Specific features of spontaneous spin reorientation in the $Y_xLu_{1-x}CrO_3$ system with a competing anisotropy

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A comprehensive investigation was made of the characteristics of the magnetic structure of the $Y_xLu_{1-x}CrO_3$ system. A comparison of the results deduced from the measurements of the torque curves, magnetization, elongation–compression magnetostriction, and shear strains demonstrated that the structure exhibited by the $Y_{0.2}Lu_{0.8}CrO_3$ system in the range $T < 77$ K is characterized by a reduction in the crystal-average values of the ferromagnetic and antiferromagnetic vectors with no change in their orientation along the crystallographic axes. A model of a spin-orientation transition is considered for magnetic materials with a competing anisotropy, making it possible to account for the experimentally observed anomalies of magnetic and magnetoelastic properties of $Y_xLu_{1-x}CrO_3$.

INTRODUCTION

As reported earlier,[1] compounds belonging to the $Y_xLu_{1-x}CrO_3$ series with $x = 0.1$, 0.15, and 0.2 exhibit a competing magnetic anisotropy [fluctuations of the magnitude and sign of the first[1)] anisotropy constant $K_1(\mathbf{r})$ in the bulk of a sample], which is responsible for a number of experimentally observed special features of their magnetic properties. Random changes in the anisotropy constant are related to its dependence on the strain of the Cr^{3+}–60^{2-} octahedra; these strains in turn are governed by the number of the Y^{3+} (Lu^{3+}) ions in the environment of a Cr^3 ion. For example, predominance of the Y^{3+} ions gives rise to a positive constant K_1, whereas predominance of Lu^{3+} gives a negative constant. The investigated system may contain regions where the sign of $K_1(\mathbf{r})$ is opposite to the sign of the average anisotropy constant of the whole material.

Generally speaking, the temperature dependences of the first anisotropy constant are also different in the ranges $K_1(\mathbf{r}) > 0$ and $K_1(\mathbf{r}) < 0$. This may be due to, in particular, fluctuations of the Cr^{3+}–60^{2-}–Cr^{3+} superexchange interaction parameters, the values of which also depend on the degree of distortion of the oxygen octahedra (we recall that in the case of $YCrO_3$ the Néel point is $T_N = 140$ K, whereas for $LuCrO_3$ it is $T_N = 112$ K). The different temperature dependences of $|K_1(\mathbf{r})|$ in regions characterized by different signs of the anisotropy constant may alter the magnitude and sign of the average anisotropy constant of the whole material as temperature is lowered, and, consequently, may give rise to a spin-reorientation (SR) transition,i.e., may rotate the ferromagnetic $\mathbf{F}$ and antiferromagnetic $\mathbf{G}$ vectors[2] from the $\mathbf{c}$ (a) axis to the $\mathbf{a}$ (c) axis. We shall consider the specific case of a change in the orientation of the ferromagnetic vector $\mathbf{F}(\mathbf{r})$.

We consider the qualitative picture of the process of orientation in the investigated compounds which exhibit a competing anisotropy. Note that regions with different signs of $K_1(\mathbf{r})$ are exchange-coupled. In the case of small regions characterized by $K_1(\mathbf{r}) < 0$ the energy of their magnetic anisotropy is frustrated and the vectors $\mathbf{F}(\mathbf{r})$ are oriented throughout the sample along the $\mathbf{c}$ axis, i.e., the usual collinear Γ_i phase is obtained ($\mathbf{F}\|\mathbf{c}$, $\mathbf{G}\|\mathbf{a}$, $|\mathbf{F}| = \text{const}$, $|\mathbf{G}| = \text{const}$). Cooling increases the size of the regions characterized by $K_1(\mathbf{r}) < 0$ and the absolute value of the anisotropy constant increases even in those cases when the size of these regions exceeds a certain critical value, so that deviation of the vectors $\mathbf{F}(\mathbf{r})$ from the $\mathbf{c}$ axis becomes possible in the regions with $K_1(\mathbf{r}) < 0$. Since the spins are exchange-coupled, these deviations are not localized within regions characterized by $K_1(\mathbf{r}) < 0$, i.e., each such region is "dressed" by a transition layer where deviations of the vector $\mathbf{F}(\mathbf{r})$ from the $\mathbf{c}$ axis increase as we approach the center of the region with $K_1(\mathbf{r}) < 0$; the thickness of the transition layer is comparable with the thickness of domain walls $\delta \approx \{\alpha/\langle|K_1(\mathbf{r})|\rangle\}^{1/2}$, where α is the inhomogeneous exchange parameter[3] and the angular brackets denote (here and later) the value averaged over the fluctuations of $K_1(\mathbf{r})$.

If there are only a few regions characterized by $K_1(\mathbf{r}) < 0$ and the distances between them exceed δ, then the deviations of the vectors $\mathbf{F}(\mathbf{r})$ from the c axis observed in different regions with $K_1(\mathbf{r}) < 0$ are not correlated, so that a fan-shaped $AS(\Gamma_4)$ asperomagnetic structure appears in the ac plane. In addition to the vectors $\mathbf{F}(\mathbf{r})\|\mathbf{c}$, the system is equally likely to include a vector $\mathbf{F}(\mathbf{r})$ for which the rotation from the $\mathbf{c}$ to the $\mathbf{a}$ axis is clockwise or counterclockwise, i.e., $\langle\mathbf{F}(\mathbf{r})\rangle\|\mathbf{c}$.

A further increase in the number and dimensions of the regions characterized by $K_1(\mathbf{r}) < 0$ as a result of cooling shortens the distances between them. Finally, they approach each other so much that the distributions $\mathbf{F}(\mathbf{r})$ in the vicinities of different regions cease to be independent and their mutual correlation gives rise to a canted phase in which the average vector $\langle\mathbf{F}\rangle$ deviates from the anisotropy axis $\mathbf{c}$. It should be pointed out that in this formulation the problem of spin reorientation from the asperomagnetic to the canted phase is equivalent to the classical problem of percolation.[4] Similarly, an orientation transition takes place from a $\Gamma_2(F_xG_z)$ phase to the canted phase.

