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PROPOSAL FOR A LARGE SCALE SPECTROMETER

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Abstract

A laser spectrometer is proposed which may offer the measurement of frequency differences between lasers at visible wavelengths with an uncertainty of 1 part in 10^{12}. The method consists in sweeping a dye laser over the frequency range in question and to sum up the frequencies measured stepwise.

Introduction

The metre definition of 1983 [1] transfers the problem of accurate length or wavelength measurement to the determination of optical frequencies. Activities in this field ended up with a frequency value for an infrared laser having a relative uncertainty of 1,3 parts in 10^{10}* and with two frequency values for visible lasers with uncertainties of 6 parts in 10^{10} and 1 part in 10^{9}, respectively. Lasers stabilized to molecular absorption peaks, however, have a reproducibility of at least several parts in 10^{11}. The current method for comparing the absolute frequency references with other visible oscillators such as stabilized lasers or molecular absorption and emission lines is interferometry. This method, although attaining uncertainties of several parts in 10^{11} [2], seems to be limited essentially by the quality of optical components and by diffraction

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* In accordance with ISO 31/0-1981(E) the decimal sign is a comma.

effects. But the efforts made to establish highly stable and reproducible frequency standards in the visible [3, 4] require considerable improvements for absolute frequency measurements in this field. Supposing that improvements will be achieved for some particular optical frequencies, the problem in comparing visible frequencies one with another will still not be resolved. A first project for comparing optical frequencies by the use of stabilized dye lasers separated by steps of 80 GHz has already been started [5]. Here, a similar experiment is proposed which also promises to solve this problem. Recent developments in establishing very stable, low-drift and high-finesse optical resonators [6, 7] may permit one to reduce the uncertainty for large-scale frequency comparisons to less than one part in 10^{11}.

Description of the spectrometer

Two similar types of spectrometers will be described. The first one is shown in Fig. 1. The essential part is a non-confocal Fabry-Pérot resonator, which will be isolated from environmental influences such as temperature and acoustic waves, as well as from variations of the refractive index of air. This can be achieved by placing the resonator in an evacuated and temperature-shielded housing, which in turn contains a temperature-controlled box having a good thermal conductivity. The mirror spacing of the resonator should be of a material having a low temperature coefficient such as zerodur, quartz or invar. Room temperature would promise a good stability, but very low temperatures, for example that of liquid helium, would not only reduce thermal expansion effects but also thermal noise. The mirrors of the resonator should have a very high reflectivity combined with a large bandwidth. Reflectivities allowing resonator finesses of more than 10000 have already been reported [8, 9]. Stabilization techniques for locking a laser on to such a resonator are possible with 2 Hz-precision at a cavity transmission linewidth of 80 kHz and a free spectral range (fsr) of 240 MHz [8]. A linewidth of 14 kHz at a fsr of 315 MHz has also been reported [9]. This resonator should not only have a good stability and a high finesse, but its fsr must also be known with a high accuracy. One way to measure the fsr is to stabilize a laser on a resonance fringe of the resonator and to stabilize a frequency-shifted part of the beam of the same laser to an adjacent fringe by means of a side-band-generating phase modulator [9, 10] or by an acousto-optic modulator (AOM). The fsr will then be identical to the modulation frequency of the modulator, which may be furnished by a calibrated frequency synthesiser with at least 1 Hz-accuracy at a frequency of 1 GHz. The frequency noise of the laser will be completely eliminated with this method.

For one of the two lasers to be compared, laser No 1, there should be established another non-stabilized monomode and tunable version, used as offset laser, which will be locked to the passive resonator by means of the phase-modulation method described elsewhere [6]. The frequency difference between the offset laser and the laser No 1, for example an iodine-stabilized He-Ne laser, will be measured by using the beat-frequency method and will be indicated as frequency f_1 (Fig. 1). Similar to the offset laser, the radiation of a prestabilized tunable dye laser is phase-modulated and mode-matched into the passive resonator. Its reflected signal will be rectified by using a double balanced mixer and will be used either for counting the resonator modes, detected as reflected fringes during the dye-laser frequency sweep, or for stabilizing the dye laser on to a resonator fringe. The frequency difference between the dye laser and the offset laser or the other laser to be compared, laser No 2, will be measured as frequency f_2 or frequency f_3, respectively (Fig. 1). The continuity in counting the cavity orders during mode changes of the dye laser may be maintained by using a wavemeter or a slave laser. In order to measure the fsr, a second phase modulator having a modulation frequency of the fsr may be applied to the dye-laser beam and a corresponding photosignal will be rectified for locking the modulation frequency to the appropriate cavity fringe [9].

