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Frequency stabilisation of femtosecond frequency combs with a reference laser

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Abstract. A solution to the key problem of femtosecond metrology - elimination of the frequency offset related to the intracavity dispersion of a femtosecond laser $-$ is proposed. The proposed method involves stabilisation of the intermode interval between equidistant spectral components in a frequency comb produced by a mode-locked femtosecond laser by phase-locking the frequency difference between a pair of discrete spectral components in this comb to the frequency of a reference laser. An introduction of a nonlinear-optical crystal for frequency doubling into the scheme for frequencycomb stabilisation allows the frequency offset related to the intracavity dispersion of the femtosecond laser to be eliminated, thus suggesting the way for absolute stabilisation of frequency combs generated by femtosecond mode-locked lasers. Radiation of a reference laser with such an approach plays the role of an anchor in the femtosecond clockwork.

Keywords: optical metrology, ultrashort pulses, nonlinear optics.

1. Introduction

The use of frequency combs generated by mode-locked femtosecond lasers for high-precision optical measurements $[1-5]$ and creation of optical clock [\[6,](#page-2-0) [7\]](#page-3-0) has led in recent years to revolutionary changes in optical metrology, allowing metrological measurements to be radically simplified both conceptually and technically. The idea of using mode-locked laser sources for high-precision frequency measurements was put forward more than two decades ago by Baklanov and Chebotayev [\[8\]](#page-3-0) (see also [\[9\]\)](#page-3-0) and by the group of Hansch [\[10\].](#page-3-0) However, the practical realisation of this approach became possible only recently due to the rapid progress of femtosecond lasers (see, e.g., the review [\[11\]\)](#page-3-0) and the advent of optical fibres of a new type $[12-16]$.

The most widespread methods of producing stabilised frequency combs for high-precision measurements are based

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on the spectral superbroadening of femtosecond frequency combs in microstructure or tapered fibres [\[3, 5,](#page-2-0) [7, 17\].](#page-3-0) Such an approach allows frequency combs spanning more than an octave to be produced. In recent experiments [\[18\],](#page-3-0) the spectral broadening of unamplified femtosecond Cr:forsterite laser pulses exceeding two octaves has been achieved with the use of tapered fibres [\[16\].](#page-3-0) The intermode interval in frequency combs broadened in an optical fibre can be stabilised, as shown in [\[6,](#page-2-0) [7\],](#page-3-0) with the use of a reference $He-Ne/CH_4$ laser.

A considerable disadvantage of the approach described above stems from the fact that the spectral broadening of ultrashort pulses in optical ébres is often a result of the joint action of a rather complicated combination of nonlinearoptical processes [\[7\].](#page-3-0) Self-phase modulation of femtosecond pulses may be accompanied under these conditions by stimulated Raman scattering, modulation instabilities, shock-wave formation, and other nonlinear optical effects, giving rise to fluctuations of spectral components and perturbing the equidistance of frequency combs. It is of crucial importance to get rid of these sources of instabilities. One of the ways to solve this problem, as will be shown below, is to exclude optical fibres from femtosecond optical metrological systems.

The main purpose of this paper is to show that the concept of femtosecond clock, which was proposed and implemented earlier i[n \[6,](#page-2-0) [7\] a](#page-3-0)nd which involves stabilisation of the frequency interval between the modes in broadened femtosecond frequency combs with the use of a reference $He-Ne/CH_4$ laser, opens the way to create fibre-free systems for high-precision frequency measurements based on the sources of ultrashort light pulses. We will show also that an introduction of a nonlinear-optical crystal for frequency doubling into the scheme for frequency-comb stabilisation allows the key problem of femtosecond metrology to be solved by eliminating the frequency offset related to the intracavity dispersion of the femtosecond laser, thus implying absolute stabilisation of frequency combs generated by femtosecond mode-locked lasers.

