Nonlinear Faraday effect in the weak ferromagnet $FeBO_3$ in ultrastrong magnetic fields up to 800 T

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A study of the Faraday effect in magnetic fields up to 800 T reveals no saturation of the effect as the field is increased. It is suggested that this result may be due to a strengthening of exchange interactions in a magnetic field or an effect of the field on electronic transitions in Fe^{3+} ions.

The $FeBO_3$ crystal is a weak ferromagnet ($T_N = 348$ K) with an easy-plane anisotropy. Its physical properties were recently reviewed by Diehl *et al.*[1] In the absence of a magnetic field, the ferromagnetic vector **m** and the antiferromagnetic vector **l** lie in the plane perpendicular to the $\bar{3}$ optic axis of the crystal. At $T = 0$ K, the weak magnetic moment is[2-4] $m_\perp = 1.3 \times 10^3$ A/m. Kurtzig *et al.*[5] studied the Faraday effect during propagation of light at an angle from the $\bar{3}$ axis, finding a specific rotation $|\alpha_\perp| = (20\text{–}2.2) \times 10^6$ deg/m in the optical region in an extrapolation to the basal plane.

We have now studied the Faraday effect during propagation of light along the optic axis, $B \| \bar{3}$ ($T = 300$ K), at the wavelength $\lambda = 0.6328$ μm in pulsed fields up to 800 T (Ref. 6). We use parallel-plate green crystals 100, 176, and 275 μm thick. The sign of the Faraday effect in the crystals and the magnitude of the field are determined from the Faraday effect in a reference sample (TF-5 glass) with a Verdet constant $V = 8.76$ deg/(cm · T). Figure 1 shows oscilloscope traces from one of the three experiments, while Fig. 2 shows the specific rotation for three samples and the intensity of the transmitted light versus the field (the upper inset). The rotation of the polarization plane is negative.

What is the reason for the change in the intensity of the light transmitted through the crystal as the field is strengthened? The wavelength $\lambda = 0.6328$ μm lies at the low-frequency edge of the absorption band corresponding to the ${}^6A_1 \rightarrow {}^4T_2$ transition in trivalent iron ions. The intensification of the transmitted light suggests that the field displaces the absorption band up the frequency scale. This shift is determined by the difference between the g-factors of the ground and excited states. In the ground state we have $g = 2.00$, and the shift of this state in a field of 800 T is $\Delta\omega \cong 800$ cm^{-1}. Even when we take the shift of the ground state into account, we find a value for the shift required to explain the observed increase in the transmission of the crystal which is correct in order of magnitude.

The specific Faraday rotation α can be written

$$\alpha = \alpha_f + \alpha_d, \qquad (1)$$

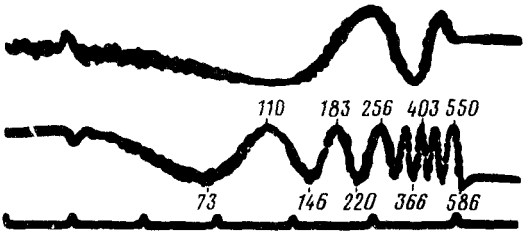

FIG. 1. Oscilloscope traces of the field-induced change in the intensity of light transmitted through the polarizer-sample-analyzer system. Top trace—$FeBO_3$ sample ($l = 0.275$ mm); lower trace—TF-5 glass reference sample ($l = 2.8$ mm). The numbers are the magnetic field in teslas.

where $\alpha_{fi} = F_{ij} m_j$ is the ferromagnetic contribution, $m_\perp$ and $m_\parallel$ are the magnetizations in the basal plane and perpendicular to it, F_{ij} is the magnetooptic tensor, $\alpha_d = V_d B$ is the positive diamagnetic contribution, and V_D is the Verdet constant. According to the data of Ref. 5, at 77 K and $\lambda = 0.6328$ μm, we would have $|\alpha_\perp| = (7.7\text{–}2.2) \times 10^6$ deg/m. Using $m_\perp = 1.3 \times 10^3$ A/m, we find $|F_\perp| = (5.9\text{–}1.7) \times 10^3$ deg/A. According to our data, in a field $B = 100$ T we have $\alpha_\parallel = -3.7 \times 10^5$ deg/m. The calculated value of the moment is $m_\parallel = 13 \times 10^3$ A/m, so we find $F_\parallel = -28.5$ deg/A, i.e., $F_\perp / F_\parallel \cong 10^2$. Consequently, there is a strong anisotropy of the Faraday effect in $FeBO_3$, and this anisotropy apparently explains the absence of the Faraday effect in fields up to 1 T.

