s-Electron–Paramagnon Scattering in Dilute Pd–Ni Alloys: Theory and Experiment

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Low-temperature electrical resistivity of dilute Pd–Ni alloys have been experimentally investigated between 2 and 30°K. A theory for *s*-electron–paramagnon scattering has been developed, and the experimental results have been interpreted on this basis. The theoretical model has also been extended to predict the temperature dependence of the thermal conductivity in strongly paramagnetic metals arising from a similar scattering mechanism.

1. BACKGROUND

IT has been recently suggested[1,2] that, in strongly paramagnetic metals, such as those near the ends of the 4-*d* and 5-*d* transition-metal series, a large renormalization of *d*-electron mass occurs as a result of *d*-electron–spin-fluctuation interaction. The theoretical calculations indicate that near the onset of ferromagnetism, the spin fluctuation enhancement of the bare-band mass, m, should vary as[2]

$$m^*/m=1+2\bar{I}\ln\{1/(1-\bar{I})\}, \quad (1)$$

where m^* is the enhanced electron mass, and $\bar{I}$ represents intraband, short-range electron-electron correlation effects. It was also shown that a measure of the strength of this mass renormalization could be obtained from the strength of the exchange enhancement of the Pauli paramagnetic susceptibility;

$$\chi/\chi_0=1/(1-\bar{I}), \quad (2)$$

where χ is the measured susceptibility and χ_0 is the Pauli band susceptibility. This theory originally predicted a spin-fluctuation-enhanced electron mass of 8 for pure Pd[2] (although later it was revised downward, for various reasons, to 3).[3] In fact, the original calculations suggested that as the paramagnetic susceptibility increases sharply near the end of the 4-*d* transition series, the peak in the 4-*d* band, inferred from heat-capacity measurements, was merely a result of increased mass renormalization. The original calculation, based on this theory, seemed to show that rather than being peaked, the 4-*d* band had a low density of states and was, in fact, rather flat.

Clearly, then, since the presence of persistent spin fluctuations, or paramagnons, can only be inferred from a comparison of low-temperature specific-heat data with heat capacities deduced from band calculations, some other independent corroborative evidence is necessary. In this paper, we shall demonstrate that low-temperature electrical-resistivity measurements of strongly paramagnetic metals like palladium can be used to substantiate the existence and presence of paramagnons in such metals.

This paper is composed of two distinct parts. In the first part, a theoretical analysis will be made of the electrical and thermal resistivities arising from *s*-electron scattering from *d*-electron paramagnons in nearly ferromagnetic transition metals. Only *s*-band conduction is assumed, and the calculations consider the paramagnon-induced *s-s* transitions in the paramagnetic state which arise from the spin-flip part of an *s-d* exchange interaction. The results of these calculations yield an electrical resistivity and thermal resistivity which, at low temperatures, is proportional to AT^2-BT^5, and CT-DT^2, respectively. Furthermore, the magnitudes of the coefficients, A, B, C, and D, and the temperature interval over which electron-paramagnon scattering occurs are strongly dependent upon the magnitude of $\bar{I}$.

In the second part of the paper we shall describe a series of measurements of the low-temperature electrical resistivities of Pd and several dilute Pd–Ni alloys. These measurements do, indeed, show that at low temperatures the electrical resistivity is proportional to T^2 and the coefficient of the T^2 term increases strongly as a function of Ni concentration. From these results, a

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[1] S. Doniach and S. Engelsberg, Phys. Rev. Letters **17**, 577 (1966); S. Doniach, S. Engelsberg, and M. J. Rice, in *Proceedings of the Tenth International Conference on Low-Temperature Physics, Moscow, 1966* (Proizvodstrenno-Izdatel'skii Kombinat, UINITI, Moscow, USSR, 1967).

[2] N. F. Berk and J. R. Schrieffer, Phys. Rev. Letters **17**, 433 (1966); in *Proceedings of the Tenth International Conference on Low-Temperature Physics, Moscow, 1966* (Proizvodstrenno-Izdatel'skii Kombinat, UINITI, Moscow, USSR, 1967). If the effects of longitudinal spin-fluctuation modes are included [see W. Brenig, H J. Mikeska, and E. Riedel, Z. Physik (to be published), and M. J. Rice and D. S. Betts (to be published)] the coefficient of the logarithmic term in Eq. (1) is increased by a factor of $\frac{3}{2}$.

