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Dielectric response of a semi-infinite layered electron gas and Raman scattering from its bulk and surface plasmons

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An exact solution of the random-phase-approximation equations is worked out for the density-density correlation function of a semi-infinite system of two-dimensional electron-gas layers, with different dielectrics outside and inside the layered system. From this solution, analytic formulas are derived for the dispersion relations of the bulk and surface plasmons and for the intensity of the light scattered inelastically from such a system. The intensity is written as a sum of the bulk and the surface terms. The theory is applied to semiconductor multilayers. The line shape of the bulk-plasmon peak, obtained after cancellation of van Hove singularities in the bulk piece by the surface piece, is compared with experiment. Conditions for observation of the Giuliani-Quinn surface plasmon are outlined.

I. INTRODUCTION

The electronic properties of a layered electron gas (LEG) have recently attracted much attention.[1–6] In particular, the predicted dispersion relation for the bulk plasmon[5,6] of the LEG, which is quite different from the dispersion relations for the plasmon in a two-dimensional[7] or a three-dimensional[8] electron gas, was confirmed experimentally by Olego *et al.*[1] in an experiment of inelastic light scattering from GaAs-(AlGa)As heterostructures. The bulk-LEG-plasmon dispersion relation is[1,5,6]

$$\omega_p(\mathbf{q},q_z)=\left[\frac{2\pi ne^2}{\epsilon m}q\frac{\sinh(qd)}{\cosh(qd)-\cos(q_zd)}\right]^{1/2}, \qquad (1)$$

where $\mathbf{q}$ and q_z are the components of the plasmon wave vector parallel and perpendicular to the planes, n is the density of the electrons (per unit area) in the plane, m is the electron mass, ϵ is the background dielectric constant, and d is the distance between two successive layers. Because electrons are confined to layers, this system has a surface plasmon only when the background dielectric constant of the LEG differs from that outside the LEG. The dispersion relation for the surface plasmon was obtained by Giuliani and Quinn,[2] by imposing the standard electrodynamic boundary conditions at the layers of a semi-infinite LEG.

To the best of our knowledge, no theory of Raman scattering from a LEG exists. In this work we present such a theory. (A short paper of parts of this work has already been published.[9]) We calculate exactly [in the random-phase approximation (RPA)] the density-density correlation function (or, effectively, the susceptibility) for a semi-infinite LEG. It can be written as the sum of a bulk and a surface term. From this correlation function we calculate the surface-plasmon dispersion relation. A full theory for the Raman scattering cross section is given, including line shapes and intensities of various collective modes. We shall see that ignoring the surface term leads to spurious peaks in the Raman spectrum at the one-dimensional Van Hove singularities of the plasmon density of states.

Following Visscher and Falicov,[10] we take the electron density to have a δ-function localization in the plane. The electrons are free to move in the plane and electrons in different planes interact only via the Coulomb interaction. The possibility of tunneling between two planes and of intraband transitions has been ignored. The planes of the two-dimensional electron gas are situated at $z=ld$ where l goes from 0 to ∞ and are embedded in a space of dielectric constant ϵ_0 for $z<-d'$ and ϵ for $z>-d'$. This system is shown schematically in Fig. 1.

The plan of the paper is as follows. In Sec. II the density-density correlation function is derived for the semi-infinite LEG described above in the random-phase approximation. In Sec. III, an exact RPA dispersion relation for the surface plasmon is obtained which is shown in the Appendix to be identical to the Giuliani-Quinn result. Section IV contains the theory of Raman scattering: the Raman intensity is written in terms of the density-density correlation function. In Sec. V the Raman spectrum is calculated theoretically and the bulk-plasmon line shape is compared with experiment. Predictions are made about

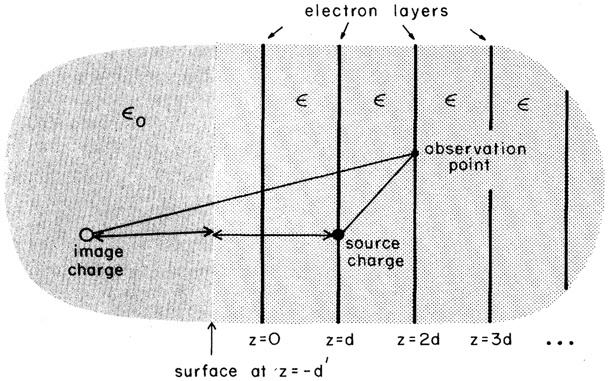

FIG. 1. The semi-infinite system of layered electron gas under consideration.

the experimental conditions in which the surface plasmon would be observable.

II. DENSITY-DENSITY CORRELATION FUNCTION

The time-ordered density-density correlation function is defined in the usual way:

$$D(\mathbf{x},z,t;\mathbf{x}',z',t')=-i\langle Tn(\mathbf{x},z,t)n(\mathbf{x}',z',t')\rangle \ , \tag{2}$$

where $\mathbf{x}$ and $\mathbf{x}'$ are vectors parallel to the planes and T is the time-ordering operator. (In the following discussion all vectors will be parallel to the plane, or perpendicular to the z axis.) The quantity $n(\mathbf{x},z,t)$ is the electron-density operator in the Heisenberg representation. $D(\mathbf{x},z,t;\mathbf{x}',z',t')$ depends on $\mathbf{x}$ and $\mathbf{x}'$ only through the difference $\mathbf{x}$-$\mathbf{x}'$ due to translational symmetry in the xy plane, and therefore one can Fourier transform it in the variables $\mathbf{x}$-$\mathbf{x}'$ and t-t' to get $D(\mathbf{q},\omega;z,z')$. The Coulomb interaction between two electrons situated at $(\mathbf{x},z)$ and $(\mathbf{x}',z')$ is given for $z\geq 0$, $z'\geq 0$ by[11]

