Phase separation in doped Mott insulators

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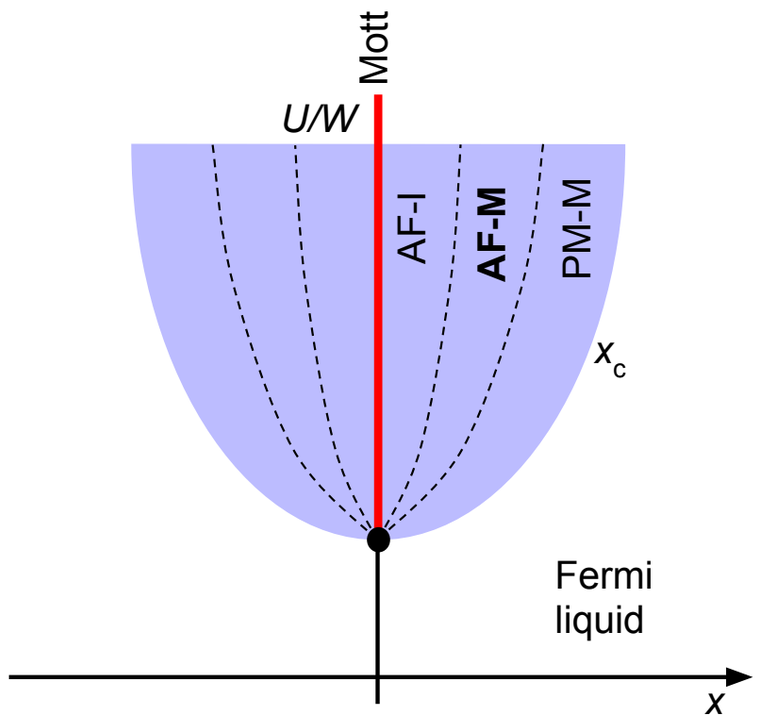
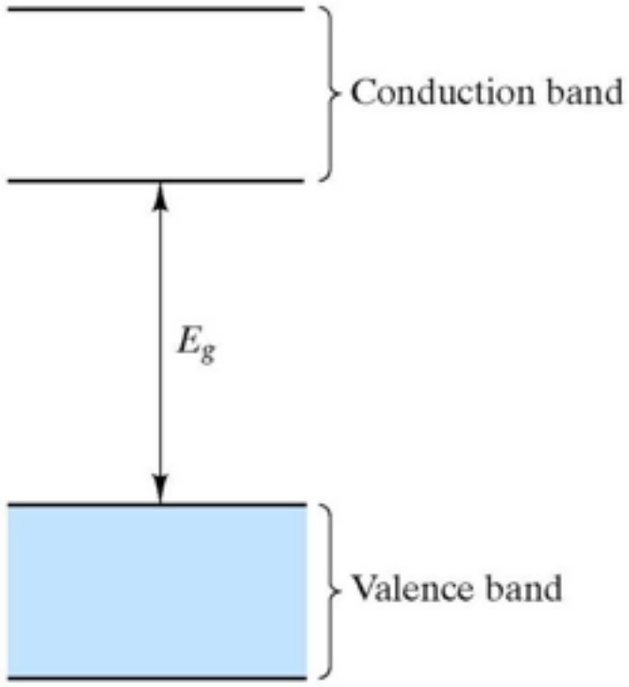
Motivated by the commonplace observation of Mott insulators away from integer filling, we construct a simple thermodynamic argument for phase separation in first-order doping-driven Mott transitions. We show how to compute the critical dopings required to drive the Mott transition using electronic structure calculations for the titanate family of perovskites, finding good agreement with experiment. The theory predicts the transition is percolative and should exhibit Coulomb frustration.

CMT Journal Club - July 8, 2014 - Tobias Meng

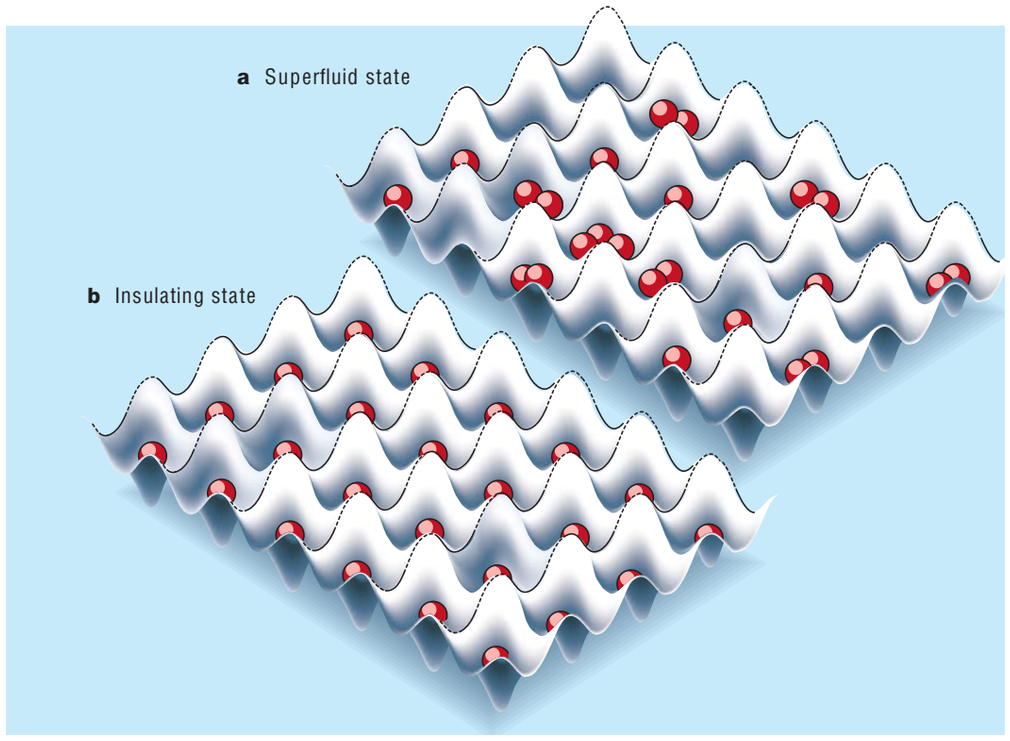


Reminder on Mott insulator

- Mott insulator: interaction driven insulator (unlike simple band insulator)
- Named after Neville Mott (Nobel price in 1977 with Anderson & van Vleck)



