

Dedicated to Prof. Dorin N. Poenaru's
70th Anniversary

CRYSTAL STRUCTURES, MAGNETIC PROPERTIES AND ELECTRONIC STRUCTURES OF RARE-EARTH-NICKEL INTERMETALLIC COMPOUNDS

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Abstract. The $\text{RNi}_{5-x}\text{Cu}_x$ with $x \leq 2$, $\text{RNi}_{5-x}\text{Al}_x$ with $x \leq 1.5$ and $\text{Gd}_x\text{La}_{1-x}\text{Ni}_5$ compounds, where R is a rare-earth, crystallize in a CaCu_5 -type structure. A $\text{HoNi}_{2.6}\text{Ga}_{2.4}$ superstructure was shown for $\text{RNi}_{5-x}\text{Al}_x$ with $x \geq 2$. Magnetic measurements on the above systems, as well as on RNi_4B compounds were performed in the temperature range 1.7–300 K and fields up to 9 T. Band structure calculations were also performed. The 4f-5d-3d exchange interactions were analysed and the contributions of 4f-5d and 5d-3d exchange interactions on R5d band polarization were estimated. As function of composition, nickel shows either an exchange enhanced paramagnetism or is a weak ferromagnet. An internal field of the order of 40 T is necessary to induce a nickel moment, at 0 K. The magnetic behaviour of nickel was analysed in models which take into account electron correlation effects in d bands.

Key words: crystal structure, electronic structure, magnetic properties, rare-earth-nickel compounds.

1. INTRODUCTION

The RNi_5 compounds, where R is a rare-earth or yttrium, crystallize in a hexagonal structure of CaCu_5 -type, having P6/mmm space group. In this structure the R atoms occupy 1a-type site, while nickel ones are distributed on 2c and 3g positions [1]. The RNi_4B compounds crystallize in a hexagonal structure of CeCo_4B -type [2]. The structure is obtained from that of CaCu_5 one by substituting every two layers Ni(2c) atoms by B. In the CeCo_4B -type lattice, the R atoms occupy two different crystallographic positions: (1a) and

(1b), the Ni atoms are located on (2c) and (6i) sites and the boron atoms occupy the (2d) positions.

The analysis of the magnetic properties of RNi_5 and RNi_4B compounds evidenced interesting properties. The Curie temperatures, T_c , are very low, the maximum values being reported for GdNi_5 and GdNi_4B . Generally, was suggested that nickel, at low temperatures, is not magnetic. Latter on, analyzing the magnetic properties of $(\text{Gd}_x\text{A}_{1-x})\text{Ni}_5$ with $\text{A}=\text{Y}$ or La [3, 4], has been shown that in GdNi_5 , at 1.7 K, the nickel moment is $M_{\text{Ni}} \simeq 0.17\mu_B/\text{atom}$, antiparallel oriented to gadolinium one. The nickel saturation moments are nil in LaNi_5 and YNi_5 compounds.

The $\text{RNi}_{5-x}\text{Cu}_x$ with $\text{R}=\text{La}$ [5] and Nd [6], for $x \leq 2$ crystallize in a CaCu_5 type structure. In case of $\text{RNi}_{5-x}\text{Al}_x$ with $\text{R}=\text{Nd}$, Gd [7], $\text{R}=\text{Dy}$ [8, 9] and $\text{R}=\text{La}$ [10] systems, for $x \geq 2$, the structure changes from CaCu_5 -type to $\text{HoNi}_{2.6}\text{Ga}_{2.4}$ -type, having also $\text{P6}/\text{mmm}$ space group. The RNi_4B compounds were reported to be ferromagnetically ordered [11, 12, 13, 14], the nickel moment being nil. Recently [15] we showed that nickel, in the above compounds, shows a spin fluctuation type behaviour [15].

In this paper we analyse comparatively, the crystal structures and magnetic properties of $\text{RNi}_{5-x}\text{M}_x$ systems with $\text{M}=\text{Cu}$ and Al , $\text{Gd}_x\text{La}_{1-x}\text{Ni}_5$ as well as RNi_4B systems. In addition, band structure calculations were performed. The magnetic behaviour of nickel in the above systems will be particularly analysed.

