Ultrasensitive Detections of Weak Resonances
– Application to Optical Frequency Standards

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We present our latest results on the ultrasensitive molecular overtone spectroscopy using the cavity-enhanced frequency modulation (FM) technique. The principle of this technique makes use of a high-finesse external cavity to enhance the intrinsic resonance contrast, while an FM modulation approach provides shot-noise limited signal recovery. Working with a 1.064 μm Nd:YAG laser, we have obtained sub-Doppler overtone resonances of C_2HD, C_2H_2 and CO_2 molecules. A detection sensitivity of 5 $\times 10^{-13}$ of integrated absorption (1 x 10^{-14} / cm) over 1-s averaging time has been achieved. The resultant high signal-to-noise ratio of the weak resonance produces excellent laser frequency stabilization.

High detection sensitivities of quantum absorptions are desirable in many research fields of fundamental physics and molecular dynamics, as well as in application areas such as target gas tracing and remote sensing. We seek a high sensitivity to use in developing visible frequency standards based on a rich spectrum of attractive and yet extremely weak molecular overtone transitions. To achieve the goal of ultrasensitive detection, it is apparent that we need to enhance the intrinsic resonance signal (contrast), and reduce the background noise, ideally to the fundamental quantum limit.

An optical cavity is placed around the quantum absorber to extend the effective interaction length between the field and matter,[1] leading to enhancement of the resonance contrast. From another view-point, for a given noise generated by transmitted dc light on the detector, the high internal build-up field of the cavity elicits a strong molecular radiation, which is resonantly coupled out for detection. The cavity power buildup permits sub-Doppler saturation spectroscopy.[2,3] In addition, the geometrical self-cleaning and matching of the two counter-propagating waves inside the cavity are important both for eliminating pointing-direction-related noises and for obtaining narrow and unshifted resonance lines.[4] In another role, a stable cavity can be used to pre-stabilize the laser frequency to reduce the detection noise. For the purpose of establishing a stable and repeatable optical frequency reference, it is also attractive to locate the absorber external to the laser to avoid various coupled instability problems associated with intra-laser-cavity absorption.

For an optimum signal recovery, this cavity-enhanced absorption contrast should be probed while avoiding various technical noises associated with the high finesse cavity and dc light field. The so-called "cavity-ring-down spectroscopy"[5] takes advantage of the cavity memory to avoid laser technical noise. It measures the cavity field decay time (which is a function of the intracavity absorption) once the input laser field is switched off. Another effective technique for avoiding the light noise at DC is to lock the laser frequency tightly on the corresponding cavity resonance and then

modulate the cavity mode around the desired molecular resonance while monitoring the cavity transmission. Essentially this approach is a simple lock-in derivative-lineshape modulation-recovery process. In order for this method to be successful, it is important to have a super-tight frequency lock loop between the laser and the cavity, since any laser frequency noise relative to the cavity will be converted to amplitude noise in detection. The cavity dither frequency is usually limited at the audio range due to the mechanical resonance and roll-off of the mirror assembly. This modulation frequency may be too low to reach the shot-noise-limited spectral region of the laser. Higher modulation frequencies are of course often used in frequency modulation (FM) heterodyne spectroscopy to achieve shot-noise limited performance. To combine the use of a high finesse cavity with the FM technique, we phase-modulate the laser beam at the cavity free-spectral-range to avoid the cavity bandwidth limit.[6,7] The biggest advantage of this configuration lies in its noise immune property relative to the laser frequency jitter. The pure FM signal is preserved by an empty cavity and a null (shot-noise) background is detected in cavity transmission, independent of the laser/cavity lock. If, for example, the central FM component is tuned onto a saturated molecular resonance and it is frequency pulled due to the intracavity molecular dispersion, then a pure dispersion signal will result in the rf beat of the cavity transmitted light after phase sensitive demodulation.

