

Ultra-narrow-linewidth continuous-wave THz sources based on multiplier chains

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Abstract We demonstrate two different sources at 1.3 THz based on multiplier chains (72nd harmonic generation), which exhibit linewidths at the level of 2×10^{-12} in relative units. The multiplication processes are shown not to contribute significantly to this linewidth. The phase noise of one of the sources and the fractional power in the carrier (76%) were determined. The application of these sources as references for quantum cascade THz lasers and for spectroscopy of ultracold molecules is suggested. Thus, rotational spectroscopy with few Hz resolution at 1.3 THz is possible with the present easy-to-use sources. An approach for reducing the linewidth by a factor on the order of 10^3 to the 1×10^{-15} level using optical technology is proposed.

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1 Introduction

Monochromatic radiation sources in the region around 1 THz are of significant interest in spectroscopy, where they can be used to probe rotational and vibrational levels of molecules [1]. At these frequencies, frequency-multiplied backward-wave oscillators (BWOs) have traditionally been used, delivering substantial power levels in the mW range.

They have been employed for numerous laboratory rotational and vibrational spectroscopy studies and have provided a wealth of information on molecular transition frequencies and on electric and magnetic dipole moments. For example, pure rotational transitions of light molecules and radicals, such as CH₂, NH₃, H₂O, and their isotopic species [2] have been accurately measured using THz sources. Low-energy bending vibrations are also accessible, e.g., the ν_2 transition of the C₃ molecule. One application of such laboratory measurements is the identification of molecular species in interstellar clouds [3]. Since astronomical receivers will soon routinely operate at THz frequencies, it is essential to have good laboratory data in order to interpret the astronomical observations.

Since BWOs are not frequency-stable, they must be frequency-stabilized to references. Synthesizer-controlled, frequency up-converted Gunn oscillators have been employed for this purpose, to which the BWOs are phase-locked. Using such sources, a high spectral resolution in rotational spectroscopy on room-temperature molecular ensembles is then achieved by using nonlinear spectroscopic techniques: Lamb dip spectroscopy has enabled sub-Doppler resolutions. Linewidths on the order of 10 kHz (10^{-7} – 10^{-8} in relative units) were measured for a number of diatomic and polyatomic molecular species, such as C₂H₂, ND₂H, and D₂OC [4], permitting to unravel fine details, such as nuclear hyperfine structure. However, BWOs are often complicated to use, need a large magnetic field and the beam properties and power change with frequency.

THz spectroscopy has also been performed using sources based on mixing two lasers [1, 5] in a nonlinear medium (photomixers [6, 9] and nonlinear-optical crystals [7, 8]), producing a wave at the difference frequency. Accurate frequency stabilization of the two lasers is necessary in order to obtain a frequency-stable THz wave. As alternative

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sources, and also to cover higher THz frequencies, quantum cascade laser sources are under intense development [8, 10, 11], toward compact, efficient sources with significant output power (at mW level). However, so far these sources require cryogenic operation.

A successful development line is efficient, room-temperature THz sources based on multiplier chains. These can be driven directly by microwave sources or by a fundamental oscillator, a Gunn oscillator. The Gunn oscillator itself is phase-locked to a quartz-stabilized multiplied microwave reference. Such sources can be wide-band and have important applications as local oscillators in THz astronomy [12, 13]. For example, in one reported source the range 1.1–1.7 THz was obtained with powers above 2 μW [14].

In this work we investigate the spectral properties of THz multiplier chain sources. We measure their linewidth and their phase noise via the microwave beat.

Our results indicate that the sources could provide a solution for two important future applications. The first is as a reference for the stabilization of THz quantum cascade lasers. These have relatively large linewidths of order 10 kHz [8, 10] and large frequency jitter (of order 1 MHz), due to environmental perturbations [8]. Moreover, an absolute frequency measurement of their radiation is desirable. Such THz lasers could be phase-locked to the multiplier chain output, thereby delivering narrow-linewidth radiation with mW power and precisely known frequency.

