Diffuse Interface Field Approach (DIFA) to Modeling and Simulation of Particle-based Materials Processes

Yu U. Wang

Materials Science and Engineering Department
Michigan Technological University

Motivation

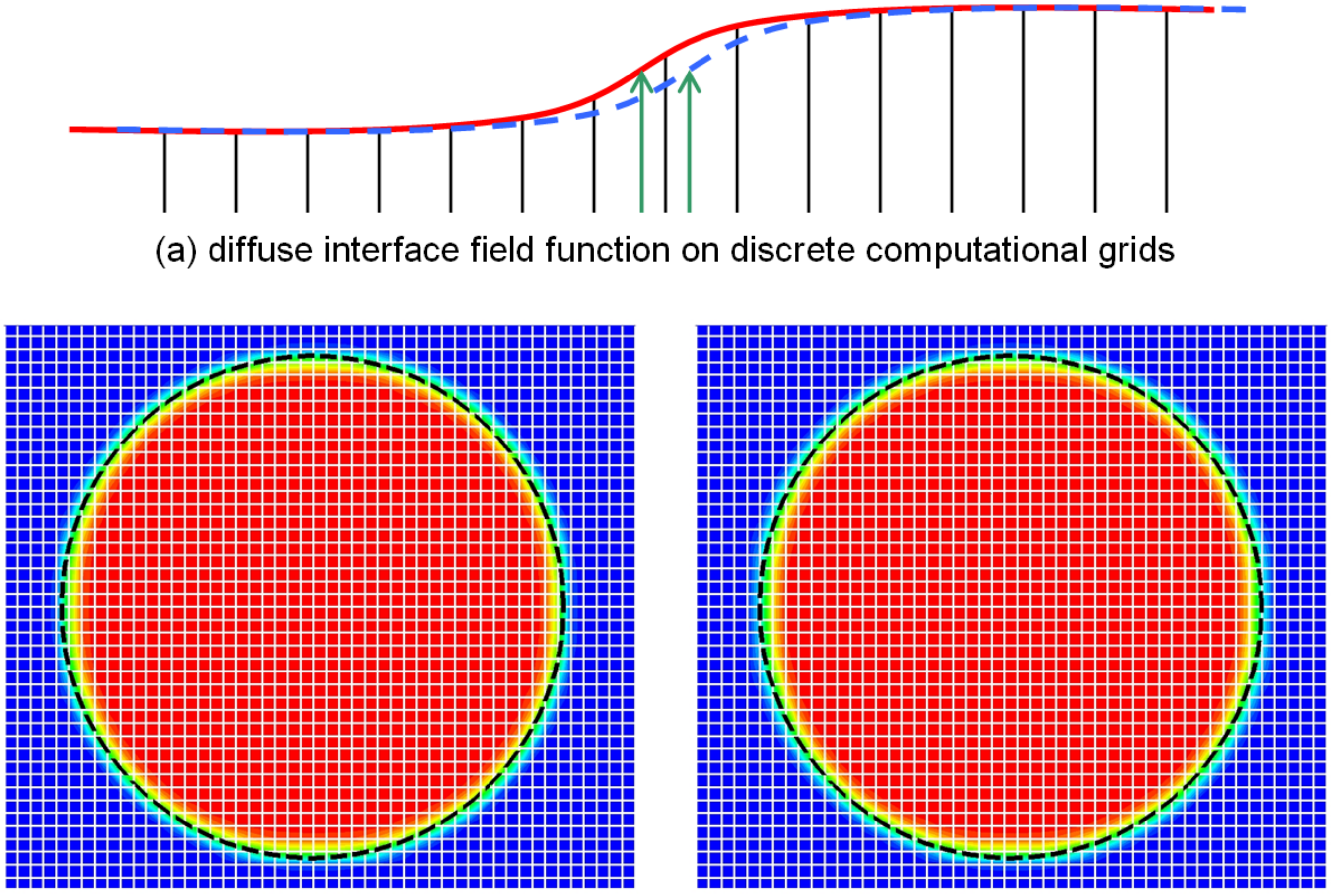
- Extend **phase field method** to model free-body solid particles

Outline

- Interesting and important issues in particle processes
  - Moving particles of **arbitrary shapes** and sizes in **close distance**: rigid-body translations and rotations
  - Short-range forces: mechanical contact, friction, cohesion, steric repulsion, Stokes drag (particle shape matters)
  - Long-range forces: electric charge, charge heterogeneity, electric double layer, electric/magnetic dipole, van der Waals (point-charge/point-dipole approximation inaccurate)
  - External forces: electric/magnetic field, gravity (field-directed self-assembly)
  - Multi-phase liquid: fluid interface evolution, capillary force on particles (surface tension, Laplace pressure via Gibbs-Duhem relation)

Model Formulation

- Diffuse interface field description: arbitrary particle shape, continuous motion on discrete computational grids, as desired for dynamic simulation



(a) diffuse interface field function on discrete computational grids

(b) centered at $X^*=86.95$, $Y^*=64$

(c) centered at $X^*=87.50$, $Y^*=64$

Model Formulation

- Short-range forces: mechanical contact, steric repulsion

$$d\mathbf{F}^{\mathrm{sr}}(\mathbf{r};\alpha)=\kappa\sum_{\alpha'\neq\alpha}\eta(\mathbf{r};\alpha)\eta(\mathbf{r};\alpha')\left[\nabla\eta(\mathbf{r};\alpha)-\nabla\eta(\mathbf{r};\alpha')\right]d^3r$$

action-reaction symmetry

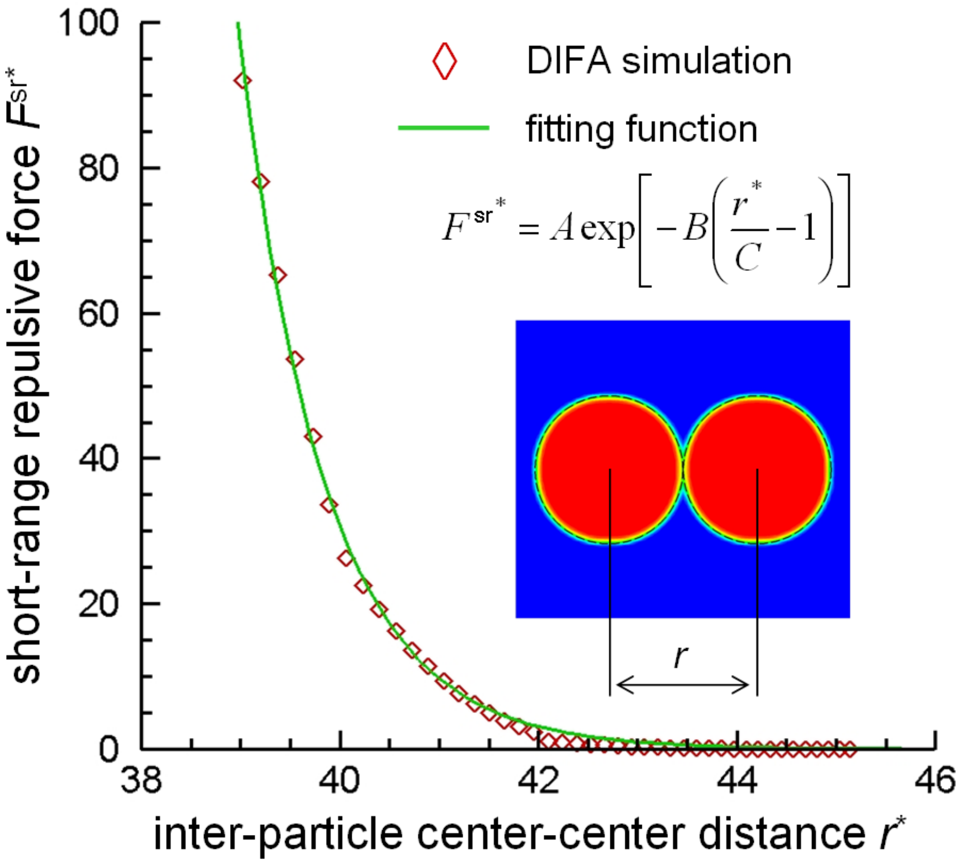
$$\mathbf{F}^{\mathrm{sr}}(\alpha)=\int_V d\mathbf{F}^{\mathrm{sr}}(\mathbf{r};\alpha)$$

soft-particle potential

$$\mathbf{T}^{\mathrm{sr}}(\alpha)=\int_V\left[\mathbf{r}-\mathbf{r}^{\mathrm{c}}(\alpha)\right]\times d\mathbf{F}^{\mathrm{sr}}(\mathbf{r};\alpha)$$

torque

Arbitrary particle shapes
without tracking interfaces



Model Formulation

- Total force and torque acting on individual particle

$$\mathbf{F}(\alpha)=\mathbf{F}^{\mathrm{sr}}(\alpha)+\boldsymbol{\xi}^{\mathrm{f}}(\alpha)$$

$$\mathbf{T}(\alpha)=\mathbf{T}^{\mathrm{sr}}(\alpha)+\boldsymbol{\xi}^{\mathrm{t}}(\alpha)$$

thermal noise for Brownian motion

- Particle dynamics in viscous liquid

$$V_i(\alpha)=M_{ij}(\alpha)F_j(\alpha)$$

$$\Omega_i(\alpha)=N_{ij}(\alpha)T_j(\alpha)$$

small Reynolds number Re<<1,
Stokes drag (friction), mobility

- Equation of motion

$$\eta(\mathbf{r},t;\alpha)=\eta\left(\mathbf{r}^0,t_0;\alpha\right)$$

$$r_i=Q_{ij}(t;\alpha)\left[r_j^0-r_j^{\mathrm{c}}(t_0;\alpha)\right]+r_i^{\mathrm{c}}(t;\alpha)$$

mapping without
error accumulation

$$r_i^{\mathrm{c}}(t+dt;\alpha)=r_i^{\mathrm{c}}(t;\alpha)+V_i(t;\alpha)dt$$

translation

$$Q_{ij}(t+dt;\alpha)=R_{ik}(t;\alpha)\,Q_{kj}(t;\alpha)$$

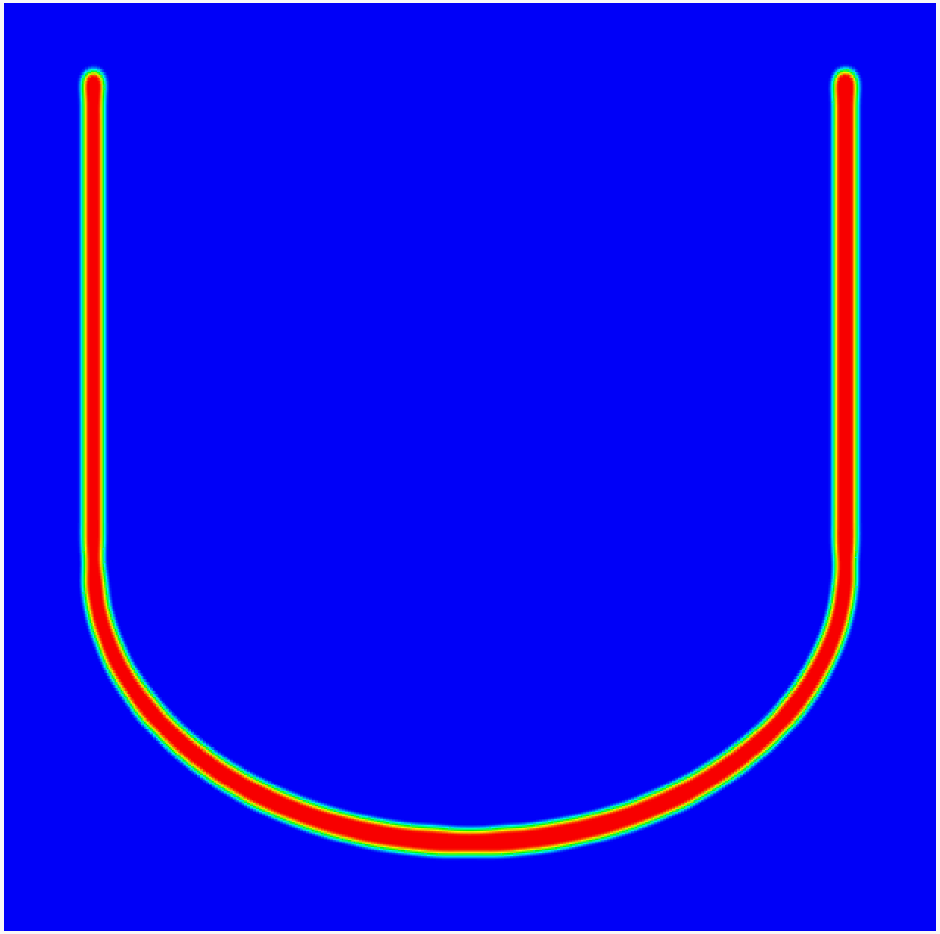
rotation

$$R_{ij}(t;\alpha)=\delta_{ij}\cos\omega+m_im_j(1-\cos\omega)-\varepsilon_{ijk}m_k\sin\omega$$

incremental rotation

Simulation

- Particle sedimentation and stacking



Simulation

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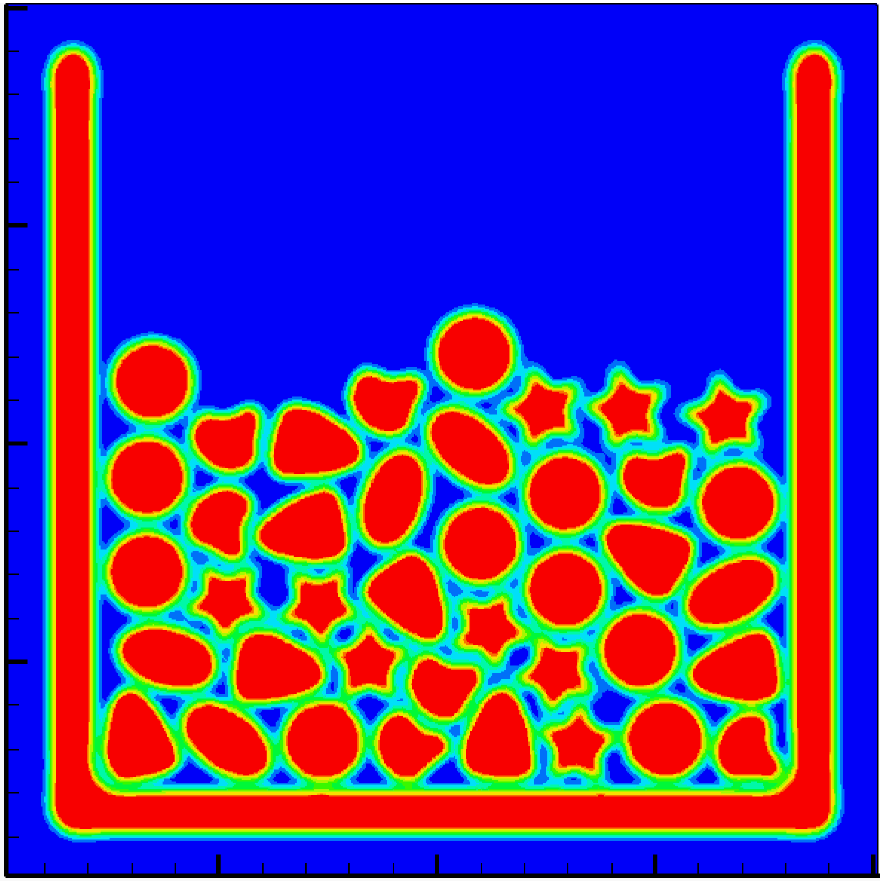
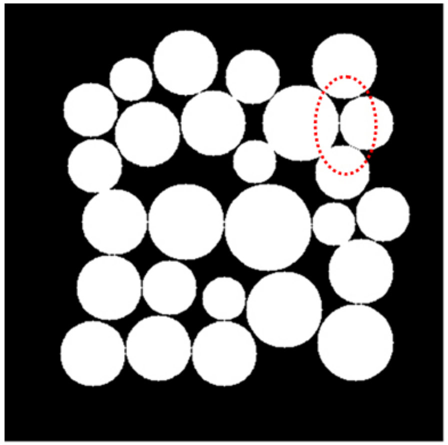
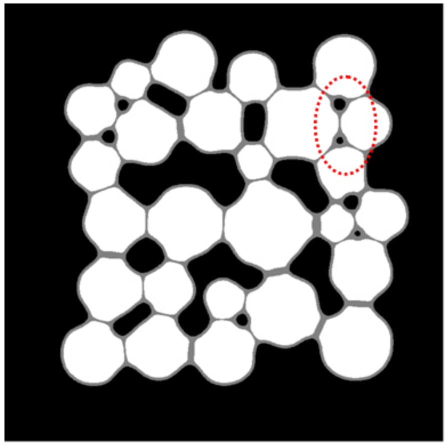


Simulation

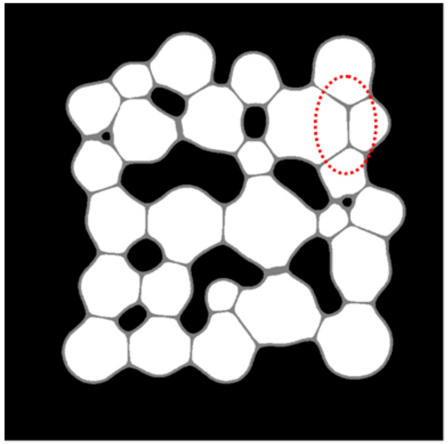
- Phase field model of solid-state sintering: rigid-body motions



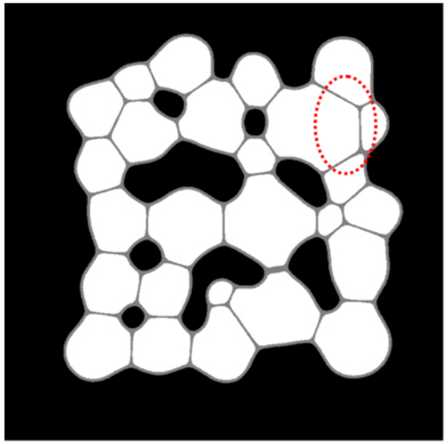
(a) t*=0



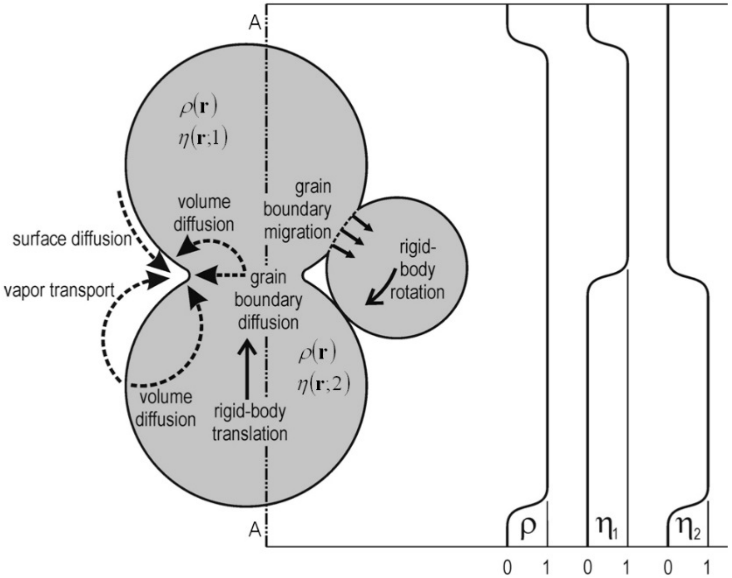
(b) t*=16

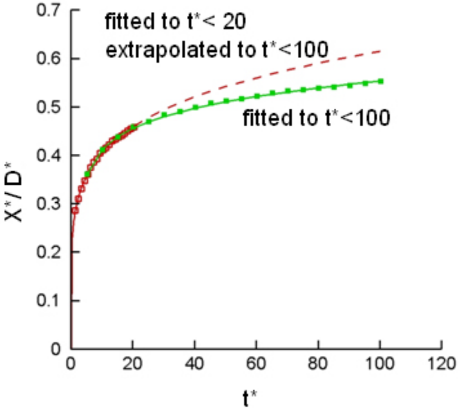


(c) t*=46



(d) t*=81





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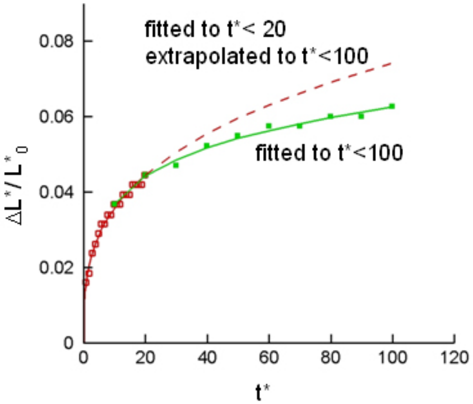


(b)

Model Formulation

- Long-range force: charged particles

$$\rho(\mathbf{r},t;\alpha)=\rho(\alpha)\,\eta(\mathbf{r},t;\alpha)$$ body charge

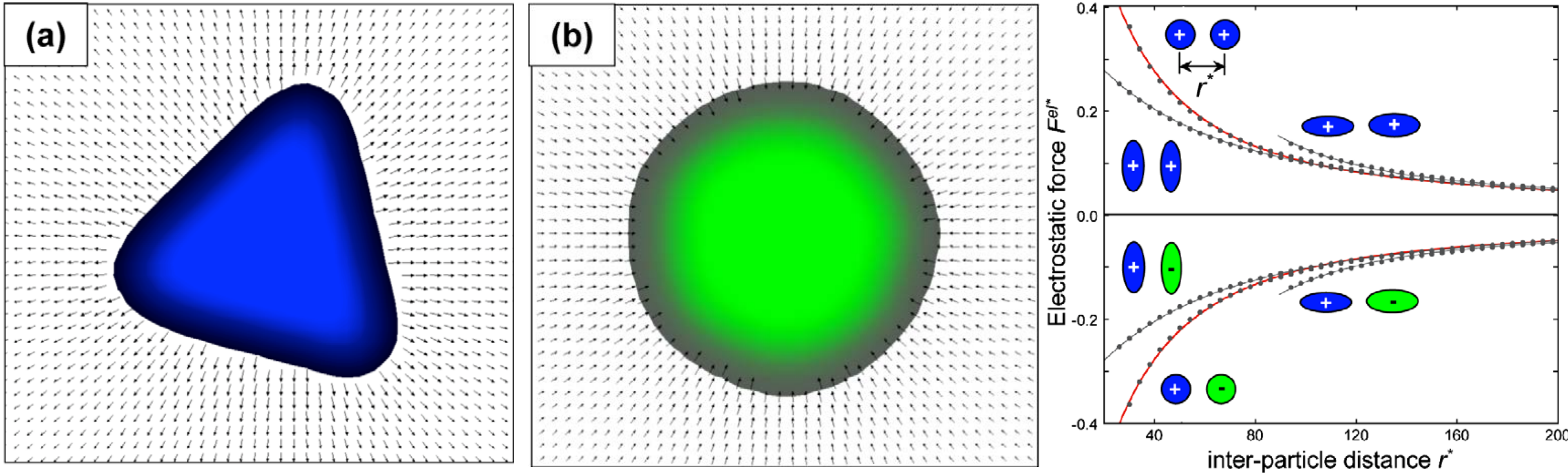
$$\rho(\mathbf{r},t;\alpha)=\rho(\alpha)\,\eta(\mathbf{r},t;\alpha)\left[1-\eta(\mathbf{r},t;\alpha)\right]$$ surface charge

$$\rho(\mathbf{r},t)=\sum_{\alpha}\rho(\mathbf{r},t;\alpha)$$

$$\mathbf{E}(\mathbf{r})=\mathbf{E}^{\mathrm{ex}}-\frac{i}{\varepsilon_0}\int\frac{d^3k}{(2\pi)^3}\frac{\tilde{\rho}(\mathbf{k})}{k}\mathbf{n}\,e^{i\mathbf{k}\cdot\mathbf{r}}$$

$$\mathbf{F}^{\mathrm{el}}(\alpha)=\int_V\mathbf{E}(\mathbf{r})\,\rho(\mathbf{r};\alpha)\,d^3r$$

$$\mathbf{T}^{\mathrm{el}}(\alpha)=\int_V\left[\mathbf{r}-\mathbf{r}^{\mathrm{c}}(\alpha)\right]\times\mathbf{E}(\mathbf{r})\,\rho(\mathbf{r};\alpha)\,d^3r$$



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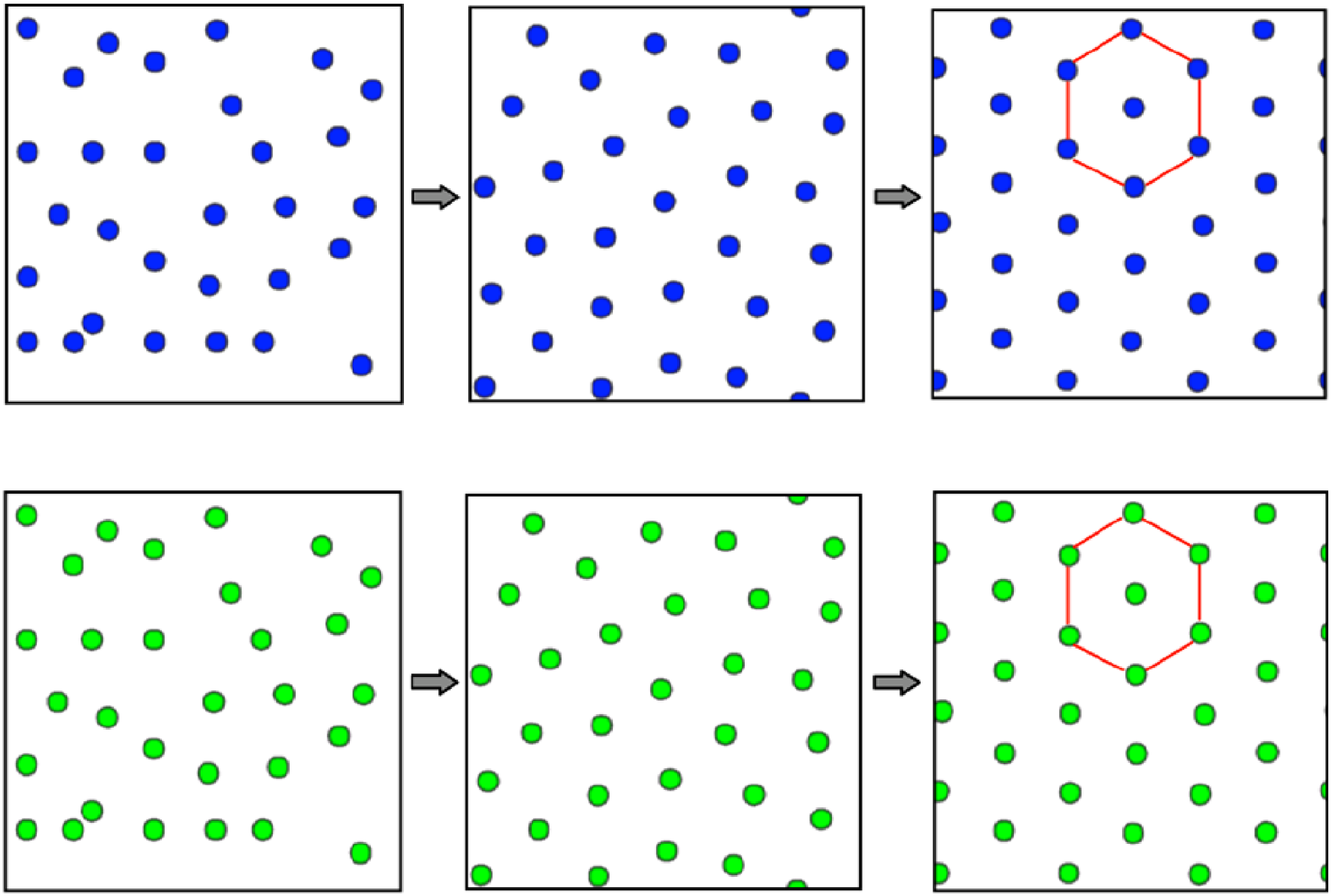
- Total force and torque acting on individual particle

$$\mathbf{F}(\alpha)=\mathbf{F}^{\mathrm{el}}(\alpha)+\mathbf{F}^{\mathrm{sr}}(\alpha)+\boldsymbol{\xi}^{\mathrm{f}}(\alpha)$$

$$\mathbf{T}(\alpha)=\mathbf{T}^{\mathrm{el}}(\alpha)+\mathbf{T}^{\mathrm{sr}}(\alpha)+\boldsymbol{\xi}^{\mathrm{t}}(\alpha)$$

