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

Generalised Dissipative Particle Dynamics with Energy Conservation: Density- and Temperature-Dependent Potentials

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SUPPLEMENTAL MATERIAL

A. Scheme to prevent numerical instabilities for the vdW EOS

A scheme to prevent numerical instabilities when using the vdW EOS as a many-body force field at low and high densities is presented. At low particle densities, a minimum value of 10^{-6} is set. At high particle densities, a maximum density, n_{max}, can be chosen such that the vdW EOS thermodynamic quantities are continuous and vary relatively smoothly beyond n_{max}, based on the specified c and Δ parameters. For the data reported in the main text, c=2 and Δ=0.05 have been used. Defining $n_{max} = \frac{1-\Delta}{b_{vdW}}$, then for $n \geq n_{max}$, the particle pressure is

$$\pi^{ex} = \pi^{ex}_{max} \left(\frac{n}{n_{max}} \right)^c \tag{1}$$

where

$$\pi^{ex}_{max} = \left(\frac{b_{vdW}\theta}{1 - b_{vdW} n_{max}} - a_{vdW} \right) n^2_{max} \tag{2}$$

The particle internal energy is then

$$u = C_V \theta - a_{vdW} n_{max} \left(\frac{n}{n_{max}} \right)^{c-1} \tag{3}$$

while the particle entropy is

$$s = C_V \ln \theta - k_B \ln \left(\frac{1}{1 - b_{vdW} n_{max}} \right) - \frac{k_B b_{vdW} n_{max}}{(c-1)(1 - b_{vdW} n_{max})} \left(\left(\frac{n}{n_{max}} \right)^{c-1} - 1 \right) \tag{4}$$

Finally, the particle Helmholtz free-energy is

$$f = u - \theta s \tag{5}$$

B. Proof of Maxwell relation for a conservative potential: Application to a many-body force field based on an equation-of-state (MB-FF-EOS) within the generalised DPDE framework

The intent of this proof is to demonstrate that if a MB-FF-EOS is implemented in the generalised DPDE framework, it satisfies a necessary condition of a conservative potential. As such, any lack of energy conservation observed in the generalised DPDE simulation is caused by standard numerical integration errors.

The proof follows the work of Warren [1] and Moore *et al* [2] who considered a many-body force field that has both a distance and density dependence, where they satisfied the necessary requirements relating the corresponding weighting functions.

Consider a MB-FF-EOS for an ideal gas in the generalised DPDE framework. For a given particle i, the conservative force based on the excess free-energy (ψ_i^{ex}) can be derived as a function of the weighted local average density (n_i) through [3]

$$\frac{\partial \psi_i^{ex}}{\partial n_i} = \frac{\eta_i \theta_i}{n_i} \tag{6}$$

where n_i is defined in Eq. (2) of the manuscript, $\eta_i = N_{CG} - 1$, which is related to coarse-grain mapping between N particles into N_{CG} particles through $\phi_i = \frac{N}{N_{CG}}$, and θ_i is the particle internal temperature. Note that $n_i = n_i(r_{ij})$, while $\theta_i \neq \theta_i(r_{ij})$. As in the manuscript, all terms are taken in reduced units.

Here, without loss of generality, the level of coarse-graining for all particles is taken to be the same, such that $\eta = \eta_i = \eta_j$.

Consider then the pairwise forces for the one-dimensional case [4],

$$F_{ij} = -\left(\frac{\eta\theta_i}{n_i} + \frac{\eta\theta_j}{n_j}\right)\omega'(x_{ij}) \tag{7}$$

where $\omega'(x_{ij}) \equiv \frac{\partial\omega(x_{ij})}{\partial x_{ij}}$.

For a three-particle system [1],

$$F_{12} = -F_{21} = -\eta\left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\omega'(x_{12}) \tag{8}$$

$$F_{13} = -F_{31} = -\eta\left(\frac{\theta_1}{n_1} + \frac{\theta_3}{n_3}\right)\omega'(x_{13}) \tag{9}$$

$$F_{23} = -F_{32} = -\eta\left(\frac{\theta_2}{n_2} + \frac{\theta_3}{n_3}\right)\omega'(x_{23}) \tag{10}$$

For each particle, the local densities are

$$n_1 = \omega(x_{12}) + \omega(x_{13}) \tag{11}$$

$$n_2 = \omega(x_{12}) + \omega(x_{23}) \tag{12}$$
$$n_3 = \omega(x_{13}) + \omega(x_{23}) \tag{13}$$

where $x_{ij} \equiv x_i - x_j$.

The total forces are

$$F_1 = F_{12} + F_{13} \tag{14}$$
$$F_2 = F_{21} + F_{23} \tag{15}$$
$$F_3 = F_{31} + F_{32} \tag{16}$$

For the one-dimensional case, a necessary condition for the MB-FF-EOS to be a conservative potential is that the following Maxwell relation is satisfied [1]:

$$D_{ij} = \frac{\partial F_i}{\partial x_j} - \frac{\partial F_j}{\partial x_i} = 0 \tag{17}$$

To verify this relation, consider particles 1 and 2. The 1st term in Eq. (17) is then

$$\frac{\partial F_1}{\partial x_2} = \frac{\partial (F_{12} + F_{13})}{\partial x_2} = \frac{\partial F_{12}}{\partial x_2} + \frac{\partial F_{13}}{\partial x_2} \tag{18}$$

Using Eq. (8), the 1st term in Eq. (18) is then

$$\frac{\partial F_{12}}{\partial x_2} = -\eta \left[\frac{\partial}{\partial x_2}\left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\omega'(x_{12}) + \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_2}(\omega'(x_{12}))\right] \tag{19}$$

Since $\theta_i \neq \theta_i(x_{ij})$

$$\frac{\partial F_{12}}{\partial x_2} = -\eta \left[\left(\theta_1 \frac{\partial}{\partial x_2}\left(\frac{1}{n_1}\right) + \theta_2 \frac{\partial}{\partial x_2}\left(\frac{1}{n_2}\right)\right)\omega'(x_{12}) + \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_2}(\omega'(x_{12}))\right] \tag{20}$$

Likewise for the 2nd term in Eq. (18)

$$\frac{\partial F_{13}}{\partial x_2} = -\eta \left[\left(\theta_1 \frac{\partial}{\partial x_2}\left(\frac{1}{n_1}\right) + \theta_3 \frac{\partial}{\partial x_2}\left(\frac{1}{n_3}\right)\right)\omega'(x_{13}) + \left(\frac{\theta_1}{n_1} + \frac{\theta_3}{n_3}\right)\frac{\partial}{\partial x_2}(\omega'(x_{13}))\right] \tag{21}$$

but $\frac{\partial}{\partial x_2}(\omega'(x_{13})) = 0$, so

$$\frac{\partial F_{13}}{\partial x_2} = -\eta \left[\left(\theta_1 \frac{\partial}{\partial x_2}\left(\frac{1}{n_1}\right) + \theta_3 \frac{\partial}{\partial x_2}\left(\frac{1}{n_3}\right)\right)\omega'(x_{13})\right] \tag{22}$$

Analogous derivations follow for the 2nd term in Eq. (17),

$$\frac{\partial F_2}{\partial x_1} = \frac{\partial (F_{21} + F_{23})}{\partial x_1} = \frac{\partial F_{21}}{\partial x_1} + \frac{\partial F_{23}}{\partial x_1} \tag{23}$$

The 1st term in Eq. (23) can also be written $\frac{\partial F_{21}}{\partial x_1} = -\frac{\partial F_{12}}{\partial x_1}$, so

$$-\frac{\partial F_{12}}{\partial x_1} = +\eta \left[\left(\theta_1 \frac{\partial}{\partial x_1}\left(\frac{1}{n_1}\right) + \theta_2 \frac{\partial}{\partial x_1}\left(\frac{1}{n_2}\right) \right) \omega'(x_{12}) + \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right) \frac{\partial}{\partial x_1}(\omega'(x_{12})) \right] \tag{24}$$

For the 2nd term in Eq. (23),

$$\frac{\partial F_{23}}{\partial x_1} = -\eta \left[\left(\theta_2 \frac{\partial}{\partial x_1}\left(\frac{1}{n_2}\right) + \theta_3 \frac{\partial}{\partial x_1}\left(\frac{1}{n_3}\right) \right) \omega'(x_{23}) + \left(\frac{\theta_2}{n_2} + \frac{\theta_3}{n_3}\right) \frac{\partial}{\partial x_1}(\omega'(x_{23})) \right] \tag{25}$$

but $\frac{\partial}{\partial x_1}(\omega'(x_{23})) = 0$, so

$$\frac{\partial F_{23}}{\partial x_1} = -\eta \left[\left(\theta_2 \frac{\partial}{\partial x_1}\left(\frac{1}{n_2}\right) + \theta_3 \frac{\partial}{\partial x_1}\left(\frac{1}{n_3}\right) \right) \omega'(x_{23}) \right] \tag{26}$$

