

well-defined gaps $\Delta_{2\alpha} > \Delta_{2\delta} > \Delta_{2\gamma} > \Delta_{2\beta}$. In the model N_α exhibits small decreases between thresholds; we have experimentally observed a slight increase ($\sim 5 \mu\text{eV}$) in $\Delta_{2\alpha}$ above the first threshold. \hat{I}_0 differs from I_0 by a phonon-trapping factor; this is consistent with the observed differences in threshold currents between samples with and without photoresist coatings. The absence of third- and fourth-gap states for injection at the gap edge in our experiments (as well as in Refs. 4 and 5) is also consistent with the model; the larger A_β which occurs in this case increases $I_{i\gamma}$, making observation of the third-gap state more difficult. Using the model and experimental data, we estimate the effective width of the boundary between regions to be $\sim 1 \mu\text{m}$. The theory of Scalapino and Huberman¹¹ indicates a length scale for the spatial inhomogeneity in Al of about $30 \mu\text{m}$, much larger than the boundary width, consistent with our assumption of uniform quasiparticle concentrations within regions. The boundary width is proportional to the phonon-trapping factor, consistent with the observed greater smearing of the gap structure in photoresist-coated samples.

The general agreement between the predictions of this model and our experimental observations supports our conclusion that we have observed an intrinsic quasiparticle diffusion instability toward a spatially inhomogeneous nonequilibrium superconducting state.

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¹G. A. Sai-Halasz, C. C. Chi, A. Denenstein, and D. N. Langenberg, *Phys. Rev. Lett.* **33**, 215 (1974).

²P. Hu, R. C. Dynes, and V. Narayanamurti, *Phys. Rev. B* **10**, 2786 (1974).

³J. J. Chang and D. J. Scalapino, *Phys. Rev. B* **10**, 4047 (1974).

⁴R. C. Dynes, V. Narayanamurti, and J. P. Garno, *Phys. Rev. Lett.* **39**, 229 (1977).

⁵K. E. Gray and H. W. Willemsen, *J. Low Temp. Phys.* **31**, 911 (1978).

⁶See also K. Hida, *Phys. Lett.* **68A**, 71 (1978).

⁷I. Iguchi, D. Kent, H. Gilmartin, and D. N. Langenberg, *Bull. Am. Phys. Soc.* **24**, 329 (1979).

⁸In this respect our samples differ significantly from those of DNG and GW. The three films in their samples were all of the same material and all three films were significantly perturbed by injection in their experiments.

⁹A similar effect has been reported in experiments on Pb films by I. Iguchi, *J. Low Temp. Phys.* **31**, 605 (1978), and **33**, 439 (1978).

¹⁰L. N. Smith, *J. Low Temp. Phys.* **28**, 519 (1977).

¹¹D. J. Scalapino and B. A. Huberman, *Phys. Rev. Lett.* **39**, 1365 (1977).

¹²A. Rothwarf and B. N. Taylor, *Phys. Rev. Lett.* **19**, 27 (1967).

¹³B. A. Huberman, *J. Chem. Phys.* **65**, 2013 (1976).

Electron-Phonon Enhancement of Electron-Electron Scattering in Al

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The influence of the electron-phonon interaction on electron-electron scattering in simple metals has been described within the framework of Landau Fermi-liquid theory. The predicted electron-electron scattering contribution to the low-temperature resistivity of Al is enhanced by a factor of ~ 20 by the electron-phonon interaction and is in excellent agreement with recent experiments.

