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On the electrical resistivity of $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ alloys in the exchange inversion region

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Résumé. — On présente les résultats de mesures de la résistivité électrique (ρ) en fonction de la température des alliages $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ (pour $x = 0; 0,5; 0,6; 0,7; 0,8$ et $1,0$). La courbe, $\rho(T)$ de $\text{Pt}_3\text{Fe}_{0,6}\text{Mn}_{0,4}$ montre deux anomalies, dont l'une est attribuée à la température de Curie et l'autre à la transition vers une structure hétérogène, non-colinéaire, magnétique, favorisant ainsi l'hypothèse que l'inversion de l'interaction d'échange arrive par l'intermédiaire d'une structure avec une orientation non colinéaire des moments magnétiques.

Abstract. — The electrical resistivity (ρ) data as a function of temperature are reported for $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ alloys (with $x = 0, 0.5, 0.6, 0.7, 0.8$ and 1.0). The $\rho(T)$ curve for $\text{Pt}_3\text{Fe}_{0.6}\text{Mn}_{0.4}$ shows two ρ -anomalies, one of which is attributed to the Curie temperature and the other to the transition to a heterogeneous non-colinear magnetic structure, thus favouring the hypothesis that the exchange reversal takes place via a system of structures with non-colinear orientation of the magnetic moments.

The solid solutions $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ are isostructural over the entire concentration range ($0 \leq x \leq 1$), ordering in an fcc lattice (Cu_3Au -type). The non-magnetic Pt atoms are ordered and occupy the 3(c) sites whereas the magnetically active Mn and Fe atoms are randomly distributed in 1(a) sites. The initial alloys have very close lattice parameters viz. $a = 3.89 \text{ \AA}$ for Pt_3Mn and $a = 3.87 \text{ \AA}$ for Pt_3Fe [1]. The unchanged crystal structure and a feeble composition dependence of the lattice parameter are ideal conditions for studying a transition in the magnetic state in $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$, from ferromagnetic in Pt_3Mn ($T_c = 390 \text{ K}$) to the antiferromagnetic in Pt_3Fe ($T_N = 170 \text{ K}$), as Fe atoms are gradually substituted for Mn, i.e. as x is increased from 0 to 1 [1, 2]. From an analysis of the neutron diffraction and magnetic measurements it was concluded that the sign of the exchange interaction $J_{\text{Mn-Fe}}$ reverses in the ferromagnetic-antiferromagnetic transition region ($0.5 \lesssim x \lesssim 0.7$) [3]. The heat capacity of $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ alloys has been measured by Kourov *et al.* [4]. In this paper we report electrical resistivity (ρ) data as a function of temperature (100-500 K) for alloys with compositions corresponding to $x = 0, 0.5, 0.6, 0.7, 0.8$ and 1.0 , and discuss the observed ρ -anomalies in the light of the existing information on the magnetic structure.

1. Experiment.

The alloys were prepared by repeated melting of the high-purity constituent metals in an electric arc furnace under argon atmosphere. The alloy buttons thus obtained were homogenized by

annealing in evacuated silica capsules at 1 150 K for one week. All the annealed specimens were found to be single-phase by X-ray analysis. The samples for electrical resistivity ($\sim 1 \times 1 \times 8 \text{ mm}^3$) were cut and reannealed for 72 hours before being taken up for measurements by standard four-probe dc method. The relative values of resistivity are estimated to be accurate to $\pm 0.05 \%$ but the uncertainty in sample dimensions limited the absolute resistivity values to be accurate to within $\pm 5 \%$. The fluctuation in temperature did not exceed 0.25 K during any particular measurement. No hysteresis was observed for cooling runs, hence only the heating runs are being reported in figure 1.

Fig. 1. — Electrical resistivity ρ as a function of temperature for $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ for different values of x . Arrows denote the magnetic transition temperatures.

2. Discussion.

The general behaviour for all the alloys is typical of magnetically ordered materials in which the scattering of conduction electrons by both the phonons and the disordered spins make significant contributions to resistivity, and comprising a point of inflection at the respective Néel (or Curie) temperature. The magnitude of the room-temperature resistivity and the temperature of the point of inflection ($T_N \simeq 170 \text{ K}$) for $x = 1.0$, i.e. Pt_3Fe are in good agreement with literature [5]. Since no « antiferromagnetic » reflections were observed in the range $0 \leq x \lesssim 0.5$ and no « ferromagnetic » reflections in the range $0.7 \lesssim x \leq 1$, in the neutron diffraction spectra [3], the temperature corresponding to the point of inflection in the $\rho(T)$ curves, viz. at 380 K and 304 K for $x = 0$

and $x = 0.5$, respectively, can be attributed to the Curie temperatures, and those at 196 K, 174 K and 170 K can be attributed to the Néel temperatures of the alloys with $x = 0.7, 0.8$ and 1.0 , respectively.

