

# Phase transitions induced by a magnetic field in the vicinity of the compensation point in gadolinium iron garnets

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The  $H$ - $T$  phase diagram for a gadolinium iron garnet single crystal is investigated in the vicinity of the magnetic compensation point in the case when the magnetic field  $\mathbf{H}$  is parallel to the direction of the difficult magnetization axis. The investigation is based on measurements of the specific heat, the magnetocaloric effect, and the absorption and velocity of ultrasound propagation in a magnetic field. It is characteristic that no anomalies of the physical properties are observed at the compensation point itself in the absence of a magnetic field. A broad sound-absorption anomaly is observed somewhat below the compensation point in the absence of the field. On application of the field, a number of anomalies in the specific heat and in the absorption and velocity of ultrasound propagation are observed. The sound absorption coefficient is smaller in the field than at  $H = 0$ . Special attention is paid to a study of phase transitions induced by weak magnetic fields. The single-crystal measurements are compared with the theory. Calculations of various parameters in the vicinity of the compensation point, based on the experimental values, are presented. The peaks in sound absorption, specific heat, and magnetocaloric effect, arising on application of a magnetic field near the compensation temperature, are analyzed.

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The vicinity of the magnetic compensation points, at which the conditions are realized for the vanishing of the resultant magnetization, of the magnetocaloric effect, of the coercive force, of the magnetostriction, of the Faraday effect, of the critical field of the transition into a canted magnetic structure, of the ferrimagnetic-resonance frequency, and of the domain-wall velocity, turned out to be a rather interesting terrain for physical research (for references see<sup>[1,2]</sup>).

The problem of the magnetic compensation point was recently attracted attention again also in the unusually interesting magnetic  $H$ - $T$  phase diagram, initially constructed by Clark and Callen<sup>[3]</sup> on the basis of Tyablikov's theory<sup>[4]</sup> without allowance for the magnetic crystallographic anisotropy, and then refined further by Zvezdin and Matveev<sup>[5]</sup> with allowance for the rotation of the sublattices and the anisotropy energy. The conclusions of Clark and Callen<sup>[3]</sup> concerning the  $H$ - $T$  phase diagrams were confirmed by experiments in strong magnetic pulsed fields, performed by Belov and Levitin (see<sup>[2]</sup>) on a number of rare-earth iron garnets. However, as follows from the theoretical work of Zvezdin and Matveev<sup>[5]</sup>, it is precisely in weak magnetic fields that the  $H$ - $T$  phase diagram near the compensation point reveals qualitatively new and important features.

There have been a number of unsuccessful attempts to determine this diagram by magneto-optical<sup>[6,7]</sup> and other methods (see, e.g.,<sup>[2]</sup>). Nonetheless, its detailed form in the region of weak magnetic fields has not yet been experimentally established. There are still no systematic investigations of a number of most important physical properties in the vicinity of the compensation point, which can be furthermore used also to establish the nature of the phase transition, and to calculate the parameters that characterize the compensation points. This pertains primarily to the specific heat, to the sound absorption and propagation velocity, to the susceptibility, and to some other characteristics.

We report here the results of an experimental attempt to reconstruct the indicated magnetic  $H$ - $T$  phase diagram on the basis of measurements of the thermal and acoustic properties of a gadolinium iron garnet, and present a comparison with the theory. We investigated  $\text{Gd}_3\text{Fe}_5\text{O}_{12}$  (GdIG) in the case when the magnetic field  $\mathbf{H}$  coincides with the difficult magnetization axis [100]. We have applied the general theory of<sup>[5]</sup> to this particular case only to an extent needed to discuss the experimental results.

## $H$ - $T$ PHASE DIAGRAM OF GdIG FOR $\mathbf{H} \parallel [100]$ NEAR THE COMPENSATION POINT

Gadolinium iron garnet is in fact a three-sublattice ferrimagnet, but in fields  $\mathbf{H} \ll \mathbf{H}_1^{\text{exch}}$  it is regarded as a two-sublattice one. In the first combined sublattice  $\{a-d\}$  the Fe ions are antiferromagnetically coupled by the exchange  $\mathbf{H}_1^{\text{exch}}$  while the second almost-paramagnetic sublattice  $\{c\}$ , which consists of Gd ions, is under the influence of a weaker exchange field  $\mathbf{H}_2^{\text{exch}}$  and is antiferromagnetically coupled with the  $\{a-d\}$  sublattice. The field  $\mathbf{H}_2^{\text{exch}}$  acts only on the spin of the Gd ion. Its orbital magnetic moment is equal to zero.

