Anisotropy of electronic correlations:
On the applicability of local theories to layered materials

B. Klebel,1 T. Schäfer,2,3 A. Toschi,1 and J. M. Tomczak1,∗

1Institute of Solid State Physics, TU Wien, A-1040 Vienna, Austria
2Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France
3CPHT, CNRS, École Polytechnique, IP Paris, F-91128 Palaiseau, France

Besides the chemical constituents, it is the lattice geometry that controls the most important material properties. In many interesting compounds, the arrangement of elements leads to pronounced anisotropies, which reflect into a varying degree of quasi two-dimensionality of their low-energy excitations. Here, we start by classifying important families of correlated materials according to a simple measure for the tetragonal anisotropy of their ab initio electronic (band) structure. Second, we investigate the impact of a progressively large anisotropy in driving the non-locality of many-body effects. To this end, we tune the Hubbard model from isotropic cubic in three dimensions to the two-dimensional limit and analyze it using the dynamical vertex approximation. For sufficiently isotropic hoppings, we find the self-energy to be well separable into a static non-local and a dynamical local contribution. While the latter could potentially be obtained from dynamical mean-field approaches, we find the former to be non-negligible in all cases. Further, by increasing the model-anisotropy, we quantify the degree of quasi two-dimensionality which causes this “space-time separation” to break down. Our systematic analysis improves the general understanding of electronic correlations in anisotropic materials, heterostructures and ultra-thin films, and provides useful guidance for future realistic studies.

I. INTRODUCTION

Some of the most intensely studied condensed matter systems are highly anisotropic, see Fig. 1: cuprates, pnictides, ruthenates, cobaltates, graphene, transition-metal dichalcogenides, and ultra-thin oxide films.

Indeed, with spherical symmetry surrendered to translational invariance, the crystal-structure of periodic solids may trigger substantially anisotropic phenomena: The coordination geometry of chemical constituents can induce crystal-fields, the lifting of orbital degeneracies, and the directional dependence of transfer integrals. In layered compounds or geometrically engineered ultrathin films, the latter effects conspire to produce low-energy dispersions that are—to a degree—confined to a two-dimensional plane. In Fig. 1 we quantify such anisotropy for a number of prominent correlated materials according to a measure α of their electronic structure, which we introduce below.

The effectively reduced dimensionality in the above mentioned one-particle ingredients has significant consequences for many-body effects2–4: First, low dimensions are the domain of non-local fluctuations in space: due to the low-coordination of the lattice geometries, the physics of the system is strongly dependent on the specific spatial configuration realized at each step of its time-evolution. As a result, in comparison to the 3D isotropic case, ordering instabilities are typically suppressed in quasi-2D materials or fully obviated in purely 2D systems (cf. Mermin-Wagner theorem5 or Kosterlitz-Thouless transitions6) by strong spatial fluctuations. The latter are reflected in significant enhancements of the corresponding susceptibilities, which affect large regions of the parameter space in 2D, while in 3D these are typically confined to the proximity of the actual phase transitions7. Second, these increased (two-particle) fluctuations may affect one-particle spectral properties, as dictated by the Schwinger-Dyson equation of motion: Corresponding renormalization effects, such as static energy shifts, quasi-particle effective weights and lifetimes may acquire strong non-local variations. The most prominent example is certainly the pseudogap regime in doped cuprates8–11, but momentum-selective coherence and quasi-particle weights can in fact originate from var-
ious non-local fluctuations of spin, charge, or orbital degrees of freedom\textsuperscript{12}. Accordingly, pseudogap physics has been evidenced in a number of correlated materials, e.g., iron pnictides\textsuperscript{13–16} and chalcogenides\textsuperscript{17}, iridates\textsuperscript{18}, and (layered) nickelates\textsuperscript{19, 20}.

The most successful local approach for correlated electrons is dynamical mean-field theory (DMFT)\textsuperscript{21}, which can be combined with density functional theory, DFT+DMFT\textsuperscript{22}, for \textit{ab initio} material calculations. Exact in $D = \infty$\textsuperscript{23}, DMFT is empirically found to be reasonable in three dimensions—except in the vicinity of a (second order) phase transition (for the reasons stated above). However, recently it was shown\textsuperscript{24} that even in the presence of strong non-local (anti-ferromagnetic) fluctuations, the dynamical part of many-body renormalizations (to linear order the quasi-particle weight) remains essentially local in 3D, while a notable variation within the Brillouin zone is engendered for static components of the self-energy. As a result, the self-energy verifies—to a good approximation—a space-time separation\textsuperscript{24}:

$$
\Sigma(\mathbf{k}, \omega) = \Sigma_{\text{static}}(\mathbf{k}) + \Sigma_{\text{local}}(\omega).
$$

