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THE PARAPROCESS IN RARE-EARTH IRON GARNETS IN THE CURIE-TEMPERATURE REGION

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The paraprocess is investigated in rare-earth monocrystalline iron garnets (Y, Er, Ho, Dy, Tb, Gd) in the Curie-temperature region. The measurements establish that the paraprocess constant has a maximum value for Y iron garnet and diminishes upon replacement of Y by heavy rare-earth ions in the dodecahedral sublattice. The data obtained are explained on the basis of molecular-field theory, on the assumption that the exchange interaction of the rare-earth ions with the Fe ions is determined, in first approximation, not by the total but by the spin moments of the rare-earth ions.

EXPERIMENT shows that in rare-earth iron garnets, the paraprocess has a significant magnitude over a broad temperature interval, from the lowest temperatures up to the Curie point. The reason lies in the fact that the rare-earth sublattice c is acted upon by a comparatively weak ($\sim 10^5$ Oe) effective exchange field from the iron sublattices a and d. Consequently the external field can exert an influence on the magnetic moments of the rare-earth ions, which are disoriented by heat motion. Far from the Curie point, this paraprocess can be described easily by means of relations obtained from the molecular-field model[1,2], since the exchange field of the a–d interaction is large ($\sim 10^7$ Oe) and the paraprocess in the a–d sublattice is very small.

The present paper treats the paraprocess in rare-earth iron garnets in the Curie-point region. In this region the paraprocess is of more complicated character, because, in consequence of the rapid diminution of the magnetization of the iron sublattices, the exchange field of the a–d interaction also diminishes, and as a result there occurs an intense paraprocess due to the a–d sublattices. In consequence of the interaction of the a–d and the c sublattices, the paraprocess produced by the external field will not be made up additively of the amounts of the paraprocess of the two sublattices, but will have a more complicated character.

We shall consider this question in more detail. The energy of interaction of the rare-earth ions with the resultant sublattice a–d and of the Fe ions can be written in the form

$$(\varepsilon_{\text{exch}})_{c-(a-d)} = -g_S \mu_B S H_{\text{eff}} \tag{1}$$

where S is the value of the spin of the rare-earth ion, and where H_{eff} is the effective field due to the negative exchange interaction of the rare-earth sublattice c with the resultant sublattice a–d of the iron ions. In paper[1] it was shown that the relation (1) leads to a satisfactory description of the change of the compensation points Θ_c and of the "low-temperature" points Θ_l of Gd, Tb, Dy, Ho, and Er iron garnets and also of mixed yttrium-terbium iron garnets.

If we assume that the interaction under consideration is determined not by the spin S but by the total moment J (that is, if we write this energy in the form $\epsilon_{\text{exch}} = -\mu_B g_J J H_{\text{eff}}$), then it is impossible to explain the orderly change of the values of Θ_c and Θ_l in the iron garnets. In this case, H_{eff} changes almost by orders of magnitude from Gd iron garnet to Tu ferrite, which is not justified physically. Thus in our calculation we suppose that $(\epsilon_{\text{exch}})_{c-(a-d)}$ is determined by the exchange interaction not of the total moments J, but solely of the spin moments S.

The analysis carried out in[1] showed that from one rare-earth garnet to another, H_{eff} is approximately constant for all rare-earth iron garnets (ions of the yttrium subgroup), since it is determined principally by the "resultant" sublattice a–d. This fact will be used in our calculations. Furthermore, we neglect exchange interaction within the rare-earth sublattice, which is small[3].

A graphic proof of the fact that the energy of interaction of the sublattice c with the resultant sublattice a–d can be taken in the form (1) is provided by the experimentally observed linear dependence of the "low-temperature" point Θ_l on the spin S of the rare-earth ion[4]. In fact, an abrupt change of long-range magnetic order occurs at a temperature (the "low-temperature" point Θ_l) such that the energy kT of heat motion is comparable with the energy of interaction of the rare-earth ion with the biassing effective exchange field (due to the action of the sublattices of iron ions); that is, $2\mu_B S H_{\text{eff}}$. Hence we get the relation

$$\Theta_l \backsim 2\mu_B H_{\text{eff}} S / k,$$

which describes the data well with $H_{\text{eff}} = \text{const}$.

