Supplementary Material

# Interfacial Properties of Binary Mixtures of Simple Fluids and their Relation to the Phase Diagram

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The Supplementary Material for the publication *Interfacial Properties of Binary Mixtures of Simple Fluids and their Relation to the Phase Diagram* contains the following points:

- the identification of the two investigated mixtures A and B in the map of the enrichment, see Ref.[4], as a function of the mixture parameters $\xi$ and $\varepsilon_2/\varepsilon_1$ (cf. Fig. S1),
- the comparison of the difference of the component 2 bulk density in the $VL_1E$ region $\Delta\rho_2 = \rho_2^{L1} - \rho_2^V$ (cf. Fig. S2) and a discussion of these results,
- two exemplary screenshots of the MD simulation of the mixtures A and B (cf. Fig. S3),
- the plots of the MD $VL_1$ density profiles from both investigated mixtures A and B at all five temperatures (cf. Fig. S4 and Fig. S5),
- the plots of the DGT $VL_1$ density profiles for mixture B at all temperatures (cf. Fig. S6),
- the $p, T$ diagram of mixture A; including the state points whose density profiles intersect in two invariant points (see Appendix of the main body of the paper for a discussion),
- and the numeric values for the computed phase equilibria and interfacial properties for both MD and DGT (Table S1-S10 for the phase equilibria and Tables S11-S20 for the interfacial properties).

## DISCUSSION OF THE RESULTS FOR THE DENSITY DIFFERENCE $\Delta\rho_2$

We have shown in an earlier work[4], that the difference of the component 2 bulk density in a $VLE$ $\Delta\rho_2 = \rho_2^L - \rho_2^V$ is directly related to the arrangement of the surface excess at the vapour-liquid interface and thereby to the enrichment of the low-boiling component 2 at the interface. It was shown in Ref.[4] that for $\Delta\rho_2 = 0$, all adsorption at a vapour-liquid interface contributes to the enrichment[4]. Fig. S2 shows $\Delta\rho_2$ in the $VL_1E$ region for the mixtures A and B. As for the phase diagrams shown in the main body of the paper, the agreement between the computer experiment and the theory is almost perfect for the mixture B. But significant deviations are observed in the case of the mixture A. The PeTS

EOS predicts $\Delta\rho_2 < 0$ for all temperatures in the entire composition range, whereas the molecular simulations predict small values $\Delta\rho_2 > 0$ for most state points. For mixture A, the function $\Delta\rho_2(x_2^{L1})$ is very different from that for mixture B. For mixtures B, $\Delta\rho_2(x_2^{L1})$ shows an almost linear increase. The slope of $\Delta\rho_2(x_2^{L1})$ decreases with increasing temperature, which is simply due to the decreasing total density difference between both phases with increasing temperature.

In contrast, in mixture A $\Delta\rho_2(x_2^{L1})$ has a highly non-linear behaviour. $\Delta\rho_2$ remains very close to zero at highly diluted mixtures ($x_2^{L1} \to 0$) and then decreases with further increasing concentration $x_2^{L1}$. This decay increases with decreasing temperature. Close to the $VL_1L_2E$ line, $\Delta\rho_2$ drops almost vertically. The PeTS EOS predicts $\Delta\rho_2$ to exhibit a minimum at the three highest temperatures ($T = 0.77\,\varepsilon k_\mathrm{B}^{-1},\ 0.825\,\varepsilon k_\mathrm{B}^{-1},\ 0.88\,\varepsilon k_\mathrm{B}^{-1}$). By definition of the critical point, the densities and compositions of both phases must equalize upon approaching the very, i.e. $\Delta\rho_2 \to 0$ at the critical point, which is verified from the EOS results.

The poor agreement between MD and the EOS in the case of mixture A is likely due to the deviations observed in the bubble line in the phase diagrams at elevated pressures. Only the convex slope of $\Delta\rho_2(x_2^{L1})$ is captured consistently from both methods.

## SIMULATION SCREENSHOTS

Fig. S3 shows screenshots from the molecular dynamics simulation from mixture A and B at $T = 0.66\,\varepsilon k_\mathrm{B}^{-1}$ during the production phase of the simulation. The liquid phase composition is similar in both simulations – just above 10% mole fraction of the low-boiling component. It is evident that the vapour-liquid interface in the case of the asymmetric mixture A shows a strong surface excess of the low-boiling component 2 at the interface which is not present in the mixture B.

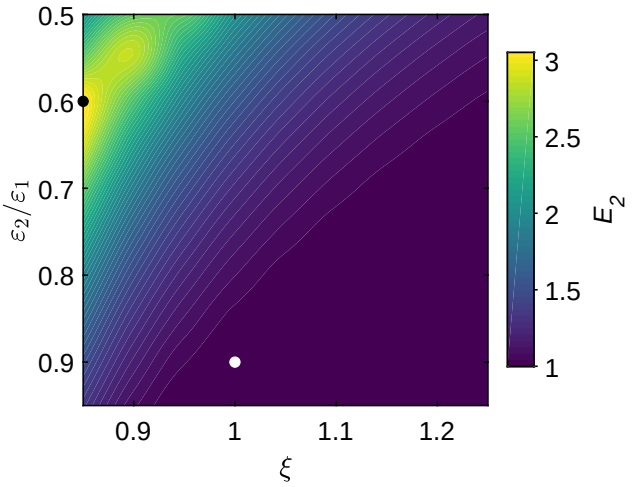



FIG. S1. Interfacial enrichment of the low-boiling component $E_2$ at vapour-liquid interfaces in binary LJTS mixtures (data from Ref.[4]) as a function of the binary interaction parameter $\xi$ and the ratio of dispersion energies $\varepsilon_2/\varepsilon_1$ at constant temperature ($T = 0.77\,\varepsilon k_\mathrm{B}^{-1}$) and liquid phase composition ($x_2^{L1} = 0.05\,\mathrm{mol\,mol^{-1}}$). The black and the white dot indicates the mixture A and B, that were investigated in the present work.

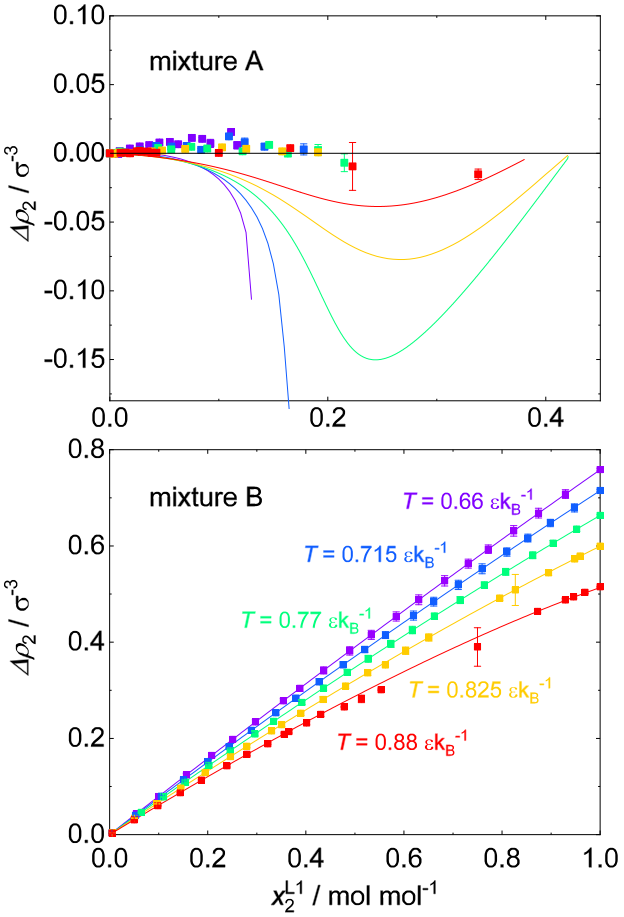



FIG. S2. Difference of the number density of the component 2 $\Delta\rho_2 = \rho_2^{L1} - \rho_2^V$ between both bulk phases. The temperatures in the top and bottom plot are colour-coded using the same scale. Symbols are MD results and lines are the PeTS EOS.

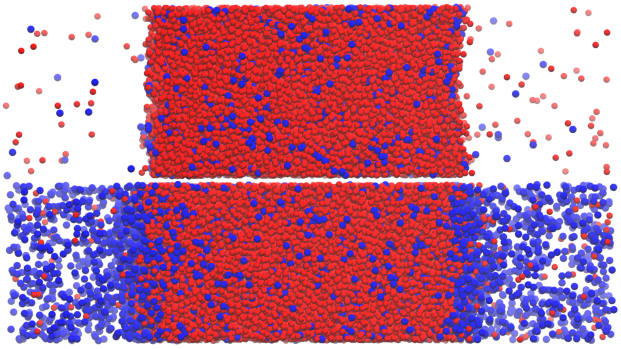


FIG. S3. Screenshots from the molecular simulations of mixture B (top) and mixture A (bottom) at $T = 0.66\,\varepsilon k_B^{-1}$. The liquid phase compositions are $x_2^{L1} = 0.11$ mol mol$^{-1}$ (top) and $x_2^{L1} = 0.12$ mol mol$^{-1}$ (bottom). The high-boiling component 1 is indicated red, the low-boiling component 2 blue.

FIG. S4. Density profiles of component 1 (dashed lines) and component 2 (full lines) for the mixture B obtained by MD. Results for all studied temperatures. The colour-code indicates the liquid phase composition.

