Noise-immune cavity-enhanced optical frequency comb spectroscopy

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We present a new method of optical frequency comb spectroscopy that combines cavity enhancement with frequency modulation to obtain immunity to laser frequency-to-amplitude noise conversion by the cavity modes and, thus, high absorption sensitivity over a broad spectral range. A frequency comb is locked to a cavity with a free spectral range (FSR) equal to 4/3 times the repetition rate of the laser, and phase-modulated at a frequency equal to the cavity FSR. The transmitted light is analyzed by a Fourier transform spectrometer with a high bandwidth detector. Phase-sensitive detection of the interferogram yields a noise-immune cavity-enhanced optical frequency comb spectroscopy (NICE-OFCS) signal. In the first demonstration, we record NICE-OFCS signals from the overtone CO₂ band at 1575 nm with absorption sensitivity of 4.3 × 10⁻¹⁰ cm⁻¹ Hz⁻¹/² per spectral element, close to the shot noise limit. © 2014 Optical Society of America

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Cavity-enhanced optical frequency comb spectroscopy (CE-OFCS) combines high spectral resolution and broad spectral coverage provided by optical frequency combs (OFCs), with vast enhancement of the interaction length with the sample obtained using high-finesse cavities [1]. Efficient comb–cavity coupling is achieved by proper adjustment of the repetition rate and the carrier–envelope offset frequency of the OFC [2]. The main source of noise in CE-OFCS is the amplitude noise, created by fluctuations of the frequencies of the comb lines relative to the center frequencies of the narrow cavity modes, a process called frequency-to-amplitude (FM–AM) noise conversion. Various detection schemes have been developed to overcome this limitation; shot noise-limited performance has been achieved using a combination of a tight comb–cavity lock and a fast-scanning Fourier transform spectrometer (FTS) with auto-balanced detection [3], and, alternatively, using a combination of dithering the OFC modes around the cavity modes and detection by a spectrograph [4]. The former method provides high resolution and wide spectral coverage, but it employs a method of active noise reduction and, thus, relies on the performance of the auto-balancing detector. The latter, on the other hand, is a simpler implementation that provides shorter acquisition times, although at the expense of a low spectral resolution and narrow bandwidth, limited by the size of the detector array.

Here, we employ a concept based on noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) [5], the most sensitive continuous wave (cw) laser-based absorption technique, in which the noise is reduced in the optical domain by a unique combination of frequency modulation spectroscopy (FMS) [6] with cavity enhancement. In NICE-OHMS, the phase of the light incident on the cavity is modulated at a frequency equal to the cavity free spectral range (FSR) so that, when the laser carrier frequency is locked to a particular cavity mode, the sidebands are transmitted through the neighboring cavity modes. All components of the FM-triplet are then transmitted through the cavity in an identical manner and are, therefore, affected by FM–AM noise conversion in the same way. Thus, the beat signal between the carrier and the two sidebands, which is detected in FMS, is free of intensity noise and gives the technique the so-called noise immunity.

In noise-immune cavity-enhanced optical frequency comb spectroscopy (NICE-OFCS) we obtain such noise immunity over a wide spectral bandwidth by performing NICE-OHMS on the modes of a frequency comb. The OFC is locked to a cavity and phase-modulated at a frequency equal to the cavity FSR. In general, the cavity FSR has to be equal to (3p ± 1)/3 times the comb repetition rate, where p is an integer, so that every (3p ± 1)th comb line is matched to every 3rd cavity mode, and the FM sidebands of the comb lines are matched to the remaining cavity modes. To perform phase-sensitive detection of the cavity-transmitted intensity, a method is required that provides broad spectral coverage and electronic bandwidth of the order of hundreds of megahertz. Thus, detection schemes employing CCD cameras or detector arrays [4,7,8] cannot be used, due to their limited electronic bandwidth. Our detection system is, therefore, based on an FTS [3,9], in which the signal is detected in the time domain by a single high-speed detector. Note that, although the theory presented below is valid for the particular case of detection by FTS, the result, i.e., the achieved noise immunity, is general.

