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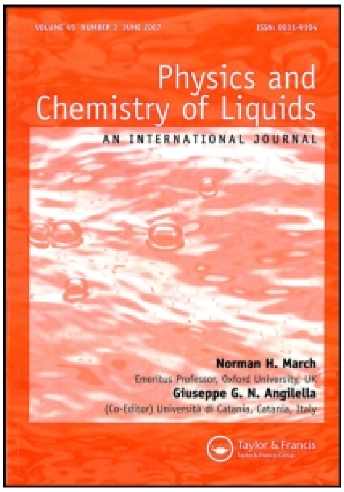

Physics and Chemistry of Liquids

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Monte Carlo Simulation of Fluid Krypton

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To cite this Article Bose, T. K. , Brostow, Witold and Sochanski, Jerzy S.(1981) 'Monte Carlo Simulation of Fluid Krypton', Physics and Chemistry of Liquids, 11: 1, 65 — 78

To link to this Article: DOI: 10.1080/00319108108079097

URL: http://dx.doi.org/10.1080/00319108108079097

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Phys. Chem. Liq., 1981, Vol. 11, pp. 65–78
0031-9104/81/1101-0065$06.50/0

Printed in the U.S.A.

Monte Carlo Simulation of Fluid Krypton

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(*Received March 13, 1981*)

The Monte Carlo procedure of Metropolis *et al.*, was used to study the states of the liquid along the saturation line as well as for densities lower and higher than those at saturation. Two different interaction potentials $u(R)$ were applied: MIST of Smith and Thakkar and the standard 6–12 potential. A number of pair radial distribution functions $g(R)$ was generated. The position of the first maximum $R_{1_{max}}$ of $g(R)$ along the saturation line shifts towards smaller values on increasing temperature. This has been found also in some diffractometric data reported in the literature and merits further experimental study. Computer-generated values of the configurational energy U^C, heat capacity at constant volume C_V and vapor pressure P are compared with experimental data. In most cases a so-called long-range correction represents an over-correction. The $u(R)$ equations used do not represent adequately properties of the liquid state. The agreement between calculation and experiment improves considerably with increasing temperature and decreasing density, i.e. going towards the gas. Some problems involved in the inclusion of multi-body contributions to $u(R)$ are pointed out.

I INTRODUCTION

Computer simulations are used more and more for correlating and clarifying properties of fluid phases, often as substitutes for theories. This makes less difference for gases, but for liquids a fundamental theory of the equilibrium properties still has to be fully developed. We believe that such a development is possible by using the formalism of the theory of information.[1–4] We also believe that a better grasp of structures of liquid phases is needed than that what has been achieved so far, and that a convenient way of characterizing irregular structures is in terms of the Voroni polyhedra.[5–8] With this philosophy, computer simulation serves mainly for the generation of configurations, and for checking various aspects of the theory.

We have devised a computer program based on the Metropolis version of the Monte Carlo procedure.[9,10] We have chosen to simulate liquid krypton, because like argon it is monatomic with spherical force fields, but has not been studied as extensively. We have used two different binary interaction potentials $u(R)$, to see whether the difference between them affects the results. Configurations have been generated for various points on the phase equilibrium diagram, as characterized in terms of values of temperature and density. We have represented the structure of the assemblies in terms of the pair radial distribution function $g(R)$. Results for some thermodynamic equilibrium properties derived from $g(R)$ are also reported here. In a separate paper, we shall relate the present work to our main scheme indicated above: the representation of liquid (or: amorphous solid) structures by the Voronoi polyhedra, and calculation of equilibrium properties from the information theory formalism.

II COMPUTATIONS

Most of our calculations were made for the basic cell of 108 atoms. This represents a reasonable optimum; for, say, 32 atoms the periodicity of the model would not have allowed extrapolations to interatomic distances of interest. On the other hand, we have made some calculations for the basic cell of 864 atoms. As expected, the results were well comparable with those for 108 atoms, while the computing times much longer. For the remainder of calculations, therefore, we have returned to 108 krypton molecules per cell.

