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PHYSICAL REVIEW B VOLUME 4, NUMBER 7 1 OCTOBER 1971

Theory of Electron-Surface-Plasmons Interactions in Tunneling, Low-Energy-Electron Diffraction, and Photoemission*

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A general theory of surface-plasmon excitation by electrons in the cases of electron tunneling, low-energy-electron diffraction, and photoemission is developed. Particular emphasis has been given to the physical concepts related to the electron-surface-plasmon interaction. The inelastic-tunneling current due to surface-plasmon emission in semiconductor-metal junctions has been calculated in detail. The surface-plasmon contributions to the inelastic spectra in low-energy-electron diffraction and to the energy distribution curves of photoemitted electrons have also been evaluated. The theory involves essentially a single parameter which determines the magnitude of the Landau damping of the surface plasmons. Using the value for this parameter as deduced from Feibelman's theory, we obtain in all cases good agreement with available experimental results, both in magnitude and line shape.

I. INTRODUCTION

A well-known property of an electron gas is its capacity to undergo collective motions, i.e., plasma oscillations (PO). The properties of these oscillations in the case of one material extending over the whole space were the subject of considerable experimental and theoretical work.[1-3] In addition to these bulk plasma oscillations (BPO), the existence of surfaces separating materials with different electronic properties introduces new modes of plasma oscillations localized around the surfaces which are called surface-plasma oscillations[4] (SPO) or surface plasmons (SP). The SPO can interact with electrons (or charged particles in general) and photons and consequently may show a considerable influence on the observable characteristics of many systems. Thus the existence of SPO has been proved experimentally, through measurement of electron energy loss,[5] u.v. radiation,[6] transition radiation,[7,8] low-energy-electron diffraction (LEED),[9,10] photoemission,[11] and superconducting tunneling.[12-14] Recently, excitations of plasmons in a degenerate semiconductor by tunneling electrons have been observed in metal-semiconductor junctions[15,16] although there is some disagreement about the interpretation of the experiments.[15-17]

There are some basic differences between bulk and surface plasmons[4]: (a) In BP, charge density is different from zero inside the material while in SP only surface charge density exists. (b) The BP are accompanied by electrostatic fields [i.e., no coupling to the transverse part of the electromagnetic (em) fields] while the fields created by SP show a mixed character having both transverse and longitudinal components. (c) The dispersion relation for BP is generally determined only by the properties of the material while for SP the geometry plays a dominant role in determining their dispersion relations. (d) The electrostatic fields associated with BP are confined strictly inside the material where the charge oscillations take place while the fields created by SP are extended outside the materials responsible for their existence.

Plasmons exist as well-defined collective excitations as long as the wavelength of the oscillations is longer than a characteristic length of the order of magnitude of the interelectronic separation. For

smaller wavelengths, the electron gas cannot be considered, even approximately, as a continuum and consequently the plasmons, being collective excitations of the whole system, have no meaning at all. Another way of saying this is that plasmons of very small wavelength can decay through energetically possible electron-hole-pair excitations and that the resulting Landau damping (LD) is so strong that the plasmons cease to exist at all even as approximate eigenstates of the system. It will be shown later that the LD of plasma oscillations plays an important role and that SPO are more heavily damped than BP. The reason is that the SPO 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 the increased damping.

To formulate plasmon excitation by external charge particles we start from the interaction energy between the external charged particle and PO. Knowing the em fields which accompany the PO, then the interaction energy is the scalar potential multiplied by the charge of the particle. The next step in the formalism[17] is the standard procedure of quantizing the fields, i.e., of expressing the amplitudes of PO and their fields in terms of creation and annihilation operators for the PO; thus a final expression of the standard form for the interaction energy results. This approach elucidates why a charged particle cannot excite BP as long as it is outside the material, since the fields of BP are limited to the region occupied by the material only. This is an important distinction between BP and SP and it may explain why there is no structure at the BP eigenfrequency in experiments involving low-energy electrons spending a very short time inside the material.

In this paper we shall examine the interaction of low-energy electrons with the SPO. This will include the normal tunneling case (the superconducting case is different[14]; because of the Josephson effect; the ac Josephson current is much more effective in exciting SP than individual electrons are), the low-energy-electron diffraction, and the photoemission problem. All these problems have the common characteristic of depending on those low-energy electrons (of the order of eV) which are near the surface of the considered material. Thus we expect that SP will play an important role in the observable quantities related with these processes.

The junctions for the tunneling experiments under consideration are either S-M (semiconductor-metal) or S-O-M (semiconductor-oxide-metal).[18,19] Experiments have been performed recently by Tsui[15] (in n-type GaAs-Pb surface-barrier tunnel junctions) and by Duke *et al.*[16] (in n-type GaAs-In contacts). Tsui has observed a structure in d^2I/dV^2 which corresponds to an increase in conductance at bias voltages near the surface-plasmon energy in GaAs. These measurements lead Tsui to explain his observations as due to the excitation of surface plasmons in GaAs by tunneling electrons. Duke, Rice, and Steinrisser[16] have also reported the observation of a broad doping-dependent structure in the d^2I/dV^2 characteristic very similar to that of Tsui's. They associate this structure with electron-bulk-plasmon interaction in the GaAs electrode. However, the position and the shape of the structure in d^2I/dV^2 resulting from their theoretical calculation do not agree with the experimental results.[16] We believe that the origin of the aforementioned tunneling anomaly is not due to BP,[20] and that the strong structure calculated by Duke *et al.*[16] is spurious arising from an overestimation of the electron (el)-BP interaction.[20] We think that a similar overestimation of the el-BP interaction is present in the work of Hedin *et al.*[21]

We summarize here the well-established[3,22] physical picture of the nature of the ground state of an interacting electron system in order to elucidate the mechanism by which a very weak el-plasmon interaction results.

The long-range nature of the Coulomb force is responsible for creating correlations in the position of the electrons. These correlations are manifested as quantum zero-point plasma oscillations which correspond to a depletion of electron density around each electron in such a way that the long tail of the Coulomb force of each electron is canceled. Thus the long range of the Coulomb force is responsible for setting up PO which in turn eliminate its long tail leaving an effective short-range field for each electron. This *residual* field gives rise to a very weak el-plasmon interaction whose only effect[22] is a very small renormalization in quantities like the electronic mass, the plasmon dispersion relation, etc.

We think that a similar physical picture holds even for highly excited states. If a particle (or hole) is introduced into the system, being initially unscreened, it can excite PO provided it is of sufficient energy. The excitation of PO in this case is simply the response of the system in its effort to oppose change by screening the additional charge. Once plasmons have been created, the long tail of the Coulomb force is ineffective owing to correlations as explained earlier. Thus again a weak residual el-plasmon interaction results.

If an electron is in the exterior of the system, in response to its long-range Coulomb field, surface charges appear which prevent the field of the particle from penetrating inside the material. This is equivalent to saying that a particle outside a system can excite only SP and not BP. A simple way to arrive at the same conclusion is to examine the

image force on a charged particle outside a conductor.[23] This force shows a resonance behavior at the SP eigenfrequency while no such behavior is exhibited at BP eigenfrequency.

The above discussion leads us to the following physical picture for the mechanism of PO excitation in electron tunneling: As long as the electron is inside the electrode, it experiences a weak residual interaction with both BP and SP, so weak that it is unable to produce any observable structure in the electron self-energy. When the electron finds itself outside the material, its field is not screened any more by the other electrons and consequently it polarizes the material by setting up surface charge density, a process which is nothing else but SP excitations by the electron in tunneling. The inelastic excitation induces an increase in the tunneling conductance,[17,24] which has been calculated[17] and agrees well with the experimental results.[15,16] Moreover, the theoretical model explains many features of the experimental observation.

The role of SP in the diffraction of low-energy electrons is the next subject to be examined here. For electrons of energy less than 100 eV, the atomic scattering cross sections are typically of about the same magnitude as the area occupied by an atom in a single atomic layer.[25] A single atomic layer is, therefore, a very efficient scatterer for low-energy electrons. Since the incoming electron is spending most of its time very close to the surface, one expects relatively large cross sections for SP excitations. Recent measurements[9,10] with LEED in a tungsten-cesium system [Cs evaporated on a (100) face of tungsten] have verified that the SP excitations have a high cross section. MacRae *et al.*[10] have interpreted their data as providing evidence for nonmetallic character of layers of low electron density due to a Wigner-Mott state.

A theory for calculating inelastic el-SP scattering in LEED can be constructed if we think of our problem as the scattering of electrons under the combined influence of two potentials U and V. U represents all the elastic and inelastic processes other than SP scattering and V is the el-SP interaction. The calculations[26] show agreement with the experiments as regards the position, width, and strength of the SP peaks.[10] The behavior of the total backscattered electrons with 1-SP emission as a function of the thickness d of the Cs layer is also in qualitative agreement with experiments.[9,10] Specifically, the observed transition from no SP contributions to strong SP contributions as the Cs coverage increases is in accordance with our theoretical results.

The theory can easily be extended to inelastic SPO excitation in photemission. We adopt here an approach similar to that of Ref. 11 accepting the three-step model[11,27] as a conveniently simple framework for performing our calculation.

Experimental evidence of structure due to plasmons (both SP and BP) has been observed by Vehse *et al.*,[28] by Callcott and MacRae,[29] and more recently by Smith and Spicer.[11] In the latter experiments the importance of SP is evident. Thus for Cs and Rb the observed structure fits quite well with that due to SP. In addition a broad peak at an energy corresponding to two SP has been observed in Cs agreeing well with the theoretical predictions of the present analysis. An unambiguous way of identifying the origin of the plasma structure would be to look at the angular dependence of the observed broad peaks; the strength of the peaks due to SP, in contrast to those due to BP, should depend strongly on the angle that the electron trajectory makes with the surface.

In Sec. II the properties of SPO in geometries related with the present work are discussed. The dispersion relations are derived and the damping of the modes is examined. The role of certain simplifying assumptions is illuminated by comparison with more realistic situations. Subsequently, in Sec. III, the SP fields derived in Sec. II are quantized and thus the el-SP interaction is given in a second quantized form for each of the cases under consideration. In Sec. IV the role of the el-SP interaction in metal-semiconductor junctions is studied. The resulting structure in the second derivative of the tunneling current with respect to the applied voltage is derived and is compared with existing experimental results. In Sec. V the role of the derived el-SP interaction in LEED and in photoemission is examined. The resulting modifications on the inelastic spectra in LEED and on the energy distribution curves (EDC) in photoemission are compared with available experimental results.

