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Phase transitions induced by magnetic field in ferrimagnets with one unstable magnetic subsystem

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Abstract

Within the molecular field theory, magnetic phase diagrams for a two-sublattice ferrimagnet with one unstable magnetic subsystem are determined. A detailed analysis is given for the experimental magnetization curve for the f–d ferrimagnet $Y_{0.75}Tm_{0.25}(Co_{0.88}Al_{0.12})_2$ measured up to 38 T at 4.2 K in which two metamagnetic transitions and a non-collinear phase between them are observed.

If any of the magnetic subsystems of a ferrimagnet exhibits a magnetic instability, i.e. a metamagnetic transition from a weakly (with magnetization M_W) to a strongly (M_S) magnetic state, a magnetic field can induce a sequence of magnetic phase transitions which differ in their nature (transitions between collinear and non-collinear structures as well as a metamagnetic transition).

In the exchange approximation within the molecular field theory, we calculated various possible magnetic phase sequences for a two-sublattice ferrimagnet of this type. In view of the basic proposition of molecular field theory that in the thermodynamic equilibrium state the sublattice magnetizations are aligned along the effective fields acting on them, we construct phase diagrams by analyzing the values and signs of these effective fields. Possible phase diagrams depend on the parameters of the system: the exchange parameter λ, the metamagnetic transition field H_m and the sublattice magnetic moments M_1 and M_2.

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In the case of a weak exchange, we can write for a small magnetic moment of the stable sublattice M_2:

$$\lambda < H_m/M_{1s}, \quad \lambda M_2 < H_m. \tag{1}$$

Under these conditions, the magnetic field first induces a non-collinear phase followed by a metamagnetic transition. For a larger magnetic moment M_2:

$$\lambda M_2 > H_m \tag{2}$$

in which case, two metamagnetic transitions with a non-collinear phase between them can occur. For larger values of the exchange parameter:

$$H_m/M_{1w} > \lambda > H_m/M_{1s}, \tag{3}$$

the existence of two non-collinear phases with a metamagnetic transition between them is possible. With a further increase of the exchange parameter

$$\lambda > H_m/M_{1w}, \tag{4}$$

the metamagnetic transition can appear before the non-collinear phase has occurred.

The substituted intermetallic compounds of the series $Y_{1-t}R_t(Co_{1-x}Al_x)_2$ (R is a heavy rare earth element, RE)

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with negative intersublattice exchange interaction λ_{fd} may serve as representatives of such ferrimagnets. The stable magnetic subsystem of these two-sublattice ferrimagnets is formed by localized moments of the 4f-shells of RE. The second itinerant, magnetically unstable, subsystem is constituted by the magnetic 3d-electrons of Co hybridized with the 5d (4d)-electrons of RE (Y).

In the $Y(Co_{1-x}Al_x)_2$ compounds, the ground state is paramagnetic for Al concentrations below 11 at% [1]. A magnetic field induces a metamagnetic transition to the ferromagnetic state, with the critical field of the metamagnetic transition, H_m, decreasing for increasing x. For $x \geqslant 0.12$ a metamagnetic transition takes place between weakly and strongly ferromagnetic states. By substituting magnetic RE for Y it is possible to vary the effective field acting on the d-subsystem in the $Y_{1-t}R_t(Co_{1-x}Al_x)_2$ compounds and realize magnetization curves of different character in dependence on RE and its concentration.

In the molecular field approximation, the thermodynamic potential of the two-sublattice f–d ferrimagnet can be written as:

$$G(M_d, H, T) = F_d - HM_d - tk_BT \ln Z. \quad (5)$$

Here F_d is the thermodynamic potential of the d-subsystem, which can be represented as an expansion in powers of M_d (see e.g. Ref. [2]). Since $F_d = \int_0^{M_d} H^{exp}(M_d)dM_d$, we used the experimental magnetization curves of the $Y(Co_{1-x}Al_x)_2$ compounds without magnetic RE (e.g. see Fig. 1, curve 1 for $x = 0.12$) in order to calculate F_d as the area under the curve $H^{exp}(M_d)$. The partition function Z of the f-subsystem is

