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# Method of Incorporating Cascade Decay Curves into Atomic-Transition-Probability Measurements

J. L. KOHL,* L. J. CURTIS, R. M. SCHECTMAN, AND D. A. CHOJNACKI

*University of Toledo, Toledo, Ohio 43606*

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The transition probability of a cascade-repopulated level can be determined from decay curves of all transitions into and out of that level. This analysis is extended to situations in which a contributing cascade is unmeasured, yielding both the transition probability sought and the mean life of the missing transition. This method is applied to the measurement of the mean lives of the $2p_9$ and $2s_5$ levels of Ne I.

INDEX HEADINGS: Neon; Spectra.

Atomic-lifetime measurements are often hampered by the difficulties introduced by cascading transitions that repopulate the upper energy level of the transition of interest.[1–4] It can easily be shown[5] that each cascade transition (either direct or indirect) adds one exponential term to the expression for the time dependence of the light emitted by the transition of interest. The corrections to the apparent mean life of this level introduced by these terms are commonly as large as an order of magnitude.[6] Although it is theoretically possible to eliminate cascade transitions by excitation of the level of interest near threshold energy,[7] experimental limitations often preclude this possibility.[4,8]

Recently, a new method for obtaining transition probabilities from decay curves was presented[9] that incorporates the decay curves (irradiance vs time) of the cascade transitions into the analysis of the decay curve of the transition of interest. Although this new analysis technique accounts exactly for cascade effects, its application was limited to cases in which the decay curves of all of the significantly intense transitions into the level being investigated can be measured. Subsequent use of this analysis technique has brought to light certain modifications in its application that account for the effects of cascades on the measurement of the transition probability of a given level even when one contributing cascade transition is not observed experimentally. Moreover, when this is done, the decay curve of the unobserved transition can be indirectly measured. It is thus possible to measure the lifetime of the cascading level, even though no radiation from this level is detected. This aspect of the method makes it possible to measure decay curves with radiation outside the range of existing detectors and might lead to measurements of decay curves due to radiationless repopulation mechanisms. The purpose of this paper is to describe this extension of the new technique, and to illustrate its use by successfully applying it to measure the mean life of the $2p_9$ level of Ne I (Paschen notation).

Perhaps the most important aspect of the reported research is that the successful application of the extension of the cascade analysis encourages applications of the original analysis, particularly to vacuum-ultraviolet transitions of highly ionized species for which all of the necessary radiation is experimentally measurable.

## METHOD

Let $A_{NJ}$ denote the transition probability that is to be measured. Here $N$ represents the level from which the decay originated and $J$ denotes the final state. If the higher-lying levels that can cascade into the state $N$ are denoted by the index $k$, and the lower-lying states into which $N$ can decay are denoted by $j$, it has been shown[9] (in slightly different form) that

$$\frac{I_{NJ}(T)-I_{NJ}(0)}{A_{NJ}}=\sum_k \int_0^T I_{kN}(t)dt-\sum_j \int_0^T I_{Nj}(t)dt, \quad (1)$$

where $I_{nm}(t)$ represents the irradiance (in photons/s) emitted in the transition $m$–$n$ at time $t$, and $t=0$ and $t=T$ are two arbitrarily chosen observation times. If the decay curves of all transitions into and out of state $N$ are measurable, then every quantity in Eq. (1) except $A_{NJ}$ can be experimentally determined, and a value for this transition probability is obtained. If, however, one of the cascade transitions, denoted by $K$, cannot be measured, a new procedure can be adopted. Equation (1) can be applied successively for two closely separated values of the observation time $T$ and $T+\Delta T$ to obtain

$$\int_T^{T+\Delta T} I_{KN}(t)dt=\frac{I_{NJ}(T+\Delta T)-I_{NJ}(T)}{A_{NJ}} +\sum_j \int_T^{T+\Delta T} I_{Nj}(t)dt-\sum_{k\neq K} \int_T^{T+\Delta T} I_{kN}(t)dt. \quad (2)$$

