Dissociative recombination of $^3HeH^+$: comparison of spectra obtained with 100, 10 and 1 meV temperature electron beams

A. Al-Khalili [a], H. Danared [b], M. Larsson [a], A. Le Padellec [a], R. Peverall [c], S. Rosén [a], J. Semaniak [d], M. af Ugglas [b], L. Vikor [a] and W.J. van der Zande [c]

[a] *Department of Physics, Stockholm University, Box 6730, S-113 85 Stockholm, Sweden*
[b] *Manne Siegbahn Laboratory, Stockholm University, S-104 05 Stockholm, Sweden*
[c] *FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*
[d] *Institute of Physics, Pedagogical University, ul. Lesna 16, 25-509 Kielce, Poland*

The heavy-ion storage ring CRYRING at the Manne Siegbahn Laboratory at Stockholm University has been used for the study of dissociative recombination of $^3HeH^+$. The new adiabatically expanded electron beam at CRYRING, which is achieved by means of a superconducting magnet, was used. The electron-beam expansion factor of 100 gave a transverse electron temperature of about 1 meV. This allowed the observation of several new resonances in the recombination cross-section.

1. Introduction

The HeH^+ ion belongs to a category of molecules for which dissociative recombination cannot occur through a neutral, resonant state crossing the ion ground state [1]. For some molecular ions the neutral state crossing occurs in such a way that only vibrationally excited levels can recombine through the resonant state. HeH^+, on the other hand, represents a "pure" case in the sense that there *is* no neutral state crossing of the ion ground state. The absence of a crossing led to suggestions that HeH^+ does not undergo dissociative recombination [2]. The merged beams experiment by Yousif and Mitchell [3] showed that there were strong reasons to believe that HeH^+ could recombine much faster than is suggested in [2] and also faster than the rate inferred from a flowing afterglow experiment [4]. A series of storage ring experiments has since then firmly established that HeH^+ recombines quite effectively even without a curve crossing [5–10].

Theoretical rationals for the dissociative recombination of HeH^+ have been put forward, combined with quantitative calculations from first principles [11–13]. Dissociative recombination of HeH^+ occurs through nonadiabatic radial coupling between the electron-molecular ion system and neutral Rydberg states that are close to the inner turning point of $v = 0$ of the HeH^+ ground state. The cross-section becomes small

above an incident electron energy of 1 eV. It increases again at 10 eV and peaks at 20 eV. The high energy cross-section is of a different origin than the one at low energy; it is also quite well understood from a theoretical point of view [14,15].

The low-energy cross-section for dissociative recombination of HeH^+ contains a number of resonances, which occur from the interference of Rydberg levels. The resonances show a clear isotope effect, as shown in [9,12]. The first storage ring observation of a low-energy resonance structure in dissociative recombination of HeH^+ [8] was made possible by the development of an adiabatically expanded electron beam [16], which lowered the transverse electron temperature in CRYRING from $kT_\perp = 100$ meV to $kT_\perp = 10$ meV. The electron beam was expanded from 1.25 cm^2 to 12.57 cm^2 in a longitudinal magnetic field, which decreased in field strength a factor of ten. In order to achieve an expansion of a factor of hundred, a superconducting magnet at the electron gun is required. This has recently been accomplished at TARN II [17] and CRYRING. The 1 meV electron beam in TARN II was used to measure the dissociative recombination of $^3HeH^+$ [17]. As a first test of the new $kT_\perp = 1$ meV electron beam we have used the same system in a study of its cross-section below 1 eV. $^3HeH^+$ has previously been studied at CRYRING at $kT_\perp = 10$ and 100 meV. The present experiment allows a comparison with the earlier data, a comparison with the TARN II data, and a consistency check of the complete set of data obtained at CRYRING between 1993 and 1997.

