Study of Capture and Cooling of H\(^-\) Ions in Radiofrequency Octopole in Magnetic Field

Š. Roučka, P. Jusko, D. Gerlich, R. Plašil, J. Glosík
Charles University, Faculty of Mathematics and Physics, Prague, Czech Republic.

Abstract. This contribution presents details of the design of a novel apparatus for studying the associative detachment reaction H + H\(^-\) \rightarrow H\(_2\) + e\(^-\). We focus on investigating the confinement of H\(^-\) ions in radiofrequency linear octopole trap and on the possibility of cooling them by collisions with atoms of He buffer gas. The possibility of using the octopole as a mass filter for calibration purposes is investigated. We further estimate the influence of radiofrequency field on the measured energy of detached electrons. Besides the ion optics, we discuss the focusing properties a hexapole magnetic lens for neutral atoms.

Introduction

This work is a part of our current project – study of associative detachment H + H\(^-\) \rightarrow H\(_2\) + e\(^-\) (AD for short). The motivation behind this study and some early design considerations were presented by Roučka et al. [2009]. Since then we moved on to putting the apparatus into operation. This paper presents the final design of apparatus for measurement of the rate constant and for the measurement of energy distribution of detached electrons. For test measurements, we decided to use trapped ion beam in octopole RF trap at room temperature instead of 22-pole mounted on cold-head which allowed us to greatly simplify the design of magnetic adiabatic collimator Roučka et al. [2009].

The ES-MPT apparatus

Our experimental apparatus under construction is designated by the abbreviation ES-MPT (Electron Spectrometer with MultiPole Trap). The apparatus is based on the guided ion beam technique and has a modular design that allows us 1) to measure rate coefficients for the disappearance of H\(^-\) and 2) to detect the detached electrons and 3) to record their energy distribution using electron spectrometer. A scheme of the apparatus with electron spectrometer is shown in figure 1. Its principal parts are:

![Figure 1. The schematic drawing of ES-MPT apparatus](image-url)

Hydrogen atom source: The atomic hydrogen is produced in an inductively coupled RF discharge in pure hydrogen. Atoms can be cooled by a PTFE accomodator mounted on cold head to less than 10 K. The produced effusive beam of atomic hydrogen can then be focused by hexapole magnet. For description of this source see [Borodi et al., 2009].
**Storage ion source:** Ions in the Storage ion source are produced by electron bombardment. They are stored in the RF field and ejected in pulsed or continuous mode. The source is transparent on axis, so the atomic beam can go straight through. Thorough description of this source can be found in [Gerlich, 1992].

**Quadrupole mass filter:** After ejection from the ion source, ions are mass-selected in the quadrupole mass filter. In case of H$^-$, the quadrupole is operated in the low pass regime, because H$^-$ is the lightest negative ion.

**Octopole ion guide:** The radiofrequency octopole serves multiple purposes in this experiment. Ions are guided in the octopole RF field to a region between two ring electrodes that can generate potential barriers for trapping of ions in this part of octopole. The trapped ions interact with the beam of atomic hydrogen and from their disappearance rate, the rate constant of AD can be calculated. For calibration of the hydrogen beam density, we will use the octopole also as a mass filter.

**Magnetic adiabatic collimator with electrostatic filter (MAC-E filter):** Electrons produced in the AD reaction are guided by the collimator magnetic field and filtered by the retarding potential barrier.

**MCP detector:** We use a microchannel plate detector for detection of ions and electrons.

**MAC-E filter construction**

The idea behind MAC is, that all produced electrons with random velocity directions are captured by magnetic field and their velocities are then collimated by special configuration of magnetic field [Beamson et al., 1980]. The basic design has been described by Roučka et al. [2009] and here we will just repeat, that the energy resolution is given by the ratio of maximal and minimal field in collimator e.g. $\Delta E/E = B_{\text{min}}/B_{\text{max}}$. The theoretical relative resolution of our collimator is 1:30. The cross section of the MAC-E filter is depicted in the figure 2. The *Main coils* generate the strong field in the reaction space. The ferromagnetic rings (blue) enhance the field in the trap and lower it outside. After filtering, the electrons are focused back to the detector using the focusing coil.

