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Magneto-phonon contribution into the Young's modulus of gadolinium

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Abstract. Theoretical and experimental investigation has been made of the magnetic contribution to the Young's modulus of rare earth ferromagnet gadolinium. Experimental study includes measurements of the Young's modulus as a function of temperature, magnetic field and magnetization of gadolinium. Theoretical analysis is based on the account of phonon anharmonicity which gives rise to the dependence of Debye temperature on magnetization. Spontaneous magnetic contribution to the Young's modulus of Gd is found to be proportional to the squared magnetization of the metal. The magnetic contribution is also induced by magnetizing magnet due to the paraprocess.

PACS. 62.20.Dc Elasticity, elastic constants – 62.20.+x Mechanical properties of solids

Introduction

Magnetic phase transitions in Rare Earths Metals (REM) and their alloys as well as an influence of the transitions on various physical properties of REM has received the valuable attention of many investigators [1–5].

One effective method to study magnetic phase transitions is the exploration of elastic properties [3,5]. Measurements of elastic characteristics allow reliably to fix such weak anomalies as those in transitions between commensurate phases of magnetic structures of REM [4,5]. Systematic study of anomalous temperature dependencies of elastic moduli of REM was started by Belov *et al.* [6,7]. They established a correlation between the elastic moduli and magnetization of Ferromagnetic REM (FREM). They considered also an effect of external magnetic field on the elastic anomalies. The effect was found to be connected with a change of magnet magnetization (ΔE-effect). An attempted theoretical description of elastic moduli anomalies near Curie point T_C was made in references [6,7] on the basis of a second order theory of thermodynamic magnetic phase transitions.

However, the developed thermodynamical approach [6,7] doesn't consider phonon anharmonicity which should take place not only in the paramagnetic region of FREM but also below T_C. The anharmonicity account is a principal point because the anharmonicity of atom oscillations in a crystalline lattice gives rise to the temperature dependence of the Young's modulus in the paramagnetic region [8]. In addition the fact should be taken into account that the term in the thermodynamic potential (ThDP) connected with phonon anharmonicity includes a characteristic Debye temperature θ which is known to depend on magnetization in a number of itinerant ferromagnets [9–11]. The reason for such dependence can be seen from the dependence of Debye temperature on bulk modulus which in its turn depends on magnetization [10,11].

Till now, there has been no clarification of the deep reasons responsible for the Young's modulus' dependence on temperature in the paramagnetic region of ferromagnets. The technique isn't developed yet for separation of the magnetic contribution from the total value of the Young's modulus. This makes it impossible to consider adequately the dependence of the magnetic contribution to the modulus on magnetization, magnetic field and temperature. For the reasons above further theoretical and experimental investigations are necessary to clear up the nature of elastic moduli anomalies in FREM.

In the present work an expression of for Young's modulus of rare earth ferromagnets was obtained on the basis of theoretical consideration of a solid body as a quantum ensemble of anharmonic oscillators [8], and magnetic phase transition theory of second order [12]. The principle point of the developed approach is dependence of Debye temperature on magnetization, ferromagnetic REM gadolinium was taken as a model for which elastic properties are considered in the framework of the present theory.

Theory

Earlier [8] for a nonmagnetic isotropic solid body with an account of thermal anharmonicity of atomic oscillations

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(the linear chain approach was used), an expression was obtained for the temperature dependence of the Young's modulus $E_p(T)$, more exactly of its lattice part:

$$E_p(T) = E_0[1 - KTD(T/\theta)], \quad (1)$$

where $E_0 = E_p(T = 0)$ is the low temperature limit of the modulus; $K = 9\beta_\infty(Lc_1/c)$ is a coefficient directly proportional to the ratio of anharmonic c_1 and harmonic c constants of restoring oscillator force and thus proportional to the degree of anharmonicity in the atomic oscillations; β_∞ is the high temperature asymptotic value of the thermal expansion coefficient in the Debye model; L is a dimensional parameter; θ is a characteristic model parameter which has the sense of Debye temperature for the oscillators chain. In a general case, parameter θ (for brevity, hereinafter Debye temperature) can be different from the real Debye temperature of matter. Tabulated Debye function $D(x)$ depends only on the relative temperature $x = T/\theta$:

$$D(x) = 3x^3 \int_0^{1/x} \frac{y^3 dy}{e^y - 1} \cdot \quad (2)$$

Thus in the limits of the Debye model the opportunity exists to connect immediately two independently measured sets of physical properties of solids: the elastic and thermodynamical ones [13].

"Magnetic" phonon anharmonicity gives rise to a change of Debye temperature with magnetization. One can state that the magnetically dependent part of Debye temperature near Curie point T_C can be expanded into a row with even powers of magnetization I:

$$\theta = \theta_0 + \theta_1(I) = \theta_0 + \frac{1}{2}\alpha_\theta I^2 + \frac{1}{4}\beta_\theta I^4 + \dots , \quad (3)$$

where it is supposed $\theta_0 = Const$ for a given magnet, and the magnetic correction to Debye temperature is small: $\theta_1(I) \ll \theta_0$. Thermodynamical reasons for equation (3) are the same as in the thermodynamical theory of Landau [12] where ThDP near the Curie point is presented as a row with even powers of magnetization. As mentioned above the similar row for Debye temperature was obtained earlier for a number of band ferromagnets [10,11] in the limits of the Stoner model. We should note that critical behavior of magnetic properties in the nearest vicinity of the Curie point can lead to deviation from that presented (3). The developing approach thus corresponds to a regular (non critical) behavior of magneto elastic properties. According to experiment [9] Debye temperature of FREM increases in the ferromagnetic region ($a_\theta > 0$) but slower than the squared magnetization ($\beta_\theta < 0$).

Expanding Debye function $D(x)$ into a row near θ_0 by small parameter θ_1 and taking into account only the three first terms we'll get the Young's modulus of FREM ($x_0 = T/\theta_0$):

$$\begin{aligned} E(x) &\approx E_p(x_0) + E_m(x_0, I) \\ &\approx E_p(x_0) + E_m^{(1)}(x_0, I^2) + E_m^{(2)}(x_0, I^4), \end{aligned} \quad (4)$$

where the lattice (phonon) part of the modulus is

$$E_p(x_0) = E_0[1 - KTD(x_0)]. \quad (5)$$

Equation (5) coincides with the result of reference [8] for non magnetic materials. E_0 in this case is the Young's modulus extrapolated to $T = 0$ from the paramagnetic region. The term dependent on magnetization $E_m(x_0, I)$ in (4) can be interpreted as a magnetic contribution to the Young's modulus' total value. The first magnetic correction in the modulus

$$E_m^{(1)}(x_0, I^2) = E_0 K \alpha_\theta Y_0(x_0) \frac{1}{2} I^2, \quad (6)$$

is proportional to the squared magnetization and vanishes in the Curie point. The second magnetic correction in the modulus is

$$E_m^{(2)}(x_0, I^4) = E_0 K x_0 \left\{ \beta_0 Y_0(x_0) - \frac{1}{2}\frac{\alpha_\theta^2}{\theta_0} Y_1(x_0) \right\} \frac{1}{4} I^4. \quad (7)$$

In equations (6, 7) notations are made:

$$Y_0(x_0) = x_0 \left[\frac{C_v(x_0)}{3R} - D(x_0) \right], \quad (8)$$

$$Y_1(x_0) = x_0 \left[\frac{C_v(x_0) + x_0 C_v'(x_0)}{3R} - (D(x_0) + x_0 D'(x_0)) \right], \quad (9)$$

where $C_v(x_0)$ is conventional Debye heat capacity at constant volume; R is gaseous constant; prime "$'$" means differentiation by argument.