In this work we explore and quantify the limits of this approximation: We monitor the momentum-dependence of many-body renormalizations of the doped Hubbard model as a continuous function of the tetragonal anisotropy in the one-electron hopping—from the cubic case in 3D to the square lattice in 2D. We find that for an anisotropy smaller than roughly one half ($\alpha < 1/2$), a local approximation to the dynamical self-energy becomes inadequate. In conjunction with the survey of electronic-structure anisotropies (Fig. 1), our results hence provide guidance for future first principle investigations of layered correlated systems: We establish a rule-of-thumb when the use of techniques beyond DFT+DMFT\textsuperscript{7, 25–27} becomes a prerequisite for reliable \textit{ab initio} calculations.

The paper is organized as follows: We detail the employed methodology in Section II. In Section III we present and discuss our results: Section III A is devoted to the \textit{ab initio} classification of materials according to the anisotropy of their electronic structure. In Section III B we study the Hubbard model for the range of anisotropy covered by the materials in Section III A. We conclude in Section IV with a synthesis of the materials classification and our many-body findings.

II. METHODS

A. \textit{Ab initio} calculations

For the \textit{ab initio} survey of materials, Fig. 1, we (i) performed (non-spinpolarized) density functional calculations (using the PBE functional) with WIEN2k\textsuperscript{28}, (ii) constructed maximally localized Wannier functions\textsuperscript{29} for the $d$-orbitals of the transition metal relevant for the low-energy band-structure with wannier90\textsuperscript{30} via wien2wannier\textsuperscript{31}, and (iii) analyzed the hopping amplitudes of the one-particle Hamiltonian in real-space: $H = \sum_{\tau, R, \sigma, \sigma'} H_{\tau\tau', LL, \rho \rho'} (\mathbf{R} - \mathbf{R'}) c_{\tau\rho, LL}^\dagger c_{\tau'\rho', LL}$. Here, $\tau$ indexes transition-metal atoms in the unit-cell, $L$ the $d$-orbitals (or a subset of them, see below), $R$ the unitcell, and $\sigma$ the spin. The measure $\alpha$ is then defined as the ratio of the in-plane and out-of-plane hopping amplitudes between nearest-neighbour transition-metal ions:

$$
\alpha = \frac{\max_{\tau, R} \sum_{LL} |H_{\tau\tau, LL, \rho\rho} (\mathbf{R})|}{\max_{\tau, R} \sum_{LL'} |H_{\tau\tau, LL', \rho\rho} (\mathbf{R})|}.
$$

To the denominator (nominator) contribute all orbital combinations of hopping elements between transition-metal ions that are nearest-neighbours in the $ab$-plane (in the $c$-direction). Taking the maximum, e.g., for the numerator, over $(\tau_{\perp}, R_{\perp}) \neq (\tau, R)$ assures selecting transition-metal ions that are nearest-neighbours. For example, $R_{\perp} = 0 (R_{||} = 0)$ for an intra-cell hopping between nearest neighbours $\tau \neq \tau_{\perp} (\tau \neq \tau_{||})$; while $R_{\perp} = a_x \hat{e}_z (R_{||} = a_y \hat{e}_x)$, or, if $a_y < a_x$, $R_{\perp} = a_y \hat{e}_y$ with $\tau = \tau_{\perp} (\tau = \tau_{||})$ indicates the hopping between the same atom but in adjacent unit-cells translated by the lattice constant $a_x (a_y)$ in the direction of the respective unit-vector $\hat{e}_x (\hat{e}_y)$. The anisotropy as described by Eq. (2) is clearly too simple to account for \textit{global} material trends. In particular, $\alpha$ largely depends on the orbital-subspace chosen to represent the pertinent low-energy dispersions (this will be detailed below for the high-$T_c$ parent compounds). However, we believe $\alpha$ to be a good indicator for trends \textit{within} a given family of compounds. As such, the anisotropy proxy of Eq. (2) could even serve as descriptor in high-throughput studies that target the optimization of some property through, e.g., chemical pressure via isovalent substitutions\textsuperscript{32}.

