MAGNETIC PROPERTIES OF $R^{(1)}_{1-x}R^{(2)}_{x}Al_2$ ALLOYS*

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Using the multiband s–f model we derive the formulae determining the Curie temperature and magnetic moment as functions of concentration x for $R^{(1)}_{1-x}R^{(2)}_{x}Al_2$ intermetallic alloys, where $R^{(1,2)}$ denote magnetic rare earth metals. These formulae, applied to $R^{(1)}_{1-x}R^{(2)}_{x}Al_2$ alloys ($R^{(1)}$ = La, Lu, Y, Zr and $R^{(2)}$ = Gd) give the linear dependence of the Curie temperature and magnetic moment versus x in full agreement with experimental data.

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1. Introduction

The Curie temperature and magnetic moment in the paramagnetic state of $R^{(1)}_{1-x}Gd^{(2)}_{x}Al_2$ samples (R = La, Lu, Y) as functions of the concentration x ($x \in [0,1]$) have been measured in [1, 2] and more recently (R = Zr) in [3]. A linear dependence of the Curie temperature T_C and of the magnetic moment M versus x has been found with T_C changing from zero ($x = 0$) to $T_C = 170$ K ($x = 1$) and M changing from zero ($x = 0$) to $M = 7.94\,\mu_\beta$ ($x = 1$), in such a way that $M(x)/x = 7.94\,\mu_\beta$ ($x \in [0,1]$).

The explanation of the observed T_C behaviour of the mentioned samples (but not of the magnetic moment) has been given in the papers [4] and [2], using the formalism invented in [5, 6].

In Ref. [7] the multiband s–f model has been succesfully applied to explain the magnetic properties of the $Gd(Al_{1-x}Me_x)_2$ alloys (Me = Cu, Ag, Pd, In, Sb, Pb, Bi, Sb, Si). The method presented in [7], after some modifications, can be also succesfully applied to the present case ($R^{(1)}_{1-x}R^{(2)}_{x}Al_2$ alloys), where we can additionally explain the behaviour of the magnetic moment of the samples as a function of the concentration x.

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2. Curie temperature and magnetic moment

In Ref. [7] the Curie temperature T_C and the magnetic moment M of $GdAl_2$ have been calculated. The corresponding formulae can be directly applied to the more general case of RAl_2, where R is a magnetic rare earth element. We recall here these formulae. The Curie temperature can be calculated from the following implicite equation:

$$T_C = \frac{S(S+1)}{3k} A, \tag{1}$$

where

$$A = \frac{5\left(I^{(5d)}\right)^2 F^{(5d)}}{2W^{(5d)}} + \frac{\left(I^{(6s)}\right)^2 F^{(6s)}}{2W^{(6s)}} + \frac{\left(I^{(3s)}\right)^2 F^{(3s)}}{W^{(3s)}} + \frac{3\left(I^{(3p)}\right)^2 F^{(3p)}}{W^{(3p)}} \tag{2}$$

$$F^{(a)} = f\left(t^{(a)} - \mu - \frac{W^{(a)}}{2}\right) - f\left(t^{(a)} - \mu + \frac{W^{(a)}}{2}\right), \tag{3}$$

$$(a = 5d, 6s, 3s, 3p)$$

,

$$f(z) = (\exp(\beta z) + 1)^{-1}. \tag{4}$$

In (2) the s–f coupling constant has been denoted by $I^{(a)}$ and the bandwidth of the corresponding band by $W^{(a)}$ with the gravity centre $t^{(a)}$. The upper index a corresponds to $5d$, $6s$ bands of R and to $3s$, $3p$ bands of Al and μ is the chemical potential.

The magnetic moment at T_C is given by:

$$M = 2B\sqrt{S(S+1)}\,\mu_B, \tag{5}$$

where

$$B = 1 + \frac{5I^{(5d)}F^{(5d)}}{2W^{(5d)}} + \frac{I^{(6s)}F^{(6s)}}{2W^{(6s)}} + \frac{I^{(3s)}F^{(3s)}}{W^{(3s)}} + \frac{3I^{(3p)}F^{(3p)}}{W^{(3p)}}. \tag{6}$$

Having in mind an alloy such as $\overset{(1)}{R}_{1-x}\overset{(2)}{R}_x Al_2$ we have to perform the averaging procedure in (1) and (5) over the alloy configurations in order to get the equation for T_C and M. Similarly to [7] we get:

$$T_C = \frac{(1-x)S^{(1)}\left(S^{(1)}+1\right)}{3k} A^{(1)} + \frac{xS^{(2)}\left(S^{(2)}+1\right)}{3k} A^{(2)}, \tag{7}$$

$$M = 2\left[(1-x)B^{(1)}\sqrt{S^{(1)}\left(S^{(1)}+1\right)} + xB^{(2)}\sqrt{S^{(2)}\left(S^{(2)}+1\right)}\right]\mu_B. \tag{8}$$

The quantities $A^{(i)} = A^{(i)}\left(I^{(i-5d)}, I^{(i-6s)}, t^{(i-5d)}, t^{(i-6s)}, W^{(i-5d)}, W^{(i-6s)}\right)$ and $B^{(i)} = B^{(i)}\left(I^{(i-5d)}, I^{(i-6s)}, t^{(i-5d)}, t^{(i-6s)}, W^{(i-5d)}, W^{(i-6s)}\right)$ $(i = 1, 2)$ are given by (2) and (6), respectively. The parameters for Al $(3s, 3p)$ in $A^{(i)}$ and $B^{(i)}$ remain unaffected.

