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# Ultrafast Quenching of the Antiferromagnetic Order in FeBO<sub>3</sub>: Direct Optical Probing of the Phonon-Magnon Coupling

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The dynamics of the optically induced phase transition from the antiferromagnetic to the paramagnetic state in FeBO<sub>3</sub> is observed using a pump-probe magneto-optical Faraday technique employing 100 fs laser pulses. At the pump energy of 1.55 eV phonon-assisted transitions dominate in the absorption of light and ultrafast heating of the lattice occurs. The quenching of the magnetic order is caused by an increase of the magnon temperature due to energy transfer from the heated lattice. The heating time of the magnon system is around 700 ps, which is a factor of 20 faster than previously reported phonon-magnon interaction times.

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The fundamental and practical limits on ultrafast magnetization dynamics are a challenging area in the physics of magnetism and a subject of vital importance for the newly developing area of spin electronics. The development of femtosecond lasers has opened the way to create external stimuli as optical and magnetic field pulses that are shorter than fundamental time scales such as spin-lattice relaxation or precession times, which can be used for extremely fast manipulation of the magnetic state of a medium [1]. So far ultrafast optical spectroscopy has been successfully applied for the study of optically induced spin polarization in semiconductors [2] and magnetization dynamics in metals [3–7] and half-metals [8]. This has resulted in some surprising observations such as a quenching of the ferromagnetic order on a time scale much faster than the spin-lattice relaxation time [4–6]. The interpretation of these exciting results is still subject to debate and is sometimes hampered by experimental artifacts that are not related to the order parameter dynamics [7]. To improve our understanding of the magnetization dynamics at very short time scales, better controlled experimental conditions are desirable. In that respect, antiferromagnetic dielectrics are promising candidates because of the localized character of their magnetic moments. Moreover, the wide use of antiferromagnets as a pinning or spacer layer in modern electronic devices such as magnetoresistive random access memories, is responsible for the broad interest in the quenching dynamics of antiferromagnetism. Such studies are especially interesting after the prediction of demagnetization times shorter than 100 fs in nickel oxide NiO [9]. However, neither Kerr nor Faraday effect can be used to monitor the magnetization dynamics in antiferromagnetic NiO in which both these effects vanish. In principle, with second harmonic generation one could monitor the spin dynamics in antiferromagnetic oxides [10], but the

complex character of their domain structure has so far prevented the obtaining of reliable experimental data with an adequate interpretation. The ultrafast dynamics of nonequilibrium spin waves was studied in the antiferromagnetic dielectric Cr<sub>2</sub>O<sub>3</sub> employing a differential transmission technique [11] but the photoinduced demagnetization in this material was not considered.

In this Letter we report the quenching of antiferromagnetic order in iron borate FeBO<sub>3</sub>, an oxide possessing weak ferromagnetism and a large Faraday rotation. One expects the mechanism of the photo-induced demagnetization in dielectric iron borate to be different from that in metallic nickel [3–7] and half-metallic strontium-iron molybdate [8], and also from that in dielectric chromium oxide [11]. In the case of metals and half-metals the photoexcitation directly heats up the electronic system, whereas in chromium oxide direct heating of the magnon system was achieved via pumping at the energy of the exciton-magnon transition. In the case of FeBO<sub>3</sub> photoexcitation is expected to first lead to an increase in the lattice temperature that is subsequently followed by an increase in the spin temperature via phonon-magnon coupling. We show that our experiment can directly probe this coupling and that the latter is 20 times faster than expected from previous results [12].

Iron borate crystallizes in a calcite type structure with space group  $R\bar{3}c$  and has a Néel temperature  $T_N \approx 348$  K. The antiferromagnetic state of FeBO<sub>3</sub> is characterized by a weak ferromagnetism due to a slight spin canting of about 1° in the (001) plane, which results in a magnetic moment oriented within this plane. Because of this magnetic moment one can turn the FeBO<sub>3</sub> sample into a single domain state with the help of a small magnetic field and linear magneto-optical effects can be used to probe the antiferromagnetic ordering. However, the mechanisms of photoinduced spin excitation and relaxation should be the

same in iron borate and nickel oxide due to the similarity of their electronic structures and optical properties. Moreover,  $\text{FeBO}_3$  has the Néel point slightly above room temperature and is characterized by good transparency in the visible spectral range. Therefore this compound is suitable for both study and potential applications.

