Designer Magnetoplasmonics with nickel nanoferromagnets

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EXPERIMENTAL PROCEDURES:

Hole-mask colloidal lithography (HCL)[1]. This bottom-up nanofabrication method relies on the use of charged colloidal nanospheres, dispersed on an oppositely charged substrate, to earn short-range ordered arrays of nanoscale features like nanodisks, nanoellipses, dimers, multimaterial nanosandwiches on the large (several cm^2) areas of the substrate. The following process steps were applied in all presented examples. Glass substrates – 10 x 10 x 1 mm pieces of microscope slide glass (VWR International). Glass cleaning – 5 min cycles of acetone, isopropanol and water ultrasonication. Spin coating of poly(methyl methacrylate), PMMA film (Microchem Coorporation, 2 wt % PMMA diluted in anisole, MW = 950000) onto a clean surface and soft baking (170 °C, 10 min on hotplate). Reactive oxygen plasma treatment (50 W, 5 s, 250 mTorr, Plasma Therm Batchtop RIE 95m), to decrease the polymer film hydrophobicity. This is done in order to avoid spontaneous de-wetting of the surface during subsequent polyelectrolyte and particle deposition steps, which would introduce inhomogenities in the particle distribution. Providing a net charge to the PMMA surface by pipetting a solution containing a positively charged polyelectrolyte onto the film (polydiallyldimethylammonium (PDDA) MW 200000–350000, Sigma Aldrich, 0.2 wt % in Milli-Q water, Millipore), followed by careful rinsing with de-ionized water in order to remove excess PDDA and blow-drying in a N2 stream. Deposition of a water suspension containing negatively charged polystyrene particles (sulfate latex, Invitrogen, 0.2 wt % in Mili-Q water) and N2 drying in a similar fashion as described above, leaving the PMMA surface covered with uniformly distributed PS-spheres. Evaporation of a thin, oxygen plasma resistant film of Au.

Removing the PS-spheres using tape stripping (SWT-10 tape, Nitto Scandinavia AB), resulting in a mask with holes arranged in a pattern determined by the self-assembled colloidal particles. Transfer of the hole-mask pattern into the sacrificial layer via an oxygen plasma treatment (50 W, 250 mTorr, Plasma Therm Batchtop RIE 95m), which effectively removes all PMMA situated underneath the holes in the film, leaving the surface covered with a thin film mask supported on a perforated, undercut polymer film. Ni deposition is done by e-beam-assisted evaporation (AVAC HVC600). Lift off was done using acetone at room temperature or 50 °C for 5–10 min.

s-SNOM[2]. Near-field imaging was done with a commercial s-SNOM from Neaspec (www2.neaspec.com). The basis of this s-SNOM is an atomic force microscopy, AFM. The tip oscillates at the mechanical resonance frequency $\Omega \approx 300$kHz with amplitude $\Delta = 30$nm while the sample is scanned. For the experiments we used silicon tips and illuminated the sample with s-polarized light ($\lambda = 632.8$ nm) under an angle of 30º relative to the sample surface. We detected the p-polarized scattered light, yielding the vertical field component[3]. The major advantage of this orthogonal excitation-detection scheme is that s-polarized light can effectively excite the sample while keeping the AFM tip unaffected. Amplitude and phase imaging has been performed by pseudoheterodyne interferometric detection[2]. Background contributions are suppressed by demodulating the detector signal at a harmonic frequency $n\Omega$ ($n \geq 2$), yielding background free near-field amplitude and phase signals Sn and φn respectively. In the presented experiments, demodulation has been done at n = 4. The simulated amplitude and phase of the vertical near-field component are calculated employing a commercial software (Lumerical).

Optical characterization. The baseline-corrected optical (absorptance) spectra are measured in transmission using Varian Cary 5000 spectrophotometer.

MOKE[4]. The magneto-optic response was measured with a home-built longitudinal MOKE setup. Two different laser sources (405 and 633nm) were employed separately. The system is based on the polarization modulation method using a photo elastic modulator (PEM) operating at 50kHz and

detecting the Kerr rotation signal by a lock-in amplifier as function of an external magnetic field. All the parameters that could lead to a MOKE sign reversal, e.g. analyzer angle, incident beam direction and magnetization field direction, are fixed during the measurements. The spectroscopical Kerr rotation response was performed with the same setup, employing a tunable supercontinuum laser source.

Calculations of Kerr rotation. The Kerr rotation is calculated based on the polarizability tensor induced by the incident field at presence of an applied static magnetic field[5]. We approximate the Ni disks to spheroids of semi-axes a, b and c (a=b), volume V and shape factor L, immersed in a medium with the dielectric constant ε_m. The polarizability tensor can be written as:

$$\alpha = V\left(\varepsilon - \varepsilon_m I\right) / \left(L \cdot \varepsilon + (I - L)\varepsilon_m\right),$$

where I is the unit matrix.

