Double beam laser absorption spectroscopy: shot noise-limited performance at baseband with a novel electronic noise canceller

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ABSTRACT

A novel electronic circuit has been developed that detects absorption in one of two (reference and signal) laser beams with shot noise-limited sensitivity. We demonstrate its use as a simple shot noise limited spectroscopy by measuring several absorption lines of I_2 vapor near 670 nm with a tunable diode laser. The noise-equivalent absorption in this double-beam method is $\sqrt{2}$ times the shot-noise-to-signal ratio of the signal beam. In our case, for 1.2 mW of power, the noise-equivalent absorption in a 1-Hz bandwidth was 4.2×10^{-8}. This noise level was confirmed, although our measurements were sometimes limited by interference fringes and laser tuning drift. These are technical, not fundamental, problems arising from unsophisticated optical equipment. This baseband method provides absorption spectra directly, eliminating laser excess noise without the somewhat elaborate signal generation and processing equipment needed in laser frequency modulation methods. The only modulation used in our system was the 0.5 to 1.0-kHz sweep of the diode laser current used to tune the laser output wavelength. The noise-cancelling circuit itself is not complex: workable versions can be constructed from readily available components at a cost of about 10 dollars.

1. INTRODUCTION

In its simplest forms, optical absorption spectroscopy is not an ultrasensitive analytical method, since one must measure a small change in a large, usually noisy, background. General-purpose dispersive and transform spectrometers have noise-equivalent absorptions on the order of 10^{-4} because of fluctuations in their incoherent lamp sources, or because the detection of these low-brightness sources is either Johnson noise-limited, or can achieve only modest shot noise statistics in a reasonable time. On the other hand, a coherent, high-brightness laser is capable of superior shot noise performance: For example, a 1-mW, 633-nm HeNe laser beam has a photon flux $N = 3.2 \times 10^{15}\ \mathrm{s}^{-1}$. In a measurement time of one second, a unit quantum-efficiency detector would therefore measure the intensity of the beam at a noise-to-signal ratio of $\sqrt{N}/N = 1.8 \times 10^{-8}$, and the noise-equivalent absorption would be approximately a factor of $\sqrt{2}$ higher.

Such precision is seldom achieved in actual measurements, even when Johnson noise in the detector and its support electronics is lower than the optical shot noise, because of excess laser noise and environmental perturbations. *Excess noise* refers to all unintended intensity modulation of the beam other than the laser shot noise. Typical causes for such noise include mode competition in the laser cavity, cavity optical feedback, power-supply instability, and physical perturbations of the cavity caused by vibration and temperature fluctuations. Vibrational and thermal perturbations also affect the optical system as a whole, as do atmospheric aerosols, air currents, and incidental Fabry-Perot fringes between nearly parallel optical surfaces, all of which contribute excess noise in the detected laser power. For many laser systems, the power spectral density (PSD) of the optical excess noise follows a $1/f$ distribution, to which may be added spurious signals due to power supply ripple and other discrete frequency components. The frequency at which excess noise has decayed below shot noise (which is evenly distributed, "white" noise) depends on the specific laser system, and can range from one to several hundred MHz. These effects limit single laser beam direct absorption spectroscopy to about the same noise-equivalent absorption as obtained with conventional light sources.

There are many ways to increase the sensitivity of single-beam laser absorption measurements, but few fully exploit the fundamental shot noise advantage of the laser. In this paper, we present a new method for measuring optical absorption which avoids most of the pitfalls of simple single- and double-beam spectroscopy while preserving their simplicity and low cost. The method is a modification of double-beam differential spectroscopy, so we first review the existing methods in order to make clear where our method differs.

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2. OVERVIEW OF SINGLE-BEAM LASER ABSORPTION MEASUREMENTS

The most common method of increasing the signal-to-noise (S/N) ratio in laser absorption measurements is to chop the laser beam and synchronously detect the detector photocurrent with a lock-in amplifier.[1] Although narrowing the detection bandwidth this way attenuates out-of-band noise (outside the passband of the lock-in), the absorption sensitivity will almost never be limited by optical shot noise for two reasons: First, the excess noise spectrum of most lasers extends far beyond the 1 to 10-kHz frequencies attainable with choppers. Second, the act of chopping intermodulates the signal with the baseband noise so that the low-frequency noise is shifted into the passband regardless of where it is.

Other methods to escape low-frequency laser noise in absorption spectroscopy involve modulating the frequency (wavelength) of the laser beam. These methods are most popular with diode lasers, whose wavelengths may be current-tuned very rapidly. The most direct approach is to apply a rapid, repetitive wavelength sweep and use a signal averager to combine data from many sweeps.[2] The sweep frequency in such systems is generally limited to fairly low values (100-500 kHz), which usually lie inside the excess noise region. Also, with diode lasers, the bias current sweep modulates the laser output amplitude as well as wavelength; small absorption peaks are easily obscured by the resulting nonlinear variation in the laser power.

Reduction of this background is a principal advantage of derivative spectroscopy.[3] In this method, a low-frequency (1-10 kHz), low-amplitude wavelength modulation is imposed on the laser output, and the photodetector output is demodulated with a lock-in amplifier referenced to the nth harmonic of the modulation frequency. As the dc bias current is scanned slowly, sweeping the laser through spectral absorption lines, the output of the lock-in produces a waveform approximating the nth derivative of the absorption spectrum. Background variations that are gradual with respect to the wavelength dither are rejected. As with straight chopping, the modulation frequency is generally too small to escape the excess noise bandwidth of most diode lasers, and measurement of weak absorptions is a slow process, requiring long lock-in time constants and scan times.

Theoretically, single-beam laser absorption spectroscopy at the shot noise limit can be achieved by rapid frequency modulation (FM) of the laser wavelength at radio frequencies (100-1000 MHz).[4,5] At these modulation frequencies, the optical spectrum of the laser beam consists of the original wavelength, or carrier frequency, and several sidebands around the carrier, separated by the modulation frequency. (Such sidebands also exist when the laser is chopped or frequency modulated at lower frequencies, but the linewidth of the laser is usually so wide that the sidebands are not resolved.) In the absence of any absorption, a dc photocurrent is produced proportional to the average power incident on the detector. Samples with absorption features comparable in width to the sideband spacing, such as Doppler broadened ro-vibrational lines of low pressure gases, cause differential absorption of the sidebands with respect to the carrier, producing ac beat signals proportional to the intensity of the absorption. By sweeping the laser carrier frequency while demodulating the detector output at the beat frequency, a spectrum of absorption features on a nominally zero baseline is obtained. The modulation frequency is chosen to lie beyond the excess noise region, so the absorption sensitivity ought to be limited only by the shot noise of the average dc photocurrent appearing in the baseline.

