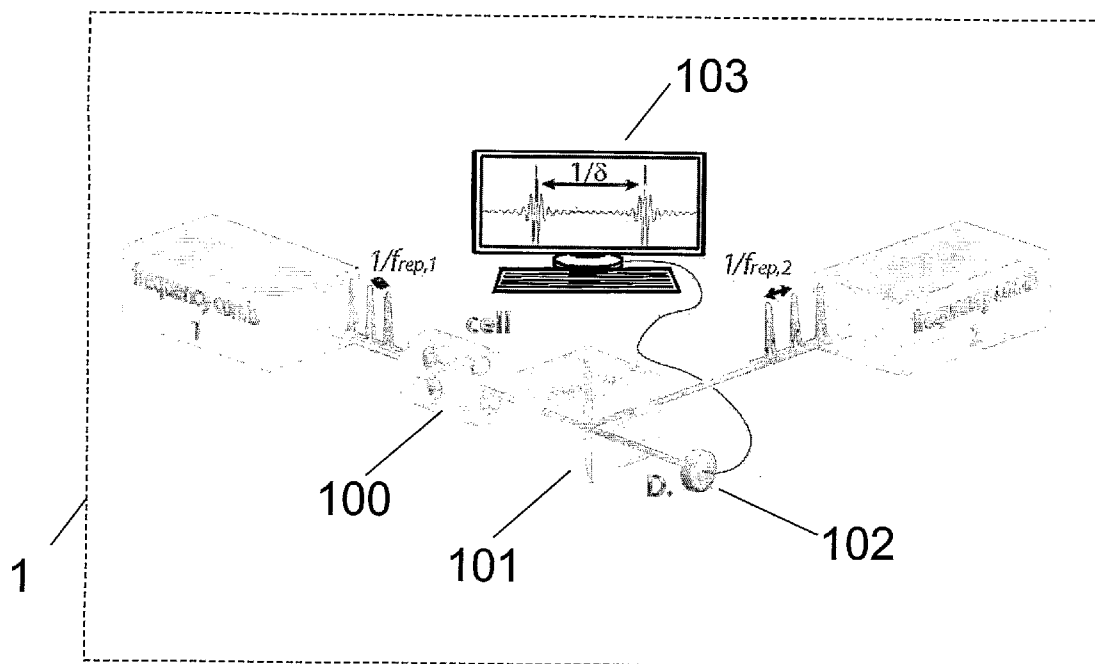


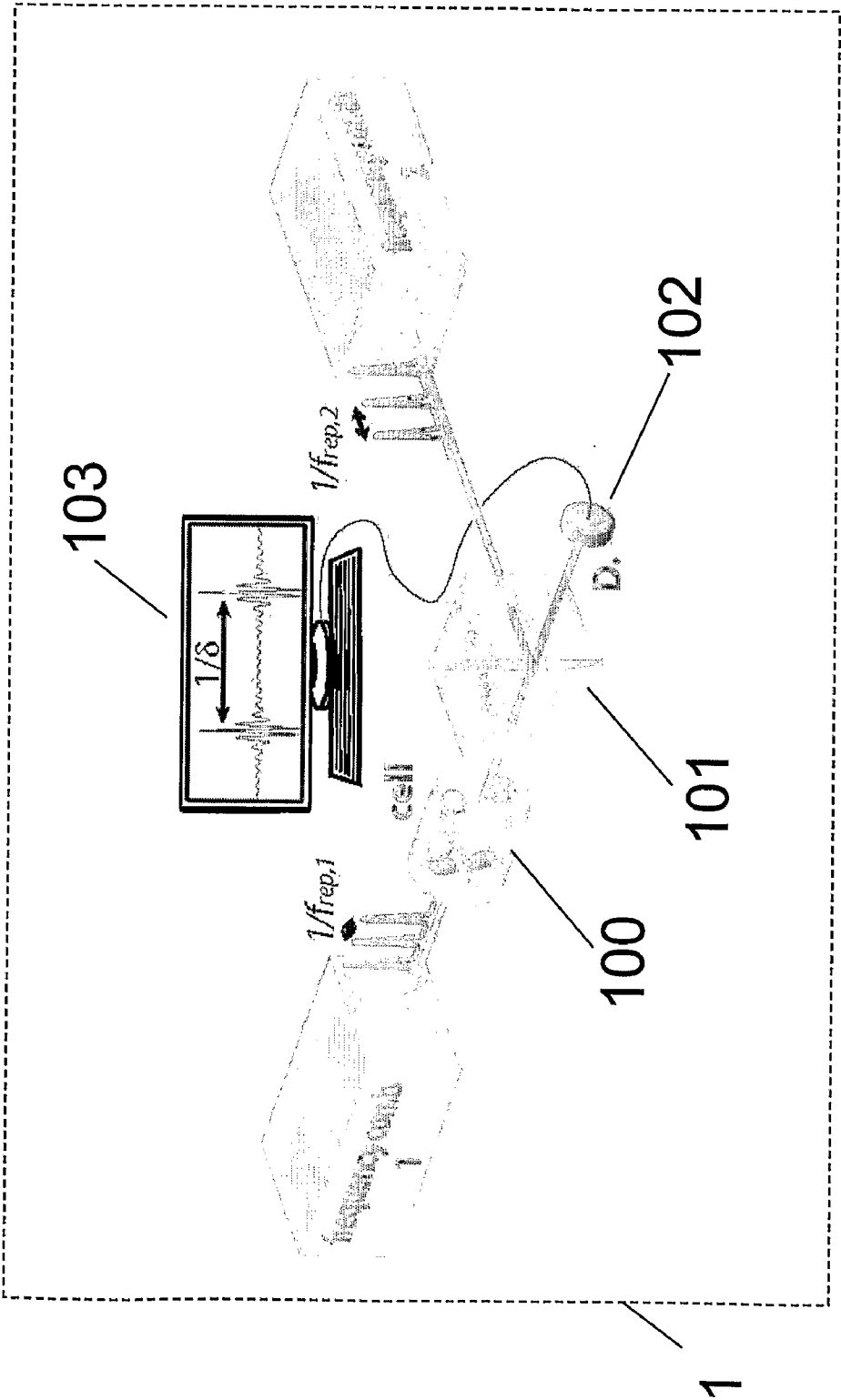


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COMBS AND SYNCHRONISATION SCHEME****Publication Classification**(75) Inventors: **Guy Guelachvili**, Etampes (FR);
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Scientifique-CNRS**, PARIS
CEDEX 16 (FR)(52) **U.S. Cl. 356/454; 356/450; 356/451**(21) Appl. No.: **13/055,909**(22) PCT Filed: **Jul. 20, 2009**(86) PCT No.: **PCT/IB2009/006288**§ 371 (c)(1),
(2), (4) Date: **Jul. 18, 2011****Related U.S. Application Data**(60) Provisional application No. 61/083,620, filed on Jul.
25, 2008.(57) **ABSTRACT**

An embodiment relates to an interferometer comprising: a first frequency comb; a second frequency comb adapted to interact with the first frequency comb in order to produce interferences; means for isolating the beating signal between a subset of frequency components among the frequency components of the two combs. This subset of frequency components may be preferably, but not necessarily, a single line of the first frequency comb and a single line of the second frequency comb; means for monitoring this beating signal and using it as a trigger or as a clock for the acquisition unit device recording the beating interference signal between the entire frequency components of the first and the second frequency combs.





