High-accuracy frequency atlas of $^{13}\text{C}_2\text{H}_2$ in the 1.5 μm region

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ABSTRACT A pair of 1.5 μm semiconductor laser frequency standards have been developed for optical telecommunications use, stabilised to Doppler-free transitions of the $\nu_1+\nu_3$ and $\nu_1+\nu_2+\nu_4+\nu_5$ combination bands of $^{13}\text{C}_2\text{H}_2$. The Allan deviation $\sigma/\tau$ for a laser locked to line P(10) of the former band follows a slope of $1.6 \times 10^{-12} \tau^{-1/2}$, reaching a minimum of $5.7 \times 10^{-14}$ at $\tau = 4000$ s. The absolute frequencies of 61 lines of the $\nu_1+\nu_3$ band and 43 lines of the $\nu_1+\nu_2+\nu_4+\nu_5$ band, covering the spectral region 1520 nm to 1552 nm, have been measured by use of a combined frequency chain and femtosecond comb, together with a passive optical frequency comb generator. The mean uncertainties for the line frequencies within each band are 1.4 kHz for the $\nu_1+\nu_3$ band and 1.9 kHz for the $\nu_1+\nu_2+\nu_4+\nu_5$ band, representing improvements on the precision of previously published data by factors of 100 and 10⁴, respectively. Improved values of the rotational constant $B''$ and centrifugal distortion coefficients $D''$, $H''$ and $L''$ of the vibrational ground state are presented.

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1 Introduction

The use of dense wavelength division multiplexing (DWDM) to increase data bandwidths in the 1.55 μm optical telecommunications band has driven the need for improved frequency standards in this region [1]. In recent years there has been considerable interest in frequency stabilising semiconductor lasers for this application using Doppler-free acetylene spectra [2–9]. Following the advent of Doppler-free acetylene systems [2], frequency measurements were carried out in Japan on the $\nu_1+\nu_3$ band, where either a passive optical frequency comb generator or a two colour pulsed fibre laser were used to span the gap between the acetylene lines and a 1.556 μm HCN standard or a 778 nm two-photon rubidium reference [3, 4, 6]. These measurements resulted in an uncertainty of 150 kHz in the absolute frequencies of over fifty transitions [3], and a reduced uncertainty of 12 kHz in the frequency value of the $\nu_1+\nu_3$ P(16) line in $^{13}\text{C}_2\text{H}_2$ [6]. Based on these measurements, the Comité International des Poids et Mesures (CIPM) adopted a value of 194 369 569.4 MHz with an uncertainty of 0.1 MHz or $5 \times 10^{-10}$ for the $\nu_1+\nu_3$ P(16) line in $^{13}\text{C}_2\text{H}_2$ [10]. Subsequent work at NMIJ/AIST (Japan) [7] and NPL (UK) [8], which made use of octave span femtosecond comb systems [11], led to a revision in the CIPM recommended value for line P(16) in September 2003 [1] to 194 369 569 385 kHz with a reduced uncertainty of 10 kHz ($5 \times 10^{-11}$). Measurements made at NRC (Canada) [9] have shown good agreement with this number and have also included precise measurements of five other $P$-branch lines. By contrast, the most accurate literature values for frequencies of the $\nu_1+\nu_2+\nu_4+\nu_5$ band have an uncertainty of 12 MHz [12]. In this paper we present the most accurate frequency values for the $\nu_1+\nu_3$ and $\nu_1+\nu_2+\nu_4+\nu_5$ bands of $^{13}\text{C}_2\text{H}_2$ yet reported, together with an improved determination of the molecular constants of the vibrational ground state.

2 Description of the acetylene systems

The two diode laser systems used in this work are commercially available extended cavity diode laser systems using the Littman configuration. The systems have a continuous tuning range of 1520–1570 nm, with an output power of ~15 mW and a free-running laser linewidth of ~2 MHz. Coarse frequency tuning is provided by a stepper motor on the extended cavity mirror, and fine tuning is available either via a piezo-electric transducer (PZT) on this mirror or through direct modulation of the laser diode injection current.

