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Pyromagnetic effect in ferrimagnets with a 'weak' sublattice

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Contents

Abstract. The temperature dependence of the residual and spontaneous magnetization in ferrites with a 'weak' sublattice may be taken as evidence for the pyromagnetic effect — a magnetic analog of the pyroelectric effect — in which the magnetization of a sample increases on cooling in the absence of an external magnetic field. A confirmation of this has been provided by the observation of a thermodynamically inverse phenomenon, the linear magnetocaloric effect, in such ferrites. These effects are due to the unidirectional exchange anisotropy characteristic of ferrimagnets with a weak sublattice.

1. Introduction

Following the extensive early work on ferroelectrics in the 1930s — when they were believed to be analogs of ferromagnets, hence the name — it was found that some of the phenomena they exhibit, in particular piezoelectricity and pyroelectricity, do not occur in magnetically ordered materials, i.e., ferro-, ferri-, and antiferromagnets. This situation, however, changed in the late 1950s when Borovik-Romanov discovered the piezomagnetic effect in antiferromagnetic crystals with certain symmetry properties [1].

The present author has recently shown [2], based on the magnetic data on ferrites with a 'weak' sublattice, that such materials should display the piezomagnetic effect due to the unidirectional exchange anisotropy (note that no direct measurements of this effect have yet been attempted).

In this paper, it will be shown that the same anisotropy causes ferrimagnets of this type to exhibit the pyromagnetic effect (the analog of the pyroelectric effect). Because of the thermodynamic relationship between the piezomagnetic and pyromagnetic effects (see Section 3), a few words on Borovik-Romanov's discovery are in order here.

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2. On the 40th anniversary of A S Borovik-Romanov's discovery of the piezomagnetic effect and the inverse phenomenon of linear magnetostriction

In the late 1950s, Borovik-Romanov [1] succeeded in measuring the piezomagnetic effect on single crystals of MnF_2 and CoF_2, antiferromagnets that had been shown to have unusual magnetic symmetry properties by Dzyaloshinskiĭ [3]. The experiment was a very difficult and intricate one, the biggest challenge being that an applied elastic compression P caused only a very slight magnetization change ΔI in the material.

The piezomagnetism discovered by Borovik-Romanov is closely related to the 'weak' ferromagnetism which magnetoanisotropic forces are known to produce in antiferromagnets. Like the magnetic moment of 'weak' ferromagnetism, that of piezomagnetism is oriented perpendicular to the vectors of spontaneous magnetization in the antiferromagnet sublattices, thus making them noncollinear to each other. And like the moment of 'weak' ferromagnetism, the piezomagnetic moment is perpendicular to the magnetization vectors of the sublattices.

One further detail brought out by Borovik-Romanov is the strong dependence of piezomagnetism on the domain structure of the antiferromagnet. Since piezomagnetism can be significantly reduced in multidomain antiferromagnets, single-domain samples are employed to observe the effect uncomplicated. The 'weak' ferromagnets, e.g., hematite (α-Fe_2O_3) and others, are also piezomagnetic.

An effect thermodynamically inverse to piezomagnetism, the so-called 'odd' (linear) magnetostriction, was also observed in Borovik-Romanov's experiments on these materials.

After the theoretical works of Dzyaloshinskiĭ [3, 4] and experimental works by Borovik-Romanov [1] the existence of linear magnetostriction in antiferromagnets was confirmed by Birss and Anderson [5] and experimentally studied in much detail in antiferromagnets with 'weak' ferromagnetism (hematite) [6] and in rare-earth orthoferrites [7].

3. Thermodynamic relations between paraprocess effects in ferrimagnets with a 'weak' sublattice

Ferrimagnets with a 'weak' sublattice possess anomalous properties compared with those having the 'Néel' magnetic structure (with a strong exchange interaction between sublattices [8]). Specifically, they show anomalous paraprocess effects, due to the unidirectional exchange anisotropy [2].

In this section, we will show that ferrimagnets with a weak sublattice should exhibit the piezomagnetic effect and its inverse phenomenon, linear magnetostriction. These, however, are due to paraprocess effects in the weak sublattice (i.e., originate in exchange) and therefore are of a different nature from their counterparts in antiferromagnets. In fact, they are a consequence of the thermodynamic relation between the paraprocess effects in a ferromagnet's weak sublattice, as also are the pyromagnetic effect (the analog of the pyroelectric effect) in ferroelectric and dielectric materials [12], and its thermodynamic inverse, the linear magnetocaloric effect.

The thermodynamic relation between paraprocess effects will be considered using the example of gadolinium iron garnet $Gd_3Fe_5O_{12}$, which is a typical weak-sublattice ferrimagnet. The weak sublattice in it is the *c* sublattice of gadolinium, whereas the strong sublattice is represented by the *ad* sublattice of Fe^{3+} cations. We limit ourselves to the temperature range between $T_B \sim 100$ K (the magnetic order – disorder transition in the gadolinium sublattice) to the magnetic compensation point $\Theta_{com} = 293$ K, a range in which the *c* sublattice is in the magnetically isotropic, single-domain, nonhysteretic state.

