Numerical evidence for thermally induced monopoles

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Motivation

Bresme and co-workers demonstrated numerically that certain polar liquids, such as water, align in response to an imposed temperature gradient, resulting in a thermally induced electric field

$$\boldsymbol{E}_{\mathrm{TP}}(\boldsymbol{r}) = S_{\mathrm{TP}} \nabla T(\boldsymbol{r}),$$

where $T(\boldsymbol{r})$ is the temperature and S_{TP} the thermo-polarization coefficient.

Frenkel recently noticed that this effect leads to a non-vanishing field flux through a surface enclosing the heat source/sink that generates the temperature gradient. In other words, the heat source/sink carries an effective charge q_{TP}:

$$\oiint \boldsymbol{E}_{\mathrm{TP}}(\boldsymbol{r}) \cdot \mathrm{d}\boldsymbol{S} \equiv \frac{q_{\mathrm{TP}}}{\epsilon_0}.$$

Consequently, a pair of heated or cooled colloidal particles immersed in such a solvent is expected to attract each other.

We show using molecular simulations that a pair of heated/cooled colloidal particles in a dipolar solvent behaves like oppositely charged electric or magnetic monopoles.

Solvent model

We employed a modified Stockmayer fluid, consisting of particles with a point dipole (colored arrows) and a Lennard–Jones center displaced along the direction of the dipole moment. The displacement is controlled by a parameter α and a non-zero value is necessary for the molecules to undergo thermo-molecular orientation.

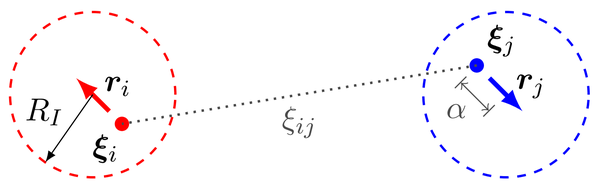

Figure 1: Two interacting solvent particles with a homogeneous mass distribution over a ball of radius R_I centered around the dipole.

Key References

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[5] F. Bresme *et al.*: *Water polarization under thermal gradients*, Phys. Rev. Lett. **101**, 020602 (2008)

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Thermally induced alignment of dipoles

We performed equilibrium and nonequilibrium molecular dynamics simulations of a heated and a cooled colloidal particle immersed into an off-center Stockmayer fluid.

To maintain a steady-state, we added energy to the hot particle and withdrew it from the cold one continuously using the eHEX algorithm. Cylindrical averages for the temperature and average dipole orientations are shown in Fig. 2.

The thermally induced charge is related to the temperature difference between the particle surface at a distance R from the center and the bulk:

$$q_{\mathrm{TP}} = -4\pi\epsilon_0 S_{\mathrm{TP}}(T_R - T_\infty)R.$$

Furthermore, solvent dipoles align with the superimposed electric field lines generated by two virtual point charges $\pm q_{\mathrm{TP}}$.