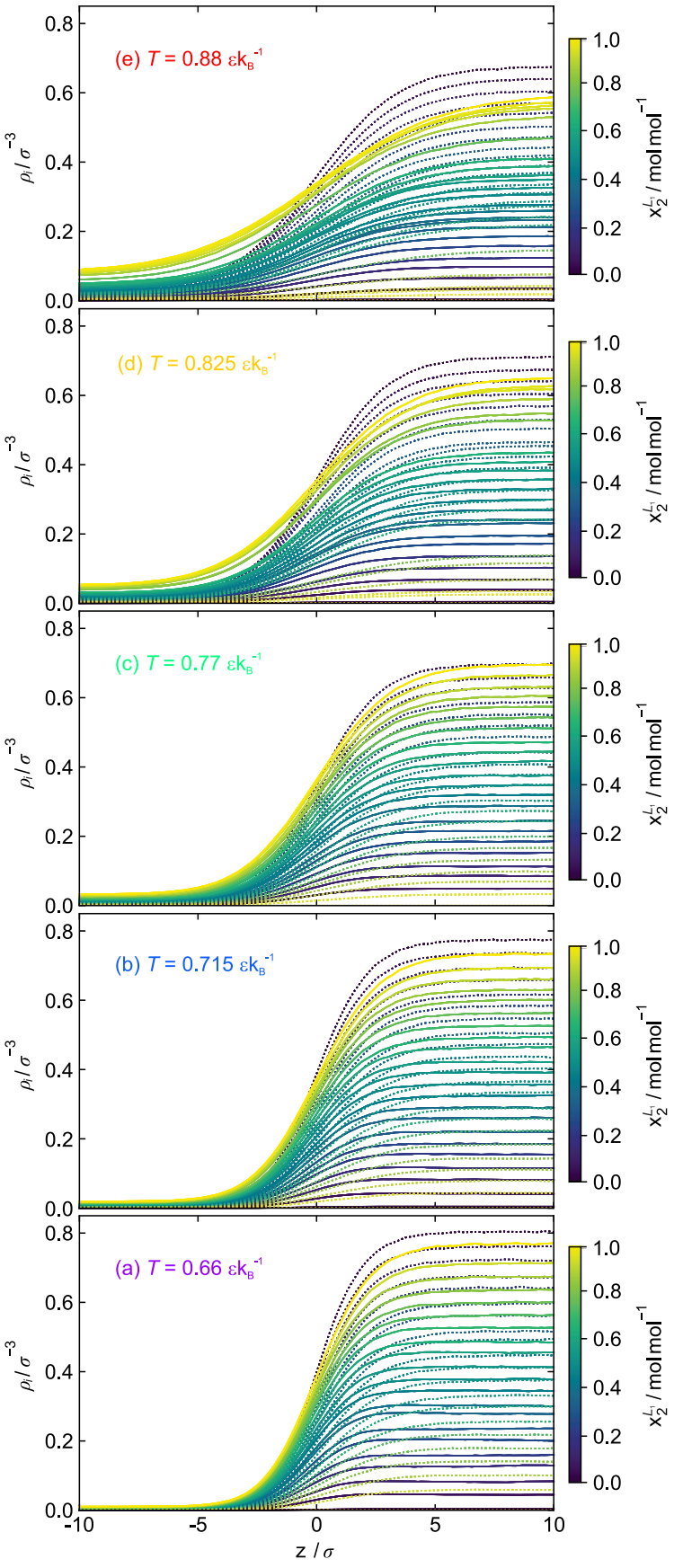


FIG. S5. $VL_1$ density profiles of component 1 (dashed lines) and component 2 (full lines) for the mixture A obtained by MD. Results for all studied temperatures. The colour-code indicates the liquid phase composition. Red squares indicate invariant intersection points of density profiles.

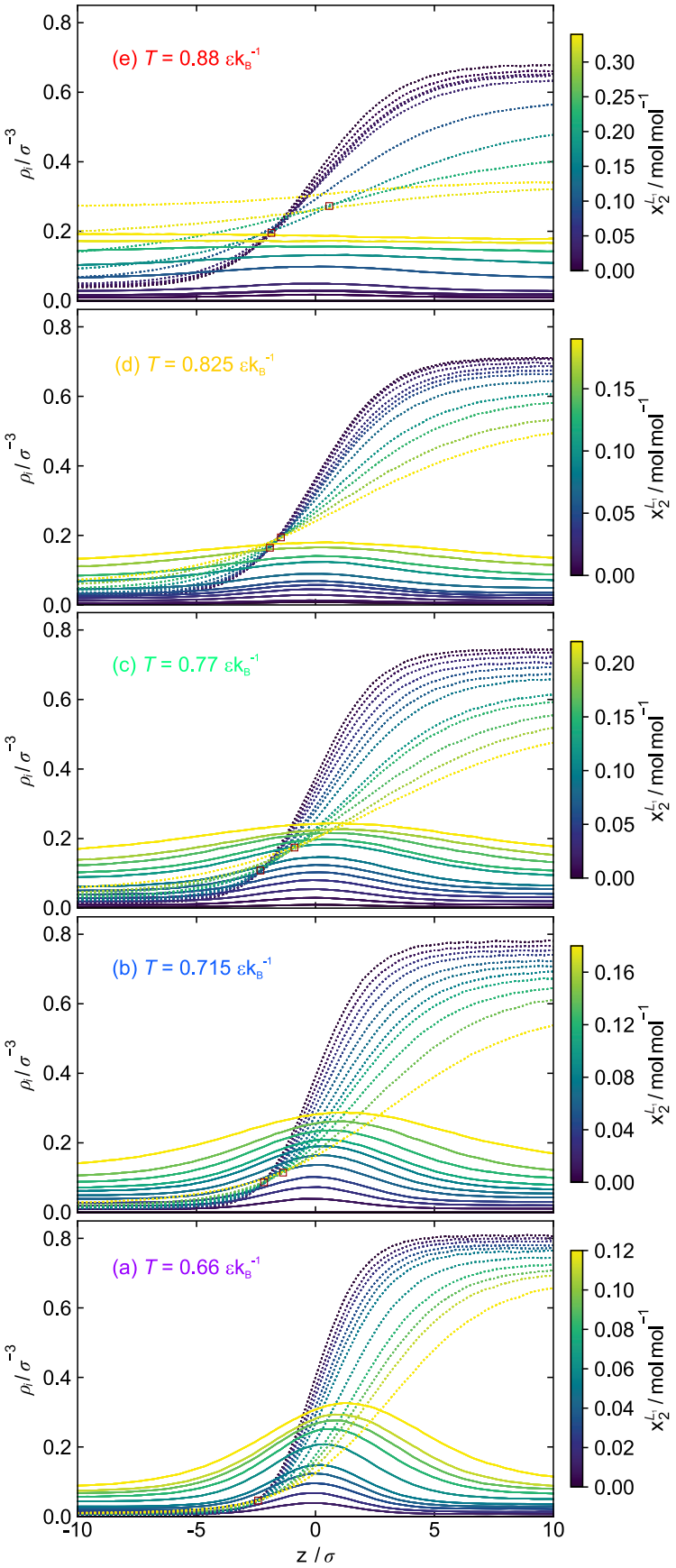
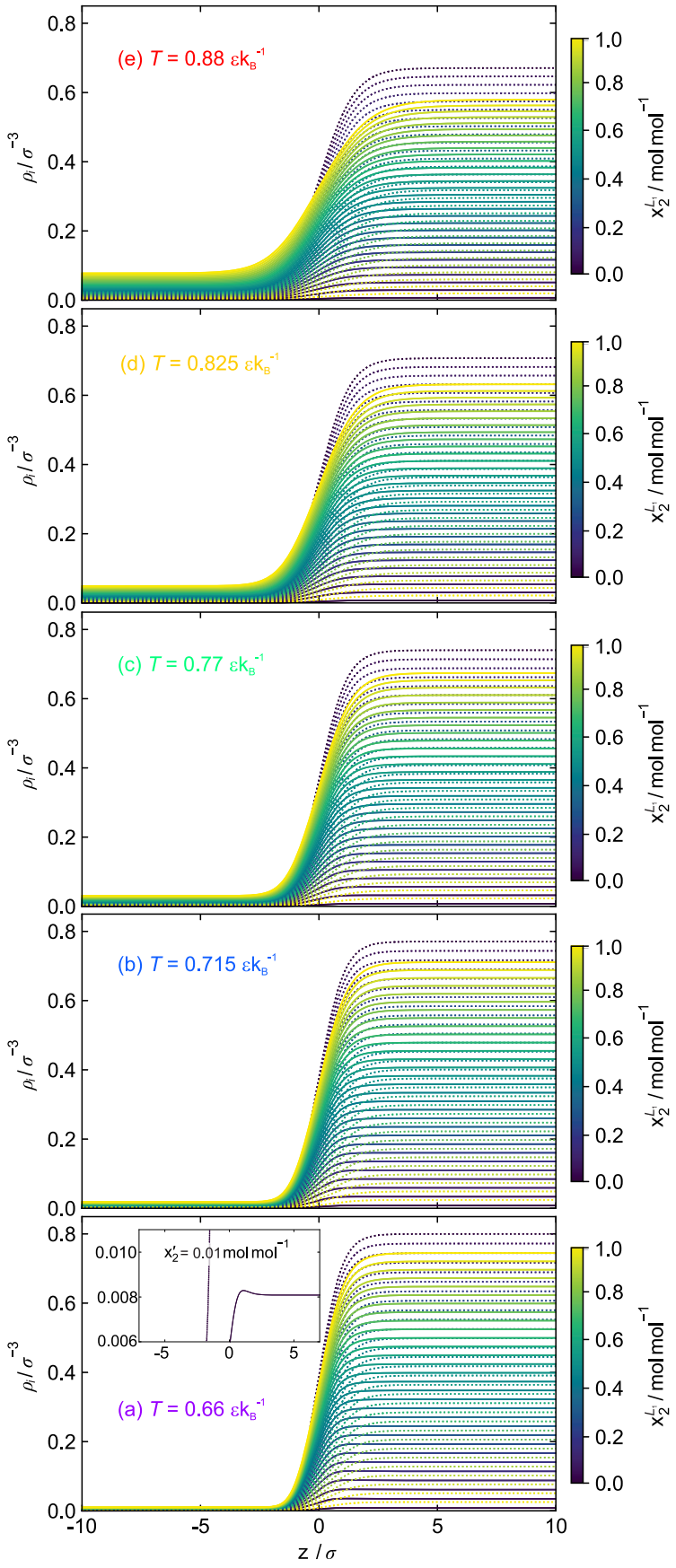


FIG. S6. Density profiles of component 1 (dashed lines) and component 2 (full lines) for the mixture B obtained by DGT. Results for all studied temperatures. The colour-code indicates the liquid phase composition.



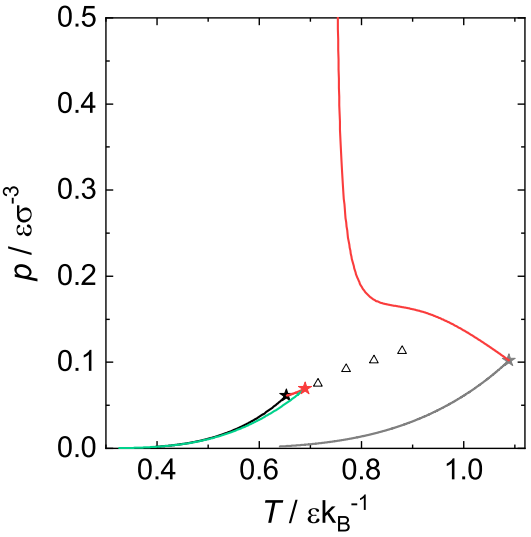



FIG. S7. Pressure-temperature diagram with characteristic curves for the mixture A – including the transition state points observed for the invariant intersection points. Pure component vapour pressure curves and critical points are the grey line and star (high-boiling component 1) and black line and star (low-boiling component 2). Red lines are critical lines of the mixture; the red star is the upper critical end point of mixture A (UCEP). The green line depicts the $VL_1L_2E$ three-phase line. The open triangles indicate the state points $p^*$ of which density profiles intersect multiple invariant points for a given temperature (see Appendix of the main body of the paper for a discussion).

