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Tomasch-oscillation amplitude decay in single-crystal lead films*

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The dependence of the amplitude of Tomasch oscillations on energy is measured for several different orientations of single-crystal lead films. Anisotropy in this amplitude is observed. Comparison is made between experiment and the predictions of several theories. In general the observed decay in amplitude with energy is more rapid than theoretical predictions. The best fit occurs with the McMillan-Anderson model and the Wolfram model with proximity effects.

I. INTRODUCTION

It is the purpose of this paper to compare experimental measurements of the dependence of the amplitude of Tomasch oscillations on energy in single-crystal lead films backed with silver with various theoretical predictions of that amplitude. For such a system, current Tomasch-oscillation theories[1–4] present the change in the quasiparticle density of states as a function of energy (ω) in a form such that, to sufficient precision for our consideration

$$\delta\rho(\omega)=A(\omega)\cos\left(\frac{2d}{\hbar v_F}\operatorname{Re}(Z\Omega)\right)\times\exp\left[-\left(\frac{2d}{l_e}+\frac{2d}{\hbar v_F}\operatorname{Im}(Z\Omega)\right)\right], \quad (1)$$

where

$$\Omega=[\omega^2-\Delta^2(\omega)]^{1/2}$$

and where l_e is the electron mean free path, d is the thickness of the film showing the oscillation, v_F is the Fermi velocity corresponding to the direction of tunneling in the film, Z is the complex renormalization constant, and Δ is the complex gap. The amplitude term $A(\omega)$ will be of primary interest here as it is independent of the parameters v_F and d, which will change from sample to sample.

II. EXPERIMENTAL DATA

Experimental measurements of the dependence of the amplitude decay on voltage were taken from voltage-current derivative curves (dV/dI) of Pb-*I*-Pb tunnel junctions (grown in this laboratory) which displayed a single Tomasch-oscillation frequency. The raw data are not unlike those reported earlier by Lykken *et al.*,[5] and the details of the experiment are as reported there. The films studied were oriented as follows: five with the ⟨100⟩ direction normal to the film surface; two with the normal 2° off ⟨100⟩ toward ⟨110⟩; one with the normal 5° off ⟨100⟩ toward ⟨110⟩; one with the normal along the ⟨110⟩; and one with the normal along the ⟨111⟩ direction. A smooth background curve was hand drawn through each of the curves. The value of the amplitude at each maximum or minimum was then measured relative to this background, and these absolute values were plotted to show the characteristic envelope of the Tomasch oscillations. A smooth curve was drawn through these points in each case (Fig. 1). These curves were compared at specified points with the predictions of the various theories. It is assumed that the Tomasch contribution to the current adds linearly, as all theories find that this term enters as an added term.

As l_e does not vary significantly over such changes in ω as are under consideration here, normalizing the data suffices to eliminate the term e^{-2d/l_e}, which is due to impurity scattering. The other exponential term in Eq. (1), which is due to phonon emission, cannot be so treated since its argument is dependent on ω. In this paper an effort is made to eliminate the major effect of this term by dividing both experimental and theoretical curves by an approximation for this term. The term $\exp[-(2d/\hbar v_F)\operatorname{Im}(Z\Omega)_{\text{poly}}]$ was constructed using values of $Z(\omega)$ and $\Delta(\omega)$ obtained by Rowell *et al.*[6] as a consequence of unfolding phonon spectra taken in tunneling studies of polycrystal lead. The effect of dividing by this term is shown graphically in Fig. 1 (dashed curve). The term $2d/\hbar v_F$ was obtained for each single-crystal film from the periodicity of the raw data, noting that extrema occur at points $n_i(\omega_i)\pi=(2d/\hbar v_F)\operatorname{Re}[Z(\omega_i)\Omega(\omega_i)]$. The method used involved indexing the extrema by $n_i(\omega)$ so that $(2d/\hbar v_F)_i=\pi/[\operatorname{Re}(Z\Omega)_{i+1}-\operatorname{Re}(Z\Omega)_i]$. Again polycrystalline data for Z and Δ were used to calculate $\operatorname{Re}(Z\Omega)$. The approximation that arises in these procedures is the assumption that the values of Z and Δ are the same for single-crystal and polycrystal films. Values of $(2d/\hbar v_F)_i$ for each junction did not vary significantly with i, indicating that $\operatorname{Re}(Z\Omega)$ for single-crystal lead of any orientation investigated could differ from that for polycrystal lead by at most a constant.

