

Comment on "Pair Interaction from Structural Data of Dense Classical Liquids"

Recently, Levesque, Weis, and Reatto (LWR) proposed¹ a method of determining pair potentials from liquid structure data. It was tested by use of a Monte Carlo-computed $g(r)$ for the Lennard-Jones (LJ) fluid and the Dagens-Rasolt-Taylor² (DRT) liquid aluminum. They stated that our method³ for extracting pair potentials was found inadequate for these systems. As our method had been tested with simulation data for DRT liquid Al and for Kr, and also applied to experimental data, their claim, if true, would be serious. Here we show that our method applied to their own data recovers the original potentials more accurately than that of LWR. We also confirmed⁴ that LWR *did not* really test our procedure.³ Instead, LWR inverted an extrapolated $g(r)$ using the modified hypernetted chain (MHNC) equation⁴ to get a potential βv_0 , which they *assumed* would correspond to our result. This is unwarranted since we had rejected³ direct inversion which is a one-to-many mapping, unless the $S(k)$ is essentially exact for *all* k . Instead, we use the MHNC "in the forward direction" where a parametrized pair potential and the hard-sphere bridge parameter⁵ η are optimized to fit the given $S(k)$, available in some range $k_{\min} \leq k \leq k_{\max}$. This avoids a model for extrapolating $g(r)$ or $S(k)$ but requires a model for the potentials. The latter is firmly based on pseudopotential theory, and constrains the one-to-many mapping to that defined by the physical model. Monte Carlo procedures can be used in our method instead of MHNC if necessary, but MHNC was found to be sufficient. Thus the strength of the method is that it avoids doubtful extrapolations and exploits a physical model.

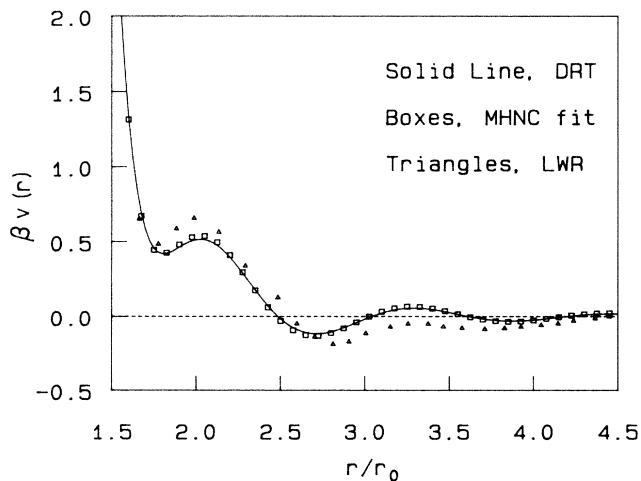


FIG. 1. Comparison of potentials for liquid Al.

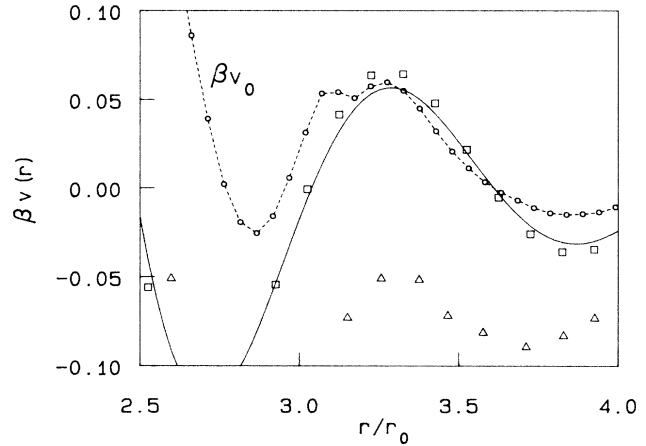


FIG. 2. LWR's βv_0 , other curves as in Fig. 1.

LWR used Verlet's extrapolation of $g(r)$ for $r > R_{\max}$ using a model $C(r)$ for $r > R_{\max}$. Hence their $S(k)$ is model dependent, and contains *small but significant errors*, especially for small k . We used the approximations to $S(k)$ given to us by LWR with our MHNC fitting method and recovered the original LJ and DRT potentials successfully. Here we give details only for the more complicated liquid-Al case. As in Ref. 3, a pair potential involving a simple pseudopotential (A_0 = well depth, R_0 = s-wave cutoff radius) and the LDA screening function³ containing the electron gas parameter r_s^* were used. More general potentials were tested in Ref. 3 and found to be unnecessary. Our parametrization is *not* based on a knowledge of the DRT potential which uses six parameters. The resulting MHNC-fitted potential is compared in Figs. 1 and 2 with the original DRT potential and the best Al potential of LWR. In Fig. 2 βv_0 of LWR is also shown. The unexpected kink in βv_0 raises questions about the quality of their data.

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