Orbital entanglement mechanism of superconductivity in cuprates

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We utilize a 1d Hubbard model to show that the superconductivity in cuprate superconductors likely arises due to the orbital entanglement between holes in the copper oxide plane mediated by orbitally-selective charge hopping. The main role of doping required to achieve superconductivity in cuprates is to suppress the Mott correlations and orbital ordering. The proposed mechanism explains superconductivity in other unconventional superconductors, and provides guidance in the search for new high-temperature superconductors.

After 35 years of intense research on superconducting cuprates, many theoretical models have been proposed \cite{1,2}. However, a generally accepted microscopic theory needed to reach one of the "holy grails" of condensed matter physics - superconductivity at room temperature at ambient pressure, has not yet emerged \cite{3}. The general features of superconducting cuprates are: i) superconductivity is hosted by the CuO\textsubscript{2} planes, ii) superconductivity requires doping parent compounds, which are antiferromagnetic (AF) Mott insulators, iii) superconductivity requires doping parent compounds, which are antiferromagnetic (AF) Mott insulators, iii) superconductivity requires doping parent compounds, which are antiferromagnetic (AF) Mott insulators, iii) the normal state at not too large doping is a correlated "strange" metal, which becomes increasingly "normal" at large doping, but the critical temperature is reduced, and iv) the superconducting correlation has a $d_{x^2-y^2}$ symmetry. Based on the observations ii) and iii), most of the theories have focused on the AF Mott correlations \cite{3}. Here, we develop a Hubbard model based only on the feature i), and show how the remaining features follow from it. Our model indicates that superconductivity results from the orbital entanglement that competes with the Mott state and orbital ordering, both of which are suppressed by doping. Our findings provide a blueprint for the search for superconductivity with high critical temperatures, and an explanation for superconductivity in many other unconventional systems including twisted multilayer graphene.

Before introducing our model, we review the electronic structure of the CuO\textsubscript{2} planes hosting superconductivity \cite{7}. These planes consist of a square lattice of copper atoms, with oxygen atoms positioned near the center between the neighboring Cu atoms to form intersecting Cu-O-Cu chains, Fig. 1(a). In the undoped state, the 2$p$ shell of O is completely filled, while the 4$s$ shell of Cu is empty and there is one hole per Cu atom in its 3$d$ shell.

In the molecular orbital approximation, the $C\text{\textsubscript{4}}$ symmetry of the Cu environment splits its five $d$-levels into three orbitally non-degenerate electronic levels derived from the orbitals with $M = \pm 2$ and $M = 0$, and a two-fold orbitally degenerate level $M = \pm 1$. In the absence of time-reversal symmetry breaking, each orbital level is also two-fold spin-degenerate. The highest-energy level $d_{x^2-y^2}$ is half-filled with one hole. The half filling and small hopping stabilize the Mott insulator state.

Antiferromagnetism arises due to the superexchange between the $d_{x^2-y^2}$ levels of the neighboring Cu atoms mediated by their hybridization with the oxygen’s $p$-orbital. For two Cu neighbors aligned along the x-axis, the $d_{x^2-y^2}$ orbitals form a σ bond with the $p_\sigma$ orbital of oxygen, Fig. 1(a). In the Mott-Hubbard language, virtual hopping of two holes from the neighboring copper atoms onto the shared oxygen atom is maximized (and kinetic energy is minimized) if their spins are antiparallel, stabilizing the AF state.

In our model, we explicitly account for the Cu-O bonding, and therefore must revise the molecular orbital picture described above. The symmetry-breaking effects of the environment of the CuO\textsubscript{2} planes (but not of the in-plane Cu-O bonding) can be approximated as a continuous $O(2)$ symmetry. Weak breaking of this continuous symmetry destabilizes the superconducting phase in favor of the Mott state, which is likely compensated by doping, as discussed below. The Cu $d$-orbitals are split by this symmetry into two two-fold orbitally degenerate levels $d_{\pm 2}$, $d_{\pm 1}$, and one orbitally non-degenerate level $d_0$. Similarly, oxygen’s $2p$ levels are split into a two-fold orbitally degenerate level $p_\pm$, and an orbitally non-degenerate level $p_0$, Fig. 1(b). The oxygen’s $p_0$ level is lower than the $p_\pm$ levels. The latter are slightly below the Fermi energy, so all the $p$-levels are filled. All the

