For a final determination of  $\eta$  we must find A in the zeroth order in a. Multiplying (B.5) by y and  $y^2$  and integration, we obtain equations for

$$W_1(z) = \int \mathscr{P}(z, y) y \, dy$$
 and  $W_2(z)$ .

We have

$$\frac{1}{4}\frac{d^2W_1}{dz^2} - \frac{1}{4}\frac{d}{dz}\left(\frac{W_1}{z}\right) - \frac{W_1}{z^2} = \frac{1}{2}\frac{dW}{dz},$$
 (B.8)

$$\frac{1}{4}\frac{d^2W_2}{dz^2} - \frac{1}{4}\frac{d}{dz}\left(\frac{W_2}{z}\right) - 2\frac{W_2}{z^2} = \frac{dW_1}{dz} - \frac{1}{2}W.$$
 (B.9)

Substituting W(z) = z in (B.8) and (B.9) we obtain

$$W_1(z) = -\frac{2}{3}z^2; \quad W_2(z) = \frac{11}{4}z^3$$

Thus, A = 11/6 and we get for  $\eta$ :

$$\eta = 1 - \frac{100}{3}a^2$$
. (B. 10)

We have  $\beta = \partial^2 \varepsilon / \partial \varphi^2 |_{\varphi=\gamma} = z + 4ay$ . Since  $\overline{y}(z) = z^{-1}W_1(z) \sim z$ ,  $a \ll 1$ , the distribution function  $W(\beta)$  takes the form (B.7) with the same value of  $\eta$ .

<sup>1</sup>P. A. Lee, T. M. Rice, and P. W. Anderson, Sol. State Comm. 17, 1089 (1975).

- <sup>2</sup>H. Fukuyama and P. A. Lee, Phys. Rev. B17, 535 (1977).
  <sup>3</sup>W. J. Gunning, A. J. Heeger, J. F. Schcegolev, S. P. Zolotukhin, Sol. State Comm. 25, 981 (1978). J.-J. Andre, A. Beeber, and F. Gauntier, Ann. de Phys. 1, 145 (1976). K. C. Carneiro, G. Shirane, S. A. Werner, and S. Kaiser, Phys. Rev. B13, 4258 (1976).
- <sup>4</sup>D. C. Mattis, Phys. Lett. A56, 421 (1976). G. Toulouse, Comm. Phys. 2, 115 (1977). J. Villain, J. Phys. C10, 1717 (1977).
- <sup>5</sup>D. J. Scalapino, M. Sears, and R. A. Ferrel, Phys. Rev. **B6**, 3409 (1972).
- <sup>6</sup>H. Schmidt, Phys. Rev. 105, 425 (1957).
- <sup>7</sup>S. A. Gradeskul and L. A. Pastur, Fiz. Nizk. Temp. 1, 277 (1975) [Sov. J. Low Temp. Phys. 1, 135 (1975)].
- <sup>8</sup>Y. Ymry and S.-k Ma, Phys. Rev. Lett. 35, 1399 (1975).
- <sup>9</sup>L. P. Gor'kov, Pis'ma Zh. Eksp. Teor. Fiz. 25, 384 (1977) [JETP Lett. 25, 358 (1977)].
- <sup>10</sup>K. B. Efetov, Zh. Eksp. Teor. Fiz. 75, 1885 (1978) [Sov. Phys. JETP 48, 949 (1978)].
- <sup>11</sup>P. W. Anderson, B. I. Halperin, and C. M. Varma, Phil. Mag. 25, 1 (1972).
- <sup>12</sup>S. A. Brazovskii, Zh. Eksp. Teor. Fiz. 76, 1000 (1979) [Sov. Phys. JETP 49, 557 (1979)].
- <sup>13</sup>R. Harris, M. Plischke, and M. J. Zuckermann, Phys. Rev. Lett. 31, 160 (1973).

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# Effect of a strong magnetic field on the spin-reorientation transition in $DyCo_{5.3}$

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Magnetization and magnetostriction measurements have been made on the intermetallic compound  $DyCo_{5,3}$ in strong pulsed magnetic fields (up to 250 kOe), at temperatures close to the spin-reorientation transition that is observed in this compound and that is of the easy-plane, easy-cone, easy-axis type. The experimental data are compared with theoretical relations obtained by consideration of a model of a two-sublattice ferrimagnet, each sublattice of which has its own magnetocrystalline anisotropy; the uniaxial anisotropy constants of the sublattices have different signs and are comparable in magnitude with the intersublattice exchange interaction. It is shown that such a model describes the magnetic properties of  $DyCo_{5,3}$  in strong fields; values of the anisotropy constants of the sublattices are obtained from the experimental data. It is significant that in the analysis of magnetization curves it was necessary to allow for a noncollinear magnetic structure that occurs in strong fields, and also for a dependence of the value of the magnetic moment of the Dy sublattice on direction.

