Extremely slow spin relaxation in a spin-unpolarized quantum Hall system

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Cyclotron spin-flip excitation in a $\nu = 2$ quantum Hall system, being separated from the ground state by a slightly smaller gap than the cyclotron energy and from upper magnetoplasma excitation by the Coulomb gap $\Delta\nu$, cannot relax in a purely electronic way but only with the emission of a short-wave acoustic phonon ($k \sim 3 \times 10^7 / \text{cm}$). As a result, relaxation in a modern wide-thickness quantum well occurs very slowly. We calculate the characteristic relaxation time to be $\sim 1$ s. Extremely slow relaxation should allow the production of a considerable density of zero-momenta cyclotron spin-flip excitations in a very small phase volume, thus forming a highly coherent ensemble of the Bose-Einstein condensate. The condensate state can be controlled by short optical pulses ($\lesssim 1 \mu$s), switching it on and off.

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Particular interest in the problem of excitation life-times in low-dimensional electron systems gained popularity in the mid-1990s, when first ideas of possible physical realizations of quantum computations were proposed. Indeed, it is known that one of the requirements (necessary though insufficient!) is the long lifetime of an excited quantum state in a two-level system, considered as a qubit. Most of those works were devoted to zero-dimensional objects – the electron relaxation in quantum dots (see [3] and references therein) or to a one-dimensional system – quantum Hall edge electrons [4]. The relaxation in strongly correlated two-dimensional systems was studied mainly in particular cases of spin relaxation in a spin-polarized quantum Hall system (i.e. in a quantum Hall ferromagnet [5, 6]).

In this letter we report on the relaxation rate calculation of the cyclotron spin-flip exciton (CSFE) in an unpolarized even-integer quantum Hall system, where an electron is effectively promoted from the fully occupied Landau level to the next fully empty level with a spin flip [1, 2] (at which point an effective hole appears in the initial level, see Fig. 1). The CSFE with spin numbers $S = S_z = 1$ is the lowest energy excitation in the system (Fig. 1). Yet it is separated from the ground state by a wide gap, which is only somewhat smaller than the cyclotron one. We will see that due to a concatenation of circumstances – a specific combination of certain CSFE features, the relaxation rate should be very slow. One of the features is the optical inactivity of the CSFE, which takes place even in the presence of spin-orbit interaction [7]. Hence the relaxation could only occur non-radiatively due to small perturbations violating the spin and energy conservations: spin-orbit and electron-phonon couplings. We estimate that the CSFE life-time should be very slow. One of the possible physical realizations of quantum computations was proposed. Indeed, it is known that one of the requirements (necessary though insufficient!) is the long lifetime of an excited quantum state in a two-level system, considered as a qubit. Most of those works were devoted to zero-dimensional objects – the electron relaxation in quantum dots (see [3] and references therein) or to a one-dimensional system – quantum Hall edge electrons [4]. The relaxation in strongly correlated two-dimensional systems was studied mainly in particular cases of spin relaxation in a spin-polarized quantum Hall system (i.e. in a quantum Hall ferromagnet [5, 6]).

Excited state — The CSFE spectrum in the $\nu = 2$ case, studied earlier both theoretically [1, 3] and experimentally [2], represents a triplet with the $S = 1$ and $S_z = 0, \pm 1$ spin components separated by the Zeeman gap. By analogy with previous works devoted to the calculation of excitations spectra in purely electronic quantum Hall systems (e.g. see [1]) as well as to excitation relaxation [3], we employ the excitonic representation technique, where exciton states are used as a basis set instead of single-electron Fermi states. The exciton states are generated by exciton creation operators [1, 2, 4, 10],

$$Q_{ab}^\dagger = \frac{1}{\sqrt{N_e}} \sum_p e^{-i q \cdot \mathbf{p}} a_p^\dagger b_p^\dagger e^{-i \phi_p} a_{p + q}^\dagger a_p - s q, \quad (1)$$

