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Femtosecond spin dynamics of epitaxial Cu(111)/Ni/Cu wedges

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The femtosecond spin dynamics in ferromagnetic metals is studied using time-resolved magneto-optics on an epitaxial Cu(111)/Ni/Cu wedge. A novel and sensitive double modulation scheme is introduced. The induced Kerr rotation and ellipticity are separately measured, and display a contrasting behavior during the first 500 fs after excitation. This proves the absence of a direct relation between the magneto-optical signal and the magnetization. A systematic study of the spin dynamics along the nickel wedge shows a surprisingly small influence of interface scattering, magnetic anisotropy, and morphology on the dynamics. © 2000 American Institute of Physics. [S0021-8979(00)64408-0]

A number of recent papers report on an ultrafast (~ 100 fs) change in the magneto-optical (MO) contrast of ferromagnetic materials after excitation by a short heating laser pulse.^{1–6} Several authors have related these MO effects to an almost instantaneous demagnetization, on a time scale faster than the electron–phonon equilibration time. Little is still known about the physical origin of the demagnetization, and even the presence of a direct relation between the observed MO effects and the magnetization is heavily debated.

In this article, we introduce a new configuration to investigate the possible existence of such a direct relation during the initial time after excitation. We argue that any difference between behavior of the induced Kerr rotation and ellipticity, which are measured separately, provides evidence for MO contributions *not* scaling with the magnetization. We studied the time-resolved MO Kerr effect (TRMOKE) of well defined and epitaxially grown Cu(111)/Ni/Cu wedges, in order to access the microscopic origin of the spin dynamics. The Cu/Ni/Cu system displays a twofold orientational phase transition.⁷ At intermediate thickness an out-of-plane magnetic anisotropy is driven by the misfit strain. At higher thicknesses, the strain is released by the appearance of misfit dislocations, causing a return to the in-plane configuration. Thus, the influence of the magnetic anisotropy and the morphology (dislocations) on the dynamics can be studied by measuring the time-resolved MO as a function of the position on the nickel wedge. Also a possible influence of interface scattering may be found this way, due to its decreasing role at increasing nickel thickness.

A Ni wedge with a thickness ranging from 0 to 8 nm, capped by 2 nm of Cu, was grown on Cu(111) by means of molecular beam epitaxy in a system with a base pressure $< 5 \times 10^{-11}$ mbar. The single crystalline substrate was treated by cycles of Ar⁺ sputtering and thermal annealing, and the surface structure was checked by low-energy elec-

tron diffraction (LEED) at various stages during preparation. The magnetic behavior was verified *ex situ* at room temperature with standard MOKE equipment. An abrupt change from in-plane to out-of-plane magnetization was found at 0.9 nm, and a somewhat more diffuse return to in-plane at 3–5 nm, in reasonable agreement with previous reports.⁷

TRMOKE experiments were performed *ex situ* at room temperature in the configuration of Fig. 1. The 70 fs laser pulses (repetition rate 82 MHz, $\hbar\omega \sim 1.7$ eV) are split in pump and probe pulses at an intensity ratio 5:1, and focused down to overlapping spots of ~ 10 μ m diameter on the sample, with a pump fluence of 0.6 mJ/cm² and an angle of incidence (probe) of 45°. The pump and probe beam are modulated by a mechanical chopper (frequency $F_{\text{pump}} \approx 60$ Hz) and a photoelastic modulator (PEM, $F_{\text{PEM}} = 50$ kHz), respectively. The PEM, a polarizer, and an analyzer are mounted in the probe beam at angles of resp. α_{PEM} , $\alpha_{\text{pol}} = \alpha_{\text{PEM}} + 45^\circ$, and α_{anal} . The dc intensity (V_{dc}), and signals at 50 kHz (V_{1f}) and 100 kHz (V_{2f}) are measured by means of a Si photodiode, and two lock-in amplifiers in series. The analogue output from the first lock-in amplifier, synchronized with the PEM, and set to a time constant $1/F_{\text{PEM}} \ll \tau = 3$ ms $\ll 1/F_{\text{pump}}$, is used as the input for a second lock-in amplifier, with $\tau = 300$ ms $\gg 1/F_{\text{pump}}$ and syn-

FIG. 1. “Double modulation” TRMOKE setup.

