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Analysis of the low-lying spectrum of the He-tetramer*

Bao Cheng-Guang(鲍诚光)[a], He Yan-Zhang(贺彦章)[a], and Li Xun-Gui(李训贵)[b]

[a] *Department of Physics, Zhongshan University, Guangzhou, 510275, China*

[b] *Department of Physics, Guangzhou University, Guangzhou 510405, China*

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There is a type of nodal surface imposed by symmetry on wavefunctions. These surfaces are crucial to the ordering of low-lying states. Based on an analysis of this type of surface and based on existing theoretical results, the feature of the low-lying spectrum of the He-tetramer is studied, and the candidates of bound states and narrow resonances are suggested.

Keywords: He-tetramer, atomic cluster, effect of symmetry
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It is well known that the interaction between two He atoms is very weak.[1,2] Therefore the binding in the cluster He_N, a system composed of N helium atoms, is also weak. The investigation of these clusters is a very interesting topic due to the theoretical interest in very weakly bound and floppy few-body systems (e.g. the search for extremely diffuse bound states), and also due to the fact that the features of these systems (e.g. how many bound states and narrow resonances would be contained in a He_N cluster) would affect the feature of the helium liquid. In 1993 an important progress along this line was achieved, namely the experimental confirmation of the existence of the bound ^{4}He dimer.[3−5] This finding greatly encouraged the search for bound states or resonances of these few-body systems. A number of theoretical studies have been devoted to ^4He_N in these years (refer to Refs.[6] and [7] and references therein). It was found recently, based on a Monte Carlo hyperspherical calculation,[6] that all the ^4He_N systems with $3\leq N\leq 10$ contain two L^Π=0^+ bound states (L is the total angular momentum, Π is the parity), denoted as 0_1^+ and 0_2^+. It is noted that 0_2^+ must contain nodal surfaces (in order to be orthogonal to 0_1^+) and thereby must have higher oscillation energy. However, the L_1^Π states with $L\neq$0 might not contain nodal surfaces in their internal wavefunction (as we shall see) and thereby have lower oscillation energy. Although these $L\neq$0 states have an additional collective rotation energy $E_{\rm rot}$, their energies might be comparable to that of 0_2^+, because the decrease in internal energy $E_{\rm int}$ might compensate for the increase in $E_{\rm rot}$. Thus, the existence of 0_2^+ is a signal that $L\neq 0$ excited bound states might exist as well. However, the calculation of $L\neq 0$ states is more difficult than that of L=0 states. To the knowledge of the author, there has been no such calculation until now. Before performing tedious quantitative calculation, it might be helpful to perform a qualitative analysis. This paper is dedicated to this purpose.

We perform an analysis on the low-lying states of the He-tetramer $^4He_4$ based on fundamental principles. It is noted that the low-lying spectrum of a four α-particle model nuclear system with the $\alpha-\alpha$ interaction containing a repulsive core and an attractive tail as

$$V(r)=V_o[-{\rm e}^{-(r/a_1)^2}+b{\rm e}^{-(r/a_2)^2}] \qquad (1)$$

has been previously studied.[8] Here V_o, b, a_1 and a_2 are adjustable parameters ($a_1>a_2$, $V_o>0$ and $b>1$). It is found that there are three states, namely the 3_1^-, 4_1^+ and 2_1^+ states, lower than the 0_2^+, and a state 2_1^- a little higher than the 0_2^+, while the other states are considerably higher. For example, when $a_1=2.105$fm and $a_2=1.428$fm (these two values are specialized for the $\alpha-\alpha$ nuclear interaction), $V_o=300$MeV and $b=2$, we have the excitation energies $E_x(L_i^\Pi)=E(L_i^\Pi)-E(0_1^+)$ in increasing order as $E_x(3_1^-)=12$, $E_x(4_1^+)=21$, $E_x(2_1^+)=22$, $E_x(0_2^+)=24$, $E_x(2_1^-)=28$, $E_x(1_1^-)=34$, $E_x(3_1^+)=40$, ... (all in MeV). Since the interaction given by Eq.(1) is very different from the real He–He interaction of atoms, the

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findings of the ordering from the four-boson model system may not be valid for the He_4 system in general, unless there is a reason beyond dynamics governing the findings. Thus we have to find the reason why the 3_1^-, 4_1^+ and 2_1^+ states are lower, so as to judge whether these quantum numbers are also associated with promising candidates of bound states of the He-tetramer.

