Uniaxial stress dependence of spatially confined excitons

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We report the first observation of the effect of two-dimensional (2D) confinement on the uniaxial stress dependence of excitons in $GaAs/Al_{0.3}Ga_{0.7}As$ quantum wells. For uniaxial stress $\mathbf{X}||[100]$, perpendicular to the quantization axis ($\hat{\mathbf{z}}||[001]$) of the exciton Hamiltonian without external stress, the exciton energy consists of two components. The first, containing hydrostatic strain terms, is the same for bulk and 2D excitons, in agreement, to within experimental error, with hydrostatic photoluminescence data. The second component contains shear strain terms and is observed to be dependent on quantum-well width. This dependence is interpreted in terms of a model based on the overlap of the light-hole (LH) and heavy-hole (HH) wave functions and on the energy differences between the LH and HH excitons.

INTRODUCTION

It is well known that as a result of two-dimensional confinement in GaAs quantum wells, the degeneracy of the light- and heavy-hole valence-band energies is lifted at $k=0$. Excitons, when produced in such an environment, exhibit two series of discrete energy levels corresponding to electrons bound to either the light hole (LH) or heavy hole (HH).[1,2] Lately there has been considerable interest[3-5] in the hydrostatic stress dependence of excitons in $GaAs/Al_xGa_{1-x}As$ quantum wells. The hydrostatic pressure coefficients α of the light-hole and heavy-hole exciton transitions, measured in photoluminescence (PL), are observed to be nearly the same (within 10%) as that of excitons in GaAs.[3,4] However, recently a systematic small decrease of α with increasing well width L_z has been reported[5] (α decreases by $\approx 5\%$ between $L_z=150$ and 50 Å). In this communication we present experimental results of the large effect of spatial confinement on the uniaxial stress dependence of exciton energies when the stress, $\mathbf{X}||[100]$, is perpendicular to the growth direction [001] of the quantum wells. Experiments were performed at low temperatures using photoluminescence excitation (PLE) spectroscopy on GaAs quantum wells so that both light- and heavy-hole excitons could be easily measured simultaneously. By monitoring the stress dependence of features from a bulk GaAs buffer layer, an accurate measure of the uniformity and magnitude of the applied stress was possible in situ. The change in the HH and LH exciton energies with stress is a sensitive function of L_z. For example, the rate of change of energy of the HH exciton confined in a 220-Å well is 1.55 meV/kbar, for low stress ($X \leq 1$ kbar), compared to 1.1 meV/kbar for bulk GaAs. This slope increases to 1.75 meV/kbar for $L_z \leq 75$ Å. We interpret this change as resulting from the sensitivity of the uniaxial stress dependence of the LH and HH exciton energies to the overlap integral of the LH and HH wave functions which in turn depends on L_z.

THEORY

The Hamiltonian of an exciton in a quantum well in the presence of uniaxial stress can be written as[6,7]

$$H=H_e+H_h+V_{ex}+H_\epsilon \ , \tag{1}$$

where H_e and H_h represent the electron effective mass Hamiltonian and the Luttinger-Kohn Hamiltonian for the hole states, respectively, V_{ex} is the electron-hole potential energy, and H_ϵ is the term introduced by the stress:

$$H_e=-\frac{\hbar^2}{2m_e}\nabla^2+V_e(z)+\Delta C \ , \tag{2}$$

$$H_h=-\frac{\hbar^2}{m_0}\left(\tfrac{1}{2}\gamma_1 k^2-\gamma_2[(J_x^2-\tfrac{1}{3}J^2]k_x^2+\text{c.p.}]\right.$$
$$\left.-2\gamma_3(\{J_x,J_y\}k_xk_y+\text{c.p.})\right)+V_h(z) \ , \tag{3}$$

$$H_\epsilon=D_d(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})+\tfrac{2}{3}D_u[(J_x^2-\tfrac{1}{3}J^2)\epsilon_{xx}+\text{c.p.}]$$
$$+\tfrac{4}{3}D_u'(\{J_x,J_y\}\epsilon_{xy}+\text{c.p.}) \ , \tag{4}$$