2. Stabilisation of the intermode interval in femtosecond frequency combs

We start with the stabilisation of a frequency comb, which is employed, in accordance with the approach developed in $[1 - 7]$, as a frequency ruler for high-precision measurements. Consider an equidistant set of spectral components (Fig. 1) corresponding to a sequence of ultrashort pulses generated by a femtosecond mode-locked laser. Because of the intracavity dispersion in the femtosecond laser [\[17,](#page-3-0) 19], the frequency v_i of the *j*th component in such a frequency comb is not an exact multiple of the intermode interval Δv (Fig. 1) as it is given by the expression:

$$
v_j = j\Delta v + v_0,\tag{1}
$$

where v_0 is the frequency offset related to intracavity dispersion (in particular, the frequency shift $\Delta \varphi$ due to the difference in the group and phase velocities of light pulses inside a laser cavity gives rise to a frequency offset equal to $\Delta \varphi/2\pi T$, where T is the cavity round-trip time [\[17,](#page-3-0) 19]).

Figure 1. Stabilisation of the intermode interval and elimination of the frequency offset in a frequency comb generated by a mode-locked femtosecond laser.

As shown in [\[6,](#page-2-0) [7\],](#page-3-0) the frequency interval Δv between the modes v_m and v_n in the comb can be stabilised by phaselocking the frequency difference between these modes to the frequency v_r of a reference laser (frequency standard). This problem can be solved through the generation of the sum frequency $v_m + v_r$ (Fig. 1) or the difference frequency $v_n - v_m$ in a nonlinear crystal. Obviously, such a stabilisation of intermode intervals is possible only when the spectral range covered by a femtosecond frequency comb is broader than the spectral range corresponding to the frequency v_r of the reference laser (Fig. 1). In particular, the intermode interval in experiments $[7]$ was stabilised using 3.39- μ m radiation of a stabilised $He - Ne/CH_4$ laser. A reference laser with such a wavelength allows stabilisation of frequency combs spanning more than 88.5 THz. Frequency combs with such a spectral width were produced in [\[7\]](#page-3-0) due to the spectral broadening of femtosecond pulses in tapered fibres.

Importantly, the possibility of stabilising the intermode interval in femtosecond frequency combs with the use of 3.39 -um reference radiation, demonstrated in [\[7\],](#page-3-0) suggests the way of performing high-precision frequency measurements without spectral broadening in optical fibres. The procedures of stabilising intermode intervals in frequency combs produced at the output of a mode-locked laser generating ultrashort pulses and frequency combs broadened in an optical ébre are illustrated in Fig. 2. The possibility of excluding optical ébres, intended to broaden frequency combs, from femtosecond metrology systems becomes clear from the stabilisation of the intermode interval with 3.39-µm radiation of a He $-Ne/CH_4$ reference laser as soon as we realise that the spectral interval of 90 THz corresponds to a pulse duration of approximately 11 fs. Modern lasers allow the generation of such pulses directly at the output of a master oscillator $[11, 20-22]$ $[11, 20-22]$.

Figure 2. Stabilisation of the intermode interval and elimination of the frequency offset in a frequency comb produced (a) by a mode-locked laser source of few-cycle pulses and (b) as a result of spectral broadening of a femtosecond frequency comb in an optical fibre.

3. Eliminating the frequency offset related to intracavity dispersion

The method of frequency-comb stabilisation proposed and implemented in [\[6,](#page-2-0) [7\]](#page-3-0) and the technique described in Section 2 permit stabilisation of the intermode interval Δv in femtosecond frequency combs. However, these methods do not eliminate the frequency offset of laser modes in femtosecond frequency combs. Below, we will discuss the way of eliminating this frequency offset and consider the possibility of solving this problem with the use of $5 - 10$ -fs laser pulses without spectral broadening of frequency combs in optical fibres.