II. SURFACE-PLASMA OSCILLATIONS

The investigation of plasma oscillations is based[4] upon Maxwell's equations together with a constitutive equation relating the vector $\vec{D}$ with the electric field $\vec{E}$. For the purposes of the present paper, electrostatic considerations suffice for an adequate description of the phenomena, as will be shown later. However, for the sake of completeness and in order to justify this approximation, we base our analysis on Maxwell's equations which, with the magnetic permeability μ taken equal to unity, can be written as

$$\vec{\nabla}\cdot\vec{D}=0\,, \tag{2.1}$$

$$\vec{\nabla}\cdot\vec{H}=0\,, \tag{2.2}$$

$$\vec{\nabla}\times\vec{E}=\frac{1}{c}\frac{\partial\vec{H}}{\partial t}\,, \tag{2.3}$$

$$\vec{\nabla}\times\vec{H}=\frac{1}{c}\frac{\partial\vec{D}}{\partial t}\,, \tag{2.4}$$

and

$$\vec{D} = \epsilon(\omega)\vec{E}\,, \tag{2.5}$$

where the frequency-dependent, k-independent dielectric function $\epsilon(\omega)$ is assumed to be known. The set of Eqs. (2.1)–(2.4) takes the familiar form of conditions of continuity of certain components across the boundaries, where the properties of the medium change discontinuously. The type of solutions we seek correspond to wave propagation along a direction parallel to the boundary surfaces separating the different materials. With the x axis normal to these surfaces, we further assume that $H_x = H_\parallel = E_\perp = 0$ in all media, where the subscripts $\parallel$, $\perp$ denote the components parallel to the direction of propagation and normal to it as well as to the x axis, respectively. Thus, we restrict ourselves to the so-called "electric (or TM) waves" and we neglect the other group of possible solutions—the "magnetic (or TE) waves"—which, being purely transverse, are of no interest to us. The solution for any component of the fields can be written as

$$\mathcal{F}_{\vec{Q}}(x, \vec{R}, t) = \mathrm{Re} F_{\vec{Q}}(x) e^{i(\omega t - \vec{Q}\cdot\vec{R})}\,, \tag{2.6}$$

where $\vec{R}$ is the component of the position vector $\vec{r}$ parallel to the boundary surfaces and $\vec{Q}$ is the two-dimensional wave vector. Equations (2.1)–(2.5) can be written as

$$E_\parallel(x) = -\frac{i}{Q}\frac{dE_x}{dx}\,, \tag{2.7}$$

$$H_\perp(x) = [\omega\epsilon(\omega)/cQ]E_x\,, \tag{2.8}$$

$$\frac{d^2E_x}{dx^2} - K^2E_x = 0\,, \tag{2.9}$$

where

$$K = [Q^2 - \omega^2\epsilon(\omega)/c^2]^{1/2}, \quad \mathrm{Re}K > 0\,. \tag{2.10}$$

The general solution of (2.9) is a linear combination of the two independent solutions e^{Kx} and e^{-Kx}. We shall examine now separately the two multiple film geometries relevant to the problem under consideration.

A. Metal-Semiconductor Junction Geometry

We identify the semiconductor-metal junction as consisting of semiconductor S and metal M, both semi-infinite, separate by a dielectric medium I (Fig. 1). The latter is introduced in order to take into account the depletion layer of the junction. In reality, the electron density tails off to zero in the depletion layer from its value in the semiconductor in a continuous way and not abruptly as the above model assumes. Recently, Feibelman[30] was able to prove that the classical SP frequency $\omega_p/\sqrt{2}$ is insensitive to the electron density profile as long as the self-consistent Hartree density is used. Bennett[31] however has shown that the SP dispersion relation is affected by the profile. Even in a S-M junction, the SP of interest has oscillations mainly confined to the S-I interface (Fig. 1). From Feibelman's results we therefore expect the effect of density tailing to be small. In fact, for the particular case when the electron density decreases to zero through several steps in a distance l, exact solutions for SPO obtained show that the correction to the SP frequency is the $O(Ql)$. Typically, for Schottky barriers l is of $O(10\ \text{Å})$. Then, as we shall see in Sec. IV, for all Q of importance, Ql is small and hence corrections to the SP effects, as calculated from our idealized junction (Fig. 1), are negligible. We take the dielectric functions in the different regions to be[32,33]

$$M:\ \epsilon_M(\omega) = 1 - \frac{\omega_M^2}{\omega^2(1 - i\omega\tau_M)}\,, \tag{2.11a}$$

$$I:\ \epsilon_I(\omega) = \epsilon_\infty\,, \tag{2.11b}$$

$$S:\ \epsilon_S(\omega) = \epsilon_\infty\left(1 - \frac{\omega_S^2}{\omega^2}\frac{1}{1 - i/\omega\tau_S}\right)\,, \tag{2.11c}$$

where τ_M, τ_S are the relaxation time for the metal and semiconductor, respectively. At the frequencies of present interest and for the case of GaAs studied by Tsui, $\epsilon_\infty = 11.3$, and $\omega_S^2 = 4\pi e^2 n/m^*$, where $m^* = 0.07m$, and n is the electron concentration.

The solution that remains finite at infinity has the form

$$M:\ E_x = A_I e^{-K_M x}\,, \tag{2.12a}$$

$$I:\ E_x = A_{II}e^{-K_I x} + A_{III}e^{K_I x}\,, \tag{2.12b}$$

$$S:\ E_x = A_{IV}e^{+K_S x}\,, \tag{2.12c}$$

where K_M, K_I, K_S are given by (2.10) with $\epsilon(\omega)$ replaced by (2.11a), (2.11b), and (2.11c), respectively. The corresponding $E_\parallel$ and $H_\perp$ can be calculated by (2.7) and (2.8); the continuity of these fields across the boundaries gives a system of four linear homogeneous equations for the four unknowns A_I, A_{II}, A_{III}, A_{IV}. Nonzero solutions exist only when the determinant of the system is zero, a condition from which the eigenfrequencies of the SPO as functions of the parameter $\vec{Q}$ are determined. In the present case the vanishing of the determinant is equivalent to

$$R_S R_M - (R_M + R_S)\coth K_I d + 1 = 0\,, \tag{2.13}$$

where

$$R_S = -\epsilon_I K_S/\epsilon_S K_I\,, \tag{2.14a}$$

$$R_M = -\epsilon_I K_M/\epsilon_M K_I\,. \tag{2.14b}$$

For each value of Q, there are two solutions for ω of Eq. (2.13) corresponding to the two possible modes of oscillation in the considered system. These two branches are schematically shown in Fig. 1, under the assumption that $\omega\tau_S \gg 1$ for the frequencies of interest. The lower part of the upper

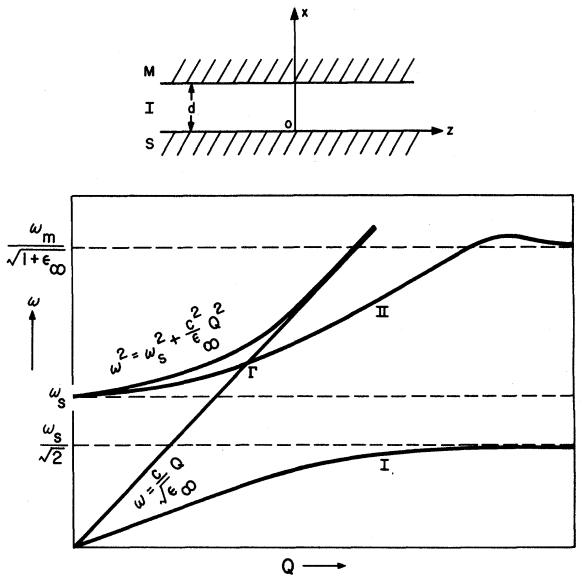

FIG. 1. Geometry and dispersion relations for SPO of an idealized metal-semiconductor junction. The analytic expressions for the curves I and II shown schematically here are derivable from (2.13). Γ corresponds to a frequency $\sim (\omega_M \omega_S)^{1/2}$.

branch (II) corresponds to oscillations of extremely small amplitude, and consequently is of no importance. Since we care for frequencies $\omega \sim \omega_S$, the upper part of the upper branch is of no interest to us. Thus only the lower branch (I) plays a role for frequencies $\omega \sim \omega_S$. The dispersion is linear for $Q \ll Q_S = \omega_S/c$, i.e.,

$$\omega = \bar{c} Q \ , \tag{2.15}$$

with $\bar{c}$ given by

$$\bar{c} = c[d/(d+\lambda_M+\lambda_S)\epsilon_\infty]^{1/2} \ , \tag{2.16}$$

where $\lambda_M \equiv 1/Q_M = c/\omega_M$, and $\lambda_S \equiv 1/Q_S = c/\omega_S$. It should be pointed out that a similar mode in superconducting tunneling junctions gives rise to the Fiske steps.[14] When $Q \gg Q_S$, the considered mode corresponds to charge oscillations confined mainly in the S-I interface and its dispersion relation is given as

$$\omega/\omega_S = (1/\sqrt{2})(1 - e^{-2Qd})^{1/2} \tag{2.17}$$

to the lowest order in ω_S^2/ω_M^2.

The results shown in Fig. 1 and Eqs. (2.15)–(2.17) are based upon the assumption that the dielectric functions are given by (2.11) with $\omega\tau_S$, $\omega\tau_M \gg 1$. This assumption is not valid at low frequencies. The reason is that for low frequencies, polar lattice vibrations strongly interact[32,33] with the electron gas in the semiconducting electrode thus modifying significantly $\epsilon_S(\omega)$ from the form assumed in (2.11c). Consequently, the results presented until now do not hold for frequencies below or about the maximum phonon frequency. However, the SP-induced structure in d^2I/dV^2 lies at higher frequencies, and thus it is not influenced by what happens at phonon frequencies. Since our purpose is to calculate the SP-induced structure only, we can neglect the lower part of branch I by introducing a lower cutoff. It should be pointed out that this lower cutoff *does not* affect at all our calculations regarding the position, strength, and shape of the SP-induced peak in d^2I/dV^2. Its only effect is to modify slightly the unimportant extreme lower tail of the peak.