2. Experiment

The experiment was performed at the heavy ion storage ring CRYRING at the Manne Siegbahn Laboratory at Stockholm University. Here is given a brief description of the experiment and data analyses and more details can be found in the paper of Strömholm et al. [9] and references therein. After mass selection, $^3HeH^+$ ions created in a hot-cathode-discharge ion source were pre-accelerated to 300 keV/amu in an RFQ and then injected into the storage ring and accelerated to an energy of 3.0 MeV/amu. In the section of the storage ring known as electron-cooler, the ion beam was merged with a beam of cold velocity-matched electrons. The ions were stored in the ring for 6 s before the measurements. During this time the ion cooling occurred, i.e., the random thermal motion of the ions was reduced by the interaction with electrons. The ion beam also shrank in diameter, and the ions relaxed through infrared emission to the ground vibrational state, which makes the results suitable for comparison with theory.

After the cooling, the longitudinal electron velocity was tuned away from the velocity-matched condition during short time intervals when data was taken. The difference of the ion and electron velocities is called the detuning velocity v_d. It defines the detuning energy E_d according to the relation $E_\mathrm{d} = m_\mathrm{e} v_\mathrm{d}^2/2$. The measurement proceeded in such a way that the time interval with electron velocity v_d alternated with time intervals when the cooler was reset to the velocity-matching condition. This procedure was followed in order to keep the ion beam translationally cooled and in

order to avoid a change of the ion beam energy due to the drag-force effect. The switching procedure was repeated five times during each injection cycle.

The neutral products from the DR process as well as neutrals produced in collisions with the rest gas in the cooler were detected by an ion-implanted surface barrier detector. The pulse-height spectra, corresponding to energy released at the detector, were recorded using a multichannel analyzer (MCA).

3. Data analysis

Dissociative recombination of $^3HeH^+$ results in the formation of two neutral atoms according to the reaction:

$$^3HeH^+ + e \rightarrow He + H + \text{kinetic energy}.$$

The probability for electron capture by the rest gas is very low at 3.0 MeV/amu, and the other collision processes in the rest gas lead to the formation of at least one charged particle. Since the neutral fragments from these processes, He and H atoms in this experiment, carry only a fraction of the kinetic energy of the stored

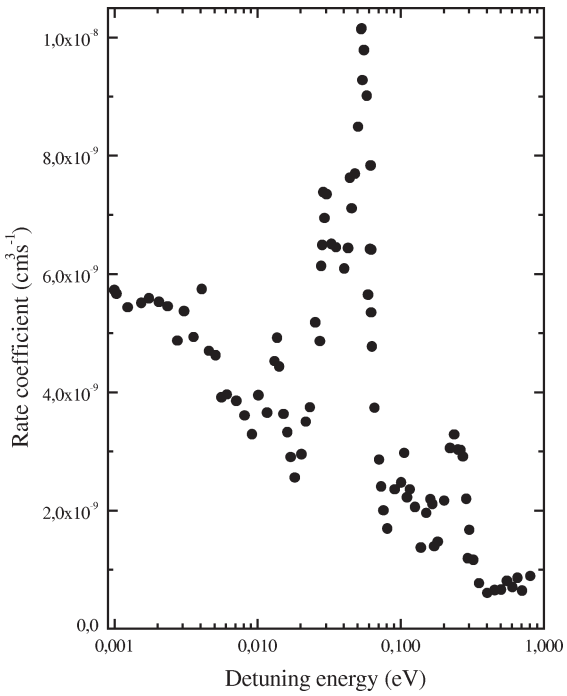

Figure 1. Rate coefficient for dissociative recombination of the $^3HeH^+$ ion obtained in the present work with a transverse electron temperature of 1 meV. Note that this is not an absolute measurement. The scale was set as described for figure 2.

beam, they result in separate "background peaks" in the MCA spectrum. The peak at maximum energy in the MCA spectrum corresponds to events when He and H atoms from the DR process impinge on the detector simultaneously. The DR rate coefficient is proportional to the ratio of the integrated number of counts from the "DR peak", divided by the integrated number of counts from the "background peak" (see, for example, [9]). The integration is performed during the time when the electrons are detuned.