FMS has previously been combined with FTS in an experiment employing a rapidly tuned phase-modulated cw laser [10]. In that implementation, an electro-optic modulator (EOM), used for phase modulation, and an absorption cell were placed behind the FTS. In NICE-OFCS, the EOM and the cavity containing the sample have to be placed before the FTS, because the Doppler shift of the comb repetition rate after the FTS would prevent precise comb–cavity matching. The different order of the components of the system modifies the theoretical description of the signal with respect to that presented in [10]. The electric field at the output of the FTS is a sum of two phase-modulated OFC fields E±, which have traveled through different arms of the interferometer. Each comb mode is characterized by a field amplitude E±n and
frequency \( \omega_n = 2\pi(nf_{rep} + f_0) \), where \( f_{rep} \) is the repetition rate, \( f_0 \) is the carrier–envelope offset frequency, and \( n \) is an integer comb mode number. In the limit of low modulation index \( \beta < 1 \), the two electric fields can be written as

\[
E_\pm = \sum_n \sum_{k=-1,0,1} E_n J_k(\beta) e^{i(\omega_n + k\omega_m)(t \pm \delta)} + \text{c.c.},
\]

where \( J_k \) is the Bessel function of order \( k \), \( T_{n;k} \) is the complex transmission function of the cavity filled with the analyte at a frequency \( \omega_n + k\omega_m \), \( \omega_m \) is the modulation frequency, \( \Delta \) is the optical path difference (OPD) between the two arms in the FTS, \( c \) is the velocity of light in vacuum, and c.c. stands for the complex conjugate. The full expression for the cavity transmission function in the presence of an absorber is given by Eq. (4) in [11]. For simplicity, we neglect the effect of cavity mirror losses and dispersion, and we use an approximation of the cavity transmission function, given by \( T_{n;k} = \exp(-\delta_{n;k}^\beta - i\phi_{n;k}) \): with \( \delta_{n;k}^\beta = 2F\delta_{n;k}/\pi \) and \( \phi_{n;k} = 2F\phi_{n;k}/\pi \), where \( F \) is the cavity finesse, and \( \delta_{n;k} \) and \( \phi_{n;k} \) are the single-pass amplitude attenuation and optical phase shift, respectively, of the \( n \)th comb line \((k = 0)\) and its sidebands \((k = \pm 1)\). The intensity incident on the detector at the output of the FTS contains a component at the modulation frequency originating from the beating of the modes of one comb \( E_+ \), with the sidebands of the other comb \( E_- \), and vice versa. This term can be written as

\[
I_{\omega_m} = \frac{1}{2} J_0(\beta) J_1(\beta) \left\{ \sin(\omega_m t) \sum_n I_n \left[ \cos(\omega_m \Delta/c) \cos(\omega_m \Delta_{c}/2c) \left[ e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \sin(-\phi_{n,0}^F + \phi_{n,-1}^F) - e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \sin(\phi_{n,0}^F - \phi_{n,-1}^F) \right] \right] + \sin(\omega_m \Delta/c) \sin(\omega_m \Delta_{c}/2c) \left[ e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \sin(-\phi_{n,0}^F + \phi_{n,-1}^F) + e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \sin(\phi_{n,0}^F - \phi_{n,-1}^F) \right] \right] + \cos(\omega_m t) \sum_n I_n \left[ \cos(\omega_m \Delta/c) \left[ -e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \cos(-\phi_{n,0}^F + \phi_{n,-1}^F) + e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \cos(\phi_{n,0}^F - \phi_{n,-1}^F) \right] \right] - \sin(\omega_m \Delta/c) \left[ e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \cos(-\phi_{n,0}^F + \phi_{n,-1}^F) + e^{-\delta_{n,0}^\beta \delta_{n,-1}^\beta} \cos(\phi_{n,0}^F - \phi_{n,-1}^F) \right] \right\}. \tag{2}
\]

The dispersion and absorption factors in the 2nd and 5th lines have the same functional form as in conventional FMS [6] or NICE-OHMS [12]. The use of FTS in NICE-OFCs causes the appearance of two additional factors (in the 3rd and 6th lines). Note that the factor in the 6th line is not equal to zero in the absence of absorption; moreover, it seems to violate the condition for noise immunity, since the attenuations of the FM-triplet components combine with the same sign. However, the amplitude of this interferogram is of appreciable size only over a small fraction of the OPD scan (in our case \( \sim 1 \) mm out of 300 mm, see Fig. 3), so the amount of noise it couples in is negligible.