   This technique, named as the noise-immune cavity-enhanced optical heterodyne molecular spectroscopy ("NICE-OHMS"), enables profitable use of the high cavity finesse without a noise penalty and has produced remarkable detection sensitivities. The ultimate sensitivity of this approach can be illustrated by comparing with the case of direct absorption, where the maximum sensitivity condition will arise if the probe field has no amplitude noise beyond its intrinsic quantum fluctuation. Given a detection bandwidth of B, the minimum detectable absorption signal under this shot-noise limit is

$$(\alpha L)_{\mathrm{min}} = \left(\frac{2eB}{\eta P_0} \right)^{1/2} , \qquad (1)$$

where e is the electron charge, η is the photodetector responsivity in Amp/Watt, and P_0 is the incident radiation power. This sensitivity can be as high as 1 x 10^{-8} at 1 s averaging for $P_0 = 1$ mW and a reasonable η (~ 0.8 Amps/Watt at 1.064 mm). Due to practical limits, this sensitivity is usually degraded by several orders. FM spectroscopy sacrifices a small signal loss while truly reaching the shot noise level. Therefore the FM sensitivity is basically that of Equation (1), multiplied by a factor of $\sqrt{2} / \left(J_0(\beta) J_1(\beta) \right)$. Here J_0 (J_1) is the zero (first) order Bessel function and β is the modulation index. The NICE-OHMS technique retains the shot-noise background from FM while the signal itself is enhanced by a factor of $(2 \cdot \mathrm{Finesse}/\pi)$ from the optical resonator. The minimum detectable absorption signal of NICE-OHMS is thus

$$(\alpha L)_{\min} = \frac{\pi}{2 \cdot Finesse} \left(\frac{2eB}{\eta P_0} \right)^{1/2} \frac{\sqrt{2}}{J_0(\beta)J_1(\beta)} \; . \tag{2}$$

Hence with a cavity finesse of 100,000, it is possible to reach a sensitivity in the 10^{-12} − 10^{-13} range with 1-s averaging time.

The experimental setup is described elsewhere.[7] First we show the noise-immune detection of the C_2HD ($v_2 + 3v_3$) P(5) transition at 1.064 μm. To demonstrate the noise-immune nature, we compare the recovered signal-to-noise ratios (S/N) under the conditions of a tight and loose laser/cavity lock. The result is shown in Figure 1. The cavity length was dithered at a low audio frequency and a lock-in was used to demodulate signals from the direct cavity transmission (DC) and from the high frequency channel of NICE-OHMS. Signal lineshapes follow modulation-broadened derivatives of absorption (DC)[8] and dispersion (NICE-OHMS).[9] The DC detection of the intracavity molecular absorption (left column) is shown to be critically dependent upon the performance of the laser/cavity lock. (A fast laser/cavity frequency-lock servo was used for the graphs obtained in the upper row while a slow and noisy servo was used for those in the bottom row.) However, increased laser frequency noise (relative to the cavity) yields little effect in our FM detection (right column).
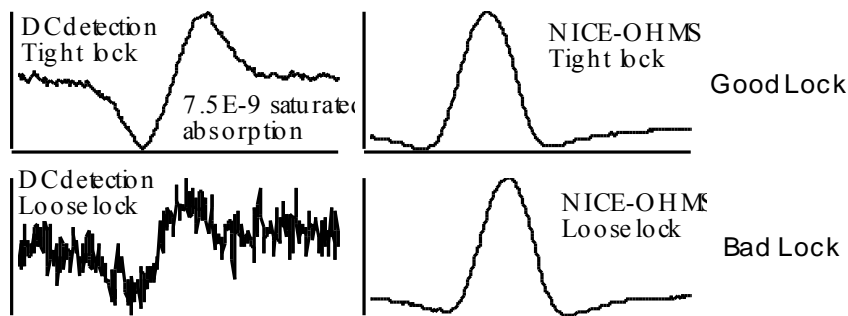

Figure 1. Demonstration of the noise-immune property. C_2HD ($v_1 + 3v_3$) P(5) transition is recovered by both the DC and the NICE-OHMS techniques, under conditions of a tight laser/cavity lock (upper row) and a substantially deteriorated lock (bottom row).

