Coherent terahertz emission from ferromagnetic films excited by femtosecond laser pulses

E. Beaurepaire[a)]
IPCMS (UMR 7504 CNRS-ULP), 23 rue du Loess, BP43, F-67034 Strasbourg Cedex 2, France

G. M. Turner, S. M. Harrel, and M. C. Beard
Yale University, Chemistry Department 225 Prospect Street, P.O. Box 208107, New Haven, Connecticut 06520-8107

J.-Y. Bigot
IPCMS (UMR 7504 CNRS-ULP), 23 rue du Loess, BP43, F-67034 Strasbourg Cedex 2, France

C. A. Schmuttenmaer[a)]
Yale University, Chemistry Department 225 Prospect Street, P.O. Box 208107, New Haven, Connecticut 06520-8107

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It is shown that the laser induced ultrafast demagnetization of ferromagnetic films results in the emission of a terahertz electromagnetic pulse. This emission has been detected from Ni films using free-space electro-optic sampling. The radiated electric field $E(t)$ is explained by Maxwell equations (radiation from a time dependent magnetic dipole), and is expected to be proportional to the second time derivative of the magnetization d^2M/dt^2, as measured in the far field. This technique opens appealing perspectives in the context of measuring and understanding the ultrafast spin dynamics as well as the interaction of electrons (both charge and spin) with electromagnetic fields. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1737467]

The study of spin dynamics is essential for technological applications, since it allows one to determine the maximum working frequency of devices for magnetic storage and information processing in applications such as magnetic random access memory (MRAM), among others. Spin dynamics are also a critical issue for magnetism since the fundamental time scales involved are not yet completely identified. Conceptually, the simplest method for changing the magnetization of a magnetic sample is through the manipulation of ultrafast (typically 50 ps rise time) magnetic fields. Such dynamics can be interpreted by the Landau–Lifshitz equations describing the precession and damping of the magnetization vector about the effective field.[1] Micromagnetic effects such as inhomogeneous magnetization distributions can be included in the theory as needed.[2] It has been shown recently that precessional switching may occur if an adequate magnetic field is applied perpendicular to the easy axis of a micron-sized element,[3] which seems to be an optimized strategy regarding the speed of magnetization reversal.[4] However, this approach bears its own physical limitations since the precessional switching frequency is related to the Larmor frequency, $\omega_L = 28$ GHz/T. Thus, truly picosecond switching requires the application of a magnetic field changing at a rate of a few Tesla/ps, which is only accessible at specialized facilities.[5]

An alternative method for the excitation of spin dynamics in metallic films utilizes application of ultrafast laser pulses. The underlying idea is that a rapid elevation of the spin temperature produces a decrease of the magnetization as the Curie temperature is approached. It was recently shown that the magneto-optical Kerr signal of a Ni film drops by ~50% in the picosecond that follows the absorption of a 60 fs laser pulse.[6,7] This was the first indication of a magnetization change faster than the spin-lattice relaxation time (~100 ps to 1 ns for Ni), and was interpreted as nonequilibrium heating of the spins, electrons, and the lattice. This observation was later supported by experiments utilizing magnetization-dependent surface second harmonic generation,[8] and time- and spin-resolved two photon photoemission.[9] Similarly, a complete loss of magneto-optical signal was also observed within less than 1 ps after absorption of the pump pulse in the high fluence regime (a few mJ/cm^2) for alloys[10,11] and ultrathin[12] films.

However, in such experiments, magnetism is probed through optical transitions, and it is questionable as to what extent at least part of the observed dynamics can be attributed to an optical effect (bleaching of optical transitions). Indeed, at least in two situations, namely pump and probe magnetic second harmonic generation for Ni(110) surface[13] and magneto-optical Kerr effect (MOKE) for Fe thin films,[14] it was concluded that nonmagnetic (charge) dynamics have significant contributions in the ''magnetic'' signals, even for delays as long as 30 ps. On the other hand, detailed studies of time dependent MOKE data for Ni ultrathin films[15] and $CoPt_3$ films[16] support the idea that for delays longer than the electron thermalization time (i.e., the time necessary for the recovery of the Fermi–Dirac distribution of electrons after absorption of the short laser pulse, which is on the order of 100 fs in metals), the spins dominate the magneto-optical response. In this context, it is imperative to develop experimental tools sensitive to magnetization variations indepen-

a)Author to whom correspondence should be addressed; electronic mail: charles.schmuttenmaer@yale.edu; beaure@ipcms.u-strasbg.fr

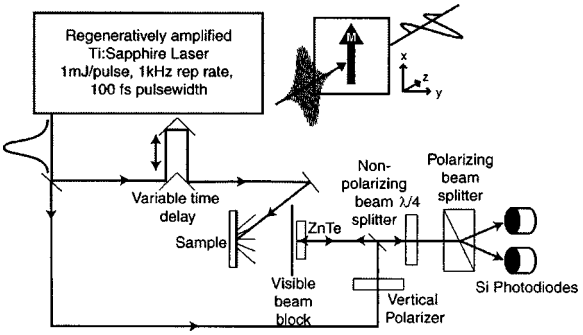

FIG. 1. Schematic diagram of experimental configuration. Inset defines coordinate axes.