In practice, frequency modulation spectroscopy encounters several limitations; the most salient is the residual amplitude modulation (AM), invariably produced along with the desired FM when either current-tuned diode lasers[5,6] or electro-optically modulated dye lasers are used.[7] The residual AM mixes with the low-frequency excess laser noise, resulting in intermodulation noise products inside the measurement passband. The magnitude of this interference is usually small, and is sometimes unimportant compared to residual Fabry-Perot fringes and other technical deficiencies in a given optical system. Therefore, almost all such experiments need sophisticated optical design[6] and some auxiliary noise suppression to reach the shot noise limit: double-beam subtraction is often used.[7,8]

3. DOUBLE-BEAM LASER NOISE CANCELLATION

Double-beam differential absorption methods have been used for years in conventional absorption spectrometers. When applied to laser absorption measurements, they have not produced shot noise-limited measurements, but in principle they could. With this technique, the laser output is split into a signal and a reference beam. The signal beam traverses an absorption cell while the reference beam travels a nearly identical path, after which each beam falls on its own detector. The resulting photocurrents are subtracted or divided, yielding an output which, since the beams have exactly the same relative excess noise, should be completely free from excess laser noise.

As conventionally practiced, subtraction requires extremely fine adjustment of the relative intensities of the beams for complete noise cancellation, which must be maintained over the full scan range, time, and temperature fluctuations of the measurement. Division suffers from the poor performance of analog dividers, which are slow and noisy. To achieve 40 dB of (electrical) noise reduction, the adjustment of the subtracter or the closed loop gain of the divider must be accurate to 1% throughout the measurement bandwidth. For a typical divider, this limits the measurement bandwidth to a few kHz. The modest performance of these techniques has restricted double-beam measurements to relatively low-performance applications.

While the usual double-beam systems are of limited use, they ought to be extremely effective since they rely only on two important properties of most optical systems: very wide temporal bandwidth and excellent linearity. These properties ensure that propagation of the beams through the optical system does not change the relative amplitudes and phases of the intensity noise modulation components. Therefore, the instantaneous fractional excess intensity noises of the beams are identical, and so are the corresponding excess photocurrent noises. Provided that path-length differences are small compared to the wavelength of the modulation (30 meters for a 10-Hz noise component), appropriate "waveform arithmetic" could cancel the excess noise perfectly, leaving only variations in the signal beam imparted by the experiment, and a background that is the root-mean-square (RMS) sum of the uncorrelated shot noise of the signal and reference beams. This does not depend on matched photodetectors, as long as they are linear. For a perfect subtracter, since the noise is exactly proportional to the average intensity, if the dc photocurrents cancel, *the noise components at all frequencies of interest cancel identically*.

The problems of an ordinary double-beam measurement can be essentially solved through the use of the laser noise cancelling circuit in Figure 1. This circuit has been described in detail elsewhere; its performance closely approximates the ideal differential system.[9] It uses a differential pair (Q1 and Q2) of bipolar junction transistors (BJT's) to subdivide the reference photocurrent so that an identical, unmodulated copy of the signal photocurrent is subtracted at the inverting input of amplifier A1. (We use the term *subdivide* to avoid confusion with analog dividers.) BJT's are nearly ideal transconductance devices, and the subdivision ratio of the pair depends only on the difference ΔV_{BE} of their base-emitter voltages, and not on the value of the currents themselves. (BJT's are unique in this.) Since the current subdivision ratio is electronically variable and there is a simple electronic criterion for finding perfect balance, namely zero dc difference current, the circuit applies feedback to adjust the subdivision ratio automatically. It has two outputs, one of which is a high-pass filtered version of the modulation on the signal beam, and the other a low-pass filtered voltage related logarithmically to the ratio of the two beam intensities:

$$V_{\log} = -\ln\left(\frac{I_{ref}}{I_{sig}} - 1\right), \tag{1}$$

where $V_{\log}$ will be given in Volts. The low-pass/high-pass cutoff frequency is set by the bandwidth of the feedback loop, and can be changed easily to suit the application, since the effective bandwidth of the noise cancellation depends only on the current-gain bandwidth product f_T of the BJT's, not on the feedback bandwidth.

At both outputs, the circuit suppresses excess laser noise down to 3 dB above the shot noise of the signal-beam photocurrent, a consequence of combining two equal photocurrents at the inverting input of A1. Extrinsic

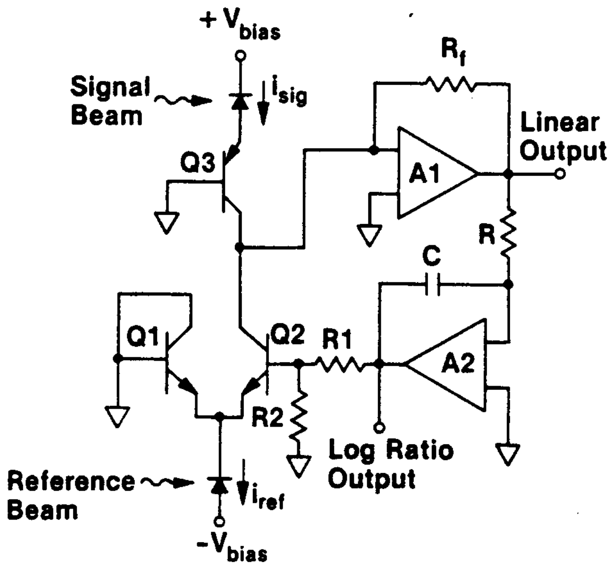

Figure 1. Schematic diagram of the laser noise canceller (see text).

