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Liquid argon: The influence of three-body interactions on atomic correlations

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Molecular dynamics calculations on a two-dimensional argon liquid at 96°K have been performed taking into account three-body interactions. The three-body forces were assumed to have the Axilrod-Teller form. For the pair interaction the Lennard-Jones potential was chosen. The differences between structural and dynamical correlations which arise from calculations with and without the presence of three-body interactions have been studied. Whereas the differences between distribution functions (pair correlation function and triplet correlation function) due to the three-body forces are not significant, time correlation functions (velocity autocorrelation function and intermediate scattering function) show systematic deviations. For example, in the case of the velocity autocorrelation function, deviations up to 25% have been observed. Furthermore, the fourth moment of the scattering law is calculated for wave numbers $0.234 \leq k \leq 2.34$ Å^{-1}. Here, also, the deviations (up to 12%) from the pair-theory values are systematic. The results indicate that the determination of pair potentials from the fourth moment (a recent method by Rahman) does not give unique pair potentials for argon and argonlike systems since three-body forces are clearly reflected in the fourth moment.

I. INTRODUCTION

Liquids are systems with strong anharmonic behavior. Even near the melting point the anharmonicities cannot be treated as small perturbations. Besides the neutron inelastic-scattering experiments, molecular dynamics (MD) calculations are important in studying liquid many-particle systems, since anharmonicity is treated without approximations. Neutron inelastic-scattering experiments and MD calculations on liquid argon have led to a remarkable increase in our understanding of the correlations in space and time in that liquid.

The coherent and incoherent scattering functions[1] were measured by Sköld *et al.*[2] using the neutron inelastic-scattering technique. Sköld *et al.* compared the experimental results to the molecular dynamics results by Rahman[3] and Levesque and Verlet.[4] The MD calculations were based on the assumption that a *two-body central force interaction* can give a reasonable description of the liquid-argon system. In these calculations *many-body forces* (three-body forces, etc.) were not considered. Comparison of the correlation functions determined experimentally to the respective data obtained by MD indicate that the pair theory describes structural and dynamical correlations well in the first approximation.

However, it is shown by Barker *et al.*[5] that three-body interactions are distinctly reflected in thermodynamic properties (pressure, energy) of liquid argon. For example, at a temperature of 100 K and a density of 1.359 g/cm^3 the contribution of the three-body terms to the pressure is about 50% and to the energy 5%. Barker *et al.* included three-body effects with success using a perturbative technique. The three-body interaction was assumed to have an *Axilrod-Teller form.*[6] Barker *et al.* did not investigate the influence of three-body interactions on the correlation functions mentioned above. To show such effects *without* using approximate theories, it is necessary to perform MD calculations in the presence of three-body interactions.

Our purpose in this paper is to demonstrate how this type of interaction (we have also used the Axilrod-Teller potential) is reflected in structural and dynamic correlations using MD. In this case, the computer time requirements are very large. Therefore we have restricted ourselves to a two-dimensional system consisting of 144 particles. Such a small system does not give relevant results for the absolute values of correlation functions. We would like to study here the differences between structural and dynamic correlations which arise from calculations with and without three-body interactions. The correlations investigated were (i) the pair correlation function and the triplet correlation function, (ii) the velocity autocorrelation function, and (iii) the intermediate scattering function and the fourth moment of the scattering law.

II. POTENTIALS OF INTERACTIONS AND THE MD MODEL

The Hamiltonian used in the calculation is given by

$$H=\sum_{i=1}^{N}\frac{p_i^2}{2m}+U_2+\lambda U_3 \qquad (1)$$

with

$$U_2 = \sum_{i<j=1}^{N} \upsilon_2(i,j) \qquad (2)$$

and

$$U_3 = \sum_{i<j<l=1}^{N} \upsilon_3(i,j,l), \qquad (3)$$

λ being an on-off parameter with values 0 and 1. For the pair interaction υ_2 we have chosen the Lennard-Jones potential, using the same parameters as Rahman in his MD calculation[7]:

$$\upsilon_2(i,j) = 4\epsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right], \qquad (4)$$

where $\sigma = 3.4$ Å, $\epsilon/k_B = 120$ K, and r_{ij} is the distance between the particles i and j. For the three-body interaction υ_3 we have used the Axilrod-Teller form[6] (the same potential was used by Barker *et al.* in the perturbation theory):

$$\upsilon_3(i,j,l) = \nu\,\frac{1 + 3\cos\theta_1\cos\theta_2\cos\theta_3}{r_{ij}^3 r_{il}^3 r_{jl}^3}. \qquad (5)$$

r_{ij}, r_{il}, r_{jl} and θ_1, θ_2, θ_3 are the sides and angles of the triangle formed by the particles i,j,l.

It is well known that the $1/r^6$ term in Eq. (4) can be obtained by applying the perturbation theory to the second order. If the perturbation calculation is extended to the third order, interactions between triplets of particles appear. The calculation of the third-order interaction energy[6] leads to the expression given by Eq. (5). $\upsilon_3(i,j,l)$ can be repulsive and attractive, which depends on the shape of the triangle formed by the three atoms i, j, and l. The parameter ν was chosen to be 73.2×10^{-84} cm^9 erg. The investigations by Barker *et al.* indicate that this value is realistic for argon.

For the MD model, $N = 144$ argon particles were arranged in a two-dimensional box such that its density corresponds to that of the three-dimensional argon liquid ($\rho = 1.374$ g/cm^3). To avoid surface effects, periodical boundary conditions were imposed on the system. For this model, the classical Hamilton equations were solved by iteration.

Using Eq. (1) for the Hamiltonian, two MD calculations have been performed. One with ($\lambda = 1$) and another without ($\lambda = 0$) three-body interactions. Both calculations were made for $T = 96$ K.

III. PAIR DISTRIBUTION FUNCTION AND TRIPLET CORRELATION FUNCTION

In a canonical ensemble, distribution functions are defined by (e.g., see Ref. 8)

$$g_\lambda(\vec{r}_1, \ldots, \vec{r}_n) = \frac{N!}{\rho^n (N-n)!} \times \frac{\int \cdots \int d\vec{r}_{n+1} \cdots d\vec{r}_N \exp(-H_\lambda/k_B T)}{\int \cdots \int d\vec{r}_1 \cdots d\vec{r}_N \exp(-H_\lambda/k_B T)}, \qquad (6)$$

where $\vec{r}_i$ is the position vector of the ith particle. First we would like to consider three-body effects by use of a perturbative technique involving a Taylor expansion in powers of the three-body coefficient ν; the result is

$$\begin{aligned} g_1(\vec{r}_1, \ldots, \vec{r}_n) &= g_0(\vec{r}_1, \ldots, \vec{r}_n) + \nu\left[\frac{\partial g_1(\vec{r}_1, \ldots, \vec{r}_n)}{\partial \nu}\right]_{\nu=0} + \cdots \\ &= g_0(\vec{r}_1, \ldots, \vec{r}_n) + \left[\frac{\langle U_3\rangle_0}{k_B T} g_0(\vec{r}_1, \ldots, \vec{r}_n) - \frac{N!}{\rho^n (N-n)!}\,\frac{\int \cdots \int d\vec{r}_{n+1} \cdots d\vec{r}_N (U_3/k_B T)\exp(-H_0/k_B T)}{\int \cdots \int d\vec{r}_1 \cdots d\vec{r}_N \exp(-H_0/k_B T)}\right] + \cdots, \end{aligned} \qquad (7)$$

where $\langle\;\rangle_0$ denotes averaging with only two-body interactions. From Eq. (7) it follows that the average of any quantity X, which is dependent on the coordinates of the N particles, can be determined in terms of two-body averages:

$$\langle X\rangle_1 = \langle X\rangle_0 + \frac{\langle U_3\rangle_0}{k_B T}\langle X\rangle_0 - \frac{\langle X U_3\rangle_0}{k_B T} + \cdots. \qquad (8)$$