2. EXPERIMENTAL

The samples were prepared in induction or arc furnaces, in purified argon atmosphere. The samples were thermally treated in vacuum at temperatures between 900 and 1000°C during 5 up to 6 days. The X-ray analyses show the presence of only one phase. The composition dependences of the lattice parameters for $\text{DyNi}_{5-x}\text{Al}_x$ and $\text{NdNi}_{5-x}\text{Cu}_x$ systems are plotted in Fig. 1. The presence of CaCu_5 -type structure was shown in $\text{RNi}_{5-x}\text{Cu}_x$ compounds with $x \leq 2$ and in $\text{RNi}_{5-x}\text{Al}_x$ ones in composition range $x \leq 1.5$. In aluminium doped compounds, a $\text{HoNi}_{2.6}\text{Ga}_{2.4}$ -type structure was shown for $2 \leq x \leq 3$. This structure is also hexagonal having $\text{P6}/\text{mmm}$ space group. The unit cell parameters of the two types of structures are related by $a_{\text{HoNi}_{2.6}\text{Ga}_{2.4}} = \sqrt{3}a_{\text{CaCu}_5}$ and $a_{\text{HoNi}_{2.6}\text{Ga}_{2.4}} = c_{\text{CaCu}_5}$ [7]. In CaCu_5 -type structure, the aluminium substitution takes place at the 3g site, situated in the $z = 1/2$ plane, which does not contains R atoms. These sites allow greater Ni-Al distances. The total filling of the 3g site by Al is not possible since Ni(2c)-Ni(3g), Al(3g)-Al(3g) or Ni(3g)-Al(3g) distances are smaller than the

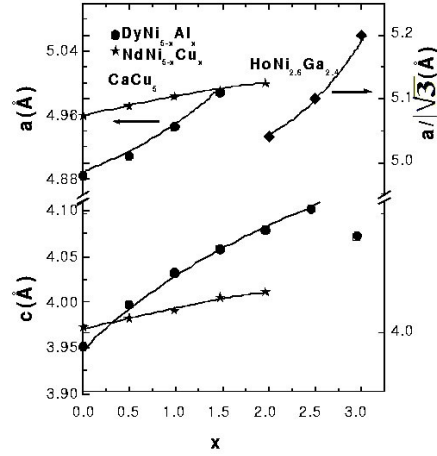


Fig. 1 – Composition dependences of the lattice parameters for $\text{NdNi}_{5-x}\text{Cu}_x$ and $\text{DyNi}_{5-x}\text{Al}_x$ compounds.

sum of metallic radii ($r_{\text{Ni}} + r_{\text{Al}}$) or ($r_{\text{Al}} + r_{\text{Al}}$). The occurrence of $\text{HoNi}_{2.6}\text{Ga}_{2.4}$ superstructure induces an increase of the distances $\text{Al}(3g)\text{-Ni}(6k)$ and $\text{Al}(3f)\text{-Al}(6k)$, making the possibility to locate aluminium in 3f and 6k sites. In this structure nickel occupies completely 6l sites and aluminium 3f sites. The 6k sites are statistically occupied by Ni and Al.

Magnetic measurements were performed in the temperature range 1.7-300 K and fields up to 9 T. The saturation magnetizations, M_s , were determined from magnetization isotherms, according to approach to saturation law, $M - M_s(1 - b/H)$, by extrapolating the measured values to $H^{-1} \rightarrow 0$. By b we denoted the coefficient of magnetic hardness. The paramagnetic contributions of conduction electrons were neglected. The errors introduced by the above assumption was of the order of $0.1\mu_B/\text{f.u.}$ In the paramagnetic range, the susceptibilities, χ , were determined from their field dependencies, according to the relation $\chi = \chi_0 + dM_0H^{-1}$, by extrapolating the measured values to $H^{-1} \rightarrow 0$. By d is denoted a presumed magnetic ordered impurity content and M_s is their saturation magnetization. By this method any possible alteration of magnetic susceptibilities, as result of the presence of small quantities of magnetic ordered phase, is avoided. For all compositions, if exist, the estimated content of magnetic ordered phase, at $T > T_c$, is smaller than 0.1%.

