# Spin dynamics in $CoPt_3$ alloy films: A magnetic phase transition in the femtosecond time scale

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We have measured the magnetization dynamics of an epitaxial $CoPt_3$ alloy film using a magneto-optical pump-probe Kerr setup with 120 fs laser pulses. At high pump fluence, the film is driven from the ferro- to the paramagnetic state with a characteristic time comparable to the pulse duration. The associated demagnetization occurs within the thermalization time of the electrons to a Fermi distribution. These results demonstrate the feasibility of ultrafast magneto-optical devices. [S0163-1829(98)00342-7]

The development of pump and probe optical techniques using ultrashort laser pulses has recently led to a breakthrough in the study of time-dependent magnetic phenomena. By using picosecond pulses, it has been shown that the thermalization of the spin dynamics to the lattice is about ~100 ps in a Gd thin film.[1] Recently,[2] we have shown that the absorption of a 60 fs duration laser pulse in a thin ferromagnetic Ni film leads to a fast magnetization decrease that can be measured using longitudinal magneto-optical Kerr effect (MOKE). From the comparison between magnetic and optical transients, it was shown that magnetization changes could be obtained in the subpicosecond time scale, before heating the lattice. A phenomenological model has been used to explain this experimental result. It takes into account energy exchange between three thermodynamic reservoirs: the electrons, the spins, and the lattice. Subsequently, experiments have been performed using a pump-probe second-harmonic generation apparatus,[3] where the transient properties of the surface of a bulk nickel sample have been measured. Ultrafast magnetization dynamics could be observed on a time scale slightly shorter than the electronic thermalization time (280 fs), the minimum of demagnetization being reached at 230 fs. These experiments require a microscopic theory for the description of magnetic interactions occurring on the femtosecond time scale. One can expect that various parameters of the ferromagnetic material such as the exchange energy, the spin-orbit coupling or the Coulomb interaction between electrons influence the spin dynamics.[4]

The preceding results, which suggest that ultrafast magneto-optical writing is feasible, might have potential applications in data storage and processing if the ultrashort laser pulse can induce a complete demagnetization. So far, only demagnetization ratios of about 50% could be obtained in the subpicosecond time scale.[2,3] Intuitively, if one assumes a temperature model, the amount of energy necessary to cross the phase transition requires overcoming the thermal inertia related to the large magnetic heat capacity at the Curie point. This process may be difficult to achieve without sample damages. These remarks prompted us to conduct additional experiments on $CoPt_3$ alloy films that have been recently studied in the context of magneto-optical recording due to their enhanced perpendicular anisotropy and Kerr rotation as well as a low Curie temperature.[5] In the present paper, we focus on the magnetization dynamics that occurs within the first hundreds of femtoseconds. On that time scale, the electrons are thermalizing to a Fermi distribution. We demonstrate that with appropriate laser intensities, the complete demagnetization of the film occurs with a characteristic time equal to or less than the pulse duration (120 fs full width at half maximum). To our knowledge, this is the fastest magnetic phase transition ever observed.

The experimental configuration used to perform the time-resolved MOKE measurements was described in Ref. 2. In the present work, we used 120 fs pulses coming from an 800 nm tunable Ti sapphire cavity amplified at 5 kHz by a YLF laser. The pump and probe beams were focused onto the sample with respective spot diameters of ~100 and 50 $\mu$m. The intensity ratio between the pump and the probe beams was 20:1. The polarization of the pump beam was either *s*, *p*, or circular, whereas the polarization of the probe beam was *p*. In the experiments reported below, the Kerr signal was measured in the polar geometry (magnetic field in the range $-3.2/+3.2$ kOe applied perpendicularly to the sample surface) at an angle of incidence of 50°. Two types of measurements, averaged over about 1000 pulses, have been performed: (1) full hysteresis loops have been measured by sweeping the magnetic field and recording the signal at a given temporal delay between the pump and probe beams (see Ref. 2). (2) Alternatively, the Kerr signal has been measured continuously as a function of the pump-probe delay in a fixed magnetic field. The results have been obtained with an epitaxial $Co_{0.25}Pt_{0.75}$ alloy film, grown at 690 K on a 16 nm Ru (0001) buffer layer deposited on a mica substrate. The $CoPt_3$ layer has a thickness of 48.4 nm. The growth conditions have been chosen to optimize the perpendicular magnetic anisotropy. The magnetic and structural characterizations of the sample are reported elsewhere.[6] The temperature dependence of the magnetization has been measured with a conventional Kerr magnetometer. Due to the chemical disorder intrinsic to this type of sample, the magnetic transition was found quite broad in agreement with previous results[7] and the remanent magnetization vanishes at 635 K.

