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Rotational Cooling of HD^+ by Superelastic Collisions with Electrons

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Abstract. Rotational cooling of HD^+ by superelastic collisions (SEC) with electrons was observed at the Heidelberg test storage ring by merging a beam of rotationally hot HD^+ ions with an electron beam at zero relative energy. Neutral fragments resulting from DR events were recorded at different electron densities using a high resolution imaging detector and a large-area, energy sensitive detector. The data allowed to deduce the time dependence of the population of three groups of rotational angular momentum states J built on the vibrational ground state of the ion together with the corresponding DR rate coefficients. The latter are found to be (statistical uncertainties only) $\langle\alpha\rangle_{0,1,2} = 3.8(1)$, $\langle\alpha\rangle_{3,4} = 4.0(2)$, and $\langle\alpha\rangle_{5,6,7} = 9.0(1.3)$ in units of $10^{-8} \text{ cm}^3/\text{s}$, in reasonable agreement with the average values derived within the MQDT approach. The time evolution of the population curves clearly reveals that rotational cooling by SEC takes place, which can be well described by using theoretical SEC rate coefficients obtained by combining the molecular R-matrix approach with the adiabatic nuclear rotation approximation. We verify the $\Delta J = -2$ coefficients, which are predicted to be dominant as opposed to the $\Delta J = -1$ coefficients and to amount to $(1 - 2) \cdot 10^{-6} \text{ cm}^3/\text{s}$, to within 30%.

1. Introduction

Collisions between electrons and molecular ions are of fundamental interest, both from a theoretical point of view as well as in connection with many applications. At low temperatures prevailing in a number of astrophysical environments, for example, inelastic collisions (IEC) between electrons and molecular ions are competing with dissociative recombination (DR), and a thorough knowledge of these processes is crucial to model the physics and chemistry taking

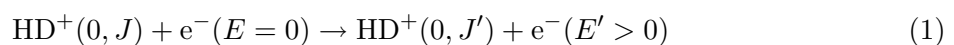
place in these plasmas. While the DR process has been investigated both experimentally and theoretically in rather detail in the last two decades [1], only very few experimental studies have been performed so far dealing with IEC processes at very low electron energies. In particular, only one quantitative experimental investigation of the superelastic collisions (SEC) process, which results in a deexcitation of ro-vibrationally excited molecular ions, has been performed so far: Krohn et al. [2, 3] measured the $\Delta v = -1$ SEC rate coefficients between the lowest vibrational states of H_2^+ . The SEC rate coefficients were found to be surprisingly large, amounting to approximately $1 \cdot 10^{-6} \text{ cm}^3/\text{s}$, about one order of magnitude larger than the corresponding DR rate coefficients. To explain these large vibrational SEC rate coefficients it turned out that detailed account has to be taken of the rotational structure and couplings of the $\text{H}_2^+ + e^-$ collision system [4].

As in H_2^+ and other symmetric molecular ions the cooling via radiative transitions is strongly hindered, and superelastic collisions seem to occur much more frequently than the destruction of the ions by the DR process, the SEC process may thus be used in storage ring experiments with merged electron-ion beams to vibrationally cool these ions. For infrared active molecular ions, on the other hand, the cooling of vibrational excitations by radiative transitions is quite fast and the molecules usually end up in their vibrational ground state within seconds. The situation is quite different, however, as far as the cooling of the rotational degree of freedom is concerned: Once the vibrational ground state is reached, the rotational cooling has to proceed along the ground state rotational band and several tens of seconds are needed to just reach equilibrium with the 300 K ambient radiation field. Assuming the SEC rate coefficients between rotational states to be of the same order of magnitude as those between vibrational states, it should be possible to use the SEC process to speed up the rotational cooling and to reach even subthermal rotational populations.

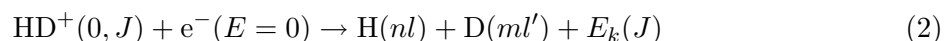
Although the occurrence of rotational cooling by SEC in storage ring experiments has been conjectured before (see e.g. [5]), no quantitative measurements of rotational SEC rate coefficients has been carried out so far. A dedicated experiment was therefore performed to study the rotational cooling of HD^+ by the SEC process [6]. This molecule is ideally suited for these investigations: Hot stored HD^+ ions were shown to vibrationally relax within $< 0.5 \text{ s}$ [7] thanks to their isotopic asymmetry, while radiative transitions along the rotational band built on the vibrational ground state are calculated to be considerably slower [8] such that the SEC process can be expected to dominate the rotational cooling process. Moreover, the small moment of inertia of HD^+ leads to a sizable spacing between the rotational states, which facilitates the determination of the rotational excitation of the ion by fragment imaging [9, 10]. And last but not least, the comparatively simple structure of HD^+ favors a theoretical description of the $\text{HD}^+ + e^-$ collision system.

