Influence of a magnetic field on the Young modulus of a hexagonal ferromagnet with easy-plane anisotropy ($Tb_{0.4}Gd_{0.6}$ single crystal)

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The method of flexural vibrations of frequency ~ 1 kHz was used to measure the Young modulus $E = s_{11}^{-1}$ (s_{11} is the elastic compliance constant) along the a and b axes in the basal plane of the hexagonal ferromagnetic crystal $Tb_{0.4}Gd_{0.6}$ (with easy-plane anisotropy). The measurements were carried out in magnetic fields up to 14.6 kOe at temperatures 78–320 K. Negative (up to 12%) and positive (up to 33%) ΔE effects were observed (this effect is the change in the Young modulus in a magnetic field). Hysteresis loops of the ΔE effect were obtained and the field dependence of the internal friction was determined. The experimental results were compared with theoretical calculations of the field dependences of the Young modulus, which were carried out using two models: (a) Displacement of domain walls under the influence of elastic stresses and of a magnetic field in the absence of rotation of the moments inside a domain (statistical theory); (b) change in the equilibrium direction of the magnetization under the influence of the same factors but in the case of complete pinning of domain walls. Both models predict a negative ΔE effect under certain specific conditions.

INTRODUCTION

Investigations of the influence of a magnetic field and of temperature on the Young modulus and mechanostriction have been made, beginning from the thirties,[1] mainly on polycrystalline ferromagnets with the cubic crystal lattice. Akulov, Kondorskii, Becker and Doring, Kersten and others have analyzed the characteristic features of the ΔE effect in these materials, but the attention has been concentrated on the maximum value of the ΔE effect because calculations of the field dependences of the elastic modulus have met with serious difficulties. Subsequently, Yamamoto and Taniguchi[2] established the important role played by the pinning of domain walls in the change of the Young modulus of a ferromagnet on application of a magnetic field. A negative ΔE effect was attributed to the presence of a maximum in the field dependence of the magnetic susceptibility $\chi(H)$. The experiments showed however, that such a one-to-one correspondence is not always observed.[3] Among the early theoretical investigations, we should mention the work of Takagi,[4] in which a theory of mechanostriction is generalized by a statistical method for the description of a cubic ferromagnet, allowing for the displacement of domain walls and for the field-induced rotation of the magnetic moments in the domains. This is one of the few theories that accounts satisfactorily for the field dependence of the Young modulus and mechanostriction not only in the case of thoroughly investigated alloys and compounds of Fe, Ni, and Co, but also in the case of rare-earth compounds such as those of the RFe_2 type (R is a rare-earth metal).[5] However, this theory does not consider changes in the Young modulus of uniaxial ferromagnets and ignores hysteresis. Among the later investigations we should mention that of Belov *et al.*,[6] where again a cubic crystal is considered and the maximum ΔE effect is calculated allowing for the rotation processes and including not only the first but also the second anisotropy constant. Magnetoelastic renormalization of the velocity of sound in cubic magnetic materials is treated in Refs. 7 and 8 allowing for the appearance of a magnetoelastic gap.[7,8]

There have been far fewer investigations of the Young modulus of uniaxial ferromagnets with the hexagonal lattice. The magnetic field dependence of the elastic moduli originating from the magnetoelastic interaction was considered in Refs. 9 and 10. However, the mechanostriction and the ΔE effect in magnetic materials with easy-plane anisotropy, such as heavy rare-earth metals and their alloys, have not yet been investigated at all. Such investigations would undoubtedly be of interest, particularly because the existence of metastable (canted) phases may give rise to peculiarities of the field dependences of the mechanostriction and of Young's modulus when hexagonal magnets are subjected to magnetization reversal in the basal plane.

We selected a single crystal of the alloy $Tb_{0.4}Gd_{0.6}$, some of whose physical properties have been investigated earlier, particularly the magnetization[11] and the basal anisotropy.[12] The temperature dependence of the Young modulus $E(T)$ along the hexagonal axis c and the corresponding internal friction of this crystal were determined earlier[13] between 4.2 and 330 K, but in the absence of a magnetic field (the field would have had little influence on the Young modulus of the sample because of the strong anisotropy along the hexagonal axis). We shall report (§ 1) the experimental temperature and field dependences of the Young modulus $E(T, H)$ in the basal plane of the same single crystal, and then we shall give the results (§§ 2,3) of a theoretical calculation of the dependences $E(H)$ for a hexagonal easy-plane ferromagnet and we shall consider two different mechanisms of the magnetoelastic renormalization of the Young

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modulus. We shall simplify the problem by considering two limiting cases: 1) complete absence of pinning of domain walls, when magnetization reversal involves displacements of these walls in low fields weaker than the anisotropy field $H \ll 36K_{66}/M_s$; 2) complete pinning of domain walls in a single-domain sample subjected to an arbitrary field. We shall follow this by an analysis (§ 4) of these two limiting cases and consider the real situation when partial pinning of domain walls occurs in a sample and there are metastable phases in which irreversible abrupt changes in the magnetization are possible, and we shall then give the conclusions.

§ 1. EXPERIMENTAL RESULTS

Measurements of the Young modulus E, of the ΔE effect, and of the internal friction Q^{-1} were made using apparatus modified somewhat compared with that described earlier[13,14]: we employed the method of flexural vibrations of a cantilevered sample with the smallest dimensions of $7\times2\times0.1$ mm. The vibrations were excited by an electrostatic method in which a change in the capacitance of the system formed by a sample and an electrode, which was part of a 50-MHz oscillator circuit, resulted in a change of the "carrier" frequency. The low-frequency component was selected by an amplifier and after amplification and passage through a phase shifter it reached the electrode thus closing the feedback circuit and exciting natural vibrations of the sample. Care was taken to ensure stabilization of the amplitude of the vibrations. The vibration frequency was measured with a frequency meter. The Young modulus E of a sample measured parallel to the long axis was proportional to the square of this frequency. Since according to our estimates the average amplitude of the relative strain experienced by the sample was only 10^{-7}–10^{-6}, when allowance was made for the smallness of the transverse dimensions of the sample and the corresponding transverse deformation, the elastic deformation could be regarded as longitudinal elongation or compression parallel to the long axis. In the case of hexagonal crystals with the long axis parallel to the crystal axis c, it was found that $E_c = s_{33}^{-1}$, whereas along the a and b axes, the corresponding relationship was $E_{a,b} = s_{11}^{-1}$, where s_{ii} are the elastic compliance constants of the material. The absolute value of E was determined to within 2–3% and the change in E to within $\pm 0.02\%$.

The reciprocal of the mechanical Q factor of the sample Q^{-1} was determined employing an amplitude discriminator and a pulse counter in the absence of the feedback circuit. The values of Q^{-1} were determined to within 2% (in the case of weak damping) or to within 8–10% (strong damping). An enclosure containing a sensor and the sample was evacuated and placed inside the gap of an electromagnet which created magnetic fields up to 15 kOe.

