A Real Space Description of Magnetic Field Induced Melting in the Charge Ordered Manganese: I. The Clean Limit

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We study the melting of charge order in the half doped manganese using a model that incorporates double exchange, antiferromagnetic superexchange, and Jahn-Teller coupling between electrons and phonons. We primarily use a real space Monte Carlo technique to study the phase diagram in terms of applied field ($h$) and temperature ($T$), exploring the melting of charge order with increasing $h$ and its recovery on decreasing $h$. We observe hysteresis in this response, discover that the ‘field melted’ high conductance state can be spatially inhomogeneous even in the absence of extrinsic disorder and find that the hysteretic response plays out in the background of a curious equilibrium physics of the melting transition. Our extensive numerical work, exploring $h$, $T$, and the electronic parameter space, is backed up by discussion of simpler limiting cases, and a Landau framework for explaining the hysteresis and field induced transitions. We compare our results in detail to experimental data on the half doped manganese. This paper focuses on our results in the ‘clean’ systems, a companion paper studies the effect of cation disorder on the melting phenomena.

I. INTRODUCTION

The manganites, $LnMnO_3$, where $Ln$ belongs to the lanthanide family, display a rich variety of phases as a function of hole doping and bandwidth. Prominent among these are the ferromagnetic metal (FM-M) and the antiferromagnetic charge ordered insulating (AF-CO-I) states. The ferromagnetic metal usually shows up in manganites with large bandwidth, i.e., large mean cation radius, $r_A$, for hole doping $x \sim 0.2 - 0.5$, while the AF-CO-I state is observed in low bandwidth materials at commensurate doping, $x \sim 0.5$. This ‘half doped’ state is especially interesting since it allows systematic study of phase competition and the role of disorder. The $x = 0.5$ state has been extensively probed experimentally [14-17,52,18,19,20,21], and also analysed theoretically [17,18,19,20,21].

It is known that low bandwidth (BW) materials have CE magnetic order with a related checkerboard charge order and concomitant orbital order are insulating (CE-CO-I), while the large BW materials have a FM-M ground state. At intermediate BW some materials have ‘A type’ magnetic order. Application of a magnetic field can melt the charge order and convert the CE-CO-I to a ferromagnetic metal. The melting transition appears to be abrupt and is accompanied by hysteresis in response to field sweep.

The general understanding of field induced melting of charge order is that an applied field destabilises the CE magnetic order in favour of ferromagnetism, and the intimate coupling of charge order and CE magnetic order results in the ‘melting’ of charge order. Some aspects of the melting problem are well studied. (i) The thermodynamic melting field $h_c$ (which is bracketed by the actual switching fields $h_{c1}^\pm$ discussed later) is small. The associated energy scale $g\mu_B h_c \ll k_BT_{CO}$, where $T_{CO}$ is the zero field melting temperature. The smallness of $h_c$ is attributed to the small energy difference between the CE-CO-I and FM-M states. (ii) The field induced transition is believed to be first order in nature and is accompanied by hysteresis arising out of competing metastable minima, particularly for materials close to the CE-CO-I - FM-M phase boundary. (iii) The field induced FM-M is believed to be homogeneous, and is so assumed in theoretical studies. These general observations set important benchmarks for any detailed theory, but some key issues remained unresolved.

(i) The nature of the finite field state: While the CE-CO-I state melts on applying a magnetic field, it was not clear that the ground state is a homogeneous metal. Recent experiments [4,5] in fact demonstrate that the melted state in actually inhomogeneous. We explicitly show that at half doping, for bandwidth materials with weak CE-CO-I state, phase separation at intermediate fields is inevitable and the ‘melted’ state is at best a percolative metal. It is only at large fields, does one recover the homogeneous FM-M final state. Thus, contrary to the belief that the field melting is a discontinuous transition, we contend that the transition is actually continuous, with equilibrium phase separated states interpolating between the initial ($h = 0$) CE-CO-I and the final homogeneous FM-M states. We suggest that in real materials with low BW and very weak disorder, the continuous nature of the transition could be probed.

(ii) Switching fields and limits of metastability: While the thermodynamic critical field can be estimated from energy balance, the upper and lower critical fields that are actually measured define limits of metastability, and have remained inaccessible within the calculations performed so far.

(iii) The effect of disorder on field melting: One expects $h_c$ to increase with reducing bandwidth, since CO is better stabilised. This indeed happens for lanthanides ($Ln$) of the form $Lnn/2Ca_{1/2}MnO_3$. For members of the $Lnn/2Sr_{1/2}MnO_3$ family [5], with very similar bandwidths, $h_c$ increases initially with decreasing BW but takes a downturn beyond a critical BW and then drops to zero, in sharp contrast to the ‘divergence’ of $h_c$ seen in the Ca
FIG. 1: Colour online: The $h-T$ phase diagram of various RE$_{1/2}$AE$_{1/2}$MnO$_3$ compounds. The materials involve a systematic decrease in $r_A$ from Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ (PSMO) to Sm$_{1/2}$Ca$_{1/2}$MnO$_3$ (SCMO). The critical CO melting temperatures increase with decreasing $r_A$. The associated hysteresis with the first order nature of transition, opens a window of metastability at low T, which tapers with increase in temperature and vanishes at $T_{CO}$. The Ca family has low disorder and shows re-entrant behaviour in $h_{co}$, which vanishes at very low bandwidths (Sm$_{1/2}$Ca$_{1/2}$MnO$_3$). Further, in the Ca family, the decrease in $r_A$, also makes the CO state more robust, with SCMO having the largest melting fields. The Sr family has larger disorder (see text) with Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ having larger $r_A$ than Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ (NSMO). While NSMO shows marked hysteresis, PSMO is rather benign.

family. We provide an explanation for this in terms of the reduced stiffness of the CO state in the presence of disorder, and consequently its melting at a lower field.

In the present work, we have used a real space Monte Carlo technique to study the melting of charge order. In our earlier work, we have addressed some of the above mentioned issues, however, this is a much detailed exposition of the phenomenon of field melting of CO in the half doped manganites. We have mapped out the $h-T$ phase diagrams capturing both hysteresis and re-entrant features as seen in experiments. In addition to thermodynamic indicators we have characterized the system at low temperature through direct spatial snapshots and by measuring the volume fraction of the CO-I and the FM-M. Using these tools we have explored the combined effects of bandwidth variation and disorder on the process of field induced melting. We have addressed all the unresolved issues, (i)-(iii), listed earlier. Further, by putting our results within a Landau-like energy landscape, we have provided a broad framework for rationalizing the observed material systematics.

The paper is organised as follows. In section II we summarize the key experimental results, and follow it in section III with a discussion of earlier theoretical work on field melting. In section IV we define our the model and describe the method for solving it. Section V discusses the zero field reference state. Section VI is the heart of the paper and discusses the results at finite field. The later sections, VII and VIII, looks at these results from alternate simpler calculations, puts forward a Landau energy landscape, and discusses the robustness of our numerical results. Section IX concludes the paper.

II. EXPERIMENTAL RESULTS

| Ca | Sr | Ba |
|----|----|----|
| 1.34 | 1.44 | 1.61 |

| La | Pr | Nd | Sm | Eu | Gd | Tb | Ho | Y |
|----|----|----|----|----|----|----|----|----|
| 1.36 | 1.29 | 1.27 | 1.24 | 1.23 | 1.21 | 1.20 | 1.18 | 1.18 |

TABLE I: Ionic radii (in Angstroms) for various AE$^{2+}$ and RE$^{3+}$ ions in the perovskite manganites.

Experimental investigations on the field melting of the CO state has probed the hysteretic melting, the bandwidth dependence of the critical melting fields and systematic effects of disorder, primarily through transport measurements and direct spatial imaging of the lattice.
undergoing the first order melting transition. We briefly review the important results here. The key experimental magnetic field-temperature ($h - T$) phase diagrams are shown in Fig.1. The material systematics were obtained by systematic decrease in $r_A$ from Pr$_{0.5}$Sr$_{0.5}$MnO$_3$ (PSMO) to Sm$_{0.5}$Ca$_{0.5}$MnO$_3$ (SCMO). The phase diagrams were obtained by sweeping up and down in magnetic fields, at fixed temperatures, on samples that have been initially zero field cooled. Fig.1 shows that the melting field in the upward sweep ($h^+$) differ the most from the ‘recovery’ field ($h^-$) in the downward sweep when $T \to 0$. This window narrows and vanishes as $T \to T_{CO}$. The evolution of $h^-$ has a ‘re-entrant’ feature that vanishes for small BW materials and is pronounced for the intermediate and low BW. Further, reducing BW (or reducing $r_A$) progressively increases the stability of the CO state, with SCMO having the largest $h^-$ and $T_{CO}$.

