CHEMICALLY-PRODUCED N_2(A) TO NO(X)
ENERGY TRANSFER IN A SUPersonic FLOW

Y. D. Jones

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Final Report

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YOLANDA D. JONES, Ph.D.
Project Officer

GERALD A. HASEN
Maj, USAF
Ch, Advanced Chemical Laser Branch

HARRO-ACKERMANN
Lt Col, USAF
Ch, Laser Science & Technology Office

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**Title:** CHEMICALLY-PRODUCED N$_2$(A) TO NO(X) ENERGY TRANSFER IN A SUPersonic FLOW

**Abstract:**
A supersonic nozzle has been used to produce N$_2$(A) by the reaction of NF$_2$ and H. The NO has been added to the flow via a hypersonic wedge. Excited state NO has been observed from energy transfer. The importance of showing chemically-produced N$_2$(A) transfer of energy to NO is for possible use as a purely chemical ultraviolet laser. The densities observed here are not sufficiently high for use as a laser; however, these experiments indicate that in principle the transfer occurs with good efficiency in a chemical system.

**Subject Terms:** Nitrogen fluoride, Excited nitrogen, Metastable nitrogen, Energy transfer, Chemical laser

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83 APR edition may be used until exhausted
All other editions are obsolete.
PREFACE

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INTRODUCTION

The $\text{N}_2\text{F}_4 + \text{H}_2$ scheme for production of $\text{NF}(a^1\Delta)$, $\text{NF}(b^1\Sigma)$ and $\text{N}_2(A^3\Sigma^+)$ is well-known from flow tube studies at low pressure and low temperature (Refs. 1-4). The $\text{N}_2(\text{A})$ is a good energy storage molecule with 6.2 eV of energy and a 2.0 s lifetime (Ref. 5). Because of the long lifetime of the species, $\text{N}_2(\text{A})$ does not make a good laser candidate. Transfer of the energy in the $\text{N}_2(\text{A})$ molecule to NO is well-documented (Refs. 6-8). The transfer is efficient, but has been demonstrated only in small-scale systems such as flow tubes and with nonchemically produced $\text{N}_2(\text{A})$. This set of experiments was designed to demonstrate efficient NO excitation by $\text{N}_2(\text{A})$ in a purely chemical system as produced via the following reactions:

$$\begin{align*}
\text{N}_2\text{F}_4 & \rightarrow \text{NF}_2 \quad (1) \\
\text{F} + \text{H}_2 & \rightarrow \text{H} + \text{HF} \quad (2) \\
\text{H} + \text{NF}_2 & \rightarrow \text{NF}(a^1\Delta) + \text{HF} \quad (3) \\
\text{H} + \text{NF}(a^1\Delta) & \rightarrow \text{N}^2(\text{D}) + \text{HF} \quad (4) \\
\text{N}^2(\text{D}) + \text{NF}(a^1\Delta) & \rightarrow \text{N}_2(\text{B}) + \text{F} \quad (5) \\
\text{N}_2(\text{B}) & \rightarrow \text{N}_2(\text{A}) + \text{h} \quad (6) \\
\text{N}_2(\text{A}) + \text{NO} & \rightarrow \text{N}_2 + \text{NO}(\text{A}) \quad (7)
\end{align*}$$

Although, the $\text{N}_2(\text{A})$ to NO transfer is efficient, when NO is added to the excited nitrogen system, the kinetics become complicated. A basic set of the $\text{N}_2(\text{A}) + \text{NO}$ reactions and rates is given in the Appendix. The transition of interest in NO is the $\text{A} \rightarrow \text{X}$. The NO($A^2\Sigma^+$) state has a lifetime of $2.0 \times 10^{-7}$ s (Ref. 9). The Franck-Condon factor for the NO($A^2\Sigma^+, v' = 0$) to NO ($X^2\Pi, v' = 1$) transition is the largest at 0.26 (Ref. 10). The NO(A-X) spectrum is complex with 8 branch bands (Refs. 11 and 12). The A-state rotational constant is
1.9965 and the Boltzmann rotational distribution is centered about $J = 7$ (Ref. 13). The stimulated emission cross section can be calculated as $1.03 \times 10^{-16}$ cm$^2$. The NO molecule is a promising laser candidate and has been lased by optical pumping (Ref. 14).

