C-axis Raman spectra of a normal plane-chain bilayer cuprate and the pseudogap

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Abstract

We investigate the Raman spectra in the geometry where both incident and scattered photon polarizations are parallel to the \( \hat{z} \)-direction, for a plane-chain bilayer coupled via a single-particle tunneling \( t_\perp \). The Raman vertex is derived in the tight-binding limit and in the absence of Coulomb screening, the Raman intensity can be separated into intraband \( (\propto t_\perp^4) \) and interband \( (\propto t_\perp^2) \) transitions. In the small-\( t_\perp \) limit, the interband part dominates and a pseudogap will appear as it does in the conductivity. Coulomb interactions bring in a two-particle coupling and result in the breakdown of intra- and interband separation. Nevertheless, when \( t_\perp \) is small, the Coulomb screening \( (\propto t_\perp^4) \) has little effect on the intensity to which the unscreened interband transitions contribute most. In general, the total Raman spectra are strongly dependent on the magnitude of \( t_\perp \).

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I. INTRODUCTION

Recently, $c$-axis properties of the high-$T_c$ superconductors have been of increasing interest both in normal and superconducting states. Among several theoretical works [1–8], a coupled plane-chain model has been proposed [9,10] to study various $c$-axis properties of YBaCuO. Due to a coupling (of magnitude $t_\perp$) between plane and chain along the $c$-axis, the properties in the $c$-direction are shown to be quite different as compared to the ones in the $a$-$b$ plane. More importantly, a finite band gap (splitting) between the two renormalized bands (arising from the finite coupling) gives a natural explanation of pseudogaps, which are mainly observed in the underdoped normal-state copper oxides [11–13] and are seen directly as a depression of the conductivity at low frequency at low temperatures.

In the past few years, Raman scattering as a probe to study high $T_c$ materials have been very successful [14–27] and have in particular played a significant role in the debate about the symmetry of the gap. Owing to the layer behavior of the high $T_c$ compounds, most experimental and theoretical studies have focused on the $a$-$b$ plane properties, taking into account different symmetrical geometries. In contrast in this paper, we want specially to study the $c$-axis normal-state Raman scattering, based on the coupled plane-chain bilayer model. This not only provides another way to test whether this model is valid in high $T_c$ cuprates, but also gives a path for extending calculations from a simple one band system to the more complicated multiband system. In particular, a better theory should take Coulomb interactions into account properly for a multiband system.

Devereaux et al. [28] have recently given a detailed account of Raman scattering in a superconducting bilayer with two identical planes. They use the effective mass approximation to derive the Raman vertex and have mainly studied the $a$-$b$ plane properties. In their calculations, they have ignored the interband transition which is not so important in the $a$-$b$ plane, but turns out to be the dominant part in some cases for the $c$-axis. This case is discussed in the present paper.

A summary of what we have done is as follows. We have derived the Raman vertex
in the tight-binding limit which is almost exact in a single bilayer limit. We have clearly exhibited in the $c$-axis Raman spectra, the role of the intra- and interband transitions which are separable in the absence of the Coulomb screening. The interband transitions contribute most in the small-$t_{\perp}$ limit. While a mixing term comes in when the Coulomb screening is included, the unscreened interband transition still dominates if $t_{\perp}$ is small. Appendix A gives the details of how one can separate the response functions needed in the calculation of the Raman spectra into intraband and interband contributions. In Appendix B, we present the analogous results for the $c$-axis Raman spectra in the case of a plane-plane bilayer model. In this instance, a simple analytic end expression can be obtained which helps gain physical insight into the $c$-axis Raman process. The total volume has been set to one ($\Omega = 1$) in this paper.

II. FORMALISM

The non-interacting Hamiltonian for the normal state of a coupled plane-chain bilayer cuprate is written as

$$H_0 = \sum_k C_k^\dagger h(k) C_k,$$

where we have defined the row and column vectors

$$C_k^\dagger = \left( c_{1k}^\dagger, c_{2k}^\dagger \right) ; \quad C_k = \begin{pmatrix} c_{1k} \\ c_{2k} \end{pmatrix},$$

and $c_{1k}^\dagger, c_{2k}^\dagger$ create an electron in the state $k$ in layers 1 and 2 respectively. The energy matrix

$$h(k) = \begin{pmatrix} \xi_1(k) & t(k_z) \\ t(k_z) & \xi_2(k) \end{pmatrix}.$$  

Here $\xi_1$ and $\xi_2$ are the (uncoupled) energy bands for the plane and chain respectively and $t$ (assumed to be real) is the perpendicular hopping coupling between plane and chain. These quantities are defined explicitly later in Eq. (22). The spin indices are suppressed throughout this paper.
To study the Raman spectra for a multiband system, we use an approach which is suitable in the tight-binding limit. The Hamiltonian (1) can be written in the Wannier representation as [29]