Time-resolved polar hysteresis loops measured with 2 $\mu$J energy per pump pulse are shown in Fig. 1. The loop obtained without pump beam (squares) is typical of materials with high perpendicular magnetic anisotropy with a coercivity of ~1 kOe. When the delay between the pump and probe

 

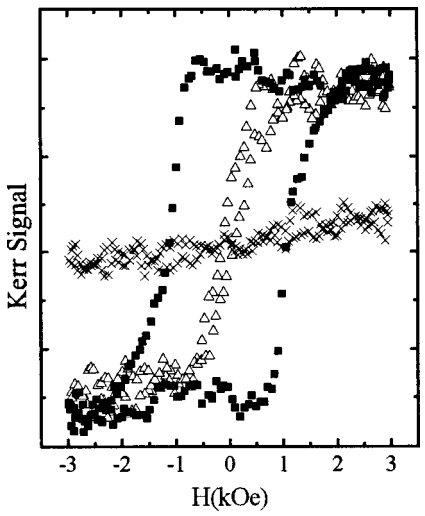


FIG. 1. Polar Kerr loops of a $CoPt_3$ alloy film at selected pump-probe delays. The pump beam of 2 $\mu$J per pulse is $p$ polarized. Squares: measured with the probe beam only, triangles: probe pulses arriving 1 ps before the pump, cross: probe beam delayed by 630 fs.

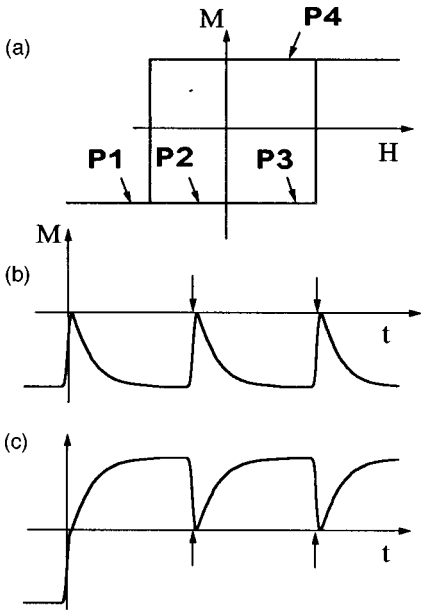


FIG. 2. Qualitative description of the spin dynamics induced by a sequence of pump pulses for different values of the applied magnetic field. (a) Static hysteresis loop of the sample, defining working points $P1-P4$. (b) Dynamics starting from point $P1$ or $P2$ showing the transient demagnetization. (c) Dynamics starting from $P3$ showing the magnetization reversal after the demagnetization induced by the first pump pulse. The arrows in the time abscissa in (b) and (c) denote the occurrence of the pump pulses.