Our single crystals were grown at the State Scientific-Research and Design Institute of the Rare-Metal Industry in Moscow: they used the Czochralski method and obtained the samples in which the microblocks were misoriented relative to one another by angles not exceeding 1°. The purity of the rare-earth metals used to grow single crystals was 99.9%. Samples of the $Tb_{0.4}Gd_{0.6}$ composition were cut by spark machining along the a and b crystallographic axes.

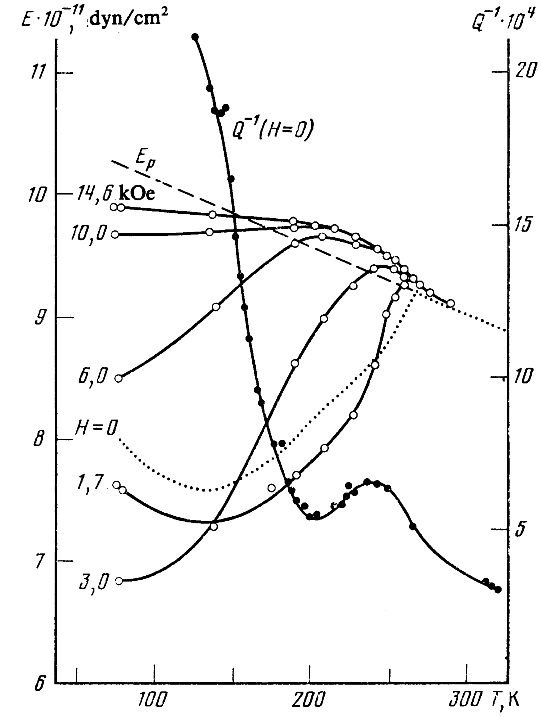

FIG. 1. Temperature dependences of the Young modulus $E = 1/s_{11}$ measured in different magnetic fields applied along the b axis to a single-crystal sample of $Tb_{0.4}Gd_{0.6}$, and of the internal friction Q^{-1} in the same sample in the magnetized state.

Figure 1 shows the temperature dependences of the Young modulus $E_b = s_{11}^{-1}$ and of the corresponding internal friction Q^{-1} of a sample cut in the basal plane along the easy magnetization axis b (a is the width and c is the thickness). The temperature dependences $E(T)$ (dotted line) and $Q^{-1}(T)$ were determined for a demagnetized sample and then the dependences $E(T)$ were recorded in magnetic fields up to 14.6 kOe applied along the b axis. Below the Curie point $\Theta = 269$ K there was a strong ferromagnetic anomaly of the Young modulus, i.e., the modulus E decreased compared with its value E_p found by extrapolation to a given temperature $T<\Theta$ of the paramagnetic value of E in the range $T>\Theta$. The anomaly varied with the field at it exhibited a change of the "sign" of the kinks on the $E(T)$ curves in the region of Θ. A small maximum of Q^{-1} was observed in the same region and this was first observed earlier at the Curie points of magnetic materials.[15]

The same results were used in Fig. 2 to plot the field dependences of the ΔE effect, i.e., of the value of $(E_H - E_0)/E_0 = \Delta E/E_0$, where E_0 is the Young modulus of a demagnetized sample and E_H is the same modulus in a field H. At 141 K the positive ΔE effect had its maximum value of 29% in a field of 14.6 kOe, and at 78 K it had its maximum negative value 11%. Similar curves were obtained also for a sample with the long axis a, which was again in the basal plane (b is the width and c is the thickness), but in this case the maximum positive ΔE effect reached 33% (at 105 K) and the maximum negative effect was 12% (at 78 K).

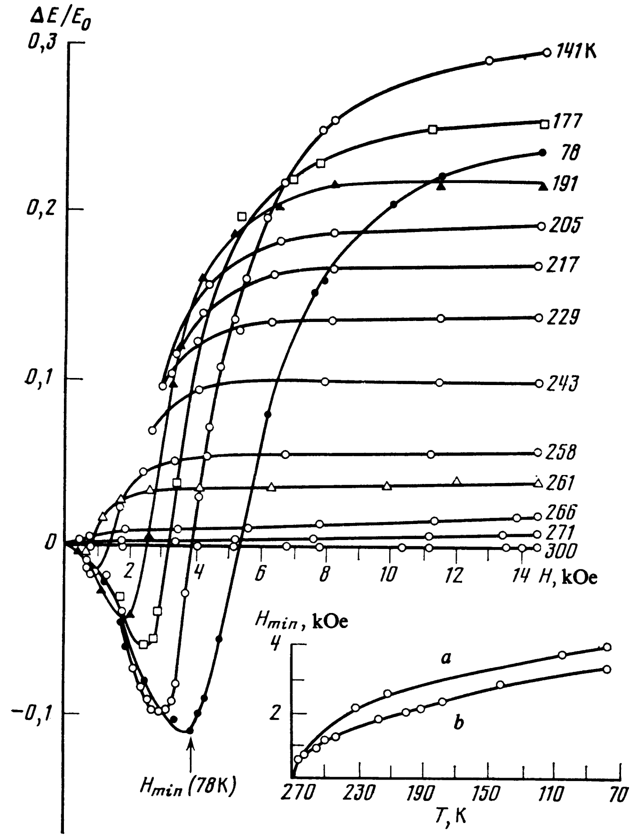

FIG. 2. Magnetic field dependences of the ΔE effect for a single-crystal sample of $Tb_{0.4}Gd_{0.6}$ cut along the b axis, determined at various temperatures. The inset shows the temperature dependences of the field corresponding to the minimum ΔE effect in samples cut along the a and b axes.

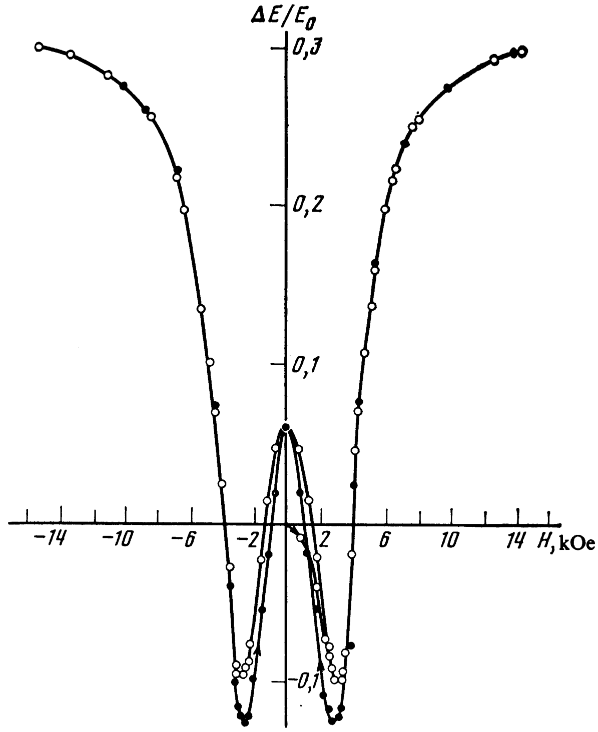

FIG. 3. Hysteresis loop of the ΔE effect in a single-crystal sample of $Tb_{0.4}Gd_{0.6}$ cut along the b axis, recorded at 141 K.