The appearance of characteristic fan-shaped $AS(\Gamma_{2,4})$ asperomagnetic structures with $\langle\mathbf{F}\rangle\|\mathbf{a},\mathbf{c}$ is the main feature of the SR transition in the investigated systems characterized by a competing magnetic anisotropy. A typical property

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of phases of this kind is SR to the canted phase Γ_4, which is induced by a weak field $H \ll H_{\text{th}} \approx |\langle K_1 \rangle|/|\langle \mathbf{F} \rangle|$ and which is accompanied by a considerable deviation of $\langle \mathbf{F} \rangle$ from the easy magnetization axis. It can be explained on the basis of the above pattern of the distribution of $\mathbf{F}(\mathbf{r})$ in asperomagnetic structures. In fact, application of a field H at an arbitrary angle to the *ac* plane results in magnetization reversal in the regions characterized by $K_1(\mathbf{r}) < 0$ where $\mathbf{F}(\mathbf{r})$ is inclined to H at an angle exceeding $\pi/2$. Such magnetization reversal is an abrupt process and occurs in a certain critical field H_c, the value of which can be easily shown to be less than the local anisotropy field in regions with $K_1(\mathbf{r}) < 0$. This feature of asperomagnetic structures gives rise to a characteristic dualism in their magnetic and magnetoelastic properties: in some experiments the compounds with a competing anisotropy exhibit properties characteristic of the collinear phases Γ_2 and Γ_4, whereas in other experiments they have properties typical of the canted phase Γ_{24}.

It should be pointed out that the behavior of these compounds in the canted phase is also characterized by a number of special features. In particular, as shown below, the long-range magnetic order [angular orientation of the vectors $\mathbf{F}(\mathbf{r})$ in this phase] can be destroyed by large-scale fluctuations of the orientation of $\mathbf{F}(\mathbf{r})$ which give rise to domains. It is known[5] that such processes are typical of disordered systems and, in particular, may be observed in spin glasses characterized by fluctuations of local orientations of the easy magnetization axes.[5]

The present paper reports an investigation of stochastic magnetic structures formed as a result of SR transitions in $Y_xLu_{1-x}CrO_3$ compounds with a competing anisotropy.

SAMPLES AND EXPERIMENTAL METHOD

Single crystals of $Y_xLu_{1-x}CrO_3$ ($x = 0.1$, 0.2) were grown from a molten solution in lead compounds. We investigated the magnetostriction along the **a**, **b**, and **c** axes of the crystals, the magnetization along the **a** and **c** axes, the torques, and the shear strains in pulsed fields oriented in different ways in the *ac*, *bc*, and *ab* planes and in a rotating static field $H = 0.5$–5 kOe. The strains were measured using a quartz piezoelectric transducer bonded to a sample. All the measurements were made at temperatures 4.2–130 K.

In using these methods it is necessary to allow for the characteristics of the crystal and magnetic symmetry of $RCrO_3$ compounds.[2] For example, if the process of SR occurs in the ac plane, the projection of the spontaneous magnetic moment **F** onto the **b** axis vanishes, so that when the field **H** lies in a plane containing the difficult magnetization axis (DMA) and the **b** axis, the expression for the torque is

$$L_{\text{DMA},b} = F(H_{\text{DMA}})\, H \sin\psi \sin\theta, \tag{1}$$

where H_{DMA} is the projection of the field along the DMA axis; θ is the angle between **F** and the **c** axis; ψ is the angle between **H** and the difficult magnetization axis. In writing down Eq. (1) is is assumed that the external field H is much less than the exchange field H_E (Ref. 2). In the range $H \ll H_{\text{th}}$, where H_{th} is the threshold magnetization field along the difficult direction, we have $\sin\theta \approx H_{\text{DMA}}/H_{\text{th}}$ (Ref. 2) and

$$L_{\text{DMA},b} \approx H^2 \sin 2\psi. \tag{2}$$

However, if the field H lies in a plane containing the easy magnetization axis (EMA) and the **b** axis, then in the range $H \ll H_E$, we have

$$L_{\text{EMA},b} = F(H_{\text{EMA}})\, H \sin\varphi, \tag{3}$$

where H_{EMA} is the projection of the field along the EMA and φ is the angle between **c** and **H**. The relationships given by Eqs. (1) and (3) will be used later to determine the nature of the magnetization curves in the easy and difficult directions. A special role in studies of stochastic magnetic structures in these compounds is played by measurements of the torque in the *ac* plane, because such measurements provide information on the characteristic (for compounds with a competing magnetic anisotropy) dependence of the average magnetic field $\langle \mathbf{F} \rangle$ on its orientation.[1]

Measurements of the torques give information on the magnitude and orientation of the ferromagnetic vector **F**, whereas measurements of the elongation–compression magnetostriction along the *a*, *b*, and *c* axes (λ_a, λ_b, λ_c) and of monoclinic distortions in the *ab*, *ac*, *bc* planes (λ_{ab}, λ_{ac}, λ_{bc}) can give the corresponding information on the antiferromagnetic vector of the investigated systems. For example, SR in the *ac* plane is characterized by

$$\lambda_i = \lambda_{i0} G^2 \sin^2\theta, \quad i = a,\ b,\ c, \tag{4}$$

where θ is the angle of deviation of **G** from the EMA and the constants λ_{i0} can have different signs for the **a**, **b**, and **c** axes.

The most sensitive method which can be used to detect extremely small deviations of the vector **G** from the **a** and **c** axes as a result of an SR transition in the *ac* plane is the determination of the monoclinic strains λ_{ac} (Ref. 6):

$$\lambda_{ac} = \lambda_{ac}^{0} G^2 \sin 2\theta. \tag{5}$$

It is clear from Eq. (5) that the sign of the shear strain λ_{ac} depends on direction of rotation of $\langle \mathbf{G} \rangle$ as a result of SR and its magnitude reaches its maximum value when $\theta = \pi/4$.

Only the average values $\langle \lambda_i \rangle$ and $\langle \lambda_{ij} \rangle$ were determined for the investigated disordered compounds with stochastic magnetic structures. We shall represent the angle $\theta(\mathbf{r})$, which describes the local orientation of the antiferromagnetic vector $\mathbf{G}(\mathbf{r})$, in the form $\theta(\mathbf{r}) = \bar{\theta} + \delta\theta(\mathbf{r})$, where $\bar{\theta} = \langle \theta(\mathbf{r}) \rangle$ determines the orientation of $\langle \theta(\mathbf{r}) \rangle$. Then, in the case of sufficiently small values of $\delta\theta(\mathbf{r})$, expanding $\sin^2\theta(\mathbf{r})$ in Eq. (4) and $\sin 2\theta(\mathbf{r})$ in Eq. (5) as a series in $\delta\theta(\mathbf{r})$, we need to include only terms through second order, and after averaging we obtain the following approximate relationships:

$$\langle \lambda_i \rangle = \lambda_{i0}\{D_\theta + \sin^2\bar{\theta}(1-2D_\theta)\}G^2, \tag{6a}$$

$$\langle \lambda_{ac} \rangle = \lambda_{ac}^{0} \sin 2\bar{\theta}(1-2D_\theta)G^2, \tag{6b}$$

where

$$D_\theta = \langle \delta\theta^2(\mathbf{r}) \rangle. \tag{7}$$

These expressions show that we can distinguish two contributions to the observed magnetostriction: the first, which we shall later call "normal," is related to rotation of the average antiferromagnetic vector $\langle \mathbf{G} \rangle$, and the second "anomalous" component is due to a change in the dispersion of the orientational fluctuations of this vector $\langle \delta(\theta^2(\mathbf{r})) \rangle$. It is shown in Ref. 1 that the anomalous contribution is respon-

sible for the singularities of the dependence $\lambda_{ac}(H)$ in the course of a field-induced $\Gamma_2(G_zF_x) \rightarrow \Gamma_4(G_xF_x)$ SR transition in $Y_{0.1}Lu_{0.9}CrO_3$. It should be pointed out that this anomalous magnetostriction can be observed also in field-induced transitions of these structures to collinear phases Γ_4 and Γ_2.

EXPERIMENTAL RESULTS

Measurements of the torques and field dependences of the magnetostriction λ_{ac} for $\mathbf{H}\|\mathbf{a}$ and $\mathbf{H}\|\mathbf{c}$[1,7] showed that for all the investigated compositions the magnetic structure at temperatures $T < T_N$ is Γ_4 with the magnetic moment oriented along the c axis. In the case of compositions with $x = 0.1$ and 0.2, we have $T_N = 113$ and 118 K, respectively. In a sample of $Y_{0.1}Lu_{0.9}CrO_3$ the magnetic moment is oriented along the **a** axis (Γ_2 phase) at temperatures $T < 80$ K (Refs. 1 and 7), whereas in the range 80–95 K the magnetic structure is modified from the high-temperature phase Γ_4 to the low-temperature phase Γ_2. It is clear from Figs. 1a and 1b that in this range of temperatures the torque curves $L_{ac}(\varphi)$ and the field dependence of the magnetostriction [the dependence $\lambda_c(H)$ is given by way of example] differ considerably from those usually observed in the case of SR transitions in $RCrO_3$ compounds. For example, the dependence $\lambda_c(H)$ is strongly nonmonotonic, suggesting the presence of two competing contributions to λ_c, which may be, in particular, the normal and anomalous magnetostrictions described above and due to the appearance at $T \leqslant 95$ K of the $AS(\Gamma_4)$ asperomagnetic structure mentioned above. This hypothesis agrees with the results presented in Fig. 1(c): the magnetostriction λ_a observed at temperatures $T \leqslant 95$ K in the $\mathbf{H}\|\mathbf{c}$ orientation can be attributed to the $AS(\Gamma_4) \rightarrow \Gamma_4$, transition induced by the field $\mathbf{H}\|\mathbf{c}$. It should be pointed out

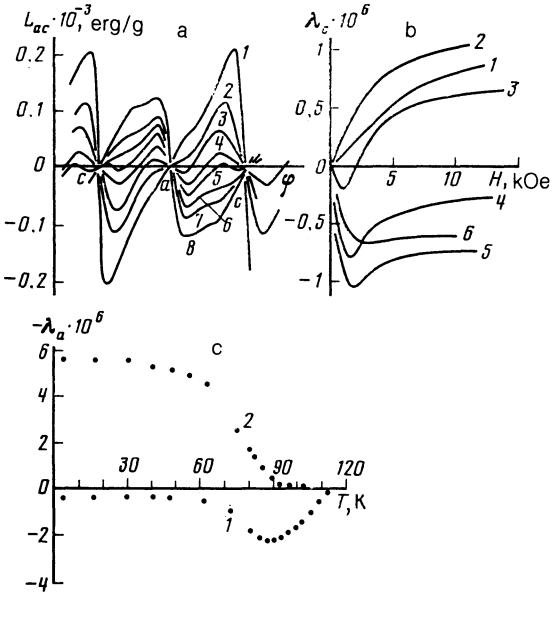

FIG. 1. Experimental results for a sample of $Yu_{0.1}Lu_{0.9}CrO_3$: a) angular dependence of the torque in a field $H = 600$ Oe in the ac plane at temperatures 73 K (curve 1), 77 K (2), 79 K (3), 81 K (4), 82 K (5), 83 K (6), 84 K (7), 87 K (8); b) field dependence of the magnetostriction at temperatures 4.2 K (curve 1), 62 K (2), 84 K (3), 91 K (4), 95 K (5), 106 K (6); c) temperature dependence of the magnetostriction λ_a^{max} in fields $\mathbf{H}\|\mathbf{a}$ (1) and $\mathbf{H}\|\mathbf{c}$ (2).

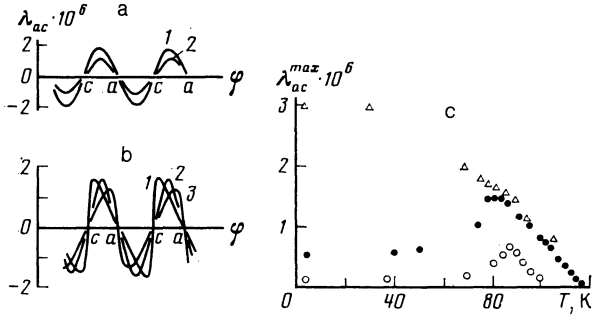

FIG. 2. Monoclinic distortions of $Y_{0.1}Lu_{0.9}CrO_3$: a) orientational dependence in a field $H = 5$ kOe at temperatures 77 K (curve 1) and 95 K (curve 2); b) orientation dependence in a field $H = 600$ Oe at temperatures 79 K (curve 1), 83 K (2), 93 K (3); c) temperature dependence of the maximum value λ_{ac}^{max} in fields $H = 80$ Oe (○), 500 Oe (●), 5500 Oe (△).

that the anomalous magnetostriction in the field $\mathbf{H}\|\mathbf{a}$ is observed at temperatures $T \leqslant 80$ K when, according to the results of measurements of $L_{ac}(\varphi)$ (Ref. 1), the magnetic moment of the sample is parallel to the **a** axis. This makes it possible to postulate the existence of the $AS(\Gamma_2)$ asperomagnetic structure at temperatures $T \leqslant 80$ K.

