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What Is the Shape of the Helium Trimer? A Comparison with the Neon and Argon Trimers

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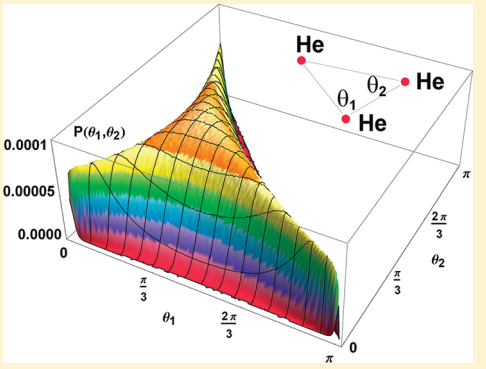
ABSTRACT: Despite its apparent simplicity and extensive theoretical investigations, the issue of what is the shape of the helium trimer is still debated in the literature. After reviewing previous conflicting interpretations of computational studies, we introduce the angle−angle distribution function as a tool to discuss in a simple way the shape of any trimer. We compute this function along with many different geometrical distributions using variational and diffusion Monte Carlo methods. We compare them with the corresponding ones for the neon and argon trimers. Our analysis shows that while Ne_3 and Ar_3 fluctuate around an equilibrium structure that is an equilateral triangle, $^4He_3$ shows an extremely broad angle−angle distribution function, and all kinds of three-atom configurations must be taken into account in its description. Classifying $^4He_3$ as either equilateral or linear or any other particular shape, as was done in the past, is not sensible, because in this case the intuitive notion of equilibrium structure is ill defined. Our results could help the interpretation of future experiments aimed at measuring the geometrical properties of the helium trimer.



INTRODUCTION

Weakly bound van der Waals noble gases clusters in the last few decades have attracted much attention from both theoreticians and experimentalists. In this family of systems probably the most intriguing one is the family of helium clusters, starting from the helium dimer. The interest origins in their highly quantum features caused by the small atomic mass and the very weak interaction potential. Due to these peculiarities, these systems can only be studied by fully quantum mechanical many particle methods. The existence of a bound state of the ^4He dimer was debated for a long time from a theoretical point of view. Starting from the '70s almost all realistic interaction potentials supported the existence of at least a bound state (see Kolganova et al.[1] for a recent review and references therein). However, it was only in 1993 that the elusive ^4He dimer was finally detected by Luo and co-workers[2] in an electron impact ionization experiment. In a subsequent and independent work, the dimer and trimer were detected by Schöllkopf and Toennies[3,4] in a diffraction experiment using a transmission grating.

The helium trimer $^4He_3$, more tightly bound than the dimer, has been the subject of theoretical studies since the early '70s.[5] Despite its apparent simplicity and the number of theoretical papers devoted to it, its properties are still not completely understood. There are a number of discrepancies or disagreements in the literature for various properties of this system that need to be settled, even in the light of helping the interpretation of future experimental work.

All modern helium−helium potentials support the existence of a single excited state. Several theoretical papers have been devoted to the investigation of the possibility that this state is a candidate for the Efimov effect, first discussed in nuclear physics. The recent paper of Kolganova et al.[1] provides a nice review of the literature of this field of research. So far, however, the experimental evidence of the existence of this state is still missing.[6]

Another debate regards the possibility that the helium trimer has states with $J > 0$. In a recent paper, Lee et al.[7] investigated the possible existence of bound states for the helium trimers with $J > 0$ using the hyperspherical coordinates method in the adiabatic approximation. For $^4He_3$, all the lowest adiabatic hyperspherical potential curves are repulsive, with no attractive well, ruling out the possibility of bound states with $J > 0$. For $^4He_2{}^3He$, the $J = 1^-$ curve has a small well, but the direct solution of the hyper-radial equation for this curve showed that this well is too shallow to support a bound state. Furthermore, they estimated that, to support a $J = 1^-$ bound state, the He−He pair potential should be made more than 20% deeper. This rules out the possibility that a different pair potential (they used the LM2M2 potential with add-on[8]), possibly including three-body terms, could support a bound state. In conclusions, no bound state with $J > 0$ was found in their work. However, a later investigation on the same system[9] reached exactly the opposite conclusion, finding at least a bound state with $J = 1$ for the $^4He_3$ system. The authors even proposed an experiment to observe such a bound state. More recent calculations[10] adopting a state-of-the-art two-body potential, including retardation effects, and three-body terms confirmed the nonexistence of bound states with $J > 0$.

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Table 1. Energies and Geometrical Structures for the Helium Trimer[a]

energy (cm^{-1})	potential	important configurations	references
−0.0872(4)	TTY	near linear contributions	Lewerenz[11]
−0.0870	LM2M2	equilateral	Nielsen et al.[12]
−0.15	LM2M2	quasilinear	González-Lezana et al.[13]
−0.1523	LM2M2	scalene + quasilinear	González-Lezana et al.[14]
−0.0876(5)	TTY	not discussed	Blume and Greene[16]
−0.0872(4)	LM2M2		
−0.0870	LM2M2	not discussed	Blume et al.[17]
−0.08784(7)	TTY	near linear contributions	Bressanini et al.[18]
−0.08785	TTY	not linear	Roudnev and Yakovlev[19]
−0.08782	LM2M2	equilateral	Barletta and Kievsky[20]
−0.08771	TTY	not discussed	Salci et al.[33]
−0.08783(4)	TTY	all kinds	this work

[a] For Monte Carlo calculations the uncertainty on the last digit is in parentheses.

