[3]Misprints in Ref. 1, Eq. (16), have been corrected [L. R. Flax (private communication)].
[4]P. D. Loly, J. Phys. C 4, 1365 (1971).
[5]F. J. Dyson, Phys. Rev. 102, 1230 (1956).
[6]P. D. Loly and B. J. S. Huett (unpublished).

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Surface-Plasmon Excitation by Electron Tunneling*†

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The role of various parameters in our theory of inelastic-electron tunneling associated with the excitation of surface plasmons is clarified in order to refute Duke's criticism. It is shown here that our theory involves only one adjustable parameter, which can be determined independently from the random-phase-approximation analysis of surface plasmons. Hence there are no adjustable parameters in fitting theory to experiment. We have fortified the claim of agreement between our theory and experiment by a calculation of the background, and a detailed comparison with raw experimental data. Predictions from both the inelastic surface-plasmon excitation model and the electron–bulk-plasmon interaction model are compared. The surface-plasmon excitation model provides a far superior fit to the experimental data.

Recently carrier-concentration-dependent broad structures in the d^2I/dV^2 characteristics of n-type GaAs–metal tunnel junctions have been observed independently by Tsui[1] and by Duke *et al.*[2] These observations have been interpreted as being caused by the mechanism of inelastic tunneling associated with the excitation of surface plasmons (SP) in the GaAs electron by Tsui.[1] Subsequently, Ngai *et al.*[3] have presented a theory of inelastic-electron surface-plasmon interactions in metal-semiconductor junctions. Their calculated inelastic SP emission structure in d^2I/dV^2 agrees with experiments[1,2] both in magnitude and line shape. A different point of view was taken by Duke *et al.*,[2] who identified such structures with self-energy effects in the GaAs electrode resulting from electron–bulk-plasmon interaction. In a recent paper,[4] Duke has criticized the reliability of the bulk-plasmon (BP) energy as arrived at by Tsui,[1] using values of the electron concentration n obtained from sample suppliers. Such doubts have been removed when Tsui and Barker[5] subsequently made infrared-reflectivity measurements to determine directly the BP energies of the GaAs samples. Their results therefore fortified the interpretation of excitation of surface plasmons in the GaAs electrode by tunneling electrodes. Furthermore, Duke,[4] though acknowledging that the line shapes as calculated by Ngai *et al.*[3] look much better than those calculated by Duke *et al.*,[2] stated that "Ngai *et al.*[3] permitted themselves the luxuries of five adjustable parameters ($\gamma, \alpha, k_{c2}, d$, and ϕ_{eff}) and an arbitrary background subtraction procedure...." One of the purposes of this communication is to refute this statement by clarifying the roles of the parameters and by showing that the theory involves essentially *one* adjustable parameter for determination of the line shape. Moreover, its value as determined by comparison with experiment[1] agrees with the prediction of the random-phase-approximation (RPA) analysis of surface plasmons.[6]

In our treatment of the inelastic current associated with SP emission, we used an average-square-barrier model[7] to represent the tunnel junction at biases in a relatively narrow energy range about the SP energy. ϕ_{eff} and d are *fixed* parameters that enter into this model description of one-electron tunneling. Their chosen values ($\phi_{eff} \approx \frac{1}{2}$ eV, $d \approx 100$ Å) are inferred from independent measurements. γ, α, and k_{c2} are parameters characteristic of SP. Plasmons of small wavelength can decay through energetically possible electron-hole-pair excitations and the result is Landau damping. For SP, Landau damping (LD) plays a vital role. The reason is that an SP oscillation of surface wave vector $\vec{Q}$ cannot be a plane wave along the direction normal to the surface and consequently the eigenfunctions along this direction contain components of very small wavelength, which are responsible for increased damping. The microscopic theory of SP[6] indicates that LD introduces a finite imaginary part to the SP frequency which is linear in Q. Introducing a characteristic wave number Q_c defined by the equation $Q_c = \omega_s/v_F$, where ω_s is the plasma energy and v_F is the Fermi velocity, we expressed[3] this result as

