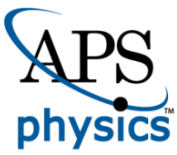

Efficient Production of Rydberg Positronium

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Phys. Rev. Lett. **108**, 043401 — Published 26 January 2012

DOI: 10.1103/PhysRevLett.108.043401

Efficient production of Rydberg positronium

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We demonstrate experimentally the production of Rydberg positronium (Ps) atoms in a two-step process, comprising incoherent laser excitation, first to the 2^3P state, and then to states with principal quantum numbers ranging from 10 to 25. We find that excitation of 2^3P atoms to Rydberg levels occurs very efficiently (~ 90%), and that the ~ 25% overall efficiency of the production of Rydberg atoms is determined almost entirely by the spectral overlap of the primary excitation laser and the Doppler broadened width of the 1^3S-2^3P transition. One would expect that the probability of producing Rydberg atoms in a stochastically populated 3-level system would be at most 33%; the observed 90% efficiency can be explained if stimulated emission back to the 2P states is suppressed, for example by intermixing of the Rydberg state Stark sublevels. The efficient production of long-lived Rydberg Ps in a high magnetic field may make it possible to perform direct measurements of the gravitational free-fall of Ps.

Although slow positron beams have been available for half a century [1], their intensities are still generally limited to ~1 pA of moderated monoenergetic positrons from small laboratory radioactive sources (e.g. 50 mCi ^{22}Na) [2]. As a result of the relatively low continuous beam positron and laser intensities, positronium-laser studies require the use of pulsed positron beams [3, 4, 5]. Recent developments in positron trapping techniques provide a way to generate intense positron pulses using a small laboratory positron beam [6]. These may be used to create high instantaneous densities of positronium (Ps) atoms [7] that are well suited to interrogation with pulsed lasers [8].

In this letter we describe experiments in which two different dye lasers are used to excite bursts of ground state Ps atoms into the 2^3P state, and then into higher lying (Rydberg) states, with principal quantum numbers ranging from 10-25. Various properties of Ps atoms in high n states make them interesting to study or to use in other experiments. For example, a precision comparison of Ps energy level separations [3] can directly address pure bound state QED physics without proton-size [9] related complications [10]. Highly excited Ps may also be useful for producing antihydrogen atoms [11], loading a stellerator to create electron-positron plasmas [12], the formation of positron-atom bound states [13], or for direct Ps gravity measurements [14].

In the present work Ps was generated using a positron accumulator [15] that delivers 2×10^7 particles in pulses that are time-bunched to widths of around 1 ns, full width at half maximum (fwhm). A porous silica film [16] was used as a target to convert incident positrons into Ps atoms in vacuum. We use single shot lifetime spectra [17] to observe laser induced changes in the Ps decay rate, and hence to perform laser spectroscopy of Ps. These spectra are analyzed to obtain the Ps "delayed fraction" f_d which is the fraction of the spectrum in the time interval 50-300 ns [18].

The lasers used in this work are both Nd:Yag pumped dye systems, very similar to those described in ref [19]. These provided up to ~ 1 mJ/pulse of ultra-violet (UV) and ~ 3 mJ/pulse of infra-red (IR) light. Both lasers were tunable, but for the present experiments the UV laser was fixed at the Ps Lyman alpha wavelength (243 nm), while the IR wavelength was varied from 733 to 764 nm. The pulse width of both lasers was ~ 5 ns.

Our experiments involve a two-step process, starting with excitation of the 1^3S-2^3P transition. 2P states have negligible annihilation rates, and decay radiatively back to the ground state with a mean lifetime of 3.2 ns. Such excitations may be observed via magnetic quenching [8]; in a relatively weak magnetic field some 2P states are composed of superpositions of singlet and triplet states, such that a 2^3P state may be Zeeman mixed [20] with a 2^1P state, and then radiatively decay to the 1^1S state, which has a mean lifetime against annihilation of only 0.125 ns. This effect, which can be used to detect the production of excited Ps states [4], does not occur for zero field, and is suppressed in strong magnetic fields > 2 T [21]. Alternatively, the 2P states may be photoionized by a second laser pulse at 532 nm. Both of these mechanisms will cause changes in the Ps decay rate that may be observed via single-shot lifetime spectra. For magnetic quenching, the increased annihilation rate is a direct result of triplet atoms essentially being converted to singlet states, whereas ionizing simply liberates a positron that then has a high probability of returning to the target and annihilating. Because incoherent transitions rapidly cycle the atoms through many Rabi oscillations, detection via magnetic quenching is quite efficient; in effect there are many chances to quench, so that even though not all 2P triplet states mix with singlet states, virtually all excited atoms can be detected in this way [21].