TABLE S1. MD and EOS results for the bulk properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.66\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^V$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^V$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0030(1) | 0.0029 | 0.00 | 0.0000 | 0.808(4) | 0.8084 | 0.0047(3) | 0.0046 |
| 0.005(1) | 0.0030(1) | 0.0029 | 0.01(1) | 0.0109 | 0.808(4) | 0.8082 | 0.0047(3) | 0.0046 |
| 0.055(1) | 0.0030(1) | 0.0031 | 0.11(1) | 0.1110 | 0.806(8) | 0.8065 | 0.0048(4) | 0.0049 |
| 0.100(1) | 0.0033(1) | 0.0033 | 0.20(2) | 0.1928 | 0.804(9) | 0.8049 | 0.0052(5) | 0.0052 |
| 0.157(2) | 0.0034(2) | 0.0035 | 0.27(2) | 0.2846 | 0.80(1) | 0.8028 | 0.0054(5) | 0.0055 |
| 0.208(1) | 0.0036(1) | 0.0036 | 0.35(2) | 0.3593 | 0.80(2) | 0.8009 | 0.0058(5) | 0.0058 |
| 0.251(1) | 0.0038(1) | 0.0038 | 0.42(2) | 0.4160 | 0.80(1) | 0.7993 | 0.0061(5) | 0.0061 |
| 0.297(1) | 0.0039(2) | 0.0039 | 0.47(2) | 0.4727 | 0.80(1) | 0.7976 | 0.0063(5) | 0.0063 |
| 0.354(1) | 0.0042(2) | 0.0041 | 0.53(2) | 0.5364 | 0.80(1) | 0.7954 | 0.0067(6) | 0.0067 |
| 0.388(2) | 0.0043(1) | 0.0043 | 0.58(2) | 0.5713 | 0.79(1) | 0.7941 | 0.0070(6) | 0.0068 |
| 0.437(2) | 0.0045(1) | 0.0044 | 0.62(2) | 0.6187 | 0.79(1) | 0.7922 | 0.0072(6) | 0.0071 |
| 0.490(1) | 0.0047(1) | 0.0046 | 0.66(2) | 0.6667 | 0.79(2) | 0.7902 | 0.0075(6) | 0.0074 |
| 0.533(1) | 0.0047(2) | 0.0048 | 0.69(3) | 0.7036 | 0.79(2) | 0.7884 | 0.0076(6) | 0.0077 |
| 0.584(1) | 0.0050(1) | 0.0049 | 0.75(2) | 0.7441 | 0.79(2) | 0.7864 | 0.0081(6) | 0.0080 |
| 0.630(2) | 0.0051(1) | 0.0051 | 0.77(1) | 0.7784 | 0.78(1) | 0.7845 | 0.0083(6) | 0.0083 |
| 0.683(1) | 0.0053(2) | 0.0053 | 0.82(1) | 0.8155 | 0.78(1) | 0.7824 | 0.0086(6) | 0.0086 |
| 0.731(1) | 0.0055(2) | 0.0054 | 0.84(1) | 0.8474 | 0.78(1) | 0.7804 | 0.0090(6) | 0.0089 |
| 0.772(1) | 0.0056(2) | 0.0056 | 0.88(1) | 0.8732 | 0.78(1) | 0.7787 | 0.0091(6) | 0.0091 |
| 0.824(2) | 0.0059(2) | 0.0058 | 0.90(1) | 0.9046 | 0.78(1) | 0.7765 | 0.0096(6) | 0.0094 |
| 0.874(1) | 0.0059(1) | 0.0059 | 0.94(1) | 0.9336 | 0.77(1) | 0.7743 | 0.0097(6) | 0.0097 |
| 0.929(1) | 0.0062(2) | 0.0061 | 0.96(1) | 0.9635 | 0.77(1) | 0.7719 | 0.0102(8) | 0.0101 |
| 1.00 | 0.0064(2) | 0.0063 | 1.00 | 1.0000 | 0.769(3) | 0.7692 | 0.0106(5) | 0.0104 |

TABLE S2. MD and EOS results for the bulk properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.715\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0058(3) | 0.0058 | 0.00 | 0.0000 | 0.779(3) | 0.7789 | 0.0087(4) | 0.0087 |
| 0.0049(2) | 0.0057(2) | 0.0058 | 0.011(3) | 0.0097 | 0.779(4) | 0.7787 | 0.0086(5) | 0.0087 |
| 0.053(1) | 0.0061(2) | 0.0061 | 0.10(1) | 0.0988 | 0.777(7) | 0.7768 | 0.0091(6) | 0.0092 |
| 0.105(2) | 0.0064(2) | 0.0064 | 0.19(1) | 0.1872 | 0.775(8) | 0.7747 | 0.0097(6) | 0.0097 |
| 0.151(1) | 0.0067(2) | 0.0067 | 0.26(1) | 0.2567 | 0.773(10) | 0.7728 | 0.0101(7) | 0.0101 |
| 0.200(1) | 0.0070(2) | 0.0070 | 0.34(2) | 0.3273 | 0.771(12) | 0.7708 | 0.0107(7) | 0.0106 |
| 0.243(1) | 0.0073(2) | 0.0072 | 0.39(1) | 0.3836 | 0.769(11) | 0.7690 | 0.0112(7) | 0.0110 |
| 0.289(2) | 0.0075(2) | 0.0075 | 0.44(2) | 0.4392 | 0.767(12) | 0.7671 | 0.0115(7) | 0.0115 |
| 0.339(1) | 0.0079(2) | 0.0078 | 0.49(2) | 0.4957 | 0.765(12) | 0.7649 | 0.0121(7) | 0.0120 |
| 0.380(2) | 0.0081(2) | 0.0081 | 0.53(2) | 0.5394 | 0.764(14) | 0.7632 | 0.0125(8) | 0.0124 |
| 0.427(2) | 0.0083(2) | 0.0083 | 0.58(2) | 0.5866 | 0.761(12) | 0.7611 | 0.0128(8) | 0.0128 |
| 0.476(1) | 0.0087(2) | 0.0086 | 0.62(1) | 0.6326 | 0.759(12) | 0.7589 | 0.0133(8) | 0.0133 |
| 0.520(1) | 0.0088(2) | 0.0089 | 0.68(1) | 0.6718 | 0.757(12) | 0.7570 | 0.0136(8) | 0.0138 |
| 0.562(1) | 0.0092(2) | 0.0092 | 0.71(2) | 0.7075 | 0.755(12) | 0.7550 | 0.0143(8) | 0.0142 |
| 0.619(2) | 0.0095(2) | 0.0095 | 0.75(1) | 0.7530 | 0.753(14) | 0.7524 | 0.0147(8) | 0.0148 |
| 0.661(3) | 0.0099(3) | 0.0097 | 0.78(2) | 0.7843 | 0.751(12) | 0.7505 | 0.0154(8) | 0.0152 |
| 0.711(1) | 0.0101(2) | 0.0100 | 0.82(1) | 0.8204 | 0.749(13) | 0.7481 | 0.0159(8) | 0.0157 |
| 0.759(2) | 0.0106(3) | 0.0103 | 0.85(1) | 0.8538 | 0.746(13) | 0.7458 | 0.0166(8) | 0.0162 |
| 0.810(1) | 0.0109(2) | 0.0106 | 0.88(1) | 0.8870 | 0.744(10) | 0.7433 | 0.0171(8) | 0.0168 |
| 0.853(1) | 0.0110(2) | 0.0109 | 0.91(1) | 0.9141 | 0.741(10) | 0.7412 | 0.0174(12) | 0.0172 |
| 0.899(1) | 0.0112(3) | 0.0112 | 0.94(1) | 0.9418 | 0.739(8) | 0.7389 | 0.0178(8) | 0.0177 |
| 0.948(1) | 0.0118(2) | 0.0115 | 0.97(1) | 0.9705 | 0.736(11) | 0.7364 | 0.019(3) | 0.0182 |
| 1.000 | 0.0119(2) | 0.0117 | 1.00 | 1.0000 | 0.734(3) | 0.7342 | 0.0190(7) | 0.0187 |

