Influence of a magnetic field on the crystal structure of dysprosium, III

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An x-ray study is made of magnetostriction of dysprosium in the ferro, antiferro- and paramagnetic states at 77—293°K in magnetic fields up to 16 kOe. It is shown that the change in the crystal lattice size in a magnetic field at $T_C < T < T_N$, due to changes in the orientation of the magnetic moments in the basal plane of the hexagonal close-packed lattice, is much greater than the effects due to magnetostriction of the para-process at $T < T_C$ and $T > T_N$. Magnetostriction deformations of the lattice in the antiferromagnetic state increase with increase of temperature.

It was shown earlier[1,2] that when the critical value of the magnetic field intensity H_{cr} is reached in dysprosium in the temperature interval 85—178°K the HCP lattice symmetry decreases to rhombic, and a jumplike change takes place in the crystal-lattice parameters. A similar behavior of the crystal structure was observed also in the first-order antiferromagnetism-ferromagnetism transition at 85°K[3,4]. This gave grounds for interpreting the structure effects in critical fields as the consequence of a magnetic transformation "with respect to the field," Unlike the usual (temperature) transformation, the phase transition in polycrystalline dysprosium in a magnetic field takes place in a certain interval of the magnetic field intensity ($\Delta H_{cr} \sim 2$—4 kOe), owing to the different orientations of the magnetic moment and of the magnetic field intensity vector H[2,5].

The presence of giant magnetostriction effects in dysprosium ($\lambda = \Delta l/l \sim 10^{-3} - 10^{-2}$) in the magnetically ordered state was noted in the literature a number of times[6-10]. The giant magnetostriction of dysprosium in relatively weak magnetic fields (~10 kOe) is obviously connected not only with the magnetoelastic deformation of the crystal lattice (rotation magnetostriction and of the para-process), but also with jumps in the crystal-lattice parameters in the antiferromagnetism-ferromagnetism transition[1] and with the reorientation of the rhombic domains of the ferromagnetic dysprosium in a magnetic field[2]. The x-ray diffraction method of measuring magnetostriction by determining the relative change of the interplanar distances (d_{hkl}) in a magnetic field[11] makes it possible to separate effects responsible for the giant magnetostriction of rare earth metals and, in particular, to measure the true deformation of the crystal lattice of dysprosium in a magnetic field.

The purpose of the present study was an x-ray diffraction investigation of the magnetic deformation of the crystal lattice of dysprosium 99.98% pure at 77—293°K in magnetic fields up to 16 kOe.

METHOD OF MAGNETOSTRICTION INVESTIGATION

The technique of x-ray diffraction experiments in a magnetic field is quite simple: the investigated flat polycrystalline samples, cooled by a stream of nitrogen vapor, are slowly rotated in their own plane in the gap of an electromagnet; the x-ray beam is normal to the surface; the beams diffracted in a "backward" geometry are recorded on photographic film. The x-rays for the diffraction by the dysprosium were produced by a chromium anticathode, and reflections from the planes (203), (210), (105), and 212) of the HCP lattice were registered. The accuracy with which the interplanar distances (d_{hkl}) were measured was not worse than 1×10^{-4} Å.

If the magnetic moment **M** makes angles α_i with the axes a_i of the orthogonal crystal lattice and is directed along the magnetic field **H**, then the magnetic deformation in the direction N_{hkl}, which makes angles β_i with the same axes, can be expressed in the form[12]

$$\lambda = \Delta d_{hkl}/d_{hkl} = \lambda_1^{\alpha,0}(\cos^2\beta_1 + \cos^2\beta_2) + \lambda_2^{\alpha,0}\cos^2\beta_3 + \lambda_1^{\alpha,2}(\cos^2\beta_1 + \cos^2\beta_2)(\cos^2\alpha_3 - 1/3) + \lambda_2^{\alpha,2}\cos^2\beta_3(\cos^2\alpha_3 - 1/3) + \lambda^{\gamma,2}[1/2(\cos^2\beta_1 - \cos^2\beta_2)(\cos^2\alpha_1 - \cos^2\alpha_2) + 2\cos\beta_1\cos\beta_2\cos\alpha_1\cos\alpha_2] + 2\lambda^{\varepsilon,2}(\cos\beta_1\cos\alpha_1 + \cos\beta_2\cos\alpha_2)\cos\beta_3\cos\alpha_3 + \ldots \quad (1)$$

