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Electrical-resistivity and ac-susceptibility measurements on Ni-Mn and Ni-Mn-Pt ternary alloys

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We present electrical-resistivity and ac-susceptibility measurements for both reentrant-spin-glass (RSG) $Ni_{76-x}Mn_{24}Pt_x$ alloys ($0<x<3$) and pure spin-glass (SG) $Ni_{72-x}Mn_{28}Pt_x$ alloys ($0<x<10$) in the 4.2–300 K temperature range. The freezing temperature (T_g) of the RSG system remains almost constant with nonmagnetic Pt impurity addition, while that of the SG system slightly increases. It is found that the SG alloys exhibit resistivity minima which shift towards higher temperatures with increasing Pt concentrations. However, the minima for the RSG system take place at temperatures below 5 K. It has been found that the resistivity curves for both the SG and RSG systems change slope at temperatures where corresponding ac-susceptibility curves exhibit maxima. These results have been interpreted in terms of domain-anisotropy and Saslow-Parker models.

Extensive studies have been done on SG systems in the last decade.[1] Various kinds of spin-glass (SG) materials have been investigated; the ac susceptibility with the appearance of a cusp at T_g and magnetization with remenance below T_g are characteristic features. From detailed magnetic studies of Ni-Mn alloys, $N_{100-x}Mn_x$ systems are found to be ferromagnetic (FM) and reentrant spin-glass (RSG) for $x<25$ and SG-like above these concentrations. The multicritical point (MCP) for these alloys lies around 24.5-at.% Mn concentration.[2] In the present study we have chosen two different alloy systems: the RSG alloy $Ni_{76-x}Mn_{24}Pt_x$ where $0<x<3$ and the SG alloy $Ni_{72-x}Mn_{28}Pt_x$ where $0<x<10$.

Here, we report measurements of resistivity $\rho(T)$ and ac-susceptibility $\chi(T)$, as functions of temperature for the alloys mentioned above. Recently, it has been reported that NiMn alloys have resistivity minima at low temperatures and the minima could reflect the emergence of a transition from an ordered to a disordered magnetic phase.[3] This assertion has been supported by magnetoresistance data that also exhibited an unambiguous peak at corresponding temperatures. We believe that our data will provide essential insight not only to clarify the origin of the minima associated with the spin-glass-like disorder but also support the proposed domain-structure model at low temperatures.

Samples in sealed quartz tubes at 0.3-atm Ar pressure were melted twice using a rf induction furnace to insure better homogenization. Cold working was employed to roll the alloys into sheets from which experimental samples, $20\times1.5\times0.2$ mm^3 in size, were cut using a diamond saw. Finally, all samples were annealed at 850 °C for 1 h in a vacuum-pumped quartz tube and quenched by allowing high-pressure Ar gas into the tube to shoot them into cold water. Resistivity measurements were carried out with the standard four-point-probe potentiometric technique. Voltage accuracy was one part in 10^5 and the current was maintained constant to one part in 10^5 as well. Temperature was stabilized to 1 mK in a conventional He cryostat using a computer-controlled data-acquisition system. Both ac and dc results were the same within the experimental accuracy of the apparatus. ac susceptibility was measured at 1.8 Oe at 17 Hz, using an ac mutual-inductance probe, which consisted of a pair of astatically wound coils.

Variation of the resistivity $\rho(T)$ with the temperature for the alloys $Ni_{76-x}Mn_{24}Pt_x$, where $x=0.0$, 0.5, 1.5, 2.0, and 3.0 at% Pt, is given in Fig. 1. The change of slope at T_g is obvious by close examination of the curves and from the peaking of dR/dT in Fig. 2, and shallow minima are suggested by the data at temperatures below 5 K, just as reported by Senoussi and Oner.[3] Resistivity minima could not be measured for these samples due to our experimental limits, however, the variation of the curves around 5 K clearly indicates that the minima would occur at lower temperatures. Furthermore, as illustrated by Fig. 2, the absolute value of dR/dT decreases with increasing Pt

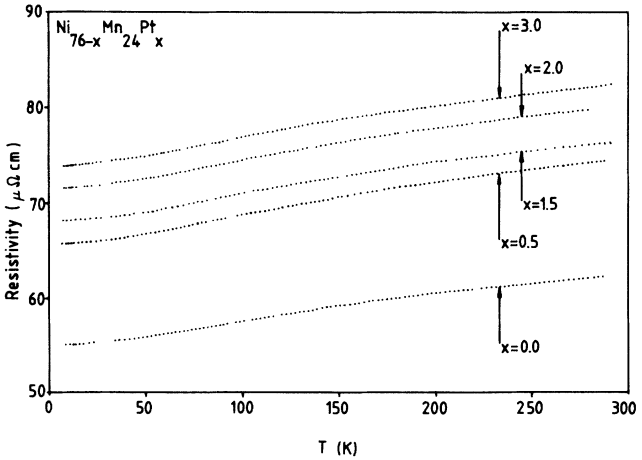

FIG. 1. Temperature variation of resistivity for $Ni_{76-x}Mn_{24}Pt_x$ alloys.

