Magnetic phase diagram of iron garnets of the system $Y_{3-x}Gd_xFe_5O_{12}$ in fields up to 50 T

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In order to investigate the magnetic phase diagram, the differential magnetic susceptibilty is measured for the iron-garnet system $Y_{3-x}Gd_xFe_5O_{12}$ ($0.01 \leq x \leq 0.2$) over the temperature range $T = 1.6$ to 4.2 K, in a magnetic field H up to 50 T. Transitions are observed from the ferromagnetic to a noncollinear phase and from the noncollinear to the ferromagnetic. From the data, the effective field of exchange interaction between gadolinium and iron ions is determined, $H_{mol} = (36.5 \pm 1.5)$ T. It is shown that the phase diagram in the H, x plane for compositions with $x \geq 0.1$ is well described by molecular-field theory. For compositions with $x \leq 0.5$, a deviation from this theory is observed; it is attributable to a decrease of indirect interaction between the gadolinium impurity atoms.

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INTRODUCTION

One of the problems in the study of magnetically ordered crystals, including rare-earth iron garnets, is the determination of the values of the intersublattice exchange interactions.[1] The magnetic system of these ferrites consists of three sublattices, of which two are formed by the magnetic moments of the iron ions, located on the octahedral (a) and tetrahedral (d) sites, while the third consists of the magnetic moments of the rare-earth ions, located on dodecahedral (c) sites. The strong antiferromagnetic exchange interaction between the a and d sublattices often justifies consideration of a resultant iron sublattice with magnetization M_{Fe} equal to the difference between the magnetizations of the d and a sublattices ($M_{Fe} = M_d - M_a$). A weaker interaction (also as a rule of antiferromagnetic type) is exchange between the rare-earth and resultant iron sublattices, which determines the state of the rare-earth sublattice. The direct interaction between the rare-earth ions is small; it can usually be neglected. The values of these interactions can be estimated from the characteristics of the noncollinear (angular) magnetic phases that occur as a result of competition between the exchange interactions and the Zeeman interaction of the sublattices with an external magnetic field.[1] This requires investigation of the magnetic phase diagrams in fields of the order of the exchange fields, *i.e.*, 10^1–10^2 T.

The yttrium-gadolinium iron garnet that we have investigated is of special interest because the rare-earth sublattice in it is formed by spins of Gd^{3+} ions that are in S states and therefore have no orbital moment, so that the magnetic phenomena that occur are not complicated by the presence of spin-orbit interaction, as is the case in other rare-earth garnets.[2] We obtained the magnetic phase diagram of the ferrite $Y_{3-x}Gd_xFe_5O_{12}$ in a range of fields comparable with the effective field of $3d$ – $4f$ exchange interaction between the resultant iron ($3d$) and gadolinium ($4f$) sublattices, and sufficient for observation of the limits of the angular phase in compositions with $x \leq 0.2$.

EXPERIMENTAL METHOD

We investigated magnetic phase transitions in polycrystalline specimens of the system $Y_{3-x}Gd_xFe_5O_{12}$ ($0.01 \leq x \leq 0.2$) by measurements of the differential magnetic susceptibility at temperatures 1.6–4.2 K in pulsed magnetic fields up to 50 T. The specimens were prepared by the usual ceramic technology from oxides of high purity (rare-earth impurity content $<10^{-3}$%).

The magnetic field was produced by discharge of a capacitor bank (capacitance 14 mF, maximum voltage 5 kV) into a many-turn solenoid. To decrease the stray currents, thyristors T-630 were used as dischargers; for discharge of two sections of the bank connected in parallel, they were inserted three in series.

The solenoid was wound with copper strip (cross section 2×3.5 mm^2), sheathed with threads of NbTi alloy. Such strip is usually used to manufacture coils of superconducting magnetic systems. Its tensile strength, $(9–10) \cdot 10^8$ N/m^2, substantially exceeds the strength of unsheathed strip. The cooling of the pulse solenoid was done with liquid nitrogen, so that the superconducting strip was in the normal state, and only its increased strength was used. The rim of the solenoid, of thickness 10–20 mm, was wound with glass fibers impregnated with epoxy resin. The construction described made it possible to obtain repeatedly, in an opening of diameter 20 mm, a field of amplitude 50 T with duration 15 msec of the half-sinusoidal pulse.

