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## Magnetization dynamics in optically excited nanostructured nickel films

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*New Journal of Physics* **10** (2008) 123004 (9pp)

Received 29 May 2008

Published 4 December 2008

Online at <http://www.njp.org/>

doi:10.1088/1367-2630/10/12/123004

**Abstract.** In this work, the laser-induced magnetization dynamics of nanostructured nickel films is investigated. The influence of the nanosize is discussed considering the timescale of hundreds of femtoseconds as well as the GHz regime. While no nanosize effect is observed on the short timescale, the excited magnetic mode in the GHz regime can be identified by comparison with micromagnetic simulations. The thickness dependence reveals insight into the dipole interaction between single nickel structures. Also, transient reflectivity changes are discussed.

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Nowadays, nanostructured ferromagnetic films are a key component of hard disks as well as magnetic random access memories. Furthermore, new concepts in which such films are utilized for logical operations by means of the stray field interaction between single elements are on their way [1]. In all these applications, the speed of certain operations is limited by the time needed to switch the magnetization of a single element. With regard to this technological impact, the magnetization dynamics of nanosized ferromagnetic elements has become a major field of research.

In 1996, Beaurepaire *et al* demonstrated the demagnetization within hundreds of femtoseconds of a nickel film as a response to a femtosecond laser pulse [2]. Since then, not only the physical origin of this effect has been discussed and examined extensively, but also this demagnetization mechanism has become a tool to induce and study magnetic motion of ferromagnetic films on a much longer timescale [3].

In this work, the transient change of magnetization and reflectivity of nanostructured nickel films in response to a femtosecond pump pulse is examined. We investigate the dynamics on a timescale of hundreds of femtoseconds as well as the transient changes in the GHz regime. Several reports on similar experiments have been recently published [4]–[6]. Comin *et al* discussed a coupling between elastic and magnetic modes of the film. In that work, the frequency of the observed magnetic mode does not alter by varying the external field, showing the importance of this magneto-elastic coupling. Kruglyak *et al* studied the angular dependence of the excited modes on the external magnetic field and found a new source of anisotropy in the magnetization dynamics, arising from the dynamic magnetic configuration. However, in response to the femtosecond laser pump pulse, at once, plasmonic, elastic and magnetic excitations as well as possible couplings between these excitations can determine the measured dynamics. Therefore, many aspects have not been addressed in the literature yet. In addition, the question of whether the ultrafast demagnetization is altered by the nanosize is of highest importance to reveal further insight into the microscopic processes. A recent study by Heitkamp *et al* clearly shows an influence of the s- and p-polarization of the exciting femtosecond laser pulse traced back to the dielectric response and the interaction with the light field [7]. They found an enhanced demagnetization as compared with continuous films. Hence, we note that the pump light polarization is an important parameter determining the response of the electronic system confined in the nanostructured antenna to the light field. To enlighten these aspects and to earn a comprehensive view, we vary structure size and pump light polarization to directly study the influence of the nanosize. On the short timescale, we observe no influence of the nanoscale on the ultrafast demagnetization while the transient reflectivity is enhanced due to plasmonic resonance during pump pulse duration. Further, we compare the experimentally examined magnetization dynamics on the longer timescale of a nanostructured film with the dynamics of a single nanoelement found by micromagnetic simulations. In particular, the thickness dependence is studied which allows to quantitatively determine the influence of the dipole interaction between single nanodiscs.

The thin film nanodot arrays are prepared by laser interference lithography (IL). Details about the IL steps [8] and the resist stack [9, 10] which are applied here, can be found elsewhere. Electron beam evaporated nickel is deposited in ultrahigh vacuum at a base pressure of  $5 \times 10^{-10}$  mbar on a thermally oxidized silicon substrate and capped by 2 nm of magnesium oxide layer. The remaining photoresist is removed by chemical etching in 1-methyl-2-pyrrolidinone at 120 °C. In our experiment, probe and pump pulses are generated by a non-commercial titanium:sapphire fs laser together with a regenerative amplifier (Coherent

