Self assembly of binary hard discs : towards quasicrystals

Etienne FAYEN, PhD student at Laboratoire de Physique des Solides

Outline

- Our system : binary hard discs mixtures

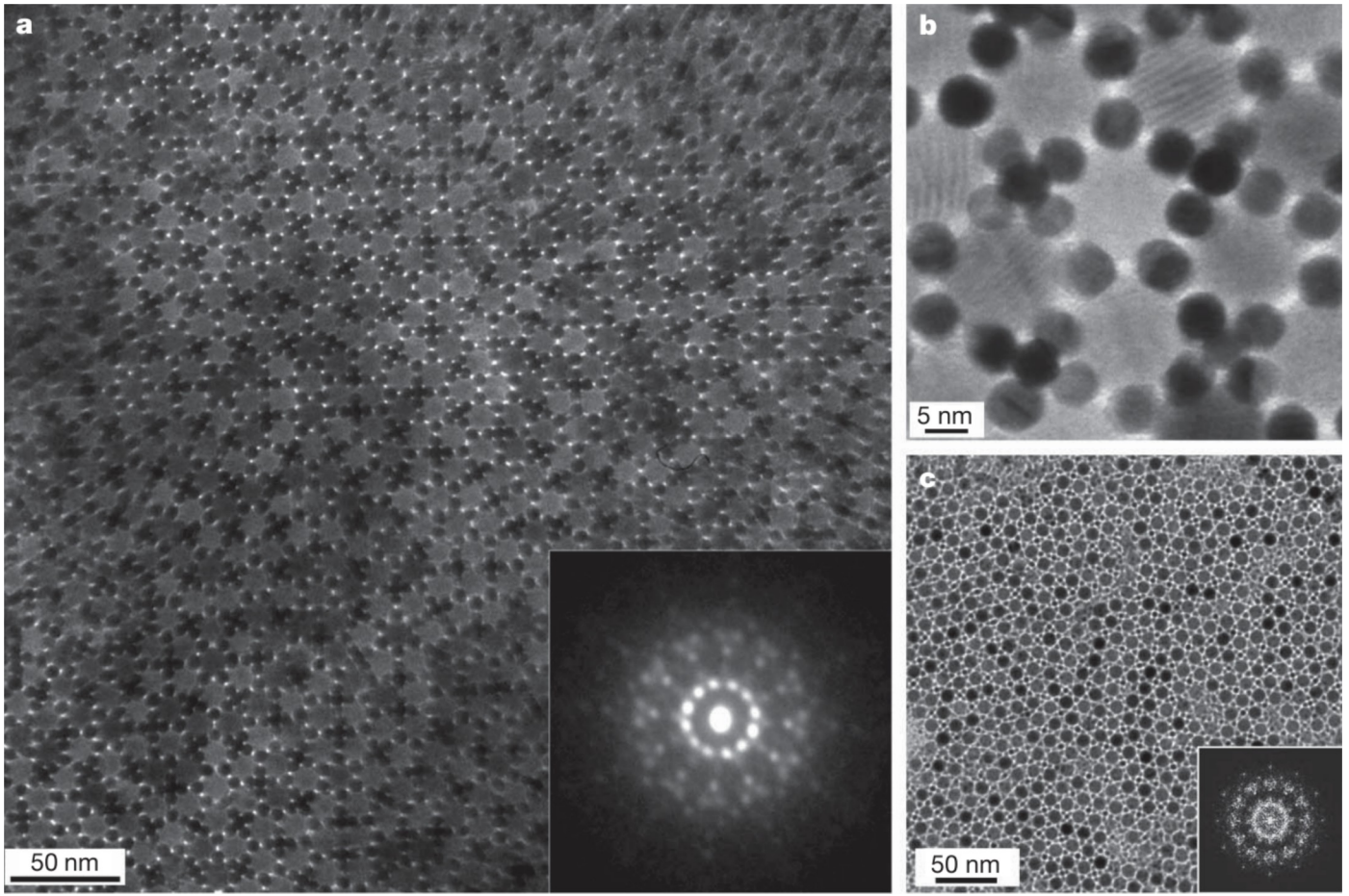
- How to build the phase diagram ?

- Floppy Box Monte Carlo simulations

- Relevance to quasicrystals

- Extension to non-additive hard discs

Why are there stable QC ?



Nanoparticles self-assembly

Talapin et al., NatLett, **461**, 964-967, 2009

Why are there stable QC ?

The SoftQC ANR team

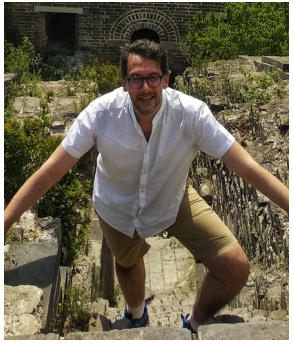
Anuradha Jagannathan
(Theory)

Benjamin Abécassis
(NP synthesis/assembly)

Brigitte Pansu
(NP synthesis/assembly)

Chiara Moretti
(NP synthesis/assembly)

Claire Goldmann
(NP synthesis)



Giuseppe Foffi
(Theory/simulations)

Jean-François Sadoc
(Theory)

Marianne Impéror
(X-rays)

Frank Smallenburg
(Theory/simulations)

Etienne Fayen
(Theory/simulations)

Why are there stable QC ?

Quasiperiodicity seems favored by interplay of length scales

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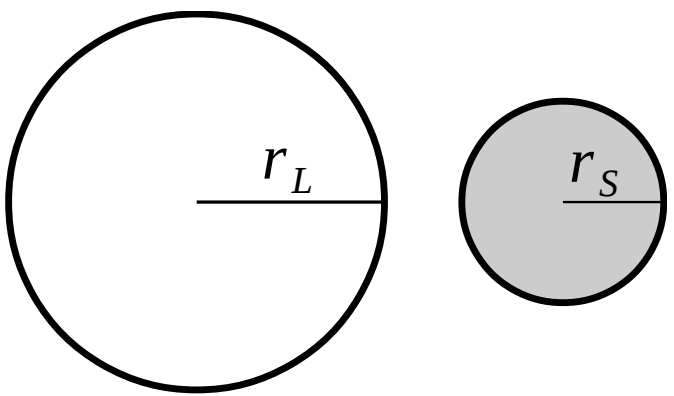
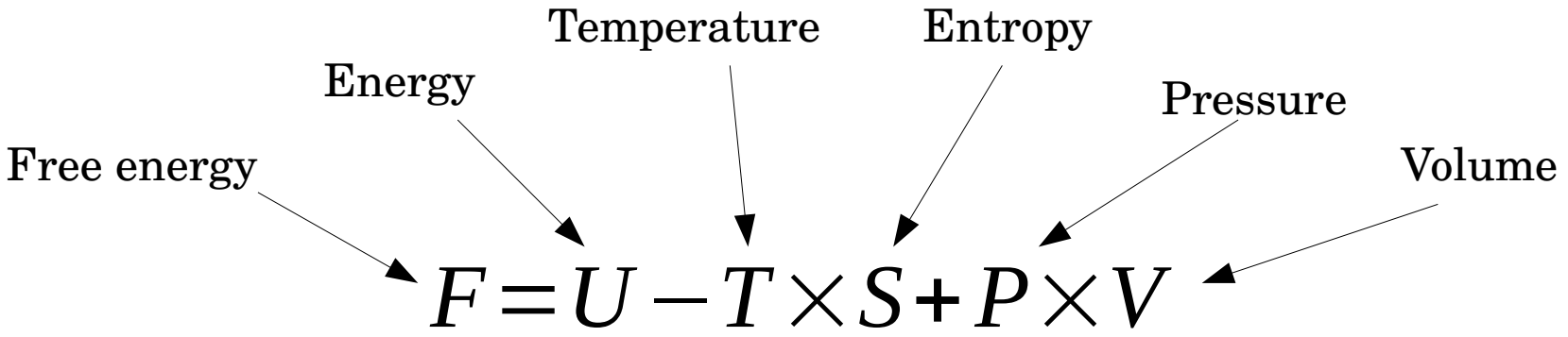
Binary hard-disc mixtures



Size ratio : $q = \frac{r_S}{r_L}$

Concentration of small discs : $p = \frac{n_S}{n_S + n_L}$

Why are there stable QC ?



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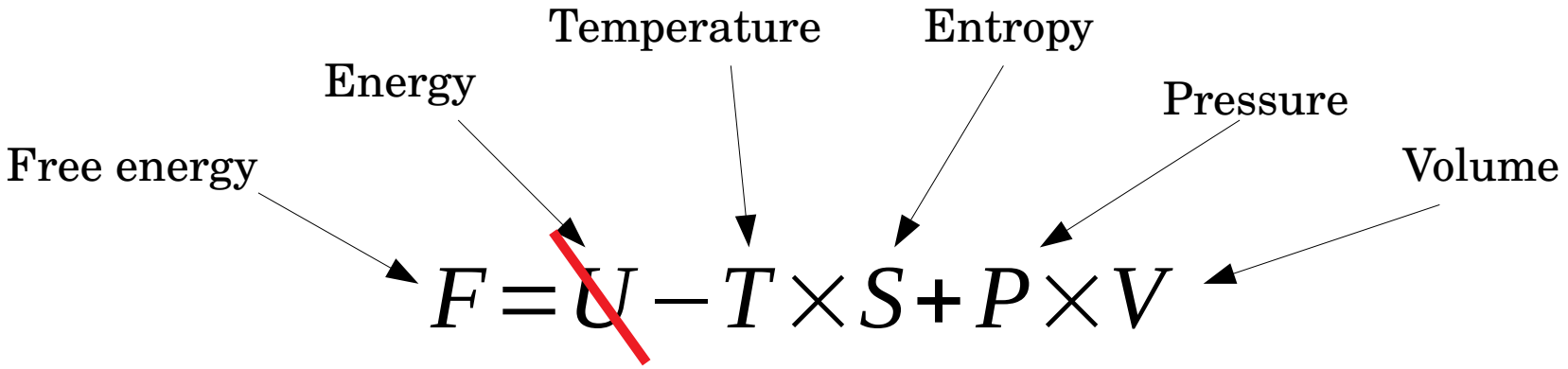
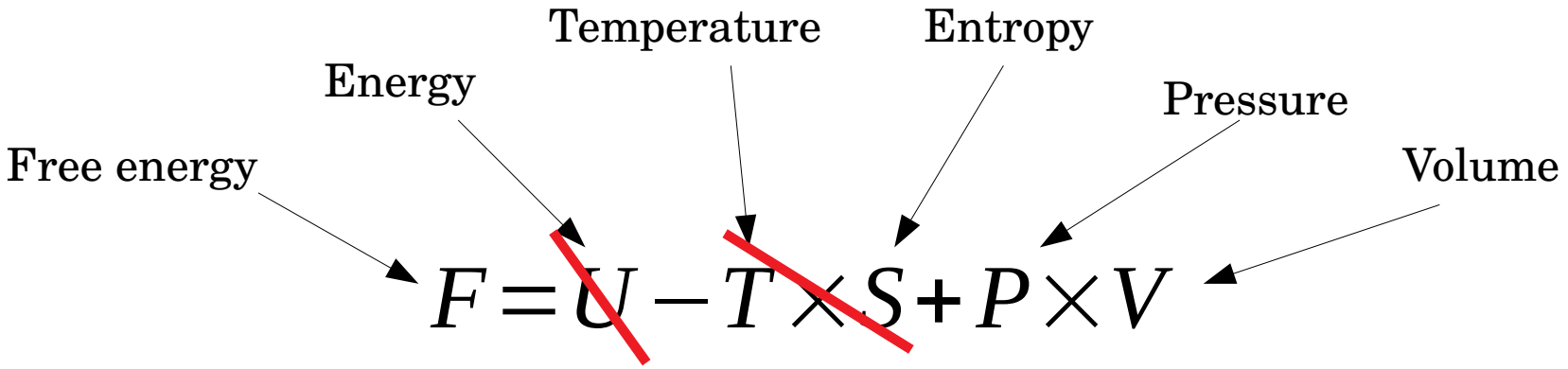


Hard interactions

Why are there stable QC ?