$$V(\mathbf{x}-\mathbf{x}';z,z')=V_0+V_I \ , \tag{3a}$$

$$V_0=e^2/\epsilon[(\mathbf{x}-\mathbf{x}')^2+(z-z')^2]^{1/2} \ , \tag{3b}$$

$$V_I=\alpha e^2/\epsilon[(\mathbf{x}-\mathbf{x}')^2+(z+z'+2d')^2]^{1/2} \ , \tag{3c}$$

$$\alpha=(\epsilon-\epsilon_0)/(\epsilon+\epsilon_0) \ . \tag{4}$$

This can also be Fourier transformed with respect to $\mathbf{x}-\mathbf{x}'$, giving

$$V(q;z,z')=V_{\mathbf{q}}f(q;z,z') \ , \tag{5}$$

$$V_{\mathbf{q}}=2\pi e^2/\epsilon q \ , \tag{6}$$

$$f(q;z,z')=e^{-q|z-z'|}+(\alpha e^{-2qd'})e^{-q|z+z'|} \ . \tag{7}$$

In the rest of the paper, we shall take $d'=0$. The case of nonzero d' can be easily obtained by replacing α by $\alpha\exp(-2qd')$ in all the following formulas.

Let $D^0(\mathbf{q},\omega;z,z')$ be the value of $D(\mathbf{q},\omega;z,z')$ in the absence of Coulomb interactions. The standard RPA treatment yields (Fig. 2)

$$D(z,z')=D^0(z,z') + \int dz_1 \int dz_2 D^0(z,z_1)Vf(z_1,z_2)D(z_2,z') \ , \tag{8}$$

where $\mathbf{q}$ and ω dependence is everywhere suppressed. From the fact that the density of electrons has a δ-function localization at $z=ld$, $D(z,z')$ and $D^0(z,z')$ must have the following structure:

$$D(z,z')=\sum_{l,l'}\delta(z-ld)\delta(z'-l'd)D(l,l') \ , \tag{9}$$

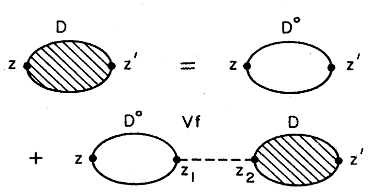

FIG. 2. Dyson's equation for the electronic density-density correlation function $D(z,z')$.

$$D^0(z,z')=\sum_{l}\delta(z-ld)\delta(z'-ld)D^0 \ . \tag{10}$$

Note that for $D^0(z,z')$ to have a nonvanishing value, both z and z' must be on the same plane because $D^0(z,z')$ is the value of $D(z,z')$ in the absence of Coulomb interactions, i.e., when there is no coupling between two different planes. D^0 must also be l independent. It is given by, in standard notation,

$$D^0(\mathbf{q},\omega+i\gamma)=2\int\frac{d^2p}{(2\pi)^2}\frac{f(\mathbf{p}+\mathbf{q})-f(\mathbf{p})}{\epsilon(\mathbf{p}+\mathbf{q})-\epsilon(\mathbf{p})-\omega-i\gamma} \ , \tag{11}$$

where $\omega>0$. D^0 has been calculated exactly by Stern[7] for $\gamma\to 0^+$ and $T=0$. The calculation can be extended to the case of finite γ and the answer is

$$D^0(q,\omega+i\gamma)=-\frac{n}{2\epsilon_F}\frac{2k_F}{q}\left[2\left[\frac{q}{2k_F}\right]-(a_+^2-1)^{1/2} + (a_-^2-1)^{1/2}\right] \ , \tag{12}$$

where $a_{\pm}=(\omega+i\gamma)/qv_F\pm q/2k_F$. The quantities ϵ_F and k_F are the Fermi energy and the Fermi wave vector, and the complex square root is chosen to be the branch with positive imaginary part. For $\omega \gg qv_F$, $\mathrm{Re}D^0(q,\omega)$ can be approximated by $nq^2/m\omega^2$.

While calculating the Raman intensity, we shall use the corrected form of D^0, suggested by Mermin,[12] which is

$$\frac{D^0(q,\omega+i\gamma)(1+i\gamma/\omega)}{[1+i(\gamma/\omega)D^0(q,\omega+i\gamma)/D^0(q,0)]} \ . \tag{13}$$

Making this correction does not lead to significant changes in the final result.

On substituting Eqs. (9) and (10) in Eq. (8) we get

$$D(l,l')=D^0\delta_{ll'}+D^0V\sum_{l_2}f(l,l_2)D(l_2,l') \tag{14}$$

with

$$f(l,l_2)=\exp(-qd\,|\,l-l_2\,|)+\alpha\exp(-qd\,|\,l+l_2\,|) \ .$$

This is the equation that we seek to solve for $D(l,l')$.

A. Bulk properties

To get the bulk properties of the system, we take the planes to be situated at $z=ld$ with l going from $-\infty$ to $+\infty$, and the dielectric constant to be ϵ everywhere so that $f(l,l')=\exp(-qd\,|\,l-l'\,|)$. The derivation of the density-density correlation function is well known but we sketch it here for comparison with the semi-infinite case. In this case, $D^b(l,l')=D^b(l-l')$ and with a change in variables Eq. (14) becomes

$$D^b(l)=D^0\delta_{l0}+D^0V\sum_{m}\exp(-qd\,|\,l-m\,|)D^b(m) \ . \tag{15}$$

Now we make the following transformation:

$$D^b(l)=\frac{d}{2\pi}\int_{-\pi/d}^{\pi/d}dq_z D^b(q_z)e^{-iq_z ld} \ , \tag{16a}$$

$$D^b(q_z)=\sum_{l}e^{iq_z ld}D^b(l) \ , \tag{16b}$$

$$D^b(q_z)=D^0+D^0V\left[\sum_l e^{ilq_zd-|l|qd}\right]D^b(q_z)\ . \tag{17}$$

The sum over l can be done and is equal to

$$\sinh(qd)/[\cosh(qd)-\cos(q_zd)]\ .$$

Thus the solution of Eq. (15) is written as

$$D^b(q_z)=D^0/\epsilon(q_z)\ , \tag{18}$$

$$\epsilon(q_z)=1-D^0V\sinh(qd)/[\cosh(qd)-\cos(q_zd)]\ . \tag{19}$$

The bulk-plasmon dispersion relation (1) follows by setting $\epsilon(q_z)$ in Eq. (19) to zero and using the small-q value for $D^0(q,\omega)$, i.e., $D^0\approx nq^2/m\omega^2$.

