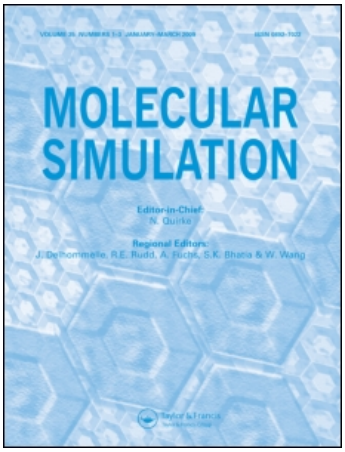

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Structure of Supercritical Fluid Krypton at Small Scattering Angle Using Parallel Molecular Dynamics Simulation

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STRUCTURE OF SUPERCRITICAL FLUID KRYPTON AT SMALL SCATTERING ANGLE USING PARALLEL MOLECULAR DYNAMICS SIMULATION

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We present a parallel algorithm for molecular dynamics involving short-range two- and three-body potentials and the pair-correlation function, $g(r)$. The method is based on a spatial decomposition of the simulation box that takes advantage of a linked-cell list, and allows a load balanced partition of the computations of both the forces and $g(r)$ over the processors. The tests of the program is conducted by evaluating the efficiency for both the thermalization phase and the production phase of the simulation. This method is successfully applied to the calculation of the direct correlation function of fluid krypton at small scattering angle along the $T = 297\,\mathrm{K}$ supercritical isotherm.

Keywords: Krypton; parallel molecular dynamics; linked-cell list; three-body potential; fluids; structure factor

1. INTRODUCTION

The liquid state theory is commonly based on the formalism of distribution functions of the classical statistical mechanics [1]. In this framework the pair-correlation function, $g(r)$, plays the central role in the sense that in principle all the physical properties of the fluid can be derived from it. For instance, its Fourier transform, which is the static structure factor, $S(q)$,

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measured by X-ray diffraction or neutron scattering, allows a direct comparison with the experiment. Once the interactions are given, molecular dynamics (MD) simulation is a powerful method [2, 3] that provides a direct link between the interactions and the physical properties since an "exact" result of $g(r)$ is obtained.

The precise determination of $S(q)$ implies the knowledge of the pair-correlation function over a wide enough range of distances. Therefore MD simulations can be done only at the price of a huge amount of computer time. Indeed, the MD method consists in solving iteratively Newton's equations of motion of N particles in a simulation box by means of a finite difference method [3]. A large number of discrete time-steps is required to produce a sufficient part of the phase space trajectory from which a statistically correct $g(r)$ is extracted. In principle, the calculation of the forces between atoms, which basically consists in performing at each time-step a double loop for a pair potential, and eventually a triple loop if a three-body potential has to be taken into account, is the most time consuming part of the simulation. Nevertheless, the determination of $g(r)$, which involves all pairs of particles, can even be more expensive from a computational point of view than the forces for large N.

Since the late eighties, much effort has been devoted to the construction of MD algorithms for parallel machines (see the paper by Plimpton [4] and references therein) to reduce the computational time and to allow the simulation of systems containing a larger number of particles ($N > 10000$). For liquids such a strategy is still developing [5], and recently, we have built a parallel MD algorithm [6] for liquids in which the calculation of the two- as well as the three-body forces has a complexity reduced to $O(N)$. It is based on a spatial decomposition (SD) of the simulation box that takes advantage of linked-cell lists, which is similar to that proposed by Esselink *et al.* [7, 8] for chain molecules. The aim of the present work is to extend our parallel algorithm to couple the calculation of the force having a $O(N)$ complexity to that of $g(r)$, which has a $O(N^2)$ complexity. We propose and compare different strategies that can be put forward to keep a good balance of the computational load amongst the processors, depending upon the number of particles and the nature of the interactions. The program is then applied to the calculation of the structure factor of fluid krypton for which recent small-angle neutron scattering experiments were done [10], representing a stringent test of the interaction model.

The layout of the paper is the following. In Sect. 2, we present the potential energy function suitable for fluid krypton as well as a parallel algorithm for the MD simulation. In Sect. 3, we illustrate the efficiencies as

well as the limitations of our algorithm and we display and discuss the physical results. Finally, in Sect. 4, we summarize and draw our conclusions.