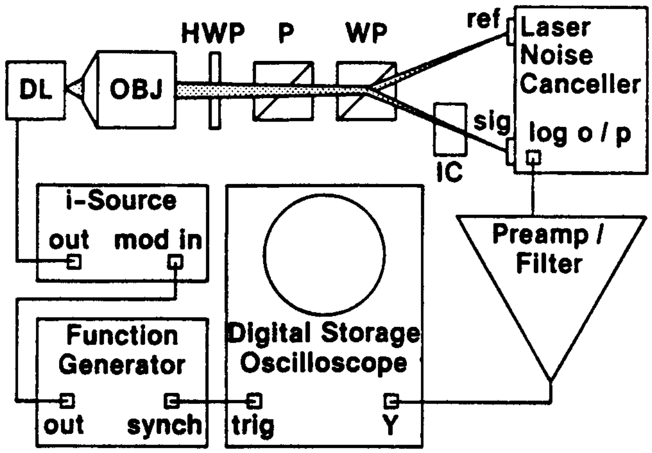

Figure 2. Diode laser absorption spectroscopy apparatus. DL = temperature controlled diode laser mount; OBJ = microscope objective; HWP = half-wave plate; P = crystal polarizer; WP = Wollaston prism; IC = I_2 cell.

emitter resistance and β nonlinearity in the BJT's limit the circuit to 50-60 dB of excess noise cancellation; fortunately, many readily available lasers are not this noisy. There are no critical adjustments; one need only ensure that the reference-beam intensity is somewhat larger than the signal-beam intensity to allow subdivision. Further explication of the circuit's operation can be found in the original reference.[9]

4. EXPERIMENTAL SECTION

To demonstrate the laser noise canceller in an absorption spectroscopy measurement, we used a Toshiba TOLD 9211 visible diode laser to measure the weak rotationally and vibrationally resolved absorption lines in the $B^3\Pi_{0_u^+} \leftarrow X^1\Sigma_g^+$ system of I_2. A schematic diagram of the experimental apparatus is shown in Figure 2. The laser was mounted in a Melles-Griot thermoelectric temperature controller, which maintained case temperature to ± 0.05 °C or better, allowing easy, stable temperature tuning. Very low noise bias current for the laser was provided by an ILX Lightwave Corporation LDX-3620 current supply, which allows external current modulation. The laser produces a 3-5 mW single longitudinal mode output near 670 nm at room temperature. Figure 3 shows typical data for both temperature and bias current tuning modes, in which the laser output wavelength was measured by counting interference fringes *in vacuo* relative to a stabilized HeNe laser in a Burleigh Wavemeter. The continuous tuning range of the laser was limited by mode hops, as is typical of diode lasers. There are several mode hops evident in the temperature tuning data (at 21.2, 22.2, and 24.8 °C), where the wavelength reading was unstable. Similarly, mode hops limited the current tuning range to the $\simeq 0.5$ cm^{-1} range shown in the current tuning data. It is not possible to tune the laser to all wavelengths within its range.

The optical system was extremely simple. The laser light was collected by an extra long working distance microscope objective (Mitutoyo M Plan Apo, NA = 0.28), to minimize optical feedback from front-surface reflections, yielding a slowly converging beam. The beam went through a rotatable half-wave plate and a crystal polarizer to a Wollaston prism, which split the beam into orthogonally polarized signal and reference beams. Each beam fell on its respective photodiode of the noise canceller. To take spectra, an absorption cell was placed in the signal beam.

The crystal polarizer was used to eliminate polarization noise, which divides unevenly in the Wollaston beamsplitter and cannot be suppressed. Rotating the polarizer adjusted the relative intensities of the signal and reference beams; for convenience, we used an orientation in which the reference beam was about twice as intense as the signal beam. The half-wave plate was used to align the beam polarization with the polarizer for optimum

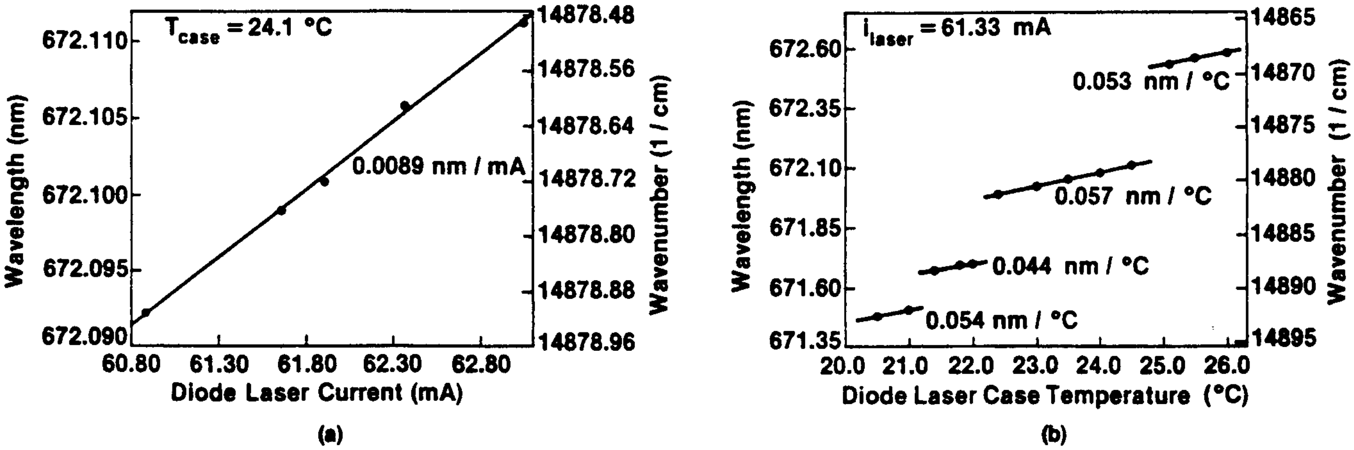

Figure 3. **Diode laser wavelength tuning rates. (a) Laser wavelength vs bias current at fixed temperature. (b) Laser wavelength vs case temperature at fixed bias current. Wavelength measurements are represented by the filled circles; the solid lines are unweighted linear least-squares fits with the tuning rates (slopes) indicated.**

transmission. Somewhat surprisingly, we found that maximizing the signal beam power was not always the best thing to do. Although doing so provides the best possible shot noise performance, inadvertent optical feedback into the laser cavity created enormous excess noise; rotating the half-wave plate only a few degrees from maximum power damped the finesse of the multiple reflections going back into the cavity.