2. Experimental Procedure

The experiment uses the DR process to probe the time evolution of the population $P_J(t)$ of the ro-vibrational states ($v = 0, J$) of HD^+ when merging a hot HD^+ beam circulating in a storage ring with cold electrons of the same mean velocity as the ions. As the relative ion-electron energies are smaller than the smallest difference between two ro-vibrational states, and the ions are vibrationally relaxed in less than 0.5 s, the only inelastic collisions which can take place thereafter are superelastic collisions between the rotational states, i.e.



with $J' < J$. The resulting time evolution of $P_J(t)$ is monitored by observing the kinetic energy release $E_k(J)$ carried by the two DR fragments applying fragment imaging [9]. Since the only open DR channel is



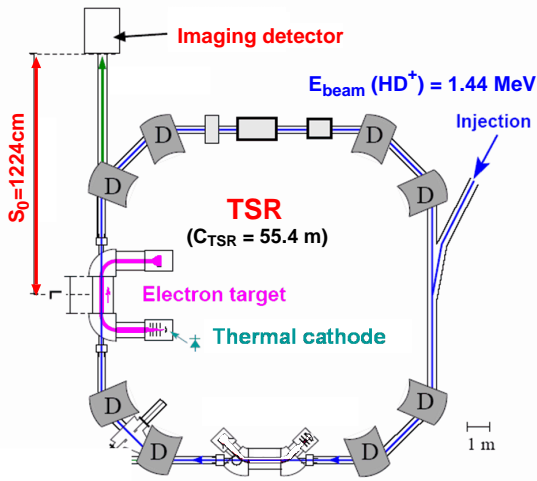


Figure 1. Schematic view of the experimental set-up. HD^+ ions were produced in a discharge ion source, accelerated to 1.44 MeV, injected into the storage ring for $30 \mu\text{s}$, and stored for times $\geq 10 \text{ s}$. The ions are merged with a velocity matched electron beam supplied by the electron target station [12]. DR fragments are observed using a high resolution imaging system based on a microchannel plate, which is attached to a phosphor screen viewed by a CCD camera [10].

with $(n, m) = (1, 2)$ or $(2, 1)$, $E_k(J)$ is given by $E_k(J) = E_0 + BJ(J + 1)$, where $E_0 = 0.726 \text{ eV}$ denotes the energy difference between the ionic ground state and dissociation channel and $B = 2.72 \text{ meV}$ is the rotational constant of HD^+ [11]; thus $E_k(J)$ is a unique function of J .

A schematic view of the experimental set-up at the heavy ion storage ring TSR operated by the Max Planck Institute for Nuclear Physics in Heidelberg is shown in Fig. 1. The electrons were produced by a thermal cathode, accelerated and expanded to form a beam of 10 mm diameter; the resulting electron energy distribution in the comoving reference frame can be described by a flattened Maxwell distribution with $kT_{\parallel} \approx 45 \mu\text{eV}$ parallel and $kT_{\perp} \approx 2.8 \text{ meV}$ transversal to the beam direction. The effective length of the electron target was $L = 1.22 \text{ m}$, which leads to an overlap factor of $\eta = L/C = 0.022$ when compared to the circumference of the TSR of $C = 55.4 \text{ m}$. The measurements were performed at three different electron densities $n_e = (0.28, 1.00, \text{ and } 1.45) \cdot 10^7 \text{ cm}^{-3}$, which we will refer to as "low", "medium", and "high", respectively. Within the first second after injection the HD^+ beam was phase space cooled by the electrons, which led to an ion beam of $< 2 \text{ mm}$ diameter and $< 25 \mu\text{rad}$ divergence.