The minimum-image distance method was used. Thus, for each pair the smallest distance between any images of the two molecules was taken as the interaction distance R. As discussed by Wood and Parker,[10] any distortions in $g(R)$ resulting from this procedure will not appear at distances less than $R_{V_{\min}}$. To describe the pair radial distribution curve, we use here the same terminology as in an earlier paper;[11] that is, the fifth minimum $R_{\mathrm{V}_{\min}}$ follows the fourth maximum $R_{\mathrm{IV}_{\max}}$ when R increases.

As already mentioned, we have used two different interaction potentials. First, we have used the all-time favorite. That is, we have taken the Mie potential—also called the Lennard–Jones potential (the first name gives the credit to whom it is due—see page 67 in Ref. 12; the second name is much more popular, however) with exponents 12 and 16:

$$u(R) = |u_{\min}| \left[\left(\frac{R_{\min}}{R} \right)^{12} - 2 \left(\frac{R_{\min}}{R} \right)^{6} \right] \tag{1}$$

where $u_{\min}$ refers to the minimum of the potential well and $R_{\min}$ is $R(u_{\min})$. We have taken $(u_{\min}/k) = 171.20$ K, where k is the Boltzmann constant; this is almost identical to the value derived from the second virial coefficient.[13] $R_{\min} = 0.40790$ nm was used,[14] equivalent to the collision diameter $R_\sigma = 0.36340$ nm. In some computations we have also used $(u_{\min}/k) = 163.10$ K,[14] with the same $R_{\min}$; those in general gave slightly worse results than the deeper potential minimum and we do not report here numerical results from this series.

The other potential we have used was MIST, proposed by Smith and Thakkar.[15,16] Incidentally, these authors have noted also that they propose a potential for krypton since sufficient work has been already done for argon. An important advantage of MIST is that it contains only one adjustable parameter. It is piece-wise analytic (with three segments) and incorporates theoretical short-range repulsion as well as the entire long-range dispersion series. It reads

$$\begin{aligned} u(R) &= 4\,|u_{\min}|\,(y^2 - y) && 0 \le R \le R_1 \\ &= \sum_{i=1}^{6} A_i (R - R_1)^{[i/2]} (R - R_2)^{[(i-1/2)]} && R_1 \le R \le R_2 \\ &= -C_6 R^{-6} \left(1 - \frac{C_8}{n C_6 R^2} \right)^{-n} && R_2 \le R \le \infty \end{aligned} \tag{2}$$

where $y = e^{C}(1 - R/R_\sigma)$, $n = 20/29$ and $[x]$ is the largest integer $\le x$. Further, $R_1 = 1.1\, R_{\min}$ and $R_2 = 1.5\, R_{\min}$. The parameters $A_i (i = 1, \ldots, 6)$ are completely determined by requiring continuity of $u(R)$, and also of the derivatives $u'(R)$ and $u''(R)$ at R_1 and R_2. The remaining parameters in (2) are: $(u_{\min}/k) = 198.55$ K; $R_\sigma = 0.35872$ nm; $R_{\min} = 0.40736$ nm; $C_6 = 126 \cdot 10^{-67}$ $\mathrm{J \cdot cm^6}$; $C_8/C_6 = 4.91 \cdot 10^{-16}$ cm^2; the Born–Mayer parameters are $A = 69.16$ $(e^2 a_0^{-1})$ and $\lambda = 1.508\, a_0^{-1}$ where e is the proton charge and $a_0 = 0.529177 \cdot 10^{-8}$ cm (Bohr radius). The values given above for the parameters are slightly different than those given in the original proposal of MIST.[15]

Calculations of the isotopic thermal diffusion factor by Aziz and Kocay[17] suggested that the slope of the repulsive wall of MIST for Kr might be too soft. Therefore, a new set of parameters has deen obtained[16,18] and this is the set we have used here.

