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Effect of electric fields on excitons in a coupled double-quantum-well structure

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We present a detailed experimental study of the influence of electric fields on exciton states in a $GaAs/Al_xGa_{1-x}As$ double-quantum-well structure. Both intrawell and interwell exciton transitions were observed. The coupling of electronic levels in the two quantum wells leads to an enhancement of the quantum-confined Stark effect (as much as 5 times that of the single-quantum-well case). From the measured exciton transitions, splittings of the quantum confined levels in a coupled double-quantum-well structure were derived without recourse to a theoretical model.

The quantum-confined Stark effect (QCSE) in semiconductor quantum-well (QW) structures has recently attracted a great deal of interest.[1-4] As an electric field is applied perpendicular to the QW layers, spatial confinement of electronic wave functions prevents the field ionization of two-dimensional (2D) excitons so that 2D exciton energies and oscillator strengths can be greatly modified. Attempts to enhance the performance of electro-optical devices based on this effect,[5] in particular to increase the exciton energy shift for a given applied field, have involved more complex structures, notably the coupled double quantum well (CDQW).[6-10] This structure consists of a pair of QW's separated by a barrier narrow enough that considerable overlap occurs between the electronic wave functions in the two wells. It has been shown theoretically that the combined effect of *coupling* between the two QW's and the QCSE of the individual QW's enhances the electric-field-induced exciton energy shift.[6] In an isolated QW, the effect of the electric field is to reduce conduction- and valence-band energies and 2D exciton binding energies.[2,3] However, in a CDQW, the split levels of each of the coupled electronic states move in opposite directions as a function of applied electric field, leading to several exciton transitions which shift rapidly to lower or higher energies as the electric field is increased. These transitions are essentially interwell-like (spatially indirect), in that they are associated with recombinations of electrons concentrated mostly in one well and holes concentrated mostly in the adjacent well. In this Rapid Communication we present a detailed study of optical transitions in a CDQW under the influence of an external electric field which clearly demonstrates some of the unique properties of the CDQW. We determine for the first time, directly from experiment, splitting energies of levels in a coupled double-quantum-well structure under flat-band conditions without recourse to a theoretical model. Our experimental results are in good agreement with theory[6] and clearly demonstrate the predicted enhanced quantum-confined Stark effect of spatially indirect exciton transitions in CDQW's.

Coupled electronic states in the conduction and valence bands of a CDQW are shown schematically in Fig. 1. In the symmetric well case (i.e., the two QW's are identical), the coupled electronic states have well-defined symmetries in the absence of external perturbations such as electric fields (we designate this the flat-band condition, $E=0$). In this situation, only transitions between electron and hole states of the same symmetry are allowed; transitions between states of opposite symmetries have zero net transition probabilities and therefore are forbidden. This is illustrated on the left-hand side of Fig. 1, in which symmetry-allowed transitions are denoted by solid lines (1, 3, 6, 8) and symmetry-forbidden transitions by dashed lines (2, 4, 5, 7). The energies of symmetric ($-$) and antisymmetric ($+$) states are given by

$$E_{\pm i}=E_{0i}\pm\Delta_i/2, \quad (1)$$

where i=electron (e), light-hole (LH), or heavy-hole (HH); E_{0i} is the energy of the corresponding state in a single quantum well (SQW) and Δ_i is the level splitting produced in the CDQW due to coupling.

When an electric field is applied to a CDQW, wave function symmetries are distorted, and all transitions be-

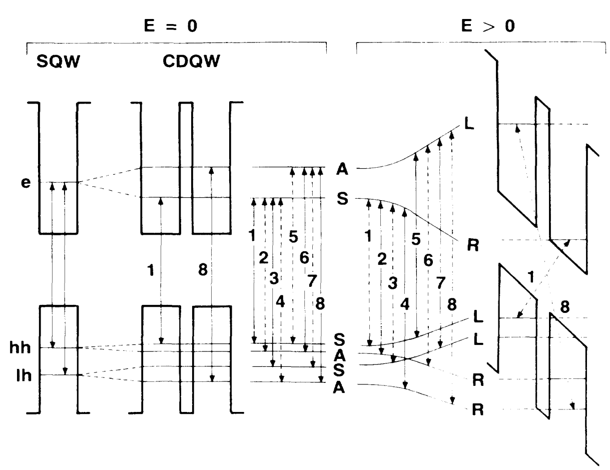

FIG. 1. Schematic diagrams of the energy levels of a single quantum well (SQW, left), and a symmetrical coupled double quantum well (CDQW) under flat-band conditions (middle), and in the presence of external dc electric fields (right). S and A correspond to symmetric and antisymmetric coupled states under flat-band conditions.

