

Supplementary Information for “Evolution of spin excitations from bulk to monolayer FeSe”

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I. BILAYER HUBBARD MODEL CALCULATIONS

We modeled the spin excitation spectrum of bulk FeSe and monolayer FeSe (FeSe/STO) using a two-orbital Hubbard model defined on a two-dimensional square lattice with $N = L^2$ unit cells, where L is the linear size of the system. The Hamiltonian is given by

$$\mathcal{H} = \mathcal{H}_{\mathcal{K}} + \mathcal{H}_{\mathcal{U}}, \quad (1)$$

where

$$\begin{aligned} \mathcal{H}_{\mathcal{K}} &= t \sum_{\substack{\langle i,j \rangle \\ \sigma, \alpha}} \left(c_{i,\alpha,\sigma}^\dagger c_{j,\alpha,\sigma} + \text{H.c.} \right) + t_{\perp} \sum_{i,\sigma} \left(c_{i,1,\sigma}^\dagger c_{i,2,\sigma} + \text{H.c.} \right) - \mu \sum_{i\sigma,\alpha} n_{i,\alpha,\sigma}, \text{ and} \\ \mathcal{H}_{\mathcal{U}} &= U \sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow}. \end{aligned}$$

Here, $c_{i,\alpha,\sigma}^\dagger$ ($c_{i,\alpha,\sigma}$) creates (annihilates) a spin σ electron in the unit cell i and orbital α , t is the intra-orbital hopping integral, $\langle i, j \rangle$ indicates a sum over nearest-neighbors, $t_{\perp}$ is the inter-orbital hopping integral between the orbitals on the same site, $n_{i,\alpha,\sigma} = c_{i,\alpha,\sigma}^\dagger c_{i,\alpha,\sigma}$ is the number operator, and U is the Hubbard interaction, which acts only between electrons in the same orbital. The total particle number is controlled by the chemical potential term μ . Because of the orbital symmetry of this Hamiltonian (the intra-orbital terms are the same for the two orbitals), and the restriction to only a local intra-orbital Hubbard interaction, one may also regard this model as a bilayer Hubbard model with layers $\alpha = 1, 2$.¹ The kinetic energy term $\mathcal{H}_{\mathcal{K}}$ can then be diagonalized and rewritten in terms of a bonding $k_z = 0$ and anti-bonding $k_z = \pi$ basis.

Throughout, we use $t = 1$ as the unit of energy and vary $U = 8t$, $t_{\perp}$, and the filling n to control the electronic structure of the system. We simulated Eq. (1) using the dynamical cluster approximation (DCA) method, where the bulk lattice system is mapped onto a periodic finite-size cluster embedded in a mean-field. The effective cluster problem was solved self-consistently by means of a continuous-time auxiliary field (CTAUX) quantum Monte Carlo method.²⁻⁴

The model's electronic structure is summarized in Fig. S1, which was obtained by extracting the single particle spectral function $A(\mathbf{k}, \omega)$ from the QMC data using the Maximum Entropy (MaxEnt) method.⁵ The first plot in the Fig. S1 shows the electronic structure of the model with $U = 8t$, $t_{\perp} = 2.0t$, and a filling $n = 1.05$. For this choice of parameters we

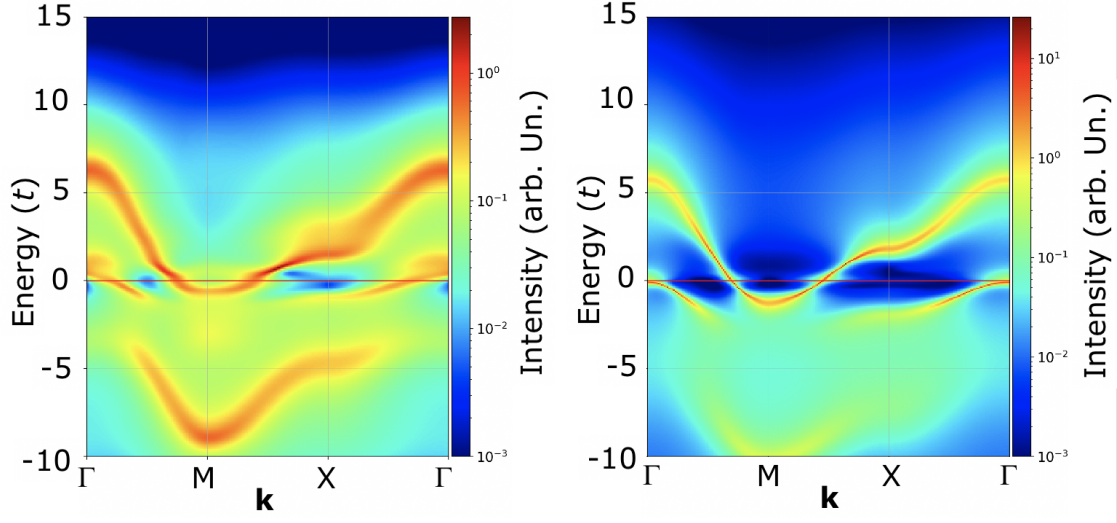


FIG. S1: Left: $A(\mathbf{k}, \omega)$ obtained by setting $U/t = 8$, $t_{\perp} = 2$, $n = 1.05$ at $T = 0.2$ on a 4×4 lattice. In this case, both the electron-like band at the M point and the hole-like band at the Γ point cross the Fermi level. Right: the single particle spectral function $A(\mathbf{k}, \omega)$, obtained by setting $U/t = 8$, $t_{\perp} = 2.5$, $n = 1.15$ at $T = 0.125$ on a 4×4 lattice. In this case, the Fermi surface has an electron-like pocket at the M point and an incipient hole-like band near the Γ point.

51 obtain a band structure that qualitatively resembles that of bulk FeSe where the hole band
 52 lies above the Fermi level at the Γ point, as shown in the right panel of Fig. S1. In contrast,
 53 for $t_{\perp} = 2.5t$, $U = 8t$, and $n = 1.15$, we obtain a band structure qualitatively similar to
 54 FeSe/STO. Specifically, we observe an electron pocket at the M point, $\mathbf{k}_{\parallel} = (\pi, \pi)$, resulting
 55 from the bonding $\mathbf{k}_z = (0, 0)$ band and an incipient hole band with $k_z = \pi$ lying slightly
 56 below the Fermi level at the Γ point ($\mathbf{k}_{\parallel} = 0$).

57 To determine the magnetic excitation spectrum, we calculated the dynamical spin sus-
 58 ceptibility

$$\chi_{\alpha, \beta}^s(\mathbf{r}_i - \mathbf{r}_j, \tau) = \langle \mathcal{T}_{\tau} S_{i, \alpha}^z(\tau) S_{j, \beta}^z(0) \rangle, \quad (2)$$

59 where $S_{i, \alpha}^z(\tau) = \frac{1}{2} [n_{i, \alpha, \uparrow}(\tau) - n_{i, \alpha, \downarrow}(\tau)]$ is the z -component of the spin on orbital α at site $\mathbf{r}_i$
 60 at time τ .

61 Additionally, to investigate the charge excitations, we calculated the dynamical charge
 62 susceptibility

$$\chi_{\alpha, \beta}^c(\mathbf{r}_i - \mathbf{r}_j, \tau) = \langle \mathcal{T}_{\tau} n_{i, \alpha}(\tau) n_{j, \beta}(0) \rangle, \quad (3)$$

where $n_{i,\alpha}(\tau) = \frac{1}{2} [n_{i,\alpha,\uparrow}(\tau) + n_{i,\alpha,\downarrow}(\tau)]$ is the density on orbital α at site $\mathbf{r}_i$ at time τ . A Fourier transform to reciprocal space and Matsubara frequencies gives the momentum and frequency dependent spin and charge susceptibilities

$$\chi_{s/c}(\mathbf{Q}, i\omega_m) = \sum_{\mathbf{r}, \alpha, \beta} \int_0^\beta d\tau \chi_{\alpha, \beta}^{s/c}(\mathbf{r}, \tau) e^{i\mathbf{Q}_{\parallel} \mathbf{r}} e^{iQ_{\perp}(\alpha - \beta)} e^{i\omega_m \tau}. \quad (4)$$

Here, $\mathbf{Q} = (\mathbf{Q}_{\parallel}, Q_{\perp})$ has both an intra-band component $Q_{\perp} = 0$ that scatters within the bonding $k_z = 0$ and anti-bonding $k_z = \pi$ bands, respectively, and an inter-band component $Q_{\perp} = \pi$ that scatters between the bands. As with the spectral functions, we obtained the dynamical susceptibilities by analytically continuing the spin and charge susceptibilities to the real axis using MaxEnt.

Figures S2 and S3 show the frequency and temperature dependence of the imaginary part of the spin and charge susceptibilities $\chi_s''(\mathbf{Q}, \omega)$ and $\chi_c''(\mathbf{Q}, \omega)$ for the case with two Fermi pockets and the case with an incipient band, respectively. In both figures, the left panel presents results for the intra-band component whereas the right panel shows results for the inter-band component of the momentum scattering vectors. In both cases, the amplitudes of the susceptibilities are largest for the inter-band scattering ($Q_{\perp} = \pi$) when compared to the intra-band case ($Q_{\perp} = 0$). In the first case, where the model has two Fermi pockets (Fig. S2), we observe a prominent peak that disperses to zero energy as the temperature decreases for the momentum scattering vector with components $\mathbf{Q}_{\parallel} = (\pi, \pi)$ and $Q_{\perp} = \pi$ in the spin channel. This behavior is due to the perfect nesting between the bonding and anti-bonding bands. Conversely, the spin excitations at this scattering vector are gapped when the hole pocket lies below the Fermi level, as in the incipient band case (Fig. S3). Whereas we observe gapped excitations in the charge channel shifted to the higher frequencies compared to the spin channel.

