Cryo-FALP study of collisional-radiative recombination of Ar ions at 40-200 K

T. Kotrík, P. Dohnal, P. Rubovič, R. Plašil, Š. Roučka, S. Opanasiuk, J. Glosík

► To cite this version:

T. Kotrík, P. Dohnal, P. Rubovič, R. Plašil, Š. Roučka, et al.. Cryo-FALP study of collisional-radiative recombination of Ar ions at 40-200 K. *European Physical Journal: Applied Physics*, 2011, 56 (2), pp.24011. 10.1051/epjap/2011110158 . hal-00746209

HAL Id: hal-00746209

<https://hal.science/hal-00746209>

Submitted on 28 Oct 2012

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Cryo-FALP study of collisional radiative recombination of Ar^+ ions at 40 – 200 K.

T. Kotrík, P. Dohnal, P. Rubovič, R. Plašil, Š. Roučka, S. Opanasiuk, and J. Glosík

Department of Surface and Plasma Physics, Faculty of Mathematics and Physics, Charles University, V Holešovičkách 2, Praha 180 00, Czech Republic, e-mail: kotrik.tomas@gmail.com

Received: date / Revised version: date

Abstract. New flowing afterglow apparatus, Cryo-FALP, was built to study ternary Collisional Radiative Recombination (CRR) of Ar^+ ions with electrons in He/Ar afterglow plasma at temperatures 40 – 200 K. The obtained ternary recombination rate coefficient at 57 K is $K_{\text{CRR}} = (3 \pm 1) \times 10^{-17} \text{ cm}^6 \text{ s}^{-1}$. It is the first time that the K_{CRR} was measured below 77 K. The measured temperature dependence $K_{\text{CRR}} \sim T^{(-4.5 \pm 0.4)}$ is in a good agreement with theoretical prediction.

Key words. Collisional radiative recombination – ternary electron assisted recombination – flowing afterglow – Ar^+ – low temperature plasma.

1 Introduction

A binary recombination of singly charged atomic cations with electrons is slow process comparing to a dissociative recombination (DR) of molecular ions (like O_2^+ or H_3^+ , etc.). In low temperature plasma an overall recombination process can be enhanced by a ternary process where part of the recombination energy is transferred to a third particle (neutral or charged). When plasma electrons act as energy-removing third bodies, the process is referred to as collisional-radiative recombination, CRR. Schematically, the mechanism of collisional radiative recombination can be described by the formula:

where A^+ denotes positive ion in general, α_{CRR} corresponds to the effective binary recombination rate coefficient of CRR. The theory of CRR was originally developed by Bates [1] and Mansbach with Keck [2]. The analytical formula derived by Stevefelt [3] for the effective binary rate coefficient of CRR has the form:

$$\alpha_{\text{CRR}} = 3.8 \times 10^{-9} T_e^{-4.5} n_e + 1.55 \times 10^{-10} T_e^{-0.63} + 6 \times 10^{-9} T_e^{-2.18} n_e^{0.37} \text{ cm}^3 \text{ s}^{-1}, \quad (2)$$

where T_e is electron temperature given in K and n_e is electron number density given in cm^{-3} . The first term belongs to the pure recombination through a collision with electron (note strong temperature dependence), the second term corresponds to the radiative recombination, where the exceeding energy is carried away by an emitted photon, and the third term accounts for the coupling between the collisional and radiative recombination. As only

higher order Rydberg states participate on the CRR process, formula (2) holds more or less for all atomic and in some extent also molecular ions (for details see ref. [3] and references herein). This formula was confirmed also by the quantum mechanical calculations [4–6]. At our experimental conditions ($n_e \sim 10^9 \text{ cm}^{-3}$, $T_e < 200 \text{ K}$) the first term (pure recombination through collision with electron) is dominant and the other two terms can be neglected. Then the recombination process can be described by the simpler ternary recombination rate coefficient: $K_{\text{CRR}} = \alpha_{\text{CRR}}/n_e = 3.8 \times 10^{-9} T_e^{-4.5} \text{ cm}^6 \text{ s}^{-1}$.

