Positron-Atom Complexes as Quantum Halo States

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The wave functions of a number of positron-atom complexes are analyzed and three of the systems, namely, e^+Be, e^+Na, and e^+He($^3S^e$), are seen to exhibit quantum halo structures with 45%–50% of their probability distribution lying in the large r classically forbidden region. The mean square distance between the large r fragments ($e^+ +$ Be, Ps + Na$^+$, Ps + He$^+$) for these systems range from 1.8 to 2.2 times larger than the square of the classical turning point, another indication of their halolike nature.

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Quantum halo states are a well known feature in nuclear structure physics [1]. They were first used to describe certain weakly bound systems in nuclear physics [2,3] and are characterized by having a large spatial extent with a large proportion of the wave function in the classically forbidden region. Thus the ability of quantum particles to tunnel into potential barriers is a significant factor in the existence of halo states. The large separation means the wave function for an effective two-body system can be regarded as two well defined clusters (e.g., a neutron plus Be^{10} in the case of Be^{11}). One recent quantitative definition [1,4] for a two-body halo state has two criteria: (i) There must be a large probability, f_c, for finding a pair of independent cluster components in the total many-body wave function. (ii) A large fraction, f_h, of the total probability density must be in the classically forbidden region of the interaction. The critical values of f_c and f_h used to denote a quantum halo have been assigned to 1/2. In the present work, the structures of a number of positron-atom bound states [5] such as e^+Be and PsNa$^+$ are analyzed, and by computing the radial density of the positron and explicitly computing the fractions f_c and f_h it is shown that these states exhibit halolike structures.

The choice of the fractions, f_c and f_h, is somewhat arbitrary since one sees a continuum of increasingly more pronounced halo effects as f_c and f_h increase in magnitude. One feature of halo states is that they have binding energies that are small when compared with the other components of the system. Another useful halo state parameter is the ratio $\langle r^2\rangle/R_0^2$. In this expression $\langle r^2\rangle$ is the mean square distance between the two objects making up the halo, while R_0 denotes the critical radius at which the motion of the objects becomes classically forbidden. A halo state is typically characterized by the condition $\langle r^2\rangle/R_0^2 > 2$ [1]. Other definitions of quantum halos have been used previously [6], but the definition used here is superior.

While there is widespread acceptance and interest in halo states in nuclear structure physics, the existence of quantum halos in atomic physics is not so widespread and the only unequivocal two-body halo states are some weakly bound diatomic molecules involving helium, e.g., ^{4}He$_2$, ^{4}He^{6}He, ^{4}He^{6}Li, etc. [7–9]. So far there are no known electronic structures of atoms that could be classified as halo states. All neutral atoms have an infinite number of Rydberg states with a valence electron located a long distance from the core. But these are not halo states since they are bound by an attractive Coulomb interaction, and thus the long-range penetration of the wave function into the classically forbidden part of the potential is not responsible for binding. The most likely candidates as halo states are ions with an electron or a positron attached to a neutral atom. The asymptotic potential here is the attractive $\alpha_d/(2r^4)$ polarization potential (α_d is the static dipole polarizability). While the polarization potential does contribute to binding, the $1/r^4$ decay of the potential at long distances means that it has less impact than the Coulomb potential in binding the particles to the atom. The potential range is defined to terminate at the boundary of the classically forbidden region; thus $\varepsilon = \alpha_d/(2R_0^4)$ can be used to define the critical radius (ε is the binding energy).

At first sight there are two classes of atomic systems that might be good candidates to be halo states. The first class includes the weakly bound $^2P^o$ negative ions of the alkaline-earth atoms [10]. However, these states are not believed to have the spatial extension required of halo states [4]. The other set includes the more weakly bound positronic atoms such as e^+Li, e^+Be, and e^+Na [5] (a more heuristic definition of e^+Li and e^+Na would be as PsLi$^+$ and PsNa$^+$).

This Letter analyzes an existing set of positronic atom wave functions (that had been generated during previous investigations about the stability of these atoms), and uses them to explicitly compute f_c and the electron-positron probability densities to determine whether any of them exhibit features characteristic of halo states. The wave functions for e^+Li [11,12], e^+He($^3S^e$) [13,14], e^+Na [11,15], e^+Be [11,16], e^+Cu [17,18], and e^+Mg [11,16] were all computed using the fixed core stochastic variational method (FCSVM) [11,19].