come allowed. The energies of states in a CDQW as a function of applied field E can be written as

$$E_{\pm i}(E)=E_{0i}(E)+\Sigma_{\pm i}(E) , \tag{2}$$

where $E_{0i}(E)$ is the energy of the corresponding state in a SQW, and $\Sigma_{\pm i}(E)$ is the level splitting produced in the CDQW due to coupling modified by the electric field effect. The + and − signs indicate high- and low-energy states. At very large applied fields, the splitting will be anisotropic ($|\Sigma_{+i}|\neq|\Sigma_{-i}|$) as a result of the large distortion of the coupled-state wave functions away from the flat-band condition.[6] Furthermore, tilting of the potential well produces a redistribution of the wave functions (Fig. 1, right). The low-energy coupled states (symmetric states under the flat-band condition) of the valence band concentrate mostly in the left well (designated L in Fig. 1), while the low-energy coupled level of the conduction band resides primarily in the right well (designated R). A similar anisotropic wave-function distribution occurs for the high-energy coupled levels (antisymmetric states under the flat-band condition).

As shown schematically in Fig. 1 (right, $E>0$), intrawell-like transitions in either the R (2 and 4) or L (5 and 7) QW's experience the regular QCSE and are weakly affected by the coupling (since the energy shifts of the electron and hole states nearly cancel each other); on the other hand, energies of interwell-like transitions undergo much more rapid decreases (1 and 3) or increases (6 and 8) with increasing applied field since these transitions couple electron and hole states either moving toward or away from each other. It is interesting to note that symmetry-allowed transitions under flat-band conditions become interwell-like transitions under an applied field (dashed lines), whereas symmetry-forbidden transitions become intrawell-like (solid lines). Under large fields, the wave-function overlap of electron and hole states localized in different wells is significantly reduced. Thus, interwell-like transitions are expected to weaken, while intrawell-like transitions approach those observed in a SQW.

Low-temperature photoluminescence (PL), photoluminescence excitation (PLE),[11] and photocurrent (PC)[12] spectroscopies were employed to study these transitions in a CDQW p-i-n structure. The samples were grown by molecular beam epitaxy on an n^+-type GaAs buffer and consisted of a single pair of 7.5-nm GaAs quantum wells (L_Z) separated by a 1.8-nm $Al_{0.27}Ga_{0.73}As$ barrier (L_B) surrounded by two 85-nm outer undoped $Al_{0.27}Ga_{0.73}As$ barriers, and a 20-nm p^+-type GaAs cladding layer on top, all grown on an n^+-type GaAs substrate. A Ni/Au-Ge/Au metallization, with 3-min 450 °C sintering, was employed to obtain an Ohmic contact to the n^+ substrate while Au-5 at.% Zn metallization provided a near-Ohmic contact to the p^+ layer. The samples, mounted on a sapphire holder, were placed in an exchange-gas liquid-He cryostat and excited with a tunable dye laser pumped with a Kr^+ ion laser.

In Fig. 2 we present typical PLE spectra as a function of bias voltage. All of the observed peaks can be explained by considering only the ground exciton states of the CDQW system. The bottom spectrum (taken with a bias of 1.6 V which corresponds to the flat-band condition since the external electric field exactly cancels the builtin internal field of the p-i-n diode), exhibited four peaks which correspond to the allowed 1, 3, 6, and 8 exciton transitions (Fig. 1, $E=0$). When a reverse bias was applied (by reducing the applied voltage), additional peaks were observed. These peaks correspond to transitions 2, 4, 5, and 7, which are symmetry-forbidden under flat-band conditions. With a net applied field small enough that the Stark effect was negligible but large enough that most of the transitions were observable, we were able to derive all

