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Atomistic versus two-body central potential models of C_{60}: A comparative molecular dynamics study

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We report on an extensive molecular dynamics investigation of two models of C_{60}. The first model is based on an effective pair, central potential obtained by integrating the interaction between two carbon atoms over the fullerene cages [L.A. Girifalco, J. Phys. Chem. **96**, 858 (1992)]. The second model explicitly takes into account the discrete, ''atomistic'' structure of the C_{60} molecules; we study two different parametrizations of the carbon-carbon interaction, one identical to that employed in the Girifalco approach, the other borrowed from previous studies on graphite [A. Cheng and M.L. Klein, J. Phys. Chem. **95**, 6750 (1991)]. We consider a temperature range spanning from 300 to 1900 K, and pressures up to 200 kbar. Results for the lattice spacing and several thermodynamic quantities, as well as for the radial distribution functions, are reported and compared among each other and with experimental data. The central pair model yields only semiquantitative predictions at typical ambient densities, whereas pressures are generally overestimated. Atomistic simulations reproduce to an overall quantitative level of accuracy the experimental C_{60} properties. A comparison is also made of the central versus the atomistic potential predictions, when using the same potential parameters in the carbon-carbon interaction. We discuss applications of the adopted modelizations to fullerene systems of current interest, as well as different strategies to optimize the values of the potential parameters.

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I. INTRODUCTION

Simulations of fullerenes and of fullerene compounds have been of great interest since the early discovery of C_{60}. The most accurate of such investigations are of course of *ab initio* type [1]; these ones, however, when applied to the solid phase of C_{60} [2] are faced with severe difficulties due to the great number of electron degrees of freedom to be taken into account.

Indeed, most simulation studies of fullerenes are based on effective pair potentials [3–9]. Such simplified interaction laws have been (or can be expected) useful for the description of a variety of physical contexts as, e.g., (i) the characterization of fullerites over wide ranges of temperatures and pressures [4,5,10,11]; (ii) the control of doping processes of fullerene crystals (see, e.g., Ref. [12]); (iii) the investigation of mixed fullerenes in the solid phase [13–16]; (iv) the determination of the high temperature phase behavior of $C_{n\geqslant 60}$ fullerenes [17–20]; (v) the description of carbon onions and ''inorganic fullerenes'' such as the metal dichalcogenides GaAs and CdSe [21].

A currently adopted effective fullerene-fullerene potential has been early derived by Girifalco [3]. Hinging on the high-temperature ($T>260$ K) chaotic rotational motion of the fullerene molecules [12], he assumed that the carbon atoms are continuously distributed over the molecular cage, approximated by a sphere, and that two carbon atoms belonging to different molecules interact via the 12-6 potential

$$V_C(r)=-A/r^6+B/r^{12}. \tag{1}$$

The integral of $V_C(r)$ over the two cages then yields the effective C_{60}-C_{60} potential in a central analytic form, with parameters A and B, and cage diameter, conveniently fixed. This approach can be applied, in principle, also to other fullerene molecules (always in the high temperature regime) as, for instance, the ellipsoidal C_{70}, provided an effective diameter of the molecular cage, is fixed. Several procedures for determining such an empirical size have been proposed by various authors [19,22–24], and computer simulation results for bulk properties of fullerites thereby determined have been published [10,11].

We shall also investigate in this work a fully ''atomistic'' representation of C_{60}, where both the shape of the fullerene molecule, and the distribution of the carbon sites over the molecular surface are taken into account [4]. In this model, carbon atoms are in a fixed position over the cage, assumed to be rigid, and their spatial distribution is directly taken from current experimental databases. Carbons on different molecules interact via the $V_C(r)$ potential, with two different choices of A and B: one corresponds to the original set proposed by Girifalco to obtain his central pair model [3]; the other one, employed by Cheng and Klein (see Ref. [4], and references therein) in their early atomistic simulations of C_{60} is obtained from a refined fit of the physical properties of graphite. This set does not reproduce correctly the well-known transition taking place in C_{60} at 260 K [12], from the high temperature orientationally disordered fcc phase to the low temperature orientationally ordered sc phase (a transition that can be obtained by endowing the carbon-carbon bonds on each C_{60} cage with fractional charges [5]). We observe that such an inability of the bare atomistic model is not relevant for the present analysis, which focuses on the fully orientationally disordered phase of C_{60}. More recent esti-

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mates of the fullerene interaction in solid C_{60} have been obtained by means of total-energy calculations [25]; also in this case, however an empirical modelization of the carbon-carbon interaction between different molecules is required, in order to accurately reproduce the experimental results.