The FCSVM uses a basis of explicitly correlated Gaussians (ECG) [11,19]. In the FCSVM, the Hamiltonian is diagonalized in a large basis of energy optimized ECGs. The core is represented by a Hartree-

Fock wave function which stays frozen during the calculation, and the core-direct and core-exchange matrix elements are evaluated exactly. Orthogonality of the valence electrons with the core is enforced with an orthogonalizing pseudopotential [11,20]. Defining the overlap with the core as $\langle P \rangle = \langle \Psi | \sum_{i \in \text{core}} |\phi_i\rangle\langle\phi_i| |\Psi\rangle$, this is less than 10^{-10} for all of the present systems. The core-polarization interactions use a semiempirical potential tuned to give the experimental binding energies of the core + electron system [11]. This ensures the accuracy of the core-valence electron interaction, and the present model Hamiltonian generally predicts single electron ionization energies, resonant oscillator strengths, and polarizabilities to an accuracy of 1% [11,21]. The positron is not very sensitive to the fine details of the positron-core interaction since the repulsive nuclear interaction keeps it away from the core. The dominant influence in binding the positron is the interaction with the accurately modeled valence electrons(s) [5,11]. For example, the valence electron polarizabilities for Be and Mg are about 100 times larger than their core polarizabilities [21,22], and complete omission of the entire core-polarization potential only changes the positron binding energies of e^+Be and e^+Mg by 2.5% and 6%, respectively. There are two positronic systems, namely, e^+Li and e^+He($^3S^e$), for which explicit comparisons with fully *ab initio* calculations can been made [11,12,14]. The FCSVM dissociation energies and expectation values are within 1% of those given by close to exact *ab initio* wave functions. The binding energies of all systems are converged to an accuracy of 10^{-5} hartree (with the exception of e^+Mg where it is 10^{-3} hartree) with respect to further enlargement of the ECG basis.

Gaussian-type basis sets have been criticized as not being suitable for the representation of the large r part of the wave function which decays as $\exp(-kr)$ [1,23] ($k = \sqrt{2\varepsilon}$ for e^+A type systems and $k = 2\sqrt{\varepsilon}$ for PsA^+ type systems). This was not a problem here since there were few active particles (three at most) and the dimensionality of the basis sets was large. However, additional ECGs, with parameters chosen to represent the large r asymptotic region were added to the basis for the four most weakly bound systems [i.e., e^+He($^3S^e$), e^+Na, e^+Be, e^+Li] to more precisely characterize the large r behavior.

An examination of expectation values suggests that these systems have well defined clusters. Both e^+Na and e^+He($^3S^e$) can be well described by a weakly bound Ps cluster orbiting a residual charged core with the mean electron-nucleus and positron-nucleus distances exceeding $15a_0$. The spin-averaged annihilation rates for both systems are about 1.9×10^9 s^{-1} [5], only 5% smaller than that of the Ps ground state. The mean electron-positron separations, $\langle |\mathbf{r}_{e^+} - \mathbf{r}_{e^-}| \rangle$, are about $3.15a_0$, which is 5% larger than that of the Ps ground state. The electron and positron densities for e^+Na shown in Fig. 1 have the correct slope for $r > 20a_0$, indicating a good description of the wave function in the classically forbidden region.

For e^+Be the radial expectations suggest that the positron does not significantly distort the structure of the Be ground state cluster. The mean electron-nucleus distance for the valence electrons in Be is $2.571a_0$; in e^+Be it is $2.654a_0$. The mean electron-electron distance in Be is $4.188a_0$; in e^+Be it is $4.214a_0$. Figure 2 shows the bulk of the positron probability distribution lies outside the peak of the electron distribution. As expected, the log-linear form of the positron density has a constant slope for $r > 10a_0$.

The cluster probability, f_c, was computed exactly by projecting the positronic atom states onto the wave functions of their large r fragments. The lowest energy disso-

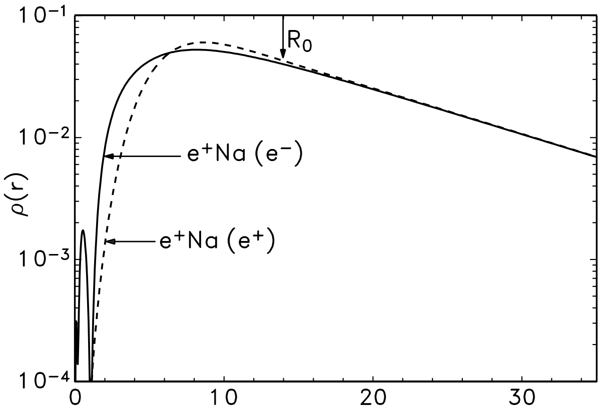

FIG. 1. The (valence) electron and positron radial densities for e^+Na as a function of r. The classical turning radius, R_0, is indicated. A least squares fit of the positron density from $r = 24a_0$ to $36a_0$ gives $k = 0.0433a_0^{-1}$ while $k = 0.0435a_0^{-1}$ is expected from the binding energy.

