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## Excitonic recombination processes in spin-polarized two-dimensional electron gases

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We report the observation of Coulomb interaction effects on the photoluminescence (PL) spectra of twodimensional electron systems close to the  $\nu = 1$  quantum Hall state. Systematic measurements of the polarized PL spectrum as a function of the separation between the two-dimensional electron gas (2DEG) and the photoexcited valence-band hole allow us to identify the recombination mechanisms. These studies demonstrate the formation of bound states between the photoexcited valence-band hole and the  $\nu = 1$  2DEG. Usually the exciton comprises a single electron in the upper spin level bound to the hole, but the PL line shape suggests that for large separations a Skyrmion-hole excitation forms the ground state. [S0163-1829(98)50632-7]

The two-dimensional electron gas (2DEG) formed in a semiconductor heterostructure provides a model system for studying the electron-electron interaction. One manifestation of the many-body interactions of particular current interest is the nature of the 2DEG close to the  $\nu = 1$  integer quantum Hall state. Theories<sup>1</sup> show that interactions act to stabilize the spin-polarized,  $\nu = 1$  ground state consisting of a filled lowest Landau level of spin- $\uparrow$  electrons. However, they also cause the low-energy charged excitations to have exotic spin structures. These large spin charged excitations are referred to as "Skyrmions" or "charged spin textures." Experiments confirm the charged excitations of GaAs systems at  $\nu = 1$  do indeed involve large spin reversal.<sup>2</sup>

Recent theoretical work<sup>3</sup> has suggested that similar spinreversed states can form in photoexcited 2DEG's close to  $\nu$ =1. These states arise from the interactions of a valence band (vb) hole with neutral (spin-wave) or charged (quasiparticle, or Skyrmion) excitations of the spin-polarized 2DEG. The form of the low-energy initial states depends sensitively on the spatial separation between the vb hole and the electron gas (i.e., on the strength of the interactions between the hole and the excitations of the 2DEG), and on the sign of  $\nu - 1$  (which determines whether the excess charges in the  $\nu = 1$  state have a positive or negative electrical charge). The recombination of these initial states can lead to the formation of elementary excitations of the electron gas (spin waves, quasiparticles, and Skyrmions), which appear as a "shake-up" structure in the photoluminescence (PL) spectrum.

We present here a comprehensive study of PL mechanisms in 2DEG's around  $\nu = 1$ . Our results demonstrate that the Coulomb interaction strongly modifies both the initial and final states of the recombination process. A key aspect to our experiment is the ability to control both the electron density and the spatial separation between the vb hole and the electron gas. This separation has been varied in two ways. First, by applying an electric field we are able to polarize the electrons and vb holes to opposite sides of a quantum well (QW). Second, by using a double QW structure and studying the recombination of electrons in a 2DEG formed in one QW with vb holes confined to the other, we have been able to investigate the recombination of very distant holes. The ability to follow the evolution of the spectral features as a function of this separation is crucial to elucidating which states contribute to PL. Furthermore, by resolving the circular polarization of the emitted light, we can distinguish between recombination involving spin- $\uparrow$  and spin- $\downarrow$  electrons, allowing an understanding of the spin structure of the states responsible for PL.

We measured the optical properties of 2DEG's formed in remotely doped GaAs/Al<sub>0.33</sub>Ga<sub>0.67</sub>As QW's, grown by molecular beam epitaxy on (100) GaAs substrates. The single-QW structure has (in order of growth) 300-Å GaAs QW, 600-Å undoped AlGaAs spacer, and 2000-Å AlGaAs Si-doped ( $10^{17}$  cm<sup>-3</sup>) layer. The two double QW's are nominally identical except that the single well is replaced by (in order of growth): 200-Å/25-Å/300-Å QW/barrier/QW

