Classical and quantum liquids induced by quantum fluctuations

Miguel M. Oliveira
CeFEMA, Instituto Superior Técnico, Universidade de Lisboa Av. Rovisco Pais, 1049-001 Lisboa, Portugal

Pedro Ribeiro
CeFEMA, Instituto Superior Técnico, Universidade de Lisboa Av. Rovisco Pais, 1049-001 Lisboa, Portugal and Beijing Computational Science Research Center, Beijing 100193, China

Stefan Kirchner
Zhejiang Institute of Modern Physics, Zhejiang University, Hangzhou, Zhejiang 310027, China

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Geometrically frustrated interactions may render classical ground-states macroscopically degenerate. The connection between classical and quantum liquids and how the degeneracy is affected by quantum fluctuations is, however, less well understood. We study a simple model of coupled quantum and classical degrees of freedom, the so-called Falicov-Kimball model, on a triangular lattice and away from half-filling. For weak interactions the phase diagram features a charge disordered state down to zero temperature. We provide compelling evidence that this phase is a liquid and show that it is divided by a crossover line that terminates in a quantum critical point. Our results offer a new vantage point to address how quantum liquids can emerge from their classical counterparts.

Liquids are characterized by the absence of long-range order. In exceptional cases, a classical liquid state may persist down to zero temperature [1, 2]. The ensuing ground-state is macroscopically degenerate and thus is characterized by a finite entropy. Such a degeneracy in the energy landscape can result from competing interactions, geometric frustration, or near phase transitions where different states compete. The proliferation of low-energy states renders the system unstable and prone to the emergence of novel and often exotic ground-states, such as phases with fractional excitations, unconventional superconductivity, valence bond solids and so on.

An interesting and still open issue concerns the relation between classical and quantum liquids, i.e., how quantum fluctuations affect the classical ground state manifold [3, 4]. Another pertinent issue of practical relevance is the stability of such liquid phases with respect to, for example, the itinerary of the frustrated degrees of freedom. Any attempt of answering these questions is faced with the principal difficulty that quantum fluctuations of competing interactions, covering a wide energy range, need to be taken into account.

In this letter, we identify liquid phases which are driven by a coupling to quantum degrees of freedom. Depending on whether the quantum variables acquire a non-zero mass, the nature of the liquid phase changes. To this end, we study the Falicov-Kimball model (FKM), a hybrid model comprised of itinerant fermions that interact arbitrarily strongly with localized charges, on a triangular lattice. This model is well suited to address some of the unresolved issues mentioned above and, in addition, allows for an effective, numerically exact Monte-Carlo sampling of its partition function [5, 6].

Originally developed to model the Mott physics in f-electron based systems [7], the FKM can be thought of as a special case of the Hubbard model with infinite mass imbalance between the two spin species thus rendering the dynamics of the heavier one classical.

The model has been instrumental in benchmarking the standard approach to strongly correlated lattice models, i.e., the dynamic mean field theory, and its extensions [8–12]. More recently, it has attracted renewed interest in the context of disorder-free many-body localization [6, 13, 14].

The position of these classical charges is annealed over an energy landscape defined by the itinerant degrees of freedom. For bipartite lattices, several exact results have been established [15, 16], including the existence of a charge density wave (CDW) at low temperature (T) for all interaction strengths. The melting of the charge-density wave state with increasing temperature was observed for commensurate fillings [15–18]. On the triangular lattice the FKM and its extensions display a variety of different ground-state phases [19–23]. For incommensurate fillings the FKM favors phase separation [5]. Recently, it was demonstrated that the half-filled model on the square lattice is non-metallic at all non-vanishing values of the interaction strength $U$ and transitions from Anderson to Mott insulators as $U$ is varied [6].