When subjected to a magnetic field H, elastic stress P, and temperature T, this sublattice exhibits various paraprocess effects as a result of spin ordering changes.

Consider first a hypothetical situation in which the weak sublattice is in a sense isolated from the strong one, i.e., the former is not affected by the exchange field produced by the latter. Given our choice of independent variables (H, P, T), the thermodynamic potential in this case is the Gibbs energy, whose total differential has the form

$$\mathrm{d}G = -I\,\mathrm{d}H - \lambda\,\mathrm{d}P - S\,\mathrm{d}T\,, \tag{1}$$

where I is the spontaneous magnetization, λ is the paraprocess magnetostriction (sometimes also called the exchange magnetostriction), and S is the magnetic part of the entropy (due to changes in spin order). The sign of $\lambda\,\mathrm{d}P$ depends on exactly what type of elastic mechanical stress, compression or tension, is applied. Accordingly, the sign of the term $S\,\mathrm{d}P$ is determined by how the thermal conditions are changed in the experiment, i.e., whether the sample is heated or cooled.

Differentiating relation (1) with respect to H, P and T, we are led to the following magnetic, magnetoelastic, and magnetothermal equations of state [12]:

$$I = \left(\frac{\partial G}{\partial H}\right)_{P,T}; \quad \lambda = \left(\frac{\partial G}{\partial P}\right)_{H,T}; \quad S = \left(\frac{\partial G}{\partial T}\right)_{P,H}. \tag{2}$$

Expanding these in series form about a certain initial state of the *c* sublattice in the temperature range between T_B and Θ_{com} and keeping only the linear terms, we obtain

$$\Delta I = \left(\frac{\partial I}{\partial H}\right)_{P,T}\Delta H + \left(\frac{\partial I}{\partial P}\right)_{T,H}\Delta P + \left(\frac{\partial I}{\partial T}\right)_{P,H}\Delta T\,, \tag{3}$$

$$\Delta\lambda = \left(\frac{\partial \lambda}{\partial H}\right)_{T,P}\Delta H + \left(\frac{\partial \lambda}{\partial P}\right)_{T,H}\Delta P + \left(\frac{\partial \lambda}{\partial T}\right)_{H,P}\Delta T\,, \tag{4}$$

$$\Delta S = \left(\frac{\partial S}{\partial H}\right)_{P,T}\Delta H + \left(\frac{\partial S}{\partial P}\right)_{H,T}\Delta P + \left(\frac{\partial S}{\partial T}\right)_{H,P}\Delta T\,. \tag{5}$$

Equations (3) – (5) describe the entire gamut of magnetic, magnetoelastic, and thermal effects caused by varying H, P, and T in an 'isolated' weak sublattice. The second term in Eqn (3) accounts for the magnetoelastic paraprocess effect due to the applied mechanical stress P (in Ref. [14] it is called a 'mechanoparaprocess', a change in magnetic order under stress P in the presence of an external field H):

$$(\Delta I_P)_{H\neq 0} = \gamma_P \Delta P\,, \tag{6}$$

where $\gamma_P = (\partial I/\partial P)_{T,H}$ is the magnetoelastic coupling coefficient. From the thermodynamics of magnetic phenomena [4] it follows that

$$\left(\frac{\partial I}{\partial P}\right)_{T,H} = \left(\frac{\partial \lambda}{\partial H}\right)_{P,T}, \tag{7}$$

i.e., there is a thermodynamically inverse phenomenon corresponding to the magnetoelastic effect — paraprocess magnetostriction [(Eqn (7) is obtained by differentiating the equalities $I = (\partial G/\partial H)_{P,T}$ and $\lambda = (\partial G/\partial P)_{H,T}$ with respect to P and H, respectively].

The third term in Eqn (3) is

$$(\Delta I_T)_{H\neq 0} = \gamma_T \Delta T\,, \tag{8}$$

where $\gamma_T = (\partial I/\partial T)_{P,H}$ is the magnetothermal coefficient characterizing the slope of the temperature dependence curve $I(T)$ in the presence of a magnetic field H. If the sample undergoes cooling in this process, then I increases due to the enhanced spin order — the reason why this type of paraprocess may be called a thermoparaprocess. The thermodynamic inverse of this phenomenon is the magnetocaloric effect [the first term in Eqn (5)]

$$(\Delta S_T)_{H\neq 0} = \gamma_S \Delta H\,, \tag{9}$$

where $\gamma_S = (\partial S/\partial T)_{P,T}$ is the magnetocaloric coefficient. Relation (9) is the expression for the magnetocaloric effect,

$$\Delta T = -\frac{T}{C_I}\left(\frac{\partial I}{\partial T}\right)_{P,H}\Delta H\,. \tag{10}$$

This expression is obtained from Eqn (9) by substituting $S = \mathrm{d}Q/T$ and $\mathrm{d}Q = C_I T$ (where $\mathrm{d}Q$ is the heat release change in the thermoparaprocess, and C_I is the heat capacity). Equation (10) can be rewritten as

$$\left(\frac{\Delta T}{\Delta H}\right)_{P,T} = -\frac{T}{C_I}\left(\frac{\Delta I}{\Delta T}\right)_{H\neq 0}, \tag{11}$$

implying that there is an inverse phenomenon to the magnetocaloric effect — the magnetothermal effect, due to the thermoparaprocess in the presence of a field. As is well known, in ferromagnets (in our case, in the 'isolated' weak sublattice) the magnetocaloric effect, as well as magnetostriction, are even effects, i.e., they are quadratic in I [14].

