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X-RAY AND SUSCEPTIBILITY STUDY OF THE FIRST-ORDER MAGNETIC TRANSFORMATION IN Mn_3Pt

E. Krén, P. Szabó, L. Pál, T. Tarnóczi,

G. Kádár and C. Hargitai

HUNGARIAN ACADEMY OF SCIENCES
CENTRAL RESEARCH INSTITUTE FOR PHYSICS

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Abstract

The first-order transformation from triangular to collinear antiferromagnetic structure /AF-AF transformation/ has been investigated in the $Mn_3Pt_{1-x}Rh_x$ and $Mn_{3+y}Pt_{1-y}$ systems by x-ray diffraction and magnetic methods.

At the AF-AF transformation temperature T_t a discontinuous change in the lattice parameter is observed with a magnitude depending on concentration but with always the same lattice parameter value of the collinear phase. This implies the existence of a critical lattice parameter value for the AF-AF transformation. Below the Néel temperature T_N an anomaly in the thermal expansion occurs indicating exchange interactions dependent on the interatomic separation. This anomaly differs in sign for the collinear-paramagnetic and triangular-paramagnetic transitions. The susceptibility shows an abrupt change at T_t and a maximum at T_N in the triangular phase. No maximum occurs for the collinear phase. The observed transition temperatures are in agreement with the earlier neutron diffraction data.

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Central Research Institute for Physics, Budapest, Hungary

In Mn_3Pt a first-order antiferromagnetic-antiferomagnetic /AF-AF/ transformation was observed by neutron diffraction [1] . On heating, the magnetic structure transforms from triangular to collinear. The concentration dependence was measured for the transition temperature T_t, the Néel temperature T_N, the lattice parameter a and the magnetic moment μ_{Mn} of the manganese atoms in the ternary $Mn_3Pt_{1-x}Rh_x$ and the binary $Mn_{3+y}Pt_{1-y}$ systems [2] . On increasing the concentrations x and y, an increase in both T_t and T_N with a simultaneous decrease in the lattice parameter were observed, while μ_{Mn} was found to be the same irrespective of the concentration and the AF-AF transformation.

In order to see the role of the lattice parameter in the AF-AF transformation and to check the transition temperatures by other methods, the variations of the lattice parameter and the magnetic susceptibility with temperature were investigated by x-ray diffraction and magnetic methods in the $Mn_3Pt_{1-x}Rh_x$ and $Mn_{3+y}Pt_{1-y}$ systems on the same samples as used in the earlier studies [1, 2].

The x-ray measurements were performed in gas atmosphere containing 30 % H_2 and 70 % N_2 in a high temperature furnace attached to a Philips diffractometer. For susceptibility measurements the specimens were sealed in quartz ampoules at 10^{-4} mmHg pressure and a magnetic balance was used.

The temperature curves for the lattice parameter in Fig. 1 show striking changes at the transition temperatures T_t and T_N. At T_t a discontinuity is observed the magnitude of which varies with concentration. On the other hand, the lattice parameter of the collinear phase at this discontinu-

ity has the same value at all concentrations. In the transition range the lattice parameters associated with the triangular and collinear phases coexist. Below T_N in a temperature interval of about 100° C the slope of the lattice parameter curves is seen to decrease for the collinear phase /see the curve for y=0,07 in Fig. 1/ and to increase for the triangular phase /see e.g. the curve for x=0,2/ with respect to the paramagnetic phase. Below the temperature range of this anomaly the values of the thermal expansion coefficients are 0,80 and 1,2 . 10^{-4} Å/°C for the collinear and triangular phases, respectively, as compared with 1,1.10^{-4} Å/°C for the paramagnetic phase, these values being independent of the concentration within the experimental accuracy.

The temperature dependence of the magnetic susceptibility is shown in Fig. 2. At T_t an abrupt change is observed, the susceptibility in the collinear is higher than in the triangular phase. At T_N the maximum which appears for the triangular phase /see the curve for x=0,5/ cannot be observed for the collinear one. The susceptibility is independent of the field strength in both the triangular and collinear phases. No reliable data could be obtained on some of the ternary alloys because of the complex magnetic properties which are, probably, due to impurities.

The values of the transition temperatures obtained from present measurements are in agreement with those from the earlier study [2] but, in contrast with the neutron results, the transformation at 505° K in the sample with x=0,1 was found to be of first-order. This observation slightly modifies the reported phase diagram [2] . The inconsistency between the experiments can be explained by the closeness of T_t and T_N in this sample.

