A kaleidoscope of phases in the dipolar Hubbard model

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We investigate the emergence of a myriad of phases in the strong coupling regime of the dipolar Hubbard model in two dimensions. By using a combination of numerically unbiased methods in finite systems with analytical perturbative arguments, we show the versatility that trapped dipolar atoms possess in displaying a wide variety of many-body phases, which can be tuned simply by changing the collective orientation of the atomic dipoles. We further investigate the stability of these phases to thermal fluctuations in the strong coupling regime, highlighting that they can be accessed with current techniques employed in cold atoms experiments on optical lattices. Interestingly, both quantum and thermal phase transitions are signalled by peaks or discontinuities in local moment-local moment correlations, which have been recently measured in some of these experiments, so that they can be used as probes for the onset of different phases.

Introduction. — Experiments with ultracold atoms on optical lattices [1–4] have stimulated the search for new paradigms in many-body physics, especially due to the possibility of controlling and engineering quantum macroscopic states [5]. A recent experimental advance is the manipulation of atoms or molecules with (electric or magnetic) dipoles [6–8]. For example, 52Cr atoms with a large magnetic moment (6µB, with µB being the Bohr magneton) form Bose-Einstein condensates (BEC’s) below \( T_c \approx 700 \) nK [9]; larger magnetic moments, \( \sim 12\mu_B \), were later obtained with Er2 molecules [10]. The first quantum degenerate dipolar Fermi gas was realized [11] with 161Dy atoms cooled down to 20% of the Fermi temperature, \( T_F \approx 300 \) nK; also, Fermi surface deformation was observed in Er atoms [12]. An ultracold dense gas of fermionic potassium-rubidium (40K−87Rb) polar molecules was also generated [13], which paved the way to trap them into 2D and 3D optical lattices [14]; more recently, a two component Er dipolar fermionic gas with tunable interactions was prepared with collisional stability in the strongly interacting regime [15].

The interest in dipolar atoms stems from the fact that their interactions are long ranged and anisotropic, such that they can be directionally repulsive or attractive. This adds extra richness to the diversity of collective states of atoms in an optical lattice [16, 17]. For instance, quantum magnetism of high-spin systems has been experimentally studied with bosonic atoms in optical lattices [18, 19], and the ability to design quantum spin Hamiltonians with cold atoms may lead to the development of error-resilient qubit encoding and to topologically protected quantum memories [20]. In addition, since one of the motivations to study cold atoms in optical lattices is the possibility of emulating condensed matter models [1–4], a detailed investigation of effects due to dipolar interactions is clearly of interest. However, since experimental studies of dipolar fermionic atoms in optical lattices are still in their infancy [13, 14], theory must take the lead in highlighting interesting effects which would make the experimental effort worthwhile. Indeed, several studies suggest that new phenomena may emerge, such as \( p \)-wave pairing [21, 22] and different density-wave patterns [23–32], some of which are analyzed through analogies with liquid crystals. However, none of these studies predicted the formation of a Mott state, which has recently been achieved in two-component dipolar fermionic systems [15]; thus, a systematic study of the interplay between Mott and competing density-wave patterns is in order.

Model and methods. — With this in mind, here we consider spinful atoms (i.e., a mixture of atoms in two hyperfine states) on a half-filled optical lattice. The system is described by the Hamiltonian,

\[
\hat{H} = -t \sum_{\langle i,j \rangle,\sigma} \left( c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.} \right) + U \sum_i n_i \sigma n_i \sigma \\
+ \sum_{i \neq j} V_{ij} n_i n_j, 
\]

(1)

where, \( c_{i\sigma} \) (\( c_{i\sigma}^\dagger \)) denotes the particle annihilation (creation) operator and \( n_i \) the number operator at site \( i \). The sums run over sites of a square optical lattice, with \( \langle i,j \rangle \) denoting nearest neighbor sites; \( \sigma = \uparrow, \downarrow \) denotes the two hyperfine states, and \( t \) is the hopping integral. An external field aligns the dipoles parallel to the unit vector \( \mathbf{d} \), specified by the usual polar angles \( \theta \) and \( \varphi \), taking \( \mathbf{z} \) perpendicular to the square lattice; see Fig. 1(d). The dipolar interaction is then written as

\[
V_{ij} = \frac{V}{r_{ij}^3} \left[ 1 - 3 \left( \hat{r} \cdot \hat{d} \right)^2 \right], 
\]

