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MAGNETOSTRICTION OF A HEMATITE MONOCRYSTAL IN FIELDS UP TO 150 kOe

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The longitudinal magnetostriction of a hematite monocrystal has been studied in pulsed magnetic fields up to 150 kOe in the temperature interval 100–300°K. The magnetostriction of hematite is due chiefly to a change of direction of the antiferromagnetism vector l. The transition, under the influence of the field, from the antiferromagnetic to the weakly ferromagnetic state is accompanied by an anisotropic change of the dimensions of the crystal: the magnetostriction in the basal plane is positive, but along the trigonal axis of the crystal is negative. It is shown that in order to describe correctly the magnetization processes of hematite in the antiferromagnetic state, it is necessary to take account of the second constant of uniaxial anisotropy. The linear (odd in the field) magnetostriction of hematite was also studied. The temperature dependence of the constant P_{11} of linear magnetostriction was obtained.

HEMATITE, α-Fe_2O_3 (space group D_{3d}^6), below the Néel temperature ($T_N \sim 950°K$) is weakly ferromagnetic: the antiferromagnetism vector l and the weakly-ferromagnetic moment m_s are perpendicular to each other and lie in the basal plane of the crystal. On cooling below $T_c \approx 250$ to $260°K$ there occurs a transition to a purely antiferromagnetic state: in this case the vector l is parallel to the trigonal axis of the crystal[1]. The magnetoelastic properties of hematite have been studied little. Mainly, the magnetostrictive effects have been measured in the weakly ferromagnetic state and in the antiferromagnetic state near the temperature T_c[2-4].

An interesting peculiarity of hematite, resulting from its magnetic symmetry, is the fact that in it, along with the usual even magnetostriction, there is possible a magnetostriction that is odd in the field (linear)[5,6]. The linear magnetostriction and its thermodynamic inverse, the piezomagnetic effect, were observed in the antiferromagnetic modification of hematite at nitrogen temperature[3,7,8], but the piezomagnetic constants calculated from these two effects are different in orders of magnitude[8].

We have made measurements of the magnetostriction of a hematite monocrystal in the temperature interval 100–300°K in fields up to 150 kOe, sufficient for the transition from the antiferromagnetic to the weakly ferromagnetic state. Together with the usual magnetostriction, even in the field, the linear magnetostriction of hematite was studied.

MEASUREMENT METHOD AND SPECIMENS

The magnetostriction measurements were made in pulsed magnetic fields, on the apparatus described earlier[9], with an external piezotransducer for the pulsed deformations. The sensitivity of the appratus was increased by about an order of magnitude by suppression of parasitic mechanical vibrations and by increase of the amplification factor of the amplifier. The accuracy of measurement of the absolute value of the magnetostriction was 10%.

Specimens in the form of rods ($10 \times 1.5 \times 1.5$ mm), oriented along various crystallographic directions, were cut from hematite monocrystals grown from a molten bath in the Institute of Crystallography of the Academy of Sciences, USSR. The deviation from the chosen directions was checked by x rays and did not exceed 3°.

A study was made of the longitudinal magnetostriction along the trigonal axis of the crystal (the z axis of a rectangular coordinate system) and in the basal plane: along a second-order axis (the x axis), and perpendicular to it, along a direction lying in a symmetry plane of the crystal (the y axis). The magnetostriction in each of these directions was measured on two or three specimens, cut from different monocrystals.

EVEN MAGNETOSTRICTION

Figure 1 shows the field dependence of the longitudinal magnetostriction λ along various directions. The curves presented in Fig. 1 describe even magnetostriction: they do not change upon change of the field direction to the opposite direction.

The longitudinal magnetostriction along the trigonal axis of the crystal (the z axis) in the weakly ferromagnetic state is close to zero (it does not exceed 10^{-7}) (Fig. 1a). Below the point of transition to the antiferromagnetic state ($T_c \approx 253°K$), the magnetostriction along the z axis in weak fields remains small; on attainment of a certain field, it increases rapidly in absolute value; and on further increase of field, it changes insignificantly (Fig. 1a). Comparison with measurements of the magnetization[10] showed that the sudden change of strain corresponds to the field H_c at which there occurs a transition of the crystal from the antiferromagnetic to the weakly ferromagnetic state (Fig. 2).