Next, the various derivative terms with respect to local density in Equations (20), (22), (24) and (26) are determined

$$\frac{\partial}{\partial x_2}\left(\frac{1}{n_1}\right) = -\left(\frac{1}{n_1}\right)^2 \frac{\partial n_1}{\partial x_2} = -\left(\frac{1}{n_1}\right)^2 \left(-\omega'(x_{12})\right) = \frac{\omega'(x_{12})}{n_1^2} \tag{27}$$

$\frac{\partial n_1}{\partial x_2}$ has been determined from Eq. (11) using the chain rule, $\frac{\partial n_1}{\partial x_2} = \frac{\partial \omega(x_{12})}{\partial x_2} = \frac{\partial \omega(x_{12})}{\partial x_{12}} \frac{\partial x_{12}}{\partial x_2}$, where $\frac{\partial x_{ij}}{\partial x_i} = -\frac{\partial x_{ij}}{\partial x_j}$ and recalling that $\omega'(x_{ij}) \equiv \frac{\partial \omega(x_{ij})}{\partial x_{ij}}$ and $x_{ij} \equiv x_i - x_j$.

The remaining terms are determined analogously using Equations (11)-(13).

$$\frac{\partial}{\partial x_2}\left(\frac{1}{n_2}\right) = -\left(\frac{1}{n_2}\right)^2 \frac{\partial n_2}{\partial x_2} = -\left(\frac{1}{n_2}\right)^2 \left(-\omega'(x_{12}) + \omega'(x_{23})\right) = \frac{\left(\omega'(x_{12}) - \omega'(x_{23})\right)}{n_2^2} \tag{28}$$

$$\frac{\partial}{\partial x_2}\left(\frac{1}{n_3}\right) = -\left(\frac{1}{n_3}\right)^2 \frac{\partial n_3}{\partial x_2} = -\left(\frac{1}{n_3}\right)^2 \left(\omega'(x_{23})\right) = -\frac{\omega'(x_{23})}{n_3^2} \tag{29}$$

$$\frac{\partial}{\partial x_1}\left(\frac{1}{n_1}\right) = -\left(\frac{1}{n_1}\right)^2 \frac{\partial n_1}{\partial x_1} = -\left(\frac{1}{n_1}\right)^2 \left(\omega'(x_{12}) + \omega'(x_{13})\right) = -\frac{\left(\omega'(x_{12}) + \omega'(x_{13})\right)}{n_1^2} \tag{30}$$

$$\frac{\partial}{\partial x_1}\left(\frac{1}{n_2}\right) = -\left(\frac{1}{n_2}\right)^2 \frac{\partial n_2}{\partial x_1} = -\left(\frac{1}{n_2}\right)^2 \left(\omega'(x_{12})\right) = -\frac{\omega'(x_{12})}{n_2^2} \tag{31}$$

$$\frac{\partial}{\partial x_1}\left(\frac{1}{n_3}\right) = -\left(\frac{1}{n_3}\right)^2 \frac{\partial n_3}{\partial x_1} = -\left(\frac{1}{n_3}\right)^2 \left(\omega'(x_{13})\right) = -\frac{\omega'(x_{13})}{n_3^2} \tag{32}$$

Using Equations (18) and (23), the terms required for Eq. (17) can now be determined.

$$D_{12} = \left(\frac{\partial F_{12}}{\partial x_2} + \frac{\partial F_{13}}{\partial x_2}\right) - \left(\frac{-\partial F_{12}}{\partial x_1} + \frac{\partial F_{23}}{\partial x_1}\right) \tag{33}$$

where Equations (27)-(32) can be substituted into these terms,

$$\begin{aligned}\frac{\partial F_{12}}{\partial x_2} = -\eta \Bigg[&\frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{12}) + \frac{\theta_2}{n_2^2}\omega'(x_{12})\omega'(x_{12}) - \frac{\theta_2}{n_2^2}\omega'(x_{23})\omega'(x_{12}) \\ &+ \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_2}(\omega'(x_{12}))\Bigg]\end{aligned} \tag{34}$$

$$\frac{\partial F_{13}}{\partial x_2} = -\eta \left[\frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{13}) - \frac{\theta_3}{n_3^2}\omega'(x_{23})\omega'(x_{13})\right] \tag{35}$$

$$\begin{aligned}\frac{\partial F_{12}}{\partial x_1} = +\eta \Bigg[&-\frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{12}) - \frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{13}) - \frac{\theta_2}{n_2^2}\omega'(x_{12})\omega'(x_{12}) \\ &+ \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_1}(\omega'(x_{12}))\Bigg]\end{aligned} \tag{36}$$

$$\frac{\partial F_{23}}{\partial x_1} = -\eta \left[-\frac{\theta_2}{n_2^2}\omega'(x_{12})\omega'(x_{23}) - \frac{\theta_3}{n_3^2}\omega'(x_{13})\omega'(x_{23})\right] \tag{37}$$

So then

$$\begin{aligned}\frac{D_{12}}{\eta} = \Bigg[&-\frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{12}) - \frac{\theta_2}{n_2^2}\omega'(x_{12})\omega'(x_{12}) + \frac{\theta_2}{n_2^2}\omega'(x_{23})\omega'(x_{12}) \\ &- \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_2}\big(\omega'(x_{12})\big) - \frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{13}) + \frac{\theta_3}{n_3^2}\omega'(x_{23})\omega'(x_{13}) \\ &+ \frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{12}) + \frac{\theta_1}{n_1^2}\omega'(x_{12})\omega'(x_{13}) + \frac{\theta_2}{n_2^2}\omega'(x_{12})\omega'(x_{12}) \\ &- \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_1}(\omega'(x_{12})) - \frac{\theta_2}{n_2^2}\omega'(x_{12})\omega'(x_{23}) - \frac{\theta_3}{n_3^2}\omega'(x_{13})\omega'(x_{23})\Bigg]\end{aligned} \tag{38}$$

Combining like terms

$$\begin{aligned}\frac{D_{12}}{\eta} = {}&\omega'(x_{12})\omega'(x_{12})\left[-\frac{\theta_1}{n_1^2} - \frac{\theta_2}{n_2^2} + \frac{\theta_1}{n_1^2} + \frac{\theta_2}{n_2^2}\right] + \omega'(x_{12})\omega'(x_{13})\left[-\frac{\theta_1}{n_1^2} + \frac{\theta_1}{n_1^2}\right] \\ &+ \omega'(x_{12})\omega'(x_{23})\left[\frac{\theta_2}{n_2^2} - \frac{\theta_2}{n_2^2}\right] + \omega'(x_{13})\omega'(x_{23})\left[\frac{\theta_3}{n_3^2} - \frac{\theta_3}{n_3^2}\right] \\ &- \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_2}\big(\omega'(x_{12})\big) - \left(\frac{\theta_1}{n_1} + \frac{\theta_2}{n_2}\right)\frac{\partial}{\partial x_1}(\omega'(x_{12}))\end{aligned} \tag{39}$$

Applying the chain rule $\frac{\partial}{\partial x_2}\big(\omega'(x_{12})\big) = \frac{\partial}{\partial x_{12}}\big(\omega'(x_{12})\big)\frac{\partial x_{12}}{\partial x_2}$, where again $\frac{\partial x_{ij}}{\partial x_i} = -\frac{\partial x_{ij}}{\partial x_j}$, so then

