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Dependence of the dissociative recombination coefficient of molecular ions Kr_2^+ with electrons on the electron and gas temperatures

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Abstract
This paper presents the measured values of the dissociative recombination coefficient of molecular Kr_2^+ ions with electrons α (Kr_2^+) as a function of both electron and gas temperatures. The measurements were made using a dual-mode (waveguide-cavity) microwave X-band apparatus. The recombination coefficient can be expressed as $\alpha_{Kr_2^+}(T_{gas}, T_e) = 1.4 \times 10^{-6}(T_{gas}/300\,K)^{-0.97}(T_e/300\,K)^{-0.53}$ ($cm^3\,s^{-1}$, K) in the temperature ranges $300\,K \leqslant T_{gas} \leqslant 500\,K$ and $300\,K \leqslant T_e \leqslant 19\,000\,K$.

1. Introduction

Krypton plasmas at moderate and high gas pressures are very often used as various light sources or in lasers based on noble gas and noble gas–fluoro halogen mixtures. At these higher pressures the dominant ions are molecular krypton ions Kr_2^+, which are produced via three-body ion conversion processes. These charged molecular krypton ions Kr_2^+ neutralize due to fast dissociative recombination processes with electrons. This process can be described by the formula

$$Kr_2^+(v) + e \Leftrightarrow (Kr_2)^*_{unstable} \rightarrow Kr^*(i) + Kr \rightarrow Kr + Kr + h\nu + \text{kin.energy}, \quad (1)$$

where the superscript + and * refer to the ionized and excited states, respectively. Generally, molecular krypton ions Kr_2^+ can be in different vibrational states v and the resulting atoms in various excited states Kr(i), which emit specific spectral lines corresponding to the transition of excited atoms in various states i to the ground state. Experimental studies of dissociative recombination in noble gas plasmas over several decades have been reviewed by Ivanov [1] and for some plasmas of active or technical gases by Adams [2]. More than 60 years of continued experimental and theoretical studies have revealed the importance of this charge neutralization process (dissociative recombination) for many diatomic, triatomic and cluster ions in a wide variety of planetary, laser, laboratory or industrial plasmas. The latest reviews [3, 4] and a book [5] were published recently.

To date, a few papers have been devoted to the measurement of the dissociative recombination coefficient of molecular krypton ions Kr_2^+ with electrons and its electron or gas temperature dependences. These results together with our own are summarized in table 1.

The first measurement of the recombination coefficient of Kr_2^+ ions with electrons was undertaken by Richardson [6] in 1952. These results were affected by the presence of a considerable amount of xenon gas, which was determined by measuring the atomic xenon spectra in the late afterglow period of a pulsed discharge. Similar recombination rate values were obtained by Lennon and Sexton [7] and they also reported the presence of nearly 0.5% of xenon gas. More precise measurements were made by Oskam and Mittelstadt [8] mainly from the point of view of krypton gas purity. They used krypton gas containing less than 2.5×10^{-3}% of xenon gas because

Table 1. Comparison of current measurement values of the recombination coefficient of Kr_2^+ ions with electrons with previously published values.

