Excitonic photoluminescence in symmetric coupled double quantum wells subject to an external electric field

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The effect of an external electric field \( F \) on the excitonic photoluminescence (PL) spectra of a symmetric coupled double quantum well (DQW) is investigated both theoretically and experimentally. We show that the variational method in a two-particle electron-hole wave function approximation gives a good agreement with measurements of PL on a narrow DQW in a wide interval of \( F \) including flat-band regime. The experimental data are presented for an MBE-grown DQW consisting of two 5 nm wide GaAs wells, separated by a 4 monolayers (MLs) wide pure AlAs central barrier, and sandwiched between \( \text{Ga}_0.7\text{Al}_{0.3}\text{As} \) layers. The bias voltage is applied along the growth direction. Spatially direct and indirect excitonic transitions are identified, and the radius of the exciton and squeezing of the exciton in the growth direction are evaluated variationally. The excitonic binding energies, recombination energies, oscillator strengths, and relative intensities of the transitions as functions of the applied field are calculated. Our analysis demonstrates that this simple model is applicable in case of narrow DQWs not just for a qualitative description of the PL peak positions but also for the estimation of their individual shapes and intensities.

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I. INTRODUCTION

Because of their technical importance and unique physical properties, semiconductor quantum wells have been the subject of intensive research since their first fabrication in 1973. A brief review of the main achievements and a number of representative references can be found, e.g., in books [1] and [2]. The tunnel-coupled quantum states in DQWs are very sensitive to both electric and magnetic fields, and changes induced by these fields have previously reported results [3,4,6,8].

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A symmetric coupled DQW consists of two identical quantum wells separated by only a thin barrier. The energy levels of the coupled QWs split owing to the interwell tunneling. In the flat-band condition \( (F = 0) \), the eigenfunctions of the DQW have defined symmetries. In this situation, only transitions between electron and hole states of the same symmetry are optically allowed. The ground-state wave function is symmetric, while that of the first excited state is antisymmetric. When an electric field is applied to the DQW, the wave functions become predominantly localized in one well only. Transitions allowed in the flat-band case evolve into spatially indirect (interwell) transitions, which shift linearly in energy as a function of the applied field. On the other hand, transitions forbidden in the flat-band case become allowed and represent spatially direct (intrawell) transitions. Two factors determine the optical transition intensities: (a) the overlap integral of the electron and hole single-particle wave functions, and (b) the population of the energy subbands for a given temperature. In order to describe the optical transitions correctly one has to account for the Coulomb interaction between electrons and holes. We use a two-particle wave function composed of the electron and hole single-particle wave functions multiplied by a function of their relative positions to get an exciton binding energy.

The purpose of this report is to demonstrate that our quite simple theory, applied to the PL results obtained from our specifically designed sample, provides excellent quantitative agreement with regard to both the energetic position of the PL peaks and their shapes and intensities as a function of applied electric fields. These results represent clearly a further improvement compared to the previously reported results [1-5].

II. THEORY

The calculation starts from the envelope-function excitonic Hamiltonian

\[
H_{ex} = H_{oe} + H_{oh} + H_{2D} + U + E_g ,
\]

where the respective electron and hole single-particle terms read [1]:

\[
H_{ov} = -\frac{\hbar^2}{2m_{\nu}(z_{\nu})} \frac{1}{\partial z_{\nu}} \frac{\partial}{\partial z_{\nu}} + V_{\nu}(z_{\nu}) - q_{\nu} F z_{\nu},
\]

\( \nu = \{ e, h \} \) denotes an electron or hole, and \( q_{\nu} \) is the respective charge. \( m_{\nu}(z_{\nu}) \) and \( V_{\nu}(z_{\nu}) \) are the respec-
tive electron or hole \( z \) dependent effective masses and confining potentials of the DQW structure. The electric field \( F \) is applied along the \( z \) axis parallel to the growth direction. \( H_{2D} \) represents the kinetic energy in the \( xy \) plane. The electron-hole exciton interaction is included by means of the Coulomb term \( U \), and the GaAs bandgap energy \( E_g \) completes the total energy \( H_{ex} \).

Eigenergies and eigenfunctions of the single-particle 1D Hamiltonian \( H_{sw} \) are found by using linear combinations of analytical functions \( \sin(\xi) \) and \( \cos(\xi) \) (wells, \( F = 0 \)), Airy functions \( A_i(\xi) \) and \( B_i(\xi) \) (wells, \( F > 0 \)), or \( \exp(\pm \xi) \) (barriers), and by matching the wave function amplitudes and their derivatives divided by the effective masses at each interface. Representative results of such a calculation made for a DQW structure with \( L_w = 18 \) ML, \( L_h = 4 \) ML (\( \approx 5.09 \) nm, and \( 1.13 \) nm, respectively), and for \( F = 30 \) kV/cm are schematically plotted in Fig. 1.

