Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Coarse-grained model of ionic liquid crystals: the effect of stoichiometry on the stability of the ionic nematic phase

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Electronic Supporting Information

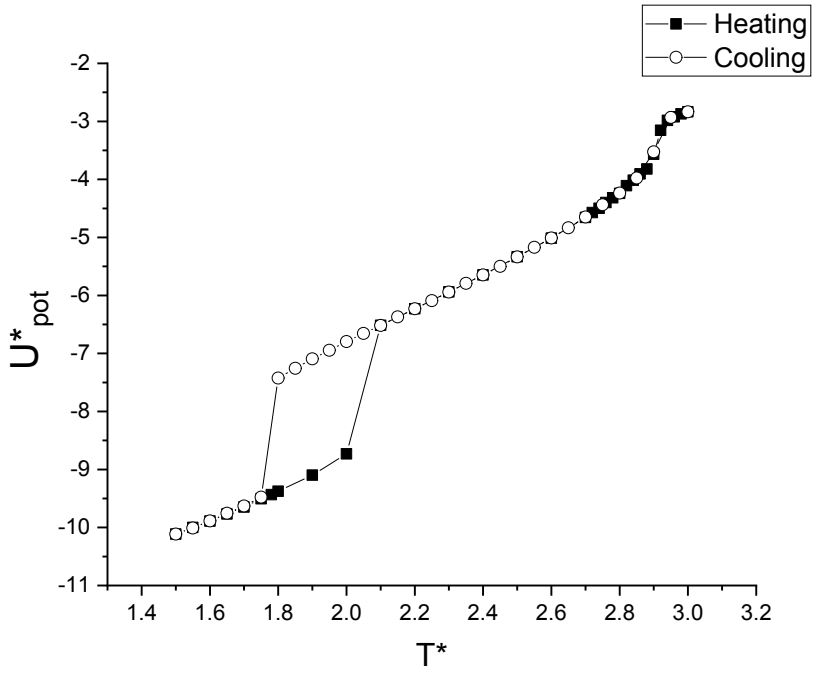

Figure S1. Reduced potential energy, U^*_{pot}, as a function of the reduced temperature for the GB:LJ 2:1 system at reduced packing fraction η^* of 0.52356. Error bars are smaller than the symbol size.

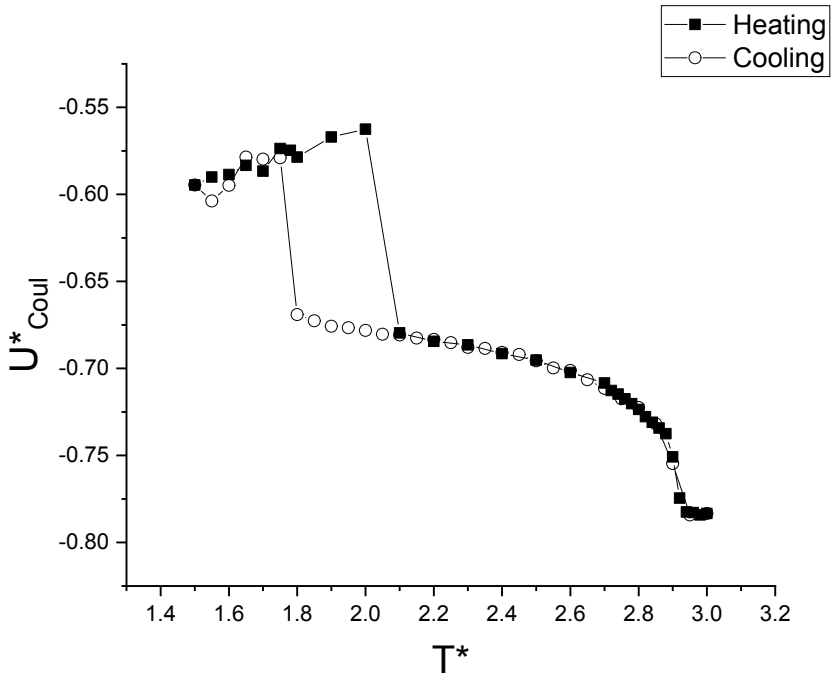

Figure S2. Reduced electrostatic energy, U^*_{coul}, as a function of the reduced temperature for the GB:LJ 2:1 system at reduced packing fraction η^* of 0.52356. Error bars are smaller than the symbol size.