α_0 ($cm^3\ s^{-1}$)	T_{gas} (K)	T_e (K)	γ_{gas}	ξ	Pressure (kPa)	Method of measurement	Method of heating	Reference
$(6–12) \times 10^{-7}$	300	300	—	—	0.8–3.3	Microwave cavity, optical spectroscopy	None *Note*: impurity	Richardson [6]
$\leqslant 11 \times 10^{-7}$	300	300	—	—	0.4–5.2	Microwave cavity	None *Note*: 0.5% Xe	Lennon and Sexton [7]
$(12 \pm 1) \times 10^{-7}$	300	300	—	—	0.8–5.8	Microwave cavity	None	Oskam [8]
42×10^{-7}	800–2500	800–2500	1.5 ± 0.1		0.47	Double probe	Shock wave	Cunningham and Hobson [10]
$>10^{-7}$	300	>300		0.5	0.1–0.3	Microwave cavity	Microwave	Nemeček [30]
1.46×10^{-7}	300–476	300–476	1.5		0.8–2.8	Microwave cavity	Electric oven	Zábudlá [29]
16×10^{-7}	300	300–8400	—	0.55	∼1.3	Microwave cavity, optical spectroscopy	Travelling microwave	Shiu and Biondi [11]
14.8×10^{-7}	300	300–21 300	—	0.53	0.4–1.6	Microwave cavity	Travelling microwave	Mikuš [12]
13.6×10^{-7}	300	300–21 300	—	0.53	0.4–1.6	Microwave cavity	Travelling microwave	Our values re-evaluated from [12]
14×10^{-7}	300–476	300–20 000	0.97	0.53	0.8–2	Microwave cavity	Travelling microwave	Our current results

it was purified by means of the cathaphoretic segregation method [9].

Cunningham and Hobson [10] using a pressure-driven shock tube technique with double-probe measurement of electron density were the first to determine the gas temperature dependence of the recombination coefficient of molecular krypton ions Kr_2^+ with electrons. In their case the electron temperature T_e, the gas temperature T_{gas} and the ion temperature T_{ion} were equal, i.e. $T_e = T_{gas} = T_{ion} = T$. The values of the temperature dependence of the measured recombination coefficient followed a $T^{-1.5}$ variation. Surprisingly, the first measurement of the electron temperature dependence of the dissociative recombination coefficient of Kr_2^+ ions with electrons was simultaneously and independently performed by Shiu and Biondi [11] and by Mikuš [12]. Both measurements showed that the variation of these coefficients with electron temperature can be described as

$$\alpha_{Kr_2^+}(T_e) \sim \alpha_{Kr_2^+}(300\,K)T_e^{-0.55} \quad \text{or}$$
$$\alpha_{Kr_2^+}(T_e) \sim \alpha_{Kr_2^+}(300\,K)T_e^{-0.53},$$

respectively, where $\alpha_{Kr_2^+}(300\,K)$ is the measured value of the recombination coefficient at 300 K (approximately at room temperature). Both measured values of $\alpha_{Kr_2^+}(300\,K)$ are in very good agreement. Therefore, the electron temperature dependence of the recombination coefficient can be expressed as

$$\alpha_{Kr_2^+}(T_e) = \alpha_{Kr_2^+}(300\,K)\left(\frac{T_e}{300\,K}\right)^{-0.54\pm0.01}. \qquad (2)$$

It is seen that the variation of the electron (and also of the gas) temperature dependences of the dissociative recombination coefficients for Kr_2^+ ions is similar, as in the case for argon and neon gases. In this paper, we present the first experimental results of the measurement of the dissociative recombination coefficient for krypton gas, when we vary both the electron and gas temperatures simultaneously and independently, as was carried out in our previous papers devoted to neon [13] and argon [14] gases.

2. Approximations to 'temperature' dependence of dissociative recombination coefficients

The theory of dissociative recombination of molecular ions with electrons has been studied and reviewed by several authors (e.g. [1, 5, 15–20]), who have used semi-classical wave or quantum mechanics. The simplest theory assumes this process as two-body collisions under a certain set of conditions. This process is characterized by the dissociative recombination coefficient, which should vary with the electron temperature T_e according to the law (e.g. [15, 17])

$$\alpha(T_e) = \alpha(300\,K)\left(\frac{T_e}{300\,K}\right)^{-0.5}, \qquad (3)$$

where α (300 K) is the value of the recombination coefficient at gas temperature 300 K.

