MAGNETIC BIREFRINGENCE IN ANTIFERROMAGNETIC MnF_2

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Pisarev, Sinii, and Smolenskii [1] have observed that the magnetooptic effect that is quadratic in the magnetization (magnetic birefringence) is large in antiferromagnetic crystals and is comparable with the linear effect (the Faraday effect) observed in ferrimagnets. In [1], the magnetic birefringence was investigated in ferrites and antiferromagnets with weak ferromagnetism. In the present study, this effect was observed and studied in the antiferromagnetic crystal MnF_2. This is a thoroughly-investigated tetragonal antiferromagnet with $T_N = 66.9°K$. The sublattice magnetization (the vector $\vec{l}$) in it is directed along the fourfold axis (the z axis in Fig. 1).

The experiment was performed at a wavelength $\lambda = 6328$ Å far from the absorption bands in this substance. Linearly polarized light with a wave vector $\vec{k}$ from an He-Ne laser was incident on the crystal along the x axis (see Fig. 1). The light-polarization vector ($\vec{E}$) was inclined 45° to the z axis in the yz plane. The resultant change in the path difference between the ordinary and extraordinary rays was cancelled out and measured with the aid of a Berek compensator placed past the crystal. The measurements were made in a wide temperature interval, 1.5 - 300°K. The temperature was measured with a gold (doped with iron) - chromel thermocouple glued to the sample, and was maintained constant within 0.1 - 0.2°K.

At 300°K the difference between the ordinary and extraordinary refractive indices in MnF_2 is $n_e - n_o = 0.026$.[1] We measured the change of this quantity

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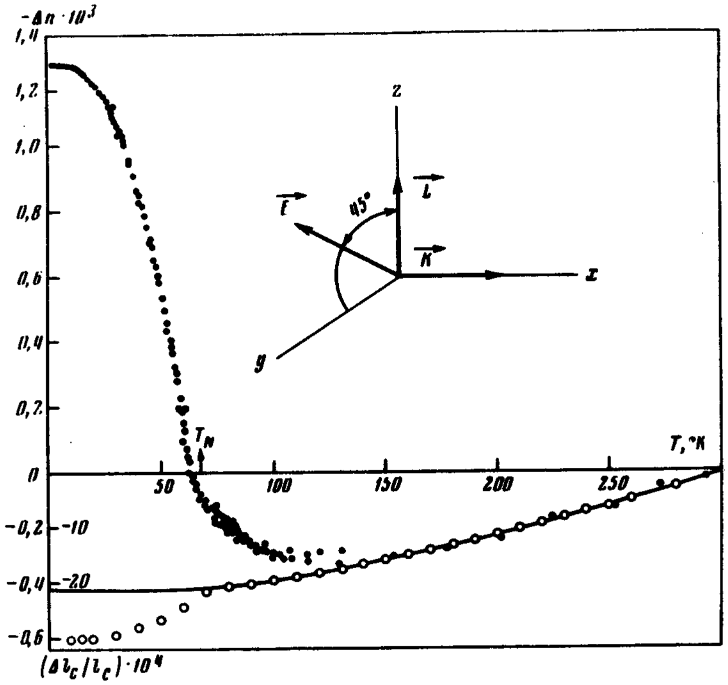
[1] The authors are grateful to V.I. Burkov for supplying these data.

Fig. 1



(Δn) as the temperature was lowered from 300°K down.

