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OPTICAL FREQUENCY SYNTHESIS AT NPL

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Abstract

Current activities relevant to laser frequency measurement are discussed. The main emphasis is an extending frequency measurement from the $3.39\ \mu\text{m}$ region to the visible, for which a particular frequency synthesis scheme is presented. Experimental results are given on phase locking gas lasers together and on checking the equivalence of frequency doubling in a phase-matched crystal and in a point contact device. Technical improvements of Josephson junction devices are mentioned.

About two years ago we obtained an accurate result for the frequency of a methane-stabilized laser at $3.39\ \mu\text{m}$, and were able to refer the results to the mean frequency of several such lasers¹. The technique involved making counts of four beat frequencies simultaneously under calculator control to avoid errors from frequency instabilities of the transfer-oscillator lasers, and the final uncertainty was estimated at ± 3 parts in 10^{11} . We hoped the result would provide a base for extending frequency measurements towards the visible and act as an interim frequency standard for length measurements. This hope remains, although continuity of operation of methane-stabilized lasers was lost at NPL shortly after that experiment was finished.

Our present main objective is to measure the frequency of a visible stabilized laser by reference to the methane stabilized laser or to the caesium standard. Dr G J Edwards has proposed a scheme to reach the $633\ \text{nm}$ I_2 stabilized He-Ne laser by using two successive non-linear mixing stages from an F-centre laser at $2.53\ \mu\text{m}$. However, our experimental results here described come from two subsidiary experiments: (i) a feasibility study on phase-locking two lasers together (at an offset frequency), and (ii) an investigation of the exactness of frequency doubling in a phase-matched crystal, as compared with a point-contact device. The former experiment involved phase-locking a small $3.39\ \mu\text{m}$ He-Ne laser and a $6.2\ \text{m-long}$ one together at a $5\ \text{MHz}$ offset frequency, and the latter was performed with CO_2 laser radiation doubled to the $5\ \mu\text{m}$ region.

The main practical interest, of course, in measuring the frequencies of visible stabilized lasers is to provide working length standards under the proposed new definition of the metre, in which the speed of light will become fixed. There are a number of visible stabilized lasers used as laboratory length standards in different parts of the spectrum, and it is also interesting to devise means of measuring the frequency differences between them, for this is intrinsically more powerful than using precise interferometry to calibrate the standards for precise interferometry itself.

Table I shows some frequency intervals between visible stabilized lasers, from which it appears that CO_2 , frequency-doubled CO_2 , CO or far-infrared optically-pumped lasers can provide suitable radiations if non-linear elements such as phase-matchable crystals can be found for the addition or subtraction of the frequencies.

Table I.- Possibilities for synthesis of some visible-laser frequency separations.

Visible stabilized lasers								
No.	Laser	Ref. Molecule	λ/nm , colour	Visible interval	Infrared laser			residual beat (GHz)
				(THz)	gas	line	f (THz)	
1	Ar*	I ₂	515 green	$f_1 - f_2$ (62.2838)	CO ₂	9.62 μm P(28)	31.1595 x 2	- 35.3
2	doubled He-Ne (or dye)	I ₂ "0"	576 yellow		CO	1 - 0 P(22)	62.2768	+ 6.9
					CO	2 - 1 P(10)	62.2711	+ 12.6
3	He-Ne	I ₂	612 orange	$f_2 - f_3$ (30.3265)	¹³ CO ₂	9.88 μm P(8)	30.3150	+ 11.5
					¹³ C ¹⁸ O ₂	9.88 μm P(20)	30.3398	- 13.3
4	He-Ne	I ₂	633	$f_2 - f_6$ (64.11)	CO ₂	9.35 μm R(6)	32.0482 x 2	+ 14
5	He-Ne	I ₂	640		CO	1 - 0 P(8)	64.0982	+ 12
6	dye	Ca (beam)	657	$f_4 - f_5$ (5.394)	CD ₃ OH	55.56 μm	5.396 (26)	-(2 ⁺ 26)
					CH ₃ OH	55.37 μm	5.4143	- 20

