Electrical resistivity of PdMn alloys: the transition from ferromagnetism to spin glass

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Results are given of measurements of the electrical resistivity of PdMn alloys (0.81 to 15 at.% Mn) in the temperature range from 1.8 to 20 K, where the transition is observed from the ferromagnetic state ($c < 5$ at.% Mn) to a spin glass ($c > 6$ at.% Mn). It was established that at low temperatures $\rho(c,T) = A(c)T^{3/2}$. The coefficient $A(c)$ decreases sharply with increasing concentration in the ferromagnetic phase and is almost constant in the spin-glass state. The temperature coefficient of the resistivity for alloys with 5 to 6 at.% Mn has two maxima. This indicates the possible existence of two phases, ferromagnetic and spin glass, for alloys of this composition.

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A broad spectrum of magnetic states is realized in Pd-rich PdMn alloys. For Pd concentrations less than 600 ppm this is the classic spin glass with the characteristic behavior of the magnetic properties.[1] In the concentration region below 4 at.% Mn the ferromagnetic state is realized,[2] while in the region from 5 to 10 at.% Mn, PdMn alloys exhibit the characteristic properties of spin glasses.[3-5] The concentration region 4 to 5 at.% Mn is the transition region and can be considered as a region of coexistence of two phases, ferromagnetic and spin glass.[6]

Such a complicated magnetic behavior of these alloys within a relatively narrow range of concentration is caused by the nature of the interaction of Mn atoms in Pd. In dilute PdMn alloys the Mn atoms possess giant magnetic moments produced by the strong polarization of the matrix atoms. The RKKY interaction between these moments leads to the spin glass state.[1] A correlation arises between these moments with increasing Mn concentration, leading to the establishment of ferromagnetic ordering.[2] In these alloys a short-range antiferromagnetic Mn–Mn interaction exists as well as the long-range ferromagnetic Mn–Mn interaction. As was suggested by Moriya[7] and established by Star *et al.*[8] in studies of the magnetic properties of PdMn alloys with Mn concentrations less than 2.5%, the antiferromagnetic Mn-Mn interaction extends to the third coordination sphere and this interaction is appreciable even for low concentrations of ~1 at.% Mn. The existence of two types of interaction, the short-range antiferromagnetic and long-range ferromagnetic, determines the spin glass state at 5 to 10 at.% Mn. Such an exchange interaction which leads to the mentioned spectrum of magnetic states, produces an appreciable effect on transport phenomena. Processes of scattering by spin excitation of the magnetic system become appreciable and determine the concentration and temperature dependences of the kinetic coefficients. An examination of the kinetic properties can therefore be used to obtain the magnetic characteristics of the alloys, not only in the temperature region of the magnetic transition, but also at low temperatures where low-frequency spin excitations become dominant.

In the present work the electrical resistivity $\rho(c, T)$ of PdMn alloys with Mn concentrations from 0.8 to 15 at.% are studied in order to reveal the features of $\rho(c, T)$ which are produced by the magnetic state and their changes on going from the ferromagnetic to the spin-glass state.

METHOD OF MEASUREMENT AND REDUCTION OF THE EXPERIMENTAL RESULTS

Alloys with Mn concentrations 0.81; 1.25; 1.65; 2.1; 2.6; 3.0; 3.75; 5; 6; 7; 8; 9; 10; 15 at.% were studied. The electrical resistance was measured by a standard method[9] in the range from 1.8 to 300 K. The temperature was measured with platinum and germanium thermometers. The results were reduced by least squares with a computer to determine the temperature dependence of $\rho(c, T)$ and the features of the temperature coefficient of resistance (tcr). Since the low-temperature region where the magnetically ordered states occur is of greatest interest, the results of the measurements of $\rho(c, T)$ in the temperature range from 1.8 to 20 K are presented here.

In analyzing the experimental results we consider the temperature dependent part of the resistivity $\rho(c, T) = \rho_{\text{tot}}(c, T) - \rho(c, 0)$, where $\rho_{\text{tot}}(c, T)$ and $\rho(c, 0)$ are the total and residual resistivities of the alloys, and not the so-called impurity part $\Delta\rho(c, T) = \rho_{\text{tot}}(c, T) - \rho(c, 0) - \rho_{\text{Pd}}(T)$, where $\rho_{\text{Pd}}(T)$ is the resistivity of the palladium matrix. It is incorrect to regard $\Delta\rho(c, T)$ in this form as the additional contribution to the resistivity of the matrix due only to the magnetic scattering mechanism, since account is not taken of departures from Matthiessen's rule caused by changes in the non-magnetic scattering mechanisms. Such changes can be important and even dominant and lead to non-monotonic $\Delta\rho(c, T)$ dependences with an extremum at a temperature which is not a characteristic of the system.[10] In addition, the procedure for subtracting the matrix resistivity from the alloy resistivity presupposes that the perturbation of the matrix on alloying is insignificant and the alloy is in the same magnetic state as the matrix. In the opposite case, when the alloy is in a different magnetic state from the matrix, the procedure for obtaining $\Delta\rho(c, T)$ loses its physical meaning and its analysis can lead to a contradiction, as occurred in the consideration of the

