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AUTOIONIZATION LIFETIMES IN LASER EXCITED Na₂ RYDBERG STATES

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In our laboratory we are studying Rydberg states of the Na₂ molecule by optical -optical double resonance. We have already analysed the rotational interactions by means of the Multichannel Quantum Defect Theory (M.Q.D.T.) [1]. The aim of this paper is to summarize another part of our work concerning the dynamical effects and the autoionization process which governs our experimental method.

Experimental results

Our experimental set-up is as follows : a supersonic sodium beam is crossed at right angle by two superimposed counterpropagating tunable dye lasers pumped by the harmonics of a pulsed YAG laser. The first dye laser selects a well defined level (v', J') of the $A^1\Sigma_u^+$ state of Na₂. The second one is tuned to the autoionizing Rydberg states (nd^1A_g). A small electric field (about 1 V/cm) collects the produced electrons which are then detected through an electron multiplier. The electron current is recorded in a micro-computer through a boxcar integrator with a variable gate aperture time.

This experimental set-up allows to measure lifetime of each line, at least for lifetimes greater than the laser pulse duration (about 6 ns). Indeed, the recorded signal S is proportional to the integral of the electron current during the boxcar gate aperture

$$S \propto \int_0^T \frac{P_0}{\tau} e^{-t/\tau} dt = P_0(1 - e^{-T/\tau})$$

where T is the gate aperture time, τ and P_0 are respectively the lifetime and the initial population of the Rydberg state. Moreover this method offers the possibility to show directly all the lifetimes of each line of a spectrum by varying T . Figure 1 shows recorded spectra for two values of the integration time. The intermediate state is the $A^1\Sigma_u^+$ ($v'=2, J'=7$) level. The observed levels are then $v=2$ (due to FCF), $J=6, 7$ and 8 (respectively P, Q and R lines) converging toward the $v^+=2$ limit of the ion. The spectra, recorded respectively with a 200 ns and a 10 ns integration time, show clearly two different regions. The $\Delta v=1$ region (between the $v^+=1$ and $v^+=2$ levels of the ion) where autoionization is very strong leading to lifetimes of the order of the ns. And the $\Delta v=2$ region (between the $v^+=0$ and $v^+=1$ levels) where the lines correspond to unresolved complex resonances : the autoionization occurs either directly toward $v^+=0$ levels or indirectly through $v=1$ Rydberg series. Thus the observed lifetimes are due to interferences between the first direct path, with $\Delta v=2$, and the second, stepwise, with two $\Delta v=1$ transitions. This leads to possible stabilization of the Rydberg states and, actually, lifetimes are very long near the $v^+=1$ limit as can be seen from the figure and from

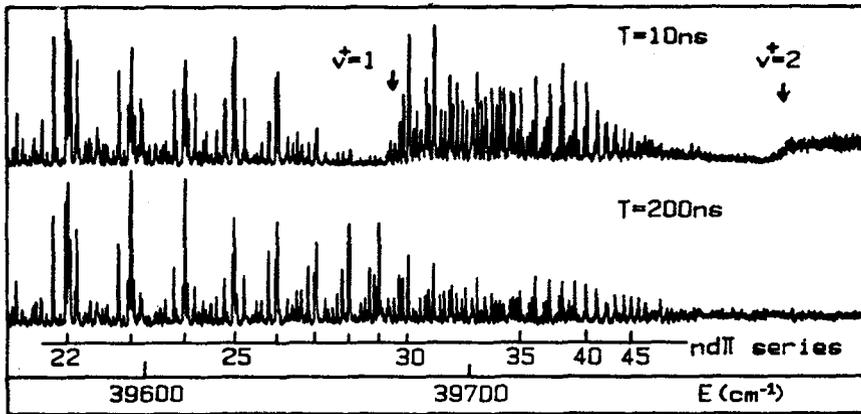


Fig. 1. Experimental spectra of autoionizing Rydberg states of Na₂ ($v^+=2, J^+=7$) for two values of the integration time. Upper : 10 ns, lower : 200 ns. The lifetimes effects are clearly seen on the upper spectrum.

Table 1. These effects become more pronounced as Δv increases but lifetimes are always of the same magnitude and the processes are fundamentally the same.

