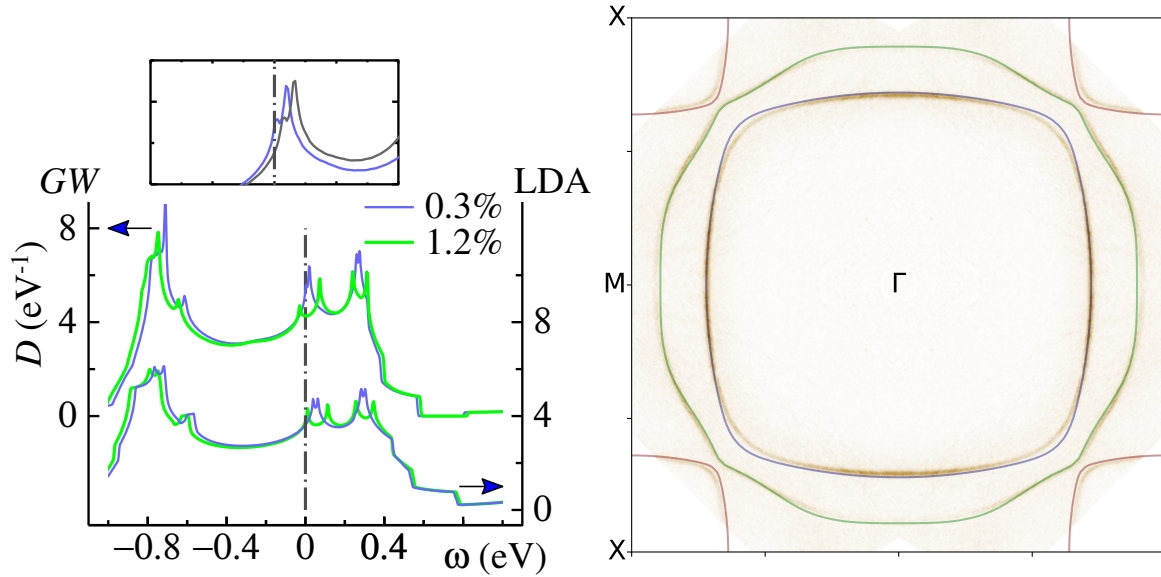


Supplementary note 1:

Crystal structure and Local density of states (DOS): Sr_2RuO_4 has a BCT lattice structure. We took the lattice parameters and internal positions from [1]. The lattice constant in the basal plane is $a=3.86448 \text{ \AA}$. The uniaxial strain was performed at constant volume.

