

The fate of the Wigner crystal in solids

part II: low dimensional materials

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Outline

- competing interactions modify/favor Wigner Crystallization
- low dimensionality: structural transitions and stabilization of Wigner crystal

Wigner crystallization (1934)

$$H = \sum_i \frac{p_i^2}{2m} + \frac{1}{2} \sum_{i,j} \frac{e^2}{|r_i - r_j|}$$

$$\frac{1}{n} = \frac{4\pi}{3} R_s^3$$
$$r_s = R_s / a_B$$

Kinetic energy

$$1/r_s^2$$

<-- competition -->

potential energy

$$1/r_s$$

- At low density, electrons in a compensating jellium **crystallize** to **minimize the electrostatic repulsion**
- Not around $r_s=1$, as one could naively expect (metal stabilized by screening!)
 - 3D, BCC, $r_s \sim 100$
 - 2D, Triangular, $r_s \sim 40$
- Very low density, hard to observe !
 - electrons at the surface of liquid helium [Grimes & Adams 79]
 - semiconductor heterostructures GaAs/AlGaAs [Yoon et al. 99]

Wigner crystal in solids?

Wigner Crystal Type of Charge Ordering in an Organic Conductor with a Quarter-Filled Band:
 $(DI-DCNQI)_2Ag$

K. Hiraki^{1*} and K. Kanoda^{1,2}

Phys. Rev. Lett. 80, 4737–4740 (1998)

**A ‘checkerboard’ electronic crystal
state in lightly hole-doped
 $Ca_{2-x}Na_xCuO_2Cl_2$**

NATURE | VOL 430 | 26 AUGUST 2004 |

T. Hanaguri^{1,2}, C. Lupien³, Y. Kohsaka⁴, D.-H. Lee^{5,6}, M. Azuma^{2,7},
M. Takano⁷, H. Takagi^{1,2,4} & J. C. Davis³

**Crystallization of charge holes in
the spin ladder of $Sr_{14}Cu_{24}O_{41}$**

NATURE | VOL 431 | 28 OCTOBER 2004 |

P. Abbamonte^{1,2}, G. Blumberg³, A. Rusydi^{1,4}, A. Gozar^{3,5}, P. G. Evans⁶,
T. Siegrist³, L. Venema⁴, H. Eisaki⁷, E. D. Isaacs^{3,8} & G. A. Sawatzky⁹

Wigner Crystallization in $Na_3Cu_2O_4$ and $Na_8Cu_5O_{10}$ Chain Compounds

P. Horsch, M. Sofin, M. Mayr, and M. Jansen PRL 94, 076403 (2005)

Can we talk about Wigner crystallization in a solid?

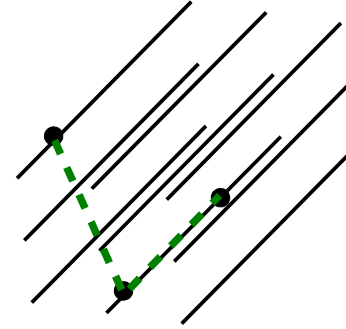
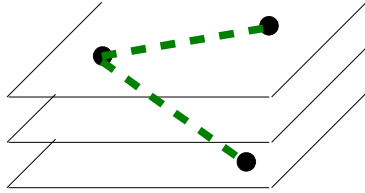
NO, in the strict sense: many **competing interactions**

- host lattice \neq vacuum, commensurability --> Baeriswyl's talk
- electron-phonon interaction --> Wigner crystal of polarons
- proximity to Mott insulating phase, magnetism --> stripes, spin polarons...
- counterions are not a uniform jellium, periodic potential/disorder --> Alloul's talk
-

YES, in a generalized sense

- all these effects **reduce the kinetic energy**, and favor charge localization, making the realization of a “Wigner crystal” easier.
- “Bad metals” are better candidates, because screening is less effective
- Often “observed” in **low-dimensional compounds**: WHY?