One of my own activities, and an increasingly arduous one, is to maintain a list of far-infrared CW laser lines as sources of coherent radiation in that part of the spectrum². Histograms of the distribution with frequency are shown in figure 1. The number of optically-pumped lines reported is still increasing, for another 450 entries now await adding to the list. However, there is little sign of a change in the distribution to fill the "gap" between about 4 THz and 24 THz in which only 8% of the lines fall. The extensive coverage by CO₂ radiation starts just above 24 THz³. It must be admitted therefore that the list may not be of great use in finding sources at a particular frequency near, say, 17 THz, although it will list what there is. (The main demand for the list is from molecular spectroscopists using far infrared lasers).

Fig.1 Spectral distribution of far-infrared cw laser lines.

Laser phase-locking at $3.39\ \mu\text{m}$

A student, J C Coveney from Cambridge University, has helped carry out some experiments on phase-locking two lasers together. A 6.2-m long He-Ne laser used as a power transfer oscillator, and a short (0.3-m) laser were phase locked together via a quartz-crystal-oscillator reference at 5 MHz.

Fig.2 Scheme for phase-locking two $3.39\ \mu\text{m}$ He-Ne lasers together.

At first a frequency offset lock based on a frequency to voltage converter was used to "assist" a weak but fast phase-lock loop (PLL) based on driving a LiNbO_3 crystal in the long laser cavity (suggested by G J Edwards). However a frequency-lock system has zero error signal under phase-locked conditions and so can not increase the lock hold range against drift, for example. Hence the frequency lock was put aside and a second, slow, phase-error feedback signal was applied to a piezoelectric transducer (PZT) driving an end mirror of the small laser, see Fig.2. The PZT had resonances in the 2 kHz region which had not to be excited. The PLL now had two drives in parallel, which were coupled via the laser beat frequency, so it was necessary to use time constants slow enough to avoid relative phase shifts between them in the region where there is net gain. With a "fast" second order loop, having $\omega_n \sim 34 \times 10^3$ rad/s, and a slow loop with integration time constant ~ 0.8 s, it was possible to achieve a phase-lock jump-in range of ~ 140 kHz (from the fast loop), and a hold-in range of ~ 10 to 17 MHz (from the slow loop), thus offering hold times of about 100s with a relative fractional drift of 10^{-9} s between the lasers. As finally set up, the fast PLL alone could only momentarily catch phase lock on its own, but with both loops connected the system was stable enough to flip from the $+5$ MHz to the -5 MHz lock sidebands when the end plate of a laser was pushed. The "slow" loop was found to be carrying significant feedback signals out to about 1 kHz.

Fig.3 Phase spectral density of beats between two $3.39 \mu\text{m}$ He-Ne lasers. Data for Fourier frequencies below about 100 Hz is interpolated from square-root Allan variance plots. The W-locus intercept gives a measure of the FWHM of the spectrum⁷.

The performance of the double PLL was constrained on the one hand by all the perturbations affecting the lasers and on the other hand by the speed of the "high voltage" driver amplifiers⁴, each having about 10 kHz bandwidth.

Once the loop was locked it was possible to investigate a number of sources of perturbations of the lasers. The laboratory was the same as that used for the methane-stabilized laser frequency measurement¹ and is on the second floor above ground of an "office-block" style building, with fan and vacuum-pump noise, but most of the frequency deviations from these causes fell below about 400 Hz, in the region where the loops have substantial gain. The perturbations affecting phase-locking were mainly associated with the dc-plasma excitation of the lasers - "white" frequency noise peaking near 4 kHz on the long laser, which strongly affected phase-locking, and possibly ion-acoustic waves in the small laser near 500 kHz, which were weak. Both could be reduced by reducing the laser current. A loop-induced oscillation tended to occur near 40 kHz. Otherwise 'spikes' seen on one occasion as vibration "bumps" at 2 to 10s intervals, and clicks corresponding to the switching of a nearby temperature-controlled soldering iron (at 13s intervals) were also capable of upsetting the quality of phase-lock, if not of unlocking the system.