[3] S. Doniach, Phys. Rev. Letters **18**, 554 (1967). The mass enhancement appears to be more effectively reduced by interatomic exchange than by Hund's rule coupling [see E. Bucher, W. F. Brinkman, J. P. Maita, and H. J. Williams, Phys. Rev. Letters **18**, 1125 (1967); and J. R. Schrieffer (to be published)].

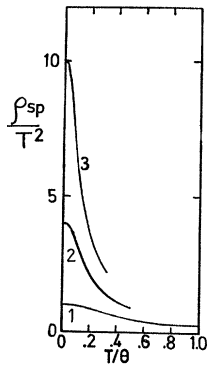

FIG. 1. Dependence on temperature and θ of ρ_{sp}/T^2 calculated from Eq. (2.9). Curve 1 is a plot of ρ_{sp}/T^2 (arbitrary units) against T/θ. Curves 2 and 3 are relative plots corresponding to reduction of the value of θ in 1 by $\frac{1}{2}$ and $1/\sqrt{10}$, respectively.

characteristic paramagnon temperature θ is deduced which can be reconciled with theory.

2. THEORY OF THE SPIN-FLUCTUATION ELECTRICAL AND THERMAL RESISTIVITIES

The electrical and thermal resistivities, ρ_{sp} and W_{sp}, respectively, that can be expected to arise from s-electron scattering from itinerant d-band spin-density fluctuations, or paramagnons, in nearly ferromagnetic transition-metal systems like Pd and dilute Pd–Ni, are investigated theoretically in this section on the basis of a simple approximation for the spin-fluctuation spectral-density function $A\mathbf{q}(\omega)$. The model, used by Rice[4] in treating an analogous transport problem in liquid He^3, predicts a specific dependence of ρ_{sp} and W_{sp} on temperature and on closeness to the ferromagnetic state.

We consider the paramagnon-induced s-s transitions in the paramagnetic state to arise from the spin-flip part of the s-d exchange interaction, which we take to be of the form

$$H_{int}=(J/N)\sum_{k,q}\{a_{k-q\uparrow}{}^{\dagger}a_{k\downarrow}\sigma_q{}^{(-)}+a_{k+q\downarrow}{}^{\dagger}a_{k\uparrow}\sigma_{-q}{}^{(+)}\},$$
$$\sigma_q{}^{(\pm)}=\sum_k d_{k+q\uparrow,\downarrow}{}^{\dagger}d_{k\downarrow,\uparrow}, \tag{2.1}$$

(N=number of atoms in the crystal), where J denotes a coupling constant, $\sigma_q{}^{(\pm)}$ are spin-density fluctuation operators for the d band, and the a's and d's are the momentum-space creation and annihilation operators for the s and d electrons (holes), respectively. Following the formalism of Mills and Lederer,[5] matrix elements for these transitions may be calculated within the Born approximation in terms of the spin-fluctuation spectral-density function $A\mathbf{q}(\omega)$. These can be treated in the usual way[6] to construct the collision term $\partial f_{k\sigma}/\partial t]_{coll}$ for use in the Boltzmann transport equation for the s-electron distribution function $f_{k\sigma}$ (σ denotes the spin orientations, "$\uparrow$," "$\downarrow$"). In doing this, we shall assume that the d band makes a negligible contribution to the electronic-transport processes. Consequently, the d-band fluctuations in spin density will be treated as being at equilibrium. We then have[4,6]

$$\frac{\partial f_{k\uparrow,\downarrow}}{\partial t}\Big]_{coll}=\frac{2\pi}{\hbar^2}\frac{J^2}{N^2}\sum_q\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}A_q(\omega)\delta(\epsilon_{k\pm q\downarrow,\uparrow}-\epsilon_{k\uparrow,\downarrow}\mp\hbar\omega)\{(1-f_{k\uparrow,\downarrow})f_{k\pm q\downarrow,\uparrow}[n(\omega)+\tfrac{1}{2}\pm\tfrac{1}{2}]-f_{k\uparrow,\downarrow}$$
$$\times(1-f_{k\pm q\downarrow,\uparrow})[n(\omega)+\tfrac{1}{2}\mp\tfrac{1}{2}]\}, \tag{2.2}$$