We begin from a study of the effect of symmetry. It is well known that the feature of a state is determined by how the wavefunction is distributed in coordinate space; i.e., in which domains the wavefunction is distributed, how extensive is the distribution, how many nodal surfaces are contained and where are these surfaces located. It is noted that how fierce is the internal motion is determined by the nodal structure of the wavefunction. If many nodal surfaces are contained in a state, the energy of the state must be high.

There is a type of nodal surface existing at regular shapes imposed by symmetry. For example, let $\boldsymbol{i}'$-$\boldsymbol{j}'$-$\boldsymbol{k}'$ be a body frame, an eigenstate of a four-body system can be expanded as

$$\Psi_{LM}(1234) = \sum_Q D^L_{QM}(-\gamma, -\beta, -\alpha)\, \Psi_{LQ}(1'2'3'4'), \tag{2}$$

where M and Q are the projections of L along a fixed Z-axis and the body $\boldsymbol{k}$'-axis, respectively. D^L_{QM} is the Wigner function for the rotation of the body-frame with the Euler angles α, β, and γ as arguments. (1234) and $(1'2'3'4')$ denote the coordinates of the four particles relative to the fixed frame and to the body-frame, respectively. Ψ_{LQ} is called a Q-component.

If the particles, say, form a cone having a threefold axis lying along $\boldsymbol{k}'$, then a rotation about $\boldsymbol{k}'$ by $2\pi/3$ is equivalent to a cyclic permutation of the three particles at the base of the cone. For bosonic systems of identical particles, wavefunctions are invariant to particle permutation. Thus we have

$$\mathrm{e}^{-\mathrm{i}2\pi Q/3}\, \Psi_{LQ}(\mathrm{CONE}) = \Psi_{LQ}(\mathrm{CONE}), \tag{3}$$

where the left-hand side arises from the rotation, and the label $\Psi_{LQ}(\mathrm{CONE})$ denotes that the coordinates just form a cone. It is clear from Eq.(3) that $\Psi_{LQ}(\mathrm{CONE})$ is nonzero only if Q=3m, here m is an arbitrary integer. In other words, there is a nodal surface in the wavefunction Ψ_{LQ} at the cones if $Q \neq$3m, disregarding the lengths of the sides and the orientation of the cone. Evidently, this nodal surface is imposed by symmetry, which is called an inherent nodal surface (INS).[9,10] Owing to the existence of the INS, the cone is inaccessible to $Q \neq$3m components.

It is recalled that the base of a cone is a regular triangle parallel to the $\boldsymbol{i}'$–$\boldsymbol{j}'$ plane. Let $\boldsymbol{n}'$ be a unit vector defined in the body-frame parallel to a side of the regular triangle, the angle between $\boldsymbol{n}'$ and $\boldsymbol{i}'$ being δ. Evidently, a rotation of the cone about $\boldsymbol{n}'$ by π together with a space inversion is equivalent to an interchange of two particles. Thus we have

$$\begin{aligned} &\mathrm{e}^{-\mathrm{i}\pi L_{n'}}\, \Psi_{LQ}(\mathrm{CONE}) \\ =&\mathrm{e}^{-\mathrm{i}\delta L_{k'}} \mathrm{e}^{-\mathrm{i}\pi L_{i'}} \mathrm{e}^{\mathrm{i}\delta L_{k'}}\, \Psi_{LQ}(\mathrm{CONE}) \\ =&\Pi\, \Psi_{LQ}(\mathrm{CONE}), \end{aligned} \tag{4}$$

where $L_{n'}$, $L_{i'}$ and $L_{k'}$ are the projections of the operator L along $\boldsymbol{n}'$, $\boldsymbol{i}'$ and $\boldsymbol{k}'$, respectively. For the Q=0 component, from Eq.(4) we have

$$(-1)^L\, \Psi_{L0}(\mathrm{CONE}) = \Pi\, \Psi_{L0}(\mathrm{CONE}). \tag{5}$$

Thus the cone is inaccessible to the Q=0 components if $(-1)^L = -\Pi$, or in other words, these components have an INS at the cones.

