MAGNETIC PROPERTIES OF HEXAGONAL FERRITES WITH WEAK EXCHANGE COUPLING BETWEEN SUBLATTICES

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Peaks are observed in the temperature curves of the first magnetic anisotropy constant K_1 and the magnetization in single crystals of the system $BaIn_xFe_{12-x}O_{19}$ for $x > 1.9$ and in a single crystal of $PbIn_{1.9}Fe_{10.1}O_{19}$. A dependence of K_1 on magnetic field strength is observed at 77°K. All three experimental facts are explained on the basis of a model in which weak exchange interaction between magnetic sublattices is assumed.

THE exchange interaction between sublattices in ferromagnets is usually greater than or comparable with the exchange interaction within each of the sublattices. Hence the magnetic properties of ferrimagnets in the region of technical magnetization do not differ qualitatively from those of ferromagnets. However, we have recently encountered single crystals of certain hexagonal ferrites in which the exhange interaction between sublattices is weak. This leads to the existence in the ferrites $Sr_3Zn_2Fe_{24}O_{41}$ and $Sr_2Zn_2Fe_{12}O_{22}$ of critical magnetic fields above which the magnetization vector of one of the sublattices is rotated into the direction of the applied field.[1,2] In addition, it was shown that in this case the magnetic-anisotropy constants K_1, K_2, . . . cease being constants and depend on the magnitude of the applied field.[2,3] Thus, the usual description of the dependence of the magnetic anisotropy energy E_K of a hexagonal crystal on the angle θ between the magnetization vector and the hexagonal axis of the crystal

$$E_K = K_1 \sin^2\theta + K_2 \sin^4\theta + \dots \quad (1)$$

becomes unsuitable. Dependence of the magnetic anisotropy constants on the magnetic field intensity in its simplest form was observed in hexagonal crystals of $BaSc_{1.8}Fe_{10.2}O_{19}$,[3] for which only the first term of Eq. (1) could be used.

In our work we have observed still another new phenomenon which can exist in crystals with weak exchange coupling between sublattices—an anomalous dependence of the anisotropy constant K_1 on temperature. This phenomenon was observed in the system of single crystals $BaIn_xFe_{12-x}O_{19}$ for $x > 1.9$ and in a single crystal of $PbIn_{1.9}Fe_{10.1}O_{19}$.

Monocrystals of these ferrites were prepared by the method of spontaneous crystallization from solution in a BaO-B_2O_3 flux. In all cases, the starting compounds, $BaCO_3$, Fe_2O_3, In_2O_3, and B_2O_3, were analytical grade reagents. Hexagonal-shaped crystals 3–4 mm in size (in the plane of the pinacoid) and 1–3 mm thick were obtained. The composition of the crystals was determined by x rays (together with some microchemical analysis), from which it was found that the In^{3+} content varied from $x = 0$ to $x = 3.4\%$.

X-ray data confirmed that these crystals all belonged to the same structural type M, the typical representative of which is the hexaferrite $BaFe_{12}O_{19}$, space group $P6_3/mmc$.

The table gives the crystal lattice parameters a and c of these crystals. They were determined from the reflections from $(11\bar{2}0)$ and (0022) at the Bragg angles $2\theta = 90$ and 140° ($Fe_{K\alpha}$ radiation). The measurement accuracy of the lattice parameters a and c is ±0.001 and ±0.005 Å, respectively.

We used three methods to investigate the magnetization processes: 1) Faraday method for taking magnetization curves and temperature behavior of magnetization in fields up to 10 kOe; 2) vibrating-magnetometer method for the same purposes, but up to fields of 15 kOe (one magnetization curve for $BaIn_{2.4}Fe_{9.6}O_{1.9}$ at liquid helium temperature was taken in fields to 50 kOe in a superconducting solenoid); 3) torque method for investigating magnetic anisotropy in fields to 25 kOe from 77 to 293°K. The crystals were made into spheres for all investigations.

