Collisional properties of atoms in high-lying doubly excited states

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Using the hyperspherical wave functions of the high-lying doubly excited $^1S^e$ states of He, we have calculated the generalized oscillator strengths between these excited states and the Born cross sections for electron-impact excitation of He in these excited states within the manifold of $^1S^e$ states. Based on the rovibrator model of the doubly excited state, we predict theoretically that for these excitation processes a He atom tends to enlarge its size while keeping its shape, as a limber "e-He^{2+}-e" linear molecule, the same as that in the initial states.

Recently there has been considerable interest in the study of planetary atoms in which two or more electrons are excited to states of high principal quantum numbers.[1,2] These excited states have negligible overlap with the ground state or first few excited states. Consequently, the planetary atoms are not produced by direct (one photon) optical excitation from lower states. They are produced by electron impact or by recombination in dense plasma. Statistically they predominate over the states which are accessible by direct optical excitation. They are expected to have long lifetimes. There should be many interesting properties of these excited species that remain to be discovered and understood, since our present knowledge about these excited species is quite poor. Particularly intrashell doubly excited (Nl, nl') states with $n=N$ were quite difficult to treat theoretically because of the existence of a strong correlation between the excited electrons. Here N ($\leq n$) is the principal quantum number of an inner (outer) electron and l (l') is an angular momentum of the excited inner (outer) electron. It is also difficult to study them experimentally because they are not produced by the direct optical excitation.

Quite recently we have witnessed remarkable progress in the theoretical understanding of the structure of the planetary atoms,[3] particularly in intrashell doubly excited states with $N=n$. Herrick and Kellman[4] have proposed the supermultiplet structure of the intrashell doubly excited states and have also presented a rovibrational collective interpretation of the supermultiplet structures that gives a physical picture which is quite easy to grasp.[5] Berry and co-workers[6,7] have confirmed this rovibrator model of the doubly excited states using the graphic display of their electron-density distribution. Lin[8] has also investigated the high-lying doubly excited states using the hyperspherical coordinates[9] and has presented a classification scheme of doubly excited states based on the supermultiplet structures.[10] The high-lying doubly excited states have also been studied in connection with the Wannier theory for two-electron breakup just above the threshold energy for this process.[11,12] Furthermore, Buckman *et al.*[13] have presented the first experimental evidence for doubly excited states of He^- with $n\simeq N\gg 1$.

For the past decade, we have been studying the collisional properties of the high-rydberg atoms.[14] We hope to extend our study to the atoms in the planetary states, particularly in the high-lying doubly excited states. Now the time is ripe to make a theoretical study on collisional properties of the atoms in the planetary states, especially in the intrashell high-lying doubly excited states. For this purpose, we have tested the validity of the hyperspherical coordinate approach for the high-lying doubly excited states of H^- and He.[15] Using the hyperspherical adiabatic potentials diabatically connected at closely avoided crossings, we have found that our energy levels of the high-lying doubly excited states of He for $N=4$ are in good agreement with the elaborate calculations based on the method of complex coordinate rotation.[16]

Here we report for the first time a theoretical study on the collisional properties of He in the planetary states, particularly in the intrashell high-lying doubly excited states. As a first step, we have calculated the generalized oscillator strengths (GOS) within the intrashell high-lying doubly excited $^1S^e$ states of He using the hyperspherical wave functions previously obtained.[15] We have also evaluated the Born cross sections for inelastic scattering of He by electron-impact excitation and found a simple and interesting propensity rule for the optically forbidden excitations between the high-lying doubly excited $^1S^e$ states by electron impact based on the rovibrational collective interpretation of the supermultiplet structures.

The computation of the GOS is straightforward except that we have adopted the hyperspherical wave functions for the high-lying doubly excited states. The hyperspherical wave functions employed here are the same as those given in Ref. 15, except that a different set of functional values is employed in the numerical integration of the variable of R to ensure the accuracy of the calculated GOS.

