Nonlinear Optics for Optical Frequency Synthesis and an Optical Divide by 3

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ABSTRACT

We review a number of new technologies show promise for the development of phase-coherent optical frequency measurement and synthesis. The ultimate goal is to have the capability to synthesize arbitrary optical frequencies; but on the shorter term it is important to measure the frequency of optical references (such as the Hg^+ transition at 282 nm and the calcium transition at 657 nm) relative to the primary cesium frequency standard. Recent progress in developing the base technologies for this effort include: improvements in our ability to measure arbitrary optical difference frequencies into the terahertz range, and using nonlinear optical techniques to extend the spectral range covered by diode lasers to the blue and the infrared. Using two nonlinear optical steps we have recently succeeded in dividing the frequency of a YAG laser (1.064 μm) by a factor of 3. This achievement should be an enabling step for a variety of optical synthesis schemes.

One of the most significant technical challenges to the viability, and general applicability of optical frequency standards is the need for a simple, robust, optical frequency measurement capability. Over the past 20 years a few optical frequency synthesis chains have been developed and used to measure a few key optical frequencies. These past and existing frequency chains have provided valuable measurements, but none of these systems fulfills the desired characteristics of being reliable, compact, and general purpose. Some new concepts have been proposed to simplify these systems, and in some cases these are being tested and developed. The relatively new concepts for optical frequency measurements include: the optical-cyclotron of Bergquist and Wineland;[1] the bisection scheme of Telle, Meschede, and Hänsch;[2] the optical frequency comb generator and OPO ideas of Wong;[3] and the 4-wave mixing frequency divider ideas of H.R. Telle.[4] All of these techniques advance the capabilities of optical frequency measurements but a number of problems remain to be solved.

An important capability for optical frequency synthesis is the ability to measure coherently the difference frequency between two laser sources in the visible. Four different technologies have been used: these include MIM diodes used with a laser local oscillator (detected difference frequencies to 2.5 THz),[5] harmonic mixing in fast

Schottky diodes,[6,7,8] optical comb generators,[9,10,11] and 4-wave-mixing in semiconductor lasers.[4] Up to now the Schottky diode systems have been limited to optical difference frequencies of about 1 THz. Optical comb generators and the 4-wave mixing systems have been improving and are now able to coherently measure difference frequencies of a few terahertz. To coherently span larger difference frequencies, it is still necessary to use nonlinear optical techniques such as optical sum and difference frequency or optical parametric oscillators (SFM, DFG, OPO).

Extending the Spectral Coverage of Semiconductor Diode Lasers

Most of the optical synthesis schemes require a number of tunable laser sources, which for practical reasons are likely to be semiconductor lasers. For this and other applications we have been exploring the high precision capabilities of commercial semiconductor lasers combined with nonlinear optical techniques and high frequency measurement methods. One of the requirements is to broaden the spectral coverage that is available from diode lasers. For example, SHG (second harmonic generation) and SFM can be used with commercial diode lasers to reach wavelengths in the blue and UV.[12] Obviously, some wavelengths are easier to reach than others. Favorable cases are those where high power diodes are available, and where noncritical phase matching can be used for the doubling and/or mixing. A well known example is SHG using $KNbO_3$ to generate wavelengths between 420 and 500 nm. In this case efficient doubling is routinely achieved and cw powers in the blue as high as 150 mW have been demonstrated. The available output power drops significantly for wavelengths less than 420 nm. However, new materials and techniques (such as quasi-phase matching in periodically poled materials) will likely change this in the very near future. A second stage of nonlinear mixing can extend the spectral coverage into the UV as has been recently demonstrated by Zimmerman et al.[13] and by Goldberg et al.[14] In these experiments diode lasers have been used to generate wavelengths as short as 212 nm (with microwatts of power) and 243 nm with 2 mW. Although only a few examples have been demonstrated, it seems possible to cover the entire spectral region between 200 and 500 nm using only diode lasers with SFM and SHG in nonlinear crystals.

Likewise, DFG can be used to extend the wavelength coverage of diode lasers toward longer wavelengths, into the infrared and even far-infrared regions. The chalcopyrite crystals, $AgGaS_2$ and $AgGaSe_2$, (and periodically poled $LiNbO_3$) are well suited to the task of converting near-IR diode laser power into tunable infrared.[15] Using only these crystals and diode lasers, it appears that it should be possible to cover the entire spectral range from 2 to 18 μm. Much of this capability remains to be proven, but a few good examples of this technique have been demonstrated. These include the generation and application of tunable IR in the 3 to 5 micron region.[15,16,17] In these initial diode-laser DFG experiments, the typical output power is a few microwatts but higher power is feasible.

