

Critical dynamics of gadolinium

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Critical propagation of ultrasonic waves in a gadolinium single crystal was investigated in the range of reduced temperatures 10^{-4} – 10^{-1} at frequencies 10–30 MHz. The critical exponents, including the dynamic exponent, and the amplitudes characteristic of the dynamics of critical fluctuations were determined. The relaxation and fluctuation contributions to the anomalous absorption were separated and their scaling functions were derived. The validity of the dynamic scaling concepts was demonstrated for absorption. Moreover, it was established that the dipole interactions played the dominant role in critical dynamics of gadolinium.

1. INTRODUCTION

Anomalies of the frequency dependence of the absorption coefficient $\alpha_k(\omega)$ and of the velocity v_l of longitudinal acoustic waves in magnetically ordered crystals near the Curie point T_c are due to the spin–phonon interaction of magnetostriction origin.^{1–8} Depending on the nature of the coupling between acoustic waves and the spin system, it is usual to distinguish between the relaxation and fluctuation mechanisms of anomalous changes in α_k and v_l . The relaxation mechanism due to the linear coupling of acoustic waves to the order parameter was first introduced by Landau and Khalatnikov¹ to account for the behavior of the propagation of sound near the λ point of helium. This mechanism is manifested only in an ordered phase when the time average of the order parameter differs from zero. The fluctuation mechanism, originating from the quadratic coupling of acoustic waves to fluctuations of the spin system, leads to anomalies of α_k and v_l both above and below T_c (Refs. 2–8).

The degree of singularity of α_k and v_l in the case of the fluctuation mechanism is governed by the nature of the exchange interaction, magnetic ordering, and anisotropy. In particular, in the case of magnetic insulators where the exchange interaction is of the short-range type, the critical absorption is due to fluctuations of the density of the spin energy and the decay of these fluctuations is due to slow spin-lattice relaxation.^{3,4} In the case of the long-range exchange interaction (encountered in magnetic metals) the main contribution to the critical absorption comes from fluctuations of the order parameter and relaxation is of spin–spin nature.^{2,3}

A theory of the critical absorption of acoustic waves in magnetic metals developed by Kadanoff (theory of interacting modes)² and Kawasaki (theory of dynamic critical variables)⁵ for the hydrodynamic range defined by $\omega\tau \ll 1$, where ω is the angular frequency of sound and τ is the relaxation time, has been confirmed experimentally by investigations of ultrasound propagating in nickel,⁶ MnP,⁷ and rare-earth metals.³ Recent theoretical treatments^{8–10} have demonstrated that α_k is described by a simple scaling function of the variable $\omega\tau$ not only in the hydrodynamic, but also in the critical range $\omega\tau \gg 1$. Experimental investigations carried out on a three-dimensional Heisenberg ferromagnet MnP (Refs. 7 and 9) and a two-dimensional Ising antiferromagnet Rb₂CoF₄ (Ref. 10) have confirmed the validity of the dynamic scaling concepts in the case of α_k . However, it has been found that the dynamic critical properties do not agree with

the Ising (Rb₂CoF₄) and Heisenberg (MnP) models, whereas the static critical properties are described well by these models.

This interesting circumstance, discovered so far for two magnetically ordered crystals, requires further theoretical and experimental studies. We therefore carried out a systematic investigation of the dynamic critical properties of magnetically ordered crystals by ultrasonic methods and shall report the experimental frequency and temperature dependences of the critical absorption and of the relative change in the velocity of longitudinal waves propagating in a gadolinium single crystal in the vicinity of the Curie point.

2. GENERAL DESCRIPTION OF GADOLINIUM AND EXPERIMENTAL METHOD

Gadolinium is a rare-earth metal with a simple magnetic order in which the magnetic moments are directed along the hexagonal axis at temperatures between ~ 230 K and T_c (Ref. 11). The Curie point of gadolinium varies within a fairly wide range, depending on the conditions during synthesis and on purity, and in the case of crystals of the highest quality it reaches 293.62 K (Refs. 12 and 13).

The static critical behavior of gadolinium is described well by the Heisenberg model with isotropic exchange and dipole interactions. For example, according to the experimental data we have $\beta = 0.375 \pm 0.005$, $\gamma = 1.39 \pm 0.02$, $\delta = 4.80 \pm 0.1$, and $\Delta = 0.55 \pm 0.05$, whereas Ref. 14 gives $\alpha = -0.09 \pm 0.005$ (here and later we shall use the generally accepted designations of the critical exponents).

