Transport properties and localized spin fluctuations in *Pt*Ni alloys

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Measurements of the thermopower of *Pt*Ni alloys with Ni concentrations from 0.33 to 4 at.% show a common linear increase with temperature at low temperatures. There is no evidence of thermopower peaks due to scattering from localized spin fluctuations (in contrast to *Pd*Ni alloys). This is consistent with resistivity measurements which suggest, in an analysis that includes large conventional deviations from Matthiessen's rule, that the localized-spin-fluctuation temperature is in the range 70 to 160 K.

I. INTRODUCTION

Spin fluctuations play a crucial role in our understanding of many of the properties of nearly and weakly magnetic systems, and of ordered magnets above their ordering temperatures.[1] Particularly noticeable are effects on electronic transport properties, reviewed for crystalline materials in Ref. 2. Recently, dramatic spin-fluctuation effects have also been seen in the resistivity of Fe-Zr glassy metals at concentrations near the ferromagnetic transition.[3]

Scattering of conduction electrons by spin fluctuations generally produces a characteristic increase in resistivity with temperature T as the population of spin fluctuations increases, initially[4,5] as T^2 and then[6] as T. (An exception[6] is the case of alloys in which well-defined virtual bound states form at magnetic impurity sites, leading to the Kondo effect in which the resistivity *decreases* as T increases.) Nearly magnetic impurities also produce giant peaks in the diffusion thermopower in the presence of both localized spin fluctuations (LSF) and a nonmagnetic impurity potential.[7] This peak scales approximately with the spin-fluctuation temperature T_s, the temperature corresponding to the peak of the spectral density of the LSF.[8]

Classic examples of spin-fluctuation systems are[4,9,10] Pd and dilute *Pd*Ni alloys. Mackliet, Schindler, and Gillespie[11] demonstrated that LSF effects also occur in the low-temperature resistivity and specific heat of dilute *Pt*Ni alloys. To investigate further the *Pt*Ni system, we have made measurements of thermopower and have extended the resistivity data to higher temperatures.

II. THERMOPOWER

We have made thermopower measurements on the *Pt*Ni alloy samples and the pure Pt sample of Mackliet, Schindler, and Gillespie[11] at temperatures in the range 1.5—120 K. The major difference between the thermopowers of the alloys and pure Pt occurs at low temperature where they have opposite sign, as shown in Fig. 1.

A metallic thermopower $S(T)$ can be written as the sum of a diffusion term $S_d(T)$ and a phonon drag term $S_g(T)$ as

$$S(T)=S_d(T)+S_g(T)\ . \tag{1}$$

The diffusion thermopower is given by Kohler's rule[12] for the *Pt*Ni alloys as

$$S_d(T)=\frac{W_{\rm Ni}(T)S_{\rm Ni}(T)}{W(T)}+\frac{W_{\rm ph}(T)S_{\rm ph}(T)}{W(T)}\ , \tag{2}$$

where $S_{\rm Ni}$ and $W_{\rm Ni}$ are the characteristic thermopower and thermal resistivity, respectively, for scattering by the Ni impurities, $S_{\rm ph}$ and $W_{\rm ph}$ are the corresponding quantities for scattering by phonons, and W is the total thermal resistivity.

Recently, analysis of measurements of the diffusion thermopower of glassy metals has confirmed that the electron-phonon enhancement is present,[13] and that there is also some evidence for velocity and relaxation-time renormalization and/or Neilsen-Taylor effects.[14] The behavior of glassy metals suggests that the total contribution of these virtual phonon effects to the absolute thermopower is significant but not large.[13,14]

As the temperature tends to zero, scattering by Ni impurities will dominate and the diffusion thermopower