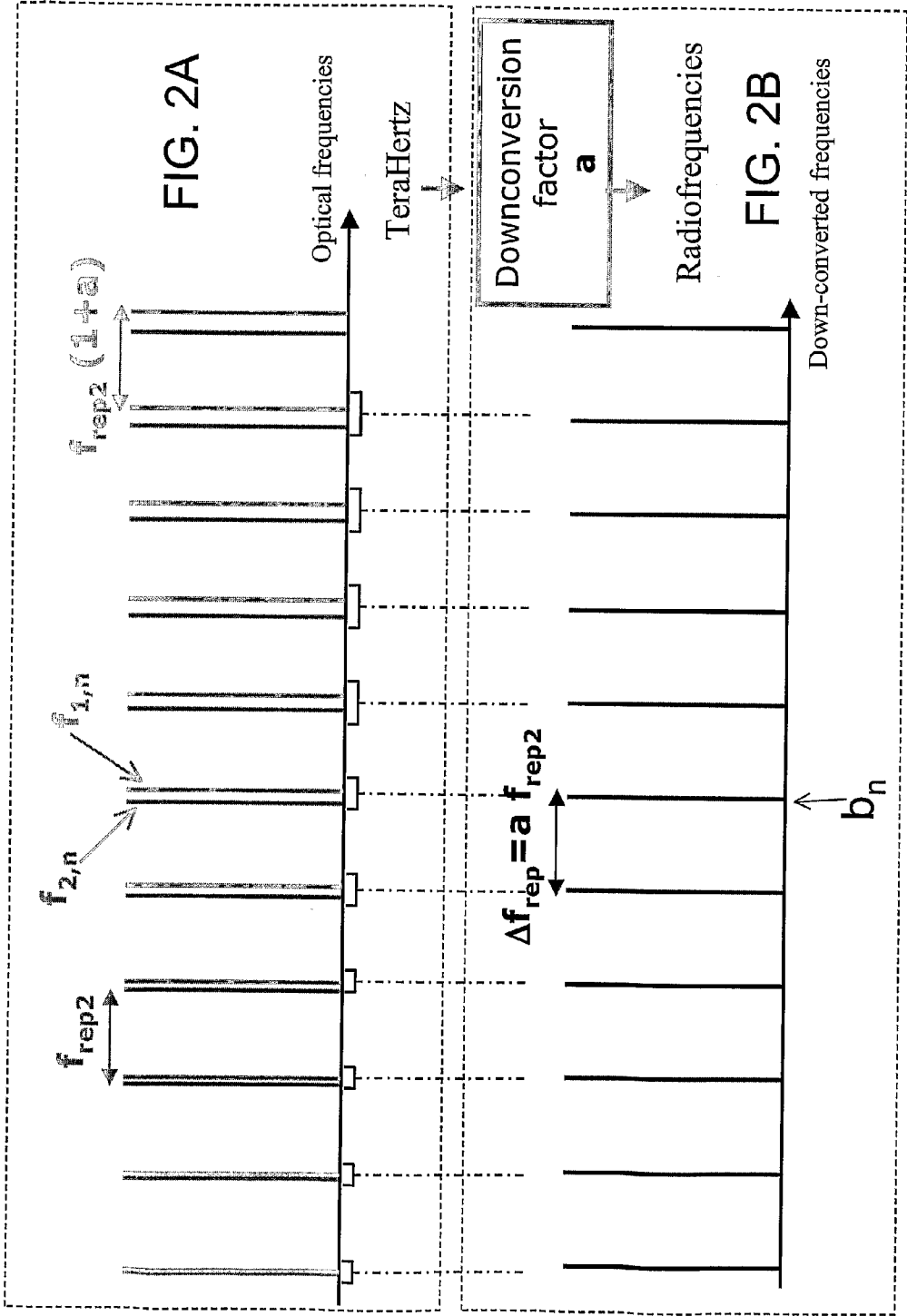


FIG. 2

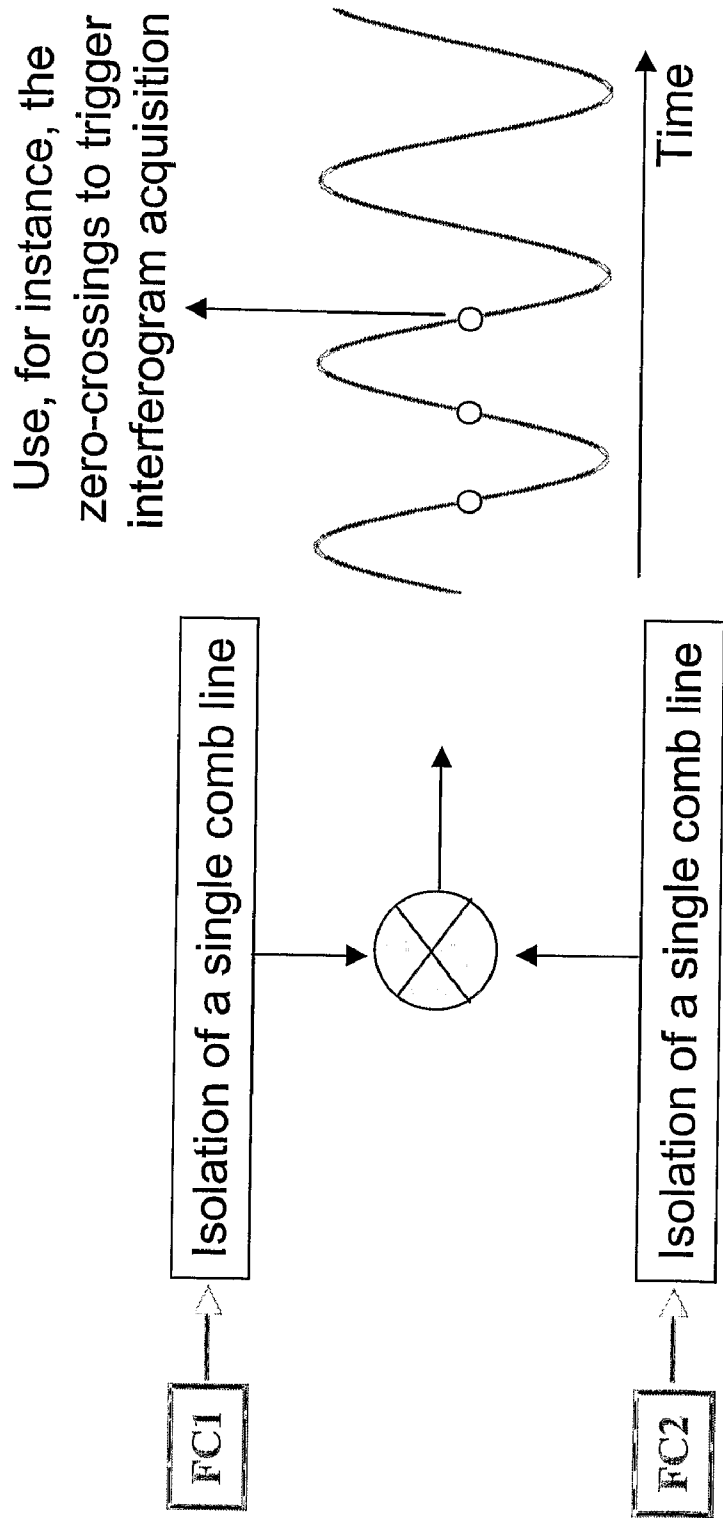


FIG. 3

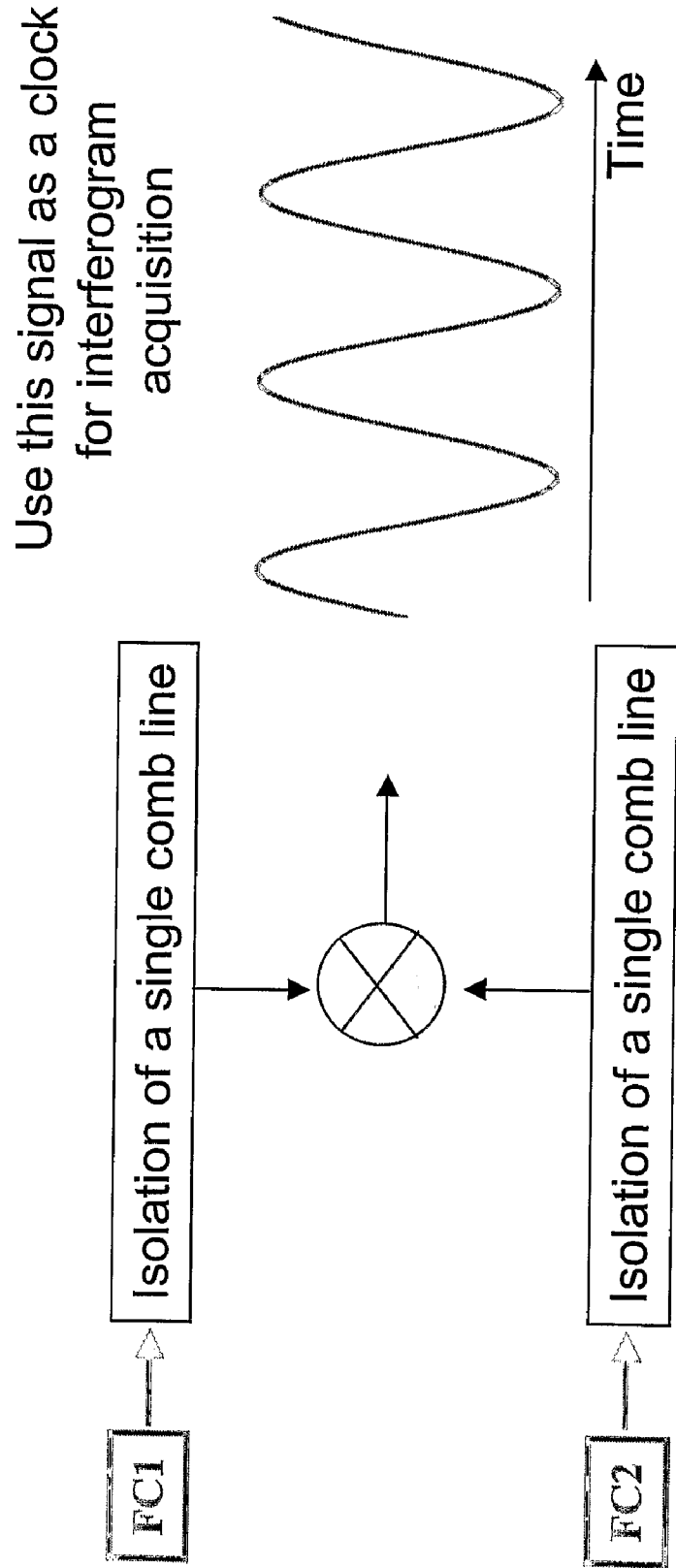


FIG. 4

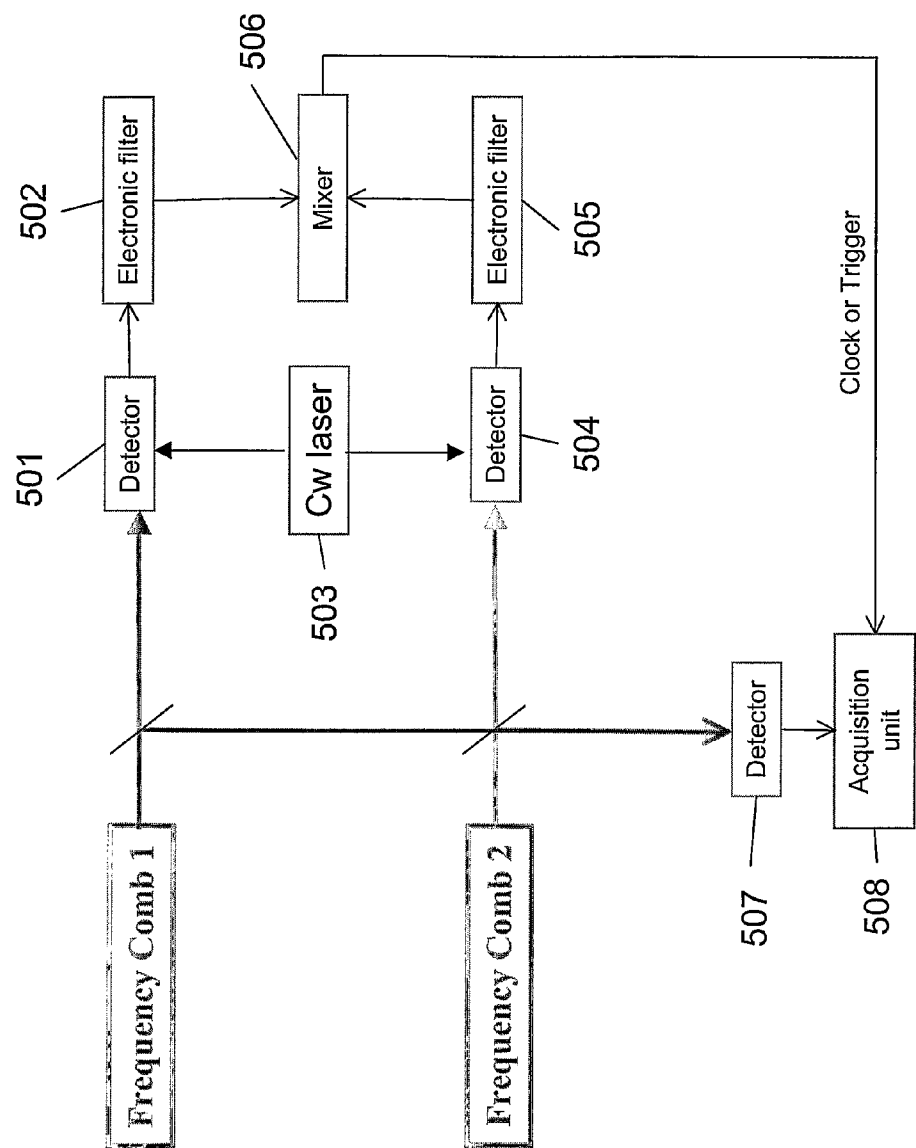


