Features of *f–d* exchange in weak ferrimagnets

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Some features of the polarization of rare-earth ions due to the weakly ferrimagnetic nature of the *d*-ion ordering in the $GdFe_{1-x}Cr_xO_3$ system are investigated experimentally and theoretically. It is shown that the antiparallel orientation of the magnetizations of the Fe and Cr sublattices results in the appearance of two points of concentration compensation of the effective fields at the Gd^{3+} ions (reversal of the field direction). The contributions to the effective field from the isotropic and anisotropic *f–d* exchange are determined. It is shown that the principal role in the polarization of the rare-earth subsystem is played by the antisymmetric Gd–Fe and Gd–Cr exchange, even though the interaction ions are of the *S* type.

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1. INTRODUCTION

The discovery of transverse weak ferrimagnetism,[1] a new type of magnetic ordering typical of weak ferromagnets with mixed Dzyaloshinskiĭ interaction, has not only demonstrated for the first time the distinguishing features of the manifestation of antisymmetric exchange, connected with the sign of the Dzyaloshinskiĭ vector, but also stimulated the investigation of a new class of magnetic materials, week ferrimagnets that have a number of anomalous magnetic properties: the appearance of points of concentration and temperature compensation of the magnetic moment, and a Néel-type temperature dependence of the magnetization.

One of the important properties of weak ferrimagnets is the possibility of quantitatively analyzing the transverse magnetic structure within the framework of the simplest molecular-field theory, owing to the smallness of the Dzyaloshinskiĭ interaction compared with the isotropic exchange that determines the basic antiferromagnetic structure of these compounds.

Transverse weak ferrimagnetism, first observed in mixed yttrium orthoferrites--orthochromites $YFe_{1-x}Cr_xO_3$ (Ref. 1), constitutes antiparallel orientation of the average weakly ferromagnetic moments of the *d*-sublattices (Fe and Cr), owing to the difference in the signs of the Dzyaloshinskiĭ vectors in the pairs Fe–Fe, Cr–Cr, and Fe–Cr.

Figure 1 shows plots, calculated by us by the procedure of Ref. 2, of the concentration dependences of the average magnetic moments of the Fe and Cr sublattices in $YFe_{1-x}Cr_xO_3$, as well as the summary moment of the *d* sublattice. The $\sigma_{Fe}(x)$ and $\sigma_{Cr}(1-x)$ dependences are similar: increasing the impurity concentration decreases the magnetization of the matrix, and the magnetization of the impurity system increases strongly with increasing impurity concentration. Except for a small region $x \approx 0.5$–0.7, the magnetic moments of the Fe and Cr sublattices are antiparallel. The original character of the behavior of the sublattice magnetizations (the presence of a minimum and the sign reversal) leads to an anomalous dependence on the concentration and on the total magnetic moment, a dependence characterized by a steep drop of the magnetization with increasing x or $(1-x)$, by the appearance of two compensation points, and also by the appearance of a minimum in the region $x \approx 0.5$. The temperature dependences of the magnetization of $YFe_{1-x}Cr_xO_3$ have the Néel character typical of ferrimagnets.[2]

It is undoubtedly of interest to investigate the influence of the weak ferrimagnetic ordering of the *d* sublattice on the behavior of the rare-earth subsystems in compounds of the type $RFe_{1-x}Cr_xO_3$. The character of polarization of the *R*-ions and its concentration and temperature dependences can yield valuable information not only on the state of the *d* subsystem, but also on the *f–d* interaction mechanisms, primarily on the relative roles of the ferro- and antiferromagnetic contributions to the effective field at the *R* ions.

