum well. Typical ESR measured around the filling factor of 2 in the AlAs quantum well is presented in Fig. 2(b). Note that the behavior of ESR observed around a filling factor of 4 is essentially the same. The coresponding microwave frequency is indicated near each curve. Surprisingly, in this material system instead of a single resonance we observed two well-resolved ESR peaks in a vast vicinity of $\nu = 2$. Such peculiar behavior contradicts not only the ESR measurements performed in conventional GaAs heterostructures, but also the data reported here for ZnO-based 2DES. Note that around odd fillings, a more conventional single-peak ESR was detected, as is illustrated in Figs. 2(c) and 2(d) for $\nu = 3$ and 5.

Let us examine the evolution of the two-peak ESR structure with varying filling factor around $\nu = 2$. At a filling factor of $\nu = 3$, a single peak is observed. If ν is gradually decreased, at a certain ν_c an auxiliary peak becomes detectable. We would like to emphasize that the resonant magnetic field of this extra ESR is always higher than the position of the original ESR line at the same radiation frequency. The amplitude of the auxiliary line is small at first, yet it quickly rises as ν approaches a value of 2. When the filling factor is less than 2, this additional ESR line becomes dominant and the ESR line originating from $\nu = 3$ gradually disappears so that at unity filling once again the single ESR line is observed. The dependence of the relative amplitude, namely $\frac{A_l}{A_r+A_l}$, on the filling factor ν is displayed in Fig. 3(a). Here A_l and A_r stand for the absolute amplitude of the left and right ESR line.

The linewidth of these two ESR lines demonstrates essentially different behavior with varying filling factor displayed in Fig. 3(b). As ν is decreased from a value of 3, the width of the left ESR originally present near $\nu = 3$ gradually increases. In contrast, the right ESR linewidth depends nonmonotonically on ν with a maximum at the exact $\nu = 2$. Note that the presence of this maximum is established well beyond experimental uncertainty, as can be seen from Fig. 3(c), where two ESR peaks measured at $\nu = 1.96$ and 1.80 are demonstrated. The almost twofold increase of the ESR line is clearly visible when the filling factor is changed between these two values. The above-described experimental findings are yet to be understood, highlighting the need for further experimental and theoretical effort.

Further insight into the origin of the double-peak ESR structure may be gained if we analyze the electron g-factors that define the resonant magnetic field B at a given radiation frequency F as $B = \frac{hF}{g\mu_B}$. The dependence of g-factors on B is displayed in Fig. 3(d) (the data in a wider range of magnetic fields may be found in one of our previous works [43]). Note that previously narrow AlAs wells were demonstrated to have large spin-orbit interaction inherited from the bulk AlAs [43]. In this case, the entanglement of the spin degree of freedom and the orbital motion of the electron causes the energy of the intra-Landau-level spin flip to depend on the index of this level [55]. Using the data of Ref. [43], we plot the values of the g-factor for the transition inside the Landau levels with indices of $N = 0$ and 1 in the same Fig. 3(d) with solid lines. As can be seen, when the magnetic field is decreased, the g-factor of the left peak continuously reaches the value of g of the spin transition for $N = 1$. At the same time, the magnetic field position of the right peak corresponds to the g-factor of the $N = 0$ one. Judging by the evolution of the ESR amplitude, width, and corresponding g-factor, we come to the conclusion that these two peaks originate from spin resonances of electrons populating both the $N = 0$ and 1 Landau levels, indicating the presence of electrons in the lower spin-split branch of the $N = 1$ Landau level and free states in the upper spin sublevel of the $N = 0$ one.

