Optical and Thermal Effective Masses of Copper

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The optical and thermal masses of copper have been calculated from a phase-shift model of the Fermi surface and Fermi velocity. The total area of the Fermi surface is also determined. The results yield a Fermi-surface average of the renormalization of the band velocity in copper by the electron-phonon interaction, and suggest that the quasiparticle current in copper is reduced by quasiparticle interactions.

I. INTRODUCTION

The free-electron model is frequently used to describe experimental observations concerning the conduction electrons in metals. The deviations of the measured values of various properties from the free-electron values are described by defining an "effective" mass to replace the bare-electron mass that enters in free-electron theory.

The thermal effective mass is a measure of the low-temperature heat capacity of the conduction electrons in a metal, and can be calculated from a knowledge of the density of states at the Fermi energy. For a metal of arbitrary Fermi surface, the thermal mass is given by[1]

$$m_{\rm th}/m^0 = (S_F/S_F^0)\, V_F^0 \langle 1/V_F^q \rangle , \tag{1}$$

where S_F is the surface area of the Fermi surface, V_F^q is the quasiparticle velocity which is renormalized by the electron-electron and electron-phonon interactions, the superscript zero denotes quantities calculated from the free-electron model, and the angular brackets indicate a Fermi-surface average of the argument.

The optical effective mass is a measure of the contribution of the conduction-band electrons to the real part $\epsilon_1(\omega)$ of the dielectric constant of a metal. The dielectric constant can be written as[2]

$$\epsilon_1(\omega) = 1 - (4\pi Ne^2)/m_{\rm opt}(\omega^2 + \tau^{-2}) + \epsilon_{1i}(\omega), \tag{2}$$

where, for a metal with an arbitrary Fermi surface, the optical mass $m_{\rm opt}$ is given by[1]

$$\frac{m_{\rm opt}}{m} = \frac{S_F^0 V_F^0}{S_F \langle V_F^b/(1+\nu)\rangle} , \tag{3}$$

where V_F^b is the band-structure Fermi velocity which is renormalized by the electron-electron, but not by the electron-phonon, interaction, and ν is a parameter which describes the influence of quasiparticle interactions on the current associated with a quasiparticle state. In the spherical approximation, ν is equal to the Landau parameter A_1^c. The second term in Eq. (2), which is the dominant term in the near-infrared region of the spectrum, represents the contribution of intraband transitions to the dielectric constant. The third term represents the contribution of interband transitions. The quasiparticle and band velocities are related by

$$V_F^b = V_F^q (1+\lambda), \tag{4}$$

where $\lambda(\vec{k})$ is an anisotropic parameter which measures the renormalization of the one-electron energy bands by the electron-phonon interaction.

In this paper we present a calculation of the thermal and optical masses of copper, using a phase-shift model of the Fermi surface. The surface area of the Fermi surface is also calculated. The thermal mass is calculated by direct evaluation of Eq. (1), which yields a result in good agreement with the experimental value derived from specific-heat data. Our calculation also yields a Fermi-surface average of the electron-phonon renormalization parameter λ. A comparison between our calculation of the optical mass and the experimental data yields an estimate of the Landau parameter A_1^c.

The electronic phase shifts at the Fermi surface of copper have been discussed elsewhere.[3] Recent high-precision measurements of the de Haas–van Alphen frequencies[4] call for some slight revision of the earlier results, which we discuss in Sec. II. In Sec. III we describe the way in which the effective masses were calculated, and in Sec. IV we present the results of our calculations and discuss their significance.

II. PHASE-SHIFT MODEL

The Fermi surface of a metal such as copper is distorted by the electron-ion interaction, which in a phase-shift analysis is represented by a short-range potential of muffin-tin form. The electronic energy levels $E_n(\vec{k})$ are roots of the secular equation

$$\det\{[(\vec{k}+\vec{g})^2 - E]\delta_{\vec{g}\vec{g}'} + \Gamma_{\vec{g}\vec{g}'}(\vec{k}, E, \eta_l(E))\} = 0, \tag{5}$$

where $\vec{g}, \vec{g}'$ are reciproacl-lattice vectors, and the augmented-plane-wave (APW) pseudopotential Γ is a function of the phase shifts $\eta_l(E)$. The explicit form of Γ has been discussed elsewhere.[3]

TABLE I. Comparison of de Haas–van Alphen frequencies computed from the phase-shift model with experimental data of Coleridge *et al.* (units 10^8 G).

