crystal has no symmetry plane parallel to the film surfaces.

We did not mention in this section the heavy holes, for which the quasiclassical quantization (1) is valid. If condition (20) is satisfied, they can be treated independently of the light holes. Allowance for the corrections relative to the parameter (20) leads to spectrum correction proportional to $M/L \ll 1$ and to an increase in the distance between the size-quantized subbands corresponding to the heavy and light holes at the points of their intersection.

Size quantization in the vicinity of the top of a degenerate valence band was considered earlier[4] with null boundary conditions used for the envelopes. This approach cannot yield Tamm states which, as shown in the present paper, alter significantly the spectrum of the size-quantized subbands in the valence band at $n_v \lesssim 1$. None the less, the positions of the extrema of the size-quantized subbands corresponding to $n_v > 1$ agree with Nedorezov's results[4] in the limit of thick films ($d \gg \pi n_v |R_v|$). A more detailed comparison of his and our results is difficult, since Nedorezov considers the opposite limiting case, where the spin-orbit interaction is assumed large and the mass of the heavy holes is assumed finite.

We note in conclusion that the boundary condition (10) retains the same form in those cases when additional fields that can be described in the language of envelopes are present in the system.

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Level crossing and instability of magnetic structure in rare-earth iron garnets

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When the ground-state levels of a rare-earth ion cross (or approach one another) in a rare-earth iron garnet, the magnetic structure of the garnet becomes unstable. This phenomenon is very close to the well known Jahn-Teller effect. If the rare-earth ions have a strong magnetic-moment anisotropy, this instability involves some distinctive anomalies of the magnetic behavior of the crystal. We have investigated a theoretical model in which the rare-earth ions are treated in an extreme anisotropic (Ising) approximation. It is shown that at different orientations of the external magnetic field the instability produced by the level crossing has a fine structure that reflects the detailed character of the magnetization reversal of rare-earth ions situated in differnt non-equivalent positions. The magnetization curves of such a system are investigated and a comparison is made with the experimental data on holmium-yttrium ion garnets.

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1. INTRODUCTION

Demidov, Levitin, and Popov[1,2] have observed an interesting phenomenon: in some mixed rare-earth iron garnets (REIG), the magnetization curves $M(H)$ exhibit magnetization jumps in strong fields at low temperatures. These jumps arise at different orientations of the external field relative to the crystal axes and are accompanied by hysteresis phenomena. The probable cause of these anomalies, in their opinion, is the crossing (or approach) of the low-lying levels of the rare-earth ions (REI) when the external magnetic field H is

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varied. We have shown in[3] that the crossing of the levels of an ion in a magnetic crystal is accompanied by an instability of the magnetic structure of the crystal, which leads to a lowering of its magnetic symmetry (the magnetic analog of the Jahn–Teller effect). The cause of the instability can be explained in the following manner. We divide the magnetic crystal into two interacting subsystems: a) magnetic ions, whose lower levels can cross; b) all the remaining magnetic ions, which we shall call the matrix. In the case of REIG, the role of the subsystem a) is played by the REI and the role of the matrix by the Fe^{3+} ions. Assume that in the initial symmetrical state ($\mathbf{m} \parallel \mathbf{H}$, where $\mathbf{m}$ is the magnetic moment of the matrix) the two lower levels of the REI intersect in a certain external magnetic field. The deformation of the magnetic structure, which leads to a lowering of the symmetry, will be characterized by the inclination θ of the magnetic field of the matrix to the equilibrium position in the initial state. This deformation causes a level splitting that is linear in θ ($\Delta E = \pm a\theta$, where a is the constant of the interaction of the ion with the matrix). The ground state energy of the ion is then lowered. At the same time, the deformation of the structure results in an energy loss $b\theta^2/2 > 0$ (b is the "rigidity" of the matrix, including the energy of its interaction with the magnetic field mH and its magnetic-anisotropy energy). The presence of terms linear in θ in the energy leads to instability of the system; the quadratic terms limit its development. The structure produced as a result of the instability is determined by the condition that the total energy be a minimum (at $T = 0$ °K):

$$E = {}^1/_2 b\theta^2 - a|\theta|. \qquad (1)$$

The minimum is reached at $\theta = \pm a/b$.

At large REI concentrations, a cooperative phase transition takes place. If the interaction constant a is large enough or the "rigidity" b is small enough, the foregoing arguments are valid also for the case when the levels approach each other. If it is assumed that the magnetization anomalies observed in[1,2] are actually due to crossing (or approach) of levels, then the onset of instability of the magnetic structure is attested here by the abruptness of the jumps and, in particular, by the presence of hysteresis. In[3] we investigated a simplified model that did not reflect certain important features of rare-earth iron garnets. In the present paper we see a more realistic model, which takes into account the actual symmetry of the REIG, and considers in greater detail than in [3] the properties of the REI.

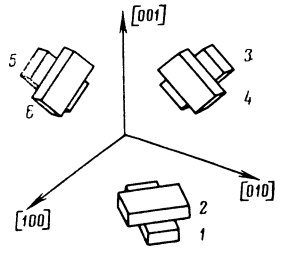

FIG. 1. Orientation of local axes of rare-earth ions and rare-earth iron garnets.

TABLE I. Orientation of rare-earth ion axes relative to the crystallographic coordinate frame.

Local axes	Non-equivalent points					
	1	2	3	4	5	6
z	[001]	[001]	[100]	[100]	[010]	[010]
y	[110]	[110]	[011]	[011]	[101]	[101]
x	[110]	[110]	[011]	[011]	[101]	[101]

2. MODEL SPIN HAMILTONIAN OF REI

The rare-earth ions occupy in the REIG c-positions whose crystal-environment symmetry is determined by the point group D_2 (222). There are six such non-equivalent positions that differ in the orientation of the symmetry axis of the C_2. Figure 1 shows the orientation of the axis for all six points (see also Table I).

