[21]P. L. Martin, O. Parodi, and P. S. Pershan, Phys. Rev. **A6**, 2401 (1972).
[22]L. R. Walker and R. E. Walstedt, Phys. Rev. Lett. **38**, 514 (1977).
[23]V. P. Mineyev and G. E. Volovik, Phys. Rev. **B18**, 3197 (1978).

Translated by J. G. Adashko

Saturation of nuclear magnetic resonance under conditions of large dynamic frequency shift

V. A. Tulin

Institute of Solid State Physics, Academy of Sciences of the USSR
(Submitted 8 June 1979)
Zh. Eksp. Teor. Fiz. **78**, 149–156 (January 1980)

In magnetically ordered crystals with strong hyperfine interaction there is a large observed frequency shift of the nuclear magnetic resonance (NMR), proportional to the magnetization of the nuclei. Under these conditions the NMR has strongly nonlinear properties. The process of locking of the nuclear magnetic resonance withn the detuning is varied in the easy-plane antiferromagnetics $MnCO_3$ and $CsMnF_3$ is investigated. It is shown that in $CsMnF_3$ the results can be described in terms of excitation of NMR on the wing of a line of the Lorentz form. For $MnCO_3$, besides this mechanism, the possibility of locking via excitation of nuclear spin waves is also assumed.

PACS numbers: 75.60. – k, 75.50.Ee

INTRODUCTION

In magnetically ordered substances with strong hyperfine interaction and large density of magnetic nuclei, coupled modes of electron-nucleus magnetic resonance appear. The nuclearlike branch of these modes, like that which has been investigated[1-5] for antiferromagnetics with small crystalline anisotropy, has the form

$$\omega=\omega_n\left[1-\frac{B\langle m\rangle}{\Omega_e^2+B\langle m\rangle}\right]^{1/2}, \tag{1}$$

where ω_n is the frequency of nuclear resonance in the hyperfine field of the electron when there is no coupling, Ω_e is the frequency of antiferromagnetic resonance in the absence of the nuclear magnetic system, and $B\langle m\rangle$ is a quantity which characterizes the coupling. Here the dependence of the coupling on the value of the mean nuclear magnetization $\langle m\rangle$ has been distinguished. The formula used for this quantity is

$$B\langle m\rangle=2\gamma_e^2H_EH_N, \tag{2}$$

where γ_e is the electronic magnetomechanical ratio, H_E is the effective exchange field of the antiferromagnetic system, and H_N is the hyperfine field on the electrons which the nuclei produce:

$$H_N=A\langle m\rangle/\gamma_e, \tag{3}$$

(A is the hyperfine interaction constant). From this we have

$$B=2\gamma_eH_EA. \tag{4}$$

The form in which the nuclearlike resonance modes are written shows that the frequency depends on the mean nuclear magnetization, which can get changed easily under the action of a high-frequency field, a phenomenon described as saturation of NMR.[6] This leads to a strong nonlinearity of nuclear magnetic resonance in the case under consideration. This nonlinearity was pointed out in the very first papers on NMR research in substances with a strong dynamic shift.[1]

If we fix the external parameters—the temperature, the external magnetic field that determines the frequency Ω_e, and the frequency of the exciting field in the range from ω, given by Eq. (1) to ω_n—then the nuclear magnetic system can be in two stable states, viz., at small power levels $\langle m\rangle\approx\langle m\rangle_T$, where $\langle m\rangle_T$ is determined by the thermostat temperature, and at a power $\langle m\rangle\approx\langle m\rangle_0$ sufficient to saturate the NMR, where $\langle m\rangle_0$ is the solution of Eq. (1) for a frequency ω equal to the frequency ω_0 of the exciting field. If $|\omega_0-\omega|>\delta\omega$, where $\delta\omega$ is the linewidth of the NMR, there is a decided difficulty in the system's making the transition from the state $\langle m\rangle_T$ to the state $\langle m\rangle_0$. This problem was considered in the paper of de Gennes and others[1] for a Gaussian shape of the NMR line. They found an exponential dependence of the amplitude of the exciting field required to take the system from the state $\langle m\rangle_T$ to the states $\langle m\rangle_0$ on the detuning $\omega_0-\omega$. Experiments made by double resonance on the saturation of NMR in $KMnF_3$ (Ref. 7) and $MnCO_3$ (Ref. 8) showed a decidedly weaker dependence. It was suggested that this dependence could be described on the basis of the defect structure in the crystals.[7,8] This point of view was maintained, in spite of existing deviations of the experimental results from the calculated dependence.