The spontaneous magnetoelastic deformations $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ are connected with the appearance of magnetic ordering, and are not determined by the described procedure[1]. The quantities $\lambda_1^{\alpha,2}$ and $\lambda_2^{\alpha,2}$ characterize the deformations of the axes a and c in a magnetic field of intensity H, while $\lambda^{\gamma,2}$ describes the lowering of the symmetry in the basal plane of the HCP Lattice, and $\lambda^{\varepsilon,2}$ describes the deviation of the hexagonal axis from the normal to the basal plane (monoclinic distortions).

The direction cosines of the measurement direction (i.e., of the normal N_{hkl} to the crystallographic plane) and of the magnetic moment can be expressed in terms of the axes of the orthogonal lattice by using simple crystallographic relations of the type

$$\cos^2\beta_1 = d_{HKL}^2 H^2/a_r^2, \quad \cos^2\alpha_1 = a_r^2 U^2/R_{UVW}, \quad (2)$$

where

$$d_{HKL}^{-2} = a_r^{-2}H^2 + b_r^{-2}K^2 + c_r^{-2}L^2,$$

$$R_{UVW}^2 = a_r^2U^2 + b_r^2V^2 + c_r^2W^2.$$

The parameters of the orthogonal (rhombic) lattice are connected with the parameters of the hexagonal lattice by the relations

$$a_1 = a_p = a_r, \quad a_2 = b_p = a_r\sqrt{3}, \quad c_3 = c_p = c_r. \quad (3)$$

The matrices for the transition from hexagonal indices [(hkl), ⟨uvw⟩] of the planes and directions to rhombic ones [(HKL), ⟨UVW⟩], are

$$\begin{pmatrix} H \\ K \\ L \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 \\ 1 & 2 & 0 \\ 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} h \\ k \\ l \end{pmatrix}, \quad \begin{pmatrix} U \\ V \\ W \end{pmatrix} = \begin{pmatrix} 1 & -1/2 & 0 \\ 0 & 1/2 & 0 \\ 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} u \\ v \\ w \end{pmatrix}. \quad (4)$$

For the case of a polycrystal with arbitrary grain

orientation, the axes a_1, a_2, and a_3 make angles γ_i with the vector **H** of the magnetic field (the Oz axis of the laboratory frame, Fig. 1). In the spherical system connected with the vector N_{hkl}, the coordinates of the crystal axes are β_i and ψ (the angle ψ is measured from an arbitrary direction in the (HKL) crystallographic plane). The magnetic moment **M** of each grain makes with the vector **H** an angle δ, for which

$$\cos\delta = \cos\alpha_1\cos\gamma_1 + \cos\alpha_2\cos\gamma_2 + \cos\alpha_3\cos\gamma_3. \quad (5)$$

The deformation (λ) of a grain whose axes are oriented at angles γ_i to the axes of the laboratory frame can be easily converted into the "true" deformation λ' (when **M** ∥ **H**)

$$\lambda' = \lambda/\cos\delta. \quad (6)$$

The grains that produce reflections from the {HKL} crystallographic planes at an angle 2ϑ differ in the values of the azimuthal angle ψ. It is easy to show that

$$\cos\gamma_i = \cos\vartheta\cos\beta_i + \sin\vartheta\sin\beta_i\cos\psi. \quad (7)$$

Averaging over the angle ψ yields[2)]

$$\langle\cos^2\gamma_i\rangle = \frac{1}{2\pi}\int_0^{2\pi}\cos^2\gamma_i\,d\psi = \cos^2\vartheta\cos^2\beta_i + {}^1/_2\sin^2\vartheta\sin^2\beta_i. \quad (8)$$

Depending on the character of the magnetic structure of the dysprosium, expression (1) is significantly altered for magnetostriction in a direction specified by the angles β_i.

