Magnetic phase transitions in samarium iron garnet and hypothesis of Ising ordering

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The low-temperature magnetic orientational phase transition in $Sm_3Fe_5O_{12}$ has been studied experimentally. In the ground state (as $T \to 0$ K) this ferrimagnetic has a magnetic structure of the $\langle uv0 \rangle$ type. This result and also the $\langle uv0 \rangle \rightleftarrows \langle 110 \rangle \rightleftarrows \langle 111 \rangle$ phase transitions, which are observed with increasing temperature in $Sm_3Fe_5O_{12}$, are explained in the model of Ising ordering of the magnetic moments of Sm^{3+} ions.

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In a recent study[1] of the Mössbauer effect in polycrystalline samples of samarium iron garnet ($Sm_3Fe_5O_{12}$) it was observed that this ferrimagnet exhibits an orientational transition in addition to the well-studied, first-order, orientational transition from $\langle 111 \rangle$ axes to $\langle 110 \rangle$ axes at $T_{t2} = 65.7$ K (Refs. 2 and 3): Below $T_{t1} = 18$ K, the magnetic moments move away from the $\langle 110 \rangle$ axes, and at $T \lesssim 10$ K they are oriented along $\langle 100 \rangle$ axes. This interpretation, however, contradicts the data of Ref. 4, according to which the $\langle 100 \rangle$ directions are not easy magnetic axes at 4.2 K.

In an effort to identify the nature of the low-temperature orientational transition in $Sm_3Fe_5O_{12}$, we have studied the initial rf susceptibility, the torque in $\{100\}$ and $\{110\}$ planes, and the magnetization along various directions over the temperature range 1.8–77 K, using single crystals grown in two laboratories.

Our results show that at 18 K there is in fact an orientational magnetic transition involving a rotation of the magnetic moments away from the $\langle 110 \rangle$ axes. This result can be seen clearly in Fig. 1a, which shows curves of the torque in the $\{100\}$ plane (the conditions $L = 0$ and $\partial L / \partial \phi < 0$ along the easy magnetic direction). This conclusion has been confirmed by the susceptibility measurements: At $T_{t1} = 18.2$ K there is a sharp maximum on the $\chi(T)$ dependence (Fig. 1b). According to our data, however, the $\langle 100 \rangle$ axes are not easy magnetic directions at low temperatures; i.e., the conclusion of Ref. 1 that $\langle 100 \rangle$ magnetic phases exist in $Sm_3Fe_5O_{12}$ is not confirmed. Figure 2 shows the temperature dependence of the orientation of the magnetic moments in the $\{100\}$ in $Sm_3Fe_5O_{12}$. We see that ϕ_L, the angle between the easy axis and the $\langle 100 \rangle$ axis, falls off gradually with decreasing temperature, but even at 2.86 K it is 29–31°.

It thus follows from the experimental data that the low-temperature transition at 18 K in $Sm_2Fe_5O_{12}$ is a second-order phase transition from the collinear $\langle 110 \rangle$ phase to the $\langle uv0 \rangle$ angular phase. In cubic magnetic materials, this transition may be described formally only by incorporating an eighth-order interaction in the magnetic-anisotropy energy (in addition to the fourth- and sixth-order interactions, which lead

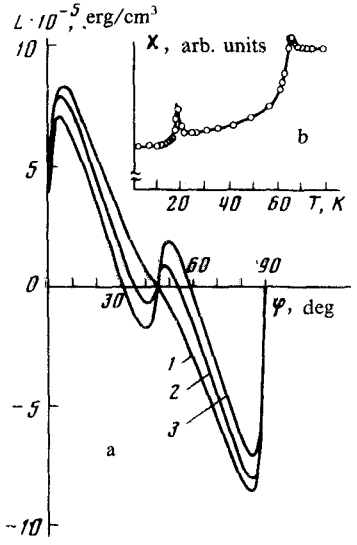

FIG. 1. a: Torque in the {100} plane in $Sm_3Fe_5O_{12}$ in a field of 10.2 kOe. 1—21 K; 2—15.5 K; 3—7.8 K. b: Temperature dependence of the initial susceptibility of $Sm_3Fe_5O_{12}$.

