Supplementary Materials

Ultrathin conformal coating for complex magneto-photonic structures

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Figure S1. Properties of randomly distributed MnFe₂O₄ nanoparticles. A thick (~ 200 nm) layer of randomly distributed MnFe₂O₄ nanoparticles (panel a) was synthesized by appropriate conditions in the microwave vessel: 0.3M precursors (Fe³⁺ and Mn²⁺) concentration in 1.5ml solvent was used to deposit the NPs on a vertically immersed optical glass slide into the microwave reaction solution, in order to have reference and comparison for the magneto-optical behavior of NPs in a disordered system. The sample was heated up to 170 °C and the set temperature was kept constant for 5 min. This layer was measured to obtain the saturated magnetic moment (~50 emu/g, see panel c) close to the bulk value. ¹ In b) we show the X-ray diffraction $\theta - 2\theta$ spectra, displaying the peaks of the spinel structure. Although the MnFe₂O₄ nanoparticles show ferromagnetic behavior at low temperatures (see upper inset of panel c) at room temperature are superparamagnetic (lower inset of panel c) Zero field cooled and field cooled (500 Oe) magnetization measurements in d) show a blocking temperature (~78 K) compatible with a size of around 5 nm.



Figure S2. High Angle Annular Dark Field HAADF-STEM image of the SiO₂ opal. An electron energy loss spectroscopy (EELS) line profile has been obtained from the inner (A) to the outer (B) part of the opal. Typical individual spectra obtained on both regions are shown on the right bottom figures. Notice that in the inner part (A), we mainly find the O K-edge corresponding to the SiO₂. When approaching to the borders, the Mn and Fe $L_{2,3}$ edges become clearer, meaning that the nanoparticles create a surface shell.



Figure S3. EELS spectrum image at the surface of a SiO₂ sphere. (a) HAADF image of a SiO₂ sphere with nanoparticles. EELS mappings carried out over the squared area in (a), show the distribution of (b) O, (c) Mn and (d) Fe over this zone. Notice that the presence of Mn and Fe is mainly found on the external surface of the sphere.



Figure S4. The coverage degree and thus optical properties influenced by nonhomogeneity in opal thickness. Figure S.4 a) presents SEM images of sample D1, having surfaces with different coverage degree. The coverage, less (zone 1) or more (zone 4) will affect the light transmission through the opal, as can be seen in optical transmittance (%) spectra (b) and the magneto-optical response, respectively (c). We observe that the position of the stop-band is red-shifted for increasing magnetic content, in agreement with the discussion of Figure 3 in the manuscript. We see, however, that an excess of magnetic loading (zones 3 and 4) leads to a degradation of the optical and magneto-optical properties (see (b) and (c)). Thus, an optimal magnetic coverage (zone 2) can be identified with optimal functional properties.



Figure S5. 3D SiO₂ opals covered with Fe_2O_3 NPs. Control of the amount of coating coverage by the reaction time. Fig S.4 a) and b) present the surface and the cross-section of an opal with incomplete coating (2 min-reaction time). The light color and small bright spots are the small nanoparticles on the surface of the silica spheres. For longer reaction times (5 min) the silica surface is almost completely covered. This could be observed in SEM images S.4 c) for surface and d) for-cross-section.



Electronic Supplementary Material (ESI) for Nanoscale This journal is The Royal Society of Chemistry 2011

1. Tang, Z. X., Sorensen, C. M., Klabunde, K. J., Hadjipanayis, G. C. Size dependent Curie temperature in nanoscale $MnFe_2O_4$ particles. *Phys. Rev. Lett.*, 67, 3602 (1991).