



Detection of Nitric Oxide (NO) at 5.3 microm

Using Difference-Frequency Laser Spectroscopy



RICE

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About Nitric Oxide

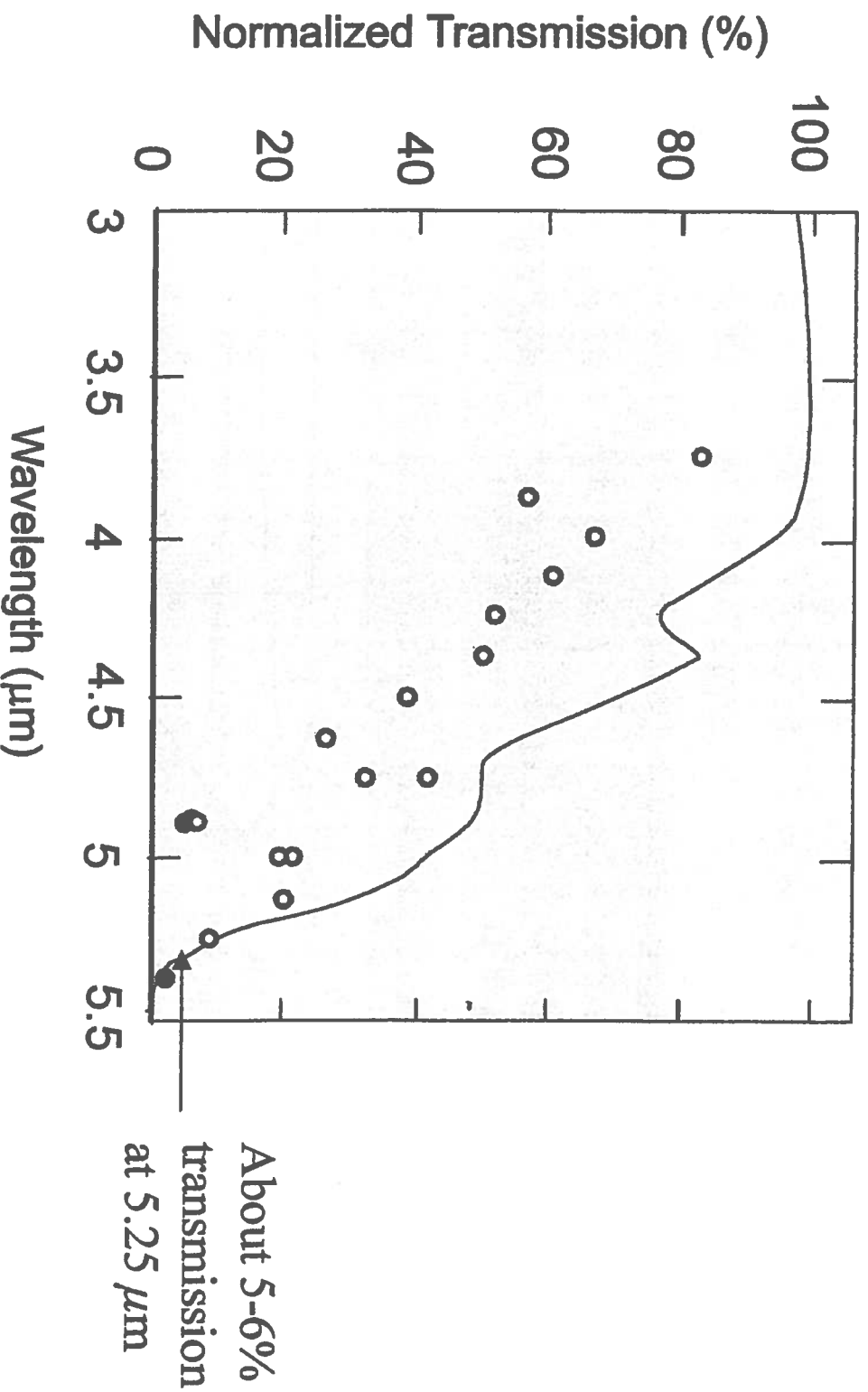
* Chemical characteristics:

- Relatively stable, no color
- Formed in many reactions involving reduction of nitric acid and solutions of nitrates and nitrites

* Biomedical and medical characteristics:

- Links between neuroscience, physiology and immunology
- Provides information how cells communicate and defend themselves
- Signal asthmatic condition, HIV infection, brain and heart activities and smoking habits
- Mediator of transition from fetal to neonatal pulmonary circulation in the case of newborn infants
- “Molecule of the year” of “SCIENCE” magazine of 1992

Measured Transmission of PPLN Crystal



Solid line: as measured by L.E.Meyers (Stanford)

Circle: current work

DFG Power Calculation with Crystal Absorption

$$P_i = \exp\left(-\frac{\alpha \cdot l}{2}\right) \cdot \frac{(16 \cdot \pi \cdot \omega_i \cdot d_{\text{eff}})^2}{c^3 \cdot n_i \cdot n_p \cdot n_s} \cdot \frac{l^2}{(W_{\text{op}}^2 + W_{\text{os}}^2)} \cdot \frac{P_p \cdot P_s}{\xi} \cdot h(\mu, \xi, \alpha)$$

Focusing point is at the center of the crystal.
 α is the power absorption coefficient for PPLN.

