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Bound states of positrons and neutral atoms

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We use many-body perturbation theory to examine the interaction of positrons with atoms. Our calculations predict positron s-type bound states with binding energies 0.87, 0.23, 0.35, and 0.045 eV for Mg, Zn, Cd, and Hg atoms, respectively. This binding is due to the positron-atom polarization potential and virtual positronium formation. A simple criterion is used to find other atoms that are likely to form bound states with positrons. Among the most probable candidates are Ti, V, Cr, Mn, Zr, Nb, and Mo.

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I. INTRODUCTION

The main aim of this paper is to suggest that neutral atoms can form bound states with positrons. The results of our calculations performed using atomic many-body theory methods show that a number of atoms (Mg, Zn, Cd, Hg, and, we believe, many others) are indeed capable of binding positrons. This binding is due to the correlation interaction between the positron and the atomic valence electrons. There are two physically distinct mechanisms contributing to the large positron-atom attraction. The first one is the dynamic polarization of the atom by the positron. The strength of this interaction is roughly proportional to the atomic dipole polarizability α. This interaction acts between an atom and an electron as well and is responsible for the stability of atomic negative ions. The second mechanism is the virtual formation of positronium (Ps) bound states by the positron and one of the valence electrons. The attraction it produces is similar to the stabilization of the symmetric state in a two-well potential. Indeed, the valence electron can be bound either to the positive atomic ion (with the binding energy equal to the ionization potential I), or to the positron (with the $|E_{1s}|$ binding energy, where $E_{1s}=-6.8$ eV is the Ps ground-state energy).

The possibility of bound-state formation of positrons with atomic and molecular systems has been extensively studied. Ore [1] proved the stability of positronium hydride PsH (or e^+H^-) and the existence of positron-anion bound states may be considered well established (see [2], and references therein). There is also experimental evidence for the existence of bound states of positrons with neutral molecules [3]. However, very few results have been obtained about the existence or nonexistence of bound positron-atom species. Aronson *et al.* [4] proved that positron binding to hydrogen is not possible which agreed with the earlier variational calculations of the e^+-H scattering phase shifts [5]. Gertler *et al.* [6] were able to show that ground-state helium would not bind a positron. Golden and Epstein [7] derived necessary conditions for the existence of bound states for positron-atom systems and showed that He, Ne, and N, in their ground states, are not capable of binding positrons, whereas the binding of positrons to H, O, Ar, and Kr could not be ruled out. Karl *et al.* [8] used the proton-atom potential and concluded that He, Li, N, O, F, Ne, Na, Al, and Ar probably do not bind positrons.

There have been a number of calculations that reported on positron–alkali-atom s-type bound states (e^+Li [9], e^+Li, e^+Na, and e^+K [10]), but while being bound in the positron-atom channel, these states are unstable to Ps emission. This is also true for the positron–alkaline-earth-atom (Ca, Sr, Ba, and Ra) bound states [11] which were obtained using the adiabatic dipole polarization potential calculated in the relativistic polarized orbital approximation. The same calculation yielded binding energies of 0.003 and 0.02 eV for Be and Mg, respectively, and the values of 5×10^{-4} and 0.0075 eV were obtained for Zn and Cd [12]. However, the accuracy of these predictions is doubtful since neither the use of the adiabatic approximation nor the neglect of other multipoles and the Ps formation can be justified in this problem.

In the present work the interaction of positrons with atoms is studied using the formalism of atomic many-body theory. Atomic many-body theory has not been applied to the type of double-well bound-state problem considered here. However, high accuracy has been obtained in the calculation of electron-atom scattering (see, e.g., [14–17]), positron-atom scattering [18,19], calculations of negative ion bound states [17,20–23], and calculations of the energy levels of atoms with one valence electron [24–26], and very high accuracy has been obtained. For example, the largest discrepancy with experimental values of the Cs energy levels was 0.005 eV [24] and was 0.01 eV for Fr [25]. The discrepancy between the experimental values of the binding energies of Pd$^-$ was 0.02 eV [17] and for He$^-$ was less than 0.01 eV [20].