FIG. 5

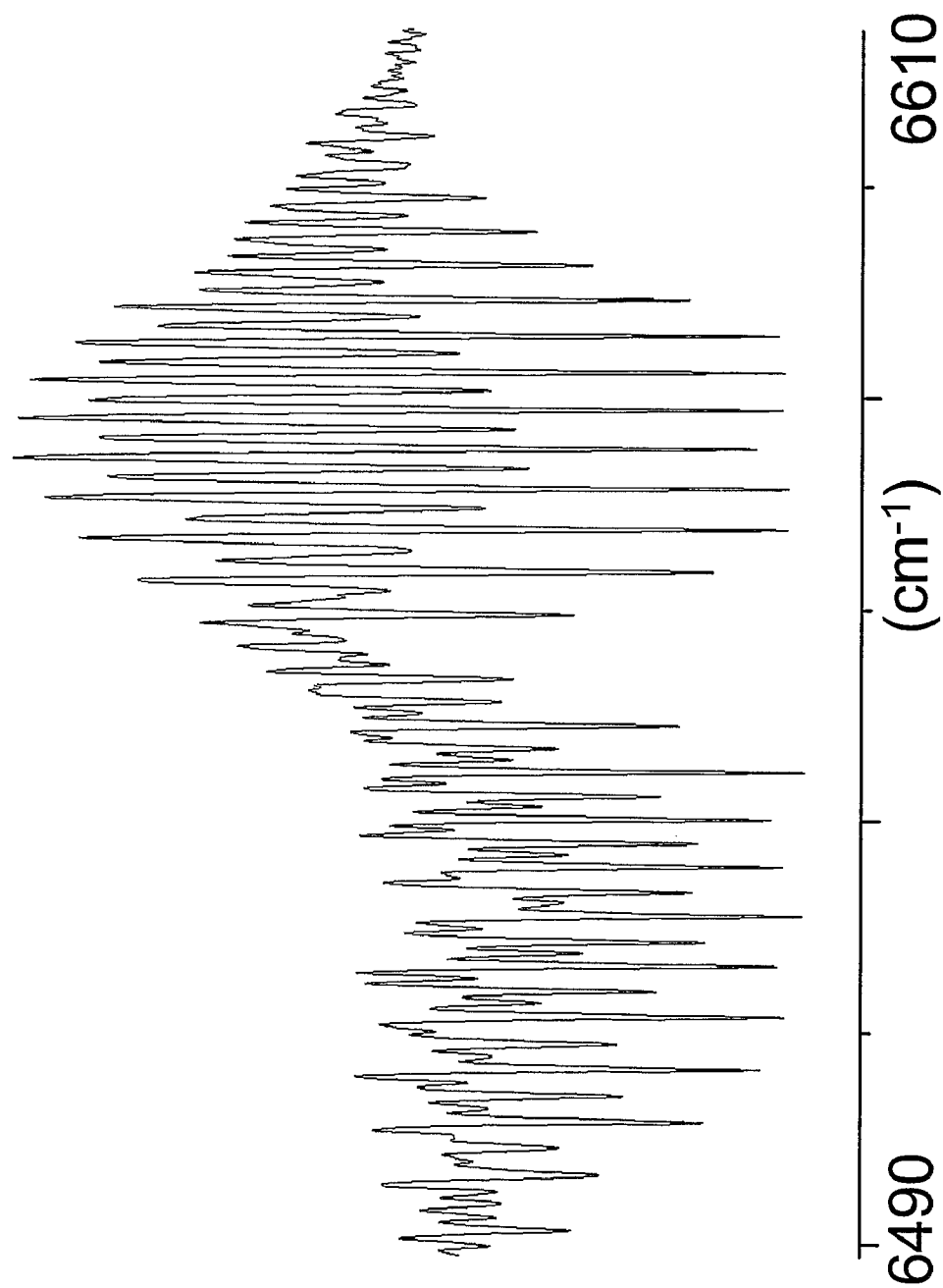


FIG. 6

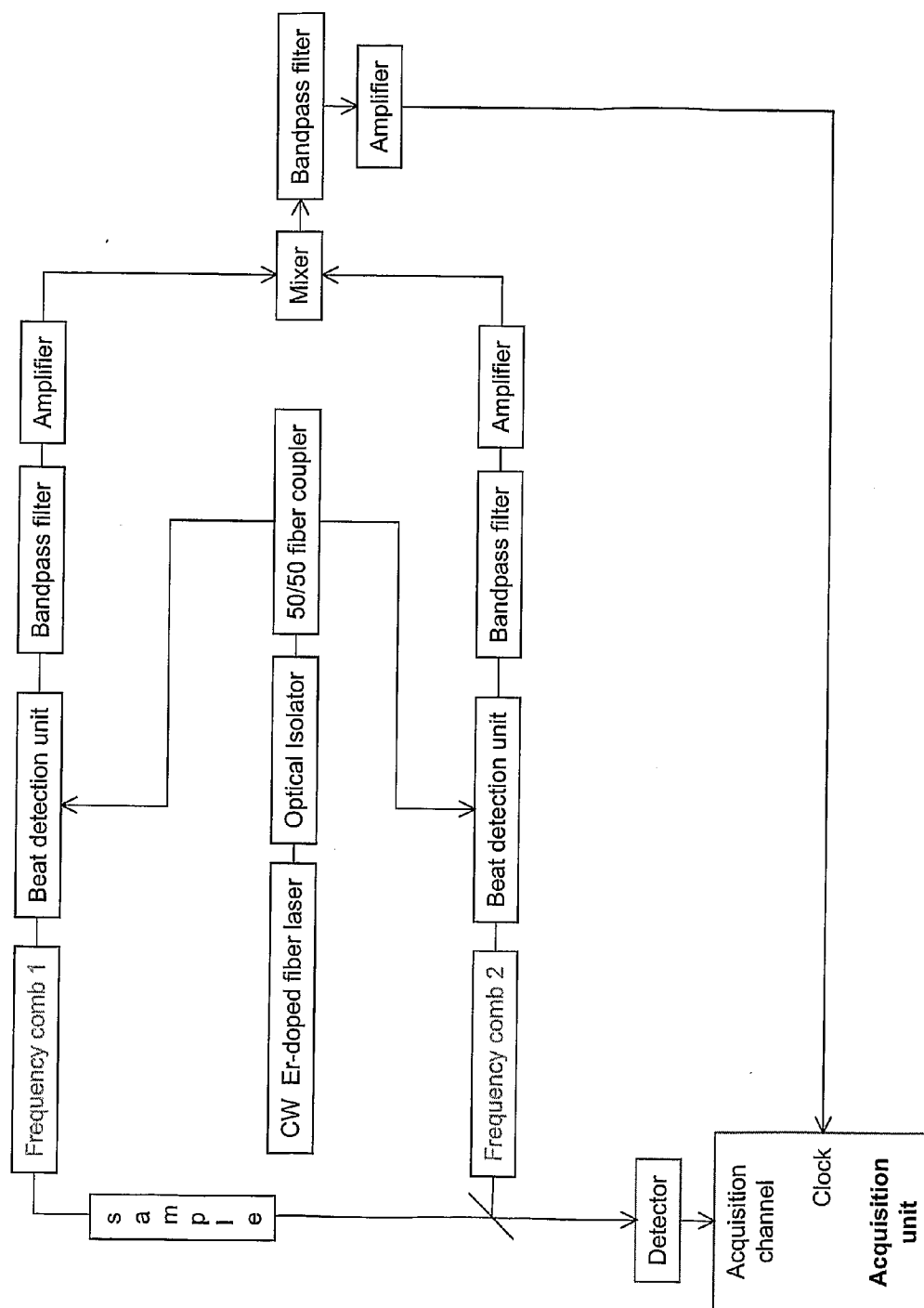


FIG. 7

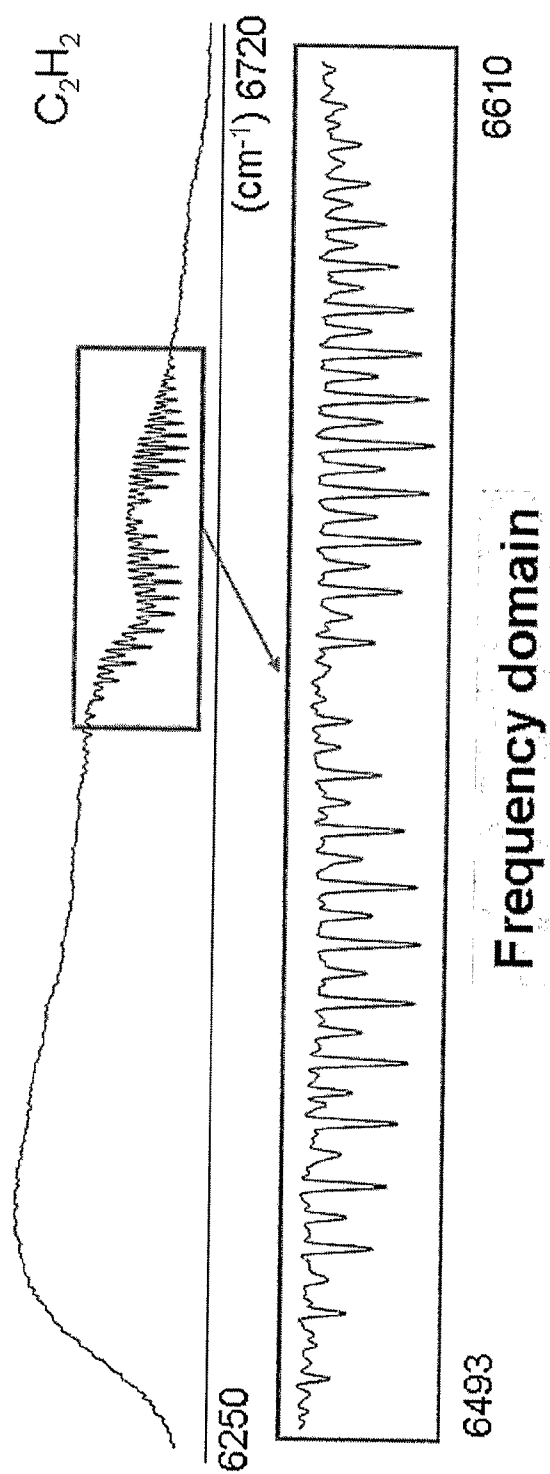
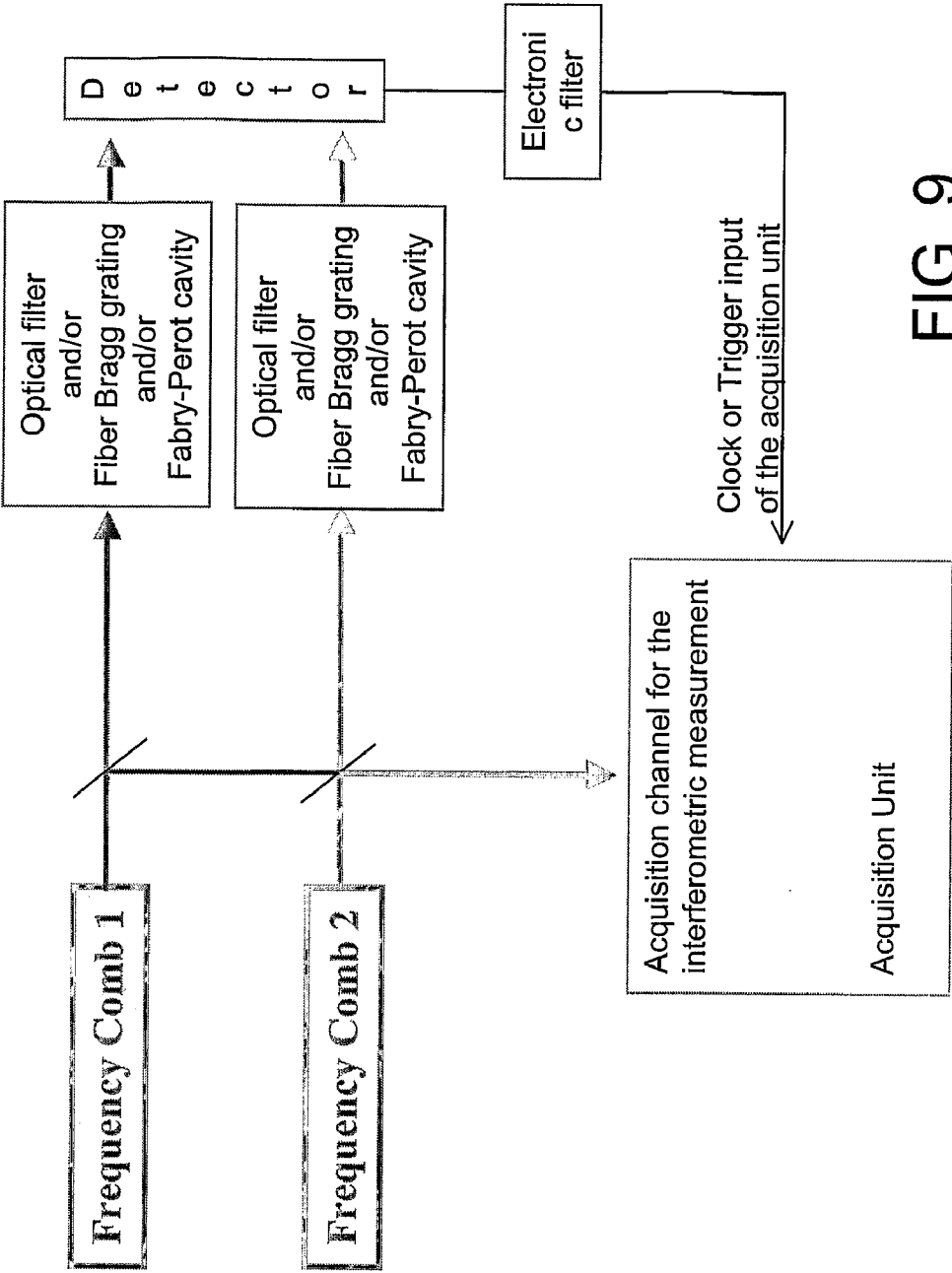


