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

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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.

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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

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duration 0.01 sec, over the temperature range 4.2 to 64 K. The error in the measurement of the magnetization in the pulsed field was 7 to 10%; 3 to 5% of this is attributable to systematic error.

The absolute value of the magnetization of a single crystal of erbium in the basal plane, along the **b** axis, was measured over the interval 4.2 to 64 K on a vibration magnetometer, with a superconducting solenoid, in a field up to 65 kOe. The error in the measurement of the magnetization did not exceed 3%. The calibration of the field of the pulse solenoid was done with a test coil and according to reference points at the known and constant fields of the transitions in erbium at 4.2 and 78 K. The error in the measurement of the field, and also of the critical fields in the phase-transition region, did not exceed 3%. After each measurement cycle in fields $H > 300$ kOe, the crystal structure of the specimen was checked.

EXPERIMENTAL RESULTS

Figure 1 shows the experimental variations of the magnetization, along the **b** axis, of a hexagonal erbium single crystal, with change of the applied magnetic field, at various temperatures. On the magnetization curve at 4.2 K there is observed at field $H = 17$ kOe, a jump of the magnetization caused by "collapse" of the helicoidal magnetic structure formed by the projections of the magnetic moments on the basal plane. The final destruction of the helicoidal structure occurs at $H = 21$ kOe; there the magnetization is 4.2 μ_B, which corresponds to an angle of departure of the magnetic moment from the axis $\theta_0 = 27.4°$.[2)] In the region of the jump, there is observed in weak magnetic fields a hysteresis in the direct and reverse traversals of the magnetization curve. On further increase of the field, up to $H = 270$ kOe, a slow increase of the magnetization in the basal plane is observed. In the field interval 100 to 272 kOe, the change of magnetization is insignificant. At $H = 272$ kOe, an anomaly is observed in the behavior of the magnetization curve: the magnetization increases by the amount $\Delta M = 0.82$ μ_B, and the curve goes to saturation with a value $M = 9.12$ μ_B per atom. This value is somewhat lower than the magnetization value obtained from measurements along the **c** axis (the easy axis) in fields up to 150 kOe,[5] and also somewhat larger than the theoretical value calculated from the formula $M = gJ\mu_B$ on the assumption of complete parallel orientation of the spins of the $4f$ electrons of the trivalent erbium ion. It is also somewhat higher than the value obtained from neutron-diffraction measurements.[10]

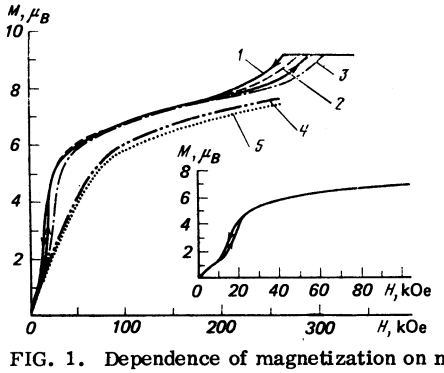

FIG. 1. Dependence of magnetization on magnetic field along the **b** axis (Curves 1, 2, 3, 4, 5 correspond to $T = 4.2$, 22, 33, 43, 64 K).

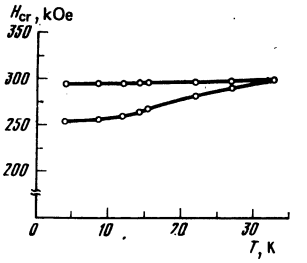

FIG. 2. Temperature dependence of critical field. Upper curve, on increase of field; lower, on decrease.

With increase of temperature, the magnetization jumps become smoother, and their amount diminishes. As in the case of magnetization along the hexagonal axis, the magnetization jumps occur with hysteresis, which diminishes with rise of temperature. At temperature 35 K, the jump disappears, and at our accuracy of measurement no anomaly of the magnetization is observed.[3)]

In the region of magnetization jumps, over the whole temperature interval investigated, curves of the differential susceptibility were taken; this makes it possible to increase the accuracy in the measurement of the critical fields H_{cr}. Figure 2 shows the temperature variations of the critical fields, obtained from measurements of the differential susceptibility, on increase and on decrease of the field. Within the temperature interval 4.2 K $\leq T \leq$ 18 K, the value of H_{cr} is practically independent of temperature; within the temperature interval 20 K $< T <$ 35 K, the critical field increases, and the hysteresis diminishes.