Figure 2 represents a graphic summary of the

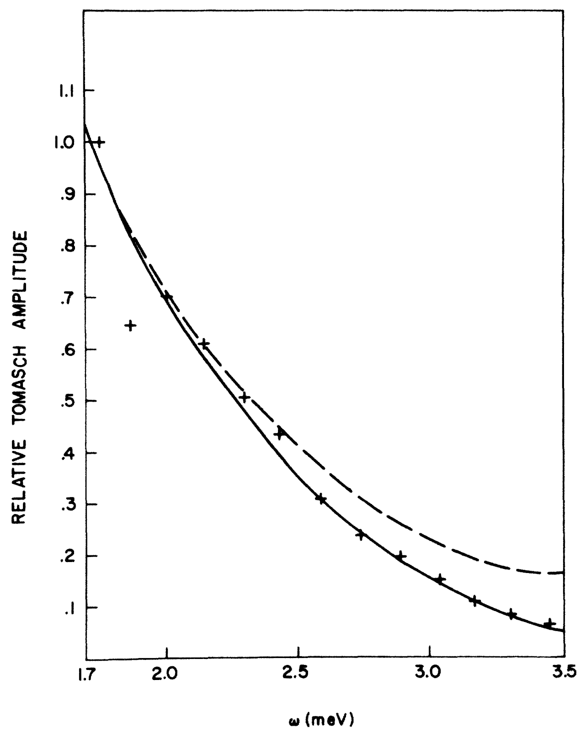

FIG. 1. Representative curve (solid line) showing smoothed curve and raw data points (crosses) used in this analysis. The dashed curve shows the effect of correcting for the exponential terms in Im $(Z\Omega)$ (see text).

experimental data adjusted using the above procedure. These data have been normalized to unity at $\omega = 2.3$ meV for convenience. At values nearer the conductance peak, considerable scatter in the data was observed. The data points represent arbitrarily chosen points on the Tomasch envelope derived by the procedure outlined earlier. The data taken for junctions oriented 2° off ⟨100⟩ were included with the data for junctions oriented with the film normal along ⟨100⟩. The data points and error bars on data designated ⟨100⟩ show the mean and standard deviation for this sample of seven films. All other points represent data for a single film of a specified orientation. While the data for films oriented 2° off ⟨100⟩ agreed quite well with those for the ⟨100⟩ films, the data for films oriented 5° off ⟨100⟩ did not agree nearly so well, as can be seen in Fig. 2. This may be an indication of the selectivity of the tunneling process or of the Tomasch mechanism in sampling only electrons traveling normal to the film surface. The good internal consistency of the ⟨100⟩ data is a modest measure of the reliability of the procedures used in obtaining this modified data. This attitude is reinforced by the fact that these junctions were fabricated by three different experimenters over a period of several years. In all future references in this paper to ⟨100⟩ data, it is understood that these include the data taken from films oriented 2° off ⟨100⟩.

From the data at voltages greater than 2.3 meV (see Fig. 2), one concludes that considerable anisotropy exists between the ⟨100⟩ orientation and the other orientations. One would like to use such information to establish the anisotropy in $Z(\omega)$ and/or $\Delta(\omega)$. Unfortunately, the dependence of the Tomasch oscillations on these parameters is too complex to establish the nature of the anisotropy in Z and Δ. It is of interest that the Tomasch behavior in the ⟨100⟩ direction appears to be uniquely different from the other principal orientations.

It is of further interest that a change of no more than 5° from the ⟨100⟩ direction in film orientation results in data that are not unlike those for the other principal directions.

