Static critical behavior of gadolinium

Kh. K. Aliev, I. K. Kamilov, and A. M. Omarov

V. I. Lenin Dagestan State University, Makhachkala

Measurements of the magnetization and differential susceptibility of two gadolinium single crystals were carried out in magnetic fields $H = 0$–5 kOe at reduced temperatures $t = 10^{-4}$–10^{-2}. The results of these measurements and an analysis of the published data demonstrated that static critical behavior of gadolinium corresponded to the crossover region and the influence of the uniaxial anisotropy was significant at reduced temperatures $t \leqslant 8.3 \times 10^{-4}$.

1. INTRODUCTION

Gadolinium is a rare-earth metal which crystallizes in the hexagonal close-packed structure and exhibits simple ferromagnetic ordering at temperatures $250 < T < T_c \approx 293$ K. Its ferromagnetism is due to the Gd^{3+} ions which are in the $^8S_{7/2}$ state. The spherical distribution of the electron density and the absence of an orbital momentum suggest that the magnetocrystalline anisotropy energy of gadolinium should be considerably less than for other rare-earth metals.[1,2] This has been confirmed by magnetic[2] and neutron-diffraction[3] investigations and it has been established that the easy magnetization axis coincides with the hexagonal axis and that the anisotropy constants K_1 and K_2 depend in a complex manner on the temperature of a sample and on the applied magnetic field H. An analysis of the influence of H and T on K_1 and K_2, carried out by Belov et al.[2] and by Yang,[4] demonstrated that the magnetocrystalline anisotropy is due to single-ion and two-ion mechanisms and that the role of the latter increases on approach to $T \to T_c$. Consequently, the anisotropy constant K_1 of gadolinium does not decrease in the limit $T \to T_c$, as expected from the magnetocrystalline anisotropy theory,[1] but increases and in the vicinity of T_c passes through a maximum remaining finite in a wide range of temperatures above T_c (Refs. 2 and 4). Investigations of quasielastic neutron scattering not only confirm these results, but they show that the magnetocrystalline anisotropy observed in the paramagnetic phase is due to the uniaxial anisotropy of the short-range magnetic order.[5]

These experimental observations and an analysis of the nature of the magnetic ordering thus demonstrate that gadolinium is a uniaxial anisotropic magnetic material and, consequently, its critical behavior may be classified as of the Ising type. However, this hypothesis is not supported by the results of experimental studies of the critical behavior of gadolinium.

The static critical behavior of gadolinium had been investigated on many occasions.[6–19] Measurements of the specific heat[6,7] thermal expansion,[8,9] magnetic properties,[10–16] and Mössbauer spectra[17] carried out on single-crystal and polycrystalline samples of gadolinium were used to determine the whole set of critical exponents $\alpha^{\pm}$, $a^{\pm}$, β, $\gamma^{\pm}$ and δ (we shall use the generally accepted notation for the critical exponents and employ the plus and minus signs for $T > T_c$ and $T < T_c$, respectively). Table I gives the experimental values of the critical exponents selected from the published data. Preference was given to those investigations which were carried out on crystals with fewest defects and in

a sufficiently wide range of reduced temperatures $10^{-4} \leqslant t \leqslant 10^{-2}$, where $t = |T - T_c|/T_c$. For comparison, Table II lists the most accurate (at present) critical exponents calculated using the renormalization group in the ε expansion for three-dimensional Heisenberg ($n = 3$), XY ($n = 2$), Ising ($n = 1$) including ε^5 (Ref. 20), and isotropic dipole including ε^2 (Ref. 21) magnetic materials.

The critical exponents of the specific heat $\alpha^{\pm}$ (Refs. 6 and 7) and of the thermal expansion $a^{\pm}$ (Refs. 8 and 9) should be, according to the theoretical predictions, equal to one another, showing that gadolinium is either a Heisenberg or an isotropic dipole magnet, since $\alpha^{\pm}$ and $a^{\pm}$ assume negative values close to the theoretical exponents. In the case of the critical exponents β, γ, and δ, we should note that all the investigations of the temperature dependences of the spontaneous magnetization M_s, characterized by the critical exponent β (see Table I), also suggest that gadolinium is a Heisenberg or an isotropic dipole magnetic material: the values of γ are closer to those predicted by the Ising model, whereas δ does not agree not only with the fluctuation theory but also with the mean field theory. An analysis of the experimental results published in Refs. 6–17 shows that this disagreement is not due to the characteristic features of the critical behavior of gadolinium, but is the result of the method adopted to determine the critical exponents. In fact, in the majority of the investigations for which the results are given in Table I the critical exponents β, γ, and δ were found by fitting the experimental M-H-T data to the scaling equation of state for the magnetization, which implies that the scaling law $\gamma = \beta(\delta - 1)$ is obeyed. Obviously, the critical exponents determined in this way should satisfy the scaling laws containing only β, γ, and δ, whereas their separate values may not correspond to the true asymptotic critical behavior since in practically any experiment the asymptotic critical region is never reached. Therefore, the experiments yield not the asymptotic but the effective critical exponents, which may differ significantly from the asymptotic values because real crystals exhibit additional interactions that perturb the initial critical behavior. For example, isotropic dipole interactions in Heisenberg magnetic materials give rise to a dependence of γ on the reduced temperature t. Moreover, in the crossover region the exponent γ passes through a minimum and the difference between the values of γ and $\gamma_{\rm eff}$ may reach 10% or more.[22]

