

Supporting Information

Crystal-to-Crystal Transitions in Binary Mixtures of Soft Colloids

Jasper N. Immink^{†,*}, Maxime J. Bergman[‡], J. J. Erik Maris^{||}, Joakim Stenhammar[†], Peter Schurtenberger^{†,⊥}

[†] Division of Physical Chemistry, Lund University, Lund, Sweden.

[‡] Department of Physics, University of Fribourg, Fribourg, Switzerland.

^{||} Inorganic Chemistry and Catalysis Group, Utrecht University, Utrecht, the Netherlands.

[⊥] Lund Institute of advanced Neutron and X-ray Science (LINXS), Lund University, Lund, Sweden.

* Tel: +46-46-2223677; E-mail: jasper.immink@fkem1.lu.se.

In Figs. 1-3, we compare the experimental RDFs at $1.25 \cdot 10^{-3}$ M KCl with theoretical RDFs of three different crystal types. These comparisons show a good match between experimental RDFs and theoretical AuCu crystal RDFs, and a dissimilarity between experimental RDFs and theoretical CsCl and NaCl crystal RDFs. We have added graphical representations of the corresponding unit cells to each figure. Unit cell representations are made with OVITO (Ref. 51 in the main paper).

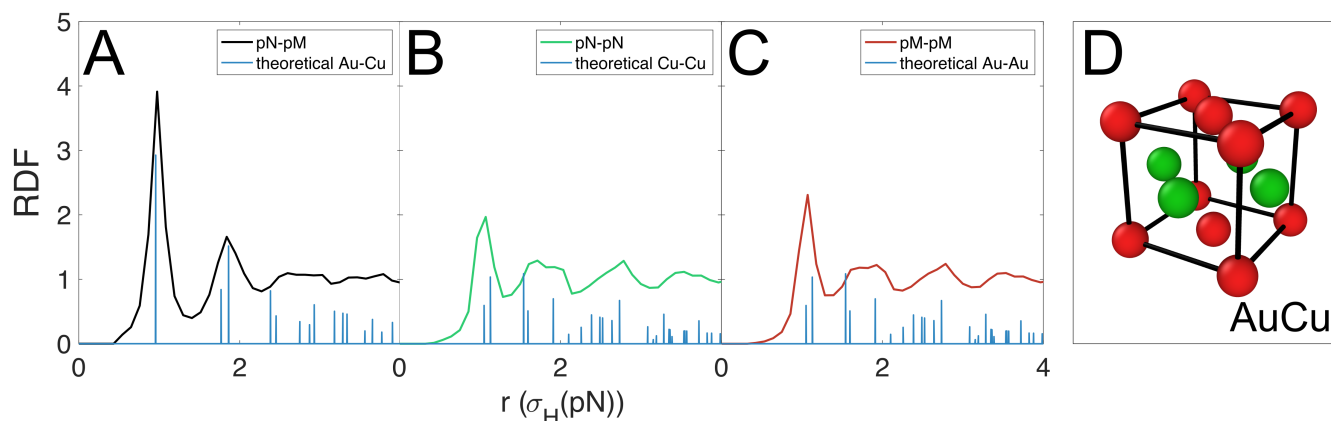


Figure 1: Radial distribution functions (RDFs) compared to the theoretical RDF of a perfect AuCu crystal, together with the corresponding AuCu unit cell. Theoretical RDF's are rescaled as in the main manuscript. The Au atom is the larger atom in an AuCu crystal. A) pN-to-pM RDF, compared to the theoretical Au-to-Cu RDF. B) pN-to-pN RDF, compared to the theoretical Cu-to-Cu RDF. C) pM-to-pM RDF, compared to the theoretical Au-to-Au RDF. D) Unit cell of an AuCu crystal.