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chronized with the chopper. This scheme assures the neglect of any but the pump-induced signals. In order to reduce interference effects between specularly reflected probe and scattered pump light, leading to apparent noise around zero delay, the retroreflector in the delay line is mounted on a piezoelectric transducer and translated at 50 Hz over several wavelengths. A simple electromagnet ($H \approx 200$ kA/m) is used to switch the magnetization between opposite remanent values ($M_{\pm}$).

We define a complex polarization rotation $\psi = \psi' + i\psi'' = \psi_0 + \Delta\psi$, where “ Δ ” indicates pump-induced contributions, and uses similar definitions for the ellipsometric ratio of p and s reflection coefficients, $\rho = R_p/R_s$. We distinguish two configurations. In the “ 0° configuration” we choose $\alpha_{\text{PEM}} = \alpha_{\text{anal}} = 0^\circ$, and find

$$V_{1f}/V_{\text{dc}} = C_1 \psi'', \quad (1)$$

$$V_{2f}/V_{\text{dc}} = C_2 \psi', \quad (2)$$

where $C_n = J_n(A_0)$, J_n is the n -th order Bessel function and A_0 is the retardation amplitude of the PEM. Both signals are independent of ρ_0 and $\Delta\rho$, i.e., nonmagnetic contributions are absent. We emphasize that such an ideal case of 100% magnetic contrast is generally not met in previous experiments.^{1,2,5,6} In the “ 45° configuration” ($\alpha_{\text{PEM}} = 45^\circ$) the $1f$ signal equals that of Eq. (1) for $\alpha_{\text{anal}} = 0$. For the $2f$ measurement, the analyzer has to be set to $\alpha_{\text{anal}} = \alpha_0 = (1/2)\cos[(|\rho|^2 - 1)/(|\rho|^2 + 1)]$ in order to adjust the pump-independent signal to zero. At this setting we find

$$\frac{V_{2f}}{V_{\text{dc}}} = C_2 \left(-\frac{\rho' \Delta\rho' + \rho'' \Delta\rho''}{|\rho|^2} + \frac{1 + \rho'}{|\rho|} \psi' + \frac{\rho''}{|\rho|} \psi'' \right). \quad (3)$$

The appearance of the first term, representing the nonmagnetic pump-induced response, allows a comparison with the electron (nonmagnetic) dynamics.

In the simple analysis of many recent papers, the MO signal is assumed to scale linearly with the magnetization, even at the shortest time scales.⁶ If this were true, it would be valid both for the rotation and ellipticity, and the equality

$$\Delta\psi'/\psi'_0 = \Delta\psi''/\psi''_0 \quad (4)$$

should be fulfilled independent of delay time. Any deviation would be a sign of “nonmagnetic” contributions.

A typical result for the different configurations and a Ni thickness in the out-of-plane region is displayed in Fig. 2. We verified that in our relatively low excitation studies the induced MO signals are roughly proportional with the laser intensity. Panels (a) and (b) show V_{1f} in the 0° and 45° configuration for opposite magnetization directions. All data are scaled to the static magnetic contrast. Also displayed are the pump-probe autocorrelation trace, and the sum and difference of M_+ and M_- signals. In agreement with the analysis presented above [Eq. (1)], panel (a) and (b) are almost identical, and the “sum” curve vanishes (except for panel (b), where a small constant offset is probably of thermal origin, and a small peak around zero delay is assigned to degenerate four-wave mixing). The difference curve represents the magnetic contrast, $\Delta\psi''/\psi''_0$. A rapid (almost instantaneous) rise is followed by an almost exponential decay at

FIG. 2. TRMOKE for a Ni thickness of 2.7 nm in the out-of-plane region, showing $1f$ (top) and $2f$ (bottom) signals in the 0° and 45° configuration at M_+ (dotted) and M_- (drawn line), as well as the sum (crosses) and difference (circles) of them, all normalized to the static magnetic contrast. The pump-probe autocorrelation is represented by the thick line.

time constant $\tau_E \approx 300$ fs to an equilibrium value different from the level found at negative delays. The magnetic contrast resolves no additional features for delay times up to 100 ps (not shown).