Investigations of the dynamic critical properties of gadolinium are reported in Refs. 3, 11, and 15–17: they show that, depending on the purity of a sample, gadolinium can be classified as an isotropic or a uniaxial Heisenberg ferromagnet. This information follows from experiments on the critical propagation of ultrasonic waves, since gadolinium is not a material suitable for the application of the traditional methods (neutron scattering, dynamic susceptibility, etc.) used in investigations of the critical dynamics. Moreover, although the published experimental data cover a fairly wide range of temperatures $10^{-3} \leq t \leq 10^{-1}$ ($t = (T - T_c)/T_c$) and frequencies 5–180 MHz, the dynamics of critical fluctuations can be studied only in the hydrodynamic range.

In a study of the characteristics of the propagation of ultrasonic waves in the critical range we need to carry out measurements in the direct vicinity of T_c ; this imposes more stringent requirements in respect of the experimental behav-

ior. In particular, the temperature of a sample should be stabilized to within 10^{-4} K and it should be varied in steps not exceeding 10^{-3} K. The critical region can be reached also by increasing the frequency of ultrasonic waves, but in this case the overall absorption rises strongly and this limits the opportunities for measurements in the ordered phase when $\omega\tau = 1$. The optimal ranges for experimental investigation of the critical dynamics of gadolinium are 5–30 MHz in respect of the frequency and 10^{-4} – 10^{-1} in respect of the relative temperature.

Our measurements of the absorption coefficient were carried out by the echo pulse method but omitting a pulse selector and an amplitude detector, and using an electron-beam null indicator of the ÉLNI type to measure the echo pulse amplitude. As a result, we were able to reduce the error in the determination of α_k to 3% and the sensitivity was at least 0.01 dB.

The relative change in the velocity $\Delta v_l/v_0$, where v_0 is the velocity at 300 K, was measured by a pulse-phase method of superposition of pulses, for which the error and sensitivity at 10 MHz were 0.05% and 2×10^{-6} , respectively.

Elastic waves were excited using X-cut quartz plates with suitable resonance frequencies. Reliable acoustic contacts were established by bonding these quartz plates with an epoxy resin to the end of a cylindrical sample which was oriented to within 0.5° along the c axis.

All the measurements were made under steady-state thermal conditions with the temperature of a sample stabilized to within 5×10^{-4} K using a modified temperature control system of the VRT-2 type. A copper-constantan thermocouple, calibrated against a platinum resistance thermometer, was used as a temperature sensor.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Typical temperature dependences of the total absorption coefficient (25 MHz) and of the relative change in the

velocity of longitudinal waves (10 MHz), plotted in Fig. 1, show that α_k passes through a maximum and $\Delta v_l/v_0$ passes through a minimum in the vicinity of T_c . It should be pointed out that, as in the case of other magnetically ordered crystals (see, for example, Ref. 3), the anomalies of α_k and $\Delta v_l/v_0$ do not occur at $T_c = 293.37$ K, which was the Curie point deduced by us from magnetic measurements by the kink method,¹⁸ but were shifted toward lower temperatures. Moreover, the temperatures of the maximum of α_k and of the minimum $\Delta v_l/v_0$ did not coincide. For example, at $\omega/2\pi = 10$ MHz, the anomalies of α_k and $\Delta v_l/v_0$ were shifted relative to T_c by 0.16 K and 0.22 K. Characteristic features of the frequency dependences of the absorption and velocity were not only an increase of the absolute value of α_k and a shift of its maximum toward lower temperatures on increase in the frequency, but also the absence of relaxation anomalies due to trivial effects of the interaction of acoustic waves with domain walls (see Figs. 1–5).

In the subsequent analysis of the experimental results and comparison with the theory it was necessary to determine the critical absorption $\Delta\alpha_k = \alpha_k - \alpha_0$ (α_0 is the background absorption) and the relative change in the velocity $(\Delta v/v)_k = \Delta v/v_0 - \Delta v/v$ ($\Delta v/v$ is the normal change in the velocity). It was assumed that near the Curie point the value of α_0 was independent of T and the background absorption was assumed to be α_k at temperatures $T > T_c$ exceeding the Curie point by 20–30 K. The normal change in the velocity was separated by extrapolation of the $\Delta v_l/v_0$ data for the paramagnetic phase to T_c (Ref. 3). Bearing in mind that in the case of gadolinium in the paramagnetic phase the value of $\Delta v_l/v_0$ depended nonlinearly on T , whereas in the ferromagnetic phase the influence of the ΔE effect did not exceed 2% (see, for example, Ref. 11), we separated $(\Delta v/v)_k$ at $T > T_c$ and $T < T_c$ using the dependence