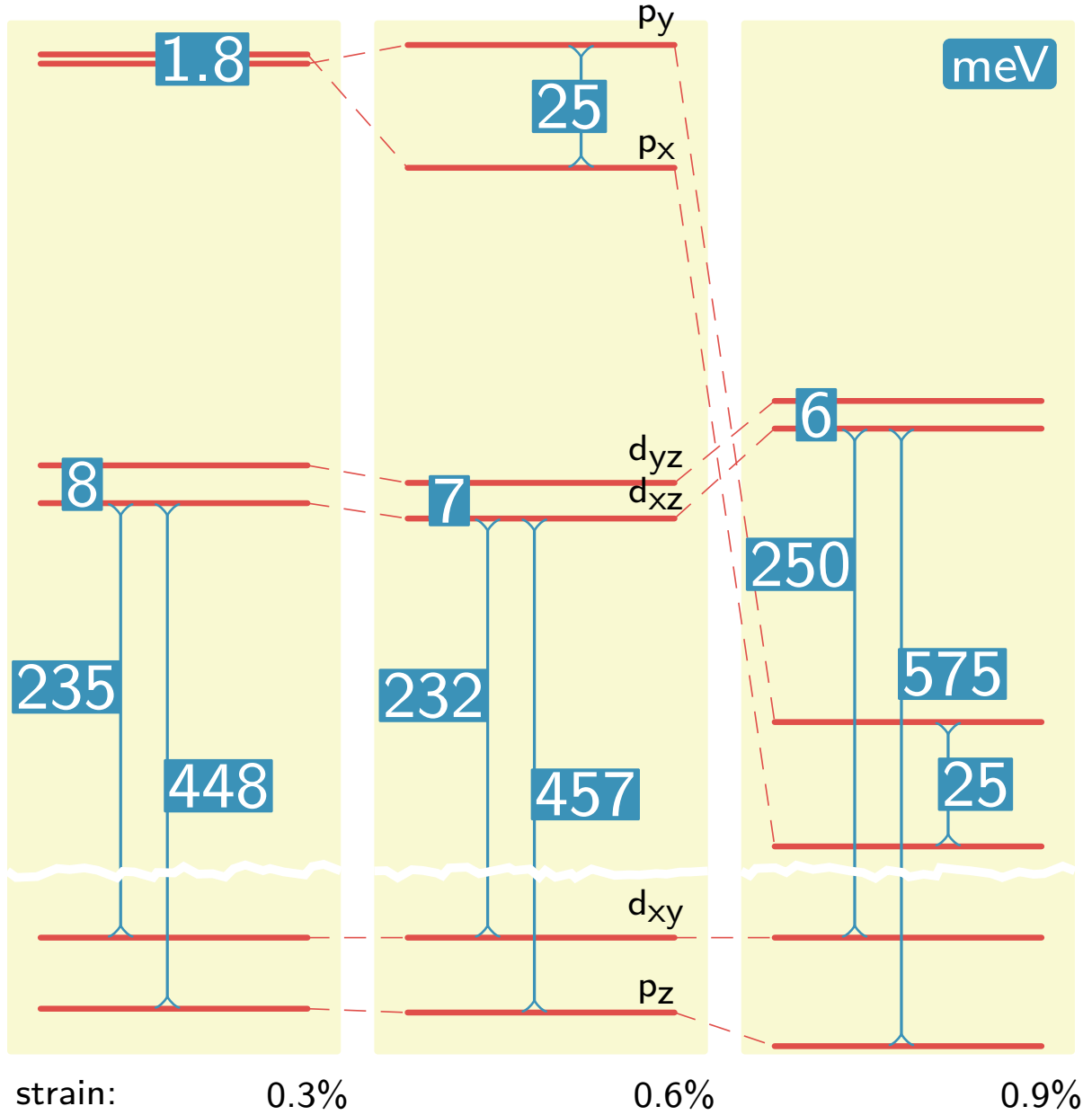
The DOS obtained from QSGW shows the presence of a sharp resonance slightly above the Fermi level in the unstrained case (Supplementary Fig. 1). Strain splits the single peak, the splitting increasing linearly with ϵ_x . The smaller, lower peak crosses E_F at $\epsilon_x=0.6\%$, which precisely coincides with the critical strain ϵ_x^* where $T_C = T_C^{\text{max}}$.



Supplementary Figure 1: **Density of States:** Top pair of curves show density of states (DOS) from QSGW, for strains $\epsilon_x = 0.3\%$ (blue) and 1.2% (green). A Van-Hove singularity appears slightly above the Fermi level when $\epsilon_x=0$. When $\epsilon_x \neq 0$ the singularity splits into two peaks, with the smaller peak crossing E_F at $\epsilon_x=0.6\%$. An intriguing possibility is to find conditions that cause the larger peak to cross E_F . Bottom pair is the corresponding DOS in the LDA. The Van-Hove singularity and its splitting are also seen, but the peaks split more symmetrically, and evolve more slowly with ϵ_x . On average, the LDA DOS is 25% smaller than the QSGW DOS, which is a consequence of the LDA's tendency to overestimate d bandwidths. Inset shows the QSGW DOS at $\epsilon_x=0.3\%$ on finer energy scale. The grey line shows the DOS with spin-orbit coupling removed. To the right, the high-resolution ARPES data [2] for the Fermi surfaces are shown (figure reproduced with due permission) in the background of our QSGW theoretical data. Weak discrepancies can be observed for the quasi-one-dimensional Fermi sheets.

