

Origin of anomalous mass renormalizations inferred from proximity tunneling experiments

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Experiments with thin Ag layers ($d_N \sim 100\text{--}700 \text{ \AA}$) backed by thicker Pb ($d_S \sim 3500 \text{ \AA}$) have led Khim and Tomasch to infer large effective renormalizations for Ag that increase with decreasing d_N , even though the electron-phonon interaction presumably retains its bulk character. Arnold has recently shown that dirty proximity superconductors violate Anderson's theorem and that, in this regime, the usual treatment of elastic scattering is inappropriate for Andreev interference phenomena. He finds that both the pairing potential, $\Delta_N(E)$, and the effective layer renormalization, $Z_N(E)$, are influenced strongly by scattering. Our reanalysis, based on Arnold's theory and on Gallagher's compound-resonance theory, assumes bulk behavior for the Ag electron-phonon interaction. We obtain successful quantitative agreement with the Ag-Pb data when the Ag mean free path nearly equals the Ag film thickness. Our best estimates of the dressed Fermi velocities are $v_{FS}^*(\text{Pb}) = (0.72 \pm 0.06) \times 10^6 \text{ m/s}$ and $v_{FN}^*(\text{Ag}) = (1.07 \pm 0.06) \times 10^6 \text{ m/s}$.

I. INTRODUCTION

A. General considerations

When one side of a Giaever tunnel diode consists of a clean metal layer (N) backed by a superconductive layer (S), the ac diode resistance (dV/dI) oscillates¹⁻³ as a function of bias (V) because coherent Andreev reflections⁴ add virtual-state structure (scattering resonances) to the N -layer's density of states. These reflections, which occur at the N - S interface, result from spatially abrupt changes in the pairing potential and, for example, correspond to condensation (pairing) of an incident quasiparticle ($k > k_F$) and liberation of a previously paired, energetically degenerate excitation ($k < k_F$). Observed level spacings, obtained with N -metal layers of known thickness (d_N), yield reasonably direct determinations of dressed Fermi velocities (v_{FN}^*) that, when divided by theoretical, bare values (v_{FN}), yield quasiparticle mass renormalizations ($Z_N(0) = v_{FN}/v_{FN}^*$) attributable to the electron-phonon interaction.

Since level spacings vary approximately as $(d_N)^{-1}$ and since all level structure is quenched at higher energies (biases) by spontaneous phonon emission, only thicker N layers produce useful virtual-state spectra. Mass renormalization estimates for thinner layers remain possible, however, because of a related effect discovered by Rowell and McMillan.⁵ Current theories,^{6,7} which describe the virtual-state regime so successfully, also predict this latter effect: namely, that N -layer densities of states contain attenuated versions of the S -metal's phonon spectrum multiplied

by the same phase factor, $\exp(i\omega_N)$, responsible for virtual levels in thick N layers, the phase being given by

$$\omega_N = \Delta k_N d_N = (2d_N/\hbar v_{FN}) Z_N(E) [E^2 - \Delta_N^2(E)]^{1/2}, \quad (1.1)$$

where $\Delta_N(E)$ and $Z_N(E)$ denote, respectively, the complex, energy-dependent pairing potential and renormalization functions of the N layer.

Khim and Tomasch⁸ have used S -metal phonon structure to infer Ag mass renormalizations for a series of reasonably thin Ag films ($d_N \sim 100\text{--}700 \text{ \AA}$) backed by substantially thicker Pb films ($d_S \sim 3000\text{--}4000 \text{ \AA}$). Use of these two metals was prompted by (1) the immiscibility of Ag and Pb, (2) the strong-coupling superconductivity of Pb, and (3) the availability of pairing potential and renormalization functions for Pb from the compilation by Rowell, McMillan, and Dynes.⁹ Use of thin Ag films was prompted by the need to achieve usefully strong Pb phonon structure and by an interest in proximity effect phenomena, which tend to gain strength as d_N decreases. Although Khim and Tomasch could parametrize their Ag-Pb results in terms of effective renormalizations, these proved larger than anticipated and, furthermore, increased systematically with decreasing d_N . Since the electron-phonon interaction in Ag is not expected to show size effects for Ag films as thick as those employed, the latter trend is particularly puzzling. These aspects of the Ag-Pb results, as well as others involving large inferred gap values and additional structural detail, have remained unexplained.

