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Radiative Lifetimes in the Resonance Series of Ne

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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-

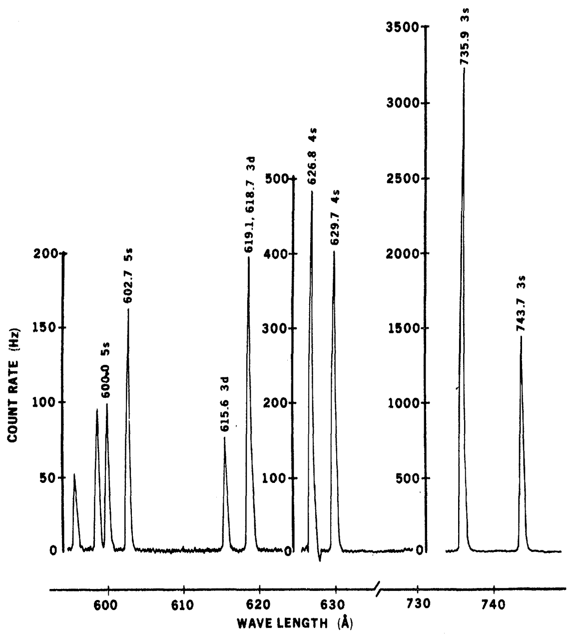

FIG. 1. Ne I lines excited by 150 V electrons. Gas pressure is 8 μ, causing some attenuation of the strongest lines.

lator strengths for the $3s-2p$ transitions.

EXPERIMENTAL TECHNIQUE

Some minor changes from the previous apparatus are as follows. The pulsed electron source is now a "planar" triode with a positively pulsed grid, whereas a diode was formerly used. With the present source the electron energy can be varied from near zero to +300 V, while the diode system was limited to the maximum pulse voltage, 50 V. The grid has a permanent dc bias of − 5 V (all potentials are referred to the filament) and the anode is normally kept near +160 V. An EH model 120D pulse generator with its output tubes paralleled, pulses the grid to + 40 V with a fall time of about 2 nsec, allowing a pulse of electrons to flow into the anode.

For measurements on the shortest lifetime reported here, 1.9 nsec, the grid pulse was shortened and sharpened by a diode clipping system. A 1N914 diode, forward biased at 50 mA, was put in series with the pulse generator output such that the pulse to the grid consisted of the stored charge of the diode. The pulse after shaping had a $\frac{1}{2}$ nsec fall time and a width of 2 nsec. The first dynode of the photomultiplier, which was used as the cathode, was vapor deposited *in situ* with a layer of CsI.

Several experimental parameters were varied in the course of measurement. The ranges were – pressure, 0.5–4.0 μ; repetition rate 2–5 MHz; channel width 0.35–0.80 nsec; pulse width 2–20 nsec. No significant variation was found, except for pressure, with any of the parameters. The statistical analysis in Frantic yielded error estimates consistently at or below 2% of the main lifetime measured.

DATA ANALYSIS

It is possible in principle to eliminate completely the problem of cascading simply by using electron energies only slightly above the threshold of the level to be examined. However, there is frequently not enough signal strength if this is done. A gain of an order or two of magnitude can be realized with uncontrolled excitation, and the problem of cascading is handled in the data analysis. The resonance lines measured have relatively simple cascading patterns and are fit well by a two-exponential model as shown in Fig. 2 for λ629.73 Å. The peak count in the channels is 2500. The channels are about 0.77 nsec wide and the full width at half-maximum of the excitation pulse is 24 nsec. The straight lines are determined by Frantic, and represent decay constants of 9.15 ± 0.18 nsec and 94 ± 9 nsec, the first being the main lifetime τ of the 4s level, and the second T the cascade lifetime. As shown in Table I, the value of T is sensitive to the record length used, but τ is rather insensitive. In most of the lines we have measured, the cascading lifetimes are less than the record length used in the fit and they are fairly well determined. The cascade lifetime determined by Frantic is the weighted average of a number of lifetimes and some longer lifetime components will not be well determined even with the longest possible record length used in the least-squares fit. This is evidenced by a large, uniformly positive difference between observed and calculated values in the tail of the decay curve. A three-component fit is sometimes better than a

