BREAKING OF PHONON BOTTLENECK IN $CsPbI_3$ NANOCRYSTALS DUE TO EFFICIENT AUGER RECOMBINATION

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ABSTRACT

Inorganics lead halide perovskite (LHP) have been became appropriate system for demonstrating light-matter interaction due to their flexible bandgap tunability, defect tolerance and high photoluminescence quantum yield nature. Although, LHPs have many hallmark properties which can support highly efficient photovoltaic devices, but they lost lot of energy in carriers-phonon scattering which slow down the recombination process and decrease the efficiency. Faster thermalization time of hot carriers support electron-hole recombination at band-edge which can be exploited in optoelectronic devices either by incorporating electrons/holes transport layer for photovoltaic or fast recombination for LED. Recently, efficient photovoltaic and light emitting devices are immediate requirement for high-speed quantum technologies. Here, we have chosen $CsPbI_3$ and Cu-doped $CsPbI_3$ nanocrystals (NCs) and addressed both issues simultaneously by using transient absorption spectroscopy. Our sample can be classified as an intermediate confinement as the size of NCs is 16 nm (32 nm) for $CsPbI_3$ (Cu-doped $CsPbI_3$) NCs which are higher than Bohr's radius (~12 nm), and give sharp excitonic peaks in ground state optical absorption with excitonic position at ~2.1 eV. Further, by femtosecond laser excitation with 400 nm and 120 fs pulse width, which is generated by second harmonic of fundamental wavelength 800 nm. The fluence-dependent measurement revealed the many-body interaction and hot carriers dynamics. At higher fluence, say 150 $\mu J/cm^2$ and above, pristine $CsPbI_3$ NCs shows breaking of phonon bottleneck effect by fast decay while Cu-doped NCs showed slow thermalization. To get insight, we have calculated Auger recombination (non-radiative) lifetime by subtractive method. The lifetime measurements clearly distinguished the appearance of contrast results due to efficient Auger process associated with pristine $CsPbI_3$ NCs. Thus, our results provide insight to incorporate metal doping and understanding about hot carrier dynamics for solar energy harvesting.

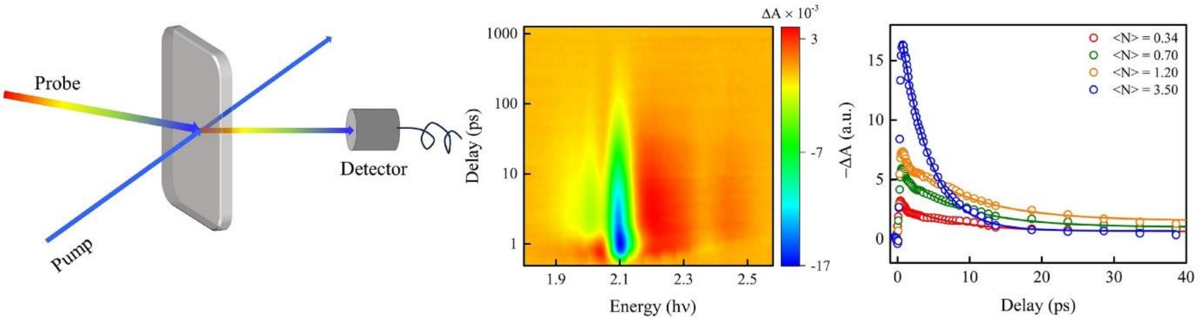

Figure 1: *From right to left*; pump-probe technique, 2D contour plot and calculated Auger recombination time.

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