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PHASE COHERENCE OF 2D EXCITONS IN GaAs-AlGaAs QUANTUM WELLS

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Abstract - We have studied photon echoes on a picosecond timescale from 2D excitons in GaAs-AlGaAs quantum wells. A theoretical model is used which considers phase relaxation during excitation. We find considerable deviations from the usual photon echo behavior for pulse lengths comparable to the phase coherence time. Line shape analyses of our experimental data yield the phase coherence times which are found to vary strongly across the inhomogeneously broadened exciton line.

I - INTRODUCTION

Coherent optical experiments in the time domain on semiconductors reveal a wide range of relaxation times /1-6/. Strongly interacting electron-hole pairs excited to high densities by band-to-band transitions show phase relaxation times of the order of a few hundred femtoseconds /1/. Photon echo experiments on impurity-related bound excitons have revealed a much weaker exciton-exciton interaction with corresponding relaxation times up to a hundred picoseconds /2/. Recently, phase relaxation times in the intermediate range have been observed by photon echoes from Wannier-like 2D excitons in GaAs-AlGaAs quantum wells /3/. 2D excitons in quantum wells are inhomogeneously broadened due to well width fluctuations along each well. Relaxation times are expected to depend strongly on the photon energy within the inhomogeneously broadened exciton line because of the spectral dependence of the exciton scattering rates /4/.

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Direct extraction of the relaxation times from the experimental data becomes difficult when the pulse length and relaxation time are comparable.

It is the purpose of this paper to discuss the influence of a finite pulse length on the total photon echo intensity as well as its time dependence on the delay between the exciting pulses. The experimental data are analyzed by means of a line shape analysis yielding the phase relaxation time.

II - THEORETICAL

In order to analyze the experimental data for a phase relaxation time T_2 and a population relaxation time T_1 comparable to the pulse length t_0 one has to incorporate relaxation during excitation in a third order density matrix theory /5/. The polarization in the $2k_2-k_1$ direction produced by the pulses $E_1(t)$ and $E_2(t-\tau_{12})$ with corresponding wave vectors k_1 and k_2 and frequency ω is given by:

$$P(t, \tau_{12}) \propto \int_0^\infty g(\omega_0) \int_{-\infty}^\infty \int_{-\infty}^\infty dt' dt'' dt''' d\omega_0 \cdot \\ \{ E_2(t' - \tau_{12}) E_2(t'' - \tau_{12}) E_1^*(t''') e^{i(\omega_0 - \omega)(t' + t'' - t''' - t)} \\ + E_2(t' - \tau_{12}) E_1^*(t''') E_2(t''' - \tau_{12}) e^{i(\omega_0 - \omega)(t' - t'' + t''' - t)} \} \cdot \\ e^{[(1/T_2 - 1/T_1)(t' - t'') + (t''' - t)/T_2]}$$

where $g(\omega_0)$ is the exciton distribution excited by the pulses.

The total diffracted energy is: $U(\tau_{12}) \propto \int_{-\infty}^\infty |P(t, \tau_{12})|^2 dt$. The time dependence of the photon echo is measured by the cross-correlation with a reference beam $E_0(t-\tau_{13})$ and is given by:

$$C(\tau_{12}, \tau_{13}) \propto \int_{-\infty}^\infty |P(t, \tau_{12})|^2 |E_0(t - \tau_{13})|^2 dt.$$

To calculate the scattered intensity when the relaxation times are comparable to the pulse length a knowledge of the electric field amplitudes is necessary. Since these cannot be measured directly we assume Gaussian pulses: $E_i(t) \propto e^{-(t/t_0)^2}$, $i=0,1,2$ with a pulse length parameter of $t_0=7.6$ ps to fit the autocorrelation width. The exciton distribution is also given by the power spectrum of our nearly transform-limited pulses.

Fig.1 shows the calculated diffracted intensity versus delay for three characteristic cases. If $T_2, T_1 \ll t_0$ the scattering curve $U(\tau_{12})$ is symmetrically centered around zero delay. The scattering curve reflects only the pulse shape properties /6/.

For $T_2, T_1 \gg t_0$ $U(\tau_{12})$ is strongly asymmetric and reflects the usual photon echo case /7/. The tail is approximately exponential ($\sim e^{-4\tau_{12}/T_2}$) and normally used to determine T_2 . The shift of the maximum towards increasing delay is an effect of the finite pulse length and does not depend on T_2 . $U(\tau_{12})$ is independent of T_1 .

In the intermediate case, where both T_2 and T_1 are comparable to t_0 , the position of the maximum of $U(\tau_{12})$ depends on T_2 and T_1 . With increasing T_2 the maximum shifts to longer delays up to a saturation value given by t_0 . If T_1 decreases for a given T_2 down to $T_2/2$ (for lifetime limited phase relaxation) the maximum shifts to shorter delays. The asymmetric tail depends only on T_2 but not in a simple exponential manner.

The reason for this behavior can be seen in Fig.2 where the maximum of the cross-correlated signal is plotted versus delay τ_{12} for the three case mentioned above. For $T_2, T_1 \ll t_0$ the diffracted signal is only produced in the temporal overlap of the two pulses. The time dependence of the position of the signal maximum is $\sim \tau_{12}/2$. Thus, the total intensity is completely given by the pulse shape. This case corresponds to the degenerate-four-wave-mixing experiments performed above the band gap of a semiconductor as described in Ref.6.