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FIG. 1. (a),(b) PL spectra recorded at 4 T with different front gate biases on the 300-Å single QW, corresponding to electron densities around  $\nu = 1$  (9.7×10<sup>10</sup> cm<sup>-2</sup>) for zero back gate bias. Insets: Schematic diagram of the excitonic recombination process seen for small electron-hole separation.

layers in the first and 200-Å/25-Å/200-Å in the second. Mesas were fabricated with Ohmic contacts to the 2DEG and depleting Schottky gates both above and below the 2DEG. By biasing the front and back gates with respect to the 2DEG, we are able to independently control both the electron density and the electric field across the 2DEG region. We determine the electric field at the back of the QW for a particular back gate bias by comparing the zero electron density Stark shift of the neutral exciton line to an effective mass calculation.<sup>4</sup>

Experimental data taken on the 300-Å single QW are plotted in Figs. 1–3. Figure 1 displays PL spectra recorded at a fixed magnetic field of 4 T, a sample temperature of 2 K, and with fixed back gate bias of zero. The spectra show a very striking polarization dependence. Notice that, as the front gate bias is changed, thereby altering the electron density and hence the filling factor, a broadening occurs on the low-energy side of the PL line seen in  $\sigma^-$  polarization [Fig. 1(a)] around  $\nu = 1$ , while, in contrast, a smooth evolution is seen in  $\sigma^+$  [Fig. 1(b)]. The low-energy broadening of the  $\sigma^$ line at  $\nu = 1$  increases with magnetic field and vanishes at higher temperature. Similar behavior has recently been reported by Plentz *et al.*<sup>5</sup> for a 250 single QW sample (without a back gate contact).



FIG. 2. As in Fig. 1, but with a finite back gate bias applied, corresponding to an electric field at the back of the QW of 10.1 kV/cm. Schematic of the nonexcitonic (c) and excitonic (d) recombination processes responsible for PL at larger electron-hole separations, for  $\nu < 1$  and  $\nu > 1$ , respectively.



FIG. 3. (a) Evolution of the PL peak energies (of Figs. 1 and 2) with filling factor ( $\nu$ ) for low and high applied electric fields. (b) Integrated PL intensity in  $\sigma^+$  polarization vs filling factor. The discontinuities at  $\nu = 1$  in (a) and (b) are due to a change in the ground state from excitonic to nonexcitonic [see insets of (b)] at high electric field. In contrast the recombination is excitonic for both  $\nu < 1$  and  $\nu > 1$  at low electric field. (c) Measured blueshift in  $\sigma^-$  as a function of the electric field at the back of the well. (d) Calculated variation of  $(B_{sw} - B_{ex})$  with electron-vb hole separation (d) using the 2D model of Ref. 3.

Rather different behavior is observed in Fig. 2, where a fixed back gate bias is applied, dropping an electric field across the QW. In this case, a sharp blueshift of the  $\sigma^-$  PL is observed with increasing density near  $\nu = 1$  [Fig. 2(a)]. A smaller blueshift is also apparent in  $\sigma^+$  polarization [Fig. 2(b)] near  $\nu = 1$ . However, the PL observed in  $\sigma^+$  for  $\nu < 1$  is relatively weak at 2 K, but strengthens sharply with sample temperature.

Figures 3(a) and 3(b), which plot the filling factor ( $\nu$ ) dependence of the PL energies and intensities, respectively, further highlight the contrasting behavior under low and high applied electric field. The PL shifts to lower energy with electron density, due to band-gap renormalization and the additional charge-induced internal electric field.<sup>6</sup> However, under high electric field, there is a sharp discontinuity in this redshift with density for  $\sigma^-$  polarization near  $\nu = 1$ , see Fig. 3(a). Notice too the sharp change in the PL intensity in  $\sigma^+$  polarization at  $\nu = 1$  under high electric field, the PL energies and intensities show a smoother evolution with filling factor. The increase in the splitting of the two lines seen in  $\sigma^-$  with the electric field at the back of the QW is plotted in Fig. 3(c).

