Measurement of Ne $2p^5 3p$ lifetimes by laser–electron-impact double excitation employing the delayed-coincidence method

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Radiative lifetimes of six Ne $2p^5 3p$ states have been measured with uncertainties lower than 1.5% by means of a pulsed electron excitation laser-induced fluorescence technique. This method allows one to obtain fluorescence signals by exciting from resonant as well as from metastable levels at low pressures. The lifetime results are 14.0 ± 0.1, 17.9 ± 0.1, 16.9 ± 0.2, 20.2 ± 0.1, 19.0 ± 0.2, and 20.6 ± 0.2 nsec for the $2p_1$, $2p_2$, $2p_3$, $2p_4$, $2p_5$, and $2p_6$ levels, respectively.

I. INTRODUCTION

The laser-induced atomic fluorescence method[1] has proved to be one of the most reliable techniques for lifetime measurements, since the selective excitation of the atomic states allows one to obtain cascade-free results. This technique has been used for excitation from metastable states of neutral atoms by Setser *et al.*,[2–4] Fujimoto *et al.*,[5] and Kandela and Schmoranzer[6] in rare gases. Eyler and Pipkin[7] employed the method for H_2 molecules. In these works metastable atoms or molecules are generated by flowing afterglow,[2–5] ion-beam neutralization,[6] and molecular-beam apparatus,[7] respectively; and a dye laser is tuned to excite the metastable atoms to a selected excited state.

In this work several useful changes have been made. With regard to resonant- and metastable-state generation, a pulsed electron beam has been used. For selective excitation of upper levels an intracavity acousto-optic pulsing laser configuration has been employed. This laser system allows one to obtain short and intense laser pulses with a high repetition frequency that allows the use of the single-photon delayed-coincidence method for level-decay measurements.

The present double-excitation method by electron impact and laser-induced fluorescence is advantageous in some cases. The experiments can be done at lower pressures than with the afterglow method, and greater accuracy in lifetime measurements can be achieved. Also it must be pointed out that laser-induced atomic fluorescence can be produced from resonant as well as from metastable states.

Although lifetimes of levels of the Ne $2p^5 3p$ configuration have been the object of several experimental[3,6,8–10] and theoretical[11,12] studies, discrepancies persist. Thus, in our opinion, it is interesting to increase the accuracy of experimental lifetime results, so that Ne transition probabilities deduced from them can be used as standards. The object of the present work has been to improve the accuracy of lifetime data by using the laser–electron-impact double-excitation method. For this purpose radiative lifetimes of six neutral atomic Ne $2p^5 3p$ levels have been measured with uncertainties lower than 1.5%.

II. EXPERIMENTAL SETUP

As mentioned above, lifetime measurements have been made by using the delayed-coincidence method.[13] An electron excitation chamber has been used. The chamber was provided with two arms, 20 cm long, for laser-beam crossing. These arms were fitted with quartz windows at Brewster's angle. The electron collision chamber had a triode configuration. A control grid was used to pulse the electron beam. Impact excitation took place in the inner part ot the anode that was a cylindrical box provided with small holes at right angles for fluorescence observation, for laser crossing, and for electron admittance. In Fig. 1 a block diagram of the experimental setup is shown.

The laser system used in this work has been described in Ref. 14. An argon-ion laser (Spectra-Physics model 165) was used to pump a pulsed dye laser consisting of a dye-laser–cavity-dumper system (Spectra-Physics models 580A and 344S, respectively). Cavity dumping is achieved with an intracavity acousto-optic modulator. This configuration allows one to obtain laser pulses of ~ 15 nsec dura-

tion and 1.7 W peak power with a repetition frequency up to 5 MHz. The dye used was rhodamine 6G. The laser spectral resolution was better than 0.5 Å.

The impinging electron's energy was in the range from 20 to 70 eV, which was previously reported as suitable for metastable excitation.[15,16] The electron beam was pulsed at a repetition rate of 28 KHz. Pulse duration was 6 μsec. The pulse ends sharply in less than 10 nsec. Laser pulses were produced 1.5 μsec after the end of the electron pulse.

A fraction of the laser beam was focused on an EMI 9781B photomultiplier (PM) in order to obtain the start signal for a time-to-amplitude converter (TAC). Laser-induced fluorescence light produced in the anode was focused on a Jovin-Yvon 5-Å bandpass monochromator. Single photons were detected by an EMI 9816B cooled photomultiplier; output pulses from the photomultiplier served as "stop" pulses for the TAC. A 1024-channel pulse-height analyzer (PHA) was used to classify and store the TAC output. The number of photons detected per laser pulse was kept lower than 5×10^{-3} to avoid pileup. The TAC was previously calibrated with an accuracy better than 0.3% by means of a quartz oscillator.