A typical measuring cycle would start at a state where the offset laser and the dye laser are locked to the same resonator fringe, which is verified by observing frequency f_2. Frequency f_1 will be memorized and as one starts the dye-laser sweep one begins to count the resonator fringes. When the frequency difference between the dye laser and laser No 2 are in a range which can be measured, the dye laser will be locked on a resonator fringe. The frequency f_3 will then be memorized as well as the new frequency f_1, which could have changed due to drifts of the resonator. The frequency of the fsr of the passive resonator as a function of the laser frequency should be determined. The frequency difference between laser No 1 and laser No 2 is then calculated as the sum of : the product of the mean fsr and the counted number of cavity fringes, the memorized frequency f_3 and the mean of the two frequencies f_1.

A second type of spectrometer (Fig. 2) compares the beat frequency between two dye lasers with an atomic-clock-controlled millimetre-wave oscillator having a frequency of the order of 100 GHz by means of a Schottky diode [5, 11]. (Complete systems of solid state millimetre-wave sources at about 60 GHz, phase-locked to an external driving frequency in the MHz range, are already available). This will give an accuracy for the frequency difference that will only be limited by the stability of the lasers involved. In this type of spectrometer, two cavities will be locked to the stabilized laser No 1 and to each of these cavities a dye laser will be locked. The frequency difference between the two dye lasers will be the sum of the frequency of the millimetre-wave oscillator f_m and the frequency f_2, the latter being of the order of 1 GHz. The dye-laser frequencies will be changed in steps of $2f_m$, covering the frequency range between the two lasers to be compared. The sum of all these successively-measured frequency steps and of the frequencies f_1 and f_3 will be the frequency difference between the two lasers in question.

Uncertainties and problems

The frequency dependence of the fsr on the phase shifts in the resonator needs to be dicussed in more detail. Phase shifts ϕ_D and ϕ_R of a resonator are induced by diffraction and by reflection on the mirrors, respectively, supposing the resonator is in vacuum. Another phase shift ϕ_{MM} may be caused by imperfect mode-matching. If the diameters of the mirrors are large compared to the beam diameter, the diffraction phase shift does not depend on the wavelength ; it will be :

$$\phi_D = (m + n + 1) \arccos \left[(1 - L/R_1)(1 - L/R_2)\right]^{\frac{1}{2}}/(2\pi) \qquad (1)$$

m, n = transverse mode-numbers

R_1, R_2 = radii of curvature of the mirrors

L = length of the cavity

The frequency of the resonant laser radiation f is calculated as follows :

$$f = (N + \phi_D + \phi_R + \phi_{MM}) \frac{c}{2L} \qquad (2)$$

N being the number of axial orders and c the speed of light. If a sideband of the laser radiation, separated from f by the frequency f_m, is tuned to another cavity resonance, differing by n axial orders from N and suffering by additional phase shifts $\Delta\phi_R$ and $\Delta\phi_{MM}$, one obtains :

$$f = \frac{N + \phi_D + \phi_R + \phi_{MM}}{n + \Delta\phi_R + \Delta\phi_{MM}} f_m \qquad (3)$$

and :

$$f_m = (n + \Delta\phi_R + \Delta\phi_{MM}) \frac{c}{2L} \qquad (4)$$

Thus, the frequency f_m, which is the fsr, depends on the change of $\Delta\phi_{MM}$ and that of $\Delta\phi_R$, which are both functions of the laser frequency. That means that f_m will not be constant. But for the described spectrometer, the frequency f_m can be measured at any laser frequency f lying in between the frequencies f_1 and f_2 of the two lasers to be compared. If f'_1 and f'_2 are the frequencies of the two cavity orders which are next to the laser frequencies f_1 and f_2, the difference $f'_2 - f'_1$ will be calculated as follows :

$$f'_2 - f'_1 = k\,\overline{f_m} \quad ; \quad \overline{f_m} = \frac{1}{k} \sum_{i=1}^{k} f_{mi} \qquad (5)$$

k being the total number of cavity orders between the two lasers.

