

## ACCURATE OSCILLATOR STRENGTHS FOR INTERSTELLAR ULTRAVIOLET LINES OF Cl I

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### ABSTRACT

Analyses on the abundance of interstellar chlorine rely on accurate oscillator strengths for ultraviolet transitions. Beam-foil spectroscopy was used to obtain  $f$ -values for the astrophysically important lines of Cl I at 1088, 1097, and 1347 Å. In addition, the line at 1363 Å was studied. Our  $f$ -values for  $\lambda\lambda 1088, 1097$  represent the first laboratory measurements for these lines; the values are  $f(1088) = 0.081 \pm 0.007$  (1  $\sigma$ ) and  $f(1097) = 0.0088 \pm 0.0013$  (1  $\sigma$ ). These results resolve the issue regarding the relative strengths for  $\lambda\lambda 1088, 1097$  in favor of those suggested by astronomical measurements. For the other lines, our results of  $f(1347) = 0.153 \pm 0.011$  (1  $\sigma$ ) and  $f(1363) = 0.055 \pm 0.004$  (1  $\sigma$ ) are the most precisely measured values available. The  $f$ -values are somewhat greater than previous experimental and theoretical determinations.

*Subject headings:* atomic data — ISM: atoms — ultraviolet: interstellar

### 1. INTRODUCTION

The chemistry of chlorine in diffuse interstellar clouds appears to be relatively simple (Dalgarno et al. 1974; Jura 1974; Jura & York 1978). In regions primarily composed of atomic hydrogen, Cl II is expected to be the dominant form of chlorine, while in molecular regions, the neutral species, Cl I and HCl, should dominate. The production of HCl is initiated by reactions involving Cl<sup>+</sup> and H<sub>2</sub>. Chlorine is unique among the abundant elements because this reaction proceeds at the fastest rate possible, the collision rate; the chemistry for all other abundant elements involves slower radiative processes or endothermic charge exchange. Although HCl has not yet been detected in diffuse clouds where visual extinctions are less than 1 mag, the available observations for Cl I and Cl II (Jura & York 1978; Harris & Bromage 1984) tend to confirm the basic ideas of this picture. One difficulty in reaching such a conclusion, however, is that the analyses have been based on rather poorly known oscillator strengths for Cl I, as has been emphasized by Keenan & Dufton (1990) and Morton (1991). In this paper, we present measurements of the fundamental atomic data necessary for carrying out more definitive analyses.

For the neutral chlorine lines at 1088, 1097, and 1347 Å of interest in the study of interstellar matter, there has been relatively little accurate information available concerning oscillator strengths. The situation for  $\lambda 1347$  is better than for the other two lines. Hofmann (1967) used a wall-stabilized arc to obtain the  $f$ -value for this transition and estimated that his result should be accurate to about 25%. Lawrence (1967) measured the mean life for  $\lambda 1335$  and computed the line strength ratio between that line and  $\lambda 1347$  with an intermediate coupling calculation to obtain an  $f$ -value of perhaps similar precision. Clyne & Nipp (1977) measured  $f(1347)$  by resonance absorption and obtained a precision of some 30%. The most accurate measurement reported is an improvement of the resonant absorption experiment of Clyne & Nip by Schwab & Anderson (1982), who quote 10% accuracy. A theoretical estimate of Ganas (1988), designed to generate excitation cross sections, is not expected to be more accurate than about 50%. The best current theoretical estimate for  $\lambda 1347$  is from the recent large-scale multiconfiguration calculation of Ojha & Hibbert (1990).

For the transitions at 1088 and 1097 Å, on the other hand, no previous experimental values have been reported. Estimates of the oscillator strengths have been extracted from astronomical data by requiring consistency between column densities obtained using observations of all three spectral lines. Jura & York (1978) deduced an  $f$ -value for  $\lambda 1097$  and Federman (1986) obtained a value for  $\lambda 1088$ . Both results were dependent upon the accuracy with which  $f(1347)$  was known at the time of the observations and upon effects of saturation of the observed absorption lines. Neither result is likely to be more accurate, therefore, than about 50%. The calculations of Ojha & Hibbert (1990) predict the  $f$ -value of  $\lambda 1097$  to be significantly larger than that for  $\lambda 1088$ , while the astronomical observations (Federman 1986) and the absorption measurements of Cantu et al. (1985) indicate that the reverse may be true.

