Inelastic light scattering from collective modes in a layered superconductor with Cooper-pair tunneling

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(Received 17 May 1995)

In a superconducting bilayer with Cooper-pair tunneling ($T_J \neq 0$) between the layers, it has been recently predicted that there should exist in-phase and out-of-phase phase and amplitude fluctuations of the order parameters of the two layers. As a way of observing these phase modes, we calculate the inelastic light scattering from a semi-infinite superconducting superlattice with a bilayer basis. For simplicity, we ignore surface modes. We show that the bulk phase modes show up as resonances in the isotropic Raman light scattering intensity at finite momentum transfer parallel to the surface. Our specific calculations are for s-wave superconductors but similar results are expected for d-wave pairing. The size of the energy gap of the out-of-phase modes is strongly dependent on the ratio of T_J and the intralayer pairing interaction g, as well as particle-hole damping.

I. INTRODUCTION

Recently, a model with strong interlayer Cooper-pair tunneling has been proposed by Chakravarty, Anderson, and co-workers[1] for the copper oxide superconductors. Within this model, Wu and Griffin[2,3] have predicted the existence of a phase and amplitude modes associated with out-of-phase fluctuations of the Cooper-pair condensates in the two layers of a bilayer, in addition to the in-phase Anderson-Bogoliubov (AB) phase and Littlewood-Varma amplitude modes. The bilayer phase modes are of most experimental interest since they couple into the density fluctuation response functions, which can be studied by inelastic light scattering. This is the subject of the present paper.

In nonsuperconducting superlattices, only the intra- and interlayer Coulomb interactions are considered, usually within the usual random-phase approximation (RPA). In this connection, Jain and Allen[4] as well as Giuliani, Hawrylak, and Quinn[5] have given a detailed discussion of inelastic light scattering in a layered electron gas (LEG) superlattice without a basis. Santoro and Giuliani[6] calculated the Raman scattering for a single normal bilayer. Griffin and Pindor[7] discussed the layer density response functions in a LEG superlattice with a bilayer basis (two sheets per unit cell). Generalizing the formalism of Jain and Allen, in the present paper we calculate the Raman light scattering from a superlattice with two superconducting layers per unit cell. This model should be relevant for Bi-Sr-Ca-Cu-O and Tl-Ba-Ca-Cu-O oxide superconductors. The superlattice period ($\equiv c$) is $\simeq 12$ Å, while the bilayer spacing ($\equiv d$) is $\simeq 3$ Å. The pairing interaction responsible for superconductivity is assumed to be confined to the two-dimensional (2D) layers due to the highly 2D character of the electronic energy bands. Since $c \ll d$, we only allow the interlayer Cooper-pair tunneling between the layers within the same unit cell and ignore the Cooper-pair tunneling between two consecutive layers in different cells.

Our main concern are the resonances in the Raman scattering which are associated with the in-phase and out-of-phase phase modes. One finds[4] that the light scattering from a surface involves a weighted sum of the layer-dependent density response functions, and as a result, it picks up the two kinds of phase modes. It has been shown that the magnitude of the energy gap of the out-of-phase Leggett mode[8] depends critically on the ratio of T_J and g,[2] and it can be above or below the pair-breaking energy 2Δ. Experimental observation of these out-of-phase phase modes and the position of the energy gaps would give unambiguous evidence for the existence of Cooper-pair tunneling as postulated in Ref. 1.

In Sec. II, we derive the layer-dependent density response functions. In Sec. III, we use these results to calculate the inelastic light scattering for a semi-infinite superconducting superlattice with a basis of bilayers, restricting the interlayer Cooper tunneling to layers within the same unit cell. In Sec. IV, we summarize our main results. Since this paper is largely an extension of the work in Refs. 2, 4, we only give a brief sketch of the formalism in Secs. II and III. For simplicity, we work with an s-wave pairing interaction and an isotropic interlayer Cooper-pair tunneling strength, and limit our analysis to $T = 0$. Similar calculations can be done for d-wave pairing using the response functions given in Refs. 3, 9. The main difference is that the out-of-phase mode is strongly damped because there is no pair-breaking gap.

II. INTRA- AND INTERLAYER RESPONSE FUNCTIONS

Using the same approach as in Refs. 4, 7 to deal with a superconducting superlattice with two sheets per unit cell, the RPA-type equation of the density-density correlation function for electronic densities in layers (l, i) and (l', j) is given by

$$\chi_{ij}(\mathbf{q}_{\|},\omega,l,l') = \bar{\chi}_{ij}(\mathbf{q}_{\|},\omega,l,l') + \sum_{\substack{l_1,l_2\\ i_1,i_2}} \bar{\chi}_{ii_1}(\mathbf{q}_{\|},\omega,l,l_1) v_{i_1 i_2}(\mathbf{q}_{\|},l_1-l_2)\chi_{i_2 j}(\mathbf{q}_{\|},\omega,l_2,l'), \quad (1)$$

where l, l' run from 0 to $N-1$ (the total number of the unit cells N is infinite for semi-infinite superlattices) and $i,j = 1,2$ label the two sheets in the bilayer within each unit cell. The irreducible two-particle Green's functions $\bar{\chi}_{ij}(l,l')$ are calculated in the absence of any Coulomb interactions. However, in a superconductor, they do include the vertex corrections (ladder diagrams) due to the intralayer pairing interaction g and interlayer Cooper-pair coupling T_J. These have been recently discussed in detail in Refs. 2, 3, 9. The Coulomb interaction in (1) is given by