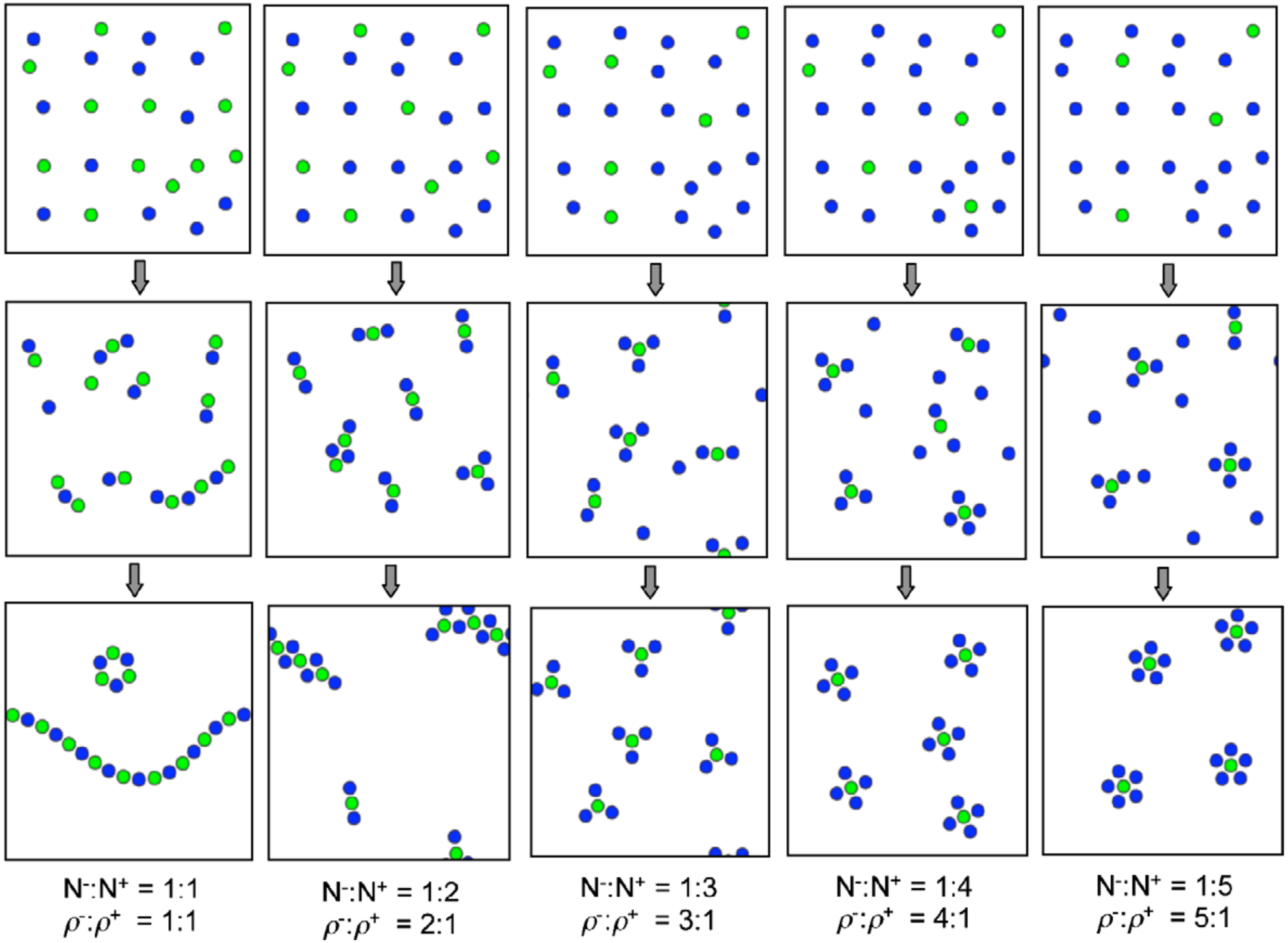
Simulation

- Particles of same charge: repulsion



Simulation

- Particles of opposite charges: attractive self-assembly

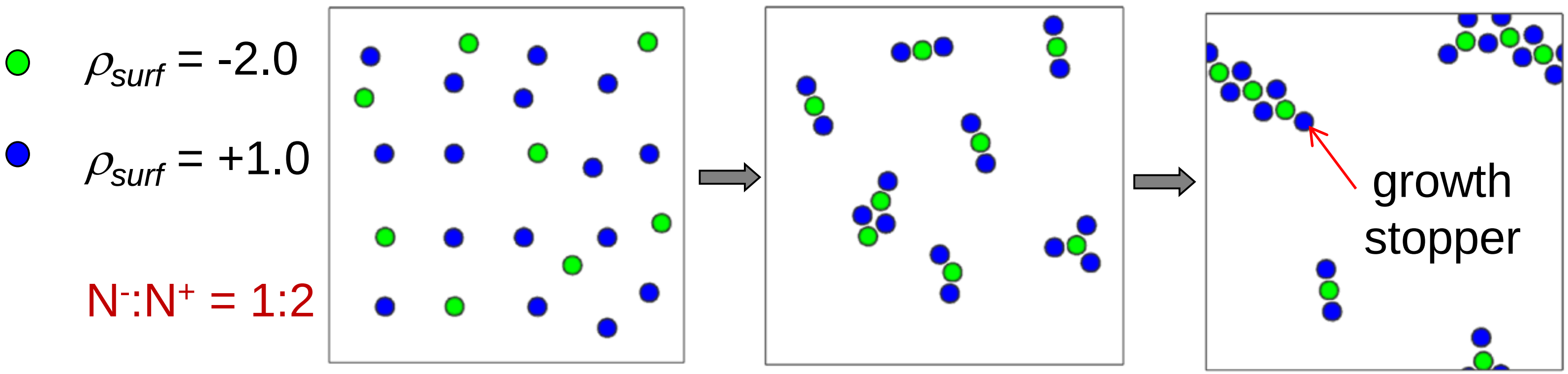


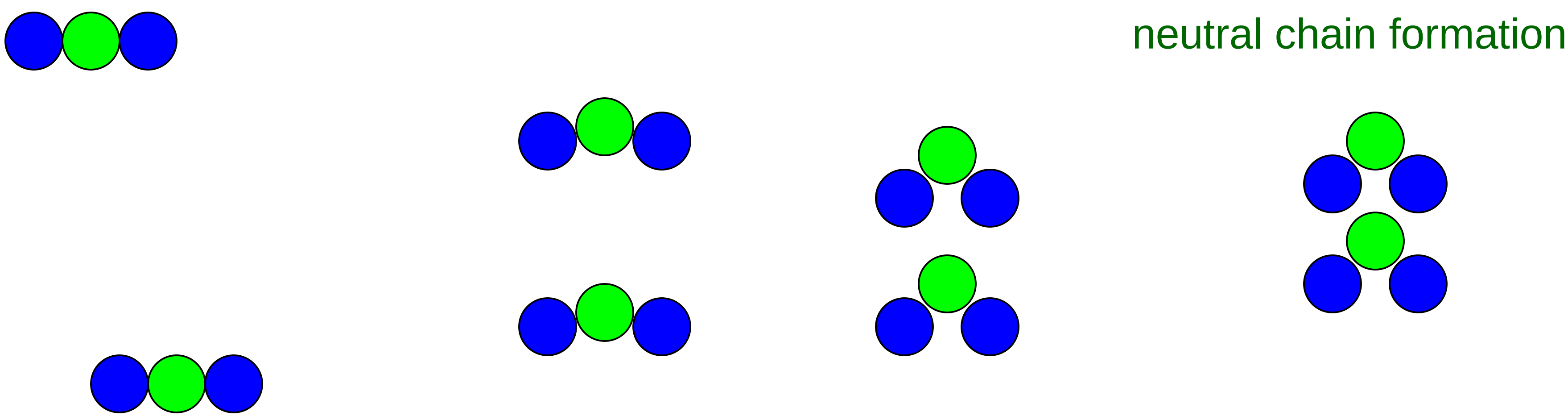
dipolar

stable

mutually induced dipoles

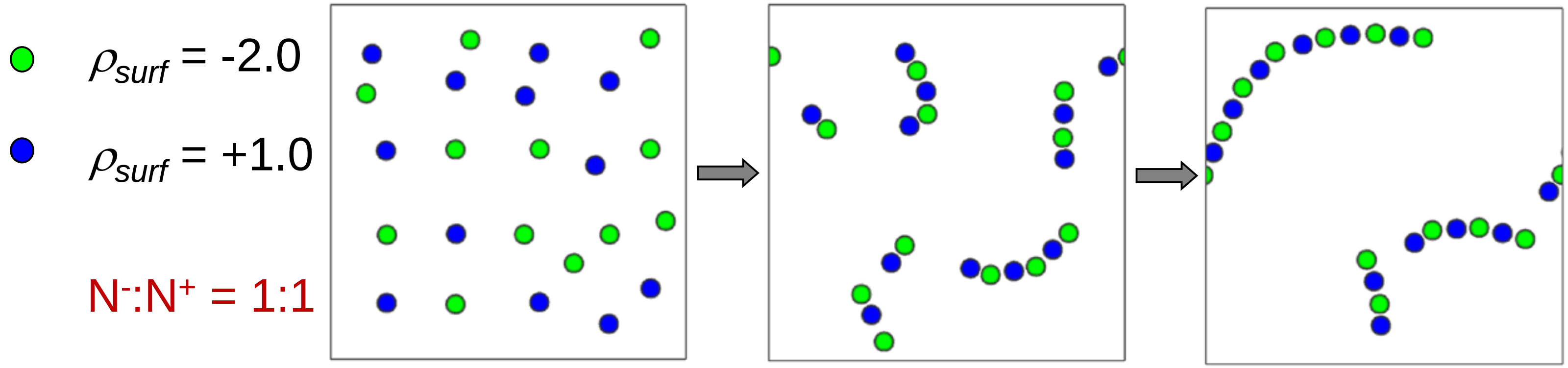
Self-Assembly Mechanisms





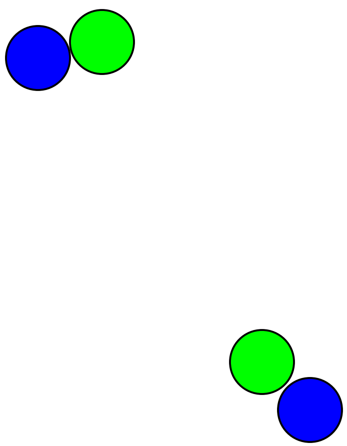
(1) neutral & symmetric (2) induced dipole (3) attraction (4) repeated growth & dipolar

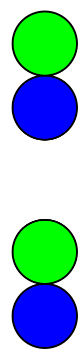
Self-Assembly Mechanisms

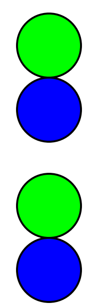


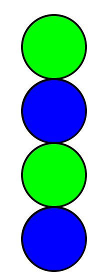
repel at long distance
attract at short distance

charged chain formation,
mutually repulsive,
as straight as possible





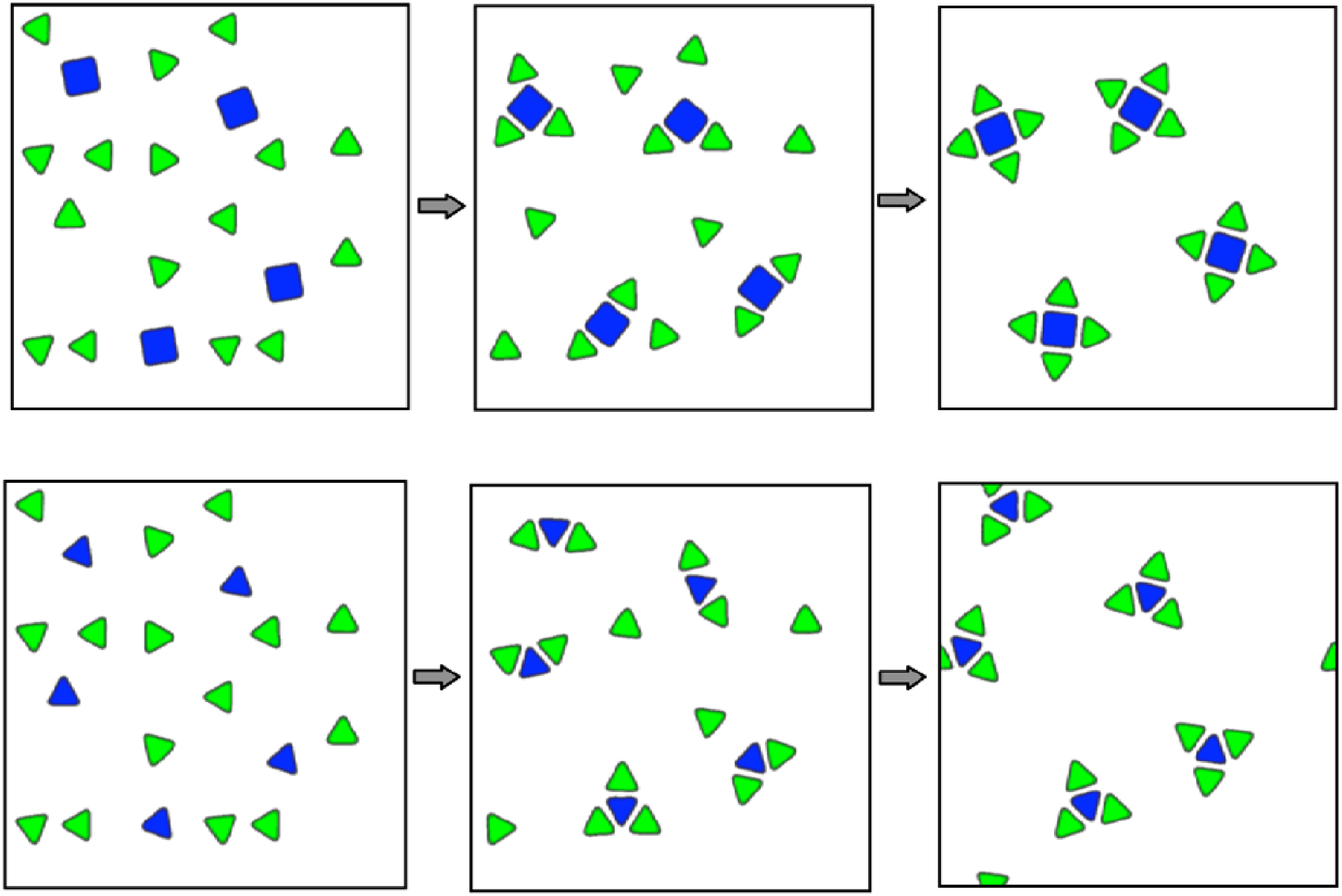




(1) charged & dipole (2) alignment (3) attraction (4) repeated growth & charged

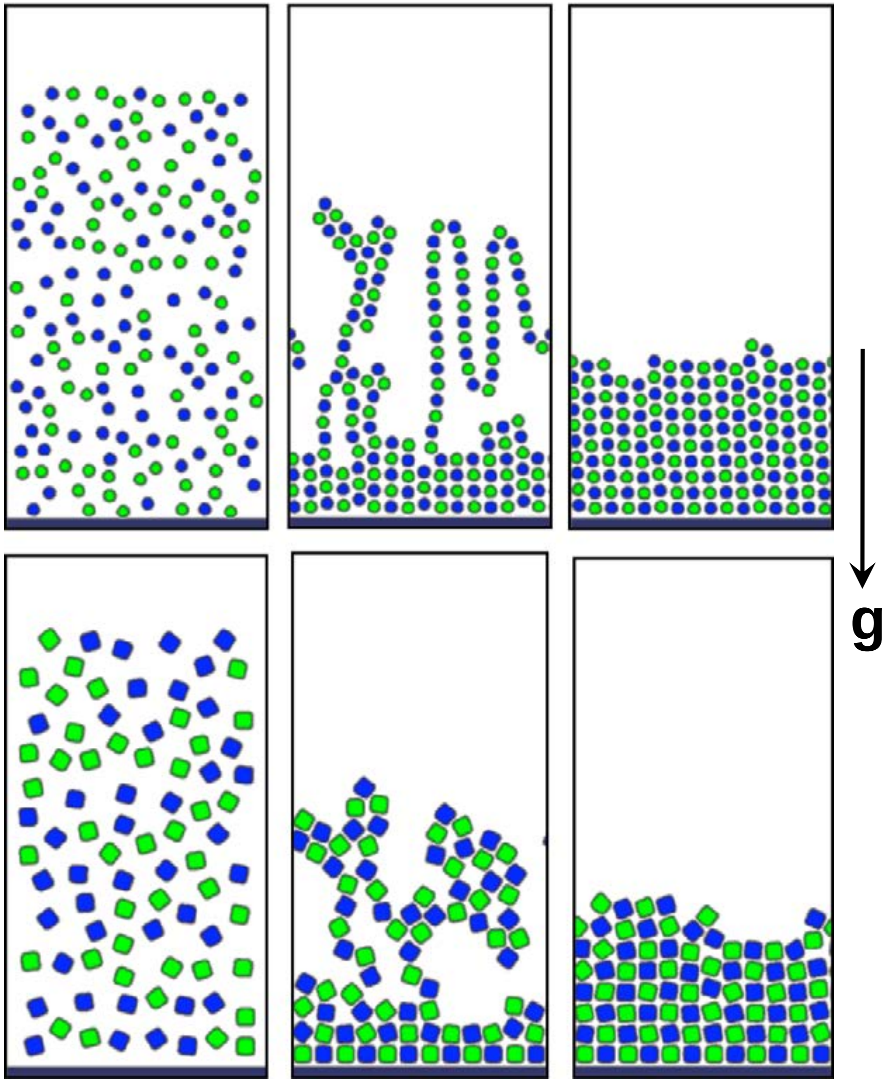
Simulation

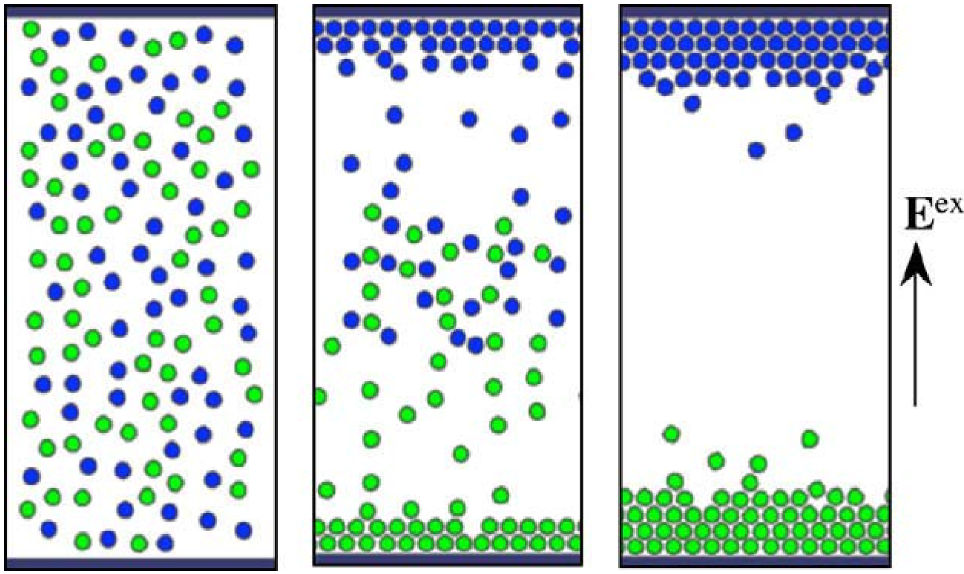
- Particles of opposite charges: non-spherical shapes



Simulation

- Stacking of charged particles under external fields





Model Formulation

- Long-range force: dipolar particles

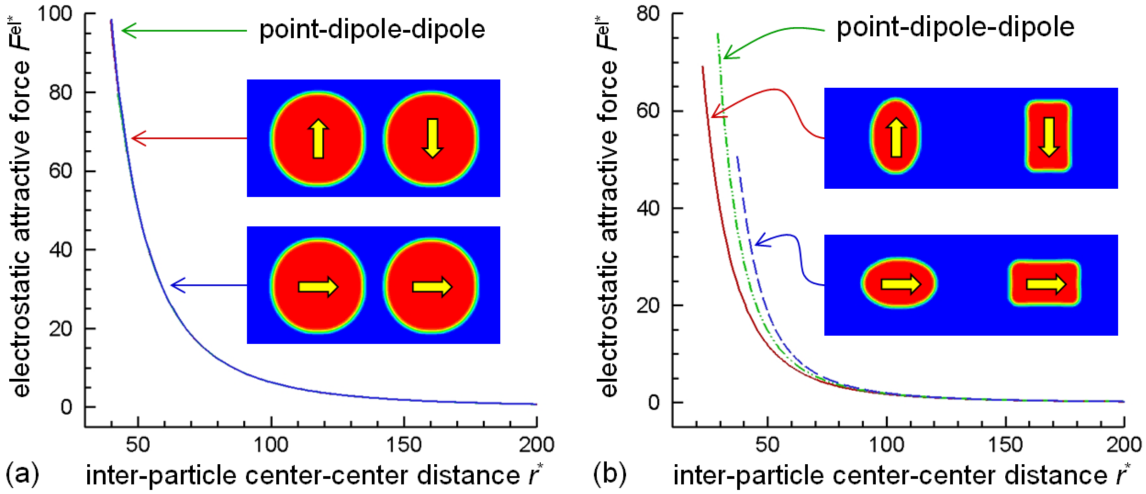
$$\mathbf{P}(\mathbf{r},t;\alpha)=\mathbf{P}(t;\alpha)\,\eta(\mathbf{r},t;\alpha)$$

$$\mathbf{P}(\mathbf{r},t)=\sum_{\alpha}\mathbf{P}(\mathbf{r},t;\alpha)$$

$$\mathbf{E}(\mathbf{r})=\mathbf{E}^{\mathrm{ex}}-\frac{1}{\varepsilon_0}\int\frac{d^3k}{(2\pi)^3}\left[\mathbf{n}\cdot\tilde{\mathbf{P}}(\mathbf{k})\right]\mathbf{n}\,e^{i\mathbf{k}\cdot\mathbf{r}}$$

$$\mathbf{F}^{\mathrm{el}}(\alpha)=\int_V\left[\mathbf{P}(\alpha)\cdot\nabla\right]\mathbf{E}(\mathbf{r})\,\eta(\mathbf{r};\alpha)\,d^3r$$

$$\mathbf{T}^{\mathrm{el}}(\alpha)=\int_V\mathbf{P}(\alpha)\times\mathbf{E}(\mathbf{r})\,\eta(\mathbf{r};\alpha)\,d^3r+\int_V\left[\mathbf{r}-\mathbf{r}^{\mathrm{c}}(\alpha)\right]\times\left\{\left[\mathbf{P}(\alpha)\cdot\nabla\right]\mathbf{E}(\mathbf{r})\right\}\eta(\mathbf{r};\alpha)\,d^3r$$



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- Long-range force: magnetic particles

$$\mathbf{M}(\mathbf{r},t)=\sum_{\alpha}\mathbf{M}(t;\alpha)\,\eta(\mathbf{r},t;\alpha)$$

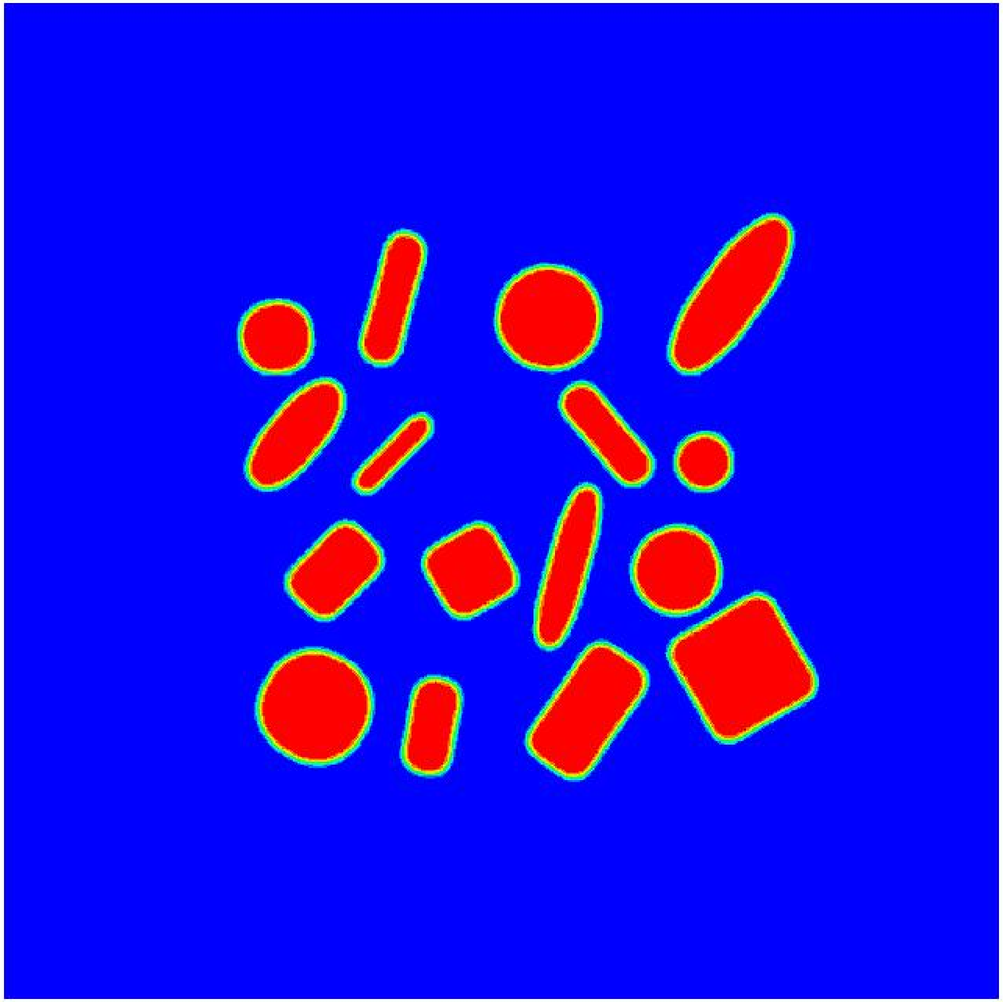
$$\mathbf{H}(\mathbf{r})=\mathbf{H}^{\mathrm{ex}}-\int\frac{d^3k}{(2\pi)^3}\left[\mathbf{n}\cdot\tilde{\mathbf{M}}(\mathbf{k})\right]\mathbf{n}\,e^{i\mathbf{k}\cdot\mathbf{r}}$$

$$\mathbf{F}^{\mathrm{mag}}(\alpha)=\mu_0\int_V\left[\mathbf{M}(\alpha)\cdot\nabla\right]\mathbf{H}(\mathbf{r})\,\eta(\mathbf{r};\alpha)\,d^3r$$

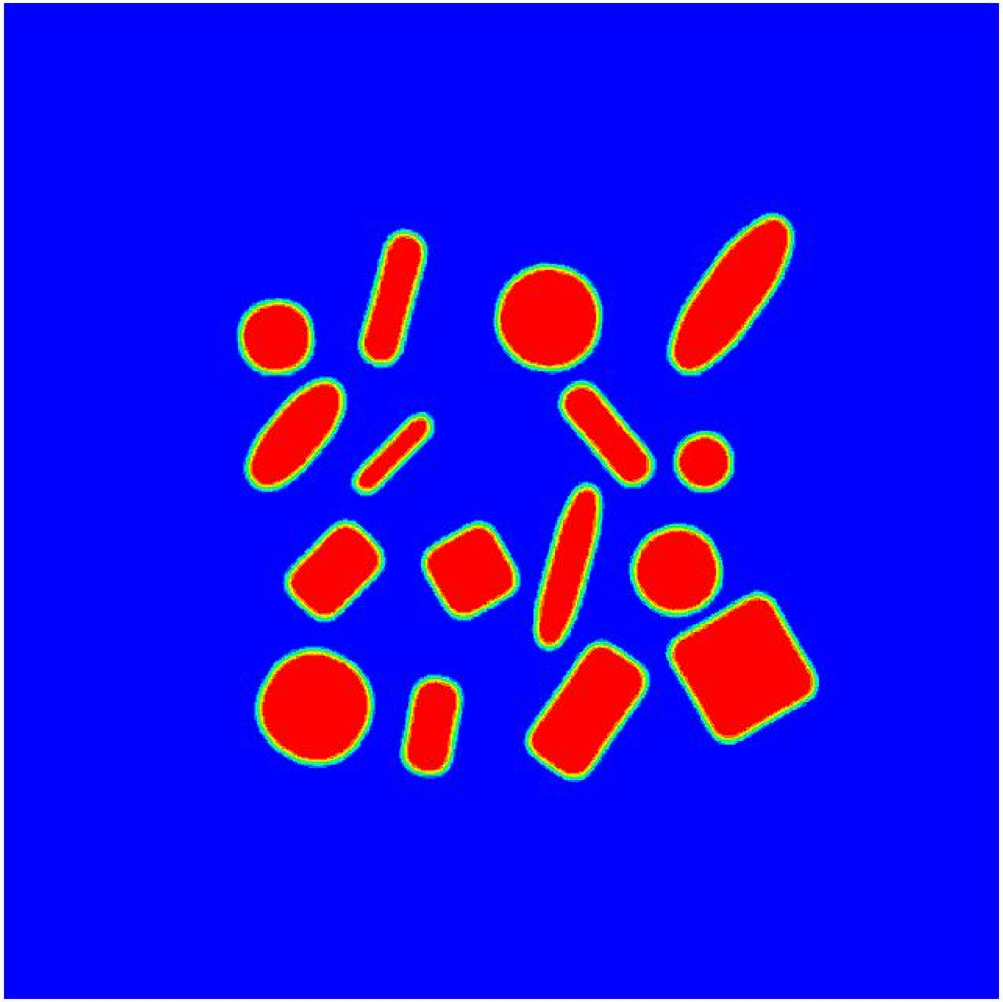
$$\mathbf{T}^{\mathrm{mag}}(\alpha)=\mu_0\int_V\mathbf{M}(\alpha)\times\mathbf{H}(\mathbf{r})\,\eta(\mathbf{r};\alpha)\,d^3r+\mu_0\int_V\left[\mathbf{r}-\mathbf{r}^{\mathrm{c}}(\alpha)\right]\times\left\{\left[\mathbf{M}(\alpha)\cdot\nabla\right]\mathbf{H}(\mathbf{r})\right\}\eta(\mathbf{r};\alpha)\,d^3r$$