To proceed with the analysis of paraprocess effects in a weak sublattice, consider a real situation in which the ferrite possesses unidirectional exchange anisotropy, i.e., an exchange field produced by the *ad* sublattice, of the form

$$(H_{\rm ex})_{\rm eff} = J_{\rm c-ad} I_1 \,, \tag{12}$$

where the parameter $J_{\rm c-ad}$ describes the exchange interaction between the weak and strong sublattices, and I_1 is the magnetization of the strong sublattice. This leads to linear magnetostriction, as was shown in Refs [2, 10] and confirmed in magnetostriction measurements on holmium iron garnet [9]. Measurement of the inverse phenomenon of piezomagnetism has not yet been undertaken.

We next formulate what is in fact the central point of the present paper: a unidirectional exchange anisotropy in the weak sublattice of a given ferrite leads, according to Eqn (9), to an odd, i.e., linear in I, magnetocaloric effect $(\Delta T/\Delta H)_{T,P}$, and to a magnetothermal effect $(\Delta I/\Delta T)_{P,H=0}$, which arises when the sample cools down as a result of the thermoparaprocess in the absence of an external magnetic field ($H = 0$) (i.e., as a result of the pyromagnetic effect, the magnetic analog of the pyroelectric effect).

4. Linear magnetocaloric and pyromagnetic effects in gadolinium iron garnet

It has been a long-held view since the work of Weiss and Forrer [13] that the magnetocaloric effect in ferromagnets (hereafter referred to as the ΔT effect) varies quadratically with magnetization I,

$$\Delta T = aI^2 \tag{13}$$

(where a is a constant), and hence is even. The same behavior is observed in Néel ferrimagnets. However, for weak-sublattice ferrites (specifically, $Gd_3Fe_5O_{12}$) the molecular field method yields at the $T_{\rm B}$ point [10]

$$(\Delta T)_{T=T_{\rm B}} = \frac{v g_S \mu_{\rm B} S (H_{\rm ex})_{\rm eff}}{{}^{\mu}C_V M_0}\, {}^{\mu}\chi_{\rm p} H \,, \tag{14}$$

where v is the number of cations in $Gd_3Fe_5O_{12}$, ${}^{\mu}C_V$ is the heat capacity at constant molar volume, H is the externally applied magnetic field, M_0 is the magnetic moment of the gadolinium sublattice, $(H_{\rm ex})_{\rm eff}$ is the exchange field due to the sublattice of Fe^{3+} cations, S and g_S are the spin and Landé factor of the Gd^{3+} cations, and ${}^{\mu}\chi_{\rm p}$ is the molar susceptibility of the paraprocess in the weak sublattice. It is seen that $(\Delta T)_{T=T_{\rm B}}$ depends linearly on the paraprocess magnetization, i.e., $I_{T=T_{\rm B}} = {}^{\mu}\chi_{\rm p} H$.

The above formula shows that even at the magnetic order–disorder phase transition, the ΔT effect is linear (odd) in magnetization. Subsequent magnetocaloric studies on weak-sublattice ferrites have confirmed this conclusion.

Figure 1 presents magnetocaloric data obtained in Refs [15, 16] on the ferrites $Gd_3Fe_5O_{12}$ and $Y_3Fe_5O_{12}$ in a magnetic field of 16 kOe. In this section, we will be primarily concerned with the ΔT effect in the temperature range from $T_{\rm B}$ (the low-temperature magnetic order–disorder transition) to $\Theta_{\rm com}$ (the magnetic-compensation point). In this range (100–280 K), the dependence of the ΔT effect on temperature is of an asymptotic nature. It is seen that the $\Delta T(T)$ curve is determined by the weak (gadolinium) sublattice because, as

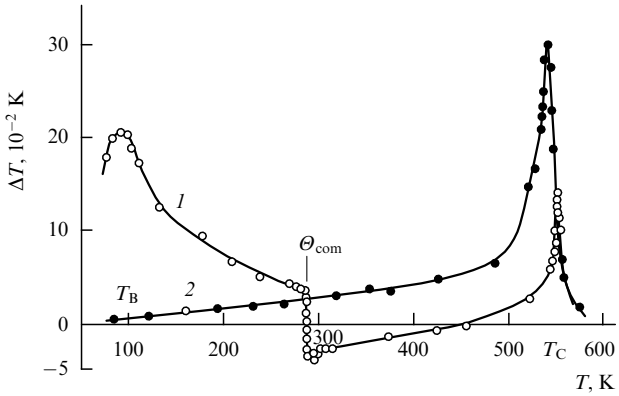

Figure 1. Temperature dependence of the magnetocaloric effect in ferrites in a field of 16 kOe: *1*, $Gd_3Fe_5O_{12}$; *2*, $Y_3Fe_5O_{12}$.

follows from Fig. 1, at low temperatures the sublattice of Fe^{3+} cations (i.e., of yttrium iron garnet $Y_3Fe_5O_{12}$) contributes only little to the magnetocaloric effect.