Figure 2 shows the experimental sensitivity we have achieved using 1.8 mTorr C_2HD gas. The cavity finesse is 100,000 and the intracavity buildup power is ∼ 300 W, giving a saturation parameter of ∼ 1.75 and a saturation peak contrast of 13.2%. The single-pass (46.9 cm long cavity) linear absorption is about 3×10^{-8}. Therefore the absolute level of saturated absorption by the intracavity molecules is 4×10^{-9}. This is verified by the DC detection of the cavity transmission, shown in the left graph of the figure. With the laser locked tightly onto the cavity with a relative noise of ∼ 1 milliHertz, the simple cavity-dither and lock-in detection of the transmission yields a S/N (amplitude / rms noise) of 130 at 1-s averaging. This corresponds to a detection

sensitivity of 3×10^{-11} (6.4×10^{-13} / cm) at 1 s. The corresponding S/N from the NICE-OHMS detection is 7700 with a 1 s time constant, as shown in the right graph of the figure. This translates into a noise-equivalent detection sensitivity of 5.2×10^{-13} at 1 s averaging, about 1.5 times worse than the shot noise limit calculated from Equation (2). Stated equivalently, the detection sensitivity for the refractive index is on the order of 1×10^{-19} at 1-s. The NICE-OHMS result is ~ 60 times better than the simple dither detection, basically because of its higher modulation frequency (319 MHz FM sideband frequency compared with 500 Hz dither frequency) and its insensitivity towards the laser frequency noise relative to the cavity.
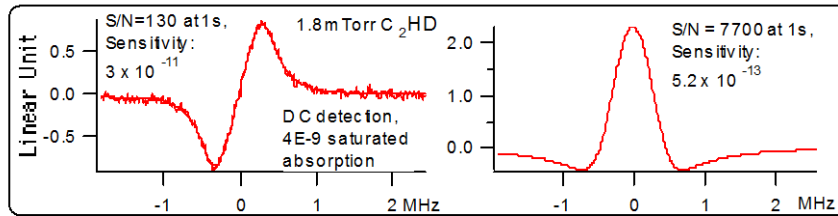

Figure 2. Sensitivity of the NICE-OHMS technique. The left graph shows the dither - based saturated absorption signal and the right graph shows the NICE-OHMS' S/N.

With this ultrasensitive capability of our spectrometer, we have measured two other weak transitions within the tuning range of the Nd:YAG laser at 1.064 μm, namely $^{12}C_2H_2$ $(2v_1 + v_2 + v_5)$ R(12) and $^{12}C^{16}O_2$ $(2v_1 + 3v_3)$ R(6), with their respective transition dipole moment of 50 μDebye and 6 μDebye. (1 Debye = 3.33564×10^{-30} C·m) They are both weaker than the C_2HD $(v_2 + 3v_3)$ P(5) transition (~ 70 μDebye). Using the same gas pressure, optical power, and cavity dither amplitude, we compare the saturated absorption signals of CO_2, C_2HD and C_2H_2 in Figure 3. The signal size of the C_2H_2 transition is about 1/4.3 of that for C_2HD. It provides another frequency reference for the Nd:YAG laser. The center frequency of the resonance is 281,612,403.278 (.025) MHz, i.e., it is 17708.458 (.014) MHz red of a frequency doubled Nd:YAG laser locked on the a_{10} hyperfine-structure component of the R(56) 32-0 I_2 transition.[10] The pressure broadening rate of 34(1) MHz/Torr is similar to that of C_2HD.[7]

The saturated absorption signal of the CO_2 transition is much weaker than the C_2HD line, by more than a factor of 350. The optical power is 410W inside, where the saturation of the CO_2 transition is estimated to be only a few percent (2%) in the free-flight regime. The recovered CO_2 lineshape is vastly different from either C_2HD or C_2H_2, as indicated in Figure 3. In fact, our theoretical model for the excellent fit of the experimental data involves two separate resonances that have different linecenters and linewidths. More importantly, the narrower negative-going peak indicates a physical process reversed from the normal saturated absorption. Extrapolated to zero pressure, the inverted peak has a FWHM of ~ 100 kHz, half of the transit time (210 kHz). These facts have led us tentatively to identify the resonance as a mixture of a

near-resonant two-photon transition in the neighborhood of the one-photon resonance.

In thinking about further advancing the detection sensitivity, it is expected that signal detection in cavity reflection should be advantageous. In this case, the heterodyne detection involves the molecular signal wave and a much reduced power at the carrier frequency, thanks to the destructive interference between the cavity-leaked
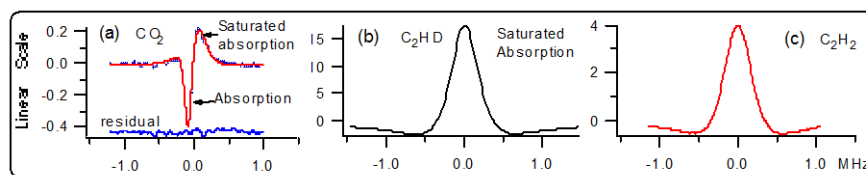

