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Plasmon-induced demagnetization and magnetic switching in nickel nanoparticle arrays

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We report on the manipulation of magnetization by femtosecond laser pulses in a periodic array of cylindrical nickel nanoparticles. By performing experiments at different wavelengths, we show that the excitation of collective surface plasmon resonances triggers demagnetization in zero field or magnetic switching in a small perpendicular field. Both magnetic effects are explained by plasmon-induced heating of the nickel nanoparticles to their Curie temperature. Model calculations confirm the strong correlation between the excitation of surface plasmon modes and laser-induced changes in magnetization. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5012857>

Plasmonic nanostructures enable strong local enhancements of the optical field in areas that are substantially smaller than the wavelength of incident light. This capability offers prospects for magnetic recording. In heat-assisted magnetic recording (HAMR), a plasmonic near-field transducer (NFT) reduces the switching field of high-anisotropy materials via local heating.^{1–3} The NFT in this application is a noble metal nanostructure that is placed near the recording medium. Excitation of the NFT at the plasmon resonance frequency efficiently transfers optical energy to a nanoscale region, enabling local switching at a reduced magnetic field. In the realm of ultrafast all-optical switching (AOS),^{4,5} the use of noble metal plasmonic antennas has also been considered. By placing gold antennas on top of a ferrimagnetic TbFeCo film, Liu and co-workers demonstrated the confinement of magnetic switching to sub-100 nm length scales.⁶

Independent from HAMR and AOS, a new discipline combining plasmonics and magnetism has emerged recently.^{7,8} Experiments on pure ferromagnetic metals demonstrate that, despite stronger ohmic damping, they also support surface plasmon resonances.^{9–11} This raises the question if one could nanostructure the magnetic medium itself to trigger magnetic switching via local enhancements of the optical field. To study the effect of plasmon resonances on magnetic switching, it is advantageous to consider ferromagnetic nanoparticles of uniform size and shape. In such nanoparticles, localized surface plasmon resonances (LSPRs) determine the magneto-optical activity via the excitation of two orthogonal electric dipoles.¹² Plasmon resonances in single ferromagnetic nanoparticles are rather broad, several hundreds of nanometers. Yet, ordering of the particles into a periodic array significantly narrows the spectral response.¹³ In this geometry, hybridization between LSPRs and the diffracted orders of the array produces intense surface lattice resonances (SLRs).^{14–17} SLR modes and, thereby, the optical, magneto-optical, and magnetic circular dichroism (MCD) properties of a ferromagnetic nanoparticle array can

be tuned by changing the period or symmetry of the lattice^{13,18,19} or the shape of the nanoparticles.²⁰ These versatile designer tools could thus be exploited to spectrally gauge the significance of the inverse Faraday effect and MCD on AOS, a topic of intense scientific debate,^{5,21–33} or to maximize their impact in monochromatic switching experiments.

The excitation of surface plasmon resonances in ferromagnetic nanoparticles can cause significant heating. In granular FePt layers where the size, shape, and environment of the ferromagnetic nanoparticles vary, near-field modifications of incident laser radiation have been found to produce an inhomogeneous magnetic switching effect.³⁴ Other optical effects such as the interference of light in GdFeCo microstructures have also been considered.³⁵ Here, we study how plasmon resonances affect optical demagnetization and magnetic switching in uniform nickel nanoparticles. We intentionally order the nickel nanoparticles into a regular array to enhance optical absorption in a narrow spectral range via the excitation of a SLR mode.¹³ This approach allows us to correlate plasmonic and magnetic effects with good spectral resolution (the LSPR mode of a random distribution of nickel nanoparticles would be too broad). Using femtosecond laser pulses of different wavelengths, we show how the excitation of a collective SLR significantly lowers the pulse fluence that is required to manipulate the magnetization state. Using a phenomenological model, we establish a link between the optical properties and temperature response of the nickel nanoparticle array during femtosecond laser excitation. From this analysis, we conclude that plasmon-induced heating of the nickel nanoparticles to their Curie temperature explains the magnetic effects.