The behavior of the ideal resistivity of Al at low temperatures has been the object of controversy for nearly a decade. Recently van Kempen and co-workers¹⁻³ have found that below 2 K $\rho_i(T)$

has a T^2 component which they interpreted as being due to electron-electron scattering, i.e., $\rho_i(T) \approx \rho_{ee}(T) = AT^2$, where $A = 2.8 \times 10^{-15} \Omega \text{ mK}^{-2}$. This result is qualitatively in accord with earlier

work by Garland and Bowers⁴ and Kaveh and Wisner⁵ but is at odds with the only published theoretical value, $A = 0.12 \times 10^{-15} \Omega \text{ mK}^{-2}$, due to Lawrence and Wilkins.⁶ Moreover, as has recently been pointed out,⁷ it is at first sight inconsistent with the absence of electron-electron scattering in the high- T Lorenz function of Al. In this Letter it will be shown that the phonon-mediated contribution to e - e scattering which may be neglected only above the Debye temperature⁸ but *has* been neglected in previous theory, dominates at low temperatures in Al. The theoretical treatment combines previous work on the Landau Fermi-liquid theory for transport phenomena in simple metals⁹⁻¹¹ with the s - p approximation introduced by Dy and Pethick¹² for the case of ³He.

According to Lawrence and Wilkins,⁶ the electron-electron scattering contribution to the electrical resistivity of a simple metal in the impurity-dominated regime is given by

$$\rho_{ee} = 2\pi^4 m^* \Delta / k_F^3 e^2 \tau_0, \quad (1)$$

where

$$\tau_0^{-1} = \frac{m^{*3} (k_B T)^2}{8\pi^4 \hbar^6} \left\langle \frac{\omega(\theta, \varphi)}{\cos\theta/2} \right\rangle, \quad (2)$$

$$\omega(\theta, \varphi) = \frac{1}{2} \omega_{\uparrow\uparrow}(\theta, \varphi) + \frac{1}{4} \omega_{\uparrow\downarrow}(\theta, \varphi), \quad (3)$$

$\omega_{\uparrow\uparrow}(\theta, \varphi)$ and $\omega_{\uparrow\downarrow}(\theta, \varphi)$ are the transition prob-

abilities for parallel- and opposite-spin scattering, respectively, and $\langle \dots \rangle$ denotes an average over solid angle. The parameter Δ appearing in Eq. (1) is a measure of the umklapp fraction for electron-electron scattering and can be estimated from pseudopotential theory (see Ref. 6). The determination of τ_0 , which is proportional to the lifetime of a quasiparticle at the Fermi surface, poses the main challenge in evaluating ρ_{ee} . In Eq. (2) the angle θ represents the angle between the initial momenta of the two quasiparticles in an electron-electron scattering event while φ represents the angle through which the relative momentum of these two quasiparticles is subsequently scattered. Equations (1)-(3) assume that the scattering amplitude is nearly independent of energy for energy transfers up to $\sim k_B T$. Because of the phonon-mediated contribution, the electron-electron scattering amplitude will have significant energy dependence on a scale of $\sim k_B \Theta_D$. For this reason Eqs. (1)-(3) are valid only in the limit $T \ll \Theta_D$ and this restriction will be implicit in the following discussion. For $\Theta_D \ll T \ll T_F$ only the Coulomb part of the scattering amplitude will contribute significantly to the resistivity and so Eqs. (1)-(3) again apply but with the understanding that electron-phonon interaction effects are not to be included in calculating the transition probabilities.

The transition probabilities in Eq. (3) are related to the four-point scattering function, ${}^0\Gamma$, by¹³

$$\omega_{\uparrow\sigma_2}(\theta, \varphi) = (2\pi/\hbar) |Z^2(k_F) {}^0\Gamma(\vec{p}_1\uparrow, \vec{p}_2\sigma_2; \vec{p}_1 + \vec{q}\uparrow, \vec{p}_2 - \vec{q}\sigma_2)|^2, \quad (4)$$

where \vec{p}_1 and \vec{p}_2 are the momenta of the incoming quasiparticles, \vec{q} is the momentum transfer for the scattering event, and the implicit energy arguments are to be evaluated at the Fermi energy.