If it is assumed that the neutral gas temperature T_{gas}, ion temperature T_i, rotational temperature T_r and vibrational temperature T_v are equal, i.e. $T_{gas} = T_r = T_v = T_i$, and that for molecular ions a Boltzmann distribution of vibration states with temperature T_v is valid, then the effective recombination coefficient as a function of electron and gas temperatures can be expressed as (e.g. [18])

$$\alpha(T_e, T_v) = \frac{\sum_{v=0} \alpha_v(T_e) \exp\left(-\frac{hf}{kT_v}\right)}{\sum_{v=0} \exp\left(-\frac{hf}{kT_v}\right)}, \qquad (4)$$

where hf/k measures the vibrational spacing, k is the Boltzmann constant, h is the Planck constant, f is the de-excitation frequency and α_v is the recombination coefficient for molecular ions in a vibration state v.

Assuming that the molecular ion is in the ground vibrational state, i.e. $v = 0$ and the recombination rate coefficient $\alpha_0 = \alpha$ (300 K), then we can obtain the simplest model, which is called the GVS model (ground vibrational

state model) [18] and for T_g larger than hf/k we can write equation (4) as

$$\alpha(T_e, T_g) \approx \alpha(300\,\mathrm{K}) \cdots \left(\frac{T_e}{300\,\mathrm{K}}\right)^{-\xi} \left(\frac{hf}{kT_g}\right) (\mathrm{cm}^3\,\mathrm{s}^{-1},\ \mathrm{K}). \tag{5}$$

This theory has been developed to explain the shock tube measurements of dissociation recombination only in Ar_2^+ (GVS model) or Ne_2^+ ions (LVS: low vibrational state model). Very good agreement with experimental results has been obtained [19].

The theory developed by Bardsley [20] for various potential energy curves using the configuration interaction method was applied to the calculation of the dissociative recombination coefficients of Ne_2^+ and NO^+ ions. His quantitative theory predicts the simple dependence of the recombination coefficients on electron temperatures $\alpha(T_e)$ and also on the electron and gas temperatures $\alpha(T_e, T_{gas})$ using a modified equation (4). Bardsley and Biondi [17] tried a more flexible approach (in order to allow both electron and ion temperatures to vary independently):

$$\alpha(T_e, T_g) \sim \alpha(300\,\mathrm{K}) T_e^{-\xi} T_g^{-\gamma}, \tag{6}$$

where ξ and γ can vary as

$$0.2 \leqslant \xi \leqslant 1.5 \qquad \text{and} \qquad -0.3 \leqslant \gamma \leqslant 1. \tag{7}$$

The very recent and detailed *ab initio* study of the dissociative recombination of He_2^+, Ne_2^+ and Ar_2^+ ions with electrons was carried out by Orel and co-workers (see references in [5]). They calculated potential energy curves and resonance parameters by means of electron scattering calculations using the complex Kohn variation method and molecular quantum chemistry techniques. They were able to calculate cross sections, recombination rate coefficients and their temperature dependences, as well as the branching fractions of the resulting atomic fragments. Their calculations more or less agree with the results of experiments.

It is worth mentioning that none of these theories was applied to the dissociation recombination of molecular krypton ions Kr_2^+. In this paper, we discuss whether our experimental results agree with any of the existing theories.

3. Experimental method and apparatus

We have used the method of stationary afterglow of a dc pulsed discharge to study the recombination processes. A dual-mode microwave afterglow apparatus in the X-band for the simultaneous measurements of electron and gas temperature dependences of the recombination coefficient was developed and described in more detail in our previous papers [13, 14, 21]. The time behaviour of electron density n_e $(\vec{r},\ t)$ during the afterglow period of inert gases under simplified conditions and moderate pressures can be described by the continuity equation

$$\frac{\partial n_e(\vec{r}, t)}{\partial t} = D_a \nabla^2 n_e(\vec{r}, t) - \alpha n_e^2(\vec{r}, t), \tag{8}$$

where D_a is the ambipolar diffusion coefficient and α is the recombination coefficient.