$$\Delta v_l/v_0 = a_0 + a_1 T + a_2 T^2, \quad (1)$$

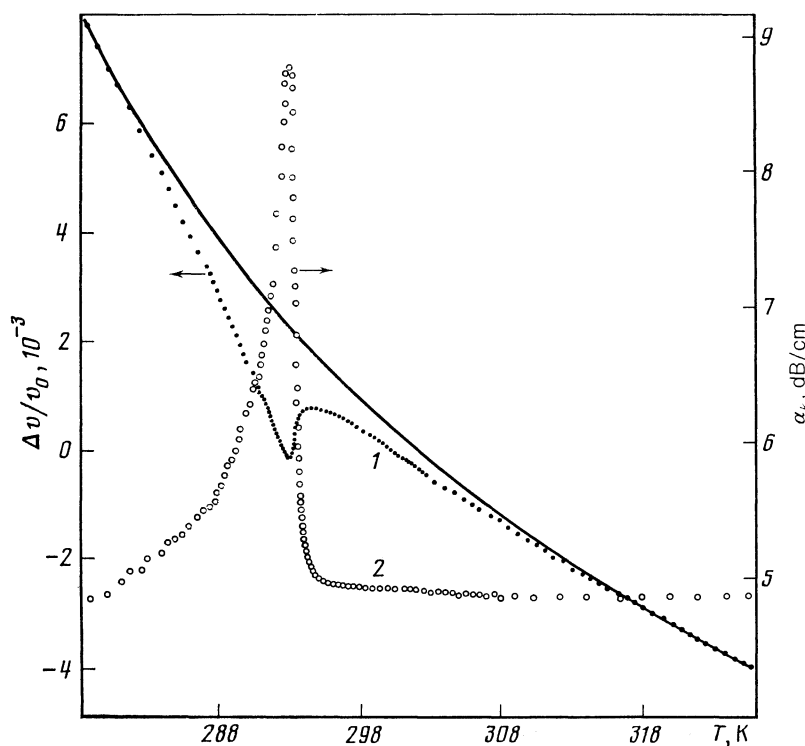


FIG. 1. Temperature dependences of the relative change in the velocity of sound at 10 MHz (1) and of the overall absorption coefficient of sound at 25 MHz (2) obtained for propagation along the hexagonal axis of gadolinium.

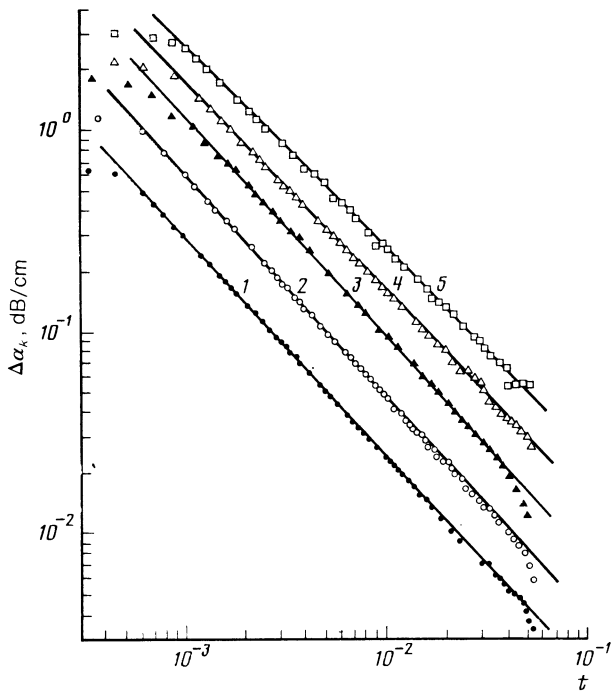


FIG. 2. Dependence of the critical absorption $\Delta\alpha_k$ on the reduced temperature t at different frequencies (MHz): 1) 10; 2) 15; 3) 20; 4) 25; 5) 30.

where $a_0 = 0.43524$, $a_1 = -2.6425 \times 10^{-3}$, and $a_2 = 3.977 \times 10^{-6}$. This dependence is represented by the continuous curve in Fig. 1.

The temperature dependences of $\Delta\alpha_k$ and $(\Delta v/v)_k$ found by the above methods are plotted in a double logarithmic scale in Figs. 2 and 3. Here, the straight lines represent the following power laws:

$$\Delta\alpha_k/\omega^2 = B_0^\pm t^{-n_i^\pm}; \quad (2)$$

$$(\Delta v/v)_k = V_0^\pm t^{-n_i^\pm}, \quad (3)$$

where the plus and minus signs refer to $T > T_c$ and $T < T_c$, respectively. The values of the critical exponents (η_i^\pm, n_i^\pm) and amplitudes (B_0^\pm, V_0^\pm) describing best the experimental results are listed in Table I.

