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DETERMINATION OF EXCHANGE INTERACTION OF SUBLATTICES IN GADOLINIUM IRON-GARNET ON THE BASIS OF THE MAGNETOCALORIC EFFECT

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On the basis of measurements of the magnetocaloric effect, the paraprocess susceptibility, and the heat capacity, in the region of temperature compensation, the effective exchange field $H_{2\text{eff}}$ is determined, which acts on the Gd^{3+} ions from the Fe^{3+} ions in the garnet structure. The measurements showed that for $Gd_3Fe_5O_{12}$ garnet the field is $H_{2\text{eff}}$ = 258 kOe, whereas for $Gd_3Ga_{0.3}Fe_{4.7}O_{12}$ it is $H_{2\text{eff}}$ = 232 kOe, which is 10% smaller than for gadolinium garnet.

IRON-garnets of the rare earths, $R_3Fe_5O_{12}$, have, as is well known, a three-sublattice magnetic structure. In the majority of cases, within the framework of the molecular-field theory, such iron garnets may be treated as two-sublattice. The two iron sublattices a and d are considered as a single sublattice Fe_{a-d}[1], in whose effective field H_{eff} are the ions of the rare earth. We have carried out a determination of the effective exchange field exerted by the Fe^{3+} ions on the R^{3+} ions.

In iron-garnets of the yttrium subgroup, possessing a compensation temperature T_c, on application of an external magnetic field in the temperature range $T < T_c$, as is well known, a magnetic moment of the rare-earth c-sublattice is established along the external magnetic field, whereas the moment of the iron Fe_{a-d} sublattice is established opposite to the field. Measurements of the magnetocaloric effect have shown[2,3] that in this temperature range there occurs in the rare-earth sublattice a paraprocess of ferromagnetic type[2,3], for which the magnetocaloric effect is positive ($\Delta T_2 > 0$ for $T < T_c$; ΔT_2 is the magnetocaloric effect due to the rare-earth sublattice). But in the iron sublattice, in this temperature range, there occurs a paraprocess of antiferromagnetic type[2].

In the temperature range $T > T_c$, the moment of the iron sublattice is aligned along the field, the moment of the rare-earth sublattice opposite to the field. In the rare-earth sublattice, in this range of temperature, there occurs a paraprocess of antiferromagnetic type: the external field, in turning the magnetic moments of the Gd^{3+} ions toward the field, simultaneously establishes an effective exchange field opposite to them, and this is accompanied by absorption of heat[2,3].

For the magnetocaloric effect ΔT_2 due to the rare-earth sublattice of iron garnets of the yttrium subgroup, one can write the expression[4]

$$\Delta T_2 = \frac{2s_2}{g_J J_2 C_v} H_{2\text{eff}}\, \chi_m H,$$

where $H_{2\text{eff}}$ is the effective exchange field exerted by the Fe^{3+} ions on the R^{3+} ions, s_2 is the spin of the rare-earth ion, J_2 is the total magnetic moment of the rare-earth ion, χ_m is the molar paraprocess susceptibility of the rare-earth sublattice, and C_v is the specific heat of the garnet.

We shall consider the simplest case, $Gd_3Fe_5O_{12}$. For a Gd^{3+} ion in an s-state, $s_2 = J_2$ and $g_J = 2$; that is,

$$H_{2\text{eff}} = C_v \Delta T_2 / \chi_m H.$$

As is seen from this formula, in order to determine $H_{2\text{eff}}$ it is necessary to measure ΔT_2, C_v, and χ_m.