Simulations based on the central potential are much easier to implement than atomistic ones, whereas the latter ones are in principle more realistic, and can be immediately adapted to molecules of any shape. On the other hand, the atomistic model still rests on relevant approximations as, for instance, the neglect of intramolecular degrees of freedom. We investigate the central and atomistic potentials in two complementary directions: on one side we compare our predictions with available experimental data. Such a study allows us to unravel the overall physical relevance of the model approximations, and paves the way to extensive applications of both approaches considered here. On the other side, we present a detailed comparison of the atomistic against the two-body model predictions, when the *same* parameters are employed for the carbon-carbon interaction. We gain in this way a direct physical insight into the approximations inherent the reduction of the fullerene-fullerene interaction to a single-site description.

The paper is structured as follows. In Sec. II we define the fullerene modelizations and interaction laws; details on the simulation procedure are also given. Results for thermodynamic and structural properties are reported and compared with experimental data in Sec. III. Conclusions and perspectives follow in Sec. IV.

II. MODELS AND SIMULATION PROCEDURE

The Girifalco potential [3], obtained from $V_C(r)$ as described in the Introduction, is written as

$$V_{C_{60}}(r) = -\alpha_1\left[\frac{1}{s(s-1)^3}+\frac{1}{s(s+1)^3}-\frac{2}{s^4}\right] + \alpha_2\left[\frac{1}{s(s-1)^9}+\frac{1}{s(s+1)^9}-\frac{2}{s^{10}}\right]. \quad (2)$$

Here $s=r/d$, $\alpha_1=N^2A/12d^6$, and $\alpha_2=N^2B/90d^{12}$, where $N=60$ and d are, respectively, the number of carbon atoms and the diameter of the fullerene particles. According to Girifalco, best values of the parameters of potential (2) are

$$A=32.0\times10^{-60}\ \text{erg cm}^6, \quad B=55.77\times10^{-105}\ \text{erg cm}^{12}, \quad d=0.71\ \text{nm}. \quad (3)$$

Note that A and B are the same constants entering potential (1).

In the atomistic simulations of C_{60} by Cheng and Klein [4], the carbon atoms interact through a standard 12-6 Lennard-Jones potential equivalent to the present $V_C(r)$ form with

$$A=23.8\times10^{-60}\ \text{erg cm}^6, \quad B=36.88\times10^{-105}\ \text{erg cm}^{12}. \quad (4)$$

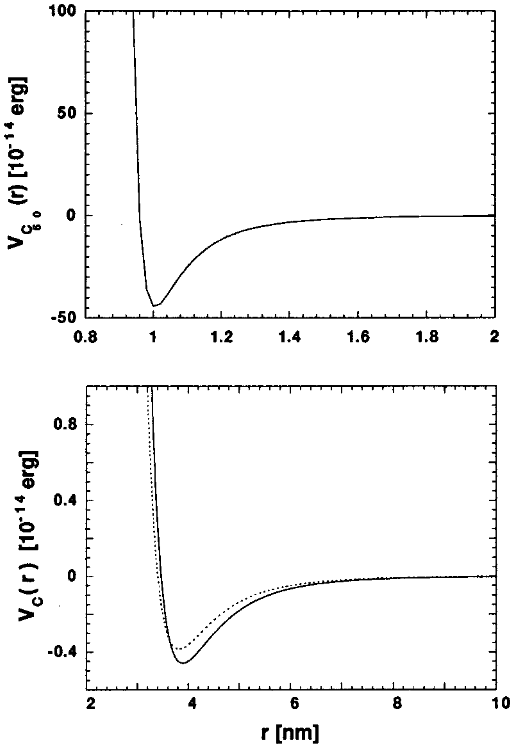

FIG. 1. Top: central, pair potential for C_{60} [Eq. (2)]. Bottom: carbon-carbon interaction [Eq. (1)] with set (3) (full line), and set (4) (dotted line). Zero-crossing distances σ, in nanometer, and potential well depths ϵ, in 10^{-14} erg, are top, [$\sigma=0.959,\epsilon=44.1$]; bottom: [$\sigma=0.347,\epsilon=0.459$, set (3)] and [$\sigma=0.340,\epsilon=0.384$, set (4)].