The crystal field in the REIG splits the ground-state multiplet of the REI into doublets in the case of Kramers ions (i.e., ions with odd number of f electrons), and into singlets in the case of non-Kramers ions. Sometimes (as, e.g., in the case of Tb^{3+} and Ho^{3+}), the very lowest levels in the REI spectrum are two (or three) closely-lying singlets, which are separated distinctly enough from the higher-lying levels[4] (see also Fig. 1 of[2]). In this case one can speak of a quasi-doublet (quasi-triplet) and describe the behavior of its levels in the magnetic field in the same manner as in the case of Kramers ions, with the aid of a g-tensor, which is strongly anisotropic here.

The dependence of the energy levels of the lower doublet (quasi-doublet, quasi-triplet) on the magnetic field in REIG is determined by the joint action of the external field and of the exchange field produced at the REI by the Fe^{3+} ions. We shall consider systems in which these fields are oppositely directed (at least at certain definite directions of the external field relative to the crystal axes; in the general case the angle between these fields is larger than $\pi/2$). This situation is realized in systems of the type $R_xY_{3-x}IG$, where R is the rare-earth element of the yttrium subgroup of the lanthanide group (from gadolinium to ytterbium) at a sufficiently low concentration x. The competition between the external and exchange fields in such systems afford a natural possibility of level crossing (in the case of Kramers ions) or approach (in the case of non-Kramers ions). If the external field ($\mathbf{H}$) is antiparallel to the exchange field ($\mathbf{H}^{ex}$), the crossing (approach) takes place at $|\mathbf{H}| = |\mathbf{H}^{ex}|$, i.e., when the REI is fully demagnetized.

According to the statement made in the Introduction, the level crossing is energywise not favored and the system, without reaching the crossing point, goes over into the canted phase and remains in it with increasing field until it leaves the "dangerous" region near the crossing point. In the canted phase, the REI energy levels are "pushed apart," inasmuch as $\mathbf{H} + \mathbf{H}^{ex} \neq 0$ in this phase. The picture is particularly obvious in the case of REI with an isotropic g-factor (Gd^{3+}) and manifests itself macroscopically as a well known phenomenon—the flipping of the sublattices of an isotropic ferrimagnet in a

magnetic field (see, e.g., [5]). In the case of strongly anisotropic REI, this practice has a number of interesting features that differ qualitatively from the isotropic case. To explain them, we consider a model in which the REI are described in an extreme anisotropic—Ising—approximation, i.e., we assume that regardless of the direction of the fields acting in such an ion, its magnetic moment, while changing in magnitude, preserves its direction. The closest prototypes of such a model are the ions Ho^{3+}, Tb^{3+}, Dy^{3+}.

Let the direction of the magnetization of the "Ising" ion in position i coincide with the axis z_i of the local coordinate system, i.e., let the only nonzero component of the g tensor in this system of coordinates be g_{zz}. We note that this choice of the ion-magnetization axis, whereby the easy-magnetization axis of the crystal lies along directions of the type [111], is the only one (this is realized also in[2]). We note also that with this choice of the axis of the "Ising" ion the positions 1 and 2, 3 and 4, and 5 and 6 become pairwise equivalent (see Fig. 1).

Under the foregoing assumptions, the spin-Hamiltonian of an ion situated in the position i in an exchange field and an external field can be represented in the local coordinate system in the form

$$\mathcal{H}=-2\mu(H_{z_i}+H^{ex}_{z_i})S_z^{eff}, \tag{2}$$

where μ is the magnetic moment of the ion and $S^{eff}=\frac{1}{2}$. We have confined ourselves to the case $S^{eff}=\frac{1}{2}$, since the qualitative conclusions (also quantitative at $T=0$ °K) do not depend on the multiplicity. We note also that we confine ourselves here to an isotropic exchange field (i.e., we assume that $-\mathbf{H}^{ex}\parallel\mathbf{m}_{Fe}$). The energy levels defined by the Hamiltonian (2) are

$$\mathcal{E}_{1,2}=\pm\mu|H_{z_i}+H^{ex}_{z_i}|. \tag{3}$$

3. MAGNETIC TRANSITIONS AT $T=0$ °K

At $T=0$ °K, the equilibrium states of the crystal are determined by minimizing its ground-state energy. We represent it in the form (per REIG molecule)

$$\mathcal{E}=-\frac{x\mu}{6}\sum_{i=1}^{6}|H_{z_i}+H^{ex}_{z_i}|-\mathbf{mH}, \tag{4}$$

where $\mathbf{m}$ is the magnetic moment of the iron sublattice ($|\mathbf{m}|=5\ \mu_B$ at $T=0$ °K), x is the REI concentration in the molecule $R_xY_{3-x}Fe_5O_{12}$. We assume $x\ll 1$ throughout.[1)] Since $-\mathbf{H}^{ex}\parallel\mathbf{m}$, it follows that $\mathcal{E}$ depends on the orientation of $\mathbf{m}$ relative to the crystal axes. The angles that specify this orientation must be determined as functions of the external field by minimizing $\mathcal{E}$. The minimization of (4) with the additional condition $|\mathbf{m}|^2=\text{const}$ determines the equation

$$\mathbf{H}+\sum_{i=1}^{6}\mathbf{H}_i^R=c\mathbf{m}, \tag{5}$$

where

$$\mathbf{H}_i^R=-\frac{x}{6}\lambda\mu_i z_i,\quad \mu_i=\mu\,\mathrm{sign}\,(H_{z_i}+H^{ex}_{z_i}), \tag{6}$$

$\mathbf{H}_i^R$ is the exchange field exerted on $\mathbf{m}$ by the REI located in position i and having a magnetic moment μ_i; λ is the exchange constant of the R–Fe interaction ($\lambda\equiv H^{ex}/m$). The left-hand side of (5) is the sum of the fields acting on $\mathbf{m}$, and c is an indeterminate Lagrange multiplier defined by the condition $|\mathbf{m}|=5\mu_B$. Generally speaking, (5) is an equation for $\mathbf{m}$, since the direction of $\mathbf{H}_i^R$, according to (6), depends on the orientation of the vector $\mathbf{m}$. From (5) it follows, in particular, that at $\mathbf{H}=0$ the diagonals of the cubic unit cell ([111] etc.) are easy axes. Indeed, the resultant of all the fields $\mathbf{H}_i^R$ along the edges of the cube is directed along one of its diagonals. The eight equivalent positions of $\mathbf{m}$ (at $\mathbf{H}=0$) can be obtained by going through all the combinations of the signs of μ_i.

