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LIFETIME AND COLLISIONAL DEPOPULATION CROSS SECTION FOR THE $14\,^2D_{3/2}$ STATE OF Rb*

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The lifetime τ and the cross section σ for depopulation induced by collisions with ground-state Rb atoms were measured for the $14\,^2D_{3/2}$ state of Rb. The experiment was performed in the gas-cell conditions in a range of Rb vapour temperatures in the vicinity of 340 K. The lifetime value τ = 1970(130) ns, agrees with the theoretical prediction with allowance for the influence of blackbody radiation as well as of the effects due to core polarizability and spin–orbit interaction. The measured cross section $\sigma = 6.5(3.1) \times 10^{-12}$ cm^2 is close to the geometrical cross section. This agrees with similar observations made by other authors for the case of the lower $n\,^2D$ states of Rb.

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1. Introduction

Lifetime values and cross sections for collisional depopulation of Rydberg states of alkali metal atoms are of interest as a contribution to data bases for spectroscopy and quantum optics as well as for other fields, like astrophysics and plasma physics.

Lifetimes of rubidium $n\,^2D_J$ states were the subject to several experimental and theoretical studies. The measurements covered all the states with n up to 13 and also the ones with $n = 15$ and $n = 18$ [1–7]. The most comprehensive set of theoretical values (for n up to 18) was delivered by Theodosiou, who calculated lifetimes for a great number of states of alkali metal atoms [8]. In these calculations the effects due to core polarizability, spin–orbit interaction and blackbody radiation were treated. The lifetime for Rb($14\,^2D$), the state of interest to the present work, had been also earlier calculated, within Coulomb approximation method, by Gounand [9].

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Experimentally determined cross sections for depopulation of $Rb(n\,^2D_J)$ states induced by collisions with ground-state Rb atoms are available for n ranging from 5 to 15, with an exception of $n = 14$ [7, 10–17]. In experiments involving different procedures, cross sections for different collisional depopulation processes were obtained: the ones for mixing between the doublet components (cross section σ_{mix}) and/or for quenching out of the doublet (σ_q) or for total depopulation encompassing both mixing and quenching (σ). To our knowledge, no theoretical counterparts have been published for $Rb(n\,^2D_J)$ states of interest. The experimental values are usually compared with respective geometrical cross sections.

In the present paper we report an experiment in which time-resolved fluorescence from the $Rb(14\,^2D_{3/2})$ state was analyzed, thus enabling the lifetime τ and the cross section σ to be deduced for this state.

2. Experimental procedure and arrangement

The excitation- and detection-scheme is sketched in Fig. 1. The $14\,^2D_{3/2}$ state was selectively populated with light pulses in two-photon excitation from the ground state $5\,^2S_{1/2}$. Time-resolved $14\,^2D_{3/2} \rightarrow 5\,^2P_{1/2}$ fluorescence (at 489.6 nm) was observed. The processes of the collisional excitation transfer, (i) within the $14\,^2D$ state (fs mixing) and (ii) out of the $14\,^2D$ state (quenching) are symbolized in Fig. 1 by curved arrows and a wavy one, respectively.

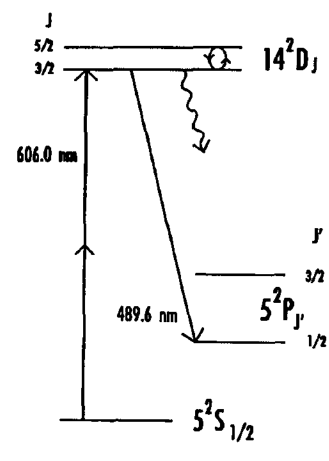

Fig. 1. The excitation- and detection-scheme.

Pulses of exciting light were produced by a dye laser (Lumonics HD 500) pumped by an excimer laser (Lumonics EX 520). The laser beam was directed into a fluorescence cell placed in a double chamber oven. The lower part of the cell contained a droplet of metallic rubidium. This part was maintained at a temperature a few degrees lower than the temperature T of the upper part, where the excitation took place. The cell was subjected to over ten days of baking up at a temperature of ca. 460°C under a vacuum better than 5×10^{-8} torr, before rubidium was distilled into it. System of photon-counting in delayed coincidence was applied to register time-resolved fluorescence signal spectrally filtered with the help of a monochromator. (See Fig. 2.)