To provide evidence for the different types of dispersion between the two-band and incipient band models, we show another set of results for a different parameter regime demonstrating that our findings are not parameter dependent. In the first plot in Fig. S4, we report the electronic structure of the model with $U = 6t$, $t_{\perp} = 2.2t$, and a filling $n = 1.05$. The obtained band structure resembles the plot in Fig. S1 representing well the bulk FeSe in which there is an electron pocket at the M point and hole pocket at the Γ point. In the right plot of Fig. S4, the electronic structure of the model with $U = 6t$, $t_{\perp} = 2.8t$, and a filling

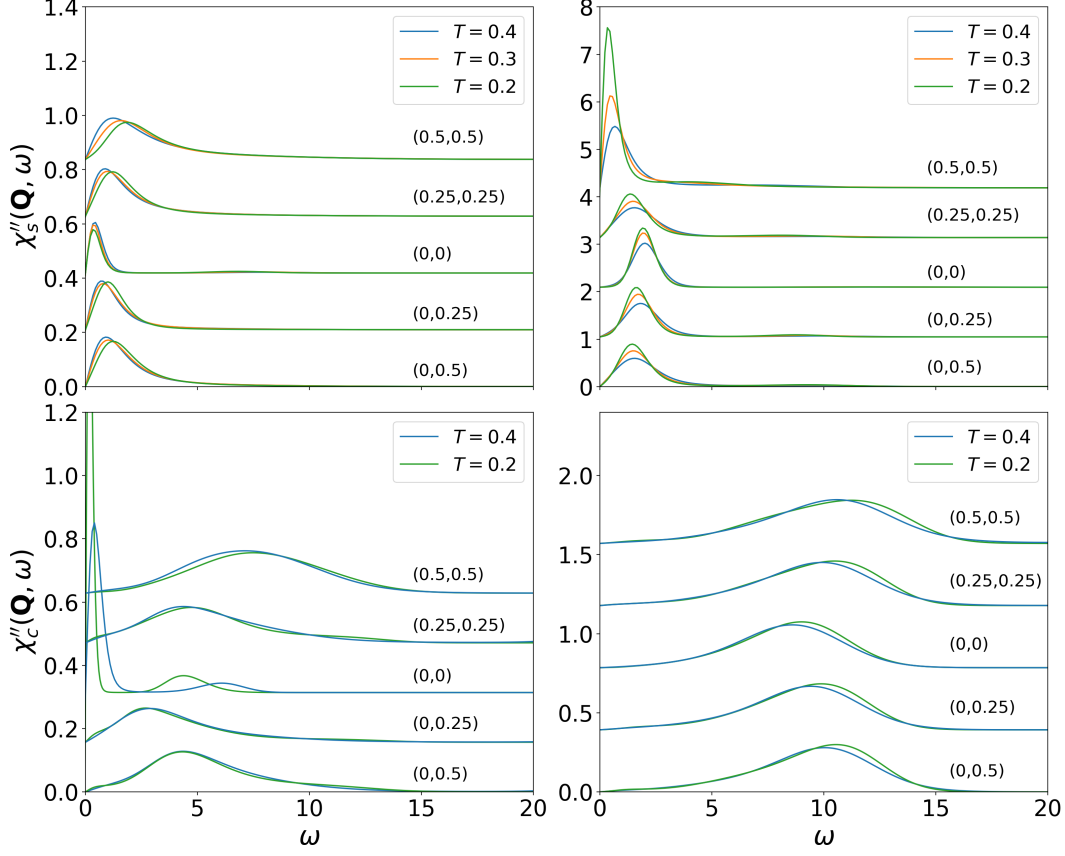


FIG. S2: Two band model. Frequency dependence of charge and spin susceptibility $\chi''_{c,s}(\mathbf{Q}, \omega)$ along high symmetry directions for different temperatures at $U/t = 8$. Results are shown here for the case with two Fermi pockets. On the top row we report the spin susceptibility and on bottom row the charge susceptibility. We report the calculations for $Q_{\perp} = 0$ or intra-band (left panel) and $Q_{\perp} = \pi$ or inter-band (right panel).

⁹² $n = 1.15$ is shown. In Fig. S4, we observe an electron pocket at the M point and an incipient
⁹³ hole band lying slightly below the Fermi level at the Γ point representing the FeSe/STO.
⁹⁴ From this parameters set, we have also calculated the spin and charge susceptibilities which
⁹⁵ are presented in the figures S5 and S6. We observe the same qualitative behaviours (acoustic
⁹⁶ spin susceptibility in the two band model vs optical spin susceptibility in the incipient band
⁹⁷ case) of the spin and charge susceptibilities both for the bulk FeSe and the FeSe/STO as in
⁹⁸ the first data set presented above.

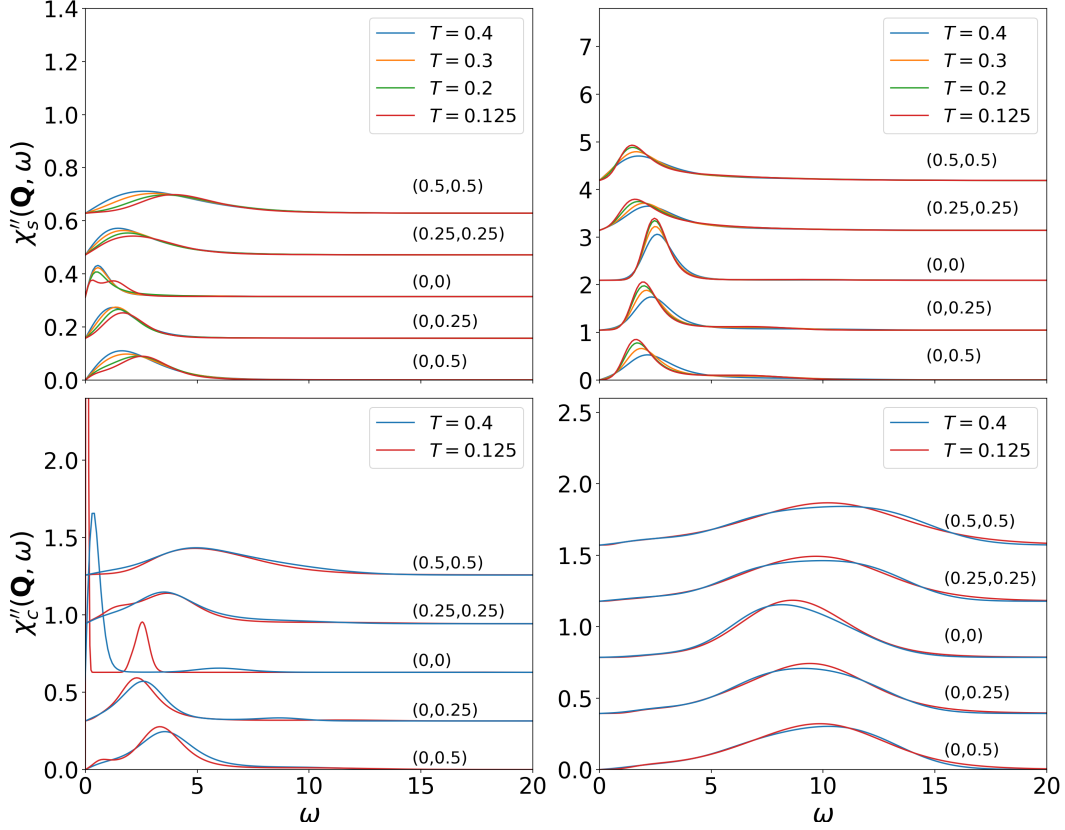


FIG. S3: Incipient band model. Frequency dependence of charge and spin susceptibility $\chi''(\mathbf{Q}, \omega)$ along high symmetry directions for different temperatures at $U/t = 8$. Results are shown here for the incipient band case. On the top row we report the spin susceptibility and on bottom row the charge susceptibility. We report the calculations for $Q_{\perp} = 0$ or intra-band (left panel) and $Q_{\perp} = \pi$ or inter-band (right panel).

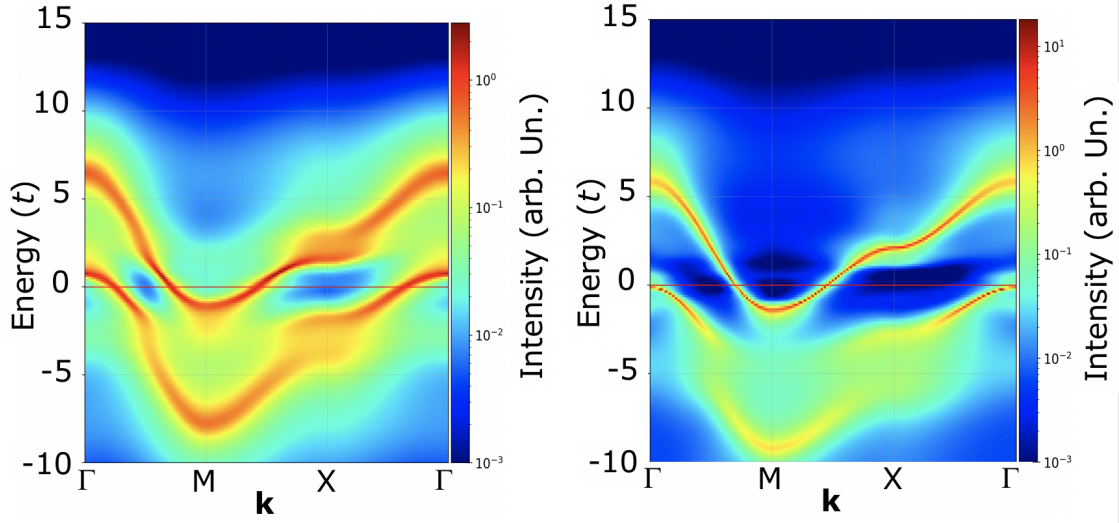


FIG. S4: Left: $A(\mathbf{k}, \omega)$ obtained by setting $U/t = 6$, $t_{\perp} = 2.2$, $n = 1.05$ at $T = 0.2$ on a 4×4 lattice. In this case, both the electron-like band at the M point and the hole-like band at the Γ point cross the Fermi level. Right: single particle spectral function $A(\mathbf{k}, \omega)$, obtained by setting $U/t = 6$, $t_{\perp} = 2.8$, $n = 1.15$ at $T = 0.2$ on a 4×4 lattice. In this case, the Fermi surface has an electron-like pocket at the M point and an incipient hole-like band near the Γ point.

