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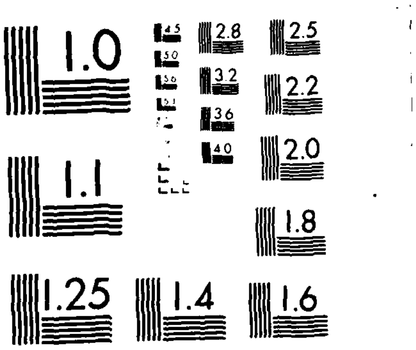

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TECHNICAL REPORT BRL-TR-2625

INVESTIGATION OF SEVERAL LOW-LYING LEVELS OF X DOUBLET PI STATE OF NCO USING ARGON LASER EXCITED FLUORESCENCE

Koon Ng Wong
William R. Anderson
Anthony J. Kotlar
John A. Vanderhoff

December 1984

US ARMY BALLISTIC RESEARCH LABORATORY
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The laser excited fluorescence spectrum of NCO from an atmospheric pressure $CH_4/N_2O/N_2$ flame is reinvestigated with a more sensitive apparatus than used previously. The NCO is pumped from the X doublet pi (1,0,0) level to the A doublet sigma plus (0,0,0) level. Emissions from the excited A state level to the following X state vibrational levels are observed: doublet pi (0,0,0), (1,0,0), and (0,0,1); doublet sigma plus (0,1,0). The previous tentative assignment of pumping transition as Q2(31) is confirmed. The confirmation

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of pumping transition solidifies our earlier interpretation that excited state spin relaxation in the flame is much faster than electronic quenching. In addition, analysis of the fluorescence spectra yields the first reported constants for the X doublet pi (1,0,0) level and spin splitting for the X doublet pi (0,0,1) level in the gas phase.

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I. INTRODUCTION

Recently we reported the laser excited fluorescence (LEF) spectra from the A-X system of the free NCO radical in a rich atmospheric pressure $CH_4/N_2O/N_2$ flame using a fixed wavelength argon ion laser.[1] NCO is of interest because thermal decomposition studies of various gun propellants have shown that HCN is formed as one of the decomposition products.[2] Shock tube studies of the oxidation of HCN indicate that NCO is an important intermediate in the further reaction of HCN.[3] All nine lines (4545 to 5145Å) of the argon ion laser pump vibrational bands of the $A^2\Sigma^+ - X^2\Pi$ electronic system. The 4658Å pump line was selected for detailed study because it pumps directly to the $A^2\Sigma^+$ $(0,0^0,0)$ level. (See the Appendix for a short explanation of the terminology used for vibrational levels.) Since no vibrational down-transfer in the excited state can occur, the resulting fluorescence spectra are simpler than for the other pump lines. In addition, though this is one of the weakest laser lines, the fluorescence from this pump line is the strongest.[1] This is probably because the line pumps from one of the lowest vibrational levels, $X^2\Pi$ $(1,0^1,0)$, while the other lines pump from higher levels. It was firmly established in Ref. 1 that NCO is pumped to $N'=31$ by this line. However, the lower rotational level (N", J") from which pumping occurs has not been confirmed due to C_2 and grating ghost interferences in the region of the laser line. In addition, emission to the $(0,0^1,1)$ vibrational level of the $X^2\Pi$ ground electronic state was not observed because of the deterioration of the laser power in the later part of the study.

The A-X system of NCO was first observed in low resolution emission spectra by Holland, et al.[4] An extensive vibrational and rotational analysis of this electronic system was performed by Dixon.[5] Bolman, et al.,[6]

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[1] W.R. Anderson, J.A. Vanderhoff, A.J. Kotlar, M.A. DeWilde, and R.A. Beyer, "Intracavity Laser Excitation of NCO Fluorescence in an Atmospheric Pressure Flame," BRL Technical Report ARBRL-TR-02527, September 1983, A134740.

[2] (a) R.A. Beyer, "Molecular Beam Sampling Mass Spectrometry of High Heating Rate Pyrolysis: Description of Data Acquisition System and Pyrolysis of HMX in a Polyurethane Binder," BRL Memorandum Report ARBRL-MR-02816, 1978, A054328. (b) C.U. Morgan and R.A. Beyer, "ESR and IR Spectroscopic Studies of HMX and RDX Thermal Decomposition," 15th JANNAF Combustion Meeting, Newport, RI, September 1978.

[3] R.A. Fifer and H.E. Holmes, "Kinetics of the HCN + NO_2 Reaction Behind Shock Waves," J. Phys. Chem., Vol. 86, p. 2935, 1982.

[4] R. Holland, D.W.G. Style, R.N. Dixon, and D.A. Ramsay, "Emission and Absorption Spectra of NCO and NCS," Nature (London), Vol. 182, p. 336, 1958.

[5] R.N. Dixon, "The Absorption Spectrum of the Free NCO Radical," Phil. Trans. R. Soc. London, Vol. 252, p. 165, 1960.

[6] P.S.H. Bolman, J.M. Brown, A. Carrington, I. Kopp, and D.A. Ramsay, "A Re-Investigation of the $A^2\Sigma^+ - X^2\Pi$ Band System of NCO," Proc. R. Soc. London Ser. A, Vol. 343, p. 17, 1975.

reinvestigated the A-X system at higher resolution. The free NCO radical has also been studied by electron paramagnetic resonance[7] and by microwave spectroscopy.[8,9] These investigations provide much information concerning the upper state, the ground state, and its associated bending vibrational levels. However, there is little information available on the two stretching vibrations of the $X^2\Pi$ ground electronic state. Using the matrix isolation technique, Milligan and Jacox[10] were able to obtain values of the two stretching frequencies, which were confirmed later by Bondybey and English[11] in their study of the LEF spectra of NCO in an argon matrix. Recently Barnes, et al.,[12] obtained spectroscopic parameters in the $(0,0^1,1)$ vibrational level of the $X^2\Pi$ state by using CO laser magnetic resonance.