In contrast to the magnetization curves taken for H || **c**,[5] the differential susceptibility for magnetization along the **b** axis does not have as sharp a maximum in the region of the anomaly. This difference is due to the smoother behavior of the magnetization curve in the region of the jump.

Figure 3 shows the temperature dependence of the derivative of the magnetization at $H = H_{cr}$, plotted from the inverse behavior of the differential-susceptibility curve. It is seen that the $\partial M/\partial H$ dependence changes little with temperature up to $T = 18$ K and departs from linearity in the interval 20 K $< T <$ 35 K.

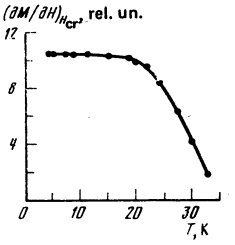

FIG. 3. Temperature dependence of the derivative of the magnetization with respect to the field.

Our experimental data on magnetization of Er along the **b** axis for $H \leq 60$ kOe agree well with the data of Ref. 7, obtained from measurements of the magnetization in constant fields of the same magnitude.

DISCUSSION OF RESULTS

Erbium may be assigned to the class of strongly anisotropic ferromagnets. The effect of the anisotropy changes the Curie temperature from the usual value $T_c^0=(g-1)^2 I_z J(J+1)/3k$ to $T_c = 1.33\ T_c^0$; that is, the anisotropy energy amounts to approximately 30% of the exchange energy.[4] We shall carry out an analysis of the dependence of the orientation of the magnetic moment on the value of the external field by two methods: a phenomenological, and a microscopic, with use of the AMF approximation. In the first case, the free energy is written in the form

$$F=K_2^0 P_2^0(\cos\theta)+K_4^0 P_4^0(\cos\theta)+K_6^0 P_6^0(\cos\theta)-MH\cos\theta', \qquad (1)$$

where K_l^0 are macroscopic anisotropy constants, $P_l^0(\cos\theta)$ is a Legendre polynomial, M is the magnetization, and θ and θ' are the angles between **M** and **c** and between **M** and **H**, respectively. The parameters K_2^0, K_4^0, and K_6^0 can be determined from three experimental quantities: the critical field H_{cr}, the amplitude of the magnetization jump, and the orientation of the magnetic moment in zero field. At 4.2 K, this gives $K_2^0 = -5.66\cdot10^{-15}$, $K_4^0 = 2.095\cdot10^{-15}$, $K_6^0 = 0.783\cdot10^{-15}$ erg/atom. We note that these values are close to data given by other authors[11] (see Table I).

Further, supposing that $M = gJ\mu_B$ (the maximum value), we carried out a calculation of the angle θ corresponding to the minimum of F as the field, oriented along the **b** axis, varied up to 340 kOe. The theoretical curve calculated by formula (1) is shown in Fig. 4. At $H=0$ it goes through the value $M=4.2\,\mu_B$ corresponding to complete destruction of the helicoidal structure. As is seen from Fig. 4, the calculated curve qualitatively reproduce the experimental. The difference in the absolute value of the amount of magnetization in weak fields is due to the fact that in the theoretical calculation, the effect of the helicoidal structure on the magnetization was disregarded. But in the region of the jump, both curves coincide, and the amplitude of the jump is $0.82\ \mu_B$. A calculation with the constants K_l^0 from Ref. 11 gave a significant difference from experiment. We note here that the K_2^0 assumed by us coincided exactly with the K_2^0 from Ref. 2 (29.3 and 29 cm^{-1}, respectively).

The phenomenological approach developed above, despite its simplicity, has definite limitations. The expansion (1) is valid if the anisotropy energy is much smaller than the exchange-interaction energy, a condition

TABLE I. Anisotropy constants in erbium.

$K_2^0\cdot10^{15}$, erg/atom	$K_4^0\cdot10^{15}$, erg/atom	$K_6^0\cdot10^{15}$, erg/atom	Note
−5.660	2.095	0.783	Our data, Eq. (1)
−4.985	0.598	0.596	Ref. 11
−3.449	−0.500	1.000	Ref. 11
−6.39	1.45	1.6	Our data, AMF