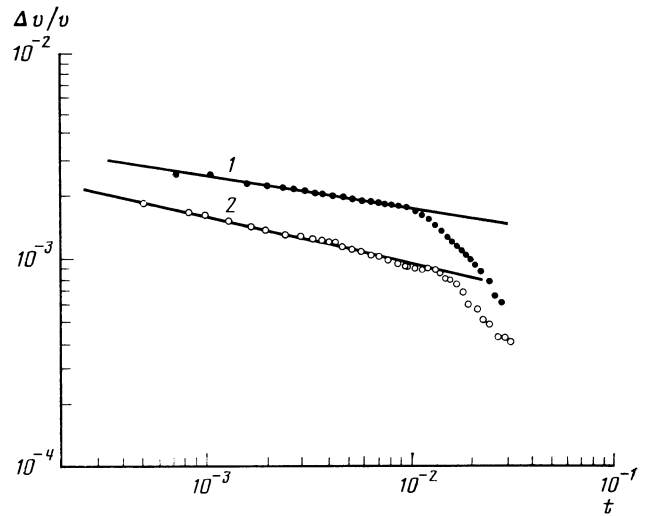


FIG. 3. Dependences of the critical relative change in the velocity at 10 MHz on the reduced temperature: 1) $T < T_c$; 2) $T > T_c$.

In comparing the experimental values of η_i, n_i, B_0 , and V_0 with the theoretical predictions for Heisenberg magnetic materials, we used the ideas that follow from the theory of interacting modes,² according to which we should have the following relationship in the limit $\omega\tau \ll 1$:

$$\Delta\alpha_k = B\omega^2\tau, \quad (4)$$

$$B = \frac{\rho k_B T_c \gamma v}{\xi_0^3} \left(\frac{1}{T_c} \frac{dT_c}{dP} \right)^2 t^{2\alpha - 2 + 3\nu}, \quad (5)$$

where ρ is the density of a crystal; k_B is the Boltzmann constant; ξ_0 and ν are the critical amplitude and exponent of the correlation length; $(1/T_c)(dT_c/dP)$, is the shift of T_c under the influence of a hydrostatic pressure P . When $\Delta\alpha_k$ is expressed in this way, we must bear in mind that B is a singular quantity, since the experimentally determined amplitude B_0 is independent of temperature. This was first pointed out by Pokrovskii and Khalatnikov¹⁹ in a discussion of the anomalous absorption of sound near the λ point of helium. They demonstrated that B has the same singularity as the

TABLE I. Critical exponents and amplitudes of gadolinium.

Critical exponents and amplitudes	$T > T_c$			$T < T_c$	
	experiments	theory		experiments	Landau-Khalatnikov theory
		uniaxial magnetic materials	isotropic magnetic materials		
η_i	1.15 ± 0.05	1.218	1.883	1.08 ± 0.05	1.0
n_i	0.20 ± 0.02	-0.03	0.122	0.14 ± 0.02	0
x	0.92 ± 0.07	1.25	1.67	0.94 ± 0.07	1.0
y	0.08 ± 0.04	0.123	0.147	0.06 ± 0.04	0
z	1.37 ± 0.10	2.17	2.48	1.39 ± 0.10	2
$B_0, \text{cm}^{-1} \cdot \text{s}^2$	$(5 \pm 0.1) \cdot 10^{-24}$	$4.72 \cdot 10^{-24}$	1.98	$(2.36 \pm 0.1) \cdot 10^{-19}$	—
V_0	$(4 \pm 0.1) \cdot 10^{-5}$	$4.16 \cdot 10^{-4}$	—	$(9.5 \pm 0.1) \cdot 10^{-4}$	—
τ_0, s	$3.06 \cdot 10^{-12}$	$3.29 \cdot 10^{-12}$	—	$1.77 \cdot 10^{-11}$	—
$B_F, \text{cm}^{-1} \cdot \text{s}^{0.92}$	$2.95 \cdot 10^{-9}$	—	—	—	—
$B_B, \text{cm}^{-1} \cdot \text{s}^{0.94}$	—	—	—	$1.29 \cdot 10^{-8}$	—

Note. The experimental data were analyzed on a computer using a program which made it possible to determine the critical exponents without preliminary separation of the background absorption or of the normal change in the velocity.

specific heat at constant pressure. Similar results are reported also for magnetically ordered crystals in Refs. 2, 20, and 21. However, it is not possible to estimate V_0 within the framework adopted in these investigations, so that we shall deduce V_0 and n_i theoretically using the familiar expression for the relaxation time ($\omega\tau \ll 1$) given in Ref. 3:

$$\tau = \frac{v}{\omega^2} \frac{\Delta\alpha_k}{|(\Delta v/v)_k|} = \tau_0 t^{-x}, \quad (6)$$

which together with Eqs. (4) and (5) gives

$$(\Delta v/v)_k = vB = V_0 t^{2\alpha-2+3\nu}. \quad (7)$$

Moreover, Eqs. (2)–(7) allow us to obtain the following relationships between the static and dynamic critical exponents:

$$\eta_i = x - 2\alpha + 2 - 3\nu = x - \alpha, \quad (8)$$

$$n_i = 2 - 2\alpha - 3\nu = -\alpha. \quad (9)$$

One of the main parameters describing the critical dynamics is the relaxation time which can be calculated from the experimental data on $\Delta\alpha_k$ and $(\Delta v/v)_k$. In the paramagnetic phase we can use for this purpose Eq. (6), whereas in the magnetically ordered phase the dependence $\tau(t)$ can be deduced from the shift of the maximum of α_k on increase in the frequency, which according to the Landau–Khalatnikov theory of relaxation¹ is governed by the condition $\omega\tau = 1$.

