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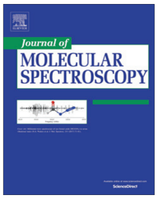

Transition dipole-moment of the $\nu_1 + \nu_3$ band of acetylene measured with dual-comb Fourier-transform spectroscopy

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ABSTRACT

The $\nu_1 + \nu_3$ vibration band of acetylene (C_2H_2) in the near infrared region was recorded with a dual-comb Fourier-transform spectrometer. We observed 56 transitions from $P(26)$ to $R(29)$ at six different column densities. The integral line intensity was determined for each recorded absorption line by fitting the line profile to Lambert-Beer's law with a Voigt function. Thanks to the outstanding capability of dual-comb spectroscopy to cover a broad spectrum in a relatively short time with high resolution and high frequency precision, we determined the reliable line strength for each ro-vibrational transition as well as the transition dipole moment for this band.

1. Introduction

Dual-comb Fourier-transform spectroscopy (DCS) is an attractive tool for precision spectroscopy [1,2]. Using a homemade dual-comb spectrometer operated in the near infrared region at wavelengths of 1.0 to 1.9 µm, we have observed the $\nu_1 + \nu_3$ and $3\nu_3$ vibration bands of acetylene (C_2H_2), the $2\nu_3$ band of methane (CH_4), and the $\nu_1 + \nu_3$ and $2\nu_2 + \nu_3$ bands of water (H_2O) with high resolution [3]. The outstanding capability of DCS has been confirmed; a broad spectrum can be measured in a relatively short time with high resolution and high precision in terms of frequency and intensity.

In the course of our study, by fitting each ro-vibrational line profile to Lambert-Beer's law with a Voigt function, we discovered an interesting phenomenon, namely that the pressure broadening coefficient depends on the nuclear-spin modification (ortho/para), and we published this finding as a letter [4]. The effects we observed are tiny but significant. In this paper we present the line strength of each ro-vibrational transition of this band, as well as the vibrational transition dipole moment of the band, which we determined for the first time using DCS and then compared them with those reported in the literature.

Numerous spectroscopic investigations have been published for acetylene. The study of the intensity of this acetylene band dates back to 1975; Varanasi and Bangaru [5] reported the line strength of each ro-vibrational transition determined from the spectrum measured by a grating spectrometer. Since then a number of papers have been published regarding the intensity of this band. The most reliable and comprehensive study was published in 2002; Hachtouki and Auwera [6] measured the band with high spectral resolution using Fourier-transform infrared spectroscopy (FTIR), and reported the intensity parameters. More recently in 2009, Li et al. [7] measured some selected transitions with high precision using continuous-wave laser spectroscopy (CW-LS). It is also noteworthy that CW-LS was employed in a highly intensive and precise study of an isotopologue, $^{13}C_2H_2$, by Kusaba and Henningsen in 2001 [8].

The present study with DCS essentially confirms the results of previous studies [5–7]. A small but systematic deviation of about 1.5% is found between the present and previously reported line strengths, which may be explained by the systematic uncertainties in the column density measurements in the present study.

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DCS essentially enabled us to measure the entire $\nu_1 + \nu_3$ band in a short time with high resolution and high frequency precision. Thanks to this outstanding capability of DCS, we were able to determine the reliable line strength for each ro-vibrational transition, as well as the transition dipole moment of this band. The present study paves the way for the practical application of DCS to the quantitative monitoring of trace gasses and for quantitatively measuring the rotational temperature of gases.

2. Experimental procedure

In the present study, the spectra were measured employing a homemade dual-comb spectrometer as shown schematically in Fig. 1(a). The dual-comb spectrometer was described in detail in previous papers [3,4]. The two optical frequency combs are generated by mode-locked erbium-doped fiber lasers. The beam of the signal comb (signal beam) passes through the sample cell, and that of the local comb (local beam) is used as a local oscillator to derive the spectroscopic signal carried by the signal beam by the heterodyne technique. The signal and local combs are operated with slightly different repetition rates of $f_{\mathrm{rep,S}}$ and $f_{\mathrm{rep,L}} = f_{\mathrm{rep,S}} - \Delta f_{\mathrm{rep}}$, respectively. After passing through the sample cell, the signal beam is merged with the local beam by a polarizing beam splitter (PBS). The merged beam is split into two by using a half-wave plate and another PBS for the balanced detection of the beat signal. After the second PBS, the transmitted and reflected beams generate opposite-phase beat signals according to the polarization [9]. These signals are balanced-detected to reduce the common-mode noise. The half-wave plate is used to adjust the power balance between the signal and local beams by rotating their polarizations. The beat signal of the two beams is repeatedly recorded at every time interval of $1/\Delta f_{\mathrm{rep}}$ (typically ~10 ms) to obtain a time-domain interferogram, which is then transformed into a transmission spectrum in the frequency domain using a fast Fourier transform (FFT) algorithm.

