Linear magnetic birefringence of hematite in the vicinity of the Morin temperature

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An experimental investigation was made of birefringence of hematite (α-Fe_2O_3) in the course of propagation of light along the C_3 axis when a magnetic phase transition was field-induced below $T_M = 260$°K. This transition was found to be inhomogeneous. Its nonhysteretic nature was confirmed. A phenomenon specific to magnetically ordered crystals was observed: an odd (in respect of the field) linear magnetic birefringence appeared in a single-domain antiferromagnetic state of hematite and this was attributed to a linear (in respect of the field) rotation of the antiferromagnetic vector away from the trigonal axis. It was found that the application of very small mechanical stresses in the basal plane could lift the degeneracy between the two antiferromagnetic domains.

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1. INTRODUCTION

Magnetooptic methods are being used extensively in investigations of the state of magnetically ordered crystals. Propagation of light through weakly absorbing materials may result in a linear magnetic birefringence,[1] in addition to the thoroughly investigated and frequently used circular birefringence (Faraday effect). In the case of antiferromagnets the importance of the former effect is greater because the Faraday effect is small. The present paper reports an investigation of this linear birefringence in hematite (α-Fe_2O_3, space group D_{3d}^6) in the vicinity of the Morin temperature $T_M = 260$ °K.

A considerable amount of work has been done (for a review see Ref. 2) on the magnetic-field-induced transition of hematite from the antiferromagnetic to the weakly ferromagnetic state. Quantitative information on this transition has been obtained mainly by measuring the magnetization and susceptibility, but this does not provide an opportunity for direct determination of the orientation of the antiferromagnetic vector. This orientation can, in principle, be determined by neutron diffraction methods but such methods are time consuming and, in practice, their precision is low.

The most thorough (at present) neutron-diffraction investigation of the field-induced transition in hematite[3] confirmed reorientation of the antiferromagnetic vector from the trigonal axis in the basal plane, but was not sufficiently sensitive to determine small deviations of the vector. A special difficulty arose from the fact that there are two types of domain in the antiferromagnetic state of hematite and these differ in respect of the sign of the antiferromagnetic vector. Therefore, methods for converting a sample to a single-domain state (with respect to the antiferromagnetic vector) are particularly important and this applies specially to the influence of mechanical stresses on the formation of domains.

Birefringence measurements provide an important opportunity for investigating reorientation of the antiferromagnetic vector induced by the magnetic field below the Morin temperature.

A linear magnetic birefringence was observed in hematite by Pisarev, Siniĭ, and Smolenskiĭ.[4] The same authors observed disappearance of magnetic birefringence on transition of hematite from the weakly ferromagnetic to the antiferromagnetic state. The existence of an anisotropic (in respect of the componets of the antiferromagnetic vector) contribution to this birefringence makes it possible to determine the orientation of the antiferromagnetic vector when the latter deviates from the trigonal axis and emerges from the basal plane in a single-domain state, and to distinguish the different single-domain states. In spite of the fact that the symmetry of the magnetoelastic tensors is the same as the symmetry of the tensors describing the magnetooptic properties and since, therefore, similar information should be obtainable from magnetostriction investigations, the magnetooptic methods have a number of advantages: a) it is easy to ensure the absence of stray mechanical stresses which may have a considerable influence on the state of the magnetic subsystem; (b) local measurements can be carried out; c) it is easy to achieve a high precision of measurements. Therefore, the use of a magnetooptic method in investigating orientational phase transitions is the basic factor that can ensure acquisition of more reliable information.