For this study, we fabricated a periodic array of single-domain nickel nanocylinders with a diameter of 100 nm and a thickness of 110 nm on a glass substrate using electron beam lithography [Fig. 1(a)].¹³ The period (p) of the square lattice was 420 nm. In all measurements, the nickel nanoparticles were immersed into an oil that matches the refractive index of the glass substrate ($n = 1.523$) to enhance the intensity of plasmon resonances. The refractive index of nickel in

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FIG. 1. (a) Scanning electron microscopy image of the nickel nanoparticle array. (b) Optical transmission spectrum. (c) Polar magneto-optical Kerr effect hysteresis loop of the nickel nanoparticle array. (d) VSM measurement of the saturation magnetization as a function of temperature, indicating a Curie temperature of about 600 K. The line represents a fit to the data using $M_s = A(T_C - T)^\gamma$, with $A = 0.117$ and $\gamma = 0.37$.

the wavelength range of our experiments varies slightly ($n = 1.98\text{--}2.05$).³⁶ Figure 1(b) shows the transmission spectrum of the array. The spectrum, which was measured using a broadband supercontinuum laser (NKT SuperK EXW-12 with acousto-optical filter), consists of a transmission maximum at the diffracted order of the lattice ($\lambda = np = 640$ nm). The transmission minimum caused by excitation of the SLR mode, such as observed at $\lambda = 660$ nm, is associated with strong optical absorption.^{14–17}

Figure 1(c) shows a polar magneto-optical Kerr effect hysteresis loop of the array. The shape of the cylindrical nanoparticles ensures a large perpendicular magnetization in zero magnetic field. The coercive field of the nickel nanoparticles is 62 mT. Using vibrating sample magnetometry (VSM), we measured the dependence of the saturation magnetization on temperature [Fig. 1(d)]. From these data, we estimate a Curie temperature of 600 K, which is only slightly smaller than the transition temperature of bulk nickel.

To study the effect of femtosecond laser pulses on the magnetization of the nickel nanoparticle array, we used a Ti:Sapphire laser with an optical parametric amplifier (OPA). We operated the laser at different wavelengths, ranging from 610 nm to 700 nm, in 10 nm intervals. The repetition rate of the laser was 1 kHz. We reduced the frequency to 100 Hz with a chopper and used an optical beam shutter to isolate single pulses with a duration of 100 fs. The laser spot size on the sample was $720\text{ }\mu\text{m}$. We used a dark-field magneto-optical microscopy technique to monitor magnetization changes. In our setup, light from a LED source illuminated the glass sample from the side and a CCD camera detected

the scattered light from the nanoparticles. We attained images with good magneto-optical contrast by aligning a polarizer and analyzer at an angle of about 80° . Figure 2 illustrates the experimental setup.

The results for femtosecond laser pulses with $\lambda = 660$ nm are summarized in Fig. 3. Prior to laser pulse illumination, the magneto-optical contrast was calibrated by full magnetization reversal in a perpendicular magnetic field. After aligning the magnetization along one of the perpendicular directions, we turned off the magnetic field and fired a single laser pulse onto the nickel nanoparticle array. The magneto-optical

FIG. 2. Schematic of the femtosecond laser setup with a dark-field magneto-optical microscope. The nickel nanoparticle array is illuminated by a single femtosecond laser pulse. Before illumination, the magnetization of the array is set by a perpendicular magnetic field. The optically induced local change in magnetization is imaged by recording light from a LED source after scattering on the nanoparticle array. Polarizing optics in the incident and scattered beams provide the magneto-optical contrast.

FIG. 3. (a)–(d) Magneto-optical microscopy images of the nickel nanoparticle array after illumination with a single femtosecond laser pulse. The pulse fluence is increased gradually. Femtosecond laser pulses are applied in zero magnetic field, and the magnetization is reset after each measurement. (e) Line scans of magneto-optical contrast through the center of the illuminated area. (f)–(i) Magneto-optical microscopy images of the nickel nanoparticle array after illumination with a single femtosecond laser pulse in a perpendicular magnetic field of 5 mT. (j) Line scans of magneto-optical contrast for this configuration. All measurements are performed with a laser wavelength of 660 nm.

microscopy images of Figs. 3(a)–3(d) depict the change in magnetization as a function of pulse fluence. We note that the magnetization was reset by an external magnetic field after each laser pulse to avoid accumulative effects. Line scans of magneto-optical contrast through the center of the illuminated area indicate that the magnetization of the array starts to decrease above a pulse fluence of 2.8 mJ/cm² [Fig. 3(e)]. A fluence of 3.9 mJ/cm² fully randomizes the magnetization of the nickel nanoparticles in the center of the laser spot. We obtained similar results when the magnetization of the nickel nanoparticles was set along the opposite perpendicular direction, i.e., starting from a normalized magneto-optical contrast of +1.

