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Binding energy and dephasing of biexcitons in $In_{0.18}Ga_{0.82}As/GaAs$ single quantum wells

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Biexciton binding energies and biexciton dephasing in $In_{0.18}Ga_{0.82}As/GaAs$ single quantum wells have been measured by time-integrated and spectrally resolved four-wave mixing. The biexciton binding energy increases from 1.5 to 2.6 meV for well widths increasing from 1 to 4 nm. The ratio between exciton and biexciton binding energy changes from 0.23 to 0.3 with increasing inhomogeneous broadening, corresponding to increasing well width. From the temperature dependence of the exciton and biexciton four-wave mixing signal decay, we have deduced the acoustic-phonon scattering of the exciton-biexciton transition. It is found to be comparable to that of the exciton transition, indicating that the deformation potential interactions for the exciton and the exciton-biexciton transitions are comparable. [S0163-1829(99)07931-X]

Biexcitonic properties of semiconductor nanostructures have attracted a lot of interest in recent years.[1] In particular, the influence of exciton confinement on the biexciton binding energy and the coherent response of biexcitonic resonances have been investigated in numerous systems. Recently, the effect of exciton localization on the binding energy[2–6] and dephasing of biexcitons[7,8] has also been studied, both in III-V and II-VI material systems. It has been observed that exciton localization leads to an enhancement of the biexciton binding for localization energies comparable to or larger than the binding energy of the unlocalized biexciton.[6] In quasi-three-dimensional (3D) and quasi-2D $GaAs/Al_xGa_{1-x}As$ structures with large localization energies, the biexciton binding energy reaches about forty percent of the exciton binding energy.[6] The effect of the exciton localization on the biexciton continuum has also been discussed recently on GaAs single and multiple quantum wells.[9] Additionally, exciton localization leads to an inhomogeneous broadening of the biexciton binding energy, resulting in a fast nonexponential decay of the biexcitonic photon echo in four-wave mixing, as reported in Ref. 8 on ZnSe multiple quantum wells. This leads to a difficult estimation of the microscopic biexciton dephasing in inhomogeneous broadened samples, and direct measurements of the dephasing of the ground state to biexciton transition have been reported only recently on a high-quality $GaAs/Al_xGa_{1-x}As$ single quantum well with negligible inhomogeneous broadening.[10] The temperature dependence of the dephasing of the exction-biexciton transition has to our knowledge not been reported in literature until now.

So far less attention has been payed to biexcitonic properties in $In_xGa_{1-x}As/GaAs$ heterostructures, in spite of their increasing importance for strained low-dimensional structures like quantum wires and quantum dots. In fact, $In_xGa_{1-x}As/GaAs$ quantum wells are qualitatively different from $GaAs/Al_xGa_{1-x}As$ quantum well, since the well material is an alloy and the barrier is a pure binary compound. A large number of publications have been devoted to this difference in the linewidth properties and in the exciton binding energy in these wells (for instance, see Refs. 11 and 12). It is interesting to investigate how this difference is reflected also in the biexcitonic properties.

In this work, we have studied the biexciton binding energy and dephasing in a series of $In_{0.18}Ga_{0.82}As/GaAs$ single quantum wells (SQW's) with well thicknesses between 1 and 4 nm by spectrally resolved, time-integrated four-wave mixing (TI-FWM). Sample temperatures and exciton densities were varied in the ranges 5–120 K and $4\times10^9-5\times10^{10}$ cm^{-2}, respectively. The well width dependence of the biexciton binding energy, and correspondingly the influence of the inhomogeneous broadening on the biexciton binding energy, are deduced. Also, the temperature dependence of the dephasing rate of the exciton-biexciton transition is reported.

