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Enigma of the vortex state in a strongly correlated d-wave superconductor

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We show that strong electronic repulsion transforms a vortex core from a metallic-type in overdoped regime to a Mott-insulator at underdoping of a strongly correlated d-wave superconductor. This changeover is accompanied by an accumulation of electron density at the vortex core towards local half-filling in the underdoped region, which in turn facilitates the formation of the Mott insulating core. We find that the size of vortices evolves non-monotonically with doping. A similar non-monotonicity of critical field H_{c2}, as extracted from superfluid stiffness, is also found. Our results explain some recent experimental puzzles of cuprate superconductors.

Introduction. Topological defects, such as, vortices have drawn significant research interests ever since Kosterlitz and Thouless [1, 2] established a melting mechanism mediated by them. Vortices are low-lying excitations of type-II superconductors in the presence of magnetic fields. In conventional superconductors, a magnetic field produces a periodic array of vortices [3, 4] with a normal metallic core of size ξ with circulating currents around the vortex on the scale of the penetration depth λ [5]. With increasing field H, the density of vortices increases. Beyond the critical field H_{c2}, overlapping cores suppress pairing amplitude everywhere and the superconductor transitions into a metal [3]. The study of vortices in unconventional superconductors has gathered recent momentum due to several experimental puzzles [6–8].

One such mystery lies in the mapping of local density of states (LDOS) at the vortex core in cuprate superconductors, a prototype of strongly correlated d-wave superconductors (dSC). Differential conductance in cuprates (both in $YBa_2Cu_3O_{7-\delta}$ [9] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ [10]) in optimal to underdoping region shows a gap structure, while weak-coupling calculations predict a large accumulation of low-lying states in LDOS at vortex core for all dopings, δ [11]. Recent experiments find similar significant pileup of the low-lying states but in the overdoped regime [7]. Several theoretical attempts have been made to understand the low doping anomalous behaviours [12–15], including the generation of sub-dominant competing orders at vortex core, such as antiferromagnetic [16–18], s-wave pairing [19], d-density wave [20, 21] and pair-density wave orders [22], augmented to weak-coupling descriptions. However, no consensus has yet been achieved to comprehend the anomaly [7, 23]. The role of strong correlations on the vortex inhomogeneities, however, have largely alluded the field of research, see however, [24–26]. After all, these strong electronic repulsions turn the parent undoped ($\delta = 0$) compound an antiferromagnetic Mott insulator [27].

Taking the route of direct inclusion of strong correlations by removing any double-occupancy within a fully self-consistent microscopic calculation, our main results in this paper are: (i) Underdoped d-wave vortex state induces charge accumulation towards local half-filling at the vortex core, and thereby promotes the emergence of 'Mottness'. (ii) The changeover of the nature of the vortex core from being Mott insulating to metallic with increasing doping, which explains the tunneling spectroscopic measurements of LDOS. (iii) The size of vortices show intriguing non-monotonic behavior. Such a non-monotonic behavior has other fascinating implications. For example, our result of superfluid density in the presence of magnetic field indicates that the upper critical field H_{c2}, shows a dome shaped evolution with δ, in agreement with experimental findings.

Model and methods. Strongly correlated materials can be described minimally by the Hubbard model [28] with $U \gg t$. In this limit the low energy physics is described by a $t - J$ model [29]:

$$\begin{aligned}\mathcal{H}_{\rm t-J} &= -t \sum_{\langle ij\rangle\sigma} \mathcal{P}\left(e^{i\phi_{ij}}\hat{c}^\dagger_{i\sigma}\hat{c}_{j\sigma} + \text{H.c.}\right)\mathcal{P} - \sum_i \mu\hat{n}_i \\ &+ J\sum_{\langle ij\rangle}\mathcal{P}\left(\mathbf{S_i}\cdot\mathbf{S_j} - \frac{\hat{n}_i\hat{n}_j}{4}\right)\mathcal{P}. \qquad (1)\end{aligned}$$