Therefore, for the reasons given above the published experimental values of the critical exponents do not allow us to determine the class of universality of the static critical behavior of gadolinium. We therefore carried out an investi-

TABLE I. Experimental critical exponents (CE) of gadolinium (published data).

CE	T_c, K	Interval of t	Values of CE	Comments	Ref.
α^+	291.31	10^{-3}-10^{-1}	-0.09(5) *	single crystal	[6]
α^-		»	-0.32(5)	$\rho_0=19$	
α^+		»	-0.20(2) }	assuming that	[7]
α^-			-0.20(2) }	$\alpha^+=\alpha^-$	
a^+	293.358	$2.5\cdot10^{-4}$-$8.6\cdot10^{-2}$	-0.06(3)	single crystal	[8]
a^-	293.425	$1.3\cdot10^{-3}$-$6.6\cdot10^{-2}$	-0.121(2)	$\rho_0=300$	
a^+	293.62	$7\cdot10^{-4}$-$4\cdot10^{-2}$	-0.25	single crystal	[9]
a^-		»	-0.32	$\rho_0=227$	
β		$1.7\cdot10^{-3}$-$6\cdot10^{-1}$	0.385	polycryst. sample	[10]
β		$2.7\cdot10^{-2}$-$1.9\cdot10^{-1}$	0.39 }	single crystal	[11]
β		$t<2.7\cdot10^{-2}$	>0.39 }		
γ	292.5	$2.2\cdot10^{-3}$-0,2	1.3(1)	reanalysis of data of Ref. 13	[12]
γ	291.1	$9.9\cdot10^{-3}$-$3.7\cdot10^{-2}$	1.24(3)	polycryst. sample	[14]
β	292.5	$t>2\cdot10^{-3}$	0.37(1) }	analysis of data of Ref. 13 using equation of state	[15]
γ			1.25(10)		
δ			4.39(10) }		
β	293.3	$t>4\cdot10^{-3}$	0.381(15) }	deduced from equation of state	[16]
γ			1.196(3)		
δ			3.615(15) }		
β	291.85	$t>10^{-3}$	0.399(16) }	Mössbauer spectroscopy	[17]
β	291.75	10^{-4}-10^{-3}	0.362(8) }		

*The numbers in parentheses denote the experimental error in the critical exponents: $\rho_0 = \rho(300\ K)/\rho(4.2\ K)$.

gation of the static and dynamic aspects of the critical behavior of gadolinium. The present paper reports the results of a study of the magnetic properties of this element and an attempt to determine the class of universality of the static critical behavior on the basis of these properties.

2. EXPERIMENTAL METHOD AND RESULTS

In contrast to the previous investigations[10-16] of magnetic properties of gadolinium, in which the critical exponents were estimated solely from the magnetization measurements, we carried out experiments in which we determined β, γ, and δ independently of one another. This was done by recording the temperature and field dependences of the magnetization M using a vibration magnetometer[23] and of the susceptibility χ using a modulation magnetometer,[24] which made it possible to carry out measurements in near-zero magnetic fields (~0.1 Oe). The temperature dependences of M and χ were recorded under quasistatic thermal conditions. The rate of change of temperature was selected in the course of measurements on the basis of a ther-

mal hysteresis and it did not exceed 0.5 K/h. The isotherms of the field dependences of M and χ were recorded keeping the temperature of a sample constant to within 5×10^{-4} K in magnetic fields up to 5 kOe, which were created by an electromagnet where the field homogeneity was within 5×10^{-4} cm^{-1}. Copper-constantan thermocouples, calibrated using a standard platinum resistance thermometer, were used to determine the temperature of a sample and as sensors used to control the temperature.

Our measurements of M and χ were carried out on two gadolinium single crystals differing from one another by the Curie points: $T_c = 293.575$ K for Gd I and $T_c = 293.370$ K for Gd II; these crystals were oriented with the aid of the Laue diffraction patterns to within 0.5°. Orientation and mechanical finishing of cylindrical crystals were followed by chemical etching in order to remove the cold-worked layer and by annealing at $T = 900$ K for 4 h. X-ray and electron diffraction investigations, as well as the high values of T_c (Table I), demonstrated that the investigated gadolinium single crystals were of high quality.

