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PHYSICAL REVIEW VOLUME 178, NUMBER 1 5 FEBRUARY 1969

Mean Lives of Some Doubly Excited Levels in Lithium I*

William S. Bickel,† Ingmar Bergström, Rudolf Buchta, Lennart Lundin, and Indrek Martinson
Research Institute for Physics, Stockholm 50, Sweden
(Received 10 October 1968; revised manuscript received 18 November 1968)

Three doubly excited levels in Li I produced upon passage of 56-keV ^{7}Li ions through a thin carbon foil have been identified. Their mean lives were measured using the beam-foil technique with the results given below:

$$\tau(1s2p^2\,{}^4P) = (6.4 \pm 0.3) \times 10^{-9} \text{ sec},$$
$$\tau(1s2s3s\,{}^4S) = (9.7 \pm 0.7) \times 10^{-9} \text{ sec},$$
$$\tau(1s2s3d\,{}^4D) = (4.5 \pm 0.4) \times 10^{-9} \text{ sec}.$$

In the optical spectrum of lithium certain spectral lines appear which cannot be assigned to the normal term schemes of Li I or Li II. It has been suggested by several authors[1–4] that these lines originate from radiative transitions between doubly excited quartet states in Li I, lying between the first and second ionization potential.

Due to selection rules these levels cannot auto-ionize through Coulomb interaction. Auto-ionization, because of magnetic interaction, is much less likely and this process is too slow to compete with radiative decay to the lowest doubly excited level.

Four lines in the visible region classified as transitions between doubly excited levels are

$1s2s3d\,{}^4D \rightarrow 1s2s2p\,{}^4P$	λ2337 Å,
$1s2s3s\,{}^4S \rightarrow 1s2s2p\,{}^4P$	λ2934 Å,
$1s2p^2\,{}^4P \rightarrow 1s2s2p\,{}^4P$	λ3714 Å,
$1s2p3p\,{}^4P \rightarrow 1s2s3p\,{}^4P$	λ4607 Å.

Applying the beam-foil technique[5] we studied the radiation coming from the first three of these transitions, and measured the intensity decay of the three different spectral lines to determine the mean lives of the upper levels.

^{7}Li ions, accelerated to 56 keV by the isotope separator at the Research Institute for Physics, were directed through a thin (10 ± 4 μg/cm^2) carbon foil. A fraction of the ions picked up an electron in the foil and emerged in various excited levels of neutral lithium. The excited beam was analyzed downstream from the foil with a low dispersion (33 Å/mm in first order) f/3.6 Jarrell-Ash 0.25 m scanning monochromator equipped with a sodium salicylate-coated photomultiplier detector. When we rotated the grating and fed the amplified photomultiplier current into a strip chart recorder, we got spectral scans of the wavelength region between λ2000 Å and λ6000 Å.

Twenty-four spectral lines were detected,[6] three of which were identified with previously reported lines from doubly excited Li I. Figure I shows a partial wavelength scan of a typical spectrum obtained. Studies of similar spectral scans, obtained with different slit widths, beam currents, and incident particle energies, assisted us in identifying the lines denoted as A, B, and C, respectively, with the first three lines given above. Unfortunately, the line expected at λ4607 Å practically coincides with the line at λ4603 Å from the $1s^24d\,^2D \rightarrow 1s^22p\,^2P$ transition in Li I.

We determined the mean lives of the upper levels in a straightforward way[7,8] by measuring the intensity $I(x)$ of radiation, at the corresponding wavelengths from the beam, for various foil positions x relative to the entrance slit of the monochromator. We assume that the data obtained this way are represented by the function

$$I(x) = \sum_1^n I_i^0 \exp(-\alpha_i x/v),$$

where the I_i^0 and α_i/v can be determined by a least-squares computer fit. The α_i are the total transition probabilities per second out of initial states i, and are found directly when the velocity v of the radiating particles is known. We measured v to within an uncertainty of 2% with an 18-cm radius 90° electrostatic analyzer, which gave both the energy of the particles and the energy spread of the beam before and after interacting with the foil. The incident particle energy E_i was 56.2 keV and had an energy profile that was essentially Gaussian with a full width at half maximum of 0.4 keV. The final particle energy E_f after interacting with the foil was 48.8 keV and also had a Gaussian profile, but with a spread of 3.0 keV at half-maximum. The measured energy loss in the foil is $\Delta E = E_i - E_f = (8 \pm 3)$ keV, a value which agrees with the one obtained by extrapolating Northcliffe's[9] data. Our value for the final particle velocity is

$$v = [(2/m)(E_i - \Delta E)]^{1/2} = (1.16 \pm 0.02) \times 10^8 \text{ cm/sec}.$$

