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Alternative approaches to cavity enhanced absorption spectroscopy

A Cygan*, P Wcisło, S Wójtewicz, P Masłowski, R S Trawiński, R Ciuryło, D Lisak

Institute of Physics, Faculty of Physics, Astronomy, and Informatics, Nicolaus Copernicus University, Grudziadzka 5, 87-100 Torun, Poland

E-mail: agata@fizyka.umk.pl

Abstract. New approaches to cavity enhanced absorption spectroscopy are presented. They are based on precise measurements of shape of cavity modes broadened and shifted in the vicinity of the absorber placed inside the high-finesse cavity. The usefulness of both techniques to accurate line-shape study is demonstrated on an example of a weak $(3 \leftarrow 0)$ $^{13}C^{16}O$ transition.

1. Introduction

A great potential of cavity-enhanced absorption spectroscopy (CEAS) techniques in application to ultrasensitive measurements of absorption grew up from significant elongation of optical path of light inside the high-finesse cavity. One of the well known variants of CEAS is the cavity ring-down spectroscopy (CRDS) which is based on variation of lifetime of the laser light in the optical cavity with absorbing medium. This lifetime can be measured from the ring-down decay time constant and is insensitive to the laser power fluctuations. It is also directly related to the spectral broadening of the cavity resonant modes. This phenomenon was originally observed by Nakagawa *et al.* [1] in 1994, however until recently it was not exploited for spectroscopy. Development of laser stabilization technique [2–4] allows measurement of the width of the single mode of the ultra stable cavity with high accuracy [5]. Very recently experimental spectrum based on cavity mode widths determination was demonstrated by Long *et al.* with the use of the FARS technique [6, 7] and by Cygan *et al.* [8] who used an ultranarrow and tunable in the range of 40 GHz external cavity diode laser to measure a weak oxygen transition. Such approach to precise measurements of cavity mode widths inspired a development of a new CEAS technique called cavity mode-width spectroscopy (CMWS) [7, 8]. Qualitative and intuitive comparison of CRDS and CMWS methods presented in [8] reveals theirs complementarity in the sense that they achieve their best precision in different pressure ranges. For low absorptions the best precision is achieved with the CRDS technique, where the ring-downs are long and decay constants can be well determined. In the opposite case, where the absorption is high and cavity modes become broader, the precision of CMWS is enhanced.

In this paper we present precise CMWS spectrum measured for a P3 $(3 \leftarrow 0)$ $^{13}C^{16}O$ transition. Contrary to our previous measurement technique of an experimental setup described in [8] here we measured cavity mode widths relative to the neighboring cavity mode, to which the probe laser is locked. We also present dispersive spectrum of $^{13}C^{16}O$ line which is a result of measurement of cavity modes pulling caused by a dispersive shift of light near the $^{13}C^{16}O$

absorption line. In contrast to the NICE-OHMS (Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy) technique [9] which is based on amplitude measurements of dispersion profiles, in this work to obtain dispersive spectrum only frequency domain quantities were used.

2. Absorptive broadening and dispersive pulling of cavity modes

Near the resonance transition of the absorbing medium placed inside the high-finesse cavity one may observe two characteristic phenomenon - absorptive broadening and dispersive shift of cavity modes. Both these effects can be described by an extended formula for electromagnetic field power transmitted through the cavity given in the form [8]

$$I_T(\nu) = I_0 \frac{(1-R)^2 e^{-\alpha(\nu)L}}{(1-Re^{-\alpha(\nu)L})^2 + 4Re^{-\alpha(\nu)L}\sin^2(\frac{\pi\nu}{\nu_{\mathrm{FSR}}(\nu)})}, \tag{1}$$

where I_0 is the incident field power, R is the intensity reflectivity of the cavity mirrors and L is the cavity length. Quantities $\alpha(\nu)$ and $\nu_{\mathrm{FSR}}(\nu)$ are the absorption coefficient and free spectral range of the cavity, respectively, and they are related to imaginary $\kappa(\nu)$ and real $n'(\nu)$ part of the complex refractive index, given generally in the form $n_a(\nu) = n'(\nu) - i\kappa(\nu)$, by equations: $\alpha(\nu) = 2k_0\kappa(\nu)$ and $\nu_{\mathrm{FSR}}(\nu) = c/(2n'(\nu)L)$, where k_0 is the value of a wave vector for the resonance transition and c is the speed of light. In case when the absorption coefficient $\alpha(\nu)$ is described by a Lorentzian line shape, formulas for $n'(\nu)$ and $\kappa(\nu)$ are simply given by Kramers-Krönig relations [10]. Using Kramers-Krönig relations we can write that

