Negatively charged excitons in coupled double quantum wells

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We study magneto-optical spectra of neutral and negatively charged excitons (X^-) in remotely doped double quantum wells with coupled conduction bands. We observe photoluminescence due to the recombination of X^- leaving an excess electron in the adjacent quantum well (QW), which anticrosses the single-QW X^- line with applied gate bias. We see also the inter-QW X^- comprising an electron in either well. [S0163-1829(97)01703-7]

The semiconductor analogs of the negative hydrogen ion, the negatively charged exciton (X^-),[1–6] and donor center (D^-),[7] provide ideal systems for studying electron-electron interactions. Recent experiments have also demonstrated the existence of the positively charged exciton (X^+).[8,9] Interest in these three particle bound complexes has been rekindled by the large increase in their binding energy caused by confinement in a quantum well (QW). The binding energy is further increased by the application of a magnetic field, leading to the stabilization of the excited (triplet) state of X^- which has a symmetric spin wave function upon interchange of the electrons.[10,9] We present here the first report of X^- in double QW structures, in which the electron wave functions in neighboring wells are sufficiently close to interact. We demonstrate behavior in the spectra due to the coupling of the *excess* electrons in the two QW's.

Photoluminescence (PL) spectroscopy has been used to study interwell coupling of neutral excitons (X) in both double QW's (Ref. 11) and superlattices.[12] In these studies spatially indirect recombination is observed between an electron and a hole in adjacent QW's, the energy of which can be varied by an electric field applied normal to the layers. At the applied electric field where the energy of the indirect X is close to that of the direct X, involving an electron and hole in the same QW, the two exciton wave functions hybridize. This produces an anticrossing of the exciton energies as a function of applied electric field, along with a sharing of the transition intensity.

The double QW structures studied here differ in that they are remotely doped so as to contain an excess of electrons whose density can be varied by means of depleting Schottky contacts. When the electron (e^-) density in either QW is of order $\sim 10^{10}$ cm^{-2}, PL is observed due to recombination of both neutral ($X \rightarrow photon$) and negatively charged ($X^- \rightarrow photon + e^-$) excitons. Our spectra display inter-QW states of X^-, in addition to the previously observed anticrossing of X. However, more prominent is the coupling of the final, single-particle e^- state of the X^- recombination. This resonance in the *final* state of the recombination is only possible for X^- and not X. We show how analysis of the PL energies and intensities yields a direct measurement of the coupling energy of the e^- subbands in the two QW's, in addition to their occupation densities.

We studied a series of GaAs/Al_xGa_{1-x}As double QW's, grown by molecular beam epitaxy on (100)-oriented, semi-insulating GaAs substrates. The growth sequence for each was 1 μm GaAs, 1 μm $Al_{0.33}Ga_{0.67}$As, 0.5 μm GaAs(25 Å)/$Al_{0.33}Ga_{0.67}$As(25 Å) superlattice, GaAs back QW, $Al_{0.33}Ga_{0.67}$As barrier, GaAs front QW, 600 Å undoped $Al_{0.33}Ga_{0.67}$As spacer, 2000 Å $Al_{0.33}Ga_{0.67}$As Si doped (10^{17} cm^{-3}), and 170 Å GaAs cap. For sample 1 the dimensions of the (back QW/barrier/front QW) layers were (300 Å/25 Å/200 Å); for sample 2 (250 Å/25 Å/200 Å); sample 3 (200 Å/25 Å/200 Å); and sample 4 (200 Å/25 Å/300 Å). Sample 5 was a single-QW control structure incorporating just a 300 Å QW. The samples display exciton linewidths of typically 0.3 meV. The wafers were processed into mesas, with Ohmic contacts to the QW layers, a semitransparent NiCr Schottky gate on the top surface and an additional contact to the back of the substrate. Although each showed qualitatively similar behavior, we present spectra taken for sample 1, for which their was no spectral overlap of the contributions of the back and front QW's.

Figure 1(a) shows the band profiles of sample 1. Since only the upper $Al_{0.33}Ga_{0.67}$As barrier layer is intentionally doped, the majority of the excess e^-'s are in the front QW under unbiased conditions. The back QW does, however, contain a small e^- density by virtue of its smaller confinement energy than the front QW. This layer structure is thus ideal for studying X^- in the back QW, whose observation requires a small excess e^- density ($\sim 10^{10}$ cm^{-2}). The e^- density in the front (200 Å) QW can be varied by applying a voltage (V_{fg}) between the front gate and the QW layers. Similarly we bias (V_{bg}) the substrate with respect to the QW layers in order to vary the back QW density predominantly.