The performance of the system, with a dc bias current running through the laser and no absorbers present in the signal beam, is shown in Figure 4. The top trace shows the noise spectrum of the laser, taken from the linear output of the noise canceller, as measured with a Hewlett-Packard 3585A spectrum analyzer. The reference beam was blocked, causing the circuit to act as an ordinary photocurrent-to-voltage (transimpedance) amplifier. There is a general $1/f$ character to the noise, as well as a spurious component at about 100 kHz due to current supply ripple. The bottom trace was taken with the reference beam unblocked. The noise spectrum is now flat, the excess noise is reduced by about 10-15 dB, and the 100 kHz spurious modulation is gone.

It is straightforward to calculate the expected noise density of the lower trace if it is limited by optical shot noise. In a 1-Hz bandwidth, the full RMS shot noise of a photocurrent is given by

$$\sigma_{i_{photo}} = \sqrt{2qi_{photo}}\,, \tag{2}$$

where q is the electron charge.[10] The RMS noise voltage from the transimpedance amplifier is R_f times this. With the reference beam blocked, the dc output level was $V_{photo} = 5.67$ V . With a feedback resistance $R_f = 20$ kΩ and a photodiode responsivity at 670 nm of about 0.3 A/W, the photovoltage corresponds to a photocurrent of $i_{photo} = 284\ \mu$A and a signal beam power of 945 μW. Thus $\sigma_{V_{photo}} = 190$ nV/$\sqrt{\text{Hz}}$, well above the 11 nV/$\sqrt{\text{Hz}}$ noise floor of the analyzer. The noise density of the lower trace was measured to be 290 nV/$\sqrt{\text{Hz}}$, 3.7 dB above the signal photocurrent shot noise PSD, in good agreement with the expected value of 3 dB.

Near 670 nm, the survey spectrum of Gerstenkorn and Luc (G&L) shows that the strongest I_2 lines have peak absorptions on the order of 1% in a 10-cm path length for I_2 vapor in equilibrium with the solid at room temperature.[11] Two spectroscopic absorption cells with path lengths of 10 cm and 1 mm were used for this work. Both cells were evacuated to below 10^{-6} torr and sealed off with their I_2 charges frozen at 77 K; so, at room temperature, the ≃0.3 torr cell pressure was almost entirely I_2 vapor. The longer cell was used for comparison spectra and setup, while the shorter one, whose absorption lines were 100 times weaker, was convenient for demonstrating ultrasensitive detection. To verify the spectroscopic origin of absorption features, the small I_2 cell was warmed with a heat gun to sublime more I_2, greatly increasing the strength of real absorption lines relative to any spurious features.

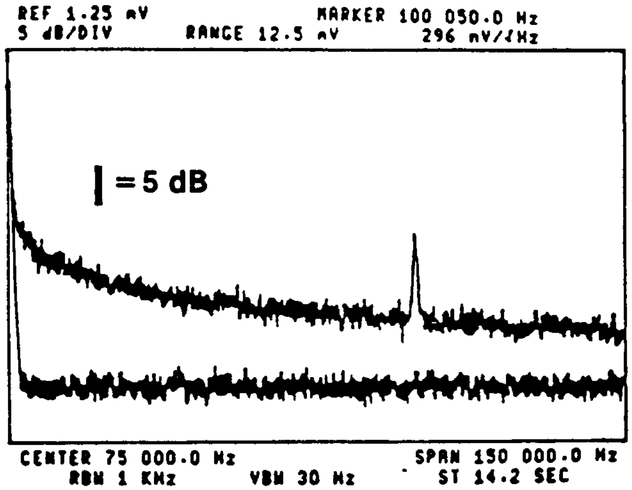

Figure 4. **Diode laser noise cancellation performance. PSD of the linear output of the laser noise canceller with the reference beam blocked (top trace) and unblocked (bottom trace). The loop bandwidth of the noise canceller was about 10 Hz. The signal beam power was 945 μW (see text).**

To obtain spectra, triangle waves from a Hewlett-Packard 3325B function generator were fed into the modulation input of the current supply (Figure 2), which had a sensitivity of 100 mA/V. With the 10-cm path absorption cell (or warmed 1-mm path cell) inserted in the signal beam, the signal from the logarithmic output of the noise canceller could be observed in real time on a LeCroy 9400A storage oscilloscope, set to trigger continuously on the synch signal of the function generator. The laser temperature, dc bias current, or the width of the optical frequency scan could then be adjusted until a satisfactory spectrum was obtained. In recording the spectra of very small absorptions, an Ithaco 1201 low-noise preamplifier/filter was used between the output of the noise canceller and the input to the oscilloscope, to control the measurement bandwidth and provide sufficient gain to allow the μV-level signals corresponding to a few parts-per-million (ppm) of absorption to be measured efficiently by the 8-bit oscilloscope. Analog filtering in conjunction with a reasonable number of sweep averages on the digital oscilloscope allowed shot-noise limited spectra of ppm absorption peaks to be recorded. For laser wavelength sweep frequencies of 500 to 1000 Hz, 1000 sweeps could be averaged in a few seconds.

5. RESULTS AND DISCUSSION

5.1 Preliminary spectral scans

The Toshiba TOLD 9211 diode laser exhibited a severe power variation as the bias current was scanned to tune the laser wavelength, making direct observation of even relatively strong (about 1%) peak absorptions nearly impossible. In Figure 5, the laser current was scanned by imposing a 1000-Hz, 4.56-mA_{pp} triangle wave on an average dc current of 49.98 mA, producing a roughly 0.9 cm^{-1}-wide wavelength scan. The center of the scan was near 14941 cm^{-1} (669.3 nm). Trace (a) of Figure 5 shows a half cycle of the current scan, recording the linear output of the noise canceller with its reference beam blocked, so the circuit acted as a transimpedance amplifier. The signal beam traversed a 10-cm path length I_2 vapor cell (in equilibrium with the solid) at room temperature. Two small absorption peaks on trace (a) (see the expansion trace (b)), are barely discernible on the steeply sloping background. These traces are a single-shot records (no averaging has been performed), but no amount of straight averaging will bring these peaks out of the sloping background. Trace (c) is a single-shot record of the log output of the noise canceller with the reference beam open. The ratiometric property of this output enhances the absorption peaks barely seen in trace (a) and many weaker peaks as well. In fact, if it were not for the coincidence of the largest peaks on trace (c) with the small features on trace (a), it would be difficult to detect any absorption peaks in trace (a) at all.

