New Cryo-FALP Experiment to Study Collisional Radiative Recombination of $\text{Ar}^+$ Ions at 40–200 K

T. Kotrik, P. Dohnal, S. Opanasiuk, P. Rubovič, Š. Roučka, R. Plašil, and J. Glosík
Charles University Prague, Faculty of Mathematics and Physics, Prague, Czech Republic.

Abstract. Novel flowing afterglow apparatus (Cryo-FALP) for studying elementary processes in plasma in the temperature range 40–200 K was designed and built. Presented is study of Collisional Radiative Recombination (CRR) of $\text{Ar}^+$ ions with electrons in helium-buffered afterglow at electron densities $10^8–10^{10}$ cm$^{-3}$ and at temperatures 50–200 K. The obtained ternary recombination rate coefficient $K_{\text{CRR}}(77 \text{ K}) = (1.2 \pm 0.6) \times 10^{-17}$ cm$^6$s$^{-1}$. The observed temperature dependence agrees well with theoretical $T^{-4.5}$ prediction. It is the first time the $K_{\text{CRR}}$ was measured below 77 K.

Introduction

A binary recombination of singly charged atomic ions with electrons is rather slow process comparing to a dissociative recombination of molecular ions (like $\text{O}_2^+$, $\text{H}_3^+$, etc.). Overall recombination process can be enhanced by collision with a third particle (either neutral or charged), which carries away the exceeding energy. If the third body is an electron, we refer to a collisional radiative recombination, CRR.

The theoretical background of CRR was originally developed by Bates [Bates et al., 1962] and Mansbach and Keck [Mansbach and Keck, 1969]. For practical purposes an analytical formula for effective binary rate coefficient of CRR derived by Stevefelt [Stevefelt et al., 1975] is commonly used:

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

(1)

where $T_e$ is the electron temperature given in K and $n_e$ is the electron number density given in cm$^{-3}$. The first term on the right-hand side of the formula (1) corresponds to the collisional recombination, the second term describes purely radiative recombination and the third term denotes the coupling between the collisional and the radiative recombination. At our experimental conditions ($n_e = 1–10 \times 10^9$ cm$^{-3}$, $T < 200$ K) the first term accounts for more than 90% of the overall recombination, hence in a good approximation the ternary recombination rate coefficient $K_{\text{CRR}}$ can be introduced:

$$
K_{\text{CRR}} = \alpha_{\text{CRR}} / n_e \approx 3.8 \times 10^{-9} T_e^{-4.5} \text{ cm}^6\text{s}^{-1}.
$$

(2)

$K_{\text{CRR}}$ is strongly dependent on temperature, but independent on $n_e$. Very recently Pohl et al. [Pohl et al., 2008] has published improved calculations which reduce the numerical factor in the first term of the equation (1) and (2) from $3.8 \times 10^{-9}$ to $2.77 \times 10^{-9}$.

Previous experimental studies were performed at higher electron temperatures (1000–4000 K, see compilation [Pert 1990]), and in few cases also at 300 K [Berlande et al., 1970; Veatch and Oskam, 1970; Tsuji et al., 2002; Skrzypkowski et al., 2004]. Only very recently, the CRR process has been studied in our group at temperatures below 300 K (77–200 K) [Kotrik et al., 2010, Kotrik et al., 2011]. In ultracold plasmas the CRR process has been studied with respect to production of antihydrogen, however in these experiments a strong magnetic field is present, that could influence the recombination process [Amoretti et al., 2004; Hu et al., 2005; Fletcher et al., 2007].

Experiment

For studying elementary processes in plasma at temperatures down to 40 K a novel version of Flowing Afterglow with Langmuir Probe apparatus—Cryo-FALP was designed and built (for details on previous versions see [Mahdavi et al., 1971; Glosik et al., 2003; Glosik et al., 2009a; Glosik et al., 2009b; Kotrik et al., 2011]). Scheme of the Cryo-FALP is shown in Figure 1.

Microwave discharge is ignited in helium gas in a glass discharge tube forming plasma composed of helium ions ($\text{He}^+$), helium metastables ($\text{He}^{m}$) and electrons. Plasma is then flowing through the stainless steel flow tube divided into three sections with different temperatures. The upstream section
Figure 1. The scheme of the Cryo-FALP apparatus. Microwave discharge is ignited in a glass discharge tube in helium gas flowing through three sections of stainless steel flow tube. Section A is kept at the room temperature (300 K), section B is cooled by liquid nitrogen to 100 – 120 K. Section C is connected by copper braids to a coldhead of helium closed-cycle refrigerator holding a 100 W of cooling power at 40 K. To adjust the temperature in the range 40–300 K the copper braids are attached to the coldhead through a copper stage equipped by small removable copper blocks and heating elements. By removing the copper blocks, heat conductance between flow tube and coldhead is reduced, thus decreasing the requirements on a heating power provided by heating elements. By combination of the copper blocks removal and/or applying appropriate heating power the temperature of the flow tube can be adjusted in the range 40–300 K within the accuracy of 1 K. In order to get rid of heat resistance across metal contacts, cryogenic vacuum grease was introduced in between the crucial metal joins.

Downstream from the discharge region argon gas is introduced into the flow tube, hereby creating $\text{Ar}^+$ dominated plasma via sequence of ion-molecule reactions [Plasil et al., 2002, Glosik et al., 2003, Novotny et al., 2006, Korolov et al., 2009, Kotrik et al., 2011]. Recombination rate coefficients are derived from the electron density decay monitored along the section C by movable Langmuir probe. By knowing the flow velocity of the gas in the flow tube the electron density decays along the flow tube can be converted into temporal decays.