3. COMPUTING METHOD

Band structure calculations for $\text{LaNi}_{5-x}\text{M}_x$ with $\text{M}=\text{Cu},\text{Al}$, $\text{DyNi}_{5-x}\text{Al}_x$, $\text{Gd}_x\text{La}_{1-x}\text{Ni}_5$ and RNi_4B were carried out by using the ab initio tight binding

linear muffin-tin orbital method in the atomic sphere approximation (TB-LMTO-ASA). The procedure of calculation was described elsewhere [16, 17, 18]. In the framework of the local density approximation (LDA), the total electronic potential is the sum of external, Coulomb and exchange-correlation potentials [19]. The functional form of the exchange-correlation energy used in the present work was the free-electron gas parameterization of von Barth and Hedin [20]. Relativistic corrections are included. For $\text{NdNi}_{5-x}\text{Cu}_x$ and $\text{TbNi}_{5-x}\text{Al}_x$ systems, band structure calculations were performed by using TB-LMTO method, within LDA+U approach [17, 18]. The LDA+U scheme is based on the Anderson impurity model in mean field (Hartree-Fock) approximation, that analyses the s and p electrons as noncorrelated, described by an orbital independent potential and d and f electrons are described by an orbital dependent potential [23]. When applied to transition metals and rare-earth compounds, the LDA+U method gives a quantitative improvement compared with LDA, for ground state properties, such as magnetic moments and interchain exchange parameters [23]. We note that this approach describes well the parallel coupling of Nd and Ni moments in NdNi_5 . In case of $\text{RNi}_{5-x}\text{Cu}_x$ system, the Cu atoms for $x = 1$, were supposed to occupy the 3g sites while for higher Cu concentrations both 2c and 3g positions. In case of $\text{RNi}_{5-x}\text{Al}_x$, for band structure calculations, the Al was introduced in 3g sites. For $\text{HoNi}_{2.6}\text{Ga}_{2.4}$ superstructure one Al was located in 3f sites and one or two aluminium atoms were distributed on 6k sites.

4. BAND STRUCTURES

The total density of states as well as the Ni(3d) projected DOS for TbNi_5 , DyNi_5 , NdNi_5 and DyNi_4Al compounds are plotted in Fig. 2. The determined nickel magnetic moments, at 0 K, are given in Fig. 3 for Ni in 2c and 3g sites in CaCu_5 -type structure as well as for 4f, 12l, 6g and 12k sites in rhombohedral R_2Ni_{17} compounds with heavy rare-earths. The magnetic moments of nickel are antiparallely oriented to those of heavy rare-earths. Values $M_{\text{Ni}}(3\text{g})$ are higher than those determined in 2c sites. The 2c sites in RNi_5 compounds have 6Ni(3g) and 3Ni(2c) atoms as well as 3R ones, while 3g sites have 4Ni(2c), 4Ni(3g) and 4R as nearest neighbours. The strength of the exchange interactions between nickel and magnetic R atoms is more strong than between nickel ones, the Ni moments being essentially induced by R atoms. Thus, the exchange splitting of Ni3d(3g) band is greater than for Ni3d(2c) one, since of the higher number of R atoms as nearest neighbours. As example, in NdNi_5 , the exchange splitting of Ni3d(3g) band, of 0.01 eV, is greater than that in Ni3d(2c) of 0.0066 eV. Similar behaviour can be evidenced in

R_2Ni_{17} compounds, where nickel magnetic moments are also dependent on their environments, intensity of exchange interactions, respectively.

