Supporting

Part 1. Synthesis and of L1₀-FePt nanoparticles, Film preparation and Characterization

Methods

1. Synthesis of L1₀-FePt nanoparticles

The L1₀-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 acid (OA, 0.68 g), and 1-octadecene (ODE 10 mL) were put into a four-neck flask. The mixture was stirred at 80 °C and degased under vacuum. Subsequently, the mixture was heated up to 120 °C 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 °C and maintained for 20 min. The mixture was then heated to 300 °C at 10 °C/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/Fe₃O₄ dispersion was stirred with magnesium(II) acetylacetonate hydrate (Mg(acac)₂·2H₂O, 700 mg), OAm (5 g), OA (1.33 g), and dibenzyl ether (20 mL) at 120 °C in an argon atmosphere. The mixture was heated up to 300 °C and maintained at this temperature for 60 minutes. After cooling down, the product was washed with ethanol/hexane and then annealed at 700 °C for 6 h in Ar/H₂ to obtain L1₀-FePt/MgO NPs. The as-annealed L1₀-FePt/MgO NPs was dissolved in ethanol at concentrations of 1mg/ml under ultrasonication. A moderate amount of concentrated HNO₃ was drop-cast into the dispersion. After 5 minutes, L1₀-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 °C for 1h. Centrifugation was then applied to remove any undispersed residue. The as-synthesized L1₀-FePt NPs were finally dispersed in toluene for further layer-by-layer assembly.

2. Film preparation

A toluene dispersion of L1₀-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 a Piranha solution (H₂SO₄/30%H₂O₂ 7:3 v/v) and rinsed with Milli-Q water. Further purification was carried out by immersion in an RCA solution (H₂O/ NH₃/H₂O₂ 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 L1₀-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 N₂ flow. By repeating this procedure in circular fashion, we can obtain (PEI/L1₀-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 Ka, 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/HNO₃ 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/L1₀-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/L1₀-FePt multilayer films 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/L1₀-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 L1₀-FePt nanoparticles](image1)

(a) fcc-FePt-Fe₃O₄ NPs, (b) fcc-FePt-Fe₃O₄/MgO NPs, (c) L1₀-FePt/MgO NPs, (d) L1₀-FePt NPs,

![Figure S2 An overview TEM image L1₀-FePt NP single layer assembled on PEI-coated TEM grids.](image2)
Figure S3 Room-temperature hysteresis loops of L1₀-FePt powders.