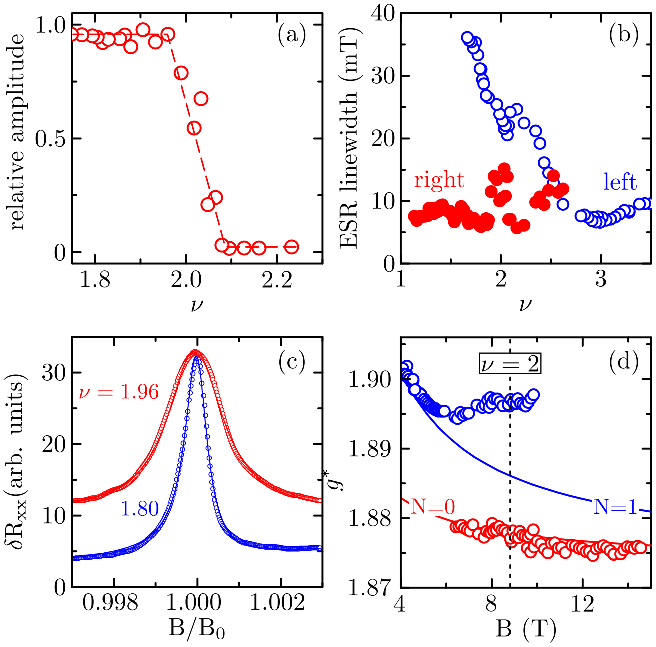

FIG. 3. (a) The filling factor dependence of the relative amplitude $[A_r/(A_r + A_l)]$ of the double-peak ESR structure observed at $\nu = 2$. Here A_r and A_l stand for the amplitude of the right and left peaks. The dashed line is a guide to the eye. (b) The evolution of the ESR linewidth for both peaks with varying filling factor. The data for the right peak are denoted by red solid circles, and those for the left one are denoted by blue open circles. (c) Typical shape of the right (in the magnetic field) spin resonance peak measured at $\nu = 1.80$ (blue circles) and 1.96 (red circles). Solid lines represent the Lorentzian fit of the data. The magnetic field is normalized by the value of the field exactly at resonance. (d) The electron g-factor defining the magnetic field position of the left (blue circles) and right (red circles) ESR peaks. Solid lines are the theoretically calculated value of electron spin resonance frequency in the presence of strong spin-orbit interaction for the $N = 0$ and 1 Landau levels.

The same pattern of Landau level population may be responsible for the anomalous ESR observed around even fillings in ZnO-based 2DES. Note that the difference in energy of the intra-Landau-level spin transition with different N is proportional to the spin-orbit constant squared [55]. The SO constant in ZnO is almost an order of magnitude smaller than in narrow AlAs wells [42], resulting in the 100 times smaller magnetic field splitting between the spin resonances of the electrons populating neighboring Landau levels. Thus the splitting of ESR into two lines is simply not resolved in ZnO-based 2DES, as the left and right ESR line should be less than 0.5 mT apart—a value smaller than the ESR linewidth.

We would like to emphasize that such an unconventional pattern of Landau level population and, as a result, nonzero spin polarization of the nominally unpolarized system is observed at low temperatures in high magnetic fields around well-developed states of IQHE and in rather clean two-dimensional systems. We believe that the observed modifications of the ground state around even fillings are brought about by the strong *e*-*e* interaction and might be viewed as a precursor for the Stoner-like ferromagnetic transition. This phenomenon may be understood in terms of the Landau level mixing caused by strong *e*-*e* interaction. Note that the Coulomb interaction is not diagonal in the set of Landau levels, and its characteristic energy exceeds any other energy scale of the 2DES studied here. Such mixing facilitates the transfer of some electrons from full Landau levels to empty ones, creating nonzero spin polarization of the electron system and allowing for the intra-Landau-level spin flips. In this sense, the data reported here may be regarded as direct evidence of the Landau level mixing in the regime of strong *e*-*e* interaction. Note that a number of examples of indirect evidence of such mixing have already been reported in the literature, including unconventional photoluminescence [19], renormalization of the electron mass and spin susceptibility [28], softening of the cyclotron spin-flip modes [20], and exchange energy renormalization [21].

Another crucial feature of the observed spin resonance lines is that the ESR signal is of a different sign at odd and even filling factors, suggesting the possible change of the detection mechanism. While the reasons for the longitudinal resistance of the 2D channel to change near ESR are well-established at exactly odd fillings [50,56], the actual mechanism at even filling remains a mystery and highlights the need for further research.

