Saturation of the ultrafast laser-induced demagnetization in nickel

D. Cheskis, A. Porat, L. Szapiro, O. Potashnik, and S. Bar-Ad
School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel
(Received 12 April 2005; published 15 July 2005)

We study the loss of magneto-optic contrast in nickel, in response to excitation by a short laser pulse, as a function of excitation density. Our measurements in the picosecond and subpicosecond regime show saturation of the contrast loss at high excitation densities. We conclude that spin relaxation is incomplete on this time scale, and explain the loss of magneto-optic contrast and its saturation in terms of a collapse of the Stoner gap due to band filling effects.

DOI: 10.1103/PhysRevB.72.014437 PACS number(s): 75.40.Gb, 78.20.Ls, 75.70.−i, 78.47.+p

The ultrafast dynamics of itinerant ferromagnets following an excitation with a short laser pulse has been the subject of an extensive debate in the last decade.[1–3] The main question is whether the magneto-optical (MO) response seen in experiments on subpicosecond time scales indeed reflects an ultrafast demagnetization process, or simply a change of the dielectric tensor elements due to band-filling effects. Before the mid-1990s it was generally believed that the spin relaxation time in itinerant ferromagnets is of the order of 100 ps, and is driven by spin-lattice relaxation.[4] A much faster loss of MO contrast in ferromagnetic nickel, on time scales of a few picoseconds, was measured for the first time in 1996 in a pump-probe experiment that employed the Magneto-Optical Kerr Effect (MOKE).[5] The loss of MO contrast was interpreted as a heating-induced demagnetization of the nickel, implying a gradual rise of the spin temperature within $\approx$3 ps. The surprisingly fast response demonstrated in that experiment has motivated several research groups to investigate this process in more detail, using various pump-probe techniques that rely on the MOKE,[5–8] magnetization-dependent surface second harmonic generation (SSHG),[9–11] and time-resolved photoemission (TRP).[12,13] The various experiments differed in detail of the samples and the time scales of the response, with the surface-sensitive SSHG experiments tending to show both a faster loss of MO contrast and a faster recovery. Nevertheless, all experiments showed a subpicosecond response. Regarding the nature of the initial, nonequilibrium situation, the SSHG experiment of Hohlfeld *et al.*[9] has shown that following thermalization of the electrons (i.e., for time delays larger than $\approx$300 fs between the pump and probe pulses) the transient magnetization is governed by the electron temperature T_e. However, strong deviations of the data from the equilibrium magnetization curve were observed for shorter time delays, suggesting that the demagnetization was not heating-induced. These findings have led to theoretical models that explained the ultrafast laser-induced demagnetization as a cooperative effect of the strong laser field and weak spin-orbit coupling.[14] TRP experiments supported the hypothesis of ultrafast demagnetization by showing evidence for a collapse of the Stoner gap on subpicosecond time scales.[13] However, the interpretation of the fast loss of MO contrast as a true demagnetization was challenged by detailed MOKE experiments,[7] which showed that pure optical processes also contribute to the magneto-optical response of the sample, and claimed that during the first hundreds of femtoseconds the MO response is dominated by band-filling effects. Similar claims challenge the data from SSHG experiments.[11] These claims have in turn been contested by more recent experiments.[8,15]

From the above discussion it can be understood that the issue of demagnetization in ferromagnetic thin films, following excitation by a short laser pulse, is still highly controversial, and there is disagreement between different authors with respect to the demagnetization time, the underlying processes, and even the interpretation of experimental data. To resolve the controversy it is essential to discover if the changes of the MO contrast actually reflect a net change of the magnetization, i.e., a net change of the difference of majority and minority spin populations. A careful analysis of the data from time-resolved magneto-optic experiments as a function of excitation density can be an important guideline for answering this question. In particular, a true magnetic response and an optical band-filling effect will show different dependencies on excitation density; while a true magnetic response will monotonically increase with excitation density, until a complete demagnetization is reached, a magneto-optical response due to band filling effects, which is not accompanied by complete spin relaxation, will eventually saturate. Such a detailed analysis has been carried out by Hohlfeld *et al.* in their experiment on nickel,[9] but unfortunately complete demagnetization was not attempted. In fact, complete demagnetization on subpicosecond time scales has never been observed *in nickel*. Conversely, complete demagnetization was reported in experiments on $CoPt_3$,[6] but the detailed behavior as a function of excitation density was not reported.

