Electron collisions with $^4\text{He}_2^+$ at the TSR

H Buhr$^{1,2}$, H B Pedersen$^1$, S Altevogt$^1$, V M Andrianarijaona$^1$, H Kreckel$^1$, L Lammich$^1$, S Novotny$^1$, D Strasser$^2$, J Hoffmann$^1$, M Lange$^1$, M Lestinsky$^1$, M B Mendes$^1$, M Motsch$^1$, O Novotny$^1$, D Schwalm$^{1,2}$, X Urbain$^3$, D Zajfman$^{2,3}$ and A Wolf$^1$

$^1$ Max-Planck-Institut für Kernphysik, Heidelberg, Germany
$^2$ Department of Particle Physics, Weizmann Institute of Science, Rehovot, Israel
$^3$ Département de Physique, Université catholique de Louvain, Louvain-la-Neuve, Belgium

E-mail: henrik.buhr@mpi-hd.mpg.de

Abstract. The dissociative recombination (DR) of $^4\text{He}_2^+$ has been investigated at the heavy-ion storage ring TSR in Heidelberg. Rate coefficients were measured up to collision energies of 40 eV. Vibrational level populations were monitored using the Coulomb explosion imaging technique showing relaxation to the $v = 0$ level (>95%) through collisions with cold electrons within 50 s. Low-energy DR rate coefficients are derived for $v = 0$, 1 and $\geq 2$ which show a strong $v$-dependence. A low-energy super-elastic collision (SEC) rate coefficient of $\alpha_{v=1\rightarrow 0,SEC}(E_d = 0) \approx 1.8 \times 10^{-7}$ cm$^3$ s$^{-1}$ was found.

1. Introduction
The dissociative recombination (DR) of $\text{He}_2^+$ has raised the interest of experimental and theoretical physicists over a long time. The molecular ion potential curve has a curve crossing with a neutral dissociative state only at relatively large internuclear distances favourable for the DR of vibrational levels $v=3$ and higher. This should result in a very low DR rate coefficient for ground state $\text{He}_2^+$ at low electron energies. This was predicted by theory for both $^3\text{He}$ $^4\text{He}^+$ and $^4\text{He}_2^+$ [1] and already confirmed by experiment for $^3\text{He}$ $^4\text{He}^+$ [2, 3]. An energy dependent DR rate coefficient for $^4\text{He}_2^+$ was calculated in the low [1] and high (up to 15 eV) energy range [4] but only measured for vibrationally hot $^4\text{He}_2^+$ [3]. Here we report on experimental results obtained for vibrationally hot and cold $^4\text{He}_2^+$ at the TSR heavy-ion storage ring in Heidelberg [5]. The absence of a dipole moment eliminates radiation as a process that changes the vibrational distribution of $^4\text{He}_2^+$, so that in contrast to the case of $^3\text{He}$ $^4\text{He}^+$, where vibrational relaxation to the ground state (>98.9 %) occurs in ~3 s [2], the effects of collisions with low-energy electrons and residual gas molecules can be studied undisturbed and over considerably longer times. Time and energy dependent DR rate coefficients under collisions with cold electrons and residual gas are presented and the changes of vibrational level populations are discussed. In addition, vibrational level dependent DR rate coefficients are derived for $v = 0$, 1 and $\geq 2$. Further details are reported in a full publication [6].

2. Experiment
The experiment was performed at the TSR at the MPI für Kernphysik in Heidelberg. $\text{He}_2^+$ was produced in a standard duoplasmatron source, accelerated to $E_i = 3.83$ or 8.32 MeV by an
rf accelerator and injected into the storage ring, where the ion beam was stored for periods of up to 100 s. The electron cooler [7] provided velocity matched electrons for phase space cooling during the first 5 s and electrons as collision partners at variable detuning velocities \( v_d \) compared to the ion beam for the collision experiments. The detuning energy is given by \( E_d = m_e v_d^2 / 2 \) (\( m_e \) is the electron mass) and describes well the average collision energy for \( E_d > kT_\perp = 11.5 \) meV, where \( T_\perp \) is the transversal electron beam temperature. During DR rate coefficient measurements [8], \( E_d \) was changed periodically following a cycle of cooling (\( E_d^c \)), measurement (\( E_d^m \)) and reference for normalization (\( E_d^r \)) with dwell times of 25–125 ms. For \( E_i = 3.83 \) MeV the electron beam density at cooling energy was \( n_e(E_d^c) = 5.5 \times 10^6 \) cm\(^{-3} \) and the reference energy \( E_d^r = 7.3 \) eV (Type A), for \( E_i = 8.32 \) MeV it amounted to \( n_e(E_d^c) = 1.1 \times 10^7 \) cm\(^{-3} \) and \( E_d^r = 23.5 \) eV (Type B). Absolute DR rate coefficients were measured similarly to the way described in [2], finding \( \alpha_{v,DR}(E_d = 7.4 \) eV)\( = 2.0(4) \times 10^{-8} \) cm\(^3\) s\(^{-1}\). Using the Coulomb explosion imaging (CEI) technique [9] the vibrational population distribution was measured for several periods of electron cooling between 5 and 50 s.