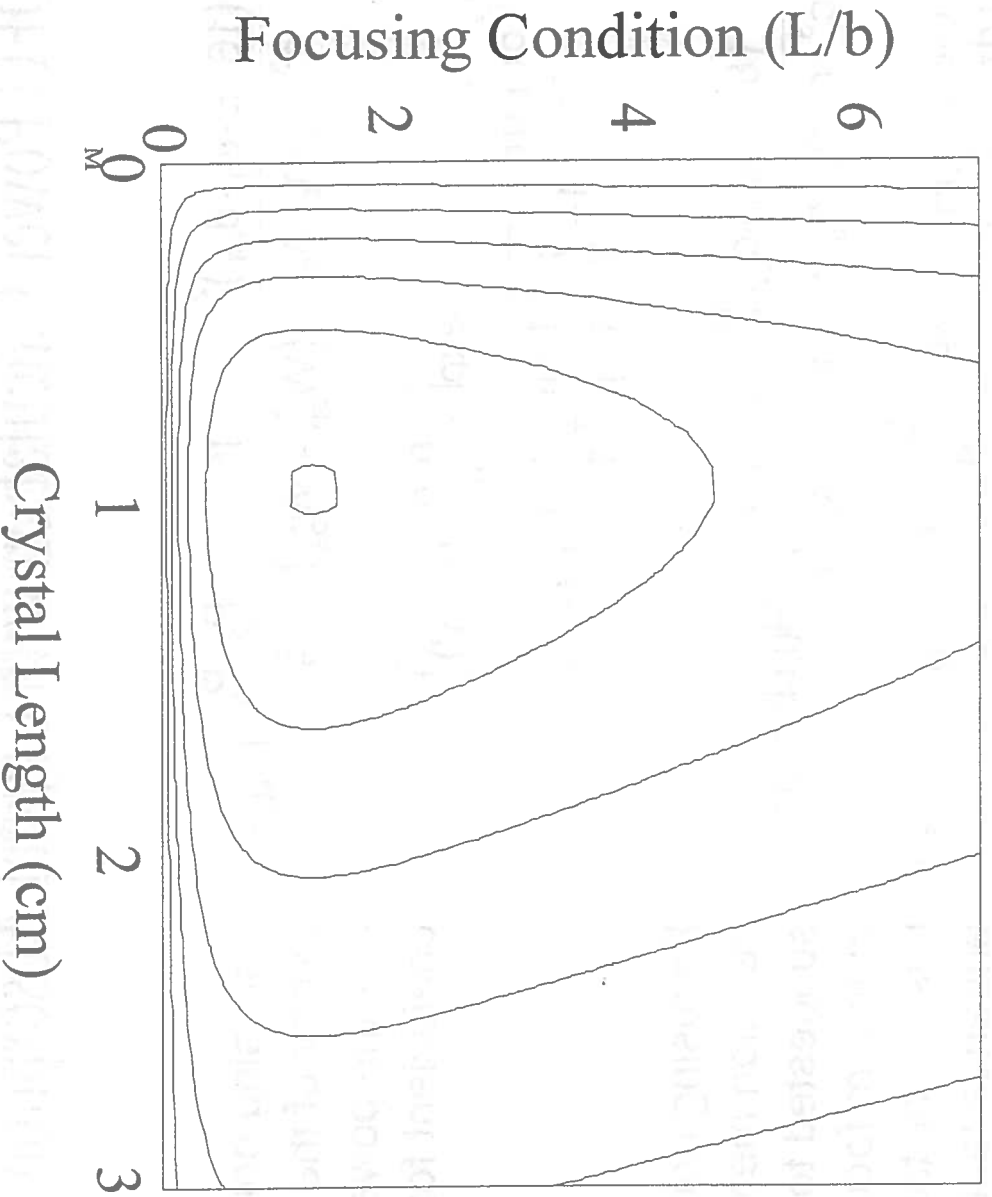
$$h(\mu, \xi, \alpha) = \frac{1}{4 \cdot \xi} \cdot \int_{-\xi}^{+\xi} d\tau \int_{-\xi}^{+\xi} d\tau' \cdot \frac{\exp\left(-\frac{b \cdot \alpha}{4} \cdot (\tau - \tau')\right)}{1 - \frac{j}{2} \cdot \left(\frac{1+\mu}{1-\mu} + \frac{1-\mu}{1+\mu}\right) \cdot (\tau - \tau') + \tau \cdot \tau'}$$

$$P_i = \exp\left(-\frac{\alpha \cdot l}{2}\right) \cdot \frac{(8 \cdot \pi \cdot \omega_i \cdot d_{\text{eff}})^2 \cdot P_p \cdot P_s \cdot b}{c^3 \cdot n_i \cdot n_p \cdot n_s \cdot \epsilon_0 \cdot (k_s^{-1} + k_p^{-1})} \cdot H(l, l_1, b, \alpha, \mu)$$

