Intersubband collective excitations at the surface of a semiconductor superlattice

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The intersubband and intrasubband collective excitations of a semi-infinite semiconductor superlattice are investigated. Surface intersubband excitations, similar to the intrasubband surface modes predicted by Giuliani and Quinn, are found for wavelengths shorter than a critical value λ^*, which depends on the material parameters.

Charge density excitations at the surface of a semiconductor superlattice have recently attracted a great deal of attention.[1-3] A new type of surface polariton, with a remarkable property of being free of Landau damping, has been predicted by Giuliani and Quinn for a surface parallel to the superlattice layers. In that study only intrasubband excitations were considered, and the superlattice could be represented as a semi-infinite periodic array of two-dimensional electron-gas layers. On the other hand, a theory of collective modes of an infinite semiconductor superlattice including miniband structure has been formulated in detail by Tselis and Quinn.[4] Rich structure associated with the intrasubband and intersubband collective modes, including depolarization shifts and excitonic effects, has been predicted using self-consistent linear response theory.

In this paper we extend this approach to the case of a semi-infinite superlattice. We employ a simple model of the electronic structure and calculate the density response of the system to an external perturbation.[5] In a simple way both intrasubband and intersubband surface modes are obtained.

The model system corresponding to the semiconductor superlattice under consideration is shown in Fig. 1. A semi-infinite array of quantum wells of thickness L, whose centers are separated by distance a from each other, occupies a half-space $z > -\delta$, of background dielectric constant ϵ. An insulator with dielectric constant ϵ_0 occupies the space $z < -\delta$. The single-particle electronic states are assumed to be of the form

$$|n,\mathbf{k},l\rangle = e^{i\mathbf{k}\cdot\mathbf{r}}\xi_n(z-la)\ . \tag{1}$$

Here n refers to the subband index, $\mathbf{k}$ is momentum in the plane perpendicular to the z axis, and the integer l denotes the quantum well centered at $z=la$. We make the assumption that the wave function $\xi_n(z)$ on different layers do not overlap, so that the minibands are flat. The energy eigenvalues are

$$\epsilon_{nkl} = \epsilon_n + \frac{\hbar^2 k^2}{2m}\ , \tag{2}$$

where ϵ_n is the energy at the bottom of each subband, and m is an effective mass.

An external potential of the form

$$v^{\rm ext}(\mathbf{r},z,t) = v^{\rm ext}(\mathbf{q},\omega,z)e^{i\omega t}e^{-i\mathbf{q}\cdot\mathbf{r}} \tag{3}$$

induces a change in the electron density, which in turn generates an induced Hartree potential δv^H. The total perturbing potential $v = v^{\rm ext} + \delta v^H$ is the sum of the external and the Hartree potentials. The density charge $\delta n(q,\omega,z)$ induced by the total potential $v(q,\omega,z)$ is given by

$$\delta n(\mathbf{q},\omega,z) = \sum_{\substack{n,n',\\ l}} \Pi_{nn'}(\mathbf{q},\omega)\langle n \mid v_l(\mathbf{q},\omega) \mid n'\rangle \times \xi_n(z-la)\xi_{n'}(z-la)\ , \tag{4}$$

where

$$\langle n \mid v_l(\mathbf{q},\omega) \mid n'\rangle = \int dz\, \xi_n(z-la)v(\mathbf{q},\omega,z)\xi_{n'}(z-la) \tag{5}$$

and polarizability $\Pi_{nn'}$ is given by

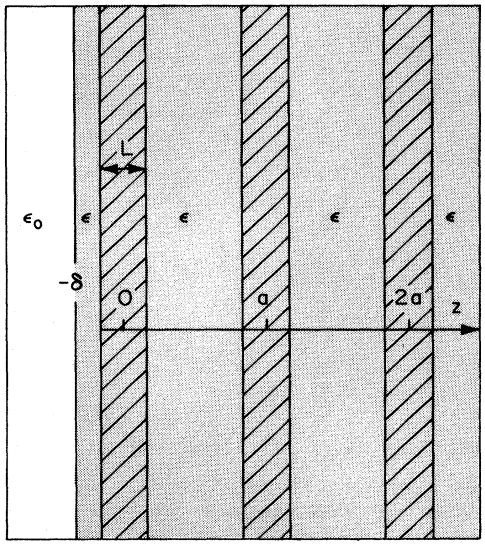

FIG. 1. A semi-infinite array of quantum wells (cross-hatched area) of thickness L, whose centers are separated by distance a, embedded in a semiconductor with dielectric constant ϵ (shaded area). An insulator with dielectric constant ϵ_0 occupies the space to the left from the interface at $z=-\delta$.