The samples were grown by molecular-beam epitaxy on semi-insulating (100) GaAs substrates at 520 °C. The back-surfaces of all the samples have been polished in order to reduce the scattered light in the TI-FWM experiment that has been performed in reflection geometry using two excitation pulses in the directions $\mathbf{k_1}$ and $\mathbf{k_2}$ with a tunable relative delay time τ. The laser source was a self mode-locked Ti:sapphire laser providing 100 fs pulses at 76-MHz repetition rate. The pulses were chirp compensated and spectrally shaped in a pulse shaper. The laser spectrum was centered on the heavy-hole 1*s* exciton resonance, avoiding to excite the heavy-hole exciton continuum. The FWM signal was detected in the reflected $2\mathbf{k_2}$-$\mathbf{k_1}$ direction, spatially selected by pinholes and spectrally resolved by a spectrometer and an optical multichannel analyzer with a spectral resolution of 0.2 meV. The polarization of the exciting beams was adjusted by Babinet-Soleil compensators.

By using the polarization selection rules in the exciton-biexciton FWM,[13] we have clear evidence of biexciton resonances in the QW's, as shown in Fig. 1(a). Biexciton binding energies are directly extracted from the FWM response in the spectral domain, by comparing the energy position of the exciton resonance for cocircular polarized beams with the biexcitonic one for cross-linear polarizations. Note that, ac-

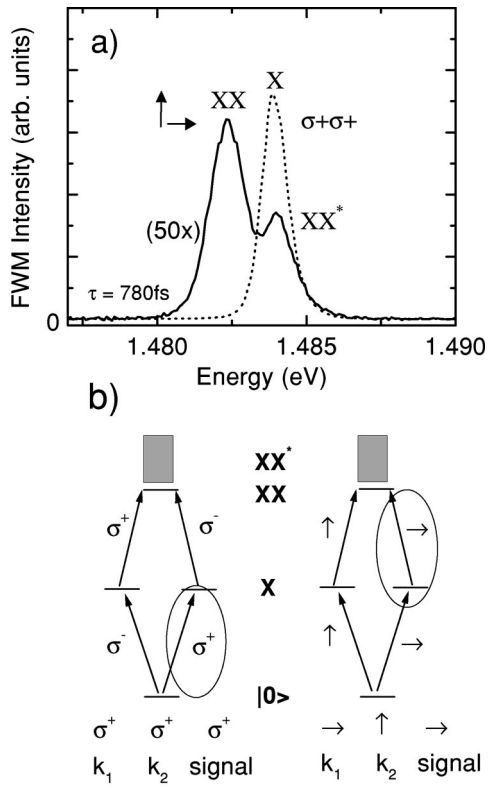

FIG. 1. (a) Spectrally resolved FWM in a 1.5 nm-wide well at 5 K, for cocircular (dotted line) and cross-linear (solid line) polarized excitation at 780 fs delay time. The excitation density was 1.5 $\times 10^{10}$ cm^{-2}. Exciton (X) and biexciton (XX) resonances and the relative scaling factor between the curves are indicated. (b) Scheme of the five-level model and the optical transitions for a circular (left) or a linear (right) polarized exciton basis. The transitions emitting FWM signal in the $2\mathbf{k_2}-\mathbf{k_1}$ direction are encircled.