The 2% uncertainty is mainly due to the statistical velocity distribution, but it also includes possible uncertainties in the energy analyzer, e.g., small variations in the distance between the plates and influence of the fringing fields. We point out that when v is determined this way, the uncertainty of foil thickness is of minor importance.

THE $1s2p^2\,^4P$ LEVEL

Figure 2 shows the data which we accumulated

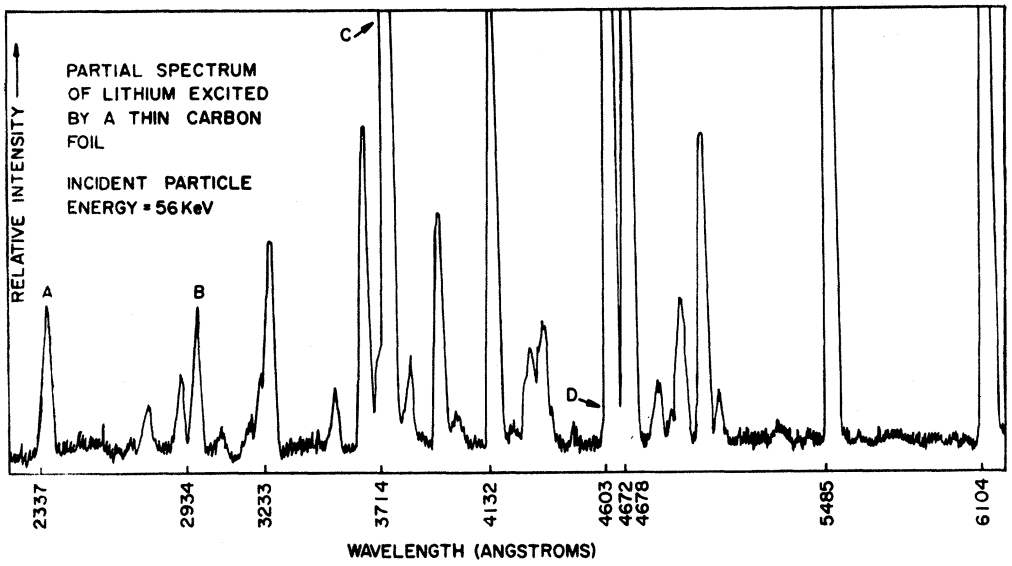

FIG. 1. Part of a spectral scan obtained from beam-foil-excited lithium atoms. The signals assigned to the decay of doubly excited levels are labeled A, B, C, and D.