Early work by Setser (Ref. 15) and Collear (Ref. 16) showed that the $N_2(A^3 \Sigma, v' = 1)$ to NO ($X^2 \Pi, v'' = 0$) transfer resulted in NO($A^2 \Sigma$) in the ratio of $v = 1$ to $v = 0$ of 1:10 (Ref. 15) and 1:2 (Ref. 16). This is the dominant process since $N_2(A)$ in $v > 1$ rapidly relaxes by V-V processes with $N_2(X)$ (Ref. 15). The most desirable NO($A-X$) transitions for lasing are the strong emission lines of $v' = 0$ to $v'' = 1, 2$. 
DEVICE DESCRIPTION

OVERVIEW

The overall system consisted of a 316L stainless steel chamber with viewing ports on four sides and has been previously described (Ref. 17). Figure 1 shows a top view of the chamber with positions shown for the gas input plumbing. The chamber was exhausted into a cooled diffuser in the transition section and two heat exchangers. The device was evacuated using two Kinney 850 cfm pumps with two M & D Pneumatics 2700 cfm blowers for a system total of 7,100 cfm.

The BCL - 16 nozzle was positioned in the chamber wall with the gas inputs. The BCL - 16 nozzle cross section is shown in Figure 2. The BCL - 16 nozzle was developed for HF/DF laser application (Ref. 18) and studied for those same systems (Ref. 19). For the N$_2$F$_4$ + H$_2$ system the combustor portion of the assembly nozzle was operated as it had been designed to produce F atoms. The hydrogen or deuterium and fluorine were injected into the combustor along with helium diluent at a molar ratio of F$_2$:D$_2$:He of approximately 1:2:50.
Figure 1. System overview.
Figure 2. Cross section of the BCL-16 nozzle.
NO INJECTOR SYSTEM

The NO was delivered to the system via a hypersonic wedge constructed from aluminum (Fig. 3). The wedge was used for preliminary examination of the NO injection. The gas was fed to the wedge by two tubes which also served as supports. The tubing could be moved along the centerline ($X_c$) of the cavity. This allowed for optimization of the NO(A-X) emission by varying the injection position and the NO flow rate. The NO flow system is depicted in Fig. 4. The $N_2$ or He could be provided through the purge system. During testing only He was used as a diluent. The entire system was operated remotely. For safety, however, an NO detector (Ecolyzer, Model 412) was placed near the device.
Figure 4. NO flow system.
NF(a^1\Delta) AND NF(b^1\Sigma) DIAGNOSTICS

The NF(a^1\Delta) diagnostic was an important part of the reaction analysis. The NF(a) and NF(b) diagnostics have been described (Ref. 20). The 874.2 nm emission from the NF(a-x) transition was detected via a 38.1 cm long spatial filter with 0.17 cm dia orifices coupled to a fused silica fiber optic. The fiber optic was bifurcated so that one end was fed into the NF(b) diagnostic. This allowed detection of NF(a) and NF(b) to be made within the same viewing volume. The diagnostic as applied to the device is shown in Fig. 5. The actual width of the flame was used to determine the volume viewed by the diagnostic. The NF(a) emission was filtered using an extremely narrow bandpass filter (FWHM 0.98 nm) centered at 874.29 nm which essentially eliminated interferences from the close-lying N\(_2\)(B) and H\(_2\)(v=3) emissions.

The NF(b) diagnostic used the same bifurcated fiber optic with the output of the other porting of the cable going to a narrow bandpass filter centered at 531.4 nm and FWHM of 9.8 nm. The 538.8 nm emission of the NF (b^1\Sigma - X^3\Sigma) transition was not masked by any near-lying emissions. The spatial filter was mounted on a remotely operated translation stage with a linear voltage displacement transducer (LVDT) to accomplish scans across the flow field of the device with a known position. Sample scans of the NF(a^1\Delta) and NF(b^1\Sigma) emissions are shown in Figs. 6 and 7.
Figure 5. Diagnostic set-up.

