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Raman intensities for collective excitations of a layered electron gas

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An analytical expression for the Raman intensity of collective excitations in a semi-infinite system of a layered electron gas is derived in terms of the induced potential of such a system. It is rigorously shown that there are no extra peaks in the Raman intensities near the bulk-plasmon band edges. The Raman line shape is calculated and the reason that the surface plasmon cannot be observed experimentally is pointed out.

Over the last few years resonant inelastic light scattering has been successfully applied to study single-particle and collective excitations in semiconductor superlattices.[1–4] Most experiments were performed with incident and scattered photons on the vacuum side of a semi-infinite superlattice. Recently, Jain and Allen[5] analytically calculated the Raman intensities for light scattering from a semi-infinite array of two-dimensional electron gas layers with some approximations. They decomposed the Raman intensities into two parts: a bulk contribution that is the same as in the bulk system and a surface contribution. In the bulk part, as pointed out by Jain and Allen,[5,6] in addition to a peak at the bulk-plasmon energy, there are two other smaller peaks at the boundaries of the bulk-plasmon band. However, when the surface part is added to the bulk part to obtain the total intensities, the extra peaks seem to disappear according to the numerical calculation. Jain and Allen[5,6] attribute the extra peaks in the bulk part to the Van Hove singularities in the one-dimensional plasmon density of states. In our previous work,[7] we demonstrated that there are no Van Hove singularities in a semi-infinite layered electron gas (LEG) system, but this cannot explain the disappearance of the extra peaks because the density of states near the two forbidden singularities also increases sharply even in this case. Does the total intensity have any extra peaks, and if so, what are their origin? These questions cannot be answered reasonably at present. In this work, we derive an analytical expression for the Raman intensity of collective excitations in terms of the induced potential of the system, from which we can see that it is due to the specific structure of the induced potential—that there are no peaks in the Raman intensities near the bulk-plasmon band edges. On the other hand, we find that the intensity at the surface-plasmon energy is very weak and that the surface mode would be difficult to observe. This could explain why there are no surface modes observed experimentally.

The model taken in this article is as follows. The electron density has 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 planes are located 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 $z>0$. We should point out here that it is straightforward to generalize the results of the paper to the case in which the subband structure is taken into account.

Following the formulation of Jain and Allen,[6] the intensity of the Raman scattered light as a function of its energy loss w for a fixed value of momentum exchange q can be written as

$$I(w)=-\sum_{l,l'}\mathrm{Im}D(\mathbf{q},w;l,l')e^{-(l+l')d/\delta}e^{-2ikd(l-l')}, \quad (1)$$

where $D(\mathbf{q},w;l,l')$ is the density-density correlation function, k and $1/2\delta$ are the real and imaginary parts of k_z, the complex z component of the photon wave vector inside the LEG. The factor $\exp[-(l+l')d/\delta]$ takes into account the decay of the photon inside the material with decay length δ, the factor $\exp[2ikd(l-l')]$ 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.

For calculating the Raman intensity $I(w)$, one can calculate $D(\mathbf{q},w;l,l')$ and then insert it in Eq. (1), but this is not necessary as will be seen in this paper. In fact, if the induced potential in the system can be given by some method, the Raman intensity $I(w)$ can also be obtained.[8] Now we define a dielectric matrix $\epsilon(l,l')$ as follows:

$$\epsilon(l,l')=\delta_{ll'}-D^0V(l,l') \quad (2)$$

where D^0 is the value of $D(\mathbf{q},w;l,l')$ in the absence of Coulomb interaction, and

$$V(l,l')=\frac{2\pi e^2}{\epsilon q}\{\exp(-qd\,|l-l'|)+\beta\exp[-qd(l+l')]\} \quad (3)$$

with

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

The the correlation function $D(\mathbf{q},w;l,l')$ can be written in a familiar way,

$$D(l,l')=D^0\epsilon^{-1}(l,l')\,, \tag{5}$$

and the Raman intensity can also be rewritten as

$$I(w)=-\sum_{l,l'}\mathrm{Im}D^0\epsilon^{-1}(l,l')e^{-(l+l')d/\delta}e^{-2ikd(l-l')}\,. \tag{6}$$

