

MAGNETIC PROPERTIES OF RARE-EARTH IRON GARNETS IN THE CURIE-POINT REGION

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The magnetic properties of rare-earth iron garnets are investigated in the neighborhood of the Curie temperature. It is shown that the paraprocess susceptibility of the Gd, Tb, Dy, Ho, and Er garnets in the immediate vicinity of the Curie point is, in the first approximation, inversely proportional to the value of the effective magnetic moment of the rare-earth ion, determined at 0° K. For the Sm and Eu garnets, this rule breaks down because of a transition of the  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  ions into an excited state on increase of temperature. The magnetic moments of the  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  ions in the Curie-point region are estimated on the basis of the intensity of the paraprocess. It is found that near the Curie point (to the right of it), the temperature dependence of the initial magnetic paraprocess-susceptibility of all the iron garnets investigated follows the relation  $\chi_0^{-1} = A(T - \Theta)^\gamma$ , where  $\gamma \approx 4/3$ .

It is known that the Curie points of all rare-earth iron garnets differ little from one another and are close to the Curie point of yttrium iron garnet; that is, they are basically determined by the exchange interaction of the iron sublattices. On the other hand, it is also known that the antiferromagnetic coupling between the "resultant" iron sublattice and the rare-earth lattice manifests itself right at the Curie point. This is evidenced by the abrupt decrease of the Curie point in rare-earth iron garnets as compared with the yttrium ferrite.

It is of interest to elucidate how this antiferromagnetic coupling affects the character of the magnetic transformation in iron garnets. For this purpose, we have carried out similar measurements of magnetization curves of polycrystalline iron garnets of Y, Sm, Eu, Gd, Tb, Dy, Ho, and Er. Measurements were also made on single crystals of the iron garnets of Y and Tb; the data from these measurements agreed with the measurements on polycrystalline specimens.

From the magnetization isotherms of the rare-earth iron garnets, measured in the Curie-point region, the temperature variation of the spontaneous magnetization  $\sigma_S$  was determined by construction of curves<sup>[1]</sup>

$$H/\sigma = f(\sigma^2). \quad (1)$$

By way of example, Fig. 1 shows the isotherms (1), which describe the paraprocess near the Curie point, for samarium iron garnet. The departures

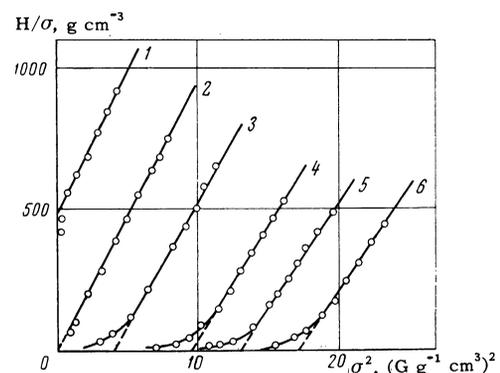


FIG. 1. Dependence of  $H/\sigma$  and  $\sigma^2$  for Sm iron-garnet: curve 1,  $t = 290.8^\circ\text{C}$ ; 2,  $289.0^\circ\text{C}$ ; 3,  $287.4^\circ\text{C}$ ; 4,  $286.0^\circ\text{C}$ ; 5,  $284.8^\circ\text{C}$ ; 6,  $283.4^\circ\text{C}$ .

of the experimental points from a straight line at weak fields is connected, on the one hand, with the effect of "remnants" of technical magnetization processes; and, on the other, with a possible effect of fluctuation processes not taken into account in the thermodynamic theory.<sup>[1]</sup> The value of the spontaneous magnetization was determined by extrapolation of the isotherms to the  $\sigma^2$  axis ( $H = 0$ ). The Curie temperature was determined on the basis of the isotherm that gives  $\sigma_S = 0$ .

In other researches, the Curie temperature has been determined by the usual construction of a tangent to the steepest part of the  $\sigma_S(T)$  curve, where  $\sigma_S$  is determined from the  $\sigma(H)$  curves by the method of linear extrapolation to  $H = 0$ ; this is inaccurate near  $\Theta$ .

