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Eliashberg function in an amorphous simple metal alloy $Sn_{1-x}Cu_x$ determined by electron tunneling

P. W. Watson III
NMT-5, MS E506, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

D. G. Naugle
Physics Department, Texas A&M University, College Station, Texas 77843-4242
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Superconducting tunneling measurements for amorphous $Sn_{1-x}Cu_x$ ($0.08 \leq x \leq 0.41$) have been inverted to determine the Eliashberg function $\alpha^2F(\omega)$ as the electron-phonon coupling parameter λ and the superconducting transition temperature T_c decrease from 1.8 to 1.0 and 6.87 K to 3.91 K, respectively, with the Cu content. The best fit to the experimental tunneling data was given with a linear dependence for $\alpha^2F(\omega)$ at low frequencies for all compositions. This linear dependence is consistent with the model for phonon damping proposed by Belitz and Wybourne (following paper) which properly incorporates electronic damping but also includes a strong nonelectronic contribution.

A question of fundamental importance to a number of topics is a correct model for the electron-phonon interaction in the presence of strong disorder. A naive approach would suggest that diffusive behavior of the electrons in highly disordered metals would reduce the screening and increase the electron-phonon interaction. This, however, is in disagreement with Pippard's result[1] that the longitudinal ultrasonic attenuation coefficient decreases monotonically with the product of the phonon wave vector and electron mean free path l. Microscopic calculations of the sound attenuation have reproduced the standard Pippard result, but the question regarding the low-frequency behavior of the Eliashberg function $\alpha^2F(\omega)$ has been more confused. Bergmann[2] predicted that $\alpha^2F(\omega)$ would be proportional to ω at low frequency on the basis of phase space arguments. Subsequently, Poon and Geballe[3] and Meisel and Cote[4] obtained an expression for α^2F for low frequencies which was identical to that of Bergmann. For very short mean free paths (high-resistivity alloys) Meisel and Cote[4] invoked the Pippard condition in an *ad hoc* fashion and predicted an ω^2 dependence. The microscopic models[5] of the electron-phonon interaction which correctly gave the ultrasonic attenuation, on the other hand, predicted an ω^3 dependence at low frequency.

The experimental situation for most amorphous simple metal alloys indicated that $\alpha^2F(\omega)$ was linear in ω at low frequency.[6] Measurements for amorphous Mo and amorphous Nb thin films,[7] however, showed a quadratic dependence for $\alpha^2F(\omega)$ at low frequency. These were transition-metal alloys, however, with significantly larger resistivities. Belitz[8] showed that *all* microscopic models yielding the Pippard result for the ultrasonic attenuation also give the same answer for $\alpha^2F(\omega)$, the Keck-Schmid[5] result with a small correction, and also suggested an explanation as to why the experiments predominantly showed a linear dependence based on asymptotic expansions in different frequency regions.

As discussed in a preliminary report of these tunneling measurements,[9] the amorphous $Sn_{1-x}Cu_x$ system is the best characterized of simple metal amorphous superconductors and is well suited for tunneling measurements to test the details of superconductivity in amorphous simple metals. The amorphous phase can be prepared over a wide composition range ($0.08 \leq x \leq 0.75$) by cocondensation of thin alloy films onto a liquid-helium temperature substrate. The superconducting transition temperature decreases linearly from approximately 6.9 to 0.9 K over this composition range. Heat-capacity measurements[10,11] which indicate the variation of the electronic density of states and the Debye temperature Θ_D over the entire composition range are also available. Unfortunately, strong-coupling deviations from the BCS reduced density of states become too small and the transition temperature too low to obtain good tunneling data for films with Cu concentration greater than about $x=0.4$ with our experimental system. Nevertheless, good tunneling measurements have been obtained over the composition range $0.08 \leq x < 0.41$ where the transition temperature changes from 6.87 to 3.91 K, the electron-phonon coupling parameter λ from 1.8 to 1.0, and the resistivity ρ from about 45 $\mu\Omega$ cm to 65 $\mu\Omega$ cm with this increase in Cu concentration. With this broad range of experimental data a reasonable set of parameters can be determined for use in the theoretical calculations of the low-frequency behavior of $\alpha^2F(\omega)$ for direct comparison with that determined from the tunneling measurements. In particular, we have tested the suggestion by Belitz[8] that Schmid's model with undamped[5] or weakly damped[8] phonons which yields a linear dependence of $\alpha^2F(\omega)$ on ω over some intermediate frequency range[5] might be consistent with tunneling measurements after all. For the parameters appropriate to $Sn_{0.87}Cu_{0.13}$ the model does not exhibit the linear behavior found in the asymptotic expansion, and the fit to the experimental data is quite poor. The model has been modified by Belitz and Wybourne[12] by incorporating strong phonon damping of nonelectronic origin into Schmid's theory of the electron-phonon coupling. The strong phonon damping is consistent with universally observed behavior in the sound propagation and thermal con-

ductivity of a wide range of glasses.[13] With this additional damping taken into account, good agreement is obtained with the tunneling density of states data.