The Rabi frequency describing the dynamics of 1S-2P excitation is $\Omega_R = \mu E_0 / \hbar$, where E_0 is the laser electric field amplitude and the dipole moment for Ps 1S-2P transitions is $\mu \equiv \langle 2P|z|1S\rangle = 4\sqrt{2}\left(\frac{2}{3}\right)^5 e a_{Ps} = 1.26\times10^{-29}$ Cm. Here $a_{Ps} = 2a_0$ is the "Ps Bohr radius". In our experiments the average laser intensity during the 5 ns pulse is ~ 2×10^{10} W/m^2. However, in the present case this power is distributed over a large

bandwidth (~ 100 GHz), and the corresponding average Rabi period is of the order of 0.5 ns. Thus, during our ~ 5 ns laser pulses we may have something like 10 Rabi oscillations (roughly), which provides multiple opportunities for quenching, and hence makes it an efficient process.

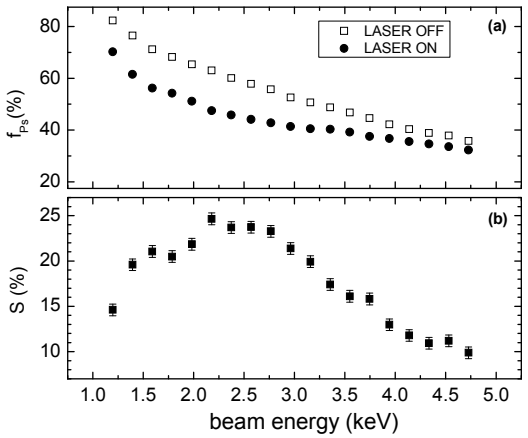

Fig 1. Positronium yield f_{Ps} leading to delayed annihilations with and without the UV laser (a) and excitation yield S (b) as a function of the positron beam energy. The magnetic field was 0.16 T. In (a) the error bars are smaller than the symbols.

Figure 1 shows the Ps yield as a function of the incident positron beam energy, with and without firing the UV laser. The laser excitation of Ps atoms is identified by the resulting destruction of ground state triplet atoms, quantified by the parameter $S \equiv \left[f_{Ps}(off) - f_{Ps}(on)\right] / f_{Ps}(off)$, where $f_{Ps}(off/on)$ refers to the Ps fraction measured with the UV laser off (on). The temperature of Ps emitted from this type of silica film depends on the implantation energy because Ps, which is initially created with about 1 eV of kinetic energy, will cool by collisions with the internal pore surfaces. In this sample the Ps energy will be constant for beam energies above 2.5 keV, and is determined by the confinement energy in the porous structure [8, 16]. The Ps emission time will also be longer for higher beam implantation energies [19]. The energy dependence of the Ps excitation yield, *S*, in Fig. 1 may then be understood in terms of two main effects; as the beam energy is increased to ~ 2.5 keV the Doppler width of the transition decreases, the spectral overlap with the laser increases and the process becomes more efficient. However, at higher beam energies the Ps energy stops changing but the emission time increases, and so the temporal overlap with the laser decreases, with a concomitant decrease in *S*.

The maximum yield observed, $S \sim 25\%$, is quite remarkable considering that the laser bandwidth is around 85 GHz fwhm, whereas the Doppler broadened linewidth, shown in Fig 2a, is almost 1 THz fwhm ($\Delta\lambda = 0.195 \pm 0.005$ nm fwhm). The spectral coverage of the laser is therefore nominally less than 10% of the width of the transition. We attribute this unexpectedly high excitation efficiency to (1) the effectiveness of the contributions in the wings of the laser frequency spectrum and (2) to power broadening, which effectively fills in the ~ 0.5 GHz gaps between the individual laser gain modes typical of an unseeded pulsed dye laser [22]. Our data also demonstrate that, even though the excitations are incoherent, the effective removal of 2P states via quenching, photoionization or further excitation means that the overall fraction of transitions can be close to 100% , rather than the 50% one would obtain for a simple two-level system.