RegA 9000, repetition rate 250 kHz, pulse width of 60–80 fs). The spot sizes of probe and pump pulses are chosen to be about 40 and 60  $\mu\text{m}$ , respectively, so that a homogeneously excited area is probed. As the typical structure size is significantly below 1  $\mu\text{m}$ , the ratio between spot and structure size is of the order of 100. Therefore, throughout this paper, any influence of the spot size on the transient changes of reflectivity and magnetization can be neglected. For the time-resolved Kerr measurements, we utilize a double modulation scheme so that the polarization of the probe pulse is modulated with a photo-elastic modulator (PEM) and the intensity of the pump pulse with a mechanical chopper. The sample is mounted at room temperature. The external magnetic field, homogeneous on the length of the sample dimension, is applied in the plane of incidence by means of a magnet coil with an iron core.

The influence of the nanoscale on the timescale of femtoseconds is discussed with respect to the dynamics we observed in an array of nickel ellipses. The nickel thickness is  $26 \pm 0.5$  nm. The long (short) axis of the ellipses averages to about 320 (130) nm. A scanning electron microscope (SEM) image of this film is shown in figure 1(a).

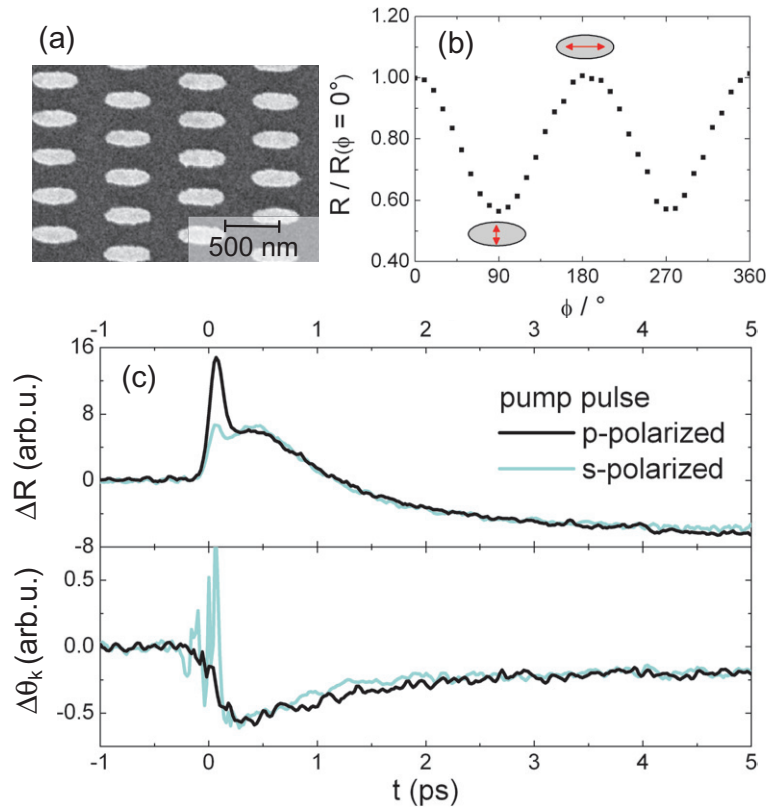
Due to plasmonic resonance [11], the reflectivity of the elliptic structures strongly depends on the polarization of the light. In figure 1(b), the reflectivity of the pump pulse as a function of its light polarization is plotted; the reflectivity is higher for p-polarized (parallel to the long-axis of the ellipse) light than for s-polarized light. This antenna effect also becomes manifest on the transient change of reflectivity as depicted in figure 1(c): during pumping the transient reflectivity of the probe pulse is enhanced for both polarization states with the enhancement being much greater for the p-polarization. After pump pulse duration, the two curves look very similar. This nanosize effect does not become manifest in the transient change of the Kerr angle that is depicted in the bottom graph of figure 1(c). It is ensured that these curves contain no transient reflectivity changes since they are formed as the difference of two Kerr transients with the external field of  $\pm 100$  mT applied in opposite direction along the sample surface. The difference between the two demagnetization curves stems from a measurement artifact, the so-called coherent artifact [12] that originates from pump–probe interference and is much more pronounced in our setup for s-polarized light due to the probe light modulation with the PEM.

