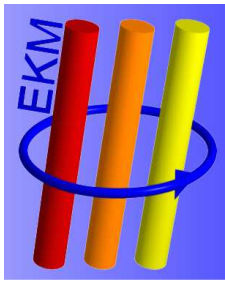
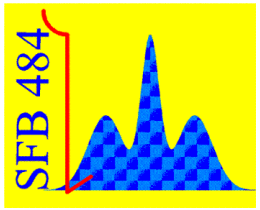


Center for
Electronic Correlations and Magnetism
University of Augsburg

Towards a global phase diagram for perovskite manganites

Sanjeev Kumar, A.P.K.,

and P. Majumdar (Allahabad)

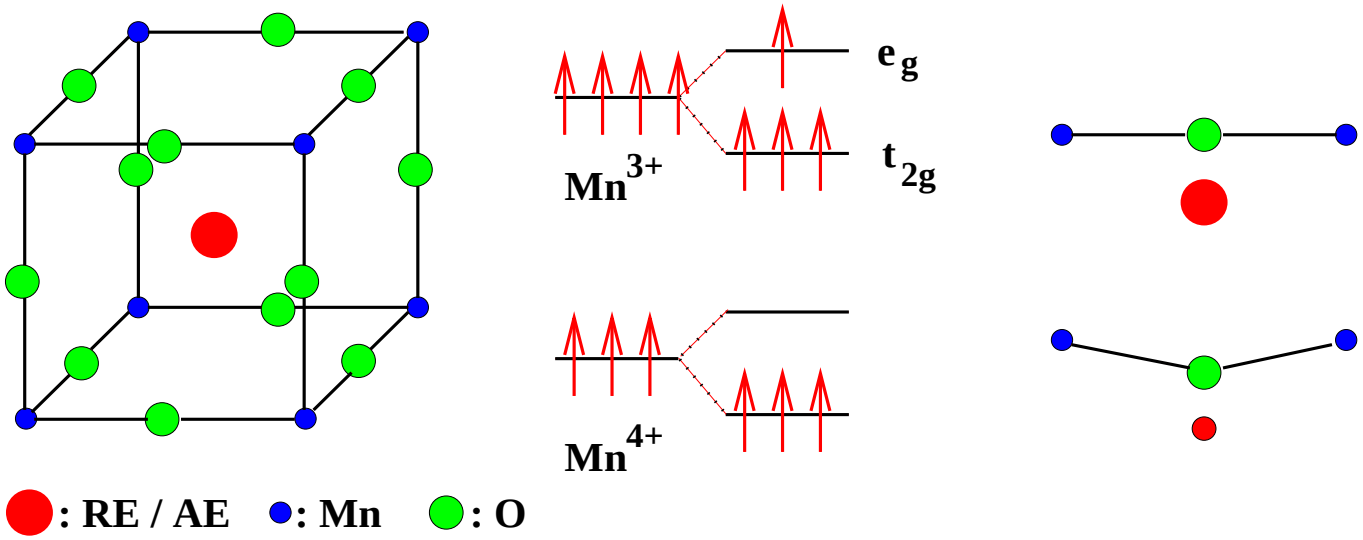


Sonderforschungsbereich 484
Kooperative Phänomene im Festkörper: Metall-Isolator-Übergänge und Ordnung mikroskopischer Freiheitsgrade

Outline:

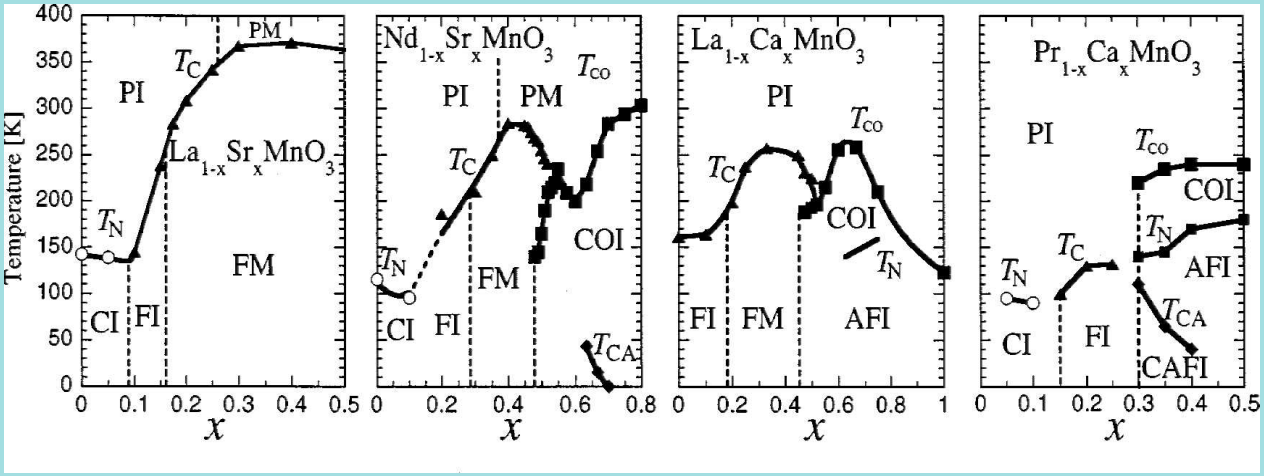
- perovskite manganites:
  crystal structure, electronic configurations
- recent experiments:
  controlling disorder and bandwidth
- model for manganites:
  2 bands, Jahn-Teller and Hund's rule coupling
- traveling cluster approximation:
  exact diagonalisation and Monte Carlo
- results:
  phase diagrams and transport
- summary

