Dynamics of electron-magnon interaction and ultrafast demagnetization in thin iron films

E. Carpene,* E. Mancini, C. Dallera, M. Brenna, E. Puppin, and S. De Silvestri
ULTRAS CNR-INFM, Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy
(Received 2 July 2008; revised manuscript received 10 October 2008; published 20 November 2008)

The electronic and spin dynamics in thin iron films have been investigated by means of time-resolved reflectivity and time-resolved magneto-optical Kerr effect. Combining the two techniques, it is possible to clarify the role of electron-phonon and electron-magnon interactions on the ultrafast magnetization dynamics. In particular, we show that the rapid (~100 fs) demagnetization is established at the electronic level through electron-magnon excitation, while the subsequent recovery of the spin order is attributed to the Elliott-Yafet spin-flip scattering process on a time scale slightly shorter than a picosecond. Both processes have characteristic time constants that undoubtedly differ from the measured electron-phonon coupling time of 240 fs.

DOI: 10.1103/PhysRevB.78.174422 PACS number(s): 78.20.Ls, 75.40.Gb, 75.70.−i, 78.47.−p

I. INTRODUCTION

Disclosing the elementary excitations in ferromagnets, and in particular the processes governing spin dynamics on the femtosecond time scale, is one of the most challenging subjects in modern solid-state science. It is well established nowadays that the excitation of a ferromagnet through an ultrashort laser pulse induces a demagnetization in the subpicosecond regime.[1–5] The undisputed fact is that photons of a femtosecond laser, focused on a metal, are absorbed by electrons close to the Fermi level, originating a nonequilibrium distribution that thermalizes with the surrounding environment (other electrons and lattice) via electron-electron and electron-phonon scattering. In ferromagnetic materials, however, the spin order is an additional factor that must be considered. Scattering events not only modify the energy and momentum of the particles involved, but they can also induce spin-flips, affecting the net magnetization of the sample. Electrons can modify their spin through different processes, mainly (i) Stoner excitations, (ii) electron-magnon scattering, and (iii) phonon-mediated spin-flip events, also known as Elliott-Yafet events.[6] Processes (ii) and (iii) dominate for low (visible and infrared (IR)) excitation energy.[7] Despite the considerable experimental evidence gathered in the last decade, only a few microscopic models of the ultrafast magnetization dynamics have been proposed. Zhang *et al.*[8] suggested an interplay between spin-orbit coupling and laser photons as responsible for the rapid demagnetization. Kazantseva *et al.*[9] modeled the subpicosecond spin dynamics in ferromagnets using an atomistic approach based on the Landau-Lifshitz-Gilbert equation, where the energy transfer between electrons and spin system is determined by a phenomenological coupling constant. The direct transfer of spin angular momentum to the lattice, through an ultrafast, phonon-mediated spin-flip scattering process has been suggested theoretically[10] and supported very recently by time-resolved x-ray magnetic circular dichroism (XMCD) experiments on Ni films.[11] Also the role of photon polarization on the spin order has recently received attention.[12] However, a clear and ultimate picture of the mechanisms governing magnetization on the femtosecond time scale is still missing.

Here, we used an all optical pump-probe approach to investigate the electronic and spin dynamics in thin iron films by means of time-resolved reflectivity and time-resolved magneto-optical Kerr effect (TR-MOKE). Although our phenomenological observations in iron substantially agree with the results on other ferromagnetic metals (similar demagnetization times and electron-phonon coupling rates), the direct comparison between reflectivity and MOKE measurements allows us to infer that ultrafast demagnetization is established at the electronic level, through transfer of spin to orbital angular momentum. Eventually, the lattice absorbs the orbital momentum, as already observed in the experiments on Ni. The main goal of this paper is to provide foundation for the suggested all-electronic demagnetization mechanism. It should be mentioned that a long-going scientific debate on whether TR-MOKE provides valid information on magnetic dynamics or is affected by nonmagnetic artifacts is still open.[13] In one of the few works involving TR-MOKE on iron films, Kampfrath *et al.*[14] showed that nonmagnetic contributions prevent the determination of the magnetization dynamics. However, this conclusion critically depends on the specific experimental conditions. Later, Bigot *et al.*[15] showed that magneto-optical signals truly reflect the spin dynamics in ferromagnets. Our results, and in particular the simultaneous measurements of reflectivity and magneto-optical effect, support the thesis that MOKE in the femtosecond regime is genuine.