This observation could help to explain the direct influence of the photonic or plasmonic field on the ultrafast demagnetization. In an early model, Zhang and Hübner explained the ultrafast demagnetization by taking directly into account the laser field [13]. The enhanced polarization within the nanostructure for p-polarized light should directly become manifest in a higher demagnetization which is not the case. Therefore, our experimental observation sets limits on how the laser field should be incorporated in a microscopic model of laser-induced demagnetization<sup>7</sup>.

For the discussion of the dynamics in the GHz regime, the magnetization and reflectivity transients of a circularly structured nickel wedge are presented. This structure consists of an square-symmetry array of nickel discs of a diameter of about 185 nm with an interdot distance of 280 nm. The slope of the nickel thickness is  $5 \text{ nm mm}^{-1}$ . An SEM image is given in figure 2(a).

The reflectivity transients are given in figure 2(d). Reflectivity oscillations of patterned films in optical pump–probe experiments have been studied since the inception of the field of picosecond ultrasonics [15]. According to the work by Lin *et al*, we can attribute the oscillatory reflectivity changes to at least two different effects: (i) in the first hundred picoseconds, probe

<sup>7</sup> Koopmans *et al* already discussed the photonic influence by means of conservation of angular momentum [14].

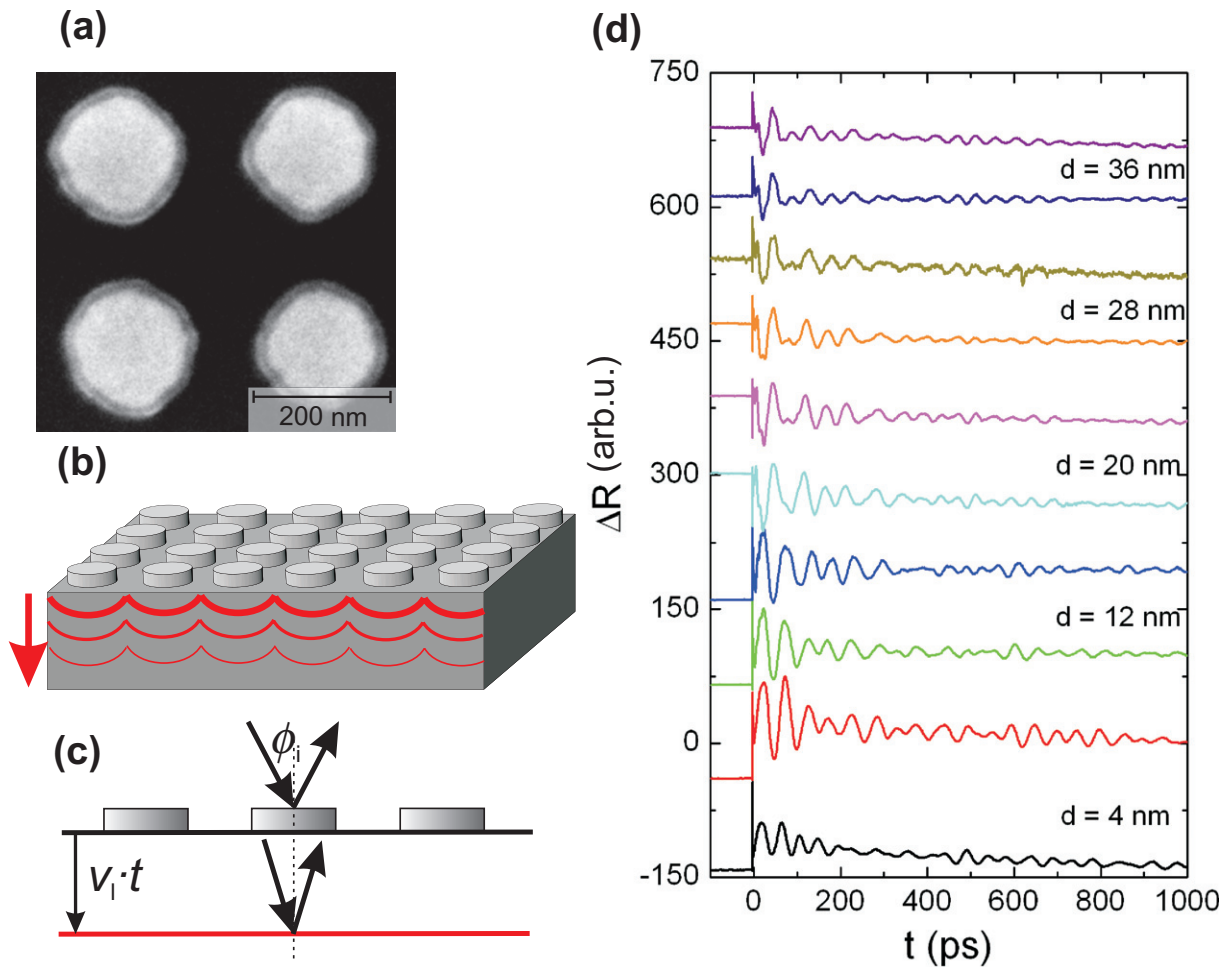