II. CORRELATION POTENTIAL METHOD

Within the atomic many-body theory the problem of interaction between a ground-state atom and a positron (or an electron) can be reduced to a single-particle (Dyson) equation (see, e.g., [13])

$$(H_0+\Sigma_E)\psi_E=E\psi_E, \tag{1}$$

where ψ_E is the quasiparticle wave function describing the motion of the extra particle of energy E, H_0 is the single-particle Hamiltonian of the zeroth approximation, and Σ_E is a nonlocal energy-dependent potential:

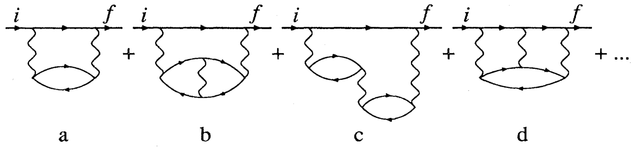

FIG. 1. Diagrammatic expansion of the positron-atom correlation potential $\langle f|\Sigma_E|i\rangle$. The thick line corresponds to the positron. Wavy lines show the Coulomb interaction between the positron and atomic electrons, and between the atomic electrons. Summation over all intermediate states is assumed. In the electron-atom case exchange diagrams must be added.

$$\Sigma_E\psi_E=\int\Sigma_E(\mathbf{r},\mathbf{r}')\psi_E(\mathbf{r}')d\mathbf{r}'. \tag{2}$$

$\Sigma_E(\mathbf{r},\mathbf{r}')$ describes the correlation interaction of the extra particle with the atom. It is equal to the self-energy operator of the single-particle Green's function. If Eq. (1) has a negative eigenvalue $E=\varepsilon_0<0$, a positron-atom bound state (or a stable negative ion if an electron is considered) exists.

It is convenient to choose H_0 to be the Hartree-Fock Hamiltonian of the atom. The correlation potential is then given by the diagrammatic expansion shown in Fig. 1. The main contribution to Σ_E is given by the second-order diagram, describing the polarization of the atom by the projectile (diagram *a* in Fig. 1). There are also important higher-order corrections to it (see, e.g., [22–24,26]), that take into account the electron-hole interaction and the screening of the Coulomb interaction in the excited atom (e.g., diagrams *b* and *c* in Fig. 1). Following [24,26] we can calculate the correlation potential in the second order in the Coulomb interaction between the projectile and atomic electrons, as shown in Figs. 2–4. The intra-atomic correlations of two types, namely, the electron-hole interaction and the screening of the Coulomb interaction, are taken into account within the polarization operator. The Feynman diagram technique and the Green function method are used to perform the all-order summation of the diagrams of the polarization operator and calculate Σ_E. The correlation potential thus calculated will be referred to as the *polarization potential* $\Sigma_E^{\rm pol}$ hereafter, since it produces the long-range asymptotic behavior

$$\Sigma_E^{\rm pol}(\mathbf{r},\mathbf{r}')\simeq-\frac{\alpha e^2}{2r^4}\delta(\mathbf{r}-\mathbf{r}'). \tag{3}$$

Note that the value of the atomic dipole polarizability α is determined by the polarization operator in Figs. 3 and 4, and corresponds to the random-phase approximation (RPA) for the excited atom. Although the dipole polarization [Eq. (3)] dominates in the asymptotic region, $\Sigma_E^{\rm pol}$ contains contributions from all multipoles, and the contributions of, say, monopole and quadrupole polarization of the target by the projectile are known to be quite important.

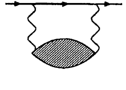

FIG. 2. Polarization contribution $\langle f|\Sigma_E^{\rm pol}|i\rangle$ to the correlation potential. In the polarization operator (shaded loop) the electron-hole interaction and screening of the Coulomb interaction are taken into account.

FIG. 3. Renormalization of the polarization operator by the residual Coulomb interaction ("screening").

The polarization potential $\Sigma_E^{\rm pol}$ (with the corresponding exchange diagrams included) was shown to be quite accurate for describing electron-atom interaction (see references in Sec. I). However, in the positron-atom case this approximation proved to be somewhat deficient [18,27]. The physical reason for this is the possibility for the positron to virtually form Ps bound states with the valence electrons. This process is represented diagrammatically by the series in Fig. 5. The contribution to the correlation potential that results from the sum of such series will be referred to as the *Ps-formation potential*, $\Sigma_E^{\rm Ps}$. In the case of electron-atom interaction the series in Fig. 5 is sign alternating and hence each successive term partially cancels the previous term, giving a small total. In the case of positron-atom interaction all the terms in the series are negative and the sum of the series of diagrams with 3, 4, 5, etc. electron-positron interactions gives an important contribution to the positron-atom attraction [27]. The result is represented by the diagram in the right-hand side of the diagrammatic equation in Fig. 5, where the shaded rectangle describes the propagation of the electron-positron pair.