Taking into account the weak dependence of the  $\{a-d\}$  sublattice magnetization on  $T$  and  $H$ , the thermodynamic potential of GdIG near the compensation point  $T_c$  is written in the form<sup>[5]</sup>

$$\Phi = \int_0^{H_{\text{eff}}} M_{\text{Gd}} dH_{\text{eff}} + K_{\text{Fe}} f(\theta, \varphi) - MH \cos \theta. \quad (1)$$

Here  $H_{\text{eff}} = \mathbf{H}_2^{\text{exch}} + H$ ,  $\mathbf{M} \equiv \mathbf{M}_{\text{Fe}}$  is the magnetization vector of the  $\{a-d\}$  sublattice,  $\mathbf{M}_{\text{Gd}}$  is that of the Gd sublattice,  $K_{\text{Fe}}$  is the anisotropy constant of the  $\{a-d\}$  sublattice, and the angles  $\theta$  and  $\varphi$  determine the direction of  $\mathbf{M}_{\text{Fe}}$  relative to the field and the crystallographic directions. We have taken into account here the fact that  $K_{\text{Fe}} > K_{\text{Gd}}$ . In the case  $\mathbf{H} \parallel [100]$  we have  $f(\theta, \varphi) = -1/4(\sin^2 2\theta + \sin^4 \theta \sin^2 2\varphi)$ . The condition that  $\Phi$  be

extremal relative to the angles  $\theta$  and  $\varphi$  takes the form (we henceforth put  $K_{Fe} \equiv K$ )

$$\frac{\partial \Phi}{\partial \theta} = MH(1 - \lambda \chi_0) \sin \theta + K \frac{\partial f}{\partial \theta} = 0, \quad (2)$$

$$\frac{\partial \Phi}{\partial \varphi} = K \frac{\partial f}{\partial \varphi} = 0, \quad \chi_0 = M_{Gd} / H_{\text{eff}}$$

and the exchange constant is  $\lambda = H_2^{\text{exch}} / M$ .

These equations have solutions  $\theta = 0$  and  $\theta = \pi$  (collinear phases) that correspond to the minimum of  $\Phi$  if  $\partial^2 \Phi / \partial \theta^2 |_{\theta=0, \theta=\pi} > 0$ . The stability limits of the collinear phases  $\theta = 0$  and  $\theta = \pi$  are given by the equations  $\partial^2 \Phi / \partial \theta^2 |_{\theta=0, \theta=\pi} = 0$ . For the case  $\mathbf{H} \parallel [100]$  they take, according to [5], the form

$$\frac{1}{T_{1,2}} = \frac{k_B B_S^{-1} [(1 \pm 4K/3MH) (\lambda M \pm H) / \lambda M_0^{\text{Gd}}]}{\mu_{Gd} (\lambda M \pm H)} \quad (3)$$

The minus sign pertains here to the quantity  $1/T_1$ , which determines the stability limit of the phase with  $\theta = 0$ , and the plus sign pertains to  $1/T_2$  and determines the limits of the stability of the phase with  $\theta = \pi$ .

We perform a numerical calculation of the stability limits of the collinear phases of GdIG. We assume for the calculations the values  $\mu_{Gd} = 7\mu_B$ ,  $\rho = 6.46 \text{ g/cm}^3$ ,  $T_C = 294^\circ \text{K}$ ,  $M_{Fe} = 135 \text{ G}$ ,  $H_{\text{eff}} = 250 \text{ kOe}$ , and  $\lambda = 1850$ . In addition, at the compensation point we have  $M_{Fe} = M_{Gd}$ , and therefore  $M_{Fe} = M_0^{\text{Gd}}{}_{7/2} (\mu_{Gd} H_{\text{eff}} / k_B T_C) = M_0^{\text{Gd}}{}_{7/2} (0.4011)$  and  $M_0^{\text{Gd}} = 780 \text{ Oe}$ , while the anisotropy constant is  $K = 6.7 \times 10^3 \text{ erg/cm}^3$  and  $H_{\text{eff}} = H_2^{\text{exch}} = M_{Fe}$ . Some of these values were taken from [1] and others were calculated by us, while the value of  $T_C$  corresponds to the experimental value of our sample.