Our OFC source is an Er:fiber femtosecond laser providing 20 mW of average power in the 1.5–1.6 μm wavelength range with a repetition rate of 250 MHz. To implement NICE-OFCs, we use a 45 cm long cavity with FSR equal to 43 times \( f_{rep} \), i.e., 333 MHz, so that every 4th comb mode is matched to every third cavity mode, and the FM sidebands (at 333 MHz) are transmitted through the two cavity modes in between, as shown in Fig. 1. In this configuration, three out of four comb lines are reflected by the
The lower frequency transitions is $\text{GHz}$.

The experimental setup is shown in Fig. 2. The output of the OFC is phase-modulated at 20 and 333 MHz using a single fiber-coupled LiNbO$_3$ EOM. The lower frequency, $f_{\text{PDH}}$, is used for error signal generation for locking of the comb to the cavity. The higher frequency, $f_m$, is matched to the cavity FSR and used for NICE-OFCS signal generation. The modulated light is coupled out into free space and spatially mode-matched to the cavity TEM$_{00}$ mode. The cavity has a finesse of 2900 at 1575 nm, and is made of two concave mirrors with radii of curvature of 5 m mounted on a 45 cm long stainless steel spacer tube. The tube is connected to a gas flow system and a vacuum pump, which allow filling the cavity with a calibrated gas sample at a desired pressure. The comb is locked to the cavity by the two-point Pound–Drever–Hall (PDH) locking scheme [11]. We choose the locking points at 1572 and 1579 nm for the $f_0$ and $f_{\text{rep}}$ locks, respectively, to transmit 30 nm of spectral range matching the CO$_2$ absorption band centered at 1575 nm. 11.5% of the incident power is transmitted through the cavity: (i) 75% of the comb lines are reflected due to the FSR–$f_{\text{rep}}$ mismatch; (ii) 30% of the power is rejected due to the choice of the locking points; and (iii) the transmission per comb line is 66% due to cavity mirror losses.

The beam transmitted through the cavity is injected into a home-built fast-scanning FTS, whose design is similar to that described in [2]. The OPD is calibrated using a stabilized He–Ne laser, whose beam is propagating parallel to the OFC beam in the FTS. The output of the interferometer is incident on a 1 GHz InGaAs detector. The detector output is demodulated at 333 MHz and resampled at the zero-crossings and extrema of the He–Ne interferogram to yield the NICE-OFCS interferogram. Both interferograms are recorded with a 2-channel data acquisition card at 5 Msamples/s and 20 bit resolution.

Figure 3 shows the out-of-phase NICE-OFCS interferogram (blue) as a function of OPD recorded in 1.6 s, when the cavity is filled with 1% of CO$_2$ in N$_2$ at 500 Torr. As a guidance to the eye, we have drawn the $\sin(a_m \Delta /2c)$ and $\cos(a_m \Delta /2c)$ envelope factors (black dashed and red dashed–dotted curves, respectively) from Eqs. (2) and (3). The amplitudes of the interferograms at OPD = 0 cm and 90 cm are virtually zero, which is expected since the absorption term multiplied by the cosine envelope is very small for the particular ratio of modulation frequency and absorption line width in our experiment (the pressure broadening of the CO$_2$ transitions is $\sim$1.5 GHz).

To obtain the final NICE-OFCS signal, we scan the FTS in a symmetric 30 cm long OPD range around the interferometer, centered at OPD = 30 cm (as marked in the figure). Such a single interferogram is acquired in 0.4 s. A zoom of the baseline of one side of the interferogram is shown in Fig. 4(b). The absorption response of CO$_2$ is clearly visible with high signal-to-noise ratio. Fast Fourier transform (FFT) of the interferogram yields a NICE-OFCS signal, shown in Fig. 4(d), with a resolution of 1 GHz and 4000 resolved elements. The absorption lines belong to the P and R branches of the $\tilde{\nu}_1$ overtone band of CO$_2$. Figure 4(e) shows a normalized CO$_2$ spectrum, i.e., divided by a background spectrum recorded when the cavity was filled with pure nitrogen.

