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## Correlated percolation and disordered Ising magnetism in doped double perovskite --Manuscript Draft--

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Abstract:	<p>Double perovskites are compounds with chemical formula <math>A_2BB'O_6</math> where B and B' ions each form face-centered cubic (fcc) lattices. Recent experiments have studied doped double perovskite <math>Ba_2Na_{1-x}Ca_xOsO_6</math> where <math>0 \leq x \leq 1</math>. For <math>x = 1</math>, the octupolar magnetism of osmium <math>Os^{6+}</math> on the fcc lattice is well described by an Ising model. To understand this system for dopings <math>x &lt; 1</math>, we construct a model where, starting from <math>x = 1</math>, each doped <math>Na^+</math> on a random B site totally suppresses the osmium Ising magnetism on its six neighboring B' sites. This leads to a correlated percolation problem on the fcc lattice of osmium ions. Using Monte Carlo simulations, we compute the percolation threshold <math>x_c \approx 0.7255</math> below which osmium <math>Os^{6+}</math> magnetism does not percolate. Based on system sizes we have studied, the fractal dimension of the critical spanning cluster in this correlated percolation model is found to be <math>df \approx 2.37</math>. We also use Monte Carlo simulations to compute the configuration averaged specific heat and transition temperature <math>T_c</math> of the Ising model on the largest cluster for <math>x &gt; x_c</math>, and compare our results on <math>T_c</math> versus doping with experiments.</p>
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# Correlated percolation and disordered Ising magnetism in doped double perovskites

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Double perovskites are compounds with chemical formula  $A_2BB'O_6$  where  $B$  and  $B'$  ions each form face-centered cubic (fcc) lattices. Recent experiments have studied doped double perovskite  $\text{Ba}_2\text{Na}_{1-x}\text{Ca}_x\text{OsO}_6$  where  $0 \leq x \leq 1$ . For  $x = 1$ , the octupolar magnetism of osmium  $\text{Os}^{6+}$  on the fcc lattice is well described by an Ising model. To understand this system for dopings  $x < 1$ , we construct a model where, starting from  $x = 1$ , each doped  $\text{Na}^+$  on a random  $B$  site totally suppresses the osmium Ising magnetism on its six neighboring  $B'$  sites. This leads to a *correlated* percolation problem on the fcc lattice of osmium ions. Using Monte Carlo simulations, we compute the percolation threshold  $x_c \approx 0.7255$  below which osmium  $\text{Os}^{6+}$  magnetism does not percolate. Based on system sizes we have studied, the fractal dimension of the critical spanning cluster in this correlated percolation model is found to be  $d_f \approx 2.37$ . We also use Monte Carlo simulations to compute the configuration averaged specific heat and transition temperature  $T_c$  of the Ising model on the largest cluster for  $x > x_c$ , and compare our results on  $T_c$  versus doping with experiments.

## I. INTRODUCTION

Double perovskite materials, with the general formula  $A_2BB'O_6$ , are of great interest due to the possibility of substituting ions at the  $A$ ,  $B$ , and  $B'$  sites in order to realize a large class compounds exhibiting a wide range of magnetic, spintronic, and optical properties [1–10]. Halide-based double perovskites have also been proposed as important materials for photovoltaic applications [11–14]. Due to a wide range of achievable  $d$ -orbital electron fillings, and the interplay of spin-orbit coupling, interactions, and lattice deformations for heavy elements [15–17], double perovskite materials also provide unusual examples of  $d$ -orbital multipolar magnetism. Quadrupolar magnetism has been reported in the  $d^1$  double perovskites where it is accompanied by lattice distortions [18–27] while Ising type octupolar magnetism has been proposed to explain the observed time-reversal breaking without any dipole moment observed in  $d^2$  double perovskites with osmium  $\text{Os}^{6+}$  on the fcc lattice [28–36].