For the experimental results shown in Fig.1, two lanthanide families have been extensively investigated, the Ca series, La$_{0.5}$Ca$_{0.5}$MnO$_3$, and the Sr series, Ln$_{0.5}$Sr$_{0.5}$MnO$_3$. The band width is varied by making materials with Ln atoms with progressively smaller radii (see Table 1). Changing the Ln atom also changes the cation size mismatch $\sigma_A$, varying the structural disorder. The disorder arises because random substitution of Ln by Sr leads to random changes in the Mn-O-Mn bond angles, $\theta$ say, modulating the local hopping, $\propto \cos^2(\theta)$. The other effect of random substitution is that if the net charge on the dopants is different than that on the parent Ln, then charge disorder creates scattering potentials.

Traditionally, the extent of structural disorder is quantified by the variance $\sigma_A$ of the average ionic radii of the A-site in A$_{1-x}$A’$_x$BO$_3$ and has been measured for the Ca, Sr, and Ba families at various hole dopings close to half filling, $x = 0.45$. While in the Ca family $\sigma_A$ is $\sim 10^{-2}$Å$^2$, for the Sr family, greater size mismatch (Table 1) leads to $\sigma_A \sim 10^{-3}$Å$^2$. In our work, we will consider the Ca family to represent the ‘clean’ manganite, while the Sr family is typical of a disordered manganite.

In addition to BW variation, explored in detail, as for example in Fig.1, there are studies that focus on the role of disorder in field induced melting. The impact of disorder on the zero field $x = 0.5$ state has been beautifully demonstrated. Experiments at half doping where CO melts on applying a magnetic field has been carried out with a particular focus on the spatial nature of the melting process, and the role of disorder.

While in the Ca family the melting fields increase with decreasing $r_A$, in the Sr family they are suppressed on decreasing $r_A$. For the Br family, with the largest disorder among the three families, the global CO correlations are destroyed even in the zero field ground state. Further, for the former two families, the systematic change in the upper and lower critical fields with $r_A$, provides information on the evolution of the various competing phases as the bandwidth changes. The phase diagrams in Fig.1, however do not provide details of the spatial nature of the field melted state. Also the detailed thermal evolution, in particular, the high temperature phases are not explored.

Some recent experiments have begun exploring the nature of the field melted state. The high field state in half doped NSMO has been probed for spatial inhomogeneity of the magnetic order in the ground state. This study reveals that the low $T$, the high field state is rather inhomogeneous with ‘poor FM’ domains coexisting with perfect FM regions. Since resistivity data implies this state is metallic, the system appears to be a percolative metal. Similar results were reported in La$_{2-x}$Pr$_y$Ca$_{2}$MnO$_3$ where again CO-I regions are shown to coexist with FM-M regions. Further experiments track the evolution of the charge ordered state with magnetic field and temperature in LCMO, PCMO, PSMO and NSMO at $x = 0.5$. The main body of results indicate coexistence, at low $T$, of competing (AF-I/FM-M) phases. Further, it is reported that the fraction of the two phases can be tuned by following different protocols in $h - T$ variation. The tunability is ascribed to the disorder induced arrest of kinetics of first order phase transition, and is a non-equilibrium effect.

While we address the results on the ‘clean’ systems such as Ca family in this work, we contrast these results with that in the disordered systems in the companion paper.

III. EARLIER WORK

These intriguing experimental results have led to attempts at a theory of the field melting in CO manganite. (i) The earliest attempt involved the mean field study of a one band model with on-site and nearest neighbour Hubbard interaction in addition to double exchange. On application of a magnetic field the zero field AF-CO-I state was shown to melt to a FM-M through a first order transition. The result shows the existence of a magnetic order dependent CO state, which is destabilised when the AF order is disrupted by a field.

(ii) A more elaborate and realistic model incorporating Jahn-Teller interaction has also been explored. A variety of magnetically ordered states (FM, G-AF, CE-AF) along with a metallic or a CO state, were examined in the presence of a field. The destabilisation of the AF-CO state with increasing $h$ was tracked for varying BW, and an increase in the field needed for the transition with decreasing $r_A$ was captured.

(iii) Finally, a two orbital model was studied, with a large family of variational states in a recent work. This established that the smallness of the (thermodynamic) melting fields is due to the closeness in energy of the CE-CO-I and the FM-M phases. It also suggested that the field melted state could be inhomogeneous, since, since for range of electron-phonon couplings, at finite $h$, a CE-CO state with defects seems preferable to the homogeneous...
neous CE-CO-I or FM-M. We believe that the result hints at field induced phase separation but the authors did not pursue the issue further.

The attempts above have added valuable insight to the field melting problem but have left three issues unexplored: (i) the spatial character of the high field state, (ii) the impact of disorder on the melting process, and (iii) the dependence of the switching field $h_c^0$ on bandwidth and disorder. We use a real space Monte Carlo method on large lattices to settle these questions.

**IV. MODEL AND METHOD**

**A. Model**

For our calculations, we consider a two band model for $e_g$ electrons, Hunds coupled to $t_{2g}$ derived core spins, in a two dimensional square (2D) lattice. The electrons are also coupled to Jahn-Teller phonons, while the core spins have an AF superexchange coupling between them. These ingredients are all necessary to obtain a CE-CO-I phase.

$$H = \sum_{\langle ij \rangle \alpha \beta} t_{ij}^\alpha \hat{c}_{i \alpha}^\dagger \hat{c}_{j \beta} - J_H \sum_i \hat{S}_i \sigma_i + J \sum_{\langle ij \rangle} \hat{S}_i \hat{S}_j - \lambda \sum_i \hat{Q}_i \tau_i + \frac{K}{2} \sum_i \hat{Q}_i^2 - \mu N - h \sum_i S_{iz}$$

Here, $c$ and $c^\dagger$ are annihilation and creation operators for $e_g$ electrons and $\alpha$, $\beta$ are the two Mn-$e_g$ orbitals $d_{x^2-y^2}$ and $d_{3z^2-r^2}$, labelled $(a)$ and $(b)$ in what follows. $t_{ij}^\alpha$ are hopping amplitudes between nearest-neighbor sites with the symmetry dictated form: $t_{aa} = t_{bb} = t$, $t_{ab} = t_{ba} = \pm t/3$, $t_{cb} = t_{bc} = \pm t/\sqrt{3}$, $t_{cb} = t_{bc} = \pm t/\sqrt{3}$, where $x$ and $y$ are spatial directions. The $e_g$ electron spin is $\sigma_i^\mu = \sum_{\sigma \sigma'} \hat{c}_{i \alpha \sigma}^\dagger \Gamma_{\sigma \sigma'}^{\alpha \beta} \hat{c}_{i \beta \sigma}$, where the $\Gamma$‘s are Pauli matrices. It is coupled to the $t_{2g}$ spin $\hat{S}_i$ via the Hund’s coupling $J_H$, and we assume $J_H/t \gg 1$. $\lambda$ is the coupling between the JT distortion $\hat{Q}_i = (\hat{Q}_{ix}, \hat{Q}_{iz})$ and the orbital pseudospin $\tau_i^\mu = \sum_{\sigma \sigma'} c_{i \alpha \sigma}^\dagger \Gamma_{\sigma \sigma'}^{\alpha \beta} c_{i \beta \sigma}$, and $K$ is the lattice stiffness, and $h$ the magnetic field. We assume it to be in the $z$ direction and coupled only to $S_i$. We set $t = 1$, $K = 1$, and treat the $Q_i$ and $S_i$ as classical variables. The chemical potential $\mu$ is adjusted so that the electron density remains $n = 1/2$ which is also $x = 1 - n = 1/2$.