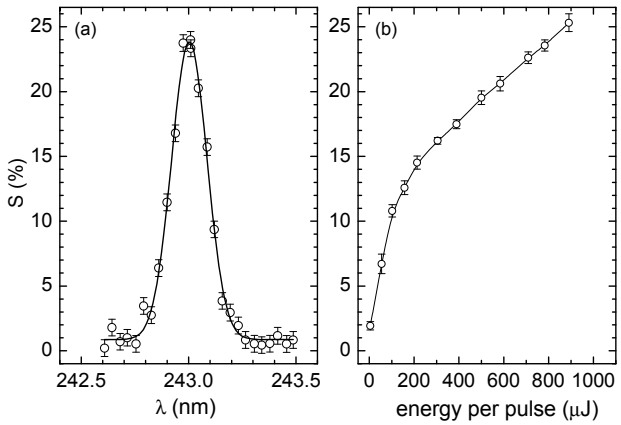

Fig 2. Doppler broadened 1^3S-2^3P linewidth (a) and power dependence of the excitation efficiency at λ = 243.0 nm (b) for a positron beam energy of 2.5 keV. The solid line in (a) is a Gaussian fit, and in (b) is a simple spline fit. The laser spot size was ~ 2.5 mm fwhm.

The excitation yield is shown as a function of the laser power in fig 2b. Based on the 1^3S-2^3P cross section, and the 50 MHz natural linewidth of this transition, we would expect the excitation to saturate for laser powers of the order of a few hundred μJ. However, after an initial steep rise in S there is a slower linear increase with power densities that are an order of magnitude above the saturation level, which supports the idea that increasing laser fluence fills in the spectral gaps in the laser bandwidth via power broadening, while enlarging the effective spectral overlap with the Ps Doppler distribution.

Having produced 2^3P Ps atoms, we then use a second pulsed IR dye laser to excite them to higher *n* states. Figure 3 shows single-shot lifetime spectra with various laser configurations, taken at magnetic fields of 0.16 T and 1 T. These data show directly the change in the Ps fraction due to the UV laser (in the 1T case there is an ionizing 532 nm pulse coincident with the UV pulse, since magnetic quenching is inhibited by the high field [21]). When the IR laser (λ = 749.6 nm,) is applied, however, n = 12 Ps atoms (lifetime ~ 1μs) are produced which *do not annihilate* until they hit the chamber wall. The resulting increase in the Ps decay rate at around (150 ± 100) ns is seen in both of the UV + IR curves of fig. 3. The Ps is produced near the middle of a small vacuum chamber, and must travel 1-2 cm to the wall. The Doppler width shown in Fig 2a corresponds to a Ps energy of ~ 60 meV, and velocities ~ 10^7 cm/s [8] which is consistent with the transit time to the wall.

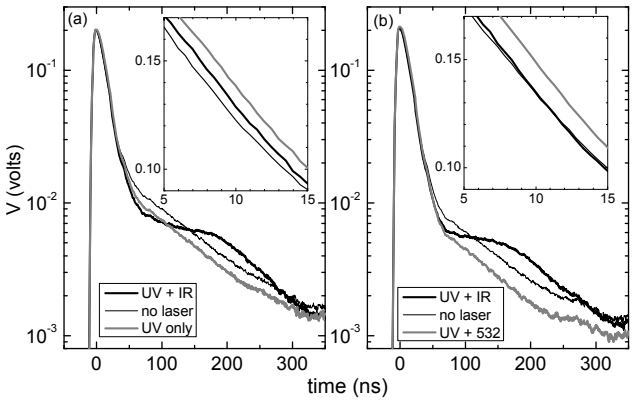

Fig 3. Single shot lifetime spectra measured at 0.16 T (a) and 1 T (b) for different laser configurations as indicated in the legend. The IR wavelength was 749.6 nm, corresponding to n = 12 Ps.

The inset of fig. 3a shows that even with the IR pulse there are still some early decays associated with the UV light, which result from magnetic quenching effect of 2P Ps, as described above [21]. Conversely, when the magnetic field is increased to 1 T there will be fewer losses of 2P states via quenching, (see inset to fig 3b), and hence a more efficient production of Rydberg Ps atoms. Moreover, when the magnetic field is increased the amount of long-lived Ps decreases because ground state m = 0 singlets and triplets become mixed [23]. However, since the m = 0 triplet lifetime at 1 T is ~ 7 ns, many of these atoms may still be excited to higher lying states, which means that both the ground state and 2P quenching processes are avoided. For higher fields the m = 0 lifetime is further reduced, and so too is the extent to which losses via ground state mixing are mitigated. The optimum magnetic field for the production of Rydberg Ps will depend on the intensities of the IR beam (which reduces 2P quenching losses) and the UV beam (which reduces 1S quenching losses); here it is ~ 1T.