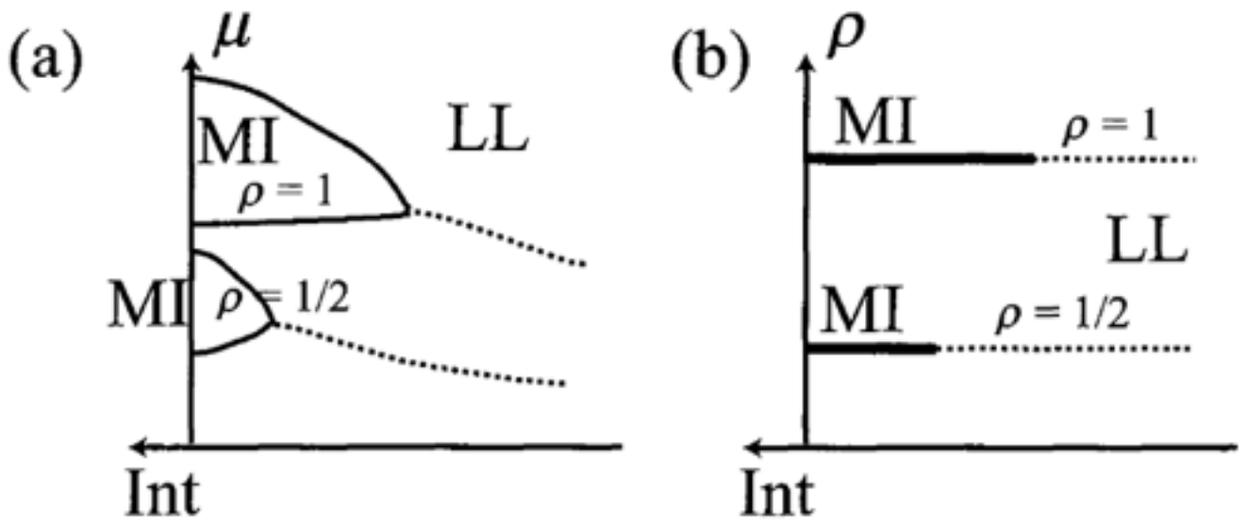
vs.



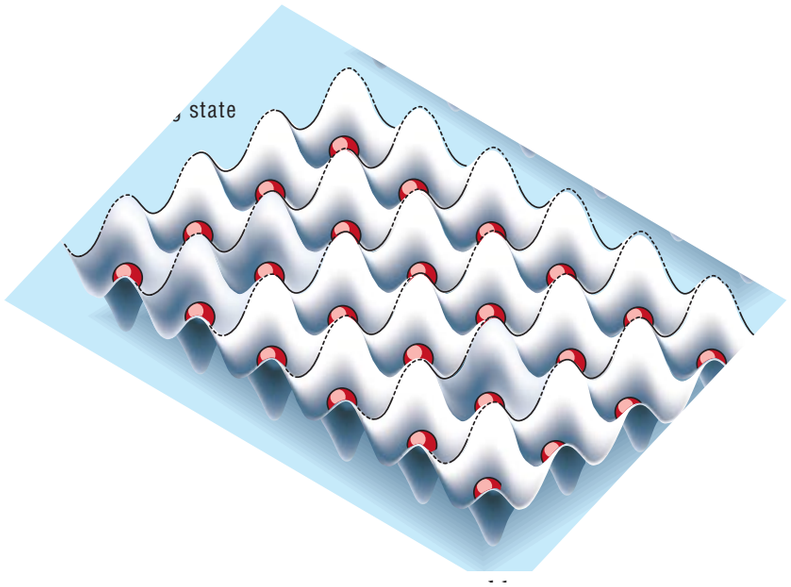
$$H_B = -w \sum_{\langle ij \rangle} \left(\hat{b}_i^\dagger \hat{b}_j + \hat{b}_j^\dagger \hat{b}_i \right) - \mu \sum_i \hat{n}_{bi} + U \sum_i \hat{n}_{bi}(\hat{n}_{bi} - 1)$$

Motivation

- Theoretically: Mott insulator exists at commensurate fillings only



[T. Giamarchi, Quantum Physics in One Dimension]



