# Structure formation in soft, polymer materials: Role of Data & Analytics

Marcus Müller

***outline*:**

- Introduction – some criteria for the potential of Data & Analytics from a perspective of a scientists
- Models, data and algorithms for structure formation in soft, polymer materials
- Concluding remarks

CoLiSA.MMP

SFB 937

Evanston, November 1 2016

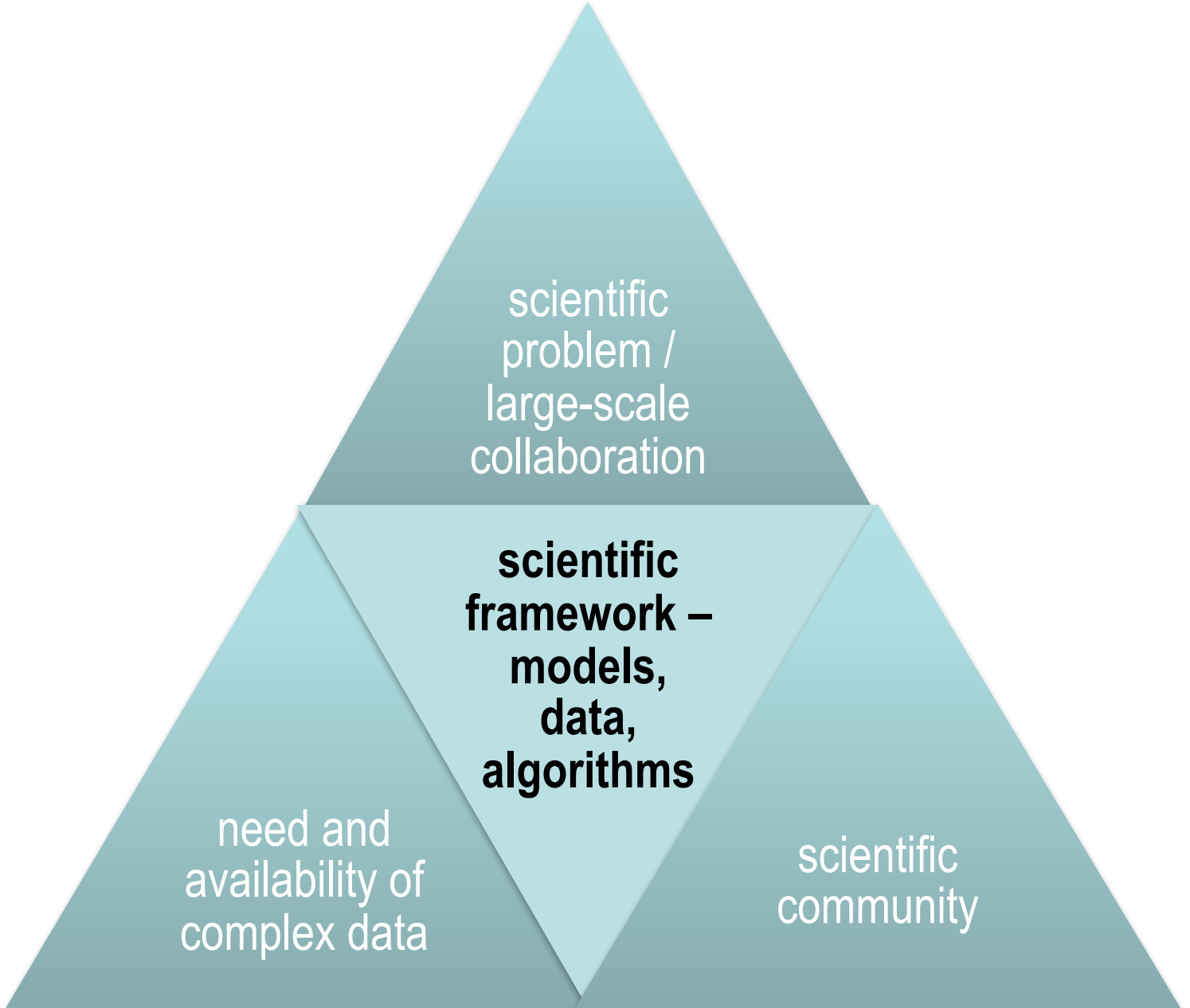
# Some criteria for potential of Data & Analytics

- ***local aspects on the scale of working groups*:**
  - re-use of data, collaborations and transfer of knowledge
  - program and data documentation / good scientific practice
  - compliance with mandates of funding agencies / government

➡ important, needs low-barrier software solution to facilitate documentation of work processes, can use problem-specific formats

- ***large-scale collaborations***:
  - an established example – high-energy physics (CERN)
  - synergistic collaborations of groups in different disciplines and locations
  - parallel development of experiment, theory, and computational infrastructure
  - building of community by common data and algorithms

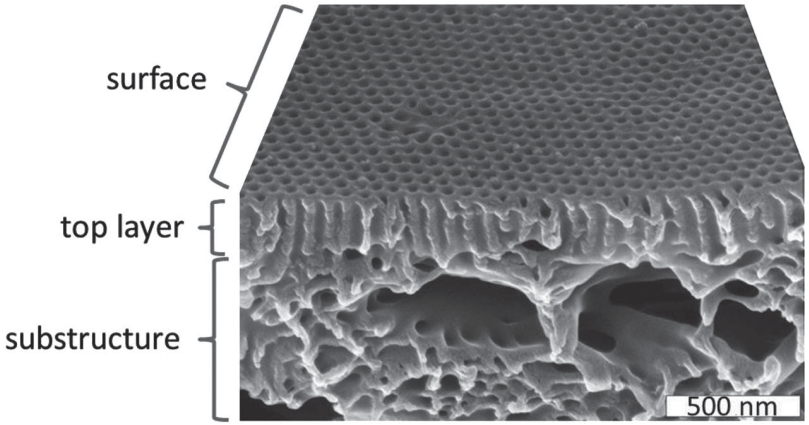
# Some criteria for the potential of Data & Analytics