At $\mathbf{H}\neq 0$ it is more convenient to investigate the behavior of $\mathbf{m}(\mathbf{H})$ in a spherical coordinate system. We consider the cases normally encountered in the experiments: $\mathbf{H}\parallel[001]$, $\mathbf{H}\parallel[110]$, $\mathbf{H}\parallel[111]$.

A. Case $\mathbf{H}\parallel[001]$. We define $\mathbf{m}$ in a coordinate system with axes [100], [010], [001] with the aid of the customarily introduced polar angle θ and azimuthal angle φ. In this coordinate system, the components $H_{zi}+H^{ex}_{zi}$ are for positions 1 and 2

$$H-H^{ex}\cos\theta, \tag{7a}$$

for positions 3 and 4

$$-H^{ex}\sin\theta\cos\varphi, \tag{7b}$$

and for positions 5 and 6

$$-H^{ex}\sin\theta\sin\varphi. \tag{7c}$$

Substituting them in (4) and introducing the dimensionless variables $E=\mathcal{E}/mH^{ex}$, $h=H/H^{ex}$, and $a=x\mu/3m\ll 1$, we obtain

$$E(\theta,\ \varphi)=-h\cos\theta-a|h-\cos\theta|-a\sin\theta(|\sin\varphi|+|\cos\varphi|). \tag{8}$$

Minimizing (8) with respect to φ we find that if $\theta\neq 0$ we have $\varphi=\frac{1}{4}\pi(2k+1)$, where $k=0,1,2,3$, corresponding to the crystal planes $(1\bar{1}0)$, $(\bar{1}10)$, $(\bar{1}\bar{1}0)$, (110). All these planes are energywise equivalent, this being the consequence of the symmetry: [001] is a fourfold axis. The presence of such a "degeneracy" can lead physically to a domain structure. Since such domains are energywise equivalent, they exist also in strong fields (so long as $\theta\neq 0$).

Minimizing (8) with respect to θ (at $\varphi=\frac{1}{4}\pi(2k+1)$), we obtain

$$\mathrm{tg}\,\theta=\frac{a\sqrt{2}}{h-a\,\mathrm{sign}\,(h-\cos\theta)}. \tag{9}$$

We note that according to (7a) we have $\mathrm{sign}(h-\cos\theta)=\mathrm{sign}\,\mu_i$.

As noted earlier, at $H=0$ there are eight equivalent directions of the vector $\mathbf{m}$ in the crystal. There can be

just as many different domains. In a relatively weak field, domains with "unfavorable" direction of the magnetic moments vanish. We shall disregard from now on the solutions corresponding to such domains. In this case these are four domains having $\tan\theta = -\sqrt{2}$ at $h=0$. Equation (9) has at small a the solution

$$\operatorname{tg}\theta = \begin{cases} \dfrac{a\sqrt{2}}{h+a}, & h<h_2=1-\dfrac{a^2}{(1+a)^2} \quad (\mu_1<0) \\ \dfrac{a\sqrt{2}}{h-a} & h>h_1=1-\dfrac{a^2}{(1-a)^2} \quad (\mu_1>0) \end{cases} \tag{10}$$

The overlap of the solutions in the region $h_1<h<h_2$ and the presence of hysteresis can be interpreted as a first-order phase transition between the phases ($\mu_1<0$) and $B(\mu_1>0)$. The transition point is determined by the condition that these phases have the same energy, equal to $h_I = 1-a^2$.

Knowing $\theta(h)$, we can determine the behavior of the magnetization of the crystal from the formula

$$M=-\partial\mathscr{E}/\partial H=-m\partial E/\partial h. \tag{11}$$

Differentiating (8) with respect to h and substituting in it $\theta(h)$ defined by formula (10), we obtain (Fig. 2a)

$$M=m[\cos\theta+a\operatorname{sign}\mu_1]=\begin{cases} m\dfrac{h+a}{[(h+a)^2+2a^2]^{1/2}}-am, & h<h_I \\ m\dfrac{h-a}{[(h-a)^2+2a^2]^{1/2}}+am, & h>h_I \end{cases} \tag{12}$$

Those branches of $M(H)$ which correspond to metastable states of the crystal near $H=H^{ex}$ will be omitted henceforth.

The results can be interpreted in the following (Fig. 3a). Positions 1–6 at $\mathbf{H}\parallel[001]$ and when $\mathbf{m}$ is rotated in the indicated planes can be divided into two groups: 1, 2 and 3–6. The ions in positions 3–6 are not acted upon by the external magnetic field; their levels are split only by $\mathbf{H}^{ex}$. The splitting is maximal when $\mathbf{H}^{ex}$ (and $\mathbf{m}$) lie in the (001) plane, and is equal to zero when $\mathbf{m}\parallel\mathbf{H}$ (see (8)). This means that these ions produce an anisotropy energy that hinders the rotation of $\mathbf{m}$ towards the [001] axis. The ions of positions 1 and 2, in turn, produce a field $\mathbf{H}^R$ that acts on $\mathbf{m}$ and is parallel to $\mathbf{H}$ in the phase A ($\mu_1<0$) and is antiparallel to $\mathbf{H}$ in the phase B ($\mu_1>0$). Thus, rotation of m towards H takes place under the influence of the field $h+a$ in phase A and under

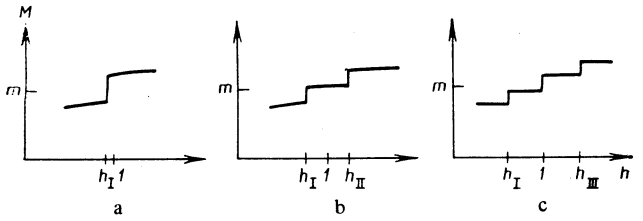

FIG. 2. Dependence of the magnetization $M(h)$ on the external field at $T=0$ K with allowance for the rearrangement of the magnetic structure (the possible hysteresis near the phase-transition points is not shown): a) $\mathbf{H}\parallel[001]$, b) $\mathbf{H}\parallel[110]$, c) $\mathbf{H}\parallel[111]$.

