

# Effects of unidirectional exchange anisotropy in ferrites

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**Abstract.** A number of anomalous effects occurring at the compensation and Curie points of ferrites are explained on the basis of the concept of unidirectional exchange anisotropy. Among these are the violation of ‘evenness’ for the magnetostrictive, magnetoresistive, and magnetocaloric effects, the antiferromagnetic paraprocess, and some other phenomena. Based on the analysis of these effects, a piezomagnetic effect is predicted to occur in ‘weak-sublattice’ ferrimagnets.

## 1. Introduction

In 1998, it was 50 years since the French physicist L Néel, Nobel prize winner, published his paper [1] in which a theory of a new class of magnetic materials, ferrimagnets, was put forward. This class of magnets includes ferrites, intermetallics — rare earth metal–iron (cobalt) compounds, rocks — magnetite, titanium magnetite, and so on. Although studies of some of these materials [2, 3] were started long before the publication of Néel’s paper, Néel was the first to establish the fact that such materials possess many-sublattice magnetic structures and so exhibit more intricate and varied magnetic properties as compared to simple (one-sublattice) ferromagnets, such as iron and nickel.

The paper by Néel initiated a great stream of experimental and theoretical investigations of ferrites and other ferrimagnets. Ferrites have found wide application as effective magnetic materials in radio engineering, microwave technology and some other fields of technology. However, a number of questions concerning their magnetic properties still remain unclarified. It is particularly true for

ferrimagnets possessing a so-called ‘weak’ magnetic sublattice (with an anomalous — asymptotic — temperature dependence of spontaneous magnetization). Among these materials are iron garnets and heavy rare-earth metal intermetallics as well as some substituted spinel-type ferrites (such as lithium ferrite–chromite). The distinguishing features of these are the occurrence of the compensation points  $\theta_c$  (low-temperature order–disorder magnetic phase transitions, or the  $T_B$  points), as well as some other phenomena.

At one time, these phenomena were interpreted on the basis of the molecular field approximation by the author and Nikitin [4, 5]. However, a number of other phenomena characteristic of these magnets have not found an adequate explanation. Among these are the violation of the ‘evenness’ for the magnetostrictive, magnetoresistive and magnetocaloric effects due to the paraprocess, and a sharp increase in the coercive force at the compensation point  $\theta_c$  found experimentally.

In the present paper, which may be considered as a continuation of review [6], it is shown that the ‘weak-sublattice’ ferrimagnets are systems where we have to do with a manifestation of unidirectional (or ‘one-sided’) exchange anisotropy. At one time, this type of anisotropy was considered in a number of experimental and theoretical works [8–12] for some ferromagnet–antiferromagnet systems. The present paper demonstrates that this type of anisotropy manifests itself in ‘weak-sublattice’ ferrimagnets as well. The use of the concept of unidirectional exchange anisotropy allows one to interpret the above-mentioned unexplained effects in ‘weak-sublattice’ ferrites.

An analysis of experimental data obtained for the Mg–Mn and Mn–Zn spinel-type ferrites and yttrium iron garnet (which do not possess a ‘weak’ sublattice at low temperatures) has revealed that they become ‘weak-sublattice’ ferrimagnets in the neighborhood of the Curie point. This means that near the Curie point (and directly above it) they are magnets in which unidirectional exchange anisotropy is present. This fact makes it possible to explain a number of anomalous effects occurring in the indicated temperature

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Received 2 February 1999  
*Uspekhi Fizicheskikh Nauk* 169 (7) 797–804 (1999)  
Translated by V M Matveev; edited by S N Gorin

regions which has not found an adequate explanation up to the present.

## 2. Three types of ferrimagnets

Depending on the relationship between the values of inter- and intrasublattice exchange interactions, there come into existence different peculiarities of the magnetic properties of ferrimagnets. Based on the analysis of these peculiarities, ferrites may be subdivided into three types.

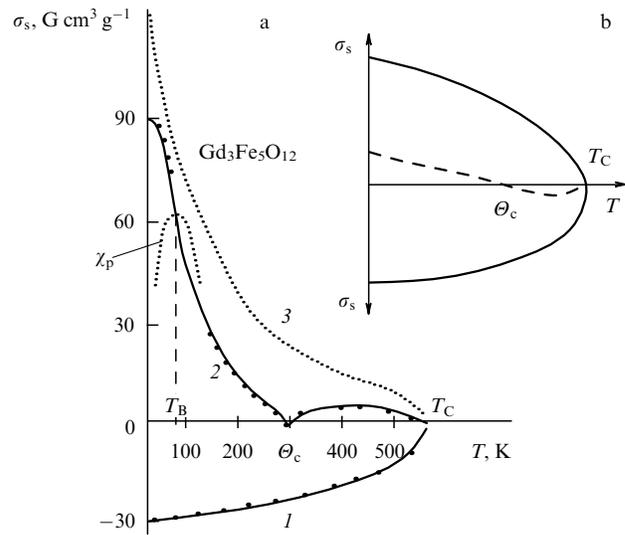
Ferrimagnets in which the intersublattice exchange interactions far exceed the intrasublattice ones should be ascribed to the first type. These ferrites possess a so-called ‘Néel’ magnetic structure (these are precisely the ferrimagnets that were examined by Néel [1]). They are characterized by strictly antiparallel orientations of the magnetization vectors of sublattices. The term ‘non-compensated antiferrimagnet’ is applicable to them, since there exists a strong antiferromagnetic exchange interaction between sublattices. Examples of such ferrimagnets are the spinel-type ferrites  $\text{NiFe}_2\text{O}_4$ ,  $\text{CoFe}_2\text{O}_4$ , and yttrium iron garnet  $\text{Y}_3\text{Fe}_5\text{O}_{12}$ . These ferrites exhibit a normal (‘Weiss’) temperature dependence of spontaneous magnetization  $\sigma_s(T)$ . Their magnetization curves  $\sigma(H)$  and hysteresis properties are similar to those of simple ferromagnets.