Tunneling barriers approximately 23 Å thick were formed by oxidizing a 1300 Å thick, 0.25 mm wide Al film, which formed the counterelectrode of the tunnel junction, in one atmosphere of dry oxygen at 85 °C for about 14 h in the same UHV deposition chamber in which the film was deposited. The system was then pumped overnight to a pressure of about 10^{-9} Torr after which the substrate was heated to 60 °C for 2 h. The substrate with the counterelectrode and surrounding thermal shields was then immediately cooled to liquid helium temperature, and four 400 Å thick Sn-Cu films were sequentially e-beam evaporated from an alloy ingot source to complete the tunnel junctions. The base pressure before evaporation was less than 2×10^{-10} Torr and rose to the mid 10^{-9} Torr range during evaporation. Variation in composition over the thickness of a single Sn-Cu film was much less than 1% based on the variation of composition between the four films determined by wavelength dispersive microprobe analysis and T_c variations. This procedure reliably produced high-quality tunnel junctions with leakage currents less than 4% of the normal-state value and no zero bias anomalies. Careful measurements of the differential conductance of the junction as a function of voltage with the Sn-Cu films in both the normal state and the superconducting state and with the Al electrode normal were made at the lowest temperature attained with the cryostat ($T\sim1.5$ K). The low-temperature energy gap Δ_0 and T_c were carefully measured. These data together with the differential conduction were used in the inversion procedure to determine the Eliashberg function $\alpha^2F(\omega)$. It was not necessary to incorporate a thin normal proximity layer at the interface between the amorphous Sn-Cu film and the barrier, as was the case with the amorphous Mo and Nb films.[7] Complete details of the experimental techniques and the analysis procedure are discussed elsewhere.[14]

Due to the finite temperature of the measurements which produces a broadening of conductance in the superconducting state at voltages near the superconducting gap (the inversion program uses the zero-temperature equations) and experimental broadening produced by the use of a lock-in amplifier together with the slow sweep of the current during the conductance measurement, there is a minimum voltage (frequency) $\omega_{\min}$ above the gap edge below which the tunneling data cannot be used. A form for $\alpha^2F(\omega)$ must be assumed below $\omega_{\min}$ (typically 1.2 to 1.9 meV for these films) to invert the data. For a given assumption for the frequency dependence of $\alpha^2F(\omega)$ below $\omega_{\min}$, $\alpha^2F(\omega)$ is varied to produce the smallest difference between the experimental tunneling density of states and that calculated with $\alpha^2F(\omega)$. For most superconductors a quadratic dependence of $\alpha^2F(\omega)$ below $\omega_{\min}$ provides good agreement between the experimental and calculated tunneling density of states. For these films, as was the case for many other amorphous alloys,[6] the assumption of a linear frequency dependence below $\omega_{\min}$ produced the best fit for all compositions. Average values of the phonon frequency $\langle\omega\rangle$ determined from the calculated Eliashberg functions increased linearly with Cu concentration as expected although the relation $\Theta_D=1.21\langle\omega\rangle$ did not hold. The electron-phonon coupling constant decreased linearly with Cu concentration, and values of the Coulomb pseudopotential, $\mu^*(\omega_{\rm ph})=0.10\pm0.02$, were approximately a constant independent of x.

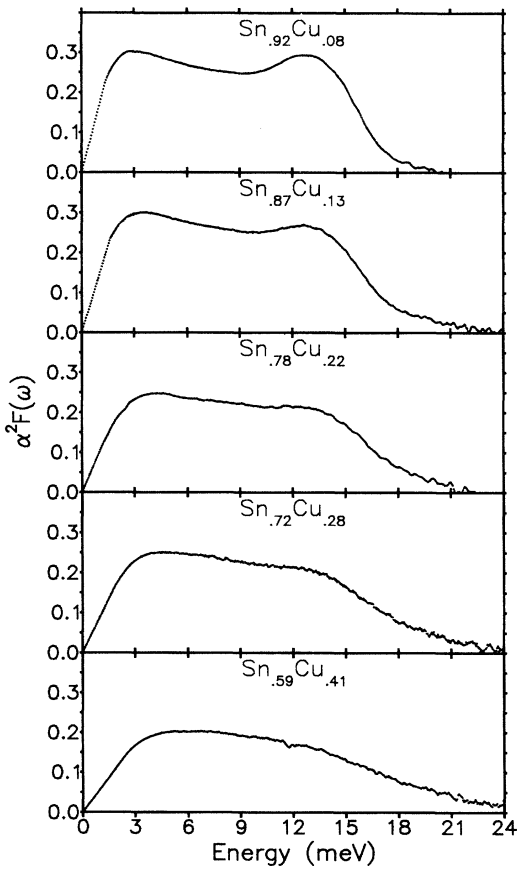

FIG. 1. The Eliashberg function $\alpha^2F(\omega)$ determined from tunneling density of states data for different $Sn_{1-x}Cu_x$ amorphous alloy films fitted with the assumption of a linear frequency dependence in the low-frequency regime.