Simulation

- Dipolar particles: agglomeration



Self-Assembly of Arbitrary-Shaped Dipolar Particles



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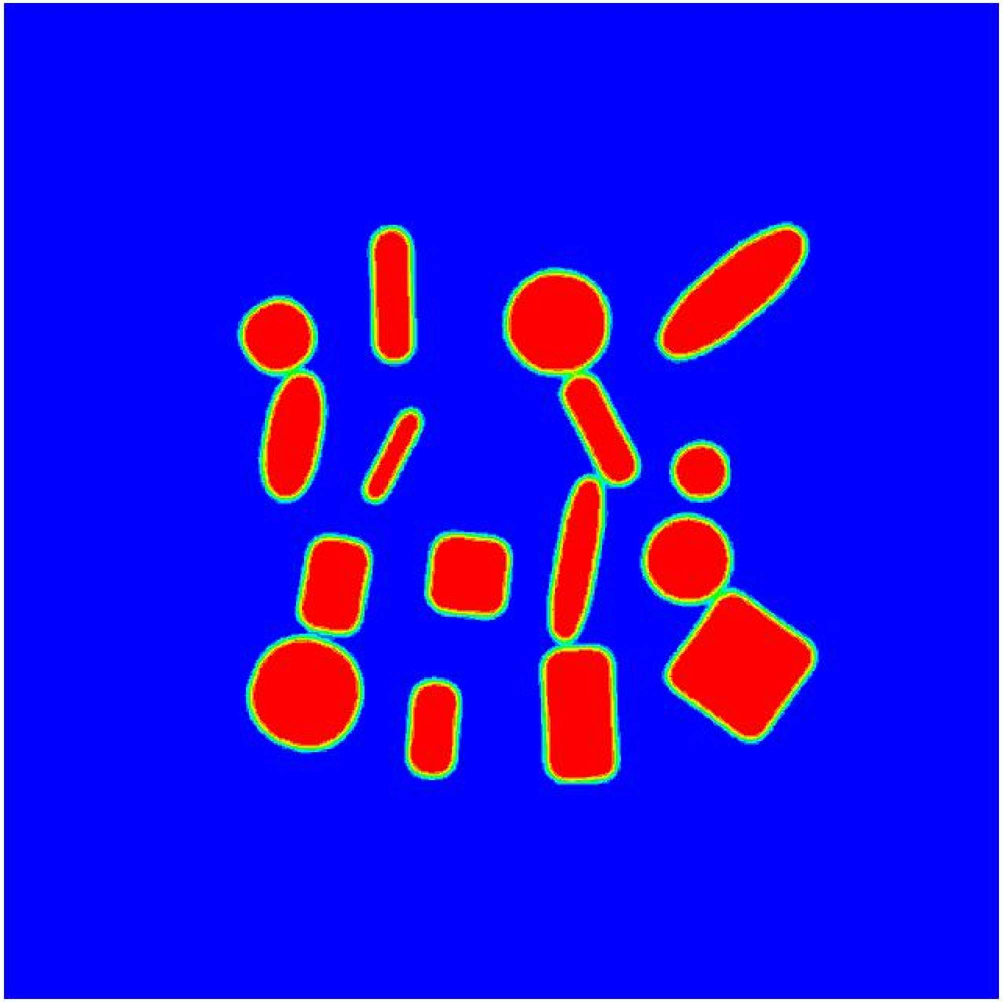

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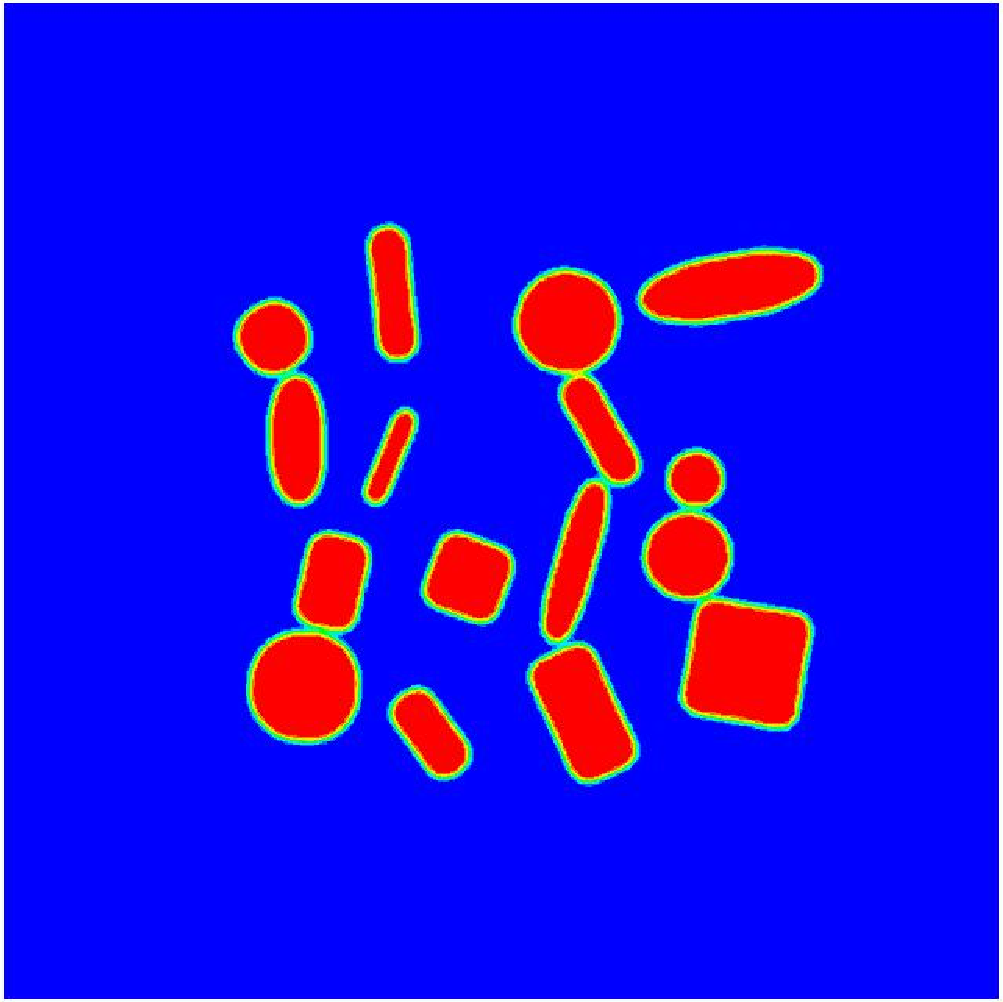

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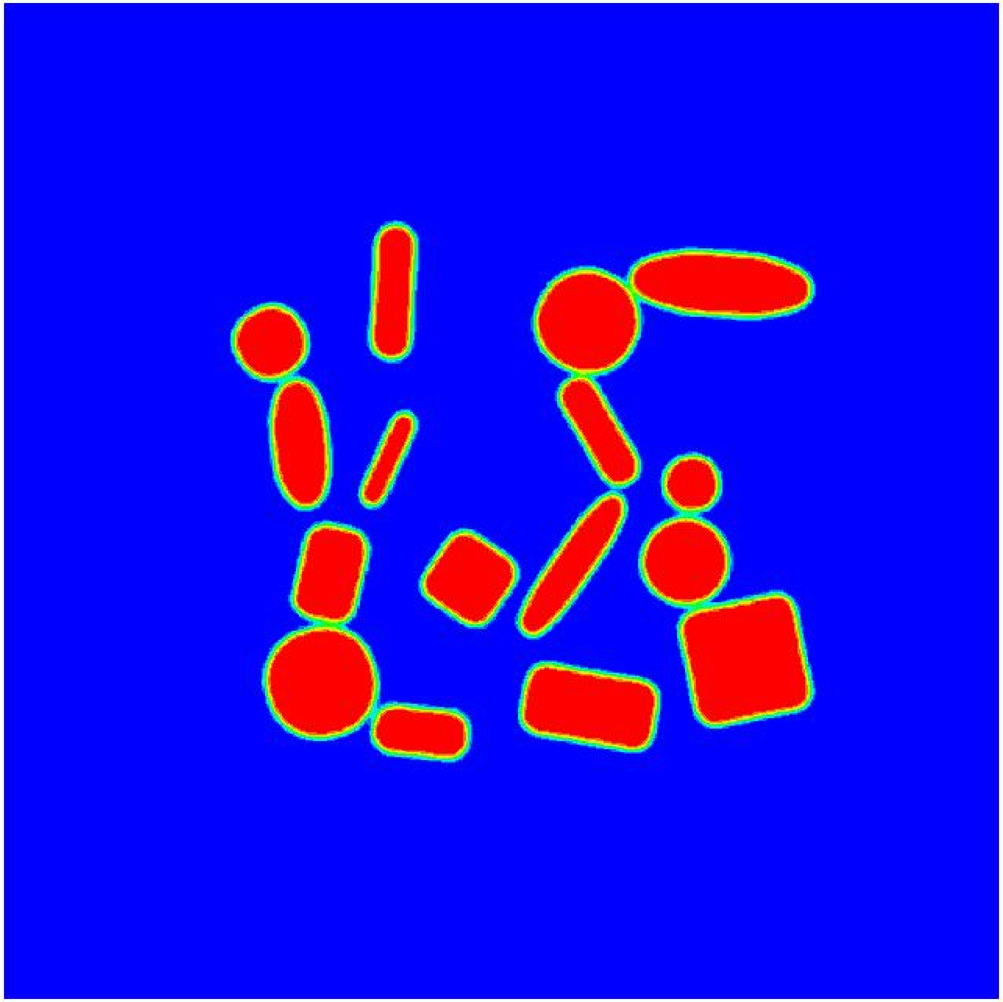

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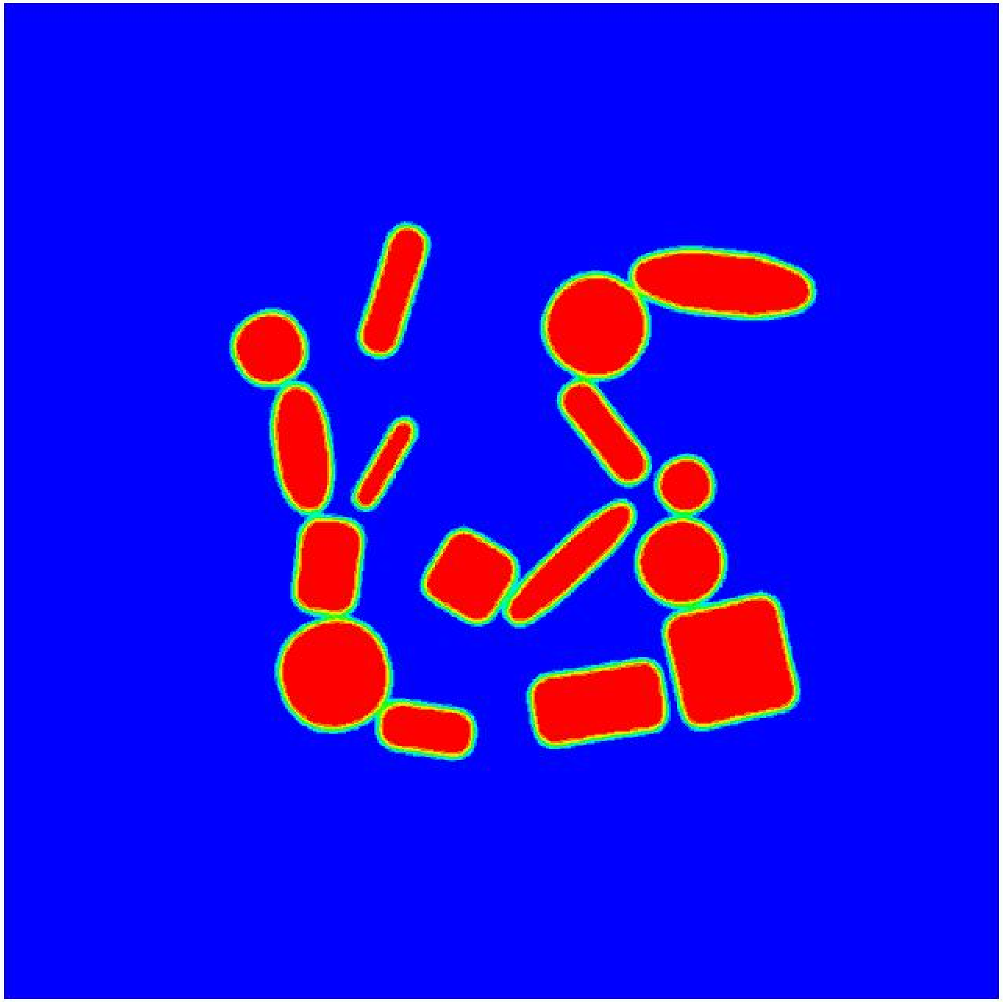

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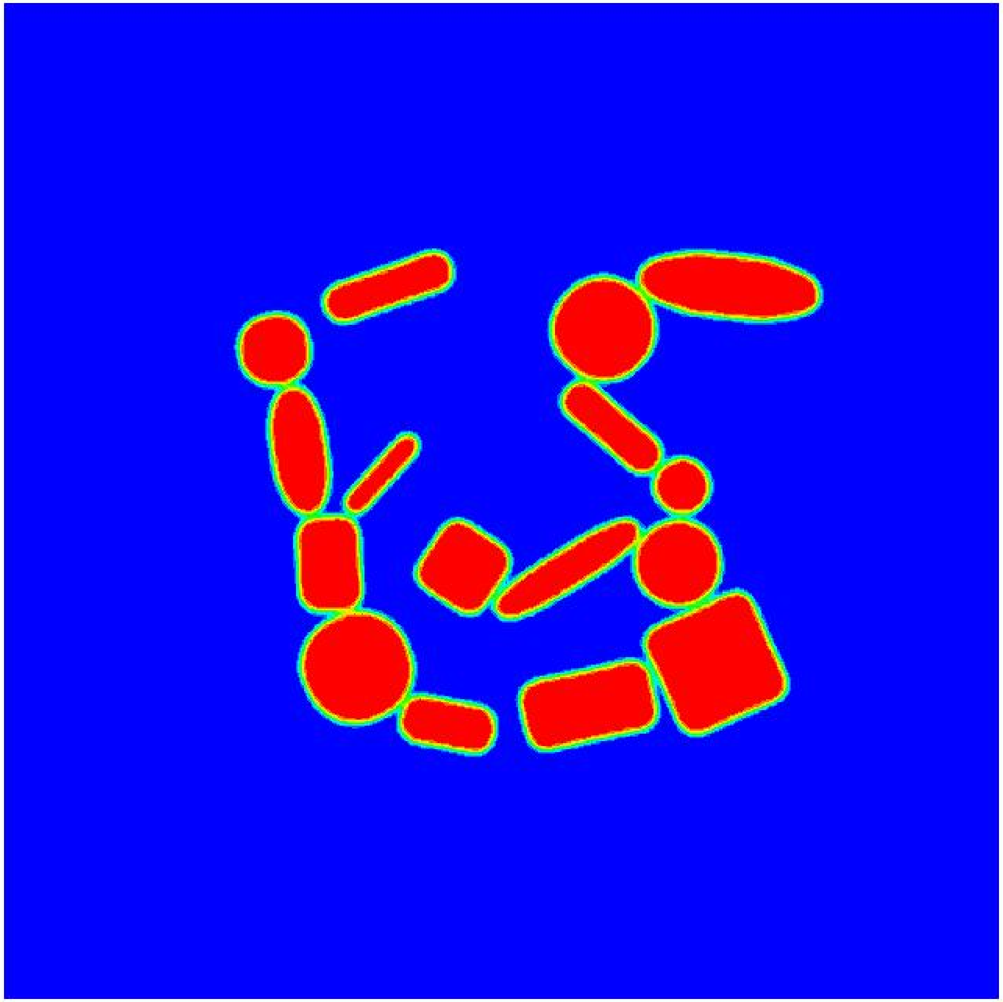

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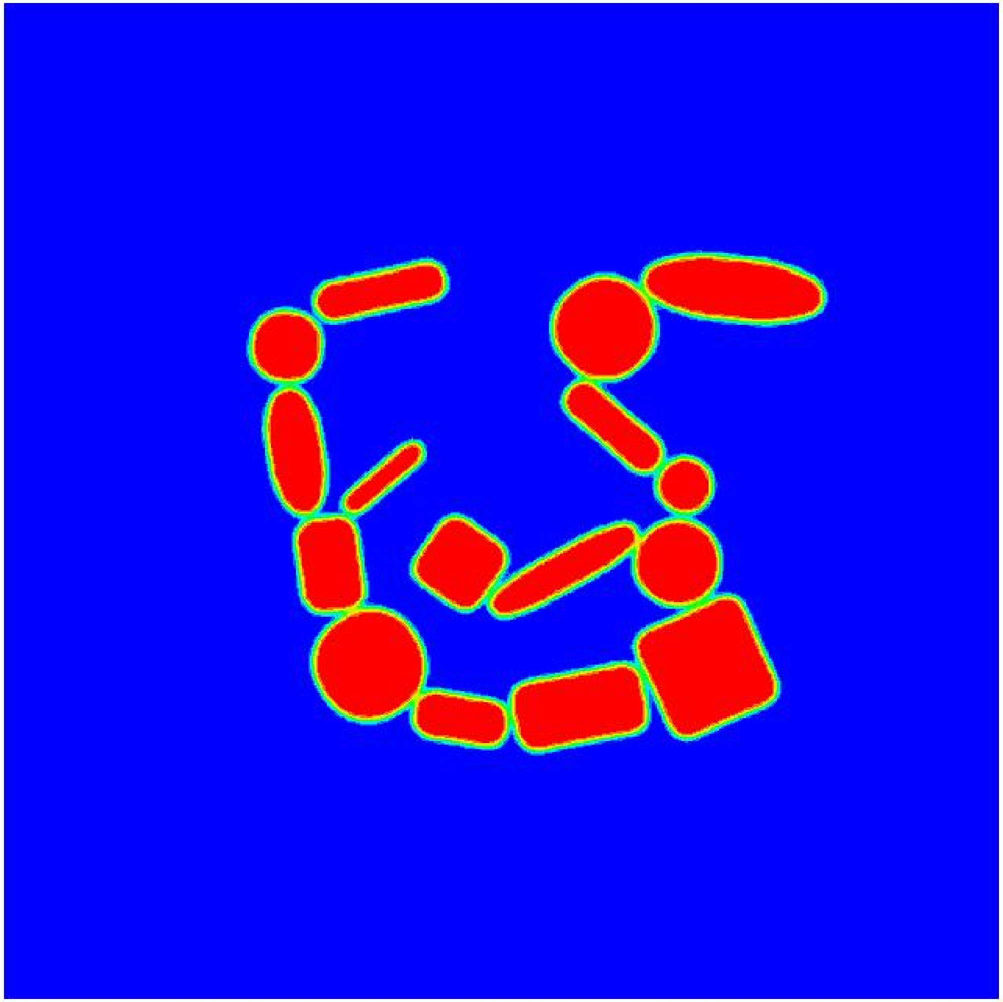

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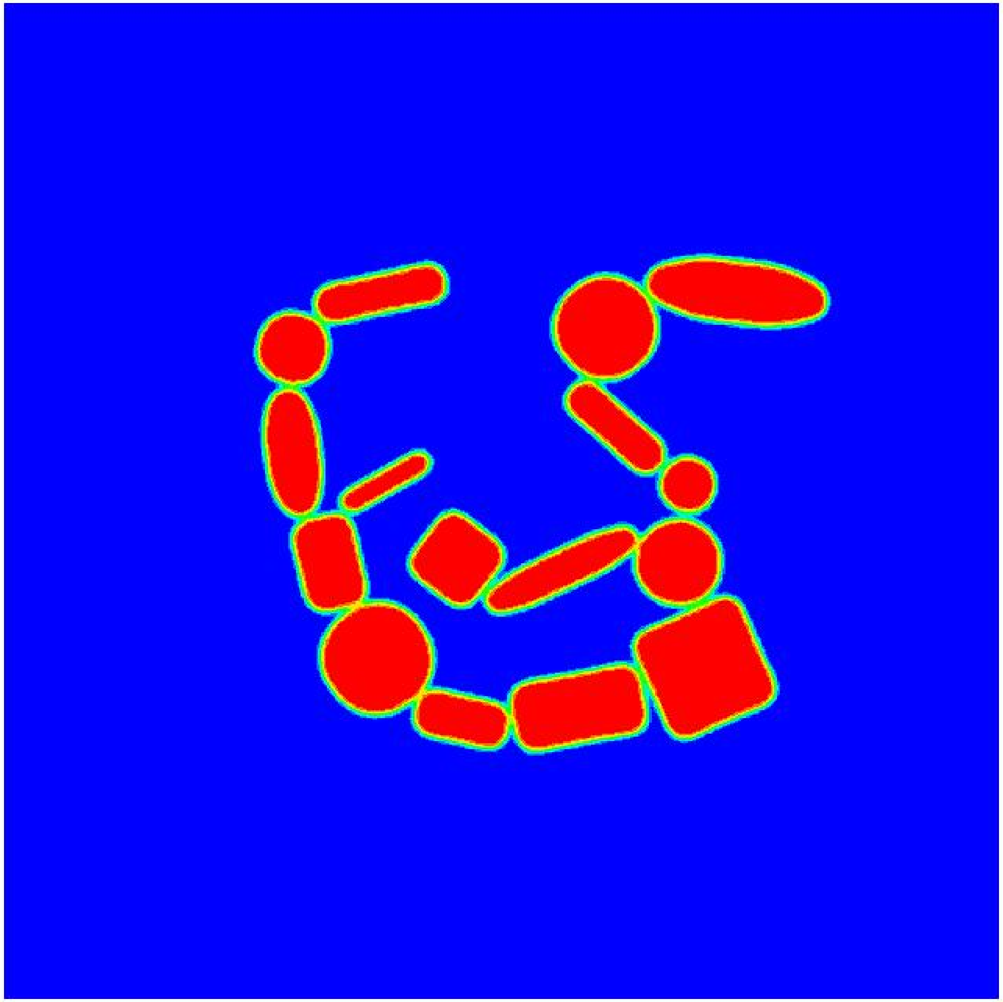

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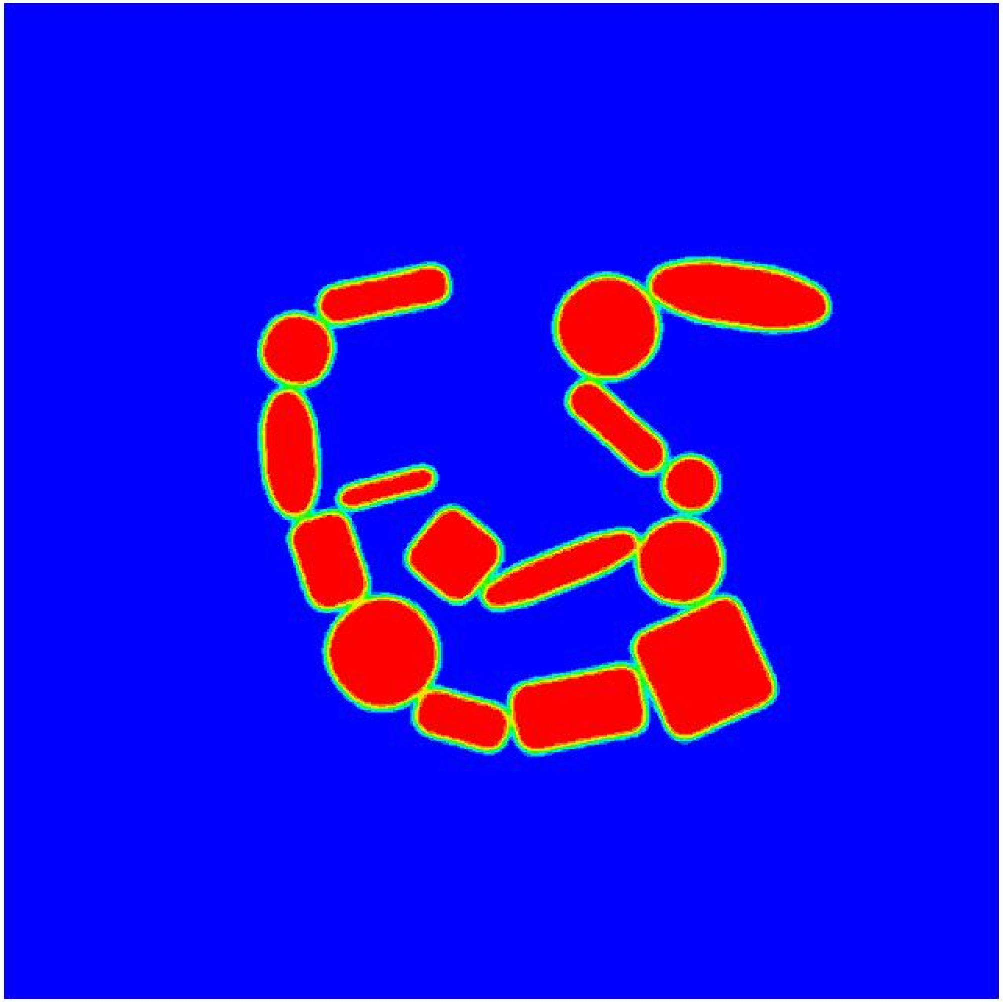

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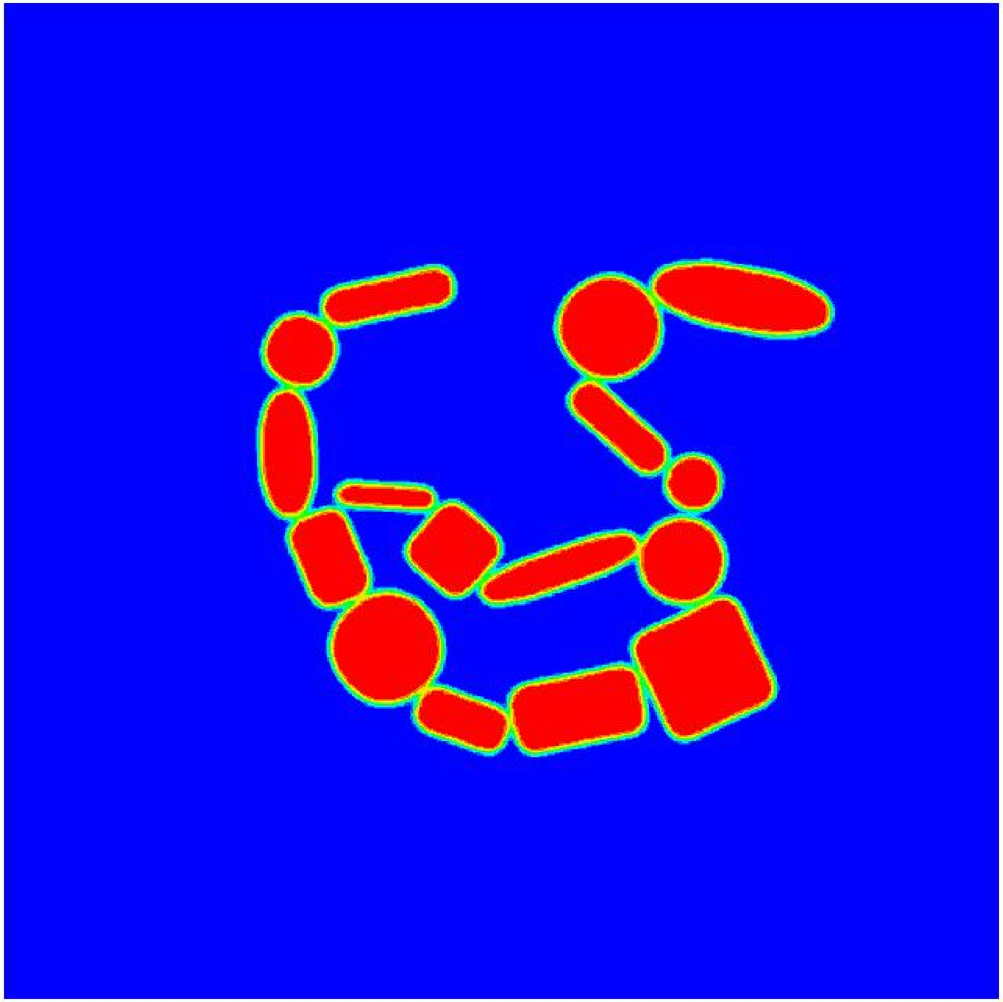

