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COLLISIONAL IONISATION OF EXCITED RUBIDIUM ATOMS

M. Chéret and L. Barbier

Centre d'Etudes Nucléaires de Saclay, Service de Physique des Atomes et des Surfaces, 91191 Gif-sur-Yvette Cedex, France

Résumé - Dans une vapeur de rubidium, la mesure des courants d'ions Rb^+ , Rb_2^+ et Rb^- a permis de caractériser les processus collisionnels de formation d'ions à partir d'atomes excités et de déterminer les coefficients de taux d'ionisation associés à ces réactions.

Abstract - Ion current measurements for Rb^+ , Rb_2^+ and Rb^- ions allowed to characterize ionizing collisional processes involving excited atoms in a Rb vapour. Rate coefficients for these reactions are determined.

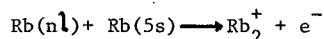
When an alkali vapor is irradiated with high power laser beams a complex medium is created and it is not very easy to separate laser fields effects from purely collisional mechanisms. It is thus important to understand first the involved collisional reactions.

Low power lasers allow to study this processes in a simple medium.

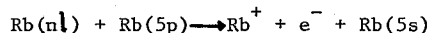
Two C.W. multimode dye lasers crossing at the centre of a cylindrical cell are used to pump, in a first step rubidium atoms in a resonant level $\text{Rb}(5p)$ and in a second step in a highly excited level $\text{Rb}(nl)$ ($l=s$ or d). The experimental set up has been described previously /1/. Typical experimental conditions correspond to: $p=10^{-4}$ Torr, $\text{Rb}(5s)=10^{12} \text{ cm}^{-3}$, $\text{Rb}(5d) = 10^{10} \text{ cm}^{-3}$, $\text{Rb}(nl)= 10^8 \text{ cm}^{-3}$.

The main processes are:

Hornbeck-Molnar associative ionization

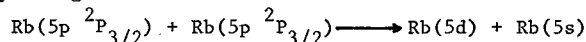


Penning atomic ionization :

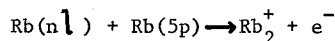


Beside these reactions the following processes have also been observed :

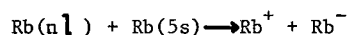
Energy pooling reaction :



Penning associative ionization :



Ion pair formation



Results concerning energy pooling reaction have been published for the Rb case /2/ and M. Allegrini presents a review on this subject at the present conference.

The other reactions correspond to ionizing mechanisms involving highly excited Rb atoms. Ions are analyzed with a mass spectrometer wired to alternately detected positive or negative ions. A typical result of mass scanning is presented in fig.1. The Rb^- current is 5000 times lower than the Rb_2^+ or Rb^+ current and consequently ion pair formation does not contribute significantly to the Rb^+ current. Photoionisation by the laser beams : $\text{Rb}(nl) + h\nu \longrightarrow \text{Rb}^+ + e^-$ is an important mechanism contributing to the Rb^+ current. The cross sections for this process are well known /3/ and the photoionisation contribution is used to determine the excited state densities /1/.

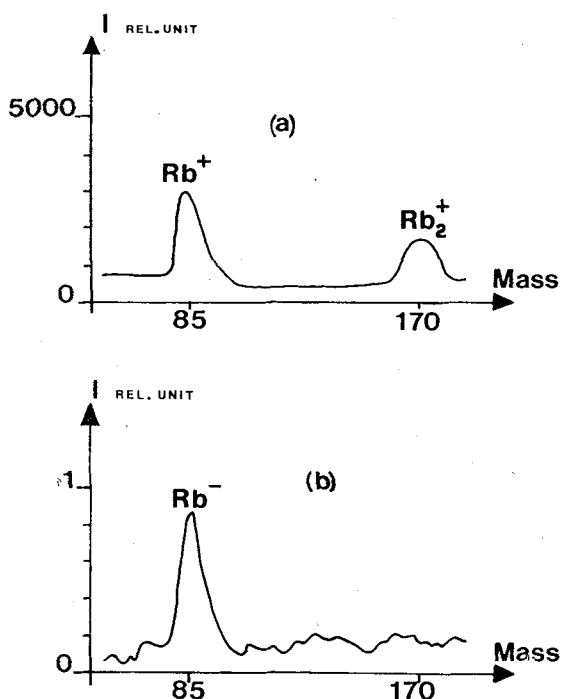
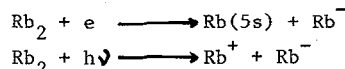


Fig.1 - Mass scanning of positive (a) and negative ions for the excitation of the $6d\ 2D_{5/2}$ level.

second one is the exchange theory; Kimura for Rb atoms found rate coefficients Γ independent lying in the range $10^{-8}\text{ cm}^3\text{ s}^{-1}$ /4/. It is then important to determine the l dependency of the Penning rate coefficient.