The small difference between the curves for the long axes a and b could be due to not only the difference between the behavior of the domains when the field was directed along a and along b, but also to a magnetic distortion of the hexagonal lattice (morphic effect[16,17]). The inset in Fig. 2 shows the temperature dependences of the field $H_{\min}$ corresponding to the minimum of the ΔE effect. For the sample cut along the easy-magnetization axis b, the $H_{\min}(T)$ curve was lower than that for the sample cut along the a axis.

Figure 3 shows a hysteresis loop of the ΔE effect obtained at 141 K for a sample with the long axis parallel to b. At 78 K the sample with the long a axis also exhibited a hysteresis loop of the ΔE effect and a similar though inverted $Q^{-1}(H)$ loop. Naturally, each branch of the loop was governed by the previous history of the domain structure of the sample, mainly in the range of fields causing displacements of domain walls, but possibly also effects other than displacements (see § 3). The almost "mirror" symmetry of the $\Delta E/E_0$ and Q^{-1} loops can be explained on the basis of a relaxation theory put forward in Ref. 15.

According to a theory of even effects, confirmed by the measurements on nickel[18] and on ferrites,[19] the ΔE effect observed in the range of fields causing the domain-wall displacements and (with a larger coefficient) in the range of fields causing rotation is proportional to the square of the relative magnetization. In the rotation range of fields this dependence follows also from a model theory for a gadolinium single crystal,[9] which can be extended (subject to small modifications) to other rare-earth metals. On the other hand, in high fields or close to Θ, where the influence of these fields on the elastic constants is due to a change in the exchange interaction or due to the paraprocess, the linear magnetostriction is very closely proportional to the square of the magnetization,[20] whereas the changes in the elastic constants should follow less closely this proportionality.[15]

We used the detailed data on the field dependences of the specific magnetization σ of the same single crystal obtained at various temperatures (Fig. 4)[11] and the data on the

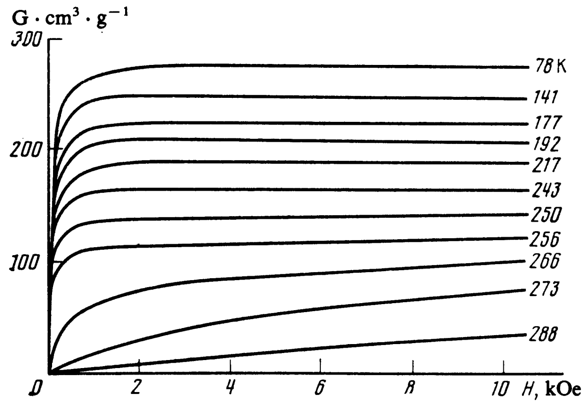

FIG. 4. Dependences of the specific magnetization of a single-crystal sample of $Tb_{0.4}Gd_{0.6}$ on the magnetic field applied at various temperatures.[11]

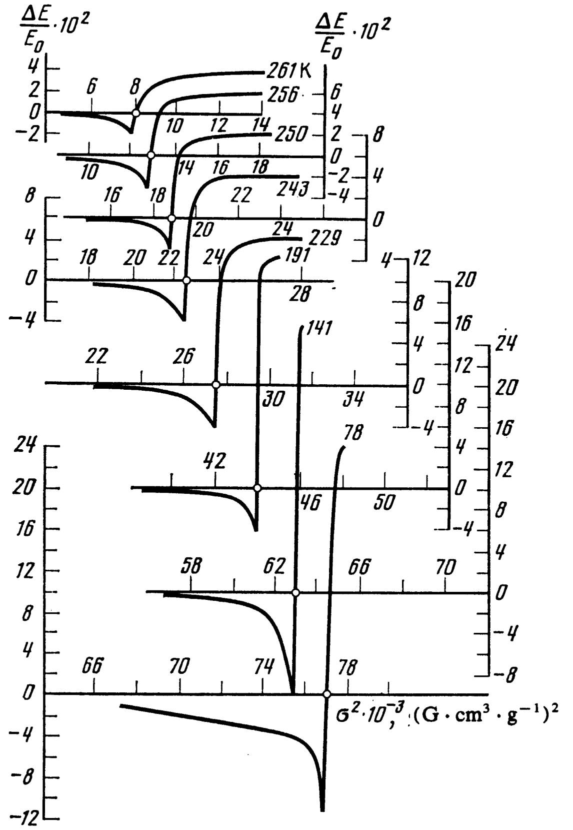

FIG. 5. Dependences of the ΔE effect in a single-crystal sample of $Tb_{0.4}Gd_{0.6}$ along the b axis on the square of its specific magnetization, recorded at different temperatures ($H \parallel b$).

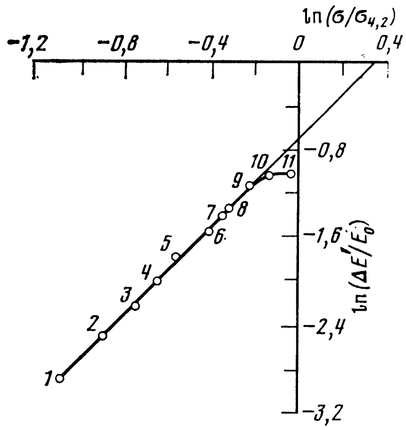

FIG. 6. Dependence of $\ln E'/E_0$ along the b axis on $\ln(\sigma/\sigma_{4.2})$, recorded at different temperatures (K): 1) 261; 2) 256; 3) 250; 4) 243; 5) 229; 6) 217; 7) 204; 8) 191; 9) 177; 10) 141; 11) 78.

dependence of $\Delta E/E_0$ on H (Fig. 2), and we found that at temperatures between 78 and 261 K the dependences of $\Delta E/E_0$ on σ^2 obtained for a crystal sample along the b axis had the form shown in Fig. 5. The majority of these curves exhibited three characteristic linear regions linked by rounded transition regions. The lower flat curve observed at 78 K corresponded to the region of displacement of domain walls and to the negative ΔE effect observed on increase in the mechanostriction strain; the upper flat part was due to the "exchange" component of the ΔE effect. The slope of the latter region decreased on approach to Θ. At 78 K this region was missing from the data, because at this temperature a field of 14.6 kOe was clearly insufficient and the $E(T)$ curves of Fig. 1 did not reach the curve representing the extrapolated (paramagnetic) modulus $E_p(T)$.

