Sub-Systems for Optical Frequency Measurements: Application to the 282-nm $^{199}Hg^{+}$ Transition and the 657-nm Ca Line

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***Abstract*– We are developing laser frequency measurement technologies that should allow us to construct an optical frequency synthesis system capable of measuring optical frequencies with a precision limited by the atomic frequency standards. The system will be used to interconnect and compare new advanced optical-frequency references (such as Ca, Hg^{+}, and others) and eventually to connect these references to the Cs primary frequency standard. The approach we are taking is to subdivide optical frequency intervals into smaller and smaller pieces until we are able to use standard electronic-frequency-measurement technology to measure the smallest interval.**

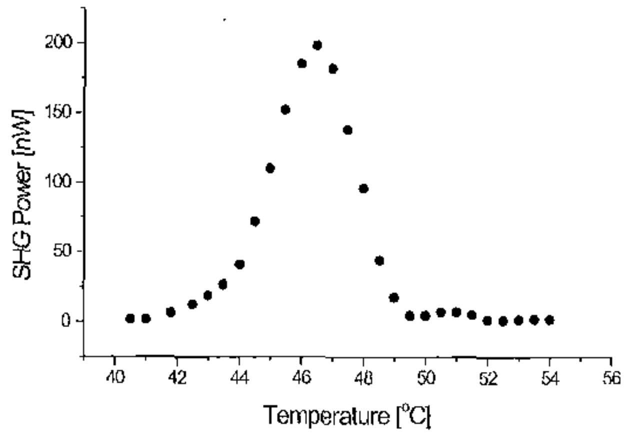

Fig. 1. Second harmonic power vs crystal temperature using 5 μW of 1315-nm diode laser radiation to produce 200 nW of 657-nm light in a single pass through a 20-mm long PPLN crystal. This is adequate power to phase lock the 1315-nm laser to the 657-nm reference laser.

I. Introduction

A. Domain-Engineered Lithium-Niobate for Optical Synthesis

The ideal optical-frequency-measurement system would be constructed from self-similar frequency dividing units that use compact lasers and simple nonlinear optical-mixing elements. These should be easy to assemble and run reliably. With this goal in mind, we have constructed and evaluated the performance of a number of diode-laser systems and optical-mixing stages. We have made progress in fabricating and using periodically poled lithium-niobate (PPLN) nonlinear crystals for most of the mixing stages in our optical synthesis system. With extended cavity diode lasers (ECDL) and a CO $\Delta v = 2$ laser, we have tested these crystals for practicality and efficiency in second harmonic generation (SHG), sum-frequency generation (SFG), difference-frequency generation (DFG), and even third harmonic generation (THG) [1]. By designing and fabricating the crystals for specific interactions we can achieve efficient nonlinear mixing (Fig. 1). Optical-grade lithium niobate (transparency window from 330 to 5500 nm) is readily available, and periodically poled crystals are now being produced commercially with various poling periods. Because not all of the poling periods that we needed were available, we developed our own poling capability at NIST. The poling is done by the electric field method, and we have now poled a number of samples with poling periods ranging from 8 to 36 μm.

Domain-engineered materials (of which PPLN is by far the most common) are a powerful and practical tool for the general task of optical-frequency synthesis [2]. We have used PPLN for a wide range of nonlinear mixing applications, some of which are summarized in Table I. For example, we have explored all possible combinations of mixing among three important wavelengths (563, 657, and 3925 nm) used in our present synthesis scheme. By pure luck, these mixings require the same poling period (12.6 μm) that we need for SHG of 1314 nm to reach 657 nm. Further evidence for the usefulness of this material as an optical mixer included weak blue and green

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TABLE I
CHARACTERISTICS OF THE DIFFERENT PPLN STAGES INVOLVED IN THE STUDIES.

PPLN stage	SHG#1	SHG#2	DFG#1	DFG#2	SHG Test	DFG Test	DFG Test	DFG Test	DFG Test
Period, μm	29.5	12.5	12.6	31.9	32.7	12.6	22.2	30.75	31.0
Temperature, °C	87	24	31	34	87	31	61	83	42
λ in, nm	3925	1308	563 3925	1308 3925	3378	657 563	810 1064	1064 2902	1064 2860
Power in, mW	100	5	15 50	4 80	200	4 20	50 250	300 65	300 48
λ out, nm	1962	654	657	1962	1689	3925	3393	1680	1694
Power out, nW	2500	200	4000	150	10 000	10	4000	2500	2500

beams that we observed coming out of a crystal designed for first-order DFG using 800 nm minus 1064 nm to produce 3.4 μm. These short wavelength beams were the two SHG signals and the SFG signal that were phase-matched at slightly different angles and high orders of the poling period [1]. In a special case, it was even possible to achieve THG of a cw laser at 3561 nm as a result of cascaded second-order nonlinearities that simultaneously phase-matched SHG and sum frequency mixing.