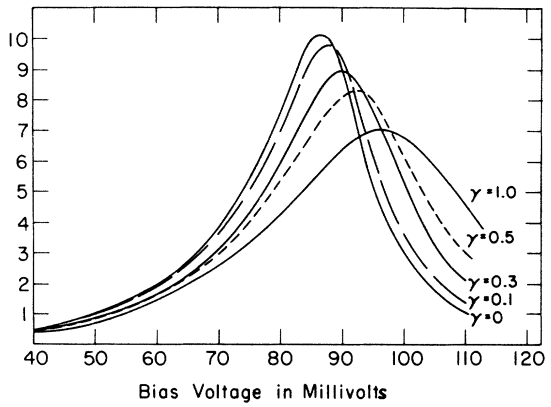

FIG. 1. Line shapes for various values of γ as indicated in figure. α is fixed at the value required by the RPA analysis. The electron concentration is $n=9.5\times10^{18}/\mathrm{cm}^3$.

$$\mathrm{Im}\omega_Q = \alpha Q/Q_c \ . \tag{1}$$

Besides the introduction of LD, the microscopic theory will yield a modified $\mathrm{Re}\omega_Q$. The RPA treatment of SP by Feibelman[6] has shown that the correction to $\mathrm{Re}\omega_Q = 2^{-1/2}\,\omega_s(1-e^{-2Qd})^{1/2}$ is of second order in Q/Q_c and thus negligible in comparison with the LD effects in $\mathrm{Im}\omega_Q$. If we put

$$\mathrm{Re}\omega_Q = 2^{-1/2}\omega_s(1-e^{-2Qd})^{1/2}(1+\gamma Q/Q_c)^{1/2} , \tag{2}$$

as we did in Ref. 3, then the RPA analysis implies $\gamma=0$. All the line shapes as reported in Ref. 3 were calculated with $\gamma=0$ in accordance with the result of the RPA analysis.[6] We defined[3] Q_{c2} (same as k_{c2} in Ref. 3) by the condition $\mathrm{Im}\omega_{Q_{c2}} = 0.2\,\mathrm{Re}\omega_{Q_{c2}}$. The results are relatively insensitive to the exact numerical factor entering the last equation, as long as this factor is of the order of unity. From Eqs. (1) and (2) it follows that Q_{c2} and α are interrelated. Since the LD is a monotonic function of Q, the SP of wave vector $Q>Q_{c2}$ are not well-defined modes of excitation. Thus contributions to the inelastic current from these SP can be neglected. Consequently we have, within the present model of SP excitations, a theory for the line shape which involves only *one* adjustable parameter, namely, α.

The dashed curve in Fig. 1 as well as in Fig. 2 of Ref. 3 in Fig. 2 has been obtained by subtracting a background from the experimental curve. This background, although not exactly known, is not arbitrary. It is smooth and it coincides with the experimentally observed curve when the difference $|\omega-\omega_s/\sqrt{2}|$ is larger than 20–30 meV. These restrictions specify the background within certain limits which have been estimated and indicated by an error bar in Fig. 2 of Ref. 3.

The one-parameter fit between theory[3,8] and experiment[1] (after the background subtraction) has been made[3] and the best fit corresponds to $\alpha/\omega_s \approx 0.035$, for all different samples studied by Tsui.[1] On the other hand, the RPA analysis of Feibelman[6] predicts that the LD of SP is of size

$$\mathrm{Im}\omega_Q = \left(\frac{\pi}{2^{15/4}}\right)\left(\frac{Q}{Q_s}\right)\left(\frac{\omega_s}{2^{1/2}}\right) , \tag{3}$$

where $Q_s=(2m\omega_s/\hbar)^{1/2}$. This can be rewritten as $\mathrm{Im}\omega_Q=\alpha_F Q/Q_c$ with $\alpha_F=(\pi/2^{17/4})(Q_c/Q_s)\omega_s$. From the data[1] both Q_c and Q_s can be calculated. We find $\alpha_F/\omega_s\approx0.03$, which is in excellent agreement with the value of $\alpha/\omega_s\approx0.035$ that gives the best fit to the data for all samples. To demonstrate further the unimportance of γ, which we have put equal to zero in all previous line-shape calculations, we here let γ vary over a large interval while keeping $\alpha/\omega_s\approx0.035$, a constant throughout. The line shapes are shown in Fig. 1. It can be seen that for γ as high as 0.1 the result remains almost unchanged.