The almost vertical linear part ("jump") in curves of Fig. 5 clearly corresponded to the region of the fastest rise of the modulus E on increase in the field (Fig. 2), which occurred in the region where the magnetization increased only slightly (Fig. 4). Clearly, the processes of domain wall displacements and some of the rotation processes, responsible for a large increase in the magnetization, were complete in fields exceeding 2 kOe, but they had little effect on the rise of the Young modulus. Such a jump in E was difficult to account for simply by the rotational processes even in the case of a single crystal. According to a theory put forward below (§ 3), such a jump may be attributed to an abrupt reorientation of the remaining non-180° domains or any easy magnetization axes in the basal plane from any of the easy-magnetization axes in the basal plane to one axis closest to the field direction. To answer some of the questions we required the knowledge of the dynamics of domains in the investigated sample, but it was difficult to observe the domain structure of rare-earth metals and alloys in high fields and at low temperatures. In the case of pure Tb it was found[21] at 4.2 K that there were narrow (500 Å) stripe domains with 180° boundaries and a certain number of "closure" domains with 60° and 120° boundaries. Our observations (obtained in cooperation with E. E. Shalygina) of the equatorial Kerr effect in the basal plane of a $Tb_{0.4}Gd_{0.6}$ crystal, carried out in alternating fields of amplitude up to 200 Oe at $T \approx 80$ K and recording displacements of domain walls particularly of the 180°-type, were used to determine the initial part of the magnetization curve which was found to be linear. The magnetization increased rapidly in such fields. However, the main processes associated with a change of E took place in stronger fields.

We shall denote the magnitude of the almost vertical jump in the Young modulus in Fig. 5 by $\Delta E'/E_0$. The dependences of $\ln(\Delta E'/E_0)$ on $\ln(\sigma/\sigma_{4.2})$, where $\sigma_{4.2}$ is the specific magnetization of a sample at $T = 4.2$ K, plotted for all the temperatures gave a straight line (Fig. 6), from which we deduced the power exponent $n = 2$ for the dependence $\Delta E'/E_0 = A(\sigma/\sigma_{4.2})^n$ which was now applicable throughout the temperature range of the existence of ferromagnetism; we also determined the maximum ΔE effect for this sample (at helium temperature) because of the jump mentioned above: $(\Delta E'/E_0)_{\max} = A = 0.5 = 50\%$.

§ 2. STATISTICAL THEORY OF THE ΔE EFFECT DUE TO DISPLACEMENT OF DOMAIN WALLS

We shall calculate the field-induced change in the Young modulus using a statistical approach similar to that employed in Ref. 4. We shall consider an easy-plane hexagonal ferromagnet in which the difficult axis c is directed along z and the easy-magnetization axis (b) coincides with

x. If the anisotropy in the basal plane is weak compared with the uniaxial anisotropy, then the magnetization switching in this plane leaves the magnetic moment in the plane, i.e., $M_z = 0$. The free energy of the system can then be represented by

$$F=F_0+F_{\mathrm{me}}+F_{\mathrm{e}}, \tag{1}$$

$$\begin{gathered} F_0=-\mathbf{H}\gamma M-K_{66}\,\mathrm{Re}\,(\gamma_y+i\gamma_x)^6, \\ F_{\mathrm{me}}=-B_{66}\left[\varepsilon_{xy}\gamma_x\gamma_y+{}^1/_4(\varepsilon_{xx}-\varepsilon_{yy})(\gamma_x^2-\gamma_y^2)\right], \\ F_{\mathrm{e}}={}^1/_2c_{11}(\varepsilon_{xx}^2+\varepsilon_{yy}^2)+c_{12}\varepsilon_{xx}\varepsilon_{yy} \\ +c_{13}(\varepsilon_{xx}+\varepsilon_{yy})\varepsilon_{zz}+{}^1/_2c_{33}\varepsilon_{zz}^2+{}^1/_2c_{66}\varepsilon_{xy}^2, \end{gathered} \tag{2}$$

where $\gamma = \mathbf{M}/M$; $\gamma_i = M_i/M$ are the direction cosines of the magnetic moment; $\varepsilon_{ij} = 1/2(\partial u_i/\partial x_j + \partial u_j/\partial x_i)$ are the components of the elastic strain tensor ($\mathbf{u}$ is the elastic displacement); c_{ij} are the elastic moduli, where $c_{66} = 2(c_{11} - c_{12})$; B_{66} is the magnetoelastic interaction constant; K_{66} is the basal anisotropy constant.

We shall assume that a magnetic field is applied along the x axis and that a stress σ_{xx} is acting. The equations

$$\partial F/\partial\varepsilon_{ij}=\sigma_{xx}\delta_{ix}\delta_{jx},$$

where δ_{pq} are the Kronecker deltas, allow us to find the equilibrium strains:

$$\begin{gathered} \varepsilon_{xx}=\frac{\sigma_{xx}}{E_s}+\frac{B_{66}}{2c_{66}}(\gamma_x^2-\gamma_y^2), \\ \varepsilon_{yy}=(E_s^{-1}-2c_{66}^{-1})\,\sigma_{xx}-\frac{B_{66}}{2c_{66}}(\gamma_x^2-\gamma_y^2), \\ \varepsilon_{xy}=\frac{B_{66}}{c_{66}}\gamma_x\gamma_y, \\ \varepsilon_{zz}=-\frac{2c_{13}}{c_{33}}(E_s^{-1}-c_{66}^{-1})\,\sigma_{xx}, \end{gathered} \tag{3}$$

where

$$E_s^{-1}=\frac{1}{c_{66}}+\left[2\left(c_{11}+c_{12}-\frac{2c_{13}^2}{c_{33}}\right)\right]^{-1}$$

is the reciprocal of the Young modulus measured along the x axis in the absence of magnetostriction ($B_{66} = 0$).