Arnold¹⁰ has recently shown that the Anderson theorem, which holds for dirty, bulk superconductors, does not hold for the proximity-effect regime and, in particular, that *s*-wave elastic scattering in the *N*-metal layer cannot be treated within the usual phenomenological approximation. The latter assumes that elastic scattering leaves $\Delta_N(E)$ unaltered and modifies $Z_N(E)$ by simply adding a damping term, $i(d_N/l_N)$, to the phase ω_N , where l_N denotes the mean free path (mfp) for elastic scattering within the *N*-layer volume. This approach is entirely consistent with the standard interpretation of Anderson's theorem and hence remains applicable for sufficiently thick *N* layers. We propose that the anomalous effective renormalizations reported by Khim and Tomasch result from the failure of this approach and that, in general, their Ag-Pb observations are successfully described by Arnold's theory.

B. Specifics

Arnold's treatment of elastic scattering in thin *N* layers yields two important effects: (1) the pairing potential tends to become homogenized over the thickness of the *NS* sandwich, meaning that $\Delta_N(E)$ approaches $\Delta_S(E)$ for thin, dirty *N*-metal layers; and (2) the effective *N*-layer thickness, appropriate for interference effects involving Andreev reflections, exceeds d_N . The first of these leads to somewhat larger values of $\Delta_N(E)$ than predicted by standard proximity-effect calculations, such as those based on Eqs. (6.7) and (6.8) of Ref. 7. According to Eq. (1.1), the second effect, if neglected, leads to experimental overestimates of the mass renormalization, $Z_N(0)$. Both effects bear on understanding the Ag-Pb results.

While effects due to elastic scattering reconcile the major difficulties with the Ag-Pb results, differences in structural detail persist. These we attribute to compound resonances of the type described in Gallagher's recent theory¹¹ and employed by Wong, Shih, and Tomasch³ to explain their observations with Zn-Pb sandwiches. In addition to Andreev reflections from the *N*-*S* interface, Gallagher also considers ordinary reflections from the free surface of the *S*-layer and obtains complicated interference effects in the *N*-layer's density of states. Compound resonances occur in the Ag-Pb observations because of the large mfp values (l_S) realized in the clean Pb backings used by Khim and Tomasch. Fortunately, these effects are confined to energies below those characteristic of the Pb phonon spectrum, since spontaneous emission of Pb phonons quenches Pb layer resonances at higher energies. In this sense, compound resonances do not complicate the central experimental task, which is to infer phases from observed Pb phonon structure.

II. THEORY

Arnold's theory of elastic *s*-wave scattering¹⁰ assumes that the *N*-layer's pairing potential, $\Delta_N(E)$, is spatially uniform and can be approximated by an average. For finite scattering times, $\tau = l_N/v_{FN}$, the modified pairing potential and renormalization functions then obey the coupled equations

$$Z_N(E) = Z_N^{\text{ph}}(E) - \frac{\hbar}{2\tau} \int_0^1 d(\cos\theta) \times \left(\frac{X_1(E)}{\Omega_N} + \frac{\Delta_N}{E\Omega_N} \int_{-d_N}^0 \frac{dx}{d_N} X_2(E,x) \right) \quad (2.1)$$

$$\Delta_N(E) = \Delta_N^{\text{ph}}(E) - \frac{\hbar}{2\tau Z_N^{\text{ph}}(E)} \int_0^1 d(\cos\theta) \frac{\Omega_N}{E} \times \int_{-d_N}^0 \frac{dx}{d_N} X_2(E,x) \quad , \quad (2.2)$$