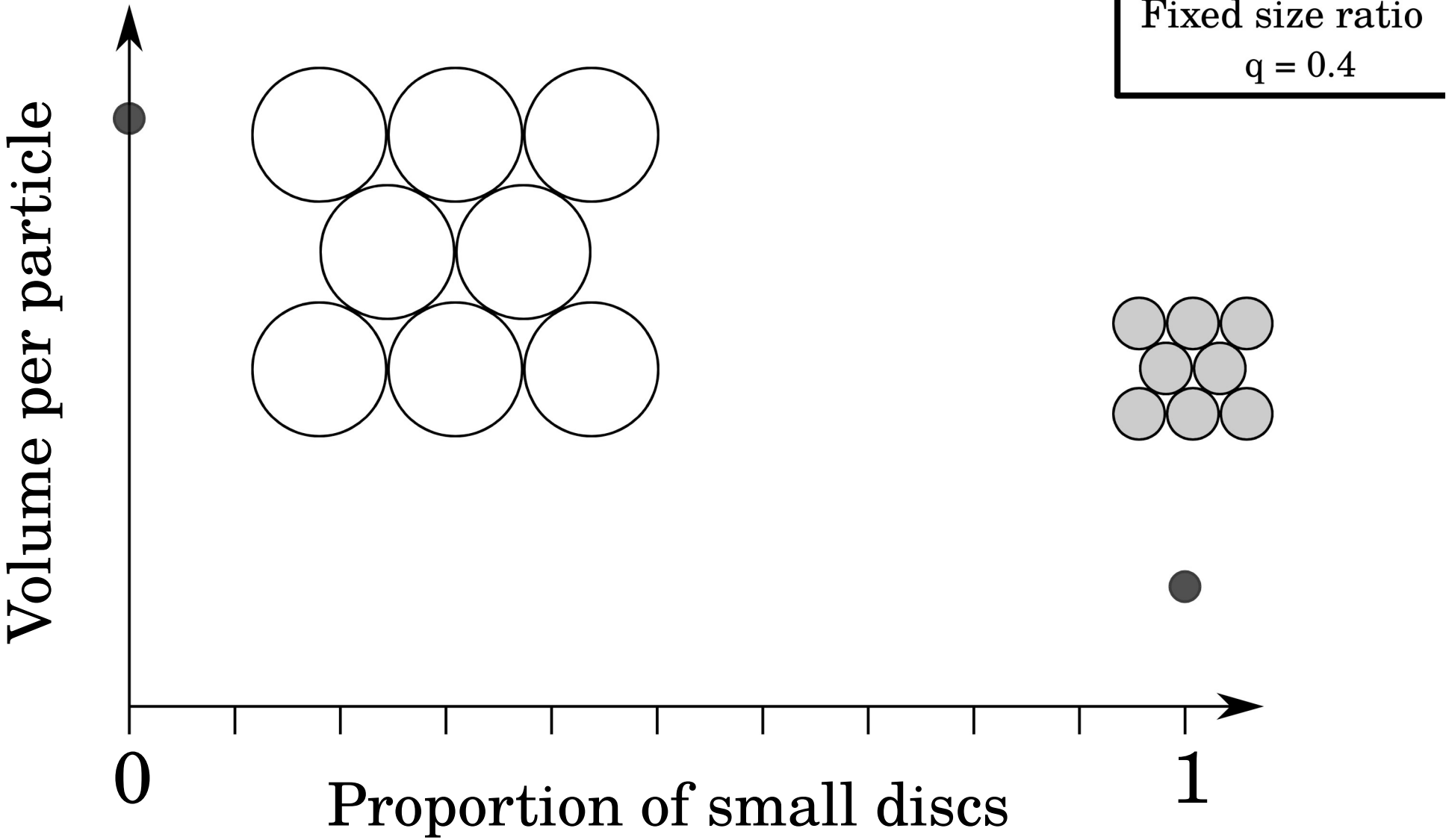


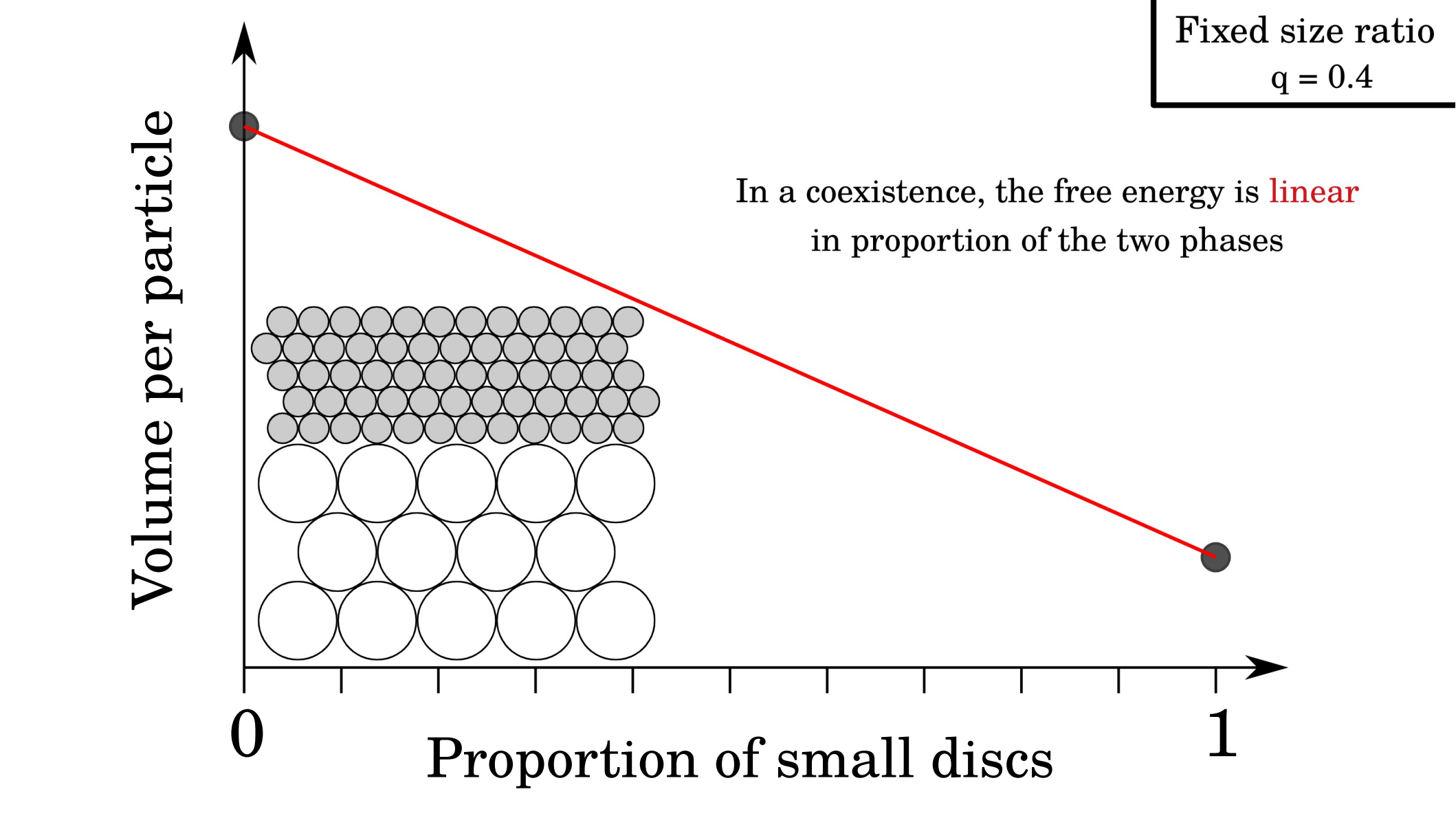
Hard interactions

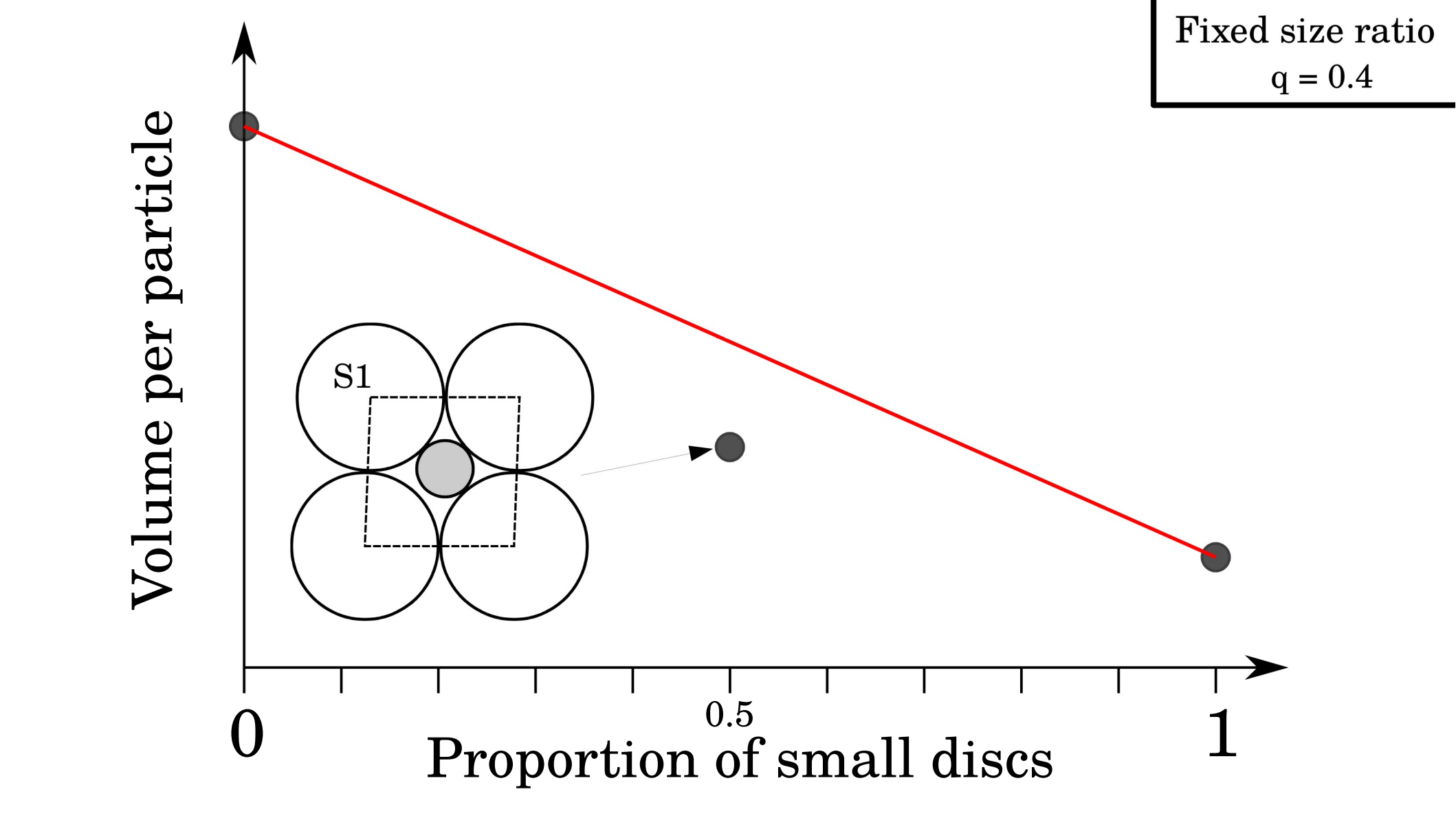
Infinite pressure limit

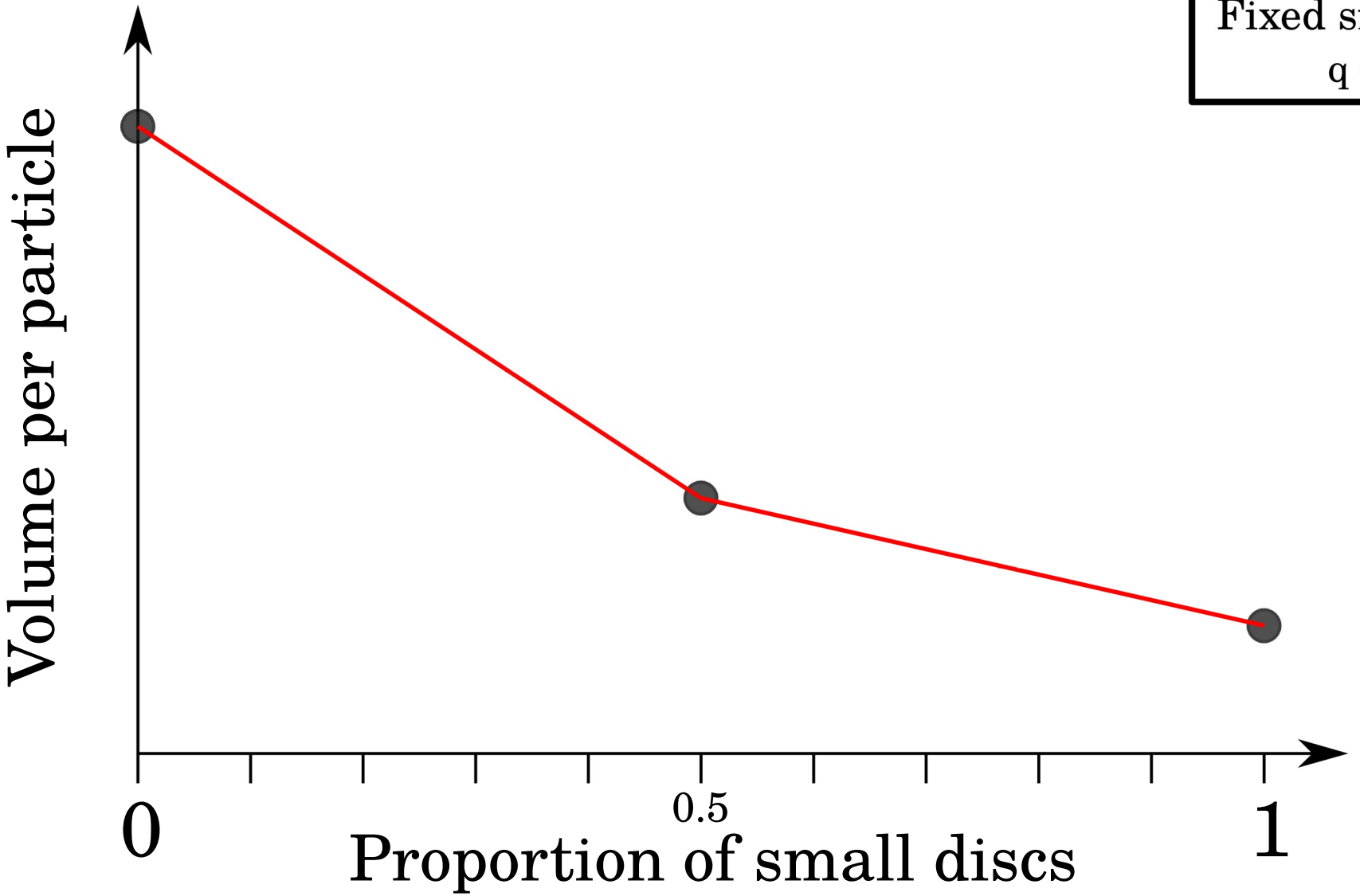
$$\frac{F}{N} = P \times \frac{V}{N}$$

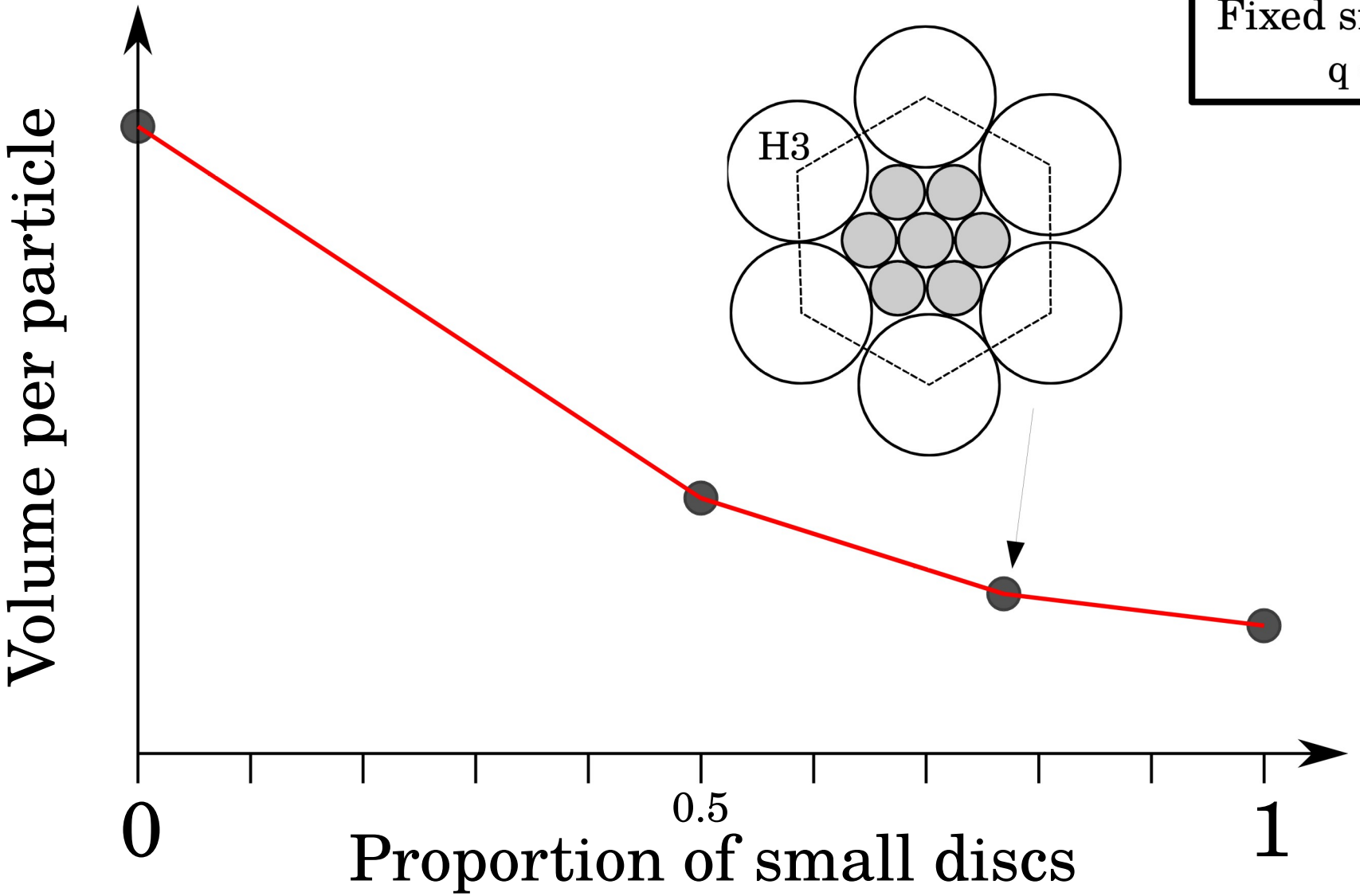
Stable means optimal packing

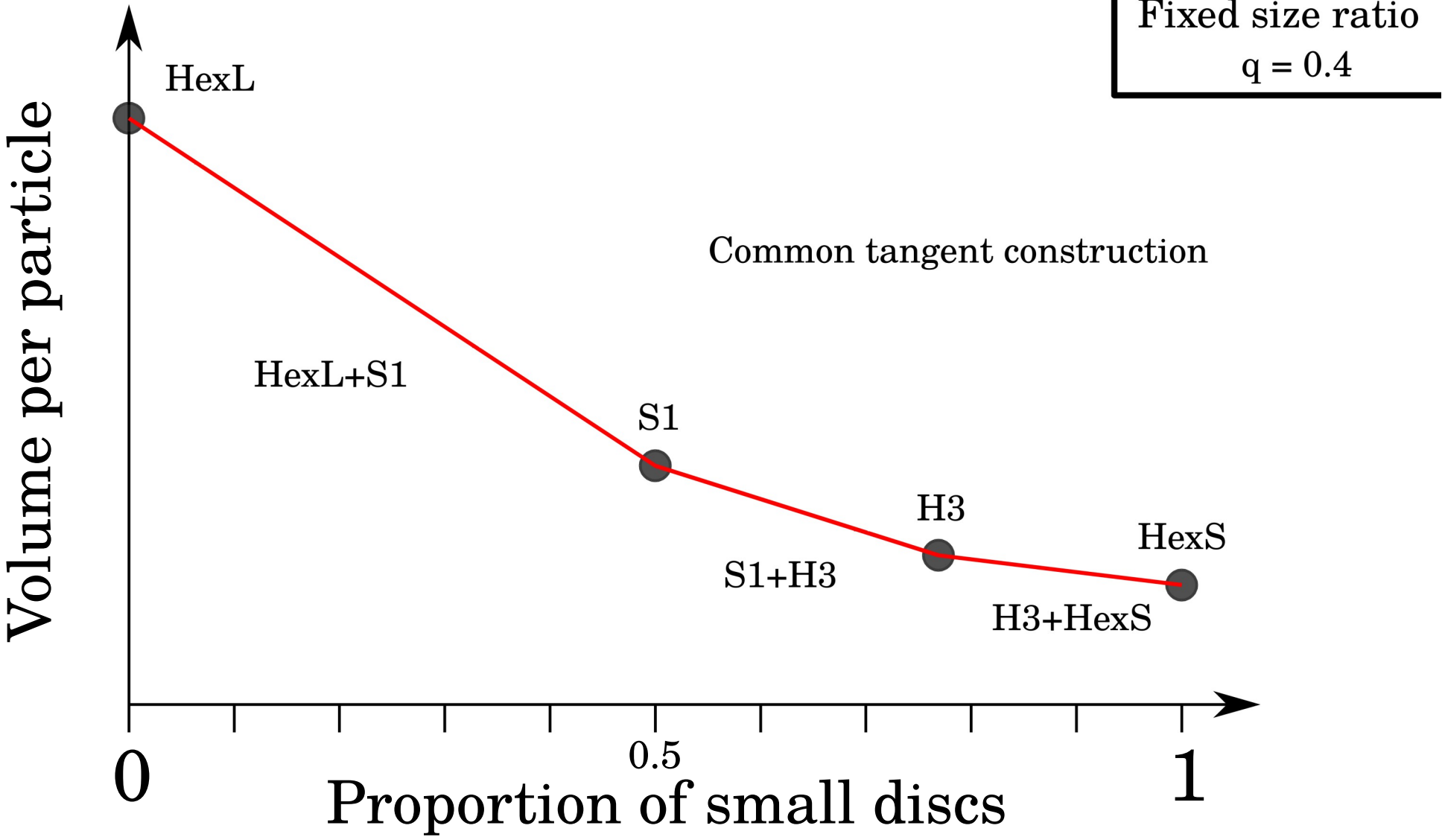

Fixed size ratio
q = 0.4
Volume per particle
0
1
Proportion of small discs

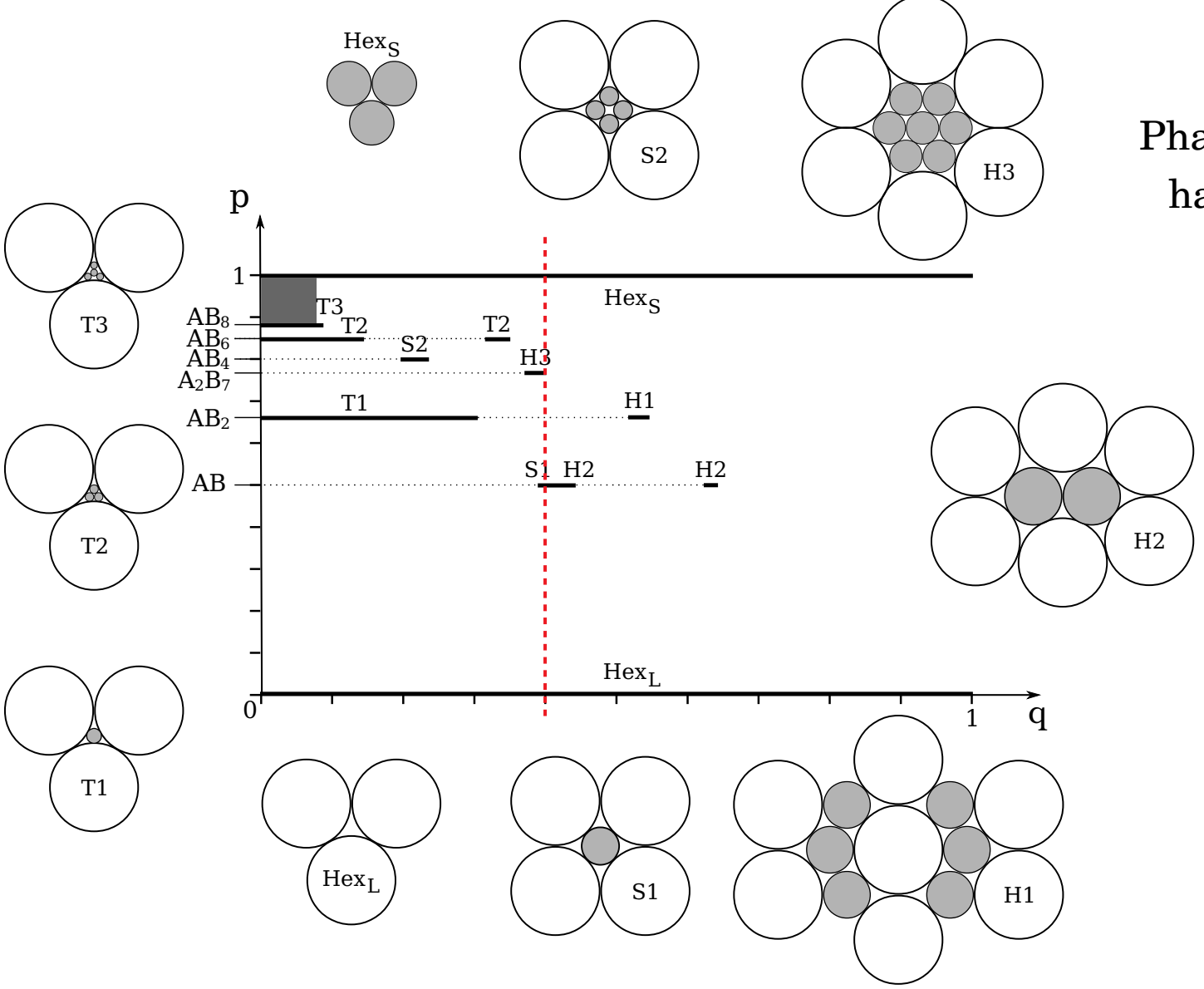

Fixed size ratio
q = 0.4
In a coexistence, the free energy is linear
in proportion of the two phases
Volume per particle
0
1
Proportion of small discs


Fixed size ratio
q = 0.4
Volume per particle
S1
0
0.5
1