My approximation for $\omega(\theta, \varphi)$ is based on the forward-scattering limit ($q = 0 = \varphi$) of ${}^0\Gamma$. In this limit¹¹

$$Z^2(k_F) {}^0\Gamma(\vec{p}_1\uparrow, \vec{p}_2\sigma_2; \vec{p}_1\uparrow, \vec{p}_2\sigma_2) = g(\vec{p}_1\uparrow, \vec{p}_2\sigma_2) + \frac{\pi^2 \hbar^2}{m^* k_F} (1 + F_0^s)^{-1} - \frac{\pi^2 \hbar^4 k_F Z}{3m^* M} \sum_{\lambda} \left\langle \left(\frac{\hat{\epsilon}_{\lambda}(\hat{q}) \cdot \hat{q}}{V_{\lambda}(\hat{q})} \right)^2 \right\rangle_{\text{av}}, \quad (5)$$

where $g(\vec{p}_1\uparrow, \vec{p}_2\sigma_2)$ is the scattering limit of the Landau interaction function, Z is the ionic charge, M is the ionic mass, m^* is the quasiparticle mass, and $\hat{\epsilon}_{\lambda}$ and V_{λ} are, respectively, the polarization vector and velocity for a phonon at long wavelength. The notation of Pines and Nozières¹⁴ is used for the Landau parameters. The second term in Eq. (5) is the forward-scattering limit of the long-range repulsive Coulomb interaction while the last term describes the long-range attractive interaction caused by induced variations in the ion density.¹⁵ The average in the last term of Eq. (5) is over directions of phonon propagation and is necessary in order to remove the crystal anisotropy, as reflected by the phonon dispersion, from the theory. The Landau interaction function can be expanded in terms of Landau parameters in the usual way as

$$g(\vec{p}_1\uparrow, \vec{p}_2\sigma_2) = \frac{\pi^2 \hbar^2}{m^* k_F} \left(\sum_l \frac{F_l^s}{1 + F_l^s / (2l + 1)} + \frac{\hat{\sigma}_2 F_l^a}{1 + F_l^a / (2l + 1)} \right) P_l(\cos\theta), \quad (6)$$

where the $\hat{\sigma}_2 = +1$ for $\sigma_2 = \uparrow$ and $\hat{\sigma}_2 = -1$ for $\sigma_2 = \downarrow$. Since the last term in Eq. (6) is readily calculated from experimental elastic constants, it follows that, in the forward-scattering limit, the scattering function on the Fermi surface may be expressed in terms of the system's Landau parameters and known quantities.

For convenience, I introduce the scattering amplitude $A(\theta, \varphi)$ defined by

$$\omega_{\uparrow\sigma}(\theta, \varphi) = (2\pi/\hbar) |A_{\uparrow\sigma}(\theta, \varphi)|^2. \quad (7)$$

Equations (5) and (6) specify $A(\theta, \varphi=0)$. To obtain the general $A(\theta, \varphi)$ I follow Dy and Pethick¹² in noting that if only s and p waves contribute to the q dependence of the scattering, the scattering amplitudes are completely specified by their $\varphi=0$ values,

$$A_{\uparrow\uparrow}(\theta, \varphi) = A_{\uparrow\uparrow}(\theta, \varphi=0) \cos \varphi, \quad (8)$$

$$A_{\uparrow\downarrow}(\theta, \varphi) = A_{\uparrow\downarrow}(\theta, \varphi=0) - A_{\uparrow\uparrow}(\theta, \varphi=0) \sin^2(\varphi/2). \quad (9)$$

This s - p approximation proved to be useful in its original applications to ³He and has recently been applied successfully to calculate transport properties of simple metals in a temperature range unaffected by the electron-phonon interaction.⁸ Since the scattering amplitude should be less q dependent for the shorter-ranged phonon-mediated electron-electron interaction than for the Coulomb interaction, the s - p approximation may be expected to be equally accurate in the present case.