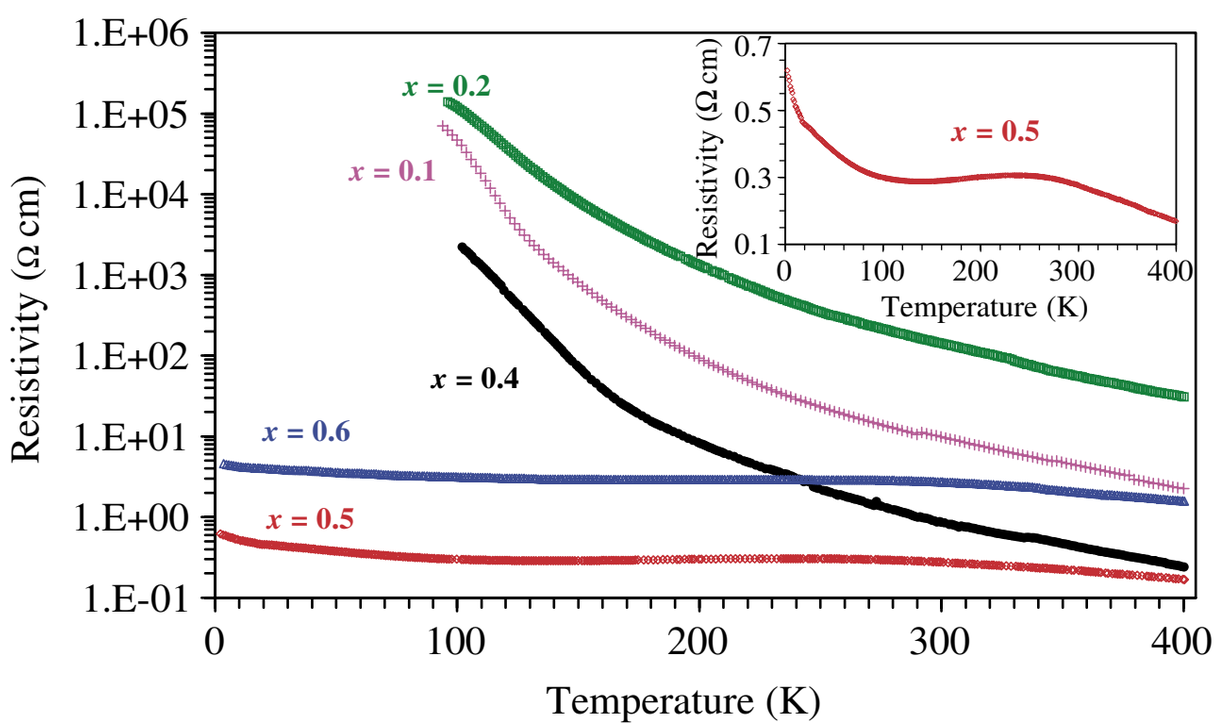


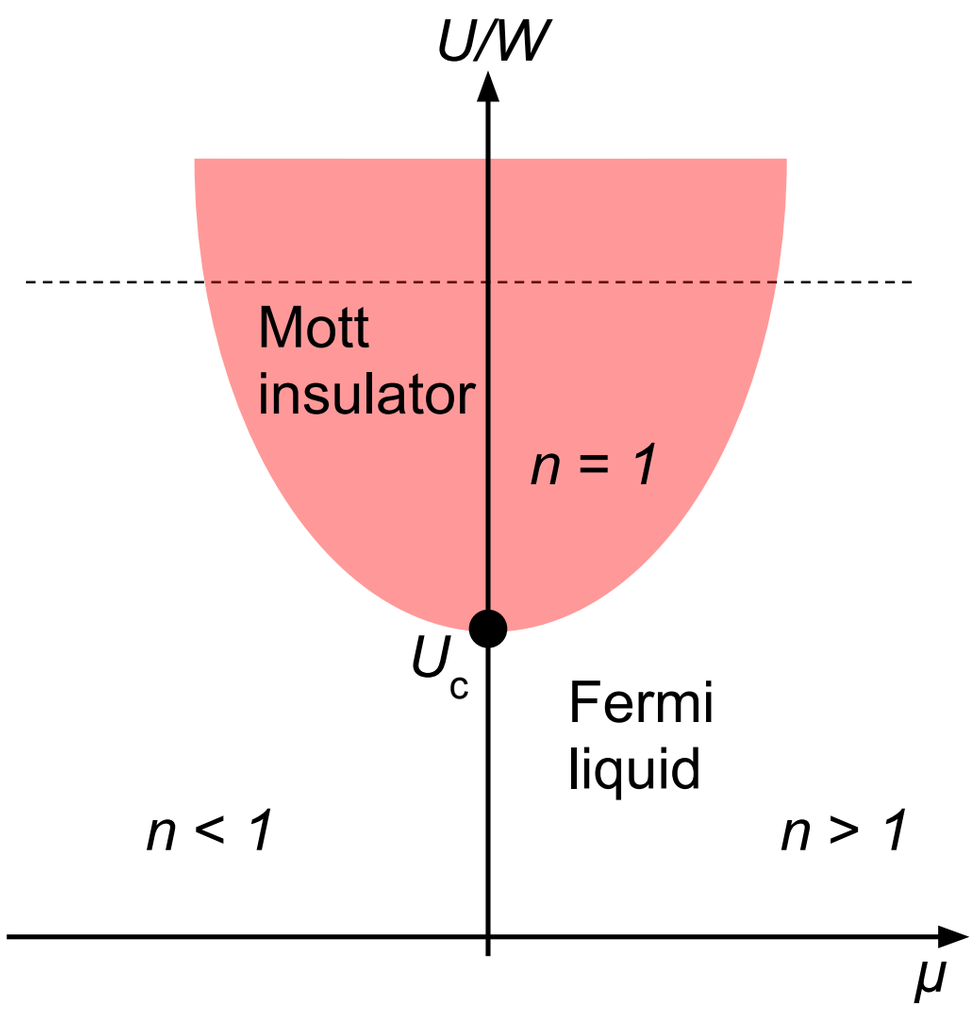
Fig. 1. Temperature dependence of resistivity in $Y_{1-x}Cd_xVO_3$. The inset shows the data for $x = 0.5$.

[A. A. Belik *et al.*, Journal of Magnetism and Magnetic Materials **310**, e240 (2007)]

- In some experiments: insulating behavior also seen at other fillings!

