Spin-lattice relaxation in ferromagnets studied by time-resolved spin-polarized photoemission

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The spin-lattice relaxation time τ_{sl} is the characteristic time needed for the energy transfer between the lattice and the spin system. It is demonstrated that τ_{sl} can be determined with laser-induced spin-polarized photoemission. Upper and lower limits are derived from a single-pulse experiment using pulses of various time durations. It is found that τ_{sl} of iron is shorter than 20 ns but longer than 30 ps. A direct measurement of τ_{sl} is possible with a pump-probe experiment. One laser pulse heats the sample and a second time-delayed pulse is used to probe the magnetic state. τ_{sl} is found to be 100 ± 80 ps for ferromagnetic gadolinium.

I. INTRODUCTION

After a fast change of the lattice temperature, a magnet needs time to establish its new equilibrium magnetization. It will be demonstrated that this characteristic time, the spin-lattice relaxation time τ_{sl}, can be determined with laser-induced spin-polarized photoemission. Apart from its evident fundamental interest the knowledge of τ_{sl} is also of technological importance: it determines, for instance, the maximum speed attainable in Curie-point writing, the most widely used technique in magneto-optical recording.

Only few experiments are able to probe the time scale of the energy transfer from the lattice to the spin system mediated by the spin-orbit coupling. Kerr-effect measurements with short laser pulses gave an upper limit of 40 ns and a lower limit of 20 ps for τ_{sl} of nickel.[1] In ferromagnetic resonance (FMR) the longitudinal relaxation time T_1 contributes among other parameters to the linewidth of the FMR signal. T_1 is determined by the time the system needs to restore the *direction* of the magnetization after a small misorientation. The spin-lattice relaxation time τ_{sl}, in contrast, is the time the magnetization needs to assume a new equilibrium *magnitude* after an abrupt change of the lattice temperature. A fast change of the lattice temperature can be induced with short and intense laser pulses. When hitting a sample surface the pulses first induce a hot electron gas which then thermalizes with the lattice within a few picoseconds.[2-4]

Photoemission with spin analysis is described in Ref. 5. The spin polarization P of the photoelectrons is defined as $P=(N^{\uparrow}-N^{\downarrow})/(N^{\uparrow}+N^{\downarrow})$, where $N^{\uparrow}$ ($N^{\downarrow}$) is the number of electrons with spin magnetic moment parallel (antiparallel) to the surface normal of the sample. In the present experiment the polarization of the total photoyield is measured without energy analysis of the electrons. P is proportional to the magnetization along the surface normal of the sample. Along this direction an external field up to 3 T can be applied. The temperature of the sample is variable from 30 K up to several hundred K. The experiment is made time resolved by using short laser pulses for photoemission.[6,7] Then the magnetization is probed in a time interval determined by the duration of the laser pulse. It has been shown in Ref. 6 that space charge does not affect the polarization of the total photoyield because of conservation of the total angular momentum. This is a basic requirement for the performance of the experiments described below.

Upper and lower limits of τ_{sl} are derived from the set of measurements described in Sec. II. In these experiments one and the same laser pulse is used to heat the sample and to emit photoelectrons. The polarization of the photoelectrons measured as a function of the laser pulse intensity decreases if the Curie temperature is reached in the laser focus and if the spin-lattice relaxation time is short compared to the laser pulse duration. During a laser pulse with a duration much shorter than τ_{sl} the magnetization has no time to change its magnitude even if the pulse intensity is sufficient to melt the sample. It is found that τ_{sl} of iron lies between 30 ps and 20 ns.

In Sec. III a pump-probe experiment is described which gives a numerical value for τ_{sl}. A laser pulse with a photon energy below the photothreshold ($h\nu<\phi$) is used to heat the sample and a second pulse with $h\nu>\phi$ probes the magnetic state after a variable time delay with respect to the first pulse. For ferromagnetic gadolinium τ_{sl} is found to be 100 ± 80 ps.

II. DETERMINATION OF UPPER AND LOWER LIMITS FOR THE SPIN-LATTICE RELAXATION TIME OF IRON

After insertion into the UHV chamber a polycrystalline iron sample of cylindrical shape (diameter 5 mm, thickness 5 mm) was cleaned by cycles of Ar-ion sputtering and heating. The only remaining contamination found with Auger spectroscopy consisted of a few percent carbon.

