

## Cyclotron spin-flip mode as the lowest-energy excitation of unpolarized integer quantum Hall states

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The cyclotron spin-flip modes of spin unpolarized integer quantum Hall states ( $\nu=2, 4, 6$ ) have been studied with inelastic light scattering. The energy of these modes is significantly smaller compared to the bare cyclotron gap. Second-order exchange corrections are held responsible for a negative energy contribution and render these modes the lowest-energy excitations of unpolarized integer quantum Hall states.

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According to Kohn's theorem,<sup>1</sup> homogenous electromagnetic radiation incident on a translationally invariant electron system can only couple to the center-of-mass coordinate. Such radiation is unable to excite internal degrees of freedom associated with the Coulomb interaction. As a result, physical phenomena originating from electron-electron interactions leave the cyclotron resonance unaffected. Hence, spin-unperturbed magnetoplasmons excited under these conditions have an energy equal to the bare cyclotron energy, irrespective of existing electron-electron correlations.<sup>2</sup> A similar statement also holds for spin excitons, intra-Landau level spin-flip excitations. In a system with rotational invariance in spin space, Larmor's theorem<sup>3</sup> dictates that Coulomb interactions do not contribute to the energy of zero-momentum spin excitons. In contrast to these magnetoplasma and spin-exciton excitations, there exist no symmetry arguments which restrict the energy of the combined zero-momentum cyclotron spin-flip mode (CSFM). It is well established that the cyclotron-spin-flip mode excited from spin-polarized ground states acquires considerable exchange energy even for zero momentum.<sup>4-6</sup> The energy of this mode may thus serve as a unique probe of many-body interactions in the electronic system.

Hitherto, it has not been considered that there is also an exchange contribution to the energy of the zero-momentum cyclotron spin-flip modes of unpolarized quantum Hall ground states at even integer fillings ( $\nu=2, 4, 6, \dots$ ). First-order perturbation calculations in the ratio  $r_c=E_C/\hbar\omega_c$  explicitly predicted a zero exchange contribution to the total energy of this combined mode of unpolarized quantum Hall ground states<sup>2</sup> ( $E_C$  is the characteristic Coulomb energy scale and  $\hbar\omega_c$  the cyclotron energy). Here, we experimentally demonstrate, however, that the energy of these modes is considerably reduced compared with the bare cyclotron gap. We corroborate with theoretical considerations that the negative energy contribution arises from *second-order Coulomb corrections* and so was not captured by previous first-order perturbation calculations.

Two high-quality heterostructures were studied. Each consisted of a single-side modulation-doped 30-nm

AlGaAs/GaAs quantum well (QW) with an electron density between 1 and  $1.2 \times 10^{11} \text{ cm}^{-2}$  and a mobility of  $5-7 \times 10^6 \text{ cm}^2/(\text{Vs})$ . The density  $n_s$  was tuned continuously via the optodepletion effect and was measured with luminescence.<sup>7</sup> Inelastic light-scattering (ILS) spectra were recorded at 1.5 K in the backscattering geometry in a split-coil cryostat. Three optical fibers were utilized. One fiber transmitted a dye laser pump beam, tuned above the fundamental gap of the QW. The remaining fibers collected the scattered light and guided it out of the cryostat. The angles between the sample surface, pump beam fiber, and collecting fibers define the in-plane momentum transferred to the electron system via inelastic light scattering. The collecting fibers selected excitations with in-plane momenta of 0.4 and  $1.0 \times 10^5 \text{ cm}^{-1}$ . The scattered light was dispersed in a triple grating monochromator and detected with a charge-coupled device (CCD) camera.

