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Peer Review File

Few-femtosecond phase-sensitive detection of infrared electric fields with a third-order nonlinearity

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Traditionally, electric field waveforms beyond radio frequencies were measured using a second-order nonlinear interaction with a laser pulse, but synthesizing such a gate pulse is difficult when sampling MIR and NIR transients. The authors present an alternative approach using a third-order nonlinear interaction with a long multi-cycle pulse to directly retrieve an electric-field transient. They demonstrate measuring the real part of a sample's dielectric function. The new method allows for MIR-to-NIR time-resolved spectroscopy that extracts spectral amplitude and phase information, which is useful for studying molecular vibrations and fundamental excitations in condensed-matter physics. The idea of the paper is interesting and intriguing, however some key information is missing, and the content is a bit obscure. I think the paper is more suitable to a journal more specialized in ultrafast lasers. I suggest a substantially major revision. My comments are listed specifically as below:
1 There are too many jargons without explanation, for example, electro-optic sampling (EOS), carrier-envelope-phase (CEP)-stable, and so forth. The authors claim the advantages of their technique comparing with those ones. However, readers may not be convinced until they understand what the jargons mean.
2 The authors used highly nonlinear fibers to change the spectral properties of the probe and signal light. The authors only described the experimental process without explaining the underlying mechanism? Furthermore, it is suggested to give typical spectra of two beams output from highly nonlinear fibers, which is helpful to understand the interaction between two beams.
3 How did authors know the duration of the probe and signal pulses output from highly nonlinear fibers. Where is the ellipsometer in Fig. 1?
4 In Fig.2 the authors performed spectral filtering and inverse Fourier transform to the measured signal. However, the details about spectral filtering are not given, for example, what are the filtering center frequency, bandwidth and profile? I think they are crucial parameters in analyzing the results.
5 The third order nonlinearities in MCTOS are called up conversion, down conversion, direct down conversion. What are those effects are specifically, for example, Kerr, Raman, four wave mixing? What do the arrows in Fig.3 mean? What the different \omega s and \Omega in Eq. 1 and 2 mean?
6 How the authors can prove their retrieved phase profiles are correct, as they do not have a ground truth? I think the measurements of the group indices of refraction in Fig. 6 are not straightforward evidence.

Reviewer #2 (Remarks to the Author):

In their manuscript, the authors describe a novel method for a phase sensitive detection of IR pulses. Similarly to the EOS, the method mixes the signal and the probe fields on a non-linear crystal to generate new photons, spectrally overlapped with the original probe field. Their interference results in a change of polarization state, which encodes the phase of the signal field. Unlike EOS, the authors utilize a higher-order $\chi(3)$ process, resulting in more lenient and practically convenient requirements on the probe bandwidth and signal frequency. The authors provide an intuitive picture of the process, as well as a rigorous mathematical description, and demonstrate the validity of the picture with the bandwidth-limited experiments. They also show the application of the technique, measuring the group index of refraction in Ge and GaSb.
The proposed method can become a valuable practical substitution for EOS and is of interest to the community. The manuscript is well written. I recommend it for publication, provided the comments below are addressed.

1. The authors mention that the resulting sensitivity range is [fc-3ΔBW/2, fc+3ΔBW/2]. Would not spectral overlap between the signal and the probe substantially complicate the measurement or even make it impossible?

2. How does the phase of the probe affect the measurement? E.g., I can imagine a strong chirp should eventually lead to stretching of the observed trace in time.

3. From the picture described in Fig. 4, the DC signals from an ellipsometer equipped with a QWP and a HWP should be pi/2 out of phase. It does not appear to be the case in Fig. 5c. What is the reason for that?

4. “An alternative successful strategy that circumvents this requirement is to exploit a highly nonlinear interaction between a strong probe pulse and the signal field...” – in one of the first works utilizing this approach, J. Phys. B: At. Mol. Opt. Phys. 51 065603 (2018), the $\chi(3)$ nonlinearity was also employed. It might be worth mentioning its relation to the described technique.

Reviewer #3 (Remarks to the Author):

The manuscript "Few-femtosecond phase-sensitive detection of infrared electric fields with a third-order nonlinearity" by H. Kempf, et al., presents an interesting demonstration that the electric field (both amplitude and phase) of a NIR laser pulse can be measured using a third-order non-linear optical process.

In general, I would say the manuscript contains suffient novel and interesting physics to be eventually published. However, I recommend major revisions (to be detailed below) in order to: (1) make the manuscript readable to a broader audience, (2) clarify the general applicability and limitations of the technique. In addition, I have some more technical/detailed recommendations.

1) General readability:

The manuscript is divided into a main article for a pedagogic explanation and a supporting information for the technical details. This is a good choice; however, I recommend to put even more technical information into the supporting information and increase the efforts to expand on general considerations in the main text.

Just to motivate the above: Personally for me, I really did not get anything useful out of the phasor description on page 4 at first reading - only after reading the entire supporting information and after some thinking, I got the idea. The problem is that the main text on page 4 is only "the supporting information without integrals", i.e., there are no extra efforts to explain the physics as compared to what is already written in the supporting information. I could live with a much reduced version of the text on page 4 and also without figure 4 in the main text. This experiment must be "understood in frequency space", which could be clarified more in the main text (see below).

For the general readability of the supporting information: The text works reasonably well until equation (S24). But the introduction of the "phasor" is difficult to understand. Behind this lies the actual experimental detection setup with the balanced detection, wave plates, and homodyning. I recommend to add more details here such that the reader can understand that the electric field hitting the detector contains a superposition of the probe field and the generated fields, and that upon squaring (in order to calculate the photon flux as the detector measures) cross terms emerge and eventually give rise to the differential current given by Eqs. (5) and (6) in the main text.