Low dimensional solids



3D solids where the electron motion is constrained to 2D layers or 1D chains by anisotropy of transfer integrals

=> **reduction of kinetic energy**

BUT

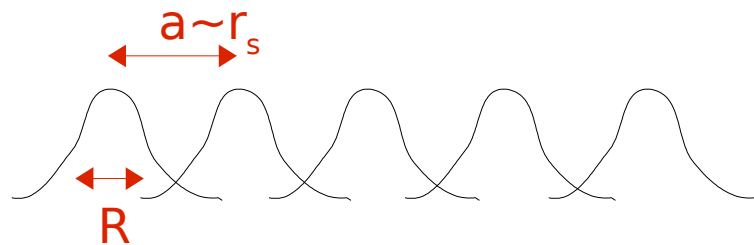
long range Coulomb interaction remains 3D and isotropic
(interlayer, interchain coupling cannot be neglected)

=> **no reduction of potential energy**

STABILIZATION OF WIGNER CRYSTAL WITH RESPECT TO PURE 3D CASE ?

Simple estimate: Lindemann criterion

The crystal melts when the radius of the localized wavefunctions in the solid attains a fraction of the interparticle distance [Pines & Nozières 1958]



$$\frac{R}{a} > \delta$$

Isotropic Wigner crystal in 3D:

- radius R related to average frequency of density oscillations (plasma frequency)

$$\omega_W = \omega_p / \sqrt{3} \sim r_s^{-3/2}$$

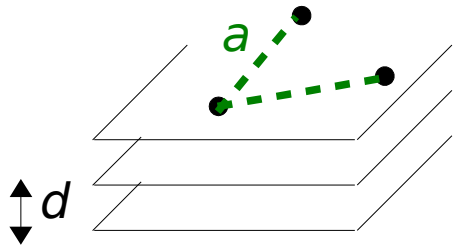
$$R = \sqrt{\frac{3\hbar}{2m\omega_W}} \sim r_s^{3/4}$$

- Interparticle distance $a \sim r_s$

$$\frac{R}{a} \sim \frac{r_s^{3/4}}{r_s} = \delta \longrightarrow r_s^c \sim 100$$

Crystal melting in layers/chains

- For a given (3D) density, the interparticle separation a in the layers depends on the interlayer separation d



$$n \sim \frac{1}{a^2 d} \sim \frac{1}{r_s^3} \longrightarrow a \sim r_s^{3/2} d^{-1/2}$$

- Squeezing the structure along z axis increases the interparticle distance a , but the localization radius R is essentially determined by isotropic 3D interactions

Lindemann criterion => **stabilization of Wigner crystal**

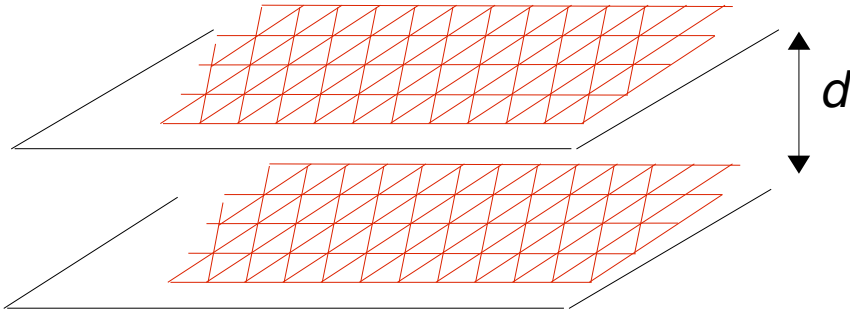
$$\frac{R}{a} \sim \frac{r_s^{3/4}}{r_s^{3/2} d^{-1/2}} = \delta \longrightarrow \boxed{r_s^c \sim 2.5 d^{2/3}} \quad \text{Layers}$$