According to neutron-diffraction data[14] and the results of magnetic measurements in strong fields[15], we have in the case of ferromagnetic dysprosium (the easy axis is ⟨100⟩)

$$\cos\alpha_1 = 1, \quad \cos\alpha_2 = 0, \quad \cos\alpha_3 = 0.$$

In the antiferromagnetic state (helicoidal magnetic structure of the simple spiral type SS[14]) we have

$$\cos\alpha_1 = \cos\omega, \quad \cos\alpha_2 = \sin\omega, \quad \cos\alpha_3 = 0, \quad (9)$$

where ω is the angle of the helicoid, i.e., the "pitch" over which the position of the magnetic moment **M** changes on going from one basal plane to the neighboring one. In this case we obtain, for the calculation of the magnetostriction, the averaged orientations of **M**:

$$\langle\cos\alpha_1\rangle = \sqrt{2}/2, \ \langle\cos\alpha_2\rangle = \sqrt{2}/2, \ \langle\cos\alpha_3\rangle = 0. \quad (10)$$

We see that the averaged values of $\cos\alpha_i$ do not depend on the angle of the helicoid, i.e., this type of magnetic ordering is approximated in our case essentially by the "easy plane."

In the paramagnetic state, when the magnetic moments are oriented along the field, we have

$$\langle\cos\alpha_1\rangle = \langle\cos\gamma_1\rangle, \quad \langle\cos\alpha_2\rangle = \langle\cos\gamma_2\rangle, \quad \langle\cos\alpha_3\rangle = \langle\cos\gamma_3\rangle. \quad (11)$$

When the magnetic structure is taken into account, Eq. (1) acquires in the case of the ferromagnetic state the form

$$\lambda_F = -{}^1/_3\lambda_1^{\alpha,2}(\cos^2\beta_1 + \cos^2\beta_2) - {}^1/_3\lambda_2^{\alpha,2}\cos^2\beta_3 + {}^1/_2\lambda^{\gamma,2}(\cos^2\beta_1 - \cos^2\beta_2) \quad (12)$$

For the antiferromagnetic and paramagnetic states we have, respectively,

$$\lambda_{AF} = -{}^1/_3\lambda_1^{\alpha,2}(\cos^2\beta_1 + \cos^2\beta_2) - {}^1/_3\lambda_2^{\alpha,2}\cos^2\beta_3 + \lambda^{\gamma,2}\cos\beta_1\cos\beta_2 \quad (13)$$

and

$$\begin{aligned}\lambda_P = {}& \lambda_1^{\alpha,2}(\cos^2\beta_1 + \cos^2\beta_2)(\langle\cos^2\gamma_3\rangle - {}^1/_3) + \lambda_2^{\alpha,2}\cos^2\beta_3(\langle\cos^2\gamma_3\rangle - {}^1/_3) \\ & + \lambda^{\gamma,2}[{}^1/_2(\cos^2\beta_1 - \cos^2\beta_2)(\langle\cos^2\gamma_1\rangle - \langle\cos^2\gamma_2\rangle) + \\ & + 2\cos\beta_1\cos\beta_2(\langle\cos^2\gamma_1\rangle)^{1/2}(\langle\cos^2\gamma_2\rangle)^{1/2}] + 2\lambda^{\varepsilon,2}[\cos\beta_1(\langle\cos^2\gamma_1\rangle)^{1/2} \\ & + \cos\beta_2(\langle\cos^2\gamma_2\rangle)^{1/2}]\cos\beta_3(\langle\cos^2\gamma_3\rangle)^{1/2}.\end{aligned} \quad (14)$$

It follows from (12) and (13) that the monoclinic deformations $\lambda^{\varepsilon,2}$ for the ferro- and antiferromagnetic states cannot be determined by the described procedure. The magnetoelastic deformations $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$ and $\lambda^{\gamma,2}$ for the given temperatures and magnetic fields were determined from a system of three linear equations of the type (12) or (13) for the crystallographic planes $(203)_h$, $(210)_h$, and $(212)_h$. For the paramagnetic state, systems of four equations of type (14) were set up, and additional data were used on the field dependence of $d_{(105)}$; these data made it possible to determine also the monoclinic deformations.

In view of the relatively low accuracy of the x-ray diffraction measurements of the magnetostriction ($\delta\lambda \sim 10^{-3}$), the field dependences of the crystal-lattice deformations were approximated by straight lines.