Another feature of the Faraday effect here is that there is no saturation in the dependence $\alpha_\parallel(B)$ at fields up to $B = 800$ T. This feature becomes even more obvious if we single out $\alpha_{f,\parallel}$ from the observed rotation by taking into account the typical value $V_d = 8\text{–}3$ deg/(cm · T) [0.05–0.02 min/(cm · G)] for diamagnets. In a field of 800 T, the diamagnetic contribution reaches $\alpha_d \cong (6\text{–}3) \times 10^5$ deg/m and can no longer be ignored. The lower inset in Fig. 2 shows the dependence $\alpha_{f,\parallel}(B)$ [for $V_D = 3.3$ deg/(cm · T)] and the calculated value of $m_\parallel(B)$, which reaches saturation at fields

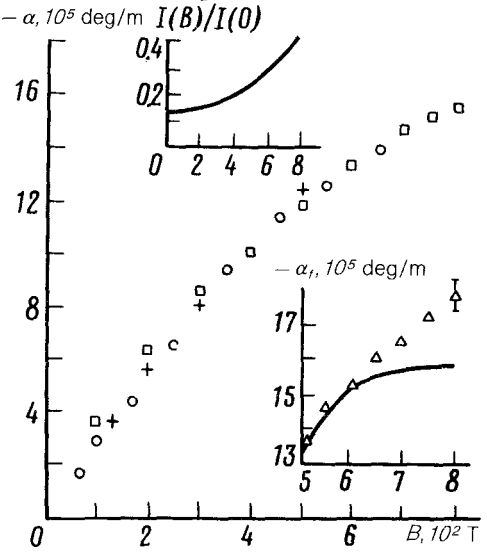

FIG. 2. Field dependence of the specific Faraday effect in three $FeBO_3$ samples. Circles—$l = 0.100$ mm; squares—$l = 0.176$ mm; plus signs—$l = 0.275$ mm. The rotation angle is determined within $\pm 10°$. The inset at the top shows the field-induced change in the intensity of light transmitted through the sample. The solid line in the inset at the bottom is the calculated behavior of the magnetic moment $m_\parallel$.

$B \cong 600$ T (solid line). The maximum molecular field in $FeBO_3$ is[7] $B_{ex} = 300$ T, and the sublattice-collapse field is $B_{coll} \cong 2B_{ex}$.

What are the reasons for the field dependence $\alpha_{f,\parallel}(B)$? An estimate of the negative paramagnetic contribution linear in the field,[8] $\alpha_p \sim -\bar{M}^2 B$, which in principle would increase the negative contribution to α_f even above the saturation of $m_\parallel$, turns out to be an order of magnitude below the observed value. The shift of the $^4T_2$ absorption band toward shorter wavelengths discussed above does not affect the change in $\alpha(B)$, since this band does not contribute to the dispersion of the Faraday rotation.[5] This rotation is dominated by the strong line $^6A_1 \rightarrow {}^4E, {}^4A_1$ which is centered in the region $\lambda_0 \cong 470$ μm, or, possibly, the two-exciton excitation $(^6A_1 + {}^6A_1) \rightarrow (^4T_1 + {}^4T)$ at $\lambda_0 = 410$ nm. There are two factors which might explain the significant effect of the field on α_f above the sublattice collapse point. The first is a strong shift of the line λ_0 induced by the field. Since the dispersion of the ferromagnetic contribution is described by

$$\alpha_f \sim -m\lambda_0^2/(\lambda^2 - \lambda_0^2), \tag{2}$$

in order to explain a 15% increase in α_f in comparison with the value in a field ≈ 600 T, we would need a red shift of λ_0 by an amount $\Delta\lambda \cong 0.02$ μm ($\Delta\omega \cong -1200$ cm^{-1}). This is quite a large shift, which we could not obtain on the basis of the transition $^6A_1 \rightarrow {}^4E, {}^4A_1$ (the sign of the shift is particularly troublesome). To some extent, this mechanism remains a possibility because of the unknown nature of the transition at 410 μm.

The absence of a saturation of the Faraday rotation in a field ≈ 800 T may be due to a magnetoelastic interaction, manifested as a growth of the exchange interaction. This mechanism is suggested, on the one hand, by the large change induced in T_N in $FeBO_3$ by an external pressure,[9] $dT_N/dp = 0.53$ K/kbar, and, on the other, by the shift of the maximum of the susceptibility $\chi_\perp$ toward a higher temperature, $dT_N/dB \cong 10$ K/T (in fields up to 0.03 T).[10] The magnetostriction of $FeBO_3$ in the case $B \parallel \bar{3}$ has not been measured (in the case $B \perp \bar{3}$, in fields up to 1 T, the magnetostriction is[3] $\tilde{\lambda} \cong 10^{-6}$). With increasing B, the decrease in distances gives rise to an effect analogous to that of an applied pressure. Assuming that dR/dp for $FeBO_3$ is on the same order of magnitude as for ruby,[11] -2×10^{-14} m/kbar, and assuming that the magnetostriction at $B \cong 100$ T reaches values $\tilde{\lambda} \cong 10^{-3}$, we find $\Delta R \cong \tilde{\lambda} R \cong -5 \times 10^{-13}$ m, i.e., $dB/dR \cong -0.2 \times 10^{15}$ T/m. If this is the case, then in strong fields we have

$$\frac{dT_N}{dB} \cong \frac{dT_N}{dp}\frac{dp}{dR}\frac{dR}{dB} \cong 10^{-1} \text{ K/T}.$$

This result leads to an increase in the molecular field to $H_{ex} \cong 350$–400 T, which can also explain the absence of a saturation in $\alpha(B)$.

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