\[
H_0 = -\sum_{i=1}^{2} \sum_{\mathbf{R}_i, \mathbf{r}_i} \sigma_i c_i^\dagger (\mathbf{R}_i + \mathbf{r}_i) c_i (\mathbf{R}_i) \\
- \frac{t_{\perp}}{2} \sum_{\mathbf{R}_1, \mathbf{R}_2} \left\{ c_1^\dagger (\mathbf{R}_1) c_2 (\mathbf{R}_2) [\delta_{\mathbf{R}_1 + \mathbf{d}/2, \mathbf{R}_2} + \delta_{\mathbf{R}_1 - \mathbf{d}/2, \mathbf{R}_2}] + \text{H.c.} \right\},
\]

where \( \mathbf{R}_i \) are the lattice vectors in a plane, \( \mathbf{r}_i \) are the displacements to the nearest neighbors of \( \mathbf{R}_i \), \( d \) is the lattice constant in the \( z \) direction, and \( \sigma_i \) and \( t_{\perp} \) are the nearest-neighbor hopping energies within and between the chain and plane layers. The Wannier representation is connected to the \( k \)-space representation by

\[
c_i (\mathbf{R}_i) = \frac{1}{\sqrt{N}} \sum_k e^{i \mathbf{k} \cdot \mathbf{R}_i} c_i (\mathbf{k}),
\]

where \( N \) is the total number of lattice sites. In the presence of a magnetic vector potential \( \mathbf{A}(\mathbf{r}) \), the tight-binding Wannier states are modified by a phase such that [30]

\[
c_i (\mathbf{R}_i) \rightarrow c_i (\mathbf{R}_i) \exp \left[ -\frac{ie}{\hbar c} \mathbf{R}_i \cdot \mathbf{A}(\mathbf{R}_i) \right],
\]

where \( e \) is the charge on the electron, \( \hbar \) is Planck’s constant over \( 2\pi \), and \( c \) is the speed of light. The assumption is made that the vector potential is slowly varying over the length scale of the crystal lattice and hence \( \mathbf{A}(\mathbf{q}) \) is strongly peaked about \( \mathbf{q} = 0 \).

Substituting (8) and (9) into (4), to second order in \( \mathbf{A} \), Eq. (4) becomes

\[
H = H_0 - \frac{e}{2mc} \mathbf{p} \cdot \mathbf{A} + \frac{e^2}{2mc^2} \mathbf{\hat{\rho}} \cdot \mathbf{A}
\]

in the \( k \)-space, where the vector

\[
\mathbf{p} = \frac{m}{\hbar} \sum_k C_k^\dagger \frac{\partial h(\mathbf{k})}{\partial \mathbf{k}} C_k
\]

and the tensor

\[
\mathbf{\hat{\rho}} = \frac{m}{\hbar^2} \sum_k C_k^\dagger \frac{\partial^2 h(\mathbf{k})}{\partial \mathbf{k} \partial \mathbf{k}} C_k.
\]
In these formulas $m$ is the bare electron mass. In electronic Raman scattering, we are interested in the transition matrix to order $A^2$. Therefore, the term $A \cdot \hat{p} \cdot A$ in (7) is used in first-order perturbation theory and the term $p \cdot A$ in (7) is used in second-order perturbation theory. As a result, Raman scattering probes an “effective density”

$$\tilde{\rho} \equiv \sum_k C_k^\dagger \gamma(k) C_k,$$

where the vertex matrix has the element

$$\gamma_{ij}(k) = (\hat{e}^I \cdot \hat{p}_{ij} \cdot \hat{e}^S) + \sum_l \left[ \frac{\langle i, k | p \cdot \hat{e}^S | l, k \rangle \langle l, k | p \cdot \hat{e}^I | j, k \rangle}{\epsilon_j(k) - \epsilon_l(k) - \hbar \omega^I} \right. \\
+ \left. \frac{\langle i, k | p \cdot \hat{e}^I | l, k \rangle \langle l, k | p \cdot \hat{e}^S | j, k \rangle}{\epsilon_j(k) - \epsilon_l(k) - \hbar \omega^S} \right].$$

In Eq. (11), the summation is over all the uncoupled bands and the dependence of the momentum transfer $q$ on $\gamma_k$ is dropped since we assume $q \ll k_F$ (the Fermi momentum). $\epsilon_i(k)$ is the energy of the $i$-band, $\hat{e}^I (\hat{e}^S)$ is the polarization of incident (scattered) photon, and the frequency $\omega^I (\omega^S)$ denotes the incident (scattered) photon energy.