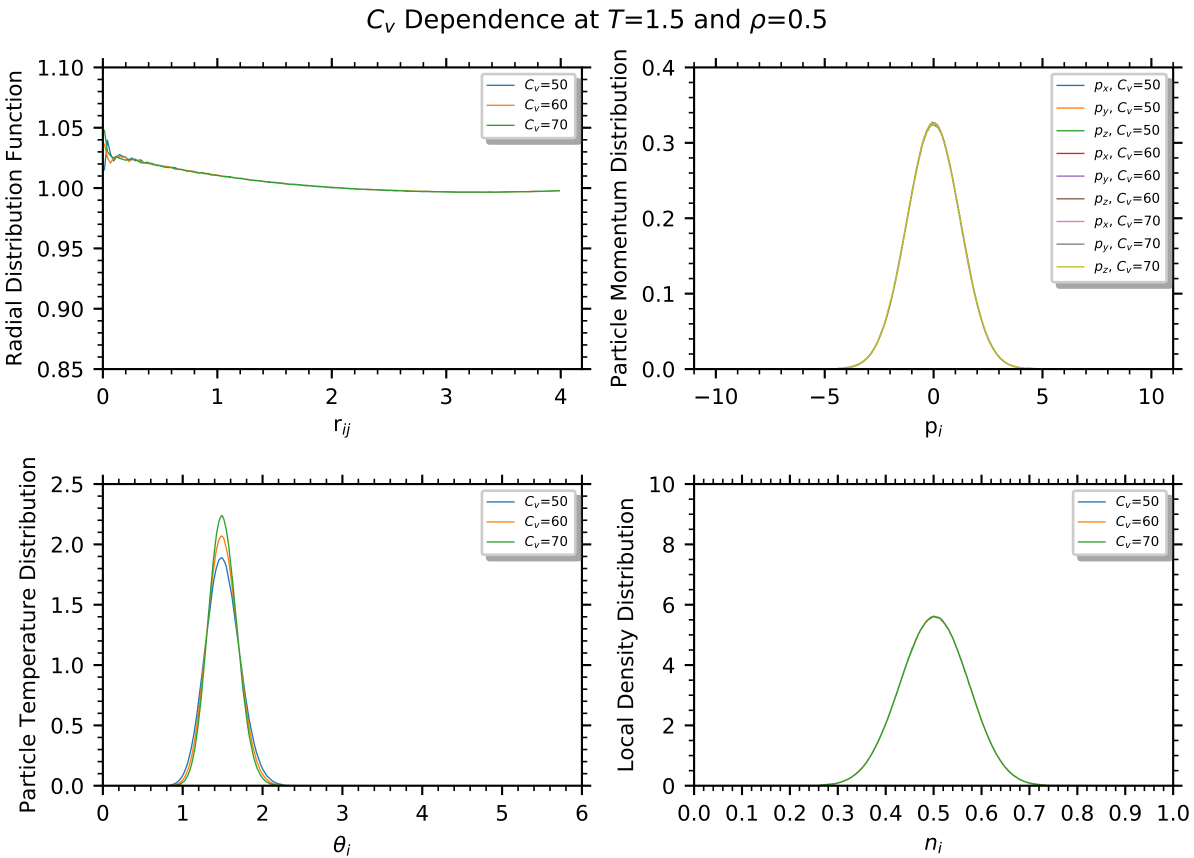
$$\frac{\partial}{\partial x_2}\big(\omega'(x_{12})\big) = -\frac{\partial}{\partial x_1}(\omega'(x_{12})) \tag{40}$$

Therefore, all terms become zero and the Maxwell relation for a conservative potential is satisfied, *i.e.*, $D_{12} = 0$.

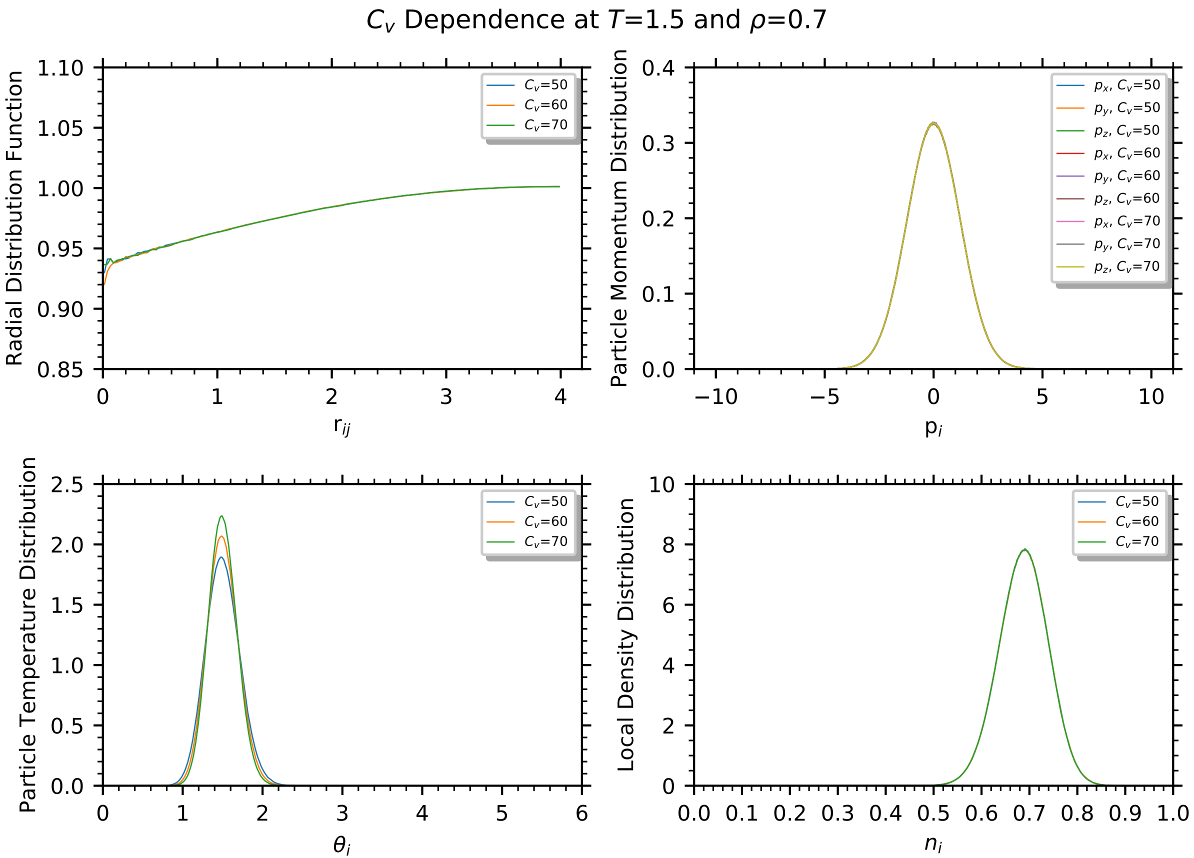
For completeness, the same procedure was applied to particles 2 and 3, to verify that $D_{23} = \frac{\partial F_2}{\partial x_3} - \frac{\partial F_3}{\partial x_2} = 0$.

C. Parametric study of the dependence of the heat capacity used in the mesoparticle equation-of-state C_V on the steady-state particle probability distributions.

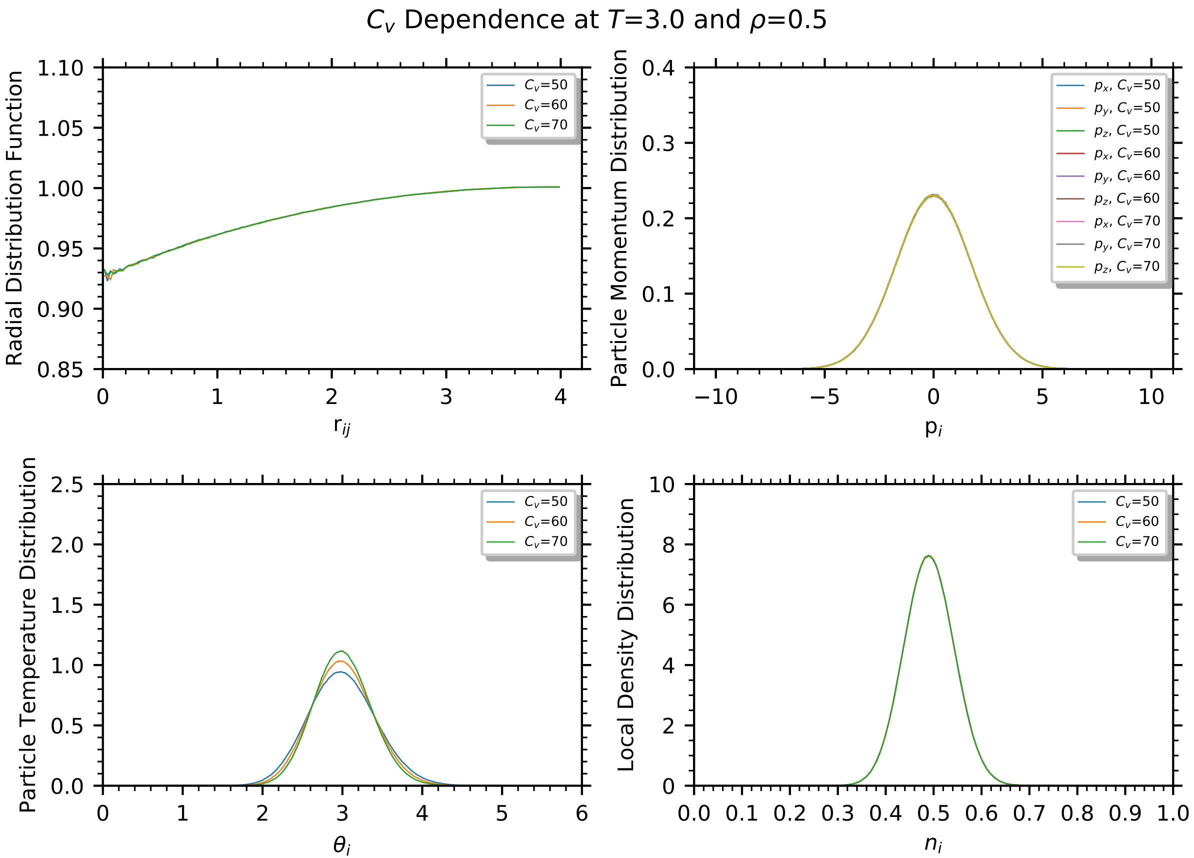
The figures in this section demonstrate the dependence of the heat capacity, C_V, on the equilibrium probability distributions for the LJ MB-FF-EOS at T = {1.5, 3.0} and ρ = {0.5, 0.7}: (top left) radial distribution function; (bottom left) particle temperature; (top right) particle momentum; (bottom right) local density.



