Excited State Muon Transfer in Hydrogen/Deuterium Mixtures

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We report the first direct observation of excited state muon transfer in hydrogen/deuterium mixtures by direct measurement of $q_{1s}$, the probability that a $\mu p$ atom, which is initially formed in an excited state, reaches the 1s ground state. The dependence of $q_{1s}$ on deuterium concentration $c_d$ was measured for two different densities at cryogenic temperatures using charge coupled devices (CCDs) to detect the muonic X rays. First results based on the analysis of the $K_{\alpha}$-lines of the two isotopes are presented.
One of the longstanding problems regarding the muonic cascade and muon catalyzed fusion ($\mu$CF) in mixtures of hydrogen isotopes is the poor understanding of excited state transfer of the muon. Excited state transfer largely influences the initial population of muonic atoms in the ground state, the starting point for a complex sequence of processes leading to muon catalyzed fusion [1, 2, 3, 4].

After a free muon is injected into a hydrogen (H), deuterium (D) or tritium (T) target, it is slowed down, and an excited muonic atom is formed [5, 6]. Rapid deexcitation to the ground state follows via various competing collisional and radiative processes [7]. In the first approach the fraction of muons reaching the 1s level of the lighter isotope was considered to be proportional to the atomic concentration [8] until the importance of the possibility of a fast excited state transfer e.g.

$$p\mu(nl) + d \rightarrow p + d\mu(nl') + 135/n^2eV$$

($p$=proton, $d$=deuteron) which can compete with the other cascade processes was pointed out [9]. A strong dependence on density $\Phi$, concentration $c_d$, and collision energy $\epsilon$ was predicted for $q_{1s}$. ($q_{1s}$ denotes the probability that a $\mu p$ atom, which is initially formed in an excited state, reaches the 1s ground state.) Several calculations of this functional dependence $q_{1s}(\Phi, c_i)$ have been published for the H/D, H/T and D/T mixtures in the last decade [9, 10, 11, 12, 13, 14, 15].

Until now, no direct measurement of $q_{1s}$ has been performed. The only experimental results on $q_{1s}$ so far have been obtained by indirect methods (“cycling rate analysis”) in the case of DT-fusion e.g. Ref.[4].

Therefore, we performed an experiment in H/D mixtures [16] using an entirely new approach based on the observation of the $\mu p$ and $\mu d$ X-ray intensities. The small energy difference and the very low energy of these muonic X rays (energy of $K_{\alpha}^{\mu}= 1.9$ keV, $K_{\alpha}^{d\mu} = 2.0$ keV) were the main challenges of this measurement.

This experiment took place at the high intensity muon beam of the $\mu$E4 area at PSI (Paul Scherrer Institut, Villigen, Switzerland). The setup is shown in figure 1. The incoming muon beam was defined by scintillation counters and an aluminum beam aperture with a radius of 35 mm for liquid (45 mm for gas) measurements. A muon momentum of $\sim38$ MeV/c ($\sim29$ MeV/c) was selected for the liquid (gas) runs.

Special silver-coated steel target cells have been developed for this experiment. 12.5$\mu$m thick Kapton foils were used as target windows for the measurements at liquid hydrogen density. For the gas measurements, 25$\mu$m thick Kapton windows had to withstand pressures of up to 6 bar at temperatures around 30 K. The shape of the windows was optimized to allow efficient detection of low energy X rays. The rest of the target cell was surrounded by superinsulation to reduce radiation heating. Another 12.5$\mu$m thick Kapton window separated the vacuum vessel from the vacuum of the charge coupled devices (CCDs) in order to protect the detector.
A new gashandling system was built for the preparation of the various H/D mixtures. The pressure and temperature of the target were monitored continually. During the measurements, various samples of the target content were extracted using a small capillary leading directly from the target to a quadrupole mass spectrometer [17] in order to determine the relative abundance of the two isotopes.