These hypotheses were checked by measuring also the shear strains in the ac plane. It is clear from Fig. 2a that the $\lambda_{ac}(\varphi)$ curves recorded in a field $H = 5$ kOe at temperatures 77 and 95 K can be described satisfactorily by a dependence of the $\lambda_{ac} \propto \sin 2\varphi$ type. This shows that a field $H = 5$ kOe rotated from the **c** to the **a** axis induced reorientation of the $\langle \mathbf{G} \rangle$ vector from **a** to **c** [this is in agreement with the results of measurements of $\lambda_c(H)$ shown in Fig. 1b] and the magnitude of $\langle \mathbf{G} \rangle$ was practically unaffected i.e., the orientational fluctuations of $\mathbf{G}(\mathbf{r})$ were suppressed and made no significant contribution to $\lambda_{ac}(\varphi)$ see [Eqs. (6) and (7)].

In the range 80–95 K the values of λ_{ac}^{max} are practically the same in fields of 5.5 kOe and 500 Oe (Fig. 2c). Moreover, in a field of just 80 Oe the ratio $\lambda_{ac} \approx (1/3)\lambda_{ac}^{max}$ ($H = 5.5$ kOe) reaches its maximum value at 80 Oe. It follows that in this range of temperatures a very weak field ($H = 500$ Oe) is sufficient to cause deviation of the vectors $\langle \mathbf{F} \rangle$ and $\langle \mathbf{G} \rangle$ by relatively large angles [we recall that monoclinic strains reach their maximum value when the angle between $\langle \mathbf{G} \rangle$ and the easy axis is $\pi/4$—see Eq. (6)].

However, for $\theta = \pi/4$, orientational fluctuations of $\mathbf{G}(\mathbf{r})$ are suppressed practically completely in fields $H \approx 5$ kOe and 600 Oe, and are reduced significantly even in a field $H \approx 80$ Oe [this is deduced from the fact that $\lambda_{ac}^{max}(H = 5\ \text{kOe}) \approx \lambda_{ac}^{max}(H = 600\ \text{Oe}) \approx 3\lambda_{ac}^{max}(H = 80\ \text{Oe})$]. The form of the $\lambda_{ac}(\varphi)$ curves obtained in a field $H = 600$ Oe (Fig. 2b) does however differ considerably from sin 2φ and, like the $L_{ac}(\varphi)$ and $\lambda_{ac}(H)$ curves (Fig. 1), is untypical of ordered $RCrO_3$, compounds. Linear sections of some $\lambda_{ac}(\varphi)$ isotherms and $L_{ac}(\varphi)$ curves (Fig. 1a) observed at the same temperatures suggest that a rotating field $H = 600$ Oe induces first-order orientational phase transitions which involve passage through an intermediate state,[8] in which different magnetic structures coexist. We shall show in the next section that these anomalies of the $\lambda_{ac}(\varphi)$ and $L_{ac}(\varphi)$ curves are typical of compounds with a competing magnetic anisotropy.

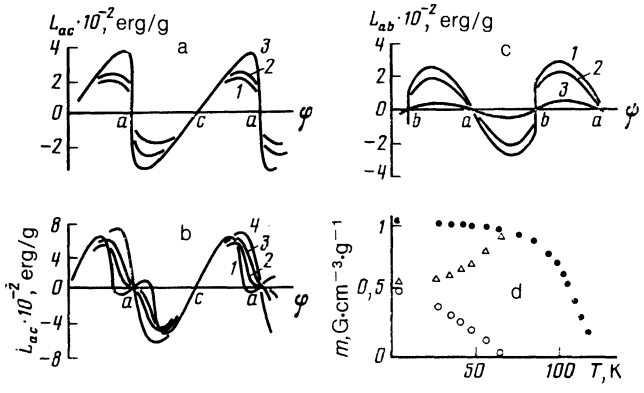

FIG. 3. Experimental results for a sample of $Y_{0.2}Lu_{0.8}CrO_3$: a) angular dependence of the torque in the *ac* plane in a field $H = 500$ Oe at temperatures 4.2 K (curve 1), 59 K (2), 90 K (3); b) angular dependence of the torque in the *ac* plane in a field $H = 4.4$ kOe at temperatures 4.2 K (curve 1), 42 K (2), 55 K (3), 90 K (4); c) angular dependence of the torque in the *ab* plane in a field $H = 500$ Oe at temperatures 4.2 K (curve 1), 51 K (2), 99 K (3); d) temperature dependence of the magnetization along the *a* axis in fields $H \gtrsim 50$ Oe (○), along the *c* axis (△), and of the saturation magnetization (●) in fields $H > H_{th}$, deduced from the field dependence of the magnetization.

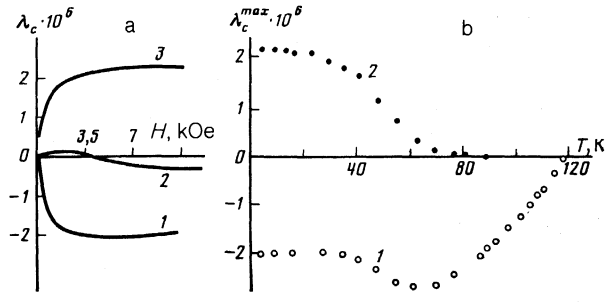

FIG. 4. Elongation–compression magnetostriction of $Y_{0.2}Lu_{0.8}CrO_3$: a) field dependence of λ_c at $T = 4.2$ K for some orientations of an external field: 1) **H**‖**a**, 2) $\varphi = 45°$, 3) **H**‖**c**; b) temperature dependence of λ_{ac}^{max} obtained for field orientations along the **a** axis (curve 1) and the **c** axis (curve 2).

A detailed investigation of magnetic structures formed as a result of SR processes was made by synthesizing a sample with a high yttrium concentration ($Y_{0.2}Lu_{0.8}CrO_3$) in which reorientation was expected at much lower temperatures. Figure 3a shows the angular dependences of the torque of $Y_{0.2}Lu_{0.8}CrO_3$ in the *ac* plane obtained at various temperatures in a field $H = 500$ Oe. The characteristic nature of the curves obtained in the **H**‖**a** configuration at temperatures $T > 77$ K (for example, at $T = 90$ K) demonstrates that a spontaneous magnetic configuration of the Γ_4 type (**F**‖**c**) exists in the range $77\text{K} < T < 118$ K. This is supported also by the results of measurements of the magnetization and magnetostriction. At temperatures below 77 K the curves again demonstrate that the projection of the spontaneous electric moment along the **a** axis vanishes, but cooling from 77 to 4.2 K reduces the magnitude of the jump of $L_{ac}(\varphi)$ proportional to the spontaneous magnetic moment $m_c \propto \langle \mathbf{F} \rangle$.