We do not quote here the equations used in the computations of the radial distribution function $g(R)$ and of thermodynamic functions of interest, since these equations are given and discussed in detail by Wood and Parker.[10] For further discussion, we only note that in general a thermodynamic function F^{corr} is obtained as

$$F^{\mathrm{corr}} = F + F^{lr} \tag{3}$$

where F is the value obtained directly from the computer average over N configurations; F^{lr} is the long-range correction, for distances larger than those included in the Monte Carlo simulation.

III RESULTS

It was interesting to see how the radial distribution function is affected by the temperature T, and by the molar volume V. Some curves showing effects of these two parameters are shown in Figure 1 and 2. The molar volumes of the liquid saturated with vapor at equilibrium (Figure 1) were obtained by graphical interpolation of the values reported by Streett and Staveley.[19]

Experimental $g(r)$ curves for Kr at a number of temperatures have been obtained by Clayton and Heaton[20] by neutron diffraction. The curves were transferred from their graphs by using a graphic-to-digital conversion and dividing the numerical data so obtained by a factor proportional to R^2, so as to account for the change from integral distributions to our presentation. Determination of structure factors from neutron scattering is believed to be more accurate than from X-ray scattering. In general, however, diffractometric techniques involve some problems. These problems have been discussed by Yarnell *et al.*,[21] as well as by Dahlborg, Davidovic and Larsson.[22] Experimental errors might be amplified in the Fourier transformation procedure, when $g(R)$ is obtained from the structure factor. This has to be taken into account when comparing experimental and calculated $g(R)$ curves.

Figures 1–4 represent only some examples of the $g(R)$ curves we have generated. Similarly, in Table I we list only some of the sets of thermodynamic functions obtained. We have listed typical data and not necessarily the best ones. Whenever possible, comparison with experimental data of Streett and Staveley[19] is provided. These authors have obtained the most

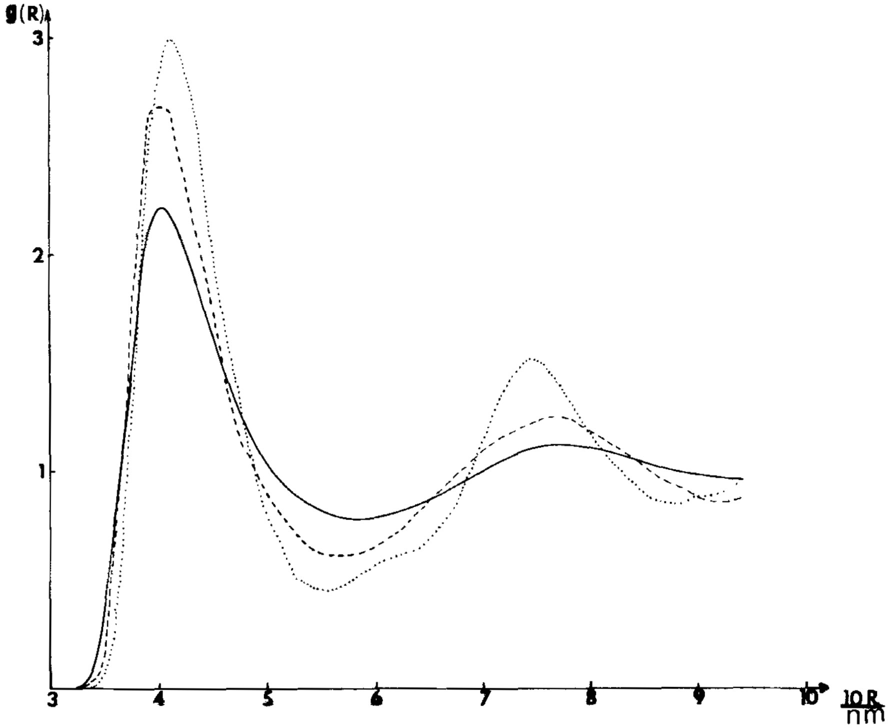

FIGURE 1 The radial distribution functions $g(R)$ for the MIST potential at T = 117.0 (dotted line), 133.0 (broken line) and 183.0 K (continuous line). The molar volumes are those of the respective saturated liquids, that is 34.22, 36.28 and 46.32 cm^3/mol.