Next, the supporting information proceeds to show that the detected current can be decomposed into a response function R(Omega) and the signal field A_S(Omega)exp(-i*Omega*t_D). This is a nice result; however, the authors fail completely in discussing this result (this discussion should be in the main text). The point is that if R(Omega) is a function which is much broader than A_S(Omega), then Eqs. (5) and (6) just reduce to a simple Fourier transform taking the signal field back to the time domain. This is the main point of the whole manuscript! And this is also what the authors are doing in Fig. 2(b) (after filtering the DDC component out). But the issue here is that if R(Omega) is not much broarder than A_S(Omega), or is at least a constant in the relevant frequency range of A_S(Omega), the inferred field (exemplified in Fig. 2b) will be a distorted version of the actual field. This point must be discussed carefully and honestly in the main text. I would recommend to show the calculated spectra of R(Omega) in the main text and compare them to the OSA signal spectrum.

2) General applicability and limitations.

In continuation of the above discussion of R(Omega), it would be useful to provide in the main text a discussion on what setting one should actually use to get the detection scheme to work best. Which choice of band pass filter (BPF) is the optimal one and why? Is it a necessity that the signal field does not overlap with the probe field in frequency space (as is the case shown in Fig. 3)? If yes, what limitations will this present? Will it be possible to achieve a situation where the R(Omega) function in Eqs. (5) and (6) can actually be taken as a constant such that the result exemplified in Fig. 2b becomes the true electric field? Would it be possible to calculate R(Omega) [from S28-S30 based on measured values of A_P(omega) by e.g. an OSA] to a level of accuracy such that the distorion could be compensated for? Evidently, this is not possible for the signal field shown in Fig. 2a and 3a (being too much on the edge of sensitivity), but it might be useful to discuss it more generally in connection to what strategy the reader should pick if he/she would like to apply the technique.

The authors mention that the signal pulse does not have an absolutely stabilized CEP. But they should explain in the main text why, and under what conditions, the technique works anyway. It seems that (Delta phi_S^0) and (Delta Phi_P^0) are assumed to share the same phase fluctuations, which is probably fair when they are derived from the same master laser. Along the same lines, the assumptions behind Eqs. (S20) and (S21) should also be commented on. Will the technique only work for bandwidth-limited pulses?

The phase matching conditions [Eqs. S16-S19] are never really discussed. Do the have any practical impact on R(Omega) defined in (S33) and (S34)? It there anything the reader should take into account if designing a similar experiment?

3) Minor things:

On page 2, it would be useful to state also the central wavelengths corresponding to the 156 THZ and 246 THz fields.

There seems to be a discrepancy between Fig. 3 of the main text and Eqs. (S13-S15). The UC process seems fine: There are two absorptions of the probe field (green arrows pointing upward and E_P has no asterisk in S13) and one emission of a signal photon (red arrow going down and an asterisk on E_S). However, for the DC process we have two downward green arrows (emission) but only an asterisk on one probe field in (S14). In general, figure 3 of the main text is very confusing. There are three frequencies at play from the probe field [e.g., omega, omega1, and omega-(omega1-Omega)], but there are only two colors (blue and green) or arrow lengths at play in the figure. Furthermore, it is not clear what is the significance of the energy gain/loss represented by the black arrows. There is energy conservation in the basic interaction for each frequency setting within the integrals in S13-S15.

In the methods section the balanced photodiode is described. It would be useful to add that this is an

InGaAs detector, or alternatively, to state the working wavelength range of 800-1700 nm.

Now we turn to the supporting information:

In the first line of section 2, there is a reference to "Sec. 4 of the main paper". But there is no section numbering in the main paper.

Please state in connection to Eq. (S2) that this is valid for a non-depleted field. It would also be useful to clarify the proper ansatz of the generated electric field E, which is allowed to vary as a function of z (just so the reader does not mistakenly take E to be on the form of S2).

Please mention why S6 is valid in the present experiment.

Equations (S10-S12) are somewhat confusing. At first glance, the reader does not understand the significance of the special definitions of omega_UC, omega_DC and omega_DDC. In fact, they are not more special/significant than omega_1. I would recommend to instead state carefully the correspondence between (S13-S15) and (a correct version of) figure 3 in the main text and argue what sum rule of frequencies apply to each equation.

It is mentioned that the special form of (S20) and (S21) is valid for a bandwidth-limited pulse. Adding a reference (for instance to a text book) would be useful.

It seems to me that there should not be an asterisk on k and \xi in (S28). Check also Eq. (S33).

Response to reviewers:

Reviewer #1 (Remarks to the Author):

Traditionally, electric-field waveforms beyond radio frequencies were measured using a second-order nonlinear interaction with a laser pulse, but synthesizing such a gate pulse is difficult when sampling MIR and NIR transients. The authors present an alternative approach using a third-order nonlinear interaction with a long multi-cycle pulse to directly retrieve an electric-field transient. They demonstrate measuring the real part of a sample's dielectric function. The new method allows for MIR-to-NIR time-resolved spectroscopy that extracts spectral amplitude and phase information, which is useful for studying molecular vibrations and fundamental excitations in condensed-matter physics. The idea of the paper is interesting and intriguing, however some key information is missing, and the content is a bit obscure. I think the paper is more suitable to a journal more specialized in ultrafast lasers. I suggest a substantially major revision. My comments are listed specifically as below:

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We acknowledge the need to explain basic concepts that are most important to the reader. In the revised version, we added short and intuitive explanations to these terms at the point of their introduction.