**B. Parameter space**

Even in the model without disorder, the parameter space with $J_H$, $J$ and $\lambda$ is rather large. So one looks to restrict the same. In this quest, the large Hund’s rule coupling, as seen in experiments is assumed to be large enough to completely enslave the electron spin to the core spin at any site. In the context of the model, this amounts to working in the $J_H/t \to \infty$ limit, and thus the relevant parameters that remain are the electron phonon coupling ($\lambda$) and the AF superexchange scale ($J$). In the present paper we do not consider disorder so the only electronic parameters are $\lambda$ and $J$. We choose $\lambda$ and $J$ such that the system is a CE-CO-I at low $T$ and $h = 0$, but close to a FM-M phase. Changing $\lambda$ is equivalent to change in (inverse) BW. This variation is needed to model the materials systematics with changing $r_A$. In reality $J$ also changes with changing BW but we will assume these two to be independent parameters and will explore only a couple of $J$ values, $J = 0.10, 0.12$, to narrow the parameter space. The justification of these values will be provided in the text. Disorder introduces another ‘dimension’ to the parameter space, those results are reported separately.

**C. Method**

The present work is in the spirit of many other calculations, where classical Monte Carlo technique has been employed to explore the nonequilibrium effects like hysteresis etc., around a phase transition. As mentioned earlier, this study aims at describing the ‘real’ space nature of field melting and to that end we a real space exact diagonalization (ED) based Monte Carlo (MC) that tracks the spin-charge-orbital degrees of freedom of the system across the melting transition. This technique defines a region (cluster) around the site at which one attempts a MC update and accepts and rejects the update based on the energy change only in the cluster. This ‘travelling cluster approximation’ (TCA) gives a huge gain compared to standard ED-MC the cost for which scales as $N^4$, $N$ being the size of the system. Within TCA the cost for the same system is $\sim \lambda N^3 N_C$, where $N_C$ is the fixed cluster size, and is linear in $N$ as opposed to $N^4$. Using this technique we have accessed sizes up to $40^2$ as opposed to the limit of $\sim 8^2$ within ED-MC. In the present work we employ $8^2$ moving cluster on system sizes between $16^2$ (for most data presented here) to $40^2$ (for detailed study of spatial evolution). For calculation of observables, we diagonalizes the full system, after the system has equilibrated. In studies such as this, where real space phase separation may play vital role in determining transport responses, it is pertinent to be able to access system sizes that are large enough to capture coexisting phases on the lattice. Such a study is not possible reliably within conventional ED-MC due to severe size limitations. Most results are obtained by sweeping the system in magnetic fields at low temperature. The issues of comparison between experimental and Monte Carlo sweep rates, etc., has been discussed later in the paper.
FIG. 2: Colour online: (a) The $\lambda - J$ phase diagram at $T = 0.01$. The various phases are indicated in colors separated by solid lines. The dashed/dotted lines, demarcate various parameter regimes in the CE-CO-I phase terms of their response to the applied fields. Clearly, the $\lambda, J$ window of interest lies where the FM-M and the CE-CO-I phases are close by, this happens for $\lambda \sim 1.6$ and $J \sim 0.1$. For large $\lambda$ values, above the big-dashed line, the CO state does not melt on applying a magnetic field. Below this line, the CE-CO-I state melts in response to an applied magnetic field. However, this region can be classified into one that recovers the CO state in a low $T$ field cycle (entire CE-CO-I region between the big-dashed line and finely-dashed line) and another that does not recover CO (the CE-CO-I region below the finely-dashed line). Below the dotted line the field melting yields a homogeneous FM-M and above it, the melting forms an inhomogeneous state. (b) The clean $\lambda - T$ phase diagram, at $J = 0.1$, and (c) for $J = 0.12$. Note, in panels (b) and (c) $\lambda$ increases from right to left. Since, these are thermal evolution with $\lambda$ at the fixed $J$, the AF(A-2D) region separating the FM-M and CE-CO-I phases at low T, is smaller for the $J = 0.1$ cross section as compared to that at $J = 0.12$.

D. Physical quantities

In order to study the evolution with applied magnetic field, we track various physical quantities in real space and momentum space which provide information about the correlations of (and between) the various d.o.f. of the system. These are further backed up with real space snapshots of the lattice across the melting transition. Here we briefly discuss these indicators.

We compute the ‘one point’ distribution of lattice distortions, $P(Q) = \sum_j \delta(Q - Q_j)$, where $Q_i = |Q_i|$, spatial $Q - Q$ correlations, $D_Q(q) = \sum_{ij} \langle Q_iQ_j \rangle e^{iQ \cdot (r_i - r_j)}$, and spin-spin correlations, $S(q) = \sum_{ij} \langle S_iS_j \rangle e^{iQ \cdot (r_i - r_j)}$. Angular brackets represent a thermal average. We also compute the volume fraction of the charge ordered region in the lattice from direct spatial snapshots of the charge distribution. To measure the volume fraction, we tag a site with a particular color if the site has $n > 0.5$ and is surrounded by the four nearest neighbor sites with $n < 0.5$ and vice versa (i.e. a site with local anti-ferro-charge correlation is marked with a particular color). Similarly, if the difference between the charge density at a site with its nearest neighbours is less than a threshold, that site is tagged by a different color, i.e., the charge uniform regions are marked by this color. For intermediate cases, we use an interpolative colour scheme. A measure of the volume fraction is necessary for studying inhomogeneous melting because the momentum space structure factors are not a good measure of the local CO in the system. Further, the spatial snapshots also directly provide visual information on the melting process. While the indicators above measure the correlations and spatial evolution, the metallic or insulating character is tracked via (low frequency) conductivity $\sigma_{dc}$, and the density of states (DOS), $N(\omega) = \langle \sum_n \delta(\omega - \epsilon_n) \rangle$, where $\epsilon_n$ are the electronic eigenvalues in some MC background and the angular brackets indicate thermal average. We track all the above quantities as a function of temperature and applied magnetic fields for studying the CO melting phenomenon.

V. RESULTS AT ZERO FIELD

Prior to studying the effects of magnetic fields on the CE-CO-I state, it is useful to look at the zero field situation. This will set up a reference state, give information about the phases that compete with the CE-CO-I phase and help us to justify an appropriate $\lambda, J$ parameter regime for calculations presented later. Accordingly, in this section we discuss in detail, the $\lambda - J$ phase diagram at low $T$ and $h = 0$. Further, this phase diagram with all the zero field competing phases also serves as a plot on which one can not only classify parameter regimes with varied responses to a typical magnetic field sweep, but also rationalize them to a large extent purely in terms of proximity to zero field phase boundaries.
A. $\lambda - J$ phase diagram at $T = 0$

Fig. 2. (a) shows the $\lambda - J$ phase diagram\textsuperscript{23} where the various phases obtained are indicated. These first order phase boundaries (indicated by solid lines) are determined by cooling the system in zero field. The dashed/dotted lines, obtained by low temperature field sweeps, indicate the boundaries of the regions showing different responses to field sweeps. While the detailed indicators allowing this classification (of regions) will be the subject of the next section, here we will just present the classification and rationalize it, to the extent possible, purely in terms of competing phases and proximity to phase boundaries.

Let us begin by describing the various competing phases. The CE-CO-I phase and the FM-M phases are separated by a thin region (A-2D) with ‘line-like’, $q = \{0, \pi\}$ spin correlations (Fig. 2. (a)). If we are at a parameter regime where the FM-M and CE-CO-I phases are very close, $\lambda \sim 1.6$ and $J \sim 0.1$, we can drive a CE-CO-I to FM-M transition by applying a small magnetic field. In the present work we restrict ourselves to two choices, $J = 0.10$ and $J = 0.12$. While $J = 0.1$ allows closer agreement of temperature and magnetic scales to experiments, as seen in Fig. 2. (a), it does not allow much room to explore the $\lambda$ dependence. The available $\lambda$ window ($\Delta \lambda$) is $\sim 0.1$. At the lower end one hits the AF-M phase and at the upper end (above the dashed red line) the CO-state cannot be destabilised using a magnetic field. For exploring the BW dependence in more detail we choose $J = 0.12$ allowing a window $\Delta \lambda \approx 0.2$.

We now describe the classification of the regions within the zero field CE-CO-I parameter space in terms on their responses to field sweeps and rationalize them in terms of the zero field phases. This will also allow easier organisation of the later data.

The dashed/dotted lines within the CE-CO-I phase demarcate parameter regimes with qualitatively different response to magnetic field sweep. The distinct regimes are determined by tracking the $q = (0, 0)$ component of the magnetic structure factor ($S(q)$), the $V_{\text{CO}}$ and the $q = (\pi, \pi)$ component of $D_Q(q)$ as a function of $h$.