II. EXPERIMENTAL DETAILS

The experiments have been conducted on a thin Fe(100) film (about 7 nm thick) epitaxially grown on MgO(100) at room temperature in ultrahigh vacuum. The film quality and its crystallographic orientation have been checked *in situ* by means of low-energy electron diffraction . The optical analysis has been performed *ex situ* with an amplified Ti:Sapphire laser, generating 60 fs, 1 kHz pulses centered at 800 nm (1.55 eV). TR-MOKE has been performed in longitudinal configuration, applying the external magnetic field along the Fe(100) easy axis and parallel to the film surface. The external field has been varied between ±10 mT, although the film coercive field was lower than 1 mT. Both pump and probe wavelengths were 800 nm with crossed polarizations. Figure 1 schematically sketches the experimental setup. Before reflection on the sample, the *s*-polarized probe beam

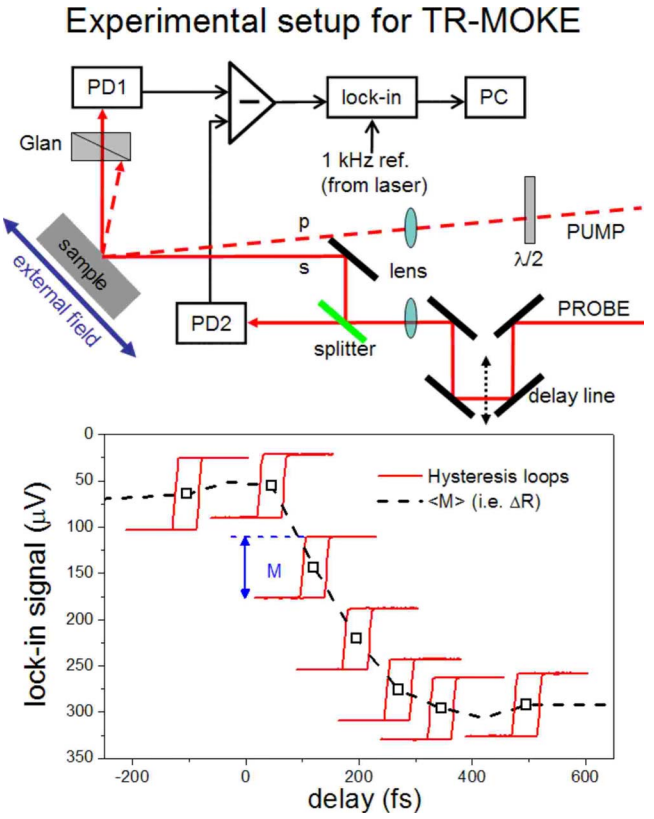

FIG. 1. (Color online) Schematic sketch of the experimental setup for time-resolved MOKE. We used a two-detector scheme and a lock-in amplifier to improve the sensitivity of the measurements, without any beam modulation. For each delay between pump and probe we varied the external magnetic field with a symmetric triangle ramp. This allowed us to obtain simultaneously the evolution of the magnetization M (from the hysteresis loops) and the transient reflectivity ΔR of the iron film.

was split.[16] One portion was focused on the sample at an incidence angle of about 45° inside the spot, 180 μm in size, irradiated by the pump pulses. The reflected beam was then analyzed with a Glan polarizer rotated at 45° with respect to the probe polarization and directed onto a photodiode (PD1). The other portion of the probe illuminated a second photodiode (PD2) identical to the first one. The electrical signals from the two detectors have been fed into the differential input of a lock-in amplifier. Before hitting the second photodiode the beam was attenuated in order to obtain the smallest signal in the lock-in (i.e., very similar photocurrents were generated by the two detectors). This allowed us to operate the amplifier at high sensitivity and at the laser frequency (1 kHz), without further beam modulation. For each delay between pump and probe the hysteresis loops have been measured by varying the external field with a symmetric, triangle shape at frequency lower than 1 Hz and averaging over at least 60 cycles. The temporal evolution of the magnetization has been deduced from the variation of the remanence (M, see Fig. 1) as a function of the delay. On the other hand, the center of the loops shifted, following the transient reflectivity signal. This technique permitted to simultaneously extract time-resolved reflectivity and magnetization under the same experimental conditions. Since the use of identical wave-