For the problem of a plane-chain bilayer, one has two bands. In the case that there is a band gap ($\Delta$) between these two bands which is very small compared to the energies of incident and scattered photons, $\Delta \ll \omega^I, \omega^S$, the second term of (11) is negligible. The Raman vertex matrix in (10) thus reduces to

$$\gamma(k) = \frac{m}{\hbar^2} \sum_{\mu, \nu} \hat{e}_\mu^I \frac{\partial^2 h(k)}{\partial k_{\mu} \partial k_{\nu}} \hat{e}_\nu^S.$$

Eq. (12) shows how one can study various geometries for the Raman spectra by choosing the appropriate incident and scattered photon polarizations. We note that while the vertices exhibited in (12) are somewhat similar to the vertex given by the famous “effective mass approximation” [31][28], the physical origin is different.

The differential cross section for Raman scattering is found to be [32]

$$\frac{d^2\sigma}{d\omega d\Omega} = \frac{1}{\pi} r_0^2 \frac{\omega^S}{\omega} \left[ 1 + n_B(\omega) \right] \text{Im} \chi_{\tilde{\rho}\tilde{\rho}}(q \to 0, i\omega_n \to \omega + i0^+),$$

where $n_B(\omega) = [\exp(\beta \omega) - 1]^{-1}$ is the Bose distribution function; $r_0 = e^2/mc^2$ is the so-called Thompson radius, and the Raman density response function
\[ \chi_{\tilde{\rho} \tilde{\rho}}(\mathbf{q}, i\omega_n) = -\int_0^\beta d\tau e^{i\omega_n \tau} \langle T_\tau \tilde{\rho}_\mathbf{q}(\tau) \tilde{\rho}_\mathbf{q}^\dagger(0) \rangle, \] (14)

which is given in terms of the effective density operator \( \tilde{\rho} \) given in Eq. (10). Taking into account the screening effect due to the Coulomb energy

\[ H_c = \frac{1}{2} \sum_\mathbf{q} \sum_{i,j=1}^2 U_{ij}(\mathbf{q}) \rho_i \rho_j \mathbf{q}, \] (15)

the Raman response function (14) in (13) will be replaced by [32-34]

\[ \chi_{\tilde{\rho} \tilde{\rho}}^{sc} = \chi_{\tilde{\rho} \tilde{\rho}} + \sum_{ij=1}^2 \chi_{\tilde{\rho} \tilde{\rho}} U_{ij}(\mathbf{q}) \chi_{\rho_i \rho_j} + \sum_{ijml=1}^2 \chi_{\tilde{\rho} \tilde{\rho}} U_{ij}(\mathbf{q}) \chi_{\rho_j \rho_m} U_{ml}(\mathbf{q}) \chi_{\rho_i \tilde{\rho}} + \cdots \] (16)

Both the intralayer and interlayer Coulomb interactions should be included. Here the real density on layer \( i \) is

\[ \rho_i \mathbf{q} = \sum_k c_{i,k}^\dagger \mathbf{q} c_{i,k} \] (17)

and the response functions \( \chi_{\tilde{\rho} \rho} \) and \( \chi_{\rho_i \rho_j} \) are defined analogously to \( \chi_{\tilde{\rho} \tilde{\rho}} \) by Eq. (14). Eq. (16) corresponds to RPA which is given diagrammatically in Fig. [1].

One can easily reduce the infinite series in (16) to

\[ \chi_{\tilde{\rho} \tilde{\rho}}^{sc} = \chi_{\tilde{\rho} \tilde{\rho}} + (\chi_{\tilde{\rho} \rho_1}, \chi_{\tilde{\rho} \rho_2})(1 - U\chi)^{-1}U \begin{pmatrix} \chi_{\rho_1 \tilde{\rho}} \\ \chi_{\rho_2 \tilde{\rho}} \end{pmatrix}, \] (18)

where \( \chi \) denotes the 2 \( \times \) 2 unit matrix and we have defined the Coulomb interaction matrix

\[ U = \begin{pmatrix} U_{11} & U_{12} \\ U_{21} & U_{22} \end{pmatrix} \] (19)

and the real density response function matrix

\[ \chi = \begin{pmatrix} \chi_{\rho_1 \rho_1} & \chi_{\rho_1 \rho_2} \\ \chi_{\rho_2 \rho_1} & \chi_{\rho_2 \rho_2} \end{pmatrix}. \] (20)

In the continuum limit, the intralayer Coulomb interactions \( U_{11} = U_{22} = \frac{2\pi e^2}{q ||} \) and the interlayer Coulomb interactions \( U_{12} = U_{21} = \frac{2\pi e^2}{q ||} e^{-q || c} \) \( (q ||) \) is the momentum transfer parallel to \( a-b \) plane and \( c \) is the bilayer spacing [33] which all go to infinity in the strong screening \( q || \rightarrow 0 \) limit. In the tight-binding strong-coupling limit, the couplings \( U_{ij} \) are themselves large. In either cases, one can approximate (18) by

\[ \chi_{\tilde{\rho} \tilde{\rho}}^{sc} = \chi_{\tilde{\rho} \tilde{\rho}} - (\chi_{\tilde{\rho} \rho_1}, \chi_{\tilde{\rho} \rho_2}) \chi^{-1} \begin{pmatrix} \chi_{\rho_1 \tilde{\rho}} \\ \chi_{\rho_2 \tilde{\rho}} \end{pmatrix}. \] (21)

