Brief Reports

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Transformation of spatially direct to spatially indirect excitons in coupled double quantum wells

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We report a detailed experimental study of the influence of electric fields on exciton lifetimes in a GaAs/$Al_xGa_{1-x}As$ coupled-double-quantum-well structure. The energy of the lowest-lying photoluminescent exciton peak was observed to decrease and its decay time was found to increase by over an order of magnitude as the internal electric field was increased. The increase in the decay time is attributed to the change of the exciton transition from spatially direct (i.e., taking place within a single well) to spatially indirect (connecting an electron level in one well with a hole level in the adjacent well).

The effect of an electric field on the optical properties of quantum-well (QW) structures has recently attracted a great deal of interest from both fundamental and practical points of view.[1–4] When an electric field is applied perpendicular to the layers of a QW structure, significant changes in the optical-absorption, reflection, and photoluminescence (PL) spectra can be observed and are attributed to modifications of the spatial confinement of the electron and hole wave functions.[5–7] A shift of the absorption edge caused by the quantum-confined Stark effect (QCSE) has been reported,[5,6] and applications of this effect have already been presented (e.g., high-speed optical modulators[8] and electrooptical bistable devices[9]). A similar shift to lower energy in the QCSE regime also occurs for the PL peak of the two-dimensional excitons in the QW.

Recently, an increasing amount of attention has been directed toward structures employing pairs of QW's, with interwell barriers sufficiently narrow that considerable overlap occurs between the electron and/or hole wave functions in the two wells. We will refer to such structures as coupled double quantum wells (CDQW's).[10–14] In an unperturbed system of two identical QW's separated by a thin penetrable barrier, degenerate single QW states split into symmetric and antisymmetric doublet states. In an isolated QW, the effect of the electric field is to reduce both conduction- and valence-band energies and exciton binding energies. In a CDQW, however, the split levels of each of the coupled electronic states move in opposite directions as a function of applied electric field. Transitions between these levels are either essentially interwell-like (spatially indirect) and associated with recombinations of electrons concentrated in one well and holes concentrated in the adjacent well, or intrawell-like (spatially direct). In the case of spatially indirect transition, the wave-function confinement will reduce the electron-hole spatial overlap, thus increasing the exciton radiative lifetime.

In a recent paper, we have studied the case of a symmetric CDQW structure, in which the two QW's were identical.[14] Low-temperature CW photoluminescence (PL), PL excitation (PLE) and photocurrent spectroscopies were employed to study the different transitions involved in a CDQW as a function of bias voltage. The salient features of our PLE results are the abundance of spectral lines, the positive and negative Stark shifts, and the crossings of some of the levels. The spectral richness results from the breakdown of parity, which usually limits the number of observable optical transitions in sym-

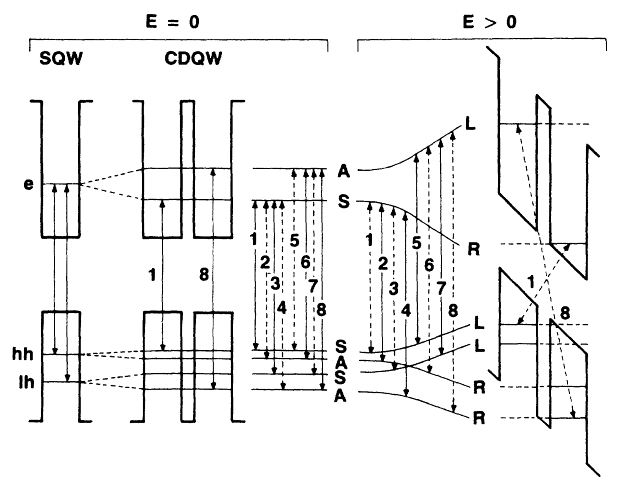

FIG. 1. Schematic diagram of energy levels and transitions in a CDQW under both flat-band and electric-field conditions.

metric QW's. In the following discussion we will refer to the schematic diagrams of the energy levels given in Fig. 1. In the absence of net internal electric fields, designated the flat-band condition, the coupled electronic states have well-defined symmetries. In this situation, only transitions between electron and hole states of the same symmetry are allowed; transitions between states of opposite symmetry have zero net transition probabilities. When the electric field is applied to the CDQW, wave function symmetries are distorted, selection rules are relaxed and all transitions become allowed. As shown in Fig. 1, the symmetry-allowed transitions under flat-band conditions become interwell-like transitions under an applied field, whereas symmetry-forbidden transitions become intrawell-like. Time-resolved spectroscopy should be of significant utility here since the decay of an interwell-like transition is expected to be much longer than that from an intrawell-like transition. In this paper, we examine the conditions under which this increase in lifetime exists. The luminescence decay time of these transitions was measured for various applied voltages in order to examine the effect of the electric field on the dynamics of excitons on a CDQW.