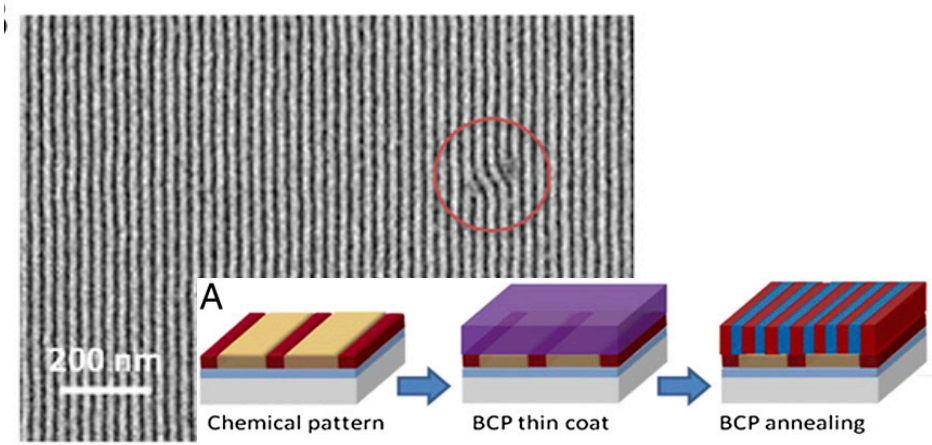


# Soft, polymer materials – scientific problem

- ***dense copolymer materials***
  (personal bias … there are many more)
  - microphase separation on the length scale from 5-100 nm
  - can be tailored in geometry and length scale by molecular
  - applications:
    - filtration membranes Abetz, *Macromol. Rapid Commun*. **36**, 10 (2015)
    - photonic materials
    - solar and fuel cells
    - DSA – microelectronic device fabrication

Hur, Thapar, Ramirez-Hernandez, Khaira, Segal-Peretz, Rincon-Delgadillo, Li, Müller, Nealey, de Pablo, *PNAS* **112**, 14144 (2015)



# Soft, polymer materials – scientific problem

- ***dense copolymer materials***
  (personal bias … there are many more)

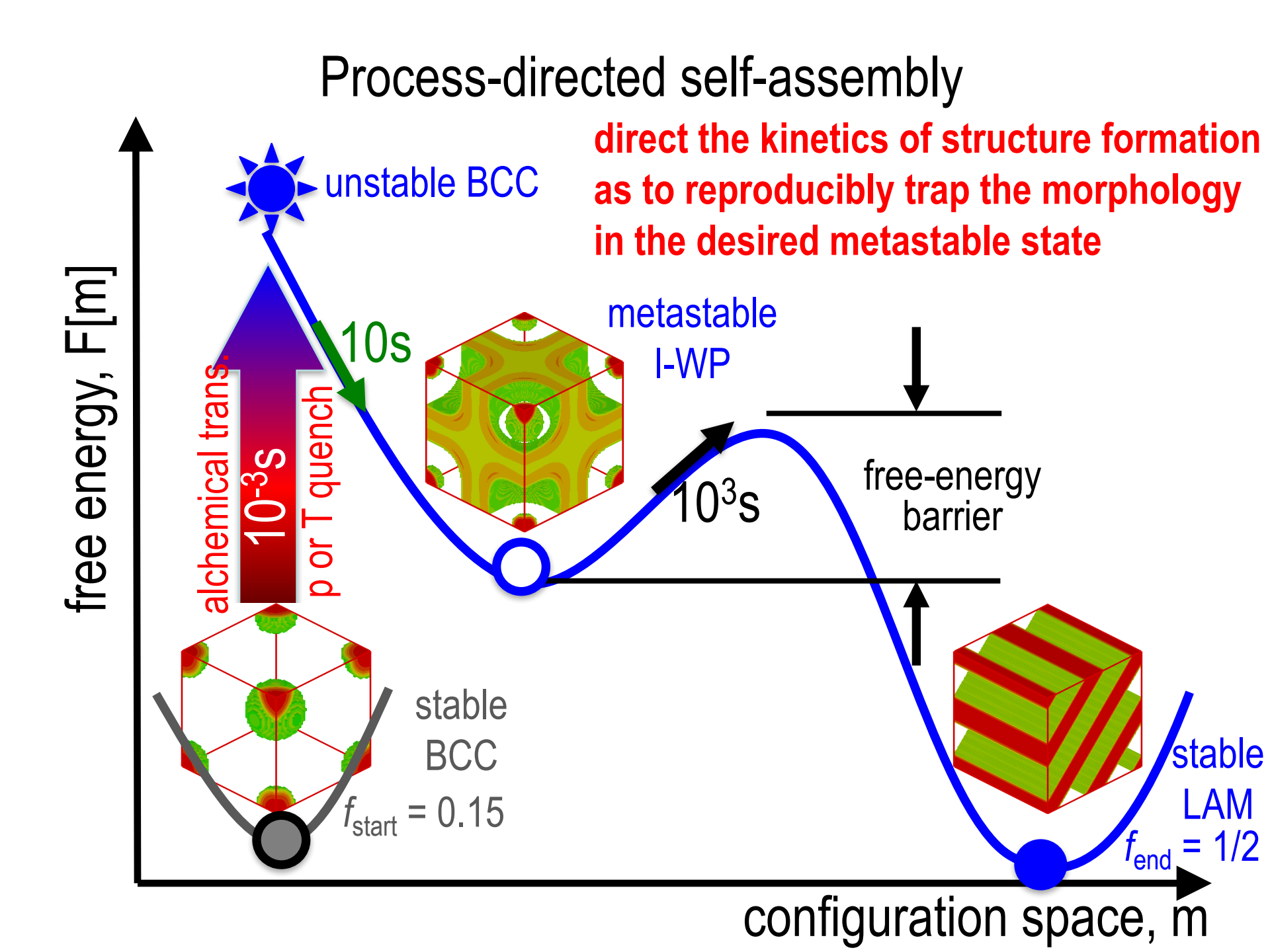
- ***self-assembly of amphilipic molecules in solution***
  (lipid membranes, lipid-protein assembly)

- ***colloidal self-assembly***
  (role of shape and depletion interactions, DNA-mediated interactions)