Every quantity occurring on the right-hand side of Eq. (2) is measurable except for the transition probability $A_{NJ}$. Moreover, for sufficiently small $\Delta T$, the integral on the left-hand side of the equation is approximately $I_{KN}(T+\Delta T/2)\cdot\Delta T$. Thus, if $A_{NJ}$ is known, Eq. (2) allows the construction of the decay curve of the missing transition. In the case of a single exponential decay, the mean life $\tau_K$ and the decay constant $\alpha_K=1/\tau_K$ can be determined using the standard least-squares curve-fitting method. Alternatively, if $A_{NJ}$ is not known, Eq. (2) can often still be utilized to determine both $A_{NJ}$ and $\alpha_K$ provided that certain assumptions can justifiably be made about the missing cascade transition. In particular, we might assume that the repopulation of the upper level of the unmeasured

transition is negligibly small, resulting in a single exponential decay. This would be the case whenever the line-excitation function (i.e., the product of the transition probability and the excitation cross section) of the unmeasured transition is large compared with the line-excitation functions of the transitions that repopulate it. The necessary information concerning the unmeasured transition can be extracted from theoretical or measured estimates of line-excitation functions, and often can be directly determined by measuring the decay curve of a transition out of the same upper level as the undetected transition. In the latter case, the decay constant $\alpha_K$ can be independently determined so that a normalization factor would be the only remaining obstacle to determining the transition probability $A_{NJ}$ using Eq. (2).

For the case in which the unmeasured cascade transition is so strongly excited, the following procedure for determining both $A_{NJ}$ and $\alpha_K$ is employed:

(a) All of the decay curves of the transitions involved in the right-hand side of Eq. (2) are measured.

(b) An estimated trial value is assumed for $A_{NJ}$.

(c) The decay curve corresponding to the unobserved transition is constructed by successive applications of Eq. (2).

(d) The process is repeated, varying the trial value of $A_{NJ}$ and determining corresponding decay curves of the unmeasured transition in an effort to search for the value of $A_{NJ}$ that yields the constructed decay curve that best fits a single exponential decay in the least-squares sense.

A number of obvious variations of this procedure also exists, including application to the case of several missing cascade transitions of similar and long lifetimes and the case of a missing transition with a more complex decay curve.

When all transitions are measured, it has been shown that Eq. (1) can be used directly to determine a unique value for the transition probability $A_{NJ}$. This cascade analysis requires only the measurement of transitions directly populating the primary level of interest $N$, but it properly corrects for every contribution to the population of level $N$ whether it be by direct cascades or from an atom that has undergone a series of cascading transitions with widely varying decay constants. This determination of $A_{NJ}$ requires only the experimentally measured decay curves, and no parametrization of the curves, such as a lifetime, is necessary. In addition, it is unnecessary to resolve spectrally all of the cascading transitions because the individual contributions are simply summed in Eq. (1). The extension of this cascade analysis to measurements involving undetectable transitions maintains the corrections and requirements of the more direct method except for corrections due to cascade transitions into the upper level of the unmeasured transition that are assumed to be negligible for the reasons described.

A consistency check on the extension of the cascade analysis is possible if we neglect all second-order cascading transitions (i.e., transitions only indirectly populating the primary level of interest). To this approximation, we can reconstruct the decay curve of the primary transition by using the theoretical expression for its decay curve given by[5]

$$I_{NJ}(t)=\left[I_{NJ}(0)-\sum_k \frac{I_{kN}A_{NJ}}{\alpha_N-\alpha_k}\right]\exp(-\alpha_N t)+\sum_k \frac{I_{kN}A_{NJ}}{\alpha_N-\alpha_k}\exp(-\alpha_k t) \quad (3)$$