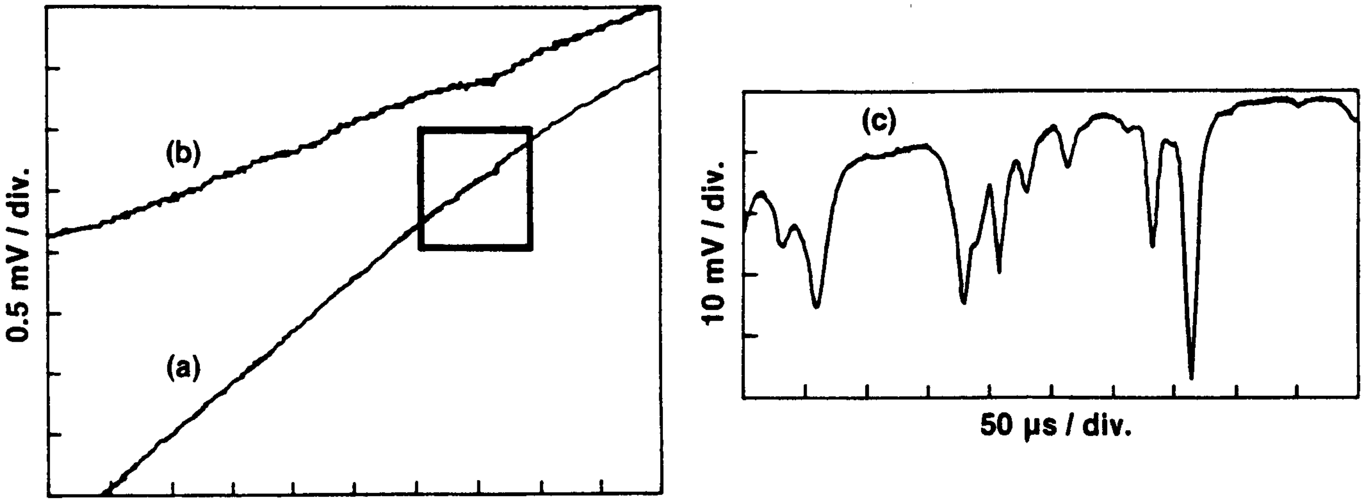

Figure 5. Iodine vapor absorption peaks in a 10-cm path cell. (a) Direct absorption signal from the linear output with reference beam blocked. (b) Expansion of the marked section of trace (a) by 5× horizontally and 2× vertically, showing two small absorption peaks. (c) Absorption signal from the log output with the reference beam open. The noise canceller loop bandwidth for these traces was about 70 kHz.

To establish that the observed peaks were I_2 absorptions, we heated the cell, increasing the I_2 pressure through sublimation. The absorption strengths grew by an order of magnitude when the cell was heated, and declined again when the cell was cooled. The shapes and relative positions of the peaks did not change measurably, nor did any appear or disappear. We did not make precise laser frequency measurements for this preliminary spectrum. The estimated values of the center wavelength and scan width were checked indirectly by comparing the pattern of spectral peaks in trace (c) with the G&L survey spectrum which, as mentioned above, shows the strongest lines near 670 nm to be about 1% in a 10-cm path. We may confirm the same peak absorption strength for our spectra in two ways. First, the largest absorption peak on trace (b) can be estimated directly; it has a peak height of 42±15 mV. The dc photovoltage of the signal beam at this point in the spectral scan is 4.75 V, so the absorption is 0.008±0.003. Second, we can compute the absorption strength for the large peak in trace (c) from Eq. 1. If the log output voltage in the absence of absorption is V_o , and that with absorption present is V, the normalized transmittance I/I_o is

$$\frac{I}{I_o} = \frac{(e^{-V_o} + 1)}{(e^{-V} + 1)} . \qquad (3)$$

The *average* log output voltage during the scan of trace (c) (as measured by a digital voltmeter) was 0.977 V, which we take as V_o. The largest peak in trace (c) is about 40 mV high, so $V = 0.977 - 0.040 = 0.937$ V. From Eq. 3, we obtain a transmittance of 0.989, or an absorption of 0.011. Owing to the absorption peaks, the baseline voltage is a little higher than the average voltage; but, even if this shift were as large as the biggest peak, 40 mV, recalculating the absorption using $V_o = 1.017$ V changes the calculated absorption by only 0.0003.

5.2 Refinements

The noise in trace (c) is primarily quantization noise from the 8-bit digitizer. To get a detectable absorption peak and baseline noise that is due to the shot noise on the same scale, we used the 1-mm path length cell to reduce the peak absorption to around 100 ppm, the Ithaco preamplifier to narrow the analog bandwidth and boost the absorption signal to use the 8-bit resolution of the storage oscilloscope efficiently, and signal averaging to further narrow the effective bandwidth, bringing the absorption peak out of the shot noise. For a scan as wide as the one in Figure 5, the power variation in the laser was also too large for the noise canceller board to completely remove, hence the sloping background in trace (c). It should be noted that the amplitude of the power

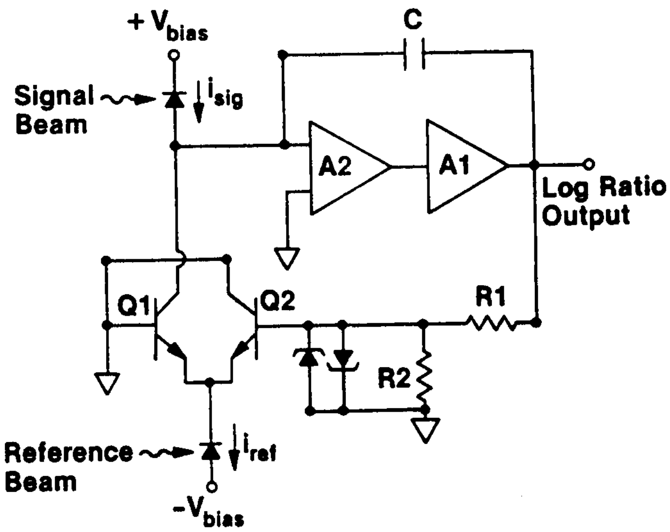

Figure 6. Schematic diagram of the modified laser noise canceller (see text).

variation in the signal beam was 3.9 V, while the residual background variation was 12 mV in the suppressed spectrum, so the circuit suppressed this variation by 50 dB, about the limit of its performance.