FIG. 8



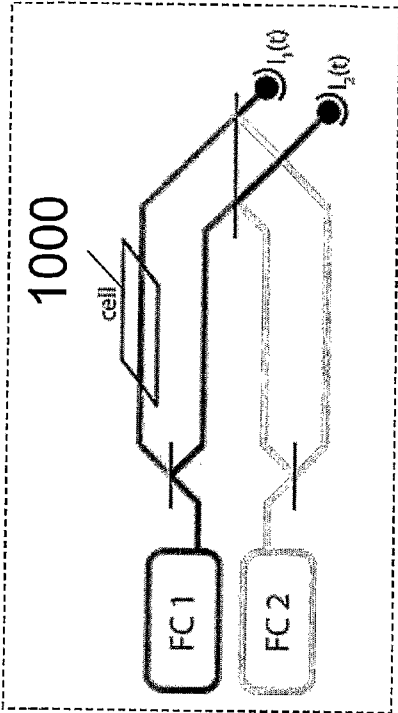


FIG. 10A

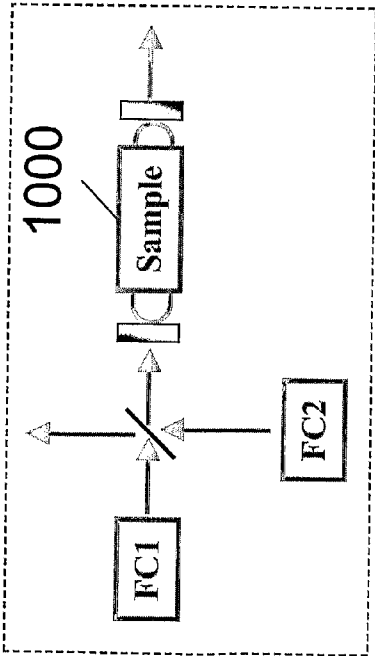


FIG. 10B

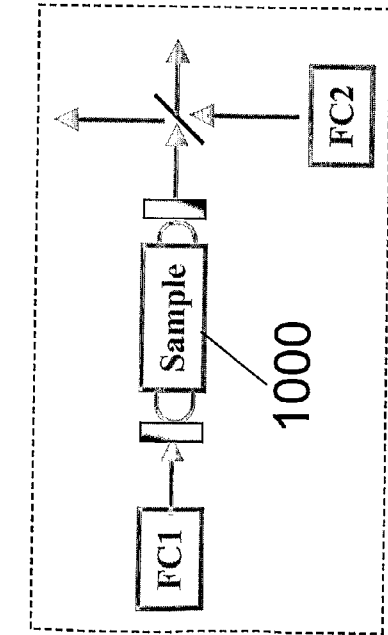


FIG. 10C

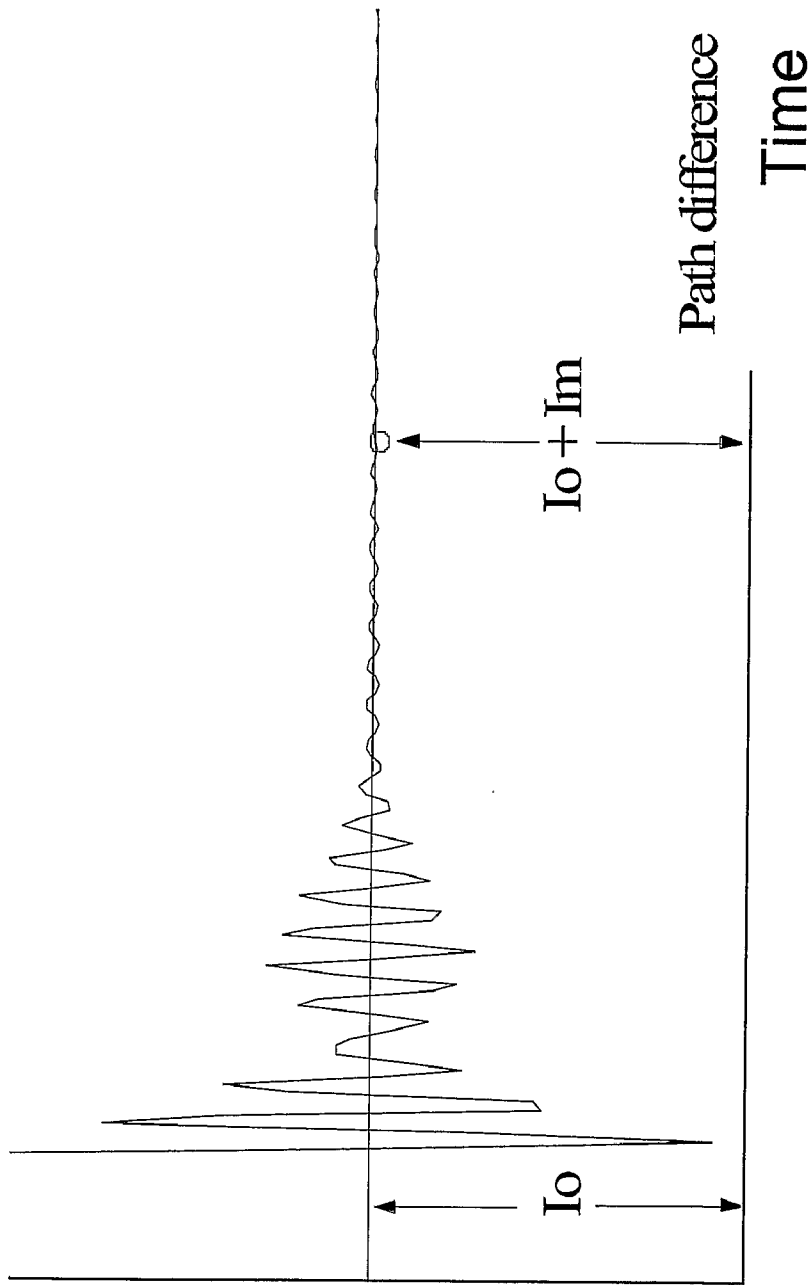


FIG. 11

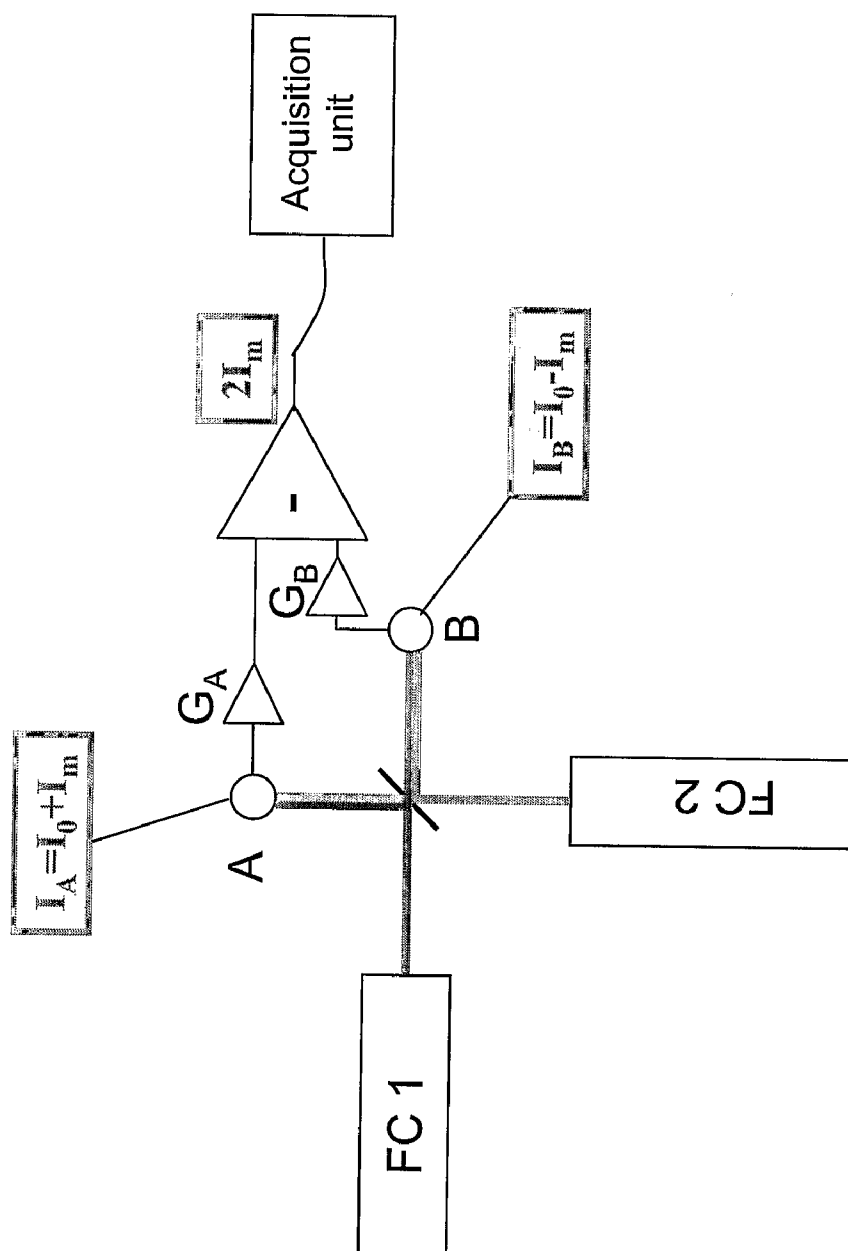


FIG. 12

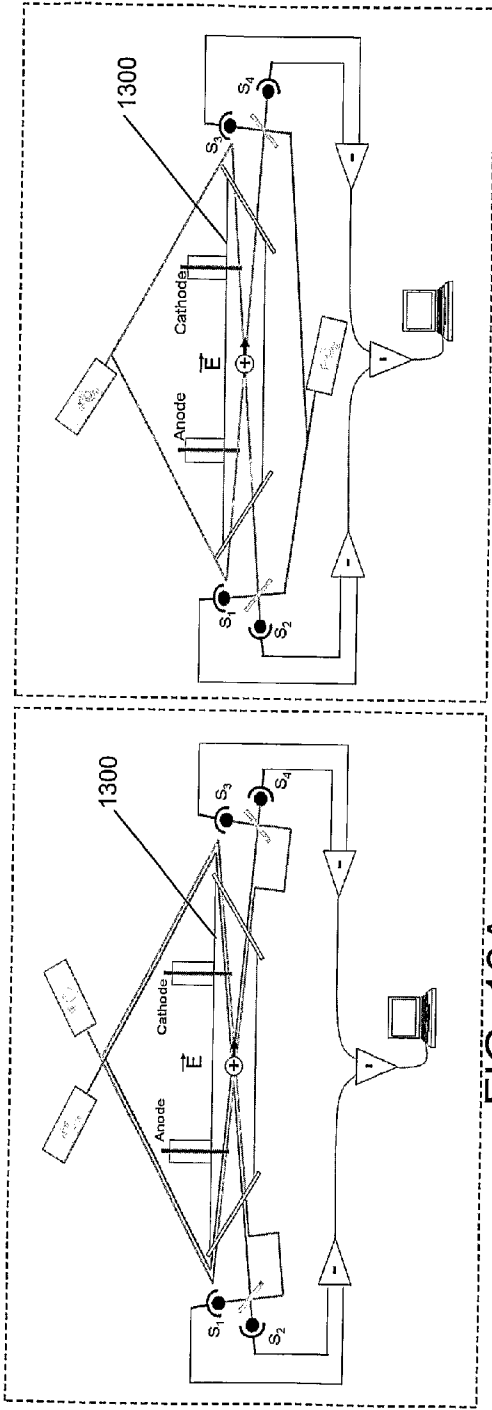


FIG. 13B

FIG. 13A

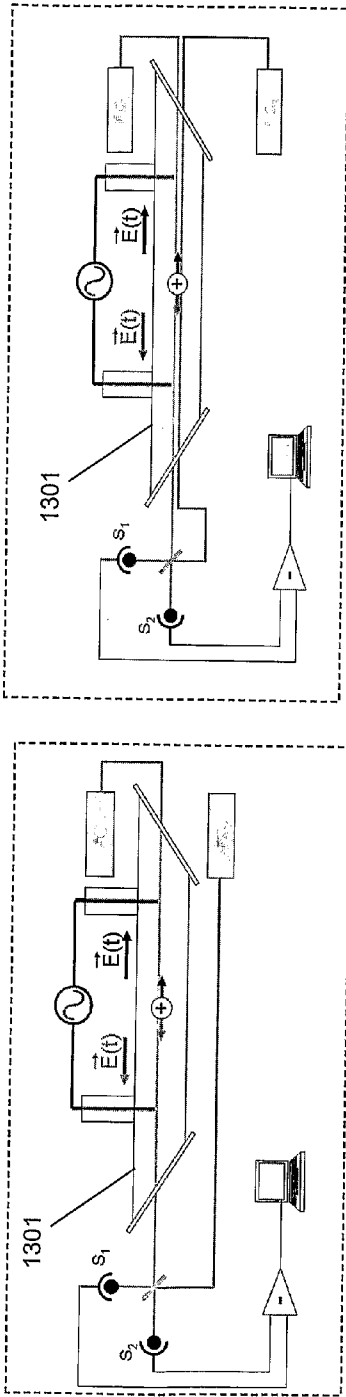


FIG. 13D

FIG. 13

FIG. 13C

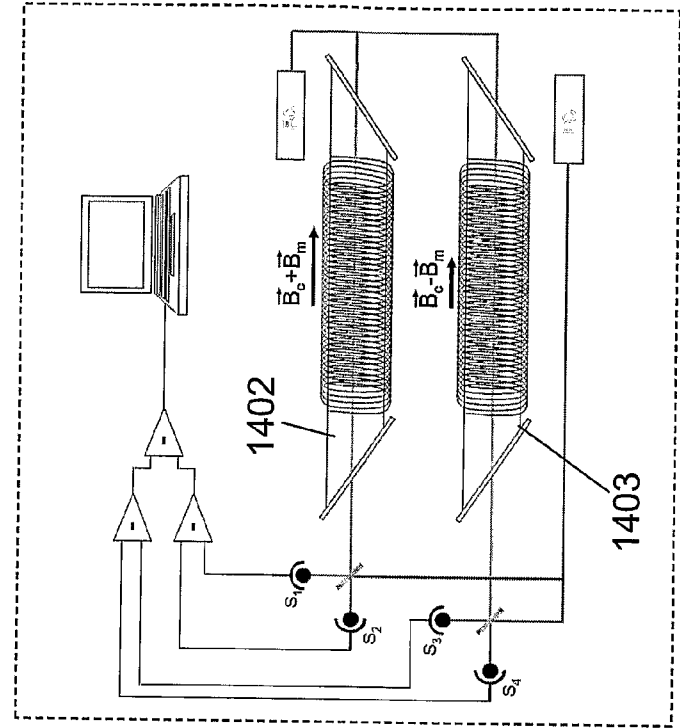


FIG. 14B

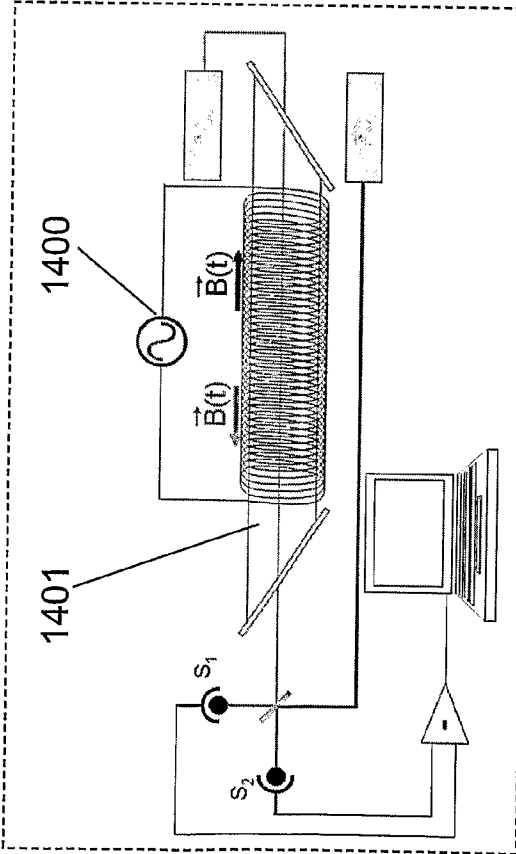
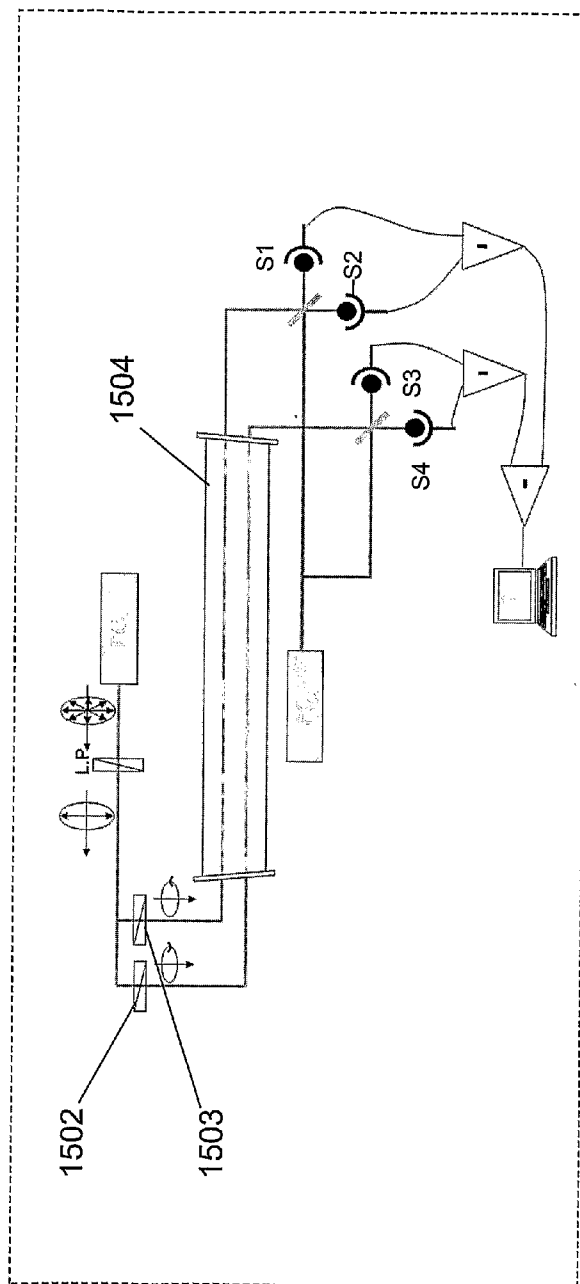
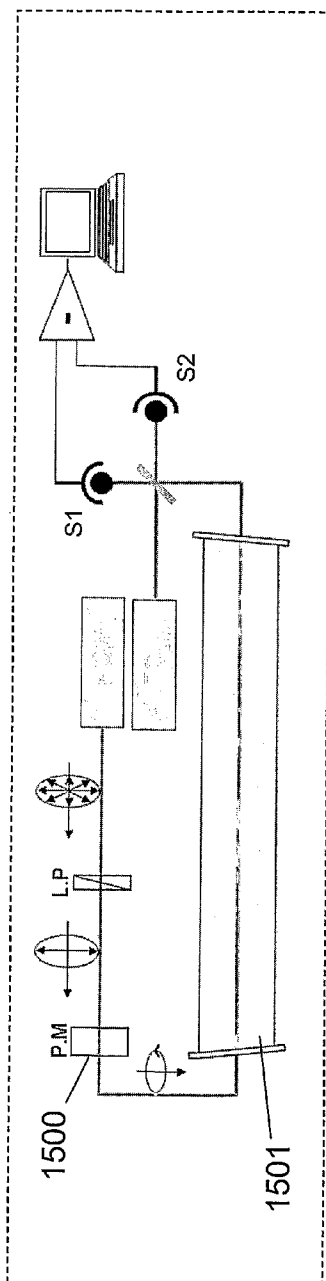


FIG. 14A

FIG. 14



INTERFEROMETER WITH FREQUENCY COMBS AND SYNCHRONISATION SCHEME

PRIORITY CLAIM

[0001] The present application is a national phase application filed pursuant to 35 USC §371 of International Patent Application Serial No. PCT/IB2009/006288, filed Jul. 20, 2009; which further claims the benefit of U.S. Provisional Patent Application Ser. No. 61/083,620 filed Jul. 25, 2008; all of the foregoing applications are incorporated herein by reference in their entireties.

RELATED APPLICATION DATA

[0002] This application is related to U.S. patent application Ser. No. _____, entitled FOURIER TRANSFORM SPECTROMETER WITH A FREQUENCY COMB LIGHT SOURCE (Attorney Docket No.: 2931-002-03) filed _____, and which is incorporated herein by reference in its entirety.

TECHNICAL FIELD

[0003] An embodiment relates to the field of interferometers. It is directed to an interferometer and to a method for measuring interferences in such an interferometer.

SUMMARY

[0004] An embodiment is more particularly directed to an interferometer comprising:

1) a first frequency comb 2) a second frequency comb adapted to interact with the first frequency comb in order to produce interferences.

3) means for isolating the beating signal between a subset of frequency components among the frequency components of the two combs. This subset of frequency components is preferably but not necessarily a single line of the first frequency comb and a single line of the second frequency comb.

4) means for monitoring this beating signal and using it as a trigger or as a clock for the device recording the beating interference signal between the entire frequency components of the first and the second frequency combs.

[0005] An embodiment therefore provides an improved interferometer with a triggering scheme based on the monitoring of the fringes variation. The synchronization method described herein is based on the determination of the beating frequency for a narrowband part of the two combs and the use of its knowledge in a trigger or clock scheme that allows to sample the interferometric signal with a new grid, independently of the combs instabilities versus time.

[0006] According to an embodiment, there is provided an interferometric method for analyzing a sample having spectroscopic absorption and dispersion signatures. The interferometric method comprises:

1) a first frequency comb which probes the sample under study

2) a second frequency comb adapted to interact with the first frequency comb in order to produce interferences. This second comb may or may not be probing the sample.

3) means for isolating the beating signal between a subset of frequency components among the frequency components of the two combs. This subset of frequency components is preferably but not necessarily a single line of the first frequency comb and a single line of the second frequency comb.

4) means for monitoring, as a function of time, this beating signal and using it as a trigger or as a clock for the device

recording the beating interference signal between the entire frequency components of the first and the second frequency combs.

5) means for recording the beating interference signal between the entire frequency components of the first and the second frequency combs using the signal described in 4) as a clock or as a trigger.

[0007] An optical Frequency Comb (FC) is an optical spectrum, which consists of phase-coherent equidistant laser lines. Frequency combs are well known in the art, for example from the patents "Generation of stabilized, ultra-short light pulses and the use thereof for synthesizing optical frequencies" R. Holzwarth, J. Reichert, T. Udem, T. W. Hansch, U.S. Pat. No. 6,785,303, 2004 or "Method and device for producing radio frequency waves", R. Holzwarth, T. Udem, T. W. Hansch, U.S. Pat. No. 7,026,594, 2006 or the publication "Optical frequency metrology" T. Udem, R. Holzwarth, T. W. Hansch Nature 416, 233 (2002), which are incorporated by reference, and are most often used in frequency metrology.