2. NUMERICAL BACKGROUND

2.1. Fluid Structure and Potential Energy Function

The central quantity of the modern theory of fluids is the pair-correlation function $g(r)$. The latter represent the density fluctuations $\rho(r)$ ($= 4\pi r^2 \rho g(r)$) around the mean number density ρ and describes the local order in the fluid. As the distance r between two particles tends to infinity, $g(r)$ tends to 1, which means that particles are no more correlated and the fluid can be considered as totally disordered. The pair-correlation function is defined by

$$g(r) = \lim_{\Delta r \to 0} \frac{N(r)}{4\pi r^2 \rho \Delta r} \tag{1}$$

where $N(r)$ is the time-average number of particles contained in a spherical shell of thickness Δr, *i.e.*, limited by two spheres of radius r and $r + \Delta r$ respectively. The calculation of Eq. (1) is achieved in two stages. Firstly, if the positions of the particles are known from a particular configuration at a time t, as it is the case in a molecular dynamics simulation, a function $N^t(r)$ is easy to determine by the following relation

$$N^t(r) = \frac{2}{N} \sum_{i=1}^{N} \sum_{j>i=1}^{N} N_i^t(r_{ij}), \tag{2}$$

where $N_i^t(r)$ is a function $N(r)$ with particle i taken as the origin. Equation (2) is therefore calculated in an average manner by taking successively all N particles as the origin. This computation, involving a double loop over the particles, is time consuming even if the symmetry property reduces it by a factor 2. Finally, $g(r)$ is statistically approached by averaging $N^t(r)$ over a set of n time-independent configurations.

A reliable description of the structural properties of fluid krypton uses a potential energy function

$$U_N(\mathbf{r}^N) = \sum_{i<j=1}^{N} u_2(\mathbf{r}_i, \mathbf{r}_j) + \sum_{i<j<k=1}^{N} u_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) \tag{3}$$

that involves a pair potential u_2 as well as a three-body potential u_3. Following Barker and Henderson [11] higher order potential energy terms can be neglected. One of the most accurate pair potential is that of Aziz and Slaman [12]

$$u_2(x) = A\varepsilon \exp(-\alpha x + \beta x^2) - F(x) \sum_{j=0}^{2} \frac{C_{2j+6}}{x^{2j+6}}, \tag{4}$$

where $x = r/\sigma$ is the reduced distance, σ being the position of the node of the potential, and ε is the potential well-depth.

$$F(x) = \begin{cases} \exp[-(\frac{D}{x} - 1)^2] & \text{if } x < D, \\ 1 & \text{if } x \geq D, \end{cases} \tag{5}$$

is a switching function between repulsive and attractive parts, and the relevant parameters of u_2 are listed in the paper of Aziz and Slaman [12]. The three-body potential takes the usual form derived by Axilrod-Teller [13]

$$u_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) = c_3 \frac{1 + \cos\theta_i \cos\theta_j \cos\theta_k}{r_{ij}^3 r_{ik}^3 r_{jk}^3}, \tag{6}$$

which is the first term of the multipole expansion representing the triple-dipole interaction. The strength of u_3 is $c_3 = 1.56 \cdot 10^{-31}$ au for krypton. We denote by θ_i, θ_j and θ_k, respectively, the angles at vertex i, j and k of the triangle (i,j,k) with sides $r_{ij} = |\mathbf{r}_j - \mathbf{r}_i|$, $r_{ik} = |\mathbf{r}_k - \mathbf{r}_i|$ and $r_{jk} = |\mathbf{r}_k - \mathbf{r}_j|$.

In MD the knowledge of the forces is needed, and expressions (7) and (9) both yield simple analytical forms, that is very convenient for our computational purposes. For Aziz and Slaman's potential, the force acting on particle i from particle j, is

$$\mathbf{F}_{ij}(r_{ij}) = \left\{ A \exp(-\alpha x + \beta x^2)(-\alpha + 2\beta x) - F(x) \left[\sum_{j=0}^{2} \frac{(2j+6)C_{2j+6}}{x^{2j+7}} - \frac{2D(D-x)}{x^3} \sum_{j=0}^{2} \frac{C_{2j+6}}{x^{2j+6}} \right] \right\} e_{ij}, \tag{7}$$