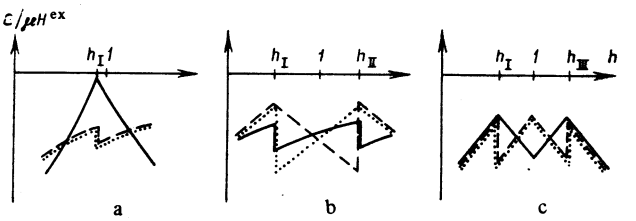

FIG. 3. Dependence, on the magnetic field h, of the energy of the ground state of the rare-earth ions in various non-equivalent points, with allowance for the rearrangement of the magnetic structure (the line $\mathscr{E}=0$ is the center of gravity of the quasi-doublet, the hysteresis is not shown, $T=0$ K): a) $\mathbf{H}\parallel[100]$, b) $\mathbf{H}\parallel[110]$, c) $\mathbf{H}\parallel[111]$; solid line—points 1 and 2; dashed—point 3; dotted—points 5 and 6.

the influence of $h-a$ in phase B, producing somewhat different $\theta(h)$ dependences in these phases and leading to a hysteresis of $\theta(h)$. We note that in this case the splitting of the levels 1 and 2 depends quadratically on θ (like $|\cos\theta-h|$), and therefore the hysteresis is narrow at small a.

The jump of the magnetization at the level crossing point ($|\cos\theta-h|=0$) is equal to $\Delta M=2ma=2x\mu/3$ and has an obvious meaning: it is the result of the reversal of magnetizations of ions 1 and 2, the concentration of which is equal to $x/3$.

B. Case $\mathbf{H}\parallel[110]$. In the coordinate system with axes $[1\bar{1}0]$, $[00\bar{1}]$, $[110]$, where the polar angle θ and the azimuthal angle φ define $\mathbf{m}$, the projections of $H_{zi}+H^{ex}_{zi}$ for positions 1 and 2 are given by

$$-H^{ex}\sin\theta\sin\varphi, \tag{13a}$$

and for positions 3 and 4 by

$$2^{-1/2}(H-H^{ex}\cos\theta+H^{ex}\sin\theta\cos\varphi), \tag{13b}$$

and for positions 5 and 6 by

$$2^{-1/2}(H-H^{ex}\cos\theta-H^{ex}\sin\theta\cos\varphi). \tag{13c}$$

Substituting them in the energy (4) we obtain in the same dimensionless variables as in (8)

$$E(\theta,\varphi)=-a\sin\theta|\sin\varphi|-2^{-1/2}a(|h-\cos\theta+\sin\theta\cos\varphi| +|h-\cos\theta-\sin\theta\cos\varphi|)-h\cos\theta. \tag{14}$$

Minimization of (14) with respect to θ and φ yields

$$\operatorname{ctg}\varphi=\frac{1}{\sqrt{2}}\frac{\operatorname{sign}\mu_3-\operatorname{sign}\mu_5}{\operatorname{sign}(\sin\varphi)}, \tag{15}$$

$$\operatorname{tg}\theta=\frac{a|\sin\varphi|+a\cos\varphi(\operatorname{sign}\mu_3-\operatorname{sign}\mu_5)}{h-2^{-1/2}a(\operatorname{sign}\mu_3+\operatorname{sign}\mu_5)} \tag{16}$$

where the signs of the magnetic moments μ_3 and μ_5 coincide with the signs of the fields (13b) and (13c) acting on them.

Equation (15) has the following stable solutions:

$$\varphi=\begin{cases} \varphi_1=\dfrac{\pi}{2}, \quad \varphi_2=\dfrac{3\pi}{2}; \quad \operatorname{sign}\mu_3=\operatorname{sign}\mu_5 \\ \varphi_3, \quad \varphi_4=-\varphi_3; \quad \mu_3>0, \quad \mu_5<0, \\ \varphi_5=\pi-\varphi_3, \quad \varphi_6=\pi+\varphi_3; \quad \mu_3<0, \quad \mu_5>0 \end{cases} \tag{17}$$

where $\sin\varphi_3=\sqrt{\frac{1}{3}}$ and $\cos\varphi_3=\sqrt{\frac{2}{3}}$. Substituting (17) in (16) we obtain $\theta(h)$:

$$\operatorname{tg}\theta=\begin{cases}\dfrac{a\sqrt{3}}{h} & (\varphi=\varphi_3,\varphi_4,\varphi_5,\varphi_6),\quad \operatorname{sign}\mu_3\neq\operatorname{sign}\mu_5, \qquad (18a)\\ \dfrac{a}{h+a\sqrt{2}} & (\varphi=\varphi_1,\varphi_2),\quad \mu_3<0,\quad \mu_5<0, \qquad (18b)\\ \dfrac{a}{h-a\sqrt{2}} & (\varphi=\varphi_1,\varphi_2),\quad \mu_3>0,\quad \mu_5>0. \qquad (18c)\end{cases}$$

Thus, three phases exist: A ($\mu_3<0$, $\mu_5<0$), B ($\mu_3>0$, $\mu_5>0$), and $C(\operatorname{sign}\mu_3=-\operatorname{sign}\mu_5)$. The regions of their existence (stability) are determined in the following manner. For example, for phase A the condition $\mu_3<0$ means that $h-\cos\theta(h)+\sqrt{\frac{2}{3}}\sin\theta(h)<0$, where $\theta(h)$ is given by (18b). Solving the corresponding transcendental equations for the critical fields, we obtain the following conditions for the existence of the phases ($a\ll1$): phase A exists at

$$h<h_3=1-a^2/(1+a\sqrt{2})^2,$$

phase B exists at

$$h>h_4=1-a^2/(1-a\sqrt{2})^2;$$

and phase C exists in the region

$$h_1=1-a\sqrt{2}-3a^2/2<h<1+a\sqrt{2}-3a^2/2=h_2.$$

Phases A and B overlap little ($h_4<h_3$). Phase C overlaps both phases A and B in the region near $h=1$. This means that first-order transitions take place between phases A, C, and B; the transition points are determined by equating the energies of the corresponding phases. The $A\rightarrow C$ transition takes place at $h_{\mathrm{I}}=1-a\sqrt{2}/2$ and the $C\rightarrow B$ transition at $h_{\mathrm{II}}=1+a\sqrt{2}/2$.

