Signal-to-noise ratio analysis in laser absorption spectrometers using optical multipass cells

Peter Werle and Franz Slemr

In high resolution absorption spectrometers with conventional light sources, the signal-to-noise ratio (SNR) is usually limited by the thermal noise level of the detector–preamplifier combination, which is independent of the light source power. However, the noise in many laser absorption spectrometers is dominated by the excess or shot noise which is dependent on the transmitted laser power, and which in turn is dependent on the number of reflections in a multipass cell. The optimum absorption path length for a high frequency modulated (FM) and a conventional wavelength modulated (WM) diode laser absorption spectrometer is investigated in this paper. The major result is that, due to the power attenuation by the multipass cell, the best SNR of a shot noise limited FM spectrometer is achieved at substantially shorter absorption paths, when compared with the excess noise limited WM spectrometer. This finding implies that the implementation of the FM technique in absorption spectrometers with multipass cells can improve the SNR only by 1 order of magnitude. Although desirable, this is substantially less than the improvement of 2 orders of magnitude expected in quantum limited conditions with a single pass cell.

I. Introduction

A large number of atmospheric trace gases can be monitored by absorption spectroscopy.[1–3] The absorptions that have to be detected are usually small, and to achieve sensitivity adequate for environmental monitoring, absorption spectrometers require long optical paths.[1] In instruments with a limited size, long absorption path lengths up to several hundreds of meters[2] have usually been provided by multireflection optical systems of which the most well known are the systems invented by White[4,5] and by Herriott *et al.*[6,7] In all these systems the sensitivity gained by lengthening the absorption path is offset by the increased attenuation of the radiation power throughput, because of the imperfect reflectivity of the mirrors. Consequently, to achieve the highest SNR each absorption spectrometer has to be operated with an optimal number of reflections in multireflection absorption systems.

The increased sensitivity of absorption spectrometers leads to a greater probability of interference by other atmospheric trace gases. Such interference is usually substantially reduced by operating the spectrometer at low pressures, where the pressure broadening is suppressed. Therefore, high resolution spectroscopy is required to achieve specificity in the measurement of atmospheric trace gases.[1] Unfortunately, conventional light sources have low spectral power density. The detectable optical density of high resolution absorption spectrometers with conventional light sources, such as a Nernst glower, is then usually limited by the thermal noise of the detector–preamplifier combination, which is independent of the incident light power. In these conditions the best SNR can be achieved by adjusting the absorption signal to a maximum, i.e., by adjusting the number of reflections to an optimal value given by the reflectivity of the mirrors. Stephens[8] derived an equation to show that the optimal number of reflections is reached when the reflection losses reduce the light power to $1/e$ of its initial value.

To overcome the limits set by the low spectral power density of conventional light sources, lasers are increasingly used in high resolution absorption spectroscopy for atmospheric trace gas monitoring.[3,9] The detectable optical density of such laser absorption spectrometers is usually limited by amplitude and phase fluctuations of the laser itself. Most of these spectrometers were operated at low modulation frequencies and in these conditions the noise is usually dominated by the $1/f$ laser noise.

The authors are with Fraunhofer Institute for Atmospheric Environment Research, Postfach 1343, D-8100 Garmisch-Partenkirchen, Federal Republic of Germany.

Received 19 January 1990.
0003-6935/91/040430-05$05.00/0.

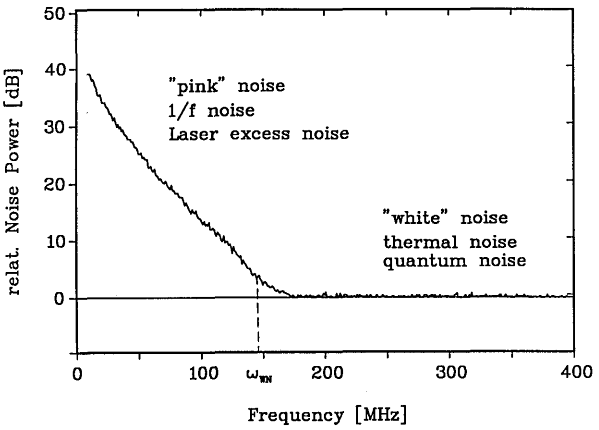

Fig. 1. Noise frequency spectrum of a lead-salt diode laser.