**Figure 1.** (a) SEM image of the elliptically nanostructured nickel film; the long (short) axis averages to 320 (130) nm. (b) Reflectivity of the pump light as a function of its plane of polarization; p-polarized (parallel to the long-axis) pump light has an increased reflectivity. (c) Transient reflectivity and the Kerr angle changes for s- and p-polarized pump light; the enhancement of the reflectivity due to plasmonic resonance during pump pulse duration finds no counterpart in the Kerr transients.

light reflected from the sample surface interferes with probe light that gets reflected from the wave front of the strain wave propagating into the substrate as schematically depicted in figure 2(d). According to Lin *et al*, the reflectivity oscillates due to this effect with a frequency of

$$f = \frac{2v_1\sqrt{n^2 - \sin^2 \phi_i}}{\lambda}, \quad (1)$$

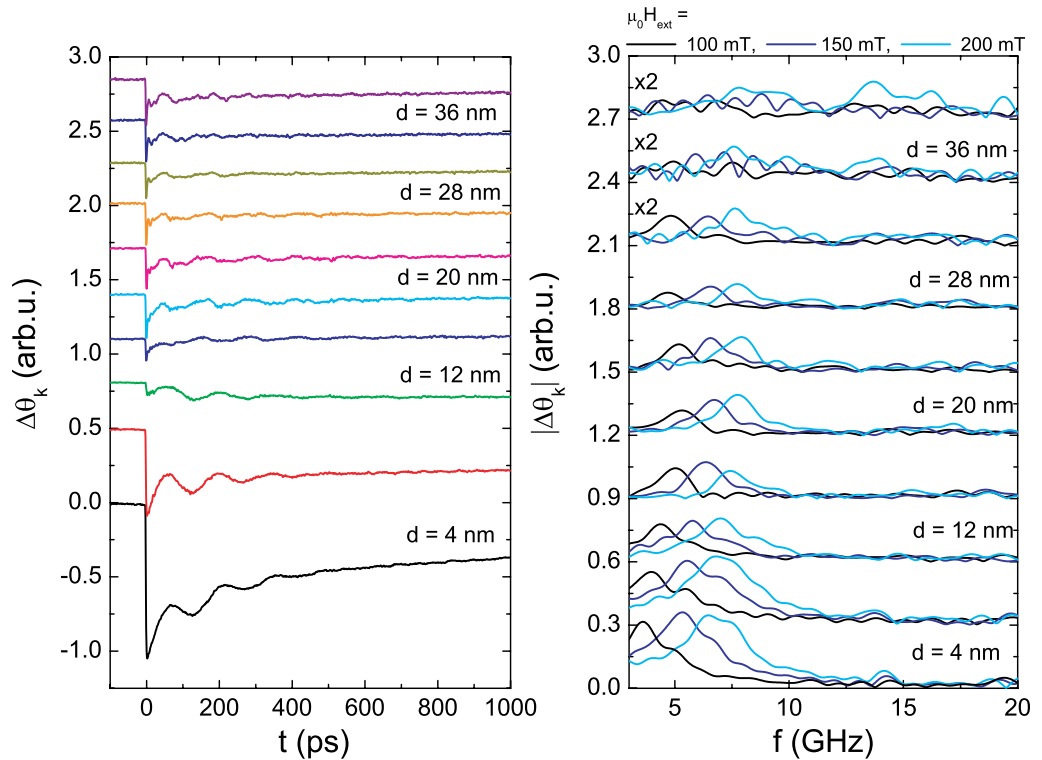
where  $\phi_i \approx 30^\circ$  denotes the angle of incidence and  $n$  and  $v_1$  refractive index and longitudinal sound velocity of the substrate, respectively. As the substrate is thermally oxidized silicon, this effect should blur out after some tens of picoseconds. Taking the material parameters of crystalline quartz  $n \approx 1.5$  for 800 nm and  $v_1 = 5970 \text{ m s}^{-1}$ , the frequency calculates to 21 GHz, which corresponds roughly to the oscillation in the first hundred picoseconds in the time domain. (ii) Furthermore, the strain wave propagating into the substrate and the thermal expansion of the nanodiscs lead to enhanced stress below the metallic structures. Therefore, on the one hand, elastic modes will be excited within single metallic structures and, on the other hand, a standing surface acoustic wave will be formed in the substrate due to the periodicity of the structures.