This has been done by populating the excited states with pulsed lasers. As an example if the $7s$ level is excited its concentration decays exponentially and populates the $6p$ level. 200 ns after the laser shot the $6p$ density becomes higher than the $7s$ density (fig.2). If one of the Penning rate coefficient for $7s$ or $6d$ level is strongly predominant the Rb^+ current will vary as the $7s$ or as the $6p$ density. If the two levels have equal rate coefficients the Rb^+ current will then decay as the sum of the $7s$ and $6d$ densities. Experimental variation (points fig.2) accounts for this last hypothesis. The same results have been obtained when the $5d$, $6d$ or $8s$ level is excited : experimental results clearly show that the Penning rate coefficient is l independent. The rate coefficients obtained with the C.W. system then can be corrected from cascading radiative mechanisms. The results are presented in fig. 3 together with the dipole-dipole calculations for $s(x)$, $p(o)$, $d(+)$ levels and Kimura's calculations (\bullet) for levels $6d$, $8s$ and $7d$.

As presented in fig.1 mass scanning of negative ions indicates the presence of a low Rb^- current. Molecules are commonly considered as responsible for negative ion formation /5/, the main reactions being :



The present paper only reports on Penning atomic ionisation and ion pair formation. The Penning ionization occurs via collision between a highly excited atom and a resonant state atom. In fact when a highly excited state is pumped the radiative cascading deexcitation populates the neighbouring levels. s and d levels essentially decay to p levels and consequently the studied medium is composed of the excited s or d level and of the various lower p levels. The total density of these p levels represents 80% of the density of the pumped s level and 20% of the pumped d level. The measured quantities correspond to the contribution of the pumped state and of the radiatively populated p levels. When a s state is populated the p level densities alone can explain the experimental observations /1/ - l dependency of the rate coefficients will be known only if the p contribution is correctly deduced.

Measured rate coefficients lie in the range $10^{-8}\text{ cm}^3\text{ s}^{-1}$. Theoretically two mechanisms are usually considered. The first one is the dipole-dipole interaction which give rate coefficients in the range $10^{-9}\text{ cm}^3\text{ s}^{-1}$ for d or p levels and in the range $10^{-10}\text{ cm}^3\text{ s}^{-1}$ for s level. The

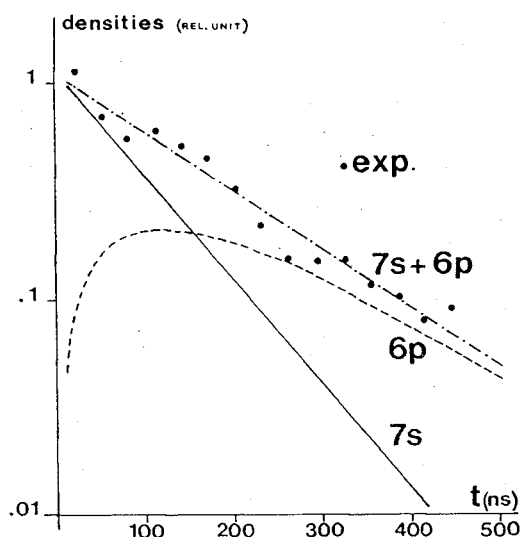


Fig.2 - Excitation of the 7s level. Time variation of the 7s and 6p densities calculated with measured lifetime. • Experimental points deduced from the Rb^+ current.