$$v_{ij}(\mathbf{q}_{\|}, l-l') = v_{2D} e^{-q_{\|}|Z_{li}-Z_{l'j}|}, \quad (2)$$

where $v_{2D} = 2\pi e^2/q_{\|}\epsilon_0$ is the Coulomb interaction for a 2D isolated layer. For the lth unit cell, we define $Z_{l,1} \equiv lc$, $Z_{l,2} \equiv lc + d$; i.e., the unit cell spacing is c and the spacing between sheets in a unit cell is d. The fact that the Coulomb interaction (2) only depends on the distance between the two sheets is a result of our assuming that the background dielectric constant ϵ_0 is the same in the superlattice ($z < 0$) as in the medium above it ($z > 0$). Treating ϵ_0 different above and below the interface, Jain and Allen[4] have shown that the Coulomb interaction leads to the appearance of surface plasmons in the case of normal semi-infinite superlattices. For simplicity, we ignore such surface modes in the present paper although they might be of interest in a future study.

Since the pairing interaction is assumed to only exist in the same layer and the interlayer Cooper-pair coupling only occurs within the same unit cell, the correlation functions $\bar{\chi}_{ij}$ will be l independent and are given by those of an isolated bilayer. This means $\bar{\chi}_{ij}(\mathbf{q}_{\|},\omega,l,l') = \bar{\chi}_{ij}(\mathbf{q}_{\|},\omega)\delta_{l,l'}$ and we can reduce (1) to

$$\chi_{ij}(\mathbf{q}_{\|},\omega,l,l') = \bar{\chi}_{ij}(\mathbf{q}_{\|},\omega)\delta_{l,l'} + \sum_{\substack{l_2\\ i_1,i_2}} \bar{\chi}_{ii_1}(\mathbf{q}_{\|},\omega) v_{i_1 i_2}(\mathbf{q}_{\|},l-l_2)\chi_{i_2 j}(\mathbf{q}_{\|},\omega,l_2,l'). \quad (3)$$

The bilayer response functions of a "neutral" superconductor $\bar{\chi}_{ij}(\mathbf{q}_{\|},\omega)$ can be assumed to be known from earlier work.[2,3] One can use an extension of the Fourier series method developed in Refs. 4, 5 to solve the system of coupled response functions in (3). One finds that the semi-infinite superlattice response functions $\chi_{ij}(\mathbf{q}_{\|},\omega;k_z,k_{z'})$ have a bulk term and a surface term.[10] Since we ignore the surface contribution in the present paper, we are left with the bulk contribution which is identical to that of an infinite superlattice, for which $\chi(l,l') = \chi(l-l')$ and l,l' range from $-\infty$ to ∞. Taking the Fourier transform of (3) appropriate to such an infinite superlattice,

$$A(q_z) = \sum_{l=-\infty}^{\infty} e^{iq_z lc} A(l)$$

$$A(l) = \frac{1}{N}\sum_{q_z} e^{-iq_z lc} A(q_z) = \frac{c}{2\pi}\int_{-\pi/c}^{\pi/c} dq_z e^{-iq_z lc} A(q_z) \ , \quad (4)$$

we obtain immediately

$$\chi_{ij}(\mathbf{q},\omega) = \bar{\chi}_{ij}(\mathbf{q}_{\|},\omega) + \sum_{i_1,i_2} \bar{\chi}_{ii_1}(\mathbf{q}_{\|},\omega) v_{i_1 i_2}(\mathbf{q})\chi_{i_2 j}(\mathbf{q},\omega), \quad (5)$$

where $\chi_{ij}(\mathbf{q}_{\|},\omega;q_z) \equiv \chi_{ij}(\mathbf{q},\omega)$. The Coulomb interactions in (5) involve layer sums which can be carried out explicitly to give (see Ref. 7)

$$v_{11}(\mathbf{q}) = v_{22}(\mathbf{q}) \equiv \tilde{v}(\mathbf{q}) = v_{2D}\left[\frac{\sinh q_{\|}c}{\cosh q_{\|}c - \cos q_z c}\right] \ ,$$

$$v_{12}(\mathbf{q}) = [v_{21}(\mathbf{q})]^* \equiv \bar{v}(\mathbf{q}) = v_{2D}\left[\frac{\sinh q_{\|}(c-d) + e^{-iq_z c}\sinh q_{\|}d}{\cosh q_{\|}c - \cos q_z c}\right] . \quad (6)$$

As one might have expected, only the *interlayer* Coulomb interactions (v_{12} and v_{21}) depend on the spacing d of the bilayers.

