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OSCILLATOR STRENGTHS FOR LINES OF THE (0, 0) AND (1, 0) BANDS OF THE $C^{1}\Pi - X^{1}\Sigma^{+}$ SYSTEM OF HCl AND THE ABUNDANCE OF HCl IN DIFFUSE INTERSTELLAR CLOUDS

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ABSTRACT

Oscillator strengths (f-values) for twenty lines of the (0, 0) band and one line of the (1, 0) band of the $C^{1}\Pi - X^{1}\Sigma^{+}$ system of HCl have been measured using quantitative photographic techniques. The f-value for the R(0) line of the (0, 0) band at 129.0257 nm, which is the stronger of the two lines that have been searched for but not detected in diffuse interstellar clouds, is 0.185 \pm 0.037. This value and the observations of absorption by chlorine species in the ζ Oph cloud have been used to reassess aspects of the chemical models of diffuse clouds.

Subject headings: interstellar: abundances — interstellar: molecules — laboratory spectra — transition probabilities — ultraviolet: spectra

I. INTRODUCTION

Column densities of molecules in diffuse interstellar clouds can be determined from measured equivalent widths only if accurate oscillator strength (*f*-value) data are available. This paper presents results of measurements of the *f*-values of rotational lines of the (0, 0) and (1, 0) bands of the $C \, {}^{1}\Pi - X \, {}^{1}\Sigma^{+}$ system of HCl at 129 nm and 125 nm, respectively.

Chlorine chemistry in diffuse interstellar clouds was first discussed in detail by Jura (1974) and by Dalgarno *et al.* (1974). The cloud models of Black and Dalgarno (1977) and of Black, Hartquist, and Dalgarno (1978) have made the only predictions of HCl column densities in specific clouds. The predictions for the ζ Ophiuchi cloud are not in accord with the observations by Wright and Morton (1979). This discrepancy and the implications of our results are discussed in § IV. Preceding that are § II, which sets forth the details of the measurement, and § III, a discussion of the data analysis and results.

A preliminary report of the *f*-value data has been given by Smith, Yoshino, and Parkinson (1980), and comments on chlorine chemistry in diffuse interstellar clouds have been made by Black and Smith (1980).

II. MEASUREMENT

The f-values were measured by determining, in a quantitative way, the absorption produced by a known column density of HCl. The procedure and apparatus were similar to those used by Smith and Parkinson (1978, here-after referred to as SP) who measured the f-values of the $\tilde{C}-\tilde{X}$ band of H₂O at 124 nm. As a consequence, only a review of the technique plus a discussion of significant differences will be given here.

The stainless steel absorption cell was 8.1 mm long and had two LiF windows. Airco electronic grade HCl, with a minimum purity of 99.99%, was used at five different pressures, 1.73, 2.13, 2.40, 2.67, and 3.20 Pa (i.e., 13 μ m Hg to 24 μ m Hg). These pressures, which were measured with a capacitance manometer, correspond to a range of column densities, $n_0 l$, from (3.46 ± 0.28) × 10⁺¹⁴ cm⁻² to (7.88 ± 0.41) × 10⁺¹⁴ cm⁻². The quantity n_0 is the molecule density in cm⁻³, and l is the cell length. The uncertainties given represent the combined effects of those in the cell length (± 0.08 mm), the pressure (± 0.13 Pa), and the gas purity. The latter was assessed by studying the absorption spectrum of significantly greater column densities of the gas (5×10^{16} – 5×10^{17} cm⁻²) in the wavelength range from 90 to 200 nm. No probable impurities were seen. The measurements were made at room temperature (295 ± 1 K).

The spectrograph, light source, and photographic plates were the same as those used by SP. The slit width was $15 \,\mu$ m. Exposure times ranged from 2 to 10 minutes. The density versus log (exposure) curve (i.e., the characteristic curve) of the photographic emulsion was determined in the same way as in the previous work. Typically, a set of six exposures, with different, metal-mesh neutral density filters, was obtained at each pressure, which was measured before and after each set of exposures was made. Absorption spectra at two pressures were obtained on each plate.