From the measurements along the z axis it can be concluded that the magnetostriction of hematite is due principally to change of direction of the antiferromagnetism vector l, whereas change of the magnitude and direction of the vector m has little influence on the magnetostriction. For example, in the weakly ferro-

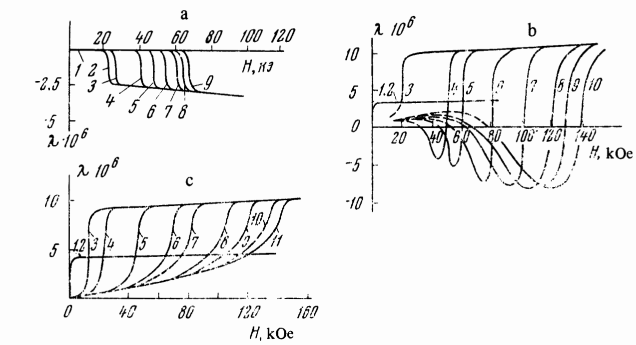


FIG. 1. Field dependence of the longitudinal magnetostriction of a hematite monocrystal along different directions. a) Magnetostriction along the trigonal axis (the z axis): curve 1 – 290; 2 – 250; 3 – 245; 4 – 230; 5 – 220; 6 – 205; 7 – 185; 8 – 120; 9 – 110°K. b) Magnetostriction in the basal plane, along the y axis: curve 1 – 295; 2 – 273; 3 – 251; 4 – 245; 5 – 230; 6 – 220; 7 – 205; 8 – 185; 9 – 165; 10 – 145; 11 – 120°K. c) Magnetostriction in the basal plane, along the x axis: curve 1 – 293; 2 – 263; 3 – 245; 4 – 230; 5 – 220; 6 – 205; 7 – 185; 8 – 165; 9 – 145; 10 – 120°K.

magnetic state the magnetostriction along the trigonal axis of the crystal is small, although, as follows from magnetic measurements [10], the magnetization m in a field of 100 kOe exceeds the weakly-ferromagnetic moment m_s by a factor of about 4.5, and the direction of the magnetization makes an angle of the order of 80° with the basal plane. This is due to the fact that in the weakly ferromagnetic state the field has no influence on the direction of the vector l (it remains in the basal plane). Below T_c the magnetostriction along the z axis also increases rapidly at the field H_c, when the antiferromagnetism vector l suddenly undergoes reorientation from the trigonal axis to the basal plane; whereas further increase of the field, having no influence on the direction of l, leads only to a small change of magnetostriction (Fig. 1,a).

Magnetostriction in the basal plane (Fig. 1b and c) in the weakly ferromagnetic state is due principally to rotation of the weakly-ferromagnetic moment and of the antiferromagnetism vector l, perpendicular to it, in the basal plane, and to displacement of the boundaries of the weakly-ferromagnetic domains. Since the effective anisotropy field in the basal plane of a hematite crystal is small (of order 1 Oe) [4], the magnetostriction saturates in weak field. A slight increase of the magnetostriction in strong fields is explained by the change of the weakly-ferromagnetic moment with field.

In the antiferromagnetic range (below $T_c \approx 253°K$),

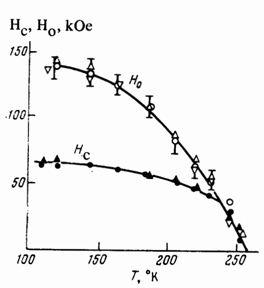


FIG. 2. Temperature dependence of the fields H_c and H_0. Field H_c: ●, according to measurements of the magnetization along the z axis; ▲, according to measurements of the longitudinal magnetostriction along the z axis. Field H_0: ○, according to measurements of the magnetization in the basal plane; ▽, according to measurements of the longitudinal magnetostriction along the y axis; △, according to measurements of the longitudinal magnetostriction along the x axis.

the magnetostriction along the x and the y axes depends differently on the field (Fig. 1,b and c). The magnetostriction along the y axis increases with field and reaches saturation at a certain field. Comparison with data of magnetic measurements [10] shows that the field for saturation of the magnetostriction coincides with the field H_0 of transition of hematite to the weakly ferromagnetic state for magnetization in the basal plane (Fig. 2).

The longitudinal magnetostriction of a hematite crystal along the x axis in weak fields is positive (Fig. 1c). On increase of the field the strain becomes negative, and then again positive, reaching saturation at field H_0. We remark that the values of H_0 and of the magnitude of the saturation strain along the x and the y axes are the same within the limits of experimental error.

