Effect of Hydrostatic Pressure on the Elastic and Inelastic Behavior of Gadolinium in the Spin-Reorientation Region

H. Klimker and M. Rosen
Nuclear Research Centre, Negev, Beer-Sheva, Israel
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The effect of hydrostatic pressures, up to 6 kbar, on the anomalies occurring in the ultrasonic wave propagation in gadolinium single crystals below the Curie point, is investigated. The elastic softening of the c_{33} elastic constants, and its pressure dependence, is discussed in terms of the molecular-field model of the magnetic-interaction Hamiltonian. It was shown that the elastic softening in gadolinium is of a magnetoelastic origin. Within the framework of the model presented in this study, it is shown why the shear elastic modes c_{44} and c_{66} do not exhibit anomalous behavior in the spin-reorientation region of gadolinium. The reason for the negligible elastic anomaly in c_{11}, in this temperature region, is also discussed. The pressure dependence of the spin-reorientation temperature T_f can be explained in terms of the pressure contribution to the first anisotropy-energy constant K_1. This calculated value of the shift in T_f with pressure is in satisfactory agreement with the experimentally determined one.

I. INTRODUCTION

The behavior of the elastic properties and ultrasonic attenuation of gadolinium in single-crystal and polycrystalline form has been investigated extensively in the vicinity of the ferromagnetic transition point (293 °K) and the spin-reorientation region[1-9] below 220 °K. Gadolinium is considered to be a "normal" ferromagnet[10] below its Curie temperature. The easy direction of magnetization lies along the hexagonal c axis down to about 220 °K, or somewhat higher depending on the sample purity.[6,8] At lower temperatures, spins begin to deviate from the c axis, thus forming a cone of easy directions of magnetization, the angle of which is temperature dependent. The temperature variation of the cone angle was determined by neutron-diffraction[11,12] and torque measurements.[13,14]

The anisotropy energy in gadolinium is considered to be smaller than the isotropic exchange energy by about two orders of magnitude,[15] 10^6 erg cm^{-3} compared with 10^8 erg cm^{-3}. The anisotropy constants in the magnetically ordered state of gadolinium have been experimentally determined,[13,14,16] and the easy direction of magnetization has been established. Below the Curie point the anisotropy constant K_1 is positive, coinciding with "normal" ferromagnetism in which the spins are aligned parallel to the hexagonal c axis. At temperatures below about 220 °K, in the spin-reorientation region, K_1 changes sign and the terms containing the anisotropy constants K_2 and K_3 describe the conical structure. The basal anisotropy was found to be very weak,[17] so that the directions lying on the cone are equivalent.

The magnetoelastic energy in gadolinium is of the same order of magnitude as the anisotropy. The magnetoelastic effect is displayed in the temperature variation of the lattice parameters[18] in the region between 200 and 300 °K of gadolinium. The coefficient of thermal expansion in the c direction is large and negative, whereas in the a direction it is small and positive. Drastic magnetoelastic effects have also been observed in the behavior of the elastic constants and ultrasonic attenuation related to longitudinal acoustic waves propagating along the hexagonal c axis of gadolinium.[3,6,8] Other pure modes do not exhibit such effects. It should be added in this context that the magnetoelastic constants are functions of the magnetostriction constants, and have been determined experimentally.[19-21]

Several experimental and theoretical investigations[22-25] have been devoted to the study of the effect of hydrostatic pressure on the magnetization behavior of gadolinium in order to determine the dependence of the exchange integral on the interatomic distance and the pressure dependence of the Curie temperature. Hydrostatic-pressure measurements are particularly important when magnetoelastic effects are observed, since the Ruderman–Kittel–Kasua–Yosida (RKKY) exchange integral is strain dependent.[26,27] Magnetization measurements under uniaxial stresses[28] showed that the Curie temperature of gadolinium is dependent exclusively on uniaxial stresses along the hexagonal c axis. This result is consistent with the anisotropy of the magnetoelastic constants in gadolinium below its Curie point.

The objective of the present study was to investigate the effect of hydrostatic pressure on the lattice-softening effect and ultrasonic attenuation anomalies occurring in the spin-reorientation region. Experimental results are discussed in terms of the first magnetocrystalline anisotropy constant K_1 and its pressure dependence. Recently, a theoretical model within the molecular-field approximation has been presented[29] explaining the experi-

mentally observed anomalies in the elastic constant c_{33} of gadolinium as a function of temperature and applied magnetic field.[8,9] In the present study, the problem will also be treated in the molecular-field approximation but using a simpler approach. It will be demonstrated why magnetoelastic effects affect only the c_{33} elastic mode and not c_{11} or any pure shear modes. This model permits also the calculation of the magnetoelastic contribution to K_1 and the pressure dependence of the spin-reorientation temperature in gadolinium, in agreement with the observed behavior.

II. EXPERIMENTAL DETAILS

The high-purity (99.9%) gadolinium single crystals, supplied by Metals Research, Cambridge, England, has been prepared by the zone-melting technique. The crystals were in the form of flat disks, 6 mm in diameter and about 5 mm thick. Sample faces were flat and parallel to better than two parts in 10^4. The thickness of each crystal was determined by means of a calibrated indicator stand to within $\pm 5\times10^{-4}$ mm.