Let us compare the behavior of the spin resonance at the nominally spin-polarized even fillings close to the ferromagnetic phase transition and away from it. While ESR is intense away from the transition, as was reported earlier in the manuscript, close to it the ESR amplitude is substantially damped [41]. The ILS resonant line corresponding to the spin exciton demonstrates [19] similar behavior around the phase transition region. The possible reason for the observed difference may be the way the ferromagnetic ordering is established when the tilt angle (or electron density, or magnetic field magnitude) is varied. We suggest that at some point on the paramagnetic side of the phase transition, the nucleating domains of the ferromagnetic phase first emerge and then grow in size as the system approaches the transition. These domains may effectively scatter spin excitons that could be created in the paramagnetic areas of the system and greatly diminish their lifetime, preventing the corresponding resonant line from being detected. However, spin excitons emanating from these domains may be observed when the total area of the domains and their size are large enough. To sum up, the amplitude of both ESR and ILS spin exciton line should first drop as the system comes closer to the phase transition and then grow once again, in qualitative agreement with the previously published data. Note that the data reported in [19] for the case in which the phase transition did not occur at a filling factor of 2 were for the values of θ differing from the transition angle by just 5 degrees—an amount smaller than the spread of angles at which the transition was observed. Thus the assumption of the nucleating ferromagnetic domains present in the nominally paramagnetic state seems to still be reasonable under such conditions, and, as a consequence, the ILS line of the spin exciton is still small around a filling factor of 2 in this regime.

Another important aspect to be discussed is that the observed effects may not be ascribed either to the disorder caused by the built-in nonuniformity of the 2DES or by the thermal fluctuations, or to the Landau level mixing due to the spin-orbit interaction. Let us consider the spin resonances detected around the fillings of 2, 3, and 5 in AlAs-based 2DES. Then if such disorder is strong enough to cause the splitting of ESR near the filling factor of 2, than it would inevitably split ESR near fillings of 3 and, especially, 5. Note that near the filling factor of 5, the energy needed to transfer an electron from the lower spin branch of the $N = 2$ level to the analogous branch of the $N = 3$ level is much smaller than the energy gap at the filling factor of 2, as the magnetic field corresponding to the filling of $\nu = 5$ is 2.5 times smaller than that of $\nu = 2$. However, only single resonance lines were detected experimentally around $\nu = 3$ and 5. Typical ESR lines measured near the indicated fillings are displayed in Figs. 2(c) and 2(d), respectively. The arrows mark the expected position of ESR corresponding to the spin flips for the adjacent Landau levels. The spin-orbit interaction is not diagonal in the basis of Landau levels and may cause their mixing as well. However, the strength of SO interaction in the MgZnO/ZnO heterojunction is almost 10 times smaller than in AlAs-based 2DES, yet the Landau level mixing observed in both samples is of comparable strength, as the intense ESR was detected around even fillings in both of them.

IV. CONCLUSION

In conclusion, the electron spin resonance of two-dimensional electrons confined at a MgZnO/ZnO heterojunction and a 4.5 nm [001] AlAs quantum well was studied near even filling factors of the integer quantum Hall effect. The electron systems under study were characterized by strong electron-electron interaction. Unconventionally, the electron spin resonance remained robust at the exact even fillings even in the case of large electron densities, where the ground state of the system was reported to be spin-unpolarized. Moreover, in AlAs quantum wells spin resonance was split into two well-resolved lines near even fillings, whereas it retained its single peak structure near odd ones. The evolution of ESR with varying filling factor around even fillings is studied in detail. The observed anomalous behavior of ESR suggests the nonzero spin polarization of the ground state around even fillings conventionally viewed as paramagnetic. Such modifications of the ground state are brought about by the Landau level mixing due to the strong *e*-*e* interactions and, in some sense, may be regarded as the precursor for the Stoner-like ferromagnetic transition.

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