Cyclotron Spin-Flip Excitations in a $\nu = 1/3$ Quantum Hall Ferromagnet

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Inelastic light scattering spectroscopy discloses a novel type of cyclotron spin-flip excitation in a quantum Hall system around the $\nu = 1/3$ filling. The excitation energy follows qualitatively the degree of electron spin polarization, reaching a maximum value at $\nu = 1/3$. This characterizes the new excitation as a $\nu = 1/3$ ferromagnet eigenmode. The mode energy exceeds drastically the theoretical prediction obtained within the renowned single-mode approximation. We develop a new theoretical approach where the basis set is extended by adding a double-exciton component representing the cyclotron magnetoplasmon and spin wave coupled together. This double-mode approximation, inferred to be responsible for substantially reducing the gap between theoretical and experimental results, shows that the cyclotron spin-flip excitation is effectively a four-particle state.

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The physics of two-dimensional (2D) electron systems in a strong perpendicular magnetic field is governed by the macroscopic degeneracy of electron states in Landau levels. Owing to this and because of the strong many-particle interaction, the ground state at the unit filling of the zeroth Landau level (LL) is an itinerant ferromagnet where electron spins tend to align even for arbitrarily small Zeeman coupling. The ground state at fractional filling $\nu = 1/3$ is believed to be somewhat analogous. In fact, the magnetization around both $\nu = 1/3$ and $\nu = 1$ behaves similarly. It reaches pronounced maxima at those particular filling factors [1,2], which is generally interpreted in favor of the formation of quantum Hall ferromagnets.

A natural way to characterize a ferromagnetic state is to study its spin-flip excitations and their modification under the influence of external parameters (magnetic field, temperature, etc.). Most relevant for the $\nu = 1$ quantum Hall ferromagnet are the spin exciton (spin waves) and the cyclotron spin-flip excitation (CSFE) modes [3,4]. Experimentally accessible long-wavelength spin excitons carry little information about the ferromagnetic order by virtue of the Larmor theorem [5]. Major efforts have therefore been concentrated hitherto around the CSFE—a long-wavelength excitation simultaneously changing the orbital and spin quantum numbers of a 2D electron gas ($\delta n = 1$ and $\delta S = \delta S_z = -1$). Studies of the CSFE by means of the inelastic light scattering spectroscopy revealed peculiarities of the Coulomb interaction in two dimensions, disclosed new magnetic phases, and helped to draw qualitative conclusions about thermodynamics of the quantum Hall ferromagnet [6]. Surprisingly, similar experiments at $\nu = 1/3$ did not produce such clear results. The cyclotron spin-flip mode was detected, yet its energy reflected the total electron density in the 2D electron gas (2DEG) rather than a variation of the electron magnetization [7]. On the other hand, if the electron magnetization has a pronounced maximum at $\nu = 1/3$, so should the CSFE energy as a measure of the ground state exchange interaction. The apparent contradiction between naive expectations and the experiment is resolved in the present work, where we report on a new cyclotron spin-flip mode whose energy variation matches the electron magnetization. Furthermore, its energy substantially exceeds the existing theoretical estimates based on the single-mode approximation (SMA) [8,9]. This is quite unusual since no experimental indications questioning the validity of the single-mode approximation [10] have ever been detected. We stress here that the SMA is not applicable to the description of complex excitations like the CSFE mode of the "soft" $\nu = 1/3$ ferromagnet [11] and emphasize the importance of double-exciton contribution to the CSFE structure.

Two high-quality heterostructures were studied. Each consisted of a single-side doped 25 nm GaAs/$Al_{0.3}Ga_{0.7}As$ quantum well with electron densities of 1.2 and 2.2×10^{11} cm^{-2} in the dark. The mobilities were 7 and 10×10^6 cm^2/V $\cdot$ s. The electron densities were tuned via the optodepletion effect and were measured by means of *in situ* photoluminescence [12]. The inelastic light scattering experiment was performed in the temperature range of 0.3–1.5 K in the magnetic field up to 14 T normal to the sample surface. Inelastic light scattering spectra were obtained using a Ti:sapphire laser tunable above the fundamental band gap of the quantum well. The laser excitation energy was varied between 1.545 and 1.575 eV. The power density was below 0.1 W/cm^2. A two-fiber optical system was employed in the experiments [12]. One fiber transmitted the pumping laser beam to the sample, the second fiber collected the scattered light and guided it out of the cryostat. The scattered light was dispersed either by a T-64000 triple spectrograph or a Ramanor U-1000 double-grating monochromator and recorded with a charge-coupled device camera. The spectral resolution of the overall detection system was about 0.03 meV.