There is now a general consensus on the ground state energy and most recent and accurate calculations agree with each other (see Table 1), but there are still few isolated papers reporting a much lower energy.

In this paper we try to resolve yet another controversy regarding the helium trimer: what is its preferred geometrical structure? Some authors described the helium trimer as an equilateral triangle, others pointed out that nearly linear configurations are important, and still others opted for a scalene shape, that is, a triangle with three different sides. This discrepancy about the shape of the helium trimer has been noticed in the past,[11−21] but no attempts have been made to resolve it.

What is striking looking at the literature for this problem is that most papers show convergent results and very good agreement for the energy of the ground state. Because structural properties are known to be less sensitive than the energetic ones, it is puzzling that different authors reached diverging conclusions about such a seemingly simple aspect of the helium trimer. In the following, we will show that most calculations in the literature agree with our results and that the disagreement on the shape of this system arises from a misinterpretation of the data, because for this very peculiar system any conclusion on its shape based only on average values or monodimensional distributions can lead to misleading interpretations. Because neon and argon trimers are not controversial in the literature, they are studied here as a comparison and to show the peculiar features of the distribution functions of the helium trimer. The approach to compare trimers of different atoms has been already used in the past[14,17,22] using different computational methods.

LITERATURE DISCUSSION

In this section, we briefly review the published papers on the helium trimer that explicitly deal with its structural properties.

In his study of small pure helium clusters, Lewerenz[11] used the TTY potential[23] and the older HFD-B(He) potential to compare the results. In the density distribution of the atoms with respect to the center of mass (c.o.m.), he noticed a slight increase toward zero. He interpreted this as a certain preference of the helium trimer to be in a nearly linear geometry, which brings one of the atoms very close to the center of mass of the system. The reasoning was that if the cluster fluctuates tightly around an equilateral triangle structure, the radial density distribution at the origin should not have a maximum, but should be rather close to zero.

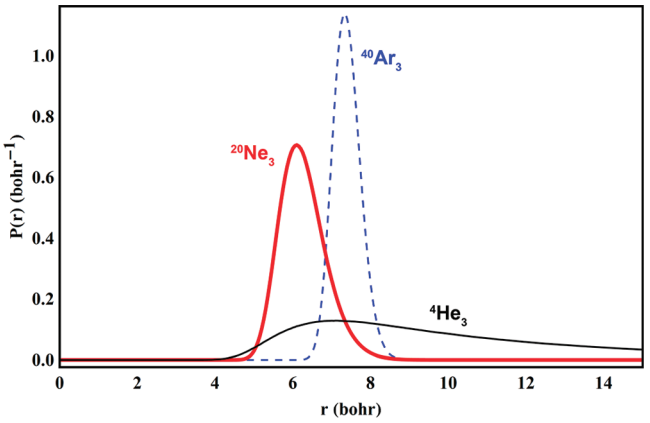

Figure 1. Pair distributions for $^{40}Ar_3$, $^{20}Ne_3$, and $^{4}He_3$.

Nielsen and co-workers[12] performed a highly accurate Faddeev type calculation on $^{4}He_3$ and $^{4}He_2{}^{3}He$ using the LM2M2 potential and hyperspherical coordinates. Their energies agree with other works with the same potential. To get a visual picture of the geometrical structure of these systems, they plotted the contour diagrams of the particle density distributions in the intrinsic coordinate system. Their conclusion was that both systems have a triangular structure, the pure system being represented by an equilateral triangle.

González-Lezana and co-workers presented a study[13] with the LM2M2 potential using Distributed Gaussians Functions (DGFs) to investigate the Efimov effect and various geometrical properties. A more detailed presentation of their work, extended also to the neon and argon trimers, is discussed below. The computed energy was below all results presented in the literature with the same or similar potentials, as noticed by Esry et al.[24] They briefly discussed the discrepancy in the literature about the main geometrical configurations contributing to the ground state. Its pair distribution function has two maxima, the first one more pronounced than the second in a ratio of about 2:1. They interpreted this bimodal distribution as being due to the presence of quasilinear configurations. This puzzling feature, as noticed by Blume et al.,[17] is in contrast with what other authors have observed. Their conclusion was that the ground state is formed by about 30% of quasilinear configurations and a negligible contribution from equilateral triangles. The two peaks placed at 4.53 and 8.81 Å of Figure 1 of their paper were interpreted as due to quasilinear configurations presenting two sides of about 4.5 Å and the third side of about 9 Å, resulting in a ratio between the two maxima of about two.

