Spectrometry with frequency combs

S. Schiller

Institut für Experimentalphysik, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany

Received December 19, 2001

A method is proposed for performing accurate spectral characterization of samples with frequency combs, e.g., from mode-locked lasers, over the spectral range covered by the combs. The essence of the method is the use of two combs of slightly different mode spacing to achieve spectral resolution. The advantages of the method are speed, frequency resolution, sensitivity, absence of dispersive components, and high spatial resolution.

OCIS codes: 300.6190, 140.4050.

Frequency combs recently revolutionized the field of optical frequency metrology. A central development has been the demonstration that the spectral width of optical combs, produced, e.g., by mode-locked Ti:sapphire lasers, can be extended to more than an octave by self-phase modulation in optical fibers.[1,2] The mode spacing (i.e., repetition rate) and the absolute frequencies can be phase locked to rf references, thereby providing a comb of ultrastable optical frequencies.[3,4] Several highly precise comparisons between optical and rf oscillators have already been demonstrated.[5–7] The huge emission width, which could be further extended by use of additional nonlinear mixing steps, and the regular mode spacing of frequency combs suggest the question of applicability to spectroscopy. Multilevel spectroscopy with a single comb has been proposed.[8]

In this Letter a novel scheme is proposed for measuring the transmission (or reflection) and the phase properties of a sample over the large spectral range furnished by a frequency comb source. Two features of the source are made use of: the broadband emission and its discrete spectral structure. By use of two combs, FC1 and FC2, with slightly different mode spacings, ω_{ax1} and ω_{ax2}, i.e., with differing repetition rates, spectral resolution is achieved without any frequency tuning. FC1 interrogates the sample; FC2 serves as a reference that allows one to read out the changes of amplitude and phase that each mode of the interrogating comb has suffered. This is achieved by means of interfering the two combs and measuring the amplitudes and the phases of the ensemble of rf beat notes that occur. The information carried by a particular rf beat arises from a specific single optical mode that interacted with the sample. In this respect the proposed method is fundamentally different from the established methods based on spatial dispersion (grating and prism spectrometers), interferometry (Fourier-transform infrared spectrometers), or tunable sources. The spectral resolution is limited only by instabilities of the mode spacings and of the absolute values of the comb frequencies. Since the comb frequencies can be precisely stabilized by locks,[3,4] spectral resolution can be extremely high. The proposed method yields a discrete spectrum, sampled at the frequencies contained in the interrogating comb. The spacing between these frequencies is equal to the pulse repetition rate, with values 1–10 GHz, suitable for many applications.[9] In this context we note that, for two femtosecond combs of equal mode spacing, precise locking as well as measurement of phase has been demonstrated.[10,11]

We begin our analysis by discussing transmissivity measurements with a frequency comb spectrometer, shown in Fig. 1. The field from FC1 propagates through the sample and is superposed with the field from FC2 at beam splitter BS2 for generation of a signal at photodetector PD2. A reference signal is generated at photodetector PD1 by superposition of the two fields from FC1 and FC2 before the former traverses the sample. Photodetectors PD1 and PD2 are assumed to be identical, and beam splitters BS1–BS4 are assumed to be 50/50 splitters.

The two electric fields, $E_1(t)$ and $E_2(t)$, emitted by the combs are $E_j(t) = {}^1\!/_2 \sum_{n=-\infty}^{\infty} a_{jn} \exp[i(\omega_{jn}t + \varphi_{jn})] + \text{c.c.}$ The modes' angular frequencies, (real) amplitudes, and phases are $\omega_{jn} = \omega_{j0} + n\omega_{axj}$, a_{jn}, and φ_{jn}, respectively. ω_{10} and ω_{20} are the two closest mode frequencies and are taken as reference frequencies. We keep the mode spacings, $\omega_{jn} - \omega_{jn-1} = \omega_{axj}$, constant, e.g., by locking them to a rf source. No particular properties of a_{jn} or φ_{jn} are assumed, except for a comparable spectral width of FC1 and FC2 and for pulse durations exceeding the repetition time difference (see below).