2 The authors used highly nonlinear fibers to change the spectral properties of the probe and signal light. The authors only described the experimental process without explaining the underlying mechanism? Furthermore, it is suggested to give typical spectra of two beams output from highly nonlinear fibers, which is helpful to understand the interaction between two beams.

Indeed, the process of nonlinear wavelength conversion in a highly nonlinear fiber was described in a minimal manner. While it is not the main theme of the current paper, we realize that it is necessary to expand on this point and therefore added explanatory comments on page 2 of the main text. In addition, Section 1 of Supplemental 1 now contains a full characterization of the broadband probe pulse including its spectrum and standard frequency-resolved optical gating (FROG) measurement. The spectra of both signal and probe pulse appear in Fig. 3a of the main text.

3 How did authors know the duration of the probe and signal pulses output from highly nonlinear fibers.

The duration of the probe pulse was evaluated with a standard FROG setup: A time-delayed sum-frequency mixing between two copies of the pulse is spectrally resolved on a spectrometer. An iterative algorithm retrieves the pulse duration from these results. FROG results and analysis were added to Sec. 1 of Supplemental 1. Information on the signal pulse duration can be obtained from the direct-downconversion (DDC) signal of MCTOS (positive offset in Fig. 2a) from which we retrieve the 40 fs estimate given in the main text.

Where is the ellipsometer in Fig. 1?

The ellipsometry setup is now contained in a highlighted frame in Fig. 1 of the main text. An ellipsometer is composed of a waveplate followed by a Wollaston prism that divides the beam according to two orthogonal linear polarizations. The balanced photodiode detection monitors slight variations (rotation or ellipticity) in the polarization of the input beam.

4 In Fig.2 the authors performed spectral filtering and inverse Fourier transform to the measured signal. However, the details about spectral filtering are not given, for example, what are the filtering center frequency, bandwidth and profile? I think they are crucial parameters in analyzing the results.

We thank the referee for pointing out this missing information. It is now added to the Methods section of the main text. Spectral filtering was achieved by multiplication of the spectral amplitude with a super-Gaussian function, $\exp\left(-\left(\frac{f-f_0}{\Delta f}\right)^6\right)$, with f_0 = 155 THz and Δf = 80 THz. This filter is designed to include the spectral range of the signal (~140-180 THz) while strongly suppressing the lower frequency content (<40 THz). Thanks to the large separation between the high (~150 THz) and low frequency (<40 THz) components, the precise functional form or parameter values has a negligible influence on the output signal in the time domain.

5 The third order nonlinearities in MCTOS are called up conversion, down conversion, direct down conversion. What are those effects are specifically, for example, Kerr, Raman, four wave mixing? What do the arrows in Fig.3 mean?

The reviewer points to the delicate issue of classification of third-order nonlinear processes. In general, all the processes involved here can be referred to as four-wave mixing – a result from a third-order nonlinearity of multiple frequencies. Raman effects include a dissipative interaction with elementary excitations of the material which is not the case here. While the DDC may be described as an optical Kerr effect (cross-phase modulation via change of the refractive index) the UC and DC cannot be classified in such a manner.

The depiction appearing in Fig. 3b is a standard way to illustrate nonlinear mixing processes (for example, *Nonlinear Optics*, Boyd R. W., Academic Press Inc.). The length of arrows, representing the participating photons, matches their energies. An upward (downward) arrow indicates annihilation (creation) of a photon. In the current version, we add black arrows to depict energy gain or loss of the initial photon to highlight the distinction between UC and down conversion DC.

What the different \omega s and \Omega in Eq. 1 and 2 mean?

In equations (1) and (2) capital Ω refers to signal frequencies whereas lower-case ω represents photons in the probe spectrum. Since four-wave mixing processes generally include multiple probe frequencies we employ subscripts to discern between those when referring to the quasimonochromatic model.

6 How the authors can prove their retrieved phase profiles are correct, as they do not have a ground truth? I think the measurements of the group indices of refraction in Fig. 6 are not straightforward evidence.

Indeed, detecting the electric field transient of an optical pulse in the mid infrared is a challenging task that has been accomplished only by a handful of papers. In fact, this is part of the motivation of the current work. There is no standard way to obtain a ground truth for such an experiment. However, we believe that the multiple measurements performed in the current study and their agreement with theory provide ample evidence that we accurately evaluate the spectral phase of the signal pulses.

In the absence of a ground truth for an absolute spectral phase it is common practice to compare the phase measurement with and without propagation in a bulk material slab and analyze the resulting change in phase (e.g. Hammond et al., J. Phys. B-At. Mol. Opt., 51, 6 (2018)). Such a measurement, carefully analyzed in Fig. 6 of the paper, reproduces the group refractive index of Ge with high precision.

Moreover, we note that our straightforward theoretical model, that does not employ any fit parameters, excellently predicts the results of measurements with multiple band-pass filters and with two separate ellipsometer configurations (see Fig. 5). The precision of the model validates that our description of the nonlinear interaction here is complete. Consequently, we can confidently state that the measured transients accurately reflect the electric field waveform (incl. spectral phase). As with any broadband measurement, the spectral response of MCTOS [see Eqs. (S30) -(S33) of Supplemental 1] should be considered in order to accurately retrieve the electric field of the pulse (see added discussion on pages 5-6 of the main text).

