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Large oscillations in the photoluminescence spectra of a GaAs quantum well in external magnetic fields: A direct measurement of charge transfer in an electron bilayer system

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Photoluminescence studies of a wide asymmetric quantum well hosting two strongly coupled electron layers demonstrate unambiguously how such a system deforms itself due to interlayer charge transfer in a quantizing magnetic field thus manifesting its overall single-layer appearance. The charge distribution inside the quantum well is balanced for total filling factor $\nu=4N$ (N integer), while it is imbalanced at most for $\nu=4N+2$. The intersubband energy oscillates enormously with the applied magnetic field and reaches values as large as the electron cyclotron energy for filling factors 4N.

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Once a two-dimensional (2D) electronic system is split into two closely spaced subsystems with appreciable coupling via Coulomb interactions and tunneling, a rich variety of qualitatively new physical phenomena arises. Prime examples are the collapse of particular integer quantum Hall states,¹ onset of the uncommon $\nu = \frac{1}{2}$ fractional quantum Hall state,² spontaneous symmetry breaking due to stabilization of phases with unequal fractional filling factors,³ and excitonic superfluidity in two half-filled Landau levels (LLs).⁴ Subtle interplay between the single-particle physics and many-body intra- and interlayer interactions can be realized and tuned by changing system parameters such as the total electron density and its distribution between the two layers and the magnitude and direction of the applied magnetic field.^{5,6}

Recently some attention has been paid to the behavior of wide quantum wells (QWs), where nonlinear depopulation with a gate bias was ascribed to exchange interaction effects.⁷ Hybrid response (mixing of single-layer and double-layer properties) was observed in quantum Hall effect studies of an imbalanced double QW, and explained by interlayer charge transfer driven by magnetic field.⁸ Further magneto-capacitance and far infrared-spectroscopy techniques⁹ confirmed this conjecture also for a wide quantum well. Hartree calculations, in turn, provided a strong support for the proposed scenario,⁹ but experimentally it was not possible to qualitatively describe the charge transfer and to extract relevant energy scales. Moreover, findings of Ref. 6 have been tried to be explained in terms of quantum Hall ferromagnetism.

Here we present our magnetophotoluminescence studies of an imbalanced wide quantum well which reveal spontaneous electrostatic deformations of the well in quantizing magnetic fields. The data indicate an effective *single-layer* behavior of the bilayer system realized through an interlayer charge transfer at a substantial capacitive energy cost. At total filling factors $v = \{6, 10, 14, ...\}$ this results in an actual locking of subbands. By scrutinizing spectral dependencies we are able to extract details of the charge transfer process.

Our sample is grown by molecular beam epitaxy and comprises a single GaAs quantum well of width 500 Å embedded in $Al_{0.3}Ga_{0.7}As$. The electrons are provided by a set of six distributed Si δ -layers situated on the top side of the

QW, with the closest doping layer 600 Å spaced apart from the QW. The as-grown carrier density is $n=1.4 \times 10^{11}$ cm⁻², and all charges reside at the top GaAs/AlGaAs interface. n^{++} -GaAs front and back gates were *in situ* incorporated into the structure at distances 1.21 and 1.05 μ m from the QW, respectively. The sample offers the privilege to have two gates and a high-quality layer of 2D electrons simultaneously and at the same time allows for highly sensitive optical experiments. Various density and charge configurations (the total density range is 1 to 6×10^{11} cm⁻²) are obtained by biasing both gates. The laser excitation at 780 nm and the collected photoluminescence (PL) signal were guided via two corresponding optical fibers. All measurements were performed at 1.5 K.

Images (a) and (b) in Fig. 1 show pseudocolor-scale plots of magnetic field and wavelength dependent PL spectra recorded at front-gate voltages -0.2 V (a) and +0.4 V (b) (bright corresponds to maximum intensity, dark to zero). The back-gate bias was kept at +5 V for both plots. Low magnetic field data (B < 1 T) clearly indicate that two LL fans are present corresponding to the filled subbands at both sides of the QW, as highlighted in the inset of Fig. 1. In this limit the energetical separation between PL lines from the lowest LLs is equal to the zero-magnetic field energy splitting $\Delta(0)$ between subbands. Therefore we simply obtain $\Delta(0)=1.1$ meV for Fig. 1(a) and $\Delta(0)=2.5$ meV for the situation in Fig. 1(b). Thus changing the front-gate bias from -0.2 V to +0.4 V promoted the density imbalance of our QW.¹⁰ Of main interest here is the distinctive response of optical spectra to magnetic fields B > 1 T, where striking oscillations of photoluminescence energies and intensities are observed.