The derivation of the bilayer response functions $\bar{\chi}_{ij}(\mathbf{q},\omega)$ in (5) is a lengthy calculation, involving treating the phase and amplitude fluctuations of the order parameters (Cooper pairs) on the two layers as well as the charge-density fluctuations. We only quote the final results of Refs. 2, 3:

$$\bar{\chi}_{11}(\mathbf{q}_{\|},\omega) = \bar{\chi}_{22}(\mathbf{q}_{\|},\omega) = A_0 - \frac{4(g+T_J)c_0^2}{\bar{D}_+} + \frac{4T_J c_0^2}{\bar{D}_+\bar{D}_-},$$

$$\bar{\chi}_{12}(\mathbf{q}_{\|},\omega) = \bar{\chi}_{21}(\mathbf{q}_{\|},\omega) = -\frac{4T_J c_0^2}{\bar{D}_+\bar{D}_-}, \quad (7)$$

where we ignore the short-range interaction g_z in the particle-hole channel. The denominators are given by

$$\bar{D}_{\pm}(\mathbf{q}_{\|},\omega) \equiv 1 - (g \pm T_J)\, B_0. \quad (8)$$

The 2D noninteracting (in the sense that there are no vertex corrections) correlation functions in (7) and (8) are defined by[2,11]

$$A_0(\mathbf{q}_\parallel,\omega+i\gamma) = \int \frac{d\mathbf{p}_\parallel}{(2\pi)^2}\frac{E+E'}{2EE'}\frac{EE'-\epsilon\epsilon'+\Delta_{\mathbf{p}_\parallel}\Delta_{\mathbf{p}_\parallel+\mathbf{q}_\parallel}}{(\omega+i\gamma)^2-(E+E')^2},$$

$$B_0(\mathbf{q}_\parallel,\omega+i\gamma) = \int \frac{d\mathbf{p}_\parallel}{(2\pi)^2}\frac{E+E'}{2EE'}\frac{-(EE'+\epsilon\epsilon'+\Delta_{\mathbf{p}_\parallel}\Delta_{\mathbf{p}_\parallel+\mathbf{q}_\parallel})}{(\omega+i\gamma)^2-(E+E')^2},$$

$$c_0(\mathbf{q}_\parallel,\omega+i\gamma) = \int \frac{d\mathbf{p}_\parallel}{(2\pi)^2}\frac{-(\omega+i\gamma)}{2E}\frac{\Delta_{\mathbf{p}_\parallel}}{(\omega+i\gamma)^2-(E+E')^2}, \tag{9}$$

where the BCS quasiparticle spectrum is $E_{\mathbf{p}_\parallel} = \sqrt{\epsilon^2_{\mathbf{p}_\parallel}+|\Delta_{\mathbf{p}_\parallel}|^2}$, with $\epsilon_{\mathbf{p}_\parallel} = p^2_\parallel/2m-\mu$ and $E\equiv E_{\mathbf{p}_\parallel}$, $E'\equiv E_{\mathbf{p}_\parallel+\mathbf{q}_\parallel}$, and $\epsilon\equiv\epsilon_{\mathbf{p}_\parallel}$, $\epsilon'\equiv\epsilon_{\mathbf{p}_\parallel+\mathbf{q}_\parallel}$. The finite broadening γ takes into account the experimental resolution and finite lifetime of the pair fluctuations. We note that the above results are only valid for s-wave intralayer pairing g and an isotropic T_J. In this case, the isotropic s-wave gap ($\Delta_{\mathbf{p}_\parallel}=\Delta$) for the bilayer is self-consistently determined by the same BCS gap equation as for a single layer, namely

$$1-(g+T_J)\sum_{\mathbf{p}_\parallel}^{\omega_c}\frac{1}{2E}=0. \tag{10}$$

Taking into account that $\bar{\chi}_{11}=\bar{\chi}_{22}$ and $\bar{\chi}_{12}=\bar{\chi}_{21}$ [see also (7)], one may reduce (5) to the matrix equation

$$\begin{pmatrix}\chi_{11} & \chi_{12}\\ \chi_{21} & \chi_{22}\end{pmatrix} = \begin{pmatrix}1-a & -b^*\\ -b & 1-a^*\end{pmatrix}^{-1}\begin{pmatrix}\bar{\chi}_{11} & \bar{\chi}_{12}\\ \bar{\chi}_{12} & \bar{\chi}_{11}\end{pmatrix}, \tag{11}$$

where

$$a\equiv\tilde{v}\bar{\chi}_{11}+\bar{v}^*\bar{\chi}_{12},$$

$$b\equiv\tilde{v}\bar{\chi}_{12}+\bar{v}^*\bar{\chi}_{11}. \tag{12}$$

The solutions of (11) will give rise to the modes which involve oscillations of the two layers in a unit cell which are in-phase or out-of-phase relative to each other. We see from (6) that $\tilde{v}$ is always real. The solution of (11) is very much simplified if $\bar{v}$ in (6) is also real and this will be assumed in the present case (we will discuss this assumption shortly). We remark that if we set $T_J=0$, we have $\bar{\chi}_{12}=0$ and then (11) reduces to the results in Appendix B of Ref. 11.

