Experimental lifetimes of some laser levels of Cl^+

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We have used the ion-beam method with a gas-jet target to measure the radiative lifetimes of three laser levels ($4p\ ^3P$, $4p'\ ^3D$, $4p'\ ^1F$) and two other levels of Cl^+. The levels and lifetimes in nanoseconds are: $4p\ ^3P$, 11.6 ± 0.6; $4p'\ ^3D_3$, 8.7 ± 0.9; $4p'\ ^1F_3$, 12.7 ± 0.6; $4p'\ ^3P_2$, 5.0 ± 0.6; and $4p''\ ^3P_2$, 6.7 ± 1.3.

INTRODUCTION

Experimentally determined lifetimes have long served as checks on atomic and molecular structure calculations and aids in the analysis of collision and radiative processes, such as those found in lasers. Also, lifetimes can be used in astrophysical calculations because of the ease with which they may be converted to oscillator strengths. For these and other reasons, experimental lifetime work can be beneficial.

In this paper we report beam-gas lifetimes for five levels of Cl^+. A few Cl II lifetimes for different levels have been measured via vacuum-ultraviolet (vuv) transitions by Lawrence[1] and by Bashkin and Martinson.[2]

EXPERIMENTAL METHOD

A plasma of Cl^+ was produced by leaking BCl_3 into an 80-MHz rf ion source. The ions produced were then accelerated (24–30 kV), focused, and mass-analyzed using standard accelerator techniques. The Cl^+ beam then entered a collision cell where the ions were excited by collision with a gas jet of He directed perpendicular to the direction of propagation of the Cl^+ beam.

A. Collision cell

The gas-jet target (see Fig. 1) was inside a cylinder with one end open to a liquid-nitrogen cold trap and diffusion pump. In the wall of the cylinder there were two small holes to allow passage of the ion beam. At the end of the cylinder opposite the pump a hypodermic needle was inserted such that the end of the needle stopped short of the beam path. With the target constructed in this manner, the He streamed out as a high-velocity jet, crossed the path of the ion beam, and then was quickly evacuated by the diffusion pump.

B. Detection systems

After passing through the collision cell, the beam entered an observation chamber. Through a fused-silica window in the observation chamber, we measured the decay in intensity of a visible transition from a level of interest as a function of the distance downstream from the collision cell. The measurements were made using a thermoelectrically cooled EMI 9789 QA photomultiplier tube attached to a Jarrell-Ash $\frac{1}{2}$-m scanning monochromator (Ebert-mount). The whole optical system was mounted on a platform which could be translated parallel to the path of the ion beam.

In order to account for small changes in beam current and target pressure changes, we used a second detection system, consisting of an EMI 6256 QB photomultiplier attached to a $\frac{1}{4}$-m Jarrell-Ash monochromator, to measure the intensity of an appropriate transition at a fixed point in the beam.

From the data obtained from these two systems, the lifetimes were extracted by standard computer-fitting techniques.[3] More details on the experimental techniques and equipment may be found in Refs. 4 and 5 and in other references cited therein.

A partial scan of Cl II is shown in Fig. 2. The scan was taken less than 1 mm downstream from the exit aperture of the collision cell. The spectral window was 4.8 Å (reciprocal linear dispersion of 16 Å/mm). As an aid in establishing spectral purity, an up-to-date spectral analysis performed by Radziemski and Kaufman[6] was used.

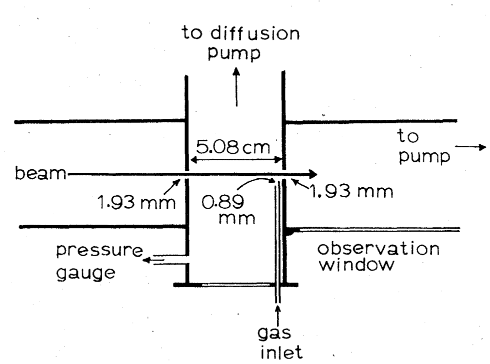

FIG. 1. Top view of a cross section of the gas-jet target.