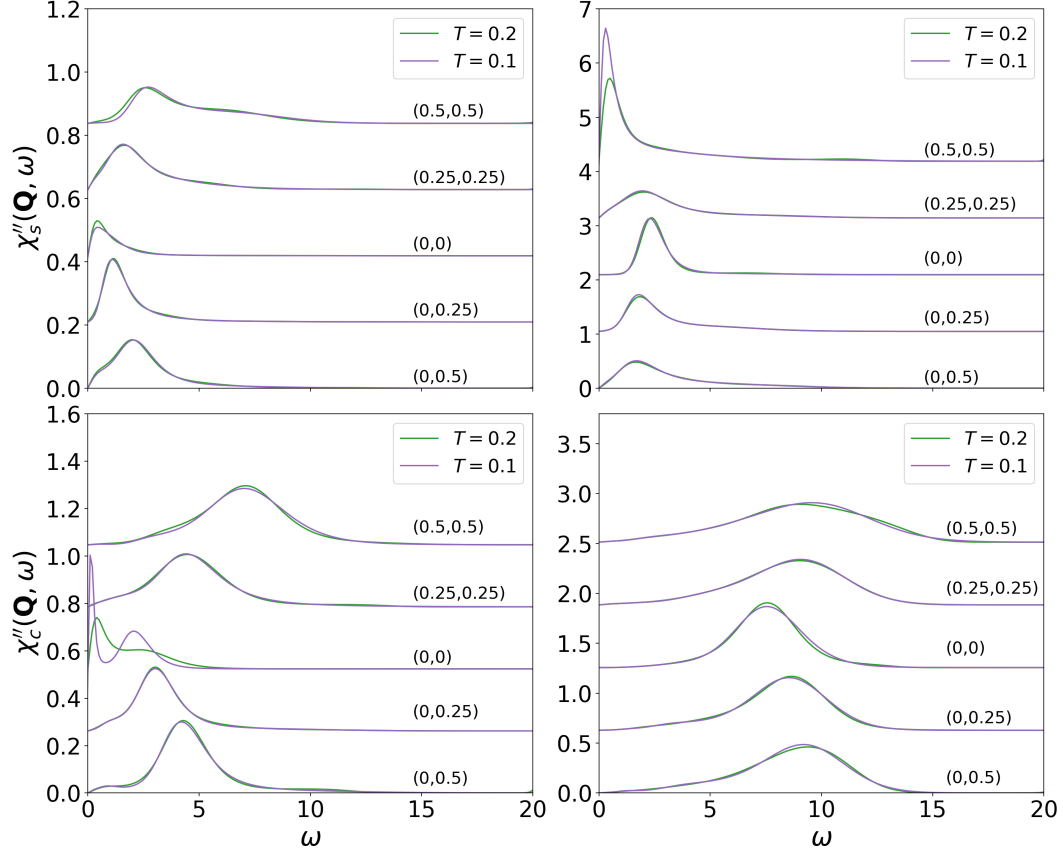


FIG. S5: Frequency dependence of the susceptibility $\chi''(\mathbf{Q}, \omega)$ of the two band model along high symmetry directions for different temperatures at $U/t = 6$. On the top row we report the spin susceptibility and on bottom row the charge susceptibility. We report the calculations for $Q_{\perp} = 0$ or intra-band (left panel) and $Q_{\perp} = \pi$ or inter-band (right panel).

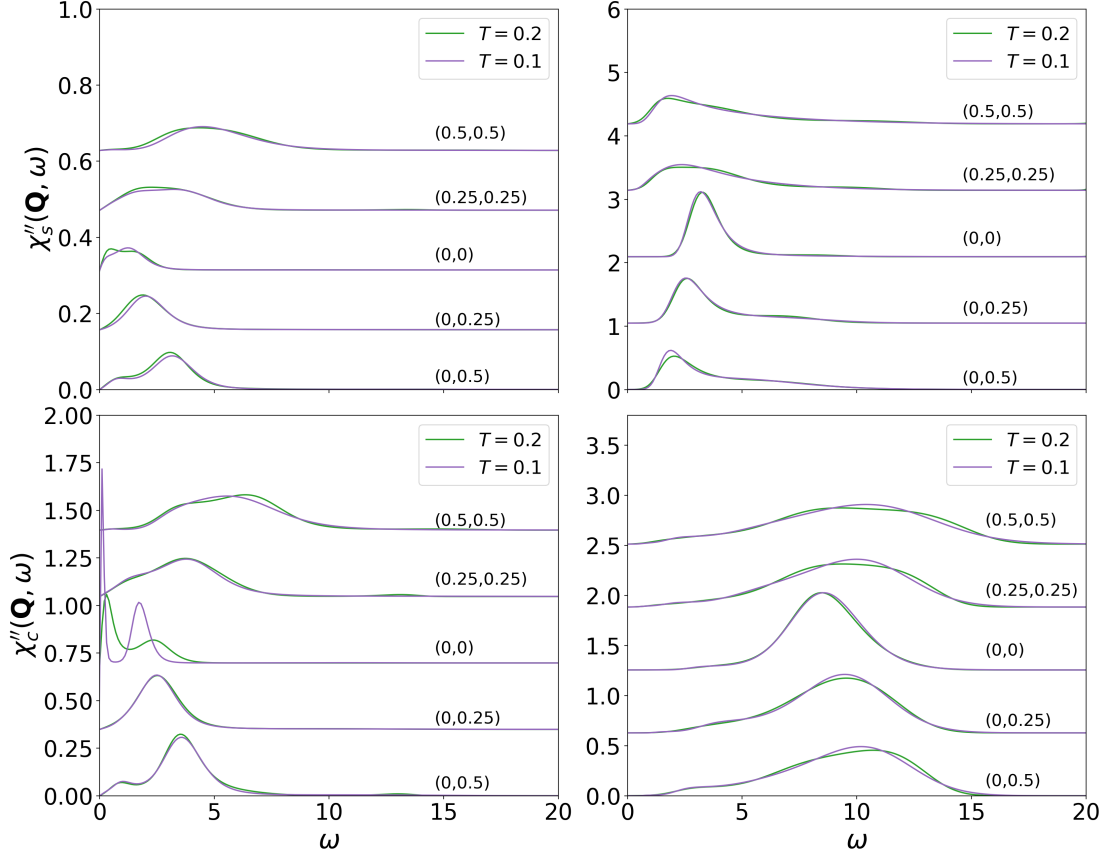


FIG. S6: Frequency dependence of the susceptibility $\chi''(\mathbf{Q}, \omega)$ of the incipient band model along high symmetry directions for different temperatures at $U/t = 6$. On the top row we report the spin susceptibility and on bottom row the charge susceptibility. We report the calculations for $k_\perp = 0$ or intra-band (left panel) and $k_\perp = \pi$ or inter-band (right panel).

99 II. A SIMPLIFIED LADDER MODEL

100 To verify the robustness of the interpretation of our 2D bilayer model results, we addi-
 101 tionally performed exact diagonalization calculations of a small 6×2 Hubbard ladder with
 102 periodic boundary conditions. This system is a 1D analog of the model considered in the
 103 main text, and is small enough that we can explicitly compute the RIXS intensity with
 104 reasonable momentum resolution.

105 The Hamiltonian of the 1D Hubbard ladder is given by

$$H = -t \sum_{l,\alpha,\sigma} \left(c_{l,\alpha,\sigma}^\dagger c_{l+1,\alpha,\sigma} + \text{H.c.} \right) - t_\perp \sum_{l,\sigma} \left(c_{l,0,\sigma}^\dagger c_{l,1,\sigma} + \text{H.c.} \right) + U \sum_{l,\alpha} \hat{n}_{l,\alpha,\uparrow} \hat{n}_{l,\alpha,\downarrow}, \quad (5)$$

106 where $c_{l,\alpha,\sigma}^\dagger$ ($c_{l,\alpha,\sigma}$) creates (annihilates) a spin σ ($=\uparrow, \downarrow$) hole on leg α ($= 0, 1$) of rung l , t
 107 is the nearest-neighbor hopping integral along the leg, $t_\perp$ is the nearest-neighbor along the
 108 rung, and U is the on-site Hubbard repulsion.

109 To understand the model's dynamical properties, we computed its single-particle spectral
 110 function $A(k, \omega)$, dynamical spin structure factor $S(\mathbf{q}, \omega)$, x-ray absorption spectra $I_{\text{XAS}}(\omega)$,
 111 and RIXS intensity $I_{\text{RIXS}}(\mathbf{q}, \omega)$ in the $\Delta S = 1$ channel, which dominates the RIXS spectra
 112 at the transition metal L -edge. As in the bilayer model, we can partition $\mathbf{q} = (q_\parallel, q_\perp)$, where
 113 $q_\parallel$ is the momentum transfer along the ladder leg and $q_\perp = 0$ and π selects the intra- and
 114 interband scattering processes.¹² Throughout this section, we work in electron language and
 115 diagonalize Eq. (5) in the $(n_\uparrow = 7, n_\downarrow = 6)$ sector to determine the ground state $|g\rangle$, which
 116 is equivalent to an electron doping of ≈ 1.08 electrons per orbital. The model parameters
 117 are $t = 1$, $U = 4t$, and $t_\perp = 3t/2$, $2t$, and $3t$.