**Figure 2.** (a) SEM image of the examined structure. Heat transfer into the substrate leads to the formation of standing surface acoustic waves (b) and also to interference effects for reflected probe light (c). (d) Traced reflectivity changes for the different nickel thicknesses.

According to Lin *et al* [15], this elastic mode can be viewed in two limiting cases either as a sole standing surface acoustic wave of the substrate or as separate vibrations of single metallic discs. Recently, Giannetti *et al* studied the coupling of these two systems in detail [16]. In our experiments, the frequencies of the oscillations in the beating patterns illustrated in figure 2(d) can be attributed to this effect with a resulting frequency shift toward smaller values as the thickness increases. This is a clear indicator that these dynamics cannot be viewed in the limiting case of pure standing surface acoustic waves. Therefore, effects stemming from magneto-elastic coupling might be incorporated in the Kerr dynamics for which some indications were found in the work by Comin *et al* [4].

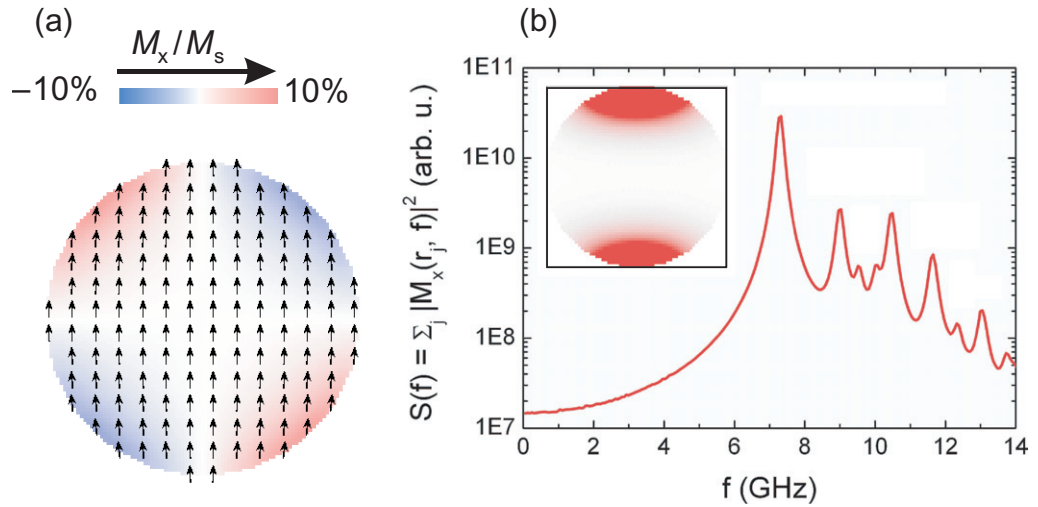
For the separation of the reflectivity changes from the Kerr dynamics, we have to trace two Kerr transients with external magnetic fields of opposite sign. Therefore, we can only detect magnetic modes that change their phase by  $\pi$  under switching the external field which might not be valid for all modes that get excited in this complex system [17]. For excitation of magnetic precessional motion, the external magnetic field is applied in an angle of  $30^\circ$  to the