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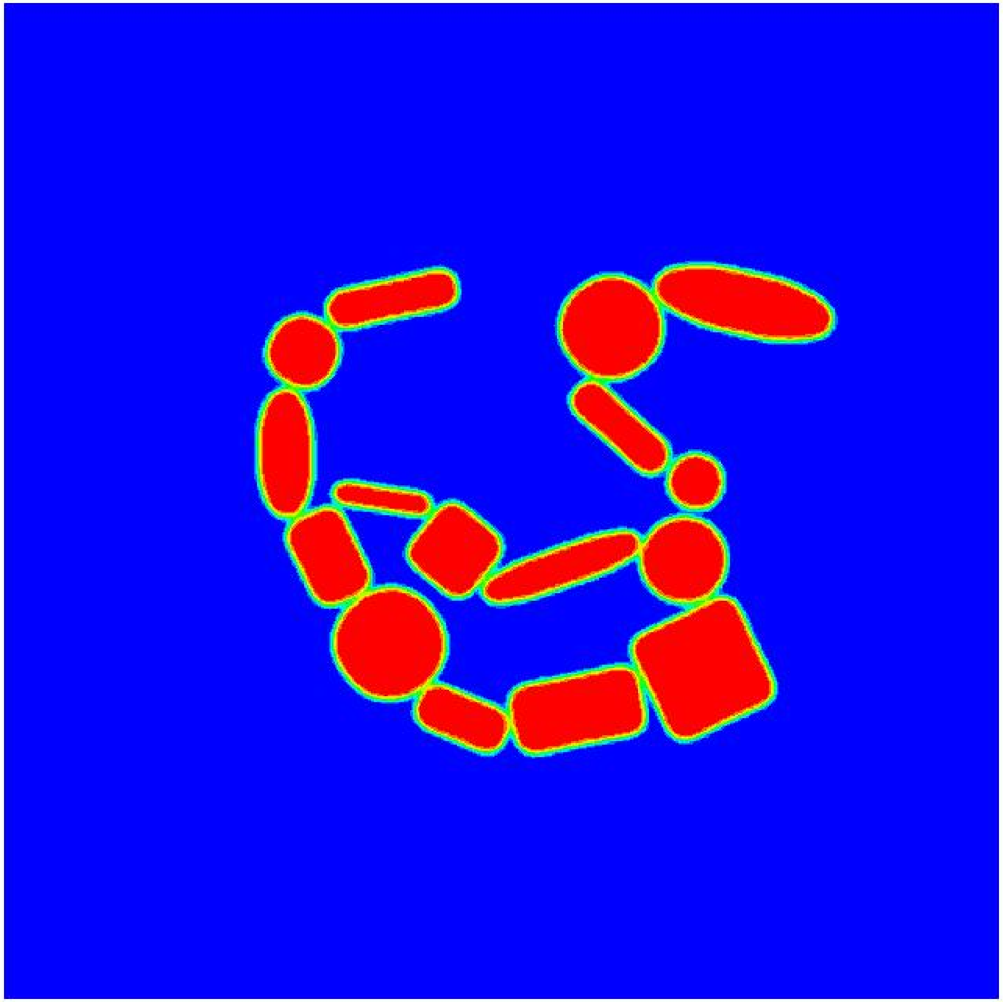

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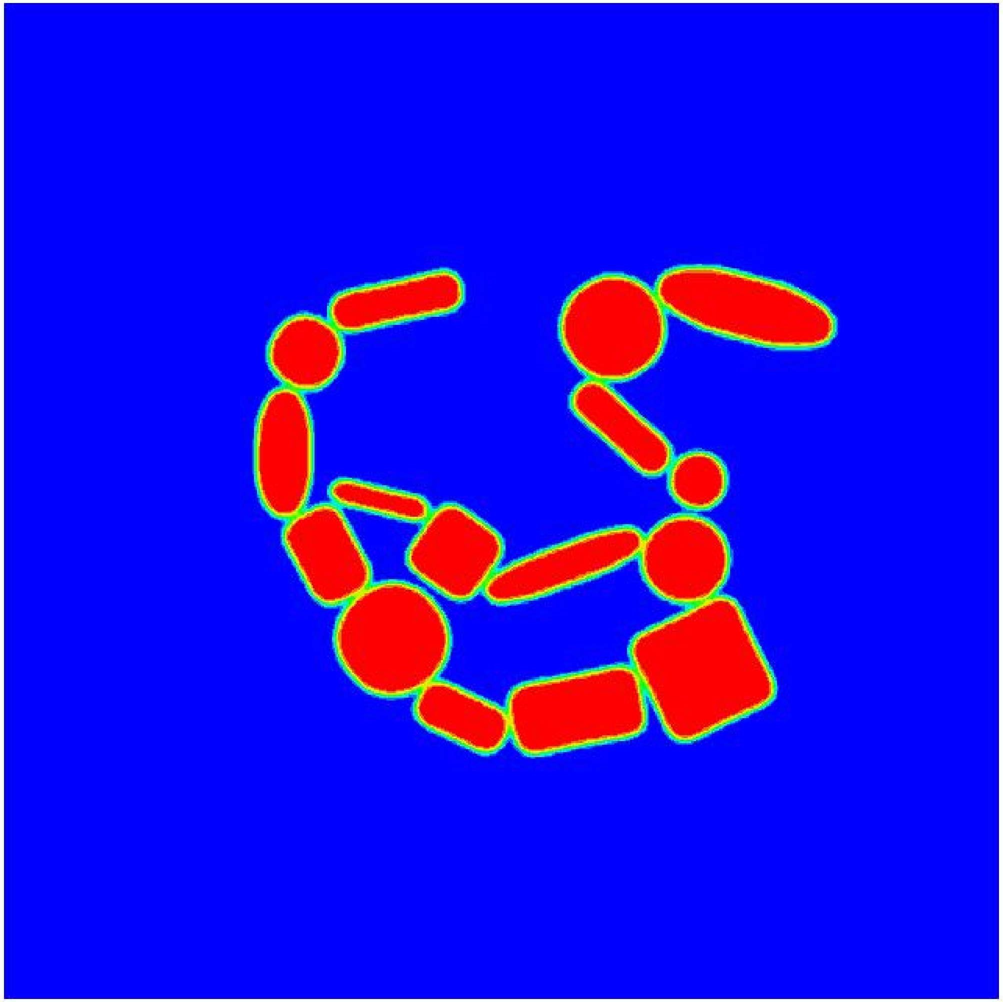

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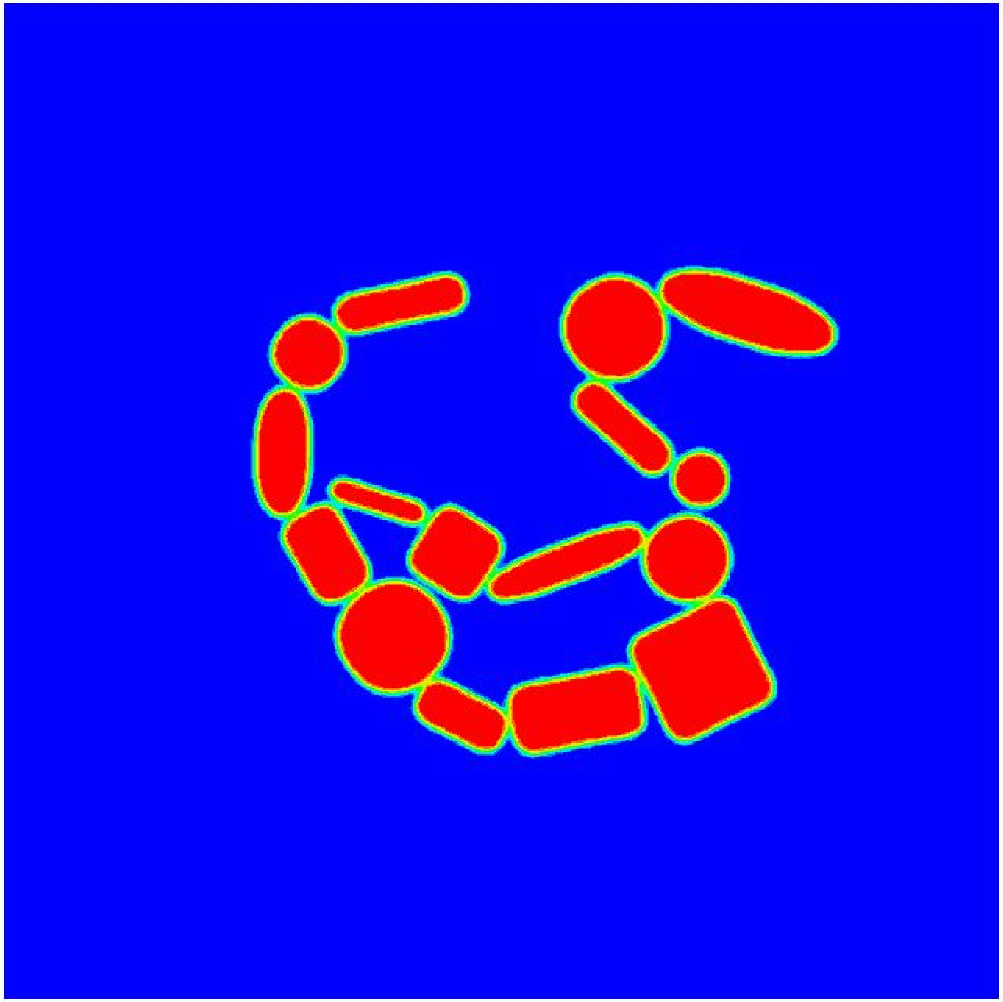

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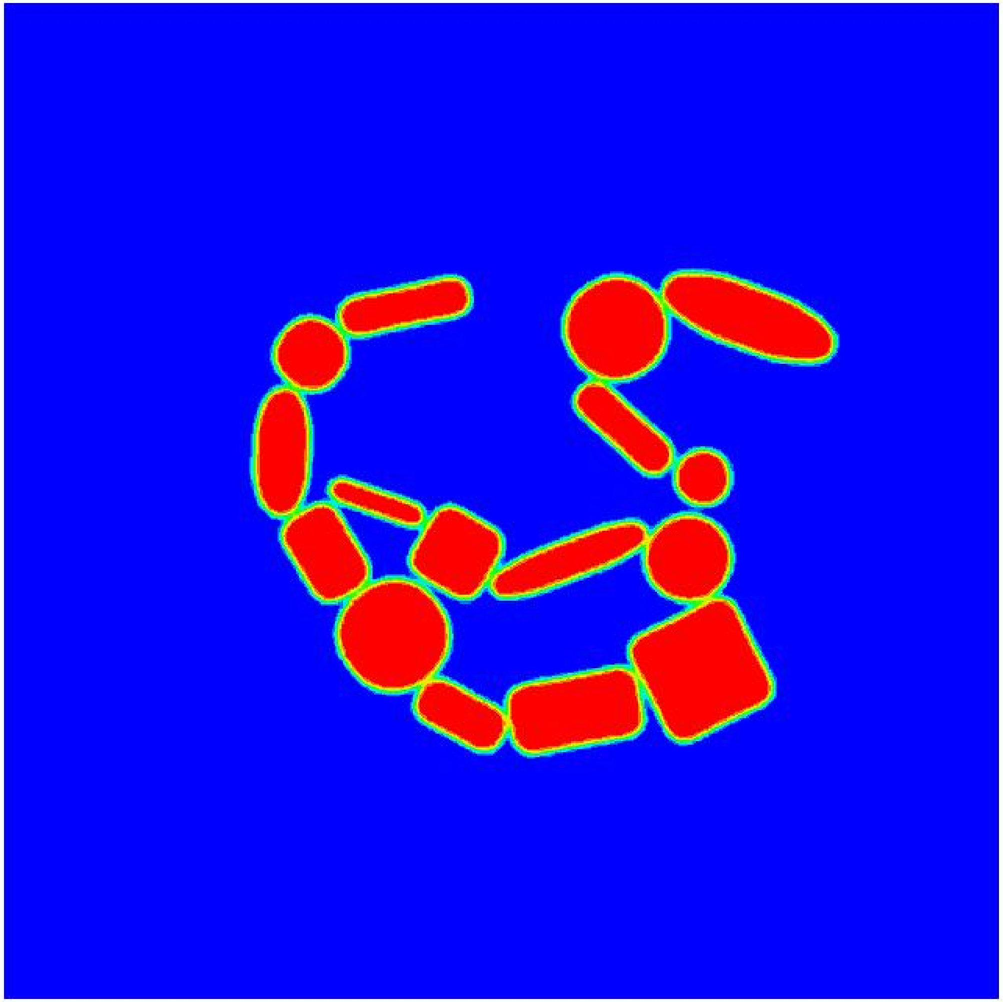

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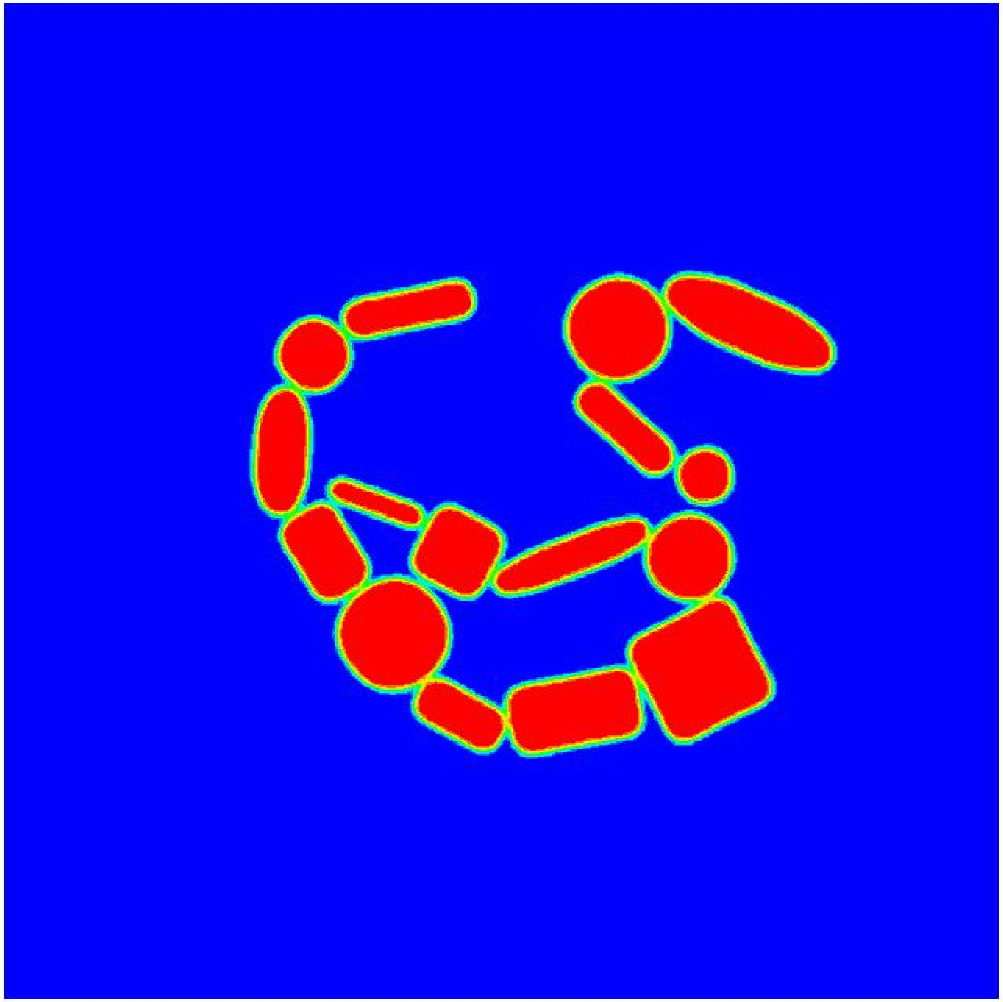

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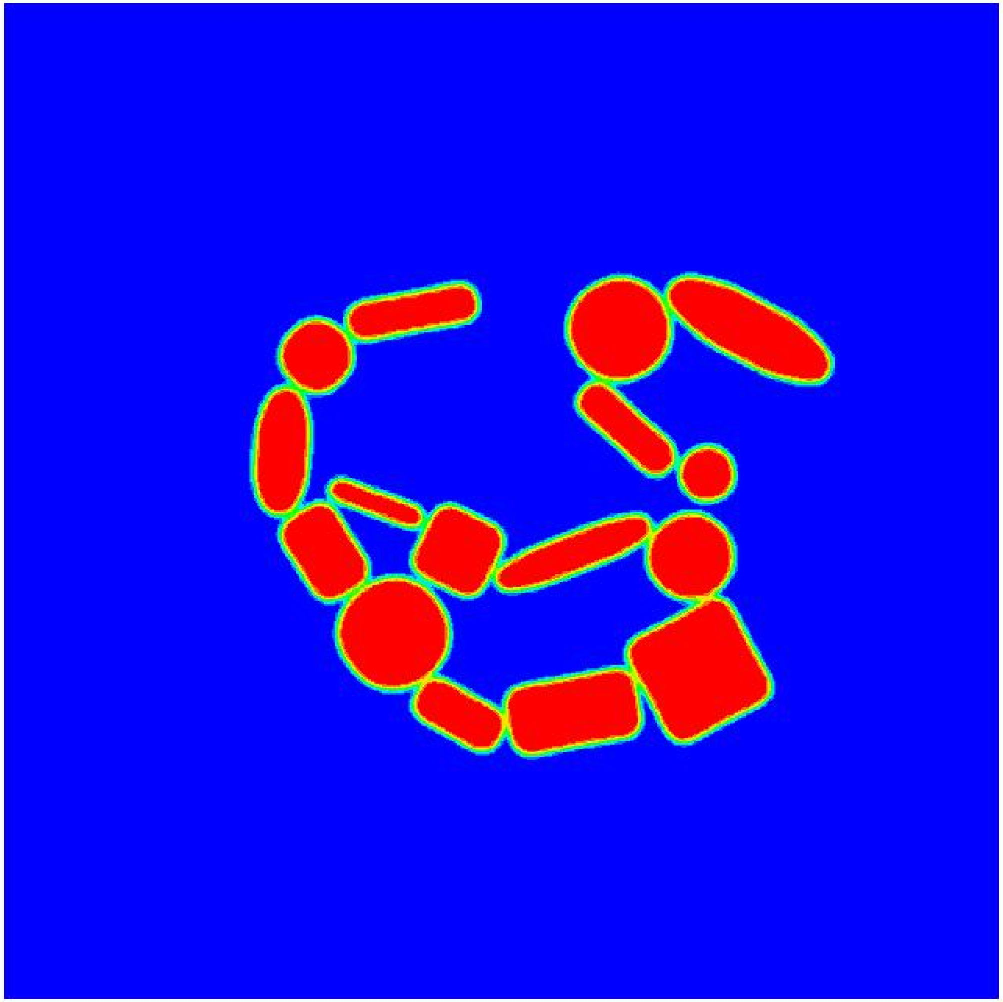

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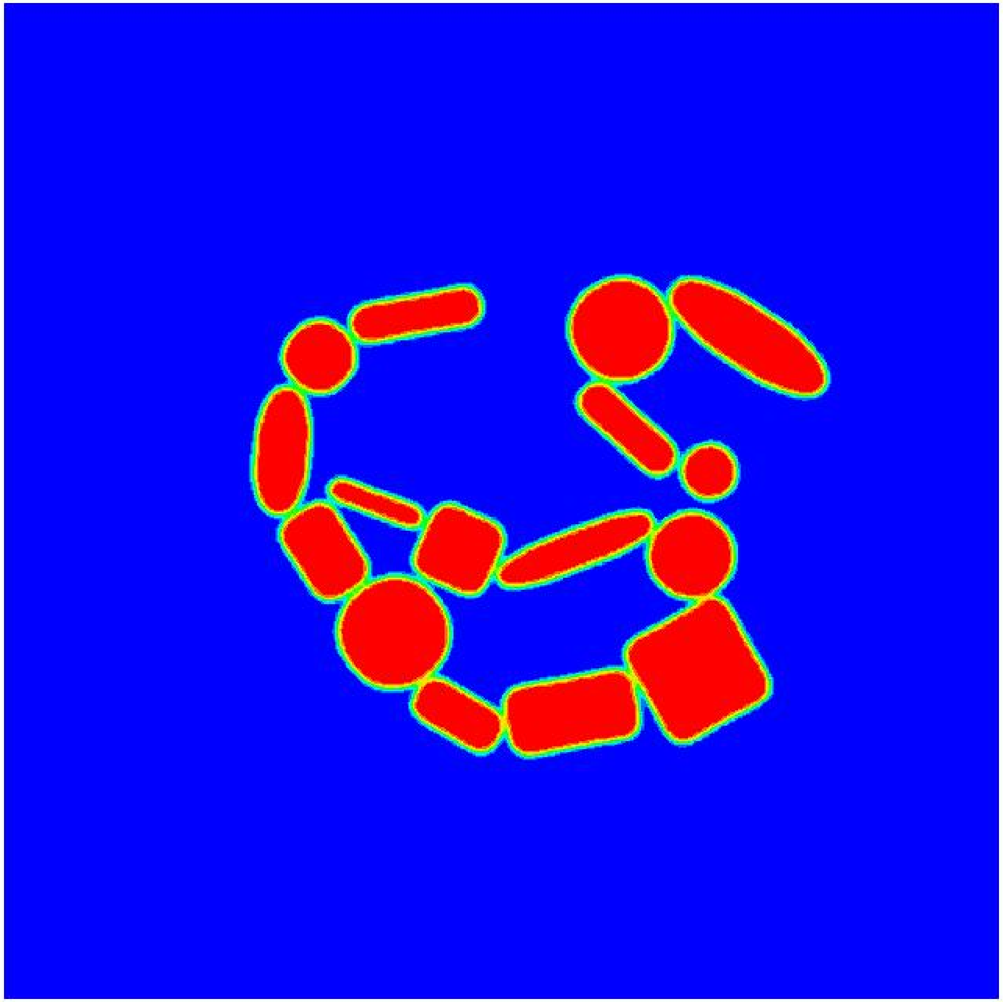

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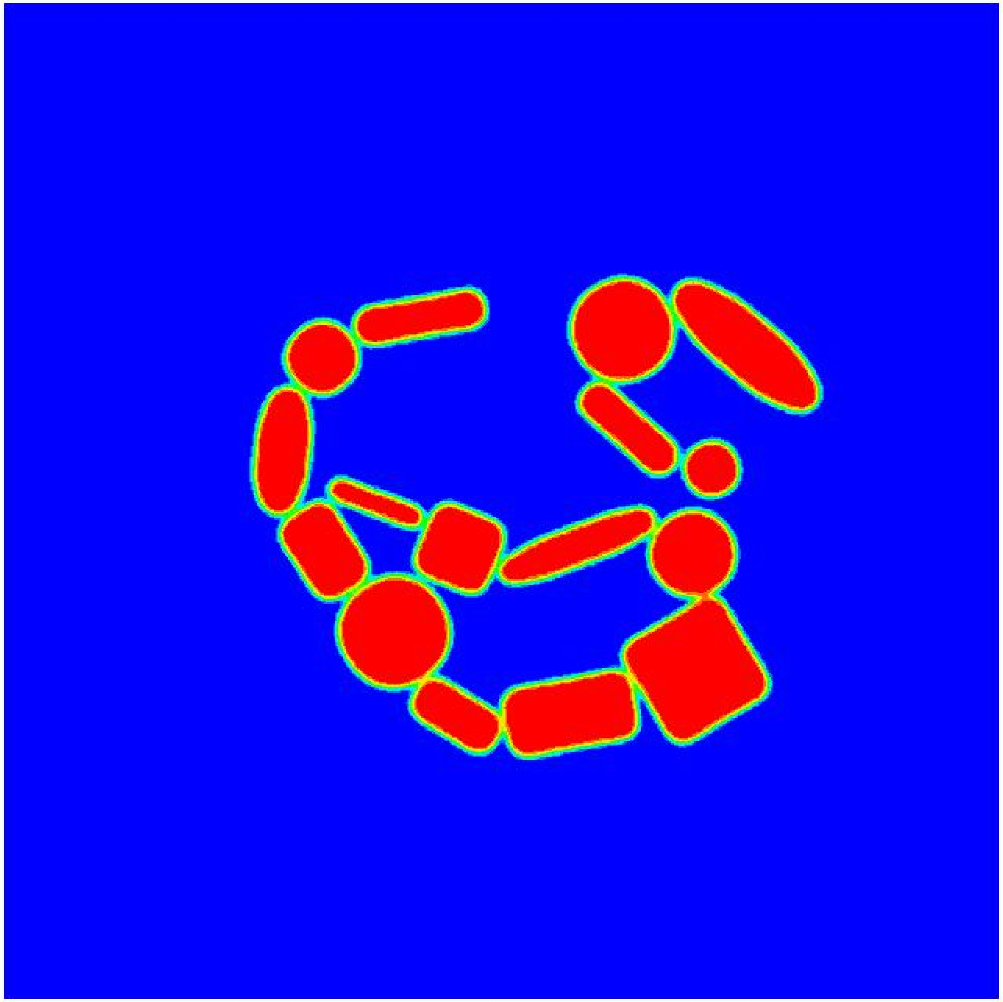

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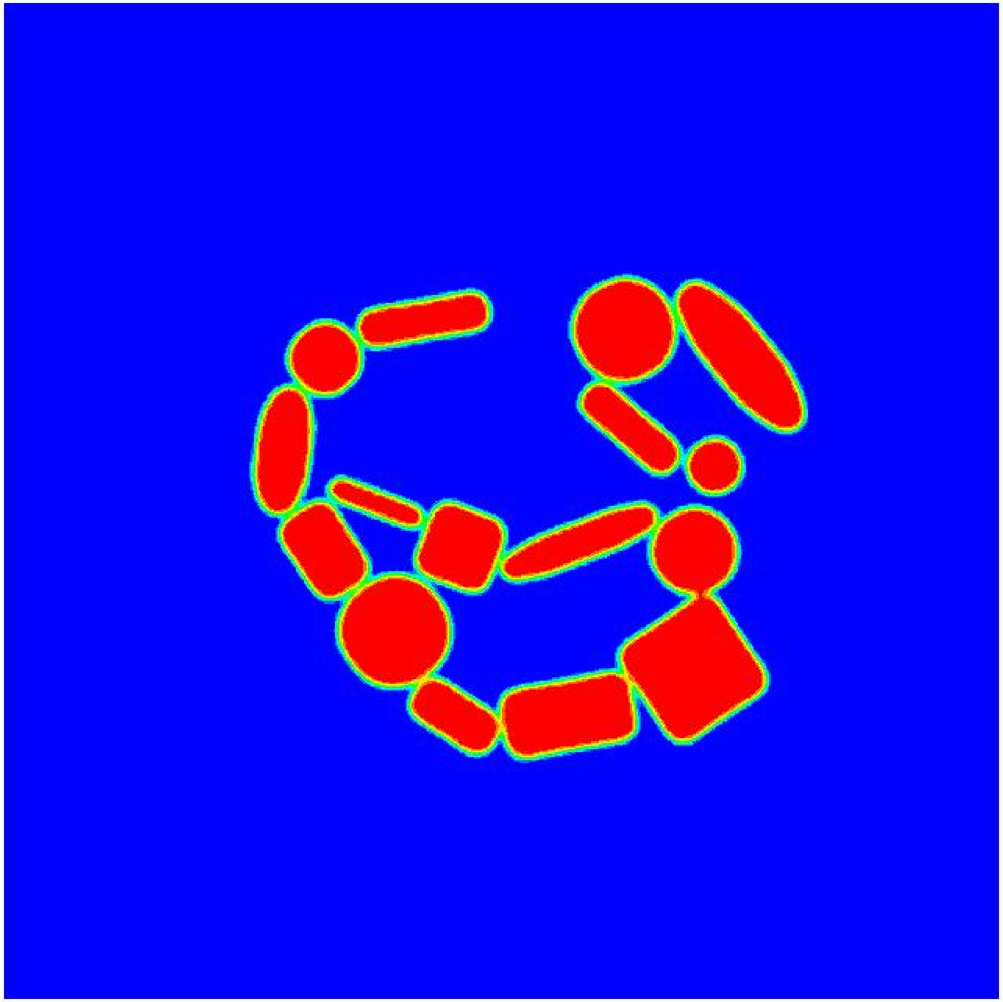

