Spectral properties of endohedrally confined helium atom*

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Based on the B-spline basis method, the properties of the helium atom confined inside an endohedral environment, such as buckminster fullerene, are studied. In our calculations, the endohedral environment is a parabolic potential well. In this situation, the phenomenon of "mirror collapse" is exhibited for energy levels of a confined helium atom. The "giant resonance" of oscillator strength of the dipole transition emerges with the variation of depth of the confining well. The physical mechanisms of these phenomena are analyzed in this paper.

Keywords: B-spline, fullerene, mirror collapse, giant resonance

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1. Introduction

With the success of La@C_{60}[1] preparation, the experimental research has successively obtained K@C_{60},[2] He@C_{60},[3] and Ne@C_{60},[4] and so on. In such an environment, which is a double-well of the atomic potential, some novel properties of atoms appear. In the theoretical research of endohedrally confined atoms,[5–17] only Connerade,[7,9,11,13] Qiao,[14–16] and Zhang[17] have focused on their spectral properties. Meanwhile, some researchers have done some experiments on the spectral properties of endohedrally confined atoms.[18–23] In our work, a B-spline functions[24] method is applied to calculate the energy level and the dipole oscillator strength of endohedrally confined helium, when the square potential well is changed into a parabolic potential one. The causes of the "mirror collapse" and "giant resonance" phenomena are analyzed.

This paper is organized as follows. In Section 2, the confined helium model and the B-spline functions are introduced. The results of energy levels and oscillator strength of the endohedrally confined helium are given in Section 3. This paper is closed by the conclusions in Section 4.

2. Method

For a helium atom in an endohedral environment, the Hamiltonian is:

$$\hat{H} = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{r_{12}} + V(r), \tag{1}$$

where $V(r)$ is the attractive potential shell. In this paper, the shape of the attractive potential is changed to soften the endohedral environment. The attractive potential shell is defined as a parabola form

$$V(r) = \begin{cases} V_0 \dfrac{r^2 - (r_A + r_B)r + r_A r_B}{(r_B - r_A)^2}, & (r_A \le r \le r_B), \\ 0, & (\text{otherwise}), \end{cases} \tag{2}$$

where the parameters r_A and r_B are the inner and outer radii of the confining potential well, and the values are 5.75 a.u. (the unit a.u. is the abbreviation of atomic unit) and 7.64 a.u.,[25] respectively. In quantum mechanics, the equivalent potential of an electron is

$$V_e(r) = \frac{l(l+1)}{r^2} - \frac{Z}{r} + V(r). \tag{3}$$

Figure 1 shows the radial distribution of the equivalent potential of the $^1P_1$ states. It is obviously a double well structure. The Coulomb well is called the inner well and the confining well is called the outer well. The parameter V_0 is the depth of the outer well.

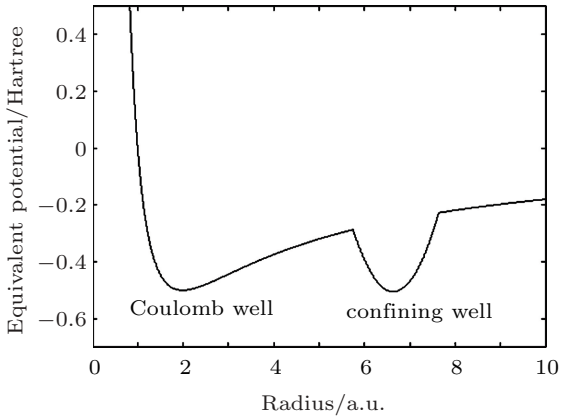


Fig. 1. Equivalent potential curve of the $^1P_1$ state. The unit 1 Hartree = 27.2114 eV.

Normally, the radial wave-functions are expanded in three spatial ranges: $0 < r \le r_A$, $r_A < r \le r_B$, and $r_B < r \le \infty$. In

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this paper, B-spline functions are used to expand the wave-functions and the boundary conditions are satisfied very simply.