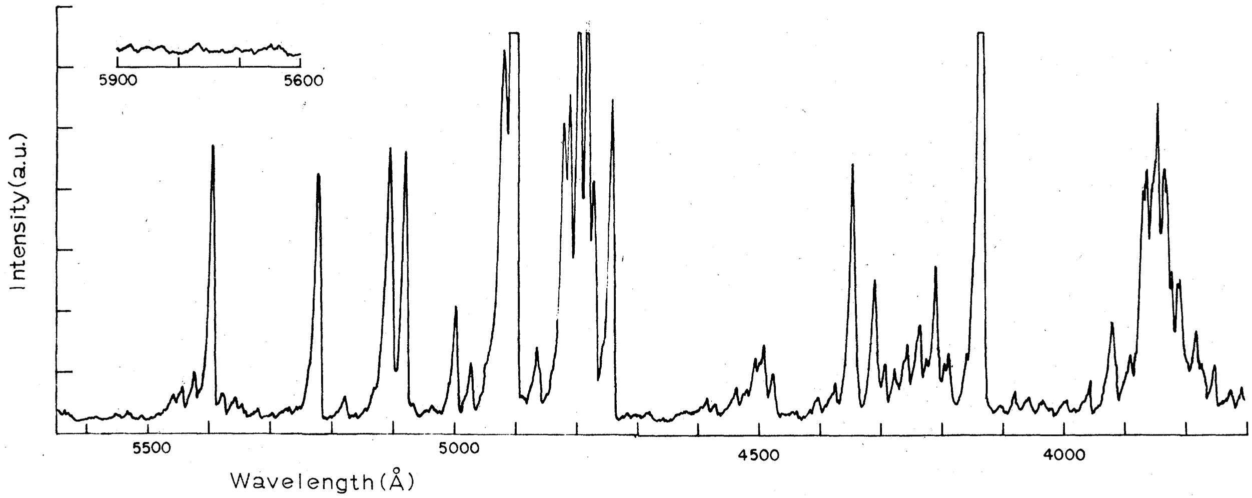

FIG. 2. Partial spectral scan of a 24.0-keV Cl^+ beam taken less than 1 mm downstream from the exit aperture of the gas-jet target. The spectral window was 4.8 Å. Intensity is in arbitrary units.

Because of the corrosive action of Cl on the ion source, long-lived and stable beams were difficult to obtain. Thus, this work was limited to reasonably intense lines which did not require narrow spectral windows.

A major concern with any gas-target excitation is the minimization of repopulation of the excited levels of the beam ions by target gas streaming into the observation chamber.[7,8] By consideration of the ratio of the cross-sectional areas of the needle and collision cell and the ratio of normal operating pressures of the collision cell (of the order of 10^{-3} Torr) and the observation chamber (of the order of 10^{-5} Torr), a conservative He number-density ratio of greater 10^4 was calculated for the beam-jet interaction region relative to the observation region. This calculation indicated that the effect of gas streaming was minimal. As a further check on the gas-streaming problem, we made spectral scans before each run to verify that no He lines were present. For example, note the absence of the 5876-Å line of He in Fig. 2.

The effect of gas streaming with only a gas-cell target was previously investigated by Head and Head[7] and by Clark and Head[8] using the 3889-Å and 5876-Å He emissions to determine the variation of the density of the He target gas as a function of the distance downstream from the exit aperture of the cell. They found that the streaming from the cell added two components to most decay curves. One could be approximated by a fast-decaying exponential $[A' \exp(-k_1 x)]$; and the other by a slowly decaying exponential $[B' \exp(-k_2 x)]$. The effect of the first could be eliminated by deleting from the decay curve points near the exit aperture or by regular curve fitting with high enough statistics. The slow component was indistinguishable from long-lived cascading and could easily be removed by standard curve-fitting techniques. With these procedures residual errors could routinely be brought down to the 10% range except for relatively short-lived levels.

The present experimental arrangement with the jet is a considerable improvement over the cell alone. Before, the target was a thick cell of low-pressure He gas with approximately uniform density, and the pressure differential between the target cell and the observation region was only in the range 10^2–10^3 to 1. With the present arrangement the effective beam interaction region is in the high-density gas jet near the end of the hypodermic needle. Of course, some of the gas from the jet forms a low-density "thick" target equivalent to that of the old arrangement, but the improvement in excitation in the target area relative to repopulation by the residual gas in the observation region is improved by at least a factor of 10^2 over the old arrangement. This is adequately substantiated by the inability to detect any He target emissions in the spectral scans of the beam in the observation region when using normal gains. The He emissions were present with the cell target.

We are convinced by these considerations, by the inability to detect any pressure-related effects, and by the success of the deletion-of-points fitting routine discussed below in removing fast components that no significant gas-streaming errors are present in our reported results.

TABLE I. Radiative lifetimes of some levels of Cl^+.

Level	λ (Å)	Lifetimes (ns) This study[a]	Theory[b]
$3p^3(^4S^o)4p\ ^3P_{0,1,2}$	5218–21[c,d]	11.58 ± 0.05 (5%)	11
$3p^3(^2D^o)4p'\ ^3D_3$	5078[c]	8.72 ± 0.22 (10%)	10
$3p^3(^2D^o)4p'\ ^1F_3$	5392[c]	12.70 ± 0.12 (5%)	11
$3p^3(^2D^o)4p'\ ^3P_2$	4344	5.04 ± 0.15 (12%)	10
$3p^3(^2P^o)4p''\ ^3P_2$	4208	6.7 ± 0.7 (20%)	≤6.6

[a] Errors following weighted means are standard deviations of means. Estimates of total uncertainties are given in parentheses.
[b] Reference 15.
[c] Known Cl^+ laser lines (Refs. 11–14).
[d] The 5218-Å line is the primary Cl^+ laser line.