Figure 1 shows typical ILS spectra of inter-Landau-level (LL) excitations as well as the magnetic-field ( $B$ ) depen-

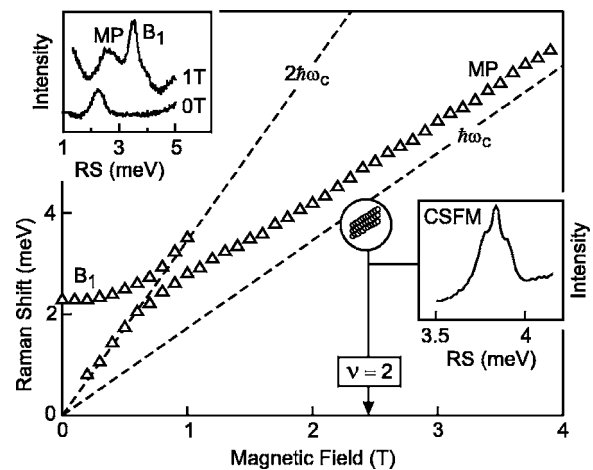


FIG. 1.  $B$  dependence of ILS line energies. Triangles mark charge-density excitations, circles the CSFM. The dashed lines correspond to  $\hbar\omega_c$  and  $2\hbar\omega_c$ . Top, left inset: ILS spectra at  $B=0$  and 1 T. Bottom, right inset: ILS spectrum at 2.4 T.

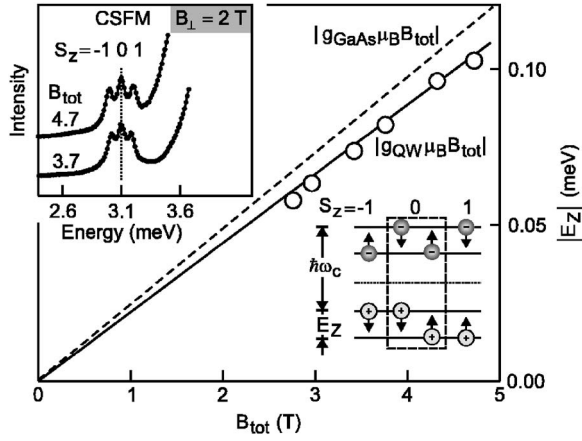


FIG. 2. Electron Zeeman energy  $E_Z$ . The dashed line plots the expected Zeeman energy when taking  $g_{\text{GaAs}} = -0.44$ . The solid line is a linear fit to the data for  $g_{\text{QW}} = -0.4$ . The inset depicts ILS spectra at a constant perpendicular field of 2 T but two different values of the total field. The right inset schematically illustrates the spin-triplet ( $S=1$ ,  $S_z = -1, 0$  and  $1$ ) cyclotron excitations for  $\nu=2$ .

dence of the energy of the various lines in these spectra in a sample with a density of  $1.2 \times 10^{11} \text{ cm}^{-2}$ . The experimental configuration selected excitations with an in-plane momentum of  $q = 1.0 \times 10^5 \text{ cm}^{-1}$ . The polarization selection rules allowed to identify that lines at low  $B$  ( $< 1$  T) correspond to charge-density excitations. The principal magnetoplasmon mode as well as a Bernstein mode ( $B_1$ ) are observed in the geometry where the incident and scattered photons have parallel polarization vectors.<sup>8</sup> At nonzero  $B$  the magnetoplasmon mode has a strong linear dispersion in the long-wavelength limit and at  $B=0$  its energy equals the plasma energy for momentum  $q$ . In contrast, the Bernstein mode is nearly dispersionless. Both modes couple through many-body Coulomb interactions near  $\sim 0.8$  T. At large  $B$ , their energies converge asymptotically to the cyclotron energy and twice the cyclotron energy, respectively.<sup>9</sup>

Of main interest here is the appearance of a triplet ILS resonance when the system is in the  $\nu=2$  spin unpolarized quantum Hall state (bottom right inset of Fig. 1). Near  $B = 2.4$  T, the central line of the triplet is clearly resolved, but the side lines only appear as shoulders. The splitting between the features corresponds approximately to the electron Zeeman energy  $E_Z$  in GaAs, so they are attributed to the three cyclotron spin-flip modes with different spin projections along the  $B$ -field axis ( $S_z = -1, 0$ , and  $1$ ). The shoulder structures are assigned to the cyclotron spin-flip modes with  $S_z = -1$  and  $1$ , and the central line ( $S_z = 0$ ) is associated with a cyclotron spin wave, i.e., out-of-phase oscillations of the two spin subsystems of the Landau levels with orbital index 0 and 1 (Ref. 10). This identification of the triplet is confirmed by measurements in tilted fields. ILS spectra in tilted fields are plotted in the left inset of Fig. 2. The triplets are much better resolved due to the larger total fields  $B_{\text{tot}}$ . Well-separated peaks appear and the spin splitting can be directly measured. The Zeeman effect is in essence a three-dimensional phenomenon and so energy gaps between the ILS triplet lines are proportional to  $B_{\text{tot}}$  rather than the per-

