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Laser-based Measurement of Radiative Lifetimes of $4p^2\ ^3P_{2,1,0}$ States of Calcium

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ABSTRACT. We have measured radiative lifetimes of $4p^2\ ^3P_{2,1,0}$ states of calcium using time resolved laser spectroscopy. The states were populated by stepwise excitation using a combination of pulse modulated tunable dye laser and a weak d.c. discharge. Measured lifetimes τ for these states are $\tau_1\ (^3P_2) = 6.6\ (4)$ns, $\tau_2(^3P_1) = 5.4\ (4)$ns and $\tau_3(^3P_2) = 7.3\ (7)$ns.

One and two electron atoms provide the important testing ground for the techniques of atomic physics. Excited states of alkaline earths (Mg, Ca, Sr and Ba) with two valence electrons exhibit dramatically different features compared to the alkalies. First of all, the coupling between the two electrons has a very significant effect on the properties of the excited states. Secondly, there are strong perturbations of the normal 4sn1 Rydberg series due to inner-shell excited configurations such as 3d4p, $4p^2$ and 5sn1 terms. The basic energy level structure of these atoms has been well established. With the advent of lasers, the experimental accuracy has improved to such an extent that it is possible to resolve and measure Rydberg states very close to the ionisation limit.

Using stepwise excitation with lasers, natural radiative lifetimes of long Rydberg series in Ba (Bhatia *et al.* 1981 and Aymar *et al.* 1982) and Sr (Grafstrom *et al.* 1983 and Jonsson *et al.* 1984) have been determined. In the case of Ba, lifetime values alongwith stark shifts and g-factors have been used to probe state mixing and coupling between the two valence electrons (*see* Gallagher *et al.* 1981 and Grafstrom *et al.* 1982). However, stepwise excitation of calcium poses a special problem. The first step excitation from the $4s^2\ ^1S_0$ ground state to the first excited 4s4p ^3P terms corresponds to the intercombination transition with a low probability whereas the first allowed transition from the ground state to the 4s4p $^1P_0$ requires

laser wavelength in the blue region (422.9nm) where the dyes are not very efficient. As a result the lifetime data on calcium is not as accurate and extensive as it is for barium and strontium. Recently, Jonsson *et al.* (1984) and Major *et al.* (1985) have reported lifetime values for some of the low lying states of calcium using first step excitation by electron impact in a mild d.c. discharge and by passing a neutral calcium atomic beam through a cold cathode penning discharge. In this paper, we report the measurement for the lifetime of $4p^2\ ^3P_{2,1,0}$ states using a pulse modulated laser spectroscopy technique originally developed by Svanberg group (*see* Gustavasson *et al.* 1979) at Lund in Sweden.

Experimental

The pulse modulated laser spectroscopy (PUMOLS) technique is basically a delayed coincidence method of nuclear physics combined with a pulse modulated dye laser. Various features of PUMOLS are discussed in detail by Gustavsson *et al.* (1979). In this technique, the time distribution of the emitted fluorescence from the excited state of an atom is stored and displayed by a multichannel analyser. Block diagram of our experimental set up is shown in Fig. 1. Atomic beam of calcium was produced by a resistively heated oven containing freshly cut chuncks of spec pure

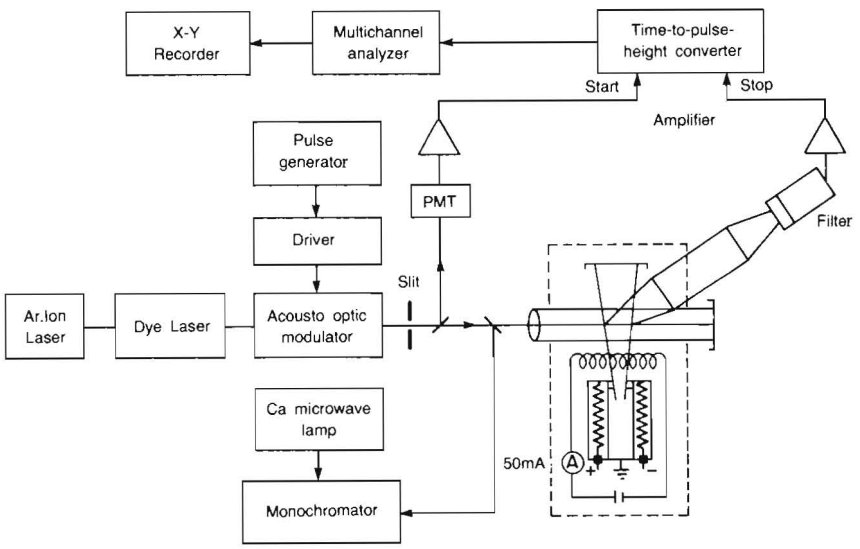

Fig. 1. Experimental set up for the measurement of atomic radiative lifetimes by Pulse Modulated Laser Spectroscopy (PUMOLS) technique