The first two single-particle eigenfunctions \( \varphi^e_i(z_e) \), \( \varphi^h_j(z_h) \) of \( H_{sw} \) are used to construct a set of basis functions for the variational calculation of the exciton states as a single product of one electron and one hole wave function multiplied by a function of relative electron hole positions \( \chi \) : \(^{3,4,5}\)

\[
\chi_{ij} = N_{ij} \varphi^e_i(z_e) \varphi^h_j(z_h) \exp \left(-\frac{\sqrt{\rho^2 + \alpha_{ij}(z_e - z_h)^2}}{R_{ij}}\right),
\]

(3)

where the normalization factors \( N_{ij} \) and the variational parameters \( R_{ij}, \alpha_{ij} \) are, in general, different for each exciton. While the \( z \) coordinates of both carriers are specified absolutely \((z_e, z_h)\), only the relative distance between them projected onto the \( x y \) plane \((\rho)\) is relevant. We minimize the total energy [Eq. (3)] for each exciton separately and obtain four wave functions \( \{\chi_{11}, \chi_{12}, \chi_{21}, \chi_{22}\} \).

In our calculation, we neglect the heavy- and light-hole bands mixing and assume strictly parabolic dispersion relations. Also, the mixing of the exciton states by the Coulomb interaction is not taken into account because of its small effect on the narrow DQW system. \(^3\) The GaAs/AlGaAs/AlAs material parameters and their compositional dependence are taken as in \(^{3,4,5}\).

The \( \chi \) is used to evaluate the optical oscillator strength (see, e.g., Ref. \(^3\)) and the electron-hole overlap integral \( F_{ij}(0) \), which is given by

\[
F_{ij}(0) = \int_{-\infty}^{\infty} \varphi^e_i(z) \varphi^h_j(z) \, dz.
\]

The normalized exciton PL intensity spectrum is then calculated as

\[
I(E) = \frac{\sum_{i,j=1}^{2} L_{ij}(E)|F_{ij}(0)|^2 e^{-\beta E_{ij}}}{\beta},
\]

\[
L_{ij}(E) = \frac{\beta \Delta_{ij}}{\pi} \int_{0}^{\infty} \frac{e^{-\beta E'} dE'}{(E - E_{ij} - E')^2 + \Delta_{ij}^2},
\]

\[
\beta = 1/k_B T, \quad k_B \text{ is the Boltzmann constant and } T \text{ the temperature. The convolution form } L_{ij}(E) \text{ expresses the normalized line-shape of the } i, j \text{th transition, which is determined by a thermal distribution of the excitonic kinetic energy and slightly diffused by a Lorenz function. The width } \Delta_{ij} = 0.5 \text{ meV is used in every case.}
\]

III. EXPERIMENTAL

Our PL experiments were performed on a sample grown by MBE at a temperature of 600°C on a semi-insulating GaAs substrate oriented in the [001] direction. The growth started with a 500 nm wide n-doped (Si, \( 2 \times 10^{18} \text{ cm}^{-3} \)) GaAs layer, followed by a 300 nm wide n-doped (Si, \( 1.4 \times 10^{18} \text{ cm}^{-3} \)) GaAlAs layer. The Al content in the GaAlAs layers was always 0.3. After this the following sequence of GaAlAs layers was grown: 500 nm intrinsic, 5 nm p-\( \delta \)-doped (C, \( 3 \times 10^{17} \text{ cm}^{-3} \)), and 100 nm intrinsic. On top of this separating layer, a sequence of three symmetric DQWs with 4 MLs (\( \approx 1.13 \) nm) wide pure AlAs central barriers in between were grown, employing growth interruptions of 20 s at each heterointerface. The well widths are 35 MLs, 26 MLs, and 18 MLs (\( \approx 10 \) nm, 7.5 nm, and 5 nm), respectively. The DQWs are separated by a 100 nm wide GaAlAs layer.
in each case. The growth then continued with another sequence of GaAlAs layers: 100 nm intrinsic, 5 nm n-δ-doped (Si, 4 x 10^{17} cm^{-3}), and 500 nm intrinsic. On the top a 300 nm wide p-doped (C, 1.4 x 10^{18} cm^{-3}) GaAlAs layer and a 20 nm wide p-doped (C, 2 x 10^{18} cm^{-3}) GaAs cap layer were grown.

The p-i-n configuration of the sample allows us to apply a bias voltage $U_{pn}$ by means of selective Ohmic contacts to the p-doped layers on the top and to the n-doped layers at the bottom. Usually, the bands in any p-i-n structure are tilted and application of forward bias is required to flatten them. We, therefore, used a special sample design comprising δ-doped layers inside the intrinsic region of the structure in order to screen the built-in electric field. Thus, the flat-band regime is obtained almost at $U_{pn} = 0$ V, avoiding high dark current present at higher forward bias. The measured devices of the size 250 µm × 250 µm were defined photolithographically and mesa-isolated. Detailed results will only be presented for the narrow DQW with $L_w = 5$ nm and $L_b = 1.13$ nm, for which the application of our simple theoretical model is justified.