Kurkin[9] calculated the critical power necessary for the transition of the nuclear system into the saturated state $\langle m\rangle_0$ in the case of a Lorentz shape of the NMR line. He also showed that the Lorentz line shape is closer to a realistic description of the situation. As could be expected in this case, the dependence of the critical power on the detuning is of a power-law type:

 0038-5646/80/010073-05$02.40

$$P_{cr} \propto \omega_1^2 = \frac{4}{27} \frac{(\Delta\omega_2 T_2)^3}{\omega_p T_2} \frac{1}{T_1 T_2}, \quad (5)$$

where T_1 is the spin-lattice relaxation time, T_2 is the spin-spin relaxation time, ω_p is the dynamic frequency shift (DFS) in the linear approximation ($\omega_p \ll \omega_n$), $\Delta\omega_2$ is the detuning, and ω_1 is the amplitude of the high-frequency field, including the enhancement coefficient, in frequency units.

The present paper presents an experimental study of the dependence of the critical power at which the transition to the saturated state occurs on the detuning, for easy-plane antiferromagnetics with large dynamic frequency shift.

EXPERIMENTAL SETUP

The objects chosen for investigation were well studied antiferromagnetics: $CsMnF_3$, a hexagonal antiferromagnetic which has the narrowest NMR line of all investigated antiferromagnetics of this class, and $MnCO_3$, a rhombohedral antiferromagnetic with weak ferromagnetism. The Néel temperature for these substances is well above 4.2 K, so that the antiferromagnetic system of crystals can be regarded as unchanged over the range of working temperatures (below 4.2 K).

The absorption of energy from a high-frequency field (600 MHz was measured) during magnetic field strength scanning of the NMR transition, for various power levels of the irradiation. The absorption cell used was a section of coaxial line of variable length, open at one end and with the other end closed with a small coil of diameter 4 mm, containing four turns of copper wire. The specimen was placed in the coil and immersed in liquid helium. The other end of the coax was brought outside the thermostat through a packing. At this end a capacitance ~0.5 pF provided coupling with the generator and the receiver. A flow-through method was used for the measurements. The figure of merit of this circuit was of the order of 200. Variable attenuators were placed between the circuit and the generator and between the circuit and the receiver. Thus the power entering the circuit could be varied while the power reaching the receiver was held constant. In this way it was possible to measure the relative change of the imaginary part of the susceptibility of the specimen and assess the degree of saturation of the NMR, eliminating possible nonlinearities of the receiver and the subsequent measuring apparatus.

RESULTS OF THE EXPERIMENT

Figures 1 and 2 shows the variation of the high-frequency field energy absorbed in the circuit (proportional to the imaginary susceptibility χ'') as a function of the strength of the static magnetic field. The parameter with which the curves are labelled is the relative amplitude of the high-frequency field h. For small power there is ordinary resonance absorption during the NMR transition. The absorption curve is an isolated peak at the magnetic field H_p that satisfies the relation (1) for frequency ω equal to that of the exciting high-frequency field and nuclear magnetization $\langle m \rangle_T$

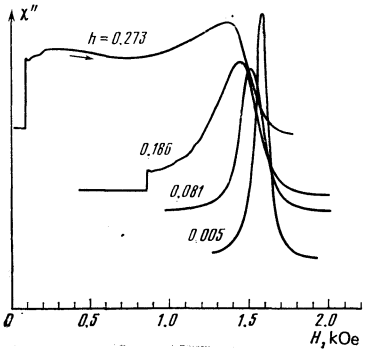

FIG. 1. Absorption curves in a specimen of $MnCO_3$ at various irradiation powers. The scale of χ'' is arbitrary, but the same for all curves. h is the relative amplitude of the high-frequency magnetic field. $f = 597$ MHz, $T = 4.2$ K.

corresponding to the cryostat temperature.

Increase of the amplitude of the high-frequency field leads in the first stage to broadening of the resonance absorption curve. This stage is well marked with $MnCO_3$, which has the broader NMR line. Further increase of the power brings about a stage in which the absorption in the specimen increases discontinuously as the static field is increased from $H = 0$, and reversal of the direction of the field scan leads to hysteresis.

The writer has shown[8] that the discontinuous rise of the absorption corresponds to a change of state of the nuclear system from $\langle m \rangle_T$ to $\langle m \rangle_0$. Fig. 3 shows the dependence of the amplitude of the high-frequency field necessary to bring the system into the state $\langle m \rangle_0$ on the magnitude of the static magnetic field at which the transition is observed. It is seen that there is a decided difference in the dependences found for $MnCO_3$ and $CsMnF_3$.

