Microscopic theory of magnetization processes in Y (Co$_{1-x}$ Al$_x$)$_2$

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Abstract
Employing *ab initio* electronic structure calculations we study the development
of the magnetic properties in $Y(Co_{1-x}Al_x)_2$ for varying Al concentration.
The effect of substitutional disorder is treated in the coherent-potential
approximation implemented within a tight-binding linear muffin-tin orbital
method. The experimentally observed reduction of the critical field of the
itinerant electron metamagnetic phase transition with increasing content of
non-magnetic Al is explained. It is shown, on the basis of a $T = 0$ K
Stoner type itinerant magnetism theory, that the alloying-induced changes in
the shape of the calculated density of states, caused by the Al substitution, lead
to (i) a stabilization of the magnetic state, (ii) a smoothening of the first-order
metamagnetic transition and (iii) a subsequent suppression of the metamagnetic
transition around $x = 0.15$. Analysing the magnetization processes in
$Y(Co_{1-x}Al_x)_2$ by varying the strength of the exchange interaction, we provide
a microscopical background to earlier phenomenological assumptions made in
the literature.

1. Introduction

The Laves phase compounds $Y(Co_{1-x}Al_x)_2$ ($x \leqslant 0.12$) show metamagnetic transitions from
paramagnetic to a ferromagnetic state in an applied magnetic field [1, 2]. This compound
was among the first where an itinerant electron metamagnetic transition was observed exper-
imentally. In addition, in the concentration range $x \approx 0.12$–0.20 spontaneous ferromagnetic
order occurs and a weak ($\approx 0.1 \mu_B$/Co) magnetic moment is detected [3]. This finding added
$Y(Co_{1-x}Al_x)_2$ to the small number of materials which appear to satisfy all the theoretical
criteria for weak itinerant ferromagnetism. Although $Y(Co_{1-x}Al_x)_2$ is a random alloy the
sharpness of the magnetic transition and the lack of sensitivity of the magnetic properties to
small fluctuations in composition near the optimum value $x = 0.15$ makes it superior to $ZrZn_2$,

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Ni_3Al or Sc_3In [4]. With all these properties $Y(Co_{1-x}Al_x)_2$ is a prototypical compound for theoretical and experimental studies of various aspects of itinerant magnetism [5–7].

Itinerant electron metamagnetism (IEM) has been the subject of intense theoretical and experimental studies for more than 30 years, starting from the pioneering work of Wohlfarth and Rhodes [8] where they predicted the possibility of a first-order phase transition in the itinerant electron system from the paramagnetic to a magnetic state in an applied magnetic field when certain conditions are satisfied. This IEM theory has been derived on the basis of the Stoner theory of itinerant magnetism.

From the very beginning the paramagnetic intermetallic compound YCo_2 was a most promising candidate to show such a type of behaviour due to a pronounced maximum in the temperature dependence of the susceptibility [9]. The very sharp IEM transition in this compound and also in isostructural $LuCo_2$ was found only in the late 1980s by Goto *et al* [10] at an applied critical field of about 70 T. However, even before these observations the IEM concept was widely used to explain the magnetic properties of the RCo_2 (R = rare earth) systems [5, 6] which in turn caused a further development of the IEM theory [11]. In particular, it has been shown that the first-order phase transitions in heavy RCo_2 are strongly related to the metamagnetic properties of the Co sublattice [12] (see also recent discussions in [13–15]).

Various further intense studies [1, 2, 16, 17] of the IEM transition in $Y(Co_{1-x}Al_x)_2$ have confirmed that the value of the critical field decreases with increasing Al concentration x. While in YCo_2 the first-order IEM transition manifests itself as a sharp jump in the magnetization at the critical field it becomes broader in $Y(Co_{1-x}Al_x)_2$ when the Al concentration is increased up to $x \leqslant 0.12$, simultaneously with a decrease of the value of the critical field [2]. For higher concentrations a weak ferromagnetic ground state (WFS) is stabilized in $Y(Co_{1-x}Al_x)_2$ [3]. At these concentrations a strong but continuous increase of the magnetization in applied magnetic field has been found [18]. However, even in these cases the inflection points of the strongly non-linear Arrott plots were interpreted as IEM transitions from a WFS to a strongly ferromagnetic (SF) state [18]. Recently Takahashi and Sakai [19, 20] have proposed a novel theory of IEM transitions which is based on the assumption that the magnetization process can be described in terms of the spin fluctuation spectrum in contrast to conventional IEM theory [8] which is based on the fine structure of the density of states (DOS) curve. For $Y(Co_{1-x}Al_x)_2$ they assumed that there are two different electronic states with different spin-fluctuation spectra: paramagnetic and weakly ferromagnetic. At the concentration $x \approx 0.19$–0.20 the weakly ferromagnetic state becomes stable and with further decrease of x, for $x < 0.12$, the paramagnetic state, which is related to a different electronic state, becomes stable with respect to the ferromagnetic state [19, 20]. For this concentration range ($x < 0.12$) the metamagnetic transition between these two states is observed in applied magnetic field. It should be noted that in this model the weakly ferromagnetic ground state for concentrations $x \approx 0.12$–0.20 and the ferromagnetic state induced by an applied magnetic field at $x < 0.12$ (in particular also the high-moment state of YCo_2 at $H > 70$ T) are assumed to have the same electronic origin.