➡

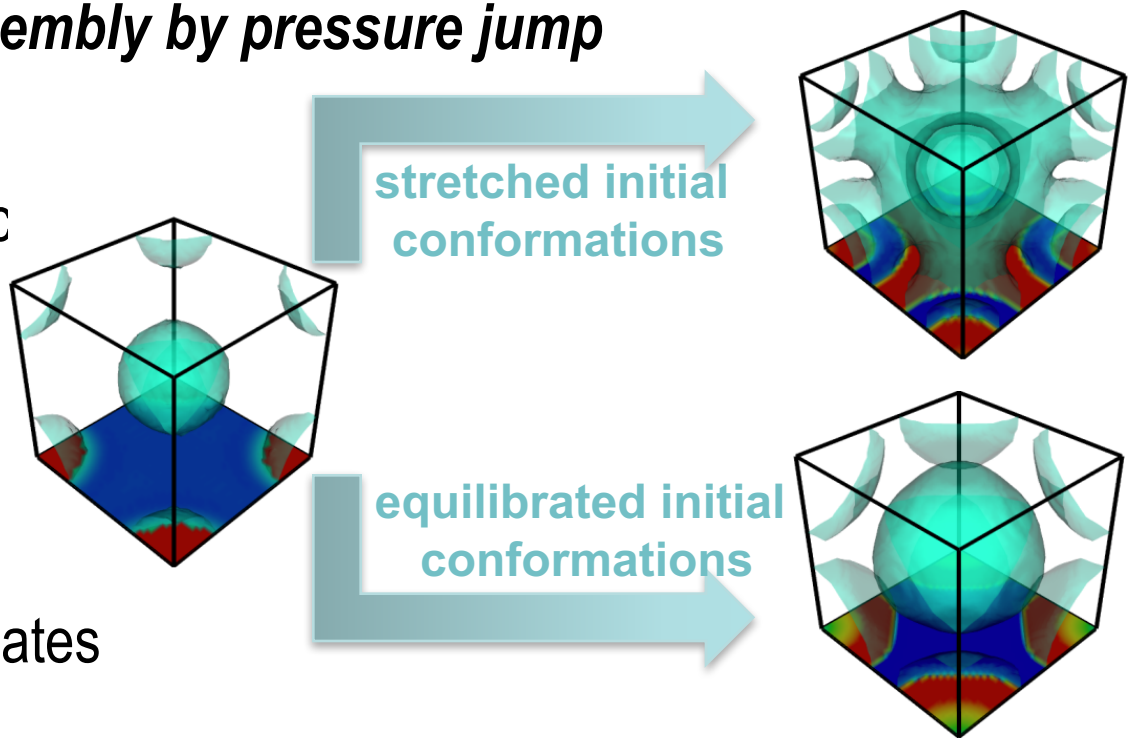
***common ingrediants that control the structure formation in soft materials*** (characteristic free-energy scale $k_BT$***)***

– kinetics of structure formation/transformation is crucial
– multitude of metastable states and intermediates
– process-directed self-assembly


Process-directed self-assembly
direct the kinetics of structure formation
as to reproducibly trap the morphology
in the desired metastable state
unstable BCC
10s
metastable
I-WP
alchemical trans.
10-3s
p or T quench
10³s
free-energy
barrier
stable
BCC
fstart = 0.15
stable
LAM
fend = 1/2
free energy, F[m]
configuration space, m

# Process-directed self-assembly

- ***process-directed self-assembly by pressure jump***
- fabrication of I-WP network morphology from BCC struc
- ***local conservation of density*** is crucial to predict pathway Jacobian of transformation from particle-based coordinates to densities