and the measured decay curves. The decay constant of level $N$ denoted as $\alpha_N=\sum_j A_{Nj}$ can be determined by a relative intensity measurement and $A_{NJ}$ because the ratio of the intensities of any two transitions out of a common upper level is proportional to the ratio of their transition probabilities. The reconstructed curve and the directly measured curve should be nearly equal, provided that the assumption of negligible second-order cascading is justified. In practice, many of the decay curves of the cascades can be least-squares fitted to either a single exponential term, indicating negligible higher-order cascading, or to a relatively short-lived and a relatively long-lived exponential, interpretable as an indication of long-lived higher-order cascades. In either of these cases, higher-order cascading has a small contribution at times immediately following excitation, so that the agreement of the reconstructed and directly measured curves should be reasonably good for the earlier section of the decay, with poorer agreement in the tail.

## EXPERIMENT

In the reported research, a pulsed electron beam is employed to populate the excited states of the atoms of a low-pressure gas. The photons emitted by the excited atoms are spectrally selected by a grating monochromator and are detected by an ITT 4034 fast-rise low-noise multiplier phototube operated as a single-photon counter. The timing arrangement employed here is similar to the delayed-coincidence method originally used for the measurement of excited atomic-state lifetimes by Heron *et al.*,[10] and later developed by Bennett.[7] This technique has since been used by numerous workers and will be only briefly described here. In the present arrangement, the time-to-amplitude converter (TAC) is started by the "sync" output of the square-pulse generator used to provide the pulsed electron beam and the single-photon signals are used to stop the converter. The shut-off time of the beam pulses is less than 1.5 ns and the excitation is cycled at 50 kHz so that only a relatively short amount of time is required to collect a large statistical sample of TAC output pulses, which are stored according to pulse height in a multichannel pulse-height analyzer. In this way, the number of photons emitted from each wavelength-

selected atomic transition of interest, as a function of the time at which they are emitted, is obtained from a large number of statistically distributed delayed-coincidence events. The analysis of these intensity-decay curves is the basis of the present research.

Because the relative magnitudes of the decay curves from several spectral lines must be compared and because they cannot be determined simultaneously with the present arrangement, it has been necessary to provide a method for normalizing the data-collection times. Variations of atomic-number density and electron flux might otherwise make relative-intensity determinations impossible. For this purpose, a separate optical system is used to monitor the photon counts from a suitable spectral line of the atomic species of interest. The total number of monitor counts for each run serves as the normalization factor. The wavelength dependence of the quantum efficiencies of the combined monochromator and detector system must also be determined. This calibration, which has been fully described elsewhere,[11] utilizes an Eppley Laboratory standard of spectral irradiance in conjunction with a set of accurately calibrated neutral density filters to reduce the light levels to the single-photon range and high-speed (100 mHz) counting electronics. Research-grade neon gas entered a differentially pumped gas target located in a Faraday cage. Decay curves were measured as described. Typical operating pressure in the target was $10^{-4}$ torr although no apparent change of the shape of the 6402-Å decay curve was observed for pressure as high as $7\times10^{-3}$ torr.

## APPLICATION

The pulsed electron beam was used to excite neon atoms, and the mean life of the $2p_9$ level of Ne I was investigated. Because the method being tested is not restricted by cascading, the energy of the beam was chosen to be high above threshold excitation, so that many levels were excited. In the notation of the previous section, $N$ denoted the $2p_9$ level, which can decay only to the $1s_5$ level, previously denoted as $J$. The decay curve for the transition $J$–$N$ at 6402 Å was obtained,

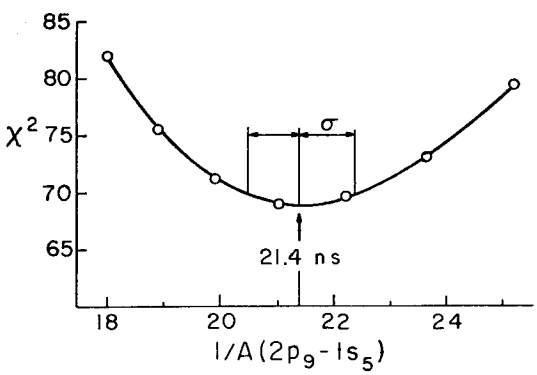


FIG. 1. $\chi^2$ vs trial value of $1/A(2p_9,1s_5)$ for fitting constructed decay curve of undetected transition to a single exponential decay curve. The abscissa has 0.529 ns per channel, and the observed standard deviation $\sigma$ is $\pm0.9$ ns.