Recent experiments and theories have explored the doped double perovskites  $\text{Ba}_2\text{Na}_{1-x}\text{Ca}_x\text{OsO}_6$  [25, 37]. Substituting a  $\text{Ca}^{2+}$  ion with  $\text{Na}^+$  ions results in six neighboring  $\text{Os}^{6+}$  ions transitioning to  $\text{Os}^{6+\varepsilon}$ . Each  $\text{Na}^+$  dopant also leads to a strong local strain field which completely suppresses the Ising octupolar magnetism on the six neighboring osmium sites [36]. In this paper, we use Monte Carlo simulations to explore the unusual percolation model which arises from the correlated impact of dopants on the osmium octupoles, and study the octupolar Ising model on the percolation cluster. The main results we obtain are: (i) the percolation threshold  $x_c \approx 0.7255(10)$  (i.e.,  $\text{Na}^+$  doping 0.2745(10)) for the correlated percolation model on the fcc lattice, (ii) the fractal dimension of the percolation cluster  $d_f \approx 2.37$  based on system sizes we have studied, and (iii) the doping dependence of the specific heat for the percolating cluster, and the octupolar transition temperature  $T_c$  which we compare with muon spin rotation ( $\mu\text{SR}$ ) data [25].

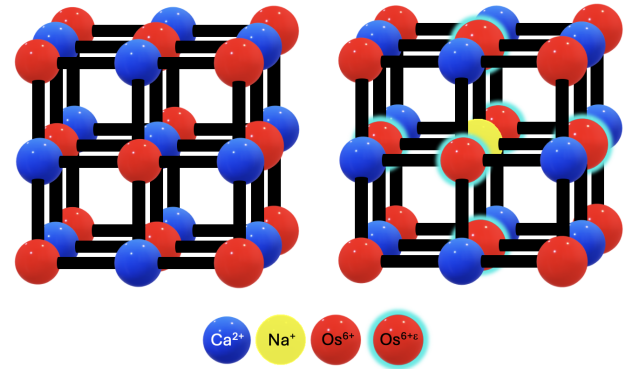


FIG. 1. Left: Ordered double perovskite structure of  $\text{Ba}_2\text{CaOsO}_6$  consisting of a three-dimensional checkerboard pattern of  $\text{OsO}_6$  (red) and  $\text{CaO}_6$  (blue) octahedra with the osmium ions exhibiting Ising octupolar magnetism. Ba and oxygen ions are not explicitly depicted. Right: Replacing  $\text{Ca}^{2+}$  (blue) by  $\text{Na}^+$  dopant (yellow) leads to local charge doping as well as strong local strain fields which completely suppress the octupole moment on the six highlighted neighboring osmium sites defining a correlated percolation problem. A nonzero octupolar  $T_c$  is only present for sufficiently small  $\text{Na}^+$  concentration ( $1 - x$ ) which can support the percolation of Ising magnetic octupoles on the active osmium sites.

## II. CORRELATED PERCOLATION MODEL

We define the correlated percolation model as follows. We start at  $x = 1$  with the ideal cubic lattice having a checkerboard pattern of  $\text{Ca}^{2+}$  and  $\text{Os}^{6+}$  ions as depicted in Fig. 1(a). We assume periodic boundary conditions (PBCs), and define the linear dimension of the system as  $L$ , so there are  $L^3/2$  ions of each type. At  $x = 1$ , this structure has the  $\text{Os}^{6+}$  ions forming an ideal fcc lattice and it exhibits Ising octupolar magnetic order. We next assume that each  $\text{Ca}^{2+}$  ion, when substituted by  $\text{Na}^+$ , suppresses the octupolar magnetism on its six neighboring osmium sites as depicted by the highlighted sites in