extensive and reliable collection of thermodynamic functions for liquid and gaseous Kr. For a given temperature, molar volume and interaction potential, we have listed the values of configurational energy U^C, heat capacity at constant volume C_V, and vapor pressure P. Quantities with the superscript corr and without a superscript are defined by Eq. (3).

IV DISCUSSION

In order to apply existing theories, non-monoatomic molecules are often treated as "argon-like" or "quasi-spherical." Even when the molecular shape gives some basis for the claim of sphericity, the force fields might be fairly complex. For instance, when dealing with the SF_6 molecule, hexadecapoles have to be taken into consideration.[23] In the case of krypton, however, we are dealing with molecules which indeed do interact via spherical force

TABLE I

Thermodynamic functions of krypton

$\frac{T}{\mathrm{K}}$	$u(R)$	$\frac{V}{\mathrm{cm^3\ mol^{-1}}}$	$\frac{-U^{C_{\mathrm{corr}}}}{\mathrm{J\ mol^{-1}}}$	$\frac{-U^{C}}{\mathrm{J\ mol^{-1}}}$	$\frac{-U^{C_{\mathrm{exper}}}}{\mathrm{J\ mol^{-1}}}$	$\frac{C_V}{\mathrm{J\ mol^{-1}}}$	$\frac{C_V^{\mathrm{exper}}}{\mathrm{J\ mol^{-1}}}$	$\frac{P^{\mathrm{corr}}}{\mathrm{J\ cm^{-3}}}$	$\frac{P}{\mathrm{J\ cm^{-3}}}$	P / J c
7.0	MIST	34.22	9300	8922	8138	15.2	8.4	−104.3	−82.0	
.0	MIST	36.28	8208	7873	7657	7.2	8.8	−28.7	−10.0	
.0	MIST	39.10	7432	7131	6966	7.0	6.7	−17.2	−2.3	
.0	MIST	46.32	6062	5857	5774	5.0	5.4	−12.3	−3.3	
.0	MIST	80.30	3728	3660	3975	4.7		2.1	3.9	
.0	MIST	55.50	5063	4921		4.9		0.9	6.1	
.0	MIST	38.50	7165	6868		6.2		45.9	61.6	6
7.0	6–12	34.22	8575	7976	8209	7.8	8.4	−57.9	−28.9	
.0	6–12	36.28	8151	7591	7657	7.6	8.8	−12.1	18.8	
.0	6–12	39.10	7463	6982	6966	7.0	6.7	−12.2	12.5	
.0	6–12	46.32	6133	5789	5774	3.9	5.4	−5.8	9.1	
.0	6–12	80.30	3506	3391	3975	3.5		1.7	4.6	
.0	6–12	400.00	812	808		2.3		3.3	3.3	
.0	6–12	140.00	1508	1470		0.6		54.8	55.4	6
.0	6–12	200.00	1049	1031		0.4		36.0	36.2	4
.0	6–12	300.00	547	538		0.2		42.4	42.4	4
.0	6–12	400.00	432	427		0.2		30.8	30.8	3