2. PERMITTIVITY TENSOR

The linear magnetic birefringence of hematite at near-infrared wavelengths (our study was carried out at the wavelength $\lambda = 1.15\ \mu$) is governed by the symmetric part of the permittivity tensor ε_{ij}, which generally depends as follows on the components of the antiferromagnetic vector $\mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$ ($\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations):

$$\varepsilon_{ij} = \varepsilon_{ij}^0 + \eta_{ijkl} l_k l_l, \tag{1}$$

where ε_{ij}^0 is the part of the permittivity tensor independent of the magnetic subsystem; η_{ijkl} is a fourth-rank tensor; i, j, k, and l assume the values X, Y, and Z. The second term in Eq. (1) describes the purely magnetic contribution to the investigated birefringence as well as the contribution due to magnetostrictive strains.[6] The selection rules for the tensor η_{ijkl} are identical with the selection rules for the photoelastic tensor.[6,7] In the general case of an arbitrary orienta-

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tion of the antiferromagnetic vector **l**, the principal axes of the tensor ε_{ij} do not coincide with the coordinate axes X, Y, and Z. However, since the deformation of the optical indicatrix due to magnetic ordering is small, the angle of deviation of the principal axis from its initial position, parallel to Z, is small and it is of the order of the ratio of the magnetic to the natural birefringence, which amounts to $\approx 10^{-3}$ rad, whereas the other two principal axes lie (with the same accuracy) in the basal plane.[8] Consequently, light incident on a plate cut at right-angles to the C_3 axis propagates practically parallel to this C_3 axis, irrespective of the orientation of the antiferromagnetic vector. In this case the birefringence reduces to a change in the phase shift between normal plane-polarized waves crossing the plate. The phase shift and the direction of the principal polarizations are governed completely by the components of the permittivity tensor in the basal plane, for which we have the following expressions (the Z axis is parallel to C_3 and the X axis is parallel to U_2)

$$\begin{aligned}\varepsilon_{XX}&=\varepsilon_0^{\perp}+\eta_1^{1}l_X^2+\eta_1^{2}l_Y^2+\eta_1^{3}l_Z^2+\eta_1^{4}l_Yl_Z,\\ \varepsilon_{YY}&=\varepsilon_0^{\perp}+\eta_1^{2}l_X^2+\eta_1^{1}l_Y^2+\eta_1^{3}l_Z^2-\eta_1^{4}l_Yl_Z,\\ \varepsilon_{XY}&=\eta_1^{4}l_Xl_Z+(\eta_1^{1}-\eta_1^{2})l_Xl_Y.\end{aligned} \quad (2)$$

Let us assume that a crystal is rotated in the laboratory coordinate system xyz through an angle ψ about the Z axis coinciding with the z axis, and that an external magnetic field **H** is applied along the x axis. Then, the components of the tensor in the laboratory coordinate system are described by the following expressions derived bearing in mind that in the course of the phase transition from the antiferromagnetic to the weakly ferromagnetic state the vector **l** remains throughout in a plane perpendicular to **H**:

$$\begin{aligned}\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})&=\eta\sin^2\theta+\frac{1}{2}\eta_1^{4}\sin 2\theta\cos 3\psi,\\ \varepsilon_{xy}&=\frac{1}{2}\eta_1^{4}\sin 2\theta\sin 3\psi,\end{aligned} \quad (3)$$

where the angle θ defines the orientation of **l** relative to the Z axis and $\eta=-\frac{1}{2}(\eta_1^1-\eta_1^2)$.

The difference between the principal values of δ and the orientation angle α of the principal axes of the permittivity tensor in the basal plane can be described by

$$\begin{aligned}\delta&=[\eta^2\sin^2\theta+(\eta_1^{4})^2\cos^2\theta+\eta\eta_1^{4}\sin 2\theta\cos 3\psi]^{1/2}\sin\theta,\\ \operatorname{tg} 2\alpha&=\frac{\eta_1^{4}\cos\theta\sin 3\psi}{\eta\sin\theta+\eta_1^{4}\cos\theta\cos 3\psi}.\end{aligned} \quad (4)$$

As pointed out earlier,[9] these dependences of δ and α on the angle θ result in complex rotation and deformation of the optical indicatrix for arbitrary angles ψ, which gives rise to certain difficulties when the traditional methods are used. In the experimental method employed in the present study these difficulties are avoided because the values of the components of the permittivity tensor are found directly in the laboratory coordinate system and not in the system linked to the principal tensor axes.