to only first-order phase transitions).[5] From the angular dependence of the torque in the {100} and {110} planes, we can estimate the magnetic-anisotropy constants of $Sm_3Fe_5O_{12}$ at 4.2 K: $K_1 = -10^7$, $K_2 = 10^8$, and $K_3 = 2.5 \times 10^7$ erg/cm^3. Since $|K_1| < |K_2|, |K_3|$, we are led to doubt whether the magnetic-anisotropy energy of $Sm_3Fe_5O_{12}$ can be written as a series in harmonic invariants at low temperatures. Furthermore, for most of the rare-earth iron garnets it would not be correct to write the magnetic-anisotropy energy as an additive part of the free energy, since in these magnetic materials the interaction of the rare-earth ions with the molecular field produced by the iron sublattice (this interaction leads to the magnetic ordering of the rare-earth ions) is weaker than the interaction of these ions with the crystal field.

For a theoretical description of the magnetic phase transitions observed in $Sm_3Fe_5O_{12}$, we will accordingly take a different approach, adopting the assumption that the magnetic ordering of the Sm^{3+} ions is of an Ising nature. We have used a similar model previously to describe the magnetic properties of holmium-yttrium iron garnets.[6–8] In the garnet structure the rare-earth ions occupy six nonequivalent dodecahedral sites (D_2 symmetry), with local axes in different orientations with respect to

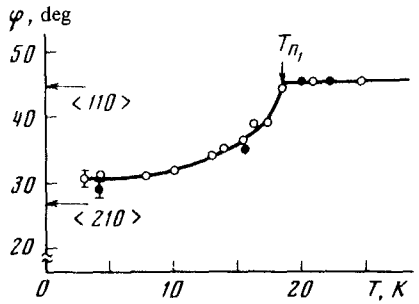

FIG. 2. Temperature dependence of the orientation of the magnetic moments of $Sm_3Fe_5O_{12}$ in the {100} plane. Open circles—from torque measurements; filled circles—from magnetization measurements.

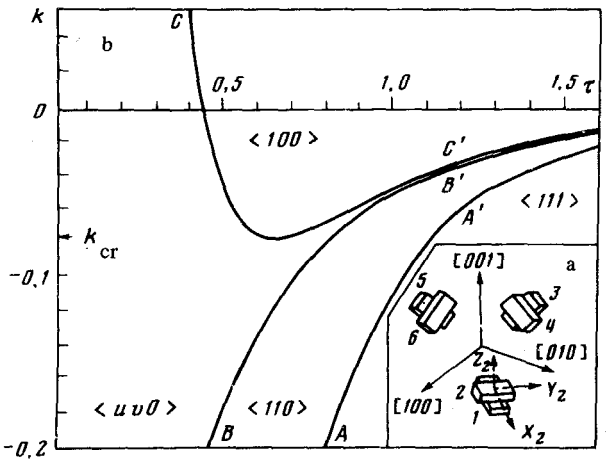

FIG. 3. a—Nonequivalent positions of the rare-earth ions in the garnet structure; b—theoretical phase diagram of $Sm_3Fe_5O_{12}$.

the crystallographic axes (Fig. 3). We assume that the Ising axis of the Sm^{3+} ion at the i-th site in $Sm_3Fe_5O_{12}$ coincides with the local $x_i(y_i)$ axis, i.e., with a $\langle 110 \rangle$ axis. For an arbitrary orientation of the molecular field, a magnetic moment of this type can thus be oriented either along or opposite the corresponding $\langle 110 \rangle$ axis, in accordance with the sign of the field projection on this axis. It follows from purely geometric considerations that for this type of magnetic ordering the resultant moment of the samarium sublattice, M_{Sm}, may be oriented only along $\langle 210 \rangle$ or $\langle 111 \rangle$ axes. The state with $\mathbf{M}_{Sm} \| \langle 111 \rangle$, however, is unstable: It corresponds to a maximum of the energy.[1)] In this model, therefore, the magnetic moment of the resultant samarium sublattice must be collinear with $\langle 210 \rangle$ axes. At equilibrium, the magnetic moment of the iron sublattice, $\mathbf{M}_{Fe}$, is parallel to $\mathbf{M}_{Sm}$. At 0 K, therefore, according to this simple Ising model, the easy magnetic axes are the $\langle 210 \rangle$ directions, for which we have an angle $\phi = 26.6°$, in approximate agreement with the value measured at liquid-helium temperature: $\phi_L = 29$–$31°$.

