

## Supporting Information

The NPs were prepared using a modification of the reported procedure by Jia *et al.*<sup>i</sup> The synthetic experiments were carried out using standard airless procedures and commercially available reagents. The syntheses were performed by heating the precursors' solution at different temperatures 150°C, 200°C, 250°C, 350° for 3 hr. At first, a solution of platinum acetylacetone (0.5 mmol), gold acetate (0.05–0.35 mmol), and 1,2-hexadecanediol (1.5 mmol) in 20-mL octyl ether and 20 mL hexadecylamine was heated up to 100° C in a three-necked, round-bottom flask under a nitrogen atmosphere. To this solution was added, via syringe, oleic acid (0.5 mmol), oleylamine (0.5 mmol), and iron pentacarbonyl (1 mmol). The mixture was heated to reflux and allowed to reflux for 30 min or 3hr resulting in a black dispersion. Then the heat source was removed and the dispersion was allowed to cool to the room temperature. The inert gas protected solution could then be opened to ambient environment. Adding 40-mL ethanol precipitated the black product. The mixture was centrifuged to isolate the particles from the brown supernatant. The particles were redispersed in hexane, precipitated with ethanol, and isolated by centrifuging. The particles were dried at room temperature in a vacuum oven to give 100–200 mg of particles. The dispersion and precipitation removed impurities. During synthesis, the relative amounts of platinum acetylacetone and iron pentacarbonyl and gold acetate were fixed in order to produce NPs with similar compositions (FePt)<sub>85</sub>Au<sub>15</sub>.

FePtAu NPs monolayers were prepared via polymer mediate self-assmbely.<sup>ii</sup> <sup>iii</sup> O<sub>2</sub> plasma treated silicon substrates were immersed in 20mg/ml chloroform solution of PEI for 5 min and then dipped in ethanol several times to wash away extra PEI. PEI covered silicon substrates were immersed into FePt (10mg/ml) hexane solution for 10 min to assemble FePtAu NPs on the surfaces. Then the substrates were rinsed with pure hexane to wash off physisorbed particles subsequently dry under N<sub>2</sub> flow.

FePtAu NPs monolayer patterns are prepared as flows. Patterns on the photoresist film were prepared using Hexamethyldisilazane (HMDS) modified silicon substrate via standard photolithography. The lithographic defined substrate was immersed into FePt (10 mg/ml) hexane solution for 10 min to

assemble FePtAu NPs on the surfaces. Then the substrates were rinsed with pure hexane to wash off physisorbed particles. Finally, the substrate was rinse thoroughly with acetone until all the photo resist was removed.

TEM images of NPs were recorded with a Philips CM-30 Twin operating at 200 kV. A drop of hexane solution of the NPs was deposited on the carbon-coated copper grid and allowing the solvent to slowly evaporate. The average NP size is determined averaging over 150 NPs from different frames of TEM images. The NP sample was analyzed by powder X-ray diffraction (XRD) analysis using a Philips X'Pert diffractometer ( $\text{CuK}_\alpha\lambda = 1.5418 \text{ \AA}$ ). XPS measurements were performed with Physical Electronics Quantum 2000 equipment. It is equipped with a spherical sector analyzer and a multi-channel plate detector. Analyzer Mode was constant pass energy. For the survey scan the pass energy was 117 eV, the X-ray beam was set to high-power mode 100 Watt/100 $\mu\text{m}$ , and the diameter beam scanned over a 1000  $\mu\text{m}$  x 500  $\mu\text{m}$  area. For element scans pass energy was 29.35 eV, the X-ray beam was set to 25 W/100  $\mu\text{m}$ , and the diameter beam scanned over a 1000  $\mu\text{m}$  x 500  $\mu\text{m}$  area. The excitation source was Al K Alpha monochromatic radiation with a source energy = 1486.6 eV. The take-off angle (analyzer angle to sample surface) was set to 30 degrees. Using both low energy electrons and ions controlled the surface potential. The temperature during analysis was 298 K, and the pressure was between 1 and  $3 \times 10^{-8}$  Torr (Argon pressure for charge control). For atomic concentration, calculation the Shirley background subtraction was employed. The sensitivity factors were provided by Physical Electronics Multipack software version 6.1A. The hydrocarbon C1s signal at 284.8 eV was used as a reference for surface charging. Magnetic hysteresis curves were then recorded in a Quantum Design vibrating sample magnetometer (VSM). This gives information about the phase transformation and ferromagnetic behavior with field up to 9T and temperature between 5K and 300K.