TABLE S3. MD and EOS results for the bulk properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.77\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0105(3) | 0.0104 | 0.00 | 0.0000 | 0.749(3) | 0.7480 | 0.0152(6) | 0.0150 |
| 0.064(2) | 0.0110(3) | 0.0110 | 0.11(1) | 0.1102 | 0.746(7) | 0.7451 | 0.0160(7) | 0.0160 |
| 0.110(1) | 0.0116(1) | 0.0114 | 0.19(2) | 0.1812 | 0.744(9) | 0.7430 | 0.0169(8) | 0.0167 |
| 0.154(2) | 0.0119(3) | 0.0118 | 0.26(1) | 0.2450 | 0.741(9) | 0.7410 | 0.0175(10) | 0.0174 |
| 0.202(2) | 0.0124(2) | 0.0123 | 0.31(1) | 0.3111 | 0.739(9) | 0.7387 | 0.0183(9) | 0.0181 |
| 0.246(2) | 0.0129(4) | 0.0127 | 0.37(2) | 0.3662 | 0.74(1) | 0.7366 | 0.0191(9) | 0.0188 |
| 0.296(2) | 0.0134(4) | 0.0132 | 0.42(1) | 0.4263 | 0.73(1) | 0.7342 | 0.0199(9) | 0.0196 |
| 0.334(1) | 0.0137(3) | 0.0136 | 0.47(2) | 0.4691 | 0.73(1) | 0.7323 | 0.020(1) | 0.0202 |
| 0.392(2) | 0.0143(3) | 0.0141 | 0.53(1) | 0.5303 | 0.73(1) | 0.7294 | 0.021(1) | 0.0211 |
| 0.436(1) | 0.0146(3) | 0.0145 | 0.57(1) | 0.5742 | 0.73(1) | 0.7271 | 0.022(1) | 0.0218 |
| 0.484(2) | 0.0150(4) | 0.0150 | 0.62(1) | 0.6199 | 0.72(1) | 0.7247 | 0.023(1) | 0.0226 |
| 0.527(2) | 0.0156(4) | 0.0154 | 0.65(1) | 0.6584 | 0.723(13) | 0.7224 | 0.024(1) | 0.0233 |
| 0.574(2) | 0.0158(3) | 0.0159 | 0.69(1) | 0.6985 | 0.720(12) | 0.7199 | 0.024(1) | 0.0241 |
| 0.617(2) | 0.0163(4) | 0.0163 | 0.73(1) | 0.7345 | 0.718(12) | 0.7176 | 0.025(1) | 0.0248 |
| 0.662(1) | 0.0168(2) | 0.0167 | 0.77(1) | 0.7700 | 0.715(10) | 0.7151 | 0.026(1) | 0.0256 |
| 0.714(2) | 0.0173(3) | 0.0172 | 0.81(1) | 0.8097 | 0.713(10) | 0.7122 | 0.027(1) | 0.0265 |
| 0.764(1) | 0.0179(3) | 0.0177 | 0.85(1) | 0.8457 | 0.710(9) | 0.7093 | 0.028(1) | 0.0273 |
| 0.808(2) | 0.0183(4) | 0.0181 | 0.88(1) | 0.8761 | 0.707(8) | 0.7068 | 0.028(1) | 0.0281 |
| 0.863(1) | 0.0188(3) | 0.0187 | 0.91(1) | 0.9133 | 0.704(9) | 0.7035 | 0.029(1) | 0.0291 |
| 0.904(1) | 0.0192(2) | 0.0191 | 0.94(1) | 0.9404 | 0.701(7) | 0.7010 | 0.030(1) | 0.0298 |
| 0.952(1) | 0.0197(3) | 0.0196 | 0.973(4) | 0.9706 | 0.698(6) | 0.6981 | 0.031(1) | 0.0307 |
| 1.00 | 0.0203(3) | 0.0199 | 1.00 | 1.0000 | 0.695(3) | 0.6957 | 0.0320(9) | 0.0314 |

TABLE S4. MD and EOS results for the bulk properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.825\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0175(3) | 0.0173 | 0.000 | 0.0000 | 0.716(3) | 0.7149 | 0.025(1) | 0.0246 |
| 0.0055(3) | 0.0176(3) | 0.0173 | 0.010(3) | 0.0092 | 0.715(4) | 0.7146 | 0.025(1) | 0.0247 |
| 0.051(1) | 0.0181(4) | 0.0180 | 0.082(8) | 0.0820 | 0.713(6) | 0.7122 | 0.026(1) | 0.0257 |
| 0.096(1) | 0.0188(3) | 0.0186 | 0.151(7) | 0.1500 | 0.710(7) | 0.7098 | 0.027(1) | 0.0268 |
| 0.145(1) | 0.0195(2) | 0.0193 | 0.213(9) | 0.2189 | 0.707(9) | 0.7072 | 0.028(1) | 0.0279 |
| 0.195(2) | 0.0201(3) | 0.0200 | 0.29(1) | 0.2844 | 0.705(9) | 0.7044 | 0.029(1) | 0.0291 |
| 0.246(1) | 0.0210(2) | 0.0207 | 0.35(1) | 0.3485 | 0.702(9) | 0.7015 | 0.031(1) | 0.0303 |
| 0.280(1) | 0.0214(4) | 0.0212 | 0.38(1) | 0.3878 | 0.700(10) | 0.6996 | 0.031(1) | 0.0311 |
| 0.330(1) | 0.0222(4) | 0.0219 | 0.44(1) | 0.4445 | 0.697(9) | 0.6966 | 0.033(1) | 0.0323 |
| 0.351(2) | 0.0224(3) | 0.0222 | 0.47(1) | 0.4668 | 0.696(10) | 0.6954 | 0.033(1) | 0.0328 |
| 0.390(2) | 0.0231(3) | 0.0228 | 0.51(1) | 0.5078 | 0.69(1) | 0.6931 | 0.035(3) | 0.0338 |
| 0.435(2) | 0.0236(3) | 0.0234 | 0.55(1) | 0.5531 | 0.69(1) | 0.6903 | 0.035(1) | 0.0350 |
| 0.481(2) | 0.0243(3) | 0.0241 | 0.60(1) | 0.5974 | 0.69(1) | 0.6875 | 0.036(2) | 0.0361 |
| 0.526(2) | 0.0248(3) | 0.0247 | 0.63(1) | 0.6389 | 0.68(1) | 0.6846 | 0.037(1) | 0.0373 |
| 0.562(4) | 0.0253(7) | 0.0253 | 0.66(1) | 0.6709 | 0.68(1) | 0.6823 | 0.043(2) | 0.0383 |
| 0.604(2) | 0.0259(3) | 0.0259 | 0.71(1) | 0.7065 | 0.68(1) | 0.6796 | 0.040(2) | 0.0394 |
| 0.651(1) | 0.0267(4) | 0.0266 | 0.74(1) | 0.7458 | 0.68(1) | 0.6764 | 0.041(2) | 0.0407 |
| 0.795(2) | 0.0289(3) | 0.0287 | 0.86(1) | 0.8577 | 0.667(7) | 0.6663 | 0.045(1) | 0.0448 |
| 0.83(4) | 0.0294(5) | 0.0292 | 0.88(3) | 0.8808 | 0.664(8) | 0.6640 | 0.046(2) | 0.0457 |
| 0.895(1) | 0.0304(4) | 0.0302 | 0.928(4) | 0.9289 | 0.658(8) | 0.6589 | 0.049(3) | 0.0478 |
| 0.948(1) | 0.0313(4) | 0.0310 | 0.965(3) | 0.9653 | 0.655(5) | 0.6547 | 0.050(1) | 0.0494 |
| 0.959(1) | 0.0313(4) | 0.0312 | 0.973(3) | 0.9729 | 0.654(5) | 0.6538 | 0.050(1) | 0.0498 |
| 1.000 | 0.0320(4) | 0.0317 | 1.0000 | 0.9934 | 0.651(3) | 0.6514 | 0.052(1) | 0.0508 |

TABLE S5. MD and EOS results for the bulk properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.88\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0271(4) | 0.0270 | 0.000 | 0.0000 | 0.679(3) | 0.6782 | 0.039(1) | 0.0384 |
| 0.0051(3) | 0.0272(3) | 0.0271 | 0.007(2) | 0.0080 | 0.679(3) | 0.6778 | 0.039(1) | 0.0386 |
| 0.050(1) | 0.0281(3) | 0.0280 | 0.075(4) | 0.0752 | 0.675(6) | 0.6750 | 0.041(2) | 0.0401 |
| 0.098(1) | 0.0291(3) | 0.0289 | 0.14(1) | 0.1434 | 0.672(6) | 0.6719 | 0.042(1) | 0.0418 |
| 0.143(1) | 0.0299(3) | 0.0298 | 0.21(1) | 0.2038 | 0.669(7) | 0.6689 | 0.044(2) | 0.0434 |
| 0.187(1) | 0.0309(3) | 0.0307 | 0.25(1) | 0.2601 | 0.666(8) | 0.6659 | 0.045(1) | 0.0449 |
| 0.240(2) | 0.0321(3) | 0.0318 | 0.32(1) | 0.3235 | 0.663(8) | 0.6623 | 0.047(1) | 0.0468 |
| 0.280(2) | 0.0328(3) | 0.0326 | 0.37(1) | 0.3697 | 0.660(9) | 0.6595 | 0.049(2) | 0.0483 |
| 0.322(2) | 0.0335(4) | 0.0334 | 0.42(1) | 0.4161 | 0.657(16) | 0.6565 | 0.050(11) | 0.0499 |
| 0.365(2) | 0.0347(3) | 0.0343 | 0.47(1) | 0.4626 | 0.653(8) | 0.6540 | 0.052(2) | 0.0512 |
| 0.356(2) | 0.0344(5) | 0.0341 | 0.46(1) | 0.4525 | 0.653(11) | 0.6532 | 0.051(4) | 0.0516 |
| 0.401(3) | 0.0354(4) | 0.0350 | 0.50(1) | 0.4997 | 0.61(2) | 0.6505 | 0.053(9) | 0.0530 |
| 0.431(2) | 0.0357(5) | 0.0356 | 0.53(1) | 0.5292 | 0.65(1) | 0.6483 | 0.054(3) | 0.0542 |
| 0.478(4) | 0.0364(7) | 0.0366 | 0.56(1) | 0.5750 | 0.645(4) | 0.6446 | 0.057(3) | 0.0561 |
| 0.514(3) | 0.0371(10) | 0.0374 | 0.60(1) | 0.6092 | 0.643(4) | 0.6416 | 0.058(3) | 0.0576 |
| 0.552(4) | 0.0383(9) | 0.0381 | 0.63(1) | 0.6435 | 0.639(4) | 0.6386 | 0.059(4) | 0.0592 |
| 0.576(6) | 0.0386(15) | 0.0387 | 0.66(2) | 0.6654 | 0.637(14) | 0.6365 | 0.060(12) | 0.0602 |
| 0.622(5) | 0.0397(15) | 0.0396 | 0.71(2) | 0.7049 | 0.633(15) | 0.6326 | 0.062(13) | 0.0622 |
| 0.651(5) | 0.0407(13) | 0.0402 | 0.74(1) | 0.7303 | 0.631(13) | 0.6300 | 0.065(11) | 0.0636 |
| 0.873(1) | 0.0453(6) | 0.0450 | 0.909(4) | 0.9063 | 0.61(1) | 0.6201 | 0.076(2) | 0.0687 |
| 0.76(9) | 0.0428(4) | 0.0426 | 0.819(10) | 0.8187 | 0.621(15) | 0.6090 | 0.070(15) | 0.0744 |
| 0.929(1) | 0.0464(5) | 0.0463 | 0.949(4) | 0.9483 | 0.605(5) | 0.6030 | 0.078(2) | 0.0775 |
| 0.946(1) | 0.0469(5) | 0.0467 | 0.959(3) | 0.9606 | 0.603(4) | 0.6012 | 0.079(2) | 0.0784 |
| 0.969(1) | 0.0473(4) | 0.0472 | 0.977(2) | 0.9777 | 0.600(4) | 0.5986 | 0.080(2) | 0.0798 |
| 1.000 | 0.0478(7) | 0.0477 | 1.000 | 1.0000 | 0.597(3) | 0.5963 | 0.081(2) | 0.0810 |

