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# The Mean Lives of Some Excited Levels in N I*

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## Abstract

We measured the mean lives of two low quartet and two low doublet excited levels of N I using the beam-foil technique. The transitions observed, their wavelengths in Å, and the mean lives of the upper levels in nanoseconds are:

| | | |
|---|---|---|
| $2s^2 2p^3\ ^4S^o - 2s^2 2p^2\ (^3P)3s\ ^4P$ | 1200Å | 2.4 ± 0.1 |
| $2s^2 2p^3\ ^2D^o - 2s^2 2p^2\ (^3P)3s\ ^2P$ | 1494Å | 2.2 ± 0.1 |
| $2s^2 2p^3\ ^4S^o - 2s 2p^4\ ^4P$ | 1134Å | 7.0 ± 0.2 |
| $2s^2 2p^3\ ^2D^o - 2s^2 2p^2\ (^1D)3s\ ^2D$ | 1243Å | 2.6 ± 0.1 |

These experimental results differ from theory by factors as large as 4.

## INTRODUCTION

There have been substantial difficulties in the way of obtaining reliable transition probabilities for electronic transitions, especially where the radiation lies in the vacuum ultraviolet. A good example of the discordant values given by experiment and theory is offered by the N I transition $2p^3\ ^4S^o - 3s\ ^4P$ (λ1200Å). An early Bates-Damgaard Coulomb approximation calculation[1] gave a total oscillator strength of 0.12; a measurement,[2] based on absorption in a gas cell, gave the conflicting value of $10^{-4}$. Recalculation,[3] using the self-consistent field approach, confirmed the original theoretical oscillator strength, and subsequent experiments[4,5] agreed with the theory.

The ultimate concordance of the foregoing theoretical and experimental results was not definitive, however. Lawrence and Savage,[6] using a phase-shift technique to measure the mean life of the $3s\ ^4P$ multiplet, deduced a transition probability larger by a factor of 2.3 than that found in Refs. 3 and 4, while still another experiment, based on the emission of light from a stabilized arc,[7] gave an even larger value of A (2.9 times that of Refs. 3 and 4). It is worth noting that a different set of calculations,[8] on other N I multiplets yielded transition probabilities several times larger even than those measured by Lawrence and Savage.

It is, therefore, abundantly clear that there is need for additional work on electronic transition rates. The theoretical calculations must take account of configuration interaction,[9] while from the standpoint of experiment a consistent and reliable method which has widespread applicability is required.

The experimental demands can be satisfied to some extent by the techniques of beam-foil spectroscopy.[10] These techniques offer an independent means of determining transition probabilities and are well suited to cases in which the observed radiation lies in the vacuum ultraviolet, in which the levels of interest are in multiply-ionized emitters, and in which the decays do not necessarily connect to the ground state. The present paper describes beam-foil measurements of the mean lives of a number of multiplets in N I; the N I multiplet discussed above is one of those studied in this paper.

## EXPERIMENT

Nitrogen atoms were obtained by partial neutralization of $^{14}N^{+}$ ions in a thin carbon foil (10 μg $cm^{-2}$) after acceleration to 2 MeV with a Van de Graaff accelerator. The radiation emitted by the excited levels was observed with a one-meter McPherson spectrometer (600 lines/mm grating blazed at 1500Å in first order). The detector was an EMR 542-G photomultiplier. The D.C. output was amplified by a Keithley picoammeter and recorded on a strip-chart, the analyzed spectrum being obtained by rotation of the grating. In order to measure mean lives the spectrometer was set to pass the required wavelength while viewing a 5 mm length of the beam. The mean lives were determined by measuring the photomultiplier signal obtained by observing the beam at different points downstream from the foil, the line of sight being perpendicular to the beam direction. The beam particle current was measured with an unshielded Faraday cup to assure its constancy. Data for each decay curve were obtained for about 14 different positions from the foil, corresponding to about 4 cm of the beam by photon counting. The energy of the particles before passing through the foil is known in this work to ± 4%. The energy loss in the foil[11] is known to within 15%, most of the uncertainty being in the measurement of its thickness, and thus the final speed is known to ± 3%. Its variation during a mean life measurement is less than 1%.

