Charged particles on a 2D plane subject to anisotropic Jahn-Teller interactions.

T. Mertelj\textsuperscript{1,2}, V.V. Kabanov\textsuperscript{1} and D. Mihailovic\textsuperscript{1,2}
\textsuperscript{1}Jozef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia and
\textsuperscript{2}Faculty of Mathematics and Physics, Univ. of Ljubljana, Slovenia
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The properties of a system of charged particles on a 2D lattice, subject to an anisotropic Jahn-Teller-type interaction and 3D Coulomb repulsion are investigated. In the mean-field approximation without Coulomb interaction, the system displays a phase transition of first order. When the long range Coulomb interaction is included, Monte Carlo simulations show that the system displays very diverse mesoscopic textures, ranging from spatially disordered pairs to ordered arrays of stripes, or charged clusters, depending only on the ratio of the two interactions (and the particle density). Remarkably, charged objects with even number of particles are more stable than with odd number of particles. We suggest that the diverse functional behaviour - including superconductivity - observed in oxides can be thought to arise from the self-organization of this type.

The standard theoretical models of strongly correlated electrons, such as the Hubbard model\textsuperscript{2} or the $t$–$J$ model\textsuperscript{2} neglect two important interactions, namely long-range Coulomb repulsion and lattice distortions caused by charged particles. Moreover, these quantum mechanical models are typically used to study $T \approx 0$ properties. As such, these models have found limited applicability in predicting the finite-temperature functional behaviour in systems such as cuprate superconductors and other oxides. An important aspect of the problem which has been of great interest recently is the existence of intrinsic mesoscale inhomogeneity in these systems, for which there is mounting experimental evidence from neutron scattering\textsuperscript{3}, XAFS\textsuperscript{4}, STM\textsuperscript{5} and time-resolved carrier dynamics\textsuperscript{6}, amongst others.\textsuperscript{7} There is emerging consensus that in doped cuprates charged carrier phases may phase segregate to form nano-scale textures. These are believed to be of importance for achieving their functional properties, and particularly superconductivity. The idea of charge segregation in cuprates appeared soon after the discovery of superconductivity\textsuperscript{8, 9, 10}, but in most cases, long-range Coulomb repulsion was not considered. More recently it was suggested that interplay of short range lattice attraction and long-range Coulomb repulsion could lead to the formation short metallic or insulating strings of polarons\textsuperscript{11, 12}. Since an isotropic interaction cannot lead to stripe formation we suggested instead that an anisotropic mesoscopic Jahn-Teller interaction between electrons and $k \neq 0$ optical phonons might lead to the formation of pairs and stripes\textsuperscript{13}. A slightly different approach, based on elasticity was considered more recently for the case of manganites by Kugel and Khomskii\textsuperscript{14} using the methods of Eremin et al.\textsuperscript{15}, and by Shenoy et al.\textsuperscript{16}. The importance of the interplay of long-range and short range forces within an Ising-like model was discussed by Low et al.\textsuperscript{17}.

The fundamental question which we try and answer here is how charged particles order in the presence of anisotropic Jahn-Teller type interaction, particularly when their density becomes large. We consider charged particles on a 2D square lattice subject to only the long-range Coulomb interaction and an anisotropic Jahn-Teller (JT) deformation. In the mean field (MF) approximation without Coulomb repulsion, the system displays a first order phase transition to an ordered state below some critical temperature. In the presence of Coulomb repulsion global phase separation becomes unfavorable and the system shows mesoscopic phase separation, where the size of charged regions is determined by the competition between ordering energy and the Coulomb energy. Using Monte-Carlo (MC) simulations we show that the system can form many different mesoscopic textures, such as clusters and stripes, depending only on the magnitude of the Coulomb repulsion compared to the anisotropic lattice attraction. Surprisingly, a feature arising from the anisotropy introduced by the Jahn-Teller interaction is that objects with even number of particles are found to be more stable than with odd number particles, which could be significant for superconductivity when tunnelling is included\textsuperscript{18}.

Let us consider the JT model Hamiltonian\textsuperscript{13}, and take only the mode of $B_{1g}$ symmetry:

$$H_{JT} = g \sum_{\mathbf{r}, \mathbf{r}'} \sigma_{3,1}(\mathbf{r})(b_{1+r}^\dagger + b_{1+r}),$$

where the Pauli matrix $\sigma_{3,1}$ describes the electronic doublet, $g$ is a constant, and $f(\mathbf{r}) = (r_x^2 - r_y^2)f_0(\mathbf{r})$ where $f_0(r)$ describes the effective range of the interaction\textsuperscript{13}.

