Supporting

Part 1. Synthesis and of $L1_0$-FePt nanoparticles, Film preparation and Characterization Methods

1. Synthesis of $L1_0$-FePt nanoparticles

The $L1_0$-FePt nanoparticles were prepared by modifying a previously reported procedure in our group. In a typical procedure, platinum acetylacetonate ($Pt(acac)_2$, 0.1 g, 0.25 mmol), oleylamine (OAm, 0.68 g), oleic aicd (OA, 0.68 g), and 1-octadecene (ODE 10 mL)were put into a four-neck flask. The mixture was stirred at 80 ^oC and degased under vacuum. Subsequently, the mixture was heated up to 120 ^oC under an argon atmosphere, and was kept at this temperature for 2 min before $Fe(CO)_5$ (0.08 ml) was injected into the solution. The temperature was quickly raised to 220 ^oC and maintained for 20 min. The mixture was then heated to 300 ^oC at 10 ^oC/min and incubated at this temperature for 10 minutes before cooled down to room temperature. The product was rinsed twice and dispersed in hexane. This $FePt/Fe_3O_4$ dispersion was stirred with magnesium(II) acetylacetonate hydrate ($Mg(acac)_2{\cdot}2H_2O$, 700 mg), OAm (5 g), OA (1.33 g), and dibenzyl ether (20 mL) at 120 ^oC in an argon atmosphere. The mixture was heated up to 300 ^oC and maintained at this temperature for 60 minutes. After cooling down, the product was washed with ethanol/hexane and then annealed at 700 ^oC for 6 h in Ar/H_2 to obtain $L1_0$-FePt/MgO NPs. The as-annealed $L1_0$-FePt/MgO NPs was dissolved in ethanol at concentrations of 1mg/ml under ultrasonication. A moderate amount of concentrated HNO_3 was drop-cast into the dispersion. After 5 minutes, $L1_0$-FePt NPs were precipitated by adding ethanol and redispersed in toluene (30 mL) with OA (0.5 mL), OAm (0.5 mL) and 1-Hexadecanethiol (0.5 mL) at 100 ^oC for 1h. Centrifugation was then applied to remove any undispersed residue. The as-synthesized $L1_0$-FePt NPs were finally dispersed in toluene for further layer-by-layer assembly.

2. Film preparation

A toluene dispersion of $L1_0$-FePt NPs and an ethanol solution of polyethylenimine (PEI) were prepared at concentrations of 15 and 5 mg/mL, respectively. Prior to LbL assembly, the quartz or silicon substrates were cleaned with an Piranha solution (H_2SO_4/30%H_2O_2 7:3 v/v) and rinsed with Milli-Q water. Further purification was carried out by immersion in an RCA solution ($H_2O/NH_3/H_2O_2$ 5:1:1 v/v/v) and thoroughly rinsed with distilled water followed by dried under a stream of nitrogen.

The substrate was immersed in ethanol PEI solution for 5min followed by rinsing with ethanol. The rising was done by dipping the substrates for 1 min each in three beakers containing ethanol, in order to remove excess PEI from the surface. The substrate was subsequently dried in gently-flowing nitrogen. The substrate was then dipped into toluene dispersion of the $L1_0$-FePt nanoparticles for 10min. After the film was rinsed 3 times in toluene for 1min each, the substrate was then placed horizontally on a strong magnet with a magnetic field normal to the film surface immediately and dried with a gentle N_2 flow. By repeating this procedure in circular fashion, we can obtain $(PEI/L1_0\text{-}FePt)_n$ multilayers assembly with desired number of layers.

3. Characterization Methods

The samples were characterized by X-ray powder diffraction (XRD) with a Rigaku D/Max-2000 diffractometer(Cu Kα, λ=1.5406 Å). The elemental composition of the samples was estimated by ICP-AES of the solution obtained after digesting the samples in a HCl/HNO_3 mixture. The transmission electron microscopy (TEM) images were obtained by a Tecnai T20 transmission electron microscope operated at an acceleration voltage of 200 kV. TEM characterization of

PEI/$L1_0$-FePt multilayer films were prepared on a carbon-coated copper grid with the same procedure as on quartz or silicon substrates. The HRTEM mages were carried out on a FEI Tecnai F30 emission analytical transmission electron microscope operated at an acceleration voltage of 300 kV. A scanning electron microscope (FEI NanoSEM 430) was employed to characterize the film thickness of the PEI/$L1_0$-FePt multilayer films and and the accelerating voltage applied was 10 kV. Infrared spectra were recorded on a Nicolet Avatar FTIR spectrophotometer in the transmission and attenuated total reflection (ATR) modes. An ATR-FTIR spectrum was obtained from 256 scans with an incident angle of 45°C and plotted after baseline correction using spectrum analyzing software (OMNIC). UV–vis absorbance spectra of PEI/$L1_0$-FePt multilayers on quartz glass were obtained by a Shimadzu UV–2550 UV–vis spectrometer system at the temperature of 23±2°C. Magnetic properties were studied using SQUID magnetometry (MPMS XL, Quantum Design).

