

# An investigation of the magnetic properties of erbium by the muon method

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The magnetically ordered and paramagnetic states of erbium at temperatures  $T < T_N$  and  $T > T_N$ , where  $T_N = 84.4$  K is the Néel temperature, have been studied by the muon method. A large crystalline specimen of erbium was used with preferred orientation of the hexagonal  $c$  axes of the individual single crystals. The correlation functions  $G_{\parallel}(T)$  and  $G_{\perp}(T)$  which describe the fluctuations of the longitudinal and transverse (relative to the  $c$  axis) components of the internal magnetic field acting on a muon at  $T > T_N$  were measured. The  $G_{\perp}(T)$  dependence was also measured at  $T < T_N$ . It is shown that for  $T > T_N$  the correlator  $G_{\perp}$  is constant over the whole temperature range investigated,  $T_N < T \leq 300$  K, while  $G_{\parallel}$  increases to a limited extent as  $T \rightarrow T_N$ . The limited range of the  $G_{\parallel}(T)$  dependence as  $T \rightarrow T_N$  is regarded as an indication that a weak first-order phase transition takes place in erbium at  $T_N = 84.4$  K.

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## §1. INTRODUCTION

It has been shown<sup>1,2</sup> that antiferromagnetic phase transitions in rare earth lanthanide group metals can be studied effectively by the muon method. The relaxation of the muon spin measured in this method is determined by the internal magnetic fields of the antiferromagnet and depends on their magnitude, direction, and rates of oscillation. The internal magnetic field at the muon is produced by the magnetic moments of the metal atoms. The atomic magnetic moments oscillate with frequency  $\sim 10^{12} \text{ s}^{-1}$  in the paramagnetic state above the Néel temperature,  $T > T_N$ , and the internal magnetic fields oscillate with the same frequency as a result. The magnetic moments are partially (or totally) ordered at  $T < T_N$ , thanks to which the internal magnetic fields at  $T < T_N$  have in general both constant and alternating components. The alternating magnetic fields lead to a relatively slow relaxation of the muon spin, while in the constant field of the magnetically ordered state the muon spin relaxation usually takes place unobservably rapidly. With the muon method, antiferromagnetic phase transitions can be fixed extremely accurately, and the fluctuational formation of the magnetically ordered phase in the paramagnetic state at  $T > T_N$  can be studied as can the dynamics of the oscillating components of the atomic spins at  $T < T_N$ .

In the present work the relaxation of muon spin in erbium in zero external field has been measured. A large crystalline high purity specimen of erbium with preferred orientation of the hexagonal  $c$  axes of the individual single crystals was used. The dependence of the relaxation on the orientation of the  $c$  axis relative to the muon polarization direction could be measured with a specimen prepared in this way.

Erbium crystallizes in a hexagonal close-packed lattice and has a rather complicated magnetically ordered structure. It is a helicoidal ferromagnet at  $T < 20$  K, a cycloidal

antiferromagnetic structure occurs at  $20 < T < 53$  K, and finally at  $53 < T < 84.4$  K there is a sinusoidal antiferromagnetic structure. Erbium becomes paramagnetic at  $T_N = 84.4$  K. The sinusoidal structure is characterized by the absence of an ordered magnetic moment in the basal plane of the crystal, and the magnitudes of the atomic magnetic moments directed along the  $c$  axis vary according to a sine law. The crystalline hexagonal axis is the symmetry axis for all the magnetic structures. In this work the internal magnetic fields in the paramagnetic and antiferromagnetic states have been studied. In the paramagnetic state the temperature dependence  $\Lambda(T)$  of the muon spin relaxation rate due to fluctuation processes has been measured for  $T \rightarrow T_N$ . It is shown that as  $T \rightarrow T_N$  the  $\Lambda(T)$  dependence does not have the singularity characteristic of a first-order phase transition. The work was carried out on the muon channel of the Leningrad Institute of Nuclear Physics at Gatchina.

## §2. EXPERIMENTS

The erbium specimen used in the present work was specially purified and the total impurity content did not exceed 0.01% by weight. The specimen was made up of six 30 mm diameter disks, each 5 mm thick, arranged so that the basal planes were parallel. The disks were prepared from polycrystalline material and were annealed while deformed in bilateral compression. The dimensions of the individual single crystals increased to 4–8 mm after annealing, and their hexagonal  $c$  axes acquired a preferential orientation perpendicular to the basal plane of the disk. The value  $\langle \cos^2 \vartheta \rangle = 0.533 \pm 0.010$  of the mean square cosine of the angle between the directions of the  $c$  axes of individual crystals and the perpendicular to the plane of the disk, averaged over all disks, was determined experimentally (see § 3). The measurements were carried out for two orientations of the specimen.