Reviewer #2 (Remarks to the Author):

In their manuscript, the authors describe a novel method for a phase sensitive detection of IR pulses. Similarly to the EOS, the method mixes the signal and the probe fields on a non-linear crystal to generate new photons, spectrally overlapped with the original probe field. Their interference results in a change of polarization state, which encodes the phase of the signal field. Unlike EOS, the authors utilize a higher-order χ(3) process, resulting in more lenient and practically convenient requirements on the probe bandwidth and signal frequency. The authors provide an intuitive picture of the process, as well as a rigorous mathematical description, and demonstrate the validity of the picture with the bandwidth-limited experiments. They also show the application of the technique, measuring the group index of refraction in Ge and GaSb.

The proposed method can become a valuable practical substitution for EOS and is of interest to the community. The manuscript is well written. I recommend it for publication, provided the comments below are addressed.

1. The authors mention that the resulting sensitivity range is [fc-3ΔBW/2, fc+3ΔBW/2]. Would not spectral overlap between the signal and the probe substantially complicate the measurement or even make it impossible?

The referee raises an important point. A partial overlap of the signal with the original probe spectrum will potentially overwhelm the MCTOS signal and degrade the measurement of spectral phase. To address this point, we have added a paragraph to the discussion on page 6 summarizing the answer given below.

We can divide the discussion to three types of signal spectra. In the first case, where the signal spectrum does not overlap with that of the probe ($f_c \pm \Delta\mathrm{BW}/2$), MCTOS can be performed straightforwardly. In case that there is partial overlap between the signal and the probe one can apply a sharp spectral filter to exclude the signal spectrum and the corresponding portion of the probe. The remaining part of the probe includes nonlinear interactions between all frequencies within the MCTOS bandwidth and would therefore still allow the retrieval of the spectral phase (see Fig. 5 of the main text). In case the signal is spectrally contained within the probe, standard linear interferometry measurements can extract the spectral phase. An extension of MCTOS will be required only if the spectrum of the signal is so broadband as to contain that of the probe. In such a case, MCTOS is probably cannot provide a reliable measurement.

Finally, we note that since the spectral range of MCTOS is tunable according to the center frequency of the probe pulse, it can be adjusted in order to mitigate the issues described above. That is, the spectral overlap between signal and probe can be avoided. Practically, tunability can be readily achieved with HNF technology by controlling the input-pulse parameters (e.g., dispersion and power) or the length of the fiber.

2. How does the phase of the probe affect the measurement? E.g., I can imagine a strong chirp should eventually lead to stretching of the observed trace in time.

The reviewer’s statement is accurate; the spectral phase of the probe does affect the detected transient. In our case, the probe exhibits a very small chirp as evidenced by the frequency-resolved optical gating (FROG) results shown in Section 1 of Supplemental 1. Therefore, the MCTOS modeling, presented here, treats the probe pulse as bandwidth limited. This approximation is justified when comparing the nearly identical spectral-response for a bandwidth-limited probe and the pulse characterized by the FROG measurements (see Fig. S3b of Supplemental 1).

As the reviewer assumes, the bandwidth of MCTOS reduces when considering a significant chirp, thus stretching the temporal envelope of the detected transient. To numerically demonstrate this effect, we calculated the spectral response of MCTOS for a linearly chirped probe (yellow, Fig. S3b). Qualitatively, the linear chirp reduces the effective momentary bandwidth of the probe and thus the bandwidth of the spectral response function. We note that this observation is common to all types of 'optical oscilloscope' methods (see, e.g. K. T. Kim et al., Nat. Photon., 7, 958-962 (2013) and M. R. Bionta et al., Nat. Photon., 15, 456-460 (2021)).

3. From the picture described in Fig. 4, the DC signals from an ellipsometer equipped with a QWP and a HWP should be pi/2 out of phase. It does not appear to be the case in Fig. 5c. What is the reason for that?

Indeed, one would expect a phase difference of $\pm\pi/2$ between the two measurement configurations shown in Fig. 5a and c. However, the measurements where not taken directly one after the other but rather about one hour apart. As a result, a drift of the relative CEP has likely occurred leading to a randomized phase difference between the two transients. A possible source for such phase noise is an uncontrolled temperature variation that modifies material dispersion. Ideally, employing a low-bandwidth stabilization procedures for the relative CEP and for the time delay between both branches would circumvent this problem.

We note that the CEP relation between multiple laser sources is generally tough to stabilize and often experiences significant drifts. In fact, such drift is often nontrivial to observe without a time-domain measurement of the pulses. MCTOS, like other techniques for time-domain detection, offers a direct characterization of CEP and waveform stability for pulsed laser sources.

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We thank the referee for drawing our attention to this relevant work which we were not aware of. In the revised version of the manuscript, we include a reference to the paper and discuss its relation with the current work (see page 2). In Hammond et al., the electric-field transient of a signal pulse is retrieved by mixing it in a non-collinear geometry with a probe pulse in a material with a third-order nonlinear response. The position-dependent interference pattern between the newly generated fields and the probe pulse, encodes the signal field waveform is retrieved from a CCD-camera image. The parallel nature of this approach enables a single-shot characterization of the field transient. On the other hand, this implementation requires relatively high pulse energies of 20-300 μJ output by a laser amplifier working in a kHz repetition rate. Such sources are significantly more complex as compared to the system used for our study; most ultrafast microscopy and spectroscopy setups rely on MHz repetition rate systems with pulse energies in the nJ regime. In the current work, MCTOS employ pulse energies of only 0.2-1 nJ, 5 orders of magnitude weaker. Such pulse energies are easily achievable even with a high repetition-rate laser oscillator. The expansion of an 'optical oscilloscope' to ubiquitous MHz repetition-rate lasers is one of the main advantages that MCTOS offers.