The torques obtained in the *ab* and *bc* planes tend to suggest a canted orientation of the spontaneous magnetic moment. In fact, at temperatures below 77 K a nonzero jump is observed in the $L_{ab}(\psi)$ curves (Fig. 3c), indicating the appearance of the component m_a. Direct measurements of the magnetization $m_a(H_a)$ and $m_c(H_c)$ also suggest the existence of a canted phase Γ_{24} at temperatures $T < 77$ K (Fig. 3d): in a field $H \approx 50$ Oe, which is considerably less than the threshold field for the $\Gamma_2 \rightarrow \Gamma_4$ transition, the magnetization along the **a** axis appears and increases as a result of cooling. The spontaneous magnetic moment along the **c** axis then decreases in magnitude.

Equally contradictory are the results of measurements of the magnetoelastic properties of $Y_{0.2}Lu_{0.8}CrO_3$. The presence of a canted phase can be deduced from the field dependence of the elongation–compression magnetostriction along the **c** axis (Fig. 4a). In fact, the opposite signs but equal magnitudes of the magnetostriction in the **H**‖**a** and **H**‖**c** cases, together with zero magnetostriction when the field H_{coerc} is along the diagonal in the *ac* plane at $T = 4.2$ K, are typical of the Γ_{24} canted phase with the orientations of the vectors $\langle \mathbf{F} \rangle$ and $\langle \mathbf{G} \rangle$ at $\theta = \pi/4$ relative to the **a** and **c** axes. The temperature dependence of the saturation magnetostriction (Fig. 4b) shows that such a structure exists in the temperature range 4.2–77 K. However, as shown in Ref. 6, the gradual magnetization reversal in a rotating magnetic field $H_{coerc} < H < H_{th}$ should be accompanied by four jump-like changes in the shear strains in the *ac* plane.

Figure 5a shows the angular dependences of monoclinic distortions of a $Y_{0.2}Lu_{0.8}CrO_3$ crystal in fields $H = 600$ Oe and 5 kOe at $T = 4.2$ K. It is clear that the dependence $\lambda_{ac}(\varphi)$ has a fundamentally different nature: the value of λ_{ac}^{max} increases with the field intensity and when the field is close to the **a** axis, the fall of the magnetostriction is steeper than in a field oriented along the **c** axis. Moreover, the field dependence of monoclinic distortions at low temperatures is also contradictory (it is shown in Fig. 5c for $T = 4.2$ K). As at temperatures $T > 77$ K (Fig. 5b), the application of a magnetic field along the **c** axis does not induce shear strains, and when the field is oriented along the diagonal in the *ac* plane, the strain reaches its maximum value λ_{ac}^{max}. This behavior is frequently exhibited by pure orthochromites outside the region of the spontaneous $\Gamma_2 \rightarrow \Gamma_4$ SR region.[1] How-

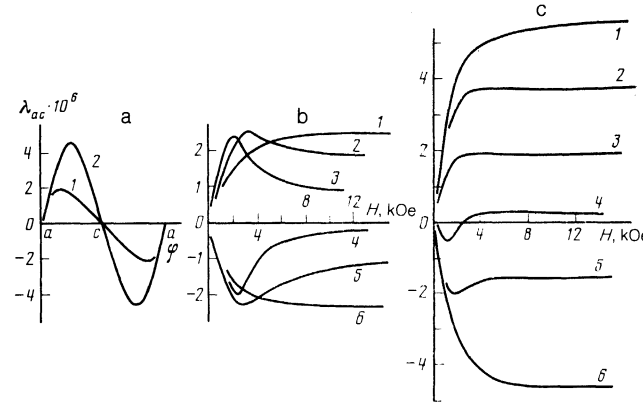

FIG. 5. Monoclinic distortions of $Y_{0.2}Lu_{0.8}CrO_3$: a) angular dependence obtained at $T = 4.2$ K in fields $H = 600$ Oe (curve 1) and $H = 5$ kOe (2); b) field dependence at $T = 85$ K in a field oriented at $\varphi = 45°$ (curve 1), 75° (2), 85° (3), − 85° (4), − 75° (5), − 45° (6) relative to the **c** axis; c) field dependence at $T = 4.2$ K in a field oriented at $\varphi = 45°$ (curve 1), 75° (2), 85° (3), − 85° (4), − 75° (5), − 45° (6) relative to the **c** axis in the *ac* plane.

ever, when the field is oriented near the **a** axis, the monolinic distortions at $T = 4.2$ K do not have bell-shaped dependences, but at temperatures above 77 K they are manifested clearly, which is the usual characteristic of the $\Gamma_4 \to \Gamma_2$ transitions.

Figure 3b gives the angular dependences of the torque $L_{ac}(\varphi)$ in a field $H = 4.4$ kOe $> H_{th}$. Throughout the investigated temperature range the **c** axis is the axis of easy magnetization, as confirmed by the results obtained in weak fields. Below 77 K there is a "dip" in the region of the $\varphi = 45°$ orientation. The magnitude of the dip increases as a result of cooling, causing reversal of the sign of $L_{ac}(\varphi)$ below $T = 20$ K for field orientations close to the **a** axis.

It follows that at $T = 4.2$ K a crystal of $Y_{0.2}Lu_{0.8}CrO_3$ has a complex magnetic structure characterized by dual manifestation of magnetic, anisotropic, and magnetoelastic properties. Some of the experimental results suggest a spontaneous configuration with the average magnetic moment vector along the **c** axis, which is smaller than the magnetic moment in the Γ_4 phase. Other results suggest that beginning with fields of $H \approx 50$–100 Oe this state can be regarded as the Γ_{24} canted phase with the orientation $\bar{\theta}$ along the diagonal in the *ac* plane. An analysis of all the experimental results demonstrates that they cannot be explained self-consistently by assuming that spontaneous SR in $Y_{0.1}Lu_{0.9}CrO_3$ and that occurring at low temperatures ($T < 77$ K) in $Y_{0.2}Lu_{0.8}CuO_3$ creates any of the known magnetic structures typical of ordered orthochromites.