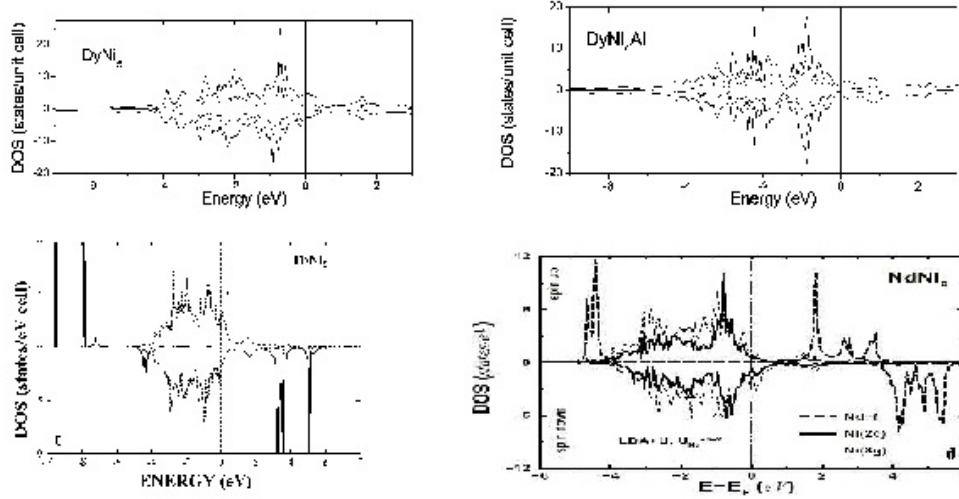


Fig. 2 – Projected densities of states for $DyNi_5$, $DyNi_4Al$, $TbNi_5$ and $NdNi_5$ compounds.

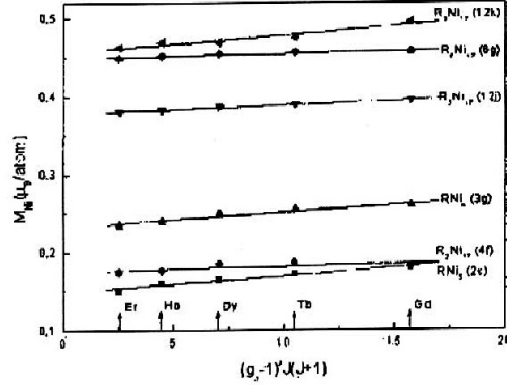


Fig. 3 – The nickel moments as function of De Gennes factor for RNi_5 and R_2Ni_{17} (rhombohedral) compounds, where R is a heavy rare-earth.

When replacing nickel by aluminium, the nickel moments, at 0 K, decrease rapidly. As example, in $DyNi_4Al$, values of 0.03 and $0.04\mu_B$ were determined at $2c$ and $3g$ sites, respectively. In $TbNi_4Al$ the nickel magnetic moments at $2c$ and $3g$ sites were 0.02 and $0.03\mu_B$, respectively. Smaller decrease of the nickel moments can be seen in copper doped compounds. Thus

in NdNi_4Cu , values of $\simeq 0.06\mu_B$ were determined both at 2c and 3g sites. The above data suggest a strong hybridization of Ni3d and Al3p bands in aluminium substituted compounds. In case of copper doped systems a rather independent Cu d-band is formed at 3.3 eV binding energy, as evidenced by XPS studies [6] and the hybridization effects are smaller. As a result of substitutions, in case of nickel heavy rare-earth compounds, there is a transition from a ferrimagnetic types ordering to a ferromagnetic one as the Al or Cu content increases.

The R5d band polarizations in RM_5 and R_2M_{17} where $\text{M}=\text{Ni}$ or Co and R is a heavy rare-earth metal, as function of De Gennes factor $G = (g_j - 1)^2 J(J+1)$ are plotted in Fig. 4. There are linear dependences described by the relation:

$$M_{5d} = M_{5d}(0) + \alpha G. \quad (1)$$

The slopes α are $\alpha = 1.4 \cdot 10^{-2}\mu_B$ for RM_5 compounds ($\text{M}=\text{Ni}, \text{Co}$) and $1 \cdot 10^{-2}\mu_B$ for R_2Ni_{17} compounds.

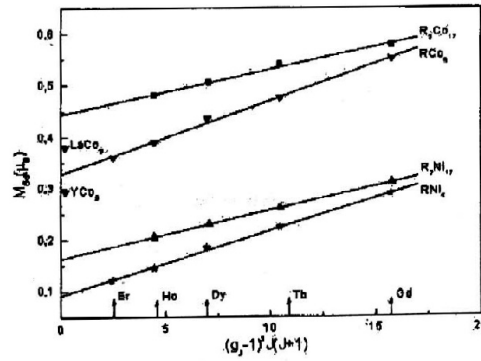


Fig. 4 – The R5d band polarizations in RNi_5 and R_2Ni_{17} heavy rare-earth compounds.

