COMMENTS AND ADDENDA

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Numerical Evaluation of the Frequency-Dependent Complex Conductivity of Superconducting Mercury

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The ratio of the frequency-dependent complex conductivity of superconducting mercury to that of normal mercury is calculated numerically from recent tunneling data of Hubin and Ginsberg.

Hubin and Ginsberg[1] have analyzed their data on aluminum/aluminum-oxide/mercury tunneling junctions using a computer program written by McMillan and Rowell.[2] For mercury, they have obtained the frequency dependence of $\alpha^2F(\omega)$, the square of the electron-phonon coupling parameter times the phonon density of states, and the complex gap parameter $\Delta(\omega)$. Using the latter and the theory of the electromagnetic response of strongly coupled superconductors,[3,4] the frequency-dependent ratio of the complex conductivity of superconducting mercury to that of normal mercury $\sigma/\sigma_n = \sigma_1/\sigma_n - i\sigma_2/\sigma_n$, where σ_1/σ_n and σ_2/σ_n are real, has been numerically evaluated. The results are valid for $T = 0$ in the Pippard and local limits. Since both σ and σ_n have the same dependence on wave vector in these two limits, the ratio σ/σ_n is dependent only on frequency.

Because of the strong coupling between electrons and phonons in mercury, the conductivity contains structure reflecting corresponding structure in the phonon density of states. However, the variations comprise only a small fraction of the conductivity, having a magnitude of less than 4% of σ_n. The major features are well described by the weak-coupling theory of Mattis and Bardeen[5] (MB) if one adds to that result a real and an imaginary term, each having a simple functional form. Thus rather than present the conductivity directly, quantities are shown in Fig. 1 which emphasize the difference between the conductivity of mercury and that calculated from the MB theory for a superconductor having the same energy gap $2\omega_g$.

In Fig. 1(a), the difference, $\Delta\sigma_1/\sigma_n \equiv \sigma_1^{Hg}/\sigma_n - \sigma_1^{MB}/\sigma_n$, between the real part of the calculated conductivity ratio of mercury σ_1^{Hg}/σ_n and that of Mattis and Bardeen σ_1^{MB}/σ_n is plotted. The δ-function term in $\Delta\sigma_1/\sigma_n$ at zero frequency is not shown. It results from a difference in the amplitudes of the δ-function terms in σ_1^{Hg}/σ_n and σ_1^{MB}/σ_n. The origin[6,7] and relative magnitudes[8,9] of these terms have been thoroughly discussed elsewhere. Structure in $\Delta\sigma_1/\sigma_n$ appearing at frequency ω is related to structure in the phonon spectrum at frequency $\omega - 2\omega_g$. Thus in Fig. 1(c), α^2F is plotted with its frequency coordinate shifted by $2\omega_g$ so that structure in this curve appears directly under structure in $\Delta\sigma_1/\sigma_n$. A discussion of the relation of the structure to the gap parameter[9] and in turn to the phonon spectrum[10] has been given elsewhere; thus we shall not consider it further.

Displaying the structure in σ_2/σ_n is somewhat more complicated. The imaginary part of the conductivity ratio contains a term $2A/\pi\omega$ which arises from the Kramers-Kronig transform of the δ-function term $A\delta(\omega)$ in σ_1/σ_n.[11] The difference, $\Delta\sigma_2/\sigma_n \equiv \sigma_2^{Hg}/\sigma_n - \sigma_2^{MB}/\sigma_n$, then contains a term $2\Delta A/\pi\omega$, where $\Delta A \equiv A^{Hg} - A^{MB}$, which at low fre-

quencies obscures the structure due to phonons. This term has therefore been subtracted. The factor ΔA has previously been calculated numerically for other strongly coupled materials[9] using the sum rule. A more direct approach used here is to evaluate $\omega\sigma_2/\sigma_n$ in the limit where ω goes to zero. Since $\omega\sigma_2/\sigma_n$ can be written as $2A/\pi$ plus a power series expansion in even, positive powers of ω for $\omega < 2\omega_g$,[12] the limit is $2A/\pi$. In the case of mercury, $A = 1.949 \times 2\omega_g$ with $2\omega_g = 1.66$ meV. For weakly coupled superconductors, $A = \pi^2/4 \times 2\omega_g$.[13] Therefore, in Fig. 1(b) $\Delta\sigma_2/\sigma_n - 2\Delta A/\pi\omega$ is plotted, where $\Delta A = -0.518 \times 2\omega_g$. The quantity thus plotted is the Kramers-Kronig transform of $\Delta\sigma_1/\sigma_n$ if the δ function is not included in the difference of the real parts of the conductivity ratios. Again, structure arising from phonons is observed. This structure is similar to the derivative of $\Delta\sigma_1/\sigma_n$[14] and has magnitude about equal to that in $\Delta\sigma_1/\sigma_n$.

It is clear that for mercury, and in fact for other strongly coupled superconductors,[9,15] it is possible for some applications to obtain a useful approximation to the conductivity ratio by adding $2\Delta A/\pi\omega$ to the imaginary part of the MB result. If $\Delta A\delta(\omega)$ is also added to the real part, the real and imaginary parts will be Kramers-Kronig transforms of each other, but σ will not obey the sum rule.[6,7] Thus one must excise caution in using the approximation, particularly in calculations where integration of σ is involved.

