Plasmons in Layered Films

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A random-phase-approximation theory is given for the electronic collective modes of a film containing N equally spaced layers of two-dimensional electron gas. Raman line shapes are predicted. The Giuliani-Quinn surface-plasmon intensity is enhanced in transmission geometry.

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Plasmon excitations have been studied both theoretically and experimentally for two-dimensional electron gas (2DEG)[1-3] and for systems with layers of 2DEG[4-7] (called layered electron gas, or LEG; these are realized experimentally in semiconductor multilayer systems). This paper studies the spectrum of a system with a finite number of layers,[6] which is interesting both in its own right, and as a way to understand the evolution from 2DEG to bulk LEG. A third reason also emerged in this study: A finite system permits transmission experiments, and these provide a natural method for measurement of the as yet unobserved Giuliani-Quinn surface plasmon[5] of the LEG.

The model system[8] is a film of thickness L containing N 2DEG layers situated at $z=0, d, \ldots, (N-1)d$, and embedded in a space with dielectric constant ϵ for $0<z<L$ and ϵ_0 for $z<0$ and $z>L$. (This geometry will be denoted ϵ_0-ϵ-ϵ_0.) Electrons are free to move in the plane, but remain in the lowest subband, and do not tunnel to other planes. This model describes GaAs-(AlGa)As heterostructures reasonably well.[7] We ignore coupling to phonons, and assume zero temperature.

The bulk plasmon dispersion relation of an infinite LEG is given by[4]

$$\epsilon(q_z)=0, \tag{1}$$

$$\epsilon(q_z)\equiv 1-D^0 V\frac{\sinh(qd)}{\cosh(qd)-\cos(q_z d)}.$$

Here $\mathbf{q}$ and q_z are the components of the plasmon momentum parallel and perpendicular to the layers, respectively. $D^0(q,\omega)$ is the polarizability[2] of the 2DEG; $V_q=2\pi e^2/\epsilon q$ is the two-dimensional (2D) Fourier transform of the Coulomb potential. This dispersion relation has been verified experimentally by Olego *et al.*[7] The plasmon can occur only for

$$|\cosh(qd)-D^0 V\sinh(qd)|=|\cos(q_z d)|\leq 1$$

which defines the bulk plasmon band. For a semi-infinite system, when the dielectric constants inside (ϵ) and outside (ϵ_0) are different, Giuliani and Quinn[5] predicted also a surface plasmon outside the bulk plasmon band. It exists only for q greater than a critical value q^* given by $q^*d=-\ln|\alpha|$, where $\alpha=(\epsilon-\epsilon_0)/(\epsilon+\epsilon_0)$. Inside the bulk plasmon band the surface plasmon is heavily damped as it can decay into bulk plasmons because of the lack of conservation of the z component of momentum.

Now we derive the dispersion relations of collective modes of a finite film of N layers which in random-phase approximation are given by the solutions of the eigenvalue equation

$$\rho(l)=D^0\sum_{m=0}^{N-1}V(l,m)\rho(m). \tag{2}$$

The relevant solutions occur in the region where $\mathrm{Im}D^0\rightarrow 0$ and to a good approximation $D^0\sim nq^2/m\omega^2$. Thus Eq. (2) is Hermitian, and has N real eigenvalues ω^2 for each $\mathbf{q}$. In Eq. (2), ρ and V are the electron density and Coulomb interaction, Fourier transformed from (x,y,t) to $(\mathbf{q},\omega)$, and with the dependence on $\mathbf{q}$ and ω suppressed. $V(\mathbf{q};l,m)$ can be readily calculated by use of the standard electrostatic image-charge method:

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

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

$$\beta=(1-\alpha^2 e^{-2qL})^{-1}.$$

This has the correct limit as $L\rightarrow\infty$ and is also symmetric under the transformation $l,m\rightarrow L-l,L-m$. Equation

(2) can be solved analytically by means of Fourier transformation:

$$\rho(q_z)=\frac{1}{N}\sum_{l=0}^{N-1}e^{-iq_zld}\rho(l),$$

$$f(q_z,k_z)=\frac{1}{N}\sum_{l,m=0}^{N-1}e^{-iq_zld}f(l,m)e^{ik_zmd}=\frac{\sinh(qd)}{P(q_z)}\delta_{q_z,k_z}+\frac{a_1-a_2(e^{iq_zd}+e^{-ik_zd})+a_3e^{i(q_z-k_z)d}}{2NP(q_z)P(k_z)},\tag{3}$$