$$\Pi_{nn'}(\mathbf{q},\omega)=2\sum_{\mathbf{k}}\frac{f(\epsilon_{n',\mathbf{k}+\mathbf{q}})-f(\epsilon_{n,\mathbf{k}})}{\epsilon_{n',\mathbf{k}+\mathbf{q}}-\epsilon_{n,\mathbf{k}}-\hbar\omega}\ . \tag{6}$$

Here we have neglected the effect of the induced exchange and correlation potentials, but inclusion of this effect is straightforward.[4] The induced density $\delta n(q,\omega,z)$ is related to the induced Hartree potential δv^H by Poisson's equation; this can be expressed in terms of the Green's function $G(z,z')$ as

$$\delta v^H(\mathbf{q},\omega,z)=\int dz'\,G(z,z')\delta n(\mathbf{q},\omega,z')\ . \tag{7}$$

The Green's function $G(z,z')$ for the geometry corresponding to Fig. 1 is given by[6] (for $z,z'>-\delta$)

$$G(z,z')=\frac{2\pi e^2}{\epsilon q}(e^{-q|z-z'|}+\alpha e^{-q(z+z')})\ , \tag{8}$$

where $\alpha=[(\epsilon-\epsilon_0)/(\epsilon+\epsilon_0)]e^{-2\delta q}$. Taking matrix elements of Eq. (7) gives

$$\langle n|\delta v_l^H|n'\rangle=\sum_{\substack{m,m',\\ l'}}\Pi_{mm'}V_{nn'}^{mm'}(l,l')\langle m|v_{l'}|m'\rangle\ , \tag{9}$$

where the symbol $V_{nn'}^{mm'}(l,l')$ is given by the equation

$$V_{nn'}^{mm'}(l,l')=\int dz\int dz'\,\xi_m(z-la)\xi_{m'}(z-la)\times G(z,z')\xi_n(z'-l'a)\xi_{n'}(z-l'a)\ . \tag{10}$$

Here we have omitted the dependence on $\mathbf{q},\omega$ for simplicity.

Making the approximation that only the lowest subband is occupied and using the fact that $v=v^{\text{ext}}+v^H$ we can write the actual response of the system as

$$\langle m|v_l|0\rangle=\langle m|v_l^{\text{ext}}|0\rangle+\sum_{n,l}\chi_{m0}V_{nm}(l,l')\langle n|v_{l'}|0\rangle\ , \tag{11}$$

where $\chi_{n0}=\Pi_{m0}+\Pi_{0m}$, $V_{nm}(l,l')\equiv V_{no}^{m0}(l,l')$, and $l,l'=0,1,2,\ldots$. This is exactly the result of Tselis and Quinn[4] except that because of broken translational invariance in the z direction, matrix elements $V_{nm}(l,l')$ depend upon both layer indices l and l' and not simply upon the difference $l-l'$. We can still define a dielectric matrix $\epsilon(l,l',n,m)$ as

$$\epsilon(l,l',n,m)=\delta_{l,l'}\delta_{nm}-\chi_{m0}V_{mn}(l,l')\ . \tag{12}$$

Equation (11) can be rewritten as

$$\sum_{n,l'}\epsilon(l,l',n,m)\langle n|v_{l'}|0\rangle=\langle m|v_l^{\text{ext}}|0\rangle\ . \tag{13}$$

Collective excitations are given by the zeros of $\epsilon(l,l',n,m)$. This can be seen by setting $v^{\text{ext}}=0$ while requiring the total potential v to remain finite. In order to solve Eq. (13) and find surface modes, we assume that mixing between different subbands is negligible. This is equivalent to taking the dielectric function $\epsilon(l,l',n,m)$ to be diagonal in subband indices [i.e., we take $V_{n,m}(l,l')=\delta_{mn}V_{mn}(l,l')$]. In this approximation surface modes are given by the solutions to Eq. (11) with $v^{\text{ext}}=0$, i.e.,

$$\langle m|v_l|0\rangle=\sum_{l'=0}^{\infty}\chi_{m0}V_{mm}(l,l')\langle m|v_{l'}|0\rangle\ . \tag{14}$$