PL spectra taken on sample 1 with a fixed back gate bias and different front gate biases are plotted in Fig. 2(a), while Figs. 2(b) and 2(c) show the variation of the PL energies and peak heights with V_{fg}. The PL originating from the front 200 Å QW is qualitatively similar to that observed for the single-QW structure (sample 5) and that reported previously by us in Refs. 2,13. For V_{fg} = 0.0 V, the PL from the 200 Å QW consists of a broad band, reflecting the spread of occu-

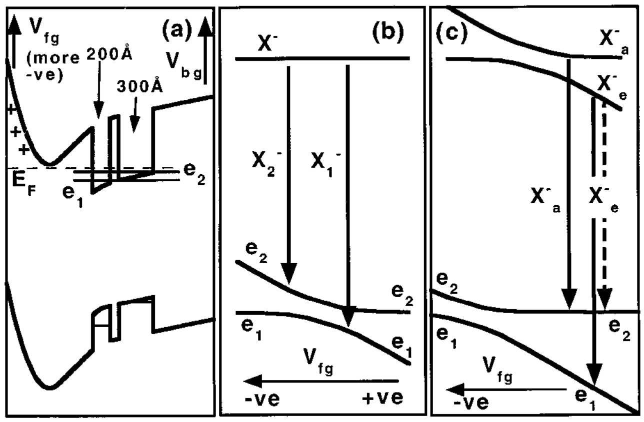

FIG. 1. (a) Schematic of the valence and conduction band profiles of sample 1. An increasingly negative bias applied to the front (back) gate raises the potentials at the front (back) of the DQW structure. (b) Schematic of possible recombination channels for X^- in the back QW, according to $X^- \rightarrow photon + e^-$, as a function of front gate bias, close to energetic resonance of the e^- levels in neighboring wells. (c) Possible X^- PL, close to resonance of the inter- (X_e^-) and intra-QW (X_a^-) excitons.

pied e^- states and allowing the density of the 200 Å QW to be estimated[13] as 1.2×10^{11} cm^{-2}. A negative bias applied to the front gate reduces the density in the front QW, producing the blueshift and narrowing of its PL band apparent in Figs. 2(a) and 2(b).[14] Around -0.40 V, the PL band narrows to a sharp peak, which is attributed to the X^- recombination in the 200 Å QW and the neutral exciton (X) line appears to higher energy. Thus, the PL of the front QW shows a similar density dependence to that of a single-QW and thereby provides a reliable measure of its e^- density.

More interesting is the dependence of the PL of the back QW on the front gate bias. The back gate bias has been fixed in Fig. 2(a) to provide an e^- density for which both X and X^- can be observed for the 300 Å QW. Notice in Fig. 2(a) that the X^- second e^- binding energy, indicated by its spectral separation from the X line, is apparently independent of the e^- density in the front QW; the splitting is (0.90 $\pm$0.02) and (0.88$\pm$0.02) meV in the V_{fg} = 0.00 and -0.70 V spectra, for which the front QW e^- densities are $\sim$1.2 and $\sim 0.0\times10^{11}$ cm^{-2}, respectively. Clearly then the X^- second e^- binding energy is quite insensitive to the charge in the other QW.

There has been some debate in the literature as to whether X^- is free in the QW plane or localized by the potential fluctuations caused by the ionized donors in the barriers.[1,3,6] Since the spacer layers are relatively thick, and the doping concentrations low, the effect of the donor ions is expected to be small in our samples.[15] However, we can reduce the amplitude of the fluctuations yet further by introducing a large e^- density into the front QW which will screen the donor potential in the plane of the back QW. The fact that we still observe X^- in the back QW after adding a large electron density to the front QW, and indeed with negligible change in binding energy, suggests that the donor potential is certainly not a prerequisite for X^- formation. X^- may exist as a free species.