DISCUSSION OF RESULTS

In an analysis of possible magnetic structures that can appear in the investigated compounds as a result of SR transitions we follow Ref. 1 and use the micromagnetism approximation by introducing a thermodynamic potential, which in [illegible] is of the form[9]

$$\Phi = \iiint \left\{ \frac{\alpha}{2} (\nabla \mathbf{G})^2 - \frac{K_1(\mathbf{r})}{2} G_x^2 - F_c^0 H_z G_x + F_a^0 H_x G_z \right\} d\mathbf{r}, \quad (8)$$

where $F_c^0 \approx F_a^0$ is the spontaneous local magnetic moment in the Γ_4 and Γ_2 phases due to the antisymmetric Dzyaloshinskiĭ–Moriya exchange. For simplicity, we assume that this interaction does not exhibit fluctuations and that $F_c^0 = F_a^0 = \text{const} = F_0$; this does not affect the qualitative results obtained below.

The random anisotropy constant $K_1(\mathbf{r})$ can be represented by its average value $\langle K_1 \rangle = \bar{K}_1$ and by a correlation function $K(\mathbf{r}_1, \mathbf{r}_2) = \langle \delta K_1(\mathbf{r}_1) \delta K_1(\mathbf{r}_2) \rangle$, which would satisfy the principle of weakening of correlations,[10] i.e., we should have $K(\mathbf{r}_1, \mathbf{r}_2) \to 0$ in the limit $|\mathbf{r}_1 - \mathbf{r}_2| \to \infty$, and the condition of statistical homogeneity of fluctuations of $K_1(\mathbf{r})$: $K(\mathbf{r}_1, \mathbf{r}_2) = K(\mathbf{r}_1 - \mathbf{r}_2)$ (Ref. 10). These requirements are satisfied by a function of the $K(\mathbf{r}_1 - \mathbf{r}_2) = D_k \exp(-|\mathbf{r}_1 - \mathbf{r}_2|/R_{ck})$ type, usually employed in a theoretical analysis of the properties of disordered magnetic materials[11]; here, D_k is the dispersion and R_{ck} is the correlation radius of fluctuations of the anisotropy. It should be pointed out that the form of $K(\mathbf{r}_1, \mathbf{r}_2)$ determines the temperatures and critical fields of SR transitions, whereas the qualitative nature of the SR process (the number of phases which occur in the course of the transition and the sequence in which they are formed) is practically independent of the set $K(\mathbf{r}_1, \mathbf{r}_2)$ if it satisfies the requirements set out above.

We can find the angle $\bar{\theta}$ governing the orientation of the average ferromagnetic vector relative to the **c** axis by introducing an effective thermodynamic potential $\Phi_{\text{eff}}(\bar{\theta})$ obtained after elimination of fluctuations $\delta\theta(\mathbf{r}) = \theta(\mathbf{r}) - \bar{\theta}$ from Eq. (8), as shown in Ref. 12:

$$\frac{\Phi_{eff}(\bar{\theta})}{\alpha/R_{ck}^2} \approx -\frac{\bar{K}_1}{2}\cos^2\bar{\theta} - \frac{\bar{D}_k \sin^2\bar{\theta}\cos^2\bar{\theta}}{\{\alpha + [\bar{K}_1 \cos 2\bar{\theta} + \bar{h}\cos(\bar{\theta} - \varphi)]^{1/2}\}^2} - \bar{h}\cos(\bar{\theta} - \varphi), \quad (9)$$

where

$$\bar{K}_1 = \bar{K}_1 R_{ck}^2/\alpha, \quad \bar{D}_k = D_k R_{ck}^4/\alpha_2, \quad \bar{h} = HF_0 R_{ck}^2/\alpha, \quad \varphi = (\mathbf{H}, \mathbf{c}).$$

The ranges in which the equilibrium phases $\bar{\theta}_{1,2} = 0, \pi/2$, and $0 < \bar{\theta}_3 < \pi/2$, exist, deduced from the conditions for a minimum of $\Phi_{\text{eff}}(\bar{\theta})$, are shown in Fig. 6. In addition to the angle $\bar{\theta}$ the stochastic magnetic structure can be described also by a correlation function $K_\theta(\mathbf{r}_1, \mathbf{r}_2)$ of the orientational fluctuations $\delta\theta(\mathbf{r})$, which in the case of these systems plays the same role as the Edwards–Anderson parameter[13] introduced for spin glasses. As shown in Ref. 14, $K_\theta(\mathbf{r}_1, \mathbf{r}_2) = \langle \delta\theta(\mathbf{r}_1)\delta\theta(\mathbf{r}_2) \rangle$ is given by

$$K_\theta(\mathbf{r}_1, \mathbf{r}_2) = \frac{D_k \sin^2\bar{\theta}\cos^2\bar{\theta}}{a(\bar{\theta})[1 + a(\bar{\theta})]^3 - \bar{D}_k \cos^2 2\bar{\theta}} \exp\left[-\frac{|\mathbf{r}_1 - \mathbf{r}_2|}{R_{ck}/a(\bar{\theta})}\right], \quad (10)$$

where

$$a(\bar{\theta}) = \left\{ \bar{K}_1 \cos 2\bar{\theta} - \frac{\bar{D}_k \cos^2 2\bar{\theta}}{\{1 + [\bar{K}_1 \cos 2\bar{\theta} + \bar{h}\cos(\bar{\theta} - \varphi)]^{1/2}\}^2} + \bar{h}\cos(\bar{\theta} - \varphi) \right\}^{1/2}. \quad (11)$$

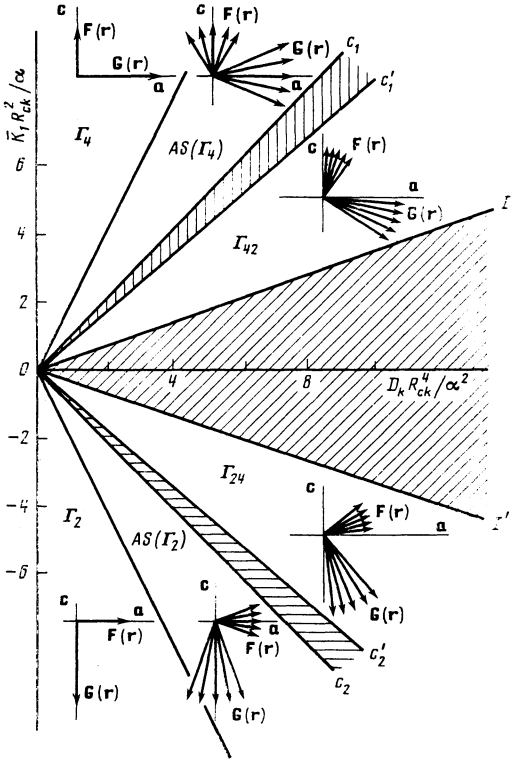

FIG. 6. Schematic phase diagram of the $Y_xLu_{1-x}CrO_3$ system. The shaded regions represent intermediate polydomain states.