We show in Fig. 1 the potential $V_C(r)$ with $[A,B]$ values as in (3) and (4), along with the Girifalco potential of Eq. (2).

We have carried out molecular dynamics (MD) simulations in the canonical ensemble to study both the atomistic models based on the $[A,B]$ pairs (3) and (4), and the central potential model (2). Different number of particles have been employed, in order to detect size effects on the simulation predictions. While for model (2) we could perform this investigation on systems consisting of even several thousands particles, in the atomistic case we had to restrict, for obvious computational time reasons, to a maximum of 256 particles. We have fixed during our simulations the time step $\tau=5\times10^{-15}$ s; runs of 1×10^6 and 5×10^4 steps have been carried out for the central potential and the 60-site models, respectively.

III. RESULTS AND DISCUSSION

We shall refer in what follows to atomistic and central potential simulations with the acronyms AS and CPS, respectively. We report in Table I the dependence of thermodynamic quantities on the sample size. It appears that CPS results hardly change for $N>500$ (see also Ref. [26]), and that the $N=256$ estimates are already satisfactorily stable. As for the AS entries, the thermodynamic quantities estimated with 256 particles would hardly vary upon further increasing N. On such a basis, most of the calculations have been performed with 256 particles.

TABLE I. Size dependence of pressure and configurational energy for MD simulations at $T=300$ K and lattice spacing $a=1.42$ nm. Results concern CPS and AS with $[A,B]$ set (3). The experimental value of the sublimation energy used to fix the $[A,B]$ pair in Ref. [3] is $U\simeq-168$ kJ/mol [27].

	$\langle P\rangle$ (kbar)		$\langle U\rangle$ (kJ/mol)	
N	AS	CPS	AS	CPS
32	−0.37	1.91	−166.5	−156.7
108	−0.38	1.47	−173.9	−167.9
256	−0.40	1.39	−174.2	−169.5
500		1.37		−169.9
864		1.36		−170.1
4000		1.34		−170.3

As far as a comparison with experimental data and previous studies on the same models is concerned, we recall that in the Girifalco approach the parameters $[A,B,d]$ entering Eqs. (1) and (2) were adjusted to reproduce at $T=300$ K the experimental crystal sublimation energy at $T=707$ K, $U\simeq-168$ kJ/mol [27]. In Ref. [3] U is calculated by summing up the potential $V_{C_{60}}(r)$ over a fcc crystal region of experimental lattice spacing $a\simeq1.42$ nm [3,12], and of extension $L=20\sigma\equiv14.2$ nm. We assess this procedure by calculating the configurational energy through simulations of the central Girifalco potential. We have employed 4000 particles in a cubic box of edge $L=14.2$ nm, arranged on a fcc lattice with $a=1.42$ nm, and obtained $U=-170.3$ kJ/mol at $T=300$ K, a result which confirms the soundness of the approach adopted in Ref. [3]. As for the AS with set (4), Cheng and Klein [4] used a sample composed of 32 C_{60} molecules, whereas our calculations involve 256 particles. The comparison among the two simulations is satisfactory: in Ref. [4] the ambient pressure is reproduced at 300 K when $a=1.414$ nm, and we have obtained the same result with $a\simeq1.412$ nm. We have also $U=-138.2$ kJ/mol, a value $\sim3\%$ different from the 32-particle estimate -142.3 kJ/mol.

Results for the configurational energy and the pressure as functions of the temperature at various lattice spacings are displayed in Fig. 2. At ambient conditions, AS based on the $[A,B]$ set (3) yield $U=-174.2$ kJ/mol, whereas simulations of potential (2) predict $U=-169.5$ kJ/mol (see also Table I). At $T=707$ K, the values $U=-164.9$, -168.2, and -131.7 kJ/mol for CPS and AS with $[A,B]$ pairs (3) and (4), respectively, can be compared with the experimental data reported above, $U=-168$ kJ/mol [27]; the overestimate of the configurational energy in the parametrization (4) of $V_C(r)$ was already noted in Ref. [4].