Substitution of these values in (3) leads to the limits (represented by the thin lines AA' and BB' of Fig. 1) of the phase stability for GdIG in the case  $\mathbf{H} \parallel [100]$  and  $H = 0$  to 22 kOe. It is easily seen that the critical fields of the transition from the collinear to the canted phase decrease gradually on both sides of the compensation point, i.e., the phase diagram has patently a tendency to broaden in the weak-field region on the (H, T) plane. To the left of the BB' line the stable phase is the one with  $\theta = \pi$ , and to the right of the line AA' the phase with  $\theta = 0$ . In the region between AA' and BB' the minimum of  $\Phi$  is observed for values of  $\theta$  different from 0 and  $\pi$ —turning of the sublattices takes place.

Inside the noncollinear phase there appears on the H-T phase diagram a region where there are two solutions with different angles  $\theta$ , namely  $\theta = \theta_1, \varphi_0$  and  $\theta = \theta_2, \varphi_0$ . This region, bounded by the curves ON and ON', is metastable. [5] The limits of the metastability region are determined from the system of equations

$$\begin{aligned} (\sin \theta)^{-1} \frac{\partial \Phi}{\partial \theta} &= MH(1 - \lambda \chi_0) - K \cos \theta (3 \cos^2 \theta - 1) = 0, \\ \frac{\partial^2 \Phi}{\partial \theta^2} &= -MH \lambda \frac{\partial \chi_0}{\partial \theta} + K \sin \theta (9 \cos^2 \theta - 1) = 0. \end{aligned} \quad (4)$$

The angle  $\theta$  changes jumpwise with increasing temperature on the ON' line and with decreasing temperature on the ON line.

In a given field  $\mathbf{H}$ , as the temperature is varied we cross the phase-transition lines AA' and BB' on the H-T diagram, anomalies of the physical properties should be observed. It is shown in [5] that transitions from the collinear phases  $\theta = 0$  and  $\theta = \pi$  to a canted phase are second-order phase transition, in accord with the general theory of phase transitions. [5] When the remaining lines of the phase diagram (OT<sub>C</sub>, ON, ON') are crossed, anomalies of the physical properties can also

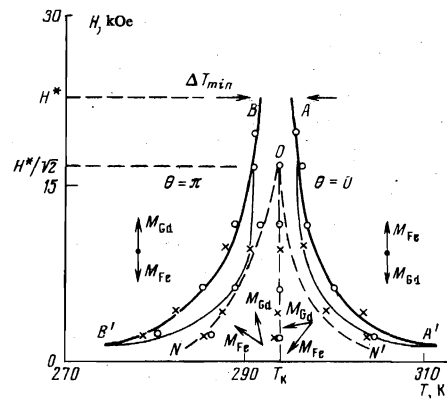


FIG. 1. Phase diagram in the (H, T) plane of gadolinium iron garnet;  $\mathbf{H} \parallel [100]$ . The thin solid lines AA' and BB' are the theoretical second-order transition lines calculated on the basis of (3). The thick curves AA' and BB' are the experimental second-order transition lines obtained from the measurements of the specific heat (O) and sound absorption at 30 MHz (X); the wave vector  $\mathbf{q}$  is perpendicular to  $\mathbf{H}$ ; OT<sub>C</sub> is the first-order phase transition line, ON and ON' are the limits of the stability region of the metastable phases, and O is the critical point. A schematic representation of the entire phase diagram is given in Fig. 3 of [5].

be observed, since the angle  $\theta$  changes jumpwise on the lines ON and ON', while on the line OT<sub>C</sub> the energies of the corresponding phases are equalized.

