Features of the magnetic behavior and of the magnetocaloric effect in a single crystal of gadolinium

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By measurements of the magnetization and of the magnetocaloric ΔT effect in a Gd monocrystal along various crystallographic directions, we have determined the aperture angle of the cone of axes of easy magnetization in the spin-reorientation process. We have also demonstrated the more complicated nature of the magnetization process at low temperatures and have shown that the field dependence of the magnetic anisotropy constants in the neighborhood of the Curie temperature leads to an abrupt change of sign of the anisotropic contribution ΔT_A to the ΔT effect. It is shown that such a change of ΔT_A should be observed near the Curie point in all magnetic materials.

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This paper poses the problem of studying the complicated magnetic behavior of monocrystalline gadolinium[1,2] by measurements of the magnetization and of the magnetocaloric effect (ΔT effect), which is an extremely sensitive indicator of magnetic transformations.[3] Investigations of the magnetocaloric effect in polycrystalline specimens[1] are not effective enough, because the magnetic anisotropy exerts an appreciable influence on the magnetic properties of gadolinium.

Earlier publications have described the technology for preparing single crystals of gadolinium, and analysis of the impurity concentration, and the orientation of gadolinium crystals along various crystallographic directions,[4] and also the method of measuring the ΔT effect.[5] The magnetization was investigated on the same specimens as was the ΔT effect, with a vibration magnetometer; this made it possible to make a study of the dependence of the ΔT effect on magnetization over the temperature interval 78–450 K.

Figure 1 shows the temperature dependence of the ΔT effect in a magnetic field of 10 kOe directed along the a or the c axis of the crystal. Noticeable features are the maximum of the ΔT effect at the Curie point, 294 K, and the breaks in the $\Delta T(T)$ curve at temperatures 220 and 250 K.

Figure 2 shows $\Delta T(H)$ isotherms, measured in the most characteristic temperature intervals for various directions of the magnetic field with respect to the hexagonal axis c. In strong magnetic fields, the ΔT effect increases linearly with increase of the field, even at low temperatures far from the Curie point. In the magnetic-field range where there is a linear increase of the ΔT effect, the magnetization curves along the a and c axes are close to saturation, and the small increase of magnetization in this field range can be attributed to the paraprocess. It is important that with change of the angle between the magnetic field and the c axis, the intercept on the ordinate axis of the extrapolation of the linear part of the $\Delta T(H)$ curves, which describes the contribution of rotation processes, changes noticeably (see Fig. 2).

To interpret the complicated behavior of the ΔT effect, it is necessary to take account of the following experimental facts. It is known[6,7] that in gadolinium there is a process of reorientation of the spins to the c axis: the magnetization vector makes a certain θ_0 with the c axis over the temperature interval 4.2–240 K, but when $T > 240$ K it orients itself along the hexagonal axis c. The complicated temperature dependence of the aperture angle of the cone is explained by the complicated temperature dependence of the magnetic anisotropy constants. In the range 230–250 K, where K_1 changes sign while $K_2 > 0$ ($K_3 \approx 0$), there occurs a spin-reorientation phase transition of the second kind.[8] It has also been established that the magnetic anisotropy constants of gadolinium strongly depend on the magnetic field.

The measured ΔT effect contains both an isotropic contribution, ΔT_P, due to the change of exchange energy with intrinsic (paraprocess) magnetization, and an anisotropic contribution, ΔT_A, due to the change of magnetic anisotropy energy[9]:

$$\Delta T = \Delta T_P + \Delta T_A, \tag{1}$$

as follows from thermodynamics,[9]

$$\Delta T_P = -\frac{T}{C_{P,H}}\left(\frac{\partial I_s}{\partial T}\right)_H \Delta H, \tag{2}$$

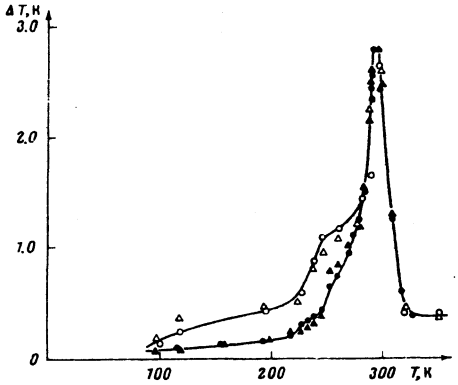

FIG. 1. Temperature variation of the magnetocaloric ΔT effect, measured along the a axis (●) and the c axis (▲), and calculated by formulas (1), (2), and (7) along the a axis (○) and the c axis (△), at $H_i = 10$ kOe.