It has been shown that $1/f$ laser noise can be substantially reduced by using high frequency modulation techniques. With a ring dye laser, Hall *et al.*[10] found that the noise level at 1 MHz is lower by 80 dB than at 10 kHz. For lead-salt diode lasers, which are used for monitoring atmospheric trace gases and for high resolution IR spectroscopy, similar measurements showed that the noise level decreased by 40 dB between 2 and 200 MHz.[11] In terms of sensitivity, modulation techniques operated at 200 MHz should improve the SNR by 2 orders of magnitude. As schematically illustrated in Fig. 1, the noise spectrum with sufficient laser power becomes frequency independent above a certain frequency ω_{WN}. The frequency, ω_{WN}, denotes the transition from the frequency dependent $1/f$ or pink noise to frequency independent white noise governed by thermal noise and quantum noise (shot noise). Based on these observations, a high frequency modulation (FM) technique[12,13] was recently developed to avoid the limitations of $1/f$ noise in conventional derivative techniques and to achieve shot noise limited performance. The basic idea of the high frequency modulation (FM) technique is to move, in modulation and detection frequency space, from the currently used low modulation frequencies (derivative technique using lock-in amplifiers = WM technique) to frequencies beyond the above mentioned ω_{WN} frequency.

Both $1/f$ noise and shot noise are strongly dependent on the laser power incident on the detector. Consequently, the optimal path length of an absorption spectrometer limited by detector thermal noise[8] will not apply to the FM and WM techniques. In this paper, therefore, we investigate the SNR of $1/f$ noise and shot noise limited laser absorption spectrometers as a function of the laser power, the mirror reflectivity, and the number of passes in a multipass absorption cell. The results show that the optimal number of reflections has to be calculated individually for the available power and the noise spectrum of each laser. Due to the different power dependences of the SNR in both cases, less reflections are optimum for shot noise limited FM spectrometers than for wavelength modulated ones. The practical consequences of the analysis are discussed.

II. Formal Description of the SNR in Derivative and FM Spectrometers Using Multipass Cells

The absorption of light by atmospheric trace gases is usually small, and then where absorption occurs signal S is proportional to the optical path length. In a multipass cell optical path length $L = n \cdot b$, where n is the number of passes and b is the base length of the multipass cell. The signal is also proportional to radiation power P_D incident on the detector after passing through the multipass cell. The number density and modulated absorption coefficient for the species under study are assumed to be constant for the present analysis. Neglecting all other losses, this power decreases with an increasing number of passes, and increases with the increasing reflectivity of mirrors R. The signal of a laser absorption spectrometer with fixed basis length is then proportional to

$$S \sim nP_D = nR^{n-1}P_0, \tag{1}$$

where P_0 is the laser power incident on the detector with a single pass of the cell.

The noise generated by a laser absorption spectrometer consists mainly of three components: thermal noise, shot noise, and laser excess noise. The laser excess noise is referred to as $1/f$ noise because it is frequency dependent. Each of these components has a different dependence on the laser power incident on the detector:

$$N_{TN}^2 = \text{constant}, \tag{2}$$

$$N_{SN}^2 = \alpha P_D, \tag{3}$$

$$N_{EX}^2 = \beta(\omega)P_D^2, \tag{4}$$

where N_{TN}, N_{SN}, and N_{EX} are the *rms* detector noise currents due to thermal, shot, and excess noise, respectively, and α and $\beta(\omega)$ are proportionality coefficients.[11] The original analysis by Stephens[8] considered only the thermal noise in detecting system N_{TN}.

