Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

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Version of record first published: 18 Oct 2010

To cite this article: L. Chioncel, E. Burzo, R. Tetean* & V. Pop (2004): Electronic Structure Of Y(CoXni1-X)2 Compounds, Molecular Crystals and Liquid Crystals, 417:1, 29-37

To link to this article: http://dx.doi.org/10.1080/15421400490478641

Mol. Cryst. Liq. Cryst., Vol. 417, pp. 29/[513]–37/[521], 2004

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400490478641

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ELECTRONIC STRUCTURE OF $Y(Co_xNi_{1-x})_2$ COMPOUNDS

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The temperature dependences of the magnetic susceptibilities for $Y(Co_x Ni_{1-x})_2$ compounds were analyzed starting from the computed density of states by LMTO-TBA method and taking into account the effect of spin fluctuations in a classical Gaussian statistics. The mean square amplitudes of spin fluctuations saturate at decreasing temperatures when increasing the nickel content is in agreement with experimental data.

Keywords: intermetallic compounds; spin fluctuations; susceptibility

1. INTRODUCTION

A large number of experimental studies were performed in order to analyze the magnetic behaviour of YCo_2 compound [1]. Since the measurements were made in a limited temperature range and no corrections for the possible presence of ferromagnetic impurities were considered, the experimental data were difficult to be analyzed. By analyzing the field dependences of the magnetic susceptibilities of YCo_2 [2] it was possible to obtain reliable data, in a large temperature range by eliminating the possible contribution due to presence of a magnetic ordered phase. These data evidenced that at low temperatures ($T < 10\,K$) the magnetic susceptibilities,

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χ, of YCo_2 increase according to a T^2 law. The susceptibilities have a maximum located at $T \cong 240\,K$ and then decrease. Above $T^* \cong 450\,K$, the χ values follow a Curie-Weiss type behaviour. The effective cobalt moment, determined at $T > 450\,K$, is close to that of Co^{2+} ion considering only spin contribution. The magnetic behaviour of this compound was analyzed in spin fluctuations model. YNi_2 is a nearly Pauli paramagnet. The susceptibilities at 1.7 K are by one order of magnitude smaller than in case of YCo_2. The χ values decrease from $2.4 \cdot 10^{-4}$ to $1.8 \cdot 10^{-4}$ emu/f.u. in the temperature range 1.7 K to 40 K and then are nearly constant around $(1.8–1.9) \cdot 10^{-4}$ emu/f.u. for $T \leq 300\,K$.

Band structure calculations were also performed on both YCo_2 [4–7] and YNi_2 [4–6] compounds. The temperature dependences of the magnetic susceptibilities, computed from band structure, were also compared with some old experimental data [5]. In this paper we analyze comparatively the temperature dependences of the magnetic susceptibilities of $Y(Co_xNi_{1-x})_2$ compounds, experimentally determined in a large temperature range, with those computed from band structures by considering the effect of spin fluctuations into account.

2. EXPERIMENTAL AND COMPUTING METHOD

The $Y(Co_xNi_{1-x})$ samples were prepared in an induction furnace in purified argon atmosphere. The samples were thermally treated in vacuum at ~1000°C for one week. The X-ray analyzes show the presence of only one phase having a cubic $MgCu_2$-type structure. The composition dependence of the lattice parameters is well described by a Veguard-type law – Table 1.

Magnetic measurements were performed in the temperature range 1.7–800 K and external field up to 80 kOe. The magnetic susceptibilities, χ_m, were obtained from their field dependencies according to the relation $\chi_m = \chi + cM_sH^{-1}$, by extrapolating the measured values χ_m to $H^{-1} \to 0$

TABLE 1 Lattice Parameters, the Parameter $\lambda = \pi/aq_m$,, the Cut-Off Wave Vector q_m and the Exchange Stiffness Constants for $Y(Co_xNi_{1-x})_2$ Compounds

Composition	a (Å)	λ	$q_m = \pi/a\lambda$ (Å^{-1})	A (cm^5Oe^2/erg)10^{-12}
1.0	7.206	2.02	0.215	5.5
0.8	7.200	1.93	0.225	5.4
0.6	7.196	1.85	0.235	5.3
0.4	7.190	1.29	0.338	5.2
0.2	7.183	1.30	0.336	5.1

[8]. We denoted by c a presumed magnetic ordered impurity content and M_s is their saturation magnetization. By this method any possible alteration of the magnetic susceptibilities, as a result of the presence of small quantities of magnetic ordered phase, is avoided.