The Landau interaction function in the presence of both electron-electron and electron-phonon interactions has been discussed in detail by Prange and Sachs.⁹ It follows from their analysis that

$$\frac{F_l^i}{1 + F_l^i/(2l+1)} = \frac{1}{(1+\lambda)} \frac{A_l^i}{[1 + A_l^i/(2l+1)]} + g_i, \quad i = a, s, \quad (10)$$

$$(1 + A_0^s)^{-1} + \sum_l \left(\frac{A_l^s}{1 + A_l^s/(2l+1)} + \frac{A_l^a}{1 + A_l^a/(2l+1)} \right) = 0 \quad (13)$$

to be satisfied. The parameter g_0 is related to λ by

$$g_0 = \lambda/(1+\lambda). \quad (14)$$

The values of λ recommended by Grimvall¹⁸ are used and g_1 is fixed by requiring

$$(1 + F_0^s)^{-1} + \sum_l \left(\frac{F_l^s}{1 + F_l^s/(2l+1)} + \frac{F_l^a}{1 + F_l^a/(2l+1)} \right) - C^{ep} = 0, \quad (15)$$

where A_l^i is the Landau parameter in the absence of the electron-phonon interaction, λ is the electron-phonon mass-enhancement factor, and g_l is the Legendre polynomial moment of the phonon-mediated scattering function discussed, for example, by Rice.¹⁶ For many simple metals, the higher moments of g_l will have considerable anisotropy¹⁷; the value to be used in Eq. (10) in this theory is the average over the Fermi surface. In any case, we will see that only the $l=0$ and $l=1$ moments which have less anisotropy will enter the final approximation for the scattering amplitudes. Recall as well that

$$m^* = m_{ee}^*(1+\lambda), \quad (11)$$

where m_{ee}^* is the quasiparticle mass in the absence of the electron-phonon interaction. To determine m_{ee}^* and A_l^i I approximate the simple metal by a uniform electron gas with the electron mass modified to m_b and the electron-electron interaction reduced by a factor ϵ so as to account for the crystalline effect on the kinetic energy and the polarizability of the core electrons. This leads to⁸

$$A_l^i = A_{l(h)}^i(m_b r_s/\epsilon), \quad (12a)$$

$$m_{ee}^* = m_b \left[1 + \frac{1}{3} A_1^s(h)(m_b r_s/\epsilon) \right], \quad (12b)$$

where r_s is the usual conduction-electron density parameter and $A_{l(h)}^i(r_s)$ is Landau parameter of the homogeneous electron gas with density parameter r_s . The values used for m_b and ϵ are discussed in Ref. 8. In evaluating Eq. (6) I have truncated the sum over l at $l=1$. This is justified because the higher moments of the Landau scattering function are expected to have little influence on properties such as the resistivity which measure some sort of average scattering rate. Comparing Eqs. (5) and (6) it is seen that F_0^s does not enter the forward-scattering limit of the scattering function while A_1^s and A_0^a are available via Eqs. (12) from calculations of the homogeneous-electron-gas quasiparticle mass and spin susceptibility, respectively.⁸ A_1^a is fixed by requiring the sum rule¹¹

TABLE I. Electron-electron scattering contributions to the electrical and thermal resistivities of simple metals. ρ_{ee} was calculated from Eq. (1) and W_{ee} from a similar formula given in Ref. 6. The columns labeled *CO* and *PM* correspond, respectively, to neglecting and including the phonon-mediated contribution to the scattering amplitudes.

Metal	W_{ee}/T (10^{-7} m/W)		ρ_{ee}/T^2 (10^{-15} Ω mK $^{-2}$)		L_{ee}/L_0		C_{ep}^a	Δ^b
	<i>CO</i>	<i>PM</i>	<i>CO</i>	<i>PM</i>	<i>CO</i>	<i>PM</i>		
Al	0.29	7.1	0.18	4.1	0.25	0.24	3.98	0.4
Pb	0.37	26	0.27	16	0.30	0.25	4.27	0.4
Cu	2.2	2.4	2.0	1.9	0.37	0.32	0.22	0.55
Ag	2.3	2.1	2.0	1.7	0.36	0.33	0.25	0.55
Au	2.9	3.8	2.5	3.0	0.35	0.32	0.15	0.55

^aThese values were calculated using the low-temperature elastic constants given by Kittel (Ref. 19).