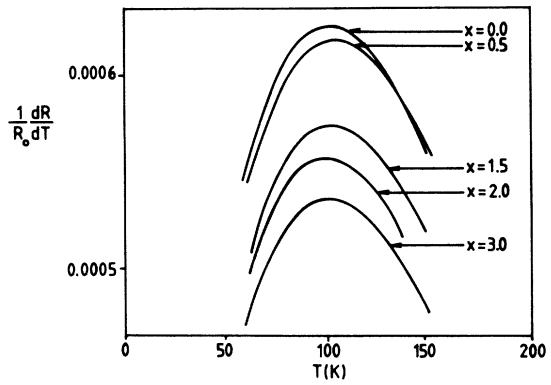

FIG. 2. Temperature dependence of the derivative of the resistivity dR/dT.

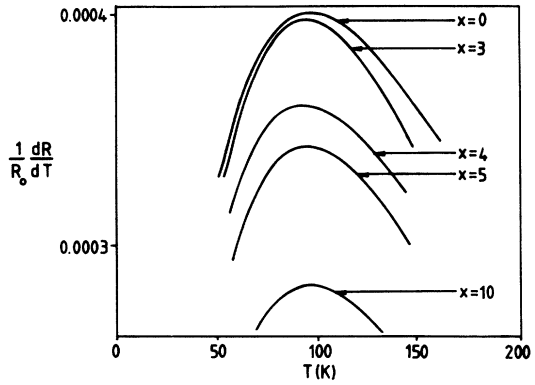

FIG. 4. Temperature dependence of the derivative of the resistivity dR/dT.

impurity, while T_g remains constant. Figures 3 and 4 show variation of the resistivity and its derivative for $Ni_{72-x}Mn_{28}Pt_x$ alloys, respectively. Although the behavior of the resistivity as a function of temperature is similar to that of 24-at.% Mn alloys, the minima are observed to shift to higher temperatures. It is clear that the well-defined minima curves are deeper for higher Pt concentrations (Fig. 5). Senoussi and Oner have reported that at lower temperatures, the minima are not logarithmic in nature (Fig. 2 in Ref. 3). Our results confirm this observation (Fig. 5 inset).

Oner and Aktas have recently reported that Pt impurities have no detectable effect on Tg of $NiMn_{24}$ alloys within experimental uncertainty.[4] This finding is in agreement with the fact that Ni and Pt are isoelectronic elements. However, in this investigation a slight increase in T_g with increasing Pt impurity level has been detected for $Ni_{72-x}Mn_{28}Pt_x$ systems [Fig. 6(a)]. The rise in T_g with Pt concentration is calculated to be 1 K per at.% Pt [Fig. 6(b)]. As there is no complete theory concerning the mean-free-path (MFP) effect of disordered, concentrated alloys, we have refrained from carrying out a MFP correction calculation on the increase of T_g. It should be emphasized that the de Gennes formula for the MFP effect on the average susceptibility $[\chi(R)]_{av}=\chi_0(R)e^{-R/L}$ is valid only for $R \gg L$. Here, L and R denote lattice spacing and MFP, respectively, for these alloys. Thus, the above condition is not expected to hold well for our case of concentrated alloys. However, we believe that the small increase in T_g originates from MFP effects. The lack of a complete theory in this area prevented us from performing a quantitative analysis. MFP effects are discussed in several reports in detail.[5-9]

Recent studies on NiMn alloys have shown that these alloys in the demagnetized state are divided into a number of small and similar domains.[4,10,11] Oner and Aktas have illustrated, by using computer simulation, that the domain structure consists of mostly antiparallel needlelike domain sets.[4] According to this study, the adjacent domains begin to align antiparallel to one other with decreasing temperature in order to reduce the total magnetostatic energy. Therefore, taking a single domain as the basic entity, magnetic ordering will set up in short range in about a few domain-size radiis, while the domain itself will be disor-