To find the resonance wavelengths for atomic fluorescence excitation, the laser beam chopped at 40 Hz was focused on a Ne hollow-cathode lamp.[15–17] Its anode voltage signal was monitored by an oscilloscope. An optogalvanic effect was easily observed when the laser wavelength was tuned to resonance.

The ultimate vacuum at the chamber was better than 10^{-6} Torr. A continuous flow of high-purity Ne was maintained in the measurements. The Ne pressure lay in the range from 40 to 150 mTorr; no dependence of $2p^5 3p$ states lifetime with gas pressure was observed within experimental uncertainties.

The present experimental system has some similarities with the one recently reported by Setser *et al.*[18] Nevertheless, the present method has some remarkable differences, the main one being the low electron energy of this work and the high repetition rate of the laser pulses that allows the use of the single-photon delayed-coincidence method. Also, the present technique enables excitation from any resonant state. This is accomplished by taking advantage of resonance trapping and by adjusting the delay between laser and electron pulses suitably.

III. RESULTS AND DISCUSSION

In Fig. 2 time dependence of the laser pulse is shown. In this figure a typical fluorescence decay has been plotted. Two methods are possible for the decay analysis. The first is to study the fluorescence decay after the end of the laser pulse, which can be fitted to an exponential decay over more than one decade. This method gives preliminary results. A more elaborate method is to perform the convolution integral of the laser pulse with an exponential decay. This method gave a good fit to the experimental points in every case. A collection time of about 1 h was enough to achieve a statistical accuracy to determine lifetime with an uncertainty less than 0.3 nsec. In Table I mean lifetime values with a statistical accuracy better than 1% are given. To calculate the total error an estimated 1% systematic error must be added.

Transitions used in this work for laser excitation

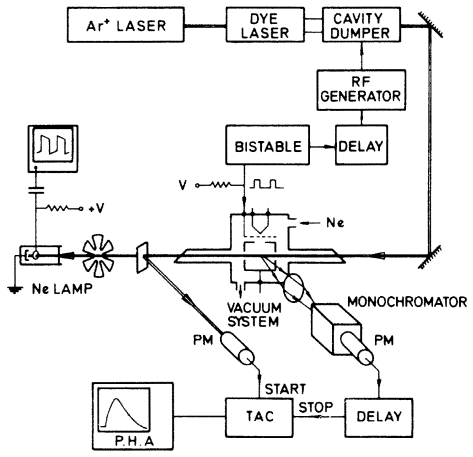

FIG. 1. Schematic diagram of the experimental setup used to measure Ne lifetimes by the method of pulsed electron excitation and laser-induced fluorescence.

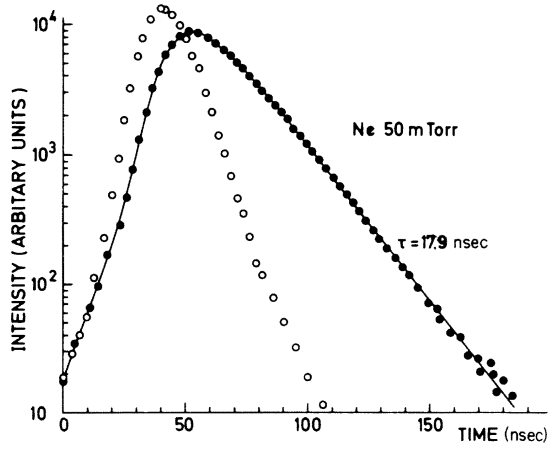

FIG. 2. Typical result for the decay of the $2p_2$ level of Ne. ○, excitation laser pulse; ●, fluorescence decay; — , convolution fitting to the experimental points.

TABLE I. Radiative lifetimes (nsec) of Ne $2p^5 3p$ states.

