Supporting Information

Power-Dependent and Ultrafast Spectroscopic Studies of Ag Ions Doped Colloidal CdSe Nanoplatelets

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S1. Experimental Section

Preparation of cadmium myristate:

We synthesized cadmium myristate [Cd $(Myr)_2$] using established methods with slight modifications.[1] 0.43 g Sodium hydroxide (NaOH) and 3.42 g myristic acid were dissolved in anhydrous methanol (300 mL) to form a sodium myristate solution. In a separate container, 1.106 g cadmium nitrate tetrahydrate was dissolved in 40 mL methanol. The cadmium nitrate solution was added dropwise to the sodium myristate solution with vigorous stirring, forming a white precipitate. After complete addition, the reaction was stirred for an additional hour, and the precipitate was washed with dried methanol and then vacuum-dried at 60°C overnight.

Synthesis of 4 ML CdSe nanoplatelets:

By adapting established procedures, we synthesized 4 monolayer (ML) CdSe Nanoplatelets (NPLs).[2] In a two-neck flask, 170 mg of $Cd(Myr)_2$, 12 mg of Se powder, and 15 mL of ODE were degassed under vacuum at room temperature for 1 hr. The mixture was heated under argon flow to 240°C. At 195°C, 40 mg of cadmium acetate dihydrate ($[Cd(OAc)_2]$. $2H_2O$) was quickly added, and the solution was kept at 240°C for 10 min. Oleic acid (1 mL) was introduced at 60°C during cooling. The mixture was dissolved in hexane, and size-selective precipitation was used to exclude nanoplatelets. The NPLs were dispersed in toluene for further use.

Synthesis of 4 ML Ag: CdSe NPLs: We have modified the previously reported procedures by synthesizing 4 ML thick CdSe NPLs.[3] The precipitated NPLs were dispersed in hexane and used for cation exchange reaction for Ag doping. Silver acetate has been taken at the required target ratio to Cd by calculating the concentration of cations. Then, Ag(OAc) was added to the CdSe solution and continuously stirred for 1 hour in ice-cold conditions. Then, the unreacted precursors were excluded from the mixture by size-selective precipitation. The NPLs were finally dispersed in toluene for further characterization and use.

S2. CHARACTERIZATION:

Step-scanned X-ray powder diffraction (XRPD) pattern was recorded using a Bruker D8 ADVANCE diffractometer with Cu-k_α X-ray radiation (1.5418 Å). The sample was prepared by drop-casting the solution onto a glass substrate. The sample was completely dried before mounting it on the goniometer. Transmission electron microscopy (TEM) measurements were done using a JEOL, JEM2100F, at an operating voltage of 200 kV. TEM samples were prepared by drop casting of nanocrystal solution in toluene on a carbon-coated Cu grid followed by the evaporation of the solvent. The X-ray photoelectron spectroscopy (XPS) measurements were carried out using an Omicron Nanotechnology instrument. Room-temperature optical absorption spectra were taken by UV−Vis spectrophotometer S4 (Shimadzu). The emission spectra of all the samples were taken with a FluoroMax-P (HORIBA Jobin Yvon) luminescence spectrophotometer. To conduct power-dependent measurements, we used a diode laser emitting light at a wavelength of 403 nm to excite the sample placed within an optical cryostat. This cryostat allows for precise control of the sample temperature. The photoluminescence spectra are recorded using a spectrograph coupled with an electron-multiplying CCD detector. We used the Andor Technology thermoelectrically cooled electron-multiplying CCD (iXon Ultra 897, with 512x512 pixel sensor), which is very sensitive in low light levels.

We have used the same fs-transient absorption spectroscopy (TAS) setup described elsewhere.[4] In brief, a mode-locked Ti: sapphire oscillator (Seed laser, Mai-Tai HP, Spectra-Physics) generates pulses of <100fs duration at 800 nm, and a repetition rate of 80 MHz. A separate pump laser (Nd: YLF laser, 527 nm, ASCEND EX, Spectra-Physics) is required for amplifying the seed pulse. The output from the amplifier was (800 nm, ~100fs, 1 KHz, pulse energy 5 mJ) sent to the spectrophotometer. BBO crystal was used to generate a 400 nm pump beam for exciting the samples. The other part of the 800 nm light was focused on a CaF_2 plate to generate a white-light continuum (WLC) and used as a probe pulse. The WLC is also divided into two parts; one part is used as a probe pulse and the other one as a reference. The reference and

transmitted probe beam are then sent to different diode arrays where the reference beam helps to account for the intensity fluctuation in the white light continuum. The power of the pump pulse (<4μJ/cm^2) was kept low enough to avoid the multi-exciton effect and sample degradation. It has to be mentioned that below the 150 fs component can be treated as an instrument response function (IRF). All recorded data were fitted using Surface Xplorer version 4.0 software and subsequently, the chirp was corrected using the same software to remove the group velocity dispersion (GVD).