The temperature dependences of the relaxation time calculated from $\Delta\alpha_k$ and $(\Delta v/v)_k$ showed that the experimental points fit two straight lines representing power laws of the type described by Eq. (6) with identical critical exponents for $T > T_c$ and $T < T_c$, but with different critical amplitudes (Fig. 4). It should be pointed out that the condition of validity of Eq. (6) is satisfied by gadolinium at reduced temperatures $t \geq 10^{-3}$ ($\omega/2\pi = 30$ MHz, $\omega\tau = 0.39$) and the absolute values of τ agree with the results reported by other authors. For example, according to Ref. 3, it is found that $\tau = 5.31 \times 10^{-10}$ s at $t = 3 \times 10^{-3}$, whereas our data obtained at the same reduced temperature give $\tau = 5.8 \times 10^{-10}$ s (Fig. 4).

The final results of an analysis of the experimental data on the temperature and frequency dependences of α_k and $\Delta v/v_0$, carried out by the method of least squares using a standard program, are presented in Table I. This table lists

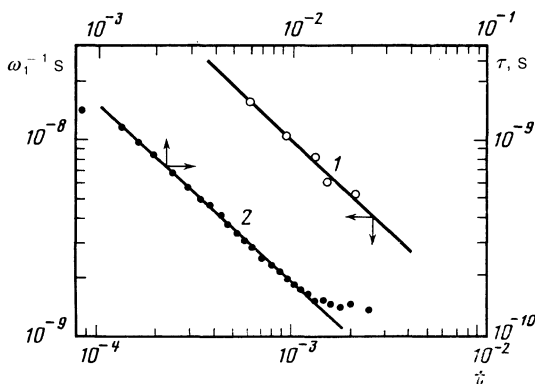


FIG. 4. Temperature dependences of the relaxation time (10 MHz): 1) $T < T_c$; 2) $T > T_c$.

also the numerical values of the critical exponents and amplitudes obtained using the theory of interacting modes. The theoretical critical amplitudes were calculated employing the following values of the parameters occurring in Eqs. (5) and (7): $v_0 = 2.9 \times 10^5$ cm/s, $\rho = 7.9$ g/cm³, $\xi = a/\sqrt{6}$, which is an estimate obtained by applying the molecular field theory to hexagonal crystals with a shortest distance $a = 3.636$ Å between the atoms²: $dT_c/dP = 1.63 \times 10^{-3}$ kbar⁻¹ was taken from Ref. 22, whereas τ_0 was estimated—as in Ref. 2—assuming that the energy of the critical fluctuations is equal to the exchange integral. The experimental and theoretical values of the dynamic critical exponent z given in Table I were calculated from $z = (\eta_i - n_i)/\nu$, which follows from Eqs. (8) and (9), and also using the familiar relationship $z = x/\nu$ (Refs. 23 and 24).

First of all, we must point out that our values of the critical exponents η_i and n_i differ from the results of other authors: $\eta_i = 1.2 \pm 0.1$ (Ref. 15), $\eta_i = 1.63 \pm 0.1$ (Ref. 3), $\eta_i = 1.8 \pm 0.2$ (Ref. 17), and $n_i = 0$ (logarithmic dependence).³ This difference is clearly associated with determination of the Curie point, assumed in Refs. 3, 17, and to be equal to the temperature at which an absorption peak was observed, whereas we found the Curie point from independent magnetic measurements in weak magnetic fields below the anisotropy and demagnetization fields.

A comparison of the experimental and theoretical critical exponents and amplitudes yielded three important results characterizing the critical behavior of gadolinium in the hydrodynamic range. Firstly, in spite of the considerable discrepancies between the theory and experiment in the case of some of the critical exponents, it follows from the theory of interacting modes that the characteristic features of the critical propagation of ultrasonic waves can be described by the isotropic Heisenberg model with a nonconserved order parameter (total spin of the system). The law of conservation of the total spin breaks down here because of the isotropic dipole interactions (these topics will be discussed in detail later). Secondly, the critical amplitudes below T_c increase on increase in T_c , which implies that the magnetically ordered phase exhibits not only the fluctuation mechanism, but also other mechanisms of the anomalous behavior of α_k and $\Delta v/v_0$, the most important of which is the Landau–Khalatnikov relaxation mechanism.¹ Thirdly, the equality of the critical exponents $x^+ = x^-$ and $z^+ = z^-$ confirms that the main dynamic scaling hypothesis²³⁻²⁶ applies to gadolinium: according to this hypothesis, the characteristic frequencies of the fluctuations are a function of one variable $\omega\tau$ throughout the whole critical range.