[0008] Interferometers based on two frequency combs are also well known in the art, for example from the publications "Time-Domain mid-infrared frequency comb spectrometer", Keilmann et al, Opt Lett. 29, 1542-1544 (2004), or "Spectrometry with frequency combs", Schiller, S. Opt. Lett. 27, 766-768 (2002), which are incorporated by reference. In these systems, the time-domain interference pattern between the two similar frequency combs with slightly different repetition rates is recorded. The key is to make a down conversion of the optical frequencies characterizing the spectrum of interest so to allow practical measurements. A single detector may be used to record, generally as a function of time, the data called interferogram. This interferogram may be Fourier transformed for spectroscopic purpose's. As this interferometer does not involve moving parts, it may lead to very fast acquisition times.

[0009] If the first comb is expressed as:

$$f_{01} + n f_{rep1}$$

and the second comb is expressed as:

$$f_{02} + n f_{rep2}$$

with f_{01} and f_{02} being the respective offsets of the first and second frequency combs and f_{rep1} and f_{rep2} the respective repetition rates of the first and second frequency combs and n an integer typically ranging between 10^5 and 10^6 , their beating signal is as follows:

$$I(t) = \sum_n A_n \cos((f_{01} - f_{02} + n(f_{rep1} - f_{rep2}))t)$$

and only depends on the differences between combs repetition rates and offsets, which are required to be constant at the time scale of a recording sequence.

[0010] The optical frequency down conversion is obtained from the interference between two similar FC. The two combs are named hereafter by FC1 and FC2. They have slightly different repetition rate frequencies f_{rep1} and f_{rep2} related to each other by

$$f_{rep1} = f_{rep2}(1+a); 0 < a < 1$$

The beating difference frequency b_n between the respective comb components $f_{1,n}$ and $f_{2,n}$, is given by

$$b_n = f_{1,n} - f_{2,n} = a n f_{rep2} + (f_{01} - f_{02})$$

which shows that a is the down conversion factor. In other words, the beat notes between pairs of lines from the two combs, which occur typically in the radio-frequency domain, provide a down-converted image of the optical spectrum. To avoid aliasing, a must be chosen such as $a < f_{rep2}/2$. This is however more than three orders of magnitude higher than the down-conversion factors induced by a moving mirror. Additionally, due to heterodyne detection of coherent signals in the radio-frequency domain, very weak signals may be detected with efficient technical noise rejection and co-additions are not necessary, unlike with an incoherent light source. This approach is multiplex and enables the simultaneous analysis of an extended spectral domain with a single detector. With the benefit of a much reduced measurement time when compared to sequential or multichannel methods, the multiplex approach has the unique advantage of extended spectral domains observation.

[0011] However, these known systems do not take into account the residual instabilities of the two combs. Even when the combs are locked, the parameters f_{rep1} , f_{rep2} , f_{01} , f_{02} encounter, on short time scales (few μ s or longer), slight fluctuations, which may be lower than one part in 10^6 . Because of these residual instabilities, the difference between the repetition rates of the two combs may vary, thus leading to errors in the interference pattern sampling. Such interferometers indeed require to keep a f_{rep2} and $(f_{01} - f_{02})$ constant during the time of a measurement with a very high level of constraint on the stability of the repetition rates of both frequency combs and on the stability of the carrier-envelope offset frequencies, when applicable. In other words, the relative coherence between the two independent comb sources must be forced within the time of a measurement, which may be very short. This constraint is stringent and prevents the practical implementation of the method with simple means. Using state-of-the-art reference cavities to lock the combs, as shown in I. Coddington, W. C. Swann and N. R. Newbury, "Coherent multiheterodyne spectroscopy using stabilized optical frequency combs", *Physical Review Letters* 100, 013902 (2008), which is incorporated by reference, excellent results may be obtained, but such systems require an extensive knowledge of the most modern and expensive tools of frequency metrology and cannot be used in a standard spectroscopy laboratory, in the industry or in field measurements. This stability constraint on the difference in repetition rates and carrier-envelope offsets of the two combs is different from those of Fourier Transform Spectroscopy (FTS) with a Michelson interferometer, where the Doppler-shifted replica of a comb source is produced by appropriate control of the moving mirror of the interferometer, independently of the comb stability. It also differs from the use that is made of frequency combs in metrology, where the phase-coherence of the comb with a radio-frequency or optical clock is needed on long time scales (several seconds).

