Observation of Long-Lived Muonic Hydrogen in the 2S State

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The kinetic energy distribution of ground state muonic hydrogen atoms \(\mu p(1S)\) is determined from time-of-flight spectra measured at 4, 16, and 64 hPa \(H_2\) room-temperature gas. A 0.9 keV-component is discovered and attributed to radiationless deexcitation of long-lived \(\mu p(2S)\) atoms in collisions with \(H_2\) molecules. The analysis reveals a relative population of about 1%, and a pressure-dependent lifetime (e.g. 30.4\(^{+21.4}_{-9.3}\) ns at 64 hPa) of the long-lived \(\mu p(2S)\) population, equivalent to a 2S-quench rate in \(\mu p(2S)+H_2\) collisions of \(4.4^{+2.1}_{-1.4} \times 10^{11} \text{s}^{-1}\) at liquid hydrogen density.

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A measurement of the Lamb shift in muonic hydrogen, i.e. the energy difference of 0.2 eV between the \(2P\)- and \(2S\)-states of \(\mu p\) atoms \[1,2,3,4,5\], is in progress at the Paul Scherrer Institute (PSI), Switzerland \[6\]. Vacuum polarization shifts the \(2S\)-levels by 0.2 eV below the \(2P\)-levels; fine and hyperfine splittings of the \(n = 2\) levels are much smaller. The finite size effect is 2% of the Lamb shift.

The \(\mu p\) Lamb shift experiment is expected to give a precise value for the root-mean-square charge radius of the proton \[6\]. Together with recent advances in H-atom spectroscopy \[4,7\], this leads to a better determination of the Rydberg constant, and to a test of bound-state quantum electrodynamics on a new level of precision \[8\].

The most important prerequisite for such an experiment, the availability of sufficiently long-lived \(\mu p(2S)\) atoms, has so far not been experimentally established. When muons are stopped in \(H_2\) gas, \(\mu p\) atoms are formed at high \(n\)-levels and then deexcite predominantly to the ground state (“muonic cascade”). A fraction \(\varepsilon_{2S}\) of a few percent reaches the metastable \(2S\) state whose lifetime is, in absence of collisions, essentially given by the muon lifetime of 2.2 \(\mu\)s.

In a gas, there is collisional \(2S\)-quenching, with very different rates depending on the \(\mu p\) kinetic energy being above or below the \(2S\)-\(2P\) threshold (\(\approx 0.3\) eV in the lab frame) \[9\]. Most \(\mu p(2S)\) atoms are formed at energies above this threshold \[10\], where collisional \(2S \rightarrow 2P\) Stark transitions (followed by \(2P \rightarrow 1S\) radiative deexcitation) lead to rather fast \(2S\)-depletion. This is the “short-lived” \(2S\)-component with a predicted lifetime \[11\] \(\tau_{2S}^{\text{short}} \sim 100 \text{ns}/p_{H_2}[\text{hPa}]\). There is, however, a competition between such Stark transitions and deacceleration \[9\]. A fraction of the \(\mu p(2S)\) atoms should therefore survive the process of slowing down below 0.3 eV, where transitions to the \(2P\) state are energetically forbidden. These \(\mu p(2S)\) atoms form the “long-lived” \(2S\)-component, with a lifetime \(\tau_{2S}^{\text{long}}\) and a population \(\varepsilon_{2S}^{\text{long}}\) (per \(\mu p\) atom).

The \(2S\)-population \(\varepsilon_{2S}\) is well determined from the measured \(\mu p\) X-ray yields \[12,13,14\]. \(\varepsilon_{2S}^{\text{long}}\) can be derived from the measured \(1S\) kinetic energy distributions \[15\] by using calculated elastic and inelastic \(\mu p(2S)\) cross sections \[16\]. At 16 hPa, for example, \(\varepsilon_{2S} = (4.40 \pm 0.17)\%\) and \(\varepsilon_{2S}^{\text{long}} = (1.16 \pm 0.12)\%\). Calculations of the radiative quenching process during collisions \[17,18\] predicted values of \(\varepsilon_{2S}^{\text{long}} \sim 600 \text{ns}\) or larger for an \(H_2\) pressure of 64 hPa, but this could not be confirmed experimentally in searches for delayed \(n = 2\rightarrow 1\) X-rays \[19\].