The SNR of a laser absorption spectrometer can be written as

$$\begin{aligned} \text{SNR} &\sim nP_D/(N_{TN}^2 + N_{SN}^2 + N_{EX}^2)^{1/2} \\ &\sim nP_D/(N_{TN}^2 + \alpha P_D + \beta(\omega)P_D^2)^{1/2}, \end{aligned} \tag{5}$$

where, in the denominator, shot noise increases with increasing power, and thermal noise is power independent. Power level $P_{\min}$ can then be defined for which shot noise equals thermal noise:

$$N_{SN}(P_{\min})^2 = N_{TN}^2 = \alpha P_{\min}. \tag{6}$$

Using the variables $\gamma(P_D)$ and $\delta(\omega)$, where

$$\gamma(P_D) = 1 + P_D/P_{\min}, \tag{7}$$

$$\delta(\omega) = \beta(\omega)/N_{TN}^2, \tag{8}$$

Eq. (5) can be rewritten in a more convenient form:

$$\text{SNR} \sim nP_D/N_{TN}[\gamma(P_D) + \delta(\omega)P_D^2]^{1/2}. \tag{9}$$

The meaning of the variables becomes clear from this expression. Variable $\gamma(P_D)$ describes how much higher the sum of thermal and shot noise is above thermal noise; at $\gamma(P_D) > 2$, shot noise dominates, whereas at $\gamma(P_D) \sim 1$, thermal noise dominates and shot noise can be neglected. $\gamma(P_D)$ is frequency independent since both thermal noise and shot noise are also frequency independent. Variable $\delta(\omega)$ takes into account the frequency dependence of the laser excess noise and is a decreasing function of modulation frequency ω_{mod}. It can be calculated from the ratio of laser excess noise to the total white noise level using

$$N_{\mathrm{EX}}^2/(N_{\mathrm{TN}}^2 + N_{\mathrm{SN}}^2) = \delta(\omega)P_D^2/\gamma(P_D). \tag{10}$$

As shown in Fig. 1 the laser excess noise decreases with increasing modulation frequency, and at frequency ω_{WN} it becomes equal to the white noise:

$$\delta(\omega_{\mathrm{WN}})P_D^2 = \gamma(P_D). \tag{11}$$

At a modulation frequency of $\omega_{\mathrm{mod}} = \omega_{\mathrm{WN}}$, the total noise level is 3 dB above the white noise level. Variable $\delta(\omega)$ is thus used to distinguish the shot noise limited FM case from the laser excess noise limited WM case.

As $P_D = R^{n-1}P_0$, expressions (9) and (7) can be rewritten as

$$\mathrm{SNR}(n) \sim nR^{n-1}P_0/N_{TN}[\gamma(n) + \delta(\omega)R^{2n-2}P_0^2]^{1/2}, \tag{12}$$

and $\gamma(n) = 1 + R^{n-1}P_0/P_{\mathrm{min}}$.

Depending on the modulation frequency, two different cases can be investigated: low frequency modulation (WM) and high frequency modulation (FM). In the WM case, $\omega \ll \omega_{\mathrm{WN}}$ and $\delta(\omega) \approx 1$. The SNR is then dictated by the laser excess noise:

$$\mathrm{SNR}(n) \sim n\, R^{n-1}P_0/N_{TN}[\gamma(n) + \delta(\omega)R^{2n-2}P_0^2]^{1/2}. \tag{13}$$

In the FM case, $\omega > \omega_{\mathrm{WN}}$ and $\delta(\omega) \ll 1$, consequently, $\delta(\omega)R^{2n-2}P_0^2 \ll \gamma(n)$ and the SNR is dominated by white noise:

$$\mathrm{SNR}(n) \sim nR^{n-1}P_0/N_{TN}\gamma(n)^{1/2}. \tag{14}$$

With an increasing number of passes the power incident on the detector will decrease until the quantum noise level becomes smaller than the thermal noise of the detector–preamplifier combination, i.e., $\lim\gamma(n) = 1$ for $n \rightarrow \infty$. Therefore, both the FM and WM cases switch to the thermal noise-limited regime with an increasing number of passes and

$$\lim_{n\rightarrow\infty} \mathrm{SNR} = nR^{n-1}P_0/N_{TN}. \tag{15}$$

This is the expression used by Stephens[8] for estimation of the optimal number of passes.