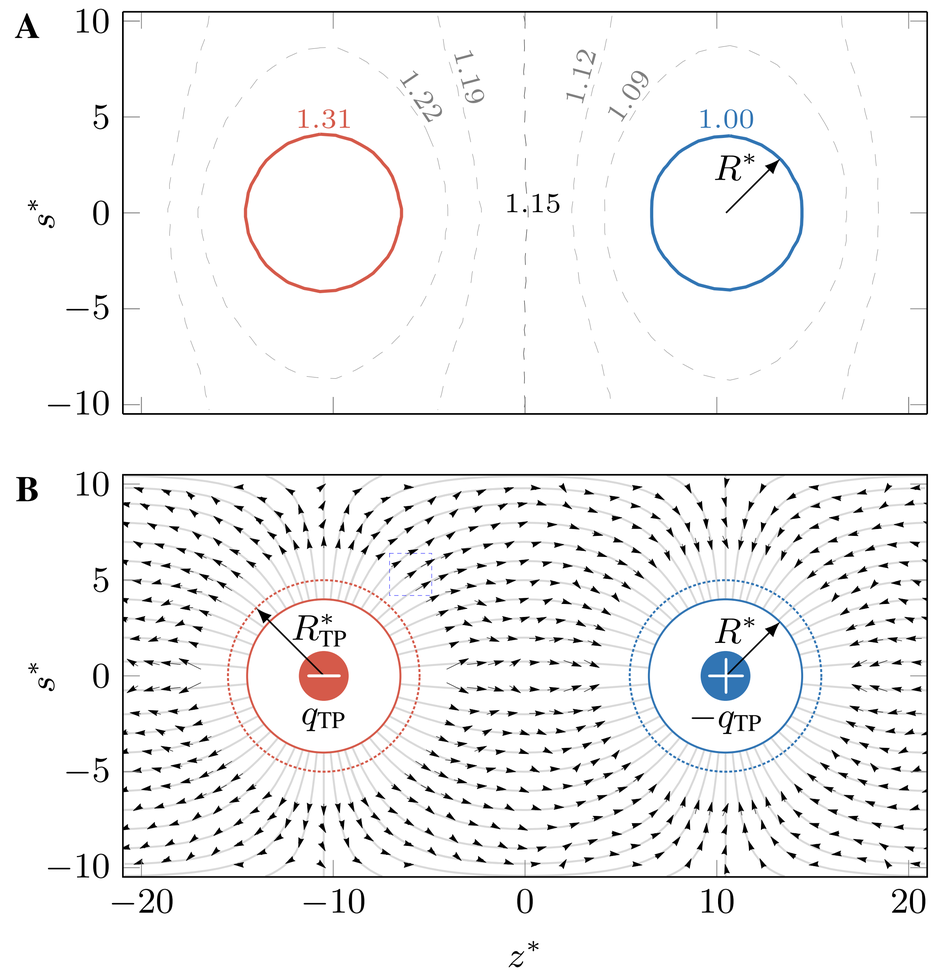

Figure 2: Cylindrically averaged temperature profile (**A**) and dipole orientations (**B**) generated by a pair of heated/cooled colloidal particles with symmetry axis z^* and perpendicular axis s^* in a fully periodic system.

Thermally induced monopoles

The analytical solution for the electric field generated by two charged spherical shells (Fig. 3A) is given by

$$\frac{\langle E_{z,\mathrm{TP}}(z)\rangle}{\tilde{E}} = \begin{cases} -1 & \text{if } |z| > z_{\mathrm{c}} + R_{\mathrm{TP}}, \\ +1 & \text{if } |z| < z_{\mathrm{c}} - R_{\mathrm{TP}}, \\ (z - z_{\mathrm{h}})/R_{\mathrm{TP}} & \text{if } |z - z_{\mathrm{h}}| \leq R_{\mathrm{TP}}, \\ (z_{\mathrm{c}} - z)/R_{\mathrm{TP}} & \text{otherwise,} \end{cases}$$

where $z_{\mathrm{h/c}} = \mp L/4$ denote the locations of the hot and cold colloidal particle, respectively, L is the box length in the z-direction, $\tilde{E} = q_{\mathrm{TP}}/(2A\epsilon_0)$ is the constant value of the averaged field between the colloidal particles, and $A = L^2/4$ is the cross-sectional area.

We can link theory and simulation by computing the field from the steady-state dipole density according to

$$\langle E_{z,\mathrm{TP}}(z)\rangle = -\frac{\langle \rho_\mu(z) - \bar{\rho}_\mu\rangle}{\epsilon_0},$$

where $\bar{\rho}_\mu = 1/L \int \mathrm{d}z \langle \rho_\mu(z)\rangle$ is the box average of $\langle \rho_\mu(z)\rangle$.

The excellent quality of agreement in Fig. 3C shows that the two colloidal particles behave as if they carry the Coulomb charges $\pm q^*_{\mathrm{TP}} \approx \mp 0.134$.