[0012] An embodiment is an improved interferometer, based on two frequency combs, accounting for the instabilities of the combs. In other words, an embodiment of a proposed device and method enable measurement of the interferences between two frequency combs where at least one comb has its line positions which are varying as a function of time. An embodiment allows monitoring the variations in the

mapping between the two frequency combs in order to overcome the accuracy and measurement time limitations of the prior art. Constraints on frequency comb stability are consequently reduced.

[0013] According to an embodiment, the interferometer comprises:

[0014] a first frequency comb generator;

[0015] a second frequency comb generator adapted to interact with the first frequency comb signal in order to produce interferences; and

[0016] a system serving as a clock synchronized on the combs instabilities or enabling to trigger the sampling of the interference process.

[0017] An embodiment also relates to Frequency Comb Fourier Transform Spectroscopy (FC-FTS) and is directed to a spectroscopic device for measuring the spectrum of a sample, at least one of said first frequency comb generator and said second frequency comb generator being adapted to interact with the sample.

BRIEF DESCRIPTION OF THE DRAWINGS

[0018] One or more embodiments will be set forth in detail with reference to the drawings in which:

[0019] FIG. 1 shows a schematic view of a frequency comb interferometer adapted in an absorption spectrometer for Fourier transform spectroscopy;

[0020] FIG. 2 comprises FIG. 2A and FIG. 2B which are plots respectively showing an example of two optical frequency comb spectra and the corresponding beating spectrum of an example frequency comb sources;

[0021] FIG. 3 shows a conceptual view of a system enabling to trigger the data acquisition according to an embodiment;

[0022] FIG. 4 shows a conceptual view of a system serving as a clock for the data acquisition according to an embodiment;

[0023] FIG. 5 shows a detailed embodiment of an interferometer, where the spectral filtering of the combs synchronization beating signal is performed with the aid of a cw laser;

[0024] FIG. 6 shows a distorted spectrum that is typically obtained without applying an embodiment;

[0025] FIG. 7 shows a block diagram illustrating an interferometric system using the synchronization method according to an embodiment, particularly according to an embodiment of the principle of FIG. 4;

[0026] FIG. 8 shows a distortion-free spectrum recorded using the interferometric system of FIG. 7;

[0027] FIG. 9 shows a detailed embodiment of an interferometer, where the spectral filtering of the combs synchronization beating signal is performed with passive filters;

[0028] FIG. 10 comprises FIG. 10A, FIG. 10B and FIG. 10C which are plots respectively showing an example of the optical layout for using the interferometer of an embodiment as a spectrometer probing absorption and/or dispersion of a sample.

[0029] FIG. 11 shows an interferogram obtained with an interferometer.

[0030] FIG. 12 illustrates an embodiment of a interferometer with differential measurements;

[0031] FIG. 13 comprises FIG. 13A, FIG. 13B, FIG. 13C and FIG. 13D and illustrates an embodiment for velocity modulation;

[0032] FIG. 14 illustrates an embodiment for Zeeman spectroscopy;

[0033] FIG. 15 illustrates an embodiment with polarization modulation.

DETAILED DESCRIPTION

[0034] One or more embodiments will be set forth in detail with reference to the drawings, in which like reference numeral refers to like elements throughout. The herein presented interferometer with synchronization technique is best understood considering the illustrative example of frequency comb Fourier Transform Spectroscopy and all related instrumental methodologies (hyperspectral imaging, microscopy, vibrational circular dichroism, attenuated total reflection, Zeeman modulation, velocity modulation, selective detection techniques, time-resolved spectroscopy . . .), but it is noted that the interferometer with synchronization technique may also be used beyond the herein described applications. For example, the herein described interferometer with synchronization technique may be adapted to be used in Optical Coherence Tomography (OCT), interferometric length measurements, Light Detection And Ranging (LIDAR), reflectometry, linear optical sampling, cross-correlation measurements between electrical fields. Other interferometric applications, based on wavefront or amplitude recombination, not specifically described herein may also benefit from the synchronization technique.

[0035] As known in the art, a frequency comb is a spectral source made of laser monochromatic emission lines regularly spaced over a spectral range. Most often, femtosecond lasers are used, leading to a femtosecond frequency comb (FFC). Alternatively, the combs may be produced by four-wave mixing in appropriate material, including toroidal microresonators or optical fibers. The combs may be also produced by a phase-modulated electro-optic device.

[0036] Summarizing, when using a mode-locked laser, ultra short pulses are periodically emitted by the mode-locked laser with a time period $T=1/v_g$ where l is the length of the laser cavity and v_g is the net group velocity. Due to dispersion in the cavity, the group and phase velocities differ, resulting in a phase shift of the carrier with respect to the peak of the envelope of each pulse. In the frequency domain the spectrum, Fourier transform of the train of periodic pulses, is made of a comb of laser modes, which are separated by the pulse repetition frequency f_{rep} . The modes frequencies obey the relation: $f_n = nf_{rep} + f_0$, with n integer and f_0 due to carrier-envelope pulse-to-pulse phase-shift. Presently the output spectrum of the comb may span more than one octave and the best level of stabilization of f_{rep} and f_0 is such that there is no deviation from a perfect grid larger than one part in 10^{19} .

[0037] In other words, a frequency comb is an optical spectrum which consists of equidistant lines. The generation of a frequency comb from a mode-locked laser requires that the periodicity applies not only to the pulse envelopes, but to the whole electric field of the pulses, including their optical phase, apart from a constant phase. Coherence between the pulses is therefore required.

[0038] As shown in FIG. 1 and known per se, a specific layout of an interferometer 1 comprises a frequency comb "Frequency Comb 1" and a frequency comb "Frequency Comb 2". "Frequency Comb 1" probes a sample contained in a cell 100. The interferences that are generated from the recombination on a beam-splitter 101 of the frequency combs "Frequency Comb 1" and "Frequency Comb 1" are measured by a detector 102 and an interferogram is measured at a computer 103. The beat notes between pairs of lines from two

combs having slightly different repetition rates, which occur in the radio-frequency domain, provide a down-converted image of the optical spectrum.

[0039] Note that the interferometer does not necessarily probe a sample 100, and that the sample 100, if present, may interact with both combs. Wavefront recombination instead of amplitude recombination may also be used. In such case, the beam-splitter 101 is not needed.

[0040] According to an embodiment, the frequency combs FC1 and FC2 have slightly different repetition rate frequencies f_{rep1} and f_{rep2} related to each other by

$$f_{rep1} = f_{rep2}(1+a); 0 < a < 1$$

[0041] According to an embodiment, frequency combs FC1 and FC2 may be generated by femtosecond mode-locked lasers with repetition rates and carrier-envelope offset stabilized or free-running. The mode-locked laser may be broadened by a non-linear optical fiber and it may be coupled to a frequency conversion system such as difference/sum frequency generation, harmonic generation and/or parametric interaction. Frequency combs FC1 and FC2 may also be generated by four-wave mixing effect or by an electro-optic modulator seeded by a continuous wave laser.

[0042] The spectrometer according to an embodiment comprises an interferometer 1 on FIG. 1 as described above. The spectrum of each laser (as shown in FIG. 2A), numbered 1 or 2, is a comb of laser modes $f_{n,i}$ separated by the pulse repetition frequency $f_{rep,i}$:

$$f_{n,i} = f_{0,i} + nf_{rep,i}$$

where $i=1$ or 2 , integer n typically in the range of 10^5 to 10^6 , and $f_{0,i}$ is the carrier-envelope offset frequency, induced by the difference between the group and phase velocities of the pulses in the laser cavities, in the case of a frequency comb based on a mode-locked laser. Repetition frequencies are generally in the radio-frequency domain (in the present example they are close to 100 MHz with $f_{rep,1} - f_{rep,2}$ ranging from 10 Hz to 20 kHz, depending on the free spectral range). One laser probes (FIG. 1) the absorbing sample and both beams are combined on a beam-mixer. Their beating signal I , recorded on a fast photodetector, is amplified and sampled using a high-resolution digitizer on a personal computer.

[0043] The beating difference frequency b_n between the respective comb components $f_{1,n}$ and $f_{2,n}$, as shown in FIG. 2B, is given by

$$b_n = f_{1,n} - f_{2,n} = nf_{rep2} + (f_{01} - f_{02})$$

which shows that a is the down conversion factor.

[0044] The interferogram which is recorded on the photodetector may therefore be written as:

$$I(t) = \sum_n A_n \cos((f_{0,1} - f_{0,2} + n(f_{rep,1} - f_{rep,2}))t)$$

where A_n is the product of the amplitude of the electric fields of the two lasers, attenuated by sample absorption. Similarly to what happens with a Michelson interferometer, the frequencies of the optical spectrum $f_{n,i} = f_{0,i} + nf_{rep,i}$ are down-converted to $f_{0,1} - f_{0,2} + n(f_{rep,1} - f_{rep,2})$. However, thanks to the absence of moving parts, the down-conversion, limited by aliasing, may lead to the radio-frequency domain between 0 and $f_{rep,i}/2$, and not to the audio-range like in Michelson-based FTS. This may result in a drastic reduction of the

recording time. The signal $I(t)$ is then Fourier-transformed to reveal the spectrum; the longer the measurement time, the better the optical resolution. The interferometric signal exhibits a periodic succession of huge bursts occurring every $1/\delta = 1/(f_{rep,1} - f_{rep,2})$ when femtosecond pulses from the two lasers coincide. Fourier-transforming a temporal sequence including more than two such bursts resolves the comb lines. As the combs parameters ($f_{rep,1}$, $f_{rep,2}$, f_{01} , f_{02}) may be straightforwardly measured with radio-frequency counters, the optical absolute frequency scale may be easily retrieved afterwards. It may also be possible to use known molecular or atomic lines present in the spectrum to rescale the optical frequency axis.

[0045] During the time of the measurement of the beating signals (which may be as short as a few microseconds), at least one parameter of one of the combs ($f_{rep,1}$, $f_{rep,2}$, f_{01} , f_{02}) may be slightly fluctuating as a function of time. In other words, b_n is a function of time. FIG. 2B therefore only provides therefore an instantaneous image of the down-converted spectrum. In practice, the fluctuations of the repetition rates ($f_{rep,1}$, $f_{rep,2}$) of the combs may be much more troublesome than the fluctuations of the carrier-envelope offset frequencies (f_{01} , f_{02}) because the fluctuations on the repetition rates are amplified by the harmonic index n . Therefore, sampling the interferogram at constant time intervals may lead to distorted information.

[0046] An embodiment provides a simple way of sampling the interferogram in a manner that is synchronous to the most troublesome comb fluctuations. An embodiment of a new sampling grid is provided. The idea consists in isolating the beating signal between two individual lines from the interfering combs and to use the resulting signal, which is a sine wave if the line positions of the combs are fluctuating in a negligible manner, as a trigger (FIG. 3 in the special example embodiment where the zero crossings of the synchronization beating signals are used as a trigger) or as a clock (FIG. 4) for the sampling process of the interferometric signal between the two entire combs. If the comb lines are not isolated well enough, i.e. if the synchronization beating signal consists in the beat note of more than one couple of lines, the synchronization process still works but maybe within a more limited time sequence, i.e. with a more limited resolution for Fourier transform spectroscopy.

[0047] According to an embodiment shown in FIG. 5, the synchronization device consists first in isolating a single line from each comb. A part of the optical signal delivered by "frequency comb 1" is sent to a fast photo-detector 501 with the aid of a beam-splitter, while most of the signal is used for the interferometric measurements (some examples are described hereafter). The beam of a continuous-wave laser 503 falls also on the same detector 501. The continuous wave laser 503 emits at the frequency f_{cw} in the same spectral region as the combs. It may be locked to a reference or free-running and its spectral purity, as well as its frequency stability, mainly determines the maximum sampling duration of the interferometric measurement. The beating signal between the frequency comb 1 and the cw laser is a comb of down-converted modes lying partly in the radio-frequency and microwave domains. With proper choice of the photodetector 501 bandwidth and further electronic filtering 502, the beating signal between a single comb line and the cw laser, which may be written $(n f_{rep,1} + f_{01} - f_{cw})$, may be isolated. The same procedure is applied to the frequency comb 2, with photodetector 504 and electronic filter 505, in order to isolate the

electric beating signal of a single line of the frequency comb 2 with the same cw laser 503, $(m f_{rep,2} + f_{02} - f_{cw})$. The two beating signals, obtained as described herein, are mixed with an electronic mixer 506 in order to suppress the contribution of the cw laser and to provide the clock beating signal between the two comb lines $(n f_{rep,1} + f_{01} - f_{rep,2} - f_{02})$. n and m may be chosen so that $(n f_{rep,1} + f_{01} - m f_{rep,2} - f_{02})$ is greater than the sampling frequency imposed by the Nyquist criterion which is $\text{Max}(f_{rep,1}, f_{rep,2})$. Special markers (for instance but not mandatorily the zero-crossings) of this sine wave which has a period varying with time are used to trigger the data acquisition of the interferometric signal on an acquisition unit 508. The interferometric signal comes from the beating signal between the entire (or filtered parts) spectra of frequency combs 1 and 2. It is measured on a fast photodetector 507 and monitored on the acquisition unit 508. Electronic filters and amplifiers may be inserted between 507 and 508 to improve the signal to noise ratio. Alternatively, as shown also in FIG. 5, this $(n f_{rep,1} + f_{01} - f_{rep,2} - f_{02})$ frequency signal may serve as a clock to the acquisition unit 508. The acquisition unit 508 may be for instance a high speed digitizer onboard a personal computer. The data points are then for instance stored on the hard drive of the computer or are coded on a field-programmable gate array or are directly Fourier-transformed before storage.