3. Results

3.1. Time dependence of the DR rate coefficient

The time dependence of the DR rate coefficient \( \alpha_{DR}(E_d) \) was measured for both Type A and B parameter sets (figure 1(a)). The rate coefficient decreases strongly especially in the beginning.

![Figure 1](image-url)  

Figure 1. Measured DR rate coefficient and population distribution of vibrational levels in \( ^4\)He\(^2+ \) as a function of time. (a) Measured DR rate coefficient at \( E_d \) as function of time for the two parameter sets and a dwell time ratio of \( t_c : t_m : t_r = 5 : 0 : 1 \). (b) Measured population distribution of \( ^4\)He\(^2+ \) with permanent electron cooling (markers, Type B) as function of time compared to model calculations using the extracted values \( \alpha_{v,DR}(E_d^c) \) including (full line) and neglecting (dotted) super elastic collisions.

This effect is much stronger for the high electron density (Type B) and after 80 s a rate coefficient lower by a factor of 2 than in the Type A experiment is reached. This indicates a strong change in the vibrational population due to electron-\(^4\)He\(^2+ \) collisions, since the observed DR rate coefficient \( \alpha_{DR}(E_d) \) is given by the weighted average of the vibrational-level specific DR rate coefficients \( \alpha_{v,DR}(E_d^c) \)

\[
\alpha_{DR}(E_d) = \sum_v p_v \alpha_{v,DR}(E_d^c)
\]

where \( p_v \) is the relative population of the vibrational level \( v \), assuming the influence of rotational excitations on the \( \alpha_{v,DR} \) to be negligible. A direct measurement of the relative populations using
the CEI technique at different times between 5 and 50 s and constant electron cooling shows that in the beginning there are 40–45% of the $^{4}\text{He}^+_2$ ions in both the $v=0$ as well as in the $v=1$ state and $\sim$15% in all other states (figure 1(b)) while $\sim$95% are in the ground state after 50 s. A least-squares-fit combining the results for $p_\alpha(t_{\text{CEI}})$ obtained at 8 different times $t_{\text{CEI}}$ and $\alpha_{dR}(E_d^v)(t_{dR})$ in equation 1 with $\alpha_{dR}^v(E_d^v)$ as fitting parameters for $v=0$, 1 and $\geq 2$ yields the results given in table 1. The times $t_{dR}$ were chosen in a way that the total time spent at $E_d^v$ matched with $t_{\text{CEI}}$, i.e., the time spent at electron cooling was equal in CEI and DR. Based on these values for $\alpha_{dR}^v(E_d^v)$ a simple model for the evolution of the relative vibrational populations $p_v$ in the stored $^{4}\text{He}^+_2$ beam was applied, starting from the distribution measured after 5 s and taking into account only ion loss due to the DR reaction while the losses due to collisions with residual gas are assumed to be $v$-independent. The $p_v$ resulting from this model are plotted as a function of time in figure 1(b) for the best values given in table 1 (dotted); they clearly do not agree with the measured time dependence. By including a super-elastic collision process assumed to change the vibrational level $v$ from $\geq 2$ to 1 and from 1 to 0 with a rate coefficient $\alpha_{\text{SEC}}^v$ of the stored ions were excited to higher vibrational levels with very high $\alpha_{\text{SEC}}^v$ and residual gas density leading to vibrational excitation of the ions.

It was also tested, whether collisions with residual gas might explain the observed time dependence. For this, after 22.5 s of electron cooling, the electron beam was switched off for a certain time and the $^{4}\text{He}^+_2$ was only subject to collisions with the residual gas. Examining these ions with CEI revealed that only marginal changes in the vibrational level population occurred towards higher excitation, while DR rate measurements after increasing non-cooling times showed a strongly increasing DR rate coefficient, indicating that very small fractions of the stored ions were excited to higher vibrational levels with very high $\alpha_{dR}^v(E_d^v)$. So the vibrational relaxation to $v=0$ as displayed in figure 1(b) is the result of super-elastic electron collisions at very low energies.

No similar time dependence of $\alpha_{dR}(E_d)$ was observed in an earlier experiment on $^{4}\text{He}^+_2$ [3], probably because SEC was suppressed due to lower electron density and higher electron beam temperature as well as due to higher residual gas density leading to vibrational excitation of the ions by collisions.