The minus sign in (21) clearly exhibits the screening effect of the Coulomb interaction.
III. RESULTS AND DISCUSSIONS

Eqs. (13) and (21) allow one to study various symmetries of the screened Raman intensities for two-band systems. In this paper, however, we will focus on the $c$ (or $z$) axis Raman intensities. We first specify the two uncoupled bands and the coupling:

\[
\xi_1(k_x, k_y) = \frac{\hbar^2}{2m}(k_x^2 + k_y^2) - \mu + \Delta \\
\xi_2(k_y) = \frac{\hbar^2}{2m} k_y^2 - \mu \\
t(k_z) = -2t_\perp \cos(k_z d/2),
\]

(22)

where $\Delta$ corresponds to the band splitting (responsible for the “pseudogap”), $\mu$ is the chemical potential, and $t_\perp$ is the plane-chain coupling strength. While the bands $\xi_1$ and $\xi_2$ with circular and linear Fermi surface are used to simplify the calculation, they give qualitatively similar results to those given by more realistic tight-binding bands [9]. Since the $k_z$ dependence only comes in through $t(k_z)$ which is assumed to have no $k_x$ and $k_y$ dependence, the only relevant geometry for the $c$-axis Raman spectra will be $(\hat{e}_S, \hat{e}_I) = (\hat{z}, \hat{z})$. Following (12), we define

\[
\gamma_c(k_z) \equiv \begin{pmatrix} 0 & \gamma_{zz} \\ \gamma_{zz} & 0 \end{pmatrix},
\]

(23)

where $\gamma_{zz} = (m/\hbar^2) \partial^2 t(k_z)/\partial k_z^2 = (t_\perp d^2/2m/2\hbar^2) \cos(k_z d/2)$. Eq. (23) is the Raman vertex used in (10) for calculating the “$c$-axis Raman intensities” throughout this paper.

In Appendix A, we have shown how one can separate all the response functions of Eq. (21) (needed in the calculation of the $c$-axis Raman spectra) into an intraband and an interband contributions. With these separations, one can easily estimate how each susceptibility $\chi$ depends on the coupling strength $t_\perp$, for both intraband and interband transitions. Of most interest, one sees that the unscreened Raman response function $\chi_{\rho \rho}$ has an intraband contribution which is proportional $t_\perp^4$ and an interband contribution which is proportional $t_\perp^2$. Therefore, the interband transitions dominate for the unscreened $c$-axis Raman intensities when $t_\perp$ is small.
In Fig. 2, we first show the unscreened Raman intensities for small $t_\perp = 2\text{meV}$ at different temperatures. Fixed parameters $\Delta = 20\text{meV}$ and $\mu = 500\text{meV}$ are used for all the results presented in this paper. To include the impurity scattering, we then introduce the scattering rates $\Gamma_i$ into the Green’s functions in (A6)

$$G_0^{-1}(k, i\omega_n) = \begin{pmatrix}
    i\omega_n - \xi_1 + i\Gamma_1 \text{sgn}(\omega_n) & -t \\
    -t & i\omega_n - \xi_2 + i\Gamma_2 \text{sgn}(\omega_n)
\end{pmatrix}.$$ (24)

For simplicity, we will assume that the impurity scattering rates are equal for both bands ($\Gamma_1 = \Gamma_2 \equiv \Gamma$) and $\Gamma$ is linear in temperature ($\Gamma = 20\text{meV}$ at $T = 100K$), but independent of frequency and momentum. As seen in Fig. 2, the $c$-axis Raman spectra can exhibit clear signatures of a pseudogap, as long as the temperature is not too high. At high temperatures for which $\Gamma \geq \Delta$, the pseudogap feature is washed out by the impurity broadening. This is, in particular, shown by the different low-frequency dependence of the intensities (see the inset in Fig. 2).

In Fig. 3, we present the $T = 20K$ unscreened Raman intensities for different coupling strengths. The inset shows that the unscreened $c$-axis Raman intensity can, in general, be separated into an intraband and an interband contribution. For better clarity of presentation, all intensities have been divided by $t_\perp^2$. This leads to a almost constant interband contribution plus a intraband contribution proportional to $t_\perp^2$. One finds at larger $t_\perp$ that the pseudogap feature arising from the interband transitions are overwhelmed by the increasing contribution of intraband transitions which tend to dominate the Raman response.