Time-resolved PL was used to study the lowest exciton state in a CDQW *p-i-n* structure as a function of applied bias voltage. The samples were grown by molecular-beam-epitaxy (MBE) on an n^+ GaAs buffer and consisted of a single pair of 7.5-nm GaAs QW's (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^+ GaAs contact layer on top, all grown on an n^+ GaAs substrate. The samples, mounted on a sapphire holder, were placed in an immersion-liquid-He cryostat and held at a temperature of 1.8 K. The excitation source used for time-resolved PL experiments was a mode-locked Argon-ion laser synchronously pumping a cavity-dumped dye laser. The excitation wavelength was 660 nm at an average power density of 30 mW/cm^2. The pulse width was less than 30 ps and the repetition rate was 4 MHz. The luminescence was dispersed by a $\frac{3}{4}$-m double spectrometer and detected by a cooled ITT-F4128F photomultiplier tube operated in the photon counting mode. The luminescence decay curves were obtained with the usual time-to-amplitude converter and pulse-height analyzer combination. The total system response to the dye-laser excitation pulses had a decay-time constant of 70 ps. By operating the system under computer control, eight spectra, corresponding to different time windows after the laser pulses, could be collected simultaneously.

As mentioned previously, the effect of electric fields is to transform a strong allowed transition into a weak one.[14] Therefore, transition 1, which is the main transition observed in PL and symmetry-allowed under flat-band conditions, becomes a transition between electrons primarily in one well and holes in the adjacent well when an electric field is applied to the CDQW. Since this transition is essentially indirect in real space (i.e., the electron and hole wave function overlap is significantly reduced), its oscillator strength declines rapidly with increasing field and an increase in lifetime is expected. On the other hand, transition 2, which is symmetry-forbidden under flat-band, becomes allowed and remains spatially direct under net internal electric field and thus one would expect a fast transient behavior. A series of time-resolved PL spectra of the CDQW sample under an external bias voltage of 1 V is shown in Fig. 2. Immediately after the excitation pulse (0–0.33-ns time window; top curve), two emission peaks are clearly visible: transition 1 at 1565.8 meV and transition 2 at 1574.1 meV. At a later time (0.65–0.98-ns time window; bottom curve) emission from transition 2 has faded. The change in the relative intensities of these two peaks for the different time windows is indicative of the different lifetimes for transitions 1 and 2. This is more readily seen in Fig. 3, which presents luminescent decay curves for peak 1 [under various bias conditions (a)–(e)] and peak 2 (f). Note the dramatic, tenfold increase in PL lifetimes for peak 1 as the internal electric field on the CDQW is increased from 0 (flat-band condition). The flat-band condition, at which the external voltage exactly cancels the built-in internal electric field of the *p-i-n* diode, was achieved for an external bias of 1.5 V. One should keep in mind, for the CDQW *p-i-n* structure used in this study, that as the external bias voltage decreases below 1.5 V, the net internal electric field increases. These results are summarized in Fig. 4 where the observed exciton transition energies and PL decay times are plotted as a function of forward bias voltage.