118 A. The single-particle spectral function

119 We begin by examining the single-particle spectral function. This quantity can be di-
 120 vided into contributions from electron addition (+) and removal (−) processes $A(k, \omega) =$
 121 $A_+(k, \omega) + A_-(k, \omega)$, with

$$A_\pm(k, \omega) = \sum_{f,\alpha,\sigma} |\langle f | c_{k,\alpha,\sigma}^\pm | g \rangle|^2 \delta(E_f - E_g - \omega) \quad (6)$$

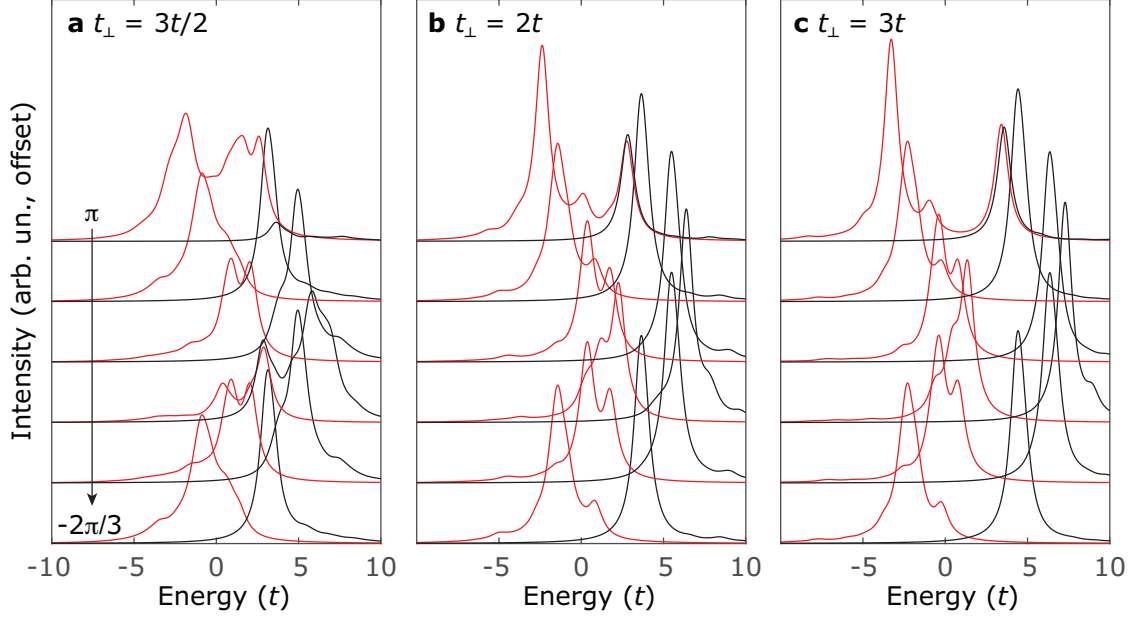


FIG. S7: Single-particle spectral function of the ladder model. The single-particle spectral function $A(k, \omega)$ for the doped ladders, divided into their addition (black lines) and removal (red lines) spectra.

Here, $|g\rangle$ and $|f\rangle$ are the ground and final states with energy E_g and E_f , respectively, and

$$c_{k,\alpha,\sigma}^+ \equiv c_{k,\alpha,\sigma}^\dagger = \frac{1}{\sqrt{N}} \sum_l e^{-ikR_l} c_{l,\alpha,\sigma}^\dagger,$$

$$c_{k,\alpha,\sigma}^- \equiv c_{k,\alpha,\sigma} = \frac{1}{\sqrt{N}} \sum_l e^{-ikR_l} c_{l,\alpha,\sigma},$$

where R_l is the position of rung l along the ladder direction.

Results for the addition (black lines) and removal (red lines) spectra are plotted in Fig. S7(a)-(c) for $t_\perp = 3t/2$, $2t$, and $3t$, respectively. In the absence of any interactions, the non-interacting bands are $\epsilon_\pm = -2t \cos(ka) \mp 2t_\perp$. We would then expect to have one band completely occupied for a slightly electron-doped system while the second band only has its states at lowest energies filled. Moreover, this band should be located approximately $4t_\perp$ above the first. While a non-zero $U = 4t$ will further dress the electronic structure, we have found that this non-interaction picture provides a useful framework for understanding our results.

Figure S7(c) shows results for $A(k, \omega)$ with $t_\perp = 3t$. Here, we see two dispersing bands. The first band consists of fully occupied states located at energies $\omega < 3t$. The second is located at higher energies and has nearly degenerate electron removal and addition states

located at its band bottom centered at $q = \pi$. Decreasing $t_\perp$ to $2t$ produces a qualitatively consistent spectral function but pushes the two bands closer to one another, as shown in Fig. S7(b). As $t_\perp$ is decreased further to $3t/2$, the first removal state at $q = 0$ becomes degenerate with an electron addition state. This observation reflects the fact that the top of the lower-energy band now lies above the Fermi level. These results show that $t_\perp = 3t/2$ produces two bands crossing the Fermi level, with hole-like and electron-like bands centered at $q = 0$ and π , respectively. Conversely, $t_\perp \geq 2t$ produces an incipient band scenario, where one band is filled, and the other has a small electron-like Fermi surface centered at the zone boundary. These cases are analogous to the 2D results shown in the main text.

B. The RIXS response and its comparison with the dynamical spin structure factor

Next, we turn our attention to the RIXS response, which is computed within the full Kramers-Heisenberg formalism. Specifically, the RIXS intensity is given by

$$I_{\text{RIXS}}(q_\parallel, \omega) = \sum_f \left| \sum_n \frac{\langle f | D_{\mathbf{k}_{\text{out}}}^\dagger | n \rangle \langle n | D_{\mathbf{k}_{\text{in}}} | g \rangle}{E_g + \omega_{\text{in}} - E_n + i\Gamma} \right|^2 \delta(E_f - E_g + \omega), \quad (7)$$

where $|g\rangle$, $|n\rangle$, and $|f\rangle$ are the initial, intermediate, and final states of the scattering process, respectively, E_g , E_n , and E_f are their corresponding energies, $\mathbf{k}_{\text{in}}$ and ω_{in} ($\mathbf{k}_{\text{out}}$ and ω_{out}) are the momentum and energy of the incoming (outgoing) photons, $q_\parallel = (\mathbf{k}_{\text{out}} - \mathbf{k}_{\text{in}})_\parallel$ and $\omega = \omega_{\text{out}} - \omega_{\text{in}}$ are the momentum and energy transferred to the ladder during the RIXS scattering process, $\Gamma = t/2$ is related to the core-hole lifetime, $D_l = \sum_{\alpha,\sigma} P_\alpha e^{iq_\parallel R_l} c_{l,\alpha,\sigma}^\dagger p_{l,\sigma}$ is a local dipole operator responsible for the $2p \rightarrow 3d$ transition and $p_{l,\sigma}$ annihilates an electron in the $2p$ core level. The phase factor P_α in the definition of the dipole operator is used to isolate the intra- and inter-band contributions to the scattering. For intra-band scattering $P_0 = P_1 = 1$ while for inter-band scattering $P_0 = -P_1 = 1$. When diagonalizing the Hamiltonian to obtain the intermediate states $|n\rangle$, we also add a local potential

$$H_{\text{ch}} = U_q \sum_{l,\alpha,\sigma} \hat{n}_{l,\alpha,\sigma} [1 - \hat{n}_{l,\sigma}^p] \quad (8)$$

to Eq. (5), which accounts for the attractive potential produced by the core hole. Here, $\hat{n}_{l,\sigma}^p = p_{l,\sigma}^\dagger p_{l,\sigma}$ is the number operator for the core hole on rung l and we set $U_q = -2t$.

160 All of our RIXS calculations are carried out with an incident photon energy tuned to the
 161 maximum in the XAS spectra. The XAS spectra are given by

$$I_{\text{XAS}}(\omega) = \sum_n |\langle n | D_{\mathbf{k}_{\text{in}}=0} | g \rangle|^2 \delta(E_n - E_g - \omega). \quad (9)$$

162 We also computed the dynamical spin structure factor

$$S(q, \omega) = \sum_f |\langle f | \hat{S}_{q,\pm}^z | g \rangle|^2 \delta(E_f - E_g - \omega) \quad (10)$$

163 to help facilitate comparisons with our results shown in the main text, where the RIXS
 164 cross section cannot be computed on large clusters. Here, $\hat{S}_{q,\pm}^z = \frac{1}{\sqrt{2N}} \sum_l e^{-iqR_l} (\hat{S}_{l,1}^z \pm \hat{S}_{l,0}^z)$,
 165 where the $+$ or $-$ sign again corresponds to the intra- and inter-band components, and
 166 $\hat{S}_{l,\alpha}^z = \hat{n}_{l,\alpha,\uparrow} - \hat{n}_{l,\alpha,\downarrow}$ is the spin- z operator of the orbital on rung l and leg α .

167 At transition metal L -edges, the magnetic excitations are typically dominated by the
 168 $\Delta S = 1$ scattering channel. The intra- and inter-band components of the RIXS spectra
 169 in the $\Delta S = 1$ channel are shown in the top rows of Figs. S8 and S9, respectively. [The
 170 incident photon energy ω_{in} in both cases was chosen to coincide with the maximum in
 171 the corresponding XAS spectra, as shown in the insets of Fig. S8, panels (a)-(c).] The
 172 corresponding intra- and inter-band components of the dynamical spin structure factors are
 173 shown in the bottom rows [panels (d)-(e)] of each figure.