Figure 4 shows lineshapes obtained across a large range of IR wavelengths for B = 0.16 T, demonstrating the production of Ps atoms with n ranging from 10 to ~ 25. The data is given in terms of the parameter f_{Ryd}, which is defined as $f_{Ryd} = (f_d[UV+IR] - f_d[UV])/(f_d[off] - f_d[UV])$. Here f_d[UV+IR] refers to the delayed fraction measured with the UV and IR laser beams, etc. Thus, f_{Ryd} = 1 corresponds to the case where the change in the long lived Ps yield caused by the UV is completely counteracted by the application of the IR beam. This can only occur when all of the 2^3P states that would otherwise have been quenched are excited to long-lived Rydberg states. The value of f_{Ryd} will be overestimated by ~ 20% due to the absence of annihilation decays of the Rydberg Ps atoms in the first 50 ns time window, and underestimated by a similar amount because of the reduced detection efficiency of high n Ps colliding with the chamber walls and the imperfect spatial overlap of the UV and IR laser pulses.

The near unity values of f_{Ryd} in Fig. 4 then imply that, despite some losses to quenching, the production of Rydberg Ps from 2^3P states is about 90% efficient. This is rather surprising since, as is shown explicitly in numerical simulations by Castelli *et al*. [11], the overall efficiency for a stochastically populated 3-level system should be ~ 33% (neglecting radiative loss from the 2^3P level). One might have expected this experiment, with simultaneous UV and IR laser pulses, to be a good approximation to a simple 3-level system, and hence to be populated stochastically. However, the high efficiency with which Rydberg atoms are produced demonstrates that this is not in fact the case. The otherwise degenerate high n Rydberg levels will be split into Stark sublevels by motional and static electric fields; we suggest that intermixing of these Rydberg sub-states, which may occur via some higher order process (e.g., ac or quadratic Stark effects) for example, inhibits stimulated emission and that the IR light is unable to drive excited states back to the 2P level. This mechanism is qualitatively similar to the 1^3S-2^3P excitation, which is almost 100% efficient (within the power broadened bandwidth of the laser) due to the removal of excited state atoms through quenching or ionisation. The number and degeneracy of the Rydberg sub-states makes this a complicated system, however, and further theoretical work is required to understand it, and in particular to identify the mechanism that leads to mixing of the levels.

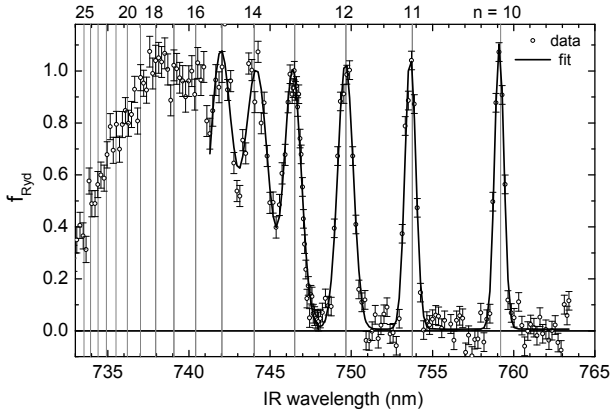

Fig 4. Lineshapes scanned from 733 to 764 nm at B = 0.16 T. The solid line is a multi-Gauss fit in the range 741-763 nm. The vertical lines indicate the expected peak positions for various Ps states based on the energy difference between each state and the 2^3P_1 level.

In Fig. 4 the transitions to Rydberg levels ranging from n = 10-15 are resolved, whereas for n > 18 and beyond the lines have become broadened, so that while it is evident that states up to n = 25 are being excited, they cannot be distinguished from one another. The six resolved lines have been fitted to a multi-Gaussian function, and there is good agreement with the expected line centers to within the ~ 0.05 nm (fwhm) accuracy of the wavelength calibration.