Our method of the solution of this problem will involve doubling of the frequency of femtosecond laser pulses in a nonlinear optical crystal. Second-harmonic generation gives rise to a new set of equidistant spectral components v_k (Figs 1, 2) with the frequencies

$$
v_k = k\Delta v + 2v_0. \tag{2}
$$

The generic idea of absolute frequency stabilisation of modes in the femtosecond frequency comb can be then based on the phase locking of the frequency difference between one of the low-frequency components of the second-harmonic frequency comb and one of the highfrequency components of the fundamental-radiation frequency comb to the frequency v_r of the reference laser (Figs 1, 2). Mathematically, such phase locking would imply that the following condition is satisfied:

$$
v_n + v_r = v_k. \tag{3}
$$

Now, if the intermode interval Δv is stabilised with the use of the same reference laser with the frequency v_r (Fig. 1), we have $v_r = N\Delta v$, where N is an integer. Thus, we arrive at

$$
M\Delta v + v_0 = 0,\t\t(4)
$$

where $M = k - n - N$ is an integer.

Since $v_0 < \Delta v$, in accordance with the physical meaning of the quantity v_0 , Eqn (4) immediately implies that the frequency offset related to the intracavity dispersion of the femtosecond laser is eliminated, and the absolute frequencies of the modes generated by the femtosecond laser are stabilised.

We emphasise once again here that the problem of eliminating the frequency offset v_0 was solved earlier [5, [17,](#page-3-0) [23\]](#page-3-0) based on the spectral superbroadening of femtosecond frequency combs in microstructure fibres. The radical difference of the approach to frequency-comb stabilisation proposed in this paper is that our method involves additional measures for stabilising the intermode interval Δv . Such a stabilisation is especially important when femtosecond frequency combs are spectrally broadened through nonlinear-optical processes in optical fibres (see Fig. 2). The intermode interval is subject to variations under these conditions due to dispersion effects, modulation instabilities, shock-wave formation in the pulse envelope, etc. [\[7,](#page-3-0) [24,](#page-3-0) [25\].](#page-3-0) As will be shown below (Section 4), the above-described technique also offers an exciting possibility of mode stabilisation with the use of lasers generating very short light pulses without spectral broadening in optical fibres, thus allowing many processes leading to fluctuations, jitters, and instabilities of frequency combs to be avoided.

The above-described method for absolute frequency stabilisation of equidistant spectral components corresponding to femtosecond pulse sequences can be experimentally implemented with the use of the same technical means as the stabilisation of the intermode interval Δv [6, [7\].](#page-3-0) A certain complication of the experimental scheme is associated with second-harmonic generation in a nonlinear crystal and the system phase-locking the frequency difference of a pair of spectral components picked from the mode sets corresponding to the second harmonic and fundamental radiation. Now, the question of fundamental importance is whether this method of absolute frequency stabilisation can be implemented without spectral broadening of frequency combs in optical ébres. This issue will be discussed in the following section.

4. Stabilising frequency combs in the regime of few-cycle laser pulses

Our approach to the elimination of the frequency offset v_0 involves the phase locking of the frequency difference of a pair of spectral components in frequency combs of the second harmonic and fundamental radiation to the frequency of a reference laser (Figs 1, 2). Let us choose, quite arbitrarily, spectral components with the frequencies $v_n = v_c + 1/2\tau$ and $v_k = 2v_c - 1/\sqrt{2}\tau$ in the frequency combs of fundamental radiation and the second harmonic, respectively (Fig. 1). Then, using Eqn (3) , we find that the spectral gap between the chosen frequency components v_n and v_k can be bridged with reference laser radiation of frequency v_r (Fig. 1) when the pulse duration τ meets the condition

$$
\frac{c}{\lambda} - \frac{1.2}{\tau} \approx v_r. \tag{5}
$$

With $v_r \approx 90$ THz (as in the case of a He-Ne/CH₄ laser), Eqn (5) leads to the following requirements to the pulse duration for Ti: sapphire ($\lambda_{Ti: S} \approx 800$ nm) and Cr: forsterite ($\lambda_{Cr:F} \approx 1.3 \text{ }\mu\text{m}$) laser radiation: $\tau_{Ti:S} \le 4.1 \text{ fs and}$ $\tau_{\text{Cr}:F} \le 8.1$ fs. Thus, the frequency offset can be eliminated with $5 - 10$ -fs Cr: forsterite laser pulses.