The total field at PD2 is $E_{\mathrm{PD2}}(t) = {}^1\!/_4 \sum_n t(\omega_{1n}) a_{1n} \times \exp[i(\omega_{1n}t + \varphi_{1n} - \phi_{12n})] - a_{2n} \exp[i(\omega_{2n}t + \varphi_{2n} - \phi_{22n})] + \text{c.c.}$, where $\phi_{jkn} = \phi_{jk}(\omega_{jn})$ are the total propagation phases of the mode n from comb j to PDk.

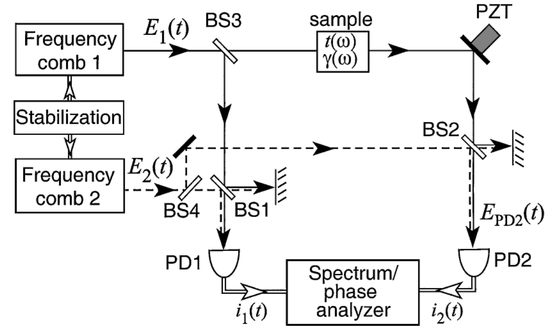

Fig. 1. Setup for measurement of transmission spectrum $t(\omega)$ and (or) phase spectrum $\gamma(\omega)$ of the sample. BS1–BS4, beam splitters; PD1–PD2, rf photodetectors; PZT, translator.

0146-9592/02/090766-03$15.00/0

ϕ_{12n} includes the sample's phase shift $\gamma(\omega_{1n})$, which consists of the propagation phase shift, $\omega_{1n}\tilde{n}(\omega_{1n})L/c$, through the sample of length L and refractive index $\tilde{n}(\omega)$, and interfacial phase jumps. $t(\omega_{1n})$ is the sample's (real) amplitude transmission coefficient for a particular mode n.

The photocurrent produced at PD2 is $i_2(t) \sim |E_{\mathrm{PD2}}(t)|^2$. It contains beats at the harmonics of the axial frequencies and at all possible combination frequencies, $\omega_{1n} \pm \omega_{2m}$. These intercomb beats are of interest here. The central idea is to have a small difference in the comb spacings, $|\omega_{ax1} - \omega_{ax2}| << \omega_{ax1}, \omega_{ax2}$; then, closely spaced low-frequency intercomb beats occur, stemming from the $n = m$ terms. They result in a contribution to the photocurrent, $i_2'(t) \sim 1/8 \sum_n \mathcal{R}_n t(\omega_{1n}) a_{1n} a_{2n} \cos[(\omega_{1n} - \omega_{2n})t + \varphi_{1n} - \varphi_{2n} - \phi_{12n} + \phi_{22n}]$, where $\mathcal{R}_n$ is the detector spectral response at $\omega_{1n} \approx \omega_{2n}$. In Fig. 2 these beats are indicated by short-dashed lines; they occur at frequencies $\Omega_n = |\omega_{1n} - \omega_{2n}| = |\omega_{10} - \omega_{20} + n(\omega_{ax1} - \omega_{ax2})|$. Thus the information about the sample's properties at the optical frequency ω_{1n}, i.e., $t(\omega_{1n})$ and ϕ_{12n}, is mapped onto the amplitude and phase of the beat note of frequency Ω_n.

An alternative picture of the measurement process can be given in the time domain. The differing pulse repetition rates $2\pi/\omega_{ax1}$ and $2\pi/\omega_{ax2}$ imply that at the photodetectors pulses from the two lasers overlap temporally only every $(\omega_{ax1} + \omega_{ax2})/2\Delta\omega_{ax}$ pulses, i.e., at a rate of $\simeq \Delta\omega_{ax}/2\pi$. We assume here that the pulse duration of one of the lasers is of the order of or exceeds the pulse repetition time difference, $|2\pi/\omega_{ax1} - 2\pi/\omega_{ax2}|$. The signal $i_2'(t)$ can then be interpreted as a stroboscopic sampling of the beat note $\exp[i(\omega_{10} - \omega_{20})t]$ between the reference frequencies, with sampling rate $\Delta\omega_{ax}/2\pi$. This results in a rf spectrum as shown in Fig. 2.

The proposed scheme has the following features:

(i) To avoid overlapping beat frequencies, one should ensure that $\Delta\omega_0$ differs from a multiple of $\Delta\omega_{ax}/2$. A suitable choice is $\Delta\omega_0 = \Delta\omega_{ax}/4$, which yields a regular spacing $\Delta\omega_{ax}/2$ between the beat frequencies, Ω_n.