Part 2. Experimental Results

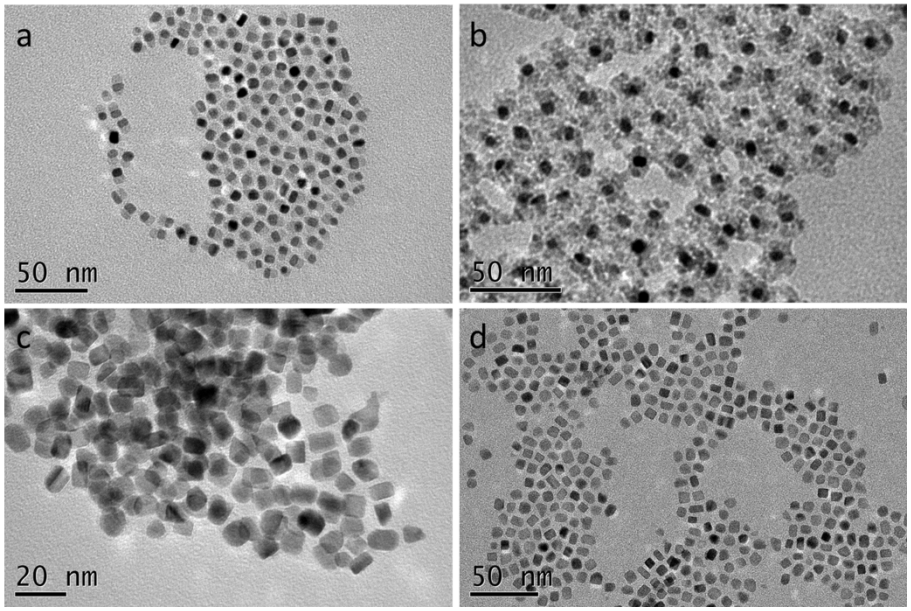

Figure S1 TEM images of the intermediate product in the synthesis process of $L1_0$-FePt nanoparticles (a) fcc-FePt-Fe_3O_4 NPs, (b) fcc-FePt-Fe_3O_4/MgO NPs, (c) $L1_0$-FePt/MgO NPs, (d) $L1_0$-FePt NPs,

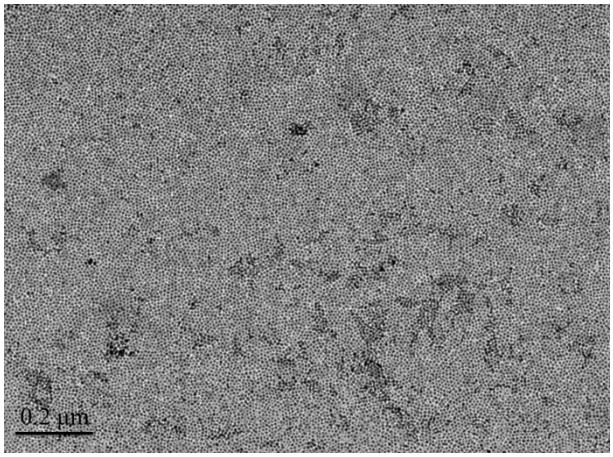

Figure S2 An overview TEM image $L1_0$-FePt NP single layer assembled on PEI-coated TEM grids.

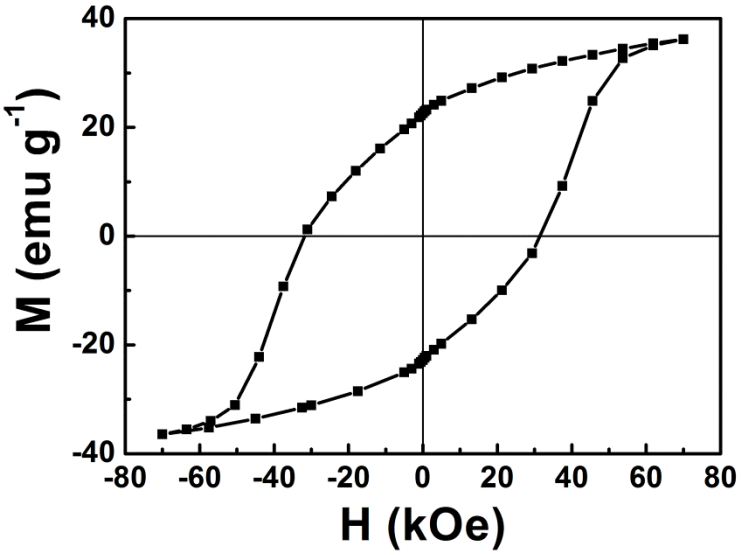

Figure S3 Room-temperature hysteresis loops of $L1_0$-FePt powders.