$$\boxed{r_s^c \sim d^{8/9}} \quad \text{chains}$$

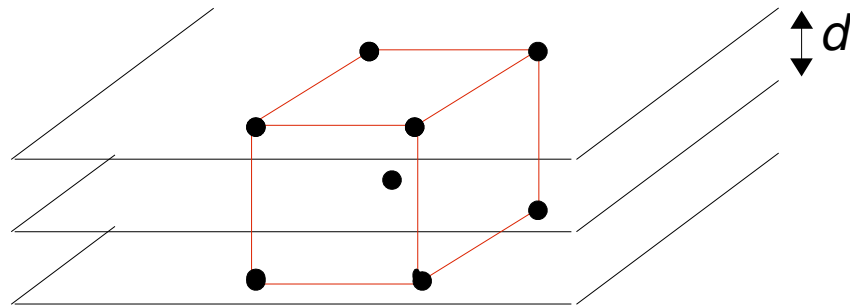
- saturates at some minimum d when interlayer tunneling sets in

Structural transitions

Vary n for fixed interlayer (interchain) distance d



- $d \gg r_s$ (high density, large interlayer distance)
weak interlayer coupling
=> staggered triangular



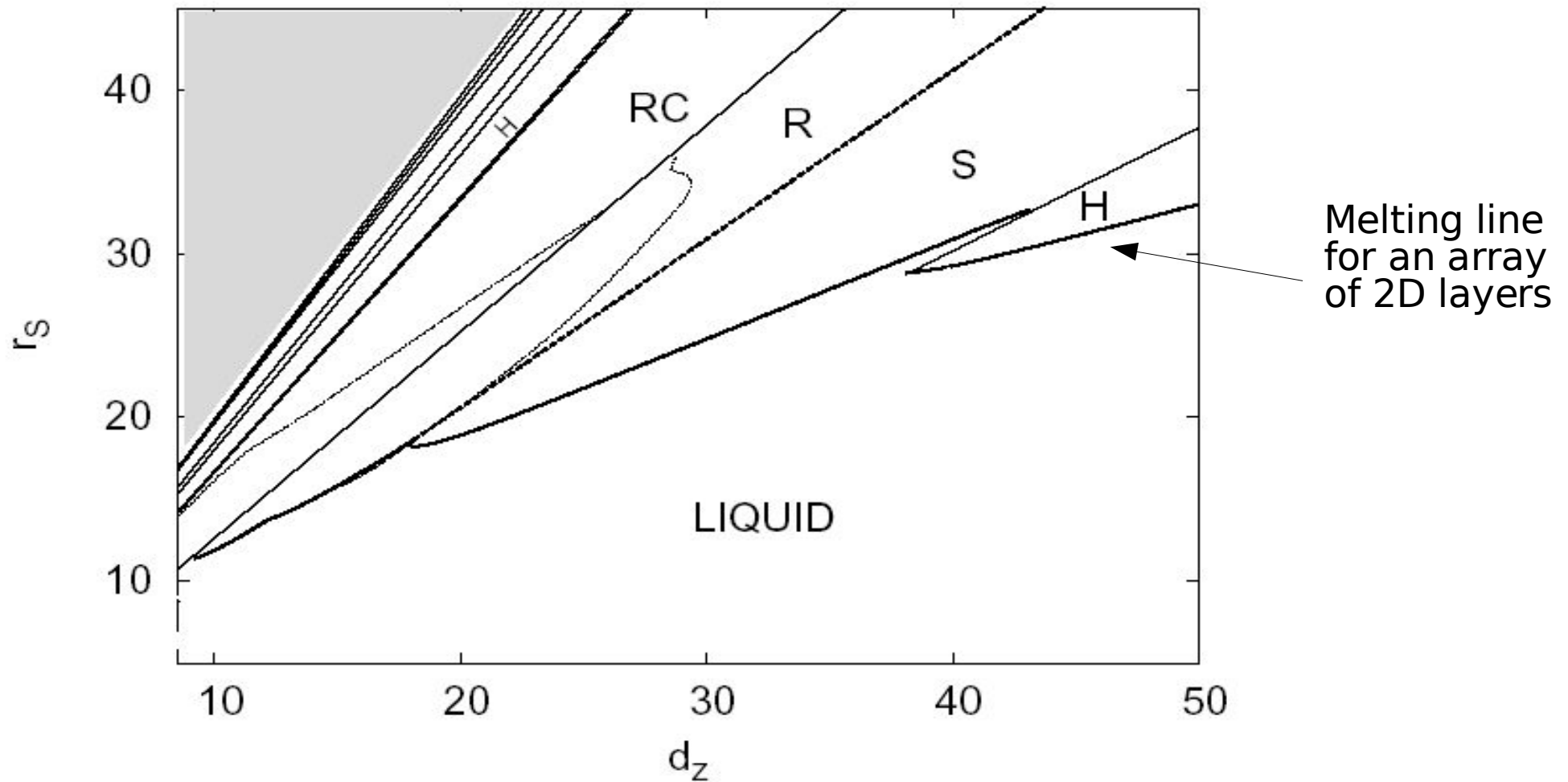
- $d \ll r_s$ (low density, small interlayer distance)
strong interlayer coupling
=> BCC in 3D space

