Study of dysprosium by a muon diffusion technique

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An experimental study by a muon diffusion technique reveals that dysprosium in its paramagnetic state exhibits a magnetically ordered phase characterized by fluctuations with a correlation time $\sim 10^{-12}$ s over the broad temperature range $T_N < T \lesssim 230$ K, where $T_N = 180$ K is the Néel point. The fluctuations have the structure of the antiferromagnetic state of dysprosium and are described by the two correlation functions $G_\perp(T)$ and $G_\parallel(T)$. The correlation function $G_\perp(T)$ does not have a singularity as $T \to T_N$. This result contradicts the interpretation of a second-order phase transition at $T_N = 180$ K in dysprosium. It can be interpreted as a weak first-order phase transition according to a renormalization group calculation. A new method is proposed for measuring the diffusion of a positive muon in a helicoidal antiferromagnet. The diffusion of muons in dysprosium at $100 < T < 175$ K is a tunneling process.

I. INTRODUCTION

Studies by the renormalization group technique lead to a fundamental limitation on the number of possible second-order phase transitions. This result has stimulated a more detailed experimental study of phase transitions which have historically been regarded as second-order. In the present paper we report the use of a muon technique to study the phase transition of dysprosium from the paramagnetic state to the antiferromagnetic state, which occurs at the Néel point, $T_N = 180$ K. Renormalization group theory predicts that this cannot be a second-order transition.[1]

The antiferromagnetic phase transition in dysprosium has been studied in several places.[2–5] The temperature dependence $M(T)$ of the ordered magnetic moment in the antiferromagnetic state of dysprosium, measured by a Mössbauer technique, can be approximated well at $(T_N - T) \gtrsim 1$ K by the expression $M(T) \propto (1 - T/T_N)^\beta$ with the parameter values[2] $T_N = (180.4 \pm 0.3)$ K and $\beta = 0.335 \pm 0.010$. This temperature dependence is qualitatively consistent with a second-order phase transition. Measurements[3] of the temperature dependence of the dysprosium lattice constants with a relative error of 10^{-6} have revealed no structural features of any sort near the temperature T_N; this result is again characteristic of a second-order phase transition. The temperature dependence of the electrical resistance, $R(T)$, and of the heat capacity, $C(T)$, of dysprosium were studied in Refs. 4 and 5 at temperatures $T > T_N$ and $T < T_N$. The experimental behavior of $R(T)$ and $C(T)$ was analyzed in detail in Ref. 5. It follows from that analysis that at $T_N = 180$ K dysprosium undergoes a complex phase transition which is difficult to describe unambiguously. The observed behavior $R(T)$ and $C(T)$ was explained qualitatively on the basis of the Kosterlitz-Thouless model in Ref. 5.

The use of a muon technique to study the properties of the antiferromagnetic transition in dysprosium is based on measurements of the temperature dependence of the relaxation rate, Λ, of the muon spin. This rate is proportional to the correlation time for the fluctuations of the magnetically ordered phase in the paramagnetic state of a metal. In the present study we show that the temperature dependence $\Lambda(T)$ reveals only a very slight increase as the temperature is lowered, and there is no singularity in the limit $T \to T_N$. This behavior of $\Lambda(T)$ can be explained in terms of a weak first-order phase transition.[6]

A second direction in this study was to investigate the diffusion of muon through a dysprosium crystal. In the method proposed here for determining the time τ for a diffusive hop of muon between neighboring interstitial positions in the crystal lattice, we make use of the particular features of the helicoidal magnetic structure of the antiferromagnetic state of dysprosium. These structural features make it possible to measure the dependence $\tau(T)$ over the temperature range $T = 90$–180 K. The results show that muon diffusion in dysprosium is a tunneling process. Preliminary results on the muon diffusion coefficient in dysprosium were reported in Ref. 7. The experiment was carried out in the muon channel of the synchrocyclotron at Gatchina.

2. EXPERIMENTAL PROCEDURE

The dysprosium sample consisted of six disks 35 mm in diameter and 5 mm thick. The disks were originally made of a polycrystalline material with an impurity content $< 0.01\%$ by weight. After the sample was annealed during two-sided compression, the dimensions of the crystal grains increased to 3–10 mm, and their hexagonal c axes became oriented predominantly perpendicular to the plane of the base of the disk. The measurements were carried out in two orientations of the sample, corresponding to angles $\alpha = 0$ and $\alpha = \pi/2$ between the muon polarization direction and the direction of the predominant orientation of the hexagonal axes c of the crystal grains in the sample. The temperature of the sample, held in a special cryostat, was determined with germanium pickups with a relative error $\delta T_{\text{rel}} = 0.05$. The absolute error in the measurements of the sample tempera-

