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Acceptor-bound excitons in GaAs/$Al_xGa_{1-x}As$ symmetric coupled-double-quantum-well structures

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The effect of an electric-field perturbation on excitons bound to Be acceptors in symmetric coupled-double-quantum-well (CDWQ) structures has been studied with photoluminescence (PL) and photoluminescence excitation spectroscopy. The Be acceptors were positioned in the central region of each well. With an electric field perpendicular to the well layers, interwell and intrawell free excitons are observed in the PL spectra of CDQW's. In weakly coupled DQW's, interwell and intrawell excitons bound to neutral acceptors can be observed simultaneously. Only one (interwell) bound exciton is observed in a strongly coupled system. Interwell and intrawell bound excitons are found to have the same bound-exciton (BE) binding energies. This indicates that it is mainly the hole of the exciton which is attracted by the neutral acceptor impurity. No variations of BE binding energies with the strength of the electric field were observed for the weakly coupled DQW's, while the binding energy increased with the electric field in a strongly coupled DQW. It was found that the BE binding energies at zero electric field in CDQW's are equal to those obtained for center-doped single-quantum-well structures with the same well widths.

Transitions related to excitons bound to neutral acceptor dopants in quantum wells have been reported by Miller *et al.*[1] The transitions are observed when the concentration of dopant atoms is of the order of 10^{16}–10^{17} cm^{-3}. The bound-exciton (BE) binding energies depend on well width and the position of the acceptor in the quantum well. Miller *et al.* found that for acceptors in the central regions of GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells the binding energies vary from 6.5 meV for 46-Å quantum-well widths to 4.0 meV for 155-Å quantum-well widths. Holtz *et al.*[2] have obtained 1*s*-2*s* transition energies for Be acceptors by two-hole transition measurements on multiple quantum wells. Theoretical calculations agree well with these results.[3–5] A system where the consequences of coupling between energy levels in adjacent quantum wells can be studied is the symmetric coupled double quantum well (CDQW) where the single-particle states in the corresponding single quantum well (SQW) are split into symmetric and antisymmetric states. At zero electric field up to four excitonic transitions related to $n=1$ states in the corresponding single quantum well can be observed in photoluminescence excitation (PLE) spectra.[6] It has been shown theoretically and experimentally that electron-hole interaction can be important for such systems,[7,8] and the ratio between the energy level splitting and the exciton binding energy determines if CDQW's are weakly or strongly coupled. No investigation of the influence of coupling effects on acceptor states has been reported, but donor binding energies in CDQW's in a magnetic field have been studied by Raganathan *et al.*[9] It was found that in weakly coupled systems the donor states have properties similar to those found in the corresponding SQW's, while coupling effects are important for donor states in strongly coupled systems.

Here the effects of an electric field on the PL spectra of Be center-doped CDQW's are reported. Under an internal electric field the symmetry of the CDQW is broken, and the coupling between the wells is modified. Up to eight transitions related to the $n=1$ states in the corresponding SQW's can be observed in PLE spectra.[6] The transitions are related to exciton states of mainly interwell or mainly intrawell nature, and the bound excitons will also be influenced in the same way. Figure 1 is a schematic diagram of energy levels and radiative transitions. To find the changes introduced by coupling in CDQW's, the PL spectra of the corresponding center-doped SQW's have been measured as well.

The samples were grown in a Varian Gen II modular molecular-beam epitaxy machine on silicon doped (*n*-type) GaAs(001) substrates. For all the samples the growth was started with a 5000-Å-thick *n*-doped (1×10^{18}

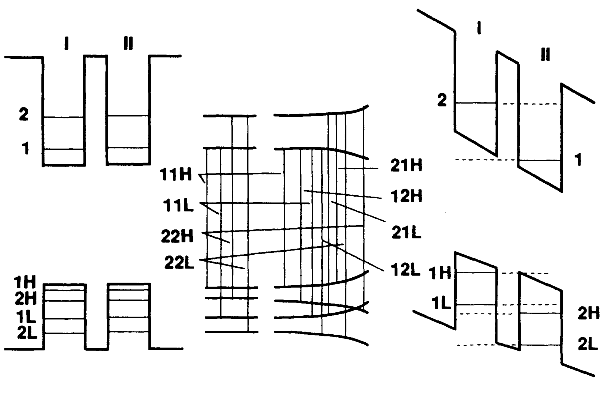

FIG. 1. Schematic diagram of energy levels and radiative transitions in a CDQW under flat-band conditions and with an electric field perpendicular to the well layers. The excitonic transition between the *n*th electron subband *n* and the *m*th heavy-hole (light-hole) subband *mH* (*mL*) is labeled *nmH* (*nmL*).