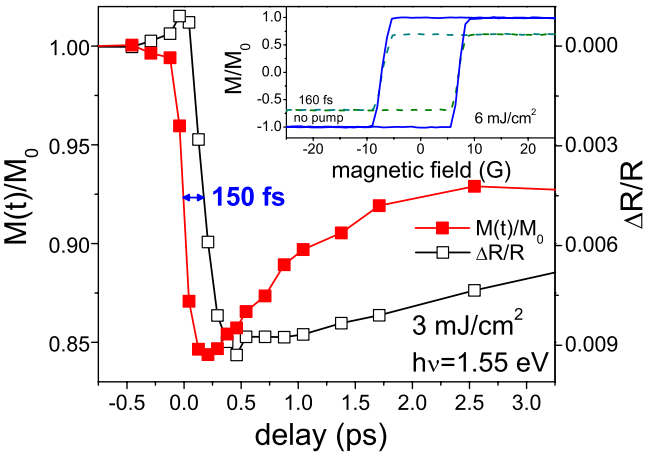

FIG. 2. (Color online) Comparison between the initial reflectivity (open symbols) and magnetization (solid symbols) curves induced by the 3 mJ/cm^2 pump pulse. The delay (~150 fs) between reflectivity and magnetization responses is emphasized. The inset reports the hysteresis loops measured with no pump (solid line) and 160 fs after the 6 mJ/cm^2 pump pulse (dashed line).

lengths for pump and probe beams might give rise to bleaching artifacts, we also checked the magnetization dynamics pumping with photons of 400 nm wavelength (second harmonic generated through a beta barium borate (BBO) nonlinear crystal) and no significant change has been found, as compared to the 800 nm pump beam. For reasons to be clarified later, transient reflectivity has also been measured with a separate experiment, using the 800 nm pump beam and varying the probe wavelength from 500 nm (2.48 eV) to 710 nm (1.75 eV), keeping the sample at remanence. The probe wavelengths have been selected from the supercontinuum beam generated by the s-polarized 800 nm photons through a 2 mm thick sapphire crystal. The white light has been focused on the sample inside the spot irradiated by the pump, and the probe wavelength has been discriminated after reflection on the sample with interferometric filters. Due to the group velocity dispersion inside the sapphire crystal the zero delay has been determined for each probe wavelength using the transient reflectivity signal of an amorphous silicon (a-Si) substrate placed under the iron film on the same sample holder and using a micrometer vertical stage to switch between them. The two samples have been carefully positioned so that no adjustment of the pump-probe overlap was necessary when switching from one to the other. Amorphous silicon has been chosen because it has a rather structureless optical response in the visible-near ultraviolet (UV) (Ref. 17) and an optical band gap lower than 1.3 eV,[18] resulting in a prompt reflectivity response for photon energy in the range 1.7–2.5 eV. This allowed us to determine the zero delay between pump and probe consistently for each wavelength.

III. RESULTS

As reported in the inset of Fig. 2, the effect of the laser excitation is to reduce the remanence (a 30% reduction is observed with a pump fluence of 6 mJ/cm^2), without affecting the coercive force. The temporal evolutions of rema-

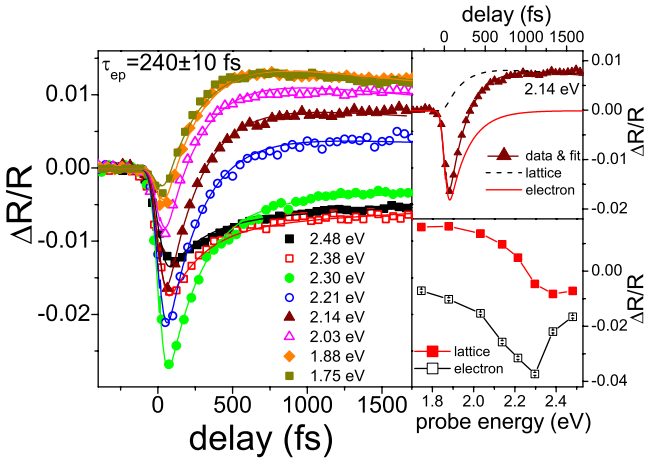

FIG. 3. (Color online) Time-resolved reflectivity curves (symbols), with corresponding fittings (lines) according to Eq. (1), obtained with 1.55 eV pump photons (800 nm) and various probe photon energies. The upper inset shows the electronic (solid line) and lattice (dashed line) contributions to the reflectivity curve measured with 2.14 eV probe. The lower inset reports their spectral weights vs the probe wavelengths.