Assuming that the mechanism of the transition described in Ref. 9 occurs in $CsMnF_3$, and in the initial part (up to the kink) in $MnCO_3$, we can plot the dependence of the detuning at which the jump in the susceptibility occurs on the amplitude on the high-frequency field.

The amplitude (relative units, as before) is calculated from the readings of the attenuator, and the detuning is

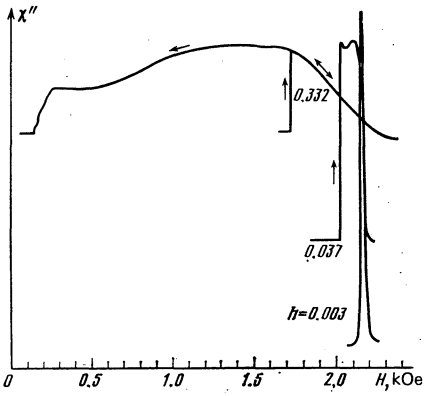

FIG. 2. Absorption curves of NMR in a specimen of $CsMnF_3$. Irradiation frequency 600 MHz, temperature $T = 1.27$ K. Arrows indicate direction of magnetic-field scan.

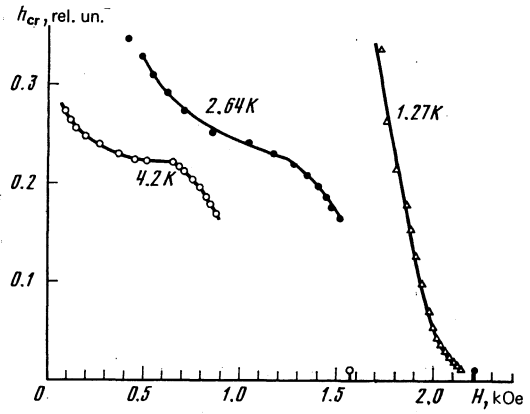

FIG. 3. Dependence of the critical high-frequency magnetic field amplitude h_{cr} at which transition of the system to the saturated state (locking) occurs on the constant magnetic field: ○, ● for $MnCO_3$, △ for $CsMnF_3$.

determined as the difference between the frequency of the electromagnetic wave and that of the nuclear resonance, found from Eq. (1) for the value of the magnetic field at which the jump in the susceptibility occurs and for the temperature of the cryostat. Since in $CsMnF_3$, the width of the NMR line is comparatively small, the jump in the susceptibility occurs for it close to the position of the NMR line. In this substance one can trace the dependence of the critical amplitude h_{cr} on the detuning, beginning at small values of the detuning. Figure 4 shows in logarithmic coordinates the dependence of Δf on h_{cr} for $CsMnF_3$, and also shows initial segments for $MnCO_3$.

The case of $MnCO_3$ is interesting because there is a change of the law connecting Δf and h_{cr}. The kink in the curve can be seen clearly in Fig. 3. The presence of this kink suggests the existence of competition between two mechanisms of the transition of the nuclear system to the saturated state.

LOCKING OF THE NMR FREQUENCY AT LARGE IRRADIATION POWER

The action of a high-power high-frequency field on a nuclear system possessing a dynamical frequency shift leads to a change of the magnitude and spatial orientation of the vector $\langle \mathbf{m} \rangle$, which leads to a change of the NMR frequency, as can be seen from Eq. (1). There can only be a decrease of the saturation magnetization, and the frequency increases correspondingly.

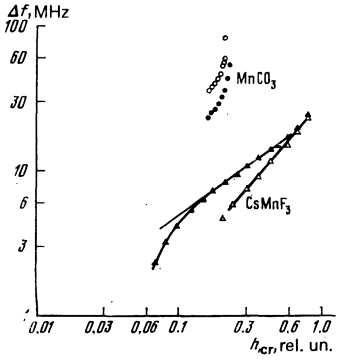

FIG. 4. Dependence of maximal frequency detuning of the high-frequency field from the NMR frequency at which the transition of the system to the saturated state occurs, on the amplitude of the high-frequency field.