^bFor Al, Δ was taken from Ref. 6 while for the noble metals the chosen value is the average of the estimates of Lawrence (Ref. 20) and Black (Ref. 21). The estimate for Pb was obtained by combining the formulas of Ref. 6 and a Heine-Abarenkov pseudopotential (Ref. 22).

where

$$C_{ep} = \frac{k_F^2}{3m^*M} \sum_{\lambda} \left\langle \left(\frac{\hat{\epsilon}_{\lambda}(\hat{q}) \cdot \hat{q}}{V_{\lambda}(\hat{q})} \right)^2 \right\rangle_{av}. \quad (16)$$

The sum rules, Eqs. (13) and (15), reflect the requirement that the parallel-spin scattering amplitude be zero when $\theta = \varphi = 0$ both with and without the electron-phonon interaction.

In Table I the value for the coefficient of T^2 in the low-temperature resistivity of Al predicted by the above approach is listed together with the value which would be predicted if the electron-phonon interaction were not included in the analysis. For the purpose of comparison the corresponding thermal resistivity of Al, W_{ee} , as well as thermal and electrical resistivities for several other cubic simple metals have been included. From Table I, we see that ρ_{ee} in Al is enhanced by a factor of ~ 20 at low temperatures because of the electron-phonon interaction. Of this a factor of ~ 2 may be ascribed to the electron-phonon enhancement of the quasiparticle mass which enters ρ_{ee} as m^{*2} . The remaining factor of ~ 10 reflects the fact that the attractive phonon-mediated electron-electron interaction is roughly speaking about four times stronger than the repulsive Coulomb interaction in Al. The numerical value for ρ_{ee}/T^2 is only $\sim 40\%$ higher than the recent experimental result of van Kempen *et al.*¹⁻³ Moreover, the predicted L_{ee}/L_0 ($L_{ee}/L_0 \equiv \rho_{ee}/L_0 W_{ee} T$) which, like ρ_{ee} , is roughly proportional to Δ , is also $\sim 40\%$ larger than the value measured by Garland *et al.*⁴ We therefore expect that most of the error in ρ_{ee} is

due to an overestimate of the value for Δ .

To illustrate the expected manifestations of this effect in other simple metals we have presented in Table I the results for the distinctly different cases of Pb and the noble metals. In Pb, which has a strong electron-phonon interaction, ρ_{ee} is enhanced by a factor of ~ 60 . However, because of the large T_c/Θ_D ratio the resistivity is likely to be dominated by electron-phonon scattering immediately above T_c and it is not likely that the enhanced ρ_{ee} can be directly observed. We expect the electron-phonon enhancement of electron-electron scattering to be most clearly observable in the superconducting simple metals with low T_c/Θ_D ratios. In the noble metals, on the other hand, the Coulomb repulsion is stronger than the attractive phonon-mediated interaction so that the total effective interaction is weakened. The competition between this reduction factor and the mass-enhancement factor leads to largely unchanged values for ρ_{ee} . This prediction is consistent with the comparison of high-temperature Lorenz function and low-temperature resistivity data in the noble metals.¹⁷ The temperature dependence of the effects discussed here will be treated in a future publication.²³

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¹H. van Kempen, J. H. J. M. Ribot, and P. Wyder, J. Phys. (Paris) Colloq. **39**, C6-1048 (1978).