It follows from the expressions in Eq. (4) that for a fixed angle θ the birefringence has a 120° anisotropy when a crystal is rotated about the C_3 axis and there are periodic changes in the positions of the principal axes of the tensor ε_{ij} and of the difference between the principal refractive indices in the basal plane. Similar results were reported for $CoCO_3$ by Kharchenko *et al.*[8] A 120° anisotropy of the birefrigence of $CoCO_3$ was observed by Pankov.[10]

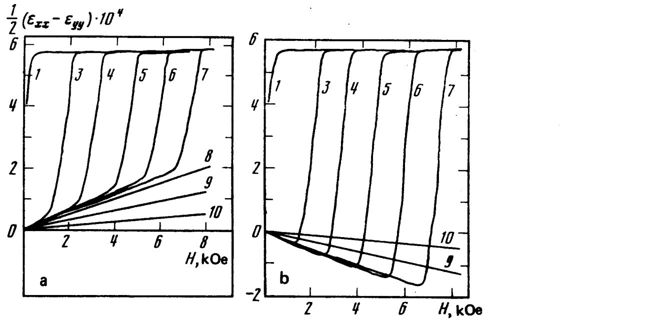

FIG. 1. Field dependences of $1/2(\varepsilon_{xx}-\varepsilon_{yy})$: a) $\psi=0°$; b) $\psi=60°$. Temperature (°K): 1) 260.8; 2) 258.9; 3) 258.0; 4) 257.2; 5) 256.4; 6) 255.6; 7) 254.8; 8) 251; 9) 238; 10) 80.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The experimental method described in our earlier communication[6] made it possible to measure signals proportional to the components $\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})$ and ε_{xy} of the permittivity tensor in the laboratory coordinate system provided sufficiently thin samples were used (the thickness was less than 0.3 mm for α-Fe_2O_3). A specially developed optical cryostat made it possible to stabilize the temperature of the sample in the range 80–300 °K to within ±0.01 °K. The cryostat was so constructed that it could be rotated together with a sample about an axis along which the light was propagated; this made it easy to vary the angle ψ. Above the Morin point T_M the dependence of the birefringence on the magnetic field was in the form of curves denoted by 1 in Figs. 1 and 2, indicating conversion of hematite from a polydomain to a single-domain state. In the range $T<T_M$ the behavior of birefringence was greatly affected by variation of the magnetic field.

Figure 1a shows the dependences of $\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})$ on the magnetic field at various temperatures $T<T_M$, obtained for a hematite sample whose U_2 axis was parallel to the external magnetic field ($\psi=0$). The sample was first cooled from room temperature in a magnetic field of +1 kOe (all the experimental records not identified by ar-

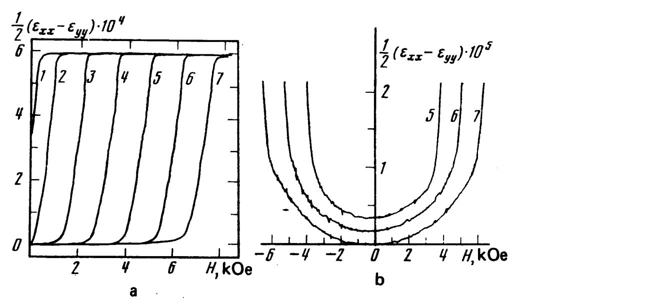

FIG. 2. Field dependences of $1/2(\varepsilon_{xx}-\varepsilon_{yy})$ obtained at different temperatures for $\psi=30°$. Temperatures are the same as in Fig. 1. In Fig. 2b the scale is increased and the initial points are displaced for clarity.