In order to estimate the influence of the correction terms in Eq. (8), we have computed and listed in Table I, as examples, the MD results for the energies of the mean-square forces. The statistical errors of the values computed is less than 0.3%. It can be seen that the corrections due to the three-body forces are relatively small and we, therefore, do not expect large differences between $g_1(\vec{r}_1, \ldots, \vec{r}_n)$ and $g_0(\vec{r}_1, \ldots, \vec{r}_n)$. Furthermore, it can be seen from Table I that in our two-dimensional system the contribution of the three-body interaction to the internal energy is about 2.8%. It

TABLE I. Energies and mean-square forces.

	$\langle U_2\rangle_\lambda/Nk_BT$	$\langle U_3\rangle_\lambda/Nk_BT$	$\langle K^2\rangle_\lambda$ (10^{-10} g cm^2 sec^{-4})
$\lambda = 1$	−3.0439	0.05529	1.31
$\lambda = 0$	−3.0145	0.05545	1.35

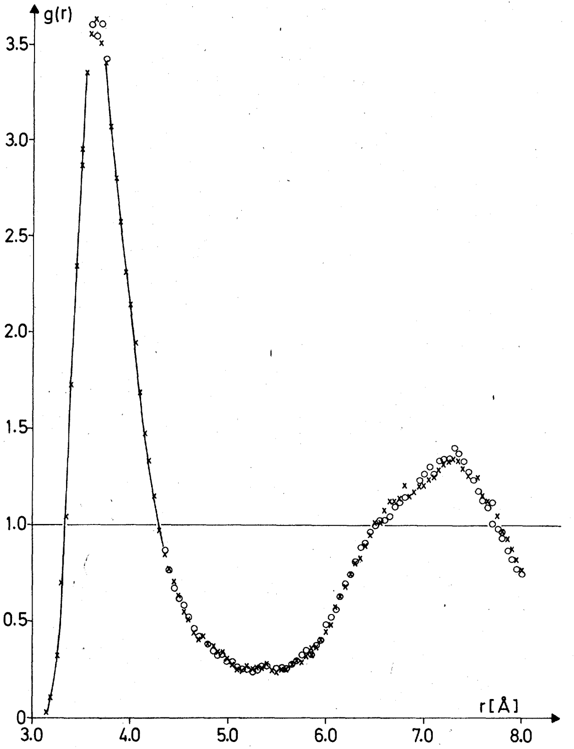

FIG. 1. Pair correlation function: crosses, MD results for $\lambda = 0$; open circles and solid curve, MD results for $\lambda = 1$.

is easy to show that in the case of a three-dimensional system this effect must be larger. As already mentioned, Barker *et al.*[5] obtained in their calculations 5%.

MD results for the pair distribution function and the triplet correlation function are plotted in Fig. 1 and Fig. 2, respectively. Since liquid argon can be considered as an isotropic and homogeneous system, both functions only depend on the relative distances r, s, and Δ of the particles from each other. We have chosen $g_\lambda(r = 3.7\ \text{Å}, s = 3.7\ \text{Å}, \Delta)$ as an example because we believe that this configuration is of particular importance in the calculations of some liquid properties and is the configuration with the best statistics (less than 5%). The statistical error of $g(r)$ is smaller than 1%. $r = 3.7$ Å corresponds to the principal peak in $g(r)$. It can be seen from Fig. 1 and Fig. 2 that there are no significant differences between the distribution functions. An extra investigation for the structure factor [the Fourier transform of $g(r)$] leads to the same conclusion. The results represented in Table I, Fig. 1, and Fig. 2 give a new justification for the application of the perturbation technique introduced by Barker *et al.*[5]

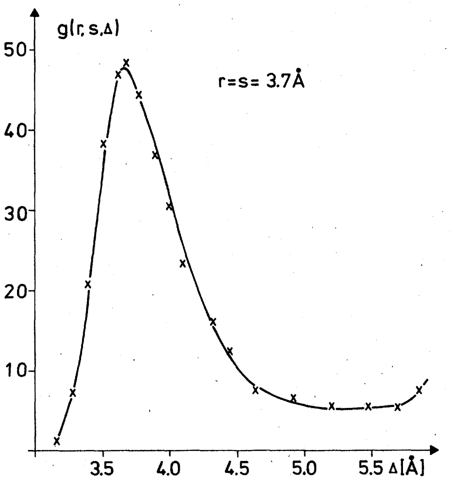

FIG. 2. Triplet correlation function: crosses, MD results for $\lambda = 0$; solid curve, MD results for $\lambda = 1$.