Here $\hat{c}^\dagger_{i\sigma}$ ($\hat{c}_{i\sigma}$) is the creation (annihilation) operator of an electron with spin σ at lattice site i in a two-dimensional square lattice, $\mathbf{S}_i$ and $\hat{n}_i$ are the spin and electron density operators, respectively, $\langle ij\rangle$ denotes nearest neighbor bonds, t is the hopping amplitude for an electron to its nearest neighbors, μ is the chemical potential fixing the average electron density ρ, $J = 4t^2/U$ is the super-exchange interaction with U being the onsite Hubbard repulsion strength. Here, $\mathcal{P}$ is the projection operator which prohibits double occupancies on each lattice site due to the strong onsite repulsive U. The orbital magnetic field is incorporated through the Peierls factor: $\phi_{ij} = \pi/\phi_0 \int_i^j \mathbf{A}.\mathbf{dl}$, where $\phi_0 = hc/2e$ is the superconducting (SC) flux quantum. We consider a uniform orbital field $\mathbf{H} = H\hat{z}$ and choose to work with the Landau gauge, $\mathbf{A} = Hx\hat{y}$.

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The effect of the projection operator is implemented by the Gutzwiller approximation (GA) [30], where restriction of double occupancy is removed in expense of renormalizing the hopping and exchange parameters: $t_{ij} \to g^t_{ij} t$, $J_{ij} \to g^J_{ij} J$, here g's are the corresponding Gutzwiller renormalization factors (GRFs) [30, 31]. GRFs, which depend on local densities n_i, are provided in the Supplementary Material (SM) [32]. Physically, the removal of double occupancy prohibits certain hopping processes across the bond $\langle ij \rangle$, and hence the average kinetic energy must reduce on that bond from a situation where double occupancies are allowed. This is incorporated by the hopping GRFs $g^t_{ij} \leq 1$. Similarly, the overall higher probability of sites being singly occupied enhances the exchange coupling through g^J_{ij}. The GA formalism has been verified [37, 38] to agree well with variational Monte Carlo calculations [39] (where the projections are exact) for homogeneous systems. We note that we refer to the strong correlations equivalently with the removal of double occupancy in this work.

We take advantage of the perfect periodicity of our square vortex lattice [59] by solving the eigenvalue problem using a fully self-consistent Bogoliubov de-Gennes (BdG) method on a unit cell typically of size $N = 24 \times 48$ and then extending the wavefunction on a system made of typically 16×8 unit cells [11, 17]. We present all energies in units of the hopping amplitude t and set the temperature $T = 0$ for our calculations. We use $J = 0.33$ – a typical value used for cuprate superconductors [40]. We consider several doping ($\delta = 1 - \rho$) values ranging from $\delta = 0.06$ (underdoped) to $\delta = 0.25$ (overdoped). To emphasize our key findings, we compared our results from Gutzwiller inhomogeneous mean-field theory (GIMT) with results from standard inhomogeneous mean-field theory (IMT), where the effects of projection $\mathcal{P}$ are ignored by taking the Gutzwiller factors to be unity, i.e., with double occupancy being allowed. In IMT, we tune J values for each doping in such a way that both IMT and GIMT yield the same d-wave gap when the magnetic field is zero [41]. The details of GIMT and IMT calculations are included in the SM [32].

