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# Optimisation of laser linewidth and cavity alignment in off-axis cavity-enhanced absorption spectroscopy

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## HIGHLIGHTS

- A model has been proposed for the estimation of cavity transmittance in off-axis cavity enhanced absorption spectroscopy.
- Cavity enhanced absorptions have been calculated for various laser linewidths and optical beam re-entrant conditions.
- Proposed approach can be used for optimisation and improvement of absorption sensitivity in off-axis CEAS.

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## ABSTRACT

Laser linewidth affects baseline mode structured variations and hence measurement absorption sensitivity in off-axis cavity enhanced absorption spectroscopy with a continuous-wave tunable laser and a stable optical cavity formed by two high reflectivity mirrors. Cavity transmittances have been calculated for various laser linewidths and different optical beam re-entrant conditions for the cavity when overlapping of the optical beams occurs on the cavity mirrors after a finite number of beam round trips within the cavity. It is shown that in order to achieve maximum absorption sensitivity both a specific laser linewidth and specific arrangement of the optical cavity have to be selected and defined using the proposed approach.

## 1. Introduction

An optical stable cavity formed by high reflectivity mirrors has been widely used in order to increase the optical path length in absorption measurements with a tunable laser, whereby the laser beam intensity at the cavity output is recorded and then processed in order to extract an absorption line profile and absorber concentration. The stable cavity can be formed by either two separate spherical or astigmatic mirrors [1,2] or three separate mirrors [3,4]. In the latter case one mirror of the combination is concave spherical or astigmatic. Integrated cavity output spectroscopy (ICOS) with a pulsed light source [5], continuous-wave (cw) cavity enhanced absorption spectroscopy (cw-CEAS) with a continuous-wave light source [6] (sometimes also called continuous-wave integrated cavity output spectroscopy (cw-ICOS)) [7] have been employed to exploit the excitation of multiple transverse and longitudinal $\mathrm{TEM}_{mnq}$ cavity modes [8,9]. If the laser beam enters the cavity through the centre of the first cavity mirror and then travels inside the cavity along the optical axis, the term on-axis cavity-enhanced absorption spectroscopy (on-axis CEAS) is often used. Typical noise-equivalent absorption sensitivities of $10^{-6}$ – $10^{-8}$ $\mathrm{cm}^{-1}\,\mathrm{Hz}^{-1/2}$ have been demonstrated in on-axis CEAS [5,6,10] and were limited by high cavity mode intensity noise.

A further reduction of cavity output intensity fluctuations and noise-equivalent absorption sensitivities of $10^{-11}$ – $10^{-10}\,\mathrm{cm}^{-1}$ $\mathrm{Hz}^{-1/2}$ have been demonstrated in off-axis integrated cavity output spectroscopy (OA-ICOS) [11–14] sometimes called off-axis cavity-enhanced absorption spectroscopy (OA-CEAS). This scheme implies an off-axis alignment of the laser beam relative to the cavity optical axis, i.e. the incident laser beam passes through a point on the cavity mirror (diameter of 7.5 mm up to 50 mm) sufficiently displaced from the cavity mirror centre. In conventional off-axis ICOS a Gaussian beam with the size of the lowest transverse $\mathrm{TEM}_{00}$ cavity mode had been coupled into a stable optical cavity at a finite off-axis angle in order to be refocused to nearly identical spot sizes when bouncing back and forward between the mirrors along various paths. The beam spot pattern (circle, ellipse or Lissajous pattern) on the cavity mirror covers almost the whole

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mirror's aperture [15,16]. It is worth noting that optical beams, which differ from the fundamental mode and have high-order mode profiles with Hermite–Gaussian, Laguerre–Gaussian [9] and even arbitrary transverse distributions, can be also focused into the optical cavity in off-axis alignment and utilised in OA-CEAS. Though for such sophisticated optical beams a model for their propagation between the mirrors, their beam spot centre distribution and an overall field distribution on the cavity mirrors can be developed, but such cases had been not considered to be used in OA-CEAS applications and models.

In OA-CEAS the overall intensity on a cavity output had been conventionally calculated under assumption of effective suppression of cavity resonances and no interference between the overlapping beam spots on the cavity mirrors [11,17]. This is despite common understanding that improved absorption sensitivity and suppression of spurious-coupling intensity noise could be achieved for laser linewidths larger than the free spectral range of the off-axis aligned cavity [11,17–19]. But it was quite often observed in OA-CEAS that if the laser linewidth (LW) and the level of laser beam overlapping on the cavity mirrors are not properly optimised, an excessive level of resonance transmission peaks can be observed within a laser frequency tuning, thus resulting in noisier absorption spectra and poor cavity enhanced absorption sensitivity. In order to reduce cavity output intensity noise in OA-CEAS with tunable lasers and hence to reduce baseline fluctuations and to improve cavity enhanced absorption sensitivity, several different approaches were proposed and exploited in the past. The first approach is based upon wavelength tuning over cavity modes with maximal rate followed by averaging of successive laser wavelength scans [11,13,14,20]. Whereas this configuration can work efficiently with current modulated diode and quantum cascade lasers, its use is limited for external cavity lasers with wide tuning range and a grating mounted on a piezoelectric transducer. The second technique exploits laser LW broadening by laser current modulation with a sinusoidal waveform [12,14,21] or white bandwidth limited radio-frequency noise [21]. The third approach includes a cavity length dithering by means of a cavity mirror mounted on a piezoelectric transducer [12,14,22]. Additionally, cavity output in OA-CEAS with high reflectivity mirrors can be very low. The optimisation of the signal-to-noise ratio of absorption spectra for different laser powers and cavity mirror's reflectivity had been explored in [23,24]. A further increase of the cavity output intensity and hence of the signal-to-noise ratio of spectra were demonstrated by adding a third mirror in front of the first cavity mirror. The beam reflected by the first cavity mirror is thereby re-injected back into the cavity [25,26]. Unfortunately, all these different approaches did not address the question: "Does an optimum combination of laser linewidth and off-axis cavity alignment exist to achieve minimum spurious cavity output intensity variations and hence maximum absorption sensitivity?". The aim of this paper is to develop a model for description of transmittance of a stable optical cavity in OA-CEAS with different laser linewidths and various off-axis beam re-entrant conditions in a two mirror stable cavity for further optimisation and improvement of absorption sensitivity.

