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ABSTRACT

Magnetic phenomena observed in *R*-Co intermetallic compounds with the *d*-magnetism instability are reviewed. The magnetic instability in these compounds is intimately related to the special position of the Fermi level in the hybridized 3d-5d (4*d*) band near to a local peak in $N(\varepsilon)$. In the presence of the *f*-*d* exchange interaction the magnetic state of the itinerant electron subsystem can essentially be modified giving rise to a number of field- and temperature-induced magnetic phase transitions. Following the band structure calculations these transitions as well as most of their fine details can be well understood theoretically. Magnetic, magnetoelastic and transport measurements of some *R*-Co compounds with *d*-magnetism instability and pseudobinary systems with *R* and Co substituted by either magnetic or nonmagnetic elements are presented and discussed.

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1. Introduction

In rare earth (RE) – cobalt, *R*-Co, intermetallic compounds the Co itinerant magnetic sublattice shows a variable magnetic moment. It shows a paramagnetic behaviour in compounds of the RE-rich side (R_3 Co), is ferromagnetic with a stable magnetic moment of 1.6 μ_B /Co in the Co-rich side (R_2 Co₁₇) (Fig. 1) [1–3]. In the middle of this series the Co magnetic moment substantially depends on the RE sublattice, i.e. the type of the RE ion. In RCo₂ intermetallics the Co sublattice changes from a paramagnetic to a ferromagnetic state depending on the strength of the *f*-*d* exchange interaction (molecular magnetic field) and changes from a weak to strong magnetic state in RCo₃ and R_4 Co₃ compounds (see, e.g. Ref. [3]).

The magnetic properties of *R*-Co intermetallic compounds with instable Co magnetic sublattice show in general more diverse and richer behaviour compared to the compounds with stable itinerant-electron magnetic sublattice. In this article some of the most characteristic effects the *R*-Co intermetallics exhibit due to the Co magnetism instability are reviewed. Much work in this field, especially in studying field-induced magnetic phase transitions in RCo_2 and RCo_3 intermetallic compounds was done by Peter Brommer with the colleagues [4–9].

Nature of magnetism is different in two electron subsystems involved in the magnetic interactions in the *R*-Co intermetallics.

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E-mail address: ashotm@stanford.edu (A.S. Markosyan). Peer review under responsibility of Vietnam National University, Hanoi. Most of the lanthanide ions retain the localized atomic character of the 4*f* orbitals and their magnetism can be well described by atomic characteristics, *L*, *S* and *J*, of a free R^{3+} ion. In contrast, the 3*d*-electrons of cobalt sublattice are itinerant and the 3*d*-states form an energy band crossed by the Fermi level ε_f with natural consequences for magnetism (see, e.g., Ref. [1,3]). The interaction between the RE and Co sublattices occurs mostly through hybridization of the 5*d* (4*d*)-states of RE and the 3*d*-states of the transition metal, which mediates the strength of the 4*f*-3*d* exchange interaction.

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The effect of the RE sublattice on the magnetic properties of the *d*-subsystem is in most cases considered as resulting in an additional shift of the majority and minority *d*-subbands, whereas the effect of the *d* electrons on the RE sublattice consists in the modification of the energy level scheme of the R^{3+} ions.

Because of a spatial localization of the 4*f* electronic shells, no direct overlap between the 4*f* wave functions takes place in the *R*-3*d* intermetallics and the *f*–*f* exchange occurs via the conduction electrons. The interactions related to the *d*-sublattice increase successively along with the content of the transition metal and the d-d interaction becomes dominating in the Co-rich compounds.

2. Itinerant magnetism of the d-electron subsystem and density of states - theoretical background

The main distinct feature of *d*-magnetism in *R*-3*d* intermetallics, which makes the magnetic properties of the Co sublattice

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Fig. 1. Co-magnetic moment versus Y and Gd content in Y-Co and Gd-Co intermetallic compounds [2].

dependent on stoichiometry, is the hybridization between the narrow 3*d* band (~3 eV) of the transition metal with a high density of states (DOS), $N_{3d}(\varepsilon)$, and the broader 5*d* band (~10 eV) of lanthanide (the 4*d* band of Y) with lower DOS. The contribution to the total DOS from the 6*s* (5*s*) band is negligible because of low $N_{s}(\varepsilon)$. The magnetic properties of the *d*-electron subsystem are hence determined by the energy dependence of $N_{d}(\varepsilon)$ near the Fermi level, ε_{f} and the position of ε_{f} itself [2,3,8].