## RESULTS

### Spectra

The particles leaving the foil are in many different charge states. Although previously the carbon foil has been used for "stripping" electrons from singly-ionized atoms passing through it, a certain fraction of the ions gain an electron, leaving them in neutral states as they leave the foil. As the energy decreases the fraction of the total number of atoms that are neutral increases. Although this has been known for some time, initial spectroscopic studies of the foil-excited beam gave little indication that neutral particles were in excited states. However, most of the previous experiments were concerned with the wavelength range above λ3500Å. In this research, where the vacuum ultraviolet region was examined down to 1050Å, many lines were seen which could be attributed to neutral nitrogen. Thus the earlier difficulties in detecting excited neutral emitters may have been due to small oscillator strengths for the transitions in the wavelength regions observed.

Figure 1 illustrates part of the observed intensity distribution for the radiation emitted from 250 keV $N^+$ ions after passing through the foil. The wavelengths were calibrated from lines of unambiguous origin, Ly α, for example, giving a wavelength uncertainty less than 1Å. In the spectral region between 1050Å and 1750Å, the most intense lines (Fig. 1) are at λ1035Å, λ1134Å, λ1184Å, λ1200Å, λ1243Å, and λ1493Å. These are identified as the resonance line of N II at λ1085Å, and lines of N I at λ1200Å, λ1134Å, λ1493Å, and λ1243Å. Other less intense lines were identified with the de-excitation of neutral nitrogen atoms to the metastable levels $2p^3\ ^2P^o$: λ1311Å, λ1412Å, and λ1743Å. Other weak lines, just above the noise signal coincided with unclassified transitions in N I, for example, λ1561Å, λ1653Å. The line at λ1243Å appears strong at much higher beam energies, but with higher resolution, it could be decomposed into two lines at 1238.5Å and 1242.5Å, which we attribute to a doublet in N V. At 250 keV the line at 1238.5Å was absent and we assumed that the line at 1243Å was caused entirely by the N I transition $2p^2 3s\ ^2D \rightarrow 2p^3\ ^2D^o$.

## Mean Lives

### $2p^3\ ^4S^o$ - $2s^2 2p^4\ ^4P$, λ1134Å

Figure 2 shows the results obtained from measuring about 20 points. The mean life 7.0 ± 0.1 nanoseconds was determined with both a computer least-squares fit, and graphically. No cascade effects were seen over a distance corresponding to three times the 1/e decay length and were negligible. This mean life was measured at two different incident particle energies, 250 keV and 500 keV. Taking into account the energy loss in the foil, we obtained the same value for the mean life. Although we expected a contribution from the transition $2s3s\ ^3S$ - $2p3s\ ^3P^o$ of N IV at this higher energy, the equality of the two values for the mean life suggests that there was a negligible contribution from the N IV transition.

### $2p^3\ ^4S^o$ - $3s\ ^4P$, λ1200Å

The intensity decay is not exponential but a good least-squares fit was obtained using two exponentials, one corresponding to the approximately exponential decay of the tail of the curve, and a shorter decay which we attributed to the mean life of the 3s $^4P$ level. The mean life associated with the level was 2.4 ± 0.1 nanoseconds, and that for the long-lived component 37 ± 10 nanoseconds. The latter corresponds perhaps to one or more cascades from the levels 3p $^4S^o$, $^4P^o$, $^4D^o$, since these levels have no other decay modes. (Transitions to the 2s2p$^4$ $^4P$ level require a two-electron transition, and are unlikely in the freely decaying beam). The mean lives of these levels are known from Richter[12] to be 28.7, 43.8, 52.6 nsec respectively, consistent with our observations on this cascade lifetime.

### $2p^3\ ^2D^o - 3s\ ^2D$, λ1243Å

The decay curve was fitted with two exponentials, one a short-lived decay attributed to the mean life of the 3s $^2D$ level, 2.6 ± 0.1 nanoseconds, and a long-lived decay of 51 ± 10 nanoseconds, attributed to cascading into the upper level of the transition. As noted earlier, we exclude any contribution from N V.

The correction to the mean life by accounting for the cascade term was 30% even though its initial intensity was only one-tenth that of the fast decay. This is a good example of the care that must be taken to correct for cascades.