(2)

where \( V \) (proportional to the square of the dipole moments) is the strength of the interaction, \( r_{ij} \equiv i - j \) is a vector joining sites on the lattice, and \( \hat{r} \) is its unit vector. The interaction of two atoms in the same optical well, \( U \), is the sum of two contributions [6, 33]: one is the...
usual contact interaction, tunable through a Feshbach resonance; the other comes from the dipolar interaction, whose behavior at small distances is limited by the finite size of the atoms \([6, 33]\).

The ground state properties of the aforementioned Hamiltonian is analysed with the Lanczos method on a \(4 \times 4\) lattice with periodic boundary conditions, in the subspace of half filling; translational symmetry and total spin projection are also incorporated in the bases used \([34, 35]\). In line with experiments in the absence of dipolar interactions, here we consider the case \(U = 8t\), which is also convenient since finite-size effects are small in the strong-coupling regime – more on this below. The finite lattice size we use forces us to truncate the dipolar interaction beyond second neighbors. Nonetheless, anisotropy and competition between attractive and repulsive couplings are preserved. We also perform strong-coupling analyses, complemented by simulated annealing, in order to check the consistency of exact diagonalization results (full lines guide the eye) for \(t = 1\), while the dashed line corresponds to the atomic limit \((t = 0)\).

Zero-temperature transitions.— Let us first fix the direction of polarization and vary the strength of the dipolar interaction, \(V\). Figures 1(a) show the correlation functions in the isotropic case, \(\theta = \varphi = 0\): spin correlations consistent with a Néel-like arrangement (a1) are completely suppressed at \(V_{\text{CB}} \approx 3.1\), beyond which charge correlations (a2) develop. The system therefore goes from a Mott phase, in which each species occupies one sublattice, to a checkerboard charge density wave (CB) phase, in which only one of the sublattices is occupied by both species; see cartoons in Fig. 1(d). Panel (a3) shows the moment-moment correlation function, which captures the increase of fluctuations at the critical point; the sharp drop in the local moment is responsible for the sharpness of \(C_m(r)\) at the transition. By contrast, when the dipoles point along the \(\hat{x}\) direction [Figs. 1(b)] the transition is from a Mott phase to a striped phase, at a smaller \(V\) than for the isotropic case; the direction of the stripes is that of the dipoles, since arranging them head-to-tail lowers the energy and skipping a row costs less energy than placing them on adjacent rows. As a result, nearest-neighbor spin correlations are now anisotropic in the Mott phase: in strong coupling, \(J_{\text{eff}} = 4t^2/(U - V_\nu), \nu = x, y\), with \(V_x < 0\) and \(V_y > 0\), so that attraction weakens magnetic correlations. By the same token, local moment fluctuations are also anisotropic, since vertically one has doublon-holon pairs while horizontally one has doublon-doublon pairs, the latter being less prone to fluctuations than the former.

Since the nature of the CDW state depends on the polarization angle, we now probe the phase transitions driven by changing the direction of the dipoles within the \(xz\)-plane (\(\varphi = 0\)), while \(V\) is kept fixed. Figures 1(c)
show that with increasing $\theta$ the cbCDW phase gives way to a Mott phase (with anisotropic correlations), and further increase in $\theta$ drives the system to another CDW phase, now with stripes along the $xz$-direction (XS); see the dash-dotted line in Fig. 1(d). This intervening Mott phase disappears at some critical $V_c$, which is not very sensitive to the presence of the hopping for a fixed $U/V$ in the physically relevant domain of $U \gg t$, as revealed by comparing with the size-independent strong coupling results; see Fig. 1(d). Note that for $V < V_{CB} \approx 3.1t$ no cbCDW state is formed, and the smaller $V$ gets, the direction of polarization must get closer to the plane in order to reach the XS phase; interestingly, below $V_{H} \approx 1.5t$ no CDW is formed.