For the reasons mentioned above, we reinvestigated the LEF spectra of NCO using the 4658Å pump line when the argon ion laser was in a good working condition. (A new, higher power tube was installed). All emissions from the $A^2\Sigma^+$ $(0,0^0,0)$ level to the ground state levels observed in the argon matrix work,[11] i.e., $(0,0^1,0)^2\Pi$, $(1,0^1,0)^2\Pi$, $(0,1^0,0)^2\Sigma^+$, and $(0,0^1,1)^2\Pi$, have been observed.* The C_2 interferences in the $(1,0^1,0)$ band have been minimized (vide infra) and our tentative assignment that $Q_2 31$ is the pumping transition has been confirmed. Furthermore, these higher S/N spectra allow analyses of the $(0,0^1,0)$, $(1,0^1,0)$, and $(0,0^1,1)$ emission bands yielding values for the two stretching vibrational frequencies, the rotational constants, and the

---

[7] A. Carrington, A.R. Fabris, B.J. Howard, and N.J.D. Lucas, "Electron Resonance Studies of the Renner Effect. I. Gaseous NCO In Its $^2\Pi_{3/2}$ (n=1), $^2\Delta_{5/2}$ (n=2), and $^2\Phi_{7/2}$ (n=3) Vibronic States," Mol. Phys., Vol. 20, p. 961, 1971.

[8] S. Saito and T. Amano, "Microwave Spectrum of the NCO Radical," J. Mol. Spectrosc., Vol. 34, p. 383, 1970.

[9] T. Amano and E. Hirota, "Hyperfine Interactions of the Free NCO Radical in the Δ Vibronic State (v_2=1)," J. Chem. Phys., Vol. 57, p. 5608, 1972.

[10] D.E. Milligan and M.E. Jacox, "Matrix Isolation Study of the Infrared and Ultraviolet Spectra of the Free Radical NCO," J. Chem. Phys., Vol. 47, p. 5157, 1967.

[11] V.E. Bondybey and J.H. English, "Fermi Resonance and Vibrational Relaxation in the $A^2\Sigma$ State of NCO in Solid Argon," J. Chem. Phys., Vol. 67, p. 2868, 1977.

[12] C.E. Barnes, J.M. Brown, A.D. Fackerell, and T.J. Sears, "The Laser Magnetic Resonance Spectrum of the NCO Radical at 5.2 μm," J. Mol. Spectrosc., Vol. 92, p. 485, 1982.

*For the sake of convenience, we will refer to them as the $(0,0^1,0)$, $(1,0^1,0)$, $(0,1^0,0)$, and $(0,0^1,1)$ emission bands from hereon.

spin-orbit coupling constants in all these levels. The $(1,0^1,0)$ band has not been reported previously in gaseous phase. The results are compared to those from the earlier studies mentioned above and also to the preliminary results of a concurrent LEF study.[13] Excellent agreement is observed.

II. EXPERIMENTAL DETAILS

The experimental arrangement is similar to that described earlier,[1] and only a brief description will be given here. A nominal 8 W (all lines) Ar^+ laser was used as the excitation source. The 4658Å line, which has a power of about 0.5 W, was selected for this study. A knife-edge burner[14] supporting a $CH_4/N_2O/N_2$ flame was placed inside the extended cavity of the laser. The intracavity circulating power was approximately 20 times more intense than the extracavity power (i.e., 10 W). The burner was placed in a horizontal position with the open channel facing the detection optics. The scattered light perpendicular to the beam was imaged onto the 100 μm slit of a 1 m monochromator. A glass dove prism was placed in front of the slit to rotate the image 90°. Instead of a photomultiplier tube with photon counting electronics, as used in our previous work,[1] an optical multichannel analyzer (OMA) with a silicon intensified vidicon tube was used to detect the LEF signal. A laboratory computer (PDP 11/34) directly interfaced to the OMA allows for the averaging of spectra for long time periods. In order to remove flame background emissions, the spectra were recorded with and without the laser, then subtracted to obtain the LEF spectra. The results were obtained with approximate flame conditions of equivalence ratio $\phi = 1.6$ with 40% dilution with N_2.*

The bands displayed in Figures 1, 2, and 3 are actually made up of two or more sections of the LEF spectra taken at different times. The arrows indicate the locations at which these components are joined together to form the entire band. A section of the entire band was taken with the monochromator set at a certain wavelength. The spectra were taken in second order. Approximately 35Å of the spectrum was accumulated into the 500 channels of the OMA at any one time. The achieved resolution was about 0.4Å ($1.8\ cm^{-1}$) FWHM.

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[13] (a) R.A. Copeland, D.R. Crosley, and G.P. Smith, "Laser-Induced Fluorescence Spectroscopy of NCO and NH_2 in Atmospheric Pressure Flames," submitted to the 20th Symposium (International) on Combustion, Ann Arbor, MI, August 1984. (b) D.R. Crosley, private communication.

[14] R.A. Beyer and M.A. DeWilde, "Simple Burner for Laser Probing of Flames," Rev. Sci. Instrum., Vol. 53, p. 103, 1982. A description of the burner suitable for purposes of understanding the spectroscopy discussed in the present work is contained in Ref. 1.

*Since some confusion has resulted in the past, we should note here that 40% dilution with N_2 means 40% of the premixed gases flowing in the burner's main channel is N_2. Small additional flows of N_2 at the two ends of the burner are used to keep the flame from curling around the knife edges. These additional flows do not alter conditions at the center of the flame where measurements are taken.

