Rapid Communications

Exchange energy renormalization in quantum Hall ferromagnets with strong Coulomb interaction

A. B. Van'kov,[1,2] B. D. Kaysin,[1,3] S. Volosheniuk,[3] and I. V. Kukushkin[1]
[1]*Institute of Solid State Physics, RAS, Chernogolovka 142432, Russia*
[2]*Laboratory for Condensed Matter Physics, National Research University Higher School of Economics, Moscow 101000, Russia*
[3]*Moscow Institute of Physics and Technology, Dolgoprudny 141701, Russia*

(Received 25 April 2019; published 25 July 2019)

An unusual behavior of the exchange energy scale of a quantum Hall ferromagnet with $\nu = 1$ was found in strongly correlated two-dimensional electron systems based on MgZnO/ZnO heterostructures. The exchange contribution, entering the energy of a collective excitation, was probed by means of inelastic light scattering. It was established that, in a wide range of electron densities corresponding to the Wigner-Seitz parameter $7 < r_s < 11$, this contribution is on the order of the cyclotron energy, which is notably different from the typical scale of $e^2/\varepsilon\ell_B$ that is typical for weakly interacting systems. The same trend was confirmed via numerical calculations.

DOI: 10.1103/PhysRevB.100.041407

Various mysteries of condensed matter physics can be revealed through the exploration of certain model problems. Therefore the effects associated with the exchange interaction of two-dimensional electron systems (2DESs) in quantizing magnetic fields are usually analyzed in the simplest case of a quantum Hall ferromagnet (QHF) with $\nu = 1$ (see Refs. [1–4]). This state is incompressible and has a spin polarization degree close to unity. Its stability is preserved even in systems with arbitrarily small Zeeman coupling because of the significant gain in exchange energy. The minimized set of embedded correlations makes this state suitable for theoretical analysis, although a consistent account of many-particle effects is possible only if the Coulomb contributions are small compared to the cyclotron energy [5]. Probing of the exchange and correlation energies of a 2DES is typically carried out by measuring the energies of the simplest collective excitations. In high-quality GaAs-based 2DESs at $\nu = 1$, the true exchange contribution has been detected by means of Raman scattering on special magnetoexcitons, cyclotron spin-flip excitations (CSFEs) [1,2], corresponding to the transition of electrons to the higher adjacent Landau level (LL) with a simultaneous spin flip. At $\nu = 1$, the CSFE energy at all momenta contains positive many-particle contributions determined by the ferromagnetic exchange energy [6,7]. These are adequately accounted for within the perturbation theory for GaAs-based systems with a moderate interaction strength ($r_s \lesssim 1$).

With the emergence of high-quality 2DESs based on ZnO [8], which have a dielectric constant $\varepsilon = 8.5$ and a relatively large electron mass $m^* \approx 0.3m_0$, the role of Coulomb correlations became much enhanced, leading to a number of unusual quantum Hall states [9,10]. The values of r_s and the LL mixing parameter $\kappa = E_C/\hbar\omega_c$ at $\nu = 1$ increase by an order of magnitude with respect to GaAs. Although there is no rigorous theory for two-dimensional (2D) electrons under such conditions, a number of estimates and calculations have been made for the activation energy of a QHF at $\nu = 1$. Various approaches based on the Landau theory of Fermi liquids [11], excitonic representation [12], random phase approximation [13], and numerical simulations [14,15] have predicted a change in the scale of the activation energy of a QHF (or its exchange energy) from the familiar scale of $e^2/\varepsilon l_B$ to $\hbar\omega_c$ at the limit of $r_s \gg 1$. Thus far, experimental evidence of this prediction is lacking, and the magnitude of the exchange energy of a QHF was reliably established only for 2DESs with a parameter of $r_s \lesssim 1$.