Dotted lines E_1 and E_2 in Fig. 1 indicate the expected behavior of PL maxima from the lowest LLs if the two layers were completely uncoupled (e.g., infinitely far apart from each other, see also the illustrative inset of Fig. 1). In contrast to this a series of alternating intensity buildups from both subbands is observed, the subband energy splitting varying from nearly zero to as much as 10 meV. Each Landau fan shifts as a whole as indicated by photoluminescence from filled higher LLs. Figure 2(a) demonstrates the dependence of PL intensities around lines E_1 and E_2 from Fig. 1(a)



FIG. 1. (Color online) Images of magnetophotoluminescence from the 500-Å-wide QW for the front-gate voltages -0.2 V [left image, corresponding to the zero-field detuning $\Delta(0)=1.1$ meV] and +0.4 V [right image, $\Delta(0)=2.5$ meV]. Lines E1 and E2 trace the expected positions of PL lines from the lowest LLs if the electronic subbands were independent, the inset also illustrates the PL fans for uncoupled electronic layers.

on the applied magnetic field. The same data plotted versus inverse magnetic field [Fig. 2(b)] clearly reveal that a *single* frequency governs all the intensity changes. It's natural then to assign this frequency to some filling factors of the whole system, and the only relevant set of filling factors is indicated in Fig. 2(b).¹¹ By tracking the PL positions [Fig. 2(c)] from the lowest LLs we conclude that at $\nu = \{4, 8, 12, ...\}$ two subbands energetically repel each other while at $\nu = \{6, 10, 14, ...\}$ a pronounced tendency for them to lock together is clearly seen. For an even more imbalanced system the effect is much more pronounced, manifesting as enormous oscillations of the intersubband energy.

We explain our findings in terms of intersubband charge transfer. An appreciable capacitive coupling of closely spaced subbands results in the charge transfer as the thermodynamic density of states changes in the applied magnetic field. This is the direct consequence of Landau quantization while minimizing the total energy of the system.

In the single-particle picture the potential energy of the system is of electrostatic nature and determined by external electric fields that result from ionized donors and gates. The zero-magnetic field kinetic energy is simply the sum of (occupied) subband Fermi energies and linearly scales with the total electron density. Thus at given density the zero-field distribution of electrons delivers minimal potential energy as this case corresponds to the minimal total energy. The magnetic quantization of an in-plane kinetic energy into LLs realizes the situation when the total energy depends on the charge distribution not only electrostatically but also through relative fillings of quantum levels in the two subbands. The interplay between these constituents determines the dynamics of our bilayer system at different filling factors.

To illustrate this point, let us consider the case of total filling factor $\nu=4$ for the system with zero-magnetic field density detuning of $\delta n/n_{tot}=10\%$ [sketch (i) of Fig. 3(a); the detuning of 10% is relevant for Fig. 1(a)]. If we assume the charge distribution has remained unchanged at $\nu=4$, then lower and upper subbands have filling factors of $\nu_1=2.2$ and

 $\nu_2=1.8$, respectively $(\frac{\nu_1-\nu_2}{\nu_1+\nu_2}=10\%)$, and the intersubband splitting equals $\Delta(0)$ as illustrated in the situation (ii) of Fig. 3(a). However, it might be possible to reduce the kinetic energy of this configuration due to transfer of electrons from the second spin-split LL of the lower subband into the partially filled sublevel of the first LL in the upper subband. This process is inevitably accompanied by an increase of the system's potential energy, and, even more important, changes the relative energies of the two subbands [case (iii) in Fig. 3(a)].

The exact quantitative description of the aforementioned phenomena takes into account the full complexity of electrostatic field distribution (due to finite thicknesses of electronic sheets, and their screening lengths that are comparable to the QW width) and the competition of intra- and interlayer Coulomb interactions.