Analysis of equations (6, 7) shows that two cases should be differentiated. The first is a case of high temperature ferromagnets ($T_C \geq \theta_0$) and the second one is the case of low temperature ferromagnets ($T_C \ll \theta_0$). Taking into account that in considering FREM Debye temperatures $\theta_0 \sim 100 \div 300$ K, pure FREM and their alloys can be regarded as high temperature ferromagnets. Examples of low temperature ferromagnets are diluted alloys of FREM with non magnetic metals. Exploring the asymptotic behavior of functions $Y_0(x_0)$ and $Y_1(x_0)$ it is easy to show that magneto elastic effects should be most clear in high temperature FREM, which are the ones considered in the present work. In this case we have near the Curie point ($x_0 \geq 1$):

$$E_m^{(1)}(x_0, I^2) \approx E_0 K \alpha_\theta \left[\frac{3}{8} - \frac{1}{10x_0} \right] \frac{1}{2} I^2, \quad (10)$$

$$E_m^{(2)}(x_0, I^4) \approx E_0 K \left\{ \beta_\theta \left[\frac{3}{8} - \frac{1}{10x_0} \right] - \frac{\alpha_\theta^2}{2\theta_0} \frac{1}{10x_0} \right\} \frac{I^4}{4} \cdot \quad (11)$$

Consequently, in the ferromagnetic region ($T < T_C$) the Young's modulus of high temperature ferromagnets will increase proportionally to the squared magnetization. The slope of the linear part of $E_m(I^2)$ dependence will increase going deep into the ferromagnetic region ($E_m^{(1)} > 0$). At high enough magnetization and low temperature, deviation from the linear will take place: dependence of $E_m(I^2)$ will become more gently sloping ($E_m^{(2)} < 0$).

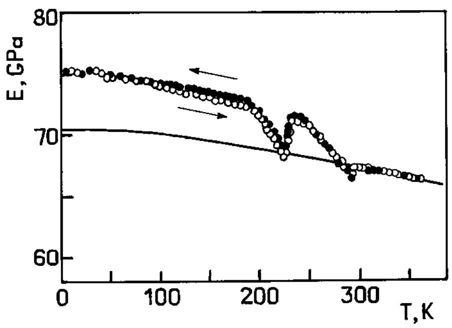

Fig. 1. Temperature dependence of the Young's modulus $E(T)$ of gadolinium single crystal along c axis. Solid line is computed curve of temperature dependence of lattice contribution $E_p(T)$ into the Young's modulus.

Table 1. Temperatures of magnetic phase transitions of gadolinium single crystals and polycrystalline sample according to the Young's modulus measurements.

Transition temperature	Axis	Heating	Cooling
T_C, K	polycrystalline	293.2	293.9
	c	292.1	292
	a	~ 300	~ 300
T_{SR}, K	polycrystalline	227.4	228.4
	c	225.2	225.5
	a	~ 240	~ 240

Experiment

For comparison of theoretical results with experimental data for FREM, measurements were carried out of temperature and magnetic field dependencies of the Young's modulus of gadolinium single crystals taken as a model object. Neutronographic investigations [14] showed that Gd is a weakly anisotropic ferromagnet below Curie temperature $T_C = 293$ K, with magnetic moments aligned along hexagonal axis c. Below the spin reorientation point $T_{SR} \approx 230$ K, magnetic moments deviate from c direction by a temperature dependent angle. The angle is determined by the anisotropy constants of Gd [15]. So, gadolinium is almost an ideal model ferromagnet for comparison between thermodynamic calculations and experimental data.

Measurements of the Young's modulus were made in a sound frequency range (1–3 kHz) for single crystals (c and a directions) and polycrystalline sample of gadolinium by the method of bending vibrations of the sample – a thin bar supported as a cantilever. Temperature region covered is $4.2 \div 370$ K, magnetic field reached 14 kOe. The measurement technique was described earlier in detail [5,13,16].

