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Reexamination of the lifetime of the $3p_8$ level of Ar I

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Using laser-induced fluorescence, the lifetime of the $3p_8$ level of argon was measured. Variation of the pressure in the observation chamber by three orders of magnitude allowed a precise extrapolation to the collision-free lifetime.

INTRODUCTION

The lifetimes of the $3p$ levels (Paschen notation) giving rise to the prominent blue argon lines have been the subject of quite a few investigations in the past.[1-8] The results from the various authors differ by as much as a factor of 3. This is surprising insofar as measuring lifetimes in the range of 100 ns and observation of fluorescence in the blue region of the spectrum should not cause any experimental difficulties. Recent measurements in our laboratory done with selective laser excitation for five of the ten $3p$ levels[7] yielded a value of 152 ns for the $3p_8$ level. Using the same technique Hirabayashi *et al.*[8] obtained 127 ns in another recent investigation. As the error bars in the two experiments did not overlap we started a new series of measurements to settle the problem.

EXPERIMENT

Metastable argon atoms were produced by extracting a beam from a hole in the bottom of a dc hollow-cathode discharge into a vacuum chamber. The discharge was operated at a pressure of $p = 0.5$ mbar and a current of $I = 1-20$ mA. For details of the source, see Schade and Helbig[9] and Schade.[10]

The metastable atoms were excited by a dye laser perpendicular to the atomic beam. For the dye laser, we chose a double-prism setup according to Racz *et al.*[11] Using a grating with 2400 lines/mm as a dispersing element, we obtained a spectral width of 0.03 nm. For the present measurements, we took Stilben 1 as the laser dye. As the pump source, we used a nitrogen laser working at a repetition rate of 80 Hz. The temporal width of the dye-laser pulse was approximately 2.5 ns.

For excitation and observation, the lines at 419.07 and at 430.01 nm were used, respectively (Fig. 1). The fluorescence was observed at right angles both to the beam of metastable atoms and to the exciting laser beam. A detection system consisting of a monochromator (Jobin-Yvon H 20) with 1-mm slits, a multiplier (Valvo XP 2020 Q), and a standard delayed-coincidence photon-counting system was employed. The decay curves were read into a multichannel analyzer and stored in a computer. For the measurement of the decay curves at different pressures, it was essential to keep the number density of the metastable atoms constant. Instead of regulating the pressure in the observation chamber by varying the pressure in the discharge, a by-pass to the hollow cathode was used for the pressure variation. The amount of argon entering the observation chamber was controlled precisely by a needle valve. Pressure variations from 1.5×10^{-4} to 1×10^{-1} mbar were obtained by running the diffusion pump with constant pumping speed at the low-pressure end and by using only the rotary pump for the high pressures. Between 1×10^{-3} and 1×10^{-2} mbar, the diffusion pump was used with reduced pumping speed by partially closing the plate valve.

RESULTS

A number of systematic errors may prevent the observation of the undisturbed lifetime in experiments like the present. Among those discussed in the literature are the influence of collisions with atoms in the observation chamber, radiation trapping, escape of the excited atoms before detection of the fluorescence, direction of the polarization in the excitation and observation channel, and

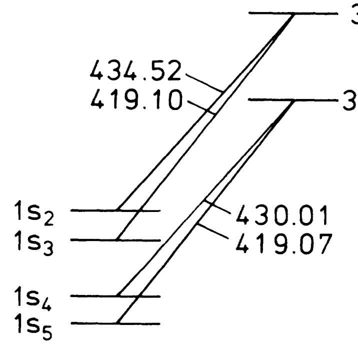

FIG. 1. Part of the Ar I energy-level diagram showing the two transitions to the $3p_8$ level used for pumping (419.07 nm) and observation (430.01 nm) and the two neighboring transitions to the $3p_4$ level (see text).

the influence of residual magnetic fields. In the following we give a discussion of those aspects that apply to our experiment.

An extensive study of the pressure dependence of the observed lifetime was carried out in order to obtain the collision-free value. The results are shown in Fig. 2. Close to three orders of magnitude in pressure are covered by the present measurements. The influence of collisions was previously only taken into account by Ralfs[7] and Hirabayashi *et al.*[8] The measurements by Ralfs covered a pressure range from 5×10^{-4} to 2×10^{-3} mbar. They agree very well with the present results and therefore have not been included in the figure. Hirabayashi *et al.* changed the pressure between 0.09 and 2 torr. The extrapolation of their results is out of scale in Fig. 2. As the difference in the pressure range covered by the respective experiments seems to be a critical point in explaining the deviating lifetime results, we would like to point out that the extrapolated value in our case differs from the last data point by only 1.2% whereas in the experiment of Hirabayashi *et al.*[8] it differs by 11%.

