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Vibrational level dependence of lifetime of NO_2 in the $\tilde{D}^2B_2$ state

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Abstract

The vibrational level dependence of the lifetime of NO_2 $\tilde{D}^2B_2$ has been investigated by using LIF, hole-burning, and photofragment excitation methods. The predissociation rates for the vibrational levels estimated from the line profiles increase sharply with excitation energy even blow the second dissociation limit to form NO and $O(^1D)$. The occurrence of the dissociation pathway into NO and $O(^1D)$ via the $\tilde{D}^2B_2$ states is confirmed by the observation of the appearance thresholds to form various J''-levels of $NO(^2\Pi_{1/2}, v'' = 0)$, which accord with the calculated dissociation limits. The dissociation mechanism is discussed based on the energy dependence of predissociation rates and $O(^1D)$ quantum yields.

1. Introduction

Nitrogen dioxide (NO_2) is one of fundamental triatomic molecules and an important component in the atmospheric and combustion chemistry. The visible absorption spectrum of NO_2 has long been the subject of intense investigation, because it shows extremely complex structure due to the strong vibronic coupling between the $\tilde{X}^2A_1$ and $\tilde{A}^2B_2$ states and the photodissociation in the visible region near the first dissociation limit to form $NO(^2\Pi_{1/2})$ and $O(^3P)$ is regarded as the bench mark system whether photodissociation occurs in the statistical nature.

The ultraviolet absorption system of NO_2 starting from 249.1 nm shows a much simpler vibronic structure than the visible one. The rotational structure of this ultraviolet absorption system was partially analyzed by Ritchie et al. [1] for the first time. They reported that the upper state has $^2B_2$ electronic symmetry. The upper state labeled $\tilde{D}$ has been characterized by Hallin and Merer [2] from the high-resolution absorption, and by our group [3,4] from the dispersed fluorescence. Detailed rotational analysis of the 249.1 nm band was reported, and the lifetime was also calculated

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from the linewidth of the $\tilde{D}^2B_2(0,0,0)$–$\tilde{X}^2A_1(0,0,0)$ transition to be 42 ps [2]. The degree of diffuseness of the band increases rapidly with increasing energy. For example, the linewidth of the (0,0,0) band at 249.1 nm is 0.127 cm^{-1}, while the rotational structure of the (0,0,2) band at 244.7 nm was not resolved because of diffuseness. It is interesting to note that the origin of the $\tilde{D}^2B_2$ state ($T_0 = 40\,125.849$ cm^{-1}), is located at energy far above the first dissociation limit (25 128.57 cm^{-1}) [5] and located at energy only 871 cm^{-1} below the second dissociation limit (40 996.43 cm^{-1}) to form NO($X^2\Pi$) and O(^1D).

The photodissociation of NO_2 in the $\tilde{D}^2B_2$ state has been investigated by several researchers [6–15]. Two groups studies the product state distribution of the nascent NO for the photodissociation of NO_2 at 248.5 nm corresponding to 40 241 cm^{-1} which is slightly lower than the second dissociation limit [7,10]. Shafer et al. [11] reported that O(^1D) fragment had a translational energy distribution peaking at low energy in the photodissociation of NO_2 at 205.47 nm. The lifetimes of the vibrationally excited levels in the $\tilde{D}^2B_2$ state of thermally averaged NO_2 were determined by spontaneous resonance Raman scattering polarization measurements [12]. More recently, strong orbital alignment has observed for the fragment O(^3P_J) in the photodissociation of NO_2 at 212.8 nm using ion imaging [13]. Evidently, we have only limited information on the vibrationally selective photodissociation of NO_2 in the $\tilde{D}^2B_2$ state, which includes the excited state lifetimes, the direct observation of O(^1D) product, and so on. In the present study, we spectroscopically investigated the vibrational level dependence of lifetime of NO_2 in the $\tilde{D}^2B_2$ state in combination with a supersonic jet technique.