The second application of THz multiplier chains is as stand-alone sources for ultra-high resolution spectroscopy, especially in molecular spectroscopy. Recent developments in quantum optics have led to the ability to produce molecules, both neutral and charged, at extremely low temperatures (mK to μK). These can be trapped for a long time and confined spatially very tightly, in an ultra-high vacuum environment. A fundamentally new regime is thereby provided: the interaction of the molecules with the environment is minimal (very low collision rate), so neither transition time broadening nor pressure broadening limits the spectral resolution. The spatial localization can be much smaller than the wavelength of THz waves (0.2 mm at 1.5 THz). In this Lamb–Dicke regime Doppler broadening can be circumvented. Consequently, the transition linewidths are expected to be extremely narrow, ultimately limited by the interaction time of the molecules with the radiation or the natural lifetime of the molecular levels. An example of an interesting molecular system is the molecular hydrogen ion HD^+ , which has been sympathetically laser-cooled [15] and where a measurement of the $J' = 1 \leftarrow J'' = 0$ rotational transition frequency (at 1.31 THz) at the level of 1×10^{-9} or better would provide a unique opportunity for comparison with ab initio theoretical predictions [16]. To make use of the ultranarrow linewidth of rotational transitions (Q-factors exceeding 10^{12}), appropriate THz sources are required, with

linewidths on the order of 1 Hz or less. A first important step in this direction has recently been demonstrated: THz spectroscopy on cold molecules at 25 K [17].

For these applications, where Doppler broadening is very small or absent, a high power is not required: μW power levels are sufficient to saturate or coherently drive the transitions. Neither phase-locked BWOs nor current THz lasers are appropriate sources. THz radiation with Hz-level linewidth and accurately known frequency could in principle be produced by difference-frequency generation of a pair of continuous-wave lasers with Hz-level linewidths. However, this approach is complex, needing very precise stabilization of each laser to an ultra-low loss, vibrationally isolated optical cavity, and measurement of both optical frequencies with a femtosecond frequency comb.

The multiplier-based approach to THz generation is an appropriate solution; it has the benefits of power levels sufficient for spectroscopic applications, turn-key and computer-controlled operation, easy modulation, easily measurable absolute frequency, ultra-narrow linewidth, and the option of upgrading to higher spectral purity.

2 Description of the apparatus

Two independent sources, similar in concept, have been investigated. Both sources are driven by commercial microwave synthesizers. Source 1 (Radiometer-Physics) (Fig. 1a) is a multiplication chain driven by an Agilent synthesizer E8241A with 8 dBm at 18.2 GHz. In sequence, the chain consists of a doubler, an amplifier, a tripler, a power amplifier (at 109 GHz), a doubler, a doubler, and a final tripler. The total frequency multiplication factor is 72, leading to 1.31 THz. The second-to-last doubler is provided with a fixed bias, the last doubler with a variable bias. The source is narrowband with a -3 dB tuning range of only ± 20 GHz around 1.31 THz, and with a maximum output power of 2.5 μW .

Source 2 (Virginia Diodes, Fig. 1b) is driven by an 18 dBm input signal at frequencies from 17.4 to 19.4 GHz, which are frequency-doubled, amplified (31 dB), and then further converted by a quadrupler, a tripler, and a final tripler, yielding a total multiplication factor of 72 as well. This source covers the range 1.25 to 1.39 THz with power between 3 and 7 μW . In the present study it was driven by an Agilent synthesizer E8257D (with option UNR, enhanced phase noise performance) at 18.2 GHz in order to match the frequency of source 1.

The 18 GHz microwave input signals to the chains are derived from the respective internal 10 MHz quartz time bases. Their frequency jitter and phase noise spectrum are of interest since they lead to corresponding jitter and linewidth