To obtain the shot noise-limited spectra shown below, the noise canceller board was modified, as shown in Figure 6. In the modified circuit, the linear output has been eliminated, so the reference and signal currents can be subtracted directly at the input of the log output integrating amplifier A2. These modifications improve the cancellation and allow the use of wider loop bandwidths, compared with the original circuit in Figure 1, without changing the essentials of its operation. Narrowing the laser wavelength scan was still necessary, not just to remove the sloping background artifact by bringing the intensity variation down to a level the noise canceller could handle, but to narrow the analog bandwidth of spectral information contained in the scan as well.

5.3 Shot noise-limited spectroscopy

The data of Figure 7 were taken at a scan rate of 493 Hz with scan limits of 63.57 mA ± 56 μA and a laser temperature of 24.1 °C. Trace (b) is slightly less than one half-cycle of the current scan, showing two closely spaced I_2 lines, and was taken when the 1-mm path length cell was still warm after using a heat gun to vaporize the I_2 in the cell. The preamplifier gain G was 5000, with its one-pole high and low pass filters set to 100 Hz and 30 kHz (3-dB points), respectively, corresponding to an analog noise bandwidth of 47 kHz. The trace is an average of 1000 individual sweeps, so the equivalent noise bandwidth for the measurement was 47 Hz. Trace (a) was taken under the same conditions, after the 1-mm cell had cooled to room temperature. The traces have been plotted on different scales to be roughly the same height; apart from the the increased noise in the room-temperature trace, they overlay almost exactly in trace (c), showing that, in this case, there are no residual interference fringes or other artifacts that distort the absorption peak shape.

The peak height of the large peak in trace (b) is 251 mV, while that in trace (a) is 98 mV. With the addition of the preamplifier and the circuit change, Eq. 1. becomes

$$V_{\log} = \frac{G}{10} \ln\left(\frac{I_{ref}}{I_{sig}} - 1\right). \tag{4}$$

G is the gain of the preamplifier, and the factors of 10 arise from a change in the voltage divider ratio (R1 and R2) at the output of the integrator; the sign changes follow from the elimination of an inverting amplifier stage.

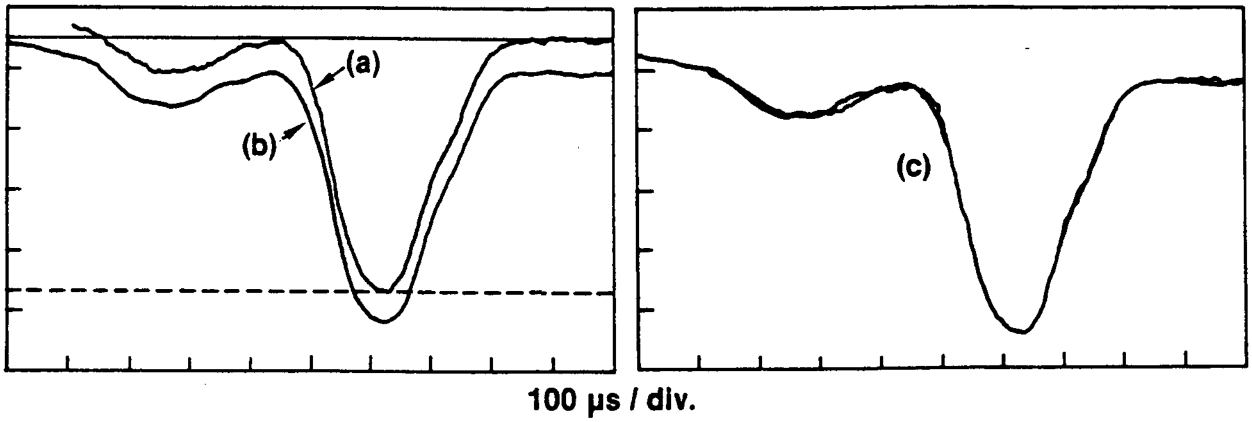

Figure 7. **Iodine vapor absorption peaks in a 1-mm path cell. (a) Cell at room temperature. (b) Cell warmed with a heat gun to vaporize the I_2 charge. (c) Superposition of (a) and (b). The horizontal cursor lines (solid and dashed) in the top panel correspond to a voltage of 98 mV for trace (a) and 251 mV for (b). The modified noise canceller loop bandwidth was greater than 100 kHz.**

(Compare Figure 1 with Figure 6.) The absorption peaks appear to be negative going in Figure 7 because we used the inverting preamplifier input for consistency with the earlier results. For this version of the laser noise canceller board, the normalized transmittance is similar to Eq. 3.

$$\frac{I}{I_o} = \frac{(e^{10V_o/G} + 1)}{(e^{10V/G} + 1)} . \tag{5}$$

The average value of the log output during the scan in Figure 7 was 0.00 V. This was a bit wasteful, since the reference beam was twice as intense as the signal beam (see Eq. 4); in principle, the reference beam could have been only slightly more intense than the signal beam. Nevertheless, the generous intensity ratio provided a safety margin for photocurrent subdivision and simplified the forthcoming calculations. Taking $V_o = 0$ and expanding the exponential in the denominator under the assumption that $10V/G \ll 1$, the fractional absorption of the signal beam is approximately

$$a = 1 - \frac{I}{I_o} = 5\frac{V}{G} , \tag{6}$$

where V is in Volts. So, with the factor of 5000 in gain, peak heights of 251 and 98 mV correspond to absorptions of 251 and 98 ppm, respectively.

The wavelength of the laser was measured by removing the noise canceller from its kinematic mount, allowing the signal beam to be directed into the wavemeter, and holding the bias current constant for a few seconds at the endpoints of the scan. At 64.13 mA, the wavelength was 14878.88 cm^{-1} and, at 63.01 mA, the wavelength was 14879.07 cm^{-1}. These wavelength endpoints were stable, and were checked before and after the Figure 7 traces were taken. This positively identifies the large peak in Figure 7 as G&L peak number 163, measured as 14878.9822 cm^{-1}. Since we made no attempt to calibrate the frequency scale in our measurements other than this bracketing procedure, or account for instrumental broadenings of the line shape, we cannot improve on the G&L value for the position of this line. The peak absorption of the line was 12% in a 200-cm path length cell at room temperature, which corresponds to an absorption of 64 ppm in a 1-mm path, which agrees well with our 98 ppm value, considering that the cell temperature was not controlled to be exactly the same as in G&L; nor was our path length measured precisely. The smaller peak in Figure 7, while not identified by G&L (it is below their stated sensitivity limit), is certainly an even weaker I_2 peak, since it changed proportionately with the large peak upon heating and cooling the cell. Its peak height is about 12 mV, so the absorption of this peak is 12 ppm, and the S/N ratio is still excellent.