The experimental arrangement of an acetylene standard is shown in Fig. 1. The electro-optic phase modulator adds 8 MHz side-band to the laser beam and the modulation index $\beta$ can be varied in the range up to ~1. The light is mode-matched into the enhancement cavity (fineness $F \approx 100$, beam waist $w_0 \approx 0.48$ mm) with an input coupling efficiency of ~75%. The one-way power in the enhancement cavity can be varied up to 200 mW, giving a one-way axial power density of up to 0.56 W mm⁻². The cavity contains a sealed 200-mm Brewster-windowed cell, containing $^{13}\text{C}_2\text{H}_2$ at a pressure of 1.0 (2) Pa. The cell and cavity are enclosed in a pressure-sealed Al-alloy box, temperature-controlled at 25.0 ± 0.5°C.

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Two separate servo loops are used in the frequency stabilisation scheme. The first uses the Pound-Drever-Hall locking technique \cite{13}, stabilising the diode laser to the build-up cavity. This lock narrows the laser linewidth to $\sim 50$ kHz (1 s time scale). A 540 Hz frequency dither is applied via a PZT to one of the cavity mirrors, modulating the laser frequency to a depth of 1 MHz. Third harmonic phase-sensitive detection is then used to recover acetylene features from the transmitted light, and the cavity length is servoed to stabilise the laser frequency to an acetylene line. Extrapolating to zero modulation and power we obtain a Doppler-free acetylene linewidth of $\sim 0.6$ MHz. A scan profile of line P(10) of the $\nu_1 + \nu_3$ band obtained using phase-sensitive detection is shown in Fig. 2.

3 Frequency stability results

An acousto-optic modulator (AOM) is used to frequency-shift the output of NPL-ECL2 by $\sim 80$ MHz, allowing a beat frequency comparison of the two lasers locked to the same acetylene line. The standard operating conditions used in this work with a 1 Pa cell were $\beta = 0.7$, modulation depth 1 MHz and one-way axial power density $0.20 \text{ Wmm}^{-2}$. An Allan deviation plot for one of the acetylene systems locked to line P(10) of the $\nu_1 + \nu_3$ band is shown in Fig. 3. The short term relative stability follows a slope of $1.6 \times 10^{-12} \tau^{-1/2}$ over the range $1 \text{ s} < \tau < 1000 \text{ s}$. For longer averaging times, the Allan deviation reaches a minimum of $5.7 \times 10^{-14}$ at $\tau = 4000 \text{ s}$.

The sensitivity of the two standards to various systematic effects has been investigated. The observed power shift is $-1.5$ kHz/Wmm$^{-2}$, which is small in the context of the normal operating condition of $0.20 \text{ Wmm}^{-2}$. Another systematic effect is the modulation shift. The two NPL acetylene systems show different modulation shifts. For example, line P(10) of the $\nu_1 + \nu_3$ band NPL-ECL2 shows a modulation shift of $-0.2$ kHz/MHz whereas NPL-ECL1 displays a shift of $+1.3$ kHz/MHz. These compare to a figure of $+4.7$ kHz/MHz observed elsewhere \cite{9}. The frequency dependence on cell temperature has not been experimentally determined but is expected to be dominated by the pressure shift \cite{14}. For our systems, the frequency shift in changing from 3 Pa cells to 1 Pa cells is $+1.0$ (3) kHz for both bands. For a cell temperature variation of $\pm 0.5^\circ \text{C}$ at $25^\circ \text{C}$, one would thus expect the frequency variation to be $<1$ Hz peak-to-peak. Unlike measurements performed elsewhere using high pressure cells \cite{12, 14}, our results do not display any $J$-dependence of the pressure shift, neither does this shift differ between $P$-branch and $R$-branch lines. In addition to effects such as power shift, modulation shift and pressure shift, it is important to consider the influence of small electronic offsets in perturbing the lock point. To this end, the output offset of the phase-sensitive detector was varied, and the systems displayed a sensitivity of $+0.1$ kHz/mV. All of the above effects are readily controlled at a level of uncertainty corresponding to tens of Hz.

In addition to evaluating the relative stability of the two systems and their sensitivity to various systematic effects, their long-term reproducibility was observed with a series of beat frequency measurements over a period of several months.