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Optical dephasing of homogeneously broadened two-dimensional exciton transitions in GaAs quantum wells

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We report optical dephasing of two-dimensional excitons in GaAs-$Al_xGa_{1-x}As$ single quantum wells by means of time-resolved degenerate-four-wave mixing and transmission experiments in the temperature range below 80 K. The observed absorption linewidths correspond directly to the excitonic phase coherence times, confirming the homogeneous broadening of the excitonic absorption. A linear temperature dependence of the homogeneous linewidth indicates acoustic-phonon scattering as the broadening mechanism.

The optical properties of two-dimensional excitons in quantum wells have been the subject of extensive studies.[1-4] The key features are (i) a large blue shift of the excitonic transition energy with decreasing well width (quantum size effect) and (ii) an enhancement (up to a factor of 4) of the excitonic binding energy due to an increased electron-hole wave-function overlap.[1,2] The large binding energy explains the presence of excitonic features even at room temperature. Nonideal growth conditions accompanied by growth steps at the interfaces cause fluctuations of the quantum-well thickness. The local excitonic energy levels thus vary strongly in a lateral direction, and the excitonic line becomes inhomogeneously broadened.[2] The dynamics of the two-dimensional (2D) exciton are correspondingly dominated by the energy-relaxation processes arising from the well width fluctuations.[3,4]

These features have been confirmed in the frequency domain by means of transmission, photoluminescence excitation, and photoluminescence spectroscopy[1,2] as well as by coherent optical experiments in the time domain.[3,4] The typical features of the inhomogeneous broadening are broad exciton lines and a shift of the emission maxima to lower energies with respect to the absorption maxima. This so-called Stokes shift has its origin in the energy relaxation of the 2D exciton before emission. Time-resolved studies involving coherent optical experiments have clarified the dynamical aspect of exciton relaxation. Excitons excited above the absorption maximum are laterally delocalized and undergo fast momentum and energy relaxation because of scattering with acoustic phonons, impurities, and well width fluctuations. Below the center of the absorption line the exciton becomes localized and the relaxation processes (spectral diffusion) are much more inefficient. Thus the homogeneous linewidth, which is a dynamical measure of the interaction of the exciton with its surrounding, increases within the inhomogeneously broadened line for increasing photon energies.[4]

We report in this paper on the dynamics of 2D excitons in high quality GaAs *single* quantum wells where inhomogeneous broadening is negligible. By means of time-resolved degenerate-four-wave mixing (DFWM) and transmission spectroscopy we demonstrate that the 2D exciton transition is truly homogeneously broadened. The homogeneous linewidth exhibits a linear temperature dependence, revealing scattering with acoustic phonons as the important line broadening mechanism.

I. EXPERIMENTAL PROCEDURES

We used for our experiments high-quality GaAs-$Al_{0.3}Ga_{0.7}As$ single-quantum-well structures (SQWS) grown by molecular beam epitaxy on n^+ substrates. The samples were polished down to wedges of a few tens of microns. The thickness of the SQWS is measured by means of transmission electron microscopy to 27.7 nm (QW 1) and 13.5 nm (QW 2). The thickness of the buffer GaAs layer and the barrier layers are 194 and 14 nm, respectively. The DFWM experiments were performed using a synchronously pumped dye laser with Styryl 9. The autocorrelation width is about 3.7 ps and the width of the power spectrum 0.9 meV. The transmission experiments were performed using a tungsten iodine lamp and a single grating monochromator with an optical multichannel analyzer (nonintensified photodiode array cooled down to $-40\,^{\circ}C$) permitting accurate measurements of transmission changes in the range of 0.1%. The spectral resolution is about 0.025 nm.

II. RESULTS AND DISCUSSION

Figure 1 shows the transmission and photoluminescence spectra of the sample at 2 K. The transmission spectrum exhibits the strong absorption line of the $N=1$ exciton at 1.515 eV as well as on the high-energy side the clearly resolved and much weaker $N=2$ and $N=3$ excitonic absorption lines originating from the thin GaAs buffer layer. At 1.520 eV the $N=1$ heavy-hole (hh) and at 1.5218 eV the $N=1$ light-hole (lh) 2D exciton transitions of QW 1 are observed. The $N=1$ hh as well as the $N=1$ lh exci-

tonic transition of QW 2 are seen at 1.5356 and at 1.5419 eV, respectively. In addition, two weak and broad absorption lines are resolved 1.3 and 4.2 meV below the $N=1$ hh exciton of QW 2.

The lower part of Fig. 1 depicts the photoluminescence spectrum obtained at a low excitation intensity of 10 mW/cm^2. The GaAs buffer layer exhibits a triplet emission structure typical for bulk GaAs consisting of emission lines at 1.515 eV due to free excitons, at 1.5143 eV due to the neutral donor bound excitons (D^0X), and at 1.5124 eV due to neutral acceptor bound excitons (A^0X).[5] The same specific line structure is also found for the SQWS. Narrow emission lines centered at the absorption maxima and belonging to the hh exciton are observed for both SQWS, whereas the emission of the lh exciton is seen only for QW 1.[6] On the low-energy side, two broad emission lines are observed for both SQWS with an increased spacing from the hh exciton emission peak for decreased well width (QW 1: 0.8 and 3.7 meV, QW 2: 1.3 and 4.2 meV). These emission lines originating from QW 2 exactly correspond to the absorption lines. We assign these emission lines to recombination of excitons with neutral donors and acceptors in analogy to the bulk case.

