

*THE MAGNETIC SUSCEPTIBILITY OF RARE-EARTH ORTHOFERRITES IN STRONG  
MAGNETIC FIELDS*

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The magnetization curves of a series of rare-earth orthoferrites in poly- and monocrystalline form were measured in pulsed magnetic fields up to 220 kOe. An analysis of the magnetic susceptibilities of the orthoferrites studied shows that at room temperature the exchange fields of the iron sublattices (as well as other crystal fields) do not affect the magnetic moments of the rare-earth ions. Thus, at the indicated temperatures the rare-earth ions in orthoferrites can be considered to be free in the magnetic sense.

1. The orthoferrites of the rare-earth elements (general formula  $MFeO_3$ , where M is an ion of a rare-earth metal) are interesting because they are antiferromagnetic substances displaying weak ferromagnetism.

A number of authors<sup>[1]</sup> have shown that the appearance of weak ferromagnetism in these compounds is connected with the emergence of non-colinearity of the magnetic moments of the iron ions under the influence of perturbing internal fields of the crystal.

According to neutron diffraction investigations, only the magnetic moments of the iron ions are oriented in the orthoferrites at room temperature; ordering of the magnetic moments of the rare-earth ions occurs only at a temperature below 20°K.<sup>[2]</sup> Nevertheless, according to<sup>[3]</sup>, exchange interaction can exist between the iron ions and the rare-earth ions even above the ordering temperature.

2. Pauthenet and Bloom<sup>[4]</sup> have established that the field dependence of the magnetization of the rare-earth orthoferrites in magnetic fields above 6000 Oe obeys the following relation:

$$\sigma = \sigma_0 + \chi H, \quad (1)$$

where  $\sigma_0$  is the specific spontaneous magnetization and  $\chi$  is the specific susceptibility.

However, for polycrystalline samples of the orthoferrites, as our measurements have shown, this relation is not fulfilled in fields up to 20 kOe, for as a consequence of the fine dispersion of the structure the coercive force of the individual grains exceeds the magnetizing field.<sup>[5]</sup>

There is an interest in more detailed studies of the magnetic properties of rare-earth ortho-

ferrites and, in particular, in the establishment of the contributions of the iron and rare-earth ions to the susceptibility. We have carried out measurements of the field dependence of the magnetization of the orthoferrites of La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, and Yb. Since we investigated polycrystalline samples along with single crystals, the measurements were carried out in strong pulsed fields up to 220 kOe.

The magnetization was measured by a ponderomotive method—by the pull on a small sample (10–100 mg) in an inhomogeneous magnetic field. An electromagnetic pickup was used for the measurement of the pull, and its signal was applied after differentiation, to a pulse oscillograph. We have described the method of measurement in more detail elsewhere.<sup>[6]</sup> The error in measuring the susceptibility by this method does not exceed 10%.

The polycrystalline samples were prepared by the usual ceramic technology<sup>[7]</sup>; the monocrystals were grown by spontaneous crystallization from solution.<sup>[8]</sup>

3. In Fig. 1 are shown the magnetization curves for the polycrystalline orthoferrite samples. As can be seen from the figure, the field dependence of the magnetization obeys the relation (1) for all compositions.

In order to be able to compare the contributions of the rare-earth and the iron ions, we made measurements on the orthoferrite  $LaFeO_3$ , in which the  $La^{3+}$  ions is non-magnetic. In this case it is obvious that the susceptibility is due to the interaction of the field with the antiferromagnetic structure formed by the magnetic moments of the  $Fe^{3+}$  ions. As is seen from Fig 1,  $LaFeO_3$  has

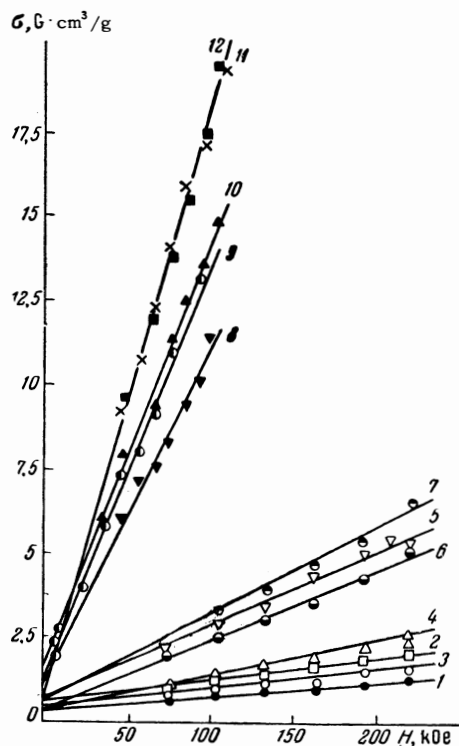


FIG. 1. Field dependence of the specific magnetization of polycrystals of the orthoferrites: 1 -  $\text{LaFeO}_3$ , 2 -  $\text{LaSc}_{0.1}\text{Fe}_{0.9}\text{O}_3$ , 3 -  $\text{LaAl}_{0.2}\text{Fe}_{0.8}\text{O}_3$ , 4 -  $\text{La}_{0.95}\text{Gd}_{0.05}\text{FeO}_3$ , 5 -  $\text{PrFeO}_3$ , 6 -  $\text{NdFeO}_3$ , 7 -  $\text{YbFeO}_3$ , 8 -  $\text{GdFeO}_3$ , 9 -  $\text{TbFeO}_3$ , 10 -  $\text{ErFeO}_3$ , 11 -  $\text{DyFeO}_3$ , 12 -  $\text{HoFeO}_3$ .

the smallest susceptibility. Upon replacement of a small portion of the  $\text{La}^{3+}$  ions by  $\text{Gd}^{3+}$  ions, which possess a large magnetic moment, the susceptibility increases markedly. The orthoferrites of Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, and Yb, in which the rare-earth ions all have magnetic moments, also have a markedly greater susceptibility than the  $\text{LaFeO}_3$ . This indicates that the principal contribution to the susceptibility of the orthoferrites comes from the rare-earth ions.