Simple thermodynamic theory

- Analyze energy densities of metallic and insulating state close to U_c



$$\epsilon_{\mathrm{m}}(\mu, U) = \epsilon_0 + d_{\mathrm{m}} \Delta U - \frac{1}{2}\kappa(\Delta\mu)^2$$

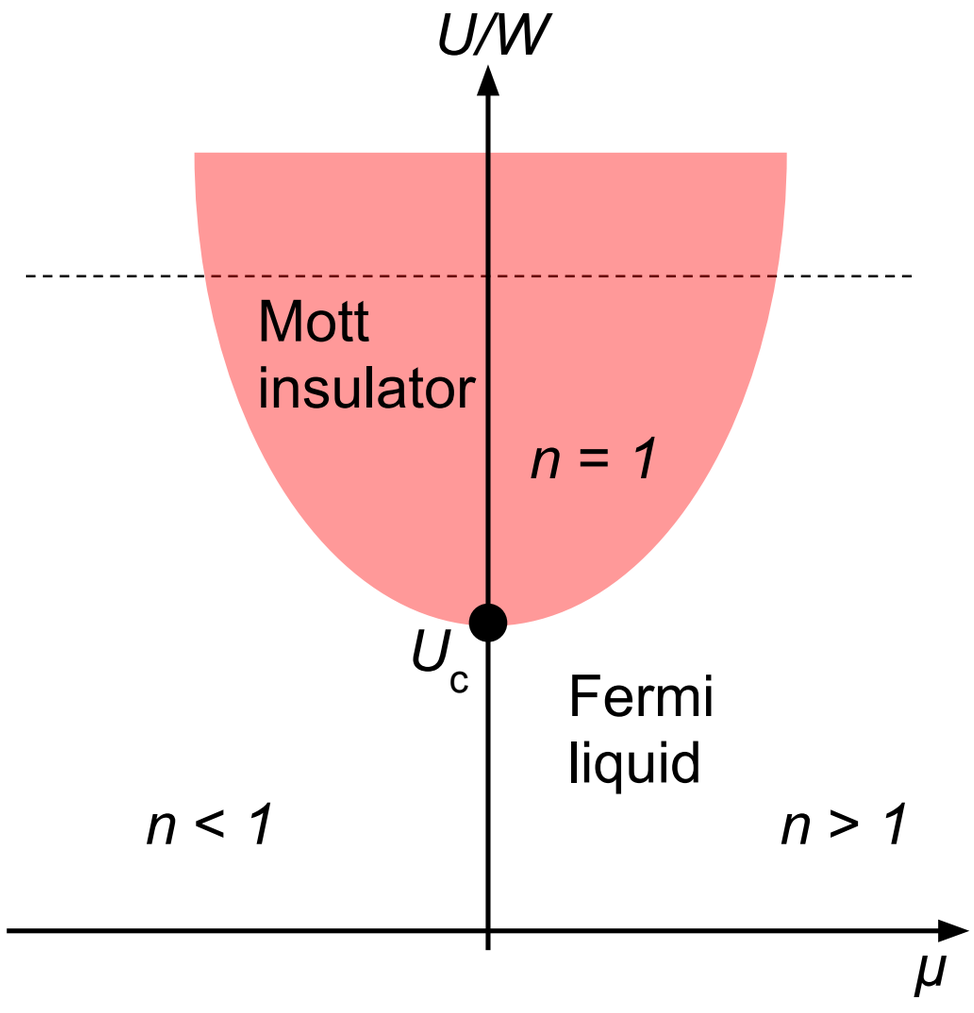
$$\epsilon_{\mathrm{i}}(\mu, U) = \epsilon_0 + d_{\mathrm{i}} \Delta U.$$

$$\Delta U = U - U_c \qquad \Delta\mu = \mu - \mu_{n=1}$$

- double occupancy per site in metal/insulator:
  $d_{m,i} = \langle n_{i\uparrow} n_{i\downarrow} \rangle$
- electron compressibility: $\kappa = \dfrac{\partial x}{\partial \mu}$
- Doping relative to half filling: $x = n - 1$

Phase boundary

- Analyze energy densities of metallic and insulating state close to U_c



$$\epsilon_{\mathrm{m}}(\mu, U) = \epsilon_0 + d_{\mathrm{m}} \Delta U - \frac{1}{2}\kappa(\Delta\mu)^2$$

