22nd International Conference on

THE PHYSICS OF SEMICONDUCTORS

Volume 1

Vancouver, Canada

August 15 – 19, 1994

Editor

DAVID J. LOCKWOOD
National Research Council of Canada
Institute for Microstructural Sciences
Ottawa, Canada

World Scientific
Singapore • New Jersey • London • Hong Kong

Proceedings Vancouver '94, ICPS 22

HIGHER ORDER PHOTON ECHO IN FOUR-WAVE-MIXING EXPERIMENTS

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Four-wave-mixing experiments on the acceptor bound exciton complex in CdS are reported. In the first order diffracted signal ($2k_2$-k_1) a photon echo is resolved at 2τ proving inhomogeneous broadening. Assuming a $T_2/4$ decay a dephasing time T_2 of about 800 ps is deduced, which is not limited by energy relaxation processes (T_1 = 1000 ps). In second diffraction order ($3k_2$-$2k_1$) a photon echo occurs at approximately 3τ and the time-integrated signal decays much faster than in first order. The experimental results are explained taking into account propagation effects using the wave equation for the electric field together with the density matrix equations for noninteracting two-level systems.

1. Introduction

Four-wave-mixing (FWM) represents one of the most powerful tools to investigate the coherent dynamics in semiconductors. However, only the first order diffracted signal has been analyzed in semiconductors up to now. The purpose of this contribution is to extent the investigations to the second order diffraction.

Coherence times of some hundred ps are reported for bound excitons in CdS[1,2,3] and CdSe.[4] These systems can be regarded as independent two-level systems not subject to the fast dephasing processes observed for free carriers as well as free and weakly localized excitons. However, the phase relaxation times T_2 deduced from the experiments are still not limited[2,3] by recombination processes.

Here, we present degenerate-four-wave-mixing (DFWM) experiments on (A^0,X) complexes in CdS with special emphasis on the second order diffracted signal. The results are used to develop a theoretical description including propagation effects in an absorbing media.

2. Experimental Setup

The DFWM experiments are performed at 1.8 K using 2 ps pulses provided by a dye laser synchronously pumped with the third harmonic of an actively mode-locked Nd:YAG laser. The diffracted signals are detected either time-integrated using Si-diodes or time-resolved using a multichannelplate photomultiplier and time-correlated single photon counting. The samples are nominally undoped bulk CdS platelets of about 15 µm thickness having the c-axis in the sample plane. The crystals investigated are selected for about 30% I_1 absorption (formation of an (A^0,X)-complex).

3. Experimental Results

Fig.1 compares the time-integrated DFWM signal in first ($2k_2$-k_1) and second ($3k_2$-$2k_1$) order of diffraction observed exciting in the I_1 absorption at 2.5356eV in CdS. As obvious

from fig.1 the first order signal decay is nonexponential but it becomes almost monoexponential for long delay times. This has been discussed in detail in previous papers[2,3] giving typical dephasing times T_2 around 800 ps at T = 1.8 K and at low excitation densities. In second diffraction order we observe a correlation peak at zero delay and a DFWM signal which rises slower but decays about 2.5 times faster than in first order. The modulation at short delay times τ is attributed to polarization interference.[5] The investigated CdS samples show an I_1 doublet (ΔE = 175 μeV) caused by the formation of bound exciton complexes at different neutral acceptors. Fig.2 compares time-resolved the diffracted signals. In first order the photon echo occurs a few ps before 2τ and its temporal width is caused by the experimental setup. In contrast, in second order the photon echo has its maximum approximately 45 ps before 3τ and is broader than the experimental resolution. A temporal width of 80 ps is estimated.

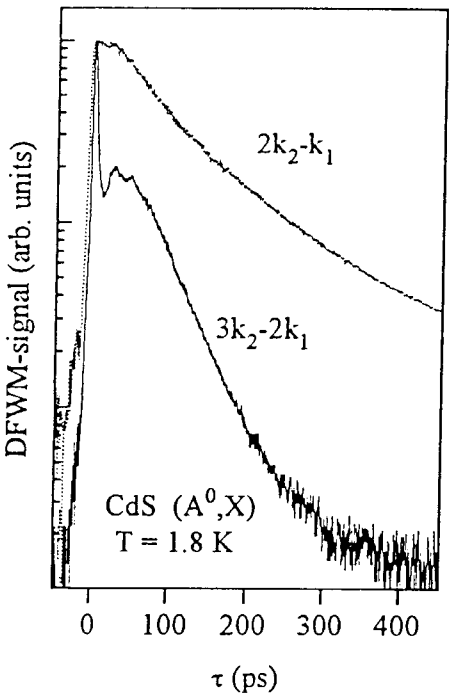

Fig.1: Normalized time-integrated DFWM signal in first ($2k_2$-k_1) and second ($3k_2$-$2k_1$) order of diffraction for the (A^0,X) complex in CdS.