In our experiments, we did not find conclusive evidence for all-optical magnetic switching. The use of linearly and circularly polarized laser pulses yielded the same results. To test the possibility of an accumulative MCD effect,^{27–32} we also illuminated our sample using multiple circularly polarized laser pulses. No evidence for all-optical stochastic switching was found. Magnetic switching could be attained by applying a small magnetic field of 5 mT (only 8% of the coercive field) during a single femtosecond laser pulse [Figs. 3(h)–3(n)]. Under these conditions, a pulse fluence of 4.2 mJ/cm² reverses the magnetization of most nanoparticles. The absence of a clear dependence on helicity points towards plasmon-induced heating as the source of optical demagnetization and field-assisted magnetic switching.

To assess the influence of heating, we now wish to establish a link between optical absorption by the nickel nanoparticle array and the required pulse fluence for demagnetization and switching. The extinction of the array, defined as $E = 1 - T$, is the sum of absorbed and scattered light intensities. Since the extinction cross section (σ_{ext}) of our sample is determined by a SLR mode that absorbs much more than it scatters,^{18,19} we can write $\sigma_{\text{ext}} = E p^2 \approx \sigma_{\text{abs}}$.

For femtosecond laser pulses, the electronic absorption of optical energy, electron-phonon thermalization, and external heat diffusion ensue successively.³⁷ The total absorbed energy (Q) during this process is given by $Q = \sigma_{\text{abs}} F \approx E p^2 F$, where F is the pulse fluence. Since it is reasonable to assume that both demagnetization and magnetic switching require heating of the nickel nanoparticles to their Curie temperature, the threshold pulse fluence (F_{th}) for both effects should scale as $1/E$. To test this assertion, we performed similar experiments as those depicted in Fig. 3 using different laser wavelengths. From these measurements, we extracted F_{th} for (1) a reduction of the magnetization to 40% of the initial remanent state, (2) full demagnetization, and (3) magnetic switching to $0.6M_s$ in a magnetic field of 5 mT. Figure 4(a) summarizes the results. It also includes the optical transmission data of Fig. 1(b), now re-plotted as $1/E$. We find a strong variation of the threshold pulse fluence with a laser wavelength. At 660 nm, where the excitation of the SLR mode maximizes the absorption of light, demagnetization and switching by a single femtosecond laser pulse are most efficient. The resemblance between the

FIG. 4. (a) Experimental threshold fluence for full demagnetization, demagnetization to $0.4M_s$ (both in zero magnetic field) and magnetic switching to $0.6M_s$ in the opposite direction (magnetic field of 5 mT). The threshold fluences are compared to the inverse extinction ($1/E$) of the nickel nanoparticle array. (b) Experimental threshold fluence (demagnetization to $0.4M_s$) and calculation of the fluence that is required to heat the nickel nanoparticles to 600 K.

wavelength dependence of F_{th} and $1/E$ confirms the crucial role of plasmon-induced heating.

As a final check, we calculate F_{th} using a phenomenological model. Just after a single femtosecond laser pulse, the brief and intense temperature increase of a metallic nanoparticle can be estimated as³⁸

$$dT = \frac{Q}{V\rho c_p(T)}. \quad (1)$$

Here, V , ρ , and c_p are the volume, mass density, and specific heat capacity of the nanoparticle. For nickel, the heat capacity varies with temperature.³⁹ The required energy for heating the nanoparticle from room temperature to a Curie temperature of 600 K can thus be obtained from

$$Q_{\text{CT}} = \int_{T=293\text{K}}^{T=600\text{K}} \frac{Q}{V\rho c_p(T)} dT. \quad (2)$$

After this, the threshold fluence is given by

$$F_{\text{th}} = \frac{Q_{\text{CT}}}{\sigma_{\text{abs}}} \approx \frac{Q_{\text{CT}}}{Ep^2}. \quad (3)$$

Figure 4(b) shows the result of this calculation for our cylindrical nickel nanoparticles. Here, we used the experimentally measured extinction as an input parameter. The agreement between the measured threshold fluence for demagnetization and the theoretical model indicates that the nanoparticles are indeed heated to their Curie temperature by the femtosecond laser pulse. Thus, the excitation of an intense SLR mode strongly modulates the pulse fluence that is required to manipulate the magnetization of the nickel nanoparticles.

In summary, we have shown that patterning of a magnetic medium into a periodic nanoparticle array affects thermal demagnetization and heat-assisted magnetic switching via the excitation of collective surface plasmon resonances. Whilst all-optical magnetic switching was not attained in our experiments on nickel, ferromagnetic nanoparticle arrays provide interesting designer tools for further research. In particular, the ability to spectrally separate plasmon-induced heating, the inverse Faraday effect, and MCD via lattice symmetry or nanoparticle shape could offer new insights into the origin of all-optical switching of ferromagnets.

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