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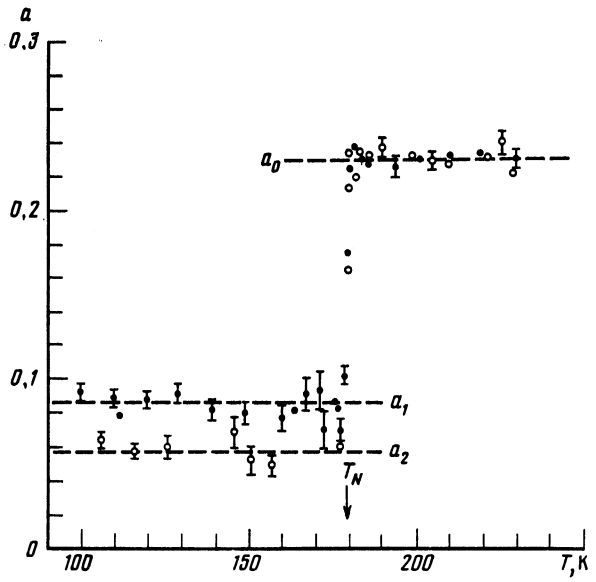

FIG. 1. Temperature dependence $a(T)$ of the measured asymmetry coefficient of the positron distribution resulting from the decay $\mu^+ \to e^+$ in dysprosium for two orientations of the sample: ○—$\alpha = 0$; ●—$\alpha = \pi/2$. The values given here for a have been averaged over intervals $\Delta T = 1$ K; $T_N = 180$ K.

ture was found by calibrating these pickups: $\delta T_{abs} = 0.3$ K at $T = 150$–200 K.

In the experiments we measure the time dependence

$$N(t) = N_0 e^{-t/\tau_0}(1 + ae^{-\Lambda t}) + B, \qquad (1)$$

which is the time dependence of the number of positrons from the decay $\mu^+ \to e^+$ which are emitted along the muon polarization direction in a zero external magnetic field. Here $\tau_0 = 2.2 \cdot 10^{-6}$ s is the muon lifetime, a is the measured asymmetry coefficient of the angular distribution of positrons from the decay $\mu^+ \to e^+$, Λ is the relaxation rate of the muon spin, and B is the background. Expression (1) assumes that the relaxation of the muon spin in dysprosium occurs in accordance with the exponential law $P(t) = e^{-\Lambda t}$. The experimental results confirm this functional dependence of $P(t)$.

Figure 1 shows the temperature dependence $a(T)$ of the measured asymmetry coefficient over the temperature range $T = 100$–300 K for two orientations of the sample, corresponding to the angles $\alpha = 0$ and $\alpha = \pi/2$. For clarity, the values of a shown in Fig. 1 have been averaged over intervals $\Delta T = 1$ K. The quantities

$$\begin{aligned} &a_0 = 0.2293 \pm 0.0010, && T > T_N, && \alpha = 0, && \alpha = \pi/2, \\ &a_1 = 0.0859 \pm 0.0015, && T < T_N, && \alpha = \pi/2, \\ &a_2 = 0.0582 \pm 0.0023, && T < T_N, && \alpha = 0 \end{aligned} \qquad (2)$$

shown in this figure are the average values of the coefficient a for the paramagnetic ($T > T_N$) and antiferromagnetic ($T < T_N$) states of the dysprosium. The reason for the decrease in a at the transition to the antiferromagnetic state is an unobservably fast precession of the muon spin in the strong interstitial magnetic fields at $T < T_N$. The interstitial magnetic fields in the antiferromagnetic state of dysprosium are $H_\mu > 10^4$ Oe and are directed perpendicular to the hexagonal c axis of the crystal.[8] The coefficients a_1 and a_2 therefore pertain to the relaxation of longitudinal components (longitudinal with respect to the field H_μ) of the muon spin. The corresponding decrease in the observed muon polarization is

$$P_{obs} = a_{1,2}/a_0 = \langle \cos^2 \theta \rangle, \qquad (3)$$

where $\langle \cos^2\theta \rangle$ is the mean square cosine of the angle between the muon polarization direction and the directions of the local fields H_μ. For the helicoidal magnetic structure of dysprosium, the quantity $\langle \cos^2\theta \rangle$ can be written

$$\langle \cos^2 \theta \rangle = {}^1/_4 \sin^2 \alpha (3\langle \cos^2 \vartheta \rangle - 1) + {}^1/_2 (1 - \langle \cos^2 \vartheta \rangle). \qquad (4)$$

Here $\langle \cos^2\vartheta \rangle$ is the mean square cosine of the angle ϑ, between the direction of the c axes of the individual crystal grains and the direction of their average orientation, i.e., the direction of the normal to the plane of the disk. For $\alpha = \pi/2$ and $\alpha = 0$ we find from expressions (3) and (4), respectively,

$$\begin{aligned} &a_1/a_0 = {}^1/_4 (1 + \langle \cos^2 \vartheta \rangle), \quad \alpha = \pi/2, \\ &a_2/a_0 = {}^1/_2 (1 - \langle \cos^2 \vartheta \rangle), \quad \alpha = 0. \end{aligned} \qquad (5)$$