The polarization of the photoelectrons emitted from Fe during 20 ns and 30 ps laser pulses is plotted in Fig. 1 as a function of the laser pulse energy. In both cases the photon energy is 2.15 eV. For photoemission the work func-

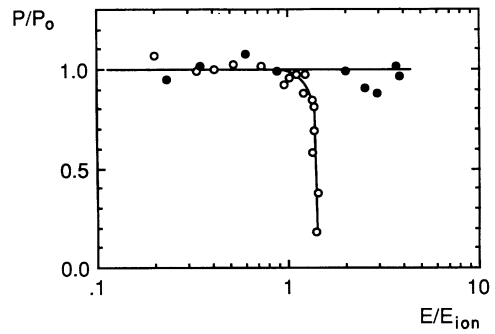

FIG. 1. Polarization of the photoelectrons emitted from polycrystalline iron during 20 ns (open circles) and 30 ps (full circles) laser pulses. A field of 0.38 T is applied perpendicular to the plane of the surface. The polarization is normalized by the spin polarization (P_0) measured with a weak laser pulse energy causing negligible heating. The energy is normalized by the threshold energy for positive ion emission (E_{ion}). E_{ion} is 90 mJ/cm^2 for the 30 ps pulses and 3.1 J/cm^2 for the 20 ns pulses.

tion of the iron sample has been lowered to 1.7 eV by depositing a submonolayer of Cs onto the surface. Cs is known to have no significant effect on the surface magnetic properties of iron.[8] The polarization is normalized to $P_0(=18\%)$, the polarization measured at a laser intensity weak enough to cause negligible heating.

The energy scale is calibrated in units of E_{ion}. This is the threshold energy, depending on the pulse duration, where the sample starts melting in the laser focus. Melting is accompanied by strong emission of positive ions[2] which are experimentally easily detected. An external field of 0.38 T is applied along the surface normal of the sample.

The sample is held at an initial temperature of 230 K. A temperature rise of 813 K (1579 K) is necessary to heat it up to the Curie temperature (melting temperature). Due to the Gaussian intensity distribution in the laser focus a significant loss of spin polarization is expected for laser energies exceeding $0.5E_{ion}$. For the 20 ns laser pulses this is observed in Fig. 1 (open circles). The behavior is in agreement with the findings reported in Ref. 6, where a noncesiated Fe sample was held in an external field of 1.7 T and irradiated with 20 ns laser pulses (photon energy 5 eV). The decrease of the polarization found in Fig. 1 (open circles) and in Ref. 6 is consistent with a spin-lattice relaxation time shorter than the duration of the 20 ns laser pulse. For a spin-lattice relaxation time much longer than the laser pulse duration the magnetization is expected to stay constant even if the intensity is sufficient to heat the lattice above T_c and even induce melting. Exactly this is observed if the experiment is performed with a 30 ps laser. The measurement is displayed by the filled circles in Fig. 1. It is found that a molten iron surface still emits polarized electrons if it is heated with 30 ps pulses. Consequently the spin-lattice relaxation time of iron must be longer than 30 ps.

For the interpretation of the measurements displayed in Fig. 1 the energy calibration in terms of E_{ion} is crucial since it establishes the only relation between pulse energy and temperature. For $E \geq E_{ion}$ the amount of positive charge emitted per laser pulse (positive ion yield) is measured by applying a negative voltage to the first electrostatic lens in front of the sample. Plotting the positive ion yield as a function of the laser pulse energy and extrapolating this curve towards zero gives E_{ion}. A yield measurement for iron is shown in Fig. 2. Using 30 ps laser pulses with a photon energy of 2.15 eV E_{ion} is found to be 38 ± 11 μJ. The diameter of the focus is 190 μm (FWHM).

A direct proof for the melting of the surface at pulse energies $E \geq E_{ion}$ is given by inspection of the irradiated area with a scanning electron microscope. In Fig. 3 a Fe surface is shown which has been hit by 400 laser pulses of 94 ± 10 μJ energy. The area with a laser-induced surface structure (LISS) has a radius of 69 ± 5 μm. LISS are known to occur if a temperature close to the melting temperature is reached in the laser focus.[9,10] Thus the energy density necessary to induce a surface structure and consequently to melt iron (E_{melt}) can be determined from Fig. 3 is the spatial flux (energy per cm^2) distribution is known in the laser focus.

The distribution of the energy flux in the laser focus is determined by measuring the size of the laser damaged area on a Polaroid film as a function of pulse energy. The resulting flux profile is shown in Fig. 4 for the 30 ps laser pulses which have been used to induce the surface damage shown in Fig. 3. The calibration of the y axis of Fig. 4 is given by

$$\int_0^\infty F(r)2\pi r\,dr = 94\ \mu\text{J}\ , \tag{1}$$

where $F(r)$ corresponds to the solid line in Fig. 4. It is a fit through the measured points using a superposition of two Gaussian curves. In the center of the 94 μJ pulse the flux turns out to be 232 mJ/cm^2. In Fig. 3 the surface has been disordered up to a distance of 69 ± 5 μm from the center of the laser spot. At this distance the laser has, according to Fig. 4, an energy density of 127 ± 17 mJ/cm^2. From Fig. 2 the threshold energy for positive ion emission (E_{ion}) was found to be 38 ± 11 μJ under the same focusing conditions. This corresponds to an energy

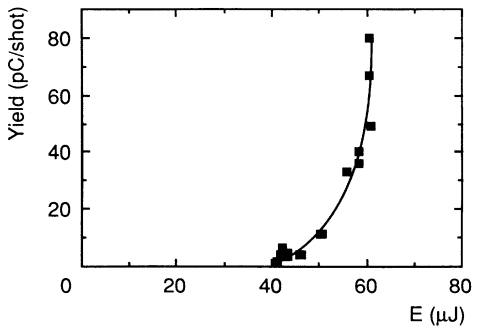

FIG. 2. Positive charge emitted from polycrystalline iron as a function of the laser pulse (duration: 30 ps, photon energy: 2.15 eV) energy. Extrapolating the curve towards zero gives the threshold energy for positive ion emission (E_{ion}). E_{ion} is found to be 38 ± 11 μJ. The diameter of the focus is 190 μm (FWHM).