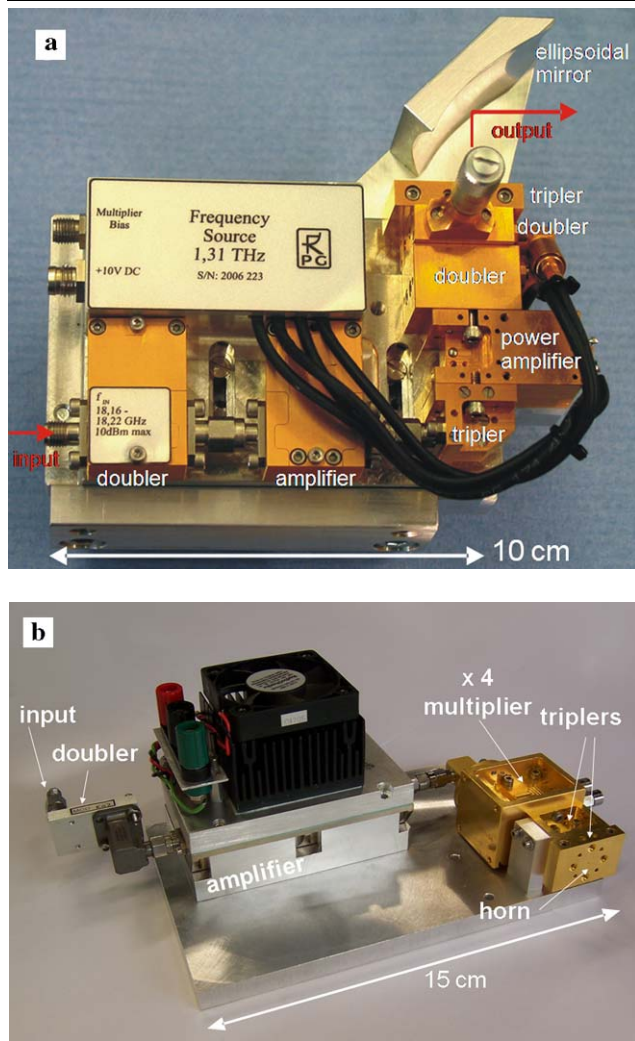


Fig. 1 **a** Narrow-band THz source 1; **b** wide-band THz source 2. Not shown are the bias and power supplies, and in case of source 2, the focusing mirror

at 18 GHz and finally at 1.3 THz. The typical single-sideband RF power spectral density $L(f)$ of the synthesizers of source 1 and 2 at 20 GHz and $f = 10$ Hz offset is -62 dBc/Hz and approx. -65 dBc/Hz, respectively, according to the manufacturer's data. In the range 100 to 600 Hz, the value is about -74 dBc/Hz for source 1 and between -80 and -94 dBc/Hz for source 2. Data for 1 Hz offset is not available for the models used but from extrapolation is expected to be approximately 25 dB higher than at 10 Hz. Both synthesizers' spectral noise at low offset frequencies (tens of Hz to tens of kHz) is specified as increasing with increasing absolute synthesizer frequency as for a pure multiplication process and thus is approximately 0.8 dB lower at the relevant 18.2 GHz compared to 20 GHz. A measurement of L_1 of synthesizer 1 at 18.2 GHz using a spectrum analyzer (Agilent E4440A) yielded values in the

range -78 ± 2 dBc/Hz for $f = 100$ –600 Hz, dropping to approximately -107 dBc/Hz at 6 kHz.

In both sources, the linearly polarized radiation is emitted from diagonal horns. As the beams are strongly divergent, they are focused by off-axis ellipsoidal mirrors located at a distance of approx. 3 cm and 7 cm from the horns' ends in sources 1 and 2, respectively.

Both sources are compact, with volumes of approximately 1 liter (excluding the power supplies, including fans) and are thus suitable for installation close to an experiment, minimizing the use of beam guiding elements and absorption by water vapor in the surrounding air.