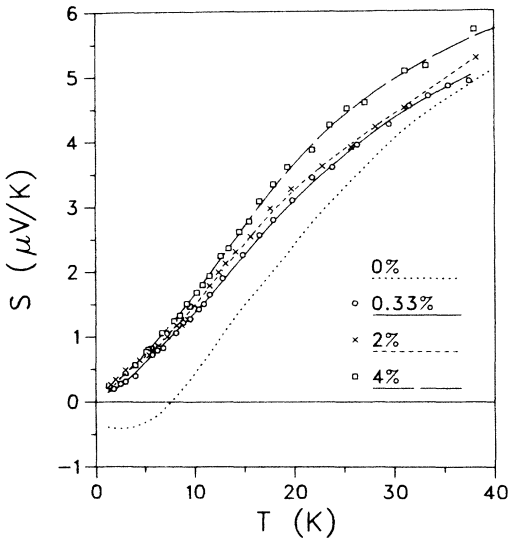

FIG. 1. Thermopower of three representative dilute *Pt*Ni alloys and Pt. The percentage figures give the Ni concentration in atomic percent.

should be simply S_{Ni}, giving a total thermopower independent of Ni concentration in the absence of large variations in drag or Neilsen-Taylor effects. This expectation is clearly verified in Fig. 1. We obtain a value for S_{Ni}/T, including virtual phonon effects, of order 0.1 $\mu V/K^2$ in the low-T limit, assuming $S_g(T)$ goes to zero faster than $S_d(T)$ as $T \rightarrow 0$. This is larger in magnitude than that for Ir and Au impurities in Pt,[15] but smaller than the value of about -1 $\mu V/K^2$ for Ni impurities in Pd.[9]

A large linear term (of either sign) is in fact predicted for the thermopower due to LSF's at low temperatures, followed by a peak at higher temperatures.[7] Such a peak is clearly seen[9] at 18.5 K in *Pd*Ni, but while S_{Ni} for *Pt*Ni *may* show a positive peak above 20 K, we are unable to verify this owing to the presence of the large phonon drag peak. There is also the complication that as the scattering of electrons by phonons increases, a pronounced nonlinear T dependence is expected in the diffusion thermopower $S_d(T)$ alone as it changes from S_{Ni} towards S_{ph} (about $-0.017T$ $\mu V/K$) (Ref. 16) as the balance of scattering changes in Eq. (3), even if S_{Ni} and S_{ph} are linear in T. Yet another complication is the expected presence of temperature-dependent enhancement and Nielsen-Taylor effects.

Turning briefly to the thermopower of "pure" Pt, our measurements agree with those of Fletcher and Greig above 20 K, but show a negative plateau of somewhat larger magnitude at very low temperatures. This observed variation in magnitude for different samples would be expected if the negative peak is due to trace Fe impurities, as suggested by MacDonald, Pearson, and Templeton.[17] This explanation of the Pt thermopower minimum is supported by the fact that it is absent in measurements on Pt films,[18] in which disorder scattering dominates over scattering from trace impurities (thus weighting out the thermopower contribution of the Fe impurities according to Kohler's rule).

III. RESISTIVITY

Mackliet, Schindler, and Gillespie[11] made detailed measurements of the resistivity of several dilute *Pt*Ni alloys from 1.2 to 4.2 K, showing how the coefficient of the T^2 spin-fluctuation resistivity term increases from 1.54×10^{-5} $\mu\Omega\,cm/K^2$ in pure Pt to 3.27×10^{-5} $\mu\Omega\,cm/K^2$ in the 4 at. % Ni alloy. We have extended these measurements to higher temperatures for the pure Pt and the 4 at. % Ni alloy (the alloy with the largest LSF resistivity term of those available). The measurements were made simultaneously in the same cryostat to minimize the error in determining the resistivity difference. The observed difference $\Delta\rho(T)$ in the resistivities of the two samples (alloy resistivity minus pure Pt resistivity) is shown in Fig. 2. The uncertainty in the ratio of the form factors for the samples (about 2%) leads a relatively large uncertainty in $\Delta\rho(T)$, which produces a large uncertainty in the slope of the temperature dependence of $\Delta\rho(T)$ at higher temperatures, as indicated by the dotted lines in Fig. 2.

