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Investigation of ultrafast spin dynamics in a Ni thin film

R. Wilks, N. D. Hughes, and R. J. Hicken[a)]
School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom

Optically induced ultrafast demagnetization has been studied in a polycrystalline nickel thin film by means of a magneto-optical pump-probe technique. The time and magnetic field dependence of the effect have been explored by measuring changes in the reflectivity, and the rotation and ellipticity associated with the linear magneto-optical Kerr effect. We find that, contrary to an earlier report, there is no significant difference in the time dependence of the rotation and ellipticity signals. Furthermore, we observe dynamic hysteresis loops with strange shapes which we believe result from a slow magnetic reorientation induced by average heating effects. These observations emphasize the importance of studying ultrafast demagnetization at saturation. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1450833]

It has recently been observed that ferromagnetic nickel may partially demagnetize on time scales much shorter than the spin-lattice relaxation time[1–4] following excitation by femtosecond optical pulses. The first experimental observations revealed demagnetization times of 260 fs[1] and 280 fs,[2] while a theoretical study suggested an intrinsic time scale for demagnetization of just 10 fs.[5] These reports imply that demagnetization occurs within the time taken for the hot electrons to thermalize. It has been suggested that spin angular momentum is removed from the electron system through the combined action of electron–electron interactions, spin-orbit coupling, and the optical field supplied by the laser pulse.[6] However, the experimental values for the demagnetization time were determined from the magneto-optical response. Two recent experiments[3,4] suggest that the initial magneto-optical signal is influenced by state filling effects, and is not of purely magnetic origin. Indeed it was suggested that the magneto-optical signal only truly reflects the magnetic state of the sample for time scales greater than 2 ps. In one of these experiments, measurements were made with a second harmonic generation technique,[4] while a time resolved linear magneto-optical Kerr effect (MOKE) technique[3] was used in the other. In the latter case a double modulation technique was used to record both the Kerr rotation and ellipticity, which were found to exhibit a qualitatively different time dependence. In this article we use a somewhat simpler detection scheme, involving an optical bridge. We directly measure changes in reflectivity that result from state-filling effects, and explore the time and field dependence of the rotation and ellipticity.

Experiments were performed on a 500 Å polycrystalline Ni film grown on Si(100) by dc magnetron sputtering at an Ar pressure of 5 mTorr. The base pressure of the vacuum system was 2×10^{-7} Torr. Optical measurements were made with a Ti:sapphire laser that produced pulses of 800 nm wavelength at a 82 MHz repetition rate. The beam was split into pump and probe parts with average powers of 280 and 12 mW, respectively. The p-polarized pump and probe beams were incident on the sample at angles of 25° and 47°, respectively. Both beams were expanded by a factor of 10, focused to a 15 μm spot diameter, and carefully overlapped while being viewed with a charge coupled device camera that gave a $\times600$ on-screen magnification factor. The width of the pump and probe pulses was about 120 fs at the sample. The experimental geometry is shown in Fig. 1. A variable delay line in the pump beam path allowed the delay between pump and probe to be adjusted through a range of 2 ns with a minimum step size of 1.67 fs. The position of the retroreflector in the delay line was moved through a few wavelengths at high frequency to remove coherence oscillations around the zero delay position that result from interference of the probe beam with diffusely scattered pump light. The reflected probe beam was directed into an optical bridge detector that allowed the reflectivity (sum signal) and rotation (difference signal) to be measured. By introducing an additional quarter wave plate in front of the detector, the ellipticity could be measured instead of the rotation.