The magnetic collimator coils and ferromagnetic shields have been constructed according to figure 1. The main coils have 300 turns and the focusing coil has 200 turns, the current through all coils is 4.5 A. We measured the generated magnetic field using Hall probe. The probe was calibrated by the field of focusing coil, which is not influenced by the ferromagnetic rings. Comparison with the calculated field is presented in figure 3. We can see, that the measured field is slightly higher in the trap area, which is caused by higher magnetic permitivity of the construction steel in the ferromagnetic rings. The difference is small and will not influence the function of MAC-E filter.
Rate constant measurement – octopole as a mass filter

During the measurement of the rate constant, we will observe the loss of ions from the trap due to the interaction with beam of H atoms. The rate of reaction in the trap at temperature $T$ is given by

$$r = \int_V k(T, n_i(x)) n_H(x) dV.$$  

We can factor out the total number of ions $N_i$ from this equation by using normalized ion density $f_i(x) = n_i(x)/N_i$ and obtain the equation

$$r = k(T) N_i N_H; \quad N_H = \int_V f_i(x) n_H(x) dV,$$  \hspace{1cm} (1)

where the integral represents an effective hydrogen atom density $N_H$. The $N_H$ must be determined experimentally and in our case we will use a reaction with well known rate constant for calibration. In this procedure we correct for the effective potential so that the distribution $f_i$ of the calibration ion is equal to the distribution of $H^-$. The ion used for calibration will be $CO_2^+$ which reacts with H and $H_2$ mainly by these reactions [Borodi et al., 2009]:

$$CO_2^+ + H \rightarrow HCO^+ + O \quad k_1 = 4.5 \cdot 10^{-10} \text{ cm}^3 \text{ s}^{-1} \hspace{1cm} (2)$$

$$CO_2^+ + H_2 \rightarrow HCO^+ + H \quad k_2 = 9.5(T/300 \text{ K})^{-0.15} 10^{-10} \text{ cm}^3 \text{ s}^{-1}. \hspace{1cm} (3)$$

The evolution of this system for initial number of $CO_2^+$ ions $[CO_2^+]_0$ is described by equations

$$[HCO^+] = \frac{k_1[H][CO_2^+]_0}{K} (1 - \exp(-Kt)) \quad [CO_2^+] = [CO_2^+]_0 \exp(-Kt)$$

$$[HCO_2^+] = \frac{k_2[H_2][CO_2^+]_0}{K} (1 - \exp(-Kt)) \quad K = k_1[H] + k_2[H_2]. \hspace{1cm} (4)$$

In order to calibrate for effective H density, we must mass select the reaction products $HCO^+$ and $HCO_2^+$. Our apparatus does not have a quadrupole mass spectrometer (QPMS) between reaction chamber and MCP detector and therefore we are planning to use the octopole itself as a mass filter in low pass regime. However, for more precise measurements of absolute rate constant, we will use the 22-pole trap with QPMS for product ions. In this experiment we expect, that we will be able to resolve between masses 29 and 45 amu and this measurement will give us enough information for calibrating the $N_H$. The low pass regime boundary for ideal multipole is given by Gerlich [1992]

$$qU_0 < \frac{1}{8} \left( \frac{gV_0}{\epsilon} \right)^2; \quad \epsilon = \frac{1}{2n^2} \omega^2 r_0^2.$$  \hspace{1cm} (5)
Where $2U_0$ is the DC potential difference between the multipole rods, $V_0$ is the RF amplitude, and $m$ is the ion mass. Other symbols use the standard nomenclature defined e.g. in the original work by Gerlich [1992]. This theoretical boundary is compared with our numerical calculation for octopole with inner radius $r_0 = 3$ mm, rod diameter 2 mm and RF frequency 7 MHz in figure 4. The results were obtained by injecting particles with temperature 300 K in a beam with diameter 4 mm. The graphs show the fraction of particles that was still trapped after 100 µs. We see that the real octopole has the boundary at a slightly different position, but it is very sharp and should be suitable for mass filtering of reaction products. For reliable determination of $N_{H}$ the ions have to be captured for a time of the order $1/K$.