We discuss now the results for the single QW. The fact that we see a sharp spectral discontinuity in high electric field spectra indicates an abrupt change in the photoexcited ground state with electron density at  $\nu = 1$ , which is absent at low field. Since the effect of the electric field is to increase the separation of the vb hole and 2DEG, this suggests that the change in the ground state is driven by the Coulomb interaction. As discussed in detail below, we suggest that at high electric field the ground state changes from nonexcitonic for  $\nu < 1$  to excitonic for  $\nu > 1$ , as shown schematically in Figs. 2(c) and 2(d). (In these diagrams we ignore the spin of the hole;  $e\uparrow$  electrons recombine with  $h\downarrow$  heavy holes in  $\sigma^-$  polarization, while  $e\downarrow$  recombine with  $h\uparrow$  in  $\sigma^+$ .) On the other hand, at low electric field, for which the Coulomb interaction is stronger, we will argue it is excitonic on either side of  $\nu = 1$ .

Such a description of the photoexcited ground states is supported by the recent theoretical work of Cooper and Chklovskii.<sup>3</sup> They considered the nonexcitonic and excitonic initial states shown in Figs. 2(c) and 2(d), respectively, for the case that the electrons and holes are confined to parallel 2D planes separated by a distance d. They demonstrated the excitonic one to be the lower energy state by an amount equal to  $(B_{ex} - B_{sw} - Z)$ , where  $B_{ex}$  is the binding energy of the  $e \downarrow$  electron and the vb hole,  $B_{sw}$  that of the  $e \downarrow$  electron and the spin hole in the  $e^{\uparrow}$  level, and Z is the Zeeman energy. In the lowest Landau level/subband approximation, and for d=0, where the electron and hole wave functions are identical, symmetry demands  $B_{ex}$  and  $B_{sw}$  are exactly equal. However, corrections arising from electron Landau-level mixing results in  $B_{ex} > B_{sw}$ .<sup>3</sup> Hence for zero, or small, electric fields, the recombination is excitonic in character for both  $\nu < 1$  and  $\nu > 1$ .

Two observations strongly support the excitonic recombination mechanism at low electric field: first, the intense PL in  $\sigma^+$  polarization for  $\nu < 1$ , and second, the low-energy broadening seen near  $\nu = 1$  in  $\sigma^-$  only. If, for the sake of argument, we ignore the formation of the excitonic state, we would expect  $e \downarrow$  electrons to relax to  $e \uparrow$  for  $\nu < 1$ , and hence the  $\sigma^+$  intensity to be weak. In fact, as is apparent in Fig. 3(b) and the spectra of Fig. 1(b),  $\sigma^+$  retains intensity for  $\nu$ <1 and, indeed, is comparably strong to  $\sigma^-$ . The intensity in  $\sigma^+$  for  $\nu < 1$  derives from the fact that it is energetically favorable at low electric field for a ground state to form with a  $e^{\uparrow}$  electron bound to to a vb hole. Turning to the second point, the low-energy broadening of  $\sigma^-$  PL peak near  $\nu = 1$ is explained by the "shake-up" of a spin-wave excitation that is left when the excitonic state recombines in  $\sigma^-$  polarization (see upper inset Fig. 1). The differing dispersions of the exciton and spin wave that form the initial and final states of this recombination process lead to the low-energy broadening of the  $\sigma^-$  PL peak.<sup>3</sup> This explains why the broadening is observed in  $\sigma^-$  polarization and not  $\sigma^+$  [see Fig. 4(c) or Figs. 1(a) and 1(b), for which a spin wave is not formed in the final state.