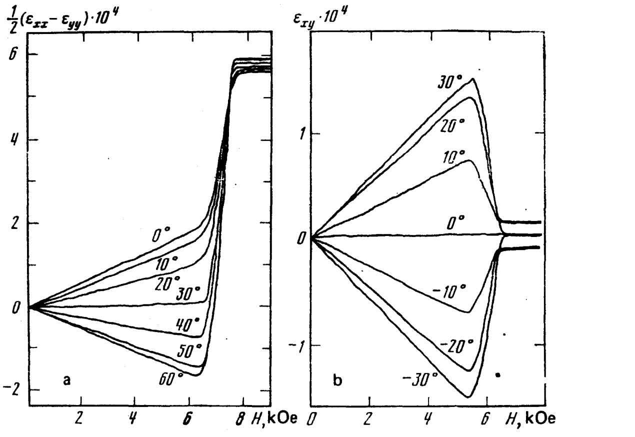

FIG. 3. Field dependences: a) $1/2(\varepsilon_{xx}-\varepsilon_{yy})$ at $T=254.8°$ K; b) ε_{xy} at $T=255.6$ °K, obtained for different orientations of the sample.

rows were made in an increasing magnetic field). In fields $H \leq 0.8H_{cr}^{\perp}(T)$ the birefringence varied practically linearly with the field. In the vicinity of $H_{cr}^{\perp}(T)$ the birefringence rose rapidly reaching its maximum value and it remained constant on further increase of the field to 10 kOe. The value of ε_{xy} was then zero within the limits of the experimental error (see curve $\psi=0°$ in Fig. 3b) indicating that the positions of the principal tensor axes were not affected and that one of these axes was parallel to **H**.

Rotation of a crystal by $\psi=60°$ reversed the sign of the slope of the linear regions (Fig. 1b). For intermediate values of ψ the curves representing $\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})$ and ε_{xy} are shown in Fig. 3. It is interesting to note that the positions of the principal tensor axes varied periodically on rotation of the crystal in a weak field:

$$\mathrm{tg}\, 2\alpha = 2\varepsilon_{xy}/(\varepsilon_{xx}-\varepsilon_{yy}) = \mathrm{tg}\, 3\psi, \tag{5}$$

and the period was 120° with respect to ψ. The difference between the principal values of the tensor was practically unaffected by changes in the orientation of the constant magnetic field in the linear region.

Like the phenomenon of linear magnetostriction, the odd birefringence was sensitive to a thermomagnetic treatment. The dependences of the birefringence on the magnetic field were obtained for a sample cooled from room temperature in a field $H=+1$ kOe (curve 1 in Fig. 4) and in a field $H=-1$ kOe (curve 2). The sign of the magnetic field of ≥100 Oe intensity at the moment of

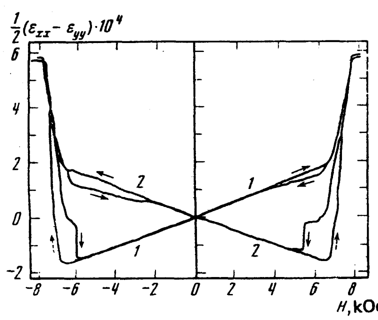

FIG. 4. Dependences of $1/2(\varepsilon_{xx}-\varepsilon_{yy})$ on H for $\psi=0°$ at $T=$ $T=254.8°$ K: 1) $H_{cool}=+1$ kOe; 2) $H_{cool}=-1$ kOe.

cooling (at $\psi=0$) resulted in stabilization of the sign and magnitude of the odd birefringence. As shown below, this thermomagnetic treatment produced a single-domain antiferromagnetic state and the subsequent study of the odd birefringence was made on this single domain.

This odd (in respect of the magnetic field) birefringence can occur only in magnetically ordered crystals belonging to 66 magnetic classes admitting linear magnetostriction.[11] In contrast to the linear (in the sense of polarization of normal waves) odd birefringence, the circular birefringence of the Faraday effect is an odd function of the external magnetic field for all the substances. Since we shall not consider the circular birefringence, the term "birefringence" will always mean the linear birefringence under consideration.