Determination of the five independent elastic coefficients in an hexagonal crystal, namely, c_{11}, c_{12}, c_{13}, c_{33}, and c_{44}, requires measurement of acoustic wave velocities of plane longitudinal waves and appropriately polarized transverse waves propagating along three crystal directions. In the present work, three single crystals were used with the following nominal orientations: crystal A with disk axis Z parallel to the hexagonal c axis, crystal B with disk axis perpendicular to the c axis, and crystal C with disk axis at an angle $\phi = 45°$ to the hexagonal c axis. X-ray back-reflection Laue photographs indicated that the actual crystal orientations were within 2° of the nominal ones, which was accepted as a satisfactory deviation.

The sound velocities and ultrasonic attenuation were measured by means of an ultrasonic pulse technique at the frequency of 10 MHz. Experimental details and method of data analysis were described elsewhere.[3,30] The ultrasonic attenuation as a function of temperature and hydrostatic pressure has been measured in the temperature range 200–300 °K, thus covering the spin-reorientation region and the Curie point of gadolinium. A Matec (Providence, R.I.) attenuation comparator model 9000, monitored automatically by a Matec attenuation recorder model 2470, was employed. Conventional cryogenic and temperature-measuring techniques were used. The temperature of the crystals was determined to within 0.1 °K.

A low-temperature high-hydrostatic-pressure system (Basset Co., Paris) was employed. The pressure-transmitting medium was high-purity helium gas. This pressure system permits attaining about 12-kbar hydrostatic pressure at liquid-helium temperatures. Using a calibrated manganin gauge, the hydrostatic pressure could be determined to within 10 bar in the kbar range. The experimental runs were performed under isobaric conditions by scanning the variation of the sound velocities and ultrasonic attenuation within the temperature range 200–300 °K. In this work experimental data were recorded during heating and cooling at several constant hydrostatic pressures, between 1 and 6 kbar.

III. THEORY

A. Hamiltonian

The free energy of a magnetically ordered crystal can be expressed in the following manner[26]:

$$H = H_m + H_e + H_{\rm me} + H_a \,, \tag{1}$$

where H_m is the term containing the isotropic RKKY magnetic interaction and H_e is the classical expression for the elastic energy of a paramagnetic crystal,[26]

$$H_e = \frac{1}{2}\sum_{ij} c_{ij}^0 \epsilon_i \epsilon_j \,. \tag{2}$$

c_{ij}^0 are the elastic constants, and ϵ_i and ϵ_j are the lattice strains in an orthogonal system. H_a is the magnetocrystalline anisotropic energy expressed by the usual macroscopic relation[31]

$$\langle H_a \rangle = K_1 \sin^2\theta + K_2 \sin^4\theta + K_3 \sin^6\theta \cos 6\phi \,, \tag{3}$$

where θ is the angle between the magnetic moment and the hexagonal c axis of the crystal. The anisotropy constants K_1, K_2, and K_3 are temperature dependent. However, in the range of interest in the present work K_3 is negligible. $H_{\rm me}$ is the magnetoelastic interaction coupling the spin system to the strain. This term includes both the one-ion and two-ion contributions to the magnetoelastic Hamiltonian and terms in the first order in strain and second order in spin components. As do Callen and Callen,[26] we express the magnetoelastic Hamiltonian in terms of the one-ion contribution. Thus,

$$\langle H_{\rm me} \rangle = -\sum_f \sum_{\Gamma, jj'} \tilde{B}_{jj'}^{\Gamma}(f) \sum_i \epsilon_i^{\Gamma, j} S_i^{\Gamma, j'}(f) \,, \tag{4}$$

where the $\tilde{B}_{jj'}^{\Gamma}(f)$ are the phenomenological magnetoelastic constants and $S_i^{\Gamma, j'}$ are the spin functions. The summation is over f, which is the index of the ion site in the crystal lattice.

B. Magnetoelastic Energy

The magnetoelastic energy term [Eq. (4)] of the total Hamiltonian can be determined by means of the molecular-field approximation.[26] In the case of gadolinium, the quantitative evaluation of the magnetoelastic coupling constants which will be

presented in this paper can be made more straightforward by using Cartesian coordinates. The magnetoelastic energy expressed in terms of Cartesian strains is therefore the following:

$$\langle H_{\rm me}\rangle = -(\epsilon_{xx}+\epsilon_{yy})\sum_f\{\tilde B^1_{xx}[(S^z_f)^2-\tfrac{1}{3}S(S+1)]+\tilde B^2_{xx}[(S^x_f)^2-(S^y_f)^2]\}-\epsilon_{zz}\sum_f\tilde B_{zz}[(S^z_f)^2-\tfrac{1}{3}S(S+1)]$$
$$-\epsilon_{xy}\sum_f\tilde B_{xy}(S^x_fS^y_f+S^y_fS^x_f)-\epsilon_{yz}\sum_f\tilde B_{yz}(S^y_fS^z_f+S^z_fS^y_f)-\epsilon_{zx}\sum_f\tilde B_{zx}(S^z_fS^x_f+S^x_fS^z_f)\ , \quad (5)$$

where the symmetric strains are given in terms of the Cartesian strains[26]

$$\begin{aligned}&\epsilon^{\alpha,1}=\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz}\ ,\\ &\epsilon^{\alpha,2}=\tfrac{1}{2}\sqrt{3}[\tfrac{2}{3}\epsilon_{zz}-\tfrac{1}{3}(\epsilon_{xx}+\epsilon_{yy})]\ ;\\ &\epsilon^{\gamma}_1=\tfrac{1}{2}(\epsilon_{xx}-\epsilon_{yy})\ ,\quad \epsilon^{\gamma}_2=\epsilon_{xy}\ ;\\ &\epsilon^{\beta}_1=\epsilon_{yz}\ ,\qquad\quad \epsilon^{\beta}_2=\epsilon_{zx}\ .\end{aligned} \quad (6)$$