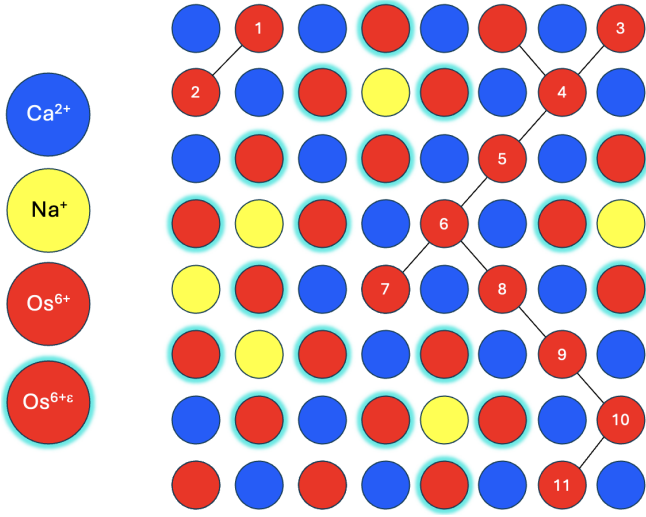


FIG. 2. Depth first search (DFS) traversal order in a 2D slice through the fcc lattice, where we start from an arbitrary  $\text{Os}^{6+}$  site on one face of the lattice and visit adjacent  $\text{Os}^{6+}$  sites to reach the opposite face. Differently colored spheres represent  $\text{Ca}^{2+}$  (blue) and doped  $\text{Na}^+$  (yellow) ions, and different osmium sites which are (i) surrounded by  $\text{Ca}^{2+}$  (red) or (ii) neighboring to a  $\text{Na}^+$  site (red highlighted with blue). Nodes have been numbered in the order in which they are visited, and we have applied PBCs. The algorithm has the following steps: (i) start at a vertex of the graph (ii) mark the current vertex as visited (iii) iterate through the adjacent vertices (iv) if an adjacent vertex has been visited, it is ignored (v) if an adjacent vertex has not been visited, recursively perform DFS on this vertex. The DFS algorithm explores a path as far as possible before backtracking and exploring other paths; for instance, the search pauses at the node-7, backtracks to node-6, and proceeds down a different branch via node-8.

Fig. 1(b). As we keep adding  $\text{Na}^+$  dopants, if we find a site where osmium magnetism is already suppressed, we assume that it remains suppressed. This leads to two types of sites at any concentration  $\delta = (1 - x)$  of  $\text{Na}^+$  dopants: those sites which are not neighbors of any dopant remain active in octupolar Ising magnetism, while a site which is a neighbor of any  $\text{Na}^+$  dopant effectively gets removed from the fcc lattice. We will use Monte Carlo simulations to realize various random configurations of  $\text{Na}^+$  for each  $x$ , and find the value of  $x$  at which the octupolar  $\text{Os}^{6+}$  ions stop percolating across the osmium fcc lattice.

### A. Percolation threshold for correlated percolation

To determine the percolation threshold for this correlated percolation problem on the fcc lattice, we vary the probability of doping from  $x = 1$  to  $x = 0$  (equivalently  $\text{Na}^+$  concentration from  $\delta = 0$  to  $\delta = 1$ ) in increments of 0.01. We generate 20000 random instances for the dopant configuration at each value of  $x$ . For each instance, we use the Depth First Search (DFS) algorithm [38] to check

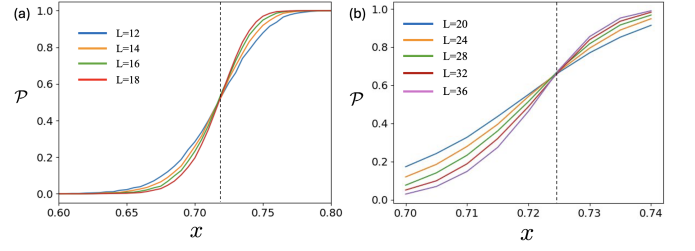


FIG. 3. Probability of percolation  $\mathcal{P}$  as a function of  $x$  for linear system sizes (a)  $L = 12, 14, 16, 18$  and larger sizes (b)  $L = 20, 24, 28, 32, 36$ . The intersection point of these graphs reveals the percolation threshold of  $x_c \approx 0.717$  on smaller sizes and  $x_c \approx 0.725$  on larger system sizes. Our best estimate using system sizes upto  $L = 52$  yields  $x_c = 0.7255(10)$ .