Domain-engineered materials such as PPLN might begin to play the role of a universal optical mixer. Unfortunately, optical nonlinearities are tiny in comparison with what can be achieved at lower frequencies with microwave harmonic mixer diodes, for example. Using low power cw lasers, we are basically limited to second-order optical nonlinearities that do not create high order mixing products efficiently. In addition, some technical challenges still limit the usefulness of PPLN in the short wavelength regions, for example SHG to the UV. Some of these cases require poling periods that are so short that they are difficult to fabricate with the present state of poling technology. As mentioned previously, going to higher order in the poling period (3, 5, 7 ...) is an alternative in some cases, but the efficiency drops rapidly as $(1/\text{order})^2$. Also, the optical power levels that can be generated in the visible/UV regions can be limited by photo-refractive effects. This, however, is not a serious problem for use in optical synthesis because we do not require high powers. Future use of waveguide mixing devices could be attractive, although waveguide technology is more challenging when the wavelengths are very different.

II. CONNECTION TO CO_2 LASERS

With a dependable connection between visible wavelengths using the CO $\Delta v = 2$ laser (~90 THz) and PPLN mixing crystals, we wanted to explore the possibility of stepping between diode laser wavelengths separated by a CO_2 laser frequency (~30 THz). Three CO_2 laser steps would then be able to span the CO $\Delta v = 2$ laser frequency interval. Thus, we set up an experiment to sum an ECDL at 844-nm wavelength with a CO_2 laser running on the $R_I(38)$ transition in the 10-μm band of the normal CO_2 isotopomer at a 10.134-μm wavelength.

The nonlinear crystal used for this experiment was Ag_3AsS_3 with a transparency window between 600 nm and 13 μm. Phase matching occurred at an angle of 22.6° with both input beams in ordinary polarization. Input powers of 250 mW at 10 μm and 1 mW at 844 nm led to an output power of 25 nW (efficiency: $\eta = 1 \times 10^{-4} W^{-1}$) at 780 nm with extraordinary polarization. A beatnote with a 25 dB signal-to-noise ratio (SNR) in a 10-kHz resolution bandwidth was sufficient to phase lock a second ECDL at 780 nm to the sum signal.

This experiment demonstrates the possibility of dividing the coarser steps of the CO $\Delta v = 2$ laser mixing stages phase coherently with a CO_2 laser. The CO_2 laser frequency could then be measured against Cs with a frequency chain [3], [4], but would not be required.

III. PRELIMINARY MEASUREMENT OF THE Hg^+ TRANSITION

Mixing of optical frequencies in PPLN opens the possibility for an accurate measurement of the Hg^+ clock transition at the 282-nm wavelength [5] using the Ca standard [6] as a frequency reference. The Ca frequency of the ${}^1S_0(m = 0) - {}^3P_1(m = 0)$ transition at $\nu(Ca) =$ 455 986 240.494 MHz has been measured by PTB against Cs with an uncertainty of ±135 Hz [7]. Even though the Hg+ standard holds great promise for a future optical frequency standard because of the very narrow optical clock transition ${}^2S_{1/2} - {}^2D_{5/2}(F = 0 \rightarrow F = 2, \Delta m_f = 0)$ with a natural linewidth of $\Delta\nu \approx 1.7$ Hz, its absolute frequency is relatively poorly known.

Our scheme to measure the Hg^+ transition is based on two fortuitous 'coincidences', namely one-half the Hg^+ frequency $\nu(\frac{1}{2}Hg^+) \approx \nu(Ca) + \nu(Ca)/6$ and $\nu(Ca)/6 \approx \nu(CO)$, where ν(CO) is the frequency of a CO $\Delta v = 2$ laser operated on the $P_{33}(15)$ transition. Hence, we can use the CO laser as a transfer oscillator between the two optical standards. Because the 282-nm transition in Hg^+ can be interrogated with a frequency-doubled dye laser oscillating at a 563-nm wavelength, the CO $\Delta v = 2$ laser can connect between the Ca reference and the dye laser frequency.