To obtain noise-immune conditions, the modulation frequency has to be carefully matched to the cavity FSR. We tune the $f_m$ manually to the optimum value by observing and minimizing the noise in the NICE-OFCS signal. To demonstrate how the signal-to-noise ratio decreases with the detuning of $f_m$ from FSR, Fig. 4(c) shows the NICE-OFCS signal for the case when the two frequencies are detuned by 20 kHz, i.e., 28% of the cavity linewidth. The corresponding interferogram is shown in Fig. 4(a). The degradation of the noise immunity for this $f_m$–FSR mismatch is evident. However, we observe that the noise is
Fig. 4. Zoom of the NICE-OFCS interferogram when the modulation frequency is (a) detuned by 20 kHz from the cavity FSR and (b) precisely tuned to the cavity FSR. NICE-OFCS signal from 1% of CO₂ in 500 Torr of N₂, when the modulation frequency is (c) detuned by 20 kHz from the cavity FSR and (d) precisely tuned to the cavity FSR. The normalized NICE-OFCS signal is shown in (e).

not increasing much for detunings up to a few percent of the cavity linewidth.

To estimate the absorption sensitivity, we take a ratio of the power spectrum to the shot noise spectrum. The standard deviation of the noise in the center of this normalized spectrum (∼1575 nm) is found to be σ = 2 × 10⁻³. This corresponds to a noise equivalent absorption coefficient of 3 × 10⁻⁸ cm⁻¹ in 0.8 s, calculated as σ/Lₐeff, where Lₐeff is defined as 2FL/π, and L is the cavity length. Finally, the sensitivity per spectral element is equal to 4.3 × 10⁻¹⁰ cm⁻¹ Hz⁻¹∕², given by σ/Lₐeff(T/M)⁻¹∕², where T is the acquisition time and M the number of resolved elements.

Under shot noise-limited conditions, the minimum detectable absorption coefficient per spectral element in FTS-based NICE-OFCS is given by

\[ a_{\text{shot-noise}} = \sqrt{\frac{eB}{\eta P J_0(\beta)J_1(\beta)}} \sqrt{\frac{M}{N L_{\text{eff}}}} \]

where e is the electron charge, B the electronic bandwidth, η the detector responsivity, P the optical power incident on the detector, and N the total number of data points in the interferogram. The first term is the shot noise limit in direct absorption; the second term originates from the fact that the power in the FMS signal is intrinsically lower by a factor of JO(β)JO(β)/√2 than in direct absorption [3]; the third term is specific to FTS and reflects both the conversion of signal-to-noise ratio from the time to the frequency domain, given by M/√N [3], and the normalization to the number of spectral elements, i.e., 1/√M, and the last term represents the cavity-enhanced interaction length. For the conditions valid in our experiment, namely, B = 2.5 MHz, η = 1 A/W, P = 0.36 mW, β = 0.35, M = 4000, N = 10⁶, and Lₐeff = 659 m, this yields 3.8 × 10⁻¹⁰ cm⁻¹ Hz⁻¹∕² per spectral element. This shows that the achieved sensitivity is close to the shot noise limit.

In conclusion, we have demonstrated for the first time noise-immune cavity-enhanced optical frequency comb spectroscopy (NICE-OFCS) and achieved shot noise-limited absorption sensitivity for detection of CO₂ around 1.57 μm. This technique implements the concept of NICE-OHMS, the most sensitive cw laser-based spectroscopic technique, over the broad spectral bandwidth of the OFC. It has the potential to become a versatile tool for broadband high resolution spectroscopy, since it reduces the amplitude noise created by the FM–AM conversion in the optical domain. Moreover, the detection system is based on commercially available detectors and electronic components. The spectral resolution can be increased by resolving the comb lines, and the sensitivity can be increased by increasing the cavity finesse. This, as well as accurate modeling of NICE-OFCS lineshapes, will be the subject of future work. Eventually, development of high speed detector arrays might allow replacing the FTS with a parallel detection scheme, which would increase the sensitivity by the square root of the number of resolved elements [3] and, thus, allow exploiting the full potential of this technique.

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