whether the configuration of  $\text{Os}^{6+}$  ions percolates across the lattice, and thus compute the fraction  $\mathcal{P}$  of all configurations which percolate. The DFS traversal order in a graph is shown in Fig. 2. DFS is appropriate for the percolation problem, as it efficiently traverses between pairs of opposite faces in the lattice. Fig. 3(a) and Fig. 3(b) show plots of  $\mathcal{P}$  as a function of  $x$  for a set of system sizes  $L = 12-18$  and larger sizes  $L = 20-36$ . We identify the intersection point of these various curves as the percolation threshold  $x_c$ . On smaller system sizes, we find  $x_c \approx 0.717$ , while on larger system sizes the threshold moves to  $x_c \approx 0.725$ . Going to larger system sizes upto  $L = 52$ , we determine  $x_c = 0.7255(10)$  at which the probability  $\mathcal{P}(x_c)$  becomes nearly system size independent corresponding to a crossing of the  $\mathcal{P}(x)$  curves. This corresponds to a critical  $\text{Na}^+$  dopant concentration  $\delta_c = 1 - x_c = 0.2745(10)$  beyond which connected  $\text{Os}^{6+}$  sites on the fcc lattice no longer percolate, so long-range octupolar Ising order can no longer exist.

We can contrast this with two other percolation thresholds we can estimate for uncorrelated percolation on the fcc lattice. If each dopant only knocks out a single  $\text{Os}^{6+}$  ion in its vicinity, the percolation problem reduces to the conventional site percolation model on the fcc lattice for which it is well known that  $x_c \approx 0.1992$  [39–41]. This means a large doping concentration  $\delta_c \approx 0.8008$   $\text{Na}^+$  would be needed to kill octupolar magnetism. If, on the other hand, we assume each  $\text{Na}^+$  dopant removes six *randomly located*  $\text{Os}^{6+}$  ions on the fcc lattice, an admittedly artificial model, then  $\delta_c \approx 0.1335$  so a much smaller doping would be needed to kill octupolar order than in our correlated percolation model.

### B. Fractal dimension of percolation cluster

Consider the number  $N_{\text{con}}$  of  $\text{Os}^{6+}$  sites on the largest connected cluster. In the percolating phase, we expect  $N_{\text{con}} \sim L^3$ , while in the non-percolating phase we expect  $N_{\text{con}} \sim L^0$ . At the percolation threshold  $x_c$ , the size of the spanning cluster is expected to be a fractal, so the number of points on the cluster will scale as  $N_{\text{con}} \sim L^{d_f}$ ,

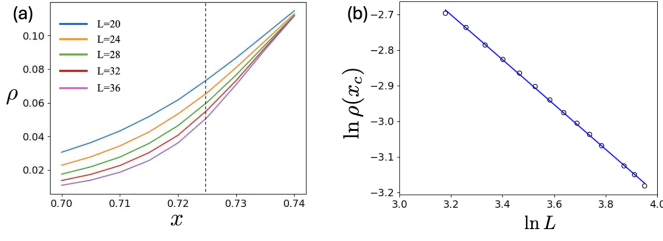


FIG. 4. (a) Cluster density  $\rho$ , defined as the largest connected cluster size  $N_{\text{con}}$  divided by total system volume  $L^3$ , plotted versus  $x$ ; dashed line indicates the percolation threshold  $x_c$ . (b) Log-log plot of the cluster density at the percolation threshold  $x_c = 0.7255$  versus the linear system size  $L$  for system sizes upto  $L = 52$ . From the slope of the linear fit we find the fractal dimension  $d_f \approx 2.37$ .

where  $d_f$  is the fractal dimension. Fig. 4(a) plots the system size dependence of the cluster density defined as  $\rho = N_{\text{con}}/L^3$ , where we expect at criticality  $\rho(x_c) \sim L^{d_f-3}$ . To extract  $d_f$ , we plot  $\ln \rho(x_c)$  versus  $\ln L$  in Fig. 4(b), where we expect  $\ln \rho(x_c) = (d_f - 3) \ln L + \text{constant}$ . A straight fit to the data on system sizes  $L = 24, 28, \dots, 52$  yields  $d_f \approx 2.37$ . Based on universality for percolation in the presence of short-range correlations, we might expect a larger  $d_f \approx 2.52$  [39, 42–44]; it is possible that the universal value will be recovered if we work on much larger system sizes. At the same time, it is important to note that it has been suggested that the fractal dimension of the percolation cluster in certain models may be non-universal [45].