$$V_{ex}=-\frac{e^2}{\kappa r} \ . \tag{5}$$

Here, m_e (m_0) is the effective (bare) electron mass, γ_1, γ_2, and γ_3 are the Luttinger parameters,[7] D_d, D_u, and D_u' are deformation potentials for the valence bands, ϵ_{ij} are components of the strain tensor, J_i are the angular-momentum matrices corresponding to a spin-$\frac{3}{2}$ state, $V_e(z)$ [$V_h(z)$] is the electron (hole) well potential barrier height, $\Delta C=C_1(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz})$ is a constant shift due to stress for the conduction band, and κ is the dielectric constant. In Eq. (3) the axis of quantization for H_h is along $\mathbf{z}||[001]$. When the stress X is also along [001] H_ϵ [Eq. (4)] contains only diagonal terms. Assuming that the valence bands are uncoupled at $X=0$, H_h contains diagonal terms only and the solution to the Schrödinger equation $H\psi=E\psi$ predicts linear stress behavior for the LH and HH 2D excitons. The change in energy of the HH and LH excitons as a function of stress, ΔE_{1H} and ΔE_{1L} respectively, are given by $\Delta E_{1H}=a(s_{11}+2s_{12})X+b(s_{11}-s_{12})X$ and $\Delta E_{1L}=a(s_{11}+2s_{12})X-b(s_{11}-s_{12})X$. As demonstrated later, the elastic compliance coefficients s_{11} and s_{12} for GaAs wells are taken to be identical to those of $Al_{0.3}Ga_{0.7}As$ barriers; $a=(D_d+C_1)$ and $b=\frac{2}{3}D_u$ are the hydrostatic and shear deformation potential constants,[8] respectively, which are taken to be identical for excitons in GaAs quantum wells and in bulk GaAs.

In our experiment $X||[100]$, so that the quantization axes of H_h and H_ϵ [Eqs. (3) and (4), respectively] are perpendicular. This leads to off-diagonal terms in the strain Hamiltonian H_ϵ, written in the basis functions which are quantized along $z||[001]$. As a consequence, the eigenvalues of the Hamiltonian H in Eq. (1) contain nonlinear stress dependent terms which are a function of the overlap integral Q of the light-hole and heavy-hole wave functions ψ_1 and ψ_2, respectively. In this case, the energies of the LH and HH excitons as a function of stress are given by

$$E_{1L\,(H)}(X)=E_g+\tfrac{1}{2}(E_2(X)+E_1(X) \pm\{[E_2(X)-E_1(X)]^2+Q^2X^2\}^{1/2}) , \quad (6)$$

where the "+" sign in front of the terms in the curly brackets corresponds to the light-hole exciton energy E_{1L}, while the "−" sign is used to obtain the heavy-hole exciton energy E_{1H}. Here,

$$E_1(X)=E_{b_1}+\Delta E_c+\Delta E_{v_1} +a(s_{11}+2s_{12})X-b'(s_{11}-s_{12})X , \quad (7a)$$

$$E_2(X)=E_{b_2}+\Delta E_c+\Delta E_{v_2} +a(s_{11}+2s_{12})X+b'(s_{11}-s_{12})X , \quad (7b)$$

and

$$Q=2\sqrt{3}\,|\langle\psi_1|\psi_2\rangle|\,b'(s_{12}-s_{11}) . \quad (7c)$$

E_g is the energy bandgap of GaAs, E_{b_1} (E_{b_2}) is the binding energy of the heavy- (light-) hole exciton in the absence of stress, and ΔE_c, ΔE_{v_1}, and ΔE_{v_2} are the lowest subband energies for the electron, heavy hole and light hole, respectively, in the quantum well. Here $b'=b/2=D_u/3$. In the absence of stress, the coupling of valence bands[9] can be incorporated in our model by introducing off-diagonal terms in H_h [Eq. (3)]. In the present experimental configuration $X||[100]$ the contribution of zero-stress valence-band mixing cannot be separated from that resulting from the stress induced off-diagonal terms in the strain Hamiltonian H_ϵ. However, for $X||[001]$, H_ϵ is diagonal and, hence, zero-stress valence-band mixing would lead to nonlinear dependence of ΔE_{1H} with X. ΔE_{1L} can exhibit nonlinear behavior with X as a result of the interaction of the LH valence band with the spin-orbit-split valence band.[8]

