Electronic Supplementary Material (ESI) for Soft Matter.
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Finite-size scaling and thermodynamics of model supercooled liquids: Long-range concentration fluctuations and the role of attractive interactions

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S-I. RADIAL DISTRIBUTION FUNCTIONS

We compared the partial radial distribution functions of the KALJ and KAWCA systems at the same temperature and found that g_{aa} and g_{ab} for both systems are very similar as observed in earlier studies[1,2], and hence their corresponding Kirkwood-Buff integral (KBI) are also very similar. We used two methods for the KBI calculation: (i) the common single integral approximation using Eq. 3 of the main text and (ii) the fitting of density fluctuations Eq. 4 of the main text. The largest difference between the two system is observed in g_{bb} and G_{bb}, especially for low temperatures. In Figure S1 we present the RDFs for the lowest temperature case $T = 0.45\epsilon/k_{\mathrm{B}}$, following the same convention of Figure 1 a,b of the main text. We plot the RDFs for both systems on the left hand side and the corresponding KBI $G^R_{\alpha\beta}$ on the right hand side. The solid lines on the right hand side represent KBIs as a function of R obtained using Equation 3 of the main text. The corresponding limits $R \to \infty$ (thermodynamic limit) using Equation 4 of the main text are presented as dashed lines.

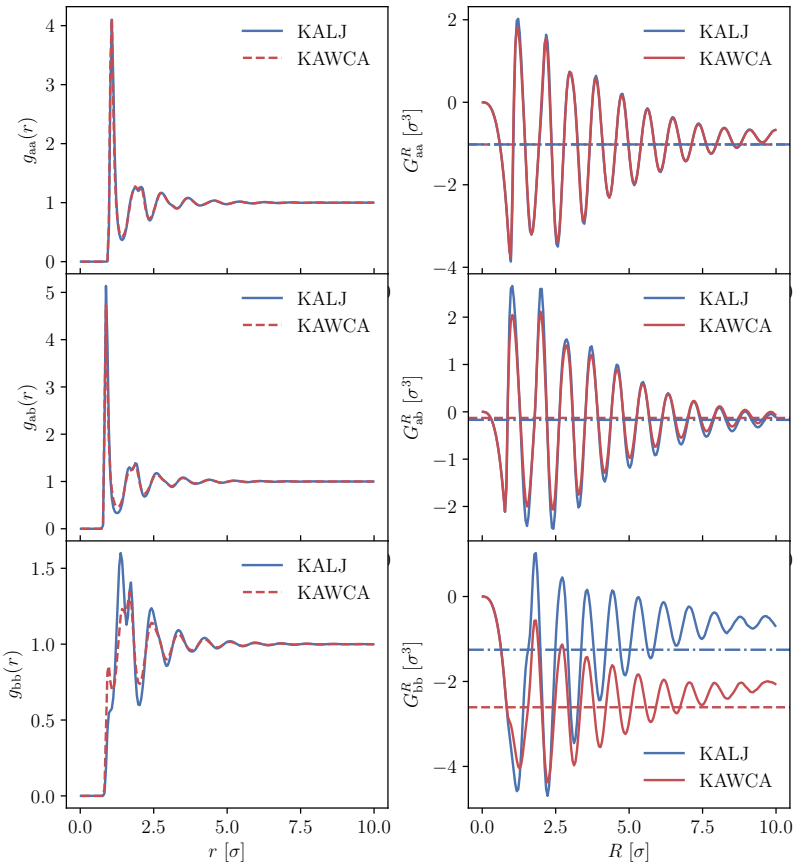

FIG. S1. Radial distribution functions g_{aa}, g_{bb} and g_{ab} and Kirkwood-Buff integrals G^R_{aa}, G^R_{bb} and G^R_{ab} for the KALJ and KAWCA systems at $T = 0.45\epsilon/k_{\mathrm{B}}$. The largest difference between the two systems is for the b − b components.

S-II. LONG-RANGE FLUCTUATION: LIMITING VALUES OF $S_{\rho\rho}$ FROM BT STRUCTURE FACTORS AND KBI

We plot the $S_{\rho\rho}$ at the lowest k of our simulation from BT structure factor using Equation 8 of the main text. The solid symbols in Figure S2 represent the data from BT structure factor. The open symbols are the limiting values using KBI (Equation 10 of the main text). Both the methods confirm that the $S_{\rho\rho}$ increases upon decreasing the temperature for KALJ system. The change in behaviour for the KALJ system is observed near the onset temperature of glassy dynamics $T = 1.00$. The qualitative result remains the same if we use any value of k upto $k < 1.5$. Note that, as stated in the main text, the low k limit of Equation 8 of the main text includes finite-size effect, thus we verified the result with a robust KBI framework.

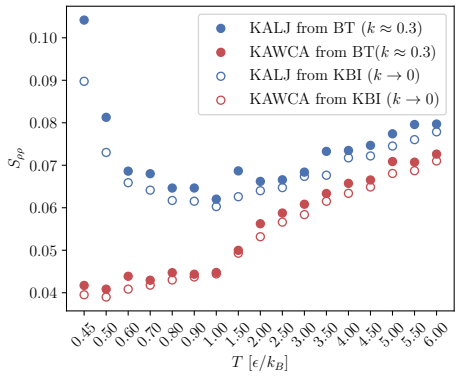

FIG. S2. The $S_{\rho\rho}$ at the lowest k from BT structure factor using Equation 7 and 8 of the main text(solid symbol), and the limiting value of $S_{\rho\rho}$ from KBI using Equation 10 of the main text(open symbol). Both the methods predict similar behaviour, confirming that the growth in the low k limit of $S_{\rho\rho}$ starts near the onset temperature $T = 1.00$ of glassy dynamics for the KALJ system, which is far off from the liquid-gas coexistence line.

S-III. AVERAGE PRESSURE

We compute the average virial pressure for KALJ and KAWCA systems. Our result suggests that the pressure values remain positive for all the state points in our simulation at $\rho = 1.2$. As expected, the KAWCA system has high pressure compared to the attractive counterpart. The pressure values for these models been reported earlier in Ref-3, stating that the state points are outside the gas-liquid coexistence curve at $\rho = 1.2$.

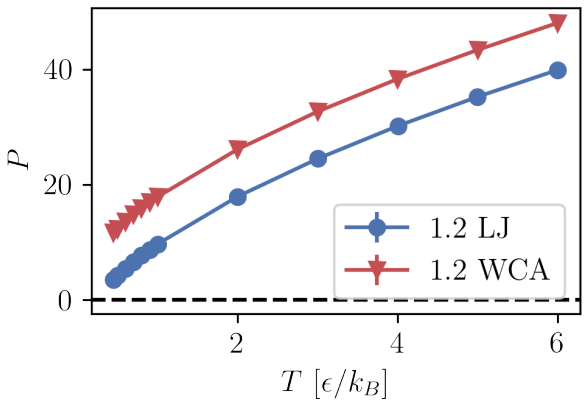

FIG. S3. Virial part of the pressure(P) with error bars for KALJ and KAWCA systems. The dotted horizontal line is drawn at $P = 0$ as a guide to the eye. The pressure remains positive fo all the simulated state points in our study.

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