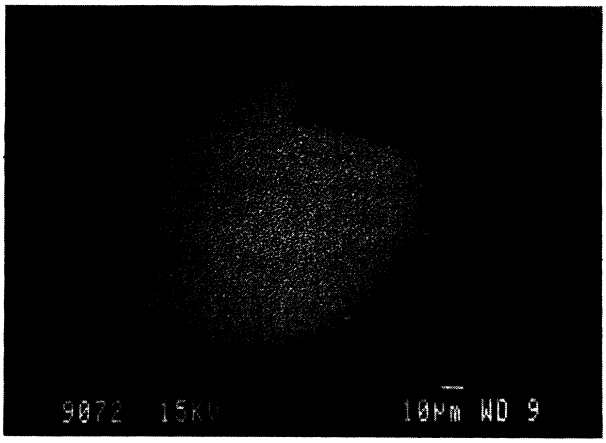

FIG. 3. Scanning electron microscope picture of the polycrystalline iron surface which has been irradiated with 400 30 ps laser pulses having an energy of 94±10 μJ. The laser-damaged area has an averaged radius of 69±5 μm.

density of [(38 μJ)/(94 μJ)] (232 mJ/cm^2)=94±35 mJ/cm^2 in the center of the focus. Therefore, within the experimental uncertainty the energy density necessary to emit positively charged ions and to induce laser damage are found to be equal.

A further confirmation that the surface starts melting at E_{ion} is obtained from an experiment involving optical spin orientation. Depending on the electronic structure polarized electrons can be emitted from a nonmagnetic material using circular polarized light.[11] The polarization P of the photoelectrons is fully determined by the crystal symmetry of the sample. Upon melting there is an abrupt decay of P from a constant value to zero.[11] Thus the melting of a surface during a short, circularly polarized laser pulse can be detected by measuring the decay of the polarization P of the optically oriented photoelectrons as a function of the pulse energy. P starts decreasing if the melting temperature is reached in the center of the laser focus.

The optical spin orientation experiment is performed with a β-Sn(001) (white tin) single crystal. The sample (cylinder: diameter 5 mm, thickness 5 mm) was cleaned by Ar-ion sputtering and was tempered at 460 K (T_{melt}=505 K) for several hours. This treatment was followed by cycles of sputtering and flash annealing cycles. Thereafter no traces of impurities were found in the Auger spectra and a reasonably sharp low-energy electron diffraction pattern was observed.

With a photon energy of 2.7 eV a transition from a non-spin-polarized initial state to a polarized final state can be induced in tin. The photothreshold of the sample was lowered below the final-state energy by deposition of Cs onto the surface.

The polarization of the optically oriented photoelectrons emitted from tin during 70 ps (full circles) and 12 ns (open circles) laser pulses is plotted in Fig. 5 as a function of the pulse energy normalized to E_{ion}. For both pulse durations P starts decreasing at an energy close to E_{ion}. The decrease of the polarization is reversible. This is checked by a control measurement at low pulse energy causing negligible heating. It is performed after each measurement with $E > E_{ion}$. If cesium had been desorbed or if the surface had become polycrystalline or amorphous because of the impact of the high-energy pulses the control measurement would have given a lower photocurrent and/or polarization. It was found that the polarization and the counting rate of the control measurement remained stable for energies up to $2E_{ion}$. The melting of the surface, which is recognized by the decrease of the polarization, starts at or close to E_{ion}. No difference is found between the heating with ns or with ps pulses. This is a reasonable result, since it is known that the energy transfer from the primary excitations caused by the laser pulse to the lattice, and correspondingly melting, occurs on a time scale of about 1 ps.[2–4]

Consequently the different dependence of the polarization as a function of pulse energy observed for iron irradiated with 30 ps and 20 ns laser pulses is due to a spin-lattice relaxation time which lies between the duration of these two pulses.

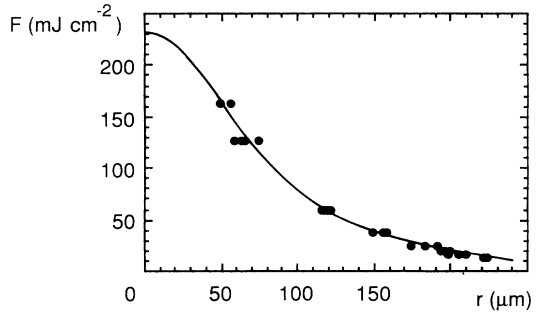

FIG. 4. Distribution of the energy flux in the laser focus of the 30 ps laser pulse. The flux profile is determined by measuring the laser-damaged area on a Polaroid film as a function of the laser pulse energy. The flux scale applies to the laser pulse which has been used to induce the surface damage shown in Fig. 3. In a distance of 69 μm from the center of the laser spot the flux is 127±17 mJ/cm^2. The solid line is a fit through the measured points by using a superposition of two Gauss curves.

