Interlayer Coupling Induced Quasiparticles

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We present an exact treatment of layered many-body electronic systems in the presence of interlayer coupling within the Schwinger functional derivative approach on the Keldysh contour. Our transparent approach allows us to clarify the definition of interlayer coupling by showing the independent roles hybridization and interactions play in generating new electronic and magnetic excitations. We find interlayer coupling to induce a variety of plasmons, magnons, and excitons, residing within the a layer, traversing between layers, or propagating along the interface. Moreover, we predict new interfacial excitations, including a new electron-hole pairing pathway, facilitated by previously ignored layer non-conserving interactions. Finally, we briefly explore the consequence of interlayer coupling on a bilayer square lattice system.

I. INTRODUCTION

Atomically thin 2D materials have proven to be one of the most exciting platforms exhibiting an extensive range of novel electronic\textsuperscript{1}, excitonic\textsuperscript{2}, valley\textsuperscript{3}, topological\textsuperscript{4}, and correlated physics.\textsuperscript{5} By combining the 2D building blocks into vertical or lateral heterostructures one may rationally engineer complex multilayer systems and artificial solids with new emergent properties giving way to direct applications in quantum information technologies\textsuperscript{6,7}, spin optoelectronic devices\textsuperscript{7,8}, and energy storage\textsuperscript{9,10}. To design and manipulate these novel layered materials a detailed theoretical description of the charge, spin, orbital, and layer degrees of freedom is crucial. However, despite vigorous experimental efforts, the development of theoretical techniques going beyond the Hohenberg-Kohn-Sham density functional theory to capture interactions in a layer dependent manner has been slow. In particular, one of the most important first-principles many-body methods used in theoretical spectroscopy for describing excitations involved in radiation-matter interaction is the so-called GW and Bethe-Salpeter Equation (BSE), still await an extension to layer dependent interactions.

The strength of interlayer coupling plays a key role in shaping the emergent properties of heterostructures composed of 2D thin-films. For example, when layers are weakly coupled, the absorption profiles of the individual layers is modified,\textsuperscript{11} along with the Raman vibrational modes.\textsuperscript{12} In the intermediate regime, the generation of new excitons (inter-layer and moiré)\textsuperscript{13–15} is facilitated along with the stabilization of superconducting phases.\textsuperscript{16} Lastly, in the limit of strong interlayer coupling, robust charge redistribution is induced\textsuperscript{17–20} and the electronic structure of the heterostructure differs considerably from its constituent layers.\textsuperscript{18,21,22}

The influence of interlayer coupling extends beyond atomically-thin 2D materials, playing a significant part in layered transition-metal oxides. In the Ruddlesden-Popper perovskite crystal structure, which includes the cuprate and iridate material families, the two-dimensional perovskite planes are interwoven with layers of alkaline earth, or rare earth metals, and are believed to behave electronically independent.\textsuperscript{23,24} However, a diversity of optimal transition temperatures is observed in the high-temperature cuprate superconductors which appears to be driven at least in-part by the choice of rock-salt layer separating the CuO$_2$ planes. For example, the highest Tc obtained in La$_{2-x}$Sr$_x$CuO$_4$ is 40k, whereas in the single layer Hg cuprate, HgBa$_2$CuO$_4$ the optimal Tc is almost 100K.\textsuperscript{25} Suggesting that the interlayer interactions between the CuO$_2$ planes and the HgO$_2$ charge-reservoir help to enhance Tc.

Previous studies using many-body perturbation theory on layered electron gas systems\textsuperscript{26–31} found the electron effective mass and quasi-particle life time gave qualitatively different results compared to isolated two- and three-dimensional systems. These models consist of a many-electron Hamiltonian with a Coulomb interaction only, where the electron-electron interaction were restricted to be with in a single layer. Therefore, there is currently no theory that addresses the many-body dynamics arising from the full spin and layer dependent interactions.

In this article we present an exact treatment of layered many-body electronic systems within the Schwinger functional derivative technique on the Keldysh contour. Our results clarify the definition of interlayer coupling by showing the independent roles hybridization and interactions play in generating new electronic and magnetic excitations. By examining the charge and magnetic response functions, along with the Bethe-Salpeter equation for the two-particle Green’s function, we predict new interfacial plasmons, magnons, and excitons facilitated by layer non-conserving interactions. We briefly explore the consequence of interlayer coupling on a bilayer square lattice system.

II. THEORY

A. Hamiltonian and Basic Notations

The Hamiltonian for a layered system with spin and layer dependent interactions is given by
where the Greek and Latin letters denote the spin and layer degrees of freedom, respectively. Our interaction index notion follows an \( in, in, out, out \) scheme inline with the diagrammatic representation. For an \( N \) layer system the Hamiltonian of the \( l \)th layer is given by \( H_{0, l}^{\alpha, \beta}(r) \). If the layers are close enough for the wave functions of adjacent layers to overlap, electrons can hop from one layer to another. The amplitude of hopping from layer \( l' \) to layer \( l \) is \( t_{\alpha, \beta}^{l, l'}(r) \). Here,\( r \) is defined over \( \mathbb{R}^3 \) and the field operators acting on a specific layer \( l \) can be written as \( \hat{\psi}(r) \equiv \hat{\psi}(r + R_l) \), where \( R_l \) is perpendicular to the interface between the layers and is the distance of the \( l \)th layer from the origin layer, \( R_0 = 0 \).

The generalized two-particle interaction takes both the spin and layer configuration into account and can be broken down into three contributions:

\[
\hat{H} = \sum_{\alpha, \beta} \int d^3 r \hat{\psi}_{\alpha r}^\dagger(r) H_{0, \alpha, \beta}(r) \hat{\psi}_{\beta r}(r) + \frac{1}{2} \sum_{\alpha, \beta, \gamma, \delta, i, j, k, l} \int d^3 r d^3 r' \hat{\psi}_{\alpha i r}^\dagger(r) \hat{\psi}_{\beta j r'}^\dagger(r') t_{\delta k r}^{l, l'}(r, r') \hat{\psi}_{\gamma k r'}(r') \hat{\psi}_{\delta \beta r}(r) \tag{1}
\]

To keep the results and the discussion general we define all operators in the contour Heisenberg picture,

\[
O(z) = U(z_0, z) \hat{O} U((z, z_0), \tag{3}
\]

with the time arguments, \( z \) and \( z_0 \), running along the Keldysh contour (\( z \in C \)). Where \( z_0 \) is an arbitrary initial time and the time evolution operator, \( U(z_0, z) \), evolves an operator \( \hat{O} \) from \( z_0 \) to \( z \) along the contour. In this picture the operators are explicitly time dependent whereas the wave functions are not. This allows us to introduce the time ordering on the contour and Wick’s theorem, connecting our results to many-body perturbation theory.\(^{34}\)

In order to treat the electronic many-body dynamics at finite temperature, we define the time-dependent ensemble average of operator \( O(z) \) as

\[
\langle O(z) \rangle = \frac{\text{Tr} \left\{ T \exp \left[ -i \int_0^T dz H(z) \right] O(z) \right\}}{\text{Tr} \left\{ T \exp \left[ -i \int_0^T dz H(z) \right] \right\}}, \tag{4}
\]

where \( \langle O(z) \rangle \) is the overlap between the initial state in thermodynamical equilibrium (for temperature \( \beta \)) at \( z_0 \) with the time evolved state at \( z \).