In a manner reminiscent of the large-$U$ limit of the Hubbard-model, an effective model for the classical charges can be derived perturbatively at sufficiently small coupling $t/U$, where $t$ is the hopping strength of the itinerant electrons. At half-filling and for large $U/t$, the FKM is equivalent to the antiferromagnetic Ising model. Thus, on the square lattice, order ensues at sufficiently low $T$ whereas the large-$U$ limit remains disordered for all $T$ on the triangular lattice [1]. While it is an interesting question how this gets modified in the presence...


The Hamiltonian of the Falicov-Kimball model is

\[ H = -t \sum_{\langle ij \rangle} c_i^\dagger c_j + U \sum_i c_i^\dagger c_i n_{f,i} - \mu_c \sum_i c_i^\dagger c_i - \mu_f \sum_i n_{f,i}, \]

where \( c_i^\dagger \) creates a \( c \)-electron and \( n_{f,i} \), a conserved quantity, counts the number of immobile classical charges on site \( i \); and \( \mu \) is the chemical potential of the system. \( t \) will be used as a unit of energy (\( t = 1 \)). The summation \( \sum_{\langle ij \rangle} \) runs over all nearest-neighbor pairs on a triangular lattice with a volume \( V = L^2 \) (\( L \) being the system’s linear dimension) and periodic boundary conditions. As the collection of \( n_{f,i} \) constitutes a set of conserved quantities, the partition function is given by a summation over non-interacting contributions for every configuration \( n_f \) of \( f \)-charges and thus can be evaluated by an efficient Monte-Carlo sampling[5, 6]. An additional difficulty arises in the present situation, as the chemical potentials \( \mu_c(T, U, L) \) and \( \mu_f(T, U, L) \) need to be self-consistently determined to ensure constant occupation as a function of \( T, U \), and of quantum fluctuations [24], the parent state at large \( U \) is already a liquid. Here, however, we will instead demonstrate that a classical liquid state can result from an ordered phase, present \( e.g. \) for one third filling on the triangular lattice, due to coupling to a quantum field.

The following, we consider a macro-canonical ensemble with the chemical potentials \( \mu_c \) and \( \mu_f \) determined such that \( \mu_c = \langle n_i \rangle / V = 2/3 \) and \( \mu_f = \langle n_f \rangle / V = 1/3 \), where \( \langle n_i \rangle = \langle \sum_i c_i^\dagger c_i \rangle \) and \( \langle n_f \rangle = \langle n_{f,i} \rangle \). Our findings are summarized in Fig.1 which shows the phase diagram of the model in the \( T-U \) plane. At large \( U \) and sufficiently low \( T \), \( T < T_c(U) \), the system develops a charge density wave (CDW). For large \( U \) and/or \( T \), the phase diagram resembles that of its counterpart on the square lattice. While this could have been anticipated, there are important differences with regard to the type of order. This will be discussed below. The most surprising feature is the two regions at low \( T \) and small \( U \), labelled classical charge liquid (CL) and quantum charge liquid (QL), respectively, that remain disordered down to the lowest \( T \). At large \( U \), the temperature-driven phase transition is continuous and within the universality class of the 3-state Potts model. For small \( U \), our data point to a discontinuous transition between the CDW and the CL down to \( T_c(U_c) = 0 \), corresponding to \( U_c \simeq 5.2 \). For large temperatures, a ‘strange metal region’ (WL) is found, where the electrons are weakly localized, followed by an Anderson insulating (AI) and finally a Mott insulting (MI) phase as \( U \) increases. The WL region is only stabilized by the finite extend of the system and is expected to vanish in the thermodynamic limit [6]. AI is

FIG. 1. (a) Phase diagram of the FK model on the triangular lattice in the \( T-U \) plane for \( x_f = 1/3 \) and \( x_c = 2/3 \). The red start indicates the quantum phase transition between the liquid states. The inset depicts a sketch of the phases in the thermodynamic limit. Band structure assuming CDW order for \( U = 2 \) (b) and \( U = 5 \) (c).