4. Modeling

The investigated neutral acceptor bound exciton complex (A^0,X) represents a localized excitation in CdS. Its Bohr radius is orders of magnitude smaller than the mean distance of the acceptors. Thus, to model our DFWM experiments we consider an ensemble of inhomogeneously distributed noninteracting two-level systems which are described by the density matrix equations. In order to account for the higher order diffracted signal the propagation of the polarization in an absorbing medium of finite width is taken into account. We start with the wave equation for the electric field E driven by the polarization P in the slowly-varying-envelope approximation[6]

$$\partial_z E + \frac{n}{c_0}\partial_t E = \frac{i}{2}\frac{k}{\varepsilon_0 \varepsilon_b} P. \qquad (1)$$

In our experiments the sample thickness d (about 15 μm) is small compared to extension of the excitation pulses (600 μm for 2 ps) and the (A^0,X) absorption is moderate with about 30 %. Thus, for further

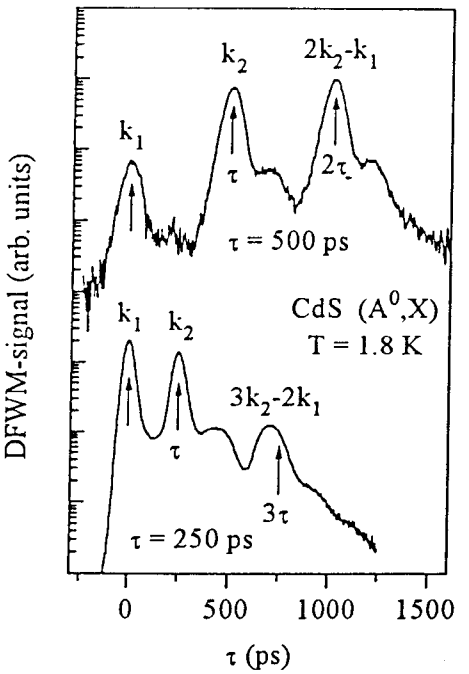

Fig.2: Time-resolved DFWM signal in first ($2k_2$-k_1) and second ($3k_2$-$2k_1$) order of diffraction for the (A^0,X) complex in CdS.

simplification of Eq. (1) the electric field can be assumed to decrease linearly within the sample and the polarization is regarded as homogeneous throughout the sample. Then we get for the electric field E_0 in the middle of the sample:

$$E_0 = E_{in} + i\frac{\hbar}{\mu} L\langle\Psi\rangle. \qquad (2)$$

E_{in} is the applied electric field, μ is the dipole matrix element, and $\langle\Psi\rangle$ is the polarization averaged over the inhomogeneous distribution. The macroscopic polarization P is given by

$$P = N\mu\langle\Psi\rangle, \qquad (3)$$

with N the density of neutral acceptors. Eq. (2) shows that the incoming electric field is corrected by an effective local field with an imaginary coupling coefficient. The coupling parameter L can be determined from the linear absorption

$$L = \frac{d}{4\pi}\int\alpha(\omega)d\omega. \qquad (4)$$

E_0 is the driving field in the density matrix equations for the (A^0,X) complex. These are solved numerically for finite pulse widths and for the macroscopic polarization the inhomogeneous broadening is taken into account. The signals diffracted in various directions are separated by a Fourier transformation with respect to the relative phase of the incoming pulses.

Fig. 3 and 4 depict the time-integrated and time-resolved DFWM signals in first and second order of diffraction calculated for independent two-level systems using Eq. (2). The phase- (T_2 = 800 ps) as well as energy-relaxation time (T_1 = 1000 ps) are taken from time-resolved experiments.[2] The inhomogeneous distribution as well as the local field parameter L (3.3 and 4.5 $\mu eV/\hbar$ of the two components of the doublet) are derived from linear absorption spectra of the sample shown in fig. 1 and 2. As obvious the calculations reproduce the basic experimental features of the second order diffracted signal, fig. 1 and 2. The time-integrated signal decays much faster than in first order and shows a correlation peak around zero delay. The time-resolved photon echo appears about 12 ps before 3τ having an FWHM of 75 ps instead of 50 ps as in first order. The

