Lowest-temperature magnetic phase transition of samarium iron garnet

G. Balestrino
Istituto di Elettronica dello Stato Solido del Consiglio Nazionale delle Ricerche, Via Cineto Romano 42, I-00156 Roma, Italy

S. Geller
Department of Electrical Engineering, University of Colorado, Boulder, Colorado 80302

B. Antonini, P. Paroli, and A. Tucciarone
Istituto di Elettronica dello Stato Solido del Consiglio Nazionale delle Ricerche, Via Cineto Romano 42, I-00156 Roma, Italy

(Received 6 December 1983)

New Mössbauer spectroscopic analysis confirms the spin reorientation below 18 K of samarium iron garnet first reported by Geller and Balestrino. However, both the Mössbauer spectroscopic analysis and measurements with an easy-axis monitor indicate that the axis of easy magnetization at the end of the spin reorientation region is along a ⟨740⟩ direction.

In a Mössbauer spectroscopic investigation of the magnetic transitions of samarium iron garnet,[1] an additional transition was found to begin at approximately 18 K and to end at approximately 10 K. At the time, it was concluded that below 10 K, the easy axis of magnetization was[2] ⟨100⟩. Subsequently, Babushkin *et al.*[3] confirmed the existence of this transition, but, by means of torque measurements, showed that the final easy axis is close to ⟨210⟩ rather than to ⟨100⟩.

We have repeated the Mössbauer spectroscopic investigation with better equipment. The fitting of tetrahedral lines $I=\frac{1}{2}$, $m_I=\pm\frac{1}{2}\rightarrow I=\frac{3}{2}$, $m_I=\pm\frac{3}{2}$ received particular attention. But more important is the determination of the angles that the local (originally cubic) axes make with the magnetization $\vec{M}$.

In Ref. 1, it was shown that in the region of the spin reorientation below 18 K, the highest-symmetry magnetic space group of samarium iron garnet must be $I2'/c'$. This space group requires the tetrahedral Fe^{3+} ions to be distributed over four crystallographically nonequivalent sets of sites in the ratio 8:8:4:4. Because it is to be expected that the relative positions of the ions do not change much with decreasing temperature, the lines from different sets may overlap.

It was also shown in Ref. 1 that for the ⟨100⟩ end point of the transition, the highest-symmetry magnetic space group must be $Ibc'a'$, which requires the tetrahedral Fe^{3+} ions to be distributed equally over three crystallographically nonequivalent sets of sites. Again, overlap is probable; in fact, the spectra (Figs. 1 and 2; see also Figs. 1 and 2 of Ref. 1) show resolution into only two tetrahedral lines, the lower-velocity line of $I=\frac{1}{2}$, $m_I=-\frac{1}{2}\rightarrow I=\frac{3}{2}$, $m_I=-\frac{3}{2}$ (Fig. 1), appearing to have twice the intensity of the higher-velocity line.

Now, although the magnetic space group of the samarium iron garnet must be $Ibc'a'$ (or lower), if it had a ⟨100⟩ easy direction of magnetization at 10 K, one might expect overlap that would make the spectrum appear to be that of a tetragonal crystal.[1] This is true because significant structural changes are not expected and the angles made by the (origi-

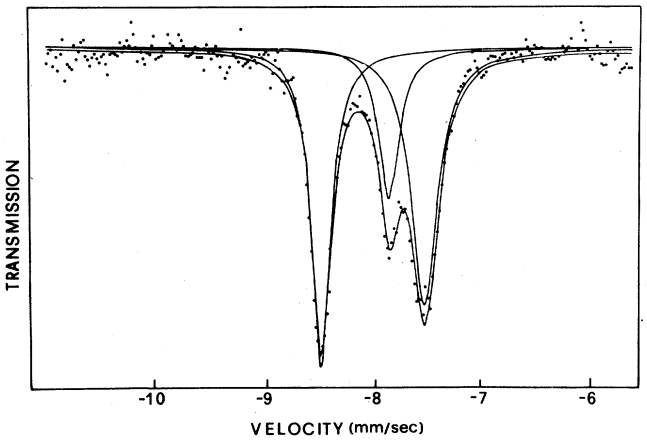

FIG. 1. Two-line fit of the ($I=\frac{1}{2}$, $m_I=-\frac{1}{2}\rightarrow I=\frac{3}{2}$, $m_I=-\frac{3}{2}$) tetrahedral lines of samarium iron garnet.

