Proposed sum-and-difference method for optical-frequency measurement in the near infrared

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Received March 4, 1994

We propose a method for the determination of optical frequencies in the near infrared that is based on the nonlinear generation of the optical sum and difference frequencies of two near-infrared lasers followed by the comparison of the sum and difference frequencies with standards in the visible and in the far infrared, respectively. We also address questions of practicability and discuss some examples open to the method.

The impressive stability exhibited by lasers locked to atomic and molecular standards has created a set of frequency fixed points, and some of these frequencies have been tied to the Cs-based definition of the second through the use of intricate frequency-transfer chains. Because of the complexity of such measurements, there is ample motivation both to exploit the standard frequencies already established by them and to find simpler frequency-measurement techniques applicable to general frequency targets. Here we highlight a method especially suitable for measuring optical frequencies in the near infrared. The basic idea is that, given two lasers of stable but unknown frequency, knowledge of the sum and the difference of their frequencies suffices to determine both unknown frequencies. For laser frequencies in the near infrared it is often possible to arrange for both the sum frequency to lie sufficiently near a standard in the visible and the difference frequency to lie sufficiently near a standard in the far infrared for such a measurement to be performed. Furthermore, the required nonlinear processes of sum- and difference-frequency generation can be accomplished with adequate efficiency. We conclude with examples of possible frequency measurements near laser wavelengths of 1.338, 1.153, and 1.126 μm.

The method proposed here is an extension of the bisection method proposed by Telle *et al.*[1] and demonstrated by Wynands *et al.*[2] and obeys the same algebra as the optical parametric oscillator technique proposed and demonstrated by Wong[3] and by Lee and Wong,[4] respectively. A special case of the method has already been demonstrated by Kourogi *et al.*[5] We start by assuming that ν_1 and ν_2 are the frequencies of two stable lasers in the near infrared; of these, we treat one as the target frequency to be measured and the other as a free variable. If milliwatt beams from these two lasers are available, then the use of two separate nonlinear processes will permit the simultaneous creation of nanowatt beams of the optical sum ($\nu_s = \nu_1 + \nu_2$) and difference ($\nu_d = \nu_1 - \nu_2$) frequencies. The idea is to choose the free-variable near-infrared frequency such that both the sum and the difference frequencies fall at (or sufficiently near) standard optical frequencies previously measured with respect to the Cs-based standard. In general, the variation of only one free parameter would not allow two such matching conditions to be met, but here we take advantage of three levels of frequency flexibility. On the first level there is a growing list of well-known frequencies in the visible,[6] with frequency spacings of the order of 20 THz, which can be compared with the sum frequency. On the second level there is a large list of lines of CO_2 lasers[7] in the 25–33-THz range, with typical frequency spacing of the order of 20 GHz, which can be compared with the difference frequency. On the third level there is the ability to measure a heterodyne beat note of more than 400 GHz in the visible,[8] and more than 30 GHz near CO_2 wavelengths,[9] between a nonlinearly derived sum or difference frequency and a visible or CO_2-based standard. The result is that significant fractions of the near infrared are accessible to the proposed technique.

The easiest way to visualize the possibilities open to the method is to draw the two-dimensional $\nu_1\nu_2$ plane, in which (through the convention $\nu_2 < \nu_1$) the arena of interest is the lower 45° wedge of the first quadrant. In the plane, lines of slope -1 represent the loci of constant sum frequency; lines of slope $+1$, the loci of constant difference frequency. Figure 1 shows such a plane, with some of the useful standards in the visible shown as candidate sum frequencies and with the CO_2-laser lines shown as a band of candidate difference frequencies. Each intersection of a visible line with the CO_2 band defines projections onto the ν_1 and the ν_2 axes, giving two regions (each approximately 4 THz wide) accessible to the method; each new visible standard developed will add another two such regions. The present state of visible standards, with spacings greater than 20 THz, combined with a range of only approximately 8 THz of available CO_2 lines, implies that certain frequencies ν_1 or ν_2 are not yet measurable by the method.