To obtain the exact expression for the self-energy, the vertex, and various other quantities we will use the Schwinger functional derivative approach.\(^{35} - 37\) To do so, we couple our Hamiltonian to a time dependent auxiliary electromagnetic field that probes the charge, spin, and layer degrees of freedom. The electric and magnetic fields are given in a compact form by

\[
\hat{\pi}(z_1) = \int d^3 r \sigma_{\alpha r}^I(1) \hat{\psi}_{\alpha r}^\dagger(1) \sigma_{\alpha r}^J \hat{\psi}_{\beta r}(1). \tag{5}
\]

Now if we wish to find the infinitesimal change in a generic, contour-ordered product of operators \( \Pi_i O_i(z_i) \) with respect to field \( \pi^I_{\alpha r}(1) \), we arrive at the following
where we have introduced the short hand anti-commutation relations of the contour hilation operators were also taken to obey the canonical \( \hat{a}^{\dagger} \hat{a} = \delta_{\alpha\beta} \). The electron creation and annihilation operator is applied. The electron creation and annihilation operator is applied.

\[
\begin{align*}
\left( i \frac{d}{dz} \delta_{\beta'\sigma} \delta_{\alpha\beta} - h^{0}_{\alpha,\beta}(1) \right) G_{\beta',\sigma}(1, 2) &= \delta(1, 2) \delta_{\alpha\sigma} \delta_{nm} - \frac{i}{2} \delta_{\gamma\delta} \delta_{\alpha\beta} \delta_{\gamma\delta} \delta_{\alpha\beta} G_{\gamma,\delta}(3, 1) G_{\gamma,\delta}(3, 1), \\
G_{\gamma,\delta}(3, 1) &= \frac{1}{2} \left( \hat{\psi}_{\gamma}(1) \hat{\psi}_{\delta}(3) \hat{\psi}_{\gamma}(3) \hat{\psi}_{\delta}(3) \right), \\
G_{\gamma,\delta}(3, 1) &= \frac{1}{2} \left( \hat{\psi}_{\gamma}(1) \hat{\psi}_{\delta}(3) \hat{\psi}_{\gamma}(3) \hat{\psi}_{\delta}(3) \right),
\end{align*}
\]

where a repeated index or variable implies a summation or integration, provided the repeated indices are on the same side of the equation.

Originally, Hedin just considered an external electric field which was used to relate the two-particle Green’s function to the functional derivative of the single particle Green’s function with respect to the probing electric field. Here, we have coupled our Hamiltonian to a layer dependent electromagnetic field allowing us to capture the intertwined charge, spin, and layer excitations of the system. Therefore, by Eq. 6, the two-particle Green’s function can be written as

\[
\begin{align*}
G_{\gamma,\delta}(3, 1) &= \frac{1}{2} \left( \hat{\psi}_{\gamma}(1) \hat{\psi}_{\gamma}(3) \hat{\psi}_{\delta}(3) \hat{\psi}_{\delta}(3) \right),
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\]

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\end{align*}
\]

where a repeated index or variable implies a summation or integration, provided the repeated indices are on the same side of the equation.
From this relation we recover the mass operator,
\[ \mathcal{M}_{\alpha n,\nu l}(1, 3) G_{\nu l,\sigma m}(3, 2) = -i \sigma_{\delta l}^{I} v_{IJ}^{lk;\nu l}(3, 1) \sigma_{\alpha n}^{J} G_{\gamma k,\sigma l,\xi,\sigma m}(1, 3, 3^{+}, 2) \]
\[ = V_{H}^{J} k_{n}(1) \sigma_{\alpha n}^{J} G_{\gamma k,\sigma m}(1, 2) + \Sigma_{\alpha n,\nu l}(1, 5) G_{\nu l,\sigma m}(5, 2). \]  

(14)

Two contributions can be readily identified, the generalized Hartree potential
\[ V_{H}^{J} k_{n}(1) = \rho_{l}^{J}(3) v_{IJ}^{lk;\nu l}(3, 1), \]  
and the exact self-energy
\[ \Sigma_{\alpha n,\nu l}(1, 5) = -i v_{IJ}^{lk;\nu l}(3, 1) \sigma_{\alpha n}^{J} G_{\gamma k,\mu s}(1, 4) \frac{\delta G_{\mu s,\nu l}(4, 5)}{\delta \pi_{l}^{I}(3)}. \]  

(16)

Where we have used the identity,
\[ \frac{\delta (G^{-1} G)}{\delta \pi} = 0 \implies \frac{\delta G}{\delta \pi} = -G \frac{\delta G^{-1}}{\delta \pi} G, \]

to pull out a factor of $G$ along with the definition of the charge and spin density,
\[ \rho_{l}^{J}(3) = -i G_{\delta l, \xi}(3, 3^{+}) \sigma_{\xi}^{J}. \]  

(18)

To uncover the richness of the self-energy, we expand the functional derivative in the self-energy in terms of the total field
\[ \Phi_{nk}(1) = \pi_{nk}^{I}(1) + V_{H}^{I} k_{n}(1). \]  

(19)

We find, just as Hedin, that the self-energy is made up of three interwoven components: the single-particle Green’s function, the screened interaction, and the vertex,
\[ \Sigma_{\alpha n,\nu l}(1, 5) = i \sigma_{\alpha n}^{J} G_{\gamma k,\mu s}(1, 4) \Lambda_{\mu s,\nu l}(4, 5; 6) W_{ak;bn}^{(J)}(6, 1). \]  

(20)

The vertex is
\[ \Lambda_{\mu s,\nu l}(4, 5; 6) = -\frac{\delta G_{\mu s,\nu l}^{-1}(4, 5)}{\delta \Phi_{ab}^{I}(6)} \]  

and the screened interaction is
\[ W_{ak;bn}^{(J)}(6, 1) = \frac{\delta \Phi_{ab}^{I}(6)}{\delta \pi_{l}^{I}(3)} v_{IJ}^{lk;\nu l}(3, 1), \]  

(22a)

\[ = \varepsilon_{a l}^{-1} L J(6, 3) v_{IJ}^{lk;\nu l}(3, 1). \]  

(22b)

To find the self-consistent equations governing $W$ and $\Lambda$, we use the equation of motion of $G$ and employ the chain-rule,
\[ W_{ak;bn}^{(J)}(6, 1) = \varepsilon_{a l}^{-1} L J(6, 3) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \left( \delta(6, 3) \delta(6, \delta(6, 4)) \right) \rho_{dc}^{M}(7) \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \varepsilon_{a l}^{-1} L J(6, 1) + \rho_{dc}^{M}(7) \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ \Lambda_{\mu s,\nu l}(4, 5; 6) = \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \lambda_{0, \nu l}^{M}(7, 8) W_{f k;bn}^{(J)}(6, 1), \]

where we used the indistinguishability of particles, $\varepsilon_{a l}^{-1} L J(6, 1) = \rho_{dc}^{M}(7) \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
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Additionally, we define the polarization as
\[ \chi_{\nu l}^{M}(7, 8) = \rho_{dc}^{M}(7) \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8), \delta(\Phi_{ab}^{I}(8))) \right) v_{IJ}^{lk;\nu l}(3, 1) \]
\[ = \lambda_{0, \nu l}^{M}(7, 8) W_{f k;bn}^{(J)}(6, 1), \]

where we used the indistinguishability of particles, $\varepsilon_{a l}^{-1} L J(6, 1) = \lambda_{0, \nu l}^{M}(7, 8) W_{f k;bn}^{(J)}(6, 1), \]

(25)

FIG. 2. (color online) Diagrammatic representation of the self-energy (a), the vertex (b), and the screened interaction (c) functions. The spin indices have been suppressed for clarity.