The two neutral fragments following the DR of HD^+ were recorded as a function of storage time t by the high resolution imaging system mounted 12.24 m downstream from the center of the electron target, which allowed to determine the projected (transverse) distance D between the two fragments with an accuracy up to $\pm 0.3\%$. A large-area energy sensitive Si detector could be temporarily placed about 1 m in front of the imaging detector to measure also the total DR rate as a function of t .

3. Analysis and Results

At matched electron-ion beam velocities the normalized projected distance distribution $f_J(D)$ resulting from the DR of a given rotational state J is only determined by the detection geometry and the kinetic energy release $E_k(J)$, the maximum projected distance being proportional to $(E_k(J))^{1/2}$ [9]. For an ensemble of rotationally excited HD^+ ions the normalized projected distance distributions $F(D, t)$ are thus given by

$$F(D, t) = (\alpha_{DR}(t))^{-1} \sum_J \alpha_J P_J(t) f_J(D), \quad (3)$$

where α_J denotes the DR rate coefficient of the rotational state J , $P_J(t)$ its population probability, and $\alpha_{DR}(t)$ the effective DR rate coefficient at time t .

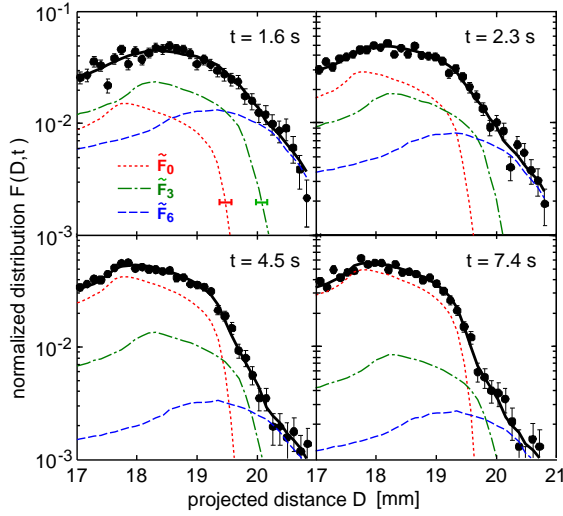


Figure 2. Projected distance distributions $F(D,t)$ recorded for distances $D \geq 17$ mm and several times t after injection. The electron density was $n_e = 1.0 \cdot 10^7 \text{ cm}^{-3}$. The broken lines are the calculated $\tilde{F}_\mu(D,t)$ distributions reflecting the contribution of three groups of rotational states ($\mu=0$ ($J=0,1,2$), $\mu=3$ ($J=3,4$), $\mu=6$ ($J=5,6,7$)), while the black solid line represents the sum of these distributions after adjusting their relative heights to the data. The horizontal error bars in the upper left panel reflect the calibration errors caused by uncertainties of the length of the merged beam region and the calibration of the imaging system.

Examples of normalized projected distance distributions (PDD) measured at “medium” electron density are shown in Fig. 2 for $D \geq 17$ mm, where these distributions are most sensitive to the rotational states populated. Because of counting statistics and the broad widths of the $f_J(D)$ distributions it was not possible to reliably deduce the individual amplitudes of $f_J(D)$ from the measured PDDs. Instead, we combined adjacent J states into three groups, denoting the groups ($J = 0, 1, 2$) by $\mu = 0$, ($J = 3, 4$) by $\mu = 3$, and ($J = 5, 6, 7$) by $\mu = 6$. Higher rotational states do not seem to contribute significantly to the DR rate at storage times larger than 2 s. Denoting the (unweighted) averages of the distribution functions $f_J(D)$ with $J \in \{\mu\}$ by $\tilde{f}_\mu(D)$, and the (unweighted) averages of the DR rate coefficients α_J with $J \in \{\mu\}$ by $\tilde{\alpha}_\mu$, we can approximate eq. 3 by

$$F(D,t) = \sum_{\mu} \tilde{F}_\mu(D,t) = \sum_{\mu} r_\mu(t) \tilde{f}_\mu(D), \quad (4)$$

$$\text{with } r_\mu(t) = \frac{\tilde{\alpha}_\mu}{\alpha_{DR}(t)} \hat{P}_\mu(t) \quad \text{and} \quad \hat{P}_\mu(t) = \sum_{J \in \{\mu\}} P_J(t).$$