example: parameters for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

- triangular ordering stable for $x > 0.2$

- square planar symmetry as observed in STM experiments is favored for $0.06 < x < 0.2$

Phase diagram for Wigner crystallization in layered solids

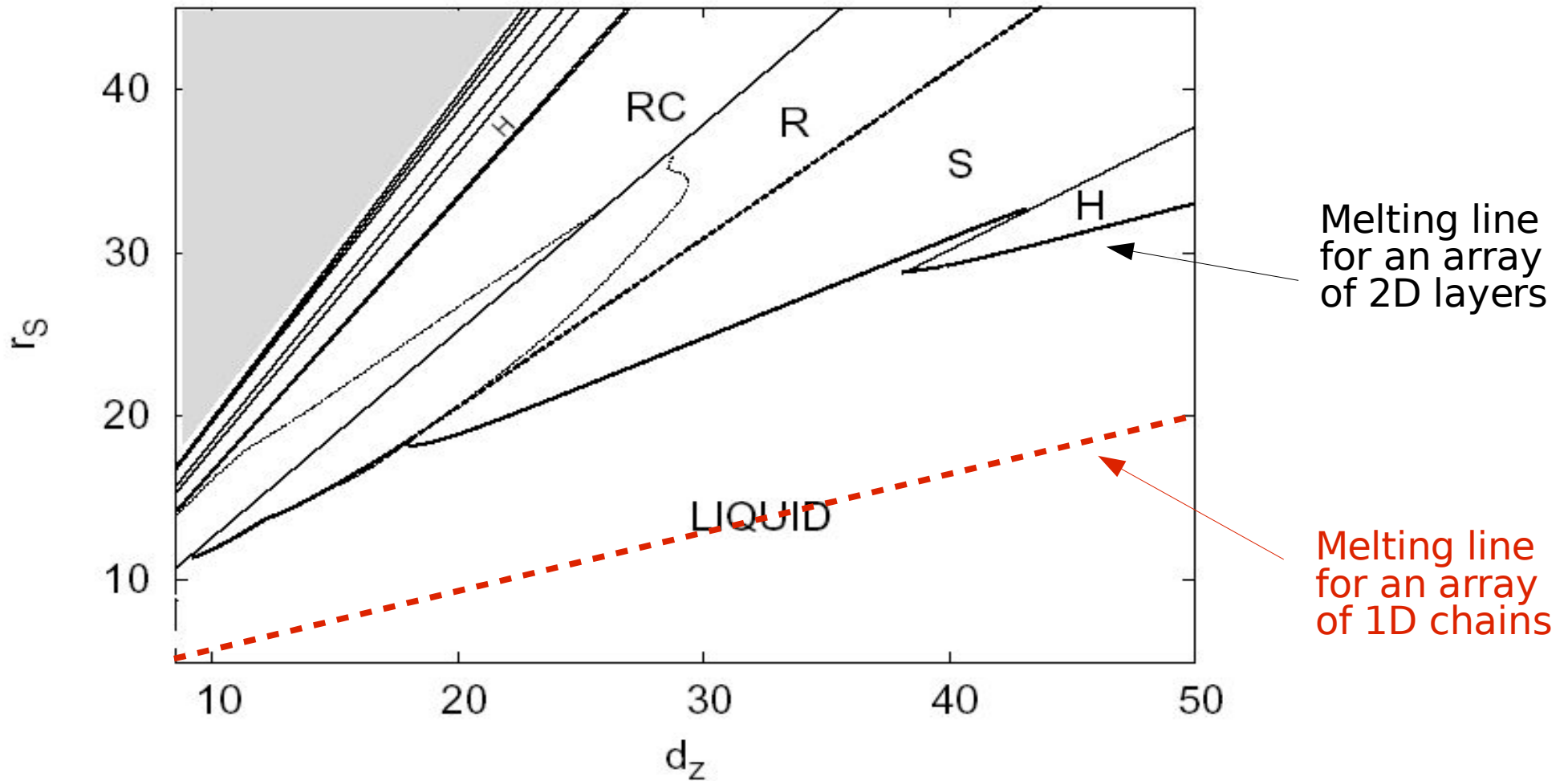


Based on quadratic hamiltonian for small displacements:

$$H = NE_M + \sum_i \frac{p_i^2}{2m} - \frac{e^2}{4} \sum_{i,j \neq i} (\vec{u}_i - \vec{u}_j) \hat{I}_{ij} (\vec{u}_i - \vec{u}_j)$$