### III. ISING MODEL ON PERCOLATING CLUSTERS

Finally, we turn to a study of the Ising model on the largest percolating cluster for  $x_c < x \leq 1$ . In the Ising model, spins are assigned values  $s_i = \pm 1$  corresponding to the two octupolar states related by time-reversal symmetry. The Ising model Hamiltonian is given by

$$H = -J \sum_{\langle ij \rangle} s_i s_j \quad (1)$$

where  $J > 0$  is the interaction energy (octupolar exchange energy) which favors a state where all  $\{s_i\} = +1$  or all  $\{s_i\} = -1$ . For  $\text{Ba}_2\text{CaOsO}_6$ , it has been estimated that  $J \approx 1 \text{ meV}$  [32], while quantum terms which could lead to dynamics are much weaker and can be ignored to leading approximation. The sites  $i, j$  are neighboring sites on the fcc lattice which form part of the connected percolating cluster and which have not been impacted by having a neighboring  $\text{Na}^+$  dopant. To calculate the properties such as the Ising phase transition, we focus on only this percolating cluster of  $\text{Os}^{6+}$  spins, which can display a thermal phase transition to long-range magnetic order.

We begin by creating random disorder configurations of the dopants and identifying the largest connected clus-

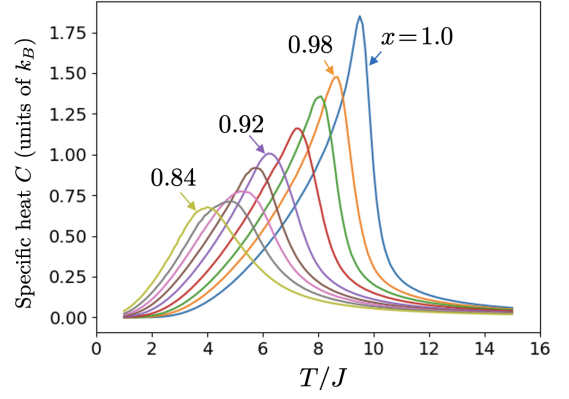


FIG. 5. Specific heat  $C$  per spin as a function of normalized temperature  $T/J$  for the Ising model on the largest percolating cluster for fixed lattice size  $L = 12$  and varying concentrations  $x = 1, 0.98, \dots, 0.84$ . The peak position of  $C$  yields the octupolar transition temperature  $T_c$ .

ter on the fcc lattice at each  $x$ . Using the Metropolis algorithm, we then perform Monte Carlo simulations of the Ising spin model on this connected cluster, fixing  $J = 1$  which is equivalent to measuring temperature in units of  $J$ . At each temperature, we carry out simulations using a single spin flip Metropolis algorithm, where we consider a spin flip and calculate the resulting energy change  $\Delta E$ . The flip is always accepted if it lowers the energy ( $\Delta E < 0$ ). If it increases the energy, the flip is accepted with probability  $e^{-\Delta E/T}$  (we set  $k_B = 1$ ). We use  $10^4$  sweeps of the percolating cluster to bring the spin configurations to thermal equilibrium, and study temperatures from  $T = 15$  down to  $T = 0.1$  in steps of 0.1. We then measure the energy over  $10^5$  sweeps of the percolating cluster, and use this to compute the specific heat per spin as a function of temperature. Finally, we average our results over 20 disorder realizations.

