Investigation of ultrafast demagnetization and cubic optical nonlinearity of Ni in the polar geometry

R. Wilks and R. J. Hicken
School of Physics, University of Exeter, Exeter EX4 4QL, United Kingdom

M. Ali and B. J. Hickey
Department of Physics and Astronomy, E.C. Stoner Laboratory, The University of Leeds, Leeds LS2 9JT, United Kingdom

J. D. R Buchanan, A. T. G. Pym, and B. K. Tanner
Department of Physics, University of Durham, Rochester Building, South Road, Durham DH1 3LE, United Kingdom

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Femtosecond optical pump-probe experiments were performed upon a Ni(720 Å)/Si(100) sample in the polar geometry with the pump beam close to normal incidence. A signal due to the ultrafast demagnetization effect was observed when the pump pulse was linearly polarized. When the pump was elliptically polarized, additional peaks were observed at zero time delay, resulting from the specular inverse Faraday effect (SIFE) and the specular optical Kerr effect (SOKE). By comparing measurements made with different pump helicities, the SIFE and SOKE peaks and the demagnetization signal were found to superpose in a linear fashion. From the dependence of the peak height upon the pump polarization, values of $\chi_{xxyy} = (1 - 3i) \times 10^{-10} \, \mathrm{rad \, cm^3 \, erg^{-1}}$ and $\chi_{xyyx} = (-9 + 2i) \times 10^{-12} \, \mathrm{rad \, cm^3 \, erg^{-1}}$ were deduced for the nonvanishing components of the local cubic susceptibility tensor. For applied fields less than the saturation value, the sudden reduction of the thin film demagnetizing field leads to an imbalance of the torques acting upon the magnetization, causing it to precess. © *2004 American Institute of Physics.*
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Excitation by intense sub-picosecond laser pulses can cause a ferromagnet to be demagnetized on time scales shorter than the spin-lattice relaxation time.[1,2] The spin dynamics may be investigated by means of magneto-optical pump-probe measurements. When pumping with elliptically polarized light, an additional transient circular and linear birefringence may be induced by means of the specular inverse Faraday effect (SIFE) or the specular optical Kerr effect (SOKE), respectively. The magnitude of the SIFE and SOKE are characterized by the values of certain components of the local cubic optical susceptibility tensor.[3] Generally in metals, a signal is observed where the pump and probe pulses overlap in time. However in certain semiconductors, pumping with circularly polarized light can create a longer-lived thermal spin population and the subsequent spin relaxation may be observed in the magneto-optical response.[4] It has yet to be shown that the magnetization of a ferromagnetic sample may be modified due to the helicity of the pump pulse. No such effect was observed in a recent experiment upon an in-plane magnetized Ni film.[5] However, the axis of the photon angular momentum and the magnetization axis were separated by a large angle, so that any optically oriented spins would be expected to precess and quickly dephase in the local exchange field. In this paper we investigate the rotation and ellipticity response of a perpendicularly magnetized Ni sample for which the magnetization and the pump photon angular momentum are almost parallel. We compare the rotation and ellipticity response for a linearly and elliptically polarized pump, and investigate the response of a partially magnetized sample. We quantify the contributions of the SIFE and SOKE to the rotation and ellipticity response and calculate the nonvanishing elements of the local cubic optical susceptibility tensor.

