Observation of a coherent signal at negative time delays in HgTe quantum dots

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Abstract: We report a decrement in the transient absorption in colloidal MIR HgTe quantum dots, resembling perturbed free induction decay (PFID), presumably the observation of a ps dephasing time at room temperature.

Quantum dots (QDs) based on semi-metals, such as HgTe, are able to span the entire MIR and parts of the FIR by tuning their size [1]. Therefore, they are extensively studied due to promising applications in lasers, photo-detectors or solar cells. We present a pump-probe study of colloidal HgTe Qds, which was initially aimed to study charge carrier multiplication [2] in these structures. Additionally to the exiton dynamics at positive pump probe delays, we observed an unexpected bleach signal at negative time delays, which reminds of perturbed free induction decay.

The colloidal HgTe particles used in this experiment were stabilized with dodecanethiol ligands and dissolved in a mixture of tetrachloroethylene and toluene. To keep the effects of sample heating, radiation damage and long-lived trap states to a minimum, a flow cell was utilized with a closed pumping cycle. The sample was probed at $\lambda_{\text{probe}} = 2\,\mu\text{m}$, in resonance with the 1s-1s exciton. For excitation we used $\lambda_{\text{pump}} = 267\,\text{nm}$ light. Before installing the flow cell, a 1 mm thick piece of ZnSe was placed at the sample position, to determine the exact temporal overlap and to estimate our temporal resolution from the rise-time of the ZnSe response, see figure 1a.

In the transient absorption data for an excitation power of 200 µW (see figures 1b and 1c) a long lived bleach signal for positive delay times is observed. This can be attributed to the photo excitation of highly excited electron hole pairs, which then rapidly relax to the band gap of the particles, thus bleaching the 1s-1s exciton transition. Around $\Delta t = 0$ a peak appears in the data, which we mainly attribute to cross-phase modulation in the CaF_2 windows of the flow cell and the solvent. More interestingly, an reduction in the transient absorption appears at negative delay times, growing in from -4 ps on. As $\Delta t = 0$ and a temporal resolution capability of better than 100 fs had been determined beforehand, this feature can not be considered an artifact.

We can also exclude radiation damage as a source of the signature, as photo-damage only becomes perceivable at excitation powers around 500 µW and above in this experimental configuration. It also manifests itself in a different manner, namely change from bleach to induced absorption. This can be seen in the transient data of the HgTe QD sample for an excitation power of 700 µW , see figures 1b and 1d. Here, the signature at negative delay times is clearly absent. We therefore presume the feature is perturbed free induction decay (PFID), which has already been observed for single epitaxially grown GaAs quantum dots at 10 K [3].

Often being an undesired artifact in pump-probe spectroscopy [4], PFID is a well known effect. It arises at negative delay times, when the system's dephasing time is longer than the cross-correlation of pump and probe pulse. In the data presented here case, however, it would represent an extraordinarily long excitonic dephasing time at room temperature.

To extract the hypothetical dephasing time τ, the following function was fit to the data

$$\Delta A(t<0) = y_0 + A * \Theta(t-t_0) \cdot \left[1 - \exp\left(-\frac{(t-t_0)}{\tau}\right)\right]. \tag{1}$$

Here, $\Theta(t)$ denotes the Heaviside step function. The fit was performed on the transient data ranging from -50 ps to -700 fs, as to exclude contributions from the cross-phase modulation signal at $\Delta t = 0$. We retrieve $\tau = 2.10 \pm 0.41$ ps for a 100 µW excitation.

At room temperature dephasing times in QDs typically are a lot shorter than 2 ps, e.g. around 200 fs in InAs systems [5,6]. Whether a factor of ten difference in dephasing time could be explained by fundamental differences in the systems, like crystal structure or size of the particles, needs further investigation.

Given our presumption is correct, this would be the, to our knowledge, first observation of ps-scale dephasing times in quantum dot systems at room temperature. This would make HgTe quantum dots the perfect model system to study fine-structure dynamics on fs timescales by exploiting 2D spectroscopy. Also studying the underlying principles of the long dephasing time itself would give valuable insight for the engineering of QDs.