(i) The big-dashed line shows the boundary above which the CO state does not melt however large the magnetic field. All melting phenomena are confined to the CE-CO-I regions below this boundary. Below this line there are CE-CO-I regions that either share first order boundary with FM-CO for $\lambda \sim 1.6 - 1.65$ or with FM-M/A-2D at lower $\lambda$. Melting of the CO can happen by a transition to the FM-M phase on applying a field for regions close to the FM-M. It is not expected that the region $\lambda \sim 1.6 - 1.65$ which shares boundary with FM-CO, will lose CO with applied field. However, we shall later see that at intermediate fields the system does phase separate between off half doping phases and thereby can lose CO. However in the limiting case of very large field (where all the spins are polarized), $J$ becomes irrelevant, and the half doping FM-CO is recovered back.

(ii) The finely-dashed line separates regions with different kinds of hysteretic response. Between the fine and the big-dashed lines the CO state melts with increasing $h$ and recovers when $h$ is swept back. Between the finely-dashed line and the AF(A-2D) boundary the melted CO state does not recover when the magnetic field is swept back to zero. This non-recovery suggests the existence of a metastable FM-M minimum, close by in energy to the CE-CO-I absolute minimum even when $h = 0$. Thus, when the field is swept back to zero the system can have FM-M volume fractions in the final state. Deeper into the CE-region, beyond the finely-dashed line, the FM-M minima is no longer metastable at $h = 0$ and the CE-CO-I is recovered on field reduction.

(iii) Finally, the dotted line is the boundary between inhomogeneous (or partial) melting and homogeneous melting. The system melts homogeneously below this line while undergoes inhomogeneous melting above it. The inhomogeneous melting at fields beyond forward melting field, $(h^+_c)$, cannot be rationalized in terms of the zero field half doping phases and as mentioned in (i) above, is related to phase separation tendency into off half doping phases at intermediate magnetic fields. This tendency is not just confined to regions with ‘FM-CO’ ‘CE-CO-I’ boundary as mentioned (i), but extends to lower $\lambda$ as well, where the boundary is between FM-M and CE-CO-I.

B. $\lambda - T$ phase diagrams at $J = 0.10$ and 0.12

While the $\lambda - J$ phase diagram gives valuable information on the low $T$ phases, issues like melting temperatures and the thermal evolution of these low $T$ phases requires constructing $\lambda - T$ phase diagrams. Together with the $\lambda - J$ phase diagram they complete the study of the ‘clean’ system at zero field.

Here we present two such $h = 0$ phase diagrams for $J = 0.10$ and $J = 0.12$. These will provide the $T_{\text{CO}}$ scales, an estimate of the stability of the CO phase with $\lambda$ and will later help to demonstrate sensitivity of the field melting scales on small changes in $J$. Fig. 2(b) and (c) show the clean $\lambda - T$ phase diagram, at $J = 0.1$ and $J = 0.12$ respectively. As a generic feature in both cases, for low $\lambda$, the system has a FM-M ground state, which gives way to a relatively narrow A-type AF phase and finally to a CE-CO-I state at higher $\lambda$. Owing to larger AF coupling $T_{\text{CE}}$ for the $J = 0.12$ case is larger than for the $J = 0.10$, while the $T_C$ scales are generally suppressed as compared to the $J = 0.10$ case. Starting from the CE-CO-I phase at low $T$, the system goes through a PM-CO to a PM-CD phase, with increase in temperature. The increase in $T_{\text{CO}}$ with $\lambda$ signals that beyond a certain $\lambda$, the CO state can be stabilized without CE magnetic order and that the stability of the CO state grows with increasing $\lambda$. However, at very large $\lambda$, the electron-phonon coupling would strongly localise electrons (onsite) and inter-site correlations would be weak, suppressing $T_{\text{CO}}$ scales.
FIG. 3: Colour online: Distinct response to field cycling as a function of $\lambda$. Top panel shows $V_{CO}$ and $S_q(0,0)$ and bottom panel shows the corresponding resistivity as a function of the applied magnetic field. The $\lambda/t$ values for the systems are indicated and $J_{AF} = 0.12$. All results are obtained at $T=0.02$. In the top panel, with increasing $\lambda$, the forward switching fields $h^+_C$ increase and for $\lambda = 1.7$, the CO does not melt, the corresponding resistivities, shown a concurrent switching with the abrupt decrease in the $V_{CO}$ and vice versa. Hysteresis is seen in all four cases, except the hysteresis for $\lambda = 1.7$ occurs only in the magnetic sector, while it occurs for $V_{CO}$, $S_{fm}$ and $\rho$, for the first three cases. Also note that with increasing $\lambda$, the melted state has successively large amounts of residual CO, which increases to unity for $\lambda = 1.7$. Further, at intermediate $\lambda \sim 1.45$, the system does not recover the CO state when the magnetic field is cycled back to zero, signalling that the FM-M is metastable even at zero field. Note, for the $\lambda = 1.4$ case, because of the structure of the AF(A-2D) phase in Fig.2(a) one has to go slightly deep into the CE-CO phase, where at zero field the CO is recovered in field cycle and hence the FM-M is unstable at $h = 0$.

We do not explore that large $\lambda$ regime. We shall later use the trends and the numbers in these phase diagrams to make detailed comparison with experiments.

Having discussed the low T, zero field phase diagrams, their thermal evolution and organisation of the parameter space in terms of the different response to field sweeps, we turn to the main theme of the paper which is to describe extensively the phenomenon field induced melting of the CO state.

VI. RESULTS AT FINITE FIELD

For studying the magnetic field effects we track the various indicators described in Sec.IV.D, by first cooling the system at $h = 0$ and then cycling the magnetic field.

This section is organized as follows. Sub-sections A and B aim at a systematic study of the evolution of the field response with $\lambda$. While in Sub. A we contrast the melting vs non melting responses, and through indicators such as DOS and P(Q) (that measures the Q-Q correlations on the lattice) hint towards an inhomogeneous melted state, in Sub. B, we discuss the complete evolution with $\lambda$. In Sub. C we present our $h-T$ phase diagrams, explicitly show real space data on inhomogeneous melting in Sub. D and in Sub. E compare our results with experiments.

A. Result of a typical field sweep

This and the next subsections revolve around Fig. 3. This illustrates the field response for four $\lambda, J$ combinations, with the CO growing stronger as we move from (a)-(d). We show the CO volume fraction, $V_{CO}$ and the $q = (0,0)$ spin structure factor in the top panels, and the corresponding resistivity $\rho(T,h)$ in the bottom panels.

In this subsection we will focus on contrasting between the melting vs non melting response. Let us start by illustrating two CO states (i) at $\lambda = 1.55$ where the CO ‘melts’ is response to increasing $h$, and (ii) $\lambda = 1.70$, where it does not. These are the last two columns in Fig.3. These two parameter points lie on opposite sides of $\lambda \sim 1.65$ in the $\lambda-J$ phase diagram (Fig. 2(a)). Above $\lambda \sim 1.65$, the FM-CO state does not melt with any applied magnetic field, i.e the charge order is not dependent on CE magnetic order for its survival. For lower $\lambda$ values the CO melts. In both these cases the applied field transforms the CE order to FM. For the $\lambda = 1.55$, there is a sharp reduction in the CO volume fraction $V_{CO}$, while for $\lambda = 1.70$ the $V_{CO}$ does not change (there is, however, a decrease in the charge disproportionation). Fig.3(g)-(h) show $\rho(h)$ at low T. For $\lambda = 1.55$ there is a large drop in resistivity associated with the magnetic transition (shown in dashed red lines). The $V_{CO}$, although drops sharply, nevertheless has a finite value. This state
independent of the magnetic field: the high field state is percolative. The data in panel Fig.4(a)-(b) shows that the CO state doesnot melt with magnetic field. For comparison, in the same panel Fig.4(b), shows a broad hump signalling a finite regions of lattice distortions, and the DOS (\(\omega\)) vs \(\lambda\) at \(h/t = 0\) and \(h/t = 1\) respectively. (c)-(d), \(N(\omega)\) vs \(\omega - \mu\) at \(h/t = 0\) and \(h/t = 0.1\) respectively. In (b), the dashed line is the \(P(Q)\) for \(\lambda = 1\), where the ground state is a uniform metal.

with a finite \(V_{\text{CO}}\) and a ‘low’ resistivity is likely to be a percolative metal. However, this by itself does not guarantee that this is the correct ground state, as low T field sweeps with multiple competing states can easily get trapped into non equilibrium states. While here we take this as an indication of possible inhomogeneous melting, we carefully separate out cases of equilibrium coexistence from nonequilibrium coexistence in later sections before making any such claims. Contrast this with the system at \(\lambda = 1.7\) where the resistivity continues to be very large and the \(V_{\text{CO}}\) stays unity even at large h.