Orbit	Notation	Experimental[a]	Phase-shift model[b]
Belly	B_{100}	5.9955 (1)	5.9954 (1)
Belly	B_{111}	5.8073 (1)	5.8073 (1)
Neck	N_{111}	0.21736 (1)	0.21737 (1)
Rosette	R_{100}	2.4604 (1)	2.4602 (1)
Dog's bone	D_{110}	2.5095 (1)	2.5095 (1)
Turning point near ⟨100⟩ in (110)	B_T	5.9558 (1)	5.9558 (1)

[a]Reference 4. [b]Present work.

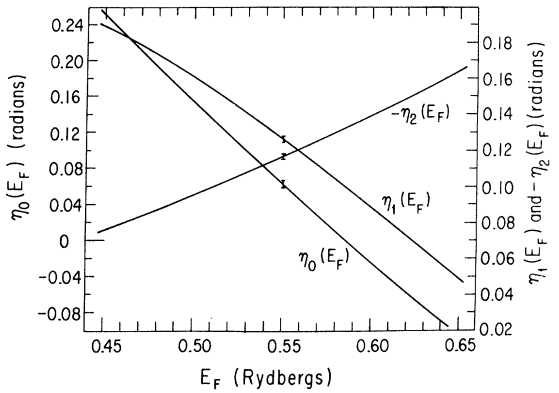

FIG. 1. Phase shifts derived by fitting experimental Fermi-surface data, plotted as a function of the assumed value of the Fermi-energy parameter E_F. The corresponding value of the f phase shift is $\eta_3 = 0.00075$ rad, and is assumed to be independent of E_F.

The phase shifts $\eta_l(E_F)$ that describe the electron-ion interaction at the Fermi surface of copper have been determined by fitting the surface of constant energy $E(\vec{k}) = E_F$ computed from Eq. (5) to experimental Fermi-surface data. In addition, a nonlocal modification of the one-electron potential due to Chodorow[5] was constructed to yield phase shifts fully in agreement with those determined from the Fermi-surface data. The experimental data on which these calculations were based yield Fermi-surface areas accurate to about 1 part in 10^3, and were analyzed in a 30-APW calculation. More recent data[4] yield Fermi-surface areas (reproduced in Table I) accurate to better than 1 part in 10^4, and justify a more accurate calculation of the phase shifts.

A 60-APW calculation was carried out to determine the s, p, and d phase shifts that give an exact fit to the experimental areas B_{100}, B_{111}, and N_{111}, for several values of the f phase shift and the Fermi-energy parameter E_F. The computed areas of the orbits D_{110}, R_{100}, and B_T were then compared with the experimental areas. Agreement was found to depend rather sensitively on the assumed value of the f phase shift, but only weakly on values of E_F in the range we investigated. Even though E_F is essentially a free parameter in our fit to the Fermi-surface data, our best estimates of the phase shifts depend sensitively on the assumed value of E_F, as plotted in Fig. 1. The phase shifts presented here are consistent with, but somewhat more accurate than, those obtained from the earlier data. The uncertainties attached to the phase shifts indicate the accuracy with which the phase shifts can be determined from the data for any assumed value of E_F. These uncertainties are dominated by the fact that the fit to the data determines the f phase shift within only a rather broad range of values, and an order-of-magnitude improvement in the absolute accuracy of the areas yields only a rather slight improvement in the accuracy with which the phase shifts can be determined.

In constructing a nonlocal modification of the Chodorow potential to reproduce the revised phase shifts, it is convenient to choose the same value of the Fermi-energy parameter ($E_F = 0.55$ Ry) as was assumed in the earlier calculations. We find the set of phase shifts listed in Table II. The angular-momentum-dependent corrections to the Chodorow potential required to reproduce these phase shifts are set out in Table III. In Fig. 2 we display the energy dependence of the phase shifts for copper calculated from the modified Chodorow potential. Comparable curves for the Chodorow potential have been given by Cooper, Kreiger, and Segall.[6] The accuracy of the extrapolation of the phase shifts away from the Fermi energy (where they are determined by the Fermi-surface data) depends largely on the accuracy of the Chodorow potential. Several authors have found that this potential gives satisfactory predictions of interband-transition frequencies in copper, and we take this as evidence that the modified Chodorow potential yields accurate energy bands in the vicinity of the Fermi level.