$$q_z,k_z=2\pi j/Nd,\quad j=1,\ldots,N,\quad P(q_z)=\cosh(qd)-\cos(q_zd),$$

$$a_1=J\beta[1+\tfrac{1}{2}\alpha Je^{2qd}+\alpha e^{qd}(\tfrac{1}{2}Je^{-qd}-\alpha e^{-qL})],\tag{4a}$$

$$a_2=J\beta[\cosh(qd)+\tfrac{1}{2}\alpha Je^{qd}+\tfrac{1}{2}\alpha Je^{qd}-\alpha^2\cosh(qd)e^{qd}e^{-qL}],\tag{4b}$$

$$a_3=J\beta[1+\tfrac{1}{2}\alpha J+\tfrac{1}{2}\alpha Je^{2qd}-\alpha^2e^{qd}e^{-qL}],\tag{4c}$$

$$J=1-e^{-Nqd}.$$

[In Eqs. (4), a_i are given for ϵ_0-ϵ-ϵ_0 geometry. For ϵ_0-ϵ-ϵ geometry, only the first two terms on the right-hand side of Eqs. (4) are retained with $\beta=1$; and for ϵ-ϵ-ϵ geometry, only the first term is retained with $\beta=1$. The limiting case $L\rightarrow\infty$ is equivalent to $e^{-qL}\rightarrow 0$ and $J,\beta\rightarrow 1$.] With this transformation, Eq. (2) becomes

$$\rho(q_z)=D^0V\textstyle\sum_{k_z}f(q_z,k_z)\rho(k_z),\tag{5}$$

which, with the help of Eq. (3), can be written as

$$\rho(q_z)=[(a_1-a_2e^{iq_zd})A_0+(a_3e^{iq_zd}-a_2)A_1]/P(q_z)\epsilon(q_z),\tag{6}$$

$$A_n=(D^0V/2N)\textstyle\sum_{k_z}e^{-ink_zd}\rho(k_z)/P(k_z).$$

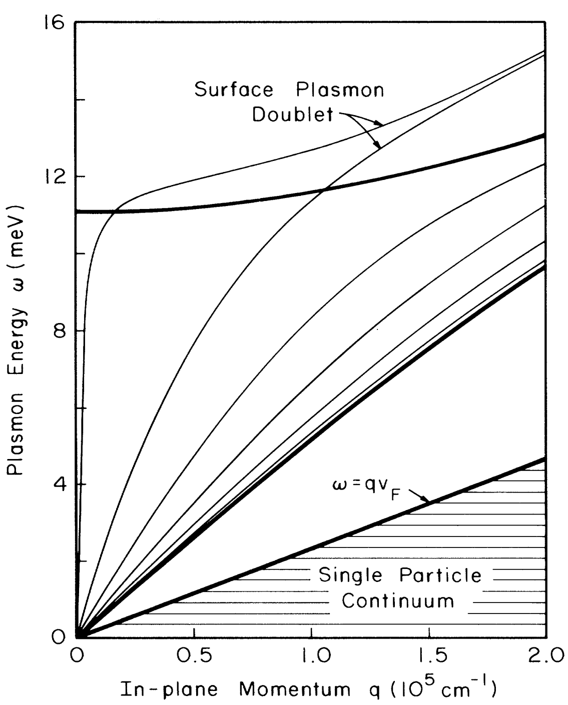

FIG. 1. Collective modes (thin lines) of a film containing six 2DEG layers. The boundaries of the bulk plasmon band (thick lines) and the single-particle continuum are also shown.

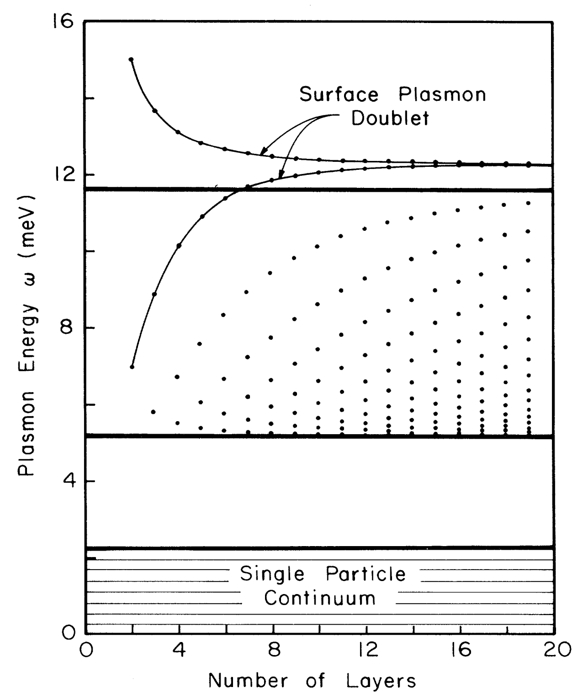

FIG. 2. Energies of collective modes plotted as a function of the number of planes for a fixed in-plane momentum $q=10^5$ cm^{-1}. The two thick horizontal lines are the bulk plasmon band edges. For $q=10^5$ cm^{-1}, the penetration depth of the surface plasmon is $\xi\sim 4d$ and the surface plasmon level starts to split at a film thickness $L\sim 3\xi$. The lines through the points are only an aid to the eye.