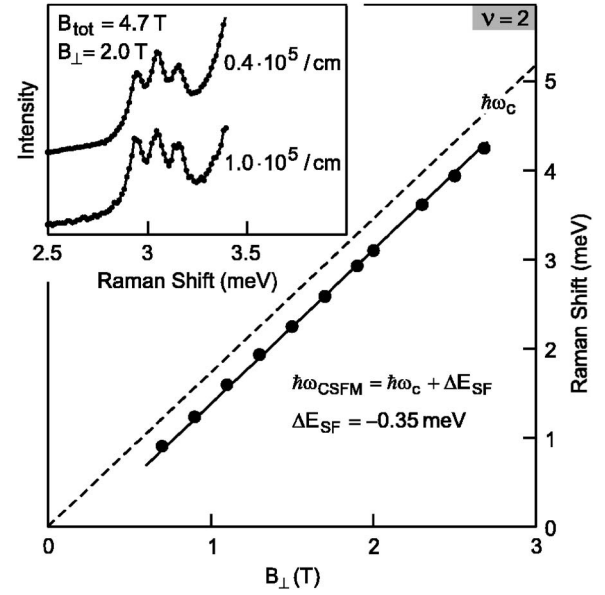


FIG. 3. The CSFM energy for the spin unpolarized  $\nu=2$  quantum Hall state vs perpendicular field. The dashed line gives the cyclotron energy. The upper inset displays ILS spectra of the CSFM for two different in-plane momenta.

pendicular component  $B_{\perp}$ . The position of the central line, however, only depends on  $B_{\perp}$  (left inset of Fig. 2). The main plot in Fig. 2 presents the measured electron Zeeman energy (open circles) as a function of  $B_{\text{tot}}$ . The data points fit well to a  $g$  factor  $g_{\text{QW}} = -0.4$  (solid line). The dashed line corresponds to  $|g_{\text{GaAs}} \mu_B B|$ , where  $g_{\text{GaAs}} = -0.44$  is the effective  $g$  factor of bulk GaAs. A significant reduction of the  $g$  factor is not uncommon in AlGaAs heterostructures and has been accounted for by band-structure nonparabolicity, confinement, and wave-function penetration effects.<sup>11</sup>

In the inset of Fig. 3 we compare ILS spectra measured at  $\nu=2$  for two different values of in-plane momenta:  $0.4$  and  $1.0 \times 10^5 \text{ cm}^{-1}$ . In agreement with existing theories,<sup>2,12</sup> the CSFM energy does not show any appreciable dispersion at momentum values accessible with ILS techniques. Therefore, the CSFM line is regarded as the energy of the cyclotron spin-flip mode when  $q \rightarrow 0$ . The key experimental finding is a downward shift of the energy of this mode with  $-0.35$  meV as compared to the bare cyclotron energy. This shift exceeds by far the single-electron Zeeman energy in GaAs at this magnetic field (0.08 meV) and we therefore assert it is strongly influenced by exchange interactions.

The  $B$  dependence of the energy of the cyclotron spin-flip mode for fixed filling  $\nu=2$  is plotted in Fig. 3. The slope is identical to the bare cyclotron energy line in GaAs. Hence, the dependence of the cyclotron spin-flip mode on  $B$  takes on the functional form  $\hbar\omega_{\text{CSFM}} = \hbar\omega_c + \Delta E_{\text{SF}}$  over a rather broad magnetic-field interval:  $0.6 \text{ T} < B < 2.7 \text{ T}$ . Here  $\Delta E_{\text{SF}}$  is the  $B$ -independent downward shift of approximately  $-0.35$  meV. It is worthwhile to note that a dimensional analysis of second-order Coulomb corrections to the energies of inter-LL excitations would yield a similar dependence on  $B$ :  $E = \hbar\omega_c + \Delta E_{\text{SF}}$ , where  $\Delta E_{\text{SF}} \sim \hbar\omega_c r_c^2$ . Indeed, if  $E_C = \alpha e^2 / \epsilon l_B$  then  $\Delta E_{\text{SF}}$  is independent of the field. The renormalization

factor  $\alpha$  is determined by the size-quantized wave function of electrons confined in the QW. In the ideal two-dimensional (2D) case  $\alpha=1$ . However, the larger the width of the 2D electron system (2DES), the smaller  $\alpha$  becomes, thereby reducing  $r_c$ . This is certainly relevant for the width of our quantum well.