B. Semi-infinite LEG

In this case $D(l,l')$ depends on l and l' separately and a simple solution is no longer possible. But it is still possible to solve Eq. (14) for $D(l,l')$ by making the following Fourier transformation:

$$D(q_z,q_z')=\frac{1}{N}\sum_{l,l'=0}^{N-1}e^{-iq_zld}e^{iq_z'l'd}D(l,l')\ , \tag{20}$$

$$D(l,l')=\frac{1}{N}\sum_{q_z,q_z'}e^{iq_zld}e^{-iq_z'l'd}D(q_z,q_z')\ , \tag{21}$$

where q_z and q_z' can take values $2\pi n/Nd$, $N=$number of planes, $n=0,\ldots,N-1$. Later we shall be interested in the limit $N\to\infty$. This transformation is similar to one used by Grecu[13] in connection with layered thin films. We tested Grecu's Fourier transform, and found that although the formulas in the intermediate stages look very different, the Raman intensity was the same by both methods.

The Fourier transform of Eq. (14) is

$$D(q_z,q_z')=D^0\delta(q_z,q_z')+D^0V\sum_{k_z}f(q_z,k_z)D(k_z,q_z')\ , \tag{22}$$

$$f(q_z,k_z)\equiv\frac{1}{N}\sum_{l,l'=0}^{N-1}e^{-iq_zld}f(l,l')e^{ik_zl'd} \tag{23a}$$

$$\begin{aligned}=\delta(q_z,k_z)\sinh(qd)/P(q_z)+[(1-e^{-Nqd})/2NP(q_z)P(k_z)]\{&(1+\tfrac{1}{2}\alpha'e^{2qd})+(1+\tfrac{1}{2}\alpha')e^{i(q_z-k_z)d}\\ &-[\cosh(qd)+\tfrac{1}{2}\alpha'e^{qd}](e^{iq_zd}+e^{-ik_zd})\}\ ,\end{aligned} \tag{23b}$$

$$P(q_z)=\cosh(qd)-\cos(q_zd)\ , \tag{24}$$

$$\alpha'=\alpha(1-e^{-Nqd})\ . \tag{25}$$

The first term of the matrix $f(q_z,k_z)$ is diagonal and corresponds to bulk LEG. The second term vanishes as N goes to ∞. However, in this limit the matrix dimension is also infinite, and the second term of (23b) will yield finite corrections to $D(l,l')$ when l or l' are near the surface ($l=0$). Formally inverting Eq. (22), we have

$$D^0D^{-1}(k_z,q_z')=\delta(k_z,q_z')-D^0Vf(k_z,q_z')\ . \tag{26}$$

The matrix f is a finite sum of factorable pieces and thus can be explicitly inverted. By direct multiplication it can be verified that

$$D(q_z,k_z)=\delta(q_z,k_z)D^b(q_z)+D^s(q_z,k_z)\ , \tag{27a}$$

where D^b is given by Eq. (18), and the surface part is

$$D^s(q_z,k_z)=[(1-e^{-Nqd})(D^0)^2V/2N\epsilon(q_z)\epsilon(k_z)P(q_z)P(k_z)Q][A-B(e^{iq_zd}+e^{-ik_zd})+Ce^{i(q_z-k_z)d}]\ . \tag{27b}$$

This is the exact RPA answer for a finite LEG of N layers. It is expressed as the bulk answer D^b plus a surface correction D^s. Corrections beyond RPA can be included in D^0 if needed. The quantities A, B, C, and Q are defined as

$$A=G\sinh^2(qd)+1+\tfrac{1}{2}\alpha'e^{2qd}\ , \tag{28a}$$

$$B=H\sinh^2(qd)+\cosh(qd)+\tfrac{1}{2}\alpha'e^{qd}\ , \tag{28b}$$

$$C=G\sinh^2(qd)+1+\tfrac{1}{2}\alpha'\ , \tag{28c}$$

$$\begin{aligned}Q=1-G[2+\tfrac{1}{2}\alpha'(1+e^{2qd})]+(H^2-G^2)\sinh^2(qd)\\ +H[2\cosh(qd)+\alpha'e^{qd}]\ ,\end{aligned} \tag{29}$$

$$G=D^0V[(1-e^{-Nqd})/2N]\sum_{q_z}1/[P(q_z)]^2\epsilon(q_z)\ , \tag{30a}$$

$$H=D^0V[(1-e^{-Nqd})/2N]\sum_{q_z}e^{iq_zd}/[P(q_z)]^2\epsilon(q_z)\ . \tag{30b}$$

In the limit $N\to\infty$, the sums over q_z in Eq. (30) can be explicitly performed:

$$\frac{1}{N}\sum_{q_z} g(q_z) \to \frac{d}{2\pi}\int_0^{2\pi/d} dq_z g(q_z) , \quad (31)$$

$$G=\tfrac{1}{2}[(b^2-1)^{-1/2}-1/\sinh(qd)]/\sinh(qd) , \quad (32a)$$

$$H=\tfrac{1}{2}[u^{-1}(b^2-1)^{-1/2}-e^{-qd}/\sinh(qd)]/\sinh(qd) , \quad (32b)$$

$$Q=\tfrac{1}{2}\{1-(b^2-1)^{-1/2}[1-b\cosh(qd)]/\sinh(qd)\} - \tfrac{1}{2}\alpha e^{qd}(b^2-1)^{-1/2}[\cosh(qd)-b]/\sinh(qd) , \quad (33)$$

$$b=\cosh(qd)-D^0 V\sinh(qd) , \quad (34a)$$

$$u=b+(b^2-1)^{1/2} . \quad (34b)$$

The imaginary part of b is always positive. Again the complex square root is chosen to be the one with imaginary part greater than zero. Notice that $|u|>1$.