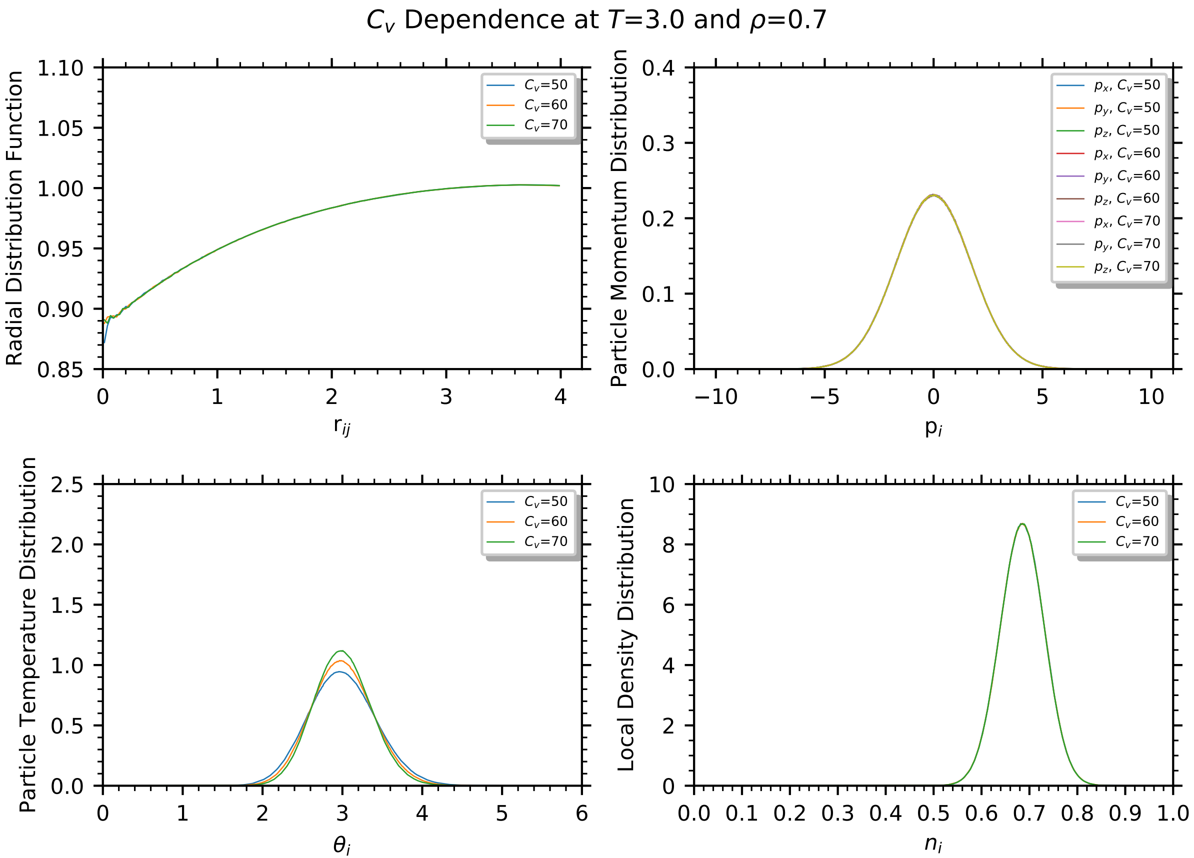
C_v Dependence at T=1.5 and ρ=0.5
Radial Distribution Function
r$_{ij}$
C_v=50
C_v=60
C_v=70
Particle Momentum Distribution
p$_i$
p_x, C_v=50
p_y, C_v=50
p_z, C_v=50
p_x, C_v=60
p_y, C_v=60
p_z, C_v=60
p_x, C_v=70
p_y, C_v=70
p_z, C_v=70
Particle Temperature Distribution
θ_i
Local Density Distribution
n_i



C_v Dependence at T=1.5 and ρ=0.7
Radial Distribution Function
r$_{ij}$
C_v=50
C_v=60
C_v=70
Particle Momentum Distribution
p$_i$
p_x, C_v=50
p_y, C_v=50
p_z, C_v=50
p_x, C_v=60
p_y, C_v=60
p_z, C_v=60
p_x, C_v=70
p_y, C_v=70
p_z, C_v=70
Particle Temperature Distribution
θ_i
C_v=50
C_v=60
C_v=70
Local Density Distribution
n_i
C_v=50
C_v=60
C_v=70



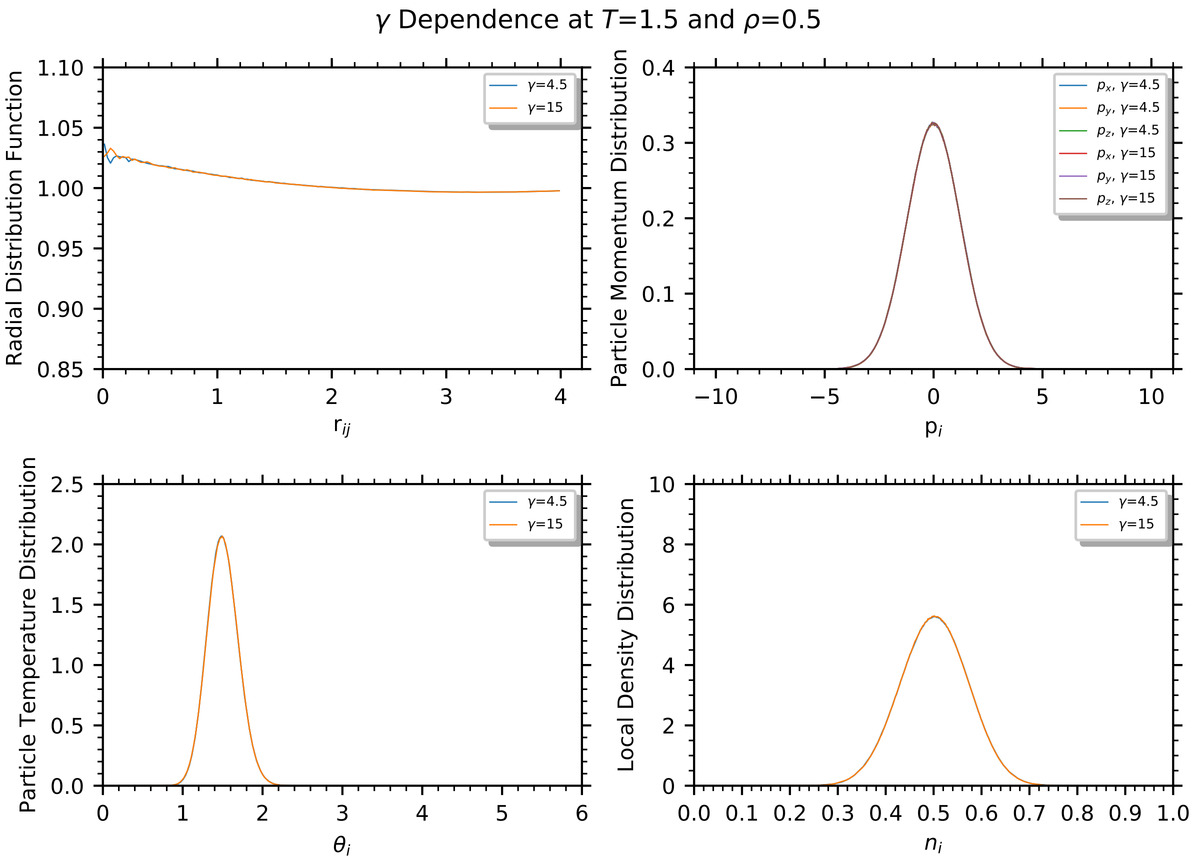
C_v Dependence at T=3.0 and ρ=0.5
Radial Distribution Function
r_{ij}
C_v=50
C_v=60
C_v=70
Particle Momentum Distribution
p_i
p_x, C_v=50
p_y, C_v=50
p_z, C_v=50
p_x, C_v=60
p_y, C_v=60
p_z, C_v=60
p_x, C_v=70
p_y, C_v=70
p_z, C_v=70
Particle Temperature Distribution
θ_i
Local Density Distribution
n_i