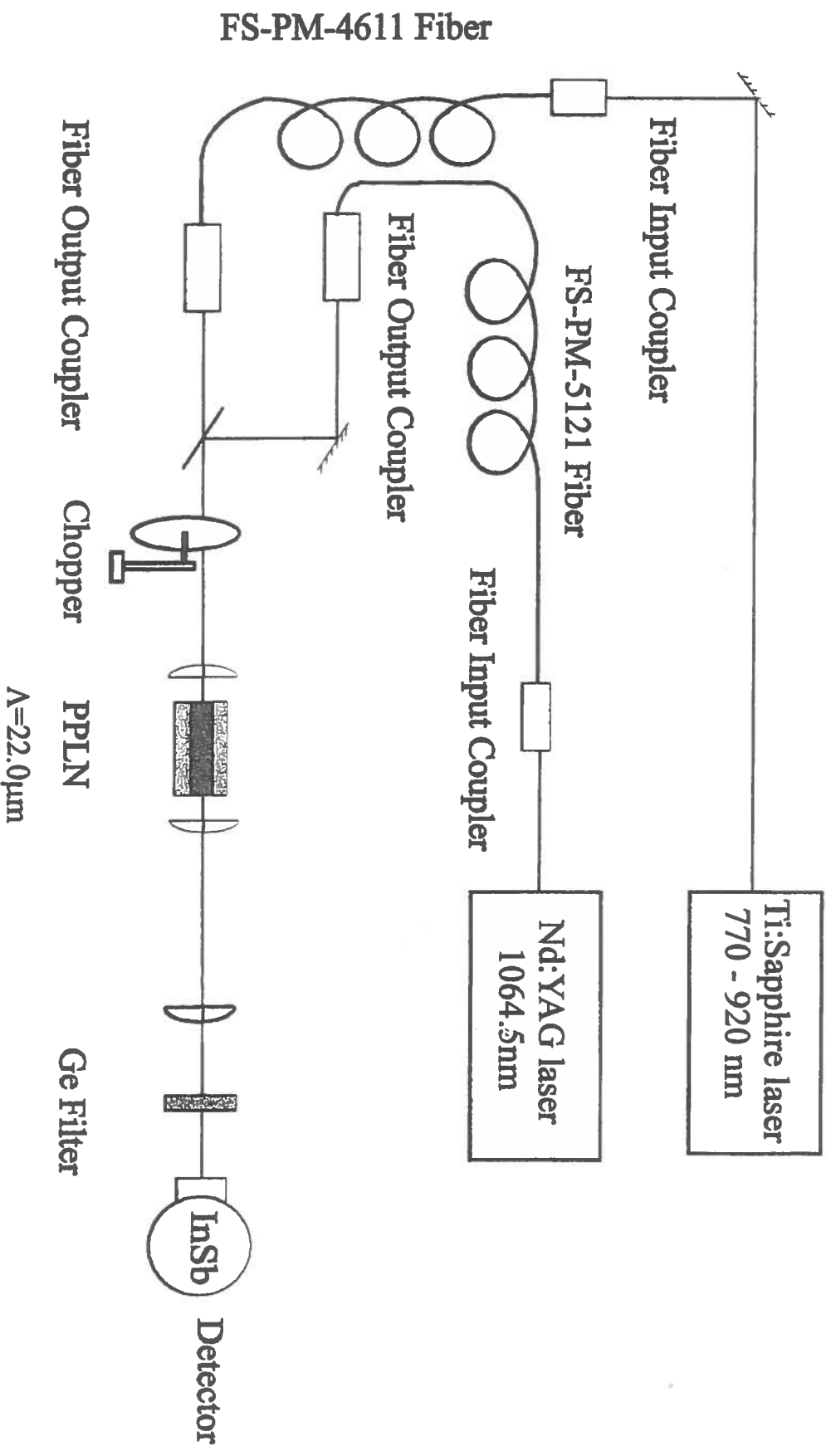
$$H(l, l_1, b, \alpha, \mu) = \int_{-2 \cdot l_1/b}^{2 \cdot (l-l_1)/b} d\tau \cdot \int_{-2 \cdot l_1/b}^{2 \cdot (l-l_1)/b} d\tau' \cdot \frac{\exp\left(-\frac{b \cdot \alpha}{4} \cdot (\tau - \tau')\right) \cdot (1 + \tau \cdot \tau')}{(1 + \tau \cdot \tau')^2 + \left(\frac{1 + \mu}{1 - \mu}\right)^2 \cdot (\tau - \tau')^2}$$

Focusing point is arbitrary.
 Iteration method is suggested to find the optimum focusing point.
 l_1 is the distance from entrance facet of the crystal to focusing point.

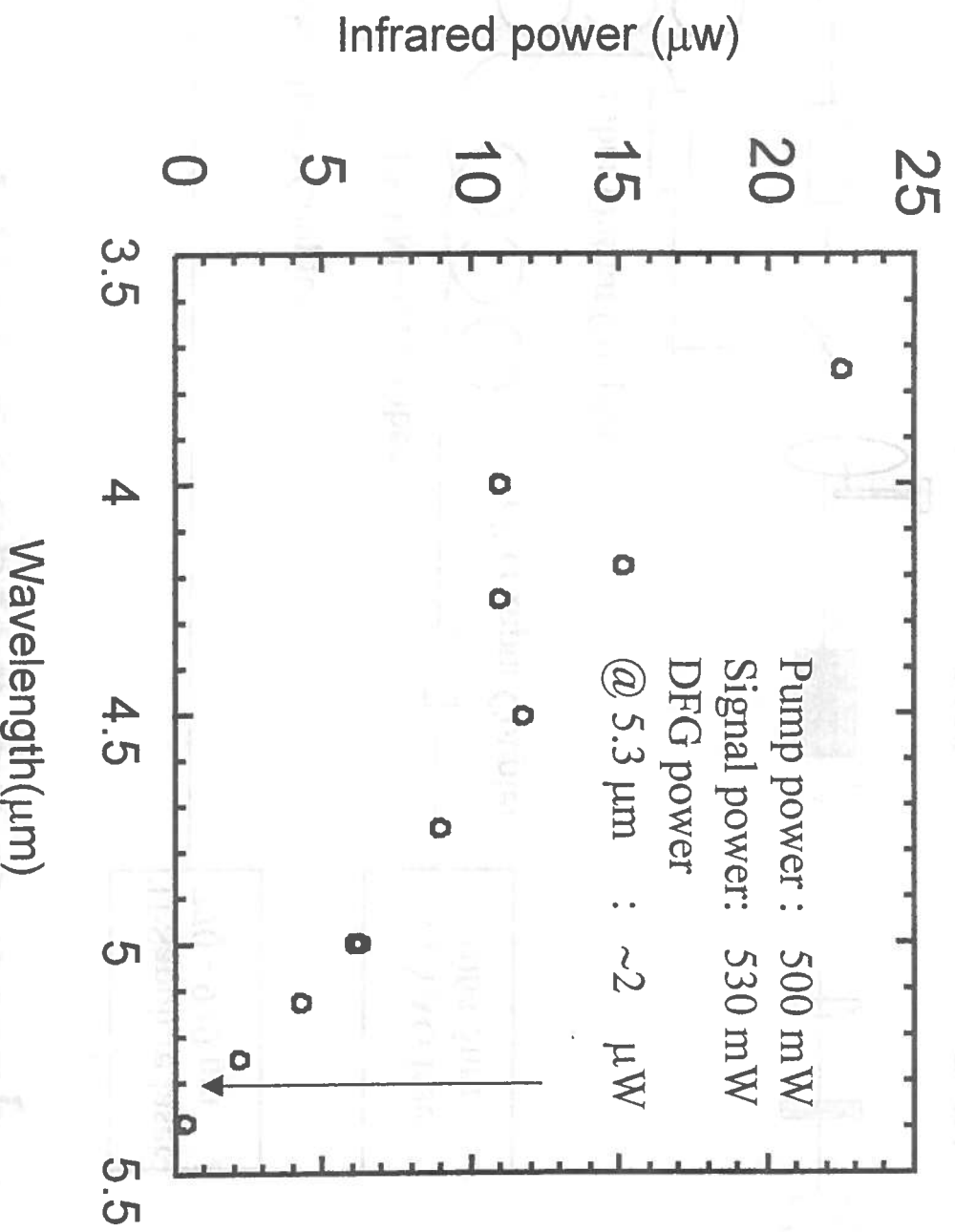
Contour Plot of DFG Power Dependence on Crystal Length