In this paper we present a comprehensive experimental study of the magneto-optical response of nickel as a function of excitation density, in the picosecond and subpicosecond regime. By analyzing the behavior of the apparent magnetization and coercivity in time-resolved MOKE measurements, we show that the loss of MO contrast at short time delays saturates at high excitation densities, while the temperature attained by the sample, at much longer time delays, keeps increasing. This behavior can only be understood in terms of the band-filling effect. Our interpretation is supported by a quantitative analysis of the excitation densities in comparison to the known band structure and specific heat data of nickel. This allows us to associate the observed saturation with the complete collapse of the Stoner gap during the elec-

tronic thermalization. From the observation that a remanent magnetization persists at high excitation densities we conclude that spin relaxation is incomplete on the time scale of the experiment.

The source of short pulses in the experiment is a Ti:Sapphire regenerative amplifier that generates ≈70 fs pulses at ≈800 nm (a photon energy ≈1.5 eV) at a repetition rate of 1 kHz. These pulses are employed in a standard time-resolved pump-probe configuration. Variable neutral density filters are used to control the power of the pump beam, with the pump:probe power ratio varying between ≈500:1 and ≈50:1. A 3:1 beam expander is used to decrease the spot size of the probe beam on the sample relative to that of pump beam, in order that the probe samples a spot with homogeneous excitation. A removable mirror allows characterization and optimization of the pulse autocorrelation at the plane of the sample, and compensation for chirp in glass elements of the setup is obtained by small adjustments of the pulse compressor in the regenerative amplifier. A half-wave plate and a polarizer are used to adjust the polarization of the probe beam: a pump and a probe with perpendicular linear polarizations are used for time-resolved differential reflection (DR) measurements, while parallel linear polarizations are employed for the longitudinal MOKE measurements. In the MOKE measurements the probe beam reflected from the sample passes through a standard analyzer, oriented perpendicular to the incoming probe polarization. The use of parallel linear polarizations in the MOKE measurements allows effective blocking of scattering of the pump beam from the sample, and further suppression is achieved by mechanically chopping the probe beam in combination with lock-in detection. This arrangement eliminates the possibility of residual, magnetic field-dependent scattering of the pump beam into the detector. The samples are ≈30 nm thick films of polycrystalline nickel, deposited in a standard evaporator on sapphire and glass substrates. The experiment is carried out at room temperature, using an electromagnet with Helmholtz coils and a maximum field of ≈50 Öersteds. When measured as function of the external magnetic field, the MOKE signal forms a hysteresis curve. As the field is increased beyond the coercive field and towards its maximum (positive or negative) value, the hysteresis curves that we measure become field independent, which allows us to identify the MOKE signal at maximum field with the spontaneous magnetization of the sample, M_s. In addition to M_s we also extract from each hysteresis curve the "effective" coercivity $\tilde{H}_c(I)$, i.e., the external field required to reduce the magnetization to zero *in the presence of pump excitation with fluence I*.

Figure 1 plots the values of M_s that we have obtained from a large set of hysteresis curves, measured as a function of laser fluence and time delay, on a sample grown on a glass substrate. The data points for time delays of 100 fs, 150 fs, 300 fs, 1 ps, and 3 ps, normalized to the value of M_s obtained with the pump beam blocked, M_0, are plotted as a function of the pump beam fluence. For clarity we have removed the error bars, and a typical error bar is displayed separately. On first impression the data appears to be noisy, but closer examination reveals several obvious and systematic trends. In particular, a clear difference is seen between