Proportion of small discs


Fixed size ratio
q = 0.4
Volume per particle
0
0.5
1
Proportion of small discs

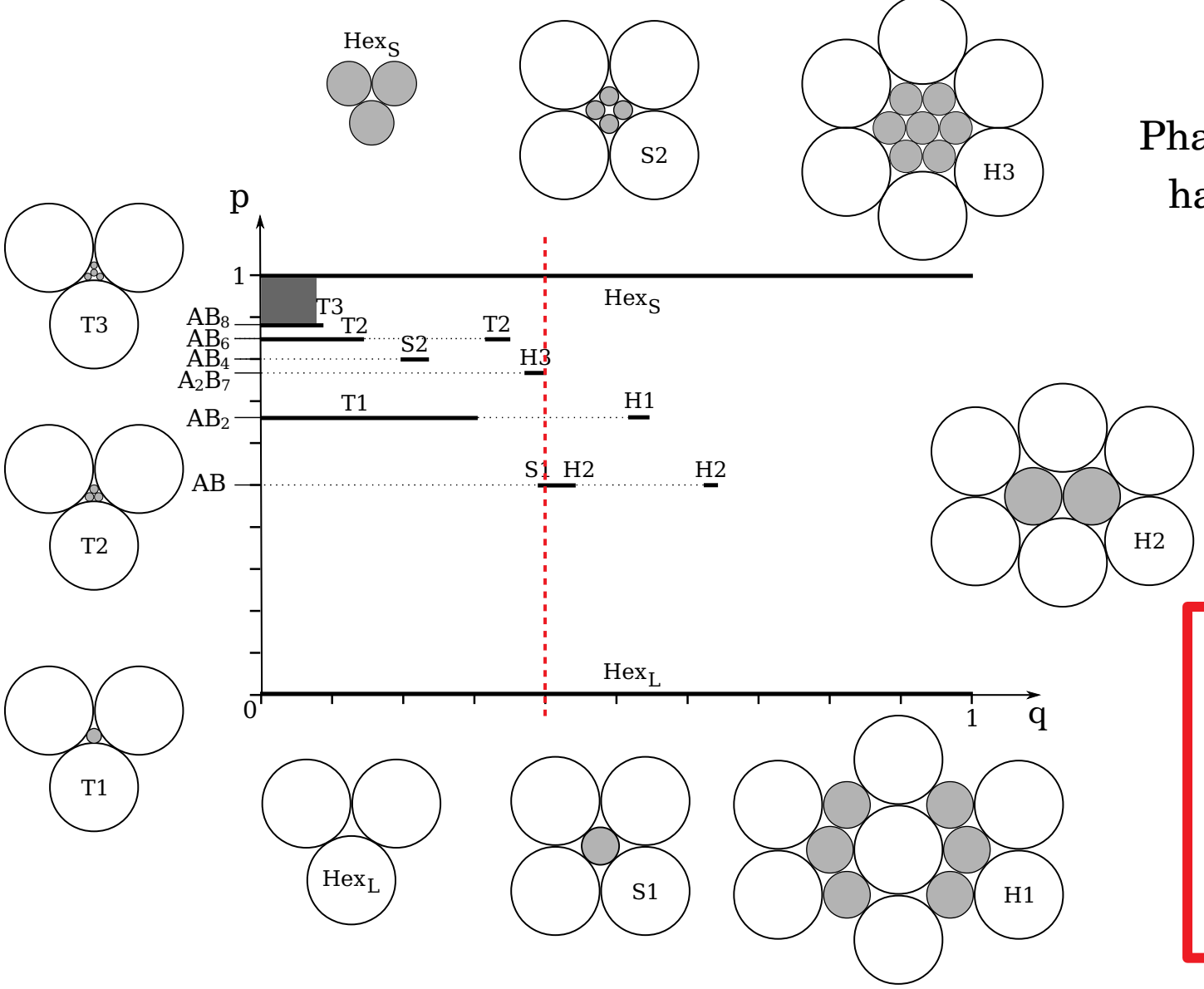

Fixed size ratio
q = 0.4
H3
Volume per particle
0
0.5
1
Proportion of small discs


Fixed size ratio
q = 0.4
HexL
Common tangent construction
HexL+S1
S1
H3
HexS
S1+H3
H3+HexS
Volume per particle
0
0.5
1
Proportion of small discs



Phase diagram for binary hard discs mixtures at infinite pressure

C.N. Likos, C.L. Henley,
PhiloMagB, **68**, 85-113, 1993



Phase diagram for binary hard discs mixtures at infinite pressure

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How can we sample candidate structures in a systematic way ?