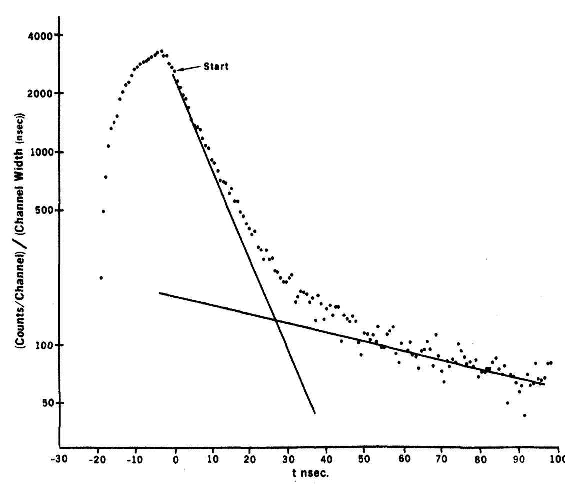

FIG. 2. Observed decay curve for Ne I, λ629.73

TABLE I. Dependence of Frantic lifetimes on record length.

Record length (nsec)	τ	T	χ^2 (modified)
50	8.98 ± 0.39	76 ± 37	1.22
67	9.08 ± 0.24	88 ± 19	1.09
98	9.15 ± 0.18	94 ± 9	1.14

a fit with two components, in the sense of yielding smaller chi squared, but the amplitude of the third component is always very small, the third amplitude and lifetime are not well determined, and the presence of a third component does not change the main lifetime by more than its error estimate.

PRESSURE DEPENDENCE OF THE LIFETIME

Resonance entrapment in the source is found to cause an increase in observed decay lifetimes as the source pressure is increased. For purposes of extrapolation to zero pressure, the Holstein formula[5] for lifetime expansion can be approximated by a linear lifetime versus pressure variation if the observed lifetime τ is kept less than a factor of 2 larger than the natural lifetime τ_0. The source was made small ($2 \times 2 \times 15$ mm) and the sample gas pressure as low as practical in order to accomplish this. The Holstein path length ρ for the source is on the order of 1 mm as shown in the argon paper. Lifetimes measured at pressures between 0.5 and 2 μ were fit to a relation $\tau = \tau_0 + bp$, where p is the pressure and b is a constant. With the exception of 735.89 Å for which measurements were made up to $\tau/\tau_0 = 2$, measurements were made with $\tau/\tau_0 < 1.1$. With such a small range in lifetime, the values of the slope b are statistically ill-defined, but have little effect on the value of τ_0.

BRANCHING RATIOS AND OSCILLATOR STRENGTHS

To transform the measured lifetimes to oscillator strengths, branching ratios have been calculated from cascade lifetimes determined in the Coulomb (Bates-Damgaard)[6] approximation. We have applied this approximation in L-S coupling using average energies. The spin-orbit term introduces singlet-triplet splitting only by wavelength factors. Lifetimes gotten in this way (except for those levels which make transitions to the ground state) are 4*s*, 19 nsec; 5*s*, 60 nsec; 6*s*, 63 nsec; 3*p*, 24 nsec; 4*p*, 97 nsec; 5*p*, 55 nsec; 3*d*, 23 nsec; 4*d*, 59 nsec; 5*d*, 92 nsec. For comparison, the 3*p* lifetimes determined experimentally[7,8] range from 15–25 nsec. The cascade lifetimes calculated are listed as Calc T's and they are in qualitative agreement with the Frantic-determined cascade lifetimes. The amount of cascading relative to the direct transition was found to be an order of magnitude less than for the corresponding cascades in Ar I. Consequently, the cascade correction introduces less uncertainty for these Ne I lifetimes than was the case in Ar I.

For those lines with no quoted error in the T's, the parameter has been held fixed at a value suggested by the B-D approximation to get a better fit. For the line at 743.70 Å, the cascade lifetime was less than, but quite near the main lifetime, and Frantic was not able to discern between the two components if it operated unconstrained. Changing the fixed T by ± 4 nsec did not change the main lifetime by more than its error estimate. For the 5*s* levels, T was arbitrarily held fixed at a value longer than the record length. In neither of these cases is the interaction between the two lifetimes troublesome, but we have tried to account for it in the error estimate of λ743.70 Å.

