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Detecting Positron-Atom Bound States through Resonant Annihilation

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A method is proposed for detecting positron-atom bound states by observing enhanced positron annihilation due to electronic Feshbach resonances at electron-volt energies. The method is applicable to a range of open-shell transition-metal atoms which are likely to bind the positron: Fe, Co, Ni, Tc, Ru, Rh, Sn, Sh, Ta, W, Os, Ir, and Pt. Estimates of their binding energies are provided.

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Our analysis has identified about 25 open-shell atoms that are likely to form bound states with the positron. We show that for many of them binding can be detected through resonantly enhanced positron annihilation.

The existence of positron-atom bound states was predicted by many-body theory calculations [1] and proved variationally [2,3] more than a decade ago. Since then positron binding energies have been calculated for many ground-state and excited atoms: He $2^1S$, Li, Be, Be $2^3P$, Na, Mg, Ca, Cu, Zn, Sr, Ag, and Cd [4–9]. They range from $\sim 10$ meV to $\sim 0.5$ eV.

In spite of this wealth of predictions, experimental verification of positron binding to neutral atoms is still lacking. To observe positron-atom bound states and measure their energies, one needs to produce sufficient numbers of atoms in the gas phase or in a beam, but more critically, to find an efficient way of populating these bound states. Thus, radiative recombination, $A + e^- \rightarrow e^+ A + h\omega$, is inefficient because of the small cross section, $\sigma \sim (\omega/c)^3$ (in atomic units in which $m_e = \hbar = e^2 = 1$ and the speed of light $c = 137$). One suggestion applicable to atoms with positive electron affinities, was to use a charge-transfer reaction for negative ions, $A^- + e^+ \rightarrow e^+ A + e^-$, and measure either its threshold energy or the electron spectrum [10,11]. The cross section of this process should be atomic-sized, but this scheme has not been realized experimentally yet.

In contrast, much is now known about positron binding to molecules. Binding energies for over 30 polyatomic species have been determined [12,13] by measuring positron annihilation with a high-resolution, tunable, trap-based positron beam [14]. The key idea of this method is that for molecules that are capable of binding the positron, the dominant annihilation mechanism is through formation of positron-molecule vibrational Feshbach resonances [15–17]. The majority of the resonances observed are associated with individual vibrational modes of the molecule. The binding energy $E_b$ can then be found from the downshift of the resonance energy $E_{\nu} = \omega_{\nu} - E_b$ with respect to the energy $\omega_{\nu}$ of the vibrational excitation [18,19]. These experiments proved the link between positron binding and enhanced annihilation rates [17].

For atoms existing theoretical predictions of positron binding are limited to species with one or two valence $s$ electrons, as these systems are easier to compute. It is expected that many other atoms with open multielectron valence shells, can bind the positron [1,10]. Physically, positron binding is facilitated by a sizeable dipole polarizability $\alpha_d$ and moderate ionization potential $I$. While there is no rigorous criterion for binding, examination of the atoms that bind, suggests the following conditions: $\alpha_d \geq 40$ a.u. and $I < 10$ eV.

Large values of $\alpha_d$ ensure that the positron experiences a strong attractive polarization potential $-\alpha_d/2r^4$ outside the atom. Small ionization potentials increase the effect of virtual positronium (Ps) formation: a process in which an atomic electron temporarily joins the positron. It gives a distinct contribution to the positron-atom attraction akin to covalent bonding [1,20,21]. The energy of the ground-state Ps is $E_{1s} = -6.8$ eV, and this effect is strongest for $I \sim 6.8$ eV. For atoms with $I < |E_{1s}|$, positron bound states increasingly have the character of a “Ps cluster” orbiting the positive ion [22]. In this case the criteria for binding change, atoms with compact cores being favored (e.g., $e^+ Na$ is bound while $e^+ K$ is not). Atoms with $I < 6.8$ eV also differ in one other important aspect: the Ps-formation channel ($A + e^+ \rightarrow A^+ + Ps$) is open at all positron energies for them.