Floppy Box Monte Carlo (FBMC) simulations

Simulate only the unit cell of a crystal (periodic BC)
Let particles position and box shape fluctuate

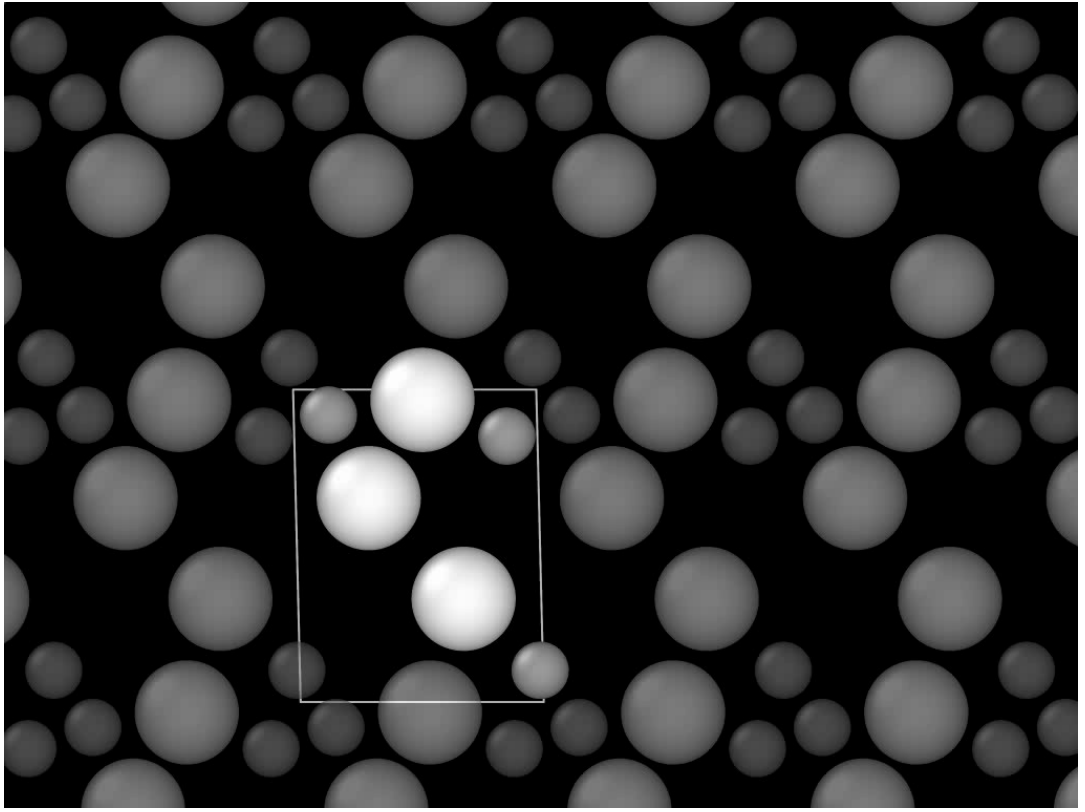
- Start from a dilute system
- Compress slowly
- Quench to infinite pressure
- Look at the resulting structure

Floppy Box Monte Carlo (FBMC) simulations

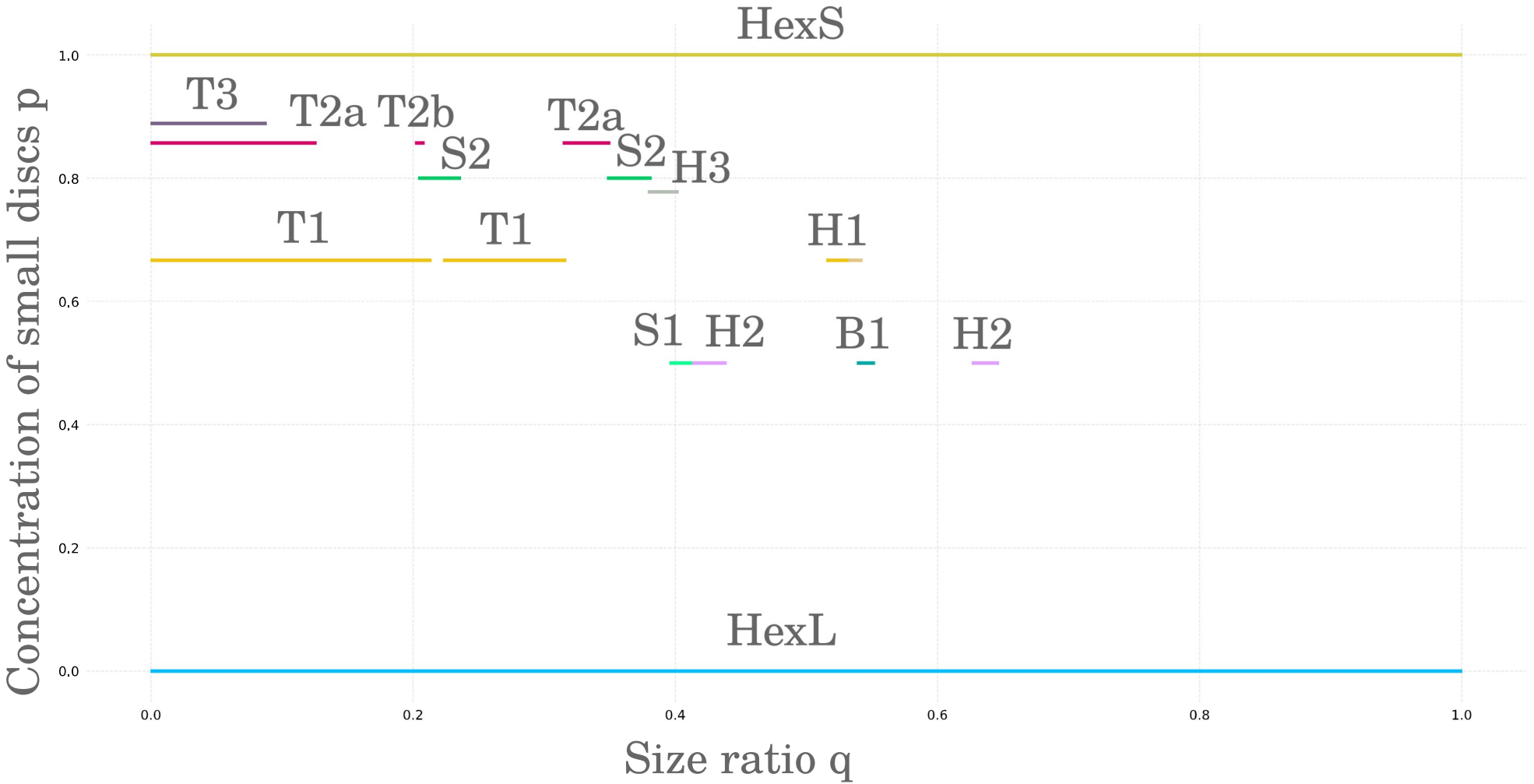
Simulate only the unit cell of a crystal (periodic BC)

Let particles position and box shape fluctuate

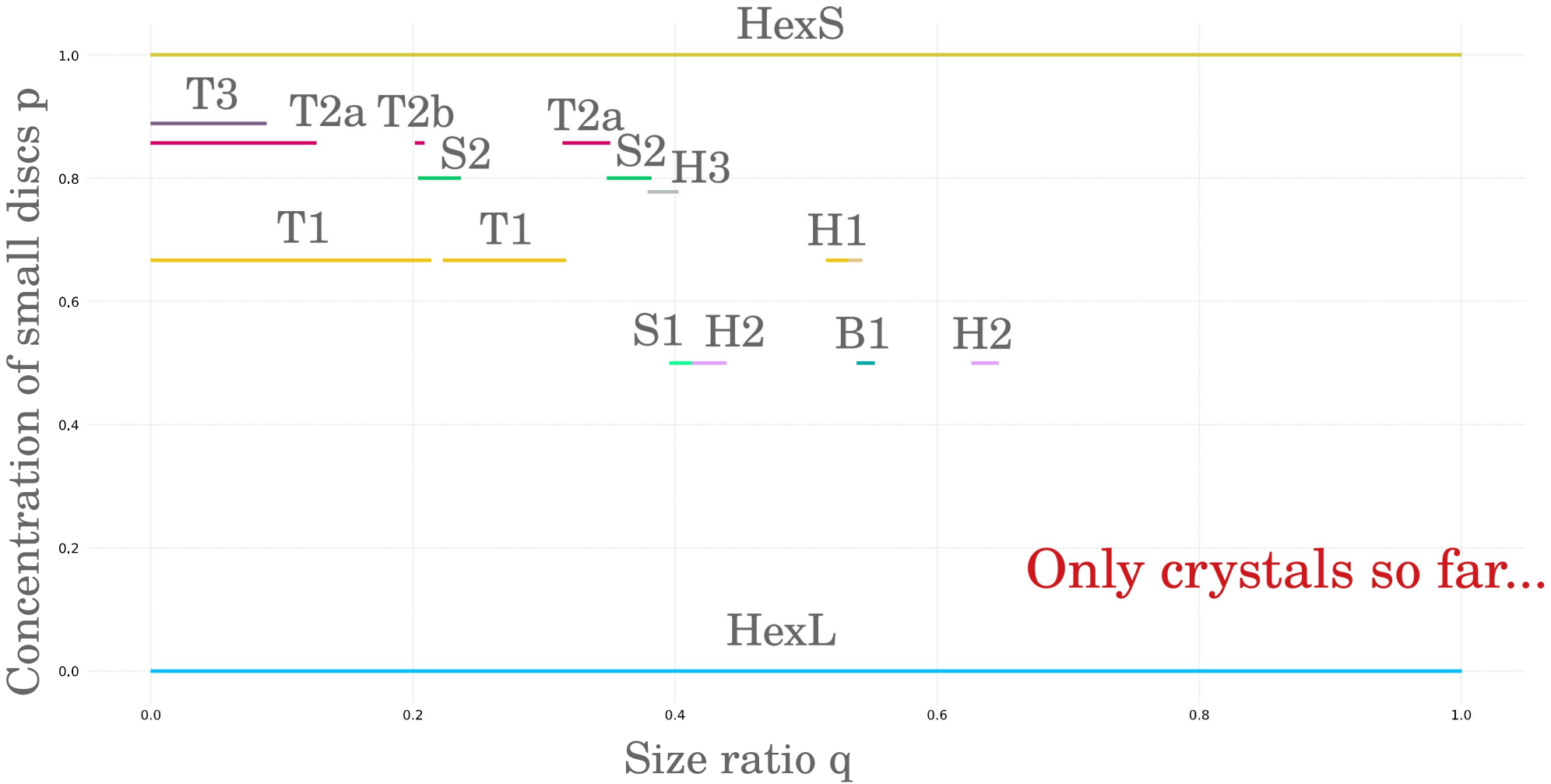
- Start from a dilute system
- Compress slowly
- Quench to infinite pressure
- Look at the resulting structure
- Start again !



