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Center-of-mass quantization of excitons in GaAs quantum-well wires

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We have studied wide quantum-well-wire structures by polarization-dependent photoluminescence and photoluminescence excitation spectroscopy. We observe an up-to-threefold splitting of the lowest heavy-hole-exciton resonance, which arises from a quantization of the excitonic center-of-mass motion perpendicular to the direction of the wires. The energy separation between these transitions depends on the active wire width L_x, which is significantly smaller than the geometrical wire width. Identifying the exciton wave vector K_{ex}^n with $n\pi/L_x$, we can reconstruct the exciton dispersion and find excellent agreement with theory if we use the $k=0$ heavy-hole mass.

In recent years the investigation of low-dimensional systems has attracted much attention. Molecular-beam epitaxy (MBE) has enabled almost routine fabrication of two-dimensional (2D) structures with sharp interfaces producing different and, sometimes, unexpected physics. The optical properties of undoped quantum wells have been shown to be strongly influenced by the carrier confinement in the growth direction,[1] leading, in particular, to a significant enhancement of the excitonic binding energy and oscillator strength.[2] Recently, interest has turned to reducing the dimensionality of these artificial structures still further. There have been several reports of optical experiments on laterally patterned structures where in each case the authors have interpreted the results in terms of an additional lateral confinement on top of the quantum-well confinement in the growth direction.[3-8] In these structures the true one-dimensional (1D) quantum limit has been reached, i.e., the valence and conduction subbands have each been quantized by the lateral confinement. However, the effects of the 1D confinement on the optical properties seem to vary and be strongly dependent on the actual fabrication procedure of these 1D structures, e.g., as blueshifts, polarization properties, enhanced luminescence efficiencies, energy splittings, and decreased diamagnetic shifts. To our knowledge, no unified picture of 1D excitons has been presented. We believe that important information should be obtained from a careful study of the transition from the well-understood 2D- to the 1D-exciton regime. For this purpose we have prepared wide quantum-well-wire (QWW) structures with about 60-nm active-wire width, which is significantly larger than the exciton Bohr radius. Here we observe a threefold splitting of the lowest heavy-hole (hh) exciton resonance, which we interpret in terms of a quantization of the center-of-mass motion (CM). For these wide wires we expect no influence of the lateral confinement on the wave function of the correlated relative motion of the electron and hole. For smaller wire widths this quantization of the exciton's motion as a whole should develop into "real" 1D excitonic states, where the lateral confinement determines the excitonic wave function in the confinement direction, while the Coulomb interaction couples the relative motion along the direction of the wires. This should then directly influence the binding energy and, in a magnetic field, the diamagnetic shift of the exciton. We also want to point out that a very similar behavior has recently been observed for the transition from 3D bulklike excitons via CM quantization into 2D quantum-well excitons.[9,10]

Quantum wire structures have been fabricated from a multiple quantum well (MQW), consisting of 25 GaAs quantum wells of 10.6 nm sandwiched between 15.3-nm-wide barriers, which were grown by MBE on top of a 1-μm-thick $Al_{0.36}Ga_{0.64}As$ optical confinement layer. This MQW configuration results in a considerable increase of the spontaneous luminescence efficiency and in a lowering by about 1 order of magnitude of the stimulated emission threshold at room temperature.[11] By holographic lithography we prepared a mask of photoresist stripes oriented along the [110] direction with a 280-nm periodicity. With three reactive ion etching steps of decreasing depth we etched rectangular grooves in a $SiCl_4$ plasma through all the MQW layers (for process parameters see Ref. 12). The overall etching depth was 760 nm, whereby in the second step the topmost quantum wells were removed. The resulting QWW structures contain 7 of the original quantum wells. Scanning electron micrographs show that the geometrical width of the wires amounts to 150 ± 10 nm. On the QWW samples a small unpatterned part was left to take 2D reference spectra.

The photoluminescence (PL) and photoluminescence excitation (PLE) spectra were excited with normally incident light of a titanium-sapphire laser, which was pumped by an Ar^+ laser. The polarization-dependent measurements were performed in a flow cryostat at 6–8 K. For magneto-optic measurements the sample was placed in a superconducting split coil magnet. A Jobin-Yvon monochromator of 1-m focal length and a photon counting system were used to analyze the PL signal. The overall spectral resolution was set to 0.3 meV.

