

Supporting Information

EXPERIMENTAL METHODS

Chemicals. Selenium powder (-100 mesh, $\geq 99.5\%$), diethylamine ($>99.5\%$), diphenylphosphine (98%), diethylammonium diethyldithiocarbamate (97%), 1,10-phenanthroline (99%), europium (III) chloride hexahydrate (99.9%), europium(III) trifluoromethanesulfonate (98%), oleylamine (OLA, 70%, technical grade), hexadecylamine (HDA, 90%), 1-octadecene (ODE, 90%), anhydrous acetonitrile ($>99\%$), absolute ethanol ($>99.7\%$), anhydrous hexanes ($>99.5\%$), isopropyl alcohol (98%), and diethyl ether (99.7%) were purchased from Sigma-Aldrich and used as received.

Eu(S₂CNET₂)₃Phen (1). The europium tris-diethyldithiocarbamate phenanthroline single-source precursor was synthesized as previously reported. Briefly, europium(III) chloride hexahydrate was dissolved in isopropyl alcohol and added to a vigorously stirred solution of diethylammonium diethyldithiocarbamate and 1,10-phenanthroline in acetonitrile at room temperature. The orange-red precipitate was collected via vacuum filtration and washed with ice-cold acetonitrile. FT-IR (v, cm⁻¹): 1490(s), 962(m).

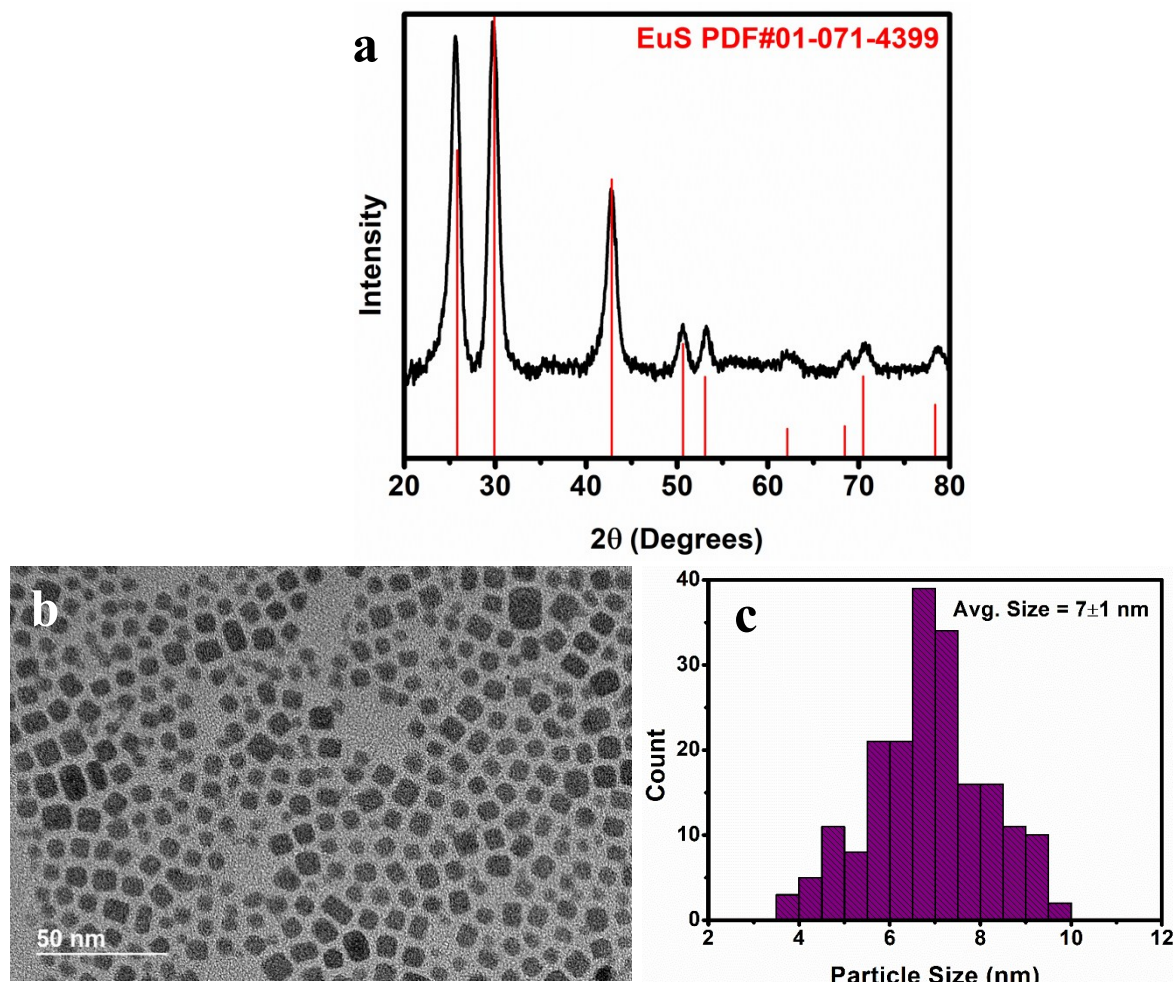
[Et₂NH][Se₂PPh₂] (DSP salt) (2). In a nitrogen-filled glovebox, selenium (7.9 g, 0.10 mol) was mixed with anhydrous ethanol (40 mL). Diphenylphosphine (8.7 mL, 0.05 mol) was added to the slurry. Excess diethylamine (5.3 mL, 0.051 mol) was injected into the reaction and lowered into a water bath at 60°C and stirred for 60 minutes. The solution turned from black to a rust-red color over the course of the reaction. The reaction was cooled to room temperature and then transferred to a freezer at -30°C. A white precipitate formed and was vacuum filtered and washed with ethanol and diethyl ether to isolate the product. FT-IR (v, cm⁻¹): 1544(w), 1433(s), 1088(m), 751(m), 691(m), 534(s), 514(s), 472(m), 450(m).

