Emergence of Quasiparticle in Doped Mott Insulators

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Doping a Mott insulator into a weakly coupled metal remains central to understanding various correlated emergent phenomena. To interpret this transition and its connection to the high-$T_c$ cuprates, we study the evolution of the single-particle spectrum for the Hubbard model using cluster perturbation theory on superclusters. With extremely low doping, we identify a heavily renormalized quasiparticle that immediately develops, crosses the Fermi level, and coexists with a polaronic band. Its spectral weight roughly grows at twice the rate of doping in the low doping regime, but this rate is halved at optimal doping. At the heavily doped regime, we find both strong electron-hole asymmetry and a vestigial presence of Mott spectral features. At last, we discuss the scope of single-band Hubbard model in describing the experimentally measured nodal spectral evolution of ARPES on La$_{2-x}$Sr$_x$CuO$_4$ ($0 \leq x \leq 0.15$). This work benchmarks the predicting power of the Hubbard model regarding the electronic properties of cuprates.

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One of the most important questions in the field of quantum materials is how the quasiparticles emerge from a Mott insulating state [1, 2]. This is a key prerequisite towards understanding the development of high-$T_c$ superconductivity via hole or electron doping from the Mott phase [3, 4], and further, why the physics of cuprates is different from other doped Mott insulators. With the single-particle momentum and energy spectrum, angle-resolved photoemission (ARPES) has a long history of addressing this question [5], which has by and large helped to determine a starting model for cuprates – the doped Hubbard or $t-J$ model. Early on it was shown that strong correlations, in the form of polaronic deformation (AFM) state from a predominantly Cu $3d^9$ configuration [6]. When a single hole is created by photoemission, it disperses from $(\pi/2, \pi/2)$ towards the $\Gamma$ point, which abruptly falls in intensity near the zone center – the so-called “waterfall” [7–10]. Rather than dispersing as a free or slightly renormalized quasiparticle, the band dispersion can be well fitted with a velocity on the scale of the spin exchange $J \sim 120$ meV, and is well described by the $t-J$ model via the concept of “spin polaron” [11–14].

However, this notion fails to describe the linewidth of the spectral function, which has observed to be rather broad in energy even at $(\pi/2, \pi/2)$ [7, 15], whereas a single hole in the $t-J$ model has no phase space to decay and should be resolution-limited sharp [11, 12]. This conclusion holds even as more realistic band structure or electron interaction parameters are considered [16–19]. Such abnormally broad energy linewidth was later captured by considering the lattice phonon polaron effect [20–23]. Coupled with the observed doping-induced spectral intensity developing distinctly from a higher-binding-energy sideband, it is believed that doped holes evolve from strongly polaronic at low doping to become less dressed by lattice distortions at higher dopings due to metallic screening [24–26].

Yet in many doped Mott insulators, such as the nickelates [27], manganites [28], and cobaltates [29], these doped carriers cause well-ordered charge structures in the form of stripes, which are insulating rather than superconducting as in the cuprates [30]. While stripe order indeed has been observed in cuprates (such as La$_{2-x}$Ba$_x$CuO$_4$ and La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$), the magnitude of charge density modulation is nowhere as strong as in other transition metal oxides [31]. This difference also led to the consideration of many material-specific degrees of freedom beyond the prototype Mott insulator, including static and dynamic lattice effects [32–40], as well as extra phenomenological parameters [41, 42]. Therefore, from the aspects of both cuprate itself and generic correlated materials, it becomes important to fully dissect the impacts of electronic correlation on
FIG. 1: Spectral function of the Hubbard model calculated using CPT: (a) for the half-filled Mott insulator and (b,c) for the 87.5% hole- and electron-doped system, respectively. The dotted line in (b) and (c) denotes the non-interacting tight-binding dispersion, while the horizontal dashed lines mark $E_F$. The Brillouin zone (BZ) cartoon shows the spectral cut between the $\Gamma$, $X$, and $M$ high-symmetry points for the square lattice. Labels in (a) denote the “Mott features”.