$$\epsilon_{\mathrm{i}}(\mu, U) = \epsilon_0 + d_{\mathrm{i}} \Delta U.$$

- Phase boundary at $\epsilon_m(\mu, U) = \epsilon_i(\mu, U)$

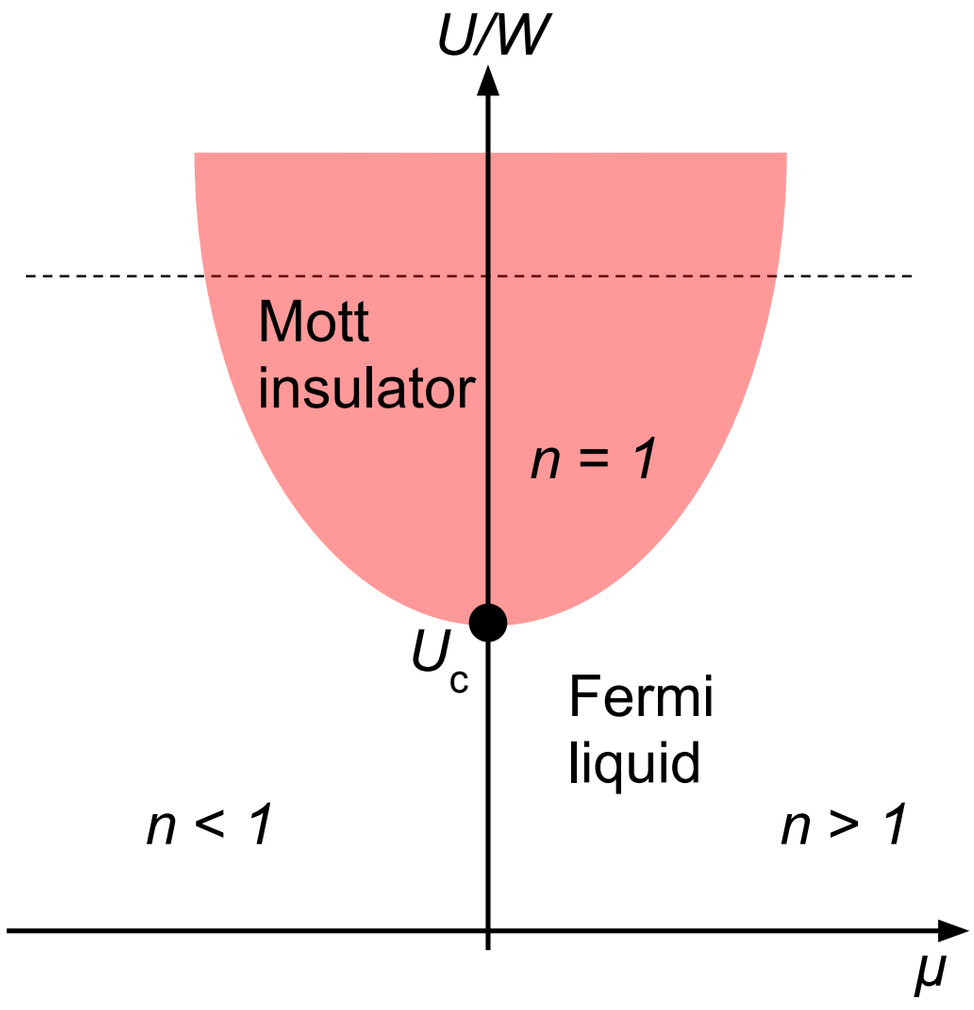
$$\Delta U = \frac{\Delta\mu^2}{2} \frac{\kappa}{d_m - d_i}$$

($\Delta U \sim \Delta\mu^2$ consistent with DMFT)

[P. Werner and A. J. Millis, PRB **75**, 085108 (2007)]

Phase boundary

- Analyze energy densities of metallic and insulating state close to U_c



$$\epsilon_{\mathrm{m}}(\mu, U) = \epsilon_0 + d_{\mathrm{m}} \Delta U - \frac{1}{2}\kappa(\Delta\mu)^2$$

$$\epsilon_{\mathrm{i}}(\mu, U) = \epsilon_0 + d_{\mathrm{i}} \Delta U.$$

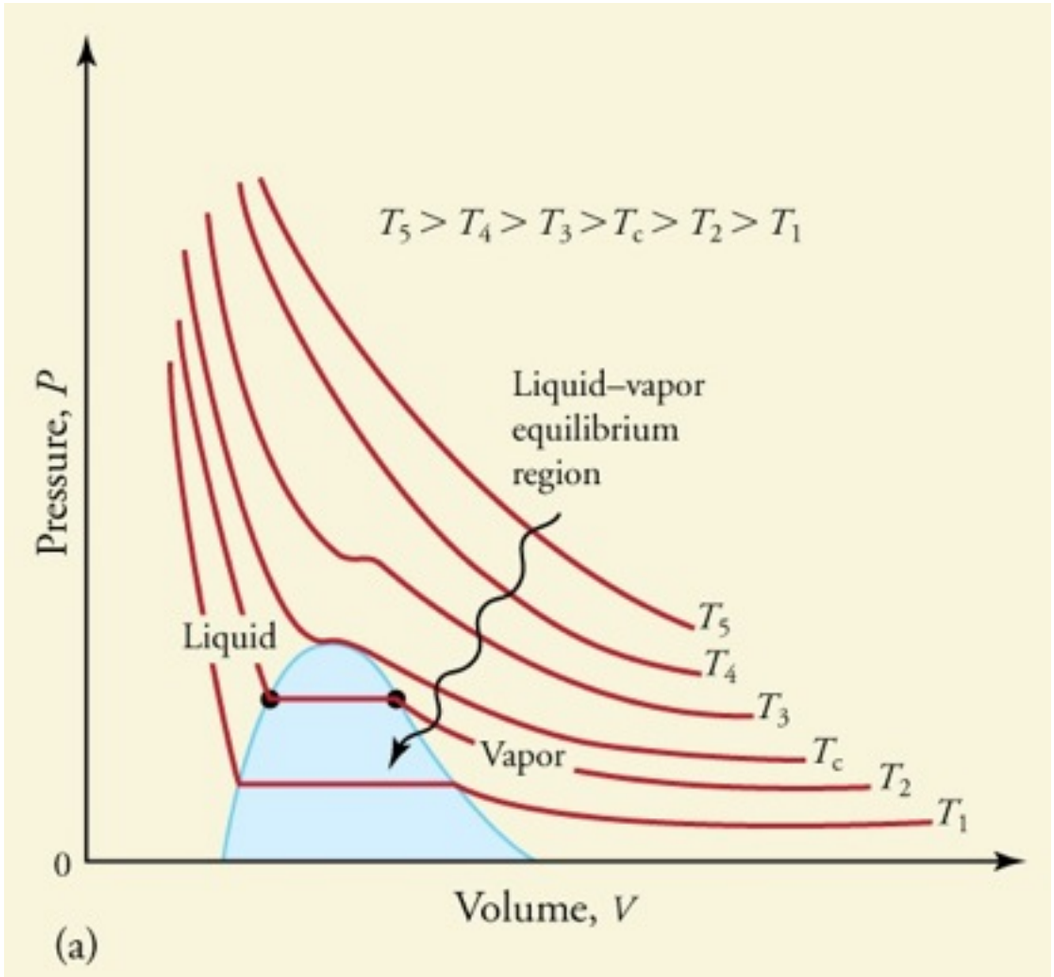
- Critical doping (metallic density)