Fiber Coupled DFG Setup



Power of Mid-IR DFG Output



DFG Power (Experimental Result)

	Power	Wavelength
Pump Beam:	120 mW	886 nm
Signal Beam:	190 mW	1064.5 nm

Focusing condition: $\xi=1.3$ Degeneracy Parameter: $\mu=0.828$

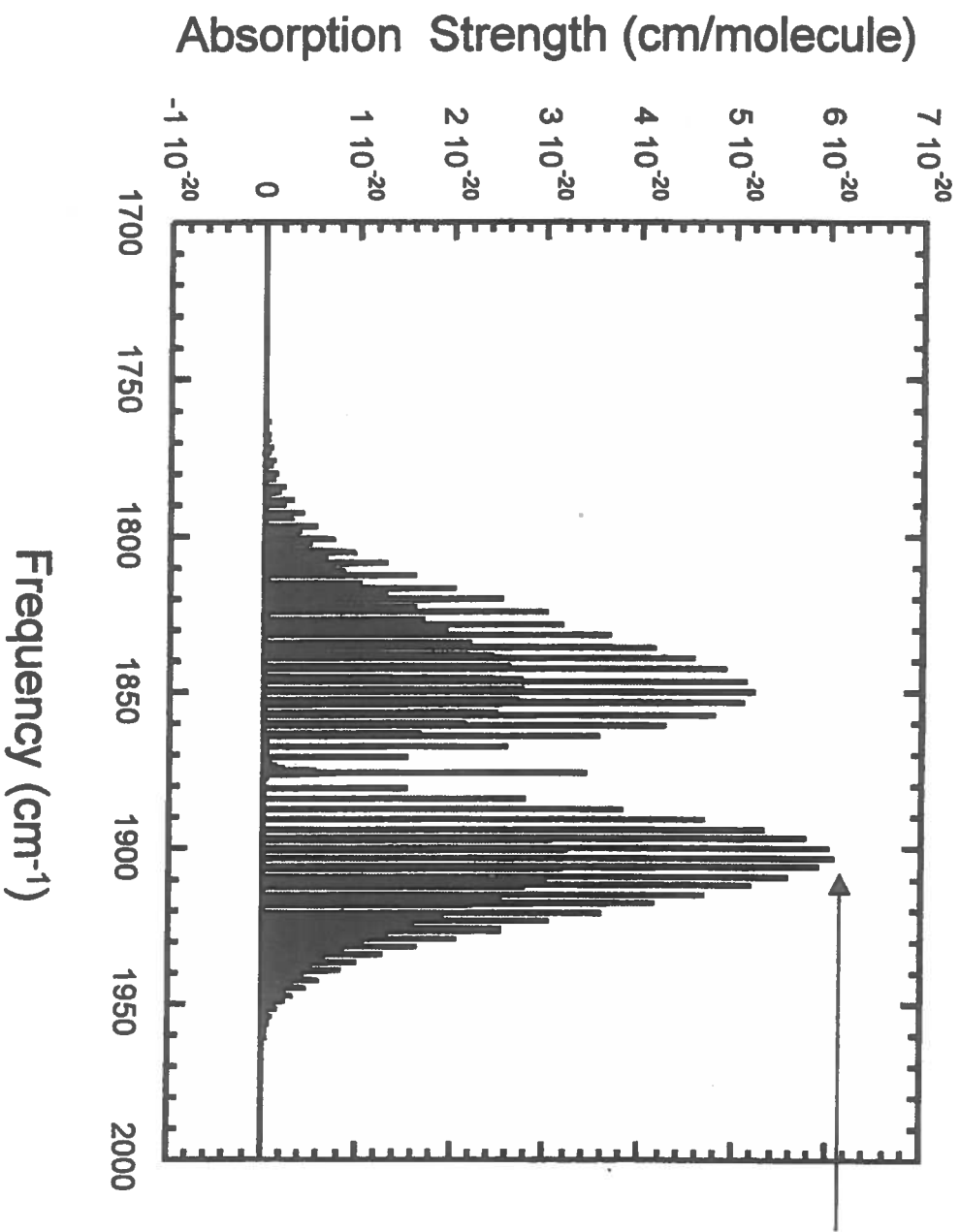
	Power	Theory
1-cm long crystal	230 nW	310 nW
2-cm long crystal	161 nW	203 nW

The above theoretical results accounts for Fresnel losses at the facets of the crystal and lens, as well as the polarization distortion caused by optical fiber.

The discrepancy is due to the mode quality after the fiber and possible beam clipping at the output of the crystal.