(ii) The transmission spectrum, $t(\omega_{1n})^2$, is recorded in parallel; i.e., information at all frequencies ω_{1n} is obtained simultaneously.[12] The required integration time is set by two factors: desired dynamic range and signal-to-noise ratio. Because adjacent beats n, $n + 1$ in the rf spectrum stem from widely different optical mode frequencies, the spectral resolution of the rf spectrum must be chosen to be much higher than the rf beat spacing, $\Delta\omega_{ax}$, so as to yield accurate spectrum intensities. Moreover, the power of each rf beat is very low, since it arises from just two individual modes. An integration time of $\sim 10^5(2\pi/\Delta\omega_{ax})$ may be suitable.

(iii) Modes ω_{1n} that are sufficiently away from ω_{10}, namely, when $|\omega_{1n} - \omega_{10}| > \omega_{ax1}\omega_{ax2}/2\Delta\omega_{ax}$, give beats of lower rf frequency with modes ω_{2n+1} (for $n > 0$) or ω_{2n-1} (for $n < 0$), than with ω_{2n}. These beats complicate the rf spectrum, and it may be desirable to attenuate these distant modes of FC1 and FC2 appropriately with optical filters. The useful spectral width of FC1 is thus given by $\omega_{ax1}\omega_{ax2}/\Delta\omega_{ax}$, and the spectrum contains $N_{sp} = \omega_{ax2}/\Delta\omega_{ax}$ sampled frequencies. The axial spacing difference $\Delta\omega_{ax}$ should be chosen to be sufficiently larger than the beat linewidths. This requirement implies the need for high repetition rates. For example, with $\omega_{ax1} \simeq \omega_{ax2} \simeq 2\pi \times 3$ GHz, and $\Delta\omega_{ax} \simeq 2\pi \times 100$ kHz, the useful width will be $\sim$100 THz.

(iv) Denoting by A the desired frequency accuracy, we find that the rms deviations of the mode frequencies must be kept at $\sigma(\omega_{jn}) < A$ during the integration time, implying $\sigma(\omega_{j0}) < A$, and $\sigma(\omega_{axj}) < A/N_{sp}$. In addition, to avoid smearing out the rf spectrum, one must ensure that the differences $\Delta\omega_{ax}$ and $\Delta\omega_0$ satisfy $N_{sp}\sigma(\Delta\omega_{ax}) \ll \Delta\omega_{ax}$ and $\sigma(\Delta\omega_0) \ll \Delta\omega_{ax}$. For example, for $A \simeq 100$ MHz and the above values of $\Delta\omega_{ax}$ and N_{sp}, the requirements are estimated to be within current capabilities.[13]

We now turn to spectral measurements of the sample phase shift, $\gamma(\omega)$. This information is contained in the phases ϕ_{12n} in $i_2'(t)$. A comparison of the phases of corresponding beats in the currents $i_2'(t)$ and $i_1'(t)$ by electronic means will yield $\Delta\phi_n = \phi_{12n} - \phi_{22n} - \phi_{11n} + \phi_{21n}$, which contains $\gamma(\omega_{1n})$ and is independent of the unknown comb phases, φ. This difference phase contains contributions from paths that drift in time, so active stabilization, e.g., by the mirror translator (PZT) in Fig. 1, may be required. Active stabilization will keep a particular difference phase constant, providing a phase reference. The simultaneous recording of the phase spectrum, $\Delta\phi_n$, for a large number of modes ω_{1n} may, however, require too complex an electronic analyzer. It is therefore of interest to consider a simpler system.

Figure 3 shows a system that employs an optical interferometer and photocurrent power spectrum