acting on the ground state $|0\rangle$. Here $a_p$ and $b_p$ are electron Fermi annihilation operators corresponding to the “initial” and “final” states of the promoted electron, and $q$ is the exciton dimensionless wave vector. Index $p$ labels intrinsic Landau level states.
\[\psi_{np}(r) = (2\pi N_0)^{-1/4}e^{ip\cdot\mathbf{r}}\varphi_n(p+\mathbf{z}),\]
where \(\varphi_n(x)\) is the oscillator wave function, \(n\) is the Landau level number, and \(N_0\) is the number of magnetic flux quanta (we measure length in units of the magnetic length \(l_B\)). \(a\) and \(b\) are binary indexes indicating both the Landau level number and the spin state. For example, for the \(S_z = 1\) component of the CSFE at \(\nu = 2\), these are \(a = (0, \downarrow) \equiv \mathbf{0}\) and \(b = (1, \uparrow) \equiv 1\), see Fig. 1 \((n\) and \(\varpi\) are spin-up and spin-down sublevels).

The set of exciton operators \((1)\) with arbitrary indexes \(a\) and \(b\) obey a closed Lie algebra. The basic property of the exciton states, \(Q_{ab}\mathbf{q}|0\rangle\) consists in the fact that they diagonalize a considerable part in the exact many-electron Coulomb interaction Hamiltonian \(H_{\text{int}}\). At \(\nu = 2\) the excitonic representation of the \(\mathbf{q} = 0\) CSFE state, \(Q_{11}\mathbf{q}|0\rangle\), allows the determination of first order corrections to the state in terms of the interaction \(H_{\text{int}}\) and thus to calculate the CSFE energy perturbatively up to the second order in terms of \(H_{\text{int}}\). Precisely this second-order result yields in this case the leading contribution to the excitation Coulomb energy \(1\).

On the basis of the results \([1, 2, 6]\) one can conclude that in the leading approximation in small parameters \(q\) and \(r_s = \alpha(e^2/\kappa l_B)/\hbar\omega_c\) (\(\kappa\) is the GaAs dielectric constant, and \(\alpha < 1\) is the averaged form-factor arising due to the finiteness of the 2D electron-layer thickness) the CSFE spectrum is

\[E(S_z, q) = \hbar \omega_c - c_2 S_z - \Delta E_C + q^2/2M_s,\]
where the negative value \(\Delta E_C\) is the Coulomb shift calculated to the second order in \(r_s\) and therefore expressed in units of \(2\text{Ry} = (e^2/\kappa l_B)^2/\hbar\omega_c = m^*e^4/\kappa l_B^2\) \([1]\), whereas \(1/M_s\) is positive \([6]\). \(\Delta E_C\) is definitely a positive value \([1]\), whereas the sign of \(1/M_s = (e^2/\kappa l_B)^2\int_0^\infty dF(p)p e^{-p^2/2}(p^2/2 - p^2/4)\) varies with a finite thickness form-factor \(F(q) = \iint dz_1 dz_2 e^{-q_1 z_1 - q_2 z_2/4} |\chi(z_1)|^2 |\chi(z_2)|^2\) (\(\chi\) is the size-quantized wave-function of an electron confined in the \(z\)-direction). In the ideal 2D case, \(F = 1\) and \(1/M_s\) is negative (though at \(q \gg 1\) \(E(S_z, q)\) increases with \(q\) \([6]\)). In the case of modern wide quantum-well structures \(F(q)\) rapidly decreases with \(q\), and \(1/M_s\) becomes positive. We will consider precisely this real situation, i.e. assume that \(1/M_s > 0\). Our final result does not depend actually on \(1/M_s\). In practice the \(\Delta E_C\) and \(1/M_s\) values can be found from experimental data. According to \([2]\) one obtains \(\Delta E_C \approx 0.35\text{ meV}\). There are no direct measurements yielding \(1/M_s\), but we can estimate this and find that

\[\Delta E_C < 1/1M_s \leq 1\text{ meV},\]
so that \(1/M_s \gg T \sim 0.1\text{ K}\). Since \(c_2 = 0.0255B\) meV and \(\hbar\omega_c = 1.73B\) meV \((B\) is in Teslas), hence the first term in Eq. \((2)\) is always much larger than all the remaining terms.

