

Double autoionization of $\text{He}^-(2s^22p^2P)$

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The double autoionization rate for the $\text{He}^-(2s^22p^2P)$ hollow atom state is calculated using a nonperturbative time-dependent close-coupling method. The nine-dimensional wave function for the three electron atom is expanded in coupled spherical harmonics. The time-dependent Schrödinger equation is then reduced to a set of close-coupled partial differential equations for the three-dimensional radial expansion functions. Relaxation in imaginary time subject to orthogonality constraints yields a $2s^22p^2P$ hollow atom state with real energy, while propagation in real time yields total and double Auger rates. The total Auger rate is in reasonable agreement with previous saddle-point complex-rotation calculations. The double Auger rate is found to be 10% of the total Auger rate, in keeping with the relatively large double to total ratio found in recent experiments for $\text{He}^-(2s2p^2^4P)$.

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I. INTRODUCTION

Double autoionization is a highly correlated process involving three electrons moving in the Coulomb field of an atomic ion core. Recently a nonperturbative time-dependent close-coupling method was used to study the double autoionization of hollow atom states, in particular the $2s^22p^2P$ state in neutral Li [1]. It was found that the nonperturbative double Auger rate was lower than previous many-body perturbation theory results [2] and in better agreement with synchrotron experimental measurements [3]. The time-dependent close-coupling method has also been successfully applied to other four-body Coulomb problems, including the triple photoionization of Li and Bi [4,5] and the electron-impact double ionization of He and H^- [6,7].

Triply excited hollow atom states of He^- have been studied for many years since they were first observed in electron scattering experiments [8]. Recently, a number of He^- hollow atom resonances have been seen in experiments using merged beams of negative ions and synchrotron radiation [9]. Of interest was the report [10] of a measurement of a double Auger rate cross section for a particular $\text{He}^-(2s2p^2^4P)$ resonance and its unexpected large fraction of the total cross section, approximately 11%. Since our previous nonperturbative calculations [1] for the $\text{Li}(2s^22p^2P)$ hollow atom state yielded a ratio of double to total autoionization of 3.7%, it would be interesting to see what happens to the ratio when the nuclear charge is reduced by one, resulting in a more highly correlated four-body Coulomb problem. Along the way we will benchmark our nonperturbative method by comparison with the highly accurate saddle-point complex-rotation calculations for the total autoionization rate [11,12]. In Sec. II we review the time-dependent close-coupling theory for the double autoionization of atoms, in Sec. III we present double and total Auger rates for $\text{He}^-(2s^22p^2P)$, and in Sec. IV we conclude with a brief summary. Unless otherwise stated, all quantities are given in atomic units.

II. THEORY

The total wave function, Ψ , for an excited state of a three-electron atom is obtained by relaxation of the time-dependent Schrödinger equation in imaginary time ($\tau=it$):

$$-\frac{\partial\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,\tau)}{\partial\tau}=H_{atom}\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,\tau), \quad (1)$$

subject to orthogonality constraints with all lower energy states. The nonrelativistic Hamiltonian is given by:

$$H_{atom}=\sum_i^3\left(-\frac{1}{2}\nabla_i^2-\frac{Z}{r_i}\right)+\sum_{i<j}^3\frac{1}{|\vec{r}_i-\vec{r}_j|}, \quad (2)$$

where $Z=2$ for He^- . If Ψ represents an excited resonance state, then propagation of the time-dependent Schrödinger equation in real time:

$$i\frac{\partial\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,t)}{\partial t}=H_{atom}\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,t), \quad (3)$$

will lead to autoionization, including both single and double Auger processes.

The total wave function, Ψ , for a given total angular momentum \mathcal{L} , may be expanded in coupled spherical harmonics:

$$\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,t)=\sum_{l_1,l_2,L,l_3}\frac{P_{l_1l_2Ll_3}^{\mathcal{L}}(r_1,r_2,r_3,t)}{r_1r_2r_3}W[(l_1,l_2)L,l_3]\mathcal{L}, \quad (4)$$

where l_i are the electron angular momenta. Angular reduction of the time-dependent Schrödinger equations, Eqs. (1) and (3), results in a coupled set of partial differential equations for the radial expansion functions, $P_{l_1l_2Ll_3}^{\mathcal{L}}(r_1,r_2,r_3,\tau)$ and $P_{l_1l_2Ll_3}^{\mathcal{L}}(r_1,r_2,r_3,t)$ respectively, see Eqs. (3) and (4) of Ref. [1] for more details. The resulting time-dependent close-coupled equations are solved by standard numerical methods

to obtain a discrete representation of the radial functions and all operators on a three-dimensional lattice. Our implementation on massively parallel computers is complete domain decomposition by partition over all radial coordinates.