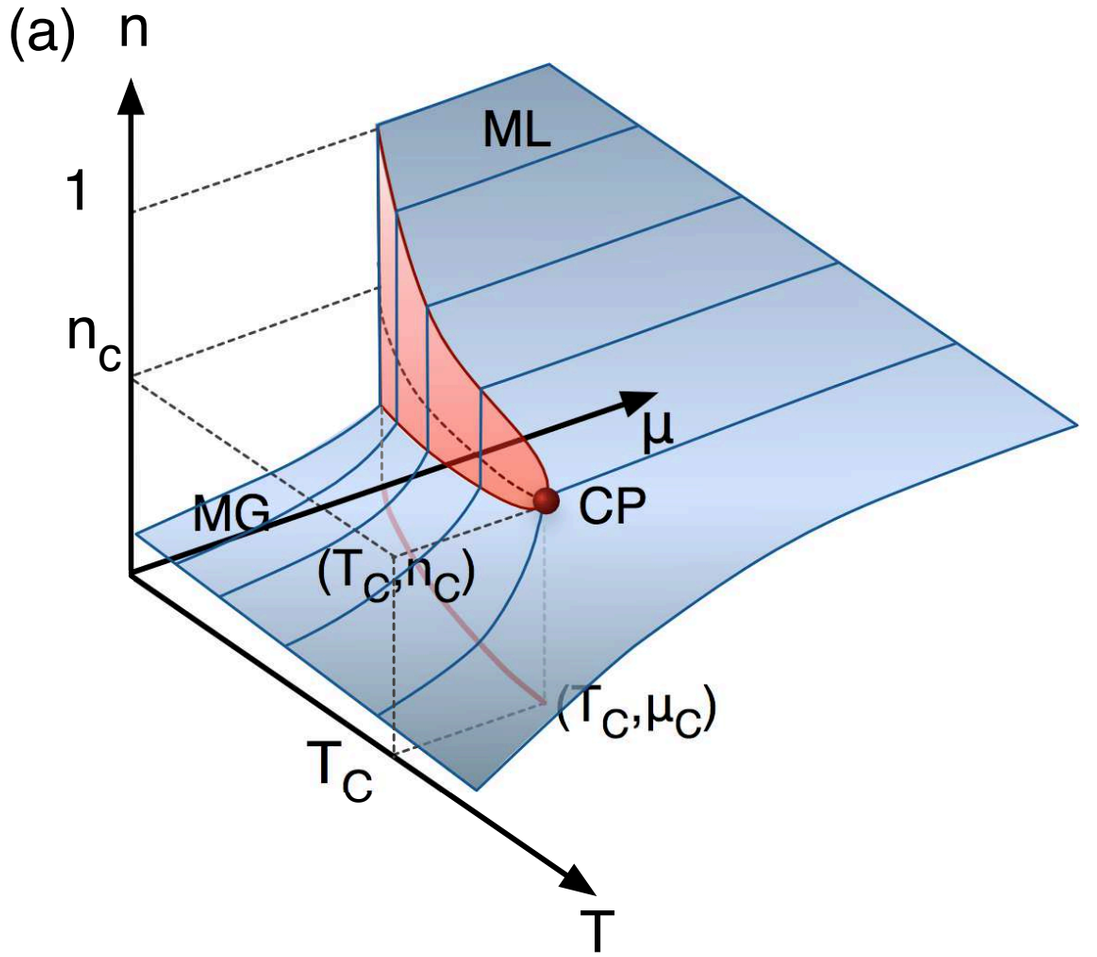
$$x = -\frac{\partial \epsilon_m}{\partial \mu} = \sqrt{\Delta U \cdot 2\kappa(d_m - d_i)}$$