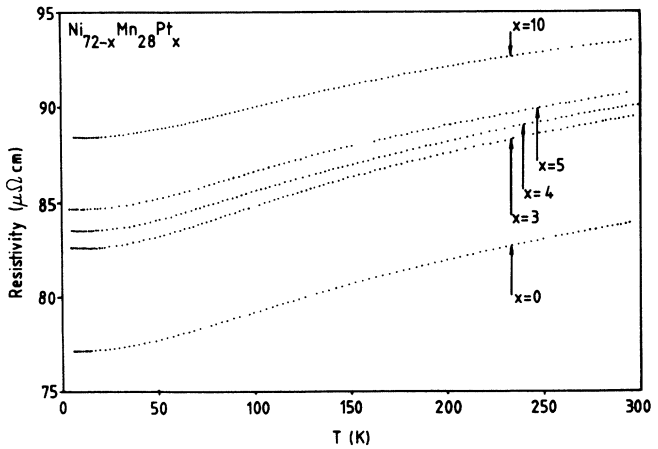

FIG. 3. Temperature variation of resistivity for $Ni_{72-x}Mn_{28}Pt_x$ alloys.

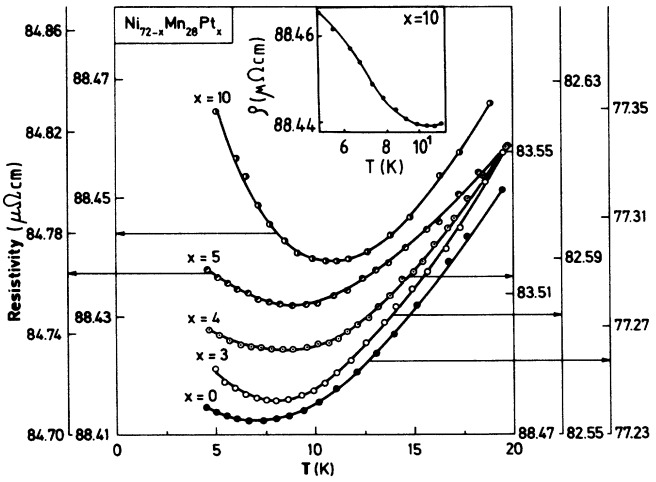

FIG. 5. Low-temperature resistivity data for $Ni_{72-x}Mn_{28}Pt_x$. Inset gives the resistivity of $Ni_{62}Mn_{28}Pt_{10}$ in a semilogarithmic scale.

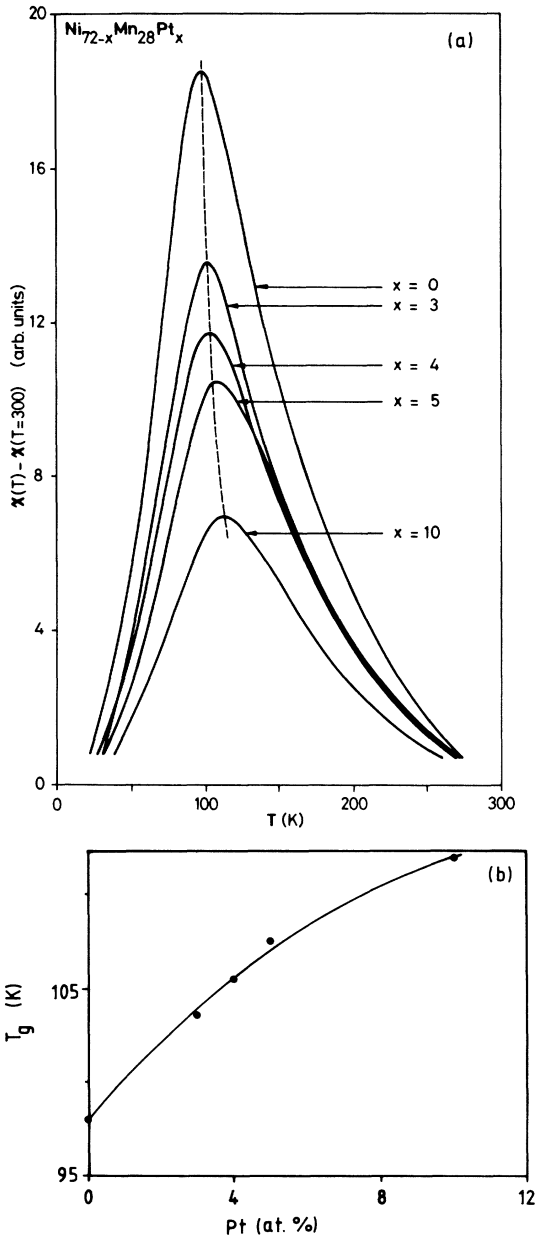

FIG. 6(a). ac susceptibility vs temperature for $Ni_{72-x}Mn_{28}Pt_x$ system. (b) The shift in T_g with impurity concentration variation.

