Predicted Raman Intensities for Bulk and Surface Plasmons of a Layered Electron Gas

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An exact analytic solution for the random-phase–approximation dielectric response including the surface correction is derived for a semi-infinite layered electron gas with different dielectric media on either side of the surface. The Raman intensity is calculated. The bulk plasmon line shape agrees with experiment; conditions for experimental observation of surface plasmons are described.

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Olego *et al.*[1] observed the bulk plasmon of a layered electron gas (LEG) by inelastic light scattering from GaAs-(AlGa)As heterostructures. This experiment confirmed the random-phase–approximation (RPA) prediction[2,3] of the bulk plasmon dispersion relation. By imposing standard electromagnetic boundary conditions at the layers of a semi-infinite LEG, Giuliani and Quinn[4] predicted the existence and dispersion relation of surface plasmons if the dielectric media outside and inside the semi-infinite LEG are different. This Letter gives a microscopic theory for the Raman line shapes of the bulk and surface plasmons. Our method involves the exact construction of the density-density correlation function in random-phase approximation for a semi-infinite LEG with different dielectric media on either side of the surface. Such an exact solution is possible because of mathematical simplifications arising from the layering of the electron gas, and is not available for electron response in more complicated surface geometries. From this correlation function, the Raman intensity is calculated. The theory has no free parameters. The bulk plasmon Raman line shape agrees well with that observed experimentally by Olego *et al.*[1] We find a surface plasmon which has exactly the dispersion relation predicted by Giuliani and Quinn,[4] and outline experimental conditions under which it should be observable by Raman scattering.

We use the model of Visscher and Falicov[5] for a LEG, which has delta-function-localized electron density in each plane. The electrons are free to move in the plane and the electrons in different planes interact only via the Coulomb interaction. The possibility of tunneling between two planes as well as of intersubband excitations within a plane is ignored. The planes of two-dimensional electron gas occur at $z = ld$ where l goes from 0 to ∞, and are embedded in a space of dielectric constant ϵ_0 for $z<0$ and ϵ for $z>0$.

The Coulomb potential energy of two electrons situated at planes l and m is given by[6]

$$(e^2/\epsilon)\{[(\mathbf{r}-\mathbf{r}')^2+(l-m)^2d^2]^{-1/2}+\alpha[(\mathbf{r}-\mathbf{r}')^2+(l+m)^2d^2]^{-1/2}\}, \tag{1}$$

where $\mathbf{r}$ is the two-vector (x,y). The second term, proportional to $\alpha=(\epsilon-\epsilon_0)/(\epsilon+\epsilon_0)$, gives the modification due to the image charge. The Fourier transform of (1) with respect to $\mathbf{r}-\mathbf{r}'$ is

$$V(\mathbf{q};l,m)=V_q f(\mathbf{q};l,m), \tag{2a}$$

$$V_q=2\pi e^2/\epsilon q, \tag{2b}$$

$$f(\mathbf{q};l,m)=e^{-q|l-m|d}+\alpha e^{q|l+m|d}, \tag{2c}$$

where $\mathbf{q}$ is a two-dimensional in-plane wave vector. The density-density correlation function is defined in the usual manner:

$$D(\mathbf{r},l,t;\mathbf{r}',m,t')\equiv -i\langle Tn(\mathbf{r},l,t)\,n(\mathbf{r}',m,t')\rangle, \tag{3}$$

where n and T are density and time-ordering operators. At zero degrees, $\langle\ \rangle$ denotes the ground-state expectation value. Let $D(\mathbf{q},\omega;l,m)$ be the Fourier transform and D^0 its value in the absence of Coulomb interaction. An exact expression for D^0 has been given by Stern[7] for $\gamma=0^+$ where γ is related to the electron mobility μ by $\gamma=e/m^*\mu$. We use a Mermin-corrected[8] value of D^0 for finite γ to take impurities