The PL spectra of the back QW do, however, show some unusual behavior over the range of voltages where the e^- states in adjacent wells are close to resonance. Notice that

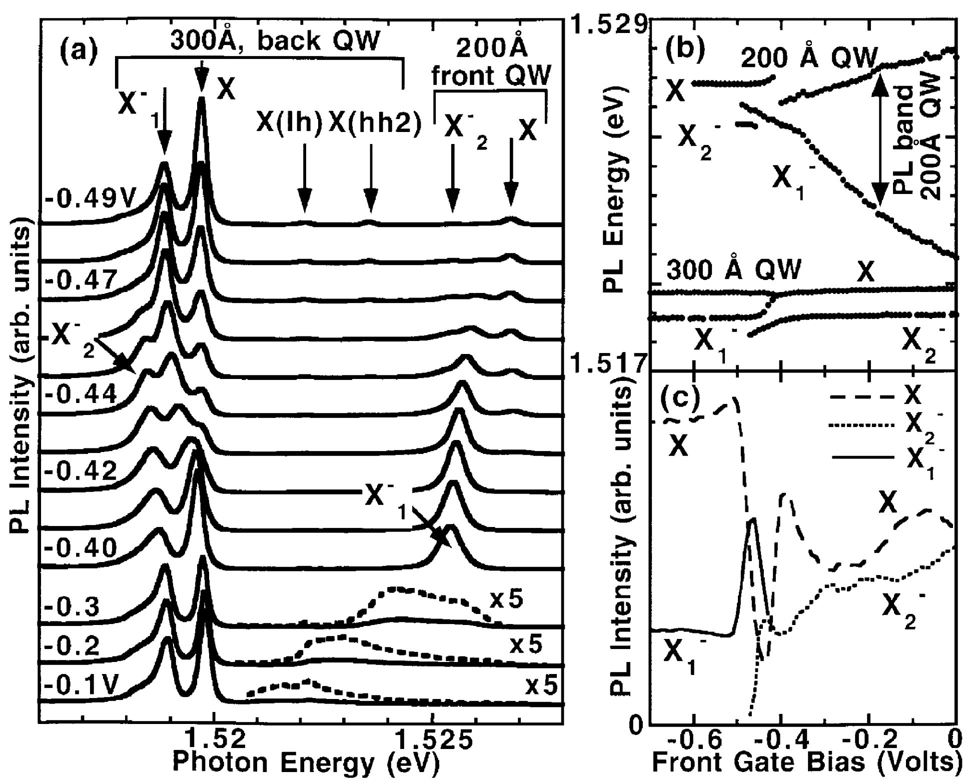

FIG. 2. (a) PL spectra recorded on sample 1 with the different front gate biases indicated, fixed back gate bias, and a sample temperature of 2.0 K. Evolution of the PL peak energies (b) and intensities (c) of the 300 Å QW with front gate bias.

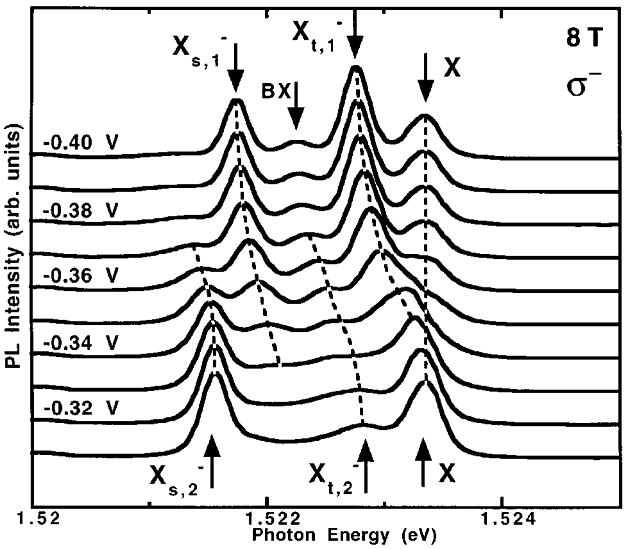

FIG. 3. Front gate bias dependent PL spectra measured for the 300 Å QW of sample 1 with an applied magnetic field of 8 T, fixed back gate bias, and a sample temperature of 2.0 K. Notice the anticrossing of both the singlet (X_s^-) and triplet (X_t^-) states of X^-. The dashed lines are guides to the eye. Peak BX is due to a bound neutral exciton and is also observed in single-QW's at very low e^- densities (Ref. 10).

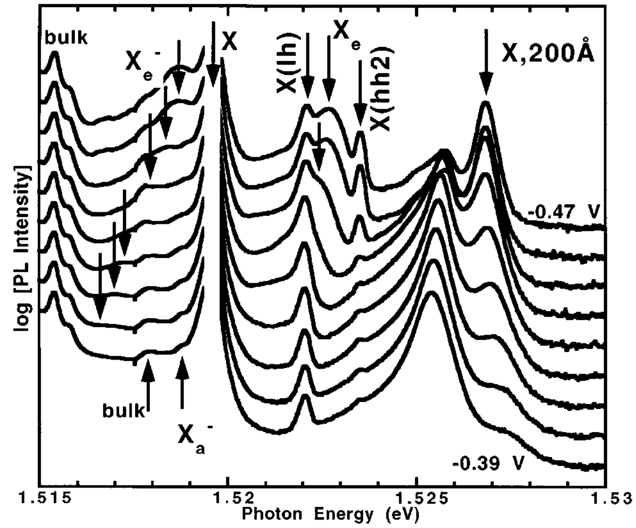

FIG. 4. PL spectra (on log scale) recorded on sample 1 at different front gate biases, and a more negative back gate bias than in Fig. 2. For this back gate bias the e^- density of the back QW is small but finite. Notice the weak peak due to the inter-QW X^- transition, X_e^- .