B. \textit{Many-body} calculations

1. \textit{The model}

We consider the one-band Hubbard model (in the usual notation)

$$
H = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}
$$

on a tetragonal lattice (see Fig. 2):

$$
\epsilon_{\mathbf{k}} = -2t_0 \left[ \cos(k_x a_x) + \cos(k_y a_y) + \alpha \cdot \cos(k_z a_z) \right]
$$

where $k_i \in [0, 2\pi/a_i]$ and we set $a_i = 1$ ($i = x, y, z$). The parameter $\alpha \in [0, 1]$ is the one-band equivalent of Eq. (2) as it scales the hopping amplitude in the $z$-direction: It allows to continuously tune the system between the 3D cubic case ($\alpha = 1$) and the 2D square lattice ($\alpha = 0$). Such change in the anisotropy directly affects, per construction, the overall kinetic energy and, hence—for a fixed interaction—the degree of electronic correlations in
In order to be able to reliably estimate how non-local fluctuations on top of DMFT affect the space-time-separation of the self-energy in Eq. (1), we use a diagrammatic extension of DMFT, the dynamical vertex approximation (DFA) in its most used ladder-implementation with Moriya corrections. In contrast to quantum cluster methods, it includes temporal and non-local correlations at all lengths scales on an equal footing. For this reason, DFA has been often exploited to describe classical and quantum phase transitions, as well as the associated fluctuations. For further technical details on the DFA calculation see App. A.

After the calculation of the momentum-dependent self-energy on the Matsubara axis $\Sigma(k, i\omega_n)$, we adapt the chemical potential $\mu$ in order to recover the system’s filling, due to the lack of full self-consistency of ladder-DFA with Moriya corrections.

Throughout the paper, we focus on many-body renormalizations near the Fermi level. To this end, we perform a Taylor series expansion, specifically

$$\Sigma(k, \omega) = \text{Re} \Sigma(k, \omega = 0) + (1 - 1/Z(k))\omega i\Gamma(k)(\omega^2 + \pi^2 T^2) + \cdots,$$

where $\gamma(k) = -\text{Im} \Sigma(k, \omega = 0) = \Gamma(k)\pi^2 T^2 + O(T^4)$ is the scattering rate and $Z(k)$ the quasi-particle weight—provided the system realizes a Fermi-liquid. These quantities are extracted from the self-energy on the Matsubara axis, $\Sigma(k, i\omega_n)$, as follows:

$$\text{Re} \Sigma(k, \omega = 0) = \text{Re} \Sigma(k, i\omega_n)|_{i\omega_n \to 0}$$

$$\gamma_k = -\text{Im} \Sigma(k, i\omega_n)|_{i\omega_n \to 0}$$

$$Z_k = \left[1 - \frac{\partial \text{Im} \Sigma(k, i\omega_n)}{\partial \omega_n}|_{\omega_n \to 0}\right]^{-1}$$

To obtain stable results for our analyzed quantities (see next subsection), we performed and averaged over polynomial fits up to order 10 before the extrapolation to $i\omega_n \to 0$.

In all our calculations we use $U = 2$ in units of $t_\alpha$, placing us—at zero doping—in the intermediate-coupling regime, yielding the highest magnetic transition temperature in 3D. In this work, we will restrict ourselves to non-local correlations stemming from magnetism in the paramagnetic regime (paramagnons).

3. Analysis tools

To analyse our results, we introduce the following measures of non-locality for the expansion coefficients defined above in Eqs. (8)-(10), $a(k) = \text{Re} \Sigma(k, \omega = 0)$, $Z(k)$, and $\gamma(k)$:

(i) the standard deviation

$$\Delta_k a(k) = \sqrt{ \frac{1}{N_k} \sum_k \left| a(k) - a_{loc} \right|^2 }$$

with respect to the local (Brillouin-zone average) value $a_{loc} = 1/N_k \sum_k a(k)$. 

FIG. 2: The tetragonal lattice: In the $xy$-plane, electrons hop with an amplitude $t_\alpha$, while the electron transfer in $z$ direction is smaller by a factor $0 \leq \alpha \leq 1$. 

the system. As this trivial effect would hide the most interesting trends emerging from our dimensional investigation, we chose to keep the kinetic energy essentially independent of $\alpha$. To this end, we adjust the overall hopping amplitude $t_\alpha$ by requiring the second moment of the density of states to be constant and fix our energy units by imposing:

$$\int_{-\infty}^{\infty} \epsilon^2 N(\epsilon) d\epsilon = \frac{1}{4},$$

where $N(\epsilon) = \sum_k \delta(\epsilon - \epsilon_k)$. This requirement yields the analytical expression