We denote the dielectric matrix $\epsilon(l,l')$ as $\tilde{\epsilon}(l,l')$ and D^0 as $\tilde{D}^0$ when setting $\mathrm{Im}D^0\to 0$. Thus the collective excitations of the semi-infinite LEG are given by the solutions of the eigenequation,

$$\sum_{l'}[\delta_{ll'}-\tilde{D}^0(w_\alpha,q)V(l,l')]\psi_\alpha(l')=0\,, \tag{7}$$

where $\psi_\alpha(l)$, an eigenvector of $\tilde{\epsilon}(l,l')$ with an eigenindex α, is the induced potential that describes propagation of the collective excitation in the system, and w_α is the energy of the corresponding collective mode. Since $\tilde{\epsilon}(l,l')$ is Hermitian, $\psi_\alpha(l)$ obeys the orthogonal relation

$$\sum_\alpha \psi_\alpha^*(l)\psi_\alpha(l')=\delta_{ll'}\,.$$

Then, with this orthogonal relation and Eqs. (2) and (7), we have

$$\sum_{l'}\epsilon^{-1}(\mathbf{q},w;l,l')\psi_\alpha(l')=[D^0(w,q)]^{-1}\{[D^0(w,q)]^{-1}-[\tilde{D}^0(w_\alpha,q)]^{-1}\}^{-1}\psi_\alpha(l)\,, \tag{8}$$

and the Raman intensity can be written as

$$I(w)=\sum_\alpha \mathrm{Im}\{[\tilde{D}^0(w_\alpha,q)]^{-1}-[D^0(w,q)]^{-1}\}^{-1}S(q,\alpha,k)\,, \tag{9}$$

where

$$S(q,\alpha,k)=\left|\sum_l \exp[-(2ik+1/\delta)ld]\psi_\alpha(l)\right|^2. \tag{10}$$

According to Ref. 7, the induced potential of the surface plasmon is given by

$$\psi_{\alpha_s}(l)=(1-e^{-2\alpha_s d})^{1/2}e^{-\alpha_s ld}\,, \tag{11}$$

where α_s is the solution of the equation

$$(e^{-qd}+\beta e^{qd})=(1+\beta)e^{\alpha_s d}\,, \tag{12}$$

and the induced potential of the bulk plasmon can be written as

$$\psi_p(l)=\frac{1}{\sqrt{2J(q,p)}}[H(q,ip)e^{ipld}-H(q,-ip)e^{-ipld}]\,, \tag{13}$$

$$J(q,p)=[(e^{-qd}+\beta e^{qd})-(1+\beta)\cos(pd)]^2+(1+\beta)^2\sin^2(pd)\,, \tag{14}$$

$$H(q,ip)=(e^{-qd}+\beta e^{qd})-(1+\beta)e^{ipd}\,. \tag{15}$$

So that the contribution to the Raman intensity from the surface state is

$$I^s(w)=\mathrm{Im}\{[\tilde{D}^0(w_{\alpha_s},q)]^{-1}-[D^0(w,q)]^{-1}\}^{-1}\exp[(1/\delta+\alpha_s)d](1-e^{-2\alpha_s d})/2[\cosh(1/\delta+\alpha_s)d-\cos(2kd)]\,, \tag{16}$$

where w_{α_s} is the energy of the surface plasmon determined by the equation

$$\frac{2\pi e^2}{\epsilon q}\tilde{D}^0(w_{\alpha_s},q)\sinh(qd)=\cosh(qd)-\cosh(\alpha_s d)\,, \tag{17}$$

and the contribution from the bulk states is

$$I^b(w)=\frac{d}{2\pi}\int dp\,\mathrm{Im}\{[\tilde{D}^0(w_p,q)]^{-1}-[D^0(w,q)]^{-1}\}^{-1}S(q,p,k)\,, \tag{18}$$

$$S(q,p,k)=2[F(q,p,k)-G(q,p,k)]\sin^2(pd)/Y(p,k)J(q,p)\,, \tag{19}$$

$$F(q,p,k)=e^{-(2/\delta)d}[(e^{-qd}+\beta e^{qd})-(1+\beta)\cos(pd)]^2+(1+\beta)^2[1-2e^{-(1/\delta)d}\cos(pd)\cos(2kd)+e^{-(2/\delta)d}\cos^2(pd)]\,, \tag{20}$$