where $Z_N^{\text{ph}}(E)$ and $\Delta_N^{\text{ph}}(E)$ denote, respectively, the renormalization and induced pairing potential functions that pertain for the proximity regime in the absence of elastic scattering. For the thin *N*-metal limit, they may be obtained from Eqs. (7.20) and (7.21) of Ref. 7; otherwise, they may be obtained more generally from Eqs. (6.7) and (6.8). The remaining quantities in Eqs. (2.1) and (2.2) are defined by

$$X_1(E) = \frac{iF(E) \cos\omega_N + \sin\omega_N}{iF(E) \sin\omega_N - \cos\omega_N} \quad , \quad (2.3)$$

$$X_2(E,x) = \frac{i \cos[\Delta k_N(x + d_N)] G(E)}{iF(E) \sin\omega_N - \cos\omega_N} \quad , \quad (2.4)$$

where

$$F(E) = (E^2 - \Delta_S \Delta_N) (\Omega_N \Omega_S)^{-1} \quad , \quad (2.5)$$

$$G(E) = E(\Delta_S - \Delta_N) (\Omega_N \Omega_S)^{-1} \quad , \quad (2.6)$$

$$\Omega_{N,S} = (E^2 - \Delta_{N,S}^2)^{1/2} \quad , \quad (2.7)$$

$$\omega_{N,S} = \Delta k_{N,S} d_{N,S} = \left(\frac{2d_{N,S}}{\hbar v_{FN,FS}} \right) \left(\frac{Z_{N,S}(E)}{\cos\theta} \right) \Omega_{N,S} \quad , \quad (2.8)$$

and where θ denotes the angle between \vec{k} and the normal to the *NS* interface. Equations (2.1) and (2.2) are readily solved, by iteration, at energies exceeding the bulk *S*-metal energy gap (~ 1.4 meV) and hence at the energies of primary interest.

Gallagher's theory of compound resonances¹¹ also assumes $\Delta_N(E)$ and $\Delta_S(E)$ to be spatially constant. In terms of our notation, the resulting density of

states is given by

$$\begin{aligned}
 N(E) = & \operatorname{Re}[D(E)(E/\Omega_N) \sin\omega_N \cos\omega_S] \\
 & + \operatorname{Re}[D(E)(\Delta_N/\Omega_N)G(E) \\
 & \quad \times (\cos\omega_N - 1) \sin\omega_S] \\
 & + \operatorname{Re}[D(E)(E/\Omega_S) \cos\omega_S \sin\omega_S] , \quad (2.9)
 \end{aligned}$$

where

$$D^{-2}(E) = 1 - [\cos\omega_N \cos\omega_S - F(E) \sin\omega_N \sin\omega_S]^2 , \quad (2.10)$$

and where $\omega_{N,S}$ is evaluated at $\theta=0$. In our subsequent discussion of numerical results, values of $\Delta_N(E)$ and $Z_N(E)$ obtained from Eqs. (2.1) and (2.2) will be substituted into Eq. (2.9), and the result used to compute corresponding ac diode resistances for specular tunneling ($\theta=0$).²

To be consistent, a theory of N - S proximity sandwiches should treat elastic scattering in both the N and S layers. It can be shown, however, that Arnold's theory regains the Anderson regime for the S layer once d_S exceeds a few bulk coherence lengths,¹² $\xi_S \sim \hbar v_{FS}^*/\pi\Delta_S$, and therefore that the phenomenological approximation describes the thick Pb backings ($d_S/\xi_S \approx 6$) employed by Khim and Tomasch. The S -layer phase appearing in Eq. (2.9) therefore becomes

$$\omega_S = (2d_S/\hbar v_{FS})Z_S^{\text{ph}}(E)\Omega_S + i(d_S/l_S) , \quad (2.11)$$

where l_S denotes the mfp for elastic scattering. In accord with the Anderson theorem, $\Delta_S(E) \doteq \Delta_{SO}^{\text{ph}}(E)$ remains characteristic of the bulk.