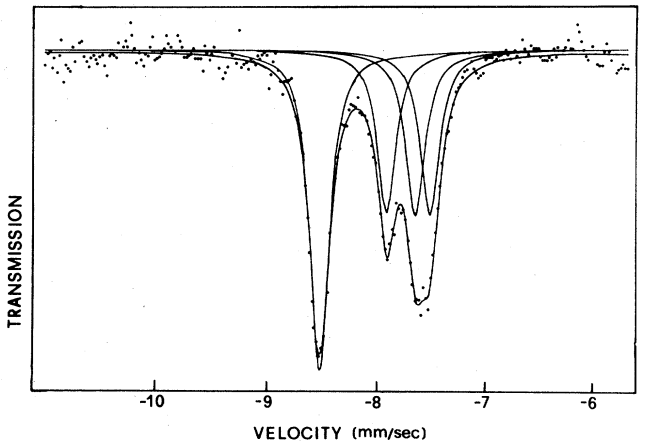

FIG. 2. Three-line fit of the $I=\frac{1}{2}$, $m_I=-\frac{1}{2}\rightarrow I=\frac{3}{2}$, $m_I=-\frac{3}{2}$) tetrahedral lines of samarium iron garnet.

nally) cubic symmetry axes with the magnetization $\vec{M}$ should not be significantly different even though the magnetic space groups must be different. In the highest-symmetry tetragonal case, the tetrahedral Fe^{3+} ions are distributed over two sets of sites in the ratio 16:8.

Fitting of the two peaks with just two Lorentzians, without constraints, gave a 26% larger full width at half maximum (FWHM) for the lower- than for the higher-velocity peak of the transition $I=\frac{1}{2}$, $m_I=-\frac{1}{2}\rightarrow I=\frac{3}{2}$, $m_I=-\frac{3}{2}$ (Fig. 1); the area under the lower-velocity peak is therefore 2.17 times that of the higher-velocity peak.

A three-line fit to *exactly* the same data is shown in Fig. 2; again, the fitting was done without constraints. Within experimental error these peaks have the same FWHM and intensity.[4] Thus it is clear that the three-line fit is considerably better than the two-line fit. This suggests only that the magnetic space group to which the samarium iron garnet crystal belongs is either $I2'/c'$ (Ref. 5) or $Ibc'a'$. In the former case, $\vec{M}$ lies in a (cubic) {100} plane; in the latter it must be along a ⟨100⟩ direction.

If the quadrupole splitting (QS) of the tetrahedral sites is known, it is possible to calculate, from the peak energies, the angles

$$\theta_1,\theta_2,\theta_3=\cos^{-1}(1-\cos^2\theta_1-\cos^2\theta_2)^{1/2}\ ,$$

that the local, originally cubic ⟨100⟩ axis make with $\vec{M}$.

At room temperature, samarium iron garnet belongs to the rhombohedral magnetic space group (Ref. 1) $R\bar{3}c'$, in which case, because of the angular factor $(3\cos^2\theta-1)$, the quadrupole interaction for the tetrahedral sites vanishes. Therefore, the fitting of a spectrum taken at 20 K (Ref. 6) was used to calculate the QS. In the calculation, it was assumed that the magnetic interaction is much larger than the quadrupole interaction. The deduced value for QS for the tetrahedral sites is 0.83 $mm\,s^{-1}$. The values of θ_1 and θ_2 from the $I=\frac{1}{2}$, $m_I=-\frac{1}{2}\rightarrow I=\frac{3}{2}$, $m_I=-\frac{3}{2}$ transition are 60.2° and 30.0°, respectively, and from the $I=\frac{1}{2}$, $m_I=\frac{1}{2}\rightarrow I=\frac{3}{2}$, $M_I=\frac{3}{2}$ transition, 61.0° and 30.2°, respectively. These results show clearly that the easy axis of the Fe^{3+} ion sublattices ($\vec{M}_{Fe}$) at 5 K is of the ⟨$u,v,0$⟩ type[7] and makes an angle of 30° with the nearest ⟨100⟩ axis.