Equations (1) and (3) yield the thermodynamic potential

$$\Phi=F-\sigma_{xx}\varepsilon_{xx}=F_0-\frac{B_{66}}{2c_{66}}\sigma_{xx}(\gamma_x^2-\gamma_y^2)+F(\sigma_{xx}), \tag{4}$$

where $F(\sigma_{xx})$ is the elastic energy independent of the direction of magnetization γ. Minimizing Eq. (4) with respect to γ_x (bearing in mind that $\gamma_x^2 + \gamma_y^2 = 1$), we can find the equilibrium directions of the magnetic moment in an arbitrary field H. Clearly, in general these directions depend on the magnetic field and on the stress σ_{xx}. We shall allow for this circumstance in analyzing the ΔE effect associated with the reorientation of the magnetic moments in a sample in which the domain walls are pinned. We shall assume here that $B_{66}\sigma_{xx}/c_{66}$ and $HM \ll K_{66}$, so that the equilibrium directions of the magnetization γ_n are practically identical with the six easy-magnetization axes in the basal plane, i.e.,

$$(\gamma_x,\ \gamma_y)_n=(\pm1,\ 0);\quad (\pm{}^1/_2,\ +\sqrt{3}/2);\quad (\pm{}^1/_2,\ -\sqrt{3}/2).$$

According to Eq. (4), the energy of each such state is

$$\Delta\Phi_n=-HM\gamma_{nx}-\frac{B_{66}}{c_{66}}\sigma_{xx}\gamma_{nx}^2.$$

We shall average Eq. (3) for the deformation along the x axis:

$$\bar{\varepsilon}_{xx}=\sum_n\varepsilon_{xx}^{(n)}f_n\Big/\sum_n f_n$$

in the approximation $\sigma_{xx}\to 0$. The weighting function f is in our case (see Ref. 4)

$$f_n=\exp\left(-\frac{\Delta\Phi_n}{\lambda}\right)=\exp\left(\frac{HM}{\lambda}\gamma_{nx}\right)\left[1+\frac{B_{66}}{\lambda c_{66}}\gamma_{nx}^2\sigma_{xx}\right], \tag{5}$$

where λ is a constant quantity which may be associated with other measurable parameters, such as the initial susceptibility of a magnetic material. Since this susceptibility (which is parallel to the easy axis) is given by

$$\chi(0)=\frac{\partial\bar{\gamma}_x}{\partial H}\bigg|_{H=0}=\frac{M^2}{2\lambda}\quad\text{for}\quad\sigma_{xx}=0,$$

we find that $\lambda = M^2/2\chi(0)$. It follows from Eqs. (3) and (5) that

$$\frac{1}{E_H}=\frac{\partial\varepsilon_{xx}}{\partial\sigma_{xx}}\bigg|_{\sigma_{xx}=0}=\frac{1}{E_S}+\left(\frac{2B_{66}}{c_{66}}\right)^2\frac{\overline{\gamma_x^4}-(\overline{\gamma_x^2})^2}{\lambda}\bigg|_{\sigma_{xx}=0}, \tag{6}$$

where E_S is the Young modulus under saturation conditions. Bearing in mind that in $H = 0$ we have $\overline{\gamma_x^2} = \frac{1}{2}$ and $\overline{\gamma_x^4} = \frac{3}{8}$, we obtain the expression for the ΔE effect:

$$\frac{1}{E_0}-\frac{1}{E_H}=\left(\frac{B_{66}}{c_{66}}\right)^2\frac{\chi}{M^2}\left(\mathrm{ch}\frac{2\chi H}{M}-\mathrm{ch}\frac{\chi H}{M}\right)\left(\mathrm{ch}\frac{2\chi H}{M}-4\,\mathrm{ch}\frac{\chi H}{M}\right)\left(\mathrm{ch}\frac{2\chi H}{M}+2\,\mathrm{ch}\frac{\chi H}{M}\right)^{-2}. \tag{7}$$

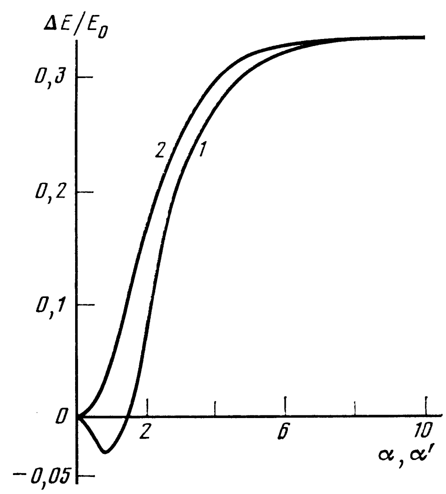

FIG. 7. Dependences of the ΔE effect on $\alpha = \chi H/M$ calculated using Eq. (7) (easy axis, curve 1), and on $\alpha' = \sqrt{3}\chi H/M$ calculated using Eq. (8) (difficult axis in the basal plane, curve 2). The graphs are plotted for the susceptibilities $\chi = 4$ assumed to be the same for the easy and difficult axes.

Curve 1 in Fig. 7 shows the calculated [using Eq. (7)] dependence of the ΔE effect on the quantity $\alpha = \chi H/M$ proportional to the field, where χ is the initial susceptibility along the easy axis. We can see from Fig. 7 and Eq. (7) that the Young modulus first falls on increase in the magnetic field (negative ΔE effect) and then rises monotonically to the value

$$\frac{\Delta E}{E_0 E_S}=\frac{1}{E_0}-\frac{1}{E_S}=\left(\frac{B_{66}}{c_{66}}\right)^2\frac{\chi}{M^2}. \tag{8}$$

A theoretical analysis shows that the average susceptibility $\chi(H)$ does not have a maximum but tends monotonically to zero. Nevertheless, it follows from Eq. (7) that the initial ΔE effect is negative.

A similar calculation carried out for the difficult magnetization axis gives the following expression for the ΔE effect:

$$\frac{1}{E_0}-\frac{1}{E_H}=\left(\frac{B_{66}}{c_{66}}\right)^2\frac{\chi}{M^2}\left(\operatorname{ch}\frac{\sqrt{3}\chi H}{M}-1\right)$$
$$\times\left(4\operatorname{ch}\frac{\sqrt{3}\chi H}{M}-1\right)\left(2\operatorname{ch}\frac{\sqrt{3}\chi H}{M}+1\right)^{-2}, \tag{9}$$

where χ is the initial susceptibility along the difficult magnetization direction in the basal plane (this value is less than for the easy axis). In this case there is no negative ΔE effect and the dependence $E(H)$ is monotonic; the value of $\Delta E/E_0$ rises and the maximum magnitude of the effect is reached on saturation when $H \gg M/\sqrt{3}\chi$. The maximum effect is again given by Eq. (8).

Curve 2 in Fig. 7 represents the theoretical dependence [plotted on the basis of Eq. (9)] of the ΔE effect on the quantity $\alpha' = \sqrt{3}\chi H/M$, proportional to the field; here, χ is the initial susceptibility along the difficult axis. The value of χ is assumed to be 4 and the same for both axes.

It has been suggested in the literature (in Ref. 2 and elsewhere) that a necessary reason for the negative ΔE effect is stabilization, which stops the motion of domain walls for one reason or another. This idea has been confirmed quite convincingly by the experimental results obtained recently by Novikov and Dolgikh.[22] However, in our calculations we have ignored any defects in the material of a sample and when measurements of E are made along the b axis of a crystal there is no anisotropy.