Temperature dependence of the Young's modulus of single crystal gadolinium in c axis direction is presented in Figure 1. Magnetic phase transition paramagnetism – ferromagnetism (PM-FM) at $T_C \approx 292$ K and spin reorientation transition (SR) at $T_{SR} \approx 225$ K are accompanied by sharp minima in $E(T)$ dependence. The points of magnetic transitions were established by the minima of $E(T)$ curve and are given in Table 1. Established in this way, temperatures T_C and T_{SR} are in good agreement with literature data [3,7,9,14]. The solid line in Figure 1 gives computed temperature dependence of lattice contribution to the modulus. The lattice contribution curve was extrapolated from the PM region of the metal to the low temperature range. Parameters of the computed curve are given in Table 2. The technique of experimental data processing on the basis of equation (6) in the paramagnetic region was discussed earlier in detail [5,13]. Weak temperature hysteresis in $E(T)$ dependence exists in the SR transition range. The hysteresis bears witness to metastable, non equilibrium behavior of magnetic structure of Gd at these temperatures.

Table 2. Parameters of computed temperature dependencies of the lattice contributions into the Young's modulus of gadolinium.

Axis	E_0, GPa	θ_0, K	$K\theta_0$	Lc_1/c
polycrystalline	68.4	184	0.076	4.76
c	70.5	339	0.082	2.07

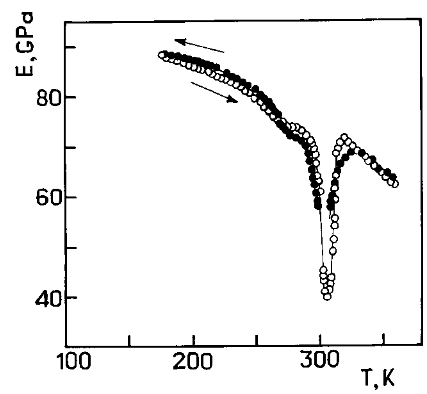

Fig. 2. Temperature dependence of the Young's modulus $E(T)$ of gadolinium single crystal along a axis.

Temperature dependence of the Young's modulus of Gd single crystal along axis a is shown in Figure 2. As well as in the c axis measurements, magnetic phase transition PM-FM is accompanied by a sharp minimum in $E(T)$ dependence. Contrary to the c axis case, SR transition along a direction is expressed weakly. Magnetic phase transition temperatures found in a axis study exceed the ones for c axis, and are less reliable (Tab. 1). Dependence $E(T)$ in a direction demonstrates visible temperature hysteresis in all temperature regions studied. Temperature dependence of the Young's modulus of polycrystalline gadolinium is much smoothen in comparison with single crystals. In general, the elastic modulus of polycrystalline sample behaves in a similar manner in the c direction. By the reasons above, only c axis data are analyzed below for gadolinium.

Temperature dependence of spontaneous (in absence of external magnetic field) magnetic contribution into the

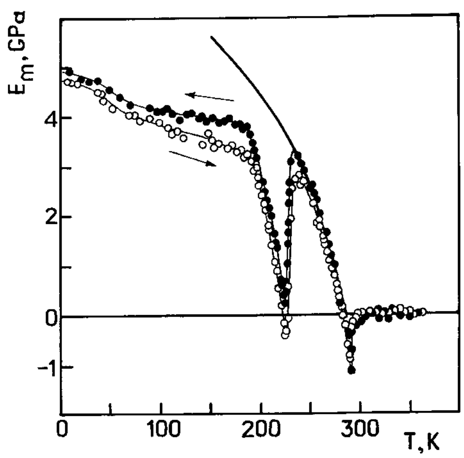

Fig. 3. Temperature dependence of spontaneous magnetic contribution into the Young's modulus $E_m(T)$ of gadolinium single crystal along c axis. Solid line is theoretical curve of the $E_m(T)$ dependence near Curie point.