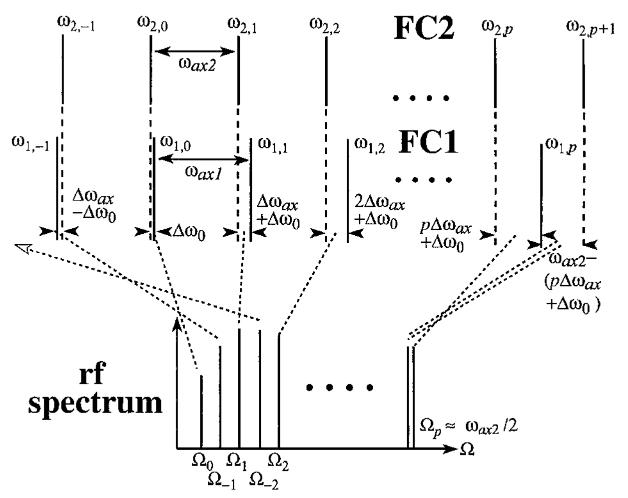

Fig. 2. Schematic of the modes of the two combs and relevant beats. The beats between modes ω_{1n} and ω_{2n} of equal index give rise to a rf spectrum with interleaving. Its discrete frequencies $\Omega_0, \Omega_{-1}, \Omega_1, \Omega_{-2}, \ldots$ arise alternatingly from modes below and above ω_{10}. Beats between modes of different index, e.g., $\omega_{2n+1} - \omega_{1n} = \omega_{ax2} - (\Delta\omega_0 + n\Delta\omega_{ax})$ decrease in frequency with increasing n and will overlap the rf spectrum of beats of equal index starting at the beat frequency $\Omega_p \simeq \omega_{ax2}/2$, once n is sufficiently large (see the double dashed lines).

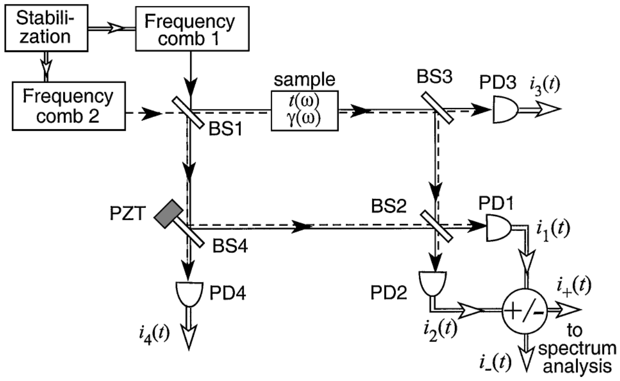

Fig. 3. Setup for measurement of the phase and the transmission spectrum. The former is determined from i_3 and i_- or i_+; the latter, from i_3 and i_4.

measurements. Here the waves of both combs travel down both arms of a Mach–Zehnder interferometer. The two output photocurrents i_1 and i_2, are summed and differenced to yield i_+ and i_-. Note that i_1 and i_2 differ because of phase jumps at beam splitter BS2. Two reference superpositions are prepared at BS3 and BS4, yielding currents i_3 and i_4. The electrical powers of the rf photocurrents arising from the beats of interest are

$$\begin{aligned} P(i_+{}', \Omega_n) &\sim (a_{1n}a_{2n})^2 \{[t(\omega_{1n})t(\omega_{2n}) - 1]^2/4 \\ &\quad + t(\omega_{1n})t(\omega_{2n})\sin^2(\delta\psi_n/2)\}, \\ P(i_-{}', \Omega_n) &\sim (a_{1n}a_{2n})^2 \{[(t(\omega_{1n}) - t(\omega_{2n})]^2/4 \\ &\quad + t(\omega_{1n})t(\omega_{2n})\sin^2(\langle\psi_n\rangle)\}, \\ P(i_3{}', \Omega_n) &\sim \frac{1}{2}(a_{1n}a_{2n})^2 t(\omega_{1n})t(\omega_{2n}), \\ P(i_4{}', \Omega_n) &\sim \frac{1}{2}(a_{1n}a_{2n})^2. \end{aligned} \tag{1}$$

Here $\delta\psi_n = \psi_{1n} - \psi_{2n}$, $\langle\psi_n\rangle = (\psi_{1n} + \psi_{2n})/2$, where ψ_{jn} is the difference between the total propagation phase shift along each arm of the interferometer for the mode ω_{jn} and includes the sample's phase shift, $\gamma(\omega_{jn})$.