The ambipolar diffusion coefficient D_a can be experimentally determined from the linear slope of the electron density decay curve in the late period of the afterglow (using a semilogarithmic scale: electron density is on the logarithmic scale and time on linear scale). From D_a the electron temperature T_e can be computed and compared with the values calculated for the incident microwave power (11). The electrons were heated by microwaves travelling continuously through the waveguide asymmetrically joined to the mantle of the open cylindrical cavity in which the discharge tube was placed. From this junction the microwaves travelled to both sides of the discharge tube. The power not absorbed by the discharge was absorbed in the matched loads at both ends of the open cylindrical cavity [21].

The calculation of electron temperature as a function of the incident microwave power is made using several simplifying assumptions: (a) the electron–neutral collision frequency ν_{en} is much less than the angular frequency ω of the heating microwave field, (b) the electron energy distribution is Maxwellian and (c) the open cylindrical microwave cavity is regarded as a circular waveguide oscillating in the non-resonant TE_{11} mode. Then the electron temperature can be expressed as

$$T_e = T_g + \frac{2}{3}\frac{e^2\langle E^2\rangle}{m_e k \delta \omega^2}, \tag{9}$$

where T_e and T_{gas} are the electron and gas temperatures, respectively, e and m_e are the electron charge and mass, respectively, k is the Boltzmann constant, $\delta = 2m_e/m_a$ and m_a is the neutral atom mass, ω is the angular frequency of the microwave heating power and $\langle E^2\rangle$ is the average electric field in the waveguide given by the relation [22]

$$\langle E^2\rangle = \frac{P}{\eta\sqrt{\left[1 - \left(\frac{\lambda}{\lambda_{crit}}\right)^2\right] S}}, \tag{10}$$

where P is the microwave power, λ is the wavelength of the microwaves in free space, λ_{crit} is the critical wavelength for the circular waveguide, S is the area of the waveguide cross section and η is a parameter depending on the waveguide properties, and in our case is equal to $1.26 \times 10^{-3}\ \Omega^{-1}$.

When all the calculations are carried out the calculated electron temperature for krypton gas at high pressure (greater than 0.5 kPa) can be expressed as

$$T_e = T_{gas} + 28.2P\ (\mathrm{K, mW}), \tag{11}$$

where T_e and T_{gas} are given in K and P in mW.

These calculated values of electron temperature were compared with the values measured in other experiments. The agreement between them was very good. These values of T_e were used to calculated the ambipolar diffusion coefficient and then to compute the electron density decay rate curves described by equation (8) for different parameters. For this purpose, we have developed our original computer program

[23] based on programs of Gray and Kerr [24] and Frommhold *et al* [25] for an infinite and a finite cylindrical geometry of the discharge tube, respectively. These computed curves were fitted to the experimentally observed curves and in this way the measured values of recombination coefficients were determined.

We measured the electron density by means of a cylindrical microwave cavity [26, 27] in the TM_{011} mode. The cavity was formed as a cylinder from a molybdenum plate with thickness 0.4 mm, which allowed us to heat it up to 650 K. The inner diameter of the open cavity was 25.5 mm and its height 200 mm. The inner radius of the quartz discharge tube was 21.5 mm.

The resonance frequency of the cavity was around 8.6 GHz. The quality factor of the cavity together with the discharge tube was around 1000 and did not change significantly with the electric oven temperature up to 650 K. A cutaway view of the open cylindrical cavity (in the TM_{011} mode) together with waveguide sections is shown in our previous papers [13, 14, 21]. Our dual-mode X-band microwave afterglow apparatus, by which we are able to measure the decay of electron density and simultaneously and continuously heat the electrons during their decay by microwave power up to 900 mW, is also described there.

Special attention was given to the vacuum apparatus and krypton gas purity. An ultimate pressure of the order 10^{-5} Pa was attained by means of rotary and oil diffusion pumps. The quartz discharge tube was permanently connected to a high vacuum system and could be heated to about 950 K for up to 24 h. To decrease the amount of active gas in commercially available spectroscopic pure krypton gas to a negligible value we evaporated a barium getter (used in TV picture tubes—SAES product) on the inner walls of a sphere (like a cold trap) in the vicinity of the discharge tube. Before filling the discharge tube the krypton gas was finally purified by means of the cathaphoretic segregation method [9] in a separate discharge tube to decrease the xenon impurity.