To take into account the possible influence of radiation trapping on the observed lifetime, the source was operated at different currents. Values up to $I=2$ A could be obtained running the hollow cathode in a pulsed mode. For details, the reader is referred to Schade.[10] As could be judged from the intensity of the fluorescence, the number density of the metastable atoms could be drastically increased by this measure. An influence on the lifetime was not observed.

As the lifetime of the $3p_8$ level is fairly long, the excited atoms in the beam may pass through the observation region before they have emitted a photon. Experimental measures have been taken to rule out an influence of this process on the present investigation. An estimate of the particle velocity in the beam and the known magnification of the image of the observation volume on the monochromator slit was used to calculate the slit width. Adjusting the image of the excitation zone to one jaw of the slit would guarantee that all of the excited atoms radiate while passing the volume that corresponds to the slit opening. Adjusting the excitation zone to the other jaw, the observed lifetimes were drastically shorter. The final measurements were carried out with a slit width of 1 mm.

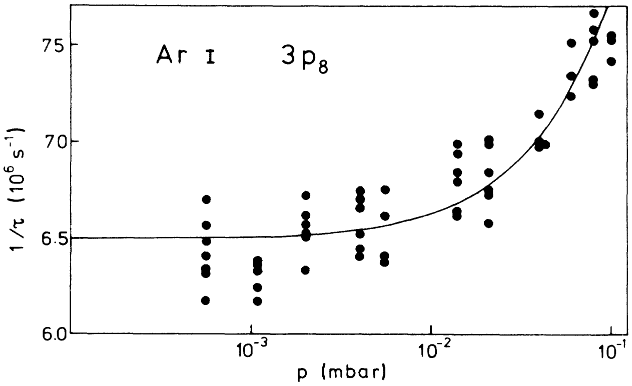

FIG. 2. Semilogarithmic plot of the pressure dependence of the inverse lifetime of the Ar I $3p_8$ level. The closed circles indicate the single values from independent decay curves. The solid line gives the fit assuming a linear dependence on the pressure.

Excitation anisotropy of the Zeeman sublevels caused by the laser excitation has been neglected for a long time in lifetime determinations. As first pointed out by Fujimoto *et al.*,[12–14] the fluorescence intensity does not necessarily follow the decay of the upper-level population. A quantitative theoretical description has been given by Hannaford and Lowe.[15] An extensive experimental study of these polarization effects has been carried out in our laboratory (Schade *et al.*[16]). The results for neon were in quantitative agreement with theoretical predictions. They may be summarized as follows: alignment produced by selectively exciting atomic levels with linearly polarized laser radiation will be destroyed in the presence of disturbing atoms by collisions. If a polarizer is used in the detection channel, the shape of the observed fluorescence curve not only depends on the natural lifetime of the excited level, but also on the angle between the polarization in the excitation and observation channel, on the cross section for alignment destroying collisions, and on the number density. Even if no polarizers are used, an influence of the effect may be present in an experiment, as mirrors, gratings, nonlinear crystals, and detectors can produce partial or complete polarization. There are two ways to measure the unperturbed decay curve. One is to use "magic-angle" excitation and the other is to do the experiment at sufficiently low pressure so that collisions can be neglected. In the present experiment we have chosen the second approach for two reasons. First, as the fluorescence intensity is very low, a long time for data collection is required in order to build up a decay curve. For optimizing the signal-to-noise ratio, it was therefore desirable to use no polarizers. Secondly, we had to extend the measurements to low pressure anyway in order to exclude the collisional depopulation of the excited levels.

The fluorescence signal of an ensemble of aligned atoms in addition to the exponential term due to the decay of the population will contain an oscillatory term in the presence of a magnetic field.[15] If the period of these Zeeman beats is short compared to the lifetime, an average over these oscillations is taken in the evaluation routine and the correct lifetime value is obtained. Problems may arise with very weak fields, such as, for instance, the Earth's magnetic field or stray fields from the laboratory equipment. They may cause the Larmor period to become comparable to the lifetime value. In such a case the apparent decay constant of the fluorescence curve will differ from the lifetime. For the present experiment, we measured the magnetic field at the position of the observation chamber and estimated the corresponding Larmor frequency to be 5 MHz. Since the decay curves could be measured for time intervals corresponding to four–six lifetimes, this shows that we averaged over about one to two periods sufficient to get the influence of the magnetic field averaged out. For the discussion to follow, it is important to mention that experiments applying a defined

TABLE I. Lifetime of the Ar I $3p_8$ level obtained by various authors.