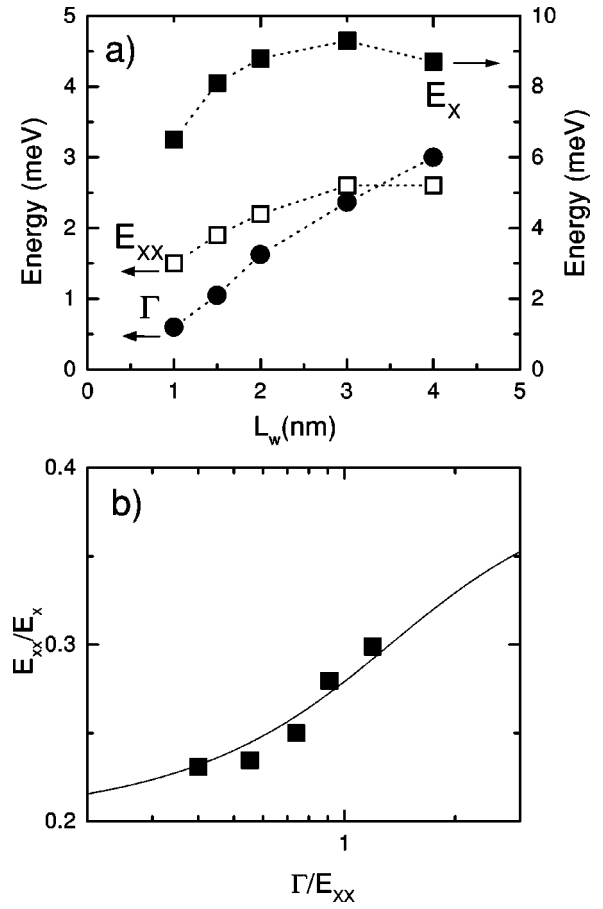

FIG. 2. (a) Inhomogeneous broadening Γ, exciton binding energy E_{X} and biexciton binding energy E_{XX} versus well width. (b) Ratio between biexciton and exciton binding energy versus ratio between inhomogeneous broadening and biexciton binding energy. The solid line is the calculated dependence according to Ref. 6. The parameter used are $\beta=0.4$, $\eta=0.2$, and $\hbar\omega_{\mathrm{loc}}=0.38\Gamma$.

cording to the five-level model[7] indicated in Fig. 1(b), the high-energy resonance observed for cross-linear polarizations corresponds to the exciton to unbound biexciton ($X-XX^*$) transition, that is shifted toward energies above the exciton resonance for strong inhomogeneous broadening.[9] The extracted binding energies are in agreement with the period of exciton-biexciton beats in delay-time observed for colinear excitation.

The measured well-width dependence of the biexciton binding energy (E_{XX}) in the investigated QW's is shown in Fig. 2(a), together with the exciton binding energy (E_{X}) and the full width at half maximum (FWHM) of the inhomogeneous broadening (Γ) of the exciton resonance. The exciton binding energies are deduced from photoluminescence-excitation measurements at low temperature while the inhomogeneous broadening is obtained by fitting the spectral profile of the exciton FWM response for long delay time, and it is in good agreement with linewidth analysis from photoluminescence and photoluminescence-excitation spectra.[14] The observed decrease of the exciton binding energy and the inhomogeneous linewidth with decreasing well width, opposite to what typically observed in GaAs/Al$_x$Ga$_{1-x}$As QW's, is a known property in shallow In$_x$Ga$_{1-x}$As/GaAs QW's, and is due to the penetration of the excitonic wave function into the binary barrier.[11,12] A similar trend is shown in Fig. 2(a) for the biexciton binding energy, again opposite to what reported in GaAs/Al$_x$Ga$_{1-x}$As QW's.[6] In order to isolate the effect of the localization on the biexciton binding from the effect of the confinement, the ratio between the biexciton and the exciton binding energies ($E_{\mathrm{XX}}/E_{\mathrm{X}}$) as a function of the ratio between the inhomogeneous broadening and the biexciton binding energy (Γ/E_{XX}) is plotted in Fig. 2(b). This ratio increases from 0.23 to 0.3 with increasing degree of localization (Γ/E_{XX}). The onset of the increasing ratio occurs when the biexciton binding energy is comparable to the inhomogeneous broadening of the exciton transition, similar to what observed in GaAs/Al$_x$Ga$_{1-x}$As QW's (Ref. 6) and in good agreement with a model proposed in Ref. 6 represented by the solid line in figure. This model treats the localization regime $\Gamma<E_{\mathrm{X}}$, which is realized in the investigated structures, and describes both the biexciton binding potential and the exciton localization as harmonic oscillator potentials. The total potential for the localized biexcitons then factorizes in two parts, separately dependent on the biexciton center-of-mass coordinate or the exciton-exciton relative coordinate. An analytical expression for the biexciton binding energy in this potential is obtained, given for QW systems by: $E_{\mathrm{XX}}/E_{\mathrm{X}}=\beta-\sqrt{\eta^2+\gamma^2}+\gamma$, were βE_{X} is the depth of the exciton-exciton interaction potential, ηE_{X} is the harmonic oscillator energy $\hbar\omega_{\mathrm{XX}}$ of the exciton-exciton relative mo-