In a first analysis step we fitted Eq. 4 to the measured PDDs in the range $17 \text{ mm} \leq D \leq 21 \text{ mm}$ to determine the relative amplitudes $r_\mu(t)$. The measured distributions are well represented by these fits (see Fig. 2). The resulting amplitudes for the measurement performed at “medium” electron density are plotted in Fig. 3a. In a second step we used the relation

$$\alpha_{DR}(t) = \left(\sum_{\mu} r_\mu(t) / \tilde{\alpha}_\mu \right)^{-1}, \quad (5)$$

which follows from the definition of $r_\mu(t)$ and the normalization $\sum_{\mu} \hat{P}_\mu(t) = 1$, to determine the averaged DR rate coefficients $\tilde{\alpha}_\mu$ for the three groups of rotational angular momentum states from the total DR rate coefficient $\alpha_{DR}(t)$ measured at “medium” electron density (see Fig. 3b). Using the relative amplitudes $r_\mu(t)$ measured at this electron density we find

$$\tilde{\alpha}_0 = 3.8 \pm 0.1, \tilde{\alpha}_3 = 4.0 \pm 0.2, \text{ and } \tilde{\alpha}_6 = 9.0 \pm 1.3$$

in units of $10^{-8} \text{ cm}^3/\text{s}$. Only relative errors are given, the absolute scale is expected to be accurate to within $\pm 20\%$.

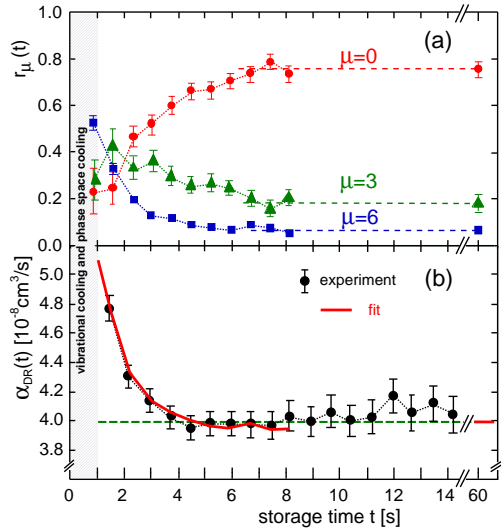


Figure 3. (a) Relative amplitudes $r_\mu(t)$ deduced from the measured projected distance distributions $F(D, t)$ and (b) the measured total DR rate coefficient $\alpha_{DR}(t)$ as a function of the storage time t . The data were taken at an electron density of $n_e = 1.0 \cdot 10^7 \text{ cm}^{-3}$. The absolute scale of the DR rate coefficient was normalized to the steady state value of $4.0(8) \cdot 10^{-8} \text{ cm}^3/\text{s}$ (dashed line), estimated for the present electron temperatures using the data collected in ref. [13]. The solid line in (b) connects the $\alpha_{DR}(t)$ calculated using Eq. 5 and adjusting the average DR rate coefficients $\tilde{\alpha}_\mu$.

In a final analysis step the $\tilde{\alpha}_\mu$ values were used to determine $\hat{P}_\mu(t)$ from the amplitudes $r_\mu(t)$. The resulting group populations are shown in Fig. 4 for the three electron densities employed. The data points in Fig. 4 clearly reveal that electron assisted rotational cooling of the HD⁺ ions takes place, the cooling getting faster with increasing density n_e of the electron beam. At times shorter than about 2 s the separation between $\tilde{F}_0(D)$ and $\tilde{F}_3(D)$ was perturbed by the strong contribution of $\tilde{F}_6(D)$, which is partly covering up the high distance tails of $\tilde{F}_0(D)$ and $\tilde{F}_3(D)$ most sensitive to J (see Fig. 2).