In Ref. [17], the temperature dependence of the specific magnetization σ was constructed graphically for the c (gadolinium) sublattice in $Gd_3Fe_5O_{12}$ ferrite, both pure and with nonmagnetic cations substituting gadolinium in the a and *ad* sublattices. It can be seen in Fig. 2 that these dependences are also of asymptotic type and that they follow the course of the $\Delta T(T)$ curve, implying a linear ΔT–σ relation. Additional evidence for such a relation is that ΔT changes sign when the magnetization of the gadolinium sublattice changes direction in passing through $\Theta_{\rm com}$ (see Fig. 1).

Further evidence for the linear dependence of the ΔT effect on I is provided by measurements in the immediate vicinity of the compensation point $\Theta_{\rm com}$ [16, 18]. Figure 3

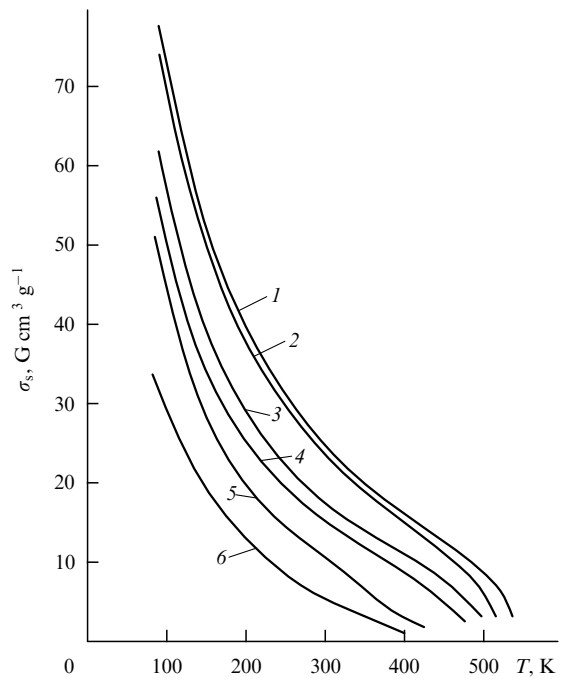

Figure 2. Temperature variation of the spontaneous magnetization of the c ('weak') sublattice for a system of substituted ferrites $Gd_{3-x}Ca_x Fe_{5-x}Sn_xO_{12}$: *1*, $x = 0$; *2*, 0.1; *3*, 0.3; *4*, 0.5; *5*, 0.7; and *6*, 0.9.

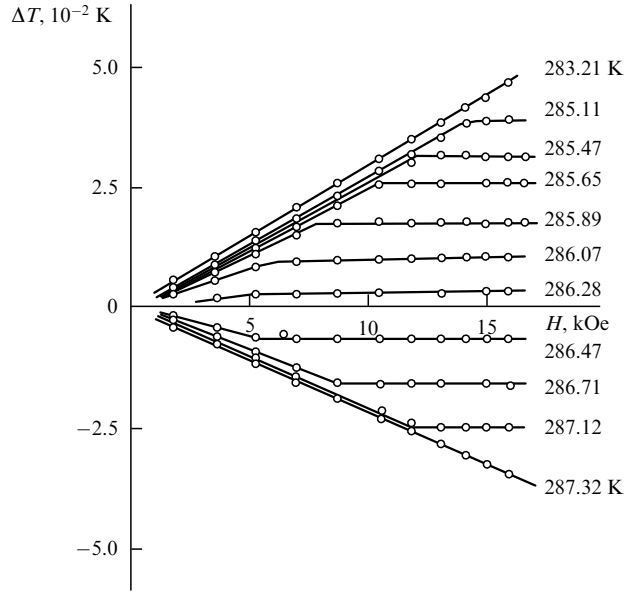

Figure 3. Variation of the magnetocaloric effect with the field near the compensation temperature $\Theta_{com} = 286.3$ K for $Gd_3Fe_5O_{12}$.

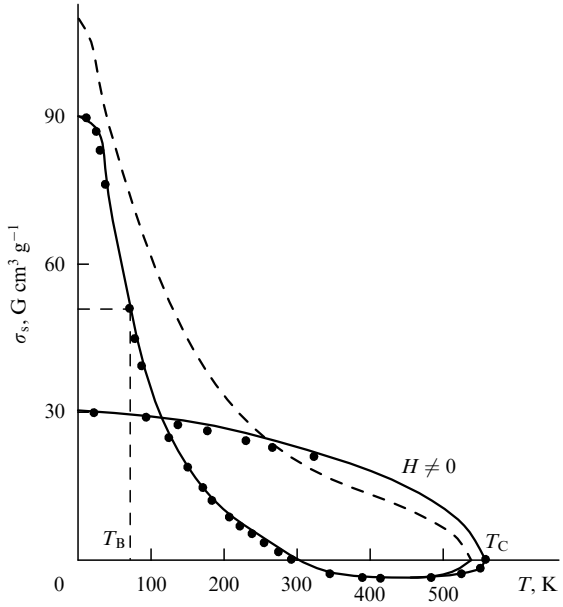

Figure 5. Temperature variation of σ_s for $Gd_3Fe_5O_{12}$ and $Y_3Fe_5O_{12}$.

shows that ΔT varies strictly linearly with H (and hence with I), because χ_p is constant near Θ_{com}.