With the order k, the B-spline functions are defined as

$$B_{i,1}(x) = \begin{cases} 1, & (t_i \le x \le t_{i+1}), \\ 0 & (\text{otherwise}), \end{cases} \tag{4}$$

$$B_{i,k}(x) = \frac{x - t_i}{t_{i+k-1} - t_i} B_{i,k-1}(x) + \frac{t_{i+k} - x}{t_{i+k} - t_{i+1}} B_{i+1,k-1}(x). \tag{5}$$

Here, $\{t_i\}$ is the knots sequence set on the x axis. In our work, the order k is set to 7, so the knots sequence is

$$t_1 = t_2 = \cdots = t_7 = 0, \tag{6}$$

$$0 < t_8 < \cdots < t_{29} = t_{30} = t_{31} = t_{32} = r_A, \tag{7}$$

$$r_A < t_{33} < \cdots < t_{35} = t_{36} = t_{37} = t_{38} = r_B, \tag{8}$$

$$r_B < t_{39} < \cdots < t_{49} = \cdots = t_{55} = r_{\max}. \tag{9}$$

The basis set is

$$\varphi_n = B_{i,k}(r_1) B_{j,k}(r_2) \Lambda^{LM}_{l_1 i l_2 j}(\Omega_1, \Omega_2) + (\boldsymbol{r}_1 \Leftrightarrow \boldsymbol{r}2), \tag{10}$$

which is used to expand the wave-functions of the system. From the Schrödinger equations, the matrix equation is obtained by

$$HC = EMC, \tag{11}$$

where H and M are respectively the Hamiltonian and overlap matrix. E and C are respectively the eigenvalues and eigenvectors, which can be obtained by solving the matrix equation (11).

In quantum mechanics, the oscillator strength of a dipole transition is calculated by

$$f_{\mathrm{i}\to\mathrm{f}} = \frac{1}{3} \Delta E \frac{1}{2J_{\mathrm{i}} + 1} |D_{\mathrm{if}}|^2, \tag{12}$$

where $\Delta E = |E_{\mathrm{f}} - E_{\mathrm{i}}|$, J_{i} is the initial state's total angular quantum number, D_{if} is the element of the dipole transition matrix and is defined by

$$D_{\mathrm{i}f} = \langle \psi_{\mathrm{i}} | \boldsymbol{r} | \psi_{\mathrm{f}} \rangle. \tag{13}$$

As a consequence, for a specific dipole transition, the oscillator strength is determined by two terms: the energy difference and the square of the dipole transition matrix element.

3. Results and discussion

By solving Eqs. (1) and (11), the energy levels of the endohedrally confined helium have been calculated. In our work, 11760 basis sets are included in the expansion of the wave function. As a validation of the B-spline functions methods, we get the ground state energy to -2.903703 Hartree in the case that the depth of the outer potential well is 0 Hartree. The reliability of the method was verified.

3.1. Energy levels

In our calculations, V_0 is taken from 0 to 6 Hartree in order to study the helium atom confined in buckminster fullerene more generally. The results of the calculation of the energy levels of the $^1S_0$ and $^1P_1$ states of a confined helium, as a function of the depth of the confining well, are shown in Fig. 2 and Fig. 3.

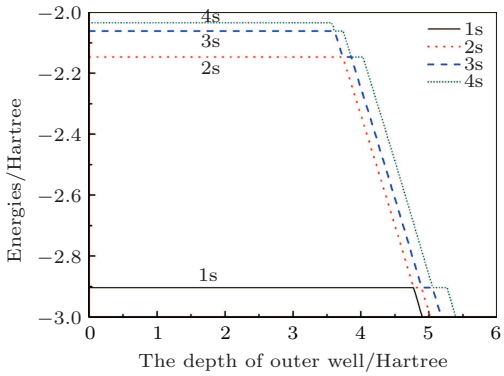


Fig. 2. (color online) $^1S_0$ states' energy levels for the helium confined in an attractive well, as a function of the depth of the outer well, with $r_A = 5.75$ a.u. and $r_B = 7.64$ a.u.

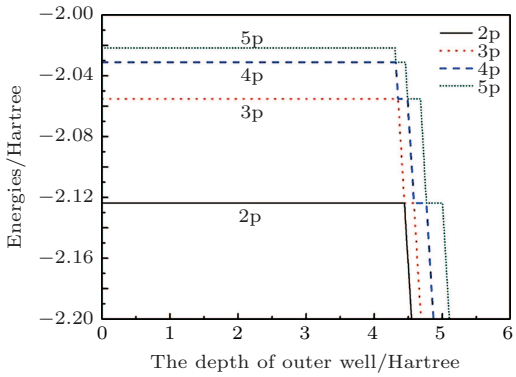


Fig. 3. (color online) $^1P_1$ states' energy levels for the helium confined in an attractive well, as a function of the depth of the outer well, with $r_A = 5.75$ a.u. and $r_B = 7.64$ a.u.