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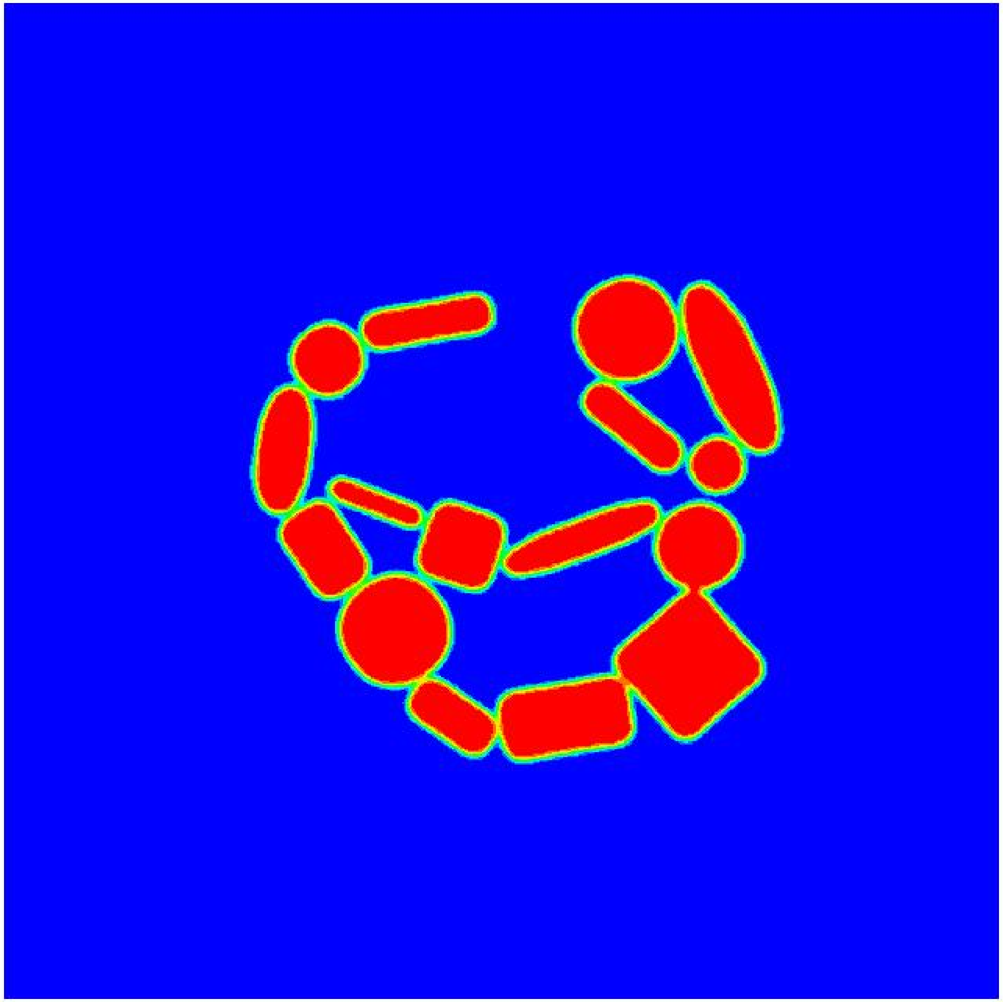
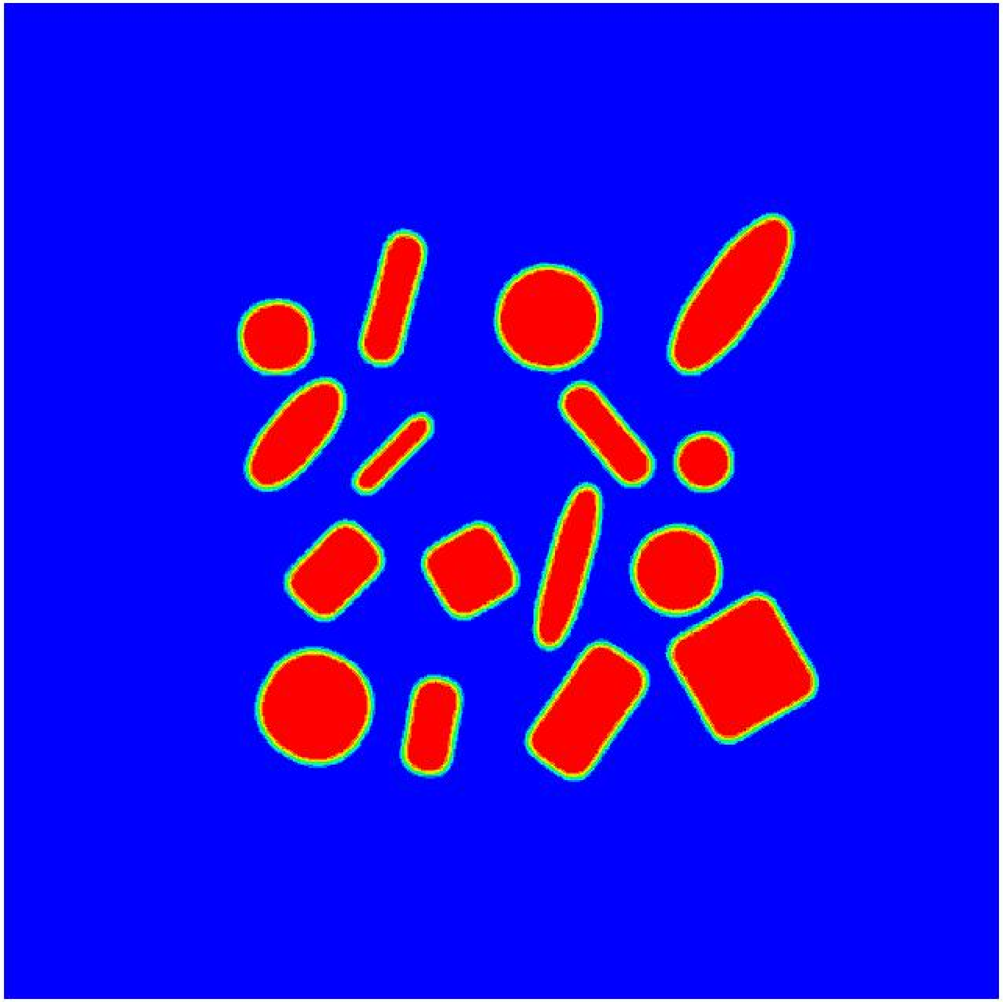
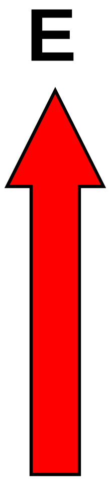


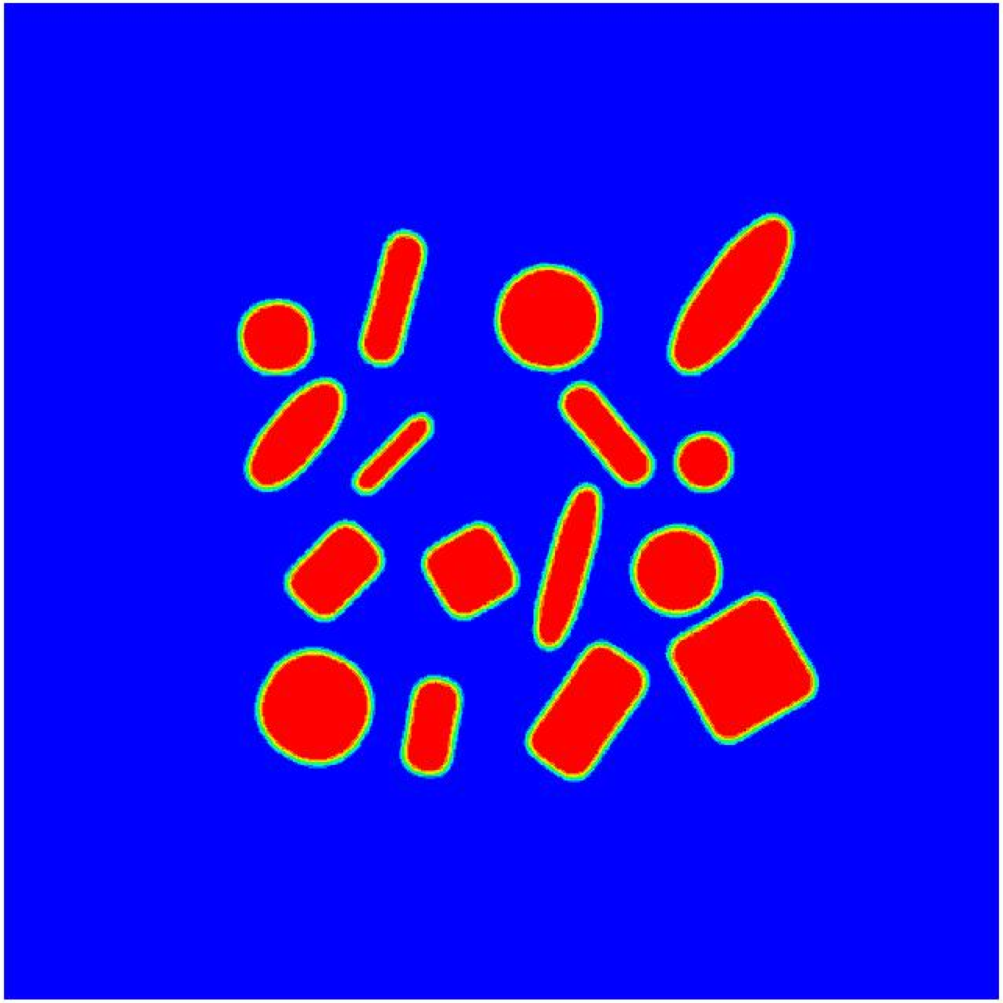
Simulation

- Dipolar particles: field-directed self-assembly



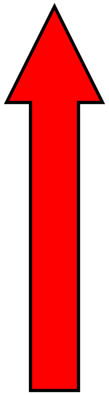
Self-Assembly of Arbitrary-Shaped Dipolar Particles

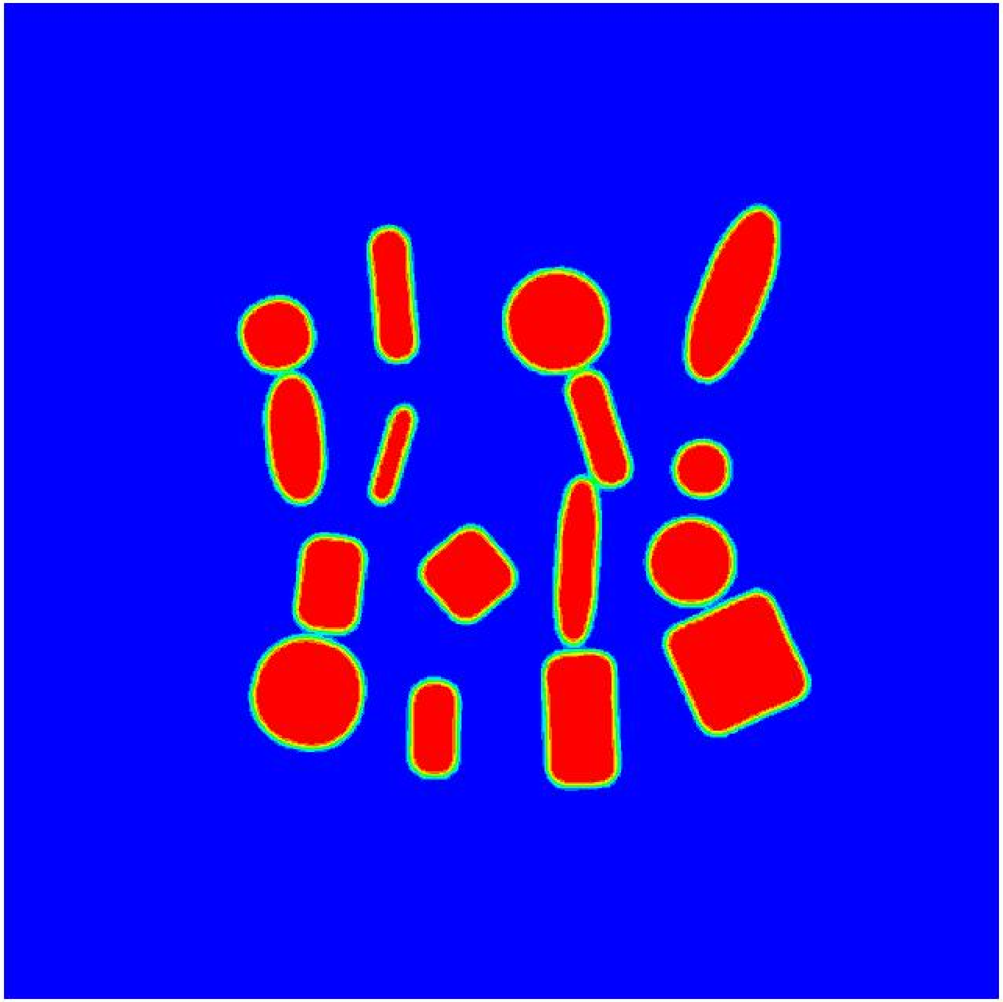




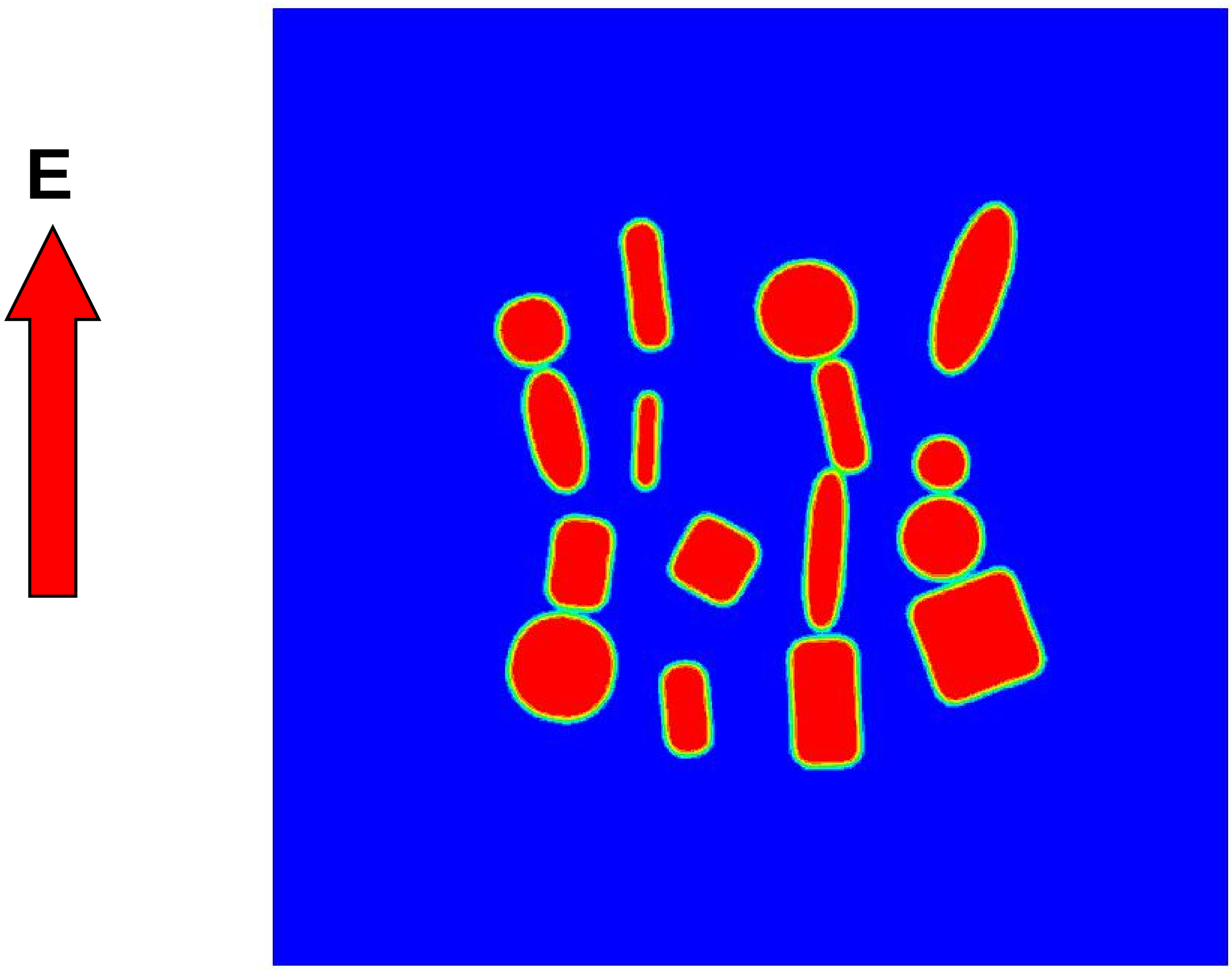
Self-Assembly of Arbitrary-Shaped Dipolar Particles

E





Self-Assembly of Arbitrary-Shaped Dipolar Particles



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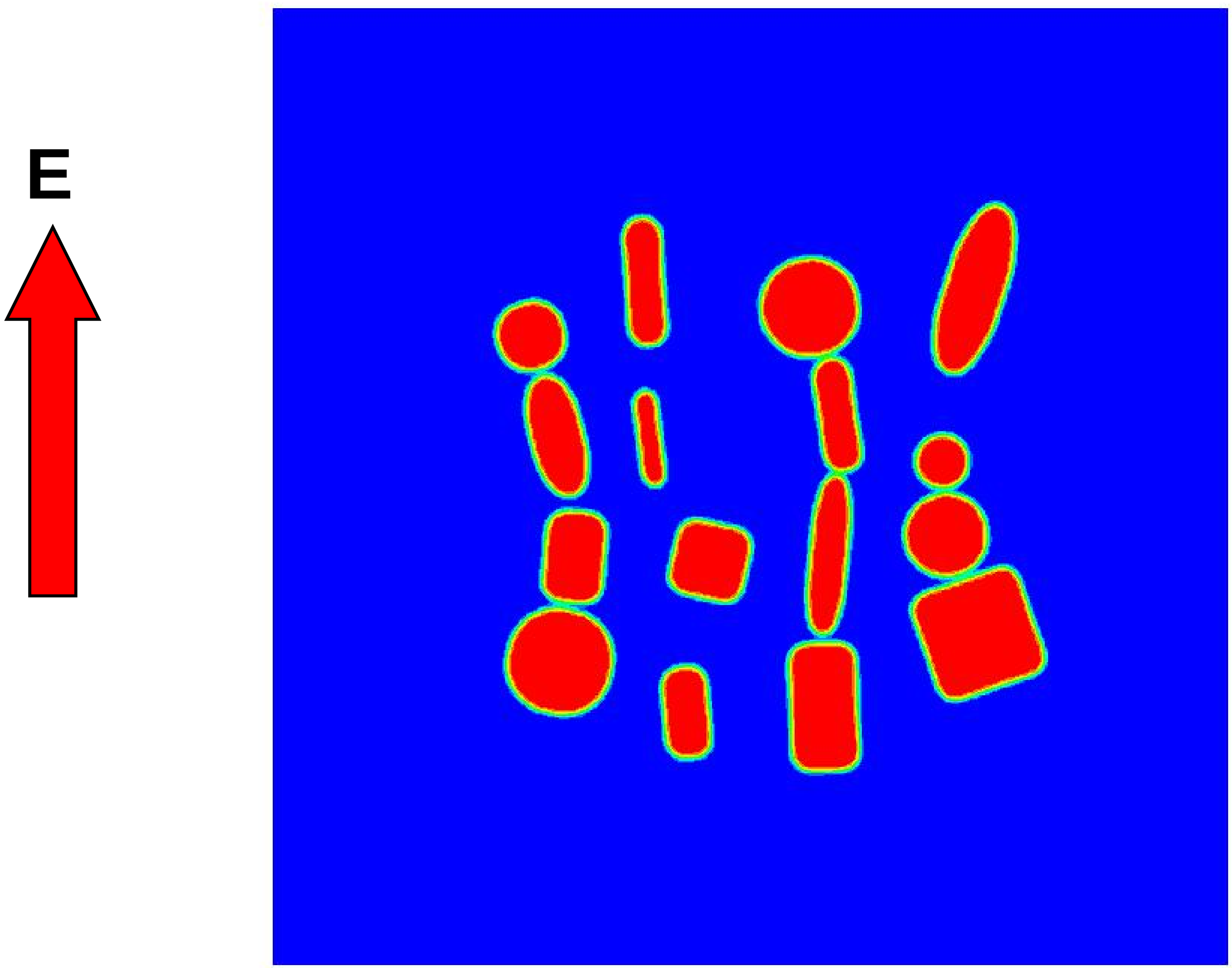

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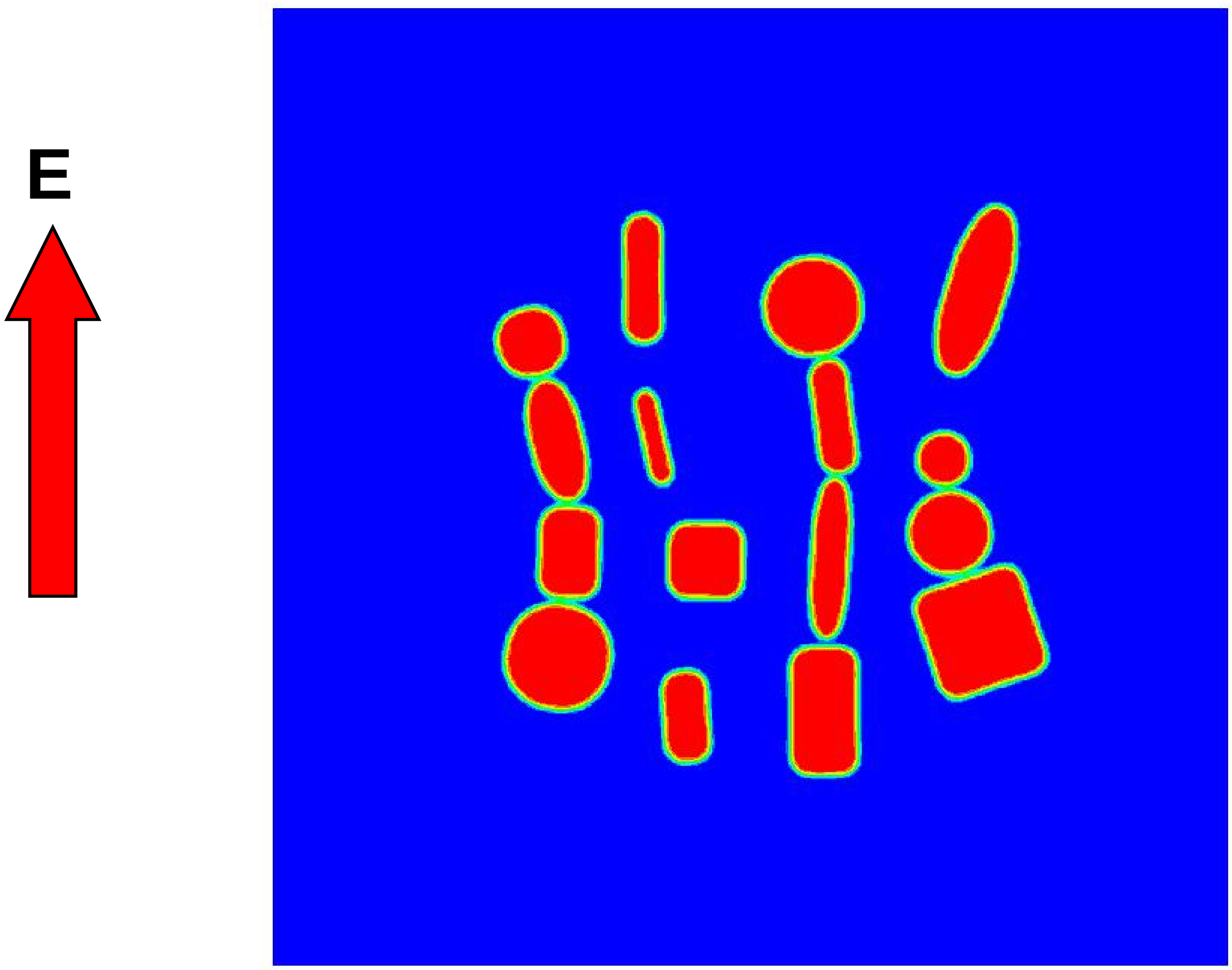

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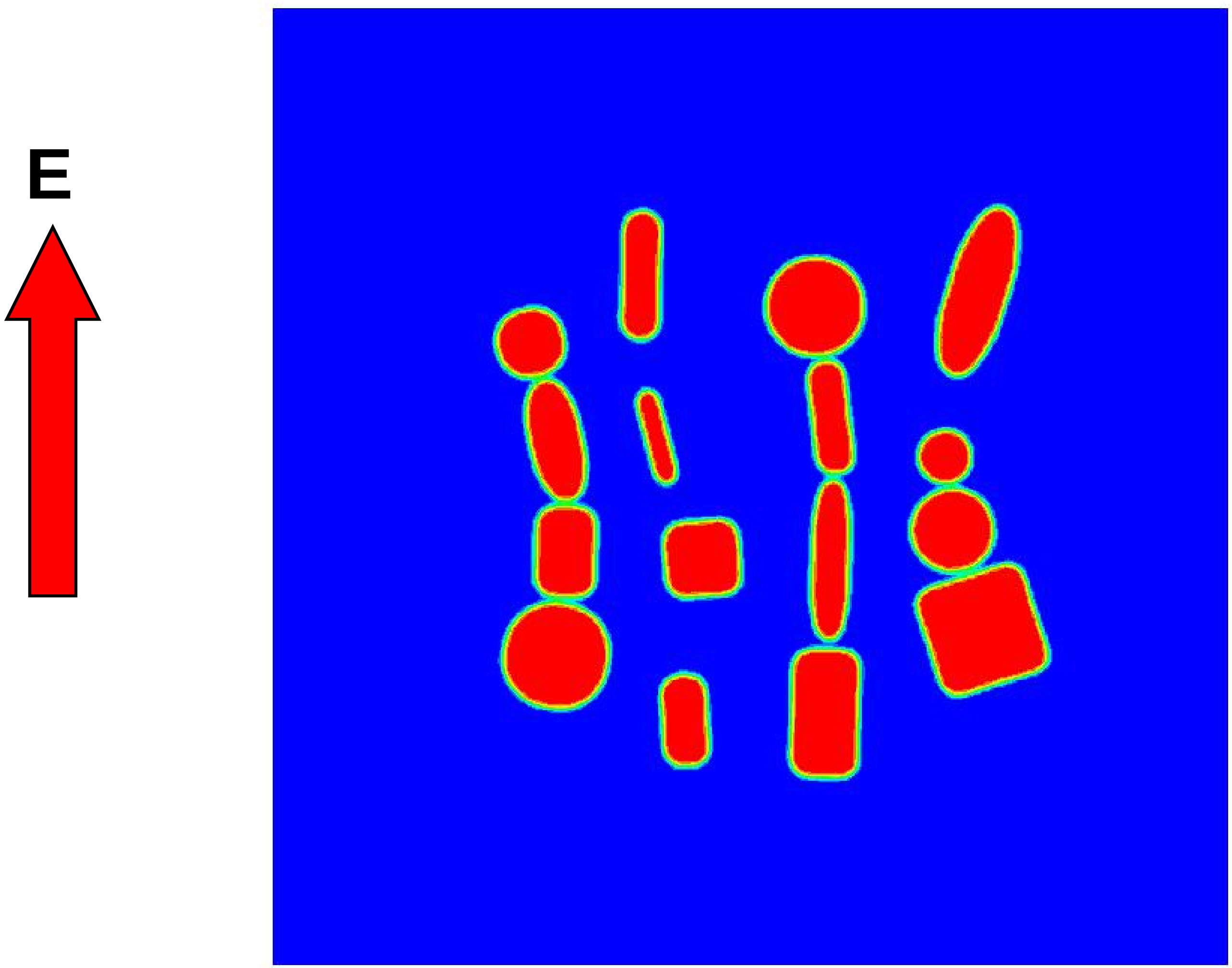

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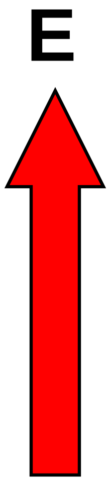