Perovskite structure & electronic configurations



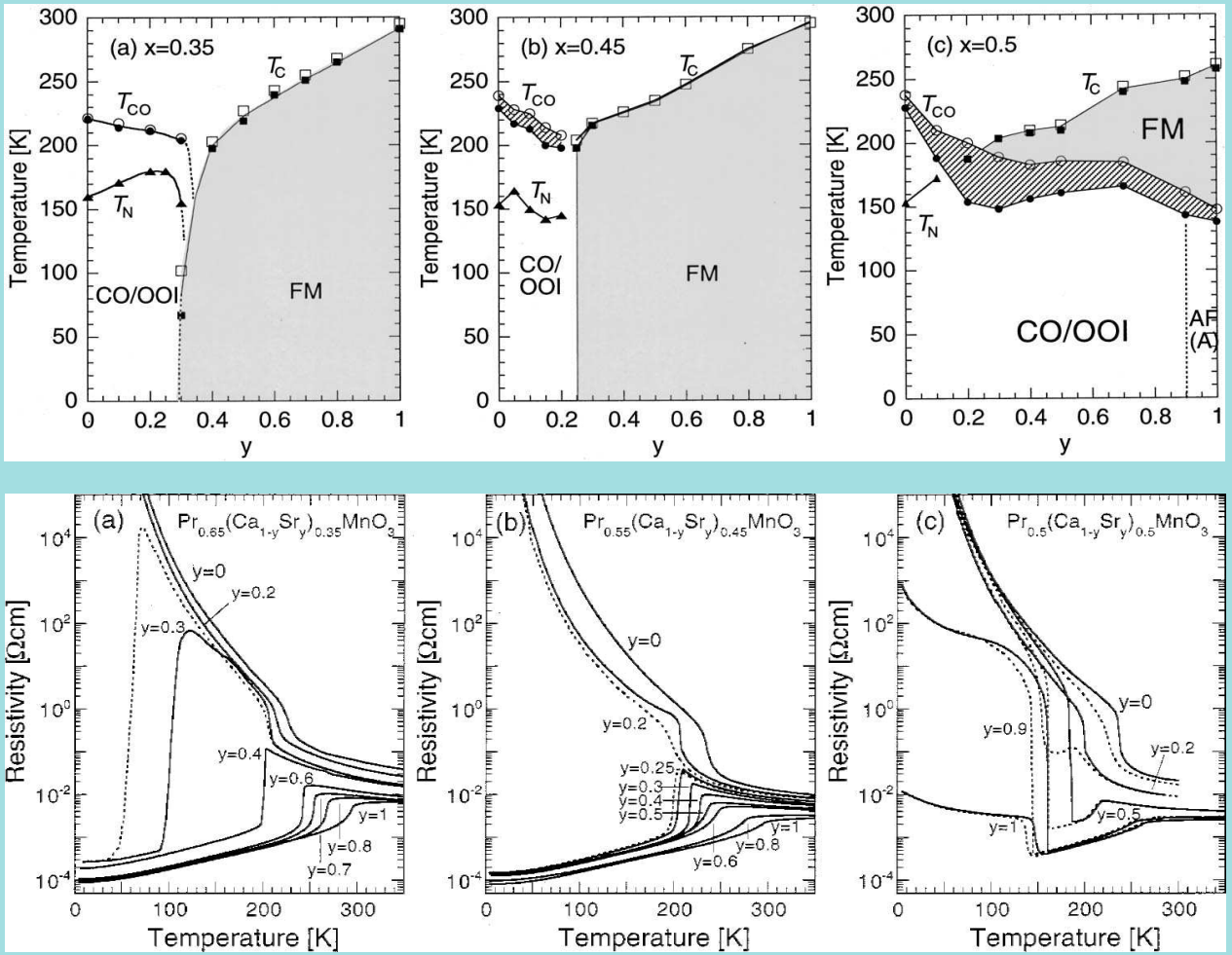
- parent compounds (e.g. $LaMnO_3$)
  A-type AFM, Mott insulator, distorted octahedra
- doped manganites (e.g. $La_{1-x}Ca_xMnO_3$)
  - RE^{3+} replaced by AE^{2+} $\longrightarrow$ hole doping
  - $CaMnO_3$: G-type AFM, band insulator

T-x Phase Diagrams. Imada *et al.* (1998)



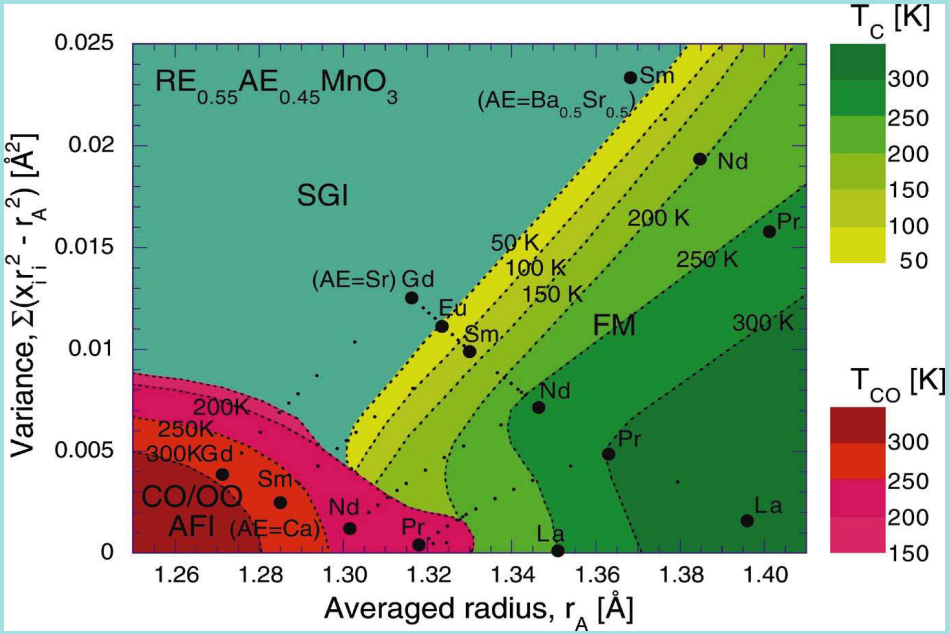
- **Large bandwidth: FM-Metal with large T_c**
- **Intermediate bandwidth: reduction in T_c, metal insulator transition, colossal magnetoresistence(CMR).**
- **Small bandwidth: Only insulating phases.**

Phase Diagrams for $Pr_{1-x}(Ca_{1-y}Sr_y)_xMnO_3$.