For the first time, CCDs [18, 19] have been employed for the observation of the muonic hydrogen X rays. Due to their excellent background suppression [20, 21, 22] they provide a unique mean for the detection of X rays in this low energy region. The size of each of the two CCD chips used was \( \sim 25 \times 17 \text{mm}^2 \) [23]. The main chip component was silicon with small absorption layers of SiN\(_3\) and SiO\(_2\) on the surface [23]. The depletion region had a thickness of \( \sim 30 \mu\text{m} \). To shorten the read-out time, each CCD chip was split into two electronically independent detection areas.

“Single pixel analysis” was used for the separation of “true” X-ray hits from charged particles or cosmic background. A “single pixel hit” was considered to be a “true” X ray if the charge content of the surrounding 8 neighbour pixels was statistically compatible with the noise peak of the CCDs [22, 24].

Systematic measurements were performed for various deuterium concentrations at two different target densities, namely at liquid hydrogen density (LHD =4.25x10\(^{22}\) atoms/cm\(^3\)) and at 1% of LHD with temperatures of 20 – 30K. Figure 2 shows the observed energy spectra of muonic deuterium, muonic hydrogen, and of an isotopic mixture at liquid hydrogen density. The small peak visible at 1.74 keV is the electronic K\(_\alpha\)-line of silicon due to fluorescence of the CCD-materials. In the mixture with \( c_d = 0.25 \) the effect of excited state transfer can clearly be seen. Without considering excited state transfer, one would expect the intensity of the deuterium K\(_\alpha\)-line to be about half of the adjoining K\(_\alpha\)\(_\mu\)-line (taking into account a \( \sim 40\% \) difference in overall detection efficiency).

In the “standard model” of the muonic cascade [4, 4, 23] the contribution of non-radiative processes to the ground state deexcitation is suggested to be only a few percent [4, 23]. This allows to determine \( q_{1s} \) in H/D mixtures by disentangling the intensities of the \( \mu p \) and \( \mu d \) K-lines. In this first analysis we considered only K\(_\alpha\) intensities. Furthermore, the atomic capture probability was considered to be equal for hydrogen and deuterium [8]. (Cohen et al. [8] calculated a difference of \( \sim 6\% \).) This approach yields

\[
q_{1s} \approx \frac{K_{\mu}^{\mu}}{1 - c_d}, \quad q_{1s} \approx \frac{1 - K_{\alpha}^{dp}}{1 - c_d}
\]

\((K_{\mu}^{\mu}...\text{Intensity of muonic hydrogen 2-1 transition, } K_{\alpha}^{dp}...\text{Intensity of muonic deuterium 2-1 transition, } c_d...\text{deuterium concentration})\).
The energy spectra were analysed using Gaussians to fit the peak areas. While the peak positions were treated as free parameters, the FWHM of the Gaussian distributions was constrained to obey the energy relation

\[
\Delta E_{FWHM}[eV] = 2.355 \times E_c \times (N^2 + \frac{F \times E}{E_c})^{1/2}
\]

(\(\Delta E_{FWHM}\) ... full width at half maximum of the fitting function [eV], 2.355 ... r.m.s. - FWHM conversion factor, \(E_c\) ... conversion energy for an electron-hole pair in silicon (3.68 eV), \(N\) ... r.m.s. transfer and readout noise of the CCDs (\(\sim 13e^{-}\) r.m.s.), \(F\) ... Fano factor (\(\sim 0.12\)), \(E\) ... energy of the X-ray [eV])

which is valid for Si-detectors \([20, 21]\). A \(\mu^+\) run and an empty target run proved that there were no background peaks visible within the observed energy region. Therefore the background function could be approximated by the sum of a constant and a term depending linearly on energy. The CERN MINUIT program package \([27]\) was employed for the fitting procedure.

An analysis of the obtained data requires the precise knowledge of the energy dependence of the detector efficiency. A Monte Carlo program \([28]\) was written to account for the various contributions to X-ray absorption by target content, windows, and CCD materials. The calculation of the intrinsic detection efficiency of the CCDs and the X-ray absorption of the Kapton foils was checked experimentally \([29]\).