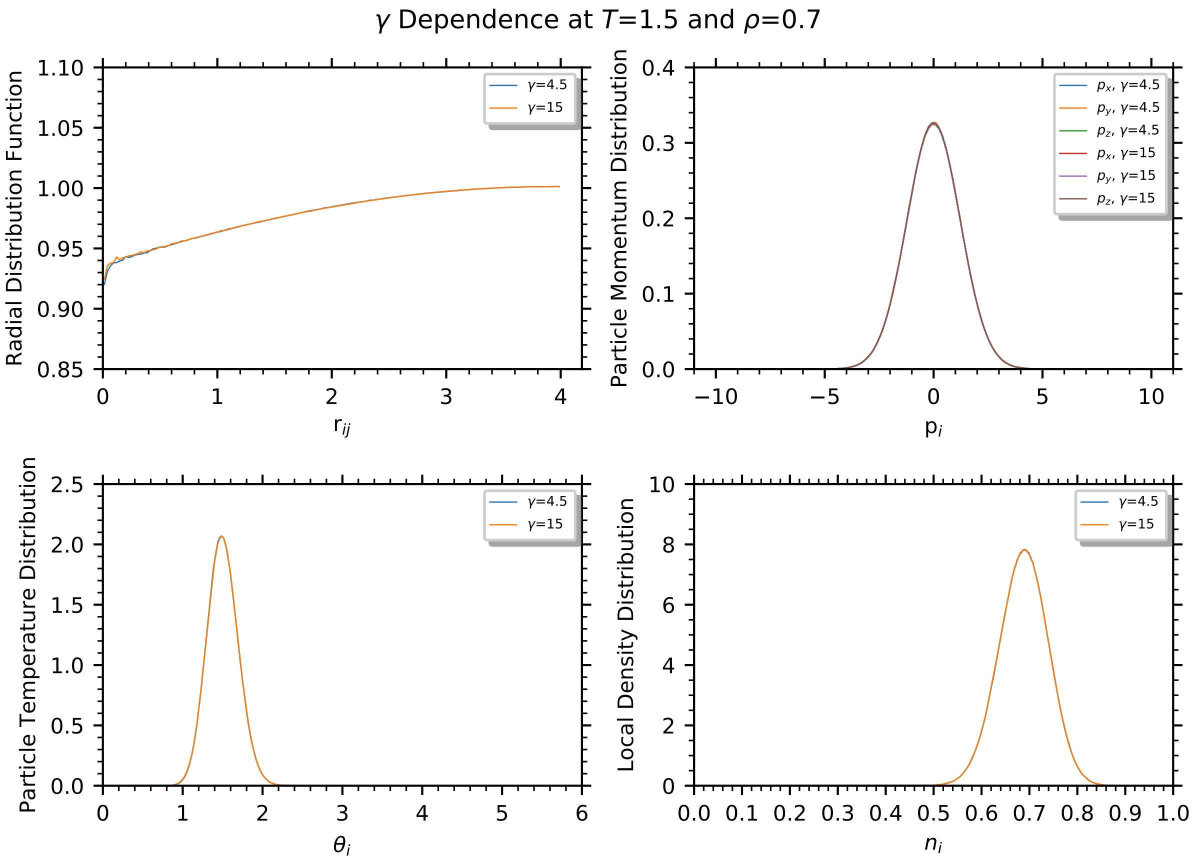
C_v Dependence at T=3.0 and ρ=0.7
Radial Distribution Function
r$_{ij}$
C_v=50
C_v=60
C_v=70
Particle Momentum Distribution
p$_i$
p_x, C_v=50
p_y, C_v=50
p_z, C_v=50
p_x, C_v=60
p_y, C_v=60
p_z, C_v=60
p_x, C_v=70
p_y, C_v=70
p_z, C_v=70
Particle Temperature Distribution
θ_i
Local Density Distribution
n_i

D. Parametric study of the dependence of the momentum coefficient γ on the steady-state particle probability distributions.

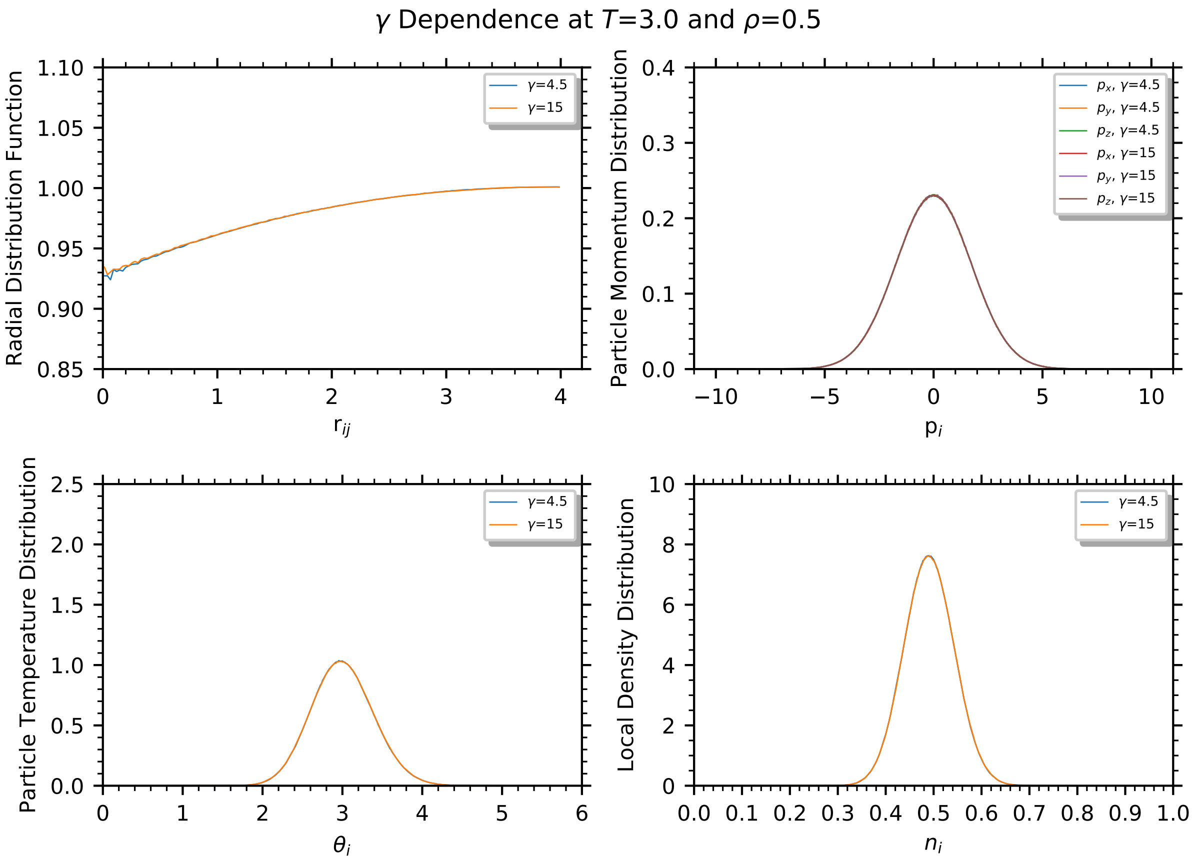
The figures in this section demonstrate the dependence of the momentum coefficient, γ, on the equilibrium probability distributions for the LJ MB-FF-EOS at T = {1.5, 3.0} and ρ = {0.5, 0.7}: (top left) radial distribution function; (bottom left) particle temperature; (top right) particle momentum; (bottom right) local density.