The secular determinant of (11) is given by $(1-a)(1-a^*)-bb^*$. This factorizes if $\bar{v}$ is real since then a and b are also real. With $\bar{v}$ real, the matrix equation (11) is easily solved to give

$$\chi_{11}(\mathbf{q},\omega)=\chi_{22}(\mathbf{q},\omega)=\frac{\bar{\chi}_{11}-\tilde{v}(\bar{\chi}^2_{11}-\bar{\chi}^2_{12})}{D_+D_-},$$

$$\chi_{12}(\mathbf{q},\omega)=\chi_{21}(\mathbf{q},\omega)=\frac{\bar{\chi}_{12}+\bar{v}(\bar{\chi}^2_{11}-\bar{\chi}^2_{12})}{D_+D_-}. \tag{13}$$

Here the new denominators are given by

$$D_\pm(\mathbf{q},\omega)\equiv 1-v_\pm(\mathbf{q})E_\pm(\mathbf{q}_\parallel,\omega), \tag{14}$$

where we have defined[7]

$$v_\pm(\mathbf{q})\equiv\tilde{v}(\mathbf{q})\pm\bar{v}(\mathbf{q}) \tag{15}$$

and

$$E_\pm(\mathbf{q}_\parallel,\omega)\equiv\bar{\chi}_{11}(\mathbf{q}_\parallel,\omega)\pm\bar{\chi}_{12}(\mathbf{q}_\parallel,\omega) = A_0-\frac{4(g\pm T_J)c_0^2}{\bar{D}_\pm}. \tag{16}$$

A key simplifying feature of (13) related to $\bar{v}$ being real is that we can separate the in-phase (+) and out-of-phase (−) phase mode contributions to the response functions in (13), namely

$$\chi_{11}=\chi_{22}=\frac{1}{2}\left[\frac{E_+}{D_+}+\frac{E_-}{D_-}\right],$$

$$\chi_{12}=\chi_{21}=\frac{1}{2}\left[\frac{E_+}{D_+}-\frac{E_-}{D_-}\right]. \tag{17}$$

This also allows a clean separation of these two contributions in the inelastic light scattering cross section (see Sec. III). A similar separation has been discussed in Refs. 7, 11. In the present case, this separation depends on $\bar{v}$ in (6) being real.

We note that in the normal phase (when both the pairing interaction g and interlayer Cooper-pair coupling T_J vanish), the functions $E_\pm$ in (16) both reduce to A_0, which in this case is the well-known Lindhard function. However, in the superconducting case, there are two possible collective mode branches for a given value of q_z, as given by the solutions of $\mathrm{Re}D_\pm(\mathbf{q},\omega)=0$ in (14). As shown in Ref. 2, the mode given by the solution of $\mathrm{Re}D_+(\mathbf{q},\omega)=0$ corresponds to the in-phase Anderson-Bogoliubov phase mode, which is renormalized by the Coulomb interaction into 2D plasmons. In contrast, $\mathrm{Re}D_-(\mathbf{q},\omega)=0$ gives a mode which corresponds to an out-of-phase phase mode which is "massive," with an energy gap at low $\mathbf{q}_\parallel$ which is modified by the intralayer and interlayer Coulomb interactions (see Sec. III). As was pointed out in Ref. 2, this kind of out-of-phase mode of two coupled Cooper-pair condensates was first discussed in another context by Leggett.[8]

In calculating the inelastic light scattering cross section in Sec. III, we will need the response functions $\chi_{ij}(\mathbf{q}_\parallel,\omega,l-l')$ for the different layers. Performing the inverse Fourier transformation of (17) using (4), with the dependence of $\mathbf{q}_\parallel$ and ω left implicit, we find that the only q_z dependence of $\chi_{ij}(q_z)$ is through $v_\pm$ via the denominators $D_\pm$ in (14). This means one only has to calculate the following quantities:

$$I_\pm(l) \equiv \frac{c}{2\pi}\int_{-\pi/c}^{\pi/c} dq_z e^{-iq_z lc}\frac{1}{D_\pm(q_z)}, \quad (18)$$

and $\chi_{ij}(l-l')$ will thus be given by

$$\begin{aligned}\chi_{11}(l-l') &= \chi_{22}(l-l') \\ &= \frac{1}{2}\left[E_+I_+(l-l') + E_-I_-(l-l')\right] \\ \chi_{12}(l-l') &= \chi_{21}(l-l') \\ &= \frac{1}{2}\left[E_+I_+(l-l') - E_-I_-(l-l')\right]. \quad (19)\end{aligned}$$

At this point, it is useful to recall the physics of the in-phase (+) and out-of-phase (−) modes in conjunction with the fact that $d \ll c$. For the + mode, we do not lose much if we work in the $d \to 0$ limit since this mode corresponds to both layers oscillating in phase. In this case, we have $\bar{v} = \tilde{v}$ [see (6)] and hence in (15) we have $v_+ = 2\tilde{v}$. In contrast, for the − mode, the essential physics is captured by considering a single bilayer (i.e., in the $c \to \infty$ limit). In this approximation, v_- in (15) reduces to

$$v_- = v_{2D} - v_{2D}e^{-q_\parallel d}. \quad (20)$$

We note that in both v_+ and v_-, the approximation that we use is such that $\bar{v}$ in (6) is real, as was originally assumed in deriving (13). Using these results to calculate $D_\pm$ in (14), after some algebra, we obtain

$$\begin{aligned}I_+(l-l') &= \frac{1}{\sqrt{b^2-1}}\left[au^{-|l-l'|} - \frac{1}{2}\left(u^{-|l-l'-1|} + u^{-|l-l'+1|}\right)\right], \\ I_-(l-l') &= \left[1 - v_{2D}(1-e^{-q_\parallel d})E_-\right]^{-1}\delta_{l,l'}, \quad (21)\end{aligned}$$

where the parameters are defined as

$$\begin{aligned}a &\equiv \cosh q_\parallel c, \\ b &\equiv \cosh q_\parallel c - 2v_{2D}E_+\sinh q_\parallel c, \\ u &\equiv b + \sqrt{b^2-1}. \quad (22)\end{aligned}$$

