ELECTRICAL RESISTIVITY AND MAGNETORESISTIVITY OF AMORPHOUS $Mn_xPd_{82-x}Ge_{18}$ ALLOYS

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Electrical resistivity and magnetoresistivity measurements of amorphous alloys, obtained by radio frequency (RF) sputtering using argon, depositing onto fused quartz substrates to thicknesses of about 20 microns, were performed. The compositions of these alloys are $Mn_xPd_{82-x}Ge_{18}$ in which x is the atomic concentration and range between 0 to 7. A well-defined minimum in the resistivity versus temperature is observed for these alloys. It is found that resistivity-minimum temperature T_m is linearly proportional to the concentration of Mn and in the temperature range where the resistivity minimum occurs, the nonmagnetic part of the resistivity of the amorphous alloys varies as eT^2 for these alloys. In the $Mn_xPd_{82-x}Ge_{18}$ alloys, the spin-compensated state is masked by the d–d spin correlation which takes place above the Kondo temperature.

1. Introduction

Since the development of techniques for rapidly cooling liquids, a large number of amorphous structures have been discovered. These materials are providing new opportunities for studying the problem of electronic and magnetic states in disordered solids. The inherent interest in such studies is augmented by the fact that disordered materials have potential uses in a variety of technological applications [1].

Ever since the advent of Kondo's theory [2, 3] to explain the resistivity-minimum phenomenon, the problem of an exchange interaction between conduction electrons and localized moments has been of considerable interest. Many have contributed to the present theoretical understanding of the problem of s–d exchange interaction. The experimental results are in general consistent with the theoretical predictions [4]. There remains, however, some uncertainty in understanding the nature of the groundstate condition-electron spins polarized in the vicinity of a magnetic spin. The reported value of the coherence length varies from 9 Å [5] to the order of 10^3 Å [6].

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In this paper, a study of Kondo-type *s–d* exchange interaction in noncrystalline alloys containing transition metals is presented. There are several reasons for the choise of this approach.

First, the results of Kondo's theory are applicable to amorphous materials since the theory does not require a crystal structure. Second, the absence of long range order in amorphous materials probably reduces the *d–d* spin interaction between magnetic atoms [7], which is favorable in the light of the model of isolated magnetic spins. Therefore, amorphous materials offer some advantages over crystalline ones to test the validity of theoretical predictions.

This paper presents the results of electrical resistivity and magnetoresistivity measurements on amorphous $Mn_xPd_{82-x}Ge_{18}$ alloys (where $x = 1$ to 7). The data are analyzed in terms of the existing theories on the *s–d* exchange interaction and the results are compared with those found in crystalline alloys containing Mn.

2. Experimental techniques

Samples of slowly varying composition were synthesized by radio frequency (RF) sputtering using argon depositing onto fused quartz substrates to thicknesses of about 20 microns.

Deposition rates were between 10 and 15 Å/sec. The argon pressure was held at 5×10^{-2} torr. Data reported in this paper are for samples deposited as a power level of 0.3 kW ($\sim$500 Å/min).

The concentration range obtained for $Mn_xPd_{82-x}Ge_{18}$ was 0–7 at % Mn. The purity for Pd and Mn were 99.99 at % and for Ge was 99.9999 at %.

The composition of the samples was determined using electron microprobe. The measurements of the resistivity were done by the standard four-probe method in the temperature range from 4.2 — 300 K. The temperature was measured with an accuracy better than ± 0.3 K by a combination of a copper-constantan thermocouple and germanium crystal thermometer. The results of the resistivity measurements are accurate to $\pm 0.02 \mu\Omega$cm. The specimens used in the magnetoresistivity measurements were the same as those used in the resistivity measurements. The transverse magnetoresistivity was measured at 4.2 K, 77 K and 293 K with the magnetic field varied from 0 to 12 kOe.

3. Results and discussions

A well-defined minimum in the resistivity versus temperature was observed for the Mn-Pd-Ge alloys as shown in Fig. 1. Two characteristics are noticed in this figure: (1) Matthiessen's rule is not satisfied ($\varrho = a + bT + c \ln T$), since the resistivity-temperature coefficient b at higher temperatures changes with Mn concentration. It was found that b is approximately proportional to the inverse of Mn concentration; (2) The resistivity--minimum temperature T_m increases with Mn concentration. Since Matthiessen's rule is not obeyed for these alloys, a simple subtraction of the resistivity of the host $Pd_{82}Ge_{18}$ alloy from that of the $Mn_xPd_{82-x}Ge_{18}$ alloy does not give a meaningful result. Therefore,

it is assumed here that the magnetic part of the resistivity is temperature dependent below T_m while it is constant above T_m. It is further assumed that the nonmagnetic part of the resistivity can be taken as an extrapolation of the resistivity at higher temperatures obeying a T^2 function, to lower temperatures, because the resistivity for the alloys containing

