

Unusual spin flip, in substituted orthochromites $Y_{1-x}Lu_xCrO_3$ with nonmagnetic rare-earth ions

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The spin-flip transitions in the compounds $Y_{1-x}Lu_xCrO_3$ with nonmagnetic rare-earth ions are observed for the first time. Their appearance is related to the presence of a competing magnetic anisotropy in the system.

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In the past, spin-flip transitions (SFT) were observed only in orthoferrites and orthochromites containing magnetic rare-earth ions under the influence of anisotropic f - d exchange. We observed SFT in the system $Y_{1-x}Lu_xCrO_3$ with nonmagnetic ions

Y^{3+} and Lu^{3+} with $x = 0.8$ and 0.9 . We shall examine below the possible mechanism for the appearance of SFT in these compounds, which does not fit into the framework of the traditional representations of the determining influence of rare-earth ions on the magnetic anisotropy and orientational transitions in rare-earth perovskites.¹

Single crystals of $Y_{1-x}Lu_xCrO_3$ were grown from a solution in a melt with lead compounds. The quantitative content of Y^{3+} and Lu^{3+} ions was determined by the method of x-ray spectral analysis. For these single crystals the torques were measured, in the temperature range 4.2–150 K on magnetic torsion scales with automatic compensation in magnetic fields up to 10 kOe. Magnetostriction along different crystallographic directions was measured in the same temperature interval with the help of a contact piezoelectric element in pulsed fields up to 80 kOe.

Curves of the dependence $M(\theta)$ (θ is the angle between the direction of weak ferromagnetic moment and the c axis), from which the anisotropy constants were determined using the relation

$$L = K_u \sin 2\theta + 4K_b \sin 4\theta,$$

were reconstructed from the experimentally measured torque curves $M(\varphi)$ (φ is the angle between the direction of the magnetic field H and the c axis of the crystal), measured in the ac plane. The correction to the angle with the transition from $M(\varphi)$ to $M(\theta)$ in fields $H \sim 10^4$ Oe, exceeding several-fold the threshold field, was not large:

$$\theta - \varphi = \arcsin \frac{M}{H(F_0 + \chi_{\perp}H)} < 10^\circ,$$

which ensured reliable determination of the anisotropy constants.

The temperature dependence of the second-order anisotropy constant K_u^{ac} , determined from the torque curves, measured at various temperatures for a composition $x = 0.9$, is shown in Fig. 1. The fourth-order anisotropy constant K_b for the given composition was small and it was difficult to determine its temperature dependence from the torque curves. At 90 K, its magnitude did not exceed 30 erg/g.

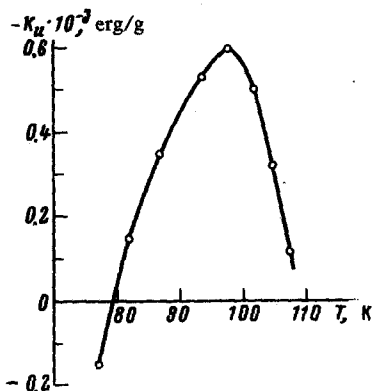


FIG. 1. Temperature dependence of the anisotropy constant in the ac plane K_u for a composition with $x = 0.9$.

We note that the anisotropy constants determined from the torque curves are related to the anisotropy constants in the expansion of the thermodynamic potential

$$\Phi = \Phi_0 + K_1 \sin^2 \theta + K_2 \sin^4 \theta$$

by the following relations:

$$K_1 = -(K_u + 8K_b); \quad K_2 = 8K_b.$$

Below the Néel point $T_N = 112$ K, the magnetic moment of this crystal is oriented along the c axis (spin configuration $\Gamma_4(G_x F_z)$, where \mathbf{F} and \mathbf{G} are the ferro- and antiferromagnetism vectors) and the anisotropy constant K_1 is positive. Near $T_r = 80$ K, the anisotropy constant, as is clearly seen from the torque curves, changes sign, indicating reorientation of the moment to the a axis: the transition $\Gamma_4 \rightarrow \Gamma_2(F_x G_z)$. This is also indicated by the field dependences of the magnetostriction, arising with the application of a magnetic field along the antiferromagnetism vector above and below T_r (Fig. 2). It is evident that for $\mathbf{H} \parallel a$ and $T > T_r$, magnetostriction due to the field-induced transition $\Gamma_4 \rightarrow \Gamma_2$ appears. Application of a magnetic field along the c axis of the crystal with these temperatures does not give rise to magnetostriction deformations. Below T_r , magnetostriction arises only for $\mathbf{H} \parallel c$ and has the opposite sign, which is related to the transition $\Gamma_2 \rightarrow \Gamma_4$ in a magnetic field.