$$t_\alpha = \frac{1}{2\sqrt{4 + 2\alpha^2}}.$$

Fig. 7 in App. B1 displays the corresponding density of states (DOS) varying continuously from three to two dimensions (see also Ref. 33). The DOS of a hypercubic lattice in infinite dimensions is—as a result of the central limit theorem—a Gaussian. In 3D, the DOS is still reasonably close to that Gaussian, yet with a flattened top. Going to 2D, however, two significant changes appear: The flat top narrows into a single peak (a logarithmic divergence: the so-called van-Hove singularity), whereas the sides develop “knees” (which become van-Hove singularities in 1D). We note that the movement of the knees towards the band-edge and the narrowing of the central plateau is somewhat linear with the change in dimensionality, while the increase in height of the narrowing plateau starts slow, and only speeds up significantly below $\alpha \leq 0.5$, i.e. for an effective dimension $D \leq 2.5$. Note that, in this work, we neglect all complications introduced by next-nearest neighbour hoppings or ionic potentials.

2. Dynamical vertex approximation

In order to be able to reliably estimate how non-local correlations in the self-energy in Eq. (1) affect the space-time-separation of the self-energy, we use a diagrammatic extension of DMFT, the dynamical vertex approximation (DFA) in its most used ladder-implementation as the associated fluctuations. For further technical details on the diagrammatic extension of DMFT, see App. A.
The oxygen octahedra surrounding the transition metal $M$. The resulting orthorhombic structures typically exhibit an increased degree of electronic correlations. Indeed, for $d^1$-perovskites, the ensuing lifting of degeneracies quenches orbital fluctuations and triggers a Mott transition$^{50,51}$. However, this physics is not driven by inter-orbital hybridizations but dominated by changes in local crystal-fields, which do not directly affect the value of $\alpha$. Indeed, for the Mott insulator YTiO$_3$ $\alpha \approx 1$ despite pronounced deviations from a perfect cubic structure.

Dimensional effects captured by Eq. (2) are instead crucial in oxides of copper: We find $\alpha \approx 0.4$ for (non-superconducting) Li$_2$CuO$_2$, while the two- and one-layer high-$T_c$ parent compounds La$_2$CuO$_4$ and HgBa$_2$CuO$_4$ have consecutively smaller $\alpha$. The quasi two-dimensional physics in cuprates seems to be indelibly connected with the occurrence of high-temperature superconductivity under doping (see, e.g., Ref. 53). Indeed, structural (and also chemical) variations will lead to trends in various electronic degrees of freedom, and empirical connections between the latter and $T_c$ have been established. Among these (interlinked) indicators are in-plane next-nearest neighbour hoppings$^{34}$, the charge-transfer energy$^{54,55}$, the $e_g$-splitting$^{56,57}$, and the magnitude of in-plane magnetic exchange couplings$^{58}$. Here, we note that, for the shown cuprates, the smaller $\alpha$, the larger $T_c$ at optimal doping.

Also in the case of the iron pnictides and chalcogenides (in their tetragonal structures), the dimensionality parameter $\alpha$ follows intuition: The 122-family is the most isotropic, the 111 less so, and the 11-chalcogenide is the most 2D-like, see Fig. 1. This trend neatly follows the magnitude of electronic correlations as monitored by, e.g., by the effective mass$^{59}$ or the fluctuating magnetic moment$^{59–61,62}$.

Comparing the $\alpha$-values of the families of cuprates and pnictides in Fig. 1, one notices that—contrary to common belief—the latter are overall as quasi two-dimensional as the former. As alluded to before, we stress that the measure $\alpha$ crucially depends on the choice of the orbital subset sought to represent the low-energy electronic structure. Comparisons of materials are therefore most adequate when they allow for a common orbital framework, thus, in particular, when the materials belong to the same family of compounds. Indeed, in Fig. 1, $\alpha$ is computed from the hoppings of the full $d$-shell of the transition metal. Contrary to the multi-orbital pnictides with nominal $d^9$-valence$^{59}$, the low-energy physics of $d^9$ cuprates is mostly dominated by a single orbital: the $d_{x^2-y^2}$ (see, however, e.g., Refs. 56, 63, and 64). The smaller orbital space limits the possibility for other dimensional effects beyond the purely geometrical ones. In Fig. 3 we therefore report $\alpha$-values for cuprates and the nickelate LaNiO$_2$ within said single-orbital framework: While trends within the cuprates remain qualitatively unchanged, the whole family substantially moves towards the 2D-limit. The anisotropy of LaNiO$_2$ surpasses 90%, but is still small as compared to the high-$T_c$
average over the Fermi surface rather than the full Brillouin zone (compare panels (g) and (h)), as it is expected in the presence of strong non-local spin fluctuations. At least at small doping the large scattering rate $\gamma$ impedes the interpretation of $Z$ as the quasi-particle weight. For instance, for $n = 0.975$ and $B = 15$ (data marked in red in Fig. 5), we find $\gamma_{loc} \approx 65\%$ of the 2D half-bandwidth, clearly invalidating the Fermi liquid picture. As a consequence, the fact that we find for this set of parameters the largest $Z_{loc} \approx 0.7$ does not contradict the fact of being the most correlated regime considered.