The complete set of self-consistent layer and spin dependent Hedin’s equations relating the electronic self-energy $\Sigma$ to the Green function $G$ and the screened interaction $W$, using the vertex $\Lambda$ and polarization function $\lambda_0$ are:
\[
\Sigma_{\alpha n,\nu t}(1,5) = i\sigma^\mu_{\alpha\gamma} G_{\gamma k,\mu s}(1,4) \Lambda^{L L}_{\mu s,\nu t}(4,5;6) W^{L l}_{ak;bn}(6,1),
\]
\[
W^{L l}_{ak;bn}(6,1) = \nu_{L l}^{ak;bn}(6,1) + \nu_{L l}^{ad;be}(6,7) \chi^{MN}_{0 ef;dg}(7,8) W^{N l}_{k;gm}(8,1),
\]
\[
\chi^{MN}_{0 ef;dg}(7,8) = -i G_{\delta c,\mu s}(7,9) \Lambda^{N}_{\mu s,\delta l}(9,10;8) G_{\nu t,\xi d}(10,7^+;\xi d)^M,
\]
\[
\Lambda^{L L}_{\alpha n,\nu y}(1,4;6) = \delta(1,6) \delta(1,4) \sigma^\mu_{\alpha\gamma} \delta_{\alpha n} \delta_{\nu y} + \frac{\delta\Sigma_{\alpha n,\nu y}(1,4)}{\delta G_{\mu s,\nu t}(9,10)} \delta_{\nu t,\xi d}(9,11) \Lambda^{L L}_{\epsilon g,\delta f}(11,12;6) G_{\delta f,\mu s}(12,10).
\]

To close the set of equations, Dyson’s equation
\(G_{\alpha n,\beta m}(1,2) = \)
\(G_0_{\alpha n,\beta m}(1,2) + G_{0 \alpha n,\nu s}(1,3) \Sigma_{\nu s,\delta l}(3,4) G_{\delta l,\beta m}(4,2),\)

links the fully interacting system to the bare non-interacting propagator,
\(G^{-1}_0_{\alpha n,\nu y}(1,4) = \)
\(\left(i \frac{d}{d\epsilon} \delta_{\alpha y} \delta_{\nu y} - h_0^{\alpha}_{\alpha n,\nu y}(1) - \Phi^{N}_{\nu y}(1) \sigma_{\alpha y} \right) \delta(1,4).\)

A diagrammatic representation of these equations are shown in Fig. 2.

FIG. 3. (color online) Diagrammatic representation of the self-energy in the GW approximation for layer conserving (a-b) and layer non-conserving (c-d) interactions. The right hand (left hand) diagrams show an electron exchanging energy and momentum with plasmons (magnons) represented by \(W^{00}\) (\(W^{IJ}\)). The boundary between layers \(l\) and \(l’\) is indicated by the red dashed line.

Before moving forward, let use interpret the meaning of the resulting self-energy and screened interaction. For simplicity and clarity, we will take
\[
\Lambda^{L L}_{\alpha n,\nu y}(1,4;6) = \delta(1,6) \delta(1,4) \sigma^\mu_{\alpha\gamma} \delta_{\alpha n} \delta_{\nu y},
\]
which yields the commonly employed GW approximation\textsuperscript{43}, where
\[
\Sigma_{\alpha n,\nu t}(1,5) = i\sigma^\mu_{\alpha\gamma} G_{\gamma k,\mu s}(1,5) \sigma^\nu_{\mu\delta} W^{L l}_{ak;bn}(5,1),
\]
and
\[
\chi^{MN}_{0 ef;dg}(7,8) = -i G_{\delta c,\mu s}(7,9) \sigma^N_{\mu s} G_{\nu t,\xi d}(8,7^+;\xi d)^M.
\]

If the hybridization between layer \(l\) and \(l’\) is assumed to be small, as is the case for vertical heterostructures composed of 2D transition metal dichalcogenides,\textsuperscript{44,45} the self-energy can be partitioned into two classes involving layer conserving and layer non-conserving interactions. We illustrate the physical meaning of each case in the following.

Layer Conserving: If a particle, \(G_{\ell\ell,\ell\ell}\), of up-spin and layer \(l\) enters the self energy \(\Sigma_{\ell\ell,\ell\ell}\), the particle exchanges energy and momentum with plasmons (magnons) represented by \(W^{00}\) (\(W^{IJ}\)). If the same particle on layer \(l\) has its spin flipped upon entering the self-energy by a spin operator \(\sigma^I_{\ell\ell}\), a magnon given by \(W^{IJ}\) is emitted. Then upon exiting the self-energy the magnon is reabsorbed, thereby flipping the spin by \(\sigma^J_{\ell\ell}\), and recovering its original spin state. We call these intra-layer plasmons (magnons). This process is illustrated in Fig. 3 (a-b).

Layer Non-Conserving: If a particle, \(G_{\ell\ell,\ell’}\), of up-spin and layer \(l\) enters the self energy \(\Sigma_{\ell\ell,\ell’}\), the screened interaction, \(W\), can ‘flip’ the layer on which the particle is propagating, as seen in Fig. 3 (c-d). As the particle changes layer, it can also emit a plasmon (magnon). On exiting the self energy, the particle is sent back to its layer of origin and reabsorbs the formally emitted plasmon (magnon). We call these interfacial plasmons (magnons), since these bosonic excitations run along the interface.

Since \(G\) is assumed to be nearly diagonal in layer in the weak hybridization limit, the screened interaction only permits two types of polarization bubbles,
\[
\chi^{MN}_{0 \ell\ell,\ell’}(T,8) \text{ and } \chi^{MN}_{0 \ell,\ell\ell}(T,8).
\]

The first bubble completely resides within a single layer and is only able to connect to layer conserving vertices. In contrast, the second polarization bubble is composed of an electron and hole residing on different layers and can only be stimulated by layer non-conserving vertices.
FIG. 4. (color online) Various polarization bubbles and collective modes induced by interlayer coupling. (left panel) Polarization bubbles present in the weak and intermediate hybridization limit connecting to layer conserving or layer non-conserving interactions, indicated by the green and blue vertices, respectively. (right panel) The various bosonic modes predicted as poles of Eq. 37. Intra- and inter-layer plasmons (magnons) are facilitated by layer conserving interactions, while the interfacial modes are generated by layer non-conserving interactions. Mixed modes are generated in the presence of spin-orbit coupling or non-collinear magnetic ordering. The boundary between layers is indicated by the red dashed line.

For intermediate strength layer hybridizations, such as those in the high-temperature superconducting cuprates\textsuperscript{46–49} and the perovskite iridates\textsuperscript{50,51}, an additional inter-layer polarization bubble is possible, \( \chi_{MN \ell k \ell k}^{0} \). (34)

For this bubble, its vertices reside on different layers, allowing connections to layer conserving interactions only. Recently, this inter-layer polarization has been found to contribute to the effective screening of the Ni 3\textit{d} orbitals from the Nd layer in the newly discovered nickelate superconductor NdNiO\textsubscript{2}\textsuperscript{52}. In the strong hybridization limit all remaining polarization bubble configurations are found and play a role in the full screened interaction. These three bubbles are sketched in Fig. 4 (left panel).