In calculating the inelastic light scattering cross section in Sec. III, we only need to consider the case of very small momentum transfer $\mathbf{q}_\parallel$. In this limit and with $\gamma = 0$, the functions A_0, B_0, and c_0 in (9) can be approximated[2,3] for $q_\parallel \ll \Delta/v_F$ by

$$\begin{aligned}A_0 &= -\frac{N(0)}{4}J(\omega), \\ B_0 &= \frac{1}{g+T_J} + \frac{N(0)}{4}\left[\bar{\omega}^2 - \frac{1}{2}\bar{q}_\parallel^2\right]J(\omega), \\ c_0 &= \frac{N(0)}{8}\bar{\omega}J(\omega), \quad (23)\end{aligned}$$

with the dimensionless functions $J(\omega)$ defined by

$$J(\bar{\omega}) = \begin{cases} \dfrac{2}{\bar{\omega}\sqrt{1-\bar{\omega}^2}}\arcsin\bar{\omega}, & \bar{\omega} < 1, \\ \dfrac{2}{\bar{\omega}\sqrt{\bar{\omega}^2-1}}\left[\ln(\bar{\omega} - \sqrt{\bar{\omega}^2-1}) + i\dfrac{\pi}{2}\right], & \bar{\omega} > 1. \end{cases} \quad (24)$$

Here the barred quantities are defined by $\bar{\omega} \equiv \omega/2\Delta$ and $\bar{q}_\parallel \equiv v_F q_\parallel/(2\Delta)$; $N(0) = m^*/\pi\hbar^2$ is the 2D density of states at the Fermi surface. The imaginary part of function $J(\omega)$ only contributes when $\omega \geq 2\Delta$. For an s-wave superconductor, there are no BCS particle-hole states below 2Δ and thus the collective modes are well defined in this region. The validity of the above approximation[12,13] is not so good for frequencies near the threshold 2Δ, namely, when $2\Delta(1-\bar{q}_\parallel/2) < \omega < 2\Delta(1+\bar{q}_\parallel^2)^{1/2}$. However, this region is negligible for small $\mathbf{q}_\parallel$, and essentially makes no contribution due to the finite experimental resolution.

Substituting (23) and (24) into (16), we obtain

$$E_\pm(\mathbf{q}_\parallel,\omega) = \frac{1}{4}N(0)J(\omega)\left[\frac{-R_\pm + \frac{1}{8}\bar{q}_\parallel^2 J(\omega)}{R_\pm + \frac{1}{4}(\bar{\omega}^2 - \frac{1}{2}\bar{q}_\parallel^2)J(\omega)}\right], \quad (25)$$

where

$$R_+ = 0, \quad R_- = \frac{1}{N(0)g}\frac{2x}{x^2-1}; \quad x \equiv \frac{T_J}{g}. \quad (26)$$

Varying the parameter x (i.e., the ratio of T_J and g) provides a simple way to compare the results when T_J is larger or smaller than g. We note that in (25), in the limit of $q_\parallel \to 0$, we find $R_+ = 0$ and hence $E_+ \to 0$. In contrast, in this limit, E_- is finite since R_- is finite. This implies that the in-phase phase mode has less weight when $q_\parallel$ is small, while the out-of-phase phase mode is not too dependent on the value of $q_\parallel$ [see (17)]. For the case of finite damping, we need only replace ω by $\omega + i\gamma$ in (24)–(26).

III. INELASTIC LIGHT SCATTERING CROSS SECTION

For an incident photon with momentum $\mathbf{q}_i$, energy ω_i, and polarization $\hat{\epsilon}_i$ and a scattered photon described by $\mathbf{q}_f$, ω_f, and $\hat{\epsilon}_f$, the inelastic light scattering cross section at $T = 0$ is found to be proportional to[4,6]

$$\frac{d\sigma}{d\omega d\Omega} \propto |\hat{\epsilon}_i \cdot \hat{\epsilon}_f|^2 I(\mathbf{q},\omega), \quad (27)$$

where

$$I(\mathbf{q},\omega) = -\sum_{\substack{l,l' \\ i,j}} \mathrm{Im}\chi_{ij}(\mathbf{q}_\parallel,\omega,l-l') \times e^{-(Z_{l,i}+Z_{l',j})/\delta}e^{-2ik(Z_{l,i}-Z_{l',j})}. \quad (28)$$

We assume that the energy transfer to the superlattice

$\omega \equiv \omega_i - \omega_f$ is very small compared to the photon frequencies, i.e., $\omega_i \simeq \omega_f \equiv \omega_0$. The momentum transfer by the light parallel to the interface (denoted by $\mathbf{q}_{\parallel}$) is very small. The momentum transfer perpendicular to the superlattice can be approximated by $2k = (2\omega_0/c)\mathrm{Re}\sqrt{\epsilon}$ where ϵ is the complex dielectric function for optical photons in the medium containing the superlattice. The damping of light in the medium is described by the fact that the photon wave vector has an imaginary part

$$\mathrm{Im}q_{z,i} \simeq \mathrm{Im}q_{z,f} = \frac{\omega_0}{c}\mathrm{Im}\sqrt{\epsilon}. \quad (29)$$

This leads to the appearance of the penetration depth δ in (28), where $\delta^{-1} \equiv (2\omega_0/c)\mathrm{Im}\sqrt{\epsilon}$. The final result given in (28) is that the inelastic light scattering cross section involves a weighted sum of the density response functions given in Sec. II.