In order to relax the constraint of polarization within the $xz$-plane, we have taken advantage of the fact that the atomic limit (i.e., $t \to 0$) captures, to a very good approximation, the essence of the phase diagrams, as discussed in connection with Fig. 1(d). Accordingly, we have mapped out the lowest energy states in the thermodynamic limit at fixed $U$ and $V$, for many values of $\theta$ and $\varphi$; consistency with data from both Lanczos diagonalizations and simulated annealing was checked in many cases. Our findings can be summarized in the $\theta \times \varphi$ phase diagram of Fig. 2(a), for $U/V = 2.22$, which displays the symmetry under a reflection of the polarization with respect to the plane of the lattice. For polarization nearly perpendicular to the plane ($0 \leq \varphi \lesssim 0.1\pi$ and $0.9\pi \leq \theta \leq \pi$), we see that the cbCDW pattern is robust against any rotation of $\mathbf{d}$ around the $z$-axis. Figures 2(b1) and 2(b2) show results from perturbation theory [40] indicating that the effect of a finite hopping is to introduce oscillations of negligible amplitudes on the border between cbCDW and Mott phases.

Beyond $\theta \approx 0.1\pi$, the patterns formed depend on $\theta$ and $\varphi$. First, striped phases emerge along either the $x$ direction (XS) for $0 \leq \varphi \lesssim 0.2\pi$ (and $0.8\pi \lesssim \varphi \leq \pi$), or the $y$ direction (YS) for $0.3\pi \lesssim \varphi \lesssim 0.7\pi$. Figure 2(c) shows correlation functions for $\theta = \pi/4$ obtained by means of Lanczos diagonalization; the ground state goes from the XS to the Mott phase around $\varphi/\pi = 0.12$ and then to YS phase at $\varphi/\pi = 0.38$. Figure 2(c3) shows that the spatial anisotropy between doublon-holon and doublon-doublon correlations is picked up by the moment-moment correlation functions as the polarization rotates around $\mathbf{d}$, thus confirming its important role in probing quantum phase transitions.

Second, for nearly in-plane polarization ($0.4\pi \lesssim \varphi \lesssim 0.6\pi$), phase separation (PS) sets in between the XS and YS phases [40]. We note that as $U/V$ increases, first the
cbCDW phase disappears (for $U/V > 2.586$); then, for $U/V > 2.71$ the PS states are suppressed (see Fig.S2 in Supplemental Material). And, finally, for $U/V > 5.33$ the striped phases disappear; in this latter regime, the system is in a Mott state for all polarization directions.

Thermal transitions.—Having characterized the ground-state phases and its transitions in terms of the dipole orientations and magnitude of the interactions, an important question, even more prominently from an experimental standpoint, refers to the robustness of these phases in the presence of thermal fluctuations. Figure 3 shows the temperature dependence of the charge-charge and moment-moment correlation functions for different directions of polarization, obtained through parallel tempering simulations in the atomic limit [40].

As expected, the charge correlations start at their ground-state values consistent with XS and YS phases (see Fig. 3(a1) and (b1), respectively), and decrease in magnitude as $T/L$ increases. An estimate of the temperature scale marking the suppression of these ordered phases can be obtained from the peak position of the moment-moment correlations, shown in Figs. 3(a2) and (b2): they are the same for both XS and YS phases, namely $T_{X}/U = T_{Y}/U \approx 0.61$, for $U/V = 2.22$. For these values of $U$ and $V$, we estimate from Figs. 3(c1) and (c2) the ordering temperature for the cbCDW phase as $T/U \sim 0.1$, which lies in a range in which parallel tempering simulations are hindered by trapped metastable configurations. Nonetheless, we are able to infer an upper bound $T_{cbCDW} < T_{X}$, which is valid for different values of the ratio $U/V (< 5.33)$. One can understand this result by noticing that charge gaps are larger for the striped phases than for the cbCDW phase, thus leading to higher critical temperatures. Finally, for polarizations leading to the Mott phase, such as $\theta = \varphi = \pi/4$ shown in Fig. 3(d1) and (d2), the atomic limit also displays a critical temperature associated with the onset of a homogeneous charge ordering, though without any manifest spin order, which is absent in this regime due to the vanishing exchange couplings when $t \to 0$.