The reduction of the critical field with increasing x in $Y(Co_{1-x}Al_x)_2$ has no unambiguous explanation. In a recent review article on the subject Gratz and Markosyan [7] have summarized the three existing attempts to explain this reduction:

 (i) a shift of the DOS at Fermi level E_F due to a change of the d-electron concentration,
 (ii) a change of the d bandwidth due to the increasing lattice parameter and
(iii) hybridization between the 3d states of Co and the 3p states of Al.

While for YCo_2 there exists a description of the IEM transition from the point of view of band structure calculations [21, 22] such a systematic treatment is absent for the $Y(Co_{1-x}Al_x)_2$

compounds. Aoki and Yamada [23] have performed non-spin-polarized self-consistent band structure calculations for the fictitious ordered Y(Co$_{0.75}$Al$_{0.25}$)$_2$ compound employing the augmented plane wave method. In an earlier paper [24] we presented a non-magnetic *ab initio* study of the disordered Y(Co$_{1-x}$Al$_x$)$_2$ employing the coherent potential approximation (CPA) for the whole range of the concentration x and discussed the formation of weak itinerant ferromagnetism in these compounds on the basis of the paramagnetic DOS.

Here we present results from *ab initio* spin-polarized band structure calculations for Y(Co$_{1-x}$Al$_x$)$_2$ compounds within density functional theory and the local spin density approximation (DFT–LSDA) for various concentrations of Al up to $x = 0.25$. The effects of alloying are treated within the CPA. Since there is no unique unambiguous interpretation of the results of the high-field magnetization measurements near and above the critical Al concentration ($x \geqslant 0.10$–0.12), the analysis of the magnetization processes in Y(Co$_{1-x}$Al$_x$)$_2$ from *ab initio* results is paid special attention.

From a general point of view, the metamagnetic transitions which occur in YCo$_2$ and Y(Co$_{1-x}$Al$_x$)$_2$ are the result of the existence of two almost degenerate local minima of the total energy $E(M)$ with a very small energy difference (about 1 mRyd/fu). It is well known that LSDA has a tendency to overestimate the band splitting in the magnetic ground state of systems with initially small magnetic moments. In the case of the strong ferromagnets, for example pure Co and Ni, this LSDA shortcoming is not essential for the determination of the magnetic ground state properties. However, when one wants to analyse the magnetization processes in Y(Co$_{1-x}$Al$_x$)$_2$, where tiny energy differences are involved, the direct use of the calculated total energies from the LSDA meets certain difficulties. Despite this fact, the calculated band structures provide a good starting point for the further analysis of the evolution of the magnetic properties with increasing Al concentration.

Our paper is organized in the following way: in the next section we describe the technical details of our *ab initio* spin-polarized calculations with particular emphasis on the above-mentioned problems of the LSDA. In section 3 the results of the LSDA–CPA calculations are presented. From these results it follows that in general, apart from the absolute values of the total energy differences, the LSDA results quite well explain the evolution of the magnetic properties with increasing Al content. In particular, the fact that substitution of Al makes the magnetic state more stable is well described. Finally, in section 4, we introduce a simple semiempirical model which is based on the calculated DOS and a pure Stoner picture of magnetic interactions. Within this model it becomes evident how the changes in the structure of the DOS caused by the presence of Al influence the metamagnetic properties of the system. One of the important results of our analysis is that it strongly supports the Takahashi–Sakai [19, 20] scenario, providing a microscopical background to the phenomenological assumptions of their theory.

2. Method of calculation

The intermetallic compound YCo$_2$ crystallizes in the C15 cubic Laves phase (MgCu$_2$ type, $Fd3m$). In Y(Co$_{1-x}$Al$_x$)$_2$ the Al and Co atoms randomly occupy Cu (Co) positions. The electronic structure of the substitutionally disordered Y(Co$_{1-x}$Al$_x$)$_2$ systems was calculated by the all-electron self-consistent tight-binding linear muffin-tin orbital (TB-LMTO) method in the atomic-sphere approximation (ASA) [25] combined with the CPA [26]. The valence bands were treated with an angular momentum cut-off $\ell_{max} = 2$; the integrations over the Brillouin zone (BZ) were performed using a uniform mesh corresponding to 8000 k-points in the full BZ. The energy integrations in the self-consistency loop were carried out using 14 nodes along a semicircle contour in the complex energy plane. The densities of states were obtained from

the self-consistent one-electron potentials using the Green function quantities calculated at energies with a small imaginary part ($\sim$10 mRyd) followed by numerical analytic continuation to the real axis [27]. The ratio of the Wigner–Seitz radii ascribed to the inequivalent sites of the C15 Laves phase was chosen as 1.25 according to previous studies for the ordered systems [21]. The radii of the Co and Al atomic spheres were set equal to each other for the whole composition range ($0 \leqslant x \leqslant 0.25$).