Thus, to identify all key features associated with electronic correlations and their evolution with doping, we systematically study the single-particle spectral features of a doped Hubbard model, with only $t$, $t'$ and $U$ and no other external ingredients. By calculating the spectral function at extremely fine doping levels, we expect to unbiasedly decipher the doping evolution of the quasiparticle dispersion, weight, and lineshape, and compare with ARPES experiments in cuprates. A better understanding of these spectral properties may provide insight on what aspects of the Hubbard model can well represent the data, and which aspects may be missing.

The Hamiltonian of the single-band Hubbard model is given by [50, 51]

$$\mathcal{H} = -\sum_{i,j,\sigma} (t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U \sum_i \left(n_{i\uparrow} - \frac{1}{2}\right) \left(n_{i\downarrow} - \frac{1}{2}\right).$$

Here, $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) and $n_{i\sigma}$ denote the creation (annihilation) and density operators at site $i$ of spin $\sigma$, respectively; $U$ denotes the on-site Coulomb interaction; and $t_{ij}$ encodes the electron hopping, restricted here to nearest-neighbors $t_{ij} = t$ and next-nearest-neighbors $t_{ij} = t'$. We chose parameters $U = 8t$ and $t' = -0.3t$, common for simulations of cuprates, and a Lorentzian broadening of 0.15t. Historically, a variety of numerical techniques have been used to investigate the single-particle spectrum of the Hubbard model, e.g. exact diagonalization [52–54], quantum Monte Carlo [55, 56], density-matrix renormalization group [57, 58], dynamical mean-field theory [59–62], CPT [63, 64] and others [65–69]. To investigate the low-temperature, finely momentum-resolved and continuously doping-dependent spectral features, CPT with superclusters is the most suitable approach. We evaluate the $4 \times 4$ cluster spectral function by an exact solver and $8 \times 8$ or $12 \times 12$ by a supercluster solver.

Figure 1 shows the calculated spectral function $A(k, \omega)$ at two extreme dopings: undoped (half-filling) and heavily doped. At half-filling, a large Mott gap separates the lower- and upper-Hubbard bands. Within each band, there are two main spectral features [see the markers in Fig. 1(a)]: one at low binding energies, describable within a spin-polaron framework [11, 14]; and a second at higher energies, which results from an effective intra-sublattice hopping [16, 18]. A “waterfall”-like step connects the above two features, constituting one of the critical spectral signatures of correlation effects. Throughout this Letter, we refer these single-particle features that exist already at the half-filled Hubbard model as the “Mott features”. In the other extreme doping limit [see
FIG. 3: (a) Energy distribution curves (EDCs) for calculated spectral functions with 1.4% hole doping. The inset shows the dashed grey area with 5x scaling. (b) False-color plot for $A(k,\omega)$ near $E_F$ along high-symmetry cuts, as indicated by the inset, for a fixed energy window and various electron concentrations with hole doping. (c,d) Same as (a,b) but for electron doping instead. In (a) and (c), the green arrows denote the quasiparticle features, while the red and blue arrows denote the remnant spin-polaron features at the lower and upper Hubbard band, respectively. In all panels, the gray dashed line denotes $E_F$.

Figs. 1(b) and (c)], the spectrum resembles that of non-interacting electrons, although the Hubbard $U$ remains unchanged [2, 4]. A quasiparticle dispersion across $E_F$ dominates the spectral function, with indiscernible residual spectral weight on the other side of the Mott gap. This feature, to which we refer as the quasiparticle, follows the tight-binding functional form of the bare band structure, but is subject to a doping-dependent bandwidth renormalization [19].

Between these two limits, we first investigate the density of states (DOS) as a function of hole- and electron-doping [see Fig. 2]. We see that a remnant Mott gap exists at all doping levels and well separates the upper and lower Hubbard bands. On top of it, infinitesimal amount of carrier introduction leads to the development of spectral weight at $E_F$ for both electron and hole doping. With the increase of doping, the spectral weight transfer gradually depletes the upper (lower) Hubbard band, and the chemical potential smoothly evolves away from half-filling (with a finite linewidth, one can still define a $\mu$ at $n = 1$). In this process, the transferred spectral weight becomes energetically mixed with the lower (upper) Hubbard band upon doping, rather than forming an entirely separate in-gap state [20, 70]. To quantify the spectral weight evolution, we integrate the three shaded regions: residual lower and upper Hubbard bands (red and blue) and the region between the Mott gap and the Fermi energy $E_F$ (green), respectively [see Fig. 2(c)]. The rapid growth of the spectral feature near $E_F$ involves spectral weight transfer from both the lower and upper Hubbard bands via doping. Our analysis shows that initially the spectral weight changes as $2.12x$ (where $x$ is the concentration of doped carriers), but is reduced to $0.84x$ at roughly optimal doping. This gradual change reflects the unraveling of electronic correlations upon doping, and presents a much more detailed doping perspective to the conventional description of $2x$ [70–72].