![Figure 4. Stability diagrams for octopole RF trap, $\Omega = 7$ MHz.](image)

**Adverse effects of RF field**

The trapping of particles in octopole has also some disadvantages in comparison to trapping in 22-pole trap, especially for buffer gas cooling of ions with lower mass than the buffer gas. The electric potential amplitude in octopole is proportional to $r^4$ in contrast to $r^{11}$ in 22-pole, hence it does not have such a wide nearly field free region [Gerlich, 1992]. Because of this, the ions often collide with the buffer gas in the regions of relatively high RF amplitude. If a light ion collides with a cold heavier particle during the oscillatory motion, its direction of motion can be completely reversed which could lead to tripling of its effective energy in the worst case.

We calculated the ion velocity distributions using the same procedure as described in [Roučka et al., 2009] for the octopole trap with helium buffer gas, $\Omega = 10$ MHz; $V_0 = 20$ V. Isotropic scattering with Langevin cross section was assumed. The results of this calculation are shown in figure 5. We can see, that the body of the energy distribution can be described as a combination of maxwellian distributions at temperatures 300 K and 1000 K with ratio 1:1. This calculation shows the energy distribution under the most unfavorable conditions. In reality the ions will be cooled mainly by collisions with molecular hydrogen, which can efficiently cool the ions by inelastic collisions due to its vibrational and rotational energy levels. Moreover, the RF frequency 10 MHz is enough for ions at 300 K, but once the get accelerated by collision, the can leave the adiabatic regime and be further accelerated by RF field. In our experiment we therefore intend to use RF frequency above 20 MHz and we expect, that the energy distribution will be much closer to 300 K Maxwell distribution.

The measurement of electron energy will also be influenced by the RF field, because the electrons will be produced in regions of nonzero RF amplitude and the uncertainty of their energy could be estimated as the local RF amplitude. We calculated the distribution of $H^-$ ions in the trap, which corresponds to the relative rate of electron production for a homogeneous H beam. The results of this calculation for RF trap with and without magnetic field are plotted in
Figure 5. Ion energy distribution function in octopole with helium buffer gas cooler.

Figure 6. The maximal distortion of electron energy distribution by RF field.

Magnetic focusing of atomic beam

In order to improve the intensity of hydrogen atomic beam, we intend to use a linear hexapole magnet as a focusing lens. The use of magnets for manipulating with atomic beams dates back to the famous Stern Gerlach experiment and has been applied to atomic hydrogen by Phipps and Taylor \[1927\]. The magnetic moment of atomic hydrogen in the ground state is determined by the electron spin, because the gyromagnetic ratio \( \gamma \) of proton is negligible in comparison with \( \gamma \) of electron for our purpose. The gradient force exerted on a magnetic dipole \( \mathbf{m} \) in magnetic field is given by \( \mathbf{F} = \nabla (\mathbf{m} \cdot \mathbf{B}) \) (see discussion by Boyer \[1988\]). The projection of electron magnetic moment along the magnetic field is given by \( \mu = -\mu_B g_s m_s \), where \( \mu_B \) is the Bohr magneton, \( g_s \) is the electron spin \( g \)-factor and \( m_s = \pm 1/2 \) is the spin quantum number. The force is then given by \( \mathbf{F} = -\mu \nabla B \). The hexapole magnet has the special property that \( \partial^2 B / \partial r^2 = M = \text{const} \) where \( M \) is so-called hexapole constant \[Kaenders et al., 1996\]. The magnet thus creates a harmonic guiding field for atoms with force equal to \( \mathbf{F} = -\mu M r \).