TABLE S6. MD and EOS results for the bulk properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.66\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0030(1) | 0.0029 | 0.000 | 0.000 | 0.808(4) | 0.8084 | 0.0047(3) | 0.0046 |
| 0.009(1) | 0.0066(2) | 0.0074 | 0.534(16) | 0.5874 | 0.806(5) | 0.8070 | 0.0106(8) | 0.0108 |
| 0.0077(5) | 0.0072(2) | 0.0067 | 0.580(18) | 0.5503 | 0.807(5) | 0.8067 | 0.0115(8) | 0.0119 |
| 0.018(1) | 0.0101(2) | 0.0119 | 0.690(15) | 0.7337 | 0.804(6) | 0.8049 | 0.0164(9) | 0.0197 |
| 0.027(1) | 0.0135(3) | 0.0161 | 0.759(9) | 0.7953 | 0.803(7) | 0.8032 | 0.022(1) | 0.0274 |
| 0.036(1) | 0.0168(2) | 0.0201 | 0.803(8) | 0.8294 | 0.801(8) | 0.8015 | 0.029(1) | 0.0351 |
| 0.045(1) | 0.0199(3) | 0.0242 | 0.824(9) | 0.8527 | 0.799(9) | 0.7997 | 0.035(1) | 0.0438 |
| 0.055(2) | 0.0235(3) | 0.0283 | 0.848(8) | 0.8687 | 0.797(11) | 0.7978 | 0.042(1) | 0.0532 |
| 0.063(3) | 0.0271(5) | 0.0315 | 0.863(8) | 0.8777 | 0.796(11) | 0.7963 | 0.050(2) | 0.0609 |
| 0.075(2) | 0.0295(4) | 0.0366 | 0.872(6) | 0.8886 | 0.793(12) | 0.7938 | 0.056(2) | 0.0750 |
| 0.085(2) | 0.0329(4) | 0.0403 | 0.880(8) | 0.8941 | 0.791(14) | 0.7919 | 0.064(2) | 0.0865 |
| 0.092(5) | 0.0364(6) | 0.0432 | 0.891(6) | 0.8974 | 0.789(13) | 0.7903 | 0.074(2) | 0.0967 |
| 0.111(2) | 0.0387(3) | 0.0504 | 0.889(9) | 0.9019 | 0.785(20) | 0.7864 | 0.081(2) | 0.1295 |
| 0.117(4) | 0.0430(4) | 0.0525 | 0.901(5) | 0.9020 | 0.784(18) | 0.7853 | 0.095(3) | 0.1429 |

TABLE S7. MD and EOS results for the bulk properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.715\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0058(3) | 0.0058 | 0.000 | 0.000 | 0.779(3) | 0.7789 | 0.0087(4) | 0.0087 |
| 0.0096(4) | 0.0106(3) | 0.0110 | 0.426(10) | 0.4476 | 0.777(5) | 0.7769 | 0.0160(8) | 0.0167 |
| 0.0129(9) | 0.0125(3) | 0.0128 | 0.496(8) | 0.5176 | 0.777(5) | 0.7762 | 0.019(1) | 0.0196 |
| 0.027(1) | 0.0184(4) | 0.0203 | 0.651(10) | 0.6775 | 0.773(6) | 0.7732 | 0.029(1) | 0.0322 |
| 0.043(2) | 0.0238(4) | 0.0278 | 0.715(11) | 0.7498 | 0.770(8) | 0.7700 | 0.038(1) | 0.0462 |
| 0.056(2) | 0.0300(4) | 0.0344 | 0.764(8) | 0.7862 | 0.767(9) | 0.7671 | 0.050(2) | 0.0598 |
| 0.071(1) | 0.0355(4) | 0.0409 | 0.786(8) | 0.8092 | 0.76(1) | 0.7640 | 0.062(2) | 0.0746 |
| 0.084(3) | 0.0415(5) | 0.0467 | 0.809(5) | 0.8233 | 0.76(1) | 0.7611 | 0.076(2) | 0.0897 |
| 0.109(2) | 0.0454(6) | 0.0575 | 0.820(8) | 0.8384 | 0.75(1) | 0.7554 | 0.085(2) | 0.1244 |
| 0.124(6) | 0.0512(8) | 0.0635 | 0.831(8) | 0.8416 | 0.75(2) | 0.7521 | 0.102(3) | 0.1497 |
| 0.142(5) | 0.0568(9) | 0.0712 | 0.836(5) | 0.8396 | 0.74(2) | 0.7480 | 0.121(3) | 0.1959 |
| 0.178(8) | 0.0639(9) | 0.1066 | 0.840(6) | 0.7357 | 0.73(3) | 0.7429 | 0.152(7) | 0.5084 |

TABLE S8. MD and EOS results for the bulk properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.77\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0105(3) | 0.0104 | 0.000 | 0.000 | 0.749(3) | 0.7480 | 0.0152(6) | 0.0150 |
| 0.0045(3) | 0.0128(3) | 0.0130 | 0.164(6) | 0.1821 | 0.748(4) | 0.7470 | 0.0185(8) | 0.0189 |
| 0.0097(6) | 0.0151(3) | 0.0160 | 0.272(13) | 0.3193 | 0.746(5) | 0.7458 | 0.022(1) | 0.0234 |
| 0.0136(9) | 0.0178(3) | 0.0182 | 0.371(12) | 0.3914 | 0.745(5) | 0.7449 | 0.026(1) | 0.0268 |
| 0.0271(9) | 0.0242(3) | 0.0257 | 0.517(8) | 0.5446 | 0.742(6) | 0.7418 | 0.036(1) | 0.0389 |
| 0.044(1) | 0.0308(3) | 0.0349 | 0.607(9) | 0.6416 | 0.737(8) | 0.7378 | 0.047(2) | 0.0551 |
| 0.056(1) | 0.0371(4) | 0.0411 | 0.652(9) | 0.6810 | 0.734(8) | 0.7349 | 0.059(2) | 0.0670 |
| 0.075(2) | 0.0435(7) | 0.0501 | 0.696(9) | 0.7194 | 0.729(9) | 0.7305 | 0.071(2) | 0.0863 |
| 0.089(1) | 0.0499(4) | 0.0570 | 0.717(5) | 0.7387 | 0.73(1) | 0.7268 | 0.085(2) | 0.1031 |
| 0.122(3) | 0.0617(6) | 0.0715 | 0.747(6) | 0.7607 | 0.72(1) | 0.7186 | 0.115(3) | 0.1464 |
| 0.146(3) | 0.0666(6) | 0.0820 | 0.753(11) | 0.7641 | 0.71(1) | 0.7123 | 0.129(3) | 0.1889 |
| 0.163(4) | 0.0731(6) | 0.0894 | 0.767(7) | 0.7598 | 0.70(2) | 0.7078 | 0.150(4) | 0.2276 |
| 0.191(8) | 0.078(1) | 0.1025 | 0.763(9) | 0.7372 | 0.69(2) | 0.7008 | 0.170(5) | 0.3148 |
| 0.215(14) | 0.084(1) | 0.1165 | 0.757(15) | 0.6988 | 0.68(3) | 0.6953 | 0.204(8) | 0.4069 |

TABLE S9. MD and EOS results for the bulk properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.825\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0175(3) | 0.0173 | 0.000 | 0.000 | 0.716(3) | 0.7149 | 0.025(1) | 0.0246 |
| 0.0041(3) | 0.0200(4) | 0.0198 | 0.114(6) | 0.1091 | 0.714(3) | 0.7139 | 0.029(1) | 0.0282 |
| 0.0083(9) | 0.0219(4) | 0.0223 | 0.173(8) | 0.1953 | 0.713(4) | 0.7128 | 0.031(1) | 0.0319 |
| 0.0177(6) | 0.0275(4) | 0.0279 | 0.308(9) | 0.3301 | 0.711(5) | 0.7103 | 0.040(1) | 0.0404 |
| 0.029(2) | 0.0327(5) | 0.0343 | 0.40(1) | 0.4314 | 0.707(6) | 0.7074 | 0.048(2) | 0.0507 |
| 0.038(1) | 0.0371(5) | 0.0397 | 0.46(1) | 0.4906 | 0.704(6) | 0.7049 | 0.055(2) | 0.0598 |
| 0.048(1) | 0.0413(4) | 0.0449 | 0.51(1) | 0.5341 | 0.701(7) | 0.7023 | 0.062(2) | 0.0692 |
| 0.069(2) | 0.0501(5) | 0.0561 | 0.57(1) | 0.5971 | 0.695(9) | 0.6964 | 0.078(2) | 0.0910 |
| 0.103(2) | 0.0633(5) | 0.0727 | 0.62(1) | 0.6477 | 0.68(1) | 0.6868 | 0.106(3) | 0.1297 |
| 0.125(3) | 0.0713(6) | 0.0829 | 0.65(1) | 0.6629 | 0.68(1) | 0.6801 | 0.125(3) | 0.1589 |
| 0.158(6) | 0.082(1) | 0.0973 | 0.66(1) | 0.6686 | 0.66(1) | 0.6700 | 0.157(4) | 0.2098 |
| 0.191(5) | 0.090(1) | 0.1110 | 0.67(1) | 0.6585 | 0.65(2) | 0.6597 | 0.184(5) | 0.2701 |