The magnetoelastic coupling coefficients can also be defined in Cartesian coordinates in the following manner:

$$\begin{aligned}&\tilde B^1_{xx}=\tilde B^1_{yy}=\tfrac{1}{2}\sqrt{3}\,\tilde B^{\alpha}_{12}-\tfrac{1}{4}\tilde B^{\alpha}_{22}\ ,\\ &\tilde B^2_{xx}=\tilde B^2_{yy}=\tfrac{1}{2}\tilde B^{\gamma}\ ,\quad \tilde B_{zz}=\tfrac{1}{2}\sqrt{3}\,\tilde B^{\alpha}_{12}+\tfrac{1}{2}\tilde B^{\alpha}_{22}\ ,\\ &\tilde B_{xy}=\tfrac{1}{2}\tilde B^{\gamma}\ ,\qquad \tilde B_{yz}=\tilde B_{zx}=\tfrac{1}{2}\tilde B^{\epsilon}\ .\end{aligned} \quad (7)$$

An alternative way[26] of expressing $\langle H_{\rm me}\rangle$ based on the isomorphism between the spin functions and the magnetization functions is by using the components of the net magnetization α^x, α^y, and α^z:

$$\begin{aligned}\langle H_{\rm me}\rangle = &-B^{\alpha}_{12}\epsilon^{\alpha,1}\tfrac{1}{2}\sqrt{3}[(\alpha^z)^2-\tfrac{1}{3}]\\ &-B^{\alpha}_{22}\epsilon^{\alpha,2}\tfrac{1}{2}\sqrt{3}\,[(\alpha^z)^2-\tfrac{1}{3}]\\ &-B^{\gamma}\{\epsilon^{\gamma}_1\tfrac{1}{2}[(\alpha^x)^2-(\alpha^y)^2]+\epsilon^{\gamma}_2(\alpha^x\alpha^y)\}\\ &-B^{\epsilon}[\epsilon^{\epsilon}_1(\alpha^y\alpha^z)+\epsilon^{\epsilon}_2(\alpha^z\alpha^x)]\ .\end{aligned} \quad (8)$$

Similarly, substituting the Cartesian strains [Eq. (6)] and the Cartesian magnetoelastic coupling coefficients [Eq. (7)], the expression for $\langle H_{\rm me}\rangle$ [Eq. (8)] becomes

$$\begin{aligned}\langle H_{\rm me}\rangle = &-(\epsilon_{xx}+\epsilon_{yy})\{B^1_{xx}[(\alpha^z)^2-\tfrac{1}{3}]+B^2_{xx}[(\alpha^x)^2-(\alpha^y)^2]\}\\ &-\epsilon_{zz}B_{zz}[(\alpha^z_1)^2-\tfrac{1}{3}]-\epsilon_{xy}B_{xy}(2\alpha^x\alpha^y)\\ &-\epsilon_{yz}B_{yz}(2\alpha^y\alpha^z)-\epsilon_{zx}B_{zx}(2\alpha^z\alpha^x)\ .\end{aligned} \quad (9)$$

The standard procedure for solving the spin function is to transform the spin components to local coordinates at each spin site.[32,33] The new Z' axis is directed parallel to the equilibrium direction of the spin, and X' and Y' are orthogonal to Z', so that X' and Z' remain in the XZ plane:

$$\begin{aligned}&S^x_f=S^{x'}_f\cos\theta+S^{z'}_f\sin\theta\ ,\quad S^y_f=S^{y'}_f\ ,\\ &S^z_f=-S^{x'}_f\sin\theta+S^{z'}_f\cos\theta\ .\end{aligned} \quad (10)$$

It was shown[17] that the anisotropy in the basal plane is very weak. Assuming that the deviation of the spin from its equilibrium direction is small, several approximations can be made:

$$\langle(S^{x'}_f)^2-(S^{y'}_f)^2\rangle\cong 0\ ,\quad \left\langle\sum_f(S^{z'}_f)^2\right\rangle\cong\sum_f\langle(S^{z'}_f)^2\rangle\ ,$$

$$\langle S^{i'}_fS^{j'}_f\rangle\cong 0\ .$$

Using these approximations and by substitution in Eq. (10), one obtains

$$\langle(S^z_f)^2\rangle=(1-\sin^2\theta)\langle(S^{z'}_f)\rangle\ , \quad (11)$$

$$\langle(S^x_fS^y_f)\rangle=\langle(S^y_fS^z_f)\rangle=\langle(S^z_fS^x_f)\rangle=0\ . \quad (12)$$

In the case of the magnetic structure of gadolinium the only nonvanishing term is

$$\sum_f[\langle(S^z_f)^2\rangle-\tfrac{1}{3}S(S+1)]$$
$$=\sum_f[\langle(S^{z'}_f)^2\rangle-\tfrac{1}{3}S(S+1)]-\sin^2\theta\langle(S^{z'}_f)^2\rangle\ . \quad (13)$$

For the equilibrium direction of the spin, $\sum_f[\langle(S^{z'}_f)^2\rangle-\frac{1}{3}S(S+1)]$ is a temperature-dependent constant. The term $\sum_f\langle(S^{z'}_f)^2\rangle$ is proportional to the reduced saturation magnetization.