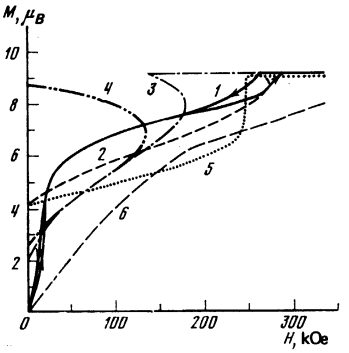

FIG. 4. Experimental (1) and theoretical (2–6) variations of magnetization with magnetic field along the hard direction (the axis **b**). Curve 2 was calculated by formula (1) with our data; 3 and 4 by (1) with the parameters K_l^0 from Ref. 11; 5 and 6 in the AMF approximation at 4.2 and 64 K, respectively.

that is not satisfied with sufficient accuracy for Er. Furthermore, this approach does not explain the magnetization jumps along the **c** axis found in the earlier paper[5] and does not give the spectrum of the system. It is also necessary to take into account that the expansion in Legendre polynomials P_l^0 may contain terms with $l>6$. We shall therefore carry out an analysis of our results on the basis of the AMF approximation developed in Refs. 3 and 4. The statement of the problem consists of the following.

Starting from the Hamiltonian

$$\hat{H}=-\sum_{i>j} I_{ij}(\hat{\mathbf{S}}_i\hat{\mathbf{S}}_j)+\sum_{ikl} A_{kl}{}^i \sum_{j=1}^{n} C_{kl}(r_j)+\sum_i g\mu_B \mathbf{HJ},$$

where we have taken into account the exchange, anisotropy, and Zeeman energies, we transform to a single-ion operator in the first term and, using equivalent operators, write $\hat{H}$ in the form

$$\hat{H}=-I_z J(g-1)^2\sigma(J_z\cos\theta+J_x\sin\theta)+{}^1/_2\alpha A_2^0\hat{O}_2^0 +{}^1/_8\beta A_4^0\hat{O}_4^0+{}^1/_{16}\gamma A_6^0\hat{O}_6^0+g\mu_B\mathbf{HJ}. \qquad (2)$$

In (2), $I_z=\Sigma_i I_{ij}$, $\sigma=J/J$ is the reduced magnetization, α, β, and γ are Stevens parameters, and $\hat{O}_l^0$ are operators tabulated, for example, in Ref. 12. The set of parameters was determined in Ref. 3 for all HREM: $I_z=43.05\ cm^{-1}$, $A_2^0=-240\ cm^{-1}$, $A_4^0=94\ cm^{-1}$, $A_6^0=133\ cm^{-1}$. A characteristic feature of the anisotropy field for these parameters in Er is the inverse arrangement of the components of the multiplet $J=15/2$: the ground state is the doublet with $m=\pm13/2$, whereas the doublet with $m=\pm15/2$ is an excited state. The molecular field parallel to the z axis, $H_{||}{}^M\sim I_z J\sigma\cos\theta$, tends to bring the components with $m=-13/2$ and with $m=-15/2$ closer together, while $H_\perp{}^M\sim I_z J\sigma\sin\theta$, on the contrary, separates the levels and "mixes" the states. In Ref. 5, a jump of amount $M=gJ\sigma\mu_B\cos\theta$ in field 110-120 kOe, parallel to the **c** axis (the z axis), was interpreted as an intersection of the levels with $m=-13/2$ and $-15/2$ This fact led us to a need to change the parameters A_4^0 and A_6^0 somewhat, to values 80 and 69 cm^{-1} respectively.[4)] Then $\Delta E=E(15/2)-E(13/2)=19.5\ cm^{-1}$ only because of anisotropy. In a magnetic field, with allowance for the fact that $H_\perp{}^M=0$ (the moment is oriented along the field), ΔE decreases

by the amount

$$I_z J(g-1)^2+g\mu_B H, \tag{3}$$

which gives, at field 120 kOe, 19.5 cm^{-1}.

Thus we shall hereafter use this set of microscopic parameters. The relation between A_l^0 and K_l^0 in the presence of weak anisotropy, and with allowance for the fact that the ground level corresponds to $m=j$, has the form[3]

$$K_2^0=\tfrac{1}{2}\alpha\langle^{15}/_2|O_2^0|^{15}/_2\rangle A_2^0, \quad K_4^0=\tfrac{1}{8}\beta\langle^{15}/_2|O_4^0|^{15}/_2\rangle A_4^0, \quad K_6^0=\tfrac{1}{16}\gamma\langle^{15}/_2|O_6^0|^{15}/_2\rangle A_6^0 \tag{4}$$

and gives $K_2^0=-6.39\cdot10^{-15}$, $K_4^0=1.45\cdot10^{-15}$, $K_6^0=1.6\cdot10^{-15}$ erg/atom. It is seen that they coincide in sign and order of magnitude and approximately in value with the parameters K_l^0 used by us in equation (1). Complete coincidence is not to be expected, since ours is the case of strong anisotropy, the ground state is a superposition of projections with $m=-15/2$ and $-13/2$, and higher harmonics were not taken into account in (1). The description in the AMF approximation is in this sense more complete.