Application of an electric field separates the electrons and holes toward opposite faces of the QW, due to the quantum confined Stark effect.<sup>7</sup> This weakens the exciton binding energy, while leaving that of the spin wave essentially unaltered. Hence for sufficient field,  $B_{ex} < B_{sw}$ , resulting in the nonexcitonic state [Fig. 2(c)] becoming lower in energy than the excitonic state for  $\nu < 1$ . Under these conditions there is a change in the nature of the initial state upon sweeping the electron density through  $\nu = 1$ ; for  $\nu < 1$  it is nonexcitonic [Fig. 2(c)] while for  $\nu > 1$  it is excitonic [Fig. 2(d)].<sup>3,8</sup> This

explains why qualitatively different behavior is observed in the PL spectra taken under high electric field to those at low field.

Several experimental observations provide support for a change in the photoexcited ground state around  $\nu = 1$  under high applied electric field. First, notice in Fig. 3(b), in contrast to the low electric field data discussed above, that the  $\sigma^+$  PL is much weaker for  $\nu < 1$  than  $\nu > 1$ . (Based on the above considerations of the photoexcited ground states, one would, in fact, expect *zero* intensity in  $\sigma^+$  polarization for the nonexcitonic initial state at  $\nu < 1$ ; the weak PL that is observed is accounted for below in terms of thermal excitation.) Second, notice in Fig. 2(a), that the PL line observed for  $\nu > 1$  in  $\sigma^-$  has the broadening on the low energy side characteristic of a spin wave being formed in the final state, while the line shape for  $\nu < 1$  is symmetric.

Since the final state of the excitonic recombination in  $\sigma^-$ (the spin wave) has larger binding energy than the initial, it has higher transition energy than the nonexcitonic one. This is the origin of the blueshift with increasing density observed at  $\nu = 1$  in  $\sigma^{-}$  polarization, plotted in Fig. 3(a). Discontinuities in the PL energy as a function of magnetic field have been observed in other systems of large electron-hole separation: very wide, unbiased single QW's (Ref. 9) or single heterojunction samples.<sup>10,11</sup> The blueshift should be simply the difference in the spin-wave and exciton binding energies,  $\Delta E = B_{sw} - B_{ex}$ . Hence the experimental observation that  $\Delta E$  increases with electric field, indicates that the excitonic initial state must rise higher in energy than the nonexcitonic one. Figure 3(c) plots the measured blueshift at 4 T as a function of the electric field at the back of the QW. This compares favorably with the dependence of  $(B_{sw} - B_{ex})$  on the electron-hole separation (d) within the 2D model described in Ref. 3 using reasonable values of d, as shown in Fig. 3(d).

We explain now the origin of the weak PL seen in  $\sigma^+$  for  $\nu < 1$  at high electric field. This PL is relatively weak at 2 K, but strengthens rapidly with temperature. As discussed above, the nonexcitonic state is the lowest energy under applied electric field for  $\nu < 1$ . As can be seen in Fig. 2(c), this state should not produce PL in  $\sigma^+$  polarization. The fact that we do see weak PL in  $\sigma^+$  for  $\nu < 1$ , the intensity of which increases with temperature, suggests that the recombining state is thermally excited. A fit of the temperature dependence of the intensity to the functional form  $T \exp(-Z/T)$ , which describes the density of spin waves at low temperatures, gives a value  $Z \sim 0.1$  meV in rough agreement with the known Zeeman energy. The fact that we also see a blueshift near  $\nu = 1$  in  $\sigma^+$  polarization suggests that the PL is due to recombination of vb holes and thermally excited spin waves. Although we expect the blueshift in  $\sigma^+$  polarization to again be  $\sim (B_{sw} - B_{ex})$ , it is smaller than that in  $\sigma^{-}$ , and consequently emerges at a higher electric field, because, due to vb mixing effects, the  $e\!\downarrow\!h\!\uparrow$  exciton seen in  $\sigma^+$  has a larger binding energy than the  $e \parallel h \parallel$  exciton seen in  $\sigma^-$ .