d-wave SC order. We begin describing our results by elaborating on the dSC order parameter calculated within the GIMT framework: $\langle \hat{c}_{i\sigma} \hat{c}_{j\bar{\sigma}} \rangle_\psi \approx g^t_{ij} \Delta_{ij}$[42, 43]. Here $\langle .. \rangle_\psi$ denotes the expectation value in the truncated Hilbert space with double occupancies removed. The spatial profile of the dSC order parameter, $\Delta^{\rm OP}_{\rm d}(\mathbf{r}_i) = \frac{J}{4} |[g^t_{i,i+\hat{x}} \Delta_{i,i+\hat{x}} + g^t_{i,i-\hat{x}} \Delta_{i,i-\hat{x}} - e^{ibx} g^t_{i,i+\hat{y}} \Delta_{i,i+\hat{y}} - e^{-ibx} g^t_{i,i-\hat{y}} \Delta_{i,i-\hat{y}}]|$ (here $b \equiv H/\phi_0$) is shown in Fig. 1. Different panels of Fig. 1 show $\Delta^{\rm OP}_{\rm d}(\mathbf{r}_i)$ for representative δ. Away from a vortex core, i.e. near the boundary of the magnetic unit cell containing a single SC flux quantum, $\Delta^{\rm OP}_{\rm d}$ attains the homogeneous Bardeen-Cooper-Schrieffer (BCS) value while it falls at the vortex core. This conical-shaped fall at the core for overdoped [Fig. 1(a)] to optimally doped [Fig. 1(b, c)] systems follows the expected $\tanh(r/\xi)$ behavior, where ξ is the SC coherence length [5]. In contrast, the fall of $\Delta^{\rm OP}_{\rm d}(\mathbf{r}_i)$ shows a strikingly different

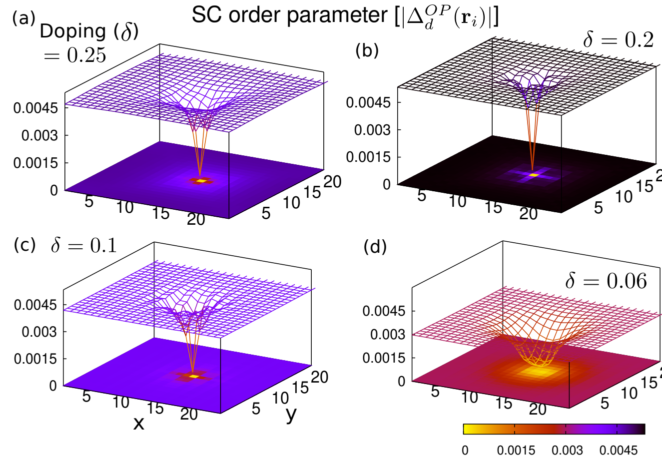

FIG. 1: **SC order parameter profiles.** d-wave SC $|\Delta^{\rm OP}_{\rm d}(\mathbf{r_i})|$ profiles around a vortex core on a magnetic cell of size 24×24 at different doping (δ) values. The fall of $\Delta^{\rm OP}_{\rm d}(\mathbf{r_i})$ at the vortex center has the conventional conical form at $\delta = 0.25$, 0.2, and takes up a form of a "flat-bottom bowl" at $\delta = 0.06$.

pattern at underdoping [Fig. 1(d)]: The region of the depletion of $\Delta^{\rm OP}_{\rm d}(\mathbf{r}_i)$ is much wider – near the core-center, the vortex resembles a "flat-bottom bowl". The weak-coupling IMT calculations preserve the conical-shaped vortex for all δ, and shrinks monotonically towards underdoping, see SM [32].

Local charge density at a vortex core. In order to develop a deeper insight into above results we next study the local charge density near the vortex core location, $\mathbf{r}_v$ [60] for different δ. In the optimally doped region ($\delta = 0.2$), the spatial density profile features a weak dip around $\mathbf{r}_v$ [Fig. 2(a)], consistent with the weak-coupling theory. Upon lowering δ, the $n_{\mathbf{r}_{\rm v}}$ rises rapidly to near unity by $\delta = 0.06$ [Fig. 2(c)]. This enhancement of $n_{\mathbf{r}_{\rm v}}$ characterizes the emergence of 'Mottness' at the vortex core region for an average doping not so close to unity. Thus, for $\delta \lesssim 0.06$ the vortex core becomes insulating and $g^t_{ij} \approx 0$ quenching the kinetic energy at the core. The effective picture of the underlying normal state in the core becomes that of an undoped patch of (antiferromagnetic) Mott insulator, described by a local Heisenberg model. This is quite unlike the Abrikosov vortex with a metallic core [5]. We note that the vortex core here is not simply serving as a window to the underlying normal state in the sense that the underlying normal state at $\delta = 0.06$ without the vortex is not yet a Mott-insulator. Instead, the Mott vortex core here is a result of strong correlations and a by-product of charge accumulation due to it. However we should also emphasize that this limit of vortex core is realized only in the proximity of the undoped Mott insulator. The reorganization of the local charge density at the vortex core as a function of doping is shown in Fig. 2(d). We find the excess local charge density at the vortex core changes sign with δ near optimal doping.