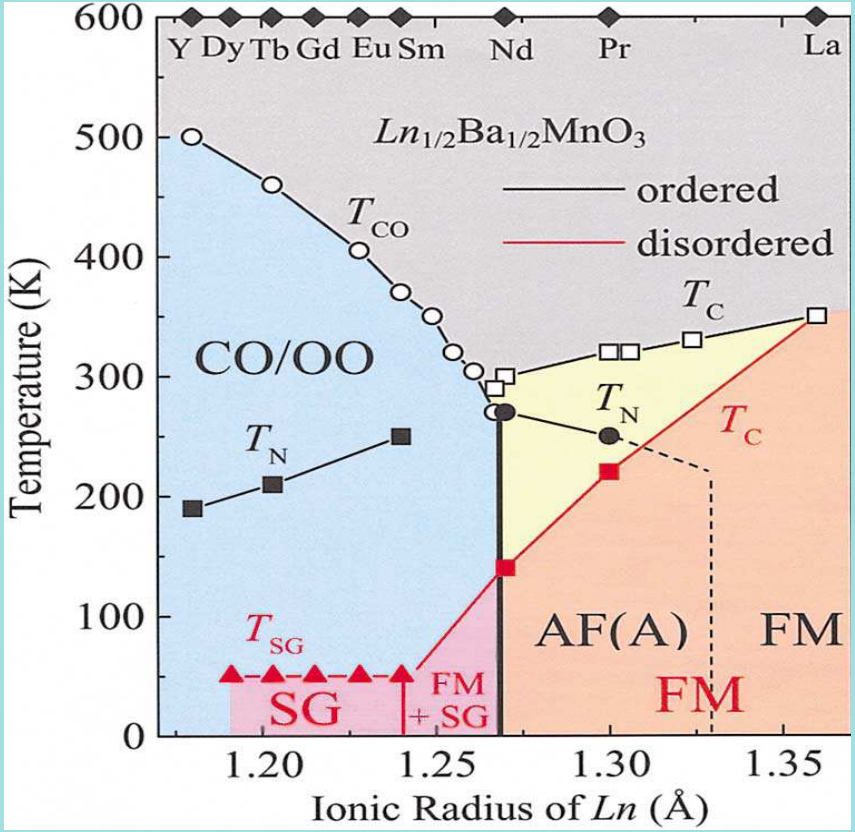
Tomioka *et al.* **(2002)**

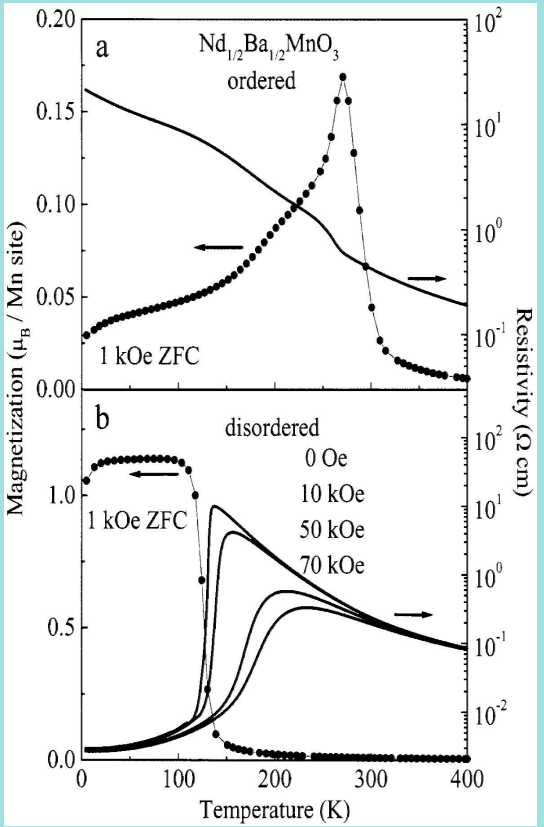
Global Phase Diagram. Tomioka *et al.* (2004)



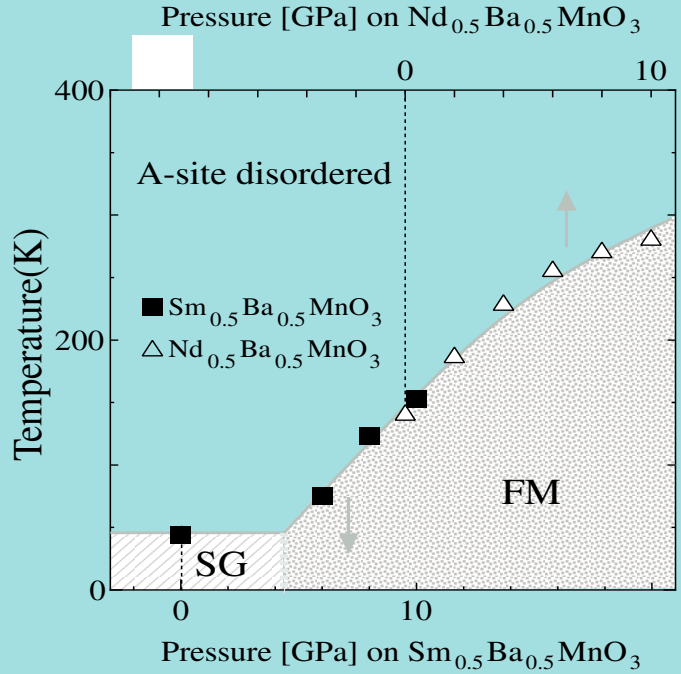
- **universal parameters: Average cation size(r_A) and variance in cation size (σ^2).**
- **more than 50 different manganites follow a universal trend in terms of r_A and σ^2.**