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electrical resistivity of PdNi alloys.[9] These reasons prompted us to analyze the magnitude of $\rho(c, T)$ and its derivatives $\partial\rho(c, T)/\partial T$ and $\partial^2\rho(c, T)/\partial T^2$ when discussing the resistivity of PdMn alloys.

RESULTS OF MEASUREMENTS AND DISCUSSION OF THEM

Figure 1 shows curves of the temperature dependences of resistivity and of the tcr of the alloys studied in the temperature range from 1.86 to 20.4 K. It can be seen that a sharp break is observed in the $\rho(c, T)$ curves for alloys with Mn concentrations up to 4 at.% at a temperature corresponding to the Curie temperature, and this is smeared out and disappears as the concentration increases. Measurements of the concentration dependence of the Curie temperature, obtained by us, are shown in Fig. 2. It follows from this figure that the maximum value of Θ is reached in the region of 3.5 at.% Mn, after which a fall in Curie temperature is observed, due to the weakening of the ferromagnetic interaction.

We shall consider the low-temperature behavior of the resistivity. The temperature and concentration dependences of $\rho(c, T)$ were determined to elucidate the scattering mechanisms in the region $T < 5$ to 6 K for non-ferromagnetic and $T < (0.3$ to $0.4)\Theta$ for ferromagnetic alloys. For this purpose $\rho(c, T)$ was approximated by two terms $A(c)T^{3/2} + B(c)T^2$. It was found that the quadratic term is negligibly small and $\rho(c, T)$ is well described by the term $A(c)T^{3/2}$ and a deviation from it is observed for $T > 5$ to 6 K. The coefficient $A(c)$ decreases strongly with increasing concentration and in the range from 6 to 10 at.% Mn remains almost constant, equal to $(36 \pm 2) \times 10^{-10}\ \Omega\cdot\text{cm}\cdot\text{K}^{-3/2}$ and increases slightly only in the alloys with 10 and 15 at.% Mn.

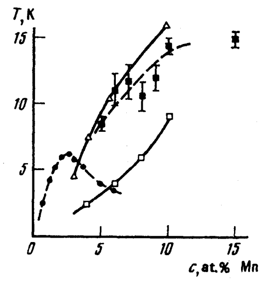

FIG. 2. Concentration dependence of Curie temperature and freezing temperature of PdMn alloys: ●) Θ, ■) T_f are obtained in the present work; □) T_f obtained from susceptibility[5]; Δ) T_f obtained from specific heat.[4]

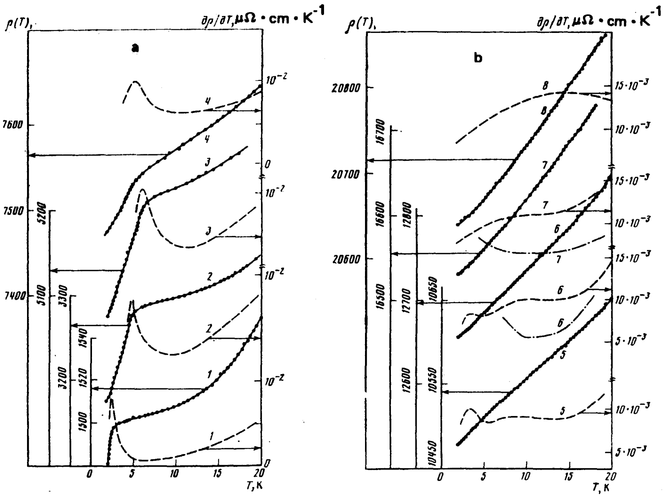

FIG. 1. Temperature dependence of $10^3\ \rho(T)$, $\mu\Omega\cdot$cm (solid lines) and of the tcr (dashed lines) for PdMn alloys: 1) 0.81; 2) 1.65; 3) 2.6; 4) 3.75; 5) 5; 6) 6; 7) 8; 8) 10 at. % Mn [the dash-dot lines are $\partial^2\rho(c, T)/\partial T^2$ in arbitrary units].