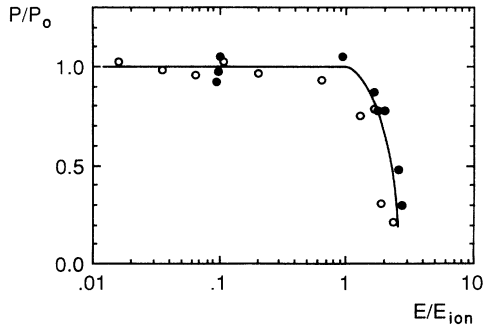

FIG. 5. Polarization of the optically oriented photoelectrons emitted from β-Sn(001) during 12 ns (open circles) and 70 ps laser pulses as a function of the pulse energy. The polarization is normalized by the spin polarization (P_0) measured with a weak laser pulse energy causing negligible heating. The laser energy is normalized by the threshold energy for positive ion emission (E_{ion}). E_{ion} is 100 mJ/cm^2 for the 70 ps pulses and 420 mJ/cm^2 for the 12 ns pulses.

III. DETERMINATION OF THE SPIN-LATTICE RELAXATION TIME FOR GADOLINIUM

A pump-probe experiment is performed in order to determine the spin-lattice relaxation time of Gd. The sample is heated with a ns laser pulse having a short rise time and a photon energy below the work function of the sample. The magnetization is probed by a time-delayed ps laser pulse with a photon energy above photothreshold. A relaxation time of 100 ± 80 ps is found for Gd. No basic experimental restrictions exist to apply the technique to other ferro- or ferromagnetic materials.

Gd has been evaporated onto a polycrystalline iron substrate (conically shaped, 6 mm long, exposed surface: 4 mm diameter) from a resistively heated W spiral. Before evaporation the iron was cleaned by sputtering and heating cycles. Film and substrate quality were tested with Auger spectroscopy. Extensive outgassing was necessary in order to produce clean Gd films. After the evaporation of Gd the Auger signal of Fe was completely suppressed. This gives a lower limit of 50 Å for the Gd film thickness. Figure 6 shows the spin polarization of the photoelectrons as a function of the magnetic field applied perpendicular to the sample surface. For a field of 0.38 T the Gd film is magnetically saturated. This small saturation field is due to the iron substrate which enhances the external field.

Due to the surface sensitivity of the photoemission experiment[12] the measured polarization reflects the magnetic state of the Gd film only. The temperature dependence of the polarization is shown in Fig. 7. A linear extrapolation of the curve points to a Curie temperature of 290 K. This value is in agreement with previous photoemission experiments[13] and with the bulk Curie temperature of Gd.[14] For the experiments displayed in Figs. 6 and 7 the full spectrum of the Hg-Xe lamp ($h\nu \leq 5.5$ eV) has been used. The photothreshold of the Gd film was 3 eV.

In order to perform time-resolved measurements the beam of an excimer laser is split into two pulses. One of them is directed over a variable beam delay line and thereafter pumps a 10 ns dye laser with a photon energy of 2.15 eV. No measurable electron emission is induced by this pulse; it just heats the sample. The absence of multiphoton excitations is due to the moderate laser pulse intensities used to heat Gd. For samples with a higher Curie temperature it might become necessary to reduce the photon energy of the heating pulse. The second part of the excimer beam pumps a 60 ps dye laser having a photon energy of 3.2 eV. The spin polarization of the electrons emitted by this second pulse is measured. By varying the delay distance the probing pulse can be moved with picosecond accuracy from 2.5 ns before to 7 ns after the onset of the heating pulse. Note that the probing pulse cannot influence the time-resolved measurement. It has been shown in Sec. II that the magnetization has no time to change during a 30 ps laser pulse. Therefore and because of its low laser pulse energy (0.5 μJ) the probing pulse has no effect on the magnetic state of the sample.

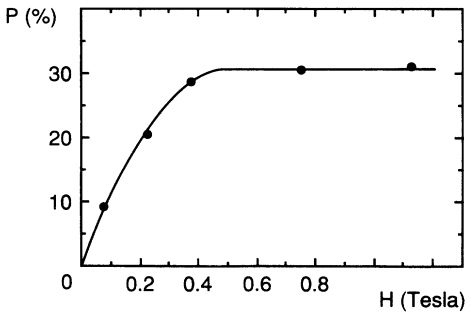

FIG. 6. Spin polarization P of the photoelectrons emitted from a Gd film as a function of the external field H applied perpendicular to the sample surface. The full spectrum of a Hg-Xe lamp ($h\nu \leq 5.5$ eV) is used as the light source. Complete alignment of the magnetization along the surface normal is achieved for external fields exceeding 0.38 T. The temperature is 45 K. The sample has a photothreshold of 3 eV.