Figure 1 shows the temperature dependences of the magnetization σ of all the crystals indicated in the table in a magnetic field of 10 kOe. The measurements were carried out in a magnetic field parallel to the hexagonal c axis. A peculiarity here is the presence of a maximum in the curves. Similar maxima for the same system were observed by Efimova and Mamaluĭ.[4] These authors followed Néel in explaining this, i.e., they said that the magnetization of the A sublattice fell more rapidly with temperature than that of the B lattice. It must be noted further, that for crystals with $x > 1.9$, saturation is not reached in fields up to 15 kOe, the magnetization increasing linearly with increasing field intensity. For the sample with $x = 2.4$, the magnetization curve was taken at liquid helium temperature in fields to 50 kOe. Even in these fields saturation was not attained. (Fig. 2).

In Fig. 3 is shown the temperature dependence of the first magnetic anisotropy constant K_1 (see Eq. (1)). Not all the curves are shown, only typical ones. Certain of the quantities not shown in the figures (magnetization σ, magnetic anisotropy constant K_1, Curie temperature T_C) can be found in the table. For the composition with $x = 3$ (curve 5), we do not give the value of K_1 at 77°K since it strongly depends on the magnitude of the magnetic field at this temperature, i.e., it is not constant. All the other

Composition	a, Å	c, Å	σ, G cm³ g⁻¹		$K_1 \cdot 10^{-5}$, erg cm⁻³		T_c, °K
			293° K	77° K	293° K	77° K	
$BaFe_{12}O_{19}$	5.883	23.195	70.5	98.0	35.4	41.4	728
$BaIn_{0.5}Fe_{11.5}O_{19}$	5.900	23.26	64.0	93.5	31.4	42.2	693
$BaIn_{1.2}Fe_{10.8}O_{19}$	5.924	23.38	60.5	88.0	19.7	24.2	626
$BaIn_{1.9}Fe_{10.1}O_{19}$	5.949	23.52	50.0	80.5	9.5	14.2	538
$BaIn_{2.4}Fe_{9.6}O_{19}$	5.968	23.61	33.5	50.0	5.3	5.9	440
$BaIn_{2.5}Fe_{9.5}O_{19}$	5.970	23.62	24.5	44.5	3.15	4.7	453
$BaIn_3Fe_9O_{19}$	5.995	23.72	5.65	26.5	from 0.1 to 0.4	depends on field	333
$BaIn_{3.4}Fe_{8.6}O_{19}$	6.000	23.79	4.00	21.0	<0.1	intensity	323
$PbIn_{1.9}Fe_{10.1}O_{19}$	5.945	23.50	37	55.0	5.7	5.85	463

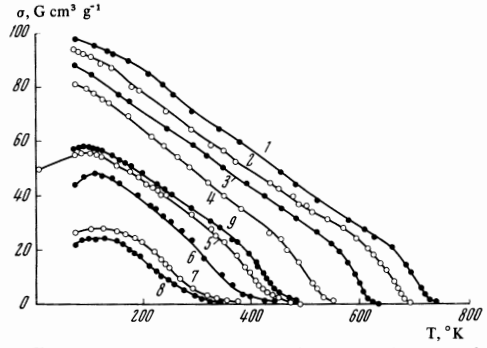

FIG. 1. Temperature dependence of the magnetization of monocrystals of $BaIn_xFe_{12-x}O_{19}$ in a field H = 10 kOe and of a monocrystal of $PbIn_{1.9}Fe_{10.1}O_{19}$: curve 1–x = 0; 2–x = 0.5; 3–x = 1.2; 4–x = 1.9; 5–x = 2.4; 6–x = 2.5; 7–x = 3.0; 8–x = 3.4; 9–$PbIn_{1.9}Fe_{10.1}O_{19}$.