As was explained in the Introduction, PO exist as well-defined oscillations only for wavelengths larger than a critical length of the order of the interelectronic separation. This is reflected in the formalism through the fact that $\epsilon(\omega, k)$ acquires an imaginary part which is quite significant as the wave vector becomes larger and larger. As a consequence, the SP frequency has a finite imaginary part and shall be written as $\omega(Q) = \mathrm{Re}\omega(Q) + i\,\mathrm{Im}\omega(Q)$. Several works on SP have appeared recently.[23,30,34,35] The results of these investigations imply that LD gives rise to an $\mathrm{Im}\omega(Q)$ which is linear in Q. Introducing a characteristic wave number Q_c defined by the equation $Q_c = \omega_S/v_F$ with v_F as the Fermi velocity, we express this result as

$$\mathrm{Im}\omega(Q) = \alpha\, Q/Q_c \ . \tag{2.18}$$

The proportionality constant α can be readily obtained by first principles by comparing (2.18) with Eq. (37) of Ref. 30 or by fitting experimental data as described later. Besides the introduction of LD, the microscopic theory will yield a modified $\mathrm{Re}\epsilon(\omega, k)$ and may change $\mathrm{Re}\omega(Q)$ from (2.17). However, the available treatment of Feibelman[30] has shown that the correction to $\mathrm{Re}\omega(Q)$ is of second order in Q/Q_c and thus negligible in comparison with the LD effects of $\mathrm{Im}\omega(Q)$. The discussion given here justifies our writing

$$\frac{\omega(Q)}{\omega_S} = \frac{1}{\sqrt{2}}\,[1 - e^{-2Qd}]^{1/2} + i\left(\frac{1}{2\tau_S\omega_S} + \alpha\,\frac{Q}{Q_c}\right) \tag{2.19}$$

for $Q_{c1} < Q < Q_{c2}$. As was explained, the lower cutoff Q_{c1} has been introduced for logical consistency only and has *no* quantitative effect on the results of our calculation. Damping of SPO increases as Q increases. Q_{c2} is defined by the condition that at Q_{c2}, $\mathrm{Im}\omega(Q_{c2}) = \mathrm{Re}\omega(Q_{c2})$, when SPO ceases to be a well-defined excitation. Obviously Q_{c2} is interrelated to α and thus (2.19) contains a single parameter only.

B. LEED and Photoemission Geometries

We idealize the Cs (cesium) absorbed on W (tungsten) as a film of thickness d bounded on one side

by a semi-infinite W plasma. The dielectric functions are taken to be

$$\text{W:}\quad \epsilon_W(\omega) = 1 - \frac{\omega_W^2}{\omega^2(1 - i/\omega\tau_W)}\,, \tag{2.20a}$$

$$\text{Cs:}\quad \epsilon_{Cs}(\omega) = 1 - \frac{\omega_{Cs}^2}{\omega^2(1 - i/\omega\tau_{Cs})}\,. \tag{2.20b}$$

Here $\omega_W(\tau_W)$ and $\omega_{Cs}(\tau_{Cs})$ are bulk plasma frequencies (relaxation times) for W and Cs, respectively. The dispersion of SP with inclusion of retardation effects can be obtained, in the same way as in Sec. IIA, through the solutions of the equation

$$R_I R_W - (R_I + R_W)\coth K_{Cs} d + 1 = 0\,, \tag{2.21}$$

where

$$R_I = -\epsilon_{Cs} K_I/\epsilon_I K_{Cs}\,, \tag{2.22a}$$

$$R_W = -\epsilon_{Cs} K_W/\epsilon_W K_{Cs}\,, \tag{2.22b}$$

and where K_I, K_{Cs}, K_W are given by (2.10) [with $\epsilon(\omega)$ replaced by ϵ_I], (2.20a), and (2.20b), respectively. The dispersion relations for SR are depicted in Fig. 2.[36] Retardation effects are not essential in our calculation because not only is the region $Q < \omega_{Cs}/C$ of Q space where they are important small, but also the associated SP fields are weak. The high-frequency mode plays no role for electrons with energy near 10 eV and will be excluded from further consideration. In the same manner as described in Sec. IIA, LD effects introduce an imaginary part to SPO frequency of the same form as that of (2.19).

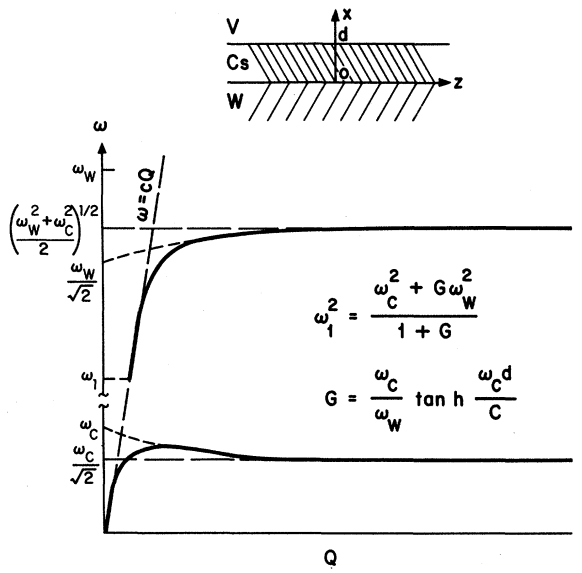

FIG. 2. Geometry and the dispersion relations for SPO of a vacuum-Cs-W system. The analytic expressions for the curves shown schematically here are derivable from (2.21). The ω_c in the legends is identical to ω_{Cs} in the text.

The SP dispersion as obtained in Fig. 2 for LEED geometry can also be used for the photoemission geometry if we let the Cs film thickness d tend to infinity. Henceforth we shall discuss, in parallel, both experimental situations for SPO by assuming that d can assume any value.

Finally we shall justify the approximation of considering even monolayers of Cs as a macroscopic film of thickness d and of dielectric function $\epsilon = 1 - \omega_{Cs}^2/\omega^2$. SPO as obtained from this point of view have been given for the tunneling and LEED geometries by (2.13) and (2.21), respectively. Let us consider (2.21) in the limit $d \to 0$, keeping the product $N = nd$ finite. Here n is the electron volume density and hence N can be interpreted as the number of electrons per unit area of a two-dimensional electron gas. For the W-Cs system we are thus examining the limit when the thickness of the Cs film tends to zero and can be expected to behave as a two-dimensional electron gas. (2.21) in this limit reduces to

$$\frac{4\pi N e^2}{m\omega^2} - \left(\frac{\epsilon_I}{K_I} + \frac{\epsilon_W}{K_W}\right) = 0\,. \tag{2.23}$$

On the other hand, the response of a two-dimensional electron gas to a longitudinal electric field of arbitrary wave vector and frequency has been calculated by Stern[37] in the self-consistent-field approximation.[38] The results have been used[37] to find the plasmon dispersion of a plane of electrons embedded in a three-dimensional dielectric. We have generalized this approach to the LEED geometry assuming the Cs is a plane ($x = 0$) of electrons. In the presence of a total field $\vec{E}(\vec{Q}, \omega) = \vec{E}_0\, e^{i\vec{Q}\cdot\vec{R} - i\omega t}$ in the plane, the induced field associated with the polarization of the electron-gas plane is proportional to

$$\exp(i\vec{Q}\cdot\vec{R} - i\omega t - K_I|x|)$$

if x is in the dielectric or to

$$\exp(i\vec{Q}\cdot\vec{R} - i\omega t - K_W|x|)$$

if x is the W region. Then the induced field in the plane $x = 0$ is found from Maxwell's equations to be

$$\vec{E}_{ind}(\vec{Q}, \omega) = -\frac{4\pi\chi(\vec{Q}, \omega)\vec{E}(\vec{Q}, \omega)}{\epsilon_I/K_I + \epsilon_W/K_W}\,, \tag{2.24}$$

where $\chi(\vec{Q}, \omega)$ is the polarizability which has been given by Stern in the self-consistent-field approximation.

We define the dielectric constant for this geometry by the relation $\epsilon(\vec{Q}, \omega) = (\vec{E} - \vec{E}_{ind})/\vec{E}$ and find

$$\epsilon(\vec{Q}, \omega) = 1 + \frac{4\pi\chi(\vec{Q}, \omega)}{(\epsilon_I/K_I + \epsilon_W/K_W)}\,. \tag{2.25}$$

The dependence of the plasmon frequency on $\vec{Q}$ is found from (2.25) by solving the equation $\epsilon(\vec{Q}, \omega) = 0$, which in the limit of large ω is identical to

(2.23) or can be checked by using Stern's expression[37] for $\chi(\vec{Q}, \omega)$.

III. QUANTIZATION OF SURFACE-PLASMON WAVE FIELDS AND ELECTRON-SP INTERACTION

In Sec. II we introduced the classical field theory of SPO. Just as in the case of the radiation field or the field of lattice vibrations in a solid, quantization of these fields gives rise to quanta which are photons and phonons, respectively, the SP wave field can be subjected to field quantization, and the resultant particle states are SP.

We consider a continuum model of an electron gas with a rigid fixed background of positive charge for any region where the conduction electron density is nonzero in our inhomogeneous system. The Lagrangian density in the ith metallic region is

$$\mathcal{L}(r_i) = (1/2n_i m_i)\,\vec{\pi}(\vec{r}_i)\cdot\vec{\pi}(\vec{r}_i) - \tfrac{1}{2}\delta\rho_i\phi(\vec{r}_i)\ ,$$

where $\vec{r}_i$ lies in the ith region. The total Lagrangian L of the system is $L=\sum_i \int \mathcal{L}(\vec{r}_i)\,d\vec{r}_i$. Here $\vec{\pi}(\vec{r}_i)$ is the momentum density and $\delta\rho_i(\vec{r}_i)$ the charge-density fluctuation from the equilibrium value $\rho_i = n_i e$ of the inhomogeneous electron gas at $\vec{r}_i$. $n_i(m_i)$ is the density (effective mass) of an electron in the ith region. $\phi(\vec{r}_i)$ is the SP wave field. Let $\vec{u}(\vec{r}_i)$ denote the vector displacement so that $\vec{\pi}(\vec{r}_i) = \partial\vec{u}(\vec{r}_i)/\partial t$ and

$$\delta\rho_i(\vec{r}_i)/\rho_i = -\vec{\nabla}\cdot\vec{u}(\vec{r}_i)\ . \tag{3.1}$$

The SP field $\phi(\vec{r}_i)$ is related to the displacement field $\vec{u}(\vec{r}_i)$ via the Poisson equation

$$\nabla^2\phi(\vec{r}_i) = -4\pi\delta\rho(\vec{r}_i) = 4\pi\rho_i\vec{\nabla}\cdot\vec{u}(\vec{r}_i)\ .$$

We shall illustrate the method for SP field quantization for the case of a vacuum-metal-metal system (Fig. 2). In Sec. II, we have seen that the field of SPO can be expressed as a superposition of eigen SP fields of wave vector $\vec{Q}$. Hence we can write $\phi(\vec{r})$ as

$$\phi = \begin{cases} \sum_{\vec{Q}} \phi_{1\vec{Q}}\, e^{-|Q|x}\, e^{i(\vec{Q}\cdot\vec{r}+\omega_{\vec{Q}}t)}, & x>d \text{ the vacuum region} \\ \sum_{\vec{Q}} (\phi_{2\vec{Q}}\, e^{|Q|x} + \phi'_{2\vec{Q}}\, e^{-|Q|x})\, e^{i(\vec{Q}\cdot\vec{r}+\omega_{\vec{Q}}t)}, & 0<x<d \\ \sum_{\vec{Q}} (\phi_{3\vec{Q}}\, e^{|Q|x}\, e^{i(\vec{Q}\cdot\vec{r}+\omega_{\vec{Q}}t)}), & 0>x\ . \end{cases} \tag{3.2}$$