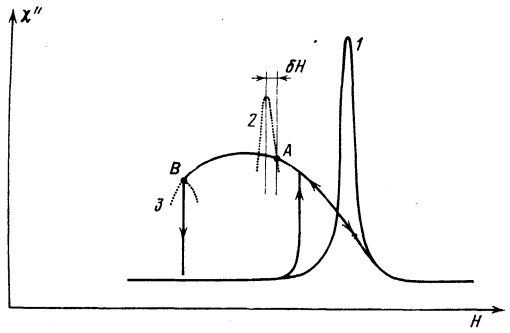

FIG. 5. Illustration of origin of absorption curves (Figs. 1, 2). Curve 1 corresponds to the unlocked state of the system; point A is an unstable locked state, and B is a stable locked state.

Fairly many analogs of this problem can be found, and therefore the study of this phenomenon is of more general interest. In any resonant system whose resonance frequency depends on some parameter which is changing as the result of the action of an exciting force, there will be a similar behavior of the system's response. In the present case of a nuclear magnetic system with DFS the dependence of the resonance frequency on the amplitude of the high-frequency field through change of the magnetization is very strong. The change of the resonance frequency through such action is much larger than the line width.

Figure 5 shows the dependence of χ'', the imaginary part of the susceptibility in NMR, on the magnetic field, illustrating the origin of the experimental curves of Figs. 1 and 2. In our situation there is a constant pumping frequency, and the position of the resonance is characterized by the magnetic field. Curve 1 corresponds to the behavior of χ'' in NMR at low power. If the power is sufficiently large, so that saturation of the NMR is possible, then, according to Eq. (1), the resonance field must decrease. If we move along the resonance curve toward decreasing field, the presence of the saturation effect will result in the resonance curve's being at a distance δH to the left of the value of H at which we are. This is shown by cirve 2 in Fig. 5. The position of the system at the point A is stable, since random decreases of the power will lead to a decrease of the quantity δH, to an increase of the absorption with subsequent restoration of the value of δH. The stability disappears when the point at which the system falls on the curve of $\chi''(H)$ is at the maximum of the absorption curve 3, at point B. In this case any random decrease of the power leads to an increase of the resonance field, and this causes a decrease of the absorption and a further increase of the resonance field, and the system moves away from the partially saturated state into the state $\langle m \rangle_T$ of thermal equilibrium with the lattice.

In the opposite motion of the field, i.e., as it increases from zero, as long as we are far from the resonance (curve 1) there can be no significant absorption associated with NMR, and therefore the nuclear system is in

equilibrium with the lattice. When we get near the resonance curve, there is some absorption on its wing, the nuclear magnetic system begins precessing, and the temperature rises above that of the lattice. Both of these changes lead to a decrease of the resonance field and to an increase of the absorption; that is, at a certain amount of detuning from the frequency of NMR (or at a certain ΔH) the system goes over into the saturated state $\langle m\rangle_0$. This transition has also been calculated in papers by de Gennes *et al.*[1] and by Kurkin,[9] in the former case for a Gaussian line shape and in the latter, for a Lorentz shape.

Experiment shows that in the case of $CsMnF_3$, the critical power for the transition is related to the detuning according to a power law. At temperature 1.27 K this relation is close to the form

$$P_{cr}^{1/2} \propto h_{cr} \propto (\Delta\omega_2)^{3/2} = (2\pi\Delta f)^{3/2};$$

which agrees with the results of Kurkin's calculation on the assumption that

$$T_2/\eta\omega_0 T_1 = \text{const}$$

(η is the amplification coefficient of the NMR).

In the case of $MnCO_3$, for detunings smaller than 30 MHz the situation is evidently similar to that observed in $CsMnF_3$. For larger detunings the plots of $\Delta f(h)$ and $h(H)$ (Fig. 3) show an appreciable change in the nature of the behavior (a weakening of the degree of dependence). This is particularly marked at the temperature 4.2 K. A similar form of the dependence is observed in $KMnF_3$.[7] The mechanism of transition of a nuclear magnetic system to the saturated state that has already been mentioned, associated with crystal defects, describes these results on the average, but without reflecting the change in the character of the dependence $\Delta f(h_{cr})$. It may be supposed that these changes reflect a change of the pumping mechanism of the NMR. Therefore we shall examine another possibility for the transition of a nuclear magnetic system to the saturated state.