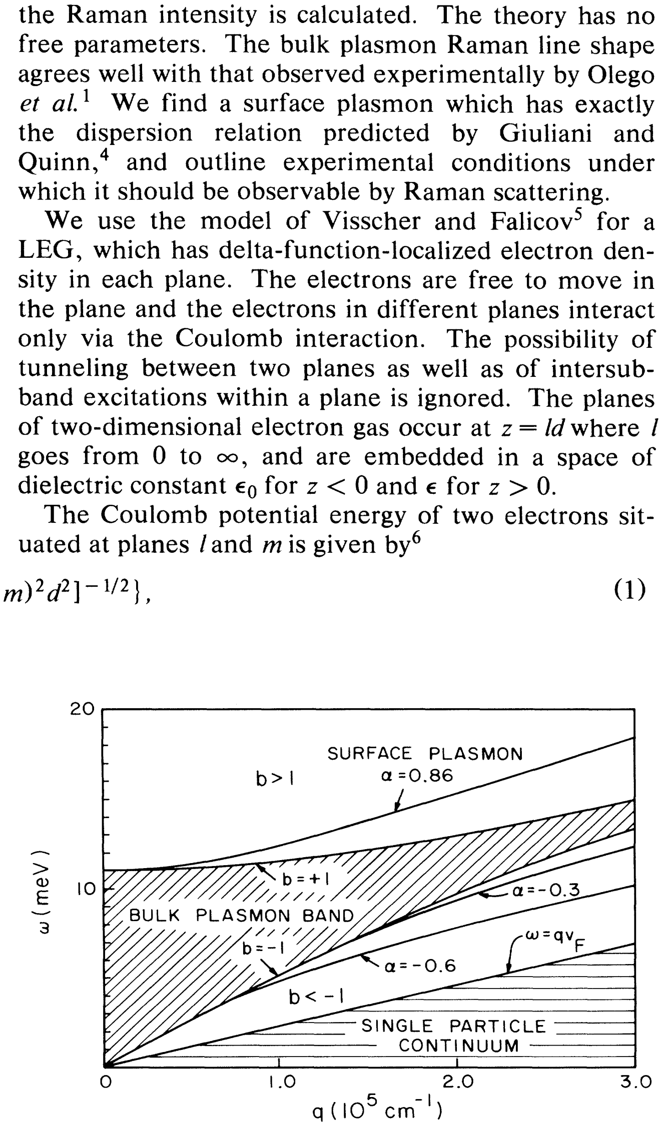

FIG. 1. Dispersion relation for the surface plasmon for several values of α. The shaded region is the bulk-plasmon band and has no surface plasmons inside it. $\alpha=0.86$ corresponds to vacuum outside and GaAs inside. $b\equiv\cosh(qd)-D^0V\sinh(qd)$.

into account.

$$D(l,m) = D^0\delta_{l,m} + D^0 V \sum_{l_1=0}^{\infty} f(l,l_1) D(l_1,m). \tag{4}$$

The dependence on $\mathbf{q}$ and ω is suppressed in this notation. To solve this equation, we make the following Fourier transformation:

$$D(l,m) = \frac{1}{N}\sum_{q_z,k_z} e^{iq_z ld} e^{-ik_z md} D(q_z,k_z), \tag{5}$$

where q_z and k_z can assume values $2\pi n/Nd$, N = number of planes, $n=0,\ldots,N-1$. (At the end, we take the limit $N\to\infty$.) We defer to a forthcoming publication[9] the long algebra that leads to the final result:

$$D(q_z,k_z) = D^b(q_z)\delta(q_z,k_z) + D^s(q_z,k_z), \tag{6}$$

$$D^b(q_z) = D^0/\epsilon(q_z), \tag{7}$$

$$D^s(q_z,k_z) = \frac{(D^0)^2 V[A - B(e^{iq_z d} + e^{-ik_z d}) + Ce^{i(q_z - k_z)d}]}{2N\epsilon(q_z)\epsilon(k_z)P(q_z)P(k_z)Q}, \tag{8}$$

$$P(q_z) = \cosh(qd) - \cos(q_z d), \tag{9}$$

$$\epsilon(q_z) = 1 - D^0 V \sinh(qd)/P(q_z), \tag{10}$$

$$A = G\sinh^2(qd) + 1 + \tfrac{1}{2}\alpha e^{2qd}, \tag{11}$$

$$B = H\sinh^2(qd) + \cosh(qd) + \tfrac{1}{2}\alpha e^{qd}, \tag{12}$$

$$C = G\sinh^2(qd) + 1 + \tfrac{1}{2}\alpha, \tag{13}$$

$$Q = 1 - G[2 + \tfrac{1}{2}\alpha(1+e^{2qd})] + (H^2 - G^2)\sinh^2(qd) + H[\alpha e^{qd} + 2\cosh(qd)], \tag{14}$$

$$G = [D^0 V/2N]\textstyle\sum_{q_z} 1/P(q_z)^2\epsilon(q_z), \tag{15a}$$

$$H = [D^0 V/2N]\textstyle\sum_{q_z} e^{iq_z d}/P(q_z)^2\epsilon(q_z). \tag{15b}$$