The analysis of the spectra demonstrates the following.

There is *no* observable Stokes shift between emission and absorption maximum. This is the expected property of the ideal 2D exciton. A single energy level on a *global* and not a *local* spatial scale is responsible for excitation as well as for emission.

The emission linewidths correspond exactly to the absorption linewidths. Both are very narrow (full width at half maximum is equal to 0.45–0.5 meV) and consistent with a homogeneous line broadening demonstrating the high quality of our SQWS.

The impurity-related emission lines are much broader than the free exciton lines. The binding energy of the bound exciton is increased due to the confinement and is in agreement with recent predictions.[7,8] The relatively large width can be explained by assuming a random distribution of the impurities in the growth direction giving rise to a statistically varying binding energy.[8]

The relatively strong impurity absorption lines seen for QW 2 as compared to the bulk where the impurity-related absorption is hardly visible demonstrate the enhanced oscillator strength of the 2D exciton due to localization which was predicted recently.[9]

Figure 2 depicts the results of the time-resolved DFWM experiments on the 2D and 3D exciton lines obtained at exciton densities below 5×10^9 cm^{-2} and at the temperature of 2 K. Two pulses of 2.8-ps duration with a variable delay τ_{12} are tuned into the excitonic resonance and focused onto the sample. The second pulse probes the coherent macroscopic polarization of the exciton ensemble left from the first pulse, thus giving rise to the coherent

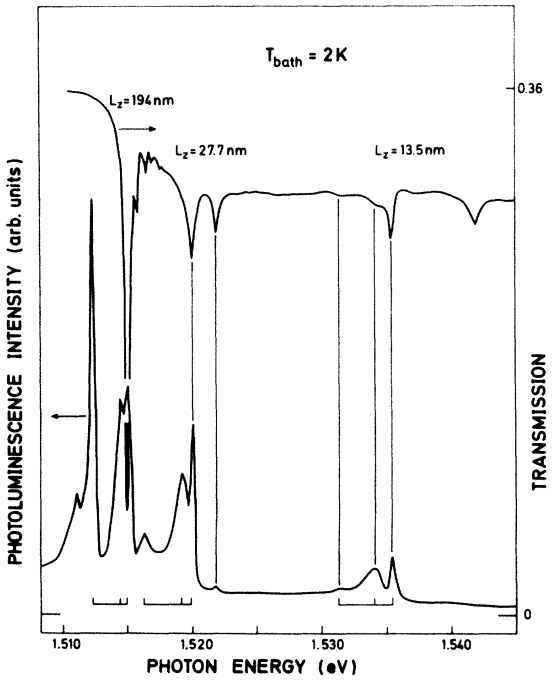

FIG. 1. Transmission (upper trace) and photoluminescence (lower trace) spectra of a sample containing two GaAs SQWS and a buffer (194 nm) GaAs layer obtained at 2 K.

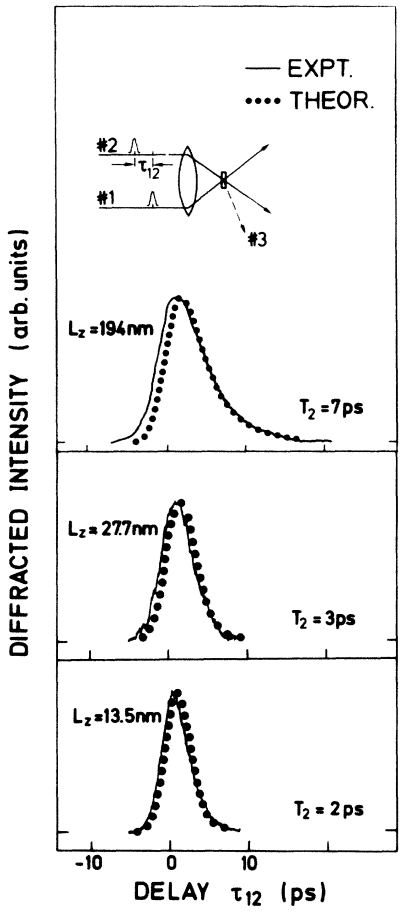

FIG. 2. Experimental (solid line) and theoretical (dotted line) diffraction curves of the GaAs buffer layer and the two SQWS at 2 K. The fit yields the phase coherence time T_2. The inset shows the schematical excitation arrangement of the two-pulse DFWM experiment.

emission (pulse No. 3) which in turn measures directly the phase coherence of the excitons. A fit to the experimental diffraction curve performed by solving the optical Bloch equations in the small signal limit[10] yields the phase coherence time T_2 of the exciton transition. We find $T_2=7\pm0.5$ ps for the bulk GaAs and $T_2=3\pm0.5$ ps for QW 1 and $T_2=2\pm0.5$ ps for QW 2.

