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To cite this version:

J. Puertolas, C. Rillo, J. Bartolome, Daniel Fruchart, S. Niziol, et al.. COMMENSURATE-INCOMMENSURATE PHASE TRANSITION IN (Co1-xMnx)2P. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-197-C8-198. <10.1051/jphyscol:1988885>. <jpa-00229110>

HAL Id: jpa-00229110
https://hal.archives-ouvertes.fr/jpa-00229110
Submitted on 1 Jan 1988

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COMMENSURATE-INCOMMENSURATE PHASE TRANSITION IN (Co$_{1-x}$Mn$_x$)$_2$P

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Abstract. - Neutron diffraction, a.c. initial magnetic susceptibility, magnetization, and heat capacity measurements allow a better understanding of the metamagnetic-like phase observed at low temperature in (Co$_{1-x}$Mn$_x$)$_2$P. The magnetic structures and phase transitions for $x = 0.6$ and 0.75 have been determined.

Introduction

The phase diagram of (Co$_{1-x}$Mn$_x$)$_2$P points out direct relations between crystal structure (orthorhombic with space group Pnma for $x \leq 0.80$ and hexagonal with space group P6$_3$2m for $x \geq 0.80$) and magnetic properties [1-3]. Co$_2$P is a Pauli paramagnet, whereas Mn$_2$P is an antiferromagnet with $T_N = 103$ K. When small quantities of Co ($x \geq 0.075$) are substituted by Mn, a ferromagnetic behaviour appears. The maximum Curie temperature, $T_c$, is achieved for the CoMnP compound [3]. Substitution of more Co by Mn reduces $T_c$ and induces an antiferromagnetic phase in the range $0.60 \leq x \leq 0.80$ at low temperatures [2, 3, 4]. Here we report on neutron diffraction, a.c. magnetic susceptibility, magnetization, and heat capacity (D.S.C.) measurements performed on powder samples, for $x = 0.60$ and 0.75. This work permits to analyse in detail the previously reported commensurate-incommensurate phase transition [4, 5] and to complete the high temperature range of the phase diagram.

Experimental and discussion

Thermal variations of the lattice constants have been determined by neutron diffraction and X-ray diffractograms. At low temperatures the cell volume remains constant, whereas above 140 K it expands continuously for $x = 0.75$ (Fig. 1). For $x = 0.60$ a drop is observed, toward the volume value close to that of ferromagnetic CoMnP. For $T \geq 400$ K the volume recovers a value which is an extrapolation of the low temperature regime. Neutron diffraction patterns evidence magnetic satellite reflections at temperatures below 150 K. This indicates an incommensurate magnetic ordering with propagation vector $q = [0, 0, q_z]$. Figure 2a gives the thermal variations of $q_z$. The high temperature measured value is close to 0.07 for $x = 0.60$ and to 0.33 for $x = 0.75$. A first order process could be invoked to justify the drops to zero at 150 K. Figure 2b shows the thermal variation of the intensity...
of some selected satellite reflections \(x = 0.75\). Similar variations were detected for \(x = 0.60\). In addition some of the two satellites attached to the nuclear lines have marked different intensities \(F_{hkl+q} \neq F_{hkl-q}\), as observed for other related compounds [6]. A selected set of relative phases between the different magnetic sublattices of the unit cell may explain this fact. Since Mn and (Mn-Co) atoms occupy 4c positions (sites 1, 2, 3, 4) of the \(P\text{mn}a\) space group, the spin modulation requires the same phase between moments of the pair sites 1-3 and 2-4 [6]. This suggest a “double-helix” magnetic structure for the incomensurate phase \((T < 150 \text{ K})\), as proposed for the iso-type ternary germanides and silicides [7, 8]. The best fit to the data is obtained for a circular helix in the \((a, b)\) plane, perpendicular to the propagation vector \(q = [0, 0, q_z]\). The incomensurate phase disappear at 150 K for both compounds. The phase diagram proposed earlier for \(x = 0.60\) by Roger [3] presents an AF-F transition and for \(x = 0.75\) and AF-Para (near 115 K !). However, the phases diagram determined by Okamoto [4] indicates that for \(x = 0.60\) there is no incomensurate-incomensurate phase transition, and that the extrapolated transition temperature is \(T = 170 \text{ K}\).

In order to precise the magnetic phase diagram complex a.c. initial susceptibility, \(\chi = \chi' - i\chi''\), measurements have been performed for \(x = 0.60\) and \(x = 0.75\), in the temperature range 80-300 K (Fig. 3). The in-phase component, \(\chi''(T)\), shows a step-like anomaly around 150 K in correspondence with the AF-F \((x = 0.60)\) and the so-called AF-Para \((x = 0.75)\) phase transitions. The out-phase component, \(\chi'(T)\), is negligible in the AF regime, whereas for \(T > 140 \text{ K}\) and \(x = 0.60\) it fastly increases indicating the rise of a net magnetic moment (F phase). Thermal hysteretic effects are clearly evidenced in the temperature range of the AF-F phase transition also indicating a first order process. For \(x = 0.75\), \(\chi'(T)\) does not show a paramagnetic behaviour above 150 K, in contrast with the previous phase diagrams [3, 4], and the absolute values are typical of a weak ferromagnetic phase (i.e. \(\chi'(x = 0.75)/\chi'(x = 0.60) = 0.09\) at 275 K).

Magnetization measurements, \(M(T)\), for \(x = 0.6\) confirm the AF-F transition and the ferromagnetic properties above 150 K \((T_c = 415 \pm 5 \text{ K})\) in consistence with neutron diffraction and a.c. susceptibility data. The \(T_c\) determined by DSC measurements is in good agreement with magnetization results. For \(x = 0.75\), the \(M(T)\) curve (Fig. 3) shows at 150 K a transition from an antiferromagnetic to a weak-ferromagnetic ordering, in agreement with neutron diffraction \((q_z \rightarrow 0)\) and a.c. susceptibility data. The Curie temperature \((400 \pm 5 \text{ K})\), is also in agreement with DSC measurements. The experimental entropy change \((0.2 \text{ mJ \text{ K}^{-1} \text{ g}^{-1}})\) is about one tenth of that measured at 415 K for the F-Para transition of \((\text{Co}_{0.4}\text{Mn}_{0.6})_2\text{P}\).

In conclusion we have resolved the magnetic structure of \((\text{Co}_{0.80-0.60})_2\text{P}\) for the compositions \(x = 0.60\) and \(x = 0.75\). The experimental evidence for a weak ferromagnetic state \((x = 0.75)\) indicates a more complicated phase diagram than that previously reported [3, 4]. More systematic measurements in the concentration range \(0.60 < x < 0.80\) are needed in order to precise the magnetic phase diagram.

Acknowledgements

Parts of this work have been accomplished at the Laboratoire de Cristallographie by some of the authors granted by CNRS (J.A.P.), by Université J. Fourier, Grenoble (S.N.), and by the French-Spanish program of “Actions Intégrées” (C.R.).