If $\bar{\theta}_{1,2}=0,\ \pi/2$, the nonzero values of the dispersion $D_\theta = K_\theta(-\mathbf{r},\mathbf{r})$ are possible only if we satisfy the condition

$$a(\bar{\theta}=0,\ \pi/2)\,[1+a(\bar{\theta}=0,\ \pi/2)\,]^3=\tilde{D}_k, \qquad (12)$$

which sets the limit of existence of the $AS(\Gamma_4)$ and $AS(\Gamma_2)$ asperomagnetic structures (Fig. 6).

We shall first consider the case when $H=0$. The angle $\theta(\mathbf{r})$, governed by the deviation of the vector $\mathbf{F}(\mathbf{r})$ from the **c** (or **a**) axis, is the order parameter of SR transitions. In the case of $AS(\Gamma_{42})$ asperomagnetic structures the correlation radius of the fluctuations $\delta\theta(\mathbf{r})$, which amounts to $R_\theta = R_{ck}/a(0,\pi/2)$, is finite, i.e., these are phases with a short-range order. On reduction of $|K_1(\mathbf{r})|$ in the case of the $AS(\Gamma_{4,2})$ phases the value of R_θ increases and becomes infinite on the OC_1 and OC_2 lines. This corresponds to a correlation of the deviations of the vectors $\mathbf{F}(\mathbf{r})$ from the **c** and **a** axes, discussed above, which appears in the vicinity of various regions characterized by a negative constant $K_1(\mathbf{r})$.

We now consider a canted phase with a long-range order in the orientation of the vectors $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$, at any point in a crystal they are inclined relative to the **c** and **a** axes. It is clear from Eq. (10) that the correlation radius of the orientational fluctuations $\delta\theta(\mathbf{r})$ is in the form of the denominator of the argument of the exponential function $R_\theta = R_{ck}/a(\bar{\theta})$, whereas the dispersion of these fluctuations $D_\theta = \langle\delta\theta^2(\mathbf{r})\rangle = K(\mathbf{r}_1=\mathbf{r}_2)$ is governed by the preexponential factor in Eq. (10). The simultaneous unbounded rise of R_θ and D_θ in the approach to $a(\bar{\theta})\to 0$ is evidence of destruction of the long-range order and the appearance, as in the case of other disordered magnetic materials,[5] of a magnetically heterogeneous state in which macroscopic domains of phases with different orientations of the magnetic moments coexist. For $H=0$, the quantity $a(\bar{\theta})$ tends to zero near the center of the SR region (when $|\tilde{K}_1|\to 0$ and $\bar{\theta}\to\pi/4$) and near the boundaries OC_1 and OC_1'.

Since the phase diagram in Fig. 6 is symmetric relative to $\tilde{K}_1=0$, we shall consider the specific case of SR from the Γ_4 to the Γ_2 phase when in the region OII' there is a heterogeneous state representing a mixture of domains of the canted phase and nuclei of the Γ_2 structure. It is shown in Ref. 14 that the nuclei appear at negative fluctuations $K_1(\mathbf{r})$ of sufficiently large absolute value or extent. Following Ref. 14, we can easily show that a polydomain state observed in the OC_1C_1' range is a mixture of domains of the canted and Γ_4 structures, and the latter are localized near strong or extended fluctuations of $K_1(\mathbf{r})$.

It therefore follows that the $\Gamma_4\to\Gamma_2$ SR process in magnetic materials with a competing anisotropy is complex. We can assume that a crystal of $Y_{0.1}Lu_{0.9}CrO_3$ exhibits all the structures shown in Fig. 6, whereas an increase in the concentration of Y^{3+} in $Y_{0.2}Lu_{0.8}CrO_3$ results in a greater (compared with the $Y_{0.1}Lu_{0.9}CrO_3$ system) positive contribution to K_1 so that $K_1>0$ does not decrease to the values admitting the existence of the canted phase, and all the way down to helium temperatures we have the $AS(\Gamma_4)$ structure.

We shall now consider the behavior of systems with a competing anisotropy when they are subjected to an external magnetic field. Figure 7a gives the angular dependence of the field $H_c^{AS}(\varphi)$ necessary to suppress the asperomagnetic structure. It should be pointed out that in fields directed

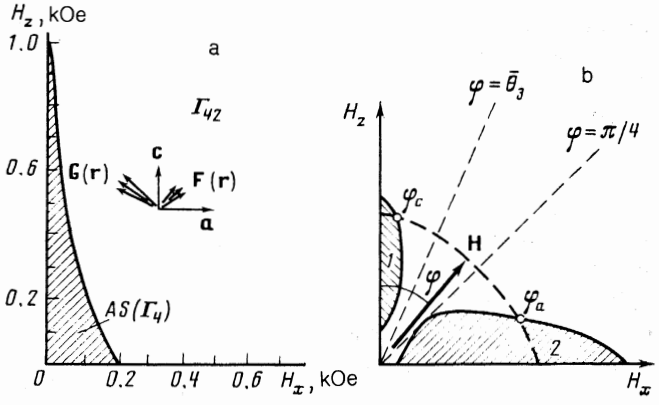

FIG. 7. Phase diagrams of a magnetic material with a competing anisotropy and an external field: a) $Y_{0.2}Lu_{0.8}CrO_3$ at $T=4.2$ K [$AS(\Gamma_4)$ phase in $H=0$]; b) canted Γ_{24} phase (shown schematically) with shaded regions representing intermediate polydomain states in which domains of the Γ_4 and Γ_{24} phases (region 1) or Γ_2 and Γ_{24} phases (region 2) coexist.

along the easy or difficult magnetization axes and exceeding the corresponding critical values of $H_{c\parallel}^{AS}$ and $H_{c\perp}^{AS}$, orientational fluctuations of the vectors $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$ are completely suppressed and we then obtain $\Gamma_4(G_x,F_z)$ and $\Gamma_2(G_zF_x)$ structures, respectively.