As the energy differences between the non-spin-polarized (NSP) and SF states involved in the IEM are quite small, their sensitivity to the computational details was studied carefully. Particular attention was paid to the inclusion of the relativistic effects and the local exchange–correlation (XC) potential. The calculated local quantities (electron charges and magnetic moments in the atomic spheres) for the pure YCo_2 at the experimental lattice constant ($a = 7.215$ Å) were hardly sensitive to these computational details. However, the calculated total-energy differences $E_{SF} - E_{NSP}$ for this system fall within a few mRyd range around zero, ranging from -5.1 mRyd/fu for a scalar-relativistic approximation [28] and the XC potential of von Barth and Hedin [29] to $+1.0$ mRyd/fu for a non-relativistic calculation with the XC potential according to Perdew and Zunger [30]. In the scalar-relativistic approximation and the XC potential of Vosko *et al* [31], reasonable agreement of our ASA calculations with results of a full-potential treatment [32] was obtained; the energy difference in both approaches was $E_{SF} - E_{NSP} \approx -4$ mRyd/fu. On the other hand, the only positive value of $E_{SF} - E_{NSP}$ (which yields a NSP ground state in qualitative agreement with experiment) was obtained in the non-relativistic approximation and the XC potential as parametrized in [30]. In view of the main aim of the present study, namely the description of alloying effects on magnetic properties, the latter option was used in the systematic calculations reported here.

Let us make a comment on the use of the calculated total energy difference between magnetic and paramagnetic states in this work. This difference is roughly proportional to the value of the critical magnetic field that should be applied for a given compound $Y(Co_{1-x}Al_x)_2$ in order to induce the IEM transition to the high-moment magnetic state (at least for the $x < 0.12$). For most of the exchange correlation potentials which were investigated during the preparation of this work, the results were similar to previous investigations [22, 32], namely, that for pure YCo_2 at the experimental lattice constant the total energy of the high-moment state is lower than the total energy of the pure paramagnetic state. This result is of course in contrast to the experimental situation where the ground state is paramagnetic and the transition to the magnetic state appears only at high magnetic fields. This qualitatively right order of the total energies was obtained only if the lattice constant was reduced in the calculations towards the theoretical DFT total energy minimum [22, 32]. Only for the XC potential parametrization according to Perdew and Zunger [30] did we find that the total energy at the paramagnetic state becomes lower than that of the high-moment state even at the experimental lattice constant. However the problem of the magnetic ground state reenters again for $Y(Co_{0.95}Al_{0.05})_2$ also when the Perdew and Zunger parametrization is used. This means that one cannot directly use the calculated difference $E_{SF} - E_{NSP}$ to estimate the critical field due to the inherent error of the LSDA as discussed above.

Apart from the above-mentioned difficulties, however, the *ab initio* results fairly well predict the mechanism of the appearance of the magnetic instability in $Y(Co_{1-x}Al_x)_2$ for $x > 0.12$ [24] and generally show that the substitution of Al favours the high-moment states energetically, but at the same time lead to a total moment reduction in the high-moment state.

Concerning possible local environment effects on the magnetic state of Co in $Y(Co_{1-x}Al_x)_2$, which within CPA are treated in lowest order, we note that NMR experiments [33] found that all Co atoms are essentially equivalent in these compounds, i.e. their magnetic state does not depend on the number of Co neighbours.

Table 1. Experimental values of the lattice constant as given by Yoshimura and Nakamura [3]. The lattice constant for $x = 0.25$ is derived by extrapolation as in [23]. Calculated magnetic moments: m_{Co} and m_Y are Co and Y moments, m_{total} is the total moment averaged per cobalt atom in the high-moment state and $E_{SP} - E_{NSP}$ the total-energy difference taken between the high-moment state and the paramagnetic state of Y(Co$_{1-x}$Al$_x$)$_2$. All calculations were done employing the Perdew–Zunger parametrization [30] for exchange and correlation.

| x, Y(Co$_{1-x}$Al$_x$)$_2$	0.00	0.05	0.10	0.15	0.20	0.25
a, lattice constant (Å)	7.215	7.244	7.275	7.312	7.342	7.380
m_{Co}, μ_B/Co	0.98	0.90	0.78	0.68	0.59	0.54
m_Y, μ_B/Y	−0.27	−0.25	−0.22	−0.19	−0.16	−0.14
m_{total}, μ_B/Co	0.84	0.76	0.65	0.56	0.48	0.44
$E_{SP} - E_{NSP}$ (mRyd/fu)	1.04	−0.68	−1.53	−1.70	−1.27	−0.88