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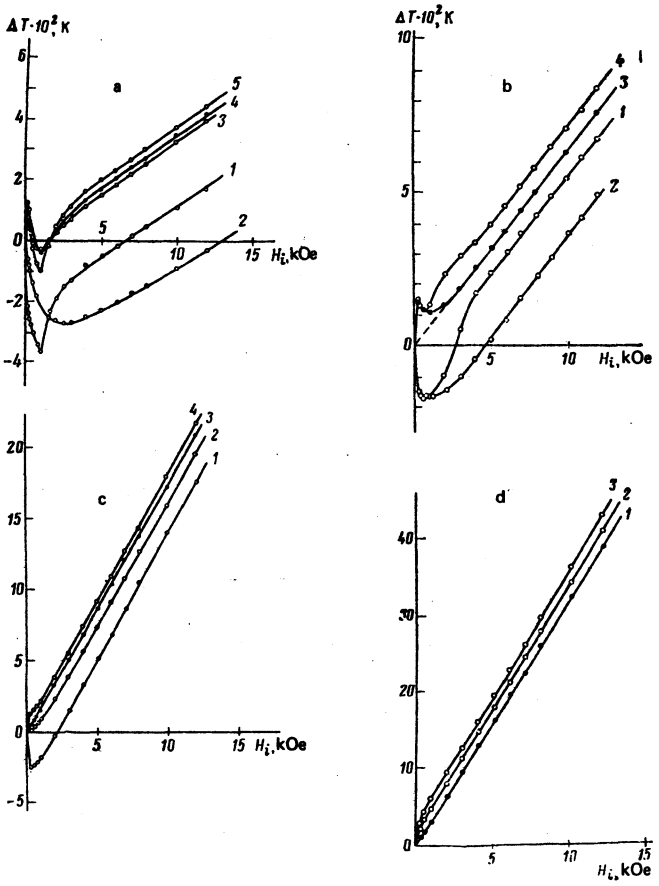

FIG. 2. Field dependence of the ΔT effect, taken along various crystallographic directions, making an angle θ_c with the c axis, a— $T=99$ K and the following values of θ_c: 1—0°; 2—30°; 3—65°; 4—70°; 5—90°; $\theta_0=70°$. b—$T=117$ K and the following values of θ_c: 1—0°; 2—30°; 3—75°; 4—90°; $\theta_0=75°$. c—$T=194.2$ K and the following values of θ_c: 1—0°; 2—50°; 3—80°; 4—90°; $\theta_0=80°$. d—$T=235.5$ K and the following values of θ_c; 1—0°; 2—50°; 3—90°; $\theta_0=0$.

where $C_{P,H}$ is the specific heat and I_S the magnetization. The contribution to the measured ΔT effect is isotropic and positive, since $(\partial I_S/\partial T)<0$.

If we neglect the influence of external and internal stresses on the magnetization, then the rotation of the vectors $\mathbf{I}_S$ will be hindered only by magnetic anisotropy forces; but the contribution ΔT_A will be due not only to the change of the free energy of magnetic anisotropy that results from rotation of the vectors $\mathbf{I}_S$, but also to the change of the magnetic anisotropy energy that results from increase of the magnetization under the influence of the applied field. Thermodynamics enables us to take account of these contributions. In fact, the free energy of magnetic anisotropy for a hexagonal crystal can be written in the form

$$F_a=K_1\sin^2\theta+K_2\sin^4\theta+K_3\sin^6\theta, \tag{3}$$

where K_1, K_2, and K_3 are the magnetic anisotropy constants and where θ is the angle that the magnetization vector makes with the c axis of the crystal. The corresponding contribution to the entropy in magnetic field H will be

$$S(H)=-\left(\frac{\partial F_a}{\partial T}\right)_H=-\left(\frac{\partial K_1}{\partial T}\right)_H\sin^2\theta-\left(\frac{\partial K_2}{\partial T}\right)_H\sin^4\theta-\left(\frac{\partial K_3}{\partial T}\right)_H\sin^6\theta. \tag{4}$$