The orientations were determined by the angles  $\alpha = 0$  and  $\alpha = \pi/2$  between the direction of the muon polarization and the perpendicular to the plane of the disks, i.e., the direction of preferred orientation of the  $c$  axes of individual single crystals.

The specimen was placed in a special cryostat where it was bathed in an intense flow of gas whose temperature was stabilized automatically. The specimen temperature was measured with germanium thermometers to a relative accuracy  $T_{\text{rel}} \approx 0.05$  K. The absolute accuracy was determined by the thermometer calibration and was  $\delta T_{\text{abs}} \approx 0.3$  K. The nonuniformity in the temperature of the specimen did not exceed 0.2 K. For  $T > 80$  K nitrogen was used for the gas stream and for  $T = 20$  to 80 K it was helium.

Muon spin relaxation was studied in an external magnetic field  $H_{\text{ext}} = 0$ , achieved to an accuracy  $\delta H_{\text{ext}} = 0.1$  Oe. The parameters describing the relaxation were determined by finding the best fit of the experiments to the calculated time dependence

$$N(t) = N_0 e^{-t/\tau_0} (1 + a e^{-\Lambda t}) + B \quad (1)$$

of the number of positrons from the  $\mu^+ \rightarrow e^+$  decay emitted in the direction of the muon polarization. Here  $\tau_0 = 2.2 \times 10^{-6}$  s is the muon lifetime;  $a$  is the experimental asymmetry coefficient of the angular distribution of positrons from the  $\mu^+ \rightarrow e^+$  decay;  $\Lambda$  is the muon spin relaxation rate;  $B$  is the background. It is assumed in Eq. (1) that muon spin relaxation follows the exponential law  $P(t) = e^{-\Lambda t}$ . Experiment confirms the exponential  $P(t)$  dependence over the whole temperature range investigated,  $T = 20$  to 300 K. In each measurement the total number of  $\mu^+ \rightarrow e^+$  events recorded was about  $10^6$ . The background was  $B \leq 0.01 N_0$ .

An example of an experimental and the corresponding calculated  $N(t)$  dependence in erbium is shown in Fig. 1. The relatively slow relaxation ( $\Lambda = 1.6 \times 10^6 \text{ s}^{-1}$ ) shown in Fig. 1

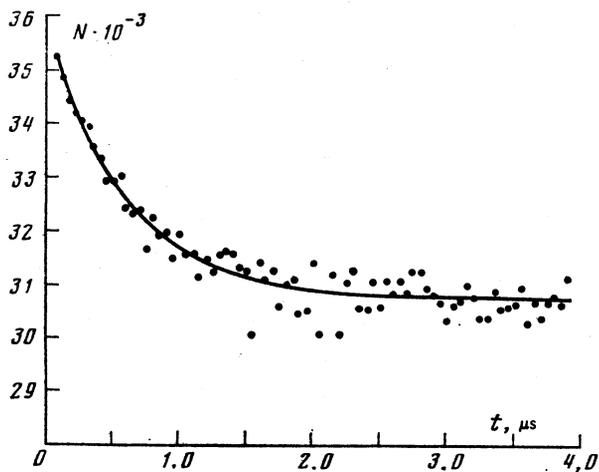


FIG. 1. Muon spin relaxation in the paramagnetic state of erbium for  $\alpha = 0$ ,  $T = 85.4$  K. The channel width of the time analyzer is  $\Delta t = 50$  ns. The smooth curve is the theoretical  $N(t)$  dependence (1) with the parameters  $N_0$ ,  $a$ ,  $\Lambda$  and  $B$  chosen by the maximum likelihood method; in particular, the parameter  $\Lambda = (1.61 \pm 0.09) \mu\text{s}^{-1}$ . The experimental and theoretical dependences shown are fitted to the muon-decay exponential  $e^{-t/\tau_0}$ .

is explained by the high frequency,  $\nu \sim 10^{12} \text{ s}^{-1}$ , of the oscillations of atomic spins in the paramagnetic state of the metal:

$$\Lambda = \sigma \frac{2\sigma}{\nu}. \quad (2)$$

Here  $\sigma \sim 10^9 \text{ s}^{-1}$  is the muon spin relaxation rate in the erbium crystal lattice with "frozen-in" magnetic moments. Figure 1 illustrates the good agreement mentioned above between the experimental  $P(t)$  dependence and the exponential assumed in the calculation.