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## INTRODUCTION

The compound  $DyCo_{5.3}$ , like other rare-earth compounds  $RCo_5$ , has a hexagonal crystallographic structure of the  $CaCu_5$  type (space group P6/mmm). This structure may be regarded as consisting of alternating layers of cobalt atoms, placed perpendicular to the hexagonal axis; the atoms of the rare-earth metal (REM) occupy hexagonal positions in every second layer.<sup>1,1)</sup> A decisive role in the formation of the magnetism in  $RCo_5$  is played by the Co-Co exchange interaction, which causes the high Curie temperatures<sup>3</sup> (about 1000 K) of these compounds. The R-Co interaction is about an order of magnitude weaker; the exchange interaction within the *R* sublattice can as a rule be neglected. Thus the REM atoms constitute a paramagnetic system located in an effective field produced by the Co sublattice. The compounds  $RCo_5$  with light REM have ferromagnetic ordering; those with heavy, ferrimagnetic.

A distinctive feature of many  $RCo_5$  compounds is the fact that at low temperatures their uniaxial magnetic

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anisotropy reaches enormous values, of the order  $10^8$  erg/cm<sup>3</sup>, comparable with the energy of R-Co exchange interaction. <sup>3-6</sup> This makes RCo<sub>5</sub> compounds good objects for study of the properties of magnetic materials in which the isotropic exchange interactions and the anisotropic interactions are comparable with each other.

It has been shown in a number of  $papers^{5-6}$  that the magnetic properties of RCo<sub>5</sub> can be treated on the basis of a model of a two-sublattice magnet in which each sublattice, R and Co, possesses its own crystalline anisotropy; the constants of uniaxial anisotropy of these sublattices may have different signs. On the basis of such a model, it has been possible to obtain a simple and graphic explanation of the phenomenon of spin reorientation that it observed in several RCo, on rise of temperature. It has been shown that in these cases, at low temperatures the dominant role is played by the negative uniaxial anisotropy of the R sublattice, and the magnetic moments of both sublattices, coupled by exchange interaction, lie in the basal plane.<sup>2)</sup> With rise of temperature, the anisotropy of the R sublattice drops considerably faster than does the positive uniaxial anisotropy of the Co sublattice, and this leads to a change of the sign of the resultant anisotropy constant and to reorientation of the magnetic moments of the sublattices to the hexagonal axis. The change of the easy direction occurs within a certain temperature interval; here an intermediate magnetic structure occurs, in which the magnetic moments of the sublattices are noncollinear with each other, and the resultant magnetic moment is directed at an angle to the hexagonal axis, forming a cone (without allowance for anisotropy in the basal plane) of directions of easy magnetization.

We note that the origin of such a noncollinear structure can be explained within the framework of a model that takes into account only the first anisotropy constant of each sublattice, but they must have different signs. Then the anisotropies of the two sublattices tend to orient the moments in two mutually perpendicular directions; this is opposed by the intersublattice exchange interaction, for which collinear arrangement of the spins is advantageous. It is easy to show that if the moments of the sublattices are not oriented along the axis of symmetry (or in the plane of symmetry) of the crystal, then the noncollinear magnetic structure will be energetically more advantageous; the degree of noncollinearity depends, of course, on the relation between the anisotropy constants and the value of the exchange interaction, and also on the angles that the magnetic moments of the sublattices make with the hexagonal axis.

Since in a spin-reorientation transition the value of the energy of uniaxial anisotropy of the R sublattice remains large and comparable with the energy of intersublattice exchange interaction (all that is small in the present case is the resultant anisotropy, which is approximately equal to the algebraic sum of the anisotropy constants of the R and Co sublattices), the value of the magnetic moment of the R sublattice depends strongly on its orientation with respect to the crystallographic axes (this mechanism for an effect of the anisotropy on the magnetic moment of a ferromagnet was first considered by E. and H. Callen<sup>7</sup>).

A jump of the magnetization in a spin-reorientation transition has been observed experimentally in the compounds  $DyCo_{5,2}$  and  $TbCo_{5,1}$ .<sup>4</sup> A semiquantitative theory has been constructed<sup>4-6</sup> to explain the presence and the amount of the jump of the magnetic moment the R sublattice in these compounds. It was shown that the crystalline anisotropy leads to a disturbance of the equidistant splitting of the levels of the REM atom in the molecular field produced by the Co sublattice. If the magnetic moments of the REM are directed along the hexagonal axis, then the lower levels of the multiplet draw together; but if the REM moments are oriented in the basal plane, they move apart. Thus the population of the multiplet levels, and consequently the value of the magnetization at nonzero temperature, depend on the direction of the magnetic moment of the R sublattice with respect to the crystallographic axes. We note, however, that jumps of the magnetic moment of the R sublattice in RCo<sub>5</sub> close to the experimentally observed were obtained<sup>4</sup> for various combinations of the constants of exchange interaction and of R-sublattice anisotropy. Therefore the question of the actual values of these interactions remains open.

Hitherto, it is principally the behavior of the  $RCo_5$ magnetization in zero field that has been analyzed and compared with experimental data; the nature of the influence of a strong magnetic field on the spin-reorientation transition in these compounds has also remained unexplained. It is clear, however, that the action of an external field whose energy of Zeeman interaction with the R sublattice is comparable with the anisotropy energy of the latter should lead to a change of the R-sublattice magnetic moment comparable with the magnetization jump caused by the spin-reorientation transition on change of temperature, and therefore should serve as a criterion for the correctness of the model being considered.