Author	Excitation	Lifetime (ns)
This work	laser	154±7
Hirabayashi *et al.*[a]	laser	127±10
Ralfs[b]	laser	152±7
Borge and Campos[c]	*e* beam	165±14
Erman and Huldt[d]	*e* beam	120±15
Malakhov and Potyomkin[e]	*e* beam	59±6
Chenevier and Goulet[f]	*e* beam	130±20
Verolainen and Osherovich[g]	*e* beam	148±12
Klose[h]	*e* beam	166±17[i]

[a] Reference 8.
[b] Reference 7.
[c] Reference 6.
[d] Reference 5.
[e] Reference 4.
[f] Reference 3.
[g] Reference 2.
[h] Reference 1.
[i] Possible systematic errors are included as in column 5 of Table II of Ref. 1.

stronger magnetic field to the observation chamber[16] lead to significantly shorter lifetime values than without a field in the case of neon. This can be explained by additional collisions caused by the gyrating particles.

Special care was taken to guarantee selective excitation as well as selective observation. Figure 1 shows part of the energy-level diagram of Ar I including the transitions considered in this paper. It is evident that two lines connected with the $3p_4$ level are close to the transitions involved in the present measurements. The spectral width of the dye laser was small enough to distinguish between the 419.1- and 419.07-nm excitation lines. Nevertheless, to prevent excitation of the $3p_4$ level, it was checked before all experiments whether fluorescence could be observed on the 434.5-nm line or not. The slit width of 1 mm of the monochromator was sufficient to resolve the 430.01- and 434.5-nm lines.

For the reduction of the data stored in the computer, the background was subtracted in a first step. The width of the single channels in the multichannel analyzer was adjusted to correspond approximately to one-tenth of a lifetime. The decay curves could be evaluated for roughly four–six lifetimes. The background was obtained by averaging the count rate of channels far enough away from those of the decay curve. In a second step, a correction was applied to account for a possible influence of the pileup effect. As input data for this correction, the ratio of the total number of excitation pulses and the number of fluorescence photons for every curve was monitored. This ratio varied from 7:1 to 10:1. Instead of fitting an exponential curve to the data in the last step of the program, a straight line was fitted to the logarithm of the count rates. This required a weighted least-squares routine, giving more weight to those channels with high counting rates.

Table I compares the present results with those of earlier experiments. In most of the investigations done so far, excitation of the level was achieved by electron impact. Only in the work done in our laboratory and in that of Hirabayashi *et al.* was selective laser excitation of the respective level used. Within the mutal error bars, the present results agree with most of the earlier results. Exceptions are the values of Refs. 4, 5, and 8. Malakhov and Potyomkin[4] use nearly identical experimental conditions as Klose.[1] Their surprisingly small value for the $3p_8$ level probably has to be traced back to undetected systematic errors in their measurements or the data reduction. The results of Erman and Huldt[5] obtained by the so called "high-frequency deflection" technique probably suffer from the short time interval between successive excitation pulses. There is the possibility that this interval is not sufficient to allow the metastable $2s$ levels to decay by collisions. This could be the reason for the exceptionally strong radiation trapping that they observe. The corrections that they have to apply probably overestimate the effect leading to the smaller lifetime that they state. The same was observed for some of the $2p$ levels.[16] To explain the deviations of the present data from those of Hirabayashi *et al.*,[8] one has to realize that their measurements have been carried out at pressures where the observed lifetimes are strongly influenced by collisions. Though they extrapolate to zero pressure, it is obvious that the scatter of the data as is shown in their Fig. 5 makes the extrapolation uncertain. Besides that, one has to realize that this extrapolation is in their case by no means a small correction to the actually measured lifetimes that range approximately from 55 to 113 ns according to their Fig. 5. Essentially collision-free measurements of the lifetime of the $3p_8$ level can be made for the pressures lower than 10^{-2} mbar as can be seen from our Fig. 2. Raising the pressure up to 10^{-1} mbar causes a shortening of the observed lifetime to the value of 130 ns, very close to that observed by Hirabayashi *et al.*[8] No definite answer can be given yet whether the magnetic field applied in the case of their experiment causes an additional collisional depopulation of the excited level, as was indicated by our experiments in neon.[16]

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