The temperature dependences of M obtained for gado-

TABLE II. Asymptotic critical exponents (CE) of three-dimensional model and dipole crystals*.

CE	n			Isotropic dipole magnetic materials
	1	2	3	
α	0.108±0.009	-0.012±0.014	-0.126±0.028	-0.135
β	0.3265±0.0025	0.3485±0.0035	0.368±0.004	0.381
γ	1.239±0.004	1.315±0.007	1.39±0.01	1.372
δ	4.795±0.017	4.773±0.017	4.777±0.014	4.454
Δ	0.511±0.030	0.537±0.030	0.561±0.035	0.425
R_χ	1.6	1.33	1.33	1.33

*Values of the critical exponents β and γ were taken from Ref. 20, whereas α and δ were calculated using the relationships $2 = \alpha + 2\beta + \gamma$ and $\gamma = \beta(\delta - 1)$. The numerical values of the critical exponents for isotropic dipole magnetic materials were based on the calculations of Ref. 21.

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M, rel. units
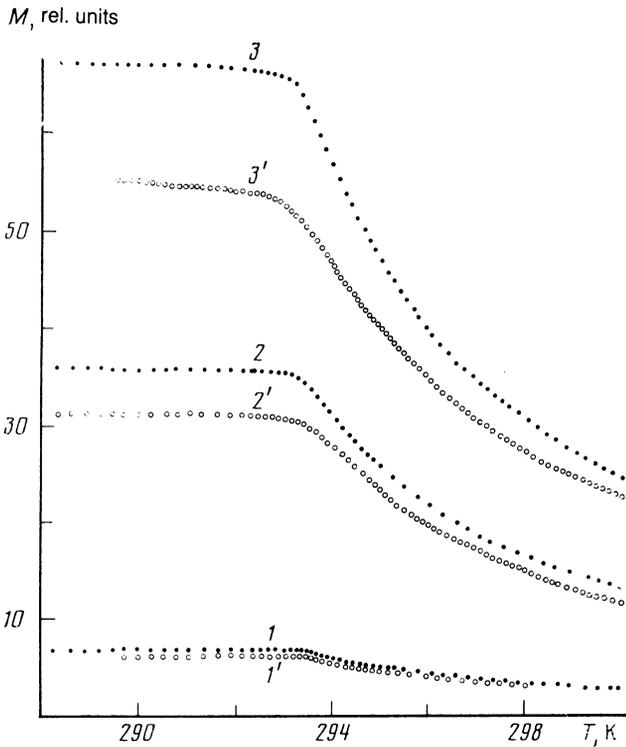
FIG. 1. Temperature dependences of the magnetization of gadolinium ($N_d = 0.452$) along the easy (●) and difficult (○) magnetization axes: 1), 1') 10.3 Oe; 2), 2') 54.1 Oe; 3), 3') 100.5 Oe.

linium along the easy and difficult magnetization axes in weak magnetic fields ($H < H_a$ and $H < H_d$, where H_a and H_d are the anisotropy and demagnetization fields, respectively) were obtained for a sample with identical demagnetization factors N_d along the easy and difficult magnetization axes (Fig. 1). It is clear from this figure that the magnetization and the easy axis remained constant in the ordered phase right up to T_c, and that there was an inflection at the, Curie point, and this inflection shifted toward lower temperatures on increase in H. The value of M for the difficult magnetization axis at temperatures $T < T_c$ decreased on increase in temperature and it was found that an increase of H or a reduction in N_d increased the degree of the change of M with T. As in the case of the easy magnetization axis, an inflection was observed in the dependence $M(T)$. Moreover, it is clear from Fig. 1 that the anisotropy of M, retained in the paramagnetic phase, increased in H.

The characteristic features of the temperature dependence of M in a magnetic field, typical of the easy and difficult magnetization axes, can be explained on the basis of the Landau theory, which predicts that anisotropic crystals of finite dimensions should exhibit second-order phase transitions of two types.[18] In a magnetic field applied along the magnetization axis a transition takes place from an inhomogeneously magnetized (polydomain) to a homogeneously magnetized (single-domain) state. The characteristic signs of this transition are constancy of the magnetization M_z along the field in a wide range of temperatures, a linear dependence of M_z on H at temperatures $T < T_c(H)$ and in fields $H < H_d$, where $T_c(H)$ is the temperature of the transition in a magnetic field, and the presence of an inflection or a kink in the case of the $M(T)$ curve at $T = T_c(H)$ or at