The modes of both the signal and local combs are phase-locked to a common CW laser emitting at 1.54 µm. By contrast, the carrier-envelope offset frequency is phase-locked to an RF synthesizer. These two phase locks reduce the relative linewidth of the two combs to less than 1 Hz. This makes it possible to decrease Δf_{rep} and thus increase the Nyquist-condition-limited spectral span enough to observe the entire $\nu_1 + \nu_3$ vibration band of $^{12}C_2H_2$ in one shot.

Dual-comb spectroscopy can be performed in two different ways as reviewed by Coddington et al. [10]: i.e. with the asymmetric and symmetric configurations shown in Fig. 1(b). With the asymmetric configuration, which is used in the present study, only the signal beam passes through the sample cell. With this approach the amplitude and phase spectra are measured simultaneously by using the local beam as a local oscillator for heterodyne detection. On the other hand, with the symmetric configuration, both beams pass through the sample cell, and only the power spectrum of the transmitted radiation can be obtained. The latter optical alignment is essentially the same as that commonly employed in FTIR spectroscopy.

In the present study, we recorded the amplitude spectrum by using an asymmetric configuration, with $f_{\mathrm{rep,S}}$ set at 48 MHz and Δf_{rep} set at 33 or 45 Hz. Interferograms were averaged over 10000 times using the coherent averaging technique [11].

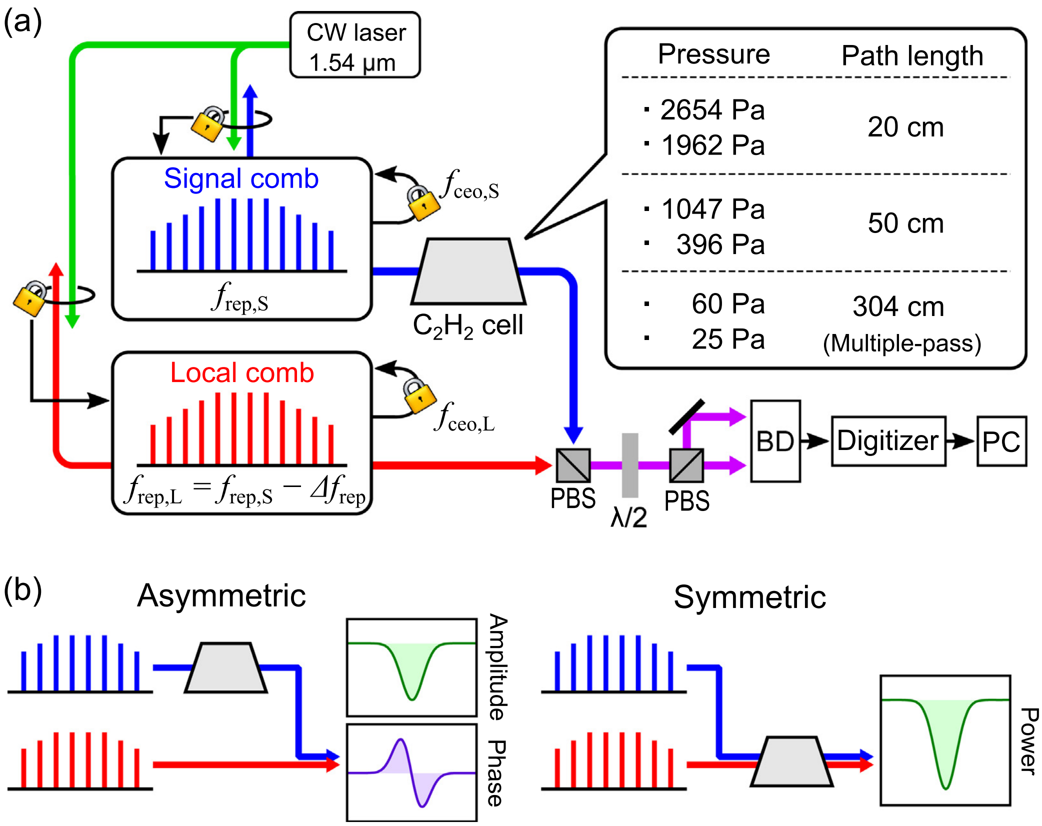

Fig. 1. (a) Experimental setup used in the present study. PBS: polarizing beam splitter, BD: balanced detector, $f_{\mathrm{ceo,S}}$ ($f_{\mathrm{ceo,L}}$): carrier-envelope offset frequency of the signal (local) comb, $\lambda/2$: half-wave plate. (b) Two configurations for dual-comb spectroscopy. In the asymmetric configuration where one comb is passed through the sample, we can obtain the amplitude and phase spectrum. In the symmetric configuration where both combs are passed through the sample, we obtain only the power spectrum.