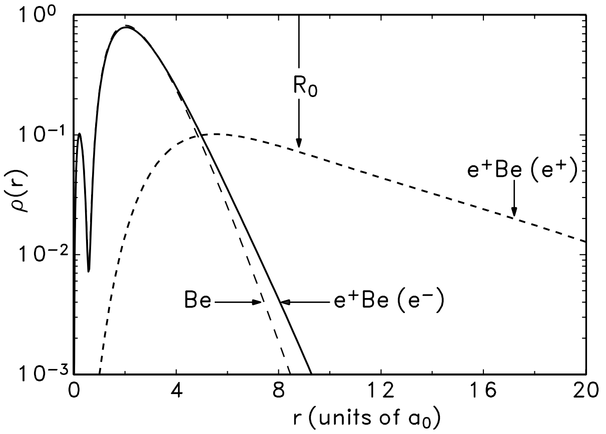

FIG. 2. The (valence) electron and positron radial densities for e^+Be as a function of r. The electron density for neutral beryllium (Be) is very similar to that of e^+Be. The classical turning radius, R_0, is indicated. A least squares fit of the positron density from $r = 12a_0$ to $20a_0$ gives $k = 0.0766a_0^{-1}$ while $k = 0.0795a_0^{-1}$ is expected from the binding energy.

ciation channel of a positronic atom depends on the parent atom ionization energy, I. If I is greater than 0.25 hartree (the binding energy of Ps), then the e^+A system dissociates into $e^+ + A$. When $I < 0.25$, the system dissociates into $\mathrm{Ps} + A^+$. The cluster fraction for e^+Cu is simplest to evaluate; it is

$$f_c = \int d^3r_0 \left| \int d^3r_1 \langle \Psi_{\mathrm{Cu}}(\mathbf{r}_1) | \Psi_{e^+\mathrm{Cu}}(\mathbf{r}_0, \mathbf{r}_1) \rangle \right|^2, \quad (1)$$

where $\mathbf{r}_1$ is the electron coordinate and $\mathbf{r}_0$ is the positron coordinate. The cluster fraction for e^+Na [or $e^+\mathrm{He}(^3S^e)$ and e^+Li] is written as

$$f_c = \int d^3R \left| \int d^3\rho \langle \Psi_{\mathrm{Ps}}(\boldsymbol{\rho}) | \Psi_{e^+\mathrm{Na}}(\boldsymbol{\rho}, \mathbf{R}) \rangle \right|^2, \quad (2)$$

where $\mathbf{R} = (\mathbf{r}_0 + \mathbf{r}_1)/2$ and $\boldsymbol{\rho} = (\mathbf{r}_0 - \mathbf{r}_1)$. [Note that an ECG basis permits an easy transformation from $(\mathbf{r}_0, \mathbf{r}_1)$ coordinates into $(\boldsymbol{\rho}, \mathbf{R})$ coordinates [24].]

Computation of f_c for e^+Be proceeds through the evaluation of

$$f_c = \int d^3r_0 \left| \int d^3r_1 d^3r_2 \langle \Psi_{\mathrm{Be}}(\mathbf{r}_1, \mathbf{r}_2) | \Psi_{e^+\mathrm{Be}}(\mathbf{r}_0, \mathbf{r}_1, \mathbf{r}_2) \rangle \right|^2. \quad (3)$$

The integrals over d^3r_1 and d^3r_2 were done analytically. However, the number of Gaussians in the final d^3r_0 integrand was $\approx (10^6)^2$, and this integral was done numerically.

The f_c coefficients for all systems are tabulated in Table I, and every system except e^+Mg has $f_c > 0.8$. One reason for this is the repulsive positron-nucleus potential. In the case of a system like e^+Be, this keeps the positron away from the region of space close to the nucleus where the electron density is largest.

The computation of f_h requires estimates of R_0, and this in turn needs estimates of the polarizabilities. The polarizability of Ps, $36a_0^3$, is known exactly. Explicit calculations of the polarizabilities of Be, Mg, and Cu using the FCSVM neutral atom ground state wave functions have not been done. However, large basis CI calculations using model Hamiltonians that are practically identical to those used for the FCSVM calculations have been performed [18,21]. The CI calculation gave $37.69a_0^3$ for Be [21], which is close to the current best estimate of the polarizability, namely, $37.76a_0^3$ [25]. The classically forbidden region is given by the condition $R_0 = \sqrt[4]{37.69/(2 \times 0.003161)} = 8.79a_0$, where 0.003 161 hartree is the e^+Be binding energy. The model atom polarizabilities, and critical radii, R_0, for e^+Mg and e^+Cu are given in Table I along with data for the $e^+\mathrm{He}(^3S^e)$, e^+Li, and e^+Na complexes.