III. COMPARISON WITH THEORY

Since the experimental data represent the amplitude of the dynamic resistance of the Tomasch contributions and since theoretical calculations of the dynamic conductance are much simpler to handle, it will be necessary to relate the two. The tunneling current at 0 °K for a superconductor-in-

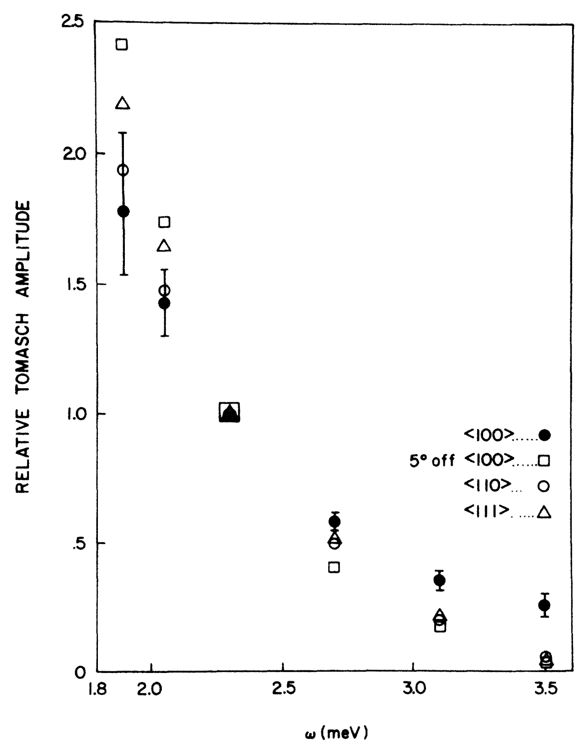

FIG. 2. Graphical summary of the adjusted experimental data used in this paper.

sulator-superconductor junction biased at a voltage V is given by

$$I_{ss} = C_N \int_{\Delta_1}^{V-\Delta_2} \rho_1(\omega)\rho_2(V-\omega)\,d\omega + C_N \int_{\Delta_1}^{V-\Delta_2} \rho_1(\omega)\delta\rho_2(V-\omega)\,d\omega \;, \qquad (2)$$

where C_N is the normal-state density of states, ρ_1 and ρ_2 are the density of states in superconducting films 1 and 2 in the absence of Tomasch contributions, $\delta\rho_2$ is the perturbation in the density of states in film 2 due to the Tomasch contributions, and Δ_1 and Δ_2 are the BCS superconducting energy gaps of films 1 and 2. The dynamic conductance can be written $(dI/dV)_{ss} = B + T$, where B/C_N and T/C_N are the derivatives with respect to voltage of the first and second integrals in Eq. (2). Since

$$\left(\frac{dV}{dI}\right)_{ss} = 1\bigg/\left(\frac{dI}{dV}\right)_{ss} = \frac{1}{B+T} = \frac{1}{B(1+T/B)} = \frac{1}{B}\left[1 - \frac{T}{B} + \left(\frac{T}{B}\right)^2 \cdots\right] = \left(\frac{1}{B} - \frac{T}{B^2} + \frac{T^2}{B^3}\cdots\right) \qquad (3)$$

and since $T \approx 0.05B$, one may approximate

$$\left(\frac{dV}{dI}\right)_{ss} \approx \frac{1}{B} - \frac{T}{B^2} = -\frac{1}{a}(B' + T') \;, \qquad (4)$$

where B' and T' are, respectively, the experimentally measured background and Tomasch contributions to the dynamic resistance. A constant has been introduced here to provide for renormalizing the experimental data relative to the theoretical calculations. Since in the absence of Tomasch oscillations it is assumed that $B' = -a/B$, it follows that $T' = aT/B^2$. Hence one can compare the experimentally measured envelopes (suitably normalized) to the envelope derived from the various theories provided that the theoretical envelope is divided by B^2.

A. Dependence on energy

The envelope data taken on films oriented with ⟨100⟩ normal to the film surface and those taken with 2° off ⟨100⟩ normal to the film surface will be combined and compared with four theoretical results in this section. The perturbation in the current due to Tomasch effects is of the general form

$$\delta I = C_N \int_{\Delta_1}^{V-\Delta_2} \rho_1(\omega)\delta\rho_2(V-\omega)\,d\omega \qquad (5)$$

at 0 °K. Since these data were taken typically at temperatures below 1.4 °K, it will be satisfactory to use this approximation. Both $\rho_1(\omega)$ and $\delta\rho_2(V-\omega)$ are complex in nature in all of these theories owing to the complex nature of Z and Δ. However, since the maximum error introduced by disregarding $\mathrm{Im}(Z)$ and $\mathrm{Im}(\Delta)$ is about 5%, these terms will be ignored, inasmuch as our experimental error is at least 5%. The first term under the integral becomes $\rho_1(\omega) = \omega/\{\omega^2 - [\mathrm{Re}\Delta(\omega)]^2\}^{1/2}$. Appropriate expressions for $\delta\rho_2(\omega)$ were used for each theoretical variation.