RESULTS AND DISCUSSION

The procedure described above was used to calculate the field dependences of the magnetostriction deformations for ferro-, antiferro-, and paramagnetic dysprosium at 77, 120, 130, 150, 160, 175, and 185°K. The corresponding isotherms are shown in Figs. 2–4.

In the ferromagnetic state (77°K), $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$, and $\lambda^{\gamma,2}$ are relatively small. When $T < T_C$ we have apparently mainly the paraprocess magnetostriction, although a slight deviation of the magnetic moments from the ⟨100⟩ directions in the basal plane (001) is not excluded in principle[15]. The magnetostriction of dysprosium in the paramagnetic state is extremely small; the observed effects exceed the measurement errors only near the Neel point.

The magnetostriction effects are large only in the antiferromagnetic state. The anisotropy of the magnetic deformation of the crystal lattice ($\lambda_1^{\alpha,2} < 0$, $\lambda_2^{\alpha,2} > \lambda^{\gamma,2} < 0$) correlates well with anisotropy of the shift of T_N following compression along the axes $a_1(a)$ and $a_3(c)$[17]; by virtue of the known relation[16]

$$\left(\frac{\partial\lambda}{\partial H}\right)_{p,T} = -\frac{T}{T_N}\left(\frac{\partial\sigma_s}{\partial T}\right)_{p,H}\frac{\partial T_N}{\partial p}, \quad (15)$$

where σ_S is the spontaneous magnetization. All the $\lambda(H)$ isotherms for $T_C < T < T_N$ terminate at $H = H_{cr}$.[3)] All the $d\lambda_i/dH$ have a tendency to decrease with increasing temperature, but the final magnetic deformation of the crystal lattice in H_{cr} increases when the Neel point is approached (Fig. 5), owing to the strong increase of H_{cr} with temperature. Unfortunately, the presence of a magnetoheterogenerous state at $H > H_{cr}$[2]

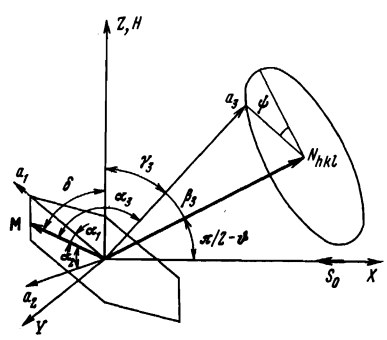

FIG. 1. Experimental setup for the study of the magnetic deformation of a crystal lattice.

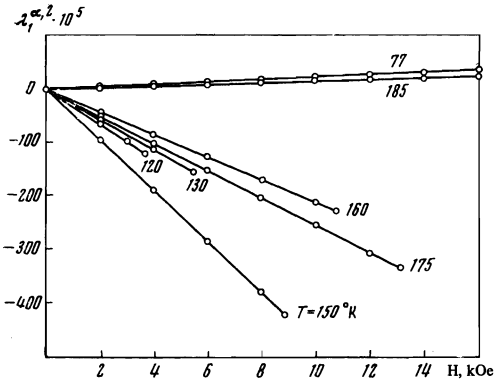

FIG. 2. Isotherms of field dependence of the magnetic deformation $\lambda_1^{\alpha,2}$.

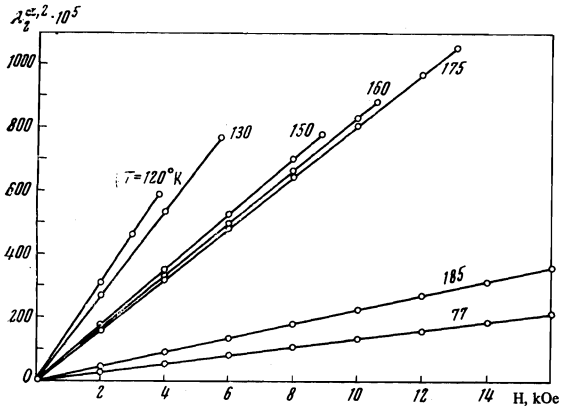

FIG. 3. Isotherms of field dependence of the magnetic deformation $\lambda_2^{\alpha,2}$.

