# Localized Spin Fluctuations in Nearly-Magnetic Dilute Alloys: Thermal Resistivity and Lorenz Number\*

#### A. B. Kaiser

W. W. Hansen Laboratories of Physics, Stanford University, Stanford, California 94305<sup>†</sup> (Received 21 September 1970)

We calculate the thermal resistivity and Lorenz number due to scattering of conduction electrons from localized spin fluctuations, using a model appropriate for nearly-magnetic transition-metal impurities in a host of similar electronic structure. The temperature dependence of the Lorenz number, which is determined by the characteristic energy of the spin-fluctuation excitation spectrum, is predicted for IrFe and related alloys assuming the spin-fluctuation model is applicable.

#### I. INTRODUCTION

We have shown<sup>1</sup> that a good account of the anomalous electrical resistivity of *Ir* Fe and related alloys is given by the scattering of conduction electrons from localized spin fluctuations (local paramagnons). In this paper we make an analogous calculation of the thermal resistivity and Lorenz number.

The spin-fluctuation model we use was first proposed by Lederer and Mills<sup>2</sup> to explain the  $T^2$  term in the electrical resistivity of dilute PdNi alloys. It applies to dilute transition-metal alloys with nearly magnetic impurities in which the host and impurity have similar electronic structure, the interference between potential scattering and spin-fluctuation scattering being neglected. The intraatomic Coulomb interaction  $U_0$  between oppositespin d electrons in the host is considered to be increased to a value  $(U_0 + \delta U)$  in the impurity cell, the Hamiltonian being

$$H = T + U_0 \sum_{i} n_{i}, n_{i} + \delta U \sum_{i} n_{i}, n_{i}$$
.

Here T is the kinetic energy and  $n_{i\sigma} = c_{i\sigma}^{\dagger} c_{i\sigma}$  is the number operator for d electrons of spin  $\sigma$ ,  $c_{i\sigma}^{\dagger}$  and  $c_{i\sigma}$  being the creation and destruction operators for the ith-cell Wannier orbital. The sum i is taken over all cells, while sum j is over just the impurity cells. Lederer and Mills calculated the generalized susceptibility response function for this Hamiltonian in the random-phase approximation. The spectral density for the spin fluctuations is given by the imaginary part of this response function.

The model should be appropriate for nearly-magnetic 3d impurities in 4d and 5d hosts from the same or nearby columns. For instance, the behavior of Fe impurities in the 4d series Ru-Rh-Pd changes from nonmagnetic in Ru to ferromagnetic in Pd as the exchange enhancement in the host increases. For the intermediate case of Fe impurities in Rh (and in the corresponding 5d-metal Ir), we suggested that the temperature dependence of the

resistivity and susceptibility could be understood in terms of strong localized spin fluctuations at the Fe sites.

Although the model is too oversimplified to allow quantitative predictions from first principles, it does seem capable of accounting for the main qualitative features of the experimental data. We calculated a universal curve for the electrical resistivity showing a gradual change from a  $T^2$  to a linear dependence at temperatures around 0.25 $T_s$ , where  $T_s$  is the characteristic temperature of the localized spin fluctuations in the alloy. Fitting this universal curve gave values of  $T_s$  for IrFe ( $T_s \sim 30\,^{\circ}$ K), RhFe ( $T_s < 1.5\,^{\circ}$ K), and PdNi ( $T_s \sim 80\,^{\circ}$ K). The essential feature of the model applied to IrFe and especially RhFe is the small energy deduced for the localized spin fluctuations.

This paper suggests a method of testing the localized spin-fluctuation model proposed for IrFe and RhFe by measuring the thermal resistivity and Lorenz number. At low temperatures, the socalled "vertical processes" due to inelastic scattering by spin fluctuations make a large contribution to the thermal resistivity, and so reduce the Lorenz number below the classical value, as shown by Schriempf, Schindler, and Mills (SSM). 3 However, the effect of these vertical processes is greatly reduced as the temperature increases above  $T_s$ , since the spin fluctuations then have insufficient energy to scatter electrons through the thermal layer. Hence, the increase of the Lorenz number with temperature indicates directly the order of magnitude of the energy of the excitations producing the inelastic scattering, i.e., gives a check on the order of magnitude of T<sub>s</sub> if the excitations are assumed to be spin fluctuations.