Eu(Se₂PPh₂)₃(MeCN)₂ (3). In a nitrogen-filled glovebox, **2** (1.3 g, 3.0 mmol) was dissolved in acetonitrile (25 mL). Europium(III) trifluoromethanesulfonate (0.60 g, 1.0 mmol) was added directly into the ligand solution. The solution started to form a precipitate upon addition of the triflate salt and was stirred for 30 minutes. The product was isolated via gravity filtration and washed with acetonitrile. FT-IR (v, cm⁻¹): 3047(w), 2296(w), 2266(w), 1480(w), 1435(m), 1323(w), 1307(w), 1178(w), 1160(w), 1092(m), 1063(w), 1025(w), 998(w), 744(m), 690(s), 620(w), 548(m), 514(s), 476(s), 447(w), 421(w).

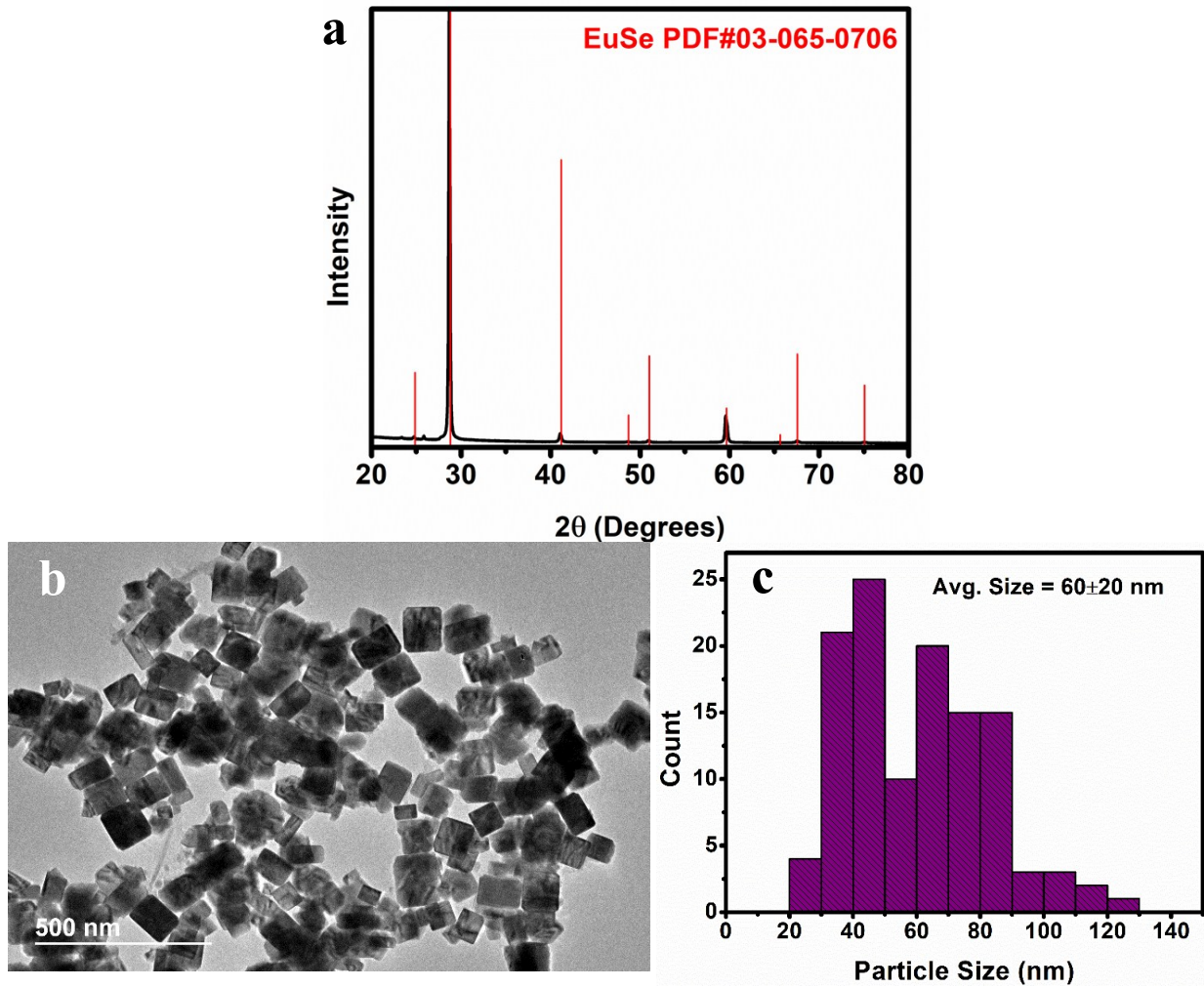
EuS/EuSe Nanoparticles. In a 3-necked round bottom flask fitted with a thermocouple for temperature control and reflux condenser, HDA (2.50 g, 10.4 mmol) and ODE (5.0 mL, 25 mmol) were degassed under vacuum at 75°C for 1 hr. In a nitrogen-filled glovebox, **1** (or **3**) (0.334 mmol) were dissolved in 3.0 mL of OLA. The HDA/ODE solution was placed under N₂ and heated to 330°C, and the OLA solution of **1** (or **3**) were injected via syringe. The color change was immediate upon injection, and the reaction was stirred for 1 hour. The reaction was then cooled from 330°C to room temperature and hexanes introduced to disperse the nanocrystals and decanted into a centrifuge tube to remove solid HDA. Nanoparticles were precipitated with EtOH and collected by centrifugation at 4500 rpm for 10 minutes. The pellet was re-dispersed in hexanes and the precipitation and centrifugation procedure was repeated a total of 4 times to wash the nanocrystals. The nanoparticles were re-suspended in hexanes or kept as powders.

Characterization. Infrared spectra were measured in the range 450-4000 cm⁻¹ as pressed pellets in KBr on a Perkin Elmer FTIR. Powder X-ray diffraction (PXRD) patterns were collected on a Rigaku Ultima IV diffractometer equipped with a high-speed DTEX silicon-strip detector using Cu K α