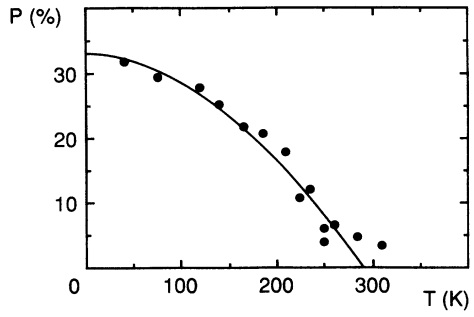

FIG. 7. Saturation polarization P of the photoelectrons measured as a function of the temperature. Saturation is achieved by applying an external field of 0.38 T. The full spectrum of a Hg-Xe lamp ($h\nu \leq 5.5$ eV) is used as the light source. The Curie temperature is 290 K.

For the time-resolved measurements described in the following an external field of 0.38 T has been applied perpendicular to the sample surface. The external field aligns the magnetization along the surface normal and in addition limits the diameter of the spot from which the electrons are collected by the electron optics. The diameter of the emitting surface area is measured by recording the photoyield during the scanning of a well-focused (151 μm FWHM) excimer laser beam over the sample surface. In Fig. 8 the counting rate is plotted as a function of the measuring position. Fitting the curve with a Gaussian function gives a diameter d_1 of 400 μm (FWHM) for the area contributing to the photocurrent. The diameter d_2 of the 60 ps probing laser pulse is 360 μm (FWHM). Both the probing laser and the external field limit the area which contributes to the polarization measurement to $d = 270$ μm ($d^{-2} = d_1^{-2} d_2^{-2}$), i.e., a region being much smaller than the heating spot diameter, which is 390 μm. Accordingly an almost homogeneously heated surface area contributes to the photoemission signal.

The effect of the heating pulse becomes visible in Fig. 9(a); the polarization decreases for $t > 0$ to $0.4P_0$. For $t < 0$ the probing pulse arrives before heating has started,

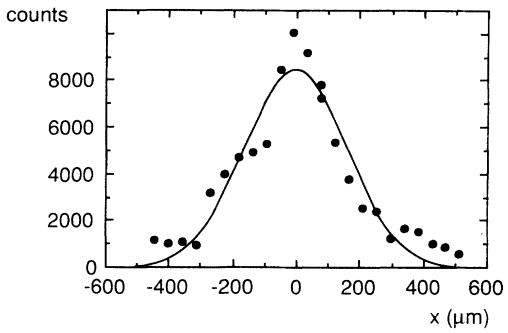

FIG. 8. Number of electrons measured in the Mott detector as a function of the measuring position. The curve is obtained by scanning a well-focused (151 μm, FWHM) excimer laser beam over the sample surface and by simultaneously monitoring the number of electrons detected in the Mott detector. An external field of 0.38 T is applied along the surface normal of the sample.

therefore $P_0(=38\%)$ measured at a negative time corresponds to the equilibrium magnetization at the initial temperature $T_0=45$ K. Due to the modest heating pulse intensity, the sample temperature remains below the Curie temperature. With the help of the $P(T)$ curve shown in Fig. 7 it becomes now possible to derive the rise of the spin temperature (ΔT_{spin}) corresponding to each polarization measurement of Fig. 9(a). The result is plotted in Fig. 9(b). Note the unique feature of the spin-polarized

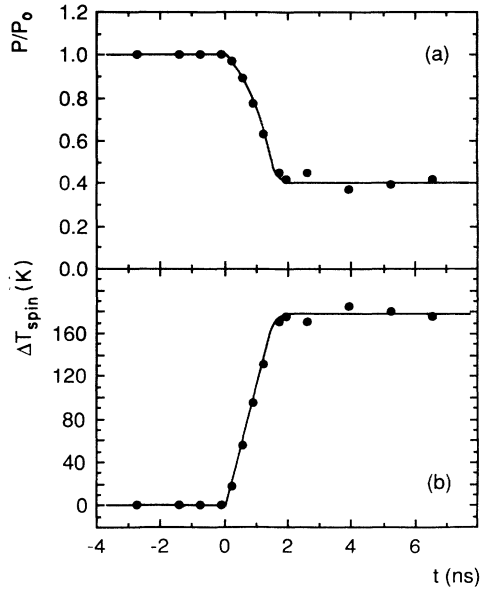

FIG. 9. (a) Pump-probe experiment using a 60 ps (3.2 eV) laser pulse as a probing pulse and a 10 ns (2.15 eV) laser pulse as a heating pulse. The sample is held at an initial temperature of 45 K. The reduced spin polarization of the photoelectrons emitted by the probing pulse is plotted as a function of the time delay between probing and heating pulse. Zero time delay corresponds to the onset of the lattice heating. (b) Rise of the spin temperature determined from measurement (a) by using the $P(T)$ curve shown in Fig. 7.