Using Eqs. (21) and (27) and the formula

$$\frac{d}{2\pi}\int_0^{2\pi/d} dq_\perp \frac{e^{iq_\perp dl}}{b-\cos(q_\perp d)} = u^{-|l|}(b^2-1)^{-1/2} , \quad (35)$$

we can write an analytical formula for the real-space correlation function $D(l,l')$ in the limit $N\to\infty$:

$$D(l,l')=D^0\left[\delta_{ll'}+D^0V\sinh(qd)(b^2-1)^{-1/2}u^{-|l-l'|}+D^0V(1-e^{-Nqd})\frac{u^2A-2uB+C}{2u^2(b^2-1)Q}u^{-(l+l')}\right] . \quad (36)$$

The first two terms on the right-hand side give $D^b(l,l')$ and the last term gives $D^s(l,l')$. As expected, $D^b(l,l')$ depends only on the difference $l-l'$, and $D^s(l,l')$ decays in magnitude as one goes far from the surface, i.e., as l and l' become large.

III. BULK AND SURFACE PLASMONS

The poles of the density-density correlation function yield the energies of collective excitations that couple to the ground state via the density operator. Thus the poles of $D(q_z,k_z)$ given by Eq. (27) will give the plasmon energies.

(i) Bulk plasmon. As has already been noted, the pole $\epsilon(q_z)=0$ of $D^b(q_z)$ leads to the dispersion relation of the bulk plasmon, Eq. (1). The relation can also be written as $b=\cos(q_z d)$, where b is defined in (34a). The range $-1<b<1$ defines the plasmon band which occurs for a fixed $\mathbf{q}$ parallel to the plane by considering all possible values of q_z.

(ii) Surface plasmon. The dispersion of the surface plasmon is given by the relation

$$Q(\mathbf{q},\omega)=0 , \quad (37)$$

which describes the pole of the surface term D^s in Eq. (27). An analytic form of Q is given in Eq. (33); the dispersion relation (37) is exact in the RPA. When $\epsilon_0=\epsilon$ or $\alpha=0$, it reduces to $b=\cosh(qd)$ which does not have any nontrivial solution as can be seen from the definition of b in Eq. (34a). However, for $\alpha\neq 0$, Eq. (37) may have solutions. This equation may be rewritten as

$$(b^2-1)^{1/2}\sinh(qd)+\alpha e^{qd}b+\cosh(qd)(b-\alpha e^{qd})=1 . \quad (38)$$

In the Appendix we show that Eq. (38) agrees exactly with the result obtained by Giuliani and Quinn.[2] In particular, because of the presence of the factor $(b^2-1)^{1/2}$, this equation does not have any solution within the bulk-plasmon band $-1<b<1$. At the boundaries of the plasmon band, namely $b=\pm 1$, it has the simple solution $e^{-qd}=|\alpha|$. For $\alpha>0$ this solution continues above the bulk-plasmon band, and for $\alpha<0$ it continues below the bulk-plasmon band. We have plotted the dispersion relation of the surface plasmon in Fig. 3 for three values of α. In light scattering only a very small momentum exchange q is accessible and therefore it is desirable to have a large $|\alpha|$ in order to be able to see the surface plasmon.

IV. THEORY OF RAMAN SCATTERING

The coupling of the system to the external laser field $\mathbf{A}(\mathbf{x},z,t)$ is given by

$$H'_{\rm int}=\sum_i [e\mathbf{p}_i\cdot\mathbf{A}(\mathbf{r}_i,t)+e\mathbf{A}(\mathbf{r}_i,t)\cdot\mathbf{p}_i+e^2[\mathbf{A}(\mathbf{r}_i,t)]^2]/2m_e , \quad (39)$$

where $\mathbf{p}_i$ and $\mathbf{r}_i=(\mathbf{x}_i,z_i)$ are the momentum and position of the ith electron. For external laser frequencies ω_L small compared to interband electron resonances, we use the standard trick[14,15] of using only the A^2 term, but with

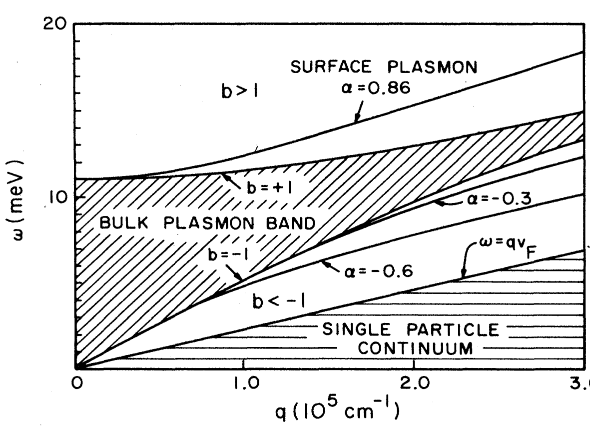

FIG. 3. Dispersion relation for the surface plasmon for certain values of α. The shaded region is the bulk-plasmon band and has no surface plasmon inside it. $\alpha=0.86$ corresponds to vacuum outside the semi-infinite LEG.

the bare mass m_e replaced by the band mass m, to account for the $\mathbf{p}\cdot\mathbf{A}$ terms. Then the interaction Hamiltonian becomes

$$H_{\text{int}}=\frac{e^2}{2m}\int d^2x\,dz\,n(\mathbf{x},z)A^2(\mathbf{x},z,t)\ . \tag{40}$$

