UNIDIRECTIONAL ANISOTROPY IN SINGLE CRYSTALS OF COBALT-IRON HEXAGONAL

FERRITES WITH A Z-TYPE STRUCTURE

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Unidirectional anisotropy is observed in single crystals of hexagonal ferrites with a Z-type structure containing cobalt ions. The nature of this anisotropy is discussed from the point of view of the possible existence in layer crystals of exchange anisotropy discovered by Meiklejohn and Bean.^[3]

1. In connection with the observation of exchange anisotropy in single crystals of hexagonal ferrites with the M structure^[1] we have made an attempt to find indications of it in hexagonal ferrites with other structures. Hexagonal ferrites are made up of successive cubic and hexagonal blocks. If a part of a block has an antiferromagnetic spin ordering while the adjoining blocks are ferromagnetically ordered, then there exists the possibility of a ferri-antiferromagnetic coupling between these blocks which leads to the appearance of exchange anisotropy. It is well known that there are antiferromagnetic blocks in hexagonal ferrites with the Z structure (Ba₃Me₂Fe₂₄O₄₁, Me is a divalent metal ion). These are the so-called T blocks (see, for example,^[2]).

The magnetic anisotropy energy of hexagonal crystals E_c is written in the following form

$$E_{\rm c} = K_1 \sin^2 \vartheta + K_2 \sin^4 \vartheta + K_3 \sin^6 \vartheta \cos 6\varphi, \tag{1}$$

where K_1 and K_2 are the first and second magnetic anisotropy constants, K_3 is the magnetic anisotropy constant of the basal plane, ϑ and φ are polar coordinates of the magnetization vector **M** with ϑ read from the hexagonal c axis. The torque L in the plane perpendicular to the basal plane is of the following form:

$$L = -\frac{\partial E_k}{\partial \vartheta} = -(K_1 + K_2)\sin 2\vartheta + \frac{K_2}{2}\sin 4\vartheta.$$
 (2)

On adding cobalt ions to crystals of the hexagonal ferrites the constant K_1 decreases and changes sign above a certain content. One can thus choose a composition such that (at room temperature) the constant K_1 be close to zero. Such a choice of the composition allows one to separate readily from the experimental torque curve terms not connected with the crystallographic anisotropy (1).

2. The appropriate single crystals were grown by the method of spontaneous crystallization from a solution of the components of the $Ba_3Co_{0.9}Fe_{1.1}^{2+}Fe_{24}^{3+}O_{41}$ ferrite in an NaFeO₂ melt. We shall refer to them briefly as CoFeZ. For these crystals we recorded magnetization curves by means of a vibrating-sample magnetometer and investigated the magnetic anisotropy by means of torque curves. All the investigations were carried out at two temperatures: 293 and 77°K. In order to determine the magnetic anisotropy constants, we expanded the torque curves in a Fourier series.

In Fig. 1 we present torque curves of CoFeZ in a plane perpendicular to the basal plane in a field of

19.5 kOe. The curve at room temperature (curve 1) is described by the following formula:

$$L = \pm j_{\rm c} \sin \vartheta$$
,

where $j_r = 3.1 \times 10^5 \text{ erg/cm}^3$. The plus sign is taken in the s range between 90 and 270°, and the minus signfrom 270 to 90°. The value of j_r in fields above 15 kOe does not depend on the magnitude of the applied magnetic field.

At $77^{\circ}K$ (curve 2) the torque curve is of a more complicated form. Its expansion in a Fourier series shows that in addition to $\sin \vartheta$ it has higher-order components.

In the basal plane the anisotropy of CoFeZ is so small ($K_3 \approx 0$) that none was observed either at 293°K or at 77°K.

Figure 2 shows the magnetization curves of CoFeZ crystals. The curves have an unusual form. Along the basal plane (curves 2 and 4) they have a critical field above which the magnetization increases linearly up to saturation.

3. The exchange anisotropy is characterized by three features^[3]: unidirectional anisotropy (a sin ϑ component in the torque curve), a shift in the hysteresis loop, and rotational hysteresis. The appearance of one of these features depends on the ratio between the ex-



FIG. 1. Torque curves of $Ba_3Co_{0.9}Fe_{1.1}^{2+}Fe_{24}^{3+}O_{41}$. Curve $1 - T = 293^{\circ}K$, curve $2 - T = 77^{\circ}K$.

FIG. 2. Torque curves of $Ba_3Co_{0.9}Fe_{1.1}^{2+}Fe_{24}^{3+}O_{41}$. Curve $1 - T = 293^{\circ}K$, H || c; $2 - T = 293^{\circ}K$, H \perp c; $3 - 77^{\circ}K$, H || c; $4 - 77^{\circ}K$, H \perp c; 5 - theoretical curve for 293°K. change interaction constant j_r and the constants of ferromagnetic and antiferromagnetic anisotropy. For the case of small crystallographic anisotropy the energy of the crystal E in a magnetic field H directed at an angle ψ to the hexagonal c axis will be of the following form:

$$E = MH\cos(\psi - \beta) + K_{AF}\sin^2\alpha - j_{\rm B}\cos(\beta - \alpha).$$
(3)

Here M is the spontaneous magnetization of the crystal, β is the angle between M and the c axis, α is the angle between the direction of the spins in the antiferromagnetic block and the c axis, and K_{AF} is the antiferromagnetic anisotropy constant. This equation has a solution only for $h = HM/j_r \ge 1$ and $j_r/K_{AF} > 1$. The presence of losses to rotational hysteresis will depend on the magnitude of the ratio j_r/K_{AF} . The larger this ratio, the sooner, i.e. the lower the fields for which the losses will disappear.^[1] In CoFeZ crystals there are no rotational hysteresis losses. Consequently, the ratio j_r/K_{AF} is large. It was shown in^[1] that for $j_r/K_{AF} > 3$ no losses to rotational hysteresis will be observed in high magnetic fields.

Of the indicated three features only one is observed in CoFeZ crystals—the presence of $\sin \vartheta$ in the torque curves. The presence of $\sin \vartheta$ is not related to an insufficiently high magnetic field. In fact, the angle of lag γ of the magnetization vector **M** behind the field vector **H** is calculated from the relation

$$\gamma = \arcsin(L / MH).$$

The maximum angle of lag $\gamma = 3^{\circ}$ which practically

does not change the shape of the curve.

Let us now calculate the magnetization curve of CoFeZ in the basal plane ($\psi = 90^{\circ}$). Differentiation of (3) with respect to α and β leads to the following two equilibrium conditions:

$$(j_c / K_{AF}) \sin (\beta - \alpha) = \sin 2\alpha, \quad h \cos \beta = \sin (\beta - \alpha).$$

The numerical solution of these equations for j_r/K_{AF} = 1.5 is shown on the lower right-hand side of Fig. 2. This solution is insensitive to a change of the ratio j_r/K_{AF} . When its magnitude changes, there is a change in the slope of the initial linear section of this curve. The theoretical curve is only qualitatively similar to the experimental curve.

The observation of the $\sin \vartheta$ component in the torque curve and the qualitative agreement of the calculated and experimental magnetization curves furnishes grounds for an explanation of the magnetic properties of CoFeZ by the presence of exchange anisotropy.

³W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956).

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²J. Smit and H. P. Wijn, Ferrites, Philips Technical Library, Eindhoven, 1959.