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Effect of the atomic electric quadrupole moment on positron binding

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The effect of an electric quadrupole moment Q is studied for positron-atom bound systems. It is demonstrated that for $Q > 50$ a.u. the electric quadrupole potential is sufficiently strong to bind a positron (or an electron) even in the absence of the dipole polarization potential. Such large values of Q are not known for atomic ground states; however, they exist in molecules and excited atoms. In the $2s2p^3P_2^o$ state of beryllium, the quadrupole contribution makes the difference between a stable bound state and an unstable state, which decays to a Be$^+$ ion and positronium. In the majority of atoms the quadrupole contribution is small and can be neglected.

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I. INTRODUCTION

The unperturbed central potential of an atom is positive at all distances and cannot bind a positron. Only when correlations between the positron and atomic electrons are taken into account can a positron be bound to a majority of atoms [1] (see also [2–10] and references therein). A simple explanation of the positron binding is provided by the polarization of the atom by the positron field. Electron-positron attraction shifts the electron cloud towards the positron, producing an electric dipole moment of the atom. This induced electric dipole moment creates a polarization potential, which at large distances behaves like $-\alpha/2r^4$, where α is the static dipole polarizability of the atom. However, many atoms also have a static quadrupole moment which produces a long-range potential which decays slower ($\sim 1/r^3$) than the polarization potential ($\sim 1/r^4$). Therefore, it looks interesting to investigate the role of the quadrupole potential in the positron binding.

There are many different techniques used to calculate the positron binding energy of atoms. The variational and configuration interaction calculations usually include the quadrupole contribution. In some other calculations, e.g., those based on the correlation potential [8–10] and coupled-cluster single-double [1,3] approaches, the positron is assumed to be in the s wave, and therefore the quadrupole contribution is not included. Note that assuming the positron to be in the s state does not mean that the contributions of higher angular momenta are totally neglected. For example, in the case of the correlation potential method [8–10] binding energy is related to the expectation value of the correlation potential $\hat{\Sigma}$: $\epsilon \sim \langle s|\hat{\Sigma}|s\rangle$, where s is the positron wave function and higher values of both electron and positron angular momenta are included in the calculation of $\hat{\Sigma}$. Therefore, the effect of virtual positron formation is taken into account (see, e.g., [8,10,11]).

In spite of the fact that many calculations do include the quadrupole contribution, it has never been presented separately. However, it is useful to know it to judge whether it could be at least partly responsible for the difference in the results in different calculations. It is also important to know whether that contribution could be large enough to provide the difference between binding and not binding. In this Brief Report we study the quadrupole contribution using a simple variational approach.

II. VARIATIONAL ENERGY OF A POSITRON IN AN ATOM

The positron trial wave function in the presence of electric dipole and quadrupole potentials takes the form

$$\Psi(r,\theta) = A(r-a)e^{-\kappa r}\left(\sqrt{1-\beta^2}Y_{00} + \beta Y_{20}\right), \tag{1}$$

where A is the normalization constant, $Y_{lm}(\theta,\phi)$ are spherical harmonics, κ and β are variational parameters, and a is a cutoff distance from the nucleus. It is assumed that the wave function is zero at $r < a$. The potential energy of the positron for $r > a$ is

$$V(r,\theta) = -\frac{e^2\alpha}{2r^4} + \frac{eQ}{2}\frac{P_2(\cos\theta)}{r^3}, \tag{2}$$

where α is the polarizability of the atom, Q is its quadrupole moment, and $P_2(\cos\theta) = (3\cos^2\theta - 1)/2$ is the Legendre polynomial. There is also an additional infinite potential barrier at $r = a$ which simulates the effect of positron repulsion from the positive electrostatic potential inside the atom. The mixing of s and d waves provides localization of the positron wave function in equatorial or polar areas depending on the sign of Q.

