

Tracking electron and hole relaxation dynamics in CdTe nanorods by 2D electronic spectroscopy

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We present new insights into the exciton dynamics from two-dimensional electronic spectroscopy on CdTe nanorods. Tracing of state-resolved energy relaxation dynamics allowed us to resolve the dynamics of both electron and hole transitions.

Quantum-confined semiconducting nanocrystals offer unique opportunities for controlling electromagnetic energy at the nanoscale. Highly complex hybrid nanostructures have been suggested for photovoltaic and photocatalytic applications [1]. For device optimization, understanding of the carrier cooling dynamics is of particular interest, the hole dynamics being of major importance. However, these dynamics are difficult to extract by conventional pump-probe spectroscopy: energetic congestion of the hole states leads to an overlap of their transient signals with broadband excitation, while state-selective narrowband excitation does not provide sufficient temporal resolution to resolve the fast dynamics. In this work we apply two-dimensional electronic spectroscopy (2DES) with 10-fs time resolution to study the carrier dynamics of small cadmium telluride (CdTe) nanorods [2] in order to examine the electron and hole dynamics of the first three excitons (S1, S2, S3). From the analysis of diagonal and cross peaks we extract 30 (45) fs hole relaxation times for the S2 (S3) exciton.

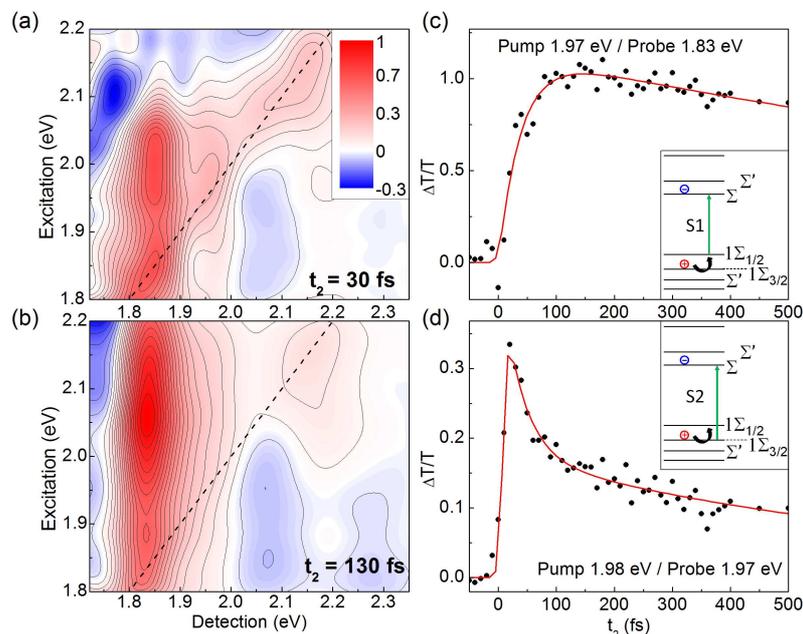


Fig. 1. 2DES maps of CdTe nanorods at waiting time delays $t_2=30$ fs (a) and $t_2=130$ fs (b). Here, red represents positive $\Delta T/T$ due to transient bleaching and blue signifies negative photoinduced absorption signals induced by transition energy shifts due to biexciton interaction. Absolute color and contour scale is set to the minimum and maximum intensity, the excitation pulse energy was 1.6 nJ. The cross peak S2/S1 and the diagonal peak S2/S2 are plotted in (c) and (d), respectively (black dots). The fit of the dynamics reveals a rise of (a) and a fast decay of (b), both of a time constant of 30 ± 10 fs, corresponding to hole relaxation, schematically shown in the insets.

[1] U. Banin, Y. Ben-Shahar, K. Vinokurov, Chem. Mater. **26**, 97-110 (2014).

[2] I. Kriegel, F. Scotognella, G. Soavi, R. Brescia, J. Rodríguez-Fernández, J. Feldmann, G. Lanzani and F. Tassone, Chem. Phys., doi:10.1016/j.chemphys.2015.08.002 (2015).