pulses is 630 fs, the field dependence of the magnetization does not show any hysteresis (crosses) but rather a weak linear increase. This behavior is typical of the paramagnetic phase. When the probe precedes the pump,[8] the hysteresis loop (triangles) is very different from what is observed without the pump: the coercivity nearly vanishes and the loop has a *s*-like shape with a saturation field of ~500 Oe. As explained below, this behavior results from the fact that the paramagnetic state of the sample is reached during the dynamics. Depending on the magnetic history of the sample, a complete transient demagnetization may lead for long delays either to a return to the initial spin direction (transient demagnetization) or to a spin reversal (magnetic switch). Two situations corresponding to different working points of applied magnetic field must be distinguished as described in Fig. 2. The first one corresponds to the points $P_1$ or $P_2$ (set by first saturating the sample in a large negative field and then increasing the field). In this case, the heating by the femtosecond pump pulse with sufficient energy leads to the demagnetized state in less than 600 fs, as demonstrated by Fig. 1. The excited part of the sample then cools down in a negative field so that the initial magnetization is restored after some time. The occurrence of subsequent pulses (arrows on Fig. 2) produces a repetition of this dynamics [Fig. 2(b)]. A transient demagnetization is therefore produced in this configuration for each pump pulse with a return to the initial states associated to $P1$ or $P2$. The second situation corresponds to the point $P3$ when a positive magnetic field, smaller than the coercitive field, is applied to the sample (also set by first saturating in a negative field and then increasing the field). In this case the sample is demagnetized by the *first* pump pulse but then cools down in a positive magnetic field, so that the magnetization is *reversed* to $P4$. Additional pulses lead to successive demagnetization and recovery to $P4$. As a result of this process, the energy provided by the pump beam allows to switch after some delay (this is equivalent to what is observed at negative delay[8]) from the metastable magnetic configuration ($P3$) to the stable one ($P4$). However, since we are using stroboscopic means with averaging over a large number of pulses (a few thousands) in order to measure the dynamics from the pump and probe experiment, we are unable to measure directly the dynamics of the magnetization switch that occurs once. This description explains the vanishing hysteresis observed in Fig. 1 (triangles). Nevertheless, the magnetization curve displays a residual hysteresis which we attribute to domains nucleation and motion during the cooling in a weak magnetic field.

In order to get a quantitative piece of information about the spin dynamics, the Kerr signal was measured as a function of pump-probe delay $\Delta t$. As seen in Fig. 3 with a pump energy of 2 $\mu$J, the corresponding dynamics occurs within the first hundreds of femtoseconds, the maximum of demagnetization being reached for a pump probe delay of ~500 fs. For these measurements, the sample was maintained in a magnetic field $H_0 = \pm 1.5$ kOe after saturation in a field of $\pm 3.2$ kOe and the Kerr signal $I_K$ was recorded as a function of $\Delta t$. The quantity reported in Fig. 3 is the normalized magnetization defined as $M(\Delta t)/Ms = \Delta I_K(\Delta t)/\Delta I_{K_0}$. This convention allows us to compare the time-dependent Kerr signal $\Delta I_K(\Delta t) = I_K(H_0, \Delta t) - I_K(-H_0, \Delta t)$ to the static one $\Delta I_{K_0} = I_K(H_0, \Delta t < 0) - I_K(-H_0, \Delta t < 0)$. Such data are reported in Fig. 3 for different pulse energies in the range 0.4–2 $\mu$J. They clearly show that the demagnetization dynamics, qualitatively discussed previously on the basis of the hysteresis curves, is achieved ~500 fs after the pump pulse. The remaining signal $M/Ms = 0.1 \pm 0.05$ observed for $\Delta t > 500$ fs at the highest fluence is due to the polarization of