In order to demonstrate that the agreement between theory and experiment as shown in Fig. 2 of Ref. 3 is not due to a "convenient" choice of the background, we summarize the results of a calculation of the background d^2I/dV^2, based on either of the two equivalent models for the barrier penetration factor D, i.e.,

$$D=(D_0/E_0)(\hbar^2k_\parallel^2/2m) \tag{4a}$$

or

$$D=(D_0/2E_0)(\xi_k+\zeta) \ . \tag{4b}$$

In Eqs. (4) D_0 is the barrier penetration factor of an electron at the Fermi energy for $V=0$, E_0 is a quantity with units of energy $E_0\sim\hbar\omega_p/2$, $k_\parallel$ is the component of the electron momentum parallel to the plane of the junction, ζ is the Fermi energy, $\zeta=\hbar^2k_F^2/2m^*$, and $\xi_k=(\hbar^2k^2/2m^*)-\zeta$. Equations (4a) and (4b) for D are identical to those used by Duke *et al.* in Ref. 2. As they note,[2] models (4a) and (4b) adequately describe the "background" characteristics of the data.

Using Eqs. (4) and following Duke *et al.*[2] (but without including the effects of the electron–bulk-plasmon interaction) one obtains for the background conductance, dI/dV,

$$dI/dV=(\mathrm{const})(\zeta+eV)^2 \ . \tag{5}$$

The background d^2I/dV^2 is given by

$$d^2I/dV^2=(\mathrm{const})(\zeta+eV) \ . \tag{6}$$

Consequently, the background d^2I/dV^2 is uniquely determined by Eq. (6) and by the observation that it must coincide with the raw experimental data when the difference $|\omega-\omega_s/\sqrt{2}|$ is larger than 20–30 meV. We have compared the experimental results (after the subtraction of the background

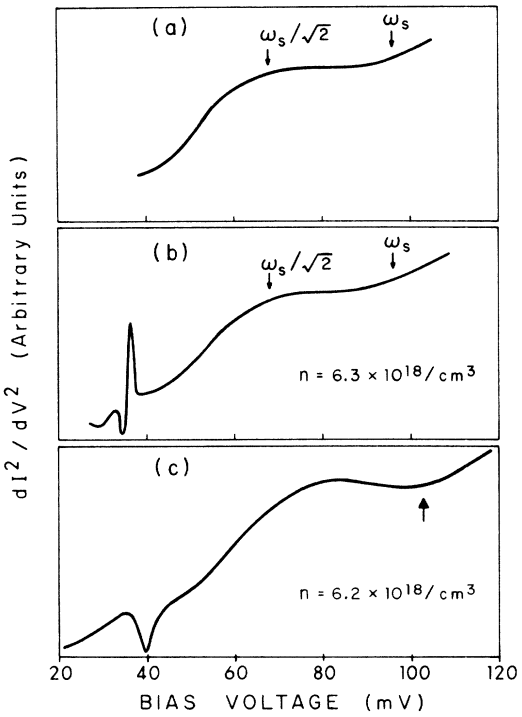

FIG. 2. (a) The $(d^2I/dV^2)_{\text{total}}$ calculated from Eq. (6) of the present work and Eq. (4) of Ref. 3 for $\hbar\omega_s = 96$ meV. The values of the parameters are $\alpha = 0.035\omega_s$, $\phi_{\text{eff}} = \frac{1}{2}$ eV, $d = 100$ Å. The results are relatively insensitive to the values of γ. (b) Tsui's data (Ref. 5) for $(d^2I/dV^2)_{\text{total}}$ from an n-type GaAs sample labeled $n = 6.3 \times 10^{18}/\text{cm}^3$; ω_s is the bulk-plasmon energy of the GaAs electrode determined optically at 78 °K. (c) Data of Duke *et al.* (Ref. 2) for $(d^2I/dV^2)_{\text{total}}$ from an n-type GaAs sample labeled $n = 6.2 \times 10^{18}/\text{cm}^3$; the arrows indicate the position of the infrared-reflectivity minimum measured on the tunneling sample at room temperature.