$$n'(\nu) = n + cNS\ \frac{\mathrm{Im}[\mathcal{I}(\nu)]}{2k_0}, \tag{2}$$

$$\kappa(\nu) = cNS\ \frac{\mathrm{Re}[\mathcal{I}(\nu)]}{2k_0} \tag{3}$$

in which n describes the value of refractive index far from the resonance transition, N is the number density of the absorber expressed in units molecule/cm^3, S is the line intensity in units cm^{-1}/(molecule/cm^2) and $\mathcal{I}(\nu)$ is a normalized complex line-shape function, i.e. $\int \mathcal{I}(\nu)d\nu = 1$. According to above relations Eq. (2) describes pulling of the cavity modes and hence the dispersive shape of the investigated transition, whereas Eq. (3) is related to broadening of cavity modes and an absorption line shape usually measured in CEAS experiments. Since presence of absorbing medium inside the cavity shortens the lifetime of laser light inside the cavity, it is clear that also widths of cavity modes have to become broader according to the Fourier transform relations. In this way frequency domain picture used in CMWS and time domain picture used in CRDS can be simply related by the formula $\delta\nu_m = 1/(2\pi n\tau)$ where $\delta\nu_m$ and τ are the main quantities measured in both these techniques, i.e. the cavity mode width and time constant of the ring-down decay, respectively.

3. Experiment and results

In order to precisely measure absorptive (CMWS) and dispersive spectra of P3 ($3 \leftarrow 0$) ^{13}C^{16}O line presented in Fig. 1. we designed an experiment independent of any cavity length changes which were the main source of noise in our previous measurements of an oxygen line [8]. In our new system the probe laser is tightly locked by the Pound-Drever-Hall [11] method to the cavity. In this way the slow drift of the cavity length as well as cavity vibrations are transferred to the probe laser, but the overlap between laser and cavity mode frequencies is maintained constantly during cavity mode widths and shifts measurements. Our spectrometer uses two-beams configuration system in which the probe laser beam is divided into two beams: one

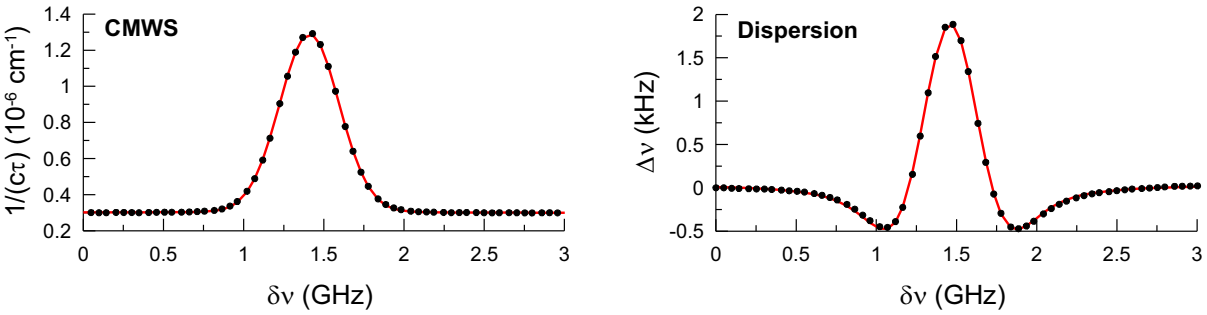

Figure 1. Absorptive (CMWS) and dispersive spectrum of the $(3 \leftarrow 0)$ $^{13}C^{16}O$ line recorded at pressure 4.8 Torr by the CMWS and dispersive techniques, respectively. Both spectra were compared with the Voigt profile.

,,lock beam" used for PDH locking of the probe laser to the cavity and the second one ,,probe beam" used for probing cavity mode widths and shifts. Both beams are mode matched to two consecutive longitudinal modes of the probe cavity. The relative scan of the ,,probe beam" around the cavity mode is realized by frequency tuning of the RF signal driving an acoustooptic modulator inserted into the optical path of the ,,probe beam". The Voigt profile commonly used in spectroscopic databases was used for comparison with absorptive (CMWS) and dispersive line shapes presented in Fig. 1. Closer look at experimental results in case of both methods suggests that the Voigt profile is not good enough to reproduce experimental data with high accuracy.

4. Conclusions

In this study two alternative approaches to cavity-enhanced absorption spectroscopy, namely CMWS and dispersive method, were proposed. We tested these new methods on the CO rovibrational transition from the $(3 \leftarrow 0)$ band, which is located in the spectral region around 6201 cm^{-1}. Both methods indicated that the Voigt profile is too simple model to describe the investigated line shape with high accuracy. Comparison of various CEAS techniques is presently of great importance for the line-shape analysis of experimental spectra, where one of the most challenging task is to distinguish the systematic errors due to experimental imperfections from these caused by the wrong choice of the line-shape model, see e.g. [12].

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