For states above n ~ 18, f_{Ryd} falls off rapidly, which may be due to field ionization of the Rydberg atoms. Although the dye used in the IR laser has a gain curve that peaks at ~ 750 nm, the power loss at 735 nm is only ~ 30%. A 50% power reduction obtained using a neutral density filter led to a

reduction in f_{Ryd} of only 15% (at 749.6 nm), which indicates that the observed reduction in f_{Ryd} for n > 20 is not due to the reduced laser power. The configuration of our vacuum chamber, and the need to accelerate the positron beam into the target (see fig 1) mean that there is a static electric field in the target region of ~ 200 V/cm. There will also be a motional Stark field of approximately the same magnitude in our 0.16 T magnetic field. Field ionisation of states with n = 20 does not agree with the calculations of Castelli *et al*. [11] who predict that this will not occur significantly until around n = 27 (for electric fields ~ 400 V/cm). This may be a consequence of level mixing, but in this case including interleaving of the n states, so that migration to higher n states facilitates ionization [24]. Since the production of Rydberg Ps with n > 20 may be experimentally useful this mechanism warrants further theoretical and experimental study.

One area where Rydberg Ps has already been used is in the production of cold antihydrogen, via a charge exchange reaction with excited state Ps [25]. In this scheme the Rydberg Ps was produced via interactions with highly excited Cs atoms before interacting with trapped antiprotons [26]. Laser excited Rydberg Ps with n ~ 25 is also expected to be a critical element in proposed experiments to produce an antihydrogen beam for gravitational tests on this anti-atom [11, 27].

Rydberg positronium has been directly produced before using lasers in an experiment with some similarities to that described here [4]. In that work, however, the positrons were produced by a pulsed linac with a 15 ns time width, producing ~ 10^4 Ps atoms per pulse. These were excited with ~ 10 ns wide lasers after a 70 ns delay, and we estimate that at most ~ 10^3 Rydberg atoms were produced per pulse, compared ~5×10^6 in our experiment. Moreover, the technique of single shot lifetime spectroscopy [17] provides a vastly improved signal to noise ratio, which makes it much easier to perform spectroscopy.

The total number of Rydberg Ps atoms produced in a single pulse could be increased using a more efficient Ps formation target. For example, using Si the Ps yield is almost 100% [28], and the Doppler width would be very similar to the present case. Other targets could produce Ps with ~ 50% efficiency but with a narrower Doppler spread [29] which would also increase the number of Rydberg Ps atoms produced. One might consider using a Doppler free two-photon transition, but the low cross section for this process means that the excitation laser will also have a high probability for photoionization [30]. Figure 2b suggests that increasing the UV laser power could make the initial excitation more efficient, but this will also lead to an increase in the photoionization of 2P states, and so would probably not result in a more efficient production of Rydberg atoms. Our accumulator is capable of generating positron pulses with up to 1×10^8 positrons, but the present buncher is too short to compress them [15]. With a longer buncher, however, and using a Si target, we would expect to be able to produce pulses containing around 2×10^7 Rydberg atoms.

We note that magnetised Rydberg Ps has also been previously observed [31]. These atoms were spontaneously formed with a low efficiency (< 1%) from cold surfaces with adsorbed layers of frozen gas. An interesting aspect of such atoms is that they tend to follow magnetic field lines, and so in principle could be guided to an interaction region without undue losses; it would be interesting to try and produce such Ps efficiently with a laser.

Our current experimental arrangement was not designed with Rydberg Ps in mind; it is too small to see long lived states decay before they collide with the walls, and has constrictions that require a high magnetic field for efficient positron beam transport. In future work a larger chamber will be used so that we can observe long lifetimes (> 1 μs) of Rydberg Ps. With a lower magnetic field it may be possible to perform high resolution spectroscopy on these states, and to investigate the production of higher-lying states. By producing Ps in a magnetic field-free region we will also be able to test the hypothesis that mixing of the Stark sublevels is responsible for the unexpectedly high efficiency of Rydberg atom production. By using a microwave field to generate high l states, very long lifetimes might be achieved (~ ms); these could be used to produce a collimated long-lived beam for direct measurements of the gravitational free fall of Ps atoms [14].

We are grateful to L. Liszkay, and co-workers for providing porous silica samples. This work was supported in part by the National Science Foundation under grant PHY 0900919 and by the US Air Force Research Office.