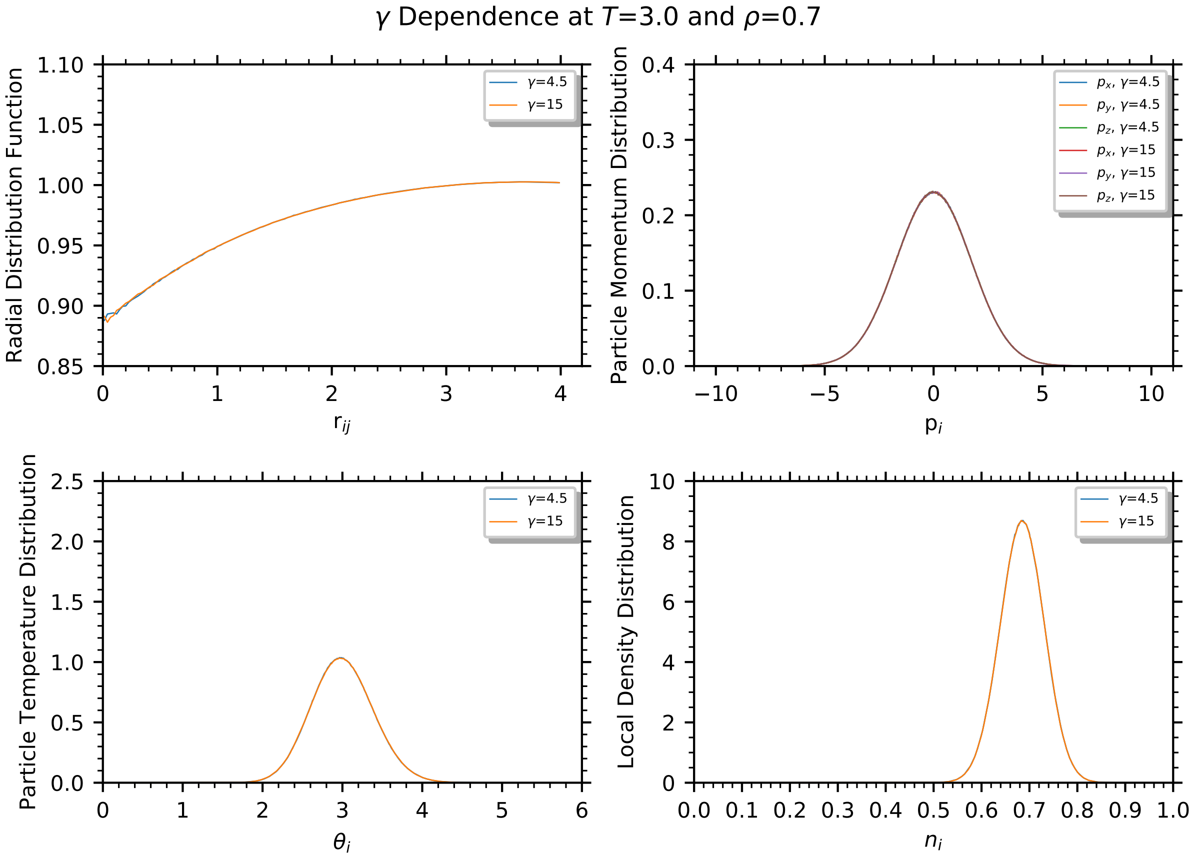


γ Dependence at T=1.5 and ρ=0.5
Radial Distribution Function
r_ij
γ=4.5
γ=15
Particle Momentum Distribution
p_i
p_x, γ=4.5
p_y, γ=4.5
p_z, γ=4.5
p_x, γ=15
p_y, γ=15
p_z, γ=15
Particle Temperature Distribution
θ_i
γ=4.5
γ=15
Local Density Distribution
n_i
γ=4.5
γ=15



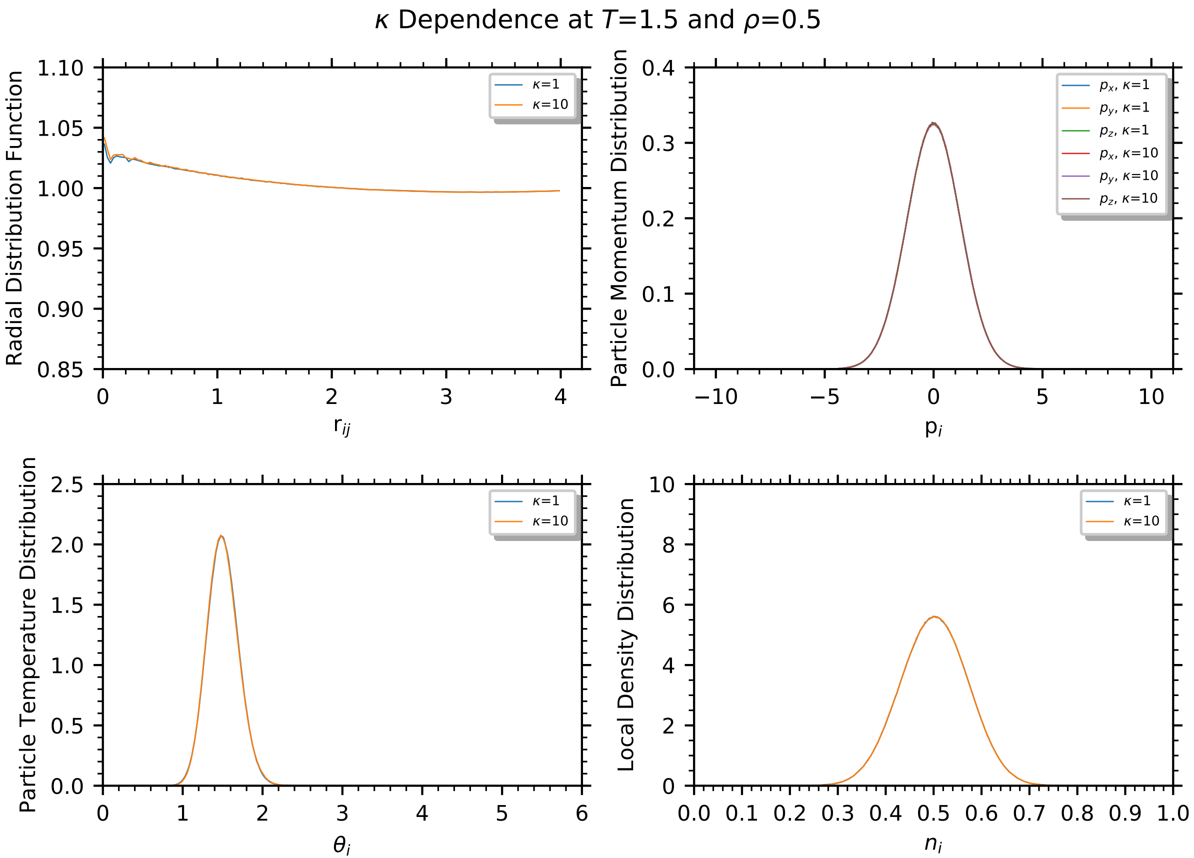
γ Dependence at T=1.5 and ρ=0.7
Radial Distribution Function
r_ij
γ=4.5
γ=15
Particle Momentum Distribution
p_i
p_x, γ=4.5
p_y, γ=4.5
p_z, γ=4.5
p_x, γ=15
p_y, γ=15
p_z, γ=15
Particle Temperature Distribution
θ_i
γ=4.5
γ=15
Local Density Distribution
n_i
γ=4.5
γ=15


γ Dependence at T=3.0 and ρ=0.5
Radial Distribution Function
r_ij
γ=4.5
γ=15
Particle Momentum Distribution
p_i
p_x, γ=4.5
p_y, γ=4.5
p_z, γ=4.5
p_x, γ=15
p_y, γ=15
p_z, γ=15
Particle Temperature Distribution
θ_i
γ=4.5
γ=15
Local Density Distribution
n_i
γ=4.5
γ=15