We next consider the screened Raman intensity based on the full prescription of Eq. (21). Due to the Coulomb screening, the presence of the second (mixing) term in (21) results in the breakdown of the intraband and interband separation for the unscreened Raman intensity. For comparison with Figs. 2 and 3, we have presented in Figs. 4 and 5 the screened $c$-axis Raman intensities. Comparing Fig. 4 with Fig. 2, one sees no major difference between the unscreened and screened intensities except for different spectral weights of the higher-frequency intensity. This result is valid for the small-$t_\perp$ case and can be established as follows. When $t_\perp$ is small, one can ignore $\chi_{\rho_1\rho_2} \sim t_\perp^2$ in $\chi$ in (20) as compared to $\chi_{\rho_1\rho_1}, \chi_{\rho_2\rho_2}$.
\( \sim t_\perp^0 \) (see Appendix A). Consequently, (21) can be reduced to a more elegant result

\[
\chi_{\tilde{\rho}\tilde{\rho}}^{\text{sc}} = \chi_{\tilde{\rho}\tilde{\rho}} - 2 \sum_{i=1}^2 \left( \chi_{\rho_i\rho_i} \right)^2 \chi_{\rho_i\rho_i}.
\]  

(25)

That is, the screening effects can be separated for each layer. However, the complete separation of intraband and interband contributions is still not possible because the intra- and interband parts (both \( \sim t_\perp^2 \)) of \( \chi_{\tilde{\rho}\rho_i} \) are equally important (although \( \chi_{\rho_i\rho_i} \sim t_\perp^0 \) is dominated by the intraband transition). Nevertheless, the mixing (second) term in (25) is proportional to \( t_\perp^4 \) and has only a small effect when \( t_\perp \) is small. Therefore, the unscreened interband transition given by \( \chi_{\tilde{\rho}\tilde{\rho}} \) still dominates. For higher values of \( t_\perp \), as used in Figs. 5 and 3, the Coulomb screening has a large effect. In fact, in view of the inset in Fig. 5, the Coulomb screening effect tends to cancel against the unscreened interband contribution, largely leaving the unscreened intraband transition.

**IV. CONCLUSIONS**

In this paper, we have studied the \( c \)-axis Raman spectra such that both the incident and scattered photon polarizations are parallel to the \( \hat{z} \)-direction, for a plane-chain bilayer two-band cuprate. This coupled plane-chain model \( [9] \) has given a natural explanation of pseudogaps \( [11-13] \) seen in the AC \( c \)-axis optical conductivity. Here we have shown in detail how one can also study pseudogaps by doing \( c \)-axis Raman scattering. Our calculations are based on a tight-binding model for the Raman vertex. Coulomb screening effects are treated properly and include both intralayer and interlayer Coulomb interactions.

When the Coulomb screening is absent, the Raman intensity can be separated into an intraband and an interband contribution. The interband contribution (\( \propto t_\perp^2 \)) dominates in the small-\( t_\perp \) limit. However, the presence of the Coulomb screening mixes up this separation and the resulting intensities are strongly dependent on the magnitude of \( t_\perp \). Nevertheless, when \( t_\perp \) is small, the mixing effect arising from Coulomb screening is small and, as a result, the interband unscreened transition still dominates.
It’s obvious that the approach derived in this paper for calculating Raman spectra in a two-band system can be easily extended to the case of a multiband system. A extension of our calculations of $c$-axis Raman intensities to the case of a superconducting plane-chain bilayer cuprate will be given elsewhere including the effect of impurities.

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APPENDIX A: INTRABAND AND INTERBAND TRANSITIONS

In this appendix, we show how one can separate the response functions needed in the calculation of the Raman spectra into intraband and interband transitions. Consider a general response function

$$
\chi_{AB}(q, \omega_n) = -\int_0^\beta d\tau e^{i\omega_n \tau} \langle T_\tau A_q(\tau) B_q^\dagger(0) \rangle, \tag{A1}
$$

where here, the operators $A, B$ can be either the effective density $\rho$ or the real densities $\rho_1$ or $\rho_2$. These operators can be written in a unified form such that in the limit $q \to 0$

$$
A = \sum_k C_k^\dagger \gamma(k) C_k = \sum_k \sum_{i,j=1}^{\rho_2} \gamma_{ij}^A c_{i,k}^\dagger c_{j,k}. \tag{A2}
$$

In (A2), the matrix $\gamma$ is $\gamma_c$ given in (23) for $\rho$ and

$$
\gamma_1 = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \text{ for } \rho_1; \quad \gamma_2 = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \text{ for } \rho_2. \tag{A3}
$$