The differential magnetic susceptibility was measured with a system of four recording coils.[3] The main and compensating coils had about a thousand turns each and were arranged coaxially. To determine the absolute values of the susceptibility, the measurement system was calibrated against the known value[4] of the susceptibility of an MnF_2 single crystal in a field perpendicular to the axis C_4 at the temperature of liquid helium. The sensitivity of the susceptibility measurements was 10^{-5}. This corresponds to a sensitivity 0.2 G cm^3 to the magnetic moment of the specimen.

The field was measured with a test coil placed near the specimen and that was calibrated against the EPR signal in diphenyl picryl hydrazyl in field 7.0 T.

EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows a typical experimental variation of the susceptibility with the field for the composition with x

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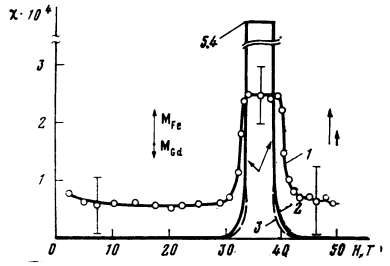

FIG. 1. Variation of differential magnetic susceptibility with field in the ferrite $Y_{2.95}Gd_{0.05}Fe_5O_{12}$ at initial temperature T_0 = 1.6 K. Curve 1, experimental data; Curves 2 and 3, calculation by molecular-field theory for adiabatic (2) and isothermal (3) magnetization conditions.

=0.05. In the field interval 30–40 T, a noncollinear phase occurs. In this phase, the susceptibility is large in comparison with the susceptibility in the collinear phases and is, in first approximation, constant. Similar relations are observed also in specimens of other compositions.

Most of the results of our experiments are well described by the two-sublattice model of an iron garnet in the molecular-field approximation.[5] The magnetic energy density in such a model can be written in the form

$$E=-\mathbf{H}(\mathbf{M}_{Fe}+\mathbf{M}_R)+\lambda_{12}\mathbf{M}_{Fe}\mathbf{M}_R+\lambda_1\mathbf{M}_{Fe}^2+\lambda_2\mathbf{M}_R^2, \quad (1)$$

where $\mathbf{M}_{Fe}$ and $\mathbf{M}_R$ are the magnetizations of the resultant iron sublattice and of the rare-earth sublattice, and where λ_{12} and λ_1, λ_2 are the molecular-field coefficients that characterize the exchange interactions between the sublattices and within the sublattices. We shall neglect exchange within the rare-earth sublattice (that is, we all set $\lambda_2=0$) and shall suppose that its magnetization depends only on the external field H and on the exchange field produced by the iron sublattice. At low temperatures, it is also possible to neglect the dependence of the magnetization of the iron sublattice on the field and temperature.

By use of this approximation, it is easy to show that at temperatures below the critical temperature T_{cr}, there occurs within the field interval H_1 to H_2 a noncollinear phase, in which neither $\mathbf{M}_{Fe}$ nor $\mathbf{M}_R$ is parallel to $\mathbf{H}$ (but the total magnetization $\mathbf{M}_{tot}=\mathbf{M}_{Fe}+\mathbf{M}_R$ is parallel to $\mathbf{H}$). When $H<H_1$, the magnetizations of the iron and rare-earth sublattices are antiparallel; when $H>H_2$, coparallel. For an iron garnet that has no compensation point, as in our case, the values of the fields H_1 and H_2 are determined by the following equations:

$$-H_1+\lambda_{12}M_{Fe}=\lambda_{12}M_{R0}B_J\left[\frac{\mu_R}{kT}(-H_1+\lambda_{12}M_{Fe})\right], \quad (2)$$

$$H_2-\lambda_{12}M_{Fe}=\lambda_{12}M_{R0}B_J\left[\frac{\mu_R}{kT}(H_2-\lambda_{12}M_{Fe})\right], \quad (3)$$