The experimentally measured asymmetry coefficient,  $a$ , of the  $\mu^+ \rightarrow e^+$  decay positron angular distribution for  $T$  between 80 and 300 K is shown in Fig. 2 for two orientations of the specimen corresponding to the angles  $\alpha = 0$  and  $\alpha = \pi/2$ . It can be seen from Fig. 2 that the  $a(T)$  dependence undergoes a sharp change at a temperature close to the assumed temperature  $T_N = 84.4$  K of the antiferromagnetic phase transition.<sup>3</sup> The experimental width of the transition, as can be seen in Fig. 2, is  $\Delta T \leq 0.2$  K. The magnitude of  $\Delta T$  is determined by the temperature interval over which the value of the coefficient  $a$  is observed to be intermediate between its values in regions  $T > T_N$  and  $T < T_N$ , and can be connected with the nonuniformity of the specimen temperature. The value  $a = a_0$  shown in Fig. 2 for  $T > T_N$  is independent of the specimen orientation. The values  $a = a_1$  and  $a = a_2$  for  $T < T_N$ , corresponding to  $\alpha = 0$  and  $\alpha = \pi/2$ , are different:

$$\begin{aligned} a_0 &= 0.180 \pm 0.001, & T > T_N; \\ a_1 &= 0.113 \pm 0.001, & T < T_N, \quad \alpha = 0; \\ a_2 &= 0.070 \pm 0.002, & T < T_N, \quad \alpha = \pi/2. \end{aligned} \quad (3)$$

The decrease of  $a$  on passing from the paramagnetic to the antiferromagnetic state comes about because of the unobservably rapid relaxation of muon spin under the action of constant magnetic fields in the magnetically ordered state. In octahedral and tetrahedral channels of the crystal lattice these fields are directed along the hexagonal  $c$  axis at  $T = 53$  to 84 K. The muon spin component perpendicular to the  $c$  axis thus relaxes rapidly at  $T < T_N$ . The longitudinal component undergoes slow relaxation ( $\Lambda \sim 10^6 \text{ s}^{-1}$ ) at  $T < T_N$  under the action of rapid oscillations of magnetic fields produced by the remaining disordered components of the atomic spins of the metal. It is this slow relaxation of the longitudinal component of muon spin which is observed at  $T < T_N$ , with the corresponding reduced experimental asymmetry coefficient  $a$ . It can also be seen from Fig. 2 that a small, 5 to 10%, reduction in  $a$  is also observed in the paramagnetic state for  $T \lesssim 88$  K.

The temperature dependence  $\Lambda(T)$  of the muon spin relaxation rate is shown in Figs. 3 and 4. The values of  $\Lambda$  in the temperature range  $T = 84.9$  to 85.2 K, corresponding to the transitional values of  $a$  ( $a_1 < a < a_0$  for the orientation  $\alpha = 0$  and  $a_2 < a < a_0$  for  $\alpha = \pi/2$ ) are not shown in Fig. 3. The values of  $\Lambda$  not shown in Fig. 3 lie within the limits  $\Lambda = (1.0$  to  $1.9) \times 10^6 \text{ s}^{-1}$ .

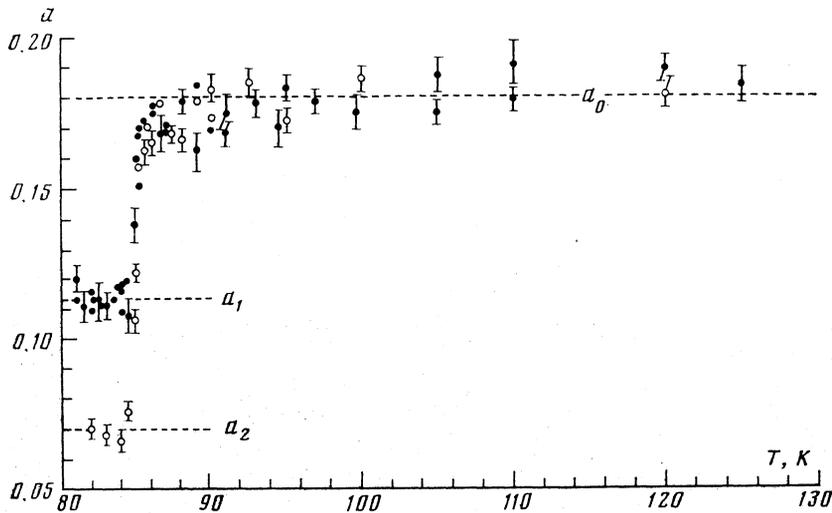


FIG. 2. Temperature dependence of the measured coefficients of angular asymmetry of the distribution of  $\mu^+ \rightarrow e^+$  decay positrons in erbium at  $T > 80$  K for  $\alpha = 0$  (●) and  $\alpha = \pi/2$  (○). The values of the coefficients  $a_0, a_1, a_2$  are given in the text.