The $\rho(c, T)$ dependence can be connected with a magnetic scattering mechanism on the assumption that electron-phonon scattering processes are ineffective in this temperature region and the electron-electron collision make a small contribution to the resistivity [the absence of a term in $\rho(c, T)$ quadratic in temperature is evidence of this].

It is known that in disordered ferromagnetic alloys, elastic scattering by randomly distributed magnetic impurities and inelastic scattering by spin-waves with quasimomentum nonconservation lead to a dependence $\rho \propto (T/D)^{3/2}$, where $D(c)$ is the coefficient of stiffness of the spin-wave dispersion law.[11] In this case the concentration dependence of the coefficient $A(c)$ will be determined by the concentration dependence of $D(c)$. It was shown theoretically that for dilute alloys of the PdFe type, $D(c)$ varies exponentially with increasing concentration and subsequently goes over into a dependence $\alpha + \beta c$ (Refs. 12, 13). The increase in $D(c)$ with increasing concentration was obtained experimentally from an analysis of magnetic measurements on PdMn alloys.[8] Since the spin-wave density decreases with increasing $D(c)$ ($g(\varepsilon) \sim \varepsilon^{1/2}/D^{3/2}$), the corresponding contribution to the resistivity decreases, and this is reflected in a decrease in the coefficient $A(c)$.

One can understand the observed temperature dependence of $\rho(c, T)$ for PdMn alloys in the spin-glass state if it is assumed that spin excitations with a quadratic dispersion law exist in them.[1)] As in the consideration of the resistivity of ferromagnetic alloys, the density of spin excitations will then be proportional to $\varepsilon^{1/2}/D^{3/2}$, which leads for isotropic scattering to a $T^{3/2}$ law for electrical resistivity.

There is a linear dispersion law for elementary spin excitations in spin glasses[15,16] and scattering by them leads to a dependence[17] $\rho(T) \sim T^4$. It has been pointed out,[15] however, that there is a possibility of spin-excitation branches existing with a quadratic dispersion law in spin glasses with a small remanent magnetization.

It was shown in a series of studies by Thomson and Thompson[1,18] on classical spin glasses of the type CuMn, AgMn, AuMn, PdMn at very low Mn concentrations that both the magnetization and the magnetic part of the specific heat are proportional to $T^{3/2}$. The authors maintained as a result of this that magnetic excitations with energy $\varepsilon \sim K^2$ were dominant in the low-temperature region. Numerical calculation and analysis of neutron data on the $\text{Eu}_x\text{Se}_{1-x}\text{S}$ system, which is analogous to PdMn alloys in that the spin-glass state (at $x = 0.4$; 0.5; 0.6) is due to existence of two types of Eu-Eu magnetic interaction, short-range ferromagnetic and long-range antiferromagnetic, also showed that the elementary magnetic excitations of this system have a quadratic dispersion law. We may, therefore, assume that elementary spin excitations with a quadratic dispersion relation exist in the PdMn spin glass studied by us, and that scattering by them gives the observed $T^{3/2}$ dependence of the electrical resistivity. The concentration dependence of $\rho(c, T)$ will then be determined by the concentration dependence of $D(c)$.

Before going on to further consideration of the resistivity we note that together with spin waves there exist in spin glasses diffusive spin modes, scattering by which can give an appreciable contrubution to the resistivity. Rivier and Adkins[20] showed that $\rho \sim T^{3/2}$ for such scattering processes, but they did not take account of conservation of the total spin. Fischer[17] obtained a more complicated temperature dependence of $\rho(c, T)$, differing from $T^{3/2}$, by taking account of conservation of total spin [in the simplest case $\rho(c, T)$ is given by a four-term expression (Ref. 17, Eq. (50)].

Comparing the dependence $\rho(c, T) \sim T^{3/2}$ obtained by us with Fischer's results[17], we conclude that scattering by diffusive spin modes does not determine the electrical resistivity of PdMn alloys.[2)]

In the simplest case $A(c)$ will be proportional to $cD^{-3/2}(c)$, so that the concentration dependence of $D(c)$ can be estimated. Calculations showed that $D(c)$ increases with increasing concentration. The freezing temperature T_f also increases with increasing concentration, i.e., there is a correlation between these quanquantities. A similar situation is found for ferromagnetics when $D \sim \Theta$ (Ref. 22). It should be noted that a similar picture exists in spin glasses of the AuFe and AgFe type,[23] when the freezing temperature increases with increasing diffusion coefficient. Neutron scattering studies on $Fe_{1-x}Cr_x$ spin glasses[24] also indicate the existence of such a correlation. It can therefore be assumed that this correlation in spin glasses does not arise by chance but is of a deeper nature, indicating a relation between the low-frequency magnetic excitations and the temperature of the magnetic transition, as occurs in other magnetically ordered systems.