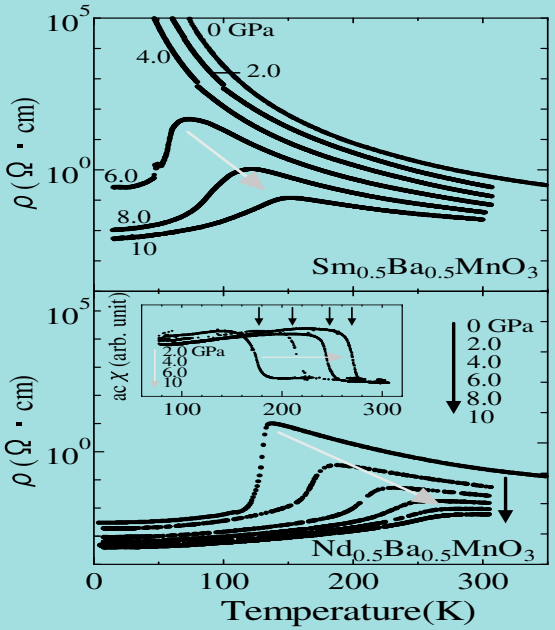
Order vs. Disorder. Akahoshi *et al.* (2003)





Bandwidth controlled by external pressure.





Takeshita *et al.* (2004)

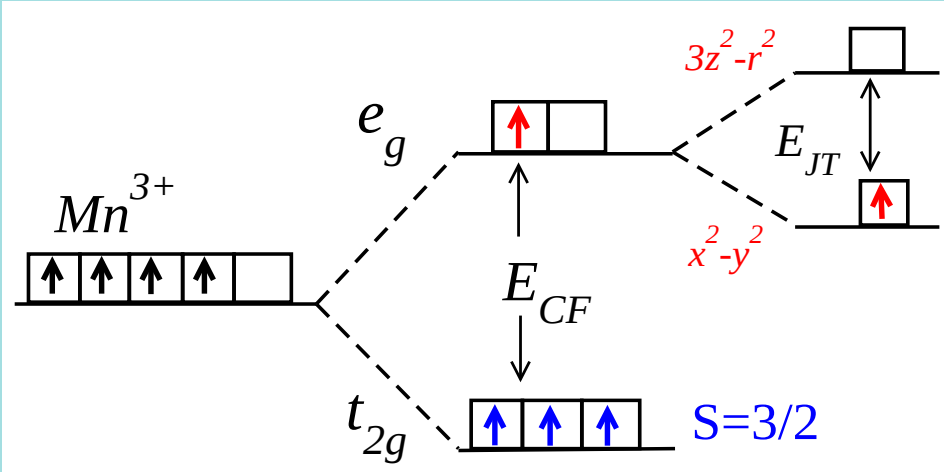
- ***SBMO* at finite pressure $\sim$ *NBMO* at zero pressure.**
- ***NBMO* at finite pressure $\sim$ *PBMO* at zero pressure.**

Summary of selected experiments

- special charge and orbital ordering phenomena at x=1/2
- insulating phases close to x=0 and x=1
- interesting doping regime for CMR: 0.25<x<0.45
- 2 overall parameters: bandwidth and disorder,
  independent of the specific chemical composition

Focus of present work:
understanding global trends away from commensurate fillings

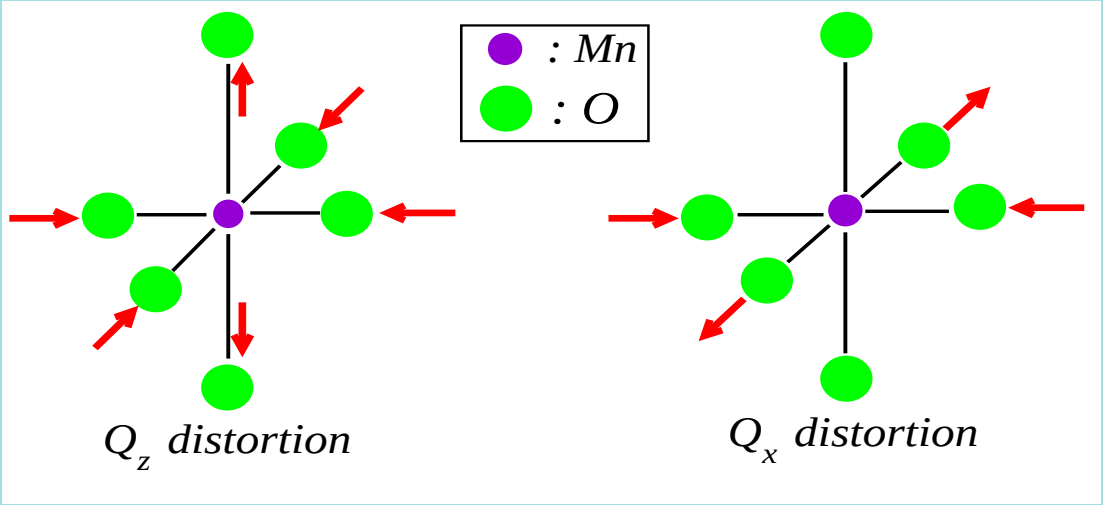
Model for Manganites



Main ingredients:

- **Kinetic energy; complicated hopping structure($t_{ij}^{\alpha\beta}$).**
- **Hund's rule coupling of t_{2g} spins (S=3/2) to e_g spin.**
- **Octahedra distortions; Jahn-Teller effect for e_g orbitals.**
- **Hubbard interactions: inter & intra orbital.**

Jahn-Teller distortions



• **Origin of JT effect: lifting of orbital degeneracy due to electron lattice coupling.**

Simplifications:

- Mn **ion is always at the centre of the octahedron.**
- O **displacements are along the** $Mn - O - Mn$ **bond.**