The revisions discussed here do not significantly alter the results for the anisotropic band velocity of copper presented in a previous paper.[7] However, the experimental value of the cyclotron mass from which the quasiparticle velocity on the neck

TABLE II. Phase shifts (in radians) derived from Fermi-surface data for copper for several values of E_F, and comparison with earlier calculations (Ref. 3) for $E_F = 0.55$ Ry.

	Present work			Ref. (3)
E_F (Ry)	0.50	0.55	0.60	0.55
η_0	0.1549	0.0638	−0.0232 (30)	0.0574 (42)
η_1	0.1600	0.1261	0.0882 (9)	0.1234 (14)
η_2	−0.0952	−0.1168	−0.1399 (5)	−0.1157 (9)
η_3	0.00075	0.00075	0.00075 (20)	0.00135 (30)

TABLE III. Angular-momentum-dependent correction to the Chodorow potential required to bring the phase shifts at energy $E_F = 0.55$ Ry into agreement with those derived from the Fermi-surface data. The correction is a constant $V(l)$ within the muffin-tin spheres, and zero outside.

l	$V(l)$ (Ry)
0	-0.0164 ± 0.0021
1	-0.0396 ± 0.0014
2	-0.0074 ± 0.0013
3	$+0.0514 \pm 0.0650$
4	0

was derived is now known to be too large. A recent experiment[8] yields the value

$$(m_c/m) = 0.455 \pm 0.006 \quad \text{(on neck)}$$

leading to a quasiparticle velocity

$$(V_F/V_F^0) = 0.418 \pm 0.006 \quad \text{(on neck)}.$$

This revised result is in better agreement with the result of a direct measurement of the neck velocity by Doezema and Koch,[9]

$$(V_F/V_F^0) = 0.427 \pm 0.006 \quad \text{(on neck)}.$$

III. METHOD OF CALCULATION

The phase-shift model of the Fermi surface of copper yields three quantities that are used in the present calculations: the Fermi radius k_F with an absolute accuracy of better than 0.01%, the quasiparticle velocity V_F^q with an absolute accuracy of about 2%, and the band velocity V_F^b with a computational accuracy of about 0.2%.

The quasiparticle velocity in copper was determined by constructing a surface of constant energy enclosing the Fermi surface, such that the differences between the areas of extremal orbits on the two surfaces are proportional to the experimental cyclotron masses. Then the local velocity at any point on the Fermi surface can be computed from the normal distance between the two surfaces of constant energy. Band velocities for copper were determined from the energy bands calculated from the modified Chodorow potential. Since band gaps predicted by this potential have been shown to be in satisfactory agreement with experimental measurements of certain optical transition frequencies, we believe that the dominant effects of the electron-electron interaction are folded into the band velocities determined in this way.

In the present analysis the phase-shift model was used to calculate the radii of the Fermi surface as a function of the radial coordinates θ and ϕ. The Fermi surface of copper has 48-fold symmetry, so the calculations were confined to a sector encompassing $\frac{1}{48}$th of the Brillouin zone, within which the radii were calculated at 150 evenly distributed points. The Fermi-surface averages in Eqs. (1) and (3) were calculated by dividing the Fermi surface into triangular elements whose vertices are three nearest-neighbor radii, as in Fig. 3. Each surface element is approximated by a spherical triangle, of radius $R = \frac{1}{3}(R_1 + R_2 + R_3)$, centered on the origin of the Brillouin zone and encompassing the same solid angle as the surface element. In this approximation the area of a single surface element is given by

$$\Delta S = R^2(\theta_1 + \theta_2 + \theta_3 - \pi)/\cos\psi\,, \tag{6}$$

where the vertex angles θ_1, θ_2, and θ_3 of the equivalent spherical triangle are calculated by standard techniques in analytical geometry. ψ is the angle between the unit vector normal to the surface and the mean-radius vector, and corrects for the tilt of the surface element. Close to the necks, the contributions to the Fermi-surface integrals were conveniently evaluated by dividing the surface into cylindrical, rather than triangular, elements. The Fermi-surface averages of the Fermi velocity and the inverse Fermi velocity, which enter in Eqs. (1) and (3), were evaluated by summing the appropriate integrands over all surface elements. The Fermi-surface area was evaluated by summing the area of the individual elements. The convergence of the calculation was checked by performing