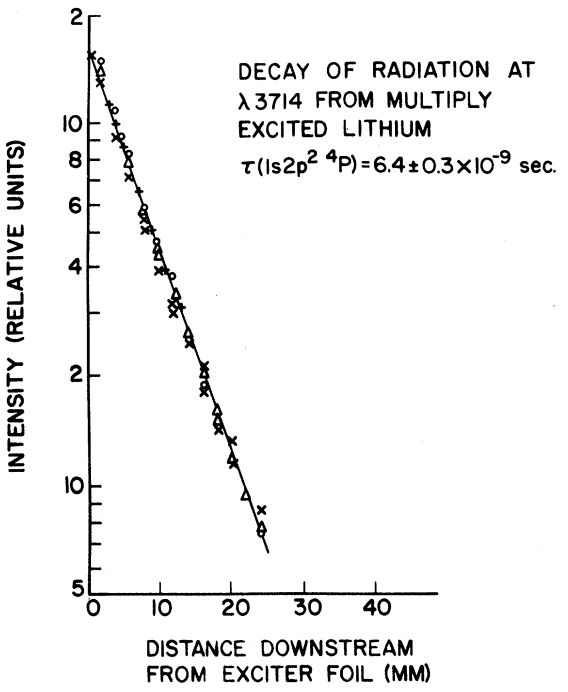

FIG. 2. The result of seven independent measurements of the intensity of radiation at λ3714 Å ($1s2p^2\,^4P \rightarrow 1s2s2p\,^4P$) from Li I as a function of distance x downstream from the exciter foil. The slope of the curve yields a mean life $\tau = (6.4 \pm 0.3) \times 10^{-9}$ sec for the $1s2p^2\,^4P$ level.

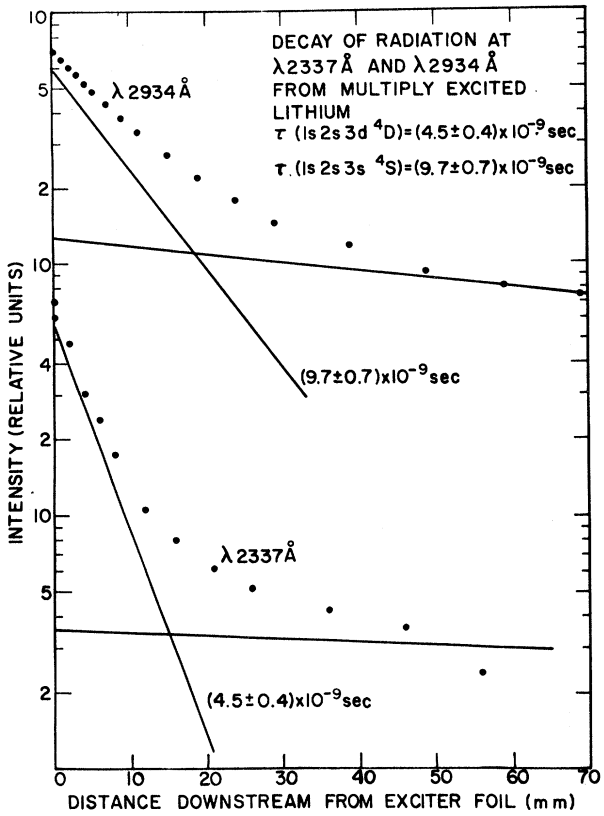

FIG. 3. The results of five independent measurements of the radiation at λ2337 Å ($1s2s3d\,^4D \rightarrow 1s2s2p\,^4P$) and λ2934 Å ($1s2s3s\,^4S \rightarrow 1s2s2p\,^4P$) from Li I as a function of distance x downstream from the exciter foil. The decays of both excited levels are affected by cascades.

by measuring the intensity $I(x)$ of the spectral line at λ3714 Å as a function of distance x downstream from the exciter foil. The straight line drawn through the data points represents a computer least-squares fit to the function $I(x) = I^0 \exp(-\alpha x/v)$, where α and $\tau = \alpha^{-1}$ have the values $(1.6 \pm 0.1) \times 10^8$/sec and $(6.4 \pm 0.3) \times 10^{-9}$ sec, respectively. The uncertainty is mainly caused by the statistics from the least-squares fit. That the best fit is a single exponential indicates that the decay of the $1s2p^2\,^4P$ level is insignificantly affected by cascades from higher levels.

THE $1s2s3s\,^4S$ AND $1s2s3d\,^4D$ LEVELS

The decay curves for the $1s2s3s\,^4S$ and $1s2s3d\,^4D$ levels are given in Fig. 3. As with the $1s2p^2\,^4P$ level, these mean lives were determined by fitting the data to a sum of exponentials. In neither case, however, can the decay be represented by a single exponential; they are apparently affected by cascades from higher levels. We choose to assign the decay with mean life $\tau = (9.7 \pm 0.7) \times 10^{-9}$ sec to the $1s2s3s\,^4S$ level, and attribute the long-lived component with $\tau = (100 \pm 20) \times 10^{-9}$ sec to the effect of cascades. That this is actually the case is not improbable because we expect the higher excited levels to have successively longer mean lives, and expect the intensity contributed to the observed spectral line by the cascade levels to be smaller than the contribution from the decaying level under observation.