FIG. 2. (Color online) Adaptation of data from Fig. 1. (a) The PL intensity oscillations when following lines E_1 and E_2 . (b) Data as in (a) but plotted versus inverse magnetic field. Note the single frequency oscillations of intensities, and identification of the relevant *total filling factors*. (c) The traces of PL positions from the lowest LLs of both subbands, for two detunings $\Delta(0)$.



FIG. 3. Physical model for the charge-transfer process. (i) The zero-field detuning is set to 10%, E_{F1} and E_{F2} are the Fermi energies of corresponding electronic layers (sheet 1 and sheet 2 in the figure). (a) The case of a total filling factor ν =4 if we keep the initial imbalance (ii), and if the system is allowed to minimize its total energy (iii), see the text for details. The horizontal thin and thick solid lines represent empty and filled parts of the spin-split Landau levels. Here the two subbands repel each other due to the charge transfer, and the system reaches the $\nu_1 = \nu_2 = 2$ distribution. (b) The analogous consideration but for the case of ν =6. Now the subbands turn out to be virtually locked.

We limit ourselves here to the simplest consideration that nevertheless perfectly elucidates the underlying physics.¹² One can approximate adjacent subbands as two infinitely thin conducting plates with electron densities n_1 and n_2 separated by distance *d*. The calculated value of *d* is roughly 200 Å for our 500-Å-wide QW and the actual electron densities; later on we will obtain the experimental estimation for *d*. The transfer of δn sheet density from one plate to another causes a difference in single electron energies between the plates of $E_c = 4\pi de^2 \delta n/\epsilon$ (here and in the following we use the CGS system), the plate being enriched moving up in energy with respect to the depleted one regardless of values for n_1 and n_2 .

Turning back to relative positions of subbands for case (ii) in Fig. 3(a), we conclude that the bilayer system at a total filling factor $\nu=4$ tends to compensate the initial zero-field imbalance by the corresponding charge transfer. Electrons flow until the system reaches the exact $\nu_1 = \nu_2 = 2$ distribution *or* LLs from two subbands with opposite spins and different orbital numbers come into alignment. The latter happens when the energy separation between subbands

$$\Delta_{\nu=4} = \Delta(0) + 4 \pi de^2 \delta n/\epsilon$$

approaches the value $\hbar \omega_c - g \mu_B B$.

An even more imbalanced system requires more electrons to change their subband index thus increasing the intersubband splitting. This fact is displayed in Fig. 2(c) which compares spectral positions of PL lines from the 0th LLs of both subbands around $\nu=8$ and $\nu=4$ for two different detunings





FIG. 4. (Color online) $\Delta_{\nu=4}$ vs $\Delta(0)$ when the 2D density of the upper electronic layer is increased by the front-gate biasing. The shaded area of the vertical axis shows the range of the cyclotron energies for the experimental points. Red lines are guided to the eyes. The steep linear region is determined by the intersubband spacial separation $d \approx 230$ Å.

 δn . The value of $\Delta_{\nu=4}$ becomes as huge as 10 meV at B=6.5 T ($\hbar \omega_c \approx 11$ meV) and the initial detuning of $\Delta(0)=2.5$ meV.

For $v_1 = v_2 = 2$ the intersubband splitting has its local maximum. This fact can be illustrated in the following way: for example, let's slightly increase the magnitude of the magnetic field thus creating total v < 4. As the degeneracies of all LLs grow equally, a possibility for electrons being transferred from the upper subband into the lower one arises. Clearly this process reduces the overall kinetic energy of the system, and decreases the energetical gap between subbands. Vice versa, one can consider the case when the total filling factor slightly exceeds the exact value of 4. Again, we come to the conclusion that the intersubband energy splitting reduces.

The previously written expression for $\Delta_{\nu=4}$ deserves a further analysis. As $\Delta(0) = \frac{\pi \hbar^2}{2m^*} \delta n$ scales linearly with δn , it can be represented in the evident form

$$\Delta_{\nu=4} = \Delta(0)(1 + 8d/a_b),$$

where $a_b = \frac{\hbar^2 \epsilon}{m^* e^2} = 100$ Å and $m^* = 0.067m_e$ are the Bohr radius and the effective mass of electrons in GaAs, respectively, and $\epsilon = 12.8$ is the GaAs static dielectric constant. In turn, for a wide QW the intersubband spacial separation *d* is a function of δn . The value of *d* is zero for a completely balanced system (when $\Delta(0)$ is the symmetric-antisymmetric gap) and grows monotonically with the detuning, nearly saturating for high δn . Therefore one may expect a rather complicated dependence of $\Delta_{\nu=4}$ on $\Delta(0)$.