In this Rapid Communication, the behavior of the exchange contribution to the energy of CSFE as a function of various parameters was studied for a ZnO-based 2DES by means of inelastic light scattering. It was found that in a wide range of electron densities corresponding to $7 < r_s < 11$, the contribution to CSFE energy at $\nu = 1$ increases approximately linearly with electron density and is comparable to $\hbar\omega_c$. This trend was also analyzed theoretically both within the framework of the Hartree-Fock approximation (HFA) and by the exact diagonalization (ED) method, where the mixing of LLs was taken into account through a screened Coulomb potential.

The experimental samples were high-quality single heterojunctions MgZnO/ZnO grown by molecular beam epitaxy with a two-dimensional electron channel formed in the ZnO layer. The parameters of the 2DES—density n_s and low-temperature mobility μ_t—were determined first by transport measurements, and then the density was specified *in situ* using the magnetophotoluminescence (PL) method, as described in previous works [16]. The actual parameters of the five experimental structures are listed in Table I. Experiments were conducted at a temperature of $T \sim 0.3$ K in a ^{3}He evaporation cryostat in the core of a superconducting solenoid. Optical access to the sample was established through two quartz fibers, one of which was used for photoexcitation, while the other was used for signal collection. The angular configuration of the fibers determined the momentum transferred from light to the 2DES. Photoexcitation was produced by a wavelength-tunable laser source operating near the direct interband optical transitions of ZnO. The excitation power density was well below 1 mW/cm^2, which prevented heating of the 2D

TABLE I. Parameters of 2DES in the set of studied samples in order of increasing electron density n_s.

Sample No.	n_s (10^{11} cm^{-2})	r_s	μ_t (10^3 cm^2/V s)
254	1.14	11.1	430
259	1.73	9.0	560
475	2.03	8.4	590
244	2.23	8.0	520
427	2.8	7.1	427

electrons. Optical spectra were detected using a spectrometer with a resolution of ∼0.2 Å. The magnetic field was tuned up to 15 T and had a normal orientation relative to the sample surface.

The inelastic light scattering signal was studied predominantly at magnetic fields corresponding to the filling factor near $\nu = 1$. Weak Raman lines from the 2DES could be distinguished from the strong PL signal based on their constant energy shift with respect to the sweeping laser's position. Their detection was facilitated by the resonant photoexcitation conditions near the interband optical transitions [17].

The identification of the Raman lines corresponding to the simplest collective excitations is made possible by considering the known properties of such excitations. Two intrasubband excitations, magnetoplasmon and spin exciton, have previously been identified and investigated in ZnO systems at different quantum Hall states [10,18]. At the long-wavelength limit, they exhibit Raman shifts close to the bare cyclotron and Zeeman energies, respectively. The CSFE energy should incorporate both of these quantities with the addition of some positive many-particle contribution [6] [see the transition diagram in Fig. 1(c)]. A CSFE is a long-lived excitation only at a filling factor of $\nu = 1$ and decays to a combination of other magnetoexcitons on either side of $\nu = 1$.

The Raman scattering signal of a CSFE at $\nu = 1$ appears as a narrow line [see Fig. 1(a)] with an energy shift exceeding the Zeeman and cyclotron energies. The CSFE line is resonantly enhanced within a spectral range of ∼2–2.5 meV. Upon deviation from $\nu = 1$, the spectral position of a CSFE changes significantly. The integrated intensity is diminished and the spectral width increases simultaneously. Therefore, the quality parameters of the Raman line of a CSFE exhibit significant reductions on either side of the filling factor $\nu = 1$ [the ν dependence of spectral intensity is shown in Fig. 1(b)]. It should be noted that line extinction is caused by its internal nature, rather than the resonant conditions for Raman scattering, because the line spectral shift in this range is several times smaller than the width of the resonant profile. This situation is reproduced at other wavelengths of incident light. From the spectral evolution of the CSFE line, one can calculate the behavior of the many-particle energy contribution. Assuming an additive structure of the excitation energy, this simply entails subtracting the single-particle terms $\hbar\omega_c$ and $g^*\mu_B B$. The residual exchange-Coulomb energy decreases on both sides of $\nu = 1$ [plotted in Fig. 1(c)], which agrees qualitatively with the expected behavior for the single-mode representation of the collective excitation CSFE [6] (indicated by dashed curves). This situation also corresponds to the behavior of