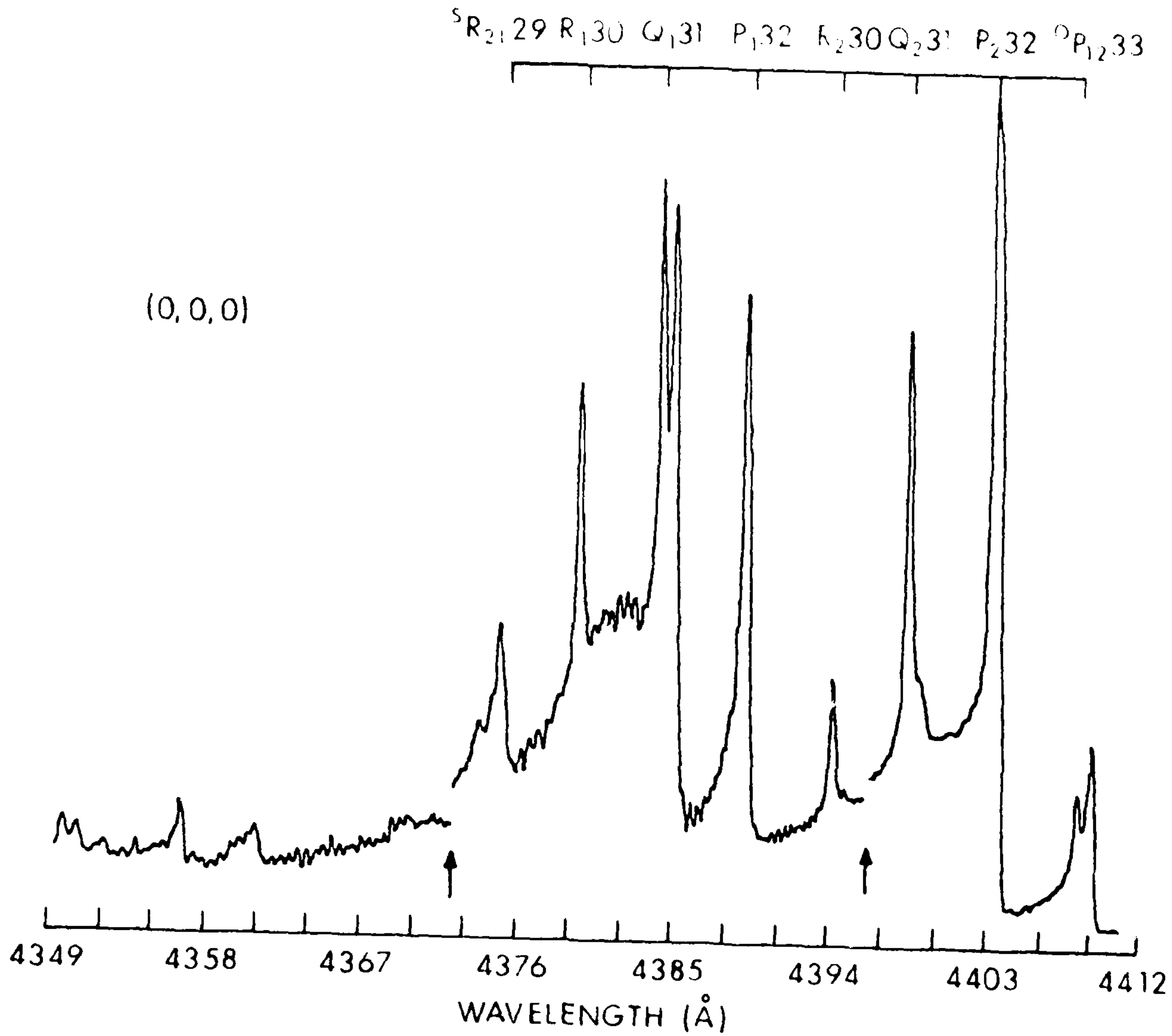

Figure 1. The $A^2\Sigma^+$ $(0,0^0,0) \rightarrow X^2\Pi$ $(0,0^1,0)$ Fluorescence Spectrum. Unlabeled peaks between 4349 and 4367Å are heads of the $A^2\Pi$ $(0,1^1,0) \rightarrow X^2\Delta$ $(0,1^2,0)$ transition.[1] The upper level is apparently excited via a small amount of vibrational up-transfer from $A^2\Sigma^+$ $(0,0^0,0)$.

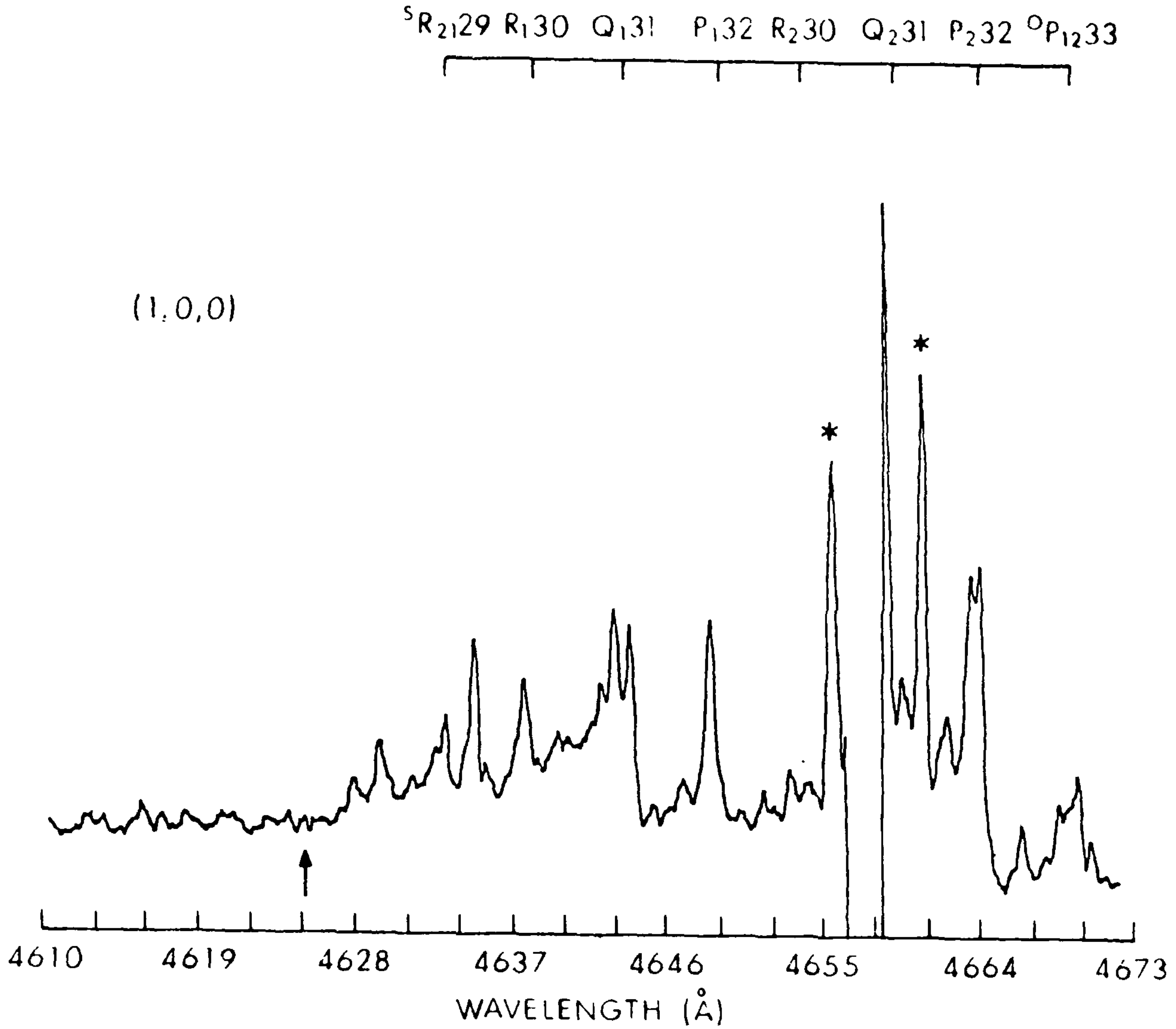

Figure 2. The $A^2\Sigma^+$ $(0,0^0,0) \rightarrow X^2\Pi$ $(1,0^1,0)$ Fluorescence Spectrum. The spectrum drops to zero near 4658Å *because of detection system saturation* by *laser scatter*. The two peaks labeled (*) are grating ghosts.