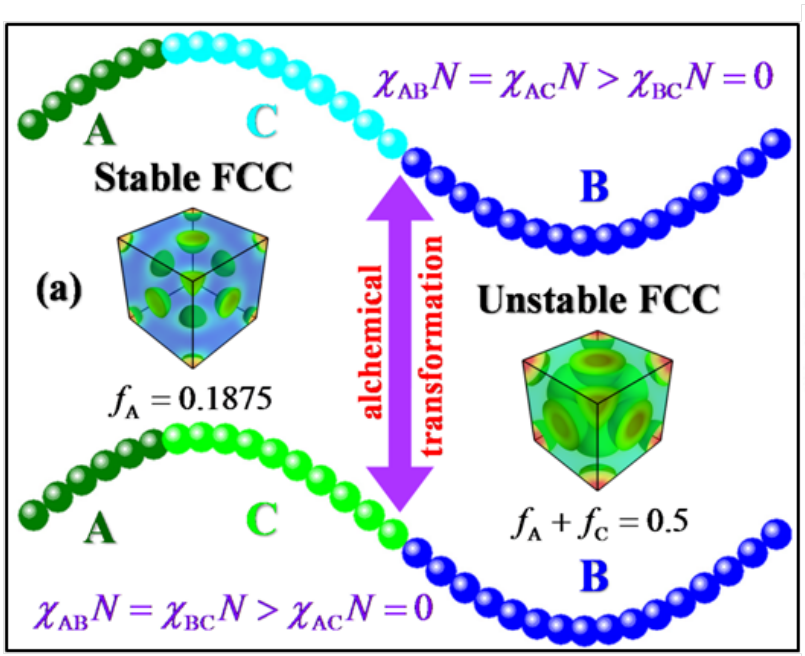


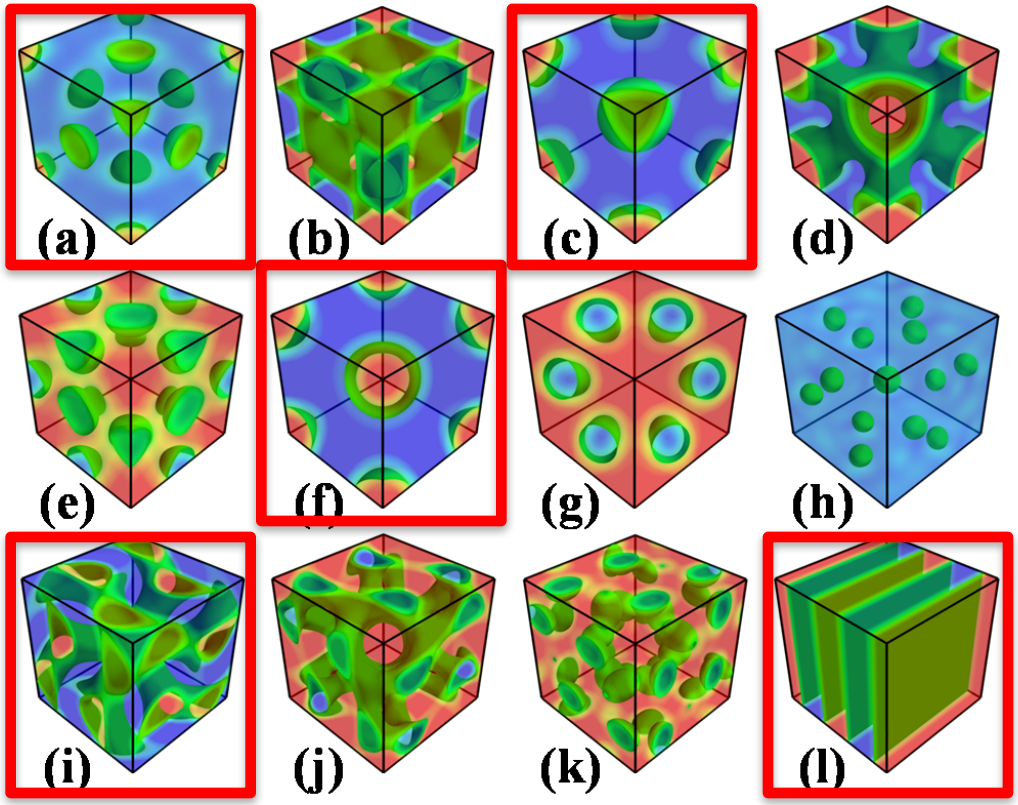

- ***importance of chain conformations being not in equilibrium*** with instantaneous density distribution

Müller, Sun, *Phys. Rev. Lett.* **111**, 267801 (2013)
Müller, Sun, *J. Phys.: Condens. Matter* **27**, 194101 (2015)

# Process-directed self-assembly

- ***process-directed self-assembly by alchemical (photochemical) transformation***



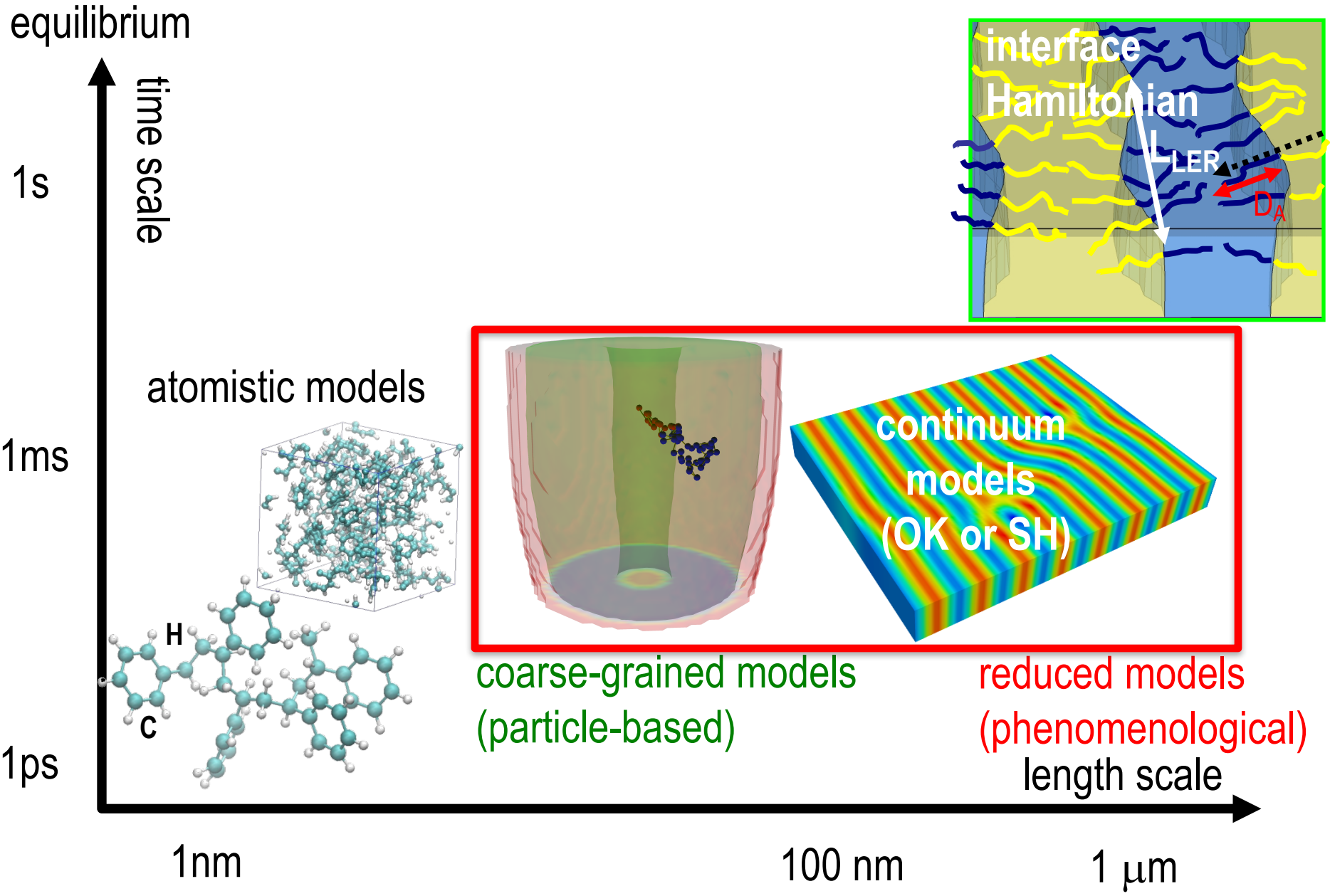




➡ variety of periodic structures:
(a) FCC, (b) F-RD, (c) BCC, (d) I-WP, (e) I-WP-S, (f) HEX,
(g) HEX2, (h) GYR-S, (i) GYR, (j) DIA, (k) DIA-S, (l) LAM