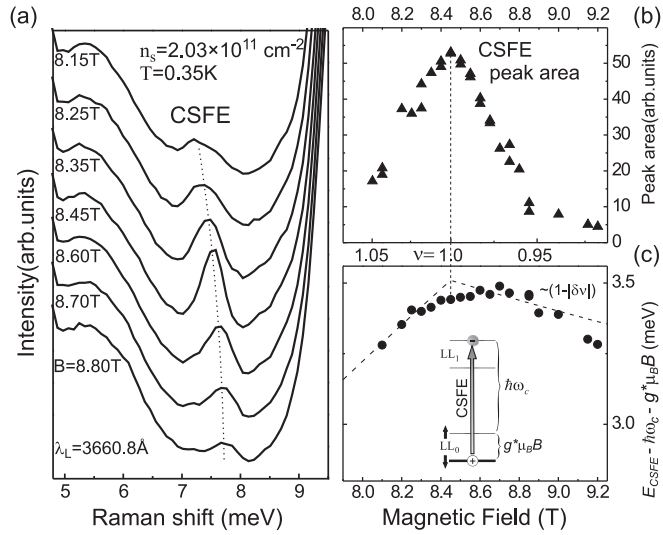

FIG. 1. (a) The waterfall of Raman spectra of CSFE at different values of the magnetic field (filling factors in the vicinity of $\nu = 1$) measured from sample No. 475. The dotted line traces the CSFE peaks. (b) Magnetic field evolution of the integral intensity of the line CSFE. (c) The magnetic field (or ν) dependence of the exchange-Coulomb energy shift of a CSFE. The fitting lines are derived from the mean-field theory (see the main text). The inset illustrates a CSFE structure as a magnetoexciton in a single-particle level scheme.

CSFE in similar experiments in GaAs [19]. The change in energy on both sides of $\nu = 1$ is associated with both a decrease in the exchange contribution [$\sim(1 - |\delta\nu|)$] and the monotonic magnetic field dependence of the Coulomb energy scale itself. The issue of the skyrmion mechanism of spin depolarization in ZnO is not even posed due to the relatively strong Zeeman coupling. The sharp maximum at $\nu = 1$ is not outlined in Fig. 1(c) either because of the incomplete spin polarization of the QHF or instrument broadening of the spectral lines.

Similar measurements of CSFE energy at $\nu = 1$ were performed on all five experimental heterostructures to establish the nature of the growth in the exchange-Coulomb contribution with electron density. Although Raman measurements were carried out with some variation in the transmission momentum, on one of the samples, the long-wavelength dispersion of the CSFE was measured explicitly. These data are presented in the inset in Fig. 2. From these data, it follows that the dispersion has a weak negative characteristic, which is confirmed by the calculations discussed below. The change in energy is insignificant and does not exceed 0.15 meV, even in the sample with the highest electron density and maximum momentum. Experimental data regarding the exchange-Coulomb contribution to the CSFE energy at $\nu = 1$ for a wide range of densities are indicated by the symbols in Fig. 2. There, the single-particle contributions $\hbar\omega_c + g^*\mu_B B$ are eliminated. One can see that the exchange-Coulomb contribution grows in an approximately linear manner and has a magnitude close to the cyclotron energy.

This energy scale is unusual for the exchange energy of QHFs and is qualitatively different from the characteristic Coulomb energy at the magnetic length (indicated in Fig. 2