The oscillator strengths of the Cl I resonance lines at 1088 and 1097 Å are difficult to compute from ab initio theory. These transitions involve levels in the  $3p^4(^3P)3d$  configuration, with excitation energies of 91,907 and 91,127 cm<sup>-1</sup>, both with angular momentum  $J = 5/2$ . This three-dimensional configuration mixes with  $3p^4(^3P)5s$  to form a complex of 22 even-parity levels between 88,000 and 93,000 cm<sup>-1</sup>. Among these are six levels with  $J = 5/2$ , whose LS-coupling designations are  $3d^2F$ ,  $4F$ ,  $2D$ ,  $4D$ , and  $4P$ , and  $5s^4P$ . However, the states are not well described by LS-coupling or by any other pure coupling scheme (such as JK-coupling which is used by some authors). In particular, the level at 91,907 cm<sup>-1</sup> has been classified as  $2D$  by Minnhagen (1961) and as  $2F$  by Radziemski & Kaufman (1969). The strong mixing which occurs among these six  $J = 5/2$  states not only confuses identification, but it also makes the computation of oscillator strengths unreliable. A large-scale configuration interaction calculation of Cl I by Ojha & Hibbert (1990) gives the best theoretical results for a large number of transitions. However, because of the effects already mentioned, these authors recommend caution in using their results for the strongly mixed levels in the  $3p^4(^3P)3d + 5s$  complex. As a prelude to our experimental work, we performed Hartree-Fock calculations with configuration interaction using both the well-known programs RCN-RCG (Cowan 1981) and MCHF77 (Froese Fischer 1969, 1978) with Pauli corrections (Ellis 1983). We also observed the sensitivity of the

RCG results to small arbitrary changes in Slater parameters. We find that the relative ordering of these  $J = 5/2$  levels is extremely sensitive to the strengths of the various interactions; it is our judgment that ab initio theory cannot reliably determine the eigenvectors of these states, nor therefore the corresponding transition probabilities. Laboratory measurements are clearly needed to determine the oscillator strengths for these lines.

In this paper, we describe the results of our laboratory measurements for the transitions at 1088, 1097, and 1347 Å, as well as that at 1363 Å. The experimental details are described in the next section, followed by a presentation of the results and a discussion of the consequences for interstellar studies.

## 2. EXPERIMENT

The VUV transitions described above were studied using the beam foil spectroscopy facility at the University of Toledo heavy ion accelerator. Beams of  $\text{Cl}^+$  ions at energies of 130 keV and 180 keV were neutralized and excited by traversal through thin ( $2 \mu\text{g cm}^{-2}$ ) carbon foils. An Acton 1 m normal incidence vacuum ultraviolet monochromator was used for wavelength selection. With 50  $\mu\text{m}$  entrance and exit slits, line profiles with FWHM of about 600 mÅ were obtained. Spectral lines in the region 1000–1100 Å were detected with a Galileo channeltron electron multiplier while those in the region 1300–1400 Å were measured using a Hamamatsu R1459 solar blind photomultiplier. Time-resolved decay curves were obtained by recording the intensity of the radiation selected as a function of the distance between the exciter foil and the monochromator entrance slits.