FIG. 2. Specific heat as a function of \( T \) for \( U = 2 \) (a), \( U = 5 \) (b) and \( U = 7 \) (c). Momentum resolved susceptibility of the f-charges \( \chi(q) \) for \( U = 5 \) (d) and \( U = 7 \) (f). (e) finite size scaling of \( \chi(Q_{\text{Max}}) \) with \( L \). PCA of the three largest eigenvalues of the susceptibility matrix \( \chi_{i,j} \) for \( U = 5 \) (g) and for the two largest eigenvalues for \( U = 7 \) (h) plotted for a range of \( T \) for \( L = 20 \) and \( L = 21 \) respectively.
characterized by a finite density of states (DOS) at the Fermi level and a volume-independent inverse participation ratio (IPR). In the MI phase, the chemical potential lies within a \( U \)-dependent spectral gap. This all closely resembles earlier findings for the FKM on a square lattice of reference \([6]\), where a full characterization of these phases can be found. In what follows, we focus on the fundamentally new features that emerge from the interplay of localized and itinerant degrees of freedom in a geometrically frustrated environment.

In the expansion in terms of \( t/U \), mentioned above, higher order terms beyond the nearest-neighbour Ising-like interaction proportional to \( t^2/U \) can be systematically derived. In next-to-leading order, i.e. \( t^3/U^2 \), it yields a term that couples the degrees of freedom on a triangular plaquette. The range of the effective interaction increases with powers of \( t/U \), increasing the frustration which eventually leads to the melting of the order. This procedure is carried out in the Supplemental Material (SM) up to order \( t^4/U^3 \) \([25, 26]\).

At large \( U \), the effective term together with the \( x_f = 1/3 \) restriction favors the existence of a low \( T \) ordered phase possessing a 3-fold degeneracy. A possible order parameter for this phase is \( \phi_{1/3} = \frac{3}{2} \sum_x e^{i \pi (r_x - r_y) q_f} \) that equals \( \phi_{1/3} = 1, e^{i \pi} \) or \( e^{i \pi} \) depending on which of the three degenerate ground states is realized. One such configuration is depicted in the inset of Fig.1, while the others can be obtained by a lattice rotation around the center of a triangular plaquette.

The order parameter symmetry implies that the associated finite-\( T \) transition belongs to the two-dimensional 3-state Potts model universality class. Indeed, this is borne out by our numerical results. We find for the correlation length exponent \( \nu_{3p} = 5/6 \) and the exponent of the \( T \) dependence of the susceptibility of the \( f \)-charges, \( \gamma_{3p} = 13/9 \) \([27]\) the values \( \nu = 0.8031(114) \) and \( \gamma = 1.4748(329) \); for details, see \([25]\).

The specific heat \( C_v \) across the charge ordering transition for \( U = 7 \) is shown in Fig. 2-(c). A high-temperature local maximum (not shown in the figure) coincides with the \( T \) scale where double-occupancy is sharply suppressed as \( T \) is lowered. At lower \( T \), the divergent \( C_v \), confirmed by the finite-size scaling, corresponds to the transition into the CDW state. The static susceptibility, \( \chi(\omega \to 0, q, T) \) is depicted in Fig.2-(f). It shows a maximum at the propagation vector \( Q_{\text{CDW}} \) of the CDW, i.e., \( q = Q_{\text{CDW}} = 2\pi/3\{1, \sqrt{3}\} \). Fig.2-(c) (orange line) shows that, for \( T < T_c \), \( \chi(\omega \to 0, Q_{\text{CDW}}, T) \propto L^2 \), in line with the existence of long-range order. The reconstructed band structure of the \( e \)-electrons within this symmetry broken phase at \( T = 0 \) is given in Fig.1-(c); \( \mu_c \) lies in the \( U \)-dependent bandgap. Assuming that the ordered phase persists as a function of \( U \) one expects an indirect closing of the bandgap for \( U = 3 \). Fig.1-(b) shows the band structure for \( U < 3 \). The charge order, however, vanishes at \( U_c > 3 \), see Fig.1-(a).