Supplementary note 2:

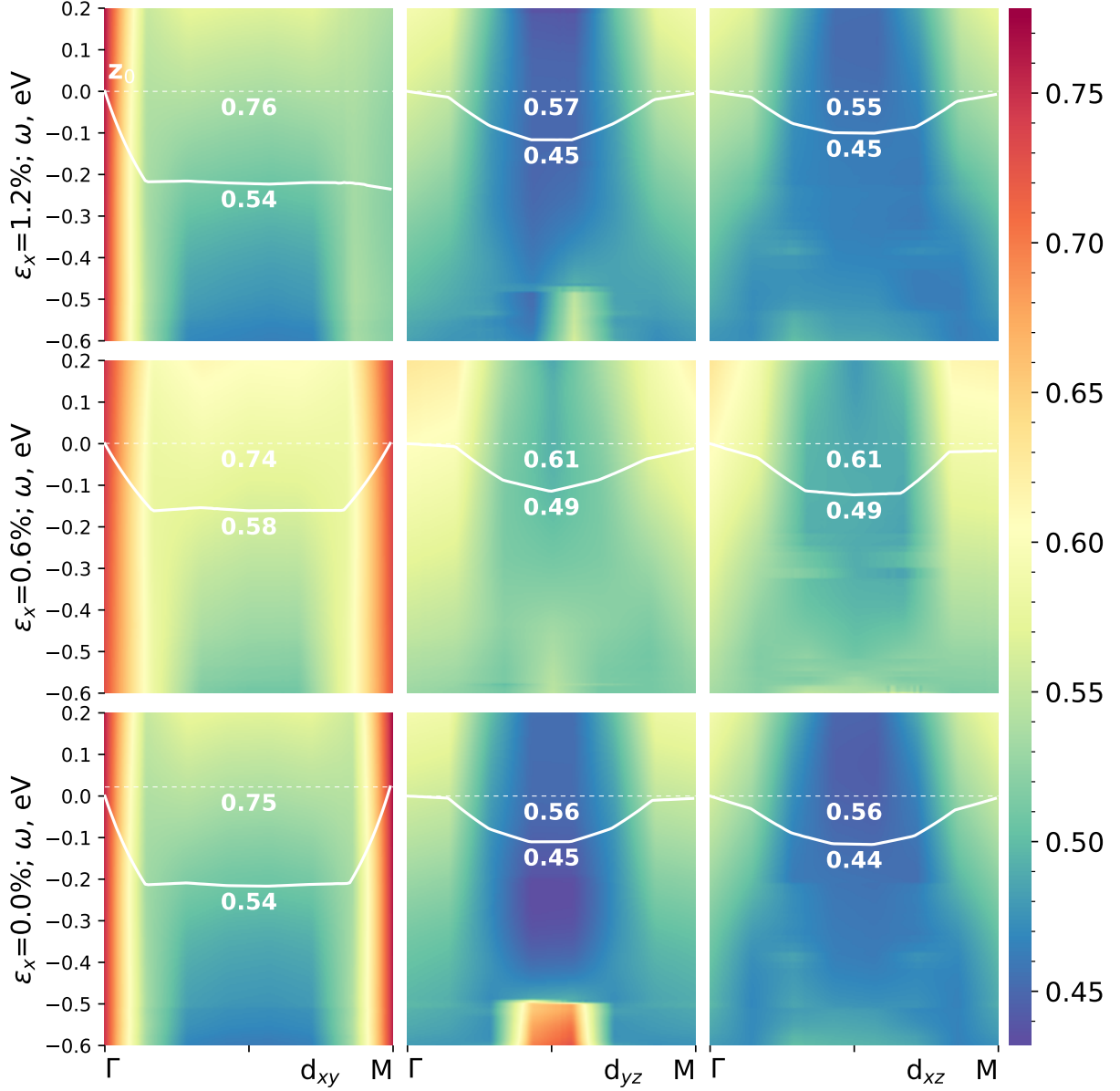
Energy levels: Strain lifts the tetragonal symmetry, splitting the degeneracy of the Ru ($4d_{xz}, 4d_{yz}$) and O (p_x, p_y) pairs. We find that the states split beyond the typical exchange scale of $\sim 4 \text{ meV}$ (Supplementary Fig. 2) when $\epsilon_x=0.6\%$. The split between Ru $4d_{xz}, 4d_{yz}$ is extremely small, and might fall below the resolution of several measurements trying to probe directly the consequences of lifting of the native tetragonal symmetry under strain. However, O p_x, p_y states split by nearly 20-25 meV.