In the second type are ferrimagnets in which one of the intrasublattice exchange interactions is comparable with the intersublattice interaction. In this case the competition of these interactions brings into existence a noncollinear (canted) magnetic structure. The first theoretical consideration of such ferrimagnets dates back to Yafet and Kittel [7]. The magnetization curves  $\sigma(H)$  of these materials have two special features — the ‘lack’ of magnetic saturation and magnetization jumps in high magnetic fields (resulting from the ‘disturbance’ of the noncollinear magnetic structure by the external magnetic field  $H$ ).

It is the author’s opinion that ferrimagnets possessing a ‘weak’ magnetic sublattice should be ascribed to the third type. A typical example of this type of ferrimagnets is gadolinium iron garnet,  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ . In this material, the  $c$  sublattice occupied by  $\text{Gd}^{3+}$  cations plays the role of a ‘weak’ sublattice, whereas the ‘net’ (or ‘combined’)  $ad$  sublattice occupied by  $\text{Fe}^{3+}$  cations acts as a normal one (it may be referred to as a ‘strong’ sublattice).

The former exhibits an anomalous (asymptotic) temperature dependence of spontaneous magnetization  $\sigma_s$  (the dotted curve 3 in Fig. 1a) similar to the temperature dependence of magnetization for paramagnets, whereas the latter shows a normal (‘Weiss’) temperature dependence  $\sigma_s$  (curve 1 in Fig. 1a). According to the earlier investigations [4–6], magnetic ordering in the ‘weak’ sublattice is set up by an effective negative exchange magnetic field  $(H_{\text{ex}})_{\text{eff}}$  which is induced by the  $ad$  sublattice (that is, by  $\text{Fe}^{3+}$  cations) and acts on  $\text{Gd}^{3+}$  cations. The ‘intrinsic’ exchange interaction between  $\text{Gd}^{3+}$  cations in the  $c$  sublattice is very weak. In such ferrimagnets there ‘easily’ arise compensation points  $\Theta_c$ . The distinguishing features of these ferrimagnets are the low-temperature paraprocess and some other phenomena. (In Fig. 1a,  $T_B$  is the point of the low-temperature order–disorder magnetic phase transition in the ‘weak’ sublattice. Such a transition was considered in papers [4–6].)

Among ferrimagnets of this type, apart from garnet-type ferrites with heavy rare-earth elements, are some rare-earth metal–iron (cobalt) intermetallics [18, 19], and lithium



**Figure 1.** Magnetization curves of different sublattices of gadolinium iron garnet ( $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ): (a) 1, magnetization curve  $\sigma_s(T)$  for the ‘combined’ sublattice (this curve is assumed to be approximated by the magnetization curve  $\sigma_s(T)$  of yttrium iron garnet,  $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ); 2, experimental magnetization curve for  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ; 3, magnetization curve for the ‘weak’ (gadolinium) sublattice; (b) how the compensation point comes into existence (according to Néel [1]).

chromites-ferrites of the system  $\text{Li}_{0.3}\text{Fe}_{2.5-x}\text{Cr}_x\text{O}_4$  with  $x = 1; 1.25$  [20]. In the last-mentioned ferrimagnets, the ‘weak’ sublattice (with asymptotic temperature dependence of  $\sigma_s$ ) is the octahedral sublattice, since a large number of  $\text{Fe}^{3+}$  ( $5d^5$ ) cations are substituted in it by  $\text{Li}^{1+}$  and  $\text{Cr}^{3+}$  cations (a  $\text{Cr}^{3+}$  cation has a low magnetic moment because its electronic configuration is  $3d^3$ ).

Other substituted spinel-type ferrites were also synthesized with compensation points  $\Theta_c$  (when  $\text{Ni}^{2+}$  and  $\text{V}^{4+}$  cations were substituted for  $\text{Fe}^{3+}$  in octahedra). Note that Néel’s theory [1] supposes that the  $\Theta_c$  points arise if both sublattices exhibit the ‘Weiss’ temperature dependence of  $\sigma_s(T)$  (Fig. 1b). In this case, however, the appearance of these points is highly improbable. In such ferrimagnets, the  $\Theta_c$  point up to the present has not been found.

## 3. Effects of unidirectional exchange anisotropy in ‘weak-sublattice’ ferrites

An important distinction of the third type of ferrimagnet from the first two is the existence of unidirectional (‘one-sided’) exchange anisotropy which arises because of the action of an effective exchange field  $(H_{\text{ex}})_{\text{eff}}$  (negative in sign) induced by the ‘strong’ sublattice on the ‘weak’ sublattice (the reverse action is negligible). As a result, the third type of ferrimagnet must exhibit anomalous effects which are characteristic of magnetic systems possessing such anisotropy (no such effects are found in the first and second type ferrimagnets).

Maiklejohn and Bean [8] were the first to observe this type of anisotropy in a somewhat exotic object — Co metal covered by a layer of cobalt oxide  $\text{CoO}$ , that is, in a ferromagnet–antiferromagnet system. On cooling this system below the Curie point of  $\text{CoO}$ , the Co ferromagnet, producing a high effective exchange field  $(H_{\text{ex}})_{\text{eff}}$ , oriented the nearby spins of the  $\text{CoO}$  antiferromagnet along the magnetization direction of Co spins, as if producing an additional

residual magnetization in Co. If an external magnetic field  $H$  was applied to the system in opposition to this magnetization, a partial ‘disturbance’ (a slight decrease) of the additional residual magnetization took place. This resulted in a displacement of the hysteresis loop.

