Sensitive and broadband measurement of dispersion in a cavity using a Fourier transform spectrometer with kHz resolution

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Abstract: Optical cavities provide high sensitivity to dispersion since their resonance frequencies depend on the index of refraction. We present a direct, broadband, and accurate measurement of the modes of a high finesse cavity using an optical frequency comb and a mechanical Fourier transform spectrometer with a kHz-level resolution. We characterize 16000 longitudinal cavity modes spanning 16 THz of bandwidth in terms of center frequency, linewidth, and amplitude. Using the center frequencies we retrieve the group delay dispersion of the cavity mirror coatings and pure N2 with 0.1 fs2 precision and 1 fs2 accuracy, as well as the refractivity of the ν1 + ν3 absorption band of CO2 with 5 × 10−12 precision. This opens up for broadband refractive index metrology and calibration-free spectroscopy of entire molecular bands.

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References and links

1. Introduction

Fabry-Perot cavities in combination with narrow linewidth continuous wave (cw) lasers are versatile tools for ultra-sensitive measurements of displacement, absorption, and dispersion. For example, high precision measurements of minute length variation of Fabry-Perot cavities have enabled detection of gravitational waves [1]. A pressure sensor based on the measurement of gas refractivity inside a cavity can outperform a manometer [2]. Cavity-enhanced molecular absorption [3, 4] and dispersion [5, 6] spectroscopies, which rely on the measurement of intracavity absorption losses and dispersion-induced cavity modes shifts, respectively, provide complementary information about the molecular transitions and high sensitivity to absorption/dispersion. However, cw lasers allow such measurements only over narrow bandwidths, typically in the sub-THz range. Optical frequency combs, whose spectra consist of thousands of equidistant narrow lines, can probe cavity modes over a much broader bandwidth. In cavity-enhanced optical frequency comb spectroscopy, absorption spectra of entire molecular bands can be acquired with high resolution in short acquisition times [7–10]. Combs are also an ideal tool for measurements of broadband dispersion induced either by the cavity mirror coatings or intracavity samples. Previous demonstrations relied on indirect measurements of the resonant condition between the comb and the cavity and required external frequency calibration [11–13]. Therefore, they did not fully benefit from the high frequency accuracy provided by the comb and suffered from poor spectral resolution (at the THz level), which was insufficient to allow observing dispersion induced by molecular transitions.

Here we present a new method for direct measurement of broadband spectra of the modes of a high finesse cavity using a frequency comb and we determine the group delay dispersion of the cavity mirror coatings and intracavity gas samples with unprecedented combination of bandwidth and precision. We achieve sub-kHz resolution in the measurement of the cavity mode frequencies and widths using a mechanical Fourier transform spectrometer (FTS) [14, 15]. Our method allows calibration-free quantification of the dispersion induced by narrow molecular transitions and opens up for improved metrology of refractive index and of entire molecular absorption bands.

2. Theory

The cavity mode frequencies, \( \nu_q \), fulfill the resonance condition on the round-trip phase shift of the electric field, \( \Phi \), which can be written as
where \( q \) is an integer mode index, \( \phi_0(v_q) = 2\pi v_q L/c + \phi_m \) is the phase shift inside an empty cavity, where \( L \) is the cavity length, \( c \) is the speed of light, and \( \phi_m \) is the phase shift induced by the cavity mirror coatings, and \( \phi_d(v_q) = 2\pi v_q [n(v_q) - 1]2L/c \) is the phase shift induced by the intracavity sample with refractive index \( n \). The cavity modes spacing, i.e. the free spectral range, \( \text{FSR} = 2\pi[(\partial \Phi / \partial v)^{-1}] \), is not constant because \( \Phi \) varies non-linearly with frequency. To evaluate the shift of the cavity mode frequencies with respect to an equidistant grid, we define a reference scale

\[
\nu_0 = q \text{FSR}_{\text{ref}} + f_0,
\]