Figure 1. (a) CuO planes hosting superconductivity in cuprate superconductors. The orbitals that in the molecular orbital picture dominate the Cu-O bonding are indicated for a Cu-O-Cu chain. (b) Atomic-level schematic of the copper $d$- and oxygen $p$-level energy splitting by the approximate $O(2)$ symmetry of the environment of the CuO\textsubscript{2} planes. Cu-O hybridization described in the Hubbard model by the hopping with coefficients $t_1$ and $t_2$ is indicated by arrows.
copper $d$-orbitals are also filled, except for the two-fold degenerate $d_{\pm 2}$ level hosting one hole. In this approximation, the Mott’s condition for half-filling is not satisfied. On the contrary, the metal-insulator transition is suppressed by the multi-orbital correlation effects that effectively enhance hopping, resulting in the emergence of a Hund’s-enhanced correlated metal state [8–10]. The effect is evaluated by analyzing the spatial overlap of the corresponding wavefunctions, as illustrated in Fig. 2 for the $d$-orbital correlations, which is essential for the proposed mechanism of singlet superconductivity in cuprates, as discussed below.

The corresponding Hubbard Hamiltonian is
\[\hat{H} = \sum_{p, \sigma} \epsilon_p n_{i \sigma} + \epsilon_d n_{j \sigma} + t_1 \hat{c}_{i \sigma}^\dagger \hat{d}_{j \sigma} + t_2 \hat{c}_{i \sigma}^\dagger \hat{d}_{j \sigma}^\dagger + U n_{i \uparrow} n_{i \downarrow} + (U - 3J) n_{i \uparrow} n_{i \downarrow}^\dagger + (U - 3J') n_{i \downarrow} n_{i \uparrow} + \text{h.c.} \quad (1)\]

where the pseudospin $\sigma$ labels the up/down orbital moments of the copper and oxygen state, the lines over symbols denote a complementary value, and up/down arrows denote the corresponding values of spin. Indices $i, j$ go over the nearest-neighbor oxygen and copper sites, respectively. "h.c." denotes hermitian conjugate for the two terms that are not hermitian. The operator $\hat{c}_{i \sigma}^\dagger$ annihilates a hole with spin $s$ in the $p_{\sigma}$ state on the $i$-th oxygen atom, $\hat{d}_{j \sigma}$ annihilates a hole with spin $s$ in the $d_{\sigma}$ state on the $j$-th copper atom, $n_{i \sigma} = \hat{c}_{i \sigma}^\dagger \hat{c}_{i \sigma}$.

The nature of the two-hole correlations is revealed by analyzing a three-atom chain that consists of two neighboring Cu atoms connected by an oxygen, as highlighted in Fig. 1(a). Exact diagonalization of the Hamiltonian Eq. (1) yields a two-fold degenerate ground state in the two-hole subspace (one hole per Cu atom) of many-particle Hilbert space. The projection of this state on the Cu $d$-orbitals has the form
\[\psi_{U(2)} = \frac{1}{\sqrt{2}} (|\uparrow \downarrow| - |\downarrow \uparrow|)(a|+\rangle + b|-\rangle), \quad (2)\]

where the two vertical arrows in the first pair of brackets denote the spin states of the two holes, while the two ± signs in the second pair of brackets denote the $d$-orbital states of the holes located on the two Cu sites. The state Eq. (2) is antisymmetric, i.e., the orbital part is symmetric, with respect to particle exchange. This is not obvious from the notations of Eq. (2), which are a mixture of single-particle notations for spin with site notations for the orbital part. Since the holes reside on different orbitals, the coefficients $a, b$ are only limited by the normalization $|a|^2 + |b|^2 = 1$.