Let us denote the state of the system composed of the electrons and photon before the scattering by $|I\rangle=|i,a\rangle$ and after the scattering by $|F\rangle=|f,b\rangle$. Here i,f are the electronic (many-body) states and a,b denote the photon quantum numbers. The scattering matrix squared and summed over the final electronic state is given by

$$\begin{aligned}\sum_f|S_{FI}|^2&=2\pi\sum_f|\langle F|H_{\text{int}}|I\rangle|^2\delta(E_F-E_I)\\&=\left[\frac{e^2}{2m}\right]^2\int d^2x\,dz\,dt\int d^2x'dz'dt'\langle i\,|\,n(\mathbf{x},z,t)n(\mathbf{x}',z',t')\,|\,i\rangle\\&\quad\times\langle a\,|\,\mathbf{A}^*(\mathbf{x},z,t)\cdot\mathbf{A}^*(\mathbf{x},z,t)\,|\,b\rangle\langle b\,|\,\mathbf{A}(\mathbf{x}',z',t')\cdot\mathbf{A}(\mathbf{x}',z',t')\,|\,a\rangle\ .\end{aligned} \tag{41}$$

This is the basic formula used for the calculation of the cross section for the Raman scattering. It contains a factor which depends solely on the electronic properties of the material, namely,

$$\langle i\,|\,n(\mathbf{x},z,t)n(\mathbf{x}',z',t')\,|\,i\rangle\ .$$

Call this factor $J(\mathbf{x}-\mathbf{x}',t-t',z,z')$ and its Fourier transform $J(q,\omega,z,z')$. At zero temperature $|i\rangle$ is the many-electron ground state and J is related to $D(\mathbf{q},\omega,z,z')$ in the following manner:

$$J(\mathbf{q},\omega,z,z')=i\theta(\omega)[D(\mathbf{q},\omega,z,z')-D^*(-\mathbf{q},-\omega,z,z')]\ . \tag{42}$$

This is a fluctuation-dissipation relation for zero temperature, i.e., when $|i\rangle$ is the many-electron ground state. It can be proved by transforming $J(\mathbf{x},z,t;\mathbf{x}',z',t')$ and $D(\mathbf{x},z,t;\mathbf{x}',z',t')$ to their spectral (Lehman) representation. The invariance of D [Eq. (9)] under $(\mathbf{q},\omega)\to(-\mathbf{q},-\omega)$ enables us to write Eq. (42) as

$$J(\mathbf{q},\omega,z,z')=-\theta(\omega)2\,\text{Im}D(\mathbf{q},\omega,z,z')\ . \tag{43}$$

The photon matrix elements in Eq. (41) are calculated in the following manner:

$$\mathbf{A}(\mathbf{x},z,t)=\sum_k\mathbf{A}_k\{a_k\exp[-i(\mathbf{k}\cdot\mathbf{x}-\omega_0t+k_zz)]+\text{H.c.}\}\ , \tag{44}$$

where $k_z=(\epsilon-\sin^2\theta)^{1/2}\omega_0/c$, ϵ is the complex dielectric constant of the material, and ω_0 is the frequency of the light incident on the surface making an angle θ with the z axis (the surface lies in the xy plane). If $\text{Re}\epsilon$ is large and θ small, we can approximately write

$$k_z=k+\frac{i}{2\delta},\quad k=\frac{\omega_0}{c}\text{Re}\sqrt{\epsilon}\ , \tag{45}$$

$$\delta^{-1}=2\frac{\omega_0}{c}\text{Im}\sqrt{\epsilon}\ . \tag{46}$$

$\mathbf{A}_k$ in Eq. (44) is related to the polarization of the photon outside the material.[16,17]

Using these formulas we find

$$\begin{aligned}&\langle a\,|\,\mathbf{A}^*(\mathbf{x},z,t)\cdot\mathbf{A}^*(\mathbf{x},z,t)\,|\,b\rangle\langle b\,|\,\mathbf{A}(\mathbf{x}',z',t')\cdot\mathbf{A}(\mathbf{x}',z',t')\,|\,a\rangle\\&\qquad=(\mathbf{A}_a\cdot\mathbf{A}_b)^2\exp\left[-i[\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')-\omega(t-t')]-i2k(z-z')-\frac{(z+z')}{\delta}\right]\ ,\end{aligned} \tag{47}$$

where

$$\mathbf{q}=\mathbf{k}_a-\mathbf{k}_b\ ,\quad \omega=\omega_a-\omega_b \tag{48}$$

and k and δ are the same as in Eqs. (45) and (46) with ω_0 replaced by ω_a or ω_b (because we are interested in the case when ω is very small and $\omega_a\sim\omega_b$).

Using Eqs. (9), (43), and (47), Eq. (41) becomes

$$\begin{aligned}&\sum_f|S_{FI}|^2/AT=2(e^2/2m)^2(\mathbf{A}_a\cdot\mathbf{A}_b)^2I(\omega)\theta(\omega)\ ,\\&I(\omega)=-\sum_{l,l'}\text{Im}D(\mathbf{q},\omega;l,l')e^{-(l+l')d/\delta}e^{-2ikd(l-l')}\ ,\end{aligned} \tag{49}$$

which gives us the probability per unit time (T) per unit area (A) of an exchange of momentum $\mathbf{q}$ (in the plane of the LEG) and energy ω. As $D(\mathbf{q},\omega,l,l')$ is symmetric under the exchange of l and l', $\exp[-2ik(l-l')d]$ can be replaced by $\cos[2k(l-l')d]$ and we explicitly see that the probability is real. The different factors in Eq. (49) are intuitively understandable. We have the usual $\text{Im}D$ which is characteristic of processes where energy is transferred to electrons by a probe coupled to the density. The factor $\exp[-(l+l')d/\delta]$ decays as we go away from the surface, and how many layers are important depends on the value of d/δ. The factor $\exp[2ikd(l-l')]$ is a coherence factor which would generate perpendicular momentum conservation if δ were infinite.