The displacement field is decomposed in the same manner, i.e.,

$$\vec{u} = \begin{cases} 0, & x>d \\ \sum_{\vec{Q}} (\vec{u}_{2\vec{Q}}\, e^{|Q|x} + \vec{u}_{2\vec{Q}}\, e^{-|Q|x})\, e^{i(\vec{Q}\cdot\vec{r}+\omega_{\vec{Q}}t)}, & 0<x<d \\ \sum_{\vec{Q}} \vec{u}_{3\vec{Q}}\, e^{|Q|x}\, e^{i(\vec{Q}\cdot\vec{r}+\omega_{\vec{Q}}t)}, & 0>x\ . \end{cases} \tag{3.3}$$

The boundary conditions that must be imposed are the continuity of the SP wave field ϕ at the two interfaces. These together with (3.1), after integration across the interfaces, enable us to express the $\phi_{i\vec{Q}}$'s in terms of the $u_{i\vec{Q}}$'s. The kinetic energy part of L,

$$\sum_i \int (1/2n_i m_i)\vec{\pi}(r_i)\vec{\pi}(\vec{r}_i)\,d\vec{r}_i\ ,$$

is given by

$$\tfrac{1}{2}n_2m_2\int(\dot{\vec{u}}_2\cdot\dot{\vec{u}}_2)\,d\vec{r}_2 + \tfrac{1}{2}n_3m_3\int\dot{\vec{u}}_3\cdot\dot{\vec{u}}_3\,d\vec{r}_3 = \tfrac{1}{2}n_2m_2\sum_{\vec{Q}}(1/|Q|)$$
$$[(e^{2Qd}-1)\dot{u}_{2Qx}\dot{u}_{2-Qx} + (1-e^{-2Qd})\dot{u}'_{2Qx}\dot{u}'_{2-Qx}] + \tfrac{1}{2}n_3m_3\sum_{\vec{Q}}(1/|Q|)\dot{u}_{3Qx}\dot{u}_{3-Qx}\ . \tag{3.4}$$

In deriving this we have used the relation $\vec{\nabla}\cdot\vec{u}(\vec{r}_i) = 0$. For the evaluation of the potential energy contribution $\sum_i \int \delta\rho(\vec{r}_i)\phi(\vec{r}_i)\,d\vec{r}_i$ to L, we recognize that $\delta\rho$ is actually the sum of two δ functions whose strengths are determined by integrating (3.1) across the interfaces. Explicitly it is

$$\delta\rho = \delta(x-d)\sum_{\vec{Q}} n_2 e(u_{2Qx}\,e^{Qd} + u'_{2Qx}\,e^{-Qd})$$
$$+\,\delta(x)\sum_{\vec{Q}}[n_3 e\,u_{3Qx} - n_2 e(u_{2Qx}+u'_{2Qx})]\ . \tag{3.5}$$

After expressing ϕ in terms of u, we arrive at

$$\sum_i \int \delta\rho(\vec{r}_i)\phi(\vec{r}_i)\,d\vec{r}_i = [\pi(n_2e)^2/|Q|]\sum_{\vec{Q}}\{(u_{2Qx}\,e^{Qd} + u_{2Qx}\,e^{-Qd})$$
$$\times[u_{2-Qx}(e^{Qd}-e^{-Qd}) + (n_3/n_2)u_{3-Qx}\,e^{-Qd}] + [u'_{2-Qx}(e^{2Qd}-1) + (n_3/n_2)u_{3-Qx}]$$
$$\times[(n_3/n_2)u_{3Qx} - u_{2Qx} - u'_{2Qx}]\}\ . \tag{3.6}$$

Lagrange equations with respect to u_{2Qx}, u'_{2Qx}, and u_{3Qx} can be written down. For a fixed $\vec{Q}$, they are represented in matrix form as

$$\begin{bmatrix} \frac{1}{2}\omega_2^2 - \omega_{\vec{Q}}^2 & \frac{1}{2}\omega_2^2 e^{-2Qd} & 0 \\ \frac{1}{2}\omega_2^2 & \frac{1}{2}\omega_2^2 - \omega_{\vec{Q}}^2 & -(n_3/n_2)\frac{1}{2}\omega_2^2 \\ 0 & -(m_2/m_3)\frac{1}{2}\omega_2^2(1-e^{-2Qd}) & \frac{1}{2}\omega_3^2 - \omega_{\vec{Q}}^2 \end{bmatrix} \begin{bmatrix} u_{2Qx} \\ u'_{2Qx} \\ u_{3Qx} \end{bmatrix} = 0 . \tag{3.7}$$

$\omega_{\vec{Q}}$, the frequency for SPO of wave vector $\vec{Q}$, is an eigenvalue of the matrix. As can easily be verified, the dispersion relation of SPO thereby derived concurs with the result of Sec. II. The components u_{iQx} of the eigenvectors are linearly dependent. Hence we can express the Lagrangian L in terms of u_{2Qx} and its derivative $\dot{u}_{2Qx}$ only. For brevity let $u_Q = u_{2Qx}$, and we have finally,

$$L = \sum_{\vec{Q}} \frac{n_2 m_2 \mathcal{F}_Q}{2|Q|} \dot{u}_Q \dot{u}_{-Q} - \sum_{\vec{Q}} \frac{\pi(n_2 e)^2}{|Q|} \mathcal{G}_Q u_Q u_{-Q} . \tag{3.8}$$

When restricted to the low-frequency branch and assuming $\eta \equiv \omega_3/\omega_2$ is $\gg 1$,

$$\mathcal{F}_Q = 2(e^{2Qd}-1) + 2/\eta_2 + [e^{4Qd}(1-\beta_Q) + 3e^{2Qd}\beta_Q - 5e^{2Qd}]/2\eta^4 + e^{4Qd}(1-\beta_Q)/2\eta^6 , \tag{3.9a}$$

$$\mathcal{G}_Q = 2(e^{2Qd}-1) + (2-\beta_Q)(e^{2Qd}-1)/\eta^2 + [(4-\beta_Q) - e^{2Qd}(5-3\beta_Q) + e^{4Qd}(1-\beta_Q)]/2\eta^4 - [e^{2Qd}(3-2\beta_Q) + e^{4Qd}(1-\beta_Q)]/\eta^6 + e^{4Qd}(1-\beta_Q)/2\eta^8 , \tag{3.9b}$$

where

$$\beta_Q = [1 - 4e^{-2Qd}(1/\eta^2 - 1/\eta^4)]^{1/2} . \tag{3.10}$$

The momentum field conjugate to u_Q is

$$\pi_{\vec{Q}} = \frac{\partial L}{\partial \dot{u}_{\vec{Q}}} = \frac{n_2 m_2}{|Q|} \mathcal{F}_Q \dot{u}_{-\vec{Q}} . \tag{3.11}$$

Then the Hamiltonian is

$$H = \sum_{\vec{Q}} \pi_{\vec{Q}} \dot{u}_{\vec{Q}} - L = \frac{1}{2}\sum_{\vec{Q}} \frac{|Q|}{n_2 m_2 \mathcal{F}_Q} \pi_{\vec{Q}} \pi_{-\vec{Q}} + \sum_{\vec{Q}} \frac{\pi(n_2 e)^2}{|Q|} \mathcal{G}_Q u_{\vec{Q}} u_{-\vec{Q}} . \tag{3.12}$$

The classical SP field is now quantized by imposing the quantum conditions

$$[u_{\vec{Q}}, \pi_{\vec{Q}'}] = i\delta_{\vec{Q}\vec{Q}'} . \tag{3.13}$$

SP creation and annihilation operators are introduced as

$$c^\dagger_{\vec{Q}} = -i\left(\frac{2n_2 m_2 \mathcal{F}_Q \omega_Q}{|Q|}\right)^{-1/2} \pi_{\vec{Q}} + \left(\frac{\pi n_2^2 e^2 \mathcal{G}_Q \omega_Q}{|Q|}\right)^{1/2} u_{-\vec{Q}} , \tag{3.14a}$$

$$c_{\vec{Q}} = i\left(\frac{2n_2 m_2 \mathcal{F}_Q \omega_Q}{|Q|}\right)^{-1/2} \pi_{-\vec{Q}} + \left(\frac{\pi n_2^2 e^2 \mathcal{G}_Q \omega_Q}{|Q|}\right)^{1/2} u_{\vec{Q}} . \tag{3.14b}$$

We can verify easily that H in (3.12) is reduced to the harmonic oscillator form

$$H = \sum_{\vec{Q}} \omega_{\vec{Q}} (c^\dagger_{\vec{Q}} c_{\vec{Q}} + \tfrac{1}{2}) , \tag{3.15}$$

with $\omega_{\vec{Q}}$ defined as

$$\omega_{\vec{Q}} = \omega_2 (\mathcal{G}_Q/2\mathcal{F}_Q)^{1/2} . \tag{3.16}$$

Using (3.9a) and (3.9b) for $\mathcal{F}_Q$ and $\mathcal{G}_Q$, and after some lengthy algebra, we verify $\omega_{\vec{Q}}$ as defined in (3.16) is equivalent to the eigenfrequency $\omega_{\vec{Q}}$ of the matrix equation (3.7) for the lower branch as it should. The displacement operator $u_{\vec{Q}}$ is equal to

$$(2|Q|/\pi m_2 n_2 \mathcal{F}_Q)^{1/2}(c^\dagger_{\vec{Q}} + c_{\vec{Q}})$$

and the SP wave field is

$$\phi(\vec{r}) = \begin{cases} \sum_{\vec{Q}} \omega_{\vec{Q}} (2/|Q|\mathcal{F}_{\vec{Q}})^{1/2}[(e^{2Qd}-1) + 1/\eta^2 - e^{2Qd}(1-\beta_Q)/2\eta^4]\, e^{-|Q|x + i\vec{Q}\cdot\vec{r}}(c^\dagger_{-\vec{Q}} + c_{\vec{Q}}) , & x > d \\ \sum_{\vec{Q}} \omega_{\vec{Q}} (2/|Q|\mathcal{F}_Q)^{1/2}\{[1 + (1-\beta_Q)/2\eta^2]e^{Qx} - [1 - 1/\eta^2 + e^{2Qd}(1-\beta_Q)/2\eta^2 + e^{2Qd}(1-\beta_Q)/2\eta^4]e^{-Qx}\} \\ \qquad \times e^{i\vec{Q}\cdot\vec{r}}(c^\dagger_{\vec{Q}} + c_{\vec{Q}}) , & d > x > 0 \\ \sum_{\vec{Q}} \omega_{\vec{Q}} (2/|Q|\mathcal{F}_Q)^{1/2}[1/\eta^2 + (1-e^{2Qd})(1-\beta_Q)/2\eta^2 - e^{2Qd}(1-\beta_Q)/2\eta^4]e^{Qx + i\vec{Q}\cdot\vec{r}}(c^\dagger_{-\vec{Q}} + c_{\vec{Q}}), & x < 0 \end{cases} \tag{3.17}$$

and the el-SP interaction is $H_{\text{el-SP}} = e\phi(\vec{r})$.