We turn now to discuss the double QW's for which we can observe 2DEG recombination with vb holes in both the same (direct PL) and the other (indirect) QW. (These assignments are based on their front and back gate bias dependencies.) This permits the interaction of the 2DEG with both near and distant vb holes to be studied in the same sample R4230



FIG. 4. (a) PL spectra recorded at 5 T in  $\sigma^-$  polarization with different front gate biases and fixed back gate bias on the 200/25/300-Å double QW. (b) Evolution of the PL peak energies with filling factor. (c),(d) Comparison of PL line shapes in  $\sigma^-$  and  $\sigma^+$  polarizations for (c) single- and (d) double-QW samples.

under the same conditions, allowing a particularly direct test of the importance of the electron-vb hole separation on the form of the PL. Our spectra taken for the indirect transition of the double QW, shows evidence for bound states involving a Skyrmion and a vb hole.

PL spectra recorded on the double QW's at 5 T for densities around  $\nu = 1$  are plotted in Fig. 4. The direct recombination shows similar behavior to the single QW at low field: a broadening on the low-energy side for  $\sigma^-$  polarization only. On the other hand, the indirect transition shows behavior more reminiscent of the single QW under high electric field: a sharp blueshift with density in  $\sigma^-$  polarization. This observation provides particularly clear confirmation that the change in the behavior seen for the single QW under applied electric field is caused by separating the vb hole from the 2DEG.

Recent theoretical work has predicted that when the vb hole is sufficiently far from the 2DEG, it will be favorable for the excitonic states responsible for recombination at  $\nu > 1$  to bind a finite number of extra spin reversals.<sup>3</sup> The resulting initial states can be interpreted as bound states of the vb hole with a (negatively charged) Skyrmion.<sup>3,12</sup> The calculations<sup>3</sup> predict that such states should only be bound

for large separations  $d > 1.4 l_B$ , where  $l_B$  is the magnetic length. At 5 T this corresponds to d > 160 Å, which is less than the separation between the centers of the wells of the two double-QW samples (275 or 225 Å). The calculations therefore show that it is reasonable to expect the Skyrmion-vb hole bound state to be observed in the double-QW structures. We suggest that the initial state responsible for the indirect PL of the double OW's at  $\nu > 1$  is of this kind: i.e., a bound state of a Skyrmion with a vb hole. Notice in Fig. 4(d), that for the indirect recombination in the double QW's at  $\nu > 1$ , both the  $\sigma^-$  and  $\sigma^+$  PL lines are asymmetric, being much broader on the low-energy side. This cannot be understood in terms of the recombination of the simplest excitonic state, as we observed for the single QW, which gives a broadening in  $\sigma^-$  only [Fig. 4(c)]. Rather, the observed shake-up in both  $\sigma^-$  and  $\sigma^+$  PL suggests that the initial state involves some additional spin reversal, as is the case for an initial state comprised of a Skyrmion bound to a vb hole. For such states, the radiative recombination in either polarization results in the creation of final-state spin waves, which one can expect to lead to lowenergy shake-up tails.

In conclusion, we have demonstrated Coulomb effects on the PL of quantum Hall systems close to the filling factor  $\nu = 1$ . The PL spectra are very sensitive to the spatial separation of the vb hole from the 2DEG. When the vb hole is close to the 2DEG, the low-energy initial states of the photo excited electron gas at filling fractions on both sides of  $\nu$ =1 are excitonic states, involving the binding of a vb hole and an  $e \downarrow$  electron. As the separation between hole and 2DEG is increased, a spectral discontinuity appears. We interpret this as a change of the  $\nu < 1$  ground state to be nonexcitonic. Our systematic investigation of the variation of the discontinuity provides a consistent interpretation of previously observed PL anomalies around  $\nu = 1.^{5,9-11,13}$  For the very large separations of the double-QW structure, we observe spin-wave shake-up tails in both polarizations of the excitonic recombination. This is experimental evidence for the formation of excitonic states involving the binding of the vb hole with a Skyrmion.

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