$R_3Fe_5O_{12}$	$\mu_{\text{eff.}}^2 / \mu_B^2$	$a_{\Theta} \cdot 10^2$	$\gamma (\pm 0.03)$	$\Theta^*$ , °K, data from literature*	$\Theta$ , °K our data
Y	0.00	24.0	1.30	560	551
Sm	0.14	21.0	1.30	573	562
Eu	0.74	16.0	1.29	568	563
Gd	7.00	11.5	1.33	564	556
Tb	7.73	11.0	1.30	568	553
Dy	7.30	11.5	1.30	563	552
Ho	6.73	12.0	1.30	567	548
Er	5.07	15.0	1.30	556	542

\*The value  $\Theta^*$  is determined from  $\sigma_S(T)$  curves, by construction of a tangent to the steepest part.

The value of the Curie temperature determined by the method described in [1], by means of the isotherms (1), is more accurate. This method provides a possibility of extracting values of the Curie temperature in the case of diffuse transformations. The presence of "tails" of the spontaneous magnetization inevitably leads to divergence between values of the Curie temperature determined by different methods. It is to be expected that this divergence will be larger, the more diffuse the magnetic transformation is.<sup>1)</sup>

The table gives values  $\Theta^*$  of the Curie temperature, according to the data in the literature, and values  $\Theta$  of the Curie temperature, determined by the method described in [1], for all the garnets investigated.

The Curie temperatures found from the isotherms (1) are somewhat lower than the temperatures determined according to the decrease of magnetization. Figure 2 gives, by way of example, the function  $\sigma_S(T)$  found by both methods, for yttrium garnet. The Curie point is determined as  $\Theta = 560^\circ\text{K}$  according to the drop in the  $\sigma_S(T)$  curve; this agrees with the value obtained in other researches.<sup>[2]</sup> The same figure shows the temperature  $\Theta = 551^\circ\text{K}$  found according to the curves (1).

From the isotherms (1) we also determined the dependence of the paraprocess susceptibility  $\chi_p$  in the neighborhood of the Curie point, and also the coefficient  $a_{\Theta}$  that describes the intensity of the paraprocess right at the Curie point. The coefficient  $a_{\Theta}$  was determined from the relation  $a_{\Theta} = (\beta_{\Theta})^{-1/3}$ , where  $\beta_{\Theta}$  is the slope of the straight part of the curves (1) at the Curie point.<sup>[1]</sup>

Figure 3 shows the temperature dependence of the paraprocess susceptibility of terbium iron garnet in the Curie-point region, for various values of the magnetic field. The form of the curves is similar to that obtained for ordinary ferromag-

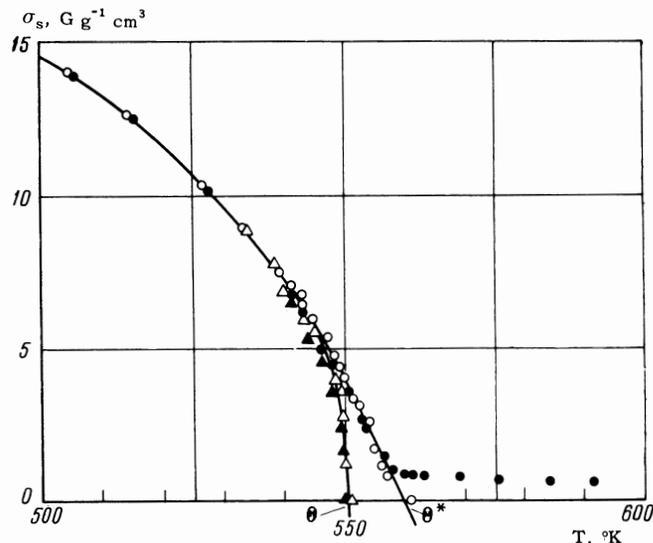


FIG. 2. Dependence of  $\sigma_S$  on  $T$ , found by two different methods:  $\Delta$ , from curves  $H/\sigma = f(\sigma^2)$  for single-crystal and polycrystalline specimens of yttrium garnet;  $\circ$ , from curves  $\sigma(H)$  for the same specimens.

nets. The maximum of  $\chi_p$ , as for ordinary ferromagnets, is shifted slightly to the high-temperature side of the Curie point. This points to a similarity between the magnetic transformation in iron garnets and the transformation in normal ferromagnets, for example in nickel.