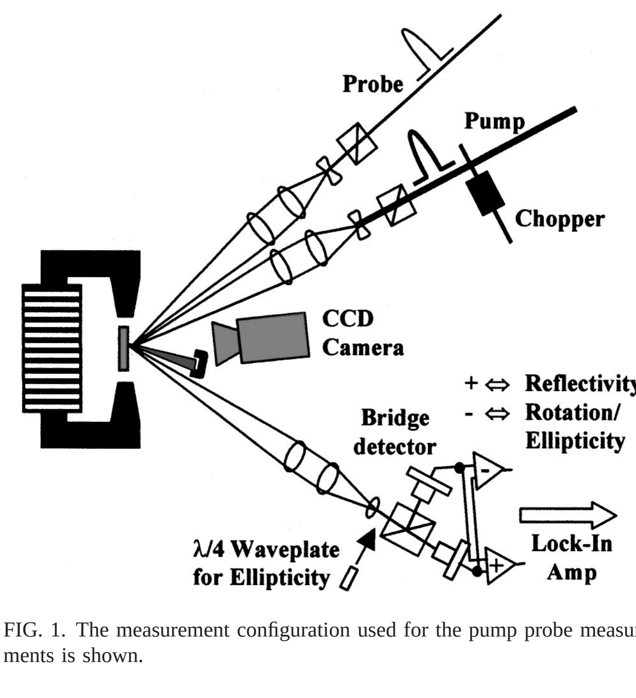

FIG. 1. The measurement configuration used for the pump probe measurements is shown.

[a)]Electronic mail: r.j.hicken@exeter.ac.uk

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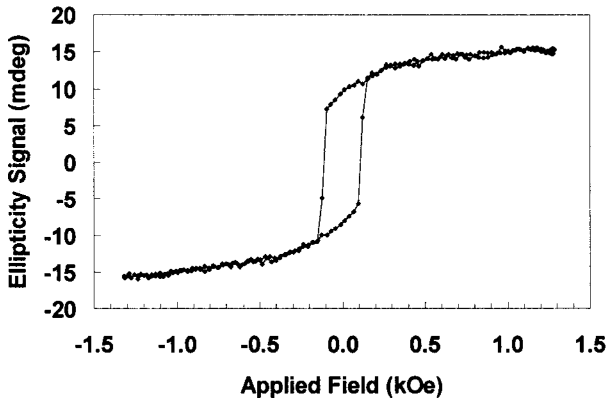

FIG. 2. A static magnetization loop obtained from a measurement of the Kerr ellipticity in the absence of the pump is shown.

A number of different measurement procedures were used. First, static MOKE loops were obtained with the pump beam blocked. Then with the pump unblocked, measurements were made either as a function of time delay in the presence of a constant magnetic field, or as the field was swept while keeping the time delay fixed. Static MOKE loops recorded with the pump beam unblocked revealed that the loop height was only slightly reduced relative to those without the pump present. The pump beam was then chopped at close to 2 kHz and the detector output was measured with a lock-in amplifier to show the small demagnetization more clearly. The output of the lock-in represents the change in the sample response induced by the pump.

A static Kerr ellipticity loop is shown in Fig. 2. It was found that the static ellipticity loop height is about 30 mdeg for our angle of incidence of 47°, while the magnitude of the rotation loop height is an order of magnitude smaller. This is consistent with calculations that we have made using optical and magneto-optical constants from the literature.[7,8]

The time resolved Kerr rotation and ellipticity signals are shown in Figs. 3(a) and 3(b), respectively. The time dependence of the signal was measured in static fields of ± 860 Oe, that the static ellipticity loop showed to be sufficient to saturate the sample. The zero time delay position was determined from a cross-correlation measurement performed with a circularly polarized pump beam. Reversing the orientation of the static magnetization should cause the sign of the time resolved Kerr signal resulting from the demagnetization to change sign as we indeed observe. However, an asymmetry can be observed between the two magnetization directions. We believe this is due to a breakthrough of the reflectivity in the difference signal. When the optical bridge is close to the balanced condition, the difference signal S varies as $S \propto (I+\delta I)(\theta_{\text{off}}+\delta\theta)$, where I is the intensity of the probe without the pump, θ_{off} is the angle by which the detector is misaligned from the balanced position, and δI and $\delta\theta$ are the changes in intensity and rotation induced by the pump beam. To first order, the lock-in signal is proportional to $\delta I \theta_{\text{off}} + I \delta\theta$. Therefore, if $\theta_{\text{off}} \neq 0$, changes in the reflected probe intensity δI may contribute to the signal. These changes are independent of the orientation of the magnetization. Therefore, we may take half the difference of the signals obtained for the two magnetization directions to obtain the true magneto-optical signal, as shown in Fig. 3. The maximum time dependent ellipticity can be compared with the static ellipticity and corresponds to a 7% demagnetization. The magnitude of the change in the magneto-optical signal is expected to be unchanged when the orientation of the magnetization is reversed. Therefore, we may obtain the nonmagnetic background signal by averaging