To calculate the expected S/N ratio and compare it with our data, we need to find the RMS fluctuations of the log output voltage V_{log} due to shot noise. From Eq. 4, the sensitivity of V_{log} to i_{sig} is

$$\frac{dV_{log}}{di_{sig}} = \frac{G}{10}\left(1 + e^{-10V_{log}/G}\right)\frac{1}{i_{sig}}. \tag{7}$$

As mentioned above, since the noise canceller in effect subtracts *two* copies of i_{sig}, the variance of the log output voltage will be twice that contributed by the signal photocurrent. So, the RMS noise density in the log output voltage is given by

$$\sigma_{V_{log}} = \sqrt{2}\,\frac{dV_{log}}{di_{sig}}\,\sigma_{i_{sig}}, \tag{8}$$

which, by combining Eqs. 2 and 7 into Eq. 8 is

$$\sigma_{V_{log}} = \frac{G}{5}\left(1 + e^{-10V_{log}/G}\right)\sqrt{\frac{q}{i_{sig}}}. \tag{9}$$

Since the average signal beam power during the scan in Figure 7 was 1.20 mW, the average photocurrent was 360 μA and, by Eq. 9, the RMS shot noise density in the log output voltage when $V_{log} = 0.00$ V should be $\sigma_{V_{log}} = 42\ \mu V/\sqrt{Hz}$. In the 47-Hz equivalent bandwidth of the spectra in Figure 7, the RMS noise voltage due only to shot noise would be $\sqrt{47}$ times higher: 290 μV. By the rule of thumb stating that the peak-to-peak excursions in the baseline should be about 5 times the RMS noise,[12] we would therefore expect the noise in the baseline of trace (a) in Figure 7 to be about 1.4 mV. Inspection of the trace shows that this is indeed the case.

A better comparison can be made by using a spectrum analyzer to look at the repetitive absorption signal waveform. If our spectral signal is shot noise-limited, it consists of the sweep frequency and its harmonics plus a white noise floor 3 dB above the shot noise of the signal-beam photocurrent. Figure 8 shows the PSD of the absorption signal that produced Figure 7, measured with a Hewlett-Packard 3562A dynamic signal analyzer. Conditions were the same as in Figure 7; the output of the preamplifier was simply connected to the analyzer. As predicted above, the signal in the frequency domain consists of several harmonics of the wavelength sweep frequency (493 Hz), riding on an essentially flat, white noise distribution. Trace (a) extended only to 12.8 kHz, but there were essentially no higher harmonics present in the spectrum. Trace (b) has a narrower span. Several spurious peaks at harmonics of 60 Hz are present, due to low-level room lights and pickup. The noise in the system is dominated by the white noise density, which is about $-85.2\ dBV/\sqrt{Hz}$. (0 dBV = 1 V RMS.) It turned out that this level was close to the noise floor of the preamplifier alone which, in trace (c) (taken with the preamplifier input grounded), is seen to be -90.0 $dBV/\sqrt{Hz}$. The noise of the log output alone is therefore -86.9 $dBV/\sqrt{Hz}$. (That is, the measured noise density is the RMS sum of the log output and preamplifier noise floors.) From above, the calculated shot noise floor is $42\ \mu V/\sqrt{Hz}$, or $-87.5\ dBV/\sqrt{Hz}$, which is in excellent agreement with the data; the 0.6-dB discrepancy is well within the baseline fluctuations of the analyzer traces.

The S/N ratio of the peak absorptions in Figure 7 has a simple form when the average $V_{log} = 0.00$ V. Putting the peak signal voltage V in Eq. 6 in terms of the fractional absorption, and dividing by the RMS shot noise voltage in Eq. 9, we obtain

$$S/N\ (dB) = 20 \log \frac{a}{2}\sqrt{\frac{i_{sig}}{q\Delta f}}, \tag{10}$$

where we have explicitly included the equivalent measurement bandwidth, Δf. For the small $a = 12$ ppm peak, the S/N ratio should be ideally 32.4 dB. Using the measured peak voltage and corrected noise density, namely

$V = 12$ mV and $\sigma_{V_{\log}} = -86.9\ \mathrm{dBV}/\sqrt{\mathrm{Hz}}$, which in a 47-Hz bandwidth corresponds to 310 μV RMS, the experimental S/N ratio is 31.8 dB.

We certainly could have measured much smaller absorptions with this system, had we used smaller path length cells or cryogenic temperature control of the cell. The noise-equivalent absorption is predicted by Eq. 10 when the S/N ratio falls to 0 dB:

$$a_{ne} = \sqrt{\frac{4q\Delta f}{i_{sig}}}. \tag{11}$$

Eq. 11 could have been motivated heuristically, as it is simply $\sqrt{2}$ times the shot-noise-to-signal ratio of the signal beam photocurrent. Under the conditions of Figure 7, $a_{ne} = 2.9 \times 10^{-7}$. In the absence of systematic errors, even such a small peak would be easy to measure by narrowing the bandwidth further, say to 1 Hz, in which case $a_{ne} = 4.2 \times 10^{-8}$, and the S/N ratio would be 16.4 dB.

Although, in this preliminary work, we did not measure peak absorptions all the way down to the limits of detectability, we believe that the measurement and analysis of the spectral signals and noise, and their agreement with the expected shot noise limits are quite conclusive. It should be emphasized that no fitting procedure has been performed in producing this agreement; only a simple calculation, based on the average laser power, fundamental constants, and a small correction for additive electronic noise that could easily be eliminated by the use of a better preamplifier, is needed to completely account for the noise performance of the system.

