Lifetime Enhancement of Two-Dimensional Excitons by the Quantum-Confined Stark Effect

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(Received 9 September 1985)

We report on picosecond luminescence studies of GaAs/AlGaAs quantum wells in the regime of the quantum-confined Stark effect. A drastic increase of the recombination lifetime is accompanied by a Stark shift of the photoluminescence of the lowest free exciton for electric fields perpendicular to the quantum-well layers. A consistent picture of the quantum-confined Stark effect is presented.

PACS numbers: 78.20.Jq, 71.35.+z, 78.55.Ds

Electric fields applied perpendicular to the layers of quantum wells (QW's) significantly change the optical absorption, reflection, and photoluminescence properties.[1-6] A shift of the absorption edge caused by the quantum-confined Stark effect (QCSE) has been reported.[1,2] It can exceed the binding energy of the lowest exciton appreciably. This feature of the QCSE originates in the polarized, yet still confined, electron-hole pairs within the wells. Novel applications of the QCSE have already been presented, e.g., high-speed optical modulators[3] or electro-optical bistable devices.[4] A similar shift toward lower energies in the QCSE regime is expected also for the *photoluminescence* of the two-dimensional excitons in the QW. In addition, an *increase of the recombination lifetime* should be observed as a consequence of the decreased electron-hole overlap. The cw and time-resolved photoluminescence (PL) experiments reported so far, however, are inconsistent with the model of the QCSE,[5,6] because neither the low-energy shift nor the increase in PL lifetime has been observed. Instead, e.g., in a 3-nm QW, strong quenching of the PL[5] and a concomitant decrease of the decay time,[6] attributed to a field-induced dissociation and subsequent tunneling of the carriers, have been found.[6]

This Letter reports on picosecond PL studies of GaAs/AlGaAs QW's in the quantum-confined Stark regime. We find a low-energy shift of the lowest exciton emission line and a corresponding increase of the recombination lifetime, both depending strongly on the quantum-well thickness. The data are explained by the field-induced charge separation and the corresponding decrease in electron-hole wave function overlap. A consistent picture of the QCSE in the luminescence is thus obtained for the first time.

Our measurements are performed with a GaAs/$Al_{0.3}Ga_{0.7}As$ sample grown by molecular-beam epitaxy and containing three decoupled single QW's (20-nm AlGaAs barriers) with nominal thicknesses of about 5, 10, and 20 nm and 100-nm-GaAs/100-nm-AlGaAs buffer layers (see inset in Fig. 1). The electric field is applied via a semitransparent Schottky contact formed by evaporation of a 20-nm-thick Au film onto the top

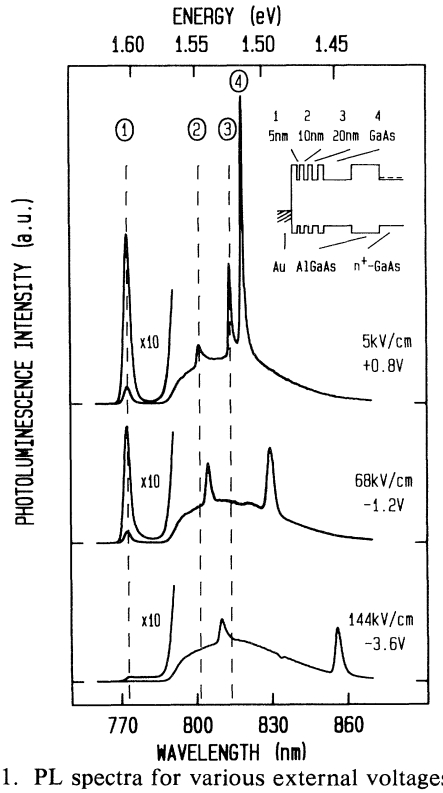

FIG. 1. PL spectra for various external voltages at an excitation energy of 1.675 eV and intensity of 1.5×10^{11} photons/cm$^2\cdot$pulse. The inset shows the band scheme of our sample under the flat-band condition.

AlGaAs cladding layer. All measurements are performed at $T=8$ K. The QW's are directly excited by picosecond pulses of a synchronously mode-locked cw dye laser with a photon energy of 1.675 eV for which the AlGaAs is transparent. The pulse duration and the repetition rate are 3 ps and 80 MHz, respectively. Two excitation intensities of 5×10^{12} and 1.5×10^{11} photons/cm^2·pulse are used which correspond to peak carrier densities of 7.5×10^{16} and 2×10^{15} cm^{-3}. The time-integrated PL spectra are measured with a spectral resolution of 0.5 meV. The time-resolved data are taken with a synchroscan streak camera with spectral and temporal resolutions of 6 meV and 20 ps, respectively.