This result was derived by taking into account only the ground Ising doublet of the Sm^{3+} ion. For a more detailed description of the magnetic structure of $Sm_3Fe_5O_{12}$ at 0 K and of its conversion with increasing temperature, we would have to take into account the higher-lying levels and also the anisotropy of the iron sublattice. In this case the thermodynamic potential of the crystal per rare-earth ion can be written

$$F = -(T/6) \sum_{i=1}^{6} \ln [\, 2\mathrm{ch}\, (\mu_x \lambda \mathbf{M}_{Fe} \mathbf{x}_i / T)] + K(T)(\gamma_x^2 \gamma_y^2 + \gamma_y^2 \gamma_z^2 + \gamma_x^2 \gamma_z^2). \quad (1)$$

The first term describes the contribution of the ground Ising doublet, split by the molecular field,

$$E_i = \pm \mu_x (\lambda \cdot \mathbf{M}_{Fe} \cdot \mathbf{x}_i) = \pm \mu_x \cdot \lambda \cdot \mathbf{M}_{Fe} \cdot \hat{\gamma} \cdot \mathbf{x}_i \quad (2)$$

μ_x is the expectation value of the magnetic-moment operator for the Ising doublet; λ is

the molecular-field constant; $\mathbf{x}_i$ is a unit vector along the x_i direction; and $\vec{\gamma} = \mathbf{M}_{Fe}/M_{Fe}$. The second term describes the anisotropy caused by the higher-lying levels of the Sm^{3+} ion and by the anisotropy of the iron sublattice.

A minimization of the thermodynamic potential in (1) with respect to $\vec{\gamma}$ shows that the following four phases can occur in this model: $\mathbf{M}_{Fe} \| \langle 111 \rangle$, $\mathbf{M}_{Fe} \| \langle 110 \rangle$, $\mathbf{M}_{Fe} \| \langle 100 \rangle$ and $\mathbf{M}_{Fe} \| \langle uv0 \rangle$. Figure 3b shows the magnetic phase diagram in terms of the variables $\tau = T/\mu_x \lambda M_{Fe}$ and $K = 6K/\mu_x \lambda M_{Fe}$. At low temperatures ($\tau \to 0$), there is the angular phase $\langle uv0 \rangle$, in which the magnetic moment lies in the $\{100\}$ plane and makes some angle ϕ_L with the $\langle 100 \rangle$ axis. If $k = 0$, the magnetic moment is parallel to $\langle 210 \rangle$ ($\phi_L = 26.6°$), and as $|k|$ increases the magnetic moment rotates slightly away from this direction. From our experimental data we find $K = -(0.3\text{–}0.6)$ in the limit $T \to 0$ for $Sm_3Fe_5O_{12}$. As the temperature is raised, there is a second-order transition to the $\langle 110 \rangle$ phase if $k < k_{cr} = -0.07$. The transition line, BB', is described by

$$k(\tau) = 1/2 \left[\operatorname{th}(1/\tau) + 2 \operatorname{th}(1/2\tau) + (1/\tau)\operatorname{th}^2(1/2\tau) - 2/\tau \right]. \quad (3)$$

A further increase in the temperature leads to a transition from the $\langle 110 \rangle$ phase to the $\langle 111 \rangle$ phase. This is a first-order transition, and it occurs along the line AA':

$$k(\tau) = -12\tau \ln[\operatorname{ch}^4(1/2\tau)\operatorname{ch}(1/\tau)/\operatorname{ch}^3(\sqrt{2}/\sqrt{3}\tau)]. \quad (4)$$

We thus see that the model adopted here not only successfully describes the sequence of phase transitions $\langle uv0 \rangle \rightleftharpoons \langle 110 \rangle \rightleftharpoons \langle 111 \rangle$ observed experimentally with increasing temperature in $Sm_3Fe_5O_{12}$ but also correctly predicts the nature of these transitions.

In this model there may also be a phase of the $\langle 100 \rangle$ type (at $k > k_{cr}$). The lower boundary of this phase is the first-order transition line CC'; this phase does not occur in samarium iron garnet.

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[1)]Three of the six Sm^{3+} ions in this state are demagnetized; i.e., their ground doublet is degenerate. This circumstance is responsible for the instability, because of the magnetic Jahn-Teller effect.[9]

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