Publ. Astron. Obs. Belgrade No. 84 (2008), 39 - 44 *Contributed Paper*

RECOMBINATION OF D_3^+ IONS WITH ELECTRONS IN LOW TEMPERATURE PLASMA

I. KOROLOV[1,2], T. KOTRÍK[2], P. DOHNAL[2], R. PLAŠIL[2] and J. GLOSÍK[2]

[1]*Research Institute for Solid State Physics and Optics, Konkoly-Thege M. 29-33, Budapest, Hungary*
E–mail: Ihor.Korolov@mff.cuni.cz

[2]*Charles University Prague, Faculty of Mathematics and Physics, V Holešovičkách 2, Prague 8, Czech Republic*
E–mail: Juraj.Glosik@mff.cuni.cz

Abstract. The flowing afterglow study of recombination of D_3^+ ions with electrons and measurements of recombination rate coefficients in He-Ar-D_2 plasma at several temperatures and buffer gas pressures, and over a wide range of deuterium number densities are reported. The influence of helium number density on overall recombination rate coefficient was observed. The results indicate the multicollision character of the recombination process in D_3^+ dominated low temperature plasma. The obtained binary and ternary recombination rate coefficients are: $\alpha_{\rm bin}$(250 K)= $(4.1 \pm 1.2) \times 10^{-8}$ cm^{3}s^{-1}, $K_{\rm He}$(250K) = $(2.1 \pm 0.7) \times 10^{-25}$ cm^{6}s^{-1}.

1. INTRODUCTION

Interactions of electrons with H_3^+ and D_3^+ ions is of fundamental importance for plasma physics, physical chemistry, astrophysics and for quantum theory. From first study of H_3^+ recombination in 1949 to present days there was over 50 attempts to measure recombination rate coefficients of H_3^+ and D_3^+ ions (see reviews Johnsen et al. 2005, Plasil et al. 2002, Larson et al. 2008). Up to 2001 the measured values of the recombination rate coefficients of H_3^+ and D_3^+ ions were scattered over several orders in magnitude and they were in contradiction with available theoretical predictions. In 2001 inclusion of non-Born-Oppenheimer Jahn-Teller coupling significantly improved the theory (Kokoouline et al. 2001). After further refinement the cross sections and the rate coefficients for H_3^+ and D_3^+ recombination were calculated (Kokoouline et al., 2003). The most recent theoretical values of the recombination rate coefficients for dissociative recombination of H_3^+ and D_3^+ ions are $\alpha_{H_3^+}(300\ \mathrm{K}) = 5.6 \times 10^{-8}$ cm^{3}s^{-1} and $\alpha_{D_3^+}(300\ \mathrm{K}) = 4 \times 10^{-8}$ cm^{3}s^{-1} respectively (Kokoouline et al., 2003, Santos et al. 2007, Kokoouline 2008). These values are in good agreement with the recent storage ring experiments (McCall et al. 2003, 2004, Kreckel et al. 2005). But the disagreement between multi- and single-collisional experiments (Plasil et al. 2002, Poterya et al. 2002) has not been solved until now. Note that in the plasmatic (mul-

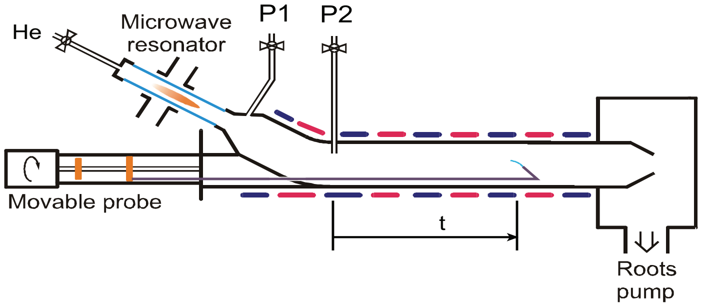

Figure 1: Schema of temperature variable Flowing Afterglow with Langmuir probe (FALP).

ticollisional) experiments plasma is usually formed in the ambient gas (He in present experiments) and small amount of hydrogen or deuterium is added to form H_3^+ or D_3^+ dominated plasma. Moreover, in plasmatic experiments in order to minimize diffusion losses the ambient gas pressure is high (100-2000 Pa) and the ions have multiple collisions with neutral particles prior to their recombination. In beam experiments there is single collision of electron with ion and ion do not collide with neutral particles. If in electron-ion interaction long-lived intermediate neutral molecule is formed, as it is in case of H_3^+ or D_3^+ ions, then we can also expect collisions of this molecule with the atoms of ambient gas. These collisions can influence the process of recombination and as a result measured overall recombination rate coefficient $\alpha_{\rm eff}$ will be dependent on the pressure. The presented studies concentrated on recombination of D_3^+ and on measurements of the pressure dependence of $\alpha_{{\rm eff}D_3^+}$.