3 Experiment

In order to characterize the spectral properties of a frequency-multiplied THz source, we have performed a beat note experiment between the two independent sources. The beat frequency measurement is performed using the CONDOR receiver [13] built for astronomical observations. As shown in Fig. 2, the waves emitted from the two sources are superposed on a highly sensitive, cryogenic hot electron bolometer (HEB) at ~ 4 K [18], cooled by a pulse-tube cooler. The HEB detector's IF signal frequency range is 1.1–1.8 GHz. Source 2 is used as the local oscillator, and its wave is collimated and focused onto the HEB detector. Source 1 is mounted so that its wave is polarized orthogonal to that of source 2. The two waves are superposed on a wire grid polarizer and then enter a polarization-rotating dual-beam (Martin–Puplett) interferometer. It produces an output wave consisting of the two THz waves with parallel polarization and therefore capable of interfering on the HEB detector and producing a beat. Since the beam of source 2 is matched to the horn of the HEB, but that of source 1 is not, source 1 deposits much lower power on the detector. The frequency difference between the two synthesizers is set such that it results in a frequency difference of the THz waves equal to 1.5 GHz, near the center of the detector frequency band. The beat signal from the HEB detector is amplified by a high electron mobility transistor (HEMT), extracted from the cryostat, further amplified, and then mixed down to a frequency around 50 kHz using a mixer (Mini-Circuits ZX05-30W) driven by a 1.5 GHz local oscillator (HP 8648B). This oscillator was locked to one of the synthesizers via the 10 MHz reference input. The low frequency allows observing the beat signal between the THz waves using a high-resolution FFT spectrum analyzer (Stanford Research Systems SR780).

4 Results and discussion

Figure 3 shows a number of spectra of the beat. At a measurement time of 2 s, the full width half maximum (FWHM)

Fig. 2 Schematic of the setup. The dashed connection was open during the characterization of the free-running behavior. LPF: low-pass filter; HEB: hot electron bolometer

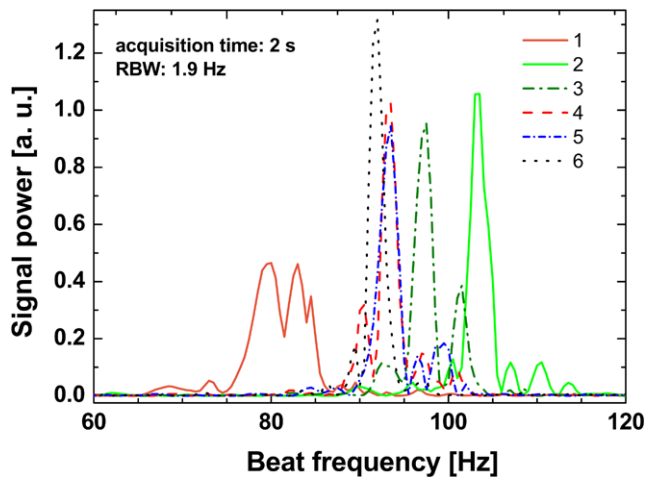
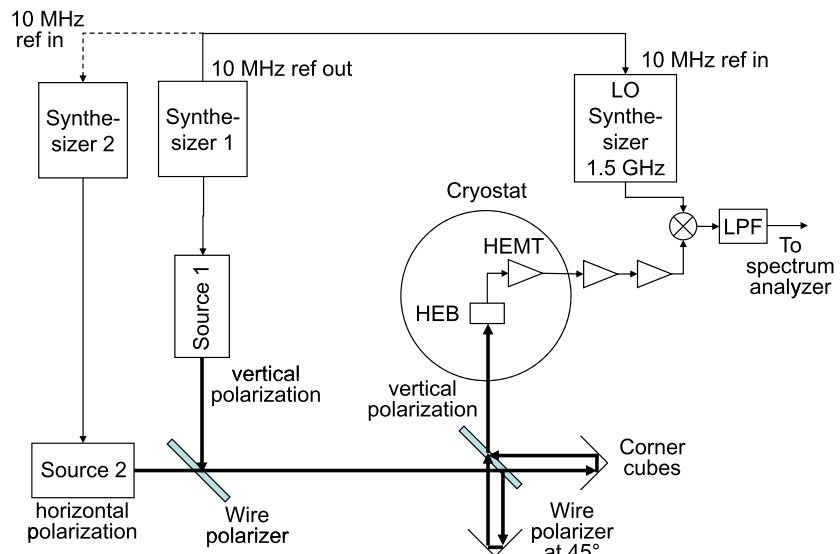