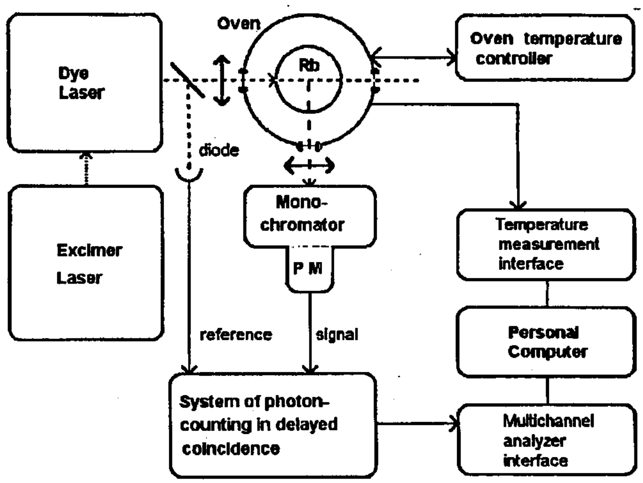

Fig. 2. A block diagram of the experimental arrangement.

Measurements were performed in a range of cell temperatures in order to enable extrapolation to collisionless conditions. The range of temperature variation of the lower part of the cell 49°C to 69.5°C corresponded to pressure range of rubidium vapour 3.2×10^{-6} to 1.9×10^{-5} torr [18].

3. Data analysis and results

It is expected that at low Rb vapour pressure, as in the case of this experiment, the temporal-development of the $14\,^2D_{3/2}$ state population and thus of the fluorescence from this state can be approximated by a single-exponential function with the decay parameter λ being the sum of the radiative decay rate $\Gamma = 1/\tau$ and the collisional depopulation rate γ. The rate γ can be expressed as $\gamma = \sigma N v$, where σ is the thermally averaged collisional cross section, $v = (8kT/\pi\mu)^{1/2}$ is the mean relative velocity of the colliding atoms, μ being their reduced mass, and N is the number density of the ground-state Rb atoms, Nv is defined by cell temperatures.

$$\lambda = \Gamma + \gamma = \frac{1}{\tau} + Nv\sigma. \tag{1}$$

A sequence of λ values was obtained by a least-squares-fitting of the exponential function to the experimental decay curves. The plot of λ versus Nv (the Stern–Volmer plot) is given in Fig. 3 together with a straight line fitted to the points. The lifetime of the $14\,^2D_{3/2}$ state of Rb is obtained as an inverse of λ extrapolated to $Nv = 0$. The slope determines the cross section σ.

In Table I(a) the measured lifetime and the relevant theoretical values of Gounand [9] and Theodosiou [8] are gathered. Theodosiou has given lifetimes for

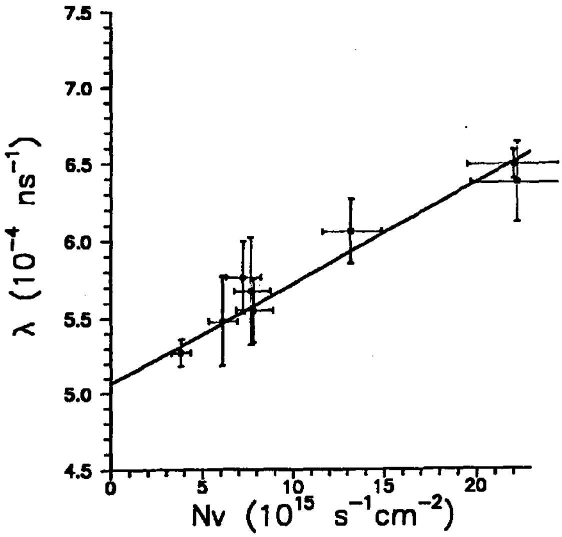

Fig. 3. Decay constant λ plotted against Nv for Rb($14\,^2D_{3/2}$) and the straight line fitted to the points (see text).