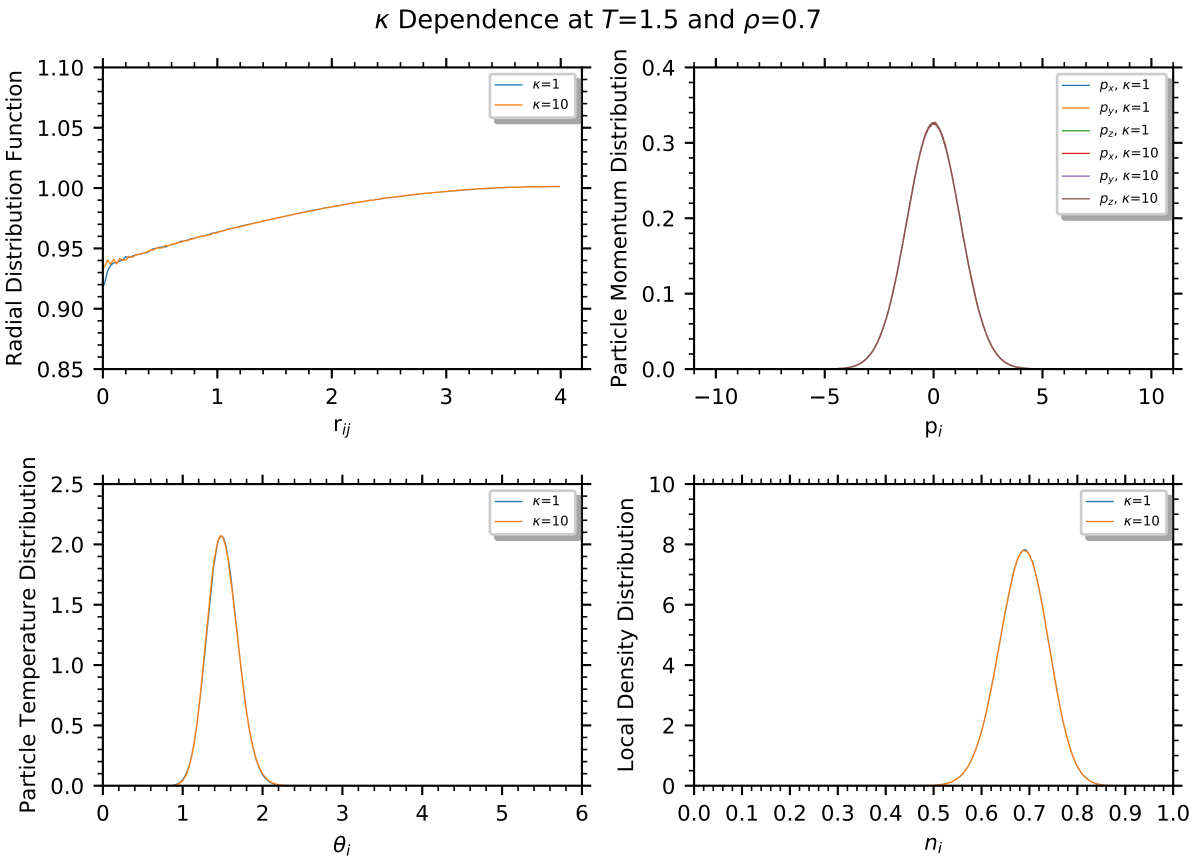

γ Dependence at T=3.0 and ρ=0.7
Radial Distribution Function
r_ij
γ=4.5
γ=15
Particle Momentum Distribution
p_i
p_x, γ=4.5
p_y, γ=4.5
p_z, γ=4.5
p_x, γ=15
p_y, γ=15
p_z, γ=15
Particle Temperature Distribution
θ_i
Local Density Distribution
n_i

E. Parametric study of the dependence of the heat coefficient κ on the steady-state particle probability distributions.

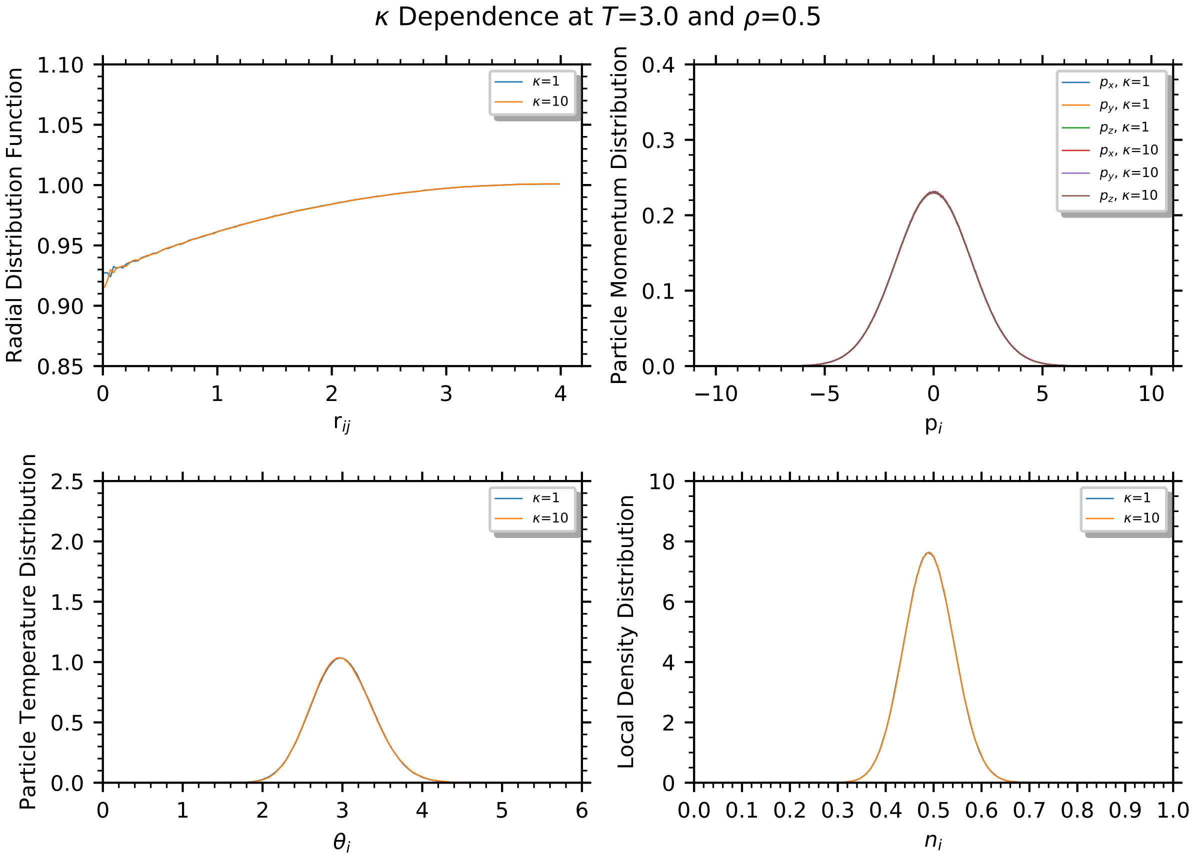
The figures in this section demonstrate the dependence of the heat coefficient, κ, on the equilibrium probability distributions for the LJ MB-FF-EOS at T = {1.5, 3.0} and ρ = {0.5, 0.7}: (top left) radial distribution function; (bottom left) particle temperature; (top right) particle momentum; (bottom right) local density.



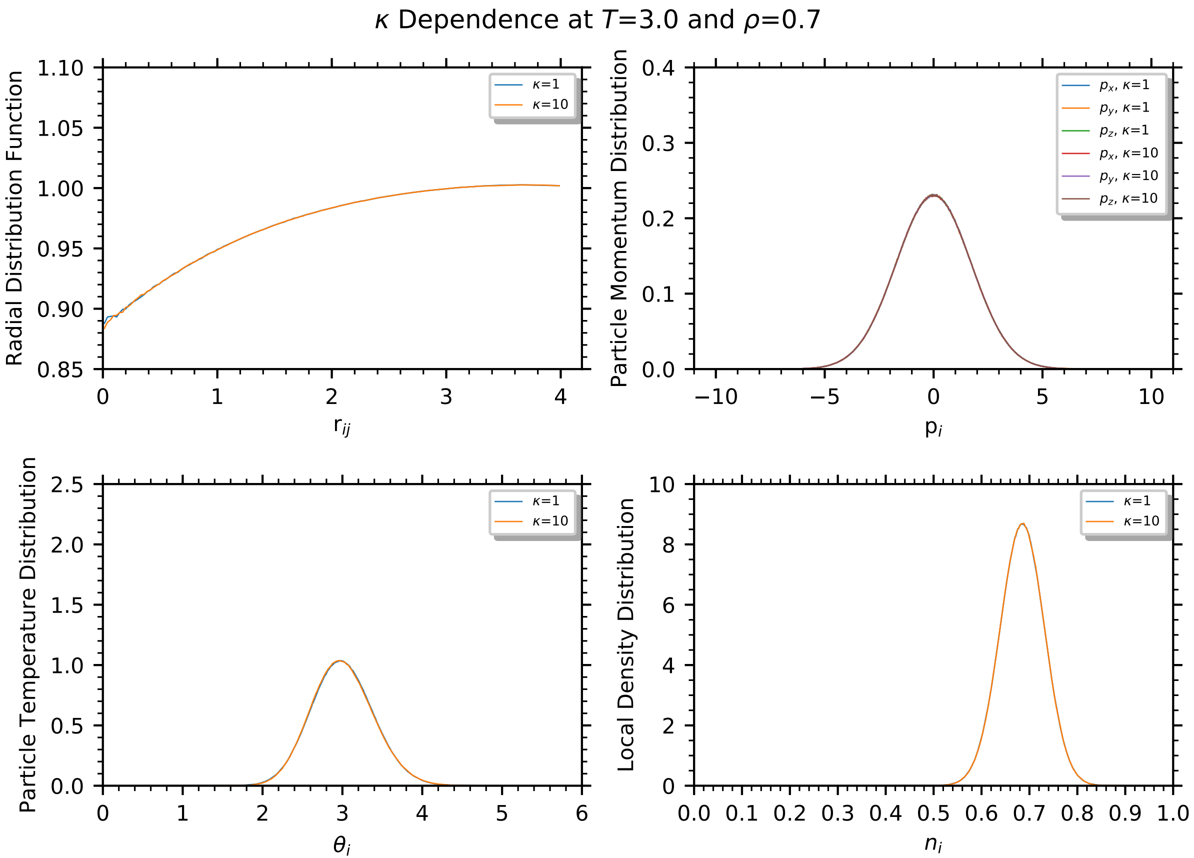
κ Dependence at T=1.5 and ρ=0.5
Radial Distribution Function
r_ij
κ=1
κ=10
Particle Momentum Distribution
p_i
p_x, κ=1
p_y, κ=1
p_z, κ=1
p_x, κ=10
p_y, κ=10
p_z, κ=10
Particle Temperature Distribution
θ_i
κ=1
κ=10
Local Density Distribution
n_i
κ=1
κ=10