Next, we turn to the $\alpha$-dependence of the local expansion coefficients: When averaged over the entire Brillouin zone, both the $Z$-factor and the scattering rate $\gamma$ hardly change when the dimensionality is reduced. However, when limiting the momentum-average to the Fermi surface, i.e. to the region where low-energy excitations are actually present, there is a significant upturn in both $Z$ and $\gamma$ (panels (c) and (h)) when $\alpha$ drops below 0.5. This trend is consistent with the behaviour of the underlying DOS (shown in the Appendix’s Fig. 7, and discussed in Section II B 1; see also the supplementary material at low energies: Indeed, the density at the Fermi level drastically increases when the effective dimension drops below 2.5 (i.e. $\alpha \leq 0.5$), providing carriers available for electronic scattering and thus driving up the collision rate. In particular at our largest dopings, this change in the trend is almost cusp-like.

Finally, we address the momentum-dependence of the quasi-particle parameters. As a function of $\alpha$ the standard deviation throughout the Brillouin zone (following Eq. (11)) of both $Z$ and $\gamma$ slightly increases from 3D to 2D. Overall, however, their momentum-dependence is moderate in this measure. The momentum-variation of static shifts $\text{Re}\Sigma(k, \omega = 0)$, on the other hand, is already large in 3D, in accordance with previous results. When reducing $\alpha$, this spread increases continuously for all investigated dopings and temperatures. In the most correlated case ($n = 0.975$), the standard deviation $\Delta_{loc}^k\text{Re}\Sigma(k, \omega = 0)$ grows from 31% (3D) to above 50% (2D) of the respective half-bandwidth (cf. Fig. 7 and 70). Static non-local renormalizations are indeed non-negligible for all studied regimes.

The variation on the Fermi surface, measured via Eq. (12) reveals, instead, a notable enhancement towards lower dimensionality: While the standard deviation of $Z$ and $\gamma$ (panels (e) and (h)) is virtually zero in 3D, it acquires consistently growing finite values for smaller $\alpha$. The spread in both quantities is, however, significantly smaller than when averaged over the whole Brillouin zone (panels (d) and (g)).

Since the averaging of the measures Eq. (11) and Eq. (12) might obfuscate a strong momentum variation carried by only a few $k$-points, we plot in panels (c), (f), (i) the maximal spread on the Fermi surface according to Eq. (13). The increased momentum resolution of this analysis reveals a much larger variation than in the $k$-averaged data for $Z$ and $\gamma$, indicating that, as a matter
of fact, the momentum dependence must be driven by small areas of the Fermi surface, where large deviations from the local average are found.

As to the trend with doping, we must note that the behavior of the momentum-spread of the scattering rate on the FS is inverted with respect to that on the whole Brillouin zone: the largest momentum-spread is realized for the largest doping. More quantitatively, we find that the difference between the maximal and the minimal scattering rate surpasses 100% of the local value \( \gamma_{FS} \), further limiting its freedom.

Also for \( Re \Sigma(k,\omega = 0) \), the \( \alpha \)-dependence is quite different when looking at peak values (panel (c)): instead of a continuous increase for shrinking \( \alpha \), the variation peaks at intermediate dimensionality and diminishes towards 2D. We speculate that the following contributes to the reversal of the trend: \( Re \Sigma(k,\omega = 0) \) describes the deformation of the Fermi surface with respect to the local DMFT starting point. For a metallic solution (and low enough \( T \)), this deformation is subject to conserving the Fermi surface volume (Luttinger’s theorem) and \( C_4 \) symmetry in the ab-plane. Now, the decreasing dispersion along the c-axis introduces a strong geometric constraint that further impedes deformations (see also Appendix B). Furthermore we find, empirically, that

\[
\hat{e}_k \cdot \nabla_k Re \Sigma(k,\omega = 0)/\hat{e}_k \cdot \nabla_k e_k|_{k=k_F} > 0,
\]

suggesting that the sign of the Fermi velocity perpendicular to the Fermi surface determines the direction of deformation, further limiting its freedom.

