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Elastically-driven polaron patterns: stripes and glass phases.

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We investigate a model of elasticity-driven polaron pattern formation on a 2-dimensional lattice at finite temperatures. We show that at high densities, polaron stripes (glass like) form when the gas is cooled. These stripes melt to a polaron gas upon heating. We attribute the emergent behavior to the role of the long-range, directional elastic interactions that favor a minimum in energy for polaron stripe formation.

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I. INTRODUCTION

Ceramic materials such as cuprates and manganites (and other transition metal oxides) exhibit novel properties of high temperature superconductivity, colossal magnetoresistance, ferro- and antiferro-magnetism as well as multiferrocity¹⁻¹¹. In many of these materials, the competition between long and short range forces, leads to the formation of long range ordered charge, spin and lattice structures such as stripes and domain walls^{2,3}, as well as self-organized and emergent properties often mediated via the lattice¹. Stripes have been observed in ferromagnetic^{12,13}, ferroelectric¹⁴, superconducting^{2,3,15,16}, as well as hybrid superconducting-anti-ferromagnetic materials¹⁷ and narrow band semiconductors^{18,19}. It has been suggested that long range correlations and resulting stripes in the electronic charge- or spin-density influences the response of the material to external controlling affecting the superconducting state^{2,3}, colossal magnetoresistance^{8,20}, or the colossal stressoresistance⁹.

We have previously proposed²¹ an approach to describe the formation of polarons in which the electronic charge couples to the elastic fields to create an effective long range and directional interaction between polarons. At low polaron density and zero temperature, the ground state of the system is a polaron stripe. When these stripes are brought in contact with a heat bath, the stripe survives up to temperatures comparable to the pairing energies of the polarons; for large temperatures, the kinetic energy of the polarons is greater than the binding energy and the system behaves as a polaron gas; at much higher temperatures the individual polarons melt. We can also expect an intermediate temperature regime, where we have small stripe segments and free polarons constantly moving and reorganizing, with a liquid like behavior. To describe the interaction with the thermal bath, we use here a hybrid adiabatic method, where we update the position of the polarons using standard Monte-Carlo (MC) techniques²². The electron density and the elastic fields are calculated self consistently (with the Newton method introduced in²¹) for every Monte-Carlo step. For large densities, as the self consistent calculation of the electronic densities is very slow, we

treat the density of each polaron as a delta function (Δ -model) and only recalculate the updated elastic fields every Monte-Carlo step.

The paper is organized as follows. In the next section we describe the Ginzburg-Landau free energy and compatibility conditions for the description of the elastic medium, and the Holstein coupling of the elastic fields with the polaron wave functions. Next we present the self-consistent Monte-Carlo method and the numerical results we obtained for the dynamics of the multipolaron system at finite temperatures. We conclude with a discussion of the different behaviors we observe (stripe, liquid and gas) for polarons at different temperatures, and the formation of polaron-stripe glass upon rapid cooling. We also discuss the short and long range correlations reflected in the distribution of polaron cluster sizes, and their pair distribution function. Our approach should be viewed as complementary to phenomenological long-range Coulomb fields introduced into polaron ordering discussions (eg.²³) in that we self-consistently deduce the form of long-range elastic fields from lattice compatibility conditions in the presence of polarons.

II. STRAIN DESCRIPTION OF POLARONS IN AN ELASTIC MEDIUM

For the description of the elastic substrate in two dimensions (2D) on a square lattice, we use symmetry adapted principal strains

$$e_1 = \frac{1}{\sqrt{2}}(\epsilon_{xx} + \epsilon_{yy}); \quad e_2 = \frac{1}{\sqrt{2}}(\epsilon_{xx} - \epsilon_{yy}); \quad e_3 = \epsilon_{xy} = \epsilon_{yx}, \quad (1)$$

where e_1 is the compressional (or dilatational) strain, e_2 the deviatoric strain and e_3 the simple shear strain associated with a unit cell. Restricting ourselves to small strains, $\epsilon_{ij} = \frac{1}{2}(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i})$ are the Lagrangian strains. The three strains in 2D are not independent but are related by the integrability or St. Venant compatibility constraint, which in terms of the symmetry adapted strains becomes

$$G = (\partial_x^2 + \partial_y^2)e_1 - (\partial_x^2 - \partial_y^2)e_2 - \sqrt{8}\partial_x\partial_y e_3 = 0$$

(this is true only if there are no dislocations or broken bones in the lattice²⁴).

The elastic energy density for a homogeneous medium is given by

$$F_{el} = \frac{A_1}{2}e_1^2 + \frac{A_2}{2}e_2^2 + \frac{A_3}{2}e_3^2$$

with A_1 , A_2 and A_3 the elastic moduli and the three strains independent. (For details about the elastic fields and the Ginzburg-Landau energy see^{25,26}).