[0048] According to a specific example of an embodiment aimed at Fourier spectroscopy of an absorbing sample, two Er-doped fiber lasers emit pulses at a repetition rate around 100 MHz and 20 mW output power. The difference between the repetition rate of the two combs is set to a value equal to 150 Hz. The repetition rate of each comb is stabilized to a quartz oscillator and the carrier-envelope offset of each comb is not stabilized. The two combs are recombined with a 50-50 beam-mixer and the two outputs of the interferometer are measured to improved measurement dynamics, as explained below. The signal is amplified and digitized with home-made electronics and acquisition systems. FIG. 6 shows the spectrum which is obtained without using an embodiment. The lines are strongly distorted by a phase error. A free-running continuous wave Er-doped fiber laser emitting at 1557.4 nm beats with each of the frequency combs, following to the principle displayed in FIG. 5, according to the detailed scheme shown in FIG. 7. In FIG. 7, the cw Er-doped fiber laser is optically isolated and its beam is split into two parts with the aid of a 50/50 fiber coupler. A part of the beam at the output of frequency comb 1 probes a sample consisting of acetylene in gas phase. It beats on a detector with a part of the beam at the output of frequency comb 2 and the beating signal is monitored using an acquisition unit, which may consist of a digitizer and proper electronics for filtering and amplifying the interferometric signal. Another part of the beam of frequency comb 1 is directed into a beat detection unit where its lines beat with the beam from the cw laser. A beat detection unit is a means to measure the radio-frequency or microwave beating signal between two optical beams. It consists in a fast photodetector and for instance optics to balance, polarize, filter one or both optical beams and electronics to filter the produced electrical signal. At the output of this beat detection unit, an electrical signal at a frequency of 42 MHz corresponding to the beat note between a single line of frequency comb 1 and the cw laser is produced, filtered and amplified and directed towards a mixer. In a similar manner, the beating signal between one line of frequency comb 2 and the cw laser is produced at a frequency of 118 MHz and is directed

towards the second input of the mixer. A beating signal between two individual lines from each comb is consequently delivered at the output of the mixer at a frequency of 76 MHz. It is filtered and amplified and used as a clock for the sampling of the interferometric signal using the acquisition unit. FIG. 8 shows the spectrum which was measured when using this 76 MHz signal as a clock for the 16-bit 180 MSamples/s digitizer used as an acquisition unit. Little or no distortion is noticeable on the spectrum of FIG. 8.

[0049] Alternative configurations may be utilized to produce the clock beating signal between the two combs. For instance, an embodiment may be relying on optical filtering, instead of using a cw laser in the synchronization scheme. As shown in FIG. 9, each comb may be frequency filtered with optical filters or fiber Bragg gratings or Fabry-Perot cavities or any combination of these items. The two filtered combs beat on a fast photodiode. After proper electronic filtering, the clock beating signal ($n f_{rep1} + f_{0,1} - m f_{rep2} - f_{0,2}$) is used as a clock or a trigger for the acquisition unit.

[0050] Further embodiments are described below. These embodiments may be used in combination with the aforementioned embodiments of the interferometer or may be used as such.

[0051] The aforementioned embodiments may be used according to a specific example embodiment aimed at Frequency Comb Fourier transform spectroscopy of an absorbing sample. Several optical configurations may be implemented, as illustrated in FIG. 10. In FIG. 10A, only frequency comb FC1 interacts with the sample 1000. The sample 1000 may be directly probed or it may be inserted inside a single pass cell, or inside a multipass cell, or inside a high finesse cavity that is resonant to the comb FC1. The two beams from combs FC1 and FC2 are recombined on a beam combiner. One or both outputs of the beam combiner may be used to measure the interferometric signal on fast photodetectors. The detectors may consist of simple photodetectors (photodiode, bolometer, antenna) or non-linear frequency converters, e.g. frequency doubling devices, coupled to photodetectors. The detectors may be microphones. In FIG. 10B, both frequency combs FC1 and FC2 interact with the sample 1000. For simultaneous absorption and dispersion measurements, the optical layout of the interferometer may be implemented as displayed in FIG. 10C. Two frequency combs, FC1 and FC2, have slightly different line spacing. One of these combs, FC1, is transmitted through the cell and heterodyned against the second comb, yielding a down-converted radio-frequency comb containing information on the absorption and dispersion experienced by each line of the comb FC1. Appropriate optics transmits the combs beams to two interferometers. The interferometer probing the sample contains the cell and leads to the sample interferogram $I_1(t)$ while the interferometer serving as a reference without the sample provides the normalization interferogram ($I_2(t)$). The two interferograms (with and without sample) are measured simultaneously and computed with a complex Fourier transform. The phase and module of the complex ratio of these two spectra give respectively the phase-shift and attenuation imposed on the detected radiation by the sample, providing access to the full characterization of the optical constants of the medium, its refractive index and absorption coefficient. Alternatively, the difference between the two interferograms $I_1(t)$ and $I_2(t)$ may be measured directly with an appropriate differential amplifier before digitization. In such case, only the interferogram of the

spectral signatures of the sample are measured, with an improved signal to noise ratio.

[0052] The frequency combs interact with a sample and experience attenuation and phase-shift due to absorption and dispersion. This interaction may be written as $\exp(-\delta\{\omega\} - i\phi(\omega))$ where δ is the amplitude attenuation and ϕ is the phase shift. The following convention is adopted: $\delta_{i,n}$ and $\phi_{i,n}$ denotes the respective components experienced by the comb i ($i=1$ or 2) at the frequency $f_{0,i} + n f_{rep,i}$. As illustrated in FIG. 10A, only comb FC1 interacts with the sample 1000. The expression of the interferogram I contains the following interesting terms:

$$I(t) \subset \sum_n \left[\begin{array}{l} e^{-\delta_{1,n}} \left[\begin{array}{l} \cos(2\pi((f_{0,1} - f_{0,2}) + n f_{rep2})t) \cos(\phi_{1,n}) + \\ \sin(2\pi((f_{0,1} - f_{0,2}) + n f_{rep2})t) \sin(\phi_{1,n}) \end{array} \right] + \\ 2e^{-\delta_{1,n}} \cos(2\pi f_{rep2}t) \\ \left[\begin{array}{l} \cos(2\pi((f_{0,1} - f_{0,2}) + n f_{rep2})t) \cos(\phi_{1,n}) + \\ \sin(2\pi((f_{0,1} - f_{0,2}) + n f_{rep2})t) \sin(\phi_{1,n}) \end{array} \right] \end{array} \right] \quad (1)$$

[0053] The first term of Eq. (1) tells that the spectral information at frequency $f_{0,1} + n f_{rep,1}$ is down-converted at frequency $((f_{0,1} - f_{0,2}) + n f_{rep2})$. The interferogram is similar to an interferogram from classical FTS. Making a synchronous detection of the signal at frequency f_{rep2} is not interesting, as it does not bring additional information.

[0054] Both combs FC1 and FC2 may also interact with the sample 1000 as shown in FIG. 10B. The expression of the interferogram contains the following interesting terms:

$$\begin{aligned} I(t) \subset \sum_n \bigg\{ & \cos(2\pi((f_{c1} - f_{c2}) + n f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) + \\ & \sin(2\pi((f_{c1} - f_{c2}) + n f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \bigg\} + \bigg\{ \cos(2\pi f_{rep2}t) \\ & \left\{ \begin{array}{l} \cos(2\pi((f_{c1} - f_{c2}) + a(n-1)f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) + \\ \sin(2\pi((f_{c1} - f_{c2}) + a(n-1)f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \end{array} \right\} + \\ & \sin(2\pi f_{rep2}t) \\ & \left\{ \begin{array}{l} \sin(2\pi((f_{c1} - f_{c2}) + a(n-1)f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) - \\ \cos(2\pi((f_{c1} - f_{c2}) + a(n-1)f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \end{array} \right\} \bigg\} + \\ & \bigg\{ \cos(2\pi f_{rep2}t) \\ & \left\{ \begin{array}{l} \cos(2\pi((f_{c1} - f_{c2}) - a(n-1)f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) - \\ \sin(2\pi((f_{c1} - f_{c2}) - a(n-1)f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \end{array} \right\} - \\ & \sin(2\pi f_{rep2}t) \\ & \left\{ \begin{array}{l} \sin(2\pi((f_{c1} - f_{c2}) - a(n-1)f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) + \\ \cos(2\pi((f_{c1} - f_{c2}) - a(n-1)f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \end{array} \right\} \bigg\} + \\ & \bigg\{ \cos(2\pi f_{rep2}t) \\ & \left\{ \begin{array}{l} \cos(2\pi((f_{c1} - f_{c2}) + n f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) + \\ \sin(2\pi((f_{c1} - f_{c2}) + n f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \end{array} \right\} + \\ & \sin(2\pi f_{rep2}t) \\ & \left\{ \begin{array}{l} \sin(2\pi((f_{c1} - f_{c2}) + n f_{rep2})t) \cos(\phi_{1,n} - \phi_{2,n}) - \\ \cos(2\pi((f_{c1} - f_{c2}) + n f_{rep2})t) \sin(\phi_{1,n} - \phi_{2,n}) \end{array} \right\} \bigg\} + \end{aligned}$$

$$\begin{aligned}
& \text{-continued} \\
& \textcircled{?} \left\{ \cos(2\pi f_{rep} t) \right. \\
& \left. \left\{ \begin{aligned} & \cos(2\pi((f_{c1} - f_{c2}) - a(n+1)f_{rep2})t) \cos(\textcircled{?} - \textcircled{?}) - \\ & \sin(2\pi((f_{c1} - f_{c2}) - a(n+1)f_{rep2})t) \sin(\textcircled{?} - \textcircled{?}) \end{aligned} \right\} - \right. \\
& \quad \left. \sin(2\pi f_{rep} t) \right. \\
& \left. \left\{ \begin{aligned} & \sin(2\pi((f_{c1} - f_{c2}) - a(n+1)f_{rep2})t) \cos(\textcircled{?} - \textcircled{?}) + \\ & \cos(2\pi((f_{c1} - f_{c2}) - a(n+1)f_{rep2})t) \sin(\textcircled{?} - \textcircled{?}) \end{aligned} \right\} \right\}
\end{aligned}$$

Ⓢ indicates text missing or illegible when filed

[0055] The first line of the expression (2) is the quantity measured by direct detection of the interferogram; without synchronous detection. Compared with what is obtained from the interferogram of Eq. (1), the detection is more sensitive, because both combs FC1 and FC2 experience interaction with the sample, but the instrumental line shape is a bit more complicated as it is frequency dependent: as frequency grows, the difference between the frequencies of the two combs increases. Also, synchronous detection at $f_{rep,2}$ frequency is interesting since the absorption and dispersion may be retrieved thanks to in-phase and in-quadrature detections.

[0056] Any interferogram may present a ratio of about 10^6 between its highest and smallest samples. An important decrease of the interferometric signal as a function of time is illustrated with the interferogram shown in FIG. 11. As a consequence, one of the main difficulties associated with Fourier transform spectroscopy, in particular—but not only—at high spectral resolution, is due to the limited dynamic range of the measurements. In Frequency comb Fourier Transform Spectroscopy, the dc component I_0 is no more a constant but becomes a function $I_0(t)$ of time t , linked to the Fourier components of the pulse repetition rates. Therefore it is difficult to satisfactorily eliminate it by electronic filtering, since this could seriously degrade the sampling. A technical consideration of importance is that, nowadays, the most performing fast acquisition board offers a limited dynamic range of 16 bits, i.e. of 65538 different levels of measurement, which essentially characterize the ADC converters. As a consequence, the recording procedure may be unable to make direct use of a commercial product.