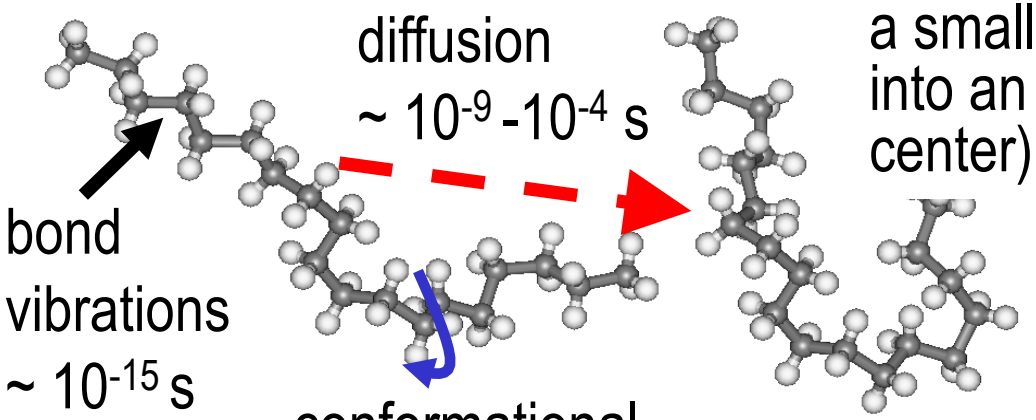
# Multitude of stable and metastable structures

- *Combinatorial screening of complex block copolymer assembly with self-consistent field theory*, Drolet, Fredrickson, *Phys. Rev. Lett.* **83**, 4317 (1999)
- *Discovering new ordered phases of block copolymers*, Bohbot-Raviv and Wang, *Phys. Rev. Lett.* **85**, 3428 (2000)
- *Design of ABC Triblock copolymers near the ODT with the Random Phase Approximation,* Cochran, Morse, Bates, *Macromolecules* **36**, 782 (2003)
- *Random isotropic structures and possible glass transitions in diblock copolymer melts*, Zhang, Wang, *Phys. Rev. E* **73**, 031804 (2006).
- *Discovering ordered phases of block copolymers: New results from a generic Fourier-space approach*, Guo, Zhang, Qiu, Zhang, Yang, Shi, *Phys. Rev. Lett.* **101**, 028301 (2008)
- *Broadly accessible self-consistent field theory for block polymer materials discovery*, Arora, Qin, Morse, Delaney, Fredrickson, Bates, Dorfman, *Macromolecules* **49**, 4675 (2016)

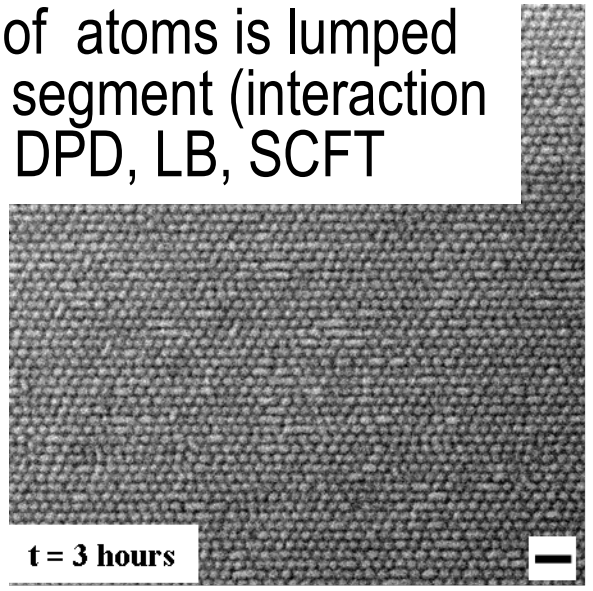
# Scientific framework – **models**, data & algorithms



# Scientific framework – **models**, data & algorithms




a small number of atoms is lumped into an effective segment (interaction center) MC,MD, DPD, LB, SCFT



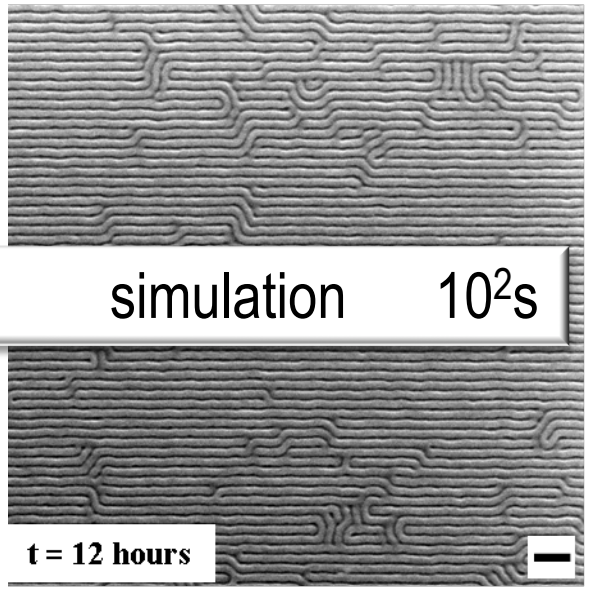

minimal coarse-grained model that captures only ***relevant interactions***: connectivity, excluded volume, repulsion of unlike segments

- incorporate essential interactions through a

$10^{-15}$s minimal, soft, coarse-grained model $10^{-5}$s simulation $10^{2}$s

  chain extension, $R_e$, compressibility $\kappa N$ and Flory-Huggins parameter $\chi N$ ➡ ***universality***
- elimination of degrees of freedom

