HELICOIDAL SPIN ORDERING IN THE HEXAGONAL FERRITE $(Sr_{0.8}Ba_{0.2})_2Zn_2Fe_{12}O_{22}$

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Magnetic superstructure reflections were observed in a $(Sr_{0.8}Ba_{0.2})_2Zn_2Fe_{12}O_{22}$ single crystal by the neutron diffraction technique. The reflections can be explained as due to helicoidal spin ordering. A critical magnetic field was found in the magnetization curves of the crystal. Exchange and magnetic dipole interactions were estimated. It was concluded that the exchange interaction was solely responsible for the existence of magnetic helicoids in the crystal. A study of the torque revealed the simultaneous existence of a plane and cone of easy magnetization.

1. INTRODUCTION

HEXAGONAL ferrites with the **Y** structure, $Ba_2Me_2Fe_{12}O_{22}$ (Ba_2Me_2**Y**), differ from hexagonal ferrites with other structures (**M**, **W**, **Z**, **X**) in that their magnetocrystalline anisotropy has an easy magnetization plane at all temperatures, irrespective of the nature of the divalent metal ion Me. However, when barium is replaced by strontium, ferrites in which Me = Zn do not exhibit spontaneous magnetization. This was observed first by Enz [1] for the ferrite $(Sr_{0.95}Ba_{0.05})_2Zn_2$**Y**. From an investigation of the magnetic properties of this crystal, Enz concluded that it had a helicoidal spin configuration.

We investigated the magnetic structure of this ferrite by neutron diffraction and we also studied its magnetocrystalline anisotropy and characteristic magnetization features.

We used $(Sr_{0.8}Ba_{0.2})_2Zn_2$**Y** crystals. For comparison, we investigated also strontium-free crystals of Ba_2Zn_2**Y**. These crystals were grown from a molten solution.

2. MAGNETIZATION CHARACTERISTICS

The magnetic properties of the crystals were investigated using a vibration magnetometer in the temperature range from 77°K to the Curie temperature. The samples were spherical. Figure 1 shows the magnetization curves of a $(Sr_{0.8}Ba_{0.2})_2Zn_2$**Y** crystal at two temperatures: 293 and 77°K. The crystal was magnetized along two directions: along the hexagonal c-axis of the crystal and along a direction lying in the basal plane. Of special interest was the magnetization curve in the basal plane (curve 1). In fields up to 700 Oe, the room-temperature magnetization σ was a linear function of the magnetic field intensity H. In a field H = 700 Oe, the magnetization increased suddenly and then again continued to rise linearly with increase of the magnetic field intensity up to 7000 Oe. In a field of 7000 Oe, the magnetization jumped again and evidently reached saturation.

At liquid nitrogen temperature the magnetization curve for the basal plane was non-linear in weak magnetic fields (curve 3). However, after an initial sharp rise (in fields up to 2000 Oe), the magnetization σ increased linearly with increase

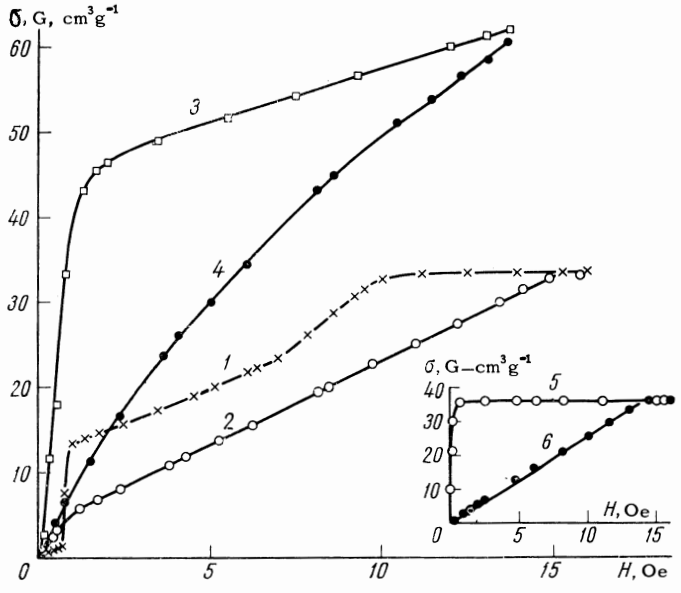

FIG. 1. Magnetization curves of $(Sr_{0.8}Ba_{0.2})_2Zn_2$**Y**: 1) H ⊥ c at 293°K; 2) H ∥ c at 293°K; 3) H ⊥ c at 77°K; 4) H ∥ c at 77°K; magnetization curves of Ba_2Zn_2**Y** at 293°K: 5) H ⊥ c; 6) H ∥ c.

in the magnetic field intensity. A field of 14 000 Oe, which was the strongest field used in the determination of the magnetization curves in liquid nitrogen, was insufficient to produce saturation, but evidently we were close to that state (curves 3 and 4 nearly converge).

