Division by 3 of optical frequencies by use of difference-frequency generation in noncritically phase-matched $RbTiOAsO_4$

Olivier Pfister

JILA, National Institute of Standards and Technology and University of Colorado, Boulder, Colorado 80309-0440

Manfred Mürtz

Institute für Angewandte Physik der Universität Bonn, D-53115 Bonn, Germany

Joseph S. Wells and Leo Hollberg

National Institute of Standards and Technology, Boulder, Colorado 80303

James T. Murray

Optical Sciences Center, University of Arizona, Tucson, Arizona 85721-0001

Received March 7, 1996

A new scheme for coherently connecting optical frequencies in a 3:1 ratio has been demonstrated. To phase lock a Nd:YAG laser at 1064 nm with a CO overtone laser at 3192 nm, we generated their difference frequency in $RbTiOAsO_4$ (RTA) and beat it against the second harmonic of 3192 nm that was generated in $AgGaSe_2$.

The ability to measure optical frequencies is a critical technology for tests of QED, as demonstrated in the beautiful experiments of Hänsch and co-workers[1] and Biraben and co-workers.[2] A number of promising new optical frequency references have been proposed in recent years, including trapped ions[3,4] and cooled Ca atoms.[5] One expects higher precision and accuracy with standards at optical frequencies rather than the Cs primary frequency standard at 9.2 GHz. The ultimate linewidth of the Hg^+-based system, for example, is 1 Hz at 10^{15} Hz, with a projected stability of $10^{-15}\ \tau^{-1/2}$ (τ is the averaging time). A serious limitation to the feasibility of optical references is that it is difficult to measure optical frequencies and to transfer the stability to lower frequencies, where it is compatible with electronics. This is also what makes the connection of these potential new references to each other and to the microwave standards an interesting if formidable task.

A few modern ideas for optical frequency synthesis are now being developed, including the optical cyclotron,[6] the bisection method of Telle *et al.*,[7] four-wave mixing techniques,[8] and optical comb generators.[9] These ideas often feature an attractive way to address the problem, which is the use of nonlinear mixing crystals together with solid-state and diode lasers. Divide-by-3 schemes are of particular interest for many reasons. First, as Wong discussed,[10] building a frequency chain that could go from the Cs clock up to visible frequencies might require only a few lasers if 2:1 and 3:1 optical parametric oscillation (OPO) frequency dividers were combined. Also, a 3:1 frequency connection is a convenient way to jump from tunable visible lasers (diodes, Ti:sapphire) to the gas lasers that are established IR frequency standards (He–Ne laser locked on CH_4 at 3.39 μm, CO_2 laser locked on OsO_4 at 10 μm). Finally, we could combine a 3:1 divider with the twice-frequency-doubled Nd:FAP laser (1126 nm), which will be used at the National Institute of Standards and Technology to excite the 282-nm Hg^+ transition.[11] Such a combination would produce a wavelength of 3378 nm, which is close to the methane reference. If this divide-by-3 system were implemented with the 1126-nm wavelength (one fourth of the Hg^+ "clock" transition), we could readily generate an array of reference lines across the visible and the near IR. Adjacent elements of this array would be spaced by one twelfth of the Hg^+ transition frequency. In this case the wavelengths would be approximately 3378, 1689, 1126, 844, 676, 563, 483, 422, 375, 338, 307, and 282 nm. For the present study, with a Nd:YAG laser instead of a Nd:FAP laser, the wavelengths would be 3192, 1596, 1064, 798, 638, 532, 456, 399, 355, 319, 290, and 266 nm. In part to test the feasibility of these schemes, we have already used difference frequency generation (DFG) to generate wavelengths close to some of those listed above: For example, a diode laser at 657 or 800 nm and a Nd:YAG laser at 1064 nm have been mixed to produce 1700 or 3200 nm,[12] respectively. Recent high-precision measurements[13] of an I_2 electronic transition near 532 nm do make the Nd:YAG laser an interesting starting point.

A 3:1 connection (through second-order frequency mixing) is achievable with two input waves, such as sum-frequency generation ($f + 2f \mapsto 3f$) or DFG ($3f - f \mapsto 2f, 3f - 2f \mapsto f$), or with a single input wave, that is, OPO ($3f \mapsto f + 2f$). An exact connection requires another mixing stage to lock the 3:1 ratio (the phase-matching requirements of the crystal are not precise enough). We need two different mixings of $(f, 2f, 3f)$ to guarantee a division by 3.