Fig. 3 Spectra (FFT magnitude squared) of the beat between the two free-running THz sources at 1.3 THz. The 6 scans were taken in the sequence as numbered, spaced by approximately 13 s. Acquisition time refers to a single scan. The frequency offset is arbitrary. RBW = spectrum analyzer resolution bandwidth

values vary between 2.5 Hz and 9 Hz. The resolution bandwidth of the synthesizer was approx. 2 Hz due to the FFT window used (flat-top). Figure 3 also shows that on the 13 s timescale between individual measurements, jitter on the order of 10 Hz occurs. This level is consistent with the type of quartz used by the two synthesizers, the quartz time base of the source 1 synthesizer being of lower stability and thus contributing dominantly. This jitter will lead to occasional broadening, e.g., on line #1 in Fig. 3. Disregarding jitter, we can state an upper limit of 2.5 Hz for the beat linewidth, or 2×10^{-12} in relative units, limited by the finite measurement time. This linewidth is the combined linewidth of the two THz waves. Note that the linewidth we consider here differs from the width of the peak of the power spectral density of

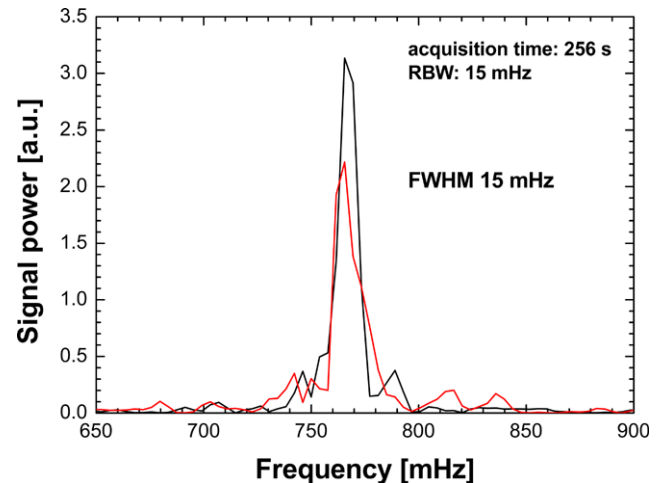


Fig. 4 Magnitude squared spectra of the beat between the two phase-locked THz sources at 1.3 THz. The two spectra were taken in sequence. The frequency scale has an offset

frequency fluctuations. We do not attempt to determine the latter in this work.

In order to test the influence of the low-frequency noise of the quartzes as compared to the multiplication chain, we locked one synthesizer to the other via the 10 MHz reference input and again took spectra of the beat note. The result of the phase-locked beat is displayed in Fig. 4. Jitter is now essentially absent, and a long acquisition time can be used. At 256 s integration time, the FWHM linewidth was 15 mHz, equal to the spectrum analyzer resolution (for the particular FFT window used). The latter was determined by analyzing a 20 kHz signal from a synthesizer. Thus, the contribution to the linewidth of the THz carrier stemming only from the frequency multiplication process (disregarding the phase noise of the synthesizer) is less than 1.2×10^{-14} in relative terms for a single synthesizer. This indicates that the