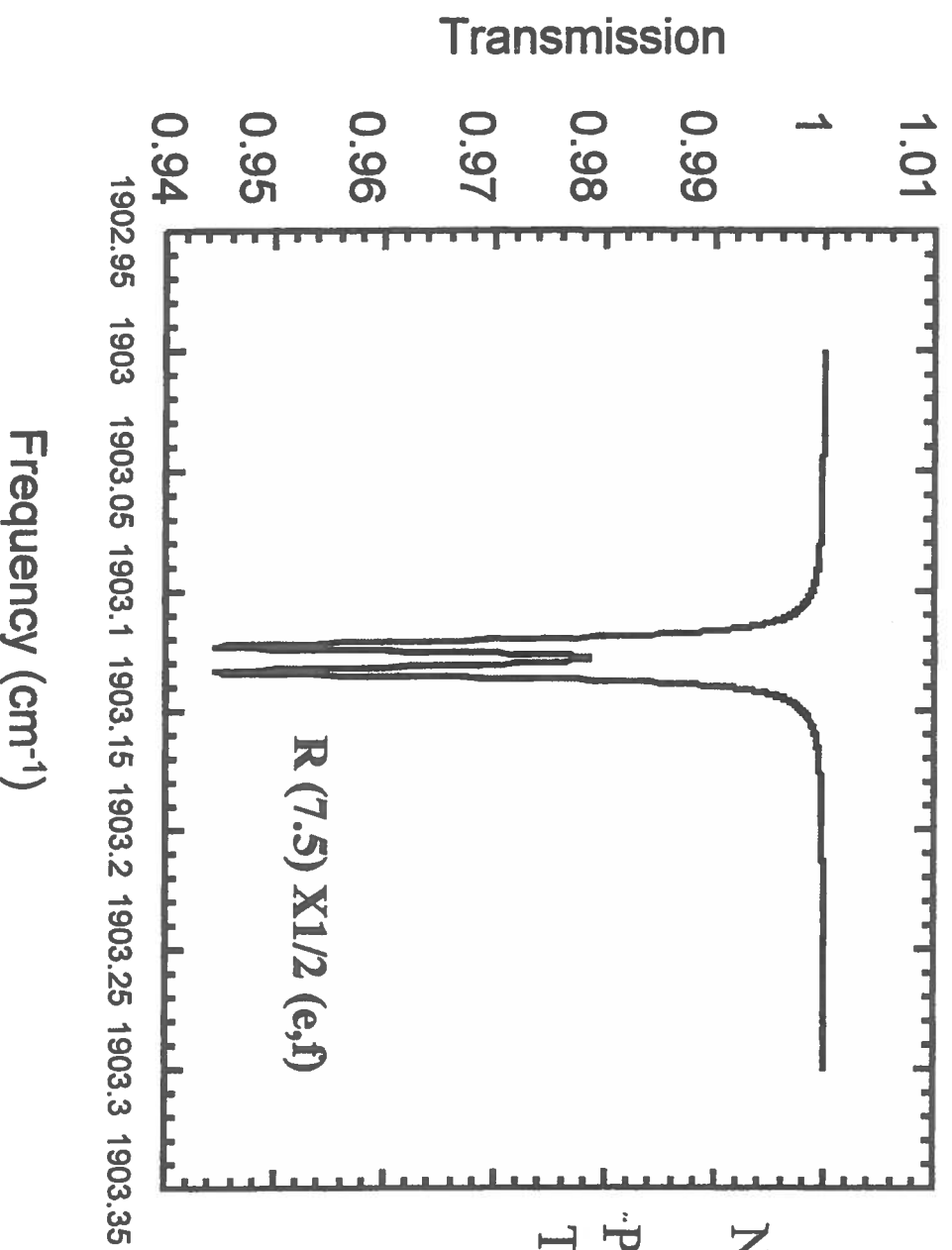
From above result, we can see that experimental result demonstrated that 1-cm long crystal is better than 2-cm long crystal in DFG and about 44% increase is achieved which is in good agreement with theoretical result.

GRISA Spectrum of Fundamental Band of Nitric Oxide



R(7.5)×1/2 (e,f)
and
R(7.5)×3/2 (e,f)
lines were
detected.