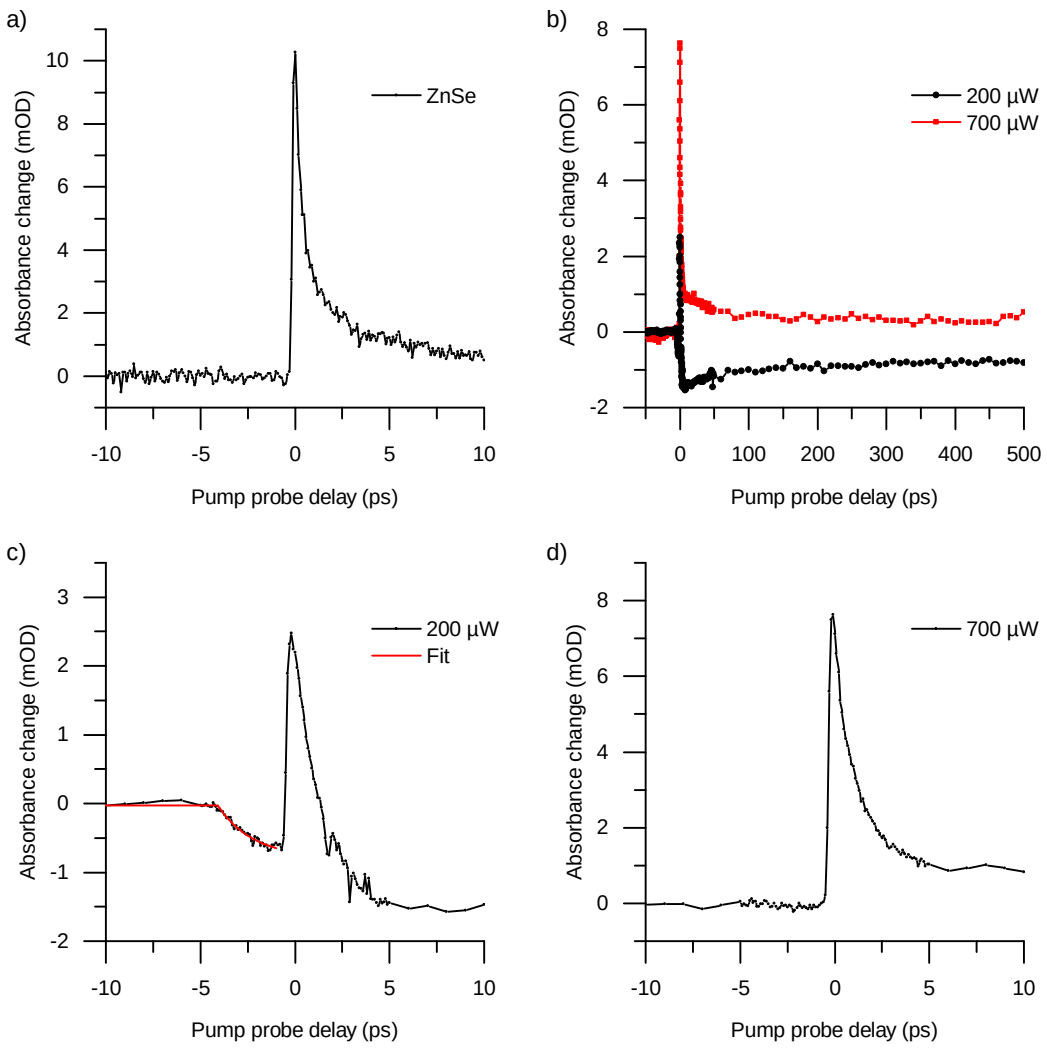

Fig. 1. a) ZnSe reference scan. b) Timetrace for HgTe QDs for excitation powers of 200 µW and 700 µW. c) Zoomed in view for HgTe QDs at 200 µW excitation, fit according to equation 1. d) Zoomed in view for HgTe QDs at 700 µW excitation. In the transient data for 700 µW the sample has undergone radiation damage.

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