As shown in Fig. 2 and Fig. 3, the energy levels of the $^1S_0$ and $^1P_1$ states of helium reveal similar phenomena. Now, as an example, the $^1S_0$ states are used to describe the phenomena. It can be seen that a few of $^1S_0$ states hold the energies of an unconfined helium atom when V_0 is small. It seems that the outer confining well has no effect on them. The phenomenon can be understood by the distribution of the radial wave-functions of these $^1S_0$ states. Figure 6(a) shows the one-body densities of the 1^1S_0 and 2^1S_0 states, which can represent the distributions of these states, with $V_0 = 1.0$ Hartree. This reveals that the radial wave-functions of these $^1S_0$ states are almost zero in the range of the outer well. So the outer confining well has no effect on them. New $^1S_0$ states emerge when the depth of the outer confining well continues to increase, and their radial wave-functions are distributed in the range of the outer well. Thus, new states are bound in attractive confining wells, but the original states are still bound in Coulomb wells. The new

$^1S_0$ states will change the existing energy sorting. For example, when a new 1^1S_0 state emerges, the original 1^1S_0 state will become a 2^1S_0 state, and 2^1S_0 states will become 3^1S_0 states, and so on.

In quantum mechanics, the condition that a system has bound states is that the depth of the confining well must achieve a certain value when the width of the confining well is fixed. In the double-well model, if the Coulomb potential is removed, the bound states will appear only when $V_0 \geq 1.018$ Hartree. To understand the influences of the Coulomb potential and the attractive confined well for the system energies, we use Table 1 and Table 2 to show the relationship between them. In Table 1, the energies of $^1S_0$ states of free helium, only confining attractive potential helium (Coulomb potential is removed) and double-well helium, are given respectively, with $V_0 = 4.0$ Hartree. The row of the 1^1S_0 state shows that the energies of the 1^1S_0 state of free helium and double-well helium are both -2.9037 Hartree. It seems that the outer well has no effect on the 1^1S_0 state. However, the 2^1S_0 state of double-well helium is a new state, whose energy is -2.3380 Hartree. This is the result of the cooperation of the Coulomb potential and the confining attractive potential.

Table 1. The energies of $^1S_0$ states of free helium, only confining potential helium (Coulomb potential is removed) and double-well helium, with $V_0 = 4.0$ Hartree.

Orbitals	E_{free}	E_{confined}	$E_{\text{double-well}}$
1^1S_0	-2.9037	-1.7368	-2.9037
2^1S_0	-2.1459	-1.6485	-2.3380
3^1S_0	-2.0613	-1.5263	-2.2494

Table 2. The V_{Coulomb} and V_{confined} are expectations of Coulomb potential and confining an attractive potential with $V_0 = 4.0$ Hartree. R is the ratio of the expectation of confining attractive potential to the expectation of total potential.

Orbitals	V_{Coulomb}	V_{confined}	R
1^1S_0	-6.753156	$-1.808361 \times 10^{-11}$	2.677801×10^{-12}
2^1S_0	-6.017686×10^{-1}	-2.862518	8.262936×10^{-1}
3^1S_0	$-6.0149648 \times 10^{-1}$	-2.8633745	8.264014×10^{-1}

In Table 2, the V_{Coulomb} and V_{onfined} are expectations of Coulomb potential and confining attractive potential with $V_0 = 4.0$ Hartree, respectively. R is the ratio of the expectation of the confining attractive potential to the expectation of the total potential. R is calculated by

$$R = V_{\text{confined}}/(V_{\text{confined}} + V_{\text{Coulomb}}). \tag{14}$$

It can be seen that the Coulomb potential makes the major contribution to the 1^1S_0 state, and the confining attractive potential makes the dominant contribution to the 2^1S_0 state. So the 1^1S_0 state is the original state, and the confining attractive potential contribution can be ignored. The 2^1S_0 state is a new state, and the Coulomb potential contribution is relatively small compared with the confining attractive potential. It is worth noting whether it is a new or an original state, the energy is the result of the combination of Coulomb potential and confining attractive potential.