In the case of substances that do not belong to these classes, we can only have an even (in respect of the field) birefringence ($\sim H^2$, known as the Cotton–Mouton effect). The odd birefringence was observed by us earlier[12]; it was later investigated and discussed by Kharchenko *et al.*[13,14] and used to observe 180° domains.[15]

If we retain only the first terms of the expansion in respect of the field, we find that the dependence of the permittivity tensor on the magnetic field is

$$\varepsilon_p = \varepsilon_p^0 + \rho_p^i H_i + R_p^q (HH)_q, \tag{6}$$

where ρ_p^i represents essentially real coefficients. In the adopted geometry, it follows from the symmetry considerations that

$$\begin{aligned} {}^1/_2(\varepsilon_{xx}-\varepsilon_{yy}) &= \rho_1^1 H \cos 3\psi + RH^2, \\ \varepsilon_{xy} &= \rho_1^1 H \sin 3\psi. \end{aligned} \tag{7}$$

These formulas for the components are in good agreement with the birefringence observations. The component quadratic in the field is manifested most clearly by the $\psi=30°$ curves shown in Fig. 2b on an enlarged scale.

Since the main contribution to the magnetic birefringence of hematite is made by the components of the antiferromagnetic vector, the dependence of this birefringence on the external magnetic field in accordance with Eq. (7) at temperatures $T<T_M$ in fields $H<H_{cr}^{\perp}$ can be explained in a natural manner by the deviation of the vector **l** from the C_3 axis.

In fact, it follows from the formulas (3) that in the case of small values of θ (we shall consider an antiferromagnetic domain with $\theta=0$ in $H=0$)

$$\begin{aligned} {}^1/_2(\varepsilon_{xx}-\varepsilon_{yy}) &= \eta\theta^2 + \eta_1^4 \theta \cos 3\psi, \\ \varepsilon_{xy} &= \eta_1^4 \theta \sin 3\psi. \end{aligned} \tag{8}$$

A comparison of these formulas with those in Eq. (7) readily yields

$$\eta_1^4 = \pm\rho_1^1 (\eta/R)^{1/2}. \tag{9}$$

The values of ρ_1^1, R, and η known from the experiment studies [the value of η near T_M was found from the birefringence in the weakly ferromagnetic phase, i.e., when $\theta=\pm\pi/2$: $\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})=\eta=(5.76\pm 0.14)\times 10^{-4}$)] were used to determine the absolute value of the constant η_1^4 at $T=260-255$ °K: $|\eta_1^4|=(1.36\pm 0.08)\times 10^{-3}$ for a high-quality

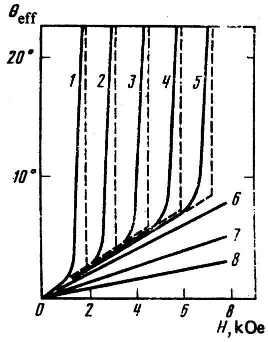

FIG. 5. Orientation of the vector l plotted as a function of H at various temperatures (°K): 1) 258.0; 2) 257.2; 3) 256.4; 4) 255.6; 5) 254.8; 6) 251; 7) 238; 8) 80.

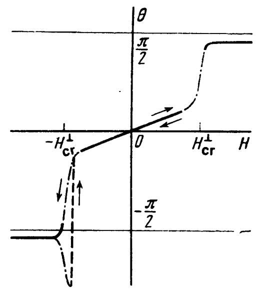

FIG. 7. Schematic dependence of θ on H at temperatures $T < T_M$ corresponding to Fig. 6. The dashed curve represents an abrupt change and the chain curve represents an inhomogeneous transition.

crystal with $T_M = 260\,°K$.

The linear dependence of the angle θ on the magnetic field is given by the formula

$$\theta = \rho_1^4 H/\eta_1^4. \tag{10}$$

Since the sign of the constant η_1^4 is not known, the angle θ can be determined but not its sign. To be specific, we shall interpret the experimental results on the assumption that $\eta_1^4 > 0$.