The physical cause of the jumplike change of the angle on the phase-diagram lines is the following: in a field  $\mathbf{H} \parallel [100]$ , the vector  $M_{Fe}$  should rotate from the [100] position ( $T < T_C$ ) to the position [100] ( $T > T_C$ ) in the planes (011) and (0 $\bar{1}$ 1) planes. The reason is that the maximum of the anisotropy energy lies along the directions [010] and [001], and the minima lie along type-[111] directions in the planes (011) and (0 $\bar{1}$ 1). The two minima of the anisotropy energy at  $M_{Fe} [111]$  and  $M_{Fe} [\bar{1}\bar{1}\bar{1}]$  are separated by an intermediate maximum at  $\theta = \pm \pi/2$ , and therefore the transition from one minimum to the other will be jumpwise. Since the positions of the energy minima change with changing field H, namely, they shift towards [011], it follows that at a certain value of the field H the turn will occur without a jump. [6] This value of the field corresponds to the point O on Fig. 1.

## EXPERIMENTAL RESULTS AND DISCUSSION

**Magnetization.** To assess the possibility of constructing the H-T diagram on the basis of magnetic measurements, we plotted 29 magnetization isotherms near  $T_C$ . It turned out that it is difficult to evaluate from the character of these isotherms the values of the critical fields at which the GdIG goes over into a phase with a canted magnetic-structure, particularly at  $T < T_C$ , when a strong paraprocess exists in the rare-earth sublattice. Nonetheless, from the isotherms for  $T > T_C$  we can see that the anisotropy field decreases with increasing temperature, and this decrease is connected with the critical field. [2] We note also that in the weak-field region the temperature dependence of the magnetization exhibits hysteresis, and the compensation point is located near  $294^\circ \text{K}$ .

**Specific heat.** Important information concerning the features of the phase diagram and the role of the fluctuations of the magnetization in the region of the field-induced orientational phase transitions can be obtained from investigations of the specific heat in a magnetic field in the vicinity of the compensation point. The first

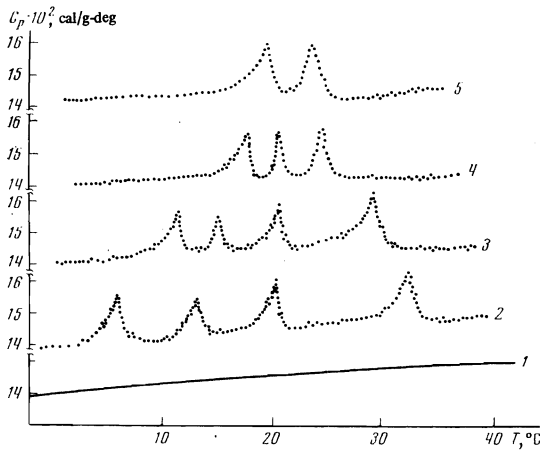


FIG. 2. Temperature dependence of the specific heat in the vicinity of the compensation point at various values of the magnetic field: 1)  $H = 0$ , 2)  $H = 2$  kOe, 3) 6 kOe, 4) 16.4 kOe, 5) 20.3 kOe;  $H \parallel [100]$ .

such investigations on polycrystalline GdIG were reported by us in<sup>[9]</sup>. Subsequently, similar measurements were made on YbIG by Ferron et al.<sup>[10]</sup>, but they investigated the case  $H \parallel [111]$ . A brief description of the adiabatic calorimeter used by us is contained in<sup>[9]</sup>, and a schematic description is given also in<sup>[11]</sup>.

The results of our measurements on single-crystal GdIG in the cases  $H = 0, 2.6, 16.4$  and  $20$  kOe are shown in Fig. 2. The magnetic-field direction coincided with the crystallographic direction  $[100]$ , i.e., with the difficult magnetization axis.

In the absence of a magnetic field, the specific heat  $C_p$  exhibits the normal temperature dependence (solid line), indicating that the compensation point at  $H = 0$  is not singular and is in no ways distinguished on the  $H$ - $T$  phase diagram.