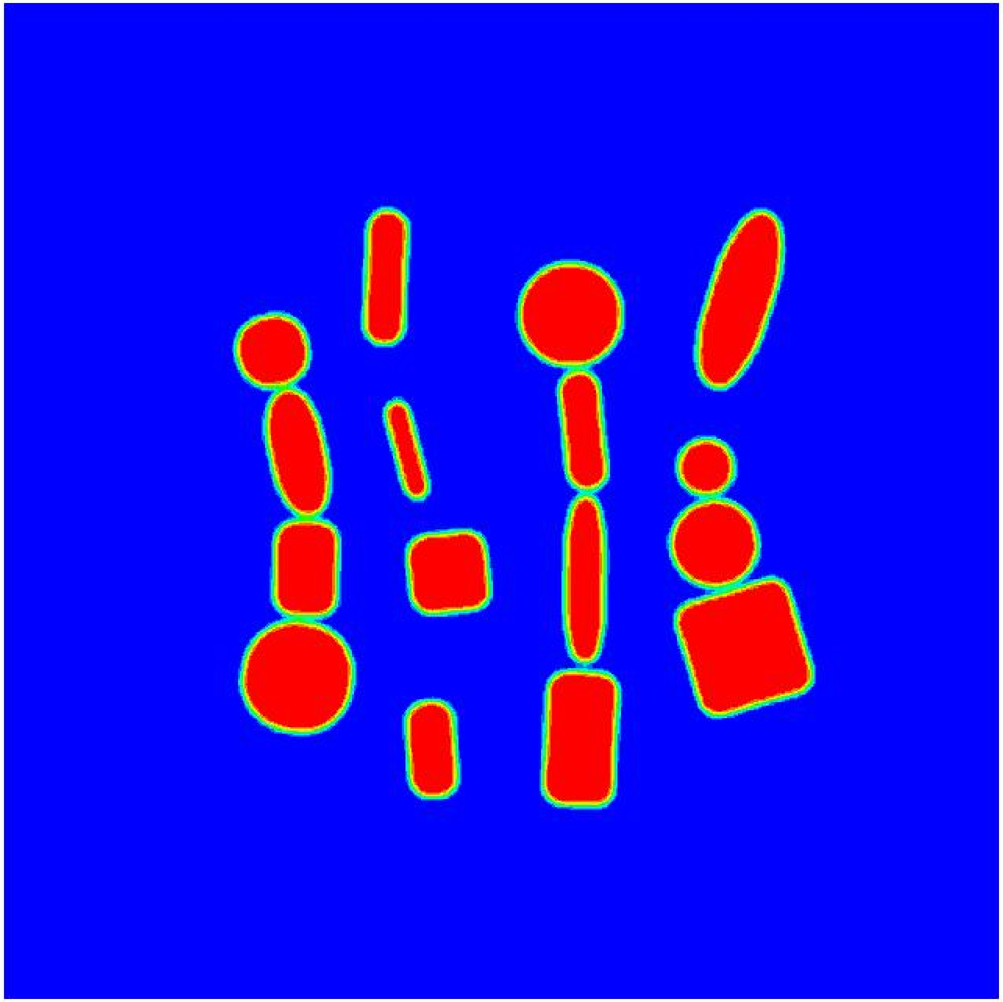

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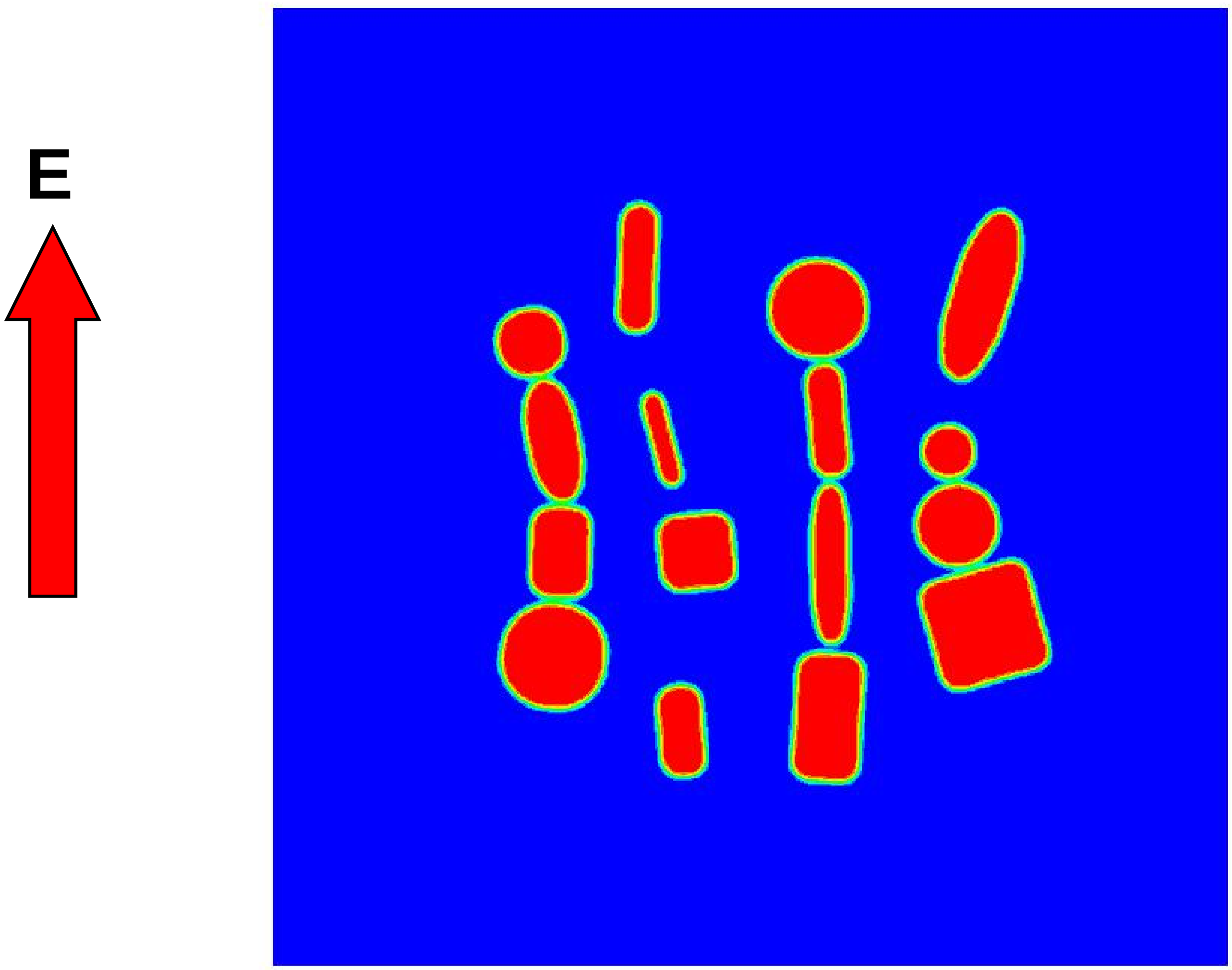

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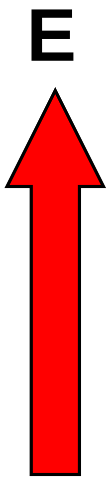

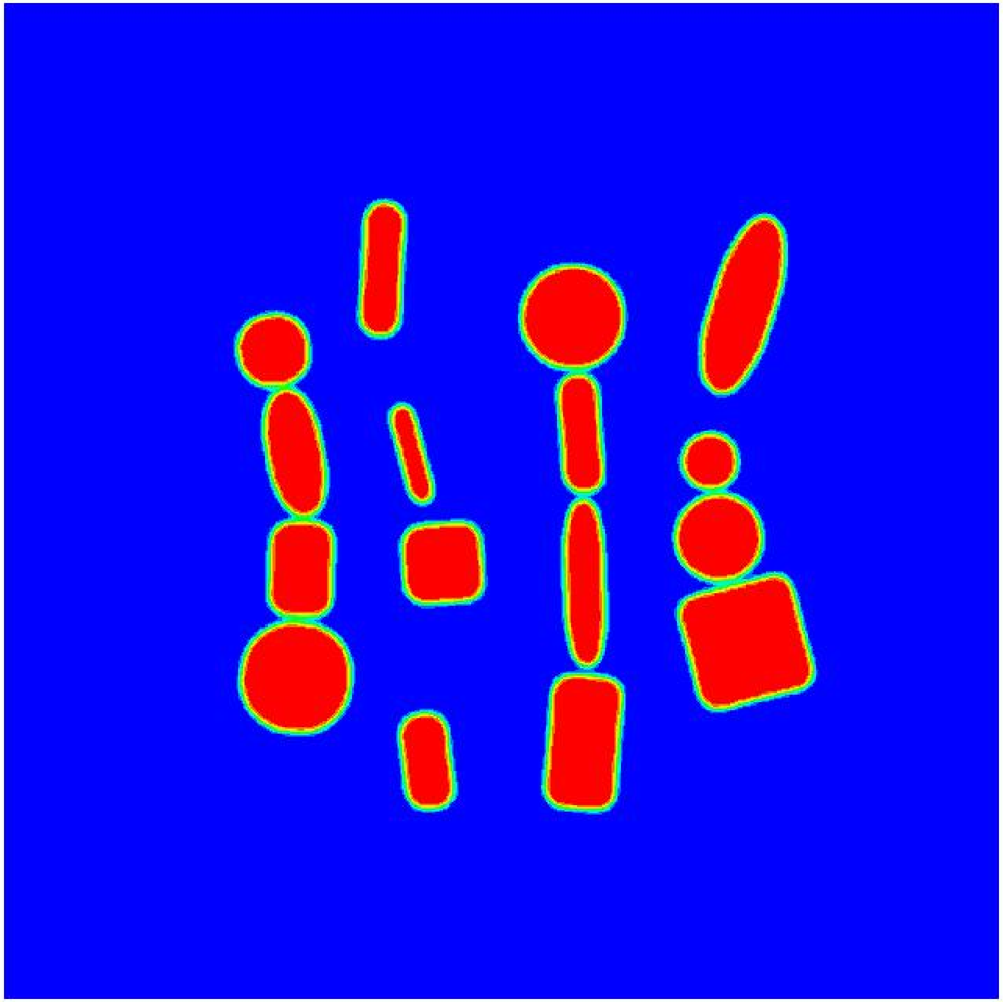

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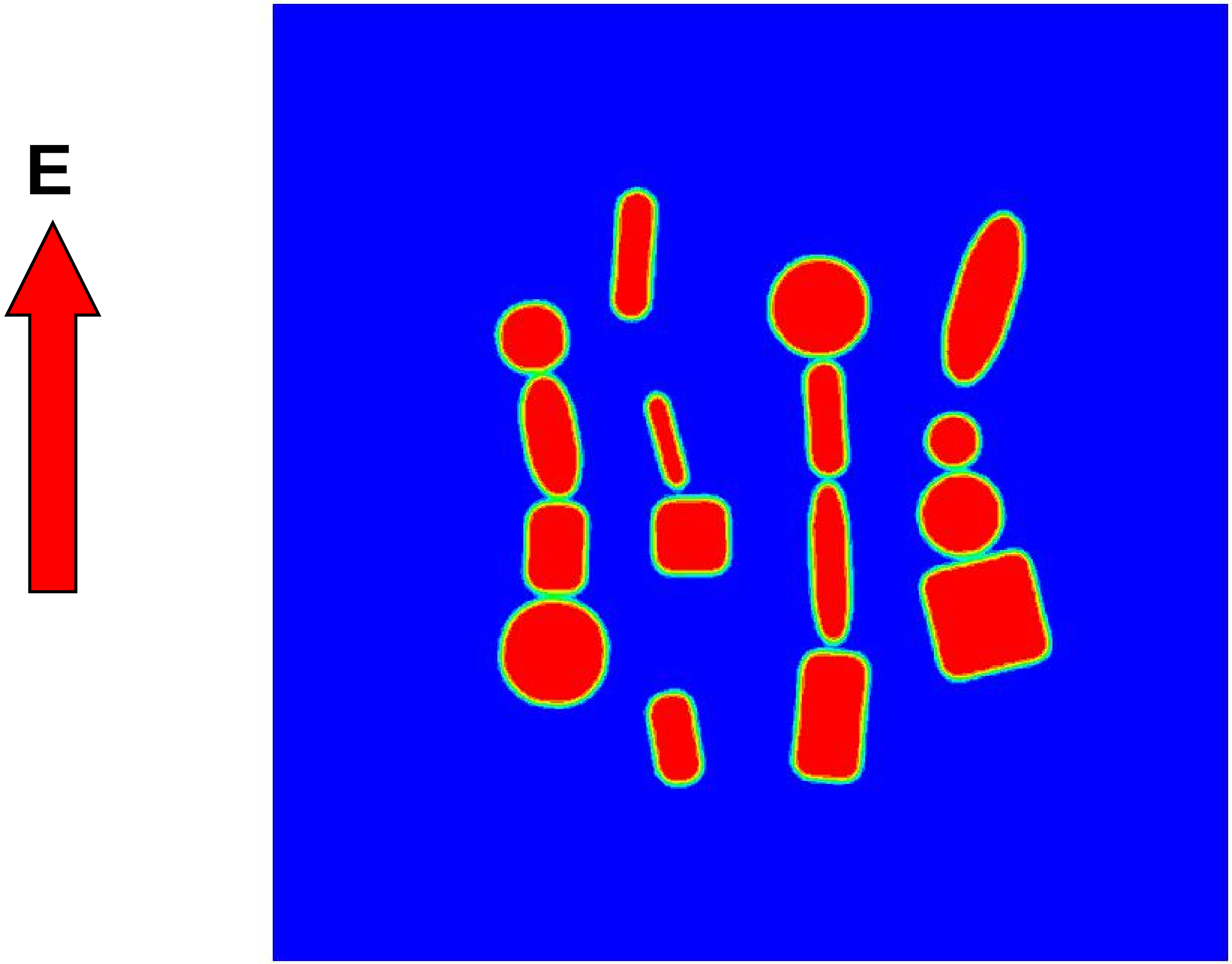

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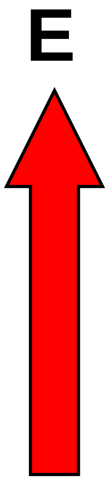

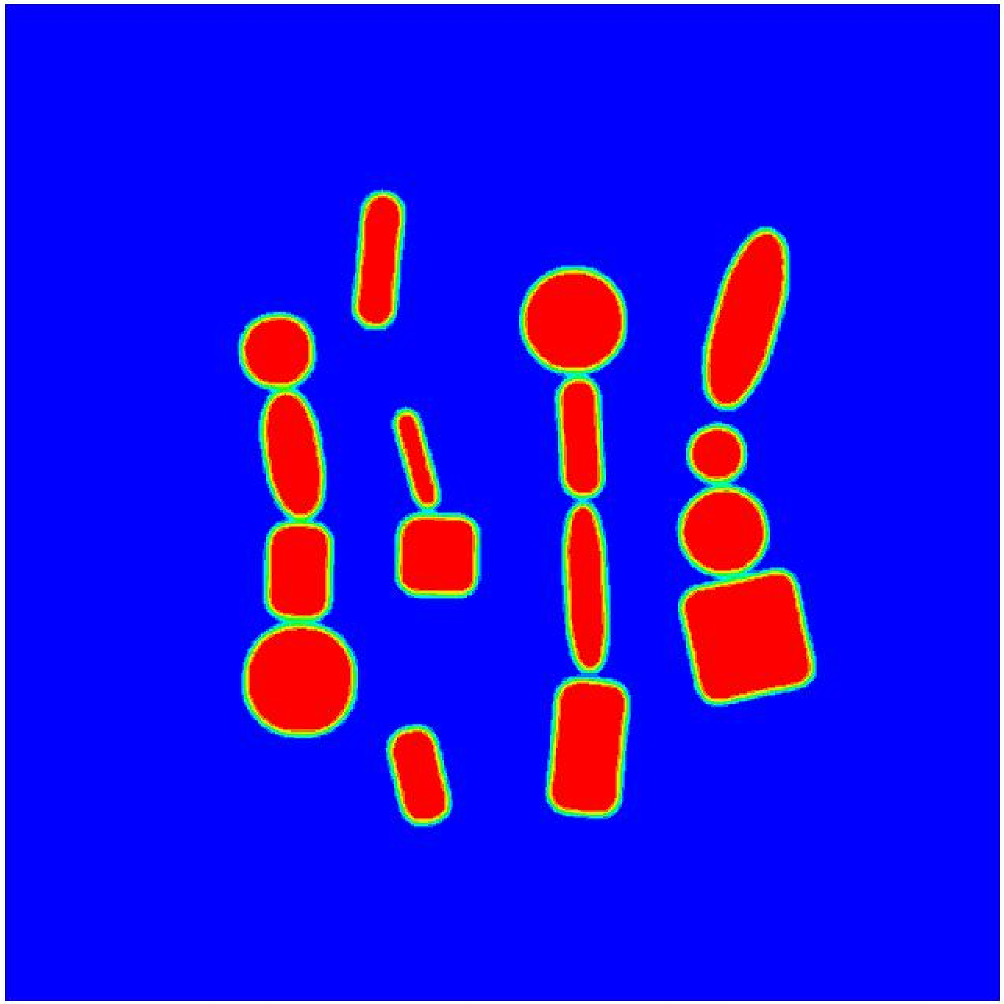

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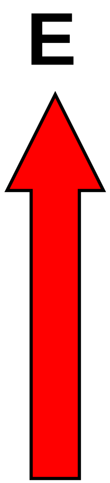

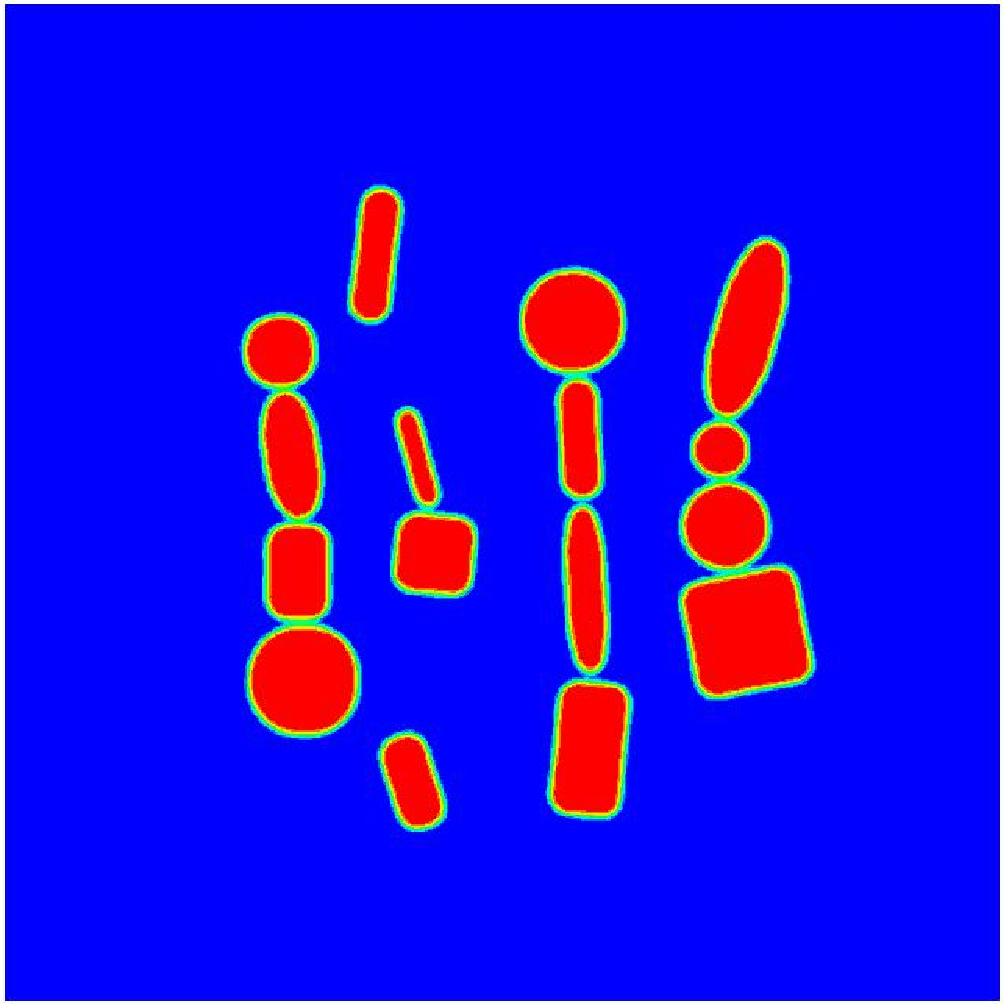

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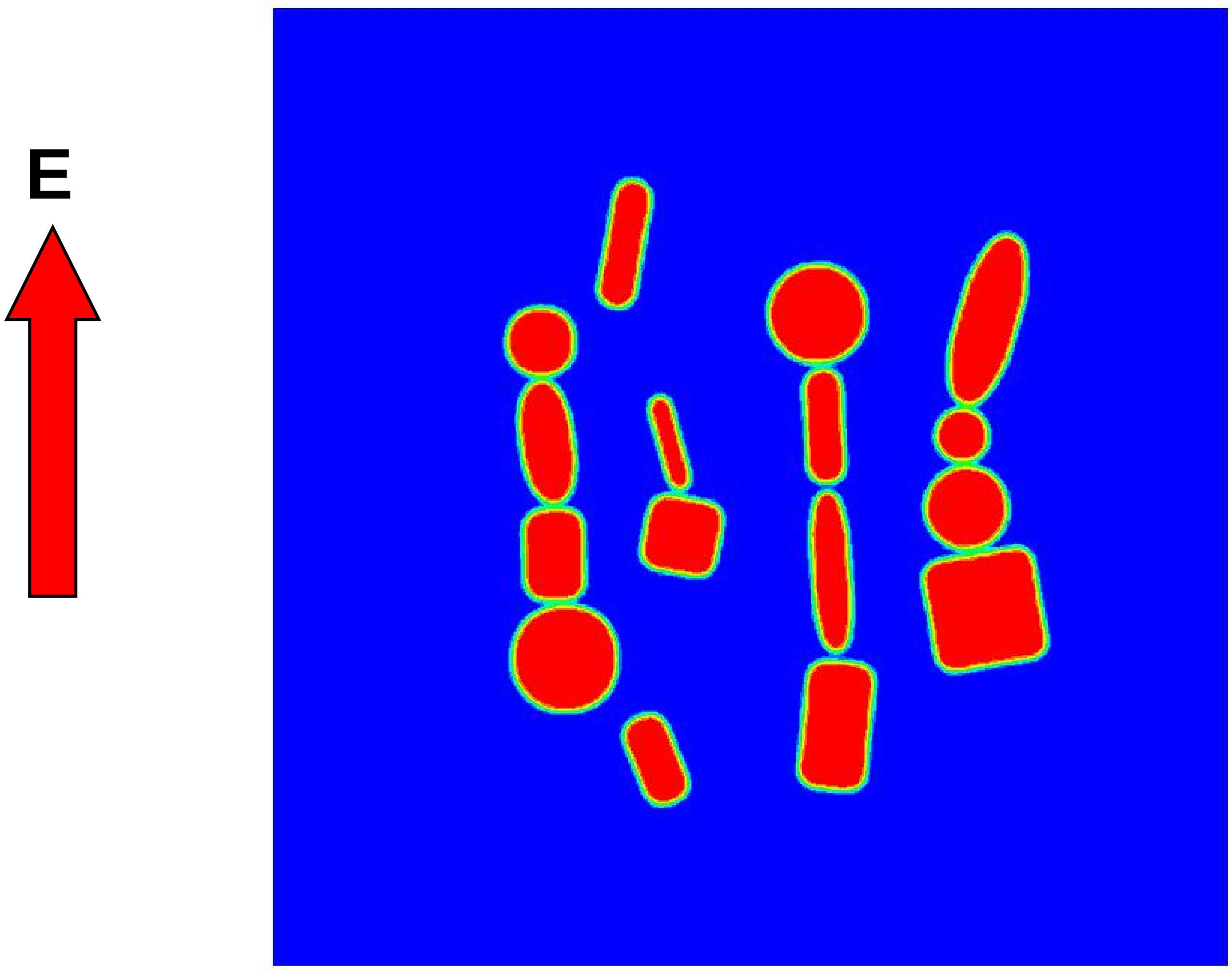

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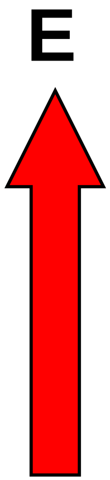

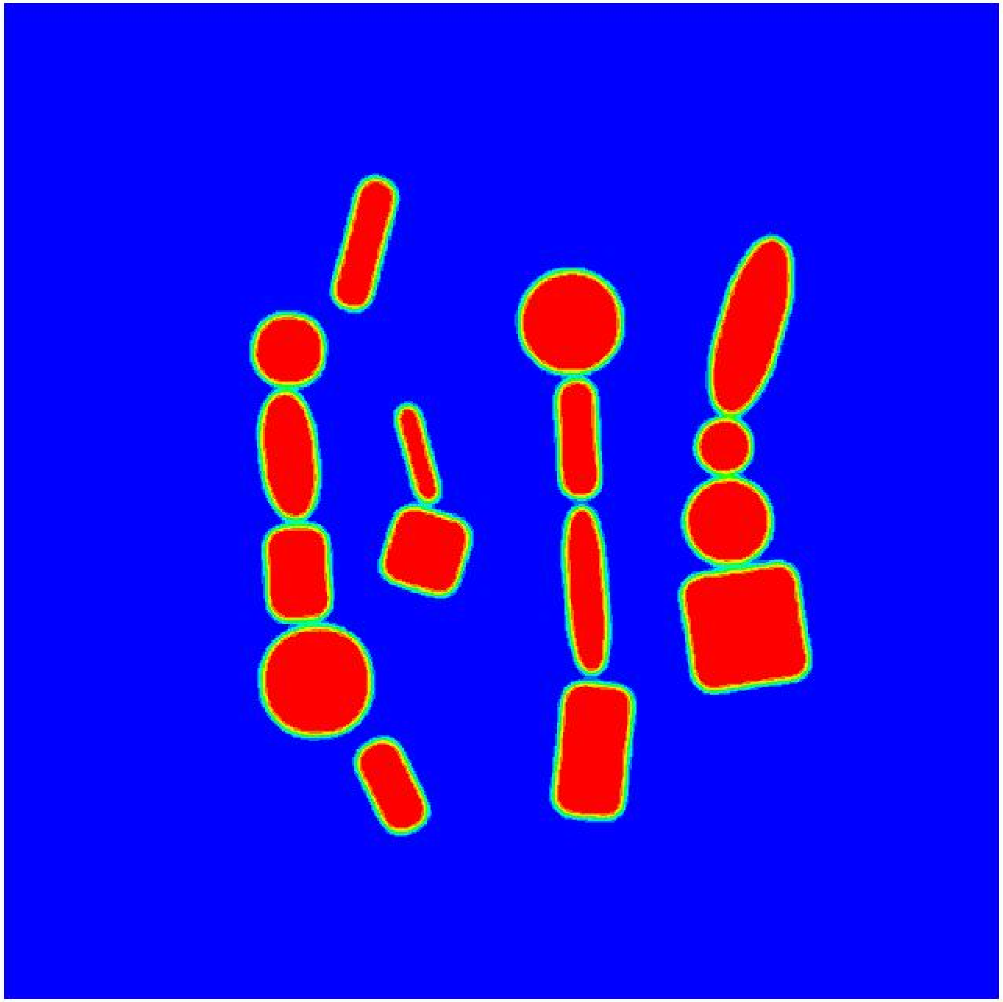

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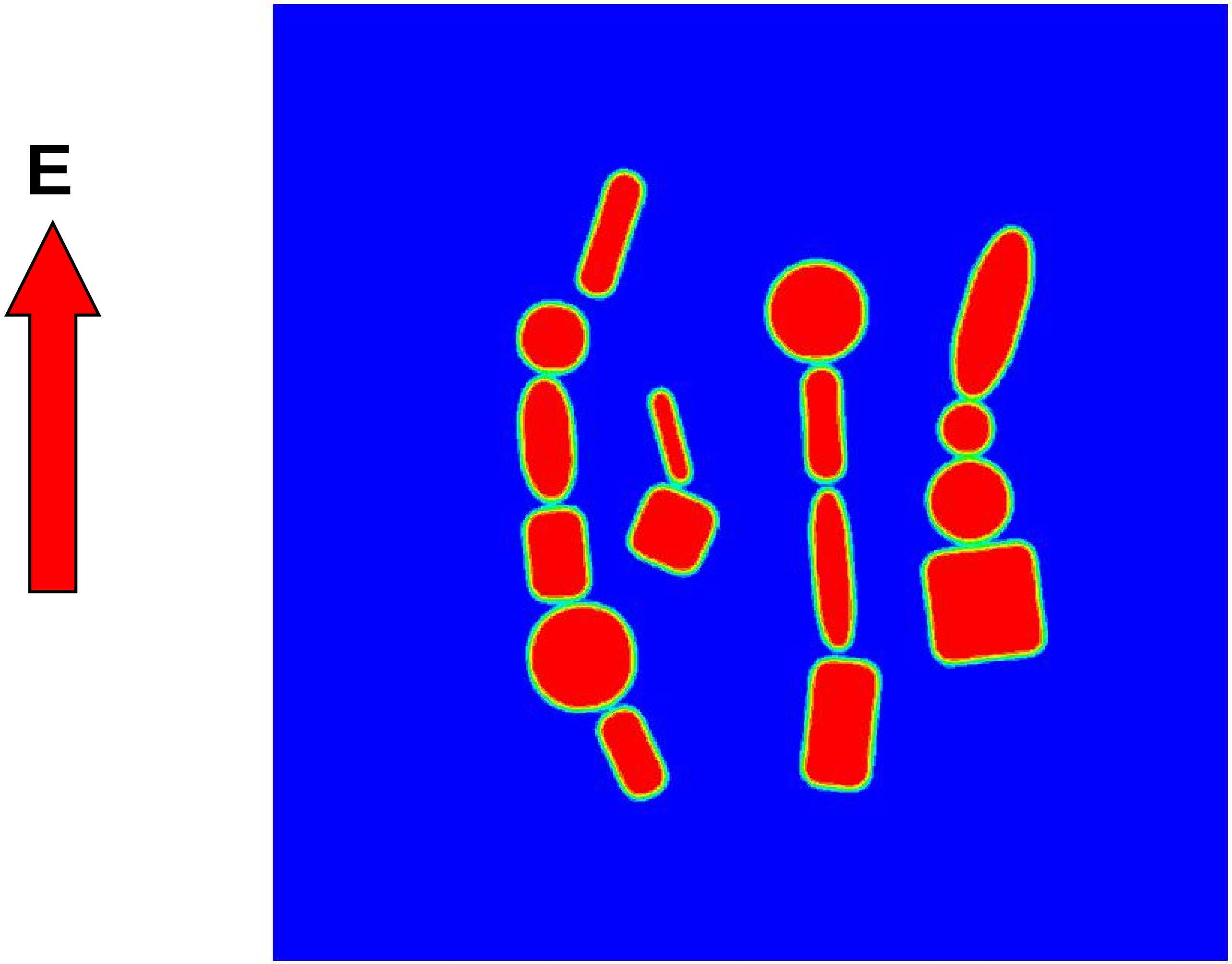

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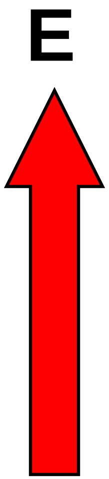