TABLE S10. MD and EOS results for the bulk properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.88\,\varepsilon k_\mathrm{B}^{-1}$. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $p$ / $\varepsilon\sigma^{-3}$ | | $x_2^{V}$ / mol mol$^{-1}$ | | $\rho^{L1}$ / $\sigma^{-3}$ | | $\rho^{V}$ / $\sigma^{-3}$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | EOS | MD | EOS | MD | EOS | MD | EOS |
| 0.000 | 0.0271(4) | 0.0270 | 0.000 | 0.000 | 0.679(3) | 0.6782 | 0.039(1) | 0.0384 |
| 0.0098(6) | 0.0325(4) | 0.0331 | 0.134(5) | 0.1479 | 0.676(4) | 0.6752 | 0.046(1) | 0.0475 |
| 0.0143(5) | 0.0350(3) | 0.0359 | 0.181(6) | 0.1993 | 0.674(4) | 0.6739 | 0.050(2) | 0.0517 |
| 0.0185(6) | 0.0377(4) | 0.0384 | 0.227(6) | 0.2391 | 0.673(5) | 0.6726 | 0.055(2) | 0.0556 |
| 0.0248(9) | 0.0401(4) | 0.0421 | 0.259(8) | 0.2902 | 0.670(4) | 0.6707 | 0.058(2) | 0.0616 |
| 0.027(2) | 0.0411(7) | 0.0437 | 0.274(8) | 0.3088 | 0.669(6) | 0.6698 | 0.060(2) | 0.0642 |
| 0.029(1) | 0.0425(4) | 0.0445 | 0.293(6) | 0.3182 | 0.669(5) | 0.6694 | 0.062(2) | 0.0656 |
| 0.0345(1) | 0.0453(4) | 0.0479 | 0.326(6) | 0.3523 | 0.667(5) | 0.6676 | 0.067(2) | 0.0712 |
| 0.038(2) | 0.0480(6) | 0.0501 | 0.353(9) | 0.3726 | 0.666(6) | 0.6663 | 0.071(2) | 0.0751 |
| 0.043(1) | 0.0501(6) | 0.0525 | 0.372(7) | 0.3923 | 0.664(6) | 0.6650 | 0.075(2) | 0.0793 |
| 0.100(2) | 0.0756(6) | 0.0824 | 0.505(9) | 0.5279 | 0.642(9) | 0.6458 | 0.126(3) | 0.1423 |
| 0.166(6) | 0.0949(11) | 0.1116 | 0.55(1) | 0.5554 | 0.612(15) | 0.6214 | 0.177(5) | 0.2320 |
| 0.223(11) | 0.1126(7) | 0.1330 | 0.52(6) | 0.5338 | 0.583(17) | 0.5981 | 0.27(2) | 0.3196 |

TABLE S11. Interfacial properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.66\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\sigma^2\varepsilon^{-1}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.00 | 0.655(26) | 0.6632 | - | - | - | - | 2.06(5) | 1.94 |
| 0.005(1) | 0.649(18) | 0.6620 | 0.003(10) | 0.002 | 1.32(13) | 1.03 | 2.05(4) | 1.94 |
| 0.055(1) | 0.647(31) | 0.6506 | 0.032(20) | 0.019 | 1.11(5) | 1.02 | 2.07(4) | 1.96 |
| 0.100(1) | 0.627(25) | 0.6405 | 0.07(2) | 0.034 | 1.08(2) | 1.01 | 2.07(6) | 1.96 |
| 0.157(2) | 0.626(23) | 0.6280 | 0.09(5) | 0.052 | 1.06(2) | 1.01 | 2.12(5) | 1.98 |
| 0.208(1) | 0.612(30) | 0.6170 | -0.11(4) | 0.067 | 0.99(2) | 1.00 | 2.11(5) | 1.98 |
| 0.251(1) | 0.604(24) | 0.6080 | 0.07(3) | 0.080 | 1.04(1) | 1.00 | 2.16(4) | 2.00 |
| 0.297(1) | 0.590(19) | 0.5984 | 0.11(7) | 0.093 | 1.03(2) | 1.00 | 2.16(5) | 2.00 |
| 0.354(1) | 0.582(28) | 0.5869 | 0.08(4) | 0.109 | 1.02(1) | 1.00 | 2.19(3) | 2.02 |
| 0.388(2) | 0.568(24) | 0.5803 | 0.08(7) | 0.117 | 1.03(1) | 1.00 | 2.22(6) | 2.02 |
| 0.437(2) | 0.565(26) | 0.5708 | 0.16(8) | 0.130 | 1.02(1) | 1.00 | 2.23(3) | 2.04 |
| 0.490(1) | 0.559(20) | 0.5606 | 0.01(13) | 0.143 | 1.02(1) | 1.00 | 2.22(4) | 2.06 |
| 0.533(1) | 0.542(23) | 0.5523 | 0.04(12) | 0.153 | 1.01(1) | 1.00 | 2.25(5) | 2.08 |
| 0.584(1) | 0.534(20) | 0.5429 | 0.11(21) | 0.165 | 1.01(1) | 1.00 | 2.28(6) | 2.08 |
| 0.630(2) | 0.524(27) | 0.5345 | 0.03(20) | 0.175 | 1.01(1) | 1.00 | 2.29(5) | 2.08 |
| 0.683(1) | 0.517(23) | 0.5251 | -0.05(11) | 0.186 | 1.01(1) | 1.00 | 2.31(5) | 2.10 |
| 0.731(1) | 0.513(27) | 0.5166 | -0.04(12) | 0.196 | 1.01(1) | 1.00 | 2.33(6) | 2.12 |
| 0.772(1) | 0.499(28) | 0.5095 | 0.1(2) | 0.204 | 1.01(0) | 1.00 | 2.36(5) | 2.12 |
| 0.824(2) | 0.494(23) | 0.5006 | -0.02(36) | 0.214 | 1.01(1) | 1.00 | 2.36(3) | 2.14 |
| 0.874(1) | 0.484(28) | 0.4920 | 0.03(56) | 0.223 | 1.01(1) | 1.00 | 2.37(5) | 2.14 |
| 0.929(1) | 0.479(28) | 0.4829 | -0.23(84) | 0.232 | 1.01(0) | 1.00 | 2.39(5) | 2.16 |
| 1.00 | 0.467(21) | 0.4729 | - | - | - | - | 2.41(5) | 2.18 |

TABLE S12. Interfacial properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.715\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.541(26) | 0.5579 | - | - | - | - | 2.34(5) | 2.12 |
| 0.0049(2) | 0.543(20) | 0.5568 | 0.004(3) | 0.002 | 1.35(12) | 1.01 | 2.31(5) | 2.12 |
| 0.053(1) | 0.537(21) | 0.5463 | 0.01(2) | 0.016 | 1.11(4) | 1.00 | 2.35(4) | 2.12 |
| 0.105(2) | 0.526(24) | 0.5350 | 0.03(3) | 0.031 | 1.06(3) | 1.00 | 2.37(5) | 2.14 |
| 0.151(1) | 0.511(22) | 0.5255 | 0.03(4) | 0.044 | 1.04(2) | 1.00 | 2.40(4) | 2.16 |
| 0.200(1) | 0.502(23) | 0.5152 | 0.10(7) | 0.058 | 1.04(2) | 1.00 | 2.44(4) | 2.16 |
| 0.243(1) | 0.497(23) | 0.5064 | 0.06(7) | 0.069 | 1.03(2) | 1.00 | 2.44(7) | 2.20 |
| 0.289(2) | 0.484(22) | 0.4973 | 0.03(7) | 0.081 | 1.03(2) | 1.00 | 2.47(7) | 2.20 |
| 0.339(1) | 0.482(16) | 0.4876 | 0.11(10) | 0.093 | 1.03(2) | 1.00 | 2.52(5) | 2.22 |
| 0.380(2) | 0.474(24) | 0.4797 | 0.09(11) | 0.103 | 1.03(2) | 1.00 | 2.51(5) | 2.24 |
| 0.427(2) | 0.465(18) | 0.4707 | 0.10(07) | 0.114 | 1.02(1) | 1.00 | 2.53(5) | 2.24 |
| 0.476(1) | 0.453(21) | 0.4616 | 0.08(5) | 0.125 | 1.02(1) | 1.00 | 2.58(6) | 2.26 |
| 0.520(1) | 0.445(22) | 0.4535 | 0.12(9) | 0.134 | 1.02(1) | 1.00 | 2.59(7) | 2.28 |
| 0.562(1) | 0.439(27) | 0.4458 | 0.05(9) | 0.143 | 1.01(1) | 1.00 | 2.63(7) | 2.30 |
| 0.619(2) | 0.429(21) | 0.4356 | 0.2(3) | 0.155 | 1.02(1) | 1.00 | 2.66(7) | 2.32 |
| 0.661(3) | 0.424(25) | 0.4283 | 0.04(23) | 0.163 | 1.01(1) | 1.00 | 2.68(6) | 2.34 |
| 0.711(1) | 0.414(25) | 0.4197 | -0.11(18) | 0.172 | 1.01(1) | 1.00 | 2.64(5) | 2.34 |
| 0.759(2) | 0.406(25) | 0.4114 | -0.04(30) | 0.181 | 1.01(1) | 1.00 | 2.67(6) | 2.36 |
| 0.810(1) | 0.398(26) | 0.4028 | 0.09(27) | 0.190 | 1.013(5) | 1.00 | 2.74(5) | 2.38 |
| 0.853(1) | 0.387(25) | 0.3957 | 0.02(34) | 0.197 | 1.010(4) | 1.00 | 2.76(7) | 2.40 |
| 0.899(1) | 0.382(23) | 0.3882 | -0.1(4) | 0.205 | 1.009(4) | 1.00 | 2.77(5) | 2.42 |
| 0.948(1) | 0.375(29) | 0.3802 | -0.8(8) | 0.213 | 1.004(3) | 1.00 | 2.83(7) | 2.44 |
| 1.000 | 0.364(13) | 0.3734 | - | - | - | - | 2.83(5) | 2.44 |

TABLE S13. Interfacial properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.77\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.454(20) | 0.4567 | - | - | - | - | 2.68(4) | 2.32 |
| 0.064(2) | 0.439(13) | 0.4433 | 0.03(3) | 0.017 | 1.09(5) | 1.00 | 2.72(4) | 2.36 |
| 0.110(1) | 0.423(16) | 0.4340 | 0.06(2) | 0.029 | 1.06(2) | 1.00 | 2.75(6) | 2.38 |
| 0.154(2) | 0.417(18) | 0.4251 | 0.02(3) | 0.040 | 1.03(2) | 1.00 | 2.76(4) | 2.40 |
| 0.202(2) | 0.410(12) | 0.4155 | 0.07(5) | 0.052 | 1.03(2) | 1.00 | 2.81(6) | 2.42 |
| 0.246(2) | 0.405(16) | 0.4071 | 0.10(6) | 0.062 | 1.04(2) | 1.00 | 2.88(8) | 2.44 |
| 0.296(2) | 0.393(15) | 0.3974 | 0.01(6) | 0.074 | 1.01(2) | 1.00 | 2.86(5) | 2.46 |
| 0.334(1) | 0.385(15) | 0.3902 | 0.12(6) | 0.082 | 1.024(13) | 1.00 | 2.90(7) | 2.48 |
| 0.392(2) | 0.376(11) | 0.3795 | 0.10(6) | 0.094 | 1.023(10) | 1.00 | 2.91(5) | 2.52 |
| 0.436(1) | 0.369(14) | 0.3715 | 0.13(9) | 0.104 | 1.021(11) | 1.00 | 2.96(8) | 2.54 |
| 0.484(2) | 0.361(16) | 0.3628 | 0.10(9) | 0.113 | 1.023(9) | 1.00 | 2.98(6) | 2.56 |
| 0.527(2) | 0.350(16) | 0.3552 | -0.02(18) | 0.122 | 1.007(16) | 1.00 | 3.00(7) | 2.58 |
| 0.574(2) | 0.343(20) | 0.3471 | 0.20(15) | 0.130 | 1.018(8) | 1.00 | 3.07(7) | 2.60 |
| 0.617(2) | 0.327(15) | 0.3396 | 0.21(12) | 0.138 | 1.016(6) | 1.00 | 3.11(7) | 2.62 |
| 0.662(1) | 0.323(17) | 0.3319 | 0.12(9) | 0.146 | 1.012(5) | 1.00 | 3.12(7) | 2.66 |
| 0.714(2) | 0.317(18) | 0.3231 | 0.18(13) | 0.155 | 1.013(6) | 1.00 | 3.17(7) | 2.66 |
| 0.764(1) | 0.319(14) | 0.3149 | 0.20(18) | 0.163 | 1.012(4) | 1.00 | 3.22(11) | 2.70 |
| 0.808(2) | 0.302(13) | 0.3077 | 0.32(15) | 0.170 | 1.011(4) | 1.00 | 3.23(7) | 2.72 |
| 0.863(1) | 0.292(11) | 0.2988 | -0.10(14) | 0.179 | 1.007(4) | 1.00 | 3.27(7) | 2.74 |
| 0.904(1) | 0.288(14) | 0.2922 | 0.04(22) | 0.185 | 1.009(4) | 1.00 | 3.27(7) | 2.78 |
| 0.952(1) | 0.283(15) | 0.2846 | 0.5(4) | 0.192 | 1.008(1) | 1.00 | 3.36(10) | 2.80 |
| 1.000 | 0.273(18) | 0.2786 | - | - | - | - | 3.38(6) | 2.82 |