The ground state Eq. (2) describes AF spin correlations, and AF orbital correlations of the two holes residing on the neighboring Cu atoms. Qualitatively, these correlations can be explained as follows. The two hopping terms describe virtual hole hopping from Cu onto O, while retaining its spin and, mostly, its pseudospin for $|t_1| > |t_2|$. The Mott’s coupling limits the hopping of another hole on the neighboring Cu atom, increasing its kinetic energy. The two-hole correlations that minimize the Mott’s coupling are i) spin-singlet that allows
the second hole to hop on both oxygen orbitals, and ii) the orbital anticorrelation described by the term in the second pair of brackets in Eq. (2) lowers the energy due to the Hund’s coupling on the oxygen site. We emphasize that \( t_1 \) and \( t_2 \) must be finite and different to stabilize the state Eq. (4). If, for example, \( t_1 = t_2 \), then the hopping hole loses its orbital information, reducing the correlations to a spin singlet without orbital entanglement. On the other hand, if \( t_2 \) (or \( t_1 \)) is negligible, then Hund’s coupling becomes dominant, stabilizing the spin triplet state. While such a state is not relevant to cuprates, it may describe some magnetic superconductors [11]. We comment more on this in the summary.

The orbital part of the ground state Eq. (2) is a \( U(2) \)-symmetric spinor with respect to the amplitudes \( a, b \). We now argue that hopping of itinerant charge carriers introduced in \( \text{CuO}_2 \) by doping, which is required to achieve superconductivity in cuprates, should reduce the symmetry of the ground state to the symmetry \( U(1) \) that ultimately leads to the superconducting state. Thus, the superconducting ”dome” on the doping-temperature diagram likely hides not one, but two quantum-critical points: one associated with the transition to the Mott state, another - with orbital magnetism.

While the effects of itinerant charge carriers are not included in our model, they can be assessed from the analysis of the correlation effects on the Mott state. For specificity, we consider a hole-doped system. In addition to one hole per \( \text{Cu} \) atom discussed above, an extra hole hops among the \( \text{Cu} \) atoms through the oxygens connecting them. If the magnitudes of \( a \) and \( b \) in Eq. (2) are different, the ”local” holes are orbitally polarized with the polarization magnitude \( |a|^2 - |b|^2 \), and with AF ordering between the nearest \( \text{Cu} \) neighbors. Hund’s coupling between an itinerant and a ”local” hole results in the orbital polarization of the itinerant hole opposite to the ”local” hole. The hopping of the itinerant hole onto the nearest \( \text{Cu} \) neighbor is then suppressed by the Hund’s mechanism, since its orbital polarization would then be the same as the orbital polarization of the ”local” hole on that atom. The state minimizing this increase of the energy of the itinerant holes is Eq. (2) with \( |a| = |b| \), i.e. an orbitally unpolarized state of the ”local” holes. The extension of this symmetry-based argument to electron doping is straightforward.

We arrive at the \( U(1) \)-symmetric two-hole state

\[
\psi_{U(1)} = \frac{1}{2} (|\uparrow\downarrow \rangle - |\downarrow\uparrow \rangle)(|+\rangle + e^{i\theta}|-)\]. \hspace{1cm} (3)

This state is remarkable in that all the components of both the spin and the orbital polarization vanish regardless of the value of \( \theta \), which is consequently unobservable and represents a gauge symmetry. This surprising property is ensured by the vanishing contributions of \( d_{\pm 1}, d_0 \) orbitals, such that the \( x \)- and \( y \)-components of orbital momentum, \( \langle L_x \rangle, \langle L_y \rangle \), identically vanish. A similar property holds for the \( p_\pm \) oxygen orbitals omitted from the \( \text{Cu} \)-projected state Eq. (2), because they are separated by \( \Delta M = 2 \). If a similar to Eq. (3) state was constructed from two orbital states that differ by \( \Delta M = 1 \), the in-plane orbital momentum would be determined by \( \theta \), so that either spontaneous or externally-induced symmetry breaking would result in orbital magnetism.

Superconductivity is an immediate consequence of Eq. (1), by an argument that can be formalized using e.g. the standard Green’s function techniques for the pair correlator described by Eq (3) following the same logic as the BCS theory of superconductivity. We leave such detailed calculations to future studies. Qualitatively, our analysis of the \( \text{Cu-O-Cu} \) three-atom chain shows that the gauge-symmetric ground state splits off below the \( d \)-level manifold due to two-hole correlations. For an extended \( \text{CuO}_2 \) plane, the collective effect of such pairwise correlations can be described in the mean-field approach as a decrease of this two-hole level’s energy with increasing density of correlated hole pairs. Spontaneous pair condensation, i.e. breaking of the gauge symmetry characterized by the parameter \( \theta \) in Eq. (4), occurs when the correlation energy gain outweighs the entropic contribution to free energy that increases with increasing correlations. It is easy to see that the gradient of \( \theta \) describes a spatial winding of the phase of the orbital wavefunction that carries a non-dissipative current.