#### A. Specific Heat

To calculate the specific heat per spin,  $C$ , we use the formula

$$C = \frac{\langle E^2 \rangle - \langle E \rangle^2}{N_c T^2} \quad (2)$$

where  $E$  is the total energy,  $N_c$  is the number of spins in the cluster, and  $T$  is the temperature. The averages  $\langle \cdot \rangle$  correspond to Metropolis configuration averages for a fixed disorder realization. Fig. 5 shows  $C$  averaged over 20 disorder realizations plotted as a function of temperature, for different dopant concentrations which correspond to  $x = 1.00, 0.98, \dots, 0.84$  in steps of 0.02. For  $x = 1$ , we find a peak in  $C$  from which we identify a phase transition into octupolar magnetic order at  $T/J \approx 9.5$ . With increasing  $x$ , we find that the peak in  $C$  moves to lower temperature, and the value of  $C$  also



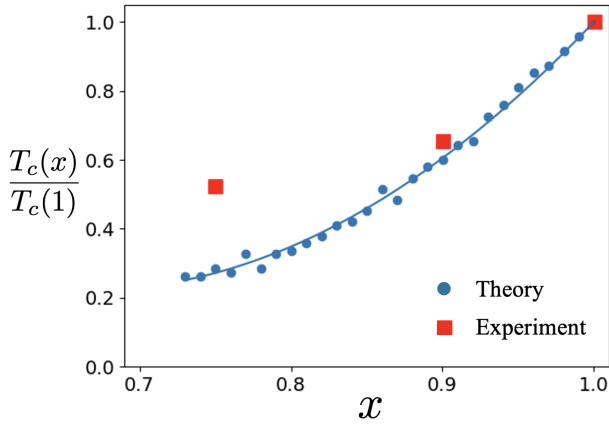


FIG. 6. Octupolar transition temperature  $T_c(x)$  at various concentrations  $x$  scaled to its value at  $x = 1$  (blue dots). Solid line is shown as a guide to the eye. We compare our results with  $T_c$  obtained using  $\mu$ SR experiments (red squares) as reported in Ref. [25].

decreases. We have checked, for consistency, that the integral  $\int dT \frac{C}{T} \approx \ln 2$  over the shown temperature range for all values of  $x$ .

### B. Octupolar transition temperature versus $x$

We finally turn to the octupolar transition temperature  $T_c$  which we identify using the peak position of the specific heat. Fig. 6 shows the Monte Carlo result for  $T_c$  as blue dots which we connect with a line as a guide to the eye. To permit a comparison of our results with experimental data, we normalize the value of  $T_c$  with its value in the clean system at  $x = 1$ . The normalized experimental data are shown as red squares in Fig. 6.

## IV. CONCLUSION

In this paper, we proposed and studied a correlated percolation problem on the face-centred cubic lattice, and an Ising model on percolating clusters, in order to explain the octupolar magnetism of the doped double perovskite  $\text{Ba}_2\text{Na}_{1-x}\text{Ca}_x\text{OsO}_6$ . We computed the percolation threshold and fractal dimension using Monte Carlo simulations of the percolation model over a wide range of lattice sizes. Our critical temperature values from Monte Carlo simulations based on the Metropolis algorithm provide a good description of the experimental result for  $T_c$  for small Na doping, but this agreement breaks down for large Na doping at  $x \leq 0.75$ . This breakdown might possibly arise from the fact that we have retained the  $\text{Os}^{6+}$  sites in our model, but dropped  $\text{Os}^{6+\varepsilon}\text{-Os}^{6+}$  and  $\text{Os}^{6+\varepsilon}\text{-Os}^{6+\varepsilon}$  interactions which might become more important at higher Na doping. Our work calls for additional experiments in the doping range  $0.75 < x < 1$  to test for the range of validity of our model and when precisely it breaks down. Additional theoretical modelling is necessary to understand the magnetism of these disordered double perovskites  $\text{Ba}_2\text{Na}_{1-x}\text{Ca}_x\text{OsO}_6$  for all dopings  $0 < x < 1$ .

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**Author contributions:** The author wrote all the Python simulation codes, carried out all the simulations, and created the figures and plots for all the results presented in this paper.

**Code availability:** The Python Monte Carlo simulation codes for the percolation model and Ising model, as well as the codes and data for generating the figures in this paper are available on GitHub (<https://github.com/Tanmay337442/percolationmagnet>).

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**Declaration of interests**

☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: