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Exciton dephasing time in CuBr quantum dots

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Abstract

We perform degenerate four-wave mixing on CuBr quantum dot samples on a femtosecond time scale. Due to inhomogeneous broadening, we observe a so called photon-echo signal. The signal dynamics measured at different pump intensities provide information on the dephasing time. We study this dephasing time at 5 K as a function of the excitation wavelength. We also determine the homogeneous absorption linewidth by spectral hole burning measurements, in the same sample, at 5 K, using a nanosecond dye laser excitation. This is an alternative indirect method to determine dephasing times and we can compare both results. We then conclude on the difficulties of measuring dephasing times in confined systems by the latter method. © 1998 Elsevier Science B.V.

1. Samples

CuBr nanocrystallites (NC) were grown in a borosilicate glass matrix, using a diffusion controlled method [1]. The thickness of the samples was typically 0.4 mm and the semiconductor concentration in the matrix was about 1%. These low concentrations allowed us to exclude all interaction effects between two distinct NC. Fig. 1 shows the absorption spectra of samples containing NC of mean radii in the range 2.8–12.5 nm. These spectra show the $Z_{1,2}$ and Z_3 bulk like excitonic absorption bands. Moreover, one can observe the blueshift of these transitions when decreasing the NC size. This displacement allowed us to determine the mean size of the NC embedded in each sample [2]. We should note as well that due to the NC size distribution in the samples [3], excitonic absorption lines were inhomogeneously broadened.

We measured the dephasing time (T_2) in the sample containing the NC with mean radii equal to 2.8 nm, in two ways: in the time domain by transient four-wave mixing (TFWM) and in the frequency domain, using the spectral hole-burning (SHB) method.

2. Transient four-wave mixing

We performed transient four-wave mixing in a two-beam configuration. The 80 fs pulses of a self mode-locked Ti:Sa laser passed a KDP crystal. We thus disposed from a laser source whose wavelength is tunable around 400 nm. Its emission was then split into two beams of equal intensity which were modulated at frequencies Ω_1 and Ω_2 and focussed onto the sample with wavevectors k_1 and k_2. A delay line permitted us to control the time delay τ between

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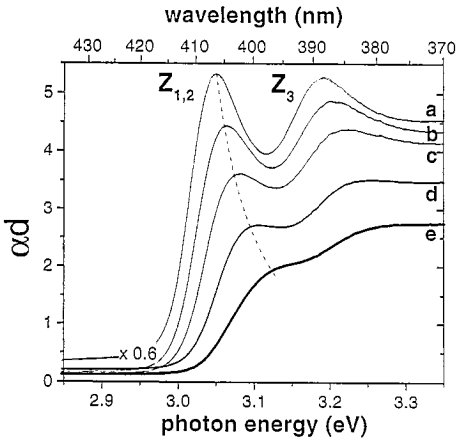

Fig. 1. Linear absorption spectra at room temperature of samples containing NCs of the following mean radii: (a) 12, (b) 9.5, (c) 5.1, (d) 3.9 and (e) 2.8 nm.

these two incident pulses. By four-wave mixing-processes, a signal in the direction $2k_2 - k_1$ was emitted. It was detected at the sum frequency $\Omega_1 + \Omega_2$ by a photomultiplier and a lock-in amplifier, as a function of τ.

Due to the inhomogeneous line broadening of our sample, we have to deal with a photon-echo-like signal [4]: it was emitted at a time 2τ after the arrival of the first pulse and decreased according to $\exp(-4/T_2)$. We had to take into account the broadening due to exciton collisions: the homogeneous linewidth Γ_{homo} was a linear function of the incident intensity with a slope β. As T_2 is related to Γ_{homo}, we found the following hyperbolic intensity dependance for the dephasing time:

$$\left.\begin{aligned} \Gamma_{\text{homo}}(I) &= \Gamma^0_{\text{homo}} + \beta \cdot I \\ T_2 &= 2\hbar/\Gamma_{\text{homo}} \end{aligned}\right\} \Rightarrow$$

$$\frac{1}{T_2(I)} = \frac{1}{T_2^0} + \frac{\beta}{2\hbar} I, \qquad (1)$$

T_2^0 and Γ^0_{homo} being the intrinsic dephasing time and the natural linewidth. They were deduced from measurements by fitting to Eq. (1) the results obtained at different excitation intensities.

Fig. 2 shows the normalized TFWM signals measured at 400 nm and 5 K, for different intensities of the incident pulses. For the strongest intensity I_0, the decay is very fast, because of a large line broadening. As the pump intensity diminishes, the signal

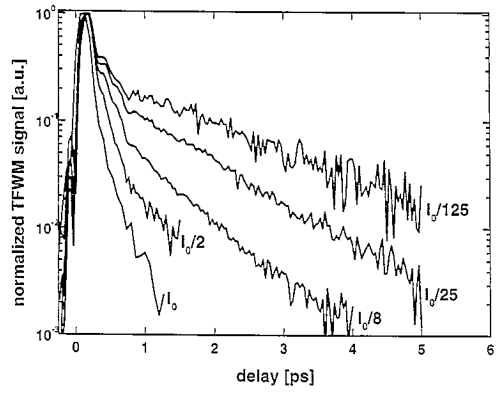

Fig. 2. Normalized TFWM measured at 400 nm and 5 K for 5 different excitation intensities.

shows longer decays and a lower signal/noise ratios. We can note also oscillations in the signal close to the temporal coincidence of the two incident pulses. Their period corresponds to an energy of about 20 meV. Oscillations can be attributed to quantum beats due to LO-phonons. The LO-phonon energy has in fact been deduced, in the same sample, from resonant Raman scattering experiments to be 20.2 meV [5].

Each experimental decay has been fitted by an exponential curve with a time constant $T_2/4$. Fig. 3 shows these dephasing times as a function of the pump intensity. The full line is the best fit (Eq. (1)) we can obtain for these experimental data, and we find the limit $T_2^0 = 6.4$ ps.

The same measurements have been carried out at different pump wavelengths. The dependence of the

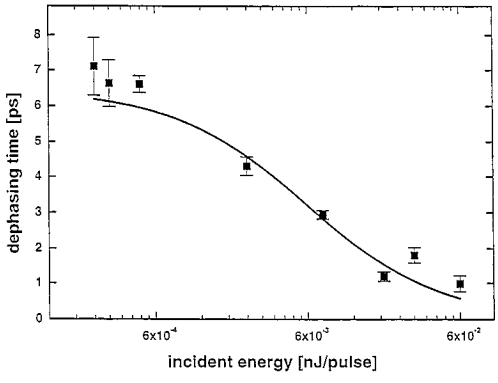

Fig. 3. Dephasing time measured by TFWM at 5 K and 400 nm as a function of excitation intensity. The full line is the hyperbolic fit given by Eq. (1).

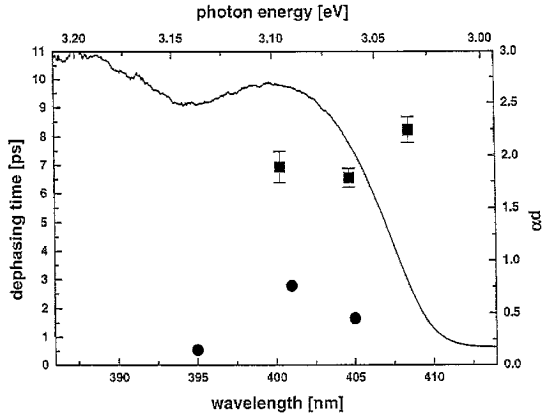

Fig. 4. Dephasing time at 5 K versus excitation wavelength measured by TFWM (■) and by SHB (●). The linear absorption spectrum at 5 K is superimposed.

dephasing time on wavelength is shown (black squares) in Fig. 4, together with the linear absorption of the sample.

3. Transient spectral hole burning

The dephasing time can be measured in the time domain with pulses of duration smaller than T_2. Its measurement in the frequency domain requires a laser whose spectral width is narrower than Γ^0_{homo}.