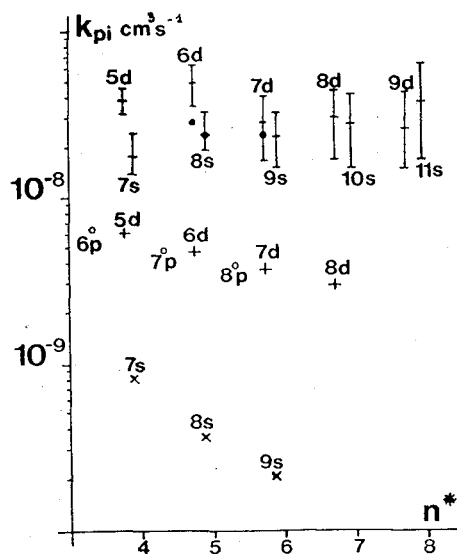
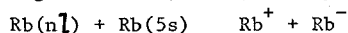


Fig.3 - Rate coefficient for Penning ionization k_{pi} against the effective quantum number n^* of the excited state.

Under our experimental conditions (cell temperature 450°K) the ratio between the dimer density and the atom density is equal to 4×10^{-6} . Experimental checks show that :

- negative ion formation requires both lasers to be resonant.
- negative ion current does not vary when the Rb_2 concentration is increased by a factor of 10 (by varying the cell temperature at constant Rb pressure).
- negative ion current varies linearly with both laser powers.

These observations rule out reactions involving any of the following particles : molecules, molecular ions, photons, electrons. Ion pair formation by a collision between a highly excited atom and a ground state atom, namely



is a reaction that fulfills all our experimental checks. This reaction involves the same partners $\text{Rb}(n\text{l})$ and $\text{Rb}(5\text{s})$ than the Hornbeck-Molnar associative ionization. The rate coefficient for ion pair formation, $k^-(n\text{l})$, can be deduced from the rate coefficient for associative ionization $k_{AI}(n\text{l})$ by comparing the negative ion current $I(\text{Rb}^-)$ to the molecular ion current $I(\text{Rb}_2^+)$:

$$k^-(n\text{l}) = k_{AI}(n\text{l}) \frac{I(\text{Rb}^-)}{I(\text{Rb}_2^+)}$$

When the $6d \ ^2D_{5/2}$ level is excited, the cell temperature varying between 383°K and 453°K at constant Rb pressure, the $k^-(6d)$ values, reported on fig.4 are measured. As the electron affinity of the Rb atom is equal to $3919.2 \text{ cm}^{-1}/4$ the reaction is exoergic by 1082.4 cm^{-1} for the $6d \ ^2D_{5/2}$ level. The

experimental values of the rate coefficient can be fitted with a cross section σ^- equal to $1.3 \times 10^{-16} \text{ cm}^2$ above the threshold and equal to zero below this limit and with a maxwellian velocity distribution $f(v)$ being assumed, leading to

$$k^- = \int_0^\infty \sigma^-(v) v f(v) dv.$$

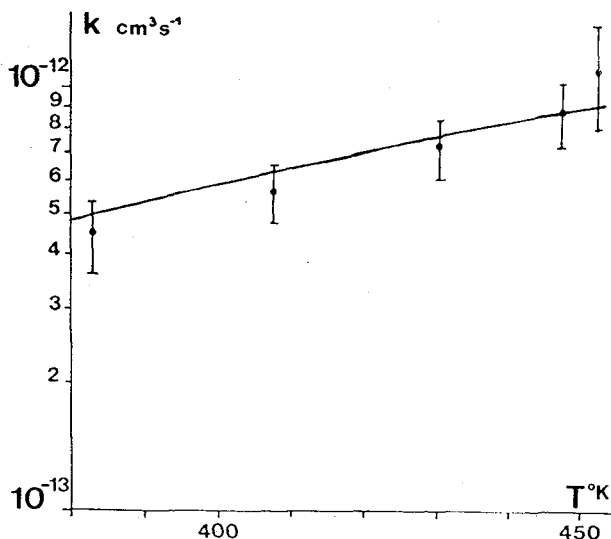


Fig.4 - $k(6d \ ^2D_{5/2})$ against cell temperature
experimental points
fitted curve with $\gamma = 1.3 \times 10^{-16} \text{ cm}^2$.

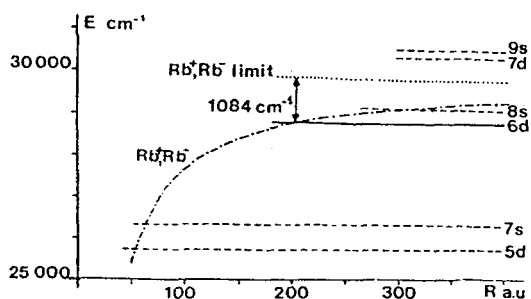


Fig.5 - Rb^+, Rb^- coulombic curve calculated with an electron affinity equal to 3919.2 cm^{-1} .