There are physical reasons which make the exact calculation of this sum very difficult. First, the electron-positron pair has bound Ps states (i.e., poles in the propagation amplitude). This makes the problem nonperturbative, and therefore it is not possible to directly sum the series. Secondly, the correct description of the bound Ps states (even if they are formed virtually) involves electron and positron states with large orbital momenta in the ladder diagram series (a similar problem was mentioned in [28] where the authors had to include *l* up to 15 in their convergent close coupling calculation of the low-energy positron-hydrogen scattering). On the other hand, to calculate $\Sigma_E^{\rm Ps}$ we only need to describe the propagation of the correlated electron-positron pair (the shaded block in Fig. 5). It was suggested in Ref. [18] that the following expression can be used to approximate the sum of the ladder diagram series:

$$\langle f|\Sigma_E^{\rm Ps}|i\rangle=\sum_n\int\frac{\langle fn|V|\tilde{\Psi}_{1s,\mathbf{K}}\rangle\langle\tilde{\Psi}_{1s,\mathbf{K}}|V|ni\rangle}{E+\varepsilon_n-\left(E_{1s}+\dfrac{K^2}{2M}\right)+i\delta}\frac{d^3K}{(2\pi)^3}, \tag{4}$$

FIG. 4. Calculation of the electron-hole interaction in the polarization operator.

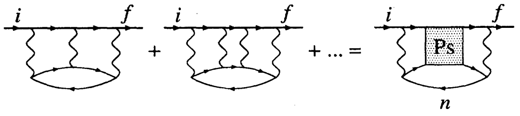

FIG. 5. Positronium-formation contribution $\langle f|\Sigma_E^{\mathrm{Ps}}|i\rangle$ to the correlation potential. The shaded block is the Ps Green function, which corresponds to the sum of the positron-electron ladder diagrams.

where $V=-1/|\mathbf{r}-\mathbf{r}_1|$ is the electron-positron Coulomb interaction, $\Psi_{1s,\mathbf{K}}=\varphi_{1s}(\mathbf{r}-\mathbf{r}_1)e^{i\mathbf{K}\cdot\mathbf{R}}$ is the wave function of the Ps atom in the ground state, moving with momentum $\mathbf{K}$, $E_{1s}+K^2/2M$ is the energy of this state, M is the mass of the Ps atom, n is the hole state, and ε_n is its energy in the Hartree-Fock approximation. The tilde above $\Psi_{1s,\mathbf{K}}$ indicates that this wave function is orthogonal to the single-electron states of the atomic ground state: $|\tilde{\Psi}_{1s,\mathbf{K}}\rangle=(1-\Sigma_n|n\rangle\langle n|)|\Psi_{1s,\mathbf{K}}\rangle$. This is necessary since the shaded block in the right-hand side of the diagrammatic equation in Fig. 5 is constructed from the excited electron states.

Since Eq. (4) is only an approximation for the series in Fig. 5, one may suspect that there is some "double counting" in using Ps intermediate states in addition to the electron and positron states in the diagram a in Fig. 1. However, this defect, if present, must be very small. First, the expression (4) has a much lower denominator. Second, it effectively involves contributions of high angular momenta $l\sim 2\pi r_+/r_{\mathrm{Ps}}$, where r_+ is the typical radius for the positron, and r_{Ps} is the radius of the Ps atom. Both effects are due to the repeated Coulomb interactions in the ladder diagram series of Fig. 5. Other physical arguments and estimates which justify the validity of approximation (4) can be found in Ref. [18]. There we also tested Σ_E^{Ps} calculated from Eq. (4) by applying it to low-energy e^+-H and e^+-He scattering where accurate variational or experimental results are available (see below). Here we would like to mention that if the binding energy of the outer atomic electron $|\varepsilon_n|\approx I$ is larger than $|E_{1s}|=6.8$ eV, the denominator in Eq. (4) is always negative (for $E\simeq 0$). This makes the contribution of Σ_E^{Ps} to the correlation potential negative, thus producing an additional attraction for the positron. The smallest absolute value of the denominator is $|I+E_{1s}-E|$. Therefore, for atoms with smaller ionization potentials the Ps-formation contribution is greater, since this process is less virtual for them. If $I-E<6.8$ eV the integral in Eq. (4) contains both negative and positive contributions. This means that for low-energy positrons the Ps formation may produce a repulsive contribution to the correlation potential. Besides that, the potential Σ_E^{Ps} in this case acquires an imaginary part due to the opening of a new reaction channel. This imaginary part is proportional to the squared amplitude of the ground-state Ps formation.