The relatively small influence of three-body forces on $g(r)$ and $g(r, s, \Delta)$ is of fundamental interest since all thermodynamic functions essentially depend on the shape of $g(r)$ and $g(r, s, \Delta)$. Another important point should be mentioned: There exist several approximations (e.g., Percus-Yevick,[9] hypernetted-chain,[10] Born-Green,[11] self-consistent method using MD[12]) which make it possible to calculate the pair potential $\upsilon(r)$ from $g(r)$. Besides other conditions these methods are based on the assumption that three-body interactions are not reflected in the distribution functions $g(r)$ and $g(r, s, \Delta)$. This condition could not be verified until now. Our results indicate that $g(r)$ and $g(r, s, \Delta)$ are nearly independent of υ_3 and justify this assumption (at least for argon and argonlike systems).

IV. VELOCITY AUTOCORRELATION FUNCTION (VAF)

The VAF $\psi(t)$ is defined by

$$\psi(t) = \langle \vec{v}(0) \cdot \vec{v}(t) \rangle / \langle \vec{v}(0)^2 \rangle, \tag{9}$$

where $\vec{v}(t)$ is the velocity at time t for one atom of the ensemble and the brackets $\langle \cdots \rangle$ denote a statistical average. The VAF is of interest not only because of the central role it plays in the theory of self-diffusion and particle motion, but also because of its significance in the determination of the incoherent scattering law $S_s(k, \omega)$.[13]

Figure 3 shows the MD results for the VAF obtained from calculations with $\lambda = 0$ and $\lambda = 1$. In both cases, the statistical error of $\psi(t)$ is smaller than 1%. It can be seen that the effect of the three-body interactions is relatively large. For example, the depth of the VAF for $\lambda = 0$ at the first minimum ($t = 0.36 \times 10^{-12}$ sec) is 25% larger than the depth of the VAF for $\lambda = 1$. This means that the presence of the three-body interactions leads to a less-

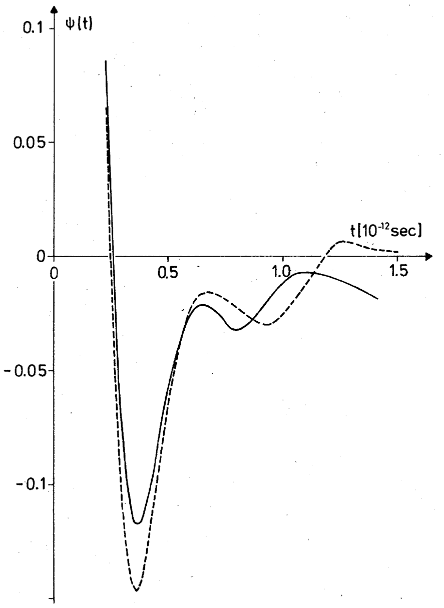

FIG. 3. Velocity autocorrelation function: solid curve, MD results for $\lambda = 1$; dashed curve, MD results for $\lambda = 0$.

pronounced oscillatory behavior of an atom in a many-particle system like argon. The diffusion constant D is given by[14]

$$D = \frac{k_B T}{m} \int_0^\infty \psi(t)\, dt. \tag{10}$$

We have found that the diffusion constant is nearly independent of three-body interactions (within the error limits). An additional investigation of the mean-square displacement $\langle r^2(t)\rangle$ leads to the same result. In this case, the diffusion constant is given by the slope of the linear part of $\langle r^2(t)\rangle$. From this, one finds the diffusion constant to be 2.5×10^{-5} cm^2 sec^{-1}.