The energy data at $a=1.42$ nm reported in Fig. 2 allow us to estimate the contribution to the specific heat at constant volume c_V due to the translational degrees of freedom for CPS, and to the translational and rotational degrees of freedom for AS. In the latter ones, because of the assumed rigidity of the molecular cages, we cannot take into account intramolecular vibrational degrees of freedom, which, as is well known, constitute the major contribution to c_V [3]. We find for $V_{C_{60}}$ $c_V=11.7$ J/(mol K) almost half the Dulong-Petit value $3R$ estimated in Ref. [3]. Atomistic simulations based on parametrization (3) yield instead $c_V=38.94$ J/(mol K), a value fairly close to $4.5R$, as early conjectured by Girifalco [3].

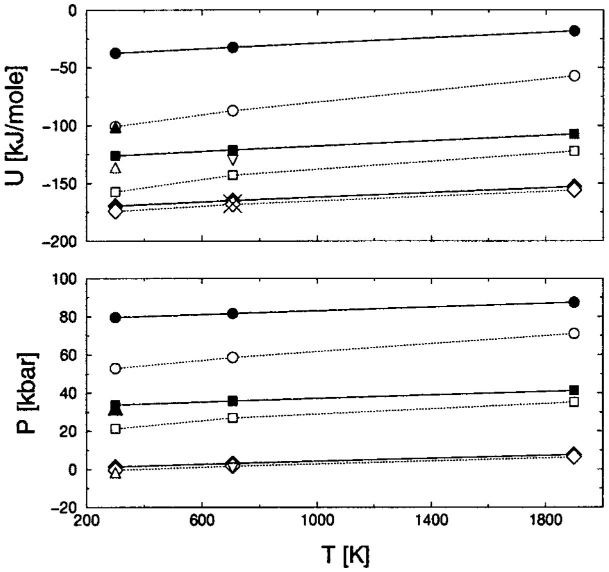

FIG. 2. Internal energy (top) and pressure (bottom) as functions of temperature for several lattice spacing a. Full symbols refer to potential (2) for $a=1.36$ nm(●), 1.38 nm (■), and 1.42 nm (◆). Open symbols refer to potential (1) with set (3) for $a=1.36$ nm (○), 1.38 nm (□), and 1.42 nm (◇). Triangles refer to potential (1) with set (4) for $a=1.36$ nm (▲), 1.4198 nm (△), and 1.42625 nm (▽). The experimental sublimation energy of Ref. [27] (×) is also reported. Lines are guides to the eye.

The comparison of simulation pressures with experimental data, reported in Fig. 3, indicates that both Girifalco models reproduce fairly well the equation of state up to 10 kbar,

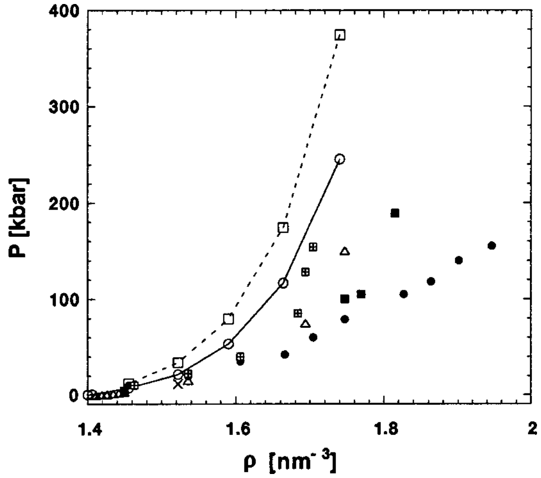

FIG. 3. Experimental and simulation pressures vs density at $T=300$ K. Symbols: two-body [Eq. (2) (□)], and atomistic [Eq. (1)] with sets (3) (○) and (4) (△) simulation results; experimental data by Duclos *et al.* [30] at 20 GPa (●), 10 Gpa (■), and 4 GPa (⊞) max hydrostatic pressure; experimental value by Fischer *et al.* [29] (×). Lines are guides for the eye.