Figure 1 shows the main experimental results. It is seen from the figure that from 300 to 100°K the value of $n_e - n_o$ changes insignificantly, but in the region of T_N it begins to vary appreciably. The maximum is $\Delta n = 1.5 \times 10^{-3}$, approximately 5% of the natural birefringence. It should be noted that Δn in MnF_2 is larger by one or two orders of magnitude than in the substances investigated in [1], containing only magnetic ions of the iron group. This anomalous behavior of Δn can be naturally related with the occurrence of antiferromagnetic ordering in the crystal. However, the reason for such a behavior can be either the direct magnetic birefringence or the change of the refractive indices due to the spontaneous striction that is produced in crystals upon magnetic ordering (cf., e.g., [2]). Since this question has not yet been definitely answered, we have compared our experimental results with the available published data [3] on the temperature dependence of the relative elongation ($\Delta\ell/\ell$) in MnF_2. It follows from [3] that the value of $\Delta\ell/\ell$ along the twofold axis (x) is practically equal to zero. The data for $\Delta\ell/\ell$ along the fourfold axis, taken from [3], are shown to the appropriate scale in Fig. 1. The plots of Δn and of $\Delta\ell/\ell$ vs. T are in very good agreement in the region 150 - 300°K. In T_N we see an additional change of $\Delta\ell/\ell$, due to spontaneous striction, but the sign of this effect is opposite to the sign of the change Δn. In addition, the spontaneous striction is of the same order as the change of $\Delta\ell/\ell$ in the region from 300 to 100°K; on the other hand, the change of Δn following the onset of magnetic ordering is approximately five times larger than the temperature change from 300 to 100°K. In our opinion, this comparison indicates clearly that the growth we observed in Δn is direct magnetic birefringence.

To separate the pure magnetic birefringence, we extrapolated the nonmagnetic part of the $\Delta n(T)$ curve to lower temperatures, assuming this curve to be proportional to the nonmagnetic part of the plot of $\Delta\ell/\ell$ vs. T. The extrapolation was by means of the Gruneisen formula, assuming that