where $\epsilon_{k\uparrow}=\epsilon_{k\downarrow}$ denotes the energy of an s electron in the momentum state $\hbar\mathbf{k}$, $n(\omega)$ is the Planck function $[\exp\{\hbar\omega/k_BT\}-1]^{-1}$, and k_B is the Boltzmann constant. Equation (2.2) may be at once fed into the standard variational procedure[6] to obtain expressions for ρ_{sp} and W_{sp}. If the deviation of $f_{k\sigma}$ from its equilibrium value $f_{k\sigma}{}^0=[\exp\{(\epsilon_{k\sigma}-\epsilon_s)/k_BT\}-1]^{-1}$ is written as $-(\partial f_{k\sigma}{}^0/\partial\epsilon_{k\sigma})\phi_{k\sigma}$ we find

$$\rho_{sp}=\frac{(1/k_BT)\int d^3k\int d^3k'V(\mathbf{k}'\downarrow;\mathbf{k}\uparrow)\{\phi_{k'\downarrow}-\phi_{k\uparrow}\}^2}{|\,(2e\hbar/m_s)\int d^3k\mathbf{k}\phi_{k\uparrow}(\partial f_{k\uparrow}{}^0/\partial\epsilon_{k\uparrow})\,|^2}, \tag{2.3}$$

$$W_{sp}=\frac{(1/k_B)\int d^3k\int d^3k'V(\mathbf{k}'\downarrow;\mathbf{k}\uparrow)\{\phi_{k'\downarrow}-\phi_{k\uparrow}\}^2}{|\,(2\hbar/m_s)\int d^3k\mathbf{k}(\epsilon_k-\epsilon_s)\phi_{k\uparrow}(\partial f_{k\uparrow}{}^0/\partial\epsilon_{k\uparrow})\,|^2}, \tag{2.4}$$

where

$$V(\mathbf{k}'\downarrow;\mathbf{k}\uparrow)=\frac{(2\pi)^3J^2}{\hbar N^2}\sum_q\int_{-\infty}^{\infty}d\omega\,A_q(\omega)n(\omega)f_{k\uparrow}{}^0(1-f_{k'\downarrow}{}^0)\delta(\epsilon_{k\downarrow}{}'-\epsilon_{k\uparrow}-\hbar\omega)\delta(\mathbf{k}'-\mathbf{k}-\mathbf{q}). \tag{2.5}$$

We have now introduced an effective-mass description $\epsilon_{k\sigma}=\hbar^2k^2/2m_s$ for the s electrons. The Fermi energy of the s electrons is denoted by ϵ_s and the electronic charge by e. In order to calculate (2.3) and (2.4), we need to know $\phi_{k\sigma}$ and $A_q(\omega)$. For $\phi_{k\sigma}$ we take the usual trial solutions,[6]

$$\phi_{k\sigma}=\mathbf{k}\cdot\mathbf{u}\times\text{const} \tag{2.6}$$

for ρ_{sp}, and

$$\phi_{k\sigma}=\mathbf{k}\cdot\mathbf{u}(\epsilon_{k\sigma}-\epsilon_s)\times\text{const} \tag{2.7}$$

for W_{sp}, where $\mathbf{u}$ denotes a unit vector in the direction of the applied electric field in (2.5) and a unit vector in

[4] M. J. Rice, Phys. Rev. **159**, 153 (1967).

[5] D. L. Mills and P. Lederer, J. Phys. Chem. Solids **27**, 1805 (1966).

[6] J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, England, 1960), Chap. 7.

the direction of the thermal gradient in (2.7). For $A\mathbf{q}(\omega)$, we employ the following model approximation[4]:

$$\begin{aligned} A_q(\omega) &= \alpha_0\omega/q \qquad (0\leq|\omega|\leq\omega_q) \\ &= 0 \qquad (|\omega|>\omega_q), \\ \hbar\omega_q &= (4/\pi)(K_0^2/\bar{I})(q/k_d)\epsilon_d \qquad (0\leq q\leq Q) \\ &= 0 \qquad (q>Q), \\ \alpha_0 &= N\hbar^2 N_0(\epsilon_d)\pi k_d/2K_0^4\epsilon_d. \end{aligned} \tag{2.8}$$