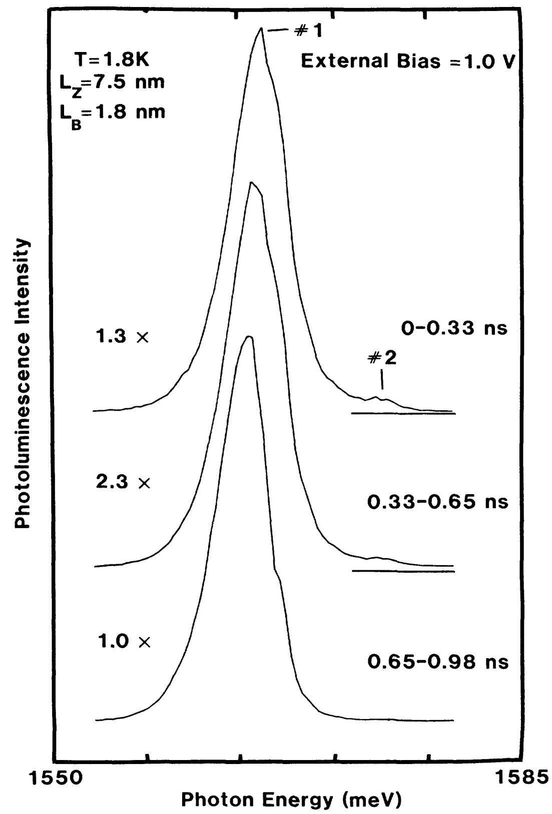

FIG. 2. A series of 1.8-K PL spectra showing the evolution of PL peaks as a function of time after pulse excitation. The time windows are indicated in the figure. An external forward bias voltage of 1 V was applied to the sample. The peaks are labeled according to the notation of Fig. 1.

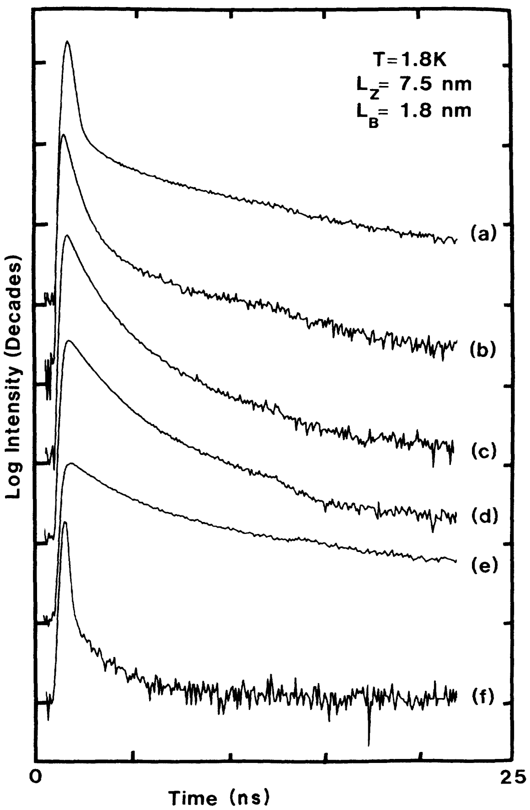

FIG. 3. The luminescence decay of exciton peaks PL [curves (a)–(e) correspond to transition 1 and curve (f) corresponds to transition 2] under various bias voltages. Each of the tick marks on the vertical scale represents one decade of intensity. The transient curves represent decays having time constants of (a) $V=1.5$ V, $\tau=0.19$ ns; (b) $V=1.0$ V, $\tau=0.31$ ns; (c) $V=0.75$ V, $\tau=0.61$ ns; (d) $V=0.50$ V, $\tau=1.0$ ns; (e) $V=0.375$ V, $\tau=1.91$ ns; (f) $V=1.0$ V, $\tau=0.10$ ns (transition no. 2).