Indirect evidence for the existence of the pyromagnetic effect $(\Delta I/\Delta T)_{P,H=0}$ in the weak sublattice of $Gd_3Fe_5O_{12}$ comes from the analysis of the isotherms $\sigma(H)$ (Fig. 4) and the temperature dependence $\sigma_s(T)$ (Fig. 5) measured [17] in the temperature range between T_B and Θ_{com}.

It is seen in Fig. 4 that the effect of the paraprocess in a field of 12 kOe on $\sigma(H)$ isotherms is small, i.e., this field causes only a slight increase in the magnetization σ_s (of order 2 – 3 G cm^3 g^{-1}) over the saturation magnetization of the *ad* sublattice (of order ~30 G cm^3 g^{-1}, Fig. 5), whereas at 83 K it is seen from Figs 4 and 5 that the magnetization increase upon cooling the sample is 25 – 27 G cm^3 g^{-1}. The reason for such an increase is the pyromagnetic effect (thermoparaprocess at $H = 0$), which occurs here due to the large, unidirectional exchange anisotropy, because the exchange field is determined by the 'total' spontaneous magnetization $(\sigma_s)_{ad}$ of the *ad* sublattice [cf. Eqn (12)].

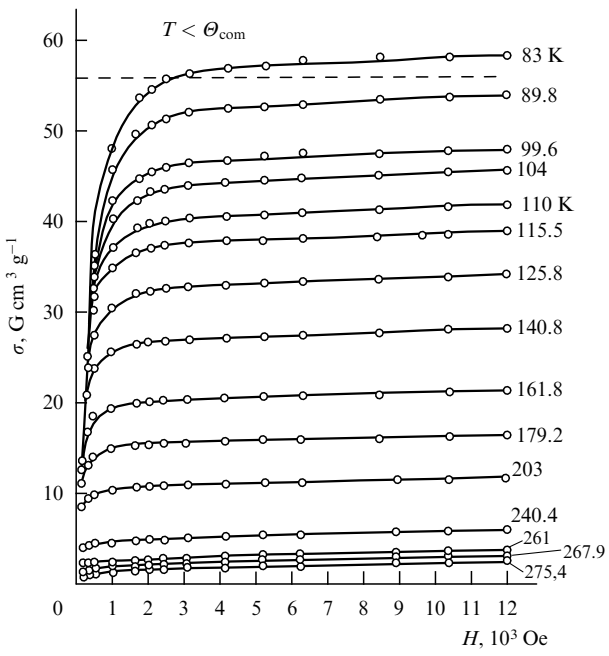

Figure 4. Isotherms $\sigma(H)$ for ferrite $Gd_3Fe_5O_{12}$ at temperatures $T < \Theta_{com}$.

Since $(H_{ex})_{eff} \sim 2 \times 10^5$ Oe according to a molecular field estimate [10], it produces a large increase in the magnetization σ_s on the isotherms $\sigma(H)$ (along the ordinate axis at $H = 0$ in Fig. 4) and gives rise to a steep, asymptotic temperature dependence of the spontaneous magnetization σ_s of the Gd sublattice, as shown by the dashed line in Fig. 5.

The $\sigma_s(T)$ curve in Fig. 5 is steepest between T_B and 150 K, but here, as the course of the $\sigma_s(T)$ curve for $Y_3Fe_5O_{12}$ (i.e., for the *ad* sublattice) suggests, the value of σ_s decreases only insignificantly, so that, according to Eqn (12), the field of the unidirectional anisotropy also remains almost unchanged in this range. This means that the dramatic asymptotic increase in the magnetization of the gadolinium sublattice σ_s with decreasing temperature is due to the pyromagnetic effect. Referring to the data of Ref. [16], this dramatic growth of σ_s along the ordinate axis is most clearly seen (for $H = 0$) in gadolinium iron garnet substituted with nonmagnetic cations (Fig. 6).

5. Direct observation of the pyromagnetic effect in gadolinium iron garnet

The pyromagnetic effect was observed by Lyubutin [19] in residual magnetization measurements on $Gd_3Fe_5O_{12}$. In Fig. 7, σ_r and the coercive force H_c of this material are plotted as functions of temperature T in the range from T_B to Θ_{com} (i.e., from 100 to 293 K).

Figure 8 shows the same for yttrium iron garnet $Y_3Fe_5O_{12}$ [19]. For both ferrites, samples of the same shape and size — 50-mm-long rods of cross section 4 × 4 mm — were used. The residual magnetization σ_r was produced by a strong magnetic field H_c and measured by the coil-drop technique.