In the above equations $1/K_0^2=1/1-\bar{I}$ denotes the random-phase approximation (RPA) exchange enhancement factor for the itinerant d band[4,7] and ϵ_d, k_d, and $N_0(\epsilon_d)$ the corresponding Fermi energy, wave vector, and unrenormalized density of states, respectively. Q is a cutoff wave vector of order k_d and is to be regarded as a disposable parameter. The model is an approximation to the actual form of the correct RPA result for $A_q(\omega)$,[7] evaluated at $T=0$ for small $q/2k_d$. We believe it to represent the essential qualitative structure of $A_q(\omega)$ close to the ferromagnetic state, i.e., the linear dependence on ω for $|\omega|<\omega_q$, and the strong peaking of $A_q(\omega)$ about $\omega=\omega_q$ as $K_0^2\to 0$.[7] Using (2.6)–(2.8) and an effective-mass approximation for the d band, expressions (2.3) and (2.4) may be evaluated (cf. Ref. 4) to give the following results for ρ_{sp} and W_{sp}:

$$\rho_{sp}=\alpha(T/\theta)^2\{J_2(\theta/T)-(T/\theta)^3 J_5(\theta/T)\}, \tag{2.9}$$

$$W_{sp}=(9\alpha/\bar{Q}^2\pi^2\theta L_0 W_k^{-1}(0))(T/\theta)w_K(T/\theta), \tag{2.10}$$

where $\bar{Q}=Q/k_d$,

$$k_B\theta=\hbar\omega_Q=(4/\pi)(K_0^2/\bar{I})\epsilon_d\bar{Q} \tag{2.11}$$

defines a characteristic paramagnon energy, L_0 denotes the ideal Lorentz ratio, and

$$\alpha=(9\pi\hbar\bar{Q}^5/8e^2\bar{I}^2k_s)(k_d/k_s)(\nu_d J/\epsilon_s)^2 \tag{2.12}$$

(ν_d=number of d electrons/atom). The functions $w_K(T/\theta)$ and $J_n(\theta/T)$ are defined by the relations

$$\begin{aligned} w_K(t) &= W_K(t)/W_K(0), \\ W_K(t) &= [J_4(1/t)-tJ_5(1/t)]+\tfrac{1}{9}(\bar{Q}^2\pi^2) \\ &\quad\times[J_2(1/t)-t^3J_5(1/t)] \\ &\quad-(\bar{Q}^2/18)[J_4(1/t)-t^3J_7(1/t)], \end{aligned}$$

$$J_n(y)=\int_0^y \frac{dx\,x^n}{(e^x-1)(1-e^{-x})} \qquad (n\geq 2). \tag{2.13}$$

For small (T/θ), the Bloch-Grüneisen integrals $J_n(\theta/T)$ may be replaced by the constants

$$J_n(\infty)\equiv J_n^0(=n!\sum_{s=1}^{\infty}s^{-n}).$$

This procedure is legitimate for (T/θ) smaller than about $\frac{1}{10}$. Equation (2.9) for ρ_{sp} then becomes

$$\rho_{sp}=(\alpha J_2^0/\theta^2)T^2-(\alpha J_5^0/\theta^5)T^5 \qquad (T\ll\theta), \tag{2.14}$$

while result (2.10) for W_{sp} has the form

$$W_{sp}=(a/\theta^2)T-(b/\theta^3)T^2+O[(T/\theta)^4] \qquad (T\ll\theta), \tag{2.15}$$

with

$$a/b=\{J_4^0(1-\tfrac{1}{18}\bar{Q}^2)+\tfrac{1}{9}(\pi^2\bar{Q}^2)J_2^0\}/J_5^0, \tag{2.16}$$

where a is a constant, independent of K_0^2 and definable from (2.10) and (2.13). Close to the ferromagnetic state, where $\bar{I}\lesssim 1$, θ is proportional to K_0^2. Then it is seen from (2.14), (2.15), and (2.12) that as $T\to 0$, ρ_{sp} varies as T^2 and W_{sp} as T, where the constants of proportionality increase with the square of the exchange enhancement factor $1/K_0^2$, i.e., with the square of the observed enhancement of the Pauli paramagnetic susceptibility χ_0. We also note that as the ferromagnetic state is approached, the *negative* T^5 and T^2 contributions found above in ρ_{sp} and W_{sp}, respectively, should eventually be able to suppress the corresponding low-temperature T^5 and T^2 electron-phonon contributions in the metal. We emphasize that (2.14) and (2.15) are only valid for small (T/θ). In Fig. 1 we show the variation of ρ_{sp} in the temperature range $0\leq T/\theta\leq 1$ as calculated from Eq. (2.9) for several relative values of θ. As θ is lowered in magnitude it is seen that while the low-temperature coefficient of T^2 increases, the domain of validity of the T^2 law rapidly diminishes. Similar conclusions obtain for the range of validity of the linear T law found for W_{sp}.[8] Further aspects of the present theory will be remarked upon in Sec. 3, where a comparison between theory and experiment will be discussed.