2. EXPERIMENT

The recombination rate coefficients were measured with flowing afterglow apparatus (FALP technique) (Novotny et al. 2006). The present version of the experimental apparatus is designed and constructed for measurement of small recombination rate coefficients (down to 5×10^{-9} cm^{3}s^{-1}) at relatively high buffer gas pressure (up to 2000 Pa), the details are described elsewhere (Korolov et al. 2006, Plasil et al. 2008). Schematic diagram of FALP is shown in Figure 1.

Helium as a buffer gas flows through the glass discharge tube where He^+ ions and He^m metastables are formed. Downstream from the discharge region argon gas is added (port P1, see Figure 1) to quench helium metastables and to form Ar^+ dominated plasma. Further downstream hydrogen (or deuterium) is added via port P2 to already relaxed plasma (Glosik et al. 1999) and D_3^+ dominated plasma is formed via the reaction sequence:

$$Ar^+ \xrightarrow{D_2} (ArD^+, D_2^+) \xrightarrow{D_2} D_3^+ \qquad (1)$$

The kinetics of formation of D_3^+ ions in deuterium containing plasmas is sufficiently well understood and has been described several times (see e.g. (Poterya et al. 2002)). In the present experiments electron number density evolution (decay) along the flowing afterglow plasma is measured using the axially movable Langmuir probe (from

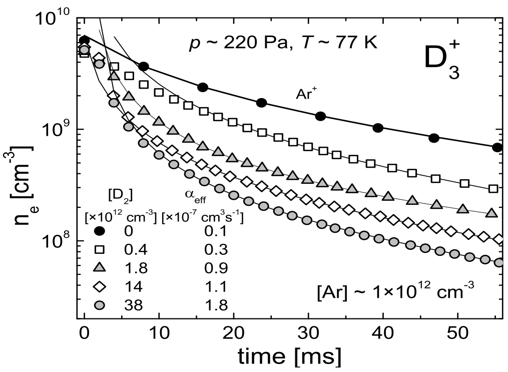

Figure 2: Measured electron density decays along the Flow Tube in He-Ar-D_2 plasmas at several concentrations of D_2. The solid lines represent fits of the data sets; the obtained rate coefficients are indicated.

the P2 port down to the end of the flow tube; 35 cm). The examples of measured electron density decays along the Flow Tube at several concentrations of D_2 (at $T =$ 77 K) are shown in Figure 2. The indicated values of $\alpha_{\rm eff}$ were calculated from the decay curves using advanced data analysis (for details see (Korolov et al. 2008)).

3. RESULTS AND DISCUSSION

In Figure 3, the obtained effective recombination rate coefficients ($\alpha_{\rm eff}$) are plotted as a function of deuterium concentration ([D_2]). The previous AISA data (Poterya et al. 2002) were also included in the figure. The observed difference between the rate coefficients measured at $T = 250$ K in AISA and FALP experiments is given by the difference in helium number densities in these experiments.

Because of large extend of deuterium densities the plasmas at low [D_2] differ substantially from plasma at high [D_2]. The difference is in number of collisions (N) of ion with deuterium molecule prior to its recombination with electron. Figure 3 is divided into three sections. In the left section with $[D_2] \leq 1 \times 10^{12}$ cm^{-3} ion has typically less than 1 collision with D_2 prior to recombination N < 1. In the right section $D_2 > 1 \times 10^{13}$ cm^{-3} ion has many collisions with D_2, N >> 1, at used temperatures and helium pressures D_5^+ ions are formed in three-body association. As a consequence decay of the plasma is influenced by recombination of D_5^+ ions and measured $\alpha_{\rm eff}$ is increasing with increasing [D_2] (Novotny et al. 2006). In the middle section (1×10^{12} cm^{-3} < [D_2] < 1×10^{13} cm^{-3}) D_3^+ ion will collide several times with D_2 before recombining with electron, N > 1. We will denote these condition as "saturated region". In these collisions ion (D_3^+) will be thermalized (internal excitation). The kinetic energy of D_3^+ will be thermalized immediately after its formation in collisions with He atoms (He density is higher by many orders of magnitude). Because of these fast relaxation processes the plasma is in the conditions corresponding to the middle section in thermodynamic equilibrium. We have studied temperature and pressure dependence of the value of recombination rate coefficient in this plasma. Figure 4