where \( \text{FSR}_{\text{ref}} \) is the empty cavity FSR evaluated around the cavity mode \( \nu_{\text{ref}} \), and \( f_0 = \nu_{\text{ref}} - q \text{FSR}_{\text{ref}} \) is an offset frequency, where \( q_{\text{ref}} = \text{floor}[\nu_{\text{ref}} / \text{FSR}_{\text{ref}}] \). The relation between the frequency shift of the cavity modes, \( \Delta \nu = \nu_q - \nu_0 \), and the intracavity phase shift, Eq. (1), can be found by Taylor expanding \( \Phi \) around \( \nu_{\text{ref}} \) to first order, which yields

\[
\Delta \nu = \nu_q - \nu_0 = \frac{\text{FSR}_{\text{ref}}}{2\pi} \left[ 2\pi q - \Phi(\nu_0) \right],
\]

where \( \text{FSR}_{\text{ref}} \) is the cavity FSR evaluated around the cavity mode \( \nu_{\text{ref}} \). The intracavity group delay dispersion (GDD) is then calculated as

\[
\text{GDD}(\nu_0) = \frac{1}{4\pi^2} \frac{\partial^2 \Phi}{\partial v^2} = -\frac{1}{2\pi \text{FSR}_{\text{ref}}} \frac{\partial^2 \Delta \nu}{\partial v^2}.
\]

The mode shift in an empty cavity, \( \Delta \nu_0 \), originates from the dispersion of the cavity mirror coatings. When the cavity is filled with a gas, an additional shift, \( \Delta \nu_n \), occurs because of the frequency dependence of the refractive index

\[
\Delta \nu_n = -v_0 \left[ n(v_0) - 1 \right].
\]

The refractive index \( n \) has two contributions \( n = n_{na} + n_{abs} \), where \( n_{na} \) is the slowly varying refractive index of a non-absorbing gas, given by e.g. the Sellmeier equation [16], while \( n_{abs} \) is the refractive index of molecular transitions, defined as

\[
n_{abs}(v_0) - 1 = -\frac{c}{4\pi^2} \rho \sum_i S_i \text{Im} \left[ \chi_i(v_0) \right],
\]

where \( \rho \) is the absorbing sample gas density, \( S_i \) is the line intensity and \( \chi_i \) is the complex line shape function of the \( i \)-th transition.

### 3. Experimental setup

The experimental setup is depicted in Fig. 1(a). The cavity with a finesse of ~1700 consisted of two dielectric mirrors with 5 \( \text{m} \) radius of curvature separated by \( L = 45 \text{ cm} \), yielding an FSR of 333 MHz. The mirrors were glued to two ends of a stainless steel tube, connected to a gas system, and a ring piezo-electric transducer (PZT) was inserted between one of the mirrors and the tube to control the cavity length. The cavity modes were probed by an amplified Er:fiber frequency comb with a repetition rate (\( f_{\text{rep}} \)) of 250 MHz, whose beam was mode-matched to the TEM\(_{00}\) modes of the cavity. Because of the difference between the cavity FSR and the comb \( f_{\text{rep}} \) the cavity acted as a filter for the comb, as shown in Fig. 1(b), and the repetition rate in cavity transmission was equal to \( f_{\text{rep}} / 4 = 3 \text{FSR} = 1 \text{ GHz} \). The
comb offset frequency, $f_{\text{com}}$, was stabilized by locking the output of an f-2f interferometer to a frequency provided by a GPS-referenced Rubidium clock, $f_{\text{clk}} = 20 \text{ MHz}$, via feedback to the current of a diode laser pumping the Er:fiber oscillator. The frequency of one of the comb lines was stabilized to a narrow-linewidth cw Er:fiber laser locked to the $P_{\text{e}(8)} \text{ CO}_2$ transition at $\lambda_{\text{cw}} = 1576.9396 \text{ nm}$ using sub-Doppler noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS [17], not shown in the figure). The linewidth of the cw laser was 2.8(6) kHz, estimated using the method described in ref [18]. The two laser beams were combined in a fiber and the beat note between the cw laser and the closest comb line was detected in free space after dispersing the spectrum by a diffraction grating. The beatnote was locked to a radio frequency, $f_{\text{DDS}}$, generated by a tunable direct digital synthesizer (DDS) referenced to the Rb clock, via feedback to an intracavity PZT and an electro-optic modulator (EOM) controlling the $f_{\text{rep}}$. This optical lock transferred the linewidth of the cw laser to the closest comb line, which was verified by measuring the width of the optical beat note below the Hz level using a spectrum analyzer. The cavity length was stabilized by locking one of the cavity resonances to the cw laser using the Pound-Drever-Hall (PDH) technique. This involved phase modulation of the electric field of the cw laser at a frequency $f_{\text{PDH}} = 20 \text{ MHz}$ using an EOM and phase sensitive detection of the cavity reflected light, which was dispersed using a diffraction grating to avoid saturation of the detector by the reflected comb intensity. The correction signal was fed to the cavity PZT with a closed-loop bandwidth of 1.1 kHz. Due to this relatively low bandwidth a frequency jitter of the cavity modes remained, yielding a mode broadening of 20 kHz, calculated from the power spectral density of the closed-loop error signal [18].