where e_{ij} is a unit vector in the $\mathbf{r}_{ij}$-direction. For the Axilrod-Teller potential, the force acting on particle i from particles j and k reads

$$\mathbf{F}_{i,jk}(r_{ij}, r_{jk}, r_{ik}) = \frac{\partial u_3}{\partial r_{ij}} e_{ij} + \frac{\partial u_3}{\partial r_{ik}} e_{ik}, \tag{8}$$

while the forces acting on j and k are respectively

$$\begin{cases} \mathbf{F}_{j,ik}(r_{ij}, r_{jk}, r_{ik}) = -\dfrac{\partial u_3}{\partial r_{ij}} e_{ij} + \dfrac{\partial u_3}{\partial r_{jk}} e_{jk}, \\ \mathbf{F}_{k,ij}(r_{ij}, r_{jk}, r_{ik}) = -\dfrac{\partial u_3}{\partial r_{ik}} e_{ik} - \dfrac{\partial u_3}{\partial r_{jk}} e_{jk}. \end{cases} \tag{9}$$

The expressions of the three partial derivatives of u_3 can be found in the paper by Hoheisel [14]. From Newton's third law, we have $\mathbf{F}_{ij} = -\mathbf{F}_{ji}$ for the two-body forces and $\mathbf{F}_{i,jk} = -\mathbf{F}_{j,ik} - \mathbf{F}_{k,ij}$ for the three-body forces. Consequently, the forces can be calculated once, thus reducing the computational time both for pair and three-body forces.

2.2. Molecular Dynamics: Parallel Implementation

Classical molecular dynamics is a simulation method in which the N atoms or spherical molecules of the liquid are treated as point mass situated in a simulation box of volume V. In most of the cases, the latter is considered as cubic for liquids. The knowledge of the forces enables one to integrate the equations of motion of all the particles in a discrete form [2, 3] by means of a finite difference method. A convenient and accurate integration scheme is Verlet's algorithm in the velocity form, since only the knowledge of the positions and velocities at time t are needed to obtain the positions and velocities at the subsequent time-step $t + dt$. Periodic boundary conditions are applied to represent the liquid as infinite and the minimum image convention is implemented in the way described by Attard [15] for three-body potentials.

The two- and three-body potentials given by Eqs. (7) to (9) are short-ranged allowing the use of a cut-off radius R_c. This means that only pairs and triplets of particles within R_c are taken into account, reducing the computation of the forces to a $O(N)$ complexity. The large simulation box with side $L = V^{1/3}$ is divided into small cubic cells whose edge length e only depends on the cut-off radius of the interactions. The most convenient choice is $e = R_c + 2\delta$, where $\delta(\ll R_c)$ is chosen by fixing the number of cells along an edge of the simulation box. Once the particles are assigned geometrically to the cells, the computation of forces between particles situated in closed cells is conveniently done by setting up the linked-cell lists (LCL). This well-known method, which is fully used for sequential algorithms, is equally important for the parallel implementation. The way of handling such LCL in MD is described in the book of Allen and Tildesley [2]. This method is extended here to the three-body-forces, and the interested reader will find the details in Ref. [6].

Parallel algorithms for MD simulations are nowadays commonly written [4] and a natural way to built a parallel MD based upon a LCL is to group cells into D sub-domains, each of these being assigned to one processor [7, 8]. As far as a parallel machine is used, the main problem is to find a good partition of the computational load over the P processors of the computer. For the sake of simplicity, we assume that the P processors are identical and an obvious manner for splitting the problem is to equally distribute, when possible, the cells among the processors, according to the isotropic properties of the liquid we deal with. Subsequently, two kinds of actions are done by a processor: (i) local actions like computations of positions, velocities, forces and local energy, and (ii) global actions such as summations of local energies and calculations of the physical quantities of the simulated system from the knowledge of all the positions, velocities and forces.