Microphotometer slits of $10 \,\mu\text{m} \times 300 \,\mu\text{m}$ were used in the measurement of the plate densities. The sampling interval was $10 \,\mu\text{m}$, i.e., the wavelength interval was 6.1×10^{-4} nm. Density data required for determination of the characteristic curve were measured at approximately 128.6 and 129.6 nm. The data were stored on magnetic tape.

III. DATA ANALYSIS AND RESULTS

a) Characteristic Curve

The transmission of the LiF windows decreased during the exposures because of the corrosive action of the HCl and the irradiation by the background continuum. This change was about 15-30% per hour. The characteristic curve was obtained by interpolating to find the density as a function of exposure (as determined by the neutral density filters) at a given time. A piece-wise linear approximation, with an estimated precision of $\pm 3\%$ in relative exposure over the linear portion of the characteristic curve, was used to convert the density data to relative intensity data.

b) Spectral Synthesis

An upper limit to the instrumental width was determined by studying the d(0) ${}^{3}\Pi_{1}-X(0)$ ${}^{1}\Sigma^{+}$ band of HCl at 122 nm (Tilford and Ginter 1971). For this work, both the microphotometer slit width and sampling interval were 5 μ m. The rotational lines of this band had a measured full width at half maximum (FWHM) of $(1.5 \pm 0.3) \times 10^{-3}$ nm $(15 \pm 3$ mÅ). The lines of the C-X band had a minimum FWHM of (8.5 $\pm 0.8) \times 10^{-3}$ nm (85 ± 8 mÅ). The large ratio of natural line width to instrumental width meant that the absorption profiles were essentially Lorentzian in character. Consequently, the intensity data were fitted to an equation of the form

$$I(\lambda) = I_0(\lambda) \exp\left[-\sum_i k_i(\lambda)l\right],$$
 (1)

where $k_i(\lambda)$ is the absorption coefficient for line *i* with wavelength λ_i and is given by

$$k_i(\lambda) = \frac{\alpha_i}{\left[(\lambda - \lambda_i)/\gamma_i\right]^2 + 1}$$
(2)

 $I_0(\lambda)$ was assumed to have the form

$$I_0(\lambda) = a\lambda + b . \tag{3}$$

The parameters *a* and *b* were not varied by the program, but were selected to give good fits to I_0 in the region of 128.7 nm and 129.5 nm. Spectra taken with no HCl in the absorption cell were used to ensure that neither the high *J* members of the *R* and *P* branches of the *C*-*X* system nor the $b^3\Pi_i - X^1\Sigma^+$ band of HCl at 128.4 nm caused errors in determining the background level.

In order to reduce the number of calculations, the data points were averaged in groups of three. Thus the fitted points were at intervals of 30 nm on the plate or 1.83×10^{-3} nm (18.3 mÅ). An iterative fitting procedure alternately varied α_i and γ_i in order to minimize the sum of the squares of the residuals. The individual line positions were not fitted by the program but were calculated from the wavenumber data of Tilford, Ginter, and Vanderslice (1970) and the known spectrograph dispersion.

The spectrum synthesis procedure required some interaction on the part of the investigators. For example, the wavelengths for individual lines were adjusted to give the optimum fit. Except for Q(8), the change in wavelength was less than 0.1 of the FWHM. An example of a fitted spectrum is shown in Figure 1.

c) Oscillator Strengths for the (0, 0) Band

The relation

$$f_{J'J''} = \frac{1}{0.0266n(J'')} \int_0^\infty k(v) dv , \qquad (4)$$

where $v \equiv c/\lambda$ (see Thorne 1974) was used to determine the *f*-values from the absorption profiles. The quantity n(J'') is the density of molecules in the absorbing level

$$n(J'') = n_0 \frac{2J'' + 1}{U} \exp\left(-E_{J''}/kT\right), \qquad (5)$$

where U is the partition function, J'' is the rotational quantum number of the lower level of the transition, $E_{J''}$ is the energy of that level, k is the Boltzmann constant, and T is the temperature in K. The value n_0 is given by

$$n_0 = L\left(\frac{P}{101325}\right)\left(\frac{273.16}{T}\right),$$
 (6)

where $L = 2.69 \times 10^{19} \text{ cm}^{-3}$ and P is the pressure in Pa (i.e., in N m⁻²).