III. Results and Discussion

Equation (12) is used to calculate the SNR as a function of number of passes for conditions usually encountered in tunable diode laser absorption spectrometers (TDLASs). The values of variable $\delta(\omega)$ are derived from the measured wideband noise characteristics of a lead-salt diode laser.[11] In this work, a 40-dB noise reduction was observed when the modulation frequency was changed from ~10 kHz, where the laser excess noise dominates, to ~200 MHz. The laser power incident on detector P_D was 521 μW and minimum power P_{min} calculated from the effective detector thermal noise was 67 μW. From these results values of $\delta(\omega)$ can be estimated for two limiting cases:

$$\begin{aligned} &\mathrm{WM:}\ \delta(\omega \ll \omega_{\mathrm{WN}}) = 0.3\mu\mathrm{W}^{-2},\\ &\mathrm{FM:}\ \delta(\omega \gg \omega_{\mathrm{WN}}) = 3 \times 10^{-5}\mu\mathrm{W}^{-2}, \end{aligned} \tag{16}$$

respectively. Using these values for δ the SNR can be calculated as a function of the number of reflections n and mirror reflectivity R.

A central point of this analysis is the laser power level in the detector. Figure 2 shows the power incident on detector P_D and variable $\gamma(n)$ as a function of the number of passes n. The dependence is calculated

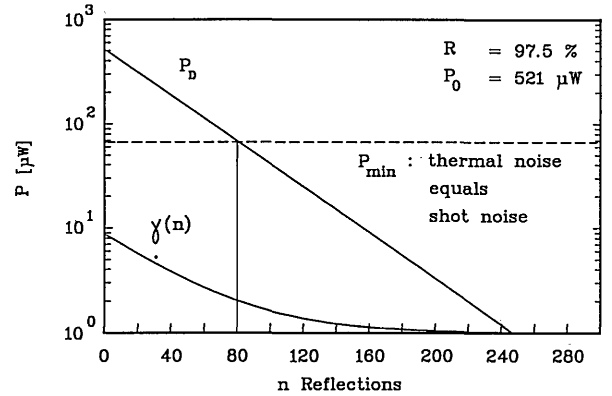

Fig. 2. Light power incident on detector P_D and variable γ as a function of number of passes in a multipass cell. Variable γ describes how much higher the white noise, consisting of thermal noise and shot noise, is in comparison with the detector thermal noise.

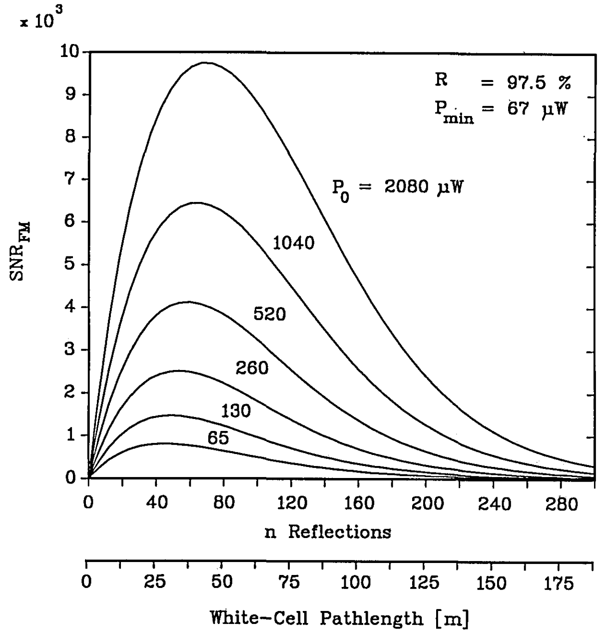

Fig. 3. Signal-to-noise ratio of a frequency modulated laser absorption spectrometer as a function of number of passes in a multipass cell for different laser output power. The calculation is based on detector noise equivalent power of 67 μW, mirror reflectivity of 97.5%, cell base length of 62.5 cm, and variable $\delta(\omega)$ (see definition in text) equal to 3×10^{-5}.