	Experimental						Theoretical	
Level	This work	Chang and Setser	Kandela and Schmoranzer	Bennett and Kindlmann	Klose	Martín and Campos	Feneuille	Mehlhorn
$2p_1$	14.0±0.1[a]			14.4±0.3	14.7±1.3	14.7±0.8	13.7	15.1
$2p_2$	17.9±0.1 17.6±0.3[a]	19.8±0.4	18.28±0.07	18.8±0.3	16.3±1.4	19.9±2.5	17.8	18.9
$2p_3$	16.9±0.2[a]		17.38±0.08	17.6±0.2	23 ±6.6	21.5±2	16.7	16.9
$2p_4$	20.2±0.1 20.2±0.3[a]	19.8±0.5	18.69±0.04	19.1±0.3	22 ±5.4	20.9±2.5	18.2	19.1
$2p_5$	19.0±0.2	19.2±0.4	19.05±0.05	19.9±0.4	18.9±2.8		18.8	20.2
$2p_6$	20.6±0.2	19.0±0.3	19.38±0.03	19.7±0.2	22 ±3.2	20.5±2	18.8	20.2

[a]Lifetime measured by means of excitation from a resonant level.

and decay measurement are listed in Table II.[19] For lifetime measurement of $2p_2$, $2p_4$, $2p_5$, and $2p_6$ levels the $1s_3$ and $1s_5$ metastable levels have been used for excitation. The resonant level $1s_4$ has been used for measurements of $2p_2$, $2p_3$, and $2p_4$ levels. The results obtained by using excitation from metastable and resonant levels for the $2p_2$ and $2p_4$ levels are in agreement within experimental errors. Finally, the $1s_2$ resonant level has been used for $2p_1$-state excitation. For the $2p_1$ and $2p_3$ lifetime study, the excitation and decay measurement lines were the same, due to the low transition probability of the other transition.[20–23] Special care was taken in this case to avoid laser reflections, and the small remaining background was subtracted.

Results are compared in Table I with previous experimental and theoretical results found in the literature. Chang and Setser[3] and Fujimoto *et al.*[5] measured Ne lifetimes with laser techniques using the flowing afterglow method. Their work was made at pressures above 0.5 Torr and a N_2 laser was used for pumping the dye laser, so a relatively low repetition rate could be achieved. Kandela and Schmoranzer[6] used a laser-beam method. Bennett and Kindlmann[8] used the electron excitation delayed-coincidence technique avoiding cascade problems by using electron energies just above threshold of a given level. Also in Table I are included the results of Refs. 9 and 10, obtained by using the same method. The theoretical values obtained by Feneuille[11] and Mehlhorn[12] in the intermediate coupling approximation are shown, too.

The lifetime values obtained in this work do not have remarkable discrepancies with the results of Refs. 6 and 8. The values of these authors tend to be slightly higher than ours except for the $2p_4$ and $2p_6$ levels. The mean discrepancy with the results of Chang and Setser[2] is 5% without a systematic tendency. Fujimoto *et al.*[5] have measured the $2p_2$ level recently by using the laser-afterglow technique; their result is 18.8±0.9 nsec in agreement within experimental errors. The present lifetimes agree with the theoretical results of Ref. 11 and with those of Ref. 12 within 4%.

TABLE II. Transitions and wavelengths used in the present work for lifetime measurements.

Level		Excitation transition	Laser wavelength (Å)	Measurement transition	Fluorescence wavelength (Å)
Paschen	jK				
$2p_1$	$[3p'(\frac{1}{2})]_0$	$1s_2$-$2p_1$[a]	5852	$2p_1$-$1s_2$	5852
$2p_2$	$[3p'(\frac{1}{2})]_1$	$1s_5$-$2p_2$ $1s_4$-$2p_2$[a]	5882 6030	$2p_2$-$1s_3$	6164
$2p_3$	$[3p(\frac{1}{2})]_0$	$1s_4$-$2p_3$[a]	6074	$2p_3$-$1s_4$	6074
$2p_4$	$[3p'(\frac{3}{2})]_2$	$1s_5$-$2p_4$ $1s_4$-$2p_4$[a]	5945 6096	$2p_4$-$1s_4$ $2p_4$-$1s_5$	6096 5945
$2p_5$	$[3p'(\frac{3}{2})]_1$	$1s_5$-$2p_5$	5975	$2p_5$-$1s_3$	6266
$2p_6$	$[3p(\frac{3}{2})]_2$	$1s_5$-$2p_6$	6143	$2p_6$-$1s_4$	6305

[a]Excitation from resonant level.

IV. CONCLUSIONS

Six Ne $2p^5 3p$ levels have been measured by means of the laser–electron-impact delayed-coincidence method, at low pressure. The reported lifetime for the $2p_1$ level has been measured for the first time by laser techniques. The method has proved to be simple and useful for lifetime measurement on levels that can be excited from metastable or resonant levels.

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