From the theory of the weak ferromagnetism of hematite [11,12] it follows that in magnetization of the crystal by a field $H_\perp$ parallel to the basal plane, the antiferromagnetism vector l rotates out of the z axis of the crystal, in a plane perpendicular to the field. The different character of the magnetostriction along the x and y axes caused by this process is due to the fact that these directions are not crystallographically equivalent. This leads to the result that in the expansion of the magnetoelastic energy in components of the antiferromagnetism vector l and components of the deformation tensor u_{ij}, a term of the form $l_z l_y u_{xx}$ is present, whereas the term of the form $l_z l_x u_{yy}$ is absent [13].

In [14], magnetostrictive deformations of a hematite monocrystal along different crystallographic directions were calculated. In particular, from [14] there follows for longitudinal magnetostriction along the y axis

$$\lambda_{yy} = A(1 - l_z^2), \tag{1}$$

and along the x axis

$$\lambda_{xx} = A(1 - l_z^2) + Bl_y l_z = A(1 - l_z^2) + Bl_z\sqrt{1 - l_z^2}. \tag{2}$$

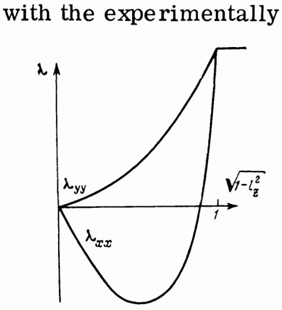
Here A and B are magnetostrictive constants, expressed in terms of the elastic moduli and of the magnetoelastic interaction coefficients. From these relations it is clear that λ_{yy} varies with l_z monotonically and that if A and B have different signs, λ_{xx} can change sign with change of l_z (Fig. 3).

From the theory of weak ferromagnetism [11,12] it follows that in magnetization of the antiferromagnetic modification of hematite in the basal plane,

$$\begin{aligned} &\sqrt{1 - l_z^2} \propto H_\perp \text{ for } H_\perp \leqslant H_0, \\ &l_z = 1 \qquad \text{for } H_\perp \geqslant H_0. \end{aligned} \tag{3}$$

Thus the curves in Fig. 3 describe the theoretical dependence of magnetostriction on field. It is seen that these curves agree qualitatively with the experimentally

FIG. 3. Theoretical dependence of the longitudinal magnetostriction on $\sqrt{1 - l_z^2} \sim H_\perp$ along the x and y axes according to formulas (1) and (2); $A > 0$, $B < 0$.



measured dependences $\lambda(H)$ in the basal plane (Fig. 1b and c). There are nevertheless differences between the experimental and theoretical curves. First, on the experimental curves there is a jump of the magnetostriction in the vicinity of the field H_0; it is especially well marked at temperatures close to T_C (Fig. 1b and c). Second, the theory does not explain the positive component of the magnetostriction along the x axis in weak fields (Fig. 1c).

The first circumstance is explained by the inaccuracy of the relation $\sqrt{1 - l_z^2} \infty H_\perp$. The papers [11,12] from which this relation follows take account only of the first constant of uniaxial anisotropy and predict a linear dependence of the magnetization of hematite on field in the basal plane, whereas experiment gives either a jump of the magnetization at $H_\perp = H_0$ (close to T_C)[15], or a nonlinear dependence of the magnetization on field[10]. Furthermore, the experimental values of H_0 are less than those calculated theoretically. In [11] the discrepancy between the experimental and theoretical data near the point T_C of hematite is attributed to a large second constant of uniaxial anisotropy. Measurements of the magnetization[10] and of the magnetostriction show that the second anisotropy constant plays a significant role also at lower temperatures. From calculations that we have made, it follows that in the temperature interval 100–230°K, allowance for the second constant explains the experimentally observed dependences of the magnetization and of the magnetostriction of hematite, in the basal plane, on the field; the ratio of the second constant to the first in this temperature interval is, according to our data, 0.3.

The nature of the positive component of the magnetostriction in weak fields is unclear. Our experiments showed that its amount varies greatly from specimen to specimen and depends on the previous history of the specimen (cooling in a magnetic field, the method of demagnetizing in the weakly ferromagnetic state, etc.). Possibly this component is due to motion of the boundaries of antiferromagnetic domains.