V. DENSITY FLUCTUATIONS

The Fourier transform of the microscopic number density of a system with N particles having positions $\vec{r}_i(t)$, where $i = 1, \ldots, N$, is given by

$$\rho_k(t) = \frac{1}{N^{1/2}} \sum_{i=1}^{N} \exp[i\vec{k}\cdot\vec{r}_i(t)]. \tag{11}$$

To describe density fluctuations, the correlation function (intermediate scattering function)

$$F(k,t) = \langle \rho_{-\vec{k}}(u)\rho_{\vec{k}}(t+u)\rangle \tag{12}$$

is of interest. In Eq. (12), $\langle\cdots\rangle$ denotes again statistical averaging. In a liquid, the intermediate scattering functions depends on $k = |\vec{k}|$. $F(k,t)$ is also independent of the initial conditions u. The averaging over u and vectors $\vec{k}$ with magnitude k is done such that the statistical error of $F(k,t)$ is always less than 0.5%. The estimation of the statistical error was based on the analysis given in Refs. 15 and 16.

In Fig. 4 we have plotted the normalized intermediate scattering function $F(k,t)/F(k,0)$ for several values of k as a function of time. The values $k = 1.75$ and 4.8 Å^{-1} span the whole region of structural detail in the structure factor $S(k) = F(k,0)$. It can be seen that the effect of three-body forces of $F(k,t)$ are negligible for times $t < t_h$, where t_h is the half-width of $F(k,t)/F(k,0)$, and are getting large with increasing time t. To show these sys-

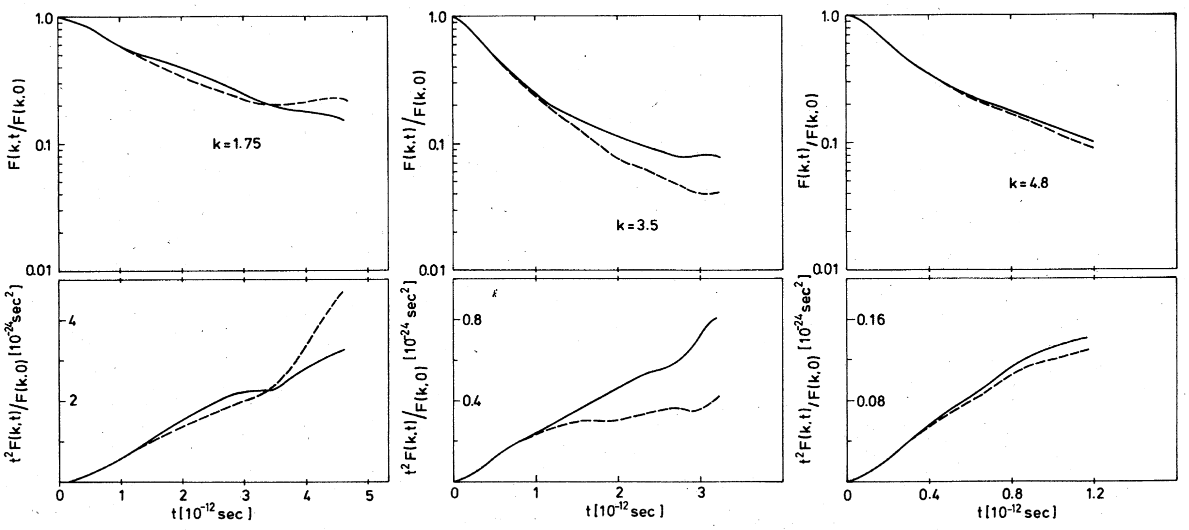

FIG. 4. Intermediate scattering function: solid curve, $F(k,t)$ and $t^2F(K,t)$ for $\lambda = 1$; dashed curve, $F(k,t)$ and $t^2F(k,t)$ for $\lambda = 0$.

tematic deviations, the values need to be plotted on a log scale. Such tendency means that υ_3 will be most reflected in the scattering law[13]

$$S(k,\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp(i\omega t)F(k,t)\,dt \qquad (13)$$

for frequencies ω which are close to zero.