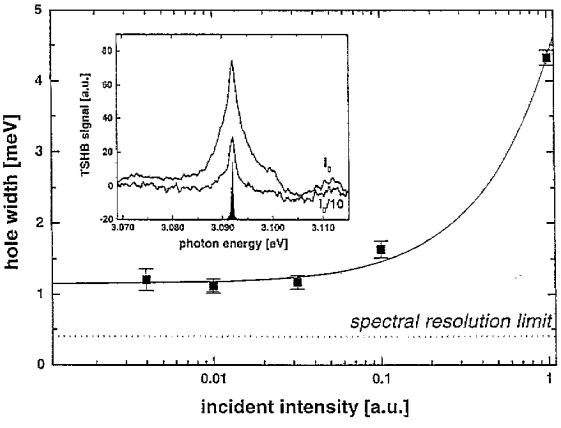

Fig. 5. Hole width measured by SHB at 401 nm and 5 K versus excitation intensity. The full line is a linear extrapolation. The insert shows differential SHB spectra obtained for two intensities (I_0 and $I_0/10$) and the laser spectrum.

For SHB we used a simple pump and probe setup. The pump is a nanosecond dye laser (PBBO dissolved in dioxane with a concentration of 0.4 g l^{-1}) pumped by a XeCl excimer laser (20 ns pulse duration). The probe was obtained from the superradiant emission of the same dye pumped by the excimer laser. Both beams were focussed at spatial and temporal coincidence onto the surface sample. We then studied the influence of the pump beam on the spectrum of the probe transmitted through the sample. Since the spectral width of the pump was narrow enough, it was able to 'burn' one homogeneous line. The probe pulses were dispersed in a single grating 3/4 m monochromator (working in its second order of diffraction) and detected by an optical multichannel analyzer connected to a computer. This system permits the detection of absorption changes $\Delta\alpha/\alpha$ of about 1%. Our spectral resolution was about 0.4 meV. Experiments have been carried out at 5 K, on the sample we studied before in TFWM experiments.

The hole which is burnt into the probe spectrum had a Lorentzien shape and its width Γ_{mes} was twice the homogeneous linewidth Γ_{homo} [6]. As discussed before, the natural linewidth is obtained by linearly extrapolating the measurements obtained for different excitation intensities to zero fluence. Fig. 5 shows the measurements performed at 400 nm, the hole width diminishes with intensity (insert of Fig. 5) and the homogeneous linewidth can be fitted with a linear decrease (full line). We found at this wavelength a value for Γ^0_{homo} of 0.490 meV, which is above the resolution limit and gives the lower limit of the exciton dephasing time $T_2 = 2.7$ ps.

The same experiments have been carried out at other wavelengths, around the $Z_{1,2}$ excitonic absorption band of the sample. The results are presented by full circles in Fig. 4.

4. Discussion and conclusion

As we have been able to measure dephasing times by two different methods, it is interesting to compare the results they give. In a general way (see Fig. 4) TFWM gives values for T_2 two or three times larger than those obtained by SHB. This discrepancy can be explained by comparing fluences and intensities of the incident beams for each experiment: SHB has

been experienced with intensities up to 100 $\mu J\ cm^{-2}$ whereas the intensity was always smaller than 3 $\mu J\ cm^{-2}$ in the TFWM experiments. The non-linear mechanisms involved in SHB assume higher excitonic populations in order to induce saturation and absorption changes. These populations have a finite lifetime of several ns during which they can scatter with other quasiparticles. This gives rise to a broadening of the line which remains even if we consider the extrapolation of the hole width to the zero fluence. In addition, the spectral hole width is close to our resolution limit for small fluences.

The TFWM experiment performed with femtosecond pulses had no spectral resolution. Therefore, we measured a mean T_2 time, averaged over NCs of different sizes. The very low intensity used in TFWM experiments permitted, however, more precise measurements of the coherence time and the homogeneous line width to be obtained than SHB, which needs high fluences.

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