In a change of the direction of the magnetization vector from the angle θ_0 to the angle θ_c at which the external field H is applied, the corresponding change of the entropy of the magnetic system will be

$$\Delta S_\theta=-\left(\frac{\partial K_1}{\partial T}\right)_H(\sin^2\theta_c-\sin^2\theta_0)-\left(\frac{\partial K_2}{\partial T}\right)_H(\sin^4\theta_c-\sin^4\theta_0)-\left(\frac{\partial K_3}{\partial T}\right)_H(\sin^6\theta_c-\sin^6\theta_0). \tag{5}$$

Since the magnetic anisotropy constants of *Gd* depend on the field, therefore even after completion of the process of rotation of the vectors $\mathbf{I}_S$, when their direction coincides with the direction of the applied field H, an increase of the magnetization in the field will occur, and this will lead to a change of the magnetic anisotropy energy and to a corresponding change of the entropy of the magnetic system, caused by the increase of field:

$$\Delta S_H=-\left(\frac{\partial^2K_1}{\partial H\,\partial T}\sin^2\theta_c+\frac{\partial^2K_2}{\partial H\,\partial T}\sin^4\theta_c+\frac{\partial^2K_3}{\partial H\,\partial T}\sin^6\theta_k\right)\Delta H. \tag{6}$$

Consequently the change of the entropy of the magnetic system will be $\Delta S=\Delta S_\theta+\Delta S_H$.

Taking into account that in adiabatic magnetization the change of the entropy of the lattice has a sign opposite to the sign of the change of the entropy of the magnetic system, we get for the anisotropic contribution to the total ΔT effect

$$\Delta T_A=\Delta T_{A\theta}+\Delta T_{AH}=\frac{T}{C_{P,H}}\left[\left(\frac{\partial K_1}{\partial T}\right)_H(\sin^2\theta_c-\sin^2\theta_0)+\left(\frac{\partial K_2}{\partial T}\right)_H(\sin^4\theta_c-\sin^4\theta_0)+\left(\frac{\partial K_3}{\partial T}\right)_H(\sin^6\theta_c-\sin^6\theta_0)\right]+\frac{T}{C_{P,H}}\left(\frac{\partial^2K_1}{\partial H\,\partial T}\sin^2\theta_c+\frac{\partial^2K_2}{\partial H\,\partial T}\sin^4\theta_c+\frac{\partial^2K_3}{\partial H\,\partial T}\sin^6\theta_c\right)\Delta H, \tag{7}$$

where θ_c is the angle at which the external field H is applied, measured from the c axis, and where θ_0 is the angle between the c axis and the direction of $\mathbf{I}_S$. In this formula there are two terms, both anisotropic; the first term describes the contribution from rotation of the vectors $\mathbf{I}_S$ against the forces of magnetic anisotropy, whereas the second term describes the contribution from change of the magnetic anisotropy constants under the influence of the applied field.

With formulas (2) and (7) it is possible to make an estimate of the contributions ΔT_P and ΔT_A, and consequently also an estimate of the total (measured) ΔT effect, if one knows the value of $C_{P,H}$ and the functions $I_S(T)$, $K_1(T)$, $K_2(T)$, and $K_3(T)$, and also the angles θ_c and θ_0. The variations of K_1, K_2, and K_3 with temperature and field have been investigated by many authors;[6,7,10] it must be remarked that both the values and the temperature behavior of the constants as determined by different authors are somewhat different.

In order to take account of the field dependence of the anisotropy constants, we carried out a calculation of the functions $K_1(H, T)$ and $K_2(H, T)$ according to the formulas of Yang.[11] In these calculations, we used our measured variations of the magnetization on temperature and field and values of the angles θ_0 obtained by us from measurements of the ΔT effect (see below). The values of $K_1(0,0)$, $K_2(0,0)$, and $\sigma(0,0)$[1)] were taken from the papers of Graham[10] and of Nigh *et al.*[12] We remark that the temperature variation of the magnetic anisotropy constant K_1 according to the results of the calculation by Yang's formula agrees well with Graham's data,[10] while the values of $K_2(T)$ in the low-temperature range deviate somewhat from his data.