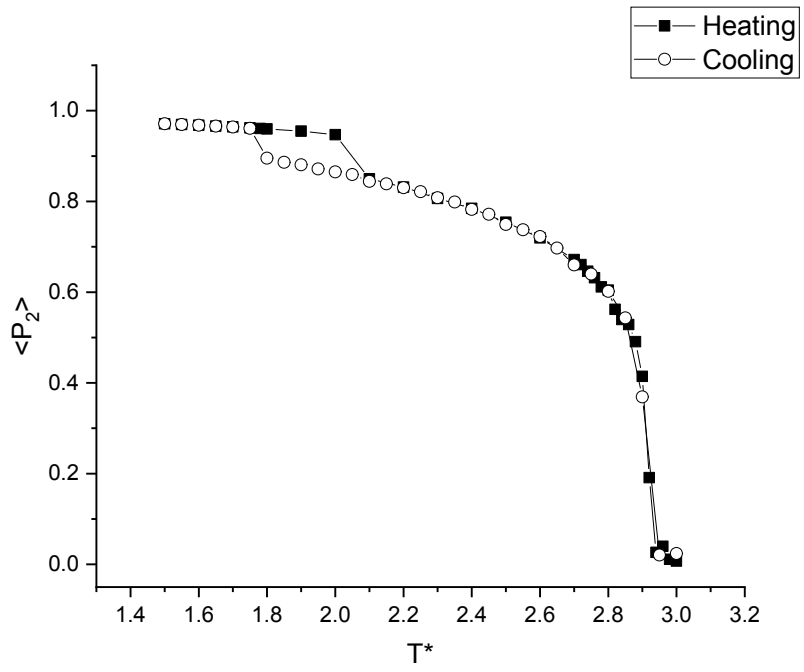

Figure S3. Orientational order parameter, $\langle P_2 \rangle$, as a function of the reduced temperature for the GB:LJ 2:1 system at reduced packing fraction η^* of 0.52356. Error bars are smaller than the symbol size.

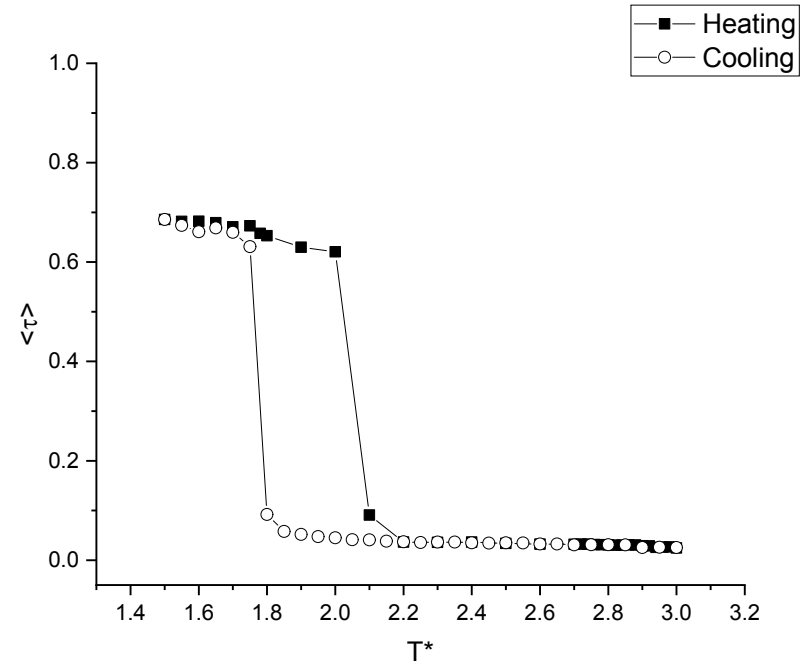

Figure S4. Translational order parameter, $\langle \tau \rangle$, as a function of the reduced temperature for the GB:LJ 2:1 system at reduced packing fraction η^* of 0.52356. Error bars are smaller than the symbol size.