The increase in the magnetic stiffness with concentration can be explained by the behavior of the susceptibility in PdMn alloys, which decreases as the Mn concentration rises from 6 to 10 at.%.[5] Zweers *et al.*[5] attribute this to the presence of clusters and to their change with concentration, but we assume that this may be due to an increase in $D(c)$, as occurs for $Fe_{1-x}Cr_x$ spin glasses[24] or for ferromagnetics on approaching Θ, when a fall in $D(c)$ produces an increase in susceptibility. The presence of spin excitations with a quadratic dispersion law also contributes to the magnetic part of the specific heat.[4] One can then understand the deviation of the low-temperature heat capacity of PdMn alloys[4] from a linear temperature dependence in the direction of higher powers of T, and also its decrease with increasing concentration, which can be explained by an increase in $D(c)$.

We studied the temperature coefficient of resistivity in order to determine the behavior of electrical resistivity in the region of freezing temperature. We differentiated the $\rho(c, T)$ curves to obtain it. In addition, we differentiated twice with respect to temperature to sort out the temperatures at which anomalies in $\rho(c, T)$ or in the tcr are observed. In analyzing the $\rho(c, T)$ function obtained and its temperature derivatives we assumed that their singularities were only brought about by magnetic scattering mechanisms and considered that electron-phonon scattering processes do not give resistivity anomalies. This is confirmed by the absence of anomalies for pure Pd and for PdMn alloys with $c < 4$ at.% Mn in the range from 7 to 20 K (Fig. 1a). The singularities in the $\rho(c, T)$ curves were revealed by the presence of a maximum of inflexion in the tcr; in the latter case the second derivative of $\rho(c, T)$ was tested for a minimum and the temperature of the minimum was taken as the temperature of the anomaly.[3),4)]

We shall consider the temperature dependence of the tcr for PdMn alloys (Fig. 1). We can distinguish between three concentration ranges.

There are sharp maxima in the $\partial\rho(c, T)/\partial T$ curves for alloys with 0.81 to 3.75 at.% Mn, due to the transition from the paramagnetic to the ferromagnetic state at the Curie temperature. As the concentration increases the values of the maximum decrease and it is smeared. The existence of a smeared maximum or an inflexion in the tcr is a characteristic of alloys with 7 to 10 at.% Mn. Two maxima in the tcr are observed for alloys with the intermediate concentrations 5 to 6 at.% Mn; the first, at low temperature, is relatively sharp and the second, at higher temperatures, is smeared.

The presence of a maximum in the $\partial\rho(c, T)/\partial T$ curve indicates that scattering processes exist in the system and undergo at the temperature of the maximum qualitative changes due to a change in the magnetic state. This occurs in ferromagnetics,[25] in Kondo systems,[26] and in systems with localized spin fluctuations,[27] in which the temperature of the inflexion in the $\rho(c, T)$ curves coincides with the Curie temperature, with the Kondo temperature, and with $T_{\text{sp.fl.}}$. It may be supposed that in the systems studied by us, the conduction-electron scattering by spin excitations, which is responsible for the low-temperature behavior of the resistivity, undergoes at a certain temperature changes due to magnetic disordering. While for ferromagnetic PdMn alloys the temperature of the $\partial\rho(c, T)/\partial T$ maximum coincides with the Curie temperature obtained from magnetic measurements, for PdMn spin glasses it is a few degrees (~5 K) above the freezing temperature obtained from susceptibility measurements and is close to the temperature of the maximum of the magnetic part of the specific heat (Fig. 2). This agreement of the temperature of the $\partial\rho(c, T)/\partial T$ maximum and of the magnetic part of the specific heat points to an important role of spin excitations at $T > T_f$ in these materials, while the temperature of the maximum together with T_f can be a characteristic temperature for a spin glass.

Finally, we look at the 5 to 6 at.% Mn concentration range which is the transition region from the ferromagnetic to the spin-glass state. It was predicted on the basis of magnetic measurements[6] that alloys in this concentration range comprise two magnetic phases: one corresponds to the ferromagnetic and the other to the spin-glass state. It can be seen from the tcr of alloys of this concentration range (Fig. 1) that the $\partial\rho(c,T)/\partial T$ curves have two maxima which can be identified with the two magnetic phases: the "low-temperature" one with the ferromagnetic phase and the "high-temperature" one with the spin-glass phase. Such an identification agrees with the behavior of tcr of PdMn alloys in the single-phase state, when only one anomaly exists in the tcr curves.