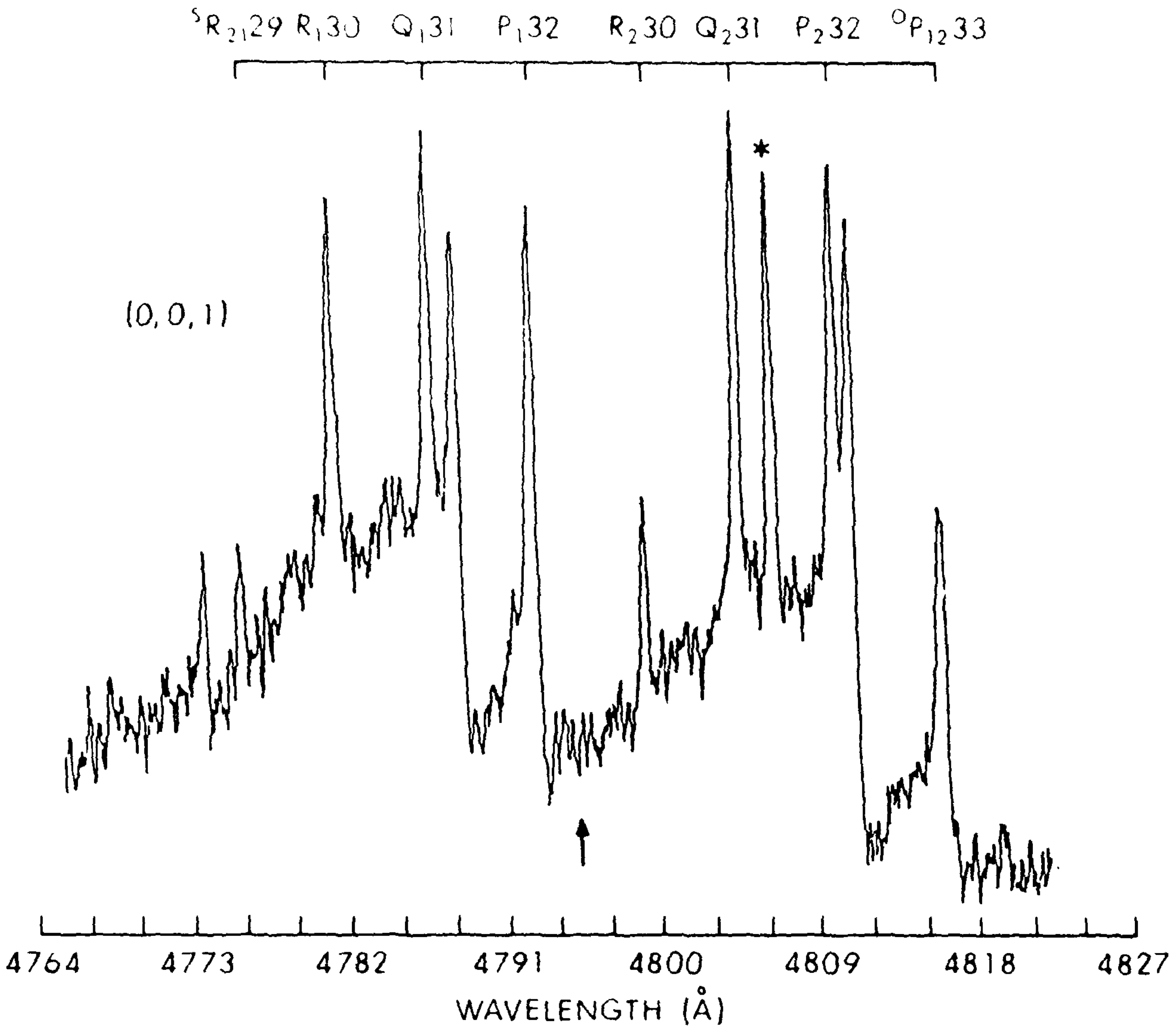

Figure 3. The $A^2\Sigma^+ (0,0^0,0) \rightarrow X^2\Pi (0,0^1,1)$ Fluorescence Spectrum. The peak at ~4772Å is unassigned. The peak labeled (*) is an argon discharge line from the laser.

Recording of spectra with the monochromator-OMA system takes much less time than with the earlier photon counting system. A 1000 scan spectrum, for instance, takes about three to four minutes. The increase in number of scans allows for a signficant increase in S/N via signal averaging. Another major advantage of the OMA is that it allows one to look at large segments of the spectrum very quickly, often in real time. However, wavelength nonlinearities have been observed across the face of the vidicon tube. These nonlinearities occur because the grating focusses various wavelengths in an arc at the monochromator exit, where the vidicon is placed. Since the vidicon face is flat, the wavelength scale is stretched near the edges. The effect also causes some loss of resolution near the edges of a scan. Fortunately, the mid-section of the spectrum, i.e., from about 100th to 400th channel, is quite linear and gives accurate frequencies (wavelengths). For example, the frequencies of the eight lines determined for the $(0,0^1,0)$ band differ with those of Bolman, et al.,[6] by less than 1 cm^{-1}, despite the moderate resolution employed in this work. For wavelength calibration, the spectra of Ar discharge lines, obtained by imaging stray light from the laser tube, and of Na emissions from a Na vapor lamp were taken under exactly the same conditions as fluorescence spectra. The wavelengths of the reference lines were taken from the MIT tables.[15]