The negative ΔE effect predicted by Eq. (7) can be explained by the fact that during the initial stage of the magnetization there is an increase in the dimensions of all domains (including the non-180° neighborhoods) for which the projection of the magnetization along the field is positive. This may be due to the presence of 60° domain neighborhoods. A further increase in the field reduces the dimensions of all the domains with the magnetizations that are not collinear with the field and the ΔE effect then rises monotonically. Therefore, the negative ΔE effect observed in the present case may be attributed to the slowing down of the growth of metastable phases in which the magnetization is directed at angles of $\pm 60°$ to the field.

We shall now estimate the theoretical value of the maximum ΔE effect for the $Tb_{0.4}Gd_{0.6}$ alloy assuming that $B_{66} = 4.6\times10^8$ erg/cm³, $c_{66} = 9.2\times10^{11}$ dyn/cm², $M = 2\times10^3$ G, and $\chi = 4$ (at 78 K). We find from Eq. (8) that $(E_H - E_0)/E_0 \approx 10^{-1}$, which is in order-of-magnitude agreement with the experimental results. However, the model in question does not account for the negative ΔE effect observed experimentally in our alloy when the field is directed along the difficult axis in the basal plane. Moreover, the model does not allow for the rotation of the magnetization vectors in the domains under the influence of the applied magnetic field.

We shall therefore calculate the change produced in the Young modulus during magnetization, by the deviation of the magnetization from the initial equilibrium position under the influence of elastic stresses. For the sake of simplicity, we shall consider a single-domain sample and assume that the domain walls are completely pinned.

§ 3. ΔE EFFECT DUE TO CHANGES IN EQUILIBRIUM DIRECTIONS OF MAGNETIZATION IN METASTABLE PHASES

The equilibrium directions of the magnetic moment in a crystal can be found from the conditions

$$\partial F/\partial\varepsilon_{ij}=\sigma_{ij},\qquad \partial F/\partial\gamma_i=0,$$

where F is given by Eqs. (1) and (2).

We shall consider the case of the field parallel to the x axis and assume that $\sigma_{ij} = \sigma_{xx}\delta_{ix}\delta_{jx}$. Minimization of the potential (4) gives

$$\frac{\partial F_0}{\partial\gamma_x}-\frac{2B_{66}}{c_{66}}\sigma_{xx}\gamma_x=0. \tag{10}$$

If we assume that the strains are small, so that $(2B_{66}/c_{66})\sigma_{xx} \ll K_{66}$, we obtain an equation describing reorientation of the magnetic moment in the canted phases (characterized by $\gamma_x \neq \pm 1$) when $|H| \leqslant 36K_{66}/M$ (in fields $|H| > 36K_{66}/M$ the magnetization is always oriented along the field, i.e., $\gamma_x = \pm 1$):

$$\frac{HM}{36K_{66}}=\frac{\gamma_x}{3}(4\gamma_x^2-3)(1-4\gamma_x^2). \tag{11}$$

The field dependence of the projection of the magnetization on the field $\gamma_x = M_x/M$ is shown for this case (when the field is parallel to the b axis of the crystal) in Fig. 8a. In the demagnetized state where $H = 0$, we have not only the $\gamma_x = \pm 1$ phases, but also the doubly degenerate canted phases with $\gamma_x = \pm 0.5$ associated with the presence of the easy axes in the basal plane. The metastable canted phases are stable in the range of fields described by Eq. (11), where $\gamma_-^2 < \gamma_x^2 < \gamma_+^2$ and $\gamma_\pm^2 = (6 \pm \sqrt{21})/20$, because we then have $\partial^2F/\partial\gamma_x^2 > 0$. At the points $\gamma_x^2 = \gamma_\pm^2$ (for $\partial^2F/\partial\gamma_x^2 = 0$) the canted phases become unstable.

We shall now consider the changes in the Young modulus due to reorientation of the magnetization in the canted phase. It follows from Eqs. (3) and (10) that

$$\frac{1}{E_H}=\frac{\partial\varepsilon_{xx}}{\partial\sigma_{xx}}=\frac{1}{E_S}+\frac{2B_{66}\gamma_x}{c_{66}}\left.\frac{\partial\gamma_x}{\partial\sigma_{xx}}\right|_{\sigma_{xx}=0}$$
$$=\frac{1}{E_S}+\frac{4B_{66}^2}{c_{66}^2}\frac{\gamma_x^2}{F_{0\gamma_x\gamma_x}}. \tag{12}$$

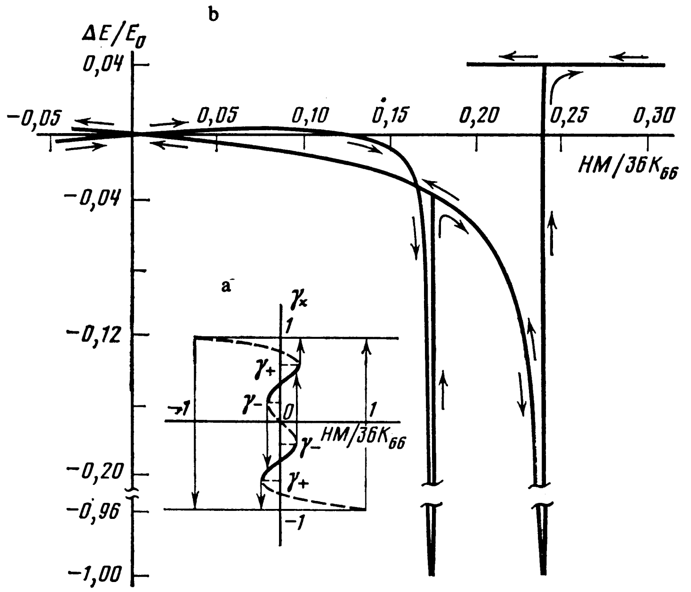

FIG. 8. a) Dependence of the projections of the magnetization $\gamma_x = M_x/M$ along the field direction on the value of $HM/36K_{66}$, proportional to the intensity of the magetic field applied along the easy axis of a single-domain sample with an initial direction of the magnetization along one of the six easy axes. b) Dependence of the ΔE effect on $HM/36K_{66}$ for the same case (calculation carried out for $\beta = 8\times10^{-3}$).