It has been suggested<sup>4</sup> that the electrical resistivity due to s-d exchange scattering from magnetic impurities may decrease as temperature decreases in the presence of strong nonmagnetic scattering (in contrast to the more usual opposite behavior<sup>5</sup>), and that RhFe and IrFe are Kondo systems of this

type (although potential scattering is weak in these alloys). If this were the case, fitting the measured susceptibilities of these alloys to a Curie-Weiss law indicates that the Kondo temperatures (about  $10\,^\circ\text{K}$  for RhFe and  $100\,^\circ\text{K}$  for IrFe) are considerably larger than the corresponding values of  $T_s$  if the localized spin-fluctuation model is applicable. The temperature dependence of the Lorenz number might therefore provide evidence to determine whether the Kondo model or the localized spin-fluctuation model is more appropriate for IrFe and RhFe.

SSM³ have made accurate measurements of the thermal resistivity of dilute PdNi alloys below 20 °K and have shown that the Lorenz number for electron-electron scattering is small, as predicted by their calculation, which is valid for temperatures  $T \ll T_s$ . We extend the calculation to higher temperature ranges to obtain predictions for IrFe and RhFe. Since the presence of lattice conductivity reduces the accuracy of measurements of electronic thermal resistivity, we consider the effect of lattice conductivity on the separation of thermal resistivity components, with reference to the PdNi data of SSM.³

We mention that a linear T term in the thermal resistivity (corresponding to a  $T^2$  term in the electrical resistivity) has been observed at low temperatures in several transition metals besides PdNi alloys: It is found that the Lorenz number for electron-electron scattering is reduced below the classical Sommerfeld value. To account for this behavior, several calculations have been made of the thermal resistivity and Lorenz number for electron-electron scattering, and in particular for scattering from uniform band paramagnons at low

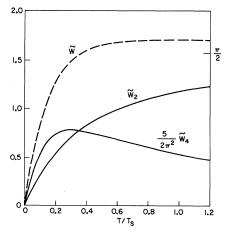


FIG. 1. Universal curves for the thermal resistivity due to scattering by localized spin fluctuations, for the horizontal  $(\tilde{W}_2)$  and vertical  $(\tilde{W}_4)$  components of scattering. The total localized spin-fluctuation resistivity  $\tilde{W}$  is shown for the case  $a_1/a_3=1$ .

temperatures<sup>9,10</sup>; the relation of these calculations to the present localized spin-fluctuation model has been discussed in some detail by SSM.<sup>3</sup> The meaning of "vertical" and "horizontal" processes for inelastic scattering from spin fluctuations has also been fully discussed by SSM.<sup>3</sup>

### II. LORENZ NUMBER

The calculation of the thermal resistivity  $W_m$  due to scattering of conduction electrons from d-electron spin-density fluctuations localized at impurity sites is given in the Appendix. We find that the normalized thermal resistivity  $\tilde{W}$  for spin-fluctuation scattering is

$$\tilde{W} = \tilde{W}_2 + \frac{3}{\pi^2} \left( \frac{a_1}{a_3} - \frac{1}{6} \right) \tilde{W}_4 . \tag{1}$$

The integrals  $\tilde{W}_2$  and  $\tilde{W}_4$  are defined by (A20) and plotted in Fig. 1 as a function of normalized temperature  $\tilde{T} = T/T_s$ , where  $k_B T_s$  is the energy of the peak in the spin-fluctuation excitation spectrum. Thus, for thermal resistivity there are two universal curves, one arising from "horizontal" processes  $(\tilde{W}_2)$  and the other from "vertical" processes  $(\tilde{W}_4)$ .

In a vertical process, which contributes to the thermal but not the electrical resistivity, the energy of a conduction electron is changed by inelastic scattering from a spin fluctuation. For instance, the heat current carried by a "hot" electron is reversed if it is scattered through the Fermi surface to become a "cold" electron, although the electrical current is not significantly changed unless the direction of motion of the electron is changed, i.e., unless there is significant horizontal scattering. The spin-fluctuation thermal resistivity is the sum of the vertical and horizontal components, whose relative magnitude is rather difficult to predict since it is determined by the value of the ratio  $a_1/a_3$  (defined in the Appendix) which depends on the angular distribution of scattering.