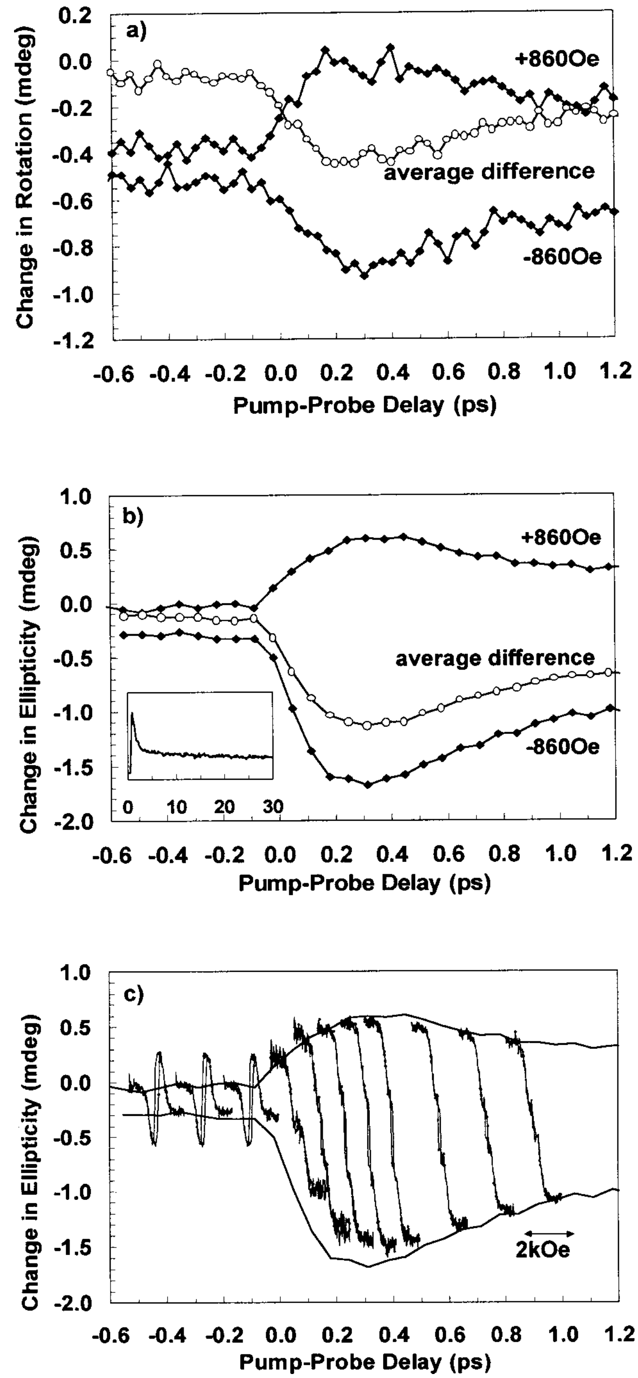

FIG. 3. The time and field dependence of the change in (a) the rotation, and (b) the ellipticity signals is shown. The diamonds show the raw time-resolved measurements at static fields of ± 860 Oe. The open circles represent half the difference of the signals measured at positive and negative field. The inset in (b) shows the ellipticity signal on a longer time scale. (c) shows field-dependent dynamic ellipticity loops at different time delays. The field scale is indicated.

