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Measurement of oxygen B–band line center frequency in reference to strontium atomic optical clock

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Abstract. We present a proof–of–principle experiment demonstrating the use of atomic optical clocks as a frequency reference in Doppler–limited molecular spectroscopy. We report the determination of an unperturbed line position with a relative uncertainty of 2×10^{-11}.

1. Introduction

We report on a molecular line position determination in the Doppler–limited regime with respect to the frequency reference provided by a strontium atomic optical clock. We apply spectra averaging which significantly reduces the uncertainty of the determined line position. It is a step towards ultra–precise spectroscopy, capable of searching for variations of physical constants in molecular systems, verifying quantum electrodynamics in molecules as well as searching for fundamental effects in atomic interactions [1].

To demonstrate the technique we chose the P7P7 transition from molecular oxygen B–band around 690 nm. New data for this and other B–band lines will enable the improvement of the global fit of the O_2 spectrum model [2]. Its accuracy in the B–band range was mostly determined by uncertainties of line positions in [3] thus we push accuracy limits in this range.

2. Experimental apparatus and measurements

The spectra were collected with the Pound–Drever–Hall locked, frequency stabilised cavity ring–down spectrometer (PDH–locked FS–CRDS) linked to an Er:fibre optical frequency comb (OFC), which was frequency shifted to 1390 nm range and subsequently frequency doubled. For details of spectrometer's operation see [4] and references therein. For the purpose of this work modifications in previously described system considering frequency measurement were applied. Previously, a heterodyne beat–note signal between the probe beam and OFC was employed. At each point of spectrum, the OFC's repetition frequency $f_{\texttt{rep}}$ was changed so that beat–note's frequency remained in the required RF–filter's range during spectra acquisition.

In this work, we used the ^{88}Sr $^1S_0 - ^3P_0$ transition of the strontium atomic optical clock as a frequency reference [5, 6]. The spectrometer's probe laser frequency, which was varying from 434 779 to 434 788 GHz and the clock frequency, which is 429 228.066 418 012(5) GHz, were linked by the OFC. A schematic diagram of the experimental setup is shown in Fig. 1. Two heterodyne beat–note signals were measured: the first one between the clock laser and

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the OFC, and the second one between the spectrometer's probe beam and the OFC. In order to simultaneously measure both beat–note frequencies, $f_{\mathtt{rep}}$ remains unchanged during entire spectrum acquisition, whereas the probe beam's frequency is additionally shifted with a double–passed acousto–optic modulator so that the beat–note frequency remains in the measurement range. In such case the spectrometer's frequency f can be calculated as:

$$f = f_c + (n_p - n_c) f_{\mathtt{rep}} + f_{\mathtt{Bc}} - f_{\mathtt{Bp}} - f_s. \tag{1}$$

Here f_c is the clock laser frequency, n_p and n_c are comb tooth numbers for heterodyne beat–note with the spectrometer's laser and clock's laser beams, respectively, $f_{\mathtt{Bp}}$ and $f_{\mathtt{Bc}}$ are the frequencies of those beat–notes and f_s is sum of all additional frequency shifts in both beams. During the measurements all frequency counters and signal generators were referenced to the same, stable 10 MHz signal [7] from the hydrogen maser [8] transferred from Borowiec near Poznań to our Institute by a 330 km fibre link [6, 9].

With the system described above, we performed measurements of P7P7 self–perturbed transition in the oxygen B–band. Data was acquired at four sample pressures in the range from 1.5 to 6 Torr. At each pressure between 38 and 113 spectra were collected and averaged. Measurements at three pressures were performed within one week, whereas the fourth one was made four months later. First three datasets were referenced to the strontium optical clock, whereas the last one was referenced to the frequency standard from the hydrogen maser.

Spectra were fitted with the speed–dependent Galatry profile (SDGP) [10] with speed dependence described by hypergeometric functions [11]. To test the influence of line shape model on the line position determination, we have also performed fits with Voigt profile (VP). We found that due to the lack of line asymmetry, line position obtained from the VP agrees within fit uncertainty with value resulting from the SDGP fit. Averaged spectrum measured at the highest sample pressure, which was 6 Torr, as well as fit residuals, are shown in Fig. 2.