cm^{-3} Si) GaAs buffer layer and continued with an undoped GaAs buffer layer. The GaAs buffer layer was succeeded by superlattice buffer consisting of 20 periods with 20-Å (7 monolayers) GaAs and 20-Å (7 monolayers) AlAs. On the buffer layers an 800-Å $Al_{0.3}Ga_{0.7}As$ barrier was grown. In the CDQW sample three different center-doped double-quantum-well structures with equal $Al_{0.3}Ga_{0.7}As$ barriers of width 14 Å (5 monolayers) were grown on top of this barrier. The individual well widths in the CDQW's were nominally 150 Å (53 monolayers), 99 Å (35 monolayers), and 59 Å (21 monolayers). The central regions of widths 31, 20, and 14 Å in the wells of the respective structures were doped with Be to a level of 1×10^{17} cm^{-3}. The double wells were separated by 800-Å $Al_{0.3}Ga_{0.7}As$ barrier layers. As a reference, a sample was grown in which the CDQW's were replaced by SQW's of the same widths as the individual quantum wells of the CDQW's and with the same doping profiles. An 800-Å $Al_{0.3}Ga_{0.7}As$ barrier layer and a 200-Å *p*-doped (1×10^{18} cm^{-3} Be) GaAs capping layer terminated the structures. Both samples were grown with As_2 from a cracker source. The GaAs growth rate was fixed at 1 μm/h. The buffer and superlattice layers were grown at a substrate temperature of 585 °C, while the $Al_{0.3}Ga_{0.7}As$ barriers and GaAs quantum wells were grown at 620 °C.

Semitransparent Schottky contacts were formed on the sample surfaces by sequential deposition of a 10-Å-thick Cr film and a 40-Å-thick Au film. With these contacts a uniform electric field perpendicular to the well layers could be applied. The samples were mounted on the cold finger in a closed-cycle cryostat and cooled down to a temperature of 12 K. The excitation source for the luminescence measurements was a tunable Ti:sapphire laser pumped by the all-lines output from an argon-ion laser. The luminescence emitted by the samples was dispersed by a double monochromator with 0.85 m focal length and 1800-grooves/mm gratings and detected by a cooled GaAs photomultiplier in a photon counting mode.

The PL spectra from the samples were recorded with excitation at a photon energy of 1.70 eV, this is under the band gap of the $Al_{0.3}Ga_{0.7}As$ barrier layers. The intensity of the laser beam used for excitation was approximately 0.1 W/cm^{-2}. Since transitions related to free excitons and band-to-band absorption are dominating in PLE spectra, PLE measurements can be used to distinguish between free-exciton- and bound-exciton-related features of the PL spectra. The PLE spectra of CDQW's are sensitive to an electric field, so they were also used to determine the voltage at which flat-band conditions occurred. The PLE spectra were obtained with detection of the luminescence from the Be transitions of lowest energy. The resulting PLE spectra are normalized with respect to laser excitation intensity variations with wavelength tuning.

Due to the *p-i-n* doping profile of the samples, there is an electric field present in the intrinsic layers at zero applied voltage. The applied voltage corresponding to zero internal electric field was determined by comparing PLE spectra for the CDQW's with those obtained for a CDQW sample with a *n-i-n* doping profile, where the built-in electric field is zero. In the PL spectra obtained for the center-doped CDQW sample at this particular voltage one intrinsic excitonic transition, related to the lowest free-exciton resonance, and one extrinsic transition, related to an exciton bound to a neutral Be acceptor, are observed for all CDQW's. There are no significant differences in BE binding energies between the corresponding CDQW and SQW structures at zero internal electric field.

Under an internal electric field strongly and weakly coupled DQW's have quite different properties (Fig. 2). For the CDQW with 150-Å wells a free-exciton (FE) transition 12*H* which was initially forbidden at zero internal electric field occurs at the high-energy side of the FE transition 11*H* with the lowest energy, and two BE