The experimental data on the birefringence were used to find a certain effective angle θ_{eff} which agreed with the angle θ calculated from Eq. (10) in the linear region and which was described by the following formula in the nonlinear region:

$$\theta_{eff} = \arcsin\left[\frac{1}{2\eta}(\varepsilon_{xx} - \varepsilon_{yy})|_{\psi=30°}\right]^{1/2}. \tag{11}$$

The dependences of θ_{eff} on H obtained for various temperatures T are plotted in Fig. 5. The slopes of the curves in the limits $H \to 0$ and $T \to T_M$ tended to a finite value and they also decreased on lowering of the temperature. The error in the determination of θ in the linear region did not exceed 10%. (In the case of poor quality crystals such a high precision could not be achieved.)

Introduction of the angle θ_{eff} is justified by the fact that the transition at $H_{cr}^{\perp}$ occurs in a very inhomogeneous manner with respect to the angle θ. In fact, if the angle θ increases monotonically and continuously on increase in H, the dependences of $\frac{1}{2}(\varepsilon_{xx} - \varepsilon_{yy})$ and ε_{xy} on H would have necessarily reached their extrema and these extrema would have been the same for different curves corresponding to a fixed value of ψ, which was contrary to the experimental data (Fig. 1b).

The high precision of the determination of the orientation of the vector l in the single-domain state made it possible to determine a slight deviation of the vector l from the basal plane in fields $H > H_{cr}^{\perp}$. This was particularly noticeable in the dependences of ε_{xy} on H in Fig. 3b obtained in fields $H > H_{cr}^{\perp}$. The 60° periodicity of the final values of ε_{xy} obtained for $H = 8$ kOe in the dependence on the angle ψ was due to the fact that the vector l of the weakly ferromagnetic phase did not fit entirely in the basal plane and during rotation of the field it emerged periodically (with a period of 120°) from the field. Using the theory of Refs. 16 and 17, we can estimate the third-order magnetic anisotropy constant in the basal plane $|d| = 1.3 \times 10^3$ erg·cm³ in the vicinity of the Morin temperature.

It is clear from Fig. 4 that there were also abrupt changes in the birefringence and, consequently, in the orientation of the vector l. Hysteresis and abrupt changes were observed mainly in fields opposite in sign to H_{cool}. Figure 6 shows the dependences of $\frac{1}{2}(\varepsilon_{xx} - \varepsilon_{yy})$ and ε_{xy} on H for $\psi = 15°$. We can see the correspondence of the abrupt changes in both curves. These experimental data are used in Fig. 7 to show schematically the dependence of the angle θ on the magnetic field for $\psi = 15°$.

The application of external mechanical stresses results in an additional crystallographic anisotropy and this can influence the orientation of the vector l and the changes in the domain structure.

The magnetic-field dependences of the birefringence

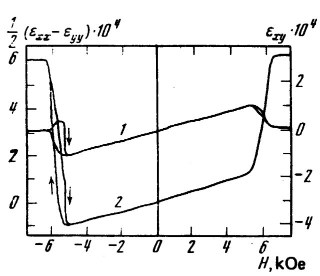

FIG. 6. Dependences of ε_{xy} (1) and $1/2(\varepsilon_{xx} - \varepsilon_{yy})$ (2) on H at $T = 255.6\,°K$ for $\psi = 15°$.

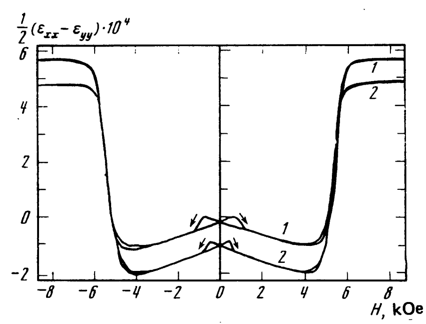

FIG. 8. Dependences of $1/2(\varepsilon_{xx} - \varepsilon_{yy})$ on H at $T = 256.6°$ K under different pressures p ⊥ U_2 kg/cm²: 1) 20; 2) 100.