Combining equations (2), (4), (5), and (6), and using U = 19.9 at T = 295 K, we get

$$f_{J'J''} \equiv f_i = (0.287 \text{ cm}^2 \text{ Pa}) \frac{\alpha_i \gamma_i}{\lambda_i^2 P} \frac{\exp\left(E_{J''}/kT\right)}{2J'' + 1}$$

To within the experimental uncertainties (see § IIId), the fitted values of γ_i (i.e., the line width) showed no dependence upon pressure and the values of α_i were proportional to pressure for the exposures with $P \leq$ 2.7 Pa. For the highest pressure used, 3.2 Pa, the values of α_i/P were systematically lower than average. We attributed this discrepancy to possible errors in determining the characteristic curve at the positions of low photographic density, i.e., at the absorption maxima of the lines. Consequently, the data from the highest pressure spectrum were discarded and the *f*-values were determined from unweighted averaged values of $\alpha_i \gamma_i/P$ for the remaining four exposures. Some systematic discrepancies in the data are discussed in § IIId.

Table 1 and Figure 2 show the results. The *f*-value and γ for R(6) are anomalously large and probably represent the sum of all *f*-values for this branch for lines with $J'' \ge 6$. The result for the R(0) line of the (1, 0) band is discussed in § III*e*.

The spectral synthesis was performed without consideration of the theoretical intensity variations

1980ApJ...238..874S



TABLE 1

Band	Line	Wavelength (nm) ^a	f	Uncertainty ^b	$\gamma (10^{-3} \text{ nm})^{c}$
(0, 0)	$R(6)^{d}$	128.9190	0.115°	+0.05/-0.08	8.2 ± 1.1
	R(5)	128.9271	0.049	+0.025	6.0 + 0.3
	R(4)	128.9388	0.056	± 0.019	5.3 ± 0.5
	R(3)	128.9550	0.065	+0.013	5.1 + 0.3
	R(2)	128.9745	0.074	± 0.015	5.9 ± 0.3
	R(1)	128.9983	0.097	± 0.019	4.8 ± 0.5
	R(0)	129.0257	0.185	± 0.037	4.4 ± 0.4
	Q(1)	129.0608	0.092	+0.031	5.0 ± 0.9
	$\hat{O}(2)$	129.0675	0.099	+0.033	5.3 + 0.5
	$\widetilde{O}(3)$	129.0786	0.098	± 0.033	5.4 ± 0.3
	$\widetilde{O}(4)$	129.0933	0.090	+0.018	5.2 + 0.2
	$\widetilde{O}(5)$	129.1117	0.085	+0.017	5.2 + 0.5
	$\tilde{P}(2)$	129.1314	0.018 ^f	+0.02/-0.01	5.5 ⁱ
	0(6)	129.1344	0.090^{f}	+0.03	5.5 ⁱ
	$\tilde{O}(7)^{g}$	129.1608	0.052°	+0.03	4.6 ± 1.4
	$\widetilde{P}(3)$	129.1718	0.035 ^e	+0.011	5.9 + 1.1
	$O(8)^{h}$	129.1891	0.043 ^e	+0.03	5.6 + 0.7
	$\widetilde{P}(4)$	129.2180	0.033	+0.010	5.2 + 0.4
	P(5)	129.2659	0.032	+0.010	5.7 + 0.6
	P(6)	129.3206	0.033	± 0.013	6.0 ± 1.0
(1, 0)	$R(0)^{j}$	124.7079	0.022	+0.02/-0.01	13.6

Oscillator Strengths and Widths for Lines of the (0, 0) and (1, 0) Bands of the $C^{1}\Pi - X^{1}\Sigma^{+}$ System of HCl from the Spectral Synthesis Fits to the Data

NOTES TO TABLE 1.—(a) Taken from wavenumber data of Tilford *et al.* (b) See text. (c) The uncertainty given is the standard deviation from the mean. (d) This includes R(6), R(7), R(8), etc. See text. (e) This value is not consistent with expected J" dependence (see Fig. 2). (f) Values for P(2) + Q(6) blend are taken from Fig. 2. (g) Obtained from 3 exposures only. (h) Obtained from 2 exposures only. Optimum fit obtained with $\lambda = 129.1937$ nm. (i) This value is estimated. (j) Obtained from one exposure only.