The above example demonstrates that an INS may exist in any shape with certain geometric symmetry. The accessibility of shapes to the Q-components of the L^Π states of a four-boson system has been investigated in detail in Ref.[11]. The main findings for $L \leq$6 states are summarized in Table 1 of this paper. In Table 1, ETH denotes an equilateral tetrahedron. If an ETH extends or contracts along one of its twofold axes, then it becomes a GETH. If an ETH twists along one of its twofold axes (a pair of particles symmetric with respect to the twofold axis rotates about the axis by an angle, while the other pair of particles rotates by the same angle but in the reverse direction), then it becomes a TETH. If an ETH extends or contracts along one of its threefold axes, then it becomes a CONE. SQ is a square. DIA (REC) is a diamond (rectangle), and is a coplanar structure having three mutually orthogonal twofold axes. "Twofold" denotes a coplanar structure with only one twofold axis. CT is a regular triangle with an extra particle at the centre. CL is a collinear chain having a twofold axis normal to the chain. When a shape is accessible to a Q-component of a L^Π state, Q is marked in the associated block. For example, the TETH is accessible to the $|Q|$=0 and two components of the 2^+ states, thus the 0 and 2 are marked in the block. The ETH is inaccessible to all the components of 1^+, 2^+, 3^+ and 5^+ states, thus

the associated blocks are empty. It is recalled that the inaccessibility of a shape implies the appearance of an INS at the shape. Thus, inherent nodal structures can be understood from the table.

Noting that ETH and SQ are the most favourable shapes for binding, because the (local) minima of the total potential energy locates at them. Therefore the accessibility of these two shapes is important to a state. We know from Table 1 that the 0^+ and 4^+ states are both ETH-accessible and SQ-accessible, the 3^- and 6^- states are ETH-accessible but SQ-inaccessible, the 2^+ states are SQ-accessible but ETH-inaccessible, while all the other $L \leq 4$ states are both ETH-inaccessible and SQ-inaccessible. This point is important to the following discussion.

Table 1. The accessibility of shapes to the Q-components of L^{Π} states. k' is adopted to be parallel to a twofold axis of the ETH, to the twofold axes of the GETH and TETH, to the threefold axis of the CONE, to be normal to the plane of coplanar structures, or to be parallel to the collinear chain CL. An empty block implies that the associated shape is inaccessible to the associated state.

	0^+	1^+	2^+	3^+	4^+	5^+	6^+
ETH	0				0,4		0,4
GETH	0		0		0,4		0,4
TETH	0		0,2	2	0,2,4	2,4	0,2,4,6
CONE	0		0	3	0,3	3	0,3,6
SQ	0		0		0,4	4	0,4
DIA, REC	0		0,2	2	0,2,4	2,4	0,2,4,6
2-fold	0		0,2	2	0,2,4	2,4	0,2,4,6
CT	0		0		0		0,6
CL	0		0		0		0

	0^-	1^-	2^-	3^-	4^-	5^-	6^-
ETH				2			2,6
GETH			2	2	2	2	2,6
TETH	0		0,2	2	0,2,4	2,4	0,2,4,6
CONE		0		0,3	3	0,3	3,6
SQ							
DIA, REC							
2-fold		1	1	1,3	1,3	1,3,5	1,3,5
CT				3	3	3	3
CL							

There are two types of nodal surface.[9] The INS belongs to the first type contained inherently in wavefunctions. When the energy of a state becomes higher, the second type, namely the dynamical nodal surface (DNS), might emerge to realize the orthogonality among states. It is easy to distinguish the INS from the DNS. The location of the former does not move in accordance with the variation of dynamical parameters (e.g. a change in the strength of interaction), while the latter does move. A series of states having the same L and Π (and having the same spatial permutation symmetry in general) will have exactly the same common nodal surface, i.e. the INS, while the DNS is distinct for different members of the series.