The total positron-atom correlation potential is calculated as the sum

$$\Sigma_E=\Sigma_E^{\mathrm{pol}}+\Sigma_E^{\mathrm{Ps}}. \tag{5}$$

It was shown in [18] that in e^+-H and e^+-He low-energy scattering the Ps-formation contribution is about 30% and 20%, respectively, of the total correlation potential. The latter provides good agreement with the results of precise variational calculations for the e^+-H scattering and with the experimental data on low-energy scattering of positrons by noble gas atoms [18,19]. The results of [19] give evidence that for the Xe atom, which has the largest polarizability among the noble gas atoms ($\alpha=27$ a.u.), the positron-atom potential may be strong enough to form an s-type bound state. The energy obtained, $\varepsilon_0=-0.005$ eV, is too small for the bound state to be considered firmly established. A 5% reduction of the correlation potential eliminates the binding, and we cannot guarantee the accuracy of the calculated correlation potential to be better than a few percent. However, if an atom had a greater dipole polarizability (and perhaps a smaller ionization potential), a stronger binding could be achieved. This would secure the result against the uncertainty in the correlation potential.

TABLE I. Parameters of the virtual and bound s-type states of positrons and neutral atoms.

Atom	Z	I [a]	α [b]	κ [c]	κ [d]	$\lvert\varepsilon_0\rvert$ [e]	z [f]
		(eV)	(a.u.)	(a.u.)	(a.u.)	(eV)	
Mg	12	7.65	68	−0.016	0.21	0.87	0.53
Cd	48	8.99	53	−0.036	0.11	0.35	0.76
Zn	30	9.39	42	−0.056	0.09	0.23	0.86
Hg	80	10.44	38	−0.080	0.05	0.045	0.95

[a] Atomic ionization potential [33].
[b] Dipole polarizabilities calculated from Σ_E^{pol}.
[c] Inverse scattering length for Σ_E^{pol}.
[d] Inverse scattering length for $\Sigma_E^{\mathrm{pol}}+\Sigma_E^{\mathrm{Ps}}$.
[e] Binding energy for $\Sigma_E^{\mathrm{pol}}+\Sigma_E^{\mathrm{Ps}}$.
[f] Normalization of the positron quasiparticle wave function.

III. RESULTS AND DISCUSSION

In the present work we consider the interaction of positrons with the atoms Mg, Zn, Cd, and Hg of the second column of the periodic table. Their polarizabilities are greater than that of Xe, and our calculations indicate that all of them can form bound states with positrons. The starting point for the calculation is the Hartree-Fock-Dirac (HFD) Hamiltonian of the neutral atom. The polarization contribution Σ_E^{pol} is obtained according to Fig. 2. The dipole polarizability values corresponding to the long-range behavior of Σ_E^{pol} are given in Table I. They are in reasonable agreement with the recommended values from [29].

The solution of Eq. (1) with $\Sigma_E=\Sigma_E^{\mathrm{pol}}$ shows that the polarization potential alone is not strong enough to produce positron-atom binding. However, it creates low-lying s-wave virtual levels. These states can be characterized by the parameter κ, which describes the low-energy behavior of the corresponding phase shift δ_0: $k\cot\delta_0=-\kappa$ (k is the positron wave number) and is equal to the inverse scattering length: $\kappa=a^{-1}$. If κ is small and negative (i.e., $-a\gg r_a$, r_a being the atomic radius) a virtual level exists at $E\simeq\hbar^2\kappa^2/2m$. The κ parameter varies linearly with respect to the potential [30], and when κ passes through zero and becomes positive, a bound state emerges at $\varepsilon_0\simeq-\hbar^2\kappa^2/2m$. The extremely small negative values of κ in Table I mean that the systems in question are on the verge of binding, and even a small increase of the correlation potential would make these states bound. We should mention that there is a noticeable discrepancy between our value of $\kappa=-0.08$ and the value of -0.025 obtained for Hg in [31], where the polarization potential was calculated by the relativistic polarized orbital method.