Figure 1 presents the key experimental result. Inelastic light scattering lines corresponding to two cyclotron spin-flip modes are clearly seen above the cyclotron energy. One (A) coincides with the inelastic light scattering line found earlier in terms of energy and linewidth [7]. The energy of the line A increases monotonically with the electron filling factor not showing any peculiarities around $\nu = 1/3$. In contrast to this, the second line (B) detected at rather strict resonant conditions demonstrates a nonmonotonic energy dependence with a pronounced maximum at $\nu = 1/3$. Moreover, the energy of line B is proportional to the 2DEG magnetization measured by Khandelwal *et al.* under similar experimental conditions [2]. This links the line B to the inelastic light scattering on a spin-flip mode of the $\nu = 1/3$ quantum Hall ferromagnet. Yet, the question arises of how two cyclotron spin-flip modes with so different properties could possibly exist in a 2DEG.

Several aspects of this problem have already been clarified in our previous publications. It was shown [6] that under certain experimental conditions a 2D electron system in the vicinity of a positive charge is unstable against formation of a spin-singlet barrier D^- complex—two electrons in a quantum well with oppositely aligned spins bound to an impurity in the quantum well barrier. In the quantum limit the barrier D^- complexes occupy a significant part of the experimentally accessible 2DEG even in the highest quality AlGaAs/GaAs structures. Because of two electron subsystems (D^- complexes and unbound electrons), two spin-flip modes exist. The line A arises from the upper branch of spin-flip excitations of barrier D^- complexes (see the inset of Fig. 1), whereas the line B originates from the free electron gas. Note that the two subsystems are not really independent. The electrons of D^- complexes interact with the unbound electrons; i.e., they are in fact many-particle conglomerates which become truly isolated only in the extreme quantum limit $\nu < 1/10$ [7]. In other words, the D^- complexes supply a degree of freedom to form an additional spin-flip mode, whose energy is not sensitive to electron magnetization. We do not consider the line A carefully studied in Ref. [7] and focus on the line B as an eigen-CSFE mode of the quantum Hall ferromagnet.

Though the CSFE energy behaves alike around $\nu = 1/3$ and $\nu = 1$ (Fig. 2), the CSFE nature at $\nu = 1/3$ is enigmatical since the CSFE energy reaches a much larger value at $\nu = 1/3$ than available calculations could predict. The electron gas in both cases is highly correlated, but the

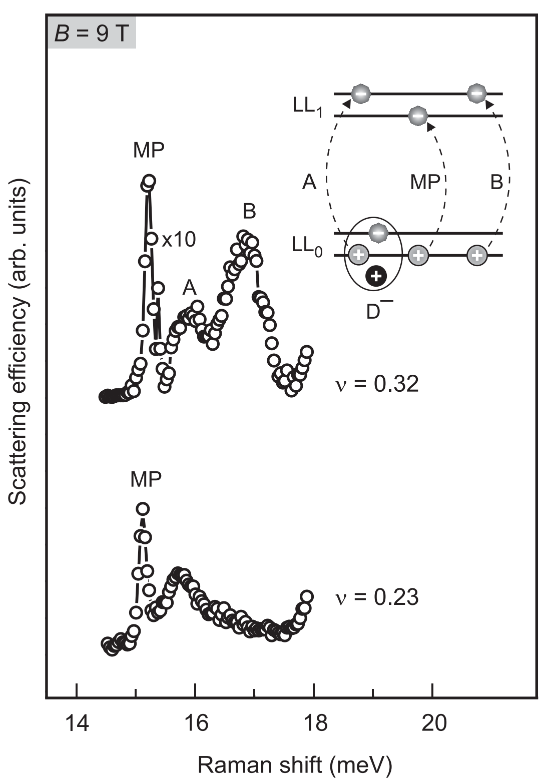


FIG. 1. Inelastic light scattering spectra at $B = 9$ T and two different filling factors, $\nu = 0.33$ and $\nu = 0.23$. Line B is the cyclotron spin-flip excitation of the $\nu = 1/3$ quantum Hall ferromagnet. Line A corresponds to the analogous excitation of D^- complexes. Line MP indicates the magnetoplasma mode. At filling factors far from $\nu = 1/3$, line B disappears from the spectrum (see the lower spectrum). The inset schematically explains the origin of the observed lines.