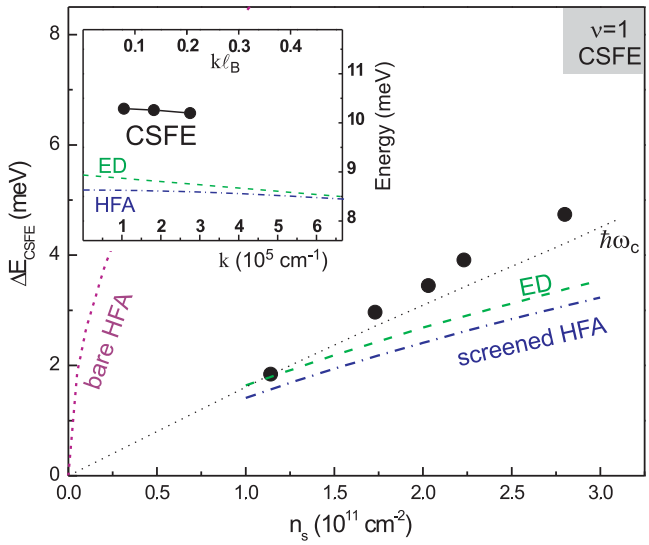

FIG. 2. Dependence of the exchange-Coulomb energy in a CSFE at $\nu = 1$ on electron density. Black symbols indicate experimental data. The black dotted line shows cyclotron energy for comparison. The green dashed line was calculated using the ED method and the blue dashed-dotted line is a simulation within a screened HFA. The purple short-dashed line shows energies obtained within an unscreened HFA. The inset compares the experimental data regarding the measured k dispersion of the CSFE in sample No. 427 ($n_s = 2.8 \times 10^{11}$ cm^{-2}) to the theoretical dispersions calculated within the ED and HFA methods. The upper axis is given in dimensionless units.

by a short-dashed curve for comparison). Although the theory of magnetoexcitons in the quantum Hall effect (QHE) regime with a strong interaction at $r_s \gg 1$ has not yet been developed, there is an approximate approach that accounts for the effects of LL mixing when considering many-particle correlations in a system. In this method, the Fourier component of the Coulomb potential can be modified by a static dielectric function calculated using the random phase approximation as follows: $V(q) = \frac{2\pi e^2}{\varepsilon q}\frac{1}{\epsilon_s(q)}$. This so-called static screening approximation [13,20] allows one to describe electronic correlations adequately at partially filled LLs, provided that $\nu > 1$ and $\kappa \gg 1$. It was previously used to calculate the Coulomb gaps in graphene in the QHE regime [21], as well as in ZnO in fractional and integer QHE states [10,22]. The same approach is applied here to qualitatively evaluate the influence of LL mixing on the scale of the exchange-Coulomb energy in CSFE. As a seed estimate, we consider expressions for both the static dielectric function and Coulomb energy terms themselves as if the electrons were confined to the lowest LL. The true configuration of the $\nu = 1$ ground state is unknown due to the strong mixing of LLs. However, as shown below, this information is not essential for tracing the qualitative transformation of the many-particle energy term.

The static dielectric function in the random phase approximation is given by [20]

$$\epsilon_s(q) = 1 - \frac{2\pi e^2}{\varepsilon q}\chi^0_{nn}(q, \omega \to 0^+).$$

Here, $\chi^0_{nn}(q, \omega)$ is the retarded density response function, which takes the following form for a noninteracting 2DES in a magnetic field,

$$\chi^0_{nn}(q) = \frac{1}{2\pi l_B^2}\sum_\sigma\sum_{k,m}|F_{k,m}(q)|^2\frac{\nu_{m,\sigma} - \nu_{k,\sigma}}{(m-k)\hbar\omega_c}, \quad (1)$$

where $\nu_{m,\sigma}$ is the filling factor of an LL m with a spin index σ. The inter-LL oscillator strength for the transition between two levels k and $m > k$ is given by

$$|F_{k,m}(q)|^2 = \frac{k!}{m!}e^{-q^2l_B^2/2}\left[L_k^{m-k}\left(\frac{q^2l_B^2}{2}\right)\right]^2\left(\frac{q^2l_B^2}{2}\right)^{m-k},$$

where $L_k^{m-k}(x)$ are the associated Laguerre polynomials.