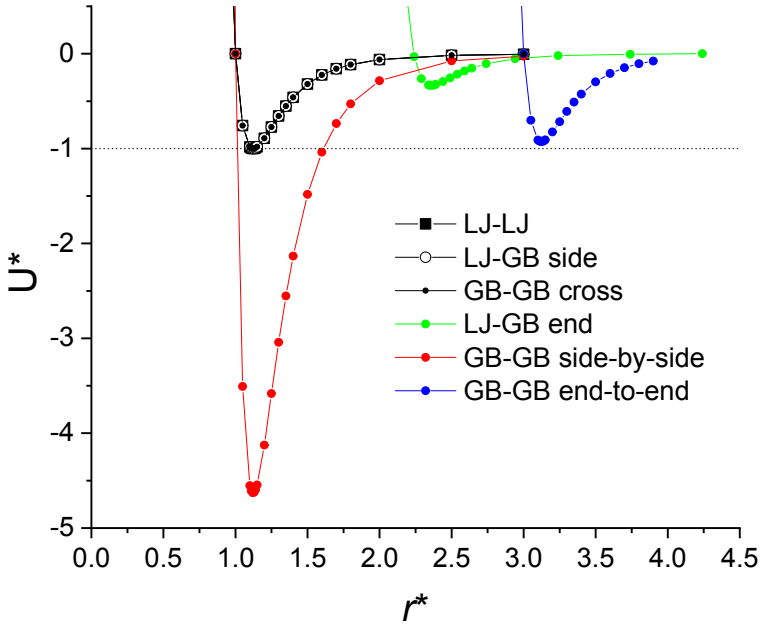

Figure S5. Pair interaction potentials for the most relevant particle-particle geometries. Test calculations have been run using the template input file below for Case 2).

```
#########################################################################################################################
##
## Example input files for MD simulations of mixtures of charged GB and LJ particles using LAMMPS
##
##   Please cite the following literature:
## - G. Saielli, K. Satoh, *Phys. Chem. Chem. Phys*. **2019** (this paper)
## - T. Margola, K. Satoh, G. Saielli, *Crystals* **2018**, 8, 371-15. DOI: http://dx.doi.org/10.3390/cryst8100371
## - G. Saielli, T. Margola, K. Satoh, *Soft Matter* **2017**, 13, 5207-5213. DOI: http://dx.doi.org/10.1039/C7SM00612H
## - T. Margola, G. Saielli, K. Satoh, *Mol. Cryst. Liq. Cryst.* **2017**, 649, 50-58. DOI: http://dx.doi.org/10.1080/15421406.2017.1303918
##
#########################################################################################################################
#
#
#
# Case 1) - Pure Gay-Berne ellipsoids, tested with version LAMMPS (17 Dec 2013).
#
# GB parameterization as in
# R. Berardi, A.P.J. Emerson, C. Zannoni,
# Monte Carlo investigations of a Gay-Berne liquid crystal,
# J. Chem. Soc. Faraday Trans., 89, 4069 - 4078 (1993)
#
#
units       lj
atom_style   ellipsoid
dimension    3

read_data    start.config

# set mass and shape
set         type 1 mass 1.0
set         type 1 shape 1 1 3

# define GB potential
pair_style   gayberne 1.0 3.0 1.0 4.0
pair_coeff   1 1 1.0 1.0 1.0 1.0 0.20 0 0 0
neighbor     0.8 bin
```

```
thermo      1000
timestep    0.0015

compute     q all property/atom quatw quati quatj quatk

dump        1 all custom 100000 trj.out &
          id type x y z c_q[1] c_q[2] c_q[3] c_q[4]

# Count number of uniaxial particles
group       uniaxial type 1
variable    dof equal count(uniaxial)

# MD simulations at constant volume
fix         1 all nvt/asphere temp 3.0 3.0 0.1

# Remove a degree of freedom for each uniaxial particle from the NVT temperature compute
compute_modify 1_temp extra ${dof}

# Only output the temperature and pressure used by NPT/asphere
thermo_style custom step c_1_temp epair pe etotal vol density press

run         1000000

write_restart restart.file

#
#
#
#
#
#
#
# Case 2) - Mixture of  non-charged Gay-Berne ellipsoids and LJ spheres, tested with  version LAMMPS (20 Jan 2015).
#
# GB parameterization as in
```

```
# R. Berardi, A.P.J. Emerson, C. Zannoni,
# Monte Carlo investigations of a Gay-Berne liquid crystal,
# J. Chem. Soc. Faraday Trans., 89, 4069 - 4078 (1993)
#
#
units       lj
atom_style   ellipsoid
dimension    3

read_data    start.config

set         type 1 mass 1.0
set         type 2 mass 1.0
set         type 1 shape 1 1 3
set         type 2 shape 1 1 1

group       gbgb   type 1
group       lennjo type 2

variable     dofGB equal count(gbgb)
variable     dofLJ equal count(lennjo)*3
compute      tempgb gbgb     temp/asphere      # for total(trans+rot) temp for GB
compute      templj lennjo   temp/asphere      # for total(trans+rot) temp for LJ
compute      kerotgb gbgb   erotate/asphere    # for rotational kinetic energy for GB
compute      kerotlj lennjo erotate/asphere    # for rotational kinetic energy for LJ
compute      ketragb gbgb   ke                 # for translational kinetic energy for GB
compute      ketralj lennjo ke                 # for translational kinetic energy for LJ

#          gayberne(shift_for_pot_min,nyu,myu,cutoff)
#
pair_style   gayberne 1.0 3.0 1.0 4.0