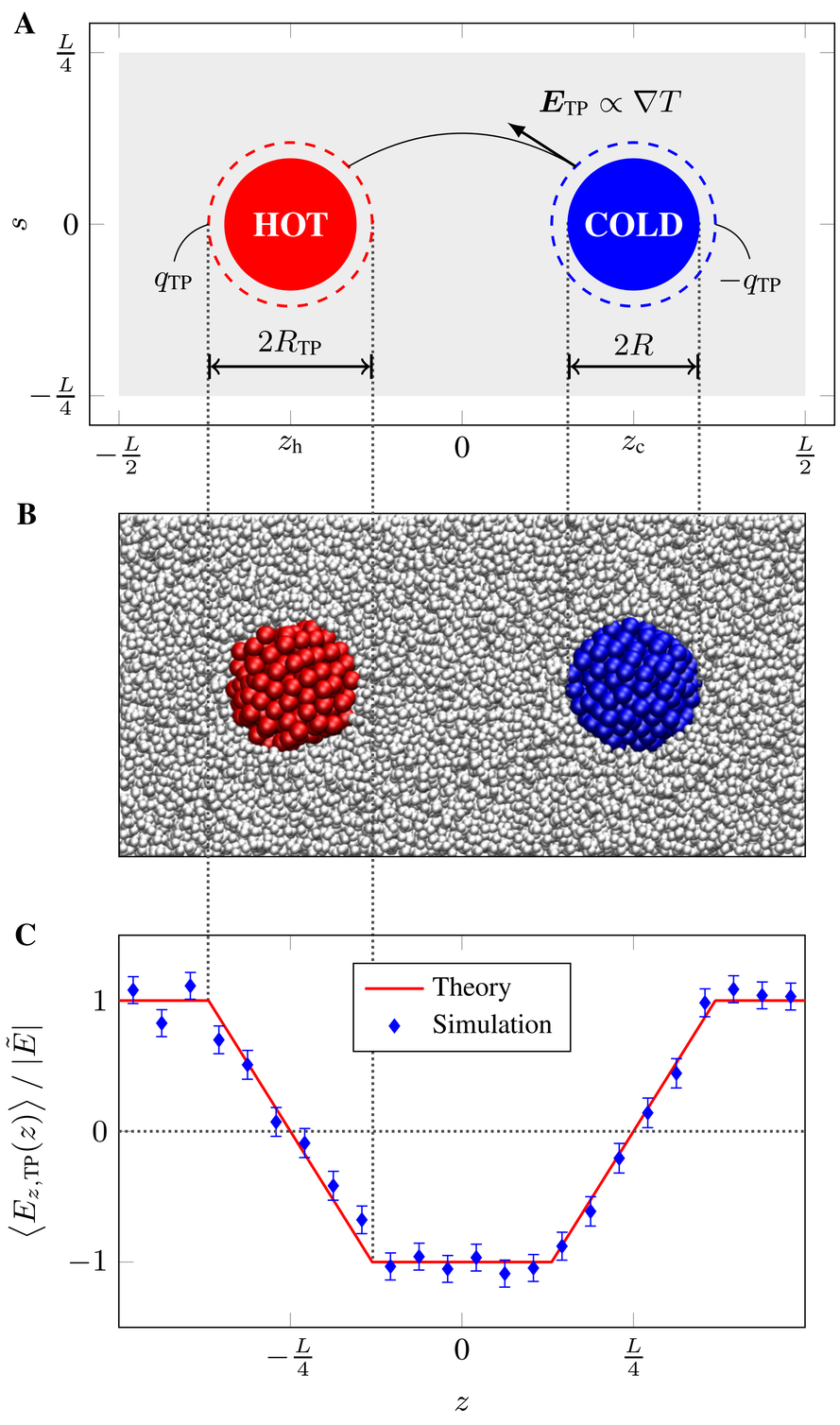

Figure 3: Illustration of the simulation setup (**A** and **B**) and induced electric field averaged over slabs perpendicular to the symmetry axis (**C**).