3. High-moment state in Y(Co$_{1-x}$Al$_x$)$_2$

The experimental lattice constants for the various Al concentrations used in the calculations were taken from Yoshimura and Nakamura [3] and are given in table 1 together with the calculated magnetic moments and total-energy differences $E_{SF} - E_{NSP}$. For pure YCo$_2$ the values of the magnetic moment in the high-moment state is in good agreement with earlier fixed spin moment studies [21] ($m_{total} = 0.88$ μ_B/Co) and LCAO spin-polarized calculations [32] giving $m_{Co} = 0.99$ μ_B and $m_Y = -0.22$ μ_B ($m_{total} = 0.88$ μ_B/Co). Since from experiment only the total moment per Co atom m_{total} in the high-moment state is readily accessible we give these values in table 1 for comparison. For pure YCo$_2$ at $H > H_c$ and $T = 10$ K the experimental value of this moment is about 0.55 μ_B/Co [10] while our calculated one is 0.84 μ_B/Co. For finite Al concentrations this disagreement becomes smaller. For concentrations $x < 0.1$ Sakakibara *et al* [17] found in fields larger than H_c moments of about 0.5–0.6 μ_B/Co. However, it should be noted that in the RCo$_2$ compounds where the field exerted by the rare earth ion saturates the Co moment the magnetic moment of Co is exactly in the range of about 0.8–1.0 μ_B/Co (see e.g. figure 5 of [7]).

The Y moment in Y(Co$_{1-x}$Al$_x$)$_2$ has the opposite sign to the Co moment in these compounds and provides a sizeable contribution to the total moment whereas the Al moments we found to be negligibly small, being $\approx$0.02–0.04 μ_B, a behaviour which is well known for s–p atoms.

The results of the calculations at the experimental lattice constants show that a high-moment state exists in Y(Co$_{1-x}$Al$_x$)$_2$ over the whole range of the Al concentration. The value of the moment in this state changes continuously as the concentration x increases. However, it remains unclear whether the high-moment state we calculate for $x = 0.15$ can be associated with the observed weakly ferromagnetic ground state in this compound, or whether it is just a metastable state. The answer on this question is strongly related to the discussion given in the introduction namely whether the inflection point of the Arrott plots for concentrations $x > 0.11$ can be interpreted as first-order phase transitions from weakly ferromagnetic states to high-moment states or whether the Takahashi–Sakai [19] scenario is valid where the weakly ferromagnetic ground state is assumed to be developed from the high-moment state as the Al concentration increases. At the present stage this question cannot be answered using bare LSDA calculations because the energy differences are too small to be treated with any degree of certainty. However, since the possibility of the IEM transitions is strongly related to the actual shape of the DOS we apply a Stoner-like model based on the calculated *ab initio* DOS in order to study the magnetization processes in Y(Co$_{1-x}$Al$_x$)$_2$ by varying the strength of the exchange interaction.
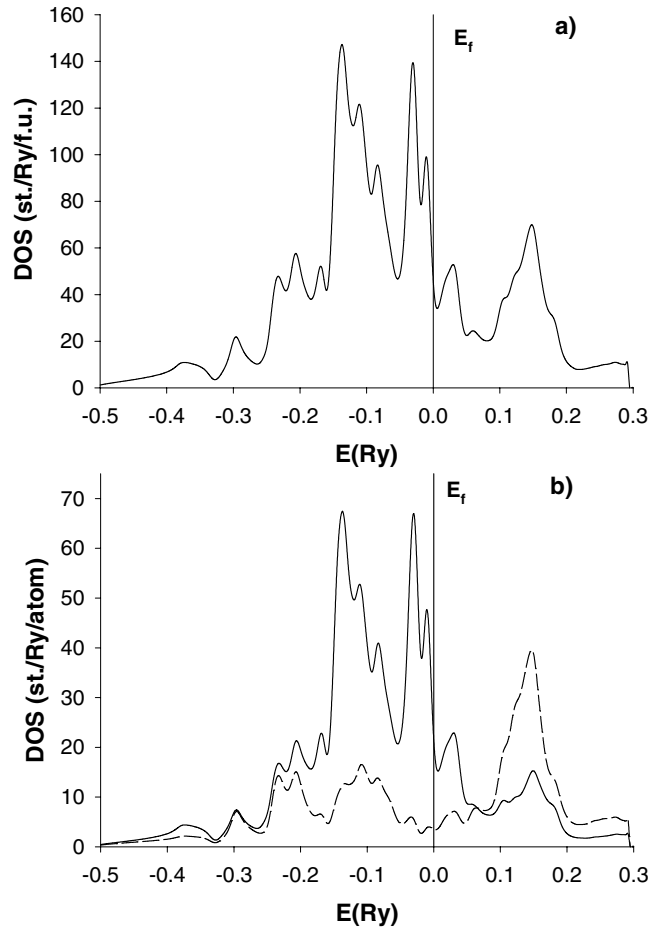
Figure 1. (a) Total DOS (states/Ryd/fu) of YCo$_2$; (b) atom-resolved DOS of YCo$_2$; Co, full curve; Y, dashed curve.

The calculated energy differences between the high-moment and paramagnetic states are given in the bottom row of table 1. Keeping in mind the discussion given above, it follows that the Al substitution strongly favours the high-moment states (the difference of the total energy $E_{SF} - E_{NSP}$ decreases) causing a subsequent lowering of the critical field. This result is in fair agreement with the reductions of the critical field which are found in experiment for $x < 0.12$ [2, 18]. For high Al concentrations the metamagnetic phase transition becomes very weak [2], making a determination of the transition point very uncertain. This weakening is also strongly related to the changes in the shape of the DOS.