The effect of displacement of the hysteresis loop was subsequently observed in NiMn alloys in the neighborhood of the Ni<sub>3</sub>Mn composition, as well as in other alloys (Ni–Fe, in particular) [9, 11, 12], since in these materials there exist small regions (clusters) possessing ferro- and antiferromagnetic properties. Vlasov and Mitsek [10] elaborated a thermodynamic theory of unidirectional anisotropy occurring in the ferromagnet–antiferromagnet systems. As was pointed out in [12], this type of anisotropy may come into being in some types of ferrimagnets.

Perekalina with co-workers [22] observed the effect of unidirectional exchange anisotropy in hexaferrites, where antiferromagnetic hexagonal blocks alternate with ferrimagnetic spinel blocks. The latter blocks act on the antiferromagnetic hexagonal blocks (with rather weak exchange interaction) by their stronger effective exchange field. The authors [22] measured torque moments  $L$  rotating single-crystal hexaferrite disks through some angle  $\alpha$  in a magnetic field. In hexaferrites with a ‘weak’ sublattice (whose role was performed by hexagonal blocks), there arose unidirectional exchange anisotropy, with the result that the measured values of torque moments  $L$  were proportional to  $\sin \alpha$  (this corresponds to the unidirectional exchange anisotropy energy  $E_{\text{ex}} \sim \cos \alpha$ ). In other hexaferrite specimens, for which exchange interactions in hexagonal and spinel blocks were comparable in magnitude (that is, in specimens without a ‘weak’ sublattice), the measured values of  $L$  were proportional to  $\sin 2\alpha$  (this corresponds to the uniaxial magneto-crystalline anisotropy energy  $E_m \sim \cos^2 \alpha$ ).

We now turn our attention to the consideration of the effects of unidirectional exchange anisotropy in the third type ferrimagnets (ferrites). The description of magnetostriction and other magnetoelastic effects is based on the expansion of the thermodynamic potential  $\Phi$  in a series in odd powers of the magnetization  $I$ , taking into account the elastic and magnetoelastic terms [28]

$$\Phi(I, P, T) = \Phi_0 + aI + bI^3 + cP + gP^2 + eIP - IH, \quad (1)$$

where  $P$  is stress. In this expansion, the next to last term is the magnetoelastic energy

$$E_{\text{me}} = eIP, \quad (2)$$

where  $e$  is the magnetostriction constant.

Differentiating  $E_{\text{me}}$  with respect to  $P$ , we obtain the magnetostriction

$$\lambda_p = \frac{dE_{\text{me}}}{dP} = eI, \quad (3)$$

which is linear in magnetization because of the influence of unidirectional exchange anisotropy. (Note that for the first and second type ferrimagnets, the next to last term in expansion (1) has the form  $eI^2P$ , so in these ferrimagnets,  $\lambda_p$  is quadratic in  $I$ .) Thus, in the third type ferrimagnets a violation of the ‘evenness’ of  $\lambda_p$  takes place.

A thermodynamically inverse effect corresponds to linear magnetostriction — piezomagnetism, that is, an occurrence of magnetization  $\Delta I$  without any magnetic field  $P$  under the

action of stress  $H$ . Differentiating  $E_{\text{me}}$  with respect to  $H$  (or  $I$ ), we obtain a relationship for the piezomagnetic effect

$$\Delta I = eP. \quad (4)$$

An important point is that, in the third type ferrimagnets, the  $\lambda_p$  and  $\Delta I$  effects come into existence owing to the paraprocess, since the energy  $E_{\text{me}}$  is of exchange nature.

In the third type ferrimagnets, the violation of the ‘evenness’ of the magnetocaloric effect and magnetoresistance also takes place.

When considering the effects of unidirectional exchange anisotropy in these ferrimagnets, it must be taken into account that the field  $(H_{\text{ex}})_{\text{eff}}$  acting on the ‘weak’ sublattice is

$$(H_{\text{ex}})_{\text{eff}} = J_{12}I_1, \quad (5)$$

where  $J_{12}$  is the exchange interaction constant (the indices 1 and 2 stand for the ‘strong’ and ‘weak’ sublattices, respectively), and  $I_1$  is the magnetization of the ‘strong’ sublattice. Since  $I_1$  is temperature-dependent, the influence of unidirectional exchange anisotropy will be especially strong at low temperatures, but, as is shown below, it also exists at the Curie point and even above this point.

The violation of the ‘evenness’ for magnetostriction  $\lambda_p$  in the paraprocess was observed in garnet-type ferrites with heavy rare-earth metals. Figure 2 shows the dependences of  $\lambda_p$  on the specific magnetization  $\sigma$  of Ho<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> in holmium iron garnet, at different temperatures (from data reported in the paper [23]). It is seen that at all temperatures  $\lambda_p$  is linear in  $\sigma$ .

The violation of the ‘evenness’ of magnetoresistance and the magnetocaloric effect due to the paraprocesses manifests itself in the neighborhood of the compensation point. As is seen from Fig. 3 and 4, where the experimental data on the magnetoresistance for lithium ferrite–chromite [Li<sub>2</sub>O(F<sub>2</sub>O<sub>3</sub>)<sub>2.5</sub>(Cr<sub>2</sub>O<sub>3</sub>)<sub>2.5</sub>] [24] and on the magnetocaloric effect  $\Delta T$  for gadolinium iron garnet Gd<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> [25] are presented,  $\Delta\rho/\rho$  and  $\Delta T$  change their signs when passing through the point  $\Theta_c$ , that is, when reversing the direction of the magnetization vector for the ‘weak’ sublattice. This is possible if  $\Delta\rho/\rho$  and  $\Delta T$  are linear in  $\sigma$ .