Band structure calculations were carried out by using the ab initio tight binding linear muffin tin orbital method (LMTO) in the atomic sphere approximation. The detailed procedure of calculation was described elsewhere [9–12]. In the framework of the local density approximation (LDA), the total electronic potential is the sum of the external, Coulomb and exchange correlation potential [12]. The frozen core approximation was employed in the generation of the potential and parametrization of the Von Barth and Hedin [13] was used for the exchange correlation part of the effective one electron potential, obtained within the local spin density approximation of the density functional theory. The calculations were performed on the ordered compounds. The integration involved a $48 \times 48 \times 48$ mesh in the Brillouin zone for the above structures. The k-space integration was performed by tetrahedron method using an algorithm to generate k-points as suggested by Jepsen and Andersen [14]. According to this algorithm, the Brillouin zone is taken as a parallelepiped spanned by the reciprocal vectors. This was first divided into tetrahedra of the optimal identical shape and then after applying the symmetry operations of the crystal, a limited set of unequivalent tetrahedra was retained and used in analytical integration. Relativistic corrections were included without the spin-orbit coupling.

3. EXPERIMENTAL AND COMPUTING DATA

The thermal variations of reciprocal susceptibilities are plotted in Figure 1. The characteristic feature for compounds with $x \geq 0.4$ is the increase of the magnetic susceptibilities up to a maximum, located a temperature T_{max}. The temperatures at which T_{max} are situated, decrease gradually when increasing the nickel content from $\sim 240\,K$ ($x = 1.0$) up to $\sim 20\,K$ ($x = 0.4$). For the compound with $x = 0.2$ the magnetic susceptibility seems to decrease in all the composition range. Above the characteristic temperature, T^*, which decrease when nickel content increase, the reciprocal susceptibilities follow a linear temperature dependence, characteristic for systems having localized magnetic moments. Assuming that nickel has a very small contribution to the magnetic susceptibility, for $T > T^*$, the effective cobalt moments were determined. These values are little dependent on the composition being situated between $3.72\,\mu_B$/atom ($x = 0.2$) and $3.87\,\mu_B$/atom ($x = 1.0$). In the low temperature range, there is a linear dependence of the magnetic susceptibility as function of T^2:

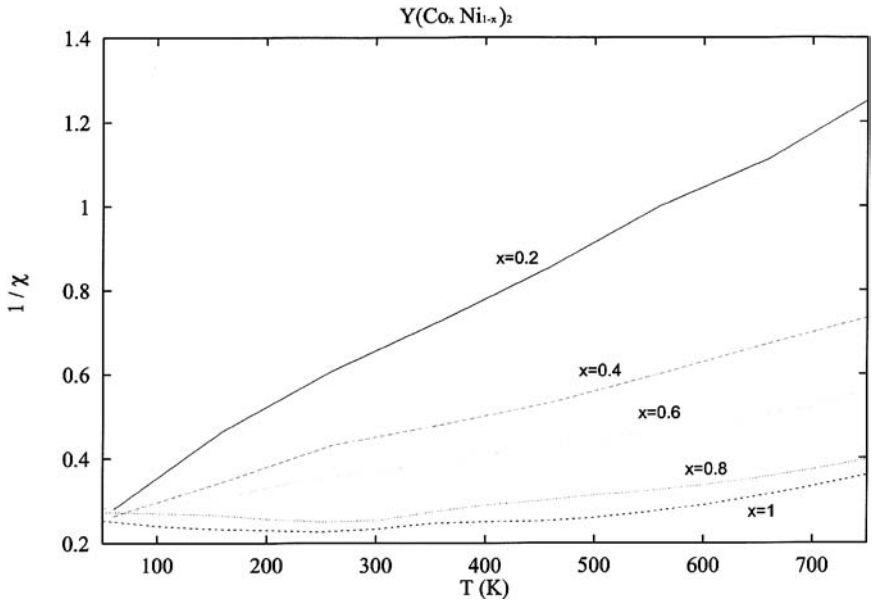

FIGURE 1 Thermal variations of reciprocal susceptibilities for $Y(Co_xNi_{1-x})_2$ compounds with $x \geq 0.2$. By solid lines are the fitted curves according to the relation (2).

$$\chi = \chi_o(1 + bT^2) \tag{1}$$

with $b = 1.8 \cdot 10^{-6}\ K^{-2}$ ($x = 1.0$) and $1.3 \cdot 10^{-6}\ K^{-2}$ ($x = 0.8$). Since of the shift of the maximum in susceptibilities to lower temperatures, when increasing nickel content, no reliable data were obtained for compounds with $x \leq 0.6$.

The band structures, for some representative compounds, are plotted in Figure 2. The Fermi level of YCo_2 lies just above a large sharp peak in DOS. The Fermi levels are shifted toward a region with lower density of states when cobalt is gradually replaced by nickel.

By using the calculated density of states and taking into account the effect of spin fluctuations, the temperature dependences of the magnetic susceptibilities were analyzed. The contribution of spin fluctuations, for the general model of itinerant electron paramagnetism was considered in the classical gaussian statistics [15,16]. In this model, it is possible to estimate quantitatively the contributions to the susceptibilities, from the spin fluctuations, in terms of a realistic density of states as [5,15,16]:

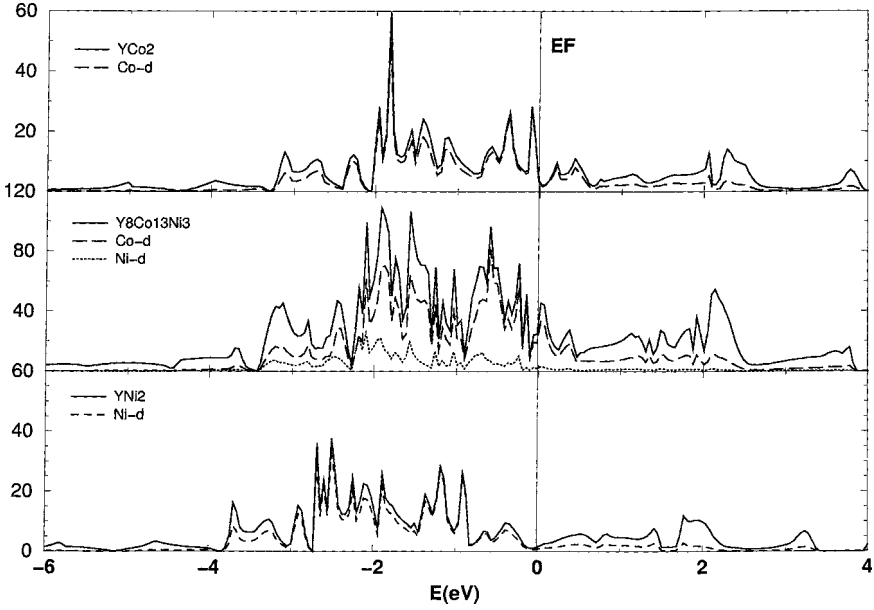

FIGURE 2 Band structures of YCo_2, Y_8Co_{13} Ni_3 and YNi_2 compounds.

$$\chi_s^{-1} = a_1 - \alpha + \frac{5}{3}a_3\,\xi^2 + \frac{35}{9}a_5\,\xi^4 + \frac{35}{3}a_7\,\xi^6 + \cdots \tag{2}$$

where the mean square value of the fluctuating magnetization, ξ^2, is given by:

$$\xi^2 = \frac{3}{2\pi^2}k_B T q_m A^{-1}\left(1 - \frac{tg^{-1}\left(q_m\sqrt{A\chi}\right)}{q_m\sqrt{A\chi}}\right) \tag{3}$$

We denoted by a_1, a_3, a_5 and a_7 the expansion coefficients of the free energy with respect to the square of the magnetization density and α, q_m and A denote the molecular field coefficient, the cut-off wave vector of spin fluctuations and the exchange stiffness constant, respectively.