The MD results are only valid[17] for times $t\leq L/c$, where L is the size of the box and c is the velocity of sound in the system. In the case of liquid argon, L/c takes the value 4.5×10^{-12} sec. Thus the behavior of $S(k,\omega)$ for small frequencies ω cannot be reliably determined from the MD results alone. In order to calculate $S(k,\omega)$ by means of Eq. (13), we have to extrapolate the results for $t>4.5\times10^{-12}$ sec. This cannot be made without additional assumptions on $F(k,t)$. Because the long-time behavior is too uncertain, we hesitate to draw any conclusions for $S(k,\omega)$ from the MD data. The solution of this problem is to use very large systems. In the future, with larger computer memories and greater speeds, it will become possible to have precise numerical data to solve this question.

The relatively large influence of three-body interactions on $F(k,t)$ at large times t suggests that the function $t^2F(k,t)$ will reflect three-body forces much more than $F(k,t)$. This is demonstrated in Fig. 4. In contrast to $F(k,t)$, the absolute values of $t^2F(k,t)$ are large in the time region of interest [note that the values of $t^2F(k,t)$ are not plotted on a log scale]. The Fourier transform of $t^2F(k,t)$,

$$R(k,\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp(i\omega t)t^2F(k,t)\,dt, \qquad (14)$$

is given by the second derivative of the scattering law $S(k,\omega)$:

$$R(k,\omega)=-\frac{d^2}{d\omega^2}S(k,\omega). \qquad (15)$$

A way of learning something about three-body forces is shown by the following method. (i) Give a model for $R(k,\omega)$ in terms of υ_2 and υ_3. (ii) Determine the pair potential υ_2. Since υ_3 is hardly reflected in structure data (see Figs. 1 and 2), a unique pair potential should be determined from the pair correlation function.[9-12] (iii) Measure $R(k,\omega)$ with high accuracy by scattering experiments. It should be mentioned that besides $R(k,\omega)$, other functions [e.g., $S(k,\omega)/\omega^2$] are qualified for the investigation of three-body forces.

VI. FOURTH MOMENT OF THE SCATTERING LAW

Often-used models for $S(k,\omega)$ are based on generalized hydrodynamics and the extension of theories of the gaseous and solid states. These developments have built up a sound general understanding of the scattering law. Most of the models involve the knowledge of moments $\langle\omega^n\rangle(k)$ of $S(k,\omega)$:

$$\langle\omega^n\rangle(k)\equiv\int_{-\infty}^{\infty}S(k,\omega)\omega^n\,d\omega,\quad n=0,1,2,\ldots. \qquad (16)$$

In a classical system all odd moments vanish.

A great deal of models have been listed and tested (using neutron-scattering data) for liquid argon by Rowe and Sköld.[18] Testing of models using our MD data lies out of the scope of this paper. Here we only want to discuss some general features. In the description of $S(k,\omega)$ the moments up to four play an important role (see Ref. 18). In some cases it appears that the knowledge of these moments is sufficient to account for the behavior of $S(k,\omega)$ or its Fourier transform $F(k,t)$ (e.g., the model by Lovesey[19] and the model by Pathak and Singwi[20]). One purpose of the calculations presented here is to investigate (i) how three-body forces are reflected in the moments up to four, and (ii) whether it is sufficient to express $F(k,t)$ and the more υ_3-sensitive quantity $t^2F(k,t)$ in terms of these moments.