Setting $f_{\text{DDS}}$ close to 83 MHz, as shown in Fig. 1(b), brought the comb lines close to resonance with the cavity modes. The transmitted light, composed of overlapping comb and cw laser beams, was analyzed with a fast-scanning FTS equipped with an auto-balancing detector [8, 19]. To achieve kHz resolution we used the method described in refs [14, 15], in which the nominal resolution of the FTS, given by the inverse of the optical path difference [20], is set equal to the comb line spacing in order to allow precise measurement of the comb line intensities without distortion induced by the instrumental line shape. We set the nominal resolution to $4f_{\text{rep}}/3$, which yielded sampling points spaced by FSR and allowed sampling of the intensities of the transmitted comb lines as well as the cw laser. To measure the profiles of the cavity modes we scanned the comb lines across the cavity modes by tuning $f_{\text{DDS}}$ in steps of 20 kHz. This in turn caused tuning of $f_{\text{rep}}$ via the feedback to the comb oscillator. The 20 kHz step was chosen to yield more than 10 points per cavity mode width and it could be made smaller if needed. Two interferograms, each lasting 2.5 s, were recorded and averaged for each $f_{\text{rep}}$ value, and 150 steps were acquired, with a total acquisition time of 20 min. The 150
spectra obtained after fast Fourier transform were interleaved [21] to yield a spectrum
containing discrete spectral pieces separated by $f_{\text{ref}} = 1 \text{ GHz}$, each covering 3 MHz.

4. Results

The spectrum of an empty cavity, spanning from 1500 to 1640 nm and containing 16000 resonance modes, is shown in Fig. 2(a). The peak intensities of the cavity modes follow the comb spectral envelope and a discrete peak is visible at $\lambda_{\text{cw}}$. The inset of Fig. 2(a) shows a zoom of a part of the spectrum containing 3 cavity modes separated by 3FSR. The spectrum of a cavity mode at 1600 nm is further enlarged in Fig. 2(b) (black markers). A model based on a Lorentzian function and a linear baseline is fitted to the data (red curve, with residuum in the lower panel). The fitted parameters were the amplitude $A_q$, the center frequency $\nu_q$, and the width $\Gamma_q$ of the Lorentzian function, as well as the offset and slope of a linear baseline. The fit returned $A_q = 1.019(3)$, $\nu_q = 18769770122.4(4) \text{ kHz}$, and $\Gamma_q = 214.4(9) \text{ kHz}$. To verify the accuracy of the mode width measurement, we measured the cavity ring-down (RD) time at 1600 nm using the comb as the light source, an acousto-optic modulator before the cavity to interrupt the beam, and a monochromator with 200 GHz resolution to disperse the transmitted spectrum. The measurement yielded a mode width of $\Gamma_{\text{RD}} = 191(2) \text{ kHz}$, which agrees with the value obtained from the fit to the mode profile when considering the mode broadening due to the remaining cavity jitter (20 kHz) and the comb linewidth (2.8 kHz).

