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# Exciton scattering in quantum wells at low temperatures

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The diffusion and the lifetime of excitons were determined via laser-induced gratings in high-quality GaAs/$Al_xGa_{1-x}As$ multiple quantum wells. The diffusion is governed by the interaction of the excitons with the interface roughness and with acoustic phonons. The exciton scattering rate was compared with the dephasing rate observed in subsequently performed photon echo experiments. The influence of interface-roughness scattering on the dephasing was found to be weak and independent of temperature for this coherent process. The exciton-phonon coupling deduced from diffusion shows an excellent agreement with theory, whereas the dephasing rates determined in our work and former contributions were found to agree only roughly with theoretically calculated values.

## INTRODUCTION

The optical properties of GaAs/(Al,Ga)As quantum wells (QW's) are dominated by excitonic resonances. In quasi-two-dimensional GaAs wells, the exciton binding energy is up to 3 times higher than in bulk material, depending on the well thickness.[1]

Although much work has been done on excitons in QW's during the last decade, experimental and theoretical results of the exciton scattering rate at low temperatures are still inconsistent. In photon echo experiments[2] a linear increase of the homogeneous excitonic linewidth with temperature was found, as predicted by theory. But the absolute values of the experimental findings differ. In addition, the predicted proportionality of the exciton phonon scattering to the inverse of the well thickness $1/L_z$ due to the changes in the density of states,[3] could not be confirmed.

The exciton diffusion at temperatures below 30 K could only be measured until now in comparatively thick samples (270 Å).[4] In these QW's the interface roughness scattering is reduced. In contrast to the results from dephasing measurements, the acoustic-phonon scattering rate, deduced from the diffusion constants at different temperatures, are in agreement with the theory.

In this contribution, we analyze the scattering rate of excitons in a multiple-quantum-well (MQW) structure in dephasing and diffusion measurements at temperatures below 30 K. We confirm the results of Ref. 4 about interface-roughness and deformation-potential scattering and demonstrate the significant differences of the exciton scattering rates measured in the coherent dephasing and the incoherent diffusion experiments.

## SAMPLES AND EXPERIMENTAL SETUP

The sample under investigation was grown by molecular-beam epitaxy on a [001]-orientated GaAs substrate which was subsequently removed by selective chemcial etching. The MQW consists of 20 periods of 103-Å GaAs layers between 150-Å $Al_{0.3}Ga_{0.7}As$ barriers. The spectral position of the lowest excitonic resonance is 1.5432 eV and the full width at half maximum (FWHM) of the absorption line is 0.9 meV.

The sample was excited with pulses of about 7-ps (FWHM) duration and 82-MHZ repetition rate, generated by a mode-locked argon-ion laser which pumps a tunable dye laser (Styryl 8). All measurements were performed in forward-scattering direction and the sample was mounted in a variable-temperature He cryostat.

For the dephasing measurements the laser is split into two beams, which are focused on the sample with a mutual angle $\theta$ between them and a spot size of abut 50 $\mu$m. They interfere coherently and set up a grating with a period of $\Lambda=\lambda/2\sin(\theta/2)$, where $\lambda$ is the wavelength of the laser tuned to the heavy-hole exciton (hhx) resonance of the MQW. Behind the sample, the diffracted signal is focused on the entrance slit of a spectrometer with a resolution of 0.2 meV and detected with an optical multichannel analyzer system. The signal decays exponentially with the time delay $t_{12}$ between the exciting pulses. The decay time $\tau_s$ determines the dephasing time $T_2=4\tau_s$,[5] if we consider the hhx resonance as an inhomogeneously broadened two-level system.

The short $T_2$ time ($\simeq 1$ ps) and the homogeneous broadening of the hhx resonace in Ref. 6 which are in contrast to our data ($T_2 \geq 50$ ps) might be a result of the

 

excitation conditions in a similar way as outlined in Ref. 7: under femtosecond excitation the spectral width of the laser pulse increases. Thus the excitonic $T_2$ time decreases partly due to the enhancement of the scattering probability with increasing excess energy. Furthermore, under these experimental conditions the intensity during a single laser pulse has to be increased, too, so that exciton-exciton collisions also contribute to the dephasing.