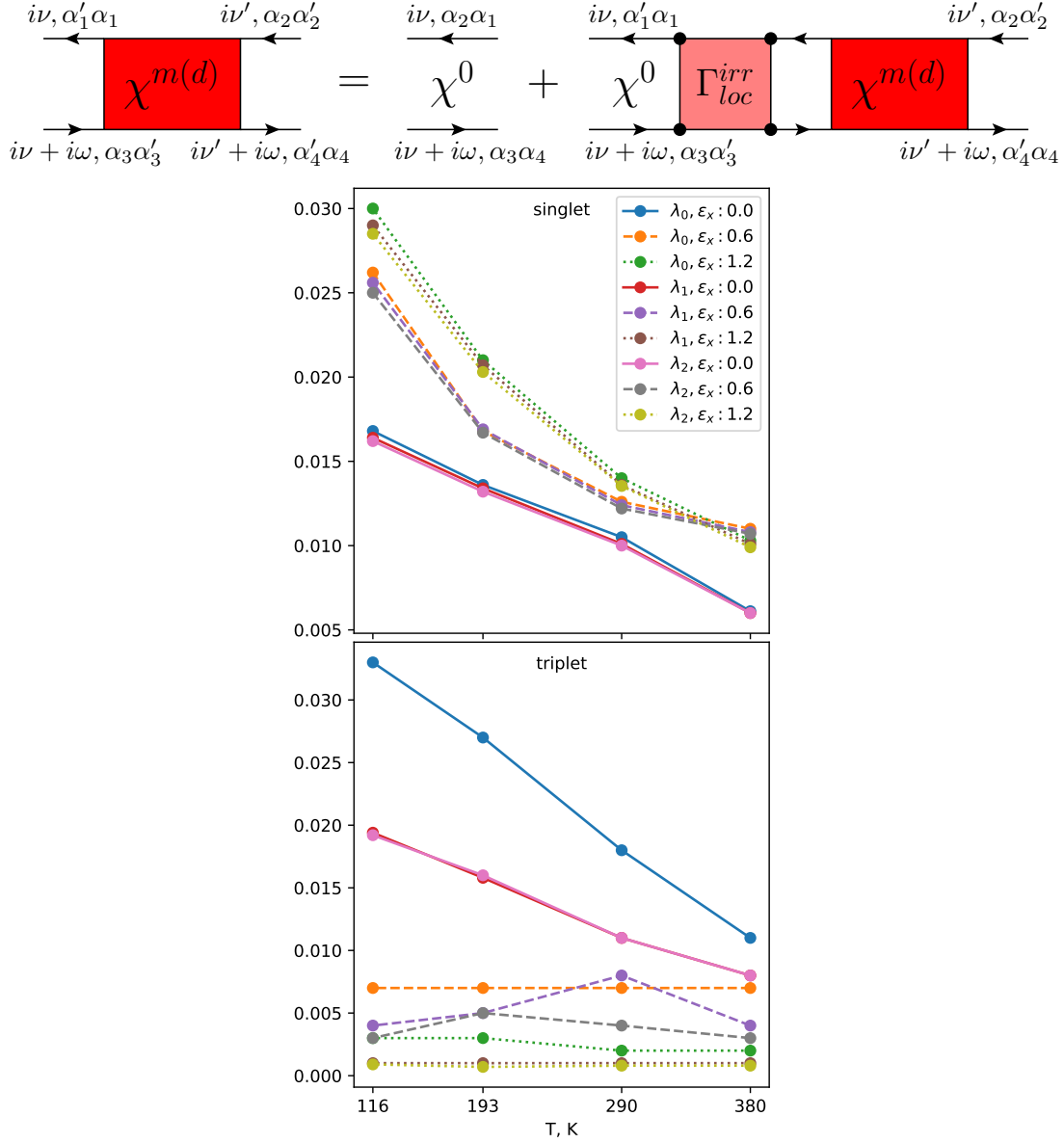
Supplementary Figure 2: **Energy levels:** Relative orientations of different active orbitals in Sr_2RuO_4 from QSGW. In strained Sr_2RuO_4 , we find typical splitting of 6-8 meV between Ru- d_{xz} and d_{yz} orbitals and about 25 meV between O- p_x and O- p_y . This energy splitting is larger than the spin exchange scale in Sr_2RuO_4 .