The computation of the forces applied on particles of a cell needs communications with nearby cells. According to the LCL method, a processor receives data from the nearby sub-domains to obtain the positions of the particles within the cut-off radius. Then, it sends the calculated forces to be used as reaction forces in the nearby sub-domains. This implies a partition of the computations over the processors according to the locality of the calculation. In a previous paper [6], we have tested two different decompositions in which the cells were either (i) grouped into cubic sub-domains (SDC decomposition) or (ii) grouped into plates (SDP decomposition), for parallel machine being composed of few processors. Using 8 processors, the SDP decomposition we deal with hereafter, is the most interesting decomposition. Indeed periodic boundary conditions allows to describe the computational domain as a torus. In this case, one send and one receive per processor are needed for the two-body forces, whereas the number of messages depends on the number of layers of cells assigned to a processor for three-body forces. One layer involves sends with the two upper processors and receives from the two lower processors, whereas two or more layers requires communications only with nearby processors.

Beside the calculation of the forces, the other major problem is the computation of the pair-correlation function $g(r)$. While the theoretical complexity of force computation decreases to $O(N)$ by the use of the cut-off assumption mentioned above, the complexity of the $g(r)$ computation is of $O(N^2)$, and it cannot be reduced since it involves all the pairs of particles. Fincham [9] was the first to discuss the possibility of calculating $g(r)$ with parallel machines. Interestingly, the calculation of $g(r)$ has no influence on the numerical results of the simulation and can be treated as a diagnostic post-process, as it is done for analyzing most of the physical properties

under interest. Unfortunately, it requires the storage of a huge amount of data due to the large number of particles involved. An alternative solution is to evaluate $g(r)$ during the MD simulation. From a parallelism point of view, the computation of $g(r)$ only induces one-way communications from processors that solve a part of the motion equations to processors which compute contributions $N_i^{\nu}(r)$ in Eq. (2), results are summed up at the end of the simulation.

For the sake of simplicity, we assume that we deal with a homogeneous liquid on a homogeneous machine composed of P processors. The parallel implementation of the thermalization phase is classical [4] since the computations costs, coming only from the forces, are linear with respect to the number of particles, and the load-balancing is obtained simply by equally distributing the cells as well as the layers of cells over the processors. During the production phase, which includes the calculation of $g(r)$ as well, the following simple parallel scheme is set up to distribute the computational load. If one equally distributes the $D(D+1)/2$ pairs of sub-domains, then one equally distributes the pairs of particles. As a matter of fact, assuming that each processor computes correlations involving its assigned particles, the computation is balanced as soon as P is odd or $D = kP$, where k is any integer greater than 2.

3. TEST OF THE METHOD AND RESULTS

3.1. Tests of the Parallel Algorithm

First of all, we illustrate of the necessity to parallelize both the forces and pair-correlation function. Figure 1 shows the CPU (central process unit) time necessary to run sequential MD during 1000 time-steps. The results are obtained at a reduced density $\rho\sigma^3 = 0.22579$ and temperature $k_BT/\varepsilon = 1.4276$, corresponding the supercritical fluid condition close the experiment of Formisano *et al.* [10]. The pair-correlation function is evaluated each 5 MD iterations. One observes that the calculation of the two-body forces is always less expensive than that of $g(r)$, which means that for large N even a parallel computation of two-body interactions, coupled to that of $g(r)$ done on a unique processor, is a dead experience. For the three-body contribution the computational cost of the forces is higher than that of $g(r)$. Nevertheless, one can see that a mimic of a load-balanced distribution of the three-body force calculation on an ideal parallel machine composed of 8 processors, which consists in dividing the dashed curve by a factor 8