Blume and Greene[16] performed a study of the ground and excited states of the clusters $^{4}He_N$ for $N = 3-10$ combining Monte Carlo methods with the adiabatic hyperspherical approximation. For the trimer they obtained an energy in full agreement with other previous independent calculations, but noticed a disagreement with the value obtained by González-Lezana et al.[13] They computed various geometric properties, such as the mean atom−atom distance, but did not explicitly discuss the shape of the trimer.

Bressanini and co-workers[18] using the TTY potential studied small ^{4}He clusters with a ^{3}He impurity. Their energetic results on

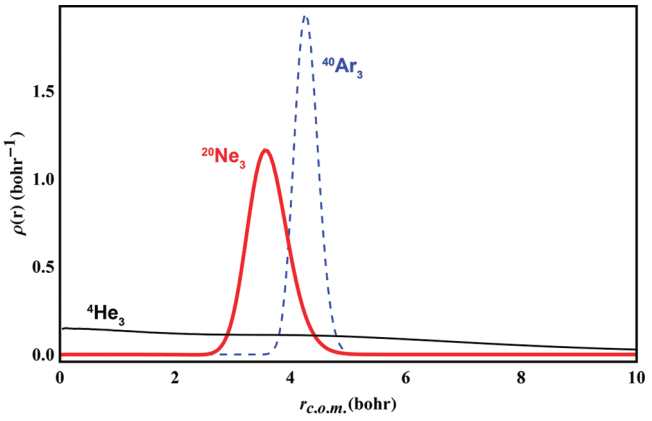

Figure 2. Radial density distributions for $^{40}Ar_3$, $^{20}Ne_3$, and $^4He_3$.

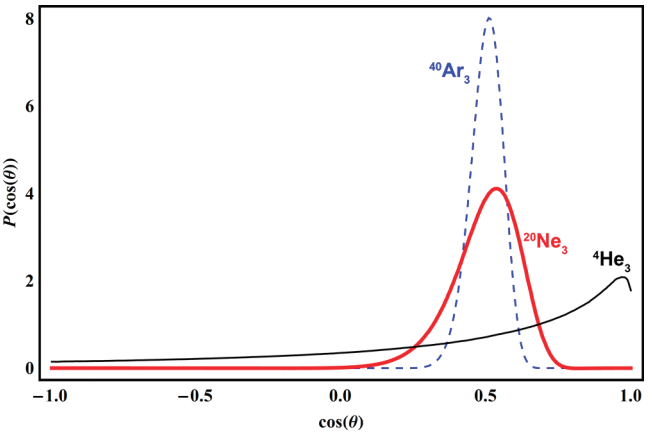

Figure 3. Cosine distributions for $^{40}Ar_3$, $^{20}Ne_3$, and $^4He_3$.

the pure trimer $^4He_3$ and the lighter $^4He_2{}^3He$ agree with previous calculations. Studying the structural properties, they confirmed the earlier observation by other researchers that the density distribution with respect to the c.o.m. for the pure helium trimer has a maximum toward zero, interpreted as a tendency of the trimer to assume nearly linear configurations. The same feature was also established for the lighter trimer $^4He_2{}^3He$.

Blume and co-workers[17] solved the Schrödinger equation in hyperangular coordinates in an adiabatic representation for the trimers of ^4He, ^{20}Ne, and ^{40}Ar. They did not explicitly discuss the shape of the helium trimer. However, they noticed broad pair and angle distributions suggesting that, at each value of the hyperradius, there is a dominant geometrical configuration. While comparing with the literature, they noticed a large difference between their energy and the one computed by González-Lezana et al.,[13] as well as a disagreement of the pair and cosine distributions. Their pair distribution shows no second maximum and their cosine distribution is significantly higher near $\cos(\theta) \cong 0$. They checked the validity of their distributions by performing an independent DMC simulation that confirmed their results and concluded that González-Lezana et al.'s results "are likely to be incorrect".

Building on their previous work[13] González-Lezana and coworkers[14] extended their treatment to the neon and argon trimers, while studying in more details the geometric properties of the helium trimer. They briefly discussed the geometry controversy citing a few previous works. To study the geometry of the trimer they assigned a weight to each basis function in their wave function and classify them into linear, equilateral, isosceles, and scalene triangular arrangements. With this approximation, they found that the equilateral configuration gives a negligible contribution, while the scalene configuration accounts for 48% and the quasilinear for 27%. They further plotted (Figure 5 of their paper) the cosine distribution, reconstructed from the average values of the first seven momenta of $\cos(\theta)$. The cosine distribution shows two peaks with ratio 1:2 for cosines value of -1 and $+1$, respectively, a feature that is in marked contrast with other papers. Finally they plotted (Figure 3 of their paper) $D(R_1,R_2)$, the bidimensional probability density function, integrated over R_3. This function shows a bimodal structure with maxima located at 4.53 and 8.81 Å. The authors interpreted this bimodal structure as being due to the presence of quasilinear geometrical configurations.

The results of this paper seem to support the significant contribution of linear configurations, as opposed to equilateral ones. However, it has been suggested[24] that the conclusions of the authors are flawed by some kind of error in their computational scheme which, despite the claim to be a variational method, produced a ground state energy which is below what is considered (see Table 1) the exact ground state energy.