The measurement was performed in the energy range $E_\mathrm{d} = 0.001$–1 eV. In the present evaluation, the relative value of the DR rate coefficient is obtained by dividing the number of counts from "DR peak" with the number of counts from the "background peak" that corresponds to He. The ion current, which is necessary for obtaining the absolute value of the DR rate coefficient, was not measured in the present experiment. The data was set to an absolute scale by adjusting the high-energy part of the spectrum to the corresponding part of the absolute DR rate coefficient spectra measured for $kT_\perp = 10$ meV [8] and 100 meV [7]. Figure 1 shows the resulting spectrum for $kT_\perp = 1$ meV. The spectra for $kT_\perp = 1$, 10 and 100 meV are presented in figure 2 on an absolute scale. The transverse electron temperature is the resolution limiting factor for $E_\mathrm{d} < 100$ meV, as can be seen in figure 2.

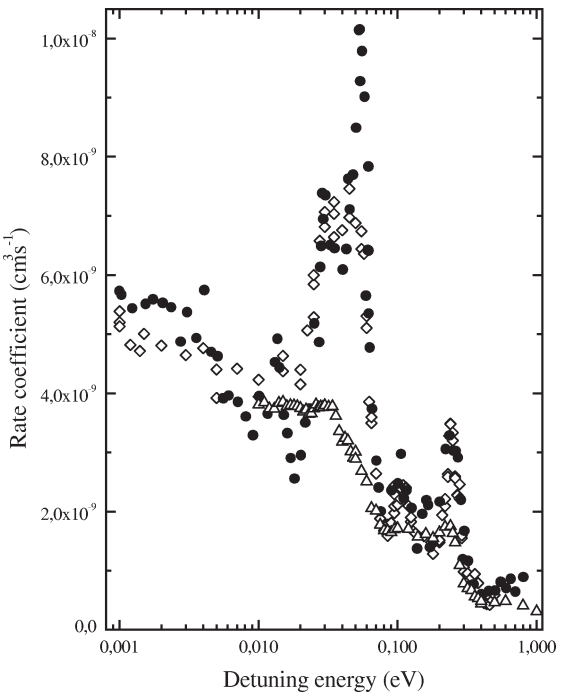

Figure 2. Rate coefficients for dissociative recombination of $^3HeH^+$ ion with electron beam of 1 meV, 10 meV [8] and 100 meV [7] transverse electron temperature. Data for 1 meV are scaled to fit the absolute data for 10 and 100 meV (see the text).

It is important to check if the differences in the DR rate coefficient spectra for $kT_\perp = 1$, 10 and 100 meV are caused only by the change of the electron temperature. The rate coefficient

$$\left\langle v_\mathrm{d}\sigma(v)\right\rangle = \int \sigma(v) v f(v_\mathrm{d}, \boldsymbol{v}_\mathrm{e})\, \mathrm{d}^3\boldsymbol{v}_\mathrm{e} \tag{1}$$

measured in the electron cooler, is an average of a velocity-weighted cross-section $\sigma(v)$ over the following "flattened" electron-velocity distribution

$$f(v_\mathrm{d}, \boldsymbol{v}_\mathrm{e}) = \frac{m_\mathrm{e}}{2\pi k T_{\mathrm{e}\perp}} \left(\frac{m_\mathrm{e}}{2\pi k T_{\mathrm{e}\|}}\right)^{1/2} \exp\left(-\frac{m_\mathrm{e} v_{\mathrm{e}\perp}^2}{2kT_{\mathrm{e}\perp}} - \frac{m_\mathrm{e}(v_{\mathrm{e}\|} - v_\mathrm{d})^2}{2kT_{\mathrm{e}\|}}\right), \tag{2}$$

where

$$v = \left[v_{\mathrm{e}\perp}^2 + v_{\mathrm{e}\|}^2\right]^{1/2} \quad \text{and} \quad \mathrm{d}^3\boldsymbol{v}_\mathrm{e} = 2\pi v_{\mathrm{e}\perp}\mathrm{d}v_{\mathrm{e}\perp}\mathrm{d}v_{\mathrm{e}\|} \text{ in eq. (1).}$$