2. Experimental

We employed three spectroscopic techniques to measure the absorption band profiles due to the $\tilde{D}^2B_2(v_1', v_2', v_3')$–$\tilde{X}^2A_1(0,0,0)$ transitions of jet-cooled NO_2, namely LIF, spectral hole-burning and photofragment excitation (PHOFEX) methods. A 1% NO_2/Ar premixed gas, kept at a backing pressure of 1 atm, was expanded through a pulsed nozzle into the vacuum chamber. In hole-burning experiments, the probe frequency was fixed to a single rotational line in one of the vibronic bands of the $\tilde{A}^2B_2$–$\tilde{X}^2A_1$ transition at 593 nm and the fluorescence light from the $\tilde{A}^2B_2$ state was detected, because the 593 nm band is relatively free from perturbation due to the strong vibronic coupling between the $\tilde{A}^2B_2$ and $\tilde{X}^2A_1$ states, and then shows a simple rotational structure [16]. In PHOFEX experiment, photofragment NO was probed by monitoring the LIF signal of the $A^2\Sigma$–$X^2\Pi$ transition of NO induced by another probe laser.

3. Results and discussion

Fig. 1 shows an LIF spectrum of jet-cooled NO_2 for the $\tilde{D}^2B_2(0,0,0)$–$\tilde{X}^2A_1(0,0,0)$ transition, called (0,0,0) band, together with the rotational assignment. Strong $^qP_0$ and $^qR_0$ branches and weak $^qP_1$ and $^qR_1$ branches were observed. Because of the nuclear spin statistics, ee and oo rotational levels are allowed to exist in the ground state of NO_2. All transitions observed in the LIF spectrum are assigned to a-type rotational transitions, which accords well with the results reported by Hallin and Merer [2]. Lineshapes of the peaks observed in the LIF spectrum are apparently broader than the linewidth of the laser used (0.25 cm^{-1}) due to the predissociation in the upper state. The $^qR_0$ branch is free from the overlap of peaks,

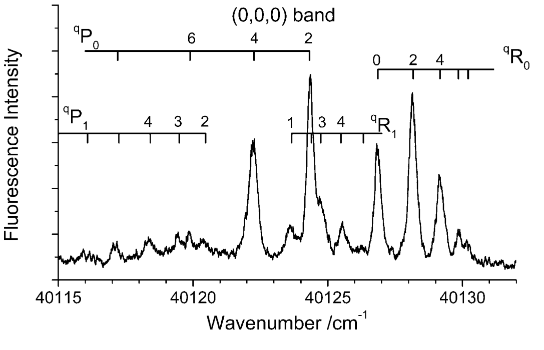

Fig. 1. LIF spectrum of the (0,0,0) band of the $\tilde{D}^2B_2$–$\tilde{X}^2A_1$ transition of jet-cooled NO_2.

and we estimate the Lorentzian width from the curve fitting operation using the Voigt profile, which is a convolution of Gaussian and Lorentzian profiles, as a fitting function. We assume the Gaussian width to be 0.25 cm^{-1}. The Lorentzian widths calculated are 0.13, 0.14, and 0.13 cm^{-1} for ${}^qR_0(0)$, ${}^qR_0(2)$, and ${}^qR_0(4)$, respectively. These values are almost the same as the reported value of 0.127 cm^{-1} [2]. We observed no vibronic bands except for the (0,0,0) band in the UV region by use of the LIF method.

The hole-burning spectrum for the (0,0,0) band is shown in Fig. 2a. The probe frequency was fixed to the ${}^qR_0(0)$ line of the 593 nm band, which means the $N'' = 0$ rotational level in the ground state was selectively monitored. The ${}^qR_0(0)$ line of the (0,0,0) band of the $\tilde{D}^2B_2$–$\tilde{X}^2A_1$ transition was observed as a dip signal at the pump frequency of 40 126.8 cm^{-1} as seen in Fig. 2a. The peak shape was roughly reproduced by the Voigt profile with Lorentzian width of 0.42 cm^{-1} and Gaussian width of 0.25 cm^{-1}. We did not use this spectrum to determine the linewidth, because the S/N ratio is much worse than that of Fig. 1 and the peak shape is broaden by saturation of absorption. We also measured various hole-burning spectra for the monitored levels of $N'' = 0$, 2, and 4, and the ${}^qR_0(0)$, ${}^qR_0(2)$, ${}^qP_0(2)$, ${}^qR_0(4)$, and ${}^qP_0(4)$ lines of the (0,0,0) band were observed. Fig. 2b shows the hole-burning spectrum for the (0,1,0) band. The linewidth of the (0,1,0) band is also reproduced by the Lorentzian profile with the linewidth of 2.4 cm^{-1}, which is unnecessary to be corrected by the laser linewidth of 0.25 cm^{-1}. The fact that the linewidth of the (0,1,0) level is much broader than that of the (0,0,0) level is consistent with other reports [2,12]. The vibrationally excited $\tilde{D}^2B_2$ levels higher than the (0,1,0) level could not been observed in the hole-burning spectra.