Comparing the $\sigma_r(T)$ [and $H_c(T)$] curves for the two ferrites is important because of the fact that whereas

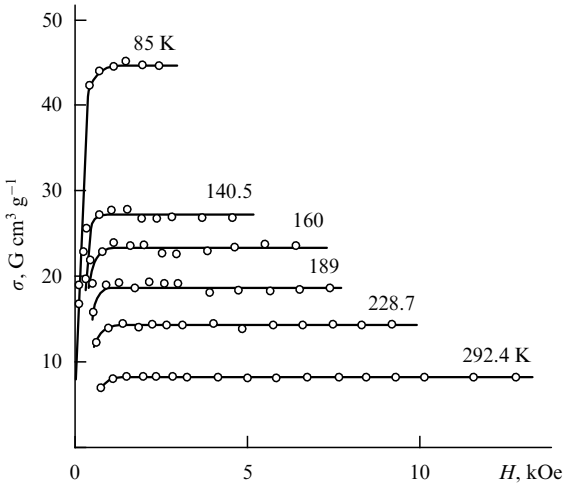

Figure 6. Isotherms of the magnetization $\sigma(H)$ of the substituted gadolinium iron garnet $Gd_3Ga_{1.5}Fe_{3.5}O_{12}$.

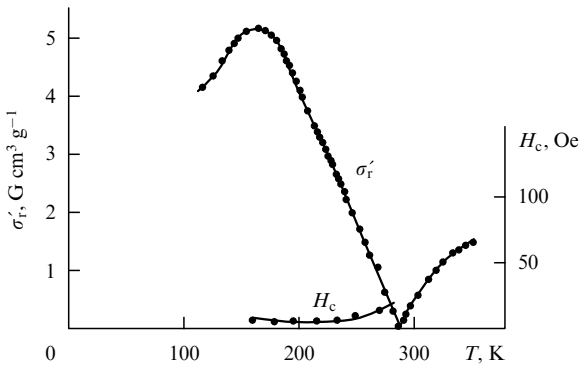

Figure 7. Variation of σ_r and H_c with temperature for $Gd_3Fe_5O_{12}$ in the range $T < \Theta_{com}$.

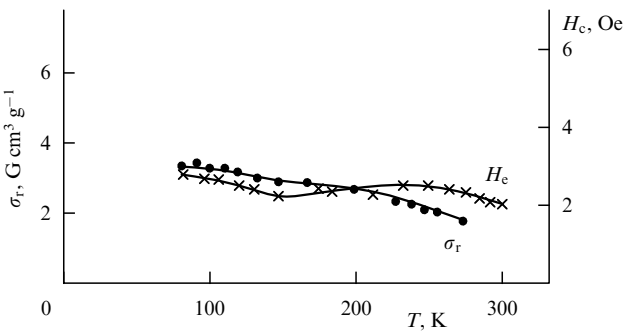

Figure 8. Variation of σ_r and H_c with temperature for $Y_3Fe_5O_{12}$ in the range $T < \Theta_{com}$.

$Gd_3Fe_5O_{12}$ possesses a weak (gadolinium) sublattice, $Y_3Fe_5O_{12}$ does not. Besides, the *ad* (i.e., Fe^{3+} cation) sublattice in $Gd_3Fe_5O_{12}$ is as if the ferrite $Y_3Fe_5O_{12}$, since the cation Y^{3+} has no magnetic moment. For these reasons, from the analysis of the $\sigma_r(T)$ and $H_c(T)$ dependences in the given temperature range the 'irrelevance' of the gadolinium sublattice to the hysteretic properties of $Gd_3Fe_5O_{12}$ can be seen.

The conclusion to be drawn from this is that the residual magnetization of a $Gd_3Fe_5O_{12}$ sample as measured by the coil-shedding technique has two components:

(1) σ_r, the true (hysteretic) component produced by the *ad* sublattice, and

(2) σ_r', the pseudo-residual (nonhysteretic) component induced in the weak sublattice by the unidirectional exchange anisotropy.

The field due to this anisotropy in this case is

$$(H_{ex})_{eff} = -J_{c-ad}(\sigma_r)_{ad}\,, \tag{15}$$

where $(\sigma_r)_{ad}$ denotes the residual magnetization of the *ad* sublattice, and J_{c-ad} is the exchange interaction parameter for the *c* and *ad* sublattices in $Gd_3Fe_5O_{12}$.

Comparing the $\sigma_r(T)$ curves for $Y_3Fe_5O_{12}$ (see Fig. 8) and $Gd_3Fe_5O_{12}$ (see Fig. 7) suggests that the large increase in the residual magnetization with decreasing temperature in gadolinium iron garnet is due to the second component, i.e., the pseudo-residual (nonhysteretic) magnetization σ_r'. In the *ad* sublattice, as follows from Fig. 7, the true residual magnetization σ_r in our temperature range has a very nearly constant value and hence, according to Eqn (15), so does the field of the unidirectional exchange anisotropy. Therefore, the increase in the pseudo-residual magnetization in Fig. 7 should be attributed to the cooling of the sample (in the case of no external field H); what we see here is in fact the magnetic analog of the pyroelectric effect, i.e., the pyromagnetic effect.