The first attempt to analyze the magnetic properties of rare-earth weak ferrimagnets, with a number of single crystals of the $NdFe_{1-x}Cr_xO_3$ as an example, was made in Ref. 4. The premise that the *d* sublattice has antiferromagnetic ordering and that the predominant contribution to the po-

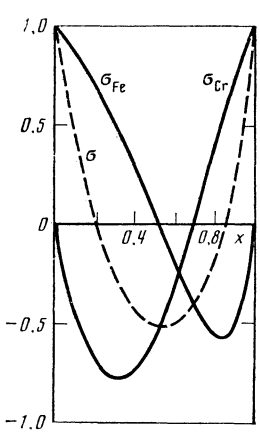

FIG. 1. Concentration dependence of the magnetizations of the Fe and Cr sublattices and of the total magnetization of the weak ferrimagnet $YFe_{1-x}Cr_xO_3$ at $T = 0$ K.

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larization is made by the Nd^{3+} ions enabled the authors of Ref. 4 to explain qualitatively some features of the magnetic behavior of a number of $NdFe_{1-x}Cr_xO_3$ compounds.

Of particular interest, in our opinion, is the $GdFe_{1-x}Cr_xO_3$ system with *S*-type *f* and *d* ions, where it might seem that it is precisely the ferromagnetic contribution due to the isotropic *f–d* exchange which should play the decisive role in the polarization of the Gd sublattice. At the same time, a detailed analysis of the magnetic properties of $GdFeO_3$ (Ref. 5) has revealed the substantial role of the anisotropic exchange of the *S*-ions Gd^{3+} and Fe^{3+} and accordingly of the antiferromagnetic contribution of the Gd sublattices to the polarization.

Additional arguments favoring this conclusion, and also an answer to the question of the role of anisotropic interactions of the *S* ions Gd^{3+} and Cr^{3+}, can be provided by an analysis of the behavior of the weak ferrimagnets $GdFe_{1-x}Cr_xO_3$.

In the present study we solved the comprehensive problem of experimentally and theoretically investigating the features of the magnetic properties of $GdFe_{1-x}Cr_xO_3$ with an aim at studying the influence of the weak ferrimagnetic ordering of the *d* sublattice on the magnetic state of the rare-earth subsystem and the role of the isotropic and anisotropic *f–d* exchange of the S ions Gd^{3+} with Fe^{3+} and Cr^{3+}.

2. EXPERIMENTAL RESULTS

Single crystals with a number of $GdFe_{1-x}Cr_xO_3$ compositions were grown from the solution in a melt of lead compounds. To determine the temperature dependence of the spontaneous magnetization we measured with a torsion anisometer the torque curves which were automatically recorded in the temperature range from 2 to 700 K.

Figure 2 shows the temperature dependences of the spontaneous magnetizations σ_c for compositions with $x = 0$ and 0.05. It can be seen that introduction of even a small amount of Cr^{3+} impurity ions decreases strongly the magnetization of the *d* ions, thus indicating directly the presence of transverse weak ferrimagnetism. For the compound $GdFe_{0.95}Cr_{0.05}O_3$ we observed also a strong decrease of the polarization of the rare-earth ions compared with $GdFeO_3$,

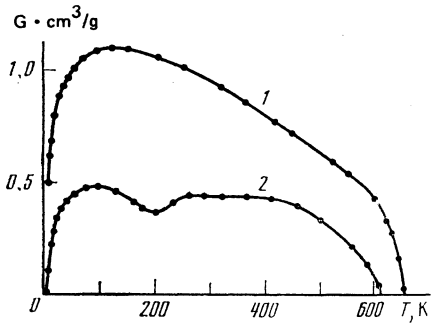

FIG. 2. Temperature dependence of the spontaneous magnetization of $GdFeO_3$ (1) and of the weak ferrimagnet $GdFe_{0.95}Cr_{0.05}O_3$ (2).

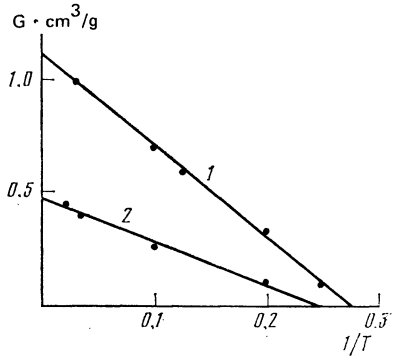

FIG. 3. Dependence of σ on $(1/T)$ for $GdFe_{1-x}Cr_xO_3$ at $x = 0$ (1) and 0.05 (2).

as is particularly clearly seen from the dependence of σ on $1/T$ (Fig. 3), which is linear for both compositions. The effective magnetic field H_c acting on the Gd^{3+} ions was determined from the slope of the $\sigma(1/T)$ line and found to be -150 Oe in $GdFe_{0.95}Cr_{0.05}O_3$ or half the value in $GdFeO_3$. In view of the negative H_c, compensation points were observed in the substituted compound, just as in $GdFeO_3$.