In the following we limit the series expansion development up to terms in ξ^2. The explicit expressions for a_i coefficients were given as [15,16]:

$$a_1 = \frac{2}{g^2\mu_B^2 N}\left\{1 + \frac{1}{6}(\pi k_B T)^2\left[\left(\frac{N'}{N}\right)^2 - \frac{N''}{N}\right] + \cdots\right\} \tag{4}$$

$$a_3 = \frac{g^2\mu_B^2}{2^2 3!} a_1^3 \left\{ 3\left(\frac{N'}{N}\right)^2 - \frac{N''}{N} + \frac{1}{6}(\pi k_B T)^2 \right.$$
$$\left. \times \left[6\left(\frac{N'}{N}\right)^4 - 13\frac{N''N'^2}{N^3} + \left(\frac{N''}{N}\right)^2 + 7\left(\frac{N'''N'}{N^2}\right) - \frac{N''''}{N} \right] + \cdots \right\} \quad (5)$$

We denoted by N the state density at the Fermi level and N', N'', N''' and N'''' are their derivatives of order from one up to four.

We analyzed the core diamagnetism and of the Pauli paramagnetic contributions due to fraction of YNi_2 to the magnetic susceptibilities of compounds with $x \geq 0.2$. The resultant contributions somewhat cancel and are smaller than 10^{-4} emu/f.u. Consequently, in the following, we neglect in our analysis this contribution.

The magnetic susceptibilities computed with the parameters from Table 1 are plotted in Figure 1 by solid lines. These describe rather well

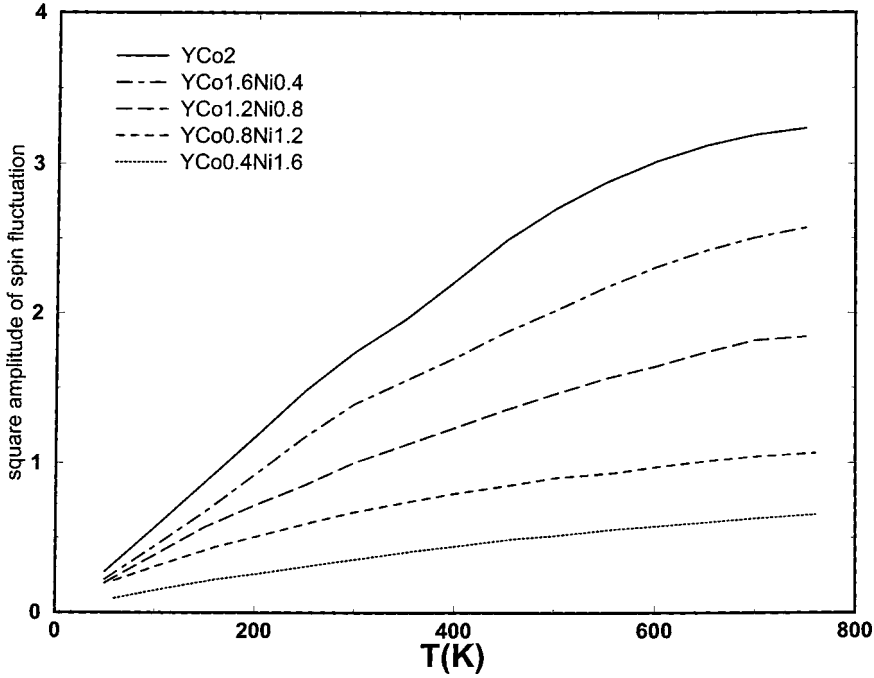

FIGURE 3 Temperature dependences of the mean square amplitudes of spin fluctuations for $Y(Co_xNi_{1-x})_2$ compounds with $x \geq 2$.