For these two cases we now look at distribution \(P(Q)\) of lattice distortions, and the DOS (\(N(\omega)\)) for possible insight into the melted state. Fig.4.(a)-(b) show \(P(Q)\) at \(h = 0\) and \(h = 0.10\) respectively. For \(\lambda = 1.55\), at \(h = 0\), \(P(Q)\) shows a bimodal distribution. The two values (peaks in \(P(Q)\)) of distortion on the lattice signifies a possible checkerboard CO ground state. This state melts into a metal at \(h \sim 0.07\), see Fig.3.(g). The \(P(Q)\) at \(h = 0.1\), Fig.4(b), shows a broad hump signalling a finite regions with localized charges. For comparison, in the same panel we have shown data corresponding to \(\lambda = 1.0\) (the dashed line), where the ground state is a uniform metal and the \(P(Q)\) is distinctly peaked close to zero. For \(\lambda = 1.7\), the data in panel Fig.4(a)-(b) shows that \(P(Q)\) is virtually independent of the magnetic field: the high field state is a ferromagnetic charge ordered polaronic insulator.

Fig.4(c)-(d) show the DOS for these \(\lambda\) and \(h\) combinations. These correlate well with the \(P(Q)\), with the field generating a finite DOS at the Fermi level for \(\lambda = 1.55\), while there is only a gap reduction (but no closure) for \(\lambda = 1.7\). The gap reduction is related to the increase in effective BW in the CO phase as the magnetic order changes from CE to FM, removing the magnetic blocking of electron hopping.

To sum up: (i) we clearly see two different kinds of response to the applied field for systems with different \(\lambda\) (or BW), and (ii) the \(V_{\text{CO}}\) and \(P(Q)\) results are indicative of inhomogeneous melting at intermediate \(\lambda\) values. In the next subsection we address the \(\lambda\) dependence more systematically.

**B. The distinct response regimes**

We now take up Fig.3(a)-(b) and their resistivities shown in Fig 3(c)-(f), and discuss how they connect with the remaining plots in Fig.3 that we discussed in the last sub-section. This will provide the generic nature of response to field sweeping with changing \(\lambda\) or BW.

Fig 3(a) & (e) are at \(J \sim 0.12\) to avoid hitting the FM-M phase (see Fig.2(a)). All the rest are at \(J = 0.12\). (i) The data at low \(\lambda = 1.40\), is shown to illustrate homogeneous melting of the CO state as opposed to inhomogeneous melting at higher \(\lambda\). As Fig.3.(a) shows, the \(V_{\text{CO}}\) reduces to less than 10% for \(h/t > 0.09\). but the CO state recovers on reducing \(h\). Note that recovery of the CO happens for this ‘low’ \(\lambda\) system even though it does not for \(\lambda \sim 1.45\) Fig. 3(b). This is so because as seen from Fig. 2(a), at \(\lambda \sim 1.4\) the A-2D phase is much wider essentially rendering the FM-M unstable at \(h = 0\). (ii) For \(\lambda = 1.45\), the A-2D region is small and the FM-M remains metastable at \(h = 0\) and the CO state is not recovered even when the field is swept back to zero. Further also in Fig.3(b), the system retains \(\sim 25\%\) of CO volume beyond \(h^c\). (iii) In Fig.3(c) the system retains \(\sim 30\%\) of CO volume beyond \(h^c\), and recovers the CE-CO-I state on field reduction. (iv) At even higher \(\lambda \sim 1.7\), as discussed earlier, the system retains a global CO at all \(h\). Thus, switching field \(h_c\) essentially diverges.

Let us summarize our findings here. The increase in the melting fields with \(\lambda\) and systematics of recovery/non recovery of the CO with \(\lambda\) bear out what was discussed in Sec V.A, i.e., if the system is (close to) far from the phase boundary (Fig.2(a)), it would (not) be able to recover the CO state. And finally with increase in \(\lambda\) from 1.4 to 1.65, there is an observed monotonic growth in the residual CO volume fraction in the field melted state, signalling towards inhomogeneous melting. Also if one takes into account the resistivity shown in the lower panel, for a certain \(\lambda\) regime, it would imply that the system is a percolative metal.

While this is indicative of the fact that by appropriately varying BW one can tune between inhomogeneous and homogeneous melting, we would caution the reader that this does not amount to saying that the melting
The intermediate issue. Alternate calculations using different protocols to settle this elaborate upon this in section VII, where we perform all states leading to nonequilibrium coexistence. We shall the system can get stuck partly in different metastable states at intermediate fields) from spurious ones, where one needs to carefully disentangle the real inhomogeneous tendencies. This numerical issue needs addressing itself lead to trapping and complicate genuine phase separation. Wide domain of metastability of competing phases can is inhomogeneous for this entire $\lambda(1.4 - 1.65)$ window.}

is the CO correlations grow stronger with $\lambda$. At low $T$, both at low $\lambda(= 1.4)$ and high $\lambda(= 1.7)$, the intermediate field state (beyond the magnetic transition) are uniform states, while for $\lambda = 1.4$ it is a FM-M, for $\lambda = 1.7$ it is FM-CO. The intermediate $\lambda(\sim 1.5 - 1.6)$, the system phase separates beyond $h^C_\lambda$, into constituents indicated (in italics). Further, the hysteresis window for $\lambda = 1.5$ extends all the way up to zero fields, implying non recovery of the CO state, increasing $\lambda$ to $\sim 1.6$, recovers the CO state. The detailed finite $T$ evolution is considered in the text.

**C. The $h-T$ phase diagrams**

This subsection complements the previous two, in that here we provide the thermal evolution of the low $T$ phases by constructing the $h-T$ phase diagram. This also allows for a comparison of trends between theory and the available experimental data in the low $\sigma_A$ (Ca based) manganites as shown in Fig. 1. We will make qualitative comparisons in this subsection that will be supported by spatial map for the real space evolution of the melting process, in Sub. D, and make quantitative comparisons are made in Sub. E.

Fig. 5 shows the $h-T$ phase diagrams obtained at $J = 0.12$ and $\lambda$ values indicated. These phase diagrams are obtained by sweeping in magnetic fields at various temperatures, by first cooling to the relevant temperature in zero field. Note, here the definitions of $h^C_\lambda$ ($h^-_\lambda$), used to denote the boundary of hysteresis, is taken to be the field at which there is a sharp increase (drop) in the FM structure factor. This is so chosen because, while for Fig.5(a)-(c) these is a CO to CD transition, in 5.(d) there is no CO melting and the magnetic transition is a good indicator of the switching fields.

**The low temperature phases:** The evolution of the low $T$ phases with increasing $\lambda$ are exactly the same as discussed in Sub.A and Sub.B. The increase in the thermodynamic melting fields with increasing $\lambda$, the homogeneous melting and subsequent recovery at low $\lambda \sim 1.4$, the inhomogeneous melting at larger $\lambda$ values along with the (non recovery) recovery at intermediate $\lambda \sim 1.45 - 1.54$ $\lambda \sim 1.55 - 1.65$ and the refusal to melting for $\lambda > 1.65$ are all seen in the $h-T$ phase diagrams at the representative $\lambda$ values in Fig. 5.

**Thermal evolution at low fields:** An increase in temperature allows the system to access larger fluctuations and thus the hysteresis window narrows with increasing temperature. This window tapers off and closes at $T_{CO}$, with the $T_{CO}$’s themselves growing progressively with increasing $\lambda$, as is also seen from Fig. 2(c). The increase in $T_{CO}$ is due to progressive increase in the stability of the CO state with increasing $\lambda$. This increase in stability of the CO state also manifests itself in the intermediate temperature physics, where at low $\lambda \sim 1.4$ the loss of the CE pattern drives the system metallic, with no residual CO correlations. With increasing $\lambda$ however, the systems have CO correlations surviving at progressively higher temperatures although the long range $(q = (\pi, \pi))$ correlations get suppressed. The accompanying magnetic state at intermediate temperatures is a line like AF phase with $q = (0, \pi)$ and $q = (\pi, 0)$ correlations. This can be looked at as a precursor to the low temperature CE phase, where the $q = (\pi/2, \pi/2)$ correlations (crucial to the CE phase) do not form at intermediate temperatures. Further increasing the temperature essentially
makes the system lose all spin-spin correlations and the systems end up in a PM phase. This is accompanied, at smaller $\lambda \sim 1.4 - 1.5$ values, by a metallic state, and at higher $\lambda \sim 1.6 - 1.7$, by a evolution into a metal only after going through a CO state. The thermal PM-CO to PM-M transition, for this higher $\lambda$ range, signals thermal liberation of carriers trapped in polaronic state. This clearly brings out the stability of CO order at larger $\lambda$ quite independent of the spin order.