The function $v_{mn}(l,l')$ can be written as

$$V_{mm}(l,l')=\frac{2\pi e^2}{\epsilon q}\int\int dz\,dz'\xi_m(z)\xi_0(z)\times(e^{-q|z-z'+(l-l')a|}+\alpha e^{-q[z+z'+(l+l')a]})\times\xi_m(z')\xi_0(z')\ , \tag{15}$$

where $l,l'=0,1,2,\ldots$. In order to obtain solutions of the equation (14) localized at the surface, we assume that $\langle m|v_l|0\rangle=e^{-\beta_m al}\langle m|v|0\rangle$. Here β_m^{-1} is a decay length of the excitation away from the interface. We require that $\text{Re}\beta_m>0$ but allow for the possibility of an imaginary part of β_m equal to $\pm i\pi/a$. If $\text{Im}\beta_m=0$ the oscillations on subsequent layers are in phase while for $\text{Im}\beta_m=\pi/a$ they are out of phase. Using this ansatz Eq. (14) can be written as

$$1=\sum_{l'=0}^{\infty}\chi_{m0}V_{mm}(l,l')\exp[-\beta_m\alpha(l-l')]\ . \tag{16}$$

Making use of Eq. (15), the sum over layer indices l' can be done explicitly. We then separate the terms that depend upon layer index l from the terms which do not. Since the solution to Eq. (16) cannot depend upon the layer index l we set the coefficient of the term containing index l to zero. Hence we arrive at the set of two equations which uniquely determines the dispersion of the surface modes. They are given by

$$1=\chi_{m0}(q,\omega)[V_{-m}(q)-G_{-m}(q)+G_{-m}(q)S(q,\beta_m)]\ , \tag{17a}$$

$$\alpha\frac{G_{+m}(q)}{G_{-m}(q)}=-\frac{e^{\beta_m a}-e^{-qa}}{e^{\beta_m a}-e^{+qa}}\ . \tag{17b}$$

Here the matrix elements $V_{\pm m}(q)$ are defined as

$$V_{\pm m}(q)=\frac{2\pi e^2}{\epsilon q}\int dz\int dz'\,\xi_m(z)\xi_0(z)e^{-q|z\pm z'|}\xi_m(z')\xi_0(z')\ , \tag{18}$$

and matrix elements $G_{\pm m}(q)$ are defined by a similar expression except for the replacement of $|z\pm z'|$ by $z\pm z'$. The function $S(q,\beta)$ is the well-known structure factor

$$S(q,\beta)=\sinh(qa)/[\cosh(qa)-\cosh(\beta a)]\ . \tag{19}$$

For intrasubband excitations ($m=0$) and for infinitely thin quantum wells Eqs. (17) reduce exactly to the pair of equations found by Giuliani and Quinn.[1] On the other hand, Eq. (17a) with $\beta_m\to -ik_z$ gives dispersion of bulk intersubband collective modes obtained by Tselis and Quinn.[4]

As an *illustration* of the formalism introduced here we determine the dispersion of intrasubband and intersubband modes using wave functions and single-particle energies corresponding to quantum wells with an infinite po-

tential barrier. In this case the matrix elements can be evaluated analytically. They are given in the Appendix. Using these approximate matrix elements we obtain a very simple form for Eq. (17b):

$$\frac{\epsilon-\epsilon_0}{\epsilon+\epsilon_0}e^{-2\delta q}(-1)^{m+1}=\frac{e^{\beta_m a}-e^{-qa}}{e^{\beta_m a}-e^{+qa}} . \tag{20}$$

Setting $\beta_m=0$ in Eq. (20) yields the critical wave vector q^* at which the surface plasmon intersects the bulk plasmon spectrum:

$$q^*=\ln|(\epsilon+\epsilon_0)/(\epsilon-\epsilon_0)|(a-2\delta)^{-1} . \tag{21}$$

The surface plasmon can exist for $q>q^*$. Clearly, q^* depends quite critically on the ratio of dielectric constants, the well thickness L and the presence of the overlayer, indicated by the factor 2δ. Note that 2δ is at least equal to the well thickness L.