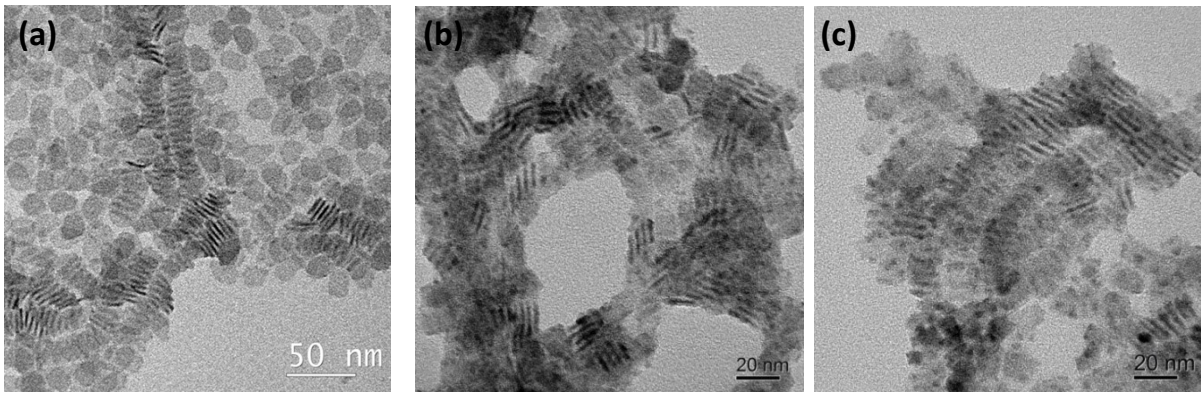
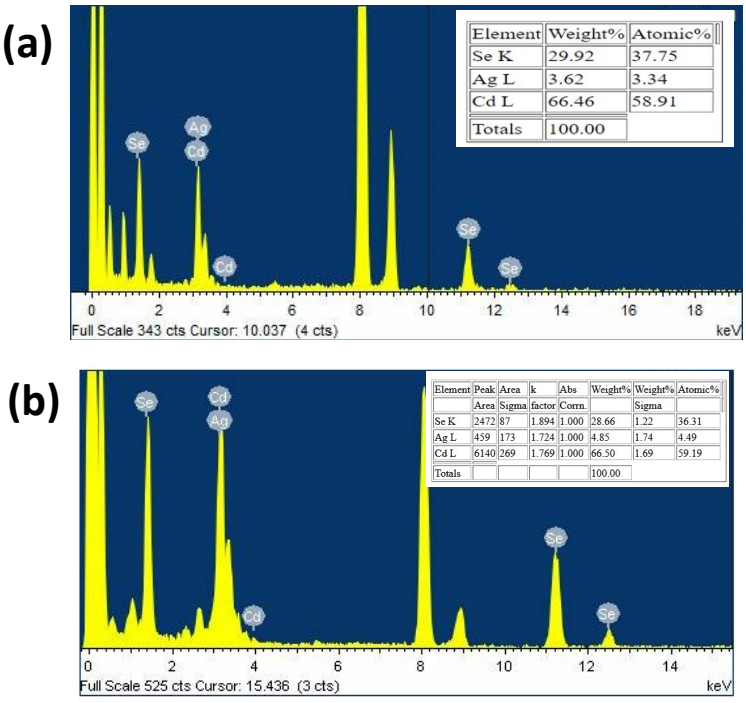


Figure. S1. (a, b, and c)TEM images of 4 ML undoped, 7%, and 11% Ag doped CdSe NPLs, respectively.