Figure 4 shows the temperature dependence of  $\chi_p$  in a field of 260 Oe. The table gives values of  $a_{\Theta}$  for all the iron garnets studied by us.

From the table and from Fig. 4 it is evident that the amount of the paraprocess is different for all the iron garnets. The most intense paraprocess is

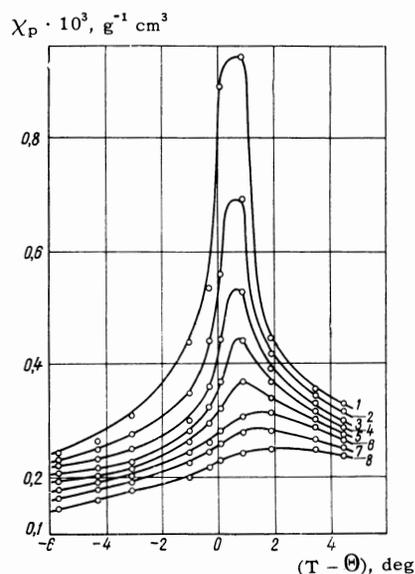


FIG. 3. Dependence of  $\chi_p$  on  $(T - \Theta)$  for Tb iron garnet: curve 1,  $H = 260$  Oe; 2, 390 Oe; 3, 520 Oe; 4, 660 Oe; 5, 790 Oe; 6, 920 Oe; 7, 1050 Oe; 8, 1180 Oe.

<sup>1)</sup>This problem has been discussed repeatedly in the literature. For more details see [1(b)].

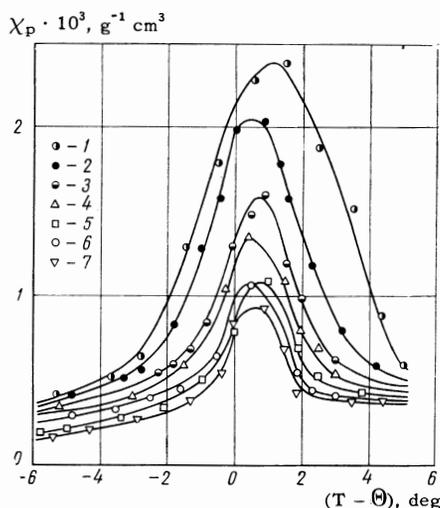


FIG. 4. Dependence of  $\chi_p$  on  $(T - \Theta)$  in field 260 Oe for iron garnets of: 1, Y; 2, Sm; 3, Eu; 4, Er; 5, Gd; 6, Dy; 7, Tb.

observed in yttrium iron garnet. In the Gd, Tb, Dy, Ho, and Er iron garnets, the paraprocess is considerably smaller; the diminution of the paraprocess is due to the antiferromagnetic orientation of the magnetic moments of the rare-earth ions. It is therefore natural to compare the intensity of the paraprocess with the value of the magnetic moment  $\mu_{\text{eff}}$  of the rare earth ion in the rare-earth sublattice of the corresponding iron garnet. The table gives values of  $\mu_{\text{eff}}$ , which were calculated from the saturation magnetization of the rare-earth iron garnets at  $0^\circ\text{K}$ .<sup>[3, 4]</sup> Figure 5 shows the dependence of the parameter  $a_\Theta$  on  $\mu_{\text{eff}}$  for all the ferrites investigated.

