

### Comment on "Pair Interaction from Structural Data for Dense Classical Liquids"

Recently Levesque, Weis, and Reatto<sup>1</sup> proposed an interesting iterative predictor-corrector method (PCM) for the determination of the pair potential from structural data for simple liquids. It is obviously a very useful alternative to the self-consistent method (SM) given elsewhere.<sup>2</sup> As demonstrated in Ref. 1 the two methods lead to different results for the pair potential (see Fig. 1 of Ref. 1). In our opinion there are three possible explanations for these differences.

(1) *Result from the SM.*—The statistical error in  $\Delta g = g(r) - g_i(r)$ , where  $g(r)$  is the "exact" and  $g_i(r)$  the approximated pair correlation function, might still be too large after the  $i$ th (in Ref. 1  $i=13$ ) iteration step and, therefore, the resulting pair potential  $v_i(r) = v(r)$  is *not yet* the correct answer. It is well known that the pair potential is very sensitive to small variations in  $g(r)$ .

(2) *Result from the PMC.*—The reference pair potential used (Lennard-Jones) is not quite consistent with the density used in the simulation of  $g(r)$ . It is known, for example, that in the case of a crystal the lattice constant (and therefore the density) is determined by minimizing the free energy, and any pair potential must not lead to the experimentally observed density. In fact, I found that my method [scheme (9) of Ref. 2] is very sensitive to small variations in density.

(3) *Different pair potentials can produce the same pair correlation function.* In other words, one would not be able to extract unique pair potentials from structure data. Such behavior cannot be excluded completely, but is in my opinion very unlikely.

The explanation given in Ref. 1 for the differences in the potentials is excluded because the SM is not based on the assumption that the function

$$\gamma(r) = g(r) \exp\{\beta v(r)\} \quad (1)$$

does not change when the pair potential  $v(r)$  is varied. After the  $l$ th iteration [scheme (9) in Ref. 2] we have

$$\begin{aligned} v_l(r) &= v_{l-1}(r) \\ &= v(r) \end{aligned} \quad (2)$$

and

$$\begin{aligned} g_l(r) &= g_{l-1}(r) \\ &= g(r), \end{aligned} \quad (3)$$

where  $g(r)$  is the experimentally determined pair correlation function. Consequently, with

$$g_l(r) = \exp\{-\beta v_l(r)\} \gamma_l(r), \quad (4)$$

$$g(r) = \exp\{-\beta v(r)\} \gamma(r), \quad (5)$$

and Eqs. (2) and (3) we obtain

$$\gamma(r) = \gamma_l(r), \quad (6)$$

and this is not an assumption but follows directly from Eq. (2). Clearly, for iteration steps  $i < l$  we have

$$\gamma_1(r) \neq \gamma_2(r) \neq \dots \neq \gamma_{l-1}(r). \quad (7)$$

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<sup>1</sup>D. Levesque, J. J. Weis, and L. Reatto, Phys. Rev. Lett. **54**, 451 (1985).

<sup>2</sup>W. Schommers, Phys. Rev. A **28**, 3599 (1983).