For the case under consideration, the Brillouin functions that describe the temperature behavior of the relative magnetization of the resultant sublattice of Fe ions and of the sub-lattice of rare-earth ions can be written, respectively, in the form

$$\bar{S}_1 / S_1 = B_{S_1}(y_1), \quad \bar{S}_2 / S_2 = B_{S_2}(y_2), \tag{2}$$

where

$$y_1 = \frac{\mu_{10}}{\nu_1 kT} H + \frac{2 z_{12} J_{12} S_1}{kT} \bar{S}_2 + \frac{2 z_1 J_1 S_1}{kT} \bar{S}_1, \tag{3}$$

$$y_2 = \frac{\mu_{20}}{\nu_2 kT} H + \frac{2 z_{21} J_{21} S_2}{kT} \bar{S}_1. \tag{4}$$

Here quantities related to the sublattice a–d are distinguished by the index 1, those related to the sublattice c by the index 2. The mean value of the spin of an ion is denoted by $\overline{S}_i$, the value of the magnetic moment at 0°K, calculated for a molecule, by μ_{i0}; S_i is the spin of an ion; ν_i is the number of atoms in a molecule for the sublattice; J_{ij} is the integral of exchange interaction of an atom of sublattice i with the atoms of sublattice j; z_{ij} is the number of nearest neighbors in sublattice j for an atom of sublattice i; and k is Boltzmann's constant.

Near the Curie point, we may expand the functions (2) as series in powers of y_1 and y_2 ($y_1 \ll 1$ and $y_2 \ll 1$ for $T \approx \Theta_C$) and may restrict ourselves to the first two terms of the series for the sublattice a—d, to the first term only of the series in the case of the sublattice c, since the exchange interaction of the rare-earth ions with the ions of the Fe sublattice is almost orders smaller than the exchange interaction within the iron sublattice, which also determines the Curie temperature ($2z_{12}J_{12}S_1\overline{S}_2 \ll 2z_1J_1S_1\overline{S}_1$, $2z_{12}J_{21}S_2\overline{S}_1 \ll 2z_1J_1S_1\overline{S}_1$). Furthermore, in the expansion we consider the case of a small magnetic field ($2z_{12}J_{12}S_1\overline{S}_2 \gg \mu_{20}H/\nu_2$).

On taking into account what has been said, we get a system of two equations for the magnetization, near the Curie point, of the sublattices a—d and c, calculated per molecule:

$$\alpha\mu_1 + \beta\mu_1^3 = H - \gamma\mu_2, \tag{5}$$

$$\delta\mu_2 = H - \gamma\mu_1. \tag{6}$$

Here the first equation describes the field dependence of the magnetization μ_1 for the sublattice of Fe ions, the second equation the field dependence of the magnetization μ_2 for the sublattice of rare-earth ions; α, β, γ, and δ are coefficients, expressed in terms of the atomic constants of the ions and of the temperature:

$$\alpha = 3\left(1 - \frac{S_1+1}{3}\frac{2z_1J_1S_1}{kT}\right)\frac{\nu_1S_1kT}{(S_1+1)\mu_{10}^2}, \tag{7}$$

$$\beta = \frac{\nu_1S_1[(S_1+1)^2+S_1^2]kT}{30\mu_{10}^4}\left(\frac{2z_1J_1S_1}{kT}\right)^3, \tag{8}$$

$$\gamma = \frac{\nu_1S_1S_2}{\mu_{10}\mu_{20}}2z_{12}J_{12}, \tag{9}$$

$$\delta = \frac{\nu_2kT}{\mu_{20}^2}\frac{3S_2}{S_2+1}. \tag{10}$$

By use of the relation (6), μ_2 can be eliminated in equation (5). As a result we get

$$(\alpha - \gamma^2/\delta)\mu_1 + \beta\mu_1^3 = (1-\gamma^2/\delta)H. \tag{11}$$