The present paper is devoted to investigation of the effect of a strong magnetic field on the magnetic structure and magnetization of a single crystal of  $DyCo_{5.3}$  in the region of the spin reorientation transition (330-370 K). Experimental data on magnetization and magnetostriction of  $DyCo_{5.3}$ , obtained in pulsed magnetic fields up to 250 kOe over the temperature range 200-500 K, are compared with theoretical results based on the model described above. A conclusion is drawn regarding the applicability of this model to a description of magnetization curves of  $DyCo_{5.3}$  in strong magnetic fields; from the experimental data, values of the anisotropy constants of the R and Co sublattices are obtained.

### SPECIMENS AND EXPERIMENTAL METHOD

The single crystals used for the measurements were the same as in the preceding paper<sup>8</sup>; they had a singlephase composition, with misorientation of the subgrains no greater than 3°. The magnetic field was measured with absolute accuracy  $\pm 7\%$ ; the specimen temperature was determined with a copper-constantan thermocouple with error  $\pm 1$  K.

The magnetization was measured by the induction method, with absolute accuracy no worse than 7%. In the magnetostriction measurements, only the relative variation of the strain with the external field was determined. The measurement method and the manner of preparation of the specimens have been described earlier in more detail.<sup>8</sup>

## **RESULTS OF THE MEASUREMENTS**

Figures 1a, b, and c show typical magnetization curve of  $DyCo_{5,3}$  along the hexagonal axis (1) and in the basal plane (2), at various temperatures: below the spin-reorientation region (a, 320 K), when the direction of easy magnetization lies in the basal plane; in the spinreorientation region (b, 350 K), when the direction of easy magnetization lies at an angle to the hexagonal axis; and above the reorientation region (c, 430 K), when the axis of easy magnetization is the hexagonal axis of the crystal. It is seen that along the hard direction there is a nonlinear increase of the magnetization, due to rotation of the resultant magnetic moment toward the field direction; in fields exceeding the saturation field,<sup>3)</sup> and also along the easy direction, there occurs a linear variation of the magnetization with the field. The following features of the magnetization behavior are noteworthy: at fields larger than the saturation field (or, at temperatures above the spin reorientation, over the whole range of fields), the magnetization along the hexagonal axis exceeds the magnetization in the basal plane; the paraprocess susceptibility (that is, the susceptibility in the ferrimagnetic collinear phase) depends on the direction. The latter fact is clearly evident from Fig. 1d, which shows the temperature variation of the susceptibility along (black diamonds) and perpendicular to (white diamonds) the hexagonal axis. The data presented show that the susceptibility in the basal plane exceeds considerably (by about 30%) the susceptibility along the hexagonal axis.



FIG. 1. a, b, c: Experimental field dependences of the magnetization of  $DyCo_{5,3}$  in a field applied along (1) or perpendicular (2) to the hexagonal axis, at temperature: a) 320 K; b) 350 K; c) 430 K. Triangles: values of the magnetization in zero field along the hexagonal axis; circles: in the basal plane. Black symbols: measurements along the easy direction; white: along the hard direction (see text). d) Temperature dependence of the susceptibility along (black diamonds) and perpendicular to (white diamonds) the hexagonal axis.



FIG. 2. Temperature variation of the magnetization of  $DyCo_{5.3}$  in zero external field. The triangles and circles denote the same values of magnetization as in Fig. 1. Squares: magnetization along an easy direction located at an angle to the hexagonal axis. Crosses: difference of magnetizations along the hexagonal axis and in the basal plane.

In Fig. 2, the black symbols show the temperature dependence of the spontaneous magnetizaton of DyCo<sub>5,3</sub>, measured along the easy (at the given temperature) direction, in fields sufficient for technical saturation of the specimen. There is a clearly marked anomalous increase of the spontaneous magnetization within the interval 330-370 K; this was first discovered by Ermolenko et al.<sup>4</sup> We note that our data on the temperature dependence of the spontaneous magnetization agree, within the limits of accuracy of the experiment, with the results of that paper. In the same figure, the white circles show the values of magnetization that would occur of, with rise of temperature, the magnetic moments remained in the basal plane. The white triangles have a similar meaning: these are the values of magnetization that would be observed if, on lowering of temperature, the moments of the sublattices remained parallel to the hexagonal axis.

We shall discuss in somewhat greater detail the reason for determining these hypothetical values of magnetization and the method of finding them. It is obvious that for elucidation of the effect of crystalline anisotropy on the value of the magnetic moment of the R sublattice, it would be simplest to compare, at a fixed temperature, the values of the magnetic moment along and perpendicular to the hexagonal axis. But at a given temperature, the direction of easy magnetization makes a single, completely determined angle with the hexagonal axis. Therefore in order to obtain the desired values of the magnetization, it is necessary by some method (for example with an external field) to turn the magnetic moments of the sublattices into the necessary direction, excluding meanwhile secondary action of the orienting force on the value of the magnetization. In our case, this was accomplished by extrapolation of the section of paraprocess increase of the magnetization, above the saturation field, to zero field (dotted in Fig. 1a, b, c). Here it was assumed, of course, that the value of the paraprocess susceptibility of such a hypothetical state would remain equal to the susceptibility above the saturation field. The fact that in the latter case the value of the susceptibility was found to be independent of the external field justifies this asumption.