The Hamiltonian

$$H = H_{kin} + H_S + H_{el-latt} + H_{dis}$$

$$H_{kin} = \sum_{\langle ij\rangle\sigma}^{\alpha\beta} \{t^{\alpha\beta} c^{\dagger}_{i\alpha\sigma} c_{j\beta\sigma} + h.c.\} \quad ; \quad H_{dis} = \sum_i \epsilon_i n_i$$

$$H_S = -J_H \sum_i \mathbf{S}_i \cdot \hat{\sigma}_i + J_s \sum_{\langle ij\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$$H_{el-latt} = \lambda \sum_i \{Q^x_i \tau^x_i + Q^z_i \tau^z_i\} + \frac{K}{2} \sum_i \{(Q^x_i)^2 + (Q^z_i)^2\}$$

$\tau^x_i = \sum_\sigma (c^{\dagger}_{i1\sigma} c_{i2\sigma} + c^{\dagger}_{i2\sigma} c_{i1\sigma})$; $\tau^z_i = \sum_\sigma (c^{\dagger}_{i1\sigma} c_{i1\sigma} - c^{\dagger}_{i2\sigma} c_{i2\sigma})$

$\epsilon_i = \pm\Delta$ **; binary disorder.**

Simplifying assumptions

- **Treat $\{Q^x, Q^z\}$ in the adiabatic limit.**
- **Treat $\{\mathbf{S}\}$ as classical variables.**
- **Justification: Large t_{2g} spin, large O mass.**
- **For a given set of $\{Q^x, Q^z\}$ and $\{\mathbf{S}\}$, the Hamiltonian is bilinear in electronic operators.**

Still a difficult problem to solve !

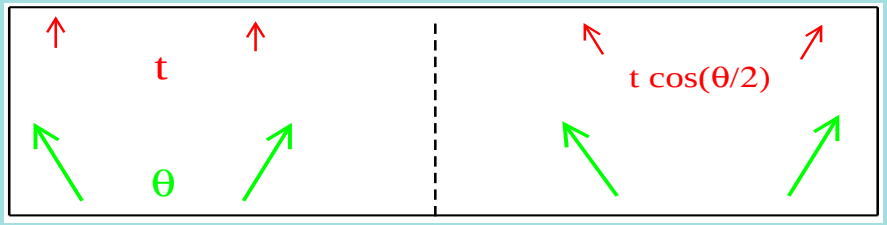
$\{Q^x, Q^z\}$ and $\{\mathbf{S}\}$ have to be determined self consistently.

Parameter space $\lambda/t, \Delta/t,\ J_H/t\ J_s/t$, n and T/t.

Q: How to determine the classical configurations?

Double Exchange Limit

$J_H/t \to \infty$; e_g **spins are slaved along the** t_{2g} **spin directions.**



$$H_{kin} = \sum_{\langle ij \rangle}^{\alpha\beta} \{ t_{ij}^{\alpha\beta} \gamma_{i\alpha}^{\dagger} \gamma_{j\beta} + h.c. \}$$

$$t_{ij}^{\alpha\beta} = t^{\alpha\beta} (\cos(\theta_i/2)\cos(\theta_j/2) + \sin(\theta_i/2)\sin(\theta_j/2) e^{\iota(\phi_i - \phi_j)})$$

θ_i, ϕ_i **: polar and azimuthal angles for spin** $\mathbf{S}_i$

- **hopping matrix specific to the perovskite manganites**

$t^{11} = t,\ t^{12} = t^{21} = \mp \frac{1}{\sqrt{3}}\ t,\ t^{22} = \frac{1}{3}\ t.$

Combining ED with Monte Carlo

Formally integrating out the fermions

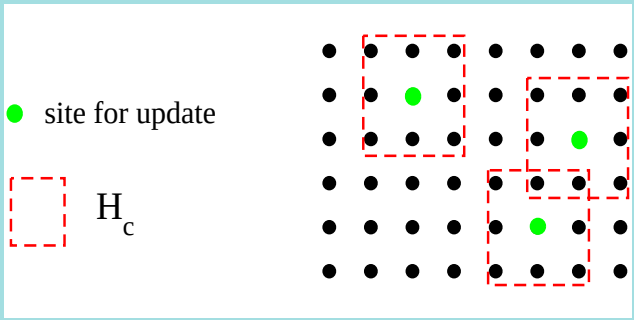
$$Z = \int \mathcal{D}\mathbf{S}\mathcal{D}Q \; Tr \; e^{-\beta H(\{Q,\mathbf{S}\})} \equiv \int \mathcal{D}\mathbf{S}\mathcal{D}Q \; e^{-\beta H_{eff}(\{Q,\mathbf{S}\})}$$

$$H_{eff}(\{Q,\mathbf{S}\}) = -\frac{1}{\beta} log \; Tr \; e^{-\beta H(\{Q,\mathbf{S}\})}$$

- **In practice one needs to know the eigenvalues of $H(\{Q,\mathbf{S}\})$ in order to evaluate the above trace.**
- **Straightforward method: combination of exact diagonalisation and classical Monte-Carlo (EDMC).**
- **Too costly in terms of cpu time.** $\tau_N \sim N^4$

Travelling Cluster Approximation(TCA)

- **Construct a Hamiltonian on a cluster of sites around the site to be updated during Monte-Carlo.**
- **Estimate ΔE for old and new configurations using the eigenvalues of the cluster Hamiltonian.**



- $\tau_N \sim N N_c^3$. **Achievable size $N \sim 1000$ for $N_c \sim 100$**
- **To compute electronic properties we use exact eigenfunctions of the full Hamiltonian in the TCA generated classical configurations.**