We shall assume that a sample is in a single-domain state and that $\gamma_x = \gamma_0$ in $H = 0$. Then, Eqs. (12) and (2) yield the following expression for the observed ΔE effect:

$$\frac{1}{E_0} - \frac{1}{E_H} = \frac{B_{66}^2}{240 c_{66}^2 K_{66}} f(\gamma_x), \tag{13}$$

$$f(\gamma_x) = \frac{(\gamma_x^2-\gamma_0^2)(\gamma_-^2\gamma_+^2-\gamma_0^2\gamma_x^2)}{(\gamma_x^2-\gamma_-^2)(\gamma_+^2-\gamma_x^2)(\gamma_0^2-\gamma_-^2)(\gamma_+^2-\gamma_0^2)}. \tag{14}$$

Since in the collinear phase we have $E = E_S$ (i.e., the magnetoelastic term is then missing from the expression for the Young modulus), it follows that the transition from the canted to the collinear phase is accompanied by an additional positive jump in the ΔE effect amounting to

$$\frac{1}{E_0} - \frac{1}{E_H} = \frac{B_{66}^2}{48 c_{66}^2 K_{66}}. \tag{15}$$

Since in $H = 0$ we have in this case $\gamma_0^2 = 0.25$, it follows from Eqs. (13) and (14) that the ΔE effect in a canted phase is always negative for $\gamma_0^2 < \gamma_x^2 < \gamma_+^2$ and it rises on approach to the point of loss of the phase stability $\gamma_x \to \gamma_+$, where $\partial^2 F/\partial\gamma_x^2 \to 0$. At this point the value of E_H tends to zero. In the range $\gamma_-^2 < \gamma_x^2 < \gamma_0^2$ there is an interval of positive values of the ΔE effect, but close to $\gamma_x^2 \approx \gamma_-^2$ it is negative, whereas for $\gamma_x^2 \to \gamma_-^2$ as well as for $\gamma_x^2 \to \gamma_+^2$, we have $E_H \to 0$. At the loss of stability point the magnetic system becomes very strongly "demagnetized" so that its differential magnetic susceptibility $\chi_{\text{diff}} \sim (\partial^2 F/\partial\gamma_x^2)^{-1}$ tends to infinity. Then, the expansion of the free energy in terms of the order parameter must include terms higher than quadratic. The action of elastic stresses gives rise to a strong modification of the magnetic structure and the dependence of the strain on the elastic stress has an additional nonlinear correction $\varepsilon_{xx} \approx \sigma_{xx}^{1/2}$. Consequently, we find that

$$(\partial\varepsilon_{xx}/\partial\sigma_{xx})^{-1} = E_H|_{\sigma_{xx}\to 0} \to 0.$$

This effect is due to the fact that when an allowance is made for the magnetoelastic interaction the soft mode of this transition is a quasielastic wave[7] which is responsible for the vanishing of the Young modulus.

The transition from a canted to a collinear phase results in an abrupt change in the ΔE effect to a positive value given by Eq. (15). Figure 8b shows the dependence of the ΔE effect on the magnetic field plotted using the formula

$$\frac{E_H - E_0}{E_0} = \frac{\beta E_0 f(\gamma_x)}{1 - \beta E_0 f(\gamma_x)}, \tag{16}$$

where $\beta = B_{66}E_0/240 c_{66}^2 K_{66}$. This dependence is plotted for $\beta \approx 8\times10^{-3}$ on the assumption that $K_{66} = 3.5\times10^5$ erg/cm^2 (Ref. 12), which corresponds to the parameters of the alloy $Tb_{0.4}Gd_{0.6}$ at $T = 78$ K. The magnitude of the positive ΔE effect due to rotation of the magnetization under the action of the field and elastic stresses is now

$$(E_H - E_0)/E_0 = B_{66}^2 E_0/48 c_{66}^2 K_{66} \approx 10^{-2}.$$

Therefore, because of the relatively strong basal anisotropy of these alloys, the ΔE effect due to the mechanism described above is small, apart however from a narrow range of fields near the lines of loss of stability by a canted phase. In such regions the ΔE effect is very large and negative.

A similar calculation for a field parallel to the y axis (which is difficult in the basal plane), gives the following expression for the change in the Young modulus along this axis:

$$\frac{1}{E_H} = \frac{1}{E_S} + \frac{4B_{66}^2}{c_{66}} \frac{\gamma_y^2}{F_{\gamma_y\gamma_y}} = \frac{1}{E_S} + \frac{B_{66}\gamma_y^2}{240 c_{66}^2 K_{66}(\gamma_-^2-\gamma_y^2)(\gamma_+^2-\gamma_y^2)}. \tag{17}$$

The observed change in the Young modulus is given by the formula

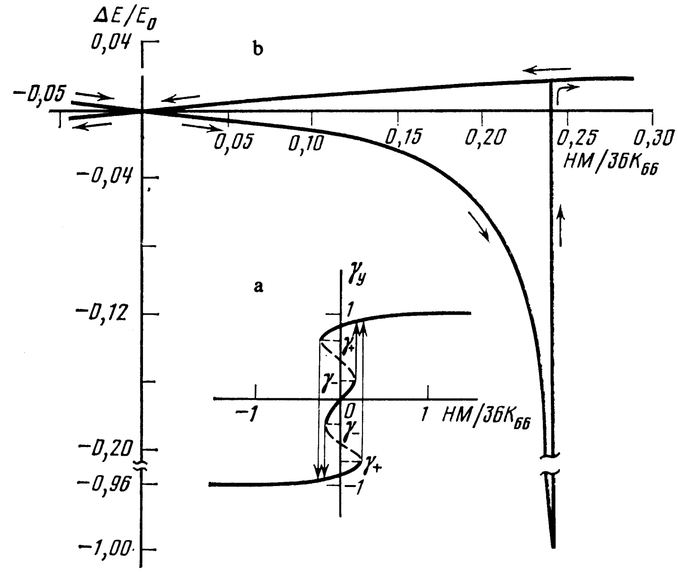

FIG. 9. Dependence of the projection of the magnetization $\gamma_y = M_y/M$ along the direction of the field on the value of $HM/36K_{66}$ proportional to the field applied along a difficult axis in the basal plane of a single-domain sample with an initial direction of the magnetization along one of the six easy axes. b) Dependence of the ΔE effect on $HM/36K_{66}$ in the same case (calculated for $\beta = 8\times10^{-3}$, $\gamma_y|_{H=0} = \sqrt{3}/2$).

$$\frac{1}{E_0}-\frac{1}{E_H}=\frac{B_{66}{}^2 f(\gamma_y)}{240c_{66}{}^2K_{66}}$$

$$=\frac{B_{66}{}^2}{240c_{66}{}^2K_{66}}\frac{(\gamma_y{}^2-\gamma_0{}^2)(\gamma_0{}^2\gamma_y{}^2-\gamma_+{}^2\gamma_-{}^2)}{(\gamma_0{}^2-\gamma_-{}^2)(\gamma_0{}^2-\gamma_+{}^2)(\gamma_y{}^2-\gamma_-{}^2)(\gamma_y{}^2-\gamma_+{}^2)},\quad(18)$$

where $\gamma_0 = \gamma_y|_{H=0}$ is the equilibrium direction of the magnetization in $H = 0$.