We note also that the phase A and B are doubly "degenerate" in φ ([110] is a twofold axis), so that domains with different directions of $\mathbf{m}$ can exist in the crystal. On going from these phases into the phase C, the degeneracy doubles; each of the domains of phase A or B is "split," thus, for example,

$$\varphi_1=\pi/2\rightarrow\varphi_3,\varphi_5,\ \text{and}\ \varphi_2=3\pi/2\rightarrow\varphi_4,\varphi_6.$$

The magnetization $M(H)$, determined by formula (11) takes the form (Fig. 2b)

$$M=m[\cos\theta+2^{-1/2}a(\operatorname{sign}\mu_3+\operatorname{sign}\mu_5)]\cdot$$
$$=\begin{cases}m\dfrac{h+a\sqrt{2}}{[(h+a\sqrt{2})^2+a^2]^{1/2}}-ma\sqrt{2}, & h<h_{\mathrm{I}}\\ m\dfrac{h}{(h^2+3a^2)^{1/2}}, & h_{\mathrm{I}}<h<h_{\mathrm{II}}\\ m\dfrac{h-a\sqrt{2}}{[(h-a\sqrt{2})^2+a^2]^{1/2}}+ma\sqrt{2}, & h>h_{\mathrm{II}}\end{cases} \qquad (19)$$

The results can be interpreted in the following manner (Fig. 3b). The ions in positions 1 and 2 add the maximum level splitting according to (13) and (14) at $\theta=\pi/2$, and zero splitting at $\theta=0$; they produce an anisotropy energy that prevents the rotation of $\mathbf{m}$ towards $\mathbf{H}$. The ions in positions 3–6 produce a field acting on $\mathbf{m}$; it is parallel to $\mathbf{H}$ at $\mu_3<0$ and $\mu_5<0$, and antiparallel in the opposite case. This causes $\theta(h)$ to be somewhat different in phases A and B and to overlap. In contrast to the case $\mathbf{H}\parallel[001]$, in this situation the splitting of the levels of ions in positions 3–6 depends linearly on the angle θ if $\varphi\neq\pi/2$ and $\varphi\neq3\pi/2$. This creates conditions for an "azimuthal" instability—the deflection of $\mathbf{m}$ in other planes near the level crossing point $h=1$. In the $A\rightarrow C$ transitions, the magnetizations of ions 3 and 4 (or 5 and 6) are reversed,[2)] and the levels of ions 5 and 6 (3 and 4) are "pushed apart." In phase C, the levels of the ions with reversed magnetization (3, 4) move apart with increasing h, and those of the ions without reversal (5, 6) come closer until their relative position becomes energywise unfavorable. At $h=h_{\mathrm{II}}$ ($C\rightarrow B$), the magnetization of ions 5 and 6 is also reversed and the system returns to the initial plane. The magnetization jumps $\Delta M=2^{1/2}ma=2x\mu/3\sqrt{2}$ have an obvious physical meaning and $\mu/\sqrt{2}$ is the projection of the magnetic moment of the "Ising" ions of type 3 and 5 on the [110] axis.

C. Case $\mathbf{H}\parallel[111]$. We define $\mathbf{m}$ in a coordinate system whose axes coincide with the directions $[1\bar{2}1]$, $[10\bar{1}]$, [111], with the aid of the polar angle θ, and the azimuthal angle φ. In this system

$$H_{z_i}+H^{\mathrm{ex}}_{z_i}=\frac{1}{\sqrt{3}}\left\{H-H^{\mathrm{ex}}\left[-\sqrt{2}\sin\theta\cos\left(\varphi+\frac{2\pi n}{3}\right)+\cos\theta\right]\right\}, \qquad (20)$$

where $n=0,1,-1$ at $i=$ respectively to 5, 6; 3, 4; 1, 2. Substituting them in (4) and using the same dimensionless variables as in (8), we get

$$E(\theta,\varphi)=-\frac{a}{\sqrt{3}}\sum_{n=0,\pm1}\left|h-\cos\theta+\sqrt{2}\sin\theta\cos\left(\varphi+\frac{2\pi n}{3}\right)\right|-h\cos\theta. \qquad (21)$$

The stationarity conditions $\partial E/\partial\theta=\partial E/\partial\varphi=0$ yield

$$\operatorname{tg}\theta=a\left(\frac{2}{3}\right)^{1/2}\frac{\cos\varphi\operatorname{sign}\mu_3+\cos(\varphi+2\pi/3)\operatorname{sign}\mu_3+\cos(\varphi-2\pi/3)\operatorname{sign}\mu_1}{h-3^{-1/2}a(\operatorname{sign}\mu_1+\operatorname{sign}\mu_3+\operatorname{sign}\mu_5)} \qquad (22)$$

$$\sin\theta[\sin\varphi\operatorname{sign}\mu_3+\sin(\varphi+2\pi/3)\operatorname{sign}\mu_3+\sin(\varphi-2\pi/3)\operatorname{sign}\mu_1]=0, \qquad (23)$$

where the signs of μ_1, μ_3, μ_5 coincide with the signs of the fields (20) acting on them.