From (5) we find

$$2a_1/a_0 + a_2/a_0 = 1, \qquad (6)$$

which agrees well with the experimental values listed for the coefficients a_0, a_1, and a_2 in (2):

$$2a_1/a_0 = 0.749 \pm 0.018, \quad a_2/a_0 = 0.250 \pm 0.013. \qquad (7)$$

From (5) we also find the value

$$\langle \cos^2 \vartheta \rangle = 0.500 \pm 0.021. \qquad (8)$$

Figure 2 shows the temperature dependence $\Lambda(T)$ of

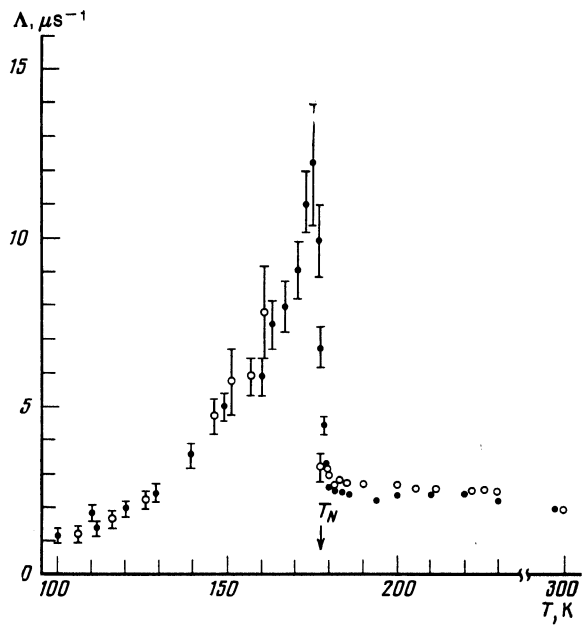

FIG. 2. The temperature dependence, $\Lambda(T)$, of the relaxation rate of the muon spin in dysprosium. ○—$\alpha = 0$; ●—$\alpha = \pi/2$. The values given for Λ have been averaged over intervals $\Delta T = 1$ K; $T_N = 180$ K.

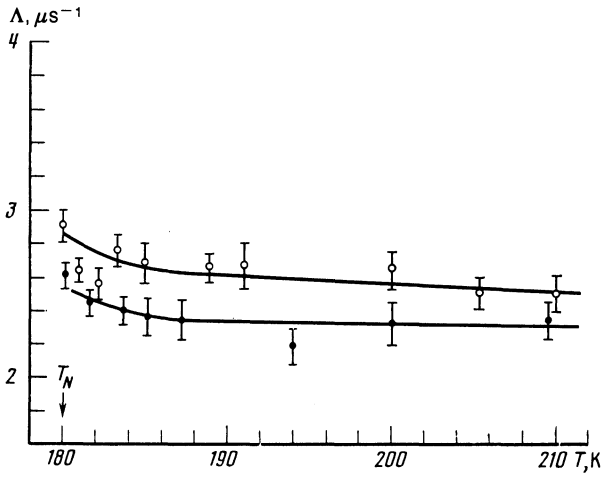

FIG. 3. Temperature dependence, $\Lambda(T)$, at $T > T_N$. ○—$\alpha = 0$; ●—$\alpha = \pi/2$. The values of Λ at $T = 300$ K for $\alpha = 0$ and $\alpha = \pi/2$ are the same: $\Lambda = 1.9 \pm 0.1\ \mu s^{9-1}$.

the relaxation rate of the muon spin in dysprosium for $\alpha = 0$ and $\alpha = \pi/2$ over the temperature range $T = 100$–300 K, again averaged over intervals $\Delta T = 1$ K. The same $\Lambda(T)$ dependence in the paramagnetic state at $T = 180$–210 K is shown in Fig. 3, on a larger scale.

It can be seen from Figs. 2 and 3 that the relaxation rate Λ increases as $T \to T_N$ in the paramagnetic state, and over a rather broad temperature interval $T_N < T \lesssim 230$ K we have

$$\Lambda(\alpha=0) > \Lambda(\alpha=\pi/2). \tag{9}$$

The reason for the increase in Λ as $T \to T_N$ is the decrease in the oscillation frequency of the atomic spins of dysprosium due to the fluctuations which develop in the magnetically ordered phase in the paramagnetic state. Inequality (9) confirms that the magnetically ordered phase which develops from fluctuations at $T > T_N$ has the structure of the antiferromagnetic state of dysprosium, with the local magnetic field H_μ at the muon directed perpendicular to the hexagonal c axis of the single crystal.

It also follows from Figs. 2 and 3 that the increase in Λ in the paramagnetic state is bounded, and there is no singularity at the phase-transition temperature. From Fig. 3 we see that the maximum values of $\Lambda(\alpha = 0)$ and $\Lambda(\alpha = \pi/2)$ found as $T \to T_N$ can be estimated to be

$$\begin{aligned} \Lambda_\parallel(T_N) &= 2{,}86\ \mu s^{-1} \text{ for } \alpha = 0, \\ \Lambda_\perp(T_N) &= 2{,}55\ \mu s^{-1} \text{ for } \alpha = \pi/2. \end{aligned} \tag{10}$$

In (10) we have adopted the streamlined notation $\Lambda_\parallel \equiv \Lambda(\alpha = 0)$ and $\Lambda_\perp \equiv \Lambda(\alpha = \pi/2)$, which we will also be using below.