To illustrate the effect of the finite size of the quantum wells on the dispersion of intrasubband plasmons ($m=0$), results for the frequencies of bulk and surface plasmons are shown in Figs. 2 and 3. The following parameters have been chosen in the numerical calculations:[7] $\epsilon=13.1$, $\epsilon_0=1.0$, $n_s=7.3\times10^{11}$ cm^{-2}, $m=0.07m_e$, and $a=890$ Å. Two different values of L have been used: $L=260$ Å (Fig. 2) and $L=130$ Å (Fig. 3) ($2\delta=L$ in both cases). In addition, the long-wavelength limit for polarizability of two-dimensional electron gas $\chi_{00}=n_sq^2/m\omega^2$ has been used. Note that the effect of increasing well thickness is to increase the critical wave vector for the surface plasmon q^* in accordance with Eq. (21). Another effect is to "push" the surface plasmon closer to the bulk spectrum. Since the Raman spectrum of bulk plasmons is broadened due to finite mobility,[3] the ability to resolve the surface plasmon from the bulk spectrum is decreased by increased well thickness.

We now turn to the intersubband excitations. In Fig. 4 bulk and surface modes are shown for the same parameters as in Fig. 2, for the mode corresponding to $m=1$. The long-wavelength limit of the polarizability

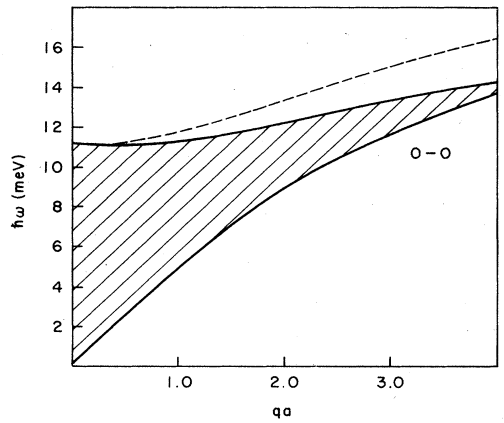

FIG. 2. Dispersion relation ω vs q for the bulk (shaded area) and surface (dashed line) intrasubband modes ($m=0$). The values of parameters are as follows: $\epsilon=13.1$, $\epsilon_0=1.0$, $n_s=7.3\times10^{11}$ cm^{-2}, $a=890$ Å, $L=260$ Å, $m=0.07m_e$, and $2\delta=L$.

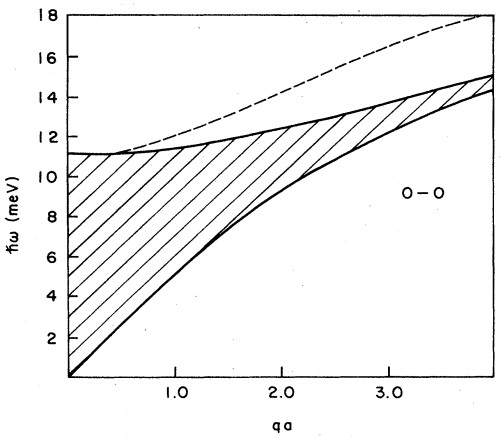

FIG. 3. Dispersion relation ω vs q for an intrasubband mode ($m=0$) for a system of thinner quantum wells; $L=130$ Å. All other parameters are the same as in Fig. 2.

$$\chi_{10}=2n_s\epsilon_{01}/[(\hbar\omega)^2-\epsilon_{01}^2]$$

has been used. Here $\epsilon_{01}=23.84$ meV so that a large depolarization shift is observed. Note the softening of the surface mode, characteristic of intersubband excitations.[4] We have also obtained surface modes for higher subbands which show qualitatively similar behavior.

Since typical semiconductor superlattices, such as GaAs-$Al_xGa_{1-x}As$, are polar materials, the background dielectric constant is a function of frequency:

$$\epsilon(\omega)=\epsilon_\infty(\omega^2-\omega_L^2)/(\omega^2-\omega_T^2) ,$$

where ϵ_∞ is a high-frequency dielectric constant, and ω_L and ω_T are the longitudinal and transverse optical phonon frequencies, respectively. By taking into account the frequency dependence of the dielectric function $\epsilon(\omega)$ we find coupled-bulk-and-surface-intersubband—optical-phonon modes as shown in Fig. 5. In the numerical calculations the following values have been taken—$\epsilon_\infty=10.9$, $\epsilon(0)=13.1$, and $\hbar\omega_L=36.2$ meV; the remaining parameters are the same as in Fig. 4. In this case we find two surface modes.