On multiplying Eq. (6) by $D^0V/2NP(q_z)$ or by $D^0V\exp(-iq_zd)/2NP(q_z)$ and summing over q_z we get two equations in A_0 and A_1 which have a nontrivial solution only if

$$\begin{vmatrix} g(a_1,a_2)-1 & g(a_2,a_3) \\ g(a_2,a_1) & g(a_3,a_2)-1 \end{vmatrix}=0, \tag{7}$$

$$g(x,y)=(D^0V/2N)\sum_{k_z}(x-ye^{-ik_zd})[P(k_z)^2\epsilon(k_z)]^{-1}.$$

Equation (7) gives the dispersion relation of collective modes of the film containing N 2DEG layers. Care must be taken to exclude solutions of Eq. (7) coinciding with $\epsilon(q_z)=0$ as we used division by $\epsilon(q_z)$ in obtaining Eq. (6).

Now we give a numerical example, choosing parameters of sample 1 of Ref. 7 for which $d=890$ Å, $\epsilon=13.1$, $\epsilon_0=1$, $m^*=0.07m_e$, and the electron density is 7.3×10^{11} cm^{-2}. The dispersion relations of the collective modes are plotted in Fig. 1 for $N=6$. The boundaries of the bulk plasmon band have also been plotted. For each branch ω vanishes as $q\rightarrow0$. This follows from the fact that a single 2D layer has $\omega\propto q^{1/2}$: The highest-energy branch has electrons in each plane oscillating in phase, in which case the film acts as a single 2DEG layer with electron density Nn, provided that $qL \ll 1$.

The modes above the bulk plasmon band are the Giuliani-Quinn surface plasmons.[5] For large values of qL the surface plasmons on the two surfaces do not couple and are degenerate, but for small qL the symmetric and antisymmetric combinations are split.[1] For negative α these surface modes occur below the bulk plasmon band,[5] and for $\alpha=0$ there are no surface modes as all the collective modes lie inside the bulk plasmon band. As the number of layers increases, the bulk plasmon band becomes more densely populated (see Fig. 2), becoming continuous as $N\rightarrow\infty$.

The penetration depth ξ of the surface plasmon is given by[5] $\epsilon(q,\omega_{\rm sp},q_z=i/\xi)=0$ which gives $\xi\sim4d$ when $q=10^5$ cm^{-1}. For this fixed in-plane momentum, the surface plasmon starts to split when L becomes comparable to $\sim3\xi$. This is shown in Fig. 2. The analogous splitting for a 3DEG film is given by $\omega_{\rm sp}(1\pm e^{-qL})^{1/2}$, and is both theoretically[1] and experi-

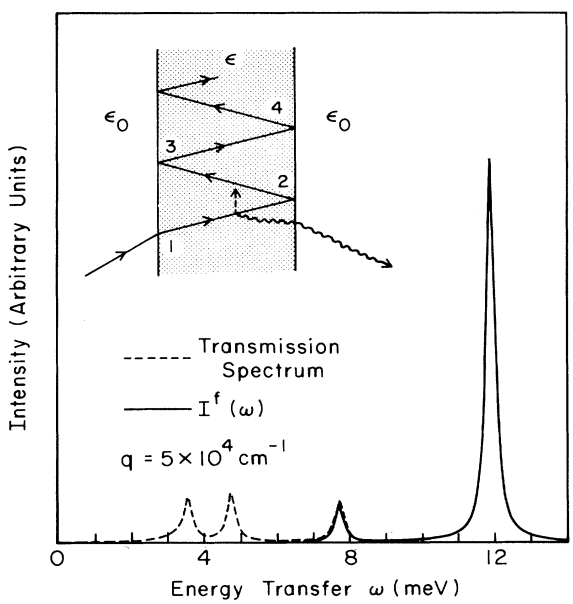

FIG. 3. Forward-scattered light $I^f(\omega)$ (solid line) for $q=5.0\times10^4$ cm^{-1} and $\gamma=0.3$ meV. The intensity is mainly concentrated at the surface plasmon energy ~12.0 meV. The total transmitted Raman intensity is different from I^f only at the dashed line. The inset shows the scattering geometry in which a photon of the incident laser beam scatters forward while exciting a plasmon.