Present far-infrared spectrometers will permit the conductivity of mercury to be distinguished from that of a weakly coupled superconductor only by the relatively large difference in the coefficients A in the ω^{-1} terms in σ_2/σ_n. Far-infrared experiments, on very thin films in which the Pippard limit strictly applies, have already revealed a value of A different from the MB value in lead,[8] amorphous bismuth,[15] amorphous gallium,[15] and β-phase gallium.[15] Observation of the details of the structure in σ/σ_n will probably be possible only by the development of improved far-infrared techniques.

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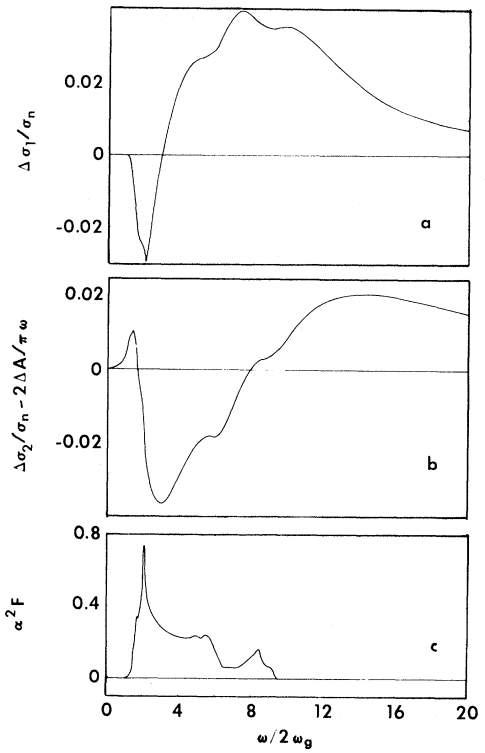

FIG. 1. Conductivity ratio and phonon spectrum of mercury as a function of reduced frequency $\omega/2\omega_g$. (a) Difference between the real part of the conductivity ratio of mercury and that of a weakly coupled superconductor having the same energy gap. (b) Difference between the imaginary part of the conductivity ratio of mercury and that of a weakly coupled superconductor having the same energy gap. The term proportional to ω^{-1} has been subtracted to better display the phonon-induced structure. (c) Square of the electron-phonon coupling parameter times the phonon density of states for mercury. The frequency scale has been shifted by $2\omega_g$ to facilitate comparison of phonon structure with structure in the conductivity ratio.

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[11]This defines A as the strength of the δ function in σ/σ_n as in Ref. 9. Other authors define it as the strength of the δ function in σ.
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Note on "Approximate Free Energies for Heisenberg Ferromagnets"

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Radcliffe[1] has published a method of treating the Heisenberg ferromagnet by using Peierls's[2] theorem on the minimization of an approximate free energy, applying the theorem to a lattice composed of clusters. The largest cluster treated is the Bethe cluster[3] of $n+1$ atoms, where n is the coordination number of the lattice.

Following Radcliffe, a cluster Hamiltonian allows us to write the approximate density matrix as a product over clusters [his Eq. (V1)] and to obtain a free energy depending on the fields acting on the central atom and its neighbors in the cluster [Eq. (V5)].

Radcliffe then assumes that the partial derivatives of the free energy with respect to these two fields are both zero, i.e., that the fields are independent. This leads to a prediction of a transition temperature (see Table I), but the consistency condition that the average magnetizations of central and neighbor atoms is the same is not satisfied. This condition, in Radcliffe's notation, is

$$\frac{\partial F_\alpha}{\partial h_\alpha} = \frac{1}{n}\frac{\partial F_\alpha}{\partial h'_\alpha} ,$$

which yields h_α as a function of h'_α. We modify Radcliffe's procedure by imposing this consistency condition. If we now further require the minimization of the free energy in terms of these two dependent variables, we also obtain a condition that predicts a transition temperature.

To investigate the difference between the transition temperatures predicted by these two methods, we have used the classical spin model[4] to simplify the arithmetic. In our "modified Radcliffe" method, the equation for the Curie temperature T_C is

$$(n+1) - (n-1)L - \tfrac{1}{3}bn(n-1)(1+L) = 0 ,$$

where $L = \coth b - 1/b$

and $b = 2JS^2/kT$,

while Radcliffe's method gives (for classical spin)

$$1 - \tfrac{1}{3}b(n-1)[1+(n-1)L^2] = 0 .$$

As shown in the table, the "improvement" of the numerical results that come from requiring consistency is in the right direction, but negligibly small. The reason for this smallness might be found in the following observation: The breakup of the lattice into Bethe clusters gives $N/(n+1)$ clusters and so $Nn/(n+1)$ exchange interactions in these clusters which are included correctly in the approximate product density matrix. The interactions between clusters involve $Nn(n-1)/2(n+1)$ exchange couplings which are considered uncorrelated, i.e., they are replaced by terms of the form $-2J\langle S_z\rangle^2$. It turns out that these are dominant in determining the Curie temperature because of their great number, and this is true whether the theory is self-consistent or not. Hence, the problem is with the method itself, in particular, with the requirement that the density matrix be written as a product.

TABLE I. Predicted transition temperature for a consistency condition.

kT_C/JS^2	$n=2$	4	6	8
Radcliffe	0.889	2.418	3.824	5.195
Consistent Radcliffe	0.874	2.407	3.817	5.195
Bethe-Peierls-Weiss method	•••	1.86	3.25	4.61