calcium. The design of the oven was similar to the one used by Jonsson *et al.* (1984) and Brinkmann *et al.* (1969). The metastable states of Ca were populated by a mild d.c. discharge at the exit hole of the oven. Attempts were also made to obtain these states by passing the neutral Ca atomic beam from the oven through a cold cathode Penning discharge as described in our earlier paper (Major *et al.* 1985). However, the d.c. discharge produced much stronger and noise free fluorescent signals. The problem of stray light from the discharge and the heater filament can be solved by proper apertures and baffles along the length of the vacuum chamber tube and by keeping the laser and atomic beam interaction region far above the oven. A typical discharge current was 25 mA at 30 volts potential difference. The density of the metastable states can be controlled by the current through the heating element.

For the second step excitation a tunable dye laser (Spectra Physics Model 375) pumped by an Ar ion laser (Spectra Physics Model 71-50) was used. Wavelength range between λ 415 nm-500 nm was covered by stillbene and coumarine dyes obtained from the Exciton Co. (Germany). All line output from the Ar ion laser was used to pump the dye laser. The typical input power into the dye laser was 8 watt. Tuning of the dye laser was monitored by a monochromator and was compared with an electrodeless calcium lamp excited by a microwave discharge. Line width of the dye laser as specified by the commercial intra-cavity etalon was 0.01 nm. The output from the laser was modulated at a repetition frequency of 0.01-05 MH_z using an acousto-optic modulator (SORO MAR-50). The pulse shape and duty cycle were controlled by HP-8013 pulse generator. An RCA 8852 photomultiplier fitted with a narrow band interference filter was used to detect the fluorescence at right angle to the atomic and the laser beam. The time between the start pulse taken directly from the pulse generator and the stop pulse by the arrival of the first fluorescent photon is measured. The decay time curves were recorded by Ortec pulse timing and analysis system consisting of time to amplitude convertor (TAC), timing filter amplifiers, constant fraction discriminator and a multichannel analyser. The data were reduced by an HP-45 minicomputer.

Results and Discussion

Partial term diagram of calcium atom illustrating the two step excitation to the $4p^2\ ^3P_{2,1,0}$ states is shown in Fig. 2. Since this configuration gives rise to even parity states it is essential to have an odd parity level as an intermediate state. Considering the transition probabilities and the wavelength limits of the tunable dye laser, it is found that the most suitable intermediate step would be the $4s(^2S)4p\ ^3P_{2,1,0}$ levels. The fine tuning of the dye laser enabled us to investigate lifetimes of the upper levels separately but no J-dependence of the lifetimes of the ^3P states were observed. Except for the $^3P_0$ state, the wavelength monitored for the fluorescence was always different than the excitation wavelength. Alternately a broad band excitation from the intermediate levels to the upper levels was used,

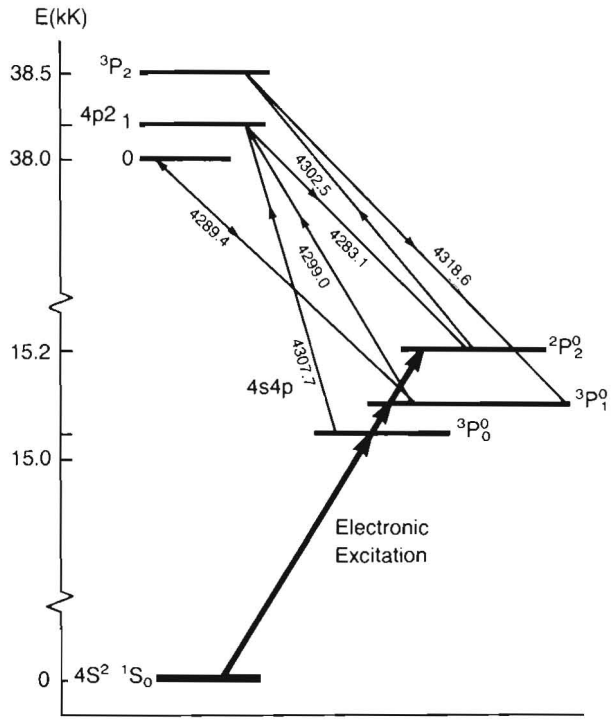

Fig. 2. Partial energy level diagram of neutral calcium

and the total decay curve recorded by the multichannel analyser was analysed using deconvolution programme with an HP-45 computer. In this method the excitation pulse profile and the equipment response function I(t) was first measured by reflecting part of the incident laser pulse using a clean quartz plate. Then, the total response function F(t) was measured. The true response function can be obtained from the deconvolution integral,