In Fig. 5 we plotted the total density of states for RNi_4B with $\text{R}=\text{Gd}, \text{Dy}$ and in Fig. 6 the dependences of nickel moments as well as of 5d band polarizations on De Gennes factor. Similar behaviour was shown as for RNi_5 and R_2Ni_{17} compounds. A value $\alpha = 4 \cdot 10^{-3}\mu_B$ was also determined. The magnetic moments of nickel atoms are nearly three times smaller than those determined in RNi_5 compounds. This fact can be correlated with the presence of boron atoms. The exchange interactions are decreased and consequently the exchange splittings of Ni3d bands are diminished.

When increasing the magnetic contributions of transition metals, for a similar rare-earth compound, the R5d band polarization is translated to higher values. The composition dependences of the R5d band polarization, described by the relation (1) suggest the presence of two contributions. The first one,

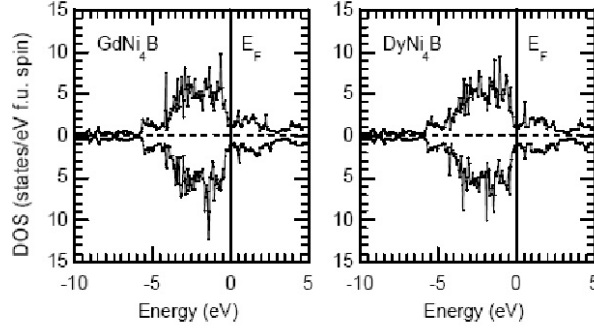


Fig. 5 – Total DOS for RNi_4B compounds with $R=Gd, Dy$.

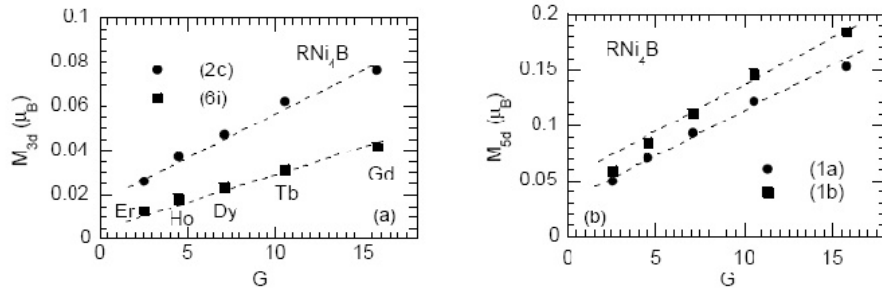


Fig. 6 – The dependences of Ni magnetic moments and of R5d band polarizations in RNi_4B as function of DeGennes factor.

αG , is related to local 4f-5d exchange interactions and seems to be essentially determined by the rare-earth moment. The second one, $M_{5d}(0)$ can be attributed to the induced polarization by short range 5d-3d and 5d-5d exchange interactions. The $M_{5d}(0)$ may be identified by extrapolation of M_{5d} vs G variation, to $G=0$ or by induced polarization in compounds with nonmagnetic R metals. The short range exchange interactions may be described by the hamiltonian:

$$H = -2 \sum_i J_{3d_i-5d} S_{5d} \sum_{n_i} S_{3d_i n_i} - 2 J_{5d-5d} S_{5d} \sum_j S_{5d_j}. \quad (2)$$

We denoted by i the number of 3d atoms situated in the first coordination shell to an R atom, n_i is the number of atoms occupying a given i site and j is the number of R nearest neighbor atoms to a given R site.

By using the molecular field approximation and supposing that the molecular field coefficients, characterizing the exchange interactions are $N_{5d-3d} \simeq N_{5d-5d}$, it result that $M_{5d}(0) \propto \sum n_k M_k$ where n_k is the number of R and M atoms situated in first coordination shell, having M_k magnetic moments.

As seen in Fig. 7, there is a linear relation described by $M_{5d}(0) = \gamma \sum n_k M_k$ where $\gamma = 0.04$.