Defining $B^{\theta}_{jj'}$ as θ-dependent magnetoelastic constants in the following manner:

$$B^{\theta}_{xx}=B^{\theta}_{yy}=(\tfrac{1}{2}\sqrt{3}\,\tilde B^{\alpha}_{12}-\tfrac{1}{4}\tilde B^{\alpha}_{22})\sum_f\langle(S^{z'})^2\rangle$$

and (14)

$$B^{\theta}_{zz}=(\tfrac{1}{2}\sqrt{3}\,\tilde B^{\alpha}_{12}+\tfrac{1}{2}\tilde B^{\alpha}_{22})\sum_f\langle(S^{z'})^2\rangle\ ,$$

and defining B^{c}_{jj} as θ-independent magnetoelastic constants such as

$$B^{c}_{xx}=B^{c}_{yy}=(\tfrac{1}{2}\sqrt{3}\,\tilde B^{\alpha}_{12}-\tfrac{1}{4}\tilde B^{\alpha}_{22})\sum_f[\langle(S^{z'}_f)^2\rangle-\tfrac{1}{3}S(S+1)]$$

and (15)

$$B^{c}_{zz}=(\tfrac{1}{2}\sqrt{3}\,\tilde B^{\alpha}_{12}+\tfrac{1}{2}\tilde B^{\alpha}_{22})\sum_f[\langle(S^{z'}_f)^2\rangle-\tfrac{1}{3}S(S+1)]\ ,$$

and substituting Eqs. (14) and (15) into Eq. (5), the magnetoelastic term of the total Hamiltonian becomes

$$\begin{aligned}\langle H_{\rm me}\rangle = &\epsilon_{xx}(B^{\theta}_{xx}\sin^2\theta-B^{c}_{xx})\\ &+\epsilon_{yy}(B^{\theta}_{yy}\sin^2\theta-B^{c}_{yy})\\ &+\epsilon_{zz}(B^{\theta}_{zz}\sin^2\theta-B^{c}_{zz})\ .\end{aligned} \quad (16)$$

The B^{θ}_{jj} are important in the spin-reorientation region of gadolinium, whereas the B^{C}_{jj} magnetoelastic constants are of importance in the proximity of the Curie temperature. The proportionality between these two sets of constants holds at all temperatures.

Finally, the general expression of the total Hamiltonian [Eq. (1)] for gadolinium in the molecular-field approximation and in the Cartesian representation is

$$\langle H\rangle = H_m + \tfrac{1}{2}\sum_{ij} c^0_{ij}\epsilon_i\epsilon_j + K_1(T)\sin^2\theta + K_2(T)\sin^4\theta + (\epsilon_{xx}+\epsilon_{yy})(B^{\theta}_{xx}\sin^2\theta - B^{C}_{xx}) + \epsilon_{zz}(B^{\theta}_{zz}\sin^2\theta - B^{C}_{zz}) . \quad (17)$$

In the spin-reorientation region the effect of hydrostatic pressure on the total Hamiltonian as expressed in Eq. (17) is therefore to make an additional contribution to the first anisotropy term $K_1(T)$. The contribution is in the form of the $\epsilon_{jj}B^{\theta}_{jj}$ terms, since both the first anisotropy and the magnetoelastic contributions have an identical θ dependence.

Equation (17) will be used in the next section to show why the experimentally observed lattice softening in the spin-reorientation region is exhibited mainly in the c_{33} elastic constant. Furthermore, by means of this expression the pressure dependence of the elastic anomalies in gadolinium will be discussed.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Lattice Softening in Spin-Reorientation Region

Several authors have reported the temperature dependence of the elastic constants of gadolinium in single crystals[1,2,4,8] and polycrystals.[3] In the present study the single-crystal elastic constants of gadolinium c_{11}, c_{33}, c_{44}, c_{12}, and c_{13} have been redetermined from liquid helium to room temperature. Our results confirm the previously published ones[2,8] to within the reported experimental errors, and therefore will not be presented here.

The elastic constants and ultrasonic attenuation display characteristic anomalies at the Curie point of gadolinium ($T_c = 291$ °K). At the beginning of the spin-reorientation region ($T_f = 231$ °K) only longitudinal waves propagating along the c axis (i.e., c_{33} elastic constant) and its ultrasonic attenuation exhibit anomalies. The elastic constants and the attenuation of the shear modes, c_{44} and c_{66}, and those of the longitudinal waves propagating along the a direction of the hexagonal gadolinium, c_{11}, do not display any anomalies in the vicinity of the spin-reorientation temperature T_f. This behavior is in agreement with the theoretical model presented in Sec. III and will subsequently be applied to gadolinium.

