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MAGNETOELASTIC PHASE TRANSITIONS AND CRITICAL BEHAVIOUR OF $(Fe_{1-x}Ni_x)_2P$ SYSTEM

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This report focuses on magnetoelastic properties and critical behaviour of $(Fe_{1-x}Ni_x)_2P$ system. For low Ni content ($x = 0.01$) an isolated critical point was found. Analysis of this critical behaviour in the frame of molecular field approximation was carried out. Moreover, thermal variations of the lattice parameters, as measured by X-ray diffraction techniques, were studied. The (P,T) phase diagrams obtained for several Ni contents show the evidence that the Curie temperature decreases with pressure. The Mössbauer spectra collected for $x = 0.025$, 0.1, 0.2, 0.25 at different temperatures are also analysed. The existence of two nonequivalent crystallographic sites occupied by iron atoms as well as local magnetic structure is discussed.

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1. Introduction

The $(Fe_{1-x}Ni_x)_2P$ series of compounds crystallise within Fe_2P-type structure ($P\bar{6}2m$ space group) in the whole composition range. For low nickel contents the Curie temperature T_C increases vs. x, however T_C strongly decreases with x for $x > 0.1$ and in the vicinity of $x = 0.8$ ferromagnetism is not observed [1–4]. Strong correlations between resistivity $R(T)$ vs. temperature variation and magnetic ordering were found. From electronic band structure calculations [3] it was concluded that the polarisation factor P varies with composition and reaches maximum value of $P = 82\%$ at $x = 0.1$, close to $x = 0.08$ when the largest value of the Curie temperature T_C and a kink in $R(T)$ were detected. The aim of this report is to analyse the magnetoelastic properties and critical behaviour of $(Fe_{1-x}Ni_x)_2P$ series of compounds.

2. Experimental

Polycrystalline samples were used in our studies. The samples were synthesised starting from 99.9% pure elements in the appropriate proportions [1, 4]. The mixtures of fine powder were mixed, then progressively heated to 850°C within 8 days in evacuated silica tubes. A final heat treatment, performed by high frequency heating, allows to melt the sample and then annealing by cooling down. The precise magnetisation and susceptibility measurements were carried out in the 80–300 K temperature range using a RG Cahn automatic electrobalance and a DC magnetometer/AC susceptibility Lake-Shore type 7225. The measurements of the a.c. susceptibility under pressure were carried out in the temperature range of 80–400 K and applied hydrostatic pressure up to 1.5 GPa. The Mössbauer spectra were recorded in transmission geometry using ^{57}Co in Rh source of 40 mCi activity. The grated samples, mixed with lucide powder, were prepared in a form of pellets of 18 mm in diameter. The low temperature measurements were carried out in a gas-flow nitrogen cryostat where temperature was stabilised by a build-in oven within an accuracy of 1 K.

3. Results and discussion

A ferromagnetic behaviour with $T_C = 260$ K is observed for $(Fe_{0.99}Ni_{0.01})_2P$. Above T_C the $M_B(T)$ curves show evidence of a phase transition with a small temperature hysteresis effect. The same type of behaviour was also confirmed in $M_T(B)$ curves [2, 5]. This field induced phase transition occurs between paramagnetic and ferromagnetic phases. In a very low field ($B = 10^{-4}$ T) the phase transition from ferro- to paramagnetic state has a discontinuous character (the width of hysteresis is about 5 K), but under fields above 1.5×10^{-2} T the hysteresis amplitude decreases below 1 K and the transition becomes rather the second order one. It may be concluded that in the field of ≈ 0.1 T an isolated critical point is observed.

It is worth noting that such a type of behaviour was also observed in Fe_2P [6] and in some other related series of compounds [7, 8]. This critical behaviour was analysed on the basis of the generalised Gibbs free energy $G_j(\sigma, T)$. The following terms were accounted for: exchange energy as calculated on the basis of molecular field approximation, Zeeman energy, elastic energy, entropy term, and pressure term [9]. Minimising the free energy, with respect to the volume in the case of no external pressure and with respect to σ when an external magnetic field is applied, we obtain an implicit expression of the temperature dependent relative magnetisation σ [7]. A numerical analysis of the magnetisation $\sigma_H(T)$ plots was carried out using the parameters $j = \frac{3}{2}$, $\eta = 1.1$, and $T_0 = 255$ K. These parameters lead to the isolated critical point, which was found experimentally, and to the best fit with our experimental results.

Electronic band structure calculations using a Korringa–Kohn–Rostoker (KKR) method were already undertaken for the parent Fe_2P compound in the paramagnetic (non-magnetic) state [7]. They reveal the existence of a deep mini-

mum in the vicinity of the Fermi energy E_F. Moreover, the value of E_F is in the region where the first derivative of the density of states is very large and the second derivative of the DOS curve calculated at E_F is positive. This indicates that a metamagnetic field induced phase transition between the para and the ferro states should be observed (metamagnetism of itinerant electrons) [8].

