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New method for cascade-free lifetime measurements

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A new method is presented for cascade-free high-precision lifetime measurements of highly excited atomic states. Experimental examples are given for the lifetimes of the Ba^+ $6p\ ^2P_{3/2}$ level and the Ne $3p'[3/2]_2$ level. The experimental arrangement uses a fast ion beam which is excited in a gas cell and, after a definite time of flight, is further excited by a continuous-wave intracavity dye-laser beam. Further possible applications of the method are mentioned.

INTRODUCTION

The selective excitation of atomic states is a well-known principle for cascade-free lifetime measurements and has been successfully applied in several cases. One of these is the optical excitation of atomic beams which was described by Koenig and Ellett[1] in 1932. The same concept, with modern instrumentation, was used by Andrä *et al.*,[2] who excited a fast-ion beam by Doppler-tuned beam laser resonance and then made high-precision time-of-flight measurements in the manner used for beam-foil experiments.[3] The number of problems, however, to which this technique can be successfully applied, is quite limited. Therefore in this paper we discuss and test a combination of broad-band excitation resulting from beam-foil or beam-gas interactions, and selective excitation from populated higher-lying levels using a tunable laser. Such a two-stage excitation technique takes advantage of all the unprecedented properties of beam-foil spectroscopy[4] (BFS) for lifetime measurements, but avoids the serious problem of perturbing cascades. The applicability of this method as well as its advantages and problems are demonstrated for the lifetimes of the Ba^+ $6p^2P_{3/2}$ level and the Ne $3p'[\frac{3}{2}]_2$ level, which have been measured previously.

THEORY

If a broad-band excitation populates higher-lying levels, and after a given time interval, a selective excitation acts on a level n and changes its population density, it is possible to avoid the perturbing cascade effect in lifetime measurements by a simple difference measurement, measuring the intensity decay curve of a transition from the nth level to a lower level with and without selective excitation. The population difference of level n can be calculated for the case of optical excitation, from or to a lower level k, by induced absorption or emission from the rate equations. The integral equations for the level n, with and without laser, result in

$$N_n^1(t) - N_n^0(t) = N_n^d e^{-t/\tau_n}, \tag{1}$$

with $t > t_0 + \delta$, where $N_n^1(t)$ and $N_n^0(t)$ are the laser-stimulated and unstimulated populations and τ_n is the mean life of the level n; t_0 represents the time interval between the two excitations and δ is the interaction time of the atoms with the laser. The quantity N_n^d is determined by the population difference of levels n and k, on which the laser acts at time t_0, as well as by the transition rates of induced absorption and emission W_{kn} and W_{nk}. With the approximation of constant laser power during the interaction time δ we obtain

$$N_n^d = \int_{t_0}^{t_0+\delta} N_k(t') W_{kn} \exp[t'/\tau_n - W_{nk}(t_0+\delta-t')]\,dt' + (e^{-W_{nk}\delta} - 1)\left(N_n(0) + \int_0^{t_0} \sum_{i=n+1}^{m} N_i(t') A_{in} e^{t'/\tau_n}\,dt'\right)$$
$$+ \int_{t_0}^{t_0+\delta} \{\exp[-W_{nk}(t_0+\delta-t')] - 1\} \sum_{i=n+1}^{m} N_i(t') A_{in} e^{t'/\tau_n}\,dt', \tag{2}$$

assuming a set of levels $n+1$ to m, from which the atoms cascade into level n. The spontaneous-transition rate between levels i and n is denoted A_{in}. Since N_n^d is a constant for times $t > t_0 + \delta$, Eq. (1) yields a pure exponential and no longer depends on cascades.

