EXCITON SPIN DEPHASING AND STRUCTURAL PROPERTIES OF SEMICONDUCTOR NANOPARTICLES EMBEDDED IN GLASS MATRIX

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Abstract: This contribution reports on experimental work of four-wave mixing spectroscopy on II-VI semiconductor quantum dots (QD) embedded in glass matrix. Exciton spin dephasing effects and the degree of asymmetry of CdS_xSe_{1-x} and CdSe QD's were investigated by analysing the polarization properties of the time integrated four-wave mixing (TI-FWM) signal.

The quantum confinement of carriers in semiconductor quantum dots implies a series of novel physical properties, which were extensively investigated in the last decade. The three-dimensional confinement of excitons leads to large enhancement of the optical nonlinearities and, moreover, quantum dots are known as prototypes for investigation of physics of quantum confinement. The importance of a clear understanding of the physics of such confined systems originates in the large range of application from spinotronics to quantum computation [1,2]. Recent efforts of the scientist point at the control of spin coherence over a practical length and time scale [3,4]. We investigated two categories of samples: 1. Schott filter glasses containing $CdS_{0.6}Se_{0.4}$ QD's embedded in glass matrix and 2. Heat treated Schott filter glasses containing CdSe QD's. Characteristics of the samples like QD's diameter and size distribution were determined separately using low wavenumber Raman techniques [5]. The electron-hole interaction plays a crucial role in the characterisation of the quantum confined systems. The interaction between excitons belonging to different QD's is negligible due to the poor volume fraction of the QD's (1%), but the Coulomb interaction between excitons belonging to the same nanocrystal cannot be neglected. Thus the lowest exciton state, formed by the electron with total spin J=1/2 and the hole having J=3/2 due to the e-h interaction splits into a set of states with J=0,1, and 2. The lowering of the symmetry leads to splitting of the lowest optically active state (spin doublet with J=1) into two states, which are active in mutually orthogonal linear polarisations [6].

Time integrated four wave mixing spectroscopy is a powerful technique to investigate the polarization properties and thus the degree of asymmetry of such confined systems. It avoids the issue of inhomogeneous broadening because the integrated FWM signal reflects the average properties of the QD's within the inhomogeneous distribution [7]. The samples kept at 10 K into an optical cryostat were probed by three 80 fs laser pulses, tuned to the same wavelength corresponding to the photon energy lying in the range of the $1S_e{-}1S_h$ transition.

We used the conventional 3-pulse geometry, with the first two pulses arriving simultaneously at the sample ($t_{12} = t_2 - t_1 = 0$) and the 3[rd] pulse being delayed ($t_{31} = t_3 - t_1 \neq 0$) with respect to the other two pulses. The first two pulses create a population grating, and the third pulse is scattered on this grating according to the Bragg condition. The FWM signal is scattered in the direction given by the phase matching condition:

$$k_s = -k_1 + k_2 + k_3 , \tag{1}$$

where the k_i are the wave vectors of the pulses with the index *i=1,2,3,s* where *s* stands for the scattered FWM signal. We studied the polarization properties of the as-received sample for different polarization geometries of the incident beams. For simplicity we use for the TI-FWM signal the notation I^{ijkl}, where *i,j,k,l* stand for σ^+ and σ^- polarization of the incident beams, respectively. According to the selection rules predicted by the non-interacting system model [7], a TI-FWM signal should appear only in the case when all three pulses have the same polarization: σ^+ or σ^-. The appearance of strong FWM signals I^{++--} and I^{+-+-} (Fig. 1), forbidden according to this model, are associated to a strong exciton-exciton coupling in 3-dimensional confined systems and to a random exchange splitting of the lowest optically active J=1 exciton state, originating from a lowered QD symmetry. Therefore information about QD asymmetry can be derived by analysing the polarization properties of the TI-FWM signal, using the inverse proportionality between the splitting Δ of the J=1 exciton state and the spin dephasing time τ^{deph}.

This method has been applied to investigate the degree of asymmetry in heat treated CdSe QD's of equal average size, grown in a glass matrix under different conditions. These experiments nicely demonstrate the wide applicability of four wave mixing techniques for the characterisation of confined systems.

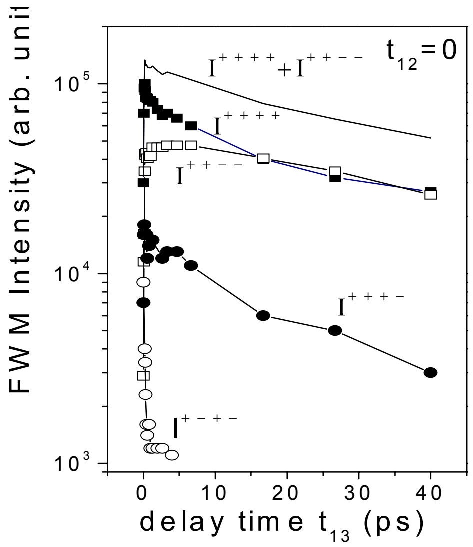

Fig. 1. TI-FWM signal of the Schott filter glass OG 550 for different polarization geometries.

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