## **Supporting Information**

# Effect of silver doping on ultrafast broadband nonlinear optical responses in polycrystalline Ag-doped InSe nano films at near-infrared

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#### Materials

The sputtering targets of InSe (99.99% purity, 6.0 cm diameter, 0.3 cm thickness and 0.2 cm thickness with copper back target) and Ag (99.999% purity, 6.0 cm diameter, 0.5 cm thickness) were all purchased from Beijing zhongcheng New Material Technology Co., Ltd.



Figure S1 The sputtering targets of InSe and Ag in the direct current-radio frequency sputtering system.

### Sample preparation

Ag-doped InSe and pure InSe nano films were precisely manufactured using a direct current-radio frequency sputtering technique. The sapphire substrates were ultrasonic cleaned in anhydrous ethanol and deionized water respectively for 20 minutes until completely cleaned and then dried. The schematic diagram of experimental procedure as presented in Figure S2. Distance between the sputtering targets to the sapphire substrates was immobilized in 8.2 cm to achieve the most excellent thickness uniformity. The direct current power and radio frequency power of the deposition process were 5 W and 50 W respectively. Under this power conditions, the reactant pressure of 3.0 Pa was available for controlling the argon flow rate to 20

sccm. The substrate temperature was raised from room temperature to 150 °C, and then deposited for 2 h to fabricate the Ag-doped InSe and pure InSe nano films.



**Figure S2** Procedure for manufacturing Ag-doped InSe nano film with 320 nm thicknesses on sapphire substrates via direct current-radio frequency sputtering technique.

#### Instruments

Morphology, thickness and grain size of Ag-doped InSe nano film were observed by scanning electron microscope (SEM, Quanta 200FEG). High-resolution transmission electron microscopy (HRTEM) and selected area electron diffraction (SAED) were detected by JEM-2100 to characterize the microstructure of Ag-doped InSe nano film. Elemental analysis of Ag-doped InSe nano film was detected by energy dispersive spectroscopy (EDS, Quanta 200FEG). The linear absorption spectrum of Ag-doped InSe nano films were detected by spectrophotometer (UV-Vis-NIR, PerkinElmer).

#### Nonlinear optical studies

The nonlinear absorption and refraction properties of Ag-doped InSe and pure InSe nano films were measured by femtosecond Z-scan measurement. The excitation wavelength at 800 nm and 1100 nm emitted from an optical parametric amplifier pumped by a Yb:KGW femtosecond laser with pulse width of 190 femtosecond. The optical parametric amplifier and beam path diagram of experimental procedure as presented in Figure S3 and S4.



Figure S3 The optical parametric amplifier in femtosecond Z-scan measurement.



Figure S4 The beam path diagram of femtosecond Z-scan measurement.

The nonlinear transient absorption and carrier recombination properties of Agdoped InSe and pure InSe nano films were measured by femtosecond transient absorption spectroscopy technique. The excitation wavelength at 620 nm emitted from an optical parametric amplifier pumped by a Yb:KGW femtosecond laser with pulse width of 190 femtosecond. The beam path diagram of experimental procedure as presented in Figure S5



Figure S5 The beam path diagram of femtosecond transient absorption spectroscopy measurement.