### 2.1. Transitions from the $3p^4(^3P)4s^2P_{3/2}$ Level

Decay curves for both transitions to the ground-state energy level  $3p^5^2P_{3/2}$  at 1347.24 Å and the transition to the higher lying fine-structure level  $3p^5^2P_{1/2}^o$  at 1363.45 Å were measured. The decays are essentially single exponential for times after excitation of less than 10 ns, with deviations due to cascade repopulation observed only for longer delays. The most significant sources of uncertainty in the mean life derived from the decay curves are the necessarily approximate treatment of the long-term time dependence and the knowledge of the velocity of the beam after traversing the foil (Federman et al. 1992). The limiting accuracy with which the mean life can be determined is about 5%. As described by Federman et al. (1992), possible systematic effects due to insufficiently accurate treatment of the energy loss in traversing the foil, to foil thickening and to beam divergence are investigated by carrying out decay curve measurements at two different beam energies and by translating the foil both in the downstream and upstream beam directions. Uncertainties here due to these causes are established to lie at the 1%–2% level. The measured value for the mean life of the  $3p^4(^3P)4s^2P_{3/2}$  excited level is  $1.51 \pm 0.07$  ns, where the quoted uncertainty here and elsewhere is  $1 \sigma$ .

To extract  $A$ -values for the two competing transitions, it is also necessary to know the branching ratio  $I(1347)/I(1363)$ . This intensity ratio was measured to be  $5.77 \pm 0.23$ , independent of distance from the foil. The detection efficiency of the monochromator-phototube combination is not expected to vary significantly over the small wavelength region between the two lines: the R1459 phototube has its maximum quantum efficiency at about 1360 Å and should not vary by more than 0.1%; the grating is blazed for 1500 Å in first order and consideration of typical grating performance (James & Sternberg

1969; Stroke 1967) suggests that the grating efficiency should change by less than 1% over this same interval. Consequently, no correction for variation in detection efficiency has been included.

### 2.2. Ground-State Transitions at 1088.06 and 1097.37 Å from the $J = 5/2$ Levels

The spectral feature at 1088.06 Å was strong and unblended. As for the two previous transitions, decay curves were measured at two energies and for different directions of travel. To improve the statistical accuracy, data from a number of decay curves measured with different foils were superposed. The resulting experimentally determined mean life was  $3.3 \pm 0.3$  ns.

On the other hand, the line at 1097.37 Å is very weak. Figure 1 shows the results of a superposition of many wavelength scans in this region. The feature at 1097.37 Å is clearly established, but is seen only as a shoulder on the stronger transition at 1098.07 Å. The strength of the line at 1097.37 Å was determined by carrying out a least-squares fit of the data to a superposition of spectral lines whose line centers were those tabulated by Kelly (1987) and whose shapes were obtained by study of the strong isolated line at 1088.06 Å. In the region 1092–1100 Å, 10 lines were superposed, and their intensities were varied to obtain the best fit. In this way, the ratio of the intensities of these two lines,  $R(1088/1097)$ , was measured at a number of foil positions corresponding to times after excitation of 0.4, 2.0, 4.0, 5.5, 7.0, and 9.5 ns. The ratio varied slowly with time, and extrapolation to  $t = 0$  gave a value for  $R$  as the atoms exited the foil of  $R = 9.3 \pm 1.1$ .

This ratio represents the product of the ratio of the respective transition probabilities with the ratio of the initial populations of the two  $J = 5/2$  excited states. While the foil excitation process is rather imperfectly understood, even for relatively simple systems (Veje 1985; Dybdall et al. 1986), it is very likely that the two  $J = 5/2$  levels arising from the same  $2p^4(^3P)3d$  configuration would be equally populated. Making this assumption results in a mean life for the excited state leading to the 1097 Å transition of  $30.7 \pm 4.6$  ns. A consistency check, showing that this hypothesis is reasonable, is shown in Figure 2. There, the intensity of the 1097 Å line is displayed as a function of time after excitation; superposed on the decay curve is a single exponential decay of 31 ns, normalized to the data. While the relatively poor statistical accuracy resulting