We now discuss how the general features of superconductivity in cuprates, as listed in the introduction, naturally emerge from the proposed mechanism. ii),iii), the charge doping effects and correlations. Based on the above analysis, both the Mott’s insulating state competing with superconductivity, and the orbital magnetism are suppressed by doping, but doping also reduces the superconducting correlations due to the orbital dephasing by the itinerant carriers, resulting in the well-known superconducting dome on the temperature-doping diagram. iv), the \( d_{x^2-y^2} \) symmetry of the order parameter derives from the symmetry of the \( \text{Cu} \) \( d_{pm2} \) orbitals hosting the superconducting holes. The relative phases of the \( d_{+2} \) and \( d_{-2} \) orbitals on the neighboring \( \text{Cu} \) atoms are locked by the correlation Eq. (3). Both components change sign upon rotation by \( 90^\circ \), so the order parameter must be a superposition of \( d_{x^2-y^2} \) and \( dx y \), which have indistinguishable symmetry with respect to in-plane rotations. The correlations are non-chiral, so this superposition is real. Based on the proposed microscopic picture, the correlations are maximized along the \( \text{Cu-O-Cu} \) chains, singling out \( d_{x^2-y^2} \) as the dominant symmetry.

Based on our analysis, we summarize the ingredients essential for the superconductivity in cuprates, which can also serve as a blueprint in the search for better high-temperature superconductors.

i) The quasi-2d electronic structure is essential for the required partial lifting of the degeneracy of orbital levels, such that only two-fold degenerate orbital levels with
$\Delta M > 1$ remain near the Fermi level. If this condition is not satisfied, in-plane orbital magnetism becomes stabilized instead of superconductivity. From this perspective, $p$ orbitals may be the most advantageous, since their hybridization (and thus crystal field effects splitting the levels) can be made stronger than for the more localized $d$-orbitals, and only $p_0$ needs to be split off.

2) The approximate rotational symmetry of the effective crystalline field of the environment of the 2d system is essential for preserving the two-fold degeneracy of the "active" orbital level. The precise symmetry constraints warrant a separate analysis outside the scope of this work. The dimensionality of irreducible representations generally decreases with symmetry lowering, ultimately quenching the orbital states. Such quenched states are well-described by the molecular orbital approximation and cannot support orbital entanglement.

3) To reduce the $U(2)$ symmetry of the two-hole wavefunction to the gauge symmetry stabilizing superconductivity, orbital magnetism competing with superconductivity must be suppressed. In cuprates, this is accomplished by correlations mediated by itinerant carriers introduced by doping. A possible promising direction for improvement is to suppress orbital magnetism, without the need for doping, by geometric frustration.

4) The insulating Mott state that competes with superconductivity is stabilized when the two-fold orbital degeneracy of the "active" $d$-level hosting one hole is lifted, for example due to the low-symmetry of the environment of CuO$_2$ planes. Such effects are inevitably present in complex oxides such as cuprates. In cuprates, Mott correlations are suppressed by doping, which also suppresses the superconducting correlations mediated by orbitally-selective superexchange. Superconductivity can be enhanced by suppressing the Mott state without doping. This can be achieved by maximizing the orbital selectivity of hopping, $t_1 \gg t_2$, which maximizes the Hund’s orbital correlations responsible for superconductivity and reduces the Mott’s spin correlations. As a result, the spin correlations are expected to become ferromagnetic, i.e. such a hypothetical superconducting order parameter is a spin-triplet state.

