An efficient method for measuring atomic and molecular lifetimes using a modulated or deflected CW dye laser beam

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AN EFFICIENT METHOD FOR MEASURING ATOMIC AND MOLECULAR LIFETIMES USING A MODULATED OR DEFLECTED CW DYE LASER BEAM

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By pulse modulating or deflecting a CW dye laser beam, cascade-free lifetime determinations at high spectral resolution can be performed with the delayed-coincidence technique using measuring times of about 1 minute. The method is illustrated by lifetime measurements for the $7^2P_{3/2,1/2}$ levels of cesium.

Pulsed tunable lasers provide a selective and efficient population of excited atomic and molecular states. Lifetimes of such states can be measured by observing the exponential decay of the laser-induced population with analog techniques using a fast transient recorder or a boxcar integrator. However, the accuracy attainable in measurements of this kind is limited by non-linearities in the photo-multiplier-tube response. The delayed-coincidence method, where the time from the laser pulse till the detection of the first fluorescence photon is measured, does not suffer from this limitation.

On the other hand, this method is inherently slow as the probability to detect a fluorescence photon must be kept low to avoid an excessive pile-up correction [1]. Thus, the measurement of a fluorescence decay curve generally requires quite a long time (one hour or more) if pulsed lasers of normal repetition rates (10—500 Hz) are used (see e.g. ref. [2]). Furthermore, advantage cannot be taken of the high peak power of the laser pulses, on the contrary, the intensity may have to be reduced to keep the excitation rate low. In order to make delayed-coincidence measurements of excited-state lifetimes more efficient, we have used a pulse-modulated or a swiftly deflected CW dye laser beam to produce short pulses at a high repetition rate, recognizing the fact that the low peak power in the pulses does not represent a serious limitation in this kind of measurements. This method for fast, cascade-free determinations of lifetimes at a high spectral resolution has been demonstrated for the $7^2P_{3/2,1/2}$ levels of $^{133}$Cs

A train of short pulses can be generated from a CW laser in several ways. By using mode-locking (internal modulation) the output power of the laser can be favourably concentrated into the pulses. With a dye laser, very short pulses can be produced ($<1$ ns) at a repetition rate determined by the cavity length [3]. Generally the pulses will then be too closely spaced for typical excited state lifetimes. The slicing out of pulses from the pulse-train as well as the operation of the mode-locked dye laser over wide wavelength regions is complicated. For general applications we have instead used an acousto-optic modulator in the external dye laser beam or have quickly spatially scanned the laser beam across a narrow slit using an ultra-fast rotating mirror.

We have used a four-side titanium prism mounted on a dentist’s air turbine drill revolving at 7000 turns/s, thus producing 28000 pulses/s. This technique has recently been employed by Rigler and Ehrenberg in connection with fluorescence relaxation studies of macro-molecules in liquids [4]. With commercially available rotating prisms, repetition rates up to 120000/s are possible and pulse-lengths below 1 ns are attainable. The short pulse-length possible with a prism is very important for measuring high-frequency quantum beats. An acousto-optic modulator has most other advantages and was used in most of our measurements. It allows a convenient matching against lifetime $\tau$ of pulse-width ($\sim 3 \tau$ for efficient pumping) and time between pulses ($\sim 10 \tau$ for a detailed study of the decay and an optimum duty cycle). We used a SORO M-AR 50 acousto-optic modulator operating at a car-

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$^{\dagger}$ First results were presented at the Nordic Symposium on Atomic and molecular transition probabilities, Lund, Sweden (March 28, 29, 1977).
rier frequency of 250 MHz. A laboratory pulse generator is used to pulse-modulate the carrier producing the periodic density variations diffracting the beam. The rise-time of the optical pulses is about 10 ns and the optical efficiency of the device about 80%. With a sinusoidal modulation it can also be used directly for phase-shift measurements of lifetimes (see e.g. ref. [5]).

Due to the narrow laser line width a highly selective excitation is obtained and very closely spaced energy levels can be investigated without blending perturbations. This can also be achieved by using an efficient electron-beam excitation in conjunction with a large analyzing spectograph as applied by Erman et al. [6]. Laser excitation, however, does not give rise to a repopulation of the studied state through cascades from higher lying levels. A high spectral resolution is very important in studies of molecular states. A selective excitation is especially valuable in measurements on atoms, where lots of cascades otherwise occur.

In fig. 1 our experimental setup for lifetime measurements employing an acousto-optic modulator is shown. The laser pulses excite an atomic beam and ordinary delayed-coincidence electronic units are used. For a demonstration of the power of the method the lifetimes of the $7^2P_{3/2,1/2}$ levels in Cs were determined. A coumarine 47 CW dye laser, pumped by the UV lines of an argon-ion laser was running at 4555 or 4593 Å in multi-mode operation (linewidth ~1 Å) at a power level of about 50 mW. A repetition rate of about 1 MHz was used. Count rates of 1000/s were typically obtained and could be further increased by single-mode operation or at least narrowing down the laser line width with an intracavity etalon to achieve a better spectral match to the narrow absorption profile of the atomic beam. The count rate obtained with this method should be compared with the one typical with ordinary pulsed lasers (1–10/s). As the probability of a detected photon following an individual laser pulse is still very small in the present method no pile-up correction needs to be applied.

In fig. 2 decay curves for the $7^2P_{3/2}$ and $7^2P_{1/2}$ states are shown, each sampled for one minute. By varying the atomic beam density over wide ranges it was shown that multiple scattering, causing apparently longer lifetimes, was absent in our measurements. For the lifetimes of the two studied states we obtained $\tau(7^2P_{3/2}) = 136(4)$ ns and $\tau(7^2P_{1/2}) = 165(6)$ ns.

The first value is in very good agreement with previous determinations whereas the second one is slightly larger than the literature values (see e.g. Deech et al. [7]).

Preliminary measurements were also performed on

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Fig. 1. Setup for delayed-coincidence measurements using a pulse-modulated CW dye laser beam.
(1) High-speed measurements (repetition rates up to 10 MHz).
(2) Cascade-free measurements.
(3) High spectral resolution (down to 0.001 Å).
(4) Negligible pile-up correction.
(5) Short pulses possible for high-frequency quantum-beat spectroscopy.
(6) Wavelength region 420—1000 nm, step-wise excitations possible.

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