The model is reduced to a lattice gas model by using the adiabatic approximattion for the phonon field\textsuperscript{14, 19}. The Hamiltonian in the pseudospin ($S = 1$) representation is given by:

$$H^{LG}_{JT-C} = \sum_{i,j} (-V_i(\mathbf{j})S_i^x S_j^x + V_c(\mathbf{j})Q_i Q_j),$$

where $Q_i = (S_i^z)^2$, $V_c(\mathbf{m}) = e^2/\varepsilon_0 a m$ is 3D Coulomb potential, $e$ is the charge of the electron, $\varepsilon_0$ is the static dielectric constant and $a$ is the effective lattice constant. $S^z = \pm 1$ corresponds to the state with $n_{1,2} = 1$, $n_{2,1} = 0$, and $S^z = 0$ corresponds to the state with $n_{1,2} = 2$, $n_{2,1} = 0$. The importance of the interplay of long-range and short range forces within an Ising-like model was discussed by Low et al.\textsuperscript{17}.

The properties of a system of charged particles on a 2D plane subject to anisotropic Jahn-Teller-type interaction and 3D Coulomb repulsion are investigated. In the mean-field approximation without Coulomb interaction, the system displays a phase transition of first order. When the long range Coulomb interaction is included, Monte Carlo simulations show that the system displays very diverse mesoscopic textures, ranging from spatially disordered pairs to ordered arrays of stripes, or charged clusters, depending only on the ratio of the two interactions (and the particle density). Remarkably, charged objects with even number of particles are more stable than with odd number of particles. We suggest that the diverse functional behaviour - including superconductivity - observed in oxides can be thought to arise from the self-organization of this type.
and \( S_i^z = 0 \) to \( n_1 = n_2 = 0 \). Simultaneous occupancy of both levels is excluded due to the large on-site Coulomb repulsion. The anisotropic short range attraction is then given by:

\[
V_l(m) = g^2 / \omega \sum_i \frac{f(i) f(m + i)}{2(A_2 + A_1 U(k))}.
\]

A similar interaction can also be derived by considering the interaction of the electronic doublet with the strain of \( B_{1g} \) symmetry, taking into account St.Venant’s compatibility conditions. Anisotropic attraction caused by elasticity has the form:

\[
V_l(m) = - \sum_k \exp(ik \cdot m) \frac{g^2}{2(A_2 + A_1 U(k))}.
\]

Here \( A_j \) are the components of the elastic modulus tensor, and \( U(k) = \frac{(k_x^2 - k_y^2)^2}{k_x^2 + 8A_1 k_x k_y} \). Compared to (3), where the range of the interaction was defined by the coupling to optical phonons, the interaction (4) decays as \( 1/r^2 \) (in 2D) at large distances. Since these attractive forces decay faster than the Coulomb repulsion at large distances, the net potential may have a minimum at short distances.

Our goal is to study the model (2) at constant average density of charged particles, \( n = \frac{1}{N} \sum_i Q_i \), where \( N \) is the total number of sites. However, to clarify the physical picture we first consider a system with a fixed chemical potential by adding the term \(-\mu \sum_i Q_i\) to the Hamiltonian (2).

Models such as (2), but in the absence of the long-range forces were previously studied on the basis of the molecular-field approximation. The mean-field equations for particle density \( n \) and pseudospin magnetization \( M = \frac{1}{N} \sum_i S_i^z \) then have the form:

\[
M = \frac{2\sinh(2zV_l/M/k_BT)}{\exp(-\mu/k_BT) + 2\cosh(2zV_l/M/k_BT)}
\]

\[
n = \frac{2\cosh(2zV_l/M/k_BT)}{\exp(-\mu/k_BT) + 2\cosh(2zV_l/M/k_BT)}
\]

here \( z = 4 \) is the number of the nearest neighbours for a square lattice in 2D and \( k_B \) is the Boltzmann constant. A phase transition to an ordered state with finite \( M \) may be of either first or second order, depending on the value of \( \mu \). For the physically important case \(-2zV_l < \mu < 0\), ordering occurs as a result of the first order phase transition. The two solutions of Eqs. (5,6) with \( M = 0 \) and with \( M \neq 0 \) correspond to two different minima of the free energy. The temperature of the phase transition \( T_{\text{crit}} \) is determined by the condition: \( F(M = 0, \mu, T) = F(M, \mu, T) \) where \( M \) is the solution of Eq. (5). When the number of particles is fixed (Eq.6), the system is unstable with respect to global phase separation below \( T_{\text{crit}} \). As a result, at fixed \( n \) two phases coexist with \( n_0 = n(M = 0, \mu, T) \) and \( n_M = n(M, \mu, T) \), resulting in a liquid-gas-like phase diagram (Fig.1).