It can be seen from Fig. 3 that  $\Lambda$  is constant and equal to  $\Lambda (T \gtrsim 200 \text{ K}) = 1 \times 10^6 \text{ s}^{-1}$  at high temperatures  $T \sim 200$  to  $300 \text{ K}$ . As the temperature is reduced to  $T \rightarrow T_N$  the magnitude of  $\Lambda$  grows, but this growth is limited and does not exceed double the value of  $\Lambda (T \gtrsim 200 \text{ K})$ . The increase in  $\Lambda$  for  $T > T_N$  is connected with the fluctuational formation of a magnetically ordered phase in the paramagnetic state and the corresponding reduction in the frequency [see Eq. (2)] of the oscillations of the atomic spins of the metal.<sup>2</sup> It follows from Fig. 3 that the fluctuational growth in  $\Lambda$  reveals a strong dependence on specimen orientation:

$$\Lambda(\alpha = \pi/2) > \Lambda(\alpha = 0). \quad (4)$$

Equation (4) corresponds to the idea that the magnetically ordered state formed by fluctuations at  $T > T_N$  has the structure of the magnetically ordered state at  $T < T_N$  when the local magnetic field at the muon in erbium is directed along the crystal  $c$  axis. The cause of the faster relaxation rate for  $\alpha = \pi/2$  is that the magnetic fields produced by fluctuations are in this case preferentially directed perpendicular to the muon spin.

The relaxation rate at  $T < T_N$  is almost equal to  $\Lambda (T > 200 \text{ K})$  in the paramagnetic state at high temperature. The reduction in  $\Lambda$  in the magnetically ordered state at  $T < 60 \text{ K}$  (see Fig. 4) is evidently connected with the rearrangement of the antiferromagnetic structure at  $T_0 = 53 \text{ K}$ .

### §3. DISCUSSION

The experimental values [Eq. (3)] of the asymmetry coefficients  $a_0, a_1, a_2$  (see also Fig. 2) agree among themselves only on the assumption that a certain fraction  $\kappa$  of muons is practically not acted upon by the constant magnetic field at  $T < T_N$ . The possible value of the constant field is in the case  $H_{\text{const}} < 20 \text{ Oe}$ , when its effect imitates the slow muon-spin relaxation determined by the coefficients  $a_1$  and  $a_2$  at  $T < T_N$ . In fact, the ratios  $a_1/a_0$  and  $a_2/a_0$ , expressed through the mean square cosine  $\langle \cos^2 \vartheta \rangle$  of the angles  $\vartheta$  between the directions of the hexagonal  $c$  axes of individual crystals, can be written in the form

$$\begin{aligned} a_1/a_0 &= \kappa + (1-\kappa) \langle \cos^2 \vartheta \rangle, & \alpha = 0, \\ a_2/a_0 &= \kappa + (1-\kappa)^{1/2} (1 - \langle \cos^2 \vartheta \rangle), & \alpha = \pi/2. \end{aligned} \quad (5)$$

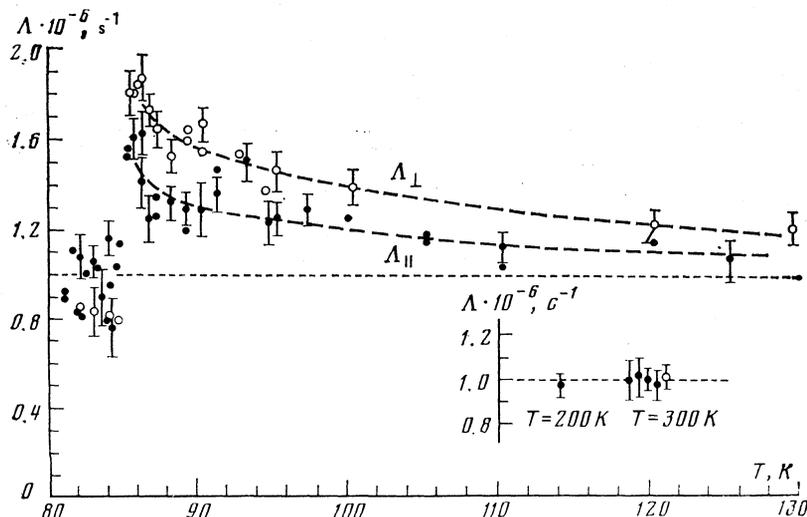


FIG. 3. Temperature dependence of the muon spin relaxation rate  $\Lambda(T)$  in erbium at  $T > 80 \text{ K}$  for  $\alpha = 0$  and  $\alpha = \pi/2$ . The smooth curves of  $\Lambda_{\parallel}(T)$  and  $\Lambda_{\perp}(T)$  are drawn using the experimental values respectively for  $\Lambda(\alpha = 0)$  and  $\Lambda(\alpha = \pi/2)$ .