Measurements were performed on a Ni film sputtered onto a Si wafer. X-ray diffraction measurements showed that the film contained $724 \pm 3 \, \text{Å}$ of Ni and a $15.5 \pm 3 \, \text{Å}$ overlayer of Ni oxide that formed upon exposure to atmosphere. The Ni had a fcc structure with a lattice constant of 3.52 Å corresponding to that of the bulk phase.[6] The average grain size was determined to be about 80 Å, and the film was found to have a preferred (111) orientation. Optical pump-probe measurements were made with 100 fs pulses with wavelength of 790 nm at a repetition rate of 82 MHz. The pump-induced rotation and ellipticity were measured in reflection as has been described in detail elsewhere.[5,7] In the present study the sample was magnetized by means of an electromagnet with a conical hole in one pole shoe that allowed optical access to the sample. The pump and probe beams had angles of incidence of 4° and 26°, respectively, and average powers of 200 mW and 4 mW, respectively. Both beams were focused to a diameter of about 15 μm on the surface of the sample. The resulting pump fluence of about 1.4 mJ/cm^2 lies within the regime where the magneto-optical response is expected to have a linear dependence upon the pump power.[2] The probe beam was always p polarized. A quarter wave plate was inserted into the initially p-polarized pump beam with its fast axis at an angle ϕ to the
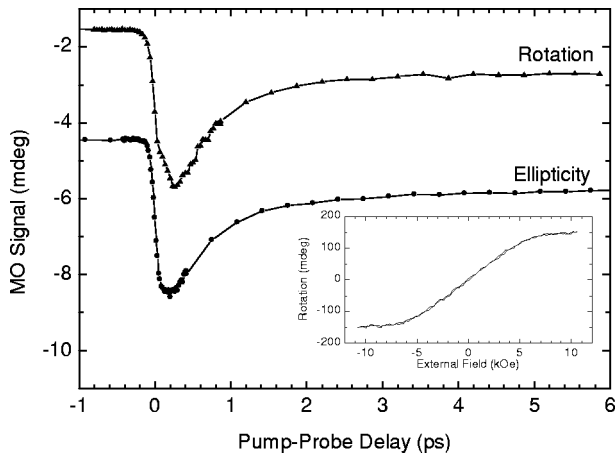
FIG. 1. The rotation (triangles) and ellipticity (circles) response of the Ni sample are shown when magnetized perpendicular to the plane by an applied field of 7.3 kOe. The curves have been offset for clarity. The inset shows the static hysteresis loop obtained with the pump beam blocked.

plane of incidence. Consequently the pump beam was *p* polarized when $\phi = 0°$, $\pm 90°$, circular when $\phi = \pm 45°$, and elliptical at intermediate ϕ values.

Static polar magneto-optical Kerr effect (MOKE) hysteresis loops were obtained with just the probe beam (inset in Fig. 1) and reveal a typical hard axis behavior with a saturation field of about 6 kOe, as expected for a Ni thin film with bulk magnetization. The half loop height of 150 mdeg agrees well with calculations made using published values of optical and magneto-optical constants.[8,9] An ultrafast partial demagnetization of about 3% was observed when the pump was *p* polarized. Figure 1 shows the transient rotation and ellipticity response of the sample in an applied field of 7.3 kOe. The rise of the signal shows a different behavior in rotation and ellipticity within the first few hundred femtoseconds, suggesting the presence of nonmagnetic contributions to the magneto-optical response.[10] The ellipticity signal has a 10–90 % rise time of 130 fs, whereas the rotation signal changes slope and reaches a maximum after 270 fs, in reasonable agreement with values reported previously.[1,2] When the pump was circularly polarized, a sharp peak was observed at zero time delay in addition to the demagnetization signal, as shown in Fig. 2. The peak width corresponds to the cross correlation of pump and probe and the sign changes with that of the pump helicity. The zero delay peak and the demagnetization signal appear to superpose in a linear fashion, the pump helicity having no effect on the longer-lived demagnetization tail.

Figure 3 shows the time-resolved rotation and ellipticity signals obtained at remanence as ϕ was varied from $-90°$ to $+90°$. Due to the shape anisotropy there is negligible remanent static polar MOKE signal and so the peak associated with the pump polarization can be observed in isolation from the demagnetization signal. A small step at zero delay, seen most clearly in the curves obtained with a linear pump, results from breakthrough of the transient reflectivity signal, caused by imperfect alignment of the polarization of the incoming pump and probe beams.[5,7] The elliptically polarized pump gives rise to a peak at zero delay but no long-lived tail.
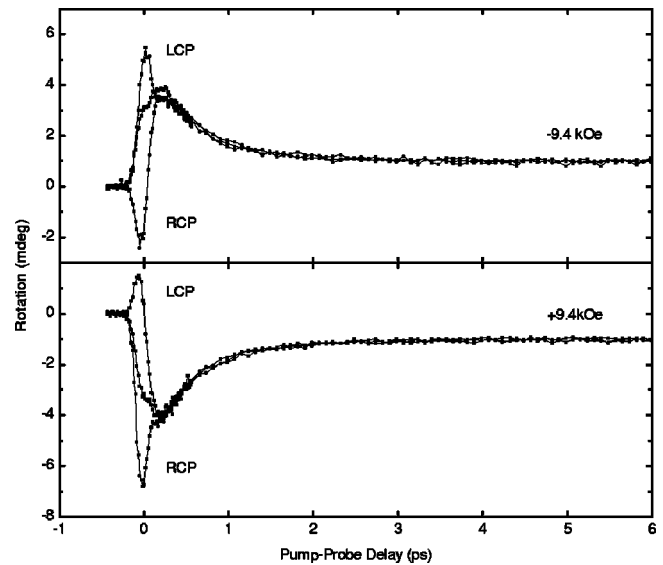