174 In the intra-band scattering channel, the RIXS response has a dispersing feature emanat-
 175 ing from $q = 0$ that originates from intra-band particle-hole excitations in the bands crossing
 176 the Fermi level. These excitations are most intense in the two-band case [Fig. S8(a)] and
 177 are suppressed in the incipient band cases [Figs. S8(b),(c)]. This change reflects the removal
 178 of the hole-like band from the Fermi surface in the latter case, which eliminates a channel
 179 for intra-band particle-hole excitations. This behavior is also reflected in the corresponding
 180 intra-band components of the dynamical structure factor, indicating that $S(q, \omega)$ provides a
 181 reasonable proxy for the low-energy magnetic RIXS response in our multi orbital toy model.

182 We obtain qualitatively different behavior in the inter-band response, shown in Fig. S9.
 183 In this case, the inter-band magnetic excitations have a continuum-like structure, similar to
 184 prior calculations for doped Hubbard ladders.¹³ For the two-band case, shown in Fig. S9(a),
 185 the excitations are gapped at $q = 0$ and disperse towards lower energies at the zone bound-
 186 aries. In the incipient band case, shown in Figs. S9(b) and S9(c), the same excitations are
 187 pushed to higher energies and are gapped by an amount proportional to $t_{\perp}$. As with the

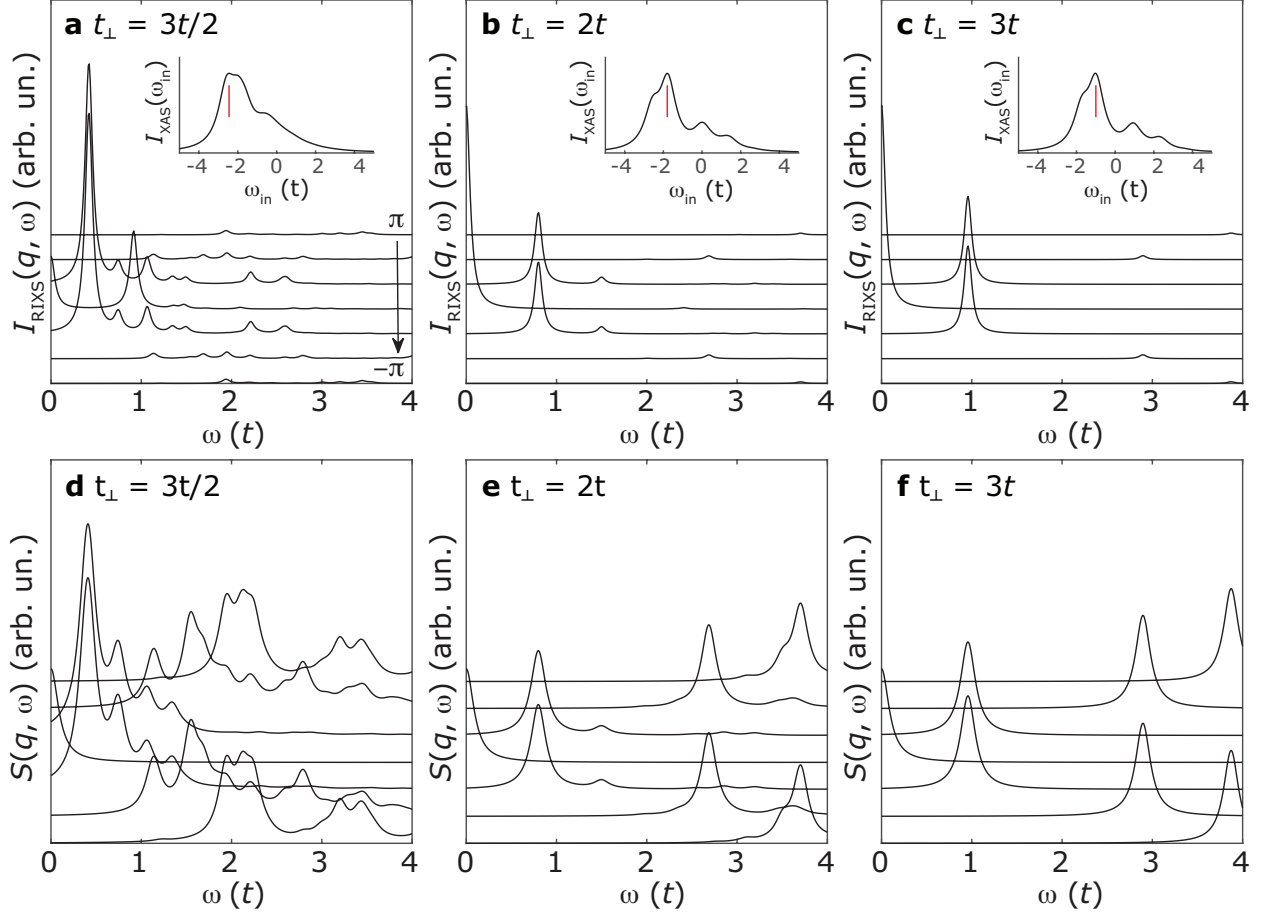


FIG. S8: Intra-band RIXS response. The top row plots the intra-band components ($q_y = 0$) of the RIXS response in the $\Delta S = 1$ channel, computed for (a) $t_{\perp} = 3t/2$, (b) $t_{\perp} = 2t$, and (c) $t_{\perp} = 3t$. The incident photon energy in each case was tuned to the resonance peak in the corresponding XAS spectra, as shown in the insets of each figure. The second row shows the intra-band scattering components of the dynamical spin structure factor for (d) $t_{\perp} = 3t/2$, (e) $t_{\perp} = 2t$, and (f) $t_{\perp} = 3t$. The RIXS spectra are plotted on the same relative intensity scale so the intensity of panels (a)-(c) can be meaningfully compared to each other. The same holds true for panels (d)-(e).

intra-band case, these features are qualitatively captured by the inter-band components of the dynamical spin structure factor, shown in Figs S9(d)-(e). All of the observations derived from this 1D toy model are consistent with those observed in our 2D bilayer model. Moreover, the qualitative consistency between the RIXS intensity and $S(q, \omega)$ provides theoretical support for the notion that the dynamical spin susceptibility can provide a qualitative description of the excitations probed by RIXS, as discussed in the main text.

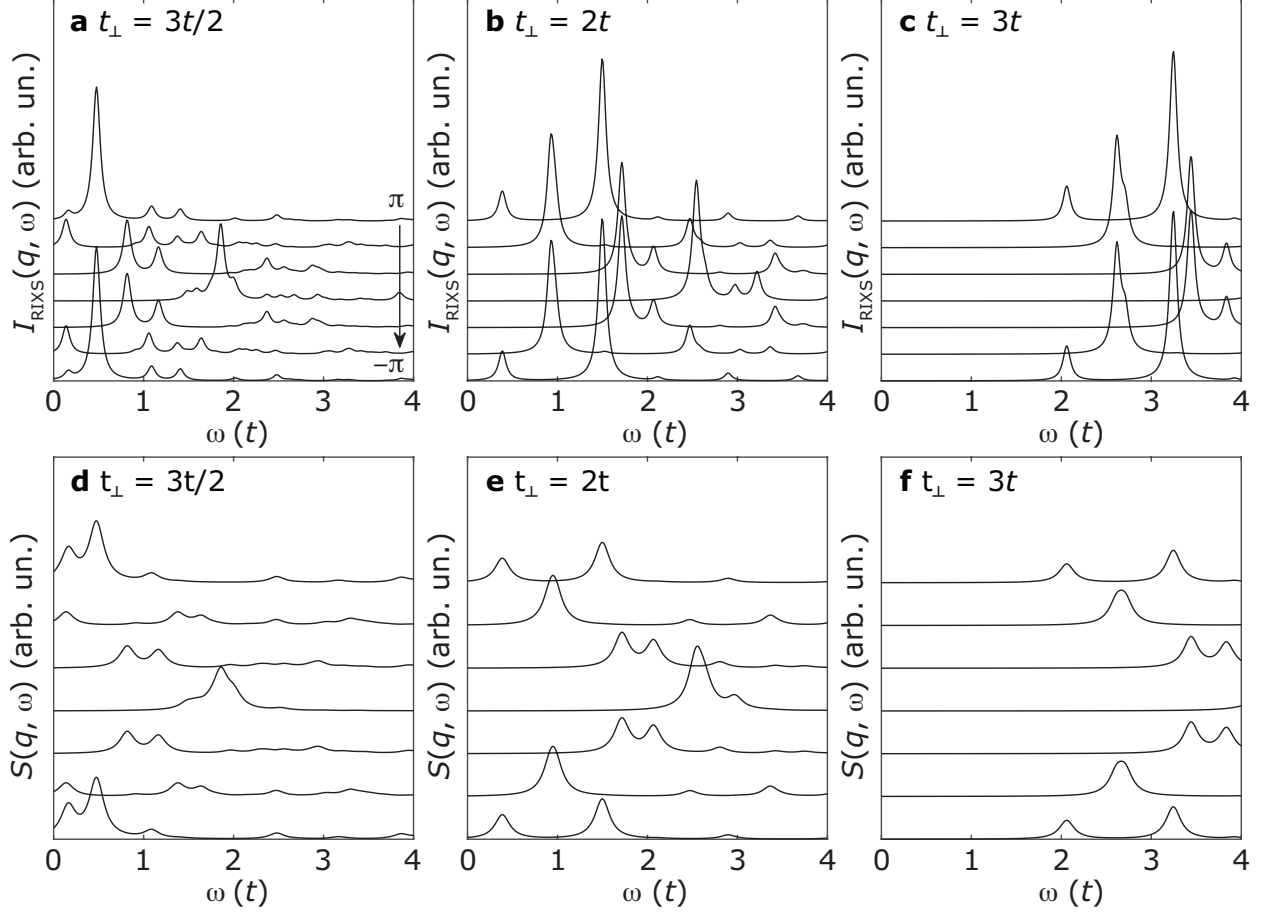


FIG. S9: Inter-band RIXS response. The top row plots the inter-band components ($q_y = \pi$) of the RIXS response in the $\Delta S = 1$ channel, computed for (a) $t_{\perp} = 3t/2$, (b) $t_{\perp} = 2t$, and (c) $t_{\perp} = 3t$. The incident photon energy in each case was tuned to the resonance peak in the corresponding XAS spectra, as shown in the insets of each figure. The second row shows the inter-band scattering components of the dynamical spin structure factor for (d) $t_{\perp} = 3t/2$, (e) $t_{\perp} = 2t$, and (f) $t_{\perp} = 3t$. The RIXS spectra are plotted on the same relative intensity scale so the intensity of panels (a)-(c) can be meaningfully compared to each other. The same holds true for panels (d)-(f).