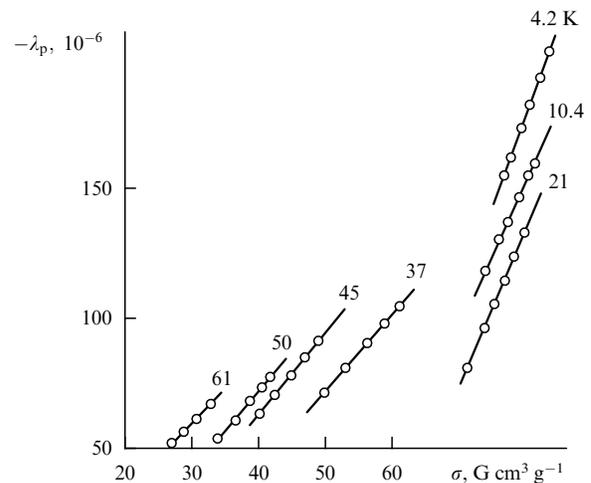
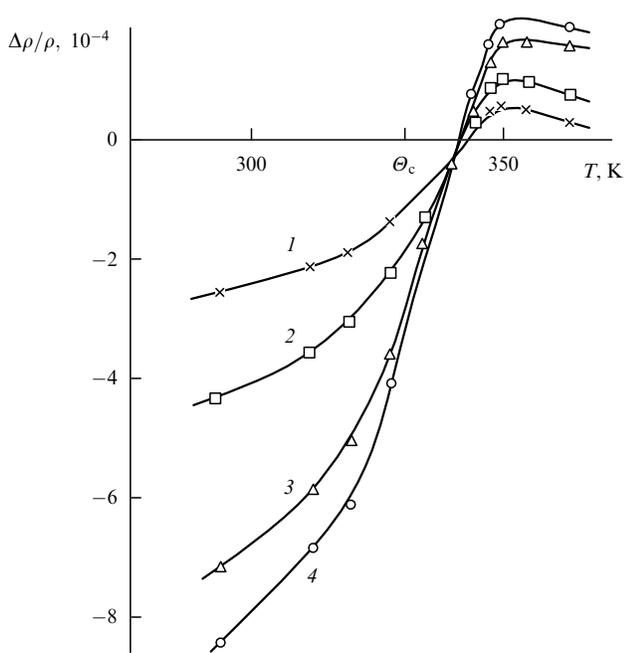
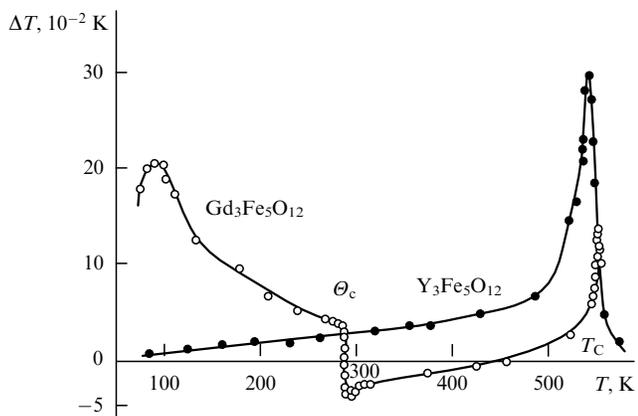


Figure 2. Magnetostriction  $\lambda_p$  in holmium iron garnet (Ho<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>) at different temperatures as a function of the specific magnetization  $\sigma$ .



**Figure 3.** Temperature dependence of magnetoresistance near the compensation point  $\Theta_c$  in the ferrite  $\text{Li}_2\text{O}(\text{Fe}_2\text{O}_3)_{2.5}(\text{Cr}_2\text{O}_3)_{2.5}$  at  $H = 1.8$  (1); 2,26 (2); 6,8 (3); 11 Oe (4).



**Figure 4.** Temperature dependence of the magnetocaloric effect  $\Delta T$  in the garnet-type ferrites  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$  and  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  at a magnetic field  $H = 16$  Oe.

It is known that in the paraprocess region the magnetoresistive effect  $\Delta\rho/\rho$  is normally negative and the magnetocaloric effect  $\Delta T$  is positive. This corresponds to a decrease of the magnetic entropy in the external magnetic field because of the alignment of all magnetic moments along  $H$  that takes place in the normal ferromagnetic paraprocess. The fact that in passing through  $\Theta_c$  both effects change their signs points to an increase of the magnetic entropy under the action of the external magnetic field  $H$ , that is, there arises an antiferromagnetic paraprocess (the field  $H$  rotates magnetic spins through  $180^\circ$  overcoming the action of unidirectional exchange anisotropy).

We now turn our attention to the following experimental fact. As is seen from Fig. 4, there is a sharp change in  $\Delta T$  when passing through the point  $\Theta_c$ . It is obvious that the value of  $\Delta T$  may be taken as a measure of the unidirectional exchange anisotropy energy at the compensation point  $\Theta_c$ .

From the aforesaid it follows that the occurrence of the antiferromagnetic paraprocess in the neighborhood of the point  $\Theta_c$  is also an effect of unidirectional exchange anisotropy that manifests itself only in ‘weak-sublattice’ ferrimagnets.

Thus, in ‘weak-sublattice’ ferrimagnets the unidirectional exchange anisotropy leads to a greater number of anomalous effects than was observed at one time for the system Co–CoO and other systems similar to it.

Of great interest is the possibility of measuring the piezomagnetic effect in ‘weak-sublattice’ ferrimagnets. This effect was first measured by Borovik-Romanov [26] in the antiferromagnetic crystals  $\text{MnF}_2$  and  $\text{CoF}_2$  possessing a definite type of crystalline symmetry which promotes its occurrence [27]. According to Refs [26, 27], the occurrence of a piezomagnetic effect is associated with a rearrangement of magnetic domains under the action of stress  $P$ . The effect was found to be very weak in these materials. The piezomagnetic effect was also measured in some noncollinear antiferromagnets (such as  $\alpha\text{-Fe}_2\text{O}_3$ ). In these materials, it is very weak as well.