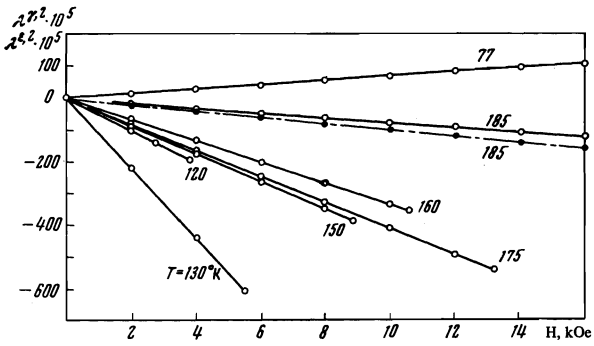

FIG. 4. Isotherms of field dependence of the magnetic deformations $\lambda^{\gamma,2}$(O) and $\lambda^{\varepsilon,2}$(O).

made it impossible to measure the magnetostriction in the field-induced ferromagnetic state in the temperature range $T_C < T < T_N$.

Although the change of the magnetic structure does not enter explicitly in the calculation formulas (13) for the antiferromagnetic state, it should be assumed that the observed large magnetostriction effects are connected precisely with the rotation magnetostriction, i.e., with the decrease of the helicoid angle ω with increasing magnetic field intensity ("twisting" of the antiferromagnetic helix is observed when the temperature is lowered[14]; it is accompanied by positive lattice deformations along the c axis and negative ones along a[3,4]).

From the result of the present earlier[1,2] studies we can deduce the main features of the antiferromagnetism-ferromagnetism phase transition in dysprosium in a magnetic field: When the temperature is raised, the

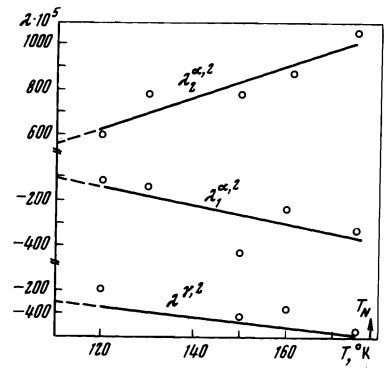

FIG. 5. Temperature dependence of the magnetic deformations at $H = H_{cr}$. The lines are linear approximations by least squares.

magnetic field that destroys the helicoidal antiferromagnetic structure (H_{cr}) increases, the jumps of the crystal-lattice parameters and the atomic volume[4)] decrease, and the magnetic deformation of the crystal lattice increases.

If the free energy F of the magnetically ordered crystal in an external magnetic field is represented in the form[12,19]

$$F = E_{exch} + E_{ell.} + E_{m.ell} \tag{16}$$

where E_{exch} is the exchange-interaction energy, E_{el} is the elastic energy, and $E_{m.el}$ is the magnetoelastic energy in an extraneous magnetic field, then the presence of an energy gap between the ferro- and antiferromagnetic states (ΔE) can be related in first order with the thermal effect of the phase transition (with the aid of the Clausius-Clapeyron equation)

$$q = \left(\frac{1}{T_c}\frac{dT_c}{dp}\right)^{-1}\Delta V = \Delta E_{exch} + \Delta E_{el}. \tag{17}$$

and with the change of the work of the magnetostriction

$$E_{m.el} = {}^1/_2 c_{11}(\lambda_1)^2 + {}^1/_2 c_{22}(\lambda_2^{\alpha,2})^2 + {}^1/_8 c^{\gamma}(\lambda^{\gamma,2})^2. \tag{18}$$

The elastic moduli c_{11}^{α}, c_{22}^{α}, and c^{γ} in (18) are connected with the elastic moduli c_{ij} in the ordinary matrix notation by the relations[12]

$$\begin{aligned} c_{11}^{\alpha} &= {}^1/_9(2c_{11} + 2c_{12} + 4c_{13} + c_{33}),\\ c_{22}^{\alpha} &= {}^2/_3(c_{11} + c_{12} - 4c_{13} + 2c_{33}),\\ c^{\gamma} &= 2(c_{11} - c_{12}). \end{aligned} \tag{19}$$

The values of c_{ij} of dysprosium were obtained in[20], the shift of the Curie point under pressure was measured in[17], and the jump of the atomic volume (ΔV) in the antiferromagnetism-ferromagnetism transition is ~0.2% at the Curie point.[4]. These data, together with the present results, enable us to calculate the change of the exchange and elastic energies in a phase transition "with respect to the field," on the one hand, and the change of the magnetic energy, on the other (Fig. 6).