The plasma velocity necessary for determination of decay time is measured by modulation of the discharge and monitoring the time delay $\Delta t$ of the distortion along the flow tube. Examples of measured dependence of the distortion time delays on the probe position $L$ are plotted in Figure 2. Gas, hence plasma, flow velocity depends on the temperature ($T$), on the gas flow ($F$) and on the pressure ($p$): $v \sim FT/p$. Because the gas temperature varies along the section B and C, the gas density and the flow velocity is also changing. As the gas flow and the pressure are constant along the flow tube, the gas velocity is proportional to the temperature. When the gas temperature relaxes to the wall temperature the velocity remains constant (see linear fits of the plots in Figure 2). In the region of constant temperature, the decay of the electron density is measured to obtain the effective recombination rate coefficient. For details on data analysis see ref. [Kotrik et al., 2011]. Because of the pronounced temperature dependence of CRR it is crucial to define experimental conditions and measurement region in which the plasma is already thermalised to the temperature of the flow tube wall. Measured dependence of the plasma velocity on the parameter $FT/p$ at different experimental conditions is plotted in Figure 3.

Results

The examples of measured ternary recombination rate coefficient $K_{\text{CRR}}$ as a function of argon number density in the temperature range 50–80 K and different experimental conditions are plotted in Figure 4. The data points at 77 K were measured in previous version of Cryo-FALP with the flow tube cooled by liquid nitrogen. In the range of experimental error are $K_{\text{CRR}}$ measured at particular temperatures constant, not dependent on the argon number density. Lines indicate theoretical values given by formula of Stevefelt et al. [1975].
Figure 2. Measurements of plasma velocity. The microwave discharge is modulated and the dependence of the time delay $\Delta t$ of the distortion measured at different experimental conditions as a function of the probe position $L$ is plotted. Plasma velocity is obtained from the linear fits of the data. Indicated temperature corresponds to the wall temperature in the section C of the flow tube.

Figure 3. The measured dependence of the plasma velocity $v$ on the parameter $FT/p$ at different experimental conditions.

Temperature dependence of measured $K_{CRR}$ is plotted in Figure 5. Shown are also experimental values obtained in our group with the previous version of Cryo-FALP apparatus with the flow tube cooled by liquid nitrogen. For comparison, experimental values at 300 K measured for Ar$^+$ by Skrzypkowski et al. [2004] and for He$_2^+$ by Berlande et al. [1970] are also plotted. Experimental results show an excellent agreement with the theory down to 50 K, thus proving the Steves and the formula over the range of many orders of magnitude. Further study of CRR in Ar$^+$ dominated plasma is in progress.
Figure 4. Measured $K_{\text{CRR}}$ plotted as a function of argon number density. Lines indicate theoretical values according to the first term of the equation (1).

Figure 5. Measured $K_{\text{CRR}}$ coefficients plotted as a function of temperature. Closed triangles and a star denote experimental values measured at fixed temperature in the Cryo-FALP apparatus with the flow tube cooled by the refrigerator and liquid nitrogen respectively. Data indicated by open circles were measured in the previous version of Cryo-FALP in regime with slowly increasing temperature of the flow tube (for details see ref. [Kotrik et al. 2011]). Closed circle and a diamond represent the data measured by Skrzypkowski et al. [2004] and by Berlande et al. [1970]. Theoretical prediction given by the first term of Stevefelt formula (see equation (1)) is plotted by a dashed line.

Discussion

The study of CRR process at temperatures below 77 K is a challenging task as other competing processes contribute to the overall deionization process in plasma. Low gas flow velocities have to be used in order to provide more time for the gas to collide and thermalise to the temperature of the flow tube wall. During this time, the diffusion losses and partially also recombination processes take place in the plasma, which effectively lower the electron number density. At lower $n_e$ the effective binary recombination for electron assisted ternary process is comparable with the helium assisted ternary process with corresponding rate coefficient $K_{\text{He}}$ and temperature dependence $T^{-2.5}$ [Bates and Khare,
Concerning argon number densities—at high argon concentrations argon clusters $\text{Ar}_2^+$ can be produced with high binary dissociative recombination rate coefficient ($\alpha_{\text{DR}} \approx 8 \times 10^{-7} \text{ cm}^3\text{s}^{-1}$ according to Okada [Okada and Sugawara, 1993]). On the other hand, at low [Ar] quenching of helium metastables via Penning ionization with argon atoms and charge transfer of $\text{He}_2^+$ ions to argon atoms is not so efficient, thus leaving enough of $\text{He}^+$ and $\text{He}_2^+$ penetrating further into the flow tube. Penning ionization of Ar by collisions with $\text{He}^+$ represents the source of fast electrons, which can influence the overall temperature of the electron ensemble. Collisions of $\text{He}_2^+$ ions with ambient He atoms at low temperatures create heavier helium clusters ($\text{He}_3^+$) [Patterson, 1968] with fast binary dissociative recombination ($\alpha_{\text{DR}} \approx 3.4 \times 10^{-6} \text{ cm}^3\text{s}^{-1}$ according to Gerardo and Gusinow [Gerardo and Gusinow, 1971]). Mentioned processes contribute to the loss of electrons from plasma, thus the experimental conditions need to be determined carefully when one wants to study the CRR process.

**Conclusion**

Measured ternary rate coefficients of CRR in $\text{Ar}^+$ dominated plasma at temperatures down to 50 K show a good agreement with the theoretical predictions.

The predicted strong temperature dependence ($T^{-4.5}$) holds over the temperature range 50–300 K giving values of the CRR coefficient in the range of many orders of magnitude.

Further study of the collisional radiative recombination in $\text{Ar}^+$ dominated plasma at temperatures below 77 K is currently in progress.

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**References**