We now address the region \( U < U_c \). Fig.2-(a) and (b) show \( C_v(T) \) in this region for a representative value of \( U \) within the QL and CL respectively. The high-\( T \) features are similar to those found for \( U > U_c \). In contrast to the \( U > U_c \) case, \( C_v(T, L) \) remains non-singular for \( L \to \infty \), indicating that the transition has been replaced by a crossover. The insets of Figs.2-(a) and (b) depict \( C_v(T) \) on a logarithmic scale and indicate that the behavior is compatible with power-law scaling in \( T \) implying gapless excitations in the system. The fact that both power-laws are distinct highlights that there are indeed two different zero temperature phases. Moreover, neither is compatible with Fermi-liquid behavior, i.e., with \( C_v(T) \propto T \). Within these phases, \( \chi(0, q, T) \) is no longer maximal for \( q = Q_{\text{CDW}} \) but rather for \( q = Q_{\text{Max}} = \pi\{1, 1/\sqrt{3}\} \), as shown in Fig.2-(d) for CL (similar for QL). Interestingly, \( Q_{\text{Max}} \) corresponds to the wave vector of a CDW expected for filling fractions \( x_f = 1/4 \) and \( x_v = 3/4 \), see Fig.2-(d). However, the order parameter \( \phi_{1/4} \) of this phase, explicitly given in \([25]\), vanishes (see below). The scaling of \( \chi(0, Q_{\text{Max}}, T) \) with \( L \) shown in Fig.2-(e) (blue line) is \( \chi(\omega \to 0, Q_{\text{Max}}, T) \propto L^a \) with \( a \approx 0.0616 \), which indicates that the CL region is incompatible with the existence of long-range order of that type. To further substantiate the characterization of the liquid region, we turn to a principal component analysis (PCA) \([28]\) of the charge excitations in CL vs CDW. This method allows for a dimensional reduction when visualizing multivariate data and has previously been applied to the 2d ferromagnetic Ising model \([29]\). Figs.2-(g) and (h) show the projection of different thermalized configurations onto the eigenvectors corresponding to the three largest eigenvalues of \( \chi_{i,j} = \langle n_{i,f}, n_{i,f,j} \rangle \) for different \( T \) and for \( U = 5 \) and \( U = 7 \) respectively. In the ordered phase, low temperature configurations cluster around one of the three ground-states, see Fig.2-(h) and \( \chi_{i,j} \) has two dominating eigenvalues that are significantly larger then all others. Within the CL region, however, configurations cluster on a four-fold symmetric structure with \( \chi_{i,j} \) having three dominating eigenvectors. Note, however, that this finding does not imply the existence of a long-range ordered four-fold state, which would be incompatible with the 1/3 filling, unless phase separation occurs. This is corroborated by a vanishing order parameter and associated susceptibility \( \chi(Q_{\text{Max}}) = \chi(L) \) as \( L \to \infty \), see Fig.2-(e) \([25]\).

Another possibility in line with the power-law behavior in \( \chi(L) \) and \( C_v(T) \) for \( T \to 0 \) is the existence of a phase transition, for a \( T \) around the low \( T \) peak of the specific heat, to a KT-like phase without long-range order. Indeed, KT transitions can occur for clock models with \( Z_q \) symmetry \([30]\). While our numerical results alone cannot exclude this scenario, this only arises for clock models with \( q > 4 \) \([30–32]\). Moreover the systems seems to retain its full symmetry, isomorphic to the permutation group.
This sharp drop is used to determine the crossover line. c

Signaling of IPR finite value as a function of vicinity of the Fermi energy and its scaling with system size. The inset of Fig.3-(a) depicts the behavior of the energy for two values of U