GEISA Spectrum of Nitric Oxide

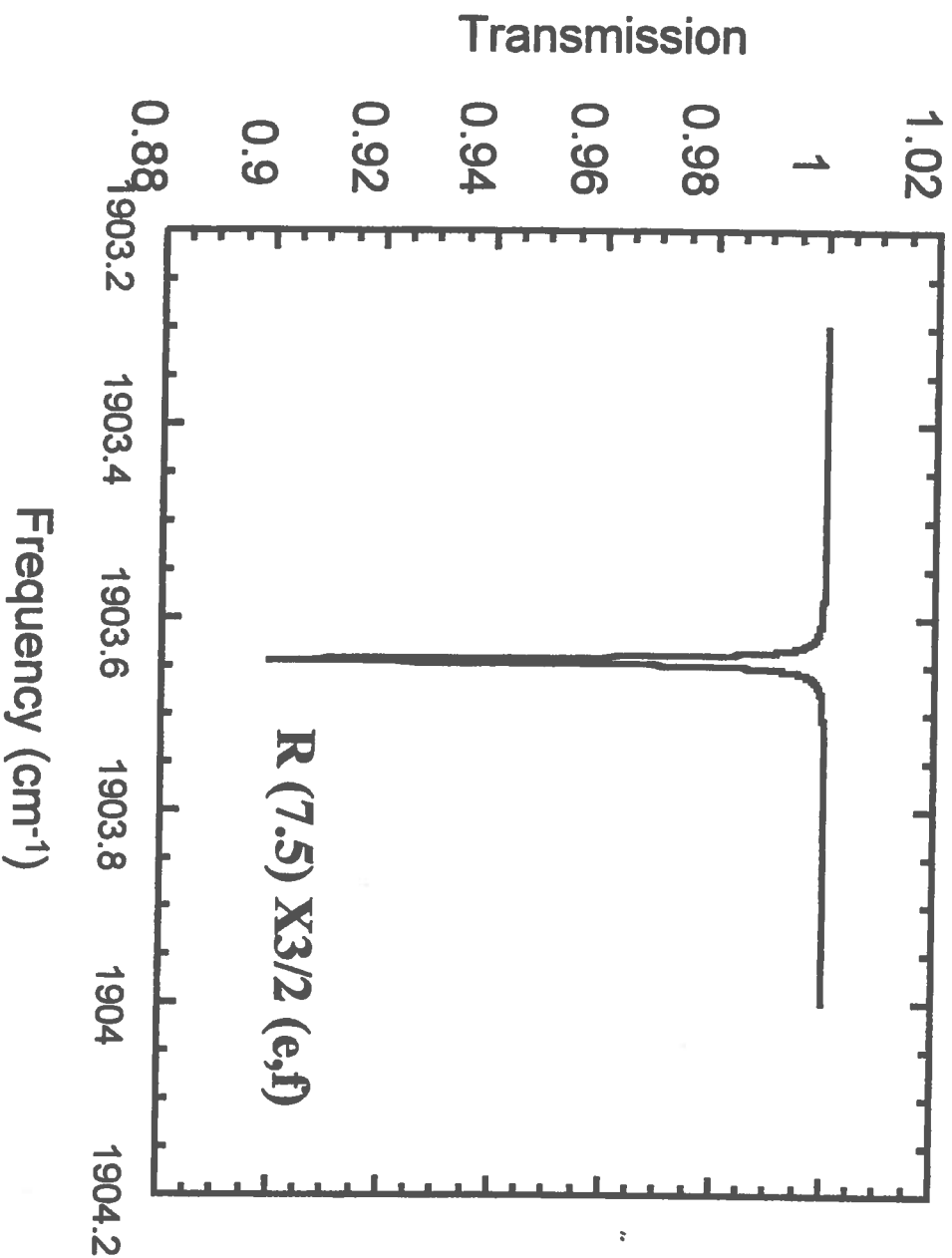


NO concentration:
2.8 ppm

Pressure: 34 torr

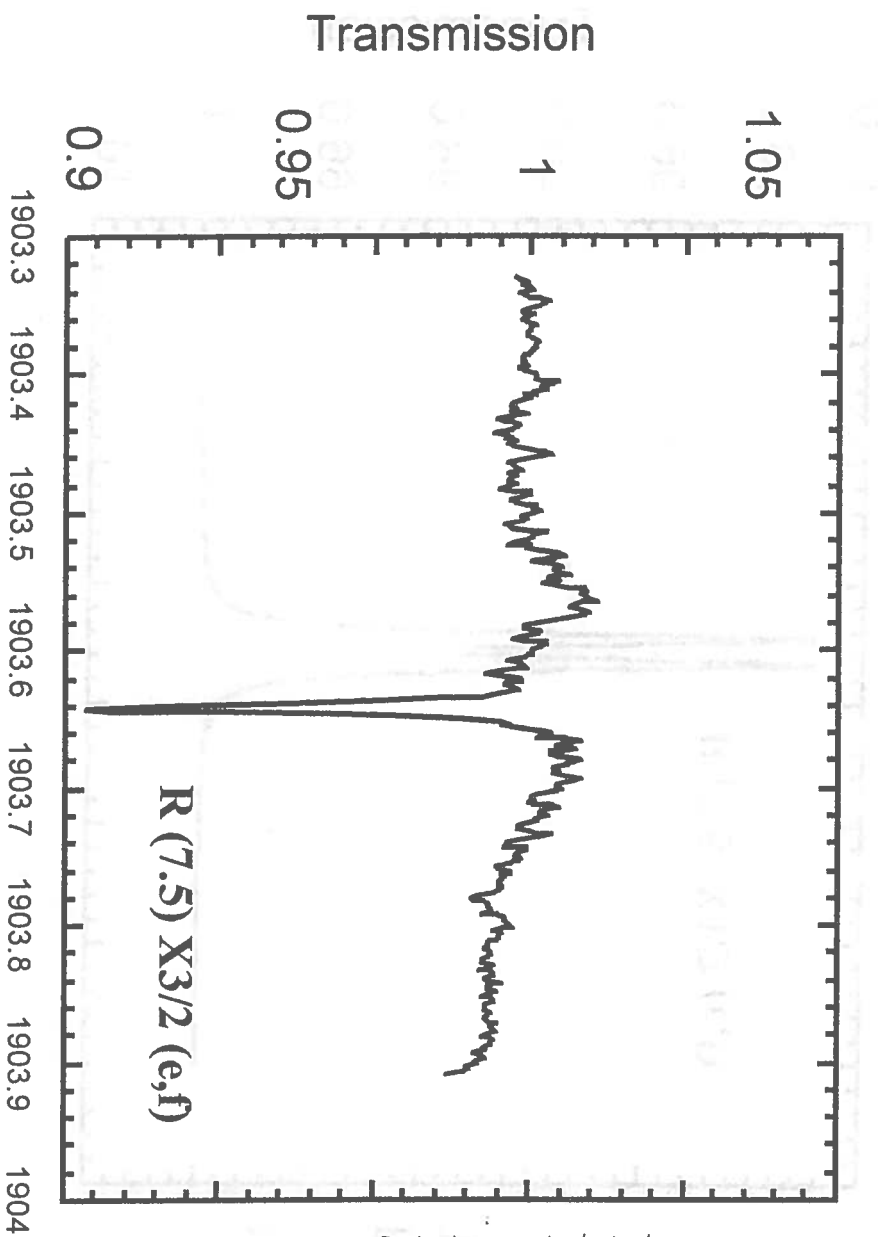
Temperature: 296 K

GEISA Spectrum of Nitric Oxide



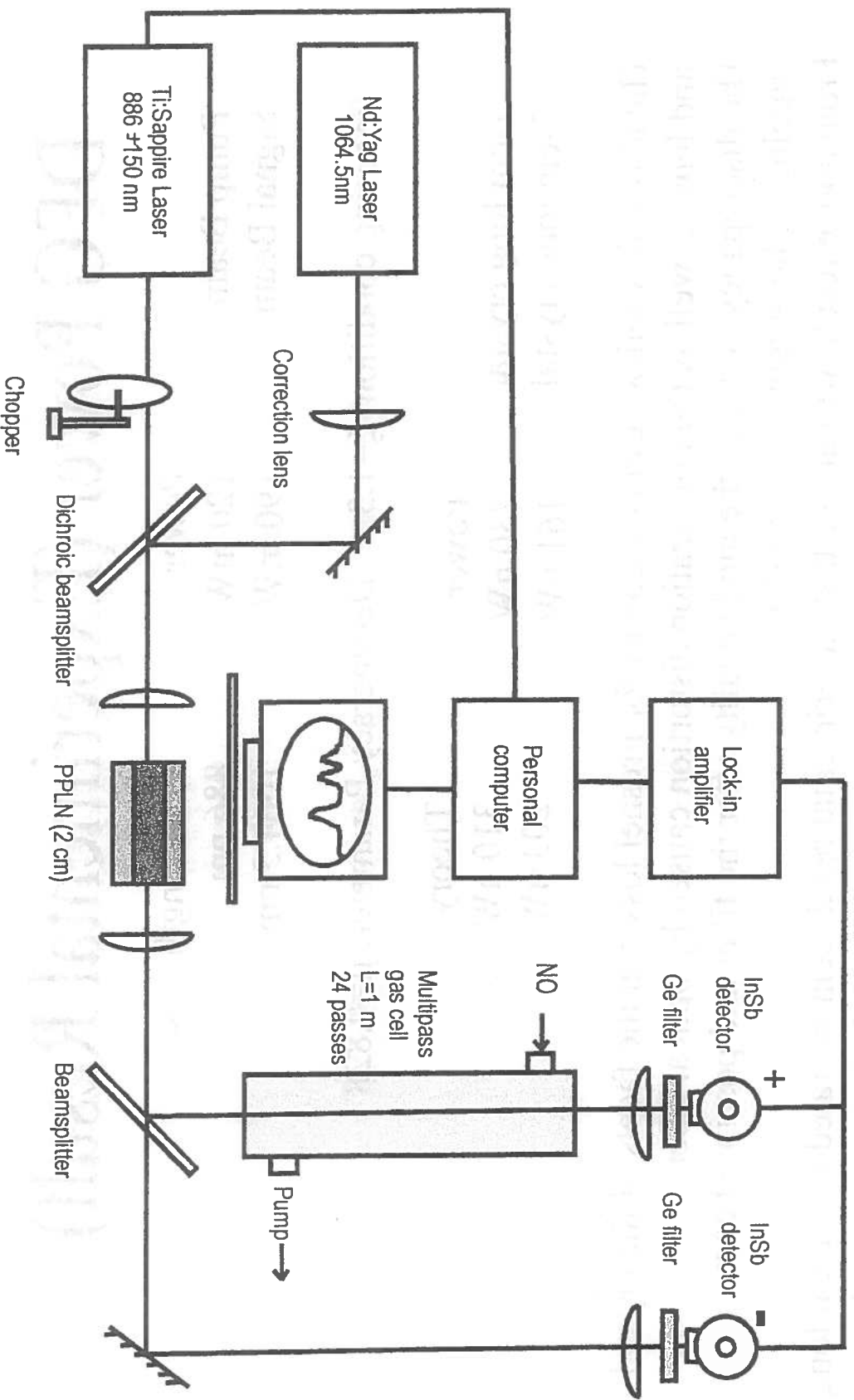
NO concentration:
5.6 ppm
Pressure: 34 torr
Temperature: 296 K

NO Spectrum at ppm Level Concentration



NO in air.
NO concentration:
5.6(\pm 0.3) ppm
Pressure: 34 torr
Temperature: 296 K

Optical Setup for the Measurement of NO by Using PPLN based DFG



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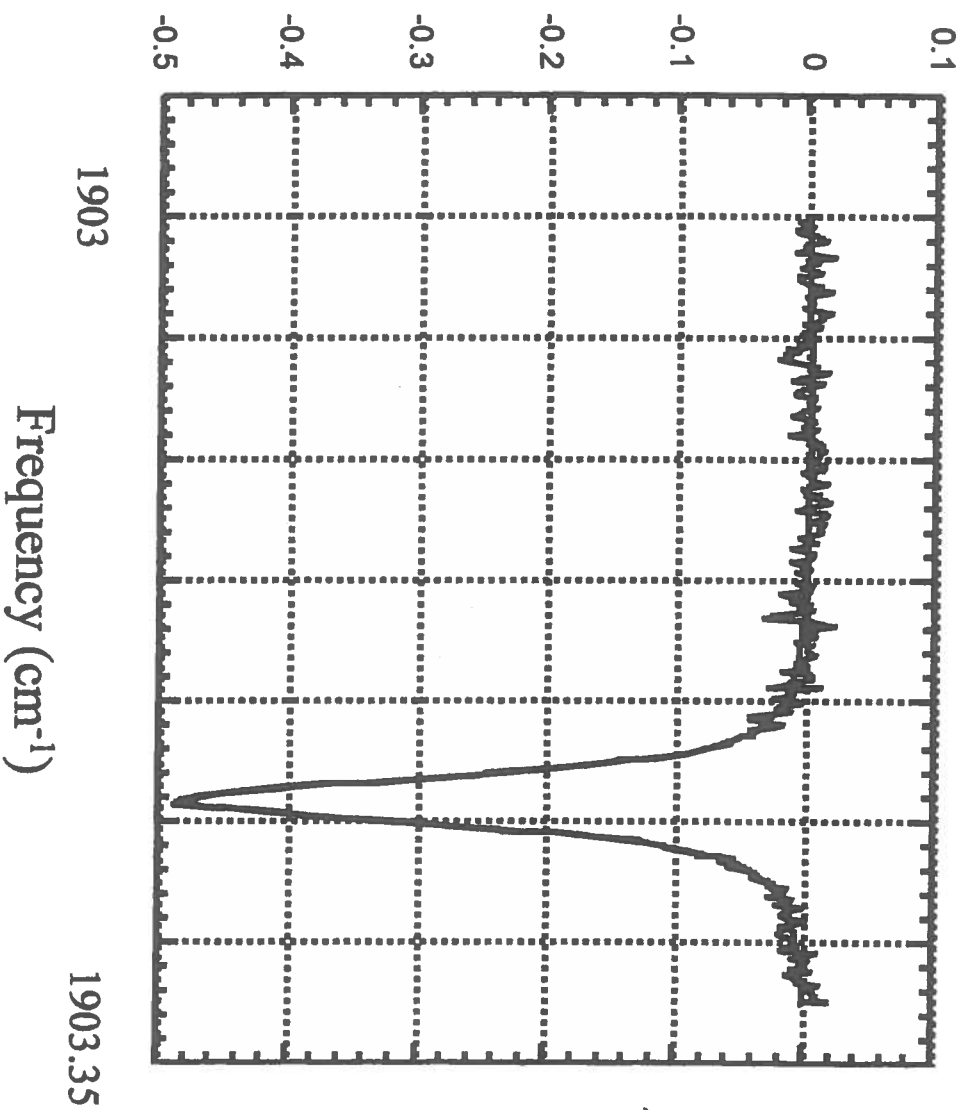
The above theoretical results accounts for Fresnel losses at the facets of the crystal and lens, as well as the polarization distortion caused by optical fiber.