$H = H_d = N_d M_s$. However, in the case when H is parallel to the difficult magnetization axis there is a transition from a state in which the magnetization component M_z along the easy axis does not vanish to a state with $M_z = 0$. The magnetization measured along the field then exhibits the same behavior as M_z in the case when the field H is parallel to the easy axis. However, in contrast to the easy axis, when we have

$$M_z = H/N_d, \tag{1}$$

the magnetization in a field H parallel to the difficult axis is

$$M_x = H/(N_d + k), \tag{2}$$

and the magnetization measured in a field can increase or decrease when T is increased. The sign of the temperature coefficient of M_x depends on the corresponding sign of the thermodynamic coefficient of the anisotropy constant $k = 2K_1/M_s^2$. For any relationship between N_d and k the value of M_x remains constant only if $K_1 \propto M_s^2$. Otherwise, M_x varies with temperature.

Therefore, in view of the weak dependence of N_d on T, the reduction in M_x of gadolinium observed on increase in temperature can be explained by an increase in k in the limit $T \to T_c$. This hypothesis is supported also by the temperature dependences of M_x, recorded for a sample with a smaller demagnetization factor and characterized by an increase in the temperature coefficient of the magnetization. It should also be mentioned that an increase in k in the limit $T \to T_c$ is observed also when K_1 is determined directly in strong magnetic fields.[2,13]

Measurements of the differential susceptibility not only confirmed the characteristic features of the M-H-T data, but also revealed additional anomalies. In particular, the value of χ measured parallel to the easy magnetization axis decreased on increase in T (Fig. 2) and it remained constant only in the direct vicinity of T_c, whereas χ parallel to the
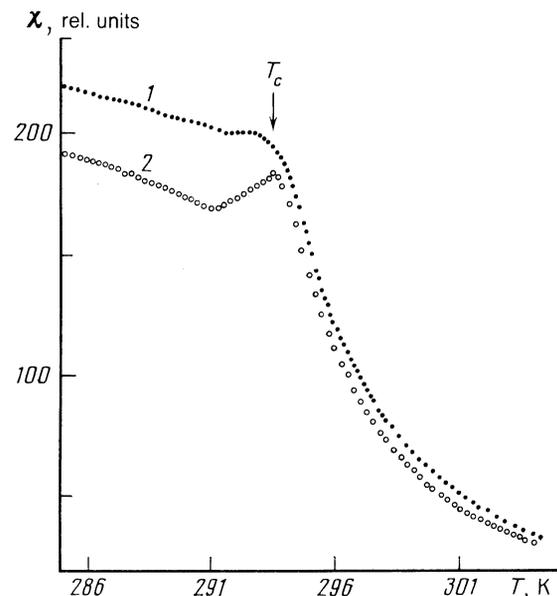
FIG. 2. Temperature dependences of the differential susceptibility of gadolinium in fields $H = 0$ and $H_- = 0.1$ Oe: 1) easy magnetization axis ($N_d = 0.358$); 2) difficult magnetization ($N_d = 0.321$).

difficult magnetization axis passed through a maximum at $T = T_c(0)$ which was retained up to $H = 650$ Oe (Fig. 1 in Ref. 19). In fields $H > 650$ Oe the temperature dependences were the same for the easy and difficult magnetization axes.

In contrast to the M-H-T data, we found that the χ-H-T results could not be explained by the Landau theory, according to which the susceptibility should remain constant in the magnetically ordered phase and it should be equal to $1/N_d$ and $1/(N_d + k)$ for the easy and difficult magnetization axes, respectively. We found that along both axes of the gadolinium crystal the value of χ decreased at temperatures down to $T = 291.5$ K which was clearly due to modification of domain walls. In fact, in the case of magnetically uniaxial crystals with K_1 that does not vanish at $T = T_c$ there should be a transition from a Bloch to a linear wall.[25,26] This type of wall is characterized by a reversal of the magnetic moment direction only when the modulus of the magnetic moment passes through zero. Investigations of the stability of a wall in a uniaxial medium have shown[25] that transition to such a medium should be expected near T_c under the condition $M_s^2/8\chi_p = K_1$ (here, χ_p is the paraprocess susceptibility). If we assume that M_s and χ_p are described by simple power laws with the critical exponents $\beta = 0.38$ and $\gamma = 1.33$ and that K_1 depends weakly on temperature in the vicinity of T_c, we find that this condition is satisfied by gadolinium at $T = 291.8$ K. It is clear from Fig. 2 that this temperature is in good agreement with the experimental results. The absence of anomalies in the case of the $M(T)$ curves is due to the fact that theoretical estimates indicate that the change in the magnetization on transition from a Bloch to a linear wall does not exceed 1% (Ref. 25).