The elastic anomaly of gadolinium in the spin-reorientation region[8] was considered to be of magnetoelastic origin and treated by Levinson and Shtrikman[31] in terms of the variation of the cone angle θ of the easy direction of magnetization as a function of strain. The lattice-softening phenomenon was treated in a manner similar to the treatment of the proximity of the displacive magnetic phase transition in erbium orthoferrite.[34]

The elastic constants are defined as the second derivatives of the lattice energy with respect to strain, $c_{ij} = d^2H/d\epsilon_i d\epsilon_j$. In the spin-reorientation region the spin easy direction is strain dependent [Eq. (17)]. Therefore, the equilibrium θ value for any strain can be evaluated by the derivative of the Hamiltonian in Eq. (17) with respect to θ:

$$\left(\frac{\partial H}{\partial\theta}\right)_{\epsilon_{ij}} = 0 = 2[K_1 + (\epsilon_{xx}+\epsilon_{yy})B^{\theta}_{xx} + \epsilon_{zz}B^{\theta}_{zz}]\sin\theta\cos\theta + 4K_2\sin^3\theta\cos\theta . \quad (18)$$

Solutions of Eq. (18) are with $\theta = 0$ for the normal ferromagnetic region of gadolinium, $\theta = \frac{1}{2}\pi$ for the maximum θ in the spin-reorientation region, and for any value of θ in this region

$$\sin^2\theta = -\frac{K_1 + (\epsilon_{xx}+\epsilon_{yy})B^{\theta}_{xx} + \epsilon_{zz}B^{\theta}_{zz}}{2K_2} . \quad (19)$$

The meaning of Eq. (19) is that the angle θ not only depends on K_1 but also is strain dependent, or pressure dependent. Since the second anisotropy-energy constant of gadolinium K_2 is positive over the whole temperature range,[14] the spin-reorientation region should begin where the numerator of Eq. (19) becomes negative.[31] On cooling gadolinium below its T_C (291 °K), θ is zero until the right-hand side of Eq. (19) becomes negative, i.e., at T_f (231 °K). Equation (19) also indicates that hydrostatic strains should decrease T_f, as is indeed borne out by experiment (Figs. 1 and 2). Without any applied strains ($\epsilon_{ij} = 0$) the spin-reorientation region and the value of θ in this region are defined by K_1 only.

It is now possible to evaluate the elastic constants in the spin-reorientation region, where the variation of θ is strain dependent. First,

$$\frac{dH}{d\epsilon_i} = \left(\frac{\partial H}{\partial\epsilon_i}\right)_{\theta} + \frac{\partial H}{\partial\theta}\frac{\partial\theta}{\partial\epsilon_i} = \frac{\partial H_i}{\partial\epsilon_i} , \quad (20)$$

since $\partial H/\partial\theta = 0$ under equilibrium conditions. Therefore, by substitution of the Hamiltonian expressed in Eq. (17) into Eq. (20),

$$\frac{dH}{d\epsilon_i} = \sum_j \epsilon_j c^0_{ij} + \frac{\partial}{\partial\epsilon_i}\langle H_{me}\rangle . \quad (21)$$

Treating separately each mode, e.g., c_{11}, we obtain by differentiating Eq. (21) with respect to strain,

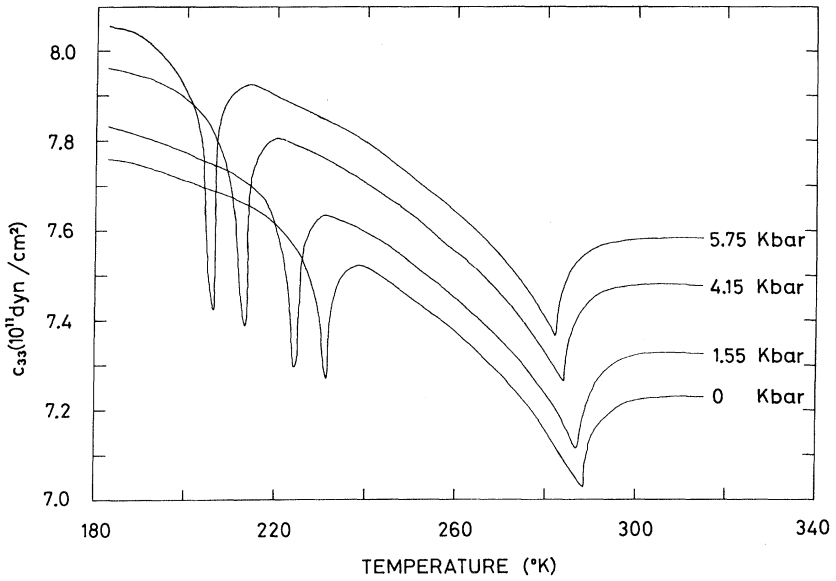

FIG. 1. Temperature dependence of the elastic constant c_{33} of gadolinum at different hydrostatic pressures.

$$c_{11} = \frac{\partial^2 H}{\partial \epsilon_{xx}^2} = c_{11}^0 + B_{xx}^\theta \frac{d(\sin^2\theta)}{d\epsilon_{xx}} \quad . \tag{22}$$

In the region where θ is constant ($\theta = 0$ or $\theta = \frac{1}{2}\pi$), $c_{ij} = c_{ij}^0$. In the spin-reorientation region, where θ is given by Eq. (19), the elastic constants will take the following form:

$$c_{11} = c_{11}^0 - (B_{xx}^\theta)^2/2K_2 \,,$$
$$c_{33} = c_{33}^0 - (B_{zz}^\theta)^2/2K_2 \,,$$
$$c_{12} = c_{12}^0 - (B_{xx}^\theta)^2/2K_2 \,,$$
$$c_{13} = c_{13}^0 - (B_{xx}^\theta B_{zz}^\theta)/2K_2 \,,$$
$$c_{44} = c_{44}^0 \,, \quad c_{66} = c_{66}^0 \,. \tag{23}$$

Equations (23) indicate that lattice-softening effects are to be expected in the spin-reorientation region in the pure longitudinal modes c_{11} and c_{33} but not in the pure shear modes c_{44} and c_{66}. This is a consequence of the statement in Eq. (17), where no magnetoelastic terms of the form $\epsilon_{xy}B_{xy}$ and $\epsilon_{zx}B_{zx}$ appear. The reason is that the corresponding spin correlation functions vanish.