Let the lowest state of a L^{Π} series be called a first-state denoted as L_1^{Π}. It has been found that the first-states do not contain the DNS.[8,11] Therefore, their nodal structures can be foreseen if the INS have been clarified. Thereby, their features can be predicted more or less definitely.

In the coordinate space there are two important domains. One is the ETH-domain containing the ETH and its neighbourhood, including the other tetrahedrons GETH, TETH and CONE (a slight deformation of an ETH would lead to the other shapes). Another is the SQ-domain containing the square and its neighbourhood, including all coplanar two-dimensional structures. These two domains are on average lower in total potential energy than other domains, and would be the first choice of low-lying

states. Since He_4 is a very floppy system, the wavefunctions would be extensively distributed and would cover the favourable ETH- and SQ-domains. If no nodal surfaces appear in these domains, the distribution would be smooth and the associated internal energy $E_{\rm int}$ would be low. Whereas if a nodal surface appears, the $E_{\rm int}$ would be much higher because the nodal surface would cause a great increase in kinetic energy.

It is clear from Table 1 that if a state is ETH-accessible, it can access any shape in the ETH-domain. Similarly, an SQ-accessible state can access any shape in the SQ-domain. Thus, the both ETH- and SQ-accessible states, e.g. 0_1^+ and 4_1^+, are inherently nodeless in the two most important domains. Their wavefunctions can be optimized without suffering any constraint from symmetry. On the other hand, all the other $L \leq 4$ states contain more or less the INS ($L \geq 5$ states are not discussed in this paper). Consequently, the $E_{\rm int}$ of 0_1^+ and 4_1^+ would be lower than other states.

For 3_1^- the INS at the SQ and at the DIA–REC would cause an increase in energy. For 2_1^+ the INS at the ETH would also cause an increase in energy. Therefore their $E_{\rm int}$ would be higher than that of 4_1^+. For the remaining $L \leq 4$ states INS exist both in the ETH- and SQ-domains. Their $E_{\rm int}$ would be much higher and are not likely to be bound (there may be an exception as discussed later).

It is clear now that the reason for the stronger binding of the 3_1^-, 4_1^+ and 2_1^+ states found in Ref.[8] is simply that little or no INS are contained in these states. This is a reason beyond dynamics, and is model-independent. Thus, these states are the most probable candidates of bound states of He_4.

Although the rotation energy $E_{\rm rot}$ of 4_1^+ is higher than that of 2_1^+, the $E_{\rm int}$ of 4_1^+ is considerably lower than that of 2_1^+. Thus, the total energy $E = E_{\rm int} + E_{\rm rot}$ of 4_1^+ might be close to that of 2_1^+. Similarly, the total energy of 3_1^- might also be close to that of 2_1^+. Thus these three states might be close to each other. They are definitely not members of a rotation band because they have distinct internal structures. The internal wavefunction of 4_1^+ is expected to be similar to 0_1^+ but more extensive in distribution due to the centrifugal barrier. The 3_1^- state is characterized by having an INS in the SQ-domain, while 2_1^+ is characterized by having an INS at the ETH shape.

It is recalled that in the previous four-boson model system 4_1^+ is lower than 0_2^+. It is noted that the energy difference $E_x(4_1^+)$ arises mainly from rotation. For the He-tetramer the size is very large, therefore the moment of inertia is also very large and the relative importance of the rotation energy is greatly reduced. Hence, it is very unlikely that 4_1^+ would be even higher than 0_2^+ in a system with a much larger moment of inertia. It has been found that the two bound 0_1^+ and 0_2^+ states of the He-tetramer have energies of $-0.389\,{\rm cm}^{-1}$ and $-0.0922\,{\rm cm}^{-1}$. It is very likely that 4_1^+ together with 3_1^- and 2_1^+ would be lower than the 3-1 threshold at $-0.0872\,{\rm cm}^{-1}$.[6] Hence, these three states are promising candidates of bound states.