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We thank the reviewer for striving to improve the clarity of the article. We full heartedly agree that the previous version of the explanations surrounding the intuitive phasor picture was not sufficiently intuitive. In the revised version of the main text, we significantly edit and rearrange these paragraphs to enhance their clarity. Moreover, we provide a thorough yet intuitive explanation for the use of phasors in ellipsometry and their graphical depiction in figure 4. We still believe, that the phasor description enables an intuitive understanding of MCTOS for readers who do not want to follow the relatively long mathematical derivation in the supplemental. The edited section, highlighted with violet text, appears in pages 4-5 of the current version.

For the general readability of the supporting information: The text works reasonably well until equation (S24). But the introduction of the "phasor" is difficult to understand. Behind this lies the actual experimental detection setup with the balanced detection, wave plates, and homodyning. I recommend to add more details here such that the reader can understand that the electric field hitting the detector contains a superposition of the probe field and the generated fields, and that upon squaring (in order to calculate the photon flux as the detector measures) cross terms emerge and eventually give rise to the differential current given by Eqs. (5) and (6) in the main text.

We agree with the reviewer on this point; the previous version of Supplemental 1 contained a gap in the mathematical description around the introduction of the phasors. This also relates to the reviewer's previous comment regarding a lack of motivation in the introduction of phasors in the main text. To make this transition smoother and relate it to the experimental setup, we added two more paragraphs to the text on page 5 of Supplemental 1 starting with the words "The new fields are polarized perpendicular to…". The text explains how the complex phasors in Eq. (S24) -(S26) correspond to the ellipsometer detection scheme for the generated photons.

Next, the supporting information proceeds to show that the detected current can be decomposed into a response function R(Omega) and the signal field A_S(Omega)exp(-i*Omega*t_D). This is a nice result; however, the authors fail completely in discussing this result (this discussion should be in the main text). The point is that if R(Omega) is a function which is much broader than A_S(Omega), then Eqs. (5) and (6) just reduce to a simple Fourier transform taking the signal field back to the time domain. This is the main point of the whole manuscript! And this is also what the authors are doing in Fig. 2(b) (after filtering the DDC component out). But the issue here is that if R(Omega) is not much broarder than A_S(Omega), or is at least a constant in the relevant frequency range of A_S(Omega), the inferred field (exemplified in Fig. 2b) will be a distorted version of the actual field. This point must be discussed carefully and honestly in the main text. I would recommend to show the calculated spectra of R(Omega) in the main text and compare them to the OSA signal spectrum.

The influence of the spectral response function on the MCTOS output is indeed an important point for discussion. As the reviewer points out, the spectral response affects not only the detected spectrum, but also the time-domain transient, as they are directly connected via a Fourier transform. Even though the previous version of the paper included the MCTOS response function (magenta line in Fig. 3a) alongside the signal and probe spectrum, it failed to fully explain the consequence of $R(\Omega)$. Following the reviewer's recommendation, we added a direct remark about the effect of $R(\Omega)$ on the MCTOS detected transient as part of a comprehensive discussion regarding the response function that was motivated by the comments in this review.

2) General applicability and limitations.

In continuation of the above discussion of R(Omega), it would be useful to provide in the main text a discussion on what setting one should actually use to get the detection scheme to work best. Which choice of band pass filter (BPF) is the optimal one and why?

Following the response for the previous comment, a discussion of the spectral response function was added to the main text on pages 5-6 and Section 6 of Supplemental 1. In short, a response function with a broader bandwidth is achieved when spectrally filtering the probe at either its high- or low-energy edge (Fig. S3a in Supplemental 1). An example of this effect is shown in Fig. 5 - the detected signal spectrum is significantly broader when a bandpass filter is set at the edge of the probe spectrum (Fig. 5 (b) and (d)) rather than at its center (Fig. 5 (f)).

We believe that the additional analysis both in the main text and supplemental provides the necessary information for future users to incorporate MCTOS in their experiments. In fact, the response function in a specific experimental setup can be straightforwardly calculated using Eqs. (S32) and (S33) of Supplemental 1.

Is it a necessity that the signal field does not overlap with the probe field in frequency space (as is the case shown in Fig. 3)? If yes, what limitations will this present?

The point mentioned by the reviewer, considering the spectral overlap between signal and probe, is certainly important and was also mentioned by reviewer #2. Please refer to the response given above (first comment of reviewer #2) or to the additional text in page 6 of the main text.

Will it be possible to achieve a situation where the R(Omega) function in Eqs. (5) and (6) can actually be taken as a constant such that the result exemplified in Fig. 2b becomes the true electric field?

A flat response spectrum ($R(\Omega)$) is achieved over a band surrounding the central frequency of the probe field. As shown in Section 6 of Supplemental 1, added to the revised version, using a bandpass filter after the nonlinear interaction further broadens and flattens the spectrum of $R(\Omega)$. In the given numerical example, a flat response is achieved over a range of roughly 100 THz (200-300 THz).

Would it be possible to calculate R(Omega) [from S28-S30 based on measured values of A_P(omega) by e.g. an OSA] to a level of accuracy such that the distorion could be compensated for? Evidently, this is not possible for the signal field shown in Fig. 2a and 3a (being too much on the edge of sensitivity), but it might be useful to discuss it more generally in connection to what strategy the reader should pick if he/she would like to apply the technique.