A qualitatively different behavior was observed for the composition with $x = 0.17$ (Fig. 4), where a hyperbolic increase of the magnetization was observed at low temperatures. This indicates reversal of the sign of the polarization of the rare-earth ions. As can be seen from Fig. 4, the low-temperature increase is preceded by a noticeable drop of the magnetization in the temperature region below 80 K. It is obviously due to the peculiarities of the temperature dependence of the *d*-sublattice magnetization and to the weak ferrimagnetic ordering, particularly to the Néel character of the $\sigma(T)$ dependence.[1,2]

An antiparallel orientation of the weak magnetic moments of the Fe and Cr sublattices in the $GdFe_{1-x}Cr_xO_3$ system is indicated also by the steep drop of the magnetization when a small amount of Fe^{3+} ions is introduced in $GdCrO_3$ (Fig. 5). This fact agrees both with the experimental data on the $YFe_{1-x}Cr_xO_3$ system,[1] and with the theoretical premises developed in Ref. 1.

A spin-reorientation transition $G_xF_z \rightarrow G_zF_x$ was observed at low temperatures in the compound $Gd_{0.1}Cr_{0.9}O_3$,

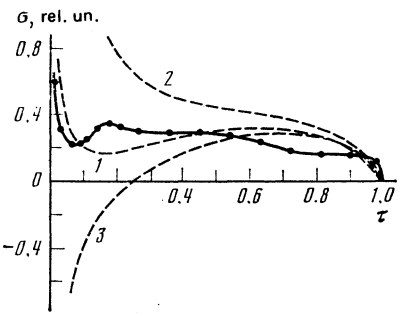

FIG. 4. Temperature dependence of the spontaneous magnetization of the weak ferrimagnet $GdFe_{1-x}Cr_xO_3$; $\tau = T/T_N$, with $T_N = 550$ K, $x = 0.17$. Points and solid curve—experimental data, dashed curves—calculation results: $1 - \alpha_{Cr} = -1.3$ kOe, $\beta_{Cr}^{as} = -4.7$ kOe; $2 - \alpha_{Cr} = -6$ kOe, $\beta_{Cr}^{as} = 0$; $3 - \alpha_{Cr} = 0$, $\beta_{Cr}^{as} = -6$ kOe.

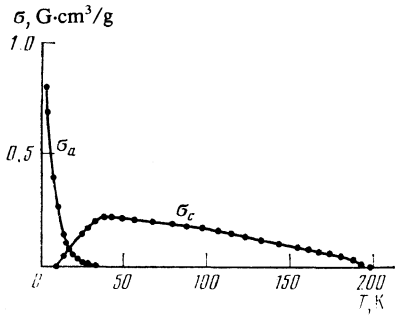

FIG. 5. Temperature dependence of the spontaneous magnetization of the weak ferrimagnet $GdFe_{0.1}Cr_{0.9}O_3$.

just as in the pure gadolinium orthochromite.[6] This transition was shifted towards higher temperatures. With decreasing temperature, the magnetization along the a axis increased hyperbolically, but the polarization of the Ge^{3+} ions turned out to be substantially lower than in pure $GdCrO_3$.