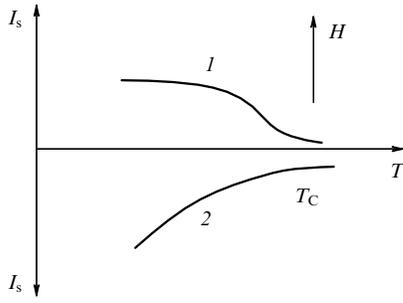
In the case of ‘weak-sublattice’ ferrites, the piezomagnetic effect must arise as a result of the orientations of spin magnetic moments, that is, of the paraprocess induced by stress  $P$ . The linear magnetostriction in the paraprocess peaks in the neighborhood of the low-temperature phase transition (at the point  $T_B$ ); therefore, the piezomagnetic effect has also to peak in this region. No measurements of this effect in ferrites were taken so far.

#### 4. Manifestations of unidirectional exchange anisotropy at the Curie points of ferrites

Most ferrimagnets (ferrites among them) do not possess a ‘weak’ sublattice at low temperatures. In such ferrimagnets the intersublattice interaction is of first importance, but there also exist intrasublattice (differing in magnitude) interactions. The latter interactions become more appreciable in the neighborhood of the Curie point ( $T_C$ ), where the intrasublattice interaction energy sharply decreases. As the temperature  $T_C$  is approached, the magnetic ordering in two different sublattices decreases at different rates. In other words, the temperature dependence of spontaneous magnetization for one of these sublattices will be approximated by a ‘Weiss’ curve, whereas for the other it will be approximated by an asymptotic curve (as is shown schematically in Fig. 5), that is, the former sublattice will be ‘strong,’ the latter one will be ‘weak.’ This means in turn that in all ferrimagnets near their Curie points there has to manifest itself unidirectional exchange anisotropy. Below is a summary of experimental results in support of the occurrence of such anisotropy near the Curie points of ferrites.

Figure 4 illustrates the temperature dependences of the magnetocaloric effect in gadolinium and yttrium iron garnets. It is seen that the maximum of the  $\Delta T$  effect in the Curie point of the former ferrite is much less than that of the latter, whereas at temperatures away off  $T_C$  this is not the case. Such a situation is explained by the existence of unidirectional exchange anisotropy at the Curie points of ferrites. This anisotropy is induced in a ‘weak’ sublattice by a ‘strong’ one. In the case of gadolinium iron garnet, the  $d$  sublattice is ‘strong,’ whereas the  $c$  and  $a$  sublattices are ‘weak.’

When an external magnetic field  $\Delta H$  is applied, the ferromagnetic paraprocess comes into play in the ‘strong’ ( $d$ )



**Figure 5.** Temperature dependences of the spontaneous magnetization of sublattices in ferrimagnets as the Curie point is approached (schematic): 1, ‘strong’ sublattice; and 2, ‘weak’ sublattice.

sublattice of gadolinium iron garnet, and according to the well-known thermodynamic formula, we have

$$\Delta T = -\frac{T}{C} \left( \frac{dI}{dT} \right)_H \Delta H \tag{6}$$

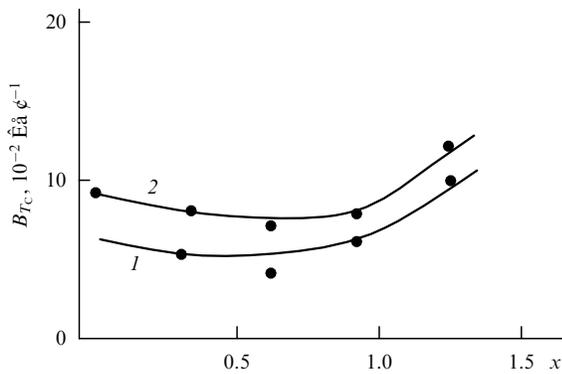
(the  $\Delta T$  effect is positive). However, this magnetic field ( $\Delta H = H$ ) simultaneously induces the antiferromagnetic paraprocesses in the ‘weak’ ( $c$  and  $a$ ) sublattices of this ferrite (such paraprocesses take place only in ‘weak-sublattice’ ferrimagnets), with the resulting formation of a negative  $\Delta T$  component, which decreases the total  $\Delta T$  effect in  $Gd_3Fe_5O_{12}$  as compared to its value in  $Y_3Fe_5O_{12}$ .

Mention should be made of another experimental fact in support of the occurrence of the antiferromagnetic paraprocess (and hence the occurrence of unidirectional exchange anisotropy) near the Curie points of ferrites. It was obtained in studies of the magnetization curve directly at the Curie point for the substituted garnet-type ferrites  $\{R_{3-x}Ca_x\}[Fe_2](Fe_{3-x}Sn_x)O_{12}$  ( $R = Gd, Tb$ ) [31]. According to [28], this magnetization curve is described by the equation

$$I = B_{T_C} H^{1/3} \tag{7}$$

(the critical magnetic isotherm), where  $B_{T_C}$  is a constant that characterizes the intensity of the paraprocess at the Curie point.

Figure 6 shows the composition dependence of  $B_{T_C}$ . Here,  $x$  is the concentration of Sn cations substituting for part of  $Fe^{3+}$  cations in the ‘strong’ sublattice, which results in a decrease in the unidirectional exchange anisotropy. This



**Figure 6.** Composition dependence of the  $B_{T_C}$  constant for garnet-type ferrites with substitutions in the tetrahedral sublattice: 1, Gd system; and 2, Tb system.

causes, in its turn, the antiferromagnetic paraprocess in the  $c$  sublattice, with the result that the  $B_{T_C}$  constant increases slightly at large  $x$ .