III. COMPUTATIONS AND CURVE FITTING

We now describe our procedures for computing theoretical diode characteristics and for fitting these to experiment. Numerical computations based on Gallagher's density of states, Eq. (2.9), require an input-set that comprises six parameters— $d_{N,S}$, $l_{N,S}$, and $v_{FN,FS}$ —and four complex functions of energy— $\Delta_{N,S}(E)$ and $Z_{N,S}(E)$. Briefly, parameter values are obtained by the following means: Layer thicknesses, $d_{N,S}$, are available from experiment.⁸ Mean free paths, $l_{N,S}$, are inferred from a trial-and-error procedure in which l_S is adjusted to reproduce observed compound resonance effects, and l_N is adjusted in attempts to accommodate both observed amplitudes and elastic scattering effects. Finally, bare Fermi velocities are approximated by appropriate literature values, $v_{FN}(1.25 \times 10^6 \text{ m/s})$ being Lewis and Lee's theoretical value¹³ for Ag and $v_{FS}(1.77 \times 10^6 \text{ m/s})$ being the [111] velocity calculated¹⁴ from Anderson and Gold's pseudopotential fit of the Pb Fermi surface.¹⁵

Of the four functions needed, the two for Pb,

$\Delta_S(E)$ and $Z_S(E)$, are approximated by their bulk behavior as tabulated by Rowell, McMillan, and Dynes.⁹ This approximation reflects the minor importance of proximity phenomena, including elastic scattering effects, in Pb films as thick as those employed ($d_S \sim 3500 \text{ \AA}$). This leaves the task of acquiring suitable numerical representations for the N -layer functions, $\Delta_N(E)$ and $Z_N(E)$. We obtain these from what is essentially a first-principles approach based on Arnold's theory of elastic scattering. Eventually the success of this procedure (described below) will be gauged by the agreement with experiment that can be realized for reasonable values of the one adjustable parameter l_N .

The theory of N -metal elastic scattering¹⁰ itself requires knowledge of $\Delta_N^{\text{ph}}(E)$ and $Z_N^{\text{ph}}(E)$, quantities already modified by proximity phenomena. In the extreme thin limit as d_N becomes small, Eqs. (7.20) and (7.21) of Ref. 7 permit calculation of these quantities from the corresponding bulk properties, $\Delta_{NO}^{\text{ph}}(E)$ and $Z_{NO}^{\text{ph}}(E)$. For bulk normal metals, this requires that $Z_{NO}^{\text{ph}}(E)$ be known. In the absence of direct experimental information for Ag, we are forced to evaluate $Z_{NO}^{\text{ph}}(E)$ from the Eliashberg equations, a procedure which presumes that the electron-phonon spectral function, $\alpha^2F(\omega)$, is known as a function of phonon energy (ω). What is known, however, is only the phonon density of states, $F(\omega)$, calculated¹⁶ from the inelastic neutron scattering measurement of Kamitakahara and Brockhouse.¹⁷ The first step towards acquiring $\Delta_N(E)$ and $Z_N(E)$, therefore, is to estimate $\alpha^2F(\omega)$ for Ag.

We approximate $\alpha^2F(\omega)$ by assuming that the electron-phonon coupling coefficient, $\alpha(\omega)$, does not vary with phonon energy and hence that $F(\omega)$ ^{16,17} and $\alpha^2F(\omega)$ differ by only a constant factor of $(\alpha)^2$. Using the Eliashberg equations, we then compute the normal-state value of $Z_{NO}^{\text{ph}}(0)$ for trial values of α , and fix $\alpha(=0.238)$ by requiring that $Z_{NO}^{\text{ph}}(0)$ equal the reasonable value 1.15.¹⁸⁻²¹ Although our assumption may represent an oversimplification near the high and low ends of the phonon spectrum, results obtained with its aid are adequate for our present needs.

