Long Path-Length Spectroscopy of O_2 Using the *NICE-OHMS* Technique

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ABSTRACT

We demonstrate high sensitivity detection in the weak magnetic-dipole band of molecular oxygen near 764 nm, using a high-finesse Fabry-Perot cavity to provide an equivalent path length of 980 meters.

Keywords: Oxygen, Molecular spectroscopy, Fabry-Perot, Optical cavity, diode laser

1. INTRODUCTION

Low concentrations of many atomic and molecular species are routinely measured using laser absorption instrumentation. Spectroscopic methods designed to avoid laser technical noise and spurious interference fringes can sometimes attain a sensitivity equal to or even slightly less than the shot noise fluctuations of the detected photo-current. For instance, a sensitivity to absorptions as small as $6 \cdot 10^{-10}$ cm^{-1} is possible with a 30 cm long path length and 1 mA of photo-current, if the shot noise level can be attained. More laser power could increase the sensitivity, but only until the photo-detector approaches saturation. In some cases higher sensitivity can be achieved by the use of stronger absorption lines, or sample pre-concentration. However, significant decreases of the minimum detectable absorption are likely to come only from drastically increasing the absorption path length. Here we discuss our progress toward more sensitive measurements by realizing long absorption path lengths.

Although path-lengths on the order of 10^2 meters are possible with multi-pass cells, longer effective path lengths ($\geq$ 1 km) in rather compact instruments are now possible by utilizing resonant optical cavities. Recently, measurements of extremely low absorption ($\approx 10^{-14}$ cm^{-1}) were demonstrated using a high-finesse cavity and a radio-frequency (RF) side-band technique, *NICE-OHMS.*[1] The acronym stands for noise immune, cavity-enhanced, optical heterodyne molecular spectroscopy. The technique is inherently continuous wave, as opposed to another measurement technique that utilizes resonant cavities, Cavity Ring-Down Spectroscopy.[2]

Our motivation is the sensitive detection of certain non-allowed transitions in the magnetic dipole "A Band" of molecular oxygen at 764 nm. An absorption signal at these transition wavelengths may have significance in terms of the quantum mechanical symmetrization postulate.[3,4] For these proceedings, we will emphasize the measurement technique, since it has the potential to be applied to other applications of interest.

2. EXPERIMENTAL APPROACH

The non-allowed absorption lines near 764 nm that we are searching for have a center frequency known accurately to about 1 GHz. The transitions would be Doppler broadened with a width of about 700 MHz. To improve on previous measurements, our minimum detectable absorption needs to be $< 10^{-12}$ cm^{-1}. One possible approach would be to frequency lock a laser to a high-finesse cavity, and monitor the transmitted power as the cavity mode is tuned through the wavelength region of interest. In addition to the center frequency sweep, a dither of the cavity frequency would allow phase sensitive detection of the transmitted power with a lock-in amplifier. The technical challeges to overcome would likely be laser intensity noise, the residual FM fluctuations of the laser in relation to the high-finesse cavity resonance, and spurious etalon fringes. A short discussion of this experimental approach is warranted because it highlights a technical advantage of the *NICE-OHMS* method.

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In such a system where the transmitted power is demodulated at the dither frequency, laser intensity noise at low Fourier frequencies may be an issue because of the cavity dither rate. The dither rate of the cavity frequency is constrained by the finite bandwidth of the feedback loop that locks the laser to the cavity mode. Thus it is difficult to use high dither frequencies. However, the laser intensity fluctuations inherent at low frequencies may be greatly reduced before coupling the laser to the cavity by an external intensity control, such as an acousto-optic modulator.

The residual FM fluctuations of the laser in relation to the high-finesse cavity resonance will be converted by the cavity to intensity noise. This FM→AM conversion noise will likely dominate the noise due to the laser intensity fluctuations, depending on the gain profile and noise floor of the frequency-locking feedback loop. With a properly designed feedback system, the residual frequency fluctuations at low frequencies may be suppressed to a level that is limited only by the shot-noise of the lock detector photo-current. However, this is often difficult to achieve experimentally, and depending on the type of laser and cavity linewidth may even require pre-stabilizing the laser before locking to the high-finesse cavity.