Depending on the conbination of the signs of μ_1, μ_3, μ_5, the following solutions (phases) of Eqs. (22) and (23) are possible:

phase A ($\mu_1<0$, $\mu_3<0$, $\mu_5<0$):
$$\theta=0; \qquad (24a)$$

phase B ($\operatorname{sign}\mu_1+\operatorname{sign}\mu_3+\operatorname{sign}\mu_5=-1$):
$$\varphi=0\ (\mu_5>0,\ \mu_3<0,\ \mu_1<0),\quad \varphi=2\pi/3\ (\mu_5<0,\ \mu_3<0,\ \mu_1>0),\quad \varphi=4\pi/3\ (\mu_5<0,\ \mu_3>0,\ \mu_1<0); \qquad (24b)$$

phase C ($\operatorname{sign}\mu_1+\operatorname{sign}\mu_3+\operatorname{sign}\mu_5=1$):
$$\varphi=\pi\ (\mu_5<0,\ \mu_3>0,\ \mu_1>0),\quad \varphi=5\pi/3\ (\mu_5>0,\ \mu_3>0,\ \mu_1<0),\quad \varphi=\pi/3\ (\mu_5>0,\ \mu_3<0,\ \mu_1>0); \qquad (24c)$$

phase D ($\mu_1>0$, $\mu_3>0$, $\mu_5>0$):
$$\theta=0. \qquad (24d)$$

The threefold degeneracy of the phases B and C is determined by the "symmetry": [111] is a threefold axis.

Substituting the values of the angles φ in (22), we obtain

$$\operatorname{tg}\theta=\begin{cases}0 & (\text{phase } A,\ \text{phase } D),\\ 2\sqrt{2/3}\,a/(h+a/\sqrt{3}) & (\text{phase } B),\\ 2\sqrt{2/3}\,a/(h-a/\sqrt{3}) & (\text{phase } C).\end{cases} \tag{25}$$

We note that any solution of the phase B goes over into the corresponding solution of phase C by the transformations $\varphi\to\varphi+\pi$ and $\mu_i\to-\mu_i$. This means that in both phases the deflection of **m** takes place in identical planes, but in different directions away from the [111] axis. Thus, for example, when considering the rotation of **m** in the (110) plane, we see that the phase B corresponds to a deflection of **m** towards the [110] axis and the phase C corresponds to a deflection towards the [001] axis. In each of the planes $(01\bar1)$ ($\varphi=0,\pi$), $(10\bar1)$ ($\varphi=2\pi/3, 5\pi/3$) and $(01\bar1)$ ($\varphi=4\pi/3, \pi/3$) it is possible to define the deflection of **m** with the aid of a single angle θ', which assumes positive and negative values. The transition $B\rightleftharpoons C$ means reversal of the sign of θ' and a corresponding reversal of the signs of all the magnetic moments μ_i.

The regions of the existence (stability) of the phases A, B, C, and D can be determined in the same manner as for the case $\mathbf{H}\parallel[110]$ above: the phase A exists at

$$h<1,$$

the phase B exists in the region

$$1-4a/\sqrt{3}<h<1+2a/\sqrt{3},$$

the phase C in the region

$$1-2a/\sqrt{3}<h<1+4a/\sqrt{3},$$

and the phase D at

$$h>1.$$

Phase overlap means that the transitions between them are of first order. Their sequence with increasing field is $A\to B\to C\to D$. The transition points are obtained from the conditions that the corresponding energies be equal and are given by a) $h_{\mathrm I}=1-2a/\sqrt{3}$ (for $A\rightleftharpoons B$), b) $h_{\mathrm{II}}=1$ (for $B\rightleftharpoons C$), and c) $h_{\mathrm{III}}=1+2a/\sqrt{3}$ (for $C\rightleftharpoons D$).

The magnetization, defined by formula (12), is given by (see Fig. 2c)

$$M=m[1+3^{-1/2}a(\operatorname{sign}\mu_1+\operatorname{sign}\mu_3+\operatorname{sign}\mu_5)]=\begin{cases}m(1-a\sqrt{3}), & h<h_{\mathrm I}\\ m(1-a/\sqrt{3}), & h_{\mathrm I}<h<1\\ m(1+a/\sqrt{3}), & 1<h<h_{\mathrm{III}}\\ m(1+a\sqrt{3}), & h>h_{\mathrm{III}}\end{cases} \tag{26}$$

The detailed picture of the transitions and of the successive magnetization reversal of the IEI takes an interesting form in this case. With increasing field ($h\to1$) the levels of all ions 1–6 in phase A come closer together (like $|1-h|$ at $\theta=0$ according to (3), (20), and Fig. 3c). At $h\sim h_{\mathrm I}$ they come so close together that the deflection of **m** (say in the $(1\bar10)$ plane) towards the [110] axis becomes energywise favored. Following this deflection, the levels of the ions 3–6 are "pushed apart," but their magnetization is still not reversed ($\mu_{3-6}<0$), while the magnetization of ions 1 and 2 is reversed ($\mu_{1,2}>0$). This fact can be understood purely geometrically. As a result of the deflection of **m** towards the (001) plane, the projection of $\mathbf{H}^{ex}$ on the magnetization axis of ions 1 and 2 decreases, so that the resultant field acting on these ions becomes positive and their magnetization is reversed; for the ions 3–6 the situation is reversed: the projection of $\mathbf{H}^{ex}$ on their axis increases, the absolute value of the resultant field decreases, and consequently the level splitting also increases. With further increase of the field, $h\to1$, the levels of the "non-remagnetized ions" continue to approach each other, and a different transformation of the magnetic structure becomes energywise profitable at $h=1$, namely the deflection of **m** in the same plane, but in the opposite direction—towards the [001] axis. The projection of $\mathbf{H}^{ex}$ on the axis of the ions 3–6 then decreases abruptly and their magnetization is reversed along the external field, while the projection of $\mathbf{H}^{ex}$ on the axis of the ions 1 and 2 increases and they are forced to reverse their magnetization. We note that the states in which all $\mu_i>0$ are here patently energywise unprofitable, inasmuch as at $\mu_i>0$ the only stable state is $\theta=0$, and at $\theta=0$ and $h\sim1$ the levels of all the ions come closest to one another. With further increase of the field, at $h=h_{\mathrm{III}}$, the deformed (canted) structure becomes unfavorable, the magnetization **m** returns to the [111] axis, and the ions 1 and 2 have their magnetization reversed along the external field. Each jump of magnetization is equal to $\Delta M=2x\mu/3\sqrt{3}$, where $\mu/\sqrt{3}$ is the projection of the magnetic moment of each "Ising" ion on the [111] axis. This is the magnetization-reversal picture when the deflection of **m** is in the $(1\bar10)$ plane, as is realized in one of the domains. In the other domains (where **m** is deflected in other planes), the sequence of the magnetization reversal of the REI will be different. The role of the magnetic-structure instability mechanism near the level crossing, referred to in the Introduction, can be clearly traced in the cases $\mathbf{H}\parallel[110]$ and $\mathbf{H}\parallel[111]$.