[0057] A specific example embodiment provides a dynamic range solution that is a combination of several points. Fast Detector signals I_A and I_B delivered, as shown on FIG. 12, at two outputs A and B of the interferometer are balanced and subtracted so to remove the unmodulated background, and to obtain only the modulated part of the interferogram. In practice, two electronic gains G_A and G_B , respectively applied to I_A and I_B , are determined before the experiment, by minimizing the difference signal ($G_A I_A - G_B I_B$). These gains are fixed within an experiment. Different electronic gains G_T are applied to the difference signal, depending on time to adapt at best the interferometric signal to the maximum input value of the acquisition unit.

[0058] The gain may be switched in real time or several recording channels may be used for a posteriori selection. Alternatively the gain change is made in a predetermined manner by computer program. In this latter case, the best measured sample is chosen a posteriori, by computer program, and the final value $G_T(G_A I_A - G_B I_B)$ is normalized to

give back the whole interferogram, well measured. Synchronous detection may be performed before digitalization as discussed below.

[0059] According to a specific embodiment, the signal measured by the two receivers, A and B, is synchronously detected at the frequency repetition rate f_{rep} . Since f_{rep} lies typically in the radio-frequency range 0.1-5 GHz, the time-domain method reduces considerably the technical noise when compared to the presently best commercial interferometers detecting interferograms at audio frequencies around 20 kHz. Additionally, absorption and dispersion parameters are given at once from the Fourier Transform of the in-phase and in-quadrature radio-frequency signals, as it may be seen from Eq (2). An embodiment is similar to frequency-modulation spectroscopy with tunable lasers. It has the additional benefits of broadband coverage, optimal modulation index and no need of external modulation.

[0060] An embodiment may also be adapted to selective-spectroscopy methodologies as described below.

[0061] Firstly, velocity modulation is intended to selectively detect the transitions of atomic or molecular ions. With FC-FTS, different schemes may be implemented as illustrated in FIG. 13. FIG. 13A and FIG. 13B make use of a dc discharge 1300. Neutral species are insensitive to the electric field, while due to the electric field, ions acquire a net drift velocity. The effect of this velocity is that the frequencies of the transitions of the ions are Doppler-shifted. Each comb, as in FIG. 13A, or only one of them, as in FIG. 13B, may be split into two counter propagating beams in order to form two interferometers. Difference between the two resulting interferograms or spectra, which may be made by differential detection as shown in FIG. 13A and FIG. 13B (where the two output of the two interferometers, respectively S1 and S2, and S3 and S4, are used according to an embodiment) or a posteriori, brings the selective information, as in one spectrum all the ion frequencies are blue shifted and in the other red shifted. All neutral signals and systematic errors are suppressed from the spectra, leaving only the two Doppler-shifted components of the ion transitions.

[0062] The FIG. 13C and FIG. 13D exploit an ac modulated discharge 1301 to acquire two interferograms with opposite Doppler shifts at a frequency rate depending on the discharge modulation or the recording time of the interferogram. Synchronous detection at the ac modulation cancels the neutral signatures. It is noted that the double differential detection scheme shown in FIG. 13A, FIG. 13B, FIG. 13C, FIG. 13D, may be preferable for signal to noise ratio improvement but not mandatory. For selective detection in the scheme of FIG. 13A or FIG. 13B, it may be for instance enough to consider the difference between the interferograms measured at detectors S1 and S3 (or S2 and S4) or the difference between the resulting spectra. For selective detection in the scheme of FIG. 13C or FIG. 13D, it may be for instance enough to consider only the signal at detector S1 (or S2).

[0063] Secondly, Zeeman spectroscopy may be performed. In a first scheme as illustrated in FIG. 14A, a magnetic field 1400 interacting with the source 1401 is periodically varied. After synchronous or differential detection, only the frequency shifted transitions by the magnetic field may be detected. However, one may consider the symmetry of the Zeeman splitting: one may have to modulate the field between $B_C + B_M$ and $B_C - B_M$, where B_C and B_M are two constants values of the magnetic field. B_C may be equal to B_M but typically cannot be zero, otherwise no signal variation will be

detected. Alternatively, as shown in FIG. 14B, the combs beams may be split into two beams, one probing a cell 1402 with a constant magnetic field $B_C + B_M$ while the other probes a cell 1403 with a constant magnetic field $B_C - B_M$ for differential detection. As it is the case for velocity modulation, it may be preferable but not mandatory to use the two outputs of the interferometers to achieve selective detection. In FIG. 14A, only the signal at detector S1 (or S2) may be measured. In FIG. 14B, it may be for instance enough to consider the difference between the interferograms measured at detectors S1 and S3 (or S2 and S4) or the difference between the resulting spectra.

[0064] Similarly to Zeeman spectroscopy, an electric field may be applied and modulated to selectively detect transitions that are sensitive to the Stark effect.

[0065] Thirdly, different schemes for polarization modulation FCFTS may be implemented as illustrated in FIG. 15. As schematized in FIG. 15A, it may be possible to use a polarization modulator 1500 on one (as illustrated in FIG. 15A with comb FC1 probing the sample 1501) or both combs FC1 and FC2 and to measure in a quick succession one interferogram with a given polarization and one second with the other polarization and to subtract them. As shown in FIG. 15B, it may also be possible to split one (FC1 in FIG. 15B) or both combs beams into two beams, with complementary polarization provided by the polarizers 1502 and 1503. The two beams from FC1 in FIG. 15B probe the sample 1504 and a direct differential detection is performed, as illustrated in FIG. 15B. Alternatively two interferograms may be acquired and subtracted a posteriori. Polarization modulation may be useful to selectively detect polarization-sensitive transitions. It may also be called linear (comparison between s and p polarization) or vibrational circular dichroism (comparison left and right circularly polarized radiation). Some examples of the subject of studies are chiral molecules or the orientation of molecules in thin solid films, liquid crystals, or Zeeman transitions from paramagnetic species. The difference between the absorption of radiation by two polarization states is often extremely small. The idea is to compare, in a quick succession, spectra resulting from the interaction of the sample with light polarized in two different states. In FIG. 15, the use of the two output of the interferometer is a specific example embodiment. As already noted for velocity and Zeeman modulations, a single detector S1 or S2 in the configuration of FIG. 15A may be used. In FIG. 15B, it may be for instance enough to consider the difference between the interferograms measured at detectors S1 and S3 (or S2 and S4) or the difference between the resulting spectra.

[0066] An embodiment may also be adapted to time-resolved applications as described below. In conventional spectroscopy, time-resolved FTS has been a powerful tool to investigate dynamic phenomena, with all the well-known advantages of FTS, especially the wide-spectral range of observation. It consists in performing a time sampling of the evolution of the observed sample, which is excited at every optical path-difference positions. At the end of the experiment, as many interferograms as time samples of the evolution of the source are obtained and transformed to give spectra, each characterizing the light source at a given time. However a major limitation is that the observed sample has to behave in a well repetitive and reproducible manner.

[0067] Time-resolved FC-FTS for dynamic studies may overcome this limitation. Various schemes may be implemented depending of the time-resolution which is looked at.

Furthermore, as acquisition time for a high resolution spectrum is only of the order of 1 millisecond or even 1 microsecond for low resolution information, a way to real-time broadband spectroscopy or single-event spectroscopy is opened with an embodiment.

[0068] For time-resolutions that are worse than the time required for measuring a full interferogram, the measurement principle is straightforward. It consists of measuring the interferograms in sequence. It may be possible to improve the time resolution up to a few μ s by varying the repetition rate of one of the combs in order to provoke the burst which occur when laser pulses from the two combs overlap. This variation in the repetition rate of one of the combs may for instance be achieved by modulating the length of one of the laser cavity with a piezo-electric transducer. If the source behaves in a repetitive manner, it may be possible to repeat several times this method to improve signal-to-noise ratio.

[0069] When the time-resolution or repetition rate of the source cannot fit the optimal sampling requirements, then specific sampling and synchronization schemes may have to be developed. The idea is to lower the interferogram sampling rate by adjusting the frequency difference between the two combs so to meet the source operating conditions.

[0070] If the desired time-resolution is higher than the sampling step of each individual interferogram resulting from the optimal frequency difference between the two combs and if the source of interest has a high repetition rate, it may be possible to perform a high frequency sampling of the time-domain interferogram and to rearrange a posteriori the samples so to generate several interferograms each of them characteristic of the absorption of the source at a given time. The source may then be synchronized to the sampling step of the first time-sample interferogram.

[0071] An embodiment may also be adapted to reflection measurements as described below. With FC-FTS, instead of looking at the light which is transmitted by the sample, it may be possible to look at the light which is reflected by the sample. The sample may be a bulk material, a layer of material (thinner or thicker than the wavelength of the comb radiation) present on a flat reflective surface.

[0072] Furthermore, attenuated total reflection may enable one to probe samples in liquid or solid phase with very easy sample preparation. A probing light beam is passed through a well chosen crystal in such a way that it reflects at least once on the internal surface in direct contact with the sample. This reflection forms an evanescent wave which extends into the sample, typically by a few micrometers. The attenuated energy from each evanescent wave is passed back to the light beam and the beam transmitted by the crystal is then collected by the second comb of the FT spectrometer as it exits the crystal. Attenuated total reflection FC-FTS may also be implemented with the two combs probing the crystal/sample assembly.

[0073] An embodiment may also be adapted to spatially resolved measurements as described below.

[0074] Firstly, as the section of a laser beam may be small, it may be possible to selectively probe tiny spatial regions of a macroscopic sample.

[0075] Secondly, an efficient way with FCFTS to map the composition of a sample may be to measure all the FT spectra simultaneously with an array detector. The object is imaged on the array detector with appropriate optics. Simultaneous measurement of spectra from each point of an image brings hyperspectral imaging.

[0076] Thirdly, microscopy with FC-FTS may bring together the advantages of microscopy with Fourier transform spectroscopy and microscopy with laser sources. Increase of spatial resolution may be obtained together with accurate spectral diagnostics and extremely short measurement times.

[0077] Fourthly, due to the low divergence and high intensity of laser beams, FC-FTS may probe long columns of sample. Alternatively, a laser beam may propagate into a long distance before reaching the sample of interest. LIDAR-type FC-FTS experiment may also be implemented: the sample-scattered light from an intense frequency comb may be collected and analyzed with the second comb.

[0078] From the foregoing it will be appreciated that, although specific embodiments have been described herein for purposes of illustration, various modifications may be made without deviating from the spirit and scope of the disclosure. Furthermore, where an alternative is disclosed for a particular embodiment, this alternative may also apply to other embodiments even if not specifically stated.

1. An interferometer comprising:

a first frequency comb generator

a second frequency comb generator adapted to interact with the first frequency comb generator in order to produce interferences;

wherein the interferometer comprises means for isolating a first single line of the first frequency comb and a second single line of the second frequency comb and beating means for generating at least one beating signal from the first single line and the second single line,

measuring means adapted to trigger data acquisition of said interferences on the beating signal.

2. An interferometer according to claim 1 further comprising a first external optical reference generator and a second external optical reference generator, wherein the beating means are adapted to generate a first beating signal of the first single line and the first external optical reference and a second beating signal of the second single line and the second external optical reference.

3. An interferometer according to claim 1 wherein the beating means are adapted to generate a beating signal of the first single line and the second single line.

4. An interferometer according to claim 1 wherein the isolating means comprise an optical filter and/or a Fiber Bragg grating and/or a Fabry-Perot etalon and/or an electronic filtering.

5. An interferometer according to claim 1, wherein the isolating means comprise a continuous wave laser adapted to beat with the first frequency comb generator and the second frequency comb generator.

6. (canceled)

7. An interferometer according to claim 1 wherein the measuring means are adapted to use the beating signal as a clock for data acquisition of said interferences.

8. An interferometer according to claim 1 having two outputs (A, B), and means for differentially measuring the two outputs.

9. A spectroscopic device for measuring the spectrum of a sample, the device comprising an interferometer comprising:

a first frequency comb generator

a second frequency comb generator adapted to interact with the first frequency comb generator in order to produce interferences;

wherein the interferometer comprises means for isolating a first single line of the first frequency comb and a second single line of the second frequency comb and beating means for generating at least one beating signal from the first single line and the second single line,

measuring means adapted to trigger data acquisition of said interferences on the beating signal,

wherein at least one of said first frequency comb generator and said second frequency comb generator are adapted to interact with the sample.

10. A spectroscopic device according to claim 8 wherein only one of said first frequency comb generator and said second frequency comb generator is adapted to interact with the sample.

11. A spectroscopic device according to claim 8 wherein said first frequency comb generator and said second frequency comb generator are adapted to interact with the sample.

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