A promising new technology is the GaAs photomixer developed at MIT Lincoln Laboratory.[18] These photomixers have been used to generate tunable radiation in the frequency range between 10 GHz and 3.7 THz, and show promise of being usable up to 5 or 6 THz. The devices are compatible with diode laser sources (in the 800 nm region) and have the potential to make compact, phase-lockable, tunable, mm-wave sources, with a variety of possible applications. The photomixers are fabricated on low-temperature-grown GaAs and radiate mm-waves that are generated from the beat note between two cw laser sources. At NIST, we have used two extended-cavity diode lasers with the photomixers to generate radiation between 10 GHz and 1.7 THz. In these initial experiments the beatnote generated output power from the photomixers is still uncertain, but appears to be about 100 nW (maximum at about 200 GHz) for a total diode laser power incident on the photomixer of about 30 mW. Power levels of about 1 μW have been achieved in other experiments with these devices. In terms of optical frequency synthesis systems, the photomixers may allow us to measure optical difference-frequencies to frequencies higher than the ~1 THz that has been possible with Schottky diode mixers.

By detecting the optically generated mm-wave radiation on a fast Schottky diode that is used as a harmonic mixer, the frequency of the mm-waves could be locked and thus provide a synthesized mm-wave source. In initial experiments along these lines we used a microwave harmonic mixer to detect the radiation from the photomixer and used the IF signal to lock the frequency of one diode laser (with a frequency offset) relative to the other laser. The beat-note signal appears to have sufficient signal-to-noise at the IF to allow phase-locking of the whole system. We have not yet determined the usable frequency range of this system, but at a minimum it shows promise of providing a synthesized sweep oscillator in the mm-wave frequency region.

Admitting some exaggeration of the potential, and simplification of the technical challenges, it now seems possible to cover a large part of the electromagnetic spectrum with coherent light generated from diode laser sources and nonlinear optical elements. These systems have already proven themselves to be useful at a number of wavelengths that span from the UV to the mm-wave spectral region.

Semiconductor Laser MOPAs

The rapid pace of the improvement of semiconductor lasers is accelerating their use in scientific applications. A significant recent advance in this technology has been the development and very importantly, the commercialization of higher power laser systems based on tapered semiconductor amplifiers.[19,20] High-power, single transverse mode, tapered-gain systems are available at wavelengths from 780 to 860 nm, near 980 nm, and very recently near 670 nm. These new devices can provide power up to 1 watt, with a single transverse mode, with a single frequency and tunability. An example of such a high-power tunable diode laser system is shown in Fig. 1. This laser is configured as master oscillator- power amplifier (MOPA) system, where the master oscillator is a standard grating-tuned extended-cavity diode laser (ECDL) and the tapered amplifier chip provides the gain to reach high output powers.

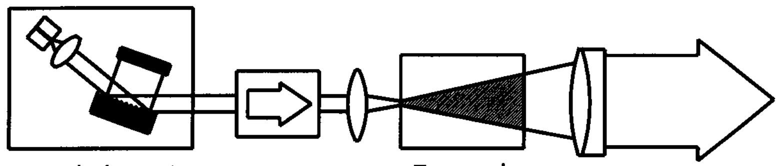

Fig. 1. Basic configuration of a tunable, single frequency, single spatial mode MOPA, which uses an extended-cavity diode laser as the master oscillator and a tapered-gain element as the power amplifier.

We have built two of these MOPA systems (near 800 nm and 675 nm) that provide similar though not identical performance characteristics. In our limited experience, the systems seem to require between 35 and 50 dB of isolation between the master and the power amplifier for stable operation. Adequate isolation also depends on other system parameters, particularly the backward power from the master laser and the facet reflectance and gain of the tapered amplifier. The MOPA system at 675 nm illustrates some of the characteristic parameters of these systems. With the master laser providing about 8 mW of usable output power, an optical isolation of ~35 dB, the output power from the tapered gain element can be as high as 500 mW. The system can be coarsely tuned from about 670 to 685 nm and has a fast-linewidth that matches that of the master laser (about 100 kHz). In this system the master laser is configured with a special design that allows broad, continuous, single-mode scans at a rapid rate (the best result of the master laser scanning is ~1.2 THz continuous single-frequency scan at a rate of ~100 s^{-1}). Using this laser as the master-oscillator in the MOPA provides high-power and broad continuous scans as demonstrated by the I_2 spectrum shown in Fig. 2.