![Fig. 2. (a) Spectrum of the empty cavity transmission spanning 16 THz consisting of 16000 discrete modes. Inset: enlargement of 3 modes separated by 3FSR. (b) A zoom of a single mode at 1600 nm (black markers) together with a Lorentzian fit (red line) and the residuum (lower panel).](image)

The center frequency of each of the 16000 cavity modes was determined from fits of a Lorentzian line shape function with an uncertainty ranging from 0.4 to 3 kHz depending on the signal-to-noise ratio of the considered mode. Figure 3(a) shows the shift of the cavity modes, calculated using Eqs. (2) and (3), for the empty cavity [$\Delta \nu_0$, black curve, left y-axis in MHz] and the cavity filled with pure N$_2$ at 750(1) Torr and 296(3) K [$\Delta \nu_{\text{N2}}$, red curve, right y-axis in GHz]. The FSR$_{\text{ref}}$ in Eq. (2) was set equal to 4/3 times the $f_{\text{ref}}$ value that maximized the transmission of the comb lines through the cavity modes around $\nu_{\text{ref}} = c/\lambda_{\text{cw}}$. This yielded FSR$_{\text{ref}} = 333.570693(1) \text{ MHz}$ and $f_0 = 63.33(5) \text{ MHz}$. When the cavity was filled with N$_2$ the change of the intracavity refractive index had to be compensated by adjusting the cavity length in order to keep the cavity modes in resonance with the comb lines. This in turn changed the value of FSR$_{\text{ref}}$ and $f_0$. The new value of FSR$_{\text{ref}} = 333.5795270(2) \text{ MHz}$ was found from the $f_{\text{ref}}$ value that maximized the transmission of the comb lines around $\nu_{\text{ref}}$, divided by the refractive index of N$_2$ at $\nu_{\text{ref}}$, calculated using the Sellmeier equation with coefficients from [22]. The new offset frequency was $f_0 = 219.40(5) \text{ MHz}$. Since both curves were referenced to $\nu_{\text{ref}}$ they cross 0 Hz at $\lambda_{\text{cw}}$. 
Fig. 3. (a) Shift of the cavity mode frequencies $\Delta \nu$ measured when the cavity is empty (black curve, left y-axis) and when the cavity is filled with pure $N_2$ at 750 Torr (red curve, right y-axis) plotted together with a calculated shift based on the Sellmeier equation for $N_2$ (blue markers). Note the three orders of magnitude difference between the two y-axis scales. (b) Residuum of a polynomial fit to the shift of the empty cavity modes. (c) Residuum of a polynomial fit to the mode shift of the cavity filled with $N_2$.

To verify the accuracy of the measurement, we calculated the shift induced by $N_2$ using Eq. (5) and the frequency dependence of the refractive index of $N_2$ from [22], subtracted the value of $\nu_{\text{ref}} (n_{\text{ref}} - 1)$ to make $\Delta \nu_{N_2} = 0$ Hz at $\lambda_{\text{cw}}$, and added the experimentally determined empty cavity shift. The resulting curve is shown with blue markers in Fig. 3(a), and it is in excellent agreement with the measured curve (relative error of the slopes is $4 \times 10^{-3}$).

To retrieve the GDD of the cavity mirror coatings and $N_2$, we fitted Eq. (3) to the two $\Delta \nu$ curves in Fig. 3 with $\Phi$ assumed as a fifth order polynomial function. The residuals of the fits are shown in Figs. 3(b) and 3(c), confirming the validity of the model and the kHz precision of the determination of the mode shift. The fitted curves were differentiated twice [see Eq. (4)] to yield the GDD of the empty cavity and the cavity filled with $N_2$. The results are shown in Fig. 4 by the black and red solid curves, respectively. The uncertainty of the GDD is below 0.1 fs$^2$ over the entire range, originating from the uncertainty of the determination of $\nu_q$ and of the fit coefficients. The GDD of the cavity filled with $N_2$ is compared with a theoretical curve (dashed curve) calculated as the sum of the experimentally determined empty cavity GDD and the GDD of $N_2$ calculated using the frequency-dependent refractive index of $N_2$ from [22]. Both curves agree within 1 fs$^2$.