The crosses in Fig. 2 show the temperature dependence of the jump of the magnetic moment of the Dy sublattice,<sup>4)</sup> determined as the difference of the mag-



FIG. 3. Experimental (symbols) and theoretical (solid lines) temperature variations of the saturation field of  $DyCo_{5.3}$  along the hexagonal axis (1) and in the basal plane (2). Circles, from measurements of the field-dependence curves of the magnetization; squares, from magnetostriction measurements.

netizations along and perpendicular to the hexagonal axis in zero external field (the latter were found either from measurements of the spontaneous magnetization or by extrapolation to zero field). It is seen that the magnetization difference within the interval 320-440 K varies little with temperature and is  $135 \pm 15 G$  (or  $1.25 \pm 0.15 \mu_B$  per formula unit of  $DyCo_{5.3}$ ); this is below the value 1.6  $\mu_B$  obtained in Ref. 4 because of the fact that in the latter paper, the magnetization jump was determined as the difference of magnetizations at different temperatures (at the beginning and at the end of the transition).

Figure 3 shows the temperature variation of the saturation fields  $H_s$  along the hexagonal axis (white squares and circles) and in the basal plane (white circles). Along the hexagonal axis, the saturation field increases rapidly with lowering of temperature and for T < 290 K is beyond the range of experimentally attainable fields. It is also evident that the saturation fields determined from magnetization measurements (circles) agree well with the fields obtained from measurements of the magnetostriction (squares). We note that the transition to the ferrimagnetic collinear phase was considerably more pronounced on the field-dependence curves of the magnetostriction than on the magnetization curves, where this transition had a smooth character; for reliable determination of the saturation field, it was necessary to have a rather extended section of magnetization increase caused by the paraprocess, above the saturation field.

## THEORY

For accurate calculation of the orientation of the magnetic moments of the sublattices with respect to the crystallographic axes and of the value of the magnetic moment of the R sublattice in  $RCo_3$  compounds, it has been proposed<sup>4-6</sup> to use the method of diagonalization of the exchange-interaction and crystalline-field operator. This methid, however, encounters serious mathematical difficulties even without consideration of the external magnetic field. For this reason, for description of the magnetic characteristics of  $DyCo_{5.3}$  we used the significantly simpler method of the magnetic moment of the R sublattice on direction was taken into

account phenomenologically, by choice of the form of the explicit dependence of the anisotropy constant on the value of the magnetization.

The nonequilibrium free energy of  $DyCo_{5.3}$  can be represented in the following form<sup>5</sup>:

$$\mathfrak{D} = \Phi_0 + \frac{1}{2} \alpha M_{\mathrm{R}}^2 - H_{\mathrm{M}} M_{\mathrm{R}} \cos \left(\theta_{\mathrm{R}} - \theta_{\mathrm{Co}}\right) - K_{\mathrm{IR}} \sin^2 \theta_{\mathrm{R}} + K_{\mathrm{ICo}} \sin^2 \theta_{\mathrm{Co}}$$

$$+ K_{2\mathrm{Co}} \sin^4 \theta_{\mathrm{Co}} - H M_{\mathrm{R}} \cos \left(\psi_{\mathrm{H}} - \theta_{\mathrm{R}}\right) - H M_{\mathrm{Co}} \cos \left(\psi_{\mathrm{H}} - \theta_{\mathrm{Co}}\right).$$
(1)

Here  $\alpha$  is a phenomenological parameter,  $H_{\rm M}$  is the molecular field exerted on the R sublattice by the Co sublattice,  $M_{\rm R}$  and  $M_{\rm Co}$  are the magnetic moments of the sublattices,  $\theta_{\rm R}$ ,  $\theta_{\rm Co}$ , and  $\psi_{\rm H}$  are the angles made by the magnetic moments of the sublattices and by the field with the hexagonal axis,  $K_{\rm 1R}$  is the first anisotropy constant of the R sublattice,  $K_{\rm 1Co}$  and  $K_{\rm 2Co}$  are the anisotropy constants of the Co sublattice, and H is the external field.

In writing the free energy (1), we have made several model approximations, the principal one of which consists in the fact that the anisotropy energy has been represented in the form of an additive correction to the exchange energy; for magnets in which these energies are comparable, this is in general incorrect.<sup>9</sup> But as will be seen from what follows, within the temperature range under consideration the anisotropy energy of the Dy sublattice is approximately an order of magnitude smaller than the energy of interaction with the molecular field, and this to a considerable degree excuses our writing the free energy in the form (1).