$$Z = \mathrm{sh}[(2J+1)|H + \lambda_{fd}M_d|\mu_f/2Jk_BT]/\mathrm{sh}[H + \lambda_{fd}M_d|\mu_f/2Jk_BT], \quad (6)$$

where J is the total angular momentum of RE. Solving the equation of state $(\partial G/\partial H = 0)$

$$H^{exp}(M_d) = H + \lambda_{fd}tM_f, \quad (7)$$

$$M_f = \mu_f B_J(|H + \lambda_{fd}tM_f|\mu_f/k_BT)$$

for all possible collinear and non-collinear phases and choosing the solution with the minimum value of the thermodynamic potential for given T and H, we can construct magnetization curves of the ferrimagnet under consideration. Examples of some magnetization curves are given in Fig. 1.

In this paper we consider in detail one case of the sequence of phase transitions that are different in nature in the compound $Y_{0.75}Tm_{0.25}(Co_{0.88}Al_{0.12})_2$, which has been chosen as a very interesting example corresponding to the situation given by formula (2). The calculations (see Ref. [3]) show that in the case of paramagnetic ground state of the unstable subsystem, less rich phase diagrams are obtained. At zero external field,

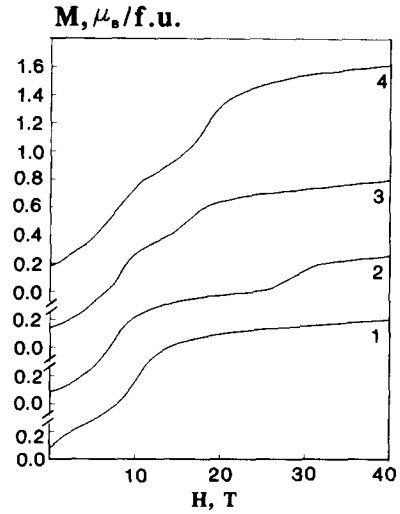

Fig. 1. Experimental magnetization curve of $Y(Co_{0.88}Al_{0.12})_2$ (curve 1) and calculated magnetization curves of $Y_{1-t}R_t(Co_{0.88}Al_{0.12})_2$ corresponding to the cases given by formulae (4) (curve 2), (3) (curve 3) and (1) (curve 4) at 4.2 K. The magnetization curves are shifted along the ordinate axis to distinguish their peculiarities.

$Y(Co_{0.88}Al_{0.12})_2$ is a very weak itinerant ferromagnet ($T_C \simeq 6$ K [4]) with the following values for the magnetic parameters: the spontaneous magnetization in the weakly ferromagnetic state is $0.08\mu_B$/f.u., the critical field of metamagnetic transition, H_m in 12 T and the magnetization in the strongly ferromagnetic state at $H \gtrsim H_m$ amounts to $0.82\mu_B$/f.u. [1]. According to Ref. [5], the magnetic moment, of Tm, μ_f, in the equivalent $Lu_{1-t}Tm_t(Co_{0.88}Al_{0.12})_2$ compounds does not depend on the concentration t and is equal to $4.3\mu_B$/f.u. Considering μ_f to be the same in $Y_{1-t}Tm_t(Co_{0.88}Al_{0.12})_2$, we obtain for the magnetic moment $t\mu_f = 0.25 \times 4.3 = 1.075\mu_B$/f.u.. As this value exceeds the maximum magnetic moment of the d-subsystem, in a weak magnetic field the magnetization of the f-subsystem is expected to align parallel to the external field, whatever the magnetic state of the d-subsystem at zero external field is.