where M_{R0} is the magnetization of the rare-earth sublattice at 0 K, B_J is the Brillouin function for the rare-earth ion with magnetic moment $\mu_R=\mu_B g_J J$ (μ_B is the Bohr magneton, g_J the Landé factor, J the total angular momentum), and k is Boltzmann's constant. In the presence of a compensation temperature T_{comp}, the sign of H_1 in formula (2) is changed in the temperature range $T<T_{comp}$. The critical temperature is determined by the relation, which follows from formulas (2) and (3),

$$T_{cr}=\frac{J+1}{3Jk}\mu_R\lambda_{12}M_{R0}. \quad (4)$$

At temperatures $T>T_{cr}$, complete demagnetization of the rare-earth sublattice occurs in field $H_{mol}=\lambda_{12}M_{Fe}$, and parallel arrangement of the vectors $\mathbf{M}_{Fe}$ and $\mathbf{M}_R$ is attained on further increase of the field, without formation of an angular phase.

In the ferrimagnetic ($H<H_1$) and ferromagnetic ($H>H_2$) collinear phases of an iron garnet without a compensation point, the magnetization of the rare-earth sublattice is described by the formula

$$M_R(T,H)=M_{R0}B_J\left(\frac{\mu_R}{kT}|H-\lambda_{12}M_{Fe}|\right), \quad (5)$$

in the angular phase, by the formula

$$M_R(T)=M_{R0}B_J\left(\frac{\mu_R\lambda_{12}M_R}{kT}\right), \quad (6)$$

so that it is independent of the field.

The total magnetization of a rare-earth iron garnet in the collinear phases is equal to the difference ($H<H_1$) or sum ($H>H_2$) of the magnetizations of the iron and rare-earth sublattices; in the angular phase, it depends linearly on the field[5]:

$$M_{tot}=H/\lambda_{12}. \quad (7)$$

Thus on the magnetization curves there should be breaks at $H=H_{1,2}$ corresponding to transitions from the Brillouin function (5) to the linear dependence (7). On the curves $\chi(H)$ of differential magnetic susceptibility vs field, these breaks correspond to jumps, which were in fact observed by us experimentally (Fig. 1). The theoretical curves in Fig. 1 were calculated by differentiation of the relations (5) and (7) (differentiations of the function $M_{Fe}(T,H)$ gives no contribution to the susceptibility at low temperatures in the field range considered). To investigate ferrites of the system $Y_{3-x}Gd_xFe_5O_{12}$, we used the following values of the magnetic parameters: $M_{Fe}=198$ G, $\mu_R=\mu_{Gd}=7\mu_B$, $M_{R0}=8x\mu_R/a^3$ ($8x$ is the number of gadolinium atoms in an elementary cell; $a=12.37\cdot10^{-10}$ m is the lattice constant). The molecular-field coefficeint $\lambda_{12}=H_{mol}/M_{Fe}=1840$ was determined from the experimental value $H_{mol}=36.5\pm1.5$ T, obtained from the phase diagram (Fig. 2) in the H,x plane.

The theoretical value of the susceptibility in the angular phase is larger than the experimental (Fig. 1) because of the fact that the observed range of existence of this phase in the compositions investigated is larger than that calculated by molecular-field theory (see Fig.

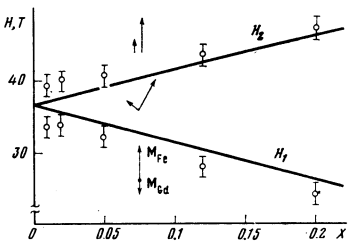

FIG. 2. Magnetic phase diagram of the system of ferrites $Y_{3-x}Gd_xFe_5O_{12}$ at initial temperature T_0 = 1.6 K. Points, experimental data; straight lines, calculation by molecular-field theory.

2). The change of magnetization of the specimens as a result of magnetization reversal of the gadolinium sublattice (that is, the area under the peak of $\chi(H)$ in Fig. 1) corresponds to the theoretical value within the limits of experimental error. The disagreement of susceptibilities just noted disappears on increase of x to 0.2, where the experimentally observed range of the angular phase almost coincides with the theoretical. We shall discuss below a possible reason for the fact that with decrease of x, the agreement breaks down.