Roudnev and Yakovlev[19] numerically solved the Faddeev equations for the helium trimer using a variety of potentials. The energy of the ground state for the TTY potential, reported in Table 1, is in agreement with many other calculations in the literature with the same potential. To study the shape of the system, they plotted what they called a "conditional density function" $\rho(r,z)$ that represents the spatial distribution of one particle with respect to the axis defined by the other two particles, a function already considered by Lewerenz.[11] Their conclusions on the geometry are not very clear: they say "the conditional density function of the ground state decreases democratically in all directions. The density function of the excited state has two distinguishable maxima demonstrating the linear structure of the cluster". They do not say it explicitly, but seem to imply that the ground state is not linear, as opposed to the excited state.

Barletta and Kievsky[20] studied the helium trimer using the LM2M2 potential and a correlated hyperspherical harmonic wave function, obtaining a ground state energy in very good agreement with many other previous calculations. They mentioned the discrepancy in the literature over the shape of the spatial arrangement of the three particles in the ground state. In order to gain some insight they computed the pair distribution function $P(r_{ij})$ and the atom distribution function with respect to the center of mass $P(r_{c.o.m.})$. Their plots are in agreement with those found in the literature. They noticed that the ratio between the average interparticle distance and the average distance of an atom from the center of mass of the system, $\langle r_{ij}\rangle/\langle r_{c.o.m}\rangle$, is very close to the ideal $\sqrt{3}$ of the equilateral triangle. They observed from their plot $P(r_{c.o.m.})$ that the probability to find an atom close to the center of mass of the system is almost zero and concluded that the helium trimer can be described as an equilateral triangle. They assigned an equilateral structure to $^4He_3$ based on the average values of the atom−atom and atom−c.o.m. distances. However, average values are really representatives of "typical" configurations of the system only when the distributions are strongly peaked, which is not the case here. Furthermore, their observation, from the plot of $P(r_{c.o.m.})$, that the probability to find an atom close to the c.o.m. is almost zero, is correct, and their plot is similar to that found by other authors and by us. However, to

correctly interpret this plot as a density, the volume element r^2 must be included in the picture, as is usually done in this case. When this is taken into account, as correctly pointed out by others in the literature,[11,13,18] this normalized distribution does not go to zero as an atom approaches the c.o.m. and, as a matter of fact, there is a maximum for $r_{\text{c.o.m.}} = 0$.

In a recent paper on the Ritz variational treatment of the helium dimer and trimer, Geltman,[21] while not explicitly studying the shape of the trimer, dismissed the quasilinear configurations in favor of the "intuitive equilateral triangular form". The reason is that the potential minimum occurs for an equilateral triangle of sides equal to the separation at the binary potential minimum. It is well-known that, for very floppy systems like the helium clusters, the equilibrium geometry does not correspond to the minimum of the potential. In fact, it is even questionable that for these systems one can define an equilibrium geometry in the same sense used for strongly bound systems. Furthermore, Geltman stated that the finding of linear structures would be hard to accept as compatible with the boson symmetry requirement of the three-atom wave function. Our calculations on the helium trimer, as all the others reviewed in this paper, employed a wave function with the correct boson symmetry: the permutational symmetry of the boson wave function has nothing to do with the space symmetry of the total wave function.

In summary, most studies agree about the floppy nature of $^4He_3$, which is well established. However, there is a controversy on the kind of configurations that are accessible to the system, what "references" geometrical configurations, if any, should be considered representative (linear, equilateral, and so on), and to what extent the system, fluctuating, deviates from those.

METHOD

For all the trimers, $^4He_3$, $^{20}Ne_3$, and $^{40}Ar_3$, the potential energy was written as a sum of two-body interactions and three-body contributions were not included. Atomic masses were taken from the NIST database.[25] For the Ne−Ne and the Ar−Ar interactions, we employed a simple Lennard-Jones potential. The parameters were taken from ref 26. The same potentials have been used in the past to compare the structural properties of the different trimers.[17]

For the He−He interaction, we performed independent simulations using both the LM2M2 and the TTY potentials, which are known to give similar results.

We approximate the ground state wave function of all systems with the pair-product form

$$\Psi_T(\mathbf{R}) = \varphi(r_{12})\varphi(r_{13})\varphi(r_{23}) \quad (1)$$

where r_{ij} is the distance between two atoms. The pair function

$$\varphi(r) = \exp\left(-\frac{p_5}{r^5} - \frac{p_2}{r^2} - p_0 \ln(r) - p_1 r\right) \quad (2)$$

has been used with success by several workers[11,18,27] for the description of small rare gas clusters. The chosen form for the trial wave function makes it impossible to compute analytically the matrix element of the Hamiltonian operator, so the variational energy and other properties must be computed by a numerical method like the variational Monte Carlo(VMC).[28,29] The VMC approach poses no restrictions on the functional form of the trial wave function, requiring only the evaluation of the wave function value, its gradient, and its Laplacian, and these are easily computed. Although the VMC approach, with a proper choice of the trial wave function, can give very good results by itself; in this work it has been mainly used to optimize good trial functions to be subsequently employed in a diffusion Monte Carlo (DMC) simulation. Both methods are well described in the literature and we do not discuss them further.