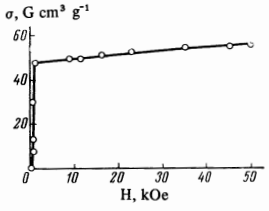

FIG. 2

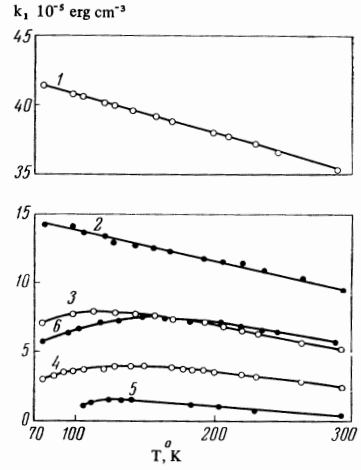

FIG. 3

FIG. 2. Magnetization curve of a monocrystal of $BaIn_{2.4}Fe_{9.6}O_{19}$ at liquid helium temperature with H ∥ c.

FIG. 3. Dependence of the magnetic anisotropy constant K_1 on temperature for monocrystals of $BaIn_xFe_{12-x}O_{19}$ and $PbIn_{1.9}Fe_{10.1}O_{19}$: curve 1–x = 0; 2–x = 1.9; 3–x = 2.4; 4–x = 2.5; 5–x = 3; 6–$PbIn_{1.9}Fe_{10.1}O_{19}$.

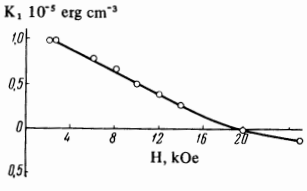

FIG. 4. Dependence of the magnetic anisotropy constant K_1 of a single crystal of $BaIn_3Fe_9O_{19}$ on magnetic field intensity.

values of K_1 given in the figure do not vary with magnetic field.

Figure 4 gives the dependence of K_1 on the intensity of the magnetic field for crystals with x = 3 at 77°K. The "constant" K_1 falls linearly with increasing magnetic field. If in fields less than 20 kOe the crystal has its axis of easy magnetization along the hexagonal axis of the crystal ($K_1 > 0$), then above 20 kOe the easy axis lies in the basal plane ($K_1 < 0$). This also happens in crystals with x = 3.4. Thus, for crystals with $x \geq 3$ at 77°K, it is impossible to speak of magnetic anisotropy constants as described by Eq. (1).

We shall now discuss these results. We make use of the model of two magnetic sublattices with a weak exchange interaction between them.[1] Then Eq. (1) must be replaced by another, which in the first approximation of perturbation theory has the following form:

$$E'_\kappa = k \sin^2\psi + k' \sin^2\psi' + k'' \cos\psi \cos\psi' + k''' \sin\psi \sin\psi', \quad (2)$$

where ψ, ψ' are the angles between the magnetic vectors of the sublattices **M**, **M'** and the hexagonal crystal axis. Here the anisotropy constants k, k', k", and k"' are constant quantities, i.e., they do not depend on the intensity of the magnetic field H.

In [2] we compared Eq. (2) with the first term of the series (1), using the following obvious relation:

$$\operatorname{tg}\vartheta = (M \sin\psi + M' \sin\psi') / (M \cos\psi + M' \cos\psi').$$

With the assumption of a weak exchange interaction between sublattices, the mutual orientation of **M** and **M'** changes with a change in magnetic field intensity H, i.e., the values of ψ and ψ' change. Consequently, the total magnetization of the crystal also changes as a function of the magnitude of the applied field (Fig. 2) or of temperature (Fig. 1). The magnetic anisotropy constant K_1 also varies as a function of the field magnitude (Fig. 4) and of temperature (for H ≠ 0) (Fig. 3). (Experimentally, we can determine only the constants K_1, K_2, ... from Eq. (1); the constants k, k', k", k"' from Eq. (2) are not subject to measurement.) The specific functions $K_1(H)$ and $K_1(T)$ differ for different special solutions $\psi(H)$, $\psi'(H)$, $\psi(T)$, $\psi'(T)$.

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