The optical properties of  $\text{FeBO}_3$  are determined by the  $d-d$  transitions in the partially filled  $d$  envelope of the  $\text{Fe}^{3+}$  ion. Figure 1 shows the relevant part of the electronic energy diagram as derived from the local symmetry of the  $\text{Fe}^{3+}$  ion. In the crystalline field, the  $3d^5$  electrons of  $\text{Fe}^{3+}$  ions occupy the ground state  ${}^6\Gamma_1^+$  ( $S = 5/2$ ), which is an orbital singlet and the only spin sextet state. The lowest excited state is triplet  ${}^4\Gamma_4^+$  ( $S = 3/2$ ). The spin degeneracy is lifted due to the spin-orbit coupling and exchange interaction.

The transition  ${}^6\Gamma_1^+ \rightarrow {}^4\Gamma_4^+$  is centered at 1.4 eV and is forbidden in the electric dipole approximation because of the selection rules imposed on parity and spin. Nevertheless, from the absorption spectra measured at 20 K four intensive spectral lines were distinguished near the first  $d-d$  transition (see Fig. 1). At higher temperatures the splitting is not seen because of increased electron-phonon interaction and phonon-assisted transitions.

The studied sample was a plate of thickness  $300 \mu\text{m}$ . The measurements were performed in a pump and probe configuration at a photon energy of 1.55 eV using amplified 100 fs pulses from a Ti:sapphire laser at a repetition rate of 1 kHz. The pump and probe beams were linearly polarized with an intensity ratio of 10 : 1. Both beams were focused on the sample to a spot diameter of about 1 mm for the pump and 0.2 mm for the probe beam. The pump fluence on the sample was around  $30 \text{ mJ/cm}^2$ . The incident angles for the pump and probe were chosen as 0 and  $20^\circ$ , respectively. In this geometry the probe is affected by the magneto-optical Faraday effect, which is proportional to the inplane magnetization of  $\text{FeBO}_3$ . The sample was placed in an inplane ac magnetic field with an

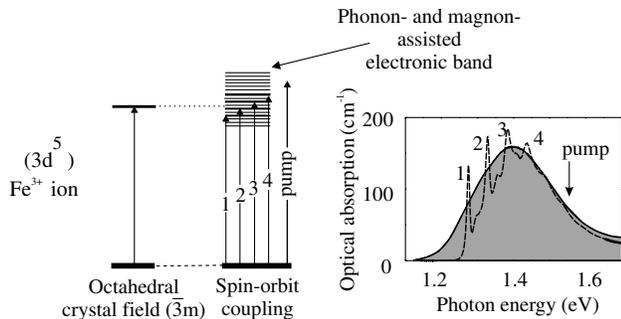


FIG. 1. The energy level scheme and the absorption spectra of  $\text{FeBO}_3$  at room temperature (solid line) and at 20 K (dashed line). The spectral lines from 1 to 4 correspond to the transitions from the ground state  ${}^6\Gamma_1^+$  to the excited state  ${}^4\Gamma_4^+$  split by the spin-orbit coupling.

amplitude of  $\approx 100 \text{ Oe}$  and a frequency of 194 Hz. The Faraday rotation was measured at this frequency with a sensitivity as high as 0.2 mrad employing a two photodiode balanced detection scheme and a lock-in amplifier. The bias temperature of the sample  $T_{\text{bias}}$  could be varied with the help of a heater located on the sample holder.

The optical pump pulse influences both the magnetic and the optical properties of the excited medium. Because the output of the used detection scheme depends on the intensity of the probe beam, knowledge of the transient transmission is necessary. This was measured by using a single-diode scheme and an amplitude modulation of the pump beam.

We calibrated the measured time dependencies of the Faraday rotation by dividing them by the associated transient transmissions [shown in Fig. 2(a)]. The resulting data show a peak during the overlap of the pulses that is followed by a slow breakdown of the antiferromagnetic order [13]. The amplitude of the initial peak was found to be a linear function of the pump fluence as shown in Fig. 2(b). The slow component of the Faraday rotation as a function of bias temperature and time is shown in Fig. 3. The dynamical changes of the Faraday effect are smaller at lower bias temperatures, while at  $T_{\text{bias}} = 346.5 \text{ K}$  the Faraday rotation rapidly decreases until a delay of about 500 ps, where the signal vanishes.