### 5. Hyperbolic temperature dependence of inverse susceptibility above the Curie points of ferrites as an effect of unidirectional exchange anisotropy

Using the molecular field approximation, Néel [1] obtained a formula for the temperature dependence of inverse susceptibility directly above the Curie point  $T_C$  of a two-sublattice ferrite (Néel’s law). It has the form

$$\frac{1}{\chi} = \frac{1}{\chi_0} + \frac{T}{C} - \frac{\sigma_0}{T - \Theta}, \tag{8}$$

where  $\chi_0$ ,  $\sigma_0$ , and  $\Theta$  are parameters depending on the molecular field coefficients  $\mathbf{n}$ , being a characteristic of the intersublattice exchange interaction, and  $\alpha$ ,  $\beta$ , being characteristics of the intrasublattice exchange interactions; and  $C$  is the Curie constant.

In the case of a three-sublattice ferrimagnet ( $Gd_3Fe_5O_{12}$ , for example), the inverse magnetic susceptibility directly above the Curie point is described by a more complex formula [32]

$$\frac{1}{\chi} = \frac{1}{\chi_0} + \frac{T}{C} - \frac{\sigma_1^2 T + m_0}{T^2 - \Theta_1 T - P_0}, \tag{9}$$

where  $\chi_0$ ,  $\sigma_1$ ,  $m_0$ ,  $\Theta_1$ , and  $P_0$  depend on the molecular field coefficients.

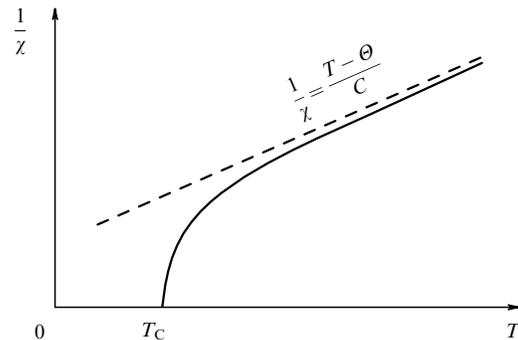
As is known, in ferrimagnets directly above  $T_C$  and antiferromagnets above  $T_N$ , the Curie–Weiss law is valid:

$$\frac{1}{\chi} = \frac{T - \Theta}{C}, \tag{10}$$

where  $C$  and  $\Theta$  are the Curie and Weiss constants, respectively.

Figure 7 schematically shows the temperature dependence of  $1/\chi$  at  $T > T_C$  for a ferrimagnet (solid line) and an antiferromagnet (dashed line). For a ferrimagnet, at temperatures directly above  $T_C$ , the  $1/\chi$  dependence on  $T$  is of hyperbolic character and only at temperatures far above  $T_C$  it adheres to the Curie–Weiss law (that is, approximates a straight line).

Experimental verification of Néel’s law for a two-sublattice ferrite was first given by Fallot and Maroni [34]



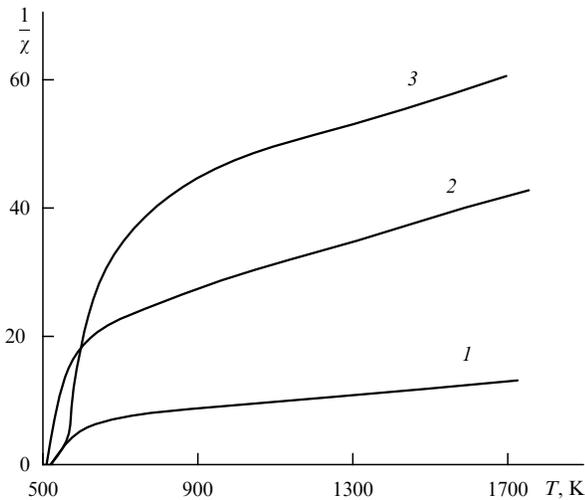
**Figure 7.** Temperature dependence of the inverse magnetic susceptibility  $1/\chi$  for a two-sublattice ferrite at  $T > T_C$  (schematic).

and then in many later papers [30]. However, up to the present, even though 50 years has elapsed since the publication of Néel's paper [1], nowhere has it been clarified what the physical mechanism of 'lowering' inverse magnetic susceptibility is near the Curie point  $T_C$  of a ferrite.

In our opinion, the hyperbolic temperature dependence of inverse susceptibility at  $T > T_C$  is simply the effect of unidirectional exchange anisotropy. At temperatures above  $T_C$ , there exist in ferrites, as before, magnetic sublattices, one of which is a 'strong' sublattice (its magnetization is aligned with the external magnetic field  $H$ ) and the other is a 'weak' one (its magnetization is opposite to  $H$ ). As a result, in addition to the ferromagnetic paraprocess in the 'strong' sublattice, there arises an antiferromagnetic paraprocess in the 'weak' sublattice, which results in raising the susceptibility  $\chi$  (that is, in lowering inverse susceptibility  $1/\chi$ ) at  $T > T_C$ .

As the temperature recedes from  $T_C$  to high temperatures, the effective exchange field  $(H_{\text{ex}})_{\text{eff}}$  of the 'strong' sublattice decreases, resulting in a decrease in the unidirectional exchange anisotropy energy, so  $1/\chi$  approaches the value that appears in the Curie – Weiss law (see Fig. 7). In formulas (8) and (9), the terms with the minus sign correspond to the action of unidirectional exchange anisotropy, that is, lead to a lowering of the inverse magnetic susceptibility.

Figure 8 illustrates experimental data [35] on the temperature dependences of  $1/\chi(T)$  above the Curie points of erbium, ytterbium and yttrium iron garnets. It is seen that in erbium and ytterbium iron garnets, the inverse susceptibility  $1/\chi$  is much less (hence,  $\chi$  is much greater) than that in yttrium iron garnet, since in the first two ferrites the antiferromagnetic paraprocess takes place in two sublattices ( $c$  and  $a$ ), whereas in yttrium iron garnet it takes place in the  $a$  sublattice only.