FIG. 2. The rotation signal obtained in positive and negative external fields of 9.4 kOe is shown. Measurements were made with *p* polarized and left and right circularly polarized pump beams.

The dependence of the peak height upon the angle ϕ has been plotted in Fig. 4, and may be described by[5]

$$\begin{pmatrix} \Delta\theta \\ \Delta\eta \end{pmatrix} = \frac{1}{2}\sin 4\phi \begin{pmatrix} \mathrm{Re} \\ \mathrm{Im} \end{pmatrix} F + \sin 2\phi \begin{pmatrix} -\mathrm{Im} \\ \mathrm{Re} \end{pmatrix} G, \qquad (1)$$

where $\Delta\theta$ and $\Delta\eta$ are the peak transient rotation and ellipticity,

$$F = -\frac{32\pi^2 I_{pump}}{c|1+n|^2} \left\{ \frac{\chi_{xxyy} + \chi_{xyyx}}{n(1-n^2)} \right\}, \qquad (2a)$$
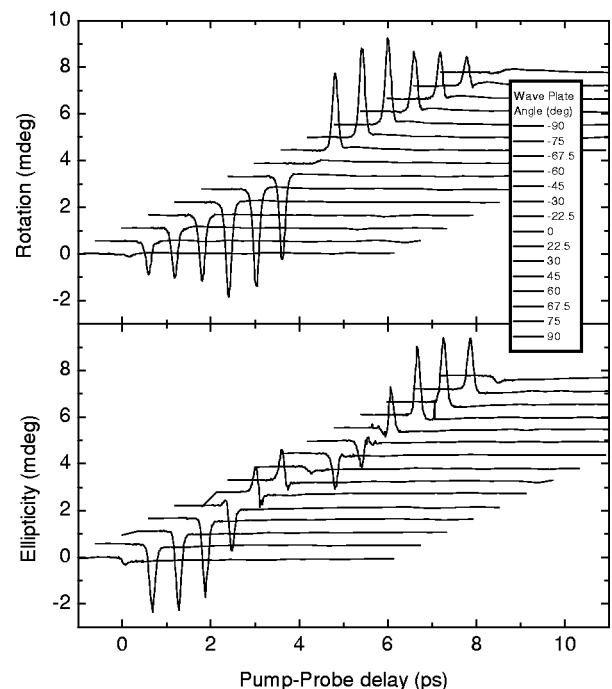

FIG. 3. The time-dependent rotation and ellipticity response in zero external field are shown. The pump beam polarization was varied by rotating the quarter wave plate from $\phi = -90°$ to $+90°$. The curves have been offset horizontally and vertically for clarity.

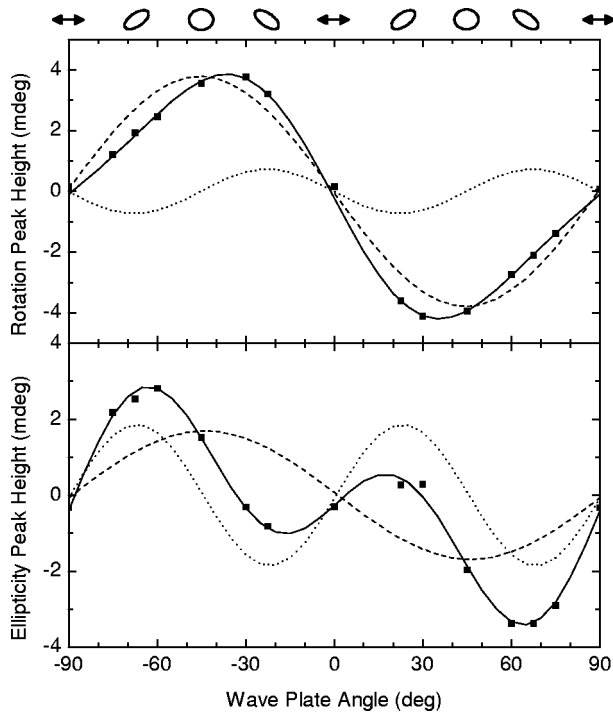J. Appl. Phys., Vol. 95, No. 11, Part 2, 1 June 2004

