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Supplementary Information

Gigantic Plasmon Resonance Effects on Magneto-Optical Activity of Molecularly-Thin Ferromagnets near Gold Surface

Minoru Osada,[*,a,b] Natália Hajduková-Šmídová,[a,c] Kosho Akatsuka,[a] Satoshi Yoguchi,[a] and Takayoshi Sasaki[a,b]

[a]*International Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), Tsukuba, Ibaraki 305-0044, Japan*

[b]*CREST, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan*

[c]*Institute of Physics, Charles University, Ke Karlovu 5, Prague 2, 121-16,Czech Republic*

E-mail: osada.minoru@nims.go.jp

Materials and Experimental Procedures

Colloidal suspensions of ferromagnetic nanosheets ($Ti_{0.8}Co_{0.2}O_2$, $Ti_{0.75}Co_{0.15}Fe_{0.1}O_2$) and non-magnetic nanosheet ($Ti_{0.91}O_2$) were synthesised by delaminating layered titatanes according to previously described procedures.[19] Sectional analysis of AFM revealed a sheet-like morphology (with the dimension of a few μm) with the average thickness of 1.1 ± 0.1 nm (Fig. 1(a)).

Nanostructured films composed of a ferromagnetic monolayer on Au nanoparticles were fabricated by a self-assembly process. The substrates used in the experiment were polished, microscope-grade glass slides (2 x 1 cm^2) that were cleaned in piranha solution (4:1 H_2SO_4 : 30% H_2O_2) and rinsed with a Millipore filtered water. Au colloidal nanoparticles, prepared by chemical reduction of $HAuCl_4$ using sodium borohydride and sodium citrate, were immobilized to the glass substrate.[21] A colloidal suspension of ferromagnetic nanosheet was used to deposit ferromagnetic monolayer on the Au-coated substrates by Langmuir-Blodgett (LB) technique.[22] A diluted colloidal suspension (ca. 0.08 g dm^{-3}) of the nanosheets was used as a subphase to form LB films without amphiphilic molecules. During LB deposition, the packing density of the nanosheets in the film could be controlled by the surface pressure of the air–water interface.

Heteroassemblies of $Ti_{0.8}Co_{0.2}O_2/(Ti_{0.91}O_2)_n/Au$ were fabricated by electrostatic layer-by-layer assembly technique previously described in detail.[30] The Au-coated substrate was immersed in poly(diallyldimethylammonium) (PDDA) chloride solution (20 g·dm^{-3}; pH = 9) for 20 min to attain a positively charged surface, followed by washing with copious amounts of pure water. Then, the substrate was dipped in a colloidal suspension of nanosheets (0.08 g·dm^{-3}; pH = 9) for 20 min and washed again. The monolayer deposition for $Ti_{0.91}O_2$ or $Ti_{0.8}Co_{0.2}O_2$ was repeated appropriate number of times (*n*) to produce a heteroassembly composed of $Ti_{0.8}Co_{0.2}O_2/(Ti_{0.91}O_2)_n/Au$.

The film was characterized by UV-visible absorption spectroscopy, atomic force microscopy (AFM), X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS). UV-visible absorption spectra were recorded in a transmission mode using a Hitachi U-4100 spectrophotometer. Film surface morphology was analyzed using an SII Nanotechnology E-sweep AFM. XRD patterns were collected by a Rigaku RINT 2200 diffractometer using monochromatized Cu Kα radiation (λ = 0.15405 nm). XPS was recorded on a Physical Electronics XPS-5700 spectrometer with Al Kα X-ray line (1486.6 eV).

Magneto–optical measurements were carried out at room temperature with a Kerr configuration in a magnetic field (10 kOe) parallel to the out-of-plane magnetization. Alternating right and left circularly polarized monochromatic lights from a Xe lamp (50 kHz) were produced by a photoelastic modulator. The reflected light was detected by a photomultiplier tube and a lock-in amplifier.