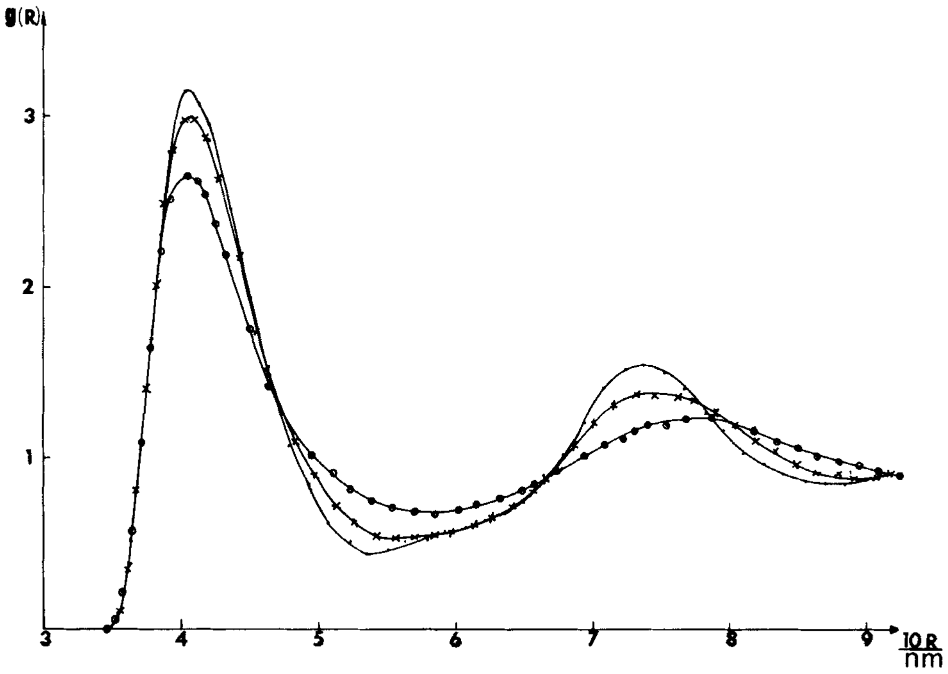

FIGURE 2 The radial distribution functions $g(R)$ for the 6–12 potential at 117.0 K, $V = 33.20$ (compressed liquid, continuous line), $V = 34.22$ (at saturation, line with crosses) and $V = 40.00\ \mathrm{cm}^3/\mathrm{mol}$ (vapor and liquid, line with circles).

fields. Some problems remain, however, and we shall now discuss them, along with some interesting features of the results.

Figure 1 shows the effect of temperature on $g(R)$. A temperature increase produces a lowering of the first peak. There is a "smearing out" phenomenon: around $R_{\mathrm{II}_{\min}}$ the curve becomes more shallow, and the second peak at $R_{\mathrm{II}_{\max}}$ becomes lower too. These observations are the expected ones. We know since the experiments on argon of Mikolaj and Pings[24,25] that the main effect of a temperature increase is a lowering of the number of nearest neighbors z_1. We also know that over the existence range of the liquid state the change in z_1 is equal to seven.[26] There is also, however, an interesting and not exactly expected feature in Figure 2: $R_{\mathrm{I}_{\max}}$ decreases along with increasing T. With this finding in mind, let us review the diffractometric data for monatomic liquids. Clayton and Heaton[20] say that there is a slight shift of what we call $R_{\mathrm{I}_{\max}}$ to higher values of R for higher temperatures. At the same time, they say that "the correctness of the results is limited by the precision of the experimental data and the validity of the corrections and analyses." Mikolaj and Pings[25] give an equation for $R_{\mathrm{I}_{\max}}$ in terms of T and