To classify the doubly excited states, we employ a set of the quantum numbers $[N(K,T)^A n]$ according to Lin.[10] The quantum numbers K and T originate from the group-theoretical approach by Herrick and Sinanoglu[17] and specify the states of the angular correlation of the two-electron system. The quantum number K is a measure of $\langle -\mathbf{r}_< \cdot \hat{\mathbf{r}}_> \rangle$, i.e., the projection of the inner electron's radius vector onto the radial vector of the outer electron, while T^2 is proportional to $\langle (\mathbf{L}\cdot\hat{\mathbf{r}}_>)^2 \rangle$. According to the rovibrational interpretation of the supermultiplet structures, $v\ (=N-K-1)$ is the quantum number of the doubly degenerate bending vibrational modes, while T is the vibrational angular momentum around the mean axis of a limber e-He^{2+}-e linear molecule. The quantum number A introduced by Lin describes radial correlation at finite R. This quantum number is set equal to $+$ $(-)$ if the angular channel function has an antinode (node) at the potential ridge $\alpha=\pi/4$ where α is a hyper angle, i.e., $\alpha=\tan^{-1}(r_2/r_1)$. Other channels are assigned to $A=0$, where their channel functions do not have substantial amplitude at $\alpha=\pi/4$ for all R but reside always at $\alpha=0$ or $\pi/2$. They are similar to singly excited states.

In the following, we will discuss our results using the label $[N(K,T)^A n]$ described above. To our knowledge, there seems to have been no experimental or theoretical data available to be directly compared with our calculated results for the GOS for the high-lying doubly excited states. Therefore, to assess the accuracy of the present calculated results, we have computed the GOS for the 1^1S-2^1S transition, i.e., the transition from $[1(0,0)^+1]$ to $[1(0,0)^+2]$ in the present classification scheme of the energy levels and compared with the elaborate calculation of the GOS for this transition by Kim and Inokuti.[18] This comparison shows that our GOS differs from their calculation by several percent around the peak in the plot of the GOS versus the momentum transfer. This is consistent with the fact that our binding energies for the 1^1S and 2^1S states are in agreement with other elaborate calculations[19,20] within 1% because an expectation value of a physical quantity other than energy includes the first-order errors. On the other hand, our binding energies of the high-lying doubly excited states with $N=4$ based on the hyperspherical approach[15] are in agreement with the elaborate calculation by Ho[16] within a few percent. Therefore, we consider that the relative errors of the calculated GOS with $N\leq 6$ around the peak value are estimated to be at most about 10%.

The GOS is found to be small for $\Delta N\ (=N'-N)\geq 2$ because overlap of the initial and final hyperradial wave functions is negligibly small. This arises from the fact that the mean sizes of the excited atoms are substantially different from one another for $\Delta N\geq 2$. Figure 1 shows a typical example of the GOS for $\Delta N=1$, i.e., between the initial $[4(3,0)^+4]^1S^e$ state and the final $[5(4,0)^+5]^1S^e$ state. For the transition with $\Delta N=1$, the GOS has a large broad peak and some negligibly small side peaks. On the other hand, the GOS for $\Delta N=0$ is rich in structures, as is seen in Fig. 2, although its absolute value is substantially smaller than that for $\Delta N=1$. The physical origin of these structures has not yet been understood because the wave functions of the high-lying doubly excited states are more complicated than those of singly excited states.[21,22]

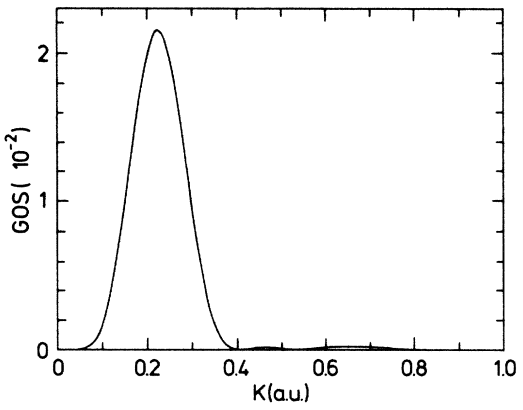

FIG. 1. Plot of the GOS against the momentum transfer between the initial $[4(3,0)^+4]^1S^e$ and the final $[5(4,0)^+5]^1S^e$ states.