3. f–d EXCHANGE IN THE WEAK FERRIMAGNET $Gd_{1-x}Cr_xO_3$. DISCUSSION OF EXPERIMENTAL DATA

When account is taken of the isotropic and anisotropic f–d interactions, the effective field at the R ion, say in $GdFeO_3$, can be represented as a sum of ferro- and antiferromagnetic contributions[3,4]:

$$\mathbf{H}=\alpha_{Fe}\boldsymbol{\sigma}_{Fe}+\hat{\beta}_{Fe}\mathbf{l}_{Fe}, \tag{1}$$

where α_{Fe} determines the contribution of the isotropic f–d exchange, $\hat{\beta}_{Fe}$ is the tensor of the parameters of the symmetric and antisymmetric anisotropic f–d interactions, $\boldsymbol{\sigma}_{Fe}$ and $\mathbf{l}_{Fe}$ are respectively the magnetization and the antiferromagnetism vector of the Fe sublattice.

Generalizing this expression to the case of rare-earth weak antiferromagnets of the $GdFe_{1-x}Cr_xO_3$ type, we obtain, with the magnetic configuration Γ_4 (G_xF_z) as an example,

$$H_c=\alpha_{Fe}\bar{\mu}_{Fe}(x,T)+\alpha_{Cr}\bar{\mu}_{Cr}(x,T)+\beta_{Fe}\bar{l}_{Fe}(x,T)+\beta_{Cr}\bar{l}_{Cr}(x,T), \tag{2}$$

where $\bar{\mu}_{Fe,Cr}$ is the average magnetic moment of the Fe or Cr sublattice, normalized for convenience to the magnetization of the Fe or Cr sublattice in the pure orthoferrite (orthochromite) at $T=0$ K, i.e, $\bar{\mu}_{Fe}(0,0)=1$ and $\bar{\mu}_{Cr}(1,0)=1$. The mean value of the antiferromagnetism vector $\bar{l}_{Fe,Cr}$ is simply connected with the mean value of the longitudinal component of the spin of the ions Fe^{3+} and Cr^{3+} at a small noncollinearity of the spins (for the sake of argument, we take the antiferromagnetism-vector components to be positive):

$$\bar{l}_{Fe}=\frac{\bar{S}_{Fe}(x,T)}{S_{Fe}(0,0)}(1-x), \quad \bar{l}_{Cr}=\frac{\bar{S}_{Cr}(x,T)}{S_{Cr}(1,0)}x, \tag{3}$$

with

$$\bar{l}_{Fe}(x,0)\approx 1-x, \quad \bar{l}_{Cr}(x,0)\approx x.$$

Thus, the unique behavior of the magnetic moments of the d sublattices of a weak ferrimagnet leads also to an unusual behavior of the effective fields at the R ions.

According to the data of Ref. 5, whose authors analyzed the isotropic and anisotropic interactions in $GdFeO_3$,

$$\alpha_{Fe}=-1.9 \text{ kOe}, \quad \beta_{Fe}=1.6 \text{ kOe},$$

and a substantial contribution to the parameter of the anisotropic interactions is made by the antisymmetric Gd–Fe exchange

$$\beta_{Fe}=\beta_{Fe}^{s}+\beta_{Fe}^{as}=(-0.5+2.1) \text{ kOe}.$$

The situation is more complicated with the analysis of the f–d interaction in $GdCrO_3$. The data of Ref. 6 make it possible to estimate the sum $\alpha_{Cr}+\beta_{Cr}=-5.5$ kOe, and also the contribution of the symmetric anisotropic Gd–Cr exchange, i.e., $\beta_{Cr}^{s}=+0.5$ kOe.

Definite information on the parameter α_{Cr} of the isotropic Gd–Cr exchange can be obtained from the data of Ref. 7, whose authors investigated the Gd^{3+}–Cr^{3+} interaction in the orthoaluminate $GdAlO_3$ with Cr^{3+} admixture, which is isostructural to $GdCrO_3$. The exchange integral $I_{GdCr}\approx 1.6$ K which they obtained with allowance for the cant angle of the magnetic sublattices Cr in $GdCrO_3$ (equal, according to Ref. 6, to 2.5×10^{-2}), would correspond to $\alpha_{Cr}\approx -6$ kOe. However, the data of Ref. 8 on the influence of Al^{+} ions on the substantial increase of the effective field in $GdFe_{1-x}Al_xO_3$ is evidence of the possibility of a noticeable decrease of the average exchange integral I_{GdCr} in $CdCrO_3$ compared with I_{GdCr} in $GdAlO_3$:Cr^{3+}. That the isotropic Gd–Cr exchange makes a relatively small contribution to the Gd-sublattice polarization in $GdCrO_3$ was indicated also by the authors of Ref. 6. As for the antisymmetric Gd–Cr exchange the theoretical results of Refs. 5 and 9 allow us to conclude that β_{Fe}^{as} and β_{Cr}^{as} are close in magnitude but are of opposite sign.