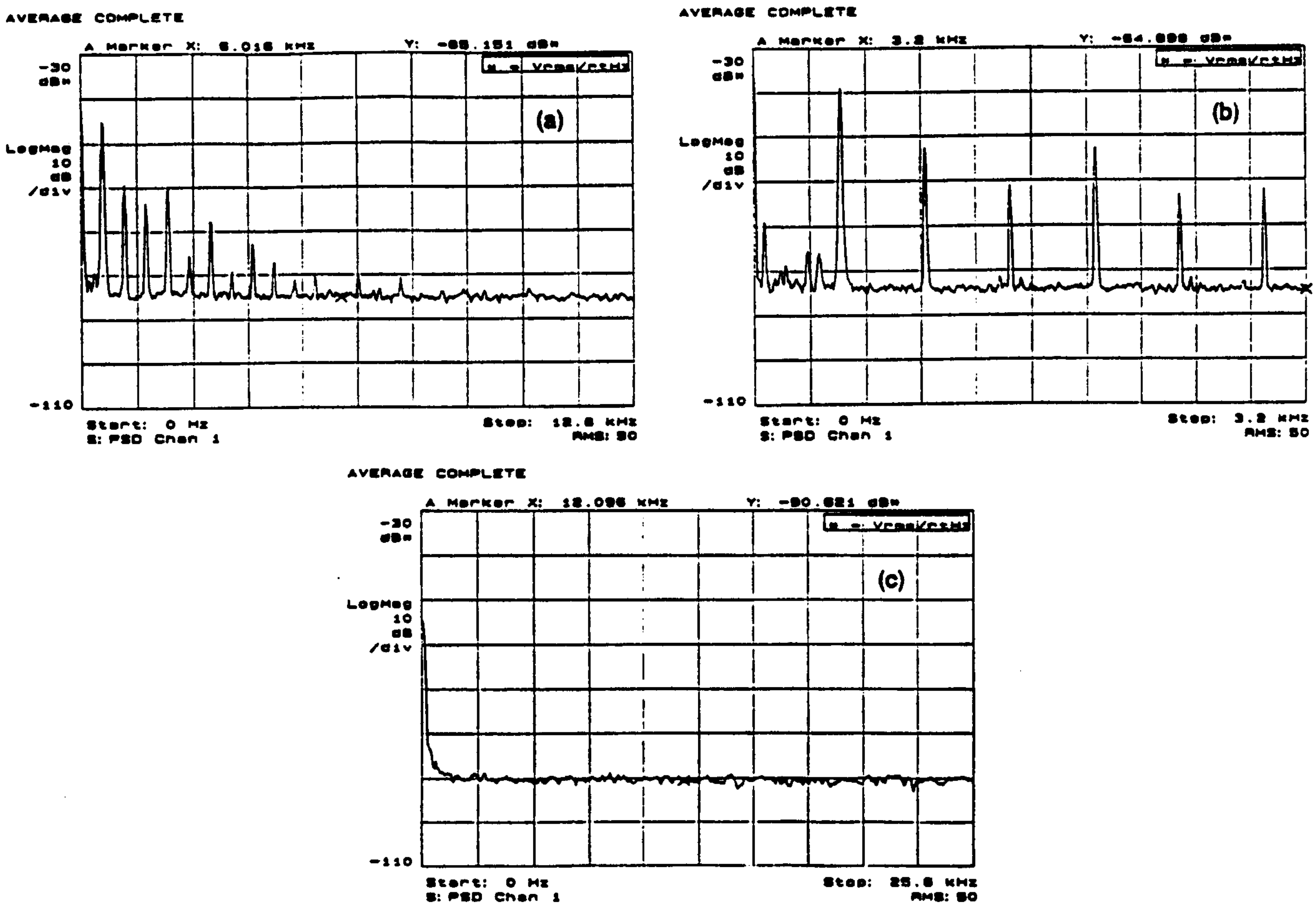

Figure 8. PSD measurements of the repetitive absorption waveform.

There are many problems common to ultrasensitive spectroscopic systems which affected ours as well, including spurious Fabry-Perot fringes, difficulty finding spectral operating regions with reasonably flat backgrounds, and development of the skill to take measurements quickly so that drift in the optical alignment was not a problem. Our setup is far from ideal. The 1-mm absorption cell was mounted in the signal beam with a three-fingered clamp, which exhibited severe position drift. Even with ultra-stable optical equipment, one might imagine that there may always be some interesting spectral region that suffers from residual interference fringes, or a laser amplitude modulation that is simply too strong for the noise canceller circuit to eliminate completely. In these cases, background subtraction will be necessary.

6. CONCLUDING REMARKS

Shot noise-limited spectroscopy is not itself new, nor does the laser noise canceller solve all the generic technical barriers to achieving it. Although the major hurdle, laser noise, has been removed, baseline features due to interferences, drift, and linearization of the frequency scale, among other things, remain troublesome. But use of the noise canceller simplifies the experimental apparatus sufficiently so that investigators can spend more effort on eliminating these residual sources of systematic error. The circuit itself is robust and uncomplicated in both design and operation, and a serviceable version can be assembled from readily available components costing on the order of 10 dollars. With this simple addition to the experimenter's arsenal, great simplification in signal processing and optical-system design is possible for spectroscopic and other laser-based ultrasensitive detection methods, a claim which we feel is amply demonstrated by our achieving the shot noise limit with such a primitive apparatus.

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Supplementary information for "Double beam laser absorption spectroscopy..." by Kurt L. Haller and Philip C.D. Hobbs

Table 1. Components for the laser noise canceller. (cf. Figure 1)		
Component	**Part number or value**	**Manufacturer***
A1, A2 op amps	OP-270	PMI, Motorola
Q1, Q2 matched BJT array	MAT04FP	PMI
Q3 cascode transistor	2N3906	Motorola, many others
R integrating amp resistor	2.1 kΩ	Ohmite Metal Devil
R_f transimpedance amp resistor	20 kΩ	"
R1 voltage divider resistor	1 kΩ	"
R2 voltage divider resistor	25 Ω	"
C integrating amp capacitor	1 nF	NPO ceramic (teflon, polystyrene OK too)
Si PIN photodiodes	BPW34	Siemens

Table 2. Components for the modified laser noise canceller. (cf. Figure 6)		
Component	**Part number or value**	**Manufacturer***
A1 booster amp	LM6321N	National S/C
A2 op amp	AD744	Analog Devices
Q1, Q2 matched BJT array	MAT04FP	PMI
R1 voltage divider resistor	301 Ω	Ohmite Metal Devil
R2 voltage divider resistor	100 Ω	"
C integrating amp capacitor	50 pF	NPO ceramic (teflon, polystyrene OK too)
Schottky clamp diodes	NBD101	Motorola
Si PIN photodiodes	BPW34	Siemens

*Identification of components by manufacturers' names and part numbers is done solely to describe experimental conditions and does not imply endorsement by the International Business Machines Corporation nor does it imply that the particular products are necessarily the best or only such products for the intended purpose.