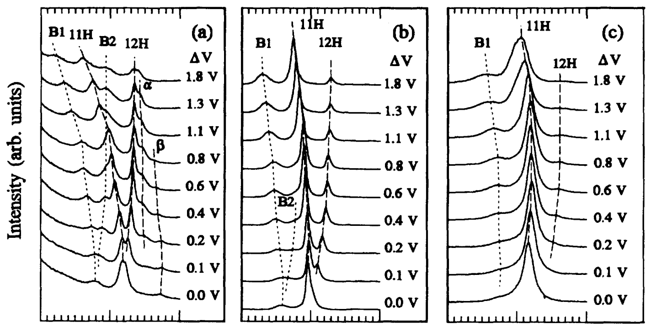

FIG. 2. PL spectra obtained at various applied voltages for center-doped CDQW's for (a) 150-Å, (b) 99-Å, and (c) 59-Å quantum wells. The dashed lines are guidelines used to label the transitions. The features *B*1 are interwell bound excitons, *B*2 are intrawell bound excitons. 11*H* and 12*H* are free-exciton transitions shown in Fig. 1, while the transitions α and β are tentatively identified as 21*H* and 11*L*. The electric field was zero at an applied voltage of $V_0=4.84$ V. The voltage ΔV given at the right side of each curve is the difference V_0-V_{applied}. (Due to resistive heating by a leakage current the sample temperature was increased with approximately 10 K at V_0. For this reason, the emission peaks are slightly redshifted.)

transitions can be observed. With increasing electric field the energy separation between the two FE features increases. The low-energy FE peak shows a distinct shift to lower energies with increasing internal electric field, while the high-energy FE transition initially has a small shift to higher energies before it reaches a near-constant transition energy. The two BE features are obviously each related to one of the FE features. The transition observed at the lowest energy in the PL spectra of the doped CDQW's is the recombination of an interwell acceptor bound exciton. The transition energy varies with electric-field strength as the transition energy of an interwell free exciton. The other extrinsic transition (which is absent in PLE spectra) occurs at an energy that varies with electric-field strength as the energy of the intrawell FE transition. Because of this variation with electric field, the transition can be identified as the recombination of an intrawell acceptor bound exciton. Thus we were able to observe this doublet of bound excitons. The intrawell excitons are probably bound to neutral acceptors in well II (see Fig. 1). At a voltage difference from flat-band conditions of approximately 0.8 V the energy levels of the lowest FE transition and the highest BE transition cross. Within the experimental resolution of 0.1 meV the Be binding energy is 4.0 meV for both interwell and intrawell acceptor bound excitons in this CDQW. The energy difference between a FE transition and its related BE transition is constant under a varying electric field.

A careful inspection of the PL spectra for the CDQW with 99-Å wells reveals features which are similar to those found for the CDQW with 150-Å wells, with a BE binding energy of 4.3 meV for both types of bound excitons. The intensities of the BE transitions are reduced and the linewidths of the peaks are slightly increased. Hence, the separation of the two BE transitions is more difficult to observe. From our experimental data it is not possible to determine if a crossing between the BE transition with the highest energy and the FE transition with the lowest energy occurs. In a strong internal electric field only three peaks are found in the PL spectra.

In contrast to the complex features of the wider structures, the PL spectra for the CDQW with 59-Å wells show only one bound exciton peak at all field strengths. The dominating FE transition shifts to lower energy with increasing internal electric field, and the BE transition follows this variation. A weak feature associated with intrawell free excitons is also observed. The BE binding energy increases with increasing internal electric field. The PL spectrum of Fig. 2(c) shows an increase from 4.6 meV at zero internal electric field to 5.5 meV at the strongest electric field. As all features here are relatively broad and the intrawell transition is weak, it will be difficult to observe an intrawell BE transition here.

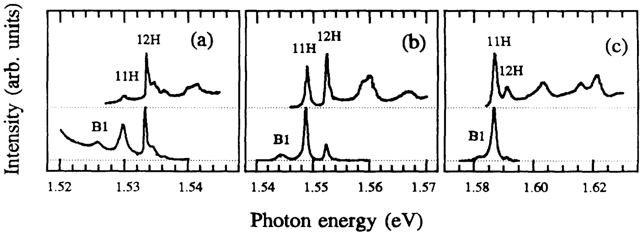

FIG. 3. PL (lower curve) and PLE (upper curve) spectra of CDQW's with an electric field perpendicular to the well layers ($\Delta V = 0.6$ V) for (a) 150-Å, (b) 99-Å, and (c) 59-Å quantum wells.