Figure 2 shows the PL spectra of the sample for various bias voltages. The sample was cooled in a closed-cycle cryostat and excited by a Ti:sapphire laser pumped by an Ar^+-ion laser. We used an excitation power of ~100 mW/cm² at a photon energy of 1722 meV (below the bandgap energy of Ga_{0.7}Al_{0.3}As). The emitted luminescence was analyzed by a monochromator with 0.6 m focal length and a 1200 grooves/mm grating, and detected by a cooled CCD camera. We applied reverse bias up to $U_{pn} = -8$ V with the maximum current of only ~10 µA. An estimation of a dissipated power and a negligible shift in position of the spectrum measured with $U_{pn} = +2$ V proved that Ohmic heating of the sample for both reverse and forward biased junction is insignificant.

IV. DISCUSSION

The sample was designed as a single (not multiple) DQW structure in order to minimize the well width fluctuations. For the same reason, the growth interruptions were employed, and a pure AlAs barrier instead of an Al$_x$Ga$_{1-x}$As one was chosen to improve its homogeneity and to avoid barrier height fluctuations. The broadening of the direct exciton peak of ~4 meV (see Fig. 2) and even lower at 10 K (~2 meV, not shown) indicates a quite good quality of the sample. Furthermore, the exciton peaks are not split due to large area monolayer fluctuations of the well widths, a phenomenon which is frequently observed on samples grown with interruptions.

The δ-doped layers shielding the DQWs within the p-n junction are effective in case of an excitation below the Ga_{0.7}Al_{0.3}As bandgap energy, when the carriers are excited exclusively in the wells. The flat band condition is found almost at zero bias voltage. On the other hand, when using a He-Ne laser excitation above bandgap energy, the δ-doped layers are neutralized and become inactive. Consequently, a forward bias corresponding to the bandgap energy is needed in order to overcome the built-in field and to establish the flat-band regime.

The experimental spectra plotted in Fig. 2 were analyzed to gain maximum data for the theoretical procedure. In the calculation, we kept the parameters $L_w$ and $L_b$ constant and varied only $T$ and $F$ for optimization. The temperature $T = 45$ K of the sample was obtained by a comparison of the measured and calculated relative peak intensities of the spectra in the linear field range. The field $F$ was deduced by means of tracing the energy distance between the first two exciton transitions, $\chi_{11}$ and $\chi_{12}$. A very good agreement of the calculated and observed transition energies is represented by Fig. 3. Notice that the optimized $T$ provides an excellent correspondence in the peak positions. The absolute positions of all the peaks including the resonant splitting of the symmetric-antisymmetric states were obtained without any free parameter. The small difference in the inten-
sities and positions of the theoretical lines $\sim 1$ meV in comparison with the experiment can be attributed to (i) the approximations used in the theory and (ii) small deviations from the intended structure of the sample.

**FIG. 3.** PL peak positions as a function of electric field $F$. The lines are calculated for the DQW depicted in Fig. 1 at $T = 45$ K. The markers indicate the peak positions of the experimental PL spectra plotted in Fig. 2.

The inset of Fig. 4 shows the deduced field $F$ as a function of the bias voltage applied to the sample. In the case of the spectrum close to flat-band regime ($U_{pn} = 0$ V), the corresponding field ($F = 1.6$ kV/cm) was determined from a linear part of this dependence. As anticipated, the dependence is linear within a limited voltage interval ($-3 \leq U_{pn} \leq 2$ V), and saturates for higher reverse bias. The linear regime of the band tilting is in very good agreement with $F$ calculated directly from the sample design (dotted line in the inset of Fig. 4). A small discrepancy can be explained as an effect of a non-homogeneous distribution of the electric field over the three DQW systems, or as a result of a too simple description of the single wave functions in the applied variational method.

**FIG. 4.** Calculated PL spectra for the DQW depicted in Fig. 1 at a temperature $T = 45$ K. The applied electric field $F$ is given beside each curve. The inset shows the electric field induced in the DQW by the bias voltage $U_{pn}$.

**V. CONCLUSION**

In this paper we have concentrated on the photoluminescence of narrow symmetric coupled double quantum wells, where the energy difference between excitonic states is high enough to allow neglecting of band mixing effects. The inclusion of exciton interaction is unavoidable for a correct description of the electron-hole states and positioning of optical transitions. We have shown that the simple model describes nearly perfectly all dominant features of the optical transitions, namely
relative intensities, absolute positions of the individual transitions, and the resonant splitting of the symmetric-antisymmetric states.

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