As elucidated by Perdew et al.,\textsuperscript{53} the exchange-correlation energy may only account for a small fraction of the total energy of a system, but it includes three key physical ingredients: the exchange energy corrects spurious effects of self-interaction and also maintains the Pauli exclusion principle, while the correlation energy accounts for Coulomb correlation effects in the many-electron environment. However, most importantly, the exchange-correlation energy plays an extremely vital role in the ‘glue’ that binds atoms together to form molecules and solids. Here, the same principle extends to layered systems. The self-energy in Eq. 26a (or Eq. 31 in the \textit{GW} approximation) provides the exchange and correlation corrections to the bare non-interacting system. Due to the explicit layer dependence, \( \Sigma^{ll'} = \Sigma_{x}^{ll'} + \Sigma_{c}^{ll'} \), one finds two types of ‘glue’, one sticking atoms together within the same layer (\( \ell = \ell' \)) and the other adhering the layers together (\( \ell \neq \ell' \)).

C. Charge and Magnetic Response

Interlayer coupling has been shown to play a pivotal role in stabilizing various magnetic orders in a layer dependent manner\textsuperscript{54} and enhancing interlayer-exchange coupling\textsuperscript{55} between 2D atomically thin-films. To analyze the instability of the ground state to various ordered phases and investigate the emergent excitations harbored in layered systems, we must observe its response to an infinitesimal time-dependent external probe \( \pi_{ij}(t) \). The response of the system due to an infinitesimal change in the external field is

\[
\chi_{m;j;ni}(1,2) = \frac{\delta \rho_{nnm}(1)}{\delta \pi_{ij}(2)}.
\]

Using the chain rule we arrive at a recursive relationship for density response due to the perturbation,

\[
\chi_{m;j;ni}(1,2) = \delta \rho_{nm}(1) \frac{\delta \phi_{ab}^{M}(3)}{\delta \phi_{ab}^{M}(2)} + \chi_{0}^{M}(1,2) + \chi_{0}^{IM}(1,3) c_{ML}^{obs}(3,4) \chi_{ij;ni}(4,2).
\]

To dissect the meaning and structure of this response function we start with its tensoral structure. Due to the generalized nature of the density and external electromagnetic field, the response \( \chi_{m;j;ni}^{IJ} \) not only contains the charge (\( I = J = 0 \)) and spin (\( I, J \in \{ x, y, z \} \)) responses, but also their mixture (\( I = 0, J \in \{ x, y, z \} \)). Moreover, each of these responses are indexed by layer (\( mj; ni \)) in which the electrons and holes reside or transition between. This fine-grained, transparent indexing structure...
we have introduced the matrix inverse of the structure of $F$. This is most readily measured by angle resolved photoemission (ARPES). Since the screened interaction ($W$) can be explicitly written in terms of the response function (Eq. 36) the resonant coupling between electrons and intra-layer (interfacial) magnons and plasmons will appear in the ARPES spectra as waterfalls and kinks in the electronic dispersion.

In contrast, the two particle spectral function is found by perturbing the ground state without changing the electron count. Here, the dynamical structure factor of the physical system is measured by scattering photons, neutrons, and electrons off the sample and measuring their change in momentum and energy. Using the fluctuation dissipation theorem the dynamical structure factor is proportional to the imaginary part of the dynamical susceptibility,

$$S(q, \omega) = \frac{\hbar}{\pi} \frac{1}{e^{\beta \hbar \omega} - 1} \text{Im} \chi(q, \omega).$$

or the dielectric function,

$$S(q, \omega) = \frac{\hbar q^2}{4\pi^2 e^2} \frac{1}{(e^{\beta \hbar \omega} - 1)} \text{Im} e^{-1}(q, \omega),$$

where the charge, spin, and layer degrees of freedom have been integrated out. Therefore, intra-layer, inter-layer and interfacial plasmon and magnon peaks should be present in the observed spectra.

### D. Two-Particle Excitations and Excitons

An important feature in the optical spectra of most semi-conductors and 2D materials is the presence of electron-hole bound pairs, or excitons. The importance of excitons in 2D materials stems from their strong binding energies as a result of the highly anisotropic screening environment, leading to many novel devices and applications. If we wish to characterize the various types of excitons that can form within a layered system, we need to examine the excitation spectrum of the 2-particle Green’s function. Our set of exact coupled equations given in Eq. 26a - 26d, do not provide a direct means to 2-particle Green’s function, but rather it is recovered by judiciously combining Eq. 24 and 25. To directly obtain the 2-particle Green’s functions we first extend $\pi_{\mu}^{-1}(1)$ to a two point function, $\pi_{\mu}^{-1}(1,2)$, and consider the infinitesimal change in the single particle Green’s function with respect to the two-point external field. Formally,
The kernel, Green's function where the self-energy is represented by an equation analogous to that for the dressed single-particle propagator $L$.

The recursion relation for the 2-particle propagator is shown in Fig. 5. A diagrammatic representation of the self-consistent Green's function is given in Fig. 5. We should note that this generalization of the single-particle Green's function, we can recover the dressed two-particle propagator $L$.

Firstly, let us comment on the structure of Eq. 40d. The spin indices of $L_0$ and $L$ are the bare and dressed two-particle propagators, respectively. We should note that $L$ is a generalization of $\chi$ where the response function can be recovered by setting

$$L_{aj;bi}(1, 2; 3, 4) = \chi_{aj;bi}(1, 2, 3, 4).$$

A diagrammatic representation of the self-consistent equation for the 2-particle propagator is shown in Fig. 5. The recursion relation for $L$ takes the form of a Dyson's equation analogous to that for the dressed single-particle Green's function where the self-energy is represented by the kernel,

$$\Xi^{NL}_{f1ec}(5, 6; 8, 9) =$$

$$v_{L_{f1ec}}^{NL}(8, 5)\delta(5, 6)\delta(8, 9) + i\sigma_N^{\epsilon} \frac{\delta\Sigma_{\epsilon}\xi \delta(5, 6)}{\delta G^{\epsilon}_{\alpha\epsilon, \beta\epsilon}(8, 9)} \sigma_{\alpha\beta}^{L}.$$  

(42)

Similar to the single-particle Green's function, we can solve for the dressed two-particle propagator $L$ in terms of $L_0$ and the 'self-energy' $\Xi$ to reveal its analytic structure. Formally,

$$L_{aj;bi}(1, 2; 3, 4) = L_{0}^{-1} - \Xi_{aj;bi}(1, 2; 3, 4).$$

where $L_0$ provides the bare two-particle excitation spectrum, and the real and imaginary part of $\Xi$ shifts the electron-hole excitations and accounts for their life time, respectively. In particular, the poles of $L$, occurring when $L_0^{-1} - \Xi = 0$, describe the pairing between and electrons and holes.

Since we wish to characterize spin and layer dependent excitons, as seen in optical spectroscopy, we will work within the GW approximation to better examine the physical content of Eq. 43 and rationalize its indexing structure. This means the kernel (Eq. 42) reduces to the difference between the bare and screened interactions,

$$v_{L_{f1ec}}^{NL}(8, 5)\delta(5, 6)\delta(8, 9) - W_{f1ec}^{NL}(6, 5)\delta(5, 8)\delta(6, 9).$$

(44)
exchange interaction is repulsive. The balance between these to opposing forces guides the creation of bound states.