### $2p^3\ ^2D^o - 3s\ ^2P$, λ1493Å

This line is affected by cascades (Fig. 2). The extent of the cascade contribution was determined from points further than 1.5 cm downstream from the foil. A least-squares analysis gave the mean life of the 3s $^2P$ level as 2.3 ± 0.1 nsec and the cascade term as 34 ± 5 nsec. The three levels 3p $^2D^o$, 3p $^2P^o$, and 3p $^2S^o$ have allowed transitions only to the 3s $^2P$ level. Their mean lives are around 50 nsec, in good agreement with the observed cascade

DISCUSSION - TRANSITION PROBABILITIES

The results are reproduced in column 3 of Table I. The comparison with the results of Lawrence and Savage[6] is satisfactory. None of the 4 multiplets were resolved. In L-S coupling the experimental decay gives the lifetime $\tau$ and the total transition probability $A = 1/\tau$ of the upper term, and the lifetime is the same for all levels in this term. There is some experimental evidence[13] that the mean lives are independent of the particular level in the upper term.

The transition probabilities (Table II) $2s^2 2p^2\ (^3P)3s\ ^4P - 2s^2 2p^3\ ^4S^o$ and $2s2p^4\ ^4P - 2s^2 2p^3\ ^4S^o$ have the same value. There are many calculations for the resonance line at 1200Å. The value $1.85 \times 10^8\ sec^{-1}$ determined in the Coulomb approximation by Bates and Damgaard[14] has been adjusted to 1.39 by Kelly,[3] who also used a Hartree-Fock approximation to obtain a value $1.53 \times 10^8\ sec^{-1}$. These theoretical results differ from our experimental result by factors of 3 to 4. This factor was considered possible by Kelly.

On the other hand, the value calculated by Kelly[3] for the transition probability of $2s2p^4\ ^4P - 2s^2 2p^3\ ^4S^o$ is apparently overestimated. Varsavsky[8] using a screened Coulomb potential produces an even higher result. Labuhn's[7] experiment gave a transition probability 1.5 times greater than this experiment.

The 3s $^2P$ level can depopulate to the $2p^3\ ^2D^o$ level giving the transition studied, but it can also go to the $2p^3\ ^2P^o$ level. The branching ratio is given by Labuhn. It is in agreement with our observed intensity ratio of about 0.4 for the lines at 1743.6Å and 1492.6Å. However, this ratio will be dependent on a slight variation of instrumental efficiency with wavelength which we have not included in this comparison. Neglecting differences in background, this kind of determination of the relative transition probabilities of two or more decay modes is essentially independent of the point along the beam at which the pertinent line intensities are measured and of cascading into the upper state. In the present instance the background intensity was the same.

The $3s\ ^2D$, $4s\ ^2P$ levels have many possible depopulating transitions, but the principal decays are those to the metastable levels $2p^3\ ^2D^o$ and $2p^3\ ^2P^o$.

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# FIGURE CAPTIONS

Fig. 1. The beam-foil spectrum of nitrogen at 250 keV, λ1080 - 1750Å. Some identified transitions are indicated.

Fig. 2. Decay curves for two N I transitions.

Table I. Lifetimes of N I

| Levels | Wavelength Å | This Experiment nsec | Lawrence & Savage nsec |
|---|---|---|---|
| $2s^2 2p^2\ (^3P)3s\ ^4P$ | 1200 | 2.4 ± 0.1 | 2.5 ± 0.3 |
| $2s^2 2p^2\ (^3P)3s\ ^2P$ | 1494 | 2.2 ± 0.1 | 1.9 ± 0.3 |
| $2s2p^4\ ^4P$ | 1134 | 7.0 ± 0.2 | 7.2 ± 0.7 |
| $2s^2 2p^2\ (^1D)3s\ ^2D$ | 1243 | 2.6 ± 0.1 | 2.2 ± 0.3 |

Table II. Transition Probabilities

| Transition | λÅ | $A_{ik}$ (measured) $10^8$ sec$^{-1}$ Us | Lawrence | Labuhn | $A_{ik}$ (calculated) $10^8$ sec$^{-1}$ Kelly | Others |
|---|---|---|---|---|---|---|
| $2p^3\ {}^4S^o - 3s\ {}^4P$ | 1200 | 4.3 | 4.0 | 5.4 | 1.53 | {1.35[a]<br>{1.39[b] |
| $2p^3\ {}^2D^o - 3s\ {}^2P$ | 1494 | 3.3 | 3.9 | 5.5 | 2.4 | |
| $2p^3\ {}^2P^o - 3s\ {}^2P$ | 1743 | 1.2 | 1.4 | 2.0 | 1.1 | |
| $2p^3\ {}^4S^o - 2s2p^4\ {}^4P$ | 1134 | 1.45 | 1.39 | 2.3 | 8.44 | 18.1[c] |
| $2p^3\ {}^2D^o - 3s\ {}^2D$ | 1243 | 3.7 | 4.1 | 4.6 | 2.68 | |