For simplicity, as shown explicitly in (27), we have only kept the *isotropic* matrix element for the Raman interaction. In most theoretical work on electronic Raman scattering in superconductors,[14,15,9] the main interest is usually on the anisotropic scattering cross sections from modes which have little wave vector dependence (dispersion). For this case, one can set the small momentum transfer to zero. In contrast, in our present work, we are interested in the scattering from phase modes with strong dispersion, which already show up in the isotropic scattering cross section given by (27).

Summing over $i,j = 1,2$ for bilayers, one obtains the more explicit expression for fixed $\mathbf{q}$

$$I(\omega) = -\sum_{l,l'}\Big[\mathrm{Im}\chi_{11}(\mathbf{q}_{\parallel},\omega,l-l')e^{-(l+l')c/\delta}e^{-2ik(l-l')c}(1+e^{-2d/\delta}) + \mathrm{Im}\chi_{12}(\mathbf{q}_{\parallel},\omega,l-l')e^{-(l+l')c/\delta}e^{-2ik(l-l')c}e^{-d/\delta}2\cos 2kd\Big], \quad (30)$$

where we have used the symmetry $\chi(l-l') = \chi(l'-l)$. By using (17)–(22) and summing over $l,l' = 0,...,+\infty$ in (30), we obtain

$$I(\omega) = -\frac{1}{1-e^{-2c/\delta}}\mathrm{Im}\Bigg\{\frac{E_+}{2}\left[1+\frac{2v_{2\mathrm{D}}E_+\sinh q_{\parallel}c(u^2e^{2c/\delta}-1)}{F\sqrt{b^2-1}}\right]\left(1+e^{-2d/\delta}+2e^{-d/\delta}\cos 2kd\right) + \frac{E_-}{2\left[1-v_{2\mathrm{D}}(1-e^{-q_{\parallel}d})E_-\right]}\left(1+e^{-2d/\delta}-2e^{-d/\delta}\cos 2kd\right)\Bigg\}, \quad (31)$$

where we have introduced the function

$$F \equiv u^2e^{2c/\delta} - 2ue^{c/\delta}\cos 2kc + 1. \quad (32)$$

These results are a generalization of those for a normal superlattice, as given in Ref. 4. In the right-hand side of (31), the first term ($\equiv I_I$) gives the contribution from the in-phase phase fluctuations, while the second term ($\equiv I_O$) is associated with the out-of-phase phase fluctuations.

For the CuO_2 layer superconductors, where $\delta \sim k^{-1}$, one can effectively set $\delta = \infty$ (i.e., ignore photon damping). In this limit, the pole due to $F = 0$ in the first term of (31) is given by the solution of $b = \cos 2kc$. This gives the in-phase (bulk) plasmon resonance for $q_z = 2k$ and corresponds to the Anderson-Bogoliubov (AB) mode, renormalized by the Coulomb interaction (for further discussion, see Ref. 11). The additional singularities of the first term in (31) are given by

$$b = \pm 1. \quad (33)$$

These correspond to the upper (+) and lower (−) limits of the bulk plasmon band for a infinite (or bulk) superlattice.[4] We note that since kc is very small for the cuprates, the AB mode given by $b = \cos 2kc$ is coincident with the upper limit of the bulk plasmon band given by $b = 1$.

In contrast, the pole of the second term in (31) associated with the solution of

$$1 - v_{2\mathrm{D}}(1-e^{-q_{\parallel}d})E_- = 0 \quad (34)$$

corresponds to the out-of-phase phase mode of a bilayer. As calculated in Refs. 2, 3, for isotropic g and T_J case and $x \lesssim 0.02$, this mode has the approximate long wavelength dispersion relation

$$\omega^2(q_{\parallel}) = \left[\omega_0^2 + \frac{1}{2}v_F^2q_{\parallel}^2\right]\left(1+\frac{m^*e^2d}{\epsilon_0}\right), \quad (35)$$

where the term involving e^2 is a result of the Coulomb interaction. More precisely, the interlayer Coulomb interaction screens the effect of the intralayer Coulomb interaction, which by itself strongly increases the energy of the out-of-phase phase mode of a neutral superconductor. The energy gap ω_0 for small x in a neutral superconductor is found to be given by[2]

$$\omega_0^2 \equiv \frac{4T_J}{N(0)g^2}(2\Delta)^2. \quad (36)$$

However, because $m^*e^2d \simeq 6$ in the high-T_c oxides of interest, one sees that the "effective energy gap" of the charged superconductor can be large even if $T_J \ll g$ and, in fact, this gap may be comparable or larger than 2Δ.

