Magnetic transformations in indium-substituted hexaferrites

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It is shown by the Mössbauer spectroscopy technique that the hexaferrite $BaIn_3M$ is characterized by the presence of an angular magnetic-disorder structure which goes over to a noncollinear structure on lowering of temperature and application of a magnetic field. The anomalous magnetic properties of ferrites having such compositions (e.g. absence of saturation in strong magnetic fields) are due to this fact, since collinear structures are not observed even at helium temperatures.

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Investigations of substituted hexaferrites $BaMe_xFe_{12-x}O_{19}$ (Me = Al^{3+}, Ga^{3+}, In^{3+} etc.) show[1-5] that the presence of anomalous magnetic properties of these compounds may be connected with the character of the disposition of the Me^{3+} ions over the crystal lattice sites. Thus, A selective disposition of the indium cations in the 2b and $4f_1$ sites leads, on the one hand, to a weakening of the principal exchange bonds and to a hexagonal bonding between the spinel blocks, and on the other hand causes a breaking of the magnetic bonds directly in the spinel blocks themselves.[5] As a result, the collinear axial model of the spin arrangement[6] becomes unstable. Mössbauer investigations of the system $BaIn_xFe_{12-x}O_{19}$ show[5] that changes of the composition in the range $2 < x < 3$ lead to an effect analogous to the "order-disorder" phase transition type, The change in the degree of ordering of the spin system is accompanied here by an anomalous decrease in the Mössbauer-spectra line intensities, a decrease that does not agree with the change in the number of In^{3+} ions in the samples.[7] All this offers evidence (in addition to the absence of magnetic saturability of the hexaferrites $BaIn_{2.4}Fe_{9.6}O_{19}$ [1] that a disordered canted magnetic structure is produced.

To observe the possible magnetic transitions in such ferrites, we have carried out Mössbauer investigations of the $BaIn_3Fe_9O_{19}$ sample at a temperature ranging to 5°K, with a magnetic field H = 16 kOe applied to the sample. In this sample, the sites 2b and $4f_1$ turn out to be almost fully occupied by nonmagnetic ions, and therefore they cannot take part in the exchange interaction. The unsubstituted Fe^{3+} ions form in this case a weakly bound complex. The magnetic coupling between these complexes is small because of the low magnetic-anisotropy energy in such compounds. This is also evidenced by the Mössbauer spectrum of the sample at room temperature, which constitutes an asymmetrical quadrupole doublet with E = 0.64 mm/sec (Fig. 1a).

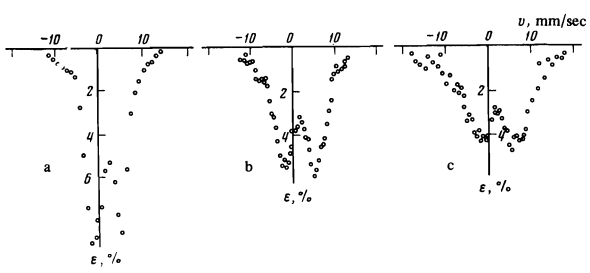

FIG. 1. Mössbauer spectra of $BaIn_3Fe_9O_{19}$ sample under various measurement conditions: a) T = 300°K, H_0 = 0; b) T = 300°K, H_0 = 16 kOe; c) T = 250°K, H_0 = 16 kOe.

Measurements performed with the temperature lowered from 300 to 100°K show that the form of the spectrum remains unchanged and retains all its characteristic features. At the same time, application of an external magnetic field H = 16 kOe on the sample broadens the spectral lines even at room temperature (Fig. 1b). A slight lowering of the temperature (to 250°K) greatly enhances the influence of the external magnetic field (Fig. 1c) and contributes to the transition of the sample into a magnetically ordered state.

In the absence of an external magnetic field, a strongly smeared transition into a magnetically-ordered state is observed only when the measurement temperature decreases below 100°K (Fig. 2a).

A qualitative estimate of this difference in the transition temperatures can be obtained by regarding the investigated sample as an aggregate of weakly bound complexes whose magnetic moments relax independently of one another under the influence of the temperature. The behavior of this aggregate of complexes is analogous to the behavior of minute supermagnetic particles, a behavior described by the expression[8]

$$\tau=\tau_0 \exp\left(KV+\alpha MH_{an}+\beta MH\right)/T. \qquad (1)$$

The first term in the parentheses in this expression describes the temperature relaxation of an individual particle, the second takes into account the magnetic dipole interaction between particles in the anisotropy field, and the third characterizes the influence of the external magnetic field on the relaxation time.

Taking the values of K, M. H_{an} for the $BaIn_3Fe_9O_{19}$ sample from [1,2] we obtain, in the absence of an external magnetic field, a temperature of about 70°K for transition to the magnetically ordered state, which is close enough to the experimentally observed 90°K.

Application of an external magnetic field will contribute, as follows from (1), to the transition of the sample

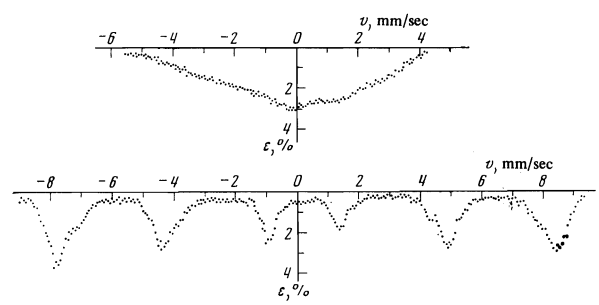

FIG. 2. Magnetic hyperfine structure of the nuclear levels of the iron ions in the $BaIn_3M$ sample: a) T = 78°K, b) T = 5°K.

into a magnetically ordered state at higher temperatures. A quantitative estimate with the aid of (1) is made difficult by the lack of information on the value of β, but even at $\beta = 0.1$ the transition temperature will amount to 200–250°K at an external magnetic field intensity of approximately 20–30 kOe.

Since our measurements did not yield a well-resolved magnetic hyperfine structure of the Mössbauer spectrum (Figs. 1c and 2a), we have performed measurements with the temperature lowered to 5°K. The Mössbauer spectrum of the sample (Fig. 2b) is in this case a sextet with strongly broadened lines. The average value of the local magnetic field, obtained from this spectrum, is 510 kOe, which is much less than the saturation fields typical of compounds of trivalent iron.[9]

Thus, Mössbauer investigations of a $BaIn_3Fe_9O_{19}$ sample indicate that a lowering of the temperature below 100°K leads to a strongly smeared transition from the disordered canted magnetic structure of the sample into a noncollinear one. Measurements with an external magnetic field H = 16 kOe on the sample show that this transition is initiated in this case at a much higher temperature ($\approx$ 250°K). Lowering the measurement temperature to 80°K leads to an increase of the degree of ordering, but no transition to a collinear magnetic structure is observed even at the very lowest temperatures.

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