Values of $\alpha^2F(\omega)$ for several compositions with $\alpha^2F(\omega)=A\omega$ below $\omega_{\min}$ are shown in Fig. 1. A fit with an $\omega^{1/2}$ produced a large positive deviation in a difference plot like that in Fig. 2, whereas, an ω^2 or ω^3 produced large negative differences. For $Sn_{0.59}Cu_{0.41}$, which exhibits a larger resistivity and might thus be expected to show an ω^2 dependence at low frequency based on the arguments of Meisel and Cote,[4] the inversion would not converge with the assumption of a quadratic dependence. Based on the predictions of Poon and Geballe[3] the slope of $\alpha^2F(\omega)$ in the linear region should be given for a free electronlike metal by

$$A=\frac{11.56\hbar^3k_F^5\rho A_0}{6\pi^5mM\Theta_D^3}, \tag{1}$$

where m is the electron mass, M the ionic mass, k_F the Fermi momentum, A_0 Avogadro's number, $\hbar$ Planck's constant over 2π, ρ the resistivity, and Θ_D the Debye temperature. Values of A calculated from Eq. (1) with the measured values of resistivity[14] and Debye temperature[10,11] are smaller than the slopes of $\alpha^2F(\omega)$ in Fig. 1 by factors of 3 to 5.

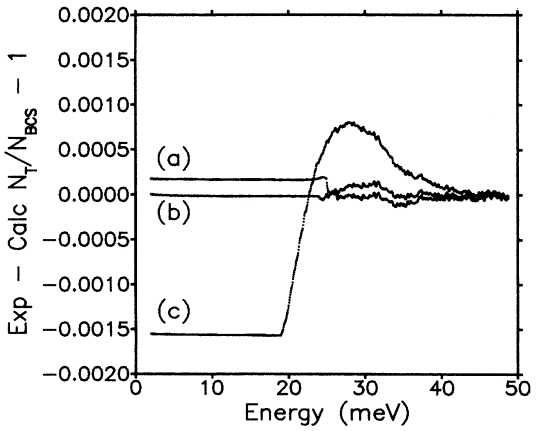

FIG. 2. Differences between the experimental and calculated tunneling density of states as a function of energy for amorphous $Sn_{0.87}Cu_{0.13}$ with different assumptions for the low-frequency behavior ($\omega \leq 1.5$ meV) of $\alpha^2F(\omega)$. Curve (a) uses values calculated from the model proposed by Belitz and Wybourne (Ref. 12) which properly incorporates electronic damping of the phonons but also includes a large nonelectronic phonon damping contribution; (b) assumes a strictly linear fit for $\alpha^2F(\omega)$; (c) uses the Keck-Schmid model (Ref. 5) which includes only electronic damping.

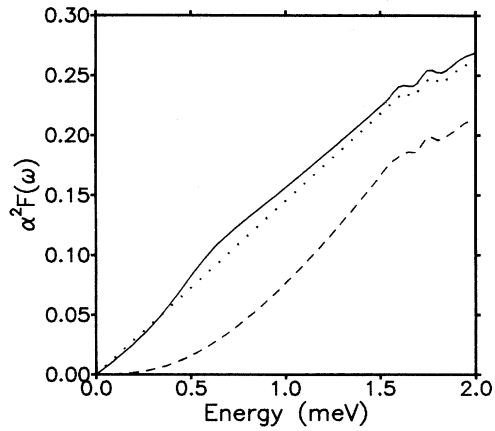

FIG. 3. The low-frequency values of $\alpha^2F(\omega)$ corresponding to models for curves (a) (solid line), curve (b) (dotted line), and curve (c) (dashed line) described in Fig. 2.