Using Schrödinger's equation the positron energy can be written as

$$E(\kappa,\beta) = \frac{\int d^3r\left(\frac{\hbar^2}{2m}\nabla\Psi^*\cdot\nabla\Psi + \Psi^* V\Psi\right)}{\int d^3r\,\Psi^*\Psi}. \tag{3}$$

Substituting (1) into (3) leads to the explicit form of $E(\kappa,\beta)$:

$$\frac{4ma^2}{\hbar^2}E(\kappa,\beta) = \mathscr{E}(x,\beta) = \frac{x^2 f(x,\beta)}{x^2+6x+12}, \tag{4}$$

where $x = 2a\kappa$ is a dimensionless variational parameter and $\mathscr{E}(x,\beta)$ is the dimensionless variational energy. The function $f(x,\beta)$ can be written in the form

$$\begin{aligned} f(x,\beta) = {} & -\Omega x^3 + (1/2 - \Omega - \Lambda\xi)x^2 \\ & + (\Lambda\xi + 1)x + 12\beta^2 + 2 \\ & + [\Omega x^3 + (2\Omega + \Lambda\xi)x^2]xe^x E_1(x), \end{aligned} \tag{5}$$

where

$$\Omega = \frac{me^2\alpha}{\hbar^2 a^2}, \tag{6}$$

$$\Lambda = 2\frac{meQ}{\hbar^2 a}, \tag{7}$$

$$\xi = \beta\sqrt{\frac{1-\beta^2}{5} + \frac{\beta^2}{3}}, \tag{8}$$

and

$$E_1(x) = \int_x^\infty dt\frac{e^{-t}}{t} \tag{9}$$

is the exponential integral [12].

Varying the energy (3) with respect to parameters x and β requires that

$$\frac{\partial\mathscr{E}(x,\beta)}{\partial x} = 0, \quad \frac{\partial\mathscr{E}(x,\beta)}{\partial\beta} = 0.$$

Solving these equations for x and β would lead to the ground-state energy $\mathscr{E}(x_0,\beta_0)$ of the positron in an atom specified by three parameters, the cutoff parameter a, the polarizability α, and the quadrupole moment Q.

The partial derivative of the energy (4) with respect to β leads to

$$\frac{\partial\mathscr{E}}{\partial\beta} = \Lambda\frac{\partial\xi}{\partial\beta}\{[xe^x E_1(x) - 1]x^2 + x\} + 24\beta = 0. \tag{10}$$

This equation can be used to express β in terms of the other parameter x,

$$\beta = \pm\sqrt{[1 \pm \lambda(x)]/2}, \tag{11}$$

where $\lambda(x)$ is a simple function of x:

$$\lambda(x) = \sqrt{1 - \frac{1}{1 + 5\left(\frac{\hbar^2}{me}\frac{a}{Q}\frac{6}{g(x)} + \frac{1}{3}\right)^2}}. \tag{12}$$

The function $g(x) = [xe^x E_1(x) - 1]x^2 + x$ is the same as the x-dependent part of Eq. (10). It is positive for all $x > 0$, and $g(x) \sim 2$ asymptotically at $x \to \infty$.

We substitute expression (11) for β into a second variational equation $\frac{\partial\mathscr{E}}{\partial x}(x,\beta(x)) = 0$. For each root x_0 of this equation there are four different values of β found from Eq. (11).

The final equation to solve for the extrema of the energy, which depend only on x after substituting $\beta(x)$, has the form

$$\begin{aligned}&\frac{-(\Omega+1/2)x^4 + (1-4\Omega-5\Lambda\xi)x^3 + (1-12\beta^2+3\Lambda\xi)x^2 + 4(1+3\beta^2)x + [6\Omega x^4 + 5(2\Omega+\Lambda\xi)x^3]xe^x E_1(x)}{-\Omega x^5 + (1/2-\Omega-\Lambda\xi)x^4 + (1+\Lambda\xi)x^3 + 2(1+6\beta^2)x^2 + [\Omega x^5 + (2\Omega+\Lambda\xi)x^4]xe^x E_1(x)}\\ &= -\frac{x^2+4x+6}{x^2+6x+12}.\end{aligned} \tag{13}$$

This equation is to be solved numerically for the roots $x = x_0$ if they exist.