Obviously, investigation of the magnetic properties of the weak ferrimagnets $GdFe_{1-x}Cr_xO_3$ can yield more detailed information on the relative roles of the isotropic and antisymmetric Gd–Cr exchange to the polarization of the rare-earth sublattice. Thus, at low temperatures and small concentrations x, the $\bar{\mu}_{Fe}(x)$ and $\bar{\mu}_{Cr}(x)$ dependences can be approximated by a linear law, where according to Ref. 1

$$\bar{\mu}_{Fe}(x)=1-x, \quad \bar{\mu}_{Cr}(x)=(S_{Cr}/S_{Fe})(2\delta-1)x, \tag{4}$$

where the parameter δ is determined by the ratio of the Dzyaloshinskiĭ-interaction parameters d_{CrFe} and d_{FeFe} and of the exchange integrals I_{CrFe} and I_{FeFe} (Ref. 1):

$$\delta=(d_{CrFe}/I_{CrFe})(d_{FeFe}/I_{FeFe})^{-1}. \tag{5}$$

Thus, the concentration dependence of the effective field at the Gd^{3+} ions at small x takes the form

$$\begin{aligned} H_c(x)&=(\alpha_{Fe}+\beta_{Fe})(1-x)+[^3/_5(2\delta-1)\alpha_{Cr}+\beta_{Cr}]x \\ &=-0.3-[6.4\alpha_{Cr}+5.2]x=-0.3+[33.2+6.4\beta_{Cr}^{as}]x, \end{aligned} \tag{6}$$

where we have used the values cited above for the parameters α and β, as well as the value $\delta=-4$ which is typical of the antisymmetric exchange in the d sublattice of $YFe_{1-x}Cr_xO_3$ (Ref. 2).

Reversal of the sign of H_c, meaning also of the direction of the Gd-sublattice polarization, takes place in our case at the critical concentration x_{cr}:

$$x_{cr}=-0.3/(6.4\alpha_{Cr}+5.2)=0.3/(33.2+6.4\beta_{Cr}^{as}). \quad (7)$$

We note that, as can be seen from Fig. 1, the validity of the linear law (4) and of relations (6) and (7), is restricted to a concentration $x_{cr}\approx 0.05$.

Particular interest attaches to the fact that in the absence of antisymmetric Gd–Cr exchange (i.e., at $\beta_{cr}^{as}=0$) the reversal of the sign of the polarization of the Gd sublattice should occur already at very low concentrations of the Cr^{3+} ions, namely at $x_{cr}\sim 0.01$. Actually, however, our experimental data (Fig. 2) indicate that $x_{cr}>0.05$ in $GdGe_{1-x}Cr_xO_3$. This result is not only experimental proof of the very existence of antisymmetric exchange of the S ions Gd^{3+} and Cr^{3+}, but makes possible numerical estimates of α_{Cr} and β_{Cr}^{as}. From the condition $x_{cr}>0.05$ we obtain at negative values of α_{Cr} and β_{Cr}^{as} the following inequalities:

$$|\alpha_{Cr}|<1.8\ \text{kOe},\quad |\beta_{Cr}^{as}|>4.2\ \text{kOe}$$

Attesting to negative α_{Cr}, meaning also β_{Cr}^{as}, is also the experimental fact that the effective field H_c is weaker at $x=0.05$ than H_c in pure $GdFeO_3$. Moreover, the experimentally established value $H_c(0.05)=-0.15$ kOe makes it possible to determine more accurately the values of α_{Cr} and β_{Cr}^{as}. On the basis of Eq. (6) we obtain

$$\alpha_{Cr}\approx -1.3\ \text{kOe},\quad \beta_{Cr}^{as}\approx -4.7\ \text{kOe}.$$