Phase-dependent contributions $P'(i_+{}')$ and $P'(i_-{}')$ to $P(i_+{}')$ and $P(i_-{}')$, respectively, can be identified by a path difference modulation (using the piezoelectric transducer in Fig. 3). The ratio $P'(i_-{}', \Omega_n)/P(i_3{}', \Omega_n)$ essentially gives the phase information of the sample, more precisely, the mean $\langle\psi_n\rangle$ of the phase at ω_{1n} and ω_{2n}. $P'(i_+{}', \Omega_n)/P(i_3{}', \Omega_n)$ gives the differential phase $\delta\psi_n$ for ω_{1n} and ω_{2n}. In the case of a sample phase that varies slowly with frequency ω, the sample's optical path-length contributions to $\langle\psi_n\rangle$ and $\delta\psi_n$ are approximately $\omega_{1n}\tilde{n}(\omega_{1n})L$ and $-(\Delta\omega_0 + n\Delta\omega_{ax})[\tilde{n}(\omega_{1n}) + \omega_{1n}\partial\tilde{n}/\partial\omega_{1n}]L$, respectively. Thus, since $|\Delta\omega_0 + n\Delta\omega_{ax}| \ll \omega_{1n}$, the information from $P'(i_+{}')$ permits an increase of the dynamic range of the phase measurement (with a corresponding reduction in sensitivity), which is useful, e.g., when the sample's length L is large. Here again, active stabilization of the interferometer may be necessary. For spectrally smooth transmissivity, $t(\omega_{1n}) \simeq t(\omega_{2n})$, the transmission spectrum is obtained from the ratio $P(i_3{}', \Omega_n)/P(i_4{}', \Omega_n) = t(\omega_{1n})^2$.

In conclusion, a new approach to spectral measurements has been proposed. The method can be applied to both transmission and reflection measurements. Advantages of the method are extremely high frequency resolution and high spatial resolution, since the laser beams can be tightly focused onto the sample. Possible applications are composition analysis of molecular gases via their spectral fingerprints and microspectrometry.

I thank P. Antonini and D. Rovera for insightful discussions.

References

1. J. K. Ranka, R. S. Windeler, and A. J. Stentz, Opt. Lett. **25,** 24 (2000).
2. W. J. Wadsworth, J. C. Knight, A. Ortigosa-Blanch, J. Arriaga, E. Silvestre, and P. St. J. Russell, Electron. Lett. **36,** 53 (2000).
3. D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, Science **288,** 635 (2000).
4. R. Holzwarth, T. Udem, T. W. Hänsch, J. C. Knight, W. J. Wadsworth, and P. St. J. Russell, Phys. Rev. Lett. **85,** 2264 (2000).
5. S. A. Diddams, D. J. Jones, J. Ye, S. T. Cundiff, J. L. Hall, J. K. Ranka, R. S. Windeler, R. Holzwarth, Th. Udem, and T. W. Hänsch, Phys. Rev. Lett. **84,** 5102 (2000).
6. T. Udem, S. A. Diddams, K. R. Vogel, C. W. Oates, E. A. Curtis, W. D. Lee, W. M. Itano, R. E. Drullinger, J. C. Bergquist, and L. Hollberg, Phys. Rev. Lett. **86,** 4996 (2001).
7. J. Stenger, T. Binnewies, G. Wilpers, F. Riehle, H. R. Telle, J. K. Ranka, R. S. Windeler, and A. J. Stentz, Phys. Rev. A **63,** 021802-1-4 (2001).
8. T. H. Yoon, A. Marian, J. L. Hall, and J. Ye, Phys. Rev. A **63,** 011402-1-4 (2000).
9. If finer sampling is desired, one can record a set of interleaving spectra by shifting both frequency combs globally in frequency by appropriate amounts.
10. R. K. Shelton, L.-S. Ma, H. C. Kapteyn, M. M. Murnane, J. L. Hall, and J. Ye, Science **293,** 1286 (2001).
11. T. M. Fortier, D. J. Jones, S. A. Diddams, J. L. Hall, J. Ye, S. T. Cundiff, and T. Windeler, Proc. SPIE **4271,** 183 (2001).
12. An opposite, sequential, mode of operation is also possible, where detection is done at a fixed beat frequency but the reference comb as a whole is scanned in frequency in steps of $\Delta\omega_{ax}$. Thereby a small frequency step $\Delta\omega_{ax}$ of one comb as a whole is effectively amplified (e.g., by a factor of 10^6) to a large step ω_{ax1} in the sampled frequency. A phase measurement as suggested in the discussion of Fig. 1 is suitable for this sequential technique.
13. One could implement the stabilization of $\Delta\omega_0$ by beating a cw laser with each comb, combining the beats to yield $\Delta\omega_0$, and stabilizing one of the combs.