III. RESULTS AND DISCUSSION

As mentioned previously, the 4658Å line of the argon ion laser pumps NCO to the $(0,0^0,0)$ level of the $A^2\Sigma^+$ electronic state. Emissions from $A^2\Sigma^+$ $(0,0^0,0)$ to all of the fundamental vibrational levels of the $X^2\Pi$ ground electronic state were observed. They are shown in Figures 1, 2, 3, and 4, corresponding to the $(0,0^1,0)$, $(1,0^1,0)$, $(0,0^1,1)$, and $(0,1^0,0)$ emission bands, respectively. The relative intensities of these bands agree qualitatively with those observed in a low temperature argon matrix.[11] The $(0,0^1,0)$ band is the most intense. Emission bands that involve excitation of the ground state stretching vibrations are stronger than that involving the bending vibration. The $(0,0^1,0)$, $(1,0^1,0)$, and $(0,0^1,1)$ emission bands do not involve the bending mode. Therefore, complications such as ℓ-type doubling and Renner effect are absent. Since these transitions all have the overall symmetry $^2\Sigma^+ - {}^2\Pi$, it is expected that similar structures be observed in these bands.[1] For a $^2\Sigma^+ - {}^2\Pi$ transition, there are three main branches and three satellite branches in each subband $^2\Sigma^+ - {}^2\Pi_{1/2}$ and $^2\Sigma^+ - {}^2\Pi_{3/2}$. However, since the spin splitting in the upper state is very small,[5,6] four of these satellite branches coincide with main branches, giving rise to only four distinct branches in each subband. The assignments of these bands are straightforward. They are given in Table 1 and illustrated in Figures 1, 2, and 3. Now, from the previous work on the $(0,0^1,0)$ band, we observed that lines from the directly pumped level N'=31 are the most prominent features of the spectra.[1] However, considerable rotational transfer in the excited state occurs, yielding lines from many other N' levels as well. Bandheads (not labeled in the figures), therefore, are also expected to appear in each of the bands just to the long wavelength side (or perhaps overlapping) the Q_1, P_1,

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[15] Atomic line positions were obtained from G.R. Harrison, Massachusetts Institute of Technology Wavelength Tables, MIT, Cambridge, MA, 1969.

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APPENDIX A.

VIBRATIONAL LEVEL TERMINOLOGY AND THE RENNER-TELLER EFFECT IN LINEAR TRIATOMIC MOLECULES

The vibrational level terminology is discussed in detail elsewhere.* A short explanation of the terminology is given here. The designation of the form (v_1, v_2^K, v_3) refers to a vibrational level as follows. v_1 and v_3 refer, respectively, to symmetric and antisymmetric stretching vibrational quantum numbers of the molecule. Here, it should be noted that although NCO has no center of inversion symmetry, the NC and CO stretching vibrations are strongly coupled.[5] Therefore, the vibrational stretching motions are similar to those of CO_2. It is thus proper to use the terms symmetric and antisymmetric in reference to the stretching motions in NCO. v_2 refers to the quantum number of the doubly degenerate bending mode. For such degenerate vibrations, the molecule possesses a quantized vibrational angular momentum designated by quantum number, ℓ . ℓ has possible values v_2, v_2-2, ..., 1 or 0. In the case of linear molecules for which Λ, the electronic orbital angular momentum quantum number is well-defined, ℓ and Λ add vectorially to yield a new quantum number K which takes on all possible values of $|\pm\Lambda\pm\ell|$. Note that K is the superscript on the v_2 quantum number in the vibrational level designation. The value of K determines the overall symmetry (electronic X vibrational) of the state, i.e., K = 0, 1, 2, 3 ... corresponds to Σ, Π, Δ, Φ ... states, respectively. Here, Σ levels are singly and Π, Δ, Φ ... levels doubly degenerate, as usual. Now, for $v_2 = 0$, we must have $\ell=0$ so that $K=\Lambda$ and only one vibrational state results. Thus, for $A^2\Sigma^+$ (where $\Lambda=0$) and $X^2\Pi$ (where $\Lambda=1$) $v_2 = 0$ can yield only a $^2\Sigma^+$ and a $^2\Pi$ vibrational level, respectively. For $v_2 > 0$, ℓ may take on values greater than 0 and several states will result. The energies of these states will differ because the bending vibration destroys the cylindrical symmetry of the molecule, removing their degeneracy. This effect is known as the Renner-Teller effect. As an example of interest in the present paper, consider the value $v_2 = 1$ in the $X^2\Pi$ electronic state. Since $v_2 = 1$, ℓ must equal 1. For the $X^2\Pi$ state, $\Lambda=1$. Thus, the sum rule $K = |\pm\Lambda\pm\ell|$ results in four values, 2, 2, 0, and 0. Of these, the two levels with K=2 will be equal in energy, yielding a single, doubly degenerate Δ state. The two K=0 values will have unequal energies yielding a Σ^+ and a Σ^- state. Therefore, if we consider the (0,1,0) level in the $X^2\Pi$ state, this level will actually be split into three levels, namely, $X^2\Delta\ (0,1^2,0)$, $X^2\Sigma^-\ (0,1^0,0)$, and $X^2\Sigma^+\ (0,1^0,0)$. Dipole radiation selection rules only allow $\Delta K=0, \pm 1$ transitions to occur. Furthermore, $\Sigma^+ \rightarrow \Sigma^-$ transitions are rigidly forbidden. Therefore, when we pump the $A^2\Sigma^+\ (0,0^0,0)$ level, the only emission to the (0,1,0) manifold which is observed is to the $X^2\Sigma^+\ (0,1^0,0)$ component.

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*G. Herzberg, Molecular Spectra and Molecular Structure. III. Electronic Spectra of Polyatomic Molecules, Van Nostrand Reinhold, New York, pp. 20-37, 1966.

APPENDIX A.

VIBRATIONAL LEVEL TERMINOLOGY AND THE RENNER-TELLER EFFECT IN LINEAR TRIATOMIC MOLECULES

13. (a) R.A. Copeland, D.R. Crosley, and G.P. Smith, "Laser-Induced Fluorescence Spectroscopy of NCO and NH_2 in Atmospheric Pressure Flames," submitted to the 20th Symposium (International) on Combustion, Ann Arbor, MI, August 1984. (b) D.R. Crosley, private communication.