Updated phase diagram of binary hard discs



Updated phase diagram of binary hard discs

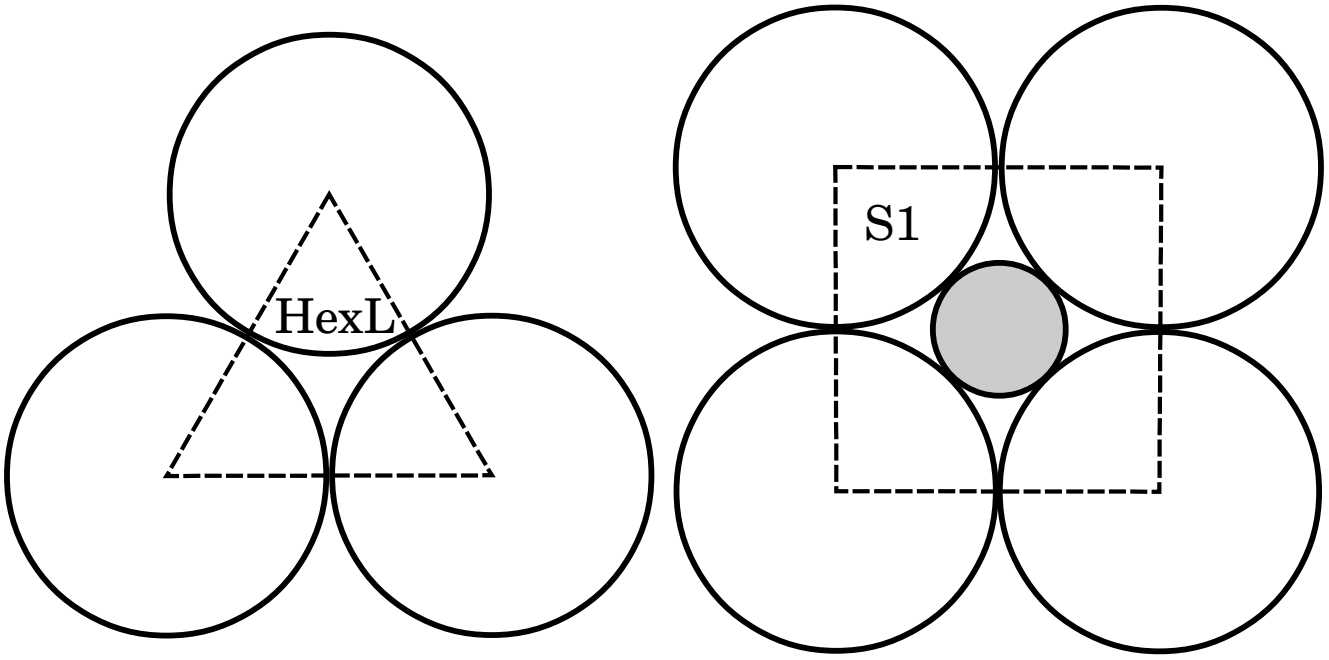


Relevance to quasicrystals

FBMC can only sample periodic structures

Rely on random tilings, ie. coexisting phases that :

- Have no boundary cost



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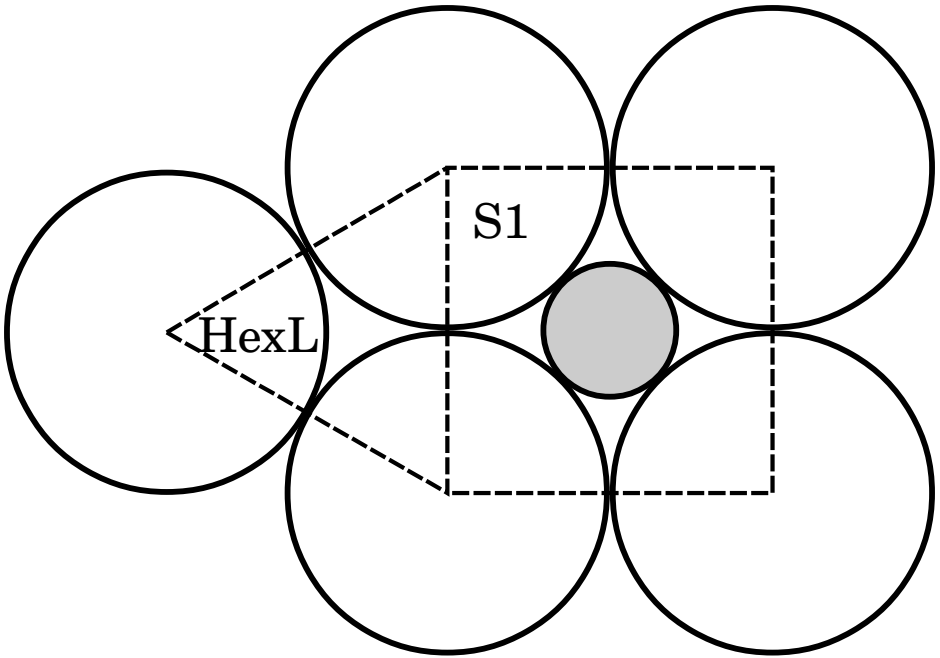
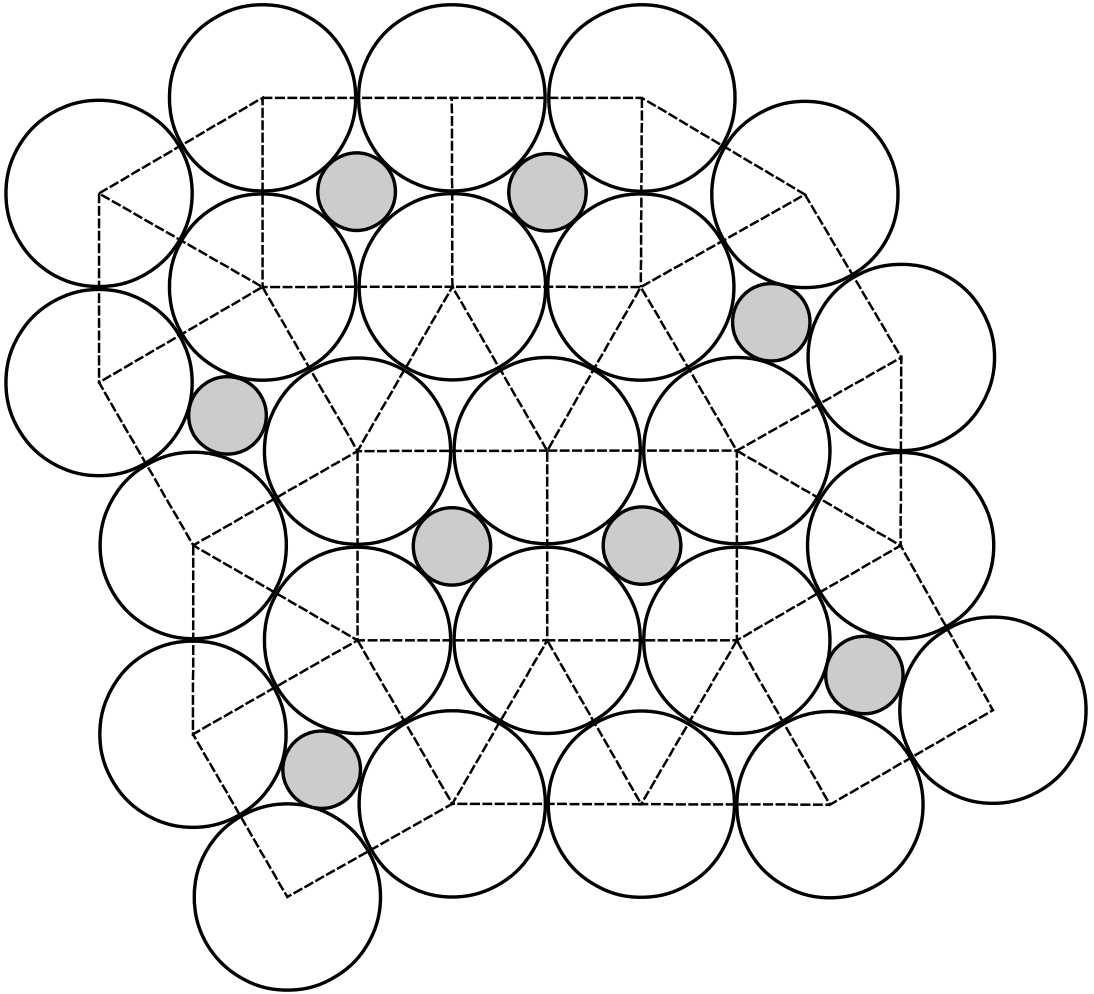


Relevance to quasicrystals

FBMC can only sample periodic structures

Rely on random tilings, ie. coexisting phases that :

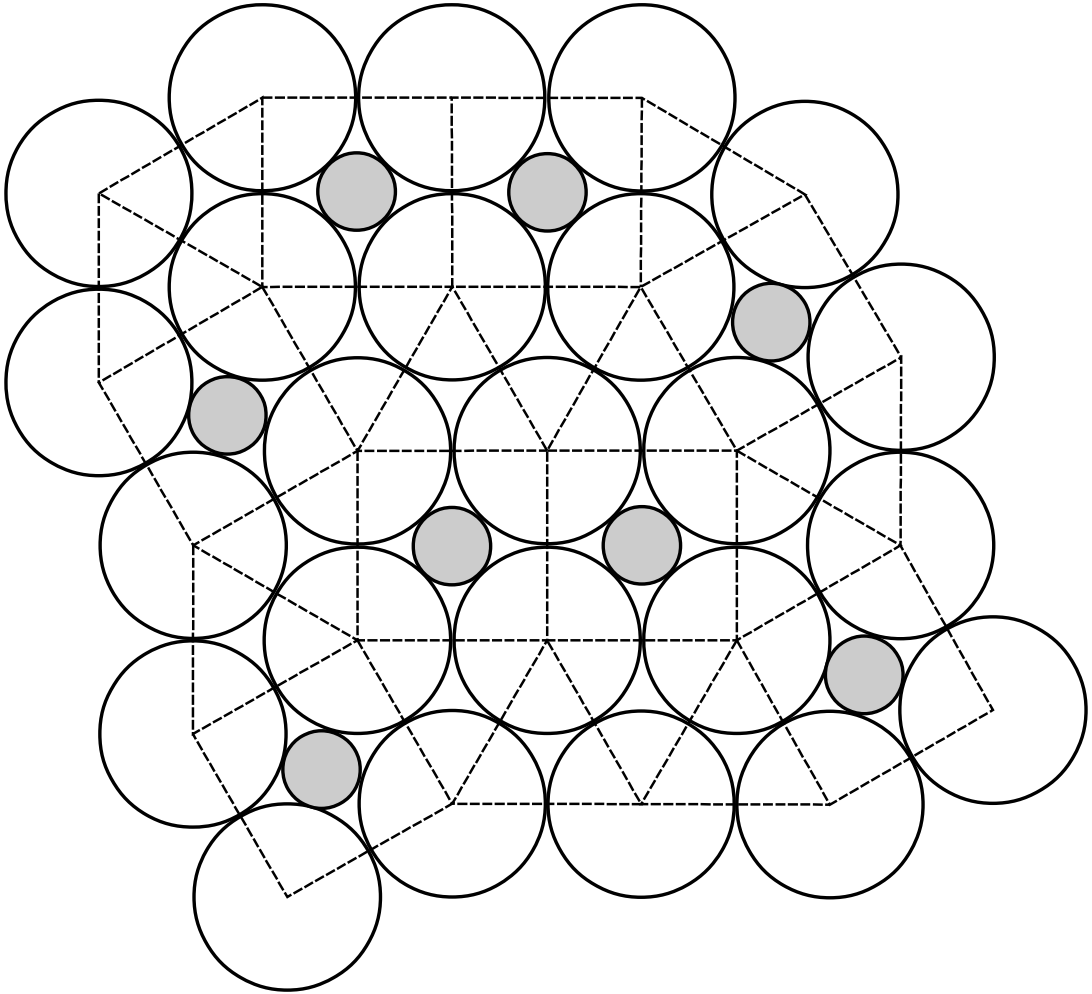
- Have no boundary cost
- Tile the plane