The charged particles are coupled to the elastic deformations of the lattice. For the interactions with the elastic fields, as well as the interaction between polarons, we use an extension of the semi-classical Holstein polaron model with an additional hard core repulsion (the model as well as details of the numerical method for the calculation of the stripes, are given in²¹ and references therein). The interaction is modeled by the Hamiltonian interaction density

$$H_{int} = \chi \sum_i |\Psi_i|^2 e_1, \quad (2)$$

where $\Psi_i = \Psi_i(\mathbf{r})$ is the wave function of the particle i and e_1 is the isotropic, dilatational strain mode, and the hard core interaction between the polarons is described by the Hamiltonian

$$H_c = \sum_{i \neq j} U_0 |\Psi_i|^2 |\Psi_j|^2, \quad (3)$$

with the parameter U_0 determining the strength of a contact interaction. The Schrödinger equation describing the polarons in the presence of the elastic field can be written as

$$i \frac{d\Psi_i}{dt} = -V \nabla^2 \Psi_i + \chi e_1 \Psi_i + U_0 \sum_j |\Psi_j|^2 \Psi_i, \quad (4)$$

where we choose the Planck constant, $\hbar = 1$ to rescale time, and set $k_B = 1$, so that energy and temperatures are measured in units of the overlap integral V . The elastic fields and the electron wavefunctions have been calculated self consistently using the method in²¹ for predefined positions of the electron, and the arrangement in a single stripe corresponds to the minimum of the energy for the system. Here we perform the same calculation, but we use a Monte-Carlo updating method to change the positions of the centers of the polarons, to account for the interaction with a thermal bath.

III. NUMERICAL SIMULATIONS

In our previous work²¹, we showed that the wave function of the polaron is very localized, whereas the elastic fields extend anisotropically to much longer range. Due to this sharp localization of the wave-function, in addition to the self consistent calculation of the polarons we

introduce a simplified calculation (Δ -model), in which we assume that the wave function of each polaron is a delta function, and we only calculate the elastic fields for each MC step. We have performed the MC simulation with the self consistent calculation of the polarons, as well as the Δ -model, and find the same qualitative behavior – the difference we obtain in the average energy (for the same temperature and after a long MC simulation) is less than 5%. The advantage of the Δ -model is that we avoid the computationally expensive self consistent calculation of the polarons. The calculations presented in this paper are performed using the Δ -model on a 55×55 lattice, but they are qualitative the same as our calculations on smaller lattices and for small numbers of polarons using the full self consistent calculation. For the Monte-Carlo simulations we typically start at zero temperature with the polarons arranged in stripes, and we increase the temperature in small steps. For every temperature increment we perform 30 to 100 thousand Monte-Carlo steps. We smooth the fluctuations of the energy curves by taking the average of the energy over 50 realizations. The energy as a function of temperature for different numbers of polarons is plotted in Fig. 1.

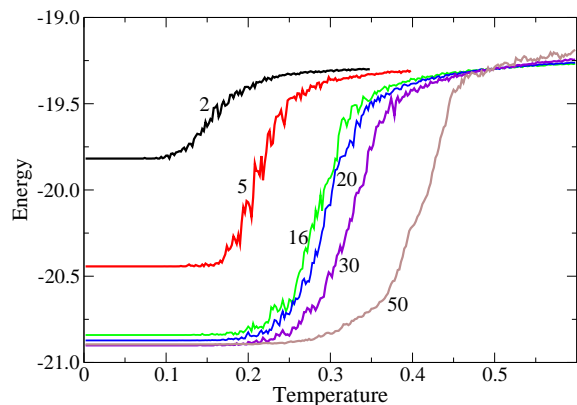


FIG. 1: Monte-Carlo results of the elastic energy with N_p polarons on a 55×55 lattice as a function of temperature for $N_p = 2, 5, 16, 30, 50$.

As we see in Fig. 1, there is a transition interval in the energy as we increase the temperature from zero to 0.6. For two polarons, the low temperature limit corresponds to a stable bound state, while the high temperature limit corresponds to uncorrelated polaron motion. In the transition interval we observe the formation of unstable bound states, with a lifetime that decreases as the temperature increases.