The PL and PLE spectra of both the unstructured and the microstructured part of the sample are displayed in the upper and lower parts of Fig. 1, respectively. The reference part shows the well-known heavy- and light-hole (lh) excitonic transitions at 1.5539 eV and 1.5666 eV, respectively. In the 2D part of the sample the peaks of the hh exciton coincide in PL and PLE, i.e., there is no detectable Stokes shift. The PL spectrum of the microstructured part of the sample is, compared to the reference,

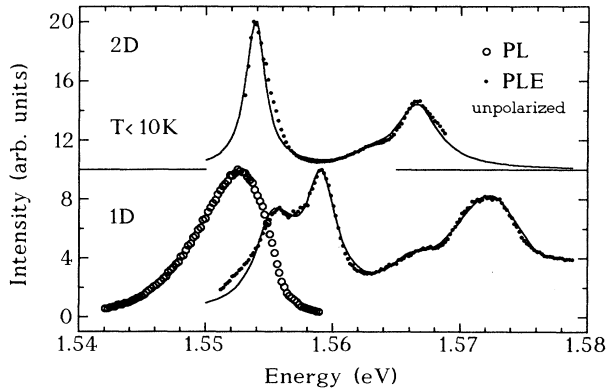

FIG. 1. PL and PLE spectra of the MQW reference (upper part) and the microstructured part of the samples (lower part). The experimental data points are fitted with Lorentzian curves.

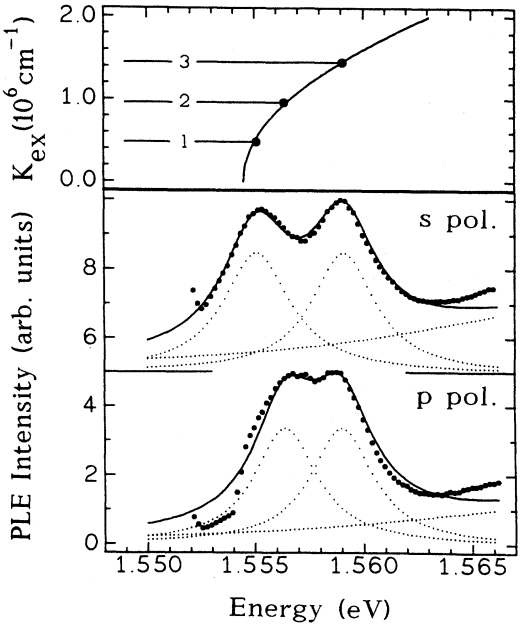

FIG. 2. Polarization-dependent PLE spectra of the CM-quantized hh exciton. The dotted curves display the contribution of the different Lorentzian broadened transitions. The top part shows the reconstruction of the CM dispersion ($M^*_{ex}=0.178m_0$) for $L_x=65$ nm.

redshifted and shows a broad low-energy tail. Therefore we conclude that mainly localized states contribute to the PL signal. The absolute intensity has dropped by a factor of 50. This means, taking into account the reduced number quantum wells and their reduced area, that the emission efficiency per quantum well has only dropped by a factor of 4. Turning to the PLE spectrum of the QWW, however, we see a clearly resolved splitting of the lowest hh excitonic transition, which is also slightly blueshifted. This paper is concerned with a careful analysis of this effect, but before we turn to a detailed discussion we want to state two other observations: The first is that the Stokes shift between the PL maximum and the lowest transition in the PLE spectrum of the QWW vanishes at higher temperatures, while the splitting remains the same. This confirms that the transitions found in PLE spectroscopy are of intrinsic nature. The second is the observation of a blueshift of the lh exciton of 4.5 meV with respect to the quantum-well reference. This blueshift is probably due to a relaxation of the $Al_{0.36}Ga_{0.64}As$ layers perpendicular to the wire direction, which strains the whole structure. X-ray diffractometry shows this clearly and will be discussed elsewhere.[13]