The following approximation is based on the fact that temperatures above ~300 K may be regarded as high for the Dy sublattice (at 300 K the magnetic moment of the Dy sublattice is down by a factor of almost two as compared with 0 K); this enables us to neglect the anisotropy constants of subsequent orders in comparison with  $K_{1R}$ . Use of the results of single-ion theory in conjunction with the high-temperature approximation provides the possibility of representing  $K_{1R}$  in the form  $aM_{\rm R}^2$ , where a is positive and independent of temperature. For the Co sublattice, on the contrary, temperatures of the order of 300 K may be considered low; therefore in the first approximation, we neglected the temperature dependence of the magnetization and anisotropy constants of the Co sublattice. Purely formally, for the purpose of symmetrizing later formulas, these constants were represented in the form  $K_{1Co} = b_1 M_{Co}^2$ ,  $K_{2Co} = b_2 M_{Co}^2$ .

The parameter  $\alpha$  in formula (1) is easily determined by considering the case when the field is directed along the hexgonal axis and the sublattices are collinear with the field: then minimization of (1) with respect to  $M_R$ gives  $\alpha = 1/\chi_{\parallel}$ .

The free energy (1) is a function of three variables:  $M_{\rm R}$ ,  $\theta_{\rm Co}$ , and  $\theta_{\rm R}$ . Minimization with respect to them enables us to obtain equations that describe the variation of these quantities with field and temperature. But it is more convenient to transform to new variables  $M_{\rm R}$ ,  $\theta_{\rm Co}$ , and  $\Delta \Theta = \theta_{\rm R} - \theta_{\rm Co} - \pi$ , where  $\Delta \theta$  is the angle that characterizes the noncollinearity of the sublattices; we shall regard it as small in comparison with  $\theta_{\rm R}$  and  $\theta_{\rm Co}$ . Hereafter we shall restrict ourselves to consideration of the two cases in which the external field is applied along the hexagonal axis  $(\psi_H = 0)$  or lies in the basal plane  $(\psi_H = \pi/2)$ . Then by minimizing (1) with respect to the new variables, we can obtain two equations, one of which corresponds to the collinear phase  $(\theta_{Co} = \psi_H, \Delta \theta = 0)$  and the other to a noncollinear arrangement of the sublattice moments:

$$M_{\rm R} = M_{\rm oR_{\rm H}} + \frac{2aM_{\rm oR_{\rm H}}\sin^2\theta_{\rm R} - H\cos\left(\psi_{\rm H} - \theta_{\rm R}\right) - \frac{1}{2}H_{\rm M}\sin^2\Delta\theta}{1/\chi_{\rm H} - 2a\sin^2\theta_{\rm R}},\qquad(2)$$

$$\sin \Delta \theta = \frac{aM_{\rm R} \sin 2\theta + H \sin(\psi_{\rm H} - \theta)}{H_{\rm H} - 2aM_{\rm R} \cos 2\theta - H \cos(\psi_{\rm H} - \theta)}; \qquad (3)$$
$$H_{\rm H}M_{\rm R}[2(K_{\rm HC} - K_{\rm H}) \cos \theta + 4K_{\rm HC} \sin^2 \theta \cos \theta]$$

$$H_{M}m_{R}[2(K_{1Co} - K_{1R}) \cos \theta + 4K_{2Co} \sin^{2} \theta \cos \theta + HM_{Co}) +H(M_{Co} - M_{R})] - (2K_{1Co} \cos \theta + 4K_{2Co} \sin^{2} \theta \cos \theta + HM_{Co}) \times (2K_{1R} \cos 2\theta + HM_{R} \cos \theta) = 0, \quad \psi_{R} = 0,$$

$$H_{M}m_{1}[2(K_{1C} - K_{1C}) \sin \theta + 4K_{1C} \sin^{2} \theta - H(M_{1C} - M_{1C})]$$
(4a)

$$-(2K_{1co}\sin\theta + 4K_{2co}\sin^{3}\theta - HM_{co})$$

$$\times (2K_{1R}\cos 2\theta + HM_{R}\sin\theta) = 0, \quad \psi_{R} = \pi/2; \quad (4b)$$

where  $M_{0R\parallel}$  is the magnetization of the R sublattice when it is oriented along the hexagonal axis in zero field (in the sense indicated above), and analogously  $M_{0R\perp}$  (see below) is the magnetization of the R sublattice in the basal plane in zero field;  $\theta \equiv \theta_{Co}$ .

It is not possible in general to obtain a solution of the system (2)-(4) in analytical form; therefore it is expedient to begin consideration of it with the solution of a simpler special case, absence of the external field (H=0). Then besides the two collinear phases ( $\theta_{Co}=0$ ,  $\theta_R=\pi$  and  $\theta_{Co}=\pi/2$ ,  $\theta_R=-\pi/2$ ), there is a solution that describes a noncollinear phase (in the spin-reorienta-tion region)<sup>61</sup>:

$$H_{\mathbf{x}}M_{\mathbf{R}}(K_{1C_0} + 2K_{2C_0}\sin^2\theta - K_{1\mathbf{R}}) - 2K_{1\mathbf{R}}(K_{1C_0} + 2K_{2C_0}\sin^2\theta)\cos 2\theta = 0.$$
 (5)