We apply the same argument to the $1s2s3d\,^4D$ level decay and assign to it the value $\tau = (4.5 \pm 0.4) \times 10^{-9}$ sec, while letting the longer-lived decay $\tau = (185 \pm 40) \times 10^{-9}$ sec, be associated with the cascading term. We make these assignments with reservation, however, since very little is yet known about the relative populations of excited levels of beam-foil excited atoms and virtually nothing for multiply excited levels.

We attach special significance to the presence of cascades in these aforementioned cases because they indicate the existence of even higher multiply excited levels which have been suggested by theory,[1,2] but not yet detected experimentally.

THE $1s2p3p\,^4P$ LEVEL

We were not able to resolve the signal at λ4603 Å which can be caused both by the $1s^24d\,^2D \rightarrow 1s^22p\,^2P$ transition of Li I at λ4603 Å and the transition of multiply excited Li at λ4607 Å. The transition

probability out of the multiply excited level causing λ4607 Å was predicted by Garcia and Mack[1] to be two orders of magnitude larger than that out of the upper multiply excited level causing λ3714 Å. A study of the decay of λ4603 Å was made, but we did not see the expected short-lived component. In fact, our experimentally measured value for τ of $(3.3 \pm 0.1) \times 10^{-8}$ sec, obtained by measuring the decay of λ4603 Å, agrees well with previous measurements of the mean life of the $1s^2 4d\,^2D$ level in Li I. Thus no definite conclusions can be drawn about the mean life of the $1s2p3p\,^4P$ doubly excited level.

Our results and those predicted theoretically by Garcia and Mack[1] are given in Table I. The theoretical transition probabilities were calculated from screened hydrogenic wave functions neglecting spin. We see that in all cases, large discrepancies exist between the theoretical and experimental values, indicating that re-examination of the theoretical calculations is necessary.

We think the appearance of these multiply excited levels at relatively low incident particle energies (< 100 keV) is significant, because it gives strong support to the hypotheses that multiple electronic excitations can be highly probable for ions of *all* charge states which emerge from the foil. We point out that in all beam-foil spectra hitherto observed, with the exception of hydrogen,[7] and helium,[10] and lithium (this work), no transitions have been detected that could be assigned with absolute assurance to neutral emitters.[11] This puzzling observation cannot be explained by: (1) too low probabilities for transition between neutral excited states; (2) too high incident particle energy (Lyman-α radiation has, for example, been detected when our incident beam was 1 MeV protons); or (3) high probability that all neutral atoms are in the ground state.

However, the existence of a high probability for the formation of multiply excited auto-ionizing states can cause this. Since the atom-foil interaction is violent, it is probable that a large fraction of the ions and atoms emerge from the foil in multiply excited levels. When the probability of the excitation by electron ejection is higher than that for photon emission (as it is in most cases), this mechanism can cause a shift of all charge states upward by 1 or 2 units thereby removing most of the neutrals. Since the transition probabilities of most auto-ionizing levels are extremely high (~ 10^{13}/sec), their mean lives are short and transitions between excited levels will yield broad and faint spectral lines which could have escaped detection with the spectroscopic exposures that have been used in beam-foil experiments so far. Four studies underway which can yield information concerning the probability of formation and relative populations of multiply excited levels of beam-foil excited atoms and ions are (1) a study of the charge state distributions downstream from the exciter foil by direct particle counting; (2) higher resolution of beam-foil spectra and detailed analysis of the energy dependence of unidentified spectral lines; (3) measurements of the energy distribution of electrons ejected by the foil and beam particles during excitation; and (4) a study of the collisional de-excitation of certain multiply excited levels, and subsequent quenching of spectral lines which occur when the foil-excited particles enter a differentially pumped gas chamber.

Evidently the beam-foil technique can be applied to the study of multiply excited auto-ionizing levels and at lower energies (< 100 keV) be exploited for studies of spectra of neutral atoms.