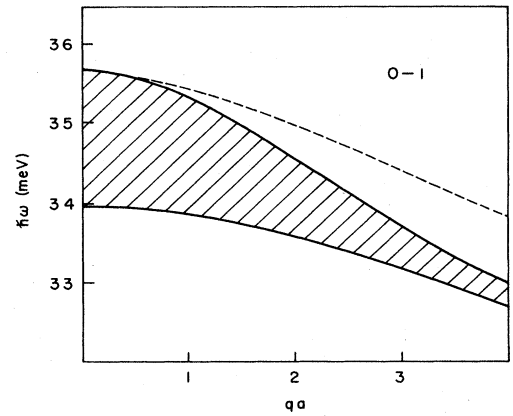

FIG. 4. Dispersion relation ω vs q for the bulk (shaded area) and surface (dashed line) intersubband excitations ($m=1$). All parameters are the same as in Fig. 2.

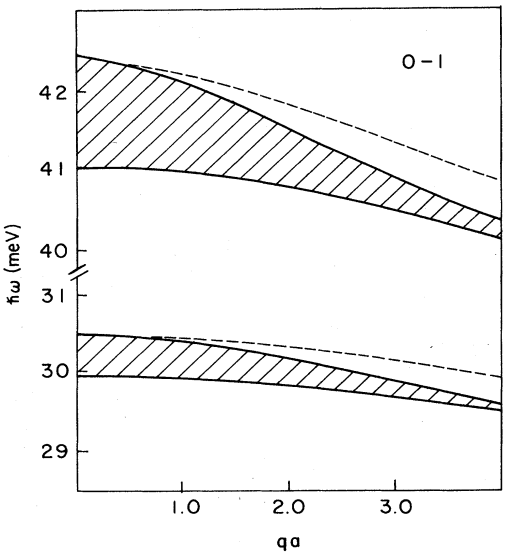

FIG. 5. Dispersion relation ω vs q for coupled-intersubband ($m=1$)—optical-phonon excitations. Shaded area denotes bulk modes and dashed lines correspond to two surface modes. All parameters here are the same except for the semiconductor dielectric constant $\epsilon(\omega)$. See text.

In summary, we have extended the Tselis-Quinn theory of collective excitations in semiconductors superlattices to the case of semi-infinite superlattices. Conditions for surface intrasubband and intersubband charge density excitations have been outlined and dispersion of these modes is predicted. We hope that this work will stimulate experiments directed toward observation of these modes either through Raman scattering or electron energy loss spectroscopy. Theoretical analysis of the former is the subject of a forthcoming publication.

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APPENDIX

Matrix elements $V_{\pm m}(q), G_{\pm m}(q)$ are evaluated using wave functions corresponding to a quantum well with an infinite potential barrier, whose center is at $z=0$ and thickness is L. The wave functions are $\xi_n(z)=(2/L)^{1/2}\sin[(n+1)\pi(z/L+\frac{1}{2})]$. From the definitions [Eq. (18)] we find

$$V_{-n}(q)=\frac{2\pi e^2}{\epsilon q}\left[qL\left[\frac{1}{x}+\frac{1+\delta_{n0}}{y}\right]-2(qL)^2\left[\frac{\pi^2[(n+2)^2-n^2]}{xy}\right]^2\times[1-(-1)^n e^{-qL}]\right] \tag{A1}$$

and

$$G_{+n}(q)=\frac{2\pi e^2}{\epsilon q}f_n(q)f_n(q)e^{qL}\,,$$
$$G_{-n}(q)=\frac{2\pi e^2}{\epsilon q}f_n(q)f_n(-q)\,. \tag{A2}$$

There $f_n(q)$ is given by

$$f_n(q)=qL\frac{\pi^2[(n+2)^2-n^2]}{xy}[1-(-1)^n e^{-qL}] \tag{A3}$$

and $x=[(qL)^2+\pi^2(n+2)^2]$, $y=[(qL)^2+\pi^2 n^2]$.

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