The spin structure has been analyzed by previous works\(^72\), so we will focus on the layer degrees of freedom. Similar to the charge and magnetic response, we find three unique cases. For the case of intra-layer excitons [Fig. 6 (left panel)], an electron and hole propagating within the same layer \(l\) interact by the screened interaction and bare exchange interaction as follows

\[
W_{ll;ll}^{t} \quad \text{and} \quad v_{ll;ll}^{t}.
\]

Here, the components of \(\Xi\) are very similar to the usual form employed in standard BSE calculations on bulk solids and thin-films, except \(\varepsilon^{-1}\) in \(W\) contains the spin and charge fluctuation contributions from the surrounding layers. If the electron and hole exist on different layers [Fig. 6 (center panel)], \(\Xi\) takes a different form with,

\[
W_{ll;kl}^{t} \quad \text{and} \quad v_{lk;kl}^{t}.
\]

III. INTERLAYER COUPLING IN A BILAYER SYSTEM

As we have shown in the preceding sections interlayer coupling, through either hybridization or electron interactions, can shape and induce various plasmonic, magnonic, and excitonic excitations. In this section we will focus on a few aspects of our findings within a concrete model. Specifically, we will explore the consequence of interlayer coupling on the magnetic ordering instabilities and spin excitations in a simple Hamiltonian for a bilayer square lattice system.

A. Non-Magnetic Hamiltonian

The Hamiltonian for a square lattice bilayer system without electron-electron interactions is explicitly written as

\[
\mathcal{H} = \sum_{lss's'} t_{ss's'}^{l} c_{lss'}^{\dagger} c_{lss'} + \sum_{lss's'} v_{ss's'}^{l} \left( c_{lss'}^{\dagger} c_{l's's'} + \text{h.c.} \right),
\]

where \(c_{ls}^{\dagger}(c_{ls})\) create (destroy) fermions on site \(s\) of layer \(l\) with spin eigenvalues \(\sigma = \pm\). The first term describes the hopping of electrons on each individual layer and the second term allows for hopping between the layers. Since the atomic sites within a given layer are organized over a square lattice, with full translation symmetry, we can Fourier transform the Hamiltonian of each layer. The Hamiltonian of each layer can then be expressed as

\[
H_{k}^{l} = \sum_{s' ss'} \left( \sum_{\langle ss' \rangle} t_{ss's'}^{l} \exp(-ik \cdot R_{ss'}) \right) c_{ls's'}^{\dagger} c_{lss'}
\]

with \(\langle ss' \rangle\) denoting that the sum is taken over successive rings of neighboring lattice sites surrounding site \(s\), and \(R_{ss'}\) is the displacement between lattice sites \(s\) and \(s'\).
Taking the sum out to the fourth nearest neighbor, we find the dispersion of each layer is given by

\[ H_k^l = -2t(\cos(k_x a) + \cos(k_y a)) \]

where \( a \) is the lattice spacing and successive primes (') denote nearest neighbors, next-nearest neighbors, and so on. Finally, the full Hamiltonian including interlayer hybridization, or \textit{bilayer splitting}, is given by

\[ H_{k\sigma} = \begin{pmatrix} H_{k\sigma} & t_{l}^k \\ t_{l}^k & H_{k\sigma} \end{pmatrix} \]

where we have assumed layer 1 and 2 have the same hopping amplitudes and the inter-layer hopping \( t_{l}^k \) can be cast as a momentum dependent bilayer splitting \( t_{l}^k \).

Here, we will use the tight binding parameterization for the bilayer bismuth-based cuprates \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \) (BSCCO) as given in Ref. 75, where the momentum dependent bilayer splitting is defined as

\[ t_{l}^k = -t_{bi} \left( \frac{[\cos(k_x a) - \cos(k_y a)]^2}{4} + a_0 \right) \]

BSCCO, first discovered in 1988,\textsuperscript{76–78} is one of the most studied cuprate compounds, owing to the weak van der Waals-like coupling between the rock-salt \( \text{SrO-BiO}_2\text{-SrO} \) charge reservoir layer and the two \( \text{CuO}_2-\text{Ca-CuO}_2 \) layers that facilitates cleaving for accurate surface studies with angle resolved photoemission spectroscopy\textsuperscript{79–84} and with scanning tunneling spectroscopy.\textsuperscript{85–89} Therefore, making it an interesting compound to examine the effects of inter-layer coupling. The hopping parameters used are given in Table II.

Figure 7 (left panel) shows the single particle spectral function in the absence of inter-layer hybridization. The band dispersion of each layer is degenerate forming a single hole-like cylinder Fermi surface centered at the corners of the Brillouin zone. For finite inter-layer hybridization [Figure 7 (right panel)], the layer basis is reorganized into bonding and anti-bonding pairs, splitting the degenerate energy levels. This produces two cylindrical Fermi surfaces of slightly different doping.

### B. The RPA susceptibilities and magnetic ordering instabilities

To calculate the magnetic instabilities we consider the density-density response

\[ \chi_0^{IJ}(q, -q', \tau) = \langle T \{ \hat{\sigma}^I_j(q, \tau) \hat{\sigma}^J_{l}(q', -\tau) \} \rangle \]

of the generalized density operator

\[ \hat{\sigma}^I(q, \tau) = \sum_k \left( \hat{\psi}^{\dagger}_{k+q\uparrow} \hat{\psi}^{\dagger}_{k+q\downarrow} \right) \sigma^I \left( \hat{\psi}_{k\tau} \hat{\psi}^{\dagger}_{k\tau} \right) \]

where \( \tau \) is the imaginary time, \( q(q') \) is the momentum transfer, \( ijkl \) index the layer, and \( I = 0 \) gives the charge density and \( I = x, y, z \) gives the spin density along each Cartesian direction. If we assume a non-interacting ground state, we can write the non-interacting susceptibilities as,

\[ \chi_0^{IJ} \]
where $\beta = 1/T$ and
\[ G_{0\alpha l,\beta k}(i\omega_n) = \sum_i V_{\alpha l,i}V^*_{\beta k,i}/i\omega_n - \epsilon_i. \] (56)

In the definition of the non-interacting Green’s function (Eq. 56) $\omega_n$ is the Matsubara frequency and $V_{\alpha l,i} = \langle i|\alpha|\beta \rangle$ are the matrix elements connecting the layer-spin and the band spaces found by diagonalizing the Hamiltonian. The retarded susceptibility in Eq. 55b is found by performing the Matsubara frequency summation and by analytically continuing $i\omega_n \rightarrow \omega + i\delta$, for $\delta \rightarrow 0^+$. To calculate the charge and magnetic response functions, we consider Coulomb interactions of the electrons on the same site and between layers in an RPA framework. We distinguish between the layer conserving intra-layer interaction $U$ of electrons on the same atomic site, and an inter-layer interaction $V$. We also take the interfacial layer non-conserving interaction into account in two different configurations, $I$ and $I'$. Where $I$ mimics a Hund’s coupling and $I'$ describes pair-hopping between the layers. Then, by including all crossed diagrams we arrive at the set of layer-dependent interactions,
\[
U = v_{s\sigma s,\sigma}^{I';I} = -v_{s\sigma s,\sigma}^{I;I'} \quad I = v_{s\sigma s,\sigma}^{I;I'} = -v_{s\sigma s,\sigma}^{I';I} \quad (57a)
\]
\[
V = v_{s\sigma s,\sigma}^{I';I} = -v_{s\sigma s,\sigma}^{I;I'} \quad I = v_{s\sigma s,\sigma}^{I;I'} = -v_{s\sigma s,\sigma}^{I';I} \quad (57b)
\]
\[
V = v_{s\sigma s,\sigma}^{I';I} = -v_{s\sigma s,\sigma}^{I;I'} \quad I' = v_{s\sigma s,\sigma}^{I;I'} = -v_{s\sigma s,\sigma}^{I';I} \quad (57c)
\]