3.1. Experimental results

First, we re-evaluated our previous published results [12] by means of the numerical solution of the continuity equation of electron density using our computer program [23]. The re-evaluated values are somewhat lower and the temperature dependence of the electron–ion recombination coefficient can be written as

$$\alpha_{Kr_2^+}(T_e) = 13.6 \times 10^{-7} \left(\frac{T_e}{300\,\mathrm{K}}\right)^{-0.53} \mathrm{cm^3 s^{-1}, K} \qquad (12)$$

for electron temperatures from 300 to 21 300 K, and krypton gas pressures in the range 0.4–1.6 kPa.

As the second step we measured the electron density in the afterglow period of a dc glow discharge as a function of the electron temperature increased by using microwave power and for different gas temperatures (300, 370, 426 and 476 K). To evaluate the dissociative recombination coefficient from the measured curves we need to know the absolute value of electron density. Therefore, we calibrated our cylindrical open

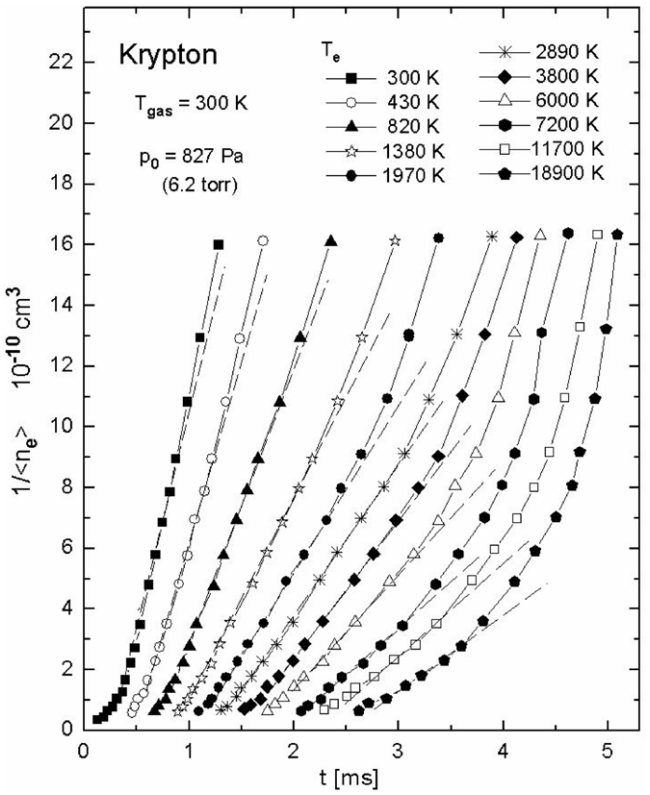

Figure 1. Reciprocal of average electron density ($1/\langle n_e \rangle$) as a function of afterglow time (t) for different electron temperatures, where the krypton gas pressure is 827 Pa and the gas temperature 300 K (zero time points of the individual curves are shifted by 0.2 ms for clarity).

TM_{011} microwave cavity by means of several long dielectric rods as we have shown in [28]. The following relation is valid for the average electron density:

$$\langle n_e \rangle = 213 \Delta f. \qquad (13)$$

The electron density is in $\mathrm{cm^{-3}}$ and the frequency shift is in Hz.

It is well known that only when the afterglow period is controlled by recombination and the electron and gas temperatures are equal is the solution of continuity equation (8) simple. In this case, the reciprocal value of electron density $1/n_e$ increases linearly with time and the slope of this straight line gives the value of the recombination coefficient. Therefore, the results of measurements are often presented as the reciprocal of electron density versus time. A few typical decay curves are shown in figure 1 for the gas temperature of 300 K and 11 values of electron temperatures (from 300 to 18 900 K). The krypton gas pressure was 827 Pa. The other results are shown in figure 2 for the gas temperature of 426 K and ten values of electron temperatures (from 426 to 19 000 K).