total value of the Young's modulus $E_m(T)$ of gadolinium c axis single crystal are given in Figure 3. The magnetic contribution was calculated as a difference between experimental values $E(T)$ and computed lattice part $E_p(T)$. The magnetic contribution is positive overall the magnetically ordered temperature range except in the narrow vicinities of phase transition points. The magnetic contribution disappears not immediately in the Curie point but visibly far into the paramagnetic region (about 310 K). This bears witness to some remaining short range magnetic ordering above T_C. This fact should be taken into account if exact calculations are necessary. Sharp minima in $E_m(T)$ dependence at transition points could be associated with critical magneto elastic effects. However, the question demands separate consideration.

Results of the ΔE-effect measurements of gadolinium c axis sample (magnetic field was parallel to the direction in study) shows that except in the vicinities of magnetic transition points, the Young's modulus increases with temperature drop (positive ΔE-effect). The increase rate increases with ascending magnetic field. The Curie point minima in $E(T,H)$ dependencies move to higher temperatures, and decrease in value with the field, so that at $H > 3$ kOe the PM-FM transition is accompanied by only bending of the $E(T,H)$ curves. On the contrary, spin-reorientation minima of $E(T,H)$ dependencies move with the field to lower temperatures and transform into plateaus at $H > 1.5$ kOe. Measured magnetization curves of gadolinium along axis c are in good agreement with literature data [17] and don't need consideration.

Discussion

Dependence of the spontaneous magnetic contribution to the Young's modulus of gadolinium along axis c is shown as a function of squared spontaneous magnetization σ_s^2 in Figure 4. It should be mentioned that specific magnetization σ is connected with a total magnetization I

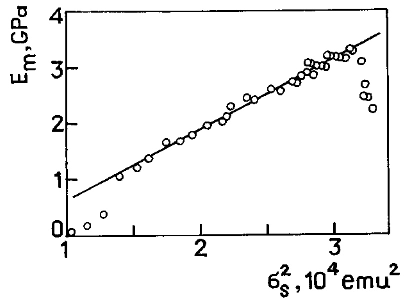

Fig. 4. Dependence of spontaneous magnetic contribution in to the Young's modulus E_m of gadolinium single crystal along c axis on the squared specific magnetization σ_s^2 (1 emu = 1 Gs cm^{3}g^{-1}). Solid line is computed straight line $E_m(\sigma_s^2)$.

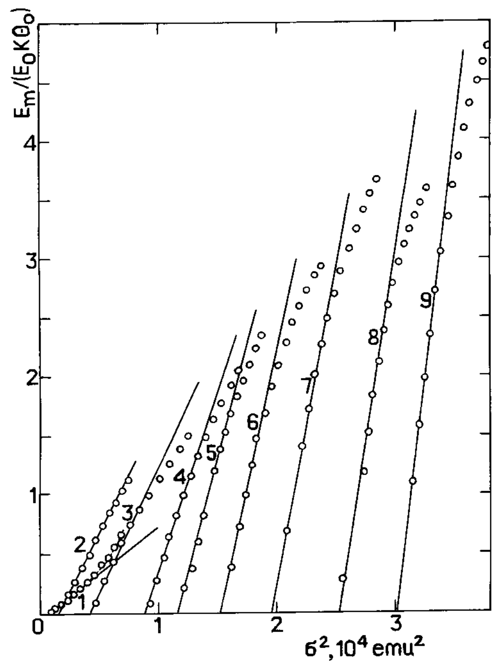

Fig. 5. Dependencies of the normalized magnetic contribution to the Young's modulus $E_m/(E_0K\theta_0)$ of gadolinium single crystal along c axis on the squared specific magnetization σ^2 (1 emu = 1 Gs cm^3 g^{-1}) at fixed temperatures. Curve 1:T = 300 K; 2:297; 3:289; 4:284; 5:278; 6:271; 7:261; 8:251; 9:233.

by the relation $I = \sigma\rho$ where ρ is density. According to theoretical consideration, the dependence $E_m(\sigma_s^2)$ is linear over almost all the ferromagnetic temperature region. This means weak temperature dependence of coefficients α_θ, β_θ in a virgin state of the metal (without field). Deviations from the linear law in the regions of small and big magnetization are associated with critical behavior of the elastic modulus near $T_C \simeq T_{SR}$, accordingly. Computed dependence of the spontaneous magnetic contribution to the Young's modulus of Gd along axis c is also shown in Figure 3 near PM-FM transition point (solid line).