Since only the horizontal component of scattering contributes to electrical resistivity, we see that

$$\tilde{\rho} = \tilde{W}_2 \tilde{T} , \qquad (2)$$

where  $\tilde{\rho}$  is the universal curve for the electrical resistivity due to localized spin fluctuations. Taking the ratio of electrical resistivity  $\rho_m$  and thermal resistivity  $W_m$  due to scattering by spin fluctuations, we obtain the Lorenz number  $L_m$  for localized spin-fluctuation scattering

$$L_{m} = \frac{\rho_{m}}{W_{m}T} = L_{0} \frac{\tilde{\rho}}{\tilde{W}\tilde{T}} = L_{0} \left[ 1 + \frac{3}{\pi^{3}} \left( \frac{a_{1}}{a_{3}} - \frac{1}{6} \right) \frac{\tilde{W}_{4}}{\tilde{W}_{2}} \right]^{-1} ,$$
(3)

where  $L_0$  is the classical Sommerfeld value of the Lorenz number:

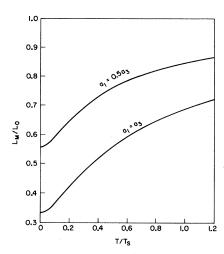


FIG. 2. Lorenz number  $L_m$  for scattering due to localized spin fluctuations.

$$L_0 = \pi^2 k_B^2 / 3e^2 \ . \tag{4}$$

Here  $k_B$  is Boltzmann's constant and e the electronic charge. At low temperatures

$$L_m(T \to 0) = L_0 5(3 + 12a_1/a_3)^{-1}$$
, (5)

which is the temperature-independent Lorenz number calculated by SSM.<sup>3</sup>

Two examples of  $L_m$  for different values of the ratio  $a_1/a_3$  are shown in Fig. 2. From (2),  $\rho_m$  and the horizontal scattering term of  $W_m$  are related at all temperatures by the classical Lorenz number  $L_0$ , so the deviation of  $L_m$  from the value  $L_0$  arises from the presence of the vertical scattering component. Hence  $L_m - L_0$  at high temperatures where the contribution of the vertical component  $\tilde{W}_4$  tends to zero. The horizontal resistivity component has its largest magnitude for the case of uniform angular scattering, in which case  $a_1/a_3 = 0.5$ , according to (A10) below. Hence, the curve for  $a_1/a_3 = 0.5$  in Fig. 2 represents the upper bound of  $L_m$  in our model.

The variation with temperature of the reduction of  $L_m$  below  $L_0$  indicates the energy of the excitations involved in the inelastic scattering processes which give rise to the vertical component of thermal resistivity. Comparison of experimental measurements of  $L_m$  with Fig. 2 should indicate the energy  $k_BT_s$  of the spin-fluctuation excitation spectrum.

## III. SEPARATION OF RESISTIVITY COMPONENTS

Making the usual assumption that the different components of electronic thermal conductivity are additive, and neglecting the lattice conductivity, we obtain the apparent resistivity due to localized spin fluctuations as

$$W'_{m} = W_{imp} - C_{imp}/T = W_{alloy} - W_{host} - C_{imp}/T$$
, (6)

where  $W_{\rm al\,loy}$  and  $W_{\rm host}$  are the measured thermal resistivities in the alloy and in the host, and  $C_{\rm im\,p}/T$  is the thermal resistivity due to nonmagnetic scattering by impurities. As for the electrical resistivity, it is assumed that the electron-phonon resistivity component is the same in the host metal and in the alloys.

Figure 3 shows the data of SSM³ for PdNi alloys replotted to illustrate the method of analysis suggested by (6). We plot  $W_m'T$  rather than  $W_m'$  so that the nonmagnetic component is a constant  $C_{\text{imp}}$ , which can easily be subtracted off. The data are consistent with this method of analysis, since  $W_m'$  is approximately linear in impurity concentration c, and in temperature below 15 °K (as expected for PdNi since the electrical resistivity data $^{9,1}$  suggest  $T_s \sim 80$  °K).