**Figure 3.** Time-resolved change of the Kerr angle for the different nickel thicknesses at an external of 200 mT and corresponding Fourier transforms for 100, 150 and 200 mT.

sample surface [3]. Via hysteresis measurements in comparison to micromagnetic simulations, we assured that in the examined field range the magnetization of a nanodisc is given by a single domain state for all thicknesses. However, at a nickel thickness above 20 nm, the magnetization would be given by a vortex state for vanishing external field. The Kerr transients for the different thicknesses are depicted in figure 3 for an external field value of 200 mT. Also, Fourier transforms for field values of 100, 150 and 200 mT are given. They are acquired by subtracting the background due to remagnetization, which is accounted for by a single exponential function, and in order to avoid frequency leakage, by application of the Hamming window function.

To understand which magnetic modes are excited we carry out micromagnetic simulations using the Object Oriented Micromagnetic Framework (OOMMF) code developed at NIST [18]. The program approximates the continuum micromagnetic theory for a three-dimensional object by a three-dimensional grid of rectangular cells. Within each cell, a three-dimensional magnetization vector represents the magnetization. The Landau Lifshitz Gilbert equation is solved by integration for discrete time steps using a Runge Kutta algorithm. A single nanodisc is modeled by means of a mesh of cells with a size of  $3 \times 3 \times 2(4) \text{ nm}^3$  for thicknesses below (above) 20 nm. The damping constant is chosen to be quite small ( $\alpha = 0.01$ ). The dynamics are induced by starting the simulation from an initial state that is slightly and statistically perturbed from the ground state which is shown in figure 4(a). The discrete solutions of the Landau Lifshitz Gilbert equation  $\mathbf{M}(\mathbf{r}_j, t_i)$  delivered by OOMMF are Fourier transformed for each cell so that the excited modes can be identified. This way, the lowest frequency mode which happens to be



**Figure 4.** (a) Ground state of a 8 nm thick nickel nanodisc. (b) Frequency spectrum of this system and the end mode corresponding to the peak at lowest frequency.

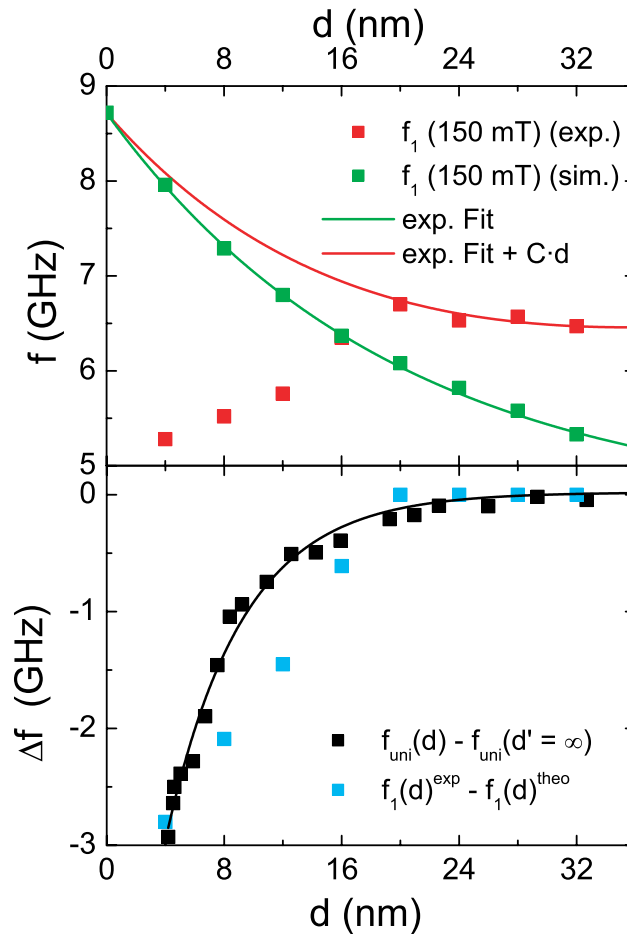
the dominant mode is identified to be of end mode-type, as depicted in figure 4(b) for an 8 nm thick structure and an external field of 150 mT. The spectrum is acquired by summing up the power transforms for all cells:  $S(f) = \sum_j |M_x(\mathbf{r}_j, f)|^2$ . It is assured that these results are not significantly altered in a simulation with vanishing out-of-plane component of the external field; therefore, the findings agree with the work of Zivieri and Stamps [19].