For construction of a phase diagram in the H, x plane, values of the limiting fields H_1 and H_2 were taken that corresponded to the half-height of the observed peak of the susceptibility, for each of the compositions investigated. The theoretical phase diagram (the solid lines in Fig. 2) was calculated by formulas (2) and (3) with the values of the magnetic parameters given above. From the calculations by molecular-field theory it follows that the effective field H_{mol} of exchange interaction between the rare-earth and iron sublattices is determined by the relation $H_{mol}=(H_2-H_1)/2$. It is with this formula that the value of H_{mol} indicated above was obtained.

It should be mentioned that by determining H_{mol} for each of the compositions investigated and taking into account that the lattice constant changes with composition, one can in principle determine the variation of the $3d$-$4f$ interaction parameter with the lattice constant of the crystal, although the accuracy of our experiments is not yet sufficient for this.

Since our measurements were made in pulsed fields, the magnetization process may not be considered isothermal. It is known that in rare-earth iron garnets there is a magnetocaloric effect that leads, during magnetization under adiabatic conditions, to cooling of the specimen, if the original temperature is above the compensation temperature.[6] This is the situation that is realized in the compositions that we investigated, since in our case the magnetization of the iron sublattice remains larger than the magnetization of the rare-earth at any temperatures, and compensation is not attained. The effect of the magnetic cooling may be as much as several degrees on magnetization from zero field to 10 T.[7] A direct estimate of the degree of adiabaticity of the process, determined by the heat-transfer time τ, is difficult under the conditions of our experiments because of absence of information about the thermal characteristics of the specimens under investigation. From the available literature data on heat conductivity[8] and specific heat[9] of iron garnets at low temperatures, values of τ are obtained that lie within wide limits from 10^{-1} to 10^{-4} sec. But a number of facts indicate that the magnetization process under our experimental conditions is nearly adiabatic. First, on the specimens with $x=0.01$ and 0.02, for which calculations by formula (4) give $T_{cr}=1.03$ and 2.06 K, we observed transitions to the angular phase already at initial temperature $T_0=4.2$ K, i.e., considerably larger than T_{cr}, above which the existence of an angular phase is impossible. Second, for initial temperatures $T_0=1.6$ and 4.2 K, approximately the same variations of the susceptibility with magnetic field were observed for each of the compositions investigated.

Consideration of phase diagrams in the H, T plane calculated by formulas (2) and (3) of molecular-field theory (Curves 1 and 2 in Fig. 3; the magnetic parameters are the same as for the preceding figures) enable us to understand the facts mentioned, if we allow for cooling during adiabatic magnetization.

The magnetocaloric effect is described by the relation

$$dT=\frac{T}{C_H}\left(\frac{\partial M_{tot}}{\partial T}\right)_H dH, \tag{8}$$

where C_H is the specific heat of the specimen at constant field. Both the crystal lattice and the spin system contribute to the specific heat of the specimen. At temperatures low in comparison with the Debye temperature ($\Theta_D\approx 550$ K for the compositions investigated), the lattice contribution to the heat capacity per formula unit is $C_{lat}=A(T/\Theta_D)^3$, with constant $A=1.08\cdot10^{-12}$ erg/K determined from experimental data on the specific heat of yttrium-iron garnet (YIG) in the helium temperature range.[9] The heat capacity of the spin system of our iron garnet is make up of the heat capacities of the iron and of the gadolinium sublattices. The heat capacity of the gadolinium sublattice can be calculated by the usual thermodynamic formula for a paramagnet[10]: $C_{magn}=xky^2dB_J/dy$, where $y=\mu_R|H-H_{mol}|/kT$. The contribution to the heat capacity from the iron sublattice was actually taken into account in the estimate of the constant A from the experimental value of the heat capacity of YIG. The dependence of this contribution on the magnetic field is slight, and we neglect it in the calculation of the magnetocaloric effect.