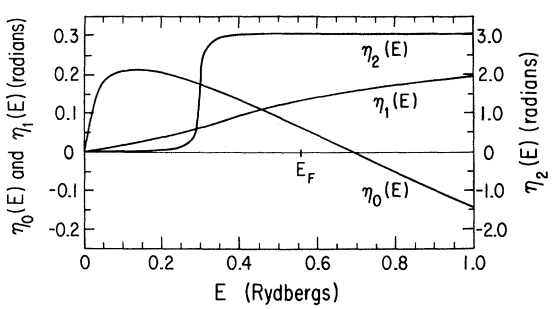

FIG. 2. Phase shifts calculated as a function of energy E from the modified Chodorow potential.

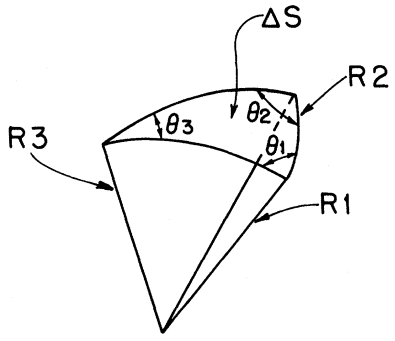

FIG. 3. Illustration of spherical triangle with pertinent variables.

the calculation with distribution of 50 and 150 points, and the results agreed within the limit of error of the calculation.

IV. RESULTS AND DISCUSSION

The total surface area of the Fermi surface of copper was found to be

$$S_F/S_F^0 = 0.982 \pm 0.002 \ . \tag{7}$$

It is reduced below the surface area of the free-electron sphere ($S_F^0 = 23.345$ Å^{-2}) by the presence of the necks.

By evaluating the expression in Eq. (1), we find for the thermal mass of copper

$$m_{\text{th}}/m = 1.41 \pm 0.03 \ . \tag{8}$$

This result is consistent with the results of experimental specific-heat measurements,[10–15] as given in Table IV, from which we derive the best estimate

$$m_{\text{th}}/m = 1.385 \pm 0.005 \quad \text{(experimental)}. \tag{9}$$

It is also consistent with independent calculations from the quasiparticle velocities, and this agreement serves as a check on our calculational procedure.

We have also evaluated the expression in Eq. (1), replacing the quasiparticle velocity by the band velocity and neglecting the anisotropy of λ. We find

$$m_{\text{th}}/m = (1.26 \pm 0.01)(1+\lambda), \tag{10}$$

and by comparing Eqs. (9) and (10) we deduce that the Fermi-surface average of the electron-phonon renormalization parameter is

$$(1+\lambda) = 1.10 \pm 0.01 \ . \tag{11}$$

Grimvall[16] has estimated lower and upper bounds on the electron-phonon enhancement of the thermal mass. From electrical resistivity and superconductivity data he finds that $(1+\lambda)$ must lie between 1.07 and 1.24. The electron-phonon enhancement is apparently far from negligible and our value of 1.10 is consistent with Grimvall's result.

TABLE IV. Comparison of experimental specific-heat masses with masses calculated by inversion of cyclotron-mass data.

Specific heat	
Martin[a]	1.383 ± 0.002
Osborne[b]	1.389 ± 0.006
Manchester[c]	1.392 ± 0.006
Griffel[d]	1.382 ± 0.006
Cyclotron mass	
Halse[e]	1.40 ± 0.03
Stark and Auluck[f]	1.40 ± 0.01
Present work	1.41 ± 0.03

[a]Reference 10. [b]Reference 11. [c]Reference 12. [d]Reference 13. [e]Reference 14. [f]Reference 15.

TABLE V. Comparison of first-principles calculations of the Landau parameter A_1^c for $r_s = 2.66$, with the result derived from the optical mass of copper.