In contrast, for the CL, we expect that an effective Hamiltonian in terms of classical charges that interact via short-range interactions, exists. However, up to fourth order neither the truncated model nor the variational one do seem to capture the properties of the CL phase. This suggest that higher order terms are necessary to fully capture the properties of this phase. We thus have obtained an unexpectedly rich phase diagram of the 1/3-filled FKM on the triangular lattice: for intermediate-to-large coupling, the temperature-driven phase transition from the charge ordered to the disordered phase belongs to the 3-state Potts model universality class; more importantly, for weak coupling and low temperatures, we show the existence of a charge liquid region divided by a cross-over line that terminates in a zero temperature transition at U_{QCP}. The classical liquid, which ensues for U > U_{QCP}, is expected to be captured by an effective,
finite-ranged classical model. However, we were not able to obtain such a model with terms up to the fourth order in terms of $t/U$. Approaching the quantum phase transition from within the classical liquid, the gap of the quantum degrees of freedom vanishes at $U_{QCP}$. This phase transition is reflected in the behavior of the specific heat which changes from $C'_v \propto T^2$ to $C'_v \propto T^4$ as $U_{QCP}$ is crossed.

Our results offer a new vantage point to address how quantum liquids can emerge from their classical counterparts. They also elucidate how classical liquids form through melting of phase separated states in the presence of frustrated interactions that favour the proliferation of domain walls. Addressing the fate of the charge liquids in the presence of a non-vanishing hybridization between the localized charges and the conduction electrons is an interesting open question with immediate experimental relevance [34, 35]. Understanding the fate of the CL phase as the FKM approaches the Hubbard model may help unraveling the phase diagram of the Hubbard model on the triangular lattice [36]. Another promising future direction is the generalization of the Ising variables to continuous symmetry groups which may shed new light on emergence of gauge fields and charge fractionalization.

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Classical liquid induced by quantum fluctuations

Miguel M. Oliveira\textsuperscript{(a)}, Pedro Ribeiro\textsuperscript{(a,b)}, and Stefan Kirchner\textsuperscript{(c)}
\textsuperscript{(a)}CeFEMA, Instituto Superior Técnico, Universidade de Lisboa Av. Rovisco Pais, 1049-001 Lisboa, Portugal
\textsuperscript{(b)}Beijing Computational Science Research Center, Beijing 100193, China
\textsuperscript{(c)}Zhejiang Institute of Modern Physics, Zhejiang University, Hangzhou, Zhejiang 310027, China

Summary: Below we provide additional technical details and further numerical results supplementing the conclusions from the main text.

Monte Carlo Algorithm
The partition function of the model can be written as

\[ Z = \sum_{\{n_f\}} e^{-\beta F(n_f)}, \]  

(S1)

with

\[ F(n_f) = -\frac{1}{\beta} \sum_{\nu} \log \left[ 1 + e^{-\beta [\epsilon_\nu(n_f) - \mu_c]} \right] - \mu_f N_f. \]  

(S2)

the free energy of a given \( n_f \) electron configuration.

The mean value of any observable \( O \) depending only on the classical charges, is thus computed as

\[ \langle O \rangle = \sum_{\{n_f\}} O(n_f) e^{-\beta F(n_f)}. \]  

(S3)

Determination of the chemical potentials
As mentioned in the main text, the triangular lattice presents an additional challenge when compared to the square lattice at half-filling. In the later, the FK model possesses a particle-hole symmetry that can be exploited to fix the system at half-filling for both electron species. On the triangular lattice, however, no such symmetry exists and therefore the chemical potentials have to be determined self-consistently in order to fix the desired fillings.

This is accomplished via the following Newton-Raphson algorithm: we start the Monte-Carlo algorithm with a guess for the chemical potentials; during the simulation we gather data that allows for the computation of the fillings of both species as well as other quantities necessary for the Newton algorithm; we then use the results of the simulation to update the chemical potentials and repeat the previous procedure until the fillings converge to the desired values. The convergence of this procedure is particularly computational resource consuming for small \( U \) and \( T \) and turned out to be the main limiting factor to our numerical analysis.