III. RIXS DATA

A. Data along (H,H)

Figure S10 reports the RIXS data acquired from bulk FeSe along the (H,H) direction. Similarly to the (H,0) direction, we detect the presence of dispersive spin excitations moving

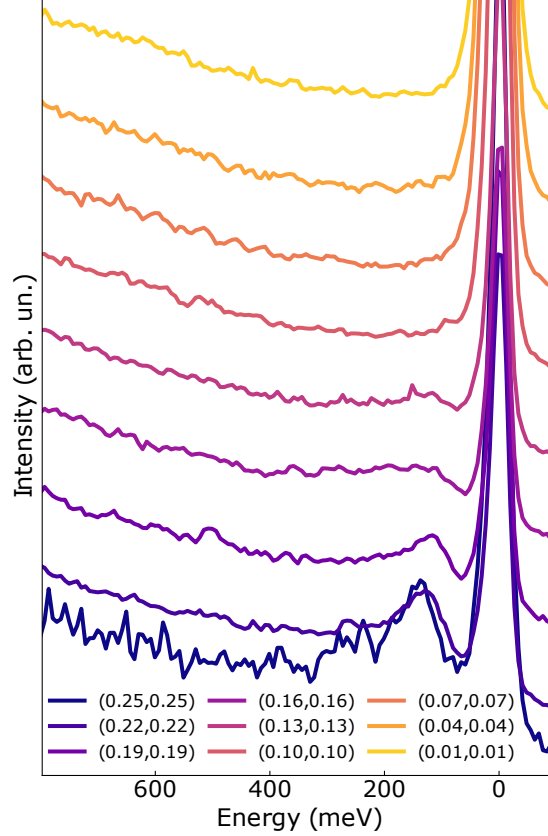


FIG. S10: RIXS data on FeSe bulk collected along the (H,H) direction.

to lower energies as we approach the zone center.

Figure S11 shows the RIXS spectra of FeSe/STO along the (H,H) direction. As with the data taken along the (H,0) direction, we observe a broad peak centered at 300-400 meV that hardly disperses in momentum space.

B. Energy detuning for FeSe/STO

As mentioned in the main text, spectroscopic measurements on a monolayer of materials are difficult to interpret. To ensure our sensitivity to the FeSe grown onto the STO substrate, we collected RIXS measurements at different incident energies and present the results in Fig. S12. As we tune the incident photon energy away from the maximum of the Fe absorption edge we observe a clear decrease of intensity. This observation corroborates the hypothesis that the RIXS signal originates from FeSe and not from the substrate or capping layer.

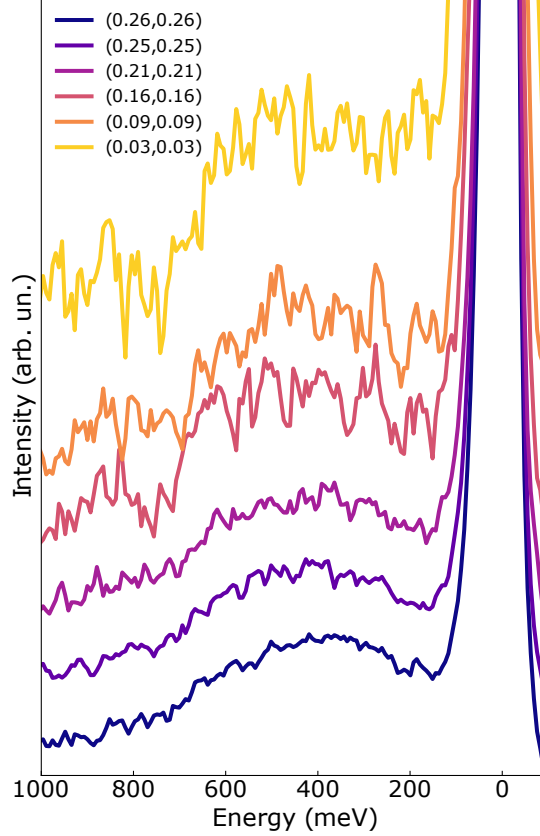


FIG. S11: RIXS data on FeSe/STO collected along the (H,H) direction.

The peak that we ascribed to the spin excitation (see main text for a detailed discussion) of FeSe/STO displays a Raman-like behaviour as the energy is changed. The peak energy does not change with the incident energy, an observation which signals that we are coherently probing an excitation of the system and not fluorescent emission (which would be at fixed emission energy, and not at fixed energy transfer). At the same time, increasing the incident energy strongly suppresses the peak observed at low energy loss.

C. Fitting of RIXS spectra in FeSe bulk and monolayer

1. Fitting of bulk FeSe

We fit the RIXS spectra of the FeSe bulk using a variation of the method employed in previous papers.^{6–11} The model consists of two gaussian curves accounting for the elastic line, and a phonon at 20-30 meV, an asymmetric lorentzian for the spin excitations, and a

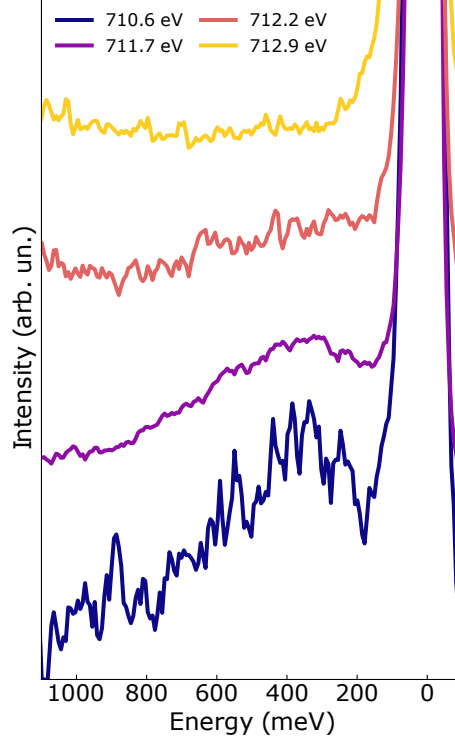


FIG. S12: RIXS data on FeSe/STO collected at different incident energies.

polynomial background. The results are reported for a single $\mathbf{Q}$ point in Fig. S13a.

2. Fitting of the FeSe/STO

For the case of FeSe/STO we performed a fitting using two gaussian curves at low energy (elastic and phonon), and an asymmetric lorentzian for the spin excitations feature. The background is less intense and could be neglected in this sample. In Fig. S13b, we show the fitting for the case of the FeSe/STO. The spectral weight in the region 50-150 meV is not perfectly reproduced. The presence of a strong elastic line due to diffuse scattering from the interfaces of the sample could lead to a broadening of the elastic line which could not reliably be fitted. The non-fitted extra spectral weight could also be due to saturation of some of the pixels of the CCD due to the strong elastic signal (with intensity leaking on either sides of the elastic peak). In any case, a more involved fitting achieved by adding an additional gaussian would not produce any additional information or affect the peak position of the spin excitations peak discussed in the main text.