Previous experimental studies have been carried out mostly at higher electron temperatures (1000 - 4000 K, see compilation in [4]) and in only few cases also at 300 K [7–10]. It is not obvious, that the Stevefelt formula holds also at low temperatures (below 300 K). Only very recently CRR was studied in our laboratory at temperatures below 200 K and K_{CRR} was obtained for temperatures from 77 up to 200 K [11]. Process of CRR has been studied also in ultracold plasmas in connection with formation of neutral antihydrogen, where it is expected that at high positron number density the production of antihydrogen via recombination of positrons with antiprotons can be enhanced by CRR process. Nevertheless in these experiments CRR is influenced by the presence of strong magnetic field confining the plasma and neutralization process can have different character [12–14].

To study CRR in low temperature plasma we constructed a new flowing afterglow apparatus, Cryo-FALP, where afterglow plasma can be cooled down to 40 K. In this paper we present the first results of the study of recombination in plasma dominated by Ar^+ ions in temperature range 40 – 77 K. It is the first FALP apparatus ever

Fig. 1. Scheme of the Cryo-FALP apparatus. The temperatures of sections A, B are fixed at 300 and 100 – 120 K respectively. The temperature of section C can be adjusted to a required value in the range 40 – 300 K. The plasma decay time is given by the position in the flow tube. The axially movable Langmuir probe is used to monitor plasma decay along the flow tube, decay time $t = L/v_{\text{PLASMA}}$.

build to operate at temperatures below the boiling point of liquid nitrogen.

2 Experiment

Modified version of the Flowing Afterglow with Langmuir Probe (FALP, for original design see ref. [15]) apparatus – Cryo-FALP (for details on the previous versions of FALP apparatuses operating at higher pressure see refs. [16–19]) was build and used to study CRR at temperatures down to 40 K. Scheme of the Cryo-FALP is shown in Figure 1. Helium buffer gas (300 – 800 Pa) is flowing through a glass discharge tube into a stainless steel flow tube. Plasma, consisting of helium ions (He^+ , He_2^+), helium metastables (He^m) and electrons, is created in pure He by a microwave discharge (2.54 GHz, 10 – 20 W) ignited in a cavity attached to the glass discharge tube. Plasma is then driven along the flow tube by the flow of helium buffer gas. Downstream from the discharge region argon gas is added to the flow tube and Ar^+ dominated plasma is formed in the sequence of ion-molecule reactions:

In order to set the optimal experimental conditions (He pressure and flow, Ar number density) at which Ar^+ ion is the dominant ion in afterglow plasma, a chemical kinetics model was developed and numerically solved (for details on reaction kinetics and the kinetics model see refs. [16, 17, 20]).

The flow tube is divided into three sections with different temperature regime. The upstream section A of the flow tube, in which the microwave discharge is ignited, is kept at 300 K. The section further downstream, B, is cooled by liquid nitrogen to 100 – 120 K. The following section C is placed in a large insulating vacuum chamber and is connected via copper braids to a cold-head of helium closed cycle refrigerator (Sumitomo). The temperature of section C can be adjusted to a value between 40

and 300 K. Further details on the design and construction will be given in a separate publication (ref. [21]).

Plasma, created in section A, undergoes many collisions with neutral particles. Through the collisions, after a short period of time outside of the discharge region ($\sim \text{ms}$), electrons and ions thermalize to the temperature of the neutral species (i.e. temperature of the walls). The thermodynamic equilibrium is reached and instead of the temperature of electrons (T_e), ions (T_i) and neutrals (T_n) only one temperature can be used $T = T_e = T_i = T_n$ (for details and discussion on plasma species thermalization in afterglow plasma see refs. [11, 22–24]). At the beginning of section B plasma is precooled via collisions with helium buffer gas to 100 – 120 K and in section C to the desired temperature adjusted in the range 40 – 300 K (see discussion later on). Temperature of the wall is monitored by sensors (silicon diodes, thermocouples type T) mounted along the wall of the flow tube.

Recombination rate coefficients are derived from the electron density decay monitored along section C by the axially movable cylindrical Langmuir probe [17, 25]. While gas flow velocity (v) (i.e. plasma flow velocity) is known, spatial position (L) of the Langmuir probe along the flow tube can be transformed into the time (t) passed since the plasma creation by the relation $t = L/v$. The decay of the electron density due to CRR and diffusion in quasineutral plasma ($[\text{Ar}^+] = n_e$) is described by the balance equation:

$$\frac{dn_e}{dt} = -K_{\text{CRR}} [\text{Ar}^+] n_e^2 - \frac{n_e}{\tau_D} = -K_{\text{CRR}} n_e^3 - \frac{n_e}{\tau_D}, \tag{4}$$

where τ_D is characteristic diffusion time. K_{CRR} and τ_D can be obtained from the fit of the measured electron density decay under the assumption of K_{CRR} and τ_D being constant (for details on the data analysis see ref. [11]).