**Thermal evolution at large fields:** Let us contrast the low field evolution with that at large finite fields. We will later join the two regimes. The expectation that, at fields beyond $h^*_5$, the system will lose all A-2D/CE correlations, is borne out well for small and large $\lambda$ cases. As discussed, while in the small $\lambda$ ($\sim 1.4$) case the CO is lost completely at high fields, in the large $\lambda$ ($\sim 1.7$) case where the CO does not melt, the systems evolve into a FM-CO phase. Increasing temperature will convert the FM-CO to a FM-M with thermal liberation of polaronic entrapments. To the temperatures accessed, the high magnetic field is strong enough to resist any further thermal transition to a PM state. In contrast to the simple evolution at very small (a) and very large (d) $\lambda$, the intermediate $\lambda$ situation (b), (c) is complicated by phase separation tendencies. These systems at fields beyond $h^*_7$, phase separate into phases that are at electron densities off half doping. The constituents vary with changing $\lambda$, in that, at $\lambda \sim 1.5$, the coexisting phases are ‘FM-M ($n_1$) + AF-CD ($n_2$)’ and those at $\lambda \sim 1.6$ are ‘FM-CO ($n_1$)+AF-CD($n_2$)’. The densities indicated in the brackets imply off half doping densities, typical values are given in Sec. VII. A. This phase separation, is the origin of inhomogeneous melting and renders the melting to be a continuous transition. The way this tuning can happen is discussed in Section VII. These regions are indicated, in Fig. 5 (b) and (c), by light colored checkerboard regions and the phases are indicated in italics. For both these $\lambda$’s the corresponding phase separated states evolve into a $n=0.5$ FM-M. (n=0.5). If one were to consider the true limit of large enough field where all the spins were fully polarized, then for $\lambda \leq 1.6$, the system would be a $n=0.5$ FM-M and above that it would be a $n=0.5$ FM-CO (which at high temperature will evolve into an FM-M). We relate the intermediate PS to the uniform asymptotic limit in Sec. VII.

**Intermediate field thermal evolution:** For all the cases shown here, the intermediate field shows a transition from the respective CE/A-2D/PM to FM state, albeit, with the hysteresis region in between, for $T < T_{CO}$. While for the low $\lambda \sim 1.4$ and large $\lambda \sim 1.7$ cases, the transitions are benign, for intermediate $\lambda \sim 1.5$ there is a transition at large $T$ and small $h$, between a PM-M and the phase separated state and between the PM-CO and the corresponding phase separated state for $\lambda \sim 1.6$.

To sum up, the $h-T$ phase diagrams are governed by the following principles: (i) The hysteretic nature of the field induced transitions, (ii) the increased stability of CO correlations with increasing $\lambda$, (iii) The ensuing decoupling of the spin and charge sectors leading to independent ordering with increasing $\lambda$ and finally (iv) the phase separation tendencies into off half doping phases at fields above $h^*_7$. This of course leaves us to clarify the fate of the theoretical spin polarized limit (of $h \to \infty$), where the phase is either CO or metallic, as will be done later in the paper.

To make a few qualitative comparisons with experimental results shown in Fig. 1, we note that, as for the Ca-family, decreasing $r_A$ or equivalently increasing $\lambda$ increases the CO melting fields and temperatures, as Further, the detailed temperature evolution of low $T$ phases.
The primary steps in the evolution are (i) nucleation of the FM-M within the CE-CO-I at low fields (as seen in the second and third columns from left) (ii) the sharp percolation of the FM-M within the CE-CO-I at low fields (as seen in the fifth column) and (iv) the subsequent recovery (the rest of the columns). While, the nucleation, typical to such transitions, is expected, it is interesting to note that the CE phase apart from giving way to FM patches, also breaks up into regions of opposite ‘handed’ stairs. Since, there is no preferred handedness, both being degenerate, small destabilisation of the CE phase results into the system creating domains of opposite handed stairs. Within each domain the CO state survives, however, its stability is locally reduced at these magnetic domain boundaries. This reduction is small and is not apparent in columns two and three, where the system started out in the CO state at $h = 0$, however is seen in the last two panels, where the system attempts to recover global CO from the melted state. Clearly some of the domain boundaries between in the CO regions coincide with that of the magnetic domains boundaries of opposite handed CE regions. This fact also makes it rather difficult to recover the CO completely on such a large systems. In column four, the system is seen to exist in a coexistent state with FM-M, AF-CD and FM-CO coexisting simultaneously. This certainly shows explicitly the inhomogeneous nature of melting arising out of the phase separation. The column five is for $h = 0.2$ which is large enough to polarize all the spins. This is deliberately shown to bring out the effects of low lying metastable states that can affect the numerics. For this value of $\lambda$ the spin polarized problem has a global FM-M ground state. However as seen, we find a coexisting FM-M and FM-CO. This happens because of large domain of metastability of the FM-CO phase, in which the system partially gets trapped. This effects the results even in column four and one needs an alternate calculation to extract the actual constituents of the equilibrum phase separated state. This is achieved by doing $\mu - n$ calculations, which is the subject of the next section. With hindsight, we mention here, that the correct constituents of the phase separation at this $\lambda$ are AF-CD and FM-M, both of which are individually at electronic densities that are off half doping, as was mentioned in subsection C.

Let us conclude this subsection by briefly summarizing what happens in such low $T$ field sweeps at other $\lambda$ values. For a system with $\lambda \sim 1.6$, the overall evolution would essentially be the same, expect that the phase separation is between an AF-CD and an FM-CO, both of which are again off half doping phases. For even larger $\lambda$ values ($> 1.65$), the system directly goes from a $(n = 0.5)$ CE-CO-I state to a $(n = 0.5)$ FM-CO state. Thus generally, for a certain $\lambda, J$ window, the systems undergo inhomogeneous melting at intermediate applied magnetic fields, where they split into regions of phases of different electron densities. This phase coexistent state, can be a percolative metal if one of the constituents is an FM-M and it manages to percolate.
E. Field melting scales

| $\lambda$ | $J$ | $T_{CO}^N(K)$ | $h_{CO}^N(K)$ | $h_{CE}^N(K)$ | $\frac{h_{CE}^N+h_{CO}^N}{N}$ |
|-----------|-----|----------------|----------------|---------------|------------------|
| 1.60      | 0.12| 200            | 170            | 65            | 102.5            |
| 1.64      | 0.12| 230            | 200            | 65            | 132.5            |
| 1.60      | 0.10| 150            | 80             | 0             | 40               |
| 1.62      | 0.10| 165            | 90             | 20            | 55               |

We end this section by making a quantitative comparison of various melting temperatures and melting fields with those found in the experiments. The above table gives the numerical and corresponding values in Kelvin of $T_{CO}$, $(h_{CO}^N)$, for $J/t = 0.1$ and $J/t = 0.12$ for similar $\lambda$ values. The numerical $T_{CO}$ values are converted to Kelvin, using the kinetic energy scale, $t \sim 0.2eV$ \cite{19} and crude finite 2D to 3D scaling of $3/2$. The magnetic fields are converted to Kelvin using $g\mu_B h/t \sim h$, again with $t = 0.2eV$. We find that for $J/t = 0.1$, the melting field is much smaller than the corresponding melting temperatures. Further, from Fig.1, as a typical low disorder example, $P_{0.5\sigma_{0.5}M_{t0.3}}$, has $T_{CO} = 250K$, while $h_{CO}^N \sim 27K$ and $h_{CO}^N \sim 17K$, giving the mean $h_{CO}^N = 22K$. Thus the smallness of the melting fields is better achieved if one tunes parameters close to $J/t = 0.1$ as compared to $J/t = 0.12$. The reason for this is seen in Fig 2(a), where, at $J/t = 0.1$, the CE-CO-I, and FM-M phases are close by, while for $J/t = 0.12$, the two phases are separated by a bigger region of A-type AF phase. Fig.7(a) and (b) show the difference of the ground state energies (per site) between the CE-CO-I and A-type AF as a function of $\lambda/t$, from the FM-M phase, which is taken to be the reference phase. $\delta E$ is defined as $E_{\text{AF-M}} - E_{\text{FM-M}}$, $N$ being the system size. It is clearly seen that the case with $J = 0.1$ has the CE-CO-I ground state energy closer to the FM-M ground state energy, while the minimum possible difference between the two for $J/t = 0.12$, is $\sim 0.03$. Crudely, this barrier to be overcome by the applied field at low $T$. So the smallest melting field achievable for $J/t = 0.12$ would be $\sim 42T$, for $\lambda/t \sim 1.4$. Where as in principle if one tunes to $J$ close to 0.1 such that the A-type AF region vanishes one can get zero melting fields. However, we do not pursue the direction of fine tuning parameters and focus on studying the melting phenomenon and the corresponding $\lambda/t$ systematics. We have chosen to work at $J \sim 0.12$ which has a larger window of $\lambda/t$ dependence for this work as discussed in Section V-A.