All parameters in our wave function were optimized using a robust estimation method by minimizing the absolute mean deviation of the local energy.[30] The optimized trial wave functions Ψ_T were employed in DMC calculations performed using different time steps. The time step bias has been eliminated by extrapolation to $\tau \rightarrow 0$.

For boson systems, like those studied here, DMC can estimate the exact ground state energy within the desired statistical accuracy. However, expectation values of operators $\hat{O}$ that do not commute with the Hamiltonian are approximated, because they are computed with respect to the distribution $\Psi_T(\mathbf{R})\phi_0(\mathbf{R})$, called the *mixed distribution*, and not with respect to the square of the exact wave function $\phi_0^2(\mathbf{R})$. For these properties we give a better estimate, which is second order on the error of the trial wave function, using the so-called second order estimate $\langle\hat{O}\rangle \cong 2\langle\hat{O}\rangle_{\text{DMC}} - \langle\hat{O}\rangle_{\text{VMC}}$.

Using the appropriate coordinates for the description of a property of a system can result in a simpler interpretation of the results. In particular, we believe that to get an insight on the shape of a floppy trimer it is not sufficient, as it has been done in the past, to study one-dimensional distributions, like the cosine or the radial distribution. The shape of a triangle is inherently a two-dimensional property and a greater understanding can be gained by studying the proper distribution. Because we are interested in the classification of the shape of the triangle, but not in its size, we believe that the distribution of two out of three angles of the triangle is a natural choice. Fixing two angles automatically defines the third one. This distribution is well suited to discuss the shape of a trimer since it does not involve distances. As mentioned previously, all distributions employed in the past involved one or two distances: while useful to get some insight on the general features of the helium trimer, like its generic floppiness, they made more difficult to extract the precise geometrical features due to the coordinate systems employed. This is probably one of the reasons why different authors attributed different geometrical structures to this system: extracting shape information from a plot involving only averaged distances is not always trivial.

RESULTS AND DISCUSSION

To check the correctness of our calculations we computed the ground state energy of all the trimers and compared them with literature calculations with the same potential. Our DMC ground state energy of $^{40}Ar_3$ is $-211.893(2)$ cm^{-1}. Blume et al.[17] obtained -211.78 cm^{-1}, while Leitner et al.[26] -211.86 cm^{-1}. For $^{20}Ne_3$, we recovered an energy of $-42.547(2)$ cm^{-1}. This value is in good agreement with other values computed with the same potential: -42.51,[26] -42.38,[17] and -42.55 cm^{-1}.[31] For $^4He_3$, we computed an energy of $-0.08783(4)$ cm^{-1}, in very good agreement with the literature (see Table 1).

The $^4He_3$ structural properties were found very similar for the TTY and LM2M2 potentials, so we show here only the results for the TTY potential. The relative insensitiveness of the structural properties with respect to the details of the potential used has been noticed also for $^{20}Ne_3$ by Baccarelli et al.[31]

For all the systems studied here the VMC distribution functions were very similar if not super imposable to the DMC

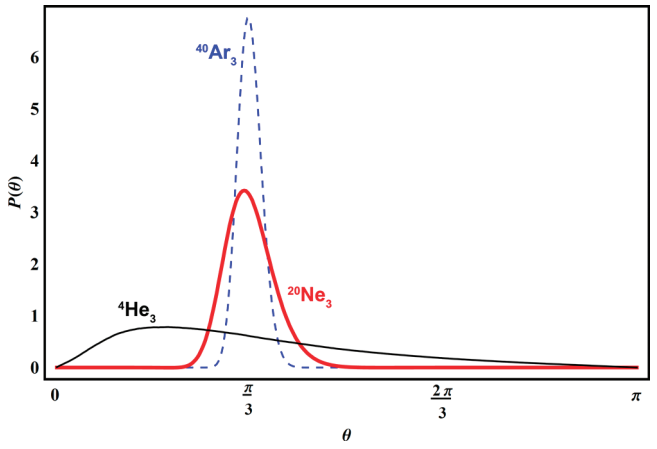

Figure 4. Angular distributions for $^{40}Ar_3$, $^{20}Ne_3$, and $^{4}He_3$.

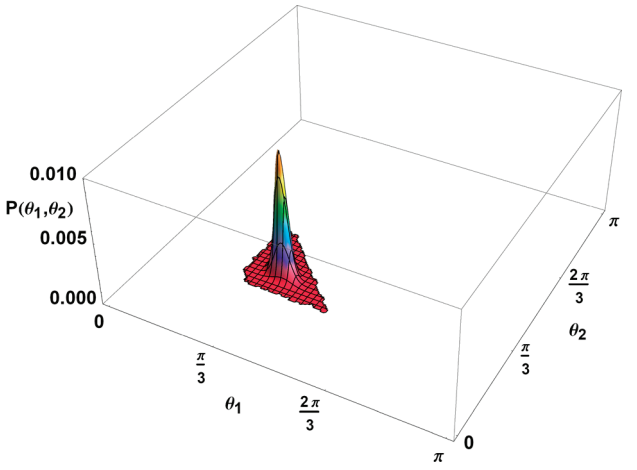

Figure 5. Angle−angle distribution function for $^{40}Ar_3$.

distributions, a sign that the trial wave functions employed were of high quality and able to correctly describe the various structural properties. In this case, the use of the second order estimator, whose distributions are shown in the figures, is justified because it corrects most of the remaining deviations from the exact distribution. In cases where the wave function is not of good enough quality, one should resort to the descendent weighting method to estimate the distributions with respect to $\phi_0^2(\mathbf{R})$.