Using (A2), the average $\langle \cdots \rangle$ in (A1) becomes

$$
\langle T_\tau A_q(\tau) B_q^\dagger(0) \rangle
= \sum_{i,j,m,l=1}^2 \sum_k \sum_{k'} \gamma_{ij}^A(k) \gamma_{ml}^B(k') \langle T_\tau c_{i,k}^\dagger(\tau) c_{j,k}(\tau) c_{m,k'}^\dagger(0) c_{l,k'}(0) \rangle
$$

\[\text{eq:A1}\]
\[= - \sum_{ijml=1}^{2} \sum_{k} \sum_{k'} \gamma_{ij}^{A}(k) \gamma_{ml}^{B}(k') \langle T_{\tau} c_{l,k}(0) c_{l',k}^\dagger(\tau) \rangle \langle T_{\tau} c_{j,k}(\tau) c_{m,k'}^\dagger(0) \rangle \]
\[= - \sum_{ijml=1}^{2} \sum_{k} \sum_{k'} \gamma_{ij}^{A}(k) \gamma_{ml}^{B}(k) G_{ij}^{0}(k, -\tau) G_{ml}^{0}(k, \tau). \quad (A4)\]

The development from the second to the third line in (A4) makes use of the usual Hartree-Fock approximation. \(G_{ij}^{0}\) are the elements of a \(2 \times 2\) non-interacting Green’s function matrix defined by

\[G_{0}(k, \tau) = -\langle T_{\tau} c_{k}(\tau) c_{k}^\dagger(0) \rangle. \quad (A5)\]

For the present coupled plane-chain bilayer model, one has (see (3))

\[G_{ij}^{-1}(k, i\omega_{n}) = \begin{pmatrix} i\omega_{n} - \xi_{1} & -t \\ -t & i\omega_{n} - \xi_{2} \end{pmatrix}. \quad (A6)\]

Substituting (A4) into (A1) and making use of the Fourier transform

\[G_{ij}^{0}(k, i\omega_{n}) = \frac{1}{\beta} \sum_{\omega_{n'}} e^{-i\omega_{n'}\tau} G_{ij}^{0}(k, i\omega_{n'}) \quad (A7)\]

and the orthogonal relation

\[\frac{1}{\beta} \int_{0}^{\beta} d\tau \; e^{i(\omega_{n}-\omega_{n'})\tau} = \delta_{\omega_{n},\omega_{n'}}, \quad (A8)\]

one obtains

\[\chi_{AB}(q \to 0, i\omega_{n}) = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{k} \sum_{ijml=1}^{2} \gamma_{ij}^{A}(k) \gamma_{ml}^{B}(k) G_{ij}^{0}(k, i\omega_{n'}) G_{ml}^{0}(k, i\omega_{n'} + i\omega_{n}). \quad (A9)\]

Furthermore, using (23), (A3), and the fact that \(G_{12}^{0} = G_{21}^{0}\), one finds

\[\chi_{\bar{p}\bar{p}} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{k} \text{Tr}[G_{0}(k, i\omega_{n'}) \gamma_{c} G_{0}(k, i\omega_{n'} + i\omega_{n}) \gamma_{c}] \]
\[\chi_{\bar{p}\bar{c}} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{k} \text{Tr}[G_{0}(k, i\omega_{n'}) \gamma_{c} G_{0}(k, i\omega_{n'} + i\omega_{n}) \gamma_{i}] \]
\[\chi_{\bar{p}\bar{i}} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{k} \text{Tr}[G_{0}(k, i\omega_{n'}) \gamma_{i} G_{0}(k, i\omega_{n'} + i\omega_{n}) \gamma_{c}] \]
\[\chi_{\bar{i}\bar{c}} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{k} \text{Tr}[G_{0}(k, i\omega_{n'}) \gamma_{i} G_{0}(k, i\omega_{n'} + i\omega_{n}) \gamma_{i}] \quad (A10)\]

The trace operator enables one to calculate \(\chi_{\bar{p}\bar{p}}, \text{etc.}\) in (A10) in any rotated frame which is convenient. We will choose a frame in which the Green’s function matrix is diagonal. This leads to results that are readily interpretable physically (see later).
We first diagonalize the Hamiltonian \( h \) in (3) and the Green’s function matrix \( G_0^{-1} \) in (A6)

\[
    h(k) \rightarrow \hat{h}(k) = U(k) h(k) U(k) = \begin{pmatrix} \epsilon_+ & 0 \\ 0 & \epsilon_- \end{pmatrix},
\]

\[
    G_0^{-1} \rightarrow \hat{G}_0^{-1} = U G_0^{-1} U = \begin{pmatrix} i\omega_n - \epsilon_+ & 0 \\ 0 & i\omega_n - \epsilon_- \end{pmatrix},
\]