Figure 3 gives the variations of K_1 and K_2 with reduced magnetization $m = \sigma(H, T)/\sigma(0,0)$ that we obtained and that were used for estimation of the total ΔT effect. It should be mentioned that this estimate is of qualitative character, because in Yang's formulas, which we used, the numerical coefficients were determined on the basis of Graham's experimental work,[10] the data of which were subsequently improved by other authors.[7]

The values of $C_{P,H}$ for this estimate were taken from the work of Belov *et al.*[1] and the values of $K_3(T)$ from that of Corner *et al.*[6] Here it should be mentioned that at all temperatures exceeding $T = 240$ K, at which the constant K_3 vanishes, formula (7) simplifies. The results of the calculations (see Fig. 1) agree with the data of measurements of the ΔT effect in the temperature range 280–330 K; but in the temperature range lying below 280 K, the values that we obtained for the ΔT effect are somewhat lower than the calculated values. One of the possible reasons for this deviation, besides the reason indicated above, may be a certain negative contribution not taken into account in the calculations and due to the more complicated character of the magnetization process that occurs in Gd in this range of temperature; of this we shall have more to say below.

The measurements of the ΔT effect along various crystallographic directions at various temperatures enabled us to trace the process of reorientation of spins to the c axis, which occurs in Gd on rise of temperature. The method of determining the aperture angle of the cone during the process of spin reorientation, by use of measurements of the ΔT effect, was described in detail earlier.[14] From analysis of formula (7) in those cases in which the contribution ΔT_A is determined solely by the first term $\Delta T_{A\theta}$ of formula (7), it follows that if the external field H is applied along various crystallographic directions at angle θ_c to the c axis, then when $\theta_c = \theta_0$, where θ_0 is the angle at which the spins are inclined during their reorientation process, the contribution $\Delta T_{A\theta}$ of rotation processes will be zero (or when $\partial K_i/\partial T = 0$). On extrapolation of this isotherm to $H = 0$, it will pass through zero.

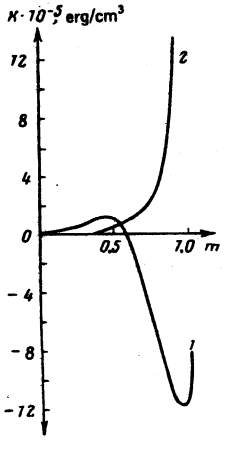

FIG. 3. Dependence of magnetic anisotropy constants (1—K_1; 2—K_2) on reduced magnetization $m = \sigma(H, T)/\sigma(0, 0)$ at $H_i = 10$ kOe.

Far from the Curie temperature, in the paraprocess region, where the process of technical magnetization is complete, ΔT varies linearly with H; therefore by extrapolating the $\Delta T(H)$ curves to zero field, one can separate out $\Delta T_{A\theta}$ for each crystallographic direction. We note that when only this contribution to ΔT_A is present, the paraprocess is isotropic; and at fields where the paraprocess is observed ($H_i > 5$ kOe), the $\Delta T(H)$ curves will be parallel to one another. From analysis of formula (7) it follows also that the contribution of rotation processes may be either positive (for $\theta_c > \theta_0$); this is explained by the fact that rotation of the vectors $\mathbf{I}_S$ to the direction of H requires a different energy when $\theta_c > \theta_0$ and when $\theta_c < \theta_0$. The second term in formula (7) is a contribution that does not vanish even after the process of rotation of the vectors $\mathbf{I}_S$ is complete, since with increase of the field there will occur also an increase of the magnetization and a corresponding change of the magnetic anisotropy constants (see Fig. 2). This will lead to the result that in strong magnetic fields, $\Delta T(H)$ curves taken along different crystallographic directions ($\theta_c \neq \theta_0$) will no longer be parallel to one another. As our measurements and numerical estimates have shown, the role of the second term in formula (7) in strong magnetic fields begins to show up only when $T > 230$ K, to the extent that the increase of magnetization in the paraprocess at temperatures $T > 230$ K is large.

