

Intracavity absorption spectroscopy of HCI isotopes, H_2O , CH_4 , C_2H_4 , and C_2H_6 in the 3.1–3.4 µm spectral range using a Cr:CdSe laser

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Abstract: We demonstrate the first application of a Cr:CdSe laser for highly-sensitive multicomponent intracavity absorption spectroscopy around $\lambda = 3.1-3.4 \,\mu\text{m}$. A detection scheme based on an integrated recording of multiple (~70) individual Cr:CdSe laser pulses after a single pump-pulse excitation is reported. The sensitivity of our system corresponds to an effective absorption path length of $L_{\text{eff}} \approx 850 \,\text{m}$. Exemplary measurements of atmospheric H₂O and CH₄, and additionally introduced gas-phase HCl, C₂H₄, or C₂H₆ are presented. The achieved noise-equivalent detection limits are in the ppb range. Possibilities for further sensitivity enhancement by up to a factor of 10⁴ are discussed.

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1. Introduction

A general trend observable in laser spectroscopy in the recent years is the development and application of mid-infrared (MIR) coherent light sources [1,2] that are capable of addressing fundamental rovibrational transitions of various species [3]. For simultaneous multi-species detection, utilization of broadband MIR laser sources is advantageous compared to systems based on multiple narrow-band lasers. This is one of the reasons why MIR frequency combs that offer broad spectral coverage, high spectral (or high time-) resolution, and a direct link to radio-frequency standards, become increasingly popular as spectroscopic tools [4–7]. These systems, however, are based on the conventional spectroscopic scheme light source \rightarrow sample \rightarrow *detector*, resulting in comparably small effective absorption path lengths ($L_{\rm eff}$, i.e., sensitivities) that are basically determined by the sample size. Although L_{eff} can be increased up to several kilometers by utilizing cavity-enhanced schemes as described, e.g., in Ref. [8], the necessary stabilization scheme makes the experiment even more complex than a regular frequency-comb setup. Furthermore, the corresponding locking of comb and cavity seems to work properly only in clean and unperturbed environments. The fact that there is almost no literature on cavity-enhanced frequency-comb measurements in challenging environments (e.g., in combustion), that go beyond proof-of-principle demonstrations, as, e.g., in Ref. [9], suggests that this complex task is not yet sufficiently mastered. Therefore, alternative broadband and highly-sensitive MIR diagnostics tools are required, especially for challenging environments. One such technique is intracavity absorption spectroscopy (ICAS) [10].

In contrast to the conventional spectroscopic scheme, with ICAS the sample is placed inside the laser resonator. The successive interaction of laser photons with the broadband laser gain and the sample's narrow-line absorption determines the laser emission spectrum. While broadband losses

(e.g., scattering and broadband absorption or beam-steering) are compensated by the laser gain medium, the narrow-line absorption originating from the sample is not compensated. The origin (and prerequisite) of this mechanism is the gain-competition of laser modes (frequencies) within the homogeneous linewidth of the gain medium. Laser modes that coincide with absorption lines of the sample experience additional losses and thus are less amplified when passing through the gain medium. With each roundtrip inside the resonator this effect accumulates thus leading to an imprint of the absorption spectrum of the sample directly onto the broadband laser emission spectrum. Since the laser light passes through the sample many times, this results in extremely large effective absorption path lengths of up to $L_{eff} = 7 \times 10^7$ m [11,12], thus enabling ultra-sensitive detection. Depending on the gain medium and operation parameters of the laser, such processes as four-wave mixing and Rayleigh and Brillouin scattering typically set the upper limit of the maximum available sensitivity. These processes circumvent the mode competition by redistributing laser photons from modes without absorption to modes with absorption [10]. As a consequence, these processes determine a certain saturation time t_s that marks the maximum sensitivity of an ICAS system, being equal to the sensitivity in continuous-wave (CW) operation. In pulsed operation with pulses shorter than t_s , the spectral sensitivity can be determined from the laser generation time t as $L_{\text{eff}} = ct$, with c being the speed of light.

With this, ICAS combines several advantages in a unique way: (i) ultra-high sensitivity originating from extreme absorption path lengths, (ii) robustness against broadband losses that are compensated by the laser gain medium, and (iii) capability of simultaneous monitoring of multiple transitions or species with a single light source due to the employment of broadband and widely tunable lasers.