Because of the temperature variation along sections B and C, gas density and flow velocity is changing along the flow tube. Plasma velocity v_{PLASMA} necessary for determination of the decay time is measured by the modulation of the discharge and by monitoring the time delay of the distortion along the flow tube (see Figure 2). Plasma velocity depends on temperature (T), on gas flow (F) and on pressure (p). Because gas flow and pressure are constant along the flow tube, gas velocity is proportional to temperature (T). When the gas temperature relaxes to the wall temperature, velocity is constant (see linear fits in Figure 2). In this region with constant temperature, from the decay of electron density, the effective recombination rate coefficient is obtained. Because of very strong temperature dependence of CRR, the measurements of the plasma velocity dependence on the flow parameters are very important in order to define conditions in which plasma is thermalized to the temperature of the flow tube wall. Measured dependence of the plasma velocity on "similarity parameter" $\phi = FT/p$ in Ar^+ and He_2^+ dominated plasma is plotted in Figure 3. To better understand the thermalization of buffer gas and plasma with the flow tube walls a simple computer model of heat transport inside the flow tube was developed and numerically solved (for details on the computer model see ref. [21]). The examples of gas

Fig. 2. The measurements of plasma velocity. The microwave discharge is modulated and the dependence of the time delay (Δt) of the responds is measured as a function of the probe position (L).

Fig. 3. The measured dependence of plasma velocity on the "similarity parameter" $\phi = FT/p$, where F is gas flow rate, T is temperature and p is pressure of the buffer gas. Indicated is dominant ion in the plasma.

temperature evolution calculated for different gas velocities at temperature of the flow tube ~ 40 K are plotted in Figure 4. The calculated buffer gas (and plasma) thermalization agrees well with the experimental insight gained from the plasma flow velocity measurements (linear fits in Figure 2) and from the measurements of the diffusion losses (see ref. [11]).

Fig. 4. Evolution of buffer gas temperature in section C calculated for different flow velocities. Solid line denotes calculated temperature of the flow tube wall, the flow tube construction, materials and heat transfers are considered.

Fig. 5. Examples of the electron decay curves measured at 57 and 77 K (temperature of section C). Experimental conditions are listed in the plot. From the fits of the plots in the region of constant plasma temperature (solid lines), the coefficients K_{CRR} are obtained. The dashed line indicate diffusion losses at 57 K, 450 Pa.

3 Results and conclusions

The examples of the measured electron density decay curves at particular experimental conditions are plotted in Figure 5. Measurements of the electron density decays were performed at various experimental conditions in order to control and verify the thermalization of plasma with the flow tube wall. At low temperatures and high enough argon number densities argon clusters Ar_2^+ with fast binary recombination rate coefficients ($\sim 10^{-6} \text{ cm}^3 \text{s}^{-1}$) can be

Fig. 6. Measured K_{CRR} coefficient as a function of temperature. Dashed line corresponds to the theoretical value calculated from the collisional (first) term of equation (2). Dotted lines indicate the theoretical values of K_{CRR} for two electron densities calculated using all terms of relation (2). Closed square and triangle are data measured by Skrzypkowski et al. [8] and by Berlande et al. [10].

created. As the present version of FALP apparatus is not equipped with a mass spectrometer, one needs to follow the optimal conditions suggested by the chemical kinetics model and/or examine the dependence of α_{eff} on $[\text{Ar}]$ in order to exclude the formation of fast recombining Ar_2^+ ions. Such measurements were performed (for results and discussion see ref. [21]) and after careful examination of the results, the eventual electron assisted ternary recombination rate coefficient K_{CRR} of Ar^+ ion obtained at particular temperature in the range 50 – 170 K is plotted in Figure 6. Open triangles denote experimental values obtained in the present version of FALP apparatus, full circles and squares correspond to values measured in the previous FALP experiment, where the flow tube was cooled by liquid nitrogen (for details see ref. [11]).