The intrinsic Faraday effect is shown in Fig. 4 as a function of bias temperature. Since the Faraday rotation is proportional to the order parameter, its temperature dependence is generally given by [14]

$$\alpha_F(T) = \alpha_0 \left(1 - \frac{T_s}{T_N}\right)^\beta, \quad (1)$$

where  $T_N$  is the Néel temperature,  $\beta$  is the critical exponent, and  $T_s$  is the magnon temperature, which drives the order parameter. Fitting Eq. (1) to the corresponding measurements that are represented by solid squares in Fig. 4 we obtained  $\beta = 0.364 \pm 0.008$  and  $T_N = 347.0 \pm$

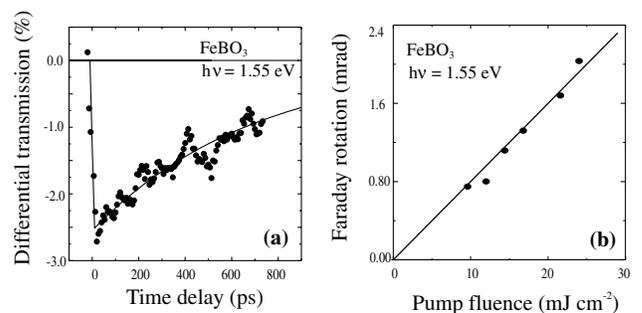


FIG. 2. (a) The transmission as a function of time delay, (b) the intensity dependence of the ultrafast Faraday rotation (symbols); linear fit with a slope of  $(8 \pm 0.6) \times 10^{-5} \text{ rad cm}^2/\text{mJ}$  (line).

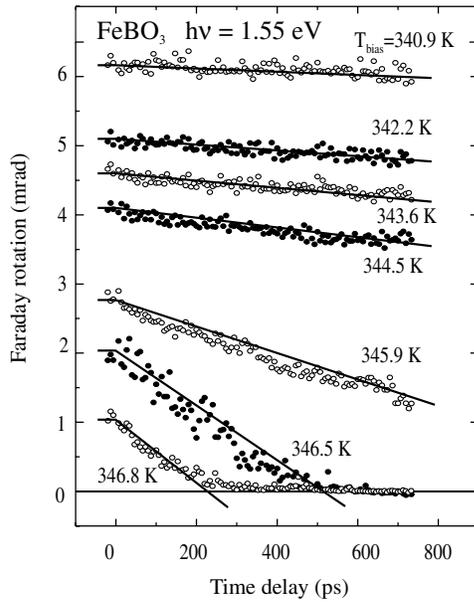


FIG. 3. The long term transient Faraday rotation measured as a function of the bias temperature. The antiferromagnetic order is destroyed at a time delay of 500 ps for  $T_{\text{bias}} = 346.5$  K.

0.1 K. These values are in good agreement with  $\beta = 0.354$  and  $T_N = 348.35$  K reported before [15].

Repeating the measurements at a negative delay of  $-20$  ps we obtained identical results but shifted about 10 K towards lower  $T_{\text{bias}}$ . This offset was due to heat accumulation caused by the repeated excitation of the sample. We minimized this effect by using the lowest possible repetition rate that yielded a reasonable signal-

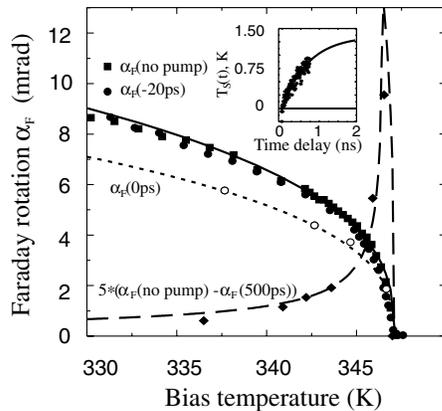


FIG. 4. The Faraday rotation without pump (solid squares), at negative (solid circles) and at zero (open circles) time delay as a function of the bias temperature with the fit to Eq. (1) (solid and dotted lines, respectively). The difference between the intrinsic magneto-optical signal and that at 500 ps is shown by rombs together with the calculation based on the fitted parameters (dashed line). The inset shows the transient component of the magnon temperature as a function of the time delay. The solid line is the fit according to Eq. (2).

to-noise ratio. The measured magnitude of the temperature offset was in good agreement with an estimate based on the optical and thermal properties of  $\text{FeBO}_3$  and was added to  $T_{\text{bias}}$  when plotting the relevant data.

Figure 4 also shows the difference between the intrinsic magneto-optical signal and that at a time delay of 500 ps. This difference increases drastically before dropping to zero at the Néel point. All these features strongly imply that the pump-induced relaxation of the magneto-optical signal is related to an increase of the magnon temperature. At a bias temperature  $T_{\text{bias}} = 346.5$  K and at a delay time of 500 ps the Néel point is reached and antiferromagnetism is broken.

The temperature dependence of the Faraday rotation at zero time delay is shown in Fig. 4 by open circles. The experimental data were fitted by Eq. (1) with  $\beta$  and  $T_N$  deduced from the previous fit and the result of the fit is shown by the dotted line. The similarity of the temperature behavior of the intrinsic Faraday rotation and that at zero time delay strongly suggests that no magnetic excitation occurs within 100 fs.