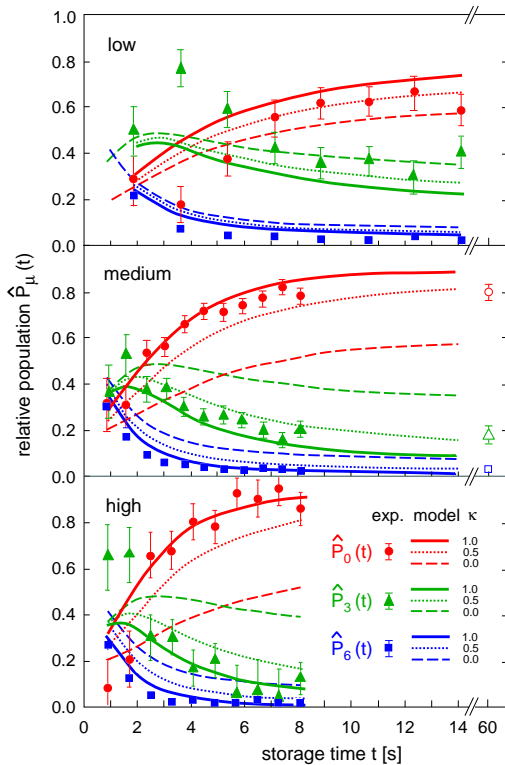


Figure 4. Relative group populations as a function of storage time t , which were extracted from the projected distance distributions measured at three different electron densities n_e of $0.28 \cdot 10^7 \text{ cm}^{-3}$ (low), $1.0 \cdot 10^7 \text{ cm}^{-3}$ (medium) and $1.45 \cdot 10^7 \text{ cm}^{-3}$ (high). The lines are the calculated group populations obtained by solving Eq. 6 with SEC rate coefficients given by $c_{J,J'} = \kappa \cdot c_{J,J'}^{ANR}$, where $c_{J,J'}^{ANR}$ are the theoretical SEC rate coefficient calculated within the R-matrix+ANR approach (see Fig. 5b), and κ is a constant factor. The solid (dashed) lines obtained for $\kappa = 1$ ($\kappa = 0$) are thus reflecting the cooling curves including (neglecting) the SEC process, while the dotted lines are the resulting curves assuming $\kappa = 0.5$.

To investigate the role of SEC in the rotational cooling process, we calculated the time evolution of the rotational populations $P_J(t)$ of the vibrational ground state by solving the

coupled set of differential equations

$$\dot{P}_J(t) = \sum_{J'} [M_{JJ'}^r + \eta n_e M_{JJ'}^c] P_{J'}(t). \quad (6)$$

Here the matrix M^r is due to the exchange of photons with the ambient radiation field; the matrix elements $M_{JJ'}^r$ are the Einstein coefficients, which can be readily taken from literature [8] assuming the radiation field to be given by a Planck distribution with a temperature of 300 K. The matrix M^c describes the population change due to interactions with electrons; for $J > J'$ the matrix elements $M_{JJ'}^c$ are given by the SEC rate coefficients $c_{J,J'}$ leading from state J to state J' , while for $J = J'$ the matrix elements are $-(\alpha_J + \sum_{J' < J} c_{J,J'})$. The initial rotational populations $P_J(t=0)$ are assumed to be given by a Boltzmann distribution for an initial temperature T_i .

We first calculated the time evolution of $P_J(t)$ assuming that only radiative transitions and cooling effects through J -dependent level depletion by DR are taking place. The DR rate coefficients α_J were set equal to the $\tilde{\alpha}_\mu$ for $J \in \{\mu\}$, and equal to $\tilde{\alpha}_6$ for $J \geq 8$, and the initial temperature was assumed to be $T_i = 1500$ K. The resulting group populations $\hat{P}_\mu(t)$ are shown in Fig. 4 by the dashed lines ($\kappa = 0$) and are clearly falling short in explaining the observed time evolution of $\hat{P}_\mu(t)$ and their dependence on the electron density n_e . In fact, the calculated $\hat{P}_\mu(t)$ are essentially independent of the electron density within the range of densities investigated, that means that radiative processes are dominating and contributions caused by state selective depletions via the DR process are small. The calculation also showed that the assumptions made for the initial level population are being washed out quite fast with increasing storage time; changes of T_i by ± 300 K affect the $\hat{P}_\mu(t)$ values at $t = 1$ s by $\pm 15\%$ and by less than 2% at $t = 7.5$ s. Thus neither rotational cooling by DR induced depletion nor radiative transitions alone can reproduce the observed group populations.