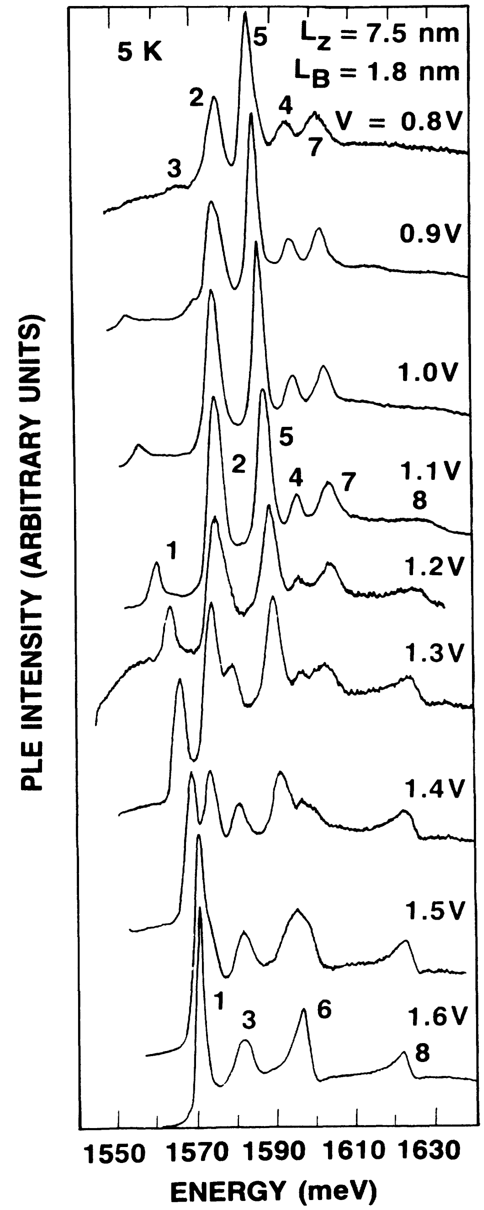

FIG. 2. Photoluminescence excitation spectra of a coupled double quantum well (CDQW) under various bias voltages. The bottom spectrum corresponds to the flat-band condition (no net electric field on the CDQW), and, as the bias voltage decreases, the net applied electric field increases. The peaks are labeled according to the notation of Fig. 1.

of the splittings due to interwell coupling using measured exciton energies [Eq. (1)]. Level splittings of the ground-state electron Δ_e, heavy-hole Δ_h, and light-hole Δ_l states in our CDQW structure were 22.1, 3.9, and 15.5 meV, respectively. With these values and the HH and LH exciton energies (transitions 1 and 3 in Fig. 1), we derived the HH exciton energy (1.582 eV) and HH-LH splitting (18.8 meV) in the QW's in the absence of coupling by employing simple arithmetic manipulation. Agreement with values measured in a reference single-quantum-well sample grown under identical conditions (1.584 eV and 19 meV, respectively) was very good.

The lowest-energy peak, an interwell-like transition labeled 1 (due to a heavy-hole transition, Fig. 1), and peak 3, a light-hole interwell-like transition, shifted rapidly to lower energies and lost PL intensity as the applied field was increased. A new, slightly higher-energy peak, labeled 2, quickly became dominant at higher fields. This new transition is due to a heavy-hole intrawell-like transition, which is forbidden under flat-band conditions (Fig. 1). It was observed to cross peak 3 at a bias voltage of about 1.1 V ($E \approx 2.7\times10^4$ V/cm), which corresponds to the crossing between *R* HH and *L* LH in Fig. 1. The highest-energy peak, labeled 8, is attributed to a interwell-like light-hole transition. With increasing field its PL intensity dropped rapidly while its energy increased, in contrast to the behavior of peaks 1 and 3 (which connect levels which move *toward* each other with increasing electric field). On the other hand, transition 8 (and also 6) connect levels moving *apart* with increasing field. The behavior of peak 6 is more difficult to follow experimentally since it quickly intersects a number of intrawell transitions and its PL intensity decreases rapidly with increasing field.