In all cases, integration was carried out numerically using limits $\Delta_1 + \delta$ and $V - \Delta_2 - \delta$, where $\delta = 0.001$ meV, in order to avoid singularities at Δ_1 and $V - \Delta_2$. The first derivative was then obtained numerically for each theory and plotted vs V by the computer. Amplitudes at the extrema in the Tomasch oscillations were then used to construct a theoretical Tomasch-oscillation envelope just as was done for the experimental data. Finally, this theoretical envelope was divided by the square of the theoretical background curve (the term B above) and normalized to unity at 2.3 meV for comparison with the experimental data.

It was expedient in the calculations to use analytic expressions for Z and Δ. An empirical fit was made to experimental curves[7] obtained for polycrystal lead in the energy interval from 1.4 to 3.5 meV. These are

$$\mathrm{Re}Z(\omega) = 2.406(1 + 0.0185\omega^2 + 0.00036\omega^4)^{1/2} \;, \qquad (6)$$

$$\mathrm{Im}Z(\omega) = -0.0003\omega^3 + 0.000167\omega^5 \;, \qquad (7)$$

$$\mathrm{Re}\Delta(\omega) = 1.36(1 + 0.03\omega^2 + 0.00125\omega^4)^{1/2} \;, \qquad (8)$$

$$\mathrm{Im}\Delta(\omega) = -0.000377\omega^3 + 0.000104\omega^5 \;. \qquad (9)$$

The different theoretical results are a consequence of two degrees of sophistication in each of two basic approaches. The first of these approaches is to consider a δ-function perturbation in the gap, χ, at the back edge of the film. McMillan and Anderson[1] (M-A) calculated the change in the Green's function to first order in the perturbation of the gap choosing χ to be a constant equal to $\delta\Delta$. Wolfram and Einhorn[4] (W-E) performed the calculation to all orders in the perturbation and introduced a χ which scaled the off-diagonal self-energy $\phi(\omega)$ so that $k_F\chi = \phi t$, where t is a constant. In the limit where $\Omega/\phi \gg (\phi/E_F)t$, the W-E exact result reduces to the M-A result. The difference between the two theories in this limit is the choice of χ. The second approach imposes a boundary condition on the Green's function for a superconductor in contact with a second metal at the interface between the two metals. The Wolfram[3] model (W) allows the second metal to be a superconductor while the McMillan[2] (M) model does not.

The M-A theory[1] predicts

$$\delta\rho(\omega) \propto \frac{\omega\Delta\delta\Delta}{\Omega^2}\mathrm{Si}\left(\frac{2d}{\hbar v_F} Z\Omega\right) \;, \qquad (10)$$

where $\delta\Delta$ is the perturbation in the gap at the far surface of the film, $\Omega = (\omega^2 - \Delta^2)^{1/2}$, and