Superelastic collisions were included into the rate equations using inelastic electron collision cross sections calculated by combining molecular R-matrix wave functions with the adiabatic nuclei rotation (ANR) approximation. These calculations followed those performed on H_2^+ [14] for a fixed bond length of $2.0 a_0$ except that they were carried out in $C_{\infty v}$ symmetry with a shifted center of mass, which results in a dipole moment of $0.85 D$. As the ANR approach assumes that the rotational states are energetically degenerate, that is that the asymptotic kinetic electron energies E_k before and after an inelastic scattering event are the same, care has to be taken in using this approach when the electron energies are comparable with

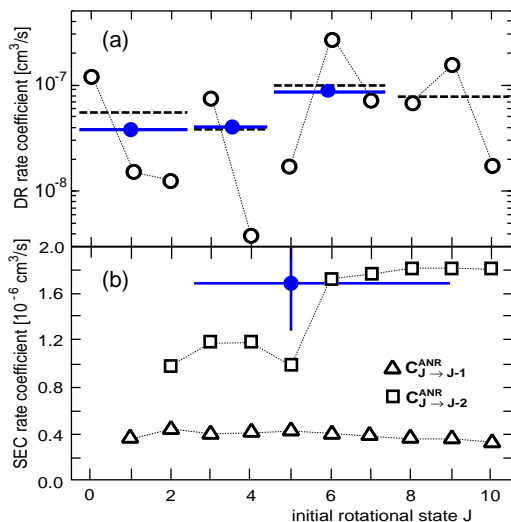


Figure 5. (a) Average experimental DR rate coefficients deduced for the three groups of J states built on the vibrational ground state of HD^+ (filled circles). They are compared to theoretical values (dashed lines) obtained by averaging the individual DR coefficients calculated within the MQDT approach (open circles). (b) SEC rate coefficients $c_{J,J'}$ calculated within the R-matrix+ANR approach and used to calculate the cooling curves shown by the solid lines in Fig. 4. The filled circle is the c_{-2} coefficient resulting from a fit of the experimental cooling curves assuming $c_{J,J-1} = 0$ and $c_{J,J-2} = c_{-2}$.

the rotational transition energies. It has been shown recently [15], however, that in view of the approximate E_k^{-1} dependence of the ANR cross sections for small kinetic energies, i.e. $\sigma_{i \rightarrow f}(E_{k,i}) \propto 1/E_{k,i}$, not only the expected threshold behavior for rotational excitation from E_{J_i} to E_{J_f} by inelastic electron scattering is secured by limiting the validity range of $\sigma_{i \rightarrow f}(E_{k,i})$ to kinetic energies $E_{k,i} > (E_{J_f} - E_{J_i})$, but that also the detailed balance relation $\sigma_{f \rightarrow i}(E_{k,f})(2J_f + 1)E_{k,f} = \sigma_{i \rightarrow f}(E_{k,i})(2J_i + 1)E_{k,i}$ is approximately fulfilled. Moreover, a quantitative assessment of the validity of the ANR approach has been performed in Ref. [15] for electron-impact rotational excitations of H_3^+ by comparing the ANR results with those obtained within the MQDT and rotational-frame-transformation approach.

The SEC cross sections calculated for HD^+ within the R-matrix+ANR approach show the expected $1/E_k$ dependence for all relevant electron energies, and – as already observed previously for ions with a moderate dipole moment [16] – the $\Delta J = -1$ cross sections are significantly smaller than those for $\Delta J = -2$. The SEC rate coefficients $c_{J,J+\Delta J}^{\text{ANR}}$ obtained by integrating the cross sections over the flattened electron distribution are shown in Fig. 5b for $\Delta J = -1, -2$ transitions; the SEC coefficients for higher transitions were found to be several orders of magnitude smaller. The resulting time evolution of $\hat{P}_\mu(t)$ obtained when taking the SEC coefficients for $\Delta J = -1, -2$ into account in the rate equation is shown by the thick lines ($\kappa = 1$) in Fig. 4. The agreement with the data is remarkable, in particular when taking into account that the only free parameter is the initial temperature T_i (assumed to be $T_i = 1500$ K), a parameter which is only moderately influencing the result within the first few seconds.