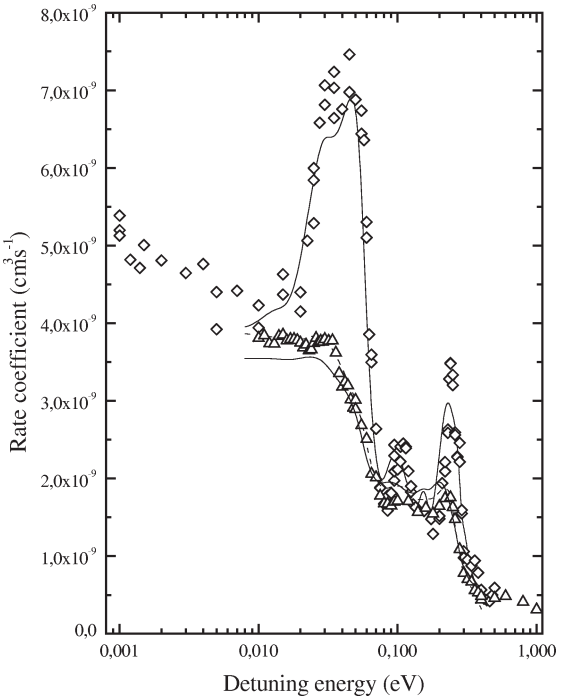

Figure 3. The measured rate coefficient for $kT_\perp = 10$ meV (open squares), shown together with the rate coefficient calculated starting from the measured data obtained for $kT_\perp = 1$ meV; the measured rate coefficient for $kT_\perp = 100$ meV (open triangles), shown together with the rate coefficients calculated starting from the measured data obtained for $kT_\perp = 1$ meV (full line) and for $kT_\perp = 10$ meV (broken line). For the calculation procedure, see text.

The measured rate coefficient at 1 meV divided by the detuning velocity, $\langle v\sigma(v)\rangle/v_\mathrm{d}$ was used as an approximation for $\sigma(v)$. This effective cross-section,

$$\sigma_\mathrm{eff}(v) = \frac{\langle v_\mathrm{d}\sigma(v)\rangle_{kT_\perp=1\ \mathrm{meV}}}{v_\mathrm{d}},$$

was inserted in the integrand in eq. (1) together with electron-velocity distributions characterised by transverse temperatures of 10 meV and 100 meV, and a longitudinal temperature of 0.1 meV. Figure 3 shows a comparison of the "theoretical", folded, rate coefficients for 10 meV and 100 meV and those measured in CRYRING when the electron-cooler beam was characterised by 10 meV [8] and 100 meV [7]. The same procedure was also followed starting with an effective cross-section

$$\sigma_\mathrm{eff}(v) = \frac{\langle v_\mathrm{d}\sigma(v)\rangle_{kT_\perp=10\ \mathrm{meV}}}{v_\mathrm{d}}$$

folded wih a $kT_\perp = 100$ meV electron distribution. It follows from figure 3 that the three sets of data are consistent.

4. Discussion

The present experiment, which was carried out using the hundred-fold expanded, $kT_\perp = 1$ meV, electron beam in CRYRING, reveals new resonances in the dissociative recombination cross-section of $^3HeH^+$. The most conspicuous features as compared with the data taken at 10 meV [8] are the double-peak structure at 30 meV and 60 meV, and the Fano-like resonance at 15 meV. The present data are also consistent with the 1 meV data taken at the TARN II storage ring [17], at least with respect to the positions of the resonances. We note that our rate coefficient at 60 meV divided with that at 20 meV is 4.0, whereas the same ratio for the TARN II data is 2.3. The difference is probably due to a slightly colder electron beam in CRYRING, or better optimised conditions during the data taking in terms of translational cooling and beam alignment.

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