It was shown in a paper by Govorkov and the writer[10] that a nuclear magnetic system can be taken over into the saturated state by parametric excitation of nuclear spin waves with parallel pumping. There is also a possibility of excitation of nuclear spin waves with the geometry used in the present study (perpendicular pumping). The insert in Fig. 6 shows a process in which two quanta of the electromagnetic field give rise to two nuclear magnons with opposite wave vectors. Several such processes can be devised (this is symbolically indicated by the sign Σ in Fig. 6). As in parallel pumping, at some elevation above threshold a change of the state of the nuclear system begins (a decrease of $\langle m\rangle$, a decrease of the wave vector of the excited nuclear spin wave, and an increase of the frequency of NMR).

Figure 6 shows the dependence of the frequency of NMR on the magnetic field. This plot can be regarded as a phase diagram of the existence of spin waves. Nuclear spin waves exist between the solid curve, the NMR frequency as a function of H, and the dashed straight line $\omega_n = \text{const}$, the unshifted NMR frequency. Now if we are given an external magnetic field H_0 and

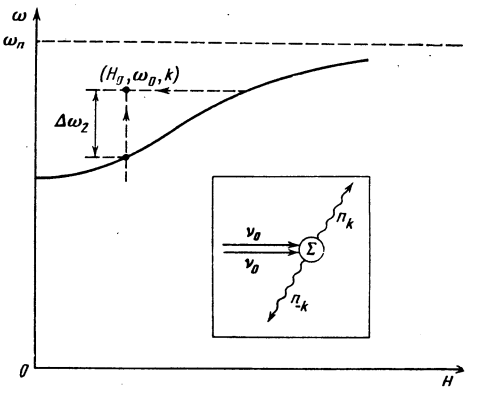

FIG. 6. Qualitative picture of the NMR spectrum in a system with dynamic frequency shift. Nuclear spin waves are localized between the solid curve $\omega(H)$ [Eq. (1)] and the dashed line $\omega_n = \text{const}$. Inset shows process of excitation of two nuclear magnons by two quanta of the high-frequency field ($2\pi\nu_0 = \omega_0$).

a frequency $\omega < \omega_0 < \omega_n$, besides the old statement that the problem contains a detuning $\Delta\omega_2 = \omega_0 - \omega$ from the NMR frequency, we can say that in the given field H_0 there are nuclear spin waves with the proper frequency ω_0 and wave number k. These nuclear spin waves can be excited by the external field, and through their excitation the system can be taken into the saturated state. Then the curve $h_{cr}(H)$ can be regarded as a sort of "butterfly" in the excitation of spin waves. These mechanisms can compete under various conditions, and evidently this is indeed manifested in the case of $MnCO_3$.

The excitation of nuclear spin waves can occur in the following way. When an alternating magnetic field of the fundamental frequency acts on an antiferromagnetic system in a NMR geometry a motion of the electronic magnetization is set up. If the alternating and constant fields lie in the plane of easy magnetization, which was the case in the present experiments, the motion of the electronic magnetization will be strongly elliptical. This can be observed easily in studying the high-frequency susceptibility. The susceptibility perpendicular to the magnetic field in the basal plane is much larger than that along the axis perpendicular to this plane, in antiferromagnetics with weak ferromagnetism ($MnCO_3$, $CoCO_3$). Owing to this practically two-dimensional motion of the electronic magnetization, characterized by a vector of constant length, oscillations of the component of the magnetization parallel to the field arise at the doubled frequency. When these oscillations reach a certain amplitude, parametric pumping of nuclear spin waves begins. The dependence of the susceptibility of the electronic system on the position of the operational point (ω_0, H) relative to the NMR spectrum branch somewhat alters both the expression for the critical amplitude and the character of the process of excitation of nuclear spin waves.

In conclusion I would like to thank M. I. Kurkin for a discussion which stimulated me to do this research, S. A. Govorkov for assistance in performing the experiments, and V. S. Lutovinov for a discussion of the possibility of transition to the saturated state via nuclear spin waves.

[1]P. G. de Gennes, P. A. Pincus, F. Hartmann-Boutron, and J. M. Winter, Phys. Rev. 129, 1105 (1963).
[2]E. A. Turov and V. G. Kuleev, Zh. Eksp. Teor. Fiz. 49, 248 (1965) [Sov. Phys. JETP 22, 176 (1966)].
[3]A. J. Heeger, A. M. Protis, Dale T. Teaney, and G. Witt, Phys. Rev. Lett. 7, 307 (1961).
[4]A. Nakamura, V. Minkiewicz, and A. M. Portis, J. Appl. Phys. 35, 842 (1964).
[5]A. S. Borovik-Romanov and V. A. Tulin, Pis'ma Zh. Eksp. Teor. Fiz. 1, No. 5, 18 (1965) [JETP Letters 1, 134 (1965)].
[6]A. Abragam, Principles of Nuclear Magnetism, Oxford, Clarendon Press, 1963.
[7]G. Witt and A. M. Portis, Phys. Rev. 135, A1616 (1964).
[8]V. A. Tulin, Zh. Eksp. Teor. Fiz. 55, 831 (1969) [Sov. Phys. JETP 28, 431 (1969)].
[9]M. I. Kurkin, Pis'ma Zh. Eksp. Teor. Fiz. 28, 675 (1978) [JETP Lett. 28, 628 (1978)].
[10]S. A. Govorkov and V. A. Tulin, Pis'ma Zh. Eksp. Teor. Fiz. 27, 345 (1978) [JETP Lett. 27, 324 (1978)].