- **Detailed comparisons, cond-mat/0406082**

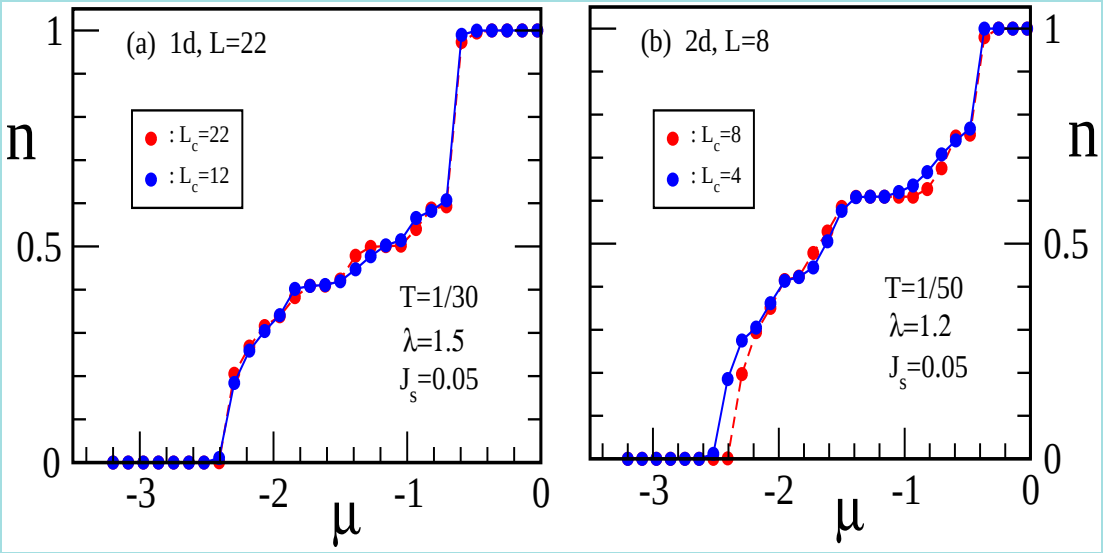
Calculated Quantities

1. **spin structure factor:** $S_{\vec{q}} = N^{-2} \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j e^{i\vec{q}\cdot(\vec{r}_i - \vec{r}_j)}$
2. **orbital structure factor:** $\tau_{\vec{q}} = N^{-2} \sum_{ij} \tau_i \cdot \tau_j e^{i\vec{q}\cdot(\vec{r}_i - \vec{r}_j)}$
3. **electronic density of states:** $N(\omega) = \frac{1}{N} \sum_k \delta(\omega - \epsilon_k)$
4. **local electronic density:** $\langle n_i \rangle = \langle \gamma_{i1}^\dagger \gamma_{i1} + \gamma_{i2}^\dagger \gamma_{i2} \rangle$
5. **conductivity using Kubo-Greenwood formula:**

$$\sigma(\omega) = \frac{\pi e^2}{hN} \sum_{ab} |\langle \psi_a | \hat{j} | \psi_b \rangle|^2 \frac{(f_b - f_a)}{(\epsilon_a - \epsilon_b)} \delta(\omega - (\epsilon_a - \epsilon_b))$$

$$\hat{j} = \mathrm{i} \sum_{\langle ij \rangle}^{\alpha\beta} (t_{ij}^{\alpha\beta} \gamma_i^\dagger \gamma_j - t_{ji}^{\alpha\beta} \gamma_j^\dagger \gamma_i)$$

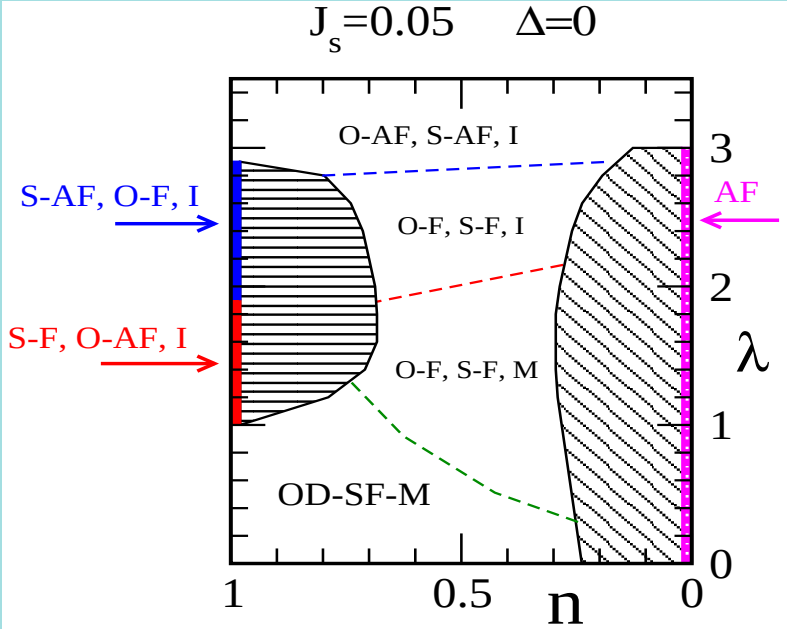
Comparison with exact results



red dots: ED results from Yunoki *et al.* PRL(1998).
blue dots: results using the cluster method.

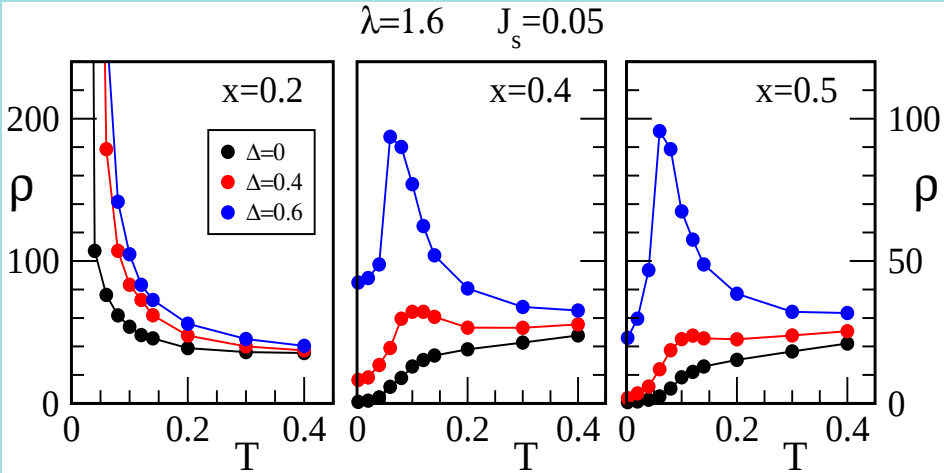
- **cpu constraints in 3d, minimum 4^3 cluster required.**