nence and reflectivity, shown in Fig. 2, reveal a ~150 fs delay between their ultrafast responses. As it will be shown, this feature relates to the optical properties of iron and it proves to be very useful in disclosing the spin dynamics. A clear interpretation of MOKE results requires that purely optical effects are disentangled from magnetic ones.[2,14] This has motivated a more detailed investigation of time-resolved reflectivity before addressing the specific mechanisms driving the spin dynamics, with a twofold purpose: (i) give an explanation of the ~150 fs delay between the ultrafast responses of magnetization and reflectivity and (ii) deduce the electron-phonon relaxation time, to be compared with the characteristic dynamics of the magnetization. The interpretation of the transient reflectivity results can be very complex, in particular for transition metals, where d bands cross the Fermi level. Although in a recent work it has been shown that the full dependence of the reflectivity upon the electron occupation must be critically taken into account in order to deduce the correct electronic dynamics,[19] several simplified approaches proved to be very successful,[20,21] especially in deducing the electron-phonon coupling constants. A refined investigation of all the spectral features relating transient reflectivity and electronic properties in iron is beyond our scope and we have opted for a simplified model. As it will be shown, our experimental result matches the theoretical value.

Transient reflectivity of the Fe(100) film, measured with probe wavelengths ranging from 500 nm to 710 nm (2.48 eV to 1.75 eV, respectively) are reported in Fig. 3, along with the fittings. The data have been reproduced according to the following phenomenological model:[21]

$$\Delta R/R = \{\alpha[1 - e^{-t/\tau_{ee}}]e^{-t/\tau_{ep}} + \beta[1 - e^{-t/\tau_{ep}}]\}e^{-t/\tau_{th}}, \quad (1)$$

where only the parameters α and β (the spectral weights) have been allowed to vary with the probe wavelength,[22] while the time constants (τ_{ee}, τ_{ep}, and τ_{th}) have been considered fitting variables, but independent of the wavelength. The first term (proportional to α) represents the electronic response, initially determined by the electron-electron thermalization (with time constant τ_{ee}) and decaying by energy transfer to the lattice with the characteristic electron-phonon relaxation time τ_{ep}. The second term (proportional to β) accounts for the lattice heating and thus rises with the same time constant τ_{ep} (the additional parameter τ_{th} is the slow thermal relaxation rate, due to heat diffusion outside the irradiated area). A typical fit of the transient reflectivity is reported in the upper inset of Fig. 3, where electronic and lattice components are explicitly shown, while the lower inset reports their spectral weights as a function of the probe wavelength. The negative peak of the electronic component at 2.3 eV nicely matches the interband transitions at 2.3–2.5 eV deduced from the optical properties of iron in the visible range.[23,24] It is clear from Fig. 3 that the relative magnitude of these two contributions produces an *apparent* delayed response when probing with long wavelengths. Comparing the initial magnetization and reflectivity dynamics of Fig. 2 with the reflectivity curves obtained at short (2.3 eV) and at long (1.75 eV) probe wavelengths in Fig. 3, it is evident how the demagnetization front promptly follows the *electronic* component of the transient reflectivity, indicating that the loss of spin order is connected to the nonequilibrium electron distribution induced by the pump pulse. Although the time constant τ_{ee} cannot be unequivocally determined from the reflectivity curves, being shorter than the pulse duration[25] ($\tau_{ee} < 30$ fs from the fittings), the electron-phonon relaxation time $\tau_{ep} = 240 \pm 10$ fs has been extrapolated. As it will be shown below, this experimental value is in excellent agreement with the one deduced from the theory of electron-phonon scattering, and it definitely differs from the time constants characterizing the magnetization.