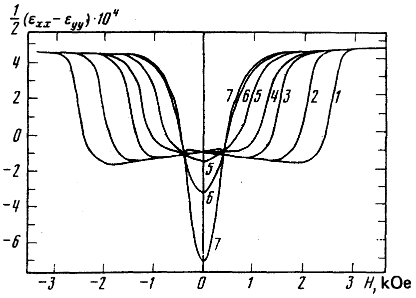

FIG. 9. Field dependences of $1/2(\varepsilon_{xx}-\varepsilon_{yy})$ in the vicinity of T_M recorded at different temperatures (°K): 1) 257.7; 2) 258.2; 3) 258.4; 4) 258.7; 5) 259.2; 6) 259.9; 7) 261.0; $|p| = 100$ kg/cm^2.

at temperatures $T < T_M$ observed on application of tensile stresses $p \perp U_2 \parallel H$ differed considerably from the dependences in the absence of mechanical stresses and became even irrespective of the magnetic field in which cooling took place (Fig. 8). The even dependence of $\frac{1}{2}(\varepsilon_{xx}-\varepsilon_{yy})$ on H and hysteresis near $H=0$ indicated a transition between two single-domain states differing in the sign of the Z component of the vector **l**. In fields $H=0$ and $H > H_{cr}$ the curves shifted proportionally to the applied load. The derivative obtained at $T = 256.6$ °K.

$$\frac{1}{2}\frac{\partial(\varepsilon_{xx}-\varepsilon_{yy})}{\partial p}\bigg|_{H=0} = -1.1\cdot 10^{-6}\ \mathrm{cm^2/kg} \qquad (12)$$

differed from the piezooptic constant $\zeta = 0.8\times10^{-6}$ kg/cm^2 (at $T > T_M$—Ref. 6). This was due to the fact that the application of mechanical stresses caused the vector **l** to deviate additionally from the C_3 axis, so that

$$\frac{1}{2}\frac{\partial(\varepsilon_{xx}-\varepsilon_{yy})}{\partial p} = \zeta + \eta_1^4\frac{\partial\theta}{\partial p}.$$

The experimental results indicated that

$$\left|\frac{\partial\theta}{\partial p}\right| = 0.24\cdot10^{-3}\ \mathrm{cm^2/kg}$$

at $T = 256$ °K. Hence, we could estimate the magnetic moment resulting from the application of a mechanical stress in the antiferromagnetic phase of hematite at $T = 256$ °K:

$$\frac{\partial m}{\partial p_\perp} = m_D\left|\frac{\partial\theta}{\partial_d}\right| = 0.5\cdot10^{-3}\ \mathrm{Oe\cdot cm^2\cdot kg^{-1}}.$$

This value was in agreement with the piezomagnetic constant $\Lambda_1^1 = 0.19\times10^{-3}$ Oe·cm^2·kg^{-1} at $T = 77$ °K (Ref. 18) if an allowance was made for the fact that the uniaxial anisotropy constant increased approximately threefold on cooling to 77 °K. We could conclude that the phenomenon of piezomagnetism in hematite is due to the formation of a magnetic moment because of deviation of the vector **l** from the C_3 axis under the action of mechanical stresses.

It is interesting to consider the behavior of birefringence when temperature was varied in the vicinity of T_M (Fig. 9) under a pressure $P = 100$ kg/cm^2. In the absence of a magnetic field at temperatures $T \ll T_M$ the vector was $\mathbf{l} \parallel z$, whereas at temperatures $T \gg T_M$ the vector was $\mathbf{l} \parallel x$ because of the mechanical stresses.[23] At $T \approx T_M$, intermediate configurations were possible. The application of a magnetic field parallel to the x axis always resulted, irrespective of temperature, in a transition of **l** to the state $\mathbf{l} \parallel y$ when the field was increased. In the vicinity of T_M there was a change in the nature of the motion of the vector **l**.

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