  ➡ ***top-down model***

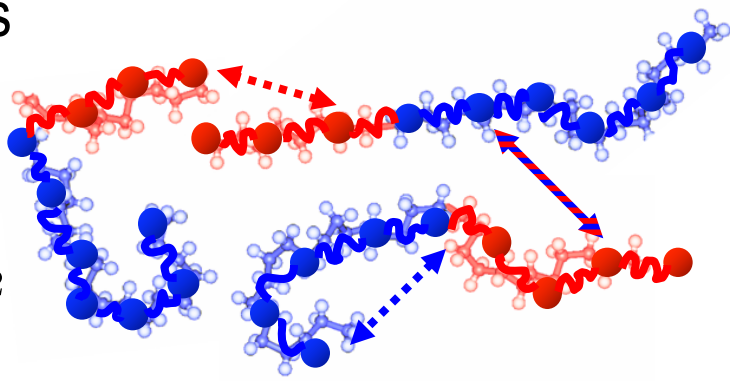



Daoulas, Müller, JCP **125**, 184904 (2006)

# Scientific framework – **models**, data & algorithms

***top-down model*** with soft, pairwise interactions

$$\frac{\mathcal{H}_{\mathrm{b}}[\mathbf{r}_i(s)]}{k_BT} = \sum_{s=1}^{N-1} \frac{3(N-1)}{2{R_{\mathrm{eo}}}^2} \left[\mathbf{r}_i(s) - \mathbf{r}_i(s+1)\right]^2$$



$$\frac{\mathcal{H}_{\mathrm{ord}}[\hat{\phi}_A, \hat{\phi}_B]}{k_BT\sqrt{\bar{\mathcal{N}}}} = -\frac{\chi_{\mathrm{o}}N}{4} \int \frac{\mathrm{d}^3\mathbf{r}}{{R_{\mathrm{eo}}}^3} \left[\hat{\phi}_A(\mathbf{r}) - \hat{\phi}_B(\mathbf{r})\right]^2$$

$$\frac{\mathcal{H}_{\mathrm{melt}}[\hat{\phi}_A, \hat{\phi}_B]}{k_BT\sqrt{\bar{\mathcal{N}}}} = +\frac{\kappa_{\mathrm{o}}N}{2} \int \frac{\mathrm{d}^3\mathbf{r}}{{R_{\mathrm{eo}}}^3} \left[\hat{\phi}_A(\mathbf{r}) + \hat{\phi}_B(\mathbf{r}) - 1\right]^2 \quad \text{with } \sqrt{\bar{\mathcal{N}}} \equiv \Phi_{\mathrm{p}} R_{\mathrm{eo}}^3$$

effective interactions become weaker for large degree of coarse-graining

➡ no (strict) excluded volume, soft, effective segments can overlap, rather enforce low compressibility on length scale of interest, $R_{\mathrm{eo}}$

$$\hat{\phi}_A(\mathbf{r}) = \frac{1}{\rho_{\mathrm{o}}} \sum_{i_A=1}^{nNf} \delta(\mathbf{r} - \mathbf{r}_{i_A})$$

$\hat{\phi}^2$ -terms generate pairwise interactions

particle-based description for MC, BD, DPD, or SCMF simulations

Müller, Smith, *J. Polym. Sci. B* **43**, 934 (2005); Daoulas, Müller, *JCP* **125**, 184904 (2006); Detcheverry, Kang, Daoulas, Müller, Nealey, de Pablo, *Macromolecules* **41**, 4989 (2008); Pike, Detcheverry, Müller, de Pablo, *JCP* **131**, 084903 (2009); Detcheverry, Pike, Nealey, Müller, de Pablo, *PRL* **102**, 197801 (2009)

# Scientific framework – **models**, data & algorithms

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) R21–R47 PII: S0953-8984(02)17948-3

**TOPICAL REVIEW**

## The standard Gaussian model for block copolymer melts

**M W Matsen**

Polymer Science Centre, University of Reading, Whiteknights, Reading RG6 6AF, UK

- "universal standard model" that defines a framework to categorize models for particle simulation, polymer DFT, molecular theory and SCFT
- starting point for generalizations (molecular architecture, EOS-effects and solvents, charges or supramolecular interactions, dynamical properties)

# Scientific framework – models, **data & algorithms**

- ***equilibrium data***:
  - phase diagrams as a function of molecular architecture (SCFT)
  - effects of compressibility and local packing structure (p-DFT)
  - exploration of potential structure (particle simulation)
  - scattering experiments
  - real-space imaging techniques (TEM/AFM/tomography+SIS)
- ***dynamic equilibrium data***:
  - rheology data
  - diffusion constant, single-chain motion in spatially modulated structure
- ***non-equilibrium collective dynamics****:*
  - structure formation, reorientation dynamics in external fields
  - defect motion and annihilation
  - process-directed structure formation