Because the Hamiltonian (2) contains the quantities σ and θ, in order to determine them and the spectrum of the system it is necessary to seek the minimum of the energy (2) on functions of the multiplet with $J=15/2$. In contrast to the isotropic case, in which min F leads to a molecular-field equation, in our case it is necessary to seek a minimum with respect to two parameters σ and θ, and this leads to solution of two coupled equations: the molecular-field equation

$$\mathrm{Sp}\{(J_x\sin\theta+J_z\cos\theta-J\sigma)\exp(-\hat{H}/kT)\}=0 \tag{5}$$

and the equilibrium-orientation equation

$$\mathrm{Sp}\left\{\frac{d\hat{H}}{d\theta}\exp\left(-\frac{\hat{H}}{kT}\right)\right\}=0. \tag{6}$$

Joint solution of equations (2), (5), and (6) constitutes the AMF approximation and gives, for Er at $H=0$ and $T=4.2$ K, $\theta=28°$. For $\mathbf{H}||\mathbf{x}$ (b), calculation gives a linear increase of θ to 49° up to field 240 kOe at 4.2 K, after which there occurs a sharp increase of it to 90° within the interval 240-250 kOe. During this, the magnetization changes discontinously from $6.79\mu_B$ to $9\mu_B$. Thus this is a phase transition of the first kind, of the spin-reorientation type.

Figure 4 gives a comparison of the theoretical results with experiment. The quantitative agreement, it is evident, is somewhat worse than in the calculation by formula (1). This may be due to the fact that the calculation was done in an isothermal version, whereas under the conditions of strong pulsed fields the process is adiabatic, and this leads to an increase of temperature. Increase of temperature decreases the amount of the jump, which brings the theoretical results closer to the measured. Calculation for $T=64$ K up to $H\approx300$ kOe gives a linear increase of $M(H)$ without a jump, as is observed in the experiment (Fig. 4).

We shall at the same time consider what the AMF gives by way of explanation of the jumps of magnetization along the **c** axis ($\mathbf{H}||\mathbf{z}$). As has been shown, in this case the levels $-13/2$ and $-15/2$ intersect for $\theta=0$ and $H=120$ kOe. But this version does not correspond to the minimum of F; that is, it is more advantageous when $H_\perp^M\neq0$ and the levels "interact" because of the nondiagonal elements $\langle m|\hat{J}_x|m'\rangle$. Solution of the AMF equations for $T=4.2$ K and $\mathbf{H}||\mathbf{z}$ gives $\theta=22°$ at $H\approx110$-120 kOe. The fact that the magnetic moment deviates form the z axis in the region of intersection of the levels, in the presence of weak exchange, was predicted in Ref. 13 and corresponds to reasoning[14] about the magnetic analog of the Jahn-Teller effect: the system tends to lift the accidental degeneracy by a change of the magnetic symmetry. But in this the jump of magnetization that is observed in the experiment should not occur. To explain it, we must take account of magnetostriction. Then inversion of the levels may result from even an insignificant change of the crystal-field parameters, because of magnetoelastic interaction, near H_{inv}: and with further increase of field, orientation of the moment along the field, i.e. $\theta=0$, will be advantageous.

We shall consider this process qualitatively. Let the two levels $m_1=-J+1$ and $m_2=-J$ have energies $\mathscr{E}_1=0$ and $\mathscr{E}_2=a$ ($a>0$) in the anisotropy field. They are acted upon by an external field h, $\Delta\mathscr{E}_1=-h(J-1)$ and $\Delta\mathscr{E}_2=-hJ$, and by exchange interaction, $H_{ex}=\hat{J}_x\sin\theta+\hat{J}_z\cos\theta$; that is, the matrix of the interaction has the form

$$\begin{pmatrix} a-h-\cos\theta & d\sin\theta \\ d\sin\theta & 0 \end{pmatrix}.$$