#  pair_coeff(type1 type2 unit_egy unit_length depth_x1 depth_y1 depth_z1
#                                  depth_x2 depth_y2 depth_z2 cutoff)
pair_coeff   1 1  1 1  1 1 0.2 1 1 0.2
pair_coeff   1 2  1 1  1 1 0.2 1 1 1
```

```
pair_coeff   2 2  1 1  1 1 1 1 1 1


neighbor     0.8 bin


thermo       100


timestep     0.0015


compute      q all property/atom quatw quati quatj quatk


dump         1 all custom 10000 trj.GB &
             id type x y z c_q[1] c_q[2] c_q[3] c_q[4]


fix          1 gbgb   nvt/asphere temp 1.65 1.65 0.1
fix          2 lennjo nvt/asphere temp 1.65 1.65 0.1


compute_modify 1_temp extra ${dofGB}
compute_modify 2_temp extra ${dofLJ}
compute_modify tempgb extra ${dofGB}
compute_modify templj extra ${dofLJ}


thermo_style custom step pe ke c_ketragb c_kerotgb c_tempgb c_ketralj c_kerotlj c_templj temp c_1_temp c_2_temp press vol


restart 10000 restart_a restart_b


run          1000000
write_restart restart.file


#
#
#
#
#
#
#
```

```
# Case 3) - Mixture of charged Gay-Berne ellipsoids and LJ spheres, tested with versions LAMMPS (20 Jan 2015), LAMMPS (28 Sep 2016) and LAMMPS (11 Ago 2017).

#

# GB parameterization as in

# R. Berardi, A.P.J. Emerson, C. Zannoni,

# Monte Carlo investigations of a Gay-Berne liquid crystal,

# J. Chem. Soc. Faraday Trans., 89, 4069 - 4078 (1993)

#

#

units       lj

atom_style   hybrid ellipsoid charge

dimension    3


read_data    start.config


set         type 1 mass 1.0

set         type 2 mass 1.0

set         type 1 shape 1 1 3

set         type 2 shape 1 1 1


group       gbgb   type 1

group       lennjo type 2


set group gbgb charge 2.0

set group lennjo charge -2.0


variable     dofGB equal count(gbgb)

variable     dofLJ equal count(lennjo)*3

compute      tempgb gbgb     temp/asphere      # for total(trans+rot) temp for GB

compute      templj lennjo   temp/asphere      # for total(trans+rot) temp for LJ

compute      kerotgb gbgb   erotate/asphere    # for rotational kinetic energy for GB

compute      kerotlj lennjo erotate/asphere    # for rotational kinetic energy for LJ

compute      ketragb gbgb   ke                # for translational kinetic energy for GB

compute      ketralj lennjo ke                # for translational kinetic energy for LJ


#          gayberne(shift_for_pot_min,nyu,myu,cutoff)
```

```
#
#pair_style   gayberne 1.0 3.0 1.0 4.0
pair_style   hybrid/overlay gayberne 1.0 3.0 1.0 4.0 coul/long 10.0
#  pair_coeff(type1 type2 unit_egy unit_length depth_x1 depth_y1 depth_z1
#                                   depth_x2 depth_y2 depth_z2 cutoff)

kspace_style pppm 1.0e-6

pair_coeff   1 1  gayberne 1 1  1 1 0.2 1 1 0.2
pair_coeff   1 2  gayberne 1 1  1 1 0.2 1 1 1
pair_coeff   2 2  gayberne 1 1  1 1 1 1 1 1

pair_coeff 1 1 coul/long #10.0
pair_coeff 1 2 coul/long #10.0
pair_coeff 2 2 coul/long #10.0


neighbor     0.8 bin

thermo       1000

timestep     0.0015

compute      q all property/atom quatw quati quatj quatk

dump         1 all custom 1000 trj.GB &
           id type x y z c_q[1] c_q[2] c_q[3] c_q[4]

fix          1 gbgb   nvt/asphere temp 1.40 1.40 0.1
fix          2 lennjo nvt/asphere temp 1.40 1.40 0.1

compute_modify 1_temp extra ${dofGB}
compute_modify 2_temp extra ${dofLJ}
compute_modify tempgb extra ${dofGB}
compute_modify templj extra ${dofLJ}
```

thermo_style custom step pe ecoul elong ke c_ketragb c_kerotgb c_tempgb c_ketralj c_kerotlj c_templj temp c_1_temp c_2_temp press vol

mass 1 1.

mass 2 1.

restart 10000 restart_a restart_b

run 1000000

write_restart restart.file