The antiferromagnetic phase transition observed experimentally in dysprosium at $T_N = 180$ K occurs over a fairly broad temperature interval $\delta T_N \approx 3$ K, in contrast with the case in erbium.[6] The values of the asymmetry coefficients a_1 and a_2 corresponding to the magnetically ordered state are reached only at $T \approx 177$ K. This feature of the antiferromagnetic phase transition in this dysprosium sample is illustrated by Fig. 4, which shows the experimental dependences $a(T)$ and $\Lambda(T)$ in the immediate vicinity of $T_N = 180$ K. The nonzero width of the antiferromagnetic phase transition in dysprosium is attributed to deformation of the individual crystal grains which occurs as this block sample is cooled. As a result of the deformation, the Néel temperatures of the individual crystal grains change, causing expansion of the phase transition observed experimentally. This explanation is confirmed by the plot of $x(T)$ in Fig. 5—the temperature dependence of the fraction of the sample which is in the antiferromagnetic state. The values of x shown in Fig. 5 were found by the maximum likelihood method from the experimental curves of $P(t)$, which were described near the temperature T_N by $(0 \leqslant x \leqslant 1)$

$$\begin{aligned} P(t) &= a_0(1-x)\exp\{-\Lambda_\perp(T_N)t\} \\ &\quad + a_1 x \exp(-\Lambda_0 t) \text{ for } \alpha = \pi/2, \\ P(t) &= a_0(1-x)\exp\{-\Lambda_\parallel(T_N)t\} \\ &\quad + a_2 x \exp(-\Lambda_0 t) \text{ for } \alpha = 0. \end{aligned} \tag{11}$$

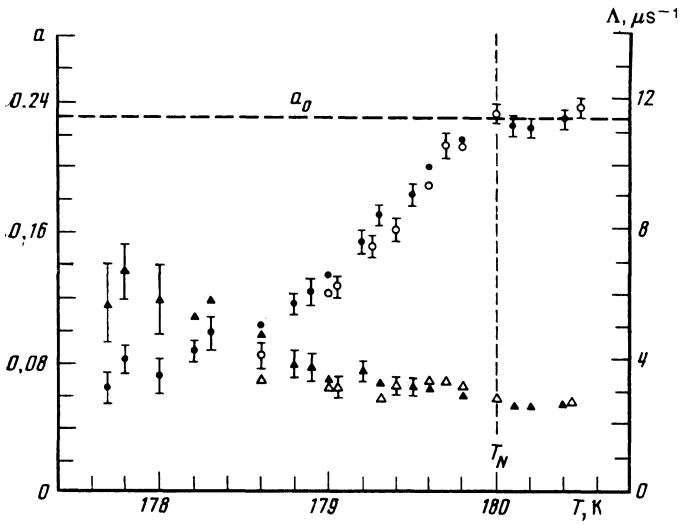

FIG. 4. Circles—The temperature dependence $a(T)$; triangles—$\Lambda(T)$. $T \lesssim T_N$. Open points—$\alpha = 0$; filled points $-\ \alpha = \pi/2$.

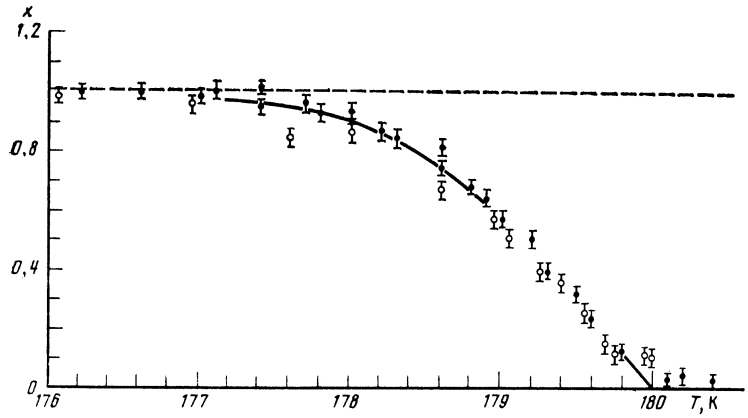

FIG. 5. The temperature dependence $x(T)$ of the fraction of the dysprosium sample in the antiferromagnetically ordered state near the phase-transition temperature T_N; ○ – $\alpha = 0$; ● – $\alpha = \pi/2$,

the coefficients a_0, a_1, and a_2 here are fixed, equal to the values given in (2); the values of the parameters $\Lambda_\perp(T_N)$ and $\Lambda_\parallel(T_N)$ are given in (10); and $\Lambda_0 = 10\ \mu s^{-1}$ is the maximum relaxation rate Λ at $T < T_N$. Figure 2 shows the dependence $\Lambda(T)$ at $T < T_N$ (see also Fig. 7 below); this dependence is the result of diffusion of muons through the dysprosium lattice. The diffusion of muons in the magnetically ordered state of dysprosium is examined in detail in Section 4.