TABLE 2 Experimentally Determined and Computed b Values

	$b \cdot 10^6$ (K^{-2})	
Composition	Exp.	Theor.
$x = 1.0$	1.80	2.06
$x = 0.8$	1.30	1.52

the experimental data. The cut of wave vectors q_m increase and the exchange shiftnes constants decrease when increasing the nickel content.

The temperature dependences of the mean square amplitude of spin fluctuations are plotted in Figure 3. There is a tendency to saturate at higher temperatures as the cobalt content increase.

The temperature dependences of the susceptibilities, in the low temperature range ($T \leq 10\,K$) were also evaluated. In the paramagnon picture [17,18] the b coefficient from the relation (1) is given by $b = \frac{\pi^2}{6}\left[2\frac{N''}{N} - 1.2\left(\frac{N'}{N}\right)^2\right]_{E_F} s^2$, where s is the exchange enhancement factor. We note that the expression of the b coefficient, in this model, is near the same as that of the T^2 term of relation (4). The b values computed from band structures are close to those experimentally determined – Table 2.

4. DISCUSSION

The interesting magnetic behaviour of $Y(Co_xNi_{17-x})_2$ compounds may be mainly attributed to cobalt atoms. The Ni, as evidenced in YNi_2, has a small magnetic susceptibility. This is also in agreement with band structure calculations of YNi_2 which show that the Fermi level is situated in the region with low density of states. The experimentally observed behaviour of cobalt for compounds with $x \geq 0.2$, may be described in the spin fluctuation model. The model considers the balance between the frequencies of longitudinal spin fluctuations, which are determined by their lifetime, and of transverse fluctuations that are of thermal origin. These effects lead to the concept of the temperatures induced moments. For a strong exchange enhanced paramagnet, as evidenced for cobalt in the present compounds, the wavenumber dependent 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 $\xi^2 = 3k_BT\sum_q \chi_q$ increases with temperature and finally reaches an upper limit at characteristic temperature T^*. For $T > T^*$, a Curie-Weiss behaviour is predicted, similar to the system having local moment. These moments are localized in q space.

The analysis of the temperature dependences of computed ξ^2 values – Figure 3 – show that as the nickel concentration is increased, there is a tendency to saturate the spin fluctuations at lower T_s^* temperatures. This is in agreement with experimental data which show that T^* values, starting from which Curie-Weiss behaviour is evidenced, decrease when increasing nickel content. However, from the comparatively analysis of the computed ξ^2 values and of thermal variations of susceptibilities it can be seen that the saturation of spin fluctuations is obtained at temperatures $T_s^* > T^*$. This may be due to the approximation used in determining q_m and A values. We limited the series development for χ up to terms in a_3. In addition we neglected a Pauli-type contribution due mainly to the fraction of YNi_2 present in a given compound as well as the core diamagnetism. To the above must be added the considered temperature range in which the linear χ^{-1} vs. T has a linear dependences. These may be connected with experimental errors as well as the little changes in slopes when small variation in ξ^2 are present, particularly in the limit of 10%, when approaching to saturation. In spite of used approximations the differences between ξ^2 values experimentally determined and those computed in high temperature range, where nearly saturation is obtained, are rather small. Thus the computed curves describe reasonably the experimental data.

The exchange stiffness constants A, decrease gradually when the nickel content increase – Table 1. This is in agreement with the decrease of the exchange enhancement factor, as evidenced from susceptibility data.

Finally, we conclude that the temperature dependences of the magnetic susceptibilities of $Y(Co_xNi_{1-x})_2$ compounds can be reasonably described starting from the calculated density of states and taking into account the effect of spin fluctuations. The estimated parameters in describing the experimental data are reasonable ones and in good agreement with those found previously for YCo_2 compound [5].

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