Table I gives f_h for the systems considered in this Letter. This was determined for e^+Be, e^+Mg, and e^+Cu directly from a tabulation of the positron density versus r. For e^+Li, $e^+\mathrm{He}(^3S^e)$, and e^+Na, f_h was taken from the fraction of the electron and positron densities in the forbidden zone. All of the systems have values of f_h lying between 0.40 and 0.50. None of the systems satisfy the formal halo state definition, but three of the systems, $e^+\mathrm{He}(^3S^e)$, e^+Na, and e^+Be, show evidence of halo formation. These states have $f_h = 0.474$, 0.484, and 0.457, respectively, and in addition the values of the $\langle r^2 \rangle / R_0^2$ parameter are 2.05, 2.20, and 1.85, respectively.

The existence of more weakly bound positronic atoms would presumably result in states which exhibit the halo effect more strongly. One possible candidate would be mercury, which has a polarizability of $(33.91 \pm 0.34)a_0^3$ [26]. So if positron binding to Hg were possible, then it is likely that the resulting complex would be more weakly bound than e^+Be and exhibit a more prominent halo.

Besides atoms, there is the possibility of positron-molecule bound states possessing halos. Although there are few explicit calculations on positron-molecule bound states [27] other than e^+LiH [28–31], there is a growing body of evidence suggesting that positron-molecule bound states are responsible for the large annihilation rates observed when positrons are injected into a number of molecular gases [32–34]. Feshbach resonances consisting of positrons bound to vibrational excitations of the ground state are thought to be responsible for the enhancement of

TABLE I. Properties of positronic atoms relevant to the existence of quantum halos. All symbols have been defined in the text, and all values are given in atomic units. The source of the static dipole polarizability is noted for Be, Mg, and Cu. The mean value of r^2 is taken to be that of the positron for e^+Be and e^+Mg. For $e^+\mathrm{He}(^3S^e)$, e^+Li, and e^+Na; $\langle r^2 \rangle$ is taken to be the mean of the electron and positron expectation values.

System	Dissociation	α_d	ε	R_0	$\langle r^2 \rangle$	$\langle r^2 \rangle / R_0^2$	f_c	f_h
$e^+\mathrm{Be}^{\mathrm{a}}$	$e^+ + \mathrm{Be}$	37.69 [21]	0.003 161	8.79	142.7	1.85	0.908	0.457
$e^+\mathrm{Mg}^{\mathrm{a}}$	$e^+ + \mathrm{Mg}$	71.35 [21]	0.016 096	6.86	58.7	1.25	0.733	0.412
e^+Cu	$e^+ + \mathrm{Cu}$	41.65 [18]	0.005 597	7.81	95.97	1.57	0.821	0.470
$e^+\mathrm{He}(^3S^e)$	$\mathrm{Ps} + \mathrm{He}^+$	36.0	0.000 586 3	13.24	360.0	2.05	0.976	0.474
e^+Li	$\mathrm{Ps} + \mathrm{Li}^+$	36.0	0.002 479	9.23	120.6	1.42	0.938	0.419
e^+Na	$\mathrm{Ps} + \mathrm{Na}^+$	36.0	0.000 473 8	13.96	428.1	2.20	0.977	0.484

[a] Further optimizations of e^+Be and e^+Mg have occurred since [16] was published.

the annihilation rate [34,35]. These positron-molecule bound states are likely to be similar in character to e^+Be, with postulated energies ranging from 5×10^{-4} to 5×10^{-3} hartree [34]. It is probable that some of these positron-molecule bound states will have quantum halos.

Because a large fraction of a halo state lies outside the potential, size and other expectation values are expected to have a simple energy dependence. For example, the product $\varepsilon\langle r^2\rangle$ should be constant. This product gives 0.203 for e^+Na and 0.211 for e^+He($^3S^e$). That the product for e^+Li, 0.299, is 50% larger is expected since e^+Li has less penetration into the classically forbidden region. It is also known that the annihilation rates of a number of positron-atom bound states (with a parent atom with $I >$ 0.250 hartree) scale simply with energy [5,36], viz.

$$\Gamma \approx 8 \times 10^9 \sqrt{\varepsilon} \tag{4}$$

(where ε is in hartree and Γ is in s^{-1}). This relation exists because most of the positron wave function lies outside the electron charge cloud (note that the range of the electron charge cloud is smaller than that of the polarization potential). This is exactly the sort of relation that should be obeyed by a halo state.

In conclusion, three systems, e^+Be, e^+He($^3S^e$), and e^+Na, exhibit most of the features used to define a halo state, although they just fail to satisfy the formal criteria of a 50% probability of finding the particles in the classically forbidden region. One expects that the identification of more weakly bound positron-atom or positron-molecule complexes will result in states with more prominent quantum halos. The open question [1,4] of whether halo states can exist in physical systems with r^{-4}-type potential fields has for most purposes been answered in the affirmative.

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