The first and second terms in expressions (11) for $P(t)$ describe relaxation of the muon spin in respectively the paramagnetic ($x = 0$) and antiferromagnetic ($x = 1$) states of the metal. The results shown for $x(T)$ for the two orientations of the sample ($\alpha = 0$ and $\alpha = \pi/2$) in Fig. 5 are the same, providing convincing support for the model which we are discussing here for the nonzero width of the antiferromagnetic phase transition of this dysprosium sample.

3. FLUCTUATIONS IN THE PARAMAGNETIC STATE OF DYSPROSIUM

Ivanter and Fomichev[8] have derived a theory which can relate the quantity Λ, which can be measured experimentally, with parameters characterizing the paramagnetic state of a metal. They showed that the time dependence $P(t)$ of the polarization of muons in a zero external magnetic field is

$$P(t) = \langle \cos^2 \chi \rangle \exp(-2G_\perp t) + \langle \sin^2 \chi \rangle \exp[-(G_\perp + G_\parallel)t]. \quad (12)$$

Here

$$G_\parallel = \frac{\gamma^2}{2} \int_{-\infty}^{+\infty} \langle H_z(t') H_z(t) \rangle dt',$$
$$G_\perp = \frac{\gamma^2}{2} \int_{-\infty}^{+\infty} \langle H_x(t') H_x(t) \rangle dt' = \frac{\gamma^2}{2} \int_{-\infty}^{+\infty} \langle H_y(t') H_y(t) \rangle dt' \quad (13)$$

are the correlation functions of the fluctuating magnetic field $\mathbf{H}_\mu$ at the muon (an octahedral or tetrahedral void); $\gamma = e/m_\mu c$ is the gyromagnetic ratio for the muon; H_x, H_y, and H_z are the components of $\mathbf{H}_\mu$ along the coordinate axes of an individual single crystal (the z axis runs along the hexagonal axis of the crystal); and $\langle \cos^2 \chi \rangle$ is the mean square cosine of the angle between the hexagonal axes of the individual dysprosium crystals and the muon polarization direction. Expression (12) is valid when the oscillation frequency of the electron spins of the metal atoms which produce the magnetic field $\mathbf{H}_\mu$ at the muon is substantially higher than the frequency at which the muon precesses under the influence of this field.

The first and second terms in expression (12) for $P(t)$ describe the relaxation of the longitudinal and transverse components, respectively, of the muon spin in an individual dysprosium crystal, averaged over all of the crystals in the sample. Expressions (13) for the correlation functions $G_\parallel$ and $G_\perp$ can be written in a more transparent form in the case in which the correlations of the fluctuating field depend exponentially on the time; e.g.,

$$\langle H_x(t') H_x(t) \rangle = \langle H_x^2 \rangle \exp(-|t-t'|/\tau_x). \quad (14)$$

Expressions (13) for the correlation functions are then written as

$$G_\parallel \approx \gamma^2 \langle H_z^2 \rangle \tau_z, \quad G_\perp \approx \gamma^2 \langle H_x^2 \rangle \tau_x. \quad (15)$$

Here $\langle H_x^2 \rangle = \langle H_y^2 \rangle$ and $\langle H_z^2 \rangle$ are the mean square components of the fluctuating field $\mathbf{H}_\mu$; and $\tau_x = \tau_y$ and τ_z are the lifetimes of the magnetic fields H_μ which develop from fluctuations.

The correlation functions $G_\parallel$ and $G_\perp$ can be found by measuring the polarization $P(t)$ for two orientations of the sample, corresponding to two values of the angle χ. In the present experiments we measured the polarization $P(t)$ for the two orientations of the sample defined by the angles $\alpha = 0$ and $\alpha = \pi/2$. The corresponding expressions for $P(t)$ are

$$P_\parallel(t) = y \exp(-2G_\perp t) + (1-y) \exp\{-(G_\parallel + G_\perp)t\}, \quad \alpha = 0,$$
$$P_\perp(t) = {}^1/_2(1-y) \exp(-2G_\perp t) + {}^1/_2(1+y) \exp\{-(G_\parallel + G_\perp)t\}, \quad \alpha = \pi/2, \quad (16)$$
$$y = \langle \cos^2 \theta \rangle = 0.500 \pm 0.021.$$

For the exponential functions

$$P_\parallel(t) = \exp(-\Lambda_\parallel t), \quad P_\perp(t) = \exp(-\Lambda_\perp t) \quad (17)$$

we find the following relations, which are quite accurate enough for our purposes, from expressions (16):