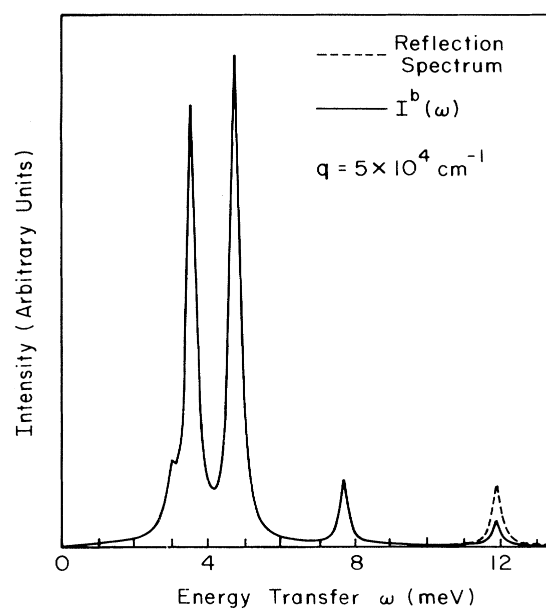

FIG. 4. The solid line is the intensity of the backscattered light, $I^b(\omega)$. The intensity is plotted on the same scale as in Fig. 3. The peaks at 3.5 and 4.5 meV appear most strongly. These are the modes that lie close to the bulk plasmon energy ~4 meV. The total reflected Raman intensity differs from I^b only at the dashed line.

mentally[9] well established.

A theory of inelastic light scattering from a semi-infinite LEG was given by Jain and Allen.[10] It was shown that one needs large in-plane momentum exchange and high-mobility samples to observe the surface plasmon with backscattered light. A thin film makes it possible to observe forward-scattered light which, as we shall see, enhances the intensity of the surface plasmons. The z momentum k_z of the photon inside the film is almost independent of the angle of incidence if $\mathrm{Re}(\epsilon)$ is large. Thus for backscattering the z momentum transfer is $2k_z$ [$\sim 5.5\times10^5$ cm^{-1} for GaAs-(AlGa) As], whereas for forward scattering it is zero.

The scattering geometry is shown in the inset of Fig. 3. First consider only the light going from point 1 to point 2 and ignore the subsequent internally reflected parts. Then the intensity of the light inelastically scattered with an exchange of in-plane momentum $\mathbf{q}$ and energy ω can be calculated as in Ref. 10. For forward scattering the resulting intensity I^f is shown in Fig. 3; for backscattering the intensity I^b is plotted in Fig. 4. [While calculating $D^0(q,\omega)$, we used $\gamma=0.3$ meV which is obtained from the mobility $\mu=5\times10^4$ cm^2/V$\cdot$s of sample 1 with the help of the relation[11] $\gamma=e/m^*\mu$.] For I^f the peak at the surface plasmon at 12 meV is the most intense, whereas for I^b the intensity is concentrated at 3.5 and 4.5 meV. The reason is that because of the approximate conservation of the z momentum, I^f and I^b have dominant contributions from collective modes with z momentum close to zero and $2k_z$, respectively. The surface plasmon travels parallel to the surface with zero z momentum and therefore appears very strongly in I^f. On the other hand, by analogy with bulk LEG, collective modes close to the bulk plasmon energy ~ 4 meV [which is a solution of Eq. (1) for $q_z=2k_z$] have z momentum close to $2k_z$, and are therefore most intense in I^b.

To take into account the effect of the internally reflected light, note that the intensity at point 2 after reflection, relative to intensity at point 1, is $\eta\sim(e^{-L/2\delta}R)^2$, where R is the reflection coefficient and δ is the decay length. For the film of Fig. 1, η is ~ 0.1. The total Raman intensity of the transmitted light is then

$$I^f+\eta I^b+\eta^2 I^f+\ldots=(I^f+\eta I^b)(1-\eta^2)^{-1}.$$

A similar formula can be written for the total Raman intensity in reflection geometry. The deviation from I^f and I^b is small, as shown in Figs. 3 and 4.

Thus the new features to be found in the Raman spectrum of layered films are the following. (1) In place of the single broad peak observed in the reflection spectrum of a semi-infinite system,[7] one would see many (two, in the case of Fig. 4) sharp peaks giving the dispersion of various collective modes. (2) To observe the surface plasmon by transmission Raman scattering one no longer needs high mobilities and large momentum exchange,[10] which should facilitate the study of the dispersion and splitting of the surface plasmon.

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