TABLE II. Lattice spacing a (in nm) as a function of the temperature (in K) close to the atmospheric pressure (in bar).

	CPS		AS, set (3)		AS, set (4)	
T	a	$\langle P\rangle$	a	$\langle P\rangle$	a	$\langle P\rangle$
280	1.42421	4	1.4182	5	1.4112	6
300	1.4245	−9	1.4185	−2	1.4118	−7
320	1.4247	2	1.4188	15	1.4123	1

and become completely wrong beyond this region. On the other hand, as early shown in Ref. [4], AS based on set (4) provide a reasonably good approximation of the experimental data, better than it can be achieved through $[A,B]$ set (3).

We have collected in Table II simulation results for the lattice spacing (or equivalently, density) as a function of the temperature at pressures close to 1 bar. Remarkably, AS based on set (3) yield atmospheric pressure at 300 K with $a=1.4185$ nm, i.e., a lattice spacing very close to the *exact* experimental value $a=1.4198$ nm. If we also consider the above reported estimate for parametrization (4), $a=1.4118$ nm, it appears that the atomistic models are quite accurate in the prediction of the lattice spacing at ambient conditions. By converse, the central model yields in correspondence of $a\simeq 1.42$ nm (adopted in the Girifalco procedure) a pressure exceeding 1 kbar at $T=300$ K, while $P\sim 1$ bar is obtained for the slightly higher value $a=1.4245$ nm (see also Ref. [10]).

On the basis of data reported in Table II, the thermal expansivity for model (4) is 2.5×10^{-5} nm/K, in rather accurate agreement with the experimental data $2.30(5)\times 10^{-5}$ nm/K [28]. Atomistic simulations based on set (3) and CPS yield instead the qualitative results 1.5×10^{-5} and 1.62×10^{-5} nm/K, respectively.

As for the isothermal compressibility at ambient conditions, K_{T}, Fischer and co-workers [29] reported the experimental value 6.9×10^{-12} cm^2/dyne, whereas Duclos *et al.* [30] obtained a bulk modulus $B=18.1$ GPa, equivalent to $K_{\mathrm{T}}=5.6\times 10^{-12}$ cm^2/dyne. There is thus a moderate discrepancy among experimental estimates. The original Girifalco calculations for the central model yielded $K_{\mathrm{T}}=6.32\times 10^{-12}$ cm^2/dyne [3], in apparent good agreement with the estimate of Ref. [29], while direct simulations on the same model now give $K_{\mathrm{T}}=5.6\times 10^{-12}$ cm^2/dyne (see also Ref. [10]), a result reproducing instead the value reported in Ref. [30]. On the other hand, the fully atomistic model (3) yields $K_{\mathrm{T}}=7.1\times 10^{-12}$ cm^2/dyne at $T=300$ K, in excellent agreement with Ref. [29]. The Cheng and Klein model yields the relatively overestimated isothermal compressibility 9.26×10^{-12} cm^2/dyne.

The different performances of atomistic models can be qualitatively explained in terms of the characteristic features of the 12-6 carbon-carbon interaction. As visible in Fig. 1, $V_{\mathrm{C}}(r)$ with set (3) is characterized by a longer zero-crossing distance σ, and a deeper well depth ϵ, than $V_{\mathrm{C}}(r)$ with set (4). These aspects imply a reduction of core repulsive effects associated with parametrization (4) at short range, with an ensuing decrease of the pressure, and less negative configurational interaction energy.