Finally, in Table III we expand the interactions in the Pauli basis. Since, the interactions do not contain any spin-flips, only the $v^{00}$, $v^{xx}$, $v^{yy}$, and $v^{zz}$ terms are non-zero. Consequently, the interactions are rotationally invariant for each layer dependent configuration.

| $v^{00}$ | $v^{xx}$ | $v^{yy}$ | $v^{zz}$ |
|---------|---------|---------|---------|
| $U_{\frac{1}{2}}$ | $-U$ | $-U$ | $-U$ |
| $V_{\frac{1}{2}}$ | $-\frac{1}{2}$ | $-\frac{1}{2}$ | $-\frac{1}{2}$ |
| $I_{\frac{1}{2}}$ | $-\frac{V}{2}$ | $-\frac{V}{2}$ | $-\frac{V}{2}$ |
| $I'_{\frac{1}{2}}$ | $-\frac{I'}{2}$ | $-\frac{I'}{2}$ | $-\frac{I'}{2}$ |

TABLE III. Layer components of the interaction in the Pauli basis. ($l \neq k$)

Using the non-interacting single-particle propagator (Eq. 56) in the polarization $\chi_0$ along with taking the bare vertex (Eq. 30) in the layer-dependent electromagnetic response function $\chi_{l,l'}^{II}$ we recover the generalized RPA susceptibilities,
\[
\chi_{l,k;l',k'}^{II}(q,\omega) = (58)
\]
\[
\chi_0 v_{l,k;l'}(q,\omega) + \chi_0 v_{m,k;m'}(q,\omega)\nu_{k,L}^{mm'}\nu_{k',l'}^{lL} \chi_{l,l'}^{II}(q,\omega),
\]
where repeated indices are summed over. For a single-band susceptibility the inclusion of interactions within the RPA approach enhances existing features in the non-interacting susceptibility as the Stoner denominator $1 - U\chi(q,\omega)$ approaches zero. In the case of a multilayer susceptibility, much like the multiorbital case,90 it is not obvious how the different structures in the spin and in the charge susceptibility are changed by the varying $U$, $V$, $I$, and $I'$. To present a simplified and transparent discussion, we varied each parameter while tracking various spin correlation functions.

Figure 8 shows the RPA spin correlations along high-symmetry line in the square Brillouin zone for intra-layer $\langle S_{01}S_{11} \rangle$, inter-layer $\langle S_{00}S_{11} \rangle$, and interfacial $\langle S_{01}S_{01} \rangle$ and $\langle S_{01}S_{10} \rangle$ spin configurations with (dotted lines) and without (solid lines) interlayer hybridization. For $U = 0.7$ eV there is a dramatic enhancement in the spin susceptibilities near $M$ in the intra-layer channel. This enhancement signals an instability toward ($\pi, \pi$) AFM order, which is in agreement with other RPA studies of cuprates91 and the experimentally observed AFM order in the BSCCO parent compound.24 Upon introducing $V$, there is an increase in the spin fluctuations at ($\pi, \pi$) in the $\langle S_{00}S_{11} \rangle$ channel, similar to the effect of $U$. Physically, this inter-layer interaction gives rise to the various AFM orderings along the $c$-axis, e.g. G- and C-type AFM orders. Finally, for finite $I$ and $I'$, spin correlations appear in the $\langle S_{01}S_{01} \rangle$ and $\langle S_{01}S_{10} \rangle$ sectors. This suggests the existence of instabilities towards interfacial magnetic ordering in the BSCCO bilayer system. Following the dotted lines, we find that a finite inter-layer hybridization tends to round-out non-analytic cusps and plateaus near $M$, eliminating competition between various AFM orders, as expected for systems in more than two dimensions.91,92
C. Antiferromagnetic Hamiltonian and Induced Magnetic Order

Proximity effects play a significant role in designing new functional heterostructures with strategically induced phases such as superconductivity, spin-orbit coupling effects, and magnetism.\textsuperscript{97–99} Additionally, the interplay between ordered phases and inter-layer coupling can shape the electronic and magnetic properties of the layered cuprate high-temperature superconductors.\textsuperscript{100,101} To explore the role hybridization and layer-dependent interactions independently play in these proximity effects, we introduce a $Q = (\pi, \pi)$ AFM order into one of the layers in our Hamiltonian for bilayer BSCCO [Eq. 51] in the mean-field. After taking the umklapp processes into account and factoring the electron-electron interactions through an auxiliary field, we arrive at the Hamiltonian in terms of the self-consistent field $m$ and occupation $n_{\sigma}$.

\[
H_{k\sigma} = \begin{bmatrix}
H_{k\sigma} & \text{sign}(\bar{\sigma})\Delta & t^k_0 & 0 \\
\text{sign}(\bar{\sigma})\Delta & H_{k+Q\sigma} & 0 & t^{k+Q}_0 \\
t^k_0 & 0 & H_{k\sigma} & 0 \\
0 & t^{k+Q}_0 & 0 & H_{k+Q\sigma}
\end{bmatrix}
\] (59)

where our wave functions take the Nambu form $\psi^\dagger = (c^\dagger_{1k\sigma}, c^\dagger_{1k+Q\sigma}, c^\dagger_{2k\sigma}, c^\dagger_{2k+Q\sigma})$, $\Delta$ is defined as $\frac{U}{T} (m + m^\dagger) = U Re (m)$, and the constant shift $Un_{\bar{\sigma}}$ is added to the chemical potential. See Appendix A for a detailed derivation of the mean-field Hamiltonian.

**Figure 9.** (color online) The self-consistent magnetic moment $S_{\text{scf}}$ and the induced magnetic moment $S_{\text{induced}}$ as a function of inter-layer hybridization for three different on-site correlation strengths, $U$.

Figure 9 shows the self-consistent spin magnetic moment $S_{\text{scf}}$ on layer 1 and the induced magnetic moment $S_{\text{induced}}$ in layer 2 as a function of bilayer splitting for three different on-site correlation strengths, $U$. For a $U$ of 2.0 eV, $S_{\text{scf}}$ has a maximum of 0.203 $\mu_B$ with no bilayer splitting. For finite $t_{bi}$, three distinct regions are observed. (I) For $0 \leq t_{bi} \leq 0.9$ eV a positive $S_{\text{induced}}$ is produced, reaching a maximum of 0.006 $\mu_B$. (II) When $0.9 \leq t_{bi} \leq 1.3$ eV, $S_{\text{induced}}$ is negative with a minimum of $-0.008$ $\mu_B$. (III) For $1.3 \leq t_{bi}$ eV both $S_{\text{scf}}$ and $S_{\text{induced}}$ are quenched. $S_{\text{induced}}$ decreases for increasing values of $t_{bi}$, with a visible kink in the line shape concomitant with the change in sign of $S_{\text{induced}}$. For larger on-site potentials, the region and moment of negative $S_{\text{induced}}$ is increased and enhanced, respectively.