It follows therefore that the anomalous behavior of χ of gadolinium near T_c can be due to crossover from the Heisenberg (Bloch wall) to the Ising (linear wall) critical behavior which, in the opinion of the authors of Ref. 10, was discovered by them first when measuring the power of the Barkhausen noise of polycrystalline gadolinium.

3. DETERMINATION OF CRITICAL EXPONENTS AND AMPLITUDES

We determined the critical exponents and amplitudes from the experimental data of the type presented in Figs. 1–3. The spontaneous magnetization and the Curie point were deduced from the M-H-T data using the kink method.[18] Clearly, an inflection or kink of the $M(T)$ curve, corresponding to the transition from the inhomogeneously to the homogeneously magnetized state, is observed in the given field when $M_s = H/N_d$ and, consequently, a determination
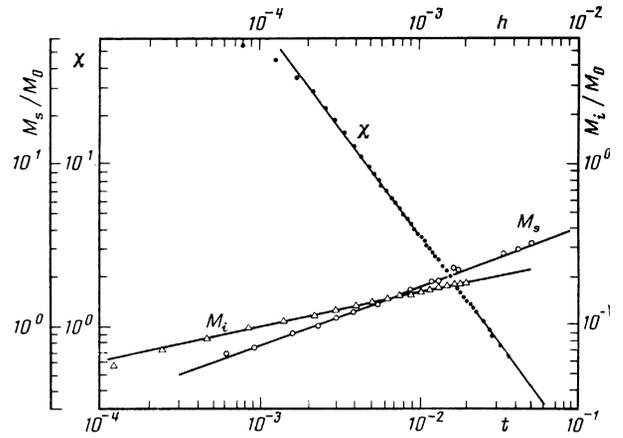
FIG. 3. Double logarithmic dependences of M_s/M_0 and χ on t, and of M_i/M_0 on $h = H_i/H_0$ (H_i is the internal magnetic field).

of the temperatures in which these inflections (kinks) are observed and of the corresponding values of H can be used to reconstruct the temperature dependence of M_s; one should know the numerical value of N_d. However, since N_d was independent of T, its value could be estimated using the M-H-T data from T_c or the susceptibility data obtained at $T = T_c$ where $\chi = 1/N_d$ is measured along the easy magnetization axis, and also by calculation from the geometric dimensions using familiar expressions.[1] In the case of gadolinium the values of N_d estimated by three different methods agreed to within 8%. It should be noted that in determination of β and γ there was no need to know the exact value of N_d so that its error did not affect the values of δ and of the critical amplitudes.

The experimental temperature and field dependences of M and χ, corrected for N_d and H_d, are plotted on a double logarithmic scale in Fig. 3. Here, different symbols represent the experimental results and the straight lines 1, 2, and 3 correspond to the following power laws:

$$M_s = M_0 Bt^\beta, \quad T \leqslant T_c, \tag{3}$$

$$\chi = \chi_0 \Gamma t^{-\gamma}, \quad T \geqslant T_c, \tag{4}$$

$$H = H_0 DM^\delta, \quad T = T_c \tag{5}$$

where $M_0 = 4\pi NSg\mu_B = 24\ 474$ G, $H_0 = k_B T_c/g\mu_B$ $S = 6.24 \times 10^5$ Oe, and $\chi_0 = M_0/H_0 = 3.92 \times 10^{-2}$, where N is the number of atoms in 1 cm^3, S is the spin number, g is the Landé factor, k_B is the Boltzmann constant, and μ_B is the Bohr magneton. An analysis of the experimental results in

TABLE III. Experimental values of effective and asymptotic critical exponents (CE) and of critical amplitudes (CA) of gadolinium

CE and CA	β	γ	δ	B	Γ
Effective	0.390±0.005	1.33±0.02	4.8±0.1	1.13±0.07	0.197±0.015
Asymptotic	0.375±0.005	1.39±0.02	4.8±0.1	1.08±0.07	0.145±0.015

CE and CA	D	R_χ	Δ	$^a\chi$	^aM
Effective	6.72±0.08	1.95±0.15	–	–	–
Asymptotic	6.72±0.08	1.21±0.15	0.55±0.05	0.73±0.03	0.27±0.03

accordance with Eqs. (3)–(5), which was carried out by us using the least-squares method and a standard program, demonstrated that the best agreement between them was obtained for the critical exponents and amplitudes listed in Table III. A comparison of the theoretical (Table II) and experimental (Table III) values of the critical exponents indicated that β and δ corresponded to isotropic Heisenberg and dipole magnetic materials. On the other hand, in the case of γ the discrepancy between the theory and experiment exceeded considerably the experimental error. In the case of the nonuniversal critical amplitudes the theoretically predicted universal ratio of these amplitudes, $R_\chi = \Gamma D B^{\delta-1}$, was much higher than the theoretical value for Heisenberg and dipole magnetic materials for which the renormalization-group theory and the ε expansion (in the first order with respect to ε) give $R_\chi = 1.33$ (Ref. 27).