In a magnetic field  $H$ , a number of peaks are observed on the temperature dependence of the specific heat  $C_{PH}$ . Thus, in fields  $H = 2$  and  $6$  kOe there are observed for peaks of  $C_{PH}$ , three at  $H = 16.4$  kOe, and two symmetrical peaks separated by  $\sim 3.8^\circ\text{K}$  remain at  $H = 20$  kOe. The interval of each transformation of  $C_{PH}$  is relatively narrow and equals approximately  $2$ - $3^\circ\text{K}$ , while the interval of the transformations of the two symmetrical peaks of  $C_{PH}$  at  $20$  kOe is somewhat larger,  $\sim 4^\circ\text{K}$ . It is typical, however, that the width of the central peak of  $C_{PH}$  decreases with increasing field, and this peak is not observed at  $H = 20$  kOe.

To evaluate the results, we marked the temperatures corresponding to the maxima of  $C_{PH}$  in a given field on the  $H$ - $T$  diagram. These values are represented in Fig. 1 by circles. As seen from the figure, the circles lie near the phase-transition lines. The values corresponding to the outermost maxima of  $C_{PH}(T)$  lie near the second-order phase transition lines (lines  $AA'$  and  $BB'$ ). The outermost peaks of  $C_{PH}(T)$  on the left thus correspond to a transition of the system from a collinear to a canted phase through the line  $BB'$ , while the outermost right-hand peaks correspond to the transition of the system from the canted to the collinear phase through the second-order phase transition line  $AA'$ . The phase-boundary curves (thick solid lines), which were drawn through the experimental points, differ from the theoretical ones in a definite interval of fields and temperatures. Agreement is observed only in the case of strong and weak fields.

The experimental points corresponding to the intermediate  $C_{PH}(T)$  maxima lie both on the metastable boundary  $ON$  and on the line  $OT_c$ .

It must be noted here, however, that theoretically, in the homogeneous case, anomalies of the physical properties should be observed in a field  $H \parallel [100]$ , including anomalies of the specific heat on the lines  $AA'$  and  $BB''$ , and one more on the lines  $ON$  and  $ON'$  or on the line  $OT_c$ . In the inhomogeneous case, i.e., when a domain structure exists in the canted phase, one cannot exclude the possibility (due, e.g., to the instability of the domain structure) of the appearance of anomalies one of the lines  $ON$  or  $ON'$ , as well as on the first-order phase-transition line  $OT_c$ . The maximum number of peaks in the temperature dependence of the specific heat corresponds to the last case. Nonetheless, there is no physically clear explanation of the simultaneous appearance of anomalies of the specific heat on the lines  $ON$  and  $OT_c$ . The reason is that the physical processes that occur in the metastable region  $NON'$  have not yet been fully analyzed. Therefore the nature of the fourth peak of the specific heat in the fields  $2$  and  $6$  kOe is likewise not quite clear to us.

The central  $C_{PH}$  peak (i.e., the peak on the line  $OT_c$ ) is observed up to a field  $H = 16.3$  kOe. This value was chosen to make the field approximately equal to  $H^*/\sqrt{2}$ ,<sup>[5]</sup> at which the jump of the angle  $\theta$  theoretically becomes equal to zero. We therefore assume that this maximum occurs at the critical point  $O$  of the  $H$ - $T$  diagram.

The results of the measurements of the specific heat indicate in general that the temperature interval of the existence of a canted magnetic structure does not vanish at  $T = T_c$ , as follows from the model in which the anisotropy field is not taken into account, but broadens in accordance with the model in which the anisotropy is taken into account.<sup>[5]</sup>

In the model of Clark and Callen<sup>[3]</sup> for rare-earth iron garnets in the canted phase the magnetization does not depend on the temperature:  $(\partial M/\partial T)_H = 0$ . In this case it follows from the general thermodynamic relation between the specific heats at constant field and constant magnetization

$$C_M - C_H = T(\partial M/\partial T)_H(\partial H/\partial T)_M \quad (5)$$

that  $C_M = C_H$ , and since  $C_M$  is independent of  $H$ , it follows that  $C_H$  in the canted phase should likewise be independent of  $H$ .<sup>[12]</sup>

We shall attempt to compare this conclusion with the experiment. In the general case,  $C_H = f(H) + C_M$ , where  $f(H)$  is the magnetic contribution due to the field  $H$ ;  $f(H)$  vanishes in the canted phase. In addition,  $C_H = C_{\text{phon}} + C_S$ , where  $C_{\text{phon}}$  and  $C_S$  are respectively the phonon and spin specific heats. In the case of GdIG, for which  $T_c$  is at room temperatures, we have  $C_{\text{phon}} \approx C_S$  and therefore in fields  $H \ll H_{1\text{cr}}$  we have  $C_H \approx f(H) + C_{\text{phon}}$  and we can separate  $C_{\text{phon}}$  from  $f(H)$ , since we have  $C_{\text{phon}} \approx C_H$  in a field  $H_{1\text{cr}} < H < H_{2\text{cr}}$ .