The possibility of the IEM phase transition in strongly enhanced paramagnetic YCo$_2$ is connected with the well known structure of the DOS of this compound which has been published several times (see e.g. [21, 34]). The Fermi level in this compound is placed between two peaks of the DOS (figure 1) in the sharply falling region of the pronounced DOS peak just below E_F. The Stoner factor is close to unity [32, 34], indicating that the system is on the verge of a magnetic instability. This causes a strong exchange enhancement, with a strong temperature variation of the susceptibility. It has been shown that the appearance of the magnetic instability in Y(Co$_{1-x}$Al$_x$)$_2$ for $x > 0.12$ is connected to the increase of the value of the DOS at the
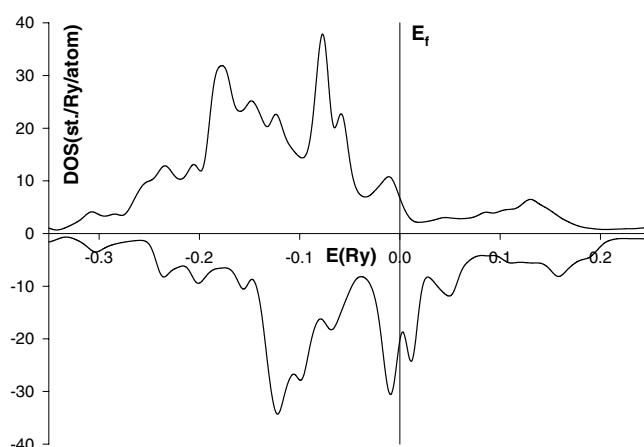
Figure 2. Spin-polarized Co-projected DOS of YCo$_2$ in the high-moment state.

Fermi level due to the broadening of the DOS peaks above and below E_F due to the increasing disorder of the Co sublattice caused by the Al substitution [24].

In figure 2 we show the spin-polarized Co DOS of pure YCo$_2$ in the metastable high-moment state. One notes that the broad peak which is above E_F for the Co-projected paramagnetic DOS is now below E_F for the spin-up DOS and simultaneously the position of the sharp peak moves above E_F for the spin-down DOS. This immediately suggests that for the determination of the expansion coefficients of the total energy in powers of the magnetization it is not sufficient to just rely on the values of the DOS near the Fermi level in the paramagnetic state as standard theories of IEM propose [8]. In order to describe the IEM transition in YCo$_2$ one needs to include the DOS structure also further off the paramagnetic Fermi level. More precisely, the magnetic splitting of the bands in the high-moment state of YCo$_2$ is about 0.08 Ryd and within this energy interval the DOS has a non-trivial structure (two peaks).

In figure 3 the DOS of the Y(Co$_{1-x}$Al$_x$)$_2$ at two concentrations ($x = 0.1$ and 0.15) for the high-spin state is shown. The substitution of Al at Co sites leads to a broadening of the DOS peaks. As a result the two peaks above and below E_F progressively disappear and form a common falling portion of the DOS (figure 3). While for $x = 0.1$ the relics of these peaks can still be seen, for $x = 0.15$ just a smooth decreasing structure of the DOS remains. Figure 4 shows these alloying-induced changes in greater detail. At the same time the splitting of the up and down bands decreases to $\approx$0.06 Ryd. These changes in the shape of the DOS cause a qualitative difference in the magnetization process under an applied external field for the compounds with low and high Al contents independently of the strength of the exchange interaction.

4. General features of the magnetization processes in Y(Co$_{1-x}$Al$_x$)$_2$

In the previous section we pointed out the drastic changes in the shape of the DOS in Y(Co$_{1-x}$Al$_x$)$_2$ which appear due to addition of Al in ordered YCo$_2$. In order to look at the influence of these changes in the DOS more closely, we apply a straightforward $T = 0$ K Stoner model of itinerant magnetism [11] but employ the full *ab initio* calculated DOS rather than making the usual Sommerfeld type expansions.

The basic assumptions of the model are the following.
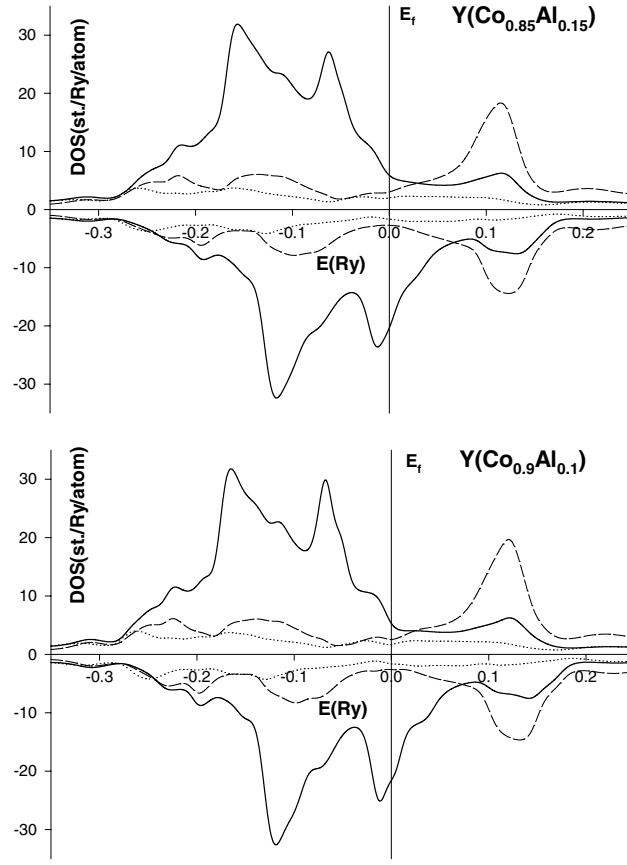
Figure 3. Spin-polarized atom projected DOS (states/Ryd/atom) of $Y(Co_{0.85}Al_{0.15})_2$ and $Y(Co_{0.9}Al_{0.1})_2$: full curve Co; dashed Y; dotted Al.