The described orbital entanglement mechanism is likely relevant to other systems that exhibit unconventional superconductivity, including iron-based superconductors [12, 13] and twisted multilayer graphene [14]. In case of graphene, the superconductivity likely arises due to the entanglement of finite-orbital-momentum electron states on Moire sites. Moreover, ferromagnetism observed in this system in immediate vicinity, in the parameter space, to superconductivity [15] can be understood as a consequence of orbital correlations similar to those described by Eq. (2), but with a triplet instead of the singlet spin component, which is stabilized by the Hund’s interaction of an electron on a Moire lattice site with another electron that virtually hops directly from the neighboring site. In contrast to the unconventional (non-BCS) mechanism of superconductivity, this mechanism stabilizing ferromagnetism is the usual direct exchange. In this context, we assert, without proof here, that Stoner ferromagnetism in multiorbital systems such as late transition metals can be interpreted as condensation of similarly orbitally-entangled spin-triplet states of electron pairs, providing a unified description for both unconventional superconductivity and magnetism as condensation of two-particle correlations akin to Cooper pairs, but mediated by orbital entanglement instead of phonons. This raises a tantalizing possibility to achieve high-temperature superconductivity in systems incorporating quasi-2d planes of ordinary 3d transition metal ferromagnets. Simple electron counting suggests that 1 hole per transition metal atom, required for the described orbital correlations, is achieved between Co and Ni. However, orbital correlations required for superconductivity are suppressed by the significant 4s component of electron wavefunctions at the Fermi level and the spin-orbit interaction. A detailed analysis of these effects will be presented in a separate publication.

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Rejection letter from PRL is appended in full below.

Re: LJ17592 Orbital entanglement mechanism of superconductivity in cuprates by Alexander Mitrofanov and Sergei Urazhdin

Dear Dr. Urazhdin,

The above manuscript has been reviewed by our referees.

On the basis of the resulting reports, it is our judgment that the paper is unsuitable for publication in Physical Review Letters. We append comments from the criticism that led to our decision.

Yours sincerely,

Dr. Abhishek Agarwal
Associate Editor
Physical Review Letters

Report of Referee A – LJ17592/Mitrofanov ———

The authors argue that superconductivity in cuprates arises due to orbitally selective charge hopping which, in turn, originates from specific symmetry-breaking due to coupling between the copper-oxygen planes and their environment. I admit that the idea is quite interesting and might possibly be related to cuprates. However, the manuscript does not contain a single quantitative results which could be compared to experimental results or other theoretical studies. The discussion is highly speculative and is entirely based on qualitative arguments. The speculative character of the discussion is reflected in numerous statements, e.g. “likely arises”, “is likely relevant”.

The main result of the manuscript is the Hamiltonian (1) which should be relevant for copper-oxygen planes. It is a complicated tight-binding model with several orbitals and seven free parameters: \( \epsilon_p \), \( \epsilon_d \), \( t_1 \), \( t_2 \), \( U \), \( J \), \( J' \). The values of these parameters which could be appropriate for cuprates are not specified. Even if these parameters were known, studying such Hamiltonian on a two-dimensional lattice is technically extremely demanding. Previous studies failed to provide unquestionable results (numerical or analytical) for much simpler Hamiltonians, and it is not clear for me why Hamiltonian (1) should allow one to overcome such technical problems. On page 3 (left column) the authors suggest that one may apply a mean-field (BCS-like) approach to discuss superconductivity. Indeed such approach seems to be technically feasible and rather straightforward. Unfortunately, the authors “leave such detailed calculations to future studies” so it is not clear whether the emerging mean-field results may reproduce at least some experimental results for cuprates. I am not convinced that a mean-field approach applied to this particular Hamiltonian may explain the physics of cuprates especially that previous mean-field studies mostly failed.

The authors solve numerically a three-site problem (still for unspecified model parameters) and report that the ground state is given by Eq. (2). Then, again purely qualitative arguments are applied to account for the presence of itinerant carriers and to obtain Eq. (3). Further on, the authors claim that superconductivity is “an immediate consequence” of the latter equations. It is not clear to me, how the presence of superconductivity can be claimed just from a single ground state obtained for three sites. There are methods which allow to recognize the presence of pairing (which should not to be confused with superconductivity) from exact diagonalization of small systems. One compares the energy of systems containing different numbers of carriers but the studied system seems to be too small even for such calculations.

The reader might be also confused about the dimensionality of the studied problem. On page 3 the authors claim that quasi-2d structure is essential, in the abstract a 1d-Hubbard model is announced but the calculations are carried out for 3-sites which essentially represent a zero-dimensional system.