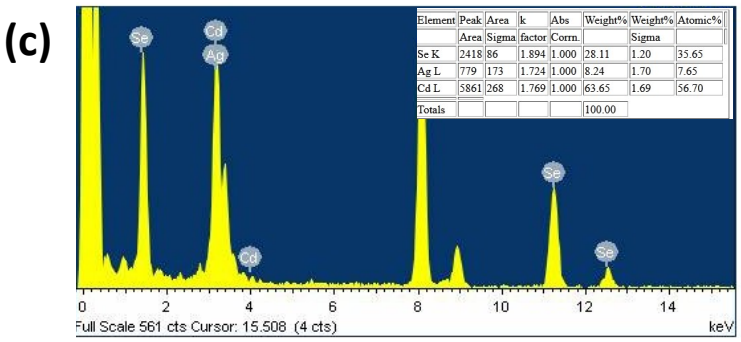


Figure S2. (a, b, and c) EDS spectra of 4 ML 5%, 7%, and 11% Ag: CdSe NPLs.

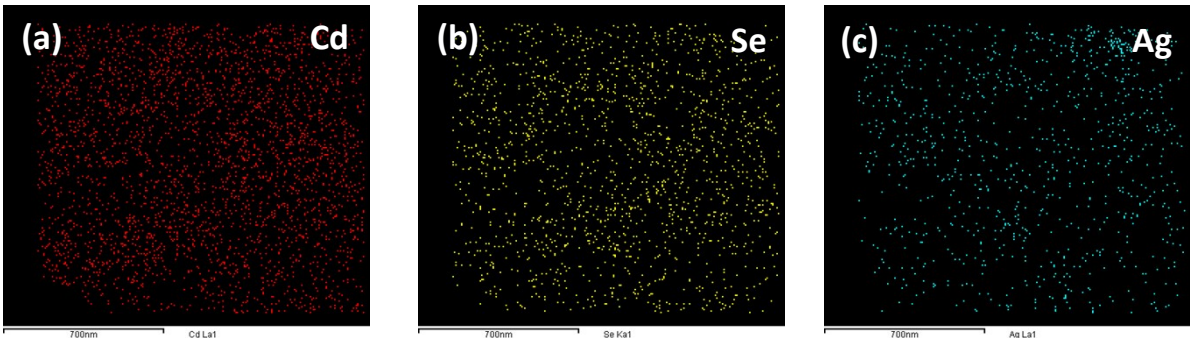


Figure S3. (a, b, c) EDS mapping of Cd, Se, and Ag respectively in Ag: CdSe NPLs.

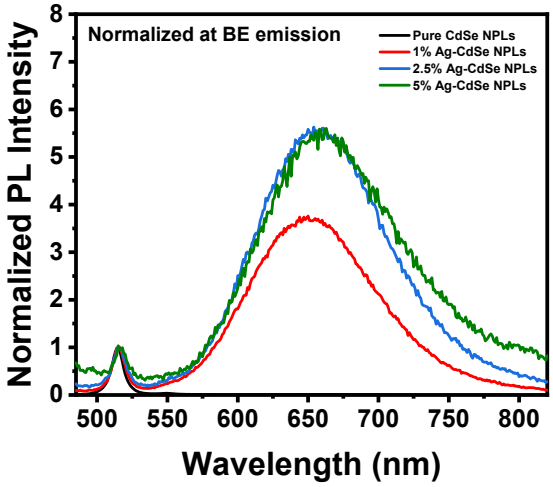


Figure S4. Photoluminescence spectra of 4 ML CdSe and Ag-doped CdSe NPLs (undoped CdSe-black; 5%-red; 7%-blue; and 11%-olive) normalized at BE emission position.