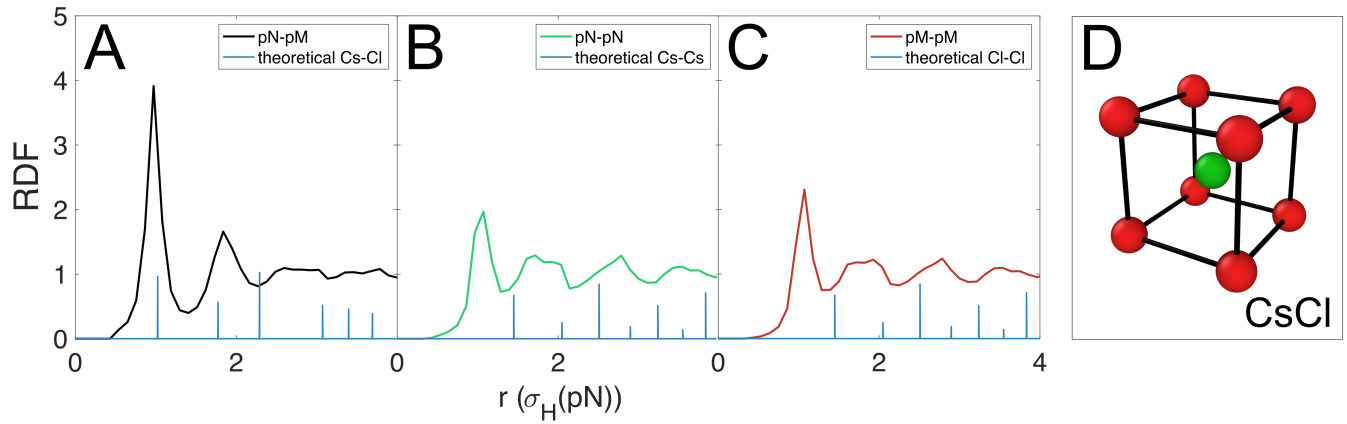


Figure 2: Radial distribution functions compared to the theoretical RDF of a perfect CsCl crystal, together with the corresponding CsCl unit cell. Theoretical RDF's are rescaled as in the main manuscript. The Cl ion is the larger atom in a CsCl crystal. A) pN-to-pM RDF, compared to Cs-to-Cl the theoretical RDF. B) pN-to-pN RDF, compared to Cs-to-Cs the theoretical RDF. C) pM-to-pM RDF, compared to Cl-to-Cl the theoretical RDF. Note the poor correspondence with the experimental RDFs compared to the AuCu structure. D) Unit cell of an CsCl crystal.