For gadolinium garnet, we made a measurement of the total magnetocaloric effect $\Delta T = \Delta T_1 + \Delta T_2$ (ΔT_1 is the magnetocaloric effect due to the sublattice of the iron ions), the paraprocess susceptibility, and the specific heat; similar measurements were also made for yttrium garnet. The specific heat was measured in a silver adiabatic calorimeter by the method described in[5]. In order to extract the magnetocaloric effect ΔT_2 due to the rare-earth sublattice in the T_c region, experimental data on the magnetocaloric effect of yttrium garnet were used; this enabled us to determine ΔT_1 and to deduce ΔT_2 (see Fig. 1), because in this case the magnetocaloric effect is entirely due to the iron sublattice, since the yttrium ions are nonmagnetic.

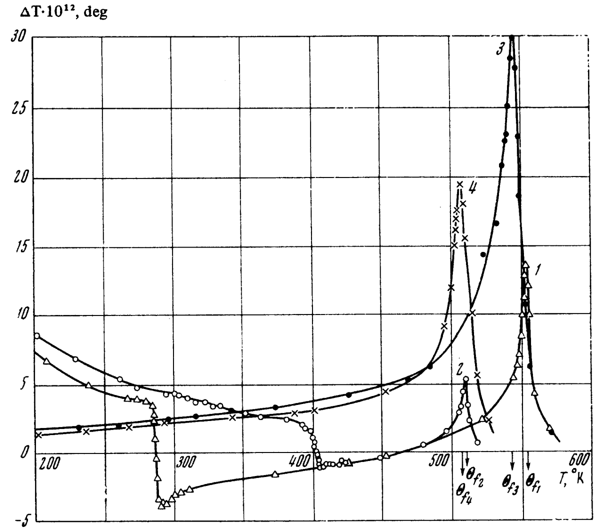

FIG. 1. Temperature dependence of the magnetocaloric effect in a field of 16 kOe for the following garnets: 1–$Gd_3Fe_5O_{12}$; 2–$Gd_3Ga_{0.3}Fe_{4.7}O_{12}$; 3–$Y_3Fe_5O_{12}$; 4–$Y_3Ga_{0.3}Fe_{4.7}O_{12}$.

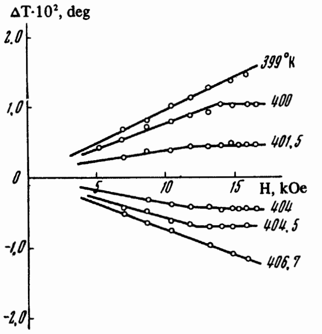

FIG. 2. Dependence of the magnetocaloric effect on external field H in the immediate vicinity of the compensation temperature for $Gd_3Ga_{0.3}Fe_{4.7}O_{12}$ garnet.

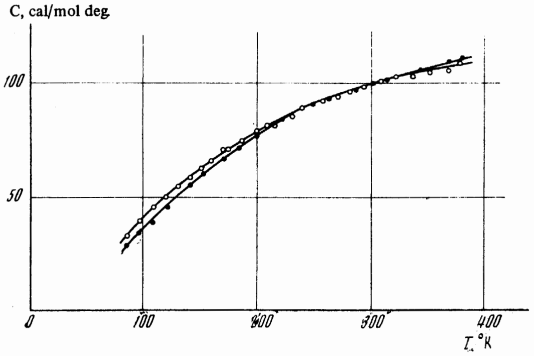

FIG. 3. Temperature dependence of heat capacity for garnets: ○, $Gd_3Fe_5O_{12}$; ●, $Y_3Fe_5O_{12}$.

As is well known, in the immediate vicinity of the compensation temperature in gadolinium garnet, there may occur an induced noncollinear magnetic structure[6]. Suitable measurements of the magnetocaloric effect in the immediate neighborhood of T_c enabled us to use in the calculation values of ΔT known to be related to the region of collinear magnetic structure. As was shown in[6], what corresponds to the collinear structure is the section in which the values of ΔT increase linearly with field (see Fig. 2 and[6]).