We take the resistivity $\rho_{LSF}(T)$ due to scattering by LSF's as given by the universal curve of Kaiser and Doniach.[5]

$$\rho_{LSF}(T)=(3T_s^2\Delta A/\pi^2)[\pi T/2T_s-\tfrac{1}{2}+(T_s/4\pi T)\psi'(1+T_s/2\pi T)]\,, \tag{3}$$

where ΔA is the increase in the low-temperature T^2 resistivity coefficient produced by LSF's and $\psi'(x)$ is the trigamma function. We also expect a contribution to the temperature-dependent resistivity in the alloy from "conventional" deviations from Matthiessen's rule (DMR) associated with the variation over the Fermi surface of the electron-phonon scattering rate. To describe this term, $\Delta(T)$, we use the theoretical two-band expression[19] with the phonon resistivity varying as T^5. As used by Kaiser[2] for *Pd*Ni alloys, this expression can be written as

$$\Delta(T)=\Delta_0/[1+(T_0/T)^5]\,, \tag{4}$$

where Δ_0 is the saturation value of $\Delta(T)$ at high temperatures, and T_0 is the temperature at which $\Delta(T)$ equals $\Delta_0/2$. This expression gives a very good description of DMR for Ti and V impurities in Pt, as measured by Azarbar and Williams,[20] although these authors gave a more sophisticated analysis taking account of the change from T^5 to T phonon resistivity at higher temperatures and allowing the two-band model parameters to be temperature dependent. For our present purpose, we suggest our simple and general expression (4) is appropriate to describe the expected shape of $\Delta(T)$ for Pt-based alloys.

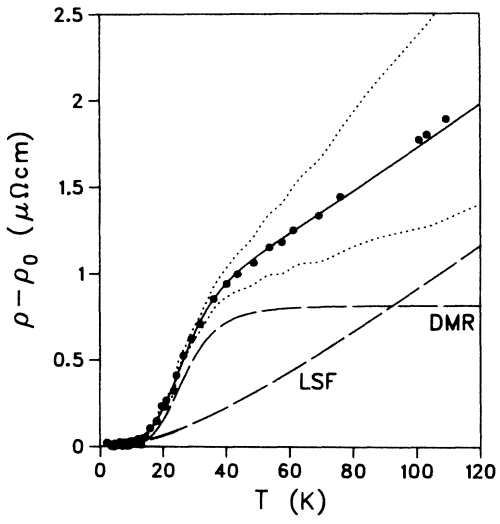

FIG. 2. Increase $\Delta\rho(T)$ in the resistivity of the *Pt*Ni alloy with 4 at. % Ni relative to that in pure Pt, with the increase $\Delta\rho_0$ in residual resistivity subtracted off to leave only the change in temperature-dependent resistivity. Our experimental data are shown by dots, with upper and lower limits given by the dotted lines. The full line is a fit of Eqs. (3)–(5) to the data with parameter values $\Delta\rho_0=3.26$ $\mu\Omega\,cm$, $\Delta A=1.8\times10^{-5}$ $\mu\Omega\,cm/K^2$, $T_s=160$ K, $\Delta_0=0.082$ $\mu\Omega\,cm$, and $T_0=27$ K. The dashed lines give the separation of the fitted resistivity into LSF and conventional DMR components.

Empirically,[20] Δ_0 (and also T_0 to a slight extent) increase with impurity concentration, in qualitative agreement with the predictions of the two-band model.

Our fitting equation is therefore

$$\Delta\rho(T)=\Delta\rho_0+\rho_{\mathrm{LSF}}(T)+\Delta(T)\ , \tag{5}$$

where $\Delta\rho_0$ is the increase in residual resistivity in the *Pt*Ni alloy. As shown in Fig. 2, we obtain a very good fit for $\Delta\rho(T)$. The size of the DMR term ($\Delta_0\sim 0.08\ \mu\Omega\,\mathrm{cm}$) is rather less than half that for *Pt*Ti and *Pt*V alloys of similar residual resistivity, while the temperature scale ($T_0=27$ K) is the same as that for *Pt*Ti and *Pt*V alloys with a similar size of DMR.