Dependencies of the normalized magnetic contribution to the Young's modulus $E_m/(E_0K\theta_0)$ of Gd along axis c on squared magnetization σ^2 at fixed temperatures

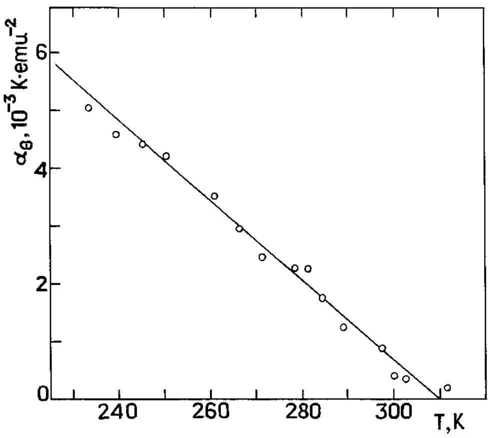

Fig. 6. Temperature dependence of thermodynamic coefficient α_θ. Solid line is computed straight line $\alpha_\theta(T)$.

are given in Figure 5. The modulus magnetic contribution was computed from the ΔE-effect measurements. In agreement with the theory, developed isotherms $E_m(\sigma^2)/(E_0 K\theta_0)$ are linear over initial parts and their slopes increase with temperature drop. With increasing magnetization the isotherms become more gently sloping, so that at high enough magnetization one can see the isotherms $E_m/(E_0 K\theta_0)$ move to saturation.

Coefficient α_θ can be easily found from the dependencies of Figure 5. As follows from equations (6, 10) slope of isotherms $E_m(\sigma^2)/(E_0 K\theta_0)$ over the initial parts are proportional to the value α_θ. Dependence $\alpha_\theta(T)$ is pictured in Figure 6. The experimental points lie satisfactorily on a straight line. This fact permits the suggestion of the following temperature dependence for the thermodynamical coefficients in equation (3) at $T < T_C$:

$$\alpha_\theta = a_0 + a_\theta(T_C - T), \ \beta_\theta = Const. \tag{12}$$

One can show that (12) is in consistent with conventional Landau second order magnetic phase transitions theory [12]. The fact that coefficient a_0 isn't equal to zero can be interpreted as evidence of remaining short range magnetic order somewhat above the Curie point. Values of the coefficients were found to be $a_\theta = 1.18 \times 10^{-3}$ K emu^{-2} and $a_0 = 6.92 \times 10^{-5}$ emu^{-2}. Coefficient β_θ equals zero within the limits of errors.

Conclusion

So the present theoretical investigation has established the spontaneous magnetic contribution to the Young's modulus of ferromagnetic REM to be proportional to the squared spontaneous magnetization. In addition, the magnetic contribution is induced by magnetizing because of a para process. Temperature dependence of the elastic modulus is determined by the phonon anharmonicity. The last one in the ferromagnetic area results in the dependence of the Debye temperature on magnetization. Reciprocal influence of phonon anharmonicity on temperature dependence of magnetization is also possible. This influence, as well as exploration of interrelation between magneto elasticity and other physical properties of matter such as heat capacity, thermal expansion, *etc.*, requires separate consideration. One sees that the magnetostriction effects studied here which form the elastic modulus anomalies, will also take place in more complicated magnetic structures of heavy REM such as antiferromagnetic, helicoidal, spin-slip ones.

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