Figure 1 displays the time-integrated PL spectra for three externally applied voltages V_{ext}. These spectra are corrected for the spectral sensitivity of our system. At $V_{ext}=0.8$ V the built-in voltage[7] is almost compensated, and the PL roughly corresponds to the flat-band condition. The four distinct PL peaks at 1.6031, 1.5459, 1.5225, and 1.515 eV are attributed to the excitonic recombination in the 5-, 10-, and 20-nm QW's and in the 100-nm GaAs buffer layer, respectively. The widths of the two-dimensional exciton luminescence peaks (FWHM) of the QW's amount to 6, 4, and 3 meV, the Stokes shifts with respect to the peaks in the excitation spectra are 5, 3, and 1.7 meV. The broad PL background between 1.44 and 1.57 eV is emitted from the n^+-GaAs substrate. This luminescence feature is independent of the applied voltage as expected for highly doped GaAs layers.

The decrease of V_{ext} significantly changes the PL of the sample as shown by the spectra at -1.2 and -3.6 V in Fig. 1. These voltages correspond to electric fields of 6.8×10^4 and 1.44×10^5 V/cm, respectively. The observed shifts of the luminescence lines of the QW's agree qualitatively with the predictions of variational calculations.[8] Strong shifts are observed for the 10- and 20-nm QW's. In the case of the 20-nm QW the excitonic PL is shifted below the bulk exciton energy for external voltages smaller than -0.4 V. In spite of the drastic line shift, the luminescence intensity does not change appreciably. The luminescence energy of the 5-nm QW remains essentially unaffected by the strong electric field, but the intensity significantly decreases for $V_{ext}<-3$ V. The luminescence from the 100-nm GaAs (bulk) buffer layer does not shift at all, but its intensity decreases rapidly with increasing field.

The experimental results for the shifts of the QW PL are summarized in Fig. 2, where the positions of the PL peaks for the different QW's are plotted as a function of V_{ext}. The field dependence is much stronger for the 20-nm QW than for the thinner wells as expected for the QCSE. Shifts as large as 100 meV are obtained at $V_{ext}=-4.4$ V. No significant changes in the luminescence peak position occur for the two different excitation intensities used in our experiments. Figure 2 also shows calculated curves for the Stark shift, obtained by solution of the Schrödinger equation for electrons and holes in an infinite QW[1] (effective electron and heavy-hole masses are $0.067m_0$ and $0.45m_0$, respectively). The corresponding exciton binding energy is taken from Greene, Bajaj, and Phelps.[9] For the calculation, QW thicknesses of 7.8, 11.6, and 18 nm are used. These values are slightly different from the nominal values and have been determined by a fit to the excitonic transition energy at zero electric field. The data are in excellent agreement with our calculations up to electric fields of 10^5 V/cm ($V_{ext}\geq-2.0$ V). Deviations between the theoretical and experimental Stark shifts occur at very high fields ($>10^5$ V/cm) where the voltage drop across the QW is comparable to the barrier height. Obviously, the model of the infinite QW does not correctly explain the Stark shift at fields $>10^5$ V/cm.

Results of the time-resolved PL studies are depicted in Fig. 3. The inset shows original streak traces for the 20-nm QW as an example. The increase in luminescence decay time with increasing field is obvious. At the highest field ($V_{ext}=-1.6$ V) the streak camera trace shows an offset which is due to the fact that the decay time becomes comparable to the separation of the laser pulses, leading to an accumulation of the signal.[10] The complete data for the 5-, 10-, and 20-nm QW are plotted in the main figure. The zero-field lifetimes (flat-band condition) decrease with thickness as

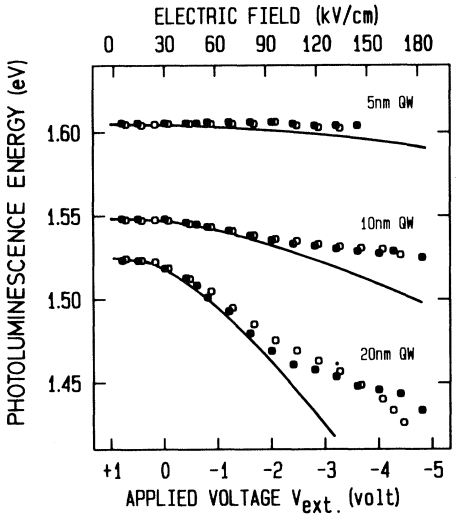

FIG. 2. PL peak energy position of the QW's as a function of the external voltage (electric field) for an excitation intensity of 1.5×10^{11} photons/cm^2·pulse (open circles) and 5×10^{12} photons/cm^2·pulse (closed circles). The excitation energy is 1.675 eV. The solid lines are calculated for an infinite QW exposed to an electric field perpendicular to the layers.