In Fig. 2, the magnetization curve of $Y_{0.75}Tm_{0.25}(Co_{0.88}Al_{0.12})_2$ at 4.2 K is shown. The measurements were performed in quasistatic magnetic fields up to 38 T (with the duration of a pulse $\sim$ 1 s) on powdered free particles, that enabled an exclusion of the influence of magnetic anisotropy [6]. As seen in Fig. 2, the magnetization extrapolated to zero field is equal to $0.24\mu_B$/f.u., which means that in the absence of the field, the magnetization of the d-subsystem equals $0.84\mu_B$/f.u., i.e. this subsystem is in the strongly ferromagnetic state. This curve is characterized by two jump-like transitions

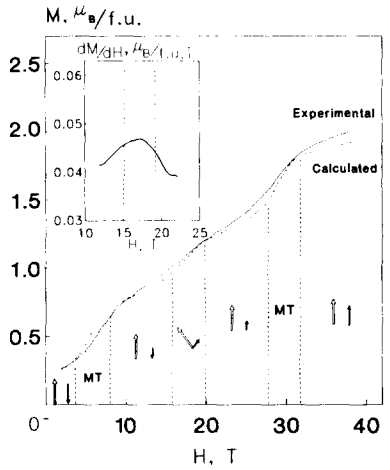

Fig. 2. The experimental and calculated magnetization curves of $Y_{0.75}Tm_{0.25}(Co_{0.88}Al_{0.12})_2$ at 4.2 K. Vertical dashed straight lines separate regions of different magnetic states (MT denotes metamagnetic transition). The magnetic structures in the different regions are schematically depicted by filled (d-subsystem) and open (f-subsystem) arrows. The inset shows the differential susceptibility in the vicinity of the bend for the experimental (solid) and calculated (dashed) curves.

at 6.5 and 28 T and the region of a well-pronounced peculiarity (a bend) between them. The bend is clearly seen as a maximum in the differential susceptibility (see the inset of Fig. 2).

The magnetization curve of $Y_{0.75}Tm_{0.25}(Co_{0.88}Al_{0.12})_2$ calculated for $\lambda = |\lambda_{fd}| = 16.4\ T/\mu_B$ f.u. (this value gives the best agreement with the experimental $M(H)$ curve), $\mu_f = 4.3\mu_B$ and the experimental $H^{exp}(M_d)$ dependence of $Y(Co_{0.88}Al_{0.12})_2$ (Fig. 1, curve 1) is shown in Fig. 2. It is seen that the calculated $M(H)$ curve qualitatively repeats all the peculiarities observed in the experimental one.

Consider now the magnetic phases of $Y_{0.75}Tm_{0.25}(Co_{0.88}Al_{0.12})_2$. At $H = 0$, the d-subsystem is in the strongly ferromagnetic state, because the molecular field $\lambda t M_f = 17.6$ T exceeds the metamagnetic transition field $H_m = 12$ T. As M_{ds} is antiparallel to the external field, the effective field $H_d = -H + \lambda t M_f(H)$ decreases with increasing H, and at $H > H_{m1} = \lambda t M_f(H_{m1}) - H_m = 5.5$ T (6.5 T in experiment) the demagnetization process of the d-subsystem continues up to $H_{cr1} = \lambda(tM_f - M_{dw}) = 15$ T, where a non-collinear phase appears existing up to $H_{cr2} = \lambda(tM_f + M_{dw}) = 19$ T. For $H_{cr1} < H < H_{cr2}$, the antiparallel alignment of the f- and d-sublattice moments is continuously changed to the parallel one. In this field region (between 12.5 and 21 T), the bend can be seen in the experimental curve. As in the parallel phase $H_d = H + \lambda t M_f(H)$, a further increase of the external field results in an increase of the effective field acting on the d-subsystem. At the critical field $H_{m2} = H_m + \lambda t M_f = 29$ T (28 T in experiment), one more metamagnetic transition occurs, connecting this time with the transition of the d-subsystem from the weakly to the strongly ferromagnetic phase. At $H > H_{m2}$, the resulting magnetization increases due to the susceptibilities of sublattices.

Thus, we have obtained a good qualitative agreement between experimental and calculated magnetization curves. Moreover, the calculations describe quantitatively, with an accuracy of several per cent, the main peculiarities of the experimental curve. Worse agreement is observed in the region where the non-collinear phase exists: the peculiarity is essentially less pronounced and extends over a wider field interval in the experimental curve than in the calculated one (see the inset of Fig. 2). This fact may be connected with effects caused by statistical inhomogeneities of the multicomponent system resulting in a smearing of the range of the non-collinear phase. The discrepancies may also be ascribed to small deviations of the aluminum content in the compounds with $t = 0$ and $t = 0.25$.

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