From this matrix it is evident that $h_{inv}=a-1$ and that the minimum of the energy ($T=0$) is

$$\min\mathscr{E}_1=\tfrac{1}{2}(a-h-\cos\theta)-\tfrac{1}{2}[(a-h-\cos\theta)^2+4d^2\sin^2\theta]^{1/2}, \tag{7}$$

that is, when $h=h_{inv}$, min $\mathscr{E}_1$ occurs at $\theta=\pi/2$. It is easy to show that if the value of a has decreased to $a-e$ in the h_{inv} region, the minimum of the energy corresponds to $\theta=0$ if $e>d^2$. Physically this follows from the fact that here the level $m=-J$ becomes the lower one, and both components of the molecular field, $H||^M$ and $H_\perp^M$, tend to separate the levels; $H_{||}^M\sim\hat{J}_z\cos\theta$ gives a larger shift than $H_\perp^M\sim\hat{J}_x\sin\theta$ (the second order of perturbation theory), and therfore the angle $\theta=0$ is more advantageous.

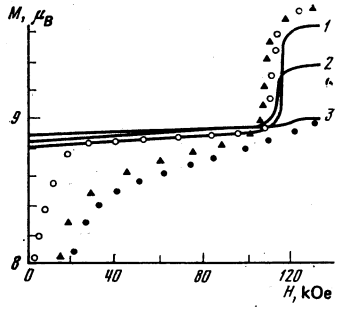

FIG. 5. Variation of magnetization with field along the **c** axis. The solid curves were calculated in the AMF model with allowance for the adiabaticity of the process and for magnetostriction (1, 2, and 3 correspond to $T=4.2$, 20, and 40 K). The experimental[5] points ○, ▲, ● correspond to $T=4.2$, 20, 40 K. The calculation took into account the contribution of the magnetization of the conduction electrons, $M_{el}=0.07M_{ion}$.

We carried out the following numerical experiment. At field $H = 115$ kOe, we "switched" into the matrix of the AMF equations the magnetostriction, which shifted the level $m = -15/2$ (the excited level) downward by a distance equal to $\frac{3}{2}a$ (a is the energy gap, which is minimal in this case and equal to 0.5 cm^{-1}). Thereupon θ changed from 12 to 0°. Further increase of the field corresponded to splitting of the levels with $m_2 = -J$ and $m_1 = -J+1$ (pure wave functions) and $\mathscr{E}_1 > \mathscr{E}_2$. In this case a jump of the magnetization occurs at 120 kOe because of an abrupt change of the wave function of the ground state.

The characteristic temperature dependence of the jump of M—lack of dependence of ΔM on T in the range 4.2 to 28 K and vanishing of ΔM at higher temperatures—is caused by lowering of the temperature by virtue of the magnetocaloric effect. This question was considered in Ref. 15. Figure 5 gives a comparison of calculated and experimental results for magnetization along the **c** axis; their close agreement is evident.[5)]

Thus, as follows from the experimental data, the transition to a collinear structure in erbium in a field **H**||**b** at low temperatures is a first-order phase transition. This has been explained by considering simple magnetization processes and by a microscopic calculation. The explanation of the jumps of magnetization along the **c** axis is based on inversion of the structure of the levels, magnetostriction, and a negative magnetocaloric effect. For the first time, saturation of the magnetization of erbium has been successfully obtained in fields ≈ 273 kOe.

[1)]An earlier experiment[1] on dilute solutions of Dy in Gd enables us to estimate the anisotropy field in Dy: $H_a \approx 600$ kOe.

[2)]It must be noted that rotation processes do not follow directly after destruction of the helicoidal magnetic structure, but begin only at a field $H \sim 35$ kOe; this, as was mentioned in Refs. 7 and 8, is due to the existence of a transitional region (metastable state) within the field interval $21 < H < 35$ kOe. Also, the value of the magnetization, and consequently of the angle θ_0, is somewhat larger than follows from neutron diffraction data ($24° \leqslant \theta_0 \leqslant 29°$).[9]

[3)]We note that the magnetization jumps are observed at temperatures considerably exceeding the temperature of transition to the helicoidal phase (20 K).

[4)]Use of the parameter values from Ref. 3 leads to qualitatively similar results. Only the amounts and fields of the magnetization jumps differ slightly.

[5)]Preliminary measurements that we have made showed that at 4.2 K, at field ~100 kOe, there occurs a discontinuity of the longitudinal magnetostriction $\Delta l/l \approx 2 \cdot 10^{-4}$. This corresponds to a change of the magnetoelastic energy ~ 0.1 cm^{-1}/atom.

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