To determine the diffusion constant $D$ of the excitons the two pump pulses arrive simultaneously and the decay of the laser-induced grating is measured by the diffraction of a third read-out beam delayed by $t_{13}$ and detected by the apparatus described above. The intensity of the diffracted beam is proportional to the square of the grating amplitude. The signal decays after a short, coherent signal with $t_{13}$ due to recombination and diffusion of the excitons, with a characteristic time $\tau_s$:[8]

$$1/\tau_s = 2/T_1 + 8\pi^2 D/\Lambda^2 \ . \tag{1}$$

By changing the grating constant $\Lambda$, the excition diffusion constant $D$ and the exciton lifetime $T_1$ both can be determined from Eq. (1).

## EXPERIMENTAL RESULTS AND DISCUSSION

The experimental results show an increase of the diffusion coefficient and of the lifetime with temperature, as shown in Fig. 1. The solid line is a fit considering the scattering rates and will be discussed below. The dashed line is a linear fit to the excitonic lifetimes as a function of temperature with a slope of 100 ps/K. The lifetime increases due to increasing thermal occupation of nonradiative states. Only excitons with an in-plane wave vector smaller than the wave vector of a photon with the transition energy may decay radiatively.[9] In addition localized states enhancing radiative and nonradiative recombination are ionized with increasing temperature.[10,11]

The momentum relaxation rate $1/\tau$ can be deduced from the diffusion constant according to the Einstein relation

$$1/\tau = k_B T/mD \ . \tag{2}$$

$m$ denotes the effective exciton mass and $T$ the exciton temperature. The in-plane mass of a exciton in a [001]-orientated GaAs quantum well is $0.206 m_0$,[12] where $m_0$ is the free-electron mass. It is interesting to consider if the same scattering events contribute to both momentum relaxation and dephasing.

Figure 2 shows the temperature dependence of the momentum relaxation rates $1/\tau$ (circles) of the excitons in dependence on the temperature deduced from diffusion and the dephasing rates $1/T_2$ (squares). At 2 K, both rates are about 50 $ns^{-1}$. The dashed line is a fit obtained from linear regression to the dephasing results. The increase of the dephasing rate with temperature shows a slope of $(2.6\pm0.7)\times10^9$ $s^{-1}K^{-1}$. The low-temperature limit ($T=0$) is 40 $ns^{-1}$, representing the residual interface-roughness scattering.

The momentum relaxation rate increases much faster with temperature and shows a nonlinear dependence. An analysis of the relaxation rates clarifies the origin of the temperature dependence and the differences to the dephasing. The solid line was calculated according to a contribution of interface-roughness scattering and acoustic-phonon scattering.[4]

The contribution from the exciton acoustic-phonon scattering rate can be approximated by[13]

$$1/\tau_{pn} = AT/L_z \ . \tag{3}$$

The temperature dependence is a linear approximation of the phonon number due to the Bose distribution in the high-temperature limit. The exciton–acoustic-phonon scattering is proportional to the quasi-two-dimensional density of states in the GaAs wells which is independent of energy, and scales with the inverse well thickness $1/L_z$. The coefficient $A$ is proportional to the exciton–acoustic-phonon interaction strength. It can be calculated for the deformation-potential scattering from the material parameters:[4,13,14] $A=50$ m/s K. If the scattering would only be due to the deformation-potential coupling with phonons as in Eq. (3), the diffusion constant should be independent of temperature. Thus the temperature dependence of the diffusion coefficient is due to additional scattering processes.