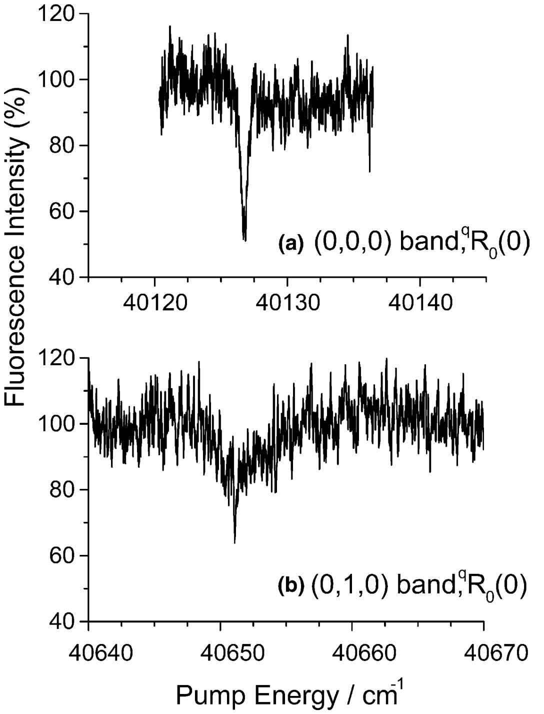

Fig. 2. Hole-burning spectra of the $\tilde{D}^2B_2$–$\tilde{X}^2A_1$ transition of NO_2. The probe laser transition was the ${}^qR_0(0)$ line of the 593 nm band. Upper and lower traces are ${}^qR_0(0)$ lines of the (0,0,0) and (0,1,0) bands, respectively.

In principle, the hole-burning spectroscopy has a great advantage for observation of the predissociative states, because it measures the light-induced depletion of population in the ground state. The hole-burning spectrum of jet-cooled NO_2 is therefore expected to be equivalent to the rotationally selected absorption spectrum. Nevertheless, we could not measure the hole-burning spectrum for the vibronic levels higher than the (0,1,0) level. The existence of N_2O_4 may prevent one to observe these levels. N_2O_4 is known to dissociate to form electronically excited NO_2 in the UV photolysis [17–19]. In our hole-burning experiment, we might detect not only the visible fluorescence of NO_2 induced by the 593 nm probe laser but also the visible NO_2^* chemiluminescence generated by the photodissociation of N_2O_4 at 250–220 nm (pump laser). The concentration of N_2O_4 in the 1% NO_2/Ar mixture of 1 atm total pressure is estimated to be 6.2% at room temperature (293 K) from the equilibrium constant. The absorption cross section of N_2O_4 monotonically increases as the light wavelength becomes shorter in the UV region below 250 nm [20]. The NO_2^* chemiluminescence due to the UV photolysis of N_2O_4 makes a background noise to the probe signal in the hole-burning spectrum, which dilutes the probe depletion fraction in the hole-burning spectrum and makes the observation of the predissociative levels locating above the (0,1,0) level difficult.

The PHOFEX spectrum was measured by fixing the probe laser frequency to the band head of P_{11} branch ($J'' = 8.5$ and 9.5) of NO $A^2\Sigma^+(v' = 0)$ $-X^2\Pi_{1/2}(v'' = 0)$ transition and scanning the pump frequency from 40 900 to 44 000 cm^{-1}. Four broad bands and a few quite diffuse bands overlapped with each other are observed in the PHOFEX spectrum. Fig. 3 shows an example band assigned to the (1,0,0) band by Coon et al. [21], the upper vibronic level of which locates about 300 cm^{-1} above the second dissociation limit indicated by an arrow. The positions of the bands observed in this experiment are almost the same as reported by Coon et al. and are summarized in Table 1, together with the lifetimes calculated from the line-widths.