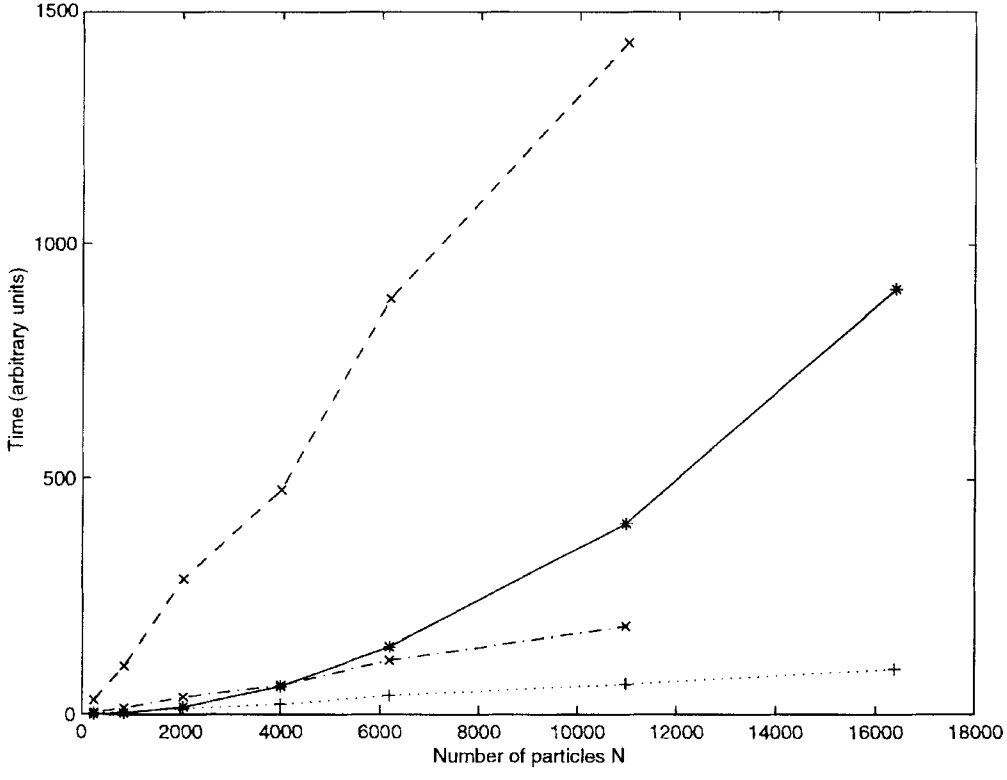

FIGURE 1 Computational costs in a sequential MD algorithm as a function of the number of particles coming from: the two-body forces (dotted curve), three-body forces (dashed curve), pair-correlation function $g(r)$ (solid line), and three-body forces divided by 8 (dash-dotted curve) to mimic an ideal parallel machine composed of 8 processors.

(dash-dotted curve), leads to computational cost which is cheaper for N greater than 4000. As a result, it is clearly necessary to develop a parallel algorithm in order to calculate $g(r)$ during the simulation in all the cases.

In the SDP method chosen here, the cells are grouped into layers, which are partitioned into D equal sub-domains. Each sub-domain is associated to one process, and the number of processors P is chosen as a divisor the number of processes. To test our parallel algorithm, the efficiency

$$E_P = \frac{\text{sequential CPU time}}{\text{sum of the } P \text{ CPU times on the parallel machine}} \tag{10}$$

is evaluated for different situations in which the number of processors and the number of layers per sub-domain is varied. The results gathered in Table I are obtained from simulations at $\rho\sigma^3 = 0.22579$ and $k_BT/\varepsilon = 1.4276$ involving $N = 16384$ particles distributed into 4096 cells using either 4 or 8 processors, associated with 8 or 16 sub-domains in each case.

First considering 8 sub-domains made of two layers of cells, one can notice that the efficiency decreases for all simulations when the number of processors P grows up. We obtain bad results for two-body computations since the relatively small density of the liquid in the present study yields to an average of 4 particles in each cell. Therefore, only a small number of pairs are involved in the computation. As expected, the computation of $g(r)$ obtained with 4 processors is efficient because the load is well balanced. On the contrary, the distribution of the couples of sub-domains is not balanced over the 8 processors, reducing the efficiency of the algorithm.

In order to balance the computation the same simulations were run with 16 sub-domains, each one corresponding to a layer of cells. In this case, a process is allocated to each sub-domain and 2 or 4 processes are put onto one processor, respectively for 4 and 8 processors. Most of the efficiency results are only slightly worst with respect to the case of 8 sub-domains,

TABLE I Efficiency E_P expressed in percent with respect to the number of processors and sub-domains. The last row corresponds to the CPU ratio of the calculations of forces and pair-correlation function with respect to the CPU time for that of the two-body forces

	Number of processors	*2-body*	*2 and 3-body*	*2-body and g(r)*
8 sub-domains	4	77.8	94.5	96.9
	8	70.7	92.6	84.5
16 sub-domains	4	69.5	90.5	92.7
	8	59.2	89.3	91.1
CPU ratios		1	17.4	20.59

essentially because the use of 16 sub-domains implies a drastic increase of communications. Nevertheless, the cost of the supplementary communications is overwhelmed by a good distribution of the computation of $g(r)$, the efficiency and CPU time of three-body force and pair-correlation function computations being of the same order of magnitude, as it can be seen on the CPU ratio displayed in Table I.

