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3d-4f METALLIC COMPOUNDS.

Magnetic moment, magnetic anisotropy and spin-reorientation phase transition in (4f-3d)-intermetallic compounds

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Résumé. — On analyse le rôle des ions 4f et 3d dans la formation du moment magnétique et de l'anisotropie magnétique dans les composés 4f-3d, ainsi que l'influence des interactions d'échange 4f-3d entre les sous-réseaux sur la variation thermique du moment magnétique et de l'anisotropie magnétique. On considère les réorientations de spin par transitions de phase spontanées et celles induites par le champ magnétique.

Abstract. — The role of 4f- and 3d-ions in the formation of magnetic moment and magnetic anisotropy of (4f-3d)-compounds is discussed as well as the influence of intersublattice (4f-3d)-exchange interaction on their temperature dependence. Spontaneous and field-induced SR-phase transitions in (4f-3d)-compounds are considered.

1. Introduction. — Great interest in the magnetic properties of intermetallic compounds of 4f-metals (R) with transition 3d-metals (T) has been displayed in the last 15 years. This is first of all connected with the fact that the (4f-3d)-alloys are used as hard [1] and magnetostrictive [2] magnetic materials, as materials for recording of information using bubble domains [3] as well as for thermomagnetic transducers using spin reorientation [4]. The potential of (4f-3d)-compounds for these purposes is determined by such characteristics as spontaneous magnetization, magnetic anisotropy, magnetostriction and Curie temperature. This paper deals with the formation of magnetic moment and magnetic anisotropy in (4f-3d)-compounds as well as spontaneous and field-induced spin reorientation (SR) phase transitions.

2. Magnetic moment of 4f-ions. — Table I shows the values of the molecular magnetic moment (μ_m) at 4.2 K of R_2Co_{17} [5], RCo_5 [6, 7], R_2Co_7 [8] monocrystals. Figure 1 gives a comparison of values of the magnetic moment per 4f-ion in a trivalent state (μ_{f3+}) calculated from these data by means of the formula

$$\mu_f = (\mu_m^{R_nCo_m} - \mu_m^{(Y,Lu)_nCo_m})/n, \quad \text{with} \quad \mu_f = gJ$$

where g is Lande factor, J is the quantum number of the total angular momentum of 4f-ion. The agreement

Fig. 1. — μ_f in R_2Co_{17} (1), RCo_5 (2), R_2Co_7 (3) monocrystals vs. gJ at 4.2 K: $\circ$ f^{3+} , $\bullet$ f^{4+} ; $\bullet$ Tatsumoto *et al.* [6].

is not very good, particularly for the light 4f-ions. However, it may be improved if we take into account the contribution from the polarization of conduction electrons in μ_m [9] as well as the influence of the crystal and exchange fields on the energy state of 4f-ions. In the case of Ce compounds it is necessary also to suppose that the Ce ions are tetravalent and give the additional 4f-electron to the 3d Co band, decreasing

μ_{Co} relative to μ_{Co} in Y_nCo_m by $\frac{n}{m} \mu_B$.

Table I.

R	Ce	Pr	Nd	Sm	Gd	Tb	Dy	Ho	Er	Tm	Lu	Y
$\mu_m^{R_2Co_{17}}/\mu_B$	24.6	32.6	33.2	27.8	13.8	8.9	7.5	7.5	9.8	13.4	26.9	27.2
$\mu_m^{RCo_5}/\mu_B$	6.6	10.4	10.4	8.5	1.42	0.69	1.71	2.06	1.71	—	—	8.3
$\mu_m^{R_2Co_7}/\mu_B$	—	10.9	13.1	9.5	4.2	7.3	—	—	—	—	—	9.6