The theoretical magnetic momentum was calculated via equation S1. In the equation,  $m$  is the magnetic momentum,  $M$  is the momentum density,  $n$  is number of NPs on the substrate,  $S$  is the area of

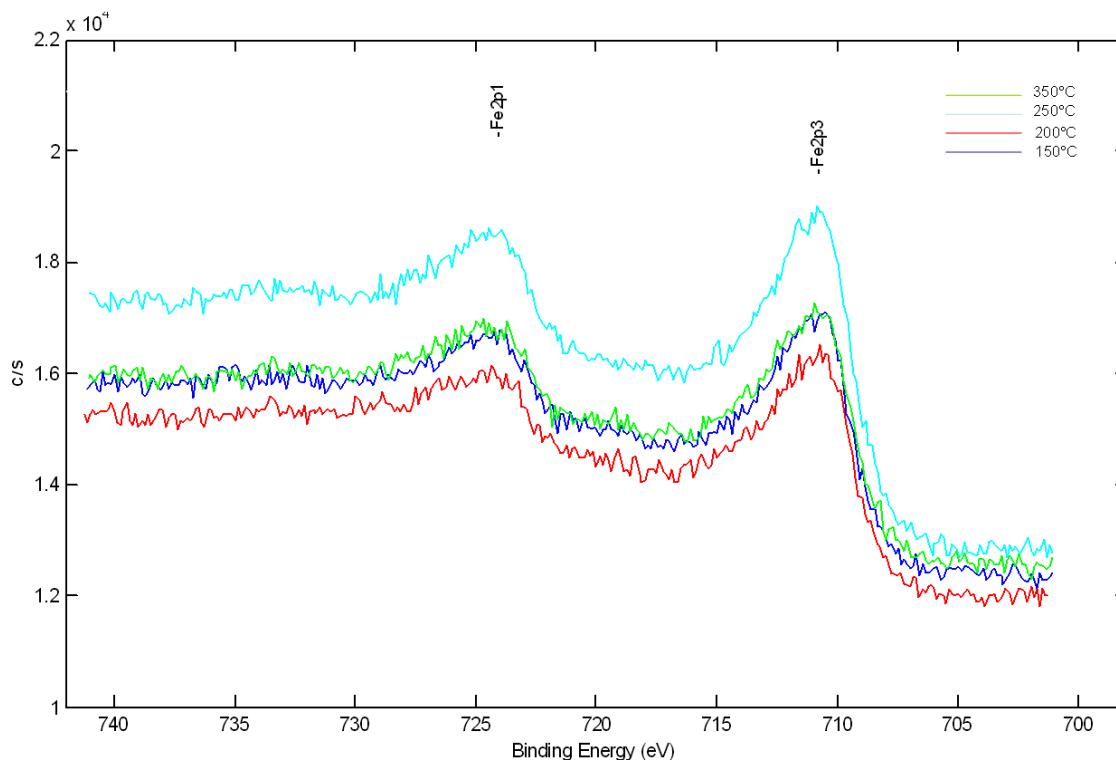
substrate,  $\rho$  is the packing density,  $r_1$  is the radius of NP,  $r_2$  is the length of the ligands and  $p$  is the ratio of FePt in FePtAu NPs.

$$m = MnV = M \frac{S\rho}{\pi(r_1 + r_2)^2} \left( \frac{4}{3} \pi r_1^3 \right) p \quad (S1)$$

Momentum density of bulk FePt is 1140 emu/cc<sup>iv</sup>. The substrate dimension is about 2.5 mm by 10 mm and the ratio of FePt in FePtAu NPs is 0.85. Assuming a hexagonal packing of NPs on substrates ( $\rho=0.9069$ ), the length of the ligands is estimated to be 1 nm. In case of FePtAu NPs anneal at 350 °C, radius of NP is 2.9 nm according to TEM analysis.