Relevance to quasicrystals

When $\frac{N_{square}}{N_{triangle}} = \frac{\sqrt{3}}{4}$

Random tiling quasicrystal



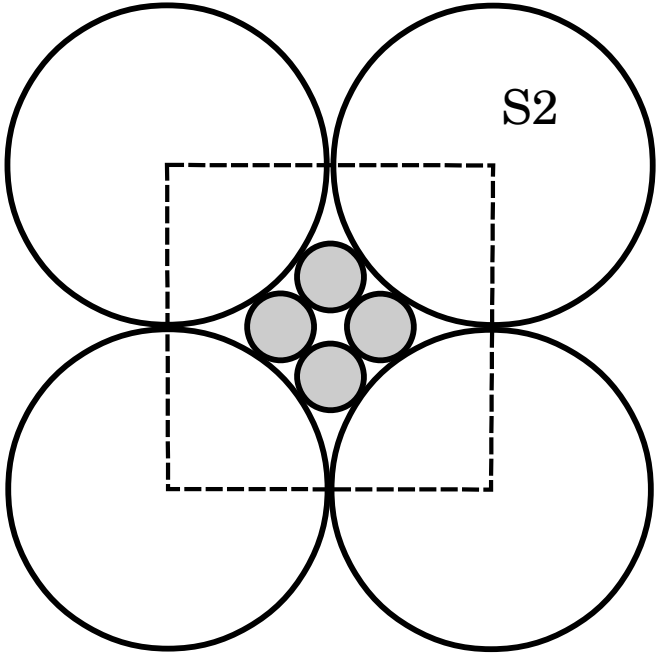
H. Kawamura, ProgThPhys, **70**, 1983

M. Oxborrow, C.L. Henley, PRB, **48**, 1993

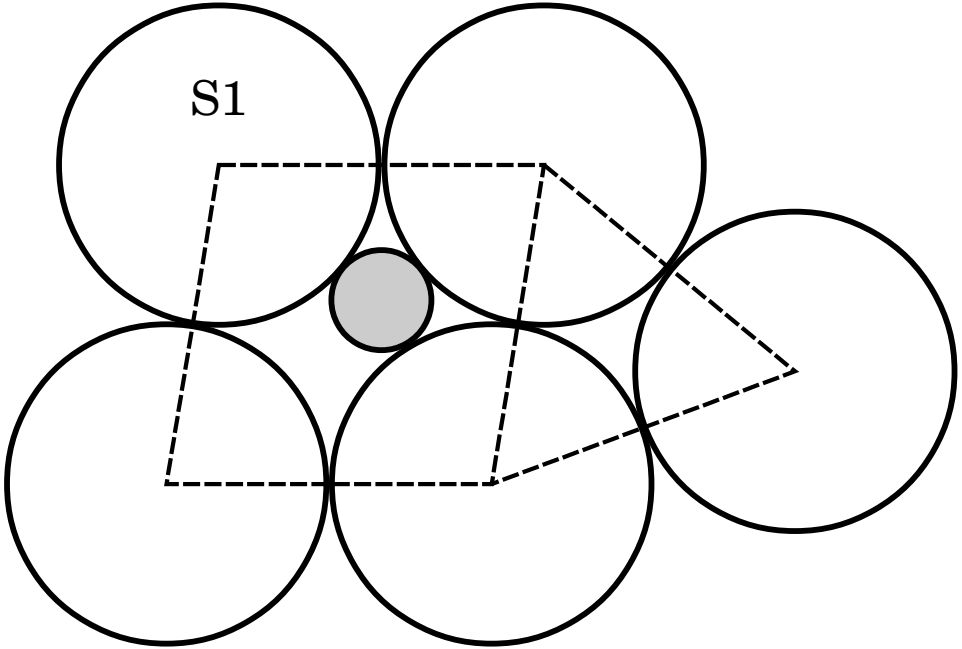
M. Widom, PRL, **70**, 1993

Relevance to quasicrystals

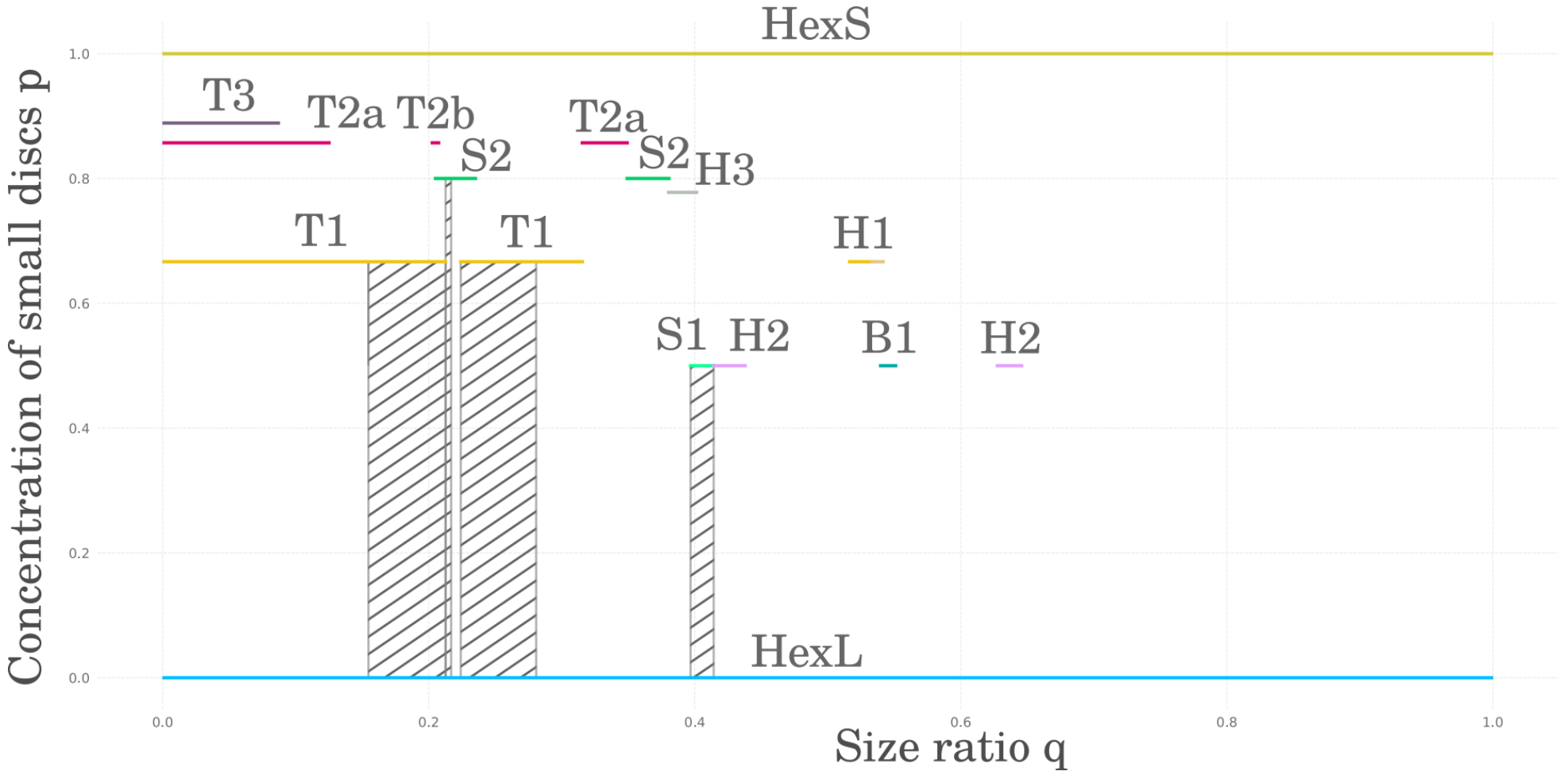
Works with other square cells:



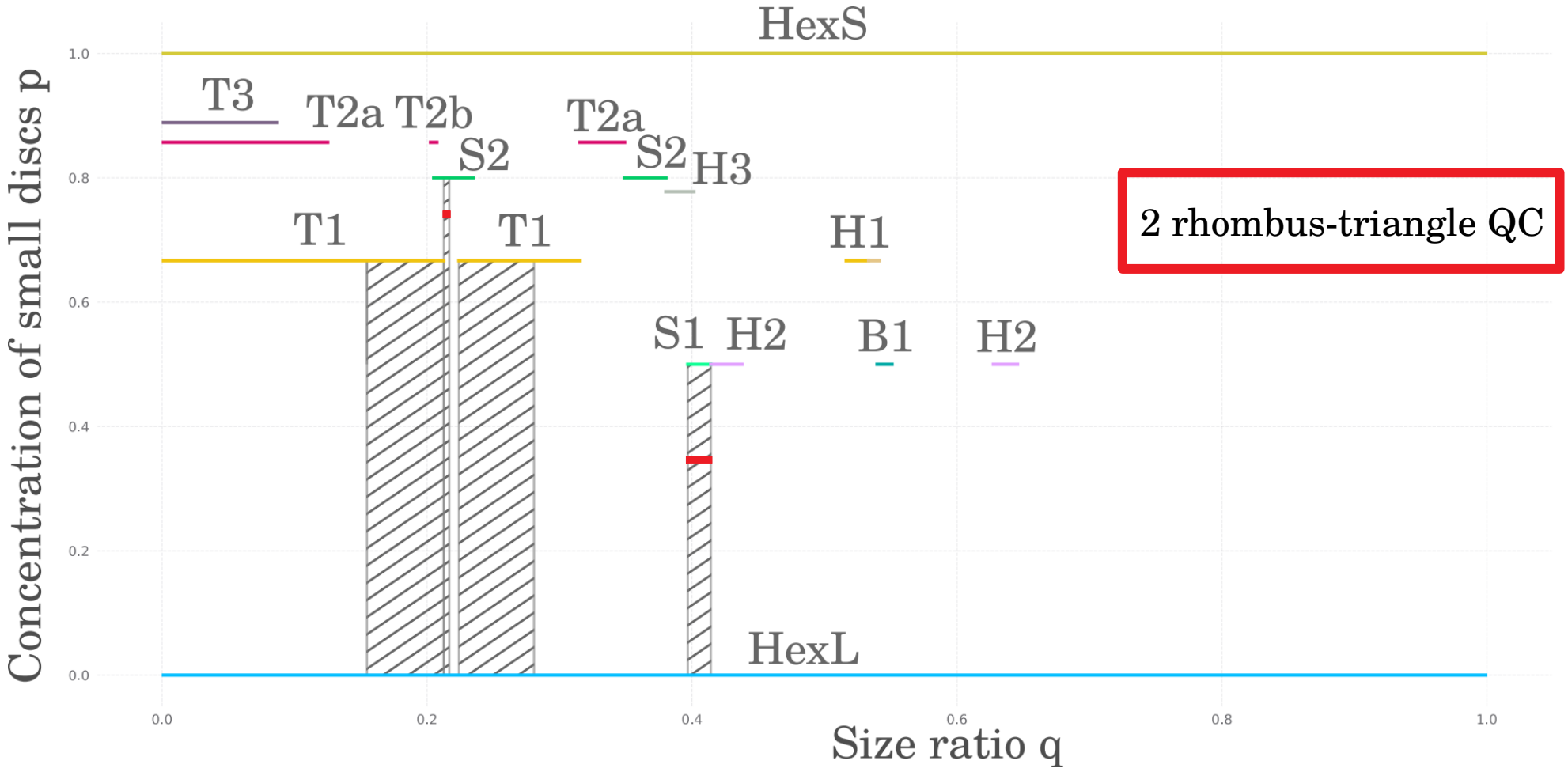
And also with rhombic cells:



Relevance to quasicrystals



Relevance to quasicrystals



Extension to non-additive hard discs

More interplaying length scales to favor quasiperiodic order ?

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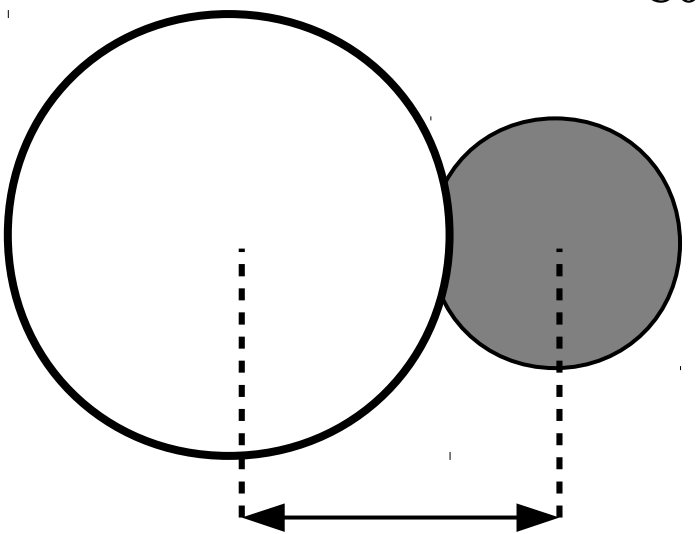
(top view)

Consider non-additive hard discs

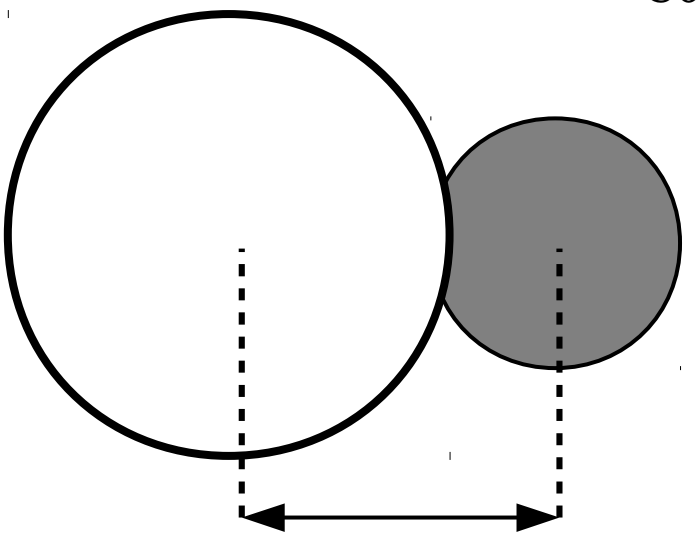


$$d_{LS} = (1+q) \times delta$$

Extension to non-additive hard discs

More interplaying length scales to favor quasiperiodic order ?

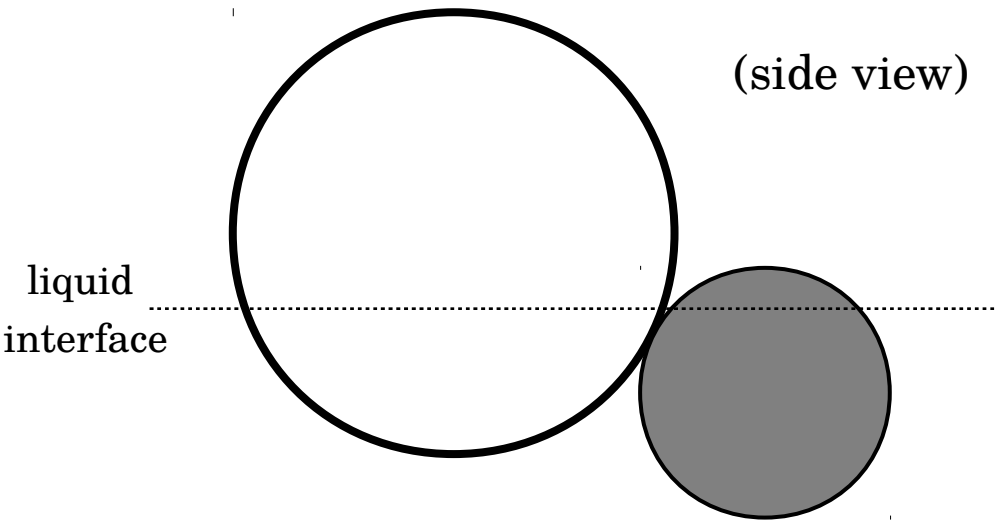
(top view)



$$d_{LS} = (1+q) \times delta$$

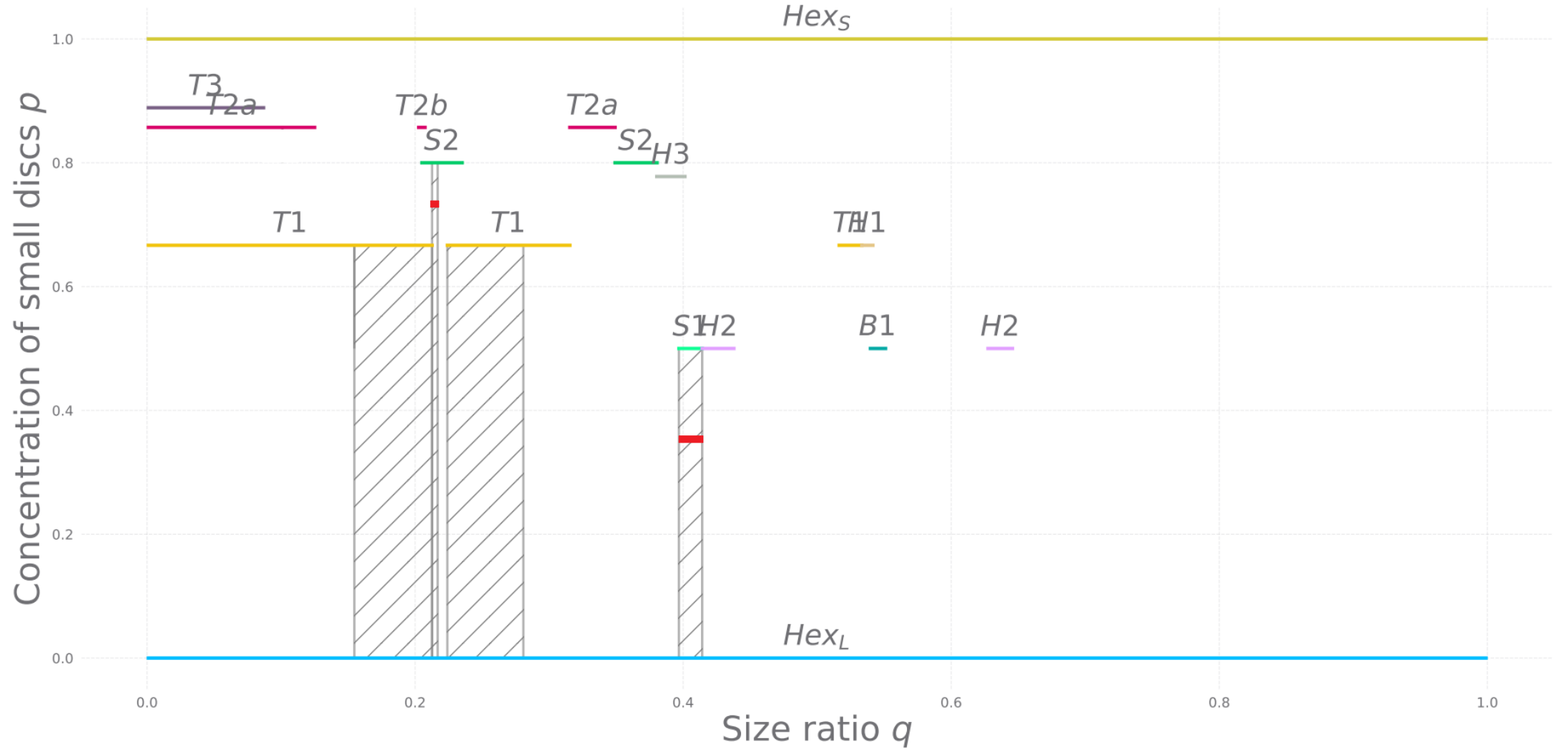
Consider non-additive hard discs

(side view)