III. RESULTS AND DISCUSSION

To use Eq. (13) to calculate the positron binding energy we need to know the atomic polarizability α, the atomic quadrupole moment Q, and the value of the cutoff parameter a. For polarizabilities and quadrupole moments we use values which can be found in the literature (either experimental or theoretical), and we treat the cutoff parameter a as a fitting parameter. We choose its value to fit the most accurate calculations of the positron binding energy. The value of the quadrupole contribution is found as the difference between the binding energy at a given value of the quadrupole moment and the value found at $Q = 0$.

In cases when the atomic polarizability or quadrupole moment cannot be found in the literature we calculate them using the configuration interaction (CI) technique [19–22]. The static scalar polarizability of an atom in state γ is given by

$$\alpha_\gamma = \frac{2}{3(2J_\gamma+1)}\sum_n \frac{\langle\gamma||\mathbf{D}||n\rangle^2}{E_\gamma - E_n}, \tag{14}$$

while the electric quadrupole moment is given by

$$Q_\gamma = -2\sqrt{\frac{J_\gamma(2J_\gamma-1)}{(2J_\gamma+3)(2J_\gamma+1)(J_\gamma+1)}}\langle\gamma\|er^2\|\gamma\rangle. \tag{15}$$

Here $|\gamma\rangle$ and $|n\rangle$ are many-electron states found in the CI calculations.

About half of all atoms do not have quadrupole moments in the ground state due to the small value of the total angular momentum J (one needs $J \geqslant 1$). Atoms with open d or f shells have large angular momenta. In Table I we present three such examples, Dy, Er, and Ir atoms. Polarizabilities α are taken from Ref. [14], quadrupole moments Q are calculated using the CI technique, and the cutoff parameter a is chosen to fit calculated binding energies presented in Ref. [1]. Since the calculations in [1] are done under the assumption that the positron is in the s wave and therefore cannot interact with the atomic quadrupole moment, the fitting is done for $Q = 0$. After the cutoff parameter a is found, Eq. (13) with values of Q from Table I is used to calculate new bound energies. The resulting energy shift ΔE is the quadrupole contribution to the binding energy. We see that it is small in all three cases. This justifies neglecting the quadrupole contribution in the calculations of Ref. [1].

The small value of the quadrupole contribution means that it can be treated by means of the perturbation theory. The first-order contribution for a positron in the s wave is zero; therefore, expansion starts from second order, and the quadrupole contribution is proportional to the square of the quadrupole moment. Using the Ir atom as a reference point, we can estimate the quadrupole contribution to the positron energy level for any atom with a small quadrupole

TABLE I. Energy shift ΔE of a bound positron in an atom due to the electric quadrupole moment (EQM) Q of the atom. I is the ionization energy, and α is the static dipole polarizability of the atomic state. The parameter a included in the positron trial function shows the minimal distance between the positron and the nucleus. E is the binding energy of the positron to the atom without the quadrupole contribution, except in the case of the Be atom, where the quadrupole effect has already been included in the binding energy [13].

Z	Atom	State	I[c] (eV)	α[a] (a.u.)	a (a.u.)	Q (a.u.)	E[b] (meV)	ΔE (meV)
				Ground states				
66	Dy	$4f^{10}6s^2\ {}^5I_8$	5.939	162.02	2.5878	0.0234	–1438	-5.36×10^{-4}
68	Er	$4f^{12}6s^2\ {}^3H_6$	6.107	150.12	2.530	0.0139	–1346	-1.98×10^{-4}
77	Ir	$5d^76s^2\ {}^4F_{9/2}$	8.967	50.26	1.8606	0.75	–101	−0.6549
				Excited states				
4	Be	$2s2p\ {}^3P_2^o$	6.597	38.33	1.60774	4.28[f],4.53[g]	–236[h]	−41.98
13	Al	$3s^23p\ {}^2P_{3/2}^o$	5.972	44.97	1.81304	5.6, 5.06[d]	0	−28.67
49	In	$5s^25p\ {}^2P_{3/2}^o$	5.512	67.45	2.13042	5.88[e]	–114	−23.15

[a] Ground-state atomic static dipole polarizabilities from Ref. [14].

[b] Recommended positron energies in Ref. [1].

[c] Ionization potential from the NIST atomic database [15].