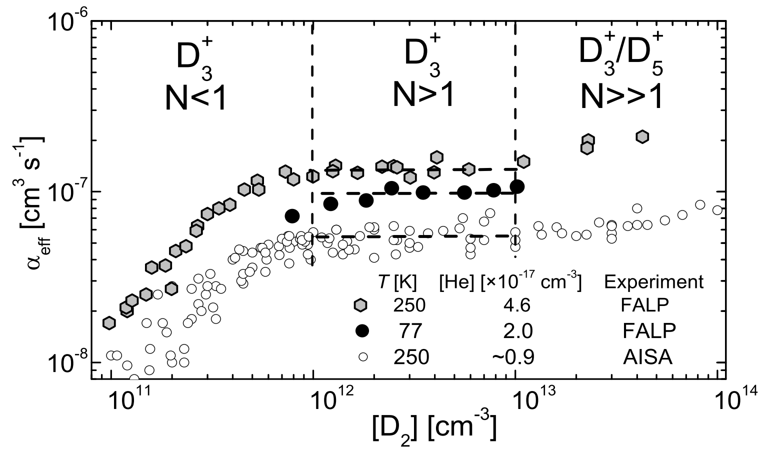

Figure 3: The effective recombination rate coefficients measured over a broad range of deuterium number densities. The dashed lines indicate values of recombination rate coefficients considered to conditions in "saturated region", i.e. in conditions where ions D_3^+ have many collisions with D_2 and the decaying plasma is in thermodynamic equilibrium with helium and deuterium.

shows the dependence of recombination rate coefficients on helium number density. The plotted rate coefficients were measured in different experiments at two different temperatures and at deuterium densities corresponding to the "saturated region". The figure also includes the calculated value of the recombination rate coefficient for binary dissociative recombination of D_3^+ at 250 K (Kokoouline et al. 2008).

The linear dependence of α_{eff} on helium density is obvious. Therefore, the dependence of measured aeff on helium number density can be written as:

$$\alpha_{\text{eff}} = \alpha_{\text{bin}} + \alpha_{\text{ter}} = \alpha_{\text{bin}} + K_{\text{He}}[\text{He}] \tag{2}$$

From the data corresponding to 250 K and 200 K we obtained: $\alpha_{\text{bin}}(250\text{ K}) = (4.1 \pm 1.2) \times 10^{-8}\text{ cm}^3\text{s}^{-1}$ $\alpha_{\text{bin}}(200\text{ K}) = (4.7 \pm 1.4) \times 10^{-8}\text{ cm}^3\text{s}^{-1}$. These values agree with the values calculated for binary dissociative recombination (DR) of the corresponding ions (Kokoouline 2008). We assume that ternary channel is coupled with formation of long-lived highly exited Rydberg molecules (D_3^*) in the interaction of electrons with D_3^+ ions. If average lifetime of D_3^* is long, then this highly excited molecule can eventually collide with buffer gas atom, affecting the recombination process:

$$\begin{array}{l} D_3^+ + e^- \rightleftarrows D_3^* \rightarrow neutrals\ Binary \\ \qquad\qquad\quad \downarrow \text{He} \\ \qquad\qquad\quad\ \rightarrow neutrals\ Ternaty \end{array}$$

The obtained values of ternary rate coefficient K_{He} for 250 K and 200 K are $(2.1 \pm 0.7) \times 10^{-25}\text{ cm}^6\text{s}^{-1}$ and $(3.5 \pm 1.1) \times 10^{-25}\text{ cm}^6\text{s}^{-1}$ respectively.