In order to be able to evaluate quantitatively the lattice-softening effect in c_{11} and c_{33} elastic constants, the magnetoelastic coupling coefficients

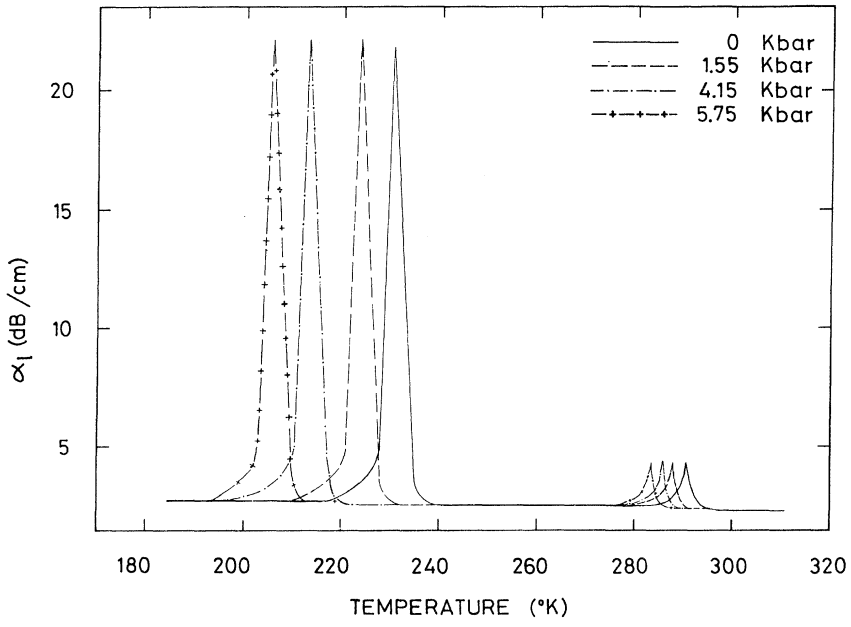

FIG. 2. Temperature dependence of the longitudinal-wave ultrasonic attenuation at different hydrostatic pressures.

B_{xx} and B_{zz} should be determined. The procedure of Callen and Callen[26] was adopted using the published magnetostriction data of gadolinium.[21] Since magnetostriction is defined as the equilibrium strain of magnetoelastic origin, one can derive it by differentiation of the magnetoelastic Hamiltonian [Eq. (8)] with respect to strain. The equilibrium condition yields

$$\begin{aligned} &-B_{12}^{\alpha}\tfrac{1}{2}\sqrt{3}\,[(\alpha^{z})^{2}-\tfrac{1}{3}]+c_{11}^{\alpha}\epsilon^{\alpha,1}+c_{12}^{\alpha}\epsilon^{\alpha,2}=0\,,\\ &-B_{22}^{\alpha}\tfrac{1}{2}\sqrt{3}\,[(\alpha^{z})^{2}-\tfrac{1}{3}]+c_{12}^{\alpha}\epsilon^{\alpha,1}+c_{22}^{\alpha}\epsilon^{\alpha,2}=0\,. \end{aligned} \tag{24}$$

The strains $\epsilon^{\alpha,1}$ and $\epsilon^{\alpha,2}$ are the equilibrium strains. These strains and the elastic constants are expressed in Eq. (24) in terms of the symmetric representation.[26] For the hexagonal symmetry they are defined as

$$\begin{aligned} c_{11}^{\alpha}&=\tfrac{1}{9}(2c_{11}+2c_{12}+4c_{13}+c_{33})\,,\\ c_{12}^{\alpha}&=(2/3\sqrt{3})\,(-c_{11}-c_{12}+c_{13}+c_{33})\,,\\ c_{22}^{\alpha}&=\tfrac{2}{3}(c_{11}+c_{12}-4c_{13}+2c_{33})\,. \end{aligned} \tag{25}$$

The equilibrium strains can be equated to the magnetostriction constants by applying saturation magnetic fields.[19] Results of such treatment are

$$\lambda_A=\bar{\epsilon}_{xx}\,,\quad \lambda_B=\bar{\epsilon}_{yy}\,,\quad \lambda_C=\bar{\epsilon}_{zz}\,. \tag{26}$$

Consequently, the magnetoelastic coupling coefficients are

$$\begin{aligned} B_{12}^{\alpha}&=-(2/\sqrt{3})\,(\lambda_A+\lambda_B+\lambda_C)\,c_{11}^{\alpha}+\tfrac{1}{3}(\lambda_A+\lambda_B-2\lambda_C)c_{12}^{\alpha}\,,\\ B_{22}^{\alpha}&=-(2/\sqrt{3})\,(\lambda_A+\lambda_B+\lambda_C)\,c_{12}^{\alpha}+\tfrac{1}{3}(\lambda_A+\lambda_B-2\lambda_C)c_{22}^{\alpha}\,. \end{aligned} \tag{27}$$