Substitution of $\alpha^2F(\omega)$ and $\mu^*=0.1$ into Eqs. (7.20) and (7.21) of Ref. 7 permits numerical solutions for the intrinsic layer properties, $\Delta_N^{\text{ph}}(E)$ and $Z_N^{\text{ph}}(E)$, appropriate for proximity superconductivity (extreme thin limit) in the absence of significant elastic scattering. Induced values of $\Delta_N^{\text{ph}}(E)$, which for Ag are quite small even in the extreme thin limit, decrease as d_N increases and proximity phenomena weaken. For Ag layers having $d_N > 500 \text{ \AA}$, we therefore set $\Delta_N^{\text{ph}}(E)$ to zero in subsequent calculations of $\Delta_N(E)$ and $Z_N(E)$ from Eqs. (2.1) and (2.2)—a reasonable procedure that improves agreement with experiment at lower biases (energies). For substantially thinner Ag layers, our use of nonzero computed

values of $\Delta_N^{\text{ph}}(E)$ has little actual effect, since now $\Delta_N(E)$ is enhanced and dominated by elastic scattering.

Similar to earlier results reported for Al,²² we find that ordinary proximity-induced superconductivity⁷ has only a minor effect on renormalizations, meaning that $Z_N^{\text{ph}}(E)$ differs only slightly from $Z_{NO}^{\text{ph}}(E)$ in the absence of substantial elastic scattering. Values of $Z_N(E)$ computed from Eqs. (2.1) and (2.2) can differ substantially from $Z_{NO}^{\text{ph}}(E)$, however, since the former are determined not only by the fundamental electron-phonon interaction but also by the extent of elastic scattering.

This situation lends itself to a semantic difficulty. Because $Z_N(E)$ differs from $Z_{NO}^{\text{ph}}(E)$ and depends on the layer parameters d_N and l_N , one can adopt either of two equivalent views: (1) if by *renormalization* one means $Z_N(E)$, then elastic scattering does in fact modify the *N-layer* renormalization; however, (2) if one means $Z_{NO}^{\text{ph}}(E)$ —the intrinsic, bulk renormalization determined by $\alpha^2F(\omega)$ —then $Z_N(E)$ represents only an effective renormalization, and elastic scattering does not change the *N-metal*'s intrinsic renormalization, which remains characteristic of the bulk. Henceforth, we will refer to $Z_{NO}^{\text{ph}}(E)$ as the intrinsic (bulk) renormalization and to $Z_N(E)$ as the effective (layer) renormalization.

IV. THEORY AND EXPERIMENT: A SUMMARY

We now examine the agreement attainable between observed and computed²³ ac diode resistance (dV/dI). All computations assume electron-phonon interactions that are independent of thickness ($d_{N,S}$) and characteristic of the bulk. Specifically, this means that all Ag layers are characterized by the same bulk $\alpha^2F(\omega)$ and hence have the same intrinsic renormalization, that of bulk Ag.

In Fig. 1, identical solid curves (displaced for con-

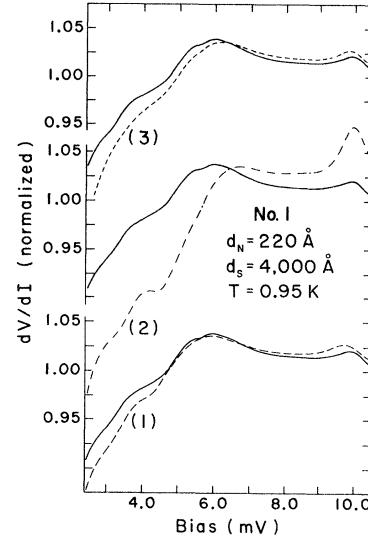


FIG. 1. Absolute ac resistance measurements, calibrated by the substitution resistor method and normalized to unity at high bias, are depicted by the repeated solid curve. Dashed curves represent three theoretical alternatives: (1) Arnold's theory of elastic scattering combined with Gallagher's theory of compound resonances, (2) Gallagher's theory by itself (see Ref. 24), and (3) Arnold's theory by itself.