The fact that spectral weight appears near $E_F$ immediately upon doping suggests that the quasiparticle already develops at this point. To resolve this development, we calculate the momentum-resolved single-particle spectral function $A(k,\omega)$. Fig. 3 shows its doping dependence along high-symmetry cuts. In contrast to the back-bending spin-polaron at half-filling, doping immediately leads to the appearance of spectral weight near $E_F$ [also see Fig. 5 for experiments], with a heavily renormalized quasiparticle dispersion consistent with the ARPES experiments on the underdoped cuprates [15, 20, 23, 48]. The spectral weight of this quasiparticle grows monotonically with doping: it gradually “fills-in” near the $M$-point for hole doping, and the $\Gamma$-point for electron doping. A clear suppression of the antinodal spectral weight at low hole doping reflects the pseudogap, while a similar suppression of the node at low electron doping indicates the hotspot [see Supplementary Materials [73]]. The
appearance of these two distinct phenomena suggests that the major normal-state quasiparticle features at optimal doping [5, 74] may also be qualitatively captured by a Hubbard model. For both hole and electron doping, the renormalization gradually decreases, and the quasiparticle dispersion smoothly evolves into the free dispersion shown in Figs. 1(b) and (c).

The difference between the Hubbard model calculation and ARPES spectrum mainly lies in the lineshape near \( k_F \). In contrast to a broad peak describable by the strong polaronic dressing at half-filling and light doping [20], the renormalized quasiparticle in Fig. 3 is always sharp near \( (\pi/2, \pi/2) \) for hole doping [also see the EDC cuts in the Supplementary Material[73]]. That means the linewidth change in the underdoped regime cannot be simply attributed to the quasiparticle dressed by spin excitations in the doped Hubbard model. A phonon polaron or correlation-enhanced polaronic dressing is required to reproduce the experimental lineshape [21]. The omission of the phonon dressing also accounts for the huge chemical potential jump \( \sim t \sim 300 \text{meV} \) in Fig. 2(b), which should be smoother in cuprate experiments [75].

Besides the qualitative spectral shape, we perform quantitative analysis of the spectral weight doping evolution in two distinct momenta and respective energy ranges. We first focus on the \( M \)-point, where the quasiparticle is most separated from the higher-energy Mott features [see Fig. 1 and Fig. 3(b)]. In Fig. 4(a), we observe rapid growth in the spectral weight associated with the quasiparticle (green), overwhelming the remnants of the lower Hubbard band (red) at a moderate hole concentration. Residual spectral weight of the latter features gradually decreases with doping until \( \sim 20\% \) [see Fig. 4(b)]. The visibility of these Mott features in doped systems has two interesting implications: on the one hand, the coupling between carriers and spin fluctuations is present even in a regime without the long-range magnetic order, consistent with several recent experimental observations [76–79]; on the other hand, the final vanishment of these Mott features may account for the transition to a more metallic phase at \( \sim 20\% \) doping in \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \) [35, 40] and \( \text{YBa}_2\text{Cu}_3\text{O}_6 \) [49].