The equation for the chemical potential μ of the alloy can be determined from the equation:

$$2\left[(1-x)\left(\langle n_{\uparrow}^{(1-5d)}\rangle+\langle n_{\uparrow}^{(1-6s)}\rangle\right)+x\left(\langle n_{\uparrow}^{(2-5d)}\rangle+\langle n_{\uparrow}^{(2-6s)}\rangle\right)+\right.$$
$$\left.+\langle n_{\uparrow}^{(\mathrm{Al}-3s)}\rangle+\langle n_{\uparrow}^{(\mathrm{Al}-3p)}\rangle\right]=(1-x)n^{(1)}+xn^{(2)}+6=$$
$$=6+n^{(1)}+x\left(n^{(2)}-n^{(1)}\right), \qquad (9)$$

where

$$\langle n_{\uparrow}^{(i-5d,6s)}\rangle \quad (i=1,2) \quad \text{and} \quad \langle n^{(\mathrm{Al}-3s,3p)}\rangle$$

are the averaged occupation numbers of electrons including the band degeneracies (cf. [7]). Each of the $\mathrm{R}^{(i)}$ atoms (i=1, 2) contributes $n^{(i)}$ electrons and two Al atoms contribute $2\times3=6$ electrons into the system, as visualized in the relation (9).

3. Numerical results

In order to get $T_{\mathrm{C}}=T_{\mathrm{C}}(x)$ and $M=M(x)$ we have to solve the system of implicite Eqs. (7) and (9) selfconsistently, and to calculate M from (8). We consider here the alloy $\mathrm{R}_{1-x}\mathrm{Gd}_x\mathrm{Al}_2$, where R = La, Lu, Y, Zr. Two of them (La, Lu) are monmagnetic rare earth elements. Therefore for them $S^{(1)}=0$, $S^{(2)}=7/2$ (Gd) and the formulae (7) and (8) become simpler.

In Figs. 1 and 2 we show the dependence of the Curie temperature calculated from (7) and (9). We have assumed the same parameters for Al as in [7]. The parameters for Gd and La or Lu are completely irrelevant because Eq. (9) determines such a position of the chemical potential μ, which is below the lower $5d$ and $6s$ band edges of Gd and La or Lu. In such a situation the mentioned bands are empty and therefore their contribution is zero. The magnetic moment M at T_{C} as a function of x, determined by (8) for $\mathrm{R}_{1-x}\mathrm{Gd}_x\mathrm{Al}_2$, (R = La, Lu), is a linear function of x in such a way that the ratio $M(x)/x=7.94\,\mu_{\mathrm{B}}$, what agrees with experiments very well [1, 2]).

In the case of $\mathrm{R}_{1-x}\mathrm{Gd}_x\mathrm{Al}_2$, where R stands for such elements as Y and Zr, the formulae (7), (8) and (9) are applicable, too. These elements are nonmagnetic ($S^{(1)}=0$, $S^{(2)}=7/2$ (Gd)) and their valence bands (Y: $4d^1 5s^2$, Zr: $4d^2 5s^2$) possess the same orbital degeneracies as $5d$, $6s$ bands of Gd, La or Lu. Also in this case the lower band edges of the $4d$ and $5s$ bands of Y and Zr lie higher than the chemical potential determined by (9). Therefore, also here, the only relevant parameters are the ones for Al. It is clear then, that the T_{C} dependence on x calculated from (7) and (9) gives the same results as in Figs. 1 and 2. The $T_{\mathrm{C}}=T_{\mathrm{C}}(x)$ dependence seems to be universal then, for all $\mathrm{R}_{1-x}\mathrm{Gd}_x\mathrm{Al}_2$ alloys (R = La, Lu, Y, Zr) and also the ratio $M(x)/x=7.94\,\mu_{\mathrm{B}}$, what is the conclusion from (8). For R = Y (see