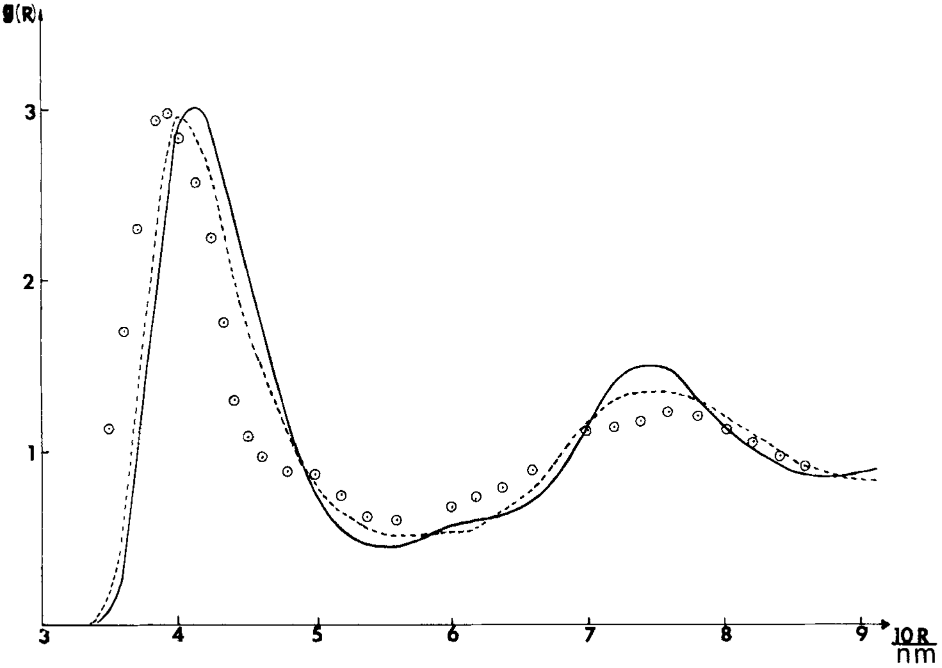

FIGURE 3 Comparison of experimental and calculated radial distribution function $g(R)$ at T = 117.0 K and saturation V = 34.22 cm^{3}mol. Circles: experimental data of Clayton and Heaton;[20] continuous line: MIST potential; broken line: 6–12 potential.

density; $R_{\mathrm{I_{max}}}$ is supposed to increase with T. On the other hand, these authors say that "the radial positions of these maxima are relatively constant, and any clear dependence on the thermodynamic state is somewhat obscured by the width of the inversion band." The Pings school has also studied liquid gallium.[27] At the lowest experimental temperature of 273.2 K they have found $R_{\mathrm{I_{max}}} = 0.283$ nm, while at their highest $T = 323.2$ K there is $R_{\mathrm{I_{max}}} =$ 0.281 nm. While this tendency agrees with our simulated $g(R)$ curves in Figure 1, the experimental uncertainty is approximately ± 0.001 nm. Thus, the shift observed is nearly within the limits of the error of the determination. Further, the same group has also studied binary mixtures of gallium and mercury.[28] Here for pure as well as for two Ga + Hg systems there is a distinct decrease of $R_{\mathrm{I_{max}}}$ when T increases. Thus, there seems to exist a *preconceived* notion that a T increase should produce an increase in $R_{\mathrm{I_{max}}}$. The experimental data and the present calculations, however, indicate that the problem at least merits further investigation.

In Figure 2 we see $g(R)$ curves obtained at a single temperature and for one interaction potential but for three densities: one at the liquid-vapor co-