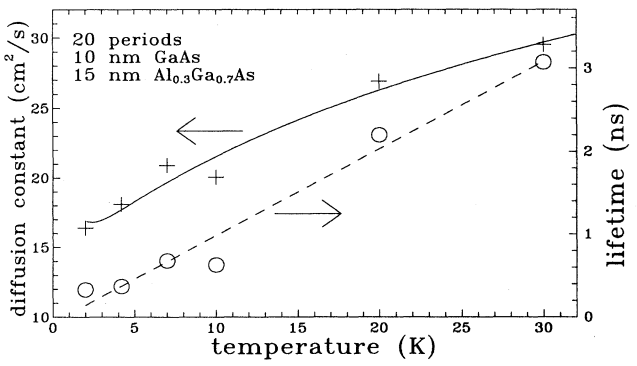


FIG. 1. Diffusion coefficient of excitons (crosses) in dependence on the sample temperature. The solid line represents a fit to the scattering rates (see Fig. 2). Exciton lifetime (circles) with a linear fit (dashed line).

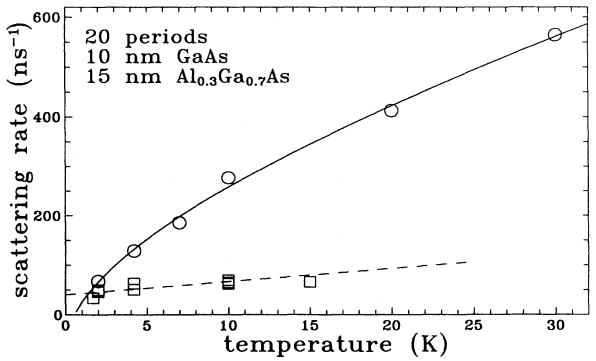


FIG. 2. Scattering rate of thermalized excitons (circles) with a second-order polynominal fit in $\sqrt{T}$ (solid line) according to Eq. (5). Scattering rate of coherent (squares) excitons with a linear fit (dashed line).

The interface roughess scattering ($1/\tau_{ir}$) is the most important scattering process in a MQW with GaAs layer thickness of about 100 Å at low temperatures.[4,15,16] It is caused by the fluctuation of the well thickness by one monolayer.

Calculations in the perturbation limit have been worked out for the case of modulation-doped quantum wells by Sakaki *et al.*,[17] showing a scattering rate with a Gaussian temperature dependence. The step in the quantization energy of the excitons in the thinner well regions is about 3 meV for our sample. It is undoped, so that the kinetic energy of the excitons corresponds to $k_B T$. The scattering process of excitons at the islands at temperatures below 30 K cannot be treated in the perturbation limit. We assume here, in a classical limit, that the exciton is a particle being scattered on every island. The mean free path of the excitons is independent of temperature. Thus interface-roughness scattering depends on the velocity of the exciton being proportional to the number of scattering centers which can be reached per time unit.

Additionally, the interface-roughness scattering increases with shrinking well thickness due to the increasing amplitude at the interface. A proportionality of the scattering rate to $1/L_z^2$ was supposed by Tsen, Sankey, and Morkoç.[4] Thus, the interface-roughness scattering can be expressed by

$$1/\tau_{ir}=(R/L_z^2)\sqrt{T}\ , \tag{4}$$

where $R$ is a constant depending on the quality of the interface. Experimentally Hillmer *et al.*[18] found a slighly larger dependence of the interface-roughness scattering on the GaAs layer thickness: $1/L_z^{2.5}$.

To test these two dominating scattering processes we tried the following fit, represented by the solid line in Fig. 2:

$$1/\tau=C+1/\tau_{ir}+1/\tau_{pn}=C+R\sqrt{T}/L_z^2+AT/L_z\ . \tag{5}$$

From the fitting procedure we obtain $A=54$ m/s K, $R=9\times10^{-6}$ m$^2$/s$\sqrt{\text{K}}$, and $C=-6.6\times10^{10}$ s$^{-1}$. The model yields an exciton-phonon interaction strength $A$ which is close to the theoretical prediction (see above). The parameters $A$ and $R$ agree with former results determined in a 275-Å MQW.[4] The small negative intercept $C$ (see Fig. 2) is an artifact, probably resulting from the linear approximation of the Bose function.[19] We have also neglected defect scattering processes caused by impurities and dislocations in the GaAs layer. Hillmer *et al.*[18] found a defect scattering rate decreasing with increasing temperature. A small contribution of these scattering processes may also enhance the negative intercept of our fit.