During the evolution of the wave function using Eq. (3), the probability for double autoionization is obtained by projection of a simple product of Ψ for a given \mathcal{L} and a total spin function for a given \mathcal{S} onto fully antisymmetric products of spatial and spin functions representing a $Z=2$ hydrogenic $1s$ bound orbital and two outgoing $Z=2$ hydrogenic Coulomb waves. The double autoionization probability is given by:

$$\mathcal{P}_{double}^{\mathcal{L}\mathcal{S}}(t) = \sum_{l_1, l_2, L, l_3} \sum_{s_1, s_2, S, s_3} \int_0^\infty dk_2 \int_0^\infty dk_3 \times |\mathcal{P}_{l_1 l_2 L l_3, s_1 s_2 S s_3}^{\mathcal{L}\mathcal{S}}(1s, k_2 l_2, k_3 l_3, t)|^2. \quad (5)$$

The partial-wave probabilities, $\mathcal{P}_{l_1 l_2 L l_3, s_1 s_2 S s_3}^{\mathcal{L}\mathcal{S}}$, are given in terms of angular and spin factor combinations of the six possible permutations of the function:

$$R(ijk, t) = \int_0^\infty dr_1 \int_0^\infty dr_2 \int_0^\infty dr_3 P_{1s}(r_i) P_{k_2 l_2}(r_j) \times P_{k_3 l_3}(r_k) P_{l_1 l_2 L l_3}^{\mathcal{L}}(r_i, r_j, r_k, t), \quad (6)$$

where $P_{kl}(r)$ are $Z=2$ hydrogenic continuum orbitals and $s_1 = s_2 = s_3 = \frac{1}{2}$, see Eqs. (9) and (11) of Ref. [1] for more details. We note that the coupled channel expansion over quantum numbers l_1, l_2, L, l_3 for Ψ is different from the summation over projection quantum numbers $l_1, l_2, L, l_3, s_1, s_2, S, s_3$ for \mathcal{P}_{double} . To avoid contamination from the continuum piece of the two-electron bound state wave functions, we restrict the sums over the electron momenta, k_2 and k_3 , to those approximately satisfying the conservation of energy, which for the double autoionization of $\text{He}^-(2s^2 2p^2 P)$ is given by:

$$E[\text{He}^-(2s^2 2p)] = E[\text{He}^+(1s)] + \frac{k_2^2}{2} + \frac{k_3^2}{2}. \quad (7)$$

Finally, the double autoionization rate is given by:

$$A_{double}^{\mathcal{L}\mathcal{S}} = \frac{\partial \mathcal{P}_{double}^{\mathcal{L}\mathcal{S}}(t)}{\partial t}. \quad (8)$$

III. RESULTS

Energy levels for selected configurations in the He isonuclear sequence are shown in Fig. 1. The He^- energies in the figure are from previous saddle-point complex-rotation calculations [12], while Hartree-Fock and hydrogenic energies are used for He and He^+ , respectively. The $\text{He}^-(2s^2 2p^2 P)$ triply excited state has its strongest single Auger transitions to $\text{He}(1s 2s^1 S)$, $\text{He}(1s 2p^1 P)$, and $\text{He}(1s 2p^3 P)$ [12]. The double autoionization of $\text{He}^-(2s^2 2p^2 P)$ may only occur to $\text{He}^+(1s)$ with an ejected energy of approximately 32.6 eV.

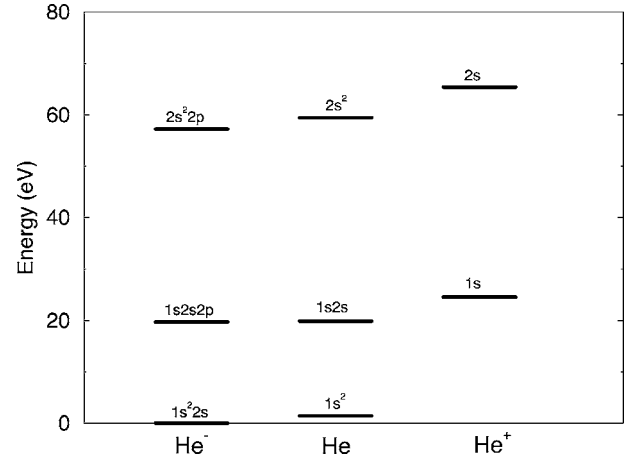


FIG. 1. Energy levels for selected configurations in the He isonuclear sequence.

The time-dependent close-coupling calculations for the double autoionization of $\text{He}^-(2s^2 2p^2 P)$ employed a $192 \times 192 \times 192$ point lattice in the (r_1, r_2, r_3) radial coordinates, with a uniform mesh spacing of $\Delta r = 0.20$. To check convergence we also employed a $256 \times 256 \times 256$ point lattice with a uniform mesh spacing of $\Delta r = 0.15$. We found that an overall box size of 38.4 was crucial for handling the large radial extent of the He^- hollow atom states. To include all $\Delta l = 1$ double promotions from the original $2s^2 2p$ configuration, we used 12 coupled channels $[(ss)Sp, (sp)Ps, (ps)Ps, (pp)Sp, (pp)Pp, (pp)Dp, (sp)Pd, (ps)Pd, (sd)Dp, (ds)Dp, (pd)Ps, (dp)Ps]$ and 10 corresponding projection functions $[(ss)^1 Sp, (ss)^3 Sp, (pp)^1 Sp, (pp)^3 Sp, (pp)^1 Pp, (pp)^3 Pp, (pp)^1 Dp, (pp)^3 Dp, (sp)^1 Pd, (sp)^3 Pd]$.