As our next example we consider an idealized semiconductor-metal junction as consisting of semiconductor S and metal M both semi-infinite

and separated by a dielectric medium of dielectric constant ϵ_∞. The Lagrangian of the system is taken as the sum of contributions from the S and M media. Proceeding in exactly the same manner as in the previous case, we can obtain the creation and annihilation operators $c^\dagger_{\vec{Q}}$ and $c_{\vec{Q}}$ for SP in the M-S contact. The interaction Hamiltonian density of an electron in the dielectric region $0<x<d$ with the SP field can be derived using techniques similar to those leading to (3.17). It is

$$H_{\text{el-SP}} = e\sum_{\vec{Q}} \phi_{\vec{Q}}(c^\dagger_{\vec{Q}} + c_{\vec{Q}})\, e^{i\vec{Q}\cdot\vec{r}} \,. \tag{3.18}$$

Here

$$\phi_{\vec{Q}} = \left(\frac{\pi\omega_Q}{Q}\right)^{1/2}\left[\left(1+\frac{\epsilon_Q}{\epsilon_\infty}\right)e^{-Qx} + \left(1-\frac{\epsilon_Q}{\epsilon_\infty}\right)e^{-2Qd+Qx}\right]$$
$$\times\left\{\left[(\epsilon_\infty+1)\left(1+\frac{\epsilon_Q}{\epsilon_\infty}\right)+(1-\epsilon_\infty)\left(1-\frac{\epsilon_Q}{\epsilon_\infty}\right)e^{-2Qd}\right]\right.$$
$$\times\left[\left(1+\frac{\epsilon_Q}{\epsilon_\infty}\right)+e^{-2Qd}\left(1-\frac{\epsilon_Q}{\epsilon_\infty}\right)\right.$$
$$\left.\left.+e^{-2Qd}\left(1+\frac{\epsilon_Q}{\epsilon_\infty}\right)\right]+(\epsilon_\infty-\epsilon_Q)e^{-2Qd}\right\}^{-1/2} \tag{3.19}$$

and

$$\epsilon_Q = \epsilon_\infty[1-(\omega_S^2/\omega_Q^2)] \,. \tag{3.20}$$

IV. INELASTIC el-SP INTERACTIONS IN METAL-SEMICONDUCTOR TUNNEL JUNCTIONS

In this section we present a calculation of the inelastic current due to el-SP interaction within the scheme of the tunneling Hamiltonian formalism. An excellent review of the formalism is given by Duke.[39] We consider the junction system to be described by the Hamiltonian $H = H_L + H_R + H_b$. H_R and H_L are the complete Hamiltonian for the isolated metal M and semiconductor S electrodes, and

$$H_b = \sum_{\vec{Q}} \hbar\omega_{\vec{Q}}(c^\dagger_{\vec{Q}}\, c_{\vec{Q}} + \tfrac{1}{2}) + \sum_{\vec{k}\,\vec{q}\alpha}(\Lambda^{(0)}_{\vec{k},\vec{q}}\, a^\dagger_{\vec{k}\alpha}a_{\vec{q}\alpha} + \text{H. c.})$$
$$+\sum_{\vec{Q}}\sum_{\vec{k}\,\vec{q}\alpha}\Lambda^{(1)}_{\vec{k},\vec{q}}(Q)[c_{\vec{Q}}(t)+c^\dagger_{\vec{Q}}(t)]$$
$$\times[a^\dagger_{\vec{k}\alpha}(t)a_{\vec{q}\alpha}(t)+\text{H. c.}] \,. \tag{4.1}$$

$a^\dagger_{\vec{k}\alpha}$, $a_{\vec{k}\alpha}$ ($a^\dagger_{\vec{q}\alpha}$, $a_{\vec{q}\alpha}$) are the creation and annhilation operators of electrons in the S (M) electrode, respectively. The first term in H_b is the Hamiltonian of the SP excitation of the junction. The second term describes the background elastic conductance. The last term represents the el-SP interaction. In this form we have excluded from consideration the multi-SP processes and the SP-induced elastic (zero SP) tunneling current. Evaluation of the tunneling current is done by calculating the linear response of the unperturbed system described by $H_L + H_R$ to the tunneling Hamiltonian H_b.[24]

The current, when the bias is V, can be expressed as a functional of temperature Green's functions and consists of an elastic part,

$$J_e(V,T) = 4e\sum_{\vec{k}\,\vec{q}}|\Lambda^{(0)}_{\vec{k}\,\vec{q}}|^2\int_{-\infty}^{\infty}\frac{d\omega}{\pi}[f(\omega)-f(\omega+eV)]$$
$$\times\operatorname{Im}G^R(\vec{k},\omega)\operatorname{Im}G^R(\vec{q},\omega+eV) \,, \tag{4.2}$$

and an inelastic part due to SP excitation,

$$J_i(V,T) = -4e\sum_{\vec{k}\,\vec{q}}\int\frac{d\omega_1\,d\omega_2}{\pi^2}[f(\omega_1)-f(\omega_1-eV)]$$
$$\times[f(\omega_2)+N(\omega_2-\omega_1+eV)]\operatorname{Im}G^R(\vec{k},\omega_2)$$
$$\times\operatorname{Im}G^R(\vec{q},\omega_1)\operatorname{Im}D^R_{\vec{k}\,\vec{q}}(\omega_2-\omega_1+eV) \,. \tag{4.3}$$

$f(\omega)$ [$N(\omega)$] is the Fermi [Bose] function, and $G^R(\vec{q},\omega)$ [$G^R(\vec{k},\omega)$] is the retarded single-electron Green's function of the M [S] electrode. The relations

$$D^R_{\vec{k},\vec{q}}(\omega) = \lim_{\delta\to 0^+}\mathfrak{D}_{\vec{k},\vec{q}}\ (i\omega_n = \omega+i\delta) \,, \tag{4.4}$$

$$\mathfrak{D}_{\vec{k},\vec{q}}(i\omega_n) = -\sum_{\vec{Q}}\int_0^{1/k_BT}\langle T_\tau[c_{\vec{Q}}(\tau)+c^\dagger_{\vec{Q}}(\tau)]$$
$$\times[c_{\vec{Q}}(0)+c^\dagger_{\vec{Q}}(0)]\rangle|\Lambda^{(1)}_{\vec{k}\,\vec{q}}(\vec{Q})|^2 e^{i\omega_n\tau}d\tau \,, \tag{4.5}$$

$$\omega_n = 2\pi nk_BT \tag{4.6}$$

can be related to the SP propagator

$$D_{\vec{Q}}(i\omega_n) = \int_0^{1/k_BT}\langle T_\tau[c_{\vec{Q}}(\tau)+c^\dagger_{\vec{Q}}(\tau)]$$
$$\times[c_{\vec{Q}}(0)+c^\dagger_{\vec{Q}}(0)]\rangle e^{i\omega_n\tau}d\tau \tag{4.7}$$

via

$$D_{\vec{k}\,\vec{q}}(i\omega_n) = -\sum_{\vec{Q}}|\Lambda^{(1)}_{\vec{k}\,\vec{q}}(Q)|^2 D_{\vec{Q}}(i\omega_n) \,. \tag{4.8}$$

The $\Lambda^{(1)}_{\vec{k}\,\vec{q}}(\vec{Q})$ that first appeared in (4.1) is defined by its relation to the SP field as

$$\sum_{\vec{Q}}\Lambda^{(1)}_{\vec{k}\,\vec{q}}(\vec{Q})[c_{\vec{Q}}(t)+c^\dagger_{\vec{Q}}(t)] = \int d^3\vec{r}\, e\phi(\vec{r},t)\psi^*_{\vec{k}}(\vec{r})\psi_{\vec{q}}(\vec{r}) \,, \tag{4.9}$$

where

$$\psi_{\vec{k}}(\vec{r}) = \frac{e^{i\vec{k}_\parallel\cdot\vec{r}}}{A^{1/2}}\chi_k(x) \quad\text{and}\quad \psi_{\vec{q}}(\vec{r}) = \frac{e^{i\vec{q}_\parallel\cdot\vec{r}}}{A^{1/2}}\chi_q(x)$$

are the one-electron basis states for the two electrodes. The boundary condition on χ is that the wave function be purely attenuating in the barrier for electrons with energy less than the barrier potential.[39] Hence, from (3.18), we have

$$\Lambda^{(1)}_{\vec{k}\vec{q}}(\vec{Q}) = [1/(L_L L_R)^{1/2} A] M(\vec{Q}; k_x, q_x)\delta(\vec{k}_\parallel - \vec{q}_\parallel - \vec{Q}) , \tag{4.10}$$

$$M(Q; k_x, q_x) = \int e\phi_{\vec{Q}}(x)\chi_{\vec{k}}(x)\chi_{\vec{q}}(x)\,dx . \tag{4.11}$$

At present we are interested in $|eV| \approx \omega_s > \omega_0$ (the LO phonon energy of S). We shall therefore replace

$$\mathrm{Im}G^R(k, \omega_2) = \pi\delta(\xi_{\vec{k}} - \omega_2)$$

and

$$\mathrm{Im}G^R(\vec{q}, \omega_1) = \pi\delta(\xi_{\vec{q}} - \omega_1) .$$

We have $\xi_{\vec{k}} = (\hbar^2\vec{k}^2/2m_L) - \zeta_L$ and $\xi_{\vec{q}} = (\hbar^2\vec{q}^2/2m_R) - \zeta_R$, where $\zeta_L(m_L)$ and $\zeta_R(m_R)$ denote the Fermi degeneracies (effective masses) in the left- and right-hand electrodes, respectively. The inelastic current in (4.3) becomes

$$J_i(V, T) = 4\pi e \sum_{\vec{k}\vec{q}\vec{Q}} |\Lambda^{(1)}_{\vec{k}\vec{q}}(\vec{Q})|^2 S(\xi_{\vec{k}}, \xi_{\vec{q}}, \hbar\omega_{\vec{Q}}, eV) , \tag{4.12}$$

where

$$S(\xi_{\vec{k}}, \xi_{\vec{q}}, \hbar\omega_{\vec{Q}}, eV) = \{[f(\xi_{\vec{k}}) + N(\omega_{\vec{Q}})]\delta(\xi_{\vec{q}} - \xi_{\vec{k}} - eV + \omega_{\vec{Q}}) + [f(-\xi_{\vec{k}}) + N(\omega_{\vec{Q}})]\delta(\xi_{\vec{q}} - \xi_{\vec{k}} - eV - \omega_{\vec{Q}})\} \times [f(\xi_{\vec{q}}) - f(\xi_{\vec{q}} - eV)] . \tag{4.13}$$