ANALYSIS AND RESULTS

The decay curves are known to follow the form[7,8]

$$I = A_k \exp\left(\frac{-x}{v\tau_k}\right) + \sum_{j>k} B_{jk} \exp\left(\frac{-x}{v\tau_j}\right) + C\ ,$$

where I is the intensity of the decay from level k; j represents a cascade level; v is the speed of ions; x is the distance; τ_k is lifetime of level k of interest; τ_j is lifetime of cascading level j; and A_k, B_{jk}, and C are constants. Experimental data were fitted to the above equation using a program run on a DEC System 10 computer. The program minimized the least-squares sum by adjusting the parameters A_k, B_{jk}, C, τ_k, τ_j. The program also computed the χ^2 integral.

The largest errors in lifetimes measured by using processes involving nonselective excitation are usually caused by cascades which are inadequately accounted for in the curve-fitting procedures. Although long-lived cascades can frequently be extracted by high statistics and good curve fittings, short-lived (or growing-in) cascades and cascades from intermediate-lived levels can cause appreciable error if not detected and removed.

The effect of the growing-in cascade is most pronounced at the upstream end of a decay curve. By following the trends of the lifetimes with successive deletion of points from the upstream end of the curve, growing-in cascade can be detected and, if it exists, minimized. The legitimacy of this procedure was shown in an earlier paper[3] and was used for the Cl^+ results reported in Table I. Computer analyses using Monte Carlo simulated decay curves also substantiate the validity of our deletion procedure and computer-fitting programs.[9]

Background counts were small in comparison with the counts at the high-intensity end of the curves, the ratio typically being 1 to 10^2. Even when the deletion-of-points routine led to the discarding of the points with the highest signal counts, the background counts constituted less than 1–5% of the signal at the start of the decay curves. Nonbeam-related background was accounted for in the

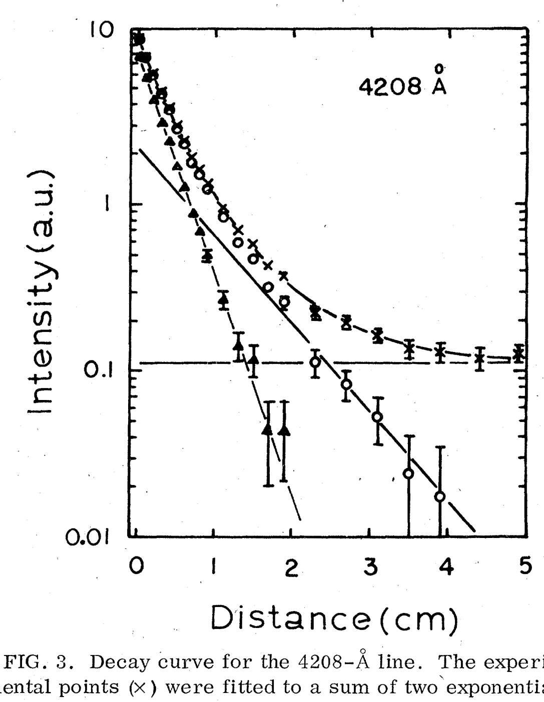

FIG. 3. Decay curve for the 4208-Å line. The experimental points (×) were fitted to a sum of two exponentials (sloped straight lines) and a constant (line of zero slope). The component identified with the decay of the $4p''\ ^3P_2$ levels is plotted with ▲'s. This particular fit gave a lifetime of 9.11 ± 0.54 ns. However, the computed lifetime dropped to 6.63 ± 1.47 ns after the four leftmost points were deleted. This trend was consistent for all decay curves taken on this level. The straight line passing through the ○'s is identified with intermediate-lived cascade.

fitting routine. We have not been able to detect any nonconstant background contributed by the beam and do not consider that such is likely, in contrast to the situation with a foil target.

Preliminary measurements for $4p\ ^3P$ and $4p'\ ^3D_3$ levels were reported earlier.[10] The new values remain essentially the same, except for a slight lowering of the $4p'\ ^3D_3$ value. The relatively large errors assigned to the $4p'\ ^3D_3$, $^3P_2$, and $4p''\ ^3P_2$ levels result from the presence of significant intermediate-lived cascades that were difficult to extract with our computer fits. See Fig. 3.

Transitions from the $4p\ ^3P$, $4p'\ ^3D$, and $4p'\ ^1F$ terms account for many of the known Cl^+ laser lines.[11-14]

We were unable to obtain measurements on other levels without excessive efforts because of intensity/spectral-purity problems. Close-lying lines from different multiplets made many of the strong lines in the scans in Fig. 2 unusable. Spectral windows small enough to eliminate overlap often gave signals too small to use. Some of the lines, of course, also originate from common levels.

We also show in Table I lifetimes computed from transition rates in the well-known NBS compilation.[15] These transition rates were calculated using the Coulomb approximation and were assigned uncertainties in the range 0–50%. For the most part our results are in relatively good agreement with the NBS work.

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