To test whether the result by Keck and Schmid, because of its approximately linear frequency dependence of $\alpha^2F(\omega)$ in an intermediate frequency range,[5] might account for the experimental observations as suggested in Ref. 8, Belitz[15] has evaluated the integrals in Ref. 5 numerically, with phonon damping due to electrons included as in Ref. 8. This numerical evaluation was necessary, since in the interesting frequency range, $\omega \leq 1.5$ meV$=\omega_{\min}$ and with parameters appropriate for $Sn_{0.87}Cu_{0.13}$, the previous analytic expressions are not applicable. The parameters used for the $Sn_{0.87}Cu_{0.13}$ alloy are $k_F=1.59\times10^8$ cm^{-1}, $v_F=1.84\times10^8$ cm/s, $\varepsilon_F=1.54\times10^{-11}$ ergs, $l=9.58\times10^{-8}$ cm, $c_l=1.60\times10^5$ cm/s, $c_t=8.10\times10^4$ cm/s, and the ratio of electronic to ionic mass density$=2.00\times10^{-5}$. Here v_F is the Fermi velocity, ε_F the Fermi energy, l the electronic mean free path, and $c_{l(t)}$ the longitudinal (transverse) velocity of sound. The velocity of sound was determined from Θ_D with the assumption that $c_l=2c_t$; k_F, v_F, and ε_F were determined from the free electron model but are not drastically different from what would be determined from the heat-capacity measurement of γ with the tunneling value of λ, and the value of l is determined from the resistivity and free electron value of k_F. The $Sn_{0.87}Cu_{0.13}$ film was chosen because a more accurate determination of $\alpha^2F(\omega)$ could be made for the more Cu-rich alloys which were more strong-coupling superconductors and because three different depositions at approximately the same composition showed consistent results. Although strong-coupling effects were somewhat greater for $Sn_{0.08}Cu_{0.92}$, the resistivity for the film could not be accurately determined, since the film agglomerated on warming to room temperature. In the fit of the tunneling data with the calculated values of $\alpha^2F(\omega)$ (which included no adjustable parameters), an adjustable scaling parameter, which is equivalent to the adjustable slope of $\alpha^2F(\omega)$ when a linear dependence at low frequency is assumed, was used. The best fit required scaling the calculated values of $\alpha^2F(\omega)$ below $\omega_{\min}$ by 0.93, but, as shown by curve (c) in Fig. 2, the difference between the measured and calculated reduced tunneling density of states was very large considering that the typical deviation from the BCS density of states values due to strong-coupling effects for this alloy is typically of the order of 1.5% over this same range. The poor agreement between the calculated value and experimental value of the tunneling density of states is readily understood when the calculated $\alpha^2F(\omega)$ (scaled by 0.93) is plotted in Fig. 3. It is much closer to an ω^2 dependence than to the linear dependence which produces curve (b) in Fig. 2.

The experiments clearly indicated that an approximately linear dependence of $\alpha^2F(\omega)$ was needed in the range between 0.5 and 1.5 meV. This indicates highly damped phonons, but the necessity to produce the Pippard result[1] indicated that a linear ω dependence could not result from the electronic mechanism[8] alone. Belitz and Wybourne[12] modified the calculation of $\alpha^2F(\omega)$ by adding an *ad hoc* nonelectronic phonon damping term which produces a phonon mean free path of 2 cm at 1 GHz and is modeled after that described by Anderson[13] to explain the universal features of the thermal conductivity of glasses at low temperatures. The model is discussed further in the following paper. The resulting low-frequency $\alpha^2F(\omega)$ (scaled by 0.86) is shown in Fig. 3. In the crucial range from 0.5 to 1.5 meV this model gives a very nearly linear dependence of $\alpha^2F(\omega)$ on frequency. The difference between the experimental and calculated tunneling density of states for this model is shown in curve (a) in Fig. 2. Although not as good as the fit with an assumed linear behavior, the agreement is good and would be considered to lie within the accuracy of the tunneling measurements.

In conclusion, tunneling measurements indicate that the low-frequency dependence of $\alpha^2F(\omega)$ in amorphous simple metal alloys must be approximately linear in frequency in the critical region from roughly 0.5 to 5 meV. This has been experimentally confirmed for an alloy system (Sn-Cu) in which T_c and λ decrease from 6.87 to 3.91 K and 1.8 to 1.0,

respectively, with increasing Cu content. The linear dependence cannot be provided by the Keck-Schmid theory[5,8] with phonon damping by electrons alone. Both theoretical[8] and present experimental arguments indicate that the Bergmann[2] and related[3,4] model calculations do not give the correct result even though they find a linear dependence on ω. Addition of a nonelectronic contribution to the phonon damping in the Keck-Schmid theory provides reasonable agreement with experiment. As discussed in the following paper, the source of this nonelectronic damping, which is observed in thermal conductivity and sound propagation in glasses remains unknown, but, at least what was once two unrelated and unresolved problems appear to have been reduced to one, the understanding of the nonelectronic phonon damping.

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