Phase separation

- Similar to liquid-gas transition: densities $0 < |x| < x_c$ forbidden

  $\Rightarrow$ phase separation





ML - Mott liquid CP - critical point

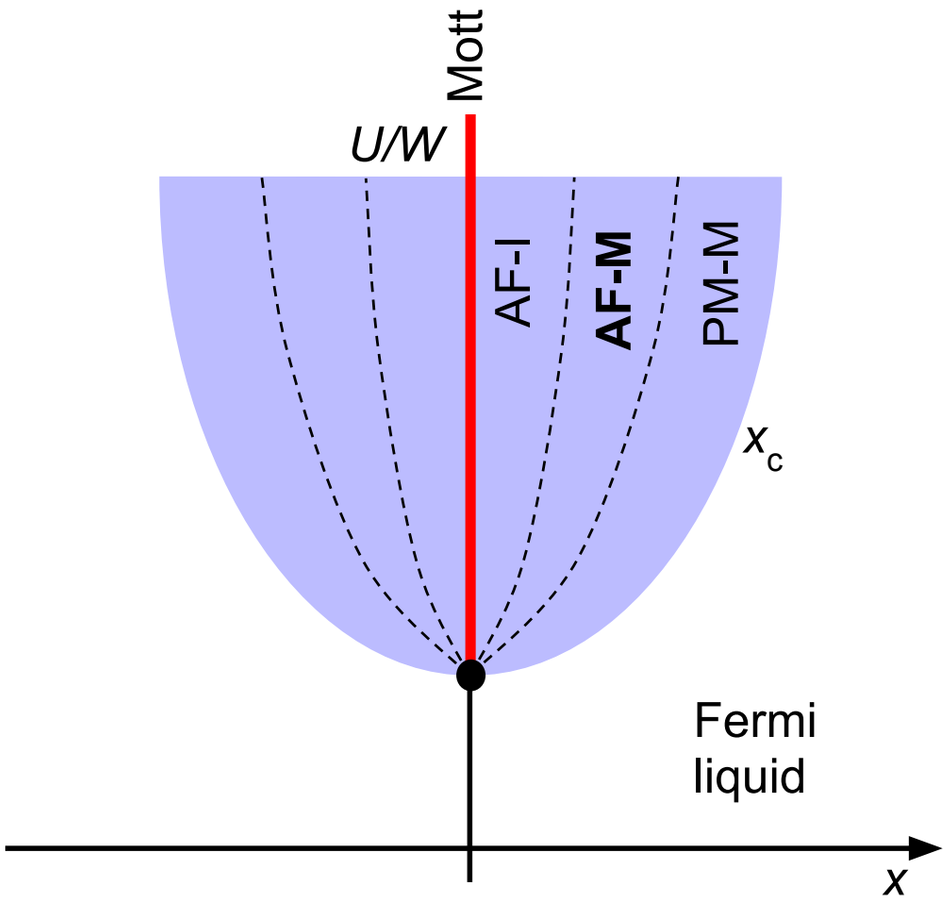
MG - Mott gas red - coexistance region

[D. Galanakis et al., Philosophical Transactions of the Royal Society A **369**, 1670 (2011)]

Percolation

- In 3D:
  - conducting regions percolate for $\frac{x}{x_c} \sim \phi_c = \frac{1}{3}$
  - Mott regions percolate for $\frac{x}{x_c} \sim 1 - \phi_c = \frac{2}{3}$

[J. W. Essam, Rep. Prog. Phys. **43**, 833 (1980)]



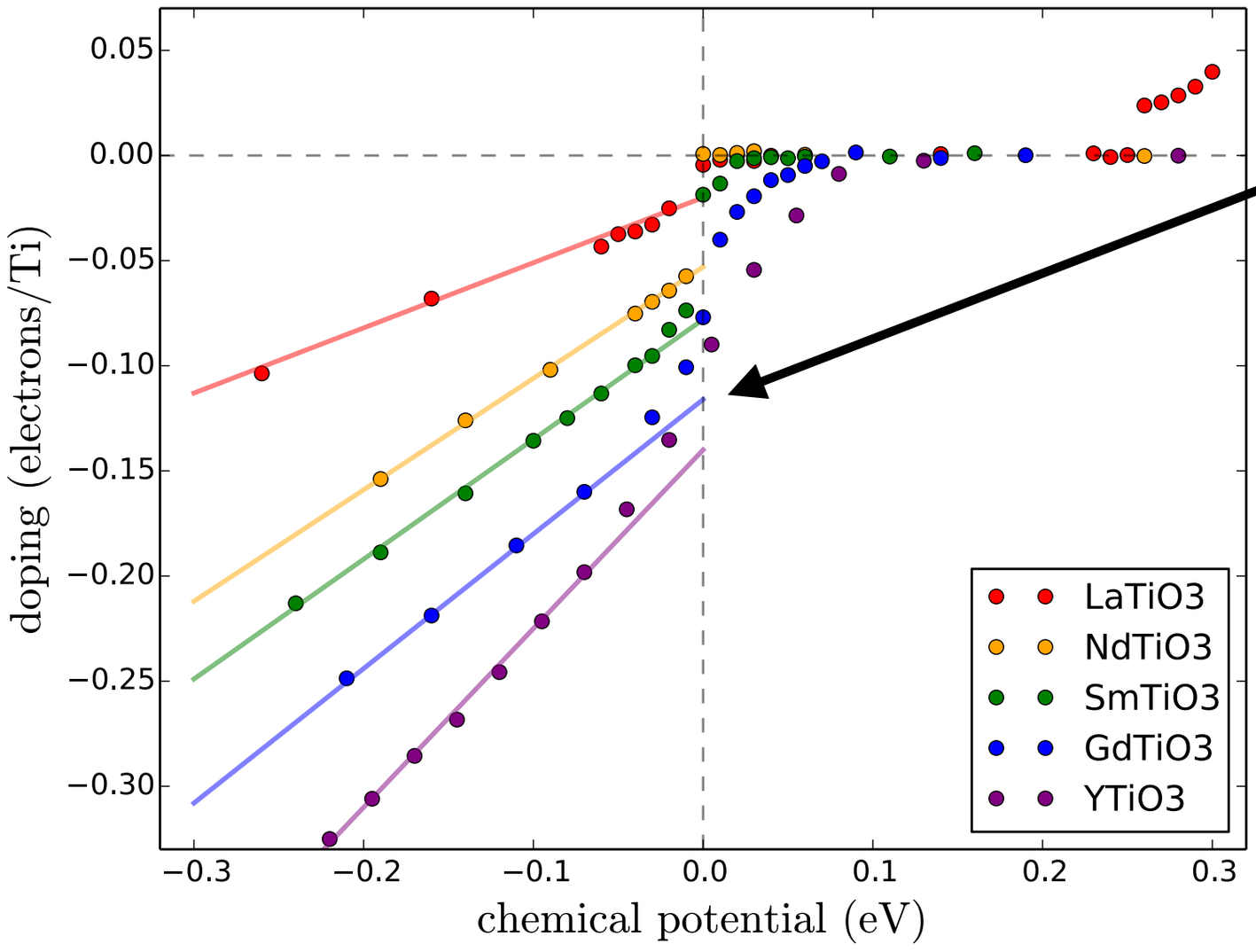
surface tension: favors one big puddle
Coulomb energy: penalizes charge imbalance