The first term on the right-hand side of Eq. (6) is the bulk term, and the second term D^s is the surface contribution. These have a clearer interpretation in real space. Using Eq. (5) one gets[6] $D^b(l,m)\sim\exp\{-\beta|l-m|\}$ and $D^s(l,m)\sim\exp\{-\beta|l+m|\}$, where β is a positive number. Thus the bulk contribution depends only on the distance between the layers, while the surface contribution decays exponentially as one goes away from the surface. The pole of the bulk term at $\epsilon(q_z)=0$ gives the well-known dispersion relation for the bulk plasmon of a LEG.[2,3] It can also be written as

$$b = \cos(q_z d), \tag{16}$$

$$b \equiv \cosh(qd) - D^0 V\sinh(qd). \tag{17}$$

For very pure samples, $\gamma\to 0^+$, $\mathrm{Im}D^0\to 0^-$, and $\mathrm{Im}b\to 0^+$. The range $|\mathrm{Re}b|<1$ defines the bulk-plasmon which is a continuum of energies that a bulk plasmon can assume for all possible values of q_z, while keeping $\mathbf{q}$ fixed. The boundaries of the bulk-plasmon band are $b=\pm 1$. The lower branch $b=-1$ corresponds to charge on neighboring planes oscillating out of phase; the upper branch $b=+1$ corresponds to its oscillating in phase.

The dispersion relation of the surface plasmon is given by the pole $Q(\mathbf{q},\omega)=0$ of the surface term D^s. Notice that Q is independent of the perpendicular momenta q_z and k_z. The other poles of D^s, $\epsilon(q_z)=0$ and $\epsilon(k_z)=0$, describe bulk plasmons with perpendicular momenta q_z and k_z and do not interfere with the pole $Q=0$ which lies outside the bulk-plasmon band.[4] In the limit $N\to\infty$, the integrals in Eqs. (15) can be performed and the dispersion relation becomes

$$(b^2-1)^{1/2}\sinh(qd) + \alpha e^{qd} b + \cosh(qd)(b - \alpha e^{qd}) = 1, \tag{18}$$

where the complex square root is chosen to be the branch with positive imaginary part. We have verified by a lengthy algebraic analysis that Eq. (18) agrees exactly with the Giuliani-Quinn dispersion relation. In particular, because of the factor $(b^2-1)^{1/2}$, there is no solution (i.e., no surface plasmon) inside the bulk-plasmon band. Outside this band a surface plasmon exists only for q greater than a critical wave vector q^*, given by $q^*d = -\ln|\alpha|$, which is the solution of Eq. (18) at the boundaries of the bulk-plasmon band, $b=\pm 1$. There is no Landau damping of this mode, and so the width of the surface plasmon is determined by γ alone. The dispersion relation of the surface plasmon is plotted in Fig. 1 for selected values of α.

The intensity of the Raman-scattered light as a function of its energy loss ω for a fixed value of in-plane momentum exchange $\mathbf{q}$ is proportional to[6]

$$I(\omega) = -\sum_{l,m} e^{-(l+m)d/\delta} e^{-2ikd(l-m)} \mathrm{Im} D(\mathbf{q}, \omega; l, m). \tag{19}$$

Here k and $1/2\delta$ are the real and imaginary parts of k_z, the complex z component of the wave vector of photon inside the LEG. For large values of Reϵ and small angles of incidence, k_z is constant and is equal to $\omega_0\sqrt{\epsilon}$. The different factors in Eq. (19) are intuitively understandable. We have the usual ImD which is characteristic of processes where the energy is transferred to electrons by a probe coupled to the density. The factor $\exp\{-(l+m)d/\delta\}$ takes into account the decay of the photon inside the material with decay length δ. The factor $\exp\{2ikd(l-m)\}$ is a coherence term which would generate perpendicular momentum conservation if δ were infinite, or, in other words, if there were translational invariance in the z direction.

All the sums can be performed in the expression for $I(\omega)$ and an analytic answer can be obtained. In Fig. 2 we have plotted the experimental line shape and the line shape calculated from Eq. (19) for sample 1 of the Olego *et al.* experiment. The experimental curve has been shifted by 0.2 meV along the ω axis, and all the curves have been normalized to have the same maximum height. Agreement with experiment is good. For comparison, the intensity given by a naive theory $I(\omega) = -\mathrm{Im}D^0/\epsilon(\mathbf{q}, \omega, 2k)$ is also plotted. This naive theory, unlike Eq. (13), does not take into account the broadening of perpendicular momentum caused by decay of the photon inside the material, and has all its width due to nonzero value of γ. For $\gamma \rightarrow 0^+$, this theory would give a delta function whereas Eq. (13) predicts the dotted curve in Fig. 2.