Since ICAS was proposed about five decades ago [13], it has been the subject of several reviews [10,14] and was exploited for various spectroscopic tasks, including atmospheric monitoring [15], medical breath analysis [16], shock tube kinetics [17,18], flames [19,20], and plasmas [21,22]. However, despite these promising results achieved with ICAS mostly in the visible and near-infrared in the recent years, there is still a great but barely used potential for significant progress in this field, namely the advancement into the MIR spectral range, where the fundamental vibrational transitions of many species are located.

Previous work on ICAS in the MIR spectral range includes the employment of Co:MgF₂ lasers around 2 μ m [23–25], a Tm/Ho fiber laser around 2.1 μ m [16], Cr:ZnSe lasers around 2.5 μ m [26–29], color-center lasers around 2.7 μ m [30], and Fe:ZnSe lasers in the 3.7–5.3 μ m range [15,31].

In this work, we demonstrate the first applications of a homemade Cr:CdSe laser for ICAS measurements of HCl, H₂O, CH₄, C₂H₄, and C₂H₆ in the 3.1–3.4 μ m spectral range. In general, Cr:CdSe lasers provide access to the spectral range of 2.2–3.6 μ m [32,33], the long-wavelength part of which is complementary to the Cr:ZnSe and Fe:ZnSe laser emission ranges [34]. From a spectroscopic point of view, the Cr:CdSe laser is highly interesting since its emission range enables accessing absorption lines of such species as, e.g., H₂O, CO₂, O₃, N₂O, CO, CH₄, NO₂, NH₃, OH, HF, HCl, OCS, H₂CO, HO₂, HOCl, HCN, CH₃Cl, C₂H₂, C₂H₆, C₂H₄, HC₃N, and other hydrocarbons [3]. Sensitive multicomponent ICAS measurements of several of these species are also demonstrated in the current work.

2. Experimental details

The experimental scheme of the Cr:CdSe laser system is shown in Fig. 1. The cylindrical plane-parallel Cr:CdSe single-crystal active element used in this work has a diameter of 27 mm and a thickness of 8.8 mm with a doping concentration of 1.7×10^{18} cm⁻³. The laser crystal has been grown at the P.N. Lebedev Physical Institute of the Russian Academy of Sciences using the physical vapor-transport technology with simultaneous Cr²⁺-ion doping [33,35].



Fig. 1. Experimental setup. M1: highly reflective concave mirror; M2: output coupler; PD: photodetector; EM: energy meter; A1: aluminum mirror; Pyrocam: pyroelectric camera.

The uncoated active element is placed under the Brewster angle ($\theta_B = 67.8^\circ$) inside the laser cavity defined by the highly-reflective concave mirror M1 (r = 400 mm) and a plane output coupler M2 (3° wedge) with a transmission of about T = 8% in the spectral range of $\lambda = 3.1-3.5$ µm. The optical distance between M1 and M2 is L = 295 mm. The active element is placed close to M2, resulting in a good overlap of the pump and laser beams. As a pump source we use a homemade flash-lamp pumped Er: YAG (5% doping) laser emitting 150-µs pulses (FWHM) around $\lambda_P = 1.78$ µm [36] with a maximum pulse energy of 200 mJ. The pump laser is operated in a single-pulse regime. The pump light is focused onto the Cr:CdSe crystal with a spot size of about 1.5 mm. The angle between the resonator axis and the pump beam is about 0.03 rad.

The pump and Cr:CdSe laser pulse-shapes and energies are measured using photo detectors (PDs in Fig. 1) and energy meters (EMs) that have been described earlier [33]. An open tube (gas cell) with a diameter of d = 14 mm and a length of l = 170 mm is installed in the resonator for ICAS measurements in gas mixtures or flows.

The spectral tuning of the Cr:CdSe laser is performed by using an intracavity CaF₂ prism (apex angle of 70°) and tilting M1 appropriately. With the current setup the laser is continuously tunable in the spectral range of 3.1–3.4 µm. Emission spectra of the Cr:CdSe laser are recorded using a homemade grating spectrometer with a maximum spectral resolution of $\delta v = 0.15$ cm⁻¹ (corresponding to $\delta \lambda = 0.15$ nm at $\lambda = 3.2$ µm). As a detector, a pyroelectric array camera Pyrocam IIIHR (MKS/Ophir, 160 × 160 pixels, 12.8 × 12.8 mm² active area) is used. The pump laser is synchronized with the camera using a pulse generator (not shown in Fig. 1).