The increase, ΔA, in the coefficient of the LSF T^2 resistivity term (largely determined by the data below 15 K) is fitted as $(1.8\pm0.2)\times10^{-5}\ \mu\Omega\,\mathrm{cm/K^2}$, in agreement with the value $(1.73\pm0.17)\times10^{-5}\ \mu\Omega\,\mathrm{cm/K^2}$ of Mackliet, Schindler, and Gillespie.[11] The fitted value of the spin-fluctuation temperature T_s is 160 K, but this is rather uncertain due to the uncertainty in the slope of the $\Delta\rho(T)$ data. Keeping the T^2 coefficient ΔA at the value determined by Mackliet, Schindler, and Gillespie,[11] the upper limit for $\Delta\rho(T)$ when fitted to our Eqs. (3)–(5) gives $T_s\sim300$ K and the lower limit $T_s\sim70$ K. However, the fit (not shown) for the upper dotted line in Fig. 2 is poorer because a value of $T_s\sim300$ K implies that $A_{\mathrm{LSF}}(T)$ shows a gradual T^2 to T change around 75 K, in disagreement with the linear behavior of the data above the knee in the DMR term. In fact, regardless of the size of the linear slope of $\Delta\rho(T)$ at higher temperatures, the presence of a significant T^2 term below but not above the main temperature variation in $\Delta(T)$ suggests that the T^2 to T change in $\rho_{\mathrm{LSF}}(T)$ occurs in the temperature range 10–40 K, i.e., T_s is the range 40–160 K. Therefore our estimate of 160 K for T_s also represents the upper limit.

IV. DISCUSSION

We have found that Ni impurities in Pt produce a relatively large and approximately linear thermopower at low temperatures, which is consistent with the presence of LSF's. However, there is no evidence of a peak due to LSF's as seen[9] in *Pd*Ni at 18.5 K, although a peak may occur above 20 K, where it could be masked by the presence of other temperature-dependent effects, particularly phonon drag. In contrast to the case of Ni impurities in Pd, the characteristic thermopower S_{Ni} of Ni impurities in Pt is positive. A somewhat similar reversal of the sign of the nearly magnetic impurity thermopower is seen for *Rh*Fe (Ref. 21) (negative LSF peak) and *Ir*Fe (Ref. 22) (positive LSF peak). In the rather over-simplified context of the one-band model,[7] this means that electrons see an attractive nonmagnetic impurity potential for the $4d$ hosts, but a repulsive potential for the $5d$ hosts.

The magnitude of the thermopower at low temperatures is an order of magnitude smaller in dilute *Pt*Ni alloys than in *Pd*Ni and the difference in the increase ΔA in the T^2 LSF resistivity term is even greater.[4,11] This weakness of LSF in *Pt*Ni leads to the expectation of a greater value of the spin-fluctuation temperature T_s in the random-phase approximation,[6] and in qualitative agreement with this our measurements of the resistivity suggest that T_s is in the range 70–160 K, compared to about 40 K for *Pd*Ni.[2] The resistivity shows relatively large conventional DMR (ascribed largely to varying phonon scattering over the Fermi surface) very similar to that seen in nonmagnetic Pt alloys.

The thermopower of "pure" Pt shows a broad negative peak at very low temperatures that is similar to that in dilute *Pt*Fe alloys, and so is ascribed to trace Fe impurities.[17] Very dilute *Pt*Fe alloys also show a characteristic LSF resistivity[23] increasing with T at low temperatures. The absence of a T^2 behavior in this resistivity down to 0.5 K suggests that $T_s<2$ K for these alloys. The negative thermopower peak[15,17] below 2 K in "pure" Pt with trace Fe impurities is therefore probably an LSF peak, Fe impurities showing much stronger LSF behavior (and so lower T_s) than Ni impurities.

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