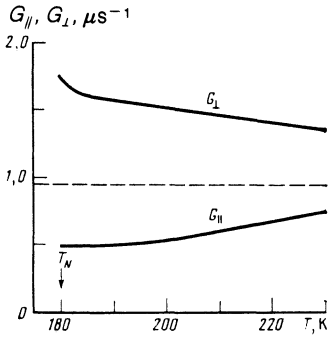

FIG. 6. Temperature dependence of the correlation functions $G_{\perp}(T)$ and $G_{\parallel}(T)$ in the antiferromagnetic state of dysprosium.

$$\Lambda_{\parallel}=2G_{\perp}y+(G_{\parallel}+G_{\perp})(1-y),$$
$$\Lambda_{\perp}=G_{\perp}(1-y)+{}^{1}/_{2}(G_{\parallel}+G_{\perp})(1-y). \qquad (18)$$

Hence

$$G_{\parallel}=\frac{\Lambda_{\parallel}+\Lambda_{\perp}}{4}-\frac{5+y}{3y-1}\frac{\Lambda_{\parallel}-\Lambda_{\perp}}{4},$$
$$G_{\perp}=\frac{\Lambda_{\parallel}+\Lambda_{\perp}}{4}+\frac{3-y}{3y-1}\frac{\Lambda_{\parallel}-\Lambda_{\perp}}{4}. \qquad (19)$$

Figure 6 shows experimental results on $G_{\parallel}(t)$ and $G_{\perp}(T)$ over the temperature interval $180<T<230$ K. These results were found from (19) through the use of the smooth $\Lambda(T)$ curves shown in Fig. 3; these smooth curves are interpolations of the measured values of Λ.

It can be seen from Fig. 6 that over the broad temperature range $T<230$ K the correlation functions $G_{\parallel}(T)$ and $G_{\perp}(T)$ differ from their paramagnetic limit (the dashed line in this figure),

$$G_{\parallel}^{\text{Para}}=G_{\perp}^{\text{Para}}=\frac{\Lambda_{\perp}+\Lambda_{\parallel}}{4}=0.95\ \mu s^{-1},$$

which corresponds to $\Lambda_{\perp}=\Lambda_{\parallel}$. It can be seen from Fig. 3 that this limit is reached at $T\leqslant 300$ K.

The correlation function $G_{\perp}$ increases with decreasing temperature, as it should if the phase which develops from fluctuations has the structure of a magnetically ordered state. As $T\to T_N$ the correlation function $G_{\perp}$ increases but remains bounded, and it has no singularity at $T=T_N$. That the $G_{\perp}(T)$ dependence does not have a singularity at the phase-transition temperature also follows from the boundednesss of the directly measurable quantities $\Lambda_{\parallel}$ and $\Lambda_{\perp}$ (Fig. 3). Furthermore, the maximum values given in (10) for $\Lambda_{\parallel}(T_N)$ and $\Lambda_{\perp}(T_N)$ of the paramagnetic state of dysprosium at $T=180$ K increase essentially not at all within the width δT_N of the experimental phase transition, as was shown by varying these quantities in a comparison of the functions $P(t)$ in (11) with experimental data. Over the temperature range of the measurements, $180<T<230$ K, the correlation function $G_{\perp}(T)$ does not satisfy the dependence $G_{\perp}\sim(T-T_N)^{-\beta}$ which is characteristic of a second-order phase transition. This result agrees with the calculations of Ref. 1, carried out on the basis of renormalization group theory. This result may be evidence that a so-called weak first-order phase transition occurs at $T=180$ K is dysprosium.

Expressions (15) can be used to estimate the scale time τ for the fluctuations of a magnetically ordered phase in the paramagnetic state of dysprosium. From (15) and Fig. 6 we have

$$\frac{G_{\perp}}{G_{\perp}^{\text{Para}}}\approx\frac{\langle H_x^2\rangle\tau_x}{\langle H_x^2\rangle^{\text{Para}}\tau^{\text{Para}}}\lesssim 2, \qquad (20)$$

from which we see that τ_x is only slightly above the paramagnetic value

$$\tau^{\text{Para}}\approx\hbar/kT_N\sim 10^{-12}\ \text{s},$$

where k is the Boltzmann constant. The observed high oscillation frequencies $\tau^{-1}\sim 10^{12}\ \text{s}^{-1}$ of the magnetically ordered phase which develops from fluctuations at $T>T_N$ describe a single magnetic fluctuational process in the paramagnetic state of dysprosium. The absence of slower fluctuations, with frequencies which cannot be measured experimentally, follows from the constancy of the asymmetry coefficient a_0 over the entire paramagnetic region of dysprosium, $T_N<300$ K (Fig. 1).

The values of the correlation function $G_{\parallel}$ at $T<230$ K shown in Fig. 6, smaller than the paramagnetic limit, can be explained in a natural way as consequences of a fluctuational decrease in the mean square field $\langle H_z^2\rangle$, which should vanish at octahedral and tetrahedral interstitial positions of a completely ordered helacoidal state.