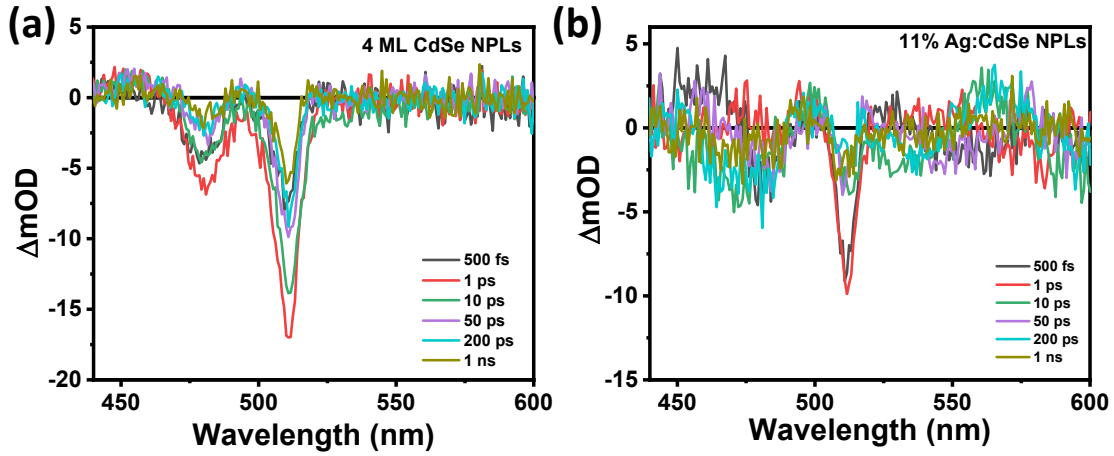


Figure S5. TA spectra of (a) undoped and (b) 11% Ag-doped CdSe NPLs.

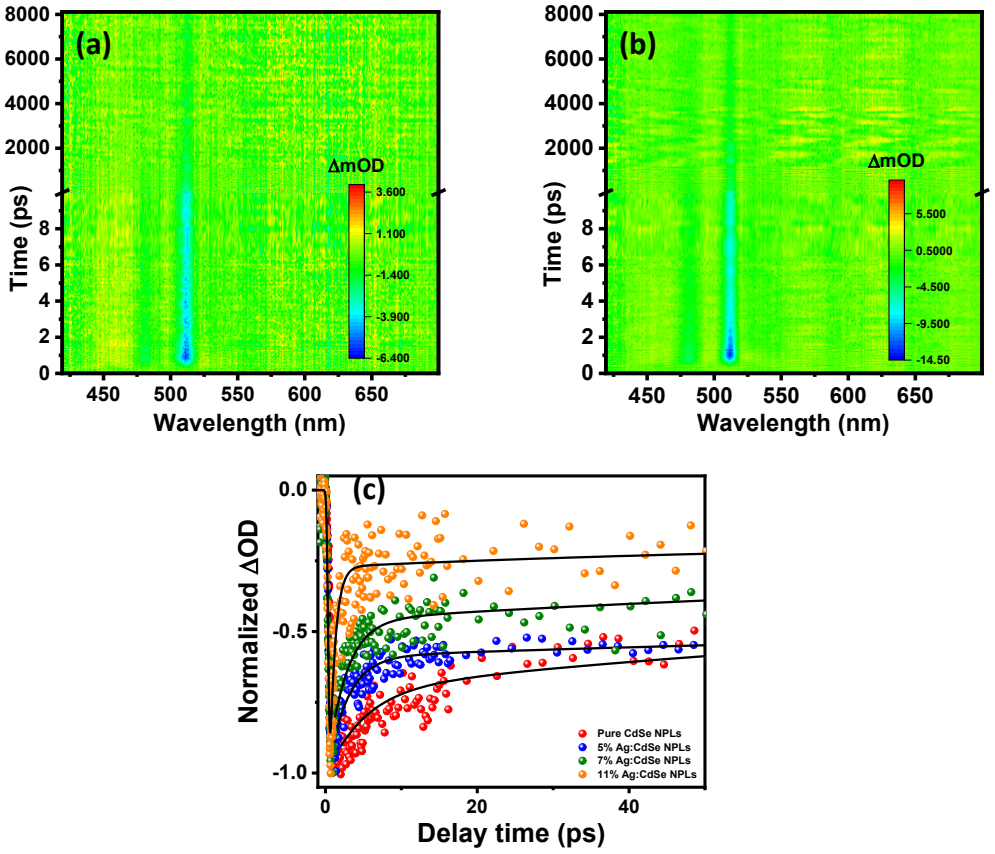


Figure S6. (a, b) 2D false color map of 4 ML 5% and 7% Ag doped CdSe NPLs, respectively. (c) Normalized bleach recovery kinetics of undoped and doped CdSe NPLs

Table S1. Time-resolved PL decay parameters for 4 ML undoped CdSe NPLs and Ag-doped CdSe NPLs.