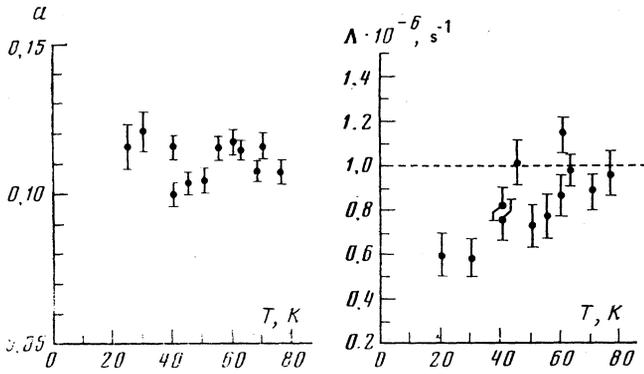


FIG. 4. Temperature dependences of  $a(T)$  and  $\lambda(T)$  for erbium at  $T = 20$  to  $80$  K for the orientation  $\alpha = 0$ .

The relations (5) and the values (3) of the coefficients  $a_0$ ,  $a_1$  and  $a_2$  lead to the following values of  $\kappa$  and of  $\langle \cos^2 \vartheta \rangle$ :

$$\kappa = 0.20 \pm 0.02, \quad \langle \cos^2 \vartheta \rangle = 0.533 \pm 0.010. \quad (6)$$

A zero magnetic field at a muon at  $T < T_N$  can occur in an octapore combined with a node of the sinusoidal wave of the ordered magnetic moments if, naturally, the magnetic and crystal structures of the metal correlate with one another. The number of such octapores is evidently a little less than the experimental value [Eq. (6)]  $\kappa = 0.20 \pm 0.02$  and is only  $\kappa_{\text{oct}} = 1/7$ , since the wavelength of the sinusoidal magnetic structure is equal to 7 layers of the lattice atoms.<sup>3</sup> The magnetic field in an erbium tetrahedral channel is always different from zero. The experimental value  $\kappa = 0.20 \pm 0.02$  can therefore be considered as an indication that the muon is localized in an octapore of the crystal lattice.

We consider now muon spin relaxation in the paramagnetic state of erbium at  $T > T_N$ . A theory of this process in rare-earth metals has been developed.<sup>4</sup> It was shown there that the dependence  $P(t)$  of the muon polarization on time in a zero external magnetic field has the form

$$P(t) = \langle \cos^2 \theta \rangle \exp(-2G_{\perp}t) + \langle \sin^2 \theta \rangle \exp(-(G_{\perp} + G_{\parallel})t). \quad (7)$$

Here

$$G_{\parallel} = \frac{\gamma_{\mu}^2}{2} \int_{-\infty}^{+\infty} \langle H_x(t') H_x(t) \rangle dt', \quad (8)$$

$$G_{\perp} = \frac{\gamma_{\mu}^2}{2} \int_{-\infty}^{+\infty} \langle H_x(t') H_x(t) \rangle dt' = \frac{\gamma_{\mu}^2}{2} \int_{-\infty}^{+\infty} \langle H_y(t') H_y(t) \rangle dt'$$

are the correlation functions of the fluctuating magnetic field  $\mathbf{H}$  at the point of localization of a muon (octapore or tetrapore);  $\gamma_{\mu} = e/m_{\mu}c$  is the gyromagnetic ratio for a muon;  $H_x$ ,  $H_y$  and  $H_z$  are components of the field  $\mathbf{H}$ , associated with the coordinate axes of the individual single crystals in such a way that the  $z$  axis coincides with the crystal hexagonal axis;  $\langle \cos^2 \theta \rangle$  is the mean square cosine of the angle between the directions of the hexagonal axes of the individual erbium single crystals and the direction of muon polarization. At studied-specimen orientation corresponding to the angle  $\alpha = 0$  we have  $\langle \cos^2 \theta \rangle = \langle \cos^2 \vartheta \rangle$ .