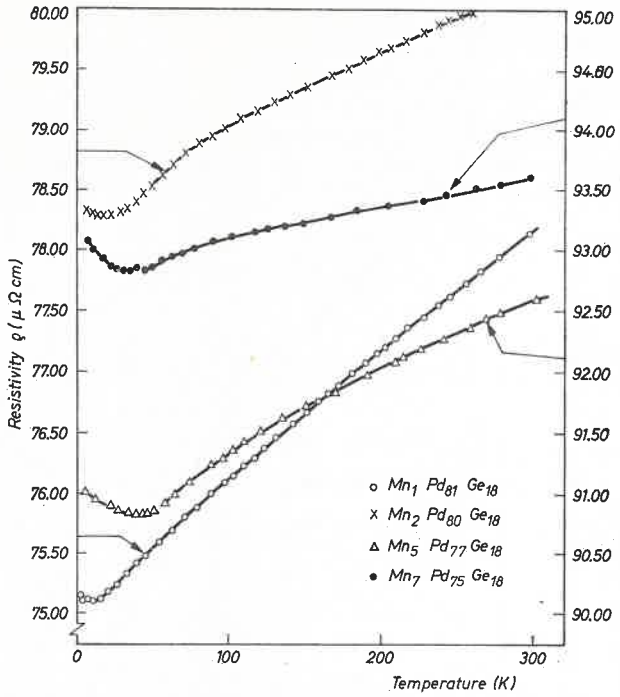

Fig. 1. Electrical resistivity versus temperature for $Mn_xPd_{82-x}Ge_{18}$ alloys. The arrows indicate the scale used

lower concentration of Mn varies as T^2 between T_m and about 80 K. Thus the magnetic part of the resistivity is determined as a difference ($\Delta\varrho$) between the measured resistivity and the nonmagnetic part defined above. From experimental data it is noticed that the temperature variation of $\Delta\varrho$ is independent of Mn concentration and that $\Delta\varrho$ varies as $-\ln T$ between 4.2 K and T_m and becomes temperature independent below 4.2 K. From these data, the resistivity of $Mn_xPd_{82-x}Ge_{18}$ alloys for $T < T_m$ can be described as

$$\varrho = a + eT^2 + c \ln T, \tag{1}$$

where a represents the temperature-independent part of the resistivity, $c \ln T$ represents the temperature-dependent magnetic resistivity part for $T < 70$ K and eT^2 is the nonmagnetic temperature-dependent part for $T_m < T < 70$ K. It was found that coefficient e is inversely proportional to Mn concentration. Equation (1) gives a resistivity-minimum temperature at

$$T_m = \sqrt{(-c/2e)}. \tag{2}$$

From the resistivity data in Fig. 1, resulted $c = -2.5 \times 10^{-2}$ μΩ cm and $e = 3.0 \times \times 10^{-4}$ μΩ cm K^{-2}.

The fact that the resistivity difference $\Delta\varrho$ levels off below about 4.2 K suggests the formation of Nagaoka's quasibound states [8] and *d–d* spin interaction between Mn atoms. The Kondo temperature T_K is probably below 4.2 K for these alloys.

It was found that the $Mn_xPd_{82-x}Ge_{18}$ alloys do not exhibit any observable magnetoresistivity at 77 and 293 K, but an appreciable negative magnetoresistivity was obtained at $T = 4.2$ K. The negative magnetoresistivity $\Delta\varrho_H$ is corrected by subtracting the positive

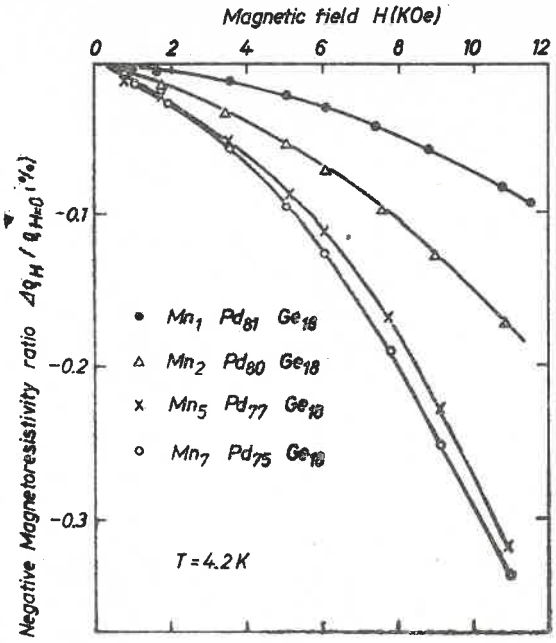

Fig. 2. Negative magnetoresistivity ratio versus magnetic field at 4.2 K for $Mn_xPd_{82-x}Ge_{18}$ alloys

magnetoresistivity of $Pd_{82}Ge_{18}$ from the observed magnetoresistivity and is divided by the resistivity at zero field, $\varrho_{H=0}$. The corrected negative magnetoresistivity at $T = 4.2$ K, shown in Fig. 2 for Mn concentrations of 1, 2, 5 and 7 at %, is found to vary as $-b\sigma_a^n$ with band n given in Table I.