For the MOKE configuration, depicted in Figure 2, the dielectric tensor is defined by the diagonal terms ε_{xx} and the off-diagonal terms ε_{yz}. The Drude model is used to describe the plasmon resonance in the particles. Thus, the diagonal term of the dielectric constant is $\varepsilon_{xx} = 1 - \omega_p^2 / \omega(\omega + i\gamma)$, where ω_p and γ are the angular plasma frequency and the collision rate, respectively. To have a better fit to the experimental dielectric constant data of Ni, ω_p=9.7 eV and γ=2.3 eV are considered. The off-diagonal term is $\varepsilon_{yz} = -i\varepsilon_{xx}Q$ where tabulated data for the MO parameter Q are used (http://www.msd.anl.gov/groups/mf/jmkerrcalc.php).

Finally, the Kerr rotation is calculated as : $\Phi_K \propto \Re(\alpha_{yz}\alpha_{xx}^*) / |\alpha_{xx}|^2$.

The size corrections (radiation and depolarization) are applied to the polarizability of the particle considered here by using:

$$\tilde{\alpha} = \alpha \Big/ \left(1 - i\frac{2V}{3}k^3\alpha - \frac{V}{D}k^2\alpha\right)$$, where k is wavenumber and $D \in$ (a, b, c).

This modifies the plasmonic behavior of the large particles, obtained using the quasistatic theory, by correcting the resonance energy and line-width. In addition, to account for the substrate effect on the LSP resonances of three samples with D = 50, 100, and 180 nm, ε_m = 1.44, 1.25, and 1.21, respectively.

REFERENCES

(1) Fredriksson, H.; Alaverdyan, Y.; Dmitriev, A.; Langhammer, C.; Sutherland, D. S.; Zäch, M.; Kasemo, B. *Adv. Mater.* **2007**, *19*, 4297-4302.

(2) Ocelic, N.; Huber, A.; Hillenbrand, R., *Appl. Phys. Lett.* **2006**, *89*, 101124.

(3) Esteban, R.; Vogelgesang, R.; Dorfmueller, J.; Dmitriev, A.; Rockstuhl, C.; Etrich, C.; Kern, K. *Nano Lett.* **2008**, *8*, 3155-3159.

(4) Vavassori, P. *Appl. Phys. Lett.* **2000**, *77*, 1605-1607.

(5) Sepúlveda, B.; González-Díaz, J. B.; García-Martín, A.; Lechuga, L. M.; Armelles, G. *Phys. Rev. Lett.* **2010**, *104*, 147401.

SUPPLEMENTARY FIGURES:

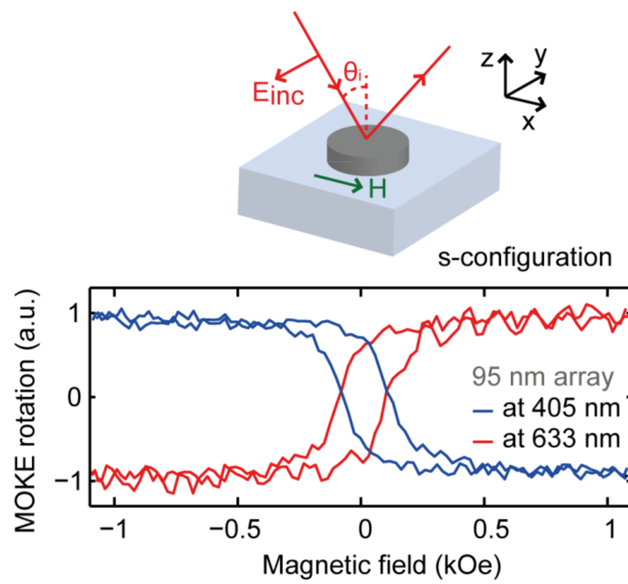

Figure S1. S-polarization MOKE on nickel nanodisks of 95 nm diameter employing two different excitation wavelengths (405 nm and 633 nm). The sketch on the top of the figure represents the geometry of the MOKE setup in s-configuration.

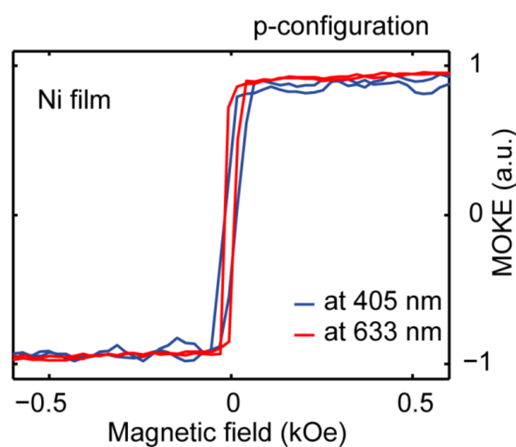

Figure S2. P-polarization MOKE on nickel film co-deposited on the same substrate, employing two different excitation wavelengths (405 nm and 633 nm).