The two terms in Eq. (7) for  $P(t)$  describe relaxation of the longitudinal and transverse components of the muon spin in an individual erbium single crystal, and are obtained as a result of subsequent averaging over all the single crystals of the specimen. It follows from Eq. (7) that relaxation of the spin component directed longitudinally along the hexagonal crystal axis is described by the correlation function  $2G_{\parallel}$ , while relaxation of the component directed, for example, along the  $x$  axis is described by the correlation function  $G_{\perp} + G_{\parallel}$ . Equation (8) for the correlators  $G_{\perp}$  and  $G_{\parallel}$  can be written in a clearer form for the case when the correlation of the components of the fluctuating field depends on time:

$$\begin{aligned} \langle H_x(t') H_x(t) \rangle &= \langle H_y(t') H_y(t) \rangle = \langle H_x^2 \rangle \exp(-|t-t'|/\tau_x), \\ \langle H_z(t') H_z(t) \rangle &= \langle H_z^2 \rangle \exp(-|t-t'|/\tau_z). \end{aligned} \quad (9)$$

Here  $\langle H_x^2 \rangle = \langle H_y^2 \rangle$  and  $\langle H_z^2 \rangle$  are the mean squares of the components of the fluctuating magnetic field at the point where the muon is localized;  $\tau_x = \tau_y$  and  $\tau_z$  are the characteristic lifetimes of the magnetic fields produced by the fluctuations. Equation (8) is then written in the form

$$G_{\parallel} \approx \gamma_{\mu}^2 \langle H_x^2 \rangle \tau_x, \quad G_{\perp} \approx \gamma_{\mu}^2 \langle H_x^2 \rangle \tau_x. \quad (10)$$

The natural assumption that the fluctuations of the field  $\mathbf{H}$  in the paramagnetic state are connected with fluctuation-produced fragments of the magnetically ordered state that is characteristic of temperatures  $T < T_N$ , leads to the inequality of correlation functions

$$G_{\parallel} > G_{\perp}. \quad (11)$$

The inequality (11) is a consequence of the fact that the magnetic moments of the erbium atoms in the ordered state produce at the muon a magnetic field directed along the hexagonal crystal axis, for which as a result, only the component  $H_z$  is not zero. This character of the fluctuations of the field  $\mathbf{H}$  at  $T > T_N$  is confirmed experimentally, as can be seen from Fig. 3 and Eq. (4). The difference between  $G_{\parallel}$  and  $G_{\perp}$  will evidently show up only sufficiently close to  $T_N$ . As the temperature is increased, the fluctuating field becomes isotropic and the values of  $G_{\parallel}$  and  $G_{\perp}$  become comparable.

Self-consistent field theory (see Moriya<sup>5</sup>) allows a more detailed prediction of the temperature dependence of the correlators  $G_{\parallel}$  and  $G_{\perp}$  near the Néel temperature to be made. According to this theory,  $G_{\perp}^{\text{th}} \sim (T - T_N)^{-1/2}$ , i.e., it becomes infinite as  $T \rightarrow T_N$ , while  $G_{\parallel}^{\text{th}}(T)$  remains finite at all temperatures and has no singularity at  $T = T_N$ . These theoretical predictions are general for arbitrary antiferromagnetic second-order transitions in metals with easy axis anisotropy. The  $G_{\parallel}^{\text{th}}(T)$  dependence predicted is observed for a number of magnetic phase transitions.<sup>5</sup> We compare below the predictions of the theory with the experimental results of the present work.

The correlators  $G_{\parallel}$  and  $G_{\perp}$  which determine the  $P(t)$  dependence (7) can be found by measuring the muon-spin relaxation for two specimen orientations. In this work  $P(t)$  was measured for specimen orientations determined by the angles  $\alpha = 0$  and  $\alpha = \pi/2$ . The corresponding expressions for the longitudinal ( $\alpha = 0$ ) and transverse ( $\alpha = \pi/2$ )  $P(t)$  depen-

dences have the form

$$P_{\parallel}(t) = x \exp(-2G_{\perp}t) + (1-x) \exp(-(G_{\parallel}+G_{\perp})t), \quad \alpha=0,$$

$$P_{\perp}(t) = \frac{1}{2}(1-x) \exp(-2G_{\perp}t) + \frac{1}{2}(1+x) \exp(-(G_{\parallel}+G_{\perp})t),$$

$$\alpha = \pi/2. \quad (12)$$

$$x = \langle \cos^2 \theta \rangle = 0.533.$$

It was pointed out in §2 that muon-spin relaxation in erbium depends exponentially on time over the whole temperature range studied. We can therefore write

$$P_{\parallel}(t) = \exp(-\Lambda_{\parallel}t), \quad P_{\perp}(t) = \exp(-\Lambda_{\perp}t). \quad (13)$$

From Eqs. (12) and (13) follow, to a high degree of accuracy, the relations

$$\Lambda_{\parallel} = 2G_{\perp}x + (G_{\parallel}+G_{\perp})(1-x), \quad \alpha=0, \quad (14)$$

$$\Lambda_{\perp} = 2G_{\perp} \cdot \frac{1}{2}(1-x) + (G_{\parallel}+G_{\perp}) \cdot \frac{1}{2}(1+x), \quad \alpha = \pi/2.$$