TABLE I

Values of b and n at $T = 4.2$ K in the expression $\Delta\varrho_H/\varrho_{H=0} = -b_a^n$ for the $Mn_xPd_{82-x}Ge_{18}$ alloys

Alloy composition	b (%μ_B^n)	n
$Mn_1Pd_{81}Ge_{18}$	0.0037	2.00
$Mn_2Pd_{80}Ge_{18}$	0.0185	1.98
$Mn_5Pd_{77}Ge_{18}$	0.0800	1.87
$Mn_7Pd_{75}Ge_{18}$	0.1210	1.70

The fact that $n = 2$ implies that the quasibound states are not fully formed at 4.2 K. To study the concentration dependence of the negative magnetoresistivity, $\Delta\varrho_H = 9\ \mathrm{KOe}/\varrho_{H=0}$ at 4.2 K is plotted against Mn concentration in Fig. 3. It is noticed that the absolute value of the negative magnetoresistivity increases with Mn concentration for $x \leqslant 5$ and decreases for $x > 5$.

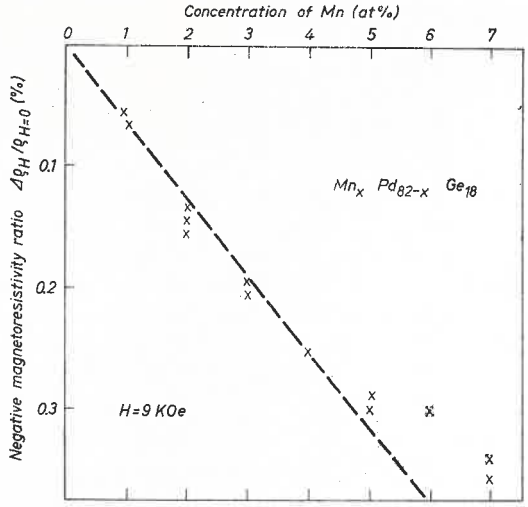

Fig. 3. Negative magnetoresistivity ratio versus Mn concentration at $H = 9$ kOe and $T = 4.2$ K for the $Mn_xPd_{82-x}Ge_{18}$ alloys

The striking result is that the concentration at which $|\Delta\varrho_H/\varrho_{H=0}|$ is a maximum very large i. e. about 5 at % when $T = 4.2$ K and $H = 9$ kOe. This is compared with the concentration of the order of less than 0.1 at % in crystalline alloys [9] in which similar maximum has been observed. These facts indicate that the d–d spin interaction between transition-metal atoms is weaker in amorphous alloys than in crystalline alloys.

These results demonstrate that the negative magnetoresistivity is due to the s–d exchange interaction. Using the result of Kondo [2, 3] ($c = 3p \times J_{sd}\varrho_m/100\,E_F$, where p is the number of electrons per atom, n the number of atoms per unit volume and E_F the Fermi energy), taking $c = -0.025 \times \mu\Omega$ cm, $S = 4.8/2$ and $E_F = 3.55$ eV, it was found $J_{sd} \simeq -0.17$ eV for Mn–Pd–Ge alloys, comparable, with $J_{sd} = -0.4$ eV for a dilute crystalline Cu–Mn alloys [11] for which $S = 5/2$.

4. *Conclusions*

The electrical resistivity and magnetoresistivity of amorphous palladium-germanium base alloys containing Mn have been studied. A well-defined minimum in the resistivity--versus-temperature curve is observed in these alloys. It is found that T_m is linearly proportional to the concentration of Mn. This is different from the case of crystalline alloys where

T_m varies as the 1/5 power of the transition-metal concentration. This difference originates from the fact that, in the temperature range where the resistivity minimum occurs, the nonmagnetic part of the resistivity of the amorphous alloys varies as eT^2 for the Mn alloys. These results indicate that the theories based on the isolated-spin assumption are applicable to these nondilute amorphous alloys.

In the $Mn_xPd_{82-x}Ge_{18}$ alloys, the spin-compensated state is masked by the *d–d* spin correlation which takes place above the Kondo temperature.

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