- structural transitions: compare energies
- melting: Lindemann criterion

Phase diagram for Wigner crystallization in layered solids



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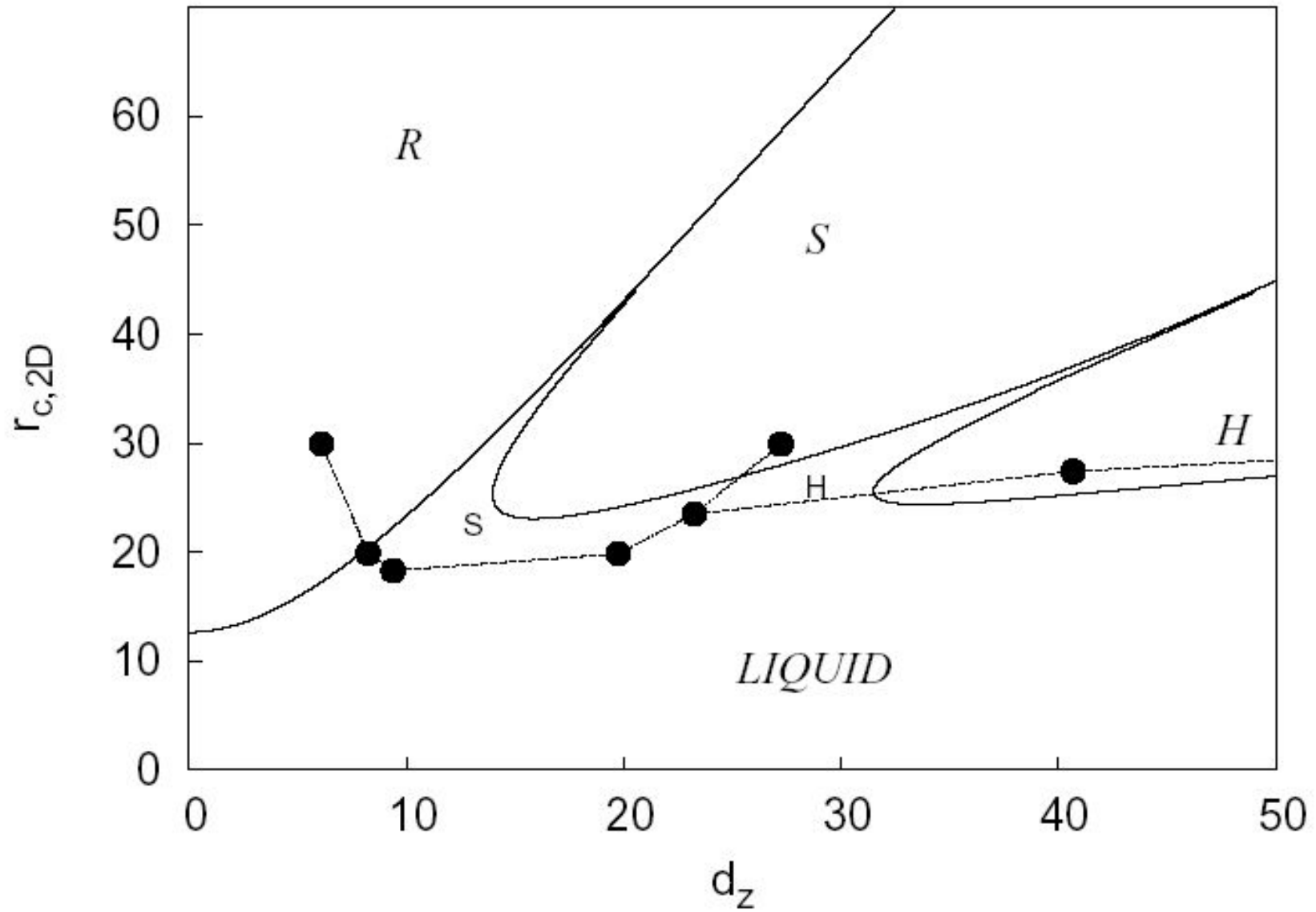
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- structural transitions: compare energies
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Conclusions