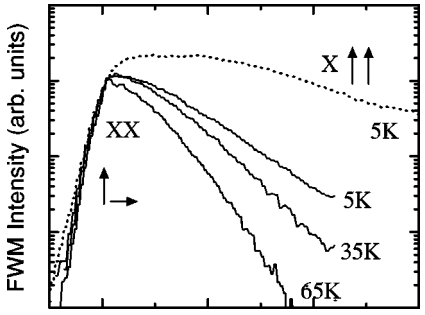

FIG. 3. Delay dependence of the FWM response at the biexciton resonance for cross-linear polarized beams, at different temperatures, in the 1 nm-wide well. The exciton density was 1.3 $\times 10^{10}$ cm^{-2}. The delay dependence at the exciton resonance for collinear polarized beams at 5 K is also shown (dotted line).

tion in the biexciton, and $\gamma = \hbar\omega_{\rm loc}/E_{\rm X}$ is the localization parameter, in which the harmonic energy $\hbar\omega_{\rm loc}$ of the localization potential is proportional to the inhomogeneous broadening. The solid line in Fig. 2(b) was obtained from this expression using the parameters indicated in the caption. Within this simple model, the increase of biexciton binding energy with increasing localization results from a quenching of the zero-point kinetic energy of the exciton-exciton relative motion in the biexciton.[6] The exciton-exciton repulsion at small distances present for electron-hole mass ratios different from unity, which is neglected in this treatment, is not changing the qualitative behavior within the weak localization regime.[6]

The delay dependence of the FWM response at the biexciton resonance for cross-linear polarized beams at different temperatures is reported in Fig. 3. The fast decay compared to the decay at the exciton resonance for collinear polarized beams at low temperature is mainly a consequence of the inhomogeneous distribution of biexciton binding energies, and is not simply related to a microscopic biexciton dephasing.[8] In fact, this decay can be simulated with zero biexcitonic homogeneous broadening.[8] In order to extract the temperature dependence of the microscopic biexciton dephasing from our experimental data, the ratio between the FWM trace at a given temperature and the trace at 5 K has been analyzed. This should cancel the effect of the inhomogeneous broadening of the biexciton binding energy, that is expected to be independent of temperature and related to disorder in the sample. By performing an exponential fit of this ratio, the additional intensity decay rate $\Delta\gamma$ due to phonon interaction has been deduced. No density dependence of this rate was observed in the range of the exciton densities investigated. In a photon echo experiment, the delay dependence of the FWM signal intensity at the exciton-biexciton transition has a decay rate twice the sum of the exciton and exciton-biexciton polarization decay rates. Let us indicate the exciton and exciton-biexciton FWHM homogeneous linewidths with γ_X and γ_{XX}, respectively. Then, the exciton and exciton-biexciton polarization decay rates are $\gamma_X/2\hbar$ and $\gamma_{XX}/2\hbar$, respectively. The phonon dephasing rate of the exciton-biexciton transition $\Delta\gamma_{XX}/2\hbar$ is then deduced by subtracting the exciton-phonon dephasing $\Delta\gamma_X/2\hbar$ from $\Delta\gamma/2\hbar$. Details about the exciton-phonon dephasing on the same samples are reported in Ref. 14. The phonon dephasing