pump-probe experiment, namely, that the magnetization acts as a thermometer indicating at each instant of time the spin temperature. Unfortunately, for the lattice and electron gas similarly simple thermometers do not exist. The form of $\Delta T_{\text{spin}}(t)$ as evident from Fig. 9 is caused by the particular intensity profile of the heating pulse shown in Fig. 10 by the full circles.[15] The intensity scale on the left of the figure applies to the pulse energy used for measurement [9(a)]. The temperature rise $\Delta T_{\text{lattice}}$ can be derived from the temporal intensity profile of the laser pulse using the classical thermal diffusion equation.[16] The reason is that in our experiment the duration of the heating pulse is much longer than the characteristic time needed to transfer the pulse energy to the lattice, a time which is of the order of 1 ps.[2–4]

The result of solving the thermal diffusion equation is shown in Fig. 10 by the open circles. For $\Delta T_{\text{lattice}} > 20$ K there is a practically linear increase of the leading edges of the optical and thermal pulse profile. Extrapolation to $I=0$ and $\Delta T_{\text{lattice}}=0$, respectively, gives a displacement of 430 ps between the two pulses. Qualitatively, the displacement is due to the fact that the energy deposited by the laser pulse depends on t quadratically when the intensity increases linearly in time. Then, as long as thermal diffusion can be neglected, the temperature in the laser focus also increases quadratically in time. For $\Delta T_{\text{lattice}} > 20$ K diffusion of the lattice heat causes the initially parabolic relation $\Delta T_{\text{lattice}} \sim t^2$ to become linear. Replacing the front edge of both pulse profiles by straight lines, which is valid to a very good approximation, results in a shift of 430 ps between the onset of the optical pulse and the heating pulse.

$\Delta T_{\text{lattice}}$ shows a similar behavior as ΔT_{spin} [Fig. 9(b)]: both quantities level off to a constant value after a delay of 2 ns. Therefore T_{lattice} and T_{spin} must always be very close and the spin-lattice relaxation time must be short compared to the rise time of the heating pulse which is of the order of 2 ns. For the plateau region the rise of the spin temperature was found to be 170 K. Accordingly the lattice temperature rise in Fig. 10 can be calibrated in

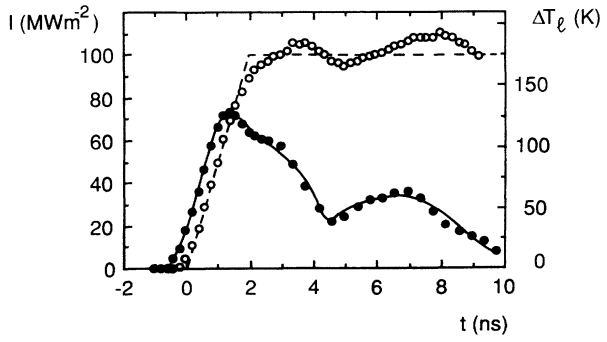

FIG. 10. Full circles: intensity of the heating laser pulse of measurement [9(a)] (left-hand scale) as a function of time. Using the solid line as a fit through the filled circles the temperature rise of the lattice $\Delta T_{\text{lattice}}$ induced by the heating laser is calculated. $\Delta T_{\text{lattice}}$ is displayed by the open circles (right-hand scale).

degrees Kelvin. Solving the thermal diffusion equation with the material parameters of the iron substrate[17] yields a maximum temperature rise (ΔT_{max}) of 85 K. With the material parameters[17] of Gd ΔT_{max} becomes 405 K. Experimentally 170 K is observed. This is reasonable since the thickness of the Gd film is smaller than the penetration depth of the light. Using the parameters of Gd or Fe only changes ΔT_{max} but not the temporal behavior of the temperature rise shown in Fig. 10.

Time-resolved measurements performed at two different heating pulse intensities are plotted in Fig. 11 on an extended t scale. For Fig. 11(b) the heating pulse energy is twice as large as for 11(a). Using these two measurements we next derive a numerical value for the spin-lattice relaxation time. The rate equation for the temperature transfer between the spin system and the lattice is[18]

$$C_{spin}\frac{dT_{spin}}{dt}=G(T_{lattice}-T_{spin}) . \quad (2)$$

C_{spin} is the specific heat of the spin system and G is the phonon-magnon coupling constant. The characteristic time for the equilibration of the temperature is then given by

$$\tau_{sl}=C_{spin}/G . \quad (3)$$

C_{spin} (not G) depends on temperature. However, in the following τ_{sl} is understood to be a T-independent, averaged (for $45<T<225$ K) quantity which makes it possible to solve Eq. (2) analytically. Guided by Fig. 10, the lattice temperature is taken to increase linearly in time,