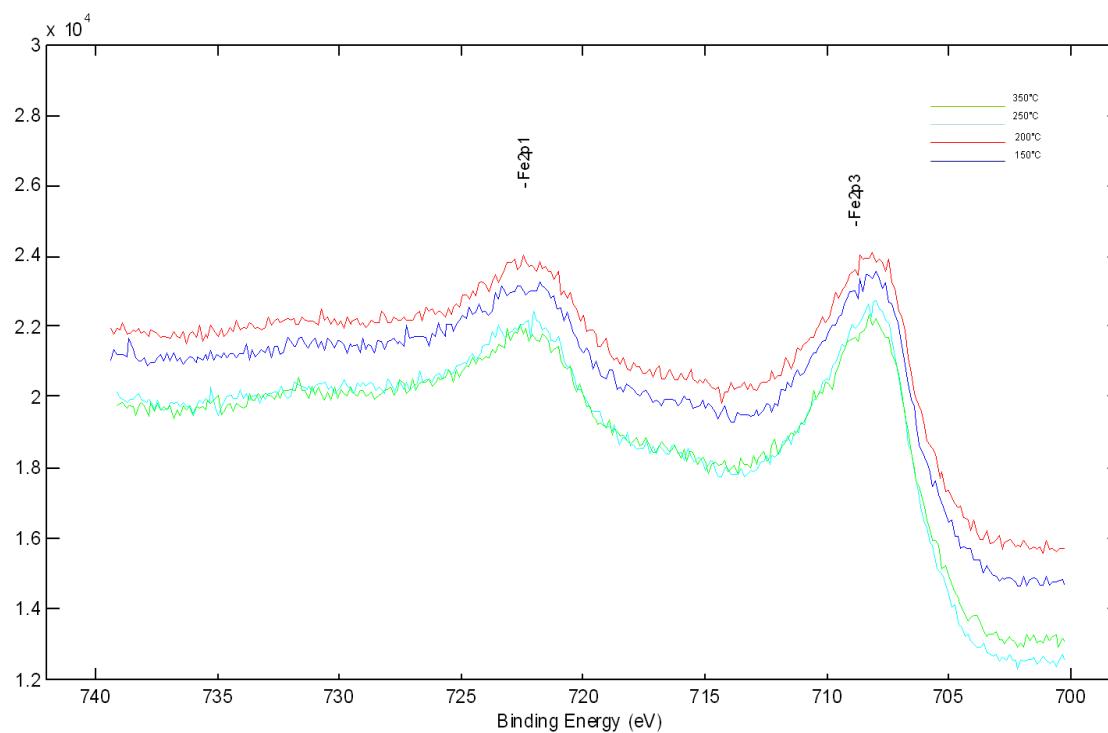
### XPS analysis of FePtAu NPs prepared via solution annealing.

In non-sputtered samples Binding Energy (BE) of (Fe<sub>2</sub>p<sub>3/2</sub>) around 710 - 711 eV, corresponds to Fe<sub>2</sub>O<sub>3</sub>. Indicating the thin surface shell consisting of Fe-oxide is present on the nanoparticle surface.



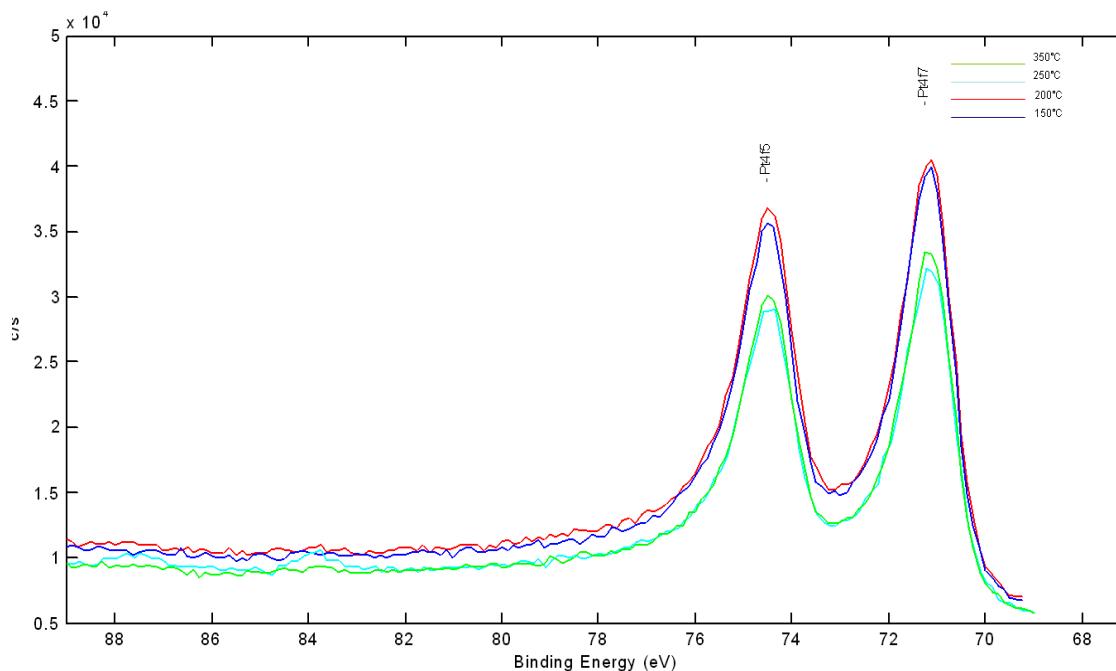
**SI Figure 1** XPS analysis of FePtAu NPs non-sputtered sample (Fe spectra)