Zero Temperature Phase Diagram in 2D



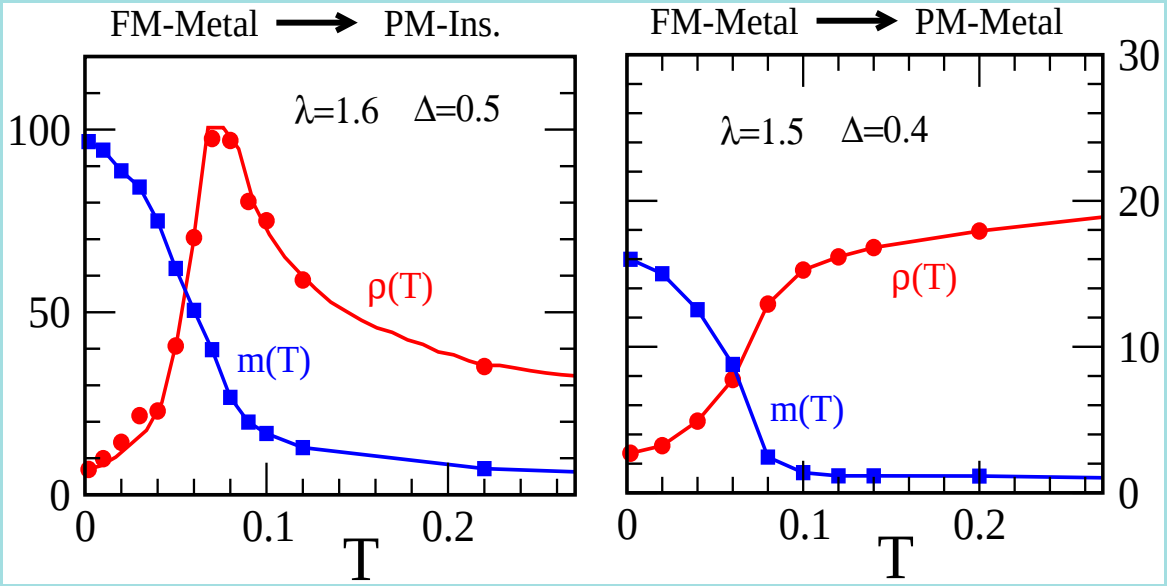
- **insulators at $n = 0$ and $n = 1$. Phase separations.**
- **FM-I phase with reduced magnetisation.**
- **larger J_s induces CO-OO-I phase at $n = 0.5$.**

Resistivity: variations in hole doping (x).



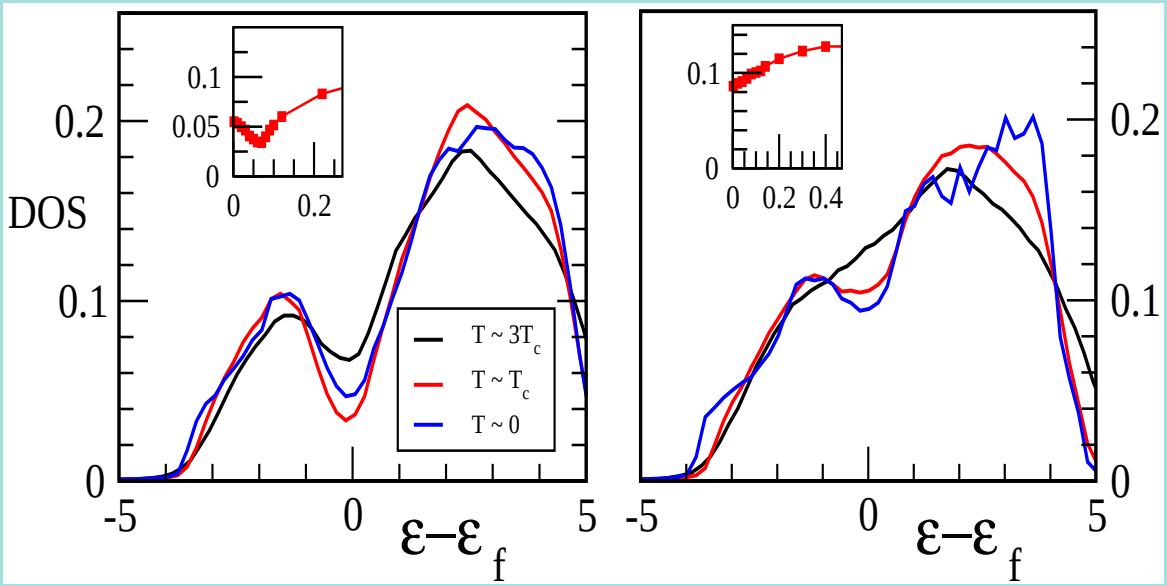
- **Low doping $\Rightarrow$ insulating phase.**
- **Disorder strongly affects the region near T_c. Inducing a metal-insulator transition.**

Resistivity and Magnetisation



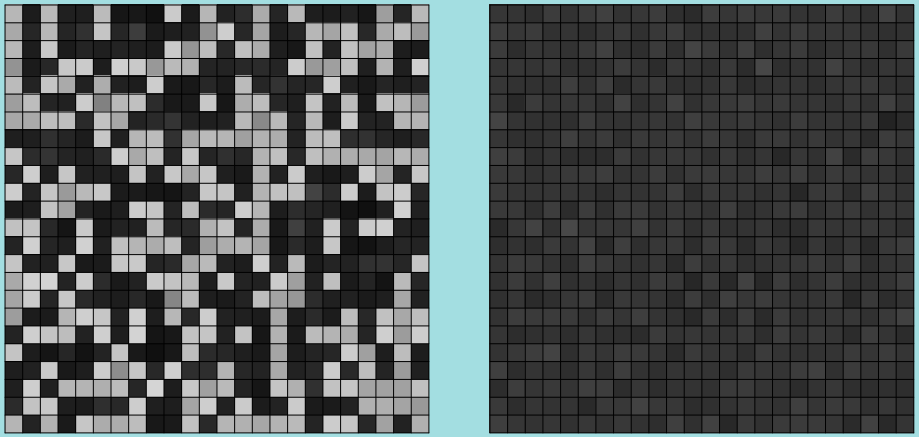
- **correlation between FM-PM and M-I transitions**
- **weak λ, Δ has typical DE like behavior.**
- **small disorder induces polaron trapping, drastically changing $\rho(T)$.**