As seen in Fig. 1, the change in the lattice parameter at T_t is sensitive to the magnitude of the anomalous thermal expansion below T_N and to the distance between T_t and T_N. If the AF-AF transformation exists, the T_N for the triangular phase is higher than T_t and can be, therefore, estimated only by extrapolation using the modified phase diagram of Ref. 2. The curves for the triangular phase in Fig. 1 are extended by dashed lines as far as these extrapolated Néel temperatures T_N'. The T_N values given in Fig. 1 for x=0,1 and y=09 are also extrapolated since they could not be obtained directly due to their closeness to T_t. As seen, the anomalous expansion decreases with increasing x and with deviation from stoichiometry. The stronger the anomaly and the larger the distance between T_t and T_N, the larger is the discontinuity observed at T_t.

The anomalous thermal expansion observed below the Néel temperatures indicates the dependence of the magnetic interactions on the interatomic separation [3] . The increased thermal expansion below the T_N of the triangular phase implies an interaction which increases with the decrease in the interatomic separation, while the decreased expansion below T_N of the collinear phase indicates an interaction which decreases with the increase in interatomic separation. Considering the spin configurations it can be assumed that while in the triangular structure it is the nearest neighbour interaction which is dominant and responsible for the anomalous thermal expansion, in the collinear structure it is the next nearest neighbour one. Thus the experimental facts can be explained by assuming two antiferromagnetic interactions, a nearest neighbour one decreasing and a next nearest neighbour one increasing with increasing interatomic separation. The x-ray measurements show the existence of a critical lattice parameter value of the collinear phase at which the AF-AF transformation takes place: $a_{crit}=3,873\pm0,003$ Å. It can be concluded that the AF-AF transformation is closely related to the ratio of the two interactions at a_{crit}.

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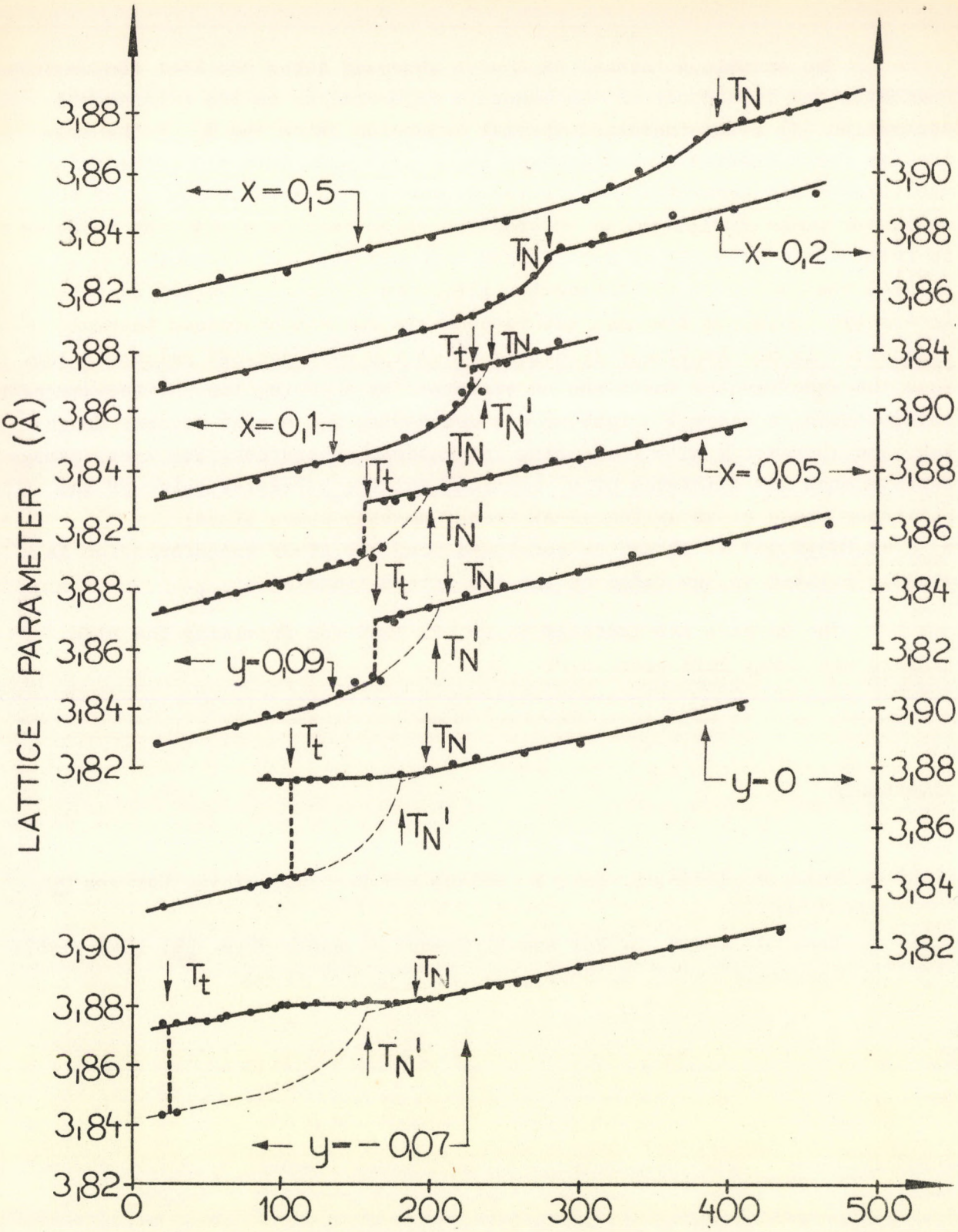