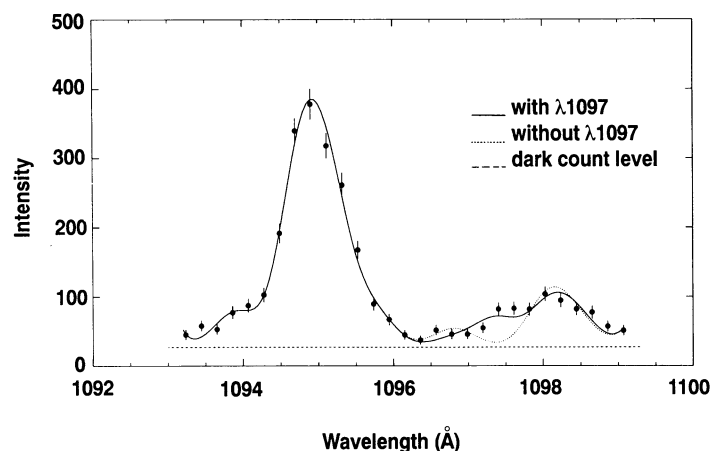


FIG. 1.—Chlorine spectrum in the vicinity of  $\lambda 1097$ . The solid curve shows the fit that includes the transition at 1097.37 Å, while the dotted curve is the fit without this transition. The dashed line indicates the dark count level.

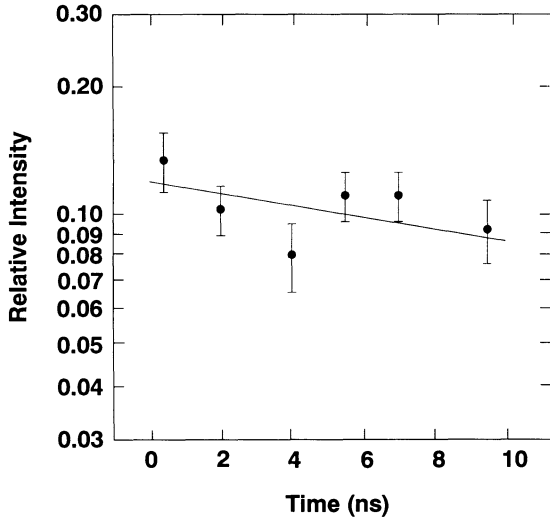


FIG. 2.—The decay curve for  $\lambda 1097$  obtained from the ratio of line intensities,  $I(1097)/I(1088)$ , normalized to  $I(1088)$ . The solid line shows a single exponential decay of 31 ns normalized to the data.

from the weakness of the spectral line prevents unequivocal conclusions from being drawn, the agreement shown is quite good. This qualitatively confirms the assumption made about the nature of the excitation process.

3. OSCILLATOR STRENGTHS

For the transitions from the  $3p^4(^3P)4s^2P_{3/2}$  level at 1347 Å and 1363 Å, the measurements of the mean life and the branching ratio suffice to determine the individual  $A$ -values and the oscillator strengths. The resulting  $f$ -values are presented in Table 1, together with previously available information.

To obtain the oscillator strength for  $\lambda 1088$  from the measured mean life, one must account for transitions to the lower lying levels of the  $3p^4(^3P)4p$  configuration. Because of the very small transition energies involved, the branching ratios to these other final states are much smaller than the 10% uncertainty in  $\tau$ , and one may therefore use  $A = 1/\tau$  for  $\lambda 1088$  to the accuracy quoted. The assumption of equal initial populations of the two  $J = 5/2$  levels built from the  $2p^4(^3P)3d$  configuration allows us to extract an  $f$ -value for  $\lambda 1097$  from the relative intensity measurement. Our results for these two transitions are compared with previous results in Table 2.

TABLE 1  
OSCILLATOR STRENGTHS FOR  $\lambda\lambda 1347, 1363$

REFERENCES	TRANSITION	
	1347.2 Å	1363.4 Å
This work .....	$0.153 \pm 0.011$	$0.055 \pm 0.004$
Previous results:		
Hofmann .....	0.090	0.035
Lawrence .....	0.113	0.039
Clyne & Nip .....	$0.10 \pm 0.03$	$0.038 \pm 0.006$
Schwab & Anderson .....	$0.109 \pm 0.010$	...
Ganas .....	0.138	0.028
Ojha & Hibbert .....	$0.132 (L)^a$	$0.049 (L)^a$
	$0.117 (V)^a$	$0.041 (V)^a$

<sup>a</sup> (L) is length form and (V) is velocity form.