Interpretation

As shown by Jungen [2] in the case of the H₂ molecule, the M.Q.D.T. is the best suited method to deal with our problem. In order to take into account both rotational and vibrational interactions, we have to introduce quantum defects as functions of the internuclear separation ($\mu_A(R)$). The reaction matrix used in the M.Q.D.T. treatment becomes

$$R_{\nu^+ N^+, \nu'^+ N'^+} = \sum_{\Lambda} \langle N^+ | \Lambda \rangle \left(\int \chi_{\nu^+} \tan \Pi \mu_A(R) \chi_{\nu'^+} dR \right) \langle \Lambda | N'^+ \rangle$$

n	P line	Lifetime (ns)	
		Q line	R line
22	11	17	13
23	10	22	9
24	10	20	16
25	9	15	11
26	10	20	10
27	60	57	42
28	200	180	67
29	-	250	-

Table 1. Lifetime measurements in the $\Delta v=2$ autoionization region(ndΠ series). Lifetimes increase rapidly just below the $v^+=1$ limit ($n=29$).

F (V/cm)	n_c	$n (\tau > 5 \text{ ns})$
3	50	48
5	45	40
10	39	37

Table 2. Electric field dependence of the lifetime in the $\Delta v=1$ region. n_c is the critical value of the principal quantum number where complete Stark mixing occurs ($n_c^3 F = \text{cste}$). n is the experimental value above which the lifetime is greater than 5 ns.

The relevant quantities $\mu_{\Lambda}(R)$ are derived from vibrational perturbations on energy levels. However this derivation leads to no more than a first order expression of μ_{Λ} as a function of R . So, as can be seen from the expression of the reaction matrix, the $\Delta v=1$ process will be treated precisely while the $\Delta v=2$ process needs some improvements. This is partly why the quantitative agreement between experiment and theory (Fig. 2) is far from being good. In fact we have to distinguish two cases. First, the $\Delta v=2$ process which is very sensitive to the parameters due to the interference effects. In that case, except just below the $v^+=1$ limit, the agreement is reasonably good. Secondly, the $\Delta v=1$ process, where only the first order well known values of $\mu_{\Lambda}(R)$ are needed and where the lifetime is expected to be proportional to n^3 . So, in that case, discrepancies should be interpreted in another way. Since an electric field (at least 1 V/cm) is always present in the experiment, field induced stabilization of the levels arises. This field mixes the observed and levels with the high angular momentum hydrogenic complex (Stark mixing) which does not interact with the core thus increasing the lifetimes. Since the magnitude of the Stark mixing is proportional to the strength of the field F and varies as n^5 with the energy, this phenomenon becomes drastic for high n values where the lifetime calculated to be of the order of 1 ns is at least 10 times greater. The field dependence of this phenomenon is clearly shown in Table 2. This process could also explain the great discrepancy between experimental and calculated lifetimes for the levels located just below the $v^+=1$ limit. Indeed, the stepwise process through $v=1$ series becomes less probable because it is concerned with intermediate levels of very high principal quantum numbers. It is worth noticing that this effect is just the opposite of that observed for rotational autoionization, where the field is found to induce autoionization toward J^+ values of the ion forbidden in the absence of an electric field [3].

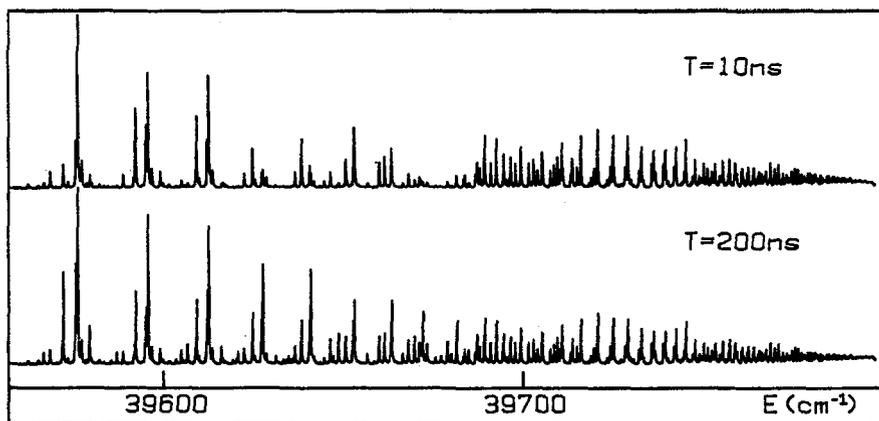


Fig. 2. Theoretical spectra corresponding to Fig. 1. Lines positions agree very well with experiment but lifetimes agree only qualitatively.

- [1] M.C. Bordas, M. Broyer, J. Chevalere, P. Labastie and S. Martin, *J. Physique* **46** (1985) 27 and ref. therein
- [2] Ch. Jungen and Dan Dill, *J. Chem. Phys.* **73** (1980) 3338.
- [3] Ch. Bordas, P. Brevet, M. Broyer, J. Chevalere and P. Labastie, *Europhys. Lett.* **3** (1987) 789.