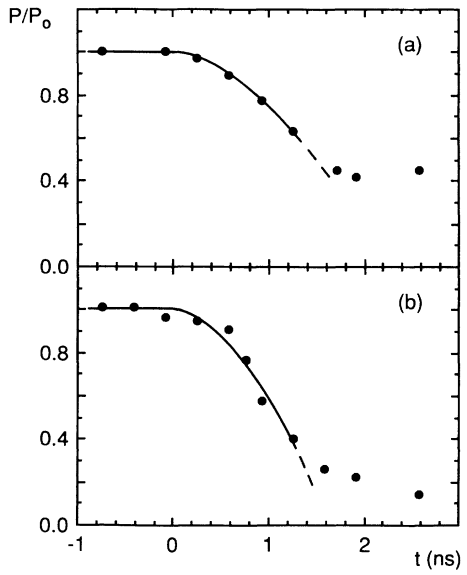

FIG. 11. Pump-probe experiment using a 60 ps (3.2 eV) laser pulse as a probing pulse and a 10 ns (2.15 eV) laser pulse as a heating pulse. For measurement (b) the heating pulse energy is twice as large as for measurement (a). The reduced spin polarization of the photoelectrons emitted by the probing pulse is plotted as a function of the time delay between the probing and the heating pulse. Zero time delay corresponds to the onset of the lattice heating. Solid lines: calculated $P(t)$ curves involving no adjustable parameters, see text.

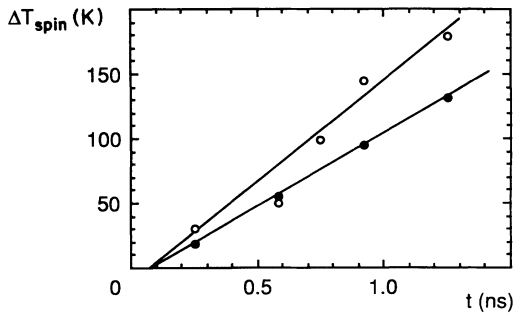

FIG. 12. $\Delta T_{spin}(t)$ derived from Fig. 7 for the data of Fig. 11(a) (solid circles) and Fig. 11(b) (open circles). The straight lines correspond to the fit $\Delta T_{spin}(t)=q(t-\tau_{sl})$. The open circle data point at 0.60 ns deviates from the straight line as does the corresponding measured polarization value in Fig. 11(b) from the $P(t)$ curve.

$T_{lattice}=T_0+qt$, where T_0 is the initial temperature at $t\leq 0$ and q is the rate of the temperature increase. Then, the solution of Eq. (2) is

$$T_{spin}(t)=T_0+q[t-\tau_{sl}(1-e^{t/-\tau_{sl}})] . \quad (4)$$

Again, as for Fig. 9(b) $\Delta T_{spin}(t)=T_{spin}(t)-T_0$ is derived exclusively from experimental data: Fig. 11 gives the polarization at a time t after the lattice heating has started. The corresponding increase $\Delta T_{spin}(t)$ of the spin temperature is obtained from Fig. 7 and is plotted in Fig. 12 for both measurements. Equation (4) shows that $\Delta T_{spin}(t)=q(t-\tau_{sl})$ for $t>\tau_{sl}$. Indeed, this linear relationship is found in Fig. 12. Extrapolation of the straight lines to $\Delta T_{spin}=0$ gives, independent of q, an intersection with the time axis at $t=\tau_{sl}$. In this way τ_{sl} is found and it amounts to 100±80 ps for both measurements shown in Fig. 11.[19] Note that the assumption $t>\tau_{sl}$ is satisfied in the extrapolation shown in Fig. 10. Since $\Delta T_{lattice}$ and ΔT_{spin} reach their constant values at comparable delay times (~1.5 ns) [see Fig. 9(b) and Fig. 10] one knows that $\tau_{sl}\ll 1.5$ ns.

The zero of the time scale is obtained from a $P(t)$ measurement at high heating pulse energy. It must lie between the last point where $P/P_0=1$ and the first point where $P/P_0<1$. The error in τ_{sl} of ±80 ps is mainly due to the uncertainty of the zero point of the time scale.

The rate of the temperature increase is given by the slope of the straight lines in Fig. 12; it is $q=115$ K/ns and $q=158$ K/ns for the measurements of Figs. 11(a) and 11(b), respectively.

Using these experimentally determined values of τ_{sl} and q together with the $P(T)$ relation from Fig. 7, the $P(t)$ curves are obtained immediately using Eq. (4). The result is shown as the solid lines in Fig. 11. Evidently the fit, without any adjustable parameter, is perfect.

IV. CONCLUSION

We have described an experiment capable of probing the time evolution of nonequilibrium magnetic states.

The technique is called laser-induced time-resolved photoemission. In a single pulse experiment the time resolution is mainly given by the duration of the laser pulse used for the photoemission.[20] This type of experiment is demonstrated to be suitable for the determination of upper and lower limits for the spin-lattice relaxation time. For iron τ_{sl} is found to be between 30 ps and 20 ns. In a more sophisticated experimental arrangement different laser pulses are used for the heating and for the photoemission. Then the time resolution is no longer given by the laser pulse duration but by the accuracy with which the beam delay distance can be varied. It is found that τ_{sl} for ferromagnetic gadolinium is 100 ± 80 ps.

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[1]M. B. Agranat, S. I. Ashitkov, A. B. Granovskii, and G. I. Rukman, Zh. Eksp. Teor. Fiz. **86**, 1376 (1984) [Sov. Phys. JETP **59**, 804 (1984)].