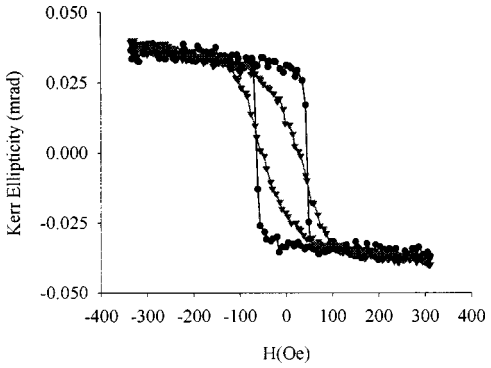

FIG. 2. Longitudinal Kerr hysteresis loops recorded along orthogonal easy (black/circle) and hard (red/triangle) magnetic axis of a Cr(30 Å)/Ni(42 Å)/Cr(70 Å) film deposited on glass. The measurement is performed using a diode laser (λ=670 nm).

dent of optical oscillator strength, which are responsive on the time scales of excitation laser pulse durations currently available.

In this letter we exploit the idea that a ferromagnetic film demagnetized on a subpicosecond time scale will generate an electromagnetic field according to Maxwell's equations. The radiated wave detected contains information intrinsically related to the spin dynamics. Similar schemes have been used to detect ultrafast intramolecular charge transfer dynamics in partially oriented molecular systems,[17] as well as carrier dynamics in semiconductors,[18,19] and Cooper-pair breaking in superconductors.[20] We have detected the corresponding electric field accompanying the changing magnetic field using time resolved electro-optic sampling schemes with a THz bandwidth, providing insight in this field of research.

A recent review on THz emitters and detection can be found in Ref. 21. The experimental geometry, shown in Fig. 1, employs a regeneratively amplified Ti:sapphire laser yielding 800 μJ/pulse at 800 nm, with pulse duration of 100 fs full width at half maximum. Its output is split into two portions, with about 99.9% of the energy being used to heat the sample after traveling along a variable delay line. A paper or polystyrene visible beam block ensures that any visible power not absorbed by the sample does not reach the detector. The other portion of the visible pulse reflects off of the beamsplitter and travels through a vertical polarizer before reflecting off of a second nonpolarizing beam splitter. It is used to detect the electromagnetic transient via free-space electro-optic sampling in a 0.5-mm-thick $\langle 110 \rangle$ ZnTe crystal.[22] Sample demagnetization is initiated by electron heating induced from absorption of a pulse with 200 μJ energy and spot size of $\sim$5 mm (leading to $\sim$1 mJ/cm^2 fluence). The sample is illuminated from the side that has the metal film, and the THz pulse emitted in the counterpropagating direction is measured. It is important to note that the THz pulse does not travel through the glass substrate, and is therefore only affected by the properties of the metal film.

The measurements presented here employ Cr(30 Å)/Ni(42 Å)/Cr(70 Å) thin films obtained by e-beam deposition under UHV on outgassed glass substrates. In-plane uniaxial anisotropy in Ni films was achieved by e-beam evaporation with an oblique incidence of $\sim$15°. Magnetic properties of these samples were studied by a conventional Kerr magnetometer, rotating the sample along its normal by steps of 5°. Uniaxial behavior was obtained with easy and hard magnetic axis separated by 90°. Typical hysteresis loops are reported in Fig. 2. THz measurements are then performed after saturating the samples along the easy axis, allowing 90% or higher remanence.

The electric field emitted upon ultrafast demagnetization of a Ni thin film is displayed in Fig. 3(a). The inset of Fig. 1 displays the coordinate system used. The excitation beam propagates along the z direction, the in-plane magnetization of the sample is along the x direction. By using wire-grid THz polarizers we have verified that the emitted radiation propagating in the z direction is indeed polarized along the y direction. As the sample is rotated about the z axis, the amplitude of the y-polarized emission varies as the cosine of the angle between the magnetization axis and the x axis. We have also verified that the emitted pulse is independent of the visible laser polarization angle relative to the x axis, and that it is independent of whether the excitation pulse is linearly or circularly polarized. The amplitude of THz emission along the surface normal from nonmagnetic films (Pt) or films with perpendicular magnetization (Co/Pt multilayers) is weaker by a factor of 50 and 10, respectively.