where the unitary matrix is

\[
    U(k) = \frac{1}{\sqrt{\epsilon_+ - \epsilon_-}} \begin{pmatrix} \frac{\xi_1 - \epsilon_-}{|t|} \sqrt{\xi_1 - \epsilon_-} & -\frac{\xi_1 - \epsilon_-}{|t|} \sqrt{\epsilon_+ - \xi_1} \\ -\frac{\epsilon_+ - \xi_1}{|t|} \sqrt{\epsilon_+ - \xi_1} & \frac{\epsilon_+ - \xi_1}{|t|} \sqrt{\xi_1 - \epsilon_-} \end{pmatrix},
\]

and the eigenvalues are

\[
    \epsilon_\pm = \frac{\xi_1 + \xi_2}{2} \pm \sqrt{\left(\frac{\xi_1 - \xi_2}{2}\right)^2 + t^2}.
\]

In an analogous way, one can rotate the vertices

\[
    \gamma_c \rightarrow \hat{\gamma}_c = U \gamma_c U = \frac{\gamma_{zz}}{\epsilon_+ - \epsilon_-} \begin{pmatrix} 2t & -\frac{t}{|t|} (\xi_1 - \xi_2) \\ -\frac{t}{|t|} (\xi_1 - \xi_2) & -2t \end{pmatrix},
\]

\[
    \gamma_1 \rightarrow \hat{\gamma}_1 = U \gamma_1 U = \frac{1}{\epsilon_+ - \epsilon_-} \begin{pmatrix} \xi_1 - \epsilon_- & |t| \\ |t| & \epsilon_+ - \xi_1 \end{pmatrix},
\]

and

\[
    \gamma_2 \rightarrow \hat{\gamma}_2 = U \gamma_2 U = \frac{1}{\epsilon_+ - \epsilon_-} \begin{pmatrix} \epsilon_+ - \xi_1 & -|t| \\ -|t| & \xi_1 - \epsilon \end{pmatrix}.
\]

For convenience, we redefine

\[
    \hat{G}_0 = \begin{pmatrix} G_{11} & 0 \\ 0 & G_{22} \end{pmatrix}, \quad \hat{\gamma}_c = \begin{pmatrix} \gamma_{11} & \gamma_{12} \\ \gamma_{12} & -\gamma_{11} \end{pmatrix},
\]

\[
    \hat{\gamma}_1 = \begin{pmatrix} \alpha_{11} & \alpha_{12} \\ \alpha_{12} & -\alpha_{11} \end{pmatrix}, \quad \hat{\gamma}_2 = \begin{pmatrix} \alpha_{22} & -\alpha_{12} \\ -\alpha_{12} & \alpha_{11} \end{pmatrix},
\]

where one can easily identify these new variables. Substituting (A17) into (A10), we obtain

\[\text{(A17)}\]
\[ \chi_{\tilde{p}\tilde{p}} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{\mathbf{k}} \gamma_{11}^2 (G_{11}G'_{11} + G_{22}G'_{22}) + \gamma_{12}^2 (G_{11}G'_{22} + G_{22}G'_{11}), \]

\[ \chi_{\tilde{p}p_1} = \chi_{\tilde{p}p_2} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{\mathbf{k}} \gamma_{11} \left( \alpha_{11} G_{11}G'_{11} - \alpha_{22} G_{22}G'_{22} \right) + \gamma_{12} \alpha_{12} (G_{11}G'_{22} + G_{22}G'_{11}), \]

\[ \chi_{\tilde{p}p_2} = \chi_{\tilde{p}p_1} = \frac{1}{\beta} \sum_{\omega_{n'}} \sum_{\mathbf{k}} \gamma_{12} \left( \alpha_{11} G_{11}G'_{11} - \alpha_{22} G_{22}G'_{22} \right) - \gamma_{12} \alpha_{12} (G_{11}G'_{22} + G_{22}G'_{11}), \]

where \( G = G'(k, i\omega_{n'}) \) and \( G' = G(k, i\omega_{n'} + i\omega_n) \). Clearly for each \( \chi \) in (A18), the first term corresponds to an intraband contribution and the second term corresponds to an interband contribution. Crudely speaking, when \( t_\perp \) is small (recall (22) and (23)), \( \gamma_{11} \sim t_{\perp}^2, \gamma_{12} \sim t_{\perp}^1, \alpha_{11} \sim t_{\perp}^0, \alpha_{22} \sim t_{\perp}^2, \) and \( \alpha_{12} \sim t_{\perp}^1 \). These in turn allow one to estimate the \( t_\perp \)-dependence of each \( \chi \), for both intraband and interband transitions.