The spectral quality of the phase lock is shown in the phase spectral density plot Fig.3, where it is compared with the free-running and frequency-locked conditions. The data is derived from both square-root Allan variance measurements and from the spectrum of the locked beat. The FWHM from phase-noise, W , ($\sim$ Halford's "fast line width") is ~ 1 Hz phase locked and ~ 20 kHz frequency-locked or free-running. The free-running linewidth FWHM on a spectrum analyser was about 100 kHz when taking about 0.1 s to cross the signal.

It seems likely, but has yet to be proved, that ordinary microwave phase-locked boxes could usefully phase lock lasers in this way, provided a dual feedback system is attached, and offset-locking beyond the usual range of frequency to voltage converters is clearly possible. In our trials the beat detector limited the useful beat frequency, but by correct biasing this can be extended to hundreds of MHz with the same type of diode.

Test of "exactness" of frequency doubling in a crystal

This experiment involves the frequency doubling of the radiation of a CO_2 laser in a crystal and subsequent 2nd-harmonic mixing with a second laser², in an MIM diode. The scheme is shown in Fig.4 and the work has mainly been done by G J Edwards and P R Pearce assisted by a student, R J H Jennings, from Bristol University. The doubling crystal was of CdGeAs_2 , and we are grateful to N Menyuk⁵ of MIT Lincoln Laboratory for supplying it⁴.

The experiment was much more difficult to set up and operate than had been anticipated. For example it was necessary to spatially filter the (~ 8 W) output of the laser incident on the doubling crystal, to convert its "doughnut" output mode to a near-Gaussian one, before adequate doubling efficiency could be realized. An iris also had to be fitted in this laser's cavity to prevent multimoding when the PZT tuning was adjusted.

With about 1 mW of $5\text{ }\mu\text{m}$ radiation from the doubling crystal, a beat with ~ 30 dB signal/noise in 100 kHz bandwidth was obtained by 2nd harmonic mixing with the second CO_2 laser in an MIM diode.

Fig.4 Scheme for comparison of frequency doubling in a crystal and in an MIM point-contact diode. The sapphire blocks $10\text{-}\mu\text{m}$ radiation.

Simultaneous frequency counting by two high-precision counters controlled by a calculator was used to test for differences between twice the fundamental frequency beat, $2\Delta f$, and the second harmonic beat, $\Delta(2f)$. Δf was set at 5 MHz by a frequency-lock loop and counting was done via filters at 5 and 10 MHz having bandwidths of about 2 MHz. The program took 50 pairs of 1-second counts, alternately reversing connections to the counters.

Fourteen such sets have been obtained in experiments on three different days, within a week. Counts were also made for 10 s averaging times, and test counts were taken in each run by putting the same, weaker, 10 MHz beat $\Delta(2f)$ into both count channels. Earlier checks were made using a synthesiser modulated with "white-frequency" f.m.

The test counts showed a system resolution of 0.02 ± 0.04 Hz. The overall result for $[\Delta(2f) - 2\Delta f]$ gave closely the same result, showing that any net difference arising in the two frequency-doubling processes has a fractional magnitude less than 7 parts in 10^{10} . Further experiment is possible and some "calibration" needs to be introduced, perhaps by introducing a time-dependent phase shift through a rise or decrease in temperature of a transmission element, for example.