The resulting form of the static dielectric function for the configuration with all electrons at the lowest spin LL is defined by the following expression,

$$\epsilon_s(q) = 1 + \frac{e^2/\varepsilon l_B}{\hbar\omega_c}\sum_{m=1}^{\infty}\frac{\sqrt{2}}{m\cdot m!}\left(\frac{ql_B}{\sqrt{2}}\right)^{2m-1}e^{-q^2l_B^2/2}.$$

One can see that the second term is proportional to the LL mixing parameter $\kappa \gg 1$, leading to a dramatic rescaling of the many-particle energy terms. These terms depend on the involved LLs and can be expressed in terms of the Coulomb matrix elements in the k domain. For the long-wavelength CSFE at $\nu = 1$, they are given by the following expression, which was derived from the HFA [1,6],

$$\Delta E^{k=0}_{\mathrm{CSFE}} = \frac{1}{(2\pi)^2}\int d^2q\,\frac{2\pi e^2}{\varepsilon q}\frac{1}{\epsilon_s(q)}\frac{q^2l_B^2}{2}e^{-q^2l_B^2/2}. \quad (2)$$

The CSFE energy calculated in this manner will be on the order of $\hbar\omega_c$ under the condition $\kappa \gg 1$. Arithmetically, this is the result of a reduction in the value of the Coulomb potential at $ql_B \sim 1$ in the integrand, where the screening factor reaches its maximum value $\epsilon_s \sim \kappa$. At these momenta, the main contribution to the many-particle energy of a CSFE is attained. In this naive picture, a crossover between the scales of $e^2/\varepsilon l_B$ and $\hbar\omega_c$ occurs at values of $\kappa \sim 3$ and asymptotically trends toward $\Delta E_{\mathrm{CSFE}} \approx 1.3\,\hbar\omega_c$ for $\kappa \gtrsim 100$. The calculation results for the actual range of electron densities in the HFA are represented by a dashed-dotted line in Fig. 2. For these calculations, the geometric form factor of the 2D Coulomb potential in the heterojunction with the calculated subband wave function was taken into account. The total effect of the form factor on the many-particle energy is small (<15%), due to the screening of the Coulomb potential and the narrowness of the calculated wave-function profiles.

To evaluate the effects of the redistribution of electrons over LLs on the calculated energy, one can make the ground-state structure less arbitrary, as well as the corresponding form of the screening function. This approach was implemented in the numerical calculation of CSFE energy by adopting exact diagonalization for $N = 11$ electrons in the basis of states at the two lowest LLs. In this case, the electrons at the two relevant levels can be left uncontrolled, but their interactions should be screened by all other LLs. In other words, the summation in Eq. (1) will begin from $m \geqslant 2$, thereby softening the artificial screening. The energy spectrum was calculated for a finite system with periodic boundary conditions in the

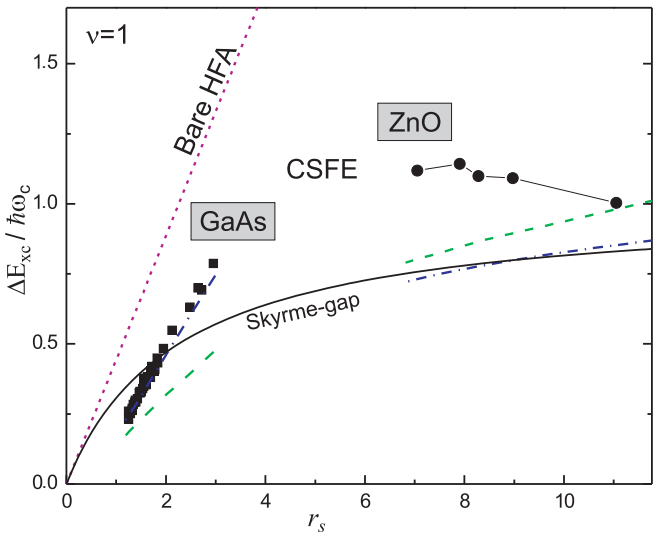

FIG. 3. Ratio of the exchange-Coulomb term in a CSFE to the cyclotron energy as a function of r_s. Square symbols represent data measured in GaAs quantum wells [2] and round symbols were recalculated from Fig. 2. HFA and ED calculations with realistic geometric form factors are indicated by dashed-dotted and dashed lines, respectively, and the bare HFA prediction is indicated by the short-dashed line. The exchange energy of a model skyrmion-antiskyrmion pair is indicated by a solid line.