When summed over $\vec{q}$, the last factor in (4.13) becomes $f(\xi_{\vec{k}} + eV - \omega_{\vec{Q}}) - f(\xi_{\vec{k}} - \omega_{\vec{Q}})$ which leads to the conductance being a step function $\theta(eV - \omega_{\vec{Q}})$ if we neglect the lifetime of SP. The major difference between SP and phonons (such as TA) should be pointed out here. Despite our having dispersion of the SP frequencies, only the SP modes with $\omega_Q \approx \omega_S$ contribute significantly to the inelastic current. The reason lies in the fact that Q-phase space for SP modes with $\omega_Q < \omega_S/\sqrt{2}$ is negligible compared with that for which $\omega_Q \approx \omega_S/\sqrt{2}$. Nevertheless, SP excitation processes are observed to have broad line shapes (in contrast to a δ function). The major cause for this is the strongly Q-dependent Landau damping (LD) for the SP modes, as we shall see. For the case $\omega_S \approx \omega_0$, we not only have el-LO phonon self-energy modification of the spectral weight function in the S electrode but also the possibility of SP-LO phonon coupling analogous to plasmon-LO phonon coupling.[32,33]

The calculation of the SP-induced inelastic current can now proceed by first evaluating $\Lambda^{(1)}_{\vec{k}\vec{q}}(\vec{Q})$ of (4.10) with $\chi_{\vec{k}}$ and $\chi_{\vec{q}}$ the basis states appropriate to the barrier potential in a S-M junction, which is a rather formidable job. It is important to keep our objectives in mind. What we want is to obtain the order of magnitude of the theoretical size of the SP effect and its line shape in d^2I/dV^2. We shall therefore adopt a square-barrier model to describe the one-electron aspects of the tunneling current. This model was also adopted by Bennett *et al.*[24] (BDS) when calculating the impurity-induced elastic and inelastic currents. To save space we shall adopt their notations. In the same approximation as used by BDS, $M(\vec{Q}, k_x, q_x)$ in (4.11) is obtained as

$$|M(\vec{Q}, k_x, q_x)|^2 = (2m_b^2 k_x q_x e^2/m_L m_R \kappa_R \kappa_L)\, e^{-\kappa_T d} |\Theta_{\vec{Q}}|^2 , \tag{4.14}$$

$$\Theta_{\vec{Q}} = \int_0^d \phi_{\vec{Q}}(x)\,dx . \tag{4.15}$$

Then

$$J_i(V, T) = 4\pi e \sum_{\vec{Q}} \int \frac{d^3k}{(2\pi)^3} \int \frac{d^3q}{(2\pi)^3} |M(\vec{Q}, k_x, q_x)|^2 \times (2\pi)^2\delta(\vec{k}_\parallel - \vec{q}_\parallel - \vec{Q}) S(\xi_{\vec{k}}, \xi_{\vec{q}}, \omega_{\vec{Q}}, eV) . \tag{4.16}$$

This equation is identical in form to the expression for the coherent one-phonon inelastic current given by BDS. The integration over d^3k and d^3q can be done as described there, and we obtain

$$J_i(V, T) = D\, e^{eV/E_b} \sum_{\vec{Q}} |\Theta_{\vec{Q}}|^2 e^{-Q^2/Q_b^2} \int_{\zeta_L}^{\infty} d\xi_k \times e^{2\xi_k/E_b}\{1 - \exp[-(1 + m_L/m_R)(\xi_L + \xi_k)/E_b]\}\, W(\xi_k, \omega_{\vec{Q}}, eV) , \tag{4.17}$$

$$W(\xi_{\vec{k}}, \hbar\omega_{\vec{Q}}, eV) = [f(\xi_{\vec{k}} + eV - \omega_{\vec{Q}}) - f(\xi_{\vec{k}} - \omega_{\vec{Q}})][f(\xi_{\vec{k}}) + N(\omega_{\vec{Q}})]\, e^{-\omega_Q/E_b} + [f(\xi_{\vec{k}} + eV + \omega_{\vec{Q}}) - f(\xi_{\vec{k}} + \omega_{\vec{Q}})][f(-\xi_{\vec{k}}) + N(\omega_{\vec{Q}})]\, e^{\omega_{\vec{Q}}/E_b} , \tag{4.18}$$

$$D = \frac{2m_b^2 e^3 A}{(2\pi)^2 \kappa_{b\mu}^2}\, e^{-2\kappa_{b\mu} d}\, \frac{Q_b^2}{(1 + m_R/m_L)} . \tag{4.19}$$

E_b, $\kappa_{b\mu}$, and d are parameters of an average-square-barrier model.[39] The integration over $d\xi_k$ is exactly of the same form as in BDS [Eq. (6.10) of Ref. 24]. We readily take over their results and find for eV near ω_Q, such that $(eV - \omega_{\vec{Q}})/E_b \ll 1$ and $(Q/Q_b)^2 \ll 1$ and at zero temperature,

$$J_i(V, 0) = D \sum_{\vec{Q}} |\Theta_{\vec{Q}}|^2 \left[1 + \exp\left(\frac{-(1 + m_L/m_R)\zeta_L}{E_b}\right)\right] (eV - \omega_{\vec{Q}})\theta(eV - \omega_{\vec{Q}}) , \tag{4.20}$$

$$\frac{dJ_i}{d(eV)} = D\sum_{\vec{Q}} |\Theta_{\vec{Q}}|^2 \left[1+\exp\left(\frac{-(1+m_L/m_R)\xi_1}{E_b}\right)\right]\theta(eV-\omega_{\vec{Q}}) , \tag{4.21}$$

and

$$\frac{d^2J_i}{d(eV)^2} = D\sum_{\vec{Q}} |\Theta_{\vec{Q}}|^2 \left[1+\exp\left(\frac{-(1+m_L/m_R)\xi_1}{E_b}\right)\right]\delta(eV-\omega_{\vec{Q}}) . \tag{4.22}$$

The assumption $(Q/Q_b)^2 \ll 1$ can be justified; we shall see later that it is valid for all Q of importance. Before we proceed to study the SP-induced line shape from (4.22), it is worthwhile to derive another form for $d^2J_i/d(eV)^2$ which resembles the results of Scalapino and Marcus.[40] This form is only rigorously correct when the background conductance is independent of bias. We make the drastic approximation of replacing $e^{-\kappa_T d}$ in (4.14) by $e^{-\kappa_b d}$ as in elastic tunneling. This implies

$$\Lambda^{(1)}_{\vec{k}\vec{q}}(\vec{Q}) = e(2m_b/\Phi)^{1/2}\delta(\vec{k}_{\parallel}-\vec{q}_{\parallel}-\vec{Q})\Lambda^{(0)}_{\vec{k}\vec{q}}\Theta_{\vec{Q}} . \tag{4.23}$$

Φ is the effective barrier potential at $eV=0$ for the square-barrier model.[39] The one-SP inelastic current is

$$J_i(V,0) = -(4m_b e^3/\Phi)\sum_{\vec{k}\vec{q}} |\Lambda^{(0)}_{\vec{k}\vec{q}}|^2 \int dx\,dx'\ [f(x)-f(x-eV)]$$

$$\times f(x')\,\mathrm{Im}G^R(\vec{k},x')\delta(x-\xi_{\vec{q}})\textstyle\sum_{\vec{Q}}|\Theta_{\vec{Q}}|^2\delta(x'-x+eV+\omega_{\vec{Q}})\delta(\vec{k}_{\parallel}-\vec{q}_{\parallel}-\vec{Q}) . \tag{4.24}$$

Using

$$|\Lambda^{(0)}_{\vec{k}\vec{q}}|^2 = \left(\frac{\partial\xi_k}{\partial k_x}\right)\left(\frac{\partial\xi_q}{\partial q_x}\right)D(\vec{k}_{\parallel},\xi_{\vec{k}}) \tag{4.25}$$

and noticing that

$$G_e(V) \equiv \frac{dJ_e(V,0)}{d(eV)} = -\frac{2e}{\pi}\int d\xi_{\vec{k}}\,\mathrm{Im}G^R(\xi_{\vec{k}},-eV)\int\frac{d^2k_{\parallel}}{(2\pi)^2}D(\vec{k}_{\parallel},\xi_{\vec{k}}) , \tag{4.26}$$

we find

$$J_i(V,0) = \frac{2m_b e^2}{\Phi}\sum_{\vec{Q}}\int dx' f(x')[f(x'+eV-\omega_{\vec{Q}})-f(x'-\omega_{\vec{Q}})]G_e(-x')|\Theta_{\vec{Q}}|^2 , \tag{4.27}$$

$$\frac{dJ_i(V,0)}{d(eV)} = \frac{2m_b e^2}{\Phi}\sum_{\vec{Q}} f(\omega_{\vec{Q}}-eV)G_e(eV-\omega_{\vec{Q}})|\Theta_{\vec{Q}}|^2 , \tag{4.28}$$

$$\frac{d^2J_i(V,0)}{d(eV)^2} = \frac{2m_b e^2}{\Phi}\sum_{\vec{Q}}|\Theta_{\vec{Q}}|^2\left(G_e(eV-\omega_{\vec{Q}})\,\delta(eV-\omega_{\vec{Q}}) - \frac{dG_e(eV-\omega_{\vec{Q}})}{d(eV)}f(\omega_{\vec{Q}}-eV)\right) . \tag{4.29}$$

Both G_e and $dG_e/d(eV)$ are obtainable from experiment, and we can show that the second term within the braces of (4.29) is two orders of magnitude smaller than the first and shall be dropped. Then this simplified form of (4.29) is directly related to (4.22) as can be seen in the special case of a metallic junction with constant conductance. When $|eV| \ll \frac{1}{2}E_b$, then

$$G_e(V) = 2\pi m_L A_1 E_b\, e^{-2\kappa_{b\mu}d} ,$$

and we can easily verify that D in (4.22) is just $(2m_b e^2/\Phi)G_e(V)$ and the connection of (4.22) with (4.29) follows.