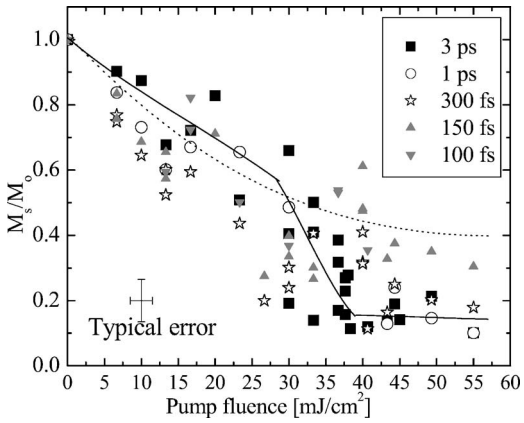

FIG. 1. The normalized spontaneous magnetization as function of laser fluence for several time delays in the picosecond and subpicosecond regime. The lines are guides to the eye: dotted, short time delays; solid, long time delays.

the temporal behavior which is observed at low pump fluences (below 20 mJ/cm^2) and that observed at high pump fluences (above 35 mJ/cm^2): At low fluences the MOKE response peaks at ≈300 fs, immediately after the excitation is over, and gradually recovers at later time delays (1 ps and 3 ps). On the other hand, at high fluences the MOKE response keeps increasing long after the excitation is over. Exactly the same behavior is observed in measurements on different samples, grown on both glass and sapphire substrates. In particular, the values of M_s are the same, and the transition from the low-intensity behavior to the high-intensity behavior occurs at the same excitation density. We note that these observations are in excellent correlation with the changes that we observe in the DR measurements, as the excitation density is increased. As shown in Fig. 2, the rise of the DR signal also extends to later time delays as the pump fluence increases. This correlation between the magnetic (MOKE) and nonmagnetic (DR) signals already suggests that the observed dynamics of the MOKE signal are not of pure magnetic origin. Furthermore, our observations allow us to draw the lines that serve as guides to the eye in Fig. 1, and depict the general trends outlined above.

The most significant feature of the data in Fig. 1 is the clear saturation of the MOKE response at high pump fluences: while the MO contrast gradually decreases with in-

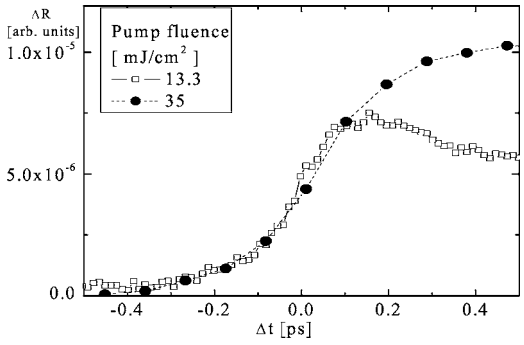

FIG. 2. The differential reflection signal as function of the time delay, for two pump fluences: 13.3 mJ/cm^2 and 35 mJ/cm^2.

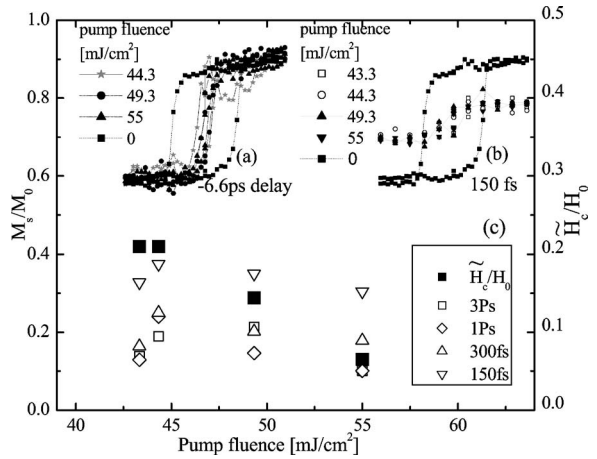

FIG. 3. The normalized spontaneous magnetization and effective coercivity at high pump fluences: (a) Hysteresis curves measured at a time delay of −6.6 ps, from which $\tilde{H}_c(I)$ is deduced; (b) Hysteresis curves at a time delay of 150 fs from which M_s is deduced; (c) M_s and $\tilde{H}_c(I)$ as a function of pump fluence.