From the table and from Fig. 5 it is evident that the parameter  $a_\Theta$  for Gd, Tb, Dy, Ho, and Er iron garnets decreases, in first approximation, in inverse proportionality to  $\mu_{\text{eff}}$ , determined at  $0^\circ\text{K}$ . It is evident that the points corresponding to these ferrites fit the straight line well. Even the point for yttrium iron garnet, where the  $\text{Y}^{3+}$  ion has  $\mu_{\text{eff}} = 0$ , fits the line. The parameter  $a_\Theta$  has its

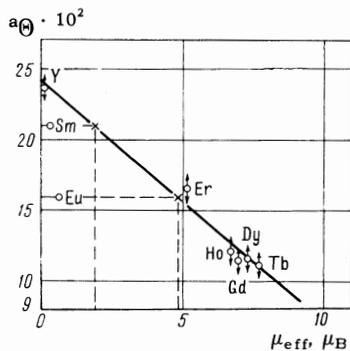


FIG. 5. Dependence of the intensity of the paraprocess on the value of the effective magnetic moment of the rare-earth ion.

largest value for this ferrite. The decrease of the intensity of the paraprocess in rare-earth iron garnets, as compared with yttrium, is explained by the fact that at the Curie point an antiparallel orientation of the magnetic moments of the rare-earth ions and of the iron ions persists. The rare-earth ions are in an effective negative exchange field of the iron sublattice. On increase of the field, the increase of the magnetization of the iron sublattice in consequence of the paraprocess entails an increase of the magnetization of the rare-earth sublattice in the antiparallel direction, since there is a negative exchange coupling between these sublattices. This diminishes also the paraprocess of the ferrite as a whole.

In Fig. 5 it is evident that the points for Sm and Eu ferrites depart widely from the straight line. The reason for this is that on increase of temperature, the  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  ions transfer to an excited state, and consequently their magnetic moments in the Curie-point region differ widely from the values at  $0^\circ\text{K}$ . We shall consider this problem in more detail for samarium ferrite.

In samarium iron garnet, in contrast to iron garnets of heavy rare-earth metals, the magnetization of the samarium sublattice, in the low-temperature region, is directed parallel to the magnetization of the resultant iron sublattice; this is explained as follows. Although the spin moments of the samarium ions are directed antiparallel to the spin moments of the iron (because of the negative exchange coupling), the orbital moment of the  $\text{Sm}^{3+}$  ion, in accordance with Hund's rule, is directed opposite to the spin moment; that is, it points in the direction of the spin moment of the iron ion. Thus the magnetization of samarium ferrite should be somewhat larger than the magnetization of yttrium ferrite, as has also been confirmed by experi-

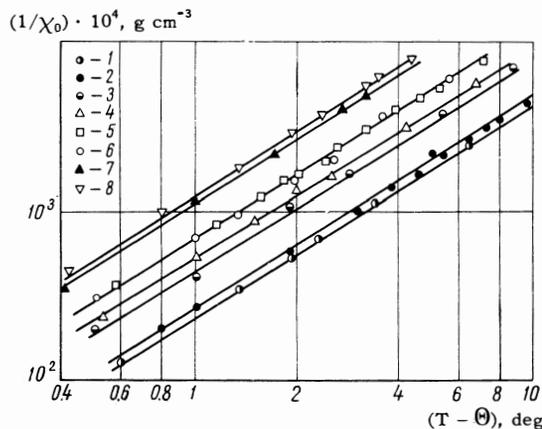


FIG. 6. Dependence of  $1/\chi_0$  on  $(T - \Theta)$  on a logarithmic scale for all ferrites investigated: 1, Y; 2, Sm; 3, Eu; 4, Er; 5, Gd; 6, Dy; 7, Ho; 8, Tb.

ment.<sup>[3]</sup> It is at low temperatures, however, that this situation occurs. With increase of temperature, in consequence of the transition of the  $\text{Sm}^{3+}$  ion into an excited state, its magnetic moment decreases appreciably.<sup>[5]</sup> It is therefore of interest to carry out measurements of paraprocess curves of samarium iron garnet at elevated temperatures, in particular near the Curie point.