Now let us focus on the analysis of the $^1S_0$ states. The 1^1S_0 state holds -2.9037 Hartree, which is the 1^1S_0 state energy of an unconfined helium atom. The first $^1S_0$ state energy line is a horizontal line, as the depth of the outer attractive well increases until its depth reaches to -4.773 Hartree. Then, the 1^1S_0 state energy decreases very quickly, and its energy shows linear decreasing with the deepening of the outer confining well. To understand this phenomenon, the one-body density of the 1^1S_0 state is calculated, as a function of the outer attractive well. For $V_0 = -4.771$ Hartree, the one-body density of the 1^1S_0 state is distributed around the nucleus, which indicates the first $^1S_0$ state is a free helium ground state, which can be seen from Fig. 6(b). For $V_0 = -4.773$ Hartree, the figure 6(c) shows that the one-body density of the 1^1S_0 state is distributed around the nucleus and the confining well, indicating that the 1^1S_0 state is the compound of a confining state and an unconfined helium ground state. In Fig. 6(d) with $V_0 = -4.775$ Hartree, the one-body density of the 1^1S_0 state is distributed in the range of the outer confining well, indicating that the first $^1S_0$ state is a bound state. The second $^1S_0$ state has a similar behavior as the first $^1S_0$ state has shown formerly. In Fig. 2, its energy holds -2.1459 Hartree when V_0 is small, which is the value of the 2^1S_0 state energy of the unconfined helium, and the state stays in the Coulomb well. At the point $V_0 = -3.728$ Hartree, the energy of the second $^1S_0$ state suddenly decreases and continues to decrease with the deepening of the outer confining well till $V_0 = -4.773$ Hartree. The state "collapses" into the confining well. Then, when $V_0 > -4.773$ Hartree, the state "collapses" into Coulomb potential and stays there. At the point $V_0 = -4.891$ Hartree, the state "collapses" into the outer attractive well again and stays there finally. Some other $^1S_0$ and $^1P_1$ states' behaviors have the same phenomena, which are shown in Fig. 2 and Fig. 3, respectively.

The phenomenon of "collapse" can also be understood from the ratio of the expectation of the confining attractive potential to the expectation of the total potential. Figure 4 shows that the ratio R is very small for the 1^1S_0 state when V_0 is small, which implies that the Coulomb potential is dominant for the 1^1S_0 state. At the point $V_0 = -4.773$ Hartree, the ratio R suddenly increases, which indicates that the 1^1S_0 state "collapses" into the confining well. Then, the ratio R is a large value when $V_0 \geq -4.773$ Hartree, indicating that the outer attractive well is dominant for the 1^1S_0 state. The 2^1S_0 state's ratio reveals the same phenomenon. The ratio R is small when the 2^1S_0 state is bound in the Coulomb well, and it is very large when the 2^1S_0 state is bound in the outer well. At the "collapse" point, the ratio R suddenly changes. The phenomenon also exists in other states of $^1S_0$ and $^1P_1$, which are shown in Fig. 4 and Fig. 5.

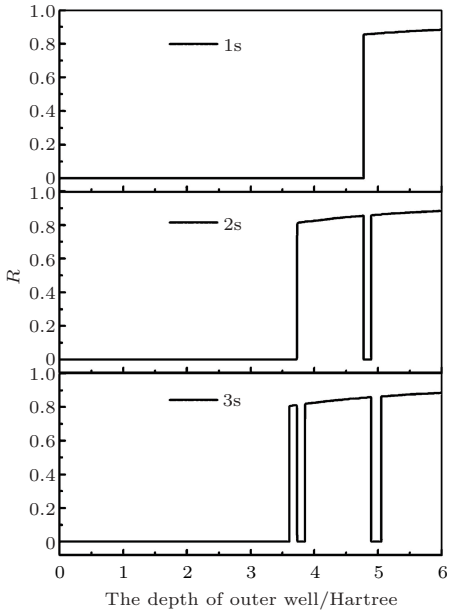


Fig. 4. Ratio R of $^1S_0$ states of the helium confined attractive well, changing the depth of the confining well. R is the ratio of the expectation of confined attractive potential to the expectation of total potential.

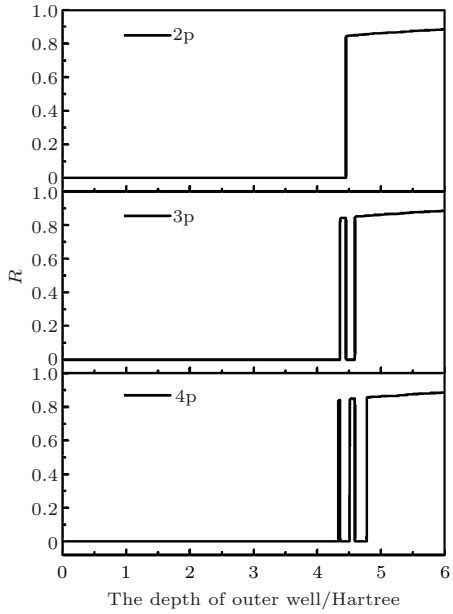


Fig. 5. Ratio R of $^1P_1$ states of the helium confined attractive well, changing the depth of the confining well. R is the ratio of the expectation of confined attractive potential to the expectation of total potential.