The discrepancy is due to the mode quality after the fiber and possible beam clipping at the output of the crystal.

From above result, we can see that experimental result demonstrated that 1-cm long crystal is better than 2-cm long crystal in DFG and about 44% increase is achieved which is in good agreement with theoretical result.

Pressure Broadening Measurements of Nitric Oxide

Voigt fitting of NO line profile



Line:

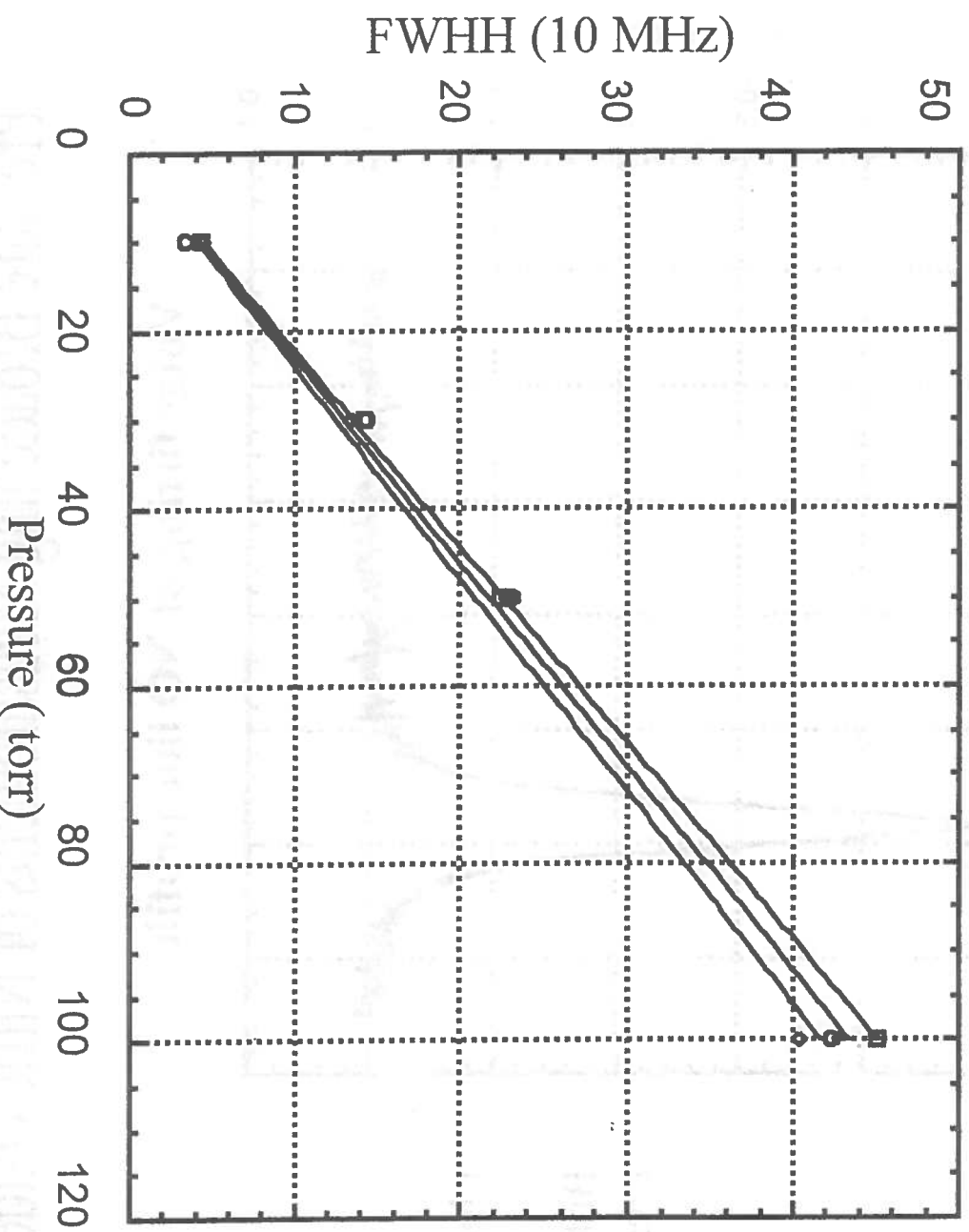
R(15/2)3/2 e,f

Buffer gas: N₂

Pressure: 10 torr

Pressure Broadening Measurements of Nitric Oxide

Full-width at half-height



Upper line:

$R(15/2)_{1/2}e$

Middle line:

$R(15/2)_{1/2}f$

Bottom line:

$R(15/2)_{3/2}e,f$

Pressure Broadening Measurements of Nitric Oxide

$l \leftarrow 0$ transition	Parity	Slope (MHz/torr)	Error (MHz/torr)	$1000 \times \gamma$ ($\text{cm}^{-1} \cdot \text{atm}^{-1}$)
$R(15/2)_{1/2}$	e	0.451	0.003	57.2(0.8)
$R(15/2)_{1/2}$	f	0.417	0.013	52.8(3.2)
$R(15/2)_{3/2}$	e,f	0.434	0.011	54.9(3.0)

Summary:

- * PPLN transmission was measured in the range of 3.5-5.3 μ m.
- * Formula of DFG power calculation with crystal absorption is given, and numerical result shows that the optimum PPLN crystal length for DFG at 5.3 μ m is 1 cm.
- * PPLN is shown to be a suitable medium to produce DFG above 5 μ m
- * Measurements of NO concentration in air at 1903 cm⁻¹ were performed with a precision of 300 ppb.
- * Pressure broadening coefficients of R(15/2) $^2\Pi_{1/2}$ - $^2\Pi_{1/2}$ (e,f) and R(15/2) $^2\Pi_{3/2}$ - $^2\Pi_{3/2}$ (e,f) lines were measured.
- * Future research directions includes:
 - Monitoring NO concentrations in human breath (~30 ppb) and in urban ambient air (~5 ppb)
 - Further development of gas sensor with compact diode or fiber laser pump sources replacing Ti:Sapphire laser
 - Optimized quasi-phase matched bulk and wave guide microstructures.