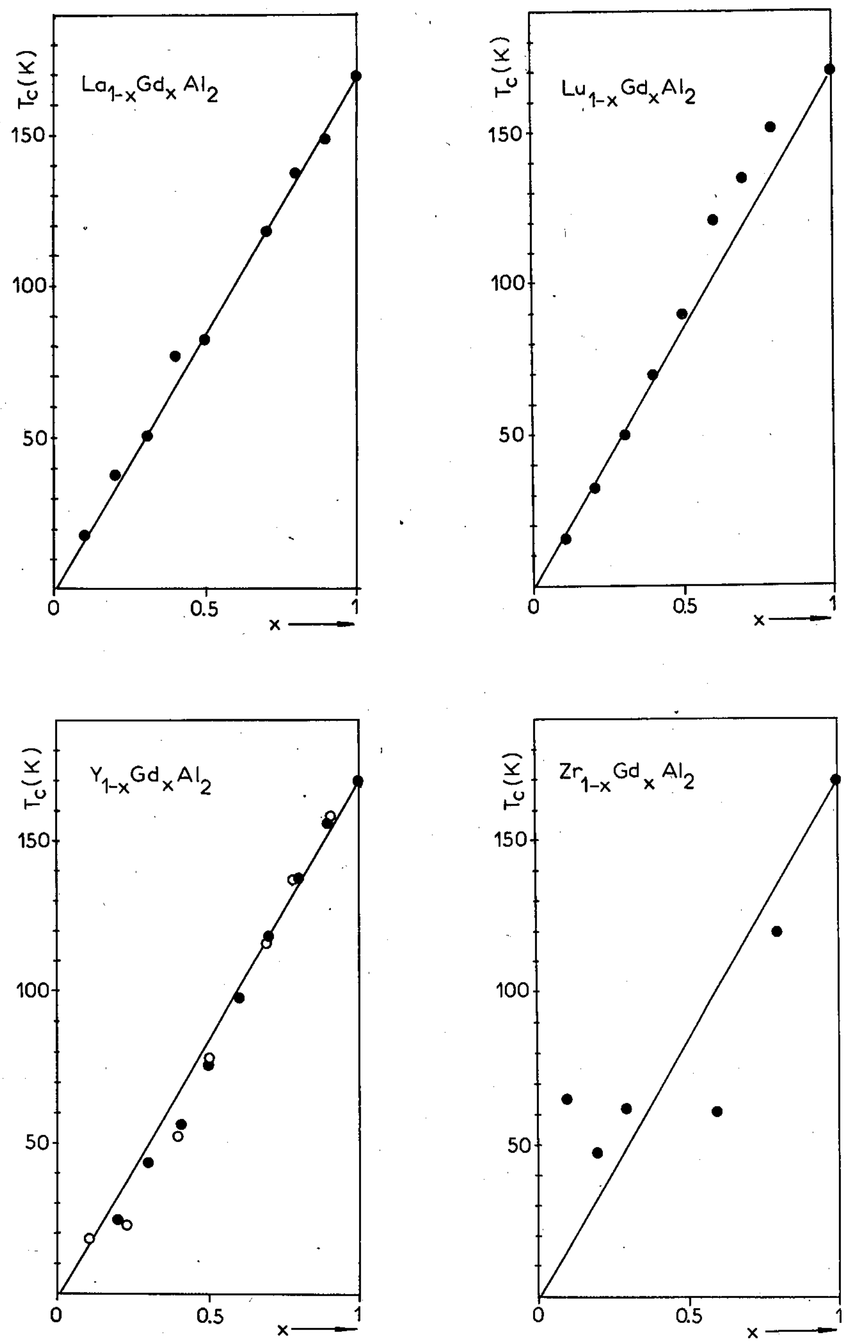

Fig. 1–4. Plot of the Curie temperature vs. concentration x, calculated from Eqs. (7) and (9). The relevant parameters in eV (only for Al) are:

$$I^{(3s)} = -0.093;\ t^{(3s)} = 0;\ W^{(3s)} = 4;\ I^{(3p)} = 0.027;\ t^{(3p)} = 2;$$

$$W^{(3p)} = 3.5;\ n^{(1)} = 3(\mathrm{La, Lu, Y}),\ 4(\mathrm{Zr});\ n^{(2)} = 3\ (\mathrm{Gd}).$$

Fig. 3) the experimental points very closely lie to the mentioned universal linear dependence of $T_C = T_C(x)$, for R = Zr (Fig. 4) the agreement is, however, not so good. In the latter case the deviation from the straight line may be caused by the clusterization process in the $Zr_{1-x}Gd_xAl_2$ samples. In our paper we have used the unperturbed density of states in a form of a rectangle for all the bands considered here $(5d, 6s, 3p, 3d, \ldots)$. In this case the density of states is a constant in the wide energy range. When the chemical potential lies in this range for $x \in [0,1]$ we can obtain linear dependence of $T_C = T_C(x)$ and $M(x)/x =$ const only. May be, in the case of $Zr_{1-x}Gd_xAl_2$ the unperturbed density of states in a form of rectangle is a poor approximation.

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