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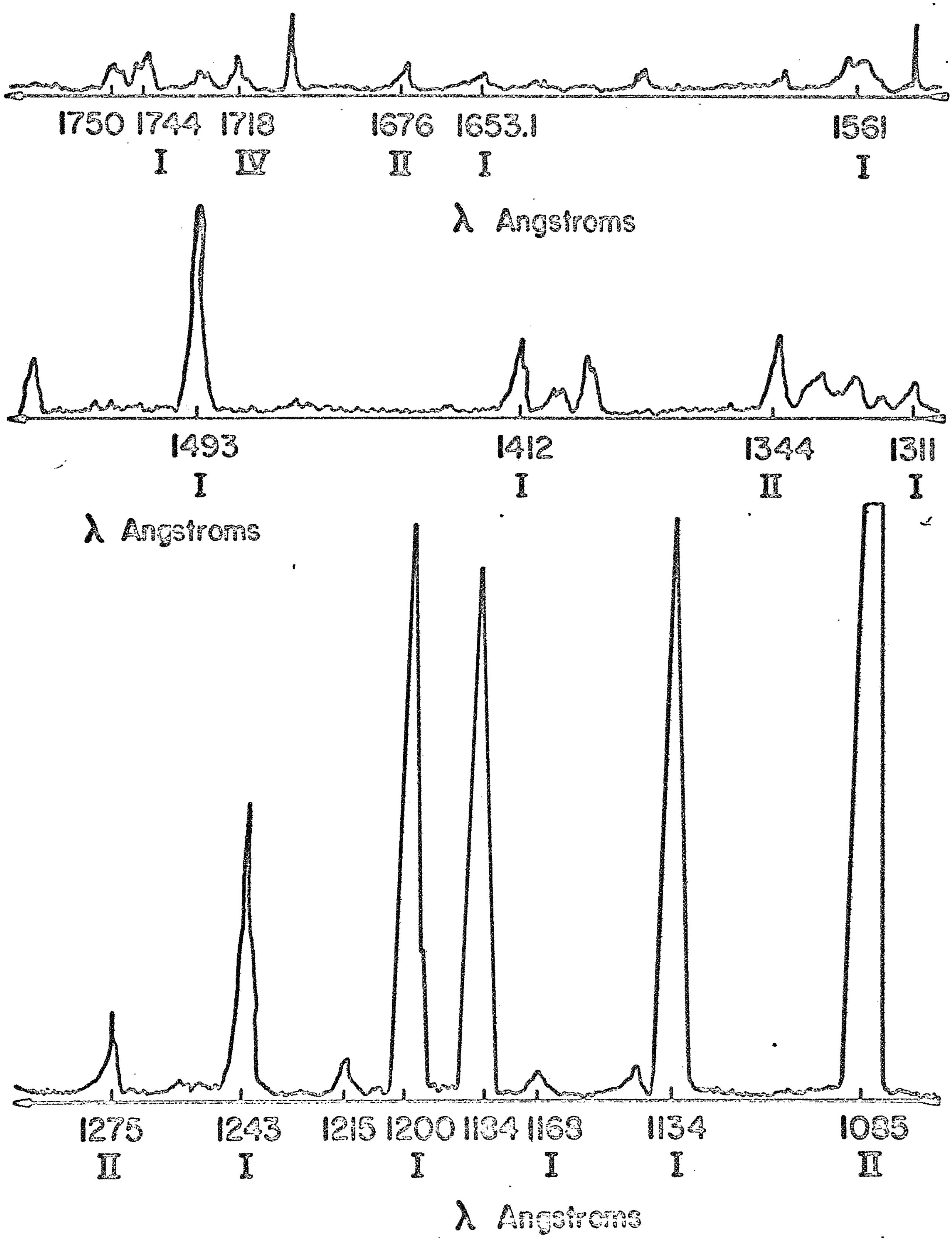
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I IV II I I
λ Angstroms
1493 1412 1344 1311
I I II I
λ Angstroms
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