For output powers less than about 150 mW, this red MOPA can be scanned continuously over frequency ranges that are larger than the mode spacing of the power-amplifier chip. With this particular system, at higher output powers (up to 500 mW) the continuous scan range of this MOPA is limited to about 20-30 GHz because of the residual facet reflectance of the tapered amplifier. Fortunately, even this red tapered amplifier has enough gain that it does not require much injected power from the master ECDL. The output power of the MOPA system begins to saturate with an input power from the master of about 3 mW.[21]

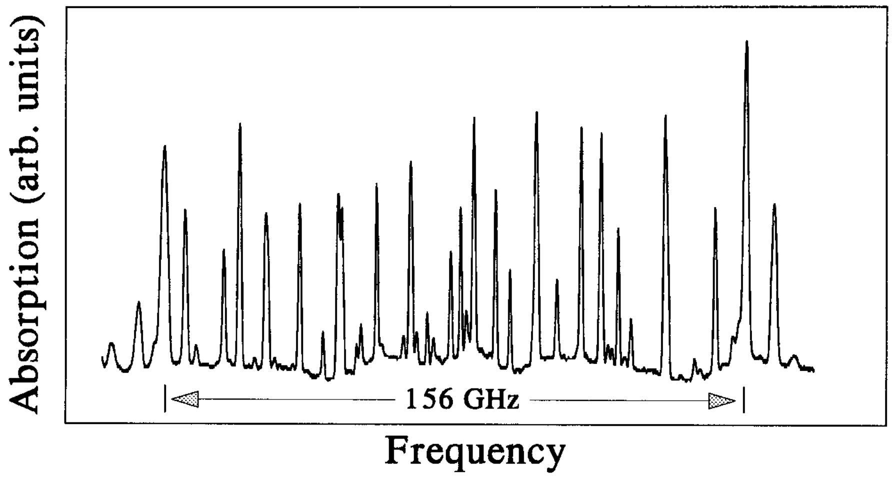

Fig. 2 Broad scan of the spectrum of I_2 at 674 nm taken with a MOPA system operating at an output power of ~150 mW. Entire 150 GHz scan was made in ~50 ms.

Semiconductor MOPAs now provide us with a powerful new tool for precision spectroscopy, optical sensing, laser cooling and trapping, and in particular for extending the spectral coverage achievable with diode lasers using nonlinear optics.

Optical Divide by 3

One of the difficulties of optical frequency synthesis is bridging the gap between the IR and the visible. Crossing this spectral region typically requires fundamental changes in the types of lasers and detector technologies. At NIST we are particularly interested in making coherent connections between optical frequency/wavelength references such as the 1.126 μm wavelength (1/4 of the Hg^+ optical "clock" frequency), the calcium line at 657 nm, the doubled YAG (1064 nm) stabilized on I_2[22], and ultimately the cesium frequency standard.

Nonlinear optical techniques are commonly used to shift optical frequencies by a factor of two (e.g. SHG and OPO). Achieving larger and arbitrary frequency ratios could simplify optical synthesis systems. In particular an optical divide by 3 system could play an important role as has been discussed by Wong.[3,23] DFG, SFM and OPO are not constrained to frequency ratios equal to a factor of two, but they do require additional measurements or mixing steps to ensure a specific frequency ratio. Unfortunately, optical phase matching over large frequency intervals can be difficult;

however there are cases where it is possible to achieve noncritical phase matching with optical frequencies that differ by a factor of three.

Part of the difficulty in implementing optical frequency synthesis chains is the lack of good quality tunable lasers in the 2 to 4 μm range. Though not very well known, there is the remarkable CO overtone ($\Delta v=2$) laser that operates on about 400 different lines between 2.6 and 4.0 μm, with output power that ranges from ~20 to 500 mW.[24] The CO laser is not continuously tunable but has a spacing between rotational lines of ~120 GHz. This density of lines provides a reasonable probability of finding a laser line near a desired frequency.

The specific divide by 3 system that we have developed uses the nonlinear crystal $RbTiOAsO_4$ (RTA) to take the difference frequency between a 3.2 μm CO laser and a 1.064 μm Nd:YAG, which generates radiation at 1.6 μm. The CO laser is simultaneously frequency doubled in $AgGaSe_2$ to also generate 1.6 μm. Then by controlling the beatnote between the two 1.6 μm signals we ensure that the frequency of the 3.2 μm laser is 1/3 of the frequency of the 1.064 μm laser. A diagram of the experimental setup is given in Fig. 3.

