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# Quenching of NO( $D^2\Sigma^+$ , $E^2\Sigma^+$ ) formed by UV photodissociation of NOCI

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The photodissociation of NOCl at 193 nm has resulted in vibrationally excited NO which was subsequently excited to the D(v = 5) and E(v = 0) states. The resulting fluorescence at 160–180 nm has been analyzed in terms of a kinetic model. Saturation of the photodissociation was observed at laser intensities above 2 MW/cm<sup>2</sup>. The quenching rate constants of NO<sup>\*</sup> by Ne, Ar, Kr, and Xe were determined to be  $k_{\text{Ne}} = (6.4 \pm 1.6) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ ,  $k_{\text{Ar}} = (1.7 \pm 0.4) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ ,  $k_{\text{Kr}} = (6.6 \pm 1.7) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ , and  $k_{\text{Xe}} = (1.0 \pm 0.2) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ . The quenching by NO and Cl was explained in terms of charge transfer mechanism and the corresponding rate constants were found to be:  $k_{\text{NO}} = (2.7 \pm 0.6) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  and  $k_{\text{Cl}} = (2.1 \pm 0.5) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ .

#### I. INTRODUCTION

The NOCl molecule has a strong absorption continuum in the ultraviolet, <sup>1,2</sup> with a peak around 200 nm. Previous studies<sup>3-5</sup> of photolysis of NOCl at wavelengths ranging from 480 to 200 nm showed that the products of the photolysis are NO and Cl, and that the resulting NO possesses a degree of vibrational excitation which is strongly dependent on wavelength; namely, at  $\lambda < 250$ nm, it is excited at least to the v = 11 level. Two more recent studies<sup>6,7</sup> have concentrated on the mechanism of photodissociation of NOCl at 248 and 193 nm, using excimer lasers; at 193 nm, vibrational excitation at least as high as v = 16 was detected with a peak at v = 14. In the present studies, we report the dissociation of NOCl by an ArF\* laser (193 nm), which is followed by further absorption of a 193 nm photon by the nascent NO molecule. Employing only moderate laser intensities, dissociation of all the NOCl molecules in the interaction volume at densities of 10<sup>16</sup>-10<sup>17</sup> molecules/cm<sup>3</sup> was observed.

The vacuum ultraviolet (VUV) fluorescence at 160– 180 nm from the highly excited NO molecules was analyzed in terms of a kinetic model. The quenching rate constants of NO\* by rare gases were determined, and the quenching mechanism by NO and Cl was discussed in terms of a curve crossing involving ionic and covalent states. The possibility of an optically pumped VUV laser was investigated.

#### **II. EXPERIMENTAL APPARATUS**

The experiments were performed with a Lambda Physics model EMG 101 excimer laser in an unstable resonator configuration. The pulses typically were 10 ns FWHM with ~50 mJ per pulse. The laser was focused into a static fill stainless steel gas cell by a  $CaF_2$  (f=50 cm) lens placed 29 cm from the interaction region (Fig. 1). In this region, the laser beam had a rectangular spatial profile 1.0×0.2 cm, with the long dimension parallel to the entrance slit of the monochromator. Because of the large attenuation of the 193 nm laser radiation by NOCl at pressures of 0.5–10 Torr, the laser entrance window was at a distance of only 3 mm from the volume viewed by the monochromator. The observed fluorescence, whose spectrum was recorded by an optical multichannel analyzer (PAR model 1205), was collected perpendicular to the laser beam by a LiF lens and imaged onto the slit of a 0.3 m evacuable monochromator (McPherson model 218). In an alternative configuration, the temporal behavior of the fluorescence was studied with a solar blind photomultiplier (EMI model G26H3152). The gases used were; NOC1 (97% pure, Matheson), Ne (99. 999% pure, Liquid Carbonic), Ar (99. 995% pure, Spectra Gases, Inc.), Kr (99. 995% pure, Air Products), and Xe (99. 995% pure, Spectra Gases, Inc.). In all experiments, these gases were mixed in the sample cell with a stainless steel/nylon magnetic stirrer and the gas pressures were measured with MKS Baratron gauges.