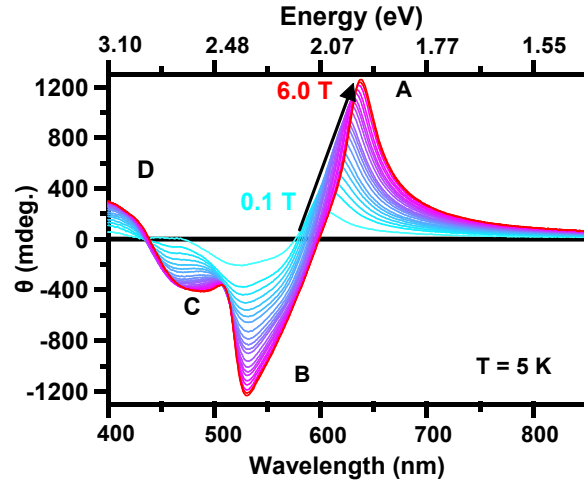
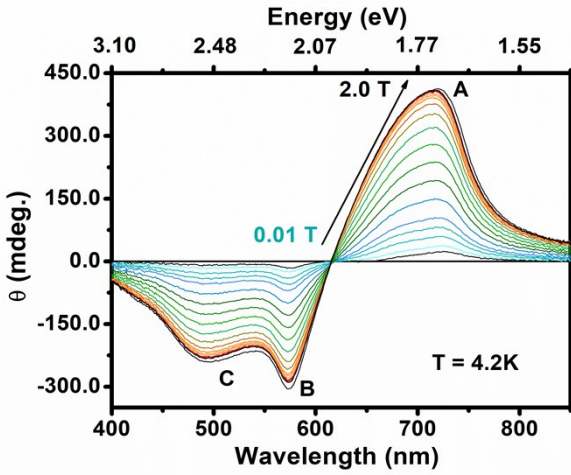
radiation at a scan speed of $1^\circ/\text{min}$ at 40 kV/44 mA. Samples were prepared for transmission electron microscopy (TEM) by drop-casting nanoparticle samples dispersed in hexanes onto amorphous carbon-coated Cu grids. High-resolution TEM (HRTEM) were performed on a JEOL JEM-2100F FEG TEM operated at 200 kV at the Advanced Imaging and Microscopy Lab in University of Maryland. EuS/EuSe samples for magnetic circular dichroism (MCD) were drop-casted as colloids in hexanes onto quartz disks or dry powders were mixed with PDMS then sandwiched between two quartz disks. Low-temperature MCD spectra were conducted with the samples placed in a superconducting magneto-optical cryostat (Cryo-Industries SMC- 1659 OVT) oriented in the Faraday configuration. At helium temperature, the sample was screened for depolarization by matching the CD spectra of a chiral molecule placed along the optical path before and after the sample. Depolarization of the thin films was less than 9%. MCD spectra were collected using an Aviv 40DS spectro-polarimeter. The same films were used for the low temperature absorption conducted using a flow cryostat (Janis STVP-100) and a Varian Cary 500 UV-VIS NIR spectrophotometer.



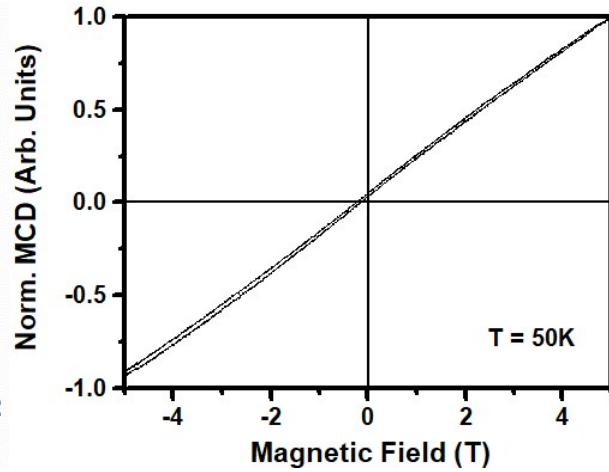
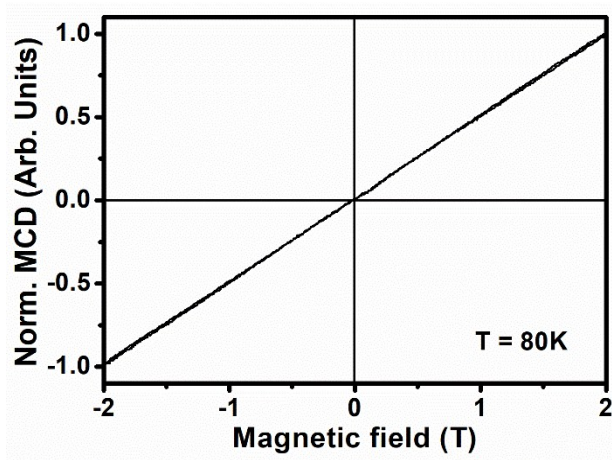
S1. X-ray powder diffraction pattern of EuS (a) and transmission electron microscopy of nanoparticles (b) with histogram (171 counts) (c).



S2. X-ray powder diffraction pattern of EuSe (a) and transmission electron microscopy of nanoparticles (b) with histogram (176 counts) (c).



S3. Field-dependent MCD spectra of EuS at 4.2K (left) and EuSe at 5K (right).



S4. Normalized field-dependent MCD intensity above T_c of EuS at 80K (left) and EuSe at 50K (right).