Requirements to the pulse duration become much less stringent if the second harmonic of reference laser radiation is employed for absolute frequency stabilisation. In this case, condition (5) becomes weaker than the requirement of Eqn (3) for Cr: forsterite laser radiation ($\tau_{Cr:F} \le 19$ fs) and leads to the requirement $\tau_{Ti: S} \leq 6$ fs for Ti: sapphire laser pulses.

5. Conclusions

The generalisation of the concept of a femtosecond clock [6, [7\],](#page-3-0) based on mode stabilisation in broadened femtosecond frequency combs with the use of a reference $He - Ne$ laser, not only allows the intermode interval to be stabilised in femtosecond frequency combs, but also opens the way of creating systems for high-precision optical measurements using laser sources of very short pulses without spectral broadening of laser pulses in optical ébres. Such an approach would eliminate the sources of errors in highprecision frequency measurements related to modulation instabilities, shock waves in pulse envelopes, and other physical processes accompanying the spectral broadening of femtosecond pulses in optical fibres.

Analysis performed in this paper shows that the introduction of a nonlinear-optical crystal, doubling the frequency of femtosecond pulses, into the scheme of frequency-comb stabilisation solves the key-problem of high-precision optical measurements, eliminating the frequency offset, which may arise, in particular, due to the intracavity dispersion of the femtosecond laser. This approach would thus make it possible to stabilise the absolute frequencies of equidistant spectral components generated by mode-locked femtosecond lasers.

Our estimates demonstrate the possibility of creating optical systems for high-precision frequency, time, and length measurements based on lasers capable of generating $5 - 10$ -fs pulses without a fibre-based spectral broadening of frequency combs.