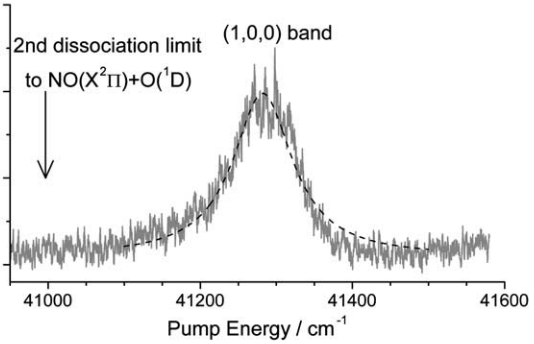

Fig. 3. PHOFEX spectrum of the (1,0,0) band of NO_2 $\tilde{D}^2B_2$–$\tilde{X}^2A_1$ transition (dark gray trace). Black trace is the Lorentzian profile simulated with FWHM of 85.9 cm^{-1}.

At the pump laser energies explored in this PHOFEX study, two possible dissociation channels are accessible: $O(^3P)$ and $O(^1D)$ channels. Slanger et al. [7] studied the nascent NO vibrational distribution by using an LIF technique for the photodissociation of NO_2 at 40 241 cm^{-1} which is slightly lower than the threshold energy of 40 996 cm^{-1} for the $O(^1D)$ channel. They reported a strongly inverted population with a maximum at $v'' = 6$–8 for the nascent NO. McFarlane et al. [10] studied essentially the same experiment except for the detection method of nascent NO, and reported a bimodal vibrational distribution with peaks at $v'' = 0$ and 5. Generally speaking, it is difficult to distinguish the dissociation channel via the predissociative $\tilde{D}^2B_2$ state from the PHOFEX spectra obtained by probing $NO(X^2\Pi_{1/2}, v'' = 0)$ fragment in the photolysis energy range from 40 996 to 42 872 cm^{-1}, because both $O(^1D)$ and $O(^3P)$ channels contribute to the $NO(X^2\Pi_{1/2}, v'' = 0)$ production. The dissociation energy of jet-cooled NO_2 required for the production of $NO(X^2\Pi_{1/2}, v'' = 0, J'')$ and $O(^1D)$, $E_{\text{limit}}(J''_{\text{NO}})$, is calculated as the sum of the second dissociation limit and the rotational energies of nascent NO. If NO_2 in the $\tilde{D}^2B_2$ state dissociates to form $NO(X^2\Pi, v'' = 0, J'')$ and $O(^1D)$, the appearance energy should accord with $E_{\text{limit}}(J''_{\text{NO}})$ in the PHOFEX spectrum obtained by probing $NO(X^2\Pi_{1/2}, v'' = 0, J'')$.

Table 1
Band positions and their assignments of NO_2 $\tilde{D}^2B_2$–$\tilde{X}^2A_1$ transition together with the corresponding lifetimes of the upper levels

Band position (cm^{-1})	Separation (cm^{-1})	Assignment (v'_1, v'_2, v'_3)	Lifetime (this work) (ps)	Lifetime (literature data) (ps)
40 126.02	0	(0,0,0)	41 ± 1.6[a]	42 ± 5[d]
40 651.4	525.4	(0,1,0)	4.8 ± 2.2[b]	2.5 ± 0.5[e]
40 844[e]	718	(0,0,2)		0.155 ± 0.015[e]
41 287	1161	(1,0,0)	0.062 ± 0.006[c]	0.075 ± 0.01[e]
41 814	1688	(1,1,0)	0.1 ± 0.01[c]	0.125 ± 0.015[e]
42 116	1990	(1,0,2)	0.034 ± 0.003[c]	
42 495	2369	(2,0,0)	0.077 ± 0.008[c]	

[a] LIF (this work).
[b] Hole-burning (this work).
[c] PHOFEX (this work).
[d] Absorption [2].
[e] Resonance Raman [12].