This result proves that the time-resolved data (cf. Fig. 3) are not directly affected by the optical excitation itself since the lifetime of the electrons in the excited state is shorter than 100 fs as deduced from the huge linewidth of the  ${}^6\Gamma_1^+ \rightarrow {}^4\Gamma_4^+$  transition. This estimate is justified by dramatic changes of the absorption spectrum with temperature in Fig. 1, the fact that the transition takes place between states with different electronic configurations  $(t_{2g})^3(e_g)^2 \rightarrow (t_{2g})^4(e_g)^1$  and earlier reports [16] which all show that this transition is accompanied by the excitation of optical phonons and that it is intrinsically broad. Possible minor contributions due to inhomogeneous broadening are negligible [16].

In order to derive information about the magnetization dynamics from the measured transient Faraday rotation  $\alpha_F(t)$  plotted in Fig. 3, we converted the latter into transient magnon temperatures  $T_s(t)$  by means of Eq. (1) for all data below  $T_N$ . Decomposing these temperatures into a static bias temperature  $T_{\text{bias}}$  and an optically induced transient component  $\Delta T_s(t)$ , we found all  $\Delta T_s(t)$  to be identical within the experimental error. Their average is shown in the inset of Fig. 4 and is characterized by a monotonic increase that was fitted by the function

$$\Delta T_s(t) = T_s^0 \left[ 1 - \exp\left(-\frac{t}{\tau_{sl}}\right) \right], \quad (2)$$

where  $T_s^0$  is the amplitude of the dynamical temperature and  $\tau_{sl}$  is the phonon-magnon interaction time. All the variables were set as fitting parameters and the result of the fit for  $T_s^0 = 1.4$  K and  $\tau_{sl} = 700$  ps is shown in the inset by a solid line.

Using the deduced parameters  $T_s^0$ ,  $\tau_{sl}$ , and  $\alpha_0$  we calculated the difference between the intrinsic Faraday rotation and that at a time delay of 500 ps as a function of

temperature (dashed line in Fig. 4). Excellent agreement with the experimental data is found.

Because of the  ${}^6\Gamma_1^+ \rightarrow {}^4\Gamma_4^+$  excitation the electron potential energy increases by only 1.4 eV, while the excess of the photon energy is transferred to either the lattice or the magnetic system. Generally magnon-assisted transitions are less intense than phonon-assisted ones [17]. Consequently, after the optical excitation the temperature of the phonons is higher than that of the magnons:  $T_l > T_s$ . This difference gradually vanishes and the magnon temperature increases with a time constant determined by the phonon-magnon interaction that is predominantly related to the relativistic spin-orbit coupling in the magnetic ions and affected by magnetostriction only in the limited spectral range near the center of the Brillouin zone [18,19]. We found that the phonon-magnon interaction in  $\text{FeBO}_3$  has a characteristic time  $\tau_{sl} = 700$  ps.

The study of magnetization reversal by pulses of microwave radiation showed that the  $\text{FeBO}_3$  lattice is thermally insulated from the magnetic subsystem during about 16 ns after excitation [12]. This value is a factor of 20 larger than the phonon-magnon interaction time obtained in the present work. This large difference originates from the fact that in our experiment the energy exchange between the magnons and phonons over the whole Brillouin zone is important, whereas in microwave experiments only magnons with small or zero wave vector are involved. As the collision integral for these quasiparticles is relatively small, due to the conservation of energy and momentum, the equilibration of spin and lattice temperatures can be a 100 times faster than the decay of magnons with zero wave vector [19].

In summary, pump-probe measurements of the magneto-optical Faraday effect were used to monitor the optical destruction of antiferromagnetism in iron borate and to study the dynamics of this phenomenon. Unlike the metallic nickel and the half-metallic strontium-iron molybdate the photoexcitation of iron borate does not lead to a heating of the electronic subsystem. The photoexcitation of iron borate is assumed to heat up mostly the lattice via phonon-assisted transitions to the excited state and nonradiative relaxation. The antiferromagnetic order is subsequently destroyed via an energy transfer from the lattice to the magnetic subsystem that leads to an increase of the magnon temperature. The rate of the order parameter relaxation is determined by the phonon-magnon interaction time, which is deduced to be around 700 ps. This value is a factor of 20 smaller than found in experiments in the microwave region. The dynamics of the Faraday effect in the subpicosecond time domain is due to tran-

sitions of the  $\text{Fe}^{3+}$  ions to the excited low-spin state ( $S = 3/2$ ), which does not lead to any magnetic excitations because of fast relaxation of the ions to the ground state ( $S = 5/2$ ) within 100 fs.

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