An experimental arrangement for such cascade-free lifetime measurements can be realized using

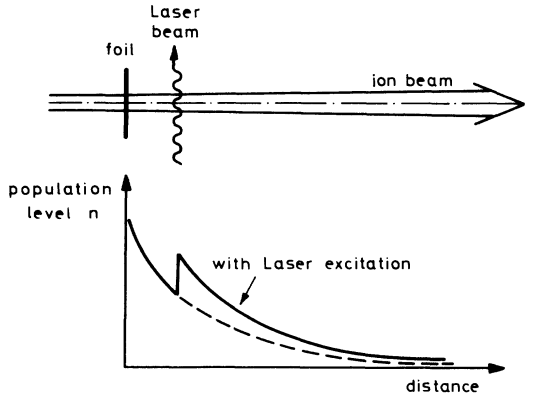

FIG. 1. Principal arrangement of foil-laser excitation and resulting population change of level n as a function of distance after the excitations.

a fast-ion beam which passes through a differentially pumped gas cell or a thin carbon foil and afterwards crosses a laser beam, which is in resonance with an optically allowed transition of the atoms (Fig. 1). The resulting intensity decay curves with and without laser action are measured by a time-of-flight experiment in the manner as used in BFS.

The strength of the detected signal is essentially determined by the efficiency of the laser used and the population difference $N_n(t_0) - N_k(t_0)$ of levels n and k at time t_0. These in turn are influenced by the special broad-band excitation process as well as by the distance between first and second excitation according to different lifetimes and cascades of the levels n and k. The effectiveness of the laser depends on the available laser output power per spectral width and on the resonance width of the atoms. Since the atomic emission lines of fast ions, which have been excited by a gas target or a thin foil, will have a Doppler broadening of about 1 Å or even more according to the divergence of the beam, it is advantageous to have a laser with a spectral width of this order. Then maximum resonance between the incoming ions or atoms and the laser radiation can be obtained.

On the other hand the spectral power densities of such broad-band lasers are in general substantially lower than those of narrow-band lasers, and therefore the transition rates of induced absorption and emission for an individual atom will be reduced.

To calculate the magnitude of W_{kn} and W_{nk} for both types of lasers, the usual expressions found in the literature[5] have to be slightly modified. Since the interaction time of the ions with the laser, which is determined by the velocity of the ions and the width of the laser beam, is typically of the order of 10^{-9} sec, the value of δ is such that $1/\delta \gg 1/\tau_n + 1/\tau_k$. So in first-order perturbation theory for single-mode laser action W_{nk} is found to be

$$W^s_{nk} = (2\pi)^{-1} A_{nk} c\lambda^2 \rho \frac{\sin^2[\frac{1}{2}(\omega_{nk} - \omega)\delta]}{(\omega_{nk} - \omega)^2 \delta^2} \delta, \qquad (3)$$

which in the case of resonance reduces to

$$W^s_{nk} = (8\pi)^{-1} A_{nk} c\lambda^2 \rho \delta, \qquad (4)$$

where c is the light velocity, λ the laser wavelength, ρ the photon density, ω_{nk} the transition frequency from level n to k, and ω the laser frequency. The resonance width of an individual atom with the laser, $\Delta\omega = 2\pi/\delta$, is determined by the duration of laser excitation and is of the order of 10^{-2} Å. Therefore only a small fraction, 1% or less, of all incoming atoms which are in an excited state k or n will be resonant with the laser radiation, and even for very high transition rates W^s_{nk} the population density of level n will not be changed by more than a few parts per thousand.

In contrast to a narrow-band laser, the efficiency of a broad-band laser is essentially dependent on the available laser power, which determines the induced transition rates. For multimode laser operation, where the frequency separation of neighboring modes is small compared to $\Delta\omega$, we obtain

$$W^m_{nk} = (\lambda^4/8\pi\ \Delta\lambda_l) A_{nk} \rho. \qquad (5)$$

When the spectral width of the laser $\Delta\lambda_l$ is limited to about 1 Å and an induced transition rate of the order of A_{nk} is sought, which for typical conditions gives a population change of a few percent, laser power of 200 W/cm^2 is necessary at $\lambda = 6000$ Å. A laser output power of this magnitude in continuous-wave operation in general will make possible a cascade-free lifetime measurement using the proposed double-excitation method.