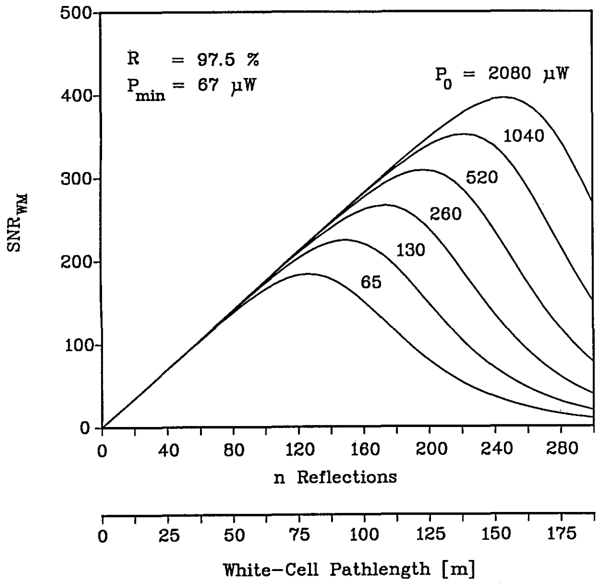

Fig. 4. Signal-to-noise ratio of a wavelength modulated laser absorption spectrometer as a function of number of passes in a multipass cell for different laser output power. The input parameters are the same as in Fig. 3 with the exception of variable $\delta(\omega)$ which is 0.3 in this case.

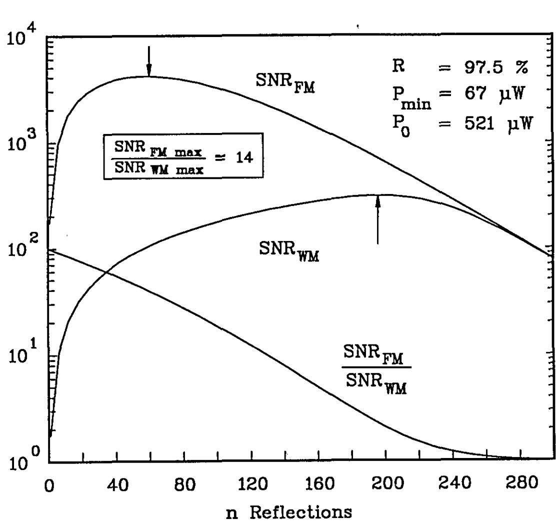

Fig. 5. Signal-to-noise ratio of frequency modulated (FM) and wavelength modulated (WM) laser absorption spectrometers and their ratio as a function of number of passes in a multipass cell for laser power of 521 μW. The input parameters are the same as in Figs. 3 and 4.

for a laser power P_0 of 521 μW and a measured reflectivity of 97.5% for the mirrors in the White cell. The latter value is lower than the specification which is ~98.6%; the difference may be caused by some dust on the mirror surface. As already mentioned, $P_{\min}$ corresponds to 67 μW, and this power level is reached after 80 passes. Therefore, shot noise limited performance of the FM spectrometer can only be expected with less than 80 passes; a higher number of passes will lead to a decrease in the SNR.

In Fig. 3, the SNR calculated from Eq. (14) for FM absorption spectroscopy is plotted as a function of the number of passes at various laser powers. $P_{\min}$ is assumed to be 67 μW, the path length is calculated using the cell base length of 62.5 cm, and $\delta(\omega)$ is 3×10^{-5} as derived in the last section. The calculated optimal number of passes varies between 40 and 70, corresponding to a path length of ~25–45 m, depending on the laser power. With increasing laser power, the optimum number of passes increases only slowly—an increase of less than a factor of 2 for a 32 times increase in laser power.

The equivalent calculation for SNR from Eq. (13) for WM absorption spectroscopy is shown in Fig. 4. With the exception of $\delta(\omega)$, which is set to 0.3, all the other parameters are the same as before. The optimum number of passes again increases with increased laser power. In comparison with the previous calculation, the number of optimal passes is much larger and increases more steeply with increasing power than in the case of FM spectroscopy. The number of optimal passes between 120 and 200 corresponds to the number of passes used in commercial WM-TDLAS instruments.[14]