Supplementary note 3:

Non-local Coulomb correlations: As Coulomb interactions are long-range, they are treated by QSGW perturbatively in a self-consistent manner through a dynamic and momentum dependent self-energy, $\Sigma(\mathbf{k}, \omega)$. We analyse $\Sigma(\mathbf{k}, \omega)$ computed within QSGW to extract the momentum-dependent quasi-particle renormalization fac-

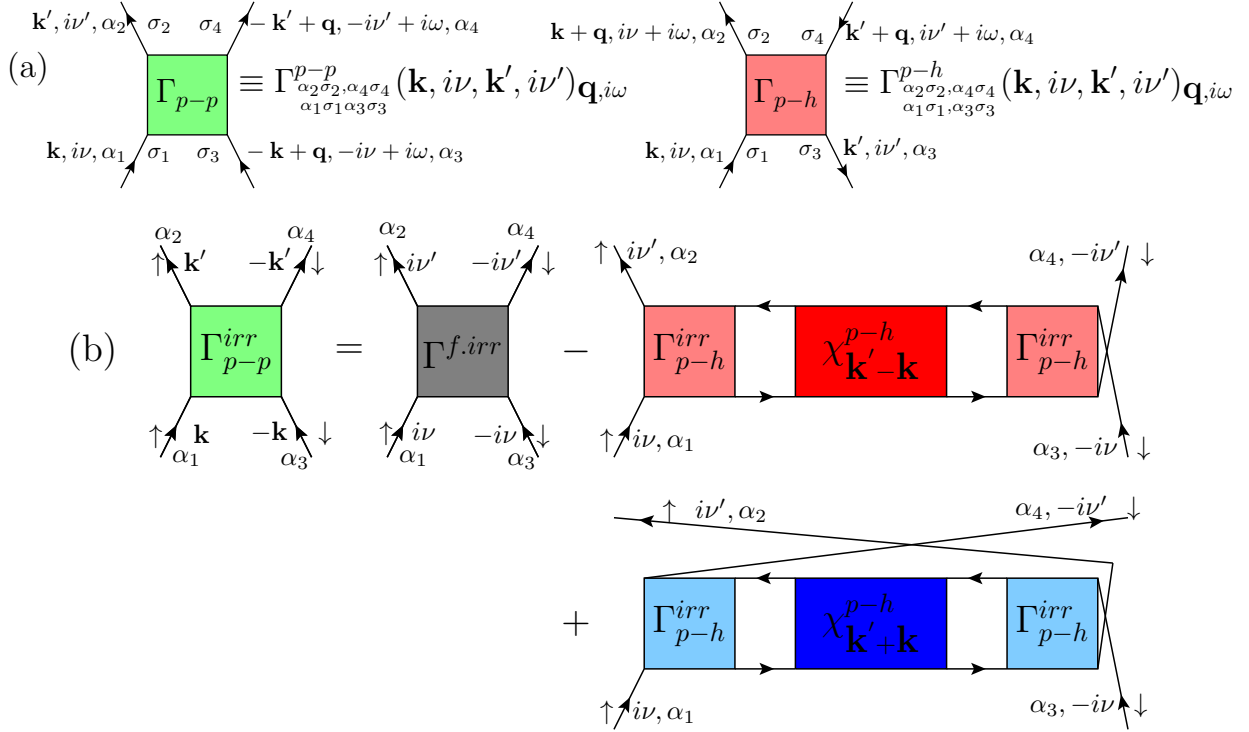


Supplementary Figure 3: **Non-local correlations:** Quasi-particle renormalization factor, $Z_k(\omega)$, computed in QSGW, as a heat map in the k - ω plane. White lines detail Z at the Fermi energy ($\omega = 0$) along the Γ -M line. The three bands present at E_F are all of Ru t_{2g} character, respectively d_{xy} , d_{yz} , d_{xz} -like on this line (from left to right). Z is a measure of how strongly nonlocality in space and time renormalize and smear out the energy bands: at $Z=1$ electrons are perfectly coherent and act like independent particles; when $Z=0$ all coherence is lost. Its k -dependence is unusually strong. To put it in perspective, in iso-structural La_2CuO_4 , Z varies by $\sim 20\%$ [3] for the Cu- $d_{x^2-y^2}$ band. Lower panels show Z_k evolves in a non-trivial, orbital-dependent fashion with strain. Spin fluctuations missing in QSGW further reduce Z .