venience) represent experimental results produced by 220 Å of Ag backed by 4000 Å of Pb, and dashed curves represent three theoretical alternatives: (1) Arnold's theory of elastic scattering combined with Gallagher's theory of compound resonances, (2) Gallagher's theory by itself,²⁴ and (3) Arnold's theory by itself. In the sense that curve (1) results from combining the actual parameter values (Table I)²⁵ used for (2) and (3), the latter two alternatives represent constituent contributions of each theory to the final result (1). Although the Gallagher contri-

TABLE I. Parameter values used to compute ac diode resistances.

d_N (Ag) ^a (Å)	d_S (Pb) ^a (Å)	$\Delta_N(0)$ ^b (meV)	$\Delta_S(0)$ ^b (meV)	l_N (Å)	l_S (Å)	v_{FN}^* (Ag) (10 ⁶ m/s)	v_{FS}^* (Pb) (10 ⁶ m/s)
100	4100	1.24	1.30	92	2730	1.07	0.75
130	3200	1.25	1.30	144	2130	1.08	0.72
220	4000	1.09	1.29	220	2670	1.07	0.77
350	4500	0.83	1.26	233	2650	1.07	0.61
510	4500	0.65	1.23	392	2250	1.07	0.76
700	4100	0.56	1.12	778	1780	1.06	0.73

^a Values are determined experimentally.

^b Values are determined experimentally from $I(V)$; see Ref. 25 for their use.

bution (2) accounts for compound resonance structure ($V < 4.5$ mV), its Pb transverse-phonon peak occurs at too high a bias ($V \approx 6.7$ mV). Conversely, the Arnold contribution (3) does better with the transverse-phonon peak, whose bias is now only slightly higher than observed, but fails with the compound resonance structure. Together these two theories yield reasonably good agreement for $d_N = 220$ Å, except for a small resistance dip missing near 5.5 mV and for a small bias shift of the Pb longitudinal-phonon peak near 9.5 mV. The agreement depicted in Figs. 2 and 3, for thinner ($d_N = 110$ Å) and thicker ($d_N = 510$ Å) Ag layers, is better.

Table I lists dressed velocities for Ag that differ somewhat from the nominal value (1.09×10^6 m/s) anticipated from the band velocity (1.25×10^6 m/s)¹³ and nominal intrinsic renormalization (1.15) that we assume initially. These differences represent minor adjustments used to fine tune our theoretical fits and, on the average, suggest either a somewhat smaller band velocity (1.235×10^6 m/s) or a larger renormalization (1.165). Our Ag films yield an average dressed velocity of $(1.07 \pm 0.06) \times 10^6$ m/s.

For Pb the anticipated dressed velocity (0.76×10^6 m/s)^{9,14,15} and the average from Table I, $(0.72 \pm 0.06) \times 10^6$ m/s, differ but little and agree with the recent value, $(0.78 \pm 0.04) \times 10^6$ m/s, inferred from compound resonances in Zn-Pb proximity sandwiches.³

Table I also documents a striking correlation between the mfp for elastic scattering (l_N) and the Ag layer thickness (d_N): namely, l_N tends to de-

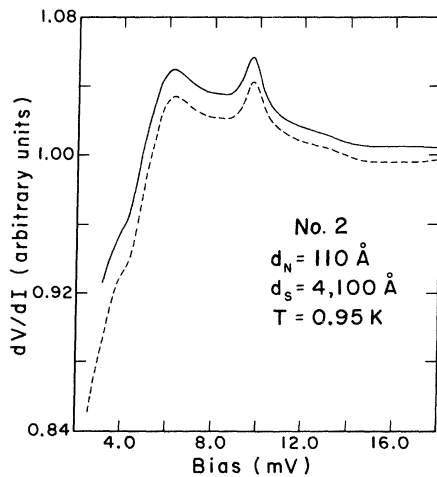


FIG. 2. Uncalibrated ac resistance measurements are depicted by the solid curve. The dashed curve represents a theoretical fit obtained with the combined theories. Vestigial compound resonance structure occurs just below 4.0 meV.