Similar behavior is observed in systems with larger numbers of polarons, as we see in Figs. 1 and 2. At low temperature we have a stable stripe. There is a transition at intermediate temperatures, and at high temperatures we have almost independent polaron motion. We identify three regimes: (A) below the transition temperature, we have stable stripes; (B) in the transition regime the stripe becomes unstable and breaks into smaller stripes

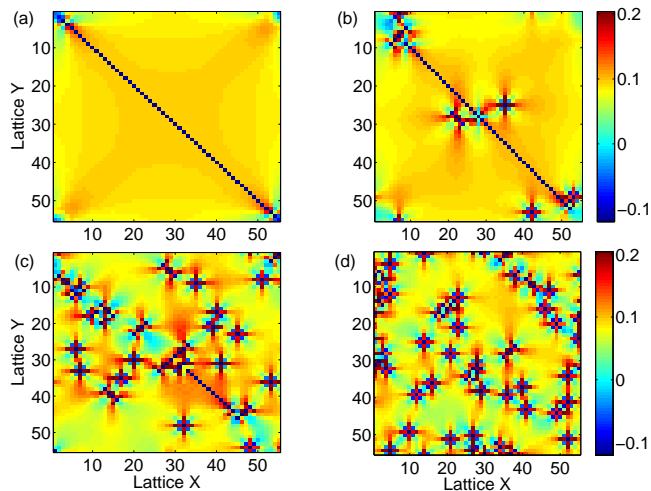


FIG. 2: The strain e_1 strain for 50 polarons on a 55×55 lattice, and for different temperatures. (a) Stripe of polarons at zero temperature. (b) Low temperature transition regime $T=0.38$. The long stripe becomes unstable and a small number of polarons escape, moving alone or forming smaller stripes. (c) Intermediate temperature transition regime with $T=0.42$. The long stripe is almost completely destroyed. Individual polarons move on the lattice and form smaller stripes with a finite lifetime. (d) High temperature regime with $T=0.6$. All polarons move independently: Small stripes are formed but their life time is very short.

and freely moving polarons, which continuously recombine to form other finite lifetime stripe segments; in this temperature range, the polarons exhibit liquid like behavior; (C) at high temperatures we have almost uncorrelated, gas-like polaron dynamics. In Fig. 2 we plot the e_1 strain for a system of 50 polaron on a 55×55 lattice.

As we can see in Fig. 2(b), at the beginning of the transition, a small number of polarons thermally escape from the large stripe. The number of polarons that escape increases as the temperature increases. These freed polarons can either move independently or they form smaller stripes with finite lifetime. This formation of small transient stripes, as well as the free polarons, can be seen in Fig. 2 (b and c). The behavior of the polarons resembles that of liquids, where there is a short range order with a finite lifetime. For high temperatures, the formation of transient stripes becomes rare, and their life time shrinks to few MC steps, as for a gas.

In Fig. 3 we plot the energy of the system with 50 polarons on a 55×55 lattice, when we slowly decrease the temperature (the temperature step is $0.001 - 0.005$, and we perform 50-100 thousand Monte-Carlo steps for every T) starting from the high temperature polaron gas (Fig. 4c). For comparison, we also plot the average energy when we heat, starting from the stripe ground state. When we decrease the temperature, depending on the cooling rate and the number of polarons, the system cannot easily reach the ground state to form single stripe. This can only happen when the number of po-

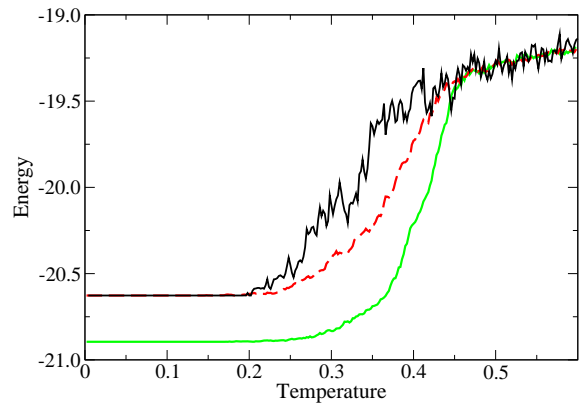


FIG. 3: Hysteresis loop when a system of 55 polarons is cooled, starting with a thermalized polaron gas at high temperatures. The upper line is the energy when cooled from the polaron gas, while the lower line corresponds to the average energy upon heating the system starting with a stripe at $T = 0$. When the system is cooled, it freezes into a higher energy glassy state with many smaller stripes. The dashed line is the average energy obtained by re-heating the system starting from the frozen, many stripes glassy state obtained upon cooling.