3. Characterization of the ICAS system

Figure 2 shows oscillograms of the pump (green) and Cr:CdSe (red, shifted along the *y*-axis) laser pulses. An irregular spike structure of the pump-laser pulse can be observed, which is typical for multimode solid-state lasers with long upper-state lifetimes.

After adjusting the total pump-pulse energy to about 120 mJ, the intensity minima of almost all pump spikes fell below the threshold of the Cr:CdSe laser, thus resulting in a pulse-train emission of about n = 70 individual pulses. In other words, the ~50% modulation-depth of the pump-laser pulse is converted into a full modulation depth of the Cr:CdSe laser, i.e. into individual pulses with pulse widths (FWHM) of about $\Delta t_p = 2-4 \,\mu s$ (inset). The fluctuation of individual pulse widths transforms into a variation of individual absorption path lengths $L_{\text{eff,n}}$. However, by integrated spectral recording of the whole Cr:CdSe pulse-train, an effective averaging over n individual emission spectra is achieved, with a resulting averaged L_{eff} that can be determined from a calibration measurement. The pulse-train to pulse-train fluctuation of the averaged L_{eff} is found to be on the order of 10%, mainly due to pump-energy fluctuations.

The main advantage of the integrated recording is the reduction of spectral-noise by a factor of \sqrt{n} , thus enabling the acquisition of a low-noise Cr:CdSe spectrum after a single pump-pulse



Fig. 2. Oscilloscope traces of the pump (green) and Cr:CdSe (red) laser pulses. The inset shows a zoomed section of the Cr:CdSe pulse train.

excitation. It should be noted that this measurement concept is demonstrated for the first time and might be especially useful for spectroscopic systems based on excitation sources that are not capable of producing short enough pulses. For future experiments it is feasible to develop an optimized pump source (i.e., with optimized pulse characteristics) to enable addressing specific experimental needs, in particular the pulse width Δt_p of individual Cr:CdSe laser pulses (determining L_{eff}) and the number of individual pulses *n* (determining the spectral noise).

Prior to specific ICAS measurements, the spectroscopic system has been characterized regarding emission bandwidth and sensitivity (i.e., L_{eff}). To record the emission bandwidth of the broadband Cr:CdSe laser, the prism has been removed, the spectral resolution of the recording system has been set to $\delta v = 0.5$ cm⁻¹, and a series of eight overlapping spectra has been recorded by stepwise tuning the spectrometer. Subsequently, these spectra have been normalized by the maximum intensity and combined to one spectrum, as described in [19]. The result is shown in Fig. 3 (bottom).

As can be seen, the broadband emission spectrum of the Cr:CdSe laser is centered around $\nu = 3160 \text{ cm}^{-1}$ ($\lambda = 3.16 \mu \text{m}$), which is shifted from the typical emission center at $\nu = 3770 \text{ cm}^{-1}$ (2.65 µm) due to the reflectivity profile of the output coupler M2. The large emission bandwidth of about $\Delta \nu = 140 \text{ cm}^{-1}$ (measured at 10% of the maximum laser intensity) is especially suitable for simultaneous multi-species or multi-line monitoring. The insertion of the CaF₂ prism leads to a slight reduction of the emission bandwidth to about $\Delta \nu = 120 \text{ cm}^{-1}$, which can be spectrally shifted (tuned) within the spectral range of $\nu = 2940-3230 \text{ cm}^{-1}$ ($\lambda = 3.1-3.4 \mu \text{m}$).

The wavelength calibration has been performed using well-known line positions of atmospheric H₂O and CH₄ from the HITRAN database [3]. Corresponding calculated spectra with partial pressures of $p_{\rm H2O}$ = 8 mbar and $p_{\rm CH4}$ = 0.002 mbar (2 ppm) are shown in the center and top diagrams of Fig. 3. To derive absolute species concentrations from the normalized measured spectrum, the flat central part of the emission spectrum has been fitted with a transmission spectrum from HITRAN by varying the absorption path length $L_{\rm eff}$ in the calculation (under consideration of the spectral resolution of our recording system). In the first step, this has been done for CH₄ lines only, since the humidity in the lab varies on a daily basis, while the CH₄ concentration is assumed to remain constant at about 2 ppm [37]. As a result, $L_{\rm eff}$ = 800 m has been obtained by visually comparing superimposed experimental and theoretical spectra. The accuracy of this procedure is about ±5%. The determined $L_{\rm eff}$ has then been used to deduce the actual concentration of H₂O, being $p_{\rm H2O}$ = 8 mbar here, by varying $p_{\rm H2O}$ in the calculation until