F-centre laser at $2.5\ \mu\text{m}$ and scheme to reach $633\ \text{nm}$

The scheme of the experiment is shown in Figure 5, and was devised by Dr G J Edwards, who is mainly responsible for the F-centre laser and the two non-linear stages to $633\ \text{nm}$. After frequency doubling the $2.53\ \mu\text{m}$ radiation in LiNbO_3 , possibly inside the F-centre laser cavity, the $1.27\ \mu\text{m}$ radiation enters another LiNbO_3 crystal inside the cavity of a large He-Ne laser operating at $633\ \text{nm}$, which acts as a near-degenerate parametric amplifier. The beat between signal and paramp idler at $1.27\ \mu\text{m}$ then measures the $633\ \text{nm}$ pump frequency.

Fig.5 Scheme to reach $633\ \text{nm}$ from a colour-centre tunable laser at $2.5\ \mu\text{m}$.

On progress, the F-centre laser is working and the basic equipment is built. However neither frequency measurement of its radiation against a CO_2 harmonic, nor adequate frequency doubling has yet been demonstrated, nor has the $633\ \text{nm}$ paramp-pump laser (6-m long) yet been run single mode with the crystal in the cavity.

All these stages must be demonstrated before the frequency synthesis experiment can be put together, in turn before any frequency measurements can be made. It seems unlikely that with our present staff of rather less than three people we can make a visible frequency measurement by this route before the recommendations on the new definition of the metre are formulated next year, but we shall try.

Development of point-contact Josephson junction devices

Work has been carried out in the group, under T G Blaney, primarily by N R Cross and Wu Pei Heng, a guest worker from Nanking University, on (i) construction of efficiently-coupled devices as mixers for submillimetre astronomy, and (ii) on making up glass-encapsulated point contacts which can be temperature cycled without serious degradation.

The efficient mixer junctions make use of conduction-cooled helium cryostats which (i) remove the junction from the liquid helium and so avoids perturbation of the input coupling by bubbling, and (ii) offer good access to, and a greater volume for, the mounting of the junction. The cryostats used are also much less bulky than those used before, and can easily be fitted on a bench. They are about 500 mm high by about 300 mm in diameter, and the 1.5 litres of helium lasts for 15 hours.

The efficiency of coupling radiation to the device, and the degree of absence of noise in the operation is demonstrated by Cross' recently obtaining a noise temperature near 400 K, double sideband, at 230 GHz, and near 2200 K at 460 GHz, for fundamental-frequency heterodyne mixing. This was done with a room-temperature IF amplifier and a rather poor IF line.

We plan experiments to try to extend frequency measurements beyond our previous limit of 4.25 THz⁶ using this new apparatus, and possibly higher energy-gap materials.

The program to try to make temperature-cyclable i.e. "permanent" Nb-Nb point contacts has met with a certain amount of success. Preset contacts are sealed into glass tubes in an inert atmosphere. Some 4 or 5 such junctions now exist that exhibit Josephson behaviour when irradiated with HCN laser radiation at 890 GHz and which have been immersed in liquid helium on several occasions. They are to be tested for measuring the laser's frequency by harmonic mixing with microwaves.

The group also looks ahead to the possibility of fabricating evaporated film devices suitable for frequency measurements at THz frequencies.

Conclusions and Comments

The recent experiments have shown up inadequacies in our equipment, partly because it is getting old and there are fewer people now to keep it going, and partly because the use of non-linear crystals in the CO₂ frequency-doubling experiment, for example, makes greater demands on the quality of the equipment than mixing with point-contact devices. The stiffness of the mounting table (not our best) and the degree of isolation achieved between crystal and laser were such that fluctuations of 2nd harmonic amplitude could be quite large.

However, we have established that phase-locking our lasers together is not too difficult, and produces a dramatic reduction in the beat spectral width, and we have obtained a useful upper limit of 7 parts in 10¹⁰ for any differences in frequency doubling between a point-contact and a phase-matched, bulk, device. There is room for hope now that we can make faster progress with the synthesis scheme to the visible using the colour-centre laser.

We welcome efforts in other laboratories to extend the frequency range of point-contact devices as mixers in the visible, and we also hope that a newly-arrived guest worker will help us extend the range of mixing in Josephson junctions beyond 4 THz, towards CO₂ frequencies.

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