**APPENDIX B: PLANE-PLANE BILAYER MODEL**

In this Appendix, we present the analogous results for the \( c \)-axis Raman intensities in a coupled plane-plane bilayer. Due to the identity of these two layers, analytical results are available and hence gives more insight into the numerical results of the plane-chain model. Similar to Eq. (22), we use the simple bands

\[ \xi_1(k_x, k_y) = \frac{\hbar^2}{2m} (k_x^2 + k_y^2) - \mu + \Delta \]

\[ \xi_2(k_x, k_y) = \frac{\hbar^2}{2m} (k_x^2 + k_y^2) - \mu, \quad (B1) \]

for the two planes which are separated by a band gap \( \Delta \) (pseudogap). The coupling between these two planes is the same as given in (22). In the case of small \( t_{\perp} \), one can approximate \( \epsilon_+ = \xi_1 \) and \( \epsilon_- = \xi_2 \) (see (A13)).

With the above simplification, the unscreened \( c \)-axis Raman intensity given by the imaginary part of \( \chi_{\tilde{p}\tilde{p}} \) in (A18) can easily be worked out to be proportional to
\[ t_{\perp}^2 \omega \left[ \frac{6 t_{\perp}^2}{\Delta^2} \frac{4\Gamma}{(h\omega)^2 + 4\Gamma^2} + \left( \frac{2\Gamma}{(h\omega + \Delta)^2 + 4\Gamma^2} + \frac{2\Gamma}{(h\omega - \Delta)^2 + 4\Gamma^2} \right) \right], \]

(B2)

where the first term corresponds to the familiar intraband transition \((\propto t_{\perp}^4)\) and the second term corresponds to the interband transition \((\propto t_{\perp}^2)\). One can see from (B2) that apart from the prefactor \(t_{\perp}^2 \omega\), the interband term contains the same factors as the intraband term except for a peak shift from \(\omega = 0\) to \(\omega = \pm \Delta\) in the Lorentzian form. At zero frequency the first Drude like contribution in (B2) will go like \(6 t_{\perp}^2 / \Delta^2 \Gamma\) while the second (interband) term will go instead like \(4\Gamma / \Delta^2\) which is the opposite dependence on \(\Gamma\) than exhibited by the first term. Also for \(t_{\perp} \to 0\) the Drude term is small compared with the second term which will then exhibit a pseudogap for \(h\omega \lesssim \Delta\) as \(\Gamma \to 0\). Using (B2), we plot in Fig. 6 the temperature-dependent \(c\)-axis Raman intensities for the plane-plane bilayer model. All the parameters used are the same as those used in Fig. 2. It is seen in Fig. 6 that a pseudogap can develop in the \(c\)-axis Raman intensities at low frequencies provided the temperature (or the impurity damping rate \(\Gamma\)) is not too high. We remark that, in analogy to the plane-chain case, the Coulomb screening has little effect on the small-\(t_{\perp}\) unscreened intensities given in Fig. 3.

The major difference between the results in Fig. 4 for a plane-chain bilayer and those in Fig. 5 for a plane-plane bilayer is that the spectral weight just beyond the pseudogap frequency is much higher in the former case than in the latter case. This can be explained as follows. For a plane-plane bilayer in which the two bands are separated by a constant band gap \((\Delta)\) all over the Brillouin zone, the interband transitions will be peaked at \(\omega = \Delta\). In contrast in the plane-chain bilayer, the band gap between the plane and chain bands varies at different \(k\) points (with minimum value of \(\Delta\)) and, as a result, the interband transitions are spreaded out for \(\omega \geq \Delta\).
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\[ \chi^{\text{sc}} = \chi + \chi_{1,2} + \chi_{1,2} + \cdots \]

**FIG. 1.** Diagrams for the screened Raman response function. Each interaction line comprises four parts corresponding to intralayer and interlayer Coulomb interactions.
FIG. 2. Unscreened $c$-axis Raman intensities for $t_\perp = 2$meV at various temperatures. We magnify in the inset the different $\omega$-dependence of the low-frequency intensities.
FIG. 3. Unscreened $c$-axis Raman intensities at $T = 20K$ with different values of plane-chain coupling $t_\perp$. For easier comparison, all intensities are divided by $t_\perp^2$. The inset shows the separation into intraband and interband transitions of the unscreened $c$-axis Raman intensity (for the $t_\perp = 10\text{meV}$ case).
FIG. 4. Screened $c$-axis Raman intensities for $t_\perp = 2$meV at various temperatures (to be compared to Fig. 2).
FIG. 5. Screened $c$-axis Raman intensities at $T = 20K$ for various values of $t_\perp$ (to be compared to Fig. 3). The inset (for $t_\perp = 10\text{meV}$) shows separably the three different contributions, namely the unscreened intraband, unscreened interband, and screening effect.
FIG. 6. Unscreened $c$-axis Raman intensities in a coupled plane-plane bilayer model for $t_\perp = 2\text{meV}$ at various temperatures. The inset shows the different low-frequency $\omega$-dependence of the intensities. This is to be compared with Fig. 2 which applies to the plane-chain bilayer.