X-ray diffraction measurements for the samples with $x = 0.01$, 0.025, and 0.1 were carried out at temperatures from 100 to 400 K. A jump in the lattice parameters associated with magnetoelastic transition was found in the studied compounds. It remains in good agreement with the results previously obtained by Becman and Lundgren [10]. Based on the lattice constants and on the atomic position parameters the interatomic distances were calculated for studied contents. It may be concluded that in the ferromagnetic–paramagnetic phase transitions: (i) the Fe(3g)–Fe(3g) atom distances significantly increase (from $3.07_5 \times 10^{-10}$ m to $3.12_3 \times 10^{-10}$ m); (ii) the Fe(3g)–(Fe,Ni)(3f) atom distances decrease (from $2.69_4 \times 10^{-10}$ m to $2.66_0 \times 10^{-10}$ m); (iii) the (Fe,Ni)(3f)–(Fe,Ni)(3f) atom distances markedly increase (from $2.50_0 \times 10^{-10}$ m to $2.68_3 \times 10^{-10}$ m). Moreover, Fe–Fe interatomic distances are higher than the critical separation distance for iron atoms (2.60×10^{-10} m). It suggests that for all studied contents the impact of ferromagnetic coupling between the iron atoms is confirmed.

A.c. susceptibility measurements under pressure were carried out for compounds with $x = 0.025$, 0.1, 0.2, 0.25. A linear decrease in the Curie temperature vs. the applied pressure was evidenced for all samples studied. It remains in a good agreement with the T_C variations with pressure previously found [4, 11]. The obtained values of dT_C/dP are equal to –9.5, –4.5, –4.5, 0.0 K/GPa for samples with $x = 0.025$, 0.1, 0.2, 0.25, respectively. Note that dT_C/dP slightly decreases with the increase in Ni content in the solid solution. The shape of the a.c. susceptibility curves changes slightly in the vicinity of T_C, however no pressure induced antiferromagnetic state, similar to the one observed in the case of Fe_2P, was detected here.

Now we discuss the Mössbauer spectroscopy results. The samples with $x = 0.2$ and 0.25 were measured above and below the magnetic transition temperature, while the compounds with $x = 0.05$ and 0.1 at room temperature only. The Mössbauer spectra are shown in Fig. 1. The least squares method was used to refine the spectra on the basis of a set of quadrupole doublets in the case of paramagnetic fractions and of a sextets for magnetically ordered patterns. Iron is located in two types of local states with different isomer shifts (IS) and quadrupole splittings (QS). According to the previous studies [2, 5] the state with low IS (0.25–0.23 mm/s) and QS about 0.30 mm/s may be assigned to the tetrahedral site and the state with high IS about 0.59 mm/s and QS about 0.76 mm/s to the pyramidal site. The results of the Mössbauer measurements can be summarised as follows. Only sample with $x = 0.25$ is paramagnetic at room temperature. A decrease in Ni content (x) increase both, in the temperature of magnetic transition and in the values of internal hyperfine magnetic fields. It is interesting to note that below T_C the finite fraction of iron in the tetragonal sites remains paramagnetic even at 80 K (9% for $x = 0.25$ and 6% for $x = 0.2$). In this case only one value of

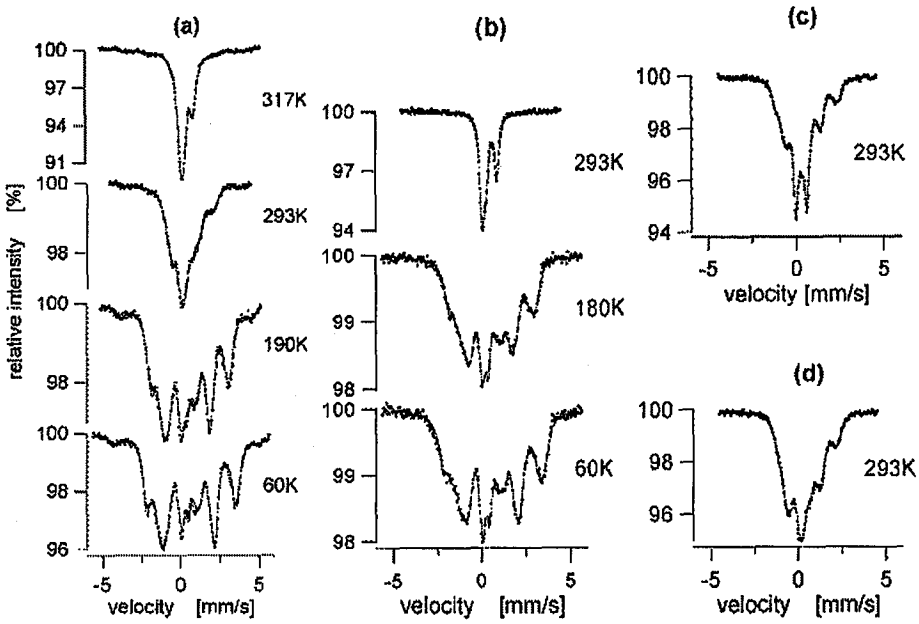

Fig. 1. Mössbauer spectra of (a) $(Fe_{0.75}Ni_{0.25})_2P$, (b) $(Fe_{0.80}Ni_{0.20})_2P$, (c) $(Fe_{0.90}Ni_{0.10})_2P$ and (d) $(Fe_{0.975}Ni_{0.025})_2P$ measured at different temperatures.

internal magnetic field (10.5 T for $x = 0.25$ and 10.7 T for $x = 0.2$) was found for the magnetically ordered fraction. In a contrary, Fe in the pyramidal site is fully magnetically ordered below the magnetic transition.

The distribution of hyperfine fields is strongly influenced by the Ni content. For $x = 0.25$ three well determined hyperfine fields (7.8 T, 15.5 T, and 17.8 T at 80 K) are observed, while for $x = 0.2$ one magnetic field dominates (17.5 T at 80 K).

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