The diagram in Fig. 7a can account for the observed features of the behavior of a sample of $Y_{0.2}Lu_{0.8}CrO_3$ at temperatures $T\leqslant 77$ K if we assume that the $AS(\Gamma_4)$ magnetic phase forms in this sample. Then, rotation of the field **H** in the *bc* plane suppresses the fluctuations of $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$ if the field is sufficiently close in orientation to the **c** axis, i.e., if $H_z > H_{c\parallel}^{AS}$. It follows from the measured field dependences of the magnetization that $H_{c\parallel}^{AS}\approx 1$ kOe and that the critical value of the projection of the field H_a' resulting in a transition of the $AS(\Gamma_4)$ structure to the canted phase is less than 100 Oe. Complete suppression of the fluctuations of $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$ and transition to the Γ_2 phase occur in a field $H_x > H_{c\perp}^{AS}$, where $H_{c\perp}^{AS}\approx 1.5$ kOe. Rotation of a field $H=500$ Oe in the *ac* plane induces the Γ_{42} canted structure (Fig. 7a) when **H** is oriented near the **c** axis. This gives rise to the experimentally observed (Fig. 3) dependence $L_{ac}(\varphi)$. A field $H=4.4$ kOe $> H_{c\parallel}^{AS}, H_{c\perp}^{AS}$ suppresses the $AS(\Gamma_4)$ asperomagnetic phase; the anomalies of the $L_{ac}(\varphi)$ curves (Fig. 3) are then due to the nonmonotonic dependence of the average magnetic moment on its orientation [$F(\bar{\theta})=|\langle\mathbf{F}(\mathbf{r})\rangle|$]; these anomalies are discussed in Ref. 1.

The hypothesis of the existence of the $AS(\Gamma_4)$ phase in $Y_{0.2}Lu_{0.8}CrO_3$ at $T\leqslant 77$ K accounts also for the observed features of the magnetostriction (Fig. 4). In fact, in this case the application of a field $\mathbf{H}\|\mathbf{c}$ gives rise only to an anomalous magnetostriction which is due to suppression of the fluctuations of $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$. The field $\mathbf{H}\|\mathbf{c}$ first induces a transition from the $AS(\Gamma_4)$ to the canted phase Γ_{42}, which is not accompanied by magnetostrictive strains; then, $\langle\mathbf{F}\rangle$ rotates toward the **a** axis. The field **H** applied along the diagonal of the *ac* plane induces a transition to the canted phase Γ_{42} and then suppresses the orientational fluctuations of $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$. The associated anomalous contribution to the magnetostriction is then small (Fig. 4). We can therefore assume that in the field $\mathbf{H}\|\mathbf{a}$ the magnetostriction is mainly due to rotation of the vectors $\langle\mathbf{F}\rangle$ and $\langle\mathbf{G}\rangle$. Then, bearing in mind that the maximum values of the magnetostriction are equal when $\mathbf{H}\|\mathbf{a}$ and $\mathbf{H}\|\mathbf{c}$, it follows from Eq. (6a) that

$$D_\theta(AS(\Gamma_4)) = \frac{\langle \delta F^2(\mathbf{r}) \rangle}{F_0} \approx \sin^2 \bar{\theta}_0, \qquad (13)$$

where $D_\theta(AS(\Gamma_4))$ is the dispersion of the fluctuations $\delta\mathbf{F}(\mathbf{r})$ and $\delta\mathbf{G}(\mathbf{r})$ in the $AS(\Gamma_4)$ asperomagnetic phase; θ_0 is the asperomagnetic angle which sets the orientation of $\langle \mathbf{F} \rangle$ in the Γ_{42} canted phase that appears in a field $\mathbf{H} \| \mathbf{a}$, $H > H_a'$ (Fig. 7a). This result is in agreement with simple qualitative ideas.

It should be pointed out that in our experiments we saw no hysteresis, i.e., a sample of $Y_{0.2}Lu_{0.8}CrO_3$ returned from the Γ_{42} canted structure to the initial asperomagnetic state $AS(\Gamma_4)$ after removal of the field, and also on rotation of $\mathbf{H}$ to the corresponding regions in the diagram of Fig. 7a. In all probability, this is due to the magnetostatic interactions. In fact, the appearance of this asperomagnetic structure results in vanishing of the projection of the magnetic moment of the sample along a direction perpendicular to $\langle \mathbf{F} \rangle$ and, consequently, provides an alternative to the formation of the usual canted structure domains [these domains cannot appear in the range where the asperomagnetic phase exists because of the absence of the long-range order of the orientations of $\mathbf{F}(\mathbf{r})$ and $\mathbf{G}(\mathbf{r})$ in this phase].

We now analyze the characteristic features of the behavior in a magnetic field of a system such as $Y_{0.1}Lu_{0.9}CrO_3$ near an SR transition. This compound exhibits an unusual angular dependence of the shear magnetostriction curve and the torque curve (Figs. 1 and 2), suggesting that rotation of a magnetic field $H = 100$ Oe in the *ac* plane gives rise to first-order SR transitions in this crystal at temperatures 82–79 K. Such transitions occur via intermediate polydomain states where the canted phase and the Γ_4 structure coexist in a field $\mathbf{H}$ lying near the **c** axis, whereas the Γ_2 phase appears for a field $\mathbf{H}$ near the **a** axis. An analysis of the phase diagram of magnetic materials with a competing anisotropy in a field $H = 0$ has shown that such states appear spontaneously as a result of loss of the long-range order by the canted phase due to large-scale fluctuations $\delta\theta(\mathbf{r})$.

We now allow for the influence of the field. Figure 7b shows the angular dependence of the critical field necessary to suppress large-scale fluctuations which give rise to intermediate domain structures (the corresponding regions are shown shaded in Fig. 7b). It is clear from Fig. 7b that rotation of a static field H results in an SR transition via intermediate polydomain states when $\varphi < \varphi_c$ and $\varphi < \varphi_a$. Observation of the corresponding linear parts of the dependence $L_{ac}(\varphi)$ and $\lambda_0(\varphi)$ makes it possible to determine the range of existence of the canted phase in $Y_{0.1}Lu_{0.9}CrO_3$, which in fact is 79–82 K.

CONCLUSIONS

Our investigations demonstrate that all the experimental results obtained for $Y_xLu_{1-x}CrO_3$ samples by the usual methods of investigation of SR processes in rare-earth perovskites cannot be explained self-consistently on the basis of a classical pattern of orientational transitions observed in ordered compounds belonging to the same class.

On the other hand, all the experimental results agree with a picture of SR processes typical of disordered magnetic materials with a competing anisotropy. This allows us to assume that the system in question has a disordered state described above. We think that the experimental methods we have developed can be used to detect and investigate stochastic magnetic structures in other disordered magnetic materials.

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[1)]The contribution of the Cr^3 ions (in the ground state $^4A_1$) to the local second anisotropy constant vanishes identically.[2]

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