From the experimental results of Fig. 2 it follows that in the canted phase, in the presence of a field, the value of  $C_{PH}$  is smaller than in its presence, and in a field  $H = 2$  kOe the value of  $C_{PH}$  in the collinear phase is larger than in the canted phase below  $T_c$ , in qualitative agreement with the advanced thermodynamic arguments. In stronger fields  $H$ , however, it is difficult to

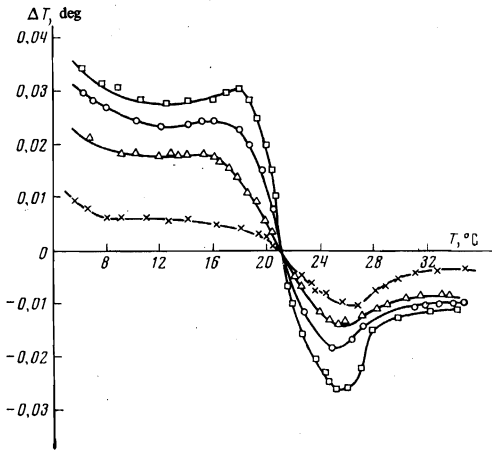


FIG. 3. Temperature dependence of the magnetocaloric effect in the region of the compensation point:  $\square$ — $H = 12.5$  kOe,  $\circ$ — $10$  kOe,  $\triangle$ — $7$  kOe,  $\times$ — $2.0$  kOe;  $H \parallel [100]$ .

make the corresponding comparison. In addition, the conclusion of the theory of<sup>[3]</sup> are approximate not only because the calculations were made within the framework of the molecular-field theory, but also because no account was taken of the anisotropy energy.

A quantitative examination of the magnetic contribution to the specific heat, again within the framework of the molecular field theory, but with allowance for the anisotropy energy, is possible on the basis of expression (1). It turns out that the magnetic part of the specific heat in the phases  $\theta = 0$  and  $\theta = \pi$  is of the form<sup>[5,13]</sup>

$$C_{PH}|_{\theta=0, \theta=\pi} = \frac{aM_{Gd}\mu_{Gd}}{k_B T^2} (H^2 \mp 2\lambda MH). \quad (6)$$

The minus and plus signs correspond to  $\theta = 0$  and  $\theta = \pi$ . The experimental data agree in general with formula (6), since the specific heat  $C_{PH}$  is smaller in the phase  $\theta = 0$  than in the phase  $\theta = \pi$  at all values of the field. In the canted phase, the magnetic specific heat is no longer equal to zero, as would be expected according to Clark and Callen.<sup>[3]</sup> The corresponding expression for the jump of the magnetic specific heat on going from the collinear to the canted phase is<sup>[5]</sup>

$$\Delta C_{PH} \approx \frac{a^2 M_0^{Gd}}{2b \mu_{Gd}} K \left( 1 + \frac{4K\lambda M}{bx_0^3 M_0^{Gd} H^2} \right)^{-1}, \quad (7)$$

where  $x_0 \equiv \mu_0^{Gd} \lambda M / k_B T_C$ , the coefficients  $a$  and  $b$  enter in the expansion  $B_S(x) = ax - bx^3 + \dots$ , of the Brillouin function, and  $x = \mu Gd H_{eff} / k_B T$ . Since  $a \approx 0.43$  and  $a^2/2b \approx 1$  for GdIG, it follows that by substituting the corresponding values in (6) and (7) we obtain

$$C_{PH}|_{\theta=0, \theta=\pi} \approx 10^{-2} \text{ cal/g-deg}, \quad \Delta C_{PH} \approx 10^{-2} \text{ cal/g-deg}.$$

Experimentally, in the fields used, we have  $C_{PH}|_{\theta=0} - C_{PH}|_{\theta=\pi} = 1$  to  $1.5\%$ , within the limits of the experimental accuracy, and  $\Delta C_{exp} = 9$  to  $12\%$  of the normal course of  $C_p$ , and is consequently larger than the theoretical value, probably as a result of the fluctuation contribution, which is not accounted for in the theory of<sup>[5]</sup>.