Whence

$$G_{\parallel} = \frac{\Lambda_{\parallel} + \Lambda_{\perp}}{4} + \frac{5+x}{3x-1} \frac{\Lambda_{\perp} - \Lambda_{\parallel}}{4}, \quad (15)$$

$$G_{\perp} = \frac{\Lambda_{\parallel} + \Lambda_{\perp}}{4} - \frac{3-x}{3x-1} \frac{\Lambda_{\perp} - \Lambda_{\parallel}}{4}.$$

The experimental temperature dependences  $G_{\parallel}(T)$  and  $G_{\perp}(T)$  are shown in Fig. 5. The values shown in Fig. 5 were obtained from Eq. (15) by using the smooth  $\Lambda_{\parallel}(T)$  and  $\Lambda_{\perp}(T)$  dependences shown in Fig. 3. These dependences are a smooth interpolation of the corresponding experimental values of  $\Lambda$  ( $\alpha = 0$ ) and  $\Lambda$  ( $\alpha = \pi/2$ ) measured at discrete temperatures. It can be seen from Fig. 5 that  $G_{\perp}$  is weakly dependent on temperature and almost coincides with its paramagnetic limit  $G_{\perp}^{\text{para}} = G_{\parallel}^{\text{para}} = 0.5 \mu\text{s}^{-1}$ , measured at  $T \approx 300$  K, when  $\Lambda_{\parallel} = \Lambda_{\perp} = 1 \mu\text{s}^{-1}$  (see Fig. 3).

The correlator  $G_{\parallel}$  increases with decreasing temperature and in agreement with Eq. (11) exceeds  $G_{\perp}$ . The difference between  $G_{\parallel}$  and  $G_{\perp}$  is observed over a fairly wide temperature range  $T_N < T \lesssim 130$  K. It follows from Eq. (5) that the experimental  $G_{\parallel}(T)$  does not have a singularity as  $T \rightarrow T_N$  and does not satisfy the theoretical relation  $G_{\parallel}^{\text{th}} \sim (T - T_N)^{-1/2}$  for second-order antiferromagnetic transitions. The absence of a singularity in  $G_{\parallel}(T)$  at the phase-transition temperature also follows from the limited range of

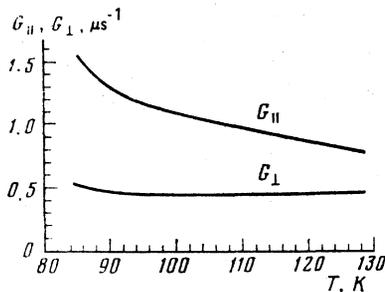


FIG. 5. Experimental temperature dependences of the correlation functions (15)  $G_{\parallel}(T)$  and  $G_{\perp}(T)$ .

the directly measured values of  $\Lambda_{\parallel}$  and  $\Lambda_{\perp}$ , which do not exceed as  $T \rightarrow T_N$  double the value  $\Lambda$  ( $T \approx 300$  K)  $= 1 \mu\text{s}^{-1}$  of the muon-spin relaxation rate in the paramagnetic state at high temperatures. This difference between the experimental and theoretical  $G_{\parallel}(T)$  dependences indicates that the phase transition in erbium at  $T_N = 84.4$  K is not a pure second-order transition and can be interpreted as a manifestation of a weak first order transition.

Zvi and Walker<sup>6</sup> predicted theoretically that there would be weak first order transitions associated with fluctuation processes at the Néel temperature for terbium, dysprosium and holmium. They pointed out, as an experimental confirmation of their theory of weak first-order transitions for helicoidal structures, the small ( $\sim 10^{-5}$ ) jump in the continuity of thermal expansion observed<sup>7</sup> for holmium. In the present study we have obtained an experimental indication of a weak first order transition for a sinusoidal antiferromagnet.

The experimental values of  $G_{\parallel}$  and  $G_{\perp}$  shown in Fig. 5 can be used for a qualitative estimate of the lifetime of the ordered state produced in erbium by fluctuations at  $T > T_N$ . From the approximate Eq. (10) and the experimental value  $G_{\parallel}/G_{\perp} < 3$  (see Fig. 5) it follows that at  $T > T_N$

$$\frac{\langle H_z^2 \rangle \tau_z}{\langle H_x^2 \rangle \tau_x} < 3,$$

i.e.,

$$\tau_z \leq 3\tau_x, \quad (16)$$

if we take  $\langle H_z^2 \rangle \approx \langle H_x^2 \rangle$ . It can be seen from Eq. (16) that the time  $\tau_z$  for formation of fragments of the ordered state over the temperature range studied is only slightly greater than the characteristic time  $\tau_x = \tau_y \sim 10^{-12}$  s of atomic spin oscillations in the paramagnetic state at high temperatures.