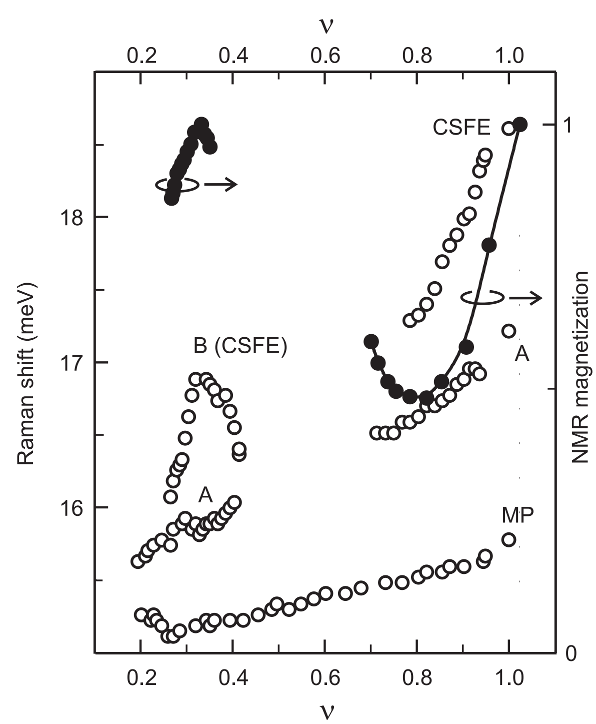


FIG. 2. Experimental ν dependence of the energies for lines A and B around $\nu = 1/3$. Also shown are the magnetoplasmon (MP) energy and energies for the corresponding lines in the vicinity of unit filling (at $\nu < 1$). Black points show the normalized electron magnetization measured with optically detected magnetic nuclear resonance in Refs. [1,2] under experimental conditions similar to ours.

$\nu = 1/3$ system still possesses a larger number of degrees of freedom than the $\nu = 1$ ferromagnet. To shed light on the interplay of diverse modes forming the CSFE in the fractional quantum Hall regime, the experimental CSFE energy is compared with existing theories, and a new theoretical approach to calculation of the CSFE energy is applied.

We note that even in the $\nu = 1$ ferromagnet the CSFE is not a single-mode (single-exciton) excitation but has a double-exciton component consisting of a spin wave (involving nonconservation of quantum numbers: $\delta S = \delta S_z = -1$ and $\delta n = 0$) and a magnetoplasmon (with $\delta S = \delta S_z = 0$ and $\delta n = 1$) [13]. Meanwhile, we first discuss the renowned SMA where the double-exciton component is ignored [9]. If the Hartree-Fock (HF) approximation is used to describe the ground state, the SMA result for the CSFE energy is easily generalized to arbitrary fillings $\nu < 1$, and at zero momentum it reads

$$\mathcal{E}_{\rm SM}^{\rm HF} = \nu \frac{e^2}{2\kappa l_B} \int_0^\infty p^3 dp V(p) e^{-p^2/2} \tag{1}$$

(this energy is counted from the cyclotron energy $\hbar\omega_c$) [4]. Here l_B is the magnetic length and $2\pi V(q)$ is the dimensionless Fourier component of the effective e-e interaction vertex in the 2D layer. [In the ideal 2D case $V(q) = 1/q$, but actually $V(q) = F(q)/q$, where $F(q)$ is the geometric form factor; the ratio $r_c = (e^2\langle F\rangle/\kappa l_B)/\hbar\omega_c$, where $\langle F\rangle$ is the averaged value, is considered to be small.] Calculation with Eq. (1) employing the usual self-consistent procedure to find the form factor $F(q)$ [14] yields in our specific case ($B = 9$ T, $\nu = 1/3$) $\mathcal{E}_{\rm SM}^{\rm HF} = 1.26$ meV. This is rather far off the experimental value $E_{\rm exp} = 1.69$ meV observed in our case (see Fig. 2). A more refined approach utilizes Laughlin's wave function for the ground state to calculate the CSFE energy within the single-mode approximation [9]. This yields the value for the correlation shift of $\mathcal{E}_{\rm SM} = 0.79$ meV. Being even smaller than the Hartree-Fock result (1), it is in a striking disagreement with the experiment.

