Background-Free Absorption Spectroscopy on Methane using Mode-Locked Cr:ZnS Laser

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I. Introduction

Infrared absorption spectroscopy is a powerful method for identifying and detecting trace amounts of molecules. However, the traditional absorption-type method has strict requirements to the stability of the light source because absorption dips may be buried in fluctuations of the laser output. In contrast, background-free (BF) methods have a great potential for ultrasensitive detection because the signal scales with the irradiance of the light source.[1]. In this study, we utilize our recently developed mode-locked Cr:ZnS laser to demonstrate BF spectroscopy based on coherent suppression of the background light with a Michelson interferometer for the $\nu_3 + \nu_4$ band of methane.

II. Experiment

Our experimental setup is shown in Fig. 1. We used a Cr:ZnS pulsed laser (2.3 μm, 60 fs, 40 MHz) described detailed in [2]. The background light was suppressed through a Michelson interferometer, whose optical arms were ideally balanced in phase and group delay. Note that the phase difference π came from the internal and external reflection of the coated beamsplitter. In order to stabilize the destructive interference, we used a feedback loop circuit driven by a collinear He-Ne laser. The BF signal was measured by a monochromator (MS2004i) and InGaAs detector (DET10D) along with the chopper and a lock-in amplifier.

III. Results

The raw spectrum of the BF signal with 0.4 nm spectral resolution is shown with blue line in Fig. 2. For comparison, the absorbance spectrum measured by commercial FTIR spectroscopy (FT/IR-4200typeA) is plotted with red line. We find that the amplitude and position are clearly corresponding to each other. We estimate the SNR of the BF signal of 170, which is several-times enhanced compared to the traditional absorption spectroscopy.

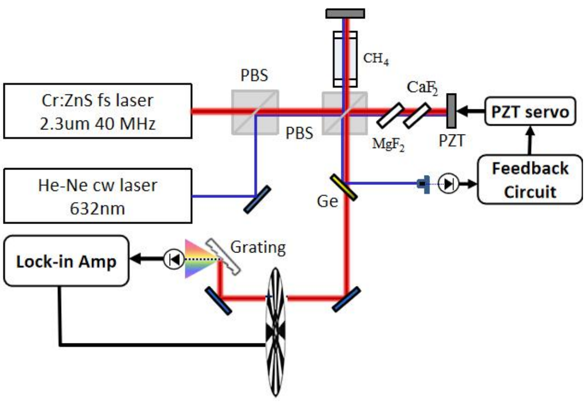

Fig. 1 The experimental setup of BF spectroscopy.

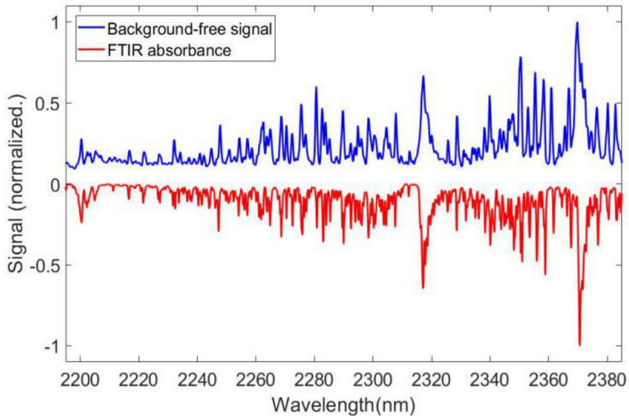

Fig. 2 The measured vibrational signals of methane for the BF measurement (blue) and a commercial FTIR (red).

[1] T. Buberl et al., Opt. Express 27, 2432-2443.(2019) [2] D. Okazaki et al., Opt. Lett. 44, 1750-1753 (2019).