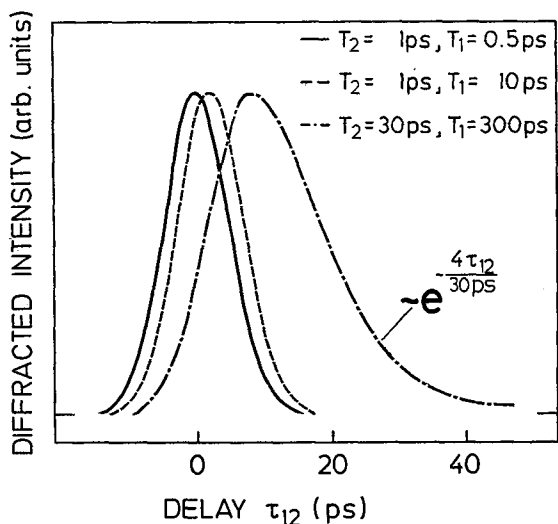


Fig. 1 - Diffracted intensity versus delay τ_{12} calculated for three different values of the phase relaxation time T_2 and the population relaxation time T_1 .

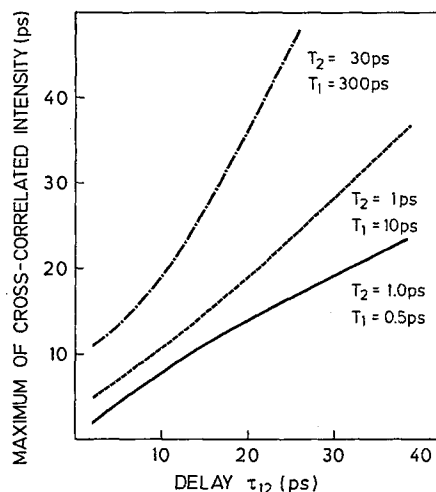


Fig. 2 - Maximum of the cross-correlated signal versus delay τ_{12} calculated for three different T_2 and T_1 .

For $T_2, T_1 \approx t_0$ the diffracted pulse shows a time dependence $\sim \tau_{12}$. Asymptotically a $2\tau_{12}$ dependence is reached. The corresponding scattering curve shows a decay time between τ_{12} and $2\tau_{12}$, depending on the absolute value of τ_{12} . For short delays the signal is delayed by an amount depending on the pulse length.

For $T_2, T_1 \gg t_0$ the usual photon echo time dependence of $2\tau_{12}$ is reached for $\tau_{12} \gg t_0$. Thus, the exponential tail in the corresponding scattering curve reflects a $e^{-4\tau_{12}/T_2}$ dependence.

III - EXPERIMENTAL RESULTS

Fig.3 shows experimental scattering curves for three different spectral positions within the inhomogeneously broadened heavy-hole transition of a 100 Å multi-quantum well sample. Near line center the scattering curve exhibits a slight asymmetry. The maximum is close to zero delay. With decreasing photon energy the asymmetry becomes stronger and the maximum shifts to longer delays.

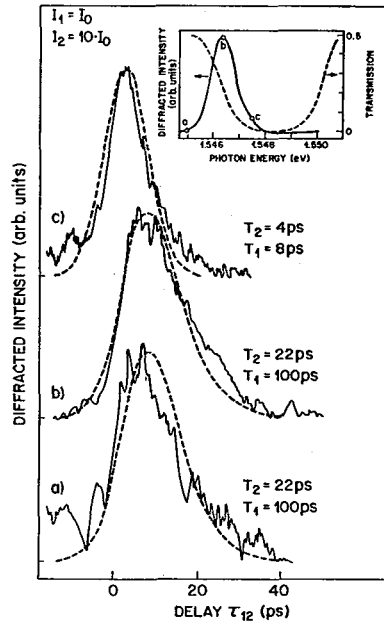


Fig. 3 - Experimental (full line) and theoretical (dashed line) scattered intensity as a function of delay for three different photon energies. The spectral positions of the exciting pulses are within the heavy-hole exciton transition and are shown in the inset. Also shown is the transmission spectrum and the diffracted intensity versus photon energy. I_0 is about $\ln J/\text{cm}^2$. The polarizations of the pulses are orthogonal.

The data can be fit to the above theory with T_1 and T_2 as parameters. T_1 has only a significant influence on the scattering curve near the line center. The small observed shift of the maximum can be fit only by taking $T_1 \approx t_0$.

Our data clearly show that there is a range of phase coherence times within the inhomogeneous 2D exciton line. For excitation near line center the short phase coherence time is dominated by population decay (spectral diffusion) whereas well below line center the phase coherence is conserved up to 22 ps. This supports the idea of a transition from 2D excitons delocalized above line center with corresponding strong dephasing interactions and localized excitons below line center with weaker interactions.

IV - CONCLUSIONS

We have studied photon echoes from 2D excitons in GaAs-AlGaAs quantum wells. Line shape analyses are used to determine the phase coherence times of 2D excitons within the inhomogeneously broadened line. We find that for excitation at line center the phase coherence time is about 4ps whereas it increases up to 22 ps for excitation well below line center.

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