For example, we can use OPO ($3f \mapsto f + 2f$) and second-harmonic generation (SHG, $f \mapsto 2f$), or we can use DFG ($3f - f \mapsto 2f$) and SHG ($f \mapsto 2f$), to obtain a beat note at $2f$, which can then be used to phase lock the OPO resonator to the $3f$ pump or to lock the two DFG input lasers together, respectively. The OPO beat note is actually much less sensitive to pump frequency noise than is the DFG beat note, for which the fluctuations of two independent lasers do not cancel each other. However, because of the additional problems related to OPO threshold, and because two good laser sources are available, we chose to build a DFG–SHG setup first. The frequencies involved are

$$f(1064\ \text{nm}) - f(3192\ \text{nm}) \mapsto f(1596\ \text{nm}). \qquad (1)$$

The input wavelengths were provided by a diode-pumped Nd:YAG monolithic ring laser (output power 530 mW) and by a CO overtone laser.[14] CO overtone lasers are not common but are nonetheless remarkable mid-IR sources. They were first developed in W. Urban's group in Bonn, Germany. They provide laser oscillation on 400 rotational lines, spread over 28 vibrational $\Delta v = 2$ transitions between 2.6 and 4.1 μm, with an output power as high as 200 mW in our case. Fortuitously, three times the frequency of the 3193.5-nm $P_{21}(14)$ line is only 4 GHz away from the center of the Nd:YAG gain curve. This falls well within the 30-GHz temperature-tuning range of the Nd:YAG monolithic oscillator.

Of the many crystals that permit phase matching for interaction (1) with reasonable nonlinear coefficients and low absorption ($LiNbO_3$, $KNbO_3$, $AgGaS_2$, proustite), none gives noncritical phase matching at a convenient temperature, which is a prerequisite if one hopes eventually to build a cw OPO system. Alkali metal–titanyl arsenate crystals (KTA, RTA, CTA) are of prime interest here: While they retain all the properties of their better-known parent KTP (reasonable nonlinearity, reduced walk-off in the $X-Y$ plane, high optical damage threshold, very high ferroelectric Curie temperature), they lack the phosphate-ion vibrational resonances that lead to significant absorption at 3.5 μm.[15] Also, RTA[16] gives noncritical phase matching at room temperature for[17]

$$f(1064\ \text{nm}) - f(3137\ \text{nm}) \mapsto f(1610\ \text{nm}). \qquad (2)$$

We observed that interaction (1) is phase matched in RTA at 335 °C. We predicted this temperature by measuring the phase-matching bandwidth at 3 μm by tuning the CO overtone laser through its $v = 23 \rightarrow 21$ emission band. A least-squares fit with a sinc^2 function gave us a 552 ± 25 GHz FWHM bandwidth, which, when divided by a temperature bandwidth of 100 °C (obtained from KTP's temperature dependence[18]), yields an approximate temperature-tuning coefficient of −5.52 GHz/ °C. Thus, to displace the phase-matching central wavelength from 3137 nm [interaction (2)] at room temperature to 3193.5 nm (−1691 GHz), one should increase the temperature by 306 °C. This is in fair agreement with the experimental determination and indicates that KTP's temperature dependence is a good first guess for arsenates as well. This large temperature acceptance gives sluggish temperature tuning, but, on the other hand, the sensitivity of the DFG interaction to temperature fluctuations is very low. We calculated the second-derivative FWHM angular bandwidth to be 8.9 °. Our attempt to measure it was not successful because of the inhomogeneity of our crystal.

The propagation in the biaxial crystal was along the X axis. Since the interaction is of type II (*oee*) in the $X-Y$ plane, the polarizations were along the Z axis for the 3192-nm ordinary beam and along the Y axis for the 1064- and 1596-nm extraordinary beams. The 7.25 mm × 5 mm × 5 mm RTA sample that we used was cut at 30° from the Z axis, so we had to use it sideways (normal axis at 30° from the X axis) with an ~65° incidence angle (Fig. 1). This alignment is close to Brewster incidence for the p-polarized light at 3192 nm, but it gives Fresnel losses of ~33% on the uncoated faces for the two other, s-polarized, beams. It also results in significant astigmatism, since the light was focused in the crystal. (Calculated waist sizes were 23 μm for the Nd:YAG beam and 40 μm for the CO beam.) The experimental setup is diagrammed in Fig. 2: The two pump beams were combined by means of a dichroic mirror (DM) and a 35-μm-diameter pinhole placed at the common waist position. A removable mirror mount of kinematic design allowed us to send the two beams into the RTA crystal, which was enclosed in a Au-plated Cu oven and heated to 330–340 °C. Two fused-silica mirrors, with transmittances of $\sim 10^{-4}$ at 1064 nm and 90% at 1.6 μm, were used between the crystal and the detector to block the powerful Nd:YAG radiation and also to absorb the less-intense 3.2 μm light. Part of the CO beam was simultaneously frequency doubled in a $AgGaSe_2$ crystal. This produced a second 1.6-μm beam, which was overlapped on a fast photodetector with the DFG signal. The ($R \sim 1/3$) beam splitter on the CO laser beam path was chosen to maximize the power on the beat detector:

$$P_{\text{beat}} \propto [(TP_1)^2 R P_1 P_3]^{1/2} \propto R^{1/2}(1 - \text{R}). \qquad (3)$$

