Reply to comment on "Sub-10-fs mirror-dispersion-controlled Ti:sapphire laser" and "Ultrabroadband ring oscillator for sub-10-fs pulse generation"

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The above Comment on our recent study[1,2] addresses three major points: (1) our statement regarding a contradiction between our recent experimental results and the conclusions of Christov *et al.* drawn from their previous theoretical research,[3] (2) comparison of the quality of sub-10-fs pulses that can be obtained from mirror-dispersion-controlled (MDC) and quartz-prism-controlled[4,5] (QPC) Kerr-lens-mode-locked[6] (KLM) Ti:sapphire (Ti:S) oscillators, and (3) the formation of solitonlike pulses in femtosecond solid-state lasers.

(1) In Ref. 3 the authors of the Comment (henceforth the authors) refer to the case in which the oscillator contains a given amount of negative fourth-order dispersion (FOD) as the ideal one for producing the shortest pulses, which suggests that introducing some negative FOD is a prerequisite for obtaining the best achievable performance of the KLM Ti:S oscillator in the sub-10-fs regime. We recently demonstrated a MDC sub-10-fs oscillator in which the second-order or group-delay dispersion (GDD) was kept nearly constant over a bandwidth of ≈ 200 nm [i.e., third-order dispersion (TOD) and FOD were negligible]. The quoted bandwidth was shown to limit the minimum achievable pulse duration to ≈ 7 fs. This pulse width could be achieved without the introduction of FOD simply by gradual reduction of the magnitude of negative GDD, in apparent contradiction to the authors' conclusion regarding the "optimum" FOD in Ref. 3.

We agree with the authors that the QPC sub-10-fs Ti:S laser is operated in a regime that is distinctly different from the operation regime of the MDC oscillator, because a significant amount of negative FOD remains in the QPC system at a center wavelength of ≈ 850 nm, where TOD is eliminated.[7] Under these conditions changing the sign of the GDD from negative to positive results in broader mode-locked spectra and shorter pulses, as the authors demonstrated both theoretically[3] and experimentally.[4] This does not imply, however, that FOD is generally required for achieving the spectral widths and pulse durations reported in Refs. 3 and 4, as suggested by the latter. As a matter of fact, in the MDC Ti:S oscillator[2] a pulse duration shorter than that reported in Ref. 4 and comparable with the theoretical result shown in Fig. 3b of Ref. 3 was achieved without appreciable FOD present in the cavity.

(2) We agree that there is no one-to-one correspondence between pulse quality and the time–bandwidth product of a pulse. However, the authors' arguments do not at all disprove our statement. The term "pulse quality" is not precisely defined; therefore it requires some elaboration. There are two major application fields of ultrashort pulses, namely, nonlinear optics and time-resolved spectroscopy. The former field often requires pulses with high prepulse contrast (A), particularly if terawatt pulses interact with solid matter. In time-resolved spectroscopic investigations of condensed matter our ability to selectively excite elementary excitations to different energy levels is traded off against the time resolution that the subsequent relaxation processes can be studied with. This problem can be alleviated by use of pulses with time–bandwidth products as small as possible (B).

The extent to which requirements (A) and (B) are met by the sub-10-fs pulses delivered by the two systems that are being compared can be readily quantified. A simple analysis of the experimental results reported in Refs. 1 and 4 yields the following results. In the absence of spectral phase modulation the instantaneous power $P(t)$ of the 8.5-fs pulses generated by the QPC Ti:S laser[4] is approximately a factor of 10^{-3} less than the peak power 25 fs before the pulse center, i.e., $P(-25\text{ fs}) \approx 10^{-3}P(0)$, as obtained by a Fourier transform of the double-peaked spectrum (with the dip reduced by a factor of 2) in Fig. 2(a) of Ref. 4. The same procedure yields $P(-25\text{ fs}) < 10^{-5}P(0)$ for the 8-fs pulses presented in Ref. 1. Hence the leading edge of the sub-10-fs pulses generated by the MDC oscillator can be orders of magnitude steeper than that of the sub-10-fs output from the QPC system. Less spectacular but more obvious is the difference between the performance of the two systems for time-resolved spectroscopy. Although it offers the same (or slightly better) time resolution, the MDC oscillator exhibits a 40% narrower bandwidth, i.e., a higher spectral purity than its QPC counterpart.

We believe that these findings justify the claim that a sub-10-fs solid-state laser operating in the regime of negative GDD with high-order dispersion eliminated from the cavity (currently embodied by the MDC Ti:S oscillator[1,2]) offers a significantly better pulse quality than a sub-10-fs oscillator operated with negative FOD and positive GDD (embodied by the QPC Ti:S oscillator[4,5]).

(3) We strongly disagree with the authors' comment on the role of solitonlike shaping in mode-locked lasers and their understanding of the major pulse-shaping ef-

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fects in KLM lasers. The results of previous investigations of femtosecond dye lasers lead the authors to the general conclusion that solitonlike shaping does not take place in femtosecond lasers in typical operation regimes. In this generalization the authors seem to have ignored striking differences between dye and solid-state lasers as well as the results of a series of theoretical and experimental studies of femtosecond solid-state lasers performed over the past 5–6 years.