[2]J. G. Fujimoto, J. M. Liu, E. P. Ippen, and N. Bloembergen, Phys. Rev. Lett. **53**, 1837 (1984).

[3]H. E. Elsayed-Ali, T. B. Norris, M. A. Pessot, and G. A. Mourou, Phys. Rev. Lett. **58**, 1212 (1987).

[4]R. W. Schoenlein, W. Z. Lin, J. G. Fujimoto, and G. L. Eesley, Phys. Rev. Lett. **58**, 1680 (1987).

[5]M. Campagna, D. T. Pierce, F. Meier, K. Sattler, and H. C. Siegmann, Adv. Electron. Electron Phys. **41**, 113 (1976).

[6]G. L. Bona, F. Meier, G. Schönhense, M. Aeschlimann, M. Stampanoni, G. Zampieri, and H. C. Siegmann, Phys. Rev. B **34**, 7784 (1986); F. Meier, A. Vaterlaus, M. Aeschlimann, M. Lutz, D. Guarisco, F. Milani, and H. C. Siegmann, J. Magn. Magn. Mater. **91**, 314 (1991).

[7]G. L. Bona, Ph.D. dissertation, ETH Zürich, 1987 (unpublished); M. Aeschlimann, Ph.D. dissertation, ETH Zürich, 1989 (unpublished).

[8]H. Alder, M. Campagna, and H. C. Siegmann, Phys. Rev. B **8**, 2075 (1973).

[9]Jeff F. Young, J. E. Sipe,and H. M. van Driel, Phys. Rev. B **30**, 2001 (1984).

[10]S. E. Clark and D. C. Emmony, Phys. Rev. B **40**, 2031 (1989).

[11]F. Meier and D. Pescia, in *Optical Orientation,* edited by F. Meier and B. Zakharchenya (North-Holland, Amsterdam, 1984), p. 295ff.

[12]D. Pescia, M. Stampanoni, G. L. Bona, A. Vaterlaus, R. F. Willis, and F. Meier, Phys. Rev. Lett. **58**, 2126 (1987).

[13]A. Cerri, D. Mauri, and M. Landolt, Phys. Rev. B **27**, 6526 (1983).

[14]James J. Rhyne, in *Magnetic Properties of Rare Earth Metals,* edited by R. J. Elliot (Plenum, London, 1972), p. 132.

[15]The temporal intensity profile of the heating pulse has been measured using an ITL TF 1850 vacuum photodiode (rise time 100 ps) and a Tektronix 2467 350 MHz oscilloscope.

[16]J. H. Bechtel, J. Appl. Phys. **46**, 1585 (1975).

[17]The material parameters of Fe (Gd) are taken at 140 K and assumed to be temperature independent: thermal conductivity 1.14 W/cm K (0.14 W/cm K), thermal diffusivity 0.26 cm^2/s (0.08 cm^2/s), optical absorption coefficient for the wavelength of the heating laser 6.59×10^5 cm^{-1} (5.2×10^5 cm^{-1}), surface reflectivity 0.52 (0.46). The laser parameters are $h\nu=2.15$ eV, pulse energy 67.6 μJ (heating pulse energy of measurement 11a), focus diameter (FWHM) 390 μm, temporal intensity profile, see solid line in Fig. 10.

[18]S. I. Anisimov, B. L. Kapeliovich, and T. L. Perel'man, Zh. Eksp. Teor. Fiz. **66**, 776 (1974) [Sov. Phys. JETP **39**, 375 (1974)].

[19]From Fig. 12 the duration of the linear increase of the lattice temperature is found to be 1.5 ns whereas from Fig. 10 this time interval appears to be rather 1.8 ns. The reason for the different times is instrumental broadening of the measurements in Fig. 10; see Ref. 16.

[20]A. Vaterlaus, D. Guarisco, M. Lutz, M. Hecht, and F. Meier (unpublished).

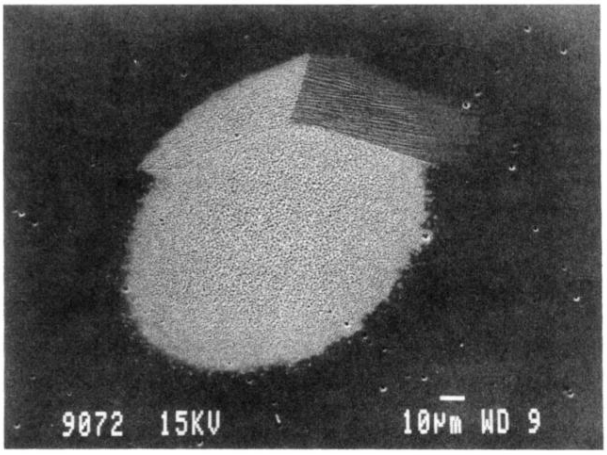

FIG. 3. Scanning electron microscope picture of the polycrystalline iron surface which has been irradiated with 400 30 ps laser pulses having an energy of 94 ± 10 μJ. The laser-damaged area has an averaged radius of 69 ± 5 μm.