Figure 1 shows the polarizabilities vs ionization potentials for atoms with $6.6 < I < 10$ eV. For most of them $\alpha_d > 40$ a.u., and according to the above criterion, they are likely to form bound states with the positron. Solid symbols identify atoms for which the binding energies have been calculated: Be, $e_b = 87$ [7]; Zn, 103 [9]; Cd, 126 [23]; Ag, 123 [5]; Cu, 170 [4]; and Mg, 464 meV [9]. The weakest binding in this group is by Be and Zn found on the bottom right in Fig. 1. For a nearby atom of
The horizontal line $\alpha_d = 40$ a.u. is an approximate boundary between binding and nonbinding atoms.

Gold positron binding occurs in the nonrelativistic approximation (which underestimates the ionization potential and overestimates $\alpha_d$). However, in a fully relativistic calculation this system is not bound [5].

Figure 1 shows that most good candidates for positron binding are transition-metal atoms with open $p$ or $d$ subshells. Many of these atoms possess low-lying excited states with energies $\sim 1$ eV, due to a fine structure or Coulomb splitting of the ground-state configuration, or $ns - (n-1)d$ transitions. Their polarizabilities are similar to those of the ground states. Hence, they are also likely to bind the positron. Depending on the excitation and binding energies, this will be either a true bound state ($\varepsilon_p > \omega_p$), or a resonant contribution to the positron-atom annihilation cross section is written using the Breit-Wigner formalism [27] as

$$\sigma_a = \frac{\pi}{k^2} \sum_\nu \frac{2J_\nu + 1}{2J_\nu + 1} \frac{\Gamma_\nu^a \Gamma_\nu^r}{(\varepsilon - \varepsilon_\nu)^2 + \Gamma_\nu^2/4},$$

where $k = \sqrt{2}\varepsilon$ is the positron momentum, $J$ is the total angular momentum of the target ground state, and $J_\nu$ is that of the resonance $\nu$. To estimate the observable effect, we average the normalized dimensionless annihilation rate $Z_{\text{eff}} = \sigma_a k/(\pi\varepsilon_\nu \epsilon_0)$ ($\epsilon_0$ being the classical electron radius) over the energy distribution in the positron beam, and obtain

$$Z_{\text{eff}}(\varepsilon) = \frac{2\pi^2 \rho_{\text{pep}}}{2J + 1} \sum_\nu \frac{(2J_\nu + 1)\Gamma_\nu^r}{k_\nu \Gamma_\nu^r} \Delta(\varepsilon - \varepsilon_\nu).$$

The last column in Table I indicates the type of electromagnetic transitions between the ground and excited states allowed by selection rules. All of the excited states have the same parity as the ground state. The majority of them have nonzero total angular momenta $J$. As a result, the most common allowed transition between the levels is $E2$. Of course, the excitation of an atom by the Coulomb field of the positron in the process of capture, is different from that by a photon. However, for the transitions of electric type, a simple estimate of the elastic width in terms of the atomic transition amplitude can be derived, which shows that $\Gamma_r \gg \Gamma_\nu$ (see below).

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where $\epsilon$ is the mean longitudinal energy of the beam, $k_e = \sqrt{2\epsilon}$, and $\rho_{e\rho}$ is the electron-positron contact density in the positron bound state, which determines its annihilation width $\Gamma_{\rho} = \pi r_0^2 \rho_{e\rho}$. The function $\Delta$ describes the positron energy distribution around the mean energy $\epsilon$, $\int \Delta(E)dE = 1$ [28,29].

The contact density can be estimated from $\rho_{e\rho} = (F/2\pi)\sqrt{2\epsilon}$, where $F = 0.66$ [16,17]. To evaluate the elastic width, we use a multipole expansion of the positron Coulomb interaction with the atom. Using the fact that for a low-energy positron, large positron-atom separations are achieved be heating the samples to temperatures ranging 1500 K, and over 2000 K for Fe, Co, and Ni, and over 2000 K for other species [30].

Detection of the resonances can thus provide the first experimental evidence of positron binding to neutral species. The above analysis indicates that positron-atom resonances could be observed with a trap-based beam used for studying resonances in positron-molecule annihilation [19]. Such a measurement requires vapour pressure of $\sim 0.01$ mtorr [18]. For the atoms in Table I this can be achieved by heating the samples to temperatures ranging from 650 °C for Sb and 1100 °C for Sn, to 1500 °C for Fe, Co, and Ni, and over 2000 °C for other species [30].