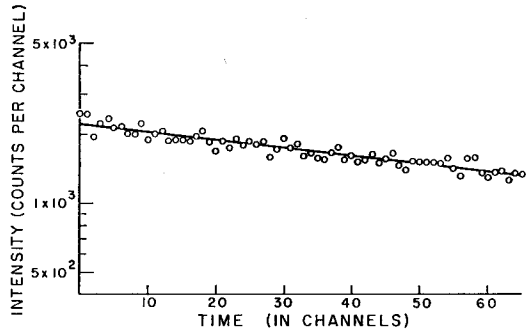


FIG. 2. Decay curve of the unobserved transition 11 180 Å, constructed by use of the new analysis technique. The best fit to a single exponential decay was obtained for $A(2p_9,1s_5)=(21.4\pm0.9\text{ ns})^{-1}$ and $\alpha(2s_5)=(239.\pm11\text{ ns})^{-1}$. The abscissa has 0.529 ns per channel.

as were decay curves for cascade transitions at 11 narrow wavelength bands between 4712 and 8377 Å.[1] Several of these decay curves may be due to unresolved blends of transitions into the $2p_9$ level, but it has been shown that this is permissible for the present analysis. Cascade decay curves within the sensitive range of the detector, but too weak to be observed, can easily be shown to make a negligible contribution to the population of the $2p_9$ level,[1] and consequently they were neglected. One important cascade transition, that from the $2s_5$ level (denoted by $K$ in the previous section), occurs at 11 180 Å. This is far beyond the infrared limit of the detector employed; hence it was not observed. However, the decay curve of this unmeasured transition is expected to be a single exponential decay, because the line-excitation cross sections for transitions into the $2s_5$ level are considerably smaller than the line-excitation cross section for the $2p_9$–$2s_5$ transition.[12,13] A very recent direct measurement of the $2p_9$–$2s_5$ transition decay curve using a cooled photomultiplier tube with an $S$-1 photocathode sensitivity has subsequently confirmed the single-exponential appearance of the $2s_5$ level of Ne I.[14] Thus, with this single undetected transition, the method described above can be utilized to determine both $A(2p_9,1s_5)$ and $\alpha(2s_5)$. The results of applying the method of the first section to the data obtained are summarized in Figs. 1 and 2. The former shows the value of chi square for fitting the constructed $2p_9$–$2s_5$ decay curve to a single exponential term, as a function of the assumed value of $A_{NJ}$. A sensitive variation is observed, and the best fit to the experimental data is given by the value $A(2p_9,1s_5)=(21.4\pm0.9\text{ ns})^{-1}$. The second figure shows the reconstructed decay curve for the single unobserved transition $2p_9$–$2s_5$ computed by use of Eq. (2) with the above value of $A(2p_9,1s_5)$. The decay curve appears to be a single exponential, which indicates that higher-order cascading is small, and yields a very accurate determination of the lifetime of the $2s_5$ level even though no radiation from the $2p_9$–$2s_5$ transition was detected. The value of $\alpha(2s_5)$ is relatively insensitive to the choice of $A(2p_9,1s_5)$ and is $(239\pm11\text{ ns})^{-1}$. This agrees well with the measurements of Chojnacki referred to above.[14]

The contribution of each term in Eq. (3) to the observed decay of the $2p_9$ level, $I_{NJ}(t)$, has been determined by fitting all 11 measured cascade decay curves and the reconstructed 11 180-Å decay curve to one or two exponentials yielding an initial intensity and lifetime for each cascade. The individual contributions of each cascade transition are shown in Fig. 3 as well as a comparison of the experimental data with the composite curve so derived. The decay curve predicted from Eq. (3) agrees well with the experimental data at the beginning of the decay with poorer agreement in the tail, as would be expected from combinations or indirect cascades into the $2p_9$ level.