The $\rho(T)$ plots for $x = 1.0, 0.8$ and 0.7 show relatively sharp change of slope at the transition temperatures (Fig. 1). The plot for $x = 0.6$, however, shows the inflection points namely at 240 K and at 300 K. This is consistent with the heat capacity results on $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ [4], where sharp λ -type heat capacity anomalies were obtained for alloys with $x = 1.0, 0.8$ and 0.7 at 164 K, 189 K and 191 K, but the $C_p(T)$ curve for the alloy $\text{Pt}_3\text{Fe}_x\text{Mn}_{0.4}$ ($x = 0.6$) showed two strongly smeared heat capacity maxima. Kourov *et al.* demonstrated that the magnetic heat capacity for this alloy ($x = 0.6$) showed a discontinuity at 240 K and suggested that the concentration ferromagnetic-antiferromagnetic transition in $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ appears to be in the vicinity of a critical composition $x_{\text{cr}} = 0.6$ [4]. Dubinin *et al.* [6] have also obtained evidence of a complex magnetic structure for $\text{Pt}_3\text{Fe}_{0.6}\text{Mn}_{0.4}$ from their neutron diffraction pattern recorded at 4.2 K which cannot be attributed to a finely dispersed mixture of ferromagnetic and antiferromagnetic phases, the formation of which would entail considerable broadening of the magnetic reflections which is not observed experimentally. These authors have used the recently developed group theoretical and molecular field methods to determine the magnetic structure, and proposed that the ferromagnetic-antiferromagnetic transition in $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ passes through a system of non-collinear magnetic structures. The magnetic phase diagram constructed by them is shown in figure 2, where $T_{\text{c,N}}/T_1$

Fig. 2. — Magnetic phase diagram of $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ system from reference [6]. T_1 denotes the Curie temperature of Pt_3Mn . Curve 1 is a calculated curve for ferromagnetic structure whereas 1' and 2 are calculated assuming non-collinear structures. Closed circles (●) denote present data from Fig. 1.

is plotted as a function of x , T_1 being the Curie temperature of the alloy Pt_3Mn and T_{c} and T_{N} are the temperatures at which coherent neutron diffraction reflections $(100)_{\text{ferro}}$ and $(1/2 \ 1/2 \ 0)_{\text{af}}$ vanish. The curve 1 is calculated for ferromagnetic structure and curves 1' and 2 are calculated for non-collinear structures [6]. It is interesting to note that the temperature corresponding to the two inflection points (*viz.* 300 K and 240 K) in the $\rho(T)$ plot for $x = 0.6$ composition agree with curve 1 and 1', respectively, implying that for this alloy in addition to the ferromagnetic domains there is also evidence of a heterogeneous non-collinear magnetic structure.

In the case of the alloy $\text{Pt}_3\text{Fe}_{0.7}\text{Mn}_{0.3}$ ($x = 0.7$) we could observe only one point of inflection corresponding to T_{N} (*i.e.* curve 2 in Fig. 2). It has been remarked that the ferromagnetic reflection (100) in this alloy is very low in intensity owing to the proximity of the concentration $x_0 \simeq 0.71$, at which it vanishes [6], making any ρ -anomaly corresponding to curve 1 unobservable. Any transition corresponding to the non-collinear structure (1') should possibly appear only below the range of temperature of present measurements.

In conclusion, the present resistivity analysis lends support to the conjecture that the ferromagnetic-antiferromagnetic transition in $\text{Pt}_3\text{Fe}_x\text{Mn}_{1-x}$ is *via* a system of structures with non-collinear orientation of the atomic magnetic moments of the alloy components. An alternate possibility that this transition could come about by the nucleation and growth of a collinear antiferromagnetic (Pt_3Fe) structure in a matrix with a collinear ferromagnetic (Pt_3Mn) structure with increase in x was ruled out since there were no signs of broadening of magnetic reflections in the transition region ($0.5 \lesssim x \lesssim 0.7$) [2].

Finally, the less than linear increase in the resistivity in the paramagnetic regime, in the case of concentration $x = 1.0$ (and to some extent in the case of $x = 0.8$ and 0.7) points to a tendency towards resistivity saturation i.e. a reduction in the electronic mean free path in such high resistivity alloys [7].

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