The theoretical dependence (2) is plotted by dashed line (collisional term only). Dotted lines denote theoretical K_{CRR} obtained from the formula $K_{\text{CRR}} = \alpha_{\text{eff}}/n_e$ for two different electron number densities when all terms of equation (2) are involved in α_{eff} calculation. The results show an excellent agreement of the measured K_{CRR} with the theory down to 50 K. Further studies of CRR in Ar^+ dominated plasma are in progress.

4 Acknowledgments

This work is a part of the research plan MSM 0021620834 and grant OC10046 financed by the Ministry of Education of the Czech Republic and was partly supported by GACR (202/07/0495, 202/08/H057, 205/09/1183, 202/09/0642), by GAUK 92410, GAUK 353811, GAUK 54010 and by COST Action CM0805 (The Chemical Cosmos).

References

1. D.R. Bates, A.E. Kingston, R.W.P. McWhirter, *Proc. Roy. Soc. A* **267**, (1962) 297.
2. P. Mansbach, J. Keck, *Phys. Rev.*, **181**, (1969) 275.
3. J. Stevefelt, J. Boulmer, J-F. Delpech, *Phys. Rev. A* **12**, (1975) 1246.
4. G.J. Pert, *J. Phys. B: At. Mol. Opt. Phys.* **23**, (1990) 619.
5. S.X. Hu, *Phys. Rev. Lett.* **98**, (2007) 133201-1.
6. T. Pohl, D. Vrinceanu, H. R. Sadeghpour, *Phys. Rev. Lett.* **100**, (2008) 223201.
7. M. Tsuji, T. Matsuzaki, T. Tsuji, *Chem. Phys.* **285**, (2002) 335.
8. M.P. Skrzypkowski, R. Johnsen, R. E. Rosati, M. F. Golde, *Chem. Phys.* **296**, (2004) 23.
9. G.E. Veatch, H.J. Oskam, *Phys. Rev. A* **1**, (1970) 1498.
10. J. Berlande, M. Cheret, R. Deloche, A. Gonfalone, C. Manus, *Phys. Rev. A* **1**, (1970) 887.
11. T. Kotrík, P. Dohnal, S. Roucka, P. Jusko, R. Plasil, J. Glosik, *Phys. Rev. A* **83**, (2011) 032720.
12. R.S. Fletcher, X. L. Zhang, S.L. Rolston, *Phys. Rev. Lett.* **99**, (2007) 145001-1.
13. S.X. Hu, D. Vrinceanu, S. Mazevet, L.A. Collins, *Phys. Rev. Lett.* **95**, (2005) 163402-1.
14. M. Amoretti et al., *Phys. Lett. B* **583**, (2004) 59.
15. D. Smith, N.G. Adams, A.G. Dean, M.J. Church, *J. Phys. D* **8**, (1975) 141.
16. J. Glosik, O. Novotny, A. Pysanenko, P. Zakouril, R. Plasil, P. Kudrna, V. Poterya, *Plasma Sources Sci. Technol.* **12**, (2003) 117.
17. I. Korolov, O. Novotný, J. Varju, T. Kotrík, R. Plašil, M. Hejduk, J. Glosík, *Contrib. Plasma Phys.* **48**, (2008) 521.
18. J. Glosík, R. Plašil, I. Korolov, T. Kotrík, O. Novotný, P. Hlavenka, P. Dohnal, J. Varju, V. Kokoouline, C. H. Greene, *Phys. Rev. A* **79**, (2009) 052707.
19. T. Kotrík, P. Dohnal, I. Korolov, R. Plašil, Š. Roučka, J. Glosík, C. H. Greene, V. Kokoouline, *J. Chem. Phys.* **133**, (2010) 034305.
20. R. Plasil, J. Glosik, V. Poterya, P. Kudrna, J. Rusz, M. Tichy, A. Pysanenko, *Int. J. Mass Spectrom.* **218**, (2002) 105.
21. J. Glosik, et al., to be published.
22. H.J. Oskam, *Philips Res. Rep.* **13**, (1958) 335.
23. I. Korolov, T. Kotrík, P. Dohnal, O. Novotny, J. Glosik, *Contrib. Plasma Phys.* **48**, (2008) 461.
24. R. Plasil, I. Korolov, T. Kotrík, P. Dohnal, G. Bano, Z. Donko, J. Glosik, *Eur. Phys. J. D* **54**, (2009) 391.
25. J.D. Swift, M.J.R. Schwar, *Electrical Probes for Plasma Diagnostics* (Illiffe, London 1970).