EXPERIMENTAL ARRANGEMENT

The simplest application of double excitation and following lifetime measurement is given for a level populated selectively from a metastable state. The attainable population difference between upper and lower levels, on which the laser acts, is much more favorable than for shorter-lived states, and the remaining signal from an original excitation, perturbed by cascades, will only be very small. Such a less complicated example of double excitation was chosen for a first application in order to test the apparatus and to optimize the laser excitation conditions.

The experimental arrangement used for a cascade-free lifetime measurement is shown in Fig. 2.

The fast-ion beam passes through a differentially pumped gas cell with He as the target gas. The ions or atoms created by charge exchange are highly excited and radiate in flight. At a distance of 0.6 m they cross a 1-mm-thick laser beam, which is tuned to an optically allowed transition of the ions. In order to increase the spectral power density for a broad-band laser action, the atoms are excited inside the cavity of a continuous wave dye laser. Figure 3 shows the essential components of the folded laser resonator.[6] The laser's active medium, which is pumped by a 6-W all-lines Ar^+ laser, consists of a 0.25-mm-thick dye-solution jet beam (rhodamine-6G in water) as applied and tested by Wellegehausen *et al.*[7] Three prisms inside the cavity allow the selection of the wavelength and reduction of dye-laser bandwidth to about 0.7 Å, while a further reduction can be made by a Fabry-Perot etalon. Prisms 1 and 2 are simultaneously used as windows to the vacuum system necessary for the ion beam. Without an etalon we obtained a dye-laser power inside the cavity of 5 W and more for a wavelength of about 5900 Å.

The detection system for a time-of-flight measurement is also to be seen in Fig. 2. A lens observing the radiating ion beam at an angle of 30°, together with a mirror and an aperture are mounted on a sliding carriage, which can be moved parallel to the ion beam. An additional mirror and lens focus a small element of the beam onto the entrance slit of the monochromator which separates the spectral line of interest from scattering light. In order to assure a difference measurement for a cascade-free signal, the laser is chopped with a frequency of 82 Hz and the signal at the exit slit of the monochromator is detected by a digital lock-in photon counting system. For normalization of the signal a current-frequency converter was used, which works as an external clock for the photon counter and regulated the chopper sampling time in accordance with the ion current. After a specified number of chopper cycles the difference signal of fluorescent radiation resulting only from laser excitation is printed out by a teletype, and the sliding carriage is moved to the next measuring position by a stepper motor.

The exact ion beam velocity, which has to be known for a lifetime measurement, is evaluated by a 90°-electrostatic energy analyzer.[8]

LIFETIME MEASUREMENTS

As a first example of selective broad-band laser excitation and following cascade-free lifetime measurement, the described technique was applied to a 240-keV Ba^+ beam to evaluate the lifetime of the Ba^+ $6p\,^2P_{3/2}$ level (Fig. 4). After populating the $5d\,^2D_{3/2}$ metastable state by beam-gas interactions, the $6p\,^2P_{3/2}$ level is excited by induced absorption from the metastable state, tuning the laser to a