Now from formula (8), by using the relations for M_R, C_{lat}, and C_{magn} and by assuming that $(\partial M_{Fe}/\partial T)_H=0$, one easily obtains the change of temperature of the ferrites under investigation during adiabatic magnetization in the collinear phases:

$$T_H^3-T_0^3=b\int_{y_0}^{y_H} yB_J'(y)\,dy, \tag{9}$$

where $b=3kx\Theta_D^3/A$; quantities with index 0 correspond to $H=0$, those with index H to field H. In the angular phase the magnetocaloric effect is nearly zero.[1] Curves 3–6 in Fig. 3 were calculated by formula (9) for $T_0=1.6$ and 4.2 K and for $x=0.02$ and 0.05.

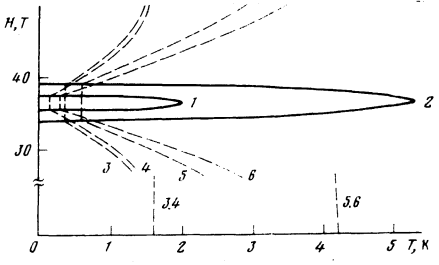

FIG. 3. Magnetocaloric effect in ferrites $Y_{3-x}Gd_xFe_5O_{12}$. Curves 1 ($x=0.02$) and 2 ($x=0.05$) bound the region of existence of the angular phase. Curves 3 and 4 represent the dependence of the temperature of the specimens with $x=0.05$ and 0.02 on the magnetic field during adiabatic magnetization for initial temperature $T_0=1.6$ K; Curves 5 and 6, the same for initial temperature $T_0=4.2$ K.

Thus the calculation shows that during adiabatic magnetization, even at initial temperature $T_0 = 4.2$ K in fields larger than 30 T, the specimens should be cooled to temperatures less than 1 K. It is now clear that observation of jumps of susceptibility during transitions to the angular phase, even at $T_0 > T_{cr}$ for specimens with $x < 0.05$, becomes possible because of the adiabatic character of the magnetization in our experiments. The coincidence of the experimental values of the fields of transition to the angular phase for different initial temperatures can also be explained by treating the magnetization process as adiabatic. In Fig. 3 it is evident that the theoretical values of the transition fields for each of the compositions are almost independent of the initial temperature of an adiabatic process (compare curves 4 and 6 for $x = 0.02$ and curves 3 and 5 for $x = 0.05$).

In conclusion, we shall discuss the discrepancy between the predictions of molecular-field theory for the phase diagram (Fig. 2) in the H, x plane and the experimental data at small gadolinium concentrations ($x \leq 0.02$). For $x = 0.01$, for example, the field interval within which the angular phase is observed is five times larger than the calculated. Furthermore, these intervals are practically the same for $x = 0.01$ and 0.02. Use of a more rigorous three-sublattice model of an iron garnet (two iron sublattices M_a and M_d and a gadolinium M_R; see, for example, Ref. 11) changes the theoretical values of the limiting fields $H_{1,2}$. An estimate shows that the corrections to the limiting fields in this case are

$$\Delta H_{1,2} \cong \frac{\lambda_{12}^2 (M_{Fe} - M_R) M_R M_a}{\lambda_{ad} M_d M_{Fe}} \sim 10x \text{ [T]}$$

(λ_{ad} is the molecular-field coefficient of the exchange interaction between the a and d sublattices). These corrections are comparable with the experimental error when $x \sim 0.01$. Thus the three-sublattice model cannot explain the differences pointed out between theory and experiment.

We suggest that these differences are caused by passage to a range of such small concentrations of gadolinium ions (antiferromagnetic impurities) that the indirect interaction of their spins via the iron sublattice (a ferromagnetic matrix) can be neglected (and it is this interaction that permits use of the concept of a magnetic sublattice $\mathbf{M_R}$). In this concentration range, the interaction of the localized spin of an impurity with the magnon field of the matrix should lead to a stepwise (specifically quantum) type of magnetization reversal of the impurity in fields near H_{mol}.[12] The field interval over which the stepwise magnetization reversal occurs is, as follows from Ref. 13, independent of the impurity concentration and remains finite on decrease of x. It would be interesting to continue the investigations of $Y_{3-x}Gd_xFe_5O_{12}$ garnets at $x < 0.01$ in order to detect certain peaks of the susceptibility, predicted in Ref. 12, corresponding to the quantum process of magnetization reversal of the gadolinium impurity.

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