In our present experiment, the "noise immune" aspect of the *NICE-OHMS* approach refers to a reduced sensitivity to the residual frequency fluctuations between the laser and the cavity. Using an electro-optic phase modulator, side-bands are impressed on the laser beam at precisely the cavity free-spectral-range frequency, f_{FSR}. When the laser is frequency locked to a cavity mode, these side-bands are transmitted through the adjoining cavity modes that are one free-spectral-range distant in frequency. All three spectral components of the cavity transmitted power are subsequently detected with a high-speed photo-detector. As with optical heterodyne laser spectroscopy without a high-finesse cavity,[5] a phase shift of any of the three spectral components, or a difference of intensity of the two side-bands will cause a signal at f_{FSR} in the detected photo-current. This signal is usually demodulated down to DC by a balanced mixer. Because our cavity free spectral range (568 MHz with our present cavity) is similar in magnitude to the width of the absorption signals, the phase of the local oscillator is adjusted so that the maximum signal corresponds to an absorption of one of the side-bands.

A dispersion-like signal is observed on the low-pass filtered mixer output when the cavity mode is scanned over an oxygen absorption line. The width of the observed signal is a function of both the transition linewidth and the cavity free spectral range. Baseline changes of this signal (for instance due to changes in the residual amplitude modulation of the crystal modulator) can be suppressed by a dither of the cavity along with the wavelength scan. We typically scan the cavity wavelength at a 0.2 Hz rate, while dithering at approximately 50 Hz. The dither amplitude is set to provide the maximum signal without significant modulation broadening (a dither of about 500 MHz, peak-to-peak). The signal output is then a second-derivative type shape familiar in wavelength modulation spectroscopy.

The system diagram is shown in figure 1. There are three servo-loops which are referred to in the diagram; One loop locks the laser to the high-finesse cavity using the Pound-Drever-Hall method, with a modulation frequency of 14 MHz. The second loop is used to lock an oscillator providing the f_{FSR} signal precisely to the cavity free-spectral-range. The third loop controls the intensity of the light incident on the cavity. A discussion of the characteristics of each of these loops is included in the following sections.

To observe the oxygen absorption lines, the high-finesse cavity is required to scan about 10 GHz. This was accomplished by mounting one of the mirrors on three piezo-electric stacks. The stacks are arranged in a 120° fashion around the mirror, approximately 3 cm from the mirror center. Each stack is capable of 8 μm of motion with 150 V, and they are driven in parallel. One mirror is flat, the second has a radius of 100 cm and they are spaced by 26.4 cm. The cavity has a finesse of 5800, which was accurately measured by observing a weak oxygen absorption line with a known cross-section. The O_2 pressure in the cavity was adjusted to result in a loss much less than the loss due to the mirrors (mirror's $R \approx 99.945\%$). The observed transmission dip when the system was tuned over the absorption profile corresponded to an effective path length, L, of 980 meters. This is related to the finesse F by $L = (2/\pi)\cdot F\, l$, where l is the cavity mirror's spacing.

2.1 The Laser and frequency Control

The 764 nm beam is produced by an extended cavity diode laser (ECDL) built in a Littrow configuration, and gives approximately 15 mW of output power in a single longitudinal mode. Smooth continuous scans over as much as 25 GHz are accomplished by moving the ECDL mirror with two piezo-electric actuators. The piezos are arranged so that one causes a mirror translation towards the grating, while the second primarily causes a mirror angle change. In order to synchronously turn the mirror angle and shift the laser cavity length, two high-voltage amplifiers with the proper gain ratio are used to drive the piezos. Furthermore, the voltage signal used for tuning was also summed to the laser injection current, which allowed wider, more stable wavelength sweeps, although at the expense of a power change during the sweep.

An error signal is derived from the cavity reflection, using the Pound-Drever-Hall method. The resulting signal is integrated and fed back to the injection current to correct the laser frequency and lock to the cavity. This "fast" loop has a