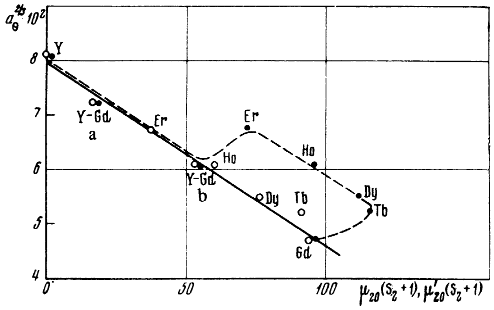
At the Curie point the coefficient $\alpha - \gamma^2/\delta = 0$, and from equations (6) and (11) we get for the resultant magnetization per molecule

$$\mu = \left[1 - \frac{2}{3}\frac{\nu_1z_{12}J_{12}S_1}{k\Theta_C}\mu_{20}(S_2+1)\right]^{3/2}\left(\frac{H}{\beta}\right)^{1/3} + \frac{S_2+1}{3S_2}\frac{\mu_{20}^2}{\nu_2kT}H. \tag{12}$$

The susceptibility at the Curie point is found by differentiation of the magnetization with respect to the field H:

$$\chi = \left[1 - \frac{2}{3}\frac{\nu_1z_{12}J_{12}S_1}{k\Theta_C}\mu_{20}(S_2+1)\right]^{3/2}\frac{1}{3\beta^{1/3}H^{2/3}} + \frac{S_2+1}{3S_2}\frac{\mu_{20}^2}{\nu_2kT}. \tag{13}$$

The last term in the expression (13) is the usual paramagnetic susceptibility of the rare-earth ions in the external magnetic field H, due to orientation of the spins of the rare-earth ions along the field H. An estimate showed that this part of the susceptibility is small (5% of the total susceptibility), and therefore we neglect it. Thus the resulting susceptibility is determined by the first term of the expression (13).



Dependence of the paraprocess constant on $\mu_{20}(S_2 + 1)$ (solid curve) and on $\mu_{20}'(S_2 + 1)$ (dashed curve) for garnets of the rare-earth elements and for mixed Y-Gd iron garnets: a – $Y_{2.47}Gd_{0.53}Fe_2O_{12}$; b – $Y_{1.3}Gd_{1.7}Fe_5O_{12}$.

In the expression (13), the quantity $\frac{1}{3}\beta^{-1/3}H^{-2/3}$ is the susceptibility of the paraprocess of the "resultant" iron sublattice when the magnetic moment $\mu_{20} = 0$. Since the magnetic moment of the Y ion is zero, this quantity $\frac{1}{3}\beta^{-1/3}H^{-2/3}$ can be taken as the paraprocess susceptibility of Y ferrite-garnet.

Experiment shows that the paraprocess at the Curie point in ferrite-garnets can be described by the relation

$$\chi = {}^1/_3 a_\Theta H^{-2/3}, \tag{14}$$

where a_Θ is a constant of the paraprocess for a given iron garnet. On denoting the quantity $\beta^{-1/3}$ by $a_{\Theta Y}$, where $a_{\Theta Y}$ is the paraprocess constant for Y iron garnet, and on substituting it into the relation (13) with allowance for (14), we get for the paraprocess constant of rare-earth ferrite-garnets the following relation:

$$a_{\Theta R}^{2/3} = a_{\Theta Y}^{2/3}\left[1 - \frac{2}{3}\frac{\nu_1z_{12}S_1}{k\Theta_C}\mu_{20}(S_2+1)\right]. \tag{15}$$

As is seen from the relation (15), the value of $a_{\Theta R}^{2/3}$ decreases with increase of the product $\mu_{20}(S_2 + 1)$.