The estimates thus obtained for the ordering of the XS, YS and cbCDW phases are gathered in Fig. 4; we recall that for $U/V > 5.33$ the ground state is a Mott ‘insulator’ for all polarization directions. Recent experiments have reached temperatures as low as $T/U \sim 0.4$ [15], but with a ratio $U/V$ too large ($\sim 50$) to probe the charge ordered phases (see Fig. 4). According to our estimates, at this $T/U$ the striped phases are accessible for $U/V \lesssim 3$, and the cbCDW phase for $U/V \lesssim 1.5$.

Summary.—We have established that dipolar fermionic atoms in an optical lattice provide a setup in which Mott and density-wave states can in principle be stabilized by a simple control of the direction of polarization. These density-wave states may be anisotropic (stripe-like) or occupy one of the sublattices; in addition, one may also find anisotropic phase-separated phases. Depending on the strength of the dipolar interaction, a rotation of the polarization around the $\hat{z}$ axis can switch between the density-wave states through a succession of phase separated states. Our results are based on exact diagonalizations of a dipolar Fermi-Hubbard Hamiltonian on a $4 \times 4$ lattice at half filling, in the regime of strong on-site repulsion. In this regime, finite-size effects are not too drastic, as evidenced by the comparison with predictions obtained.
in the atomic limit (hopping $t \to 0$), aided by simulated annealing. By now the use of moment-moment correlations has proven to be a powerful tool to probe different phases in experiments with ultracold atoms [36–39], so that our theoretical predictions for this quantity should provide guidance in the experimental search for these phases. Indeed, despite the low temperatures achieved in recent experiments, the large $U/V \sim 50$ [15] regime prevented this kaleidoscope of phases from being accessible. If experiments were able to reduce $U/V \lesssim 3$, our parallel tempering simulations predict that for $T/U \sim 0.4$ the striped phases will be within reach.

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Supplementary Material:
A kaleidoscope of phases in the dipolar Hubbard model

ATOMIC LIMIT \((t = 0)\) PHASE DIAGRAM

Here we discuss the phase diagram of the dipolar Hubbard model (dHM) in the atomic limit \((t = 0)\),

\[
H_{\text{at}} = U \sum_i n_i \hat{n}_i + \sum_{i \neq j} V_{ij} \hat{n}_i \hat{n}_j. \quad (S1)
\]

As mentioned in the main text, up to next-nearest neighbors \(V_{ij}\) becomes

\[
\begin{align*}
V_x &\equiv V \left(1 - 3 \sin^2 \theta \cos^2 \phi \right) \quad (S2) \\
V_y &\equiv V \left(1 - 3 \sin^2 \theta \sin^2 \phi \right) \quad (S3) \\
V_{d1} &\equiv \frac{V}{2^{3/2}} \left[1 - \frac{3}{2} \sin^2 \theta (1 + \sin 2\phi) \right] \quad (S4) \\
V_{d2} &\equiv \frac{V}{2^{3/2}} \left[1 - \frac{3}{2} \sin^2 \theta (\cos 2\phi + \sin 2\phi) \right] \quad (S5)
\end{align*}
\]

The eigenstates of Eq.\((S1)\) are product states (classical states) and the ground state (GS) is the one which minimizes the energy for the given values of \(U, V, \theta\) and \(\phi\). For instance, when \(U \gg V\), double occupancies are suppressed due to the high energy penalty \(U\), and the GS corresponds to a Mott insulator. Physical intuition can be used to set up other possible GS classical states; see, e.g., Fig. 1 of the main text and Fig. S1.