Analysis of the behavior of the isotherms $\Delta T(H)$ enabled us to determine the angle θ_0 at which the spins are inclined during their reorientation process (see Fig. 4a). The same figure shows also data from neutron-diffraction investigations.[13] As is seen from Fig. 4a, the values of the angles determined by measurements of the ΔT effect and by neutron diffraction agree quite well over the temperature range 200–260 K and differ slightly when $T < 200$ K, although they lie within the limits of measurement error indicated for the neutron-diffraction investigations. From Fig. 1, where measured and calculated values of the ΔT effect along the a and c axes are presented, it is seen that when $T < 280$ K there is a noticeable divergence of the measured and calculated values of the ΔT effect. Furthermore, at low temperatures, on the isotherms $\Delta T(H)$ in the low-field region, there is a noticeable negative component even for crystallographic directions that lie on the cone of axes of easy magnetization ($\theta_c = \theta_0$). This suggests that not all the directions of magnetization on the surface of this cone are energetically equivalent.

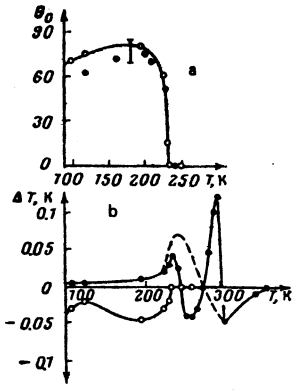

FIG. 4. a—Temperature dependence of the aperture angle θ_0 of the cone of axes of easy magnetization, determined from measurements of the ΔT effect (○) and from neutron diffraction data (●) [13]. b—Temperature dependence of the anisotropic contribution ΔT_A, measured along the c axis (○) and along the a axis (●) of a Gd crystal. The dashed curve shows the contribution $\Delta T_{A\theta}$ calculated by formula (8).

The magnetization process at these temperatures proceeds in such a manner that with increase of the field H there occurs a rotation of the projections of spins initially in the basal plane, as a result of which a magnetic structure is formed that is energetically more advantageous (the maximum on the $\Delta T(H)$ curves), at fields ~0.1 kOe (see Fig. 2a); then with further increase of the field, this magnetic structure is destroyed, for which energy is expended that is withdrawn under adiabatic magnetization from the lattice (the minimum on the $\Delta T(H)$ curves at $H_i \sim 1$ kOe; see Fig. 2a); after this, the spins depart from the cone of axes of easy magnetization, setting themselves at $h_i \sim 5$ kOe along the direction of the applied field.

Thus during the magentization process, with increase of field there is a suppression first of the magnetic anisotropy in the basal plane, and then of the uniaxial magnetic anisotropy. With increase of temperature (see Fig. 2b or c), the behavior of the $\Delta T(H)$ curves in weak fields becomes progressively simpler; the process of reorientation of spins to the c axis is completed, and at $T = 235.5$ K the c axis becomes the axis of easy magnetization (See Fig. 2d). From the behavior of the curves in Fig. 2a, b, and c it is seen that at temperatures 78–200 K the role of the field dependence of the anisotropy constants is not yet in evidence, the paraprocess is isotropic, and the $\Delta T(H)$ curves at fields $H_i > 5$ kOe run parallel to one another. At temperature $T = 235.5$ K, the field dependence of the anisotropy constants is already beginning to show up (see Fig. 2d); this is also confirmed by numerical estimates that we have made. For temperatures $T > 230$ K, we determined the contributions to ΔT_A along the a axis not by extrapolation of the $\Delta T(H)$ curves to zero field, but as the difference between ΔT^c and ΔT^a, measured at $H_i = \text{const} = 10$ kOe.

Figure 4b gives the contributions ΔT_A of rotation processes to the total ΔT effect, as we determined them experimentally for crystallographic directions corresponding to the a and c axes of the crystal. The contribution ΔT_A^c remains negative until completion of the spin-reorientation process and vanishes when $T = 235.5$ K. Since for $T > 235.5$ K the c axis remains the axis of easy magnetization, the contribution of rotation processes along the c axis remains zero at higher temperatures also. The complicated temperature dependence of ΔT_A^a (Fig. 4b) can be explained by the fact that with approach to the Curie temperature, the contribution ΔT_{AH} increases; in the neighborhood of the Curie temperature, this contribution not only affects the magnitude of ΔT_A but even changes the sign of this term.

In fact, in the absence of field-dependence of the anisotropy constants, the contribution ΔT_A, measured along the a axis, can be determined according to the formula

$$\Delta T_A^a = \Delta T_{A\theta}^a = \frac{T}{C_{P,H}}\left(\frac{\partial K_1}{\partial T} + \frac{\partial K_2}{\partial T}\right), \tag{8}$$

since $\sin\theta_c = \sin 90° = 1$, while $\sin\theta_0 = \sin 0 = 0$ and $K_3 = 0$.