b. Renormalizations on the Fermi surface. The two measures of non-locality on the Fermi surface—Eq. (12): FS-average, Eq. (13): maximal absolute difference—yield notably different results, suggesting largely inhomogenous renormalizations. Here we investigate this further by explicitly plotting in Fig. 6 \( Re \Sigma(k,\omega = 0) \) (left column, with respect to the Fermi surface average, \( 1/N_{k_F} \sum_{k_F} Re \Sigma(k_F, \omega = 0) \)), \( Z(k) \) (middle), and \( \gamma(k) \) (right) on the Fermi surface for the set of parameters given by \( n = 0.9, \beta = 20 \) and various \( \alpha \) (rows).
FIG. 6: Renormalizations on the Fermi surface: $\text{Re} \left[ \Sigma(k, \omega = 0) - 1/N \sum_{k_F} \Sigma(k_F, \omega = 0) \right]$, $Z(k_F)$, and $\gamma(k_F)$ for $n = 0.90$, $\beta = 20$ and representative values of $\alpha$ on the respective DGA Fermi surface.
In 3D ($\alpha = 1$, top row in Fig. 6), the values of the $Z$-factor and the scattering rate $\gamma$ are virtually homogeneous on the Fermi surface. The momentum-differentiation grows but remains overall small down to $\alpha = 0.5$. Below this mid-point between 3D and 2D, when the Fermi surface gets visibly quasi two-dimensional, we notice the emergence of a pronounced momentum-selectivity in $Z(k_F)$ and $\gamma(k_F)$. Akin to hole-doped cuprates, excitations at the anti-node ($(\pi, 0, k_z)$-direction and equivalent) become more short-lived than at the node ($(\pi, \pi, k_z)$). Recently, it has been suggested that such pseudogap physics is intimately linked to the proximity to a van-Hove singularity\textsuperscript{71,72}. In our model, such a feature exists for $\alpha = 0$ and is located at half-filling. Regarding the trend with doping, the variance throughout the Brillouin-zone is indeed largest for the smallest doping. However, as far as the momentum-selectivity of the scattering rate $\gamma$ and the quasi-particle lifetime $\tau = 1/(Z\gamma)$ on the Fermi surface is concerned, it is largest for the doping that places the chemical potential furthest away from the van-Hove singularity.

Non-local renormalizations also deform the Fermi surface. Here (left column in Fig. 6), we show the pertinent $\text{Re}\Sigma(k_F, \omega = 0)$ with respect to $\text{Re}\Sigma_{FS} = 1/N_{k_F} \sum_{k_F} \text{Re}\Sigma(k_F, \omega = 0)$. The latter Fermi-surface average can be seen as defining an effective local theory, obtained by a (partial) momentum-projection of the DFT self-energy. Following the arguments detailed in Appendix B, the Fermi surface expands (retracts) where $\text{Re}\Sigma(k, \omega = 0) - \text{Re}\Sigma_{FS} > 0 (< 0)$. In 3D ($\alpha = 1$), the Fermi surface is a body with tube-like openings in all axial directions. Non-local many-body effects shrink the tubes while expanding the inner core where the tubes intersect. When the hopping in $z$-direction decreases ($\alpha \leq 0.5$), the static non-local shifts of $\text{Re}\Sigma$ expand the Fermi surface near the $xy$-basal plane, but narrow the tubal forms oriented along the $z$-axis, thus magnifying the effective dimensional reduction for correlated electrons.

### c. Renormalizations at finite energies.

The fact that the self-energy coefficients have a larger variation over the entire Brillouin zone than over the Fermi surface ($\Delta_{k_F}^{\text{loc}} a(k) > \Delta_{k_F}^{FS} a(k)$) suggests that non-local effects are larger for excitations at finite energies, $|\epsilon_k| > 0$. For $\text{Re}\Sigma(k, \omega = 0)$ this can be rationalized as follows (using for simplicity a static self-energy): The momentum-derivative of $\text{Re}\Sigma(k)$ at $k_F$ reduces the effective mass (see, e.g., Eq. (1) in Ref. 24). Away from the Fermi surface to first order $\text{Re}\Sigma(k) \approx (k - k_F) \cdot \nabla_k \text{Re}\Sigma(k)|_{k = k_F}$. Assuming the derivative / the effective mass to be roughly constant, this effect increases the band-width via a shift $\text{Re}\Sigma(k) - \mu > 0$ (for $\epsilon_k > 0$) that grows (linearly) with the distance to the Fermi surface. In this sense of widening dispersions, non-local correlations counteract local (dynamical) correlations.