geometry of a torus using the Coulomb matrix elements provided in Ref. [22]. The resulting CSFE energy at $k = 0$ for a set of actual densities is plotted in Fig. 2 as a dashed line. Again, these results are in qualitative agreement with the experimental results. Therefore, we can conclude that the use of a statically screened potential in the calculation of many-particle energy terms yields a reasonable estimate, even if one does not know the exact structure of the ground state of a QHF. Explicit accounting for ground-state blurring over the several lowest LLs does not lead to principal changes if this effect is properly accounted for in the screened potential. Calculation using ED with a larger number of explicit LLs was not performed because this would inevitably require a further decrease in the number of electrons, leading to more coarse results.

The density evolution of the exchange correlation term in the CSFE energy for the case of a strongly interacting 2DES can be reasonably compared to previous data for GaAs-based systems in terms of the dimensionless units r_s and $\Delta E_{xc}/\hbar\omega_c$. These data are plotted in Fig. 3, where the small r_s points were recalculated from the data in Ref. [2], which were measured in finite-width quantum wells. The geometric form factor considerably weakens the Coulomb contribution in GaAs-based systems, but the trend is still linear as a function of r_s, as predicted by HFA (plotted as a short-dashed line). In contrast, the large r_s points of the ZnO-based system approach saturation at $\Delta E_{xc}/\hbar\omega_c \sim 1$, which is qualitatively explained by the considerations discussed above. Calculations using screened ED and HFA are plotted as dashed lines.

More sophisticated approaches to the calculation of the many-particle energies of quantum Hall ferromagnets were proposed earlier in the context of the problem of skyrmion-antiskyrmion excitations. Although this type of neutral excitation has a qualitatively different structure, the many-particle contribution to its energy is comparable to that of a CSFE. At the limit of a strong magnetic field for a strictly 2D case with $\nu = 1$, each of them is exactly half of the specific exchange energy of the zeroth LL. At $r_s \to \infty$ and a vanishing electron g-factor, the analytical result for the skyrmion-antiskyrmion creation energy is exactly $\hbar\omega_c$, which also agrees with our result for the many-particle contribution to CSFE at $k = 0$. The analytical curve [12] for a Skyrme gap in a strict 2D case is plotted in Fig. 3 for comparison. We guess the crossover between the energy scales in both cases at approximately $r_s \sim 3$–6.

The near-linear growth pattern of the exchange-enhanced gap at $\nu = 1$ has already been observed in a series of magneto-transport experiments on GaAs-based 2DESs [3,4]. However, even when LL mixing is considered, quantitative agreement with theory was greatly hampered by the uncontrollable effect of the disorder on the transport characteristics. From this perspective, the probing of a system using inelastic light scattering is much less vulnerable because the many-particle contribution to the energy of long-wave magnetoexcitons is gained at relative distances on the order of the magnetic length. This method works equally well both in a strong-field limit on GaAs-based systems and in a weak field here in ZnO-based systems. At the qualitative level, the observed renormalization of the many-particle energy of strongly correlated 2DES in the QHE regime can serve as an explanation for the well-established pattern of the crossing of fictitious Landau levels of quasiparticles with a renormalized effective mass and g-factor. It intriguing that in ZnO-based 2DES with a strong interaction the extracted energy gaps grow almost linearly with magnetic field [9,18], that is, the linearized combination of exchange and Coulomb contributions is already embedded in these gaps.

In conclusion, the scale of the exchange energy in a quantum Hall ferromagnet with $\nu = 1$ was probed in the regime for the Wigner-Seitz parameter $r_s > 7$. The exchange-Coulomb contribution to the energy of collective excitation CSFE was measured through inelastic light scattering on a series of ZnO-based 2DESs. This contribution exhibited maximum values at $\nu = 1$ and decreased with the deviation of the filling factor due to the perturbation of the ferromagnetic order. We determined that the magnitude of the exchange-Coulomb energy scales on the order of $\hbar\omega_c$, rather than $e^2/\varepsilon l_B$. This phenomenon is caused by the strong mixing of LLs, which leads to the effective renormalization of the Coulomb interaction, expected in theory [11–13]. It was demonstrated that a good estimate for the many-particle energy terms can be obtained by performing calculations even within HFA with a statically screened Coulomb potential.