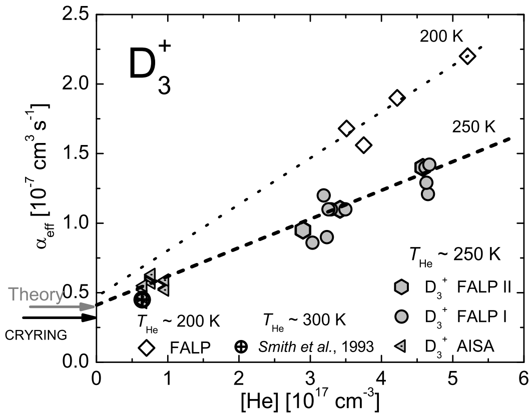

Figure 4: Effective recombination rate coefficients of D_3^+ measured in He-Ar-D_2 afterglow experiments. The arrow "Theory" indicates calculated values for 250 K (Kokoouline 2008). The dashed and dotted line connect measured data with theoretical value of rate coefficient for binary dissociative recombination (Kokoouline et al. 2008) for 250 K and 200 K. The arrow "Cryring" indicates the value of recombination rate coefficient obtained from the cross section measurements in CRYRING experiment (Padellec et al. 1998, Larsson et al. 1997). The thermal value obtained by Smith and Spanel in FALP experiment (300 K) is also included.

4. CONCLUSIONS

The measurements of recombination rate coefficient of D_3^+ ion at well defined conditions of afterglow plasma in a FALP experiment were performed. We observed strong pressure and temperature dependence of the rate coefficient indicating the existence of ternary channel in the overall recombination process. The observed ternary process is 100 times faster than previously observed three body recombination (Cao et al. 1991) and indicates extraordinary mechanism of D_3^+ recombination. The study of temperature dependence of the ternary channel of D_3^+ recombination process is in progress. The dependence of the effective rate coefficient on the D_2 density at $[D_2] < 1 \times 10^{12}$ cm^{-3} can indicate that different rotational and nuclear spin states of D_3^+ are not at the equilibrium on the timescale of the present experiment.

Acknowledgements

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 (202/07/0495, 202/08/H057) by GAUK 53607, GAUK 124707 and GAUK 86908.

References

Cao, Y. S. and Johnsen, R.: 1991, *J. Chem. Phys.*, **94**, 5443.
Glosik, J., Bano, G., Plasil, R., Luca, A., Zakouril, P.: 1999, *Int. J. Mass Spectrom.*, **189**, 103.

Johnsen, R.: 2005, *J. Phys.: Conf Series*, **4**, 83.
Kokoouline, V., Greene, C. H., Esry, B. D.: 2001, *Nature (London)*, **412**, 891.
Kokoouline, V., Greene, C. H.: 2003, *Phys. Rev. A*, **68**, 012703.
Kokoouline, V.: 2008, *private communication.*
Korolov, I., Novotny, O., Kotrik, T., Plasil, R., Varju, J., Hejduk, M., Glosik, J.: 2008, *Contrib. Plasma Phys.*, in print.
Korolov, I., Novotny, O., Plasil, R., Hlavenka, P., Kotrik, T., Tichy, M., Kudrna, P., Glosik, J., Luca, A.: 2006, *Czech. J. Phys.*, **56**, 854.
Kreckel, H. et al.: 2005, *Phys. Rev. Lett.*, **95**, 263201.
Larsson, M. and Orel, A.E.: 2008, Dissociative Recombination of Molecular Ions, Cambridge University Press, Cambridge.
Larsson, M., Danared, H., Larson, A., Le Padellec, A., Peterson, J. R., Rosen, S., Semeniak, J., Stromholm, C.: 1997, *Phys. Rev. Lett.*, **79**, 395.
McCall, B. J. et al.: 2003, *Nature*, **422**, 500.
McCall, B. J. et al.: 2004, *Phys. Rev. A*, **70**, 052716.
Novotny, O., Plasil, R., Pysanenko, A., Korolov, I. and Glosik, J.: 2006, *J. Phys. B: At. Mol. Opt. Phys.*, **39**, 2561.
Padellec, A., Larsson, M., Danared, H., Larson, A., Peterson, J. R., Rosen, S., Semeniak, J., Stromholm, C.: 1998, *Phys. Scripta*, **57**, 215.
Plasil, R., Glosik, J., Poterya, V., Kudrna, P., Rusz, J., Tichy, M. and Pysanenko, A. : 2002, *Int. J. Mass. Spectrom.*, **218**, 105.
Plasil, R., Korolov, I., Kotrik, T., Glosik, J.: 2008 *Int. J. Mass Spectom. Ion Process.*, in print.
Poterya, V., Glosik, J., Plasil, R., Tichy, M., Kudrna, P. and Pysanenko, A.: 2002, *Phys. Rev. Lett.*, **88**, 044802.
Santos, S. F., Kokoouline, V. and Greene, C. H.: 2007, *J. Chem. Phys.*, **127**, 124309.
Smith, D. and Öpatnel, P.: 1993, *Int. J. Mass Spectrom. Ion Process.*, **129**, 163.