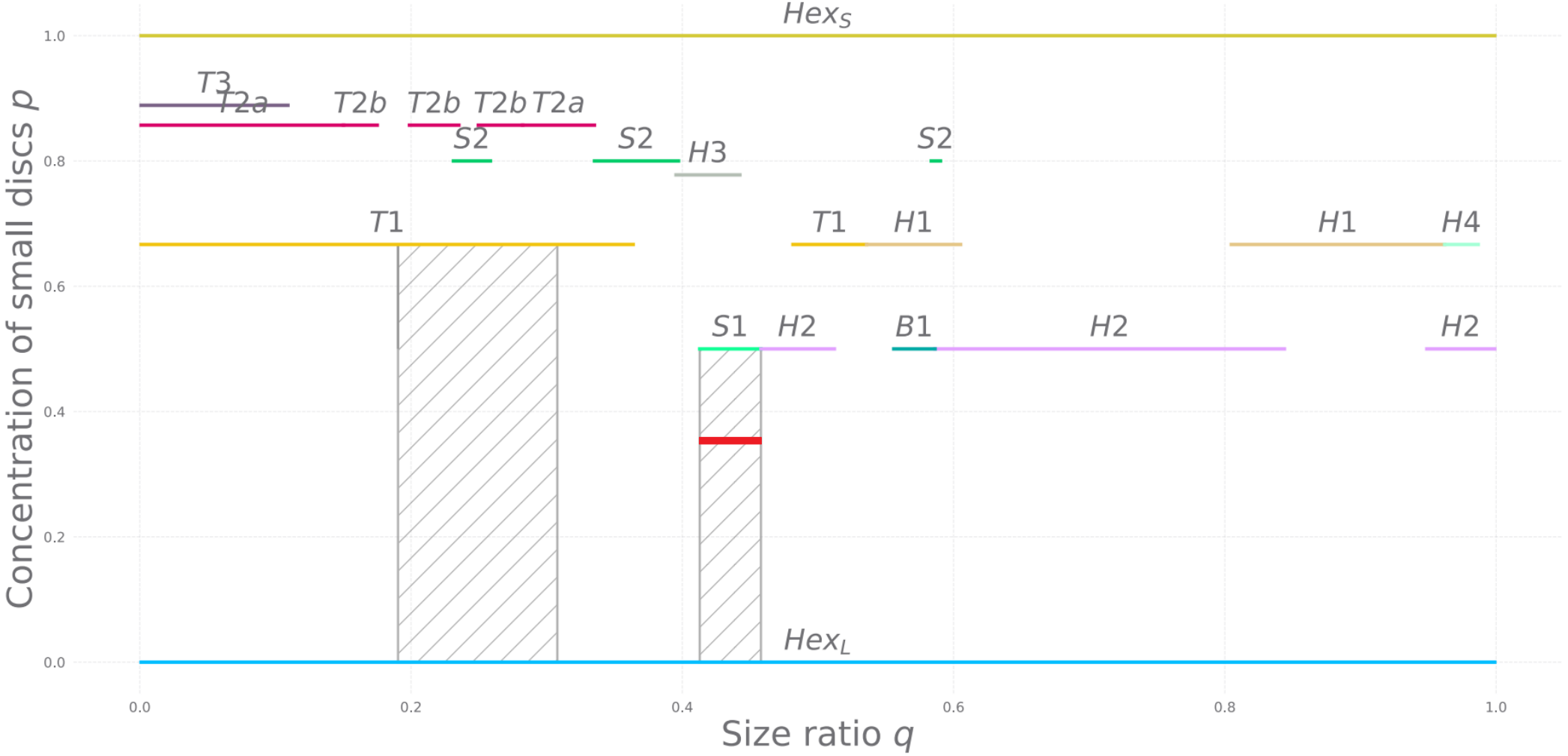
Delta = 1.00

Extension to non-additive hard discs



Delta = 0.97

Extension to non-additive hard discs



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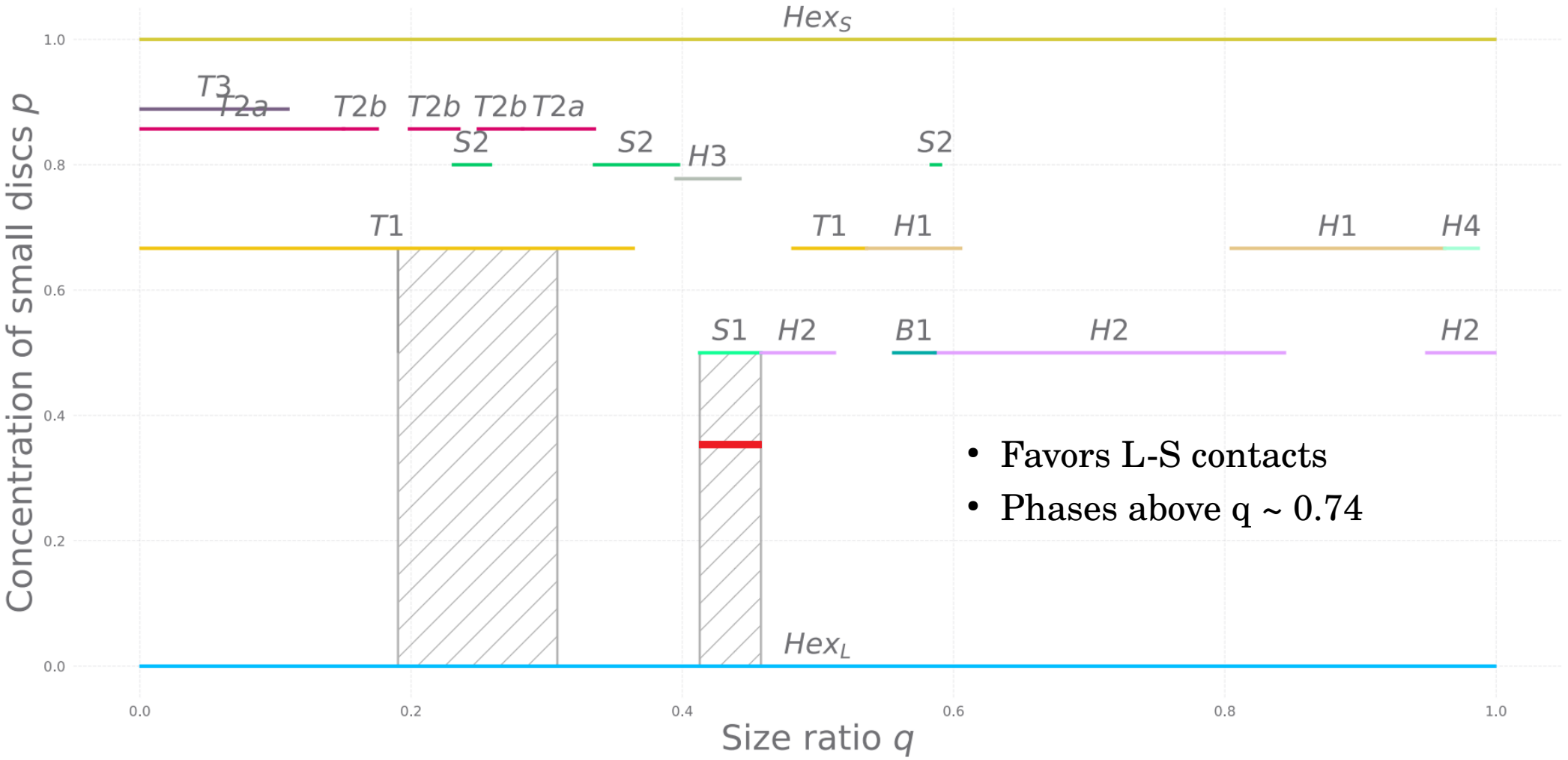
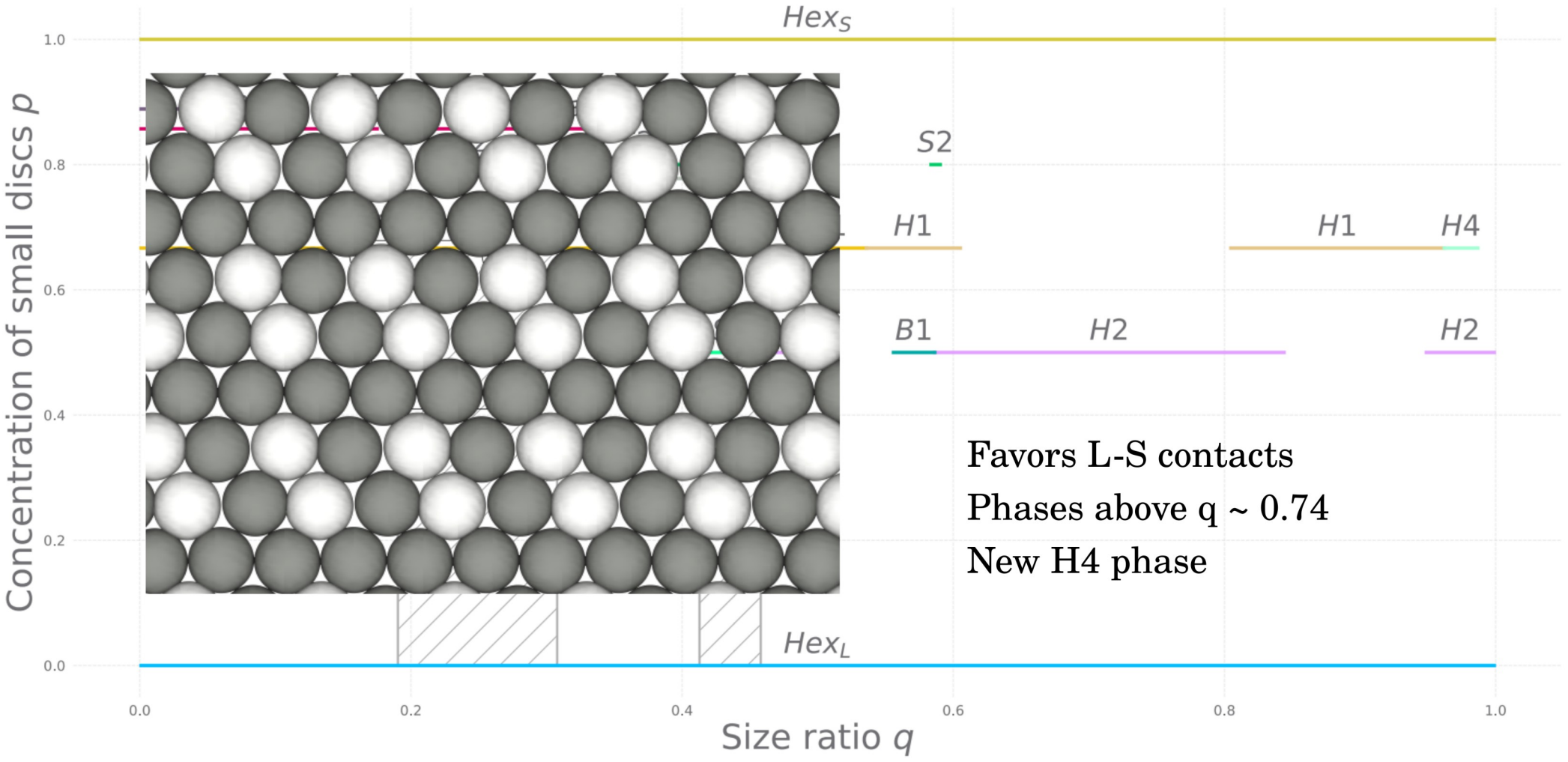
Extension to non-additive hard discs