dered as a whole. Domain size is also estimated by taking into account the experimental results obtained on disordered Ni-(24.5-at.% Mn alloys by Goldfarb and Patton.[12] They measured the magnetization of these alloys as a function of both temperature and magnetic field and analyzed their data with the help of the Langevin expression in the superparamagnetic region to deduce the available volume per cluster and concluded that it is 2.6×10^{-20} cm^3, corresponding to a spherical cluster separation of 37 Å. On the other hand, a rough estimate for the MFP of these alloys through the Boltzmann transport equation with an assumption of 0.8 conduction electrons per atom yields about 20–30 Å.[13] In light of the above discussion, it can be concluded that the MFP and the estimated average domain size are close to each other. It appears then that a reasonable explanation of the resistivity slope change at the freezing temperature can be suggested; as MFP is comparable to the domain size, it is obvious that a significant fraction of the total number of conduction electrons will be influenced by the short-range magnetic ordering of the domains that would be set up with decreasing temperature. Consequently, it is reasonable to attribute the slope change in resistivity to short-range domain ordering at lowering temperatures. This process would, of course, be accompanied by the freezing of spins within individual domains.

At first sight, the low-temperature minimum found in the resistivity of NiMn alloys is reminiscent of the Kondo effect which occurs in non-FM crystalline metals with a small amount of FM impurity.[14] However, since magnetic ordering of the local spin destroys the spin-flip freedom, it is unlikely that the Kondo effect can explain this minimum. The resistivity of some metallic glasses gives a minimum which resembles the minimum seen in crystalline Kondo systems. Two-level-tunneling-system models have been used to try to explain this unusual electronic-transport behavior in metallic glasses.[15] This mechanism can also be ruled out safely since, according to the theoretical predictions, presence of a small amount of a magnetic impurity would remove the resistivity singularity.[16] Studies on Ni-Mn alloys made by Senoussi and Oner proved that resistivity does not exhibit any logarithmic behavior.[3] Our experiments also yielded the same result. Thus the suggestion that predicts logarithmic increase of $\rho(T)$ with T does not seem to be a candidate to explain the minima in the NiMn systems.

It should be noted that the zero-field electrical resistance of these alloys at low temperatures is actually magnetoresistance considering the multidomain structure. If we consider the phenomelogical model for an isotropic SG system,[17] we can write the magnetoresistance for one domain in the zero external applied field as

$$\frac{\rho(H_{\rm int})-\rho(0)}{\rho(0)} = -\alpha\langle(\chi_0\mathbf{H}_{\rm int}+\mathbf{M}_r)^2\rangle ,$$

where χ_0 is paramagnetic susceptibility, M_r is the average remanent magnetization over a single domain, α is a constant of proportionality, and $H_{\rm int}$ is the average internal field within the domain. Indeed, the resistivity $\rho(T)$ that we have measured is the $\rho(H_{\rm int})$ in the expression given above. As a result, resistivity increases with decreasing average remanence. According to Saslow and Parker,[18] the neighboring spins will be canted in order to reduce the system's free energy; as the temperature further decreases the canting spreads to more and more spins a greater distance from the frustrated spin. Thus the increasing number of canted spins with decreasing temperature disturb more and more of the magnetic order and the average remanence over the whole domain will decrease and the observed resistivity will increase with decreasing temperature. In summary, there seems to be two competitive effects: a diminishing resistivity due to domain ordering and a rising resistivity due to frozen spins in each domain. We believe that the resistivity minima occur as a result of

these two competing factors. The shift in the resistivity minima towards higher temperatures is a measure of the severity of disordering in individual domains. Indeed, being a superposition of increasing and decreasing components, the resultant resistivity curve would rise and shift to the right as disorder becomes more and more severe. Furthermore, the shifting minima would be more shallow and the resistivity curves would have smaller slopes. This model is consistent with higher resistivity values seen in the shifting shallow minima (Figs. 1, 3, and 5) and lower resistivity slope, dR/dT, observed with added Pt (Figs. 2 and 4). In support of our proposal, adding 2-at.% Pt to NiMn has been reported to lead to a large broadening in the hysterisis cycle without any detectable change on the FM-SG transition.[4]

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