The linear part of the magnetization curve in weak fields was observed only in the temperature range 270—330°K. For comparison, Fig. 1 includes (in the lower right-hand corner) the magnetization curves of Ba_2Zn_2Y at 293°K; a neutron-diffraction study of this compound showed no helicoidal spin configuration.

The Curie temperatures of the $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ and Ba_2Zn_2Y crystals were equal: 390 ± 10°K.

3. MAGNETOCRYSTALLINE ANISOTROPY

The magnetocrystalline anisotropy was investigated by the torque method in the temperature range between 77°K and the Curie temperature. Figure 2 shows the dependence of the torque L on the angle ϑ between the c axis and the direction of magnetization in $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ at 293°K (curve 1) and at 77°K (curve 2). For comparison, the same figure shows, in the lower left-hand corner, the torque curve of Ba_2Zn_2Y at 293°K. Ba_2Zn_2Y has an easy magnetization plane at all temperatures but the behavior of the torque curves

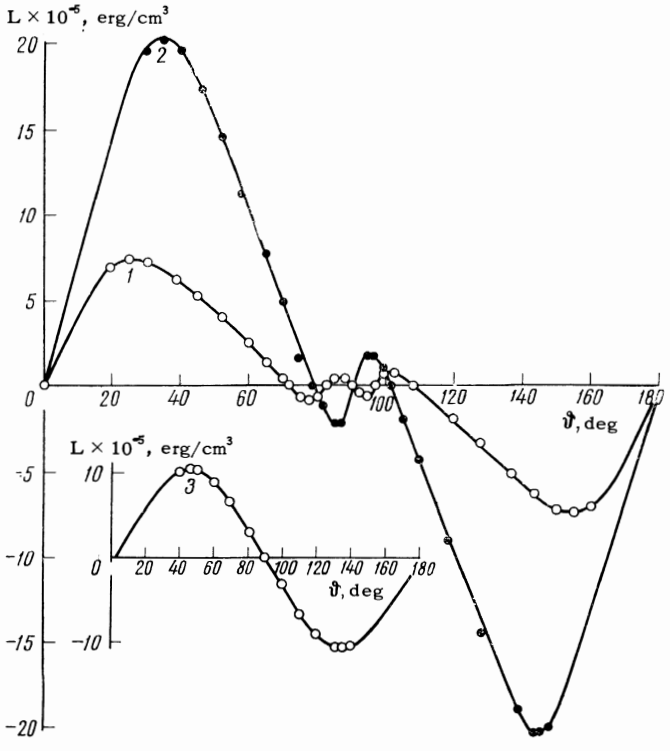

FIG. 2. Torque curves of $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$: 1) at 293°K, 2) at 77°K, and the torque curve of Ba_2Zn_2Y at 293°K (3).

of $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ is more complex. It is found that, at room temperature, not only directions in the basal plane ($\vartheta_0 = 90°$) but also directions lying on the surface of a cone with a vertex angle $\vartheta_0 = 72°$, measured with respect to the hexagonal axis, are all easy magnetization directions. At liquid nitrogen temperature (curve 2 in Fig. 2) the torque curve of $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ is characterized by an easy magnetization cone with a vertex angle $\vartheta_0 = 78°$.

The combination of an easy magnetization plane and a cone is observed only in the temperature range 270—330°K. At lower temperatures, $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ crystals have only an easy magnetization cone which is not greatly affected by temperature (Fig. 3); above this range of temperatures these crystals still have a cone whose vertex angle rapidly decreases with increase in temperature and, 20 deg below the Curie point, i.e., at 370°K, the cone becomes an easy magnetization axis ($\vartheta_0 = 0°$). The combination of an easy magnetization plane and a cone exists only in fields above 7000 Oe. In weaker fields the direction of the resultant magnetization lies in the basal plane.

It is worth mentioning that there have been no published reports of crystals with the Y structure having an easy magnetization cone and particularly an easy magnetization axis (with the exception of Ba_2Co_2Y, which has an easy magnetization cone at 77°K).