The presence of lattice conductivity will reduce  $W'_m$ . At low temperatures, the lattice conductivity is expected<sup>11</sup> to vary as  $T^2$ :

$$(W_{1at})^{-1} = DT^2$$
, (7)

where D is a constant. Fletcher and Greig<sup>12</sup> have deduced values of  $(W_{\rm lat})^{-1}$  in PdAg alloys with Ag concentrations from 5% to 20%. These measurements suggest<sup>13</sup> that D for dilute Pd alloys is of the order of  $3\times10^{-5}$  to  $10^{-4}$  (W/cm)/ $^{\circ}$ K³. The lattice conductivity in dilute PtAu and PtIr alloys is similar. <sup>12</sup>

The total thermal resistivity W in an alloy or host with electronic resistivity  $W_{\rm el}$  and lattice conductivity (7) is

$$W = W_{e1}(1 + W_{e1}DT^2)^{-1}. (8)$$

Figure 4 shows the effect of lattice conductivity reducing the apparent resistivity  $W_m'$  deduced from (6) below the actual spin-fluctuation resistivity  $W_m$  (taken as linear in T) for parameter values corresponding approximately to PdNi, with  $D=10^{-4}$ 

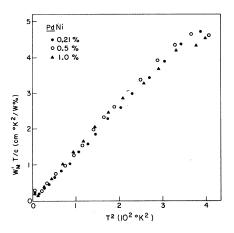


FIG. 3. Thermal resistivity component  $W_m'$  for PdNi alloys derived from the data of  $SSM^3$  using (6).

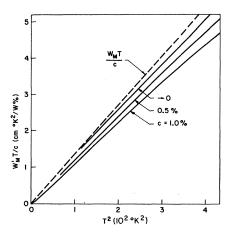


FIG. 4. Apparent thermal resistivity  $W_m'$  for localized spin-fluctuation scattering, showing the effect for different impurity concentrations c of a lattice conductivity (7) with  $D=10^{-4}(\mathrm{W/cm})/\mathrm{^{\circ}K^3}$ . As an approximation for  $Pd\mathrm{Ni}$  below 20 °K,  $W_{\mathrm{el}}$  is taken as  $W_{\mathrm{el}}=(1.1+13.8c)/T+(3.0+13.5c)\times10^{-3}T+2.55\times10^{-4}T^2$  cm °K/W, where T is measured in °K and c in atomic percent.

 $(W/cm)/^{\circ}K^{3}$  taken as the same in pure Pd and the alloys.

The effect of lattice conductivity is roughly proportional to D and to impurity concentration c. Assuming that the addition of impurities affects the electronic conductivity much more strongly than the lattice conductivity, the magnitude of the reduction of  $W_m'/c$  by the lattice conductivity will increase as c increases (although taking  $C_{\rm imp}$  as the intercept on a plot of  $W_m'T$  vs  $T^2$  will tend to reduce this c dependence).

Since the PdNi data in Fig. 3 show little concentration dependence, it is unlikely that the reduction in slope of  $W_m'T/c$  vs  $T^2$  due to lattice conductivity is more than 10%. Taking  $W_m/c=1.32\times 10^{-2}T$  (cm/W)/% for T<15°K, and by an analogous analysis of electrical resistivity data<sup>3</sup>  $\rho_m/c=1.45\times 10^{-4}T^2$  ( $\mu\Omega$  cm/°K²)/%, we get for PdNi when  $T\ll T_s$ :

$$L_m = 1.1 \binom{+0.1}{-0.2} \times 10^{-8} \text{ V}^2/^{\circ}\text{K}^2$$
, (9)

so that  $L_m/L_0$  is between 0.35 and 0.50. To observe the increase in  $L_m$  predicted by our calculation, it would be necessary to extend the PdNi measurements to higher temperatures (of the order of  $T_s$ ).

The value of  $L_m$  in (9) is similar to the value of the Lorenz number for all electron-electron scattering (i.e., including scattering by spin fluctuations in the host Pd) obtained by SSM,  $^3$  using a rather different method of analysis, for each of the PdNi alloys investigated. In contrast, the Lorenz numbers for nonmagnetic scattering from impurities

were found<sup>3</sup> to be within 2% of the classical value  $L_0$ .