creasing time delay, there is no apparent decrease as function of pump fluence beyond ≈ 35 mJ/cm^2, for any of the time delays. Indeed, in all of our measurements in the picosecond and subpicosecond regime we observe a residual MO contrast at the maximum attainable pump fluence, and attempts to increase the latter beyond ≈ 60 mJ/cm^2 invariably result in irreversible damage to the sample (probably due to melting, which occurs at considerably longer time delays). The saturation of the MOKE response at high pump fluences is demonstrated in more detail in Fig. 3. Figure 3(a) shows a set of 3 hysteresis curves, measured with 3 (increasing) pump fluences at a time delay of −6.6 ps, superimposed on the hysteresis curve measured with the pump beam blocked. These measurements correspond to an actual time delay of ≈ 1 ms (the period of the laser system), and therefore M_s recovers to M_0. On the other hand $\tilde{H}_c(I)$ decreases monotonously with increasing pump fluence. This decrease reflects a heat-assisted magnetization switching process that occurs at a considerably longer time delay,[16] when the temperature of the nickel may be assumed to be well-defined. We therefore use $\tilde{H}_c(I)$ (which we extract from the data for negative time delay) as a measure of the actual excitation density and of the maximum temperature attained by the sample in each excitation cycle. The values of $\tilde{H}_c(I)$, normalized to the coercivity measured with the pump beam blocked, are plotted as full squares in Fig. 3(c) [note that one of these data points was omitted from Fig. 3(a) for the sake of clarity]. The decrease of $\tilde{H}_c(I)$ as a function of pump fluence verifies that the actual excitation density has indeed increased. In contrast, the loss of MO contrast saturates, as the normalized values M_s/M_0 in Fig. 3(c) show [these were analyzed from the hysteresis curves that we measured at *positive* time delays, such as the curves for $\Delta t = 150$ fs shown in Fig. 3(b)]. Figure 3(c) therefore clearly demonstrates that at high fluences the MO contrast no longer decreases as the excitation density is increased.

The observed dependence of the MO contrast on pump

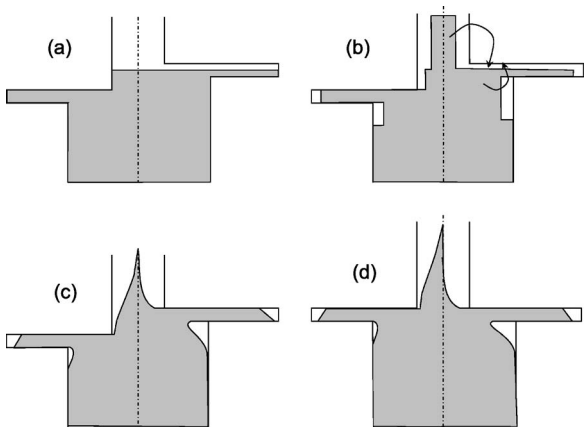

FIG. 4. An illustration of the thermalization-induced collapse of the Stoner gap discussed in the text. The bands are shown schematically, with the highest peak corresponding to the *d* band which is partially filled in equilibrium.

fluence can only be understood in terms of band filling effects, i.e., the changes of occupation of the nickel energy bands that result from the optical excitation and from the ensuing carrier dynamics. To illustrate these effects we refer to the band structure of nickel, where the density of states near the Fermi level is dominated by a completely filled majority-spin *d* band and a partially-filled minority-spin *d* band.[17] In the simplest form of the Stoner model,[18] the magnetization is proportional to the difference of electron densities in these two *d* subbands, and to the Stoner gap that separates the two. In thermal equilibrium at zero temperature there are 0.54 holes per nickel atom in the minority-spin *d* band.[19] In the following we explain how the optical excitation may result in the filling of these holes, leading to an instantaneous decrease of the Stoner gap, accompanied by an instantaneous change of the MO contrast. The explanation implies that at sufficiently high pump fluences the fillings of the two *d* subbands become comparable, and the Stoner gap collapses completely. At this point there can be no further change of the instantaneous response, and the MO contrast saturates at a value determined by the net (nonequilibrium) spin polarization. Our argument focuses on the dynamics of minority-spin states, but may be generalized to include majority-spin states. The equilibrium state is depicted in Fig. 4(a). The effect of the pump beam is the excitation of electrons with energies of up to 1.5 eV below the Fermi level to states with energies of up to 1.5 eV above the Fermi level. This is shown schematically in Fig. 4(b). These photoexcited electrons then thermalize, within ≈ 0.5 ps.[13] When considering the dynamics of minority-spin states, it is reasonable to assume that the thermalization process is dominated by scattering into empty states in the minority-spin *d* band, due to its high density of states [see arrows in Fig. 4(b)]. Another plausible assumption is that the thermalization process is dominated by spin-conserving elastic scattering events, in which every photoexcited electron gives its excess energy to several electrons just below the Fermi level. While this may result in scattering of electrons *out* of the partially-filled minority-spin *d* band, many other electrons would scatter *into* that band from lower-lying states. A quantitative analy-