We believe that the reviewer's idea for compensating the distortion due to the spectral response function, $R(\Omega)$, should generally work quite well. While we do not follow precisely this procedure, our results indicate such a correction can be implemented over a spectral band with a sufficient signal-to-noise ratio.

As proposed by the reviewer, we calculate the spectral response function based on the probe spectrum, as measured with an OSA (see Fig. 3a). We then numerically calculate the expected MCTOS output spectrum – the product of the signal spectrum measured with an OSA (Fig. 3a) and the calculated response function. The comparison with the measurements, shown in Fig. 5 of the main text for three different bandpass filters, demonstrates an excellent agreement between numerical and experimental results. Thus, the opposite process, a reconstruction of the original signal by dividing the measured spectrum with a calculated response function should also perform reliably.

The authors mention that the signal pulse does not have an absolutely stabilized CEP. But they should explain in the main text why, and under what conditions, the technique works anyway. It seems that (Delta phi_S^0) and (Delta Phi_P^0) are assumed to share the same phase fluctuations, which is probably fair when they are derived from the same master laser.

The reviewer is correct in stating that MCTOS relies on identical CEP fluctuations for both signal and probe pulses. In the revised version of the paper, we added a sentence on page 4 to highlight that this is indeed the case in our setup. As mentioned in the question, CEP locking between the signal and probe is achieved here since both are derived from the same oscillator. We note that a relative CEP stability is a minimal condition for all 'optical oscilloscope' techniques, whereas some techniques require even an absolute CEP stability for both signal and probe pulses.

Along the same lines, the assumptions behind Eqs. (S20) and (S21) should also be commented on. Will the technique only work for bandwidth-limited pulses?

Before addressing this point, we emphasize that we only assume bandwidth-limited pulses for the probe pulse while the spectral phase of the signal is not constrained [$\phi_S^{ho}(\Omega)$ in Eq. (S20)].

In our analysis, the phase of the probe is assumed to be constant $\Phi_P^{ho}(\Omega) = 0$ [see Eq. (S19)]. This is supported by the FROG characterization of the probe beam, added to Sec. 1 of Supplemental 1. In principle, MCTOS also works for non-bandwidth-limited probe pulses, but using a chirped probe would result in modification of the spectral response function (see added Fig. S3b in the Supplemental 1) and the detected transient. After characterization, the additional spectral phase of the chirped probe can be included in the analytical model (see Sec. 6 of Supplemental 1) to arrive at the spectral response function of MCTOS (similarly to the above discussion regarding the distortion of the transient). In the revised version, this point is addressed within the discussion regarding the spectral response of MCTOS (page 6 of the main text).

Finally, we note that the issue of the probe's spectral phase pertains not only to MCTOS but rather to all 'optical oscilloscope' methods (see, e.g. K. T. Kim et al., Nat. Photon., 7, 958-962 (2013) and M. R. Bionta et al., Nat. Photon., 15, 456-460 (2021)).

The phase matching conditions [Eqs. S16-S19] are never really discussed. Do the have any practical impact on R(Omega) defined in (S33) and (S34)? It there anything the reader should take into account if designing a similar experiment?

We thank the reviewer for bringing the missing information to our attention. In the current version, it is given in page 6 (Sec. 5) of Supplemental 1. Naturally, the phase-matching conditions should be thoroughly considered in MCTOS. As for all nonlinear processes, the chromatic dispersion in the nonlinear medium can limit the interaction bandwidth. A mathematical treatment of this aspect is included in the spectral response function as the phase-matching factors [Eq. (S15)]. In the case of a thick detection crystal, the phase-mismatch will modulate the spectral response function. When implementing MCTOS, this can be circumvented by either phase matching with a birefringent crystal or by using a thin detection medium. In our work, we relied on the latter option, employing 16 µm-thick Si and 25 µm-thick GaSe crystals.

3) Minor things:

On page 2, it would be useful to state also the central wavelengths corresponding to the 156 THZ and 246 THz fields.

We added the corresponding wavelengths.

There seems to be a discrepancy between Fig. 3 of the main text and Eqs. (S13-S15). The UC process seems fine: There are two absorptions of the probe field (green arrows pointing upward and E_P has no asterisk in S13) and one emission of a signal photon (red arrow going down and an asterisk on E_S). However, for the DC process we have two downward green arrows (emission) but only an asterisk on one probe field in (S14). In general, figure 3 of the main text is very confusing. There are three frequencies at play from the probe field [e.g., omega, omega1, and omega-(omega1-Omega)], but there are only two colors (blue and green) or arrow lengths at play in the figure. Furthermore, it is not clear what is the significance of the energy gain/loss represented by the black arrows. There is energy conservation in the basic interaction for each frequency setting within the integrals in S13-S15.

Following this comment, we realized that Fig. 3 of the main text can be difficult to interpret. Therefore, we have added a sentence in page 3 that details the role of different arrows:

“We divide the four-wave mixing interaction into three distinguishable processes which contribute to the output. These are depicted in Fig. 3b in a standard arrow scheme; the leftmost arrow is the probe input whereas rightmost arrow stands for the nonlinearly generated probe output.”

With regards to the specific comments: The color of an arrow represents its energy (or frequency). MCTOS, being a four-wave-mixing process, generally involves four frequencies. A signal photon (red in Fig. 3), a higher-frequency probe (blue) and two lower-frequency probe photons (green). It is true that the use of identical colors for the two lower-frequency photons can seem misleading as they do not necessarily have the same energy. However, since they are generally quite close in wavelength and in order to avoid confusion with the addition of further colors, they are represented with the same green shade in Fig. 3b.