TABLE I

Results of the present experiment and relevant theoretical values: (a) lifetime (in nanoseconds), (b) cross section for total depopulation induced by Rb–Rb collisions, and geometrical cross section.

a)

	This experiment	Theory			
		Theodosiou [8]			Gounand [9]
Temperature / State	327–348 K	0 K	350 K	460 K	0 K
$14\,^2D_{3/2}$	1970(130)	2359	2038	1937	2910
$14\,^2D_{5/2}$		2275	1978	1884	

b)

	Depopulation cross section σ this experiment	Geometrical cross section σ_{geom}
$14\,^2D_{3/2}$	$6.5(3.1) \times 10^{-12}$ cm^2	5.73×10^{-12} cm^2

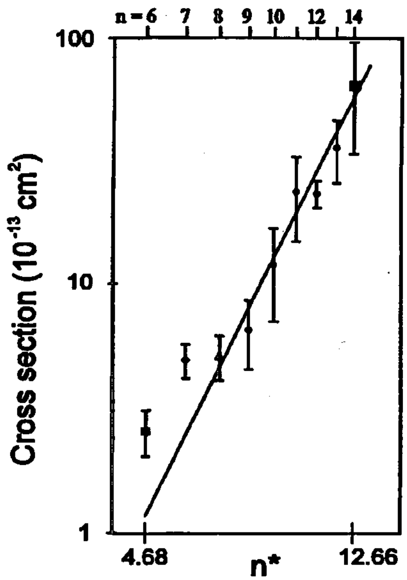

Fig. 4. Cross section for total depopulation of Rb($n\,^2D_{3/2}$) induced by collisions with ground-state Rb atoms, plotted in a log–log scale against effective quantum number n^*. The notation is ■ — σ, this work; • — σ, Ref. [10]; full triangles — ($\sigma_{\rm mix}+\sigma_{\rm q}$), Ref. [15]; full diamonds — ($\sigma_{\rm mix}+\sigma_{\rm q}$), Ref. [14]; and □ — ($\sigma_{\rm mix}+\sigma_{\rm q}$), Ref. [16]. The full curve represents n^* dependence of the geometrical cross section.

three different temperatures of blackbody radiation. The lifetime measured in the present work, attributed to the mean temperature of ca. 340 K, agrees within the uncertainty limits with the interpolated Theodosiou's result. By comparison with Gounand's theoretical value calculated by using Coulomb approximation, this seems to prove that for this state, calculations beyond Coulomb approximation are necessary. In Table I(b), together with the experimentally determined collisional cross section σ, the geometrical cross section $\sigma_{\rm geom}$ for the collisions between Rb(14D) and Rb(5S) atoms is given. As a measure of $\sigma_{\rm geom}$ the following formula was used:

$$\sigma_{\rm geom} = \pi\langle(r_{14D}+r_{5S})^2\rangle \cong \pi(\langle r_{14D}^2\rangle + 2\langle r_{14D}\rangle\langle r_{5S}\rangle + \langle r_{5S}^2\rangle), \tag{2}$$

with mean radii $\langle r\rangle$ and mean square radii $\langle r^2\rangle$ from hydrogenic expectation values [19]:

$$\langle r\rangle = n^{*2}\left\{1+\frac{1}{2}\left[1-\ell(\ell+1)/n^{*2}\right]\right\}a_0, \tag{3}$$

$$\langle r^2\rangle = \frac{1}{2}n^{*2}\left[5n^{*2}+1-3\ell(\ell+1)\right]a_0^2, \tag{4}$$

n^* is the effective principal quantum number, ℓ — the orbital angular momentum quantum number and a_0 — the Bohr radius. As can be seen, our experimental value is quite well reproduced by $\sigma_{\rm geom}$. In Fig. 4 total depopulation cross section data known from the literature for other Rb($n\,^2D_{3/2}$) states and the result of this work are plotted against n^*. The full curve represents n^* dependence of the geometrical cross section. Those values from the literature are generally also close to the respective geometrical cross sections.

In our experiment the contributions to σ from fs mixing ($14\,^2D_{3/2} \rightarrow 14\,^2D_{5/2}$) and quenching have not been evaluated separately. For a range of caesium $n\,^2D_{\mathcal{J}}$ states Pendrill has shown that at low Cs vapour density the rate of fs mixing collisions exceeds significantly the rate of any other inelastic collisional processes [20]. As mentioned in Sec. 1, for some rubidium $n\,^2D_{\mathcal{J}}$ states, both experimentally determined cross sections σ_{mix} and σ_{q} are available [14–16]. Although these σ_{q} values are smaller than the respective σ_{mix} values, they are not negligible. Therefore it seems reasonable to hold that also in depopulation of the rubidium $14\,^2D_{3/2}$ state quenching plays a significant role and that the obtained cross section presented in Table I(b) characterizes total collisional depopulation of $14\,^2D_{3/2}$, comprising $14\,^2D_{3/2} \rightarrow 14\,^2D_{5/2}$ mixing as well as quenching of $14\,^2D_{3/2}$ resulting in population of other states than $14D$.

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