FIG. 2.—The *f*-values for the C(0) ${}^{1}\Pi - X(0)$ ${}^{1}\Sigma^{+}$ band of HCl as functions of the rotational quantum number J''. The squares, triangles, and circles refer to the *P*, *Q*, and *R* branches, respectively. The solid symbols represent the results of the spectral syntheses. Uncertainties are given in Table 1. The open symbols show results that have been adjusted to fit the expected J'' dependence shown by the dashed lines. The arrows show the extent of the adjustment from the spectral synthesis results for *P*(3), *Q*(7), *Q*(8), and *R*(6). Separate *f*-values for the blended lines *Q*(6) and *P*(2), obtained by extrapolation, are shown by the \times and +, respectively.

expected in molecular spectra. From Kovacs (1969) we see that

$$f_{J'J''} = \begin{cases} \kappa (J''-1)/(2J''+1) & \text{for } P \text{ branch }, \\ \kappa & \text{for } Q \text{ branch }, \\ \kappa (J''+2)/(2J''+1) & \text{for } R \text{ branch }, \end{cases}$$

where κ is a constant of proportionality.

The dotted curves in Figure 2 show these functions of J" with κ scaled to fit the data. Some of the blended and/or weak lines [e.g., P(3), Q(7), Q(8), R(6)] deviated from the expected behavior by more than the experimental uncertainties. Synthetic spectra were generated by using values of α for these lines that were adjusted to give f-values consistent with the expected J" dependence shown in Figure 2. R(7) was included and Q(6) and P(2) were calculated separately in these syntheses; average values of γ (viz., $\gamma = 5.5 \times 10^{-3}$ nm) were used and α was selected in the manner just described.

The resulting synthetic spectra did not give as good a fit to the data as did the original spectra, which were adjusted to the optimum fit by the computer. The Q(6) line fitted well, but the R(6), R(7), the P(2), and the P(3) portions were underestimated and the Q(7) and the Q(8) portions were overestimated. These discrepancies are what one would expect as a result of the use of the extrapolated f-values.

878

d) Uncertainties

The statistical uncertainty in the results for the seven best-resolved lines [R(0), R(1), R(2), R(3), Q(4), Q(5), and Q(6) + P(2)] was typically $\pm 10\%$. The results were not distributed at random; the values of γ_i obtained at P = 2.13 Pa were systematically 10%smaller than the mean of the other data while the values of α_i/P for P = 1.73 and 2.13 Pa were 8%smaller, on the average, than those for P = 2.40 and 2.67 Pa.

There are several possible causes of these deviations. The uncertainty of the pressure determinations is about 6%. The position of the background continuum, I_0 , could be determined to within 1%. Spectral fits with backgrounds that were deliberately misplaced by this amount, gave *f*-values for the stronger lines that changed by 2% from the previously determined values. Weaker lines varied by more. The slope of the linear portion of the characteristic curve could be determined with an uncertainty of about $\pm 5\%$. A spectral fit with a curve changed by this amount gave *f*-values that were changed by 6-12%, depending on the line strength. These three effects could produce systematic deviations of the magnitude discussed in the previous paragraph.

The effects of additional possible systematic errors, which would affect all the results, were estimated. The most likely of these involved the nonlinear portions of the characteristic curve which were difficult to determine precisely. The effects of uncertainties in gas purity and temperature were neglected.

The uncertainties given in Table 1 are estimated standard deviations; those for the best-resolved lines are $\pm 20\%$. For the other lines, the quoted uncertainties are more subjective. This situation reflects the facts that these lines are weak and/or blended, that they deviate from the expected dependence upon J'', and that the quality of the computer fits for these lines was not sensitive to large changes in α or γ .

e) The R(0) Line of the (1, 0) Band

The R(0) line of the (1, 0) band, at 124.7079 nm, is significantly weaker than the R(0) line of the (0, 0)band and, therefore, is unlikely to be useful for astronomical observations. However, because Wright and Morton (1979) looked for absorption in interstellar clouds by both lines, a measurement of the fvalue of the R(0) line of the (1, 0) band was made. This determination was made with somewhat less precision than was used for the (0, 0) band measurements. Only one spectrum, of absorption by a column of 1.5×10^{16} cm⁻² of HCl, was analyzed and the lines extended beyond the linear portion of the characteristic curve. The f-value is 0.022. The estimated uncertainty is a factor of 2. The value of γ is 0.014 \pm 0.005 nm.