The six states of equilibrium are then described by

$$\gamma_y=0,\quad \gamma_y=\pm\sqrt{3}/2.$$

The equilibrium direction of the magnetization in a magnetic field $\gamma_y(H)$ is then given by the equation

$$HM/36K_{66}={}^1/_3\gamma_y(4\gamma_y{}^2-3)(4\gamma_y{}^2-1).\quad(19)$$

A solution of this equation yields a "hysteresis loop" $\gamma_y(H)$ shown in Fig. 9a.

Stable canted phases exist in a range of fields defined by the conditions $\gamma_y^2<\gamma_-^2$ and $\gamma_y^2>\gamma_+^2$, because in these intervals we have $\partial^2F_0/\partial\gamma_y^2>0$. The field dependence of the ΔE effect is given by Eqs. (18) and (19).

Figure 9b shows the dependence of the ΔE effect on the quantity $HM/36K_{66}$, which is proportional to the field intensity; the dependence is plotted for the case of $\gamma_0=\sqrt{3}/2$ using the formula

$$\frac{E_H-E_0}{E_0}=\frac{\beta E_0 f(\gamma_y)}{1-\beta E_0 f(\gamma_y)},\quad(20)$$

where $\beta\approx8\times10^{-3}$. On approach to the loss of stability point $\gamma_y^2\to\gamma_+^2$ the negative ΔE effect becomes much stronger and then it changes abruptly to a positive value because of a transition to a stable canted phase characterized by $\gamma_y^2>\gamma_+^2$. This is followed by a smooth rise of the ΔE effect to a positive value described by

$$\frac{E_H-E_0}{E_0}=\frac{B_{66}{}^2E_0}{85c_{66}{}^2K_{66}}\left[1-\frac{B_{66}{}^2E_0}{85c_{66}{}^2K_{66}}\right]^{-1}.\quad(21)$$

If the demagnetized state of a sample corresponds to the $\gamma_y=0$ phase, the ΔE effect is always negative, as demonstrated by an analysis of Eqs. (18)–(20), and it rises in the absolute sense on approach to the loss of stability line of this phase when $\gamma_y\to\gamma_-$. In the demagnetized state a sample contains domains corresponding to all six phases. In the case of complete pinning of domain walls the total ΔE effect is found by averaging Eqs. (13) and (18) over the phase states of the system.

§ 4. DISCUSSION OF RESULTS AND CONCLUSIONS

It follows from § 3 that the presence of metastable canted phases may give rise to a negative ΔE effect when a sample is magnetized along easy or difficult directions in the basal plane.

The hysteresis of $\Delta E/E_0$ illustrated in Figs. 8b and 9b near $H=0$ may be manifested during magnetization of a sample even when domain walls are not pinned because canted phases are present in a real sample subjected to weak fields. The experimental dependences of $\Delta E/E_0$ on H (Fig. 3) show clearly such a hysteresis. It cannot be explained satisfactorily by the model of displacement of domain walls. In stronger fields the abrupt changes in $\Delta E/E_0$ shown in Figs. 8b and 9b are clearly masked by the processes of domain wall displacement. These processes reduce the contribution, discussed in § 3, of metastable canted phases to the negative ΔE effect.

If the transition to a stable canted phase, occurring by domain wall displacement, terminates in fields that do not reach the loss of stability lines (when $36\chi K_{66}/M^2\ll1$), an abrupt change in E cannot be observed and the main contribution to $\Delta E/E_0$ is due to the first of the mechanisms discussed above.

However, if a sample contains defects etc. which can pin domain walls, the changes in the Young modulus due to the loss of stability by metastable phases result in a rapid rise of the negative ΔE effect in the appropriate range of fields. The inhomogeneity of the magnetostatic demagnetization field in metastable phase domains results in "smearing" of the abrupt changes in E, so that even in this range of fields they are smoother.

When the energy of the basal anisotropy and of the demagnetization fields are comparable, i.e., when $36\chi K_{66}/M^2 \approx 1$, the processes of displacement of domain walls and reorientation of the magnetizations in the domains on increase in the fields are superimposed and they cannot be separated. In this case both mechanisms contribute to the ΔE effect. Such a situation clearly occurs also in the crystal of the $Tb_{0.4}Gd_{0.6}$ alloy. However, far from the points of loss of stability by canted phases the dominant contribution to the ΔE effect is made in this crystal by the processes of domain wall displacement. We cannot exclude the possibility that the large magnitude of the negative ΔE effect observed in this alloy at low temperatures is due to the presence of defects mentioned above, which can result in partial pinning of domain walls during the initial stage of the magnetization of a sample $[H \ll M/\chi(0)]$. However, it should be noted that the large value of the maximum negative ΔE effect observed along the easy and difficult directions for the Tb–Gd alloy can hardly be explained fully by the two mechanisms under discussion: estimates obtained using the model of wall displacement give much lower values of the ΔE effect and, according to the second model, this effect exists only in a very narrow range of fields. It is necessary to analyze in greater detail the influence of defects on the pinning of walls and on the internal friction in the investigated samples. The questions mentioned here cannot be answered without a more detailed study of the magnetization of a sample during the initial stage of the rise of the field and, moreover, it is necessary to obtain information on the dynamics of the domain structure during the magnetization of a sample in the region where metastability is lost.

We shall now draw some conclusions. The above theoretical analysis shows that the model of displacements of domain walls in a hexagonal magnetic material with the easy-plane anisotropy during the magnetization of a sample along the easy direction gives rise to a negative ΔE effect which is not associated with the existence of a susceptibility maximum in a finite magnetic field. If there are no defects in a sample, a calculation of this kind does not predict a negative ΔE effect when a sample is magnetized along a difficult direction in its basal plane.

The magnetoelastic contribution to the Young modulus associated with the deviation of the magnetization from the equilibrium direction under the action of elastic stresses gives rise to a hysteresis in the presence of metastable phases in a sample and to abrupt changes in the Young modulus in the region where these phases become unstable.

These theoretical models account qualitatively for the field dependence of the ΔE effect in the investigated Tb–Gd alloy, but the first of these models underestimates the maximum negative ΔE effect in the alloy because of failure to allow for the presence of defects which can pin domain walls in a sample and thus retard the magnetization in relatively weak fields. Crystal defects may also be responsible for the very existence of metastable domain phases in a sample in relatively high fields.

The second model allows for metastable phases and predicts a negative ΔE effect without invoking displacement and pinning of domain walls, but the effect occurs only within narrow ranges of magnetic fields.

The experimentally obtained dependence of the ΔE effect of $Tb_{0.4}Gd_{0.6}$ on the square of its magnetization have revealed the influence of various processes during magnetization of an alloy single crystal on the ΔE effect. The temperature dependence of the jump of the ΔE effect in the investigated sample is also described well by a quadratic dependence on the magnetization. The temperature dependence of the ΔE effect has not been considered theoretically in the present paper.

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