- ²J. H. J. M. Ribot, J. Bass, H. van Kempen, and P. Wyder, *J. Phys. F* **9**, L117 (1979).
- ³J. H. J. M. Ribot, J. Bass, H. van Kempen, and P. Wyder, to be published.
- ⁴J. C. Garland and R. Bowers, *Phys. Kondens. Mater.* **9**, 36 (1969).
- ⁵M. Kaveh and N. Wiser, *Phys. Lett.* **51A**, 89 (1975).
- ⁶W. E. Lawrence and J. W. Wilkins, *Phys. Rev. B* **7**, 2317 (1973).
- ⁷A. H. MacDonald and M. J. Laubitz, *Phys. Rev. B* (to be published).
- ⁸A. H. MacDonald and D. J. W. Geldart, to be published.
- ⁹R. E. Prange and A. Sachs, *Phys. Rev.* **158**, 672 (1967).
- ¹⁰V. Heine, P. Nozières, and D. Wilkins, *Philos. Mag.* **13**, 741 (1966).
- ¹¹W. F. Brinkman, P. M. Platzman, and T. M. Rice, *Phys. Rev.* **174**, 495 (1968).
- ¹²K. S. Dy and C. J. Pethick, *Phys. Rev.* **185**, 373 (1969).
- ¹³P. Nozières, *Theory of Interacting Fermi Systems* (Benjamin, New York, 1964).
- ¹⁴D. Pines and P. Nozières, *The Theory of Quantum Liquids* (Benjamin, New York, 1966).
- ¹⁵L. J. Sham and W. Kohn, *Phys. Rev.* **145**, 561 (1966).
- ¹⁶T. M. Rice, *Phys. Rev.* **175**, 858 (1968).
- ¹⁷C. R. Leavens and J. P. Carbotte, *Can. J. Phys.* **51**, 398 (1973).
- ¹⁸G. Grimvall, *Phys. Scr.* **14**, 63 (1976).
- ¹⁹C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1971).
- ²⁰W. E. Lawrence, *Phys. Rev. B* **13**, 5316 (1976).
- ²¹J. E. Black, *Can. J. Phys.* **56**, 708 (1978).
- ²²W. A. Harrison, *Pseudopotentials in the Theory of Metals* (Benjamin, New York, 1966).
- ²³A. H. MacDonald and D. J. W. Geldart, to be published.

Spin-Density-Wave Fluctuations in Copper Nuclear Spins

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A spherical-model calculation of the spin-density-wave fluctuations in copper nuclear spins is presented. The results are compared with an experiment where copper nuclear spins have been cooled to below $0.1 \mu\text{K}$. The agreement, with no adjustable parameters, is found to be satisfactory for spin susceptibility, entropy, and internal energy. A considerable depression of the transition temperature from its mean-field value, in qualitative agreement with the experiments, is also found.

The thermodynamic properties of nuclear spins cooled to temperatures of the order of tenths of a microkelvin become interesting for two reasons.¹ On the one hand, the spin-spin interaction energy becomes comparable to temperature and one might expect the nuclear spins to undergo a phase transition into an ordered state. Such a condensed state would be the lowest-energy many-body state reached so far. On the other hand, since the interaction between nuclear spins is known, one expects their thermodynamic properties to be predictable. In other words, these systems become "ideal systems" on which theoretical techniques can be examined for their quantitative accuracy.

Copper nuclear spins interact via the magnetic dipole-dipole interaction and via the conduction-electron-mediated Ruderman-Kittel (RK)² ex-

change interaction. Whereas the former interaction is overall ferromagnetic, the latter is oscillatory and in copper, overall antiferromagnetic. The resulting ground-state spin arrangement has been studied by Kjälman and Krukijärvi³ for varying strength of the exchange interaction. For the strength appropriate for copper, they find a spin-density wave (SDW) state. This paper is an account of the thermodynamic effects of the SDW fluctuations. We calculate the susceptibility, the entropy, and the internal energy of the copper nuclear spins. We also discuss the depression of the transition temperature caused by the same fluctuations. The calculations have been done within the spherical model⁴ which is known to be exact for a large coordination number⁵ or a large number of the components of the order parameter.⁶ Copper nuclear spins interact with