The basic features of the electric field can be easily interpreted from the classical Maxwell theory of electromag-

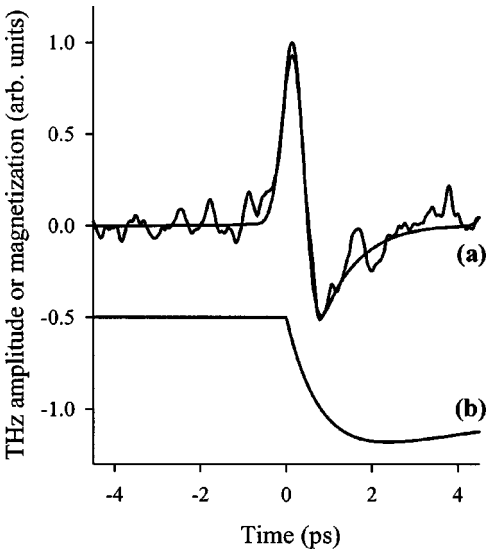

FIG. 3. Part (a) is the THz pulse generated upon ultrafast laser heating of 42-Å-thick in-plane magnetized Ni film. The smooth line is a simulation assuming the time-dependent magnetization of part (b) convoluted with a Gaussian instrument response function (full width at half maximum 540 fs).

netism. If one assumes that pump pulses coherently excite elementary magnetic dipoles in the film producing a time-varying magnetization, the electric field emitted in the far field (polarized in the y direction), propagating in the z direction is

$$E_y(t) = \frac{\mu_0}{4\pi^2 r} \frac{\partial^2 M_x}{\partial t^2}(t - r/c), \tag{1}$$

where r is the distance to the dipole. The description presented above suggests that the measurement of transient electric field emitted by the sample is related to the second time derivative of the magnetization.

In order to verify this relationship between the electric and magnetic fields let us assume a temporal variation of the magnetization [Fig. 3(b)] similar to the one previously observed in pump-probe experiments[16]

$$\Delta M(t) = \{-\Theta(t)[k_1(1-e^{-t/\tau_1})e^{-t/\tau_2} + k_2(1-e^{-t/\tau_2})]\} \otimes G(t), \tag{2}$$

where t is time, $\Delta M(t)$ is the time-dependent change in magnetization, $\Theta(t)$ is the Heaviside step function centered at $t=0$, k_1 and k_2 are constants depicting the relative amount of transient response versus long-term values. In the present context, τ_1 and τ_2 may be viewed as phenomenological constants describing the initial decay and recovery time of the magnetization.[6,7,15,16] $\otimes G(t)$ represents convolution with a Gaussian instrument response function. The best fit of the second derivative of the magnetization after convolution with the detector response function is plotted as the smooth line in Fig. 3(a).

The present work shows that the ultrafast demagnetization process in a ferromagnetic film is intimately related to emission processes in the far infrared. Therefore, one should consider the interaction of the emitted radiation with the spin polarized electrons on the same footing in order to obtain a satisfactory description of the overall dynamics when energy and angular momentum are conserved. From such a perspective, the present model is far too simple since it does not describe the mechanisms involved in the spin dynamics. From a microscopic point of view, a plausible scenario is that the sample is optically excited by the pump pulse inducing spin conserving optical transitions. At a later time, spin-flip events in the electron relaxation process occur via the spin orbit coupling, which should not give rise to a complete demagnetization as has been experimentally observed.[10] Simultaneously, the relaxation of electrons (spins) between the majority and minority subbands, assisted by photon emission in the far infrared, occurs, contributing to the demagnetization process. This description would be the extension of a previously proposed model where the coherent interaction between the photons and electrons has been considered.[23]

In conclusion, we have observed THz radiation attributed to the ultrafast demagnetization of thin ferromagnetic films. The symmetry properties of the emitted electric field (polarization direction with respect to the direction of the magnetization) are in agreement with a time-dependent magnetic dipole described by classical electromagnetic theory. Further experimental and theoretical developments are now necessary in order to fully take into account effects related to heat diffusion and the propagation of the far-infrared pulse inside the metallic film. Such efforts will allow this technique to realize its full potential in directly characterizing time-varying magnetizations in samples of arbitrary construction. It is an approach that will allow profound insights into the mechanisms associated with the demagnetization since it gives direct access to a process (the photon emission) not observable in pump-probe experiments. It is therefore a very useful and complementary methodology that could be systematically employed to unravel the subtleties of the ultrafast spin dynamics in metals.

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