### 3.2. Energy dependence of DR rate coefficient as function of time

The strong time dependence of the vibrational populations $p_v$ under electron cooling and hence of $\alpha_{dR}(E_d^v)$ implies also a strong effect on the energy dependent DR rate coefficient. This can be seen in figure 2 which compares $\alpha_{dR}(E_d)$ for different vibrational population distributions reached by different times and intensities of electron cooling. One finds that for $E_d<7$ eV $\alpha_{dR}(E_d)$ is decreasing with increasing electron cooling, where the effect is strongest for small $E_d$. At higher energies $\alpha_{dR}(E_d)$ does not change. Comparison with the measured $p_v$ (figure 1(b)) suggests that the rate coefficient for Type B ($\geq 58.5$ s) is dominated by the vibrational ground state ($p_0>95$%).

This vibrationally cold DR rate coefficient shows a broad structure between 2 and 12 eV with the peak at $\sim 7.5$ eV, which is also present, but less prominent in the measurements of Type A. Between 20 and 35 eV another structure with 3 peaks is found. Similar structures were observed in the DR of $^3\text{He}^4\text{He}^+_2$ [2]. The structure around 7.5 eV is also present in the calculated DR rate coefficient for the vibrational ground state [4] with the peak at 7.1 eV. However, the calculated height is considerably smaller than the one measured here, namely $0.91 \times 10^{-8}$ cm$^3$ s$^{-1}$.

### Table 1. Vibrational-level specific low-energy DR rate coefficients $\alpha_{dR}(E_d^v)$ derived for $v=0$, 1 and $\geq 2$.

| vibrational level $v$ | 0          | 1          | $\geq 2$   |
|-----------------------|------------|------------|------------|
| $\alpha_{dR}^v(E_d^v)$ ($10^{-9}$ cm$^3$ s$^{-1}$) | 3.4(2.0)   | 3.2$^{+17.8}_{-3.2}$ | 202$^{-153}_{+37}$ |
Figure 2. Measured DR rate coefficient of $^4\text{He}_2^+$ as function of detuning energy in the range of $E_d=10^{-4}$–$40\text{ eV}$ for different total electron cooling: Type A parameters and storage times of 7–14 s and 33.75–58 s and Type B parameters and storage times $>58.5$ s. The electron beam temperature was $kT_{\perp} = 11.5\text{ meV}$.

vs. $2.0(4)\times10^{-8}\text{cm}^3\text{s}^{-1}$. Support for both values comes from previous measurements of the absolute DR rate coefficient of $^3\text{He}^4\text{He}^+$ [2, 3]. The values of the resonant dissociative excitation rate coefficient from calculation and the present experiment agree well [4].

4. Conclusions

Using results of CEI and DR rate coefficient measurements, we deduced vibrational level dependent DR rate coefficients, which increase by 2 orders of magnitude when going from $v=0\ (3.4(2.0)\times10^{-9}\text{cm}^3\text{s}^{-1})$ to $v\geq2\ (202^{+153}_{-62}\times10^{-9}\text{cm}^3\text{s}^{-1})$. Similar increases were found in theoretical [1] and previous experimental [2] works on $^3\text{He}^4\text{He}^+$. The strong decrease of $\alpha_{DR}(E_d)$ as a function of time reflects the vibrational cooling of the stored ions due to SEC with a rate coefficient of $\alpha_{SEC}(E_d)=1.8\times10^{-7}\text{cm}^3\text{s}^{-1}$.

Acknowledgments

This work has been funded in part by the German-Israeli Foundation for Scientific Research (GIF) under contract I-707-55.7/2001. H. B. P. acknowledges support from the European Community program IHP under contract No. HPMF-CT-2002-01833. X. U. is Senior Research Associate of the F.R.S.-FNRS.

References

[1] Carata L, Orel A E and Suzor-Weiner A 1999 Phys. Rev. A 59 2804
[2] Pedersen H B et al 2005 Phys. Rev. A 72 012712
[3] Urbain X et al 2005 J. Phys. B 38 43
[4] Royal J and Orel A E 2007 Phys. Rev. A 75 052706
[5] Habs D et al 1989 Nucl. Instrum. Methods Phys. Res. B 43 390
[6] Buhr H et al 2008 Phys. Rev. A 77 032719
[7] Kilgus G, Habs D, Schwalm D, Wolf A, Badnell N R and Müller A 1992 Phys. Rev. A 5730
[8] Al-Khalili A et al 2003 Phys. Rev. A 68 042702
[9] Wester R et al 1998 Nucl. Instrum. Methods Phys. Res. A 413 379