It follows from the magnetostriction measurements that the transition from the antiferromagnetic to the weakly ferromagnetic state is not purely volumetric, but is accompanied by an anisotropic change of the parameters of the crystal cell: the magnetostriction in the basal plane is positive, along the trigonal axis negative.

LINEAR MAGNETOSTRICTION

As has already been mentioned, hematite possesses, in addition to even magnetostriction, magnetostriction that is odd in the field (linear)[3,5]. In [5] it was shown that the linear magnetostriction of the antiferromagnetic modification of hematite in a direction with direction cosines α_x, α_y, α_z is

$$\lambda = P_{11}(\alpha_x^2 - \alpha_y^2)H_x + 2P_{24}(\alpha_x H_y - \alpha_y H_x)\alpha_z - 2P_{11}\alpha_x\alpha_y H_y, \quad (4)$$

where P_{11} and P_{24} are the constants of linear magnetostriction. It follows from formula (4) that the longitudinal linear magnetostriction along the coordinate axes is

$$\lambda_{yy} = \lambda_{zz} = 0, \qquad \lambda_{xx} = P_{11}H_x. \quad (5)$$

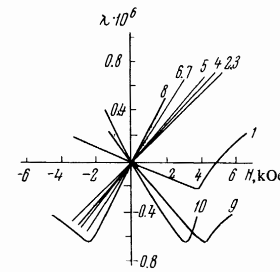


FIG. 4. Field dependence of the linear magnetostriction along the x axis. Curve 1 – without magnetic heat treatment, T = 220°K; curves 2-8, treatment in magnetic field + 100 Oe (2 – T = 119; 3 – 134; 4 – 145; 5 – 165; 6 – 183; 7 – 205; 8 – 220°K); curves 9 and 10, treatment in magnetic field –100 Oe (9 – T = 165°K; 10 – T = 205°K).

The measurements showed that, in agreement with theory, the linear magnetostriction is different from zero along the x axis (Fig. 4) and equal to zero along the two other axes. Our experiments also corroborate the data of papers [3,5] showing that the magnitude of the linear magnetostriction depends on the previous history of the specimen (Fig. 4). If the specimen is cooled below T_C in the absence of a field, the magnitude of the linear component is small (Fig. 4) and not reproducible from experiment to experiment. With cooling in a field that exceeds 100 Oe, the linear magnetostriction increases and becomes reproducible; the sign of the linear strain changes with change of the sign of the field in which the cooling occurs (Fig. 4). This effect of magnetic heat treatment is explained by the influence of antiferromagnetic domain structure on the magnitude of the linear magnetostriction[3,5]. Cooling in a field contributes to the formation of an antiferromagnetic structure with a preferred direction **I**, and this leads to increase of the linear strain. Our experiments indicate that the degree of single-domain tendency of specimens treated in a field is great. This is confirmed by the good reproducibility of the values of linear strain (and of the values of P_{11} calculated from them) from experiment to experiment and by the closeness of the values for different specimens (Fig. 5).

Furthermore, the values of P_{11} found by us at nitrogen temperature differ little from the value $P_{11} = 1.9 \times 10^{-10}$ Oe^{-1} obtained in paper[5]. Such coincidence, in the presence of a multidomain antiferromagnetic structure, is unlikely. Apparently, therefore, the difference between the values of the piezomagnetic constants calculated from the linear magnetostriction and from piezomagnetic measurements cannot be explained by the presence of antiferromagnetic domains in the measurement of the linear strain. We remark that the value of the piezomagnetic constants determined from the influence of pressure on the electron paramagnetic resonance spectrum of Fe^{3+} and Al_2O_3 ions[16] agree in order of magnitude with data from measurements of the linear strain.

We succeeded in observing linear magnetostriction only in weak fields (or order 4 to 6 kOe). With further increase of the field, the linear strain goes over to even positive strain. The transition is seen especially

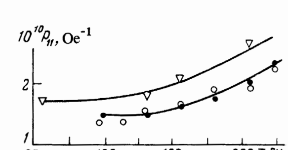


FIG. 5. Temperature dependence of the coefficient of linear magnetostriction P_{11}: Δ,– specimen No. 1; ○, ● – specimen No. 2 (the white and black circles refer to different experiments).

clearly when the linear strain is negative (Fig. 4, curves 1 and 8-10).

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