The non-linear effects of GRFs in the effective chemical potential μ_i, obtained while minimizing the total ground state energy of the system, play a key role in driving vortex cores

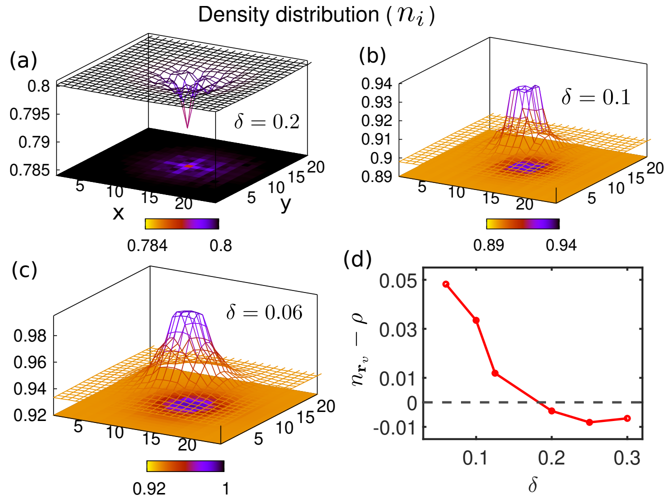

FIG. 2: **Electronic charge density distribution.** Local density n_i maps around a vortex core for different δ. At the vortex core, at $\delta = 0.2$ [panel (a)], n_i features a dip, and at $\delta = 0.1, 0.06$ (panel (b) and (c)]), the electronic charges accumulate to form a hill (with core density approaching unity). Panel (d) shows the profile of $n_{\mathbf{r}_v} - \rho$ Vs δ. The $n_{\mathbf{r}_v}$ values are less than ρ for $\delta > 0.18$ and greater than ρ for $\delta < 0.18$. At $\delta = 0.06$, $n_{\mathbf{r}_v}$ approaches unity leading to formation of a Mott insulating core.

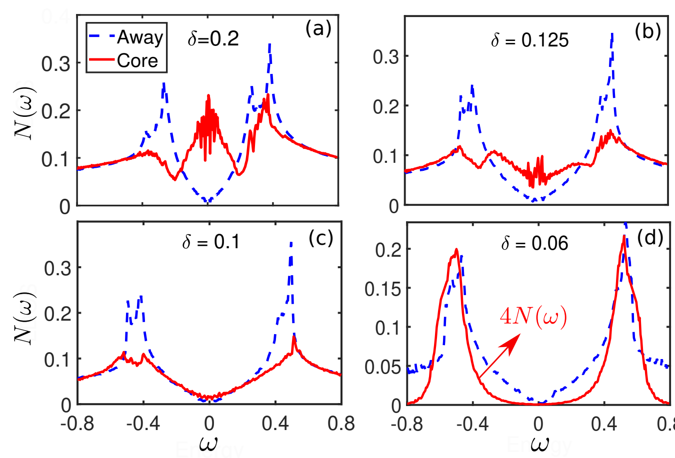

FIG. 3: **Local density of states.** LDOS at the vortex core (red traces) and away from the core (blue traces) for $\delta = 0.2$ (a), $\delta = 0.125$ (b), $\delta = 0.1$ (c), and $\delta = 0.06$ (d). For $\delta = 0.2$, the LDOS features a mid-gap peak which gradually reduces with decreasing δ. For $\delta = 0.06$, a hard gap opens with sharp peaks at $\omega \approx \pm J_{\text{eff}}/2$. In panel (d), the vortex core LDOS is scaled up by a factor of 4, for visual clarity.

towards Mottness, see SM for additional details [32]. Such effects not only drive the vortex core towards Mottness but also helps the nearby sites of the vortex core to attain local half-filling forming a near plateau in n_i [Fig. 2(c)]. The occurrence of a plateau in n_i in the core region is ultimately connected to the "flat bottom bowl" structure of $\Delta_{\text{d}}^{\text{OP}}$. The charge fluctuations freeze on these sites, as $t_{ij} \approx 0$, depleting dSC order over an extended region.