The temperature dependence of the 4f-sublattice magnetic moment for R_2Co_{17} as shown in figure 2 [5] is approximated by the Brillouin function

$$\frac{\mu_R(T)}{\mu_R(0)} = B_j \left(\frac{gJ\mu_B H_m}{kT} \right) = B_j \left(\frac{2A(g-1)J\langle s \rangle}{kT} \right) = B_j \left(\frac{\xi_0 m_d(T)}{T} \right) \quad (1)$$

with a suitable choice of the parameter

$$\xi_0 = 2A(g-1)J\langle s \rangle/K.$$

ξ_0 represents the magnitude of the (4f-3d)-exchange interaction (H_m is the molecular field, A is the resulting exchange parameter, $\langle s \rangle$ is the mean spin value of 3d-ion, $m_d(T) = \mu_d(T)/\mu_d(0)$ is the relative magnetization of the 3d-sublattice, k is the Boltzmann constant). The value of ξ_0 for $R_2(Co_{1-x}T_x)_{17}$, where $R = Sm, Er$ and $T = Fe, Ni, Al$ increases approximately linearly with μ_d [10, 11] as well as with the spin moment of the 4f-ions in R_2Co_{17} compounds [5].

Fig. 2. — Temperature dependences of 4f-sublattice magnetic moment (points-experiment, lines-equation (1)) in R_2Co_{17} .

3. Magnetocrystalline anisotropy. — The magnetic anisotropy of (4f-3d)-compounds may, as a first approximation, be represented as $K = K_f + K_d + K_{fd}$. At low temperatures the 4f-sublattice anisotropy is dominant. The constancy of the values of the anisotropy coefficients per Nd-ion in $Nd_{1-x}Y_xCo_5$ when Nd is substituted by non-magnetic Y, and the small change in lattice parameters a and c (figure 6) points to a single ion type of anisotropy [6]. If the parameters change greatly, K_{2Nd}^0 decreases approximately as the lattice sums in the expression for the crystal field potential. This is clearly seen in the example of the $Nd_{1-x}U_xCo_5$ compound. Figure 3 shows by solid and dashed lines the dependence

$$\frac{K_{2Nd}^0(x)}{K_{2Nd}^0(0)} = 1 - 3 \left[\left(\frac{a(0)}{c(0)} \right)^3 - \frac{\Delta c(x)}{c(0)} - \right.$$

$$\left. - \frac{3}{2} \frac{\Delta a(x)}{a(0)} \right] \cdot \left[\left(\frac{a(0)}{c(0)} \right)^3 - \frac{3}{2} \right]^{-1} \quad (2)$$

for $Nd_{1-x}U_xCo_5$ and $Nd_{1-x}Y_xCo_5$ [7].

Fig. 3. — See text.

The magnetic anisotropy of (4f-3d)-compounds can be changed in a more subtle way when one 3d-ion is substituted by another. Figure 4 gives as an example the concentration dependence of the anisotropy constants for $Er_2(Co_{1-x}Fe_x)_{17}$ [10]. This dependence can be explained if we assume that the anisotropy of these compounds at low temperatures is mainly determined by the Er-sublattice anisotropy, and the crystal field acting on the Er-ions is changed in a complicated way because of an uneven replacement of Co-ions by Fe-ions with unequal charges. In this case we have for $K_1^0(x)$ [11, 12]

$$K_1^0(x) = K_1^0(0) + \omega_1(q_{Fe} - q_{Co}) \times \left[\lambda_d^{(1)} \omega_d^{Fe} + \lambda_f^{(1)} \omega_f^{Fe} + \lambda_B^{(1)} \left(\frac{1}{2} x - \frac{9}{2} \omega_d^{Fe} - 3 \omega_f^{Fe} \right) \right], \quad (3)$$

where $\lambda_d^{(1)}$, $\lambda_f^{(1)}$, $\lambda_B^{(1)}$ are lattice sums for positions 6g and 12k (d-plane), 12j (fd-plane) and 4f (dumbbell), ω_d^{Fe} and ω_f^{Fe} are the probabilities of filling lattice sites (at a given Fe concentration) by Fe ions in d- and fd-planes respectively, q_{Co} and q_{Fe} are the charges of

Fig. 4. — Concentration dependence of the magnetic anisotropy of $Er_2(Co_{1-x}Fe_x)_{17}$ (—●— experiment, - - - calculation in the case of random substitutions (2), or preferential ordering of Fe and Co (3)).