In Fig. 5 the FM and WM limiting cases are compared for a laser with a power of 521 μW. The ratio of $\mathrm{SNR_{FM}}$ and $\mathrm{SNR_{WM}}$ is also plotted as a function of the number of passes. The ratio shows the highest value, ~100, at very short path lengths. The ratio corresponds to the calculated improvement in sensitivity by application of the FM technique.[13] Given a practical number of passes, however, the sensitivity improvement is substantially smaller. At an optimal number of passes for the FM technique of ~60, the $\mathrm{SNR_{FM}}$ is only about 40 times better than $\mathrm{SNR_{WM}}$; and when the FM and WM techniques are each operated at an optimal number of passes, introduction of the FM technique will only improve the SNR by about a factor of 14. The optimal number of passes for the FM technique is only about one-third of the optimal number of passes for the WM technique. In practical terms, cells with a lower number of passes can be built more cheaply and adjusted more easily than cells designed for a high number of passes.

Another interesting aspect can be derived from Fig. 5. If a conventional derivative multipath spectrometer is equipped with FM modulation and detection circuitry and the adjustment of the absorption cell is not changed, no substantial sensitivity gain will be found. To achieve better sensitivity with the FM technique the number of reflections must be reduced.

The SNR for WM and FM techniques are shown in Figs. 6 and 7, respectively, as a function of the reflectivity of the mirrors. For this calculation the same parameters were used as for Fig. 5. In both cases the optimal number of passes and the achievable SNR increase rapidly with the improvement of mirror reflectivity. Improving the reflectivity from 96.5 to 98.5% improves the achievable SNR in both cases by almost a factor of 2.5, and it increases the optimal number of passes by a factor of ~2.5. This result stresses the need for high reflectivity mirrors in multipass systems for trace gas analysis.

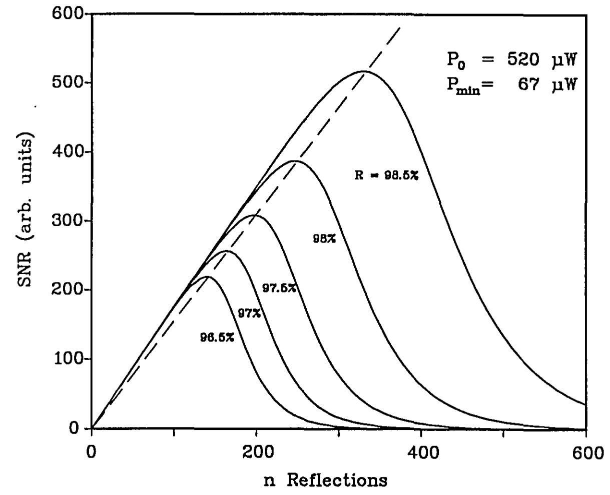

Fig. 6. Signal-to-noise ratio of a wavelength modulated (WM) laser absorption spectrometer as a function of number of passes at different mirror reflectivities.

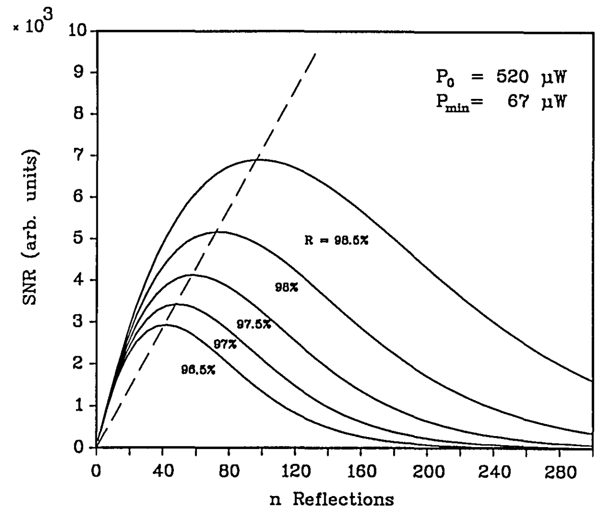

Fig. 7. Signal-to-noise ratio of a frequency modulated (FM) laser absorption spectrometer as a function of number of passes at different mirror reflectivities.