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FIG. 4. The variation of the height of the peak at zero delay shown in Fig. 3 (squares) with the orientation of the wave plate is shown. The solid curve is a fit to Eq. (1) while the dashed and dotted curves show the contributions from the SIFE and SOKE, respectively.

$$G = -\frac{32\pi^2 I_{pump}}{c|1+n|^2}\left\{\frac{\chi_{xxyy}-\chi_{xyyx}}{n(1-n^2)}\right\}, \qquad (2b)$$

I_{pump} is the pump intensity, c is the speed of light, n is the index of refraction, and χ_{xxyy} and χ_{xyyx} are the incoherent parts of the nonvanishing components of the local cubic susceptibility tensor of an isotropic material. The first and second terms in Eq. (1) correspond to the SOKE and SIFE, respectively. The data in Fig. 4 have been fitted to Eqs. (1) and (2). Taking $n = 2.46 + 4.35i$ (Ref. 11) we obtain $\chi_{xxyy} = (1.3 - i2.7) \times 10^{-10}$ rad cm^3 erg^{-1} and $\chi_{xyyx} = (-9.0 + i1.7) \times 10^{-12}$ rad cm^3 erg^{-1} in good order of magnitude agreement with a value of $|\text{Im}(\chi_{xxyy} - \chi_{xyyx})| = 3 \times 10^{-10}$ esu at 810 nm obtained by Bennett *et al.* from SIFE measurements alone.[12]

For magnetic field values less than the saturation value, the MOKE signal was observed to oscillate with a period of the order of 100 ps, as shown in Fig. 5. The oscillations were found to be independent of the pump polarization, but their phase was observed to shift by 180° as the direction of the applied field was reversed. We believe that the oscillations are associated with precession of the magnetization stimulated by an optically induced reduction of the demagnetizing field.[13] For field values less than the saturation value, the magnetization is canted relative to the surface normal such that the demagnetizing field is equal and opposite to the applied magnetic field. The pump pulse induces a sudden reduction of the magnetization, and hence the demagnetizing field, leaving a net field about which the magnetization begins to precess. When the static field exceeds the saturation
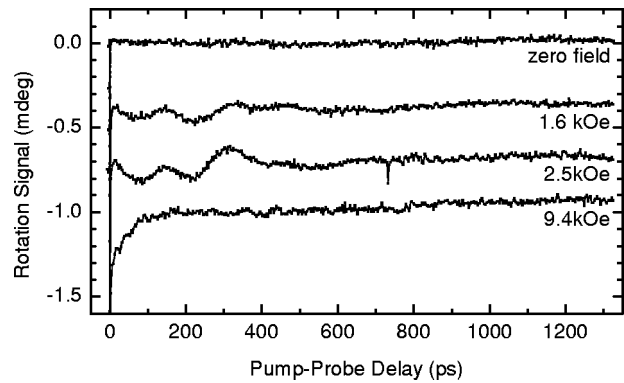

FIG. 5. The time-dependent rotation signals obtained with a *p*-polarized pump at applied fields of 0, 1.6, 2.5, and 9.4 kOe are shown. Precession of the magnetization is observed at fields smaller than the saturation value.

value, the net field is parallel to the static magnetization and exerts no torque to stimulate the precession.

In summary, we have investigated the rotation and ellipticity response of a perpendicularly magnetized Ni sample by means of magneto-optical pump-probe measurements. We observed a difference in the transient rotation and ellipticity induced by a linearly polarized pump, indicating that nonmagnetic signals contribute to the response. When the sample was pumped with elliptically polarized light, a peak at zero delay due to the SIFE and SOKE added to, but did not modify, the demagnetization signal. This indicates that the pump helicity cannot be used to modify the spin polarization on time scales greater than the pulse width. From the variation of the height of the peak at zero delay, the nonvanishing elements of the cubic susceptibility tensor have been calculated. Finally, optical demagnetization was observed to induce magnetization precession when the strength of the static applied field was smaller than the saturation value.

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