Physicaly, the increase in $t_{bi}$ can be facilitated by uni-axial compressive strain where the two CuO$_2$ layers are brought into closer proximity, allowing greater wave function overlap. The change in sign of $S_{\text{induced}}$ suggests a change from C-type to G-type AFM order purely due to hybridization. A similar type of behavior is observed in the Bilayer CrI$_3$ where different layer stacking configurations induce AFM or FM coupling between the layers.\textsuperscript{102} Furthermore, the delicate interlayer hopping between IrO$_6$ planes in Sr$_2$IrO$_4$ can be disrupted by an external laser pulse, changing the magnetic symmetry of the system.\textsuperscript{51,103–105}

**Figure 10.** (color online) The single-particle spectral function of a bilayer AFM-Metallic system with and without inter-layer hybridization. (top) Shows the spectral weight for Layer 1 with $(\pi, \pi)$ AFM order and an uncorrelated metallic Layer 2. (bottom) Shows the effect of inter-layer hybridization on the spectra of each layer.

**Figure 10** (top) shows the single-particle spectral function without bilayer splitting. Layer 1 exhibits a 1 eV AFM band gap at the X point and along the $M-\Gamma$ direction in the square Brillouin zone. Since correlations were turned off in layer 2, it is a metal. The bottom panels of Figure 10 display the effect of inter-layer hybridization on the single particle states. Firstly, the spectra of both layers are present in the projected spectral weight of each layer. This is produced by $t_{bi}$ forming bonding (anti-
bonding) pairs between various layer quantum numbers. Moreover, the wide AFM gap of layer 1 is clearly seen, along with a very slight induced gap produced in the originally metallic band of Layer 2.

### D. Spin Waves in the Presence of Interlayer Coupling

To mark the effect of the various layer dependent interactions and bilayer splitting on the spin wave dispersion in Layer 1 and metallic character of layer 2, we calculate the imaginary part of the transverse spin susceptibility $\Im \chi^+_{\omega}(q,\omega)$ in the random phase approximation [Eq. 58]. The results are presented in Figure. 11 and are organized as follows. The right and left panels show $\Im \chi^+_{\omega}(q,\omega)$ with and without bilayer splitting, respectively. The rows in each panel present the data for the various layer-dependent interaction combinations used in the RPA. The specific interactions used are noted on the left. The value of $U$, $V$, $I$, and $I'$, employed are 1.5 eV, 1.0 eV, 1.0 eV, 1.0 eV, respectively. For brevity, only the relevant non-zero tensor components are given.

Figure 11 (left panel, row one) shows the non-zero components of $\Im \chi^+_{\omega}(q,\omega)$ along high symmetry lines in the Brillouin zone for just an on-site potential $U$. In layer 1 (channel 00:00), a clear gapless spin wave dispersion is observed, with its energy minimum at $(\pi, \pi-\delta)$. The spin excitation is clear throughout the Brillouin zone, never entering the continuum and damping out. In contrast, layer 2 (channel 22:22) exhibits a gapless particle-hole continuum, consistent with its metallic band structure.

Furthermore, the interfacial channel 20:02 is non-zero exhibiting a faint gaped spin excitation band at 0.35 eV on top of the particle-hole continuum. When inter-layer and interfacial interactions $V$ (row two) and $I'$ (row four) are introduced, the spectra is relatively unchanged except for an enhancement in the gapped interfacial spin mode in channel 20:02. Lastly, $I'$ generates a new non-zero interfacial matrix element (channel 22:00) with a similar structure to that of channel 20:02.

Interestingly, a finite layer non-conserving interaction $I$ (row three) dramatically damps the magnon dispersion in layer 1 by mixing in the metallic particle-hole continuum of layer 2. Moreover, the zero of the dispersion is shifted to surrounding $\Gamma$ and $X$. Since $I$ mixes the excitation spectrum of layer 1 and 2, a magnon dispersion is now induced in layer 2, similar to layer 1. Additionally, a new interfacial non-zero channel 02:02 is found, displaying characteristic features of layer 1 and 2.

If a finite bilayer splitting is included (right panel), the spectrum of $\Im \chi^+_{\omega}(q,\omega)$ is very similar to that of the isolated case, except for a few key aspects. The magnon dispersion in layer 1 [seen in row one, two, and four] now has its minimum at the $M$ point in the Brillouin zone. Moreover, along $X - \Gamma$ an avoided crossing appears and the magnon mode becomes incoherent near $\Gamma$, due to the admixture of metallic features from layer 2. In the interfacial channel 20:02, the spin wave band from layer one is clearly seen extending into the continuum.

In summary, layer-dependent interactions are able to modify magnetic ordering tendencies and magnon dispersions, and induce collective modes in neighboring layers all without inter-layer hybridization. This illustrates
the key role these interactions play in designing and manipu-
ling various charge and magnetic phases and exci-
tations in 2D atomically-thin film heterostructures and
layered correlated compounds, such as the perovskite
transition-metal oxides.

IV. CONCLUDING REMARKS

We have derived a generalization of Hedin’s equations
for a layered system with arbitrarily strong interlayer
coupling. Our approach was made sufficiently general
to accommodate non-local interactions and non-equilibrium
quantum phases through the Keldysh and Schwinger
techniques. We have thus opened a pathway for examin-
ing the interplay of charge, spin, orbital and layer degrees
of freedom in layered heterostructures and their phase di-
agrams including relativistic magnetic interactions, along
with the evolution of electronic spectra with pressure and
doping.

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Appendix A: Mean-Field Interactions and AFM

In order to include staggered AFM order on the atomic
sites, we include an on-site Hubbard interaction term
to the Hamiltonian of Eq. 51. Specifically, the double-
occupancy energy penalty, \( U \), is placed on the single
effective band crossing the Fermi level. The Hubbard in-
teraction can be written in momentum space as

\[
\frac{U}{2} \sum_{\sigma, kk'/Q} \left( c_{k\sigma}^\dagger c_{k'\sigma} c_{k'\bar{\sigma}} c_{k\bar{\sigma}} + c_{k+Q\sigma}^\dagger c_{k\sigma} c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} \right). \tag{A1}
\]

where \( \bar{\sigma} \) denotes \(-\sigma\). Due to momentum conservation,
the interaction depends on both the crystal momentum,
\( k(k') \), of the electrons and the momentum transferred,
\( Q \), during the interaction. The momentum transfer gives
rise to umklapp processes where electrons can scatter to
neighboring Brillouin zones, which are the key for de-
scribing various density-wave instabilities. Here we take
\( Q = (\pi, \pi) \) following the experimentally observed AFM
order. Thus, the full single-band Hamiltonian is

\[
\mathcal{H} = \sum_{\sigma} \sum_k \left( H_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + H_{k+Q\sigma} c_{k+Q\sigma}^\dagger c_{k+Q\sigma} \right)
- \mu \sum_{\sigma} \sum_k (\hat{n}_{k\sigma} + \hat{n}_{k+Q\sigma})
+ \frac{U}{2} \sum_{\sigma, kk'} c_{k\sigma} c_{k'\sigma} c_{k'\bar{\sigma}} c_{k\bar{\sigma}} + c_{k+Q\sigma} c_{k\sigma} c_{k'\bar{\sigma}} c_{k'\bar{\sigma}}. \tag{A2}
\]