f) Band Strengths

Following the suggestion of Tatum (1967) we calculated the band strengths from our oscillator strength

data; using his notation

$$=\frac{2.366\times10^{-61}\lambda\varpi_1f\times(2-\delta_{0,\Lambda''})(2S+1)(2J''+1)}{\text{H\"onl-London factor}}$$

where the band strength is in units of coulombs² m², λ is in Å, and the normalization of the Hönl-London factors is such that the sum of them for all those transitions that have a common lower level is 2J'' + 1. In our case, $\varpi_1 = 1$, and $\Lambda'' = 0$.

The 14 *f*-values data for the (0, 0) band denoted by solid points in Figure 2 gave $S[C(0), X(0)] = 5.54 \times 10^{-59}$ coulombs² m² with a standard deviation of 0.39×10^{-59} coulombs² m². The uncertainty in S is estimated to be $\pm 20\%$.

For the (1, 0) band, $S[C(1), X(0)] = 6.5 \times 10^{-60}$ coulombs² m², with an uncertainty of a factor of 2.

IV. HCl in diffuse interstellar clouds

Interstellar absorption lines of HCl have been searched for but not detected toward 10 stars by Jura and York (1978) and toward ζ Oph by Morton (1975) and Wright and Morton (1979). The chemistry of interstellar chlorine has been discussed by Jura (1974) and by Dalgarno *et al.* (1974). Jura (1974) proposed that Cl can be ionized by photons at wavelengths between 91.2 and 95.6 nm, that the resulting Cl⁺ reacts rapidly with H₂ to form HCl⁺, and that this reacts with H₂ to form H₂Cl⁺, the immediate precursor of HCl. Consequently, one might expect HCl to abound in regions containing both H₂ and chlorine-ionizing photons.

The most severe test of the proposed sequence of chemical reactions is provided by the observations of chlorine species toward ζ Oph. Use of the lower limit to our measured *f*-value of the *R*(0) line of the (0, 0) band, f > 0.138, with the upper limit to the equivalent width, W < 1.9 mÅ (Wright and Morton 1979), gives an upper limit to the column density: *N*(HCl) < 9.3 $\times 10^{11}$ cm⁻². This limit is a factor of 28 smaller than the column density predicted by the model of the ζ Oph cloud of Black and Dalgarno (1977). The column densities of Cl, 1.1×10^{14} cm⁻², and of Cl⁺, (1.2-4.3) $\times 10^{13}$ cm⁻², have also been measured (Morton 1975). The model (Black and Dalgarno 1977) was designed to match the observed amount of Cl, but it fails to reproduce the column density of Cl⁺; the calculated value (5.4×10^{12} cm⁻²) being a factor of about 4 too small.

Jura and York (1978) predict a ratio of column densities $N(\text{HCl})/N(\text{Cl}) \approx 0.01$ in typical diffuse clouds with high concentrations of H₂. This ratio is smaller than any of the limits established by Jura and York (1978) in their *Copernicus* observations of interstellar chlorine. This ratio is also smaller than that predicted by Black and Dalgarno (1977) for the ζ Oph cloud because Jura and York (1978) adopted a value for the rate of photoionization of Cl which is much smaller than that used by Black and Dalgarno (1977). A recent survey of interstellar absorption lines with the ultraviolet spectrograph of the IUE (Black et al. 1980; Black 1980) has extended observations of Cl and HCl to more highly-reddened background stars than were accessible to Copernicus. Details of the observations will be published elsewhere, but the data on the diffuse cloud toward HD 46223 suggest already that it provides another severe test of the chlorine chemistry. The equivalent width of the 129.0257 nm line of HCl is W \leq 7.4 mÅ, implying a column density N(HCl) \leq 2.7 $\times 10^{12}$ cm⁻². The equivalent width of the 134.7420 nm line of Cl I of $W = 82.5 \pm 5$ mÅ indicates a column density, $N(\text{Cl}) \approx 1.5 \times 10^{15} \text{ cm}^{-2}$. This is derived from a preliminary curve-of-growth analysis of a number of interstellar lines of neutral atomic species. The resulting small ratio, $N(\text{HCl})/N(\text{Cl}) \leq 0.0018$, is in conflict with the expectations of Jura and York (1978).