V. INTENSITY OF RAMAN SCATTERED LIGHT

Now we can substitute the expression for $D(l,l')$, Eq. (36), into Eq. (49) and obtain the intensity of the Raman scattered light as a function of its energy loss for a fixed value of momentum exchange. After performing the sums over l and l', $I(\omega)$, which is proportional to the intensity, is given by

$$I(\omega)=-\mathrm{Im}D^0\left[(1-e^{-2d/\delta})^{-1}\left[1+\frac{D^0V\sinh(qd)(u^2e^{2d/\delta}-1)}{(b^2-1)^{1/2}E}\right]+\frac{D^0Ve^{2d/\delta}(u^2A-2uB+C)}{2Q(b^2-1)E}\right], \tag{50}$$

$$E=u^2e^{2d/\delta}+1-2ue^{d/\delta}\cos(2kd)\ . \tag{51}$$

The first term on the right-hand side of Eq. (50) gives the bulk contribution $I^b(\omega)$ and the second term gives the surface contribution $I^s(\omega)$.

A. Bulk plasmon

Now we compare this result with the experiment of Olego *et al.*[1] The intensity given by Eq. (50) is plotted in Fig. 4, as is the intensity observed in the experiment. For comparison we also include the intensity given by a naive theory, i.e., $I(\omega)\sim-\mathrm{Im}D^0/\epsilon(q,2k)$. The naive formula for the intensity, unlike Eq. (50), does not take into account the broadening of the perpendicular momentum k, caused by decay of the photon inside the material due to the lack of translational invariance in the z direction. The complete theory, Eq. (50), gives a much better agreement with the experiment. In order to calculate $I(\omega)$, the values of parameters have been chosen to be the same as those of sample 1 of the experiment of Olego *et al.*[1] They are $q=4.8\times10^4\text{cm}^{-1}$ (which corresponds to $\theta=20°$ in their notation), effective mass $m=0.07m_e$, static dielectric constant $\epsilon=13.1$, electron density $n=7.3\times10^{11}$ cm^{-2}, $d=890$ Å, $\delta=6000$ Å, $2kd=4.94$, $\epsilon_0=1$, electron mobility $\mu=5\times10^4$ cm^2/Vs, and[18] $\gamma=e/m\mu=0.3$ meV.

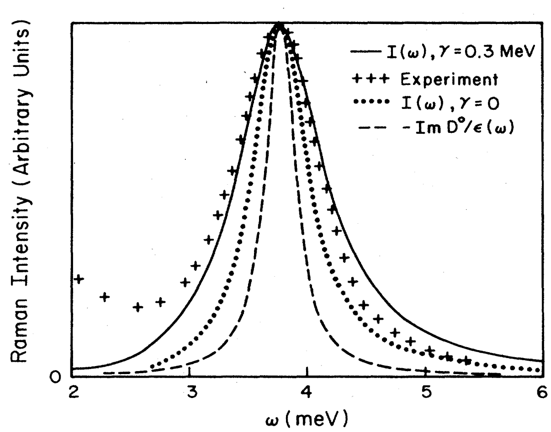

FIG. 4. Comparison between the experimental and theoretical line shapes of the bulk-plasmon peak in the Raman spectrum. The experimental peak has been shifted along the ω axis to align it with the other peak. The result of a naive theory $I(\omega)=-\mathrm{Im}\,D^0/\epsilon(\omega)$ is also shown. All the spectra are normalized separately.

To see the influence of the purity (or of γ) of the sample on the line shape, we plot also the intensity for $\gamma=0^+$, or mobility $\mu=\infty$ (and all the other parameters same as before), in Fig. 4. This shows that the width of the plasmon peak arises mainly from the spread of the perpendicular momentum k and the decay of the photon, with electronic damping giving a smaller contribution. Even for a completely pure sample the plasmon peak would not be a δ function [as one would get from the naive formula $I(\omega)\sim-\mathrm{Im}D^0/\epsilon(q,2k)$].

B. Van Hove singularities

Hidden in the calculated intensity $I(\omega)$ in Fig. 4 is a surprising cancellation between bulk and surface parts at the boundaries $b=\pm1$ of the bulk-plasmon band. This is shown in Fig. 5 in which $I^b(\omega)$, $I^s(\omega)$, and $I(\omega)$ have been plotted separately for $\gamma=0.1$ meV. $I^b(\omega)$ has an interesting structure which is explained as follows. The denominator of Eq. (50) becomes zero or small at three separate frequencies given by

$$b=\mp1\ , \tag{52}$$

$$b=\cos(2kd)\ , \tag{53}$$

where Eq. (53) is the solution of $E=0$ in the limit $d/\delta\rightarrow0$, and is the same as the dispersion relation of a

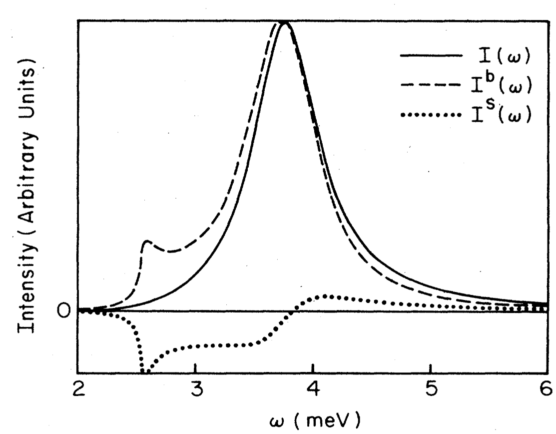

FIG. 5. $I^b(\omega)$, $I^s(\omega)$, and $I(\omega)$. When the surface term I^s is added to the bulk term I^b to get I, it cancels the peak in I^b at $\omega_{\min}\cong2.5$ meV.