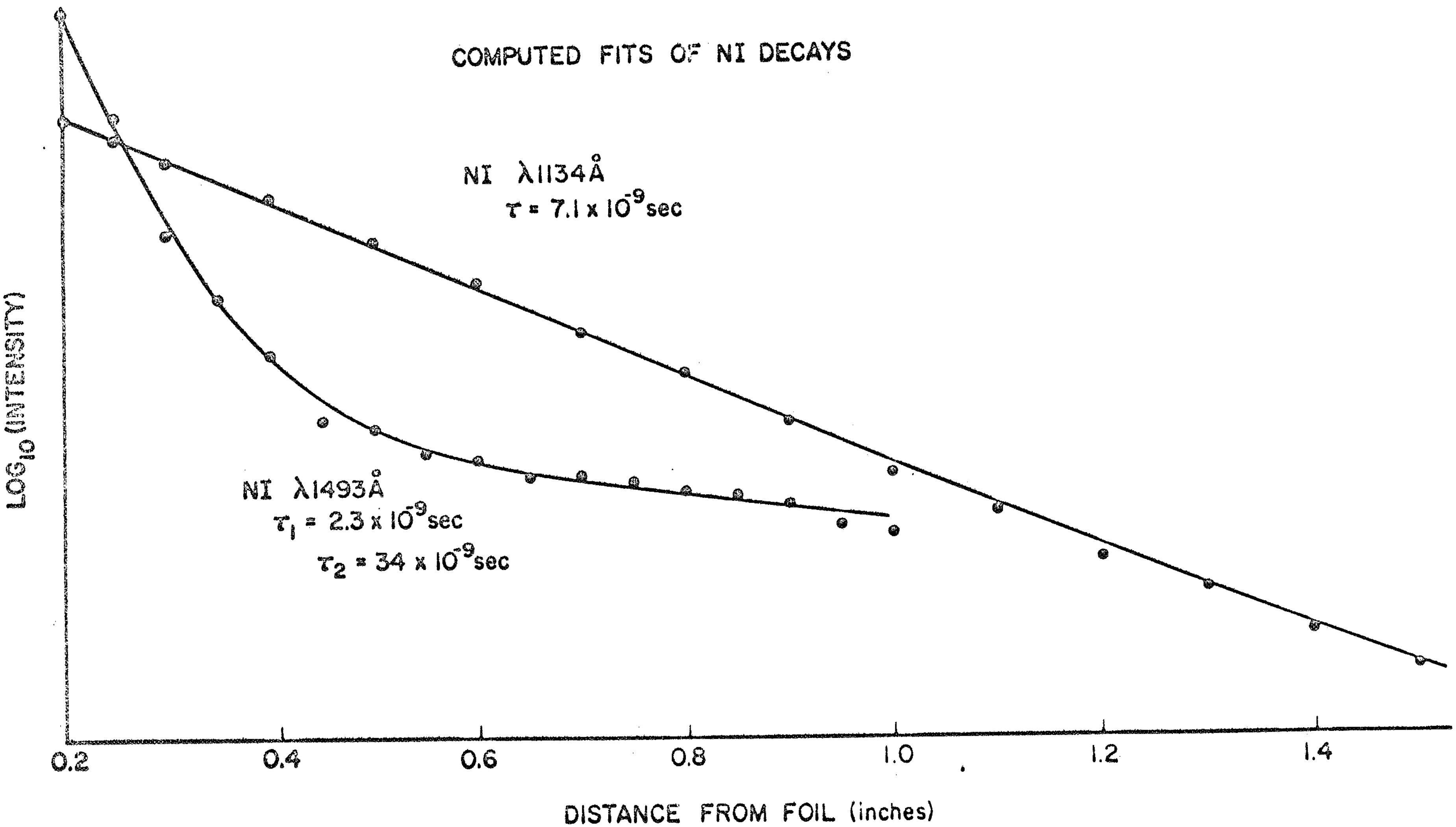
II I I I I II
λ Angstroms


COMPUTED FITS OF NI DECAYS
NI λ1134Å
τ = 7.1 x 10⁻⁹ sec
NI λ1493Å
τ₁ = 2.3 x 10⁻⁹ sec
τ₂ = 34 x 10⁻⁹ sec
LOG10 (INTENSITY)
0.2
0.4
0.6
0.8
1.0
1.2
1.4
DISTANCE FROM FOIL (inches)