To estimate the sensitivity of the present experiment to the SEC rate coefficients several tests have been performed. The group populations expected when assuming $c_{J,J'} = \kappa \cdot c_{J,J'}^{\text{ANR}}$ with $\kappa = 0.5$ are depicted in Fig. 4 by the dotted lines; they clearly underestimate the observed cooling. The contribution of the $\Delta J = -1$ coefficients to the cooling is found to be small; the effect of setting, e.g., all $c_{J,J-1}$ coefficients equal to zero can be compensated by increasing κ by about 15%. Moreover, a fit of the cooling curves assuming $c_{J,J-1} = 0$ and $c_{J,J-2} = c_{-2}$ results in $c_{-2} = 1.7(4) \cdot 10^{-6} \text{cm}^3 \text{s}^{-1}$, which agrees well with the average value of the calculated $c_{J,J-2}$ coefficients (see Fig. 5b), in particular when taking into account the expected increase caused by setting $c_{J,J-1} = 0$. The influence of heating processes on the cooling curves other than those induced by radiation were found to be negligibly small. Neither those caused by inelastic electron scattering in the bending section of the electron beam, where kinetic energies of up to 1.5 eV are available, nor those induced in the straight section by the tail of the Maxwellian energy distribution were found to change the group populations by more than a few percent. We conclude from these tests that our experiment verifies the dominant $\Delta J = -2$ SEC rate coefficients calculated within the R-matrix+ANR approach to within about 30%.

4. Summary and Conclusion

The average DR rate coefficients $\tilde{\alpha}_\mu$ deduced for the three rotational angular momentum groups μ are plotted in Fig. 5a together with the individual DR rate coefficients α_J calculated within the MQDT approach. These calculations were performed along the lines discussed in Refs. [17, 4], taking detailed account of the rotational structure and rotational couplings, and result in strongly J dependent DR rate coefficients. The values obtained by simply averaging the calculated α_J for $J \in \{\mu\}$ are consistent with the measured values, in particular when taking into account the $\pm 20\%$ uncertainty connected with the absolute scale of the measured values. Although the J dependent variations are strongly reduced by the averaging procedure, the observed increase of more than a factor of 2 between $\tilde{\alpha}_3$ and $\tilde{\alpha}_6$ constitutes another experimental verification of the strong J dependence of the DR cross sections for molecular hydrogen molecules [18, 10].

More importantly, the present experiment allows for the first time to obtain a quantitative estimate of rotational SEC rate coefficients. The observed cooling of the rotational degree of freedom induced by bathing rotationally hot HD^+ ions in cold electrons of $T \approx 10$ K was

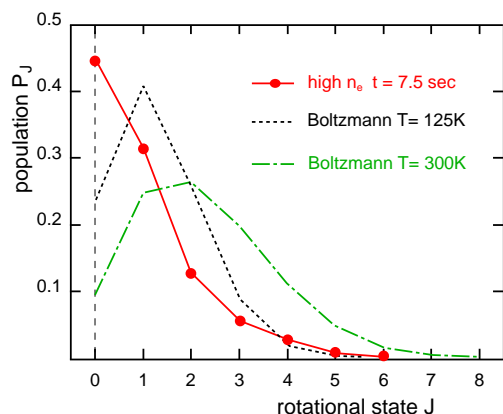


Figure 6. Population of the ($v = 0, J$) states in HD^+ reached after 7.5 s of electron cooling at an electron density of $n_e = 1.45 \cdot 10^7 \text{ cm}^{-3}$ (filled circles). For comparison, Boltzmann distributions expected for rotational temperatures of 300 K (dashed dotted) and 125 K (dotted) are shown.

found to be well described by theoretical rate coefficients calculated by combining the R-matrix approach with the ANR approximation (see Fig. 5b); in particular, we could verify the size of the dominant $\Delta J = -2$ coefficients to within about 30%. These rate coefficients are of the order of $(1 - 2) \cdot 10^{-6} \text{ cm}^3\text{s}^{-1}$ and are thus of the same size as the vibrational SEC rates observed for $\Delta v = -1$ transitions in H_2^+ [2, 3]. Superelastic collisions can thus be used to speed up the rotational cooling of molecular ions in storage ring experiments and to realize even subthermal distributions. This is demonstrated by Fig. 6, which displays the population of the individual ($v = 0, J$) states of HD^+ reached after cooling for 7.5 s at the highest electron density investigated, which was calculated assuming the theoretical SEC rate coefficients shown in Fig. 5b. Although the resulting population cannot be represented by a Boltzmann distribution, a comparison of the centroids shows that the SEC controlled distribution corresponds to a rotational temperature of $\approx 125 \text{ K}$, which is well below the radiative equilibrium of 300 K; without electron cooling, moreover, the latter would only be reached after $\approx 30 \text{ s}$ of storage.

Acknowledgments

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