4. DIFFUSION OF A MUON IN DYSPROSIUM

Figure 7 shows the temperature dependence of the relaxation rate of the muon spin, $\Lambda(T)$, at $T=100$–175 K. The relaxation of the muon spin in this temperature interval is explained on the basis of diffusion of a muon through the dysprosium crystal. As mentioned earlier, at $T<T_N$, be-

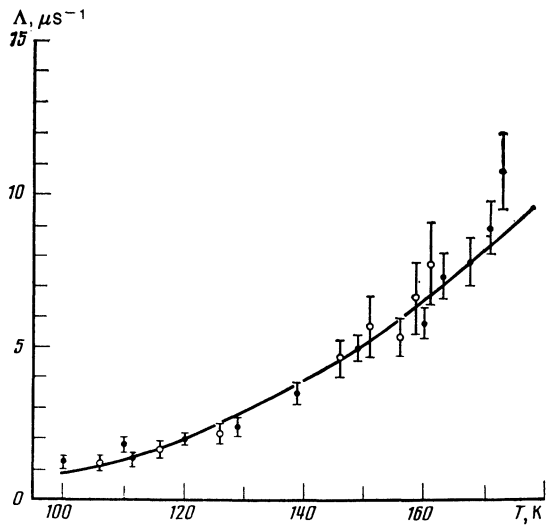

FIG. 7. Experimental temperature dependence $\Lambda(T)$ of the relaxation rate of the spin of a diffusing muon in the antiferromagnetic state of dysprosium, along with theoretical function (26). ○—$\alpha=0$; ●—$\alpha=\pi/2$.

cause of the high local field H_μ at the octahedral and tetrahedral interstitial positions in dysprosium, only the longitudinal component (along the direction of the field H_μ) of the muon spin, σ_{long}, can be observed experimentally. When a diffusion muon hops between two neighboring interstitial positions, the longitudinal component of its spin decreases, becoming equal to $\sigma_{\text{long}} \cos\varphi$, where φ is the angle between the directions of the field H in these interstitial positions. We show below that the diffusion of a muon along interstitial positions in the antiferromagnetically ordered state of dysprosium gives rise to an exponential dependence $P(t) = e^{-\Lambda t}$ of the observed polarization with

$$\Lambda = (\varkappa/\tau) \sin^2\varphi, \tag{21}$$

where τ is the scale time for a diffusive hop by the muon, and $\varkappa \lesssim 1$ is the relative probability for a diffusive hop by the muon to an interstitial position in the neighboring atomic layer, with atomic spins and thus local magnetic fields H_μ rotated through an angle φ. The upper limit $T = 175$ K of the temperature interval in which this diffusion process occurs was chosen so that the nonzero width of the phase transition in the sample would clearly have no effect on the values found experimentally for Λ.

To derive expression (21) we use the notation

$$P(t) = \sum_i P_i(t) \cos\theta_i, \tag{22}$$

where $P_i(t)$ is the contribution to the measured polarization $P(t)$ from muons which at the time t are in an interstitial position in which the local magnetic field H_μ makes an angle θ_i with the direction of the original polarization of the muons. The summation is over all interstitial positions. As a muon diffuses through the crystal, the rate of change of $P_i(t)$ is described by

$$dP_i/dt = W \cos\varphi \{[P_{i-1}(t) + P_{i+1}(t)]/2\} - WP_i(t), \tag{23}$$

where $W = \varkappa/\tau$. The first term in (23) describes an increase in $P_i(t)$ due to the diffusion of muons to interstitial position i from the neighboring interstitial positions $i-1$ and $i+1$; the second term describes the decrease in $P_i(t)$ due to the diffusion of muons away from interstitial position i. From relations (22) and (23) we find

$$dP/dt = -WP(t) + {}^1/_2 W \cos\varphi \sum_i [P_{i-1}(t)\cos\theta_i + P_{i+1}(t)\cos\theta_i].$$

Alternatively, changing the summation indices, we can write

$$dP/dt = -WP(t) + {}^1/_2 W \cos\varphi \sum_i [P_i(t)(\cos\theta_{i+1} + \cos\theta_{i-1})]. \tag{24}$$

Since the local fields H_μ in the dysprosium single crystal are coplanar (perpendicular to the hexagonal axis of the crystal) and are rotated through an angle φ with respect to each other, we can easily see that the following relation holds:

$$\cos\theta_{i+1} + \cos\theta_{i-1} = 2\cos\theta_i \cos\varphi.$$

Expression (24) can then be written as

$$dP/dt = -WP(t) \sin^2\varphi,$$

from which we conclude that $P(t)$ has an exponential dependence in expression (21) for the relaxation rate Λ.