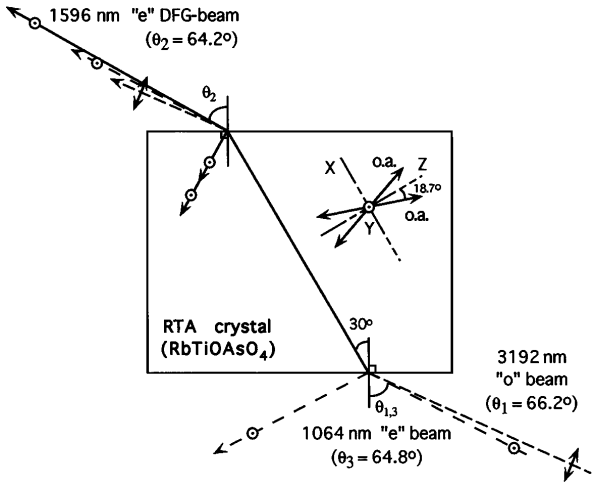

Fig. 1. RTA crystal: beam alignments and polarizations, optical axes (o.a.), and principal axes (X, Y, Z).

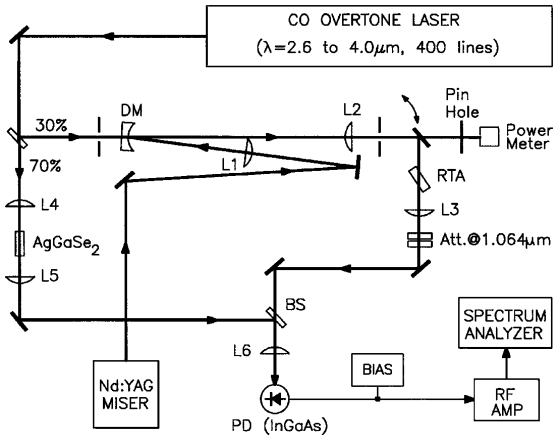

Fig. 2. Experimental setup: L1–L6, lenses; BS, beam splitter; PD, photodiode.

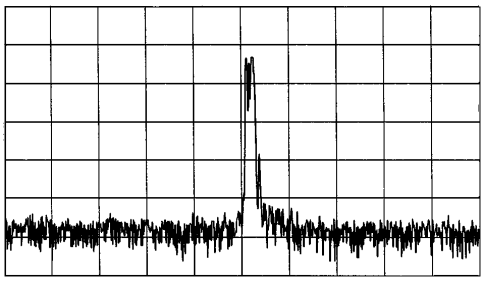

Fig. 3. Spectrum of the beat note between DFG and SHG 1596-nm waves. Vertical scale, 10 dB/division; horizontal scale, 500 kHz/division; resolution bandwidth 30 kHz; center frequency 6 MHz.

The powers at 1596 nm at the photodetector were 25 nW for the DFG beam and 1 μW for the SHG beam. A 60-μm-diameter InGaAs photodiode (bandwidth 5 GHz) was used to detect the beat note, which was amplified (40 dB gain, 60 MHz bandwidth) and sent to a spectrum analyzer (Fig. 3). The 40-dB signal-to-noise ratio should be adequate to phase lock the two lasers together. Fast frequency control of the CO laser is now being developed for this purpose. A frequency lock between the lasers has been established and gives a beat note with a 150-kHz linewidth (owing to the jittering of a narrower peak under acoustical and discharge noise in the CO laser) and a 1-kHz center frequency stability.

We have demonstrated a coherent frequency connection in an exact 3:1 ratio between 1064 and 3192 nm. The fact that RTA noncritically phase matches this process is encouraging for the extension of this research to an OPO system. We also look forward to realizing the 1126–3378-nm link on the way to connecting the Hg^+ 282-nm transition to the primary Cs standard.

We thank G. M. Loiacono (Crystal Associates, Inc.) for growing the RTA crystal, R. A. Stolzenberger (Crystal Associates, Inc.) for information about RTA, and S. Waltman and J. L. Hall for significant contributions to this research. This research is supported in part by the U.S. Air Force Office of Scientific Research. M. Mürtz acknowledges support from the German Science Foundation; O. Pfister acknowledges a Lavoisier grant from the Ministère des Affaires Etrangères of France.

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