For an \(L \times L\) lattice with periodic boundary condition, the energy per particle at half filling for the competing ground states may be written as

\[
E_0^{\text{cdCDW}} = \frac{U}{2} + 2(V_{d1} + V_{d2}) \quad (S6a)
\]
\[
E_0^{\text{Mott}} = V_x + V_y + V_{d1} + V_{d2} \quad (S6b)
\]
\[
E_0^{\text{XPS}} = \frac{U}{2} + 2V_x \quad (S6c)
\]
\[
E_0^{\text{YPS}} = \frac{U}{2} + 2V_y \quad (S6d)
\]
\[
E_0^{\text{d1PS}} = \frac{U}{2} + 2(V_x + V_y + V_{d1} + V_{d2}) \quad (S6e)
\]
\[
E_0^{\text{d2PS}} = \frac{U}{2} + 2(V_x + V_y + 2V_{d2}) \quad (S6f)
\]
\[
E_0^{\text{d2PS}} = \frac{U}{2} + 2(V_x + V_y + 2V_{d1} + V_{d2}) \quad (S6g)
\]
\[
E_0^{\text{d2PS}} = \frac{U}{2} + 2(V_x + V_y + 2V_{d1}) \quad (S6h)
\]

where XPS and YPS denote \(\hat{x}\)- and \(\hat{y}\)-oriented phase-separated states, while d1PS and d2PS denote \(\pm \pi/4\)-oriented phase-separated states; see Fig. S1.

By comparing the energy of these different classical states one is able to draw the atomic-limit \(\theta \times \phi\) phase diagrams presented in Fig. 2(a) of the main text. From the outset we note that the last term in the energy of all phase-separated (PS) states vanish as \(L \to \infty\), so that all PS states become degenerate in the thermodynamic limit. For \(U/V = 2.22\), we identify the following transitions, depending on the values of \(\theta\) and \(\phi\): (I) cbCDW-Mott, (II) Xs(or Ys)-Mott, (III) Xs(or Ys)-PS and (IV) PS-Mott; see Figs. 1 and 1. Indeed, starting from the isotropic case, \(\theta = 0\), when the GS is a cbCDW, there is a transition to a Mott state at a critical \(\theta_{c1}\), given by

\[
\theta_{c1} = \arcsin \left( \pm \sqrt{ \frac{2}{3} + \left( \frac{\sqrt{2}}{2 - 4\sqrt{2}} \right) \frac{2U}{3V} } \right), \quad (S7)
\]

where the \(\pm\) apply to \(\theta_{c1} > \pi/2\) or \(\theta_{c1} < \pi/2\), respectively, with the proviso that the cbCDW phase disappears for \(U/V > (4\sqrt{2} - 2)/\sqrt{2} \approx 2.586\), which would lead to a complex \(\theta_{c1}\). This point is marked in Fig. 4 of the main text, at the corresponding onset of the
Mott phase at zero temperature. The fact that \( \theta_{c1} \) is independent of \( \varphi \) gives rise to the straight horizontal line phase boundaries in Fig. 2(a) of the main text.

Increasing \( \theta \) above \( \theta_{c1} \) leads to attractive dipolar interactions along the \( \hat{x} \) (or \( \hat{y} \)) direction while still being repulsive along \( \hat{y} \) (or \( \hat{x} \)). This energetically favors stripes along \( \hat{x} \) (or \( \hat{y} \)), which we denote by XS (or YS); their regions of stability in the \( \theta \times \varphi \) plane are shown in Fig. 2(a) of the main text. In-between the XS and YS phases there is a Mott region, whose boundaries depend on both \( \theta \) and \( \varphi \), for fixed \( U/V \).

When \( \theta \approx \pi/2 \), the average dipolar interaction is attractive and the phase-separated states compete with both XS (YS) and the Mott state. For \( \theta = \pi/2 \), an XS-YS transition takes place at \( \varphi_{cs} \approx 0.15\pi \) for \( 0 \leq \varphi \leq \pi/4 \), or at \( \varphi_{cy} \approx 0.35\pi \) (YS-PS transition in this case) for \( \pi/4 \leq \varphi \leq \pi/2 \), see Fig. 2(a). The PS state has the global minimum energy within the range \( \varphi_{cs} < \varphi < \varphi_{cy} \), due to the fact that the components of the dipolar interaction are attractive, \( V_x, V_y, V_d < 0 \), thus favoring the condensation of the particles. In this case, the PS competes with the Mott phase as the dipole direction deviates from \( \theta = \pi/2 \); see Fig. 2(a). The Mott-PS transition occurs in a line of the phase diagram whose critical value of \( \theta \) is given by