For the case of incoherent diffusion of a muon through a crystal, the temperature dependence $\tau(T)$ can be written[9]

$$1/\tau = \nu e^{-Q/T}. \tag{25}$$

From expressions (21) and (25) we then find

$$\Lambda_{\text{theo}} = \varkappa\nu e^{-Q/T} \sin^2\varphi. \tag{26}$$

The parameters $\varkappa\nu$ and Q are determined by the least-squares method in a comparison of the theoretical function $\Lambda_{\text{theo}}(T)$ (26) with the experimental values of Λ shown in Fig. 7. It should be kept in mind here that the angle φ depends strongly on the temperature in dysprosium. Measurements by means of neutron diffraction show that the angle φ increases almost linearly from $\varphi = 26.5°$ at $T = 90$ K to $\varphi = 43.2°$ at $T = 180$ K (Ref. 10). The theoretical function $\Lambda_{\text{theo}}(T)$ found for this function $\varphi(T)$ is shown by the solid line in Fig. 7. The values of the parameters $\varkappa\nu$ and Q turn out to be

$$\varkappa\nu = 10^{8.28 \pm 0.07}\ \text{s}^{-1},\quad Q = (393 \pm 20)\ \text{K}. \tag{27}$$

The values in (27) for the parameters $\varkappa\nu \approx \nu$ and Q are typical for the diffusion of muons in metals and correspond to a tunneling diffusion.[9]

The method described above for measuring the function $\tau(T)$ in dysprosium in principle makes it possible to determine whether the diffusion of a muon in this metal occurs by a sequence of hops to adjacent interstitial positions or is an unobservably rapid motion (e.g., a band motion) through the crystal between relatively remote local capture centers formed by (usually) impurities. In the latter case the relaxation rate of the muon spin should be independent of the function $\varphi(T)$, which determines the structure of the magnetic ordering of the crystal. Unfortunately, at the level of the statistical error of the given experiment we can make only the qualitative assertion that an interstitial diffusion of a muon in the dysprosium is more probable. As measures of how well the theoretical functions $\Lambda(T)$ for these two diffusion processes agree with the experimental results we find $\chi^2 = 32$ for the function $\varphi(T)$ given above (Fig. 7) and $\chi^2 = 37$ for the case $\varphi = \text{const}$, with a mean statistical value $\chi^2 = 18$.

It follows from relation (21) that the only quantity which must be measured in order to determine the characteristic time τ for a diffusive hop of a muon in dysprosium is the relaxation rate Λ. The fact that the time τ does not depend on the local magnetic field H_μ at the muon should be regarded as an important advantage of the method described above for measuring the diffusion of a muon in dysprosium. This is of course a general method, which can also be used to measure the correlation time τ in other, similar magnetically ordered structures.

Diffusion is not the only process which causes the muon spin to relax at $T < T_N$. The alternating magnetic fields which result from oscillations of the atomic spins in a mag-

netic material, which are not completely ordered at $T > 0$, also produce relaxation of the muon spin. The relaxation rate of the muon spin corresponding to oscillations of the atomic spins, Λ_{osc}, is not large and is less than the paramagnetic value $\Lambda_{osc} \lesssim \Lambda(T = 300\ \text{K}) = 2\,\mu\text{s}^{-1}$. This result has been tested experimentally for erbium,[6] in which there is no diffusive relaxation of the muon spin at $T < T_N$.

5. CONCLUSION

This study of a block-oriented dysprosium sample by a muon method has yielded the following results.

1. Over the broad temperature range $T_N < T < 230$ K of the paramagnetic state of the metal, we observe the formation of a magnetically ordered phase with an antiferromagnetic structure. The lifetime of the observed fluctuations is $\sim 10^{-12}$ s. There are definitely no slower fluctuations.

2. Fluctuations of the paramagnetic state of dysprosium are described by the two correlation functions $G_\perp(T)$ and $G_\parallel(T)$. The function $G_\perp(T)$ increases to a limited extent as the temperature is lowered, and it does not have a singularity in the limit $T \to T_N$, where $T_N = 180$ K is the Néel point. The function $G_\parallel(T)$ falls off in the limit $T \to T_N$. This behavior of $G_\perp(T)$ and $G_\parallel(T)$ is typical of fluctuations whose magnetic structure corresponds to the antiferromagnetic state of dysprosium, and it contradicts the interpretation of the phase transition at $T_N = 180$ K as second-order. This conclusion agrees with Ref. 1, where it was shown by a renormalization group method that a second-order phase transition could not occur at $T = T_N$ in dysprosium, holmium, or terbium.

3. The nonzero width $\delta T_N \approx 3$ K observed for the phase transition in this study is attributed to the appearance of stresses in the individual single crystals of the block dysprosium sample which we used. The width is therefore of an instrumental nature.

4. A method has been proposed for measuring the diffusion of muons in the antiferromagnetically ordered state of dysprosium. With a sufficiently low statistical error this method would make it possible to determine whether the diffusion of a muon in this metal is the result of hops between interstitial positions or is the consequence of the capture of a muon by capture centers. The method proposed here does not require determining the local magnetic field at the muon, H_μ. The results show that the diffusion of a muon in dysprosium is a tunneling process at $T = 100$–175 K. The frequency of the diffusive hopping of a muon over this temperature interval is $\tau^{-1} \sim 10^7\ \text{s}^{-1}$. The method proposed here can also be used to study the diffusion of muons in other metals having a similar magnetic structure.

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