The "easy-axis monitor"[8] was also used to determine the position of $\vec{M}$ in samarium iron garnet at temperatures between 27 and 4.2 K.

For this experiment, a single crystal of samarium iron garnet was grown from a flux and oriented by Laue photography. A small (100) disk was cut from the crystal; the orientation was rechecked by Laue photography, and found to be within 2°. The disk was annealed in air for 2 h at 900 °C to remove internal stresses, after which it was mounted in the easy-axis monitor. The specimen was then cooled to liquid-helium temperature and then allowed to warm freely, during which the rotation of the easy axis was recorded. The results are shown in Fig. 3.

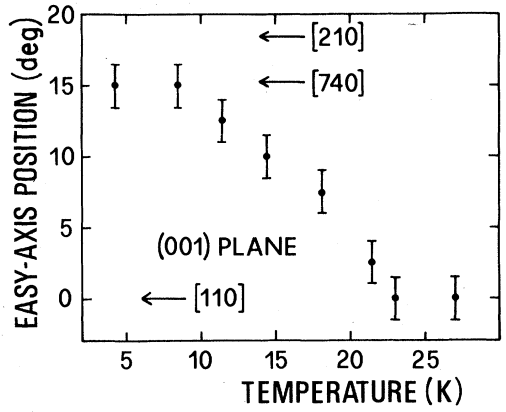

FIG. 3. Easy-axis position measured with respect to a [110] axis, as a function of temperature.

The temperature of the second-order ⟨$u,v,0$⟩ → ⟨110⟩ transition appears, from these measurements, to be somewhat higher than that reported earlier[1] and confirmed by Babushkin *et al.*[3] This is attributed to anomalous behavior of the specific heat of samarium iron garnet near the transition: an increase in the heat capacity of the specimen near the transition temperature could cause the temperature of the thermocouple to be higher than the specimen temperature.

At and below about 8 K (Fig. 3), $\vec{M}$ is oriented in a direction 15° from a [110] direction in the {100} plane of the crystal, showing that the easy axis of the crystal is 30° from the nearest ⟨100⟩ direction at the end of the spin reorientation. Babushkin *et al.*[3] have given reasonable arguments as to why $\vec{M}$ should be along a ⟨210⟩ direction. However, a ⟨210⟩ direction makes an angle of 26.6° with the nearest ⟨100⟩ direction. The experimental results, including that of Babushkin *et al.*,[3] indicate that the angle is 30°, which means that $\vec{M}$ is unequivocally not along a ⟨210⟩, but rather along a ⟨740⟩ direction. This may be attributable to the truly lower symmetry of the crystal and suggests the need for a crystal structure determination of a single domain crystal at 5 K.

---

[1]S. Geller and G. Balestrino, Phys. Rev. B **21**, 4055 (1980).

[2]We use the symbol of a "form" of zone axes to indicate that the easy direction may be along any of the *originally* cubic directions in the form.

[3]G. A. Babushkin, V. A. Borodin, V. D. Doroshev, A. K. Zvezdin, R. Z. Livitin, and A. J. Popov, Pis'ma Zh Eksp. Teor. Fiz. **35**, 28 (1982) [JETP Lett. **35**, 34 (1982)].

[4]The results are as follows: the FWHM of the two lower-veolcity peaks are the same and are 3% wider than the higher-velocity peak. The largest difference in peak heights is 0.7%.

[5]It is assumed that the peaks from the two fourfold sets of positions overlap exactly.

[6]At this temperature, the phase is orthorhombic (Ref. 1) with ⟨110⟩ easy axis.

[7]As indicated earlier, space-group considerations established that this is the case in the spin reorientation region thought originally to go to $\vec{M}$ parallel to ⟨100⟩. The magnetic point group $2'/m'$ to which the crystal belongs at all temperatures less than 18 K *requires* that $\vec{M}$ be of the ⟨$u,v,0$⟩ type.

[8]B. Antonini, S. D'Angelo, A. Foco, B. Maturi, and P. Paroli, IEEE Trans. Magn. **MAG-18**, 1562 (1982).