4. NEUTRON-DIFFRACTION INVESTIGATIONS

A neutron-diffraction investigation of the magnetic structure was carried out using the same samples as were employed in the magnetic studies. The neutron diffraction patterns near the 006, 009, 0012 and other peaks had magnetic superstructure reflections (satellites) whose positions varied with temperature and with an external magnetic field.

Figure 4 shows the diffraction patterns re-

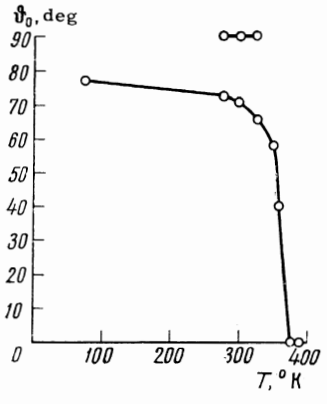

FIG. 3. Temperature dependence of the vertex angle ϑ_0.

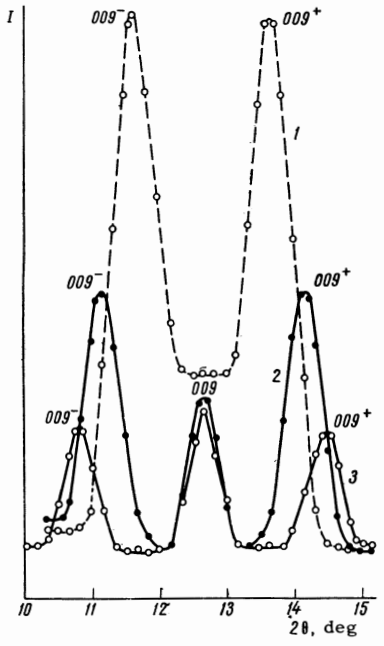

FIG. 4. Neutron-diffraction patterns of $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ in the region of the 009 reflection (1 denotes the number of pulses per minute, in relative units; θ is the Bragg angle). The neutron wavelength was 1.07 Å. 1) 4.2°K; 2) 293°K; 3) 370°K.

corded when a sample and a counter were set in motion (1:2) near the 009 reflection. The satellites symmetrical with respect to the 009 structure reflection are denoted by 009^- and 009^+. Such superstructure can be explained by the existence [in $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ crystals] of helicoidal spin ordering with a propagation vector directed along the c axis. Helicoidal ordering of spins in a hexagonal ferrite has already been found by us in a neutron-diffraction study of the system $BaCo_xW$.[2] Figure 4 shows clearly the temperature dependence both of the intensity of the satellites and of their positions with respect to the structure peak.

At 4.2°K, the superstructure period τ is 60 Å. This period remains constant up to 230°K. In the temperature range 240–270°K the period of the helicoid decreases to 43 Å. The period τ then remains at this value up to 320°K. In the temperature range 320–350°K, τ again decreases, to 32 Å, and then remains constant until the helicoidal ordering is destroyed at 370°K, i.e., below the Curie temperature (400°K). The application of a magnetic field in the ab plane alters the helicoidal structure period from 43 Å in H = 0 at 293°K to 29 Å in H = 1500 Oe at 293°K. The period then remains at 29 Å up to H = 7000 Oe. Further increase of the field intensity destroys the helicoid: the intensity of the satellites decreases and they tend to merge with the central peak. The satellites disappear completely in fields stronger than 14 000–16 000 Oe.

The intensity of the 009 structure reflection also increases considerably with increase of the field intensity; in H = 0 this reflection has practically no magnetic contribution. A magnetic field directed along the c axis does not alter the parameters of the helicoid. The helicoidal ordering is destroyed completely by fields of 18 000–20 000 Oe directed along the c axis.

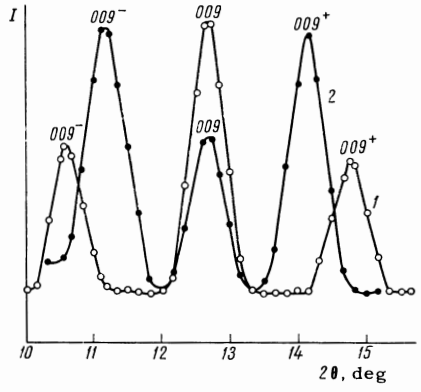

FIG. 5. Neutron-diffraction patterns of $(Sr_{0.8}Ba_{0.2})_2Zn_2Y$ in the region of the 009 reflection at 293°K. The neutron wavelength was 1.70 Å. 1) H = 3000 Oe, $\mathbf{H} \perp \boldsymbol{\varepsilon}$; 2) H = 0.