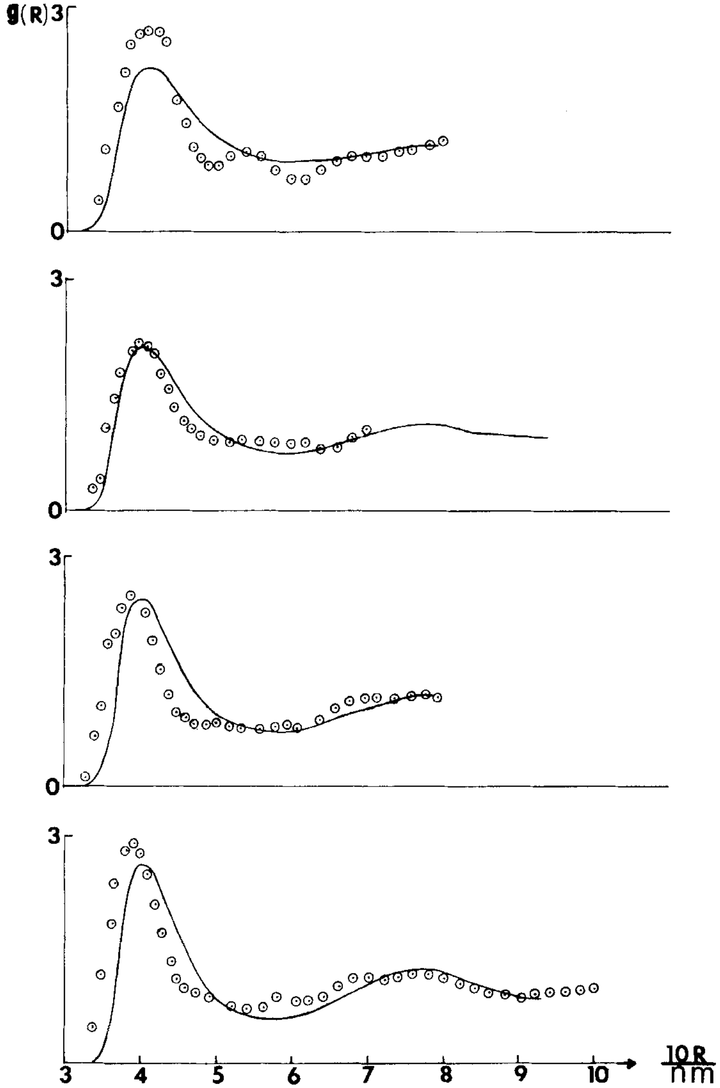

FIGURE 4 Comparison of experimental (circles are date of Clayton and Heaton[20]) and calculated with the MIST potential (continuous line) radial distribution functions $g(R)$ for $T = 133.0$, 153.0, 183.0 and 210.0 K. The diagram for the lowest temperature is at the bottom, for the highest at the top.

existence line and on both sides of the line. Qualitatively, results of a decrease in ρ are similar to that of an increase in T. We note that even a relatively small increase in molar volume V, from 33.20 to 34.22 cm^3 mol^{-1}, but from a compressed liquid to a saturated liquid, produces sizeable effects. The outward shift of $R_{\mathrm{II}_{\mathrm{max}}}$ with increasing V is particularly pronounced.

As mentioned in the introduction, we were interested in differences between results obtained by using two different potentials. From this point of view, the results shown in Figure 4 are rather disappointing. There is a distinct though not very great difference between diffractometry and the Monte Carlo simulation. At the same time, $g(R)$ curves obtained from the two $u(R)$ relations are nearly superimposable. If anything, the 6–12 potential, in spite of its simplicity, renders slightly better results. After many of the present calculations have been completed, we became aware of molecular-dynamics simulations of krypton by Vermesse and Levesque,[29] apparently performed nearly simultaneously with ours. These authors wanted also to find effects, if any, of using different $u(R)$ relations on simulation results. They have applied two potentials which they called Lennard–Jones, namely 12–6 and 9–6, and a multiparameter potential of Barker *et al.*[30] On the basis of simulation of the compressibility factor PV/kT and of viscosity they conclude that none of the potentials used are compatible with experiment. In our case, we bear in mind fairly wide margins of experimental error in $g(R)$ mentioned in Section 3. The discrepancies between our computation and experiment are not very large; but they cannot be explained by errors in diffractometry alone.

Figures 3 and 4 together show us the effect of temperature on experimental and computed curves. The extent of agreement between the two kinds of curves is seen to improve considerably along with T increase. Since the experimental accuracy is not so strongly temperature dependent, apparently the MIST interaction potential is better at higher temperatures.

