

Electronic Supplementary Information

Improved Synthesis of $\text{PbS}_x\text{Se}_{1-x}$ Ternary Alloy Nanocrystals and Their Nonlinear Optical Properties

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Materials and Supplies: All chemicals are used as received. Selenium(Se, AR) is purchased from Shanghai Zhongqin Chemical Reagent. Lead chloride (PbCl_2 , AR), sulfur (S, 99.5%) are purchased from Tianjin Guangfu Fine Chemical Research Institute. Oleylamine (OLA, 96%) is obtained from J&K Chemical. Anhydrous ethanol (AR), anhydrous chloroform (AR) are obtained from Tianjin Kaitong Chemical and anhydrous tetrachloroethylene (TCE, AR) is purchased from Xilong Chemical.

Synthesis of $\text{PbS}_x\text{Se}_{1-x}$ NCs: All of the alloyed nanoparticles are synthesized using a one-pot, hot injection method in sealed two-neck flasks. In a typical reaction, PbCl_2 (1 mmol, 280 mg) were added to 5 mL of oleylamine at room temperature in sealed reaction system, and the mixture was heated to 110°C under vacuum for 1 h to distill off volatiles. During the reaction, the PbCl_2 -OLA mixture turned into a homogeneous and clear solution. Argon gas was subsequently admitted, and the mixture was left for 30 min under vigorous stirring. Elemental selenium (0.5 mmol, 40 mg) and sulfur (0.5 mmol, 16 mg) was dissolved in 3 mL of OLA by heating at 240°C in oil bath to give a orange-yellow Se/S solution, which was thereafter injected into the PbCl_2 -OLA mixture at the same temperature. The heat was removed after the reaction proceeded for 5 min and the reaction solution was cooled naturally to room temperature, resulting in a black colloidal solution. And then 15 mL of chloroform was added to the reaction mixture, which was thereafter centrifuged for 5 min at 3000 rpm to eliminate any solid byproducts, especially unreacted lead chloride. A minimum amount of ethanol was added to the supernatant and the solutions were

subsequently centrifuged to induce precipitation of the NCs. The resulted supernatant was discarded, and the precipitate was redispersed in chloroform. The as-prepared NCs were then further precipitated with ethanol and redispersed in chloroform to remove any further trace of ligand. The eventual NCs dispersions were dried under Argon stream and redispersed in chloroform to remove any trace of ethanol from the solution.

Z-scan measurement: The Z-scan technique is employed to measure the nonlinear optical properties of PbSe NCs. Briefly, 4 ns (FWHM), 532 nm laser pulses with a repetition rate of 10 Hz from a frequency-double Q-switched Nd:YAG laser (Continuum, Model Surelite SL-I-10) was used as the light source. The spatial and temporal profiles of the laser pulses presented an approximately Gaussian distribution, with energies that could be varied from 10 to 100 $\mu\text{J}/\text{pulse}$. The laser beam was split into two parts by a beam splitter and two corresponding pyroelectric detectors (Laser Probe, RJ-735; with RJ7620 dual channel power meter) were used to measure changes in laser transmission. Under the open-aperture configurations, the aperture placed before the detector was kept open and the nonlinear absorption of the samples was measured. The other part of laser beam was simultaneously measured by another detector with a partially closed aperture set in front of it, which is the close-aperture configurations. The intensity change detected herein contained both nonlinear absorption and nonlinear refraction. The samples were placed in quartz cells with thickness of 2 mm, mounted on a translation stage that was controlled by a computer to move along the z axis with respect to the focal point.