TABLE S14. Interfacial properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.825\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.350(17) | 0.3598 | - | - | - | - | 3.06(7) | 2.62 |
| 0.0055(3) | 0.347(14) | 0.3587 | 0.00(1) | 0.001 | 1.24(12) | 1.00 | 3.09(8) | 2.64 |
| 0.051(1) | 0.343(16) | 0.3497 | 0.04(2) | 0.012 | 1.09(5) | 1.00 | 3.17(8) | 2.66 |
| 0.096(1) | 0.329(14) | 0.3408 | 0.01(3) | 0.023 | 1.04(2) | 1.00 | 3.18(7) | 2.68 |
| 0.145(1) | 0.323(17) | 0.3314 | 0.04(5) | 0.034 | 1.03(2) | 1.00 | 3.26(9) | 2.70 |
| 0.195(2) | 0.311(15) | 0.3221 | 0.01(4) | 0.044 | 1.02(2) | 1.00 | 3.30(9) | 2.74 |
| 0.246(1) | 0.306(14) | 0.3125 | 0.03(7) | 0.055 | 1.02(2) | 1.00 | 3.33(9) | 2.78 |
| 0.280(1) | 0.302(19) | 0.3064 | 0.08(7) | 0.062 | 1.02(1) | 1.00 | 3.35(7) | 2.80 |
| 0.330(1) | 0.282(20) | 0.2973 | 0.07(5) | 0.072 | 1.02(1) | 1.00 | 3.39(7) | 2.84 |
| 0.351(2) | 0.285(17) | 0.2936 | 0.07(9) | 0.076 | 1.02(1) | 1.00 | 3.47(10) | 2.86 |
| 0.390(2) | 0.280(20) | 0.2867 | 0.03(10) | 0.084 | 1.02(2) | 1.00 | 3.46(6) | 2.88 |
| 0.435(2) | 0.272(20) | 0.2788 | 0.06(7) | 0.092 | 1.02(1) | 1.00 | 3.50(9) | 2.92 |
| 0.481(2) | 0.263(20) | 0.2709 | 0.08(8) | 0.100 | 1.02(1) | 1.00 | 3.53(8) | 2.94 |
| 0.526(2) | 0.253(18) | 0.2632 | 0.05(12) | 0.108 | 1.013(4) | 1.00 | 3.62(12) | 2.98 |
| 0.562(4) | 0.253(16) | 0.2572 | 0.05(10) | 0.114 | 1.002(12) | 1.00 | 3.54(12) | 3.02 |
| 0.604(2) | 0.242(19) | 0.2503 | 0.05(23) | 0.121 | 1.012(7) | 1.00 | 3.75(12) | 3.04 |
| 0.651(1) | 0.237(19) | 0.2425 | -0.07(17) | 0.129 | 1.008(7) | 1.00 | 3.71(10) | 3.08 |
| 0.795(2) | 0.207(14) | 0.2194 | 0.04(20) | 0.150 | 1.007(3) | 1.00 | 3.96(9) | 3.20 |
| 0.83(4) | 0.199(13) | 0.2145 | 0.03(26) | 0.154 | 1.007(3) | 1.00 | 3.96(12) | 3.22 |
| 0.895(1) | 0.196(12) | 0.2040 | 0.28(18) | 0.163 | 1.008(4) | 1.00 | 4.09(10) | 3.30 |
| 0.948(1) | 0.189(11) | 0.1959 | -0.08(33) | 0.170 | 1.008(3) | 1.00 | 4.16(12) | 3.34 |
| 0.959(1) | 0.189(15) | 0.1942 | 0.35(31) | 0.171 | 1.008(1) | 1.00 | 4.20(16) | 3.36 |
| 1.000 | 0.180(8) | 0.1896 | - | - | - | - | 4.20(11) | 3.40 |

TABLE S15. Interfacial properties of the binary LJTS mixture B ($\varepsilon_2/\varepsilon_1 = 0.9$ and $\xi_{12} = 1$) at $T = 0.88\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.264(14) | 0.2675 | - | - | - | - | 3.67(9) | 3.04 |
| 0.0051(3) | 0.253(14) | 0.2665 | 0.002(7) | 0.001 | 1.43(9) | 1.00 | 3.68(8) | 3.04 |
| 0.050(1) | 0.247(8) | 0.2582 | 0.006(13) | 0.011 | 1.11(2) | 1.00 | 3.75(12) | 3.06 |
| 0.098(1) | 0.239(12) | 0.2494 | 0.03(3) | 0.020 | 1.10(2) | 1.00 | 3.83(9) | 3.12 |
| 0.143(1) | 0.233(15) | 0.2412 | 0.05(2) | 0.029 | 1.07(1) | 1.00 | 3.90(11) | 3.18 |
| 0.187(1) | 0.226(11) | 0.2334 | 0.02(4) | 0.038 | 1.05(2) | 1.00 | 3.95(10) | 3.20 |
| 0.240(2) | 0.213(14) | 0.2242 | 0.01(4) | 0.047 | 1.04(1) | 1.00 | 4.01(9) | 3.28 |
| 0.280(2) | 0.208(13) | 0.2174 | 0.07(5) | 0.055 | 1.06(1) | 1.00 | 4.04(13) | 3.32 |
| 0.322(2) | 0.204(13) | 0.2102 | 0.08(4) | 0.062 | 1.04(1) | 1.00 | 4.03(15) | 3.36 |
| 0.365(2) | 0.197(15) | 0.2029 | 0.09(4) | 0.069 | 1.03(1) | 1.00 | 4.17(11) | 3.40 |
| 0.356(2) | 0.202(16) | 0.2045 | 0.08(5) | 0.068 | 1.04(1) | 1.00 | 4.18(12) | 3.38 |
| 0.401(3) | 0.194(17) | 0.1970 | 0.05(7) | 0.075 | 1.03(1) | 1.00 | 4.22(19) | 3.44 |
| 0.431(2) | 0.188(17) | 0.1921 | 0.04(7) | 0.080 | 1.03(1) | 1.00 | 4.26(13) | 3.48 |
| 0.478(4) | 0.180(18) | 0.1845 | 0.05(6) | 0.087 | 1.01(2) | 1.00 | 4.37(16) | 3.54 |
| 0.514(3) | 0.174(19) | 0.1787 | -0.01(9) | 0.093 | 1.00(3) | 1.00 | 4.49(3) | 3.58 |
| 0.552(4) | 0.164(18) | 0.1728 | 0.10(10) | 0.098 | 1.02(2) | 1.00 | 4.46(19) | 3.64 |
| 0.576(6) | 0.164(18) | 0.1689 | -0.07(6) | 0.102 | 1.01(3) | 1.00 | 4.6(4) | 3.66 |
| 0.622(5) | 0.156(16) | 0.1619 | -0.02(8) | 0.108 | 1.01(2) | 1.00 | 4.66(5) | 3.74 |
| 0.651(5) | 0.148(21) | 0.1573 | -0.08(7) | 0.112 | 1.01(2) | 1.00 | 4.7(3) | 3.76 |
| 0.873(1) | 0.113(8) | 0.1245 | 0.20(19) | 0.139 | 1.010(4) | 1.00 | 5.2(2) | 4.16 |
| 0.76(9) | 0.119(11) | 0.1411 | 0.31(32) | 0.126 | 1.01(6) | 1.00 | 5.03(2) | 3.94 |
| 0.929(1) | 0.108(10) | 0.1165 | 0.19(20) | 0.144 | 1.008(2) | 1.00 | 5.38(19) | 4.26 |
| 0.946(1) | 0.103(11) | 0.1141 | 0.0(3) | 0.146 | 1.007(3) | 1.00 | 5.38(24) | 4.30 |
| 0.969(1) | 0.103(12) | 0.1109 | -0.2(5) | 0.148 | 1.008(3) | 1.00 | 5.63(26) | 4.36 |
| 1.000 | 0.097(13) | 0.1080 | - | - | - | - | 5.70(26) | 4.38 |