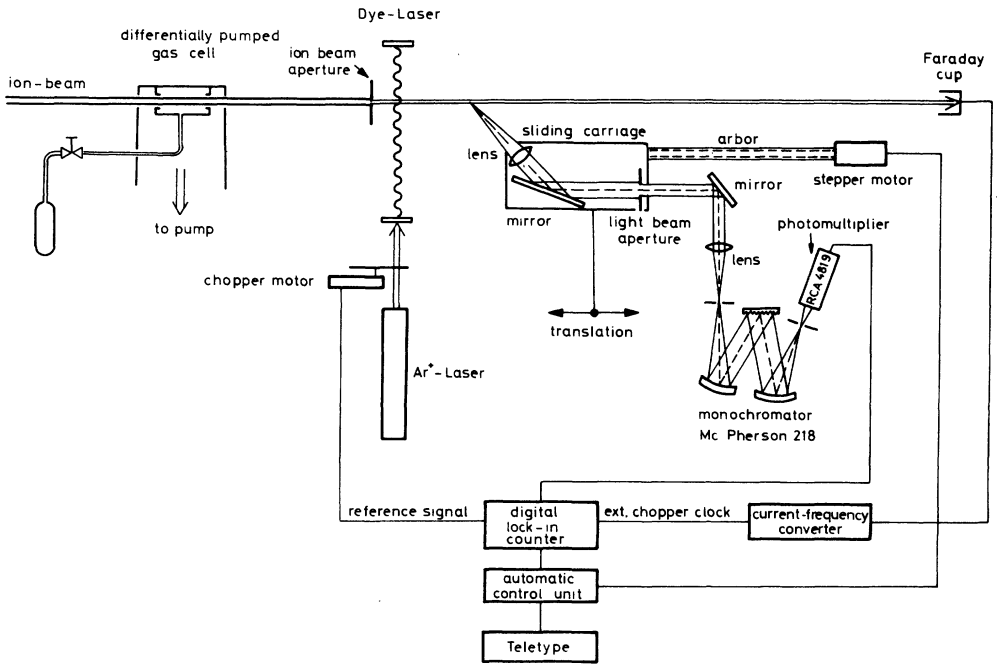

FIG. 2. Experimental arrangement of gas-laser excitation and following cascade-free lifetime measurement.

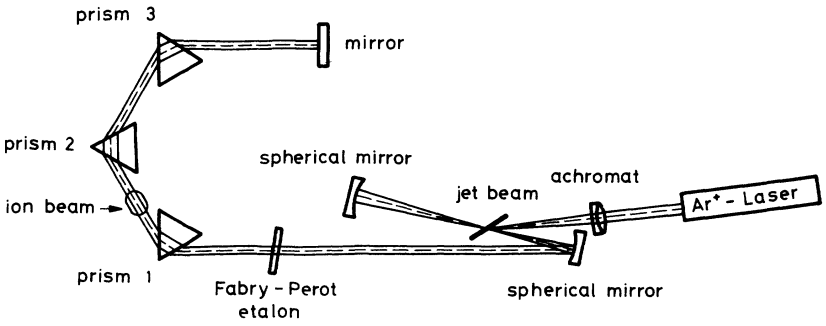

FIG. 3. Schematic drawing of the dye-laser resonator.

wavelength of $\lambda = 5854$ Å. Afterwards it is most advantageous to measure the intensity decay curve for the resonance transition $6s\,^2S_{1/2}$-$6p\,^2P_{3/2}$ with $\lambda = 4554$ Å.

Using a time interval of 1 μsec between both excitations, the ions are concentrated in the $5d\,^2D$ states and the ground state, while the $6p\,^2P_{3/2}$ level is emptied. Therefore the population density of the metastable can be enriched to about 5%. With an available dye-laser power of 5 W inside the cavity and a spectral width of 0.7 Å, about 6% of the ions in the metastable state will be pumped to the $6p\,^2P_{3/2}$ level. For a 500-nA Ba^+ beam of 1.5 mm height and 4 mm width, and a 1-mm-diameter laser beam, a counting rate of 4.5×10^4 counts/sec in 1 mm distance after laser excitation is expected, which agrees well with the experiment. We estimated that about 1.5×10^{-5} of all the light from the transition is detected.

For this particular case of Ba^+, however, we should mention that it is not necessary to excite the ions by a gas target. Since the ion source already populates the $5d\,^2D_{3/2}$ state, and the time of flight from the ion source to the laser excitation takes about 2×10^{-5} sec, one expects nearly the same fraction of ions in the metastable state as with gas cell excitation. Under such conditions the spectral width of the laser can be reduced by an etalon to provide an increase in the power per mode. The observed counting rate then rises to 6×10^4 counts/sec.

