Saturation of the ultrafast laser-induced demagnetization in nickel

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(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: 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. 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. 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). The loss of MO contrast was interpreted as a heating-induced demagnetization of the nickel, implying a gradual rise of the spin temperature within 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, magnetization-dependent surface second harmonic generation (SSHG), and time-resolved photoemission (TRP). 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. has shown that following thermalization of the electrons (i.e., for time delays larger than 300 fs between the pump and probe pulses) the transient magnetization is governed by the electron temperature Te. 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. TRP experiments supported the hypothesis of ultrafast demagnetization by showing evidence for a collapse of the Stoner gap on subpicosecond time scales. However, the interpretation of the fast loss of MO contrast as a true demagnetization was challenged by detailed MOKE experiments, 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. These claims have in turn been contested by more recent experiments.

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, 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, 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 Oersteds. 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 forms a hysteresis curve. As the field is increased beyond the coercive field and towards its maximum forms a hysteresis curve. As the field is increased beyond the coercive field and towards its maximum forms a hysteresis curve. As the field is increased beyond the coercive field and towards its maximum forms a hysteresis curve. As the field is increased beyond the coercive field and towards its maximum forms a hysteresis curve. As the field is increased beyond the coercive field and towards its maximum forms a hysteresis curve.

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 increasing pump fluence, the temporal behavior which is observed at low pump fluences (below 20 mJ/cm²) and that observed at high pump fluences (above 35 mJ/cm²): 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 $M_z$ 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.

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increasing time delay, there is no apparent decrease as function of pump fluence beyond ≈35 mJ/cm², for any of the time delays. Indeed, in all of our measurements in the picosecond and subpicosecond regime we observe a residual MO contrast no longer decreases as the excitation density is increased.

The observed dependence of the MO contrast on pump 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. In the simplest form of the Stoner model, 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. 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. 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 anal-
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 \( \sim 35 \text{ mJ/cm}^2 \). With an absorption coefficient of 0.4, a photon energy of 1.5 eV, a beam diameter of 200 \( \mu \text{m} \), a sample thickness of 30 nm, and the known lattice parameters of nickel, this fluence translates to an excitation of \( \sim 0.15 \) 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 \( \sim 0.14 \text{ cal/g K}^{20,21} \) 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 \( \sim 60 \text{ mJ/cm}^2 \)). 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 suggests that the actual number of electrons in the Fermi level is lower than the number of holes in the minority-spin band to \( \sim 0.54 \) electrons per atom in minority spin. This is lower than the number of photoexcited electrons, but is sufficient to completely block the empty states in the minority-spin d band.

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.