$$F(t) = \int_0^t f(t') \cdot I(t-t')dt$$

using the decay in the exponential form so that

$$f(t) = f_0\ e^{-t/\tau}$$

The best computer fit for F(t) was obtained when f(t) is expressed as

$$f(t) = f_1\ e^{-t/\tau_1} + f_2\ e^{-t/\tau_2} + f_3\ e^{-t/\tau_3}$$

This indicated the simultaneous decay of three different levels even though the three lifetimes were not very much different as indicated earlier by the method of selective excitation. The excitation pulse and the decay curve for $^3P_2$ state is given in Fig. 3, and the measured lifetimes for the three states are listed in Table 1. The stated errors shown in the brackets include both the statistical scattering and estimated systematic errors. A critical evaluation of the numerical results obtained by the deconvolution programs was made, with particular concern for their sensitivity to the precise form of the exictation function and level and form of the background. Further experimental tests to define the limits of systematic errors are desirable. Therefore, pending a more thorough analysis, the error limits indicated are probably more than the expected inherent accuracy of the system.

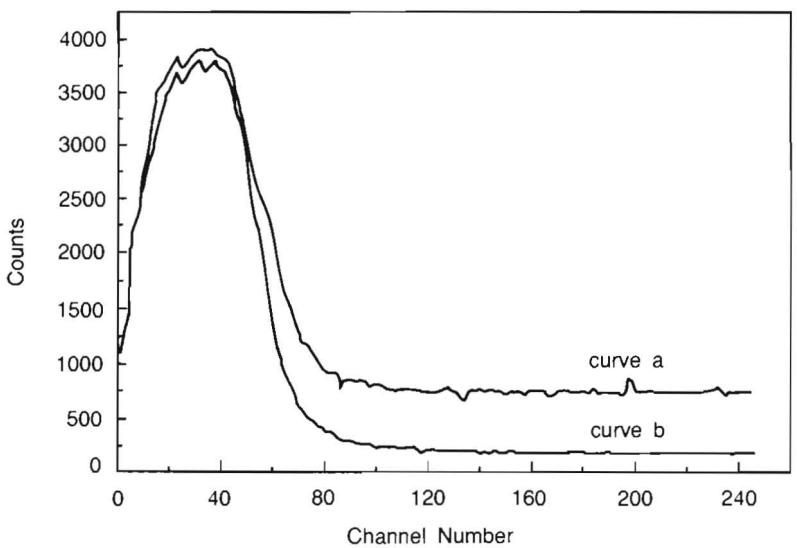

Fig. 3. Plots of data obtained on the lifetime of $4p^2\ ^3P_2$ state. Curves a and b correspond to the fluorescence and excitation functions respectively

Table 1. Lifetime values for $4p^2\ ^3P_{2,1,0}$ states

State	λ excitation	λ monitored	τ (this work)	Other
τ_1 ($^3P_2$)	4302.5	4318.6A°	6.6 (4)	–
τ_2 ($^3P_1$)	4299.0	4283.1	5.4 (4)	–
τ_3 ($^3P_0$)	4307.7	4307.7	7.3 (7)	–
τ ($^3P_{2,1,0}$)	–	Broad band decay	6.8 (6)	6.9(4*)

* Value quoted by Jonsson *et al.* (1984) Physica Scripta.

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قياس الأعمار الاشعاعية للمستويات $4P^2\ {}^3P_{2,1,0}$ في الكالسيوم باستخدام الليزر

ك. س. باهاتيا و م. مرافي

قسم الفيزياء ـ جامعة الكويت ـ ص. ب (٥٩٦٩) ـ الكويت

تم قياس الأعمار الاشعاعية لمستويات الكالسيوم $4P^2\ {}^3P_{2,1,0}$ المثارة باستخدام طيف الليزر المميز زمنياً. وقد زودت المستويات بالذرات (أهلت) بطريقة الاستثارة المتعاقبة وبمرحلتين وذلك باستخدام نبضة متغيرة من ليزر صبغيّ، علاوة على استخدام التفريغ الكهرستاتيكي الضعيف. حيث تم الحصول على الأعمار كما يلي:

العمر الاشعاعي للمستوى $^3P_2$ يساوي (٤) ٦,٦ نانو ثانية.

العمر الاشعاعي للمستوى $^3P_1$ يساوي (٤) ٥,٤ نانو ثانية.

العمر الاشعاعي للمستوى $^3P_0$ يساوي (٧) ٧,٣ نانو ثانية.