Hence it is easy to obtain the conditions corresponding to the beginning ( $T_1 = 330$  K) and end ( $T_2 = 370$  K) of the spin-reorientation transition:

$$H_{\mathbf{x}}M_{0R\perp}(K_{1Co}+2K_{2Co}-K_{1R})+2K_{1R}(K_{1Co}+2K_{2Co})=0, \quad T=T_{1}$$
(6a)  
$$H_{\mathbf{x}}M_{0R\parallel}(K_{1Co}-K_{1R})-2K_{1R}K_{1Co}=0, \quad T=T_{2},$$
(6b)

and also the value of the jump of magnetic moment for a fixed temperature:

$$\Delta M_{\rm Ro} = M_{\rm oR_{\perp}} - M_{\rm oR_{\parallel}} = \frac{2aM_{\rm oR_{\parallel}}}{1/\chi_{\parallel} - 2a}.$$
(7)

In the presence of an external field, one can obtain simple relations for the values of the susceptibility of the R sublattice in the collinear phase  $(\theta = \psi_H, \Delta \theta = 0)$ :

$$\gamma^{-1} = \gamma_{\rm H}^{-1} - 2a\sin^2\psi_{\rm H}, \tag{8}$$

whence the difference between the longitudinal and transverse susceptibility (along the hexagonal axis and the basal plane) is

$$\Delta \chi = \chi_{\parallel} - \chi_{\perp} = -2a\chi_{\perp}\chi_{\parallel}. \tag{9}$$

An exact solution (when  $\chi$  is independent of *H*) can be obtained also for the saturation fields. By saturation field we understand in this case the critical field for transition from the noncollinear phase to a ferrimagnetic collinear phase. We purposely do not use here the term "anisotropy field" because for ferrimagnets, especially ones possessing different anisotropy constants for the two sublattices, there is in general no well-defined anisotropy field. This is evident in the example of a ferrimagnet possessing a magnetic compensation temperature (near this temperature, there is a region where, over the whole magnetic-field range, for fields applied along the hard axis, collinear ferrimagnetic structure is absent<sup>9</sup>).

On transition from the noncollinear phase to the ferrimagnetic collinear (as has been shown,<sup>9</sup> this transition is a phase transition of the second kind) the relations  $\theta = \psi_H$ ,  $\Delta \theta = 0$  are valid, and equations (4) reduce to ones quadratic in *H*; their solution has the form

$$H = Q \mp (Q^{2} - P)^{\frac{1}{2}};$$
(10)  
$$Q = \frac{\frac{1}{2}A (M_{co} - M_{0R\parallel}) - (b_{1}M_{co} + aM_{0R\parallel}) + 2a\chi_{\parallel} (AM_{0R\parallel} + b_{1}M_{co})}{(1 - A\chi_{\parallel}) (1 - 2a\chi_{\parallel})},$$
(11a)  
$$P = \frac{-2A (b_{1}M_{co}^{2} - aM_{0R\parallel}^{2}) + 4ab_{1}M_{0R\parallel}M_{co}}{(1 - A\chi_{\parallel}) (1 - 2a\chi_{\parallel})},$$

in the case when the field is applied along the hexagonal axis, and

$$Q = \frac{{}^{i}/{}_{2}A \left(M_{co} - M_{0R_{\perp}}\right) + \left(bM_{co} + aM_{0R_{\perp}}\right) - 2a\chi_{\perp} \left(AM_{0R_{\perp}} - bM_{co}\right)}{\left(1 - A\chi_{\perp}\right) \left(1 + 2a\chi_{\perp}\right)} \quad (11b)$$

$$P = \frac{2A \left(bM_{co}^{2} - aM_{0R_{\perp}}^{2}\right) + 4abM_{0R_{\perp}}M_{co}}{\left(1 - A\chi_{\perp}\right) \left(1 + 2a\chi_{\perp}\right)}, \quad b = b_{1} + 2b_{2}$$

in the case when the field is directed in the basal plane. In formulas (11), A is the parameter of R-Co exchange interaction:  $H_{\rm M} = A M_{\rm Co}$ . We recall that  $K_{1\rm R} = a M_{\rm R}^2$ ,  $K_{1\rm Co} = b_1 M_{\rm Co}^2$ , and  $K_{2\rm Co} = b_2 M_{\rm Co}^2$  (the last two relations are purely formal). The upper sign in formula (10) corresponds to the saturation field, the lower to the critical field for transition from collinear ferrimagnetic phase to a high-field noncollinear phase (the latter, on further increase of the field, transforms to a collinear ferromagnetic phase). In contrast to the case treated earlier,<sup>9</sup> where anisotropy of only one sublattice was taken into account, formulas (10) and (11) describe the more general case of a ferrimagnet each sublattice of which possesses its own anisotropy.

It is evident from formula (10) that the saturation field and the critical field have meaning only when the radicand is nonnegative. In the contrary case, no collinear ferrimagnetic structure occurs, and the lowfield noncollinear structure, on increase of the field, transforms directly to the high-field noncollinear structure. From the condition for vanishing of the radicand in (10), it is easy to find relations for the critical coordinates  $H^*, T^*$  on the phase diagram, at which the collinear ferrimagnetic phase degenerates to a point:

$$H^{*} = Q(T^{*}),$$
  
2|b<sub>1</sub>M<sub>co</sub>-aM<sub>R</sub>(T<sup>\*</sup>)|=A((M<sub>co</sub>)<sup>V<sub>1</sub></sup>-(M<sub>R</sub>(T<sup>\*</sup>))<sup>V<sub>1</sub></sup>)<sup>2</sup>, (12)

here  $M_{\rm R} = M_{\rm ORH}$  or  $M_{\rm ORH}$ , respectively, for a field parallel to the hexagonal axis or lying in the basal plane.