Similar results were obtained at krypton gas temperatures of 370 and 476 K. As can be seen in both figures, the experimental decay curves increase not strictly linearly with time, which is a result of the increasing importance of ambipolar diffusion. In particular for higher electron temperatures they show a large curvature. The numerical solution of equation (8) provides a satisfactory fit to the measured data.

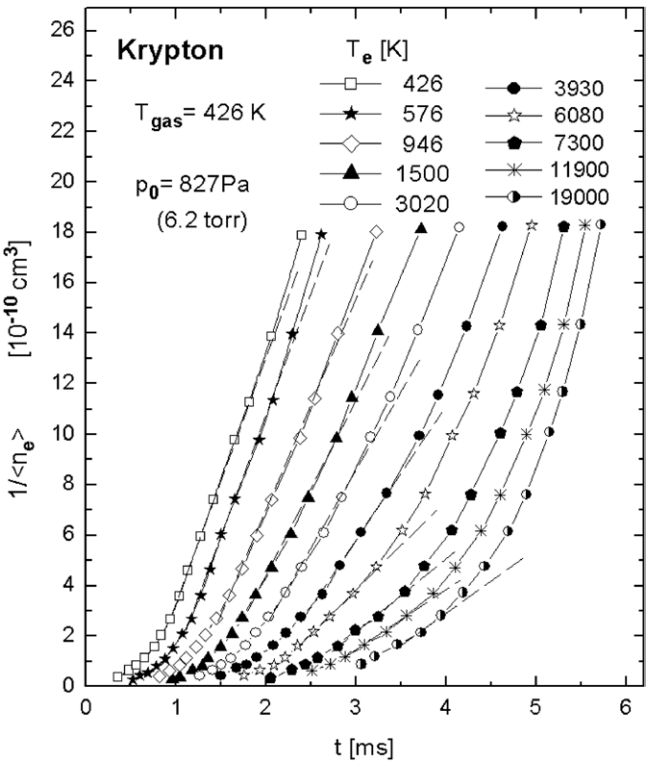

Figure 2. Reciprocal of average electron density ($1/\langle n_e \rangle$) as a function of afterglow time (t) for different electron temperatures, where the krypton gas pressure is 827 Pa and the gas temperature 426 K (zero time points of the individual curves are shifted by 0.2 ms for clarity).

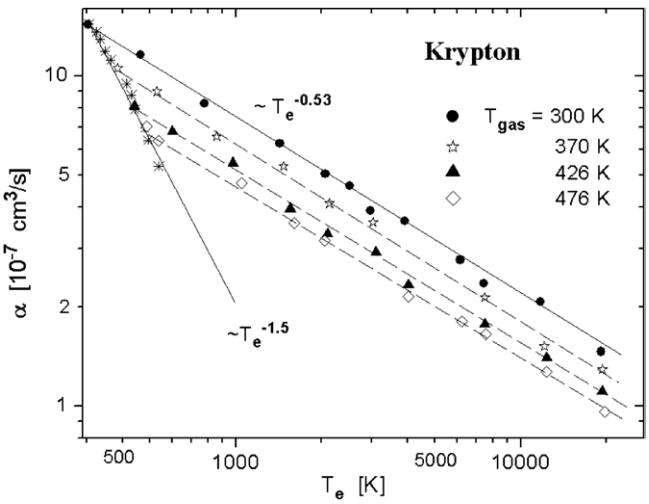

Figure 3. Dissociative recombination coefficients of molecular ions Kr_2^+ with electrons, as a function of electron and gas temperatures.