#### III. RESULTS AND DISCUSSION

#### A. Pure NOCI

Structured fluorescence in the spectral region 160-180 nm was observed upon irradiation of NOCl at 193 nm (Fig. 2) in the pressure range 0.5-10 Torr. Similar fluorescence had been observed following photodissociation of N<sub>2</sub>O at 193 nm, with additional lines in the range



FIG. 1. Schematic diagram of the experimental setup in the photodissociation studies of NOC1.

180-230 nm.<sup>8</sup> These additional lines in the longer wavelength region were not seen in the present study because of absorption by NOCl, which has a large cross section<sup>2</sup> in the 180-230 nm interval (see Fig. 2). The fluorescing lines were identified<sup>8</sup> as corresponding to the following transitions<sup>9</sup> in NO:  $\epsilon$  (5, 1),  $\epsilon$  (5, 2),  $\gamma'$  (0, 1),  $\gamma'$  (0, 2), and  $\epsilon(5, 4)$ .

The production of NO\* is the result of a stepwise twophoton absorption. The observed time-integrated fluorescence arises from the final stage in the following model:

photodissociation	$\text{NOCl} + h\nu_1 \rightarrow \text{NO}(X, v'') + \text{Cl}$ ,	$K_1 = \sigma_1 I_1$ ,	(1)
photoexcitation	$NO(X, v'') + h\nu_i \rightarrow NO^*$ ,	$K_2 = \sigma_2 I_1 ,$	(2)

radiative decay  $NO^* \rightarrow NO(X, v'') + h\nu_f$ ,  $K_r$ , (3)

quenching 
$$NO^* + M \rightarrow NO + M$$
,  $K_A = k_A[M]$ , (4)

of pholaser NO and Cl. Tuus,  $K_4 = k_N c [NO] + k_{Cl} [Cl]$  in this circumstance in the limit of complete molecular dissociation.

The photodissociation of NOCl at 193 nm [Eq. (1)]proceeds efficiently with a cross section<sup>2</sup>  $\sigma_1 \approx 5.6 \times 10^{-17}$ cm<sup>2</sup>. The subsequent photoexcitation of the nascent NO (X, v'') depends on accidental coincidences between the laser wavelength and allowed transitions in NO. The laser output has a spectral width<sup>10</sup> of ~100 cm<sup>-1</sup>, centered at 193.3 nm. Within this range lie the D + X(5,7) and E - X (0,5) lines of NO [ $\epsilon$  (5,7),  $\gamma'$  (0,5), respectively]. The cross section for the  $\epsilon(5,7)$  transition was estimated on the basis of the known oscillator strengths of other  $\epsilon$  lines, <sup>11</sup> the respective Franck-Condon factors, <sup>12</sup> and the ratio of laser bandwidth to Doppler width  $\sigma_{2,\epsilon} \approx 1 \times 10^{-17} \text{ cm}^2$ . Similarly, from the Franck-Condon factors and the ratio of electronic matrix elements of the X - D and X - E transitions<sup>8</sup>  $\sigma_{2,x}$  $\approx 1.2 \times 10^{-18}$  cm<sup>2</sup>. From Fig. 2, it is evident that under our experimental conditions, the individual lines partially overlap, and therefore, they cannot be treated separately. However, from the amplitudes of the peaks, a rough estimate was obtained for the ratio of populations of the nascent NO in the v = 5 and v = 7 states:  $r \equiv N_{\nu=5}/N_{\nu=7} = 2 \pm 1$ . This figure cannot directly be compared with the time-of-flight measurements<sup>7</sup> of vibrational distribution in NO, because v = 5 and v = 7 have negligibly small populations<sup>7</sup> compared to the peak (v = 14). From the above estimates the weighted average of the cross section for photoexcitation [Eq. (2)] is

$$\sigma_2 = \frac{\sigma_{2.6} + \gamma \sigma_{2.7}}{1 + \gamma} \approx (4.1 \pm 1.5) \times 10^{-18} \text{ cm}^2 .$$
 (6)