According to our experimental data, the molar heat capacity calculated for $Gd_6Fe_{10}O_{24}$ is $C_V = 200$ cal/mol-deg; the molar susceptibility is $\chi_m = 0.143$. From measurements of the total magnetocaloric effect ΔT below and above $T_c = 286.3°K$[6] in a field $H = 10^4$ Oe, at 285.3°K, $\Delta T = 2.5 \times 10^{-2}$ deg; whereas at 287.3°K, $\Delta T = -2.3 \times 10^{-2}$ deg.

On the other hand, for $Y_6Fe_{10}O_{24}$ near T_c we have $\Delta T_1 = 2.0 \times 10^{-2}$ deg. From these data one can find the magnetocaloric effect due to the gadolinium sublattice. It must be taken into account that for $T < T_c$, the sublattice of magnetic moments of the gadolinium ions is directed along the field and gives a positive magnetocaloric effect, whereas the iron sublattice is disordered by the field and gives a negative magnetocaloric effect ($\Delta T_1 = -2.0 \times 10^{-2}$ deg for $H = 10^4$ Oe). Consequently, $\Delta T_2 = \Delta T - \Delta T_1 = 4.5 \times 10^{-2}$ deg.

For $T > T_c$, the iron lattice is directed along the field, therefore it is ordered by the field and gives a positive magnetocaloric effect $\Delta T_1 = +2.0 \times 10^{-2}$ deg; but the resultant magnetocaloric effect of the garnet in this range of temperature is nevertheless negative ($\Delta T = -2.3 \times 10^{-2}$ deg); this is explained by the larger magnitude of the magnetocaloric effect ΔT_2 due to disordering of the sublattice of gadolinium ions, which is directed opposite to the field (paraprocess of antiferromagnetic type[2]). Therefore $\Delta T_2 = -4.3 \times 10^{-2}$ deg. Thus from measurements made in the immediate vicinity of the compensation point, one can determine the effective exchange field exerted on the rare-earth sublattice by the iron ions; $H_{2\text{eff}} = 258$ kOe.

We also made similar measurements of the magnetocaloric effect and the paraprocess susceptibility in the T_c region for the mixed garnet $Gd_3Ga_{0.3}Fe_{4.7}O_{12}$ (see Figs. 1 and 2). Starting from the assumption that the heat capacity of such a mixed garnet differs little from the heat capacity of gadolinium garnet, an assumption that is supported by the behavior of the C(T) curves for gadolinium and yttrium garnets (see Fig. 3), we made for this garnet also an estimate of the effective exchange field exerted on the Gd^{3+} ions. In order to estimate the magnetocaloric effect of the sublattice of iron ions, we made an yttrium garnet with a similar mixture in the iron sublattice, of composition $Y_3Ga_{0.3}Fe_{4.7}O_{12}$, for which also measurements were made of the magnetocaloric effect and the susceptibility (see Figs. 1 and 2). In the mixed gadolinium garnet, as is seen from Fig. 1, both the Curie point Θ_{f_2} and the compensation point are shifted. Shift of the Curie point to a region of much lower temperatures, which is evident from Fig. 2 by the shift of the maximum of the magnetocaloric effect, corresponding to the Curie point, indicates a weakening of the exchange interaction by about 8%.

As is seen from Fig. 1, the compensation temperature of the mixed gadolinium garnet rose; this may be due either to weakening of the Fe_{a-d} sublattice, or to the fact that the temperature dependence of the magnetic moment of the gadolinium sublattice is different in the mixed garnet. Evidence in favor of the first assumption, however, is a similar shift of the Curie point Θ_{f_4} in the mixed garnet of yttrium in relation to Θ_{f_3} for yttrium garnet.

Our measurements enabled us to determine, for $Gd_3Ga_{0.3}Fe_{4.7}O_{12}$ garnet, $H'_{2\text{eff}}$ in the immediate vicinity of the compensation point: $H'_{2\text{eff}} = 232$ kOe; this is approximately 10% smaller than the value obtained by us for the effective exchange field acting on the Gd ions in gadolinium garnet.

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