Density of states



- **pseudogap in the DOS.**
- **DOS at E_f has different behavior in the two cases.**
- **does provide hints for different resistivity behaviors.**

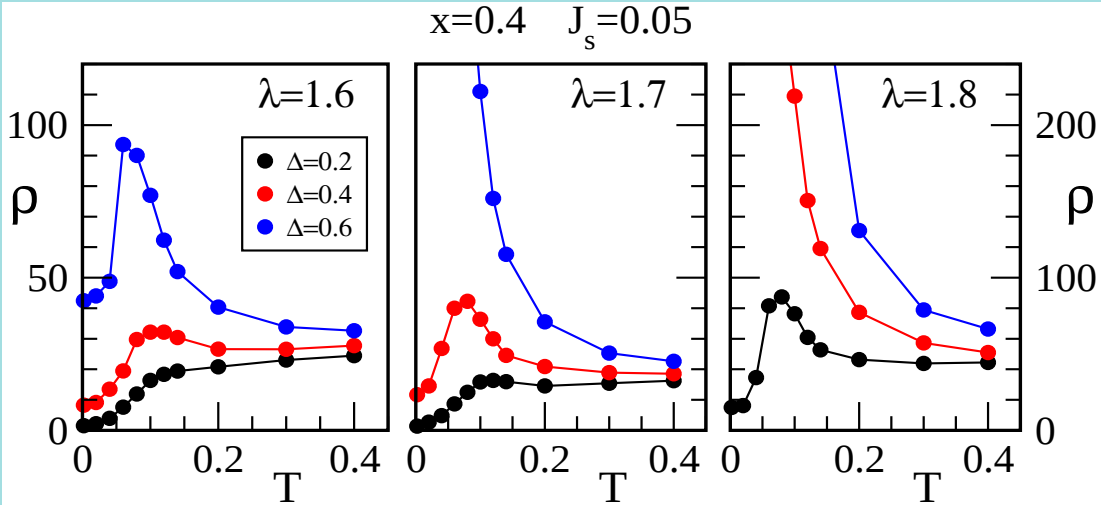
Real Space Density Profile



Understanding the MIT: Two type of states

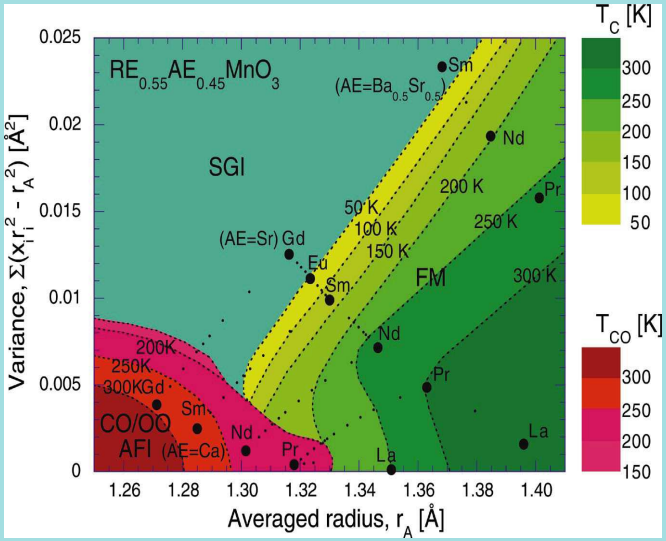
- **Mixture of polaronic and extended states at $T = 0$.**
- **Rise in $\rho(T)$, scattering of mobile electrons.**
- **Additional spin disorder manages to trap extended states $\Rightarrow$ insulating paramagnetic phase.**

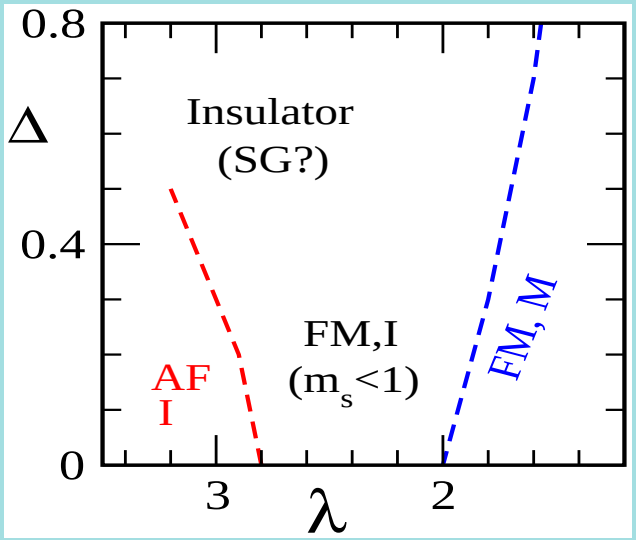
Resistivity: variations in bandwidth and disorder



- **change in $t \Rightarrow$ change in λ/t, J_s/t and Δ/t.**
- **we explore the effect of changing λ/t and Δ/t.**
- **3 transport regimes on increasing λ or Δ or both.**

Summary





- **Similar trends as seen in the experiments.**
- **spin glass phase in our model, yet to be confirmed.**
- **More realistic modeling: disorder in hoppings, changes in J_s/t.**
- **Work still in progress...**