As for the comparison of Fig. 3 of the main text (pasted below for convenience) with the analytical treatment in Supplemental 1, we appreciate the reviewer’s attention to details, but after careful consideration maintain that the equations are correct. As mentioned in the added text and following the standard practice in such a representation, the first three arrows from the left represent the incoming waves and correspond to the fields on the right side of Eqs. (S12) -(S14). The fourth (rightmost) arrow represents the emerging field (E_{DC}, E_{DC} and E_{DDC}) in these equations.

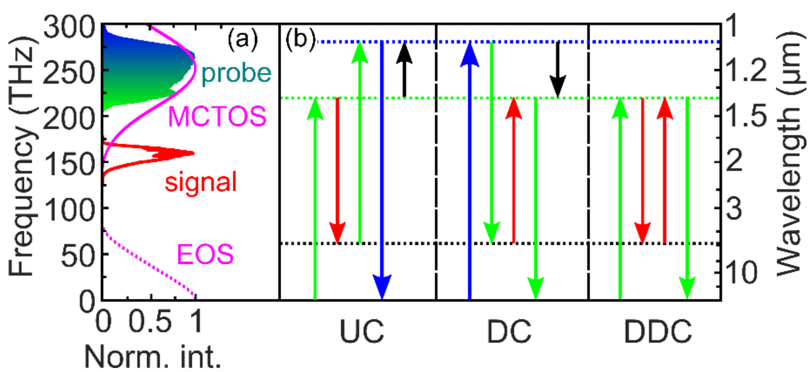

Fig. 3 Third-order nonlinear processes contributing to MCTOS. a Normalized spectral intensities of the probe (turquoise color gradient) and signal (red) pulses. The solid and dashed magenta lines represent the spectral response function of the probe pulse for MCTOS and EOS, respectively. **b** Third-order nonlinear interactions. The length and color of the arrows illustrate the energy of the participating photons. The black arrows represent the effective frequency change of the initial probe photon. UC/DC: up-/downconversion. DDC: direct downconversion.

For the DC process, for example, the input fields are composed of two probe and one signal frequencies. The high-frequency probe and the signal photons are annihilated and are therefore plotted as upward-pointing arrows. The third input field stimulates the emission of a lower-energy probe photon (green) and is therefore plotted as a downward-pointing arrow. As for the output, the newly generated field component is depicted as the rightmost arrow (green) that points downwards to represent the emission of a probe photon. In the right-hand side of Eq. (S13), the three input fields appear as $E_P[\omega + (\omega_1 - \Omega)]$, $E_P^*(\omega_1)$ and $E_S(\Omega)$ where a complex conjugate of the field is taken for the emitted photon.

In this scheme, the black arrows represent the energy difference between the leftmost (input) and rightmost (output) probe photons. An upward- (downward-) pointing arrow depicts energy gain (loss) in an upconversion (downconversion) process.

In the methods section the balanced photodiode is described. It would be useful to add that this is an InGaAs detector, or alternatively, to state the working wavelength range of 800-1700 nm.

This is indeed helpful information; we added it in the methods section.

Now we turn to the supporting information:

In the first line of section 2, there is a reference to "Sec. 4 of the main paper". But there is no section numbering in the main paper.

We thank the reviewer for noticing this point and corrected it in the revised version.

Please state in connection to Eq. (S2) that this is valid for a non-depleted field. It would also be useful to clarify the proper ansatz of the generated electric field E, which is allowed to vary as a function of z (just so the reader does not mistakenly take E to be on the form of S2).

As the reviewer noticed, the previous version of Supplemental 1 was inaccurate when introducing the general form of the solution to the nonlinear wave equation. We thank the reviewer for pointing this out. The amended Eq. (S2) -(S6) reflect the z dependence of the electric-field amplitude and phase (apart from the trivial oscillating wave). The results from Eq. (S7) and onwards, relying on the undepleted beam approximation, remain unchanged.

Please mention why S6 is valid in the present experiment.

Eq. (S5) expresses the slowly varying envelope approximation. We note that when phase-matching conditions are approximately fulfilled (as assured by the choice of a thin detection crystal), the phase mismatch term ξ can be expanded according to $\exp(i\Delta kz) \sim 1 + i\Delta kz + \frac{(i\Delta kz)^2}{2}$. In the case of undepleted beams this leads to

$$\frac{d^2E_p}{dz^2} = \frac{dE_p}{dz} \cdot i\Delta k.$$

To a good approximation, the condition in Eq. S6 becomes

$$\Delta k \ll k_p$$

which strongly holds in the case of our experiments. For example, considering four-wave mixing in the GaSe crystal and considering characteristic signal and probe frequencies we obtain $\frac{\Delta k}{k_p} \approx 1\%$.

Equations (S10-S12) are somewhat confusing. At first glance, the reader does not understand the significance of the special definitions of omega_UC, omega_DC and omega_DDC. In fact, they are not more special/significant than omega_1. I would recommend to instead state carefully the correspondence between (S13-S15) and (a correct version of) figure 3 in the main text and argue what sum rule of frequencies apply to each equation.

Indeed, the correspondence between Eqs. (S9) -(S11) and Fig. 3b is nontrivial. To assist the reader, we added a paragraph in page 4 of Supplemental 1 explaining this relation. The added text also helps in understanding the relation between Eqs. (S12) -(S14) and Fig. 3b.