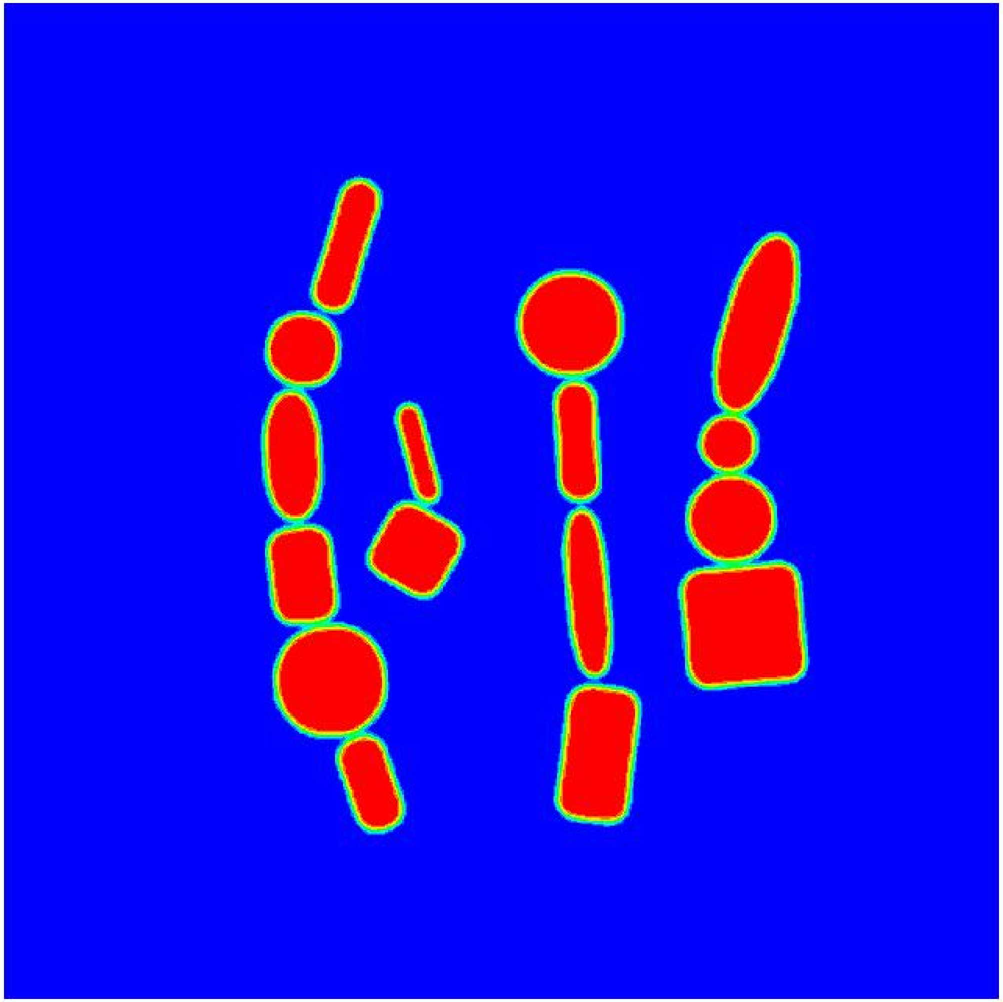

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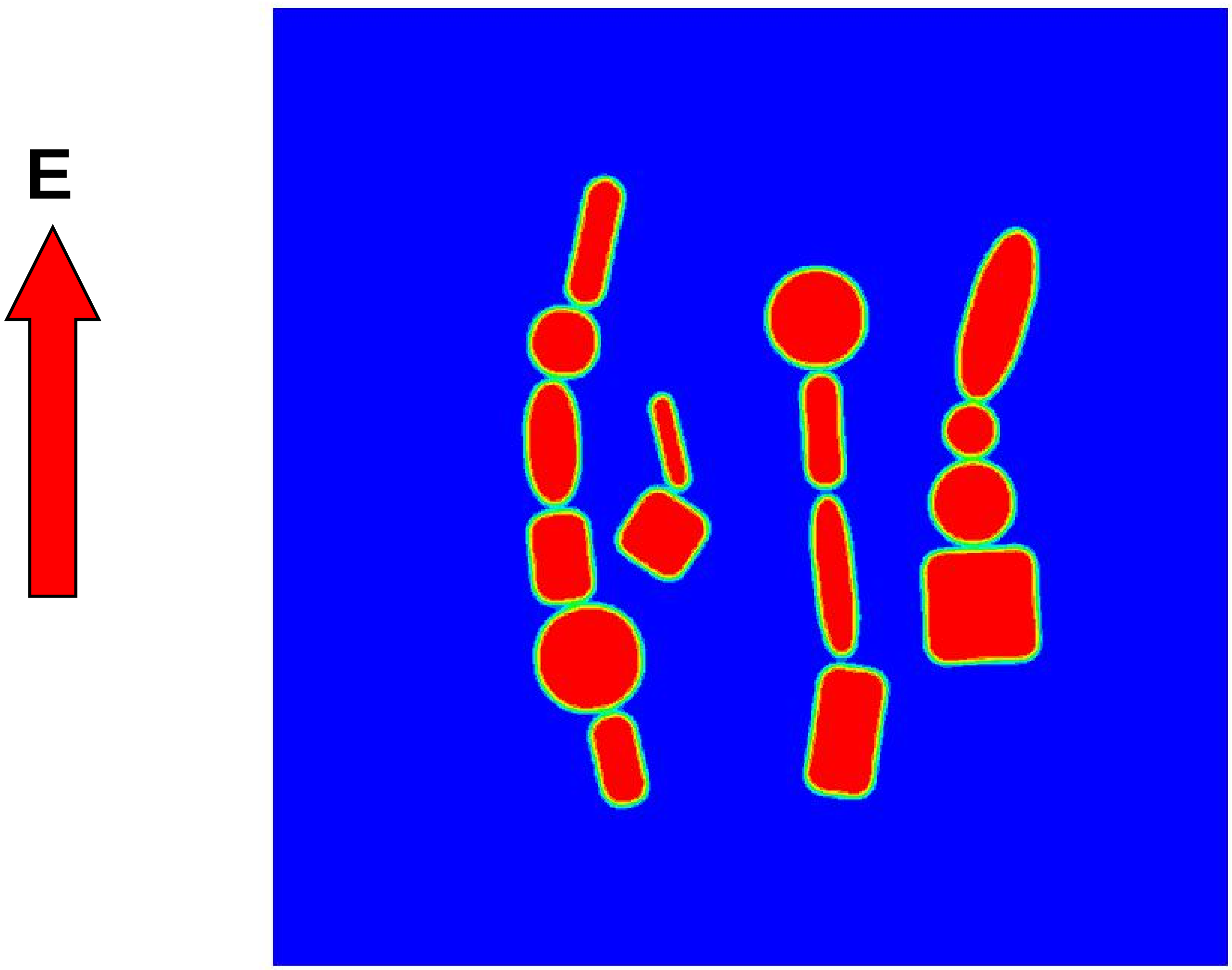

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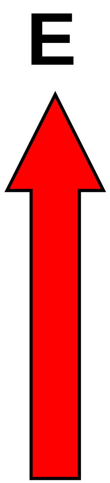

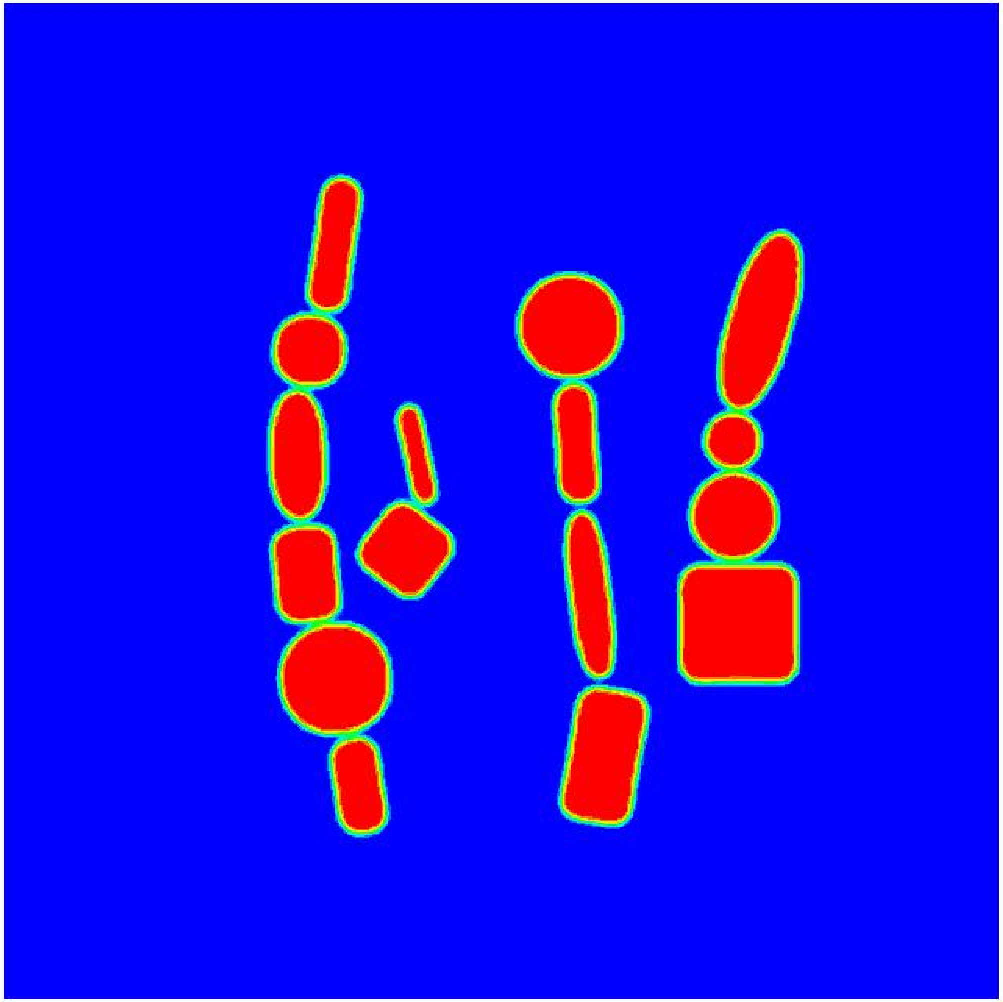

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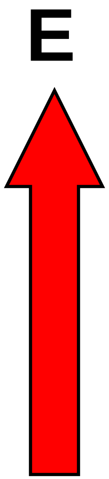

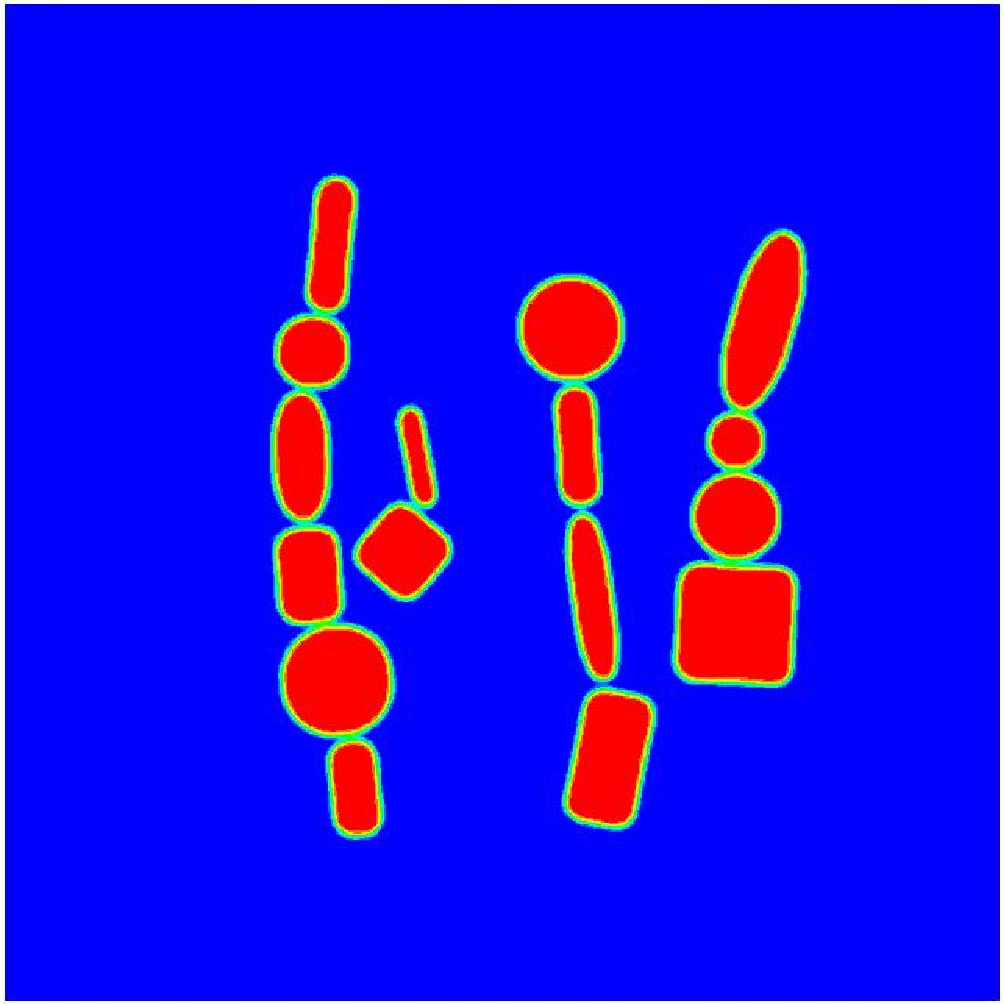

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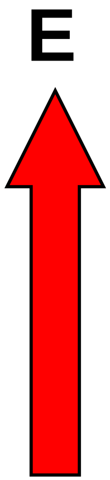

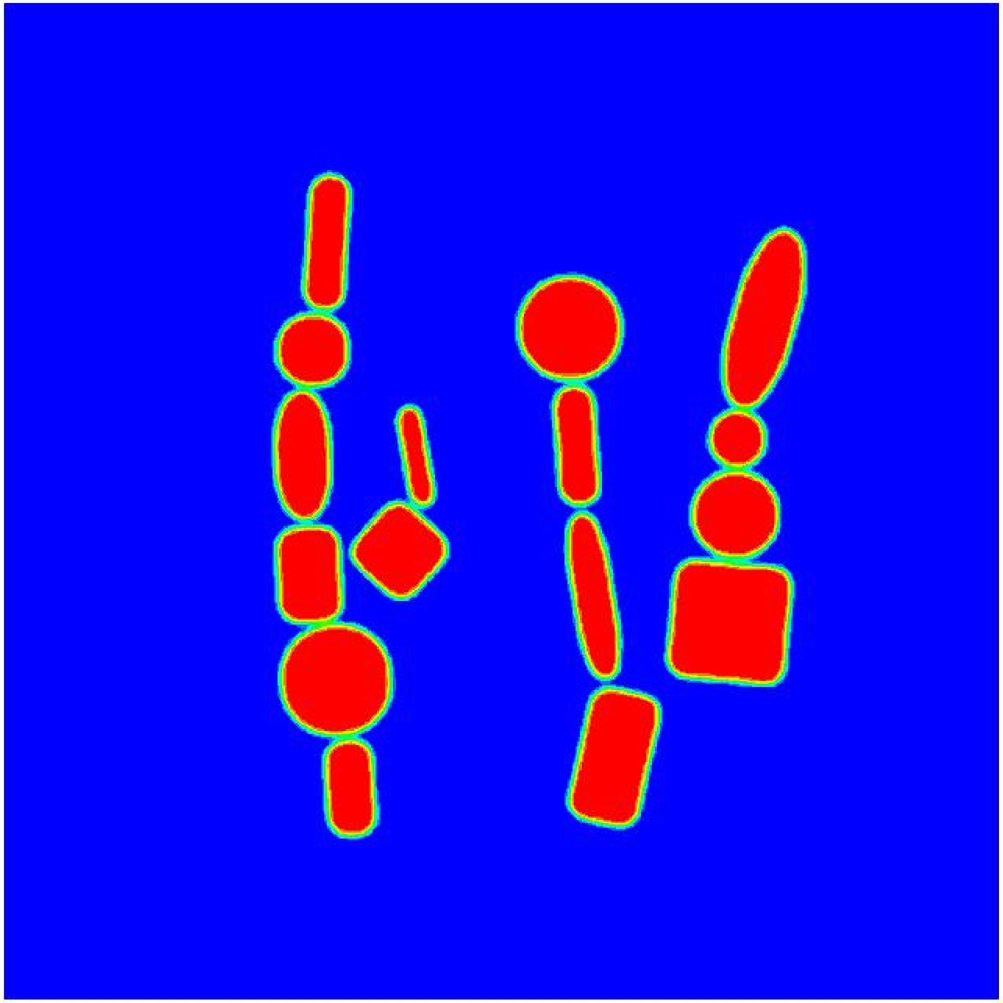

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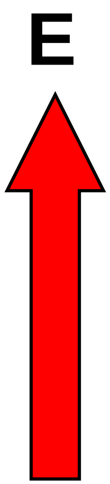
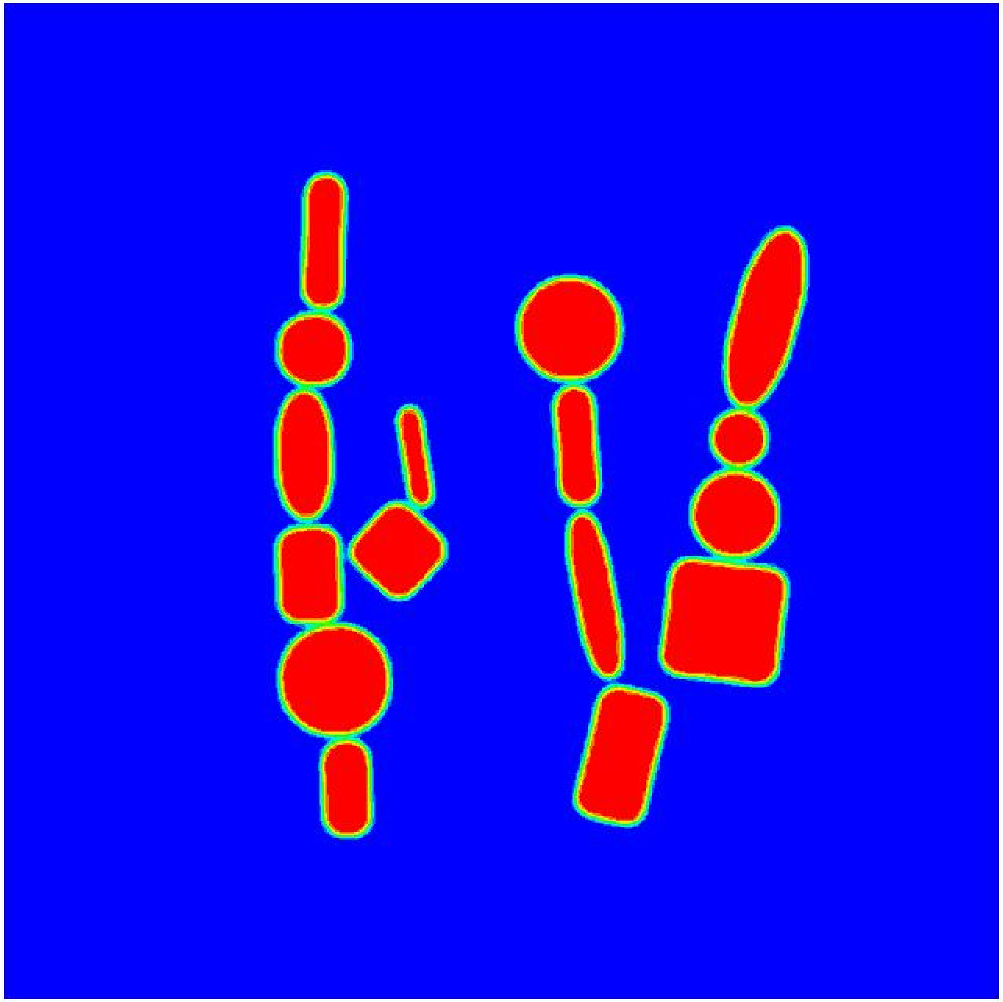
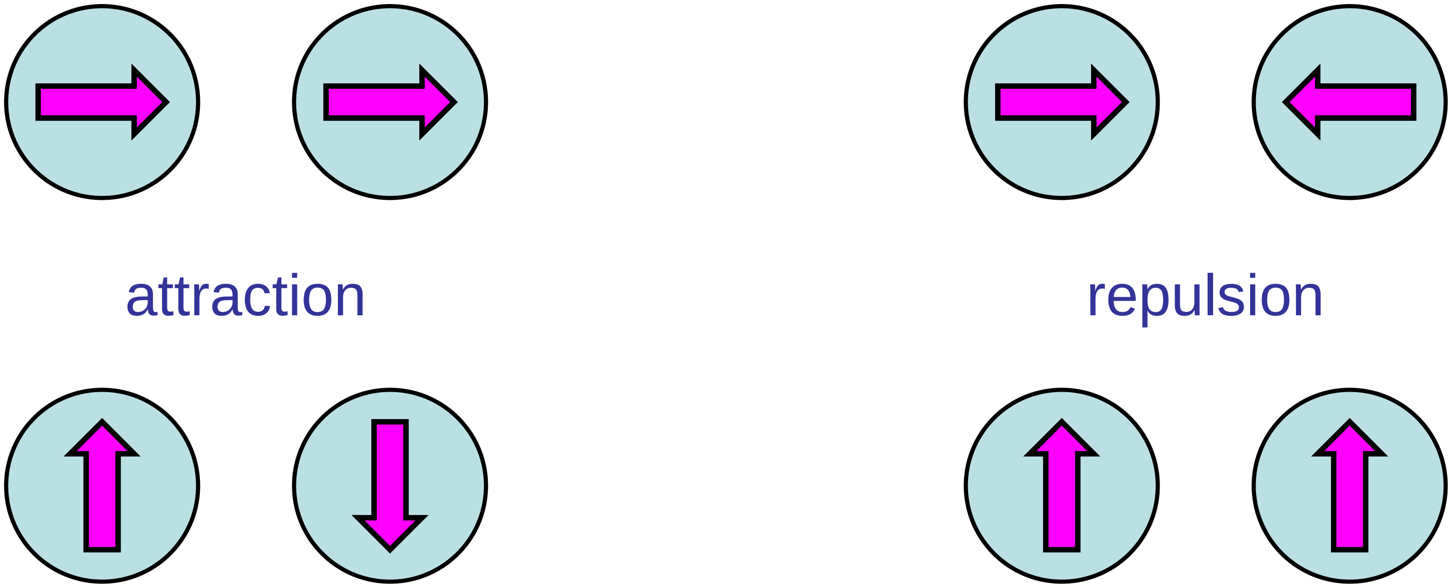




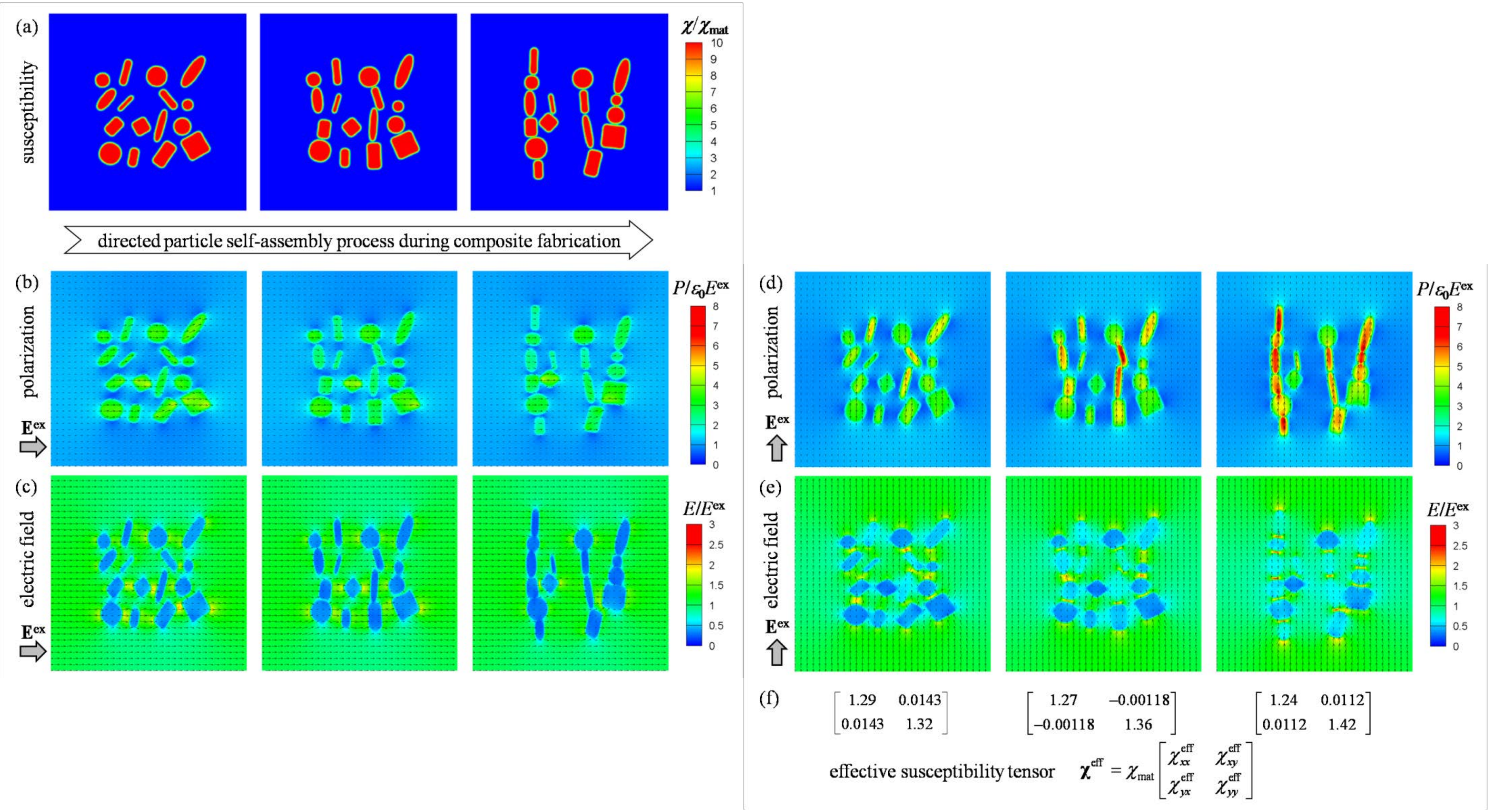
Processing-Microstructure Relationship

- Mechanisms of Filler Particle Self-Assembly
  - Strongly anisotropic force that can be tuned by external field
  - Rigid-body motion (translation and rotation) of colloidal particles in liquids (water, organic solvent, polymer melt, etc.)



Simulation

- Phase field model of dielectric/magnetic composites



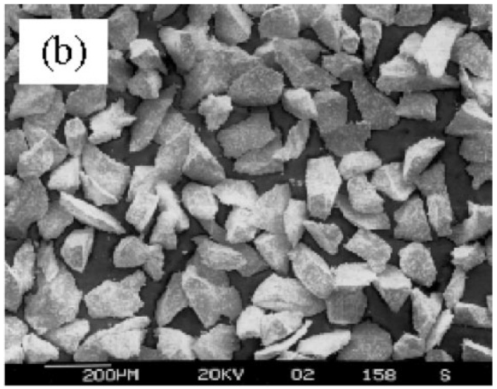
Particle-Filled Polymer-Matrix Composites

- Alignment of irregular-shaped functional filler particles

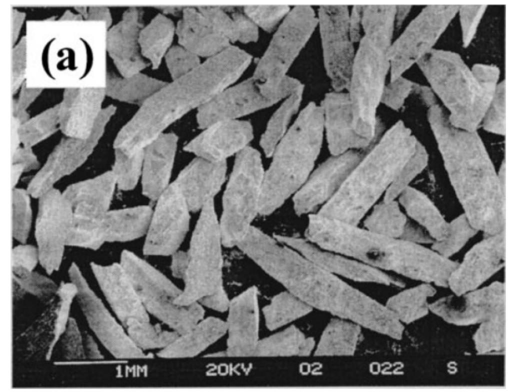
Dielectric: PZT fillers　　Electro-Optic: $PbTiO_3$ nanoparticles

Magnetostrictive: Terfenol-D particles

equiaxed irregular



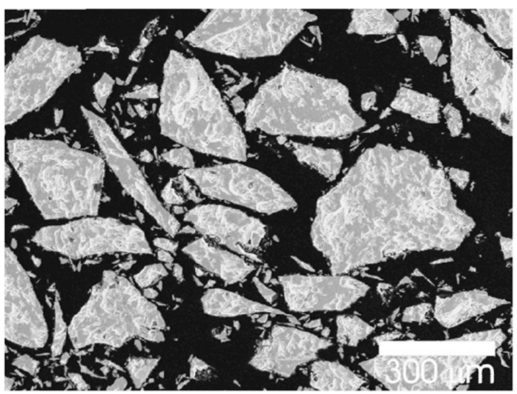
needle-shaped irregular



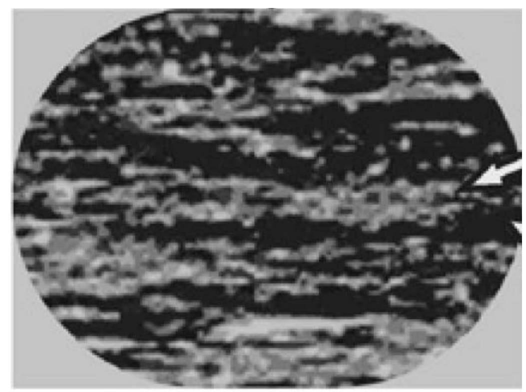
Duenas et al, *J. Appl. Phys.*, **90**, 2433, 2001.

Or et al, *J. Appl. Phys.*, **93**, 8510, 2003.

random



aligned



Shanmugham et al, *J. Mater. Res.*, **19**, 795, 2004.

Or et al, *J. Magn. Magn. Mater.*, **262**, L181, 2003.