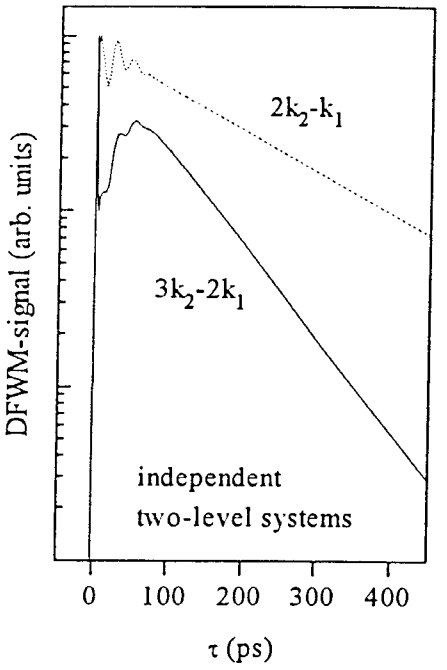

Fig. 3: Normalized calculated time-integrated DFWM signal in first ($2k_2$-k_1) and second ($3k_2$-$2k_1$) order of diffraction for independent two-level systems.

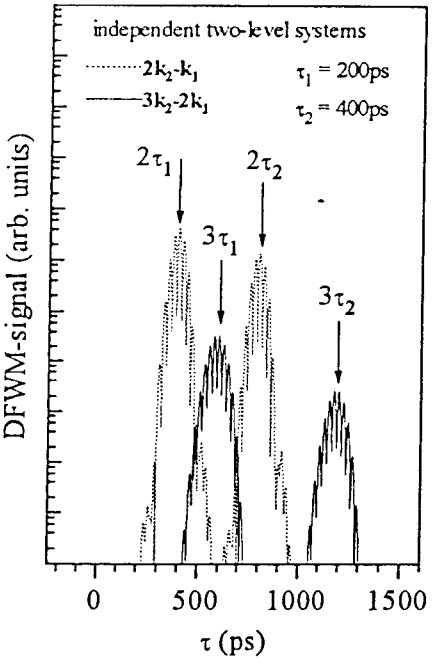

Fig. 4: Calculated time-resolved DFWM signal in first ($2k_2$-k_1) and second ($3k_2$-$2k_1$) order of diffraction for independent two-level systems.

polarization beats in the real time signal, fig. 4, are not resolved in our experiments, fig. 2, due to the experimental resolution.

5. Discussion

Fig.1 and 2 show strikingly different properties of the first and second order diffracted signals in a DFWM experiment. It is obvious, that both macroscopic signals are caused by the same coherently driven ensemble of inhomogeneously distributed two level systems ((A^0,X) complexes). The microscopic properties of the two level systems are their phase (T_2) and energy (T_1) relaxation times. The second order diffracted signal results from propagation effects as is demonstrated by the calculations. We interpret this as the diffraction of the first order diffracted signal ($2k_2$-k_1) occuring at 2 τ at the grating (k_2-k_1) generated by the two incoming pulses at τ. This is basically a nondegenerate FWM process resulting in a signal in the direction $3k_2$-$2k_1$. In case of an inhomogeneously broadened system the photon echo is now expected at 3 τ. In a similar way it is obvious that in the n^{th} diffraction order the echo occurs at $(n + 1)\ \tau$. In fact, the photon echo is found to appear a few ps earlier due to shaping by the polarization decay. The shift deduced from the model ($\approx$ 12 ps) is smaller than the experimental one ($\approx$ 45 ps). We attribute this to the fast nonexponential decay of the polarization. The time-integrated DFWM signal is measured over τ and, thus, decreases faster in higher order due to the increasing delay of the photon echo. Due to the generation process (the population grating decays with T_1) the decay of the time-integrated higher order DFWM signals depend not only on T_2 but also on T_1. The second order signal ($3k_2$-$2k_1$) should decay with $(8/T_2 + 2/T_1)^{-1}$. In fact both the experimental and the calculated second order signal decays with approximately $T_2/10$. The corresponding T_2/T_1 ratio of 1 corresponds well to the value of 0.8 deduced from the first order diffracted signal and the luminescence decay. In principle, the second order diffracted signal allows to determine T_1 without setting up a nondegenerate FWM- experiment.

6. Conclusion

In conclusion, we have demonstrated for the first time higher order photon echo signals in semiconductors. In case of the (A^0,X) complexes model calculation prove that the higher order diffracted signal is a result of propagation effects. On principle, higher order diffracted signals should be observable in each absorbing media. It is shown that in the n^{th} diffraction order the photon echo should occur at approximately $(n + 1)\ \tau$.

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