#### DISCUSSION OF EXPERIMENTAL RESULTS

Formulas (10) and (11) were used to plot the theoretical phase diagrams shown in Fig. 3 by solid lines. The parameters entering these equations were assigned

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as follows. The value of the molecular field acting on the Dy sublattice was determined by us earlier<sup>8</sup> from measurements of the susceptibility in the basal plane near the compensation temperature:  $H_{M} = 950 \pm 50$  kOe  $(A = 1050 \pm 50)$ . The constant *a*, which determines the anisotropy of the Dy sublattice, can be found from the relations (7) for the jump of magnetic moment or (9) for the difference of paraprocess susceptibilities, the value of a determined by both methods agreed within the limits of experimental error and was  $260 \pm 40$ . The magnetic moment of the Co sublattice in the present temperature interval was taken from the paper of Nowik and Wernick<sup>10</sup>; with allowance for the deviation from stoichiometric composition in our compound, we set  $M_{Co} = 890G$  (7.7  $\mu_B$  per formula unit). The constants  $K_{1Co}$  and  $K_{2Co}$ , which characterize the anisotropy of the Co sublattice, were used as adjustable parameters for the purpose of satisfying equations (6); this gave  $K_{1Co}$  $= 5.8 \cdot 10^7 \text{ erg/cm}^3$ ,  $K_{2Co} = 0.63 \cdot 10^7 \text{ erg/cm}^3$ , which are sufficiently close to the anisotropy constants of YCo<sub>3</sub> at room temperature:  $K_{1Co} = 5.3 \cdot 10^8 \text{ erg/cm}^3$ ,  $K_{2Co}$ ≈ 0.<sup>11</sup>

We note that without allowance for the second anisotropy constant  $K_{2Co}$ , it is impossible to satisfy equations (6), even by changing (within reasonable limits) the anisotropy constant and magnetization of the R sublattice: the temperature interval of spin reorientation in this case was found to be considerably narrower. All the other parameters were taken directly from experimental data.

It is seen from Fig. 3 that there is good agreement between the theorteical variations of the saturation fields and the experimental values [this is somewhat surprising, in view of the limited accuracy of determination of the parameters in (10) and (11)]. The theoretically determined critical temperature  $T^*$  was found to be 285 K ( $H^* = 300$  kOe); this is considerably higher than the magnetic compensation temperature in DyCo<sub>5.3</sub> (124 K).

We note that in rare-earth iron garnets the difference between the compensation temperature and the critical temperature  $T^*$  does not exceed 10-15 K (see, for example, Ref. 9). The fact that the anisotropy of each of the sublattices in  $DyCo_{5,3}$  is large and tends to orient their moments in two mutually perpendicular directions promotes the occurrence of a noncollinear magnetic structure, and a collinear ferrimagnetic structure in an external field is not observed even at temperatures greatly different from the compensation temperature. Above the spin-reorientation transition, equation (12) has no solution, the radicand in (10) is always positive, and a collinear ferrimagnetic structure exists (within a certain field interval) up to the Curie temperature. Some divergence between the experimental data and the theoretical relation at high temperatures (external field directed in the basal plane) is presumably due to our neglect of the temperature dependence of the anisotropy constants and magnetic moment of the Co sublattice.

Figure 4 shows experimental (1) and theoretical (2) field dependences of the magnetization along the hard



FIG. 4. Experimental (1) and theoretical (2) field dependences of the magnetization of  $DyCo_{5,3}$  along the hard direction: a) T = 320 K, field directed along the hexagonal axis; b) T = 400 K, field directed in the basal plane.

direction, for temperatures below (T = 320 K, field along the hexagonal axis) and above (T = 400 K, field)directed in the basal plane) the spin-reorientation transition. The theoretical relations were obtained by numerical solution of the system of equations (2)-(4)with use of the parameters determined above. It is seen that here also there is satisfactory agreement between the experimental and the calculated curves. One notices a somewhat unusual form of the field dependence of the magnetization: a sharp rise in weak fields (up to  $\sim 0.2 H_s$ ) and a smoother increase of the magnetization on further increases of the field.<sup>7)</sup> A supplementary analysis showed that such a form of the magnetization curves is due both to the occurrence of a noncollinear magnetic structure in an external field, and to the dependence of the magnetic moment of the Dy sublattice on field and direction, in particular a faster rotation of the sublattices in the initial fields. The maximum angle  $\Delta \theta$  between the sublattices, at 320 K, occurs when  $\theta_{Co} \approx 38^{\circ}$  and is 15°; this justifies the above treatment of  $\Delta \theta$  as a small parameter.

Similar peculiarities of behavior of M(H), but in weaker form, are observed also on magnetization of the crystal in the basal plane (Fig. 4b). The maximum value of the angle  $\Delta\theta$  in this case is attained when  $\theta_{Co} \approx 28^{\circ}$  and is 4.5°.