Nonetheless, at this moment we cannot be sure that 4_1^+, 3_1^- and 2_1^+ are all bound. They (or some of them) might be resonances if they are higher than the 3-1 threshold. If all or some of them are resonances, they would emerge from the 3-1 channel, and would have a narrow width due to the centrifugal barrier. In particular, if the 4_1^+ is a resonance, it would be narrower and easier to identify due to the centrifugal barrier of the G-wave.

It is recalled that both the ground states of the trimer and dimer have angular momentum zero and parity even. Therefore both the 3-1 and 2-2 channels have $\Pi = (-1)^L$. Thus, for all the $\Pi = -(-1)^L$ states of the tetramer, no outgoing channels are available except those close to the break-up channel. With this in mind, let us observe again all the $\Pi = -(-1)^L$ states with $L \leq 4$. The 0_1^- and 1_1^+ states have to be dropped because they have too many INS. Among 2_1^-, 3_1^+ and 4_1^-, their $E_{\rm int}$ might be close to each other due to having similar numbers of INS, however 2_1^- might be lower because it has a smaller $E_{\rm rot}$. Although $E_x(2_1^-)$ must be higher than $E_x(2_1^+)$ due to having more INS, 2_1^- has a considerably higher outgoing threshold. Thus, 2_1^- is also a candidate for bound states.

In conclusion, this paper is devoted to a qualitative aspect. The four lowest $L \neq 0$ states (bound state or resonance) of He_4 are suggested to be the 4_1^+, 3_1^-, 2_1^+ and the extra 2_1^- states. The first three might have their energies close to each other, and they are found to be less constrained by symmetry, no (or little) INS are contained in them. This is the reason why they are lower. The energy of 2_1^- would be considerably higher; it might be bound simply because it has a much higher outgoing threshold.

The existence of the INS is a reality. The effect of the INS is absolutely not negligible. Thus, knowledge of the INS would help us to better understand the underlying physics. Incidentally, the argument presented in this paper is essentially model-independent.

Thus, the 3_1^-, 4_1^+ and 2_1^+ states would also be close to each other and would be the lowest $L \neq 0$ states of other four-boson systems. In heavier atomic clusters, e.g. Ne_4 and Ar_4, a stronger binding is expected. In these cases they have a much larger possibility to be bound.

Incidentally, the predicted ordering of levels of the first-states based on the inherent nodal structures has been found to coincide nicely with accurate theoretical calculation and/or experimental data, e.g. in the cases of four-valence-electron atoms,[12,13] dipositroniums[14,15] and quantum dots.[16] Nonetheless, it remains to check all the suggestions on the He-tetramer.

References

[1] Tang K T, Toennies J P and Yiu C L 1995 *Phys. Rev. Lett.* **74** 1546

[2] Aziz R A and Slaman M J 1991 *J. Chem. Phys.* **94** 8047

[3] Luo F, McBane G C, Kim G, Giese C F and Gentry W R 1993 *J. Chem. Phys.***98** 3564

[4] Schöllkopf W and Toennies J P 1994 *Science* **266** 1345

[5] Schöllkopf W and Toennies J P 1996 *J. Chem. Phys.* **104** 1155

[6] Blume D and Greene Chris H 2000 *J. Chem. Phys.* **112** 8053

[7] González-Lezana T *et al* 1999 *J. Chem. Phys.* **110** 9000

[8] Li X G and Bao C G 2001 *Few-Body Systems* **31** 51

[9] Bao C G 1992 *Few-Body Systems* **13** 41

[10] Bao C G 1993 *Phys. Rev.* A **47** 1752

[11] Bao C G, Li X G and Xie W F 1998 *Few-Body Systems* **23** 201

[12] Bao C G 1998 *Phys. Lett.* A **250** 123

[13] Moore C E 1971 *Atomic Energy Levels* (Washington, DC: National Bureau of Standards) p 467

[14] Bao C G 1998 *Phys. Lett.* A **243** 215

[15] Kinghorn D B and Poshusta R D 1993 *Phys. Rev.* A **47** 3671

[16] Ruan W Y, Liu Y Y, Bao C G and Zhang Z Q 1995 *Phys. Rev.* B **51** 7942