3. EXPERIMENTAL

To test the preceding theory, and, indeed, to show that paramagnons do exist, low-temperature electrical-resistivity measurements were made on a series of dilute Pd–Ni alloys. Samples of pure palladium (having a resistance ratio $R_{300}/R_{4.2}$ in excess of 1000) and palladium containing 0.5, 1.0 and 1.66 at.% nickel were examined; the temperature region investigated was from 2 to 30°K. These alloys were chosen because of their unique magnetic properties. Pure palladium is strongly paramagnetic and it has been estimated that χ/χ_0 may be as large as 50.[2] Additions of up to 2.25% Ni cause the magnetic susceptibility to increase still further, yet the alloys remain paramagnetic.[9] Since these alloys do not exhibit any complicating localized magnetic-moment phenomena,[10] it appears likely that the

[7] S. Doniach, Proc. Phys. Soc. (London) **91**, 86 (1967).

[8] A detailed evaluation of this temperature dependence may be found in Ref. 4, where a similar result was predicted for the thermal resistivity of liquid He^3.

[9] R. M. Bozorth, D. D. Davis, and J. H. Wernick, J. Phys. Soc. Japan **17**, Suppl. B-1, 112 (1962).

[10] D. Shaltiel, J. H. Wernick, and H. J. Williams, Phys. Rev. **135**, A1346 (1964).

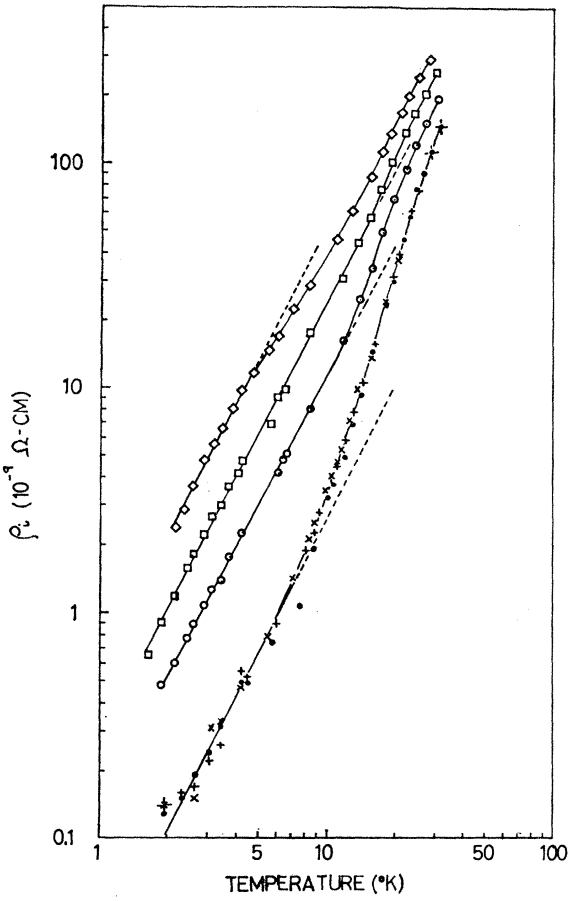

FIG. 2. The ideal electrical resistivity versus temperature. ●, ×, +, three samples of high purity Pd; ○, Pd–0.5% Ni; □, Pd–1% Ni; ◇, Pd–1.66% Ni.

large increase in their magnetic susceptibility can be attributed to an increase in their exchange enhancement. As a result, we should expect that the fraction of the electrical resistivity resulting from electron-paramagnon scattering would increase strongly as a function of nickel content.

Procedure

The alloys were prepared by induction melting 5*N* pure nickel and palladium. Quartz-lined, stabilized-zirconium crucibles were employed, and the specimens were melted under a purified argon atmosphere. Two specimen shapes were employed. For the pure palladium samples, three samples approximately 2 cm long by $\frac{1}{2}$ cm^2 cross-sectional area were spark-cut from a cylindrical ingot; for the alloys, 0.015-in. diam by 2 cm long wires were swaged and cut from the resultant ingots. Following the final swaging pass, the alloy wires were all carefully annealed at 1200°C for 20 h.