Apart from the need for visible and far-infrared standards for comparison, this method requires ways to generate adequate power levels at the sum and the difference frequencies to be heterodyned against those standards. The nonlinear processes used will

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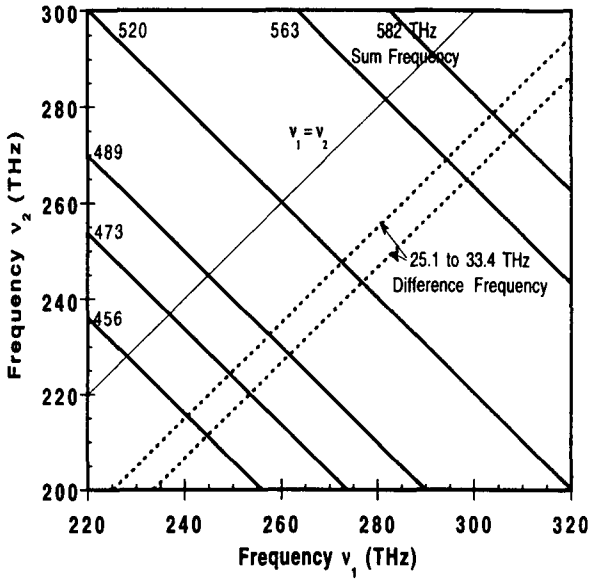

Fig. 1. A portion of the $\nu_1\nu_2$-frequency sum-and-difference plane, which shows loci of constant sum frequency for some interesting visible-laser frequencies, and the band of constant difference frequencies available from CO_2 lasers.

thus need to be phase matched, either by angle or by temperature tuning. For generating the sum frequency one solution is temperature-tuned noncritically phase-matched sum-frequency generation in lithium niobate ($LiNbO_3$). The dispersion curves for the pure material[10] give phase-matching loci (for the $o + o \rightarrow e$ process, where o and e denote ordinary and extraordinary axes) shown in the $\nu_1\nu_2$ plane in Fig. 2; the temperatures required are feasible (and will differ somewhat if congruently melting, or MgO-doped, material is used). For generating the difference frequency the nonlinear material must be transparent beyond 10 μm; one attractive possibility is silver selenogallate[11] ($AgGaSe_2$). Its dispersion curves[12] give the room-temperature phase-matching locus for noncritically phase-matched difference-frequency generation (for the $e - o \rightarrow o$ process) shown in Fig. 2. The extent to which $AgGaSe_2$ can be temperature tuned is not yet known; angle tuning of the material ($\theta < 90°$) makes accessible the region above and to the left of the locus shown. For regions inaccessible to $AgGaSe_2$ a useful material (although of smaller nonlinear coefficient) is proustite[13] (Ag_3AsS_3), which at room temperature will give angle-tuned $e - o \rightarrow o$ difference-frequency generation in the required region for angles $\theta = 16$–$20°$.[14]

One more choice can be exercised in the measurement of the difference frequency $\nu_d = \nu_1 - \nu_2$. The most obvious method is to illuminate the nonlinear material with input beams at frequencies ν_1 and ν_2 and to extract the output beam at ν_d. This beam, and one from a CO_2 laser, can then illuminate a far-infrared photodetector to generate the beat signal. An alternative method is to use the nonlinear material as an upconverter, illuminating it with frequency ν_2 and a beam from the CO_2 laser, to produce an output $\nu_{up} = \nu_2 + \nu_{CO_2}$; this output can be combined with the beam of frequency ν_1 to form a beat note on a photodetector sensitive in the near infrared (where detectors of greater sensitivity and bandwidth are available). The phase-matching conditions and the polarization requirements on the nonlinear material are the same for the two alternatives.

One of the attractions of the proposed method is that the only use made of the sum and the difference frequencies is to compare them by direct heterodyning with reference lasers in the visible and the far infrared. Given the power available from visible and CO_2 standards serving as local oscillators, the generation of very modest powers at the sum and the difference frequencies will suffice to give adequate beat signals at the detectors. For example, sum-frequency generation[15] in a 5-cm crystal of $LiNbO_3$ with two 10-mW input beams will give an output power of the order of 10 nW; when combined on a photodetector with a visible-standard beam of power 100 μW, this output will generate a beat note with a signal-to-noise ratio[16] of approximately 48 dB in a bandwidth of 100 kHz. Similarly, even angle-tuned upconversion in a 0.5-cm-long sample of proustite[17] is efficient enough that a 10-mW beam at ν_2, combined with a CO_2 beam of 100 mW/mm^2, will give an upconverted output of the order of 10 nW; when combined on a photodetector with a 10-mW beam at frequency ν_1, this output suffices to give a signal-to-noise ratio of 52 dB in the same bandwidth. Such beat-note signal strengths are sufficient to permit tight phase locking of the lasers at frequencies ν_1 and ν_2 to the visible and CO_2 standards, if desired. Thus there is no need to create new transfer-standard oscillators at the sum and the difference frequencies.