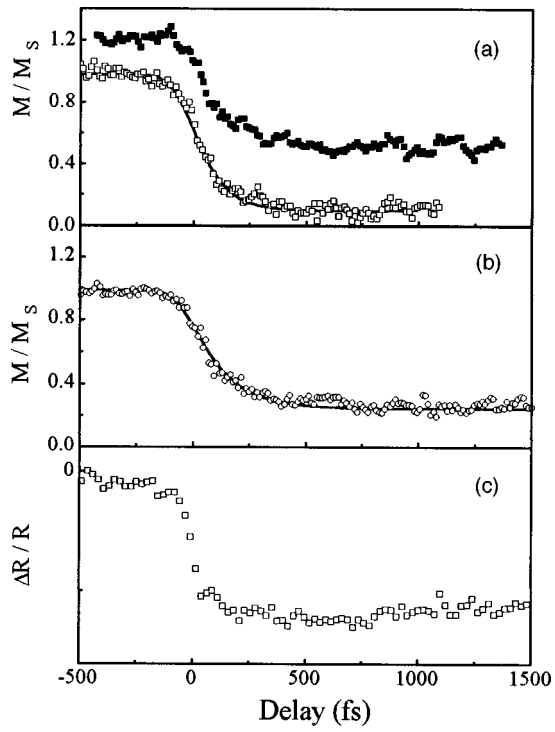


FIG. 3. Time resolved polar Kerr and reflectivity measurement. (a) Time dependence of the normalized Kerr signal for 0.4 $\mu$J (■) and 2 $\mu$J (□) *p*-polarized pump pulses; the data obtained at 0.4 $\mu$J are offset for clarity. (b) 1.5 $\mu$J circularly polarized pump pulses. (c) Transient reflectivity after excitation by 1.6 $\mu$J *p*-polarized pump pulses. The lines are fits assuming exponential magnetization decays and Gaussian pulse temporal profile 120 fs FWHM (see text).

the disordered Co moments in the applied magnetic field (as expected for a paramagnet). This agrees with the time-resolved measurements presented in Fig. 1, and with the static Kerr measurement performed at elevated temperature.

In order to evaluate the characteristic time for the demagnetization, we propose to analyze the above results assuming an exponential decay of the magnetization after excitation by the pump beam $M(\Delta t)/Ms = 1 - \exp(-\Delta t/\tau)$. The response time $\tau$ is deduced by fitting the Kerr signal with a convolution between $M(\Delta t)$ and the pump and probe intensity profiles. A pulse width of ~120 fs full width at half maximum (FWHM) is determined by the autocorrelation of the pump and the probe beams. The fits in Figs. 3(a) and 3(b) give a value $\tau = 100 \pm 60$ fs within the pump energy range 0.4–2 $\mu$J used in the experiment. The dynamics of the Kerr signal $\Delta I_K(\Delta t)$ is not affected by the polarization state of the photons, as shown in Fig. 3(b) using a circular polarization of the pump beam. In Fig. 3(c), the reflectivity normalized signal obtained for a pump energy of 1.6 $\mu$J is represented. The corresponding magnetic and electronic dynamics are very similar.

The relaxation time $\tau$ of ~100 fs, which is within the actual temporal resolution of the setup, is much smaller than the usual spin-lattice relaxation in metals. Typical reported values are at least of a few tens of picoseconds.[1,9,10] As previously stated in the case of nickel films,[2] this shows that the underlying mechanisms involved in the demagnetization dynamics cannot be attributed to the heating of the spins by the lattice.