An *analytical* calculation of the second-order correction to the CSFM energy is performed in terms of small  $r_c$ . The theory is based on the following general features of the system. The state of the system is described by the exact quantum numbers  $S$ ,  $S_z$ , and  $\mathbf{q}$  and by the “good” quantum number  $\delta n$  characterizing the excitation kinetic energy  $\hbar\omega_c\delta n$  ( $\delta n$  is good but not exact due to LL mixing). The relevant excitations with  $\mathbf{q}=0$  and  $\delta n=1$  may be presented in the form  $\hat{K}_{S,S_z}^\dagger|\mathbf{0}\rangle$ , where  $|\mathbf{0}\rangle$  is the ground state and  $\hat{K}_{S,S_z}^\dagger$  are “raising” operators:  $\hat{K}_{0,0}^\dagger=\sum_{np\sigma}\sqrt{n+1}c_{n+1,p,\sigma}^\dagger c_{n,p,\sigma}$ ,  $\hat{K}_{1,0}^\dagger=\sum_{np\sigma}\sqrt{n+1}\times(-1)^\sigma c_{n+1,p,\sigma}^\dagger c_{n,p,\sigma}$ , and  $\hat{K}_{1,\pm 1}^\dagger=\sum_{np}\sqrt{n+1}c_{n+1,p,\uparrow/\downarrow}^\dagger c_{n,p,\downarrow/\uparrow}$  [ $c_{n,p,\sigma}$  is the Fermi annihilation operator corresponding to the Landau-gauge state  $(n,p)$  with spin index  $\sigma=\uparrow,\downarrow$ ]. The commutators with the kinetic-energy operator  $\hat{H}_1$  are  $[\hat{H}_1,\hat{K}_{S,S_z}^\dagger]\equiv\hbar\omega_c\hat{K}_{S,S_z}^\dagger$ . The total Hamiltonian is  $\hat{H}_{\text{tot}}=\hat{H}_1+\hat{H}_{\text{int}}$ , where  $\hat{H}_{\text{int}}$  is the exact Coulomb-interaction Hamiltonian. If  $|\mathbf{0}\rangle$  is unpolarized, we have  $\hat{S}_z^2\hat{K}_{S,S_z}^\dagger|\mathbf{0}\rangle\equiv S(S+1)\hat{K}_{S,S_z}^\dagger|\mathbf{0}\rangle$  and  $\hat{S}_z\hat{K}_{S,S_z}^\dagger|\mathbf{0}\rangle\equiv S_z\hat{K}_{S,S_z}^\dagger|\mathbf{0}\rangle$ . Moreover, the identity  $\langle 0|\hat{K}_{S,S_z}^\dagger[\hat{H}_{\text{int}},\hat{K}_{S,S_z}^\dagger]|0\rangle\equiv 0$  holds ( $|0\rangle$  to describe the zeroth order ground state). It implies that first-order Coulomb corrections vanish for both the spin-unperturbed or singlet magnetoplasmon (where  $S=0$ ) and the combined CSFM triplet ( $S=1$ ). At the same time,  $[\hat{H}_{\text{int}},\hat{K}_{0,0}^\dagger]\equiv 0^1$  but  $[\hat{H}_{\text{int}},\hat{K}_{1,S_z}^\dagger]\neq 0$ . Hence, whereas the magnetoplasmon has no exchange energy correction calculated to *any order* in  $r_c$ , the combined modes should have second- and higher-order exchange corrections.

