Recombination in low temperature Ar^+ dominated plasma

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

Department of Surface and Plasma Science, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic

Introduction

Recombination of atomic and molecular cations with electrons is very important loss process of charged particles in plasma. At 300 K, molecular ions usually undergo dissociative recombination to neutral products with typical rate coefficient $\alpha \sim 10^{-7}\,\mathrm{cm^3s^{-1}}$. Atomic ions usually recombine radiatively with rather small rate coefficient $\alpha \sim 10^{-12}\,\mathrm{cm^3s^{-1}}$. In plasma at temperatures below 100K ternary recombination processes with very strong temperature dependences start to influence overall recombination process. There are no experimental data for these processes at temperatures below 77 K except [1, 2].

The ternary recombination can be described as a two step process. After the collision of e^- with cation A^+ a neutral excited intermediate state A^* is formed. In following collision with another particle the complex is stabilized and the recombination process is terminated. In case of collisional radiative recombination (CRR) the third particle is another e^-:

$$A^+ + e^- + e^- \xrightarrow{K_{\mathrm{CRR}}} A + e^- \,. \tag{1}$$

This process can be described by ternary recombination rate coefficient K_{CRR}. If CRR is the only loss process, the process can be described by effective binary recombination rate coefficient α_{CRR}

$$\alpha_{\mathrm{eff}} = K_{\mathrm{CRR}} \cdot n_{\mathrm{e}} \,, \tag{2}$$

where n_{e} is electron number density. Dependence of K_{CRR} on T and n_{e} was predicted by Stevefelt [3]:

$$K_{\mathrm{CRR}}(T,n_{\mathrm{e}}) = 3.8 \cdot 10^{-9} T_{\mathrm{e}}^{-4.5} n_{\mathrm{e}} + 1.55 \cdot 10^{-10} T_{\mathrm{e}}^{-0.63} + 6 \cdot 10^{-9} T_{\mathrm{e}}^{-2.18} n_{\mathrm{e}}^{0.37}\,\mathrm{cm^6s^{-1}}\,. \tag{3}$$

At our conditions ($n_{\mathrm{e}} \sim 10^9\,\mathrm{cm^{-3}}$ and $T < 100\,\mathrm{K}$) only the first term is dominant and we can write

$$K_{\mathrm{CRR}}(T,n_{\mathrm{e}}) = 3.8 \cdot 10^{-9} T_{\mathrm{e}}^{-4.5} n_{\mathrm{e}} \,. \tag{4}$$

The second ternary process which have to be considered is recombination through collision with neutral particle of the buffer gas M:

$$A^+ + e^- + M \xrightarrow{K_{\mathrm{M}}} A + M \,. \tag{5}$$

In experiment, K_M can be obtained from the dependence of the effective recombination rate coefficient on the number density of neutral particle [M] and

$$\alpha_{\mathrm{eff}} = K_{\mathrm{M}} \cdot [\mathrm{M}]\,. \tag{6}$$

Theoretical value given by Bates [4] is in our case (He as a buffer gas)

$$K_{\mathrm{He}}(T) = 5.9 \cdot 10^{-28} \cdot \left(\frac{300}{T}\right)^{-2.91} \mathrm{cm}^6\mathrm{s}^{-1}\,. \tag{7}$$

Experimental apparatus

In this study we used Cryo–FALP II apparatus (see figure 1). It is based on a classic flowing afterglow with Langmuir probe (abb. FALP) design [5]. Buffer gas is flowing through a flow tube. Discharge (2.45 GHz, ∼ 20–25 W) is ignited in a microvawe cavity at the beginning of the flow tube. Downstream, reactant gas (or gases) are added and plasma decay is monitored by means of Langmuir probe, from which we can determine concentration and temperature of electrons n_e and T_e. Position of the probe is converted into the decay time by known gas velocity [2].

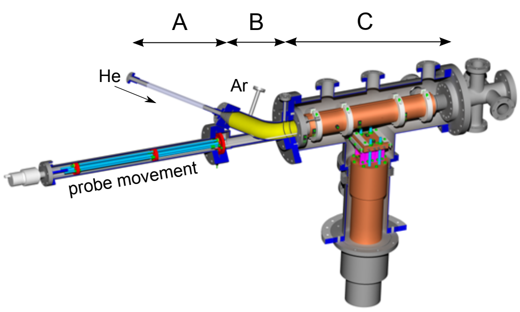

Figure 1: Drawing of Cryo–FALP II apparatus.

At temperatures below 300 K, plasma decay is driven by two processes – its recombination and ambipolar diffusion. Accounting quasineutrality of plasma and presumption that we have only one dominant ion, time evolution of electron concentration n_e can be described by following equation [6]:

$$\frac{\mathrm{d}n_{\mathrm{e}}}{\mathrm{d}t} = -\alpha_{\mathrm{eff}} n_{\mathrm{e}}^2 - \frac{n_{\mathrm{e}}}{\tau_{\mathrm{D}}}\,, \tag{8}$$

where α_{eff} is an effective recombination rate coefficient and τ_D is a characteristic time of ambipolar diffusion. α_{eff} can be rewritten with (2) and (6) and accounting binary recombinatio rate coefficient (negligible in this experiment) as:

$$\alpha_{\mathrm{eff}}(T) = \alpha_{\mathrm{bin}} + K_{\mathrm{CRR}} n_{\mathrm{e}} + K_{\mathrm{He}}[\mathrm{He}]\,. \tag{9}$$

One can see, that from measured dependence of α_{eff} on helium number density (i.e. pressure) both K_{CRR} and K_{He} can be obtained.