We report here on the first measurement of a non-radiative deexcitation mechanism for long-lived \(\mu p(2S)\) atoms at gas pressures \(p_{H_2} = 4, 16, \) and 64 hPa at room temperature \[15\]. It was found in the analysis of time-of-flight (TOF) spectra which were measured to determine the \(\mu p(1S)\) kinetic energy distributions at low densities. A pronounced fast component showed up, corresponding to a surprisingly high \(\mu p(1S)\) kinetic energy of 0.9 keV.

The basic idea of the experiment is to form \(\mu p(1S)\) atoms near the axis of a cylindrical gas target and to deduce their kinetic energy from the measured TOF they need to reach the gold-coated cylinder surface. The muon transfer reaction \(\mu p + \text{Au} \rightarrow \mu \text{Au}^+ + 1\) to highly excited \(\mu \text{Au}^+\) takes place, and several \(\mu \text{Au}\) X-rays with energies up to 9 MeV are emitted and detected by a CsI(Tl) scintillator. The cylindrical geometry gives a better kinetic energy resolution than earlier planar geometries \[21\].

Figure \[1\] shows the experimental setup. Negative muons with a momentum of \(p = 10\) MeV/c from the high-intensity \(\pi E5\) channel at PSI entered a 1 m long, 20 cm inner diameter solenoid operated at 5 T and evacuated to \(10^{-5}\) hPa. Its fringe field reduced the radial beam size to a few mm in the region of full field. About 5000 \(\mu^/-\) reached a 16 \(\mu\)m thick Mylar entrance moderator. The muons were detected by a parallel-plate avalanche counter (PPAC) with a total areal density of 300 \(\mu\)g/cm\(^2\). The PPAC is a double gas chamber filled with 16 hPa isobutane, with a detection efficiency up to...
The muon detection efficiency of the stack-MCP assembly between the signals from PPAC and MCP. An incoming muon was identified by a delayed stop, the electrons were detected in a microchannel plate. Accelerated these electrons, and frictional cooling enhanced the muon stop rate. After traversing the target, the electrons were detected in a microchannel plate (MCP). An incoming muon was identified by a delayed coincidence between the signals from PPAC and MCP. The muon detection efficiency of the stack-MCP assembly was $\sim 40\%$ for a target gas pressure of 4 hPa and decreased to $\sim 15\%$ at 64 hPa. The radial beam distribution was measured to be approximately Gaussian-shaped with spreads $\sigma_x \approx \sigma_y \approx 1.8$ mm in the 5 T field. The cyclotron radii of the muons were $\sim 1$ mm at 5 T.

The gas target vessel was a 26 cm long Al cylinder. Its inner cylindrical surface was coated with a 500 nm thick evaporated gold layer. The side walls were covered with polypropylene. Measurements were performed with two target inner radii, $R = 29$ mm and 10 mm. A target with polypropylene instead of gold was used to determine the background. Muon transfer to carbon does not produce X-rays within the accepted $\mu$Au X-ray energy window.

The upstream and downstream target windows were thin Formvar foils (1 cm diameter) of 10 $\mu$g/cm$^2$ at $p_{H_2} = 4$ and 16 hPa, and 40 $\mu$g/cm$^2$ at 64 hPa. H$_2$ gas with $<3$ ppm impurities was continuously flushed through the target (exchange rate 2/min). Outgassing from walls and pipes resulted in a total impurity concentration of order $10^{-4}$. The measured $\mu p$ formation rate was $\sim 20$ s$^{-1}$.

A 20 cm long cylindrical CsI(Tl) scintillator of 9.2 cm inner and 19 cm outer diameter was placed around the target. It had a high probability to detect at least one of the $\mu$Au X-rays. The CsI(Tl) crystal was surrounded on all but the outer side by an 8 mm thick plastic scintillator acting as anti-coincidence detector against charged particle background. The fast light component of the plastic scintillator was distinguished from the "slow" CsI light component (decay time $\sim 1.5$ $\mu$s) by pulse shape analysis.

For each incoming muon, the time difference between the MCP signal and a CsI signal from a $\mu$Au X-ray was recorded. The true $\mu p(1S)$ TOF, however, starts at the formation of a $\mu p(1S)$. Time calibration measurements were done with a few per cent of xenon admixed to the H$_2$ target gas, because at such a low Xe concentration $\mu p$ and $\mu p$ formation times are nearly identical. The $\mu$Xe X-rays were detected in the CsI(Tl) crystal, too. Several Xe concentrations were used and the $\mu p(1S)$ formation time distribution was obtained by extrapolating to zero Xe concentration. A careful analysis of the $\mu$Xe time spectra separated the time distribution of direct $\mu$Xe-formation from the (delayed) transfer reaction $\mu p + Xe \rightarrow \mu Xe^* + p$. Small residual deviations not taken into account by the Xe calibration (like the $\mu p$ cascade time and discriminator threshold effects) were also considered.