Then, to obtain the total correlation potential (5), the Ps-formation contribution is calculated from Eq. (4). As one would expect, only the formation of Ps by the electrons from the outer shell noticeably contributes to Σ_E^{Ps}. The relative strength of Σ_E^{Ps} is essentially determined by the smallest value of the energy denominator in Eq. (4), $|\varepsilon_n-E_{1s}|\approx I-|E_{1s}|$, where n is the outer atomic orbital. This energy difference (equal to the Ps-formation threshold) is rather small, especially for Mg, which makes Σ_E^{Ps} very sensitive to the magnitude of ε_n. To improve the accuracy of the calculation of Σ_E^{Ps} we use experimental values of $\varepsilon_n=-I$ rather than the HFD ones, for the outer orbital. In principle, this could be done within the many-body theory by including the self-energy corrections to the hole state n in Fig. 5.

The inclusion of Σ_E^{Ps} into the correlation potential produces the s-type positron-atom binding. The binding energies $|\varepsilon_0|$ are given in Table I together with the values of κ, obtained from the corresponding s-wave phase shifts.

Naturally, one should be very careful when predicting a new kind of atomic system, to be sure of one's results. The accuracy of the polarization potential has been shown to be very high for many atoms. However, the accuracy of the Ps-formation potential is not as well established. Thus it is important to consider how accurately we need to know the Ps-formation potential to be sure that the bound states actually exist. Comparing the κ values for Σ_E^{pol} and the total Σ_E, one can see that even a fraction of the present Ps-formation potential would be sufficient to form bound states (from about 60% for Hg to only 7% for Mg). Therefore, there is a great deal of certainty in our prediction of the existence of these bound states.

Calculating the eigenvalue ε_0 of Eq. (1) at arbitrary E: $(H_0+\Sigma_E)\psi=\varepsilon_0\psi$, one can find the dependence of the eigenvalue on E: $\varepsilon_0(E)$[the equation $\varepsilon_0(E)=E$ gives the correct energy of the bound state]. This enables one to calculate the corresponding residue of the Green's function (see, e.g., [13]),

$$z=\left(1-\frac{\partial\varepsilon_0(E)}{\partial E}\bigg|_{E=\varepsilon_0}\right)^{-1}<1, \qquad (6)$$

which estimates the contribution of the "positron plus atom in the ground state" component to the wave function of the positron-atom bound state. In other words, it is the degree to which the positron-atom bound state is a single-particle state. The values of z are given in the last column of Table I. They show that the positron-atom states obtained are highly correlated, since the z values are noticeably smaller than unity. In general they are smaller than those for the negative ion states [20,22], where the correlation potential is accurately approximated by Σ_E^{pol} (with the exchange diagrams added). This is explained by the fact that Σ_E^{Ps} has a much stronger energy dependence than Σ_E^{pol} (since the denominator of Σ_E^{Ps} is approximately $|E_{1s}|$ smaller than that of Σ_E^{pol}). Therefore for an atom A with the ionization potential close to 6.8 eV the positron-atom bound-state wave function is not described very well by the e^+A component and the positron-atom complex has rather a moleculelike structure. Its wave function can be qualitatively described as a linear combination of the e^+A and $\mathrm{Ps}A^+$ components.

Now, since our many-body calculations predict bound states for positrons with Mg, Zn, Cd, and Hg atoms, it would be interesting to answer the following question: Is positron binding to these atoms an exception, or rather a common phenomenon throughout the periodic table? In order to get some insight let us recall that it is the polarization potential alone that creates low-lying positron-atom virtual levels. Once this has happened a relatively small additional attraction is capable of turning these virtual levels into true bound states. Therefore the first thing to be estimated is the strength