The corresponding phase-coherence times obtained from the linewidth Γ (assuming homogeneous broadening) given by $T_2=2/\Gamma$ are $T_2=6.5\pm0.2$ ps for the bulk GaAs (Ref. 11), $T_2=2.8\pm0.1$ ps for QW 1, and $T_2=2.6\pm0.1$ ps for QW 2. The good agreement between the linewidth and the phase coherence time confirms the homogeneously broadened nature of the exciton line in our samples.

In Fig. 3 we show the homogeneous linewidths obtained from the transmission spectra versus the temperature. At temperatures below 80 K the homogeneous linewidth Γ depends linearly on the temperature T for all the exciton transitions:

$$\Gamma=\Gamma_0+\gamma^* T \ .$$

The linear scattering coefficients γ of the hh and lh transitions of QW 1 are 10 μeV/K and 5 μeV/K for QW 2. The linear scattering coefficient for the 3D exciton in the buffer layer (not shown in Fig. 3) is about 17 μeV/K.[12]

This linear temperature dependence demonstrates for the first time that optical dephasing of the *delocalized* 2D excitons is caused by *one-phonon* scattering. The resonantly excited excitons with negligible wave vector parallel to the layer have vanishing kinetic energy. By absorption of an acoustic phonon the exciton is scattered up along the two-dimensional exciton dispersion curve. This one-phonon anti-Stokes scattering is a T_1 process because the final state differs from the initial one. The linear temperature dependence also indicates that higher-order phonon scattering involving two or more phonons are negligible. Pure dephasing processes necessitating at least two phonons do not seem to be important.

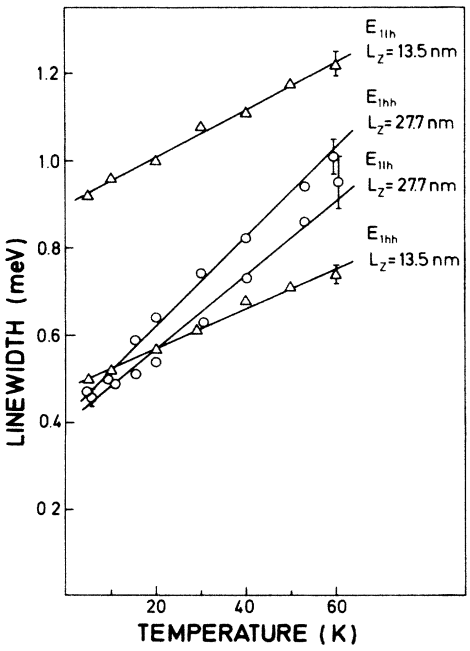

FIG. 3. Homogeneous linewidth (full width at half maximum of the absorption peak) vs temperature for the heavy-hole (hh) and light-hole (lh) exciton transition of the two SQWS.

Our findings differ from previous luminescence studies exhibiting much broader and probably inhomogeneously broadened lines.[13] Therefore the temperature dependence did not show unambiguously any influence of acoustic phonon scattering.

Our main results can be summarized as follows.

(a) The scattering rates of the hh and the lh exciton are the same within 10%.

(b) The scattering rate decreases with decreasing well width in contrast to recent theoretical predictions.[14]

A theoretical understanding of these findings is not yet possible, partly because a realistic calculation of the scattering rates has to implement the quasi-2D character of the excitons in the quantum well with a finite thickness. Detailed knowledge of the excitonic wave function is needed which is impeded by the complicated electron-hole Coulomb interaction. The second problem arises from the dependence of the scattering rates on the exciton mass. Following theoretical arguments the masses of the 2D excitons in the quantum well are expected to be changed compared to the bulk material. Experimentally, however, the masses are not known accurately enough.

Published theoretical results are quite controversial. Whereas the theory of Takagahara[14,15] predicts scattering rates to depend strongly on the masses, the approach of Lee, Koteles, and Vassell[13] reveals more or less the same scattering rates (within 20%) for the hh and the lh exciton.

The observed temperature-independent scattering contribution Γ_0 can be attributed mainly to scattering with impurities in particular neutral donors.

The much faster optical dephasing of the 1h exciton of QW 2 is due to the energetic overlap with the continuum states of the hh exciton. Because the electron-hole Coulomb interaction mixes not only the different electronic subbands but also the different valence bands all the excitons are coupled together. The resonant interaction between the discrete 1h exciton and the hh exciton continuum leads to a Fano-type resonance, which in our case is nearly symmetric, implying a weak interaction. The optical dephasing of the 1h exciton then is given by the coupling coefficient as well as by the much more rapid dephasing of the continuum states which is too fast to be detected with our present long pulse duration.

III. CONCLUSIONS

We have reported on the ultrafast dynamics of 2D excitons in GaAs SQWS by means of transmission and time-resolved DFWM experiments. We have observed for the first time homogeneously broadened exciton transitions due to the high quality of our samples. Thus, the optical dephasing time is equivalent to the measured linewidth. We find that the homogeneous linewidth is limited by scattering with acoustic phonons below 80 K. The residual temperature-independent scattering is attributed to scattering with impurities.

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