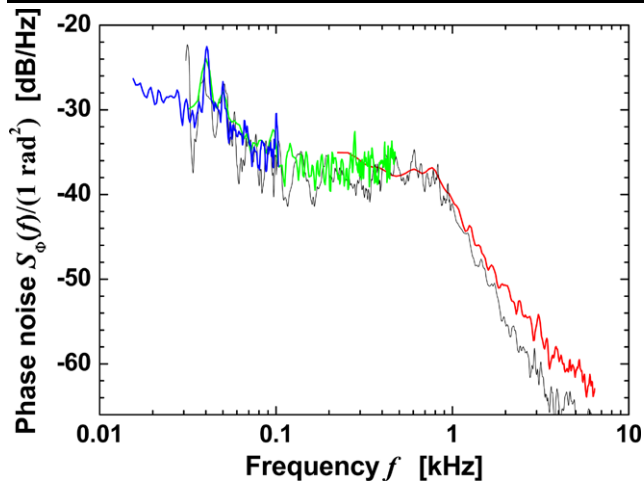


Fig. 5 Source 1 + 2 combined one-sided phase noise power spectral density at 1.3 THz, obtained from the beat between free-running sources. Shown are an average over 5 spectra measured with 200 Hz span and 1.9 Hz RBW (blue), an average over 9 spectra measured with 800 Hz span and 3.8 Hz RBW (green), and an average over 10 spectra measured with 12 800 Hz span and 121 Hz RBW (red). The thin black curve is the measured phase noise of synthesizer 1 at 18.2 GHz, increased by a factor 72^2 , and smoothed. In the frequency range above 0.1 kHz, the nominal phase noise of the spectrum analyzer used to measure it was subtracted. Below 100 Hz, the plotted values include the spectrum analyzer noise and are thus an upper limit for the synthesizer noise

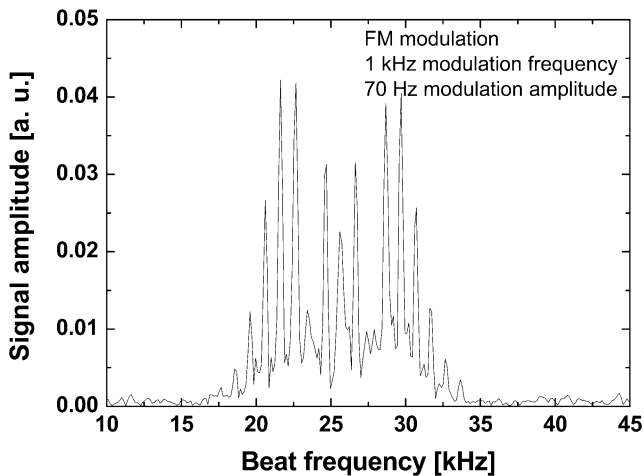


Fig. 6 Magnitude spectrum of the beat between the free-running THz sources, one of which is frequency-modulated

observed THz beat linewidth with *free-running* synthesizers is dominated by the (low-frequency) quartz noise. Since the frequency noise of the synthesizer quartz of source 2 is lower than that of source 1, we expect the frequency noise of source 1 to be the main contributor to the observed beat linewidth.

The phase noise power spectral density $S_{\phi}(f)$ of the free-running beat is shown on a larger frequency scale in Fig. 5. It has been derived from spectra of the beat, which is

possible in the case of weak phase noise and negligible amplitude modulation noise. In the present case of a beat, the spectral density is the sum of the individual spectral densities of the two sources. The phase noise of the 1.5 GHz LO synthesizer makes a negligible contribution. The spectral density exhibits a rather large noise pedestal. It can be characterized by the integral over the phase noise spectrum, which yields a root mean square (rms) phase deviation $\sigma_{\text{rms}} = 0.53$ rad in a bandwidth from $f = 16$ Hz to 6.4 kHz. The fractional power contained in the carrier is $\eta = \exp(-\sigma_{\text{rms}}^2) = 0.76$. The origin of the substantial noise contribution (0.24) is the high multiplication factor of the chain. As is well known, a frequency multiplication process by a factor M (here $M = 72$) reduces the fractional power η_0 in the carrier at the fundamental frequency to the level $\eta_0^{M^2}$ at the multiplied frequency. This corresponds, for weak phase modulation, to an increase in the phase noise power spectral density by a factor M^2 , or 20 dB per frequency multiplication decade. In our case, a phase noise power increase by $20 \log(M) \cong 37.1$ dB from 18.2 GHz to 1.3 THz is expected. For comparison, Fig. 5 also shows the phase noise of synthesizer 1 scaled by this factor, $2M^2L_1(f)$. The nominal noise of synthesizer 2 is significantly smaller. As can be seen, the scaled synthesizer 1 noise is comparable to the THz phase noise. Thus, the THz phase noise plateau in the frequency range 0.1–1 kHz originates from the multiplied-up plateau of synthesizer 1.