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References

- 1. Udem Th., Reichert J., [Holzwarth](http://dx.doi.org/10.1103/PhysRevLett.82.3568) R., Hänsch T.W. Phys. Rev. Lett., 82, 3568 (1999).
- 2. Reichert J., Niering M., [Holzwarth](http://dx.doi.org/10.1103/PhysRevLett.84.3232) R., Weitz M., Udem Th., Hänsch T.W. Phys. Rev. Lett., 84, 3232 (2000).
- 3. Diddams S.A., Jones D.J., Jun Ye., Cundiff S.T., Hall J.L., Ranka J.K., Windeler R.S., [Holzwarth](http://dx.doi.org/10.1103/PhysRevLett.84.5102) R., Udem T., Hänsch T.W. Phys. Rev. Lett., 84, 5102 (2000).
- 4. Jones D.J., Diddams S.A., Ranka J.K., Stentz A., Windeler R.S., Hall J.L., Cundi S.T. Science, 288, 635 (2000).
- 5. Holzwarth R., Udem T., Hansch T.W., Knight J.C., [Wadsworth](http://dx.doi.org/10.1103/PhysRevLett.85.2264) W.J., Russell P.St.J. Phys. Rev. Lett., 85, 2264 (2000).
- 6. Bagayev S.N., Chepurov S.V., Klementyev V.M., Kuznetsov S.A., Pivtsov V.S., Pokasov V.V., Zakharyash V.F. Appl. Phys. B, 70, 375 (2000).
- 7. Bagayev S.N., Dmitriyev A.K., Chepurov S.V., Dychkov A.S., Klementyev V.M., Kolker D.B., Kuznetsov S.A., Matyugin Yu.A., Okhapkin M.V., Pivtsov V.S., Skvortsov M.N., Zakharyash V.F., Birks T.A., Wadsworth W.J., Russell P.St.J, Zheltikov A.M. Laser Phys., 11, 1270 (2001).
- 8. Baklanov Ye.V., Chebotayev V.P. Appl. Phys., 12, 97 (1977).
- 9. Bagayev S.N., Chebotayev V.P., Klementyev V.M., Pyltsin O.I. Proc. X Int. Conf. On Laser Spectroscopy (Font-Romeau, France, 1991).
- 10. Eckstein J.N., [Ferguson](http://dx.doi.org/10.1103/PhysRevLett.40.847) A.I., Hänsch T.W. Phys. Rev. Lett., 40, 847 (1978).
- 11. Brabec T., [Krausz](http://dx.doi.org/10.1103/RevModPhys.72.545) F. Rev. Mod. Phys., 72, 545 (2000).
- 12. Knight J.C., Birks T.A., Russell P.St.J., Atkin D.M. Opt. Lett., 21, 1547 (1996).
- 13. Knight J.C., Broeng J., Birks T.A., Russell P.St.J. [Science](http://dx.doi.org/10.1126/science.282.5393.1476), 282, 1476 (1998).
- 14. Cregan R.F., [Mangan](http://dx.doi.org/10.1126/science.285.5433.1537) B.J., Knight J.C., Birks T.A., Russell P.St.J, Roberts P.J., Allan D.C. Science, 285, 1537 (1999).
- 15. Fedotov A.B., Zheltikov A.M., Mel'nikov L.A., [Tarasevitch](http://dx.doi.org/10.1134/1.568334) A.P., von der Linde D. JETP Lett., 71, 281 [\(2000\);](http://dx.doi.org/10.1134/1.568334) Alfimov M.V., Zheltikov A.M., Ivanov A.A., Beloglazov V.I., Kirillov B.A., Magnitskii S.A., Tarasishin A.V., Fedotov A.B., Mel'nikov L.A., Skibina N.B. JETP Lett., 71, 714 (2000); [Zheltikov](http://dx.doi.org/10.1070/pu2000v043n11ABEH000839) A.M. Phys. Usp., 43, 1203 [\(2000\).](http://dx.doi.org/10.1070/pu2000v043n11ABEH000839)
- 16. Birks T.A., Wadsworth W.J., Russell P.St.J. Opt. Lett., 25, 1415 (2000).
- 17. Udem T., Reichert J., Holzwarth R., Diddams S., Jones D., Jun Ye., Cundi S., Hänsch T., Hall J., in The Hydrogen Atom: Precision Physics of Simple Atomic Physics (Berlin: Springer, 2000, p. 125).
- 18. Akimov D.A., Fedotov A.B., [Podshivalov](http://dx.doi.org/10.1134/1.1434286) A.A., Zheltikov A.M., Ivanov A.A., Alfimov M.V., Bagayev S.N., Pivtsov V.S., Birks T.A., Wadsworth W.J., Russell P.St.J. JETP Lett., 74, 460 (2001).
- 19. Reichert J., Holzwarth R., Udem Th., Hansch T.W. Opt. Commun., 172, 59 (1999).
- 20. Xu L., Tempea G., Poppe A., Lenzner M., Spielmann Ch., Krausz F., Stingl A., Ferencz K. Appl. Phys. B, 65, 151 (1997).
- 21. Sutter D.H., Steinmeyer G., Gallmann L., Matuschek N., Morier-Genoud F., Keller U., Scheuer V., Angelow G., Tschudi T. Opt. Lett., 24, 631 (1999).
- 22. Apolonski A., Poppe A., Tempea G., Spielmann Ch., Udem Th., [Holzwarth](http://dx.doi.org/10.1103/PhysRevLett.85.740) R., Hansch T.W., Krausz F. Phys. Rev. Lett., 85, 740 (2000).
- 23. Holzwarth R., Reichert J., Udem Th., Hänsch T.W. Laser Phys., 11, 1100 (2001).
- 24. Agrawal G.P. Nonlinear Fibre Optics (Boston: Academic, 1989).
- 25. Nakazawa M., Tamura K., Kubota H., Yoshida E. Opt. Fibre Technol., 4, 215 (1998).