If all f_{mi} are known, there is no need to care about phase shifts. Moreover, if the dependence of the f_{mi} on the laser frequency is continuous and shows no strong oscillations of a period of the order of the fsr or less, it will be sufficient to measure the fsr for only a fraction of the k cavity orders and to interpolate to find the values of the other ones. It is known that interference mirrors show a stronger frequency dependence on the reflection phase shift than do metal mirrors, but the structure of this dependence should not be considerably different from that of the frequency dependence of their reflection coefficient. Also, when scanning the laser frequency, a sort of quantum frequency noise may appear for the fsr, if the size of the beam spot on the mirrors changes by the radius of a surface molecule. At a nearly confocal resonator with a fsr of 1 GHz this condition is fullfilled for a frequency change of 1 fsr. But the surface of an atom relative to that of the whole beam spot is about 3×10^{-10}. Even if one single atom would cause an extra phase shift of π, the resulting noise amplitude would be 0,3 Hz. More important might be a phase noise produced by surface defects. But this noise should appear at frequencies larger by some orders of magnitude. In all cases, the overall uncertainty will be increased as a function of the number of non-vanishing derivatives of the frequency dependence of the fsr and as a function of its statistical noise observed in scanning the laser frequency. The idea to measure the absolute frequency by using (2) [9, 10] would be more difficult because the absolute phase shifts ϕ_D, ϕ_R and ϕ_{MM} are unknown.

The uncertainty of the frequency difference between laser No 1 and laser No 2 will be the root-mean-square combination of : the uncertainty of $\overline{f_m}$ (comprising phase shifts and temperature drifts) multiplied by the square root of the counted number of cavity fringes k, the uncertainty of f_1 and the uncertainty of f_3.

As an example, the recommended frequencies of visible lasers [1] and other laser frequencies capable of being stabilized will be considered (Table 1). The maximum frequency difference between adjacent lasers is about 31 THz. Assuming a fsr of 1 GHz for the passive resonator, the number of cavity fringes to be counted between adjacent lasers is equal to the number of GHz given in Table 1. Supposing that a minimum accuracy for the mean fsr will correspond to an uncertainty of 100 Hz, then the total uncertainty arising from the resonator will be 18 kHz. That part of the uncertainty of either f_1 or f_3 which arises from the offset laser or from the dye laser, respectively, both being stabilized to the passive resonator, may be 30 Hz. Thus the root-mean-square combination of the spectrometer contributions of the uncertainty will be 18 kHz or 3×10^{-11} of the laser frequency for a frequency difference of 31 THz.

The solution that eliminates the problem of the frequency-depending fsr is the use of a millimetre-wave oscillator. If a Cs-clock-stabilized 100 GHz oscillator having an uncertainty of less than 1 Hz is used and if the dye lasers and the lasers to be compared have an uncertainty of 30 Hz and 10 Hz, respectively, the total uncertainty of the spectrometer will be 530 Hz or 1×10^{-12} of the laser frequency, for the largest frequency difference in Table 1. Here, the limitation in uncertainty is mainly determined by that of the dye lasers. The uncertainty could even be reduced if an oscillator with a higher frequency were used. Commercial carcinotrons, for example, are available for up to more than 600 GHz.

Other problems may arise for large frequency differences between the two lasers to be compared. Limitations are given by the tuning range of the dye laser and by the bandwidth of the mirror coatings of the passive resonator. The limitation in the tuning range of the dye laser, which might be about 50 nm of the wavelength, can be overcome by using a second or a third dye laser having the complementary spectral ranges. The limitation in the mirror coating of the resonator may be important for the first type of spectrometer and could be overcome in the following way. A second resonator having different mirrors is needed. At the frequency limit between the two resonators the dye laser will be locked to the resonator first being used. A second dye laser tuned to about the same frequency will be locked on the second resonator. After the difference frequency between the two dye lasers has been measured the second dye laser continues the sweep on the second resonator correspondingly. Another way to solve this problem would be to establish intermediate independently-stabilized lasers and to do the frequency measurements between adjacent stabilized lasers by using appropriate dyes for the dye laser and appropriate mirrors for the passive resonator. There is already a certain number of wavelengths at which lasers have been stabilized (Table 1) and which may be sufficient for this purpose. At any other wavelength larger than 500 nm, dye lasers could easily be stabilized on iodine absorption features ; at wavelengths smaller than 500 nm, Te_2 absorption lines may be suitable as reference frequencies.