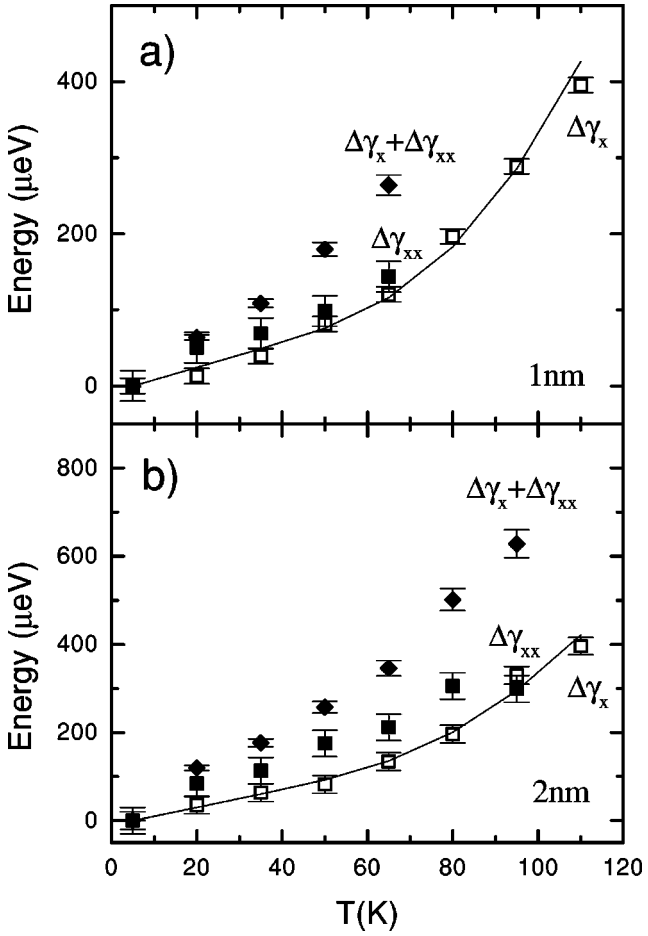

FIG. 4. Phonon dephasing rate of the exciton (open square) and exciton-biexciton (closed square) transitions versus temperature in the 1 nm- (a) and 2 nm-wide (b) well. Solid lines are fits of the exciton-phonon dephasing rates.

rates of the exciton and the exciton-biexciton transitions as a function of temperature are shown in Fig. 4 for 1 and 2 nm-wide wells. For the 1-nm wide well the analysis at the exciton-biexciton transition has been performed only up to 65 K, since for higher temperatures the system becomes mainly homogeneously broadened, and the FWM decay rate for positive delay at the biexciton resonance shows only the exciton dephasing rate. Solid lines in Fig. 4 are fits of the exciton-phonon dephasing rates according to the expression $\gamma_X = \gamma_0 + aT + bN_{\rm LO}$, where a,b are the acoustic and optical phonon coefficient respectively, and $N_{\rm LO}$ is the LO-phonon occupation number (for details see Ref. 14). From the fits we find $a = 1.6 \pm 0.2$ μ eV/K for the 1 nm-wide well and $a = 2.0 \pm 0.1$ μ eV/K for the 2 nm-wide well, in agreement with the expected well-width dependence of the acoustic phonon coefficient in thin QW's.[14,15] Concerning the phonon dephasing rate of the exciton-biexciton transition, we observe from Fig. 4 that $\Delta\gamma_{XX}$ and $\Delta\gamma_X$ have comparable temperature dependencies, with $\Delta\gamma_{XX}$ slightly larger then $\Delta\gamma_X$.

The interaction of excitons with acoustic phonons via deformation potential is given by the modulation of the band gap by the lattice deformation. Since exciton-biexciton and exciton transition energies differ only by the biexciton binding energy, both transitions involve a virtually equal region of the band structure and should undergo comparable defor-

mation potential interactions. This is in agreement with our experimental findings.

In summary, the binding energy and the temperature dependence of the dephasing of biexcitons in $In_xGa_{1-x}As/GaAs$ single quantum wells is reported. We find an increase of the biexciton binding energy from 1.5 to 2.6 meV with increasing well width from 1 to 4 nm. The ratio between exciton and biexciton binding energy increases from 0.23 to 0.3 with increasing inhomogeneous broadening. This is attributed to a quenching of the zero-point kinetic energy of the exciton-exciton relative motion in the biexciton, in agreement with a recent model proposed in literature. The temperature dependence of the dephasing of the exciton-biexciton transition due to phonon interaction is found to be comparable with the temperature dependence of the exciton-phonon dephasing, as explained by comparable deformation potential interactions for the exciton and the exciton-biexciton transitions.

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