Next we evaluate $|\Theta_{\vec{Q}}|^2$. From (3.19), (4.15), and (4.22) we find

$$|\Theta_{\vec{Q}}|^2 = \frac{\pi\omega_{\vec{Q}}}{2Q^3}\frac{1-e^{-Qd}}{1+e^{-Qd}} , \tag{4.30}$$

$$\frac{d^2J_i}{d(eV)^2} = \frac{\pi D}{2}\sum_{Q}\left(\frac{\omega_{\vec{Q}}}{Q^3}\right)\left(\frac{1-e^{-Qd}}{1+e^{-Qd}}\right)\delta(eV-\omega_{\vec{Q}}) . \tag{4.31}$$

The summation over Q is changed to integration over ω, where $\omega=\omega_Q$ with Jacobian $\omega/(\omega_S^2-\omega^2)d$, then

$$\frac{d^2J_i}{d(eV)^2} = Dd\int^{\omega_S/\sqrt{2}} d\omega\left(\frac{1-(1-2\omega^2/\omega_S^2)^{1/2}}{1+(1-2\omega^2/\omega_S^2)^{1/2}}\right)$$

$$\times\left(\frac{2\omega^2}{\omega_S^2-2\omega^2}\right)\frac{1}{ln^2(1-2\omega^2/\omega_S^2)}\,\delta(eV-\omega) . \tag{4.32}$$

The integrand has a singularity of the form

$$\{[(\omega_S/\sqrt{2})-\omega]\,ln^2\,[(\omega_S/\sqrt{2})-\omega]\}^{-1}\ .$$

We shall see later that convolution of this singularity with a Lorentzian function determines the position of the peak and its line shape.

As we have discussed in Sec. II, real SP have finite lifetime caused by Landau damping. To include this lifetime effect, we replace the δ function in (4.32) by a Lorentzian[41]

$$\omega_2(Q)/\pi\{[\omega_1(Q)-eV]^2+\omega_2^2(Q)\}\ ,$$

where $\omega_1(Q)=\mathrm{Re}\omega_Q$, $\omega_2(Q)=\mathrm{Im}\omega_Q+\frac{1}{2}\Delta\omega_\tau$, and $\Delta\omega_\tau$ represents the collision broadening of electrons. LD is strongly Q dependent and contributes a term $\alpha Q/Q_c$ to $\mathrm{Im}\omega_Q$. Since SP of higher Q are strongly attenuated, we cut off the integration over Q in (4.31) at Q_{c2}, where $\mathrm{Im}\omega_{Q_{c2}}\approx\alpha Q_{c2}/Q_c$ is a sizable fraction of $\mathrm{Re}\omega_{Q_{c2}}$ so that the SP is no longer a well-defined mode of excitation. Since SP do not exist for wavelengths shorter than the interelectronic distance, we expect $Q_{c2}<k_F$. Further we take $\Delta\omega_\tau$ as calculated from the measured value of the electron mobility in the S samples. Hence we have, within the present model of SP excitation, a *one*-adjustable-parameter theory for the line shape, namely, α (or Q_{c2} which is related to α).

Sample line shapes for several values of Q_{c2} have been given in Fig. 2 of a previous brief report.[17] It is observed therein that the line shape depends very sensitively on the choice of Q_{c2}. The best fit to experiment corresponds to $Q_{c2}/Q_c\approx 2$ for all samples.[17] This is no coincidence but a natural consequence of our SP-induced inelastic tunneling current model. For, in the theory of LD, the parameter $Q_c\equiv\omega_S/v_F$ has a physical meaning. From the definition of Q_{c2}, the ratio Q_{c2}/Q_c is expected to be invariant with respect to different samples. Quantitative comparison of the value of α, obtained from the best *one*-parameter fit, with predictions of model theories of SP shall now be made. For this purpose we consider the recent work of Feibelman[30] who has computed the LD of SP. His result predicts

$$\mathrm{Im}\omega_Q=(\pi/2^{15/4})(Q/Q_S)(\omega_S/\sqrt{2})\ , \qquad (4.33)$$

where $Q_S=(2m\omega_S/\hbar)^{1/2}$.

This is 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 Tsui's data[15] both Q_c and Q_S can be calculated. We find $\alpha_F/\omega_S\approx 0.03$ which is in excellent agreement with the value of $\alpha/\omega_S\approx 0.035$ that gives the best fit to the data for all samples.[17]

We have thus seen from Fig. 2 of Ref. 17 that the present theory leads to a line shape in agreement with experiment. Moreover, it explains quite satisfactorily all the puzzling features as noted by Tsui: First, the long tail at low biases is due to the fact that the dispersion for SP is given by $(\omega_S/\sqrt{2})(1-e^{-2Qd})^{1/2}$ instead of $\omega_S/\sqrt{2}$ for one interface. Second, the maximum does not occur at $\omega_S/\sqrt{2}$ as one may first expect,[15] but at a lower value (see Table I of Ref. 15). This is due to the combined effect of dispersion and considerable LD at Q near Q_{c2}. We have thus removed the discrepancy between the value of the observed "threshold" and $\omega_S/\sqrt{2}$.[15] The way LD pushes the maximum to a lower value than $\omega_S/\sqrt{2}$ can be seen from (4.32) when $\delta(eV-\omega)$ is replaced by the Lorentzian as described. The pole singularity

$$\{[(\omega_S/\sqrt{2})-\omega]\,ln^2\,[(\omega_S/\sqrt{2})-\omega]\}^{-1}$$

at $\omega=\omega_S/\sqrt{2}$, when convoluted with the Lorentzian, will shift the maximum to a lower value. Third, the broad line shape is due mainly to considerable LD near Q_{c2} and to a much less extent due to SP dispersion or collision broadening. Tsui's observation of "no systematic dependence of this half-width on the electron mobility of the sample" can now be explained. Tsui has observed a strong dependence of the half-width on the electron concentration but not on electron mobility. Our present theory indicates that the half-width is determined roughly by the sum of collision broadening and LD at Q near Q_{c2} (indeed at such high values of Q, LD dominates over collision broadening). Referring to (4.33) it is clear that the magnitude of LD depends linearly on ω_S or (concentration $n)^{1/2}$. The observed strong dependence of half-width on n is in accordance with the theory. To further demonstrate this point we have plotted in Fig. 3 the measured half-widths from Tsui's data versus $n^{1/2}$. The linear dependence is clear.[42] Fourth, the reason for the absence of inelastic volume plasmon excitation by tunneling electrons is that the field of the bulk plasmon, unlike that of SP, is essentially zero in the barrier region.

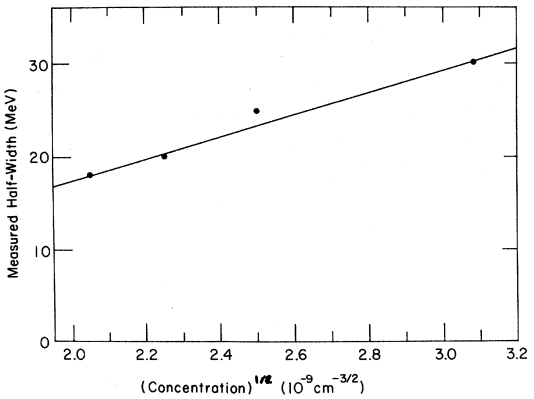

FIG. 3. Plot of the measured half-widths (from Tsiu's data) versus the square root of the electron concentration.

The magnitude of $d^2J_i/d(eV)^2$ predicted by the theory and given by either (4.22) or (4.29) can be calculated by taking $d \approx 100$ Å, $\Phi \approx \frac{1}{2} eV$, and G_e as deduced from the experiment. The calculated value of the change in the second derivative of the tunneling current, due to SP emission, is consistent with the experimentally reported size of this change.

V. SP EXCITATION IN LOW-ENERGY-ELECTRON DIFFRACTION AND IN PHOTOEMISSION

In this section we provide a theory for calculating el-SP inelastic scattering in LEED. In Sec. III we derived the el-SP interaction Hamiltonian. Although it takes several forms in different media, we can express it as

$$e\phi(\vec{r}) = e\sum_{\vec{Q}} [\pi\omega_Q/2Q\mathfrak{D}(Q)]^{1/2}\phi_Q(x)\, e^{i\vec{Q}\cdot\vec{r}}(c^\dagger_{\vec{Q}} + c_{\vec{Q}}) \ . \tag{5.1}$$

$$\phi_{\vec{Q}}(x) = A_1(Q)\, e^{-Qx} \ , \qquad x > d \qquad \text{(vacuum region)}$$

$$= A_2(Q)\, e^{Qx} + A_2'(Q)\, e^{-Qx} \ , \qquad d > x > 0 \qquad \text{(Cs region)}$$

$$= A_3(Q)\, e^{Qx} \ , \qquad x < 0 \qquad \text{(W region)} \ .$$

When compared with (3.17), $\mathfrak{D}(Q) = (4/\pi)\mathfrak{F}(Q)$ and $\phi_{\vec{Q}}(x)$ are just the expressions within the braces. Since $\eta^2 \ll 1$, we can expand these quantities in powers of η^2. To lowest order, we have $A_1 \approx (e^{2Qd} - e^{-2Qd})$, $A_2 \approx (1 + e^{-2Qd})$, $A_2' \approx -(1 + e^{-2Qd})$, $A_3 \approx (1 - e^{-4Qd})\eta^2$, and $\mathfrak{D} \approx (e^{2Qd} + 1 - e^{-2Qd} - e^{-4Qd})$. Note that the el-SP interaction in the W region is smaller by a factor of η^2.