$$\mathrm{Si}(x) = \int_x^\infty \frac{\sin y}{y}\,dy \;. \qquad (11)$$

Maki and Griffin[7] have presented an approximation

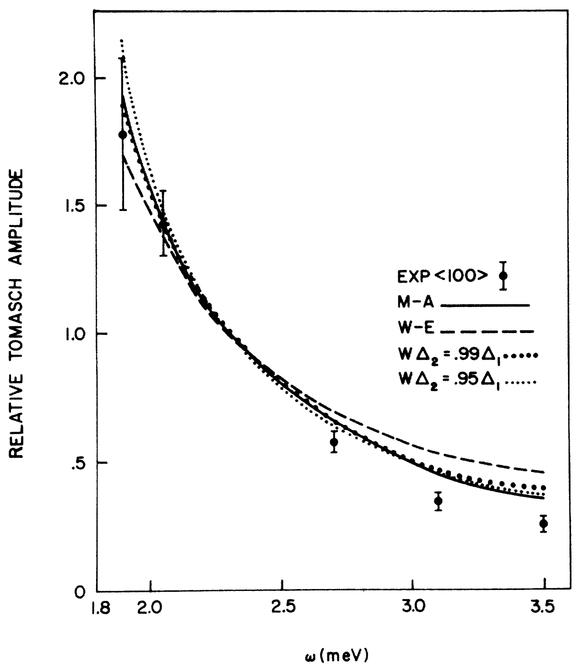

FIG. 3. Comparison of experimental Tomasch envelopes with three theoretical formulations. For W, Δ_2 is the gap in silver, Δ_1 that in lead.

for the Si function so that when imaginary terms are dropped,

$$\mathrm{Si}\left(\frac{2d}{\hbar v_F}Z\Omega\right)=\cos\left(\frac{2d}{\hbar v_F}Z\Omega\right)\Big/\left[\left(\frac{2d}{\hbar v_F}\right)^2 Z^2\Omega^2+\left(\frac{d}{l_e}\right)^2\right]^{1/2} . \tag{12}$$

The mean free path in these films was not obtained directly but was obtained by comparing resistivity ratios here to those of Anderson and Hines.[8] This comparison was used to scale the mean free path here to that obtained by Anderson and Hines. Typically d is 4 μm and l_e is 16 μm in our study. Since $(2d/\hbar v_F)\,\mathrm{Re}(Z\Omega)$ is greater than 15 in all cases, (9) reduces to

$$\delta\rho(\omega)\propto\frac{\omega\Delta\delta\Delta}{Z\Omega^3}\cos\left(\frac{2d}{\hbar v_F}Z\Omega\right) . \tag{13}$$

In this case, $\delta\Delta$ was chosen as constant and absorbed in the normalizing process. The resulting Tomasch envelope divided by B^2 is shown in Fig. 3.

W-E[4] scaled the perturbation at the far surface (χ) with the off-diagonal self-energy parameter so that $k_F\chi=\phi t$. They also added a term for higher-order contributions which made their expression

$$\delta\rho(\omega)\propto\frac{\omega\Delta^2}{\Omega^3}\cos\left(\frac{2d}{\hbar v_F}Z\Omega\right)+K\frac{\omega\Delta^4}{\Omega^4}\sin\left(\frac{2d}{\hbar v_F}Z\Omega\right) , \tag{14}$$

where K is a constant determined by the size of the Tomasch effect. If the Tomasch contribution is 10% of the unperturbed contribution $[\delta\rho(\omega)/\rho(\omega)=0.1]$, then $K\approx 0.21$. This value of K was tried in these calculations but made no significant change in $\delta\rho(\omega)$ in the energy region of interest in this study. Only the results for $K=0$ will be presented here. The resulting Tomasch envelope divided by B^2 is shown in Fig. 3.

Wolfram's[3] Green's-function boundary-condition approach yields a harmonic series for the Tomasch term. In this paper only the first term of the series is calculated. This approach provides for an overlay on the film which may also be superconducting. The general expression is

$$\delta\rho(\omega)\propto\frac{\Delta_1}{\Omega_1}\left(\frac{R_2\Delta_1-R_1\Delta_2}{R_1R_2-\Delta_1\Delta_2}\right)\cos\left(\frac{2d}{\hbar v_F}Z\Omega_1\right) , \tag{15}$$

where $\Omega_i=(\omega^2-\Delta_i^2)^{1/2}$ and $R_i=\omega+\Omega_i$. The subscript 1 refers to the superconducting film exhibiting Tomasch oscillations and the subscript 2 refers to the superconducting overlayer. In this experiment the overlayer was a silver film about 0.15-μm thick, and would be subject to proximity effects. In order to allow for this possibility, the gap in silver (Δ_2) was assigned the following fractions of that in lead (Δ_1): $\Delta_2/\Delta_1=0.0$, 0.5, 0.8, 0.95, 0.99, and 0.999. The resulting Tomasch envelopes divided by B^2 are shown in Fig. 4, and those for $\Delta_2/\Delta_1=0.95$ and 0.99 are repeated in Fig. 3.