The second-order calculation is based on the excitonic representation (ER) technique.<sup>13–15</sup> It utilizes exciton states  $\hat{Q}_{ab\mathbf{q}}^\dagger|\mathbf{0}\rangle$  as a basis set, instead of single-electron states of a degenerated LL. The exciton creation operator is defined as<sup>13–15</sup>

$$\hat{Q}_{ab\mathbf{q}}^\dagger = N_\phi^{-1/2} \sum_p e^{-iq_x p} b_{p+q_y/2}^\dagger a_{p-q_y/2}. \quad (1)$$

Here,  $N_\phi=A/2\pi l_B^2$  stands for the number of magnetic flux quanta and  $\mathbf{q}=(q_x,q_y)$  is given in units of  $1/l_B$ . The binary indices  $a$  and  $b$  denote both the LL number and the spin index:  $a,b=(n_{a,b},\sigma_{a,b})$ . All three CSFM states have certainly the same exchange energy, and it is sufficient to calculate this, e.g., for the state with  $S_z=-1$ . The zero-order approximation is thereby  $|SF,-\rangle=N_\phi^{-1/2}\hat{K}_{1,-}^\dagger|\mathbf{0}\rangle_{r_c=0}=\hat{Q}_{010}^\dagger|\mathbf{0}\rangle$  [i.e.,  $a=(0,\uparrow)$  and  $b=(1,\downarrow)$ ]. To calculate the first-order corrections to the  $|SF,-\rangle$  state or, equivalently, the second-order correction to its energy, we follow the standard perturbative approach<sup>16</sup> using the “excitonically nondiagonalized” part  $\hat{\mathcal{H}}_{\text{int}}$  of the Coulomb Hamiltonian<sup>14</sup> in the ER form,

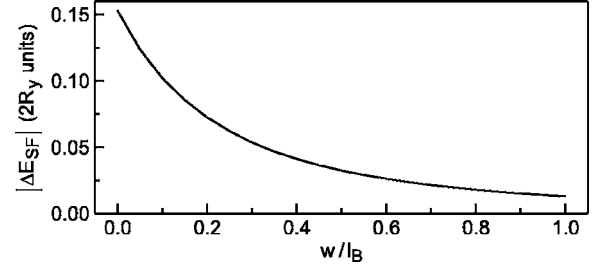


FIG. 4. The CSFM exchange shift calculated from Eq. (3) with the modified Coulomb interaction  $V(q)=q^{-1}e^{q^2 w^2} \text{erfc}(qw)$ . Its absolute value at  $w=0$  equals  $(1-\ln 2)\text{Ry}$ .

$$\hat{\mathcal{H}}_{\text{int}} = \frac{e^2}{2\epsilon l_B} \sum_{\mathbf{q},a,b,c,d} V(\mathbf{q}) [h_{n_a n_b}(\mathbf{q}) \delta_{\sigma_a \sigma_b} \hat{Q}_{ab\mathbf{q}}^\dagger] \times [h_{n_c n_d}(-\mathbf{q}) \delta_{\sigma_c \sigma_d} \hat{Q}_{cd-\mathbf{q}}^\dagger] \quad (2)$$

(cf. Ref. 14), where  $2\pi V(\mathbf{q})$  is the Fourier component of the dimensionless Coulomb potential (in the strict 2D limit  $V=1/q$ ), and  $h_{kn}(\mathbf{q})=(k!/n!)^{1/2}e^{-q^2/4}(q_-)^{n-k}L_k^{n-k}(q^2/2)$  are the ER “building-block” functions ( $L_k^n$  is the Laguerre polynomial,  $q_\pm = \mp(i/\sqrt{2})(q_x \pm iq_y)$ ; cf. Refs. 2, 14, and 15). For calculation details, we refer the reader to Ref. 17. Here, we limit ourselves to reporting the final result,

$$\Delta E_{\text{SF}} = - \sum_{n=2}^{\infty} R_n \frac{1-2^{1-n}}{n(n^2-1)}, \quad (3)$$

with

$$R_n = \frac{2}{n!} \int_0^\infty dq q^{2n+3} V^2(q) e^{-q^2},$$

in units of  $2\text{Ry}=(e^2/\epsilon l_B)^2/\hbar\omega_c \approx 11.34\text{meV}$ . For the ideal 2D system with zero width  $R_n \equiv 1$  and the summation can easily be performed. It yields  $\Delta E_{\text{SF}}=(\ln 2-1)/2 = -0.1534, \dots$