$$G(q,p,k)=2(1+\beta)[e^{-(1/\delta)d}\cos(2kd)-e^{-(2/\delta)d}\cos(pd)][(e^{-qd}+\beta e^{qd})-(1+\beta)\cos(pd)]\,, \tag{21}$$

$$Y(p,k)=(1+e^{-(2/\delta)d})^2-4(1+e^{-(2/\delta)d})e^{-(1/\delta)d}\cos(2kd)\cos(pd)+2e^{-(2/\delta)d}[\cos(2pd)+\cos(4kd)]\,, \tag{22}$$

where the bulk-plasmon energy w_p is determined by the equation

$$\frac{2\pi e^2}{\epsilon q}\tilde{D}^0(w_p,q)\sinh(qd)=\cosh(qd)-\cos(pd)\,. \quad (23)$$

It should be noted that $I^s(w)$ as defined here is quite different from the surface part defined by Jain and Allen.[5,6] For distinction, we call $I^s(w)$ the surface-state contribution, and for the same reason we call $I^b(w)$ the bulk-state contribution.

Apparently, the surface-state contribution can only contribute one peak at the surface-plasmon energy, while the bulk-plasmon peak and other peaks (if they exist) arise from the bulk-state contribution. For confirming the possibility of the existence of the extra peaks near the bulk-band edges, we now concentrate on the bulk-state contribution. As is well known the influence of the purity (or of r) of a sample on the line shape merely increases the peak width at some energies, and cannot change the number of peaks. So, it is sufficient to limit ourselves to the case of $r\to 0$ for studying the problems whether the extra peaks exist or not and what the origin is. To understand better the problems, it is instructive to recall how the extra peaks come from the bulk part defined by Jain and Allen.[5,6] The bulk part can be obtained from the bulk-state contribution given in (18) and (10) if we discard the surface influence, i.e., replace (13) by $\psi_p(l)=e^{ipld}$. In this case, $S(q,p,k)$ can be written as

$$S(q,p,k)=1/2e^{d/\delta}[\cosh(d/\delta)-\cos(p-2k)d]\,. \quad (24)$$

Using the limit $r\to 0$ and the small-q formula for $\tilde{D}^0 \sim nq^2/mw^2$, we have[6]

$$I^b(w)=\tfrac{1}{2}\pi\tilde{D}^0 wN(q,w)S(q,w,k)\,, \quad (25)$$

$$N(q,w)=\frac{2\Omega^2}{\pi w^3}\sinh(qd)(1-K^2)^{-1}\,, \quad (26)$$

$$K=\cosh(qd)-(\Omega^2/w^2)\sinh(qd),\ \ \Omega^2=2\pi ne^2q/m\epsilon\,. \quad (27)$$

$S(q,w,k)$ in Eq. (24) is $S(q,p,k)$ from Eq. (23) evaluated at $\cos(pd)=K$, which is the bulk-plasmon dispersion relation. Since the density of states $N(q,w)$ increases sharply near the two band edges, even though there are no Van Hove singularities in the allowed spectrum, the extra peaks also exist in the bulk part defined by Jain and Allen.[5,6] However, if we take into account the surface influence in (10), the induced potential of the bulk plasmon changes dramatically and contributes a factor $(1-K^2)$ in $S(q,p,k)$, which will cancel out the singular factor $(1-K^2)^{-1/2}$ in $N(q,w)$ exactly. Hence, owing to the special structure of the induced potential when the surface is presented, there are no extra peaks near the bulk-plasmon band edges.

In order to calculate $I(w)$, the values of the parameters are chosen to be the same as those of sample 1 of the experimental Olego, Pinczuk, Gossard, and Wiegmann.[2] They are $q=4.8\times10^4$ cm^{-1}, the electron mass $m=0.07m_e$, the static dielectric constant $\epsilon=13.1$, the electron density $n=7.3\times10^{11}$ cm^{-2}, $d=890$ Å, $2kd=4.94$, $\delta=6000$ Å, $\epsilon_0=1$, the electron mobility $\mu=5\times10^4$ cm^2/V s, and $r=e/m\mu=0.3$ meV. In Fig. 1 we give the intensity of the Raman scattered light predicted by Eqs. (16)-(23). The intensity is mainly concentrated at the bulk-plasmon energy ~3.7 meV, the peak at the surface plasmon energy ~11.5 meV is very small, the ratio of the intensities at these two energies equals about 45, so that the surface-mode peak cannot be observed within the experimental accuracy.[3]