In Fig. 1, we have drawn the dispersion relation for

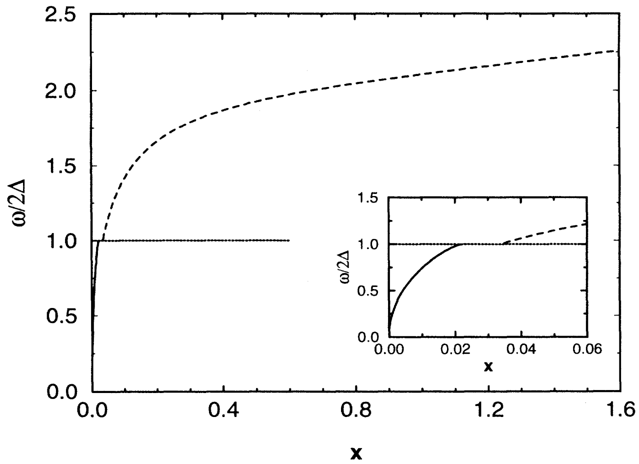

FIG. 1. The frequency of the out-of-phase phase modes in an s-wave superconductor with isotropic interlayer pair tunneling vs the value of x ($\equiv T_J/g$). The dashed line for energy above 2Δ implies that the modes are damped due to the pair-breaking mechanism. The parameters used are $m^* = m$, $\epsilon_0 = 2$, $q_{\|} = 0.006\Delta/v_F$, and $gN(0) = 0.25$.

the out-of-phase phase modes, emphasizing the strong dependence of the energy gap on the magnitude of x. Including the effect of the Coulomb interaction, the out-of-phase phase modes with $x \lesssim 0.02$ and frequency below 2Δ are well defined, but those at larger values of x and above 2Δ are strongly damped due to the p-h damping.

For comparison, we have plotted the Raman light scattering intensity based on (31) in Fig. 2 for $x = 0.01$ and Fig. 3 for $x = 1.5$, corresponding to two different regimes. We choose the following parameters appropriate for the high-T_c cuprates: superlattice period $c = 12$ Å, bilayer spacing $d = 3$ Å, pairing strength $gN(0) \equiv 0.25$, 2D hole density $n = 1.5 \times 10^{14}$ cm^{-2}, layer effective mass $m^* \equiv m$, background dielectric constant $\epsilon_0 = 2$, in-layer

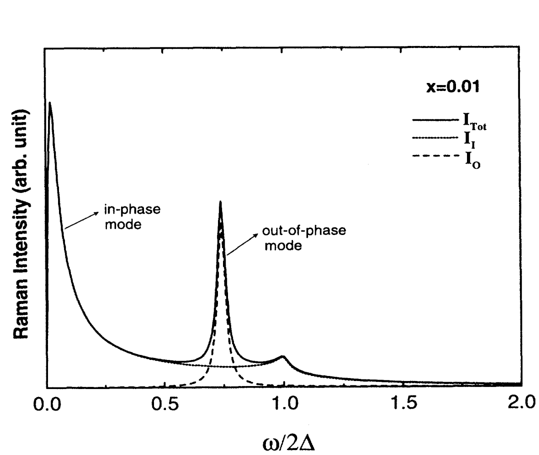

FIG. 2. The Raman light scattering intensity from phase modes is plotted for a semi-infinite superconducting superlattice with a basis of bilayers, with Cooper-pair tunneling occurring between the two layers. The parameters used are given after (36), with tunneling strength $T_J = 0.01g$ ($x = 0.01$).

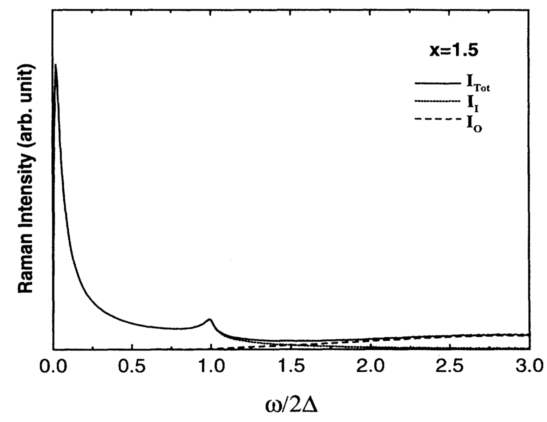

FIG. 3. The same plot as in Fig. 2, for $T_J = 1.5g$.

momentum transfer $q_{\|} = 0.006\Delta/v_F$, photon momentum in the z direction $k = 1.0 \times 10^5$ cm^{-1}, penetration depth $\delta \sim 1/k = 1000$ Å, and $\gamma = 0.04\Delta$.

The scattering intensity in Fig. 2 shows a strong peak corresponding to an energy gap of the out-of-phase phase mode of $\omega = 0.73(2\Delta)$, when $x = 0.01$ (see Fig. 1). In contrast in Fig. 3, the expected out-of-phase phase mode with energy $\omega = 2.36(2\Delta)$ for $x = 1.5$ is not visible due to the strong particle-hole damping (see Fig. 1). The spectrum above the threshold 2Δ is a continuum with a broadened maximum at about $\omega = 3(2\Delta)$. The peak at low frequency in both Figs. 2 and 3 corresponds to in-phase 2D plasmons, while the cusps correspond to $\omega = 2\Delta$. We can use the fact that $c, d \ll \delta, k^{-1}$ for high T_c layered superconductors to simplify (31). One can then easily check that the intensity of in-phase mode is not too dependent of the values of δ and k. In contrast, the intensity of out-of-phase mode is very dependent on the values of δ and k. Indeed, if the light was completely undamped as it penetrated the superlattice ($\delta^{-1} \to 0$), the intensity of the out-of-phase modes would be maximum (for a given value of k). We also note that if we let both δ^{-1} and k go to zero, then one can see from (30) in conjunction with (17) and (19) that the out-of-phase mode has zero weight in the light-scattering cross section.