At time $\tau = 0$ the radial expansion functions needed to solve Eq. (1) are given by:

$$P_{l_1 l_2 L l_3}^{\mathcal{L}}(r_1, r_2, r_3, \tau = 0) = P_{2s}(r_1) P_{2s}(r_2) P_{2p}(r_3) \delta_{l_1, 0} \delta_{l_2, 0} \delta_{L, 0} \delta_{l_3, 1}, \quad (9)$$

$$P_{2s}(r_1) P_{2p}(r_2) P_{2s}(r_3) \delta_{l_1, 0} \delta_{l_2, 1} \delta_{L, 1} \delta_{l_3, 0}, \quad (10)$$

$$P_{2p}(r_1) P_{2s}(r_2) P_{2s}(r_3) \delta_{l_1, 1} \delta_{l_2, 0} \delta_{L, 1} \delta_{l_3, 0}, \quad (11)$$

where $\mathcal{L} = 1$ and $P_{nl}(r)$ are $Z=2$ hydrogenic bound orbitals. Relaxation of the coupled equations in imaginary time on a $(192)^3$ lattice used 20 000 time steps of $\Delta \tau = 0.04$, while a $(256)^3$ lattice needed 40 000 time steps of $\Delta \tau = 0.02$. To guard against relaxation to any state involving $1s$ character, a Schmidt orthogonalization is carried out at each imaginary time step, see Eq. (7) of Ref. [1] for more details. The total energy on the lattice for the $\text{He}^-(2s^2 2p^2 P)$ hollow atom state was found to be -0.794 a.u. for the $(192)^3$ lattice and -0.796 a.u. for the $(256)^3$ lattice, which compares well with a recent saddle-point complex-rotation value of -0.801 a.u. [12].

At time $t = 0$ the radial expansion functions needed to solve Eq. (3) are given by

$$P_{l_1 l_2 l_3}^{\mathcal{L}}(r_1, r_2, r_3, t=0) = P_{l_1 l_2 l_3}^{\mathcal{L}}(r_1, r_2, r_3, \tau=800). \quad (12)$$

Propagation of the coupled equations in real time on a $(192)^3$ lattice used 2500 time steps of $\Delta t=0.008$, while a $(256)^3$ lattice needed 5000 time steps of $\Delta t=0.004$. A mask function was employed at the lattice boundary to prevent reflection of the large fast moving part of the wave function due to autoionization. The total autoionization probability is given by:

$$\mathcal{P}_{total}^{\mathcal{L}}(t) = 1.0 - \sum_{l_1, l_2, l_3} \left| \int_0^\infty dr_1 \int_0^\infty dr_2 \int_0^\infty dr_3 \right. \\ \left. \times P_{l_1 l_2 l_3}^{\mathcal{L}}(r_1, r_2, r_3, t=0) P_{l_1 l_2 l_3}^{\mathcal{L}}(r_1, r_2, r_3, t) \right|^2. \quad (13)$$

The derivative of $\mathcal{P}_{total}^{\mathcal{L}}(t)$ yields a total autoionization rate for the $\text{He}^-(2s^2 2p^2 P)$ hollow atom state of 76 meV for the $(192)^3$ lattice and 72 meV for the $(256)^3$ lattice, which compares well with a recent saddle-point complex-rotation value of 72 meV [12]. Finally, using Eqs. (5)–(8), the double autoionization rate for the $\text{He}^-(2s^2 2p^2 P)$ hollow atom state was found to be 7.9 meV for the $(192)^3$ lattice and 7.2 meV for the $(256)^3$ lattice. Thus, the ratio of double to total autoionization for this particular resonance increases from 3.7 to 10.0% in going from Li to He^- . The ratio increase is in keeping with the idea that correlated four-body Coulomb

processes become stronger for three electron atoms as the nuclear charge is reduced.

IV. SUMMARY

In conclusion, we have applied a nonperturbative time-dependent close-coupling method to calculate the double autoionization rate for the $\text{He}^-(2s^2 2p^2 P)$ hollow atom state. The energy and the total autoionization rate for $\text{He}^-(2s^2 2p^2 P)$ agree well with recent highly accurate saddle-point complex-rotation calculations [12]. The ratio of the double autoionization rate to the total autoionization rate for $\text{He}^-(2s^2 2p^2 P)$ is found to be 10%, a factor of almost three higher than that found earlier for $\text{Li}(2s^2 2p^2 P)$ [1]. The large ratio is in keeping with recent experimental findings [10] of a large double to total Auger cross section ratio for a similar He^- triply excited hollow atom state. In the future, we plan to continue the development of the time-dependent close-coupling method for double Auger processes, especially in regard to its ability to efficiently locate and converge to $\mathcal{L}\mathcal{S}$ states within a triply excited configuration.

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