To investigate the effects of the long range-forces, we performed MC simulations on the system (2). The simulations were performed on a square lattice with dimensions up to \( L \times L \) sites with \( 10 \leq L \leq 100 \) using a standard Metropolis algorithm in combination with simulated annealing. At constant \( n \) one MC step included a single update for each site with nonzero \( Q_i \), where the trial move consisted from setting \( S_i = 0 \) at the site with nonzero \( Q_i \) and \( S_i = \pm 1 \) at a randomly selected site with zero \( Q_i \). A typical simulated annealing run consisted from a sequence of MC simulations at different temperatures. At each temperature the equilibration phase (10^4—10^6 MC steps) was followed by the averaging phase with the same or greater number of MC steps. Observables were measured after each MC step during the averaging phase only. For \( L \gtrsim 20 \) we observe virtually no dependence of the results on the system size.

Comparing the MC results in absence of Coulomb repulsion shown by \( t_{\text{crit}} \) in Fig. 1 with MF theory we find the usual reduction of \( t_{\text{crit}} \) due to fluctuations in 2D by a factor of \( \sim 2 \).

Next, we include the Coulomb interaction \( V_c(r) \). We use open boundary conditions to avoid complications due to the long range Coulomb forces and ensure overall electroneutrality by adding a uniformly charged background electrostatic potential (jellium) to Eq. (2). The short range potential \( V_l(i) = V_l(i)e\alpha/\epsilon^2 \) was taken to be nonzero only for \(|i| < 2 \) and is therefore specified only for nearest, and next-nearest neighbours as \( V_l(1,0) \) and \( V_l(1,1) \) respectively.

The anisotropy of the short range potential has a profound influence on the particle ordering. We can see this if we fix \( V_l(1,0) = -1 \), at a density \( n = 0.2 \) and vary the next-nearest neighbour potential \( V_l(1,1) \) in the range from \(-1 \) to \( 1 \). When \( V_l(1,1) = 0 \), the attraction is "ferrodistortive" in all directions, while for positive \( V_l(1,1) > 0 \) the interaction is "antiferrodistortive" along the diagonals. The resulting clustering and ordering of clusters at \( t = 0.04 \) is shown in Fig. 2a. As expected, a more symmetric attraction potential leads to the formation of more symmetric clusters. On the other hand, for \( V_l(1,1) = 1 \), the "antiferrodistortive" interaction along diagonals prevails, resulting in diagonal stripes.

In the temperature region where clusters partially order the heat capacity \( c_L = \partial \langle E \rangle_L / \partial T \) where \( E \) is the total energy) displays the peak at \( t_{\text{co}} \). The peak displays no scaling with \( L \) indicating that no long range ordering of clusters appears. Inspection of the particle distribution snapshots at low temperatures (Fig. 2a) reveals that finite size domains form. Within domains the clusters are perfectly ordered. The domain wall dynamics seems to be much slower than our MC simulation timescale preventing domains to grow. The effective \( L \) is therefore limited by the domain size. This explains the absence of the scaling and clear evidence for a phase transition near \( t_{\text{co}} \).
We now focus on the shape of the short range potential which promotes the formation of stripes shown in Fig. 2a). We set $v_{1}(1,0) = -1$ and $v_{1}(1,1) = 0$ and study the density dependence. Since the inclusion of the Coulomb interaction completely suppresses the first order phase transition at $t_{c_{1}}$, we measure the nearest neighbor density correlation function $g_{pL} = \frac{1}{L^{2}} \sum_{i=1}^{L} \langle Q_{i} - n \rangle$, as the characteristic crossover temperature related to the formation of clusters at which $g_{pL}$ rises to 50% of its low temperature value. The dependence of $t_{c_{1}}$ on the density $n$ is shown in the phase diagram in Fig. 1. Without Coulomb repulsion $V_{c}(r)$, $t_{c_{1}}$ follows $t_{c_{2}}$, as expected. The addition of Coulomb repulsion $V_{c}(r)$ results in a significant decrease of $t_{c_{1}}$ and suppression of clustering. At low densities we can estimate the onset for a cluster formation by the temperature, $t_{c_{1}}$, at which $g_{pL}$ becomes positive. It is interesting to note that $t_{c_{1}}$ almost coincides with the $t_{c_{2}}$ line at low $n$ (Fig. 1).