\[
\theta_{c2} = \arcsin \left[ \pm \sqrt{\frac{2 + U \sqrt{2}}{3V + 2 \sqrt{2}}} \right],
\]

where \( \pm \) respectively correspond to the critical \( \theta \) for \( \theta < \pi/2 \) and \( \theta > \pi/2 \); again, note that \( \theta_{c2} \) is independent of \( \varphi \). For \( U/V > \frac{1 + 2\sqrt{2}}{\sqrt{2}} \approx 2.71 \), Eq. (S8) yields \( \sin \theta_{c2} > 1 \), so that the PS phase is suppressed, with the Mott state dominating the whole region \( \varphi_{cs} < \varphi < \varphi_{cy} \) of the phase diagram.

Summing up, as \( U/V \) increases, first the cbCDW phase disappears (for \( U/V > 2.586 \)), then for \( U/V > 2.71 \) the PS states are suppressed; see Fig. S2. And, finally, for \( U/V > 5.33 \) the striped phases disappear; in this latter regime, the system is in a Mott state for all polarization directions.

Since experiments with ultra-cold atoms can’t always be considered as ‘in the thermodynamical limit’, one must comment on how these results are affected by a finite \( L \). We recall [see Eqs. (S6)] that while the energies per particle for the Mott, XS (or YS), and cbCDW states are independent of system size, \( L \), the PS states have contributions proportional to \( 1/L \), due to ‘interface’ contributions [see Fig. S1]. Therefore, in a finite system PS states with different orientations may be formed due to the anisotropic nature of the dipolar interaction. For instance, the strip in which the XPS (or YPS) phase is stable when \( L \rightarrow \infty \) shrinks to a small lobe emerging from the striped phases when, say \( L = 4 \). By contrast, the boundaries involving non-PS phases are hardly affected by a finite \( L \).

**SECOND ORDER PERTURBATION THEORY**

Let us now discuss how a small hopping \( (t \ll V,U) \) affects the atomic-limit phase diagrams, resorting to perturbation theory (PT). The correction to the atomic-limit energies up to second order perturbation theory, \( E^{(2)} \), is described by the effective Hamiltonian \([41]\)

\[
\langle \phi_0^i | H_{\text{eff}} | \phi_0^i \rangle = \langle \phi_0^i | H | \phi_0^i \rangle + \sum_{m>0} \frac{\langle \phi_0^i | K | \phi_m \rangle \langle \phi_m | K | \phi_0^i \rangle}{E_0 - E_m},
\]

where \( E_m \) and \( | \phi_m \rangle \) are the respective eigenvalues and eigenstates of \( H_{\text{at}} \), \( H_{\text{at}} | \phi_m \rangle = E_m | \phi_m \rangle \). \( H_{\text{eff}} \) is therefore an operator which acts in the subspace of the degenerate ground states of \( H_{\text{at}} \), \{ | \phi_0^i \rangle \}, and the perturbation \( K \) is the hopping term of the dHM.

The correction \( E^{(2)} \) is the lowest energy of \( H_{\text{eff}} \). The cbCDW and the XS(YS) atomic-limit ground states form a subspace that is two-fold degenerate in each case, so \( H_{\text{eff}} \) is a \( 2 \times 2 \) diagonal matrix. In these cases, we obtain

\[
E^{(2)}_{\text{cbCDW}} = \frac{2t^2}{U - 4V_x - 3V_y + 4V_d l + 4V_d^l},
\]

and

\[
E^{(2)}_{\text{XS(YS)}} = \frac{2t^2}{U - 4V_x(y) - 3V_y(x) - 4V_d l - 4V_d^l}.
\]