The unusual spin flip was observed for the composition $x = 0.8$. At temperatures below $T_N = 112$ K, the magnetic moment of the crystal was oriented along the c axis and magnetostriction, due to the induced transition $\Gamma_4 \rightarrow \Gamma_2$, appeared only when $\mathbf{H} \parallel a$, indicating rotation of \mathbf{F} from the c axis to the a axis of the crystal. Below 30 K, magnetostrictions appearing with reorientation of the field along the a and c axes, were comparable in magnitude, which corresponds to orientation of the magnetic moment \mathbf{F} at an angle of 45° to the c axis in the ac plane. The temperature dependence of the angle of rotation of the spins, obtained from measurements of the magnetostriction, is shown in Fig. 3.

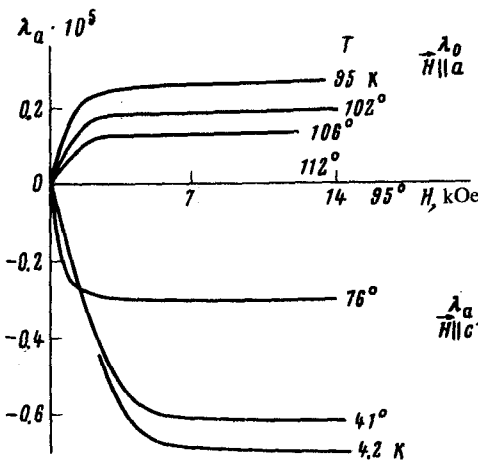


FIG. 2. Magnetostriction isotherms $\lambda_a(H)$ with the magnetic field oriented along the a and c axes, $x = 0.9$.

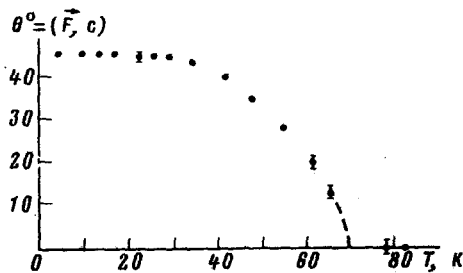


FIG. 3. Temperature dependence of the angle θ , which determines the orientation of the magnetic moment in a composition with $x = 0.8$.

The peculiarities of orientational transitions in the compositions $Y_{1-x}Lu_xCrO_3$ can be explained by viewing them as systems with competing anisotropy. The anisotropy constant of Cr^{3+} ions K_1 in $LuCrO_3$ (magnetic structure F_2) in the ac plane of the crystal is negative, in contrast to the other orthochromites, including $YCrO_3$, where $K_1 > 0$ (magnetic structure F_4).² It may be expected that at the concentration $x_c \approx 0.9$ ¹⁾ the sign of K_1 will change. In compositions with x close to x_c , due to concentration fluctuations, the probability for the existence of exchange-coupled regions with different signs of the constant K_1 is high: $K_1(r) > 0$ with $x(r) < x_c$ and $K_1(r) < 0$ with $x(r) > x_c$. Since the Néel temperature in the system $Y_{1-x}Lu_xCrO_3$ decreases monotonically with increasing x from 141 K to 112 K, it may be expected that the temperature dependence of $K_1(r)$ in these regions is also different, especially near T_N . It is logical to assume that at high temperatures $T \approx T_N$, the dominant anisotropy is the magnetic anisotropy of the regions with lower concentration $x(r) < x_c$, which have a higher ordering temperature and that the average anisotropy constant of the system (\bar{K}_1) is positive. At low temperatures, the sign of the average constant \bar{K}_1 is determined primarily by the relative volume concentration of the regions with $K_1(r) > 0$ and $K_1(r) < 0$ or, which is equivalent, with concentrations x . For $x > x_c$, $K_1(T \approx 0) < 0$, i.e., the sign of the average anisotropy constant changes with temperature, leading to spin flip.

As shown in Ref. 3, the nature of the orientational transitions in systems with competing anisotropy is strongly affected by the orientational fluctuations of the vectors F and G , caused by fluctuations in $K_1(r)$. Their contribution to the thermodynamic potential of the system leads to renormalization of the constant \bar{K}_1 and to the appearance of an effective fourth-order anisotropy $K_2 > 0$ [in pure $RCrO_3$ $K_2 \approx 0$ (Ref. 2)]. This expands the temperature range in which the spin flip occurs and could be the reason for the appearance of the stable angular orientation of spins observed in a composition with $x = 0.8$.

¹⁾The estimate of x_c was obtained from the data on the anisotropy constants of $YCrO_3$, $LuCrO_3$, and $Y_{0.5}Lu_{0.5}CrO_3$ ² at $T = 0K$.

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³E. V. Sinitsyn and I. B. Bostrem, *Zh. Eksp. Teor. Fiz.* **85**, 661 (1983) [*Sov. Phys. JETP*, to be published].

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