determined as explained above) with our theoretical results for $d^2\delta I/dV^2$ as modified when one uses in Eq. (4) of Ref. 3 the value of dI/dV given by (5). The changes from the results shown in Fig. 2 of Ref. 3 are minor and tend to improve the agreement between theory and experiment.

In Fig. 2 we present for comparison some of the raw experimental data of Tsui[1,5] and Duke *et al.*[2] together with our theoretical calculations for $(d^2I/dV^2)_{\text{total}} \equiv d^2I/dV^2 + d^2\delta I/dV^2$, where d^2I/dV^2 is given by Eq. (6) and $d^2\delta I/dV^2$ by Eq. (4) of Ref. 3. It should be noted that the theory of Duke *et al.* predicts a characteristic structure (a dip followed by a bump as shown in Fig. 5 of Ref. 2) in $(d^2I/dV^2)_{\text{total}}$ located at $eV \approx 1.7\hbar\omega_p$. In addition, the magnitude of this characteristic structure seems to be inconsistent (much larger) with the magnitude of the observed structure (see Fig. 6 of Ref. 2).

Duke[4] has also criticized Tsui[1] for attributing his observation of "no systematic dependence of the half-width on the electron mobility of the sample" to "the nonideal boundary between the electron plasma and the tunneling barrier." This criticism is appropriate, but we can explain the observation in a natural way from the predictions of our detailed theory.[3,8] Tsui has observed a strong dependence of the half-width on the electron concentration but not on electron mobility. Our theory[3,8] indicates that the half-width is determined roughly by the sum of electron collision broadening and LD at Q near Q_{c2} (indeed at such high values of Q, LD dominates over collision broadening). It is clear from Eq. (3) that the size of the LD depends linearly on ω_s, i.e., is proportional to $(n)^{1/2}$. Indeed, a plot of the measured half-widths from Tsui's data versus $n^{1/2}$ exhibits a linear behavior.[3,8]

In summary, by attributing the observed structure to SP excitations we were able to explain all the relevant experimental features. As we have shown here, our theory involves essentially one adjustable parameter (α) which can be determined independently from the RPA analysis of surface plasmons.[6] On the other hand, the alternative explanation attributing the observed structure to BP-electron interactions in the electrode predicts a minimum in d^2I/dV^2 at an energy $\hbar\omega_p(q_0)$, where q_0 is the maximum BP wave vector,[4] in disagreement with the available experimental results.[2,3] Thus the SP excitation picture appears to be the natural explanation for both Tsui's and Duke *et al.*'s data.

In conclusion, it should be noted that the calculation of the electron self-energy by Duke *et al.* suffers from two unjustified approximations: (i) replacing the vertex part Γ by unity[9] and (ii) replacing the electron propagator by the free-electron propagator. As has been pointed out by Lundqvist,[9] who used approximations (i) and (ii), it is very difficult to justify approximation (i) since little is known about the vertex part Γ for energy transfer of the order of $\hbar\omega_p$. Recently Pendry[10] has demonstrated by summing ladder diagrams that the effective electron-BP coupling will be reduced as a result of his calculation. Regarding the second approximation, it seems to be inconsistent with Duke *et al.*'s results; they find that the electron propagator is drastically different from the free-electron propagator.

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[1] D. C. Tsui, Phys. Rev. Letters 22, 293 (1969).

[2] C. B. Duke, M. J. Rice, and F. Steinrisser, Phys. Rev. 181, 733 (1969).