Fig. 3. Bottom: Emission spectrum of the Cr:CdSe laser recorded without a prism in the resonator. Center and top: Calculated transmission spectra of atmospheric H₂O and CH₄ using the HITRAN database, with $L_{\text{eff}} = 800 \text{ m}$ and $\delta v = 0.5 \text{ cm}^{-1}$.

the visually best agreement with the experiment has been found. This method is used throughout the current work.

After this characterization, the spectral resolution of the recording system has been set to the maximum, i.e., to $\delta v = 0.15$ cm⁻¹, and ICAS measurements of several exemplary gas-phase species have been performed in different spectral regions. The results are presented in the following sections.

4. ICAS measurements

4.1. Isotopes of HCI

To generate a small concentration of HCl in the laser cavity, a droplet of aqueous solution of HCl has been released into the gas cell. During the subsequent evaporation, ICAS spectra with HCl absorption have been recorded. An example spectrum is shown in Fig. 4 (bottom, red).

To fit a calculated spectrum to the measured one, the following parameters have been used: $L_{\text{eff}} = 800 \text{ m}, \delta \nu = 0.15 \text{ cm}^{-1}, p_{\text{CH4}} = 2 \text{ ppm}$ and $p_{\text{H2O}} = 10 \text{ mbar}$. The additional concentration of HCl in the cavity has been determined to be $p_{\text{HCl}} = 7 \text{ ppm}$ with the partial concentrations of the main isotopes H³⁵Cl and H³⁷Cl being $p_{\text{H35Cl}} = 5.3 \text{ ppm}$ and $p_{\text{H37Cl}} = 1.7 \text{ ppm}$. The cumulative calculated spectrum of H₂O, CH₄, and HCl is shown in Fig. 4(B), revealing good agreement with the experiment. Diagrams C, D, E, and F show calculated spectra of individual relevant species with their corresponding concentrations. For more clarity, vertical dashed lines mark absorption lines of CH₄, H³⁵Cl, and H³⁷Cl.

With the estimated noise level in the measured spectra of about 3% (RMS), the following noise-equivalent detection limits have been determined: $p_{min,H2O} = 7$ ppm, $p_{min,CH4} = 52$ ppb, $p_{min,H35Cl} = 59$ ppb and $p_{min,H35Cl} = 49$ ppb. These results already demonstrate the capability of our ICAS system to perform broadband, highly-sensitive multicomponent measurements, exceeding the performance of modern spectroscopic systems, e.g., based on frequency combs operating in a similar spectral range [6,7]. Furthermore, it should be noted that the detection limits reported in the current work can be improved significantly by (i) noise reduction of at least one order of magnitude when averaging over a larger number of individual pulses [10,38],



Fig. 4. A: Emission spectrum of the Cr:CdSe laser with absorption of atmospheric H_2O and CH_4 and of HCl from the gas cell. B: Calculated spectrum of H_2O , CH_4 and HCl. Diagrams C, D, E, and F show individual calculated spectra of the relevant species with the corresponding concentrations.

and (ii) by increasing the duration of individual Cr:CdSe laser pulses to several milliseconds, as demonstrated with similar solid-state lasers based on Cr:forsterite [12] and Cr:ZnSe [29] crystals. With this, at least a factor of 10⁴ of sensitivity enhancement can be expected after corresponding modifications of the current ICAS setup, thus enabling detection limits in the low ppt range.

4.2. Ethylene (C_2H_4)

Measurements of C_2H_4 have been performed in the spectral ranges around 3000 cm⁻¹ and around 3160 cm⁻¹. For this purpose, a highly diluted mixture of about 10 ppm of C_2H_4 in N₂ has been prepared in a gas cylinder. This gas mixture has been injected into the gas cell for a short time and, subsequently, ICAS measurements with suitable (i.e., not saturated) absorption signals of C_2H_4 have been performed. Figure 5(A) shows such spectra recorded in two different spectral regions.