(i) The magnetic splitting of the up and down 3d bands of Co is rigid. This assumption is justified by the comparison of the *ab initio* calculated DOS in the paramagnetic and high-moment states of $Y(Co_{1-x}Al_x)_2$ (see e.g. figures 1– 3).

(ii) The exchange interaction is treated using an effective (Stoner) parameter I and is taken to be equal to $-IM^2$ [11], where M is the magnetization of the Co atoms.

Compared to the usual treatment of itinerant electron magnetism, this model has the advantage of accounting for the DOS in an exact way. As it was shown in the previous section this feature is important in case of $Y(Co_{1-x}Al_x)_2$ with low Al concentration since the DOS has a pronounced peak in the vicinity of the Fermi level. However, the results of this model can be regarded only as semi-quantitative due to the simplifications that Stoner theory makes on the character of the exchange interaction. The usefulness of this approach for our purposes follows from the fact that it allows us to vary the strength of the exchange interaction in order to study the universal properties of the magnetization process at $T = 0$ K which are related entirely to the shape of the DOS of the given compound.

Putting the paramagnetic (zero-band-splitting) Fermi energy equal to zero the magnetic part of the energy reads

$$F(M) = E(M) - E(0) = \int_0^{\Delta_1} \varepsilon N(\varepsilon)\,d\varepsilon - \int_{-\Delta_2}^0 \varepsilon N(\varepsilon)\,d\varepsilon - IM^2 - HM \tag{1}$$
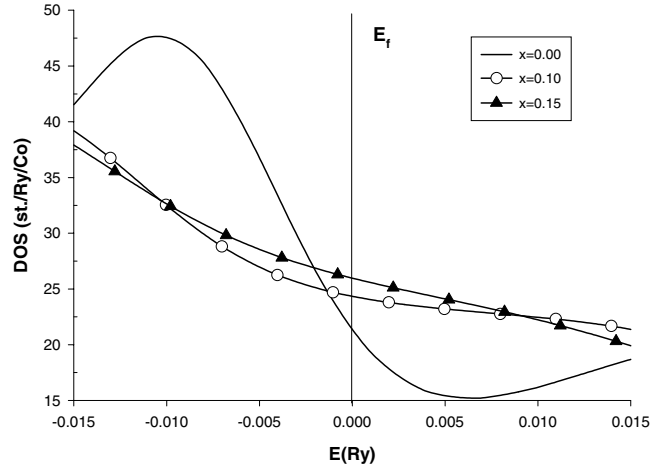
Figure 4. Atom-projected DOS (states/Ryd/atom) of Y(Co$_x$Al$_{1-x}$)$_2$ for $x = 0.00$, 0.10 and 0.15 near the Fermi energy. The changes in the value of the DOS at E_F and in the respective curvature are responsible for the dramatic changes of the magnetization behaviour.

where $N(\varepsilon)$ is the Co-projected DOS taken per spin direction, M is the magnetization and H is the applied external field. The quantities Δ_1 and Δ_2 are the respective shifts of the up- and down-spin bands in the magnetic state, so that the total band splitting amounts to $\Delta = \Delta_1 + \Delta_2$. The first two terms in equation (1) express the increase in kinetic energy due to the band splitting, the third term is Stoner exchange and the last one is the energy from the applied magnetic field. It is obvious that the values of M, Δ_1 and Δ_2 depend on each other. By fixing one of them the remaining two can be determined from the expressions

$$\int_0^{\Delta_1} N(\varepsilon)\, d\varepsilon - \int_{-\Delta_2}^0 N(\varepsilon)\, d\varepsilon = 0 \tag{2}$$

$$M = \int_{-\Delta_2}^{\Delta_1} N(\varepsilon)\, d\varepsilon \tag{3}$$

where equation (2) is the condition that the number of electrons is conserved.

Using the calculated DOS for non-spin-polarized Y(Co$_{1-x}$Al$_x$)$_2$ and choosing the value of the effective exchange constant I, the magnetization curves $M = M(H)$ can be drawn by fixing the value of the external field H and carrying out the numerical minimization of the magnetic energy (equation (1)) under the additional conditions provided by equations (2) and (3).