For the *ns* levels, theoretical relative oscillator strengths are easily obtained from the formula of Peterson.[9] Agreement is fair for 3*s* and 4*s*, better for 5*s*. For the *nd* levels, pair coupling[10] predicts equal strengths for the three components. However, we undertook a diagonalization of the spin-orbit matrix (with a least-squares fit to the observed energies) in order to obtain spin-orbit parameters and thus calculate the coefficients $|\langle 3d,\, {}^1P_1^0 | 3d, J=1\rangle|^2$ which determine the relative strengths of the lines. These are 0.266, 0.552, and 0.182 for the lines which radiate at 615.62 Å, 618.67 and 619.09 Å, respectively. These results and our experimental lifetimes indicate that pair coupling is not a good assumption in this case. The spin-orbit parameters obtained from our diagonalization are $\zeta_d = 0.000$, $\zeta_p = 521.427$, $F_2 = 16.14$, $G_1 = 2.97$ and $G_3 = 0.059$. These are in reasonable agreement with the results of Condon and Shortley[2] who quote $\zeta_d = 1.178$, $\zeta_p = 530.11$, $F_2 = 15.51$, $G_1 = 2.98$, and $G_3 = 0.002$.

ERROR ESTIMATES AND COMPARISON OF RESULTS

The errors (in the lifetimes) quoted in Table II are twice the error of the pressure fit made to the Frantic-determined lifetimes, or 5%, whichever is larger. They are upper bounds on the possible error. Cascading is the major source of uncertainty, and we have tried to estimate its effects in these error limits.

Comparison of our oscillator strengths for the 3*s* levels (which are not affected by branching) with several experimental and theoretical results is given in Table III. The Hartree-Fock calculations of Gold and Knox[11] should be accurate, as quantum defect plots[12] reveal no configuration perturbations at the 3*s* levels. Our value of the 3*s* oscillator strength is in excellent agreement

TABLE II. Ne I lifetimes and oscillator strengths.

nl	(Å)	τ (nsec)	Branching A[d] (10^6/sec)	f value ($\times 10^3$)	Theory relative f values	T (nsec)	Calc[d] T's (nsec)
$3s$	743.70	31.7 ± 1.6	0	7.8 ± 0.4	7.8	20	19, 23, 25
$3s$	735.89	1.87 ± 0.18	0	130 ± 13	102[a]	16 ± 4	19, 23, 25
$4s$	629.73	9.67 ± 0.50	55	8.6 ± 1.0	8.6	140 ± 20	60, 95
$4s$	626.82	7.78 ± 0.80	55	13.0 ± 2.0	11[a]	90 ± 8	60, 95
$3d$	619.09	13.2 ± 0.6	43	5.7 ± 1.0	5.7	c	55, 75, 134
$3d$	618.67	7.25 ± 0.6	43	16.0 ± 2.0	17[b]	c	55, 75, 134
$3d$	615.62	12.3 ± 0.6	43	6.4 ± 1.0	8.8[b]	c	55, 75, 134
$5s$	602.73	19.5 ± 0.5	17	5.7 ± 1.0	5.7	170	63, 91, 170
$5s$	600.04	23.1 ± 1.5	17	4.2 ± 1.0	4.1[a]	170	63, 91, 170

[a]Peterson formula (Ref. 9).
[b]Intermediate coupling (This work).
[c]No detectable cascading.
[d]Coulomb approximation (Ref. 6).

TABLE III. Comparison of oscillator strengths for the $3s$ levels.

	743.70	735.89
This work	0.0078 ± 0.0004	0.130 ± 0.013
HF (Gold, Knox)		
Theory (1) (Ref. 11)	0.011	0.110
Theory (2) (Ref. 11)	0.012	0.120
Intermediate Coupling (Ref. 10)	0.035	0.140
Line broadening (Ref. 14)	0.012 ± 0.002	0.168 ± 0.02
Electron impact (Ref. 13)	$f_{743}+f_{735}=0.14 \pm 0.01$	
f-sum rules (Ref. 12)	$f_{743}+f_{735}=0.163$	

with the experimental work of Geiger.[13]

Lewis[14] has made an analysis of rare-gas line-broadening data and has given f values for the $3s$ lines of Ne I and the $4s$ lines of Ar I. The geometric mean of Lewis's four-f values is a factor of 1.27 larger than the geometric mean of the corresponding numbers reported here and in the argon paper. It seems doubtful that the scale of the lifetime measurements could be this much in error.

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