***multitude techniques***: SCFT, p-DFT, molecular theory, particle simulation, GISAX/GISAS, TEM, AFM, tomography+SIS

# Scientific framework – models, **data & algorithms**

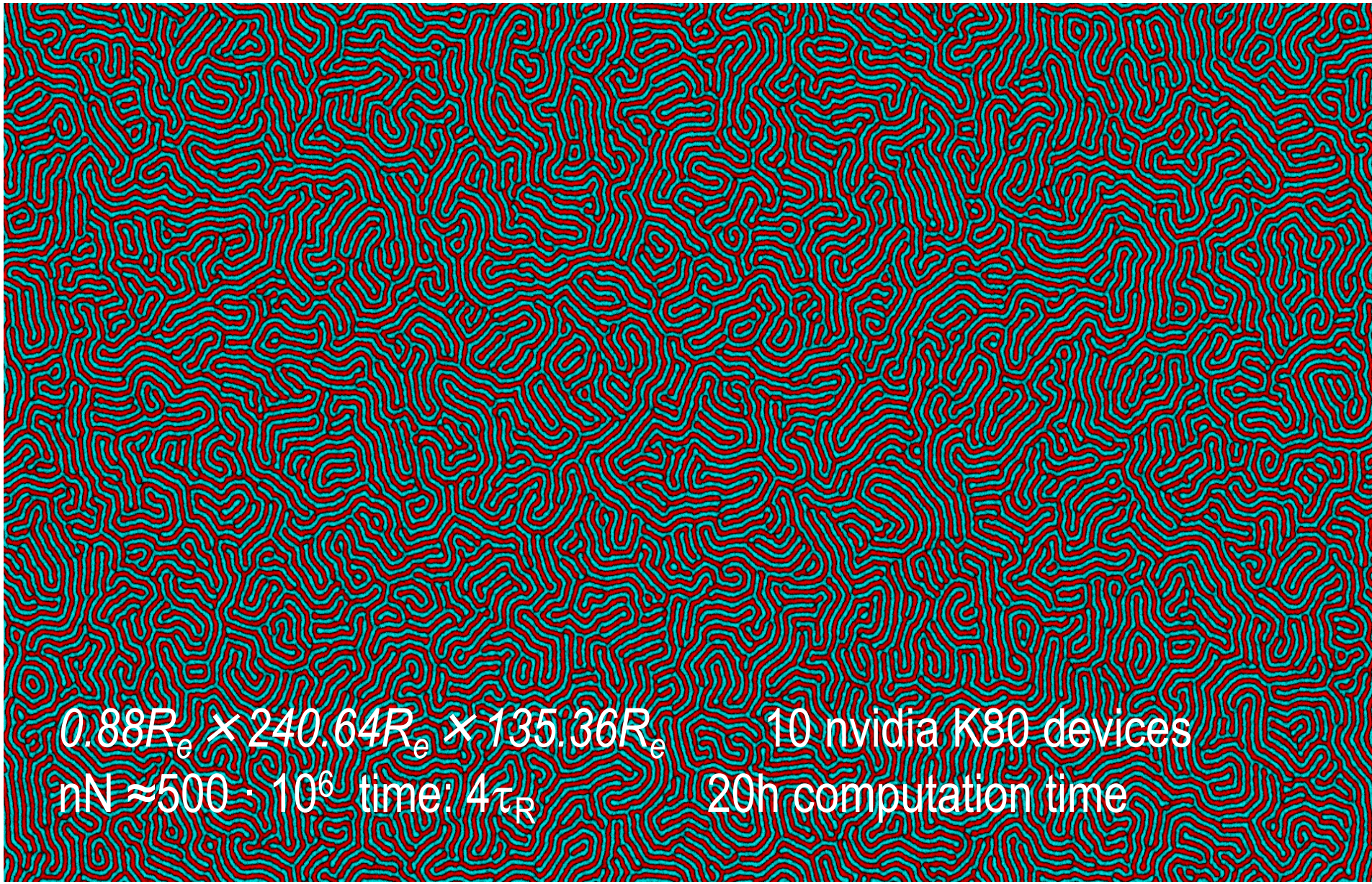
***example***: particle simulations on GPUs

- ***MPI***
  - no domain decomposition
  - polymer block parallel
- ***OpenACC 2.5***
  - fine grain parallel (polymer)
  - #pragma GPU parallel
  - Imemory offloading
  - PGI compiler
- ***OpenMP***
  - like OpenACC
  - any compiler

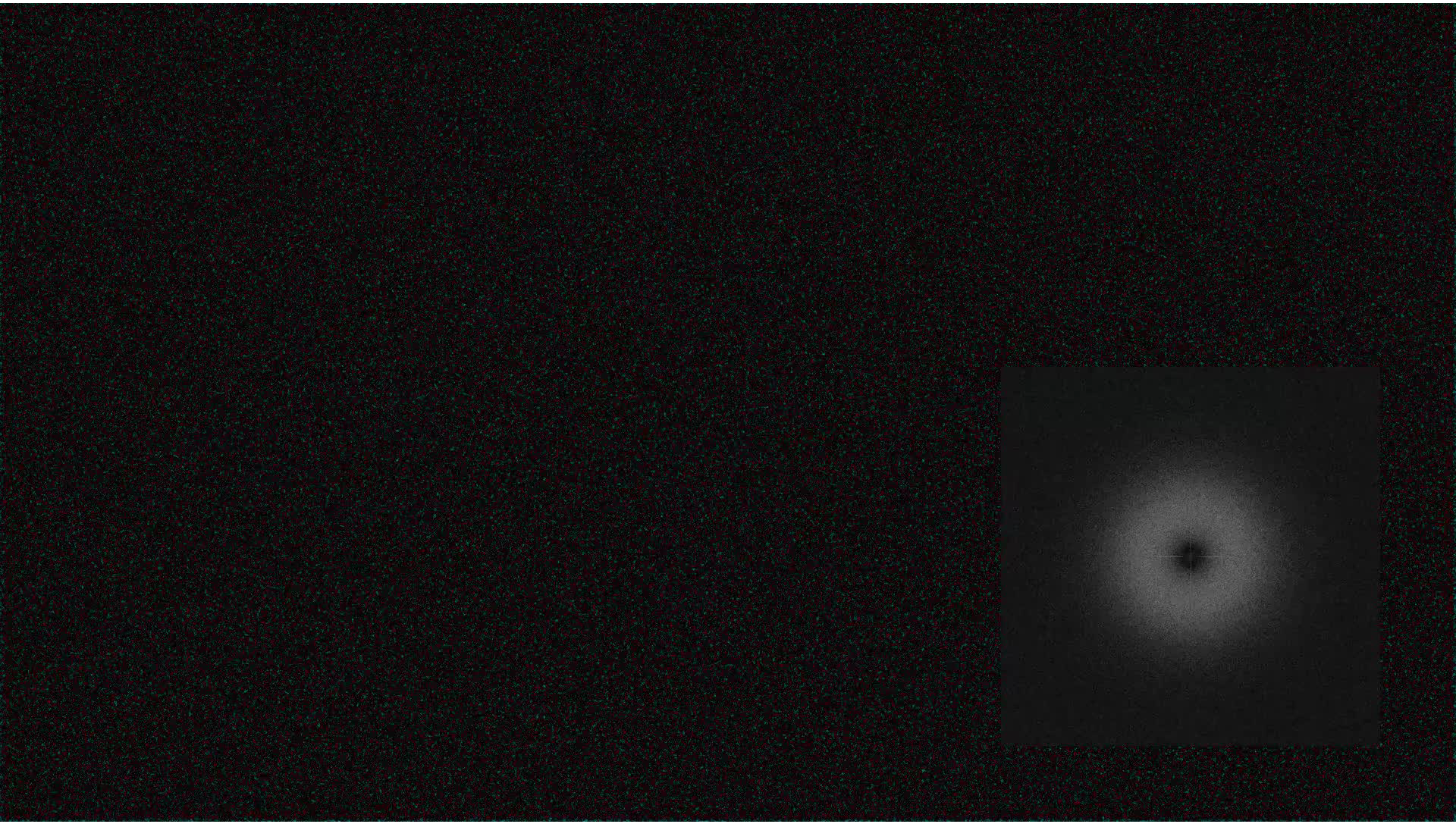
- ***features***
  - Smart Monte-Carlo moves
  - Distributed computation via MPI / OpenMP
  - single source code multiple architectures: CPU and GPU (via OpenACC)
  - ***hdf5 i/o format***, parallel i/o
  - up to 64 polymer types