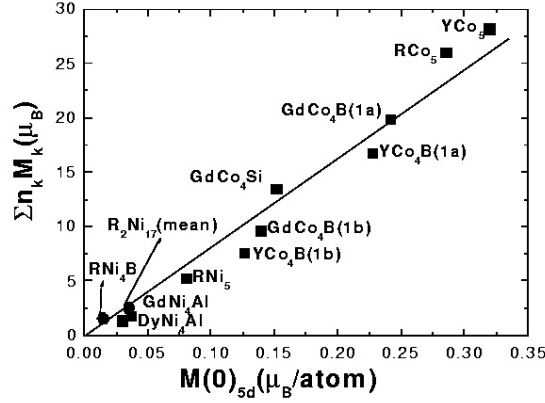


Fig. 7 – The $M_{5d}(0)$ contributions to R5d band polarization due to short range exchange interactions as a function $\sum n_k M_k$.

5. MAGNETIC DATA

The thermal variations of spontaneous magnetizations for $(\text{Gd}_x\text{La}_{1-x})\text{Ni}_5$ compounds are plotted in Fig. 8.

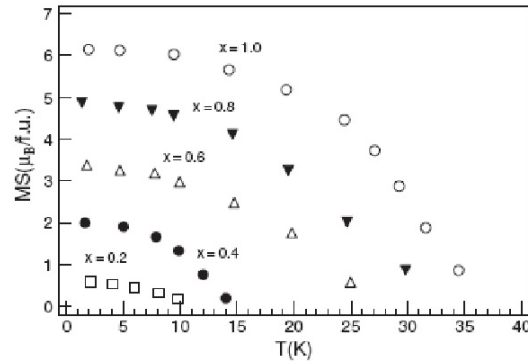


Fig. 8 – Thermal variations of spontaneous magnetizations for $\text{Gd}_x\text{La}_{1-x}\text{Ni}_5$ system.

The magnetizations, at 1.7 K, decrease nearly linearly when increasing La content – Fig. 9. On the same figure, the magnetizations obtained from band structure calculations are also given [4]. There is a good agreement between

experimentally determined and computed values per formula unit. The differences are smaller than $0.3\mu_B$. The Curie temperatures decrease gradually from $T_C = 35$ K and finally, the LaNi_5 is a paramagnet. Similar results were obtained for other systems. Thus, in case of $\text{DyNi}_{5-x}\text{Al}_x$ the Curie temperatures decrease from 13 K ($x = 0$) to 8 K ($x = 1$) and 5 K ($x = 0$). The Curie temperature of NdNi_5 is $T_C = 9$ K and diminishes to 6 K in NdNi_4Cu . These data show that the exchange interactions decrease both as result substitution to R or Ni sites by nonmagnetic elements.

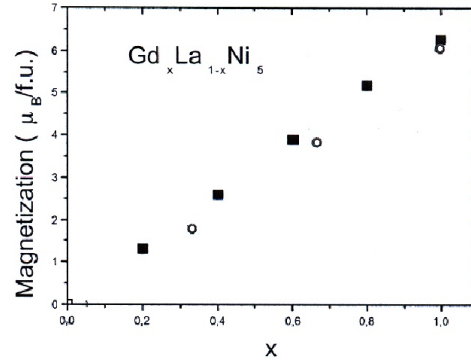


Fig. 9 – Composition dependences of saturation magnetizations, at 1.7 K, in $(\text{Gd}_x\text{La}_{1-x})\text{Ni}_5$ compounds. The data obtained from band structure calculations (\circ) are also plotted.

The thermal variations of reciprocal susceptibilities for some representative compounds with magnetic rare-earths are shown in Fig. 10a, and in Fig. 10b with nonmagnetic ones, as $\text{LaNi}_{5-x}\text{Cu}_x$.

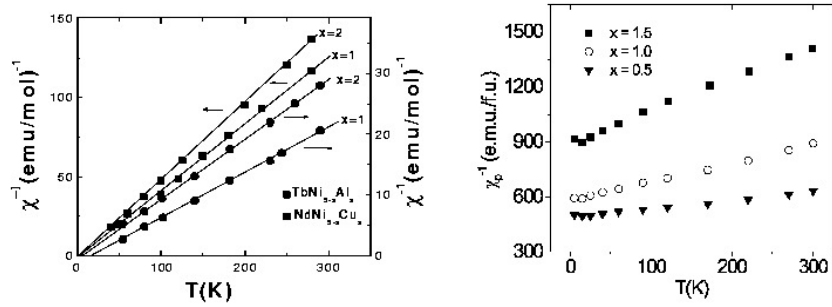


Fig. 10 – Thermal variations of reciprocal susceptibilities for:
a) $\text{TbNi}_{5-x}\text{Al}_x$ and $\text{NdNi}_{5-x}\text{Cu}_x$ compounds with $x = 1$ and 2;
b) $\text{LaNi}_{5-x}\text{Cu}_x$ systems.