We have made measurements of the values of a_Θ for Y, Er, Ho, Dy, Tb, and Gd iron garnets and for the system of mixed Y-Gd iron garnets at $T = \Theta_C$. The investigations were carried out on monocrystalline specimens of small dimensions in fields up to 15 kOe, on a pendulum magnetometer[5,6] 1). An experimental determination was made of the value of a_Θ^{-3}, which is the slope of the magnetization isotherms H/σ as a function of σ^2; in the fields investigated, in the Curie-temperature region, these are straight lines with a slowly changing slope (in a temperature interval of about 15°, the value of a_Θ changes by no more than 3%).

The measurements showed that the coefficient a_Θ decreases with advance along the series of rare-earth iron garnets in the order Y, Er, Ho, Dy, Tb, Gd. In the mixed yttrium-gadolinium garnets, a_Θ decreases with

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1) The investigations on monocrystals, carried out over a wider range of fields, enabled us to improve the accuracy of the data, by measurement of a_Θ, obtained earlier [5] on polycrystalline specimens.

increase of the content of Gd ions replacing the Y ions.

In the figure, the solid line gives the experimental dependence of $a_{\Theta R}^{2/3}$ on $\mu_{20}(S_2 + 1)$ found by us. For μ_{20} we took the values of the saturation magnetization at 0°K obtained experimentally for iron garnets in the paper of Geller et al.[7] It is seen that in agreement with formula (12), this dependence is in fact of linear character.

The results obtained can be interpreted as follows. In rare-earth iron garnets there is a negative exchange interaction between the sublattice of rare-earth ions and the sublattice of Fe ions; therefore on increase of the magnetization of the iron sublattice (in consequence of the paraprocess), there occurs an increase of this negative exchange interaction, that is an increase of H_{eff}, and consequently also an increase of the magnetization of the rare-earth sublattice in the opposite direction. This effect is especially large in the immediate neighborhood of the Curie point, since here the paraprocess of the iron sublattices attains its maximum value. Therefore the paraprocess constant in rare-earth iron garnets in the Curie-point region is smaller than in Y iron garnet.

It follows from formula (15) that the paraprocess constant changes from one rare-earth garnet to another in direct proportion to the product $\mu_{20}(S_2 + 1)$. This deduction is in agreement with the results that were obtained on the basis of molecular-field theory, with similar assumptions about the nature of the exchange interaction, for the Θ_c and Θ_l points of rare-earth iron garnets. It was shown[1] that Θ_c changes from one rare-earth garnet to another also in proportion to $\mu_{20}(S_2 + 1)$, whereas Θ_l changes in proportion to the spin S.

We point out that the linear dependence of $a_{\Theta R}^{2/3}$ on $\mu_{20}(S_2 + 1)$ is observed only when the experimental value of μ_{20} is taken. It is seen from the figure that if instead of μ_{20} we take the theoretical value $\mu'_{20} = \nu\mu_B g_J J$ (that is, the value for free ions), then the linear dependence of $a_{\Theta R}^{2/3}$ on $\mu'_{20}(S_2 + 1)$ is violated. The experimental values of μ_{20} (determined from low-temperature measurements) are somewhat smaller than μ'_{20}. The difference between the values of μ_{20} and μ'_{20} is at present explained on two possible grounds:

1) Partial quenching of the orbital moment of the rare-earth ion by the crystalline field.

2) Existence of noncollinear magnetic moments of the sublattices of the ferrite.

This suggests that these causes, responsible for the lowered values of the magnetic moments of the rare-earth sublattices, apparently remain valid also near the Curie point. Similar deductions can be made also by consideration of the Θ_c points of the rare-earth garnets.

The possibility is not ruled out that a more accurate calculation of the paraprocess susceptibility of rare-earth iron garnets, with allowance for ''partnership'' of the orbital moment of the rare-earth ions in the exchange interaction with the Fe ions, might make a contribution to the value of the paraprocess susceptibility. It is to be expected, however, that the ''orbital'' correction to this exchange interaction should be, presumably, very small.

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