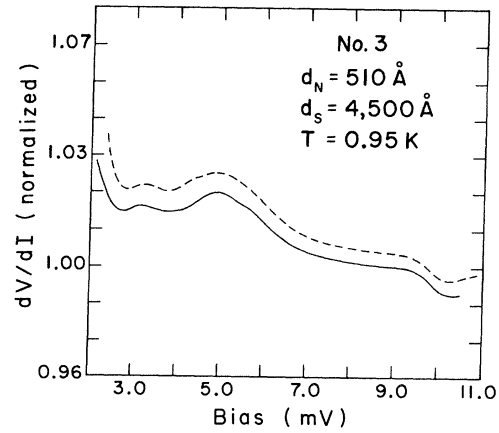


FIG. 3. Absolute, normalized ac resistance measurements are depicted by the solid curve. The dashed curve represents a theoretical fit obtained with the combined theories.

crease with decreasing d_N so that, on the average, $d_N/l_N = (1.1 \pm 0.2)$. We are unable to offer plausible reasons for this behavior, since elastic scattering presumably depends on the volume density of defects. Although incoherent scattering at the Ag-Pb interface leads naturally to $l_N \approx d_N$, we know of no *a priori* reason for believing that surface scattering closely mimics volume scattering, especially to the extent of yielding appropriate numerical values for l_N . This aspect of our work requires further study.

Our central conclusion is that Arnold's theory provides a natural account of the Ag-Pb proximity tunneling results⁸ without the *ad hoc* introduction of enhanced effective renormalizations.⁸ For sufficiently thin N -metal layers, according to Arnold's theory, elastic scattering substantially alters the renormalization behavior of the N -metal layer, even though the intrinsic electron-phonon interaction remains characteristic of the bulk. In addition to changes in the effective renormalization, Arnold's theory also produces substantial changes in the complex pairing potential, so that resultant structure in the density of states depends on a complicated interplay between these two factors as a function of energy. Consequently, there is no very direct connection between Arnold's effective layer renormalization and the empirical renormalizations inferred by Khim and Tomasch.⁸ In the extreme thin limit and in the presence of extreme scattering ($d_N/l_N \geq 3$), Arnold's theory predicts a gap dependence on d_N coinciding with that given by McMillan's proximity effect theory.²⁶ This may account for Khim and Tomasch's

success in fitting observed gap values with McMillan's theory even though their Ag-Pb sandwiches violated the weakly coupled layer aspect of the theory. Finally, use of clean, thick Pb backings can introduce significant compound resonance structure, even for Ag layers as thin as 100–200 Å.

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²³All theoretical diode characteristics are computed for specular tunneling by the methods of Ref. 2.
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²⁵ $\Delta_N(E)$ and $Z_N(E)$ are conveniently calculated only for $E > \Delta_S^{\text{ph}}(E)$ ($=1.4$ meV), but their numerical values are needed down to $E=0$ when computing ac resistances from $N(E)$. Below $E=2.0$ meV, we approximate $\Delta_N(E)$ by the experimental gap value (Table I), inferred from tunneling, and $Z_N(E)$ by its computed value at 2.0 meV. Owing to the minor influence of $N(E < 2.0$ meV) on ac resistances in the range of interest ($eV \approx E > 3$ meV), computed ac resistances in this range are reasonably insensitive to these approximations. Also, when computing $N(E)$, we scale $\Delta_{SO}^{\text{ph}}(E)$ values taken from Ref. 9 by the ratio $[\Delta_S(0)/1.4] \sim 0.9$, where $\Delta_S(0)$ now denotes the experimental gap value (Table I) inferred from tunneling and 1.40 meV is the gap edge adopted in Ref. 9.
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