- in solids, the concept of Wigner crystallization must be generalized to account for a number of competing interactions
- in low dimensional compounds, the constraints on the electron motion induced by the anisotropy of the transfer integrals can stabilize the Wigner crystal to much higher densities than in vacuum
- could explain why “Wigner crystals” are “often” “observed” in low-dimensional materials

Check: bilayer heterostructures

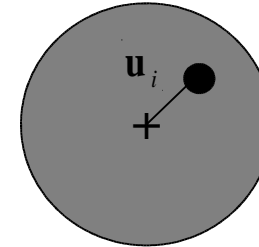


$$H = \sum_i H_i + \frac{1}{2} \sum_{i,j} H_{ij}$$

Mean Field (Wigner)

$$H_i = E_{\text{madelung}} + \frac{p_i^2}{2m} + \frac{1}{2} m \frac{\omega_P^2}{3} u_i^2$$

Neutral spheres do not interact, $\mathbf{E}=0$

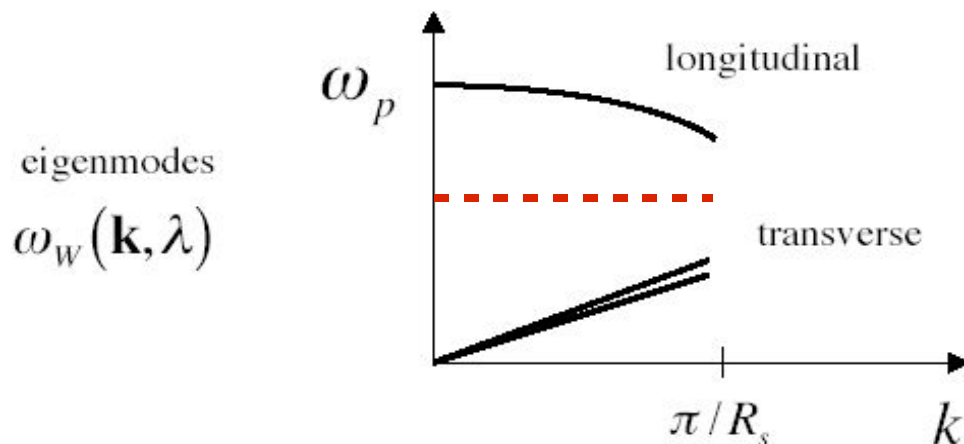
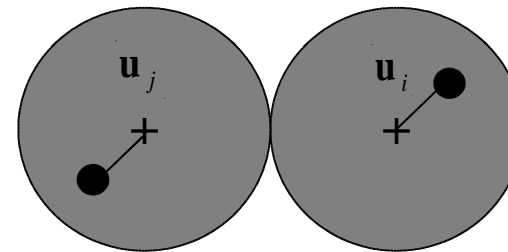


Beyond mean field

$$H_{ij} = \sum_{\alpha\beta} u_i^\alpha \Lambda_{ij}^{\alpha\beta} u_j^\beta$$

$$\Lambda_{ij}^{\alpha\beta} = e^2 \frac{R_{ij}^2 \delta_{\alpha\beta} - 3R_{ij}^\alpha R_{ij}^\beta}{R_{ij}^5}$$

Dipole-dipole interactions



Warning !! : not all crystallized structures are MECHANICALLY stable.