VII. $\mu - n$ CALCULATIONS, NATURE OF TRANSITION & AN ORGANIZING FRAMEWORK

While the above results on inhomogeneous melting are promising, as we saw, a system can get stuck, at low temperatures, in states with wide domain of metastability, especially if there are multiple competing phases close in energy, as in the present case. Thus there is a need to clearly separate genuine inhomogeneous melting from any spurious one. This issue is addressed in the following subsection by performing $n - \mu$ calculations. This will be an alternate way to conclude about phase separation at intermediate fields. Once having established the genuineness of the inhomogeneous melting, we will provide a Landau energy landscape as an organizing framework for the entire field melting systematics with changing BW.

A. Field induced phase separation

We compute the 'chemical potential'-electron density' ($\mu - n$) curves at various fixed magnetic fields and $\lambda$ values. The $\mu - n$ curves are obtained from low temperature $\mu$ scans of the system, at fixed applied magnetic fields, in a protocol that does not retain the memory of previous $\mu$ steps during the $\mu$ sweeps. While runs without memory avoid hysteresis effects, the grand canonical calculation allows the system to choose the 'best' possible $n$, thereby allowing the system to get into the correct phase at any given field. Moreover, we ensured that the system has annealed well enough by checking that our results hold up to large number (8000) of Monte Carlo steps at each $\mu$. We also cross checked these results with those obtained from a different protocol of coming down in temperature and fixed magnetic fields, for a finely discritized set of $\mu$ values. This ensures that the results are well annealed and free from low temperature Monte Carlo problems.

As seen in Fig. 8 (a), for CE-CO-I systems close to the FM-M phase, $\lambda \sim 1.5$, the CO is quickly lost (beyond $h \sim 0.02$). At a slightly higher field, the system prefers an FM-M state with $n = 0.57$ up to a certain $\mu$. 

![Image](https://via.placeholder.com/150.png?text=Figure+8)
and then directly go to an ('A-type' AF)-CD phase at $n = 0.44$ leaving the $n = 0.5$ to be an unstable density. Thus, the fixed $n(= 0.5)$ problem is phase separated and infact a percolative metal. The constituents of the PS are the FM-M and the AF-CD phases. This is true for all systems with $\lambda \sim 1.45 - 1.6$, at intermediate fields. The situation is different for larger $\lambda \sim 1.6 - 1.65$. For a typical case, $\lambda \sim 1.6$ (Fig. 8 (b)), the system prefers either a AF-CD at $n = 0.52$ up to a certain $\mu$ and then an AF at $n = 0.44$ at intermediate fields. Such a system if forced to be at $n = 0.5$, as was done previously, will phase separate into the above constituents again leading to an inhomogeneous melted state. At larger fields, both in Fig. 8 (a) and (b), the $n = 0.5$ state becomes stable, recovering the correct asymptotic limits of FM-M for $\lambda = 1.5$ and AF-CD for larger $\lambda \sim 1.6$. Thus apart from ratifying the earlier conclusion of inhomogeneous melting, this calculation helps identify the real components of the phase separation. Thus, if we look at the spatial snapshots for $h = 0.08$ and $h = 0.2$ in the field increasing run in Fig.6, we find that the existence of the FM-CO in the ground state (for a $\lambda = 1.55$ system) was purely due to it having a wide domain of metastability. This fact is not a surprise because, at least at large fields, where the $J$ becomes negligible, the FM-M and FM-CO share a first order boundary, as seen for small $J$ in Fig.2(a) around $\lambda = 1.6$. For the same reason FM-M fractions are trapped in the large field FM-CO ground state, for $\lambda \sim 1.6 - 1.65$. In the fixed density problem, for $\lambda < 1.6$, we explicitly checked that annealing the PS system (with FM-M AF-CD and FM-CO) to higher temperature first makes the FM-CO component disappear rapidly, showing that it is purely due to trapping in metastable FM-CO state. With this background we now discuss the nature of the melting phenomena and its relation to the sweep rate in the Monte Carlo and in the experiments.

**B. Nature of field induced transition & sweep rate dependence**

From the $\mu - n$ calculations, it is apparent that the melting for a window of couplings is inhomogeneous. While for low $\lambda \sim 1.4$ and high $\lambda > 1.65$, the respective transitions are abrupt, for the intermediate $\lambda(\sim 1.45 - 1.65)$, the melting is inhomogeneous and the transition is continuous. Here we discuss a schematic of such a transition. We use the ferromagnetic structure factor as the order parameter for this discussion. For intermediate coupling the magnetic phase separation is between an FM and AF states. From their densities one can work out the volume fractions of the two constituent magnetic phases. Fig.9(a) shows, the magnetization with increasing applied field. The dashed line shows the notional abrupt (first order) transition. The continuous line depicts the actual transition whereby the magnetization grows continuously (from CE-type AF state) with increasing magnetic field to a FM state. The hysteresis occurs in the background of this unusual equilibrium physics. Since the ‘switching’ in hysteresis depends on the sweep rate let us clarify the experimental and simulation timescales.

The *local* relaxation time $\tau_{loc}$ in electronic systems is $\sim 10^{-12}$ seconds, but collective relaxation times $\tau_{coll}$, say, can be macroscopic, $\sim 100$ seconds in the CO manganite. This measurement was at $\sim 0.9T_{CO}$ and $\tau_{coll}$ is likely to be much greater at low $T$. The field cycling periods $\tau_{per}$ that we could infer from field melting experiments were $\sim 10ms$. Overall $\tau_{loc} \ll \tau_{per} \ll \tau_{coll}$. Our MC results are broadly in the same window. The ‘microscopic’ timescale is the MC step. The sweep periods were $10^3 - 10^4$ MC steps (bigger in smaller systems) but still $\ll 10^{12}$ that one would need to avoid trapping in a metastable state.

The sweep rate dependence of the switching is illustrated schematically in Fig.9, for an intermediate coupling system. The left panel, (a), is for a quasistatic sweep, $\tau_{per} \gg \tau_{coll}$. In this case there would be only pro-
gressive melting and no hysteresis, the system is always in equilibrium. Panel (b) illustrates the regime $\tau_{\text{per}} \sim \tau_{\text{coll}}$, where the sweep rate is still ‘slow’ but the system cannot quite track the equilibrium state. In this case there could be successive switching. This regime is also out of computational reach for the system sizes we use. Panel (c) is for our regime $\tau_{\text{loc}} \ll \tau_{\text{per}} \ll \tau_{\text{coll}}$. The system switches at $h^*_c$ on field increase, but not necessarily to the underlying equilibrium state. The magnetization, $V_{\text{CO}}$, etc, are determined by the presence of metastable states. For $h \gg h^*_c$, where the equilibrium state is a homogeneous FM (at this $\lambda$) the low temperature system can still remain trapped in the metastable state. Finally, (d) is for an ultrafast sweep, $\tau_{\text{per}} \sim \tau_{\text{loc}}$, where the system is unable to respond at all to the changing field.

As shown in panel (c), for sweep rates typical in the experiments and in our calculation, the high field state is influenced by the equilibrium PS and nearby metastable phases. So, in any low $T$ field sweep for $\lambda = 1.55$ (say), the observed state is a mixture of the equilibrium AF-M + FM-M combination and a metastable FM-CO. Increasing $h$ converts the AF-M to FM-M but the metastable FM-CO fraction (also seen in Fig. 6, at $h = 0.2$) can be removed only by thermal annealing. At larger coupling, $\lambda \geq 1.6$, the high field low $T$ state would have stable (equilibrium) FM charge order.