It is interesting to see what $I(\omega)$ would look like if one used only the bulk value of D in Eq. (19). A typical curve for the resulting intensity $I^b(\omega)$ is plotted in Fig. 3. Besides the peak at the bulk plasmon energy, it has two other smaller peaks at $\omega_{\min}$ and $\omega_{\max}$ which define the boundaries of the bulk-plasmon band for $q = 1\times10^5$ cm^{-1}. These peaks arise from the van Hove singularities in the one-dimensional plasmon density of states $N(\mathbf{q}, \omega)$ defined as

$$N(\mathbf{q}, \omega) = N^{-1} \sum_{q_z} \delta(\omega - \omega_p(\mathbf{q}, q_z)), \tag{20}$$

where $\omega_p(\mathbf{q}, q_z)$ is the bulk plasmon energy. When the surface contribution is added to $I^b(\omega)$, a cancellation of these peaks takes place. For $\alpha = 0$, perfect cancellation can be verified analytically. For $\alpha \neq 0$, the total intensity has no structure at $\omega_{\min}$ and $\omega_{\max}$ provided the surface plasmon is sufficiently separated from the bulk plasmon band, as is the case in Fig. 3. This shows the importance of the surface term in Eq. (19). A strict phase relation $\Delta\phi = 0$ or π for charge oscillations in neighboring layers (corresponding to $\omega_{\max}$ or $\omega_{\min}$) is needed to give a van Hove singularity but is destroyed by the presence of the surface. A similar cancellation has been found in an analysis of surface vibrational resonances by Stroscio *et al.*[10]

The surface plasmon has not yet been detected experimentally. For the usual geometry with vacuum outside, the surface plasmon exists only for $q > 1.7\times10^4$ cm^{-1} for sample 1 of Olego *et al.*[1] To get a good spectral weight at the surface plasmon, it is desirable to have high $\mathbf{q}$ and small γ, i.e., large mobili-

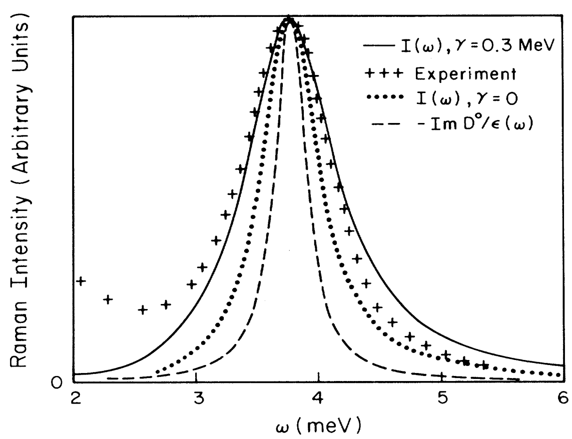

FIG. 2. Comparison between the experimental and theoretical line shapes of the bulk plasmon peak in the Raman spectrum. The experiment is from Ref. 1; $\gamma = 0.3$ corresponds to a mobility $\mu = 5\times10^4$ cm^2/V·s, used in Ref. 1.

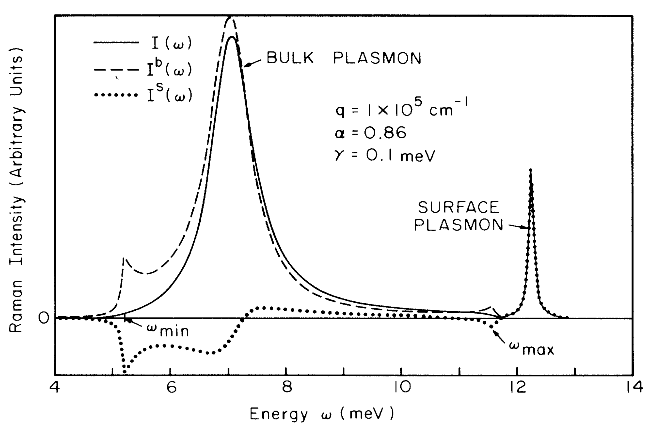

FIG. 3. Raman intensity $I(\omega)$ with its bulk part $I^b(\omega)$ and surface part $I^s(\omega)$ shown separately. Peaks in $I^b(\omega)$ at the bulk plasmon band edges, $\omega_{\min}$ and $\omega_{\max}$, are cancelled when $I^s(\omega)$ is added to it to obtain $I(\omega)$. The bulk and surface plasmons occur at 7 and 12.2 meV, respectively.

ty. In Fig. 3 we plot the Raman intensity for $q = 1.0 \times 10^5$ cm^{-1} and $\gamma = 0.1$ meV which are experimentally accessible. The bulk and surface parts of the intensity are shown separately. The surface plasmon appears at about 12.2 meV.

In a future publication[6] we plan further line-shape calculations for other experimental situations. In particular it would be interesting to search for the Giuliani-Quinn surface plasmon below the bulk plasmon continuum. This occurs if $\alpha < 0$, which is harder to achieve experimentally. Although the surface-plasmon intensity diminishes for smaller $|\alpha|$ or $q \rightarrow q^*$, we find that it does not disappear abruptly when it enters the bulk-plasmon band; a resonance remains with an intensity enhanced by the van Hove singularity at the band edge.

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