Since in the temperature range $T > 230$ K we have $|\partial K_1/\partial T| \gg |\partial K_2/\partial T|$ (the constant $K_2 = 0$ at $T = 290$ K), the sign and character of this contribution should be determined principally by the first term, that is by the temperature dependence of K_1. It is known[7] that K_1 increases over the temperature range 230–280 K; this increase becomes gradually slower with increase of temperature, and at $T = 280$ K the constant K_1 goes through a maximum. But the contribution $\Delta T_{A\theta}^a$ does not vanish here because $\partial K_2/\partial T \neq 0$ at this temperature. Thereafter, with further increase of temperature, when K_1 decreases ($\partial K_1/\partial T < 0$) and $K_2 = 0$ at 290 K, the contribution $\Delta T_{A\theta}^a$ should remain negative and gradually decrease in value (see Fig. 4b).

But estimates that we have made show that in the neighborhood of $T = 280$ K, where the value of $\Delta T_{A\theta}$ is nearly zero, there is a large increase of ΔT_{AH}; at $T = 280$ K the value of $\Delta T_{AH} > 0$, and this also determines the sign of the positive total contribution ΔT_A. With approach to the Curie temperature the contribution ΔT_{AH} increases, remaining positive; but then it abruptly changes sign, becomes less than zero, and gradually decreases in value, remaining negative (see Fig. 4b). In Fig. 4b the curve of the contribution $\Delta T_{A\theta}$ is plotted with dashes. From a comparison of this curve with our experimental curve it is seen that near the Curie temperature, the value and sign of ΔT_A^a are determined by ΔT_{AH}; that is, by the contribution that arises from the presence of a field dependence of the anisotropy constants.

It must also be remarked that the estimation of the contribution ΔT_{AH} is itself quite difficult, since it requires finding the derivative with respect to T of the product of two other derivatives, $(\partial K_1/\partial m)(\partial m/\partial H)$, and therefore is very approximate. Nevertheless this estimate, both in order of magnitude and in the character of the $\Delta T_A^a(T)$ curve, confirms the behavior of our experimental curve. The character of the $\Delta T_A^a(T)$ curve for $T < 280$ K can also be revealed by comparison of the curves ΔT^a and ΔT^c of the total ΔT effect measured along the a and c axes of the crystal, shown in Fig. 1.

It should be mentioned that on the basis of general

ideas of the theory of magnetism,[15-18] the magnetic anisotropy constants are ultimately determined by the value of the magnetization (within a domain). Therefore the quantity

$$\frac{\partial^2 K}{\partial H \partial T} = \frac{\partial}{\partial T}\left(\frac{\partial K}{\partial m}\frac{\partial m}{\partial H}\right)$$

which determines the behavior of ΔT_{AH} in the Curie-point region (see formula (7)), is larger than zero for $T<\theta$ and less than zero for $T>\theta$ as a result of the fact that $\partial m/\partial H$, as is well known, goes through a maximum at the Curie point (the maximum of the paraprocess susceptibility); therefore the $\Delta T_A(T)$ curve should change sign for all ferromagnets. Thus the change of sign at ΔT_A in the Curie-point region should be a phenomenon common to all ferromagnets; the actual character of the $\Delta T_A(T)$ curve will be determined by the specific form of the $K(m)$ curve for the given magnetic material.

In conclusion, it should be remarked that over the range 90–230 K, in analysis of the behavior of the magnetization and of the ΔT effect it is necessary to take into account the change, with rise of temperature, of the apeature angle of the cone of axes of easy magnetization and the reorientation of the spins at $T = 235$ K. In the low-temperature range, the effect of magnetic anisotropy in the basal plane becomes important, and this leads to the result that among the directions of magnetization on the surface of the cone with angle θ_0, there appear energetically more advantageous axes of easy magnetization. From the results of our investigation the conclusion must be drawn that the field dependence of the magnetic anisotropy constants in the Curie-temperature region determines the value and sign of the ΔT_A effect and exerts an important influence on the magnetization process and the magnetocaloric effect.

In conclusion, the authors express their deep gratitude to K. P. Belov and R. Z. Levitin for discussion of the results.

[1)]Here $\sigma(0, 0)$ is the value of the specific magnetization of absolute saturation.

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