4. DYNAMIC SCALING CONCEPTS IN ABSORPTION

It follows from experimental and theoretical investigations⁷⁻¹⁰ that in the case of magnetically ordered crystals with the long-range exchange interaction the overall absorption of ultrasonic waves is due to both the relaxation processes and the fluctuations of the order parameter. Below T_c both mechanisms are active, whereas in the paramagnetic phase only the fluctuation mechanism is retained, i.e.,

$$\Delta\alpha_k^+ = \alpha_F^+, \quad \Delta\alpha_k^- = \alpha_F^- + \alpha_R, \quad (10)$$

where α_F and α_R are the fluctuation and relaxation contributions to the critical absorption $\Delta\alpha_k$.

According to the dynamic scaling concepts and the theory of interacting modes,^{5,8} throughout the critical region the value of α_F is described by the scaling function of the variable $\omega\tau$:

$$\alpha_F^\pm = B_F^\pm \omega^{1+y^\pm} f^\pm(\omega\tau^\pm), \quad (11)$$

where $f_F^\pm(\omega\tau^\pm)$ is the scaling function, the specific form of which is not given by the theory, but the experimental results are usually analyzed on the assumption that f_F^\pm has the Lorentzian profile⁷⁻¹⁰:

$$f_F^\pm = \frac{(\omega\tau^\pm)^{1-y^\pm}}{C + (\omega\tau^\pm)^{1-y^\pm}}. \quad (12)$$

Here, C is a positive constant; $y^+ = \alpha/x^+$; $B_F^+ = B_F^-$; $\alpha_F^+ \neq \alpha_F^-$ because $\tau_0^+ \neq \tau_0^-$. We can easily see that

$$\alpha_F^-(t) = \alpha_F^+(qt), \quad q = (\tau_0^-/\tau_0^+)^{1/x}. \quad (13)$$

Hence, using Eq. (10), we can separate α_F and α_R in the magnetically ordered phase. It has been shown that not only α_F , but also α_R can be described by a scaling equation⁸

$$\alpha_R(t) = \Delta\alpha^-(t) - \alpha_F^+(qt) = B_R \omega^{1+y^-} f_R(\omega\tau^-), \quad (14)$$

where

$$f = \omega\tau^- / [1 + (\omega\tau^-)^2]. \quad (15)$$

A similar result follows also from the Landau-Khalatnikov theory if we assume that $y^- = 0$.

In an experimental check of the scaling equations (11) and (14) we have to determine y^+ , y^- , and C . We can use the limits of the functions f_F and f_R at $T = T_c$ and $T = T_{max}$. Since at these temperatures we have $\omega\tau \rightarrow \infty$ ($T = T_c$) and $\omega\tau = 1$ ($T = T_{max}$), it follows that

$$\alpha_F^+ = B_F \omega^{1+y^+} = \alpha_c, \quad T = T_c, \quad (16)$$

$$\alpha_R = 1/2 B_R \omega^{1+y^-} = \alpha_{max}, \quad T = T_{max}. \quad (17)$$

Figure 5 shows the frequency dependence of $\Delta\alpha_k$ for gadolinium at these temperatures: it shows that the experimental points fit the dependences (16) and (17) in the frequency range 5–30 MHz. The value of $\Delta\alpha_k$ corresponding to

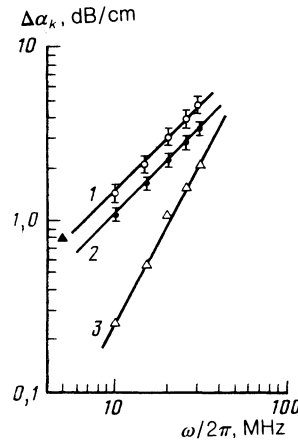


FIG. 5. Frequency dependences of the critical absorption at various temperatures: 1) $T = T_{max}$; 2) $T = T_c$; 3) $t = 10^{-3}$.

5 MHz was taken from Ref. 17. As expected, within the limits of the experimental error we have $y^+ = y^-$ (Table I).

An estimate of the constant C was made using the experimental values of $\Delta\alpha_k$ at $T = T_c$ and assuming that $\omega\tau^+ = 1$. Equation (12) and the temperature dependence of $\Delta\alpha_k$ yield $\alpha_F^+/\alpha_c = 1/(C+1) = 0.75$ and $C = 0.33$.