We emphasize that the removal of double occupancy is crucial for the aforementioned charge accumulation at the core and subsequent effects. Without the removal of double occupancy, we verified that the weak dip in n_i at the vortex core, a feature of overdoping continues until the lowest doping, see SM [32].

LDOS at the vortex core. The emergence of Mottness has important implications for the LDOS at the vortex core as we discuss below. In an s-wave superconductor, Andreev-like zero-energy bound states [44] were predicted theoretically to appear in the vortex cores and have also been observed experimentally in tunneling measurements [45]. For a dSC, similar accumulation of the low-energy core states (LECS) is also predicted within IMT calculation [11], even though true bound states are not found due to the collapse of the d-wave gap along the nodal directions. Such LECS are reminiscent of the metallic nature of the vortex core. However, the differential tunneling conductance map in cuprates shows no signatures of LECS in underdoped to optimally doped samples, beyond some sub-gap features [9, 10]. In contrast, recent experiments in overdoped samples showed prominent LECS at the vortex core [7].

To uncover this mystery, we show in Fig. 3 the LDOS with varying doping δ in GIMT. Within GIMT, the LDOS is calculated using [41, 46]: $N(\mathbf{r}_i, \omega) = N_e^{-1} \sum_{k,n} g_{ii}^t [|u_n^k(\mathbf{r}_i)|^2 \delta(\omega - E_{k,n}) + |v_n^k(\mathbf{r}_i)|^2 \delta(\omega + E_{k,n})]$, where $\{u_n^k(\mathbf{r}_i), v_n^k(\mathbf{r}_i)\}$ are the local Bogoliubov wavefunctions, $E_{k,n}$ are corresponding energy eigenvalues (see SM [32]), and N_e is the total number of eigenstates. As shown Fig. 3(a) the LDOS near the vortex cores is found to feature a peak near zero-energy for optimal doping $\delta = 0.2$. We find a similar peak at $\omega \approx 0$ in LDOS near vortex core for doping $\delta > 0.2$. Thus, LECS are present in the overdoped to optimally doped region, which also agrees with the weak-coupling predictions [11]. However, the vortex core LDOS at $\delta = 0.125$ in Fig. 3(b) shows a depletion in zero energy states and subgap features. With decreasing doping the low energy states get further suppressed and no LECS can be seen in Fig. 3(c). Upon further lowering doping to $\delta = 0.06$, the vortex core LDOS exhibits a U-shaped (hard) gap, as depicted in Fig. 3(d). This gap can be explained by the change in the nature of the vortex core with core density approaching unity for $\delta = 0.06$ as seen in Fig. 2(c). The Mott cluster of sites at the vortex core, being described by an effective Heisenberg model as discussed already, features lowest lying excited states beyond a spin gap $\approx J_{\text{eff}}$ [43, 47]. The tantalizing similarity of our finding of LDOS with experiments is truly intriguing. In IMT calculations, prominent LECS are always present at the vortex core for all δ, see SM [32].

Non-monotonicity in the core size. The unfolding of Mottness causes an intriguing non-monotonic variation of the core size with δ, as we examine below.