Polarization-dependent PLE spectroscopy (Fig. 2) further reveals that the splitting of the hh exciton is threefold. The ground state (1) can only be observed in *s*-polarized PLE, i.e., the electric-field vector of the exciting and detected light is perpendicular to the plane of incidence and parallel to the wires. The highest-energy state (3) is observed in both polarizations, while the intermediate transition (2) is only observable in *p* polarization. We ascribe these energy splittings to a CM quantization of the excitons in the wide quantum wires. This behavior is expected for $L_x \gg a_B$, where the lateral wire confinement does not quantize the electronic wave functions directly. However, due to the wire structure the motion of the exciton as a whole is not free in the plane of the original well, since it is reflected by the lateral boundaries. In terms of quantum mechanics, the reflection leads to the formation of quantized standing waves perpendicular to the direction of the wires. Consequently, the original continuous 2D energy dispersion of the exciton's motion now becomes a set of discrete points, at least in the direction of the lateral confinement.

The kinetic energy for the center-of-mass motion is given by $E^{CM}_{kin}=\hbar^2 K^2_{ex}/2M^*_{ex}$, where $M^*_{ex}=m_e^*+m_{hh}^*$ is the the total mass of the exciton. In the direction perpendicular to the wires K_{ex} can have only quantized values:

$$K^n_{ex}=\frac{\pi}{L_x}n,\quad n=1,2,3,\ldots. \tag{1}$$

We determine the exact positions of the transitions by fitting the PLE spectra with Lorentzian curves, as is shown by the dotted curves in Fig. 2. Within 5% accuracy the spectral positions E_n obey the following empirical relation:

$$E_n=1554.5+C_1 n^2\ (\text{meV}),\quad C_1=0.5\pm 0.02\,, \tag{2}$$

and show a characteristic quadratic n dependence of an "infinite" square-well confinement. The second term therefore yields the experimental quantization energies of the confined center-of-mass motion. The small shift of the excitonic ground-state energy of 0.6 meV compared to the unpatterned part of the sample may be explained by inhomogeneities of the original MQW or possibly could be due to the relaxation of the $Al_{0.36}Ga_{0.64}As$ layers. To evaluate the quantization length L_x we use $M^*_{ex}=m_e^*+(\gamma_1+\gamma_2)^{-1}=0.178m_0$, where the in-plane mass of the heavy hole is taken from the diagonal elements of the Luttinger-Kohn matrix. Under this assumption L_x is evaluated to be 65 nm, and, as can be seen in the upper part of Fig. 2, the fit of the experimental data to the dispersion of the exciton's center-of-mass motion is

reasonable.

The confining width is significantly smaller than the geometrical wire width. Hence, we can deduce that, on both sidewalls of the wires, there is a relatively thick area over which the properties are significantly altered during the etching process probably due to the penetration and channeling of ions into the semiconductor. But the above value for L_x does not necessarily need to display exactly the width of the active central region. This parameter depends on the penetration depth of the exciton's wave function into the damaged regions. If we assume that the exciton occupies a rigid volume, we have to add roughly $2a_B$ to the value obtained above. On the other hand, we want to note here that x-ray-diffraction measurements, which directly probe the crystalline part of the etched wires, also led to 60 nm width.[13] This is in excellent agreement with the value of L_x which we determine from the splitting of the hh exciton. However, it should be noted that the diagonal approximation underestimates the heavy-hole in-plane mass, since the coupling due to the off-diagonal elements in the Luttinger-Kohn matrix at $k \neq 0$ leads to a flattening of the dispersion.[14] If we take this effect into account, it increases the hh mass and therefore decreases the value derived for L_x.

An interesting feature is the strong polarization dependence of the spectra, in particular the appearance of the peak $n=2$ only in p-polarized PLE. At present we are not sure whether this polarization dependence is an intrinsic property or whether grating coupler effects induce the emission of X-mode quantum-well excitonic polaritons.[15] However, since the energy separation between the exciton and the X-mode exciton-polariton is small[16–18] and is exceeded by the linewidth by about 1 order of magnitude, we are not sensitive to it concerning the small difference in transition energy. D'Andrea and Del Sole[19] evaluated reflectance spectra for wide QWW's, which structural properties are not too different from ours. They found that in s-polarized reflectance spectra only CM-quantized states with *odd n* are observable. This agrees well with our observation. Unfortunately, it is not possible to analyze their p-polarized spectrum in such a straightforward manner, especially for small n.