A typical decay curve as a function of distance after excitation, shown in Fig. 5, consists of a superposition of ten different runs. The measurement was evaluated by a least-squares fit with a pure exponential yielding a lifetime of

$$\tau(6p\,^2P_{3/2}) = 6.28 \pm 0.03 \text{ nsec}.$$

To this error, resulting from the counting statis-

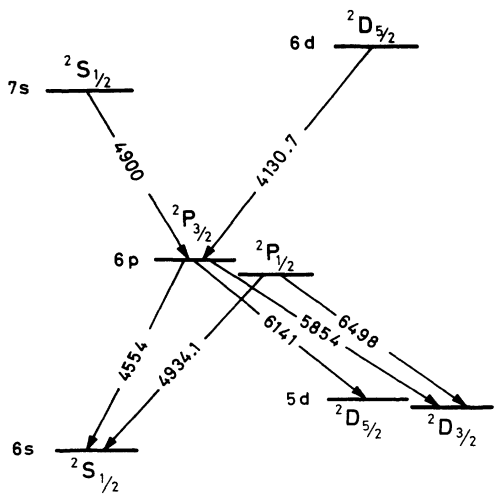

FIG. 4. Fine-structure level diagram of Ba II.

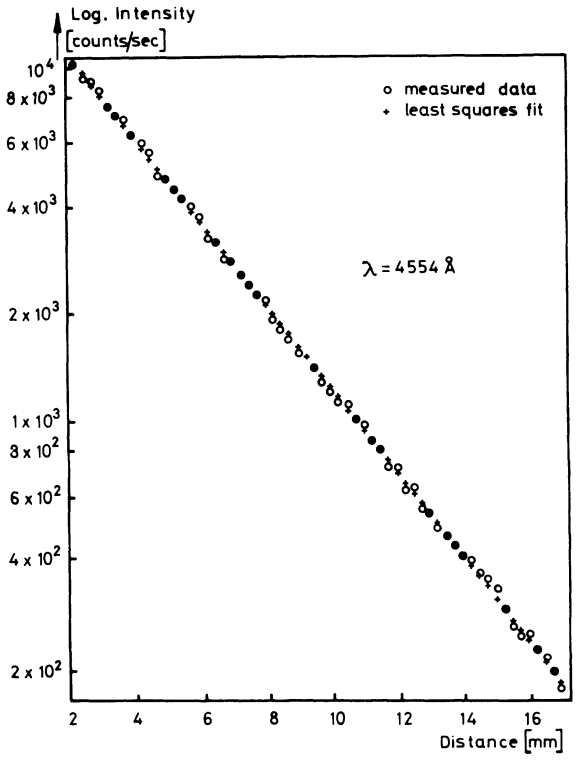

FIG. 5. Semilog plot of the intensity decay curve of the $6p\,^2P_{3/2}$ level as a function of distance after dye-laser excitation.

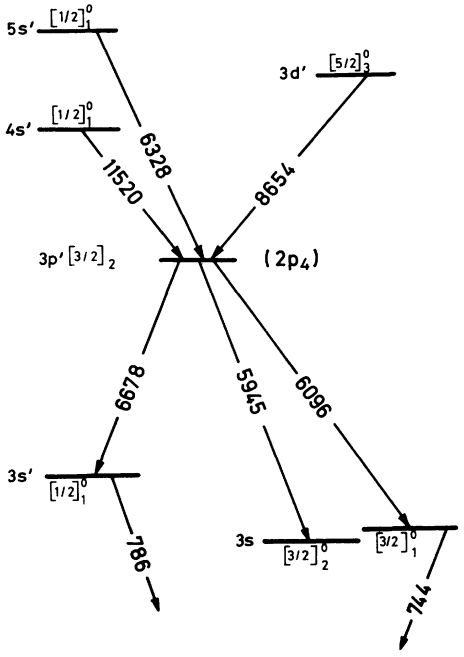

FIG. 6. Fine-structure level diagram of Ne I.

tics and the uncertainty of the ion velocity, was added an additional error due to the alignment of the optical detection system and the stability of the laser power. The final result

$$\tau(6p^2P_{3/2}) = 6.28 \pm 0.06 \text{ nsec}$$

is in excellent agreement with earlier results (Table I) and shows the high accuracy possible with this method.