Supplementary Figure 4: The Feynman diagram for the Bethe-Salpeter equation in the spin (charge) channel. The non-local susceptibility is obtained by replacing the local propagator by the non-local propagator. In lower panel first (λ_0), second (λ_1) and third (λ_2) eigenvalues with and without strain as function of temperature in singlet and triplet channels are shown.

tor $Z_k = (1 - \partial\Sigma(\mathbf{k}, \omega)/\partial\omega)^{-1}$. Supplementary Fig. 3 shows how the non-local $\Sigma(\mathbf{k}, \omega)$ renormalizes bands differently on the Γ -M line. The variation is orbital-dependent, fluctuating by $\sim 40\%$ in a nontrivial manner with $\mathbf{k}$, ω , and strain. This has a significant effect on the d bandwidth, and is one important reason why QSGW bands are significantly narrower than their LDA counterparts. Bandwidths are nevertheless much larger than ARPES measurements, and they get further renormalized by strong spin fluctuations included in DMFT.



Supplementary Figure 5: (a) The spin, orbital, momentum, and frequency labels of vertex functions in the particle-particle ($p-p$) channel and the particle-hole ($p-h$) channel. (b) The decomposition of the irreducible vertex function in the particle-particle channel Γ_{p-p}^{irr} .

Supplementary note 4:

Magnetic, charge and Superconducting susceptibilities: Here we show the Feynman diagram representation of the Bethe-Salpeter equation in the particle-hole ($p-h$) channel (see Supplementary Fig. 4).

In order to extract Γ_{loc}^{irr} , we employ the Bethe-Salpeter equation which relates the local two-particle Green's function (χ_{loc}) sampled by CTQMC, with both the local polarization function (χ_{loc}^0) and Γ_{loc}^{irr} .

$$\Gamma_{loc, \alpha_3, \alpha_4}^{irr, m(d)}(i\nu, i\nu')_{i\omega} = [(\chi_{loc}^0)^{-1} - \chi_{loc}^{m(d)-1}]_{\alpha_1, \alpha_2}^{\alpha_3, \alpha_4}(i\nu, i\nu')_{i\omega}. \quad (1)$$

The non-local polarization bubble in the $p-h$ channel is computed from

$$\chi_{\alpha_3\sigma_3, \alpha_4\sigma_4}^0(i\nu, i\nu')_{\mathbf{q}, i\omega} = -\frac{T}{N_k} \sum_{\mathbf{k}} G_{\alpha_2\alpha_1, \sigma_1}(\mathbf{k}, i\nu) \cdot G_{\alpha_3\alpha_4, \sigma_3}(\mathbf{k} + \mathbf{q}, i\nu + i\omega) \cdot \delta_{i\nu, i\nu'} \cdot \delta_{\sigma_1\sigma_2} \cdot \delta_{\sigma_3\sigma_4} \quad (2)$$

With these two quantities we can get the full dynamic and non-local spin and charge susceptibilities.

We show how our computed singlet and triplet eigenvalues behave with temperature and strain in Supplementary Fig. 4. Further, we show the Feynman diagram representation of the $p-p$ vertex and its decomposition using the $p-h$ vertex function computed in the magnetic and density channels in Supplementary Fig. 5.

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- [1] Q. Huang, *et al.*, Journal of Solid State Chemistry 112, 355 (1994)
 - [2] A. Tamai, *et al.*, *Phys. Rev. X* **9**, 021048 (2019).
 - [3] S. Acharya, *et al.*, *Phys. Rev. X* **8**, 021038 (2018).