We now focus on the phenomenological aspects of the spin dynamics, in particular within the first few picoseconds after the laser pulse. The time evolution of the remanence is shown in Fig. 4 for different pump fluences. Nonlinear (i.e., multiphoton) effects are ruled out, since M scales proportionally to the pump fluence. Regardless of the pump intensity, three distinct regimes can be identified: (i) the demagnetization time is roughly 160 fs, (ii) a subsequent, partial recovery of the spin order is observed within about 3 ps, (iii) the original value of M is not reestablished within the investigated time window of 100 ps. The last temporal feature is attributed to heat diffusion outside the irradiated area and will not be further investigated. In order to extract quantitative values, we have fitted the experimental magnetization curves with a phenomenological approach similar to Eq. (1): the demagnetization front has been modeled with an exponential function proportional to the term $(1-e^{-t/\tau_{em}})$, while the subsequent recovery has been assumed to follow a biexponential law, including a fast rate τ_s (~1 ps) representing the spin relaxation, and a slow rate (>100 ps) embodying the effect of heat dissipation.[22] The demagnetization time constant τ_{em} ranges from 50 to 75 fs, increasing with the pump fluence, as displayed in the inset of Fig. 4. For comparison, the electronic response extrapolated from the reflectivity data of Fig. 3 is reported as well, revealing the similarity between the *initial* spin and electron dynamics. The

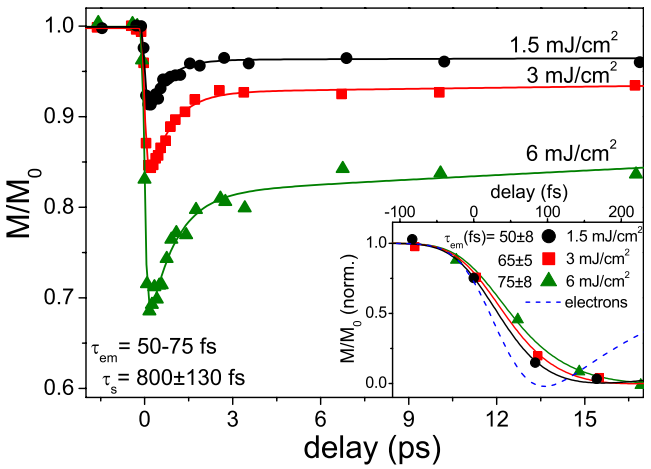

FIG. 4. (Color online) Magnetization curves at three different pump fluences (symbols) and corresponding fittings (lines). The inset reports the normalized demagnetization fronts in greater detail, with the corresponding values of the time constant τ_{em}. For comparison, the electronic component of the transient reflectivity, as extracted from the curves of Fig. 3, is shown as well (dashed line).

spin relaxation constant is $\tau_s = 800 \pm 130$ fs with a weaker dependence on the pump intensity. This value is definitely larger than the electron-phonon coupling time of 240 fs, establishing that electrons and spins follow diverse relaxation paths after optical excitation.

IV. DISCUSSION

We will now substantiate our results with some theoretical arguments, starting from the computation of τ_{ep}. The interaction between electrons and phonons in metals has been extensively investigated in the past,[26] even in the ultrafast regime.[27] The theory is based on the rate of change of the electron and phonon distributions due to collisions. Following the method proposed by Allen,[27] the electron-phonon coupling time τ_{ep} relates to the electronic temperature T_e, the coupling constant λ_p (~ 0.5, see Ref. 28), and the Debye frequency ω_D (for Fe, $\hbar\omega_D = k_B\Theta \sim 40$ meV, $\Theta = 470$ K being the Debye temperature[29]). The electronic temperature T_e has been deduced from the absorbed laser energy per unit volume E_a using the relation $E_a = \gamma(T_e^2 - T_0^2)/2$, with T_0 being the initial electronic temperature (room temperature) and $\gamma = 0.7$ mJ/cm^3 K^2 being the electronic specific heat of iron.[29] E_a has been estimated according to the optical parameters of iron as $E_a = [1 - e^{-d/\lambda}]\phi(1-R)/d$, where ϕ is the incident laser fluence, $R = 0.5$ is the reflectivity of iron at the pump wavelength,[30] $\lambda = 17$ nm is the absorption length,[31] and $d = 7$ nm is the film thickness. Depending on the laser fluence, we estimated $T_e \sim 1000 - 2000$ K, leading to

$$\tau_{ep} = (2\pi k_B T_e)/(3\hbar\lambda_p\omega_D^2) \sim 150 - 300 \text{ fs}. \quad (2)$$