Potts transition
In order to characterize the temperature driven phase transition for large \( U \) we have computed the order parameter susceptibility

\[ \chi_{\phi_{1/3}} = \beta V \left( \langle |\phi|^2 \rangle - \langle |\phi| \rangle^2 \right) \]  

(S4)

and the Binder cumulant

\[ U_4 = 1 - \frac{\langle |\phi|^4 \rangle}{3\langle |\phi|^2 \rangle}. \]  

(S5)

These quantities are given in Fig.S1. The crossing point of the Binder cumulant was used to determine the critical temperature. Based on the finite size scaling of the susceptibility and the scaling of the derivatives with respect to beta of the Binder cumulant and the order parameter, we obtained the critical exponents quoted in the main text. The finite size scaling analysis used followed closely what is described in Ref. [37].

Double occupancy
The number of doubly occupied sites is defined as

\[ D = \frac{1}{V} \sum_i \langle n_{f,i} c_i^\dagger c_i \rangle. \]  

(S6)
FIG. S1. Temperature driven transition for large $U$ ($U = 30$). Order parameter’s amplitude (a); Susceptibility (b); and Binder cummulant (c) plotted as a function of temperature for different systems sizes. The inset shows a zoom of the crossing point.

FIG. S2. Double occupancy (a) and specific heat (b) as a function of temperature for $U = 4$. Phase diagram (c) where the red line marks the position of the high temperature bump in the specific heat, which is associated with the suppression of double occupancy. This quantity is explicitly computed as

$$
\langle n_{f,i} c_i^\dagger c_i \rangle = \sum_{\{n_f\}} n_{f,i} \left( \sum_\alpha |\langle i | \alpha \rangle|^2 n_F(\epsilon_{\alpha}) \right) e^{-\beta F(n_f)}
$$

where $|\alpha\rangle$ is an eigenvector of the single-particle Hamiltonian $H(n_f)$ with energy $\epsilon_{\alpha}$ obtained for a given configuration of the classical charges. $n_F(\epsilon_{\alpha})$ is the Fermi occupation number of the eigenstate $|\alpha\rangle$.

Figs. S2 (a) and (b) show that the high temperature peak of the specific heat coincides with the abrupt suppression of the double occupancy. Fig. S2 (c) depicts in red line the maximum of the high temperature peak in the $C_v$. The low temperature phases studied in the main text arise for temperatures one order of magnitude lower than the sudden suppression of doubly occupied states and thus the phenomena reported in the main text cannot be attributed to it.

Nature of the CL-CDW phase transition
Our numerical results indicate that the transition between the CL and CDW phases, shown in the phase diagram of Fig. 1, is of a differed nature from that between CDW and MI.

Figs. S3 (a,b,c) and S3 (d,e,f) show the histograms of the energy (a,b,c) and of the order parameter (d,e,f) for different values of $U$ across the CL-CDW phase transition for $L = 12$.

FIG. S3. Histograms of the energy (a,b,c) and of the order parameter (d,e,f) for different values of $U$ across the CL-CDW phase transition for $L = 12$.

Phase separation
As mentioned in the main text, the liquid phase is characterized by a susceptibility $\chi(Q)$ which is enhanced for the same wave-vectors as those of the ordered phase that ensues for fillings $x_f = 1/4$ and $x_c = 3/4$. The order parameter of that phase is given by

$$
\phi_{1/4} = 4 V \sum_i \left[ e^{2\pi i \delta_{A,i}} + e^{\pi i \delta_{B,i}} + e^{3\pi i \delta_{C,i}} + e^{3\pi i \delta_{D,i}} \right] n_{f,i},
$$

where $\delta_{A,i}$ is a function that is 1 when $i$ belongs to sublattice $A$ and 0 when it is not. Since an ordered phase of this type would only occupy one of the four sub-lattices, this order parameter is able to distinguish between those four different ground-states. As already stressed in the main part of the manuscript, this type of order is absent in the CL phase which follows from $\phi_{1/4}$ vanishing as $1/L$. This also rules out the occurrence of phase separation with a non-vanishing volume fraction of a $\phi_{1/4}$ phase. Interestingly, the effective classical model obtained by truncating the $t/U$ expansion to fourth order does exhibit phase separation with a $\phi_{1/4}$ phase.