Another lifetime measurement was carried out for the He-Ne laser level $2p^5(^2P^0_{1/2})3p'[\frac{3}{2}]_2$ of Ne ($2p_4$ in Paschen notation). This level is not optically connected to the ground state but can be populated from the $3s[\frac{3}{2}]^0_2$ metastable state using a laser with a wavelength of $\lambda = 5945$ Å (Fig. 6).

Since the transition probabilities as well as the production of metastables after charge exchange are less favorable than for the case of Ba^+, we could only expect a counting rate of a few hundred counts per second for a Ne^+ beam of 1 μA and

TABLE I. Comparison with earlier results for the lifetime of the Ba^+ $6p\ ^2P_{3/2}$ level.

$\tau(6p\ ^2P_{3/2})$ (nsec)	Ref. (year)
6.29	9 (1968)
7.0 ± 0.6	10 (1966)
11.5 ± 0.6	11 (1971)
6.27 ± 0.25	12 (1968)
6.21 ± 0.06	2 (1973)
6.28 ± 0.06	This work

TABLE II. Comparison with earlier results for the lifetime of the lower laser level $3p'[\frac{3}{2}]_2$ of Ne.

$\tau(3p'[\frac{3}{2}]_2)$ (nsec)	Ref. (year)
110 ± 2.5	13 (1960)
83 ± 11	14 (1934)
11.0 ± 1	15 (1933)
40	16 (1962)
22 ± 4.5	17 (1966)
19.1 ± 0.3	18 (1966)
19.6 ± 0.2	This work

52 keV. Measuring the intensity decay curve of the transition $3s[\frac{3}{2}]^0_1$-$3p'[\frac{3}{2}]_2$ with $\lambda = 6096$ Å, for different runs over five hours, a least-squares fit with a pure exponential yields a lifetime of

$$\tau(3p'[\tfrac{3}{2}]_2) = 19.6 \pm 0.2 \text{ nsec}.$$

The specified error represents the different uncertainties as described before. Table II gives a comparison with previously measured results of the lifetime and shows very good agreement with Ref. 18.

FURTHER APPLICATIONS

The experiments with Ba^+ and Ne demonstrate the high accuracy and the cascade-free lifetime measurements possible with the new technique. A large number of differently charged ground states and metastable states can be produced as a result of beam-gas interactions and afterwards higher-lying levels selectively populated. However, this method of laser excitation with following cascade-free lifetime measurement can also be successfully applied to shorter-lived states, which are still strongly populated just after a foil excitation. When a laser is available to change the population density of the upper or lower level (on which the laser acts) by at least a few percent, hundreds of levels can be investigated in this manner.

In addition to a pure lifetime measurement, there is another interesting application of double excitation. Since the interaction time of the laser with the ions is about 10^{-9} sec, close neighboring levels like hyperfine or Zeeman sublevels will be excited coherently. Moreover, atomic levels populated by a laser are polarized. Therefore, quantum beats[19] should be observed yielding hyperfine-structure splitting factors and g_j factors. For the present measurements on Ba^+ and Ne no quantum beats could be expected, since ions and atoms with nuclear spin $I = 0$ were used, and no external magnetic field was applied. Further applications of laser excitation as already mentioned by Andrä *et al.* (Ref. 2) are possible for Hanle-effect or double-resonance experiments.[20]

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