A similar kind of dependence of  $\sigma$  on  $H$  is also observed for monocrystals of the orthoferrites of La, Pr, Nd, Sm, Eu, Gd, and Yb (Fig. 2), and the values of the susceptibilities of the monocrystals are in good agreement with the values for the corresponding polycrystalline samples. The field dependence for all monocrystals was taken along the axis of weak ferromagnetism (the  $a$  axis for Sm, the  $c$  axis for all the others).

The values of the molar susceptibility were calculated from the experimental data for all the orthoferrites investigated. Subtracting from the molar susceptibility of the orthoferrites with magnetic rare-earth ions the susceptibility of  $\text{LaFeO}_3$ , which, as indicated above, is due to

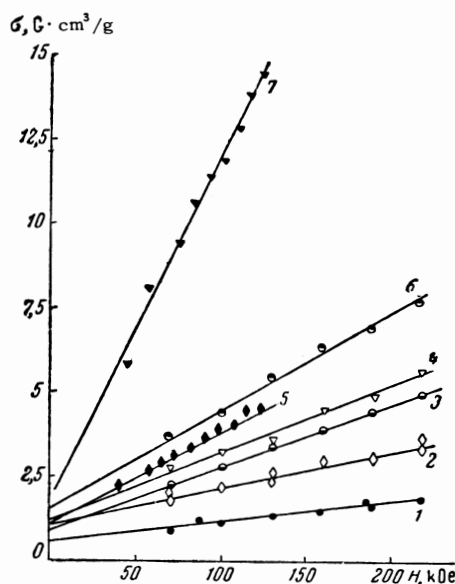


FIG. 2. Field dependence of the specific magnetization of monocrystals of the orthoferrites: 1 -  $\text{LaFeO}_3$ , 2 -  $\text{SmFeO}_3$ , 3 -  $\text{NdFeO}_3$ , 4 -  $\text{PrFeO}_3$ , 6 -  $\text{EuFeO}_3$ , 6 -  $\text{YbFeO}_3$ , 7 -  $\text{GdFeO}_3$ .

iron ions, we obtained the susceptibility of the rare-earth ions in orthoferrites at room temperature. It was assumed here that the values of the susceptibility of the iron sublattices were the same for all the orthoferrites. (It should be kept in mind that the exchange interaction between the iron ions in orthoferrites apparently decreases with increasing atomic number of the rare-earth element. Evidence for this is that the Curie point of lanthanum orthoferrite is  $100^\circ$  higher than the Curie point of ytterbium orthoferrite.) In order to test our assumption, we made measurements of the susceptibility on some substituted compo-

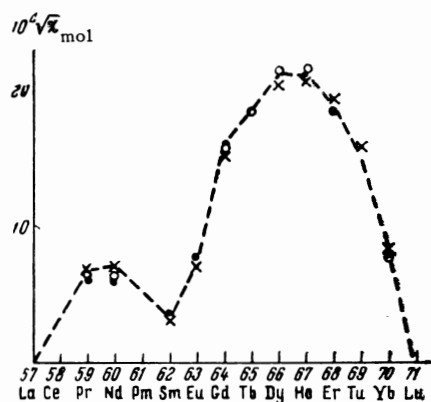


FIG. 3. Dependence of the molar susceptibility of ions of rare-earth elements in orthoferrites and oxides on their atomic number: open circles - orthoferrites (polycrystals), filled circles - orthoferrites (monocrystals), crosses - rare-earth oxides; dashed line - theoretical curve.

sitions, in which a portion of the  $\text{Fe}^{3+}$  ions in the lanthanum orthoferrite was replaced by non-magnetic ions  $\text{Al}^{3+}$  and  $\text{Sc}^{3+}$ . Although the Curie points of these substituted compositions differed among themselves by more than  $100^\circ$ ,<sup>[8]</sup> no significant change in the susceptibility was observed (Fig. 1).

Figure 3 shows the values of  $(\chi_{\text{mol}})^{1/2}$  calculated in this fashion for the rare-earth ions in orthoferrites as a function of the atomic number of the rare-earth element. (The quantity  $(\chi_{\text{mol}})^{1/2}$  is proportional to the effective magnetic moment  $\mu_{\text{eff}}$  of the ion.) In the same figure are shown the theoretical values of  $(\chi_{\text{mol}})^{1/2}$  for the free trivalent rare-earth ions calculated from the Van Vleck theory,<sup>[9]</sup> as well as the experimental values of this quantity for ions of the rare earths in oxides.<sup>[10]</sup>

As is seen from Fig. 3, there is good agreement between the molar susceptibilities we have obtained for rare-earth ions in orthoferrites and the theoretically computed values for the free trivalent ions.

This offers evidence that at room temperature there is practically no effect of the exchange field of the iron sublattices on the magnetic moments of the rare-earth ions in orthoferrites, and that no "freezing" of the magnetic moments of these ions by the crystalline field of the orthoferrite lattice occurs. Thus, the ions of rare-earth elements in orthoferrites can be considered free in the magnetic sense.

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