Our problem is the scattering of electrons under the combined influence of $U(\vec{r})$—the crystal potential, and $V(\vec{r}) = e\phi(\vec{r})$—the SP wave field. This falls into the general category of two potentials scattering.[26,43,44]

For a calculation of the SP-emission transition probability, it is possible to circumvent the difficulty of acquiring the complicated LEED wave functions by taking $U = U_0(x)$ and thus ignoring diffraction. Naturally, this model can give an order-of-magnitude estimate to the SP scattering intensity only. $\chi_f^-(\chi_i^+)$, the eigenfunctions of U_0 for ingoing (outgoing) wave condition, will now be a product of an SP wave function $\Phi_{n_f}(\Phi_{n_i})$, a plane wave $e^{i\vec{K}_f\cdot\vec{r}}(e^{i\vec{K}_i\cdot\vec{r}})$ for electron motion parallel to the interface, and a normal wave function $\xi^{\pm}_{k_f}(x)$. $\xi^{\pm}_{k_f}(x)$, for example, is the solution to the one-dimensional Schrödinger equation

$$[-(\hbar^2/2m)(d^2/dx^2) + U_0(x) - \mathcal{E}]\xi(x) = 0 \ ,$$

with $\mathcal{E} = \hbar^2 k_f^2/2m$. The state χ_f^- labeled by $\{k_{fx}, \vec{K}_f, n_f\}$ has total energy

$$E_f = \hbar^2(k_{fx}^2 + K_f^2)/2m + \sum_{\vec{Q}} \hbar\omega_{\vec{Q}}\,(n_{f\vec{Q}} + \tfrac{1}{2}) \ .$$

$n_{f\vec{Q}}$ is the number of SP of wave vector $\vec{Q}$, and $\sum_{\vec{Q}} n_{f\vec{Q}} = n_f$. Let us first consider the intensity of electrons scattered via 1-SP emission. Such an intensity R is defined as the ratio (flux of electrons that have undergone 1-SP emission)/(incident flux), and to first order in V, it is given as

$$R = (2\pi m/\hbar^2|\, k_{ix}|)\sum_f |\langle \chi_f^-|\, V\,|\chi_i^+\rangle|^2 \delta(E_f - E_i) \ . \tag{5.2}$$

Since V is just $e\phi(\vec{r})$ in (5.1), the momentum conservation $\vec{K}_f + \vec{Q} = \vec{K}_i$ must be obeyed for a nonvanishing matrix element. Summation over f in (5.2) implies summation over $\vec{Q}'$, $\vec{K}_f$, and k_{fx}. Replacing the summation over k_{fx} by an integral over $\mathcal{E}_{fx}(\equiv \hbar^2 k_{fx}^2/2m)$ with the Jacobian $dN_x/d\mathcal{E}_{fx}$, which is the density of states for motion perpendicular to the interfaces, (5.2) becomes

$$R = \left(\frac{m}{\hbar^2}\right)^2 \sum_{\vec{Q}'}\sum_{\vec{K}_f} \int \frac{d\mathcal{E}_{fx}}{|k_{ix}k_{fx}|} \times |\langle e^{i\vec{K}_f\cdot\vec{r}}\,\xi^-_{k_{fx}}(x)\Phi_1(\vec{Q}')|\, V\,| \times e^{i\vec{K}_i\cdot\vec{r}}\,\xi^+_{k_{ix}}(x)\Phi_0\rangle|^2 \delta(E_f - E_i) \ . \tag{5.3}$$

Here $\Phi_1(\vec{Q}')$ is the SP state function with one SP of wave vector $\vec{Q}'$ present, and Φ_0 is the SP ground state. To calculate the intensity $R^{(2)}$ of electrons scattered via 2-SP emission defined similarly to R, we consider the second term of the Born series for the T matrix. This term can be written as $\sum_g V_{fg}V_{gi}(E_i - E_g + i\epsilon)^{-1}$, where states designated by g are labeled by $\{k_{gx}, \vec{K}_g, n_g\}$. Summing over k_{gx} reduces it to

$$\sum_{\vec{K}_g, n_g} \frac{(-im/2\hbar^2)V_{fg}V_{gi}}{\hat{k}_{xg}} \ ,$$

where $\hat{k}_{gx}$ is determined by the equation $E_g - E_i = 0$. Then the contribution to the reflectivity via emission of two SP's of wave vector $\vec{Q}_1$ and $\vec{Q}_2$ to final states $f \equiv \{k_{fx}, \vec{K}_f, \vec{Q}_1 + \vec{Q}_2\}$ is

$$R^{(2)} = \left(\frac{m}{2\hbar^2}\right)^4 \sum_{\vec{Q}_1\vec{Q}_2 K_f} \int \frac{d\epsilon_{fx}}{|k_{ix}k_{fx}|} \times \left| \sum_{\vec{K}_g, n_g} \frac{V_{fg}V_{gi}}{|\hat{k}_{gx}|} \right|^2 \delta(E_f - E_i) \ . \tag{5.4}$$

The matrix elements guarantee momentum conservation: $\vec{K}_f + \vec{Q}_1 + \vec{Q}_2 = \vec{K}_i$. The matrix element in (5.3) introduces a factor $\delta(\vec{Q}, \vec{Q}')\delta(\vec{K}_f, \vec{K}_i + \vec{Q})$ and reduces R to

$$R = \frac{\pi}{2}\left(\frac{me}{\hbar^2}\right)^2 \sum_{\vec{Q}} \frac{\hbar\omega_Q}{\mathfrak{D}(Q)} \int \frac{d\mathcal{E}_{fx}}{|k_{ix}k_{fx}|} \, |\langle \xi^-_{k_{fx}}(x)|\, \phi_{\vec{Q}}(x)| \times \xi^+_{k_{ix}}(x)\rangle|^2 \delta(\mathcal{E}_i - \mathcal{E}_f - \hbar\omega_{\vec{Q}}) \ . \tag{5.5}$$

We change integration variables from $\vec{Q}$, $\mathcal{E}_{fx}$ to $\vec{Q}$, $\mathcal{E}_f$ and define $dR/d\mathcal{E}_f$ via $R = \int (dR/d\mathcal{E}_f)d\mathcal{E}_f$. Then

$dR/d\mathcal{E}_f$ is the quantity to be compared directly with the LEED inelastic spectrum. We have seen in previous sections that SP excitation is strongly attenuated by Landau damping, which contributes an imaginary part to its energy according to (2.18). To include various inelastic processes other than SP emission that an electron can undergo,[45] the initial- and final-state electron energies should have imaginary parts $\Gamma(\mathcal{E}_i)$ and $\Gamma(\mathcal{E}_f)$, respectively. The energy-conserving δ function in (5.4) should then be replaced by a Lorentzian with a width $\Gamma = \mathrm{Im}\omega_Q + \Gamma(\mathcal{E}_i) + \Gamma(\mathcal{E}_f)$. $\mathrm{Im}\omega_Q$ has been given in (4.33) and both $\Gamma(\mathcal{E}_i)$ and $\Gamma(\mathcal{E}_f)$ can be estimated from the line shape of the elastic peak. We get

$$\frac{dR}{d\mathcal{E}_f} = \frac{\pi}{2}\left(\frac{me}{\hbar^2}\right)^2 \int d\vec{Q} \times \frac{\hbar\omega_Q}{|k_{ix}k_{fx}|\mathfrak{D}(Q)} \frac{\Gamma}{\pi[(\mathcal{E}_i - \mathcal{E}_f - \hbar\omega_Q)^2 + (\frac{1}{2}\Gamma)^2]} \times |\langle \xi^-_{k_{fx}}(x) | \phi^+_{\vec{Q}}(x) | \xi^+_{k_{ix}}(x)\rangle\rangle|^2 \quad . \tag{5.6}$$

Integration over $\vec{Q}$ in (5.6) should be carried out only up to $Q = Q_{c2}$ at which point $\mathrm{Im}\omega_{Q_c}$ is comparable with $\mathrm{Re}\omega_{Q_c}$. This physical restriction on the size of Q occurs because of LD which has the effect that SP with wavelengths smaller than the interelectronic spacing are so strongly damped as to be nonexistent. From the form of (5.6) we can easily see that $dR/d\mathcal{E}_f$ peaks at $\mathcal{E}_f = \mathcal{E}_i - \hbar\omega_{Cs}/\sqrt{2}$ because as Fig. 2 indicates there is a large volume of phase space for which $\mathrm{Re}\omega_Q \approx \omega_{Cs}/\sqrt{2}$. We have performed numerical calculations[26] in the $W(100)$-Cs case studied experimentally.[9,10] The potential U has been taken approximately as a steplike potential with the magnitude of the steps determined by the work functions and Fermi energies of the materials. Results of this calculation have been previously reported.[26] The position, width, and strength of the calculated SP peaks[26] are in agreement with experiment[9,10] if we take $\mathrm{Im}\omega_Q$ as in (4.33) (which will give a value of $Q_{c2} \approx Q_{Cs} \equiv \omega_{Cs}/v_F$) and both $\Gamma(\mathcal{E}_i)$ and $\Gamma(\mathcal{E}_f)$ of the order of 1 eV.[46] The behavior of the total backscattered electrons with 1-SP emission as a function of the thickness d of the Cs layer is also in qualitative agreement with experiment.

The method presented can also be applied to SP excitation in photoemission if we adopt Berglund and Spicer's model[11] for photoemission processes. The photoexcited electron on traveling to and escaping across the surface can emit SP's. The contribution to the EDC due to SP emission is given in exactly the same form as (5.6), except new sets of scattering states ξ^-_f and ξ^+_i appropriate to the photoemission geometry have to be used, and d has to be taken as infinite. Numerical calculations for Cs at photon energy ~10 eV, with the same SP parameters as used previously for calculation of the $W(100)$-Cs LEED spectrum, predict that photoelectrons which have suffered 1-SP inelastic scattering constitute a sizable fraction ($\sim\frac{1}{2}$) of the primary and show a broad peak at the SP frequency $\omega_{Cs}/\sqrt{2}$ whose width is ~2 eV. This result has verified that the excitation of SP contributes significantly to photoemission processes as was first pointed out by Calcott and MacRae[29] and recently observed in the alkali metals by Smith and Spicer.[11] We conclude by commenting briefly about the hypothesis of a Mott transition in the intermediate second Cs layer with 2×2 structure inferred by MacRae *et al.*[10] from the lack of SP structure.[47] The SP modes under consideration exist only when $Q > 1/d$. However, SP are defined only for $Q < Q_c$. Thus if $1/d \gtrsim Q_c$, the SP do not exist at all. As the Cs coverage increases, $1/d$ decreases and Q_c increases so that eventually $Q_c \gg 1/d$ and then SP become well-defined eigenmodes. Thus a smooth transition from no SP contribution to strong SP contribution should be expected on the basis of the present analysis as the Cs coverage increases, if one assumes that the density of free carriers changes smoothly. Consequently a nonsmooth change in the SP structure would imply a nonsmooth change in the density of free carriers. However, before the Mott transition hypothesis can be considered as established, it seems that further experiments are needed to prove beyond any doubt the nonsmooth nature of the variation of the SP structure as the Cs coverage changes and to clarify the nature of the 2×2 layer.

VI. CONCLUSIONS

We have studied the SP excitations in interfaces, quantized them, and worked out the el-SP interaction Hamiltonian. Then in the case of electron tunneling, we have calculated the extra current contribution due to emission of SP as electrons tunnel, thereby opening up new conductance channels. In LEED and in photoemission, we have calculated modifications of the electron inelastic spectrum and the EDC, respectively, due to inelastic SP scattering by using the "two-potentials" scattering formalism. In each of these calculations good agreement between theory and experiment has been obtained for both magnitude of the effect and its line shape. Moreover, in the case of electron tunneling, the sensitive dependence of the theoretical line shape on only a single parameter α characteristic of SP Landau damping enables a first determination of it from tunneling data. This experimentally determined value of α is in excellent agreement with the theoretical estimate given by Feibelman.[30] This seems to confirm the interpretation of the experimental results as due to inelastic scattering by SP.

*Part of this work was sponsored by the Department of the Air Force.

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