Our experimental value of $\tau_{ep} = 240 \pm 10$ fs excellently fits inside the expected range, reinforcing our interpretation of the transient reflectivity data. The fact that the sample remains in its magnetized state (see Fig. 4) even if the electronic temperature exceeds the Curie value [$T_C \sim 1000$ K for Fe (Ref. 29)] is not surprising given the strong far from equilibrium condition induced by femtosecond pulses. This observation is supported by the recent simulations of Kazantseva *et al.*[9]

In order to explain the spin dynamics, our starting point is the spin-orbit (LS) coupling. It can formally be written as $\mathbf{L}\cdot\mathbf{S} = L_zS_z + (L^+S^- + L^-S^+)$, where $L^\pm$, $S^\pm$ are the standard raising and lowering operators for orbital and spin momentum, respectively. In particular, the terms L^+S^- and L^-S^+ allow exchange between electron spin and orbital angular momenta. Based on the properties of L_z, one can easily verify that $T_z = dL_z/dt = -\partial U/\partial\phi$, where T_z is the torque in the z direction (i.e., the quantization axis), U is the potential energy of the electron, and ϕ is the rotation angle about the z axis.[32] Since U has the symmetry of the crystalline environment, $\partial U/\partial\phi$ is not independent of ϕ, and L_z is not a constant of motion (as it would be in the atomic case). Any variation of the orbital angular momentum will be rapidly quenched as the result of the torque exerted on the electron by the crystal field.[33] Therefore T_z can be regarded as the rate of angular momentum transferred between the electronic system and the lattice that ultimately acts as a reservoir of angular momentum.

Having recognized that LS coupling allows exchange between spin and orbital angular momenta, and that the latter is rapidly quenched by the crystal, it is now legitimate to wonder how these facts link to the femtosecond spin dynamics set off by the laser pulse. Under equilibrium conditions and for temperature $T \ll T_C$, the magnetization deviates from its saturation value due to low-energy spin excitations (magnons), the number of which follows the Bose-Einstein distribution.[34] Scattering events can modify this condition, but electronic collisions are hindered by the exclusion principle and limited to a small volume in the momentum space (within an energy of k_BT across the Fermi level). A femtosecond laser pulse, however, induces drastic changes promoting electrons to unoccupied levels and opening new scattering channels. Electron-electron interaction rapidly leads to a hot electronic distribution where electrons exchange energy, linear and angular momentum as well. In $3d$ ferromagnetic metals, the number of unoccupied levels for minority spin electrons is large, while majority bands are almost filled. Therefore, spin-flip processes should preferentially transform a majority electron into a minority one (i.e., the term L^+S^- prevails), reducing the net magnetization. Considering that magnon excitations are energetically favored, as compared to Stoner excitation, collective spin modes triggered by electronic scattering events should be expected. Through LS coupling (L^+S^-), the creation of magnons leads to a decrease in the spin momentum and to an increase in orbital angular momentum, but the latter is rapidly quenched by the crystal field. Therefore, in the proposed scenario, the ultrafast demagnetization dynamics is solely determined by the rate of magnon creation. This picture is substantially in agreement with the conclusions drawn by Koopmans *et al.*[10] to explain the rapid demagnetization, but it involves the mediation of electrons orbital angular momentum rather than phonons. Failure to directly observe the transfer between spin and orbital angular momenta with time-resolved XMCD measurements (see Ref. 11) does not strictly imply it is not present: if

an ultrafast quenching of the angular momentum occurs on a time scale shorter than the experimental time resolution it can hardly be revealed.

Although our previous considerations have at least in part speculative character, our Ansätze find quantitative support in the data, as we discuss in the following. We considered the close analogy between phonons and spin waves, and adapted the theory of electron-phonon interaction to evaluate the electron-magnon coupling time, as already suggested by Schäfer *et al.*[35] in the calculation of the electron self-energy. This can be achieved by replacing the Debye phonon dispersion relation $\omega \propto k$ [used to obtain Eq. (2)] with the magnon dispersion, $\omega \propto k^2$. Such a modification is equivalent to substituting the term $\lambda_p \omega_D^2/2$ in Eq. (2) with $\lambda_m \omega_m^2/5$, where ω_m and λ_m are the *magnon* cut-off frequency and coupling constant, respectively. Recent experiments on Fe have shown that the surface magnon cut-off frequency is $\hbar\omega_m \sim 170$ meV and the estimated coupling constant is $\lambda_m = 0.2$,[35] while the bulk magnon cut-off frequency is located at $\hbar\omega_m \sim 270$ meV.[36] Using $\lambda_m = 0.2$ and $\hbar\omega_m = 200$ meV leads to

$$\tau_{em} = (5\pi k_B T_e)/(3\hbar\lambda_m \omega_m^2) \sim 40 - 80 \text{ fs}. \quad (3)$$