Model Formulation

- Particles in multi-phase liquid: capillary forces

$$F = \int \left[f\left(\{c_\alpha\},\{\eta_\beta\}\right) + \sum_\alpha \frac{1}{2}\kappa_\alpha \left|\nabla c_\alpha\right|^2 \right] dV$$

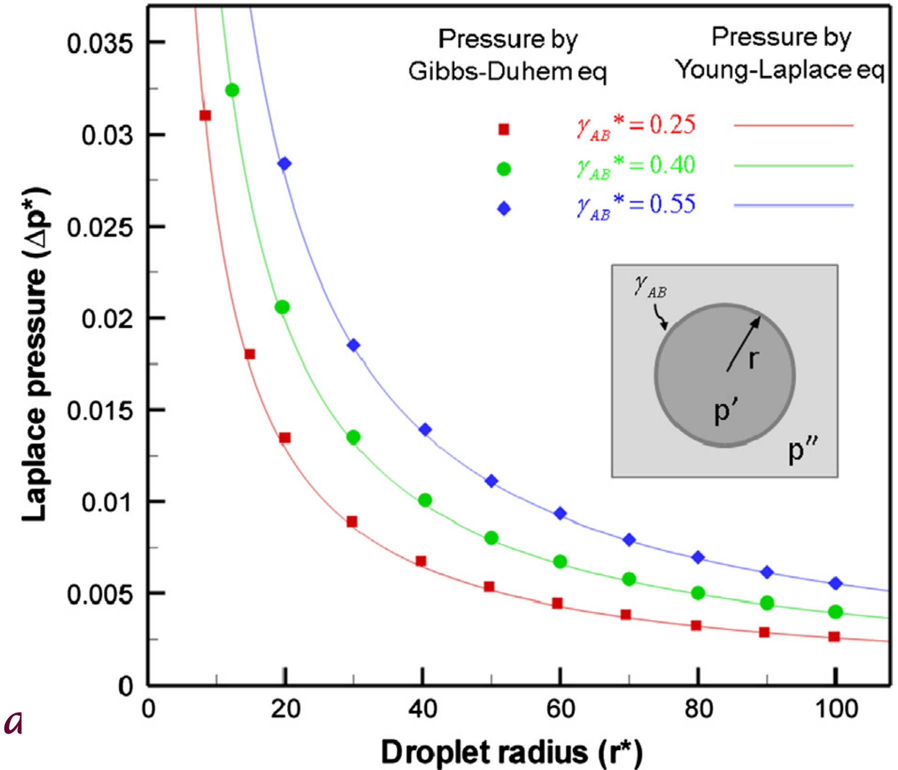
Landau polynomial

$$f\left(\{c_\alpha\},\{\eta_\beta\}\right) = A\left[\sum_{\alpha=1}^{2}\left(3c_\alpha^4 - 4c_\alpha^3\right) + \sum_\beta \left(3\eta_\beta^4 - 4\eta_\beta^3\right) + 6\left(\chi c_1^2 c_2^2 + \sum_\beta \sum_{\alpha=1}^{2} \lambda_\alpha c_\alpha^2 \eta_\beta^2\right)\right]$$

$$\frac{\partial c_\alpha}{\partial t} = \nabla \cdot \left(M_\alpha \nabla \frac{\delta F}{\delta c_\alpha} \right)$$ Cahn-Hilliard

$$dp = c_A d\mu_A + c_B d\mu_B$$
$$p - p^0 = c_1\mu_1 + c_2\mu_2$$
$$\mu_\alpha = \partial f / \partial c_\alpha$$
Gibbs-Duhem

$$\Delta p = \frac{\gamma}{R}$$ Young-Laplace

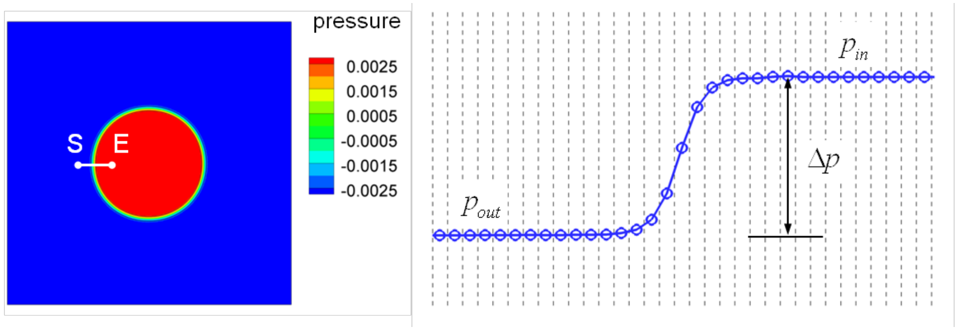


Model Formulation

- Particles in multi-phase liquid: capillary forces

Laplace pressure

$$d\mathbf{F}^{\mathrm{LP}}(\mathbf{r},\beta) = \kappa_{\mathrm{P}} \nabla \eta(\mathbf{r},\beta)\, p(\mathbf{r})\, dV$$

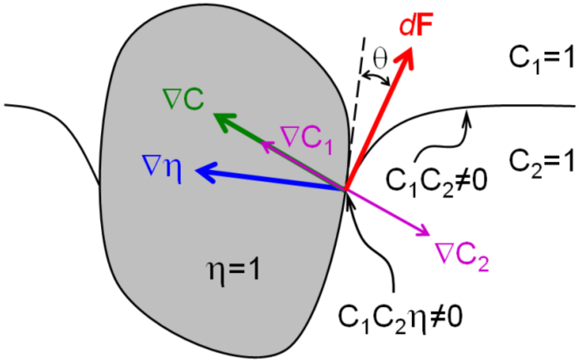


interfacial tension

$$d\mathbf{F}^{\mathrm{IT}}(\beta) = \kappa_{\mathrm{T}}[\nabla c \times (\nabla c \times \nabla \eta_\beta)]dV$$

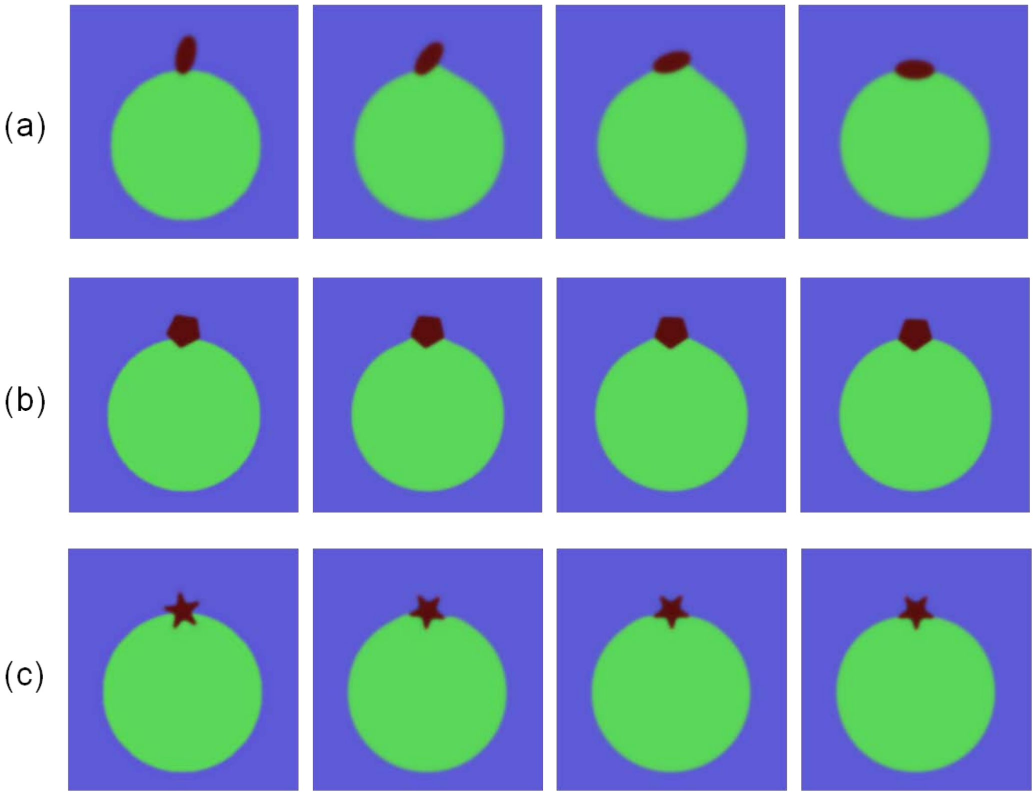
$$= \kappa_{\mathrm{T}}[(\nabla c \cdot \nabla \eta_\beta)\nabla c - |\nabla c|^2 \nabla \eta_\beta]dV$$

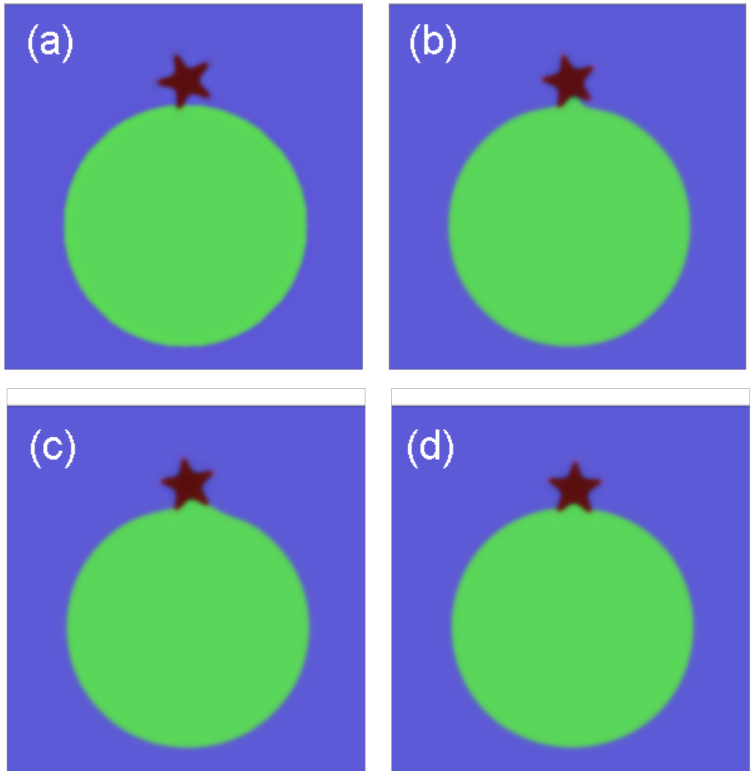
$$\nabla c = c_1 c_2 (\nabla c_1 - \nabla c_2)$$



Simulation

- Irregular-shaped particle at curved fluid interface

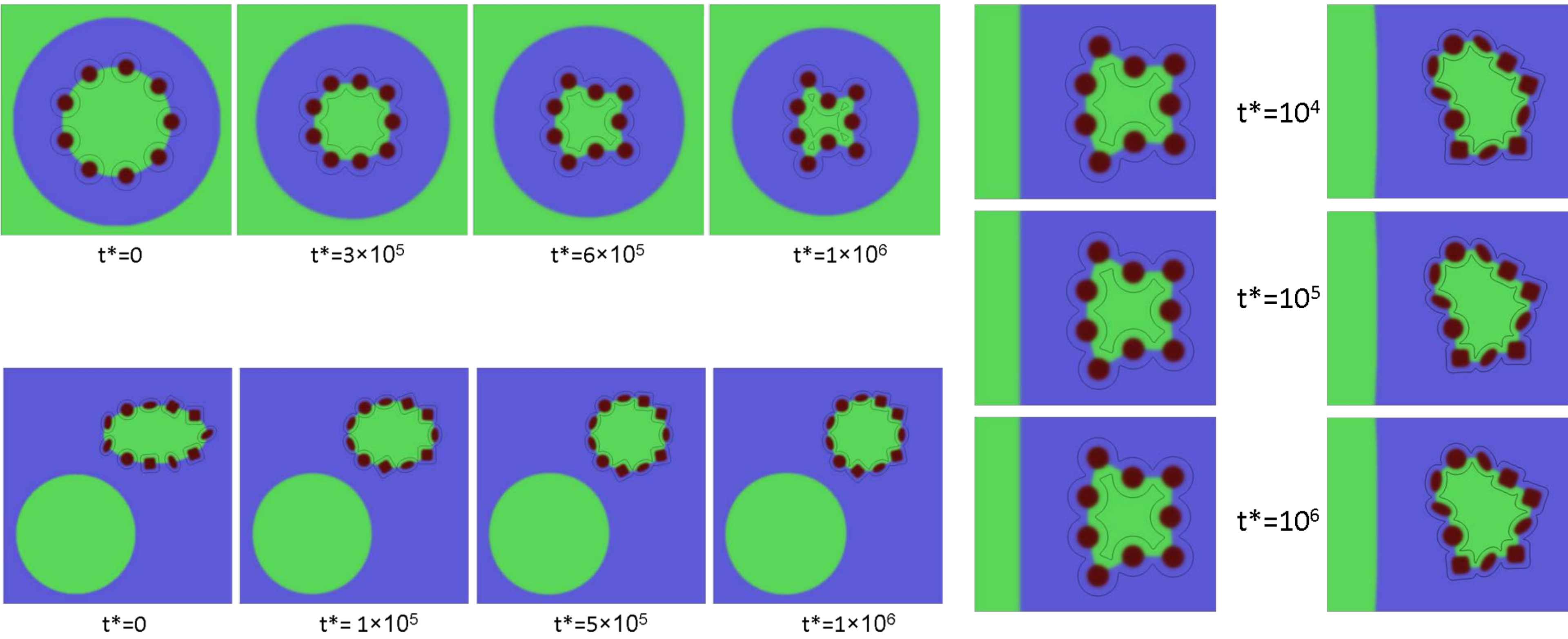




Simulation

- Particle self-assembly directed by fluid interface: encapsulation

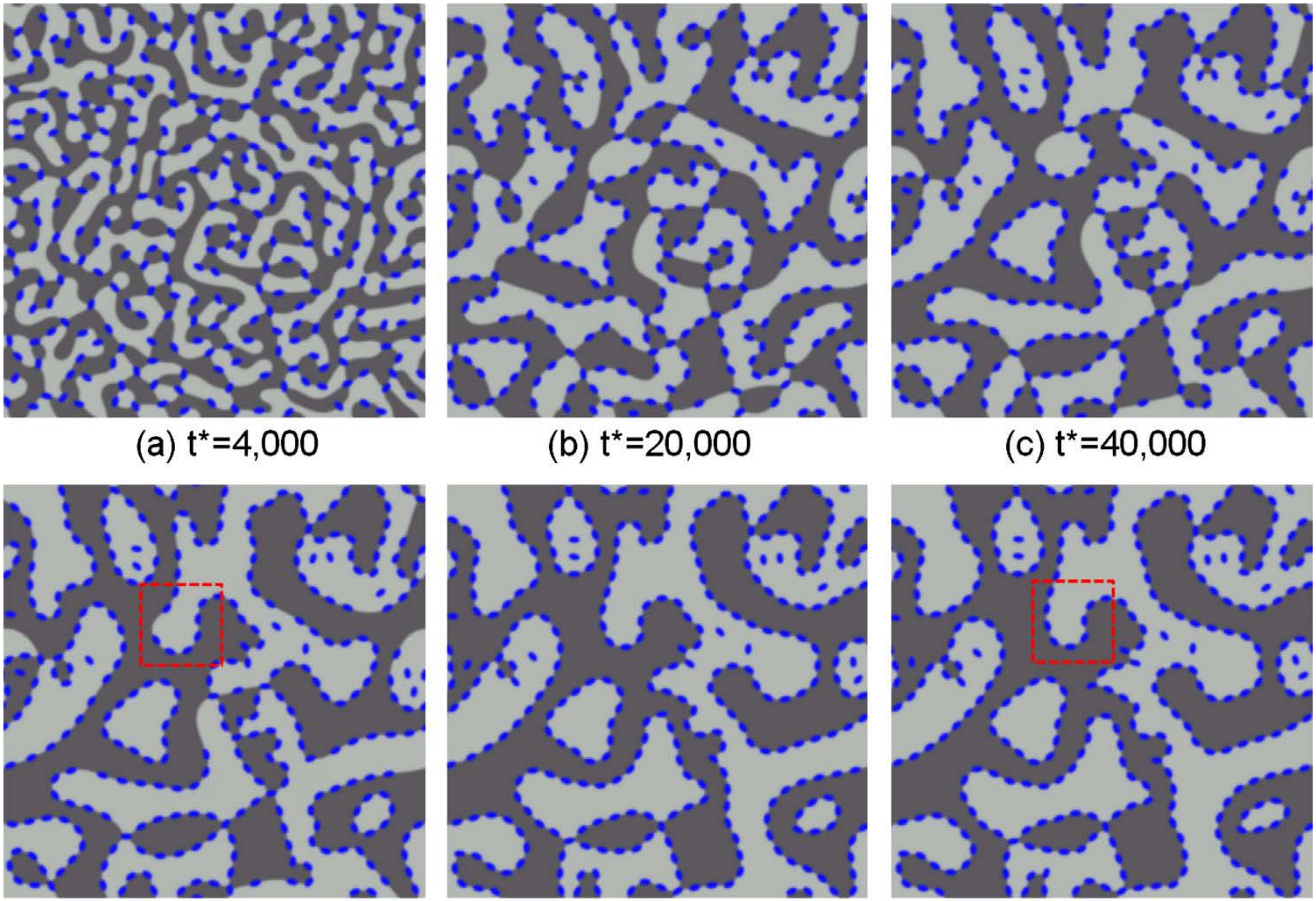
negative pressure | zero pressure



positive pressure

Simulation

- Bijel: bicontinuous interfacially jammed emulsion gels

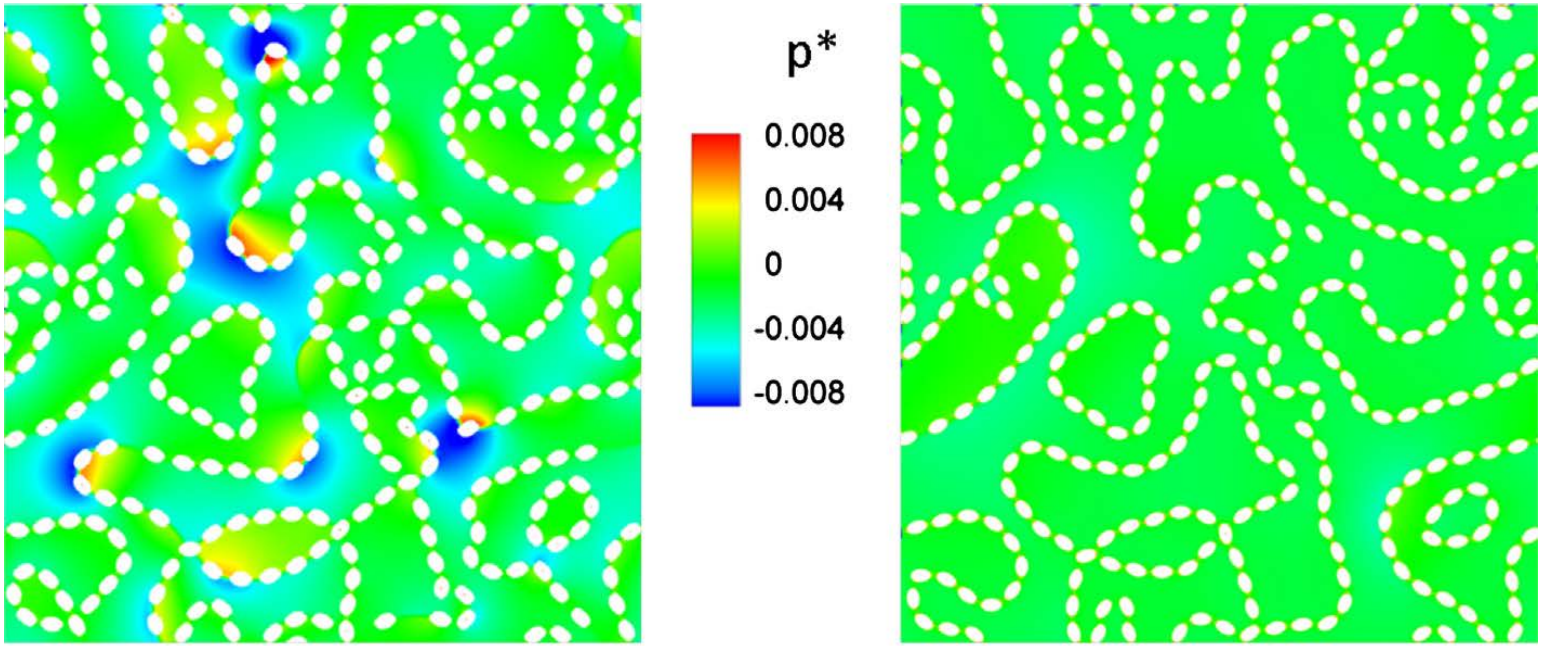


(a) t*=4,000 (b) t*=20,000 (c) t*=40,000

(d) t*=60,000 (e) t*=200,000 (f) t*=400,000

Simulation

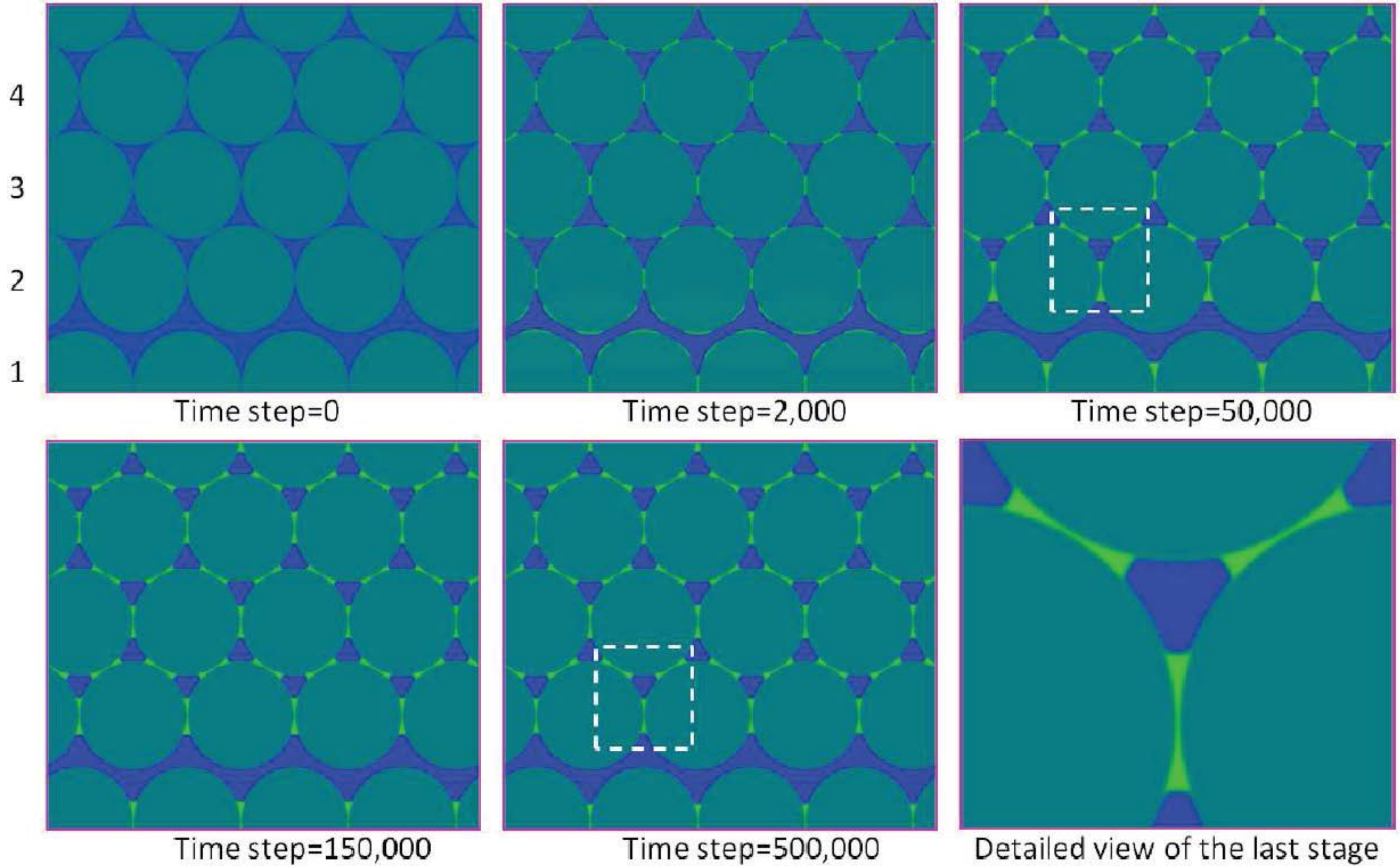
- Bijel: bicontinuous interfacially jammed emulsion gels



(a) t*=60,000 (b) t*=200,000

Simulation

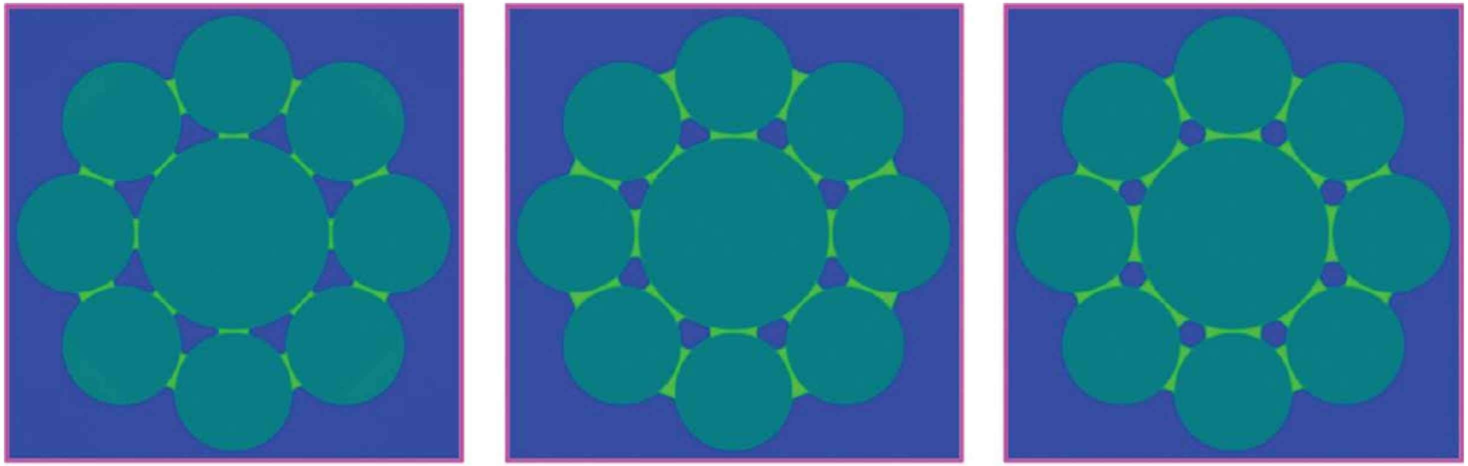
- Capillary bridges for in-situ firming of colloidal crystals



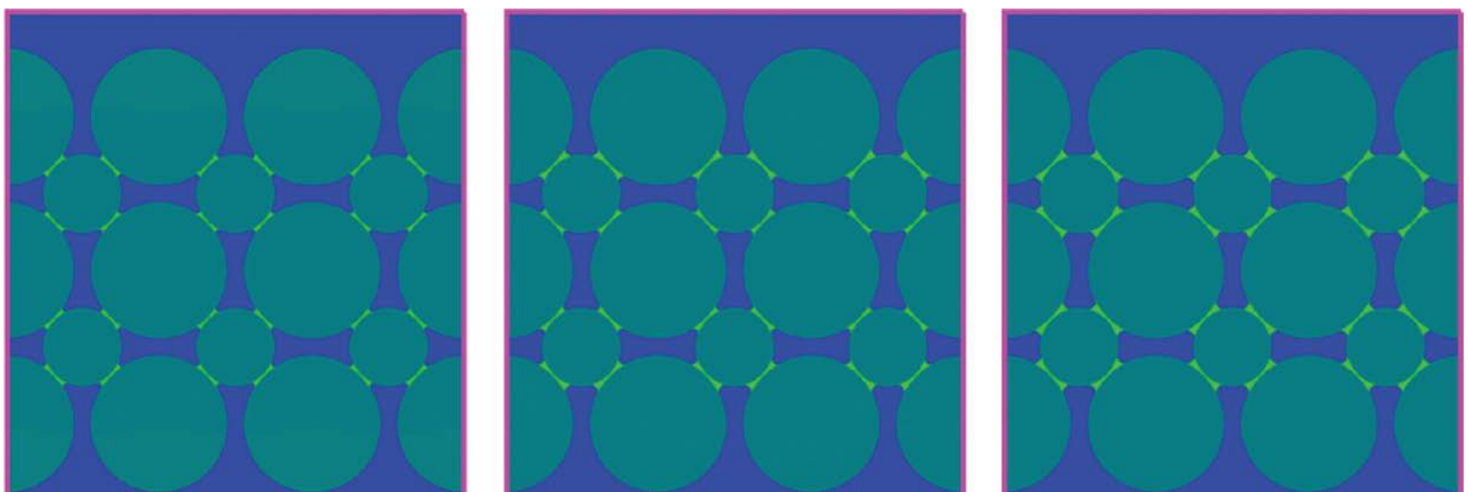
100 nm, 10,000 Pa

Simulation

- Capillary bridges for in-situ firming of colloidal crystals



Time step=10,000 Time step=50,000 Time step=800,000



Time step=10,000 Time step=50,000 Time step=800,000

Acknowledgement

- NSF DMR-0968792; TeraGrid supercomputers.

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Diffuse Interface Field Approach (DIFA) to Modeling and Simulation of Particle-based Materials Processes

Yu U. Wang

Materials Science and Engineering Department
Michigan Technological University