The pair distribution function for $^{40}Ar_3$, shown in Figure 1, is strongly peaked around 7.4 bohr. The neon trimer is known to be less rigid than the argon trimer, as can be seen from the slightly larger pair distribution. In contrast the pair distribution function of $^{4}He_3$ is much extended, as has been already pointed out many times in the literature. Comparing these curves it is clear that the structure of the helium trimer is quite different from the other two. The same picture emerges by looking at the curves of the radial density with respect to the c.o.m. The curves for argon and neon, shown in Figure 2, are peaked, respectively, around 4.1 and 3.6 a.u. The curve for helium is completely different. Unlike the plot for Ar and Ne, the maximum for the He curve is located at zero. This is the plot that led some authors[11,18] to the conclusion that nearly linear configurations, that is, where one particle is close to the center of mass, are important in the structural description of this very floppy system.

The cosine distribution for the argon trimer, shown in Figure 3, is peaked around 1/2, which is the cosine of $\pi/3$, the angle of an equilateral triangle. The distribution for $^{20}Ne_3$, still peaked around 1/2, is much broader, and close inspection of its values shows that it is greater than zero, albeit very small, even for values close to −1. This suggests a structural picture of an equilateral triangle that fluctuates mainly around the equilibrium structure, but can be found occasionally even in much distorted shapes. Blume et al.[17] studied the cosine distribution of the neon trimer as a function of the hyper-radius and concluded that at the equilibrium geometry the system is well described by an equilateral triangle. However, as the hyper-radius is increased, going toward the fragmentation into a diatomic molecule and an atom, the preferred geometry becomes linear.

From the analysis of the radial distribution functions of $^{20}Ne_3$ estimated by their DGF method, Baccarelli et al.[31] concluded that the ground state has a predominant equilateral structure. They then proceeded to analyze the importance of the different DGFs to the description of the ground and several excited states. As already mentioned they classified each arrangement as "flat isosceles", "tall isosceles", "equilateral", "scalene", and "collinear", associating each basis function to a particular triangular arrangement taking into account some variations in the length of the sides of the triangle. From their analysis the authors concluded that the ground state of $^{20}Ne_3$ is described by a contribution of all families, with about 25% of the total weight each, with the exclusion of the collinear one.

The cosine distribution of $^{4}He_3$ is unlike those of the other studied systems. It does not show a maximum around 1/2, but rather extends all the way from −1 to +1, with a maximum close, but not exactly, to +1. Because a cosine of +1 corresponds to an angle of zero, this curve, like the previous one, suggests that nearly linear configurations are important.

Blume et al.[17] in their comparative study of $^{4}He_3$, $^{20}Ne_3$, and $^{40}Ar_3$ report a cosine distribution very similar to ours, except their plot is cut close to +1, so it is not possible to infer where the maximum of their curve is exactly.

The same picture emerges examining the curves for the angular distributions shown in Figure 4. Both the $^{40}Ar_3$ and $^{20}Ne_3$ curves are centered nearly on $\pi/3$. Note that the average angle for any trimer, regardless of the distribution, is always $\pi/3$, due to the constraint that the sum of the three angles in a triangle is equal to π. The shape of the angular and radial distributions leaves no space to alternative descriptions of the argon and neon trimers other than an equilateral triangle that fluctuates around the equilibrium structure, the neon trimer being less rigid than the argon trimer.

We believe that a natural way to discuss the shape of a trimer is by an angle−angle two-dimensional distribution function, and this is trivially done in a Monte Carlo simulation. This distribution gives the probability to find the first angle at a given value and the second at another one. A two-dimensional array is set up during the simulation and the values of two angles of the instantaneous atomic configuration are gathered. This procedure leads naturally to a distribution function that gives the probability to find the triangle with particular values of all the angles, the third angle being automatically fixed by the other two.

We note that these two-dimensional distribution functions are trivial to compute using QMC simulations, but could be difficult to estimate using other computational methods employed in the past to study these systems. This could be one of the reasons why they have never been employed, to our knowledge, to describe

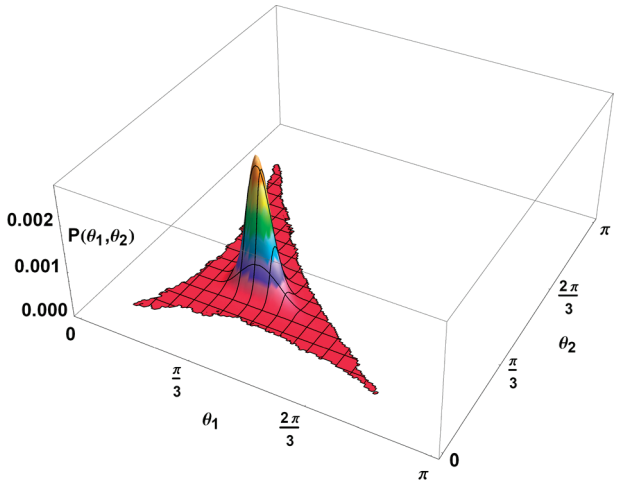

Figure 6. Angle−angle distribution function for $^{20}Ne_3$.