Fig. 4 shows the PHOFEX spectra for the (1,0,0) band obtained by probing various rotational levels of the photodissociation product of NO($X^2\Pi_{1/2}$, $v'' = 0$, J''). The shape of the PHOFEX spectrum strongly depends on the rotational quantum number J''. In each spectrum, one can determine the appearance energy to observe a specific rotational level of NO($X^2\Pi_{1/2}$, $v'' = 0$, J''). The appearance energies are determined to be 41 237, 41 280, and 41 323 cm^{-1} for $J'' = 11.5$, 12.5, and 13.5, respectively. These values agree well with $E_{\text{limit}}(J''_{\text{NO}})$, which are indicated by bold arrows in Fig. 4. When NO_2 photodissociates at the photon energy lower than $E_{\text{limit}}(J''_{\text{NO}})$, O(^{3}P) should be produced as a counter product of NO($X^2\Pi_{1/2}$, $v'' = 0$, J''). The clear observation of the NO($X^2\Pi_{1/2}$, $v'' = 0$, J'') production threshold at the corresponding $E_{\text{limit}}(J''_{\text{NO}})$ means that the counter product of NO($X^2\Pi_{1/2}$, $v'' = 0$, J'') is O(^{1}D). This experimental result provides a rigorous evidence showing that NO_2 in the $\tilde{D}(1,0,0)$ level predissociates into NO($^2\Pi$) and O(^{1}D). Uselman and Lee [6] measured the neopentyl alcohol production yield through the insertion reaction of neopentane with O(^{1}D) as a function of NO_2 photolysis energy, and suggested that O(^{1}D) is produced after the excitation through the $\tilde{D}^2B_2$–$\tilde{X}^2A_1$ bands located above the threshold energy of 40 996 cm^{-1}. Our spectroscopic result accords well with their result obtained from the reaction product analysis. They also suggested that the formation of O(^{3}P) is also competitive above the second dissociation limit [6]. There is no indication that the O(^{3}P) channel contributes to the PHOFEX signal, which implies that nascent NO as a counter product of O(^{3}P) is not populated in the $v'' = 0$ level of the $X^2\Pi_{1/2}$ state. Bigio and Grant [22,23] studied the two-photon dissociation of NO_2 near the second dissociation limit, where the probable principal pathway is $\tilde{X}^2A_1 \rightarrow \tilde{A}^2B_2 \rightarrow \tilde{D}^2B_2$ [9]. They mentioned the O(^{1}D) and O(^{3}P) channels to produce NO($X^2\Pi$, $v'' = 0$) and NO($X^2\Pi$, $v'' \sim 6$), respectively. Our result obtained from the one-photon dissociation experiment is consistent with their result derived from the two-photon dissociation.

Fig. 5 shows the lifetimes obtained from the lineshape analysis in this work and the available literature data as a function of the excitation energy [2,12]. The literature data are available only for the energy region below 42 000 cm^{-1}. The agreement between the three data sets seems quite good. One set of data are determined by a resonance Raman method, and probably provide the short-time limits of the lifetimes [12]. The linewidth of the (0,0,0) band was measured to be 0.127 cm^{-1} from the absorption spectrum obtained at room temperature [2], which is almost the same value as ours. It should be mentioned

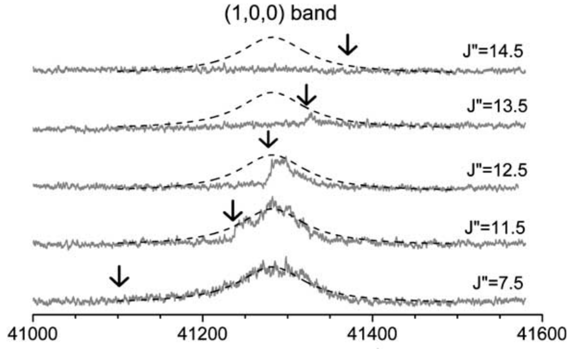

Fig. 4. PHOFEX spectra of the (1,0,0) band obtained by monitoring various J'' values of a photodissociation product NO($X^2\Pi_{1/2}$, $v'' = 0$). The broken lines indicate the simulated Lorentzian profile of Fig. 3. The bold arrows indicate the dissociation limit $E_{\text{limit}}(J''_{\text{NO}})$ to form a specific rotational level of NO($X^2\Pi_{1/2}$, $v'' = 0$, J'') and O(^{1}D).