When $[E_2(X)-E_1(X)]\gg Q^2X^2$, higher-order terms in X can be neglected and the change in the energy of the light- (heavy-) hole exciton as a function of stress can be simplified to

$$\Delta E_{1L\,(H)}[X]=E_{1L\,(H)}(X)-E_{1L\,(H)}(0) =a(s_{11}+2s_{12})X\pm\left[b'(s_{11}-s_{12})X+\frac{1}{2}\frac{Q^2X^2}{\{[E_l(0)-E_h(0)]+2b'(s_{11}-s_{12})X\}}\right] . \quad (8)$$

$E_l(0)-E_h(0)$ is the energy separation of the light- and heavy-hole excitons at $X=0$. The error introduced by such a simplification is less than 2% for the stress range covered in this experiment. In Eq. (8) the only terms sensitive to spatial confinement of the excitons (i.e., L_z) are $E_l(0)-E_h(0)$ and Q^2 [which is also related to $E_l(0)-E_h(0)$]. All other terms depend on the bulk material properties of the constituents of the structure and, thus, are independent of L_z; they will be taken as constants. $E_l(0)-E_h(0)$ is directly measured in our experiment and, hence, we are left with one fitting parameter, Q^2, which varies with L_z. In the limit of small L_z for low stress ($X\le 1$ kbar), Eq. (8) reduces to the form

$$\Delta E_{1L\,(H)}=a(s_{11}+2s_{12})X\pm b'(s_{11}-s_{12})X$$

and is similar to the case of bulk GaAs except that the shear deformation potential $b'=b/2$. Recent experiments have shown that the hydrostatic coefficient a, for 2D excitons in narrow wells, may be slightly different (≈5%) from that of bulk GaAs (Ref. 5) but, in any case, the difference is within our experimental error.

EXPERIMENTS AND DISCUSSION

The samples used in this study were grown by molecular beam epitaxy along the [001] axis of a semi-insulating GaAs substrate. A typical sample consisted of a 0.75-μm, undoped GaAs buffer layer on which were grown a multiple-quantum-well (MQW) layer of 20 periods of 110-Å GaAs quantum wells and 100-Å $Al_{0.3}Ga_{0.7}As$ barriers and a 220-Å GaAs single quantum well (SQW). Figure 1 shows the 4.5 K photoluminescence spectra of this sample. Peaks due to HH excitons in the two different wells are clearly seen, as is structure due to the GaAs buffer layer.[10] At 4.5 K only states associated with heavy-hole excitons are populated and, therefore, observable by PL. In contrast, photoluminescence excitation spectroscopy reveals information about both HH and LH excitons. This is because PLE spectroscopy (where the wavelength of the spectrometer is fixed in order to monitor the intensity dependence of some relevant energy level while the exciting energy is scanned) is analogous to absorption spectroscopy and thus is sensitive to oscillator

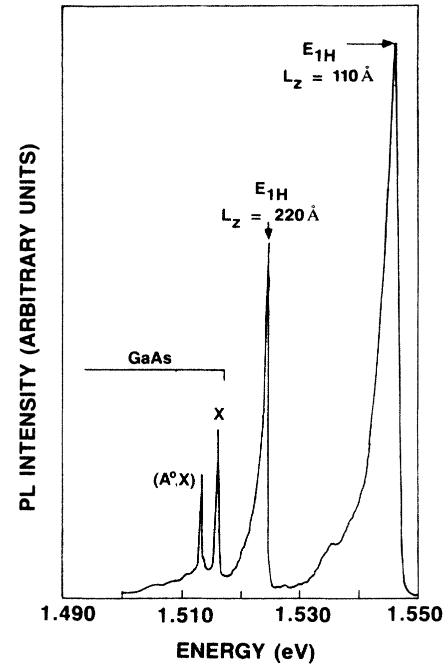

FIG. 1. 4.5 K photoluminescence spectrum of a GaAs/$Al_{0.3}Ga_{0.7}As$ quantum well sample with well widths, $L_z=220$ and 110 Å.

strengths of transitions independent of thermal population effects. This is illustrated in Fig. 2(a), which shows the PLE spectrum of the light hole (LH) and heavy hole (HH) excitons in the 220-Å SQW (E_s identifies the fixed energy of the spectrometer). As uniaxial stress X is applied along