This estimate nicely agrees with our experimental values of $\tau_{em} \sim 50-75$ fs. Furthermore, its dependence on the electronic temperature T_e is compatible with the dependence on the pump fluence we observed (see inset of Fig. 4). Although we cannot exclude *a priori* that the laser field can directly influence the spin polarization, as suggested by Zhang *et al.*,[8] our experimental evidence, and in particular the fluence dependence of τ_{em}, suggests this might not be the main demagnetization mechanism.

In order to complete the description of spin dynamics, we must address the recovery of the spin order following the ultrafast demagnetization. According to Elliott's formulation,[6] the spin relaxation time τ_s relates to the (Drude) electron momentum scattering time τ_D, and in particular $\tau_s \sim \tau_D/c^2$, where c is a dimensionless parameter related to the spin-orbit interaction. Although, to our present knowledge, no experimental value of c is available for iron, it has been extracted for other $3d$ metals from electron spin resonance, obtaining $c^2 \sim 10^{-2} - 10^{-3}$.[37] On the other hand, τ_D in Fe is about 2 fs (Ref. 34); therefore, one expects $\tau_s \sim 200-2000$ fs. Our experimental value of τ_s falls well inside the expected range, supporting the picture of Elliott-Yafet spin-flip process for the subpicosecond recovery of the magnetization.

V. CONCLUSIONS

In conclusion, we have shown that laser-induced ultrafast demagnetization of epitaxial Fe thin films can be understood in terms of electron-magnon interaction, taking place on a time scale <100 fs. Hot electrons can efficiently excite magnons, leading to a rapid reduction in the magnetization. This process is mediated by the spin-orbit coupling, allowing the transfer from spin to orbital angular momentum that is eventually absorbed by the lattice. The subsequent recovery of the spin order takes place with a characteristic time constant of ~ 800 fs, that is longer than the electron-phonon relaxation time (240 fs), and supports the picture of Elliott-Yafet spin-flip scattering. Based on general grounds and endorsed by simple calculations, we have provided a microscopic interpretation of the phenomena, the validity of which is not limited to iron, but extends to other ferromagnetic metals as well. It would be significant if these results stimulated further theoretical investigations on this topic in the future.

ACKNOWLEDGMENTS

The authors wish to thank F. Ciccacci for the enlightening discussions. C. Manzoni and D. Brida are gratefully acknowledged for their help with the optical experimental setup.

*ettore.carpene@fisi.polimi.it

[1] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996).
[2] B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, Phys. Rev. Lett. **85**, 844 (2000).
[3] H.-S. Rhie, H. A. Dürr, and W. Eberhardt, Phys. Rev. Lett. **90**, 247201 (2003).
[4] M. Lisowski, P. A. Loukakos, A. Melnikov, I. Radu, L. Ungureanu, M. Wolf, and U. Bovensiepen, Phys. Rev. Lett. **95**, 137402 (2005).
[5] M. Cinchetti, M. Sanchez Albaneda, D. Hoffmann, T. Roth, J. P. Wustenberg, M. Krauszs, O. Andreyev, H. C. Schneider, M. Bauer, and M. Aeschlimann, Phys. Rev. Lett. **97**, 177201 (2006).
[6] R. Elliott, Phys. Rev. **96**, 266 (1954); Y. Yafet, *Solid State Physics* (Academic, New York, 1963), Vol. 14.
[7] M. Plihal and D. L. Mills, Phys. Rev. B **58**, 14407 (1998).
[8] G. P. Zhang, Y. Bai, W. Hübner, G. Lefkidis, and T. F. George, J. Appl. Phys. **103**, 07B113 (2008), and references therein.
[9] N. Kazantseva, U. Nowak, R. W. Chantrell, J. Holhfeld, and A. Rebei, Europhys. Lett. **81**, 27004 (2008).
[10] B. Koopmans, J. J. M. Ruigrok, F. Dalla Longa, and W. J. M. de Jonge, Phys. Rev. Lett. **95**, 267207 (2005).
[11] C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E. F. Aziz, M. Wietstruck, H. A. Dürr, and W. Eberhardt, Nature Mater. **6**, 740 (2007).
[12] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Tsukamoto, A. Itoh, A. Kirilyuk, and Th. Rasing, Phys. Rev. Lett. **98**, 207401 (2007).
[13] P. M. Oppeneer and A. Liebsh, J. Phys.: Condens. Matter **16**, 5519 (2004), and references therein.
[14] T. Kampfrath, R. G. Ulbrich, F. Leuenberger, M. Münzenberg, B. Sass, and W. Felsch, Phys. Rev. B **65**, 104429 (2002).
[15] J.-Y. Bigot, L. Guidoni, E. Beaurepaire, and P. N. Saeta, Phys.