**Figure 8.** Temperature dependence of  $1/\chi$  for the ferrites  $\text{Er}_3\text{Fe}_5\text{O}_{12}$  (1),  $\text{Yb}_3\text{Fe}_5\text{O}_{12}$  (2), and  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (3) at  $T > T_C$ .

## 6. Peculiarities of the order–disorder magnetic phase transitions in ferrites, induced by unidirectional exchange anisotropy

In the 'weak-sublattice' ferrites, there exist two such transitions: a low-temperature transition (at the point  $T_B$ ) and a transition at the Curie point  $T_C$ .

First, we consider the transition at the  $T_B$  point. According to Refs [4, 5], magnetic ordering in the 'weak' sublattice results from the action of an exchange field  $(H_{\text{ex}})_{\text{eff}}$  induced by the 'strong' sublattice. Calculations in the molecular field approximation yield the following estimate for  $T_B$ :

$$T_B \approx \frac{\mu_B g_s S (H_{\text{ex}})_{\text{eff}}}{k}, \quad (11)$$

where  $S$  is the total spin of the 'weak-sublattice' cations. In the case of  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ , this formula gives  $T_B \sim 65$  K. The experimental value of  $T_B$  is about 100 K.

Compared to the phase transition at the Curie point  $T_C$ , the magnetic phase transition at the point  $T_B$  exhibits the following special features. First, it is as if an incomplete transition, since thermal motion only partially destroys magnetic ordering in the 'weak' sublattice at the  $T_B$  point (at  $T > T_B$ , the field continues to act on this sublattice).

Second, the transition at the  $T_B$  point exhibits no fluctuations (in contrast to the transition in a ferromagnet at  $T_C$ ), since magnetic fluctuations are suppressed by the action of the field  $(H_{\text{ex}})_{\text{eff}}$  of the 'strong' sublattice or, what is the same, by the action of unidirectional exchange anisotropy.

Consider now the magnetic phase transition at the Curie point of ferrites. The 'strong' sublattice plays a leading part in the mechanism of this transition, since the magnetization of this sublattice is greater than that of the 'weak' sublattice. In the neighborhood of  $T_C$ , it exhibits intense magnetic fluctuations, much as this takes place at the Curie points of ferromagnets. As for the 'weak' sublattice, it is free of such magnetic fluctuations because of the action of unidirectional exchange anisotropy. This leads to the fact that magnetic fluctuations (both 'temporal' and 'spatial') arise as if not over all the volume of the ferrite specimen. As a consequence, these fluctuations are less intense than those at the Curie points of ferromagnets.

Another distinction between the order–disorder magnetic phase transitions at the Curie points of ferrites (and other ferrimagnets) and ferromagnets lies in the fact that in the former magnets this transition is more spread in temperature than in the latter ones. In ferromagnets this 'spreading' is determined by structural inhomogeneities and the origination of a short-range magnetic order, whereas in ferrites the influence of unidirectional exchange anisotropy, which inhibits the disturbance of the magnetic order near  $T_C$ , must be added to these causes of 'spreading.' Owing to this anisotropy, a magnetic order exists above the Curie point in the temperature region immediately adjacent to  $T_C$ , as indicated by very low values of the inverse magnetic susceptibility directly above  $T_C$  (Néel's law).

All the aforesaid regarding the peculiarities of the magnetic phase transition at the Curie point should be taken into consideration in studies of the magnetic critical state (scaling [33, 36, 37]). The matter is that for solving this problem it is important to determine from experiment the true values of the critical exponents  $\beta$ ,  $\gamma$  and  $\delta$  appearing in the relationships for the temperature dependences of spontaneous magnetization  $I_s$  near  $T_C$ , the susceptibility  $\chi$  above  $T_C$ , and the critical isotherm (the magnetization curve directly at  $T_C$ ):

$$I_s = A(T - T_C)^\beta, \quad (12)$$

$$\chi = C(T - T_C)^{-\gamma}, \quad (13)$$

$$I = BH^{1/\delta}. \quad (14)$$

In work [38] these critical exponents were determined for ferromagnets (predominantly for nickel). However, in a number of other works they were measured for spinel-type ferrites [39, 40] and garnet-type ferrites [37], reasoning that for these materials the critical exponents are identical to those for simple ferromagnets.

As follows from the consideration of the peculiarities of the phase transition at the  $T_C$  point of ferrites, this is obviously not the case. In particular, equation (13) does not apply to ferrites, since Néel's law holds at  $T_C$ .

## 7. Conclusion

In conclusion it should be said that this paper is devoted to the analysis of experimental data for a variety of anomalous effects in the neighborhood of the compensation points and the Curie points of ferrites. It was inferred that unidirectional exchange anisotropy manifests itself in these temperature regions. The concept of this anisotropy makes it possible to explain many phenomena which up to now have not found an adequate interpretation. Among these are

(1) the violation of 'evenness' for the magnetostrictive, magnetoresistive, and magnetocaloric effects in 'weak-sublattice' ferrimagnets;

(2) the antiferromagnetic paraproces, arising at the compensation point and at the Curie point, is an effect of unidirectional exchange anisotropy;

(3) hyperbolic temperature dependence of inverse susceptibility directly above the Curie points of ferrites (Néel's law) results from the action of unidirectional exchange anisotropy (antiferromagnetic paraproces);

(4) unidirectional exchange anisotropy leads to a partial suppression of magnetic fluctuations and 'spreading' of the magnetic transition at the Curie point;

(5) based on the analysis of manifestations of the above-mentioned phenomena, the possibility of occurrence of a piezomagnetic effect in the 'weak' sublattice of ferrite in the neighborhood of the low-temperature transition ( $T_B$  point) is predicted.