Nozieres & Pines approx.[a]	+0.03
Hubbard approx.[a]	0.00
Screened exchange[b]	−0.045
Rice[b]	+0.020
Expansion method[b]	−0.031
Lee and Nowak[c]	(−0.07 ± 0.05)

[a]Reference 18. [b]Reference 19. [c]Reference 17.

From Eqs. (1), (3), and (4) it follows that, neglecting the anisotropy of λ and ν, the thermal and optical masses are related by

$$(m_{\text{th}}/m_{\text{opt}}) = (S_F/S_F^0)^2 \langle V_F^b \rangle \langle 1/V_F^b \rangle (1+\lambda+\nu). \tag{12}$$

Schwartz's inequality for the Fermi-surface averages yields

$$(m_{\text{th}}/m_{\text{opt}}) \geq (S_F/S_F^0)^2 \ (1+\lambda+\nu). \tag{13}$$

Our best estimate of the experimental optical mass of copper[17] is

$$(m_{\text{opt}}/m) = 1.39 \pm 0.06. \tag{14}$$

Inserting Eqs. (7), (10), (11), and (14) into Eq. (13) yields an upper limit for the parameter

$$\nu \leq -0.02. \tag{15}$$

In deriving this upper limit, we have allowed for the substantial uncertainty in our best estimate of the optical mass. This upper limit is consistent with the value of ν, previously estimated by the authors to be[17]

$$\nu = -0.07 \pm 0.05. \tag{16}$$

We have pointed out above that in the spherical approximation ν reduces to the Landau parameter A_1^c. A comparison between this result and first-principles calculations[18,19] of A_1^c is set out in Table V. The first-principles calculations vary substantially among themselves, but are qualitatively consistent with our result. The microscopic calculations assume an electron fluid in equilibrium with a uniform background of positive charge, thereby neglecting band-structure effects. Thus the results of the microscopic calculation are not strictly comparable with our result, and the most one can expect is qualitative agreement.

V. CONCLUSIONS

A phase-shift analysis of the most recent experimental Fermi-surface data for copper calls for a slight revision of earlier results. The principal change is a reduction in our best estimate of the f phase shift. For a given value of the Fermi-energy

parameter E_F, the revised phase shifts are numerically closer to the results of the Korringa-Kohn-Rostoker (KKR) calculation of Cooke, Davis, and Wood,[20,21] which suggests that the changes are in part a consequence of the more complete convergence of the present calculations.

A substantial increase in the accuracy of the experimental data yields only a slight increase in the accuracy of the phase shifts. This is because the f phase shift is rather poorly determined by fitting the Fermi-surface data, and uncertainty in the f phase shift is reflected in our best estimates of the other phase shifts. A least-squares fit to all the area data would yield more accurate phase shifts, but with present techniques such a calculation would be prohibitively costly.

The results presented in Table I suggest that a four-phase-shift model is capable of reproducing the area data with an accuracy of about 1 part in 10^4. Even though an analysis of more complete experimental data would be needed to substantiate this claim, phase-shift analysis is evidently a rather accurate technique for the inversion of Fermi-surface data.

Our calculation of the thermal and optical masses of copper was undertaken to test the velocity distributions reported in a previous paper.[7] The consistency between our calculated thermal mass and the specific-heat data provides a check on the quasiparticle velocities, although the check is perhaps less convincing in view of a discrepancy for divalent metals noted recently by Stark and Auluck.[15] The Fermi-surface average of λ is consistent with independent estimates by Grimvall.[16] This provides some evidence for the correctness of the band velocities, although the range of values of λ that is consistent with Grimvall's results is unfortunately rather wide.

In the calculations described here we have assumed that the electronic energy bands calculated from the modified Chodorow potential are fully renormalized by the Coulomb (i.e., electron-electron) interaction. Several arguments may be advanced to support this assumption:

(i) The Chodorow potential was constructed to fit the empirical spectrum of the free copper ion, and presumably includes the dominant effects of exchange and correlation within the ion core.

(ii) The modified potential has been adjusted to fit Fermi-surface data exactly, and it is known that in principle the Coulomb renormalization of the conduction bands can be folded into a nonlocal effective potential.

(iii) Energy gaps calculated from the modified potential are in good agreement with the corresponding optical-transition frequencies, which depend only on the difference between the Coulomb-renormalized energies in the initial and final states.