[1] W. H. Cherry, *Secondary electron emission produced from surfaces by positron bombardment*, PhD Dissertation, Princeton University, 1958).
[2] *Positron Beams and Their Applications*, edited by P. Coleman (World Scientific, Singapore, 1999).
[3] S. Chu and A. P. Mills, Jr., Phys. Rev. Lett. **48**, 1333 (1982).
[4] K. P. Ziock, R. H. Howell, F. Magnotta, R. A. Failor, and K. M. Jones, Phys. Rev. Lett. **64**, 2366 (1990).
[5] K. Michishio *et al.*, Phys. Rev. Lett. **106**, 153401 (2011).
[6] C. M. Surko and R. G. Greaves, Phys. Plasmas **11**, 2333 (2004).
[7] A. Rich, Rev. Mod. Phys. **53**, 127 (1981).
[8] D. B. Cassidy, T. H. Hisakado, H. W. K. Tom, and A. P. Mills, Jr., Phys. Rev. A **81**, 012715 (2010).
[9] R. Pohl *et al.*, Nature **466**, 213 (2010).
[10] S. G. Karshenboim, Phys. Rep. **422**, 1 (2005).
[11] F. Castelli, I. Boscolo, S. Cialdi, M. G. Giammarchi, and D. Comparat, Phys. Rev. A **78**, 052512 (2008).
[12] T. S. Pedersen *et al.*, J. Phys. B **36**, 1029 (2003).
[13] J. Mitroy, M. W. J. Bromley, and G. G. Ryzhikh, J. Phys. B **35**, R81 (2002); Xiang Cheng, D. Babikov, and D. M. Schrader, Phys. Rev. A **83**, 032504 (2011).
[14] A. P. Mills, Jr. and M. Leventhal, Nucl. Instr. and Meth. B **192**, 102 (2002).

[15] D. B. Cassidy *et. al*., Rev. Sci. Instrum. **77**, 073106 (2006).
[16] The silica targets are described in P. Crivelli *et al*., Phys. Rev. A **81**, 052703 (2010).
[17] D. B. Cassidy, S. H. M. Deng, H. K. M. Tanaka, and A. P. Mills Jr., Appl. Phys. Lett. **88**, 194105 (2006).
[18] We convert the delayed fraction into the Ps fraction using the relation $f_{Ps} \approx 2\times(f_d - 0.05)$, D. B. Cassidy, T. H. Hisakado, H. W. K. Tom, and A. P. Mills, Jr., accepted for publication Phys. Rev. B (2011).
[19] D. B. Cassidy, T. H. Hisakado, H. W. K. Tom, and A. P. Mills, Jr., Phys. Rev. A **82**, 052511 (2010).
[20] S.M. Curry, Phys. Rev. A **7**, 447 (1973); C.D. Dermer and J.C. Weisheit, Phys. Rev. A **40**, 5526 (1989).
[21] D. B. Cassidy, T. H. Hisakado, H. W. K. Tom, and A. P. Mills, Jr., Phys. Rev. Lett. **106**, 173401 (2011).
[22] We note that other laser systems suitable for Ps excitation have been proposed that will not have the same spectral properties as dye lasers. S. Cialdi *et al.*, Nucl. Instrum. Methods Phys. Res. Sec. B **269**, 1527 (2011).
[23] O. Halpern, Phys. Rev. **94**, 904 (1954).
[24] Level mixing is known to affect field ionization rates for excited state atoms. E.g., M. G. Littman, M. L. Zimmerman, and D. Kleppner, Phys. Rev. Lett. **37**, 486 (1976); T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein , Phys. Rev. A **16**, 1098 (1977); M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. Lett. **41**, 103 (1978).
[25] J. W. Humberston, M. Charlton, F. M. Jacobsen and B. I. Deutch, J. Phys. B 20 L25 (1987).
[26] E. A. Hessels, D. M. Homan, and M. J. Cavagnero, Phys. Rev. A **57**, 1668 (1998); C. H. Storry, *et. al*., (ATRAP Collaboration), Phys. Rev. Lett. **93**, 263401 (2004).
[27] A. Kellerbauer *et al.*, Nucl. Instrum. Methods Phys. Res. B **266**, 351 (2008).
[28] D. B. Cassidy, T. H. Hisakado, H. W. K. Tom, and A. P. Mills, Jr., Phys. Rev. Lett. **106**, 133401 (2011).
[29] E.g., P.J. Schultz and K.G. Lynn, Rev. Mod. Phys. **60**, 701 (1988).
[30] M. S. Fee, et. al., Phys. Rev. A **48**, 192 (1993).
[31] J. Estrada, T. Roach, J. N. Tan, P. Yesley, and G. Gabrielse, Phys. Rev. Lett. **84**, 859 (2000); C. J. Baker *et. al*., J. Phys. B: At. Mol. Opt. Phys. **41,** 245003 (2008).