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Magnetic properties of Al–Gd–Ni orthorhombic compounds

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Abstract

The magnetic properties of the Al_4GdNi, Al_2GdNi and $AlGd_2Ni_2$ compounds have been investigated using magnetic measurements in the temperature range 4–800 K and magnetic fields up to 9 T and X-ray photoelectron spectroscopy at room temperature. The compounds Al_4GdNi and $AlGd_2Ni_2$ order antiferromagnetically at T_N = 20 and 25 K, respectively, and Al_2GdNi orders ferromagnetically at T_C = 39 K. The results are discussed in terms of the Ruderman–Kittel–Kasuya–Yosida theory, the polarization of 5d electrons by the local exchange interaction 4f–5d and the spin fluctuations on Ni sites.

Keywords: Rare-earth transition metal compounds; Magnetization; XPS spectra; Spin fluctuations

1. Introduction

In the ternary metallic system Al–Gd–Ni, 14 intermetallic compounds with different crystallographic structures are reported [1]. The nearest-neighborhood of Gd and Ni ions, as well as, the distances Ni–Ni, Gd–Gd, Gd–Ni are different in each compound. This leads to different states of Ni atoms and also influences the Gd–Ni interaction in these compounds. The electronic 3d bandwidth of the transition metals (T) is determined by the overlap between the d orbitals of adjacent atoms and depends on the number of nearest-neighbors and the hopping integral J_h, which is very sensitive to the T–T distances [2]. On the other hand, the hybridizations between the 3d Ni and 5d6s Gd states and 3d Ni and 3sp Al states may lead to a partial or complete filling of the Ni 3d band. In many rare-earth-nickel compounds, nickel atoms do not carry a magnetic moment because of charge transfer of rare-earth conduction electrons to the 3d band [3]. It is expected in such a system to observe at low temperatures very different magnetic structures, such as ferromagnetism, ferrimagnetism or antiferromagnetism. Earlier experimental data on $Al_kGd_mNi_n$ compounds revealed three behaviors: (a) the Ni 3d band is partially filled. Ni atoms carry a magnetic moments like in the metallic Ni. The interactions Gd–Ni are of antiferromagnetic type and the compounds are ferrimagnetic at lower temperatures ($Al_3Gd_2Ni_5$, $Al_9Gd_2Ni_8$) [4]; (b) the Ni 3d band is completely filled. The magnetic properties of these compounds are governed by an indirect exchange coupling (RKKY-type) between the Gd moments, responsible for the ferromagnetic ($AlGd_3Ni_8$, $Al_8Gd_5Ni_2$) [5–7] and antiferromagnetic order (Al_3GdNi_2, $Al_{16}GdNi_3$) [4,8] at low temperatures and (c) the Ni 3d band is almost filled. The density of states at the Fermi level is drastically reduced in comparison with metallic nickel. The interactions between the Gd magnetic moments are of RKKY-type, but temperature-induced magnetic moments at Ni sites were also evidenced ($AlGdNi_4$) [9].

The aim of this paper is to study the magnetic properties of the new compounds Al_4GdNi, Al_2GdNi and $AlGd_2Ni_2$ to elucidate the magnetic state of Ni ions and to evaluate the effective s–f exchange integrals Γ in these compounds. The three orthorhombic compounds Al_4GdNi, Al_2GdNi and $AlGd_2Ni_2$ crystallize in the Al_4NiY, BRe_3 and B_2CoW_2

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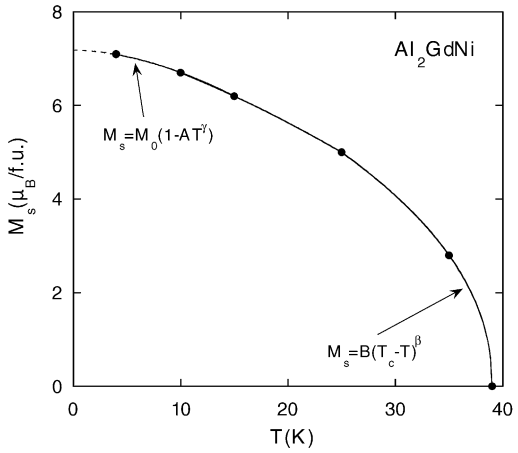

Fig. 1. Thermal variation of the spontaneous magnetization of Al_2GdNi.

structure types, respectively (Pearson symbols oC24, oC16 and, respectively, oI10 and space groups *Cmcm*, *Cmcm* and, respectively, *Immm*) [1].