Co and Fe-ions, and $\omega_1 = -2^{-l+1} e^2 \langle r_f^l \rangle / K_l(J)$, where e is the electronic charge, $\langle r_f^l \rangle$ is the mean of the 4f-electronic shell radius in l -th power, $K_l(J) = \alpha_l 0_l^0$ (α_l and 0_l^0 are respectively Elliott-Stevens parameters and operators). Taking into consideration that values and signs of lattice sums are different for different sites of the $\text{Th}_2\text{Ni}_{17}$ type structure, one can generally speaking obtain theoretically different $K_l^0(x)$ values depending on the preferential filling of the lattice sites by Fe-ions and on the different values of the ionic charges. The best agreement of the experimental $K(x)$ dependence with the theoretical one $K_l^0(x)$ is obtained by assuming that the Fe-ions occupy the sites in the following sequence $6g + 12k$, $12j$, $4f$ and that $q_{\text{Co}} = 0$, $q_{\text{Fe}} = 0.7 q_{\text{Er}}$ (figure 4).

The temperature dependence of anisotropy constants for R_2Co_{17} are shown in figures 5 and 6 [5]. From these data for compounds with the light 4f-ions we determined $K_1^0(T)$ for the 4f-sublattice and compared them (figure 8b) with the theoretical ones using a model of crystal field single-ion anisotropy

$$\begin{aligned} K_1^0(T)/K_1^0(0) &= \hat{I}_{1+1/2}[\xi^{-1}(\mu_R(T)/\mu_R(0))] \\ &= \xi_1^J[\mu_R(T)/\mu_R(0)], \end{aligned} \quad (4)$$

where ξ_1^J depends on ξ_0 and is tabulated in [13]. The agreement between the experimental and calculated $K_1^0(T)$ is very good, if we take ξ_0 from [5]. Similar results were obtained by us for NdCo_5 [7]. Thus, by changing ξ_0 , e.g. changing μ_d , we can vary the temperature dependence of the magnetic anisotropy of the 4f-sublattice and, therefore, that of the (4f-3d)-compound as a whole [11].

Fig. 5. — a) K_1 and K_2 vs. T in R_2Co_{17} compounds; b) $K_1^0(T)/K_1^0(0)$ vs. T (points-experiment, lines-calculation on Eq. (4)).

Fig. 6. — K_1 and $K_s = K_1 + 2K_2 + \dots$ vs. T of R_2Co_{17} compounds (--- calculation [5]).

For R_2Co_{17} with heavy 4f-ions the $K(T)$ is more complex especially for $\text{Tb}_2\text{Co}_{17}$, $\text{Dy}_2\text{Co}_{17}$ and $\text{Ho}_2\text{Co}_{17}$ (figure 6). This is probably connected with the fact that a non-collinear magnetization can appear along the hard direction and give an additional increase of the magnetization [14, 15]. Taking into consideration this effect, we theoretically obtained the temperature dependence of the effective anisotropy constant for $\text{Tb}_2\text{Co}_{17}$ (dashed line) in agreement with the experimental $K_s(T)$, as well as the temperature dependence of the true anisotropy constant of the 4f-sublattice [5].

The $K_1(T)$ of the Co-sublattice in (4f-Co)-compounds is satisfactorily defined by the Akulov-Ziner-Carr law [16]

$$K_1(T)/K_1(0) = (1 - \alpha T/T_c [\sigma_s(T)/\sigma_s(0)]^3) \quad (5)$$

Fig. 7. — K_1 vs. T of Y_2Co_{17} , YCo_5 , Y_2Co_7 , $\text{UCo}_{5.3}$ (● experiment, — calculation on Eq. (5)). YCo_5 Frederick *et al.* [17].

with a suitable choice of the coefficient α , as shown in figure 7.