The critical exponents and amplitudes found by fitting the data to simple power relationships (3)–(5) give not the asymptotic but the effective values,[22,28] which in most cases are smaller than the theoretical values. In particular, the exponent γ may be underestimated because of a number of factors, the most important of which is the difficulty of attainment of the asymptotic critical range in experiments. The value of γ is also influenced strongly by the occurrence of crossover from one critical behavior to another, particularly in the crossover region.[22] In any case, the correct determination of the critical exponents and amplitudes requires correction to the scaling results,[28,29] i.e., when an analysis is made of the experimental data, it is necessary to allow not only for the leading term, but also for the less singular terms. The first two terms of the correction to the scaling are of interest, but in the case of the majority of three-dimensional crystals the temperature dependences of the magnetization and susceptibility were analyzed in accordance with the expressions

$$M_s = M_0 B t^\beta (1 + a_M t^\Delta),\tag{6}$$

$$\chi = \chi_0 \Gamma t^{-\gamma}(1 + a_\chi t^\Delta),\tag{7}$$

where a_M and a_χ are the corresponding nonuniversal amplitudes of the correction, and Δ is the universal correction to the critical exponent. The correction procedure is not usually applied to a critical isotherm, because the asymptotic critical range is certainly reached here. The correction associated with the nonlinearity of the scaling fields gives rise to unimportant corrections to the dependence (5), as shown in Ref. 29.

The final results of the analysis of the experimental data in accordance with Eqs. (6) and (7) are presented in Table III, and the graphs demonstrating the validity of the adopted correction procedure are given in Fig. 4. It follows from Fig. 4, which gives the dependences of the critical amplitudes B and Γ on t on a double logarithmic scale, that the maximum scatter of the fitting parameters does not exceed the experimental error in the investigated temperature ranges $2 \times 10^{-3} \leqslant t \leqslant 3.7 \times 10^{-2}$ and $6 \times 10^{-4} \leqslant t \leqslant 5.1 \times 10^{-2}$ for χ and M, respectively.

It therefore follows that in the correction to the scaling all the critical exponents (including γ) and the universal ratio R_χ between the critical exponents of gadolinium are in good agreement with the theoretical values predicted for three-dimensional Heisenberg and dipole magnetic materi-
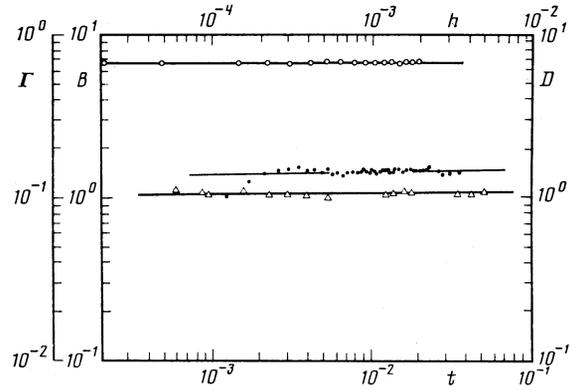
FIG. 4. Double logarithmic dependences of the critical amplitudes B ($\triangle$) and Γ ($\bullet$) on t, and of D ($\bigcirc$) on h.

als with isotropic interactions. However, the experimental values of the critical exponents and of R_χ are insufficient to distinguish between these universality classes. It is therefore necessary to estimate the influence of the dipole and other forces on the critical behavior of gadolinium.

4. DIPOLE NATURE OF THE CRITICAL BEHAVIOR OF GADOLINIUM

Following Refs. 30–33, we shall estimate the contribution of the dipole and anisotropic forces to the critical behavior of gadolinium from the crossover temperatures t_d and t_a found from the relationships

$$t_a = g_a^{1/\Phi_a} = \left(\frac{\Theta^c - \Theta^a}{\Theta^c} \right)^{1/\Phi_a},\tag{8}$$