Infrared lasers like the methane-stabilized He-Ne laser at 3,39 µm wavelength could be compared with visible ones by mixing their frequencies with that of other lasers [12, 13] or by using a multiple of their frequencies, obtained by harmonic generation in a non-linear medium, the two methods offering an optical frequency coinciding with the tuning range of a wide-band tunable laser like a dye laser.

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Table 1.- He-Ne laser, Ar^+ laser and special dye laser wavelengths λ to be stabilized on Ca or I_2 absorption features, their successive frequency differences Δf and appropriate dyes for the scanning dye laser.

λ	Δf	dyes
657 nm		
	12 231 GHz	DCM
640		
	5 394	DCM
633		
	16 268	RB (DCM)
612		
	30 326	R6G
576		
	31 373	R110
544		
	30 911	C6
515		
	14 875	C102
502		

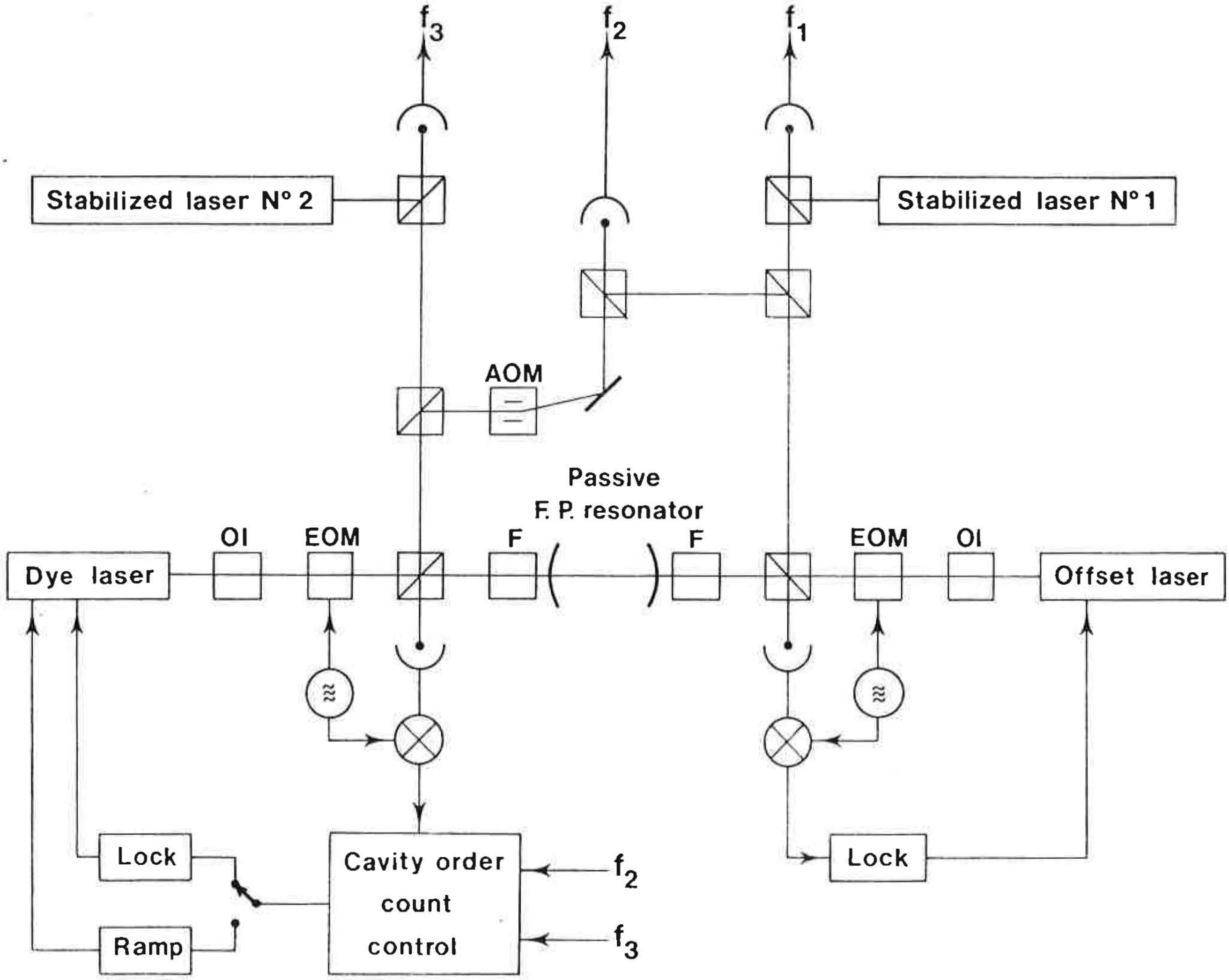

Fig. 1.- Spectrometer set-up. EOM : electro-optic modulator ; AOM : acousto-optic modulator ; OI : optical isolator ; F : Faraday rotator ; f_1, f_2, f_3 : frequencies to be measured.