Discussion relating to (*i*). It has already been mentioned above that $\langle\omega^0\rangle(k)=S(k)$ does not depend on υ_3. Furthermore, in simple liquids the second moment $\langle\omega^2\rangle(k)=k^2k_BT/m$ is not dependent on any interaction (see Ref. 21). $\langle\omega^4\rangle(k)$ is the first moment which involves explicitly the interaction potential. It is given in terms of the potential energy $U=U_2+\lambda U_3$ by[21]

$$\langle\omega_\lambda^4\rangle(k)=3\left(\frac{k_BT}{m}k^2\right)^2+\frac{k^2}{m^2}\left\langle\frac{1}{N}\sum_{i,j}\frac{\partial U}{\partial x_i}\frac{\partial U}{\partial x_j}\times\exp[ik(x_i-x_j)]\right\rangle. \qquad (17)$$

We have calculated $\langle\omega_\lambda^4\rangle(k)$ for $0.234\leq k\leq2.343$ Å^{-1}. To show that the deviations of $\langle\omega_1^4\rangle(k)$ from $\langle\omega_0^4\rangle(k)$ are systematic, we have plotted in Fig. 5 the quantity

$$\alpha(k)=[\langle\omega_1^4\rangle(k)-\langle\omega_0^4\rangle(k)]/\langle\omega_0^4\rangle(k).$$

The statistical error of both functions computed is less than 0.5%. It can be seen from Fig. 5 that the presence of three-body interactions leads to deviations up to 12%. With increasing k, the first term of the right-hand side of Eq. (17) is getting large compared to the second term involving the interaction. Therefore the deviations of $\langle\omega_1^4\rangle(k)$ from $\langle\omega_0^4\rangle(k)$ are small for large values of k.

Discussion relating to (*ii*). We have calculated some $F(k,t)$ for wave vectors k for which

$$\langle\omega_1^4\rangle(k)=\langle\omega_0^4\rangle(k).$$

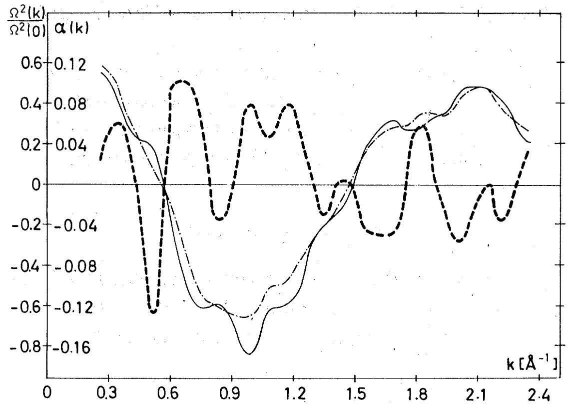

FIG. 5. Fourth moment of the scattering law: dashed curve, $\alpha(k) \equiv [\langle\omega_1^4\rangle(k) - \langle\omega_0^4\rangle(k)] / \langle\omega_0^4\rangle(k)$; solid curve, $\Omega^2(k)/\Omega^2(0)$ for $\lambda = 1$; dot-dashed curve, $\Omega^2(k)/\Omega^2(0)$ for $\lambda = 0$.

For those values of k (e.g., $k = 1.75$ Å^{-1}; see Fig. 5), three-body interactions should not be reflected in $F(k,t)$ if other quantities depending on interaction do not play an important role in the determination of $F(k,t)$. However, the example for $k = 1.75$ Å^{-1} (see Fig. 4) shows distinctly the presence of υ_3. This means that a precise description of $F(k,t)$ must involve, besides the moments, up to four other quantities (e.g., higher-order moments[22-24]) depending on the interaction.

It should be mentioned that Rahman[25] has recently proposed a method for the determination of pair potentials from $S(k)$ and $\langle\omega^4\rangle(k)$. In this investigation the quantity

$$\Omega^2(k) = \frac{1}{mk_BT}\left\langle \frac{1}{N} \sum_{\substack{i,j \\ i\neq j}} \frac{\partial U}{\partial x_i}\frac{\partial U}{\partial x_j} \exp[ik(x_i - x_j)]\right\rangle \tag{18}$$

is of main interest. We have calculated $\Omega^2(k)$ for $\lambda = 0$ and $\lambda = 1$. Figure 5 shows the results for the normalized quantity $\Omega^2(k)/\Omega^2(0)$ for both cases. It can be seen that the deviations of the two curves from each other are systematic. As for $\alpha(k)$, the presence of three-body forces lead to an additional oscillatory component in $\Omega^2(k)/\Omega^2(0)$. The amplitude of this component is relatively large for some values of k. For example, at the first minimum of $\Omega^2(k)/\Omega^2(0)$, the absolute value is, in the case of $\lambda = 1$, about 30% larger than for $\lambda = 0$. We conclude that the method proposed by Rahman cannot give a unique pair potential for argon and argonlike systems since three-body interactions are clearly reflected in $\Omega^2(k)$.