The fitting of decay curves to sums of exponential terms is a commonly used technique for approximating the effects of cascading transitions on atomic-lifetime measurements. If the cascades have lifetimes that are reasonably different from the transition of interest, then the cascade contributions can be included as a single effective exponential and a two-exponential fit can be made. A simple two-exponential, four-parameter, fit to the 6402-Å decay curve has been made for comparison purposes and this yielded a very satisfactory fit with a value for the short-lived component of 26.2±0.6 ns and 230±9 ns for the longer-lived term. It can be seen from Fig. 3 that the rather large overestimate of the $2p_9$ lifetime that would have resulted from following this curve-fitting procedure is principally due to a blending of the 8377- and 6402-Å terms. This example demonstrates clearly the importance of including the measured decay curve of the cascade transitions in the data analysis of the decay curve of the transition of interest.

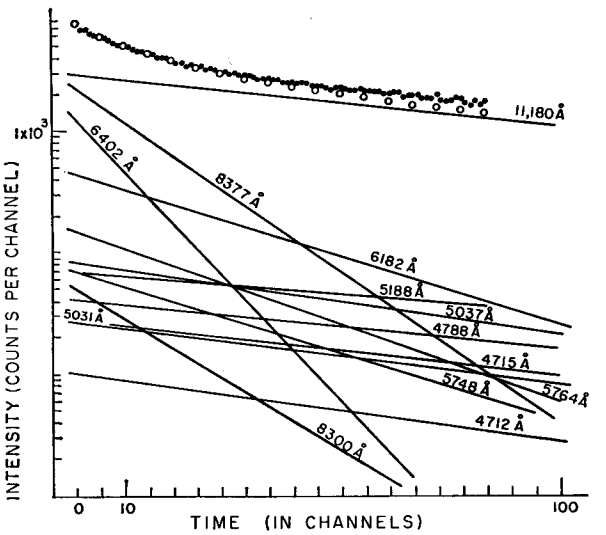


FIG. 3. Individual contributions to the decay curve of the 6402-Å transition of Ne I. Each solid line in the figure refers to an indicated term of Eq. (3), the open circles indicate the decay curve predicted from Eq. (3), and the filled circles make up the directly measured 6402-Å decay curve. The abscissa has 0.529 ns per channel.

## CONCLUSIONS

We have obtained values for the mean life of the $2p_9$ level of Ne I using a data-analysis technique that explicitly accounts for effects of cascade transitions. The resulting value of 21.4±0.9 ns is in good agreement with the results of Klose[15] (22.5±0.9 ns) and is also in reasonable agreement with the threshold measurement of Bennett and Kindlmann[7] (19.5±0.6 ns). We have also indirectly measured the mean life of the $2s_5$ level and this result (239±11 ns) is expected to be more reliable than the earlier measurement of Bennett,[16] which was subject to effects of collisional de-excitation. The indirectly measured value is in excellent agreement with Chojnacki's[14] value (240 ns) obtained by use of a more direct measurement.

The new method of analysis has allowed a measurement at high excitation energies that is comparable in accuracy to the results of the threshold measurements, and should be equally applicable to situations unsuited to threshold excitation. Moreover, this measurement demonstrates an analysis technique that allows construction of the decay curve of an important cascade transition in a situation in which the radiation from the transition cannot be observed. This aspect of the analysis may prove useful both for the study of lifetimes involving radiation outside the range of available detectors and for the possible study of radiationless de-excitation mechanisms. Furthermore, it offers considerable promise for lifetime measurements of highly ionized atoms for which cascade corrections often cause considerable difficulty.

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