From the above discussion it follows that the concept of unidirectional exchange anisotropy makes it possible to find a unified explanation of many phenomena that manifest themselves in ferrites. In ferrites, this anisotropy determines considerably more effects than have been found before in ferromagnet-antiferromagnet systems.

## References

- Néel L *Ann. Phys. (Paris)* **3** 137 (1948) [Translated into Russian; in *Antiferromagnetizm* (Ed. S V Vonsovskii) (Moscow: IL, 1956)]
- Weiss P J *Phys.* **3** 5 433 (1896)
- Snoek J L *New Developments in Ferromagnetic Materials* (New York: Elsevier, 1947) [Translated into Russian (Moscow: Nauka, 1949)]
- Belov K P *Zh. Eksp. Teor. Fiz.* **41** 692 (1961) [*Sov. Phys. JETP* **14** 499 (1965)]; Belov K P *Ferrity v Sil'nykh Magnitnykh Polyakh* (Ferrites in Strong Magnetic Fields) (Moscow: Nauka, 1972)
- Belov K P, Nikitin S A *Izv. Akad. Nauk SSSR. Ser. Fiz.* **24** 95 (1970); Belov K P, Nikitin S A *Phys. Status Solidi* **12** 453 (1965)
- Belov K P *Usp. Fiz. Nauk* **166** 669 (1996) [*Phys. Usp.* **39** 623 (1996)]
- Yafet Y, Kittel C *Phys. Rev.* **87** 290 (1952)
- Maiklejohn W H, Bean C P *Phys. Rev.* **102** 1413 (1956)
- Kouvel J S, Graham C D J *Appl. Phys.* **30** 312 (1959)
- Vlasov K B, Mitsek A I *Fiz. Met. Metalloved.* **14** 487, 998 (1962)
- Volkenshtein N V, Turchinskaya M I *Izv. Akad. Nauk SSSR Ser. Fiz.* **27** 12 (1963)
- Vlasov K B et al. *Izv. Akad. Nauk SSSR Ser. Fiz.* **28** 423 (1964)
- Perekalina T M, Shurova A D, Fonton S S *Zh. Eksp. Teor. Fiz.* **57** 749 (1969) [*Sov. Phys. JETP* **30** 383 (1969)]
- Jacobs I J *Chem. Sol.* **11** 1 (1959)
- Bertaut F, Forrat F C. R. *Acad. Sci.* **242** 382 (1956)
- Geller S, Gillo M A *Acta Crystallogr.* **10** 239 (1957)
- Pauthenet R C. R. *Acad. Sci.* **242** 1859 (1956)
- Buschow K *Phys. Status Solidi* **7** 199 (1971)
- Belov K P et al. *Fiz. Met. Metalloved.* **34** 470 (1976)
- Gorter E W *Philips Res. Rep.* **9** 125, 321, 453 (1954)
- Blasse G *Crystal Chemistry and Some Magnetic Properties of Mixed Metal Oxides with Spinel Structure* (Eindhoven: Philips Research Laboratories, 1964) [Translated into Russian (Moscow: Metallurgiya, 1968)]
- Perekalina T M, Shurova A D, Fonton S S *Zh. Eksp. Teor. Fiz.* **57** 749 (1969) [*Sov. Phys. JETP* **30** 383 (1969)]
- Belov K P, Sokolov V I *Izv. Akad. Nauk SSSR Ser. Fiz.* **30** 1073 (1966)
- Belov K P et al. *Zh. Eksp. Teor. Fiz.* **38** 1914 (1960) [*Sov. Phys. JETP* **11** 1378 (1960)]
- Belov K P et al. *Pis'ma v Zh. Eksp. Teor. Fiz.* **7** 423 (1968) [*JETP Lett.* **7** 393 1 (1968)]
- Borovik-Romanov A S *Zh. Eksp. Teor. Fiz.* **38** 1088 (1960) [*Sov. Phys. JETP* **11** 1346 (1960)]
- Dzyaloshinskii I E *Zh. Eksp. Teor. Fiz.* **33** 807 (1957) [*Sov. Phys. JETP* **6** 642 (1958)]
- Belov K P *Magnitnye Prevrashcheniya* (Magnetic Transitions) (Moscow: Fizmatgiz, 1959) [Translated into English (New York: Consultants Bureau, 1961)]
- Belov K P et al. *Zh. Eksp. Teor. Fiz.* **62** 2183 (1972) [*Sov. Phys. JETP* **35** 1142 (1972)]
- Pauthenet R, Bochirol L J *Phys. Rad.* **12** 249 (1951)
- Belov K P, Shlyakhina L P *Fiz. Met. Metalloved.* **30** (1) 29 (1970)
- Néel L C. R. *Acad. Sci.* **234** 2172 (1952)
- Patashinskii A Z, Pokrovskii V L *Fluktyatsionnaya Teoriya Fazovykh Perekhodov* (Fluctuation Theory of Phase Transitions) 2nd ed. (Moscow: Nauka, 1982) [Translated into English (Oxford: Pergamon Press, 1979)]
- Fallot M, Maroni P J *Phys. Rad.* **12** 256 (1951)
- Aleonard R J *Phys. Chem. Sol.* **15** 167 (1960)
- Fisher M *The Nature of Critical Points* (Boulder, Colo.: Univ. of Colorado Press, 1965) [Translated into Russian (Moscow: Mir, 1968)]
- Kamilov I K, Aliev X K *Statisticheskie Kriticheskie Yavleniya v Magnitouporyadochennykh Kristallakh* (Statistical Critical Phenomena in Magnetically Ordered Crystals) (Makhachkala: Izd. DNTs RAN, 1993)
- Kouvel J S, Rodbell D D J *Appl. Phys.* **38** 979 (1967)
- Miyatani K, Yoshikawa K J *Appl. Phys.* **41** 1272 (1970)
- Onbayachi S, Iida S J *Phys. Soc. Jpn.* **25** 1187 (1968)