plasmon with wave vector $(\mathbf{q},2k)$. For the parameters given above, b is equal to -1, $+1$, and $\cos(2kd)$ at $\omega_{\min}=2.5$ meV, $\omega_{\max}=11.2$ meV, and $\omega_p=3.7$ meV. The two energies $\omega_{\min}(q)$ and $\omega_{\max}(q)$ give the limits of the plasmon band which is the continuum of energies a plasmon can assume with a fixed in-plane wave vector $\mathbf{q}$ but an arbitrary wave vector q_z, and define the shaded region in Fig. 3. $I^b(\omega)$ has peaks at $\omega_{\min}$ and ω_p, and also one at $\omega_{\max}$ which is too small to be visible on the scale of the plot. For $\gamma \gtrsim 0.1$ meV the peak at $\omega_{\min}$ is not very well defined and at $\gamma=0.3$ meV there is only a bump at $\omega_{\min}$. To understand better the origin of these peaks in $I^b(\omega)$, it is instructive to use Eqs. (21), (27), and (49) to write

$$I^b(\omega)=-\frac{1}{N}\mathrm{Im}\sum_{q_z} D^0 S(q_z)/\epsilon(q_z)\ , \tag{54}$$

$$[S(q_z)]^{-1}=2e^{d/\delta}\{\cosh(d/\delta)-\cos[(q_z-2k)d]\}\ . \tag{55}$$

Using the small-q formula for $D^0 \sim nq^2/m\omega^2$, we can write $\epsilon(q_z)$ in plasmon pole approximation as

$$\epsilon(\mathbf{q},q_z,\omega)=1-\omega_p^2(\mathbf{q},q_z)/\omega(\omega+i\eta)\ , \tag{56}$$

where ω_p is defined in Eq. (1). Equation (54) now becomes

$$I^b(\omega)=\tfrac{1}{2}\pi D^0 \omega N(\mathbf{q},\omega) S(\omega)\ , \tag{57}$$

where $N(\mathbf{q},\omega)$ is the one-dimensional density of plasmon states at $(\mathbf{q},\omega)$ given by

$$N(\mathbf{q},\omega)=\frac{1}{N}\sum_{q_z}\delta(\omega-\omega_p(\mathbf{q},q_z))$$
$$=(\Omega^2/\pi\omega^3)\sinh(qd)(1-K^2)^{-1/2}\ , \tag{58}$$

$$K=\cosh(qd)-(\Omega^2/\omega^2)\sinh(qd),\quad \Omega^2=2\pi n e^2 q/m\epsilon\ . \tag{59}$$

$S(\omega)$ in Eq. (57) is $S(q_z)$ from Eq. (55) evaluated at $\cos(q_z d)=K$, which is the plasmon dispersion relation (1). Thus $S(\omega)$ is peaked at ω_p with a width determined by d/δ. The density of states $N(\mathbf{q},\omega)$ has one-dimensional Van Hove singularities at $K=\pm 1$, i.e., at $\omega_{\max}$ and $\omega_{\min}$. The functions $S(\omega)$ and $\omega N(\omega)$ are plotted in Fig. 6. The origin of the extra peak of $I^b(\omega)$ shown in Fig. 5 at $\omega_{\min}$ is clearly a density-of-states effect made visible by the finite width of $S(\omega)$ which arises from the decay length δ of the laser photon.

When we add $I^s(\omega)$ to $I^b(\omega)$ to obtain the total intensity $I(\omega)$, the peaks in $I^b(\omega)$ at $\omega_{\min}$ and $\omega_{\max}$ are completely canceled by negative peaks at the same energies in $I^s(\omega)$, and $I(\omega)$ has no structure at all at $\omega_{\min}$ and $\omega_{\max}$. This remarkable disappearance of these two peaks can be seen analytically in the case $\epsilon_0=\epsilon$ or $\alpha=0$ by adding the two terms in Eq. (50) and noticing that the numerator is proportional to $(b^2-1)^{1/2}$ which exactly cancels in the denominator the factor $(b^2-1)^{1/2}$ which was responsible for these two peaks. We are not certain about the physical origin of this cancellation, which seems to be similar to a cancellation found by Stroscio *et al.*[19] in an analysis

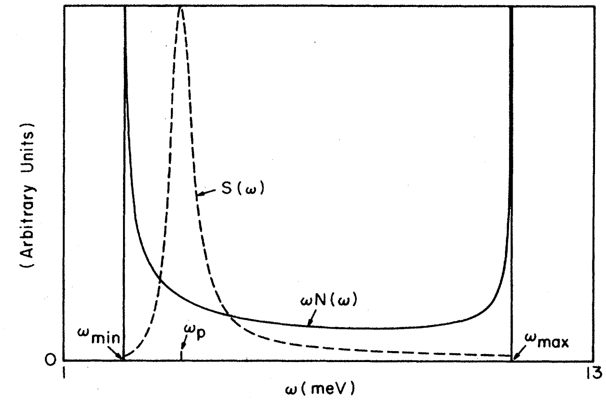

FIG. 6. The plasmon density of states $\omega N(\omega)$ [Eq. (58)] and structure factor $S(q_z)$ [Eq. (55)] evaluated at $\omega=\omega_p(q,q_z)$ for the same parameters as in Fig. 5. As shown in Eq. (57), the product of these two functions determines $I^b(\omega)$ and accounts for the peaks shown in Fig. 5.

of surface vibrational resonances. Apparently the presence of the surface destroys the possibility of collective oscillations with neighboring planes strictly in phase $(\Delta\phi=q_z d=0)$ or out of phase $(\Delta\phi=q_z d=\pi)$ which are needed to obtain Van Hove singularities.

The line shape of the plasmon peak obtained by this theory is in excellent agreement with the experiment as shown in Fig. 4. Note that there is no free parameter in the theory. No bump was observed at $\omega_{\min}$—but, indeed, for a better verification of the cancellation of the spectral weight at the bulk-plasmon band edges the experiment would have to be repeated on purer samples.