With the three employed synthesizers again all referenced to one 10 MHz reference, the actual value of the beat note frequency gives partial information about the synthesizers' internal synthesis algorithms accuracy. The THz beat note frequency, measured with the FFT spectrum analyzer, differed only by 0.16 Hz (1.2×10^{-13} in relative units) from the value calculated from the three synthesizers' frequency settings. This value corresponds to ~ 2 mHz when referred to the synthesizers' fundamental frequencies (18.2 GHz). A later test measurement consisting in mixing the three synthesizers' signals at approx. 1.5/17.2/18.7 GHz directly, and measuring the beat frequency did not show any deviation of its value from the expected one at a level of 1 mHz. It is likely that the 2 mHz discrepancy is due to a drift of the FFT analyzer quartz between the two measurements performed a few months apart.

A significant advantage of the multiplier chain approach is the simplicity of implementing frequency modulation, using the modulation options of the microwave source. As an example, the result of a frequency modulation of source 2 is shown in Fig. 6. One use of modulation is a spectral broadening of the THz radiation, which is useful for speeding up the initial search for molecular transitions if their precise frequencies are unknown, i.e. by making larger frequency steps. The exact frequency can then be determined by progressively reducing the modulation parameters.

5 Conclusion and perspectives

We have described THz sources based on commercial synthesizers that exhibit free-running THz radiation linewidth below 2×10^{-12} in relative units and reasonably high spectral purity, notwithstanding the large increase of phase noise by the large frequency multiplication factor. Such performance can in principle enable a 10^3 -fold improvement of the resolution in molecular ro-vibrational spectroscopy if combined with appropriate spectroscopic techniques. In order to make full use of this low linewidth, the jitter and drift of the quartz references on time scales above a few seconds must be reduced by stabilizing them, e.g., to a commercial atomic frequency standard. The modulation capabilities of appropriate synthesizers should also enable THz pulses with tailored shapes. This will allow coherent excitation (Rabi oscillations) of rotational or vibrational states in molecules.

It was shown that the multiplication chain by itself can permit a fractional linewidth of 1×10^{-14} . To achieve this in an actual THz source, rf or microwave oscillators with much lower phase noise and frequency jitter than the present synthesizers are required. For example, frequency-multiplied quartz oscillators with phase noise at the level of $(-62, -76)$ dBc/Hz for $f = (1, 10)$ Hz offsets and 10 GHz carrier, and with high frequency stability on time scales of seconds have been demonstrated [19]. A hydrogen maser is a particularly suitable source, providing both low phase noise (typical values corresponding to the above are $(-60, -72)$ dBc/Hz [20]), and in addition very high frequency stability (fractional instability below 1×10^{-14} for integration times of hours). Long-term stability is then obtainable by locking to GPS signals or to an atomic frequency standard. All of these sources are turn-key systems.

An approach leading to even higher spectral purity is possible using optical technology. Here, an optical reference is used, e.g., a laser locked to an ultrastable optical cavity. A femtosecond frequency comb is locked to this reference. The repetition rate of the comb, or a multiple of it, provides a microwave signal with excellent spectral purity [21]. At low offset frequencies (1–10 Hz), phase noise levels 40 dB below those of high-quality commercial sources such as those above have been demonstrated. Using the multiplier chain approach, such microwave performance could lead to THz radiation with mHz linewidths. A 1×10^{-15} relative frequency instability could then also be obtained, thanks to the high short-term stability of the optical cavities [22]. The frequency measurement of the THz wave could be easily performed on the microwave. While the optically generated ultra-low-noise microwave sources are still research systems, current progress in frequency comb and ultrastable laser technology could enable turnkey systems in the near future.

In conclusion, we have shown that current, commercially available and easy-to-use THz sources reach a fractional

linewidth of a few 10^{-12} . These sources are suited as frequency references for stronger THz radiation sources and for laboratory spectroscopy on molecules at milli-Kelvin temperature, allowing a resolution of, e.g., less than 3 Hz at 1300 GHz. In addition, we found that the multiplier chains increase the fractional linewidth of the THz radiation by less than 10^{-14} . Therefore another decrease in linewidth by a factor of several hundred should be obtainable by using as input signal a state-of-the-art microwave source, hence allowing THz applications fractional frequency resolution and accuracy in the 10^{-15} range.

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