Finally, substitution into Eq. (7) permits determination of the magnetoelastic coupling coefficients B_{xx} and B_{zz} of gadolinium. Their temperature dependence is shown in Fig. 3. The behavior of the magnetoelastic coupling coefficients of gadolinium explains why at T_f lattice softening occurs only in the c_{33} elastic constant, and not in c_{11}. At T_f, $|B_{xx}| \ll |B_{zz}|$. It is expected that the ratio of the dips in the elastic constants at T_f, $\Delta c_{11}/\Delta c_{33} \sim (B_{xx}/B_{zz})^2$, is according to Eq. (23), and therefore the lattice softening in c_{11} should be negligible compared to that in the c_{33} elastic constant. To summarize, the theoretical model presented here indicates that anomalous behavior of the pure elastic modes in gadolinium at T_f should be limited to c_{33} only. Experimental results obtained in the present investigation confirm the validity of this statement.

Figures 1 and 2 show, respectively, the temperature dependence of the c_{33} elastic constant and ultrasonic attenuation at different hydrostatic pressures, up to 6 kbar. Two main pressure-dependent features are displayed. The first is a linear increase in the absolute values of c_{33} by a rate of 5.47×10^{9} dyn cm^{-2}kbar^{-1} at temperatures in the paramagnetic region and at 4.09×10^{9} dyn cm^{-2} kbar^{-1} below T_C, e.g., at 273 °K. The second pressure-dependent effect exhibited in Figs. 1 and 2 is the shift in both T_C and T_f as a function of hydrostatic pressure. The significance of this behavior will be discussed in Sec. IV B.

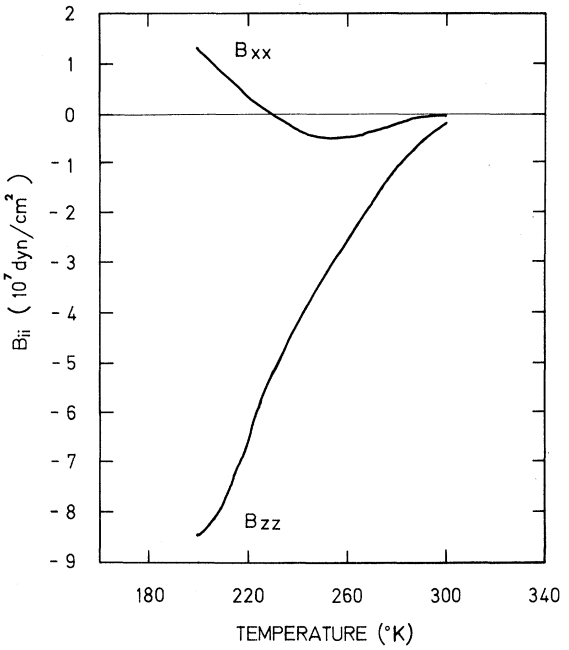

FIG. 3. Temperature dependence of the magnetoelastic coupling coefficients B_{xx} and B_{zz} of gadolinium.

At any hydrostatic pressure, the temperature variation of c_{33} in gadolinium is the sum of two components, Eqs. (22) and (23): c_{33}^{0} of the paramagnetic lattice with a normal temperature dependence and a magnetoelastic contribution including the θ-dependent and θ-independent parts.

The magnetoelastic contribution is not directly pressure dependent. It depends on the magnetic ordering which is in turn affected by the corresponding shifts in T_C and T_f with hydrostatic pressure (Fig. 4).

The increase in c_{33} with pressure (Fig. 1) is due to the variation of c_{33}^{0} with hydrostatic pressure through the elastic constants of higher order, which are temperature independent. The difference in the rate of change with pressure dc_{33}/dP of 5.47×10^{9} dyn cm^{-2}kbar^{-1} compared with 4.09×10^{9} dyn cm^{-2}kbar^{-1} for the paramagnetic and ferromagnetic regions, respectively, can be attributed to the increased stiffness of the gadolinium lattice in the ferromagnetic region.

B. Pressure Dependence of T_C and T_f

The anomalies in the temperature dependence of the elastic constant c_{33} and ultrasonic attenuation of gadolinium at T_C and T_f shift to lower tempera-