4. Spin reorientation phase transitions. — A detailed review of the studies concerning this problem was recently given by K. P. Belov *et al.* [18]. Here we shall only give some examples of such phase transitions in (4f-3d)-compounds. Figure 8 shows the temperature dependence of the angle φ between the c -axis and the direction of magnetization in $\text{Nd}_{1-x}\text{U}_x\text{Co}_5$ monocrystals [7]. When T increases from T_1 , the angle φ gradually decreases and when $T \geq T_2$ the magnetization lies in the basal plane of the lattice. At such SR-transitions the magnitude of μ_t is preserved, as shown in [19] for TbCo_5 . The angle φ is determined by the equation $\varphi = \arcsin(-K_1/2K_2)^{1/2}$ which is experimentally confirmed for these phase transitions of the second type ($K_2 > 0$) accompanied by heat capacity anomalies [20]. Changing $K_1(T)/K_2(T)$ by change of the uranium concentration, it is possible to shift the temperature of the SR-transitions in $\text{Nd}_{1-x}\text{U}_x\text{Co}_5$. This latter fact will be of practical importance, if the (4f-3d)-compounds with such transitions really find application in thermomagnetic transducers.

Fig. 8. — φ vs. T for $\text{Nd}_{1-x}\text{U}_x\text{Co}_5$ monocrystals (O from the measurement on torque magnetometer,

$$\bullet - \varphi = \arcsin \sqrt{(-K_1/2K_2)} [7]).$$

Another example of SR-transitions are those occurring when magnetizing materials with certain relations between K_1 and K_2 . A theoretical analysis is given in [21-23]. In the case of easy-axis materials with H applied perpendicular to this axis there exists a field H_k at which the magnetization of a single-domain increases in a jump from $m_k = \sigma_k/\sigma_s$ to 1, if $-1 \leq K_2/K_1 \leq -1/6$ (see insert in figure 9), where

$$\begin{aligned} H_k &= 4 K_1 (-K_1/K_2)^{1/2} / 3 \sqrt{6} \sigma_s \quad (a); \\ m_k &= (-K_1/6 K_2)^{1/2} \quad (b); \end{aligned} \quad (6)$$

When decreasing H , the saturated state becomes absolutely unstable at a field

$$H'_k = 2(K_1 + 2 K_2)/\sigma_s.$$

Fig. 9. — σ vs. applied field H in $\text{Er}_2(\text{Co}_{1-x}\text{Fe}_x)_{17}$ monocrystals at 4.2 K : - - - $H \parallel c$, — $H \perp c$.

As we see, an hysteresis $\Delta H = H_k - H'_k$ takes place in this phase transition of first order. If at $H = 0$ the material exhibits a domain structure, during magnetization a nuclei of a new magnetic phase with magnetization parallel to the basal plane is formed in a field $H_n < H_k$ at $m_n = \sigma_n/\sigma_s$, where

$$H_n = (2 K_1/\sigma_s) m_n + (4 K_2/\sigma_s) m_n^3 \quad (a); \quad (7)$$

$$m_n = \frac{1}{3} \left[\left(\frac{-3 K_1}{K_2} - 2 \right)^{1/2} - 1 \right] \quad (b);$$

The transition during magnetization of an easy-plane material along the c -axis, with $4 K_2 - K_1 \leq 0$, may occur in a similar way. In this case the corresponding transition parameters are

$$H_k = -\frac{4(K_1 + 2 K_2)}{3 \sigma_s} \left(\frac{1}{3} - \frac{1}{6} \frac{K_1}{K_2} \right)^{1/2} \quad (a); \quad (8)$$

$$m_k = \left(\frac{K_1 + 2 K_2}{6 K_2} \right)^{1/2} \quad (b);$$

$$H'_k = -2 K_1/\sigma_s \quad (9)$$

$$H_n = \frac{2(2 K_2 - K_1)}{\sigma_s} m_n + \frac{4 K_2}{\sigma_s} m_n^3 \quad (a); \quad (10)$$

$$m_n = \frac{1}{3} \left[\left(\frac{3 K_1}{K_2} + 4 \right)^{1/2} - 1 \right] \quad (b);$$