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**Table I.** Atoms with low-lying excited states in which positron binding and annihilation resonances are expected.

<table>
<thead>
<tr>
<th>Z</th>
<th>Atom</th>
<th>Ground state $I$ (eV)</th>
<th>$\alpha_d$ (a.u.)$^a$</th>
<th>$\epsilon_p$ (eV)$^b$</th>
<th>Excited state(s) $\epsilon_p$ (eV)$^c$</th>
<th>$\omega_{\rho}$ (eV)$^d$</th>
<th>Transition type $^e$</th>
</tr>
</thead>
<tbody>
<tr>
<td>26</td>
<td>Fe</td>
<td>$3d^44s^23p^2D_4$</td>
<td>7.902</td>
<td>56.7</td>
<td>0.28</td>
<td>3d$^4$4s$^2$F_{3/2}</td>
<td>0.09</td>
</tr>
<tr>
<td>27</td>
<td>Co</td>
<td>$3d^44s^23p^2F_{9/2}$</td>
<td>7.881</td>
<td>50.7</td>
<td>0.28</td>
<td>3d$^4$4s$^2$F_{3/2}</td>
<td>0.08</td>
</tr>
<tr>
<td>28</td>
<td>Ni</td>
<td>$3d^44s^23p^2F_{5/2}$</td>
<td>7.640</td>
<td>45.9</td>
<td>0.24</td>
<td>3d$^4$4s$^2$F_{2}</td>
<td>0.24</td>
</tr>
<tr>
<td>43</td>
<td>Tc</td>
<td>$4d^55s^2F_{5/2}$</td>
<td>7.280</td>
<td>77.0</td>
<td>0.46</td>
<td>4d$^5$5s$^2$F_{5/2}</td>
<td>0.23</td>
</tr>
<tr>
<td>44</td>
<td>Ru</td>
<td>$4d^55s^2F_{5/2}$</td>
<td>7.361</td>
<td>64.9</td>
<td>0.21</td>
<td>4d$^5$5s$^2$F_{5/2}</td>
<td>0.21</td>
</tr>
<tr>
<td>45</td>
<td>Rh</td>
<td>$4d^55s^2F_{5/2}$</td>
<td>7.459</td>
<td>58.1</td>
<td>0.20</td>
<td>4d$^5$5s$^2$F_{5/2}</td>
<td>0.20</td>
</tr>
<tr>
<td>50</td>
<td>Sn</td>
<td>$5s^25p^3P_0$</td>
<td>7.344</td>
<td>52.0</td>
<td>0.02</td>
<td>5s$^2$5p$^3$P_{3/2}</td>
<td>0.02</td>
</tr>
<tr>
<td>05l</td>
<td>Sb</td>
<td>$5s^25p^3P_{3/2}$</td>
<td>8.608</td>
<td>44.6</td>
<td>0.05</td>
<td>5s$^2$5p$^3$P_{3/2}</td>
<td>0.05</td>
</tr>
<tr>
<td>73</td>
<td>Ta</td>
<td>$5d^65s^2F_{5/2}$</td>
<td>7.550</td>
<td>88.5</td>
<td>0.45</td>
<td>5d$^6$5s$^2$F_{5/2}</td>
<td>0.45</td>
</tr>
<tr>
<td>74</td>
<td>W</td>
<td>$5d^65s^2D_{0}$</td>
<td>7.864</td>
<td>75.0</td>
<td>0.46</td>
<td>5d$^6$5s$^2$D_{0}</td>
<td>0.46</td>
</tr>
<tr>
<td>76</td>
<td>Os</td>
<td>$5d^65s^2D_{0}$</td>
<td>8.438</td>
<td>57.4</td>
<td>0.47</td>
<td>5d$^6$5s$^2$D_{0}</td>
<td>0.47</td>
</tr>
<tr>
<td>77</td>
<td>Ir</td>
<td>$5d^65s^2D_{0}$</td>
<td>8.967</td>
<td>51.3</td>
<td>0.46</td>
<td>5d$^6$5s$^2$D_{0}</td>
<td>0.47</td>
</tr>
<tr>
<td>78</td>
<td>Pt</td>
<td>$5d^65s^2D_{0}$</td>
<td>8.960</td>
<td>43.9</td>
<td>0.27</td>
<td>5d$^6$5s$^2$D_{0}</td>
<td>0.27</td>
</tr>
</tbody>
</table>