The PLE spectra for the structures at a moderate electric field (0.6-V bias difference from flat-band conditions) show that the three CDQW's have different intrinsic properties (Fig. 3). The $11H$ transition for the CDQW with the widest wells has an almost negligible oscillator strength compared to the strength of the transition $12H$. For the CDQW with 99-Å wells, the oscillator strength of the transition $11H$ is only slightly lower than for the transition $12H$. For the CDQW with the narrowest wells, the transition $11H$ has high absorption strength at this value of the electric field. The evolution of the ratio of the FE transition intensities with varying electric field can be understood from the nature of the excitons. Under an electric field perturbation, excitons in CDQW's can be roughly classified as intrawell and interwell excitons.[6] At zero electric field only transitions with $n = m$ are allowed by an electric dipole selection rule (see Fig. 1), but at nonzero electric field the transitions $12H$, $21H$, $12L$, and $21L$ are also allowed. At a sufficiently strong electric field the lowest excitonic state $11H$ has an interwell nature where the electrons and holes are mainly localized in opposite wells. These states show a distinct redshift with increasing electric field. The value of the overlap integral between electron and hole wave functions is significantly reduced. The oscillator strength for transitions between this exciton state and the ground state is therefore small, and the lifetime of the exciton with the lowest energy will be relatively long compared with lifetimes in SQW's. The excited state $12H$ has an intrawell nature, i.e., electron and hole wave functions are localized in the same well. The radiative lifetime of these excitons will be short compared to the interwell excitons.[10] For this reason, the intrawell excitons provide an important radiative recombination channel in the CDQW's with wide wells. If the radiative lifetime of these excitons is long compared to the lifetime with respect to scattering to the lowest exciton state, the two exciton levels will be at a thermal quasiequilibrium. The ratio of the intensities of the transitions $12H$ and $11H$ is then given by

$$\frac{I_{12}}{I_{11}} = \frac{O_{12}}{O_{11}} e^{-\Delta E / k_B T} , \tag{1}$$

where I_{ij} are integrated intensities of the transitions ijH, O_{ij} are oscillator strengths of the same transitions, and ΔE is the energy difference between the exciton states $12H$ and $11H$. Our PL and PLE results are consistent with Eq. (1) if the excitons are assigned a temperature T of approximately 30 K.

Due to the complex valence-band structure in GaAs/Al$_x$Ga$_{1-x}$As heterostructures it is difficult to obtain theoretical results for binding energies of excitons to neutral acceptors. Theoretical calculations show that the ground-state binding energy for Be acceptor at the well center vary slowly for SQW's wider than 100 Å and that this variation has its origin from the shift of the valence-band edge due to confinement of hole states in the wells.[3] The spatial extent of the neutral acceptor potential which traps excitons will be comparable to this distance of 100 Å, and this potential will not be significantly influenced by coupling in the CDQW's with wide wells. BE binding energies are in general not particularly sensitive to small changes of the confining potential, so a shift in binding energy due to coupling between the wells will be small.

A probable explanation for the observation of equal BE binding energies of interwell and intrawell excitons in wide CDQW's is that the impurity potential of the neutral acceptor mainly attracts the hole. The hole of the bound exciton is then confined by the impurity potential of a neutral acceptor in well I for interwell excitons (Fig. 1). The electron is mainly in well II and consequently separated from this impurity potential. The hole of an intrawell exciton is confined by neutral acceptors in well II. Here the electron is located in the same well, that is within the range of the impurity potential. If the electron only interacts relatively weakly with the neutral acceptor, the BE binding energy will be the same as for the interwell bound exciton, in accordance with the experimental results. The concentration of acceptor impurities in our samples is so low that it can be justified to use a model where one exciton interacts with only one neutral acceptor. It should be noted that the assumption that it is the hole of the bound exciton which is bound to a neutral acceptor is not an exact model for excitons bound to neutral acceptors in bulk GaAs. In reality, binding of excitons to neutral acceptors is a complex three-particle problem.[11] That no variation of BE binding energies with electric field is observed in weakly coupled DQW's is an indication that the neutral impurity potential and the wave function associated with the hole of the bound exciton is not too strongly perturbed by the electric field. The symmetry splitting of heavy-hole energy levels at zero electric field is smaller than BE binding energies for all CDQW's here, and the hole of the bound exciton will be confined in one quantum well of a CDQW by the neutral acceptor potential. The acceptor states in the CDQW's with 59-Å wells can be influenced by coupling between the wells, but an explanation for the increase in BE binding energy with increasing internal electric field requires a detailed model of a bound exciton in an electric field in a CDQW.

In summary, free and bound excitons in strongly and weakly coupled doped DQW's have been studied by PL spectroscopy under varying electric fields. In weakly coupled DQW's both interwell and intrawell bound excitons can be observed. The BE binding energies for the two types of bound excitons were identical. In these CDQW's the BE binding energies were constant (within the experimental resolution) under variations of the electric field, while the BE binding energy for a strongly coupled DQW increased with increasing electric field. Within experimental uncertainty the BE binding energies at zero electric field in the CDQW's studied here were identical to those found in the corresponding SQW structures.

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