In all cases, at high temperatures, Curie-Weiss type behavior was evidenced. The effective nickel moments determined from Curie constants are plotted in Fig. 11. The effective nickel moments in doped aluminum systems decrease more rapidly than in case of copper ones, suggesting a strong hybridization of Ni3d and Al3p bands, as evidenced already by band structure calculations.

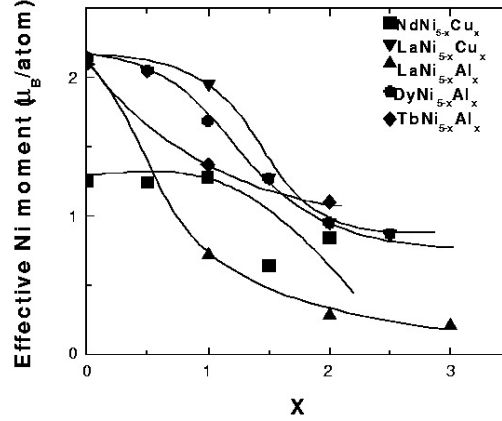


Fig. 11 – Composition dependences of the effective nickel moments.

In LaNi₅ or YNi₅-based compounds, the magnetic susceptibilities increase up to a characteristic temperature, T_{max} . The T_{max} values are shifted to lower temperatures when increasing copper or aluminum content. Above a characteristic temperature, T^* , the reciprocal susceptibilities follow a Curie-Weiss type dependence. The effective nickel moments follow the same trend, as function of composition, as in case of magnetic rare-earth compounds. In the range $T \leq 10$ K, the magnetic susceptibilities of LaNi_{5-x}M_x (M=Al,Cu) follow a dependence described by the relation

$$\chi = \chi_0(1 + aT^2). \quad (3)$$

The coefficient a was computed, by using band structure calculations. Considering the paramagnon model [24] we have

$$a = \frac{\pi^2}{6} \left[\left(2 \frac{\eta''(E_F)}{\eta(E_F)} \right) - 12 \left(\frac{\eta'(E_F)}{\eta(E_F)} \right)^2 \right] s^2. \quad (4)$$

We denoted by s , the Stoner enhancement factor and $\eta(E_F)$, $\eta'(E_F)$ and $\eta''(E_F)$ are the density of states at the Fermi level and their first and second derivatives, respectively. The computed a values, according to relation (4), agree reasonably with those experimentally determined. The Curie temperatures of RNi₄B vary linearly with De Gennes factor. The effective nickel

moments are nearly constants and around $1.8\mu_B/\text{atom}$, close to those determined in RNi_5 based compounds.

6. CONCLUSIONS

The results of magnetic measurements and band structure calculations suggest that the exchange interactions in R-Ni based compounds, where R is a magnetic rare-earth, can be described by the Campbell model [25]. The magnetic coupling between rare-earths and transition metals takes place through 4f-5d-3d exchange interactions. The R4f electrons polarize the R5d band through local exchange interactions. The induced R5d band polarizations in heavy rare-earth compounds is parallel to the 4f moments. There are also short range exchange interactions of 5d-3d or 5d-5d types which induce additional 5d band polarizations. The band structure calculations show that Ni moments, at 0 K, decrease very fast in $\text{RNi}_{5-x}\text{M}_x$ ($\text{M}=\text{Al},\text{Cu}$) systems in the composition range $0 \leq x \leq 1$ and are near nil for higher Al or Cu substitution. In $\text{Gd}_x\text{La}_{1-x}\text{Ni}_5$ the Ni moments at 2c site decrease from $0.2\mu_B$ ($x = 0$) to $0.08\mu_B$ ($x = 0.67$) while the Ni moments of atoms situated in 3g sites vary from $0.255\mu_B$ ($x = 0$) to $0.12\mu_B$ ($x = 0.67$). There is a linear correlation between Ni moments at 2c and 3g sites and exchange splitting of their 3d bands – Fig. 12. There seems to be a critical value of the exchange splitting for which Ni moment will appear, of the order of $2.5 \cdot 10^{-2}$ eV. This corresponds to an exchange field of $\simeq 0.4$ MOe. A value of critical exchange field of $\simeq 0.3$ MOe has been suggested from magnetic measurements, close to the above one [4].