Our measurements showed that the paraprocess in Sm iron garnet in the Curie-point region is somewhat smaller than in yttrium; that is, the paraprocess in this ferrite has the same character as in iron garnets of the heavy rare-earth elements. The reason for this is that in the excited state, the orbital component of the magnetic moment of the  $\text{Sm}^{3+}$  ion decreases greatly, so that in the Curie-point region the spin component becomes the dominant component of the magnetic moment of the ion, and it is antiparallel to the spin moment of the iron atom. Therefore the intensity of the paraprocess in samarium garnet in this temperature range should be less than in yttrium garnet, as is also confirmed by the results of our measurements. From Fig. 5 it is possible, on the basis of the values of  $a_0$ , to estimate the values of  $\mu_{\text{eff}}$  for the  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  ions for temperatures in the Curie-point region. For the  $\text{Sm}^{3+}$  ion,  $\mu_{\text{eff}} \approx 2\mu_B$ ; for  $\text{Eu}^{3+}$ ,  $\mu_{\text{eff}} \approx 5\mu_B$ . These values differ significantly from those determined at  $0^\circ\text{K}$  (see the table).

Our data on the intensity of the paraprocess testify to the existence of antiparallel correlations of the spins in the Curie-temperature region in rare-earth garnets.

As is well known, great attention has been paid recently to detailed investigation of spin correlations near magnetic phase transitions, since the results of these investigations are of interest for the development of a statistical theory of phase transitions.<sup>[6-9]</sup> Spin correlations in simple ferromagnets lead to a complication of the temperature dependence of the initial magnetic paraprocess susceptibility  $\chi_0$  near the Curie point (on the high-temperature side). According to theory,<sup>[6]</sup> this dependence has the form<sup>2)</sup>  $\chi_0^{-1} = A(T - \Theta)^\gamma$ .

Experiments have shown that for Fe, Ni, and Gd, within the limits of experimental error,  $\gamma \approx 4/3$ .

The question arises whether this relation holds for ferrimagnets, in particular for iron garnets in which antiparallel spin correlation occurs.

We determined the value of  $\gamma$  for the ferrites investigated by extrapolation of the straight parts of the curves (1) to the value  $H/\sigma = 0$ . Above the Curie point, these straight lines intersect the axis of ordinates, with an intercept equal to  $1/\chi_0$ . Further, straight lines  $1/\chi_0$  against  $(T - \Theta)$  on a logarithmic scale were plotted, and from the slopes of these lines the values of  $\gamma$  were determined for all the ferrites investigated (Fig. 6). It was found that for the iron garnets of Y, Sm, Eu, Gd, Tb, Dy, Ho, and Er the values of  $\gamma$  were close to  $4/3$  (see the table).

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<sup>2</sup> B. Lax and K. J. Button, Microwave Ferrites and Ferrimagnetics (McGraw-Hill Book Company, Inc., New York, 1962).

<sup>3</sup> S. Geller, H. J. Williams, R. C. Sherwood, J. P. Remeika, and G. P. Espinosa, Phys. Rev. 131, 1080 (1963).

<sup>4</sup> S. Geller, J. P. Remeika, R. C. Sherwood, H. J. Williams, and G. P. Espinosa, Phys. Rev. 137, A1034 (1965).

<sup>5</sup> J. H. Van Vleck, Theory of Electric and Magnetic Susceptibilities (Oxford University Press, London, 1932).

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<sup>7</sup> G. A. Baker, Jr., Phys. Rev. 124, 768 (1961)

<sup>8</sup> J. S. Kouvel and M. E. Fisher, Phys. Rev. 136, A1626 (1964).

<sup>9</sup> S. V. Tyablikov, Metody kvantovoi teorii magnetizma (Methods in the Quantum Theory of Magnetism), Nauka, 1965 (Translation: Plenum Press, New York, 1967).

<sup>2)</sup>The coefficient A depends on the magnetic and crystalline structures.