On the other hand, the atomic-limit Mott states form a macroscopically degenerate subspace, and \( H_{\text{eff}} \) becomes an anisotropic SU(2) Heisenberg Hamiltonian \([41, 42]\)

\[
H_{\text{eff}} = J_x \sum_i \vec{S}_i \cdot \vec{S}_{i+\hat{x}} + J_y \sum_i \vec{S}_i \cdot \vec{S}_{i+\hat{y}} - \frac{N}{4} (J_x + J_y),
\]

where the exchange couplings \( J_x = 4t^2/(U - V_x) \) and \( J_y = 4t^2/(U - V_y) \) depend on the dipolar angles \( \theta \) and \( \varphi \).
atomic limit

\[ C_{m}(r) = \begin{cases} \frac{1}{2}m_{2}^{2} - \frac{1}{2}m_{2}^{2} & \text{for } r = (0,1), \frac{1}{2}m_{2}^{2} - \frac{1}{2}m_{2}^{2} & \text{for } r = (1,0), \frac{1}{2}m_{2}^{2} - \frac{1}{2}m_{2}^{2} & \text{for } r = (1,1), \frac{1}{2}m_{2}^{2} - \frac{1}{2}m_{2}^{2} & \text{for } r = (2,0) \end{cases} \]

Figure S3. (Color online) \( \theta \times \varphi \) phase diagram obtained with second order perturbation theory for \( U = 100t \) (full red lines) and \( U = 8t \) (dashed red lines). The results for the atomic-limit are also presented.

\[ E_{\text{Mott}} = E_{\text{Mott}}^{(0)} + E_{\text{Mott}}^{(2)}, \quad E_{\text{cbCDW}} = E_{\text{cbCDW}}^{(0)} + E_{\text{cbCDW}}^{(2)} \]

and

\[ E_{\text{XS(YS)}} = E_{\text{XS(YS)}}^{(0)} + E_{\text{XS(YS)}}^{(2)}, \]

we have established that the main effect of the hopping is to enlarge the region dominated by the Mott phase in the \( \theta \times \varphi \) diagram. The critical angle \( \theta_{c1} \) associated to the Mott-cbCDW transition decreases in comparison with the atomic-limit case. Further, due to the presence of anisotropic AFM correlations, \( \theta_{c1} \) acquires a tiny dependence on \( \varphi \), as it can be seen from Fig. 2(b) of the main text. In addition, the lobes of the \( \theta \times \varphi \) phase diagram dominated by the XS and YS phases shrink as we decrease the value of \( U \) to \( U/t = 8 \); see Fig. S3.

**MOMENT-MOMENT CORRELATIONS**

As shown in the main text, the moment-moment correlations, \( C_{m}(r) = \langle m_{i}^{2}\rangle \langle m_{j}^{2}\rangle - \langle m_{i}\rangle \langle m_{j}\rangle \), can be used to identify not just the different transitions described by the dHM, but also the nature of the anisotropic CDW phases. In this section we take a closer look at the local moment, \( \langle m_{i}^{2}\rangle \), at the different Mott-CDW transitions discussed in the main text.

We first consider the transitions occurring as one varies \( \varphi \) for fixed \( \theta = \pi/4 \); see Fig. 2(c) of the main text. In this case, Fig. S4(a1) shows that \( \langle m_{i}^{2}\rangle \) is close to saturation in the Mott phase, but sharply decreases in the striped phases; a similar behavior occurs as \( \theta \) varies with fixed \( \varphi \), as in Fig. S4 (b1). By contrast, Figs. 1(c) and 2(c) show that \( C_{m}(r) \) is peaked at the different transitions for some specific directions \( r \). For the Mott-XS(YS) transition, for instance, the peak of \( C_{m}(r) \) occurs when the doublon-holon fluctuations are the strongest; see Fig. S4(b1), and the discussion in the main text. A peak in \( C_{m}(r) \) is also observed at the cbCDW-MottAFM transition, see Fig. S4(b2). Thus, the sharp drop in the local moment is responsible for the sharpness of \( C_{m}(r) \) at the transition.

**PARALLEL TEMPERING IN THE ATOMIC LIMIT**

To estimate the critical temperatures \( T_{\alpha} \) signaling the onset of the different ordered classical phases, \( \alpha \), listed in Eq. (S6), we use the parallel tempering [45] (or replica exchange method) of the Hamiltonian (S1). In summary, we use a Monte Carlo (MC) sampling of the occupations of both species \( \{\uparrow \text{ and } \downarrow\} \), promoting random swaps of site occupancies, complemented by random creation and destruction of particles at different temperatures. These moves are implemented as to obey the detailed balance condition, in a particle-hole symmetric version of Eq. (S1). This guarantees that on average one keeps \( \langle n_{\downarrow}\rangle = \langle n_{\uparrow}\rangle = 0.5 \). After a single MC sweep, an attempt of swap of the configurations related to adjacent temperatures in a given range is induced and accepted with probability