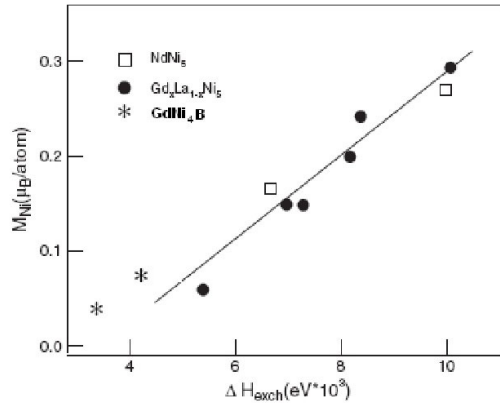


Fig. 12 – The correlation between Ni magnetic moments at 2c and 3g sites in RNi_5 and 2c and 6i sites and in GdNi_4B at 2c and 6i sites and the exchange splitting of their 3d bands.

The magnetic susceptibilities of $\text{LaNi}_{5-x}\text{M}_x$ compounds show, at $T \leq 10$ K, a T^2 dependence. Above a characteristic temperature, T^* , a Curie-Weiss behaviour was evidenced. As already mentioned, the nickel at 0 K, has a weak moment or shows an enhanced magnetic susceptibility. In the high temperature range, the linear χ^{-1} vs. T dependences suggest the presence of localized nickel moments. The above behaviour may be analysed in models which take into account the electron correlations effects in d-band, as spin fluctuation model [26] or dynamical mean field theory [27]. The above models reconcile the dual character of electron, which as a particle, requires a real space description and as a wave, a momentum space description. The spin fluctuation model considers the balance between the frequencies of longitudinal spin fluctuations which are determined by their lifetime and transverse fluctuations which are of thermal origin. These effects lead to the concept of temperature induced moment. For a weak ferromagnet as Ni in $\text{RNi}_{5-x}\text{M}_x$ ($x \leq 1$), and RNi_4B or an exchange enhanced paramagnetism as for compositions $x \geq 2$, or in $\text{LaNi}_{5-x}\text{M}_x$ (M=Cu,Al) systems, the wave number depend susceptibility, χ_q , has a large enhancement due to electron-electron interaction for small q -values. The χ_q shows a significant temperature dependence only for q values close to zero. The average amplitude of spin fluctuations $\langle S_{loc}^2 \rangle = 3k_B T \sum \chi_q$ increases with temperature and reaches an upper limit, at a temperature T^* . For $T > T^*$, a Curie-Weiss behaviour is predicted, as in systems having local moments. The moments are localized in q -space. If the spin fluctuations are saturated, feature which characterize the systems having high exchange enhancement factors ($s \simeq 8 - 10$), the effective moments are given by the electronic configuration of 3d ion. The effective nickel moments in RNi_5 ($\simeq 2.15\mu_B$) or in RNi_4B ($\simeq 1.80\mu_B$) are somewhat lower than the value expected for Ni^{2+} ion ($2.82\mu_B$). This decrease may be attributed to the diminution of electron correlations due hybridization and/or magnetic dilution effects.

The magnetic behaviour of nickel in RNi_5 -based systems or RNi_4B ones may be analysed also in the dynamical mean field theory DMFT [27] combined with the standard LDA band calculations (LDA + DMFT) [28]. In a strongly correlated system, leading Curie-Weiss behaviour, at high temperatures, is predicted. For an itinerant electron system, the time dependence of the correlation function results in a temperature dependence of $\langle S_{loc}^2 \rangle$. Fluctuating moments and atomic like configurations are large at short time. The moments are reduced at larger time scales, corresponding to a more band like less correlated electronic structure near Fermi level.

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