- Favors L-S contacts
- Phases above q ~ 0.74

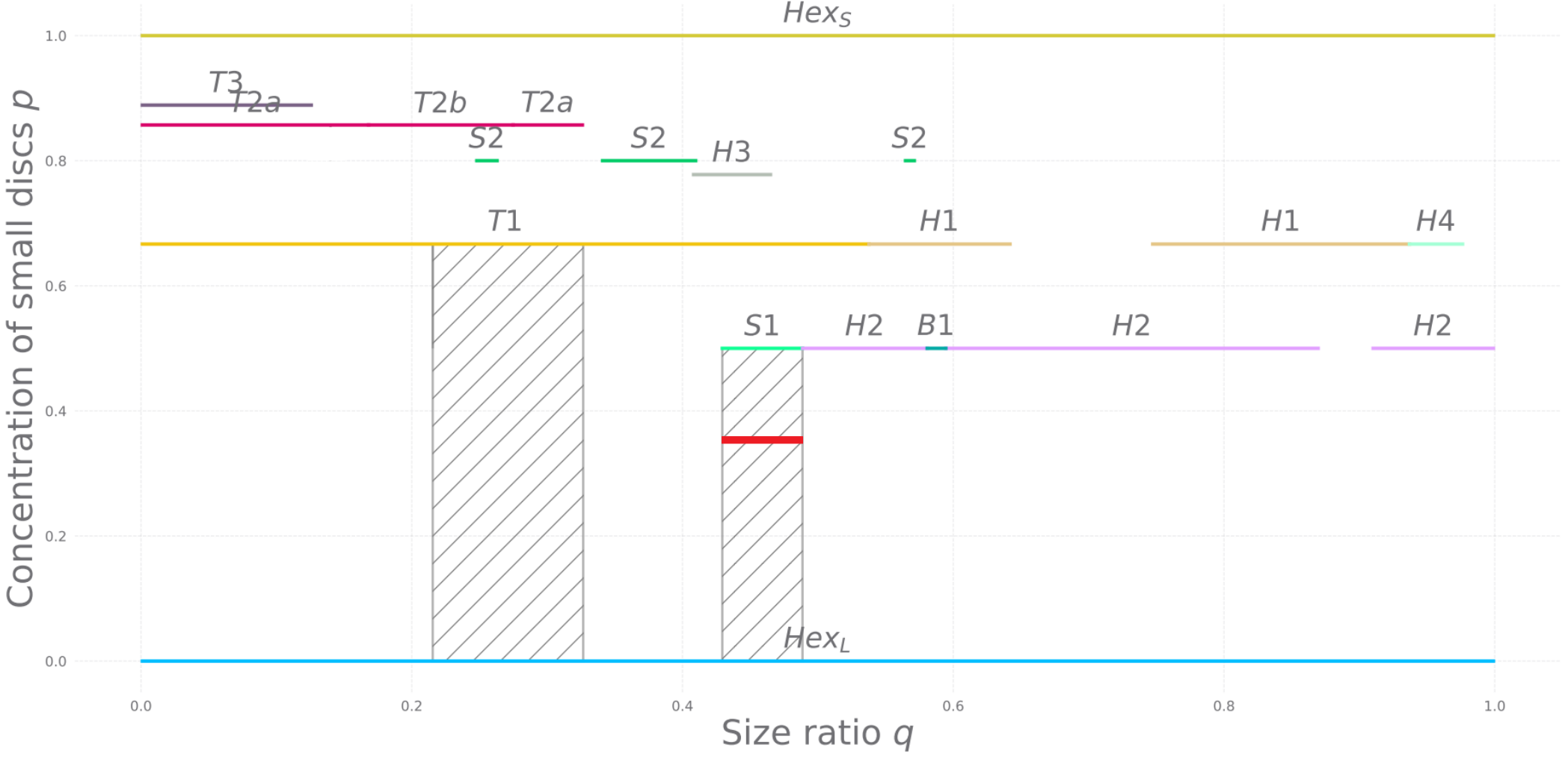
Delta = 0.97

Extension to non-additive hard discs



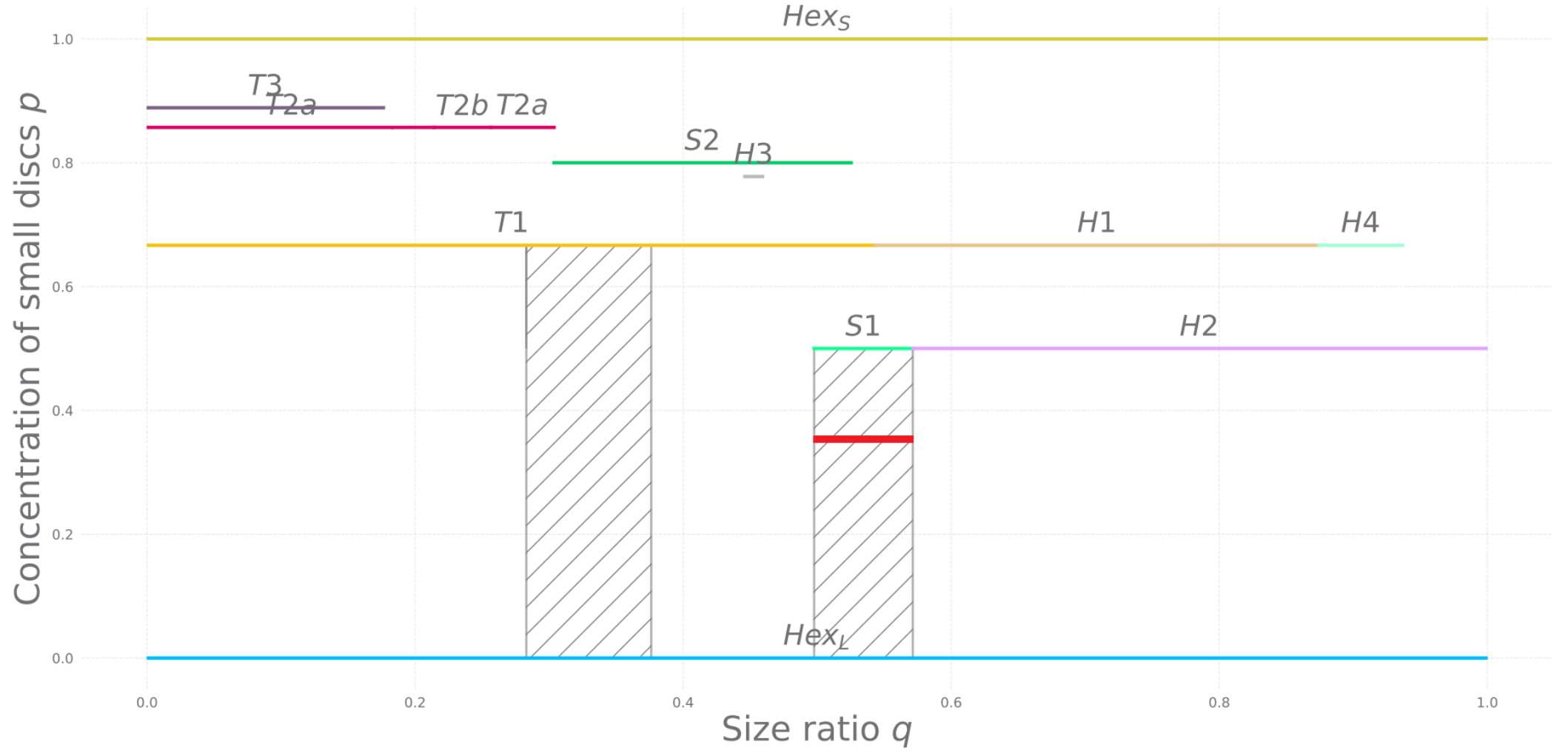
Delta = 0.95

Extension to non-additive hard discs



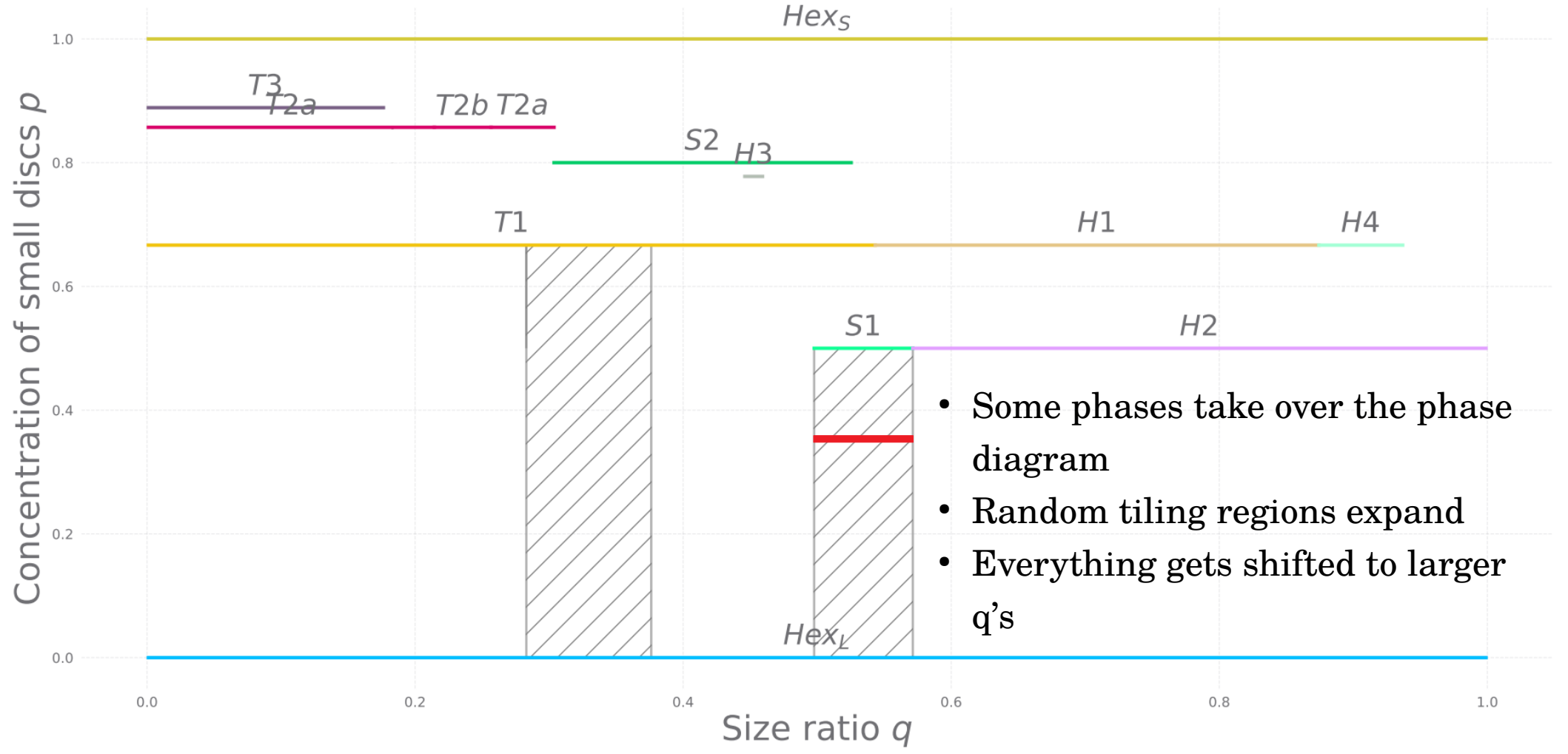
Delta = 0.90

Extension to non-additive hard discs



Delta = 0.90

Extension to non-additive hard discs



- Some phases take over the phase diagram
- Random tiling regions expand
- Everything gets shifted to larger q's

Conclusion

FBMC: reliable method to generate candidate structures for the phase diagram

=> updated phase diagram of binary mixtures of hard discs at infinite pressure

=> 2 random tiling QC regions

Non-additivity (extra length scale) increases the range of stability of the QC

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FBMC: reliable method to generate candidate structures for the phase diagram
=> updated phase diagram of binary mixtures of hard discs at infinite pressure
=> 2 random tiling QC regions

Non-additivity (extra length scale) increases the range of stability of the QC

Perspectives

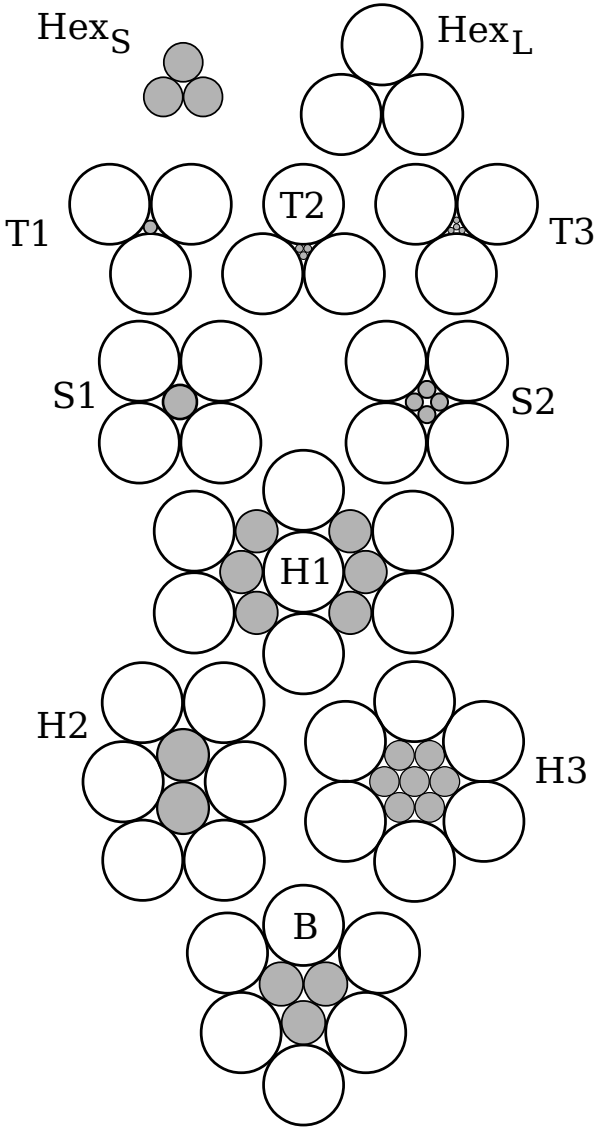
Try and equilibrate some of those random tiling QCs using large scale molecular dynamics simulations.

Acknowledgments

Frank SMALLENBURG
Giuseppe FOFFI
The SoftQC team

Project funded by the ANR grant n°18-CE09-0025

Thank you for your attention


Hex$_S$
Hex$_L$
T1
T2
T3
S1
S2
H1
H2
H3
B