The sample resistances were measured, using a conventional four-probe dc potentiometric circuit. A Tinsley-Diesselhorst potentiometer and galvanometer amplifier having a noise level of 10^{-9} V was employed; sample currents of 0.3 to 0.5 A were used. A sample holder which could accommodate up to 5 samples was employed. For temperatures below 4.2°K, the holder and samples were immersed directly in pumped liquid helium. Temperatures were measured by measuring the He vapor pressure above the bath. For resistance data above 4.2°K, the liquid-He level was allowed to drop below the bottom of the sample holder, and measurements were made during the slow warmup; the warmup rate was never more than 10°K/h up to 30°K. Temperatures in this range were measured using a calibrated Au-0.03% Fe versus Ag-0.37% Au thermocouple.

4. RESULTS AND DISCUSSION

The experimental results of this investigation are shown in Figs. 2–5. In Fig. 2, the ideal electrical resistivities defined as ρ measured minus ρ residual, of pure Pd and the three Pd–Ni alloys are plotted versus temperature on a log-log plot. At the lowest temperatures, the resistances of all the samples vary as T^2. We interpret this as the low-temperature electron-paramagnon scattering contribution which was deduced from Eq. (2.9) for low values of T/θ. However, as the temperature is increased a marked deviation from the T^2 behavior is noted. We consider this variation to be the result of both the temperature dependence of ρ_{sp} as well as that of ρ_{ph}, the usual intraband electron-phonon scattering term which is present for all metals. For pure Pd, this results in a T^2 dependence to 7°K. If we assume that, over the entire region investigated, $\rho_{sp}=AT^2$ for pure Pd [i.e., $T\leq 30°K\ll\theta(\mathrm{Pd})$], then we can extract $\rho_{ph}(T)$ from our measured value of $\rho_i(T)$. This has been done, and the result is shown in Fig. 3. Although simple

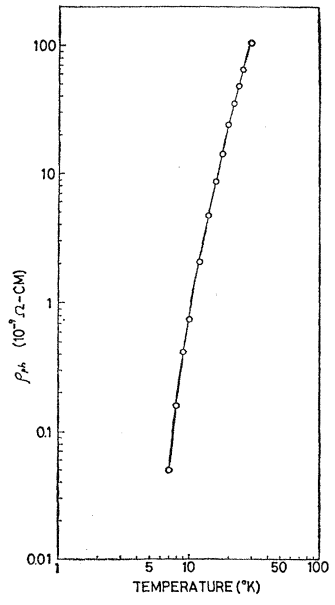

FIG. 3. ρ_{ph} versus temperature for pure Pd.

electron-phonon scattering theory would suggest that this term should go as T^5 for low T, our measurements indicate only a rather limited region where ρ_{ph} varies as T^5.

As one adds Ni to Pd, the coefficient of the T^2 term in the resistivity increases; in addition, the region over which a T^2 dependence is found is a strong function of the temperature. For the Pd–$\frac{1}{2}$%Ni specimen, the resistivity goes as T^2 to 11°K; for Pd–1%Ni to 17°K; and for Pd–1.66%Ni to 5°K. Since $\rho_{sp} \propto T^2$ for values of $T/\theta \lesssim \frac{1}{10}$, this would appear to imply that θ is not a monotonically decreasing function of Ni concentration. This apparent discrepancy with theory can be resolved by looking directly at only the spin-flip contribution to the electrical resistivity.

To extract ρ_{sp} from ρ_i for all the alloys requires the knowledge of the dependence of ρ_{ph} as a function of Ni concentration. Since it seems reasonable to assume that adding up to only 1.66% Ni would not significantly affect the ρ_{ph}, i.e., Matthiessen's rule is reasonably well obeyed, we shall, as a first approximation, assume that ρ_{ph} for all the alloys is equal to that of pure Pd. Subtracting this quantity from the alloy data yields the results shown in Fig. 4. Here we have plotted (ρ_{sp}/T^2) as a function of temperature to illustrate the interval over which ρ_{sp} can be approximated by AT^2. Clearly, this interval extends to approximately 14°K for Pd–$\frac{1}{2}$%Ni, to 10°K for Pd–1%Ni, and to somewhat under 3°K for Pd–1.66%Ni. If we now use the T/θ-$\frac{1}{10}$ rule, the data would suggest $\theta \simeq 140$, 100 and 30°K for the three alloys, respectively. Using these values of θ, we have calculated ρ_{sp} from Eq. (2.9) and the results are shown as the solid lines in Fig. 4; these are in fairly good accord with the experimental data.

