Exciton dynamics in Cu-doped InAs colloidal quantum dots

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Abstract: Femtosecond transient absorption spectroscopy has been used to investigate the exciton dynamics in native and Cu-doped InAs quantum dots from three respects: 1) Auger recombination; 2) hot exction cooling; 3) absorption cross section.
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1. Introduction

Doping is an attractive way to control the physical and chemical properties of colloidal quantum dots (QDs). For example, doping can change the Fermi level by increasing electron or hole concentration and achieve the desired absorbing wavelength, which will broaden their technological application in microelectronics and optoelectronics [1]. So it is very important to understand the role of dopants in the QDs and how they affect the properties of QDs such as multi-exction dynamics. Our recent work focus on the comparing ultrafast exciton dynamics in InAs quantum dots with those in otherwise identical Cu-doped crystals by using femtosecond transient absorption spectroscopy. In this case, copper partly donates its valence electrons to the QDs, leading to n-type doping [1].

2. Results and Discussion

Figure 1 shows the transient difference spectra after excitation by 640 nm in different doped QDs, heavily doped (right), lightly doped (middle) and undoped (left).When irradiated at 640 nm, the bleaching at the band edge, which is attributed to the 1S-1S transition, is observed. Meanwhile at early times a positive-broad absorption is also observed which evolves during the process of exciton cooling.

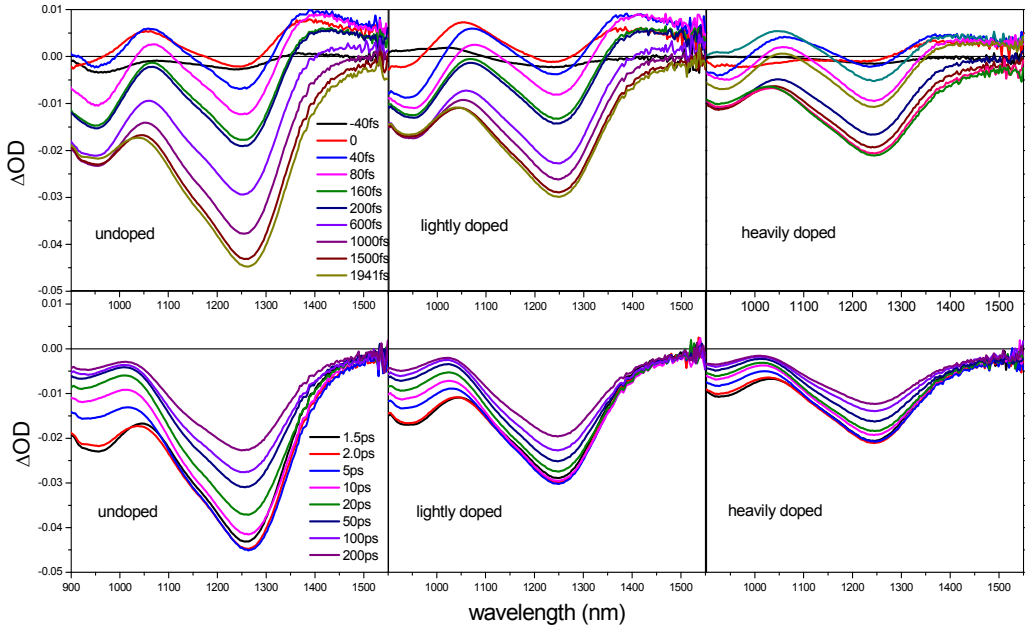

Fig.1 Transient spectra at different time delays after excitation at 640 nm in different doped InAs QDs

After initial rapid evolution, a gradual decay is observed at longer delays (t > 2ps), which is expected to be the exction annihilation. At low fluence, the bleach exhibits a slow decay which is assigned to the single exciton decay. As the fluence is increased, rapid bleach decay due to Auger recombination of the multi-excitons is observed along with an incessant slow decay of the single exciton. In different doping level QDs, the slow decay which is attributed to the single exciton cooling is similar, and the fast decay which is attributed to the Auger recombination of multiple excitons does not show dramatically different. The decays are fit to a biexponential function resulting in a slow time constant of 259 ps and fast time constants of about 19 ps for three samples irrespective of level of doping.

The early positive absorptions are assigned to the absorption by the hot excitons and the decay following it reflects the nascent exciton cooling process. In previous experiments the positive absorption was assumed to shift of the transient energy level due to the biexciton shifting effect[2] and the apparent red shift of the band edge is due to large amplitude of the bleach. However, we observe that the amplitude of the bleach at the same delays decrease dramatically as the doping level increases, while the positive absorptions are relatively unchanged. This indicates that the positive absorption is not related with the change of the energy level, and is attributed to the absorption of the hot exciton.

To further probe doping effects on the photo-physics, a comparison of QD absorption cross sections and the bleach induced by a single relaxed exciton was conducted for doped and undoped samples. To do that, two measurements were performed. In the first, the distribution of exciton number states was measured as a function of pump fluence well above the band edge. This is obtained from comparing bleach immediately after photoexcitation (A) with that surviving Auger recombination (B) after photoexcitation at 640 nm. Assuming Poisson statistics for multiple absorptions and the known fluence, this ratio can be simulated to produce σ_{640nm}, from which the band edge cross section is deduced from the linear spectrum. As shown in table 1, this cross section is essentially unchanged by doping. The bleach induced by a single relaxed exciton at the band edge can be determined by measuring the absolute value of band edge bleach ~2 ps after excitation, as a function of the density of absorbed photons. Unlike the cross section for absorption, the bleach cross section for a single exciton is reduced dramatically by the doping with Cu.

Tablet.1 The cross section at band edge in the samples by two methods

Pump at 640nm	Undoped(10^{-15}cm^2)	Lightly doped(10^{-15} cm^2)	Heavily doped (10^{-15} cm^2)
A/B	0.42±0.07	0.44±0.01	0.40±0.03
BLEACH	0.31±0.03	0.25±0.01	0.109±0.005

3. Conclusions

We have investigated ultrafast exciton dynamics in native and Cu-doped InAs quantum dots by femtosecond transient absorption spectroscopy. And our experimental results show that:

1) Exction dynamics like Auger recombination of multi excitons, single exciton decay and hot exciton cooling process are similar in InAs QDs irrespective of level of doping. As n-type doping QDs, the extra electrons were expected to have a dramatic effect on the dynamics of photon generated charges in QDs. However the similar relaxation time constants of multi excitons and single exciton demonstrate that the electrons offered by Cu dopants are not involved in carrier dynamics in our experiments.

2) The cross section derived from the band edge bleach is always smaller than that from the A/B ratio method, especially in heavily doped QDs. These differences indicate that initial an apparent change in the band edge degeneracy, drastically reducing the bleach contributed by a single relaxed exciton at the band edge.

4. References

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