14. R.A. Beyer and M.A. DeWilde, "Simple Burner for Laser Probing of Flames," *Rev. Sci. Instrum.*, Vol. 53, p. 103, 1982. A description of the burner suitable for purposes of understanding the spectroscopy discussed in the present work is contained in Ref. 1.

15. Atomic line positions were obtained from G.R. Harrison, Massachusetts Institute of Technology Wavelength Tables, MIT, Cambridge, MA, 1969.

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3. R.A. Fifer and H.E. Holmes, "Kinetics of the HCN + NO_2 Reaction Behind Shock Waves," J. Phys. Chem., Vol. 86, p. 2935, 1982.

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stretching vibrations, about which little is currently known. Excited state spin relaxation, with retention of N' identity, is faster than electronic quenching by flame molecules.

The $(0,1^0,0)$ level involves one quantum of bending vibration in the $^2\Pi$ ground state. Because of the Renner effect and spin-orbit splitting, there are four substates, $^2\Sigma^+$, $^2\Delta_{5/2}$, $^2\Delta_{3/2}$, and $^2\Sigma^-$, for this level.[5] Since the upper state pumped by the laser is $^2\Sigma^+$, one expects emission only to the $^2\Sigma^+$ substate due to appropriate selection rules. For a $^2\Sigma^+ - {}^2\Sigma^+$ transition, the selection rule $\Delta N = \pm 1$ holds, $\Delta N = 0$ being forbidden. Furthermore, the ground state spin splitting is larger than that of the excited state.[5] Thus, one expects to observe doublet P and R branches. This is exactly what is observed in the $(0,1^0,0)$ emission band displayed in Figure 4. There is an extensive analysis of this band in Ref. 6 so that further analysis here would be superfluous. Using these results, the peaks at 4473.2 and 4473.9Å are assigned as the $R_2 30$ and $R_1 30$ and those at 4482.8 and 4483.6Å as the P_2 and P_1 heads, respectively. $P_2 32$ and $P_1 32$ are not resolved from their respective heads.

Information about excited state energy transfer can also be obtained from these spectra, as previously discussed.[1] As mentioned earlier, the $Q_1 31$ and $Q_2 31$ transitions have approximately equal intensities while $R_1 30$ is about two times stronger than $R_2 30$ (Figures 1-3). Now, the Q_1, Q_2, and R_1 branches all have unresolved satellites, namely, $^QP_{21}$, $^QR_{12}$, and $^RQ_{21}$, respectively. One expects linestrengths of Q_1 and Q_2 to be nearly equal and stronger than $^QP_{21}$ and $^QR_{12}$ which are nearly equal. Calculations[6,18] show the linestrengths of $R_2 30$, $R_1 30$, and $^RQ_{21} 30$ all to be nearly equal. Therefore, comparison of either the Q-branch or the R-branch intensities indicates the laser produces approximately equal densities in the N'=31 spin components. This could occur via equal pumping rates of the two spin components. However, we have shown unambiguously that $Q_2 31$ in the $(1,0^1,0)$ band is the pumping transition. Equal pump rates are not possible in this branch[1] because the $Q_2 31$ and $^QR_{12} 31$ occur at nearly the same frequency and strongly overlap at 2500 K. Further, the Q_2 linestrength is about three times that of the $^QR_{12}$.[6,18] Therefore, our earlier tentative interpretation that the molecule undergoes fast excited state spin relaxation with retention of N' identity in the flame is solidified. Furthermore, we point out that the spin relaxation rate must be much faster than electronic quenching. Otherwise, the nascent spin distribution excited by the laser would be frozen, since quenching would occur before spin redistribution.

IV. CONCLUSIONS

Laser excited fluorescence spectra of NCO in a $CH_4/N_2O/N_2$ flame have been observed using the 4658Å line of an argon ion cw laser. Four bands were obtained as a result of emission from the $A^2\Sigma^+ (0,0^0,0)$ level to different levels of the ground state. Three of the four bands display similar, 8-branch structures because they possess the same vibronic species in the upper and lower state, i.e., they are all $^2\Sigma^+ \rightarrow {}^2\Pi$ transitions. The structure of the other band is considerably simpler because it is a $^2\Sigma^+ \rightarrow {}^2\Sigma^+$ transition. All of the prominent lines in these bands originate from the N'=31 rotational level of the $A^2\Sigma^+ (0,0^0,0)$ state. The region of the laser line, i.e., the $A^2\Sigma^+ (0,0^0,0) \rightarrow X^2\Pi (1,0^1,0)$ transition, has been reinvestigated under improved experimental conditions and the previous $Q_2 31$ pumping transition assignment is confirmed. Rotational analyses of the $(0,0^1,0)$, $(1,0^1,0)$, and $(0,0^1,1)$ emission bands have also been performed. The spin-orbit coupling constants and the rotational constants were obtained for these states. This analysis yields valuable information concerning the two ground state

TABLE 2. COMPARISON OF RESULTS OF ANALYSIS FOR SPECTRAL CONSTANTS (cm^{-1}) OF NCO

LEVEL	A" (SPIN-ORBIT PARAMETER)			B" (ROTATIONAL CONSTANT)		
$(0,0^1,0)$	-95.585[a]	-95.6[b]	-95.35 ± 0.44[c]	0.38952[d]	0.3895[b]	0.3913 ± 0.0030[c]
$(1,0^1,0)$	-------	-90.0[b]	-90.17 ± 0.40[c]	-------	0.3883[b]	0.3892 ± 0.0027[c]
$(0,0^1,1)$	-------	-97.3[b]	-97.16 ± 0.13[c]	0.38618[e]	0.3861[b]	0.3865 ± 0.0009[c]

ν_1	-------	1275[f]	1272[g]	1272.0[b]	1270.3 ± 2.7[c]
ν_3	1921.4[e]	1922[f]	1923[g]	1922.5[b]	1921.06 ± 0.89[c]
T_o	22754.06[a]	-----	-----	------	22755.3 ± 3.0[c]

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a - Ref. 6
b - Ref. 13
c - This work. Error limits are one standard deviation.
d - Derived from microwave data[8,9] in Ref. 6. Rounded to five digits.
e - Ref. 12, rounded to five digits. $B"_{0,0,1}$ is obtained by combining $B"_{0,0,0}$ and α_3. The value for ν_3 is increased by 0.8 cm^{-1} from the value 1920.6 cm^{-1} given in Ref. 12 (see text).
f - Ref. 10, Ar matrix results.
g - Ref. 11, Ar matrix results.