three peaks can be observed in the band edge region (1.518–1.5205 eV) of the 300 Å QW for $-0.42 \geqslant V_{\mathrm{fg}} \geqslant -0.46$ V. The dependence of the two lower energy peaks on V_{fg} and V_{bg} demonstrates that both require the presence of an excess e^-. This, along with the nature of the anticrossing behavior described below, leads us to ascribe the two lower energy peaks to X^- recombination leaving an excess electron in either of the two coupled e^- subbands.

Figure 2(b) demonstrates the apparent anticrossing of the two lower energy peaks as V_{fg} is varied. The anticrossing is unusual in that it occurs in the *final* state, i.e., the single electron state that is left behind when one of the electrons and the hole in X^- recombine according to $X^- \rightarrow photon + e^-$. Figure 1(b) illustrates the anticrossing of the e^- levels which occurs when an increasingly negative bias is applied to the front gate, thereby raising the potential on the left-hand side of Fig. 1(a). At the gate biases where the e^- levels in the two QW's are close to resonance, the surviving e^- can be left in either of the two hybridized states after recombination, producing two possible recombination energies and therefore two X^- peaks in the PL spectra. We label these two peaks X_1^- and X_2^- according to whether the e^- is left in the first or second subband, themselves labeled in order of increasing energy. The minimum energy separation fitted to the two X^- peaks gives a direct measure of the coupling energy of the single e^- states, of (0.55 ± 0.05) meV for this e^- density, close to the value expected from a self-consistent solution of the Poisson-Schrödinger equations.[13]

The hybridization of the e^- levels in the two QW's also produces an anticrossing in the X^- transition of the front 200 Å QW. Indeed two X^- peaks from the 200 Å QW can just be discerned in some of the spectra in Fig. 2(a). Notice in Fig. 2(b) that, consistent with our assignments, the energy shift of the X^- transition with V_{fg} has opposite sign for the front and back QW's. The anticrossing for the 200 Å QW is most readily observed in spectra (not shown) taken for different V_{bg} and fixed V_{fg}, for which it is qualitatively similar to that seen in Figs. 2(a) and 2(b) for the 300 Å QW as a function of V_{fg}.

When a magnetic field is applied normal to a single QW, another X^- peak emerges below the X line, due to an excited state where the spin wave function is symmetric upon interchange of the two electrons; the spin-triplet states.[10,9] In the present study of double QW's, the coupling of the e^- states is manifested as an anticrossing of both the ground, singlet state (X_s^-) and excited, triplet state (X_t^-) at finite magnetic field. Figure 3 plots PL spectra taken at different V_{fg} and fixed V_{bg} under an applied magnetic field of 8 T. The emitted light is polarized in σ^- polarization. Notice the anticrossing of both X_s^- and X_t^- near -0.36 V, with similar gap energies of $\sim$ (0.50 ± 0.05) meV, respectively. Similar anticrossing behavior is observed in σ^+ polarization.

The intensities of the two X^- peaks depend upon the overlap of the initial and final states for each transition, as well as the population of X^-, and hence the density of excess e^-'s, in the *back* QW. For $V_{\mathrm{fg}} \geqslant -0.44$ V, the e_2^- wave function lies predominantly in the back QW and hence has larger overlap with the X^- wave function than e_1^- , producing the strong X_2^- PL peak for the back, 300 Å QW. Close to resonance, the amplitude in the back QW of e_2^- declines, while that of e_1^- grows, leading to the transfer of transition strength to the X_1^- peak seen in Fig. 2.