**Magnetocaloric effect.** The use of an adiabatic calorimeter has made it possible to measure not only the specific heat but also the magnetocaloric effect (henceforth called the  $\Delta T(H)$  effect). Measurements of the temperature of a body following application of a mag-

netic field were used successfully in the past to estimate a number of parameters in the vicinity of the compensation point, including GdIG.<sup>[2,14]</sup>

Our measurements of the  $\Delta T(H)$  effect were made not only because no such measurements had been made on single crystals in a specified field direction relative to the crystallographic axes, but also to determine accurately the compensation temperature  $T_C$ , to compare the results with the  $C_{PH}$  data, and then to calculate the exchange-interaction constant.

Figure 3 shows the results of measurements of the magnetocaloric effect at various values of  $H$  near  $T_C$ , at  $H \parallel [100]$ . At the compensation point, at all values of the field, an inversion of the  $\Delta T(H)$  effect is observed as a function of the temperature, in agreement with<sup>[2,14]</sup>; this is connected with the character of the paraprocess in the paramagnetic gadolinium sublattice. Thus, the magnetic moment  $M_{Gd}$  is larger than  $M_{Fe}$  below  $T_C$  and is directed along the field  $H$  and  $H_{eff}$ . The field  $H$  decreases the entropy of the spin subsystem of the gadolinium sublattice (paraprocess of the ferromagnetic type), a fact accompanied by an increase of the thermal energy of the phonon sublattice. The latter corresponds to a positive sign of the  $\Delta T(H)$  effect and is realized in experiment. Above  $T_C$  we already have  $M_{Fe} > M_{Gd}$ . Therefore  $M_{Fe}$  is directed along the field  $H$  and is antiparallel to  $H_{eff}$  and  $M_{Gd}$ . In this case the field  $H$  increases the entropy of the spin subsystem of the gadolinium sublattice (paraprocess of the antiferromagnetic type), this being accompanied by cooling of the entire system, and thus leads to a negative sign of the  $\Delta T(H)$  effect. These arguments are valid, naturally, if we are dealing with the transition of the system from one collinear phase to another through the compensation point without allowance for the turning of the magnetic moment and the canted structure. However, the real phase diagram of GdIG does not correspond in this case to the situation of a jumplike transition from one collinear arrangement of the magnetic moments to another. As seen from Fig. 1, these two regions are separated by a rotation region, where a canted magnetic structure is produced at the same time. It is clear that this circumstance will influence the behavior of the  $\Delta T(H)$  effect in the vicinity of  $T_C$  and must therefore be taken into account.

The theory of the  $\Delta T(H)$  effect at the compensation point, within the framework of the molecular field theory, was developed by Belov and Nikitin<sup>[2,13]</sup> but without allowance for the rotation of the lattice and the canted structure.

An expression for the  $\Delta T(H)$  effect with allowance for the magnetic-anisotropy energy was obtained in<sup>[5]</sup>. It follows from this expression that  $(\partial T/\partial H)_S$  in the canted phase differs from zero. On the other hand, if the anisotropy constant  $K = 0$ , we obtain the well known result  $(\partial T/\partial H)_S = 0$ .<sup>[3]</sup> Our experiment yields  $(\partial T/\partial H)_S \neq 0$  in the canted phase, just as in<sup>[13]</sup>.

A characteristic feature of our experimental results is the presence of extrema of the  $\Delta T(H)$  effect both below and above  $T_C$ . Above  $T_C$  these extrema are sharper and were noted in<sup>[14]</sup>, where they were attributed to the influence of the domains. However, a comparison of the temperatures of the extrema with the phase-boundary lines on Fig. 1 shows that these extrema lie approximately on the boundaries of the phase lines. Conse-

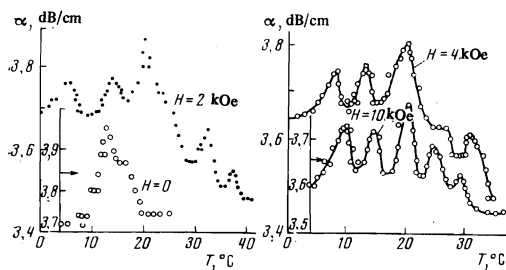


FIG. 4. Temperature dependence of the sound absorption coefficient at 30 MHz;  $q \perp H \parallel [100]$ .

quently, the region between these extrema corresponds to the region of the existence of the canted phase.