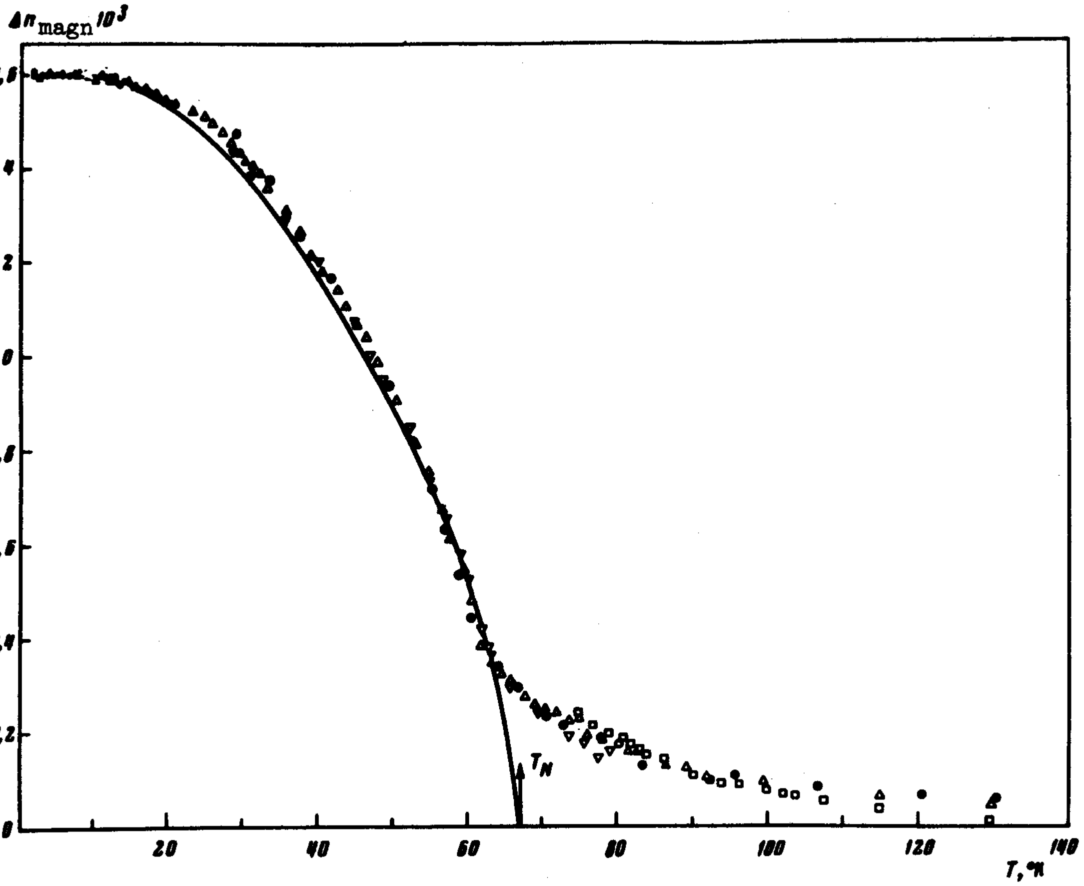


Fig. 2

$$\frac{\Delta l}{l} = \frac{l_T - l_{300^\circ}}{l_{300^\circ}} = K\left[\int_0^T C_D dT - \int_0^{300^\circ} C_D dT\right]. \tag{1}$$

C_D was chosen to be the Debye function calculated for $\theta = 455°K$, which agrees well with the experimental values of the specific heat [4] in the temperature range 150 - 300°K. The coefficient K in (1) is determined by comparison with the $\Delta l/l$ curve in the 150 - 300°K region. The curve calculated from formula (1) is shown solid in Fig. 1. The difference between the experimental and theoretical curves, which represents the temperature dependence of the magnetic birefringence, is shown in Fig. 2.

According to the theoretical notions [1, 5, 6], the value of Δn_{magn} is an antiferromagnet should be proportional to the mean-squared magnetization of the sublattices. The most accurate temperature dependence of the sublattice magnetization M was obtained from MnF_2 from measurements of the nuclear magnetic resonance (NMR) [7]. A comparison of the values of $M^2(T)$ obtained in that reference, referred to our experimental data in a helium bath (solid curve of Fig. 2), with Δn_{magn} shows a good agreement in a wide temperature range, thus confirming the relation $\Delta n_{magn} \sim \langle M^2 \rangle$. Near T_N, however, the values of Δn deviate strongly from the values of M^2 obtained from NMR. Δn_{magn} vanishes only at T = 120 - 130°K, whereas $M^2_{NMR} = 0$ at $T = T_N = 66.9°K$. Such an anomalous temperature dependence of Δn_{magn} is apparently due to the interaction of the light with the magnetic-moment fluctuations that are produced above T_n. These fluctuations could not be observed in experiments on AFMR [6] and NMR [7], since the period of the oscillations of the electromagnetic wave is in these cases larger by several orders of magnitude than the lifetime of an individual fluctuation. In experiments with light, these times are comparable in magnitude, and therefore the presence of such fluctuations in the crystal leads to a change of its refractive index.

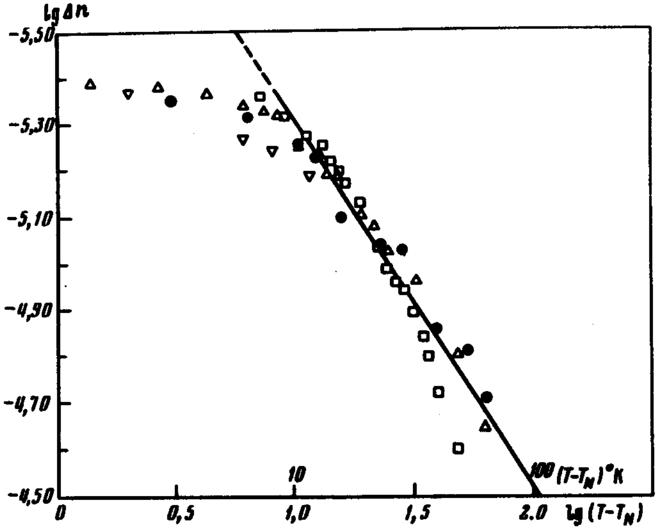


Fig. 3

Figure 3 shows our results for temperatures above T_N. It is seen from the figure that Δn_{magn} has a power-law variation at $T - T_N >$ 10°K:

$$\Delta n_{magn} = a/(T - T_N)^{(0,79 \pm 0,1)}. \qquad (2)$$

In conclusion, we thank P.L. Kapitza for interest in the work, S.V. Petrov for preparing the MnF_2 crystals, and S.M. Elagin for help with the experiments.

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