- Diffuser
- CaF$_2$ window
- Flow
- Bifurcated fiber optic connected to PMT's
- Origin
- V$_{TOTAL} = V_1 + V_2$
- $D = 2r_0$
OPTICAL MULTICHANNEL ANALYZER (OMA)

The OMA III 1460R system (EG&G PAR) was used to monitor the change in emission over a wide wavelength range (usually 300-900 nm) at a fixed point within the device. The OMA III system consisted of a nonintensified diode array head (Model 1412) coupled to a Model 1233 polychromator.
N\textsubscript{2}(A) TO NO TRANSFER STUDIES

The system N\textsubscript{2}(A) production was optimized and has been described previously (Ref. 21). Maximum N\textsubscript{2}(A) production was found to occur when D\textsubscript{2} was used in place of H\textsubscript{2} due to better penetration of the higher molecular weight gas and fewer loss mechanisms for the NF(a\textsuperscript{1}A) (Ref. 22). The two effects cannot, at this time, be decoupled because of only preliminary flow studies. The N\textsubscript{2}(A) was monitored by the visible N\textsubscript{2}(B) emission in the flow. The N\textsubscript{2}(B) relaxes to N\textsubscript{2}(A) with the emission of a photon. The N\textsubscript{2}(A) may also be produced directly. Therefore, the N\textsubscript{2}(b) gave a lower limit to the actual N\textsubscript{2}(A) production. The additional N\textsubscript{2}(A) contribution was evaluated by examination of the N\textsubscript{2}(c) population in previous experiments (Ref. 23). The N\textsubscript{2}(b) was determined to be accurate to within a factor of 10. Without the hypersonic wedge in place, maximum N\textsubscript{2}(B) was about 2 \times 10^{11} molecules/cm\textsuperscript{3}. The wedge interfered slightly with the flow field and, thus, mixing of the NF\textsubscript{2} and H\textsubscript{2} streams. The maximum N\textsubscript{2}(B) detected with the wedge in place was about 1 \times 10^{11} molecules/cm\textsuperscript{3}. Variation of the injection point along the X\textsubscript{C} was first performed. Figure 8 shows the variation of NO(A-X) emission (or γ-bands) with wedge position. The distance measured was from the backside of the wedge which had a width of 0.599 cm. The maximum NO(A-X) emission was at 1 to 2 cm from the nozzle exit plane (NEP).

Flow rate variation is shown in Figs. 9 and 10. Figure 9 shows the NO(A-X) emission as a function of flow rate of NO. The NO emission decreased with increased NO flow rate. This is probably due to disturbance of the mixing of primary, secondary and trip jets by large flow rate injection. The flow rate range was limited by the flow control system. The NO can react with N(2\textsuperscript{D}) rapidly (Ref. 23), \( k = 7.0 \times 10^{11} \text{ cm}^3/\text{molecules-s} \), providing a competitive pathway for N\textsubscript{2}(A) formation. There is a definite decrease in N\textsubscript{2}(B) with increasing NO flow rate (Fig. 10). The decrease is most probably due to a combination of these effects in addition to self-quenching. The effect of He diluent is demonstrated by the data on the graph in Fig. 9 at 0.181 g/s NO. The He effectively dilutes the flow and does not aid mixing.
Figure 8. NO emission with variation in injector position.
Figure 9  NO(A) emission as a function of NO flow rate.
Figure 10. Variation of $N_2(B)$ emission as a function of NO flow.
A minimum number of runs were performed with trip jet injection of the D$_2$ and secondary injection of the N$_2$F$_4$. These were designed to determine if mixing could be improved in this configuration, providing greater number densities of NO(A). Table 1 contains information from the tests. There is some increase observed in the reversed configuration. The NO does not appear to interfere with NF(a$^1\Delta$) or NF(b$^1\gamma$) production significantly as evidenced by the high concentrations in the tests.

Table 2 contains information from several tests where data was taken with and without NO flow. For the two high NO flow rate conditions, the efficiency is 30 to 50 percent due to the creation of turbulence in the flow. At the low flow rate, using the N$_2$(B) emission yields a greater than 100 percent transfer. What is actually occurring is that not all of the N$_2$(A) is accounted for by the N$_2$(B) emission. This also implies that the NF$_2$ + D$_2$ reaction sequence forms some N$_2$(A) directly instead of all N$_2$(B) which then relaxes to N$_2$(A). This is possible since the N$_2$(A) is not detectable directly in this system because of interferences from NO emission from contaminants in the N$_2$F$_4$.