We will now calculate the focusing properties of the hexapole magnet. We have a setup schematically described in figure 7. Particles are starting with random direction from point
source located at \( z = 0 \). The length of the hexapole magnet is \( l_2 \) and it starts at distance \( l_1 \) from the origin. Particles are detected at \( z = L_3 \). Let us now calculate the trajectory of a particle with velocity \( v_0 \) and angle \( \phi_0 \) with respect to \( z \) axis. The particle will enter the hexapole at a radial distance \( r_1 = v_0/l_1/v_z \). In the hexapole, the particle will perform oscillatory motion described by \( r_{II}(t) = A\cos(\omega t + \delta) \) with angular frequency \( \omega = \mu M/m \). The trajectory can be obtained by substituting the time for \( z/v_0 \). The constants \( A \) and \( \delta \) will be found by matching this solution to the linear trajectory before entering the magnet \( r_I(z) = v_0/z/v_0 \). The possible refraction at the ends of the hexapole can be neglected if we assume that the axial energy of the particle is much higher than the hexapole potential, \( E_z \gg \frac{1}{2}\mu Mr^2 \). For a magnet with \( M = 30800 \) T/m² the hexapole potential at radius 5 mm is equal to 22 µeV. The trajectory then must be smooth up to its first derivative:

\[
A \cos(\omega l_1/v_z + \delta) = r_1; \quad -A\omega \sin(\omega l_1/v_z + \delta) = v_0.
\]

These transcendental equations can be solved analytically to give

\[
A = r_1\sqrt{1 + \left(\frac{v_0}{\omega r_1}\right)^2}; \quad \delta = -\arctan\frac{v_0}{\omega r_1} - \frac{\omega l_1}{v_0}.
\]

After leaving the magnet, the particle will continue in linear motion \( r_{III}(z) = r'_{II}(L_2)\cdot(z-L_2) + r_{II}(L_2) \), where the prime denotes derivative w.r.t. \( z \). Position of the particle in the detector plane is given by \( r_3 = r'_{II}(L_2)l_3 + r_{II}(L_2) \). After substituting we obtain

\[
r_3 = A\cos\left(\frac{\omega L_2}{v_z} - \delta\right) - A\frac{\omega l_3}{v_z}\sin\left(\frac{\omega L_2}{v_z} - \delta\right).
\]

Now we can calculate the amplification of the beam by evaluating the maximal initial angle (acceptance angle) \( \phi_0 \) for which the atoms still enter a specified aperture. The angle cannot be expressed analytically from equation (8) and it has to be solved numerically. In this approximate calculation we will use following parameters: \( l_1 = 50 \) mm; \( l_2 = 105 \) mm; \( l_3 = 1000 \) mm; \( M = 30800 \) T/m²; Aperture radius \( r_a = 2 \) mm (this corresponds to “magnet II” described by Borodi et al. [2009]). The amplification is obtained by dividing the solid acceptance angle (square of acceptance angle for small angles) by the solid acceptance angle without magnet. Its values are plotted in figure 8. The peaks are are cut off because of an additional condition, that the amplitude of oscillations \( A \) cannot be higher than the inner radius of hexapole magnet \( r_H = 7 \) mm. Influence of other apertures is not considered in this calculation.

**Conclusion**

We presented our design of a novel apparatus for studying of the associative detachment reaction and especially for measurement of the energy of detached electrons. The magnetic...
adiabatic collimator for electrons has been constructed and its measured magnetic field is in good agreement with our initial design. It was shown, that it will be possible to calibrate the H atom beam intensity in our setup. This will allow us to measure the absolute values of rate constant in addition to relative measurement. Our simulations of buffer gas cooling show, that it is difficult to achieve thermal equilibrium of H⁻ with buffer gas. The situation can be improved by using cold effusive beam of molecular hydrogen for cooling (hydrogen atom source without discharge), because its temperature can be varied down to 10 K and the ions will collide mainly in the field free regions. The effects of RF field on measured EEDF were estimated and a method for their elimination was proposed. Finally we discussed the focusing of atomic beam by a magnetic lens. The amplification of the beam as a function of atom energy was calculated.

**Acknowledgments.** This work is a part of the research plan MSM 0021620834 financed by the ministry of Education of the Czech Republic and was partly supported by GACR (205/09/1183, 202/09/0642), GAUK 54010, and GAUK 25709

**References**