The McMillan[2] Green's-function boundary-condition approach is only applicable to the case in which the overlayer is normal. Ignoring imaginary terms, one finds that

$$\delta\rho(\omega)\propto\frac{\omega-\Omega}{\Omega}\cos\left(\frac{2d}{\hbar v_F}Z\Omega\right) . \tag{16}$$

This yields normalized results that are indistinguishable from those for the Wolfram model with $\Delta_2/\Delta_1=0$ and is shown in Fig. 4.

As Fig. 3 shows, all of these methods predict a decay in the Tomasch envelope with increasing ω that is less rapid than that which is experimentally observed. Among them, however, the M-A model and the W model with silver gap of from 0.95 to 0.99 of the gap in lead seem to fit the best.

B. Amplitude

A bit more information can be obtained by comparing the magnitude of the Tomasch oscillations with the predictions of the various theories. As Eq. (1) indicates, the amplitude in all of these theories is of the form $T=G(\omega)\,e^{-2d/l_e}$, where $G(\omega)$ is an energy-dependent term which differs from theory to theory. In this experiment, e^{-2d/l_e} is about 0.6. One can get a normalized dI/dV experimental curve as a function of ω by dividing $(dV/DI)_{nn}$ by

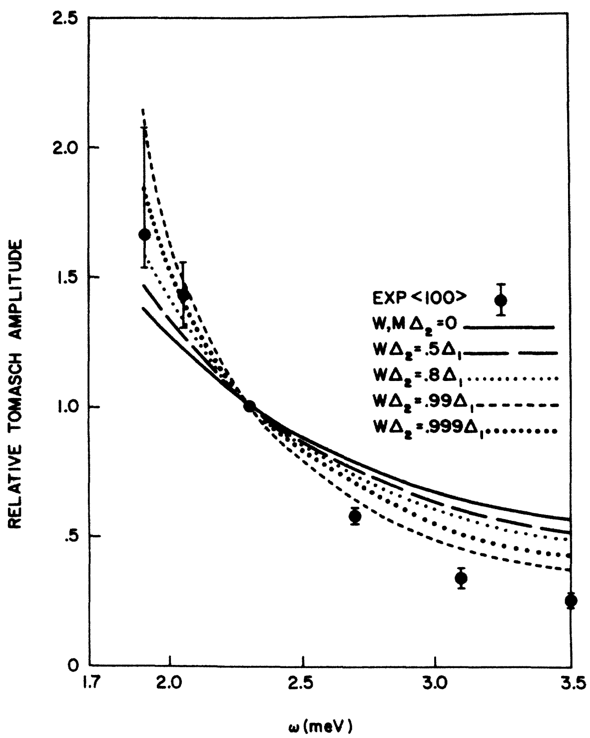

FIG. 4. Comparison of experimental Tomasch envelopes with theory for differing proximity contributions. For W, Δ_2 is the gap in silver, Δ_1 that in lead.

$(dV/dI)_{ss}$, where $(dV/dI)_{nn}$ and $(dV/dI)_{ss}$ are the dynamic resistances of the junction measured with both films, first normal and then superconducting. The experimental value of the Tomasch amplitude of the normalized dynamic conductance for $\omega = 2$ meV is $\delta(dI/dV)_{exp} \approx 0.10 \pm 0.05$. If one calculates $G(2.0)e^{-2d/l_e}$ for various values of Δ_2/Δ_1 (the ratio of the gap in silver to that in lead) in the W theory, one obtains the results of Table I. One sees that setting $\Delta_2/\Delta_1 = 0.99$ gives rise to very good agreement with theory on this point.

The M-A theory, which also fits the decay of the amplitude of the Tomasch oscillations with increasing energy fairly well, contains an undetermined parameter and cannot be compared directly to the experimental data reported here.