obtained from the fit to the $(0,0^1,0)$ data of Bolman, et al., was used and assumed to be the same for $(1,0^1,0)$ and $(0,0^1,1)$. (This value differs from that of Refs. 5 and 6, which may be ascribed to the presence of several higher order terms in the Hamiltonian used and to the fact that several of the constants of Bolman, et al. were obtained from other sources and fixed. All of the parameters in the present fit to the $(0,0^0,0)$-$(0,0^1,0)$ band were allowed to vary.) Also, to insure that reliable values for G", A", and B" could be obtained from a set of eight transitions having the same value of N', the appropriate eight transitions were selected from the data of Bolman, et al., and were fitted in the same manner as the LEF data. The results for G", A", and B" were within error limits of those from the fit of all the $(0,0^1,0)^2\Sigma^+ - (0,0^0,0)^2\Pi$ band transitions and had rather small error limits. The experimental results for the $(0,0^1,0)$ band were next fitted. The results are shown in Table 2 and exhibit excellent agreement with Refs. 5 (not shown), 6, and 8 (though the error limits are larger, as expected, because of the moderate resolution and the small number of available lines).* These results illustrate the validity of the technique. Finally, the $(1,0^1,0)$ and $(0,0^1,1)$ bands were fitted. For the $(1,0^1,0)$ band, the laser wavelength[15] was used as the $Q_2 31$ line position. Now, Q_2 branch line spacings near $Q_2 31$ in the $(0,0^1,0)$ band are ~1 cm^{-1}.[5,6] One would expect similar spacings for this branch in the $(1,0^1,0)$ band. Thus, the laser line must be within ~0.5 cm^{-1} of $Q_2 31$ in the $(1,0^1,0)$ band, or the laser would pump a different Q_2 line. As 0.5 cm^{-1} is the approximate precision limit of the measurements, it was deemed reasonable to use the laser line position in the fit. The standard deviations of the fits for the $(0,0^1,0)$, $(1,0^1,0)$, and $(0,0^1,1)$ bands were 0.60, 0.54, and 0.18 cm^{-1}, respectively. The constants are listed in Table 2.

There are a few things to point out about these results. First, Table 2 shows that the spin-orbit coupling constant changes significantly with different vibrations. Second, the values obtained for the $(0,0^1,1)$ emission band are much more precise than the rest, probably because of the extremely sharp and well-defined peaks of this band (Figure 3). Third, despite the moderate resolution used and the limited number of transitions, satisfactory results are obtained (see Table 2). For example, the constants obtained for the $(0,0^1,0)$ level agree with those determined with much higher resolution[5,6,8] to within one standard deviation. Similarly, constants for the $(0,0^1,1)$ band agree with those from laser magnetic resonance (LMR) data[12] and from matrix studies.[10,11] Note that ν_3 from Ref. 12 is slightly smaller than it should be because the authors, having only LMR transitions in the $^2\Pi_{3/2}$ component available, were forced to use $A''_{0,0,0}$ from Ref. 6 in their analysis of the $(0,0^1,1)$ level. (This should not affect results for $B''_{0,0,1}$.) To first order, their result for ν_3, 1920.6 cm^{-1}, may be corrected by adding half of the difference in A" values for the two levels, ~0.8 cm^{-1}, to obtain 1921.4 cm^{-1}. Comparing our data with the preliminary results of Copeland, Crosley, and Smith,[13] we see that the agreement is excellent except for ν_3. Gas phase constants for the $(1,0^1,0)$ level and for $A''_{0,0,1}$ have not previously been reported.

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*For argument's sake, we repeated this fit, but assumed N'=30 and then N'=32. The results did not agree with those of the earlier references at all.

Swan system fluorescence in this region also excited by the 4658Å line.[16] However, the relative flame density profiles (Figure 11, Ref. 1) indicate that the concentration of C_2 drops much more quickly than that of NCO in the post-flame region. Thus, it is expected that in this region the concentration of NCO will increase relative to C_2. The best $(1,0^1,0)$ spectrum that could be obtained in this manner is shown in Figure 2. It is seen that most of the C_2 peaks disappear (compare to Figure 9, Ref. 1) and the spectrum is amenable to analysis. A pair of strong peaks about the laser line are grating ghosts (determined experimentally by removing the flame). At the violet end of the band complications arise from unexpected lines. These complications may come from the $(0,0^0,0)\ ^2\Sigma^+ - (0,2^1,0)^2\Pi^+$ band, which appears because of a Fermi resonance between the $(0,2^1,0)$ level and the $(1,0^1,0)$ level.[11] In spite of these problems, features clearly analogous to those in Figures 1 and 3, namely $P_2 32$, $P_1 32$, $Q_1 31$ (and their associated bandheads) and $R_1 30$, are easily recognizable in Figure 2. In addition, small features assignable as the $^{O}P_{12}33$ (and associated bandhead), $R_2 30$ and $^{S}R_{21}29$ are present. The only feature not present is the $Q_2 31$, which is hidden by the intense laser scatter. The laser line position is centered between the two strong grating ghosts (see Figure 2). Clearly, the spacing of prominent lines is proper for assignment at this position as the $Q_2 31$, unambiguously identifying this line as the pumping transition and confirming our previous tentative assignment.[1] No attempts were made to analyze lines from the $(0,2^1,0)\ ^2\Pi^+$ band.

The vibrational term value, G", the spin-orbit coupling constant, A", and the rotational constant, B", were determined for the $(0,0^1,0)$, $(1,0^1,0)$, and $(0,0^1,1)$ levels of the ground state using the assigned transitions of the appropriate LEF spectra (Figures 1-3). A standard doublet Hamiltonian and weighted nonlinear least squares fitter[17] were used to perform these calculations. Since the eight transitions available as input to the fitter for each of the three bands form a very small data set, and since each band has the same upper state, a separate determination of the A state constants was performed first. This was done by fitting the $A^2\Sigma^+ (0,0^0,0) - X^2\Pi (0,0^1,0)$ data of Bolman, et al.,[6] while allowing all the necessary upper and ground state constants to vary.[18] The upper state constants thus obtained were held fixed for the subsequent calculations. The data from the LEF spectra were then fitted to determine the ground state constants G", A", and B". For these calculations, the lambda doubling parameters and centrifugal distortion constant, D", could not be determined from the data due to the moderate resolution of our apparatus. The lambda doubling parameters were set equal to zero while a value of $D" = 3 \times 10^{-7}\ cm^{-1}$, the approximate value

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[18] A.J. Kotlar, unpublished results.