And so, on the basis of the results presented, we may conclude that the proposed phenomenological model describes well the magnetic properties of the compound  $DyCo_{5.3}$  in strong magnetic fields and may be used to describe the magnetic characteristics of other compounds of similar type. It is important that in analysis of magnetization curves of highly anisotropic manysublattice magnets, it is necessary to pay attention to the noncollinear magnetic structure that originates in an external field. An important role is also played by allowance for the dependence of the value of the magnetic moment of the R sublattice on direction, and by the paraprocess, which occurs, in contrast to an isotropic ferrimagnet,<sup>13</sup> even in the noncollinear phase.

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<sup>&</sup>lt;sup>1)</sup>Some surplus of Co atoms in the compound under investigation is caused by the fact that in practice it is not possible to obtain a single-phase composition with the stoichiometry  $DyCo_5$ ; surplus pairs (dumbells) of cobalt atoms in this compound replace atoms of the rare-earth metal.<sup>2</sup>

- <sup>2)</sup>For R = Nd, Dy, Tb; for R = Ho, Pr the easy directions at 0 K are located at an angle to the hexangonal axis.
- <sup>3)</sup>An analog of the anisotropy field in a ferrimagnet (see below).
   <sup>4)</sup>The magnetization jump in the Co sublattice has been supposed to be negligibly small.
- <sup>5)</sup>In the present case, we have used a Landau expansion of the nonequilibrium thermodynamic potential as a power series in the magnetic moment of the R sublattice,

 $\Phi = \Phi_0 + \frac{1}{2} \alpha M_R^2 + \frac{1}{4} \beta M_R^4 + \dots;$ 

 $\alpha$  and  $\beta$  are phenomenological parameters. In the paramagnetic state,  $\alpha > 0$ , and the second term of the expansion may be neglected. The remaining terms in formula (1) have an obvious physical meaning.

- <sup>6)</sup> The relation (5), without allowance for  $K_{2Co}$ , was obtained earlier.<sup>4</sup>
- <sup>7)</sup>An attempt to process the experimental curves by the Sucksmith-Thompson method,<sup>12</sup> which is based on a representation of the anisotropy energy in the form  $K_1 \sin^2 \theta + K_2 \sin^4 \theta$ and does not allow for noncollinearity of the sublattices, showed that the magnetization curves, in coordinates  $H/\sin\theta$ or  $H/\cos\theta$  vs  $\sin^2\theta$ , were clearly nonlinear. This indicates inapplicability of this method in the case of DyCo<sub>5.3</sub>.

- <sup>2</sup>W. A. J. J. Velge and K. H. J. Buschow, J. Appl. Phys. **39**, 1717 (1968).
- <sup>3</sup>A. V. Deryagin, Usp. Fiz. Nauk **120**, 393 (1976) [Sov. Phys. Usp. **19**, 909 (1976)].

- <sup>4</sup>A. S. Ermolenko, E. V. Rozenfel'd, Yu. P. Irkhin, V. V. Kelarev, A. F. Rozhda, S. K. Sidorov, A. N. Pirogov, and A. P. Vokhmyanin, Zh. Eksp. Teor. Fiz. 69, 1743 (1975) [Sov. Phys. JETP 42, 885 (1975)].
- <sup>5</sup>V. V. Druzhinin and S. P. Zapasskiĭ, Fiz. Met. Metalloved. 44, 929 (1977) [Phys. Met. Metallogr. (USSR) 44, No. 5, 24 (1977)].
- <sup>6</sup>V. V. Druzhinin, S. P. Zapasskiĭ, and V. M. Povyshev, Fiz. Tverd. Tela **19**, 159 (1977) [Sov. Phys. Solid State **19**, 90 (1977)].
- <sup>7</sup>E. R. Callen and H. B. Callen, J. Phys. Chem. Solids 16, 310 (1960).
- <sup>8</sup>A. G. Berezin, R. Z. Levitin, and Yu. F. Popov, Zh. Eksp. Teor. Fiz. **79**, 268 (1980) [Sov. Phys. JETP **52**, 135 (1980)].
- <sup>9</sup>K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, Orientatsionnye perekhody v redkozemel'nykh magnetikakh (Orientational Transitions in Rare-Earth Magnetic Materials), Nauka, 1979.
- <sup>10</sup>I. Nowik and J. H. Wernick, Phys. Rev. **140A**, 131 (1965).
- <sup>11</sup>A. V. Deryagin, A. V. Andreev, and S. M. Zadvorkin, Deposited paper, All-Union Institute of Scientific and Technical Information No. 3312-76.
- <sup>12</sup>W. Sucksmith and J. E. Thompson, Proc. Roy. Soc. (London) A 225, 362 (1954).
- <sup>13</sup>K. P. Belov, Ferrity v sil'nykh magnitnykh polyakh (Ferrites in Strong Magnetic Fields), Nauka, 1972.

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<sup>&</sup>lt;sup>1</sup>G. Dworchak and Y. Khan, J. Phys. Chem. Solids 15, 1021 (1974).