Leggett[8] first pointed out that the out-of-phase phase mode is somewhat similar to the excitonlike mode for the s-wave (L=0) superconductors in the presence of an additional short-range d-wave attractive interaction g_2.[16] Unlike the excitonlike modes which only exist below 2Δ (since we must have $g_2 < g_0$), the bilayer out-of-phase phase modes can exist above as well as below 2Δ (apart from the different damping), since there is no limit on the magnitude of T_J.

In normal superlattices considered by Jain and Allen,[4] it has been shown that proper inclusion of the surface contribution to the layer response functions results in two effects. (1) Surface plasmons show up as resonances in the inelastic light scattering if the background dielectric constant ϵ_0 is taken to be different above and below the superlattice. (2) There is a negative weight contribution to $I(\omega)$ which cancels out the bulk mode Van Hove singularity associated with $b = -1$ in (33). We expect the analogous phenomena to arise in the superconducting superlattice under consideration.

Although we have only presented the results of Raman scattering intensities for an s-wave superconductor, similar results are expected for $d_{x^2-y^2}$-wave superconductors. The major difference between s-wave and d-wave superconductors[3] arises from the different p-h damping since for d-wave superconductors, pair breaking is allowed at frequencies below 2Δ (for some regions on the Fermi surface) as well as above 2Δ (here Δ is the maximum value of the energy gap). Due to the strong p-h damping below 2Δ for d-wave superconductors, it is estimated that the out-of-phase phase modes are only well defined and hence visible as resonances in $I(\omega)$ for $x \lesssim 0.002$ and $\omega \lesssim 0.2(2\Delta)$ (where the p-h damping is negligible). The low frequency in-phase phonon phase modes are always well defined for small $\mathbf{q}_{\|}$.

IV. CONCLUDING REMARKS

We have presented results for the isotropic Raman inelastic light scattering intensity for finite momentum transfer in a semi-infinite superconducting superlattice with a basis of bilayers, generalizing the work by Jain and Allen[4] for a normal superlattice of electron gas layers. We allow Cooper-pair tunneling between layers within the same unit cell . Due to this coupling of Cooper pairs in the two layers, there exists "out-of-phase" collective modes in addition to "in-phase" Anderson-Bogoliubov phase and Littlewood-Varma amplitude modes. These out-of-phase modes were recently studied thoroughly by Wu and Griffin[2,3] for both s-wave and d-wave pairing, and are the signature of the Chakravarty-Anderson Cooper-pair tunneling model.

In the present paper, we have concentrated on showing how the collective modes of the Cooper-pair order parameter show up as resonances in the inelastic light scattering from the surface of a layered superconductor with a bilayer basis. We have ignored the surface modification of the superlattice response functions and thus our results only include the "bulk" superlattice modes. The present results, however, are sufficient to give a qualitative picture of what one can expect from such inelastic light scattering studies. One could extend the present results to include the surface contribution, analogous to the work of Jain and Allen.[4] However, we might note that it is the out-of-phase phase oscillations that are of greatest interest and these are not expected to be modified much since they are associated with a single bilayer (i.e., they don't depend much on the intercell Coulomb interactions).

In addition to the order parameter *phase* fluctuations we have studied in this paper, there are also in-phase and out-of-phase *amplitude* fluctuations.[2,3] However, these have zero weight in the density response functions which enter into the inelastic light scattering cross section. Some other sort of experimental probe is needed to study the amplitude modes.

ACKNOWLEDGMENTS

W.C.W. thanks Professor T. Timusk and Professor B. W. Statt for useful discussions. This work was supported by a research grant from the NSERC of Canada. A.G. would also like to acknowledge the hospitality and financial support of the Universita di Trento during a sabbatical.

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[1] S. Chakravarty, A. Sudbø, P. W. Anderson, and S. Strong, Science **261**, 337 (1993).
[2] W. C. Wu and A. Griffin, Phys. Rev. Lett. **74**, 158 (1995).
[3] W. C. Wu and A. Griffin, Phys. Rev. B, **51**, 15 317 (1995).
[4] J. K. Jain and P. B. Allen, Phys. Rev. B **32**, 997 (1985).
[5] G. F. Giuliani, P. Hawrylak, and J. J. Quinn, Phys. Scr. **36**, 946 (1987).
[6] G. E. Santoro and G. F. Giuliani, Phys. Rev. B **37**, 8443 (1988).
[7] A. Griffin and A. J. Pindor, Phys. Rev. B **39**, 11 503 (1989).
[8] A. J. Leggett, Prog. Theor. Phys. (Kyoto) **36**, 901 (1966).
[9] W. C. Wu and A. Griffin, Phys. Rev. B **51**, 1190 (1995).
[10] M. Plamondon and A. Griffin (unpublished).
[11] R. Côté and A. Griffin, Phys. Rev. B **48**, 10 404 (1993).
[12] H. A. Fertig and S. Das Sarma, Phys. Rev. B **44**, 4480 (1991).
[13] H. Monien and A. Zawadowski, Phys. Rev. B **41**, 8798 (1990).
[14] M. V. Klein and S. B. Dierker, Phys. Rev. B **29**, 4976 (1984).
[15] T. P. Devereaux, Phys. Rev. B **45**, 12 965 (1992).
[16] A. Bardasis and J. R. Schrieffer, Phys. Rev. **121**, 1090 (1961).