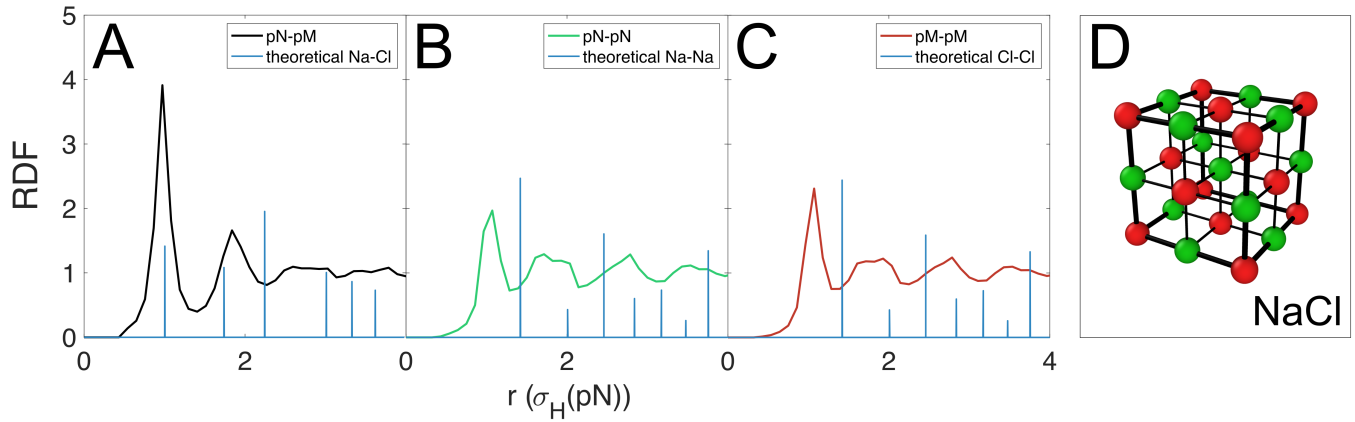


Figure 3: Radial distribution functions compared to the theoretical RDF of a perfect NaCl crystal, together with the corresponding NaCl unit cell. Theoretical RDF's are rescaled as in the main manuscript. The Cl ion is the larger atom in a NaCl crystal. A) pN-to-pM RDF, compared to Na-to-Cl the theoretical RDF. B) pN-to-pN RDF, compared to Na-to-Na the theoretical RDF. C) pM-to-pM RDF, compared to Cl-to-Cl the theoretical RDF. Note the poor correspondence with the experimental RDFs compared to the AuCu structure. D) Unit cell of an NaCl crystal.

In Tables 1 and 2, we show the average q_l and \bar{q}_l values of all particles in $2.0 \cdot 10^{-3}\text{M}$ KCl or $1.25 \cdot 10^{-3}\text{M}$ KCl, together with their standard deviations. It is important to note that values diverge from figures in Table 1 due to experimental noise and inherent limitations, particularly from a limited resolution in microscopy, and due to the values encompassing defect particle figures. Furthermore, the \bar{q}_l values are generally lower than the q_l values: this arises from \bar{q}_l comprising not only the deviations from perfect crystalline order in the immediate surrounding, but also the surrounding of neighboring particles. Using \bar{q}_l rather than q_l is nonetheless necessary to suppress individual FCC-particles, that randomly have an AuCu-type surrounding, to be designated as belonging to an AuCu phase.

Table 1: Obtained 3D local bond order parameters for all particles in $2.0 \cdot 10^{-3}\text{M}$ KCl. q_l is split in $q_l(\text{all})$, $q_l(\text{equal})$ and $q_l(\text{unequal})$.

Crystal	q_4	q_6	q_8	\bar{q}_4	\bar{q}_6	\bar{q}_8
$q_l(\text{all})$	0.174 ± 0.058	0.460 ± 0.040	0.347 ± 0.036	0.088 ± 0.027	0.415 ± 0.031	0.283 ± 0.026
$q_l(\text{equal})$	0.356 ± 0.119	0.546 ± 0.089	0.458 ± 0.088	0.148 ± 0.060	0.441 ± 0.052	0.316 ± 0.043
$q_l(\text{unequal})$	0.385 ± 0.140	0.555 ± 0.101	0.469 ± 0.108	0.169 ± 0.064	0.430 ± 0.055	0.311 ± 0.048

Table 2: Obtained 3D local bond order parameters for all particles in $1.25 \cdot 10^{-3}\text{M}$ KCl. q_l is split in $q_l(\text{all})$, $q_l(\text{equal})$ and $q_l(\text{unequal})$.

Crystal	q_4	q_6	q_8	\bar{q}_4	\bar{q}_6	\bar{q}_8
$q_l(\text{all})$	0.271 ± 0.085	0.386 ± 0.073	0.367 ± 0.060	0.160 ± 0.045	0.287 ± 0.053	0.263 ± 0.041
$q_l(\text{equal})$	0.768 ± 0.147	0.644 ± 0.198	0.744 ± 0.136	0.604 ± 0.131	0.456 ± 0.140	0.558 ± 0.101
$q_l(\text{unequal})$	0.445 ± 0.093	0.400 ± 0.091	0.370 ± 0.094	0.346 ± 0.075	0.280 ± 0.054	0.234 ± 0.049