Ex: Simple Cubic $\omega_w(\mathbf{k}, \lambda)^2 < 0$

STM: hole crystallization?

Measure tunneling I vs. bias V with atomic resolution

=> conductance $dI/dV = g(r,V) \sim$ Local DOS

FT of local DOS => spots in reciprocal space

- q varies with bias => qp interference, $g(q,V) \sim A(k,V) A(k+q,V)$

- dispersionless q , $4a_0 \times 4a_0$ modulations, 30-80 Å corr. length

=> charge ordering

Several interpretations (crystal of superconducting pairs, condensate with spatial modulations, stripes, Wigner crystal of holes...)

A Wigner crystal with periodicity 4×4 can be stabilized provided that the holes are subject to an additional localizing effect whose characteristic energy scale is ~ 150 meV, the same that is observed in $\sigma(\omega)$ candidates => polaron energy E_p , exchange energy

References

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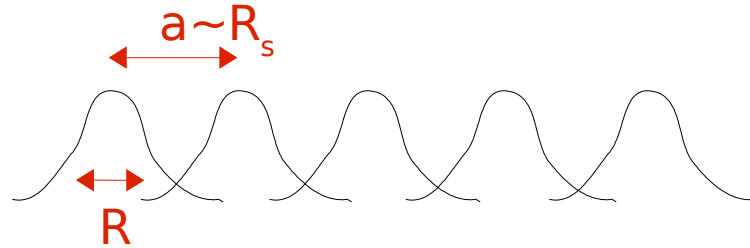
[G.R., S.F. & P.Q.: Eur. Phys. Jour. B, 42, 305 \(2004\)](#)
on the modulations observed by STM

G.R and S.C.: Phys. Rev. B 71, 184303 (2005)

G.R. and S.F.: in preparation
on the stabilization of a WC in low-dimensional solids

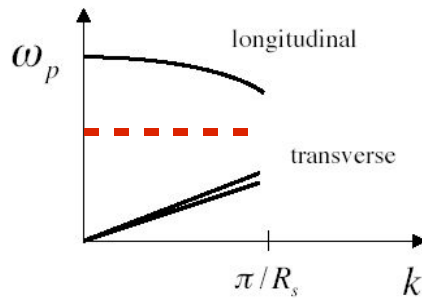
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$$\frac{R}{a} > \delta$$

eigenmodes
 $\omega_W(\mathbf{k}, \lambda)$

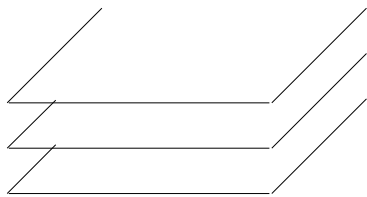


BCC Wigner crystal in 3D,
radius R related to
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$$\omega_W = \omega_p / \sqrt{3} \sim r_s^{-3/2}$$

$$R = \sqrt{\frac{3\hbar}{2m\omega_W}} \sim r_s^{3/4}$$

$$\frac{R}{a} \sim \frac{r_s^{3/4}}{r_s} = \delta \longrightarrow r_s^c \sim 100$$



Same WC structure,
now with a layered
constraint:
out of plane fluctuations
are forbidden

$$R = \sqrt{\frac{2\hbar}{2m\omega_W}}$$

$$r_s^c(\text{layers}) \simeq \frac{4}{9} r_s^c(3D) \sim 45$$

$$r_s^c(\text{chains}) \simeq \frac{1}{9} r_s^c(3D) \sim 11$$