### IV. DISCUSSION

We have extended the calculation by SSM³ of the Lorenz number  $L_m$  for spin-fluctuation scattering to temperatures of order  $T_s$  and larger. The curves in Fig. 2 illustrate how the temperature dependence of  $L_m$  depends on  $T_s$  and on the magnitude of the reduction in  $L_m$ . This reduction of  $L_m$  below  $L_0$  is caused by the contribution of vertical processes to the thermal resistivity and its magnitude depends on the angular distribution of scattering, which cannot be reliably predicted by our isotropic model. However, if the magnitude of  $L_m$  as  $T \to 0$  is determined experimentally, the increase of  $L_m$  as T increases indicates the value of  $T_s$  for localized spin fluctuations in the alloy.

For IrFe the electrical resistivity data<sup>14</sup> indicate that  $T_s \sim 30\,^{\circ}$ K, so this alloy should be suitable for checking the predicted increase of  $L_m$  with T. For RhFe the  $T^2$  dependence of electrical resistivity was not observed, <sup>15</sup> because the value of  $T_s$  is apparently much smaller (below 1.5  $^{\circ}$ K), so the linear T dependence of  $W_m$  may not be observable. We can make the interesting prediction that  $L_m$  should be approximately equal to  $L_0$  down to very low temperatures for RhFe if the spin-fluctuation model is applicable.

Fortunately, the lattice conductivity in the transition metals Pd and Pt is roughly an order of magnitude smaller than in the noble metals.  $^{12}$  It should be possible to obtain useful data for  $L_m$  in IrFe and RhFe if the lattice conductivity in these alloys is no larger than in the PdNi alloys  $^3$  discussed in Sec. III. To identify the effects of lattice conductivity on  $W_m$ , it is desirable to make measurements at several different impurity concentrations. Another possible complication is deviations from additivity of the different thermal resistivity components,  $^{16}$  although the constancy of the Lorenz number for the PdNi data of SSM suggests that, at least for this case, such deviations are not too significant.

We conclude that measuring the Lorenz number  $L_m$  for IrFe and RhFe should show whether the excitations producing the resistivities  $\rho_m$  and  $W_m$  are consistent with the low-energy localized spin fluctuations postulated to explain the temperature dependence of  $\rho_m$ .

# ACKNOWLEDGMENTS

I would like to thank Professor S. Doniach and Dr. W. J. Lawrence for helpful discussions.

# APPENDIX

In this Appendix we use the standard variational procedure<sup>3,17</sup> to calculate the thermal resistivity

 $W_m$  due to scattering of conduction electrons from localized spin fluctuations. To represent this scattering process we employ a simple isotropic two-band model, one band (s-like) contributing to the conductivity and the other (d-like) giving rise to magnetic properties. Then the thermal resistivity  $W_m$  is

$$W_{m} = \frac{27\hbar^{2}}{8\pi^{4}k_{F}^{4}k_{B}(k_{B}T)^{4}} \iint dk \, dk' \, f(\epsilon_{k})[1 - f(\epsilon_{k'})]$$

$$\times \int_{0}^{2k_{F}} q \, dq \, Q(\vec{k} - \vec{k}')(\omega^{2}k_{F}^{2} + \epsilon_{k}\epsilon_{k'}q^{2}) , (A1)$$

where  $Q(\vec{k} \rightarrow \vec{k}')$  is the scattering rate from filled conduction-electron state  $\epsilon_k$  to empty state  $\epsilon_{k'}$ ,  $f(\epsilon_k)$  is the Fermi-Dirac function,  $k_F$  is the Fermi wave vector for the conduction band, q is the magnitude of the change  $\vec{q}$  in the wave vector upon scattering:

$$\vec{q} = \vec{k}' - \vec{k} , \qquad (A2)$$

and  $\omega$  is the change in energy:

$$\omega = \epsilon_{k'} - \epsilon_{k} . \tag{A3}$$

As for electrical resistivity,  $^1$  (A1) is an average over initial and final states  $\vec{k}$  and  $\vec{k}'$  of the scattering rate  $Q(\vec{k} - \vec{k}')$ , but because of the contribution of vertical processes depending on energy change  $\omega$ , the weighting factor for  $Q(\vec{k} - \vec{k}')$  is not simply the change in forward momentum (which is proportional to  $q^2$ ).