### IV. PERSPECTIVE AND CONCLUSIONS

When tackling a many-body problem—a model Hamiltonian or a strongly correlated material—knowledge about strength, nature and structure of correlation effects is a prerequisite for choosing the most efficient, yet adequate methodology: Does the system allow for a perturbative\textsuperscript{75,76} or weak-coupling\textsuperscript{77,78} treatment? Is the low-energy physics known to be dominated by a single microscopic process or fluctuation channel\textsuperscript{79}? Is potential pseudogap physics mostly driven by short-range fluctuations\textsuperscript{80,81}? Does the self-energy verify certain properties: Is it (to a sufficient degree) local (à la DMFT\textsuperscript{21,23}), static (à la Hartree-Fock, DFT+U\textsuperscript{82}), or does it approximately obey space-time separation\textsuperscript{24}, Eq. (1): $\Sigma(k, \omega) = \Sigma_{\text{static}}(k) + \Sigma_{\text{local}}(\omega)$? The validity of Eq. (1) in 3D for the Hubbard model\textsuperscript{24} and isotropic cubic materials\textsuperscript{26}, advocates that the realistic methodology DFT+DMFT (that combines DMFT with density functional theory) can be vastly improved by supplying it with an adequate static but non-local potential ($\Sigma_{\text{static}}(k)$ beyond DFT). The latter highlights the merits of approaches such as GW+DMFT\textsuperscript{83,25,84}, QS$GW+$+DMFT\textsuperscript{85,86}, S$Ex$+DMFT\textsuperscript{87}, or space-time separated GW\textsuperscript{24}, in all of which the non-local contribution to the self-energy (in the Wannier sense) is empirically found to be essentially static at least at low energies.\textsuperscript{88}

As a consequence, it is valuable to know the limits of validity of Eq. (1). Our results suggest that the error incurred by assuming space-time separability of the self-energy becomes prohibitive at an anisotropy corresponding to about half-way ($\alpha \approx 0.5$) between the isotropic 3D and the planar 2D case.

Our findings hence support the view (the expectation) that in cuprates ($d^9$ nickelates\textsuperscript{85,89,90}) no local theory can account for the rich many-body physics at play. Clearly, in 2D the self-energy does not obey space-time separation. At least at half-filling other simplifications may, however, apply for the Hubbard model.\textsuperscript{91}

The iron pnictides and chalcogenides reside in an $\alpha$-regime that suggests the presence of sizable non-local renormalizations, see Fig. 1. Concurrently, diagrammatic fluctuation techniques recently demonstrated\textsuperscript{92,93} that static non-local renormalizations are indeed non-negligible—in line with previous experimental analyses\textsuperscript{94}. Non-local corrections to dynamical renormalizations, however, were previously shown\textsuperscript{95} to be small, at least away from the systems’ Néel temperatures. These recent results substantiate earlier claims based on perturbation theory, and strengthen the space-time separability of the self-energy in the iron pnictides and chalcogenides proposed in Refs. 44 and 85\textsuperscript{96}.

According to the anisotropy classification of materials in Fig. 1, we ascertain that perovskite oxides are firmly in the realm of Eq. (1): dynamical self-energies will be essentially local. However, we found static non-local renormalizations (beyond DMFT) to be non-negligible also for cubic systems—motivating the above approaches to in-
clude a $\Sigma_{\text{static}}(k)$ (beyond DFT). Yet, most importantly, our findings strongly suggest that geometric constraints in (simulations of) oxide-based heterostructures\textsuperscript{97–99} or ultra-thin films\textsuperscript{12,100–104} may result in strong dynamical non-local effects—posing in particular limits on the applicability of DMFT. We believe that our anisotropy measure of Eq. (2) can provide guidance to identify such systems prior to actual many-body calculations. Moreover, our findings call for reexamining layered systems such as sodium cobalates and 124-ruthenates with realistic many-body approaches beyond DMFT\textsuperscript{7,25,26}.

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Appendix A: Details on the dynamical vertex approximation calculations