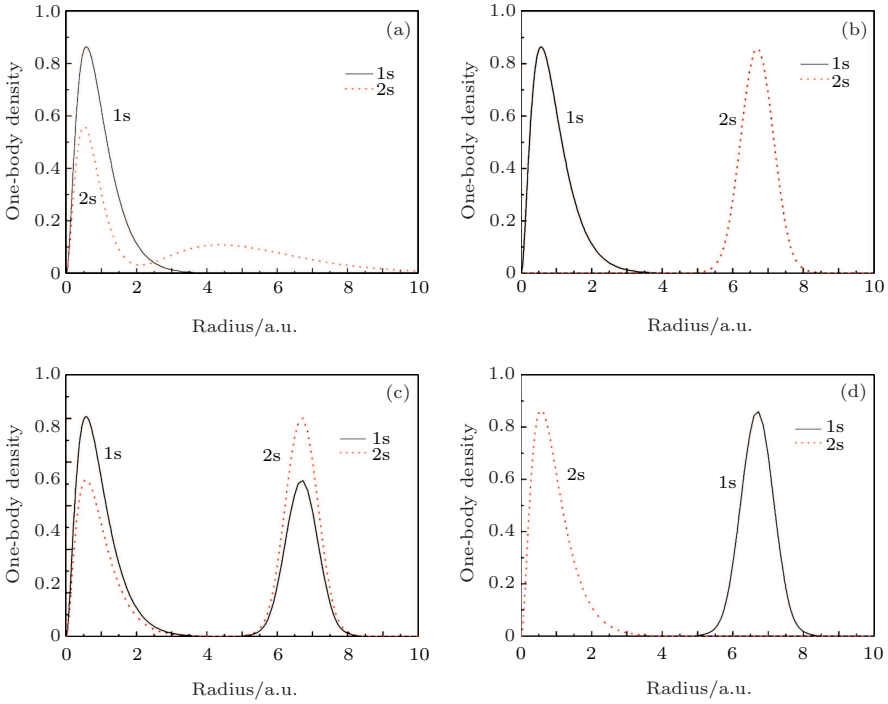


Fig. 6. (color online) One-body densities of the 1^1S_0 and 2^1S_0 states of endohedrally confined helium atom. The depth of an attractive well is: (a) $V_0 = 1.0$ Hartree, (b) $V_0 = 4.771$ Hartree, (c) $V_0 = 4.773$ Hartree, (d) $V_0 = 4.775$ Hartree, respectively.

At the "collapse" point, $V_0 = -4.773$ Hartree from Fig. 6(c), the first and the second $^1S_0$ states are both strong mixtures of an unconfined helium state and a confined helium state, which leads to the result that their energy lines do not cross. Thus, at the avoided crossing point, $V_0 = -4.773$ Hartree, the first $^1S_0$ state suddenly "collapses" into the outer confining well, which previously is bound in the inner Coulomb well. Simultaneously, the second $^1S_0$ state "collapses" into the inner Coulomb well, which previously is bound in the outer confining well. The behaviors of the $^1S_0$ states are called "mirror collapse." Figures 2 and 3 show that other states also have the same phenomena. The point of avoided crossing is a cut-off point of the horizontal lines and the oblique lines, which reveals that those states are respectively bound in the inner Coulomb well and the outer confining well. It is worth noting that all oblique lines are parallel, which demonstrates that the energies of all states bound in the outer attractive well decrease at the same rate, i.e. at the same

rate as the deepening of the outer well.

3.2. Dipole oscillator strengths

The $^1S_0$–$^1P_1$ dipole oscillator strengths of the endohedrally confined helium are calculated with Eqs. (12) and (13) in our work. For a certain dipole transition, such as 1^1S_0–2^1P_1 dipole transition, the energy difference is certain, so the dipole transition matrix element determines the oscillator strength. The dipole transition matrix element is related to the overlap of the wave-functions of the states involved in the transition. However, these wave functions are closely related to their one-body densities. So, the dipole transition matrix element is closely related to the overlap of one-body densities of the states involved in the transition.