The first application of this sum-and-difference method was demonstrated by Kourogi *et al.*,[5] who locked a solid-state laser at 1.06 μm and a diode laser

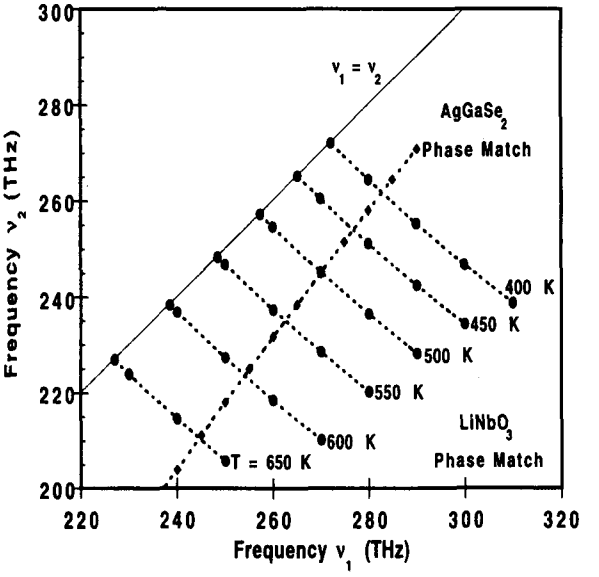

Fig. 2. Phase-matching loci in the $\nu_1\nu_2$-frequency plane. The family of curves gives the phase-matching loci for temperature-tuned type-I sum-frequency generation in pure $LiNbO_3$, calculated with the data of Ref. 10. The dashed curve gives the phase-matching locus for type-I difference-frequency generation in $AgGaSe_2$ at room temperature, calculated with the data of Ref. 12.

at 1.55 μm by beating their sum frequency against an I_2-stabilized 633-nm He–Ne laser and their difference frequency against a CH_4-stabilized 3.39-μm He–Ne laser. With ν_s and ν_d thus fixed there is no slack in the system, and it is indeed fortunate that the required ν_1 falls within the range of Nd-based solid-state lasers and that the resulting ν_2 is of such interest to the optical-fiber communication band.

As another possible application of the method, we mention the availability of diode-pumped monolithic Nd:YAG lasers[18] at wavelengths of 1.064, 1.319, and 1.338 μm; the first two of these have had their doubled frequencies stabilized to absorption lines in iodine cells,[19] and because of their compact size and their excellent passive stability all three are attractive candidates for absolute-frequency measurements. The three frequencies available are near 281.63, 227.26, and 224.06 THz, respectively. With the available CO_2 lines the last of these is directly accessible to the sum-and-difference technique: if we choose the 1.338-μm laser to give ν_2 = 224.06 THz and build a 1.201-μm laser (using available diode lasers or the Cr-doped forsterite[20] laser system) to provide ν_1 = 249.55 THz, then the sum frequency lies at the 473.61-THz 633-nm He–Ne standard, and the difference lies at 25.49-THz, near enough to the 25.475-THz $P(38)$ line of the $^{13}CO_2$ hot band.

Another example is a remeasurement of the 1.153-μm He–Ne laser frequency (whose second harmonic, stabilized by absorption in iodine, has already provided a 520-THz standard in the visible). The direct remeasurement of the frequency ν_1 = 260.103 THz at 1.153 μm is possible, given a commercially available diode-laser source at 1.305 μm to provide ν_2 = 229.77 THz, because the sum frequency of 489.880 THz matches the I_2-stabilized 612-nm He–Ne visible standard, and the difference frequency of 30.326 THz can be compared with the $^{13}CO_2$ $P(8)$ line at 30.315 THz. This remeasurement could provide a very tight consistency check on frequencies already measured by means of more complicated frequency chains.

A final example is a measurement of the 266.188-THz frequency of the 1.126-μm Nd:FAP laser, whose fourth harmonic matches the clock transition of the mercury ion-trap standard.[21] The use of a commercially available 1.446-μm, 207.394-THz diode-laser system would yield a sum frequency within 30 GHz of the 633-nm I_2-stabilized He–Ne standard and a difference frequency of 58.794 THz at the $P_5(13)$ line of the $^{12}C^{16}O$ laser.[22] This carbon monoxide frequency could in turn be stabilized with a metal–insulator–metal diode to double and to mix with the 29.407-THz $R(28)$ line of the $^{12}C^{16}O_2$ laser. The necessary sum- and difference-frequency generation can be phase matched by temperature tuning in $LiNbO_3$ and by angle tuning in $AgGaS_2$, respectively.

We thank U. Simon and J. L. Hall for stimulating discussions and NASA and the U.S. Air Force Office of Scientific Research for supporting this research.

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