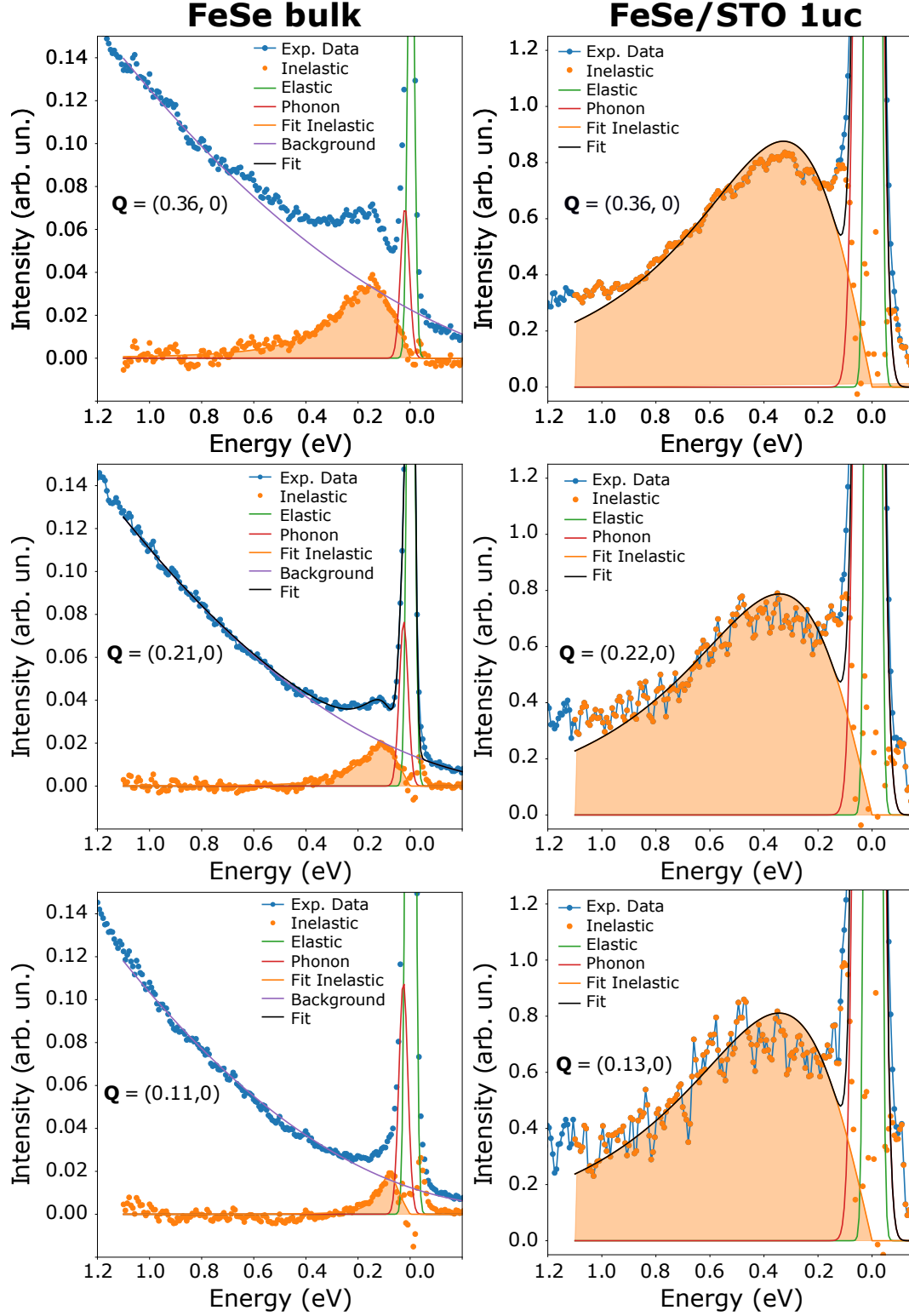


FIG. S13: Representative fitting for the bulk and monolayer FeSe at high, medium and low momentum transfer.

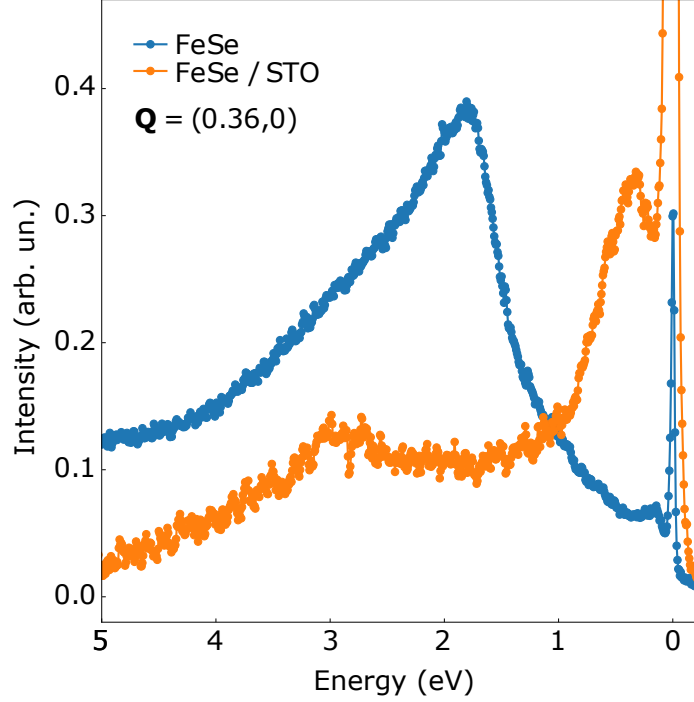


FIG. S14: Comparison at high energy loss of FeSe and FeSe/STO at $\mathbf{Q} = (0.36, 0)$.

3. Comparison of high energy loss

In Fig. S14 we display a comparison between FeSe and FeSe / STO at high energy loss up to 5 eV. The spectral intensity has been adjusted for better visualization. The fluorescence line of FeSe resembles that of Fe pnictides, but the lineshape of FeSe/STO is highly distorted by the limited thickness. For a more exhaustive comparison, measurements with higher statistics are needed.

IV. COMPARISON OF RIXS DATA WITH SUSCEPTIBILITY

In Fig. S15 we report a comparison of charge susceptibility and the RIXS experimental data for FeSe/STO and the incipient band model **a** and FeSe bulk and the two band model **b** for different momenta. We include both intra- (orange) and interband (blue) charge susceptibility. To convert the energy from units of t to energy we used the same values reported above for the spin susceptibility (90 and 160 meV for the bulk and incipient band cases). In both models the interband charge susceptibility peaks at much higher energy than the interband and shows a flat gapped dispersion. The intraband susceptibility (blue

traces) presents a dispersion qualitatively similar to the spin component but much steeper. In both incipient and two band cases the calculations of the charge susceptibility do not agree with the experimental data.

In Fig. S16 we report a comparison of the sum of the spin and charge susceptibility with the experimental data at different momenta. We indicate as orange trace the interband component and as blue trace the intraband one. In this figure we observe the dominance of the interband susceptibility over the intraband one. In the case of the incipient band comparison this leads to a pretty good agreement between the interband susceptibility (dominated by the spin component) and the experimental data. For the case of the two band the model the agreement of the interband susceptibility is poor compared to the experimental data due to the lack of dispersion. However a good agreement between the intraband susceptibility (dominated by the spin component) is observed.

In Fig. S17, we display as a color plot the charge susceptibility as a function of Q and energy and the experimental data. In panels **a,c** (**b,d**) we report the intraband and interband charge susceptibility for the two band model (incipient band model). As white dots and diamonds we report the experimental data for the monolayer and bulk cases, respectively. We used the same t parameters of Fig. 3 of the main text to rescale the energy. Contrary to the plot of the spin susceptibility we observe a poor agreement between the charge dispersion and our experimental data, proofing that RIXS our experimental observations are connected to the spin components rather than the charge one.

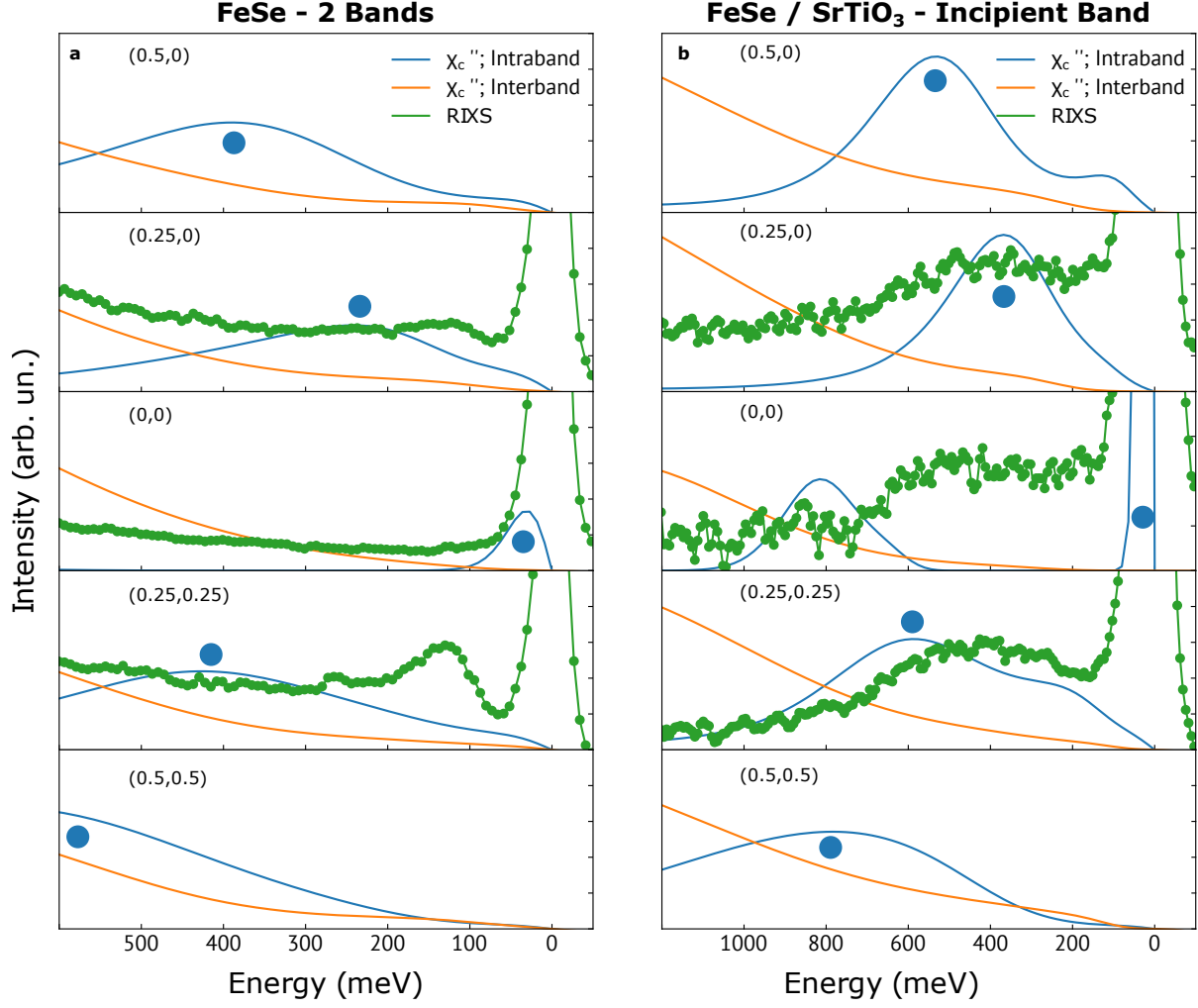


FIG. S15: Comparison of charge susceptibility and RIXS data. Left (right) panel: FeSe bulk (FeSe/STO) RIXS data and $\chi''_c(\mathbf{q}, \omega)$ from a two bands (incipient) band model for intra- (blue line) and interband (orange line) charge susceptibility. Blue circles indicate the energy position of the maximum of the spin susceptibility in the intraband cases.