It is interesting to note that the experimental  $G_{\parallel}(T)$  relaxation at  $T_N < T < 130$  K shown in Fig. 5 is reasonably well represented by the expression

$$G_{\parallel}(T) = G_{\parallel}(T_N) - A(1 - T_N/T)^{\beta},$$

where  $\beta = 0.48$ . We cannot suggest any interpretation of this form of the  $G_{\parallel}(T)$  temperature dependence.

As was pointed out above, the slow muon spin relaxation observed experimentally in the magnetically ordered state at  $T < T_N$  occurs under the action of the rapidly oscillating disordered components of the atomic magnetic moments. According to Ivanter and Fomichev<sup>4</sup> [see also Eq. (7)], the expression for  $P(t)$  then has the form

$$P(t) = \langle \cos^2 \theta \rangle \exp(-2G_{\perp}t), \quad T < T_N. \quad (17)$$

The correlator  $G_{\perp}$  now describes fluctuations of the transverse magnetic fields  $H_x$  and  $H_y$  (as for  $T > T_N$ ,  $\langle H_x^2 \rangle = \langle H_y^2 \rangle$ ) in the presence of a constant magnetic field  $H_z$ , ordered by a sinusoidal wave with wave vector directed along the  $z$  axis. The exponential dependence  $P(t) \sim e^{-\Lambda t}$  leads to the expression

$$\Lambda = 2G_{\perp} \approx 2\gamma_{\mu}^2 \langle H_x^2 \rangle \tau_x, \quad T < T_N. \quad (18)$$

It follows from relation (17) that the rate of the slow muon

spin relaxation at  $T < T_N$  is independent of the specimen orientation. This conclusion is confirmed experimentally (see Fig. 3).

It can be seen from Fig. 5 that the correlator  $G_1$  remains constant at  $T_N < T < 300$  K. The experimental equality  $\Lambda(T < T_N) \approx \Lambda(T = 300 \text{ K})$  (see Fig. 3) and Eq. (18) show that the magnitude of  $G_1$  does not change also on going to the magnetically ordered state, i.e., at  $T < T_N$ . The roughly constant value of  $G_1$  over a wide temperature range means that the reduction in the transverse magnetic fields  $H_x$  and  $H_y$ , which comes about when the longitudinal component of the field  $H_z$  is ordered, is compensated for by an increase in the transverse correlation time  $\tau_x = \tau_y$ .

#### §4. CONCLUSION

The following deductions about the properties of the antiferromagnetic phase transition and the magnetically ordered state of erbium can be drawn from measurement of muon spin relaxation in a magnetically oriented specimen of this metal:

1. The formation of ordered fluctuations is observed over a wide temperature range  $T_N < T \lesssim 130$  K of the paramagnetic state, and their magnetic structure corresponds to the antiferromagnetic ordering for  $T < T_N$ .

2. Fluctuations of the longitudinal and transverse components, relative to the direction of the hexagonal crystal axis, of the magnetic field  $\mathbf{H}$  in the interstices of the erbium crystal lattice are described by two correlation functions  $G_{\parallel}(T)$  and  $G_{\perp}(T)$ . The values of  $G_{\parallel}$  and  $G_{\perp}$  are different in the region of ordered fluctuations of the field  $\mathbf{H}$  and coincide at high temperatures when the magnetic fluctuations are isotropic. The correlator  $G_{\parallel}$  grows to a limiting value with decreasing temperature as  $T \rightarrow T_N$ . The limit on  $G_{\parallel}$  follows directly from the limit on the temperature dependence  $\Lambda(T)$  of the muon spin relaxation rate at  $T < T_N$ . The correlator  $G_{\perp}$

stays constant over the whole temperature range studied at  $T > T_N$ . The limit on  $G_{\parallel}$  as  $T \rightarrow T_N$  contradicts the theoretical dependence  $G_{\parallel}^{\text{th}} \sim (T - T_N)^{-1/2}$  for a second-order antiferromagnetic transition and can be considered as an experimental indication that the antiferromagnetic ordering of erbium at  $T_N = 84.4$  K is a weak first-order phase transition.

3. The correlator  $G_1$  does not change when erbium goes into the magnetically ordered state at  $T < T_N$ . The correlator then describes the fluctuations of disordered components of the magnetic field  $\mathbf{H}$  on a muon in the interstitial pores of the crystal.

4. In the magnetically ordered state at  $T < T_N$  an appreciable fraction  $\kappa = 0.20 \pm 0.02$  of muons do not experience the action of the constant magnetic field produced by ordered magnetic moments. This result can be explained by the localization of muons in octahedral interstices of the erbium crystal lattice, since the magnetic field is not zero in tetrahedral interstices of erbium.

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