From the previous sections we know that the melting can either be homogeneous or inhomogeneous. For $\lambda$ less than 1.6, the system can melt the CO simply by lowering the energy of the FM-M minima with increase in the applied field. The increase in field can either lead to a simple first order transition, as happens for low $\lambda \sim 1.4$ or as happens for intermediate $\lambda \sim 1.5$, lead to a field induced phase separation. In either case the loss of CO volume fraction is guaranteed. However for $\lambda \sim 1.6 - 1.65$, the FM-CO is the one closest in energy to the CE-CO-I (and also the true ground state in the limit of $h \rightarrow \infty$) and without the intermediate field phase separation, would have simple gone over to the FM-CO phase, as happens for $\lambda > 1.65$. Thus the phase separation is crucial for the destabilisation of the CO for this $\lambda$ window.

With this general understanding, let us discuss the Landau landscape shown in Fig. 10. This has three panels depicting the underlying free energy landcapes with increasing magnetic fields for three successively increasing values of $\lambda$.

**Small $\lambda$ response:** Panel (a) of Fig. 10 corresponds to $\lambda \sim 1.5$, where the CO state melts beyond a critical field but does not recover when the field is swept back. In the energy landscape, the $h = 0$ landscape has CE-CO-I as the global minima, however the FM-M is metastable. This metastable FM-M would be necessary to explain the non recovery of the CE-CO-I state when the field is swept back to zero, as some part of the system remains stuck in the metastable state. At $h = h_1$ and up to $h_2$, the FM-M minima lowers as expected with increasing field. If the $\lambda$ is very small $\sim 1.4$, this continues leading to a first order transition to a homogeneous FM-M. However, if $\lambda \sim 1.5$, at $h_2$, the system phase separates into off half doping phase (AF-CD + FM-M) phases, these two minima are depicted in panel (a) at $h_2$. On further increasing the field the system evolves to the asymptotic

**C. Landau landscape for melting**

Over the earlier sections, we made a number of conclusions regarding the BW or $\lambda$ dependence of the response to magnetic field sweeps. By invoking a Landau like landscape, with a relevant number of competing phases, we organize the broad systematics of the field response with changing BW within a single framework.

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**FIG. 10:** Colour online: The low temperature Landau free energy landscape with the magnetic field axis going into the plane of the paper and the other axis, $\Phi$, being the general order parameter axis. The three panels (a), (b) and (c) are representative of $\lambda = 1.5$, $\lambda = 1.6$ and $\lambda > 1.65$ respectively. All the phases are at electron density $n = 0.5$, except the ones for which a density are shown in brackets along with the name of the phase. The intervening A-type AF region is not shown to avoid cluttering.
large field ground state of a $n = 0.5$ FM-M. The phase that is closest in energy to this is the FM-CO, as is seen in Fig.2 (a) at low J and $\lambda \sim 1.5$. Given that the system tends to get trapped in the FM-CO, we depict this state to be metastable at large fields.

**Intermediate $\lambda$ response:** Panel (b) shows a similar landscape for $\lambda \sim 1.6 - 1.65$, range. Here there are a few important differences in contrast to that in (a): (i) From Fig. 2 (a) the FM-CO phase is the closest in energy to the CE-CO-I phase, that can be accessed by a magnetic field. (ii) Since we know that the CE-CO phase is recovered when the field is swept back, in the $h = 0$ landscape the FM-CO has to be unstable, as opposed to the FM-M being metastable at $h = 0$ in (a). (iii) the phase separation at intermediate fields is between FM-CO and AF-CD as depicted, which are again not half doping phases. (iv) Finally at large field values the FM-CO minima is the global minima with the FM-M minima being metastable, as is seen from Fig. 2 (a) at low J.

**Large $\lambda$ response:** This is shown in panel (c). Just like the very small $\lambda$ systems, the very large $\lambda$ systems have a rather benign evolution. As in (b), the phase closest in energy to the CE-CO state is the FM-CO state and since the CE-CO state is recovered back when the field is swept back to zero, this FM-CO state should be unstable at $h = 0$. With increasing field, the FM-CO would gradually lower and finally replace the CE-CO as the global minima. At large fields (not shown) the CE-CO would become unstable. Note here the CO does not melt and in this view, if the intermediate field induced phase separation did not occur for $\lambda \sim 1.6 - 1.65$, CO melting would not have been possible.

To sum up, we have qualitatively described the evolution of the free energy landscape with magnetic field and BW variations. Before concluding, we briefly discuss some general numerical issues and the large field asymptote.

**VIII. ASYMPTOTIC LIMIT & NUMERICAL CHECKS**

**A. The large $h$ limit**

All through, we have maintained that the large field state is rather difficult to capture in the field sweep protocol. The large field or spin polarized limit would yield the critical $\lambda$ below which the system is a FM-M and above which it is a FM-CO. Since this limit is spin polarized, we are effectively at the $J = 0$ axis of Fig. 2(a). There are two equivalent ways of determining the critical $\lambda$. One is setting $J = 0$ and annealing the system at zero field from high to low temperature. The other is to freeze all the classical spins to a fixed direction and then to anneal the orbital/lattice variables. Fig. 11 (a) presents the $\lambda - T$ phase diagram of the later approach. The parameter space, at low temperature, is divided into low $\lambda < 1.6$ metallic phase and large $\lambda \geq 1.6$ CO-OO phase. The $T_{CO}$ increases with increasing $\lambda/t$ as is expected. Let us now look at the electronic charge disproportionation $\delta_n$ as a function of $\lambda/t$. This is calculated by averaging the modulus of the difference of the onsite charge densities from $n = 0.5$ over all sites ($\delta_n = \frac{1}{N} \sum_{i=1}^{N} |0.5 - n_i|$). This although is a bulk measurement, judging by the trend of monotonic growth in $\delta_n$ in Fig. 11 (b), it is clear that even for $\lambda/t < 1.6$, there is some residual local CO regions. We have checked this explicitly by looking at snapshots. While above $\lambda = 1.6$, the system has global CO, the residual CO in the lower $\lambda$ case are due to trapping in metastable state. Thus, although we have cooled the system and there is no low temperature sweep involved, still there is some amount of phase mixing due wide domain of metastability of the FM-CO. Thus it is bound to affect the low T field sweeps in the earlier calculations. We briefly discuss some general numerical issues below regarding stability of the results on annealing time and mention some general numerical ways to ascertain if a phase coexistent state is an equilibrium state or a result of a system remaining stuck partially in a metastable state.

**B. Numerical checks**

(i) Although TCA gains much in accessing large system sizes, even with TCA performing MC runs with $10^5$ sweeps on large systems at every temperature is pretty demanding computationally on large systems. So to ensure that we have annealed well enough, we repeated a few calculations on smaller systems employing standard exact diagonalization-Monte-Carlo. This was particularly done at parameter points where the system phase separated in the TCA calculations, we presented the same state (obtained using the same protocol) to very large annealing ($\sim 40000$ MC relaxation steps) on these small systems. The phase separation remained stable even after such long runs.
(ii) One way to ascertain if the coexistent state (as is our intermediate field phase separated states) is the equilibrium ground state would be by diagonalizing the system in the background of the each of the competing phases separately and comparing their energy with that of the coexistent state. Still another way would be to do alternate calculation which would make the coexistent phases separately and comparing their energy with that of the coexistent state. We have dealt with the 'clean' case, we have mapped the $\mu - n$ calculation showing the phase separation tendency at intermediate fields which unambiguously pointed out the coexistent state naturally plausible. In our case the $\mu - n$ calculation made extensive comparison between the thermal and the magnetic CO melting scales of our calculations and experiments. Finally, from our numerical results we have established the way in which the free energy landscape would evolve with increasing magnetic fields and changing $\lambda$ (or $r_A$). We take up the disorder effects on the CO melting in the companion paper.[11]

IX. CONCLUSIONS

We have reported the first controlled results on the field melting of charge order in half doped manganites using an unbiased Monte Carlo method. In this paper we have dealt with the 'clean' case, we have mapped put the $h - T$ phase diagram exhibiting both hysteresis and re-entrant features. We found the melting to be inhomogeneous at intermediate magnetic fields and this stands as a testable predictions in this regard. We have made extensive comparison between the thermal and the magnetic CO melting scales of our calculations and experiments. Finally, from our numerical results we have established the way in which the free energy landscape would evolve with increasing magnetic fields and changing $\lambda$ (or $r_A$). We take up the disorder effects on the CO melting in the companion paper.[11]

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