Neglectig the phonon scatteirng, the mean-free path $l$ of excitons is given by

$$l=(L_z^2/R)\sqrt{k_B/m}=640\ \text{Å}\ . \tag{6}$$

$l$ is comparable to the area of monolayer flat regions in our sample. The mean-free path of excitons is larger than the exciton diameter [about 100 Å (Ref. 3)] so that our ansatz for the interface-roughness scattering is justified.

The difference between momentum relaxation and dephasing seems surprising at first but we find a rather reasonable explanation: the coherent excitions suffer only scattering with acoustic phonons. Their momentum $\mathbf{k}_\parallel$ and thus their velocity in the plane of the quantum well are almost negligible due to the optical excitation. Therefore, the interface roughness scattering is very small. To feel this scattering process the excitons must first thermalize with the lattice by some scattering processes with phonons, but then their phase coherence is already gone and they no longer contribute to the photon echo signal. This idea is confirmed by the fact that the scattering with acoustic phonons in Eq. (5) alone leads to a similar slope as found for $1/T_2$. From the slope of the dephasing versus temperature, we can deduce an exciton-phonon coupling of $A=31$ m/s K. The contribution of the interface-roughness scattering to the dephasing for coherent excitons is independent of temperature and corresponds to the intercept.

The contribution of exciton-phonon scattering rates to dephasing and momentum relaxation of excitions can now be determined. We like to emphasize that, in general, the results deduced from diffusion measurements show a better agreement with the theoretical prediction; however, this method is more indirect and the interface-roughness scattering has to be substracted first.

Figure 3 shows the exciton-phonon coupling $(\tau_{pn}T)^{-1}=AL_z^{-1}$ in dependence on the well thickness. The results of this contribution (circles) are compared with previous works on this field[2,4] (triangles). The solid symbols represent results obtained from diffusion measurements. The solid line shows the theory according to Eq. (3). The open symbols show the results of photon echo experiments. Whereas the phonon-scattering rate of thermalized excitons is well described by the theory, the dephasing rate is only in rough agreement. The open symbols show even a different slope with temperature.

These differences may be tentatively explained by the following considerations. The energetic distribution of coherent excitons is determined by the spectral line shape of the exciting laser beam, and the absorption spectrum which is strongly sample dependent. If the linewidth of the laser pulse is significantly smaller than the hhx reso-

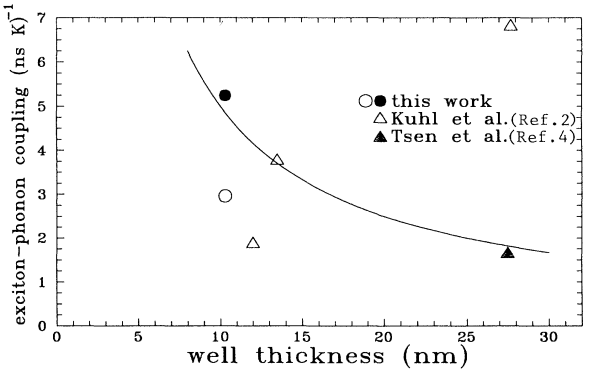


FIG. 3. Exciton-phonon coupling determined by diffusion (filled symbols) and dephasing (open symbols) measurements. The solid line represents the theory according to Eq. (3).

nance the dephasing also depends on the excitation energy due to localized states.[20] Thus satisfactory agreement with theory can only be expected if the hhx resonance is narrow, and if no Stokes shift between absorption and luminescence appears. Furthermore, in high-quality samples it is not always obvious if the hhx resonance is more inhomogeneously or homogeneously broadened, depending on the excitation conditions (see above). Since we use very-high-quality samples and rather weak laser pulses, we think that the data of Ref. 4 and ours are more reliable.

## CONCLUSION

We have determined the exciton scattering rates at low temperatures. The results emphasize the temperature dependence of the interface-roughness scattering of thermalized excitons. The exciton-phonon interaction strength was found to coincide with theoretical predictions and previous measurements on thicker quantum wells.

The phonon scattering rate could be related to the phase coherence time in subsequently performed photon echo experiments, where the interface roughness scattering is independent of temperature.

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