A similar analysis for electron doping, now at the \( \Gamma \)-point, is shown in Fig. 4(c). The Mott features persist until even higher doping. Although the finite-size effect in CPT precludes definitive assessment of order, the correlation effects at heavy doping suggest a substantial impact of short-range spin correlations. This matches the recent discovery of Fermi surface reconstruction outside the AFM phase in \( \text{Nd}_{2−x}\text{Ce}_x\text{CuO}_4 \) [44]. Here, the visibility of Mott features at higher doping (up to \( \sim 40\% \)) than the hole-doped side reflects the more robust magnetic correlations in the electron-doped Hubbard model [80–82]. To resolve the above dichotomy within occupied states and to compare our observations directly with experimental measurements, we focus our attention on the spectrum near \( k_F \) along the nodal direction, where the quasiparticle penetrates \( E_F \). Contrasting the spectral weight at two momentum-energy windows – one close to \( E_F \) and a second at higher binding energy – we can quantitatively distinguish the evolution of the quasiparticle spectral weight from the Mott features (specifically spin-polaron in the calculation) at low doping. As shown in Fig. 5(d), following a rapid exchange of spectral weight below 2% hole doping, both features begin to saturate and coexist (with a relative intensity close to 1). We also observe consistent behavior in experimental spectra at the extremely underdoped regime of the \( \text{La}_{2−x}\text{Sr}_x\text{CuO}_4 \) [see Fig. 5(a–c)]. After the immediate development of quasiparticle at 1% doping, the spectral weight ratio between the quasiparticle and polaronic feature saturates in a large range of doping. This agreement confirms our theoretical prediction that strong correlations continue to

FIG. 4: (a) EDCs at the \( M \)-point for different hole doping levels. The green shaded region highlights the growth of the quasiparticle, while red marks residual Mott features in the lower Hubbard band. (b) Integrated spectral weight of features in (a). (c) A similar analysis performed in electron-doped systems at the \( \Gamma \)-point instead, where blue marks the corresponding integrated weight in the upper Hubbard band.

FIG. 5: (a–c) ARPES experiments on underdoped \( \text{(La, Sr)}_2\text{CuO}_4 \) for half-filling, 1% and 12% doping near \( k_F \). The red and blue boxes in (b) denote the lower and higher energy windows near \( k_F \), corresponding to the quasiparticle and polaronic feature, respectively. (d) The spectral weight ratio between the lower and higher energy windows obtained from calculations (open circles) and experiments (solid squares). The shaded region denotes the AFM phase in \( \text{(La, Sr)}_2\text{CuO}_4 \).
play an essential role on the quasiparticle up to relatively high doping levels.

In summary, we have presented a comprehensive benchmarking study of the single-particle spectral function of cuprates upon hole and electron doping, from the perspective of the Hubbard model. By dissecting different spectral features originated from distinct origins, we analyzed the doping evolution of them. Many of the observations match with experiments on a qualitative level. Starting from an extremely underdoped regime, doped carriers induce quasiparticle-like states near the Fermi level. Both the momentum-resolved calculations and ARPES experiments reveal that this itinerant quasiparticle has no analog in the Mott insulating state at half-filling, including the Mott gap, spin-polaron, and the high-energy intra-sublattice features. Instead, it coexists with these Mott features in a wide range of doping. Though heavily renormalized and lightly-weighted at low doping, this quasiparticle gradually unravels from the Mott feature at a rate roughly twice that of the doping. At heavy doping (∼20% for hole-doping), the continued presence and electron-hole asymmetry of correlations are consistent with the observations in recent ARPES and RIXS experiments [43–46, 76–78].

From these aspects, the Hubbard model seems to effectively capture the essence of the emergence of low energy quasiparticles. However, in contrast to these qualitative consistencies in spectral weights and dispersions, the almost unchanged lineshape calculated from the Hubbard model cannot address the observed broadband in parent compounds or lightly doped cuprates. It also fails to reproduce the widely observed low-energy kinks at the nodal quasiparticle bands [32–34]. Towards a more comprehensive picture, additional lattice polaronic coupling may further act as another channel to destroy the quasiparticles’ coherence and itineracy via Franck-Condon principles, meanwhile contributing to the material-specific dependence both for various cuprate families and other transition metal oxides.