**Relaxation — How can the CSFE decay?** This process is determined by two necessary conditions: by the availability of an interaction that does not conserve the electron system spin, and by a mechanism of energy dissipation making the relaxation process irreversible. If comparing to possible mechanisms of the spin-exciton relaxation in a quantum Hall ferromagnet \([6]\), the analysis shows that in our case the only CSFE relaxation channel is governed by the spin-orbit and electron-phonon couplings, leading to acoustic phonon emission. The decay probability is determined by the Fermi golden rule,

\[w_{fi} = (2\pi/\hbar)|\mathcal{M}_{fi}|^2 \delta(E_f - E_i),\]
where in the initial state \(|i\rangle\) the number of SCFEs is greater by one than in the final \(|f\rangle\), and conversely the number of phonons is greater by one in \(|f\rangle\) than in \(|i\rangle\), \(\mathcal{M}_{fi}\) being the relevant matrix element.

Methodically it is useful to develop the approach as applied to the general case where \(\nu = 2n + 2\) \((n = 0, 1, 2, ...).\) The spin-orbit coupling is described by a single electron term of the total Hamiltonian, namely \(H_{so} = \alpha(\mathbf{q} \times \mathbf{\sigma}) + \beta(q_y\sigma_y - q_z\sigma_z),\) where \(q = -\sqrt{\epsilon}eA/\hbar\). This operator represents a combination of the Rashba \((\sim \alpha)\) and Dresselhaus \((\sim \beta)\) terms \([11]\) and does not violate translational symmetry. As usual \([3]\) it is convenient to employ a bare single-electron basis diagonalizing the single-electron Hamiltonian \(q^2/2m_e^* + H_{so}\). Within the leading order in the \(H_{so}\) terms one obtains the basis states,

\[\Psi_{np} = \left(\sqrt{n\pi}e^{im\phi}\Psi_{n+1,p} + i\sqrt{m\pi}e^{im\phi}\Psi_{n-1,p}\right)\]
\[\Psi_{np} = \left(-\sqrt{n\pi}e^{im\phi}\Psi_{n-1,p} + i\sqrt{m\pi}e^{im\phi}\Psi_{n+1,p}\right),\]
where \(n = \beta\sqrt{2}/l_B\hbar\omega_c\) and \(v = \alpha\sqrt{2}/l_B\hbar\omega_c\) are small dimensionless parameters. Thus the single-electron states acquire a chirality labeled by subindex \(n\) or \(\overline{n}\) instead of pure spin-up \((b)\) and spin-down \((a)\) quantum numbers. The definition of the exciton creation operator formally remains the same [Eq. \((1)\)], although the \(a_p\) and \(b_p\) operators now describe annihilation in some states \([3]\). In particular, in the case of the CSFE \(a_p\) corresponds to annihilation in the state \(\Psi_{np}\) and \(b_p\) to creation, resulting in the \(\Psi_{np}\). Exactly this transition from \(\Psi_{np}\) to \(\Psi_{np}\) now represents the \(\overline{n} \rightarrow n + 1\) promotion, and our task is to calculate the relaxation of the \(Q_{11}\mathbf{q}|0\rangle\) state.

The Hamiltonian of electron coupling to 3D acoustic phonons with momenta \(k = (\mathbf{q}, k_z)\) is written as

\[\hat{H}_{e-ph} = \frac{\hbar^3}{L_z^{1/2}} \sum_{\mathbf{q},k_z,s} U_s'(k) P_{k,s}^1 H_{e-ph}(\mathbf{q}) + \text{H.c.} \quad (4)\]
(e.g., see Ref. \([3]\)), \(L_z = 2\pi N_0 l_B^2\) is the 2D area, \(L_z\) is the thickness of the slab, \(P_{k,s}^1\) is the phonon creation Bose operator, i.e. state \(P_{k,s}^\dagger|0\rangle\) represents a combination of the 2D electron ground state and a 3D phonon with momentum \(k\) and polarization \(s\), \(H_{e-ph}(\mathbf{q}) = \int e^{i\mathbf{q}\mathbf{r}}\hat{\Psi}(\mathbf{r})\hat{\Psi}(\mathbf{r})d^2r\), and \(U_s'(k)\) is the renormalized vertex, \(U_s'(k) = U_s(k)\Phi(k),\) where