sis of these competing processes is beyond the scope of this paper. However, the proximity of the minority-spin *d* band to the Fermi level suggests that the actual number of electrons that scatter into the minority-spin *d* band is significantly larger than the number of photoexcited electrons. In combination with partial depletion of the majority-spin *d* band, this may result in the complete suppression of the population difference of the two *d* subbands [Fig. 4(c)], and a collapse of the Stoner gap [Fig. 4(d)]. Note that the total difference between the number of majority and minority spins remains unchanged, but is distributed differently among the bands [see Fig. 4(d)], leading to the apparent demagnetization observed in the experiment. A quantitative analysis of the pump fluence at which saturation is observed supports this hypothesis: The saturation is reached at a pump fluence of ≈ 35 mJ/cm^2. With an absorption coefficient of 0.4, a photon energy of 1.5 eV, a beam diameter of 200 μm, a sample thickness of 30 nm, and the known lattice parameters of nickel, this fluence translates to an excitation of ≈ 0.15 electrons per nickel atom. This is lower than the number of holes per atom in minority *d* band (0.54), and while it shows that the density of photoexcited electrons by itself is insufficient to completely block the empty states in the minority-spin *d* band, it also suggests that the redistribution of energy during thermalization can easily increase this blocking. It is also interesting to compare the energy density needed to achieve this blocking to the energy density which is necessary to elevate the sample's equilibrium temperature to the Curie point. Using the above experimental parameters and an average *total* specific heat of ≈ 0.14 cal/g K,[20,21] we find that a pump fluence of 35 mJ/cm^2 is sufficient to elevate the sample's temperature by almost 900 K (which is in agreement with the melting of the sample at ≈ 60 mJ/cm^2). Moreover, the same fluence would result, in the absence of heat transfer to the lattice, in significantly higher electron temperatures. The fact that a MO contrast is still measured under such conditions on one hand shows that analysis of the experimental data in terms of heat-induced demagnetization is unjustified, but at the same time demonstrates that even partial thermalization of the electrons can indeed result in complete collapse of the Stoner gap. We emphasize that the complete collapse of the Stoner gap that we predict at high excitation densities is not contradictory to the partial collapse and recovery reported in TRP measurements at somewhat lower densities.[13] In fact, the data in Fig. 1 does show that at lower excitation densities the magnetization recovers after 3 ps. We claim, however, that even spin-conserving processes can result in the apparent demagnetization (in contrast to the explanation suggested in Ref. 12).

In the scenario presented above, the residual MO contrast that we observe at high pump fluences is related to the assumption that the thermalization process is dominated by spin-conserving scattering events, and implies that spin-relaxation is incomplete on the time scale of our experiment (in agreement with Ref. 12). The conclusion from the results and discussion presented above is that the ultrafast magneto-optic response of nickel to short pulse laser excitation is a demagnetization effect only in the sense of collapse of the Stoner gap, but not in the sense of spin relaxation. This means that following the thermalization process, the non-equilibrium situation still involves different populations of majority and minority spins, but the distribution of the spins among the bands is different from that in equilibrium. In particular, this nonequilibrium situation cannot be simply described in terms of a magnetic temperature. This conclusion, which is based on our new experimental evidence, is in agreement to earlier indications to the role played by band-filling effects in experiments on nickel.[7] It is seemingly in contradiction with the experimental data for $CoPt_3$,[6,15] but this may be due to different spin relaxation times in the two materials. Further experimental work is required to confirm this point.

This research was supported by the Israel Science Foundation. We are thankful to A. Gerber, M. Karpovski, and R. Bron for their assistance and advice.

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