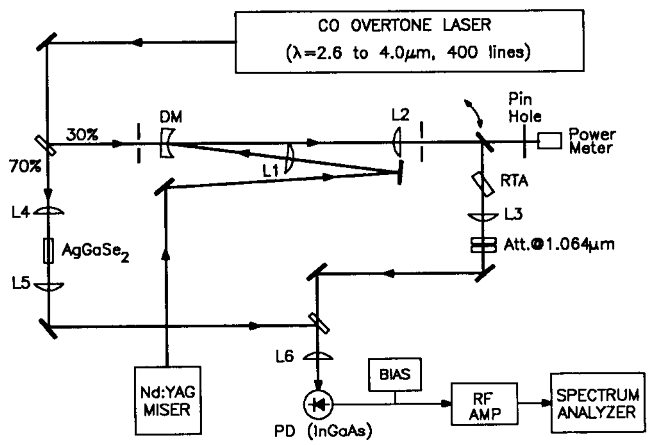

Fig. 3 Experimental setup for the optical divide by three of the frequency of a Nd:YAG laser at 1.064 μm. RTA is used to take the difference frequency and $AgGaSe_2$ is used for the SHG.

RTA is a biaxial crystal which is not commonly used and the Sellmeier coefficients are not known very well beyond 3 μm, but we have found RTA to be a good choice for a divide-by-3 system in the 1 to 3 μm region because it can be noncritically phase-matched for DFG. This scheme has some attractive features, it provides a convenient method to jump from tunable visible lasers to the existing IR gas lasers. In addition the two nonlinear crystals are both relatively efficient, which means that sufficient

SHG and DFG are generated in single pass to give a beatnote with good signal-to-noise ratio (see fig. 4). This beatnote was generated with 1 μW from the SHG beam and ~20 nW from the DFG beam and provides a signal-to-noise of ~40 dB in a 100 kHz bandwidth. This should be adequate for phase-locking the two lasers. The power available from the two lasers at these wavelengths were 500 mW from the diode-pumped YAG and 150 mW from the CO laser.

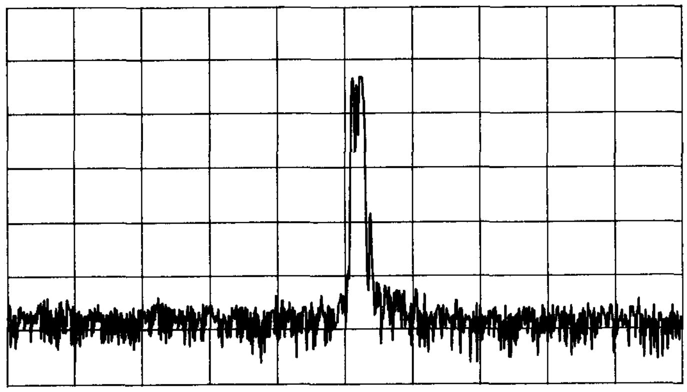

Fig. 4 Spectrum analyzer display of the beatnote at 1.6 μm between the SHG of the CO laser ($P_{21}(14)$ line at 3.192 μm, where 21 is the lower vibrational level) and the 1.6 μm radiation generated as the difference frequency between the CO laser and a 1.064 μm Nd:YAG laser. Horizontal axis is 500 kHz/Div and the vertical is 10 dB/div (resolution bandwidth 30 kHz, sweeptime 17 ms/div).

This type of divide-by-3 system could be used to generate a coarse array of reference lines across the visible and the near IR. Implementing it using 1.126 μm as the starting point (which is 1/4th of the narrow Hg^+ "clock" transition at 282 nm)[21] would produce adjacent elements of the array spaced by 1/12 of the Hg^+ clock transition frequency. The various lines could be generated by SFM and DFM. In the particular case of the Hg^+ transition these wavelengths would be approximately 3378, 1689, 1126, 844, 676, and 563 nm. In part, to test the feasibility of these schemes we have used DFG mixing to generate wavelengths nearby those listed above. For example: $AgGaS_2$ was used to take diode($\approx$800 nm) - YAG(1064 nm) $\rightarrow$ 3.2 μm ; and $LiNbO_3$ was used to take diode(657 nm) - YAG(1064 nm) $\rightarrow$ 1.6 μm. A successful divide by 3 and generation of 1/12 intervals of the Hg^+ transition would give us the beginning reference grid for other optical frequency measurements. Further division of the frequency of the Hg^+ transition could proceed using a number of different schemes. An interesting case might be to use Wong's OPO divider scheme[3] to subdivide the interval between 1126 and 1689 nm. Also, it would be fairly straightforward to trisect or bisect the 1/12 intervals using DFM with CO_2 and/or CO laser lines. Using standard techniques we can now connect directly to other reference wavelengths in the IR (such as the CH_4/HeNe or the OsO_4/CO_2 lasers) that have previously been measured by other frequency chain systems.

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