As seen from the figure, in spite of the decrease[5)] of $\Delta E_{exch} + \Delta E_{el}$ with increasing temperature, the energy gap between the antiferro- and ferromagnetic states of dysprosium increases, owing to the appreciable increase of the magnetoelastic energy in the magnetic field.

If, as assumed above, the spontaneous and induced deformations of the crystal lattice following a change in temperature and magnetic field are connected with processes of rotation of the magnetic moment in the basal plane (i.e., with the "twisting" of the helicoid), then the results obtained here can be treated from the point of view of the change of the magnetic structure. By normalizing (in accordance with the data of[4,14])

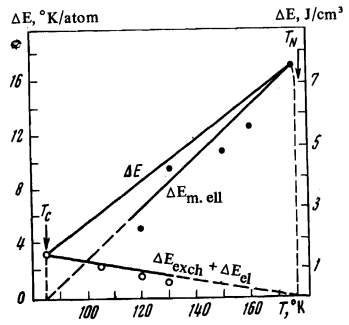

FIG. 6. Temperature dependence of the exchange and elastic (○) and of the magnetoelastic (●) energies and of the width of the nergy gap between the antiferro- and ferromagnetic states.

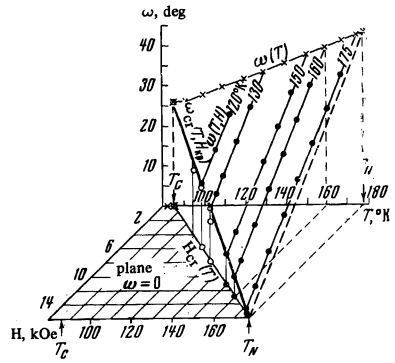

FIG. 7. Temperature and field dependences of the helicoid angle ω (scheme): X—neutron-diffraction data [14]; $\circ - \omega(T, H) = \omega(T) + \Delta\omega\lambda_2^{\alpha,2}/\Delta\lambda_c$; $\bullet - \omega(T, H) = \Delta\omega_{T_C}\frac{\Delta V}{V}(T) \Big/ \left(\frac{\Delta V}{V}\right)_{T_C}$

the change of the helicoid angle ω relative to the maximum effect, or the lattice deformation λ_c along the principal axis (at $T_C < T < T_N$), we obtain $\Delta\omega/\Delta\lambda_c \sim -3.3 \times 10^3$ deg. We can also normalize the angle of the helicoid against the jump of the volume at T_C ($(\Delta V/V)^{-1}\Delta\omega \sim +1.3 \times 10^4$ deg).

With the aid of these criteria we plotted a semiquantitative $\omega - T - H$ diagram (Fig. 7). As seen from the diagram, the helicoid angle decreases with decreasing temperature and increasing magnetic field. The quantities ω_{cr} (at $H = H_{cr}$), estimated from the magnetic deformation $\lambda_2^{\alpha,2}$ and from the jump of the volume in H_{cr}, are in satisfactory agreement. The value of ω_{cr} decreases with increasing temperature. We note once more that these constructions are only schematic; the postulated linearity of $\omega(\lambda_2^{\alpha,2})$ and $\omega(\Delta V/V)$ can in fact be more complicated.

[1)]The spontaneous deformations of the axes a and c below the Neel point can be obtained by extrapolating the a(T) and c(T) curves from the paramagnetic region [13]: $\lambda_1^{\alpha,0} = (a_{meas} - a_{extr})/a_{extr}$, $\lambda_2^{\alpha,2} = (c_{meas} - c_{extr})/c_{extr}$.

[2)]Since the magnetostriction effect is even, $\cos\gamma_i$ averages out.

[3)]The critical field H_{cr} was determined in the experiment by the appearance on the x-ray patterns of the first lines of ferromagnetic (rhombic) dysprosium.

[4)]And also of the magnetization jump $\Delta\sigma$ [14].

[5)]I.e., the decrease of the jumps of the crystal-lattice parameters [2].

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