We conclude that, as in experiment, the exchange interaction lowers the energy of the CSFM relative to the singlet magnetoplasmon mode. The absolute value of the shift  $|\Delta E_{\text{SF}}|$  obtained with Eq. (3) is reduced when taking into account the nonzero thickness of the 2DES. The Coulomb vertex should be written as  $V(q)=F(qw)/q$ , where  $F(qw)$  is a form factor capturing the Coulomb softening.<sup>18</sup> The effective thickness parameter  $w$  characterizes the spread of the electron wave function in the growth direction. If a variational wave function of the form  $|\psi(z)|^2 \sim \exp(-z^2/2w^2)$  is chosen, then  $F(qw)=e^{w^2 q^2} \text{erfc}(qw)$  (Ref. 19). Note that for a second-order energy correction this form factor enters twice. The calculation of  $|\Delta E_{\text{SF}}|$ , including the influence of finite thickness, is plotted in Fig. 4. A similar value for  $|\Delta E_{\text{SF}}|$  as in experiment is obtained when  $w \approx 0.5l_B$ , which agrees well with the effective width for a 30-nm GaAs QW structure.

We note that inelastic light-scattering studies at  $\nu=2$  were carried out previously in Ref. 20. The authors obtained similar spectra with a nonzero energy shift, but explained their observations in terms of transitions to the roton minimum at

large wave vector. This assignment was plausible as it was based on information from Ref. 2 in which a nonzero energy shift was predicted only for the roton minimum but not for  $q=0$  due to the first-order approximation in the interaction. The authors were forced to invoke disorder to account for the large momentum transfer required for scattering into the roton minimum. The energy shift was reported to fit to a square-root dependence over the investigated  $B$ -field range as anticipated for rotons. In contrast, we assign the spectra in our experiments to the properties of the cyclotron spin-flip mode at  $q=0$ . If we were to ascribe the signals to indirect inelastic light scattering into the roton minimum, we would expect a second much larger direct resonance at  $q=0$  since the density of states is large for both the  $q=0$  extremum and the roton minimum. Moreover, we find a field-independent energy shift  $\Delta E_{SF}$  over a large  $B$  range. Well-resolved triplet spectra and negative exchange energy shifts are not only obtained at  $\nu=2$ , but also at filling  $\nu=4$  and  $\nu=6$ . Figure 5 illustrates, for instance, ILS spectra measured at  $\nu=4$ . Note that the negative exchange energy contribution at  $\nu=4$  is only half of the value at  $\nu=2$  due to the larger spatial extent of the wave functions of exciton states of higher Landau levels. The observation of well-resolved triplet modes at  $\nu=4$  and  $\nu=6$  excludes an interpretation of our data at momenta of the roton minimum. In this case, two and three roton minima would appear for  $\nu=4$  and  $\nu=6$ . This would result in significant broadening and the triplet structure would be smeared out.

In conclusion, the inelastic light-scattering response from

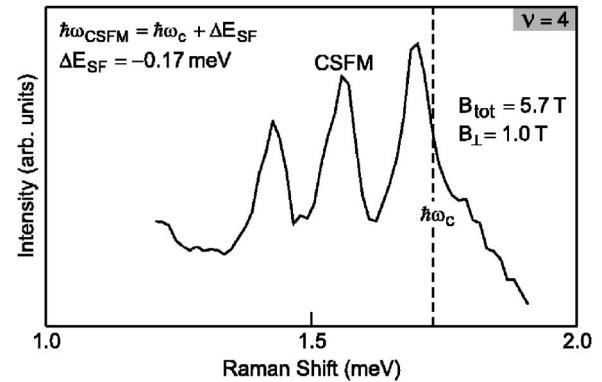


FIG. 5. ILS spectrum of the cyclotron spin-flip mode at the  $\nu=4$  unpolarized quantum Hall state at the indicated values for  $B_{\perp}$  and  $B_{\text{tot}}$ .

the combined cyclotron spin-flip modes of unpolarized quantum Hall states at  $q=0$  has been studied. A negative energy term was found to decrease their energy and was attributed to many-body Coulomb exchange interaction. A second-order perturbation theory of the Coulomb interaction explains the experimental results.

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