The 1^1S_0–2^1P_1 oscillator strengths are calculated and shown in Fig. 7(a) as functions of the depth of the confining potential well. When V_0 is a small value, 1^1S_0 and 2^1P_1 states are both bound in Coulomb potential and the overlap of one-body density of them is normal, so that the oscillator strength has the same value corresponding to that of an unconfined helium atom, which can be seen from Fig. 7(a) and Fig. 7(b). At $V_0 = -4.449$ Hartree, the 2^1P_1 state collapses into the outer well, but the 1^1S_0 state is still bound in the Coulomb well. Then the oscillator strength suddenly decreases to a very small value from Fig. 7(a), but it is not zero. The reason is that the overlap of one-body densities of 1^1S_0 and 2^1P_1 states is close to zero from Fig. 7(c) with $V_0 = -4.6$ Hartree. By increasing the depth of the outer well, when the depth $V_0 = -4.773$ Hartree, the 1^1S_0 state collapses into the outer well. In this case, from Fig. 7(d) with $V_0 = -4.9$ Hartree, 1^1S_0 and 2^1P_1 states are all bound in the outer well so that their one-body densities overlap almost completely. So the value of oscillator strength is a large value, which is two orders of magnitude greater than the normal value of an unconfined helium atom. This phenomenon of very strong resonance enhancement of oscillator strength is called "giant resonance." At the same time, from Fig. 8(a), the 1^1S_0 and 2^1P_1 states are bound in the Coulomb well when V_0 is small, so the oscillator strength is normal. At the point $V_0 = -4.6$ Hartree, the 1^1S_0 state is still bound in the Coulomb well, but the 2^1P_1 state is bound in the confining well. At that moment, the oscillator strength is very small. When $V_0 \geq -4.773$ Hartree, the 1^1S_0 and 2^1P_1 states are both bound in the outer well, the oscillator strength is enhanced and the phenomenon of "giant resonance" appears. Not only the oscillator strength of 1^1S_0–2^1P_1 dipole transition, but also other dipole transitions have the phenomenon about "giant resonance," for example, 1^1S_0–3^1P_1 dipole transition, 2^1S_0–3^1P_1 dipole transition, and so on. Figure 9 shows the results of the oscillator strength of 1^1S_0–3^1P_1 dipole transition and the corresponding one-body densities of 1^1S_0 and 3^1P_1 states in different depths of the outer attractive well. Figure 10 shows the results of the oscillator strengths of the dipole transitions of 2^1S_0–2^1P_1 and 2^1S_0–3^1P_1. Figures 8(b), 8(c), and 8(d) show the ratio R of states that are included in the dipole transitions, and corresponding to the 1^1S_0–3^1P_1 dipole transition, 2^1S_0–3^1P_1 dipole transition, and 2^1S_0–3^1P_1 dipole transition, respectively.

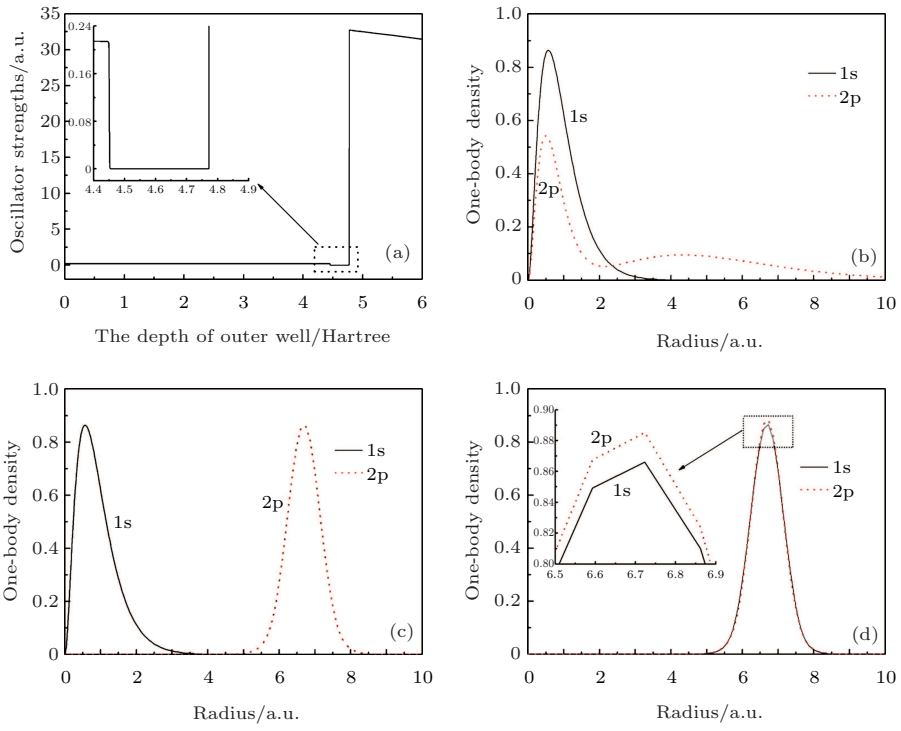


Fig. 7. (color online) (a) The 1^1S_0–2^1P_1 oscillator strengths of confined helium with changing depth of outer confining potential well; inset shows the enlarged figure of the dotted box. (b)–(d) one-body densities of 1^1S_0 and 2^1P_1 states of endohedrally confined helium atom. The depth of an attractive well is: (b) $V_0 = 4.4$ Hartree, (c) $V_0 = 4.6$ Hartree, (d) $V_0 = 4.9$ Hartree, respectively.