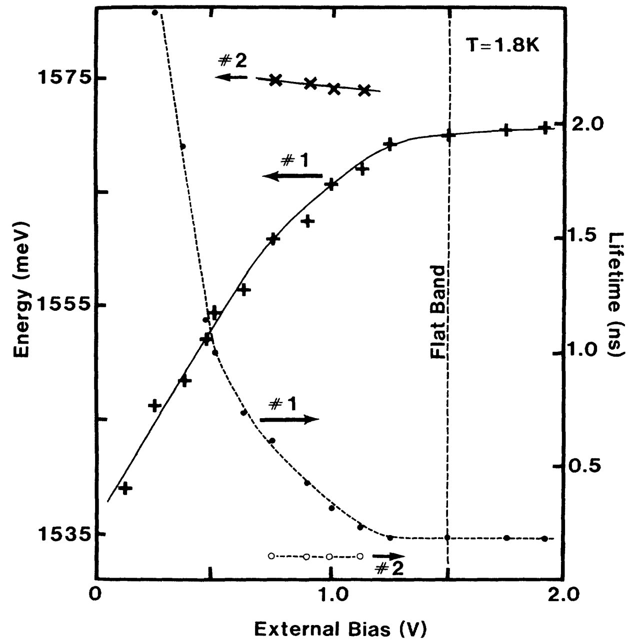

FIG. 4. Energies (crosses) and lifetimes (solid circles) of exciton peaks as a function of forward bias voltage measured using time-resolved photoluminescence spectroscopy. Solid and dashed lines through the data points are guides to the eye. The flat-band condition corresponds to the case where there is no net electric field on the CDQW. Because of the *p-i-n* structure used in this study, the net internal electric field increases as the external bias voltage decreases. The identities of transitions No. 1 and No. 2 are discussed in the text.

As one can see, the dependence of the energy of transition 1 on bias voltage is quadratic-like at low fields (near flat-band conditions) and becomes linear at higher fields in agreement with theory.[10] From the electric-field dependence of the lifetime peak 1, we assign this lowest energy peak to an interwell-like transition involving an electron and a heavy hole. This is in good agreement with the CW results obtained previously using the PLE technique.[14]

The energy dependence of transition 2 on the applied external voltage was monitored only for some values of external bias ranging from 0.75 to 1.125 V. This emission, which appeared only weakly in the PL spectra, was observed more clearly using the PLE technique.[14] The spectral position of peak 2 was found to be almost constant from 0.75 to 1.125 V as shown in Fig. 4. Transition 1, however, was found to change by more than 6 meV over the same bias range. We attribute transition 2 to a heavy-hole intrawell-like transition in agreement with the PLE results obtained previously.[14] This transition, which is symmetry forbidden under flat-band conditions, remains spatially direct in an electric field and thus does not lose oscillator strength. The lifetime for such a recombination is expected to be fast because of the spatially direct nature of this transition and thus the strong overlap of the electronic wave functions. The transient PL decays of transition 2 were recorded for the various external bias voltages and are also summarized in Fig. 4. The lifetimes were all found to be fast and constant at ~ 100 ps, independent of the applied field. Transition 2 is faster than transition 1, even under flat-band conditions, since its higher energy permits it one more decay channel than transition 1, i.e., decay to transition 1.

In summary, we have presented a detailed optical study of the influence of external electric fields on the lifetime of exciton transitions in a coupled-double-quantum-well structure. This work is strong evidence that the model we proposed earlier is accurate and details the clear observation of the transformation of a spatially direct exciton transition into a spatially indirect one.

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[1]E. E. Mendez, G. Bastard, L. L. Chang, and L. Esaki, Phys. Rev. B **26**, 7101 (1982).

[2]G. E. Stillman, C. M. Wolfe, C. O. Bozler, and J. A. Rossi, Appl. Phys. Lett. **28**, 544 (1976).

[3]T. H. Wood, Appl. Phys. Lett. **48**, 1413 (1986), and references therein.

[4]T. H. Wood, C. A. Burrus, D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, and W. Wiegmann, Appl. Phys. Lett. **44**, 16 (1984).

[5]D. A. B. Miller, D. S. Chemla, T. C. Damens, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. B **32**, 1043 (1985).

[6]D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. Lett. **53**, 2173 (1984).

[7]J. A. Kash, E. E. Mendez, and H. Morkoç, Appl. Phys. Lett. **46**, 173 (1985).

[8]T. H. Wood, C. A. Burrus, D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, and W. Wiegmann, IEEE J. Quantum Electron. **QE-21**, 117 (1985).

[9]D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Appl. Phys. Lett. **45**, 13 (1984).

[10]E. J. Austin and M. Jaros, J. Appl. C **19**, 533 (1986).

[11]H. Kawai, J. Kaneko, and N. Watanabe, J. Appl. Phys. **58**, 1263 (1985).

[12]A. Yariv, C. Lindsey, and U. Siran, J. Appl. Phys. **58**, 3669 (1985).

[13]H. Q. Le, J. J. Zayhowski, and W. D. Goodhue, Appl. Phys. Lett. **50**, 1518 (1987).

[14]Y. J. Chen, Emil S. Koteles, B. S. Elman, and C. A. Armiento, Phys. Rev. B **36**, 4562 (1987).