A typical OMA III spectra with NO is shown in Fig. 11. The NO(A-X, $V' = 0$) and NO(A-X, $V' = 1$) peaks were identified. Specifically, the (0-1), (0-2), (0-3), (0-4), (0-5), (0-6), (1-6), (1-7) and (1-8) peaks were detected. Possible B-band peaks are also identified in the figure. Figure 12 was taken on the same test with the NO flow turned off and is included for comparison. The OMA III scans shown are not corrected for wavelength response of the detector.
### TABLE 1. D₂ Trip Jet Injection.

<table>
<thead>
<tr>
<th>Wedge (at 0 position)</th>
<th>Primary F₂ in He³ (25%)</th>
<th>Combustor D₂⁺</th>
<th>Trip D₂⁺</th>
<th>Secondary He⁺ N₂F₄⁺</th>
<th>N₂(B) x10¹⁰</th>
<th>NO(A) x10¹⁰</th>
<th>NF(a¹Δ) x10¹⁵</th>
<th>NF(b¹Σ) x10¹²</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.20</td>
<td>0.14</td>
<td>0.006</td>
<td>0.02</td>
<td>0.12 0.33</td>
<td>0.40</td>
<td>-</td>
<td>neg1.</td>
<td>6.1 4.10</td>
</tr>
<tr>
<td>0.22</td>
<td>0.13</td>
<td>0.006</td>
<td>0.02</td>
<td>0.12 0.33</td>
<td>1.80</td>
<td>3.08</td>
<td>3.2 4.25</td>
<td></td>
</tr>
<tr>
<td>0.22</td>
<td>0.13</td>
<td>0.006</td>
<td>0.02</td>
<td>0        0.33</td>
<td>1.80</td>
<td>8.13</td>
<td>3.2 4.18</td>
<td></td>
</tr>
<tr>
<td>0.28</td>
<td>0.14</td>
<td>0.006</td>
<td>0.02</td>
<td>0.12 0.32</td>
<td>1.54</td>
<td>7.31</td>
<td>3.2 3.92</td>
<td></td>
</tr>
<tr>
<td>0.34</td>
<td>0.13</td>
<td>0.006</td>
<td>0.02</td>
<td>0.12 0.31</td>
<td>1.30</td>
<td>6.58</td>
<td>3.2 3.50</td>
<td></td>
</tr>
<tr>
<td>0.34⁺</td>
<td>0.13</td>
<td>0.006</td>
<td>0.02</td>
<td>0.12 0.28</td>
<td>1.39</td>
<td>6.25</td>
<td>2.0 3.34</td>
<td></td>
</tr>
<tr>
<td>0.34⁺</td>
<td>0.13</td>
<td>0.006</td>
<td>0.02</td>
<td>0        0.28</td>
<td>1.40</td>
<td>6.32</td>
<td>2.0 3.36</td>
<td></td>
</tr>
</tbody>
</table>

*aAll flow rates in g/s.

bWedge at 0.64 cm position.
TABLE 2. $N_2$(A) to NO Transfer Data.

<table>
<thead>
<tr>
<th>Wedge</th>
<th>Primary</th>
<th>Trip</th>
<th>Secondary</th>
<th>(Molecules/cm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO$^a$ (at 0 position)</td>
<td>$F_2$ in He$^a$</td>
<td>$D_2^a$</td>
<td>He$^a$</td>
<td>$N_2F_4^a$</td>
</tr>
<tr>
<td>0.183</td>
<td>0.12</td>
<td>0.007</td>
<td>0.02</td>
<td>0.03</td>
</tr>
<tr>
<td>0</td>
<td>0.12</td>
<td>0.007</td>
<td>0.02</td>
<td>0.03</td>
</tr>
<tr>
<td>0.238</td>
<td>0.13</td>
<td>0.007</td>
<td>0.02</td>
<td>0.03</td>
</tr>
<tr>
<td>0</td>
<td>0.13</td>
<td>0.007</td>
<td>0.02</td>
<td>0.03</td>
</tr>
<tr>
<td>0.33</td>
<td>0.12</td>
<td>0.007</td>
<td>0.02</td>
<td>0.03</td>
</tr>
<tr>
<td>0</td>
<td>0.12</td>
<td>0.007</td>
<td>0.02</td>
<td>0.03</td>
</tr>
</tbody>
</table>