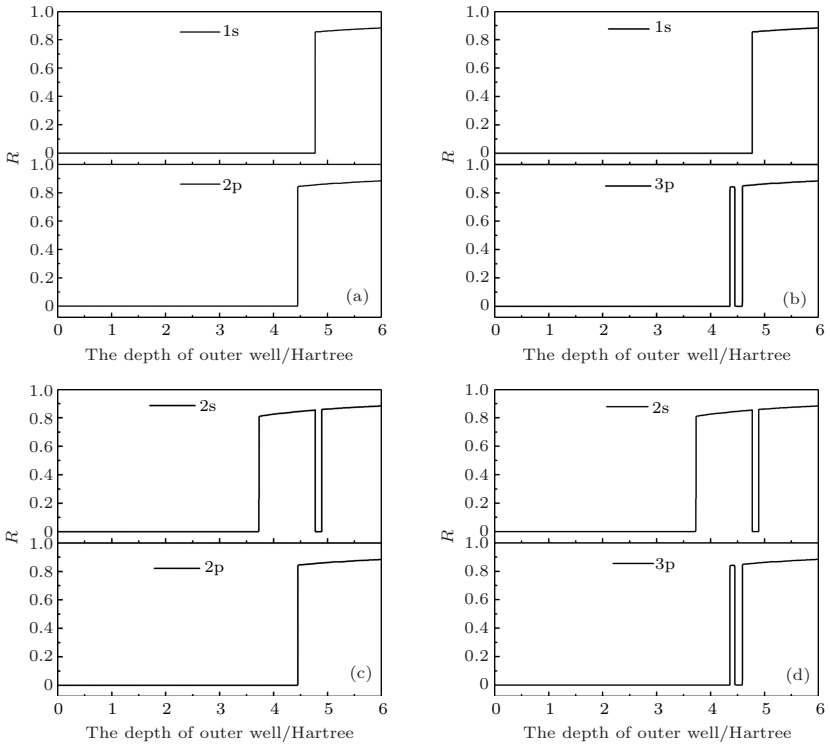


Fig. 8. The ratio of the states involved in the dipole transition. (a) The dipole transition of 1^1S_0–2^1P_1, (b) the dipole transition of 1^1S_0–3^1P_1, (c) the dipole transition of 2^1S_0–2^1P_1, (d) the dipole transition of 2^1S_0–3^1P_1.