Expression for the fourth moment. It is easy to derive an expression for the fourth moment in terms of both the pair potential υ_2 and the triplet potential υ_3. Besides the pair correlation function $g(r)$ the triplet correlation function $g_3(r,s,t)$ has to be known. From Eq. (17) one obtains

$$\langle\omega_1^4\rangle(k) = \langle\omega_0^4\rangle(k) + \frac{k^2}{m^2}k_BT\rho^2 \times \int\int d\vec{r}\,d\vec{s}\,g_3(r,s,t) \times (1-\cos kx)\frac{\partial^2\upsilon_3(1,2,3)}{\partial x^2}, \tag{19}$$

where $\langle\omega_0^4\rangle(k)$ is the pair-theory value

$$\langle\omega_0^4\rangle(k) = 3\left(k^2\frac{k_BT}{m}\right)^2 + \frac{k^2}{m^2}k_BT\rho \times \int d\vec{r}\,g(r)\,(1-\cos kx)\frac{\partial\upsilon_2(1,2)}{\partial x^2}. \tag{20}$$

The main difficulty in estimating $\langle\omega_1^4\rangle(k)$ from Eq. (19) is the lack of experimental information about the triplet correlation function. The measurement of the pressure dependence of $g(r)$ makes it possible to test different expansions for $g_3(r,s,t)$, but its direct determination is not possible in this way.[26-28] Although some molecular dynamics and Monte Carlo calculations about $g_3(r,s,t)$ do exist[29-32] for liquid argon and liquid rubidium,[33] the data are not sufficient to solve Eq. (19) numerically.

VII. SUMMARY AND FINAL REMARKS

The influence of three-body interactions on structural and dynamical correlations has been studied for liquid argon by means of molecular dynamics. The three-body forces were assumed to have the Axilrod-Teller form. The investigations by Barker *et al.*[5] indicate that this potential is realistic for argon. Whereas the differences between distribution functions (pair correlation function, triplet correlation function) due to the three-body forces are not significant, time correlation functions [velocity autocorrelation function, intermediate scattering function $F(k,t)$] show systematic deviations. Our results for $F(k,t)$ suggest that the second derivative of the scattering law $R(k,\omega) = -d^2S(k,\omega)/d\omega^2$ should be more sensitive to three-body forces than $S(k,\omega)$ and should therefore be a proper quantity to test models for three-body potentials. This can be done if a reliable model for $R(k,\omega)$ is known. Our investigations of the fourth moment of $S(k,\omega)$ indicate that such a model must involve, besides the moments up to four, other quantities (e.g., higher-order moments) depending on the interaction.

Since the pair correlation function and triplet correlation function are nearly independent of three-body interactions, a unique pair potential should be determined from these functions. Recently Abel *et al.*[34] showed that both the repulsive and the long-range part of the pair potential for liquid rubidium can be extracted successfully from structure data using an extended Born-Green equation. Also the self-consistent method[12] mentioned above should be qualified, which allows the determination of a pair potential from the pair correlation function using molecular dynamics. The method by Rahman[25] [determination of pair potentials from $S(k)$ and $\langle\omega^4\rangle(k)$] does not give a unique pair potential for argon and argonlike systems since three-body interactions are clearly reflected in $\langle\omega^4\rangle(k)$. This method should give an effective pair potential.

It should be mentioned once more that our study was made for a two-dimensional system. Effects which are marginal in three dimensions could be exaggerated in two dimensions and vice versa.

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