Figure 2 shows the $\mu$Au time spectra measured at 64, 16, and 4 hPa for both target radii, and the $\mu p$ formation time distributions deduced from the $\mu$Xe data. The background was studied with the polypropylene target. It is dominated by bremsstrahlung from muon-decay electrons, with an additional component (depending on pressure) at times preceding $\mu p$ formation, correlated to muons stopping in high-Z materials near the target. All $\mu$Au time spectra exhibit a wide distribution whose maximum corresponds to $\mu p$ kinetic energies $E_{kin}$ of 5...20 eV. Such energies result from Coulomb deexcitations $\mu p + H_2 \rightarrow \mu p_{\pi < n} + E_{kin} + p + ...$ during the cascade, a well known effect for $\mu p$ and $\pi p$ atoms.

The data measured in the $R = 29$ mm target, which is most sensitive to high kinetic energies, show an unexpected peculiarity: The 64 hPa spectrum features an additional pronounced peak at $t \approx 100$ ns. This peak is also visible at 16 hPa, but almost disappears at 4 hPa. The corresponding kinetic energy is about 0.9 keV, indicated in Fig. 2 as dotted line at 0.074 $\mu$s which is the calculated TOF for 29 mm, the shortest distance from the beam center to the Au wall.

Such a high-energy $\mu p(1S)$-component has never been reported before. It can only originate from radiationless deexcitation of $\mu p(n = 2)$ atoms in collisions with H$_2$ molecules. The $n = 2 \rightarrow 1$ transition energy of 1.9 keV is shared according to momentum conservation between the formed $\mu p(1S)$ and a proton of the H$_2$ molecule:

$$\mu p(n = 2) + H_2 \rightarrow \mu p_{1S}(0.9 \text{ keV}) + p(1 \text{ keV}) + \ldots .$$

A detailed analysis of the measured time spectra was performed using a set of Monte Carlo (MC) generated TOF spectra extending over the whole expected energy range ($\sim 10^{-2} - 10^3$ eV). In the MC, the target and detector geometries, axial and radial muon stop distributions, X-ray detection efficiencies, and scattering of $\mu p$ atoms on H$_2$ molecules and on the gold surface of the target cylinder were taken into account. Cross sections for $\mu p(1S) + H_2$ scattering calculated by Adamczak for...
between 9 eV and 1000 eV we approximately included in the fit function; the most important components (a) at 0.9 keV originates from non-radiative µp(2S)-quenching and was convoluted with the fitted 2S-lifetime. The high energy Coulomb deexcitation lines with main components (b) \( n = 3 \to 2 \) (166 eV) and (c) \( n = 4 \to 3 \) (58 eV), and a continuous distribution at energies below 60 eV (d). The dashed curve (s) is the \( \mu p \) formation time distribution obtained in the Xe calibration measurements.

First the data sets at 64 and 16 hPa were analyzed independently of each other. (At 4 hPa the 0.9 keV component is too small to be treated in an independent fit.) Free fit parameters were the 2S-quench time \( \tau_{2S}^{\text{quench}} \equiv 1/\lambda_{2S}^{\text{quench}} \), the amplitude \( A_{2S} \), the time offset \( \Delta \tau_{2S} \), and the amplitudes of the Coulomb deexcitation components. The results are given in Tab. II. For \( \Delta \tau_{2S} \) a value of 8 \( \pm 1.9 \) ns was obtained at 16 hPa, and roughly 4 times less at 64 hPa, i.e. much smaller than \( \tau_{2S}^{\text{quench}} \).