At a bias voltage of 0.8 V ($E \approx 4\times10^4$ V/cm, top spectrum, Fig. 2), the PLE spectrum was dominated by four peaks due to intrawell-like transitions of the *R* (2 and 4) and *L* (5 and 7) QW's. Interwell-like transitions were much weaker and could barely be detected. The PC spectrum (not shown) exhibited such a strong signal for the interwell-like transitions, that we were able to determine the exciton energy of peak 1. Intrawell-like transitions, at this applied field and beyond, behaved similarly to the SQW case (the regular QCSE). Transitions involving heavy-hole states (2 and 5) were stronger than those connected with light-hole states (4 and 7). With further increases in electric field, PL intensities of all the transitions became weaker (not shown) due to the field-induced reduction of the lifetimes of the states (the electron and holes were no longer completely confined to the QW's). At bias voltages less than 0.7 V ($E > 4.6\times10^4$ V/cm), the PC spectrum was of higher quality than the PLE spectrum (a higher signal-to-noise ratio) and usable spectra were measured down to a bias voltage of -1.4 V.

In Fig. 3 we summarize the observed exciton transition energies as a function of bias voltage (applied electric field). Intrawell-like transition energies are presented as open circles, while interwell-like transition energies are shown in closed circles. The dependence of interwell exciton energies on bias voltage is quadratic at low fields and becomes linear at higher fields in agreement with theory.[6]

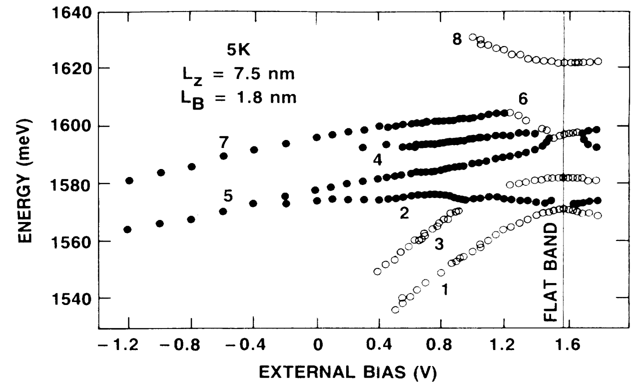

FIG. 3. Energies of exciton peaks as a function of applied external voltage measured using photoluminescence excitation and photocurrent spectroscopies. The open circles are interwell-like exciton transitions and the closed circles are intrawell-like exciton transitions (see text).

It is interesting to note that the electric field dependence of peak 8 is of the same magnitude (but of opposite sign) to that of peak 3. This is evidence that the splitting energies of the two electron and light-hole electronic states are approximately equal at low fields. The energies of all the intrawell-like transitions, which became dominant at higher applied fields, had nearly linear relationships with the field strength. The anomalous behavior of transition 2 at low applied fields is related to its crossing with transition 3. At higher fields, transitions 2 and 4 have very similar characteristics. At still higher fields, peaks 2 and 5 (connected with heavy-hole levels) and peaks 4 and 7 (connected with light-hole levels) converged and at $V < -0.4$ V ($E > 1\times10^5$ V/cm), only intrawell transitions of the left well (5 and 7) remained. This is consistent with the theoretical model in that at large fields the two wells are nearly decoupled and the states of the left well have a relatively longer lifetime than those of the right well.[6] At the highest fields studied, the oscillator strength of the light-hole transition (7) finally disappeared and only the heavy-hole transition (5) remained. This occurred because the heavy-hole level, which is more strongly bound within the well, possesses a relatively longer lifetime.

In summary, we have presented a detailed optical study of the influence of external electric fields on exciton transitions in a coupled double quantum well. The experimental data provide clear evidence of features unique to this system. In particular, the electric-field-induced energy shifts of transitions 1, 3, and 8 can only be understood within a coupled well model. Under large electric fields, the behavior of intrawell-like transitions (5, 7, 4, and 2) is analogous to that observed in SQW's. It is important to note that because of the coupling between the two QW's, the quantum-confined Stark effects of some of the exciton states are considerably enhanced. This is best illustrated by the observation that, in this sample, the field-induced shifts of the coupled exciton states (i.e., transitions 1, 3, or 8) are as much as 5 times larger than those of the single-quantum-well case (analogous, for example, to the behavior of intrawell-like transitions 5 and 7 at large fields). Potentially these large-field-induced shifts of exciton resonance energies, in particular transition 1, can be utilized in electro-optical devices to yield large extinction ratios. Since the oscillator strengths of these interwell-like exciton states weaken rapidly with electric fields they are best used in device configurations with long interactions lengths.

Note added. Since the present work was submitted for publication, Islam, Hillman, Miller, and Chemla[13] have reported similar findings for modes 1, 2, and 3 in a CDQW waveguide structure.

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