



Measurement of absolute line intensities in the $\nu_5-\nu_4$ band of $^{12}\text{C}_2\text{H}_2$ using SOLEIL synchrotron far infrared AILES beamline

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ABSTRACT

Absolute intensities of about 120 lines of the $^{12}\text{C}_2\text{H}_2$ molecule are reported for the $\nu_4-\nu_5$ band between 65 and 192 cm^{-1} , with an average accuracy of 5%. Vibrational transition dipole moment squared values and empirical Herman–Wallis coefficients are obtained allowing modelling the rotational dependence of the transition dipole moment squared. Special care is taken to accurately determine an apparatus function for the Bruker IFS 125-HR coupled to the synchrotron SOLEIL far infrared AILES beamline in order to minimize its effects on the line parameter retrieval.

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1. Introduction

The infrared spectroscopy of the acetylene molecule C_2H_2 is important for atmospheric, planetary, and astrophysical applications (prebiotic molecule), as well as for industrial applications. This molecule is present as a trace constituent in the upper atmosphere of giant planets and satellites such as Titan where it results from methane

photodissociation and has been detected for example in spectra recorded with the infrared radiometer-infrared spectrometer (IRIS) instrument on board Voyager 1 and 2 [1]. Acetylene was also observed in the circumstellar shell of cool carbon stars such as IRC+10216 [2], and in interstellar clouds. A few years ago, it was possible to deduce the stratospheric distribution of acetylene in Uranus from spectra obtained with the Infrared Space Observatory instruments (ISO) [3]. Recent and future instruments as e.g. SPITZER [4,5], ALMA [6], or HERSCHEL [7] will operate in the FIR spectral region where only very few line intensities have been measured.

The present data have been obtained using the AILES-A (Advanced Infrared Line Exploited for Spectroscopy) beamline of SOLEIL synchrotron. This beamline located at the third generation Synchrotron Radiation source SOLEIL allows high resolution spectroscopic measurements of

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molecules in the entire infrared and THz range. The performances concerning flux, spectral range and stability can be particularly advantageous for high resolution FTIR spectrometer as the beamline optics permits the entire source to be used without aperture stop (entrance iris), even for measurements at highest resolution of $\sim 0.1 \mu\text{eV}$ (10^{-3}cm^{-1}). For this study, the spectral region ($50\text{--}600 \text{cm}^{-1}$) was exploited. This frequency range comprises two spectral regions of the acetylene absorption, corresponding to the $\Delta P=0$ and 1 sequence of vibrational transitions [8], P being a pseudo-quantum number equals to $5\nu_1+3\nu_2+5\nu_3+\nu_4+\nu_5$, where $\nu_1, \nu_2, \nu_3, \nu_4,$ and ν_5 are the quantum numbers associated with the normal modes of vibration of the molecule in the ground electronic state. Extensive work on this spectral region has been performed by Kabbadj et al. [9] who assigned vibro-rotational lines for 5 hot bands around 100cm^{-1} ($\Delta P=0$) and 5 hot bands around 500cm^{-1} ($\Delta P=1$). Among them, line intensities have been studied only for some lines of the $\nu_5^1-\nu_4^1$ intense band, by Robert et al. [10]. The superscript in the notation of this band refer to $\ell = |\ell_4 + \ell_5|$, ℓ_t being the vibrational angular momentum quantum number associated with the degenerated bending mode t . From the measurement of these line intensities, the vibrational transition dipole moment squared of the $\nu_5^1-\nu_4^1$ band has been deduced with an accuracy expected to be close to 20% in Ref. [10]. A synthetic spectrum of this band was then calculated and included in the Cologne Database for Molecular Spectroscopy (CDMS) [11] (273 lines from 1 to 242cm^{-1} , namely up to J equal 51 for various branches) with obvious astrophysical applications for the Herschel Space Observatory [12] but also for all instruments performing detection in this spectral range.

The goal of this work was to measure absolute line intensities at room temperature in the $50\text{--}600 \text{cm}^{-1}$ spectral region for all the bands observed in Ref. [9] in the two spectral regions of the acetylene absorption. To perform intensity measurements, several spectra have been recorded with a Fourier transform spectrometer Bruker IFS 125-HR. The SOLEIL synchrotron facility allows recording spectra with superior brilliance in the far infrared while providing an increased signal-to-noise ratio compared to classical sources. An important preliminary study on the apparatus function calculation and its effects on the line parameter retrieval was firstly performed. Taking into account the allowed beamtime, the experimental spectra have only been recorded with a single absorption path of around 10 m. Then, line intensities could be measured only in the $\nu_5^1-\nu_4^1$ band. For a complete and accurate analysis of the other bands, a larger absorption path will be required (around 100 m). In this work, a set of 120 absolute line intensities has been obtained with an average accuracy of 5%, and transition dipole moment squared values have been deduced. The next section of the paper will be devoted to the experimental conditions, to the analysis of the apparatus function and to its effects on the line parameter retrieval. The present results concerning line intensity measurements will be presented in Section 3, together with a comparison of transition dipole moments squared obtained in this work and those of Robert et al. [10].

2. Experimental conditions and preliminary studies

2.1. Experimental conditions

Six spectra have been recorded with the rapid scan Bruker IFS 125 HR interferometer coupled to the far infrared beamline AILES of the SOLEIL synchrotron. This beamline utilizes infrared synchrotron radiation from both edge emission [13] and constant field synchrotron source. The far infrared synchrotron emission is extracted and transported under vacuum in AILES beamline and is focused on the entrance aperture of the high resolution interferometer. The beam spot appears to be highly symmetric in the horizontal plane, while large asymmetry is observed along the vertical axe. This shape showing a peak-like distribution from the edge emission and the flat distribution of the constant field is in excellent agreement with the ray tracing calculations (SRW) at this wavelength [14]. This narrow profile explains how the AILES beam is well adapted for high resolution measurements. Indeed, with this distribution, no iris is needed even for the highest resolution measurements. For lower energy, the natural cone opens up as described in [15], and remains compatible with high resolution as the maximum iris size increases for decreasing energy. This advantage turns out to be particularly valuable when combining the high resolution interferometer with a multipass cell (White-type configuration).

For the present measurements, the Bruker IFS 125 is equipped with a $6 \mu\text{m}$ Mylar beam splitter, and a 4.2 K Si-bolometer detector. Experimental conditions are gathered in Table 1. An optical filter covering the spectral region between 50 and 600cm^{-1} was employed. The whole optical path is under vacuum and a metal multipass White-type cell (2.5 m base length) was aligned to provide a $10.14 \pm 0.01 \text{m}$ absorption path length. The cell was equipped with $50\text{-}\mu\text{m}$ -thick polypropylene windows. The temperature of the gas inside the cell has been obtained

Table 1
Experimental conditions and characteristics of the spectra recorded using the rapid-scan interferometer of SOLEIL-AILES beamline.

Commercial sample (Air Liquide Alphagaz)				
Natural C_2H_2				97.760% of $^{12}\text{C}_2\text{H}_2$
Stated purity				99.55%
Total absorption path				10.29 m
Collimator focal length				418 mm
Beam size at focal point				See text
Spectrum number	Total pressure (hPa)	Temperature (K)	Bruker-resolution ^a (cm^{-1})	Number co-added scans
1	0.598	300.1	0.002	300
2	1.204	300.0	0.002	266
3	2.450	300.2	0.002	299
4	4.886	300.0	0.003	336
5	9.798	299.6	0.003	292

^a Bruker-resolution = $0.9/\Delta_{\text{max}}$, Δ_{max} being the maximum optical path difference.

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