For definiteness, we define the vortex length scale ξ_{c} as the distance from the vortex center where the order parameter $\Delta_{\rm d}^{\rm OP}(i)$ recovers 80% of its maximum value. The red trace in Fig. 4(a), representing $\xi_{\rm c}(\delta)$, captures the two trends above and below the optimal doping $\delta \approx 0.2$. For $\delta > 0.2$, $\xi_{\rm c}$ shrinks as the doping value is decreased. This is consistent with the BCS expectation, where $\xi_{\rm c} \sim v_{\rm f}/\pi E_{\rm gap}$, with $v_{\rm f}$ and $E_{\rm gap}$ being the Fermi velocity and the energy-gap, respectively. Since, $E_{\rm gap}$ increases with decreasing δ within a d-wave BCS description, the vortex core shrinks. In the region below $\delta \approx 0.2$, $\xi_{\rm c}$ ceases to follow the $v_{\rm f}/\pi E_{\rm gap}$ trend and starts to increase continuously as doping is lowered towards $\delta \to 0$. As discussed earlier, in the strong underdoped limit the congregation of Mott sites makes the variation of $\Delta_{\rm d}^{\rm OP}$ near the vortex core flatter. Our findings indicate that the enhancement of $\xi_{\rm c}$ in underdoped regime is intimately connected with formation of Mott-cluster. It is indeed fascinating that the non-monotonicity in the vortex state tracks the non-BCS behavior [27]. A similar non-monotonic doping dependence has been theoretically discussed also for the SC coherence length in strongly correlated superconductors [39].

To further highlight the prominent dependence of the vortex core size on strong correlations, we also include the trace of $\xi_{\rm c}$ from IMT calculations in Fig. 4(a), which shows only a monotonic increase with δ in the entire range.

Superfluid stifness and critical magnetic field. Having encountered the non-monotonic dependence of $\xi_{\rm c}$ with δ, we next turn our attention to superfluid stiffness D_s which gives rise to Meissner effect [5]. Here we focus on the δ-dependence of $H_{\rm c2}$ within GIMT framework. In what follows, we calculate D_s using the Kubo formalism [48]: $D_s/\pi = \langle -k_x \rangle - \Lambda_{xx}(q_x = 0, q_y \to 0, \omega_n = 0)$, where $\langle -k_x \rangle$ is the average kinetic energy along x-direction and $\Lambda_{xx}(\mathbf{q}, \omega)$ is the transverse current-current correlation function. In order to obtain the $H_{\rm c2}(\delta)$, in Fig. 4(b) we plot D_s as a function of H, at different values of δ. Because the BdG technique does not include quantum phase fluctuations of SC order, D_s is not driven to zero by the fluctuations in the dSC pairing amplitude alone (which are fully included in BdG method). However, because BdG calculation results in a significant reduction of D_s to a low value, it is expected that quantum phase fluctuations, riding on top of the fluctuations in the pairing amplitude, would guide D_s to zero. We thus consider a small threshold value of $D_s/\pi = 0.1$ to mark off $H_{\rm c2}$. Even though such extraction of H_{c2} will not be an accurate estimate of the upper critical fields, we believe it to represent the qualitative doping dependence of the *true* $H_{\rm c2}$.

The behavior of the extracted critical field $H_{\rm c2}$ in the inset of Fig. 4(b), features a dome-shaped profile with its maximum residing at $\delta \approx 0.2$ (optimal doping). Similar non-monotonic behavior in $H_{\rm c2}$ versus δ has been recently observed in cuprate superconductors [49]. Interestingly, this finding gels well with the size of vortex core, because in Ginzburg – Landau theory theories, $H_{\rm c2} = \phi_0/2\pi\xi^2$, where the coherence length ξ is the characteristic length scale of the vortex core. Thus a non-monotonicity in the core size, as seen in Fig. 4(a), implies a non-monotonicity in $H_{\rm c2}$ as well. Interestingly, in cuprates