We shall now consider the configuration energies U^{C} listed in Table I. For both interaction potentials and at all temperatures other than 210.0 K the uncorrected U^{C} values agree better with the experiment than those where the long-range correction has been included. A detailed analysis of ways of calculating U^{lr} has been made by Wood and Parker.[10] As discussed for instance in Ref. 31 in connection with some calculations of Kohler[32] for argon, we know that interactions other than those between the nearest neighbors are important. Since the interrelation between $U^{\mathrm{C}_{\mathrm{corr}}}$, U^{C} and $U^{\mathrm{C}_{\mathrm{exper}}}$ is not the same at all temperatures, we cannot say whether the overestimate of U^{lr} is due to approximations in $u(R)$ functions only. Approximations in the correction procedure itself may play a role also.

One more point in connection with the U^{C} values is worth noting. The disagreement between calculated and experimental values is the worst at

117.0 K, the lowest temperature studied. This agrees with the observation made above for $g(R)$ functions. It would indicate that the interaction potentials used are somehow "high-temperature functions," better for higher T and/or lower ρ. The many-body interaction would thus be the culprit, a conclusion also reached by Vermesse and Levesque[29] from their molecular-dynamics simulations.

In the case of the specific heat capacity at constant volume a long-range correction has not been made. As discussed by Wood and Parker,[10] this would require the triplet correlation function. We note that for the MIST potential the worst agreement between C_V and C_V^{exper} is again at the lowest temperature.

The vapor pressure results are the most disappointing. All uncorrected values are here better than corrected ones. Quite a few computed values are negative, a phenomenon not exactly unknown in computer simulations. Meirovitch and Alexandrowicz[33] note that negative pressures are typical in the two-phase region in all calculations based on the virial theorem. Once again, there is a clear improvement if we go to higher values of T and V. In other words, we are really dealing here with vapor-phase potentials. The conclusion of Vermesse and Levesque[29] that none of the potentials used are compatible with experiment is clearly confirmed by our calculations.

V SOME CONCLUDING REMARKS

We would like now to single out two of the topics discussed above. First, we really have conflicting evidence in the matter of dependence of $R_{\mathrm{I}_{\max}}$ on temperature along the saturation line. While different behavior of different materials is not impossible, we are more inclined to believe that there is a common denominator. We recommend the problem to the attention of experimental diffractometrists.

Second, we have supported above the conclusion of Vermesse and Levesque concerning the quality of available interaction potentials $u(R)$. We have found that the effective two-body potentials are really gas-phase potentials. As for many body interactions, Vermesse and Levesque mention an improvement obtained for argon by Barker *et al.*[34] by inclusion of the triple-dipole energy of Axilrod and Teller and Muto.[35] We would like to point out that there are many third-order non-additive interaction energies that can compete strongly with the non-expanded triple-dipole energy when charge-overlap effects are important. Ree and Bender[36] have studied non-additive forces in molecular hydrogen; the Axilrod-Teller-Muto (ATM) expression turned out to be inadequate at high densities, where four-body and higher order interactions play a role. O'Shea and Meath[37,38] have conducted a more general study of the problem of non-additivity. They have concluded[38] that the ATM representation of the many-body interaction, with or without charge overlap corrections, is of doubtful validity until $R \gg R_{\min}$. We recall that $R_{\min}$ corresponds to the minimum of $u(R)$ curve, and that we have[26] $R_{\min} = R_{1_{\max}}$ at vanishing density.

Acknowledgements

This work was supported by le Ministère de l'Education du Gouvernement du Québec and the Natural Sciences and Engineering Council of Canada.

Professor Vedene H. Smith, Jr., of the Department of Chemistry, Queen's University, Kingston, Ontario, has kindly communicated to us his not yet published set of MIST potential parameters for krypton. Correspondence with Dr. Alan L. Mackay of the Department of Crystallography, Birbeck College, University of London, is acknowledged also. The numerical calculations were performed at the Centre Informatique, Université du Québec, Trois-Rivières, Québec, Canada.

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