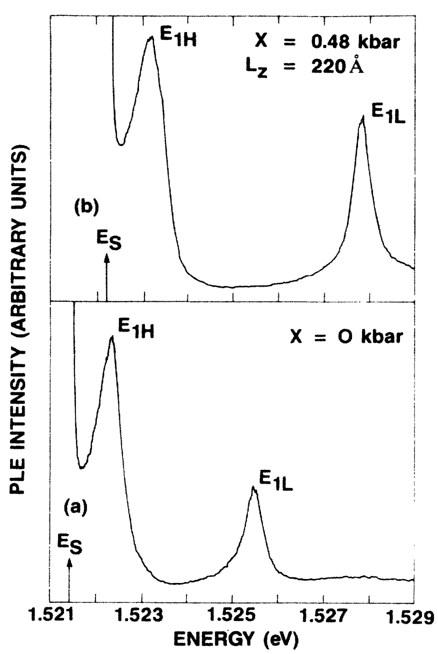

FIG. 2. Photoluminescence excitation spectra of excitons in 220-Å-wide quantum wells. E_s labels the energy of the fixed spectrometer setting and E_{1H} and E_{1L} label peaks due to heavy- and light-hole excitons with uniaxial stress of (a) 0 kbar and (b) 0.48 kbar.

the [100] direction, both ground-state excitons move to higher energies [Fig. 2(b)]. The absence of broadening in E_{1L}, the fastest moving peak, indicates that the applied stress is uniform. In order to achieve uniform stress the sample was held between two polished discs at the bottom of a stress cell. Double-sided tape was used to cushion both ends of the sample. The stress cell is similar in principle to the one described in Ref. 11. Modifications were made in the design so as to incorporate it in an exchange gas cryostat. Typical sample dimensions were $0.5\times2\times8$ mm^3 with the long axis parallel to [100]. The 0.5 mm dimension, imposed by substrate thickness, does lead to slight bending of the sample, which results in some uncertainty in the actual stress applied.[12] The absolute value of stress was determined by observing the well-understood stress dependence of E_{1L} of the GaAs buffer layer.[12] Since this GaAs buffer layer is a fraction of a micron away from the quantum wells being examined, the uniaxial stress experienced by both the buffer and the quantum wells is essentially the same and accurate determination of small differences in the uniaxial stress behavior of bulk and 2D excitons is possible. Furthermore, by observing the stress dependence of the first excited levels of the ground-state excitons[13] we were able to determine that over our experimental range, exciton binding energies were independent of stress.

Figure 3 shows the shift in energy of the HH and LH excitons as a function of stress, ΔE_{1H} and ΔE_{1L}, respectively. The open circles, crosses, and solid triangles represent experimental data for quantum wells of thicknesses $L_Z=220$, 110, and 40 Å, respectively. The dashed lines represent the behavior of HH and LH excitons in bulk GaAs. A systematic increase in the slope of $\Delta E_{1H}(X)$ is observed with decreasing L_Z. In contrast, the slope of $\Delta E_{1L}(X)$ for $L_Z=220$ and 110 Å appears to be the same and decreases for $L_Z=40$ Å.

Since GaAs and $Al_xGa_{1-x}As$ are closely lattice matched, their elastic coefficients are expected to be very similar.[14] Both the hydrostatic, a, and shear deformation potential, b, constants of semiconductors are related to the Phillip's ionicity.[15] Since the magnitude of this parameter is nearly the same for GaAs and AlAs, a and b should be similar for these two materials. From all the experimental data on deformation potential constants a and b can be estimated[15] to be the same for GaAs and $Al_{0.3}Ga_{0.7}As$ to within 10%. Thus, the large observed differences in the slopes of ΔE_{1L} and ΔE_{1H}, as a function of X for the quantum wells of different L_z must be due to the effects of confinement on exciton energies and are not related to the presence of $Al_{0.3}Ga_{0.7}As$ layers in the sample.