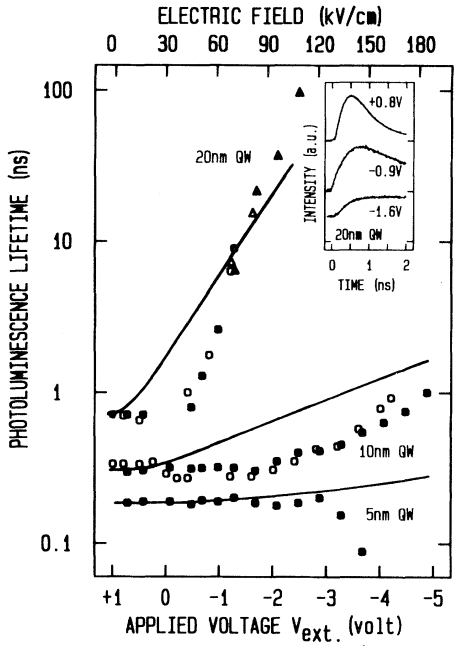

FIG. 3. Time dependence of the PL intensity for various external voltages. The excitation energy is 1.675 eV. Excitation intensity: 1.5×10^{11} photons/cm^2·pulse for the open circles and triangles (Ref. 10), and 5×10^{12} photons/cm^2·pulse for the closed circles and triangles (Ref. 10) and for the streaks in the inset.

a result of the two-dimensional carrier confinement.[11] The carrier lifetime of the 5-nm QW is essentially field independent up to electric fields of 1.25×10^5 V/cm ($V_{ext}=-3$ V). Quite differently, the luminescence decay times of the wider quantum wells are strongly prolonged by an electric field. In particular, the PL lifetime of the 20-nm QW increases from 660 ps under the flat-band condition to about 100-ns at $V_{ext}=-2.4$ V. This lifetime enhancement with the electric field prevents the measurement of luminescence decay times for $V_{ext}<-2.4$ V with the present setup. As the luminescence intensity remains almost unchanged up to $V_{ext}=-4.4$ V, we conclude that the influence of nonradiative processes on the exciton lifetime is small for $V_{ext}>-4.4$ V. A simple extrapolation of the decay times therefore predicts PL lifetimes as large as tens of microseconds for $V_{ext}=-4.4$ V.

The same model of an infinite quantum well which has been applied to calculate the quantum-confined Stark shift is used to evaluate the luminescence decay times with the assumption that radiative recombination dominates. The radiative recombination lifetime τ_r is given by $\tau_r=\tau_0/|M_{cv}|^2$ in the one-particle picture, where M_{cv} is the overlap integral of the electron and hole wave functions,[8] and τ_0 is the lifetime under the flat-band condition. The effective widths of the QW's are already fixed by the calculated excitonic luminescence energy under the flat-band condition. The only additional parameters in the curves are the luminescence lifetimes under the flat-band condition, which are 180, 280, and 660 ps, for the 5-, 10-, and 20-nm QW's, respectively. The theoretical calculations are presented by the solid lines in Fig. 3. The good agreement between theory and experiment demonstrates that the lifetime enhancement in an electric field is mainly caused by charge separation within the QW.[12]

For the 5-nm QW at fields above 130 kV/cm ($V_{ext}<-3.2$ V), we observe a sharp decrease of the PL lifetime. This decrease together with the quenching (Fig. 2) can be attributed to quantum-mechanical tunneling of electrons and holes through the barriers as calculated by Austin and Jaros.[13] The tunneling will be most important for the thinner QW because of the higher subband energy.

Our data clearly show that, besides the shift of the absorption edge, the QCSE manifests itself in the luminescence and, in particular, leads to an appreciable increase of the recombination lifetime. This increase of the lifetime, however, can be observed only in high-quality samples where competing nonradiative processes are of minor importance.

In summary, we have shown for the first time that significant quantum-confined Stark shifts up to 100 meV can be observed in the luminescence of a GaAs/AlGaAs quantum-well system. An enhancement of the luminescence lifetime by more than two orders of magnitude is found and explained by the spatial separation of electrons and holes. Our data are described by a simple theoretical model of an infinite quantum well exposed to an electric field perpendicular to the layers. Together with previous absorption data and theoretical considerations, a complete and consistent understanding of the quantum-confined Stark effect is now obtained.

The authors gratefully acknowledge the expert technical assistance of K. Rother and H. Klann. We are also grateful to R. F. Kopf for assisting in the molecular-beam-epitaxy growth. The Schottky contacts have been evaporated by the group of Dr. H. U. Habermeier. Many thanks are also expressed to Professor H.-J. Queisser and Dr. G. Fasol for the critical reading of the manuscript and stimulating discussions.

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$$\tau_d = -\{\ln[PL_0/(\Delta PL + PL_0)]\}^{-1} t_s.$$

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