Physically, the explanation of the pyromagnetic effect is that decreasing the temperature of the weak sublattice of $Gd_3Fe_5O_{12}$ decreases the detrimental influence of thermal motion on magnetic order, thus giving rise to a thermopara-process (at $H = 0$) because $(H_{ex})_{eff}$ in Eqn (15) is constant in the temperature range considered.

The above conclusion is valid if the weak sublattice does not show magnetic hysteresis. That the gadolinium sublattice does not participate in determining the hysteretic properties of $Gd_3Fe_5O_{12}$ (at temperatures above T_B) is deduced from the measurements of the magnetocrystalline anisotropy constant K_1. In Ref. [20], the contribution ΔK_1 of the Gd^{3+} cations to the constant K_1 of $Gd_3Fe_5O_{12}$ was found to be extremely small (Fig. 9), and its actual value was obtained by subtracting the anisotropy constant K_1 of the ferrite $Y_3Fe_5O_{12}$ obtained in Ref. [21] (see Fig. 9b) from the total value of the K_1 of $Gd_3Fe_5O_{12}$. In Fig. 9 it can be seen that the Gd^{3+} cations of the *c* sublattice contribute to K_1 only at temperatures below T_B (~ 100 K).

For $T > T_B$, the constant K_1 in this ferrite is determined entirely by the Fe^{3+} cations of the sublattice *ad*. The *c* sublattice is in the isotropic state and contributes nothing to the hysteretic properties of $Gd_3Fe_5O_{12}$. The true value of σ_r and H_c is zero in this lattice, unlike the spontaneous magnetization σ_s induced by the unidirectional exchange anisotropy [2].

The analogy between the pyroelectric and pyromagnetic effects is of course formal because their physical mechanisms are different. In pyroelectric materials, the spontaneous polarization P_s varies with temperature as a result of an electric charge redistribution (for $E = 0$).

In the case of a pyromagnetic material, the magnetic order (at $H = 0$) induced in the weak sublattice by the exchange field $(H_{ex})_{eff}$ varies according to Eqn (15) due to the residual magnetization σ_r present in the *ad* sublattice.

Although these two effects have different mechanisms, their manifestations have much in common. First, both occur in dielectric and magnetic materials in which there exists a spontaneous polarization P_s and a spontaneous magnetiza-

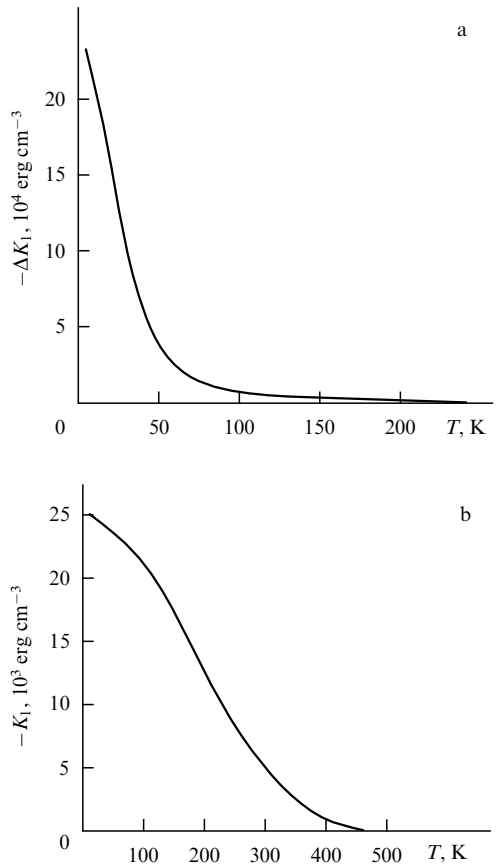

Figure 9. Temperature variation of ΔK_1 (a) for $Gd_3Fe_5O_{12}$ (after [20]; and (b) for $Y_3Fe_5O_{12}$ (after [21]).

tion σ_s, and both the pyroelectric and pyromagnetic materials are in the single-domain state. Another point of similarity is that, as follows from Fig. 7, the pyromagnetic effect (i.e., the value of σ_r') is linear in T.

There is one further thing to note about the way in which the pyromagnetic effect manifests itself. From Eqn (15), σ_r' depends on the value of the true σ_r, and since this latter is structure-sensitive, so must the pyromagnetic effect. That this is indeed the case follows from residual magnetization σ_r' measurements in the temperature range under study [19], which showed that the $\sigma_r'(T)$ curves shift along the T axis in $Gd_3Fe_5O_{12}$ substituted with nonmagnetic cations.

Note however that the pyromagnetic effect always comes together with its piezomagnetic counterpart. The reason is that changing the temperature causes a thermal expansion (or shortening) of a ferrimagnetic sample, and since this can be considered as an additional elastic stress ΔP, a (presumably small) quantity $\Delta I'$ of piezomagnetic origin will be added to the experimentally observed pyromagnetic effect. A similar situation occurs in ferroelectrics when the pyroelectric effect is measured [25].