Thus, the antisymmetric Gd–Cr f–d exchange makes the largest contribution to the polarization of the Gd sublattice of the rare-earth weak ferrimagnets $GdFe_{1-x}Cr_xO_3$ compared with the isotropic Gd–Cr exchange and the symmetric anisotropic Gd–Cr interaction. The sign of the parameter β_{Cr}^{as} turns out to be opposite to the sign of β_{Fe}^{as}, in full accord with the conclusions of the microscopic theory.[5,9] The values of the Gd–Cr and Gd–Fe isotropic exchange turn out to be close.

Knowledge of the numerical values of the parameters $\alpha_{Fe,Cr}$ and $\beta_{Fe,Cr}$ has enabled us to calculate, within the framework of the molecular-field theory, the concentration and temperature dependences of the average effective field H_c, the magnetization of the Gd sublattice, and the total magnetization of $GdFe_{1-x}Cr_xO_3$ in the entire range of variation of x. The exchange integrals and the Dzyaloshinskiĭ-interaction parameters in the d sublattices were chosen equal to the corresponding values for $YFe_{1-x}Cr_xO_3$,[1,2] and the susceptibility of the Gd^{3+} ions was taken in accord with the data of Ref. 10.

Figure 6 shows the calculated concentration dependence of $H_c(x)$ at $T=0$ K; it has an unusual form with two compensation points—at small and relatively large concentrations of the Cr^{3+} ions.

Figure 7 shows the calculated temperature dependence of the total magnetization of $GdFe_{1-x}Cr_xO_3$ for a successive series of concentrations x. Whereas at $x\approx 0.05$ compensation of the total magnetic moment is still observed, at $x=0.10$ the reversal of the sign of Hc leads to a hyperbolic increase of $\sigma(T)$ in the low-temperature region. The effect due to the features of the weak ferrimagnetic ordering of the d sublattice, namely the appearance of a minimum on the $\sigma(T)$ curve in the temperature region preceding the hyperbolic growth of the magnetization, manifests itself here first

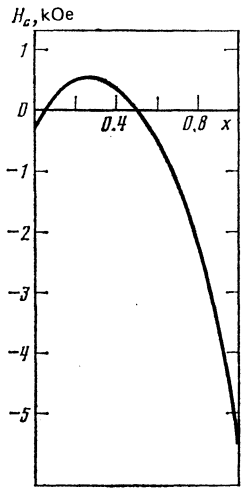

FIG. 6. Calculated concentration dependence of the average effective field at the Gd^{3+} ions in the weak ferrimagnet $GdFe_{1-x}Cr_xO_3$ at $T=0$ K.

weakly, and becomes more and more pronounced with further increase of x. This is a consequence of the Néel-type temperature dependence of the magnetization of the d sublattice of the weak ferrimagnet $GdFe_{1-x}Cr_xO_3$ (dependence of type a in the notation of Ref. 1) at positive polarization of the Gd sublattice.

This effect is very clearly seen on the experimental $\sigma(T)$ curve for the composition with $x=0.17$ (Fig. 4). We have shown in the same figure the calculated curve at the values of α_{Cr} and β_{Cr} obtained by us (curve 1), as well for a comparison of the results of the calculation assuming absence of antisymmetric Gd–Cr exchange ($\beta_{Cr}^{as}=0$, curve 2) or of isotropic Gd–Cr exchange ($\alpha_{Cr}=0$, curve 3). The satisfactory agreement between the theory (curve 1) and the experiment is one more proof of the reliability of our conclusions concerning the relative role of the isotropic and anisotropic Gd–Cr interaction.