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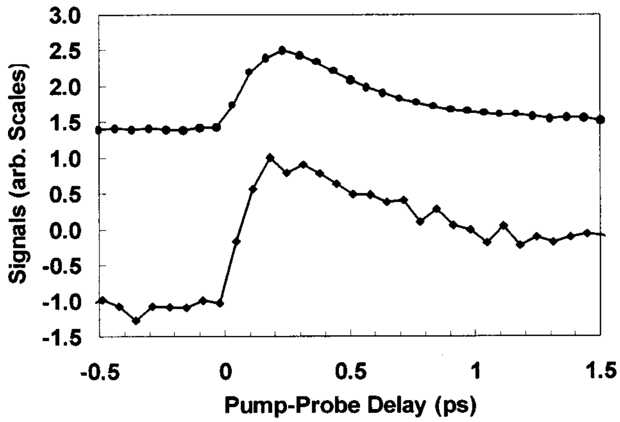

FIG. 4. The diamonds show the average of the raw ellipticity scans obtained in positive and negative field, and represent the nonmagnetic contribution in the measured magneto-optical signal. The circles show the time dependence of the pump induced reflectivity signal. No cross-correlation measurement was made in the latter case and the zero delay has been set to match that of the diamonds. Although the vertical scale is arbitrary and different for the two curves, the curves are seen to have similar shape.

the magneto-optical signals obtained for the two field directions so that the magnetic contributions cancel. The background signal from the ellipticity and the time dependent reflectivity are plotted together in Fig. 4. The two curves are seen to have similar form, suggesting that our analysis of the detector output is correct.

Figure 3(c) shows a series of dynamic hysteresis loops taken at different constant time delays. These loops show the change in the Kerr signal induced by the pump beam rather than the full Kerr signal that appears in a static hysteresis loop. Despite the strange loop shapes the signal at saturation agrees well with that observed in the time scans. We discuss the possible origin of the loop shape in the next section.

Most existing models of the demagnetization process have assigned different temperatures to the lattice and various electron subsystems, and then described the heat flow between the subsystems with rate equations.[1,2] Reflectivity changes in noble metals have been modeled by taking account of nonthermal electrons and allowing for heat transport into the interior of the film.[9,10] This model is not easily extended to Ni due to the overlap of the *sp* and *d* bands at the Fermi level. However, the reflectivity curve in Fig. 4 is of similar shape to others reported previously.[11]

From Figs. 3(a) and 3(b) we see that, after accounting for reflectivity breakthrough, the ellipticity and rotation time scans both rise in a time of about 300 fs. Contrary to earlier reports[3] we see no significant difference in the time dependence of the rotation and ellipticity scans. This suggests that the thermalization time of 300 fs can be still considered as an upper limit for the demagnetization time. The strange shape of the dynamic loops in Fig. 3(c) might result from a number of mechanisms. These include the ultrafast demagnetization, nano- or picosecond magnetic reorientation due to precession or domain wall motion, or a slower reorientation of the magnetization associated with the increase in average temperature when the chopper blade is open. We do not expect the ultrafast demagnetization to depend upon the small applied field since in our experiment the system is never close to a phase transition to the paramagnetic state. The dynamic loops at negative delay are at a positive time delay of about 12 ns relative to the previous pump pulse. Any precessional motion will be damped out on time scales much shorter than 12 ns. Since the loop shape does not depend upon the position of the focused spots upon the sample, a domain wall process seems unlikely. We therefore suggest that the inverted loops seen at negative time delay result from a reorientation of the macroscopic magnetization associated with sample heating on time scales comparable to the period of the chopper. This heating also causes the 1% background demagnetization observed in the saturated state at negative delays. The dynamic hysteresis loops show that the reduction of the spontaneous magnetization associated with ultrafast demagnetization can only be reliably measured when the sample is saturated.

In summary we have measured changes in reflectivity, Kerr rotation, and Kerr ellipticity as a function of both time and applied field in a polycrystalline nickel film. The rotation and ellipticity signals are found to have the same time dependence. The field dependent measurements suggest the presence of a much slower magnetic reorientation and emphasize the importance of studying the ultrafast demagnetization effect at saturation.

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