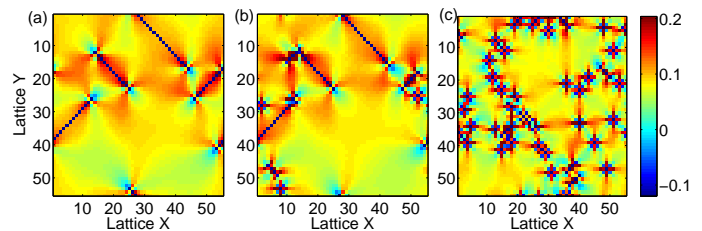


FIG. 4: The strain field e_1 obtained from cooling an initially thermalized polaron gas at high T . (a) The frozen glassy state with many stripes ($T = 0$). (b) Intermediate temperature $T = 0.34$ where the glassy state is almost formed. (c) The initial high temperature polaron gas for $T = 0.6$.

larons is small (of the order of 10-15) and the cooling rate is slow (100 thousand Monte-Carlo steps for each temperature step, with very small temperature steps). The energy landscape exhibits many local minima, each corresponding to a different stripe configuration, separated by energy barriers. The height of these barriers increases with the number of polarons. For small number of polarons, the temperature fluctuations can overcome the barriers and the system relaxes to the absolute ground state faster. For a large number of polarons, or fast cooling rates, there is a hysteretic formation of many small stripes (Fig. 4b). As the temperature decreases, the lifetime of these stripes increases and, as a result, the polarons are trapped into this meta-stable state (Fig. 4a). As we see in Fig. 3, the energy of this meta-stable state is higher than the ground state, but the energy barrier separating it from the ground state is very large, limiting transitions between them.

From Figs. 2 and 4 we see that there is a formation

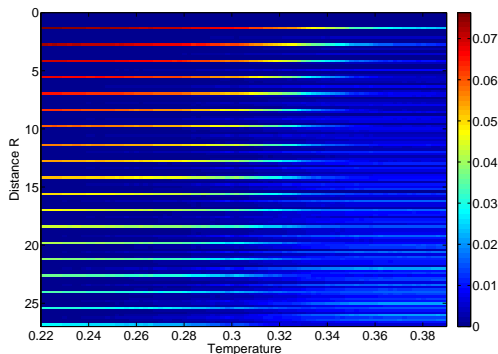


FIG. 5: Pair distribution function density plot as a function of temperature for $N_p = 30$ on a 55×55 lattice. A regular arrangement of delta-function peaks with zero background fluctuations is seen for small temperatures, but as the temperature increases the long range correlation becomes weaker and the corresponding peaks are replaced by a background fluctuation. At even higher temperatures, even the short range correlations disappear and the pair distribution becomes almost uniform.

of polaron clusters and the distribution of cluster sizes changes with temperature (see also movies in supplementary materials²⁷). The distribution of cluster sizes is a robust delta function at low and high temperatures (centered around N_p and 1, respectively). For intermediate temperatures the system exhibits a broad distribution of cluster sizes and associated time scales. Multi-time scale responses are likely but beyond the statistics of our small system sizes.

We also calculate the pair distribution function for the polarons, and follow its changes with temperature. We start heating the system with the polarons arranged in the ground state of a single stripe for $T = 0$. At this temperature the pair distribution function consists of well defined delta-function peaks. These peaks are seen in Fig. 5 as lines in the density plots. As the temperature increases, the long range correlations disappear, and the peaks corresponding to large separation between the polarons decrease, while the background of the pair distribution function becomes more uniform. At higher temperatures, all the peaks disappear, and the pair distribution function becomes uniform (to the degree permitted by the discretization of our simulation box). As we see from Fig. 5, the correlation length of the stripe decreases as we increase the temperature, from the total length of the stripe at $T = 0$ to zero at high temperatures. In the transition region the pair distribution function confirms that we have liquid like behavior; there is no long range correlation, but we do have short range correlation, and the formation of smaller stripes. This is typical behavior of the pair distribution function for liquids.

IV. CONCLUSION

We have used a classical Monte-Carlo method to study the variations with temperature of a many polaron system coupled adiabatically through an elastic substrate. The ground state of the system corresponds to the formation of an extended stripe, with all the polarons arranged parallel to the $\langle 11 \rangle$ direction of the underlying square lattice. When the temperature is increased, the system undergoes a transition from a single stable stripe to smaller meta-stable stripes with liquid like behavior, and finally, at higher temperatures, to a weakly correlated polaron gas. When cooling this polaron gas, we observe the formation of a hysteretic glassy meta-stable state with many stripe segments at low temperature. There is a high energy barrier that does not allow the polarons to evolve from this glassy state, to the single stripe ground state. The formation of the glassy state depends on the cooling rate and the density of polarons. We also calculate the pair distribution function for the polarons, and find that at intermediate temperatures there is a short range correlation but the long range correlation is lost, consistent with a liquid-like phase. At higher temperatures, even the short range correlation is lost, and the system behaves like a polaron gas.

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