Here we must note that the conventional way to estimate the effective exchange constant from *ab initio* calculations cannot be applied in the present case. The usual procedure would be to calculate the *ab initio* energy difference between a magnetic and the non-magnetic state and to divide it by the square of the respective magnetic moment, an approach which is strictly only valid in the paramagnetic limit [35]. Even if this method works well for most of the ferromagnetic materials and provides reasonable values for I, in the present case, where one is always very close to a magnetic instability and has to deal with extremely small energy differences, tiny uncertainties in the exchange parameter lead to dramatic changes in the magnetization process.

In Stoner theory the exchange parameter I is related to the intra-atomic exchange integral and for a given type of atom in a metallic solid depends only slightly on the actual chemical composition. In the present case of Y(Co$_{1-x}$Al$_x$)$_2$ all physically significant results can be
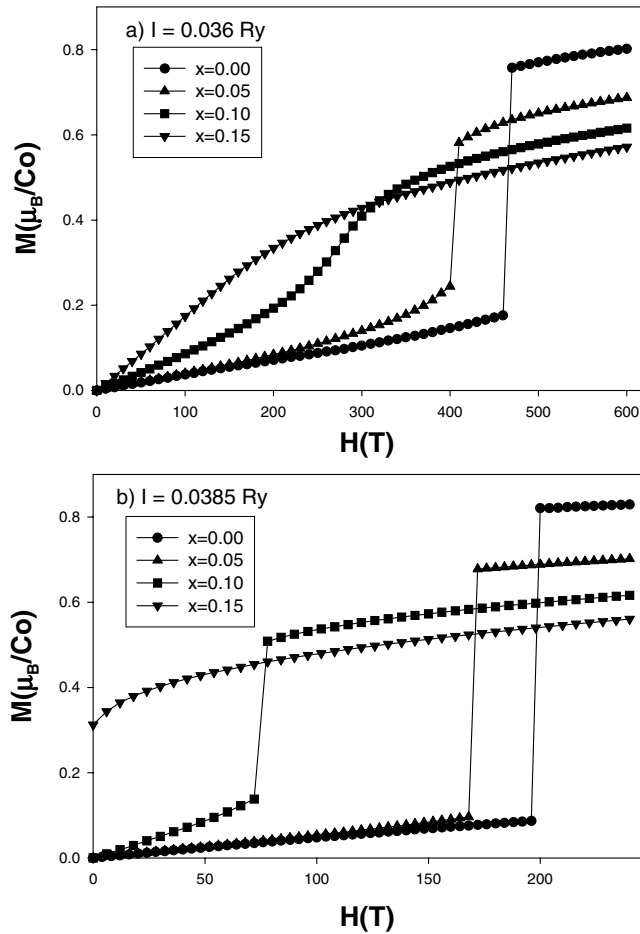
Figure 5. Magnetization curves of $Y(Co_{1-x}Al_x)_2$ for varying Al concentrations for two different values of the exchange parameter I.

obtained by assuming I to be in the range 0.035–0.040 Ryd, being close to the value of 0.036 Ryd as calculated for pure Co [36]. We start our investigation taking this 'canonical' value $I = 0.036$ Ryd for cobalt and calculate the respective magnetization curves (figure 5(a)). The first things to note are qualitatively different magnetization processes for pure YCo_2 ($x = 0$) and for $x = 0.05$ as compared to those for $x = 0.10$–0.15. While in the first two cases a first-order metamagnetic transition occurs, the magnetization curves for the latter ones are continuous. At the same time the magnetic susceptibility (first derivative with respect to the field) increases with increasing Al concentration. It can be concluded that smoothening of the DOS due to alloying drives the system even closer to the magnetic instability (this point that has been discussed in detail in [24]) and concomitantly the sharpness of the IEM transition becomes smeared out. These features are also observed experimentally [2]. The ground state at $H = 0$ of $Y(Co_{0.85}Al_{0.15})_2$ is non-magnetic under the canonical value of I, and also the critical field of the IEM of YCo_2 is much too high as compared to the experimental value of 70 T.

For the curves presented in figure 5(b) we used a value $I = 0.0385$ Ryd. In this case the critical field of YCo_2 is greatly reduced towards the experimental value, and $Y(Co_{0.9}Al_{0.1})_2$ becomes metamagnetic as well. Now $Y(Co_{0.85}Al_{0.15})_2$ is ferromagnetic in the ground state

with moment ∼0.32 μ_B/Co. These results resemble the experimental situation much better. Since the model applied here is also approximate (the approximation lies in the fact that I is taken as an effective mean field parameter) the ideal value for I satisfying all experimental values for all the concentrations is hard to find. From the comparison of the results given in figures 5(a) and (b) it follows that, depending on the value of the exchange interaction Y(Co$_{0.9}$Al$_{0.1}$)$_2$ is either a metamagnet showing a first-order phase transition or just a strongly enhanced paramagnet. If we further increase the value of the exchange parameter, for example by fitting it to the experimental value of the critical field of pure YCo$_2$, also Y(Co$_{0.9}$Al$_{0.1}$)$_2$ will show a magnetic ground state. It must be noted that there does not exist a single value of I where Y(Co$_{0.9}$Al$_{0.1}$)$_2$ shows a magnetic ground state and simultaneously a metamagnetic first-order phase transition under an applied field.