Fig. 4. Group delay dispersion (GDD) of the empty cavity (black solid curve), and the cavity filled with pure $N_2$ at 750 Torr (red solid curve). The dashed curve is the sum of the GDD of $N_2$ calculated using the Sellmeier equation and the experimentally determined empty cavity GDD (dashed black curve).

The high precision of the retrieved cavity mode frequencies allows also the measurement of dispersion induced by molecular transitions. Figure 5(a) shows the refractivity of the $3v_1 + v_3$ band of $CO_2$ (black markers) obtained using Eq. (5) and the cavity mode frequencies...
measured when the cavity was filled with 1.00(5)% of CO₂ in N₂ at 750(1) Torr at room temperature [296(3) K]. The red solid curve shows a fit of the molecular refractivity, Eq. (6), calculated using the imaginary part of the complex Voigt profiles and CO₂ line parameters from the HITRAN database [23], with the sample density as the fitting parameter. The slowly varying background originating from the N₂ and cavity mirror dispersion was removed by fitting a fifth order polynomial function together with the model. The residual of the fit, shown in Fig. 5(b), has a standard deviation equal to 5 × 10⁻¹², demonstrating the high precision of the measurement and a good agreement with theory. The CO₂ concentration retrieved from the fit was 0.95(1)%. The relative error of 5% with respect to the specified concentration, and the structure remaining in the residuum, are presumably caused by the fact that the pressure broadening and shift parameters in HITRAN are defined for CO₂ in air while our sample was CO₂ in N₂.

Fig. 5. (a) Refractivity of the 3ν₁ + ν₃ absorption band of 1% of CO₂ in N₂ at 750 Torr (black markers) together with a fit (red curve). (b) Residual of the fit.

5. Conclusions and outlook

To summarize, we measured cavity transmission spectra spanning 16 THz of bandwidth and containing 16000 modes with ~200 kHz linewidth using a comb-based Fourier transform spectrometer. These measurements provide evidence that the resolution of a comb-based mechanical FTS is given by the comb linewidth, and not by the maximum optical path difference, provided that the nominal resolution of the FTS is matched to the comb line spacing. Each cavity mode was fully characterized in terms of amplitude, width, and center frequency. From the shift of the center frequencies we retrieved the GDD of the cavity mirror coatings and of pure N₂, as well as the refractivity of the entire CO₂ absorption band. The precision of the GDD measurement was at the 0.1 fs² level and the accuracy of the N₂ measurement was within 1 fs² of a model based on the Sellmeier equation, while the precision of the refractive index of the molecular transitions was 5 × 10⁻¹².

Our method offers at least one order of magnitude improvement on the signal-to-noise ratio compared to the previous demonstration of direct measurement of cavity resonance modes using a dual-comb spectrometer [24], allowing retrieval of the cavity mode parameters with significantly improved precision. The demonstrated precision of the dispersion measurement is more than an order of magnitude better than that obtained using white light sources [25] and comparable to that achieved with other comb-based methods [11–13]. However, our approach offers at least 5 orders of magnitude improvement in frequency resolution by directly measuring the cavity transmission spectrum. This in turn enables the measurement of cavity mode shifts induced by molecular transitions, which was previously possible only with cw laser-based techniques [5, 6]. The ability to measure dispersion of entire molecular bands without any prerequisite on the knowledge of the cavity parameters will allow determination of transition line parameters with improved accuracy.

In conclusion, direct measurement of cavity resonance modes using a comb-based Fourier transform spectrometer provides means to measure simultaneously the group delay dispersion...
of cavity mirror coatings, the dispersion of the refractive index of gases, and the resonant
refractivity of entire molecular bands. Complementary information about the molecular
transitions can be obtained from the linewidth and intensity of the cavity modes, opening up
for full characterization of the real and imaginary parts of molecular bands and fundamental
tests of the Kramers-Kronig relations.

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