κ Dependence at T=1.5 and ρ=0.7
Radial Distribution Function
r_ij
κ=1
κ=10
Particle Momentum Distribution
p_i
p_x, κ=1
p_y, κ=1
p_z, κ=1
p_x, κ=10
p_y, κ=10
p_z, κ=10
Particle Temperature Distribution
θ_i
Local Density Distribution
n_i



κ Dependence at T=3.0 and ρ=0.5
Radial Distribution Function
r_ij
κ=1
κ=10
Particle Momentum Distribution
p_i
p_x, κ=1
p_y, κ=1
p_z, κ=1
p_x, κ=10
p_y, κ=10
p_z, κ=10
Particle Temperature Distribution
θ_i
κ=1
κ=10
Local Density Distribution
n_i
κ=1
κ=10


κ Dependence at T=3.0 and ρ=0.7
Radial Distribution Function
r_ij
κ=1
κ=10
Particle Momentum Distribution
p_i
p_x, κ=1
p_y, κ=1
p_z, κ=1
p_x, κ=10
p_y, κ=10
p_z, κ=10
Particle Temperature Distribution
θ_i
κ=1
κ=10
Local Density Distribution
n_i
κ=1
κ=10

FIGURES

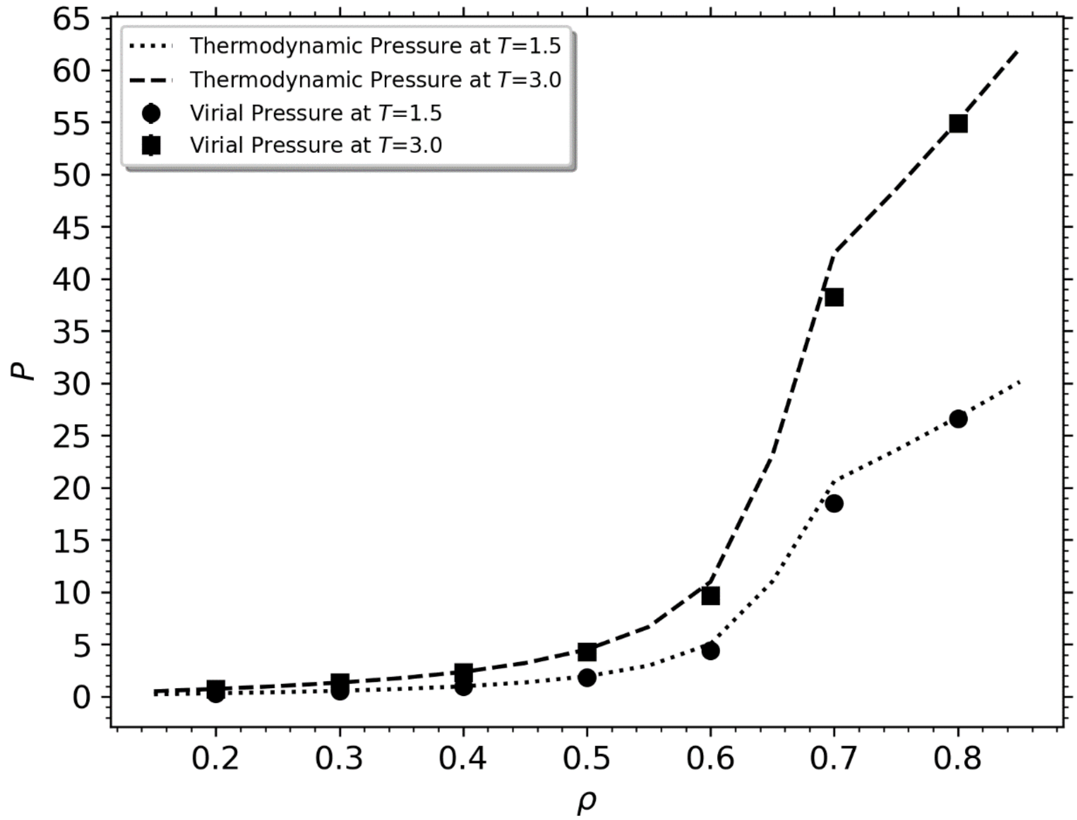


Figure S1: Comparison of the thermodynamic pressure calculated from the analytical van der Waals EOS (dashed lines) and the virial pressure simulated using the van der Waals EOS in the generalised DPDE method (symbols) at T=1.5 (circles) and T=3.0 (squares). The changes in slope at ρ~0.7 correspond to the chosen value of n_{max} described in Section A. Standard deviations calculated from the generalised DPDE simulations are smaller than the plotted data points.

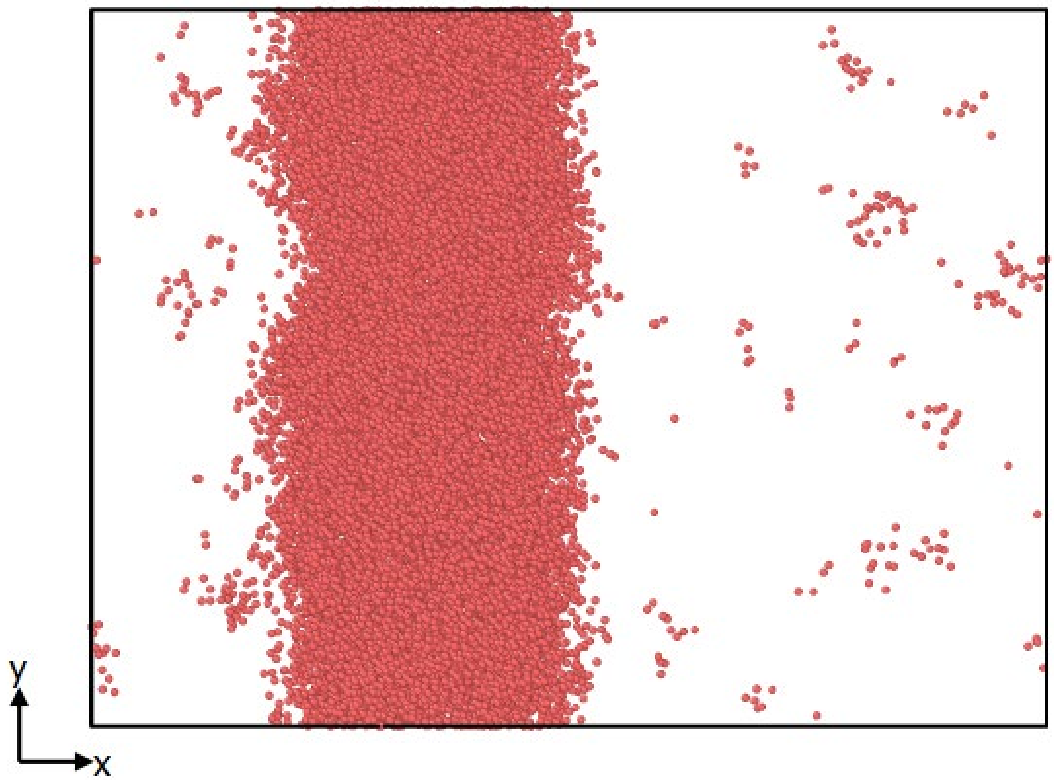


Figure S2: Configurational snapshot of vapour-liquid phase separation of the Lennard-Jones EOS [5] at T=1.0 and ρ=0.2 using the generalized DPDE method. Particles with a local density smaller than 0.1 are not displayed for visual clarity. The image was generated using the OVITO visualization software [6].

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