C. Surface plasmon

According to Eq. (50), the surface plasmon whose dispersion relation is $Q(q,\omega)=0$ should be detectable by Raman scattering. In Fig. 7 we give the intensity of the

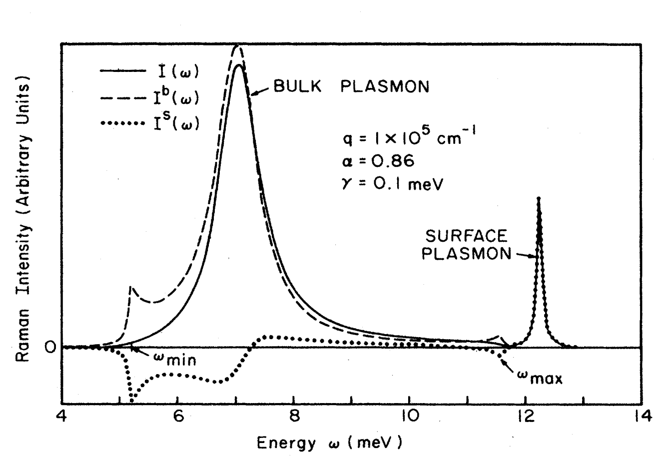

FIG. 7. The bulk and surface contributions of the intensity [$I^b(\omega)$ and $I^s(\omega)$, respectively] and the total intensity $I(\omega)$ are plotted for the values of parameters shown in the picture. $\alpha=0.86$ corresponds to vacuum outside the LEG. The surface plasmon is seen at 12.2 meV.

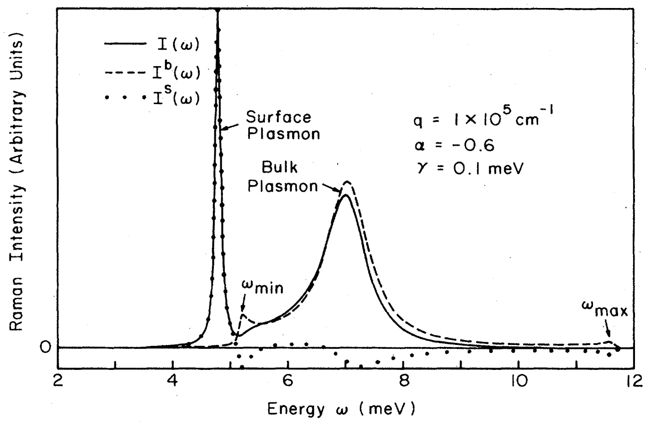

FIG. 8. $I^b(\omega)$, $I^s(\omega)$, and $I(\omega)$ are plotted for $\alpha=-0.6$ which means that the dielectric constant ϵ_0 outside the semi-infinite LEG is greater than the background dielectric constant ϵ inside the LEG. The surface plasmon now appears below the bulk-plasmon band at 4.8 meV.

Raman scattered light predicted by Eq. (50) for $q=1.0\times10^5$ cm^{-1}, $\gamma=0.1$ meV, and $\epsilon_0=1$ (vacuum in $z<0$ space). The surface plasmon is visible at ~12.2 meV. The intensity at the surface plasmon is enhanced as the mobility of the electrons increases, or as γ decreases. The width of the surface plasmon comes from γ alone which means that it is free from Landau damping, as pointed out by Giuliani and Quinn.[2]

A surface plasmon on the lower side of the bulk-plasmon band can be observed if the dielectric constant ϵ_0 outside the LEG can be increased significantly above ϵ, making α negative. In Fig. 8 we give the energy spectrum of the scattered-light intensity for $\alpha=-0.6$. Such a large value of α can be possible if the experiment is done on a layered system made of a material with a low dielectric constant ϵ.

Note added in proof. We have recently[20] worked out the response functions of a film of finite thickness, using the same method. This calculation shows that transmission Raman spectroscopy provides a way of enhancing the surface plasmon intensity.

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APPENDIX

In this appendix we show the equivalence of the dispersion relation of the surface plasmon given by Eq. (38) with the one obtained by Giuliani and Quinn in Ref. 2. Equations (1) and (2) of Ref. 2 can be written in our notation as

$$b=\cos(q_z d)\ , \tag{A1}$$

$$e^{d/\xi}=\epsilon/[\epsilon_0\sinh(qd)-\epsilon\sinh(qd)+2\epsilon b]\ . \tag{A2}$$

For definiteness consider the regime $b>1$. Then according to Ref. 2, $q_z=i/\xi$ and Eq. (A1) becomes $b=\cosh(d/\xi)$ whose solution for ξ is

$$e^{d/\xi}=b+(b^2-1)^{1/2}\ . \tag{A3}$$

The other solution $b-(b^2-1)^{1/2}$ is discarded as it corresponds to a negative value of ξ which is unphysical (ξ is the penetration depth[2]). Now ξ can be eliminated from Eqs. (A2) and (A3) and the surface-plasmon dispersion relation obtained in Ref. 2 becomes

$$\epsilon_0\sinh(qd)+\epsilon[b+(b^2-1)^{1/2}-\cosh(qd)]=0\ . \tag{A4}$$

Now it takes a lengthy and straightforward algebraic manipulation to show that Eq. (A4) is exactly the same as Eq. (38).

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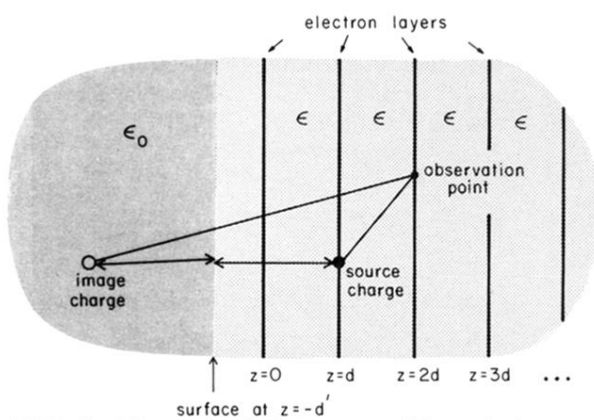

FIG. 1. The semi-infinite system of layered electron gas under consideration.