It should be noted that two maxima were observed in the $\partial\rho(c,T)/\partial T$ curves for dilute PdFe alloys ($c \approx 0.1$ to 1 at.% Fe). The first (sharply pronounced) occurred at the Curie temperature, and the second (smeared) at a higher temperature (in appearance it was similar to the maximum in tcr of PdMn alloys with $c > 4$ at.%). The observed behavior of $\partial\rho(c,T)/\partial T$ in PdFe alloys were explained[28] as a result of scattering of conduction electrons by a spin system in which at $T > \Theta$ there exist both short-range magnetic order and a long-range order of the fluctuations in the short-range magnetic order. In the opinion of Kawatra *et al.*[28] the change in concentration leads then to a change in the inter-cluster interaction and produces a change in the ratio of these contributions. This explanation is in principle a particular case of one more general, that relaxation is produced in disordered magnetic systems electron both by local scattering effects determined by short-range order, and by collective magnetic excitations determined by long-range order. The ensuing singularities of the kinetic properties are a reflection of qualitative changes in the magnetic structure.

Comparison of the results of our studies of the electrical resistivity of PdMn alloys with studies of $\rho(c,T)$ of PdFe alloys makes it possible to suggest that in the latter a magnetic state of the type of a spin glass can be realized at $T > \Theta$ and is reflected in the bahevior of the tcr.

CONCLUSION

The analysis of the electrical resistivity of PdMn alloys which undergo a transition from a ferromagnetic to a spin glass state with increasing Mn concentration thus showed that the main scattering mechanisms in the region $T < 5$ K (for spin glasses) and $T < 0.3\,\Theta$ (for ferromagnets) are scattering by spin excitations with a quadratic dispersions law, leading to the relation $\rho(c,T) = A(c)T^{3/2}$. A characteristic feature of the resistivity of spin glasses is then that the concentration is independent of $A(c)$.

Consideration of $\rho(c,T)$ and the tcr in the temperature range from 2 to 20 K revealed anomalies in these properties [a maximum or inflection in the $\partial\rho(c,T)/\partial T$ curves], which can be due to a magnetic transition as the temperature changes. While the temperature of the anomaly in $\partial\rho(c,T)/\partial T$ coincides with the Curie temperature for ferromagnetic PdMn alloys, it lies above the freezing temperature of PdMn spin glasses and close to the temperature of the maximum magnetic contribution to the specific heat.

Two maxima of the tcr exist for alloys with 5 to 6 at.% Mn. Taking account of the magnetic data, it can be assumed that this region is one of coexistence of two phases, ferromagnetic and spin glass.

1) The density of low-temperature spin excitations and the dynamic structure factor for Pd + 10 at. % Mn spin glass were calculated by Ching *et al.*[14] The good agreement with the experimental results on the low-temperature part of the specific heat and with the neutron scattering data enable one to consider that it is spin excitations (spin waves in that study) which have determined these properties.

2) Such a comparison can be valid when diffusive spin modes can exist in concentrated spin glasses of the PdMn type, analogous to those discussed by Fischer.[17] The results of Nieuwenhuys and Verbeek[21] can serve as indirect confirmation of such a possibility for PdMn alloys.

3) The absence of a maximum in the $\partial\rho(c,T)/\partial T$ curves for some alloys is connected with the existence of a strong temperature dependence of the tcr, due for example to electron-phonon scattering processes. Either a maximum or an inflection can be observed in the resulting $\partial\rho(c,T)/\partial T$ curves, depending on the relation between $\partial\rho(c,T)/\partial T|_{\text{phon}}$ and $\partial\rho(c,T)/\partial T|_{\text{mag}}$, and the existence of $\partial\rho(c,T)/\partial T|_{\text{phon}} > 0$ makes the observed temperature of the anomalies in $\partial\rho(c,T)/\partial T$ lower than the temperature of the anomalies connected with ρ_{mag}.

4) Ford and Mydosh[23] noted that $\partial^2\Delta\rho(c,T)/\partial T^2$ has a minimum in the region of the freezing temperature. In this case the procedure for obtaining the minimum of $\partial^2\Delta\rho(c,T)/\partial T^2$ leads to the same result as proposed by us, since in the absence of $\rho(T)$ anomalies in the matrix in this temperature region singularities in $\partial^2\rho(c,T)/\partial T^2$ and $\partial^2\Delta\rho(c,T)/\partial T^2$ should occur at one and the same temperature.

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