In summary, I think that the manuscript may be considered as an interesting proposal for future studies rather than as a description of clear results which may convince other researchers. In my opinion, the manuscript presents just a rough idea for the future studies and does not make any important contribution to the understanding of either cuprates or other unconventional superconductors. I cannot recommend it for publication.

Report of Referee B – LJ17592/Mitrofanov ———

This paper provides a theoretical study on the superconducting mechanism in cuprate high-temperature superconductors. Based on the one-dimensional Hubbard model, the authors claim to have found that “orbital entanglement” due to orbital-selective hopping is important for superconductivity. The authors also claim that the doping effect can be understood by suppressing the Mott correlation and orbital order.

However, there is little indication of why we should consider the "orbital entanglement" picture that is the subject of this paper, how it relates to many previous theories, and the objective evidence for the validity of their picture. It seems that the model used here is far from cuprates. In many places, discussion is made without adequate explanation or citation. Furthermore, the paper is not easy to read because the figures are not fully explained and there are no data, graphs, or tables.

For these reasons, I cannot recommend the publication of this paper.

The specific problems are as follows.

1) This paper proposes a physical picture of cuprate superconductors. However, it is unclear why we have to consider the picture of "orbital entanglement", which is the main subject of this paper. I think that the authors need to provide objective evidence for the following points based on analytical and numerical calculations...
and discussion with experimental facts, using graphs, pictures, or tables: (1) the advantages and limitations of this theory, (2) the validity of this theory, and (3) the relationship between this theory and many previous theories.

2) Since the main idea of this paper is "orbital entanglement", the information of the orbital is important. The authors introduce the orbitals "M=+2 -2" and "d+2 d-2" on page 1. I cannot find any description of the meaning of this number. Are they z-components of the orbital magnetization (=l_z)? Does M=2 and d+2 represent same orbital? If yes, the crystal field splitting of the Cu d-orbital in CuO2 plane is largely different from Fig. (b). I think authors ignore the effect of O2− ions in CuO2 plane. Therefore, Eq. (1) is not a good starting point of cuprates.

3) The authors discuss superconductivity based on Eq. (2). It is important to note that this is not a direct solution to the 2-dimensional Hubbard model Eq. (1), but a solution to the Cu-O-Cu cluster (0-dimensional). There is no objective reason why the ground state of the Cu-O-Cu cluster is valid for the ground state of the CuO2 plane in cuprates.

4) The authors' writing style is quite misleading. In relation to 2), the term "molecular orbital" is used to describe orbitals " +2 -2". However, I this it is not a molecular orbital but just a d-orbitals split by crystal field. In relation to 3), the authors state that they are discussing 2-dimensional CuO2, but, in the abstract, they refer to a 1-dimensional Hubbard model, but it seems that what they are discussing is just a 3-site cluster.

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Report of Referee C – LJ17592/Mitrofanov

The manuscript suggests novel orbital entanglement mechanism of superconductivity (S.C) observed in cuprates. Instead of the conventional one-band Hubbard model with crossing Fermi level of dz² − y² orbital, authors suggest the two-orbital (or four-orbital including oxygen hybridization) Hubbard model with 1/4-filling at Mott insulator of parent compounds, due to the multi-orbital correlations. They show the mechanism of S.C, through a solution of the three atom-chain (multi-orbital) Hamiltonian computed by an exact diagonalization method. I think that this suggestion might be the valuable to understand unresolved nature of S.C in cuprate, because high-Tc S.C is observed in some multi-orbital systems like iron-type materials. On the other hand, I am not sure how broadly interesting or novel this work is as a lot of studies have been already performed on single-band Hubbard model. To my knowledge, there is an understanding of the key to S.C within one-band Hubbard model. In addition, I am not sure whether the solution of the simple three atom-chain Hamiltonian provides an evidence of an obvious clue for S.C. Therefore, I suggest transfer of this paper to a specialized journal if my questions in below can be fully answered.

I have two questions.

(1) The DFT results indicate single-band system on parent compounds. On the other hand, in order to account for this mechanism the two-orbital model would be needed on parent compounds. Can the authors explain this inconsistency?

(2) In order to make Mott insulator of two-orbital system on parent compounds, larger values of U and J than bandwidth would be needed. On the other hand, value of U is comparable with bandwidth. Can the authors explain this inconsistency?

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