[3] K. L. Ngai, E. N. Economou, and M. H. Cohen,

Phys. Rev. Letters 22, 1375 (1969).
[4]C. B. Duke, Phys. Rev. 186, 588 (1969).
[5]D. C. Tsui and A. S. Barker, Jr., Phys. Rev. 186, 590 (1969).
[6]P. J. Feibelman, Phys. Rev. 176, 551 (1968).
[7]A. J. Bennett, C. B. Duke, and S. D. Silverstein, Phys. Rev. 176, 969 (1968).
[8]K. L. Ngai and E. N. Economou, Phys. Rev. B (to be published).
[9]B. I. Lundqvist, Physik Kondensierten Materie 6, 193 (1967).
[10]J. B. Pendry, J. Phys. C 3, 1711 (1970).

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Temperature Dependence of the Surface-Sheath Nucleation Field in Strong-Coupling Superconductors

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In an earlier paper the temperature dependence of H_{c3} in a strong-coupling type-I superconductor was determined and compared with the theory of Eilenberger and Ambegaokar (EA). An error in the EA theory is now taken into account. The dc resistivity appropriate to the surface-sheath region has been obtained by a more consistent method. When these new factors are considered, the results suggest that the discrepancy between theory and experiment is due to a more fundamental reason than just experimental inaccuracy.

Recently[1] we have presented data on the temperature variation of the surface-sheath nucleation field (H_{c3}), in a strong-coupling type-I superconductor (0.25 at.% Bi in Pb). In that paper we compared our experimentally determined value for the slope $(dH_{c3}/dT)|_{T_c}$ (− 234 G/°K) with a prediction for that quantity based on the theory developed by Eilenberger and Ambegaokar[2] (EA) for this material (− 188 G/°K). The latter is a phenomenological theory in which the Landau-Ginzburg equations have been generalized to include strong-coupling effects. We attributed most of the discrepancy between theory and experiment to possible uncertainty in several experimental parameters which must be put into the EA equations in order to extract a value for the slope. In particular, we noted that since we had no direct means for determining a value for the dc conductivity in the surface sheath, of necessity we used the conductivity of the bulk material, which is easily measurable. Since the presence of surface scattering processes would make the resistivity in the surface larger than that of the bulk, we were aware that our calculated slope could be expected to be too small, as was the case.

It is the purpose of this paper to point out several subsequent developments which relate to the results and discussion in the above-mentioned paper. We find that, when these new factors are considered, the disagreement between theory and experiment is somewhat increased, and we now believe that the discrepancy is of a more fundamental nature than had previously been supposed.

Of primary significance is a recent paper[3] by EA in which they pointed out an error in their original work. As a result, each of their predicted values for $(dH_{c3}/dT)|_{T_c}$ should be decreased by the factor 0.68. The authors pointed out that the fairly good agreement which had been demonstrated between the theory and data taken on pure lead, must now be considered as being spurious. The same correction factor would apply to our material.

We have also managed to make progress toward obtaining a value for the dc conductivity in the surface of our alloy. We have used the following result by Goodman[4] which relates the Ginzburg-Landau parameter κ in dilute alloys to that of the pure material through the dc resistivity:

$$\kappa = \kappa_0 + 7.5\times 10^3 \rho \gamma^{1/2}. \quad (1)$$

Here, the subscript refers to pure lead, ρ is the dc resistivity, and γ is the coefficient of the electronic specific heat. We have determined κ from our data by plotting $\kappa(T) = H_{c3}(T)/2.4H_c(T)$ vs T, and extrapolating to T_c. Several authors have measured $H_{c3}(T)$ in pure lead,[5−10] so that it should be possible to determine κ_0 from their data by the same method. However, these measurements disagree somewhat with each other. This disagreement is presumably due to differences in the surface conditions of the samples used. To obtain a value relevant to our work we have measured $\kappa(T)$ in a pure single-crystal lead sample which had been carefully prepared in a manner identical to that used in the case of our alloy. Thus, the sur-