The theoretical fits to the ΔE_{1H} data in Fig. 3(a), solid lines labeled A, B, and C for the 220, 110, and 40 Å wells, respectively, were calculated using Eq. (8) and taking Q^2 as an adjustable parameter. $a=7.93$ eV and $b'=0.98$ eV for all the three curves. $E_{1L}(0)-E_{1H}(0)$ was experimentally determined to be 3.2, 9.3, and 26.08 meV for $L_z=220$, 110, and 40 Å, respectively. Best fits were obtained when $Q^2=7.0\pm1.0$, 7.5 ± 1.0, and 4.0 ± 2.0 $(\text{meV/kbar})^2$ for $L_z=220$, 110, and 40 Å, respectively. For other narrow wells, not shown here, Q^2 was found to

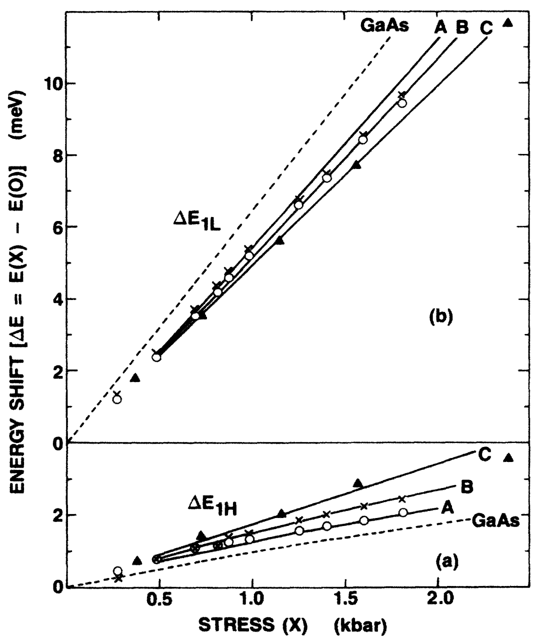

FIG. 3. Experimental [circles (220 Å), crosses (110 Å), and solid triangles (40 Å)] and calculated (solid lines A, B, and C corresponding to $L_z=220$, 110, and 40 Å, respectively) energy shifts of (a) heavy-hole excitons (ΔE_{1H}) and (b) light-hole excitons (ΔE_{1L}) as a function of stress X. The calculated lines were fit to the data of (a) using experimentally determined values in Eq. (8) with Q^2 as an adjustable parameter. The dashed lines represent the behavior of HH and LH excitons in undoped GaAs.

be 4.0±2.0 (meV/kbar)^2 for $L_z=55$ and 73 Å. These values of a and b' are in good agreement with the corresponding values for bulk GaAs.

Using the values obtained from the ΔE_{1H} fitting, Eq. (8) was employed to calculate the curves A, B, and C of ΔE_{1L} versus X in Fig. 3(b). The agreement with experiment is good with the exception of data from the 220-Å well (curve A). Preliminary experiments on the stress dependence of higher-level transitions indicate an anticrossing between E_{13H} (the exciton transition between the $n=1$ electron and the $n=3$ heavy-hole levels) and E_{1L}. For $L_z=220$ Å ($E_{13H}-E_{1L}\approx 10$ meV), this anticrossing occurs at about 2 kbar resulting in sublinear behavior of the E_{1L} feature. However for $L_z \leq 100$ Å, $E_{13H}-E_{1L}\geq 30$ meV, the level repulsion occurs at a stress much higher than that used in our present experiment. This would qualitatively explain the observed deviation from theory of the experimental data for the 220-Å well, while a good fit between experiment and theory is found for $L_z=110$ Å and 40 Å. A detailed account of the stress dependence of exciton transitions between higher subbands will be published later.

In conclusion, we have demonstrated the effect of quantum confinement on the uniaxial stress X dependence of 2D exciton energies when X||[100] is perpendicular to the growth axis [001]. Using the square of the overlap integral of the LH and HH wave functions Q^2 as a fitting parameter in our model, we obtain good agreement between the experimental data and theory. Q^2 is observed to be a constant ≈ 7 (meV/kbar)^2 for $L_z \geq 100$ Å and is seen to decrease by about 40% for $L_z \leq 75$ Å. This behavior is similar to other properties of 2D excitons (e.g., binding energy) which are sensitive functions of L_z, especially for $L_z \leq 100$ Å.

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