To investigate the possibility of a first-order phase transition from a low- to a high-moment state we performed a set of calculations for Y(Co$_{0.85}$Al$_{0.15}$)$_2$ which are shown in figure 6. It can be seen that such a situation never occurs; even more a strict statement can be formulated: the DOS of Y(Co$_{0.85}$Al$_{0.15}$)$_2$ has a structure that does not allow for a first-order IEM transition to occur at all. This compound can be either magnetic or paramagnetic, but not metamagnetic because all magnetization curves given in figure 6 are continuous. However, as can be seen, at certain values of the effective exchange (e.g. $I = 0.0384$ Ryd) the Arrott plot for this compound can become extremely nonlinear.

Turning back to figure 5(b), we see that the magnetic ground state at higher Al concentrations becomes developed from the high-moment state which is metastable at low Al concentrations in full agreement with the phenomenological assumptions made by Takahashi and Sakai [19, 20].

To interpret the results of this section from the point of view of the changes in the DOS caused by the Al substitution (figures 1–3) we note that a peak structure near the Fermi level which can be found in YCo$_2$ progressively disappears with increasing Al content. The pronounced first-order phase transition can be found only for compositions where this peak structure of the DOS can still be detected (as in Y(Co$_{0.9}$Al$_{0.1}$)$_2$—see figure 3). For Y(Co$_{0.85}$Al$_{0.15}$)$_2$ the DOS already shows a smooth falling portion in the vicinity of the Fermi level which makes the magnetization process also smooth.

5. Conclusion

In this paper we have shown that the pronounced changes in the magnetization processes in Y(Co$_{1-x}$Al$_x$)$_2$ are related to the progressive smoothening of the DOS with increasing Al concentrations. The weakly ferromagnetic ground state at increased concentrations of Al develops from the high-moment metastable state which at lower concentrations is only accessible via a first-order IEM. The latter observation provides a microscopical background for the earlier phenomenological assumptions made by Takahashi and Sakai [19]. The smoothening of the DOS leads to the disappearance of the peak structure near the Fermi level and causes an increase of the DOS at the Fermi level [24] which finally leads to the observed magnetic instability at Al concentrations $x > 0.12$. At the same time, this mechanism also leads to the suppression of the possibility of a first-order metamagnetic transition. For intermediate Al concentrations the smoothening of the DOS causes a progressive smearing out of the originally sharp metamagnetic transition. The magnetization processes can be described on the basis of a $T = 0$ K Stoner model using the *ab initio* calculated DOS for varying Al content. The calculated changes in the magnetization behaviour follow the experimentally observed trends. From our results we conclude that these changes are solely driven by the alloying-induced variations of the DOS of the Co–Al sublattice.
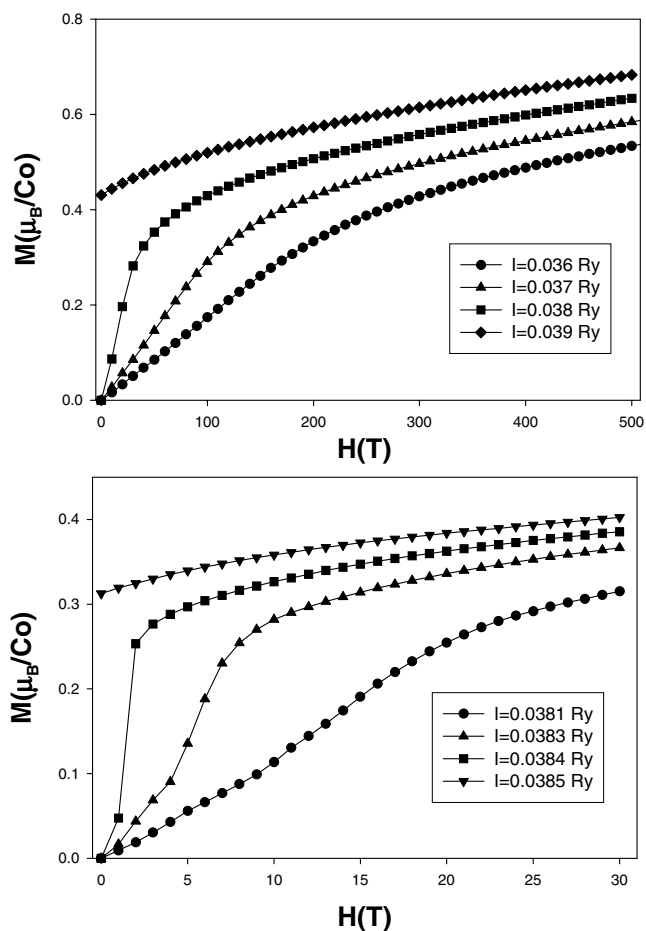
Figure 6. Magnetization curves for $Y(Co_{0.85}Al_{0.15})_2$ for various values of I.

Acknowledgments

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