The intensity ratio of the surface to bulk modes is enhanced as the mobility of the electrons increases, or as r decreases. This can be seen from Fig. 2 for $r=0.1$ (the

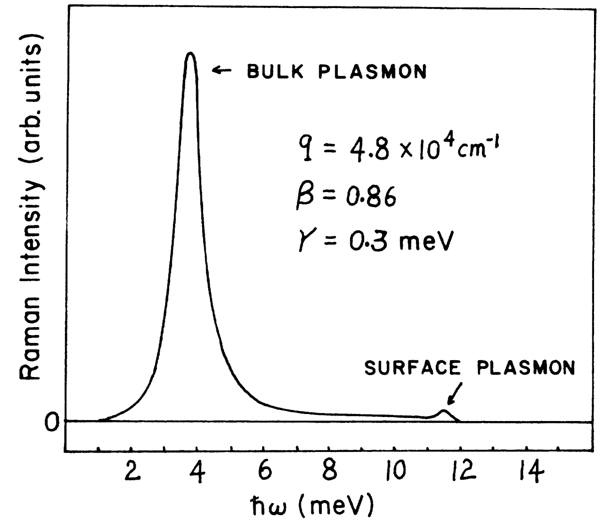

FIG. 1. Raman intensities for collective excitations of semi-infinite LEG. The bulk and surface plasmons occur at 3.7 and 11.5 meV; the intensity at the bulk-plasmon energy is much stronger than that at the surface-plasmon energy. All the parameters are the same as those of sample 1 of the experiment of Olego *et al.* (Ref. 2).

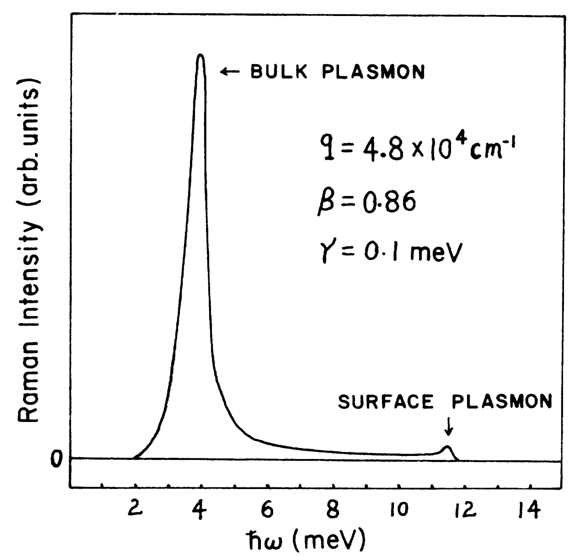

FIG. 2. Raman intensities of bulk and surface plasmons which occur at 3.7 and 11.5 meV, respectively, for $r=0.1$ (the other parameters are the same as in Fig. 1).

other parameters are the same as in Fig. 1), but the surface mode, even in this case, is also difficult to observe experimentally.

It should be noted that the intensity of the Raman-scattered light calculated in this paper is different from that obtained by Jain and Allen.[6] We think this probably due to the incorrect form of the density-density correlation function given by (36) of Ref. 6, which can be realized from the following considerations. Since (5) holds in the calculation of Jain and Allen for $D(\mathbf{q},w;l,l')$, then, according to Ref. 7, the eigenvector $D(\mathbf{q},w;l,l')$ can be written as

$$\psi_\alpha(l) = H(q,\alpha)e^{\alpha ld} - H(q,-\alpha)e^{-\alpha ld}, \quad H(q,\alpha) = (e^{-qd}+\beta e^{qd}) - (1+\beta)e^{\alpha d}, \tag{28}$$

where α cannot only be taken as pure imaginary, $\alpha = ik$ with k real, and α_s determined by $H(q,\alpha_s) = 0$, which corresponds to the bulk and surface modes, respectively, but α can also be taken as all other complex values within the condition $|\alpha| < q$. The latter is unphysical because in this case the eigenvector given by (28) cannot obey the boundary conditions, i.e., cannot remain finite when $l \to \infty$. Hence, the $D(\mathbf{q},w;l,l')$ obtained by Jain and Allen gives an incorrect form for the density-density correlation function for real systems and is probably responsible for the difference in Raman intensities calculated in Ref. 6 and in this work.

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