$^a$All flow rates in g/s.
Figure 11. OMA III scan with NO flow on.
Figure 12. OMA III scan without NO flow.
Efficient $N_2(A^3\Sigma) \rightarrow NO(X^2\Pi, V^\ast = 0)$ transfer has been demonstrated using a purely chemical production method for $N_2(A^3\Sigma)$. The $N_2(A)$ production system is complex and is not completely understood. Improvements in nozzle design could increase the $N_2(A)$ production which would allow for greater NO(A) densities. Improvements in the $N_2(A)$ production have been discussed in Reference 21. The maximum production level of NO(A) in these experiments was about $10^{11}$ molecules/cm$^3$. Levels on the order of $10^{14}$ are required to demonstrate gain over the short path length provided by this nozzle and system. Therefore, for nozzle development not only increased $N_2(A)$ production should be examined, but also gain path length. The $N_2(A)$ to NO(X) energy transfer system is promising for development into a chemical laser system.
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APPENDIX

N₂(A) to NO(X)

KINETICS
TABLE A-1. Kinetic Rates.

1. $N_2(A) + NO(X) + N_2(X) + NO(A)$ \quad $k_1 = 1.5 \times 10^{-10}$ (Ref 1)
2. $NO(A) + NO(X) + NO(X) + NO(X)$ \quad $k_2 = 1.7 \times 10^{-10}$ (Ref 2)
3. $NO(A) + N_2(X) + NO(X) + N_2(X)$ \quad $k_3 = 1.1 \times 10^{13}$ (Ref 2)
4. $NO(A) + N_2(B) + NO(X) + N_2(A)$ \quad $k_4 = 2.4 \times 10^{-10}$ (Ref 3)
5. $N_2(A) + NO(X) + N_2(X) + NO(X)$ \quad $k_5 = 7.0 \times 10^{-11}$ (Ref 4)
6. $N_2(A) + NO(X) + N_2(X) + NO(X)$ \quad $k_6 = 3.6 \times 10^{-11}$ (Ref 5)
7. $N_2(A) + O(3P) + N_2(X) + O(3P)$ \quad $k_7 = 2.8 \times 10^{-11}$ (Ref 6)
8. $O(1D) + N_2(X) + O(3P) + N_2(X)$ \quad $k_8 = 6.9 \times 10^{-11}$ (Ref 8)
9. $O(1S) + O(3P) + O(1D) + O(3P)$ \quad $k_9 = 1.8 \times 10^{-11}$ (Ref 9)
10. $O(1D) + NO(X) + O(3P) + NO$ \quad $k_{10} = 8.5 \times 10^{-11}$ (Ref 10)
11. $O(1S) + NO(X) + O(1D) + NO(X)$ \quad $k_{11} = 4 \times 10^{-10}$ (Ref 11)

All rates are in cm$^3$/molecule - s.
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Washington, DC 20301

US Air Force Academy
Attn: OFP
Colorado Springs, CO 80840

Naval Research Laboratory
Attn: Code 2627, Tech Lib
Washington, DC 20375

Physical Sciences Inc.
P.O. Box 3100, Research Park
Andover, MA 01810

SRI International
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Menlo Park, CA 94025

McDonnell-Douglas Astro Co
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Huntington Beach, CA 92647

Naval Postgraduate School
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Department of the Navy
Monterey, CA 93940

Nimitz Library
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U.S. Naval Academy
Annapolis, MD 21402

Rockwell Int'l, Rocketdyne Div
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Canoga Park, CA 91304

Sandia National Laboratories
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P.O. Box 5800
Albuquerque, NM 87115

TRW Electronics & Defense Sector
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One Space Park
Redondo Beach, CA 90278

United Technologies Research Corp
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400 Main Street
East Hartford, CT 06108

US Army Ballistic Missile Defense
Advanced Technical Center
P.O. Box 1500
Huntsville, AL 35807

KAMAN TEMPO
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2560 Huntington Ave, Suite 500
Alexandria, VA 22303

KAMAN TEMPO
Attn: DASIAC/DETIR, D. Reitz
P.O. Drawer QQ
Santa Barbara, CA 93102

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Attn: LSE
Maxwell AFB, AL 36112

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