Expressing  $Q(\vec{k} \rightarrow \vec{k}')$  in terms of the spectral density  $A(q, \omega)$  for localized spin fluctuations, <sup>1,2</sup> we obtain

$$\begin{split} W_{m} &= \frac{\rho_{0}}{L_{0}} \frac{3k_{B}\beta^{4}}{\pi^{2}} \int_{0}^{\infty} d\omega \, n(\omega) [\,\omega^{2}I_{0}(\omega)\overline{A}_{1}(\omega) \\ &+ \omega I_{1}(\omega)\overline{A}_{3}(\omega) + I_{2}(\omega)\overline{A}_{3}(\omega) \,] \,, \quad (A4) \end{split}$$

where  $\beta = (k_B T)^{-1}$ , and  $n(\omega)$  is the Bose function. As before,  $p_0$  is

$$\rho_0 = \left(\frac{JN(\epsilon_F)}{4}\right)^2 \frac{\nu}{n} \frac{m}{ne^2 \tau_F} , \qquad (A5)$$

where J is the coupling constant for scattering of conduction electrons from localized spin fluctuations,  $\nu$  and n are the number of atoms and conduction electrons, respectively, per unit volume,  $N(\epsilon_F)$  is the density of conduction electrons at the Fermi energy  $\epsilon_F$ , m is their effective mass, and  $\tau_F = \hbar/\epsilon_F$ . The  $\overline{A}_n(\omega)$  is the weighted average over wave vector of the spectral density:

$$\overline{A}_{n}(\omega) = (1/k_F^{n+1}) \int_0^{2k_F} dq \ q^n |F(q)|^2 A(q, \omega) ,$$
 (A6)

where F(q) is a form factor for the impurity d orbital, and

$$I_n(\omega) = \int_{-\infty}^{+\infty} d\epsilon_k f(\epsilon_k) [1 - f(\epsilon_k + \omega)] \epsilon_k^n. \tag{A7}$$

 $I_n(\omega)$  is easily evaluated for  $k_BT\ll\epsilon_F$ , giving

$$W_m = \frac{\rho_0 k_B \beta^4}{L_0} \int_0^{\infty} d\omega$$

$$imes rac{eta^{-2}\omega\overline{A}_{3}(\omega)+3\pi^{-2}\omega^{3}\overline{A}_{1}(\omega)-(2\pi^{2})^{-1}\omega^{3}\overline{A}_{3}(\omega)}{(e^{eta\omega}-1)(1-e^{-eta\omega})}$$
 . (A8)

We can give a physical explanation of the three terms in this expression. The first term, which includes in  $\overline{A}_3(\omega)$  the weighting factor  $q^2$  for angular scattering, represents the horizontal component of scattering, and so is the same as in the expression for electrical resistivity. The second term represents the vertical scattering component, having an additional energy-weighting factor of  $\omega^2$  but not including the  $q^2$  factor in  $\bar{A}_1(\omega)$ . The third term is a negative cross term having both the angular- and energy-weighting factors  $q^2$  and  $\omega^2$ , which arises as follows (cf. Ziman, <sup>17</sup> p. 390). In a scattering process in which the vertical and horizontal components are both large, the electron is changed from, say, a hot electron moving in one direction to a cold electron moving in the opposite direction, so there is little net effect on the heat current. Hence, the total resistivity is less than the sum of the vertical and horizontal components acting separately. Since the cross term has the same temperature dependence as the second term in (A8), we regard it as part of the vertical scattering component.

We introduce now the explicit form of the spectral density for localized spin fluctuations<sup>1</sup>

$$\overline{A}_n(\omega) = a_n \frac{\tilde{\omega}}{1 + \tilde{\omega}^2} \quad , \tag{A9}$$

where  $a_n$  is a constant independent of energy

$$a_n = 2c\alpha\delta u \left( \sqrt{2R} + \frac{\sqrt{2}}{\alpha\delta u} \frac{\overline{\chi}_{2I}}{\overline{\chi}_I} \right)$$
 (A10)

and

$$\tilde{\omega} = \omega / k_B T_S = \alpha \delta u \, \overline{\chi}_I . \tag{A11}$$