The values of the dissociative recombination coefficient for molecular krypton ions Kr_2^+ with electrons $\alpha(Kr_2^+)$ evaluated from the data shown in figures 1 and 2 (and others) are presented as a function of T_e for four values of T_{gas} in figure 3.

It can be seen that a simple power law of electron temperature dependence as $\alpha_{Kr_2^+}(T_e) \sim \alpha_{Kr_2^+}(T_{gas}) T_e^{-0.53}$ fits all of the pure krypton data very well. This dependence is in very good agreement with that shown by Shiu and Biondi [11] and Mikuš [12]. When $T_e = T_{gas} = T$ the dissociative recombination coefficient decreases as $T^{-1.5}$ for temperatures up to 500 K. This dependence is in good agreement with the GVS model developed by O'Malley *et al* [19] for argon and with our previous measurements [29].

Eliminating the electron temperature dependence of the recombination coefficients, we can describe the gas and electron temperature dependence in the form proposed by Bardsley and Biondi [17] as

$$\alpha_{Kr_2^+}(T_e, T_{gas}) = \alpha_{Kr_2^+}(T_{gas}) \left(\frac{T_e}{300\,\mathrm{K}}\right)^{-0.53} \mathrm{cm^3\,s^{-1}, K} \qquad (14)$$

or

$$\alpha_{Kr_2^+}(T_e, T_{gas}) = 14 \times 10^{-7} \left(\frac{T_{gas}}{300\,\mathrm{K}}\right)^{-0.97} \times \left(\frac{T_e}{300\,\mathrm{K}}\right)^{-0.53} \mathrm{cm^3\,s^{-1}, K} \qquad (15)$$

for the range $300\,\mathrm{K} \leqslant T_{gas} \leqslant 500\,\mathrm{K}$ and $300\,\mathrm{K} \leqslant T_e \leqslant 20\,000\,\mathrm{K}$.

In table 1 we summarize the previously measured data of the recombination coefficient together with their gas or electron temperature dependences for molecular krypton ions with electrons. Our results for the value of $\alpha(Kr_2^+)$ agree very well with those obtained by Shiu and Biondi [11] and with their results of the electron temperature dependence of the recombination rate coefficient. It is interesting that our results for the gas temperature dependence of $\alpha(Kr_2^+)$ agree very well with the shock tube results of Cunningham and Hobson [10] but for different gas temperature intervals. It is surprising that no theoretical calculation has been made, according to our knowledge. The presented results agree well with the model presented in [17]. The errors of our measurements are less than about 10%. These errors arise in measurements of resonant frequency of the cavity and time.

The dissociative recombination process is multichannel. Each channel is characterized by the partial recombination coefficient and the recombination products of atoms in excited and ground states. Richardson [6] and Shiu and Biondi [11] observed from optical spectroscopy excited Kr atoms in the 5p state (Paschen notation). Barrios *et al* [31] showed by measurements of the centre-of-mass kinetic energy release with the time-of-flight (TOF) technique that the primary excited atom products were in the 5s states, which correspond to the spectral line in the UV region. It is expected that the ratio between atomic fragments can change with electron or gas temperature. Therefore, the simultaneous measurement of the time behaviour of spectral line intensities under different gas discharge conditions can give us more important information about both temperature dependences of the recombination coefficient.

4. Conclusions

In this paper, we have investigated simultaneously the electron and gas temperature dependence of the dissociative recombination coefficient of molecular ions Kr_2^+ with electrons using a dual-mode X-band microwave apparatus in the

afterglow of a dc glow discharge. Our measured value of α_0 at gas room temperature is in good agreement with the results of Shiu and Biondi [11]. The same is true for the electron temperature dependence up to $T_e = 2 \times 10^4$ K while the gas temperature dependence is similar to that measured by Cunningham and Hobson [10] for a gas temperature higher than 500 K.

We were able to show that the temperature dependence of the dissociative recombination coefficient fits well with the proposal of Bardsley and Biondi ([17] (see (6)) for restricted ranges of the gas and electron temperatures.

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