

Prism-based Broadband Optical Cavity (400 – 1600 nm) for High-Sensitivity Trace Gas Sensing by Cavity Enhanced Absorption Spectroscopy





Gaoxuan Wang^{1,}, Lingshuo Meng¹, Qian Gou², Azer Yalin³, Tong Nguyen Ba¹, Cécile Coeur^{1,}, Alexandre Tomas⁴, Weidong Chen¹

¹ Laboratoire de Physicochimie de l'Atmosphère, Université du Littoral Côte d'Opale, Dunkerque, France ² School of Chemistry and Chemical Engineering, Chongqing University, China ^{3.} Department of Mechanical Engineering, Colorado State University, Fort Collins, CO 80523, USA ⁴ IMT Lille Douai, Département Sciences de l'Atmosphère et Génie de l'Environnement, France

* Email : chen@univ-littoral.fr





Introduction The use of high-reflectivity dielectric mirrors to form an optical cavity allows one to achieve effective optical path lengths of up to several kilometres for high-sensitivity spectroscopy applications [1,2]. However, the high reflectivity of a dielectric mirror is obtained via constructive interference of the Fresnel reflection at the interfaces produced by multilayer coatings of alternate high and low refractive index materials. This wavelength-dependent coating limits the bandwidth of the mirror's high reflectivity to only a few percent of the designed central wavelength [3]. In this work, we report the development of a novel optical cavity based on prism in CaF₂ used as cavity reflector through total internal reflection combined with Brewster angle incidence [4,5], which offers a high-finesse optical cavity operating in a broadband spectral region from 400 to 1600 nm. Cavity Enhanced Absorption Spectroscopy (CEAS) of NO₂ and H₂O vapour was applied to determine the prism reflectivity and to validate its broadband spectral capability.

Prism-cavity based on total internal reflection : material losses limited reflectivity





Prism-based cavity consists of two prism reflectors, which rely on total internal reflection of light beam at near the Brewster angle incidence. Light L_{in} is coupled into the cavity at R_{4} and coupled out (L_{out}) at R₁. All surfaces are flat with exception to EF, which has a 6 m convex curve [5]. Effective prism reflectivity is controllable by tuning the beam incident angel around the Brewster angle ($\theta_{\rm B}$), within ~ 1°.



Modeled braodband high-reflectivity with a Brewster angle optimized for λ =1000 nm

Experimental demonstration of broadband cavity performance



✓ Laser @ 1074.6 nm with 1.2 mW P component light

- ✓ CRD decay time : 2.65 μ s => R = 99.925% (by CRD)

BBCEAS measurement of NO₂ absorption using diode lasers at 405, 450, 532 nm



BBCEAS spectra $I_0(\lambda)$ and $I(\lambda)$ of N_2 and NO_2

Linear dependence of $(I_0-I)/I$ vs. NO₂ concentration

In Broadband CEAS (BBCEAS), gas absorption coefficient is expressed :

$$\alpha = \left(\frac{I_0}{I} - 1\right) \times \left(\frac{1 - R}{d} + \alpha_{Ray, N2}\right) = \sum_i \sigma_i(\lambda) \times N_i$$

where d is the cavity length; R the prism reflectivity; $\alpha_{Rav,N2}$ the gas Rayleigh scattering; $\sigma_i(\lambda)$ and N_i are the reference absorption cross section and number density of i species.

Using known concentration N of calibration gas, prism reflectivities were deduced : 405 nm : 99.48±0.01% 450 nm : **99.58 ± 0.01%** 532 nm : **99.77**±0.02%

CEAS measurement of H₂O at 1074.6 nm & 1500 – 1520 nm



H₂O vapour absorption spectra at 1074.6 nm in a multipass cell (L_{eff}=152 m) and the prism cavity

H₂O absorption simulation vs. experimental spectrum in the prism cavity over 1500 - 1520 nm

- CaF, prism reflectivity : 99.917 ± 0.004% (vs. 99.925% from CDR) @ 1074.6 nm deduced from the H₂O vapour absorption spectrum whose concentration was determined by direct absorption spectroscopy in a multipass cell (L_{eff}=152 m).
- CaF, prism reflectivity : 99.972 ± 0.001% at 1507.25 nm determined from the measured high-resolution H_2O absorption spectra over 1500-1520 nm.
 - Prism offering material losses α limited reflectivities (R_{ideal}=1- α) : CaF₂ prism

Summary

Measured CEAS spectra of NO₂ in the visible and of H_2O vapour in the near-infared (NIR) using the developed CaF₂ prism-cavity demonstrated its broadband capability and allowed for determination of its reflectivities : 99.48% (405 nm), 99.58% (450 nm), 99.77% (532 nm), 99.917% (1074.6 nm) and 99.972% (1507.25 nm).

The prism reflectivity is mainly limited by its optical losses including bulk absorption & scattering, surface scattering, residual birefringence induced losses (converting P to S polarization) due to either strain or misalignment of the optic axis of the prism substrate material, and fraction loss related to incident angle's deviation from the Brewster's angle. Further improvement using lower losses SiO₂ prism : ~50 ppm @ 1570 nm in the NIR.

losses of ~500 ppm at 500 nm [5]

R_{ideal} ~ 99.95% (@ 500 nm) vs. R_{exp} ~ 99.77% (@ 532 nm)

References

[1] S. S. Brown, Chem. Rev. 103 (2003) 5219-5238. [2] M. Mazurenka, A. J. Orr-Ewing, R. Peverallb and G. A. D. Ritchie, Annu. Rep. Prog. Chem. Sect. C 101 (2005) 100-142. [3] G.R. Fowles, *Introduction to Modern Optics* (Rinehart and Winston, 1975), p. 328. [4] P. S. Johnston and K. K. Lehmann, Opt. Express 16 (2008) 15013-15023. [5] B. Lee, K. Lehmann, J. Taylor and A. Yalin, Opt. Express **22** (2014) 11583-11591. **Acknowledgements** This work is supported by the French national research agency (ANR) under MABCaM (ANR-16-CE04-0009) and CaPPA (ANR-10-LABX-005) contracts.