The measured spectrum has been fitted using the HITRAN database. The resulting best-fit parameters are as follows: $L_{\text{eff}} = 800 \text{ m}$, $\delta v = 0.15 \text{ cm}^{-1}$, $p_{\text{CH4}} = 2 \text{ ppm}$, $p_{\text{H2O}} = 10 \text{ mbar}$ and $p_{\text{C2H4}} = 10 \text{ ppm}$. The corresponding cumulative calculated spectrum of H₂O, CH₄, and C₂H₄ is presented in Fig. 5(B), showing good agreement with the experiment. For comparison, diagrams C, D, and E show calculated spectra of individual relevant species with their corresponding concentrations. The noise-equivalent detection limit for C₂H₄ is determined to be $p_{\text{min,C2H4}} = 320$ ppb.



Fig. 5. A: Emission spectra of the Cr:CdSe laser recorded in two different spectral regions with absorption of atmospheric H_2O and CH_4 and of C_2H_4 from the gas cell. B: Calculated combined spectrum of H_2O , CH_4 , and C_2H_4 . Diagrams C, D, and E show individual calculated spectra of the relevant species with the corresponding concentrations.



Fig. 6. A: Emission spectrum of the Cr:CdSe laser with absorption of atmospheric H_2O and CH_4 and of C_2H_6 from the gas cell. B: Calculated combined spectrum of H_2O , CH_4 , and C_2H_6 . Diagrams C, D, and E show individual calculated spectra of the relevant species with the corresponding concentrations.

4.3. Ethane (C_2H_6)

Similar to the experiments with C_2H_4 discussed in the previous section, measurements of C_2H_6 have been performed in the spectral range around 3000 cm⁻¹. Figure 6(A) shows an exemplary ICAS spectrum with atmospheric absorption and additional C_2H_6 in the gas cell.

The experimental spectrum has been fitted using the HITRAN database. The resulting best-fit parameters are as follows: $L_{\text{eff}} = 900 \text{ m}$, $\delta v = 0.15 \text{ cm}^{-1}$, $p_{\text{CH4}} = 2 \text{ ppm}$, $p_{\text{H2O}} = 15 \text{ mbar}$ and $p_{\text{C2H6}} = 600 \text{ ppb}$. The cumulative calculated spectrum of H₂O, CH₄, and C₂H₆ is presented in Fig. 6(B), showing good agreement with the experiment. For comparison, diagrams C, D, and E show calculated spectra of individual relevant species with their corresponding concentrations. The noise-equivalent detection limit for C₂H₆ is determined to be $p_{\min,\text{C2H6}} = 13 \text{ ppb}$.

5. Conclusions

In this work, the first application of a Cr:CdSe laser for highly-sensitive multicomponent ICAS measurements is reported. In particular, an emission bandwidth (i.e., simultaneous spectral coverage) of up to $\Delta v = 140 \text{ cm}^{-1}$ is demonstrated, while the tuning range for a Cr:CdSe laser can be about v = 2780-4545 cm⁻¹ ($\lambda = 2.2-3.6$ µm), thus enabling spectroscopic investigations of a variety of species. A detection scheme involving an integrated (averaged) recording of about 70 individual Cr:CdSe laser pulses after a single pump-pulse excitation is demonstrated for the first time. The sensitivity of the newly developed ICAS system currently corresponds to an effective absorption path length of $L_{\rm eff} \approx 850$ m. Exemplary measurements of atmospheric H₂O and CH₄, and additionally introduced HCl, C2H4, or C2H6 are presented. The achieved detection limits in the investigated spectral regions are: $p_{\min,H2O} = 7$ ppm, $p_{\min,CH4} = 52$ ppb, $p_{\min,H35Cl} = 59$ ppb, $p_{\min,H35Cl} = 49$ ppb, $p_{\min,C2H4} = 320$ ppb, and $p_{\min,C2H6} = 13$ ppb. However, it should be noted that these detection limits can be further improved significantly by (i) noise reduction of at least one order of magnitude when averaging over a larger number of individual pulses, and (ii) by increasing the duration of individual Cr:CdSe laser pulses to several milliseconds. With this, at least a factor of 10^4 of sensitivity enhancement can be expected after corresponding modifications of the current ICAS setup, thus enabling detection limits in the lower parts per trillion (ppt) range.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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