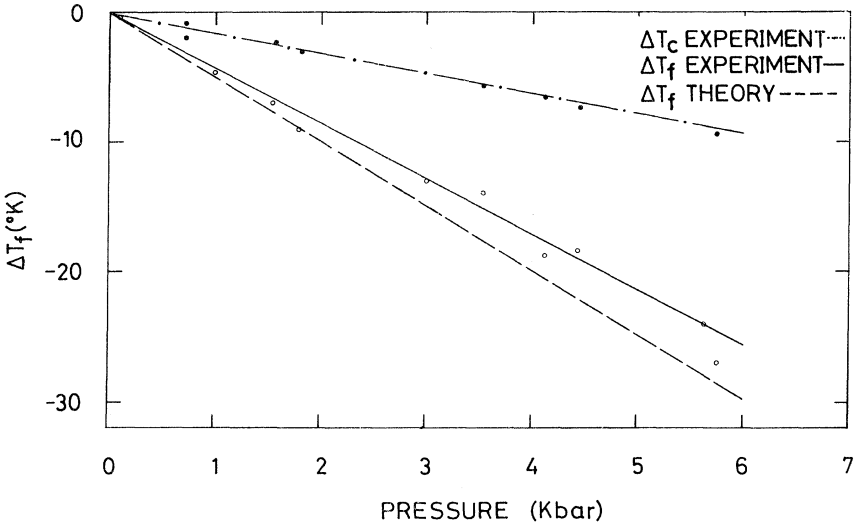

FIG. 4. Effect of hydrostatic pressure on the shift of the critical points T_c (Curie point) and T_f (spin reorientation) in gadolinium.

tures upon application of hydrostatic pressure. This behavior is exhibited in Figs. 1, 2, and 4. The Curie temperature T_C and the spin-reorientation temperature T_f are found to decrease linearly with increasing pressure (Fig. 4), although with differing slopes of (-1.3 ± 0.1) and (-4.3 ± 0.2) °K kbar^{-1}, respectively. The shift of T_C with pressure as determined in the present paper compares satisfactorily with high-pressure magnetization[28] and magnetoresistance[23] measurements -1.55 and -1.4 °K kbar^{-1}, respectively. However, Fleming and Liu[25] report a theoretical value of $\partial T_C/\partial P = -2.3$ °K kbar^{-1}. The magnetization measurements were performed under uniaxial stresses and the reported values[28] were $\partial T_C/\partial P_{cc} = 1.55$ °K kbar and $\partial T_C/\partial P_{aa} = 0$ °K kbar^{-1}. This behavior is in accord with the pressure dependence of T_C as determined from the elasticity data in the present paper. Moreover, this behavior is also expected from the temperature variation of the corresponding magnetoelastic constants (Fig. 3). Near the Curie point, B_{xx} is -0.1 erg cm^{-3} compared with -0.7 erg cm^{-3} for B_{zz}. Therefore, it is expected that $|\partial T_C/\partial P_{aa}| < |\partial T_C/\partial P_{cc}|$, as is indeed experimentally observed from the shift in the anomalies of c_{11} and c_{33} at T_C in gadolinium.

Figure 4 shows a linear shift of the spin-reorientation temperature T_f with hydrostatic pressure as determined from the measurements of the elastic constants and ultrasonic attenuation. The slope of dT_f/dP is (-4.3 ± 0.2) °K kbar^{-1}. It was shown in Sec. IV A that the spin-reorientation region, and consequently the lattice softening, is correlated to the temperature T_f where the first anisotropy-energy constant K_1 changes sign. This consideration is valid if the sample is strain free. Upon application of hydrostatic pressure the spin-reorientation region occurs at temperatures where K_1^{eff} changes sign. K_1^{eff} is the numerator of the right-hand side of Eq. (19) and is expressed as

$$K_1^{\text{eff}} = K_1 + \Delta K_1(\epsilon) = K_1 + (\epsilon_{xx} + \epsilon_{yy}) B_{xx}^{\theta} + \epsilon_{zz} B_{zz}^{\theta} \,. \tag{28}$$

The magnetoelastic contribution to K_1 is

$$\Delta K_1(\epsilon) = (\epsilon_{xx} + \epsilon_{yy}) B_{xx}^{\theta} + \epsilon_{zz} B_{zz}^{\theta} \,.$$

It was shown in Eq. (19) that the magnetoelastic contribution $\Delta K_1(\epsilon)$ is of the same θ-functional dependence as the first anisotropy-energy constant K_1.

The uniaxial strains induced by a hydrostatic pressure P can be expressed in terms of the linear compressibilities:

$$\epsilon_{xx} = \epsilon_{yy} = -\beta_{\perp} P \,, \quad \epsilon_{zz} = -\beta_{\parallel} P \tag{29}$$

where $\beta_{\perp}$ and $\beta_{\parallel}$ are the linear compressibilities perpendicular and parallel, respectively, to the hexagonal c axis of gadolinium. $\beta_{\perp}$ and $\beta_{\parallel}$ are experimentally determined quantities, derived from elasticity measurements.

From Eqs. (28) and (29) the hydrostatic-pressure derivative of the first anisotropy-energy constant can be determined:

$$\frac{dK_1}{dP} = -(2\beta_{\perp} B_{xx}^{\theta} + \beta_{\parallel} B_{zz}^{\theta}) \,. \tag{30}$$

The pressure contribution causes the K_1^{eff} to change sign at lower temperatures when hydrostatic pressure is increased. The pressure dependence of the spin-reorientation temperature T_f can therefore be calculated from

$$\frac{dT_f}{dP} = \frac{dK_1/dP}{(dK_1/dT)_{T_f}} \,,$$

where dK_1/dP can be determined by Eq. (30). The calculated value for dT_f/dP was found to be -4.9 °K

$kbar^{-1}$. In this calculation the contribution from the anomalous thermal expansion in the c axis of gadolinium[18] in this temperature region was taken into account. The calculated value of dT_f/dP is therefore in satisfactory agreement with the experimental one, $(-4.3 \pm 0.2)\,^{\circ}K\,kbar^{-1}$, as shown in Fig. 4.

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