Figure 3. Lineshape comparison among the transitions of (a) CO_2, (b) C_2HD, and (c) C_2H_2, all with 5 mTorr of gas, 410 W intracavity power and 50 kHz peak-to-peak cavity dither.

field and the direct reflection of the incident field. The shot noise level can eventually be set by the molecular signal itself, instead of the residual carrier, leading to a much enhanced detection S/N. This approach relies critically on the perfect mode-match between the incident field and the cavity leakage field, since the spatial mode-matching of the local oscillator with respect to the molecular signal is no longer automatic as in the transmission case. We foresee the necessity of using an auxiliary optical cavity before the high finesse resonator to stabilize the mode direction and structure of the incident field.

At present the instrumental resolution bandwidth of our spectrometer still exceeds the natural linewidth of the molecular resonance. Although such a transit time limited linewidth can be narrowed by optical selection of slow-moving molecules, the result comes with a penalty in the reduction of S/N. To have a longer interaction time, we either need heavier molecules or use an optical cavity with a larger mode size. We are exploring the idea of pressure-tuning the radius of a high-finesse cavity mirror.

A high detection sensitivity along with a high resolution are welcome to explore new phenomena that are otherwise hidden from us. Our spectrometer is ready to search for the predicted permutation splitting associated with exchange of the hydrogen atoms when their vibrational amplitudes become large in the high order overtone band ($5v_3$) of $^{13}C_2H_2$.[11] Another important prospect is high precision null tests on a number of fundamental physical principles. The NICE-OHMS detection technique can also be applied to the case of two-photon resonances. When the optical intensity is reduced, the fluorescence S/N decreases as I^{-1} while the NICE-OHMS S/N decreases as $I^{-1/2}$. Therefore NICE-OHMS should win at the low intensity range. Using a relatively low optical power is favorable for a frequency standard, as the power-related systematic shifts on the center frequency can be reduced. NICE-OHMS is especially valuable when the fluorescence is weak and the fluorescence detection efficiency (quantum and/or collection efficiency) is low.

In the work of optical frequency metrology, NICE-OHMS technique can provide us with thousands of weak molecular lines as high quality visible frequency/wavelength references. A fast servo loop narrows the laser fast linewidth, with the limit usually set by vibrations of the reference cavity. The combined laser/cavity system is then locked onto the intracavity molecular dispersion for long-term stability. We checked the quality of this overtone-stabilized laser at 1.064 μm against the known Nd:YAG/I_2 reference system. The absolute frequency of the P(5) line in the ($\nu_2 + 3\nu_3$) band of $^{12}C_2HD$ has been determined.[7] (See the paper by Hall *et al.* in this proceeding for the heterodyne result between two different laser systems.)

Linewidth narrowing is offered by slow molecules since the natural life-time of the overtone transition is about 300 times longer than our current transit time. A low power is necessary so that the low Rabi frequency leads to appreciable saturation only for the slowest molecules. In our < 2 mTorr sample gas, the mean-free-path of molecules is ~ 30 times longer than the transverse field dimension, resulting in the transit-time regime. Slow molecules with their whole life-time inside the field will have a constant and velocity-independent saturation parameter, controlled primarily by the collisional broadening. Faster moving molecules see a reduced saturation and mostly contribute to the wings of the resonance. Using a power that is reduced by 75 times from normal cases, Figure 4 shows a resonance with a linewidth of ~ 20 kHz. This is thirteen times narrower than that set by the room temperature transit-time-limit, and is mainly limited by the relatively high pressure (1.8 mTorr). With an improved S/N this approach will enable us to access the information of free molecules with minimized second order Doppler shift (~ 2 x 10^{-14}).
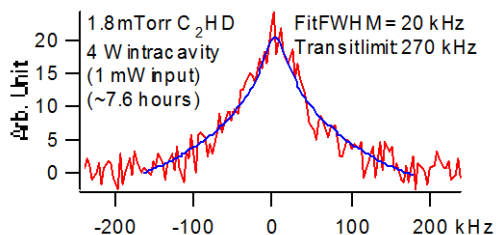

Figure 4. Narrow resonance with slow molecules. With low power and gas pressure, slow molecules give a linewidth 13 times below the room-temperature transit limit.

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