Even more interesting effects should result from further increase of the concentration x. The appearance of a point of compensation of the d-sublattice magnetization together

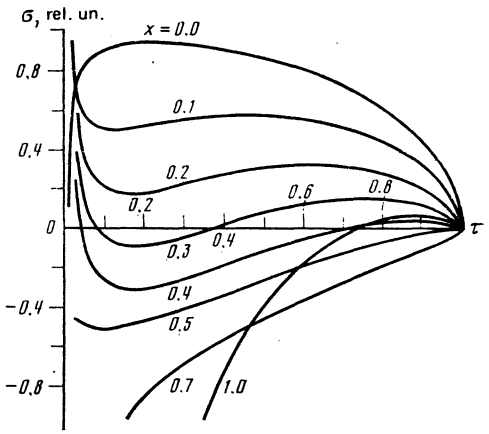

FIG. 7. Calculated temperature dependences of the total magnetization of a number of weak ferromagnets $GdFe_{1-x}Cr_xO_3$, $\tau=T/T_N$.

with the positive polarization of the Gd^{3+} ions results in two compensation points.

Calculation shows that at $x \approx 0.27$ there appears in the region $\tau \approx 0.17$ a compensation point that subsequently doubles, with one (high-temperature) compensation point moving towards T_N with increasing x, and the other (low temperature) towards $T = 0$ K. At $x > 0.5$ the compensation points vanish. Only for compositions directly adjacent to pure gadolinium orthochormite is the compensation again observed, and with increasing concentration of the Fe^{3+} ions the compensation point shifts from $T_k = 110$ K in pure $GdCrO_3$ to $T_k = T_N$ at $x \approx 0.95$.

The calculated temperature dependences of the magnetization of $GdFe_{1-x}Cr_xO_3$ at large values of x agree satisfactorily with the experimental data, for example with those for the composition $GdFe_{0.1}Cr_{0.9}O_3$ investigated by us (Fig. 5). Naturally, in this case the experimental values of $\sigma(T)$ must be taken with a minus sign.

4. CONCLUSION

The weak ferrimagnetic character of the ordering of the sublattice of the rare-earth orthoferrites-orthochromites $RFe_{1-x}Cr_xO_3$, due to the different signs of the Dzyaloshinskiĭ vectors in the pairs Fe–Fe, Cr–Cr, and Fe–Cr and manifesting itself in an antiparallel orientation of the average magneticmoments of the Fe and Cr sublattices, leads to an anomalous concentration and temperature dependence of the effective magnetic field acting on the R ions, and in particular to the appearance of two points of concentration compensation (sign reversal) of the field.

The qualitative and quantitative analysis of the magnetic properties of a number of single crystals of the system $GdFe_{1-x}Cr_xO_3$ has made it possible not only to explain the unusual character of the concentration and temperature dependences of the magnetization, but also to obtain important information on the magnitude of the isotropic and anisotropic *f–d* Gd–Cr exchange. Thus, it has been shown that the isotropic Gd–Cr exchange is antiferromagnetic and close in magnitude to the Gd–Fe exchange. Of greatest interest is the conclusion that the main contribution to the polarization of the Ge^{3+} ions, from both the Fe and Cr sublattices, is made by the antisymmetric Gd–Fe and Gd–Cr exchanges. This fact is due entirely to the suppression of the potentially large isotropic *f–d* exchange in the pairs Gd–Fe and Gd–Cr at the basic antiferromagnetic structure of the *d* sublattice,[5] so that relatively weak anisotropic interactions of the S ions Gd^{3+} with Fe^{3+} and Cr^{3+} assume the principal role in inducing the effective field at the rare-earth ions in $GdFe_{1-x}Cr_xO_3$.

Finally, we note the need for further experimental investigation of the rare-earth weak ferrimagnets $RFe_{1-x}Cr_xO_3$ both from the viewpoint of studying various *f–d* interactions, and of the possibility of obtaining better magnetic properties than in pure $RFeO_3$ and $RCrO_3$. From this viewpoint, interest attaches to our prediction that compositions with two magnetization compensation points can occur in the $GdFe_{1-x}Cr_xO_3$ system.

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