This cross section value is not theoretically explained. A Landau-Zener calculation /5/ at the crossing point between the unperturbed atomic curve and the coulombic Rb^+, Rb^- ion pair curve (fig.5) give a negligible contribution : it is not surprising as the crossing point (203 a.u) is passed diabatically.

Negative ion current measurements have been performed on levels lying energetically under (5d, 7s, 6d, 8s) or above (7d, 9s) the Rb^+, Rb^- limit. In fig.6 are plotted the $k(nl)$ values against the energy defect of the reaction. The corresponding cross section are reported in fig.7.

As for the 6d level the Landau-Zener calculation at the crossing point between the atomic curve and the ionic curve does not explain these results for levels under the Rb^+, Rb^- limit. For levels above such a calculation is not possible. More sophisticated theoretical approaches are then needed. One may consider crossing points at shorter distances obtained by splitting the excited level /6/ or by manifold crossings in the molecular system (fig.8). Another possibility is a delocalised calculation as applied by Sidis /7/ to the H^+, H^- system. (fig.9) These various methods require first the knowledge of the molecular potential curves.

For the first time ion pair formation have been observed in an excited vapor. As the exit channel for the reaction (i.e. the Coulombic Rb^+, Rb^- curve) is well known this process appears appealing to check the other involved molecular potential curves.

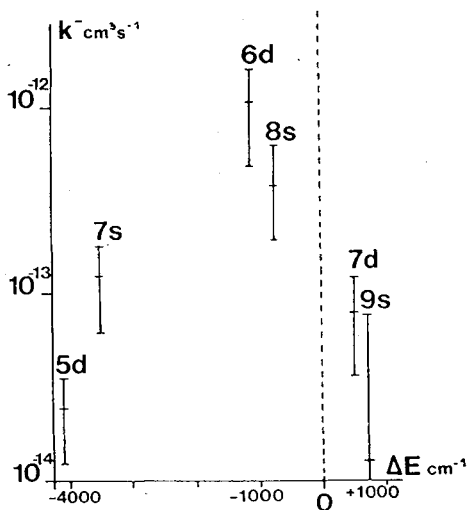


Fig.6

rate coefficient (fig.6) and cross section (fig.7) for ion pair formation versus the energy defect of the reaction.

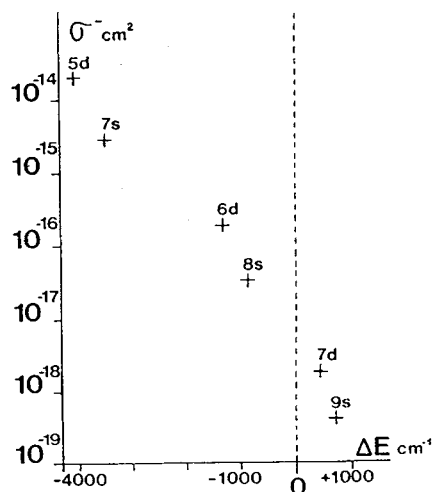


Fig.7

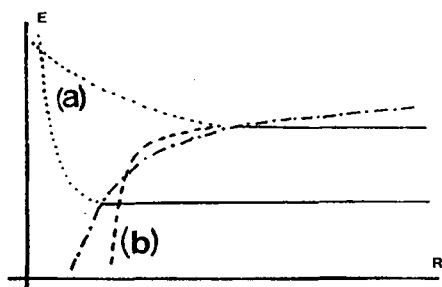


Fig.8 - (a) splitting of the atomic curve
(b) crossing in the molecular system.

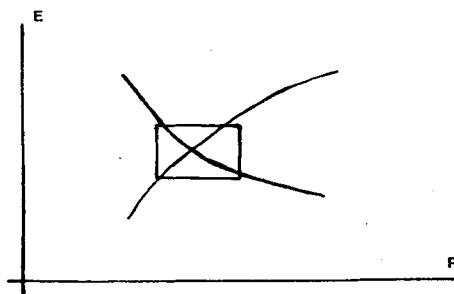


Fig.9 - Delocalised scheme in
the molecular system.

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