Using a PPLN crystal with a 12.6 μm poling period,

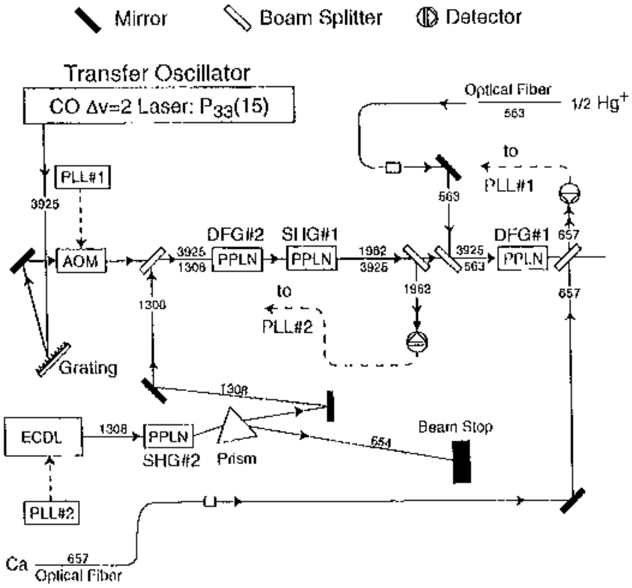

Fig. 2. Block diagram of system for frequency measurement of Hg^+ with respect to CA (AOM = acousto-optic modulator; PLL = phase-locked loop).

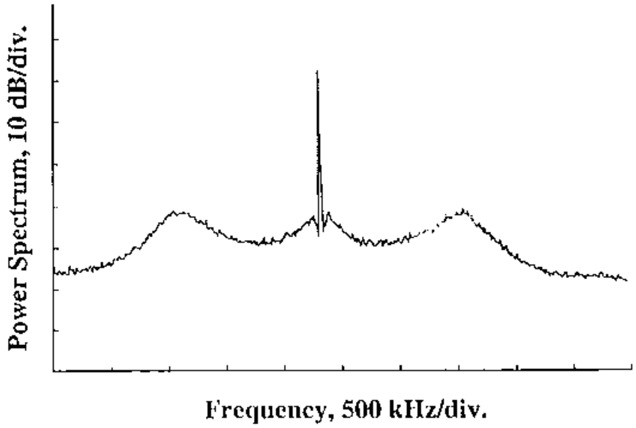

Fig. 3. Beat signal between the doubled CO laser (SHG#1) and the difference signal between the ECDL and the CO laser (DFG#2) used for phase-locking the ECDL (center frequency, 21.7 MHz; span, 1 MHz/div; Power, 10 dB/div, and resolution bandwidth, 30 kHz).

we generated a difference signal between the dye laser frequency ($\nu(\frac{1}{2}Hg^+)$) and the CO $\Delta v = 2$ laser frequency ν(CO). With input powers of 15 mW and 70 mW, respectively, we generated 5 μW in the difference signal. A beatnote between this signal and the ECDL from the Ca reference was achieved with a 3.6-GHz offset and a 20-dB SNR in a 100-kHz bandwidth. Careful alignment of the CO laser resonator and centering the CO $\Delta v = 2$ laser on its gain curve lead to a preliminary measurement of the Hg^+ transition frequency, $\nu(Hg^+) = 1\,064\,721\,600.4 \pm 20$ MHz. Knowledge of the CO $\Delta v = 2$ laser frequency was obtained by generating another beat signal with a CO $\Delta v = 1$ laser running on the $P_{33}(12)$ transition and a 20-GHz microwave source in seventh mixing order on a MIM diode. By centering both lasers carefully on their gain curves, the CO $\Delta v = 2$ laser frequency could be estimated from the beatnote frequency and the previously measured value of the CO $\Delta v = 1$ laser frequency [8] with an uncertainty of 10 MHz. This measurement of the Hg^+ optical clock transition frequency relative to Ca is a proof-of-principle demonstration of our measurement technique along our path toward a more precise measurement of the Hg^+ standard. To achieve an uncertainty of less than 1 kHz, we will also measure the CO $\Delta v = 2$ laser frequency relative to the Ca frequency (Fig. 2).

For this purpose, the CO laser is stabilized by phase locking it (with a frequency offset) to the frequency difference, $\nu(\frac{1}{2}Hg^+) - \nu(\text{Ca})$. The use of different nonlinear mixing stages in PPLN permits an effective multiplication of six times the CO laser frequency for comparison with ν(Ca). An ECDL with a frequency ν(DL) at a 1.3-μm wavelength is phase locked to $3 \times \nu$(CO) and at the same time frequency-doubled to $2 \times \nu$(DL), which is near ν(Ca). An optical frequency comb generator will provide the necessary connection between ν(Ca) and $6 \times \nu$(CO). A value for the CO laser transition frequency will then give a new accurate value for the frequency of the Hg^+ ion clock transition.

A difference signal (DFG#1) between ν(CO) and $\nu(\frac{1}{2}Hg^+)$ with a power of 3.5 μW is generated in PPLN, and a beat signal with a 3.6-GHz offset to ν(Ca) can be used for phase locking the CO laser. We have demonstrated phase locking of the CO laser by an acousto-optic modulator with a SNR of 40 dB in a 300-Hz bandwidth.