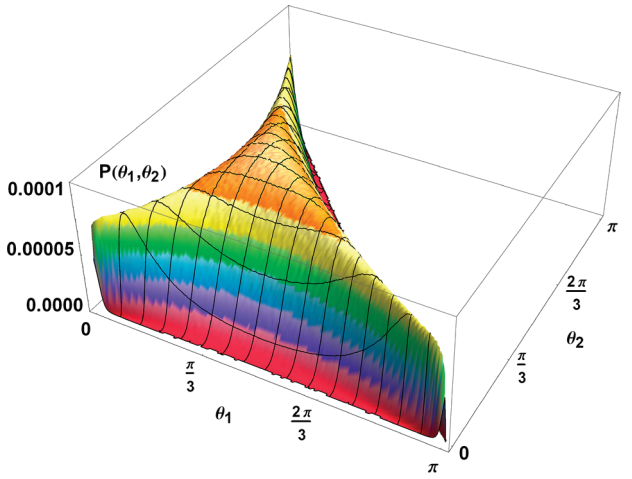

Figure 7. Angle−angle distribution function for $^{4}He_3$.

the shape of these systems, despite the fact that they lead to a natural pictorial interpretation of their global shape.

The function for $^{40}Ar_3$, shown in Figure 5, is strongly peaked around the angle values $(\pi/3,\pi/3)$ showing that the classification of its structure as an equilateral triangle is a natural one. However, the system is not as rigid as a typical triangular molecule that vibrates around its equilibrium structure, and angles as large as $\pi/2$ can be found, albeit with an extremely small probability.

The angle−angle distribution function for $^{20}Ne_3$, shown in Figure 6, was obtained during a simulation sampling 10^7 configurations. The distribution is peaked where two of the angles, and necessarily also the third, are at about $\pi/3$, confirming the picture of the equilateral triangle already suggested by the cosine distribution. However, due to the floppier nature of the neon trimer, caused by the lighter mass and a shallower potential compared to the argon trimer, the system can be found in a variety of triangular configurations, isoscele and scalene, around the equilateral one. It is also interesting to note that the distribution stretches toward the points $(0,0)$, $(0,\pi)$, and $(\pi,0)$, corresponding to almost linear configurations, where the trimer can be sporadically found, albeit with extremely small probability. This is completely in agreement with a recent coulomb explosion experiment[32] where the structures of the argon and neon dimer, trimer, and tetramer were investigated. The analysis of the kinetic energy release (KER) spectrum after the coulomb explosion of the neon trimer showed a contribution not only from strictly equilateral configurations, but also from a larger variety of triangular configurations.

Having shown the validity of the angle−angle distribution function to naturally interpret the structure of neon and argon trimers, where there is consensus in the literature about their general shape, we now proceed to apply this analysis to the more controversial case of the helium trimer, where different authors have given different interpretations.

The shape of the angle−angle distribution function for $^{4}He_3$ shown in Figure 7 is completely different from the analogous plots for neon and argon. There is a weakly pronounced maximum again located at about $(\pi/3,\pi/3)$, but the distribution is almost flat, extending in all directions with slowly decaying probabilities such that almost all possible angle pairs, including quasilinear configurations, are reached. This is a fundamentally different picture than for Ne_3 and Ar_3 whose angle−angle probability distributions show a peaked distribution around the equilateral structure. So, the angle−angle distribution emphasizes the floppiness of the helium trimer.

From this plot it seems very clear that all kinds of configurations must be taken into account in the description of this very peculiar system. Classifying $^{4}He_3$ as either equilateral or linear or any other particular shape is not sensible, because this system explores almost all possible configurations with probability roughly of the same order of magnitude and does not show any particularly preferred shape.

At first, it might seem puzzling that the angle−angle distribution function has a maximum at $(\pi/3,\pi/3)$ while both the cosine and the angle distributions, in Figures 3 and 4, have a maximum for quite different values. However, there is no contradiction: while $(\pi/3,\pi/3)$ is a single point, in the one-dimensional distribution for each angular value many points contribute to the integral and their contribution is large as the angle−angle probability distribution is fairly flat. Near the edges of the angle−angle plot the trimer has near-linear configurations, but the probability has decreased only to roughly 50% of the maximum value. So near-linear configurations give a significant contribution to the one-dimensional distributions, as observed by many authors.

This phenomenon is absent in $^{20}Ne_3$ and $^{40}Ar_3$ because the angle−angle distribution is strongly peaked around $(\pi/3,\pi/3)$ and goes quickly to zero, moving out from the maximum. So, most of the configurations give zero contribution to the integrals needed to compute the various one-dimensional distributions.