To compare this data with the experimental data, we determine the thickness dependence of this end mode and also extract the frequency of the broad maximum from the spectra found in the experiment; both are plotted versus nickel thickness in the upper graph of figure 5. It turns out that the thickness dependence from the micromagnetic simulations follows an exponential function that coincides for vanishing nickel thickness with the uniform precession frequency of a thin film. This frequency is given by  $\frac{\gamma_0}{2\pi} \sqrt{H_{\text{ext}}(M_s + H_{\text{ext}})}$  ( $\gamma_0 = \mu_0 |\gamma|$ , where  $\gamma$  is the gyromagnetic ratio and  $M_s$  the saturation magnetization; see, e.g. [20]). The experimentally determined frequency dependence looks quite different. However, the dipole interaction between nickel nanodiscs has been completely neglected within the simulations. In a simple model, we incorporate the dipole interaction as an additional external magnetic field that acts on a single disc. This additional field increases with the magnetic moment of a disc linear in the nickel thickness ( $H_{\text{ext}} + C' \cdot d$ ). For external fields below the saturation magnetization, the frequency of magnetic modes close to the uniform one is given mainly by  $\frac{\gamma_0}{2\pi} \sqrt{H_{\text{ext}} M_s}$ . Therefore, in our model, the frequency will alter to first order about an expression linear in nickel thickness  $d$ :

$$f \rightarrow f + \frac{\gamma_0}{2\pi} C \cdot d. \quad (2)$$

By using the exponential fit to the simulated frequencies as the expression for  $f$ , the frequency behavior is adequately explained for thicknesses above 20 nm, where the constant is determined to be  $\mu_0 C = 1.25 \pm 0.05 \text{ mT nm}^{-1}$ . Therefore, we do not only recognize the experimentally dominant mode as an end mode, but also find an quantitative expression for describing the dipole interaction in this nanostructured film. Below 20 nm, the frequencies deviate from the proposed





**Figure 5.** Thickness dependence of the dominant mode from experiment and simulation; while the simulated data follow an exponential function, the experimental data has to be corrected by a term linear in  $d$  to account for dipole interaction (upper graph). At lower thicknesses, a surface effect reduces the precessional frequencies as is also found in a continuous film (lower graph); the line is a guide to the eye only.

model. This drop in frequency, also found in the continuous nickel films, arises from an altered anisotropy as the film thickness is decreased [21]. In the lower graph of figure 5, the frequency deviation, found in a continuous nickel wedge at lower thicknesses, is plotted together with the difference between the experimentally determined frequencies in the nanostructured wedge and the fitting curve in the upper graph. Considering the fact that the demagnetizing factors for a disc are different from the ones for a continuous film, the similarity between the two plots is sufficient to relate both deviations to the same effect.

In conclusion, we have investigated the influence of the nanoscale on laser-induced magnetization dynamics in nickel, both on the timescale of hundreds of femtoseconds as well as in the GHz regime. The fact that there seems to be no influence on the short timescale might help to narrow possible microscopic models for the ultrafast demagnetization. On the other hand, the GHz dynamics of magnetization are significantly altered due to the nanostructuring.

The thickness dependence of the excited magnetic mode allows us to quantitatively study the dipole interaction between single nickel nanodiscs. A possible coupling between magnetic and elastic modes could not be proven or ruled out in this work so far.

## Acknowledgment

Financial support by the Deutsche Forschungsgemeinschaft within the priority program SPP 1133 is gratefully acknowledged.

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