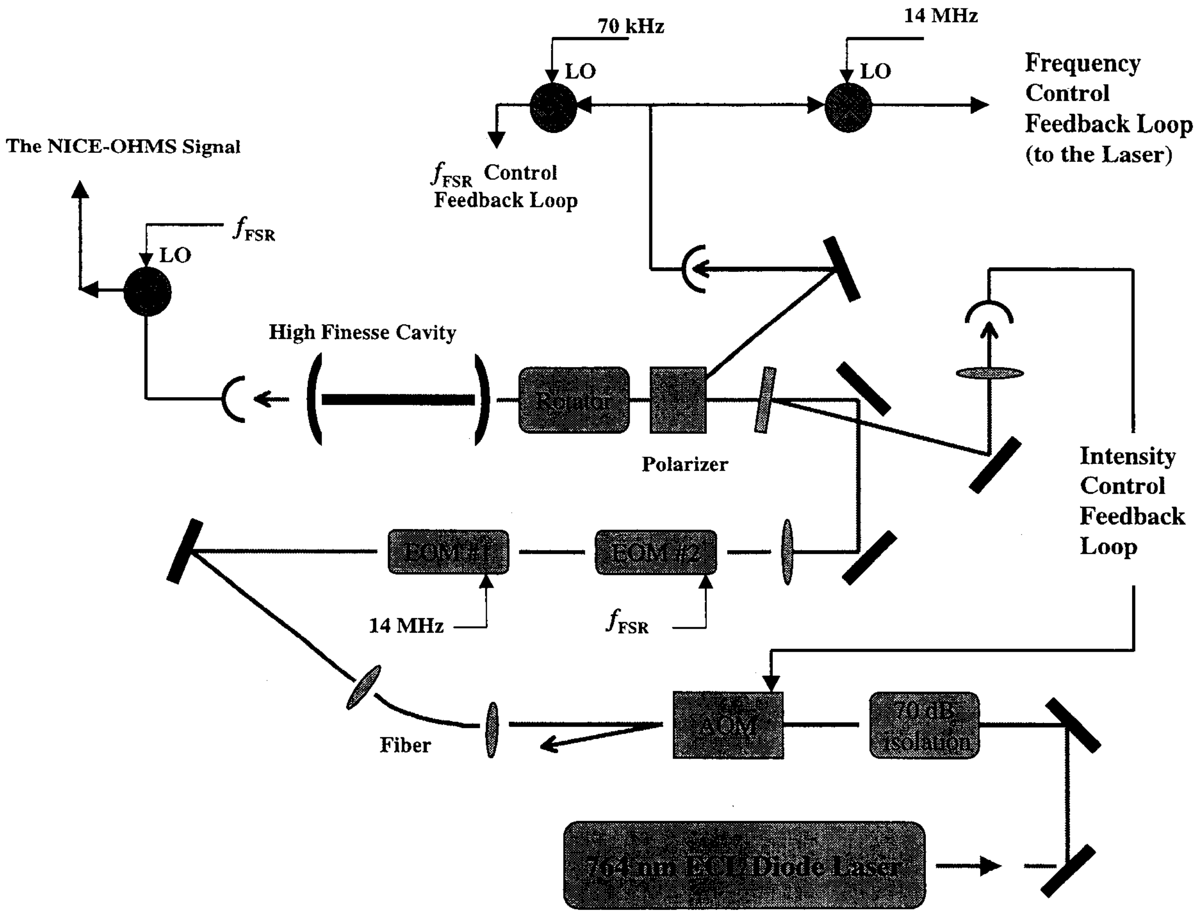

Figure 1. The system diagram. Electro-optical modulator #1 produces sidebands used to lock the laser to the cavity, and modulator #2 is used to produce sidebands at the cavity free-spectral-range. The laser beam is sampled prior to the cavity with a beamsplitter in order to drive an acousto-optic modulator (AOM) and control the intensity incident on the cavity. The high-finesse cavity is actually in a vacuum chamber (not shown) with Brewster-cut entrance and exit windows.

bandwidth of approximately 1.5 MHz. The integrator gain is leveled off at 100 kHz in order to compensate for the loop phase shift due to the 100 kHz wide optical cavity. The correction voltage is also integrated with a slower loop and fed back to adjust the piezos (and dc injection current level) so that the fast loop correction voltage remains near zero.

Both the fast and the slow loops are of the same basic design, two inverting operational amplifiers arranged to provide high gain at low frequencies to the servo feedback gain profile. This is achieved by connecting the output of one of the amplifiers to the positive input of the second, as shown in Figure 2.

Changes of the ECL internal alignment were occasionally necessary when the laser wavelength was tuned to various oxygen lines. A short section of single-mode optical fiber used in the beam path (see Fig. 1) facilitated the alignment between the laser and the cavity, and also served as a spatial filter. The fiber ends were polished at a slight angle to avoid interference fringes.

2.2 Side-bands at f_{FSR}

A resonant optical phase modulator was constructed using a 50 mm long anti-reflection coated electro-optic crystal. The modulator design is a simple LC tank circuit, with the inductance provided by a copper loop.[6] The Q-factor is about 100, resulting in side-bands at f_{FSR} with an intensity of approximately 10% of the carrier with 300 mW of RF power.