Using the above estimates of y^+ and C , we analyzed the experimental data for α_F^+ in the frequency range 10–30 MHz by the least-squares method. The fitting parameters were x^+ , y^+ , and C , selected to be close to 0.9, -0.08 , and 0.33, respectively. The best agreement between the experimental points and Eq. (11), represented by the dashed curve in Fig. 6, was obtained for $x^+ = 0.91$, $y^+ = -0.12$, and $C = 0.30$. These values were in reasonable agreement with x^+ , y^+ , and C obtained by direct determination (Figs. 4 and 5, and Table I).

The relaxation contribution to the anomalous absorption α_R was separated using Eqs. (13) and (14) with $q = 6.8$. The scaling function for α_R obtained on a double logarithmic scale is also shown in Fig. 6. Different shapes of the symbols are used in Fig. 6 to denote different frequencies, and the dashed curve obeys Eq. (14) with $y^- = -0.12$. The

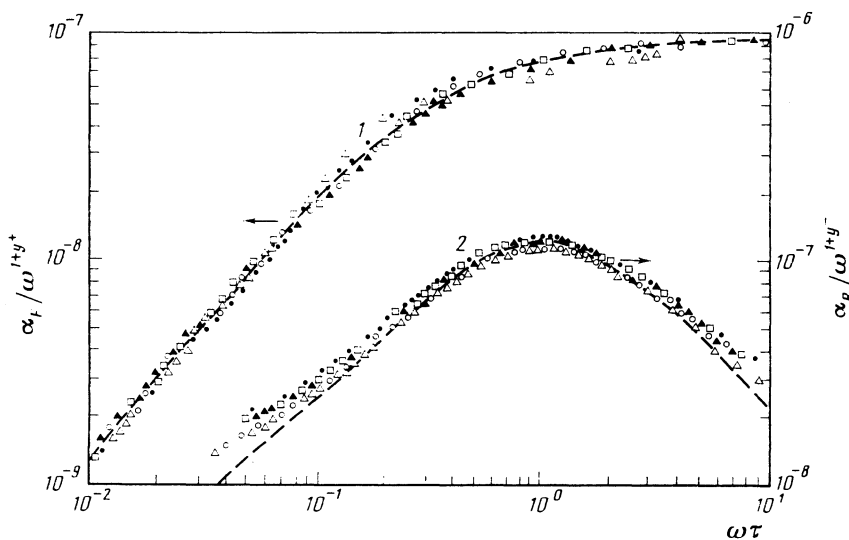


FIG. 6. Scaling equations for the fluctuation (curve 1, $T > T_c$) and relaxation (curve 2, $T < T_c$) contributions to the anomalous absorption. The different symbols for the different frequencies are the same as in Fig. 2.

numerical values of the other parameters needed in the calculation are listed in Table I. It is clear from Fig. 6 that the experimental frequency and temperature dependences of α_R obtained in the range 10^{-1} –7.0 of $\omega\tau$ fit quite well Eq. (14). The maximum of α_R does not occur at $\omega\tau = 1$, as expected on the basis of the Landau–Khalatnikov theory, but is shifted slightly toward higher values of $\omega\tau$. This shift, amounting to $\omega\tau = 1 - y^- = 1.12$, is due to the singularity of the critical amplitude of the absorption coefficient, which in turn shifts the maximum of $\Delta\alpha_k$ and the minimum of $\Delta\nu/\nu$ relative to one another.

5. DIPOLE NATURE OF THE CRITICAL DYNAMICS OF GADOLINIUM

It is clear from Table I that the experimental values of the critical exponents differ considerably from the theoretical exponents expected for the Ising ($n = 1$) and Heisenberg ($n = 3$) models. In particular, the critical exponent η_i is much less than the value which follows from Eq. (8) for the systems with $n = 3$, but it is close to the value for systems with $n = 1$. On the other hand, the sign and the absolute magnitude of n_i agree best with the Heisenberg model. The critical exponents x and z are far from the values predicted by the theory in the case of model crystals. This is true even in the case of ferromagnets with a nonconserved spin, although the agreement between the theory and experiment then improves (see, for example, the second row for z in Table I). The anisotropic and dipole interactions of gadolinium may result in breakdown of the law of conservation of the total spin.