To analyze the Fe state inside the nanoparticles, samples were sputtered (1 nm). BE (  $\text{Fe}_{2p_{3/2}}$ ) around 707 - 708 eV, corresponds to Fe pure metal, indicating that the inner core pure Fe metal.



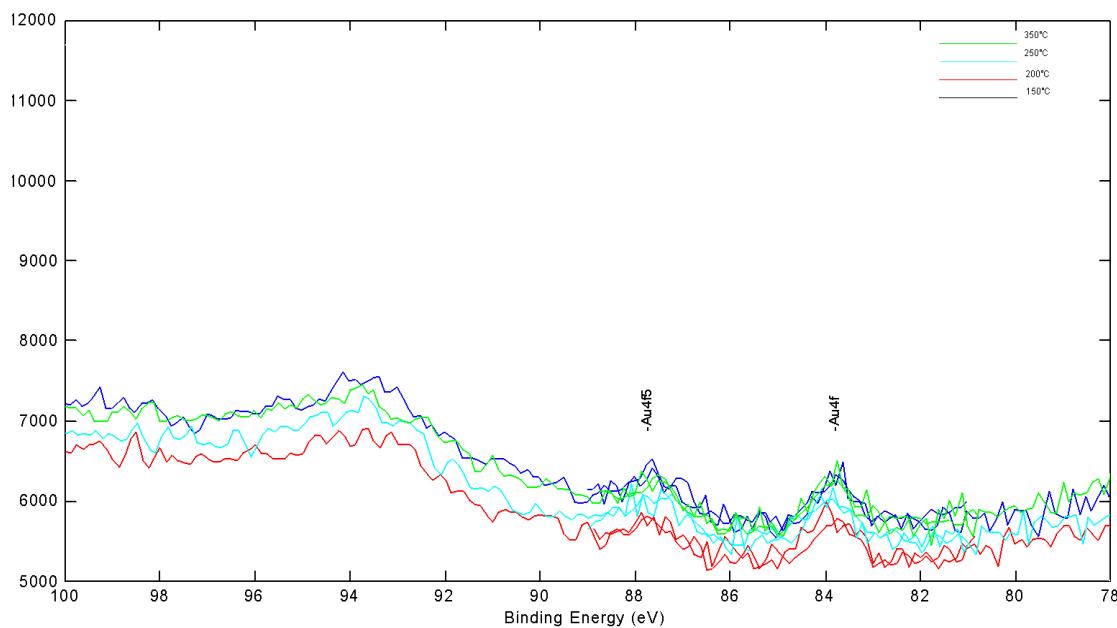
**SI Figure 2** XPS analysis of FePtAu NPs sputtered sample (Fe spectra)

Pt is in pure metallic state in all the samples BE (Pt $4f_{7/2}$ ) around 71.2 eV, corresponds pure metal.



### SI Figure 3 XPS analysis of FePtAu NPs non-sputtered sample (Pt spectra)

Au is in pure metallic state in all the samples BE( Au4f7/2) around 73.8 eV, corresponds Au pure metal.



### SI Figure 3 XPS analysis of FePtAu NPs non-sputtered sample (Au spectra)

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#### References

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