## 2. Model

### 2.1. Transmittance of a static stable optical cavity

In some specific arrangements of a stable optical cavity formed by two separated and static (not moving) high reflectivity mirrors with an optical beam incident on the first mirror at a finite angle relative to the optical cavity axis passing through a point on the cavity mirror which is located off the central mirror point, the optical beam re-entrant condition can be fulfilled after an integer number $K$ of round trips (from two passes up to hundreds). After the $K$ round trips of the optical beam between the mirrors, optical beam spots can overlap completely on the mirror surface and the incident and retro-reflected beam directions of propagation can become coincident [1,27,28]. To estimate an overall beam intensity on the cavity output we assume that the two mirrors are specified with the same optical quality and their amplitude transmittance and reflectance are, respectively, denoted by $t_{\mathrm{m1}}$, $t_{\mathrm{m2}}$ and $r_{\mathrm{m1}}$, $r_{\mathrm{m2}}$. The power transmittance and reflectance are given by $T = t_{\mathrm{m1}} \times t_{\mathrm{m1}} = t_{\mathrm{m2}} \times t_{\mathrm{m2}}$ and $R = r_{\mathrm{m1}} \times r_{\mathrm{m2}}$, respectively, whilst for lossless cavity mirrors $T + R = 1$. Under the assumption of an optical field stabilised in amplitude, but with random phase modulation within a time interval $t$, and linear interaction of the optical beam with an absorbing sample, the electric field after travelling through the absorbing gas between the two mirrors is given by

$$E(t,\nu) = E_0 \exp\left(i(2\pi\nu t + \varphi(t))\right) \times \exp(-\delta_A(\nu) - i\varphi_A(\nu)), \tag{1}$$

where $E_0$ is the stabilised electric field amplitude, $\nu$ is the optical frequency, $\varphi(t)$ is the phase that follows a stochastic variation, $\delta_A(\nu)$ is the amplitude attenuation caused by the sample and $\varphi_A(\nu)$ is the accompanying optical phase shift [29,30]. The fluctuations of the optical wave phase $\varphi(t)$ deteriorate the temporal coherence of the optical beam and result in broadening of the optical field bandwidth or linewidth $\Delta\nu$ defined as a half width at half maximum (HWHM). As a consequence the coherence time ($t_{coh} = 1/2\Delta\nu$) and the coherence length ($L_{coh} = \upsilon t_{coh}$, where $\upsilon$ is the light speed in gas medium) decrease with broadening of the optical wave. For ergodic random processes, the time average is equal to the ensemble averages. For an optical spectrum with a Lorentzian lineshape it can be shown [29] that the time average of the optical phases $\varphi(t)$ and $\varphi(t+\tau)$ can be expressed via the linewidth $\Delta\nu$ as

$$\overline{\exp(-i\varphi(t))\exp(i\varphi(t+\tau))} = \exp(-2\pi\tau \times \Delta\nu), \tag{2}$$

where the time delay $\tau \geqslant 0$.

If two mirrors are separated by a distance $h$, then the round trip time $t_r$ is given by

$$t_r = \frac{2h}{\upsilon}. \tag{3}$$

In the case of an ideal geometrical coupling of the optical fundamental Gaussian beam into the cavity, and if the beam does not miss the cavity mirror after an infinite number of round trips and is refocused inside the cavity after each reflection by the mirror, it is reasonable to assume that a square root of a laser beam geometrical coupling parameter $c_g$ is equal to unity. In case of a strongly focused or defocused incident laser beam the geometrical coupling parameter $c_g$ is less than unity. The total electrical field $V(t)$ at the plane behind the second cavity mirror can be expressed as a sum of periodically reflected and transmitted electrical fields

$$\begin{aligned} V(t,\nu) = c_g[&E_0 T \exp(i(2\pi\nu t + \varphi(t))) \times \exp(-\delta_A(\nu) - i\varphi_A(\nu)) \\ &+ E_0 T R \exp(i(2\pi\nu(t - t_r) + \varphi(t - t_r))) \times \exp(-3\delta_A(\nu) - i3\varphi_A(\nu)) \\ &+ \ldots + E_0 T R^{(m-1)} \exp(i(2\pi\nu(t - (m-1)t_r) + \varphi(t - (m-1)t_r))) \\ &\times \exp(-2(m-1)\delta_A(\nu) - i2(m-1)\varphi_A(\nu)) + \ldots]. \end{aligned} \tag{4}$$

After rearrangement we obtain

$$\begin{aligned} V(t,\nu) = {} & c_g E_0 T \exp(i2\pi\nu t) \times \exp(-\delta_A(\nu) - i\varphi_A(\nu)) \times \sum_{m=1}^{\infty} R^{(m-1)} \exp(-i2\pi\nu(m-1)t_r) \\ & \times \exp(i\varphi(t - (m-1)t_r)) \times \exp(-2(m-1)\delta_A(\nu) - i2(m-1)\varphi_A(\nu)). \end{aligned} \tag{5}$$