In order to assess the approximations inherent the reduction of the multisite interaction to a two-body, central potential form, now we concentrate on a detailed comparison among the thermodynamic and structural properties obtained by CPS and AS when the $[A,B]$ pair (3) is used in the basic carbon-carbon interaction. As already observed, in the solid state regime, $T=[300–700]$ K, the configurational energy of the two-body model is systematically higher than the atomistic one (see Fig. 2); the two values approach at high temperatures, provided the density is not too high ($\rho<1.4577$ nm^{-3}). As for the pressure, the CPS value can be even dramatically higher than the AS one. In Fig. 2, this emerges in particular for $a=1.36$ nm, a lattice spacing which corresponds, in the fcc arrangement of C_{60}, to a nearest-neighbor distance of ~ 0.96 nm, i.e., the position of the zero of $V_{\mathrm{C}_{60}}$ (see Fig. 1). At this distance, a pair of fullerene molecules begins to test a very strong repulsion, which reflects in the rapid increase of both the energy and the pressure, with a large discrepancy with respect to the atomistic results. The mismatch among the two pressure estimates reduces to about 30% in passing from 300 to 700 K and persists at lower densities. If we also consider the pressure vs the lattice spacing, it emerges that the integration of the carbon-carbon interactions in the Girifalco scheme results in an overestimate of repulsive effects at short range. We recall that another central, two-body model for C_{60} has been derived in Ref. [9] from first-principles studies of the dispersion interaction among fullerene molecules. Such a model is very similar to the Girifalco potential, except for a tiny reduction of the range of the repulsive part, supporting in this respect the observations of the present analysis.

We now examine results for $T=1900$ K. It is known that at this temperature the central, pair model exhibits both a solid and a liquid phase [17], with triple and critical points quite accurately estimated [18,20]. The comparison of energy and pressure for these two phases is also reported in Fig. 2, where a fairly good agreement between CPS and AS results is documented. A substantial coincidence also emerges in the equation of state, as shown in Fig. 4. The CPS and AS results are expected to approach when the temperature increases, since the smeared out distribution of carbon sites over the spherical molecular cage (the basic Girifalco approximation) becomes more realistic under chaotic rotational motion conditions of fullerene particles. It clearly appears, however, that when the density increases the two approaches yield rather different predictions for both energy and pressure.

As far as structural predictions are concerned, we display in Fig. 5 radial distributions functions $g(r)$ of the two-body against the atomistic model at ambient temperature and at $T=1900$ K, and for pressures which, for the atomistic case, are reasonably close to ambient value. It appears that the differences among the two sets of structural functions are minor ones. Peaks in the patterns at $T=300$ K are typical of a solid configuration and are located according to the metric relationships of the fcc arrangement. All features are systematically higher in the central model $g(r)$, indicating again an overestimate of repulsive effects with respect to the atomistic case. The differences in the peak heights reduce when the

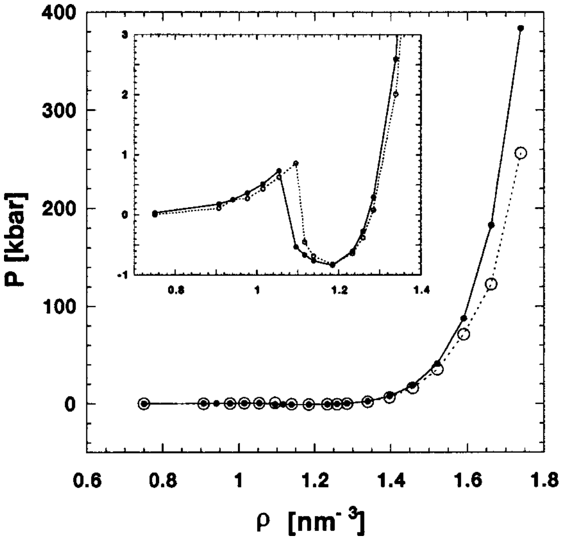

FIG. 4. Equation of state at $T=1900$ K: Girifalco potential (2) (●); AS with set (3) (○). Inset: magnification of the liquid-solid transition region.

temperature increases, and practically disappear at 1900 K (see also Ref. [17]). The close reproduction through atomistic simulations of the central model equation of state, energy, and structure, indicates that, should a complete determination of the phase diagram through the atomistic description approach be undertaken, one would likely confirm the existence of a stable liquid phase also for such a detailed representation of C_{60}.