To investigate the collisional properties of these excited species, we have calculated the Born cross sections of the atoms in the planetary states for inelastic collision by electron impact, i.e.,

$$e+\mathrm{He}^{**}(i)\rightarrow e+\mathrm{He}^{**}(f)\ ,$$

where i and f are specified by the labels $[N(K,T)^A n]^1S^e$ and $[N'(K',T')^A n']^1S^e$. The planetary states studied here are autoionizing ones with a finite decay time. However, the above collision processes are well defined because their collision times are usually short compared with the lifetimes of these excited states,[16] although it seems quite difficult to study them experimentally. To test the accuracy of our computation, we have calculated the Born excitation cross section for the 1^1S-2^1S transition using our GOS and compared it with that calculated from the GOS by Kim and Inokuti.[18] This comparison shows that our Born cross sections are in agreement with those calculated from their GOS within 1% accuracy in the energy range where the Born approximation is valid. Judging from the fact that our hyperspherical wave functions reproduce the

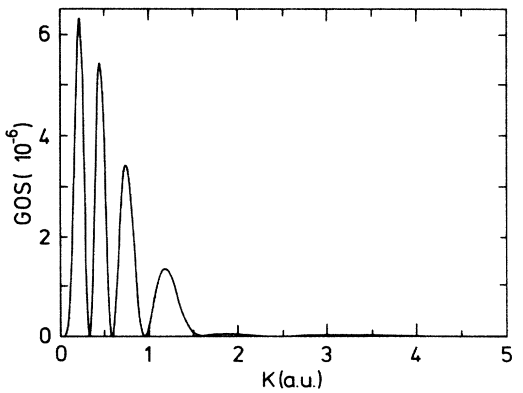

FIG. 2. Plot of the GOS against the momentum transfer between the initial $[4(3,0)^+4]^1S^e$ and the final $[4(1,0)^+4]^1S^e$ states.

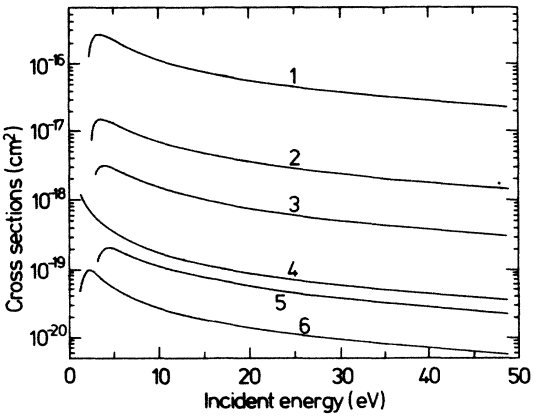

FIG. 3. Born cross sections of the atom in $[4(3,0)^+4]$, (0.4025) $^1S^e$ state for electron-impact excitation. The final $^1S^e$ states are (1) $[5(4,0)^+5]$, (0.2629), (2) $[5(2,0)^+5]$, (0.2422), (3) $[5(0,0)^+5]$, (0.2237), (4) $[4(1,0)^+4]$, (0.3666), (5) $[5(-2,0)^+5]$, (0.2000), and (6) $[4(-1,0)^+4]$, (0.3266) states, respectively. The numbers in parentheses are the binding energies of each excited state, in rydbergs, calculated from the channel functions constructed from 81 basis functions.

TABLE I. Relative order of the magnitude of the Born cross sections for the four initial states.