Another development of the existing theory should account for the multicomponent feature of CSFE. In a fractional quantum Hall ferromagnet, charge-density waves (intra-Landau-level excitations) are split from the ground state by a considerable energy gap [8]. Because of this, we will utilize a model where charge-density waves are ignored but the double-exciton component corresponding to coupled spin wave and magnetoplasmon is taken into account. The present approach is thus a projection of the $\nu = 1$ CSFE theory [13] onto the fractional ν case. Specifically, we employ the double-mode approximation (DMA). The CSFE state is then represented as

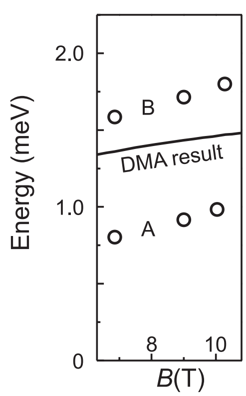
$$|{\rm SF}\rangle = \mathcal{Q}^\dagger_{0\bar{1}\mathbf{0}}|0\rangle + AN_\phi^{-1/2}\sum_{\mathbf{q}} \psi(q)\mathcal{Q}^\dagger_{0\bar{0}-\mathbf{q}}\mathcal{Q}^\dagger_{01\mathbf{q}}|0\rangle \tag{2}$$

(N_ϕ is the magnetic flux number in the 2D electron gas studied). The definition of the exciton creation operator $\mathcal{Q}^\dagger_{ab\mathbf{q}}$ in terms of electron annihilation and creation operators can be found, e.g., in Ref. [13] (see also commutation rules for the $\mathcal{Q}$ operators there and references therein). Here a and b are binary indexes labeling Landau levels and spin sublevels: $a = 0$ to denote the $(n, S_z) = (0, \uparrow)$ state, $b = \bar{0}$ is the $(0, \downarrow)$ state, $b = 1$ corresponds to $(n, S_z) = (1, \uparrow)$, and $b = \bar{1}$ is the $(1, \downarrow)$ state. In the $\nu \leq 1$ case, we have $\langle 0|\mathcal{Q}_{ab\mathbf{q}}\mathcal{Q}^\dagger_{ab\mathbf{q}'}|0\rangle = \nu\delta_{a,0}\delta_{\mathbf{q},\mathbf{q}'}$ if $b \neq 0$. We consider the ground state $|0\rangle$ in the HF approximation. Then one can also find that $\langle 0|\mathcal{Q}_{aa\mathbf{q}}\mathcal{Q}^\dagger_{aa\mathbf{q}'}|0\rangle = \nu\delta_{a,0}\delta_{\mathbf{q},\mathbf{q}'}(1 - \nu + \nu N_\phi\delta_{\mathbf{q},0})$.

Because of the same symmetry reasons as those given in Ref. [13], the "wave function" $\psi(q)$ in Eq. (2) should be presented as an expansion over orthogonal functions $L_m(q^2)e^{-q^2/2}$, where m is an odd number (L_m is the Laguerre polynomial). However, we also can limit ourselves to a single-term approximation by choosing $\psi(q)$ equal to $L_1(q^2)e^{-q^2/2}$. Then, considering A as a fitting parameter, we find the CSFE energy after a variational procedure. The result is again given by the largest root of a 2×2 secular equation. The latter takes the form

$$\det|(E - \mathcal{E}_i)\delta_{i,k} + (1 - \delta_{i,k})\mathcal{D}_{ik}| = 0 \qquad (i, k = 1 \text{ or } 2), \tag{3}$$