\[\Phi(k) = \int e^{ikz}\chi(z)^2dz.\]
When substituting \(\hat{\Psi}(\mathbf{r}) = \sum_{np}[a_{np}\hat{\Psi}_{np} + b_{np}\hat{\Psi}_{np}],\) we only keep in \(H_{e-ph}\) the terms governing the
decay of the $Q_{n+1}^{q}$ state, and the dimensionless operator $\mathcal{H}_{e-ph}$ relevant to the case takes the form $\mathcal{H}_{e-ph}(q) = \sqrt{N_{\phi}}G_{n}(q)\mathcal{Q}_{m+1}^{q}$ where $G_{n}(q) = L_{n}^{1}(q^{2}/2)e^{-q^{2}/4}[v(q^{2} + iq^{2}/2)/(n + 1)]^{1/2}$. $L_{n}^{1}$ is the Laguerre polynomial.

Further manipulations are simplified in view of a basic feature of the studied relaxation process – only ‘hard’ phonons (with frequency $\approx \omega_{c}$) are generated at the CSFE distribution. Besides, the calculation shows that only phonons emitted almost parallel to the $z$-direction are relevant, i.e. $q \lesssim 1 \ll k_{x} \approx \omega_{c}/c_{s}$. In this connection, there are also apparent simplifications for the vertex $U$, namely: (i) only the contribution of the deformation phonon field where the amplitude is proportional to $\sqrt{k}$ has to be taken into account, and (ii) only LA phonons ($s = l$) give rise to the deformation potential in the GaAs lattice [12].

For the 3D vertex one only needs the expression for the square (see [6]); $|U|^{2} = \pi \delta_{cph}(k_{y})/\hbar^{2}D$. For the exciton-exciton scattering process: $|i| = \mathcal{Q}_{m+1}^{q} \mathcal{Q}_{m+1}^{q}$, and $|f| = \mathcal{Q}_{m+1}^{q} \mathcal{Q}_{m+1}^{q}$. Respectively, the matrix element is equal to $M_{j_{1}}(k_{x}, q_{0}) = \left< l_{q} \mid \sum_{q} U_{1}(k_{x}, q_{0}) \right. \mathcal{Q}_{m+1}^{q} \mathcal{Q}_{m+1}^{q} \left| \right. k_{x}, q_{0} \right.$, or to $M_{j_{2}}(k_{x}, q_{1}, q_{2}) = \left< l_{q_{1}} \mid \sum_{q} U_{1}(k_{x}, q_{2}) \mathcal{Q}_{m+1}^{q} \mathcal{Q}_{m+1}^{q} \left| \right. k_{x}, q_{0} \right.$, where $M_{j_{1}}(0) = \langle 0 | \mathcal{Q}_{m+1}^{q_{1}} | 0 \rangle = \delta_{q_{1}q_{0}}$ and $M_{j_{2}}(0) = \langle 0 | \mathcal{Q}_{m+1}^{q_{1}} \mathcal{Q}_{m+1}^{q_{2}} | 0 \rangle$ (the latter expectation is calculated with the help of excitonic commutation algebra [1, 2]).