Rev. Lett. **93**, 077401 (2004).
[16] We followed the two-detector scheme of J. M. Florczak and E. Dan Dahlber, J. Appl. Phys. **67**, 7520 (1990).
[17] S. Adachi and H. Mori, Phys. Rev. B **62**, 10158 (2000).
[18] M. Durandurdu, D. A. Drabold, and N. Mousseau, Phys. Rev. B **62**, 15307 (2000).
[19] V. V. Kruglyak, R. J. Hicken, P. Matousek, and M. Towrie, Phys. Rev. B **75**, 035410 (2007).
[20] C.-K. Sun, F. Valleè, L. H. Acioli, E. P. Ippen, and J. G. Fujimoto, Phys. Rev. B **50**, 15337 (1994).
[21] N. Del Fatti, C. Voisin, M. Achermann, S. Tzortzakis, D. Christofilos, and F. Valleè, Phys. Rev. B **61**, 16956 (2000).
[22] The fitting function has been convoluted with a Gaussian-shaped temporal profile in order to account for the cross-correlated time width (~90 fs) of the laser pulses.
[23] T. Nautiyal and S. Auluck, Phys. Rev. B **34**, 2299 (1986).
[24] J. H. Sexton, D. W. Lynch, R. L. Benbow, and N. V. Smith, Phys. Rev. B **37**, 2879 (1988).
[25] V. V. Kruglyak, R. J. Hicken, M. Ali, B. J. Hickey, A. T. G. Pym, and B. K. Tanner, Phys. Rev. B **71**, 233104 (2005).
[26] G. Grimvall, *The Electron-Phonon Interaction in Metals* (North-Holland, Amsterdam, 1981).
[27] P. B. Allen, Phys. Rev. Lett. **59**, 1460 (1987).
[28] P. B. Allen, Phys. Rev. B **36**, 2920 (1987).
[29] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1986).
[30] We used the complex index of refraction and the Fresnel relations to estimate the reflectivity of iron for *p*-polarized pump photons impinging on the sample at 40°.
[31] J. H. Weaver and H. P. R. Frederikse, in *Handbook of Chemistry and Physics*, edited by D. R. Lide, (CRC Press, Boca Raton, FL, 2008).
[32] P. W. Atkins, *Molecular Quantum Mechanics* (Oxford Univerity Press, Oxford, 1983).
[33] In magnetic transition metals, $\langle L_z \rangle$ is not totally quenched due to the spin-orbit interaction. This can be explained in terms of band filling effect.See O. Eriksson, L. Nordstrom, A. Pohl, L. Severin, A. M. Boring, and B. Johansson, Phys. Rev. B **41**, 011807 (1990).
[34] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Saunders College, Philadelphia, 1976).
[35] J. Schäfer, D. Schrupp, E. Rotenberg, K. Rossnagel, H. Koh, P. Blaha, and R. Claessen, Phys. Rev. Lett. **92**, 097205 (2004).
[36] X. Y. Cui, K. Shimada, M. Hoesch, Y. Sakisaka, H. Kato, Y. Aiura, S. Negishi, M. Higashiguchi, Y. Miura, H. Namatame, and M. Taniguchi, J. Magn. Magn. Mater. **310**, 1617 (2007).
[37] P. Monod and F. Beuneu, Phys. Rev. B **19**, 911 (1979).