The above evidence suggests that the previously adopted chlorine chemistry is not correct in detail. The modifications required to bring the predictions of the ζ Oph model into agreement with the observations must have the effect of increasing the relative abundance of Cl⁺ at the expense of the HCl. A recent, improved calculation of the cross section for photoionization of Cl indicates a value near threshold of $\sigma \approx 1.8 \times 10^{-17}$ cm² (Brown, Carter, and Kelly 1978), which implies that the photoionization rate should be 3 times smaller than that adopted by Black and Dalgarno (1977). This only exacerbates the difficulty of accounting for the large ratio $N(\text{Cl}^+)/N(\text{HCl})$, toward ζ Oph. We have recomputed the rate coefficient of radiative recombination

$$Cl^+ + e \rightarrow Cl + hv$$
, (7)

using the new photoionization cross section (Brown, Carter, and Kelly 1978) and following the prescription of Gould (1978). At low temperatures ($T \lesssim 100$ K) the rate coefficient is $\alpha = 1.0 \times 10^{-10} T^{-0.56}$ cm³ s⁻¹. The reaction

$$Cl^+ + H_2 \rightarrow HCl^+ + H$$
, (8)

not only leads to the formation of HCl, but also dominates the removal of Cl⁺ in regions containing H₂. Although this reaction is known to have a rate coefficient of $k = 7 \times 10^{-10}$ cm³ s⁻¹ at T = 298 K (Fehsenfeld and Ferguson 1974), it is exothermic by only a small amount, 0.22 ± 0.02 eV, and the existence of a modest energy barrier cannot be excluded. The presence of an energy barrier would reduce the rate coefficient at the temperatures, $T = 20{-}100$ K, expected in diffuse interstellar clouds with the result that the predicted concentration of Cl⁺ would increase, while that of HCl would decrease. It is possible that the subsequent reaction

$$HCl^{+} + H_{2} \rightarrow H_{2}Cl^{+} + H$$
 (9)

(Fehsenfeld and Ferguson 1974) is also only slightly exothermic, and that it has a small energy barrier that decreases its rate at low temperature. If reaction (9) is very slow at interstellar temperatures, then the concentration of HCl⁺ will be enhanced and that of HCl will be reduced while the concentration of Cl⁺ will be largely unaffected. An experimental investigation of the temperature dependences of reactions (8) and (9) is therefore crucial for a complete understanding of the chemistry of interstellar chlorine.

Because the rates of reactions (8) and (9) at low temperatures are not known, it is possible to choose values for them that bring the predictions of the model of the ζ Oph cloud into harmony with the observations for Cl, Cl⁺, and HCl. Good agreement with the observations can be achieved when these rate coefficients behave approximately as $k \approx$ $10^{-9} \exp(-\Delta E/kT) \text{ cm}^3 \text{ s}^{-1}$, with energy barriers in range given by $\Delta E/k \approx 100-200 \text{ K}$.

It is worthwhile to reiterate Jura's (1974) comment, viz. that a sensitive astronomical search for lines of the $A^{2}\Sigma^{+}-X^{2}\Pi$ system of HCl⁺ is feasible. Franck-Condon factors and accurate spectroscopic constants for this system in the near ultraviolet have been published recently (Saenger, Zare, and Mathews 1976), and recent lifetime measurements (Möhlmann, Bhutani, and de Heer 1977) indicate that the oscillator strengths of the lines arising in the lowest rotational level of the (0, 0), (1, 0), and (2, 0) bands will all be $f \approx 1.5 \times 10^{-3}$. Although HCl⁺ is likely to be too rare to be detected easily in diffuse clouds, limits upon its abundance might help to constrain the theory further.

During the course of these measurements, the authors obtained high resolution laboratory spectra in the wavelength range from 100-140 nm for a wide range of measured column densities of HCl. Some band assignments in this wavelength region have been made (Tilford, Ginter, and Vanderslice 1970; Tilford and Ginter 1971; Douglas and Greening 1979), and M. L. Ginter is extending the analysis and preparing an atlas of the absorption spectrum. Additional *f*-values, with uncertainties of the order of a factor of 1.5-2 could be obtained from our data if there is a specific scientific need.

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