To illustrate this behaviour, in Fig. 2b) we show snapshots of the calculated MC particle distributions at two different temperatures for different densities. The growth and ordering of clusters with decreasing temperature is clearly observed. At low $n$, the particles form mostly pairs with some short stripes. With further increasing density, quadruples gradually replace pairs, then long stripes appear, mixed with quadruples, etc.. At the highest density, stripes prevail forming a labyrinth-like pattern. The density correlation function shows that the correlation length increases with doping, but long range order is never achieved (in contrast to the case without $V_{c}$). Note that while locally, there is no four-fold symmetry, the overall correlation function still retains 4-fold symmetry.

To get further insight in the cluster formation we measured the cluster-size distribution. In Fig. 3 we show the temperature and density dependence of the cluster-size distribution function $x_{L}(j) = \langle N_{p}(j) \rangle / (nL^{2})$, where $N_{p}(j)$ is the total number of particles within clusters of size $j$. At the highest temperature $x_{L}(j)$ is close to the distribution expected for the random ordering. As the temperature is decreased, the number of larger clusters starts to increase at the expense of single particles. Remarkably, as the temperature is further reduced, clusters of certain size start to prevail. This is clearly seen at higher densities (Fig.3). Depending on the density, the prevailing clusters are be pairs up to $n \approx 0.2$, quadruples for $0.1 \lesssim n \lesssim 0.3$ etc.. We note that for a large range of $v_{1}(1,0)$, the system prefers clusters with an even number of particles. Odd particle-number clusters can also form, but have a much narrower parameter range of stability [24].

The robust prevalence of the paired state in a wide region of parameters (Fig. 3 c,d) is particularly interesting from the point of view of superconductivity. In contrast to Bose condensation of mobile intersite bipolarons discussed by Alexandrov and Mott [27], it has been suggested that pair tunnelling between objects such as shown in Fig. 2 can lead to an insulator-to-superconductor transition [13]. A similar situation occurs in manganites and other oxides with the onset of a conductive state at the threshold of percolation, but different textures are expected to arise from the different magnitude (and anisotropy) of $V_{1}(n)$, and static dielectric constant $\epsilon_0$ in the different materials [25].

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Fig. 1. a) The phase diagram generated by $H_{JT}$ (2) with, and without the Coulomb repulsion (CR). The dashed line is the MF critical temperature, while the full triangles ($\blacktriangle$) represent the MC critical temperature, $t_{\text{crit}}$, without CR. The open circles (o) represent $t_{\text{cl}}$, without CR. The open triangles ($\blacktriangle$) represent $t_{\text{cl}}$ while the diagonal crosses (x) represent the onset of clustering, $t_{0}$, in presence of CR. The cluster-ordering temperature (see text), $t_{\text{co}}$, (also incl. CR) is shown as crosses (+). The size of the symbols corresponds to the error bars. b) Typical temperature dependencies of the nearest neighbor density correlation function $g_{0L}$ for $n = 0.18$ in ab-

Fig. 2. a) Snapshots of clusters ordering at $t = 0.04$, $n = 0.2$ and $\nu_{f}(1,0) = -1$ for different diagonal $\nu_{f}(1,1)$ (given in each figure). Grey and black dots represent particles clusters in state $S_{x}^{l} = 1$ and states $S_{x}^{l} = -1$ respectively. The preference for even-particle-number clusters in certain cases is clearly observed, for example for $\nu_{f}(1,1) = -0.2$. b) Snapshots of the particle distribution for two densities at two different temperatures $t = 0.64$ and $t = 0.1$ respectively.

Fig. 3. The temperature dependence of the cluster-size distribution function $x_{L}(j)$ (for the smallest cluster...
sizes) as a function of temperature at two different average densities $n = 0.08$ (a) and $n = 0.18$ (b). $x_L(j)$ as a function of $n$ at the temperature between $t_0$ and $t_{cl}$ (c), and near $t_{co}$ (d). The ranges of the density where pairs prevail are very clearly seen in (d). Error bars represent the standard deviation.