In order to study processes which occur at temperatures below 77 K, we built a new Cryo–FALP II apparatus [2]. Flowtube itself is divided into the three main parts. The first part (A), where microwave discharge is ignited is kept at room temperature. The second part (B), where reactant gases are added (argon in this case) can be precooled by liquid nitrogen to ~ 110 K temperature. The third part (C) is connected to the coldhead and can be cooled down to ~ 40 K.

Results

We studied collisional radiative recombination of Ar^+ ions:

$$Ar^+ + e^- + e^- \longrightarrow Ar + e^- . \tag{10}$$

Temperature dependence of measured coefficient of collisional radiative recombination is displayed in figure 2. Our current data (red triangles), older data measured at Cryo-FALP (open green circles) [1], data from other experiments (green open square and orange rhombus) [7, 8] and theoretical dependence (dashed line) [3] can be seen. Very good agreement across the whole temperature range is observed.

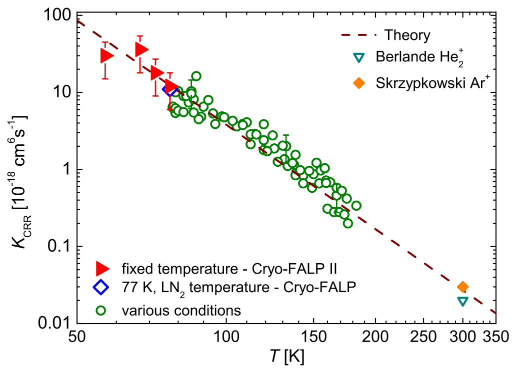

Figure 2: Temperature dependence of K_{CRR}.

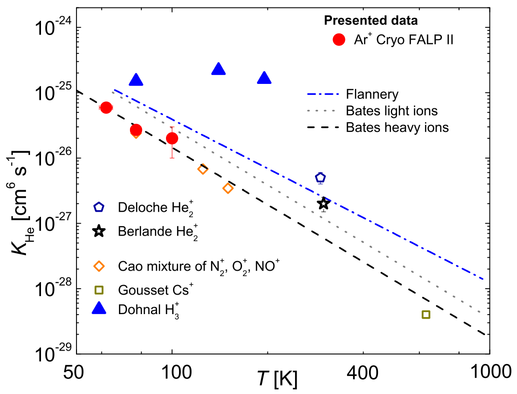

Figure 3: Temperature dependence of K_{He}.

Ternary recombination of Ar^+ ions

$$Ar^+ + He + e^- \longrightarrow Ar + He \tag{11}$$

was also studied. Temperature dependence of the measured ternary recombination rate coefficient of Ar^+ ions with electrons is shown in figure 3 (red circles) together with the data form other experiments [7, 10, 9, 13]. The obtained rate coefficients follow the $T^{-2.9}$ temperature dependence in agreement with theory for ternary recombination of atomic ions [4, 12] and agree well also with the data from other experiments with exception of H_3^+ ion [13]. To our knowledge, these data are the first one for ternary recombination measured below 77 K.

Discussion

We studied recombination of Ar^+ ion with electrons in the temperature range of 57–100 K using Cryo-FALP II apparatus equipped with Langmuir probe. Obtained results for collisional radiative recombination and helium assisted ternary recombination rate coefficient are in a good agreement with theories.

Acknowledgments: This work was partly financed by the research grant OC10046 from the Ministry of Education of the Czech Republic and was partly supported by GACR (205/09/1183, P209/12/0233), by SV 265 302, by GAUK 92410, GAUK 353811, GAUK 54010 and by COST Action CM0805 (The Chemical Cosmos).

References

[1] T. Kotrik, P. Dohnal, S. Roucka, P. Jusko, R. Plasil, J. Glosik, R. Johnsen, Phys. Rev. A **83**, 032720 (2011)

[2] T. Kotrik, P. Dohnal, P. Rubovic, R. Plasil, S. Roucka, S. Opanasiuk, J. Glosik, Eur. Phys. J. Appl. Phys. **56**, 24011 (2011)

[3] J. Stevefelt, J. Boulmer, J. Delpech, Phys. Rev. A **12**, 1246 (1975)

[4] D. Bates, S. Khare, Proc. Phys. Soc. Lond. **85**, 231 (1965)

[5] M. R. Mahdavi, J. B. Hasted, M. M. Nakshbandi, J. Phys. B: Atom. Molec. Phys. **4**, 1726 (1971)

[6] F. F. Chen, Introduction to plasma physics, Plenum Press, New York (1974)

[7] J. Berlande, M. Cheret, R. Deloche, A. Gonfalone, C. Manus, Phys. Rev. A **1**, 887 (1970)

[8] M. P. Skrzypkowski, R. Johnsen, R. E. Rosati, M. F. Golde, Chem. Phys. **296**, 23 (2004)

[9] R. Deloche, P. Monchicourt, M. Cheret, F. Lambert, Phys. Rev. A **13**, 1140 (1976)

[10] Y. S. Cao, R. Johnsen, J. Chem. Phys. **94**, 5443 (1991)

[11] G. Gousset, B. Sayer, M. Ferray, J. Lozingot, J. Quant. Spectrosc. Radiat. Tranfer **19**, 239 (1978)

[12] M. R. Flannery, J. Chem. Phys. **95**, 8205 (1991)

[13] P. Dohnal, M. Hejduk, J. Varju, P. Rubovic, S. Roucka, T. Kotrik, R. Plasil, J. Glosik, J. Chem. Phys. **136**, 244304 (2012)