where \( H_k \) is written in terms of \( Q \) explicitly by restricting
\( k(k') \) to the smaller AFM Brillouin zone. We now rewrite
the interaction in terms of the mean-field and expand the
number operator in terms of fluctuations away from the
mean electron count per state, \( \langle n_{k\sigma} \rangle \):

\[
\langle n_{k\sigma} \rangle = \langle n_{k\sigma} \rangle + (n_{k\sigma} - \langle n_{k\sigma} \rangle) \tag{A3}
= \langle n_{k\sigma} \rangle + \delta_{n\sigma},
\]

where \( \delta_{n\sigma} \) is the fluctuation away from \( \langle n_{k\sigma} \rangle \). We substi-
tute into the interaction of Eq. A2 assuming fluctuations
are small, \( \delta_{n\sigma} \approx 0 \), giving

\[
\frac{U}{2} \sum_{\sigma, kk'} \left( c_{k\sigma}^\dagger c_{k'\sigma} c_{k'\bar{\sigma}} c_{k\bar{\sigma}} + c_{k+Q\sigma}^\dagger c_{k\sigma} c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} \right) + \langle c_{k+Q\sigma} c_{k\sigma} c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} \rangle \tag{A4}
\]

In order to treat the various matrix elements in Eq. A4,
we consider the average charge and spin densities as a
function of momentum transfer, \( q \),

\[
\langle \rho(q) \rangle = \sum_k \left( \left( c_{k+q\uparrow}^\dagger c_{k+q\downarrow} \right)^2 \right) \tag{A5a}
= \sum_k \langle c_{k+q\uparrow}^\dagger c_{k\uparrow} \rangle + \langle c_{k+q\downarrow} c_{k\downarrow} \rangle
= N_\epsilon \delta_{q,0},
\]

\[
\langle S^z(q) \rangle = \frac{1}{2} \sum_k \left( \langle c_{k+q\uparrow}^\dagger c_{k+q\downarrow} \rangle - \langle c_{k+q\downarrow}^\dagger c_{k+q\uparrow} \rangle \right) \tag{A5b}
= \frac{1}{2} \sum_k \langle c_{k+q\uparrow} c_{k\uparrow} \rangle - \langle c_{k+q\downarrow} c_{k\downarrow} \rangle.
\]

Therefore, for \( q = Q = (\pi, \pi) \),

\[
\langle \rho(Q) \rangle = \sum_k \left( \langle c_{k+Q\uparrow} c_{k\uparrow} \rangle + \langle c_{k+Q\downarrow} c_{k\downarrow} \rangle \right) \tag{A6}
= 0,
\]

which implies,

\[
\langle c_{k+Q\uparrow} c_{k\uparrow} \rangle = - \langle c_{k+Q\downarrow} c_{k\downarrow} \rangle. \tag{A7}
\]

Also, by hermiticity we have the equivalence,

\[
\langle c_{k+Q\sigma}^\dagger c_{k\sigma} \rangle = \langle c_{k\sigma}^\dagger c_{k+Q\sigma} \rangle. \tag{A8}
\]
Using the relation in Eq. A7 we find \( \langle S^z(Q) \rangle \),

\[
\langle S^z(Q) \rangle = \frac{1}{2} \sum_k \left( c_{k+Q+}^\dagger c_{k} - c_{k+Q-}^\dagger c_{k} \right) = \sum_k \left( c_{k+Q+}^\dagger c_{k} \right).
\]

(A9)

The preceding relations allow us to cast staggered magnetization and electron density as,

\[
m = \sum_k \left( c_{k+Q+}^\dagger c_{k} \right) = \sum_k \left( c_{k+Q-}^\dagger c_{k} \right),
\]

(A10a)

\[
n_\sigma = \sum_k \left( c_{k\sigma}^\dagger c_{k\sigma} \right).
\]

(A10b)

Inserting these definitions and simplifying we arrive at the Hamiltonian in terms of the self-consistent field \( m \) and occupation \( n_\sigma \),

\[
H_{k\sigma} = \begin{pmatrix}
H_{k\sigma} + U n_\sigma & \text{sign}(\bar{\sigma}) \Delta \\
\text{sign}(\bar{\sigma}) \Delta & H_{k+Q\sigma} + U n_\sigma
\end{pmatrix}
\]

where our wave functions take the Nambu form \( \Psi = (c_{k\sigma}, c_{k+Q,\sigma}^\dagger) \) and \( \Delta \) is defined as \( \frac{U}{2} (m + m^\dagger) = U \text{Re} (m) \).

To self consist \( m \) and \( n_\sigma \), their expectation value can be written in terms of the diagonalized system. Let the quasiparticle creation (\( \gamma_{k\sigma}^\dagger \)) and annihilation (\( \gamma_{k\sigma} \)), operators in the diagonalized system be defined as

\[
c_{k\sigma} = \sum_i V_{\sigma,i} \gamma_{k\sigma i} \quad \text{and} \quad c_{k\sigma}^\dagger = \sum_i \gamma_{k\sigma i}^\dagger (V_{\sigma,i})^\dagger.
\]

(A12)

where \( i \) indexes the bands. Therefore \( m \) and \( n_\sigma \) are given by

\[
n_\sigma = \sum_i \sum_k \left( (V_{\sigma,i})^\dagger V_{\sigma,i} + (V_{\sigma,i+Q})^\dagger V_{\sigma,i+Q} \right) f(\epsilon_{k\sigma i}),
\]

(A13a)

\[
m = \sum_i \sum_k \left( (V_{\sigma,i}^k) V_{\sigma,i}^k + (V_{\sigma,i}^{k+Q}) V_{\sigma,i}^{k+Q} \right) f(\epsilon_{k\sigma i}).
\]

(A13b)

for \( k \) in the AFM Brillouin zone and \( f \) being the Fermi function. The self-consistently obtained values of the expectation value of \( m \) and \( n_\sigma \) are calculated within a tolerance of \( 10^{-5} \) at a temperature of 0.001 K.

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By the chain rule

\[ \frac{\delta G_{\mu_\alpha \nu_\beta}(A_4,5)}{\delta \pi_{\mu_\alpha}(3)} = \frac{\delta G_{\mu_\alpha \nu_\beta}(A_4,5)}{\delta \Phi_{\alpha_\beta}(6)} \frac{\delta \Phi_{\alpha_\beta}(6)}{\delta \pi_{\mu_\alpha}(3)} \]

where the first and second terms contribute to the vertex and screened interaction, respectively.

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To find the dispersion of the collective excitations, we first use the fact that

\[ [1 - F] = \sum \limits_i [V_i V_i^\dagger - V_i^\dagger V_i] \]

\[ = \sum \limits_i V_i^\dagger [1 - \lambda_i] V_i \]

where \( \lambda_i \) is the \( i \)th eigenvalue of \( F \). Thus when \( \lambda_i = 1 \), \( 1 - F \) is singular and a pole is produced in \( \chi_{\mu_\alpha \nu_\beta}^{ij} \). Therefore the dispersion of the collective mode is given by tracing energy versus momentum where \( \lambda_i = 1 \). This approach is similar to those discussed in Refs. 108–112.

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