4. *H-T* PHASE DIAGRAMS

The thermodynamic potential of the system can be represented in the quasidoublet approximation in the form

$$\Phi=-\mathbf{mH}-\frac{Tx}{6}\sum_{i=1}^{6}\ln\left[2\operatorname{ch}\frac{\mu(H_{z_i}+H_{z_i}^{ex})}{T}\right], \tag{27}$$

where $H_{z_i}+H_{z_i}^{ex}$ are determined by formulas (7), (17), and (20), respectively for $\mathbf{H}\parallel[100]$, $\mathbf{H}\parallel[110]$, and $\mathbf{H}\parallel[111]$.

We consider first the case $\mathbf{H}\parallel[111]$. Assuming a concentration $x\ll1$, we confine ourselves to the behavior of the system at small angles θ and fields close to $\mathbf{H}^{ex}$. It is convenient to introduce the dimensionless variables

$$f=\frac{\Phi}{mH^{ex}\theta_0^2},\ \tau=\frac{3}{8}\frac{T}{\mu H^{ex}a},\ W=\frac{\sqrt{3}(1-h)}{4a},\ \Theta=\frac{\theta}{\theta_0},\ \theta_0=\left(\frac{8}{3}\right)^{1/2}a; \tag{28}$$

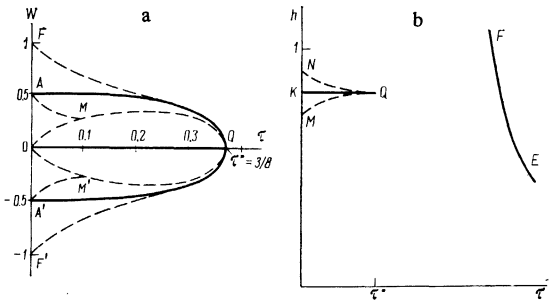

FIG. 4. $H-T$ phase diagrams in relative coordinates: a) $\mathbf{H} \parallel [111]$, b) $\mathbf{H} \parallel [100]$.

Then, omitting the terms independent of Θ, we find that f takes the form

$$f = \frac{\Theta^2}{2} - \tau \sum_{n=0,\pm 1} \ln \left[\mathrm{ch} \frac{W - \Theta \cos(\varphi + 2\pi n/3)}{2\tau} \right]. \qquad (29)$$

We note that the dependence on the physical parameters x, H^{ex}, m, and μ enters in this expression only via the scales of the quantities θ, H, and T, so that we can confine ourselves to citing the results of an investigation of the phase diagram in numerical form. The corresponding analytic expressions are cumbersome and not illustrative. The equilibrium values of the angle φ are here the same as at $T = 0$ °K, and we shall therefore assume, as before, that the angle Θ take on positive and negative values in the "equilibrium" planes $(1\bar{1}0)$, $(10\bar{1})$, $(01\bar{1})$, and set φ in (29) equal to zero.

The results of the former problem of minimization and numerical calculations reduce to the following (Fig. 4a).

1. The collinear phase $\Theta = 0$ (the phases A and D) is absolutely stable (in equilibrium) in the region outside the curve AQA'. It is metastable in the regions OQA and OQA'.

2. The canted phase B $(\Theta_B(W, \tau))$, defined by the equation

$$W = \Theta/4 \pm \tau \operatorname{Ar\,ch} [\Theta^{-1} \operatorname{sh}(3\Theta/4\tau) - \operatorname{ch}(3\Theta/4\tau)], \qquad (30)$$

is absolutely stable in the region bounded by the curve AQA, and is metastable in the regions AQF and $A'M'QO$.

3. The canted phase C $(\Theta_C(W, \tau) = -\Theta_B(-W, \tau))$ is absolutely stable in the region bounded by the curve OQA', and is metastable in the regions $F'QA'$ and $AMQO$. We note that certain details connected with the overlap of the two metastable phases and the presence of supplementary (metastable) canted solutions in the regions AMO and $A'M'O$ have been left out of Fig. 4a. Thus, even at finite temperatures the realignment of the magnetic structure near the level crossing point proceeds via three jumps (first-order phase transitions) on the lines AQ, OQ, and $A'Q$. At $\mathbf{H} \parallel [110]$, the H-T phase diagram of the system in the quasi-doublet approximation coincides in main outline with the diagram described by us in[3].

We consider now the case $\mathbf{H} \parallel [001]$. For finite temperatures, the equilibrium values of the angle φ will be the same as at $T = 0$ °K. The free energy (8) then takes the form

$$f = -h \cos\theta - \tau a \left[2 \ln \mathrm{ch} \left(\frac{\sin\theta}{\tau\sqrt{2}} \right) + \ln \mathrm{ch} \left(\frac{\cos\theta - h}{\tau} \right) \right], \qquad (31)$$

where $\tau = T/\mu H^{ex}$. An analysis of the free energy (31) in the small-concentration approximation leads to the following picture (see Fig. 4b):

1. At sufficiently low temperature $(T < T^* = 2a^3 \mu H^{ex})$ there exist two phases, the regions of existence of which overlap near H^{ex}. The phase A $(\mu_1 < 0)$ exists in the region below the NQ curve, and the phase B $(\mu_1 > 0)$ exists in the region above the MQ curve. The KQ curve is the line of the first-order phase transition between the two canted phases. The point $T = T^*$ is a critical point of the vapor—liquid type, above which only one canted phase exists.

2. At sufficiently high temperatures (for example, $T \sim a \mu H^{ex}$ at $H \sim H^{ex}$) the canted phase goes over smoothly (second-order transition) into the phase $\theta = 0$ on the line FE.