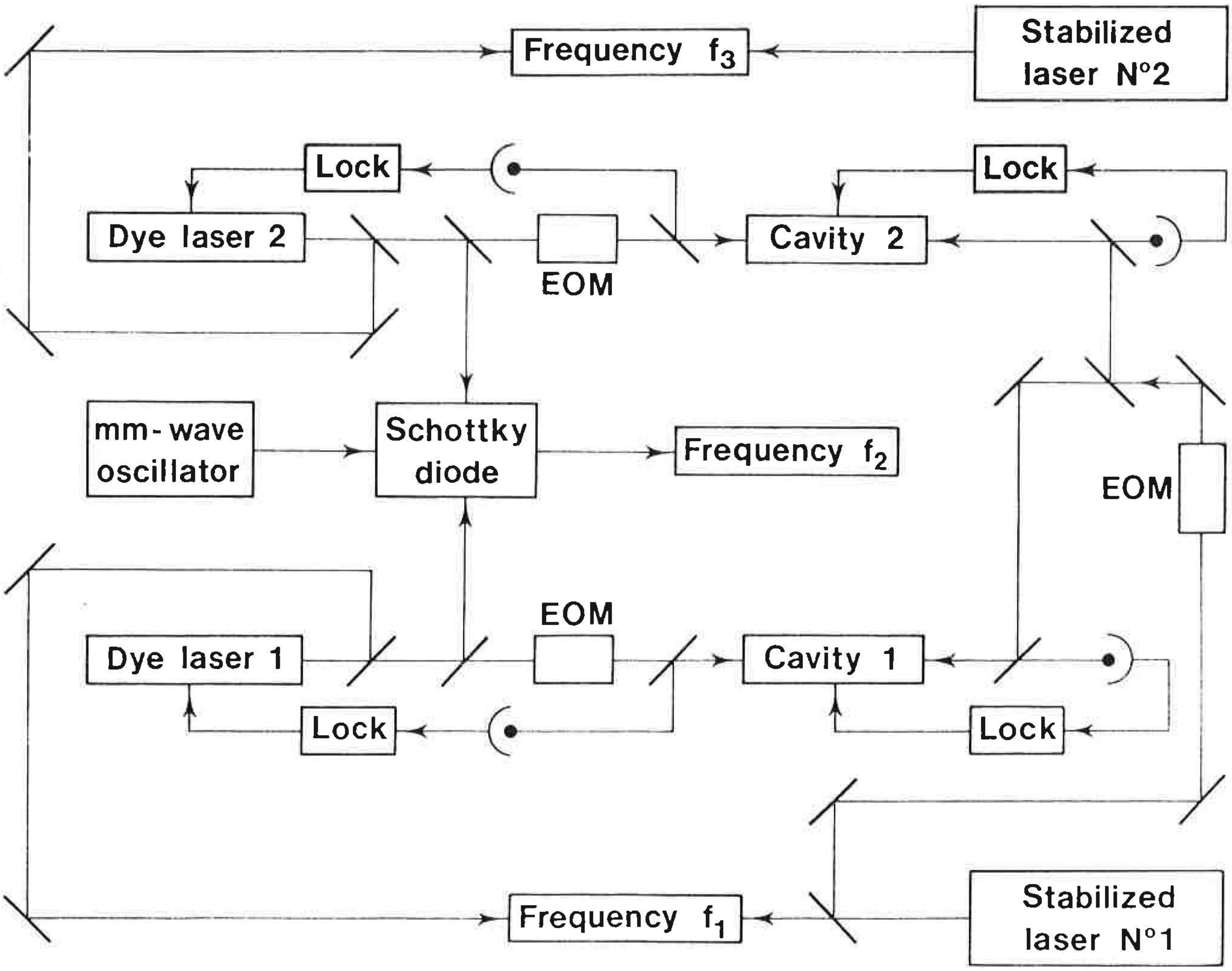

Fig. 2.- Spectrometer with a millimetre-wave source (MMW) and two dye lasers.