$M_{2} = \delta_{q_{1}q_{2}}\delta_{q_{1}q_{3}}\delta_{q_{2}q_{4}}\delta_{q_{3}q_{4}} - \frac{2 \cos \phi}{\hbar^{2}D} \delta_{q_{1}q_{2}} \delta_{q_{3} + q_{4}}$, where $\phi = (\langle q_{1} + q_{2} + q_{3} \rangle)/2$. Generally, in order to calculate the CSFE relaxation rate, $R = \sum_{i} w_{j_{i}}$, one should know the distribution $N_{q}$ of excitons over the $q$ wave numbers. At any moment the CSFE distribution is quasi-equilibrium one, and characterized by an ‘adiabatic’ chemical potential, since the thermodynamic equilibrium is certainly established much faster than the CSFE decay processes occur. (The excitons obey Bose statistics because their number in any state determined by a certain $q$ may be macroscopically large.) Our conditions are as follows: (i) initially the total number of excitons $N_{x} = \sum_{q} N_{q}$ excited by a short external optical pulse is rather large: $N_{x} \sim 0.01 N_{\phi}$; (ii) the relevant values are $q \sim \sqrt{M_{x}T} \ll 1$, and $N_{q} \sim e^{-q^{2}/2M_{x}T}$ if $q^{2} \sim M_{x}T$. First, we estimate the single-exciton relaxation rate:

$$R_{1} = \frac{2\pi}{\hbar} \sum_{k_{x}, q_{0}} \left| M_{j_{1}}^{(1)}(k_{x}, q_{0}) \right|^{2} \delta(h\omega_{c} - \hbar \omega_{c} \sqrt{k_{x}^{2} + q_{0}^{2}}/\hbar^{2}B)$$

$$\approx \frac{2\pi}{\hbar^{2}D} \sum_{q_{0}} N_{q_{0}} G_{n}(q_{0})^{2} \sim N_{x}/\tau_{1},$$

where $1/\tau_{1} = (M_{x}T)^{2}/\tau$ – see $\tau$ below defined in Eq. (7). When calculating the rate due to the two-exciton scattering, one finds that under our condition the dominant contribution to the rate is provided by the $\sim 1/N_{\phi}$ term of the expectation (6) (also $\phi = 0$ has to be set), and the rate is

$$R_{2} = \frac{\pi}{\hbar} \sum_{k_{x}, q_{1}, q_{2}} \left| M_{j_{2}}(k_{x}, q_{1}, q_{2}) \right|^{2} \times N_{q_{1}} N_{q_{2}} \delta(h\omega_{c} - \hbar \omega_{c} \sqrt{k_{x}^{2} + q_{1}^{2} + q_{2}^{2}}/\hbar^{2}B)$$

$$\approx \frac{2\pi}{\hbar^{2}D} \sum_{q_{1}, q_{2}} G_{n}(q_{1})^{2} N_{q_{1}} N_{q_{2}} \sim 2N_{x} N_{q}^{2} (u^{2} + v^{2}).$$

The summation in this expression is reduced to $\sum_{q_{1}, q_{2}} G_{n}(q_{1})^{2} N_{q_{1}} N_{q_{2}} \sim 2N_{x} N_{q}^{2} (u^{2} + v^{2})$. Finally, if $n_{x} = N_{x}/N_{\phi}$ is the CSFE concentration, then the kinetic equation takes the form $dn_{x}/dt = n_{x}^{2}/\tau$ with characteristic inverse relaxation time

$$\tau = \frac{4\omega_{c}^{2} \Phi(\omega_{c}/\epsilon)}{\hbar^{2}D} \left( u^{2} + v^{2} \right) / \left( 1 + n_{x}^{2} \right).$$

Obeying equation $n_{x}(t) = n_{x}(0)/(1 + n_{x}(0)t/\tau)$, the SCFE density decays nonexponentially. Yet, note that due to the nonexponentiality of the relaxation, the real value that should be compared with the experimental results is not $\tau$ but $\tau' \sim \tau/n_{x}(0)$.

Meanwhile the single-exciton relaxation, though exponential, occurs much more slowly – the characteristic relaxation time $\tau_{1}$ is by a huge factor $\sim (M_{x}T)^{-2} \sim 10^{-4} - 10^{-3}$ longer than in the case of exciton-exciton scattering (7). This feature is determined by considerable enhancement of the relaxation phase volume in the case of two-exciton scattering processes.