Diagrammatically the approximation made in DMFT is assuming the one-particle irreducible self-energy to be purely local also in finite-dimensional systems: $\Sigma(k, i\omega_n) \approx \Sigma(i\omega_n)$. The dynamical vertex approximation (DΓA)\textsuperscript{7,36} raises this assumption to the two-particle analog of the self-energy, the fully irreducible two-fermion scattering vertex $\Lambda_{i\omega_n,i\omega_n'}^{\nu \nu'} \approx \Lambda_{i\omega_n,i\omega_n'}$. This leads to a systematic inclusion of non-local correlations on every length scale. Without an \textit{a priori} knowledge of which physical scattering channel dominates the physics\textsuperscript{79,105}, for DΓA calculations the (computationally very demanding) parquet equations have to be solved self-consistently. In this work, we restricted ourselves to non-local correlations stemming from magnetism. Then the DΓA equations can be considerably simplified, since the full parquet DΓA can be restricted to its single-shot ladder (Bethe-Salpeter) version with Moriyaesque $\lambda$-corrections in both the charge and the spin channel channel [Eq. (6) in Ref. 38]. We used the code available at Ref. 106. We obtained the necessary two-particle Green’s function after a self-consistent DMFT calculation from an exact diagonalization impurity solver (with four bath sites), whose results we carefully checked against the ones from continuous time quantum Monte Carlo\textsuperscript{107}. For the Bethe-Salpeter ladders and Dyson-Schwinger equation we used a momentum grid with a maximum linear mesh sizes of $N_q = 60$ and $N_k = 20$ and the total number of fermionic as well as bosonic Matsubara frequencies being $N_{i\omega_n} = N_i\Omega = 120$. We calculated the self-energy for $N_{k,\Sigma} = 2601$ points in the Brillouin zone.

Appendix B: Effects of Re$\Sigma$($k_F, \omega = 0$)

The Fermi surface (FS) using the FS-averaged DΓA self-energy, $\text{Re}\Sigma^{FS} = 1/N_{k_F} \sum_{k_F} \text{Re}\Sigma(k_F, \omega = 0)$, is given by

$$\epsilon_{k_F} + \text{Re}\Sigma^{FS} = 0. \quad (B1)$$

Here and in the following we shall always absorb the chemical potential $\mu$ in Re$\Sigma$. Since FS-averaged DΓA defines a local theory, the Fermi surface is by construction the same as within DMFT. The Fermi surface of the full DΓA solution is obtained as:

$$\epsilon_{k_F} + \text{Re}\Sigma(k_F) = 0 \quad (B2)$$

$$\epsilon_{k^0_F} + \text{Re}\Sigma(k^0_F) + \delta_k (\epsilon_k + \text{Re}\Sigma(k))_{k=k^0_F} = 0 \quad (B3)$$

where we linearized around the above $k^0_F$: $\delta_k = k_F - k^0_F$ (and, for keeping the notation light, omitted complications introduced by the directional dependence of the momentum-derivatives). Now we introduce

$$\Delta\text{Re}\Sigma(k^0_F) = \text{Re}\Sigma(k^0_F) - \text{Re}\Sigma^{FS}$$

and use Eq. (B1), then:

$$\Delta\text{Re}\Sigma(k^0_F) + \delta_k \partial_k (\epsilon_k + \text{Re}\Sigma(k))_{k=k^0_F} = 0 \quad (B4)$$

yielding the Fermi-surface deformation

$$\delta_k = \Delta\text{Re}\Sigma(k^0_F) \times \frac{m^* Z}{m v^*_k} \quad (B5)$$

where we used the definition of the effective mass (e.g., Eq. (1) in Ref. 24). We find $\forall v \forall k \in \text{FS}: \epsilon_k \cdot \nabla_k \epsilon(k) > 0$ as expected for hole-doping. Hence the sign of $\Delta\text{Re}\Sigma(k^0_F)$ determines whether for that $k$ the FS expands (+) or shrinks (−).

* tomczak.jm@gmail.com

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FIG. 7: Density of states $N(\epsilon)$ of the tetragonal dispersion of Eq. (4) for the dimension $D$ varying continuously between 3D ($\alpha = 1$) and 2D ($\alpha = 0$).
A notable outlier is LaFeAsO (see Fig. 3), which in our measure, Eq. (2), is the most 2D-like pnictide considered. This finding is congruent with the fact that the Fe-Fe distance in this 1111-compound is larger by about 50% than in the other pnictides considered. However, effective mass renormalizations in LaFeAsO is comparable to BaFe$_2$As$_2$, the most 3D-like pnictide here. This deviation in the trend is ascribed to the lower value of the screened interaction in the 3d-orbital manifold of the 1111-material.

As a caveat to the trends described below, let us note that with decreasing $\alpha$, the system is located further and further away from the spin-ordered phase. Therefore, proximity effects are expected to be stronger closer to 3D than for (quasi-)2D, which should be considered when discussing the overall effect of changes in the dimensionality.

For $\alpha < 1$ magnetic transition temperatures $T_N$ displayed as shadings are only guides to the eye. The $T_N$-curve for $\alpha=1$ has been reproduced from Refs. 24 and 42.

Indeed, we previously noted in 3D that in this regime $\gamma$ involves corrections in Fermi liquid theory in its temperature dependence, as neither DMFT nor DΓA verify a $T^2$ behaviour.

See Supplemental Material available under ...