It is mentioned that the special form of (S20) and (S21) is valid for a bandwidth-limited pulse. Adding a reference (for instance to a text book) would be useful.

The expression in Eq. (S19) for a bandwidth-limited pulse can be found, e.g., in Eq. 3.10 of Weiner, Ultrafast Optics, Wiley 2009. For clarification, we added this reference to the supplemental.

It seems to me that there should not be an asterisk on k and \xi in (S28). Check also Eq. (S33).

We thank the reviewer for drawing our attention to Eq. (S32); a missing asterisk was added in the revised version. However, the presence of complex conjugation on k and ξ in Eq. (S27) is correct. This is delicate mathematical issue that originates in the expression in Eq. (S30). The signal is proportional to the imaginary part of the sum of the two phasors. However, the exponents in the two phasors have opposite signs.

In order to simplify the expression, we note that generally

$$\mathrm{Im}[P_{DC}(t_D) + P_{UC}(t_D)] = \mathrm{Im}[P_{DC}(t_D) - P_{UC}^{*}(t_D)],$$

where now both terms of the sum share the same exponential factor. The complex conjugation of P_{UC} results in a mutual exponential term for the two phasors. This enables us to finally express the MCTOS output as a Fourier transform of the signal spectrum. A byproduct of this simplification is that k and ξ are also conjugated in the response function for UC [Eqs. (S27) and (S32)].

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed my comments satisfactorily, and I am in support of their publication.

Reviewer #2 (Remarks to the Author):

The authors addressed all my comments in the new revision. I recommend the current version for publication.

Reviewer #3 (Remarks to the Author):

I believe that the authors have addressed the concerns of all three reviewers satisfactorily, and the present manuscript is essentially acceptable for publications. I have some minor remarks below, which I recommend the authors to consider. This can be handled between the authors and the editors without another round of review.

In connection with the description of arrows in Fig. 3, the authors write that the "rightmost" arrow stands for the generated probe. But this should be the fourth arrow (the rightmost one is the black arrow).

While Eq. (1) is a suitable representation of the more accurate (S21), there seems to be an error when comparing (2) to (S23). The phase factor is missing, and the frequencies do also not seem correct.

5 lines below Eq. (2): field -> fields.

There seems to be some "asymmetry" in presenting only E_UC and E_DDC in Eq. (1) and (2) [i.e. leaving out E_DC], while at the same time, all three relevant fields are present in (3-5).

The authors use "norm square", |...|^2, in Eqs. (2) and (5). This gives a risk that the reader will interpret these amplitudes as complex valued.

The phase parameter "\Delta\Phi" is not defined in Fig. 4b and 4d. Maybe do this in the figure caption.

The addition of section 6 in the Supplemental is important. I recommend to add a reference to this in the last paragraph on page 4. For instance: The FT of the MCTOS output provides a direct measurement of the spectral amplitude and phase of the signal pulse within the bandwidth offered by he response functions, see Section 6 of Supplemental 1.

Below Eq. (7). Shouldn't the Taylor expansion of \phi_S^ho be carried out in terms of powers of (\Omega-\Omega_0), where \Omega_0 is the center frequency of the signal field? Note, this equation is also present in the Supplemental.

Response to reviewers. Second round of revision for "Few-femtosecond phase-sensitive detection of infrared electric fields with a third-order nonlinearity" in Communications Physics.

Reviewer #1 (Remarks to the Author):

The authors have addressed my comments satisfactorily, and I am in support of their publication.

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In connection with the description of arrows in Fig. 3, the authors write that the "rightmost" arrow stands for the generated probe. But this should be the fourth arrow (the rightmost one is the black arrow).
We implemented the changes suggested by the reviewer.

While Eq. (1) is a suitable representation of the more accurate (S21), there seems to be an error when comparing (2) to (S23). The phase factor is missing, and the frequencies do also not seem correct.
We thank the referee for pointing this out and added the missing phase factor in Eq. (2). However, we retain the frequencies as are. We note that in Eq. (2) of the main text we treat the quasi-monochromatic case in which we consider only one signal frequency Ω.

5 lines below Eq. (2): field -> fields.
We believe that the singular form is correct here. Perhaps the editorial team can advise.

There seems to be some "asymmetry" in presenting only E_UC and E_DDC in Eq. (1) and (2) [i.e. leaving out E_DC], while at the same time, all three relevant fields are present in (3-5).
The reviewer is correct that there is a certain asymmetry. However, in order not to overload the reader with equations, we wanted to include only the most important ones in the main text. In our opinion, Eqs. (3)-(5) are crucial to the qualitative understanding of MCTOS (with the help of Fig. 4 in the main text), and thus we provided the expressions for all three processes. Eqs. (1) and (2) help to understand the derivation of Eqs. (3)-(5). In comparison, the omitted equation for E_{DC} is quite analogous to that of E_{UC} and does not significantly contribute to the clarity of the topic.

Small text edits

- The authors use "norm square", |...|^2, in Eqs. (2) and (5). This gives a risk that the reader will interpret these amplitudes as complex valued.
- The phase parameter "\Delta\Phi" is not defined in Fig. 4b and 4d. Maybe do this in the figure caption.
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- Below Eq. (7). Shouldn't the Taylor expansion of \phi_S^ho be carried out in terms of powers of (\Omega-\Omega_0), where \Omega_0 is the center frequency of the signal field? Note, this equation is also present in the Supplemental.

We thank the referee for mentioning these points which we adapted in the revised version.