

Ultrafast dynamics of excitons and charge carriers in 2D colloidal semiconductor nanocrystals

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The development of ultrathin two-dimensional (2D) semiconductor nanostructures have generated considerable interest over the past years. Among these nanostructures, transition-metal dichalcogenide nanosheets, 2D hybrid perovskites and colloidal semiconductor nanoplatelets have a thickness controlled with atomic precision (defined number of monolayers) which leads to strongly tunable and exceptionally narrow optical features.^[1] For II-VI semiconductor nanoplatelets, the recent development of heterostructures of various shapes and compositions have allowed to access new optical properties and thus opened new fields of applications.^[1]

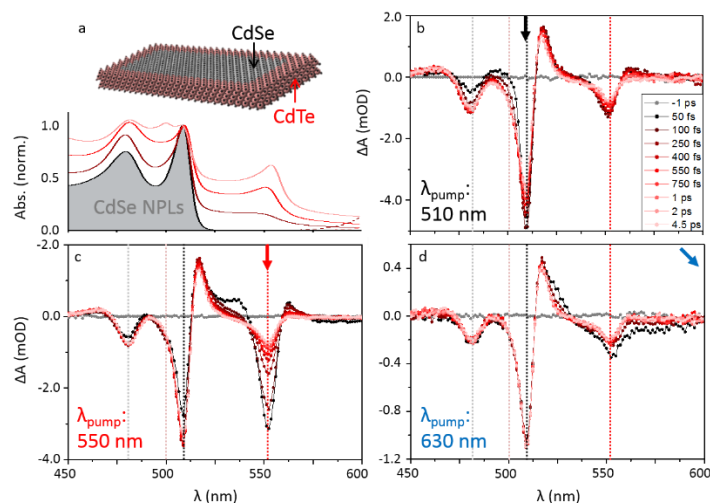


Figure 1. a) Schematics of CdSe-CdTe core-crown nanoplatelets and absorption spectra of three different samples (bare CdSe nanoplatelet spectrum displayed in grey). b-d) Transient spectra at various pump-probe times for three different excitation wavelengths. The position of the heavy hole and light hole exciton transition is highlighted with the black (red) and grey (pink) dotted lines for CdSe (CdTe), respectively.

Here we use femtosecond transient absorption spectroscopy to study the ultrafast dynamics of excitons and charge carriers in CdSe-CdTe core-crown nanoplatelets (Fig. 1a).^[2] In this system, the well red-shifted photoluminescence emission was previously attributed to recombination at the charge transfer state, with the electron preferentially localized in CdSe and the hole in CdTe (type-II emission).^[3] Instead, our measurements reveal an instantaneous bleach of the CdTe optical transitions when pumping selectively at the CdSe transitions, and reciprocally (Fig. 1b,c). These results are explained by the direct delocalization of the electron wavefunction over the heterostructure following the optical excitation. By comparing the early time dynamics of the different bleach transitions for different pump wavelengths and on several samples (different lateral dimensions and crown compositions), we conclude the photoluminescence involves hole trap states, either localized at the CdSe-CdTe interface or at the surface of the CdTe crown.^[4]

We will also present the current progress in the development of a new two-dimensional electronic spectroscopy setup operating in the visible and ultra-violet range to investigate the sub-100 fs dynamics of colloidal nanocrystals after optical excitation well above the bandgap of the semiconductor nanostructures.^[5]

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