\[ p = \min\{1, \exp\{-\beta_{i}\langle E_{j} - E_{i}\rangle\} \}, \]

where \( \beta_{i} = 1/T_{i} \) is the inverse temperature of a given configuration \( i \) whose associated energy for the Hamiltonian (S1) is \( E_{i} \).
We typically use square lattices up to \( L = 32 \), and 20,000 MC sweeps, with approximately 300 different temperatures chosen in a way to ensure that the range encompasses the associated critical temperatures \( T^* \). It is a known difficulty of the parallel tempering scheme on how to choose the optimal set of temperatures [46, 47] which overcome the trapping of metastable configurations when \( T \to 0 \). Although sub-optimal, we used a simple approach of evenly spaced ones, which is more than sufficient to resolve the critical temperatures associated to the onset of the different phases.

As an example, we report in Fig. S5 the comparison of \( N_q \) for two striped phases, XS (\( \theta = \pi/2 \) and \( \phi = 0 \)) and YS (\( \theta = \phi = \pi/2 \)) in panels (a) and (b), respectively. We note that the structure factor has a symmetric role for different channels: while for XS the \( q = (0, \pi) \) channel displays an extensive behavior at low temperatures, \( q = (\pi, 0) \) reflects this corresponding behavior for the YS phase. For very low temperatures, however, the aforementioned trapping of metastable configurations occurs, preventing the observation of a fully formed plateau; this, in turn, signals that the correlation length for this ordering has reached the linear size of the system. Nonetheless, the critical temperatures \( T_{cXS}^{KS} \) and \( T_{cYS}^{KS} \) lie way above the temperatures where these problems begin to occur. As an estimation, we also display as a vertical line in these panels the thermal peak-positions of the moment-moment correlation functions (as in Fig. 3 of the main text), which are very close to the regime where the curves for different system sizes start displaying an extensive behavior. Conversely, for the channels \( q = (\pi, 0) \) [Fig. S5(a1)] and \( q = (0, \pi) \) [Fig. S5(b2)] for the XS and YS phases, respectively, \( N_q \) is approximately independent of the system size, thus confirming the nature of the charge periodicity.

Lastly, we perform similar parallel tempering simulations for the case of isotropic interactions, i.e., \( \theta = \phi = 0 \), where the ground state of Eq. S1 displays cbCDW order. Figure S6 shows the temperature dependence of the \( q = (\pi, \pi) \) channel for the charge structure factor, \( \hat{\mathbf{n}} \) oriented dipoles. The extensive behavior at low temperatures signals the onset of the cbCDW order. As before, the vertical line depicts the peak position of the temperature dependent moment-moment correlation functions. We choose the ratio of interactions \( U/V = 1 \).

\[
N_q = \frac{1}{L^2} \sum_{i,j} \langle e^{i \mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \mathbf{n}_i \mathbf{n}_j \rangle_{\text{MC}},
\]  

\( S17 \)

which becomes an extensive quantity in the presence of a given charge order with wave-vector \( \mathbf{q} \).

![Figure S5](image-url)

Figure S5. (Color online) Temperature dependence of the charge structure factor for the stripe phases XS in (a) and YS in (b). We select two channels with \( q = (\pi, 0) \) in (a1) and (b1), whereas (a2) and (b2) display the \( q = (0, \pi) \) results. Vertical lines depict the peak position of the temperature dependent moment-moment correlation functions, signaling the thermal transition. We fix the interaction ratio \( U/V \) to 2.22.

Similarly to the quantum version of the Hamiltonian, in the main text we present local correlations \( \langle C_{\mathbf{r}}(\mathbf{r}) \rangle \) which help to identify the charge distribution in all classical phases. Here, to complement this analysis and describe a fully developed order, we compute the associated charge structure factor,

\[
N_q = \frac{1}{L^2} \sum_{i,j} \langle e^{i \mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \mathbf{n}_i \mathbf{n}_j \rangle_{\text{MC}},
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