The most known *R*-Co intermetallics with the *d*-magnetism instability are the *R*Co₂ compounds, in which the *d*-electron subsystem exhibits itinerant electron metamagnetism (IEM), i.e., a first-order field-induced magnetic phase transition from a paramagnetic to ferromagnetic state [10–12]. For the YT₂ compounds with T = Fe, Co and Ni several authors have published band structure calculations. Although for these calculations different methods have been used (e.g. Ref. [13,14]), the common result of all these calculations confirms the existence of a strong hybridization between the 3*d* states of the transition metal and 4*d* states of yttrium (or 5*d* states in the case of a lanthanide). The calculated energy dependence of DOS is qualitatively similar in shape for all these intermetallics. At low energies $N(\varepsilon)$ exhibits a relatively narrow peak (due to the 3*d* electronic states) followed by a flat range with lower DOS at greater energies (primarily due to the 4*d* states).

In Fig. 2, $N(\varepsilon)$ near ε_f of YFe₂, YCo₂ and YNi₂ are compared [13]. Among them YNi₂ has the lowest value of $N(\varepsilon_f)$. The Stoner criterion of ferromagnetism $IN(\varepsilon_f) \ge 1$ (I is the d-d exchange integral) is by far not fulfilled, the product $IN(\varepsilon_f) = 0.21$. YNi₂ is nonmagnetic and shows a very weak temperature dependence of susceptibility. In contrast, $IN(\varepsilon_f) = 2.6$ for YFe₂, which is therefore is a ferromagnet with a spontaneous magnetization $M_S = 1.4 \mu_B/Fe$ at 4.2 K. Since M_S of YFe₂ is considerably smaller than M_S for metallic Fe (=2.2 μ_B/Fe), YFe₂ is a non-saturated ferromagnet, i.e. the spin-up and spindown bands both are not filled. For YCo₂ the Stoner criterion is nearly fulfilled: $IN(\varepsilon_f) = 0.9$. This causes a strong exchange enhancement with a pronounced temperature variation of the magnetic susceptibility. The average value of χ is much larger than the Pauli susceptibility.

For YT₃ compounds, the calculated energy dependence of DOS is shown in Fig. 3 [14]. The shapes of DOS of YFe₃, YCo₃ and YNi₃ are again more or less similar to each other as in the case of YT₂ compounds. While ε_f of YFe₃ is located near the highest peak of the DOS that of YCo₃ is located near a steep descent of the *N*(ε) and that of YNi₃ is located just above a small peak. As a result, YCo₃ is a weak



Fig. 2. Calculated local DOS of the 3*d* electrons of *T* and 4*d* electrons of Y for YFe_2 (a), YCo_2 (b), and YNi_2 (c) in the paramagnetic state [13].

itinerant electron ferromagnet with T_C varying from 280 to 301 K and M_S from 1.35 to 1.45 $\mu_B/f.u$.

The calculated electronic structure of Y₄Co₃ is shown in Fig. 4 [15]. This compound has a Ho₄Co₃-type hexagonal crystal structure with three inequivalent Co sites (6h), (2d), (2b) and two inequivalent Y sites. As the unit cell includes three formula units, the Co(2b) sites are half-filled (50%) and the number of atoms in the unit cell is equal to 21. Thus, in this crystallographic model, Y₄Co₃ cannot be regarded as an ordered compound, but as a disordered alloy with (2b) sites occupied randomly by cobalt atoms and vacancies. The ferromagnetic state obtained from spin-polarized computations is attributed to the Co atoms located on the (2b) sites, being the only magnetic atoms among 21 ones in the unit cell, and forming a quasi-one dimensional magnetic chains. As seen, in this case the Fermi level is located on an expressed minimum of DOS. Thus application of either external or internal molecular magnetic field shall result in an increase of the total DOS and a stronger polarization of the *d*-band with corresponding increase of the magnetic moment per Co.

In the above *R*-Co series, substitutions of non-magnetic Y by magnetic RE induces a substantial increase of μ_{Co} . Within the scope of the itinerant model, this effect is ascribed to the *f*-*d* exchange interaction. The total molecular field acting on the *d* subsystem reads [1,3].

$$B_{mol}^{(Co)} = \lambda_{dd} M_d + \lambda_{Rd} M_R, \tag{1}$$

where $\lambda_{dd} = z_d I_{dd}/2\mu_B^2$ and $\lambda_{dd} = (g_R - 1)z_R I_{Rd}/2g_R \mu_B^2$ are the corresponding molecular field coefficients, I_{dd} and I_{Rd} denote the d-d and R-d exchange integrals, z_d and z_R are the numbers of T and R atoms in the nearest-neighbour surrounding to a T atom. In the presence of external magnetic field, the total effective field acting on the Co sublattice can be represented as

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