TABLE 2  
OSCILLATOR STRENGTHS FOR  $\lambda\lambda 1088, 1097$

REFERENCES	TRANSITION	
	1088.06	1097.37
This work .....	$0.081 \pm 0.007$	$0.0088 \pm 0.0013^a$
Previous results:		
Federman .....	$0.069 \pm 0.020^b$	...
Federman .....	$0.094 \pm 0.026^c$	...
Jura & York .....	...	$0.014 \pm 0.004$
Jura & York .....	...	$0.019 \pm 0.005^e$
Ojha & Hibbert .....	$0.016 (L)^d$	$0.042 (L)^d$
Ojha & Hibbert .....	$0.006 (V)^d$	$0.017 (V)^d$

<sup>a</sup> Using assumed excitation.  
<sup>b</sup> Redetermined from original data.  
<sup>c</sup> Revised using  $f(1347)$  obtained in this work.  
<sup>d</sup> (L) is length form and (V) is velocity form.

4. DISCUSSION

Our  $f$ -values for  $\lambda 1347$  and  $\lambda 1364$  are systematically some 40% larger than those reported earlier, necessitating a reevaluation of astrophysical conclusions based upon the older, less accurate, results. In particular,  $f$ -values obtained from astronomical observations which rely upon knowledge of  $f(1347)$  must be revised. Agreement between our measurements and the recent theoretical predictions of Ojha & Hibbert (1990) is, for these lines, quite satisfactory. The disagreement between our measurement and that of the careful study by Schwab & Anderson (1982) is puzzling.

Our measured  $f$ -values for the transitions at 1088 and 1097 Å indicate that the relative strengths obtained from studies of interstellar spectra are correct: in particular,  $\lambda 1088$  is the stronger of the two lines. Adjusted to incorporate our new determination of  $f(1347)$ , the analysis of Federman (1986) yields a value for  $f(1088)$  of  $0.094 \pm 0.026$  which is in good agreement with the value reported here of  $0.081 \pm 0.007$ . A similar revision for the oscillator strength of  $\lambda 1097$  from the analysis of Jura & York (1978) gives 0.019, accurate to, perhaps, 25%; our result, however, is  $f(1097) = 0.0088 \pm 0.0013$ . The difference suggests that although Jura & York included the effects of saturation on  $\lambda 1347$  in their study, our larger  $f(1347)$  would require an even larger correction to the column density of Cl I from  $\lambda 1347$ . In particular, in order to bring the laboratory and astronomical results into agreement, the saturation correction appears to be about 3.0, as opposed to the value 1.5 used by Jura & York for the data toward  $\lambda$  Ori.

Although it is beyond the scope of this paper, we briefly comment on possible saturation effects in the astronomical studies of the 1088 Å line (Federman 1986). The lines at 1088 and 1347 Å are significantly stronger than the line at 1097 Å. These two stronger lines are undoubtedly saturated along many lines of sight; in fact  $\lambda 1347$  probably shows saturation effects for all the reported measurements. Preliminary estimates suggest that the neglect of saturation increases the uncertainty in the  $f$ -values obtained astronomically from the reported 25% to, perhaps, 50%. This issue will be developed more fully in a subsequent paper, where a study of chlorine chemistry based on our new  $f$ -values will be described.

In summary, we measured the lifetimes and deduced the oscillator strengths for the neutral chlorine lines at 1088, 1097, and 1347 Å, which are important to studies of interstellar matter. The line at 1363 Å was also studied. Our results for

$\lambda\lambda 1088, 1097$  are the first laboratory determinations; the results clearly establish that  $\lambda 1088$  is the stronger of the two lines, as had been inferred from astronomical observations. For  $\lambda\lambda 1347, 1363$ , our measurements are the most accurate laboratory data available. Our oscillator strengths for these two lines are somewhat larger than earlier determinations. Since our oscillator strengths differ from the values used in previous

studies of chlorine in diffuse interstellar clouds, the chemical picture that emerged from those studies will be reexamined.

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