$$t_d = g_d^{1/\Phi_d} = (C/T_c)^{1/\Phi_d},\tag{9}$$

where Φ_a and Φ_d are the crossover critical exponents for the anisotropic and dipole critical behavior, Θ^c and Θ^a are the paramagnetic Curie points for the c and a axes, and C is the Curie constant. The parameters g_a and g_d representing the anisotropic dipole interactions can be estimated using the molecular field theory.[1,2] These estimates indicate that $g_a = 1.41 \times 10^{-4}$ and $g_d = 1.35 \times 10^{-3}$ for $K_1 = 2 \times 10^5$ erg/cm^3, $N = 3.02 \times 10^{22}$ cm^{-3}, $S = 7/2$, and $T_c = 293.575$ K, which shows that $t_a = 8.31 \times 10^{-4}$ and $t_d = 8.02 \times 10^{-3}$ if $\Phi_a = 1.25$ (Ref. 30) and $\Phi_d = 1.37$ (Ref. 31). It is thus found that the temperature of the crossover to the Ising critical behavior is an order of magnitude closer to the Curie point than t_d and, therefore, the former cannot affect significantly the critical exponents. The temperature of the dipole crossover t_d occurs approximately in the middle of the investigated range of the reduced temperatures t and, consequently, the experimental situation corresponds to the dipole crossover region. This is supported also by the absolute value of χ (Fig. 3), which at $t = 2.62 \times 10^{-2}$ satisfies the condition $\chi = 1$ dividing the critical range into the exchange ($\chi \ll 1$) and dipole ($\chi \gg 1$) intervals.[32] Consequently, beginning from $t = 2.62 \times 10^{-2}$ the critical behavior of gadolinium is governed by isotropic dipole forces. In this case we can estimate the amplitude of the correction to the scaling, the shift of the Curie temperature of an isotropic Heisenberg magnetic material by the dipole forces $t_c = [T_c(g) - T_c(0)]/T_c(0)$, and the effective value of the

critical exponent γ_{eff} from the theory of the renormalization group and the ε expansion.[33]

In estimating these parameters we can use the scaling equations of state for the susceptibility and the ideas developed in Ref. 33 for dipole systems, according to which the renormalization mass r (representing the reciprocal of the susceptibility) obeys the crossover scaling function

$$r = C_0^{-\tau_H} t^{\tau_H} X^{-1}(y), \qquad (10)$$

where C_0 is a nonuniversal constant, $t = [T_c - T_c(0)]/T_c(0)$, γ_H is the Heisenberg critical exponent of χ, and $y = x/\dot{x}$ $(x = g_d/t^{\Phi_d}, \dot{x} = g_d/t_c^{\Phi_d})$. The asymptotic dipole critical behavior $(t \ll g_c^{1/\Phi_d})$ in Eq. (10) is described by a function $X(y)$, which has a singularity at $y = 1$. Expanding $X^{-1}(y)$ near the point $y = 1$ and bearing in mind that $1 - y \approx (\Phi_d/t_c)t$, we find that if $t \ll t_c$ Eq. (10) yields

$$\chi = A t^{-\gamma_d} [1 - B(1) \Phi_d^{\Delta_d} t_c^{-\Delta_d} t^{\Delta_d}], \qquad (11)$$

where $i = [T_c(g_d) - T_c(0)]/T_c(0)$; γ_d and Δ_d are the dipole critical exponent of χ and the correction to the scaling, respectively. The complete expression for the nonuniversal constant $B(1)$ is given in Ref. 22 and will not be repeated here because it is too cumbersome. A comparison of Eqs. (7) and (11) shows that

$$a_\chi = -B(1)(\Phi_d/t_c)^{\Delta_d}. \qquad (12)$$

If we substitute in Eq. (12) the values $B(1)$ and Φ_d calculated using the ε expansion, then in the case of three-dimensional crystals we find that

$$a_\chi = 0.099 t_c^{-\Delta_d} \qquad (13)$$

and at a temperature $t_c = 4.17 \times 10^{-3}$ (estimated from the molecular field theory) when $\Delta_d = 0.425$ (Ref. 21), we find that $a_\chi = 1.017$ for gadolinium. This result differs considerably from the experimental value (Table III). A more correct method for the determination of a_χ was proposed in Ref. 33. This method is based on a comparison of the expressions deduced from the macroscopic and microscopic theories of χ, and it makes it possible to express the susceptibility of a substance in terms of the principal parameters of the microscopic theory.

$$\chi = g_d/4\pi r. \qquad (14)$$

Obviously, we should have $g_d = r$ at some temperature $\dot{t}_m$ Using this condition in Eq. (10) and expressing C_0 in terms of t_c $(t_c = (C/d)g_d^{1/\gamma_H})$, as in Ref. 33, we obtain an expression