IV. Conclusions

The signal-to-noise ratio in optical multipass cells used for monitoring trace gas concentrations was studied using high frequency (FM) as well as conventional derivative (WM) laser absorption spectrometers. In conditions usually encountered in TDLASs, the optimal number of passes for FM spectrometers was found to be much smaller than the optimal number of passes for WM spectrometers. This finding implies that, if both techniques are operated with an optimal number of passes, the introduction of FM techniques can improve the ultimate SNR in spectrometers using optical multipass cells by only about an order of magnitude. Although still highly desirable, this is substantially lower than the 2 orders of magnitude potential improvement derived solely from the noise analysis, without considering the use of multipass cells.

The smaller number of passes required for optimal SNR in FM spectrometers has other practical consequences. Cells with a smaller number of passes are cheaper to construct and to adjust. Since they can be designed with smaller volume, faster measurements can be made. This property is especially important because only in cells with a small volume can the potential high speed of FM measurements be utilized.

If, on the other hand, ultrasensitive measurements require long absorption path length, high reflectivity mirrors and high power lasers are a prerequisite to get the full potential of FM techniques.

This work was carried out with support from the German Ministry of Research & Technology and it is part of EUROTRAC, an environmental program of EUREKA.

References

1. P. L. Hanst, "Spectroscopic Methods for Air Pollution Measurements," Adv. Environ. Sci. Technol. **2,** 91 (1971).
2. E. C. Tuazon, A. M. Winer, R. A. Graham, and J. N. Pitts, Jr., "Atmospheric Measurements of Trace Pollutants by Kilometer Pathlength FT-IR Spectroscopy," Adv. Environ. Sci. Technol. **10,** (1979).
3. D. R. Hastie, G. I. Mackay, T. Iguchi, B. A. Ridley, and H. I. Schiff, "Tunable Diode Laser System for Measuring Trace Gases in Tropospheric Air," Environ. Sci. Technol. **17,** 352A–364A (1983).
4. J. U. White, "Long Optical Paths of Large Aperture," J. Opt. Soc. Am. **32,** 285 (1942).
5. J. U. White, "Very Long Optical Paths in Air," J. Opt. Soc. Am. **66,** 411–416 (1976).
6. D. R. Herriott, H. Kogelnik, and R. Kompfner, "Off-Axis Paths in Spherical Mirror Interferometers," Appl. Opt. **3,** 523 (1964).
7. D. R. Herriott and H. J. Schulte, "Folded Optical Delay Lines," Appl. Opt. **4,** 883 (1965).
8. E. R. Stephens, "Long-Path Infrared Spectroscopy for Air Pollution Research," Appl. Spectrosc. **3,** 80–84 (1958).
9. G. Hübler, D. Perner, U. Platt, A. Tönissen, and D. H. Ehhalt, "Ground Level OH Radical Concentration: New Measurements by Optical Absorption," J. Geophys. Res. **89,** 1309 (1984).
10. J. L. Hall, T. Baer, L. Hallberg, and H. G. Robinson, "Precision Spectroscopy and Laser Frequency Control Using FM Sideband Optical Heterodyne Techniques," in *Laser Spectroscopy V,* A. R. W. McKellar, T. Oka, and B. P. Stoicheff, Eds. (Springer-Verlag, Berlin, 1981), p. 16.
11. P. Werle, F. Slemr, M. Gehrtz, and C. Braüchle, "Wideband Noise Characteristics of a Lead-Salt Diode Laser: Possibility of Quantum Noise Limited TDLAS Performance," Appl. Opt. **28,** 1638–1642 (1989).
12. G. C. Bjorklund, "Frequency-Modulation Spectroscopy: a New Method for Measuring Weak Absorptions and Dispersions," Opt. Lett. **5,** 15–17 (1980).
13. P. Werle, F. Slemr, M. Gehrtz, and C. Braüchle, "Quantum Limited FM-Spectroscopy with Lead-Salt Diode Laser," Appl. Phys. B **49,** 99–108 (1989) and references therein.
14. H. I. Schiff, G. W. Harris, and G. I. Mackay, "Measurement of Atmospheric Gases by Laser Absorption Spectrometry," in *The Chemistry of Acid Rain: Sources and Atmospheric Processes,* R. W. Johnson, G. E. Gordon, W. Calkins, and A. Z. Elzerman, Eds., ACS Symposium Series No. 349 (ACS, Washington, DC, 1987).