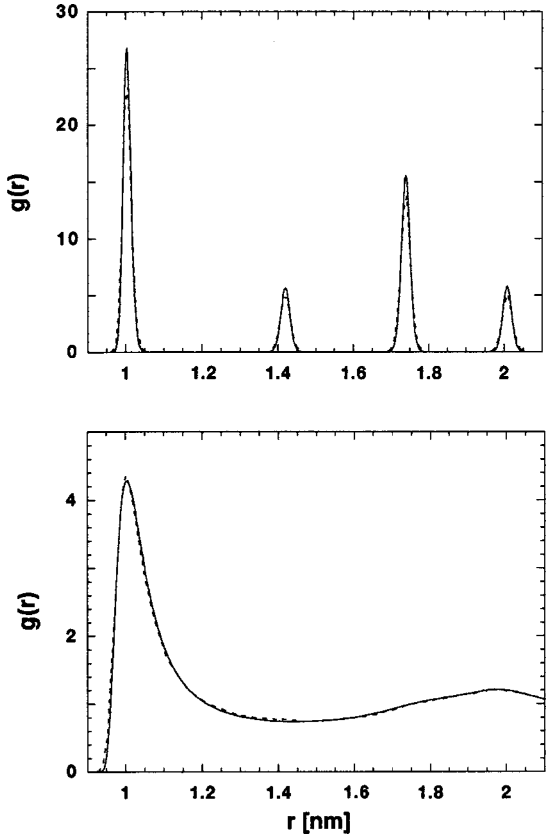

FIG. 5. Radial distribution functions at $T=300$ K in the solid phase (top) and at $T=1900$ K in the liquid phase (bottom). CPS (full lines) and AS with set (3) (dashed lines) results are shown.

IV. CONCLUSIONS AND PERSPECTIVES

We have reported the results of extensive molecular dynamics simulations of atomistic and central, pair potential models of C_{60}, in the temperature range $T=300$–1900 K, and up to pressure of 200 kbar, where the fullerene molecules are always in a fully disordered orientational state. The comparison of simulation results among each other, and with experimental data allows us to establish the suitability of the carbon-carbon potential, in order to reproduce the physical properties of real-life C_{60}, as well as to assess the approximations inherent the reduction of the multisite fullerene-fullerene interaction to a two-body, central potential form.

Fairly stable predictions of thermodynamic properties can be obtained with 256 particles (see also Ref. [26]); size effects in the envisaged simulations can be thus relatively easily overcome. Atomistic simulations based on the Girifalco parametrization (3) yield good overall predictions at ambient conditions, especially as for the lattice spacing and the internal energy. The equation of state is better reproduced by the graphite fit parameters (4), though the configurational energy is rather poorly estimated. The different performances of the two models can be explained qualitatively in terms of the characteristic features of the carbon-carbon interaction.

We also find a fair consistency with the solid state calculations originally employed by Girifalco to fix his potential parameters. Two-body and atomistic predictions with set (3) tend to overlap at high temperatures, especially for densities close to ambient conditions; by converse, substantial discrepancies manifest in the high-density regime. We deduce that repulsive effects are overestimated in the central, pair model, and conjecture that a tiny reduction of the fullerite diameter might result in an improvement of the Girifalco model in the high-density–high-pressure regime. We quote in this context the good overall performances of the Girifalco scheme in predicting the equation of state of C_{84} [11], suggesting that this approach possesses flexibility properties hitherto not fully exploited. We plan to investigate, in this instance, whether an optimal strategy can be devised to fit the potential parameters of C_{60} and higher order fullerenes, by requiring, e.g., the model to reproduce the first and second derivatives of the free energy, that is, the ambient pressure and compressibility.

Nowadays computing facilities make it conceivable to attempt a best fit of the potential parameters to the physical properties of the $C_{n \geqslant 60}$ family, within different modelizations, with specific applications, e.g., to fullerene mixtures. Indeed, the mixing properties of these materials in the solid phase are still controversially reported [13–16], and atomistic simulations could elucidate the phase behavior of such systems. Also, the multisite approach allows a realistic description of the interaction of impurities with host fullerene matrices; accurate simulations of impurity diffusion in solid fullerenes at relatively high temperatures could be useful to anticipate the effectiveness of the permeation devised, as well as to characterize the doped crystal. Of course, the smearing out of the carbon sites on the spherical cage, advocated in the two-body description, would constitute in this case a serious approximation, since it affects crucial aspects of the impurity dynamics such as selective localization in the crystal interstices, or over the molecular surface. In this context, though simple models can still be useful for a qualitative description, atomistic simulations should be considered as the primary tool for reliable investigations.

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