Sample (NPLs)	τ^g (ps)	τ_1^r (ps)	τ_2^r (ps)	τ_3^r (ps)
CdSe	0.41	5±0.25 (29.8%)	140±5 (41.5%)	1 ns (28.7%)
5% Ag: CdSe	0.3	2.6±0.1 (53%)	138±7(19.3%)	>1ns (27.7%)
7% Ag:CdSe	0.2	2.5±0.12(55.5%)	113.6±5.6(17.6%)	1 ns (26.9%)
011% Ag:CdSe	0.16	0.8±0.04 (87%)	72.1±3.6 (8%)	1 ns (5%)

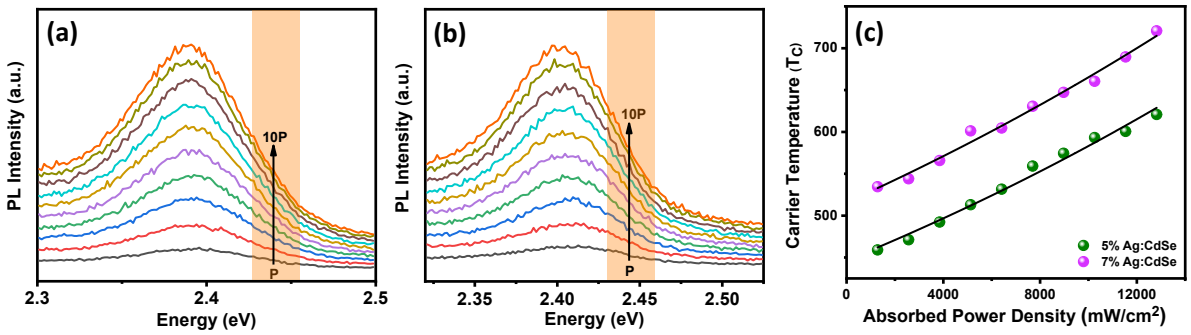


Figure S7. (a, b) Power-dependent PL of 5% and 7%Ag: CdSe NPLs respectively. (c) Carrier temperature increases with increasing power density (olive- 5% Ag: CdSe and purple- 7% Ag: CdSe NPLs)

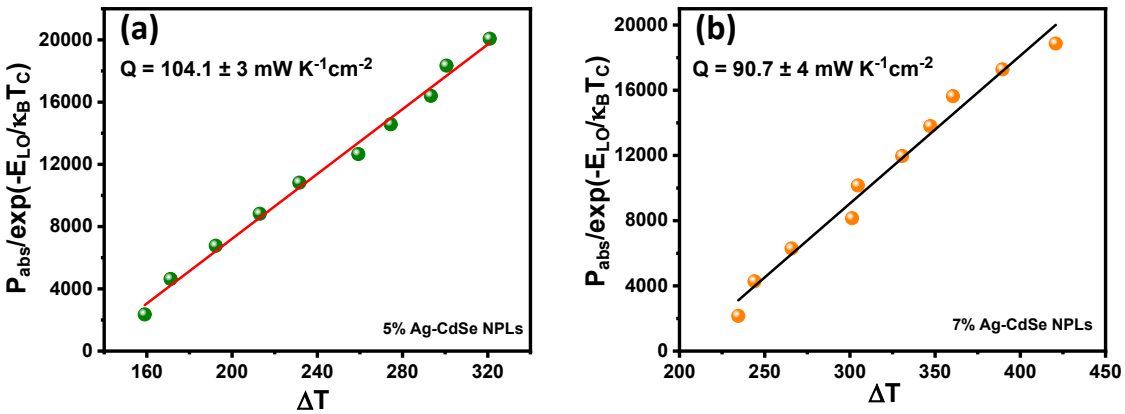


Figure S8. (a, b) $P_{abs}/exp(\frac{E_{LO}}{\kappa_B T_C})$ plot as a function of ΔT, where slopes give the thermalization coefficient values (Q) for 5% and 7% Ag-CdSe NPLs respectively.

References

(1) Tessier, M. D.; Spinicelli, P.; Dupont, D.; Patriarche, G.; Ithurria, S.; Dubertret, B. Efficient Exciton Concentrators Built from Colloidal Core/Crown CdSe/CdS Semiconductor Nanoplatelets. *Nano Lett.* **2014**, *14*, 207-213.

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(3) Khan, A. H.; Pinchetti, V.; Tanghe, I.; Dang, Z.; Martín-García, B.; Hens, Z.; Van Thourhout, D.; Geiregat, P.; Brovelli, S.; Moreels, I. Tunable and Efficient Red to Near-Infrared Photoluminescence by Synergistic Exploitation of Core and Surface Silver Doping of CdSe Nanoplatelets. *Chem. Mater.* **2019**, *31*, 1450-1459.

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