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[1] X.-G. Wen and P. A. Lee, Phys. Rev. Lett. 76, 503 (1996).
[2] P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. 78, 17 (2006).
[3] W. Stephan and P. Horsch, Phys. Rev. Lett. 66, 2258 (1991).
[4] E. Dagotto, Rev. Mod. Phys. 66, 763 (1994).
[5] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003).
[6] F. Zhang and T. Rice, Phys. Rev. B 37, 3759 (1988).
[7] F. Ronning, K. M. Shen, N. P. Armitage, A. Damascelli, D. H. Lu, Z.-X. Shen, L. L. Miller, and C. Kim, Phys. Rev. B 71, 094518 (2005).
[8] W. Meevasana, X. Zhou, S. Saharakorpi, W. Lee, W. Yang, K. Tanaka, N. Mannella, T. Yoshiida, D. Lu, Y. Chen, et al., Phys. Rev. B 75, 174506 (2007).
[9] T. Valla, T. Kidd, W.-G. Yin, G. Gu, P. Johnson, Z.-H. Pan, and A. Fedorov, Phys. Rev. Lett. 98, 167003 (2007).
[10] B. Xie, K. Yang, D. Shen, J. Zhao, H. Ou, J. Wei, S. Gu, M. Arita, S. Qiao, H. Namatame, et al., Phys. Rev. Lett. 98, 147001 (2007).
[11] G. Martinez and P. Horsch, Phys. Rev. B 44, 317 (1991).
[12] J. Bala, A. Oleś, and J. Zaanen, Phys. Rev. B 52, 4597 (1995).
[13] A. Macridin, M. Jarrell, T. Maier, and D. Scalapino, Phys. Rev. Lett. 99, 237001 (2007).
[14] E. Manousakis, Phys. Rev. B 75, 035106 (2007).
[15] K. M. Shen, F. Ronning, D. Lu, F. Baumberger, N. Ingle, W. Lee, W. Meevasana, Y. Kohsaka, M. Azuma, M. Takano, et al., Science 307, 901 (2005).
[16] Y. Wang, K. Wohlfeld, B. Moritz, C. Jia, M. van Veenendaal, K. Wu, C.-C. Chen, and T. P. Devereaux, Phys. Rev. B 92, 075119 (2015).
[17] M. Kohno, Phys. Rev. B 92, 085128 (2015).
[18] Y. Wang, K. Wohlfeld, B. Moritz, C. J. Jia, M. van Veenendaal, K. Wu, C.-C. Chen, and T. P. Devereaux, Phys. Rev. B 97, 199903 (2018).
[19] Y. Wang, B. Moritz, C.-C. Chen, T. P. Devereaux, and K. Wohlfeld, Phys. Rev. B 97, 115120 (2018).
[20] K. Shen, F. Ronning, D. Lu, W. Lee, N. Ingle, W. Meevasana, F. Baumberger, A. Damascelli, N. Armitage, L. Miller, et al., Phys. Rev. Lett. 93, 267002 (2004).
[21] A. Mishchenko and N. Nagaosa, Phys. Rev. Lett. 93, 036402 (2004).
[22] A. Mishchenko and N. Nagaosa, Phys. Rev. B 73, 092502 (2006).
[23] K. Shen, F. Ronning, W. Meevasana, D. Lu, N. Ingle, F. Baumberger, W. Lee, L. Miller, Y. Kohsaka, M. Azuma, et al., Phys. Rev. B 75, 075115 (2007).
[24] O. Rösch, O. Gunnarsson, X. Zhou, T. Yoshiida, T. Sasagawa, A. Fujimori, Z. Hussain, Z.-X. Shen, and S. Uchida, Phys. Rev. Lett. 95, 227002 (2005).
[25] C. Slezak, A. Macridin, G. Sawatzky, M. Jarrell, and K. Wohlfeld, Phys. Rev. B 98, 144503 (2018).
[79] K. Ishii, M. Fujita, T. Sasaki, M. Minola, G. Dellea, C. Mazzoli, K. Kummer, G. Ghiringhelli, L. Braicovich, T. Tohyama, et al., Nat. Commun. 5 (2014).
[80] T. Tohyama, Phys. Rev. B 70, 174517 (2004).
[81] B. Moritz, S. Johnston, T. Devereaux, B. Muschler, W. Prestel, R. Hackl, M. Lambacher, A. Erb, S. Komiya, and Y. Ando, Phys. Rev. B 84, 235114 (2011).
[82] Y. Wang, C. Jia, B. Moritz, and T. P. Devereaux, Phys. Rev. Lett. 112, 156402 (2014).