$^a$Dipole polarizabilities from Ref. [25].
$^b$Binding energies $\epsilon_p = |e|\mid$ for atoms in ground-state configurations obtained using $\Sigma = \chi \Sigma^{(2)}$ with $\chi = 2$ (see text).
$^c$Binding energies for atoms in excited-state configurations obtained with $\Sigma = \chi \Sigma^{(2)}$, $\chi = 2$.
$^d$Energies of excited states from Ref. [26], such that $0.2$ eV $\leq \omega_{\rho} \leq E_{\rho} + 0.15$ eV.
$^e$When several transitions are allowed, the most probable is indicated.

assuming that the quadrupole amplitude is atomic-sized, one obtains $\Gamma_{\rho} \sim 1–10$ meV. Hence, these resonances are sufficiently narrow to produce observable sharp features in the energy dependence of $Z_{\text{eff}}$. Estimating the annihilation width for $\epsilon_{\rho} = 150$ meV, we obtain $\Gamma_{\rho} = 4 \times 10^{-7}$ eV, hence, $\Gamma_{\rho} \gg \Gamma_{\rho}$. In this case, $\Gamma_{\rho} / \Gamma_{\rho} = 1$, and the contribution of such resonance to $Z_{\text{eff}}$, Eq. (3), is close to maximum possible. For a positron beam with energy spread $\delta \epsilon \sim 25$ meV, using $\Delta_{\text{max}} / \sqrt{\Delta_{\text{max}}} / \delta \epsilon$, the peak resonant value of the annihilation rate from Eq. (3) is given by $Z_{\text{eff}} \sim \pi F \sqrt{\epsilon_{\rho} / \epsilon_{\rho} / \delta \epsilon / \delta \epsilon \sim 10^3}$. This estimate remains valid even if the elastic width is suppressed by up to 3 orders of magnitude, e.g., for a higher-multipole transition, or a transition mediated by the relativistic (spin-orbit) interaction.
atoms, and first estimates of the binding energies. While a Feshbach resonance only signifies binding to an excited state, the binding energy in the ground state is expected to be similar if it has the same electronic configuration.

Resonant enhancement can also be observed with thermalized positrons. Depending on the exact position of the resonances, it can lead to a nontrivial dependence of the annihilation rate on the positron temperature, with greater rates measured at higher temperatures. Such behavior would be in sharp contrast with that observed in nonresonant systems, such as the noble gases [31].

One should mention that earlier experimental searches for positron resonances in the vicinity of electronic excitation thresholds (for H₂, N₂, CO and Ar) yielded negative results [32]. However, these systems are quite different from the open-shell metal atoms considered here. None of them is expected to bind the positron in the ground state, and the electronic excitations lie above the Ps-formation threshold. In addition, the relative role of resonances in the annihilation is much more prominent than in the elastic or total scattering measured in Ref. [32].

The case of transition-metal atoms is also markedly different from that of Be, in which positron binding to the excited 2s2p³P state was predicted in configuration-interaction calculations [8]. This excited state lies above the Ps-formation threshold, but a large positron binding energy of 250 meV ensures that the bound state is 40 meV below the Be²⁺ + Ps threshold. Such strong binding by the excited state is promoted by its large dipole polarizability. For comparison, the positron binding energy by the ground-state Be atom is 87 meV [7].

Besides the Feshbach resonances, positron annihilation can be increased by shape resonances. These resonances are supported by the strong polarization attraction and the centrifugal barrier. Thus, calculations predict a sharp p-wave resonance in positron scattering from Mg at 95 meV, with $Z_{\text{eff}} = 1300$ at the peak, and similar but broader resonances at ~0.45–0.65 eV in Cu, Zn, and Cd, with $Z_{\text{eff}} \sim 100$ [9,23]. Compared with the Feshbach resonances, the shape resonances do not indicate positron binding. They also have much larger widths, e.g., ~0.1 eV in Mg.

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