$$[(t_c + \dot{t}_m)/d t_c]^{\tau_H} = X(y_m) = X(t_c/(t_c + \dot{t}_m)),$$

from which we find that in the case of three-dimensional $(d = 3)$ crystals

$$t_c = 0.349 \dot{t}_m. \qquad (15)$$

Thus, in finding t_c from Eq. (15) we need simply to determine the temperature $\dot{t}_m$ at which the experimental value is $\chi = 1/4\pi$ $(\chi = 1$ in the SI system). As pointed out already, the susceptibility χ of gadolinium satisfies this condition at $t = 2.62 \times 10^{-2}$, (Fig. 3). It then follows from Eq. (15) that $t_c = 9.16 \times 10^{-3}$, which is approximately twice as
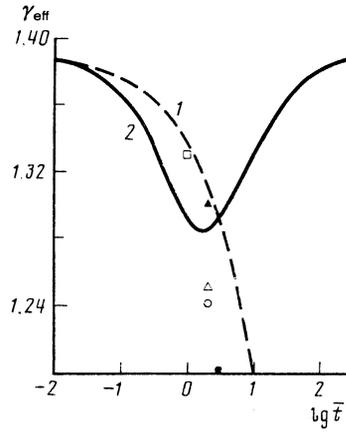
FIG. 5. Dependence of γ_{eff} on $\log \bar{t}$ in the crossover range: ●) Ref. 16; △) Ref. 15; ▲) Refs. 12 and 13; ○) Ref. 14; □) results of the present study.

large as the corresponding estimate obtained from the molecular field theory. Substitution of this value of t_c into Eq. (13) on the assumption that $\Delta_d = 0.55$ gives $a_\chi = 0.727$, which is in excellent agreement with the estimate of a_χ obtained from an analysis of the experimental data in accordance with Eq. (7).

Another piece of evidence in support of the dipole nature of the critical behavior of gadolinium is provided by a comparison of the effective critical exponents $\gamma_{\text{eff}} = -\partial \lg \chi/\partial \lg t$ calculated from the experimental results and theoretically. The definition of γ_{eff} and Eqs. (7) and (13) yield

$$\gamma_{\text{eff}} = \gamma_d - a_\chi \Delta_d \bar{t}^{\Delta_d} = \gamma_d - 0.0544 \bar{t}^{\Delta_d} \qquad (16)$$

Here, $\bar{t} = t/t_c$. If we allow for terms of higher order, the correction to the scaling of γ_{eff} can be found from the scaling function for the susceptibility in the crossover range.[33] Equation (10) and the definition of γ_{eff} yield

$$\gamma_{\text{eff}} = (1-y)^{1/\Phi_d} \left[\gamma_H + \frac{\Phi_d y \gamma_d}{1-y} + \frac{\Phi y P'(y)}{P(y)} \right], \qquad (17)$$

where $y = (1 + \bar{t})^{-\Phi_d}$; $P(y) = X(y)/(1-y)^{-\gamma_d}$; $P'(y)$ is the first derivative of $P(y)$ with respect to y. The expressions for the calculation of $P(y)$ and $P'(y)$ are given in Refs. 22 and 33. The dependences of γ_{eff} on $\bar{t}$ calculated from Eq. (16) (dashed curve) and from Eq. (17) (continuous curve) are presented in Fig. 5. Here, for comparison with the theory, we plotted the experimental values of γ_{eff} of gadolinium. The points with different configurations in Fig. 5 correspond to the values of γ_{eff} listed in Table I. We must bear in mind that γ_{eff} cannot be determined from the experimental results at each temperature and, therefore, Fig. 5 gives only one value corresponding to the average temperature in the interval where the critical exponent was determined, bearing in mind that $\bar{t}$ should be calculated as the arithmetic mean of $\log t$ (Ref. 33). An estimate of $\bar{t}$ based on our results gives $\bar{t} = 0.995$ and $\log \bar{t} = -0.0022$. In Fig. 5 this value of $\log \bar{t}$ is represented by an experimental point labeled by a square, which is in good agreement with the theoretical value deduced from Eq. (16). Consequently, in an analysis of the experimental data for gadolinium we need to consider only the first term of the correction to the scaling. This conclu-

sion is supported also by the results reported in Refs. 12 and 13, which indicate that $\gamma_{\text{eff}} = 1.33$ and $\log \bar{t} = 0.339$ (Table I), and by the agreement between the values of a_χ deduced from the experiments and from Eq. (13). In the other case only the first term of the correction to the scaling is allowed for. Moreover, it is clear from Fig. 5 that all the experimental studies of magnetic properties of gadolinium correspond to the crossover range and it is therefore not surprising that the critical exponent γ is undervalued compared with the asymptotic exponent.

It therefore follows that an analysis of the experimentally determined magnetic properties carried out by us allowing for the possibility of crossover and also used in dealing with the correction to the scaling, demonstrates that all the critical exponents as well as the universal relationship between the critical amplitudes and the amplitude of the correction to the scaling are in good agreement with the theoretical values for magnetic materials whose critical behavior is determined by isotropic dipole interactions.

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Translated by A. Tybulewicz