Translated by W. H. Furry

Magnetization of a single crystal of erbium in the basal plane in strong magnetic fields

V. L. Yakovenko, V. E. Bril', V. V. Druzhinin, R. Z. Levitin, V. M. Mel'nikov, and R. E. Osipova

Engineering-Physics Institute, Moscow
(Submitted 19 June 1979)
Zh. Eksp. Teor. Fiz. 78, 157–164 (January 1980)

The magnetization of erbium has been measured in the basal plane, along the **b** axis, in fields ~300 Oe and over the temperature range 4.2 to 64 K. In the range 4.2 to 35 K, a magnetization discontinuity $\approx 0.82\ \mu_B$ per atom is detected at a field ≈270 kOe; thereafter, saturation sets in. In the temperature range in which a discontinuity occurs, the temperature dependence of the critical fields is obtained from measurements of the differential susceptibility. It is shown that the low-temperature anomalies of the magnetization of erbium in the basal plane and along the hexagonal axis **c** can be explained in the anisotropic molecular field (AMF) approximation.

PACS numbers: 75.30.Cr

INTRODUCTION

A considerable amount of work has been devoted to investigation of the anisotropy of heavy rare-earth metals (HREM). But there is not yet any diminution of interest in this problem, since there are a number of phenomena in HREM that still await explanation. One of the important problems is overcoming the anisotropy field, which in HREM reaches values of order of magnitude $H_a > 500$ kOe.[1)] The obtaining of such fields entails considerable difficulty. Still more complicated is the problem involved in measurements of the magnetic characteristics in such magnetic fields.

In certain HREM the anisotropy field may be smaller. As has been shown by investigations of the magnetization of Er_xGd_{1-x} alloys,[2] the anisotropy fields in Er, obtained by extrapolation of the magnetization curves into the high-field region, have a value $H_a < 400$ kOe. In the same paper, for the alloy with 20% erbium content, it was shown that saturation along the hard direction is attained at a field $H \approx 50$ kOe, and that there is an anomaly on the magnetization curve: saturation is attained jumpwise. Consequently it might be possible to try to obtain saturation of pure erbium by assuming a similar behavior of the magnetization curve in fields appreciably lower than 400 kOe. Saturation by discontinuity along the hard direction in erbium also follows from a calculation in the anisotropic molecular field (AMF) model, given below, with $H_a < 300$ kOe.

The present paper gives the results of measurements of the magnetization and of the differential susceptibility of a single crystal of erbium in the basal plane, along the **b** axis, in strong fields, reaching 340 kOe, over the temperature range 4.2 to 64 K. A jump of the magnetization to saturation was observed at a field ~300 kOe over the temperature range 4.2 to 35 K. On the basis of the AMF approximation, a quantitative explanation is given of the characteristics of the magnetization in the basal plane, and also along the **c** axis; the latter were measured earlier[5] by some of the authors of this article.

SPECIMENS; EXPERIMENTAL TECHNIQUE

The single crystals of erbium were grown by the method of crucibleless zone melting with induction heating, in an atmosphere of gaseous helium, at the A. A. Baĭkov Institute of Metallurgy. The composition and homogeneity of the specimens were determined by the method of atomic-absorption analysis. The specimens for magnetization measurements were cut from bulk single crystals in the form of bars of dimensions $10 \times 1.4 \times 2$ mm and $5 \times 1 \times 1$ mm by the electric-spark method and were oriented on a diffractometer by the method of inverse Laue mapping, along various crystallographic directions. The accuracy of the orientation in the experimental apparatus was 2 to 3°. The magnetization measurements were made by the induction method[6] in pulsed magnetic fields up to 340 kOe, with pulse

 0038-5646/80/010077-05$02.40