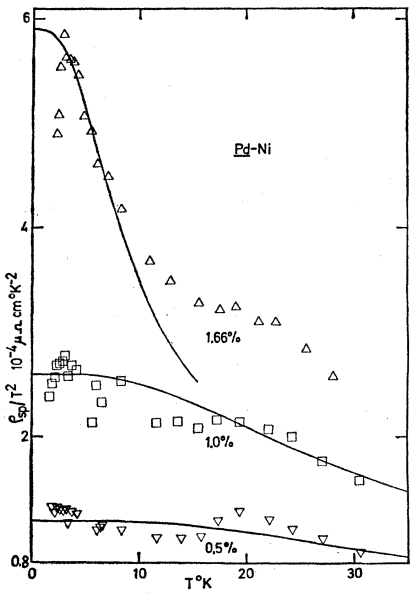

FIG. 4. (ρ_{sp}/T^2) versus temperature for three Pd–Ni alloys. The solid lines have been calculated using Eq. (2.9).

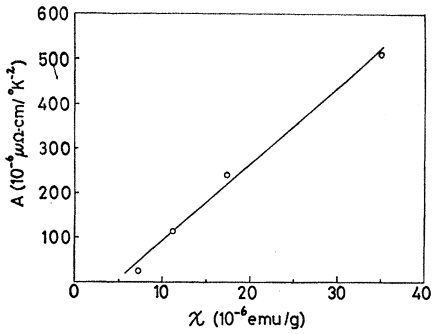

FIG. 5. A versus magnetic susceptibility, the latter measured at 4.2°K.

One can now understand the origin of the peculiar temperature dependence of ρ_i for the three alloys. Simply put, the second term of ρ_{sp} in Eq. (2.9) yields a negative T^5 term to ρ_i, which increases as a function of Ni concentration. Mixing this contribution with the Bloch-Grüneisen T^5 resistivity results in an apparent electron-phonon contribution which decreases as Ni is added to Pd. For the Pd–$\frac{1}{2}$%Ni specimen, this mixing results in the T^2 dependence of ρ_i extending to 11°K. Almost complete cancellation of the T^5 terms occur for the Pd–1.0%Ni specimen, and ρ_i goes as T^2 to 17°K. For Pd–1.66%Ni, the negative T^5 contribution of ρ_{sp} has become so large that a negative deviation in ρ_i occurs at approximately 4.5°K.

We consider the latter qualitative features of the experimental results to constitute strong evidence for the existence of paramagnons in these alloys.

One aspect of the theory which is not obeyed is the relationship between the coefficient of the T^2 term $(\alpha/\theta^2 = A)$ of ρ_{sp} and the exchange enhancement. The theory would suggest that this coefficient should vary as $(\chi/\chi_0)^2$. Since small additions of Ni to Pd are unlikely to affect χ_0, the theoretical prediction is seen to suggest that A should vary as χ^2. Measurements of χ for pure Pd and the three Pd–Ni alloys were performed at 4°K by Williams of this laboratory, and the variation of A versus χ is shown in Fig. 5. Clearly, in contrast with the theory, A is found to increase much less rapidly than as χ^2. Presumably, the latter disagreement could be rectified by using in place of (2.8) the complete RPA result for $A_q(\omega)$, for which, in the higher frequency region $|\omega| > \omega_q$, $A_q(\omega)$ is not enhanced over its value for the noninteracting d band by as much as $1/(1-\bar{I})^2$.

The resistivity data presented above do not enable us to estimate the value of θ for pure Pd. In fact, in the analysis, it was tacitly assumed that θ for pure Pd was much larger than the maximum temperature used in the measurements. This enabled us to use a T^2 dependence for ρ_{sp} in our subtraction process from ρ_i to obtain ρ_{ph}. However, within the framework of the present theory we can estimate a value of θ for pure Pd. To do this we shall assume the Fermi surface of palladium to be composed of three spherical d-hole

sub-bands each containing 0.12 hole/atom, and a spherical s band containing 0.36 electrons/atom.[11] If we take[12] $m_d=5.4M_e$, we have[7] $\epsilon_d=0.25$ eV and $\bar{I}=0.925$. Using (2.11), these values lead to an estimate of θ in terms of $\bar{Q}$,

$$\theta=220°\text{K}\,\bar{Q}.$$

We have no real way of estimating $\bar{Q}$. However, we shall make a self-consistent picture by setting $\bar{Q}=1.4$. This gives us a value of 308°K for θ for Pd. Furthermore, using Eq. (2.12) and remembering that $A=\alpha/\theta^2$, we find that to fit the experimentally determined value of A for palladium requires J to be 0.3 eV. This can in turn be used to estimate s-electron mass enhancement due to interaction with d-band paramagnons.[13]