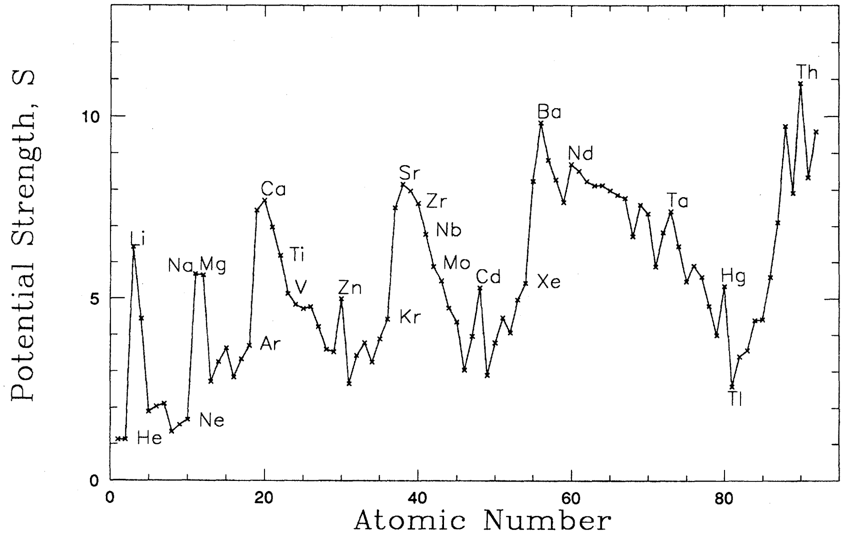

FIG. 6. Strength of the positron-atom polarization potential. Atoms with $S\gtrsim5$ (and $I\gtrsim6.8$ eV) are likely to form bound states with positrons.

of the polarization potential. This can be done for a local positron-atom potential $U(r)$ by calculating the dimensionless integral [32]

$$S=\frac{2m}{\hbar^2}\int |U(r)|r dr \tag{7}$$

over the domain where $U(r)<0$. There are two basic features of the positron-atom interaction which can be approximated within a local potential $U(r)$. First, at large distances it is proportional to the atomic dipole polarizability: $U(r)=-\alpha e^2/(2r^4)$. Second, the potential inside the atom (i.e., at $r<r_a$) is strongly repulsive. If we assume that the atomic radius is given by the classical turning point of the outer electron in the ionic Coulomb field: $r_a=e/I$, the integral (7) calculated from r_a to infinity yields the following estimate for the potential strength:

$$S=\frac{m\alpha I^2}{2\hbar^2}. \tag{8}$$

This estimate of the potential does neglect the nonadiabatic effects, as well as the quadrupole polarizability, etc. However, we compared the S values of H, He, Ar, Kr, Xe, Mg, Zn, Cd, and Hg with the results of our accurate many-body calculations (the results of the present work and of [18,19]) and we found that the trends shown by the S values agree with the trends seen in our many-body calculations.

In Fig. 6 we plotted the strength of the positron-atom polarization potential S from Eq. (8) for all atoms through to $Z=92$ (we used the polarizabilities from Ref. [29] and the ionization potentials from [33]). The results of the present paper and the absence of binding for the rare gas atoms He, Ne, Ar, and Kr [19] indicate that the strength has a critical value $S_c\simeq 5$. Atoms with $S\gtrsim S_c$ are likely to have bound states with positrons, whereas those with $S<S_c$ are not. Of course, if the ionization potential of an atom is smaller than 6.8 eV, the state is unstable against Ps emission, unless $I+|\varepsilon_0|>6.8$ eV. The latter may easily be the case for Cr, which has $I=6.77$ eV. The situation $I\simeq 6.8$ eV should, in fact, give rise to a large Ps-formation contribution, probably even stronger than that for Mg. Besides Cr, there are a number of other atoms which look quite favorable in the sense of binding positrons: Ti, V, Mn, Zr, Nb, and Mo ($S=6.2$, 5.1, 4.7, 7.6, 6.7, and 5.8, respectively). Mn is included in this list as it has an ionization potential just greater than 6.8 eV [$I(\mathrm{Mn})=7.43$ eV] and hence the Ps-formation contribution should be very large for it.

IV. CONCLUSIONS

The results of our calculations and estimates suggest that many atoms in the periodic system can form bound states with positrons. The question of binding is decided by the magnitude of the dipole polarizability and the ionization potential of the atom. Virtual Ps formation is vitally important for the binding. The wave function of the "(positron) + (atom)" bound state is strongly correlated and contains a large contribution of the "Ps + (positive ion)" component.

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