➡ synergies in co-development of programs and unified data formats: GROMACS, LAMMPS, ESPRESSO, HOOMD

# Need and availability of complex data



# Need and availability of complex data



# Need and availability of complex data

***wealth of information*** (even in this deceptively simple example):

- single-chain motion in spatially modulated environment
- kinetics of collective structure formation (initial, spinodal)
- defect motion and annihilation
- grain growth and grain boundary motion
- large-scale connectivity of domains

requires analysis on different scales, using different models and comparison to various experimental techniques;
interaction between simulation (theory) and experiment

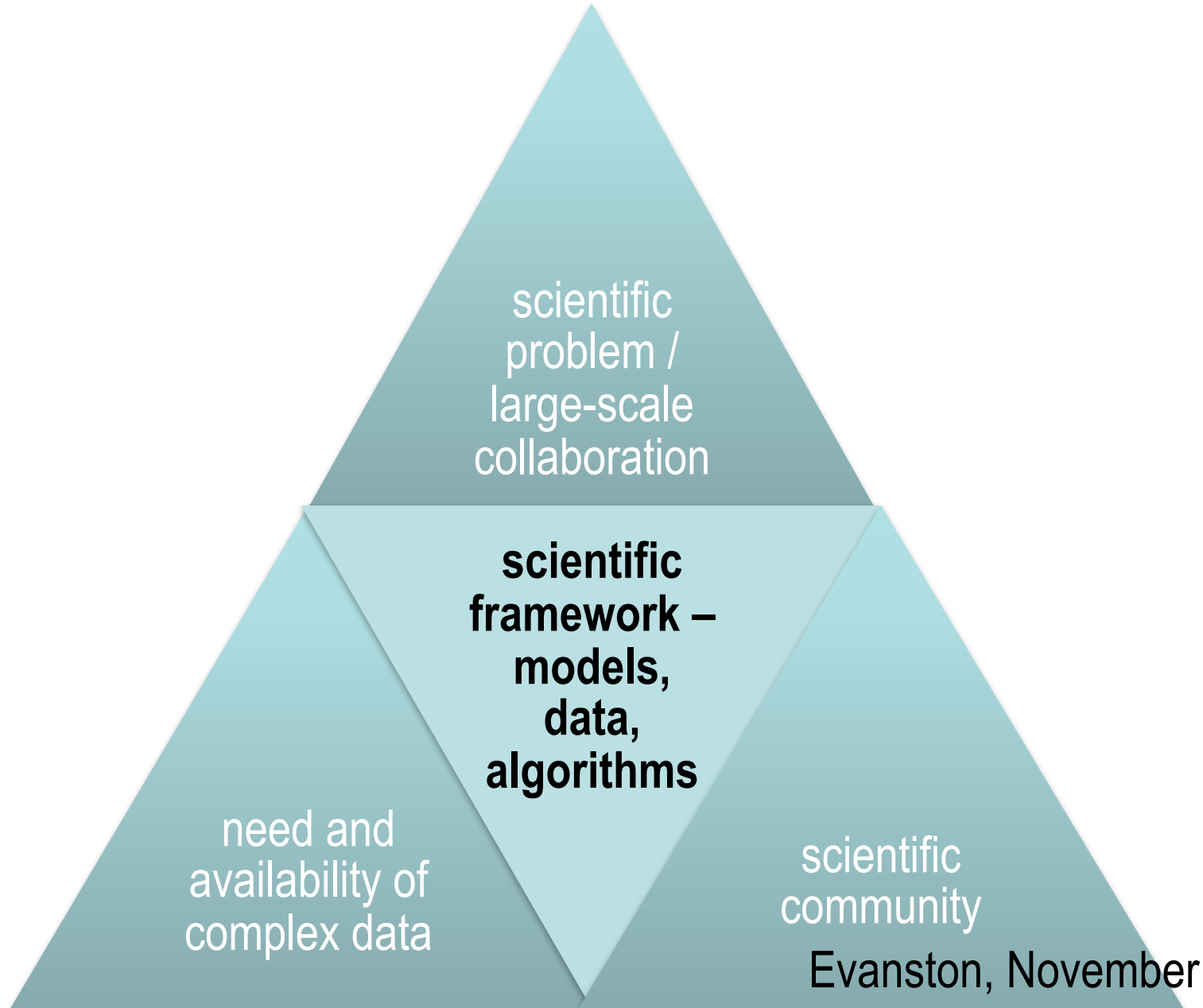
➡

"computing gets more and more intertwined with data analysis"
US DOE ASCAC Report, *Synergistic challenges in data-intensive science and exascale computing*, 2013

➡

***involvement of different scientific communities using common standards***

# Opportunities for Data & Analytics in Soft Matter research

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# Opportunities for Data & Analytics in Soft Matter research

- ***scientific problem***:
  - relevant for technological applications
  - complex, rich behavior due to multitude of competing structure
- ***framework***:
  - universal standard model (top-down approach)
    connecting computational models to experiments
- ***availability of data***:
  - advances in numerical methods (SCFT, GPU-based simulation) and experiments
  - wealth of information (from single-molecule motion to large-scale morphology)
- ***scientific community***:
  - numerous groups in America, Europe, and Asia
  - common standards and community-building required

complex data

Evanston, November 1 2016

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disclaimer – specific case of structure formation in dense copolymer materials
but similar for other Soft Matter areas

complex data