Initial $N(K,T)^A n$	v	Final $N(K,T)^A n$	v	Initial $N(K,T)^A n$	v	Final $N(K,T)^A n$	v
$4(3,0)^+4$	0	$5(4,0)^+5$	0	$5(4,0)^+5$	0	$6(5,0)^+6$	0
		$5(2,0)^+5$	2			$6(3,0)^+6$	2
		$5(0,0)^+5$	4			$6(1,0)^+6$	4
		$4(1,0)^+4$	2			$6(-1,0)^+6$	6
		$5(-2,0)^+5$	6			$5(2,0)^+5$	2
		$4(-1,0)^+4$	4			$5(-2,0)^+5$	6
						$5(0,0)^+5$	4
						$6(-3,0)^+6$	8
$4(1,0)^+4$	2	$5(2,0)^+5$	2	$5(2,0)^+5$	2	$6(3,0)^+6$	2
		$5(4,0)^+5$	0			$6(5,0)^+6$	0
		$5(0,0)^+5$	4			$6(1,0)^+6$	4
		$4(-1,0)^+4$	4			$6(-1,0)^+6$	6
		$5(-2,0)^+5$	6			$5(0,0)^+5$	4
						$6(-3,0)^+6$	8

energy levels of the high-lying doubly excited states, in agreement with those found by elaborate calculations[19,20] to within a few percent, we consider that our Born cross sections for the planetary states are reliable within an accuracy of several percent.

We have calculated the Born cross sections of the initial $[N(K,T)^+n]^1S^e$ states for electron impact excitation with $N \leq 6$ and $\Delta N \leq 1$, except for the transitions from $N=3$ to 4. As a typical example, we present the Born cross sections for the initial $[4(3,0)^+4]^1S^e$ state in Fig. 3. The largest cross sections are of the order of magnitude of 10^{-16} cm^2 for $N=4$ and 5. We have discovered some systematics for the excitation cross sections within the $^1S^e$ manifold. To see these systematics explicitly, we give the relative order to the magnitudes of the Born cross sections for the four initial states in Table I. In this table, we also give the vibrational quantum number v corresponding to the doubly degenerate bending vibrational motion of the limber e-He^{2+}-e linear molecule according to the rovibrational collective interpretation of the supermultiplet structures.[5]

Table I shows the following systematics for $\Delta L=0$, and with A unchanged, i.e., $A=+$: (i) the inelastic cross section σ for $\Delta N=1$ and $\Delta v\ (=v'-v)=0$ is the largest within the transitions for the fixed N and $\Delta N \leq 1$, (ii) σ decreases with an increase of $|\Delta v|$ for ΔN fixed, and (iii) $\sigma(\Delta N=1) \gg \sigma(\Delta N=0)$ for N and Δv fixed. However, for $N=2$, there are some exceptions (not shown here) for the systematics (ii). This arises from the fact that the rovibrator model may be less reliable for low N.

Summarizing the systematics found here, we have a simple propensity rule for the excitation of the atoms in the high-lying doubly excited states, by electron impact, in which the excitation with $\Delta v=0$ and $\Delta N=1$ take place favorably within the manifold of the $^1S^e$ excited states for $N \leq 6$. In other words, according to the rovibrator model, we can interpret that the atoms in the high-lying doubly excited $^1S^e$ states tend to enlarge their sizes while keeping their shapes, as the limber "e-He^{2+}-e" linear molecules, the same way as those in the initial states, i.e., $\Delta v=0$. Judging from the fact that there is strong similarity between the cases of $N=4$ and 5, we may conjecture that this propensity rule may apply to the cases with higher N. The prediction obtained here remains to be confirmed by experiment. Theoretical studies on $^1S^e$-$^1P^o$ and $^1S^e$-$^1D^e$ excitations are now in progress.

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[3]In Ref. 1, the atoms are called "planetary" in which two or more electrons are excited to the states of high principal quantum numbers, while in Ref. 2 only the doubly excited (Nl,nl') states with $n \gg N \gg 1$ are classified as planetary. We follow the original definition given in Ref. 1.

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