In our measurement we clearly observed a change in the muonic X-ray intensity pattern of the hydrogen and deuterium cascade in isotopic mixtures at various deuterium concentrations and densities. This can be directly attributed to the existence of excited state transfer during the muonic cascade. Our results for the \(q_{1s}\) values at different densities are displayed as a function of deuterium concentration in figure 3. Statistical errors and systematic errors due to uncertainties in the efficiency of the setup contribute to the given error bars. The data point at 20% deuterium concentration is the result of a previous test experiment \([22, 30]\). For liquid hydrogen density the solid line demonstrates the theoretical expectation of Ref.\([11]\) for a collision energy \(\epsilon = 1\) eV, the dotted line shows the calculation of Ref.\([11]\) for \(\epsilon = 6\) eV, and the dashed line was calculated in Ref.\([12]\).

The data clearly reveal that the dependence of \(q_{1s}\) on deuterium concentration is much weaker than predicted by standard cascade theory. Various new ideas were published recently which offer alternative explanations and show better agreement with our results at liquid hydrogen density. Czaplinsky et al. \([11]\) assumed higher collision energies \(\epsilon\) of the muonic atoms. New calculations of the muonic cascade in hydrogen \([25]\) suggest that the energies of the muonic atoms - influenced by various cascade processes - are distributed in a complex way, especially Coulomb deexcitation processes have to be taken into account. The resulting values for \(q_{1s}\) are given in fig.3 (dashed-dotted line) \([13]\). A new mechanism for deexcitation of the muonic atom via an excited \((d\mu)^{\ast}\) molecule was proposed in \([13, 14]\) which suggests
an alternative non-radiative transition to the ground state. The existence of this “resonant sidepath” in the \(\mu\)CF-cycle - up to now one calculation exists for D/T mixtures only - would influence significantly the theoretical expectation for \(q_{1s}\) and its dependence on \(c_d\).

Within error limits, our results agree with the ones obtained by analysis of cycling rates in the case of D/T mixtures given in Ref.[4].

Figure 4 displays the density dependence of \(q_{1s}\) at different deuterium concentrations. A large difference between data taken at liquid hydrogen density and at 1% of LHD can be seen. A more detailed result on the density dependence of \(q_{1s}\) is expected from the final analysis, which will include measurements at 4% and 8% of LHD.

Financial support by the Austrian Science Foundation, the Austrian Academy of Sciences, the Swiss Academy of Sciences, the Swiss National Science Foundation, Paul Scherrer Institute and the German Federal Ministry of Research and Technology is gratefully acknowledged. It is a pleasure to thank D.Sigg for his software support. We are grateful to E.Steining and H.Weiss for their help during the experiment.

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Figure 1: Top view of the setup of our experiment for the measurements in gaseous H/D mixtures. Two separated vacuum vessels were used for the target (V1) and the CCDs (V2). The scintillation counters 3 and 3a were used to define incoming muons; the electron counter served to detect electrons following muon decay. For the measurements at liquid hydrogen density a smaller target cell (50 x 50 x 34 mm) was used.
Figure 2: Energy spectra of a) liquid D$_2$, b) a liquid H/D mixture containing 25% deuterium and c) liquid H$_2$. The solid lines indicate the fits of the various peaks.
Figure 3: The dependence of $q_{1s}$ on deuterium concentration. Filled dots display the results for liquid hydrogen density measurements, the point at $c_d = 0.2$ is taken from Ref. [22, 30]. Triangles show the measurements at 1% of LHD. The theoretical expectations for $q_{1s}$ at liquid hydrogen density are taken from Ref. [11] calculated for a collision energy $\epsilon = 1$ eV (solid line), $\epsilon = 6$ eV (dotted line), Ref. [12] (dashed line), and Ref. [15] (dashed-dotted line) (scaling factor $k_t = 0.5$).
Figure 4: Dependence of $q_{ls}$ on target density for three different deuterium concentrations. Dots refer to measurements at $c_d = 0.1$, stars to $c_d = 0.25$ and triangles to $c_d = 0.5$. The density is given relative to LHD.