In strong contrast with that for dye lasers, self-amplitude modulation (SAM) is far less effective than self-phase modulation (SPM) in additive-pulse mode-locked (APM) and KLM solid-state lasers. The SAM and SPM coefficients, κ and ϕ (Ref. 8), respectively (in the notation of Haus *et al.*, γ and δ, respectively[9–11]) have the same units and can be directly compared. In typical KLM and APM systems $\phi/\kappa \geq 10$; i.e., the SPM-induced round-trip phase shift is an order of magnitude larger than the SAM-induced dynamic change in the round-trip gain.[10–12] As a consequence, irrespective of the pulse duration, SPM rather than SAM dominates pulse formation in the overwhelming majority of KLM and APM solid-state lasers. A clear manifestation of the predominance of SPM is the emergence of strongly chirped pulses with nearly rectangular spectral shapes in the regime of positive cavity GDD[13] and solitonlike pulse formation in the negative GDD regime.[5] There are two major effects that are characteristic of solitonlike pulse formation: a linear dependence of pulse duration on GDD and the emergence of sidebands in the frequency spectrum. The former was predicted by the master-equation theory of Haus *et al.*[9–11] and subsequently verified experimentally in KLM Ti:S lasers for pulse durations ranging from 7 to 200 fs,[2,5,14,15] as well as in a KLM Cr:forsterite laser in the range 25–85 fs.[16] Soliton sideband generation owing to dispersive and periodic perturbations was theoretically treated by Kelly *et al.*[17] and experimentally observed in a number of bulk and fiber solid-state systems (see, e.g., Refs. 5 and 18).

In addition to those effects mentioned in Ref. 11, solitonlike shaping is perturbed by a few other effects that are inherent in KLM systems, such as the separate action of SPM and GDD (discreteness)[5,8,12] and space–time effects[19,20] (as pointed out by the authors). We investigated the implications of the former[8] and found that it contributes to the steady-state pulse duration as given by Eq. (1) of Ref. 2. The authors are incorrect when stating that "there is no proof that this formula gives the steady-state pulse duration." In fact the model[8] predicts a difference in the steady-state pulse duration at the dispersive and the nonlinear cavity ends, which was verified experimentally.[21] More importantly, the formula predicts a minimum pulse duration of $\tau_{\min} \approx \sqrt{|D|}$ for a given value, $D(<0)$, of the intracavity GDD, unless limiting effects (see below) come into play. This prediction was also quantitatively verified in a number of APM fiber lasers that used active fibers with positive dispersion, in which discreteness is more pronounced, as was demonstrated by Dennis and Duling recently.[18] The large number of experimental results (gathered and in part obtained by Duling and co-workers) that confirm our prediction over a wide dynamic range of pulse durations[18] gives us great confidence in the formula criticized by the authors.

As was pointed out previously, Eq. (1) of Ref. 2 loses its validity as soon as the spectral extension of the solitonlike pulse exceeds the wavelength range over which $D(<0)$ is approximately constant in the cavity. This limit could be approached by reduction of $|D|$ and (or) an increase in the intracavity pulse energy in the MDC oscillator. In Ref. 2 we presented experimental evidence that the constant-GDD bandwidth is the dominant effect that currently limits the performance of the solitonlike Ti:S oscillator to $\approx$7 fs. It is hoped that increasing this bandwidth will allow the generation of even shorter high-quality solitonlike pulses from the Ti:S oscillator. However, even in the hypothetical case of infinitely broad bandwidth, Eq. (1) of Ref. 2 breaks down at some small but finite negative values of D, depending on the values of κ, ϕ, and the intracavity pulse energy, as was pointed out in Ref. 8. Therefore the authors may not be surprised that Eq. (1) does not account for the behavior of a system operated in the regime of zero or positive cavity GDD.

Our major conclusions from the results presented in Refs. 1 and 2 are that (i) the solitonlike interplay between positive SPM and negative GDD is capable of generating high-quality ultrashort pulses down to $\approx$7 fs in the absence of high-order dispersion and in the presence of efficient SAM and (ii) this performance is limited by the finite bandwidth over which the negative GDD is approximately constant. The currently available experimental results do not rule out the feasibility of generating high-quality 5-fs or possibly even sub-5-fs solitonlike pulses from the Ti:S laser by an increase in constant-GDD bandwidth and slightly more efficient SAM. Of course, while we are attempting to exploit this possible potential fully, it is important to think simultaneously about novel pulse-shaping mechanisms that can push the limit of the solitonlike regime without compromising pulse quality.

Note added in proof: After submission of our reply further experimental work relevant to the above discussion was reported. Keller and co-workers demonstrated the generation of 6.5-fs pulses from a hybrid mirror-prism-dispersion-controlled KLM Ti:S laser operated in the same regime as the MDC oscillator discussed above.[22] Kasper and Witte performed high-dynamic-range autocorrelation measurements to compare the performance of MDC and QPC oscillators. Although they used "first-generation" chirped mirrors with a significantly narrower GDD bandwidth than those used in Refs. 1 and 2, their MDC oscillator yielded a substantially higher contrast ratio than the corresponding QPC system in the sub-20-fs regime.[23]

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