Figure 4 demonstrates the experimental data for the behavior under discussion; here we keep the sheet density for one subband constant and increase it in the other subband by varying the front-gate bias. Two regimes can be clearly observed in this plot. The first regime shows a steep linearity on $\Delta(0)$ in the range 0.8–1.2 meV, while in the second one $\Delta_{\nu=4}$ approaches the electron cyclotron energy and depends on $\Delta(0)$ only through the change in the total electron density (and the corresponding increase of the cyclotron energy at $\nu=4$). Hence we conclude that starting from the zero-field detuning of approximately 1.5 meV, the intersubband splitting $\Delta_{\nu=4}$ reaches its upper limit (roughly $\hbar\omega_c$), and the system loses its ability to attain the perfect $\nu_1 = \nu_2 = 2$ distribution. At the same time the slope of the linear dependence for smaller $\Delta(0)$ allows for the estimation of the intersubband spacial separation *d*, and it gives a very reasonable value of d=230 Å.

Obviously the same consideration is also true for any total filling factor that can be written in the form $\nu = 4N$ where N is a positive integer. The system will balance itself up to critical fillings when $\hbar\omega_c$ becomes comparable to E_c .

However at total filling factors $\nu=4N+2$ the minimization of the overall system energy gives rise to a drastically different result. For example, by analyzing the case of $\nu=6$ in the same way as for $\nu=4$, we observe that the fillings of $\nu_1=3.3$ and $\nu_2=2.7$ [$\delta n/n_{tot}=10\%$, Fig. 3(b)] are unstable against the electron redistribution which *tends to detune the system even further*. At a small cost of the increase in the Zeeman energy (as compared to the cyclotron energy and $\Delta(0)$ in the displayed magnetic field range), electrons from the upper subband decrease their potential energy by populating the lower subband until appropriate Landau levels align. In this case the intersubband splitting becomes as small as the Zeeman spin splitting.

Therefore the moderately detuned bilayer system in strong magnetic fields has a remarkable symmetry at even total filling factors: either it is perfectly balanced in a sense of charge distribution (at $\nu=4N$) or it is nearly balanced in

the sense of relative positions of the lowest energy levels of subbands (at $\nu = 4N+2$).

Within the suggested picture the alternating PL intensity buildups from both subbands are directly explained. Due to the electrostatic deformations, optically excited holes track the changes in the potential profile of the quantum well. Therefore if there is some transfer of electrons from one QW side to the other, the holes move in the same direction. This changes the overlap of hole and electron wave functions for both subbands. The immediate consequence is the severe decrease of intensity from the subband being depleted. As it was discussed earlier, the depletion of one subband also reduces its energy with respect to another one, and this point completes the qualitative explanation of main features of the data shown in Fig. 1.

To summarize, we have observed a peculiar oscillating behavior of magnetophotoluminescence spectra from a wide quantum well containing two imbalanced electronic layers. The features in PL intensities and spectral positions are clearly explained in terms of charge transfer between subbands. The intersubband energy splitting is shown to oscillate with a single frequency determined by the total 2D electron density in two layers, and reaches giant values up to the electron cyclotron energy.

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- ¹⁰ The intersubband splitting is minimal and equals the symmetricantisymmetric gap when the QW is perfectly balanced (or symmetric). In that case the wave functions and corresponding subbands are delocalized in a sense they belong to both layers simultaneously. Here we consider the situation when the system is far away from its balance, so in fact each wave function is confined within corresponding layer. Therefore the terms subband and layer are equivalent throughout this discussion.
- ¹¹As that set corresponds to the total density 5.3×10^{11} cm⁻², approximately the same value is extracted from the sum of optically measured concentrations in each subband.
- ¹² The following reference presents somewhat analogous consideration but it is limited to the case of zero-magnetic field: P. P. Ruden *et al.*, Appl. Phys. Lett. **59**, 2165 (1991).