Next, \( \tau_{2S}^{\text{quench}} \) was set to be proportional to \( 1/p_{H_2} \), because non-linear effects (like three-body collisions) can be excluded at our conditions. A combined fit of the data at all three pressures gave nearly the same values as the independent fit (Tab. II). The corresponding quench rate at room temperature is

\[
\lambda_{2S}^{\text{quench}} = 5.1^{+2.4}_{-2.1} \times 10^5 \text{s}^{-1} \times p_{H_2} \text{[hPa]}
\]

or, normalized to liquid hydrogen atom density (LHD = \( 4.25 \times 10^{22} \text{atoms/cm}^3 \)),

\[
\lambda_{2S}^{\text{quench}} \text{(LHD)} = 4.4^{+2.1}_{-1.8} \times 10^{11} \text{s}^{-1}.
\]
The errors result from the quadratic sum of statistical and systematic errors. The combined fit gave a reduced \( \chi^2 \) of 1.030 (\( \chi^2 = 529.5 \) for 514 degrees of freedom). \( \chi^2 = 559.2 \) resulted for \( \tau_{S\text{long}} \) (64 hPa) fixed to 9 ns, i.e. a short 2S-lifetime is statistically excluded at the 5\( \sigma \) confidence level.

Various sources of systematic errors were investigated: \( \mu \)\( \nu \) formation time, scattering on \( \text{H}_2 \) and gold, radial size of the muon stop distribution, and background shape. Uncertainties in the formation time gave a contribution of \( 5.0 \) ns to \( \tau_{S\text{quench}} \) (64 hPa), \( \text{H}_2 \)-scattering one of \( 1.0 \) ns, whereas all other errors were considerably smaller. Gas impurities modified \( \tau_{S\text{quench}} \) (64 hPa) by \( < 0.1 \) ns. The resulting total systematic errors are given in Tab. I.

The large value found for \( \lambda_{S\text{quench}} \) can be attributed to the formation of an excited muonic molecular ion in a resonantly enhanced reaction with \( \text{H}_2 \),

\[
\mu \nu \text{S}(2S) + \text{H}_2 \rightarrow \{[(\nu \nu \mu)]^* \}^* \text{pee}^*,
\]
similar to the well-known formation of ground state muonic molecules in muon catalyzed fusion \[28\]. There is a series of resonant states just below the \( \mu \nu \) dissociation limit \[23\]; rovibrational excitations of the electronic molecule absorb the small excess energy from the formation of the \( \{[(\nu \nu \mu)]^* \}^* \) “pseudonucleus”. A subsequent Auger decay to states with \( \geq 16 \) eV binding energy stabilizes the \( \{[(\nu \nu \mu)]^* \}^* \) against back decay. Mixing of the excited \( \{[(\nu \nu \mu)]^* \}^* \) with the repulsive \( \nu \nu \mu \) ground state finally leads to \( \mu \nu \text{S}(1S) \) atoms with 0.9 keV kinetic energy.

A theoretical estimate of the molecular formation rate \[23\] gave \( \approx 5 \times 10^{10} \text{ s}^{-1} \) at LHD, ten times smaller than our result (Eq. (1)). The calculation neglected the correlated motion of the three interacting atoms which is important in low-energy \( \mu \nu \text{S}(2S) \) collisions because of its long-range polarization potential. The tritatomic molecule has a much denser vibrational-state spectrum with more ways to exchange excitation energy, leading to longer effective lifetimes of the compound system and correspondingly higher molecular formation rates.

The populations \( \varepsilon_{2S} \) resulting at 16 and 4 hPa agree with the values of \( \langle 1.16 \pm 0.12 \rangle \% \) and \( \langle 1.03 \pm 0.09 \rangle \% \), respectively, evaluated for the \( \mu \nu \text{S}(2S) \) that slow down to below 0.3 eV \[12\]. This means that there is little room for radiative 2S \( \rightarrow \) 1S deexcitation processes and explains why no delayed 1.9 keV X-rays were found in previous experiments \[11, 12, 24\]. The dominance of non-radiative 2S-decay resulting from our measurements is in accord with recent calculations of the radiative and non-radiative decay rates of \( \{[(\nu \nu \mu)]^* \}^* \) molecules \[29, 32\].

In summary, we have measured that \( \approx 1 \% \) of all muons stopped in \( \text{H}_2 \) gas between 4 and 64 hPa form long-lived \( \mu \nu \text{S}(2S) \) atoms that decay non-radiatively into \( \mu \nu \text{S}(1S) \) atoms of 0.9 keV kinetic energy. For the current \( \mu \nu \) Lamb shift experiment, a pressure of \( \approx 1 \) hPa is needed where both 2S-population (\( \approx 1 \% \)) and lifetime \( \tau_{S\text{long}} = 1.04^{+0.29}_{-0.21} \mu\text{s} \) at 1 hPa) meet the experimental requirements \[4\]. First measurements at a new low-energy muon beam at PSI showed that enough \( \mu \nu \) atoms can be formed in a small gas volume at such pressures.

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