2. Experimental

The investigated compounds Al_4GdNi, Al_2GdNi and $AlGd_2Ni_2$ were prepared by the standard arc-melting technique using a water-cold crucible in an argon atmosphere. The samples were melted repeatedly in the same atmosphere to ensure homogeneity. The weight loss of the final materials was found to be less than 1%. The purity of the starting materials was 99.9% for Gd and Ni and 99.999% for Al. X-ray powder diffraction measurement showed that the compounds are simple phases with the expected structure type. The lattice parameters agree with those reported in Ref. [1].

The magnetic measurements were performed with a vibrating magnetometer in the temperature range 4–300 K and magnetic fields up to 9 T. The XPS spectra were recorded using a PHI 5600ci ESCA spectrometer with monochromatized Al Kα radiation at room temperature. The pressure in the ultrahigh vacuum chamber was in the 10^{-10} mbar range during the measurements. Because of the high chemical reactivity of the rare-earth compounds the samples were broken in situ. Sample cleanliness was checked by monitoring the oxygen and carbon 1s levels.

3. Results and discussion

The temperature dependence of the spontaneous magnetization of Al_2GdNi is shown in Fig. 1. The values and variations of magnetization with field and temperature suggest that this compound exhibits a collinear ferromagnetic arrangement of Gd moments below the Curie temperature $T_C = 39$ K. At low temperatures, $T < 15$ K, a spin-wave-like dependence of the type $M_S(T) = M_S(0)(1 - AT^{\gamma})$ is observed with a spontaneous magnetization $M_S(0) = 7.19\ \mu_B$/f.u. and an exponent

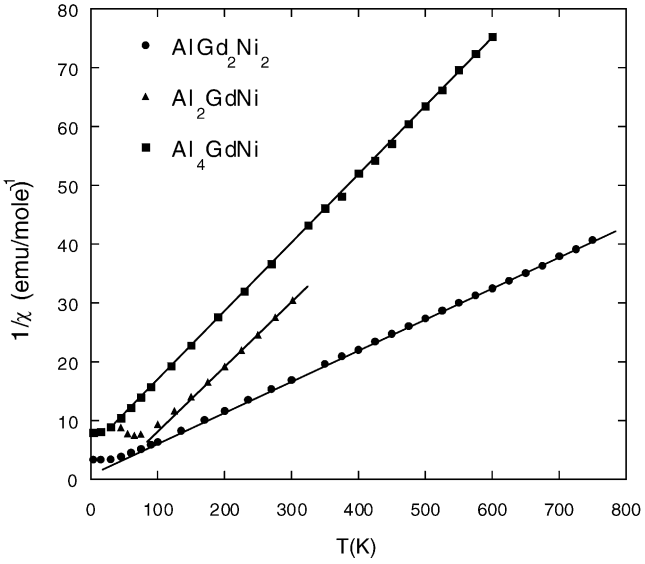

Fig. 2. Thermal variation of the reciprocal susceptibilities of Al_4GdNi, Al_2GdNi and $AlGd_2Ni_2$ compounds.

$\gamma = 1.8$, which is indicative of an essentially two-dimensional spin wave spectrum similar to that reported for pure Gd [10]. The spontaneous magnetization at 0 K is slightly higher than the theoretical value for the Gd^{3+} ion (7 μ_B). For the thermal variation of $M_S(T)$ when approaching the Curie temperature, the fit of the $M_S(T) = B(T_C - T)^{\beta}$ leads to the exponent $\beta = 0.46$, a value which is close to 1/2. This may be indicative of a mean field-like behavior in this temperature range.

The temperature dependence of the reciprocal susceptibilities of Al_4GdNi, Al_2GdNi and $AlGd_2Ni_2$ compounds are shown in Fig. 2. The $1/\chi(T)$ curves for Al_4GdNi and $AlGd_2Ni_2$ present an upturn to a constant value at 20 and 25 K, respectively, temperatures which were assigned as transition temperatures (T_N) from paramagnetic to antiferromagnetic state. The lack of a cusp in the $\chi(T)$ dependence for these two compounds is due to a very small concentration of the magnetic impurities, which at very low temperatures have a contribution to the measured susceptibility. The temperature dependence of the reciprocal susceptibilities for all investigated compounds is linear above the transition temperatures (Fig. 2). The magnetic susceptibilities obey the Curie–Weiss law, $\chi = C/(T - \theta_p)$, with the paramagnetic Curie temperature θ_p and the effective magnetic moments μ_{eff} given in Table 1. The effective magnetic moments in all three compounds are higher than the theoretical moment of 7.94 μ_B for the Gd^{3+} ion. Intrinsic effects such as a contribution from Ni atoms or from conduction electrons lead to slightly larger magnetic moments.

In order to distinguish between these two effects, we analyze both the valence band and Ni 2p core level spectra. The valence bands of Al_4GdNi, Al_2GdNi and Ni metal are shown in Fig. 3. One can observe marked differences both in the shape and in the position of the valence band for the three materials. The maximum of the valence band is shifted to higher