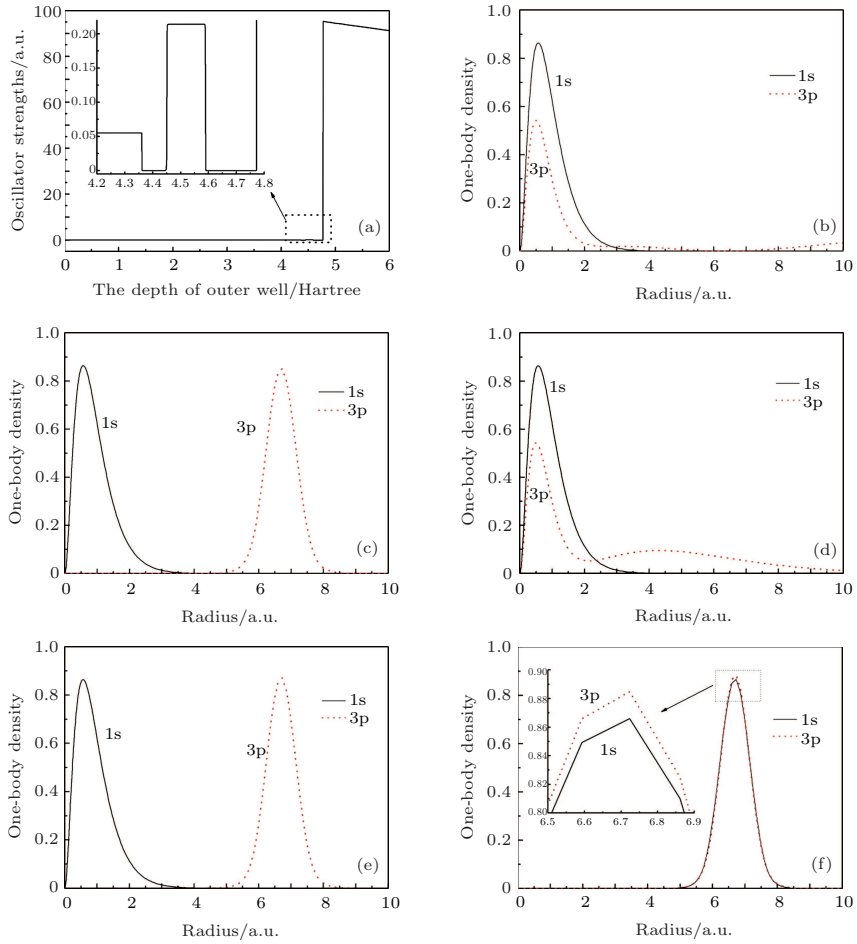


Fig. 9. (color online) (a) The 1^1S_0–3^1P_1 oscillator strengths of confined helium with changing depth of outer confining potential well; inset shows the enlarged figure of the dotted box. (b)–(e) one-body densities of 1^1S_0 and 3^1P_1 states of endohedrally confined helium atom. The depth of an attractive well is: (b) $V_0 = 4.3$ Hartree, (c) $V_0 = 4.4$ Hartree, (d) $V_0 = 4.5$ Hartree, (e) $V_0 = 4.7$ Hartree, (f) $V_0 = 4.9$ Hartree, in which the inset shows the enlarged figure of the dotted box, respectively.

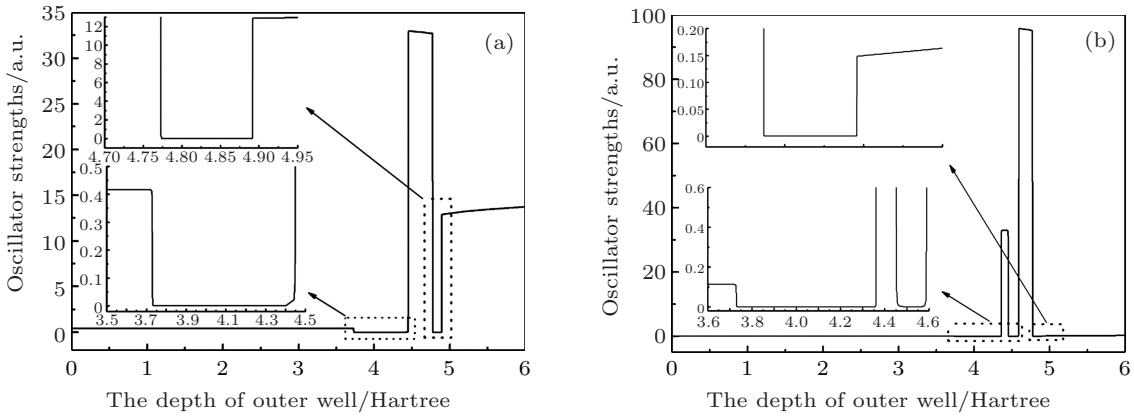


Fig. 10. The $2^1S_0 - 2^1P_1$ and 2^1S_0–3^1P_1 oscillator strengths of confined helium with changing depth of the outer confining potential well; inset shows the enlarged figure of the dotted box. (a) The 2^1S_0–2^1P_1 oscillator strength, (b) the 2^1S_0–3^1P_1 oscillator strength.

4. Conclusion

The spectral properties of an endohedrally confined helium atom were calculated with the B-spline functions method. With the evolution of the energy spectrum as a function of the depth of the outer attractive well, the phenomenon of "mirror collapse" exists in endohedrally confined helium. And the phenomenon of "giant resonance" of the dipole oscillator strength appears with the increase of the depth of the confining well. These phenomena are due to the jumps of the wavefunction between the Coulomb well and the confining attractive well, with the changing depth of the outer attractive well. It is readily apparent that the phenomena can also be understood from the major potential contribution changes between the Coulomb potential and the confining attractive potential, with the deepening of the outer attractive well.

The endohedral environment gives rise to some new questions about confined atoms. For example, the control of laser population, the prominent resonance in the photoionization cross section, and multiple photoionization, with endohedrally confined atoms.[26–28] Related work is being pursued.

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