If one chooses to calculate the amplitude in the W-E theory[4] for the case $K = 0$ [see Eq. (13)], one finds that the theory contains a constant of proportionality t which is undetermined. This constant represents an inverse scaling factor on the gap perturbation χ. Using the experimental data contained in this paper and the Maki and Griffin expansion for the Si function, one can calculate χ, which is typically of the order of 2.4×10^4 meV Å. One might interpret this number as a measure of the product of the perturbation of the gap at the back surface of the film and the distance over which the perturbation occurs.

Rowell and McMillan[9] have shown that the envelope predicted by the M theory has the same energy dependence as one observed in an iron-backed lead film. This means that the W theory with the gap in iron set equal to zero would also fit those experimental data. The M theory does not predict the correct amplitude for the oscillations, and neither would the W theory with the gap set equal to zero.

C. Anisotropy

Since anisotropy in the gap has been measured,[5,10] the effect of changing the gap from that observed in polycrystal lead is of interest. The value of $\delta\rho(\omega)$ has been calculated for values $\Delta(\omega)$ differing from those for polycrystal lead using a simple scaling factor. That is, $\Delta(\omega)$ as computed was multiplied by a constant b, where b is the ratio of Δ_0 for a single-crystal film to that for a polycrystal film. In all cases, this involved decreasing $\Delta(\omega)$ from its polycrystal values and had the general effect in all theories of further decreasing the decay rates and making the fit less satisfactory.

The real part of the renormalization Z entered into the envelope calculations in the M-A theory as a factor $1/Z$. Any anisotropy in real Z which varies Z by a constant factor would not enter into the normalized curves.

Two other possible sources of anisotropy are the real and imaginary terms in $Z\Omega$ which appear in Eq. (1). However, the term $\mathrm{Re}(Z\Omega)$ appears in the argument of the cos term and determines the periodicity of the Tomasch oscillations. Since all of the maxima and minima can be indexed using one analytic form of $\mathrm{Re}(Z\Omega)$, this term probably differs by at most a constant from orientation to orientation. Without assuming v_F to be known (which is not done here) it is not possible to determine such constants.

Assuming that the observed anisotropy in the amplitude dependence of the Tomasch oscillations (see Fig. 2) is due to anisotropy in the term $\mathrm{Im}(Z\Omega)$, one can calculate $\mathrm{Im}(Z\Omega)$ for orientations other than ⟨100⟩ compared to that for the ⟨100⟩ orientation.

This has been done for the McMillan-Anderson formulation assuming that the dependence of $\mathrm{Im}(Z\Omega)$ on ω for films oriented with ⟨100⟩ normal

TABLE I. Tomasch amplitude for $\omega = 2$ meV for various gaps in silver.

Δ_2/Δ_1	$G(2.0)\, e^{-2d/l_e}$
0.95	0.45
0.99	0.096
0.999	0.0096

to the film surface is the same as that of polycrystal lead (as was done earlier). The value of $\mathrm{Im}(Z\Omega)$ for the "off ⟨100⟩" films is much larger (about four times greater) than that for the ⟨100⟩ films at all energies. This would seem to indicate that the quasiparticle lifetime is much shorter in the former case if this calculation has any meaning.

IV. SUMMARY

In summary one may state the following.

(i) Some anisotropy is observed in the amplitude dependence of Tomasch oscillations with energy. In particular, the dependence for ⟨100⟩-oriented films agrees with that for polycrystalline films but is different from that for the other orientations reported here. All the other orientations show the same amplitude dependence.

(ii) All theoretical calculations of the amplitude dependence considered here predict a decay with energy which is less than that observed.

(iii) The McMillan-Anderson formulation and the Wolfram formulation with a gap in the silver of 0.95 to 0.99 of that in lead fit the experimental amplitude dependence best.

(iv) Scaling the perturbation ($\delta\Delta$), as is done in the Wolfram-Einhorn approach, makes the fit with experiment less satisfactory. Some other scaling might be made to work.

(v) The amplitude of the Tomasch oscillations (as opposed to the energy dependence of that amplitude) predicted by the Wolfram formulation with a gap in the silver of 0.99 of that in lead agrees remarkably well with experiment.

(vi) From (iii) and (v) above one should note that in the case of the Wolfram theory, a gap in the silver of about 0.99 of that in lead gives rise to both the proper amplitude and the dependence of amplitude on energy of the Tomasch oscillations reported here.

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