The excitonic intensities are also remarkably sensitive to intra-QW many-body e^--e^- interactions, through their dependence on the population of X^- (and hence e^-) in the back QW. Notice the sharp maximum in the intensity of X_1^- near -0.46 V in Fig. 2(c) and the dip in the strength of X. These features are apparent over a wide range of different back gate biases. They can be explained by an increase in the e^- density of the back QW upon depletion of the front QW, due to the difference in the many-body exchange-correlation potential in the two QW's.[16] In the random phase approximation, an excess e^- density (N) creates an exchange-correlation potential which changes the conduction band energy by (for bulk) $V_{xc} \propto -N^{1/3}$. Close to depletion of the front QW, there is a sharp reduction in the magnitude of its exchange-correlation potential, producing a larger drop in its e^- density than would occur without this many-body effect. Since the total excess e^- density is determined by the device capacitance and the applied voltages, this has the effect of increasing the density in the back QW (by around 10^{10} cm^{-2} for this structure). This increase in e^- density raises the population of X^- in the back QW, while lowering that of X correspondingly. This produces the sharp strengthening of the X_1^- transition, whose optical matrix element dominates over that of X_2^- at this bias, as well as the weaker maximum in the X_2^- intensity and the large dip in strength of X.

So far we have discussed only coupling of the final, e^- state of the X^- recombination, $X^- \rightarrow photon + e^-$. In Fig. 2 the two X^- transitions of the 300 Å QW are from the same initial (intra-QW) excitonic state, consisting essentially of two electrons and one hole in the back QW. One could also envisage an *inter*-QW X^- consisting of one electron and hole in one QW and the second e^- in the other QW. This will couple to the intra-QW X^- as a function of gate bias, as shown schematically in Fig. 1(c). The resonance of the X^- states is shifted to less $-$ve V_{fg} (or, alternatively, more $-$ve V_{bg}) from that of the e^- states, because the second e^- binding energy of the intra-QW X^- is larger than that of the inter-QW X^-.

Notice in Fig. 1(c) that the inter-QW X^- can relax to either of the e^- subbands producing two possible transitions. We do not observe the inter-QW X^- transition leaving the excess e^- in the other QW to the hole. This is because the binding energy of the excess e^- in the front QW to the exciton in the back QW will be small and the transition is therefore not resolved from the neutral exciton of the back QW. We do, however, see the inter-QW X^- PL leaving an excess e^- in the same QW as the hole [dashed in Fig. 1(c)], as discussed below. Although one could expect the latter to be the weaker of the two inter-QW X^- transitions, its observation is aided by the fact that it is shifted from X.

Figure 4 plots PL spectra taken with different V_{fg} and a fixed V_{bg}, for which the e^- density in the back QW is lower than in Fig. 2 but still finite. Measurements taken at more $-$ve V_{bg}, for which the back QW is depleted, demonstrate that the peak marked X_e in Fig. 4 is due to the inter-QW neutral exciton. (At these more $-$ve V_{bg}, the X_e peak blueshifts with increasingly $-$ve V_{fg} and anticrosses the intra-QW X.) The X_e peak weakens very rapidly with excess e^- density in the back QW, explaining why it is only observed at the most $-$ve V_{fg} in Fig. 4. We suppose this is because the lifetime of a hole in the 300 Å QW decreases rapidly with increasing e^- density,[6] thereby quenching the much slower indirect recombination.

Notice the weak peak, marked X_e^- in Fig. 4, which lies around 4.2 meV to a lower energy than X_e and which also blueshifts with increasingly $-$ve V_{fg}. This feature, which is only observed for a small e^- density in the back QW, is ascribed to recombination of the inter-QW X^-, leaving an e^- in the back QW [dashed line in Fig. 1(c)]. This is suggested by the fact that the X_e^- peak shifts to higher energy with increasingly $-$ve V_{fg}, i.e., the opposite behavior to that discussed previously (Fig. 2), indicating that the inter-QW coupling is in the initial [Fig. 1(c)], rather than final [Fig. 1(b)] state of the recombination. Also consistent with the X_e^- assignment is its relatively large splitting from X_e of 4.2 meV. One expects a large binding for the ''second'' e^-, since it is placed in the same QW as the hole and the opposite one to the ''first'' e^- in X_e.

In conclusion, the PL spectra of X^- in double QW's show an anticrossing due to mixing of excess e^- states of the two QW's. In a magnetic field both singlet and triplet states of X^- undergo this splitting. The minimum splitting of the PL lines gives a direct measure of the coupling energy of the e^-'s. On the other hand, the excitonic intensities are very sensitive to e^- transfer between the QW's due to their differing exchange-correlation potentials. Away from resonance, the X^- second electron binding energy is not effected by a large e^- density in the other well, demonstrating that it is insensitive to the donor localization potential. The inter-QW X_e^- comprising an e^- in either well, is also observed at the lowest excess e^- densities.

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