We cannot rule out the possibility that the energy bands calculated from the modified Chodorow potential may be less than fully renormalized by the Coulomb interaction, a possibility that has been stressed by Christiensen.[22] If this were so, then our value of λ could not be interpreted as determined by the electron-phonon interaction alone, and our value of ν would represent only a fraction of the full Coulomb renormalization. The true λ would necessarily be more positive, and the true ν more negative than we found, in such a way that the sum of λ and ν should remain unchanged. Nevertheless, our optical-mass calculation attests to the consistency of our assumption that the energy bands derived from the modified Chodorow potential are fully renormalized by the Coulomb interaction, and appears to rule out the possibility that they can be regarded as the "bare" band structure.

The thermal and optical masses are related by an inequality that depends on parameters of the Fermi surface. It has been suggested that information about (S_F/S_F^0), and hence the topology of the Fermi surface, can be deduced from the ratio of these masses. Unfortunately the many-body corrections λ and ν confuse the interpretation, as is evident from Eq. (13). Several authors have noted inconsistencies in applying this inequality to the data for copper. Our calculations show that if the integrals are evaluated carefully, taking into account the anisotropies of the Fermi surface and the velocity distributions, then one obtains estimates of the total surface area and many-body corrections that are consistent with the inequality.

In summary, the energy bands derived from the modified Chodorow potential, when interpreted as being largely or fully renormalized by the Coulomb interaction, are consistent with the Fermi-surface data and optical-transition frequencies, with the thermal and optical masses, and with what is known about many-body corrections to the one-electron energy bands of metallic copper.

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Thermal and Electrical Conductivity of Pure Tin from 4.5 to 77 °K†

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The thermal and electrical conductivities of two pure single crystals of tin have been measured. The samples were oriented at 72° and 6° with respect to the tetragonal axis of tin. The thermal conductivity values for the two samples were fit to the relation $(\alpha T^n+\beta/T)^{-1}$ from 4 to 12 °K with n approximately 3.2 for both samples. This large value is attributed to dispersion in the phonon spectrum which causes an approximate 4.2-power dependence with temperature of the specific heat over the same temperature range. The electrical-resistivity value of the two pure samples was found to obey the Bloch-Grüneisen expression over a wide range with a Debye temperature of 125 °K and with a small additional residual resistivity. The anisotropy of the thermal conductivity exhibits a slight maximum at 10 °K, attributable to the relative effect of impurity scattering and an anisotropic band structure. The anisotropy of the electrical conductivities exhibits a more pronounced maximum around 20 °K because of an additional effect of area differences of Brillouin-zone segments on the Fermi surface when small-angle scattering dominates. The ratio of electrical anisotropy to thermal anisotropy at the maximum was found to be approximately 1.27, which compares well with the theoretically predicted value of 1.26 from the area differences at the zone segments in the 90° and 0° orientations.

I. INTRODUCTION

This paper reports the results of thermal and electrical-conductivity measurements on two pure single crystals of tin from 4.5 to 77 °K. These two samples had different crystallographic orientations with respect to the tetragonal axis of tin, one being nearly parallel (6°) and the other nearly perpendicular (72°). A variable-temperature cryostat consisting basically of a thermally isolated Swenson-type heat exchanger[1] cooled by circulating helium vapor was employed for the measurements. The thermal conductivity of the samples was measured by the longitudinal-heat-flow method. The temperature gradient was determined by calibrated germanium resistors. The electrical resistivity of both samples was measured potentiometrically along the identical length as for the thermal-conductivity measurements. An evaluation of the electrical-resistivity anisotropy defined by $a_E = \rho(\parallel)/\rho(\perp)$ could then be obtained from the measured resistivity of the two samples. Previous measurements on the anisotropy of the electrical resistivity for pure tin[2,3] have indicated that a maximum occurs in the vicinity of 20 °K, and it was of interest to observe the behavior of the thermal anisotropy $a_T = \kappa(\perp)/\kappa(\parallel)$ given by the ratio of thermal conductivities of the two samples. A comparison of the electrical and thermal anisotropies for two pure samples of tin above 4.2 °K has not been previously reported because of the lack of thermal-conductivity measurements on pure oriented crystals in this temperature