Panels (c) and (d) show the corresponding V_{2f} data. At 0° , the difference curve represents the MO ellipticity. Comparison of (a) and (c) show that after 2 ps Eq. (4) holds, $\Delta\psi'/\psi'_0 \approx \Delta\psi''/\psi''_0 \approx 0.7\%$. At short delay times, however, a different profile is observed which will be discussed below. The nonmagnetic information is obtained from the sum trace in panel (d). The enhanced noise around zero delay is due to the interference artifact. Also, it should be realized that for the Cu/Ni/Cu system and $\hbar\omega = 1.7$ eV we have $\psi'_0:\psi''_0 \approx 1:7$, which means that rotation data are generally more noisy.

A detailed comparison of rotation and ellipticity is shown in Fig. 3(a). The clear difference observed during the first picosecond demonstrates that *during this initial stage*

FIG. 3. (a) TRMOKE at $w=2.7$ nm, comparison of $\Delta\psi'/\psi'_0$ (open symbols), $\Delta\psi''/\psi''_0$ (filled), and $\Delta\psi'/\psi'_0 - \Delta\psi''/\psi''_0$ (line). (b) $\Delta\psi''/\psi''_0$ at several Ni thicknesses.

there is no direct relation between the MO response and the magnetic moment of the nickel film. A collection of data at different positions on the wedge is also depicted in Fig. 3(b). So far, we were not able to detect any significant thickness dependence within the experimental accuracy.

Finally, we turn to a preliminary interpretation of the observed features. The almost exponential relaxation of the signal between 0.4 and 2 ps, and the fact that the time constant for the magnetic channel resembles that of the nonmagnetic one, indicate that both effects are related to the energy relaxation of the electron system with the lattice, driven by the electron–phonon coupling.

As reported in several articles, we observe a rapid loss of MO contrast after excitation. In agreement with Ref. 6, the rise time is basically limited by the pulse width. However, the clear difference of rotation and ellipticity means that we cannot assign this MO phenomenon to an (almost) instantaneous demagnetization. Although we cannot exclude that a part of the sharp rise at $t=0$ is related to magnetic phenomena, the observed deviation from Eq. (4) has to be formulated in terms of (possibly magnetization-conserving) changes in the occupied density of states during the rapid thermalization of the hot electron gas. Recently, we were able to experimentally identify these “transfer-of-spectral-weight” effects in spectroscopic studies on magnetic semiconductors.⁸ Now we also demonstrated the relevance for metallic systems. Nevertheless, the observation that after 1–2 ps Eq. (4) is fulfilled, and the absence of additional features on the time scale up to 100 ps, are suggestive for a full equilibration of electrons, phonons, and spins. Thus, demagnetization might well take place within 1–2 ps.

As to the thickness dependence, Fig. 3(b) displayed no dependence on the position whatsoever. From this observation a number of conclusions can be drawn. First, our method provides a reliable and very reproducible measure of

the spin and electron dynamics. Second, the observed features are intrinsic to the dynamics of the Ni layer and not due to heat transfer between film and substrate. Finally, it provides some additional support for the MO—rather than a magnetic interpretation.

In summary, we provided clear evidence for nonmagnetic contributions to the femtosecond MO response in ferromagnetic nickel from a difference between pump-induced rotation and ellipticity during the first 0.4 ps. Still our results make likely that the spin system has come into equilibrium with the electrons and the lattice within 1–2 ps. It will be one of the challenges of the field to really access the magnetization dynamics at these time scales, and to isolate the underlying spin scattering mechanisms. A preliminary attempt in this direction displayed a surprisingly weak dependence of the observed dynamics on the Ni film thickness.

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