We also have performed magnetic-field-dependent measurements up to 7 T with the field oriented along the growth direction. We find that the lowest CM-quantized hh-exciton state in the QWW shows essentially the same diamagnetic shift as the corresponding hh transition in the 2D reference, i.e., $\Delta E^{\mathrm{dia}}=0.039(2)B^2$ (meV T^{-2}). Since the diamagnetic shift is a measure for the extent of the excitonic wave function in the plane perpendicular to the magnetic field,[20] this observation ensures that the relative motion of the electron and hole is not affected by the lateral confinement. Conversely, for the dependence of the highest observable CM-quantized transition we find a stronger increase with magnetic field [$\Delta E^{\mathrm{dia}}=0.050(2)B^2$ (meV T^{-2})]. This is shown in Fig. 3 where we have plotted the energy difference E_3-E_1 for these two transitions. It can be seen clearly that the separation increases with magnetic field, and we interpret this observation in terms of an enlarged Bohr radius for the higher transition. This may be due to the fact that already at $B=0$ the $n=3$ CM-quantized state is mixed up with higher excitonic states (e.g., $2S$ state), since their energy separation is small. The quasidegeneracy may then cause the formation of eigenstates consisting of the intermixed wave functions of the different involved states. We also want to mention briefly that the degenerate valence-band structure does not allow, in principle, a complete decoupling of the relative and center-of-mass motion.[21,22] This may also influence the magnetic-field behavior of the higher CM-quantized excitonic states.

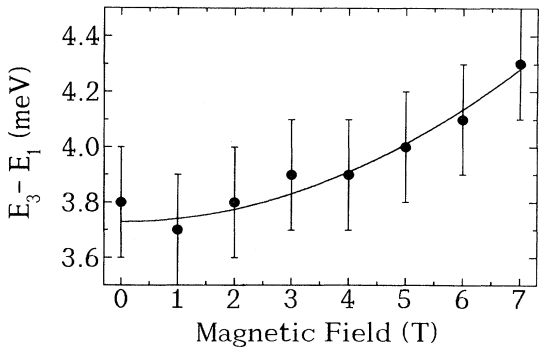

FIG. 3. Energy difference between the $n=3$ and $n=1$ CM-quantized transitions in a magnetic field. The stronger diamagnetic shift of the $n=3$ state is reflected by an increasing separation of the transitions.

We have shown that the interpretation of our experimental data in the framework of the simple picture of the CM quantization leads to a quantitative understanding. However, the above discussion already shows that a more sophisticated picture is probably involved. Here we additionally want to point out that, what has not been considered in theory so far,[19] in QWW the CM-quantized states can be degenerate with transitions between 1D subbands labeled e_1h_j ($j=1,2,3$), where h denotes the 1D hole subbands in the valence band. In QWW's no subbands exist (even at $k=0$) with a pure heavy- or light-hole character.[23] If we assume a wire width of 75 nm, the separation of the hole subbands calculated in Ref. 23 well reproduces our splitting. However, we stress that the two transitions with $j \neq 1$ are forbidden and, since the exciton binding energy is much larger than the 1D hole subband separation, the states are mixed by the Coulomb potential. On the other hand, this observation may account for the fact that we are near the region where the continuous transition between the different types of confinement should occur. Recently, the same kind of observation was also reported for wide CdTe quantum wells, where the transition from 3D bulklike behavior towards the 2D quantum-well limit was investigated.[9]

In conclusion, we have prepared wide QWW, which show the quantization of the center-of-mass motion of the hh quantum-well exciton. The energetic positions of the threefold splitted hh excitonic state well reconstruct its original continuous CM dispersion, when a quantization length of 65 nm is used. This is in close agreement with the crystalline central part of the wires determined to be 60 nm by x-ray diffraction.

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