We turn our attention next to the following experimental fact [19]. Referring to Fig. 7 shows that, as the temperature is lowered, the $\sigma_r'(T)$ curve shows a maximum at ~ 160 K and then declines.

In our opinion, this is due to the fact that as the temperature is lowered and the exchange energy in the weak sublattice increases (due to an enhanced magnetic order), so — and even more intensely — does the magnetic dipole interaction between the magnetic moments of magnetic cations Gd^{3+}, the faster growth rate being due to the long-range nature of this interaction. As a result, the gadolinium sublattice (which is in the single-domain state) tends to be demagnetized, thus causing the domain-formation process. As a result of the magnetic dipole interaction, which thus comes into play to compete with the exchange interaction in the weak sublattice, the magnetic system of this latter becomes metastable and a concomitant relaxation effect arises.

Finally, we may mention a magnetic viscosity maximum for a weak field H (Fig. 10) and a maximum of internal friction Q^{-1} in a zero field (Fig. 11), observed respectively in Ref. [22] and Ref. [23] in the ferrite $Gd_3Fe_5O_{12}$ in the same temperature range as was discussed here. An internal friction maximum was also found [24] in lithium–chromium ferrite between T_B and Θ_{com} in a weak field H (or even at $H = 0$ in

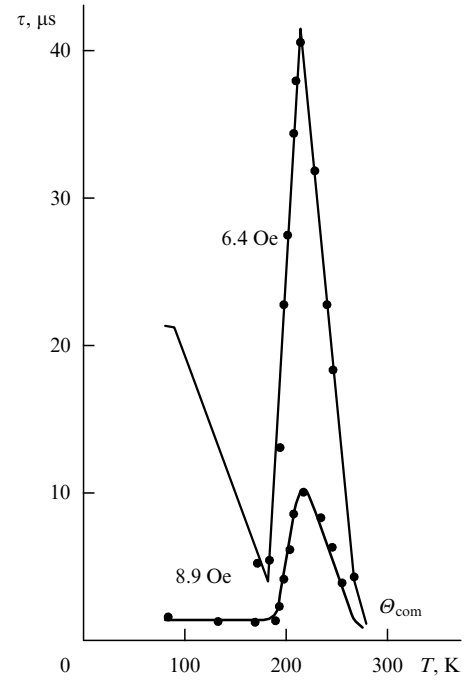

Figure 10. Maximum of magnetic viscosity τ (magnetization reversal time) in $Gd_3Fe_5O_{12}$ below the magnetic compensation point Θ_{com}.

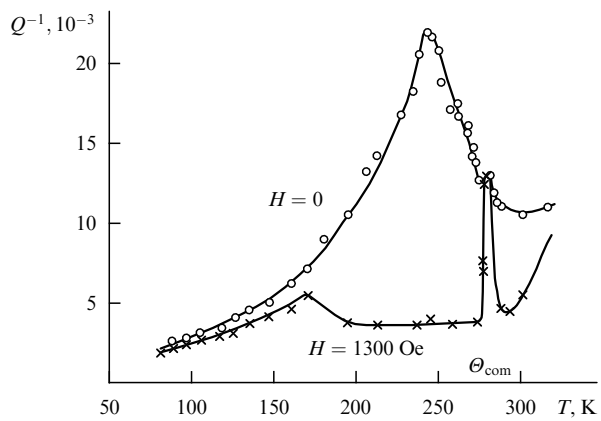

Figure 11. Maximum of internal friction Q^{-1} (in field $H = 0$) in $Gd_3Fe_5O_{12}$ at temperatures below Θ_{com}.

the case shown in Fig. 10). Note that the maximum of Q^{-1} observed in a strong magnetic field of 1300 Oe at the compensation point of $Gd_3Fe_5O_{12}$ (see Fig. 11) is due to the instability that arises in the collinear magnetic structure at this point.

6. Conclusion

Summarizing, we may say that in the 1960s two new research areas emerged in the study of magnetically ordered materials.

First, A S Borovik-Romanov and I E Dzyaloshinksiĭ pioneered the study of antiferromagnets in which unusual magnetic symmetry properties lead to the phenomenon of 'weak' ferromagnetism and give rise to a noncollinear magnetic structure. In these materials, the magnetic analogs of piezoelectricity — the piezomagnetic effect and linear magnetostriction — were discovered, as were some other interesting effects absent in normal antiferromagnets.

The pioneering investigations of the second kind, led by S A Nikitin and the present author, were concerned with the anomalous properties of ferrimagnetic materials with a 'weak' sublattice (with a unidirectional exchange anisotropy and an asymptotic behavior of the spontaneous magnetization in one of the sublattices). Since the 1960s, new phenomena, such as the low-temperature magnetic order–disorder transition (the T_B point), the linear magnetocaloric effect, linear magnetostriction, and the pyromagnetic effect have been observed, and the existence of an exchange-related piezomagnetic effect has been predicted [2]. Also, other magnetic properties absent in normal, Néel type, ferrimagnetic materials, have been revealed.

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