The radiative rate of NO\* [Eq. (3)] was estimated to be  $K_r = 1 \times 10^8 \text{ s}^{-1}$ . This value takes into account the decrease in radiative lifetime<sup>13,14</sup> of the vibrational levels of the  $D^2\Sigma^*$  state, from 16 ns at v = 0 to 10 ns at v = 3. The  $E^2\Sigma^*$  state has a longer lifetime, because of a smaller value of the E-X matrix element.<sup>8</sup> Thus, a weighted average of D(v'=5) and E(v'=0) was estimated to yield  $K_r \approx 1 \times 10^8 \text{ s}^{-1}$ . Time-dependent studies of the fluorescence showed that its duration was that of the laser pulse. This fact determined a lower limit for  $K_3 = K_r + k_4 [\text{NOC1}], K_3 \gtrsim 10^8 \text{ s}^{-1}$ , in agreement with our estimates.

In Eqs. (1)-(5),  $I_i$  is the laser intensity in units of photons cm<sup>-2</sup>s<sup>-1</sup>,  $\nu_i$  and  $\nu_f$  are the frequencies of the laser and of the fluorescence, respectively, the k factors are the appropriate rate constants, the  $K_i$  represent the total rates, the  $\sigma_i$  are the absorption cross sections, and [M] is the concentration of the quenching species M. For experiments in which no foreign gas is added, M is primarily the sum of NO and Cl, and  $k_4$  denotes the average rate constant for quenching of NO\* by both

 $I_f = K_r \int_0^\infty [\operatorname{NO}^*(t)] dt \; .$ 

FIG. 2. Lower curve (right-hand ordinate): The fluorescence spectrum from NO\*. NOCl pressure was p=1 Torr, the laser intensity was  $I_1 = 3$  MW/cm<sup>2</sup>. Upper curve (left-hand ordinate): The absorption cross section of NOCl (from Ref. 2), showing the transmission "window" for the observation of the NO\* side fluorescence.





FIG. 3. The integrated NO\* fluorescence as a function of the laser intensity. The NOCl pressure was 1 Torr. The solid line is the best fit, using the exact analytical expression (6) (see the text).

The analytical solution of the system of rate equations representing Eqs. (1)-(5) above is

$$I_{f} = K_{\tau} \delta \{ [(K_{3} - K_{1})^{-1} (K_{3}^{-1}A_{3} - K_{1}^{-1}A_{1}) - (K_{3} - K_{2})^{-1} (K_{3}^{-1}A_{3} - K_{2}^{-1}A_{2})] + K_{3}^{-1} \exp(-K_{3}t_{p}) \times [(K_{3} - K_{1})^{-1} (A_{1} - A_{3}) - (K_{3} - K_{2})^{-1} (A_{2} - A_{3})] \}.$$
(6)

In Eq. (6),

$$K_{3} \equiv K_{r} + k_{4} [\text{NOC1}] ,$$

$$A_{i} \equiv \exp(-K_{i}t_{p}) - 1 \quad (i = 1, 2, 3) ,$$

$$\delta \equiv \alpha_{v}, \frac{\sigma_{1}\sigma_{2}}{\sigma_{2} - \sigma_{1}} I_{i} [\text{NOC1}] ,$$
(7)

in which  $t_p$  is the laser pulse duration  $(t_p = 10^{-8} \text{ s})$ , and  $\alpha_{v'}$ , is the fraction of NO molecules in the v'' = 5, 7 vibrational states of the ground electronic state.

Expression (6) can be used to analyze the dependence of the integrated fluorescence  $I_f$  on the laser intensity  $I_i$ . At low intensities, when

$$K_i \ll 1/t_p$$
,  $K_3(i=1,2)$ , (8)

only a small fraction of the NOCl molecules is dissociated, and of those dissociated, only a small fraction is further excited. In this case,  $I_f$  varies as the square of the laser intensity. Conversely, in the high-intensity limit,

$$K_i \gg 1/t_p$$
,  $K_3$   $(i=1,2)$ , (9)