According to results of our simulations displayed in Figure 1, the following rules can be outlined: firstly, when simulations involve a very small number of particles N lower than say 500, sequential computations are faster than parallel ones, which are penalized by communication costs; for a number of particles lower than 4000, a good choice is, for example, 8 processors for the forces, and 1 processor for $g(r)$. This implies the send of all the positions to same processor dedicated to the calculation of $g(r)$. For a large number of particles, the computation of $g(r)$ is also implemented with a parallel scheme, requiring all-to-all sends and receives of the positions. Moreover, when only a two-body potential is involved, a sequential computation of the forces associated to a parallel scheme for $g(r)$, is a solution to reduce communications.

As far as parallel algorithms are considered, more general schemes can be designed in order to remove assumptions made on the number and the homogeneity of the processors mentioned above for our algorithm and the isotropy of the liquid. For example, one may think of a distribution of the computation of $g(r)$ with respect to a cell decomposition rather than a layer decomposition, or the use of processors devoted to the computation of the forces or that of $g(r)$. Another choice is to distribute the calculation of $g(r)$ over the processors, then to distribute the computation of the forces so as to remove the potential lack of balance and this can be done with multi-threading techniques [16].

3.2. Structure Factor of Fluid Krypton at Small Angle

We come now to the application of our program to the determination of the structure of fluid krypton. The general shape of $S(q)$ is essentially governed by the short-range repulsive part of the potential energy function, while its behavior at small scattering angle (low-q) is very sensitive to the long-range attractive part of the potential, and reflects the detailed nature and the peculiarities of the interactions [17]. An accurate representation of $S(q)$ at small-q lies therefore on the use of large simulation boxes and a reliable potential energy function. In the present work, we have done simulations with a system containing 6912 particles and 12 layers distributed on $P = 6$