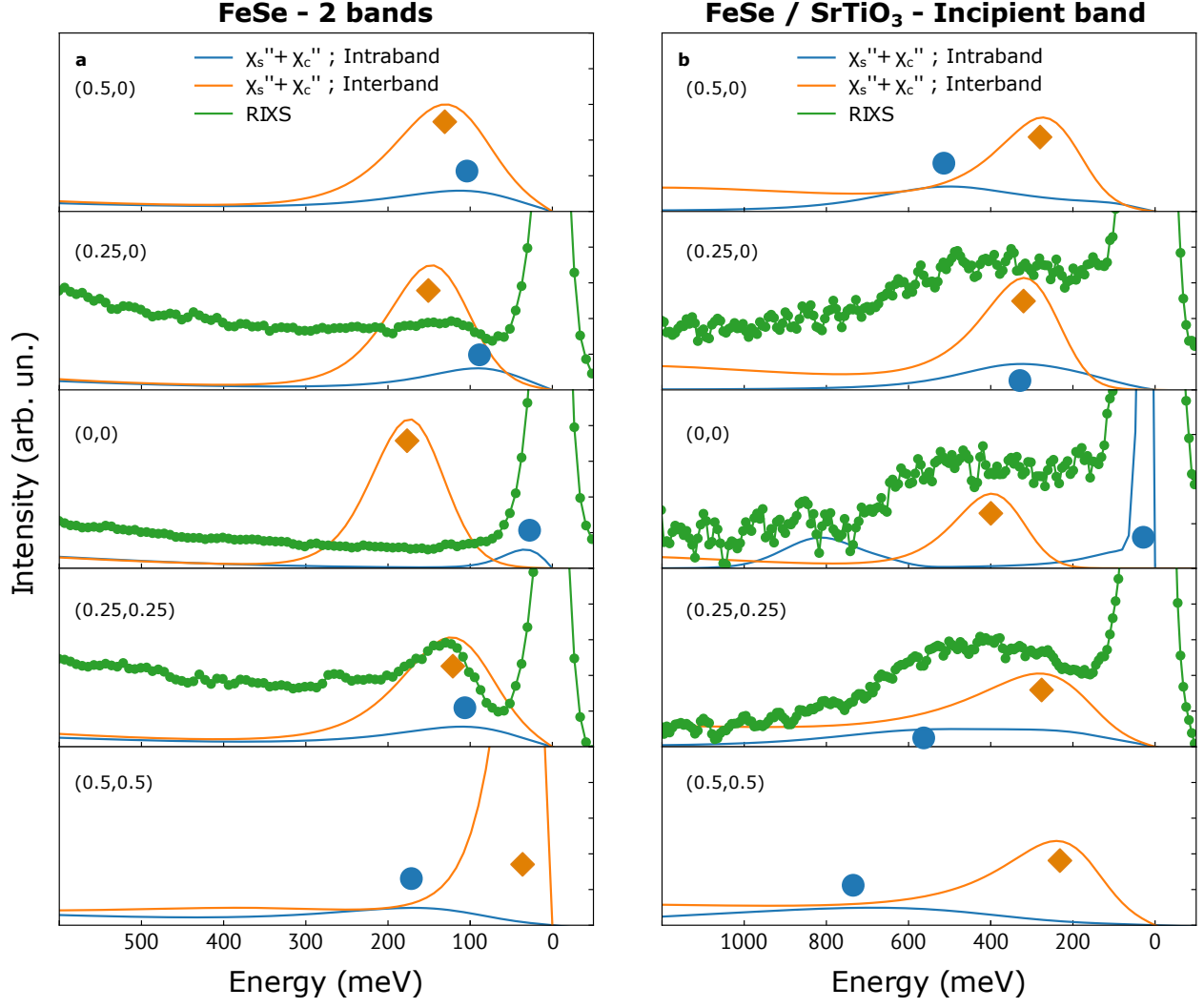


FIG. S16: Comparison of total susceptibility (charge and spin) susceptibility and RIXS data. Left (right) panel: FeSe bulk (FeSe/STO) RIXS data and $\chi''_{tot}(\mathbf{q}, \omega)$ from a two bands (incipient) band model for intra- (blue line) and interband (orange line) charge susceptibility. Orange circles (blue) indicate the energy position of the maximum of the spin susceptibility in the interband (intraband) cases.

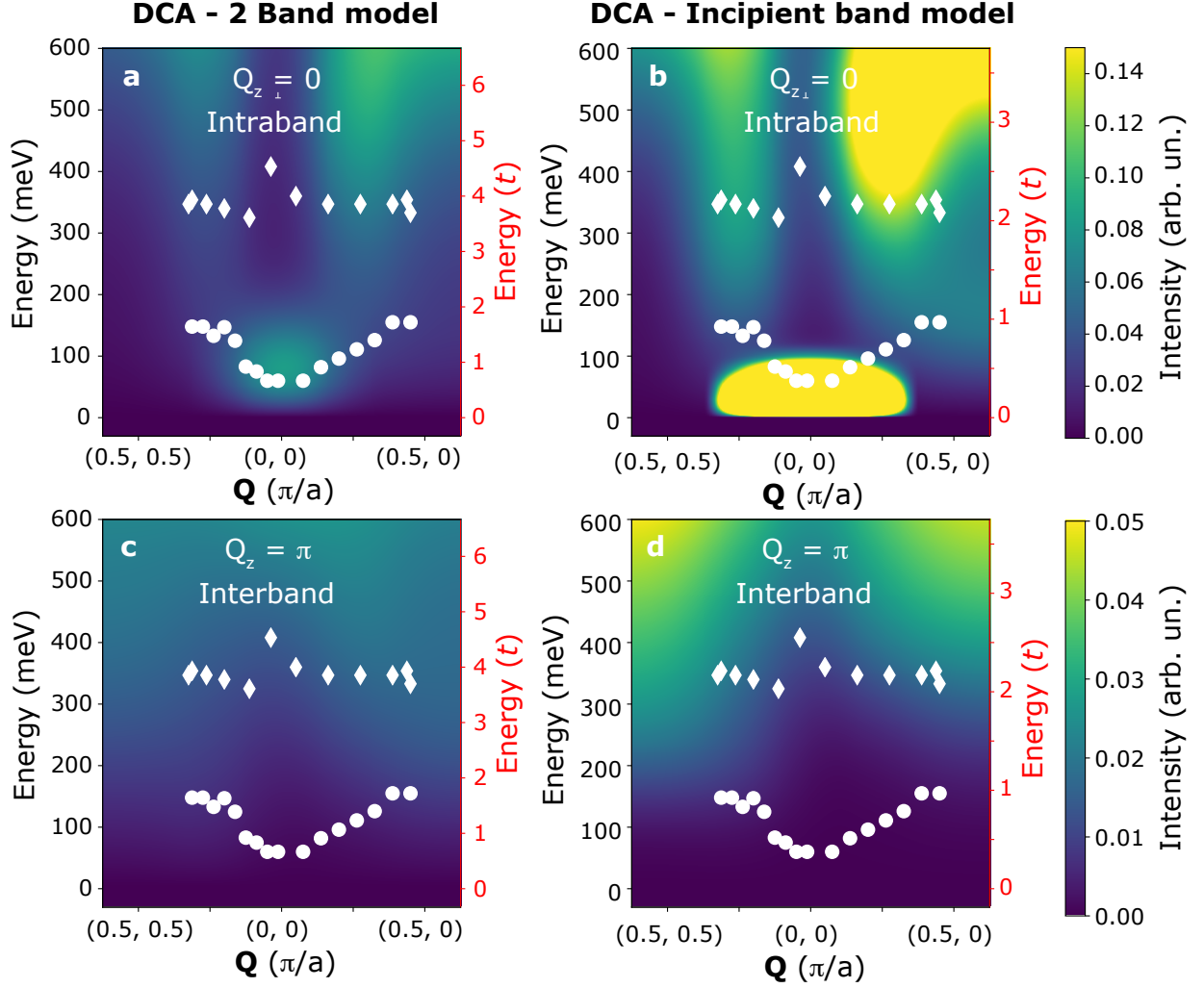


FIG. S17: Dynamical Charge susceptibility from DCA calculations. **a-d**: DCA calculations of the imaginary part of the charge susceptibility $\chi''(\mathbf{q}, \omega)$ for the two-band Hubbard model (**a**: intraband $Q_z = 0$; **c**: interband $Q_z = \pi$) and the incipient band Hubbard model (**b**: intraband $Q_z = 0$; **d**: interband $Q_z = \pi$). Red circles (white diamonds) indicate the energy position of the peak detected by RIXS in bulk (monolayer) FeSe. The uncertainties associated with peak fitting are smaller than the markers.

V. SUPERCONDUCTING GAP OF FESE/STO

The T_c of the FeSe/STO has been characterized by ARPES before applying the capping layer of amorphous Se. In Fig. S18 we show ARPES data of the superconducting gap. The gap has been estimated to be in 13.7 meV, corresponding to a $T_c \approx 60 - 65$ K.

ARPES on FeSe/STO before capping

EDC fo SC gap

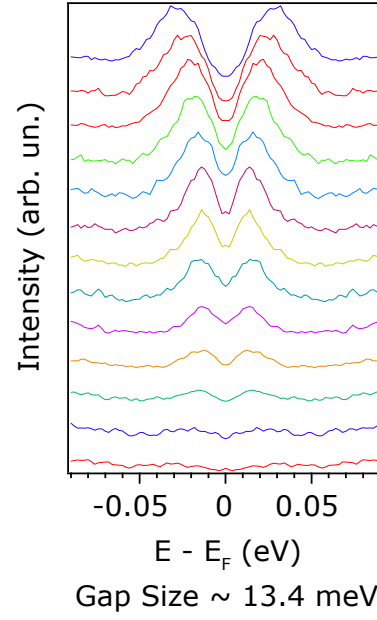


FIG. S18: ARPES measurements of superconducting gap in FeSe/STO before capping.

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