ACKNOWLEDGMENTS

We thank J. Hilke for the capable assistance he gave us while the experimental work was being done, and we are grateful to S. Bashkin and J. D. Garcia for helpful discussions. It is also our pleasure to thank H. Gordon Berry for his assistance with the computer analysis of the decay curves, and Georg Sidenius for valuable advice concerning the electrostatic analyzer. One of us (W. B.) gratefully acknowledges the good hospitality extended to him during his stay at the Research Institute for Physics in Stockholm.

TABLE I. Experimental and theoretical values for the mean lives and transition probabilities of the doubly excited levels of Li I.

λ (Å)	Transition	τ_{exp} (nsec)	A_{exp} (units of 10^8/sec)	A_{th}[a] (units of 10^8/sec)
3714	$1s2p^2\,^4P \rightarrow 1s2s2p\,^4P$	6.4 ±0.3	1.6 ±0.1	29
2934	$1s2s3s\,^4S \rightarrow 1s2s2p\,^4P$	9.7 ±0.7	1.0 ± 0.1	0.17
2337	$1s2s3d\,^4D \rightarrow 1s2s2p\,^4P$	4.5 ±0.4	2.2 ±0.2	52

[a] In Ref. 1 the values 59, 0.33 and 88, respectively, are given. The term scheme proposed was, however, partly based on an unsatisfactory experimental number. The theoretical transition probabilities given above have been corrected for this fact.

*Supported in part by NASA, ONR, and The Research Institute for Physics, Stockholm, Sweden.

†Present address: Department of Physics, University of Arizona, Tucson, Arizona.

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PHYSICAL REVIEW VOLUME 178, NUMBER 1 5 FEBRUARY 1969

Radiative Lifetimes in the Resonance Series of Ne

George M. Lawrence and H. S. Liszt*
Douglas Advanced Research Laboratories, Huntington Beach, California 92647
(Received 14 October 1968)

Direct measurements have been made of radiative lifetimes of the nine $J=1$ levels of the $3s$, $4s$, $5s$, and $3d$ configurations in Ne I. Branching ratios have been calculated in the Coulomb approximation, and oscillator strengths are given for all of the above transitions. The $3s$ lifetimes agree well with previous experimental results. The cascade lifetimes determined in a two-exponential model are consistent with known lifetimes of visible transitions.

INTRODUCTION

In this paper we report on direct measurements of the radiative lifetimes of the first nine lines of the "resonance" series of Ne I. A spectrum of Ne I is shown in Fig. 1. Energy levels in Ne I have been compiled by Moore[1] and an energy-level diagram is given by Condon and Shortley.[2] The ground state of Ne I is $1s^2 2s^2 2p^6\ {}^1S_0$ and dipole transitions between excited states and the ground state are allowed only from ${}^1P_1^0$ components of the excited states. Intermediate coupling holds in Ne I and all $J=1$ levels will have significant ${}^1P_1^0$ components. Hence, all $J=1$ levels will make transitions to the ground state. The lines studied here originate in the $J=1$ levels of the $3s$, $4s$, $5s$, and $3d$ configurations. The two $3d$ levels which radiate near 619 Å are not resolved in the spectrum shown here, but were observed individually for lifetime measurements.

The experimental apparatus and method of data analysis have been described in a paper on Ar I lifetimes[3] (hereafter called the argon paper). The resonance transitions are excited by electron collision in low-pressure (~1 μ) neon and the time decay of photon emission is recorded in a multichannel analyzer after a sharp (~1 nsec) cutoff of the electron beam. An iterative computer program Frantic[4] is used to make a least-squares fit of a sum (two or three components, some of which may be held fixed) of exponentials to the measured data.

The statistical error estimates to be quoted and referred to are calculated by Frantic and are *a priori* standard deviations σ_i of the points. These include uncertainty in the channel widths and statistical fluctuation in channel count ($\sqrt{N}$ type). An additional $\frac{1}{2}\%$ error is added to these to allow for any systematic errors in calibration. The Frantic least-squares fit uses weights $W_i = 1/\sigma_i^2$ and the estimated standard deviations in the parameters are obtained from the diagonal elements of the inverse of the least-squares matrix.

The major interpretative complications are radiative cascading and resonance photon entrapment. These will be discussed in the body of the paper, and our results will be compared with some previous determinations of the oscil-