[d] Experimental value from Ref. [16].

[e] Experimental value from Ref. [17].

[f] EQM of a Be atom from Ref. [18].

[g] EQM of a Be atom from Ref. [17].

[h] Positron binding energy to the excited state of Be from Ref. [13].

moment:

$$\Delta E = -0.6549\left(\frac{Q}{0.75}\right)^2 \text{ meV}, \tag{16}$$

where Q is in atomic units (ea_0^2). The values given by this formula differ from those presented in Table I by 13% for Er and 19% for Dy. We expect similar accuracy for other atoms with small quadrupole moments.

In Table I we also present three other results for the quadrupole contribution to the positron binding energy. We consider the excited state of the Be atom, for which accurate calculations of the positron binding energy is available [13], and we consider the upper components of the fine-structure doublets of the ground state of Al and In. In all these cases the value of the quadrupole moment is relatively large. So is the quadrupole shift. The case of the Be atom is interesting because the quadrupole contribution makes an important difference for the positron binding. The calculated binding energy (−236 meV) does include the quadrupole contribution. However, if this contribution is neglected, the system becomes unstable against the emission of positronium (Be + $e^+ \rightarrow$ Be$^+$ + Ps).

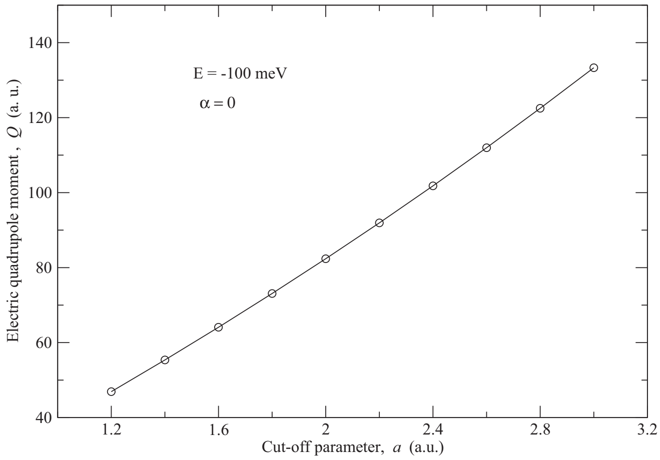

FIG. 1. The distance of the boundary of the wave function of the positron from the nucleus a (in a.u.) vs the required minimum electric quadrupole moment Q to get a bound state with the energy $E = -100$ meV when the dipole polarizability is supposed to be zero, $\alpha = 0$.

In some excited atomic states the polarizability may be very small or even negative [23]. Therefore, it is interesting to check if the quadrupole alone (for $\alpha = 0$) could provide the positron binding. We use Eq. (13) to estimate what value of the quadrupole moment is needed to provide the positron binding. Figure 1 shows a plot of the quadrupole moment corresponding to the 100-meV binding energy as a function of the cutoff parameter a. We use the estimation $a \sim e^2/I$ to find a reasonable range of values for a. Here I is the ionization potential. In this estimation we assume that the boundary of the atom beyond which the positron cannot penetrate is defined as a classical turning point for the outermost electron where its kinetic energy is zero and the electron energy E is equal to the potential energy $U = -e^2/a$, i.e., $I = |E| = |U| = e^2/a$.

We see that required values of the quadrupole moments are large. No atom in the ground state has so large a quadrupole moment. However, the quadrupole moment can be large in the excited state, $Q \sim \nu^4$, where ν is the effective principle quantum number [$E = -1/(2\nu^2)$]. Large values of Q proportional to their squared size may also exist in molecules.

IV. CONCLUSION

A simple quantum-mechanical variational method was used to estimate the contribution of the positron interaction with atomic quadrupole moment to the positron energy in an atom.

It was found that the contribution is small for the binding in the ground state. This validates the calculations in which this contribution is neglected. The quadrupole contribution can be significant in excited states. For example, in the excited state of beryllium it makes a difference between a bound state and the decay into a beryllium positive ion and positronium.

The quadrupole contribution probably plays an important role in positron binding to molecules which have large quadrupole moments.

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