Another PPLN crystal allows doubling of the CO laser frequency ν(CO), delivering 2.5 μW of power in the second harmonic (SHG#1) at a 1962-nm wavelength. The SHG#1 radiation can be used to generate a beat signal with the difference frequency (DFG#2) between the ECDL and the CO laser at a 1962-nm wavelength that is generated in a third PPLN for a divide-by-three scheme of the ECDL. We achieved a 150-nW DFG#2 signal with input powers of 4 and 80 mW for the ECDL and the CO laser, respectively, leading to a beatnote of 55-dB SNR in a 30-kHz bandwidth between the SHG#1 and DFG#2. We have shown that this beat signal can be used for phase locking the ECDL to the third harmonic of the CO laser (Fig. 3).

A last PPLN crystal is used for doubling the ECDL (SHG#2) to $2 \times \nu$(DL), close to the Ca reference frequency ν(Ca). A power of 200 nW in the SHG#2 was achieved using 5 mW of input power of the ECDL.

All of the required nonlinear mixings, beatnotes, and phase locks have now been demonstrated. The last remaining step is to operate the whole system simultaneously and to measure the 2.24-THz frequency gap between the SHG#2 at 654 nm and the Ca reference at 657 nm. This interval will be bridged by an optical frequency comb generator [9], [10] that is currently being built by our group.

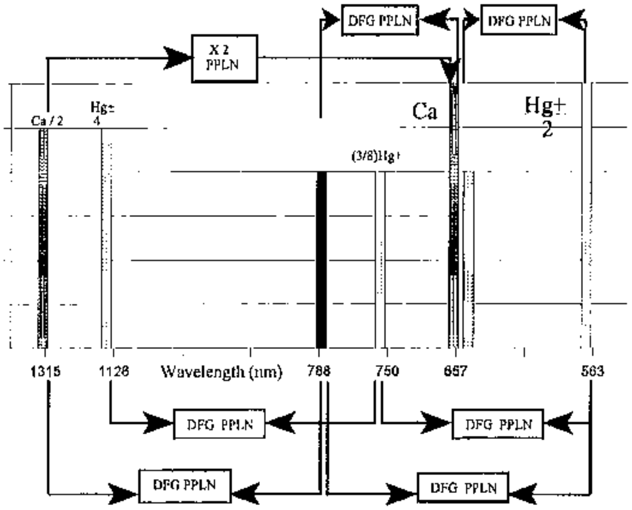

Fig. 4. Diagram of proposed optical frequency measurement system to connect the Hg+ and Ca references to the Cs clock. This relies on the subunits described previously, and we have added two new lasers: one at 788 nm that bisects the frequency interval between 563 and 1315 nm, and one at 750 nm that bisects the frequency interval between 563 and 1126 nm ($\frac{1}{2}$ the 563-nm laser frequency). The boxes labeled "DFG PPLN" imply difference frequency generation in periodically poled lithium niobate.

IV. Route from the Visible to the Cesium Clock

Our next major step is to measure the frequencies of Hg^+, Ca, and other important optical references relative to the primary Cs atomic frequency standard. The scheme we are developing for this purpose is diagramed subsequently.

Three separate but interconnected, frequency bisections are indicated in Fig. 4; these require making the difference-frequency between each end point and the mid point equal. Alternatively, these could be implemented by the SHG plus SFG bisection method described by Telle *et al.* [11]. Whichever approach is most appropriate can be chosen for the particular wavelengths involved. Until recently, we had planned to continue the frequency chain shown in Fig. 4 by using CO_2 lasers as described in Section II. However, our present plan is to take advantage of the revolutionary new results from Udem *et al.* [12] who have shown that it is possible to measure frequency intervals as large as 20 THz using femtosecond mode-locked lasers. Using their method, we plan to measure the interval between the 788- and 750-nm ($\sim$19 THz) lines shown in Fig. 4 with a pulsed Ti:sapphire laser and, additionally, to measure the interval between $\nu(657)_{Ca}$ and the $\nu(657)_{Bi}$, the bisector of the interval between 563 and 788 nm ($\approx$ 0.283 THz) with a comb generator. Simultaneous measurements of the 19.1-THz interval ($\Delta\nu_1$) and the 0.283-THz interval ($\Delta\nu_2$) yield the frequency of both Ca and $\frac{1}{2}Hg^+$ with respect to cesium: $\nu(\frac{1}{2}Hg^+) = 28 \times \Delta\nu_1 - 8 \times \Delta\nu_2$ and $\nu(Ca) = 24 \times \Delta\nu_1 - 8 \times \Delta\nu_2$. Much work remains to be done, however, with these new improved tools, such as diode lasers, PPLN, and mode-locked lasers, and, with some of the suggested schemes discussed, we believe that a path to precise optical frequency measurements is clear. We also feel that these new measurement systems are coming together just as the optical references are also reaching interesting performance levels. The hope is these optical standards will be able to compete favorably with microwave standards in terms of stability and accuracy.

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