Referencing measurements to the frequency of the atomic optical clock enabled us to avoid systematic uncertainties caused by frequency standard, whereas spectra averaging led to significant reduction of statistical uncertainty. Other uncertainty sources are associated with
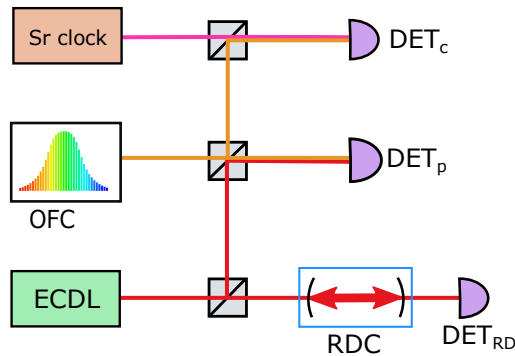
Figure 1. Schematic diagram of optical connections between main parts of the experimental setup. Sr clock is strontium atomic optical clock, ECDL – spectrometer probe laser, RDC – spectrometer's ring–down cavity, DET_c, DET_p and $\mathrm{DET}_{\mathrm{RD}}$ are detectors for heterodyne beat–notes with clock, probe beam and detector for acquisition or ring–down decays, respectively.
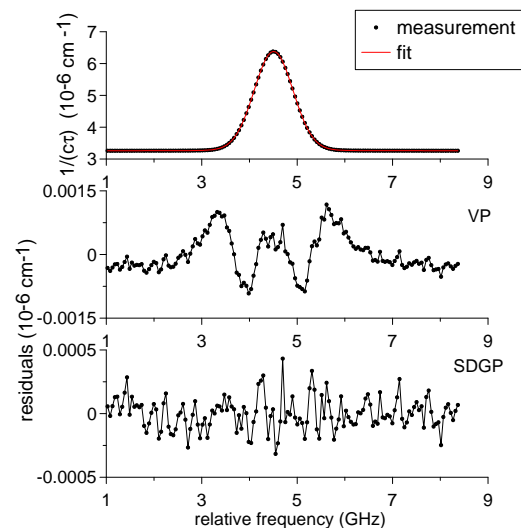
Figure 2. Top panel: Line P7P7 obtained at pressure of 6 Torr with averaging of 38 scans. Central and bottom panels: fit residuals obtained for VP and SDGP, respectively.

Table 1. Uncertainty budget for the determination of unperturbed line position ν_0 and collisional shift coefficient Δ/N. Systematic fit offset indicates differences in line positions obtained while performing fits pressure–by–pressure and multispectrum fit approach.

Uncertainty source	$u\,(\nu_0)$ (kHz)	$u\,(\Delta/N)$ (%)
Fit uncertainty	8.1	0.65
Systematic fit offset	2.1	0.12
Pressure reading & calibration (max 5.6 mTorr)	0.60	0.065
Temperature: variations, gradient, reading & calibration (max 240 mK)	0.86	0.094
Total	8.5	0.7

pressure and temperature measurement and calibration, as well as choice of spectra fitting technique. Their estimated influence on the uncertainty of unperturbed transition frequency and collisional shift coefficient is shown in Table 1. We have determined the unperturbed frequency of the P7P7 transition in the oxygen B-band equal to 434 783.508 485 7(85) GHz, which corresponds to relative uncertainty of 2×10^{-11}, and its collisional shift coefficient equal to $-0.09381(66) \times 10^{-19}$ GHz/(molecule/cm^3)) which corresponds to relative uncertainty of 7×10^{-3}.

3. Conclusions

We performed a proof–of–principle experiment showing application of atomic optical clocks as frequency reference in molecular spectroscopy. This experimental scheme can be applied for precise and accurate determination of hydrogen line positions and other of lines that cannot be easily accessed by sub–Doppler spectroscopy techniques and are important for fundamental studies.

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