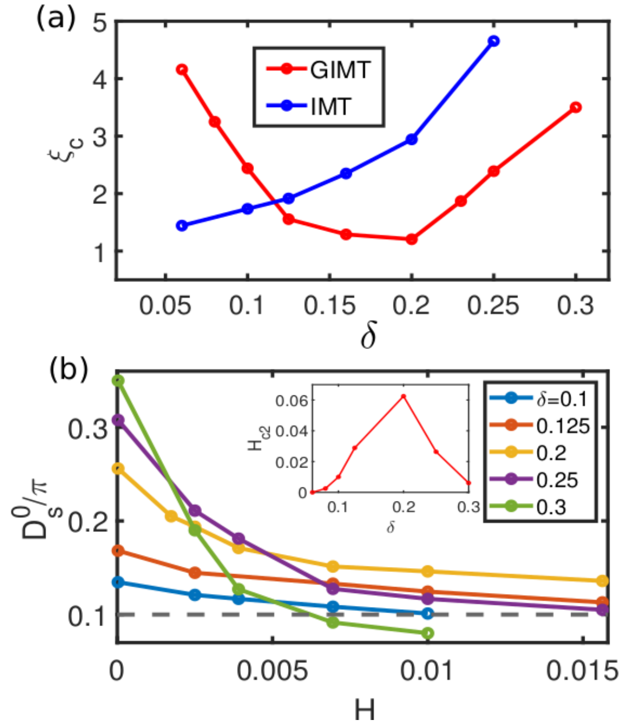

FIG. 4: **Vortex core size and critical magnetic field from superfluid density.** Panel (a) depicts the variation of the vortex core length scale $\xi_{\rm c}$ as a function of doping, from IMT (blue trace) and GIMT (red trace) calculations. In IMT, $\xi_{\rm c}$ shrinks monotonically with decreasing doping. In GIMT, $\xi_{\rm c}$ shows a non-monotonic behavior. Values of $\xi_{\rm c}$ are in the unit of the lattice spacing. Panel (b) shows the variations of superfluid density D_s^0 as a function of magnetic field H at different doping values. The threshold value for estimating the critical magnetic field $H_{\rm c2}$ is set at $D_s^0/\pi = 0.1$, as marked by the black horizontal line. The inset in panel (b) shows the behavior of the obtained $H_{\rm c2}$ with respect to δ, featuring a dome like profile. The H values are represented in the unit of ϕ_0.

the maximum of $H_{\rm c2}$ occurs near the optimal doping [50, 51], similar to our findings.

Conclusion. We illustrated how the nature of the vortex core changes from metallic-type in overdosed regime to a Mott-insulating one upon approaching undoping of a strongly correlated dSC. This changeover is accompanied by accumulation of the electronic charge at vortex core towards half-filling, which in turn facilitate the formation of Mott insulating core. It will be interesting to track the charge of vortices using cavity electromechanics measurements [52]. The change of the nature of vortex explains the anomaly in LDOS with dopings. The shape of the vortices do change as well, leading to a non-monotonic evolution of the vortex core size, which in turn explains the experimental signatures of $H_{\rm c2}$. These features stem from the non-BCS features due to the proximity to a Mott insulator. A high value of H_{c2} near optimal doping is also sometimes associated to the presence of a quantum critical point in the literature [53]. Our results do not depend on the presence of any quantum critical point near optimal doping. However, it will be an interesting future direction to connect our findings to a possible quantum critical point. Possible presence of competing orders can fine-tune the scenario by bringing in additional length scales. It should also be noted that our real space calculations naturally produce competing superconducting orders like extended s-wave order. However, the amplitudes of the extended s-wave order is extremely small and thus unlikely to have a significant effect on the LDOS. Our findings can have important implications on properties of other materials like Fe-based superconductors and twisted bilayer graphene, where strong correlation physics is believed to play a crucial role [54–57].

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[59] While a triangular vortex lattice is energetically favorable within a continuum Ginzburg-Landau theory, which ignores underlying lattice symmetries. However, it is numerically challenging to study the triangular vortex lattice with a underlying square lattice of finite size. Also, the connection between the structure of the vortex lattice and the crystal lattice symmetry is observed experimentally in conventional s-wave superconductors [58]

[60] While $\mathbf{r}_v$ represents the center of a vortex, for a better resolution of different local observables, e.g. LDOS at vortex core, we gather statistics not just at the vortex center but on a 2×2 lattice sites around the vortex center. Thus $\mathbf{r}_v$ represents the location of the 'vortex core region'.