An estimate of the influence of the anisotropic and dipole forces on the critical behavior of ferromagnets can be obtained from the relevant crossover temperatures: $t_A = (g_A)^{1/\Phi_A}$ and $t_d = (g_d)^{1/\Phi_d}$, where g_A and g_d are the parameters that represent the strength of the anisotropic and dipole interactions; $\Phi_A = 1.25$ and $\Phi_d = 1.37$ are the crossover exponents. Numerical estimates indicate that $t_A = 8.31 \times 10^{-4}$ and $t_d = 8.0 \times 10^{-3}$, so that in the investigated range of temperatures we would expect the crossover to the dipole behavior rather than to the anisotropic behavior. This conclusion is supported also by the anisotropy parameter $g_A = 5 \times 10^{-5}$ given in Ref. 2 and by the results of experimental investigation of the static critical behavior of gadolinium.^{3,13} In particular, an analysis of the published values of the static critical exponents and also of our data (which will be published in future) show that practically the whole of the investigated temperature range corresponds to the dipole critical behavior. For example, the experimental values of the susceptibility indicate that the condition $4\pi\chi = 1$, that demarcates the dipole ($4\pi\chi \gg 1$) and exchange ($4\pi\chi \ll 1$), critical regions²⁶ is satisfied at $t = 2.62 \times 10^{-2}$. Consequently, in an analysis of the experimental data on the critical propagation of ultrasonic waves in gadolinium we have to allow for the influence of isotropic dipole forces. However, this topic has not yet been tackled theoretically and we shall therefore use a general theory of the influence of the dipole forces on the critical dynamics of ferromagnets, which was presented in the reviews of Maleev.^{26–28} According to this theory, ferromagnets can have the normal and anomalous (hard) dynamics. The former is characterized by the critical exponents $x = (2 - \eta)\nu$ and $z = 2 - \eta$. Calculations carried out by Teitel'baum²⁹ using

the ε expansion gave a similar result: $z = 2 + 0.94\eta$. The hard dipole dynamics, due to the interaction of fluctuations longitudinal and transverse to the momentum, leads to $z = (5 - \eta)/2 - 1/\nu$ and $x = [(5 - \eta)/2 - 1/\nu]\nu$. If we use the values of the Fisher critical exponents η and ν obtained by the ε expansion,²³ we obtain $z = 1.977$ and $x = 1.368$ for the normal dynamics, whereas for the harder dynamics we obtain $z = 1.043$ and $x = 0.722$. The experimental values of z and x lie between them and we cannot give preference to any one of the dynamics. However, it is known that in the case of ferromagnets an increase in S reduces the region of the hard dynamics²⁷ and in the case of gadolinium ($S = 7/2$) it is more realistic to expect the normal dynamics. However, this still leaves large discrepancies between the theoretical and experimental values of z and x , which in our opinion are due to the following circumstances. Firstly, in the exchange region the dipole forces give rise to what is known as the Huber damping,³⁰ which is characterized by a reduction in the relaxation time in the limit $T \rightarrow T_c$: $\tau \sim t$. Since in the case of gadolinium we have $4\pi\chi = 1$ already at $t = 2.62 \times 10^{-2}$, the influence of the Huber damping on the temperature dependence of $\Delta\alpha_k$ should be slight. Nevertheless, it may reduce the critical exponent η_i and, consequently, both x and z . Secondly, practically the whole of the investigated range of temperatures corresponds to the crossover region in the case of gadolinium, so that by analogy with the static critical exponents, the experiments give not the asymptotic but the effective values of x and z . For example, the critical exponent γ of the static susceptibility, which can be used to describe η_i ($\eta_i = \gamma - \alpha$ applies in the case of the normal dynamics and $\eta_i = [\gamma(5 - \eta)/(2 - \eta)] - 1 - \alpha$ corresponds to the hard dynamics) is at least 10% less than the asymptotic value.³¹ It is not permissible to ignore this circumstance in discussing the experimental results. We must also bear in mind that the above discussion is purely qualitative because of the absence of theoretical investigations of the critical propagation of ultrasonic waves in the crossover region.

Finally, we must mention that investigations carried out by other methods^{32–35} also support the hypothesis of a considerable influence of the dipole forces on the critical dynamics of gadolinium. This dynamics was investigated by the ESR method at 9 GHz (Ref. 32) and it was found that the width of the resonance line in the paramagnetic phase passed through a maximum typical of dipole magnetic materials.³⁶ Moreover, the dependence $\Delta H T \chi_{\perp} = f(\chi_{\parallel})$ (where χ_{\parallel} and χ_{\perp} are the longitudinal and transverse—relative to the field—values of the static susceptibility, and ΔH is the line width) obeys a power law with an exponent of 0.84, whereas in the Huber damping case this exponent should be 7/4. An investigation of the critical dynamics of gadolinium by the method of perturbed γ – γ angular correlations^{33–35} shows that the fluctuations are isotropic, at least in the range $t \gg 3 \times 10^{-3}$ and the dynamic critical exponent z found from the autocorrelation time has the following values: 1.3–1.52 (Ref. 33), 1.786 (Ref. 34), and 1.73 (Ref. 35), which are in fairly satisfactory agreement with our data and with the theoretical values for the normal dipole dynamics.

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