Discussion — When numerically estimating $\tau$, one faces the basic difficulty related to uncertainty in the form-factor $\Phi(\omega_{c}/\epsilon)$ value. This strongly depends on the poorly observable function $\chi(z)$. However, it is clear that for a wide quantum well with effective thickness $d \approx 20$ nm and for a magnetic field, e.g. equal to 5 T, a considerable incommensurability of $d$ and $\omega_{c}/\epsilon = k_{x} \approx 3$ nm takes place. This fact tremendously reduces the Fourier component $\chi(z)$. When estimating $\chi(z)$ by means of three models: (i) Fang and Howard; (ii) Takada and Uemura (see [14] and references therein); and (iii) the simple model where quantum-well walls are considered to be infinitesimally high, one finds that $\Phi(k)^{2} \approx C/(kd)^{6}$ where $C \approx 4.7 \times 10^{-4}, 6.5 \times 10^{-5}$, and $3.1 \times 10^{-5}$ respectively (in all three models is set equal to the average penetration length of the charge into the semiconductor). Substitution of this estimate into Eq. (7) and the assumption that $u^{2} + v^{2} = 10^{-3}/B$ (B is in Tesla) [6] yields for a quantum well with $d = 20$ nm: $\tau \approx 10^{-2} \times B^{3}/\epsilon$, and hence $\tau' \sim B^{3}/\epsilon$. At $B = 5$ T
even for the ‘fastest’ Fang-Howard model describing the $\chi(z)$ wave-function, one gets estimates of $\sim 7\text{ ms}$ and $\sim 0.7\text{ s}$ for the $\tau$ and $\tau'$ times respectively.

The CSFE was diagnosed in experiment [2] as a result of Raman scattering. However, the direct photo-absorption of photons with angular momenta $-1$ ($\sigma^-$-photons) in the vicinity of laser carrier frequency $\omega_{\text{exc}} \approx \omega^0_c + \omega_{\text{c}} + (E_g + \epsilon_2)/2 - 3|g_h\mu_B B|/2)/\hbar$ would seem to be more intensive way of CSFE creation $[E_g(B)]$ is the GaAs/AlGaAs forbidden gap depending on the magnetic field; $\omega^0_c$ and $g_h$ describe the cyclotron frequency and the $g$-factor of the valence-band heavy-holes]. This pulse creates the $S_z = 1/2$ electrons in the first Landau level and the $S_z = -3/2$ heavy-holes in the first Landau level within the valence band. The latter, due to some fast relaxation and recombination processes, should convert into the effective $+1/2$ hole in the electronic zeroth Landau level. As a result the CSFE emerges. One could also quench the CSFE by using, e.g. a zeroth Landau level. As a result the CSFE emerges.

Finally, we concern the relaxation of other excitations with energies higher than but close to the same $\omega$, depending on the magnetic field; $\sigma^-$ pulse at the frequency $\omega_{\text{exc}} \approx (E_g - \epsilon_2)/2 + 3|g_h\mu_B B|/2)/\hbar$ and simultaneously a $\sigma^+$ pulse at the frequency $\sim (E_g + \epsilon_2)/2 - 3|g_h\mu_B B|/2)/\hbar$ filling the zeroth electronic Landau level. As a result one can conclude that even the pulses’ widths $\lesssim 1\mu\text{s}$ would be sufficient for exciting/quenching, and the ratio of the excited state lifetime $\tau'$ towards the time of the state switching on/off is greater than $10^6$.

correspondingly canceling a thermal-activation transition to a radiatively relaxing state. The CSFE mode could be controlled optically by exciting and ‘quenching’ pulses with widths $\lesssim 1\mu\text{s}$. The long-lived excitons have to form an exciton Bose-Einstein condensate. The number of excitons in the condensate is governed by the amplitude and width of the optical pulse exciting the system. This state should be similar to a ‘thermodynamic condensate’ of spin waves in a quantum Hall ferromagnet [2], where excitons’ momenta are not strictly equal to zero due to an inhomogeneity caused by an external disorder, but belong to a phase volume that is much smaller than the total number of excitons in the condensate (see Ref. [6]). Decay of the condensate should also be characterized by time $\tau$.

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