A more appropriate way of obtaining $\bar{Q}$ can be had from an experimental determination of the Wiedemann–Franz ratio. From Eqs. (2.9), (2.10), (2.12), and (2.13), we see that the low-temperature T^2 electrical resistivity and the linear T thermal resistivity are related by an effective Wiedermann-Franz ratio $L=L_0f(\bar{Q})$, where

$$f(\bar{Q})=\left\{1+\frac{9J_4{}^0(1-\bar{Q}^2/18)}{\pi^2J_2{}^0\bar{Q}^2}\right\}^{-1}. \tag{4.1}$$

Thus, if L_0 could be determined experimentally, (4.1) could be used to provide an estimate of the cutoff parameter $\bar{Q}$. However, because of our use of the simple variational functions (2.6) and (2.7), Eq. (4.1) is not yet sufficiently accurate to warrant such a procedure. The main source of error here is to be found in the employment of Eq. (2.7), which probably results in an overestimate of W_{sp}, and hence an underestimate in $f(\bar{Q})$, by a factor of order 2.[14]

We can, at least, use Eq. (4.1) to predict the order of magnitude of the thermal resistivity linear in T from the observed values of the T^2 electrical resistivity. For pure Pd we find the thermal resistivity linear in T to be $\sim5\times10^{-3}T$ cm deg/W. The T^2 electron-phonon thermal resistivity in Pd was found by White and Woods to be $W_{\text{el-ph}}=3.6\times10^{-4}T^2$ cm deg/W.[15] Thus the spin-fluctuation thermal resistivity linear in T should be distinguishable from the T^2 electron-phonon thermal resistivity below about 10°K for pure Pd. In Pd–Ni alloys, the spin-fluctuation thermal resistivity should dominate over $W_{\text{el-ph}}$ in much the same way as ρ_{sp} was observed to dominate ρ_{ph} in this paper.

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APPENDIX: S-ELECTRON-PARAMAGNON MASS ENHANCEMENT

We may use the value $J\sim0.3$ eV, arrived at in Sec. 3, to estimate the effective s-electron mass $m_s{}^*$ due to the interaction with the d-band paramagnons. In terms of Green's functions, the single s-electron self-energy $\sum_\sigma(\mathbf{k},\omega)$ due to this interaction is, within the model (2.1),

$$\sum_{\uparrow}(\mathbf{k},\omega)=(-i)\left(\frac{J}{N}\right)^2\sum_{\mathbf{q}}\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}G_{\downarrow}{}^0(\mathbf{k}+\mathbf{q},\omega+\omega')\times3\chi_{\mathbf{q}}{}^{-+}(\omega'), \tag{A1}$$

where $G_\sigma{}^0(\mathbf{k},\omega)$ denotes the unperturbed single s-electron Green's function and $\chi_{\mathbf{q}}{}^{-+}(\omega)$ the RPA d-electron spin-fluctuation (paramagnon) propagator for a single sub-band, defined explicitly in Ref. 1. Following an approximate calculation of analogous corrections to the d-electron effective mass,[1] (A1) may be evaluated to give the estimate

$$m_s{}^*/m_s\simeq1+9(J/I)^2(m_s/m_d)(\nu_d/\nu_s)^{1/3}\times\ln[1+(\bar{Q}^2\bar{I}/12K_0{}^2)]. \tag{A2}$$

Using $J=0.3$ eV, and taking $\bar{Q}=1.4$ and (m_s/m_d) to be as large as $\frac{1}{3}$, (A2) yields $m_s{}^*/m_s-1\sim3\%$ for Pd. The s-electron mass enhancement due to interactions with the d-band paramagnons is therefore very small in Pd. However, because of the logarithmic factor in (4.2) the s-electron mass enhancement can become significant in the region of the ferromagnetic instability ($\sim2\%$ Ni). Nevertheless, its contribution to the enhancement of the electronic specific heat will still be of the order $(J/I)^2(m_s/m_d)$, smaller than that arising from the corresponding d-electron mass enhancement, which also diverges logarithmically at the ferromagnetic instability.[1,2]