Looking at Figure 7, it is not surprising that some authors, by looking only at mean values or one-dimensional distributions, averaging out one or more dimensions, were misled in their judgment about the shape of the helium trimer: for such a broad distribution any average is not really representative of the system.

Recently some Coulomb explosion experiments have been performed with the aim to experimentally measure the shape and the different geometric parameters of the Neon and Argon trimers[32] and it seems likely that in the near future a similar investigation on the Helium trimer can be carried out. In a typical experimental setup, all the atoms are singly ionized using a very short laser pulse. After ionization, the system breaks up and all the momentum vectors of the different ions are measured in

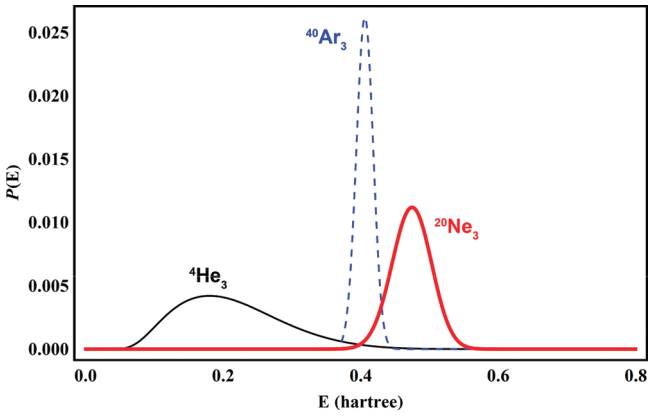

Figure 8. KER distribution in the classical approximation.

coincidence. These experiments cannot directly measure the geometric parameters of the trimer under study such as the interatomic distances. Rather there is the necessity to invert the experimental data of the momentum vectors to reconstruct the geometrical structure of the system. An easily accessible experimental quantity measured in these experiments is the total kinetic energy. An assumption that is made in the interpretation of the experimental data is that immediately after the ionization the system explodes and, considering the ions as classical point charges, the Coulomb repulsive potential energy pushes the ions apart while it is converted into kinetic energy. To recover the interatomic distances, an initial guess must be made on the system geometric shape, choosing the initial position of the various atoms based on some probability distribution. Then the classical equations of motion are numerically solved, following the ions until complete breakup, and the resulting distributions are compared with the experimental data. The initial guess on the interatomic distances is adjusted until the numerical simulation and the experiment agree. So the correct extraction of the geometrical parameters requires the knowledge of the geometrical shape of the trimer.

To help experimentalists to interpret Coulomb explosion experiments of these systems, we computed the KER distribution in the classical approximation because it is simply the Coulomb potential energy after all the atoms have been singly ionized, that is, $1/r_{12} + 1/r_{13} + 1/r_{23}$. Figure 8 shows the different features of the three systems studied here. The curve for $^{40}Ar_3$ is the sharpest of the three: again, a consequence of the relative higher rigidity of the system. For a completely rigid classical system, the curve would be a Dirac's delta. Neon's plot is broader and shifted to higher energies due to the fact that, as shown in Figure 1, the neon atoms are closer and after ionization the Coulomb repulsion is higher. On the other hand, the curve for $^{4}He_3$ is shifted toward zero, because helium atoms are usually far from each other but has a relative long tail, because the trimer explores a variety of configurations where He atoms can approach to a relatively short distance.

CONCLUSIONS

We performed quantum Monte Carlo simulations for the trimers $^{4}He_3$, $^{20}Ne_3$, and $^{40}Ar_3$ and compared various geometric probability distributions in order to resolve a long-standing controversy in the literature about the shape of the helium trimer. We introduced a two-dimensional distribution, namely, the angle−angle distribution, that provides a clear and intuitive way to interpret the geometrical shape of any trimer. This distribution is not easily accessible using computational techniques different than Monte Carlo methods, and this could be a reason why it has not been used in the past. Computing this distribution for the different trimers the almost equilateral structure of $^{40}Ar_3$ emerges naturally. $^{20}Ne_3$ has still an equilateral structure, but its floppier nature makes it possible to find this system even in more distorted shapes, albeit with low probability. The analysis of the angle−angle distribution function for $^{4}He_3$ reveals an almost flat distribution, with a very weakly pronounced maximum close to the equilateral structure and a distribution that extends in all directions covering roughly all possible shapes before going to zero almost at the boundaries. All kinds of configurations must be taken into account in the description of $^{4}He_3$, and in our opinion, classifying it as either equilateral or linear or any other particular shape is meaningless, because, in this case, the intuitive notion of equilibrium structure is ill defined. Data on cluster geometry can be obtained by Coulomb explosion experiments.[32] To extract the internuclear distances between the atoms in a trimer from the data gathered in these experiments, an assumption on the geometrical configuration of the system must be made. Our results for neon and argon trimers agree with recent experimental data.[32] We hope that our analysis of the shape of the helium trimer can be useful in interpreting future Coulomb explosion experiments aimed at investigating its geometrical properties.

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