 $\delta U$  is the increase in the intra-atomic exchange interaction in the impurity cell, and  $\alpha$  is the susceptibility enhancement factor for localized spin fluctuations:

$$\alpha = (1 - \delta U \overline{\chi}_R)^{-1} \tag{A12}$$

and

$$_{n}\overline{\chi}_{2R} = (1/k_{F}^{n+1}) \int_{0}^{2k_{F}} dq \ q^{n} |F(q)|^{2} \chi_{R}^{2}$$
, (A13)

$$_{n}\overline{\chi}_{2I} = (1/k_{F}^{n+1}) \int_{0}^{2k_{F}} dq \, q^{n} |F(q)|^{2} 2\chi_{R}\chi_{I}$$
, (A14)

$$\overline{\chi}_R = (1/N) \sum_{\vec{q}} \chi_R$$
, (A15)

$$\overline{\chi}_I = (1/N) \sum_{\vec{n}} \chi_I , \qquad (A16)$$

where  $\chi_R$  and  $\chi_I$  are the real and imaginary parts of the generalized susceptibility  $\chi(q,\omega)$  in the host metal, and N is the total number of atomic cells.

Defining dimensionless variables

$$\tilde{W} = W_m L_0 / \rho_0 k_B a_3 , \qquad (A17)$$

$$\tilde{T} = T/T_{S} , \qquad (A18)$$

and substituting in (A8), we get

$$\tilde{W} = \tilde{W}_2 + \frac{3}{\pi^2} \left( \frac{a_1}{a_3} - \frac{1}{6} \right) \tilde{W}_4 ,$$
 (A19)

where

$$\tilde{W}_{n} = \frac{1}{\tilde{T}^{n}} \int_{0}^{\infty} d\tilde{\omega} \, \frac{e^{\tilde{\omega}/\tilde{T}} \tilde{\omega}^{n}}{(e^{\tilde{\omega}/\tilde{T}} - 1)^{2} (1 + \tilde{\omega}^{2})} \quad . \tag{A20}$$

Taking the limit  $\tilde{T} \rightarrow 0$ , we obtain a linear dependence on  $\tilde{T}$  corresponding to the  $\tilde{T}^2$  dependence of the electrical resistivity in this limit,

$$\tilde{W}(\tilde{T} \to 0) = \left[ \frac{\pi^2}{3} + \frac{4\pi^2}{5} \left( \frac{a_1}{a_3} - \frac{1}{6} \right) \right] \frac{T}{T_S} ,$$
 (A21)

which is equivalent to the result of SSM<sup>3</sup>. In the limit  $\tilde{T} \rightarrow \infty$ ,

\*Research supported by the Advanced Research Projects Agency through the Center for Materials Research at Stanford University, Stanford, Calif.

†Present address: Physics Department, Victoria University of Wellington, P. O. Box 196, Wellington, New Zealand.

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$$\tilde{W}(\tilde{T} \rightarrow \infty) = \frac{\pi}{2} + \frac{T_S^2}{T^2} \frac{3}{\pi^2} \left( \frac{a_1}{a_3} - \frac{1}{6} \right) \int_0^{\tilde{\omega}_m} \frac{\tilde{\omega}^2 d\tilde{\omega}}{(1 + \tilde{\omega}^2)}$$
(A22)

Hence, at high temperatures the horizontal component gives a constant resistivity, while the vertical component goes to zero as  $T^{-2}$ . Note that the general expression (A9) for the spectral density shape becomes inaccurate for large energies, especially for n=1. We introduce a cutoff to prevent an unphysical infinity in the integral of (A22), in which the  $\omega^2$  weighting factor for vertical processes enhances the contribution of higher energies. The magnitude of the vertical-process term above  $T_s$ as it decreases towards zero depends on the shape of the high-energy tail of the excitation spectrum and cannot be represented by a universal curve.

At high temperatures the local enhancement factor  $\alpha$  decreases, leading to a decrease of electrical resistivity below the linear law.  $^1$  Since  $a_3$  defined by (A10) is roughly proportional to  $\alpha$ , from (A17),  $W_m$  will reflect directly the decrease in  $\alpha$ , i.e., the normalizing factor for W will decrease at high temperatures. Thus, the temperature dependence of  $W_m$  is rather complex. After a linear increase with T at low temperatures,  $W_m$  becomes more or less constant above  $\frac{1}{2}T_s$ ; then, at still highter temperatures it decreases as  $\alpha$  decreases.

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