where $\mathcal{E}_1 = \int_0^\infty q dq V(q)[\nu\epsilon(q) + (1 - \nu)\varepsilon(q)]$, $\mathcal{E}_2 = \mathcal{E}_{\rm SM}^{\rm HF}$ [see Eq. (1)], and $\mathcal{D}_{12} \equiv \mathcal{D}_{21} = \sqrt{\nu}\int_0^\infty q dq V(q) d(q)$ with $\epsilon = 2q^2(1 - q^2)^2 e^{-3q^2/2} + \frac{1}{2}(4 - 5q^2 + q^4)e^{-q^2} + \frac{1}{16}(q^2 - 4)^3 e^{-3q^2/4} + (2 - q^2/2) \times e^{-q^2/2}$, $\varepsilon = \frac{1}{2}[(q^2 - q^4)e^{-q^2} - (q - q^3/4)^2 e^{-3q^2/4}]$, and $d = q^2(q^2 - 1)e^{-q^2}$. In the present experimental situation one obtains from Eq. (3) the DMA energy $E = \mathcal{E}_{\rm DM} = 1.43$ meV; see Fig. 3. The DMA result thus exceeds the SMA energy (1) and brings the theory and the experiment into rather closer agreement. Note that, although Eq. (3)



Ground state model \ Number of modes	SMA	DMA
Hartree-Fock wave function	Eq. (1) and Ref.[9]: **1.26 meV**	The present work calculation: **1.43 meV**
Laughlin's state	Ref.[9]: **0.79 meV**	?

FIG. 3. Left: Experimental magnetic field dependence of the energies for lines A and B (CSFE). The result of calculation within double-mode approximation is also shown as a solid line. Right: Table shows the comparison of CSFE energies calculated at $B = 9$ T and $\nu = 1/3$ within existing theoretical frameworks and for Hartree-Fock and Laughlin's ground states.

was formally derived for any $\nu < 1$, it becomes irrelevant when gapless charge-density waves can propagate in the electron system. The double-mode approximation evidently fails in this case. We expect that soft charge-density wave modes coupled to the CSFE exciton should effectively reduce the CSFE energy. Indeed, experimentally a striking reduction in CSFE energy is observed when ν is offset from the $\nu = 1/3$ value corresponding to the ferromagnet state.

To clarify the specific place of the double-mode approximation among other theoretical models, all available results for calculations of the $\nu = 1/3$ CSFE energy at $B = 9$ T are presented in the table (right-hand side of Fig. 3). Relevant references in this table are given in the cells located at the crossing of the appropriate column (number of modes considered) and row (the ground state descriptions). The cell with the question mark corresponds to the double-mode approximation where Laughlin's ground state would be used for the CSFE calculation. However, the analysis reveals that when the double-exciton component is taken into account and the problem effectively becomes a four-particle one, the result of the calculation (at variance with the single-mode approximation) cannot be expressed in terms of the two-particle correlation function [15] but can only be presented in terms of the three-particle correlation function. This function depending on three scalar arguments has actually never been calculated, and nothing is known about it at the present time.

To conclude, we briefly outline the general meaning of the reported results. We have employed the inelastic light scattering experimental technique which seems to be the only microscopic tool for a direct access to energies and momenta of collective excitations in a strongly correlated 2D electron system. This has enabled us to find a new CSFE mode unrelated to any translation symmetry imperfections. The CSFE is thus a purely electronic excitation. The CSFE energy has been measured in the range of filling factors close to 1/3 and shown to "feel" the degree of spin polarization having a pronounced maximum at $\nu = 1/3$. Although the CSFE was theoretically predicted a long time ago [3], before the work [13] it was only considered within the single-mode approximation [3,4,9] neglecting the double-exciton component [16]. We emphasize that the double-exciton component formally contributes to the CSFE on an equal footing with the single-exciton mode. The first-order CSFE energy ($\sim e^2\langle F\rangle/\kappa l_B$) is determined by both components, and the CSFE is effectively a four-particle state. Yet, until now, the difference between the double- and single-mode approximations turned out to be small numerically and undetectable experimentally (a fine example is the $\nu = 1$ ferromagnet, [6,13]). In this Letter we show that this difference becomes significant for the $\nu = 1/3$ quantum Hall state. Despite the recognized similarity for the $\nu = 1$ and $\nu = 1/3$ states, the latter represents evidently a "softer" type of quantum Hall ferromagnet. As a result, participation of the double-exciton component in the CSFE becomes more weighty. Its inclusion substantially improves the agreement between the experiment and the theory, indicating thereby a composite nature of the CSFE in the fractional quantum Hall regime.

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