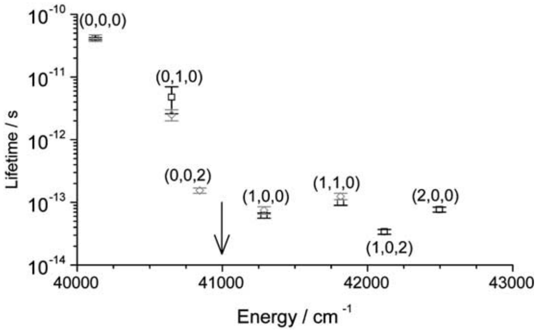

Fig. 5. Lifetimes of various (v_1', v_2', v_3') levels obtained from this work and the available literature as a function of excitation energy: □, this work by LIF/hole-burning/PHOFEX; ○, absorption [2]; ◇, resonance Raman [12]. The bold arrow indicates the dissociation limit $E_{\text{limit}}(J''_{\text{NO}} = 0)$ to form a specific rotational level of NO($X^2\Pi_{1/2}$, $v'' = 0, J'' = 0$) and O(^{1}D).

that the rotational level ($N' = 9$, $K' = 3$) analyzed in absorption spectrum by Hallin and Merer is different from those ($N' = 1$, 3, 5 of the $K' = 0$ manifold) in our experiment. This implies that the lifetimes are independent of the rotational quantum number; in other words, the predissociation proceeds through homogeneous interaction. The homogeneous interaction between the $\tilde{D}^2B_2$ and $\tilde{A}^2B_2$ states is one of the probable candidate to explain the predissociation mechanism of the present interest.

It is found that all the vibronic bands studied by the PHOFEX method have extremely broad linewidths, and the corresponding lifetimes are shorter than that of the (0,0,0) band. More importantly, the steep decrease in lifetime occurs beyond the (0,0,0) band. The lifetime reaches about 100 fs at the (0,0,2) level, which is located 152 $\mathrm{cm^{-1}}$ below the second dissociation limit, and no marked energy dependence has been noticed in the energy region above the (0,0,2) level. It is clearly demonstrated that the second dissociation limit is not important to determine the lifetime. This trend of lifetimes indicates that the predissociation rate to form $NO(^2\Pi)$ and $O(^3P)$ increases sharply as the vibrational energy increases in the $\tilde{D}^2B_2$ state. Roughly speaking, the predissociation rate of the (0,0,2) levels is 260 times faster than that of the (0,0,0) level. If the predissociation occurs via the $\tilde{A}^2B_2$ state as mentioned before, the linewidth Γ of the peaks is expressed in the Fermi's golden rule as $\Gamma = 2\pi W^2 \rho$, where W is the interaction term between the $\tilde{D}^2B_2$ and $\tilde{A}^2B_2$ states, and ρ is the density of states in the $\tilde{A}^2B_2$ state. The density of the $\tilde{A}^2B_2$ state is quite high because energy gap between the (0,0,0) levels in the $\tilde{D}^2B_2$ and $\tilde{A}^2B_2$ states is as large as 30 394 $\mathrm{cm^{-1}}$. At least we can safely say that the density of states of the coupled states does not change drastically over the excitation energy region of 40 000–41 000 $\mathrm{cm^{-1}}$, where we recognize a sharp rise in the predissociation rate.

This trend of the predissociation rate seems consistent with that of the $O(^1D)$ quantum yield obtained by Uselman and Lee [6]. The quantum yield of $O(^1D)$ is almost constant around 0.5 above the second dissociation limit. The predissociation rate reaches about 10^{13} $\mathrm{s^{-1}}$ at (0,0,2), which locates below the second dissociation limit. This observation implies that the predissociation rate into the $O(^3P)$ channel is high enough to compete with the $O(^1D)$ channel in the energy region above the second dissociation limit. The competition of the predissociation channel results in almost the same values of quantum yields of $O(^1D)$ and $O(^3P)$ above the second dissociation limit.

In summary, the LIF, hole-burning, and PHOFEX spectra were measured for jet-cooled NO_2 and the vibrational level dependence of the lifetime for NO_2 in the $\tilde{D}^2B_2$ state has been investigated. The lifetimes for the vibrational levels in the $\tilde{D}^2B_2$ state are estimated from the line profiles of the LIF, hole-burning, and PHOFEX spectra, which increase steeply as the excitation energy increases even below the second dissociation limit to form $NO(^2\Pi_{1/2})$ and $O(^1D)$. The appearance thresholds were observed in the PHOFEX spectra obtained by monitoring various J''-levels of nascent NO, which accord with the calculated energies required for forming $NO(^2\Pi_{1/2}, v'' = 0, J'')$ and $O(^1D)$. This observation clearly indicates the occurrence of the $O(^1D)$ dissociation pathway via the $\tilde{D}^2B_2$ state.

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