TABLE 1. WAVELENGTHS (Å) AND ASSIGNMENTS OF THE $(0,0^1,0)$, $(1,0^1,0)$, AND $(0,0^1,1)$ EMISSION BANDS[a]

$(0,0^1,0)$	$(1,0^1,0)$	$(0,0^1,1)$	Assignments
4408.99	4669.42	4815.46	$^OP_{12}$ head
4408.17	4668.92		$^OP_{12}33$
4403.31	4664.05	4810.50	$P_2 + {}^PQ_{12}$ head
	4663.51	4809.57	$P_2 32$
4398.36	4657.94[b]	4803.89	$Q_2 31$
4393.75	4653.06	4798.47	$R_2 30$
4388.92	4648.56	4792.02	P_1 head
			$P_1 31$
4384.69	4643.90	4787.97	$Q_1 + {}^QP_{21}$ head
4383.98	4642.92	4786.29	$Q_1 31$
4379.18	4637.75	4780.78	$R_1 30$
4374.94	4632.69	4775.46	$^SR_{21}29$

---

a - Table entries between two assignments indicate overlapped lines.

b - Argon laser wavelength from Ref. 15.

P_2, and $^OP_{12}$ branch prominent lines. All of the expected eight prominent lines from N'=31 and four associated bandheads appear in each of the $^2\Sigma^+ - {}^2\Pi$ vibrational bands (except for the one obscured by laser scatter). Note the strikingly similar features of these spectra. For example, in all three bands, $R_1 30$ is about two times stronger than $R_2 30$ whereas $Q_1 31$ and $Q_2 31$ are of similar intensities. In the $(1,0^1,0)$ band shown in Figure 2, the shoulder at 4632.69Å has been assigned as the $^SR_{21}29$ instead of the larger peak at 4633.17Å, based on rotational line spacing considerations. This is the only assignment which gives a reasonable fit in our rotational analysis (vide infra).

In our previous work, the region of the laser line at 4658Å was investigated in order to identify the rotational transition pumped by this line. Although evidence suggested that the Q_2 branch line is pumped, the assignment was not firm because of interferences from grating ghosts and C_2

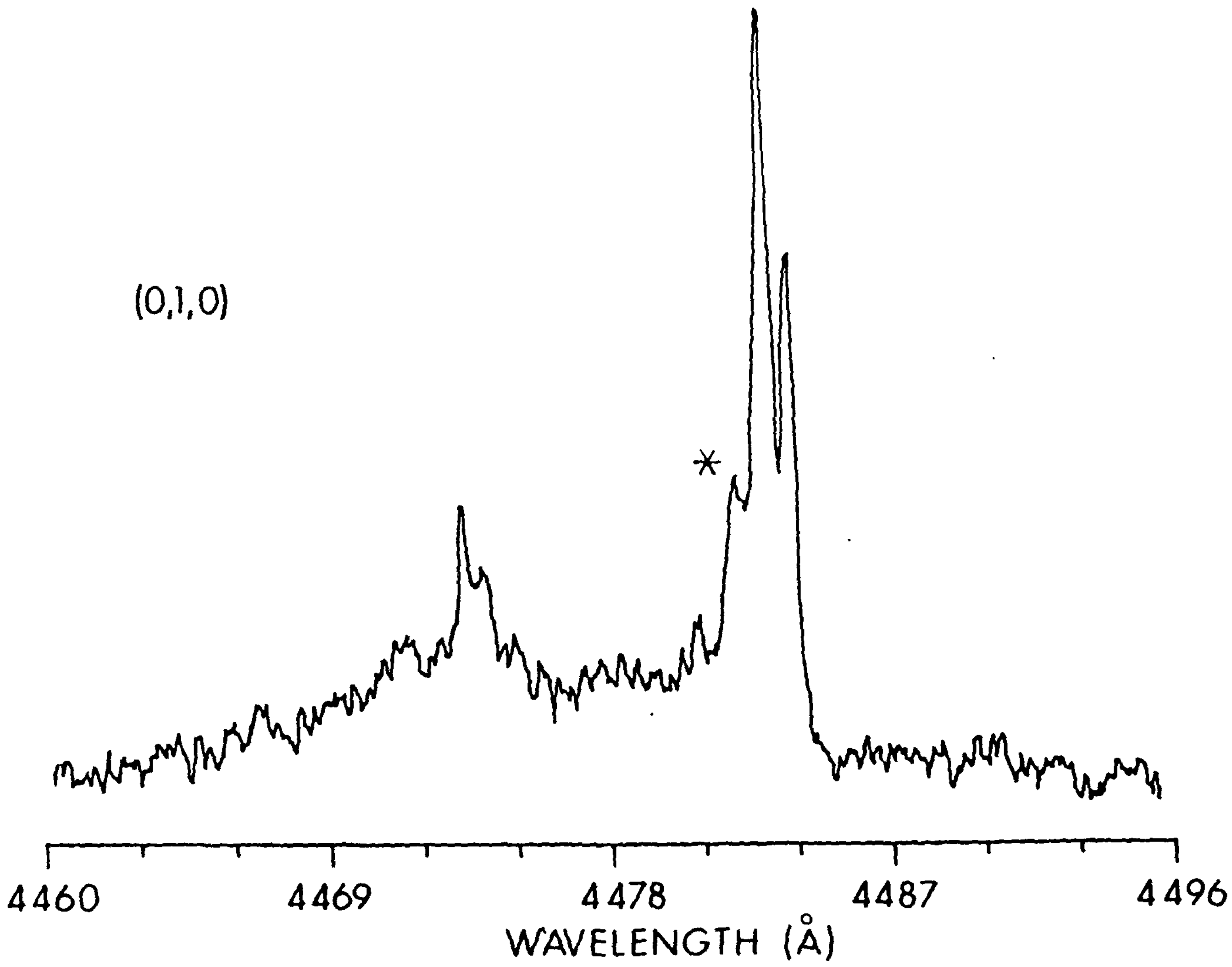

Figure 4. The $A^2\Sigma^+ (0,0^0,0) \rightarrow X^2\Sigma^+ (0,1^0,0)$ Fluorescence Spectrum

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