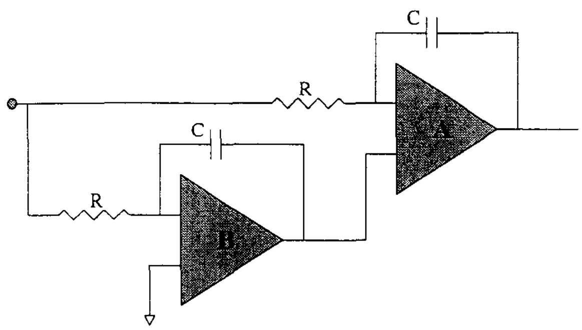

Figure 2. The simple op-amp circuit that is used to provide a higher electronic gain than available from a single integrator. The RC product of Amp B is adjusted to be some 8 to 10 times larger than that of Amp A.

Any deviation of the RF frequency from the cavity free-spectral-range, even with no inter-cavity absorption present, produces a modulation on the power transmitted from the cavity. This of course must be minimized, since it represents a baseline change and/or noise in the signal. The change in the cavity's free-spectral-range as the cavity length is tuned amounts to 1.5 kHz per GHz of optical frequency shift. This change, plus the RF source's frequency fluctuations and thermal drift can be corrected for by locking the source frequency to the cavity free spectral range. A simple method to accomplish this is to note that there is a minimum in the reflected power from the cavity when the side-bands are exactly at f_{FSR} . To lock the RF to the free spectral range, we dither the RF source at a moderate rate (70 kHz), and synchronously detect the reflected power in order to derive an error signal.[6] This dither rate, and the subsequent feedback loop bandwidth, are limited by the modulation characteristics of the commercial RF signal generator. The loop bandwidth is approximately 10 kHz, and is constructed with the same basic design as shown in Figure 2 in order to have high gain at our signal dither rate (approximately 50 Hz).

2.3 Intensity Noise reduction

Unfortunately, the free-running intensity noise of our extended cavity diode laser is increased when the laser is locked to the high-finesse cavity. This is due to the electronic feedback to the injection current, which modulates the laser power within the bandwidth of the lock-loop.[7]

The absorption signals detected in this *NICE-OHMS* experiment are of course proportional to the laser power incident on the photo-detector (e.g., as in the technique of wavelength modulation spectroscopy). Ideally, the noise background is not appreciably affected by power fluctuations, since these fluctuations change the shot noise floor only slightly. However, in practice the phase modulator is also a source of residual amplitude noise on the laser beam at the modulation frequency. Power fluctuations will modulate this residual AM signal, causing additional noise. These low frequency laser power fluctuations can be reduced by the use of an acousto-optic modulator. A fraction of the light incident on the cavity is detected, as shown in Fig. 1, and used in a control loop to diffract excess light away from the primary experiment beam.

3.0 OXYGEN SPECTROSCOPY

To investigate a wavelength region, we first tune the laser to the desired wavelength with the aid of a six digit wavemeter. A resonance of the high-finesse cavity is then tuned to the same wavelength, and the gain of the locking electronics switched on. The cavity may then be scanned and the *NICE-OHMS* signal recorded by a computer controlled analog-to-digital board. An example is shown in Fig. 3.

As previously discussed, we are building a spectrometer with a sensitivity goal of at least 10^{-12} cm^{-1}. If the shot noise level of sensitivity can be attained, the power transmitted from the cavity (≈ 450 μW) is sufficient to reach this goal in a 1 s averaging time, with the cavity mirrors we now have. However we are presently limited by spurious interference fringes, and the

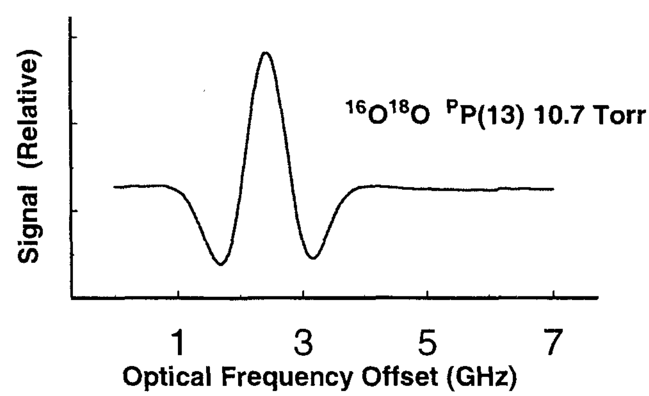

Figure 3. An oxygen absorption line measured with the *NICE-OHMS* method. This is an allowed transition of $^{16}O^{18}O$ at 764.475 nm, and represents about 1% absorption in the 980 M path-length.

detection limit is about one order of magnitude higher than our goal. Continuing efforts to reduce the interference effects and increase the sensitivity include vibrating several of the system components and averaging.

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