The authors are grateful to the Russian Science Foundation (Grant No. 19-42-04119) for their support in performing experiments on Raman scattering and to the Russian Foundation for Basic Research (Grant No. 17-02-00751) for support in performing the calculations. We thank S. M. Dickmann for fruitful discussions.

[1] A. Pinczuk, B. S. Dennis, D. Heiman, C. Kallin, L. Brey, C. Tejedor, S. Schmitt-Rink, L. N. Pfeiffer, and K. W. West, Phys. Rev. Lett. **68**, 3623 (1992).
[2] A. B. Vankov, L. V. Kulik, I. V. Kukushkin, V. E. Kirpichev, S. Dickmann, V. M. Zhilin, J. H. Smet, K. von Klitzing, and W. Wegscheider, Phys. Rev. Lett. **97**, 246801 (2006).
[3] A. Usher, R. J. Nicholas, J. J. Harris, and C. T. Foxon, Phys. Rev. B **41**, 1129 (1990).
[4] V. S. Khrapai, A. A. Shashkin, E. L. Shangina, V. Pellegrini, F. Beltram, G. Biasiol, and L. Sorba, Phys. Rev. B **72**, 035344 (2005).
[5] C. Kallin and B. I. Halperin, Phys. Rev. B **30**, 5655 (1984).
[6] J. P. Longo and C. Kallin, Phys. Rev. B **47**, 4429 (1993).
[7] S. Dickmann and V. M. Zhilin, Phys. Rev. B **78**, 115302 (2008).
[8] J. Falson, Y. Kozuka, M. Uchida, J. H. Smet, T. Arima, A. Tsukazaki, and M. Kawasaki, Sci. Rep. **6**, 26598 (2016).
[9] J. Falson, D. Maryenko, B. Friess, D. Zhang, Y. Kozuka *et al.*, Nat. Phys. **11**, 347 (2015).
[10] A. B. Vankov, B. D. Kaysin, and I. V. Kukushkin, Phys. Rev. B **98**, 121412(R) (2018).
[11] S. V. Iordanski and A. Kashuba, J. Supercond. **16**, 783 (2003).
[12] S. Dickmann, Phys. Rev. B **65**, 195310 (2002).
[13] A. P. Smith, A. H. MacDonald, and G. Gumbs, Phys. Rev. B **45**, 8829 (1992).
[14] I. Mihalek and H. A. Fertig, Phys. Rev. B **62**, 13573 (2000).
[15] R. Price and S. Das Sarma, Phys. Rev. B **54**, 8033 (1996).
[16] V. V. Solovyev, A. B. Van'kov, I. V. Kukushkin, J. Falson, D. Zhang, D. Maryenko, Y. Kozuka, A. Tsukasaki, J. H. Smet, and M. Kawasaki, Appl. Phys. Lett. **106**, 082102 (2015).
[17] A. B. Vankov, B. D. Kaysin, V. E. Kirpichev, V. V. Solovyev, and I. V. Kukushkin, Phys. Rev. B **94**, 155204 (2016).
[18] A. B. Van'kov, B. D. Kaysin, and I. V. Kukushkin, Phys. Rev. B **96**, 235401 (2017).
[19] A. S. Zhuravlev, L. V. Kulik, I. V. Kukushkin, V. E. Kirpichev, and V. E. Bisti, Pis'ma Zh. Eksp. Teor. Fiz. **85**, 128 (2007).
[20] I. L. Aleiner and L. I. Glazman, Phys. Rev. B **52**, 11296 (1995).
[21] W. Luo and R. Cote, Phys. Rev. B **88**, 115417 (2013).
[22] W. Luo and T. Chakraborty, Phys. Rev. B **93**, 161103(R) (2016).