TABLE S16. Interfacial properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.66\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.655(26) | 0.6632 | - | - | - | - | 2.06(5) | 1.94 |
| 0.009(1) | 0.601(22) | 0.6152 | 0.07(1) | 0.075 | 4.6(6) | 7.12 | 2.19(6) | 2.06 |
| 0.0077(5) | 0.609(28) | 0.6222 | 0.07(1) | 0.064 | 5.8(3) | 7.10 | 2.18(5) | 2.04 |
| 0.018(1) | 0.573(24) | 0.5659 | 0.12(1) | 0.157 | 4.5(3) | 7.22 | 2.28(5) | 2.20 |
| 0.027(1) | 0.536(25) | 0.5201 | 0.19(2) | 0.238 | 4.3(3) | 7.23 | 2.45(5) | 2.34 |
| 0.036(1) | 0.493(34) | 0.4768 | 0.25(2) | 0.318 | 4.2(2) | 7.08 | 2.58(6) | 2.48 |
| 0.045(1) | 0.477(35) | 0.4317 | 0.31(3) | 0.406 | 4.0(2) | 6.74 | 2.73(3) | 2.66 |
| 0.055(2) | 0.430(30) | 0.3871 | 0.35(2) | 0.498 | 4.0(1) | 6.32 | 2.92(4) | 2.88 |
| 0.063(3) | 0.393(30) | 0.3535 | 0.48(4) | 0.573 | 4.1(3) | 5.98 | 3.15(8) | 3.08 |
| 0.075(2) | 0.366(37) | 0.2988 | 0.53(4) | 0.709 | 3.7(2) | 5.38 | 3.31(10) | 3.48 |
| 0.085(2) | 0.328(31) | 0.2594 | 0.64(6) | 0.825 | 3.7(1) | 4.93 | 3.59(7) | 3.88 |
| 0.092(5) | 0.295(17) | 0.2285 | 0.75(5) | 0.933 | 3.8(3) | 4.58 | 3.00(14) | 4.28 |
| 0.111(2) | 0.260(19) | 0.1493 | 0.75(6) | 1.360 | 3.3(1) | 3.67 | 4.30(17) | 6.12 |
| 0.117(4) | 0.215(32) | 0.1253 | 1.08(5) | 1.594 | 3.5(2) | 3.38 | 5.08(16) | 7.22 |

TABLE S17. Interfacial properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.715\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.541(26) | 0.5579 | - | - | - | - | 2.34(5) | 2.12 |
| 0.0096(4) | 0.511(22) | 0.5189 | 0.05(1) | 0.056 | 3.7(4) | 4.93 | 2.45(5) | 2.22 |
| 0.0129(9) | 0.494(17) | 0.5055 | 0.08(1) | 0.076 | 3.9(4) | 4.90 | 2.50(7) | 2.26 |
| 0.027(1) | 0.452(25) | 0.4496 | 0.14(2) | 0.162 | 3.4(2) | 4.73 | 2.69(5) | 2.44 |
| 0.043(2) | 0.413(18) | 0.3944 | 0.18(2) | 0.252 | 3.0(2) | 4.48 | 2.89(7) | 2.68 |
| 0.056(2) | 0.363(32) | 0.3466 | 0.30(3) | 0.336 | 3.1(1) | 4.19 | 3.19(9) | 2.92 |
| 0.071(1) | 0.319(31) | 0.3008 | 0.38(4) | 0.422 | 3.0(1) | 3.87 | 3.44(8) | 3.20 |
| 0.084(3) | 0.273(29) | 0.2608 | 0.44(3) | 0.504 | 2.9(1) | 3.56 | 3.9(1) | 3.52 |
| 0.109(2) | 0.257(19) | 0.1891 | 0.50(5) | 0.675 | 2.5(1) | 2.95 | 4.0(1) | 4.36 |
| 0.124(6) | 0.208(26) | 0.1514 | 0.64(6) | 0.784 | 2.5(1) | 2.59 | 4.6(2) | 5.10 |
| 0.142(5) | 0.172(22) | 0.1061 | 0.77(6) | 0.932 | 2.43(6) | 2.11 | 5.4(2) | 6.52 |
| 0.178(8) | 0.112(24) | 0.0330 | 0.96(7) | 0.180 | 2.13(8) | 1.02 | 6.8(4) | 5.76 |

TABLE S18. Interfacial properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.77\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.454(20) | 0.4567 | - | - | - | - | 2.68(4) | 2.32 |
| 0.0045(3) | 0.439(28) | 0.4423 | 0.019(4) | 0.019 | 2.8(4) | 3.58 | 2.74(6) | 2.38 |
| 0.0097(6) | 0.411(18) | 0.4258 | 0.03(1) | 0.041 | 2.5(3) | 3.53 | 2.78(7) | 2.44 |
| 0.0136(9) | 0.406(14) | 0.4138 | 0.05(1) | 0.057 | 2.9(4) | 3.50 | 2.87(8) | 2.48 |
| 0.0271(9) | 0.366(23) | 0.3734 | 0.10(1) | 0.113 | 2.7(1) | 3.36 | 3.01(6) | 2.64 |
| 0.044(1) | 0.331(22) | 0.3250 | 0.14(3) | 0.184 | 2.4(1) | 3.15 | 3.2(1) | 2.88 |
| 0.056(1) | 0.303(20) | 0.2935 | 0.20(3) | 0.232 | 2.5(1) | 3.00 | 3.5(1) | 3.06 |
| 0.075(2) | 0.270(22) | 0.2485 | 0.27(2) | 0.304 | 2.2(1) | 2.76 | 3.7(1) | 3.38 |
| 0.089(1) | 0.219(22) | 0.2154 | 0.29(2) | 0.360 | 2.22(6) | 2.56 | 4.0(1) | 3.68 |
| 0.122(3) | 0.170(18) | 0.1507 | 0.45(3) | 0.474 | 2.06(8) | 2.12 | 4.9(2) | 4.52 |
| 0.146(3) | 0.150(17) | 0.1090 | 0.49(5) | 0.544 | 1.88(6) | 1.80 | 5.1(2) | 5.40 |
| 0.163(4) | 0.115(16) | 0.0834 | 0.59(6) | 0.566 | 1.83(6) | 1.58 | 6.1(3) | 6.14 |
| 0.191(8) | 0.088(19) | 0.0492 | 0.60(6) | 0.492 | 1.66(4) | 1.26 | 6.7(4) | 7.28 |
| 0.215(14) | 0.063(18) | 0.0282 | 0.68(11) | 0.318 | 1.56(4) | 1.07 | 8.0(7) | 7.72 |

TABLE S19. Interfacial properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.825\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.350(17) | 0.3598 | - | - | - | - | 3.06(7) | 2.62 |
| 0.0041(3) | 0.346(16) | 0.3493 | 0.02(1) | 0.013 | 2.5(2) | 2.70 | 3.19(5) | 2.68 |
| 0.0083(9) | 0.335(13) | 0.3387 | 0.02(1) | 0.026 | 2.1(3) | 2.67 | 3.19(8) | 2.72 |
| 0.0177(6) | 0.303(16) | 0.3159 | 0.05(2) | 0.054 | 2.2(1) | 2.60 | 3.35(8) | 2.82 |
| 0.029(2) | 0.280(14) | 0.2901 | 0.08(2) | 0.087 | 2.1(2) | 2.51 | 3.5(1) | 2.96 |
| 0.038(1) | 0.268(12) | 0.2690 | 0.12(2) | 0.115 | 2.1(1) | 2.43 | 3.6(1) | 3.10 |
| 0.048(1) | 0.245(15) | 0.2487 | 0.15(2) | 0.142 | 2.0(1) | 2.35 | 3.7(1) | 3.24 |
| 0.069(2) | 0.212(22) | 0.2071 | 0.18(3) | 0.198 | 1.8(1) | 2.17 | 4.1(1) | 3.56 |
| 0.103(2) | 0.163(12) | 0.1497 | 0.25(4) | 0.278 | 1.74(6) | 1.88 | 4.8(2) | 4.24 |
| 0.125(3) | 0.130(12) | 0.1177 | 0.29(3) | 0.320 | 1.64(6) | 1.70 | 5.3(2) | 4.78 |
| 0.158(6) | 0.095(19) | 0.0783 | 0.37(4) | 0.358 | 1.54(7) | 1.45 | 6.4(3) | 5.74 |
| 0.191(5) | 0.064(17) | 0.0486 | 0.37(7) | 0.349 | 1.42(4) | 1.25 | 7.2(6) | 6.82 |

TABLE S20. Interfacial properties of the binary LJTS mixture A ($\varepsilon_2/\varepsilon_1 = 0.6$ and $\xi_{12} = 0.85$) at $T = 0.88\,\varepsilon k_\mathrm{B}^{-1}$. Results from MD and DGT+PeTS. Both methods are calculated at the same liquid phase composition $x_2^{L1}$. The number in the parentheses indicates the statistical uncertainty in the last decimal digit.

| $x_2^{L1}$ / mol mol$^{-1}$ | $\gamma$ / $\varepsilon\sigma^{-2}$ | | $\Gamma_2^{(1)}$ / $\sigma^{-2}$ | | $E_2$ | | $L_{10}^{90}$ / $\sigma$ | |
|---|---|---|---|---|---|---|---|---|
| | MD | DGT | MD | DGT | MD | DGT | MD | DGT |
| 0.000 | 0.264(14) | 0.2675 | - | - | - | - | 3.67(9) | 3.04 |
| 0.0098(6) | 0.236(13) | 0.2481 | 0.01(1) | 0.022 | 1.77(19) | 2.08 | 3.81(8) | 3.16 |
| 0.0143(5) | 0.227(16) | 0.2394 | 0.03(1) | 0.032 | 1.81(13) | 2.06 | 3.89(9) | 3.20 |
| 0.0185(6) | 0.225(15) | 0.2316 | 0.05(1) | 0.041 | 1.85(13) | 2.03 | 4.0(1) | 3.26 |
| 0.0248(9) | 0.214(14) | 0.2201 | 0.04(2) | 0.055 | 1.69(12) | 1.99 | 4.0(1) | 3.36 |
| 0.027(2) | 0.210(14) | 0.2153 | 0.06(1) | 0.060 | 1.62(17) | 1.98 | 4.1(1) | 3.38 |
| 0.029(1) | 0.209(19) | 0.2128 | 0.06(1) | 0.063 | 1.72(11) | 1.97 | 4.1(1) | 3.40 |
| 0.0345(1) | 0.201(18) | 0.2028 | 0.08(1) | 0.075 | 1.70(11) | 1.93 | 4.3(1) | 3.50 |
| 0.038(2) | 0.192(10) | 0.1961 | 0.09(3) | 0.083 | 1.71(12) | 1.91 | 4.4(2) | 3.56 |
| 0.043(1) | 0.184(9) | 0.1891 | 0.09(2) | 0.091 | 1.71(8) | 1.88 | 4.4(1) | 3.60 |
| 0.100(2) | 0.105(16) | 0.1085 | 0.19(3) | 0.186 | 1.52(5) | 1.55 | 5.7(2) | 4.74 |
| 0.166(6) | 0.056(14) | 0.0476 | 0.22(5) | 0.228 | 1.28(6) | 1.24 | 7.5(6) | 6.66 |
| 0.223(11) | 0.021(18) | 0.0187 | 0.19(8) | 0.185 | 1.14(3) | 1.07 | 10.3(20) | 8.88 |