5. DISCUSSION OF RESULTS

A comparison of the anomalies of the magnetization of $Ho_xY_{3-x}IG$ in a strong magnetic field[2] with the theoretical conclusions that follow from the considered model show them to be patently similar. Attention is called to the following: a) the equality of the number of magnetization jumps observed in experiment and predicted by the theory at all investigated directions of the external field; b) the equal spacing of the jumps and the equality of their magnitude at $\mathbf{H} \parallel [111]$ and $\mathbf{H} \parallel [110]$; c) the behavior of the susceptibility at different field directions; both in theory and in experiment the susceptibility is maximal at $\mathbf{H} \parallel [001]$ and minimal at $\mathbf{H} \parallel [111]$; d) the similarity of the H-T phase diagrams.

We present somewhat more detailed estimates, using the experimental results of[2] (see Fig. 3 of[2]). In our model there are only two indeterminant quantities: H^{ex} and the magnetic moment μ of the "Ising" ion, which enters in the reduced concentration $a = x\mu/3m$. The quantity H^{ex} enters in the theory in the form of the scale of the field, and it can be determined in the following manner. According to the theory, the central jump of $M(H)$ at $\mathbf{H} \parallel [111]$, the mid-point between the jumps at $\mathbf{H} \parallel [110]$, and (approximately) the jump at $\mathbf{H} \parallel [001]$ correspond to $h = 1$, i.e., $H = H^{ex}$. From Fig. 3 of[2] it follows with good accuracy that all these points correspond to $H^{ex} \approx 120$ kOe. This quantity agrees with the known data.[6] To determine μ we can use the following characteristics of the $M(H)$ curves: the field intervals between the jumps are the values ΔM of the individual magnetization jumps. All are determined by a single unknown quantity μ. According to theory, we have at $\mathbf{H} \parallel [111]$

$$\Delta H_{[111]} = 2aH^{ex}/\sqrt{3}, \quad \Delta M = 2am/\sqrt{3};$$

at $\mathbf{H} \parallel [110]$ we have

$$\Delta H_{[110]} = \sqrt{2}\, aH^{ex}; \quad \Delta M = \sqrt{2}\, am;$$

and at $\mathbf{H} \parallel [001]$

$$\Delta M = 2am.$$

According to Fig. 3a of[2], $\Delta H_{[111]} = 17$ kOe and $x = 0.25$, $m = 5\mu_B$; then $\mu = 7.5\ \mu_B$. Determining the same quantity from $\Delta H_{[110]} = 23$ kOe, we obtain $\mu = 8.1\ \mu_B$. The obtained values of μ agree within the limits of experimental accuracy. The values of ΔM were measured with lower accuracy, and we confine ourselves therefore to an order-of-magnitude estimate. At $\mathbf{H} \parallel [111]$ we have $\Delta M \lesssim 1\ \mu_B$ (see Fig. 3 of[3]), where $\mu \approx 10\ \mu_B$. We note that the values of the measured jumps of $M(H)$ increase on going from $\mathbf{H} \parallel [111]$ to $\mathbf{H} \parallel [110]$ and to $\mathbf{H} \parallel [001]$, also in accord with the theory. Thus, the values of μ determined from different independent characteristics of the magnetization curves $M(H)$ of $Ho_xY_{3-x}IG$ turn out to be close to one another (within the limits of experimental accuracy). This indicates that the magnetic-structure instability mechanism due to the crossing of the levels (the magnetic analog of the Jahn–Teller effect) is indeed realized in the system $Ho_xY_{3-x}IG$.

Let us estimate the critical temperature T^* at which the first-order phase-transition lines converge at $\mathbf{H} \parallel [111]$ (Fig. 4a). The dimensionless critical temperature is $\tau^* = \frac{3}{8}$ (Fig. 4a). According to (28) we have

$$T^* = \frac{x\mu^2}{3m} H^{ex},$$

which yields $T^* \approx 15$ K at $x = 0.25$ and $\mu \approx 10\ \mu_B$. The corresponding value of T^* for $\mathbf{H} \parallel [110]$ is of the same order; the critical temperature at $\mathbf{H} \parallel [001]$ is much lower.

Let us discuss the question of the energy gap Δ, which is usually present between the singlet levels that make up the quasi-doublet (quasi-triplet). We have assumed it equal to zero. It is physically obvious (and is confirmed by calculations) that if the gap is much smaller than the additional level splitting due to the instability, then the influence of the gap can be neglected. The splitting in our case is

$$\delta\mathscr{E} \approx 2amH^{ex} = 2(x/3)\mu H^{ex}.$$

At $x = 0.25$ we have $\delta\mathscr{E} \approx 10$ cm^{-1}. For example, Ho^{3+} in YGaG has $\Delta \sim 5$ cm^{-1} [4] (see also Fig. 1a of[2]). The most interesting effect due to the presence of the gap Δ is the presence of a concentration threshold. At small x, such that $\delta\mathscr{E} < \Delta$, the instability of the magnetic structure becomes energywise unprofitable.

We have considered a model of an Ising ion, whose magnetization direction coincides with the z axis of the local environment of the rare-earth ion in the garnet. At other direction of this magnetization axis (x or y), the behavior of the system can differ strongly from the investigated one. This is apparently the case with $Tb_xY_{3-x}IG$.

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[1)] A lower bound on x is also imposed by the fact that we are considering only matrix magnetic-structure deformations that are homogeneous over the sample and are characterized by an angle θ. This is not a very strong limitation, as can be seen from the following qualitative reasoning: The perturbation $\delta\theta$ produced by the REI propagates in the matrix, owing to exchange interaction, to a region with linear dimensions on the order of $\sqrt{A/b}$, where A is the constant of the exchange between the iron ions and b is the magnetic "rigidity." It follows therefore that at $x \gg a_0^3(b/A)^{3/2}$ (a_0 is the distance between atoms) we can neglect the coordinate dependence of θ. For REIG this condition is satisfied at $x \gg 10^{-3}$—10^{-4}.

[2)] These probabilities are equivalent. We are dealing in fact with different domains, in one of which the first to reverse magnetization are ions 3 and 4 and in the second ions 5 and 6.

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