Figure 5 shows the diffraction patterns near the 009 reflection at 293°K, recorded in the following magnetic fields: 1) H = 3000 Oe, perpendicular to the neutron scattering vector $\boldsymbol{\epsilon}$; 2) H = 0.

5. DISCUSSION OF RESULTS

We shall now compare the experimental data with a phenomenological theory used by Enz.[1] The resultant magnetic moment in each basal plane is rotated by an angle α with respect to the direction of the magnetic moment in the neighboring basal plane. Enz ascribes this angle to the competition between two exchange interactions: a positive exchange interaction between the nearest-neighbor planes, and a negative interaction between the next-nearest planes. The energy of this interaction, per molecule, is written in the following form

$$E_{exc} = -\mu_s^2(I_1 \cos\alpha - I_2 \cos 2\alpha), \qquad (1)$$

where μ_S is the spontaneous magnetic moment per molecule. The exchange parameters are $I_1 > 0$ and $I_2 < 0$. If $|4I_2| > |I_1|$, the helicoidal structure is stable and the angle α_0 (the equili-

brium value of the angle α) is found from the condition

$$\cos\alpha_0 = -I_1/4I_2. \quad (2)$$

In weak fields the magnetization σ of a helicoidal antiferromagnet depends linearly on the magnetic field intensity

$$\sigma = \chi H, \quad (3)$$

where the susceptibility χ is

$$\chi = -\left\{32I_2\sin^4\frac{\alpha_0}{2}[1+2\cos\alpha_0(1+\cos\alpha_0)]\right\}^{-1}. \quad (4)$$

In a field H = 0, the magnetic moments have a purely helicoidal distribution. In weak fields, in which the linear law (3) applies, the helicoidal structure is still retained. When the magnetic field intensity is increased to a critical value H_c, the helicoid becomes deformed. The value of H_c is given by the expression

$$H_c = -7.76\mu_s I_2\sin^4\frac{\alpha_0}{2}. \quad (5)$$

We shall use this treatment to discuss the experimental results. We shall calculate H_c at room temperature. Using the data given in Sec. 4, we find the angle α_0 (or more correctly, the average value of this angle since the distances between ions along the c axis of the Y structure are not equal). We use Eq. (3) to calculate χ and Eq. (4) to calculate I_2 from the known values of $\alpha_0 = 13^\circ$ and χ: $I_2 = -60\times10^{23}$ G^2/erg. Consequently, it follows from Eq. (2) that $I_1 = 230\times10^{23}$ G^2/erg. Now we use Eq. (5) to determine the critical field H_c, giving $H_c = 570$ Oe. Comparison of the calculated value, $H_c = 570$ Oe, with the experimental value, $H_c = 700$ Oe, shows satisfactory agreement between experiment and theory.

We estimate the parameters of the exchange interaction between the nearest neighbors n_1 and between the second-nearest neighbors n_2:

$$n_1 = \mu_s^2 I_1 = 12\cdot10^{-14}\text{ erg}, \quad n_2 = \mu_s^2 I_2 = 3\cdot10^{-14}\text{ erg}.$$

These values are comparable with the exchange interaction energy determined from the Curie temperature: $E_{exc} = kT = 5.5\times10^{-14}$ erg. (A calculation of the magnetic dipole interaction energy $E_{dip} = \mu_s^2/r^3$, where r is the distance between layers, gives a value which is two orders of magnitude lower.)

The study of the magnetic anisotropy shows that the equilibrium direction of the magnetization in a field of about 7000 Oe changes from a plane to a cone. The theory of the helicoidal magnetic structure does not deal with such a transition. Therefore, we cannot compare theory with experiment for these and stronger fields.

We are unable to explain the simultaneous existence of a plane and a cone of easy magnetization. The expansion of the magnetic anisotropy energy in the form of a power series

$$E_c = k_1\sin^2\vartheta + k_2\sin^4\vartheta \quad (6)$$

predicts only the existence of a cone of easy magnetization and does not describe the observed behavior of the torque curve near the basal plane. It is quite likely that a calculation of the torque curves should include the exchange energy of Eq. (1), as well as the magnetic anisotropy energy of Eq. (6).

[1] U. Enz. J. Appl. Phys. Suppl. to 32, No. 3, 22S (1961).

[2] I. I. Yamzin, R. A. Sizov, I. S. Zheludev, T. M. Perekalina, and A. V. Zalesskiĭ, JETP 50, 595 (1966), Soviet Phys. JETP 23, 395 (1966).

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