$\Rightarrow$ domains have size determined by this competition
(Coulomb frustration of phase separation)

- In 2D: $\phi_c = \frac{1}{2}$ - no coexistence (no AFM-M phase)

Numerics

- Use DFT+DMRG: calculate doping x as function of chemical potential μ



$$x = -\frac{\partial n_m}{\partial \mu} = \sqrt{\Delta U \cdot 2\kappa(d_m - d_i)}$$

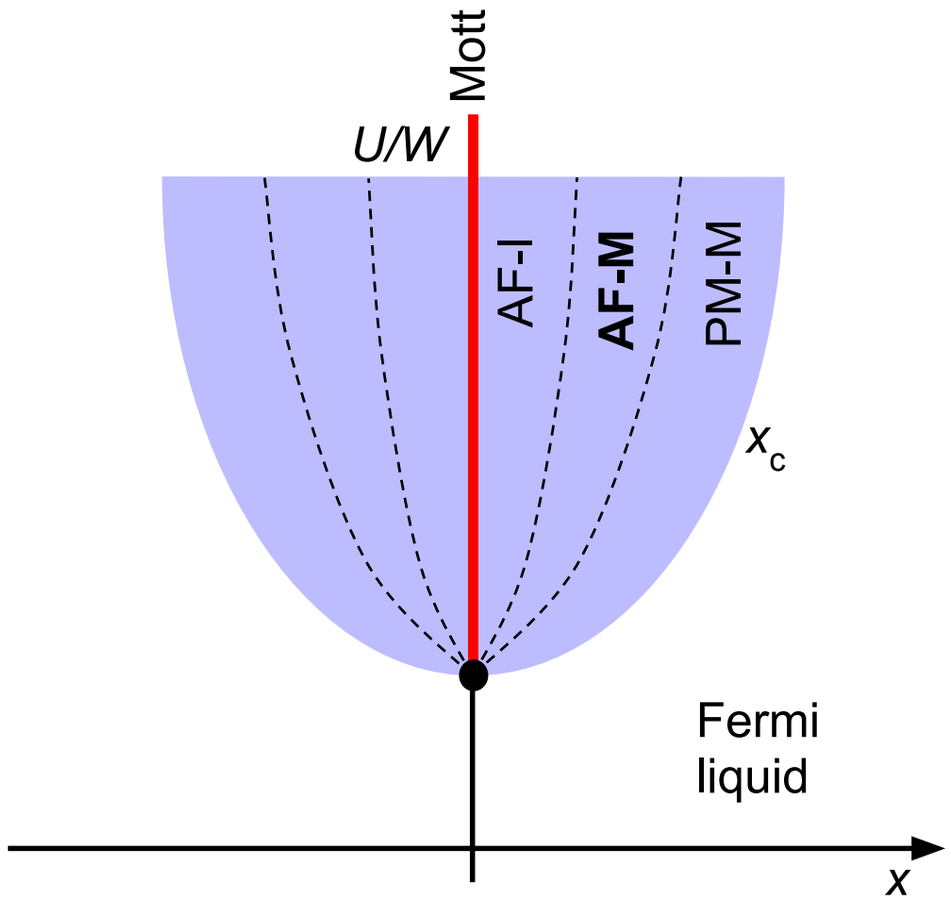
extract parameters from numerics:

Compound	κ (e/eV)	d_m	d_i	U_c (eV)	x_c
$LaTiO_3$	0.20	0.15	0.13	8.8	4%
$SmTiO_3$	0.22	0.22	0.19	6.0	20%
$YTiO_3$	0.28	0.23	0.20	4.7	27%

size ↘ : correlations ↗

qualitative agreement (also with experiment: off by a factor of 2) - no disorder & polarons

Summary



Summary – We have outlined a theory for the first-order filling-controlled Mott transition, which predicts intrinsic electronic phase separation when a Mott insulator is doped away from half-filling, and demonstrated explicitly how to calculate the critical doping x_c in electronic structure calculations.

- Simple thermodynamic theory

$$\epsilon_{\rm m}(\mu, U) = \epsilon_0 + d_{\rm m}\Delta U - \frac{1}{2}\kappa(\Delta\mu)^2$$

$$\epsilon_{\rm i}(\mu, U) = \epsilon_0 + d_{\rm i}\Delta U.$$

- Allows to determine critical doping

$$x_c = \sqrt{\Delta U \cdot 2\kappa(d_m - d_i)}$$

- For $0 < |x| < x_c$: percolation of phases

Simple theoretical description: Mott-Hubbard

- Example for bosons on lattice ($[b_i, b_j^\dagger] = \delta_{ij}$) with
  - coordination number Z
  - hopping w
  - on-site interaction U

- Bose-Hubbard Hamiltonian:

$$H_B = -w \sum_{\langle ij \rangle} \left(\hat{b}_i^\dagger \hat{b}_j + \hat{b}_j^\dagger \hat{b}_i \right) - \mu \sum_i \hat{n}_{bi} + U \sum_i \hat{n}_{bi}(\hat{n}_{bi} - 1)$$

- Mean field phase diagram:

$$\langle b_i^\dagger b_i \rangle = n \quad \text{in Mott lobe } n$$

[S. Sachdev, Quantum phase transitions]