Experiments showing the possibility of such transitions are given in [23] for $\text{Pr}_2(\text{Co}_{1-x}\text{Fe}_x)_{17}$ and in figure 9 for $\text{Er}_2(\text{Co}_{1-x}\text{Fe}_x)_{17}$. In the uniaxial case ($x = 0, 6$) one obtains from figure 9

$$H_a = \frac{2 K_1}{\sigma_s} = 72 \text{ kOe}, \quad m_n = 0.23.$$

From Eq. (7a, b) we obtain $H_k = 24.1 \text{ kOe}$ and $H_n = 15.7 \text{ kOe}$, which is close to the experimental value 17.5 kOe. Thus we can assume that the SR-phase transitions occurs by the shift of domain walls,

in agreement with the fact that $\frac{d\sigma}{dH} \Big|_{H \approx H_n}$ is approximately equal to the demagnetizing factor of the sample N (in our case $N = 4\pi/3$). We did not find any hysteresis (within an accuracy of about 100 Oe) which is, probably, connected with the easy formation of a transitional domain structure on crystal defects when H is decreased and with small pinning of domain walls on lattice defects.

In case of *easy plane* type anisotropy ($x = 1.0$) we have, from figure 9

$$H_a = 2(K_1 + 2K_2)/\sigma_s = 154 \text{ kOe} \quad \text{and} \quad m_n = 0.31.$$

From Eqs. (8), (10) we obtain $H_k = 61 \text{ kOe}$ and $H_n = 57 \text{ kOe}$ which is also close to the experimental value of 48 kOe.

Interesting examples of SR-phase transitions observed in the (4f-3d)-compounds are the field-induced (metamagnetic) phase transitions, where the type of magnetic ordering is changed. Figure 10 shows the magnetization curves of a Gd_3Co monocrystal at $T = 8 \text{ K}$ along the a -, b -, c -axes [24]. This compound is a noncollinear antiferromagnet with a magnetic structure of a *cross*-type, the Gd magnetic moments (Co has no magnetic moment) lying in the bc -plane of the Fe_3C type lattice [25].

Fig. 10. — σ vs. internal field H_i for monocrystals of Gd_3Co (—•—•—) and $(\text{Gd}_{0.2}\text{Tb}_{0.8})_3\text{Co}$ (—•—) at 8 K along different crystallographic directions. (Each point was measured 30 s after application of the field.)

When H is increased along the b - and c -axes σ increases linearly at first, at $H_k = 8.1 - 8.4 \text{ kOe}$, σ rises in a jump to $\sigma_k = 0.73 \sigma_s$ and then increases gradually to σ_s . μ_{Gd} being equal to gJ that shows the ordering of μ_{Gd} in the state of magnetic saturation to be ferromagnetic.

When H is decreased, we observe an hysteresis $\Delta H = 300 \text{ Oe}$ in the steepest part of $\sigma(H_i)$ (not shown in figure 10). Thus we see that the phase transition is of the first type. Noting that $\frac{d\sigma}{dH} \Big|_{H \approx H_k} = N'$ we may conclude that the transition, as in the previous case, occurs by moving of the metamagnetic walls [26]. When defects are introduced in the crystal the phase transition is field spread [24]. This shows that walls can be nucleated near the crystal defects.

ΔH is considerably increased if we increase the magnetic anisotropy, as seen in the example of the isostructural compound $(\text{Gd}_{0.2}\text{Tb}_{0.8})_3\text{Co}$ [1, 27]. This compound has a noncoplanar magnetic structure with a ferromagnetic component along the c -axis and an antiferromagnetic one along the a - and b -axes similar to the structure of Tb_3Co [28]. Noting that $\frac{d\sigma}{dH} \Big|_{H \approx H_k} = N'$ holds here as well, we may assume that the AF-F phase transition occurs by motion of the metamagnetic walls. Since this compound has a value of magnetic anisotropy ($\sim 10^8 \text{ erg/cm}^3$) comparable with the exchange energy, the walls are narrow (several inter-atomic parameters). As we known, such walls have a high intrinsic coercivity [29]. This fact explains in our opinion the high magnetic hysteresis in the AF-F transition. At a proper choice of fundamental magnetic characteristics K and A , hysteresis may be so large ($\Delta H > H_k$) that the crystal will be in a single domain state at $H = 0$ as when the magnetization is applied along the c -axis. However, in the last case the magnetization along the c -axis does not change the magnetic structure. The crystal has a gigantic value of the energy product ($\sim 100 \text{ mln. gauss.Oe}$) [1, 30] and may be used as a permanent magnet at low temperatures.

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