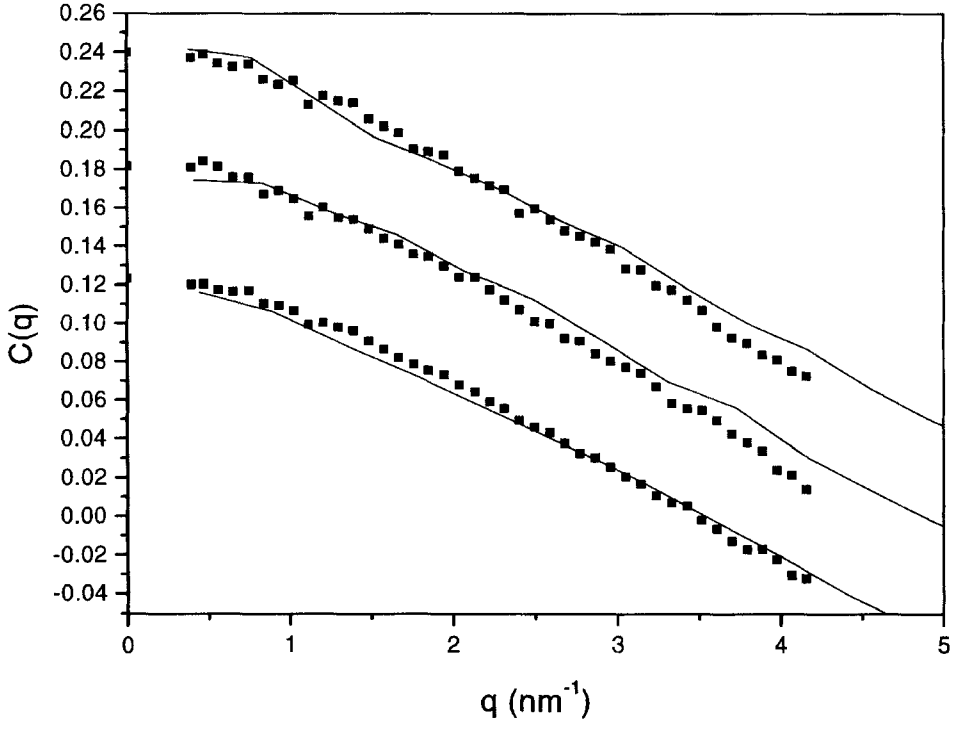

FIGURE 2 Direct correlation function $C(q)$ for $\rho = 1.52\,\mathrm{nm}^{-3}$, $\rho = 1.97\,\mathrm{nm}^{-3}$ and $\rho = 2.42\,\mathrm{nm}^{-3}$ from the top to the bottom (the curves corresponding to $\rho = 1.52\,\mathrm{nm}^{-3}$ and $\rho = 1.97\,\mathrm{nm}^{-3}$ are shifted upwards by an amount of 0.05 and 0.1 respectively). The full lines correspond to the MD simulations involving Aziz and Slaman's [12] two-body potential combined to the Axilrod-Teller three-body contribution. Open squares represent the experimental data of Formisano *et al.* [10].

processors, which is sufficient to extract the Fourier transform of the direct correlation function

$$C(q) = (S(q) - 1)/\rho S(q). \tag{11}$$

The simulations are conducted in the microcanonical ensemble (NVE) over 10^5 time-steps $\Delta t = 5 \cdot 10^{-15}$ s. First, the temperature of the system is maintained around the reference value $k_B T/\varepsilon = 1.4276$, corresponding to $T = 297$ K, by rescaling periodically all the velocities within the thermalization period of 20000 time-steps. Then, follows the production period during which the pair-correlation function is extracted over a sample of 8000 time-independent configurations, taken every $10\Delta t$. We have also performed a simulation with 16384 particles and $P = 8$ processors. Essentially the same results were obtained, albeit with a smaller mesh, showing that they are not sensitive to the system size, within the statistical errors.

In Figure 2, we display the curves of $C(q)$ for fluid Kr at a temperature $T = 297$ K and densities $\rho = 2.42\,\text{nm}^{-3}$, $\rho = 1.97\,\text{nm}^{-3}$, and $\rho = 1.52\,\text{nm}^{-3}$, corresponding to thermodynamic states studied by Formisano *et al.* [10] in their recent small-angle neutron scattering experiments. The calculations were carried out by using Aziz and Slaman's (AS) pair potential and the Axilrod-Teller (AT) three-body contribution given by Eqs. (7) to (9). Our results compare favorably with the experiments, demonstrating that the AT three-body term cannot be ignored. Therefore, Aziz and Slaman's pair potential combined with the Axilrod-Teller three-body term provide a reliable representation of the interactions for fluid krypton. It is worth mentioning that the AS potential contains the most important dipole–dipole terms of the multipole expansion in its attractive part as well as a contribution that takes the three-body exchange energy into account in its repulsive part. Nevertheless, the interplay between the AS and AT potentials and their respective role for the description of the structural and thermodynamic properties, are not fairly well understood.

4. SUMMARY AND CONCLUSION

The purpose of the present study was to implement a parallel algorithm suitable for a limited number of processors, in order to simulate accurately the physical properties of homogeneous dense liquids, and especially the structure factor that can be measured by scattering experiments. In such simulations, a large number of particles have to be undertaken, which poses

two major problems: (i) the calculation of the forces containing two- as well as three-body contributions, and (ii) the calculation of the pair-correlation function during the course of the simulation to avoid the storage of the huge amount of data. In a first step, we have proposed a parallel algorithm based on a spatial decomposition method [6] to ensure the linearity of the calculation of the forces with respect to the number of particles, and in this paper we have successfully included in our parallel algorithm the calculation of the pair-correlation function in addition.

As an application of our parallel program, we have presented results of the structural properties of fluid krypton along the supercritical isotherm $T = 297\,\mathrm{K}$. The good agreement found between the MD and the experiments demonstrated that a simple and accurate representation of the interactions for fluid krypton lies in the combination of Aziz and Slaman's pair potential and the Axilrod-Teller three-body contribution. Last but not least, we have confirmed that the self-consistent integral equation, and its extension to the three-body potentials studied in a previous work [18], is very accurate in this range of densities and reinforces our previous conclusions based on the pair-correlation function. Therefore, the present methodolgy has been useful to shed more light to understanding of the interactions in noble gases. Moreover, it will be a useful tool to study the structural and thermodynamic properties of covalent materials such as silicon, dominated by three-body interactions, for which recent experiments were done inthe liquid and supercooled states [19]. Work along this line is under progress.

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