**Sound absorption.** Besides measurements of the specific heat and of the  $\Delta T(H)$  effect, we measured also the sound absorption in the vicinity of the magnetic compensation point. The sound absorption coefficient was measured<sup>[15]</sup> at 30 MHz in fields  $H = 0, 2, 4,$  and  $10$  kOe. The results are shown in Fig. 4. The sound propagation direction coincided with  $H \parallel [100]$ .

It is known that near the phase-transition points the sound absorption coefficient increases, possibly as a result of the growth of the order-parameter fluctuations in the system, or the anomalous growth of the relaxation time.<sup>[16-18]</sup> The absorption maximum may also be the result of resonant sound absorption in the presence of a field, or of natural magnetoelastic resonance in the absence of a field.<sup>[19]</sup> The sound-absorption coefficient can therefore be used to observe phase transitions in the vicinity of the compensation point, and consequently to reconstruct the  $H-T$  phase diagram.

It is seen from the measurement results (Fig. 4) that, in contrast to the specific heat, a broad sound-absorption maximum is observed in the absence of a field, somewhat below the compensation point. Since the compensation point in the absence of a field is not a phase-transition point (as follows from the measurements of  $C_p$ ), the sound-absorption peak at  $H = 0$  can be ascribed to domain processes in the vicinity of but somewhat lower than  $T_C$ . These processes are due to acoustic waves propagating through the crystal or to some other magnetic relaxation phenomena. This interpretation is also confirmed by the fact that the peak vanishes when a field is applied.

The results of measurements in a magnetic field differ strongly from the case  $H = 0$ . In a field  $H$ , just as in the case of the specific heat, a number of absorption maxima appear. The number of sound-absorption peaks, however, is even larger than in the case of  $C_p$ . This shows once more that the processes that occur in the canted phase must be subjected to a more thorough theoretical analysis.

As seen from Fig. 4, the amplitude of the absorption maxima is smaller than the amplitude of the absorption peak at  $H = 0$ , and in addition the sound-absorption maxima are observed both at the point  $T_C$  itself and away from this point. With increasing field  $H$ , the temperature interval within which the absorption maxima are observed becomes narrower, and this agrees with the results of the specific-heat measurements. Very complicated results were obtained by also for the speed of sound, for which a number of minima was observed. If the temperatures of the sound-absorption maxima are

plotted on the  $H-T$  diagram (the crosses on Fig. 1) then, as seen from Fig. 1, they lie near the corresponding values of the specific heat. Just as in the case of the specific heat, the temperatures corresponding to the outermost maxima lie near the second-order phase transition lines  $AA'$  and  $BB''$ , and the remaining ones lie near the boundaries of the metastable phases  $ON$  and  $ON'$  and the line  $OT_C$ . Therefore the interpretation of the sound-absorption maxima is the same as in the case of  $C_p$ .

A feature of the dependence of the sound-absorption coefficient near  $T_C$  on the intensity of the external magnetic field is an initial decrease with the field, a tendency to saturation, and the absence of a resonant magnetoelastic interaction between the sound and the spin system. The field dependence of the absorption is also in qualitative agreement with the  $H-T$  phase diagram.

Thus, the entire set of measurements performed near  $T_C$  confirms the important role played by the magnetic crystallographic anisotropy near the compensation point in relatively weak magnetic fields, and corroborates the qualitative conclusions of the theory of Zvezdin and co-workers concerning the magnetic phase diagram in the  $(H, T)$  plane. For a better agreement it is necessary to take into account, within the framework of the molecular-field theory, at least the possibility of formation of a canted magnetic structure in the combined iron sublattice, and the spin fluctuations near the second-order phase transition lines.

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