Effective Classical Model
In this section we outline the derivation of the effective
a) b)

FIG. S4. (a) Configuration of an effective classical model with only a square plaquette term. (b) Typical configuration of the FKM within the CL phase for $U = 5$ and $T = 0.0085$.

classical model, obtained from an expansion in small $t/U$, at finite temperature. Results for zero temperature were already given in ref. [26].

For simplicity, the representation was changed from occupation to spin variables $s_i = 2n_{f,i} - 1$. Next, we expand the free energy in terms of $t/U$,

$$ F(s) = F_0(s) + \frac{1}{\beta} \sum_{k=1}^{\infty} \frac{(-1)^k}{k} \text{Tr} [(G_0T)^k] \quad (S9) $$

where

$$ F_0(s) = -\mu_f N_f - \frac{1}{\beta} \text{Tr} \left[ \log \left( -G_0^{-1} \right) \right], \quad (S10) $$

$T$ is the adjacency matrix defined as $T_{i,j} = t$ if $i$ and $j$ are nearest neighbours on the lattice and

$$ G_0^{-1}(i\omega_n) = i\omega_n - \frac{U}{2}(S + 1) + \mu_c. \quad (S11) $$

where $S$ is a diagonal matrix whose entries are the spin variables $S_{i,j} = \delta_{i,j} s_j$.

After summing over the Matsubara frequencies and going over rather tedious combinatorics, we obtain up to $4^{th}$ order and for $T = 0$

$$ E = -\frac{1}{2} \left( \mu_f - \mu_c + \frac{6t^3}{U^2} \right) \sum_i s_i + \left[ \frac{t^2}{2U} - \frac{6t^4}{U^3} \right] \sum_{|i-j|=1} s_is_j + $$

$$ + \frac{3t^3}{2U^2} \sum_{\Delta} s_{\Delta} + \frac{5t^4}{2U^3} \sum_p s_p + \frac{3t^4}{2U^3} \sum_{|i-j|=\sqrt{3}} s_is_j + $$

$$ + \frac{t^4}{U^3} \sum_{|i-j|=2} s_is_j + O \left( \frac{t^5}{U^4} \right). \quad (S12) $$

These coefficients coincide with those given in Ref. [26]. The finite temperature expressions can be obtained in a similar way but will not be reproduced here as they are lengthy and not particularly instructive. In Eq(S12), the $\sum_{|i-j|=1} s_is_j$ term corresponds to an Ising-like interaction over nearest neighbors, $\sum_{|i-j|=\sqrt{3}} s_is_j$ over opposite ends of a rhombus and $\sum_{|i-j|=2} s_is_j$ over next-nearest neighbors. $\sum_{\Delta} s_{\Delta}$ corresponds to a sum over interactions on triangular plaquettes, where $s_{\Delta}$ is a product of the spins belonging to it and $\sum_p s_p$ represents the same for square plaquettes. A sketch of all the mentioned interactions is shown in the inset of Fig. 4- (b) of the main text.

**Variational Classical Model**

The variational model referred to in the main text was obtained assuming a free energy of the form of Eq.(S12) but with arbitrary coefficients $J_a=1,2$.

$$ E = h \sum_i s_i + J_1 \sum_{|i-j|=1} s_i s_j + J_2 \sum_{\Delta} s_{\Delta} + $$

$$ + J_3 \sum_p s_p + J_4 \sum_{|i-j|=\sqrt{3}} s_i s_j + J_5 \sum_{|i-j|=2} s_i s_j. \quad (S13) $$

The couplings were fixed considering 100000 configurations of the classical charges generated with the FKM and fitting the free energy to that of Eq.(S13) with a linear regression on the $J_a=1,2.$ parameters. This procedure was done in Ref.[33] to show that an effective Hamiltonian obtained via linear regression can be used to propose more efficient Monte-Carlo updates and thus speed up the dynamics of the original model. The variational coupling constants obtained in this way are shown to vary smoothly with $U$ in Fig.S5.