The ultrashort response time $\tau$ excludes a demagnetization induced by the lattice. Two other mechanisms are discussed below. First, one can raise the question of a demagnetization directly induced by the pump photons. Such an effect, which relies on the transfer of angular momentum between the photon field and the electrons, is expected to depend strongly on the polarization of the pump beam. It can be ruled out from the results of Fig. 3, where it is seen that the dynamics are identical both for linear and circular polarization. Let us notice that in this case, the pump beam was at normal incidence on the sample and propagating along the direction of the applied magnetic field. Second, one can consider a demagnetization induced by the interacting electron populations. It is noticeable in Fig. 3(c) that the demagnetization dynamics follows the reflectivity changes. The time delay when the extremum of the reflectivity occurs is usually related to the thermalization time of the electron population (i.e., the time required by the nascent electronic population to acquire a Fermi-Dirac distribution). This thermalization time has been observed to be a few hundreds of femtoseconds in gold films.[11] More recently the study of the initial transmission and reflection pump-probe signals in nickel films showed that a similar thermalization of the electrons occurs within ~300 fs.[12] The mechanism of this thermalization is attributed to the electron-electron collisions that are mediated by the Coulomb interaction. The present experimental results in $CoPt_3$ show that the demagnetization occurs simultaneously with this electron thermalization. Such fast demagnetization dynamics which also occurs in nickel films[3,12] can be accounted for using a model with four coupled baths (nonthermalized electron populations, thermalized electrons, spins and lattice).[12] The coupling between the nonthermal electrons and the spins may occur via the spin-orbit interaction: during the collisions, the total spin of the electron population is not conserved, due to this spin-orbit coupling, resulting in the demagnetization. Let us mention that the initial nonthermal electron population was not taken into account in Ref. 2 where we focused on the equilibrium dynamics between the electrons, spins, and the lattice that takes place in the first few picoseconds.

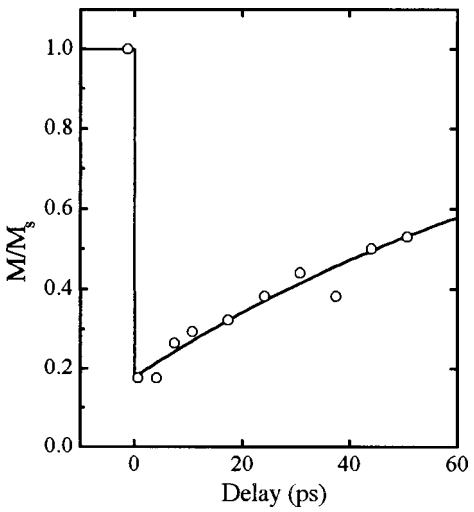


FIG. 4. Extended time scale for the demagnetization with a pump pulse energy 0.8 $\mu$J. The line is an exponential fit to the Kerr signal recovery at positive delays.

In connection with the technology of magneto-optical devices, the dynamics for the cooling down of the spin system is also of interest, since it determines the highest repetition rate at which a thermomagnetic device can operate. The recovery of the Kerr signal at longer delays is reported in Fig. 4. Exponential fits to these data reveal a characteristic time for the cooling down of ~90 ps. In fact, this time is comparable with the exponential decay time observed for the reflectivity of the sample (about 80 ps). It is known that, in this dynamical regime, the electron and the lattice are in equilibrium. The magnetization increase that we observe in Fig. 4 is therefore mainly related to the cooling of the temperature of the sample, as reported by Guarisco *et al.*[13] According to the simulations of Ref. 14, for Ni films, the temperature dynamics for delays larger than a few tens of picoseconds is related to the homogenization of the temperature profile within the metallic film. This process is followed by the heat propagation into the insulating substrate.

In conclusion, using time-resolved polar MOKE experiments, we have observed that femtosecond laser pulses can induce a complete demagnetization of a ferromagnetic film. This effect, observed here in $CoPt_3$ films and which looks similar to the ferro- to paramagnetic phase transition observed in static measurement, occurs in a time scale when the electrons and spins are not in equilibrium with the lattice. An analysis of the Kerr signal relaxation shows that the characteristic time for the demagnetization (about 100 fs) is comparable to the pulse width. Therefore, most of the demagnetization occurs during the thermalization of electron populations to a Fermi-Dirac distribution.[15] Possible mechanisms for the demagnetization are discussed. The ''heating'' of the spin system either from the spin-lattice interaction or from direct interaction with the photon field have been shown to have no major influence. We suggest that the ultrafast demagnetization can be explained taking into account the interactions between the spin-polarized electron population following the optical absorption. However, the description within the framework of a microscopic theory of the spin dynamics during the time scale where the relaxation of electronic population occurs is still an open question.

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