both photodissociation and photoexcitation are saturated, and  $I_f$  is independent of  $I_I$ . In the intermediate regime, the process with the larger cross section only is saturated, and  $I_f$  is linear in laser intensity. In our experiment, the photoexcitation has a smaller cross section than the photodissociation  $\sigma_2 < \sigma_1$  because of the large laser linewidth. The use of a narrow linewidth laser, of course, would allow this condition to be reversed. Figure 3 depicts the intensity variation of the integrated fluorescence at 1 Torr NOC1. The solid line is the best fit with  $\sigma_1 = 5.6 \times 10^{-17}$  cm<sup>2</sup> and  $\sigma_2 = 4.1 \times 10^{-18}$  cm<sup>2</sup>. Also, we used  $k_4 = 4.8 \times 10^{-9}$  cm<sup>3</sup> s<sup>-1</sup>, which is the value we obtained in our quenching measurements (see the following section). The only variable parameter was the vertical scale factor, which is just proportional to the collection efficiency of the detection system. As can be seen from Fig. 3, at laser intensities  $I_1 \gtrsim 1.5$  MW/cm<sup>2</sup>, all the NOC1 molecules in the interaction region were dissociated. Extrapolation of the line in Fig. 3 (not shown in the figure) predicts saturation of the photoexcitation as well at intensities  $I_1 \gtrsim 50$  MW/cm<sup>2</sup>.

#### B. Collisional effects of inert gases

The inert gases, Ne, Ar, Kr, and Xe were added to NOCl to determine the quenching rate coefficients of NO<sup>\*</sup> in the D and E states. At the laser intensities used in our experiment, the inert gases did not interact with the laser, and they were also transparent at the fluorescence wavelength. The only effect was interaction with NO<sup>\*</sup> causing quenching of the fluorescence, probably by inducing D,  $E \rightarrow C$  transitions<sup>15</sup> in NO. At NOCl, pressures sufficiently high that  $K_1$ ,  $K_2 \ll K_3$ , expression (6) for  $I_f$  is reduced to the familiar Stern-Volmer equation

$$I_{f}(P_{Rg}) = I_{f}(P_{Rg} = 0) \frac{K_{\tau} + k_{4}[\text{NOC1}]}{K_{\tau} + k_{4}[\text{NOC1}] + k_{Rg}[Rg]} , \quad (10)$$

where  $k_4 = k_{NO} + k_{C1}$  from Eq. (4). For the circumstance  $I_1 \approx 2$  MW/cm<sup>2</sup>, the above condition was satisfied at pressures higher than approximately 1.5 Torr. The  $\epsilon(5, 2)$  and  $\gamma'(0, 1)$  lines were integrated together and displayed in Stern-Volmer plots. The slopes *m* of the Stern-Volmer plots are, from Eq. (10),

$$m = \frac{k_{\text{Rg}}}{K_r + k_4 [\text{NOC1}]} . \tag{11}$$

The values of *m* varied significantly as the NOCl pressure was changed. In Fig. 4, the inverse slopes  $m^{-1}$  are plotted vs NOCl pressure for Ar and Kr. The slopes of these plots (Fig. 4) are  $k_4/k_{\rm Rg}$  and the intercepts are  $K_r/k_{\rm Rg}$  from Eq. (11). For  $K_r = 1 \times 10^8 \, {\rm s}^{-1}$ , the quenching rates coefficients of the rare gases were determined and the values are listed in Table I. As shown in Table I, the quenching coefficient increases with increasing atomic size of the rare gas.

The quenching rate coefficient  $k_4$ , which is the sum of the rate coefficients for quenching by both NO(X, v) and Cl, was found from the same data to be  $k_4 = (4.8 \pm 1.0) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ . Previous values of  $k_{\text{NO}}$  are in the range<sup>13,15-17</sup> (0.3-3.5)×10<sup>-9</sup> cm<sup>3</sup> s<sup>-1</sup>. Comparison of the value of  $k_4$  found in this study with the other published values suggests that the quenching rate constant by Cl atoms of NO\* is on the order of  $k_{\text{Cl}} \sim 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ .

The quenching mechanism of NO\*(D, E) by Cl atoms can be viewed in terms of a charge transfer<sup>16,18,19</sup> process in the following way. The covalent potential curve correlating to NO\*(D, E) + Cl crosses the ionic curve which correlates to NO\*( $a^{3}\Sigma^{*}$ ) + Cl<sup>-</sup> at  $R_{x} \approx 5.1$  Å. The same ionic curve then crosses the covalent curve which corresponds to NO\*( $C^{2}\pi$ ) + Cl at  $R'_{x} \approx 3.8$  Å. Thus, the

State(S)	D(v = 5) + E(v = 0)	E(v=0) + E(v=1) $+ E(v=2)$ $Bof 22$	D(v = 1) + D(v = 5) $+ E(v = 0)$	D(v=0)
Gas		Kel. 23	Nel. 8	Nel. 15
Neon	$6.4 \pm 1.6$	0.75		
Argon	$16.6 \pm 4.2$	7.5	$16.6 \pm 3.6$	15
Krypton	$66 \pm 17$			
Xenon	$100 \pm 25$			

TABLE I. Quenching rate coefficients of NO\* by rare gases (in units of 10<sup>-11</sup> cm<sup>3</sup> s<sup>-1</sup>).<sup>a</sup>

<sup>a</sup>The quenching rate constants of NO\* by rare gases. The data from Ref. 23 were calculated for a radiative lifetime 40 ns for the E state. Those from Ref. 8 were corrected to reflect the radiative lifetime of ~10 ns (Refs. 13 and 14) rather than 25 ns.

entrance channel NO(D, E) and the exit channel NO(C) are connected by means of an ionic curve and two curve crossings. The  $C^2\pi$  state predissociates into N(<sup>4</sup>S) and O(<sup>3</sup>P). In the Landau-Zener formulation of inelastic transitions in collisions, the cross section  $\sigma$  is related to  $R_x$  by<sup>20</sup>

$$\sigma = \pi R_x^2 e^{-\alpha R} x , \qquad (12)$$

in which  $0.0 \le \alpha \le 0.1$  for NO(*D*).<sup>16</sup> The curve crossing corresponding to Cl as the collision partner is at  $R_{x,Cl} \approx 5.1$  Å, a value which can be compared to the cross-ing point corresponding to NO collisions, <sup>16</sup>  $R_{x,NO} = 5.5$  Å. Using Eq. (12) and taking into account the effect of the reduced mass on the rate constant, we have estimated



FIG. 4. The inverse of the Stern-Volmer quenching slopes as a function of NOCl pressure, for Kr and Ar. The straight lines are the least square fits, based on Eq. (11) in the text. Similar lines were obtained for Ne and Xe. The quenching rate constants were determined from these lines.

$$k_{\rm C1}/k_{\rm NO} = 0.96(\sigma_{\rm C1}/\sigma_{\rm NO}) \approx 0.80$$
 (13)

Using Eq. (13) and our result  $k_4 \equiv k_{NO} + k_{C1} = (4.8 \pm 1.0) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ , we have deduced the individual rate constants for quenching NO\*;  $k_{NO} = (2.7 \pm 0.6) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ ,  $k_{C1} = (2.1 \pm 0.5) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ , values consistent with the measured quenching rates of NO. <sup>13,15-17</sup>

#### **IV. CONCLUSIONS**

In conclusion, an ArF\* laser at 193 nm was used to photodissociate NOCl molecules into NO(X, v) and ground state Cl atoms. Subsequent excitation of the photofragments NO(X, v) generated NO in the D(v=5)and E(v=0) states, which then produced fluorescence in the 160–180 nm range. A kinetic model was used to analyze the intensity dependence of the pure NOCl and the quenching rates of NO, Cl, and buffer gases.

It should be noted, that with laser linewidths of  $\leq 5$ cm<sup>-1</sup>, the photoexcitation stage can be saturated even with modest intensities of a few  $MW/cm^2$ . If the rotational-vibrational population distribution in NO, following photodissociation, is favorable, sizable population inversions can be obtained between the D, E states and certain rovibrational levels of the X state. Indeed, stimulated emission in the infrared has been observed following dissociation of NOCl in the near UV. 21,22 Stimulated emission in the infrared takes place on several rovibrational transitions in NO, involving v = 9. The recent results<sup>7</sup> on the vibrational population distribution of NO indicate that population inversion may occur at least below v = 14. Correspondingly, the mechanisms studied in this work indicate that it may be possible to generate a two-photon optically pumped laser in NO, at 160-170 nm.

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