Fig. 1 Temperature dependence of the lattice parameter for $Mn_3Pt_{1-x}Rh_x$ and $Mn_{3+y}Pt_{1-y}$ alloys

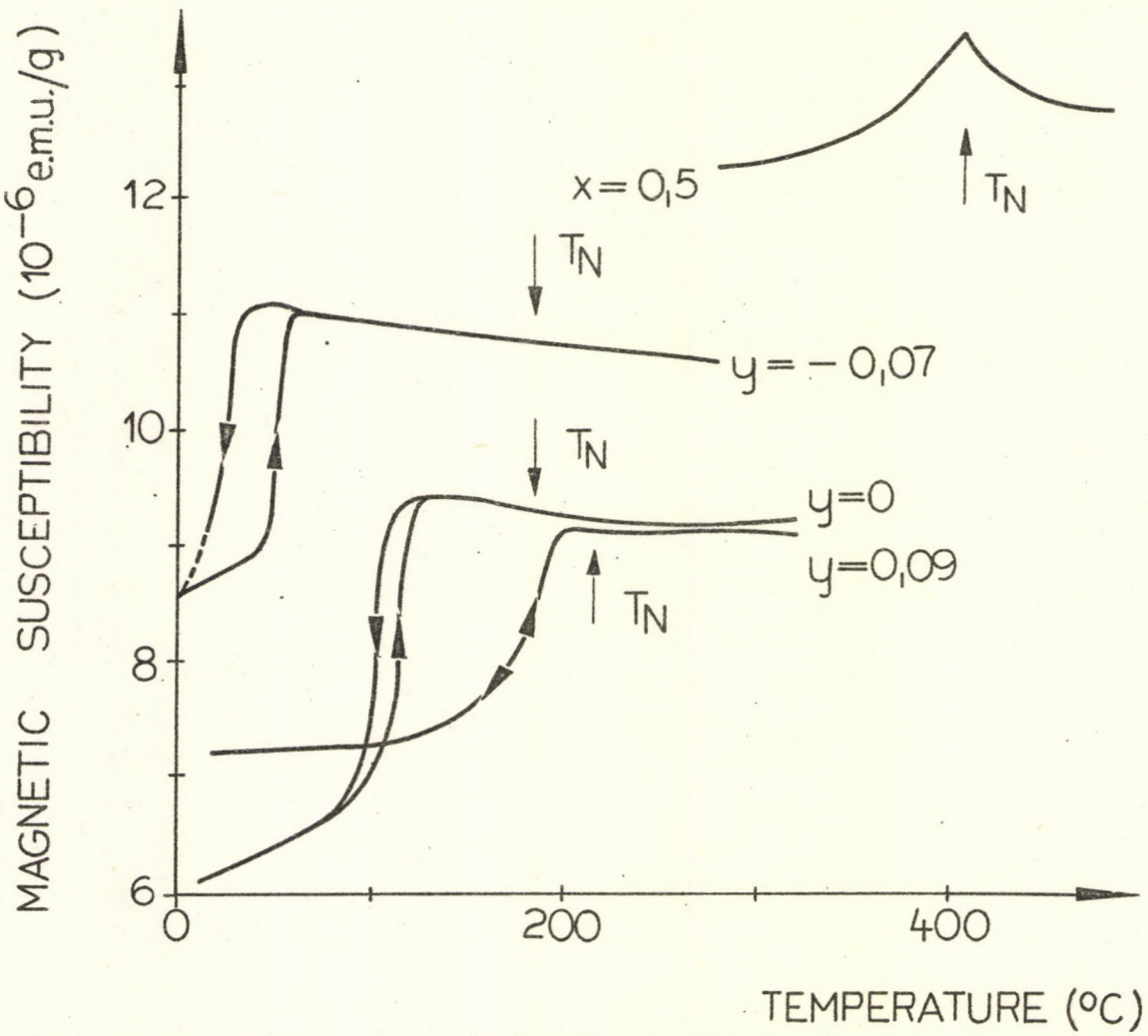

Fig. 2 Temperature dependence of the magnetic susceptibility for $Mn_3Pt_{1-x}Rh_x$ and $Mn_{3+y}Pt_{1-y}$ alloys

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