Enhancing terahertz generation from a two-color plasma using OPA waste light

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We show experimentally that the terahertz (THz) emission of a plasma, generated in air by a two-color laser pulse (containing a near IR frequency and its second harmonic), can be enhanced by the addition of an 800-nm pulse. We observed enhancements of the THz electric field by a factor of up to 30, at some input pulse energies. This provides a widely accessible means for researchers using optical parametric amplifiers (OPA) to increase their THz yields by simply adding the residual pump beam of the OPA to the plasma generating beam. We investigate the dependence of the THz electric field enhancement factor on the powers of the two-color beam as well as the 800 nm enhancement beam. Numerical calculations using the well-known photocurrent model are in excellent agreement with the experimental observations.

The emerging applications of THz radiation in a wide array of technologies including chemical sensing and non-destructive material and biomedical imaging [1,2], as well as in fundamental materials research [3,4], make the efficient generation of high-field THz radiation an ever-increasing need. Especially desirable are table-top THz sources that would be widely accessible to researchers. In addition to the more traditional techniques of photoconductive switching [5] and optical rectification in nonlinear crystals [6,7], plasma generation is also arising as a promising route to intense THz generation. In short, this method typically involves combining an ultrafast laser pulse with its second harmonic in a tight focus to generate a plasma from which THz radiation is emitted [8,9]. The THz radiation emitted from a two-color plasma is particularly attractive for its continuous, broadband spectral characteristics spanning from 0.1 THz to as high as 200 THz, depending on pulse duration [10]. Recent work by Kim and coworkers has shown that this emission is explained by a transient photocurrent model, wherein the nonlinearity of THz generation is due to the exponential nature of photoionization [11,12].

Having established the two-color filament generation technique, many researchers are now investigating ways to optimize or improve the THz output [14,17]. Of particular relevance to this work, several researchers have explored the idea of using additional pulses prior to the two-color pulse in order to introduce a preexisting plasma and alter the THz emission [1] [19,21]. In another vein of investigation, Clerici et al. recently showed that THz yields are drastically improved at longer pump wavelengths, so much that the losses incurred during conversion in an OPA to longer wavelengths are more than made up for in the THz generation [10]. Additional theoretical work has since been reported supporting these measurements of wavelength scaling [22]. Optimal tabletop plasma THz generation is therefore likely performed using an OPA, however, the residual pump beam to the OPA is typically not used. This letter discusses a practical and value-adding use for this often wasted light in enhancing the THz yield of a two-color plasma.

All reports involving the use of an additional enhancement pulse that we are aware of use an 800 nm beam, with its second harmonic, as both the prepulse and the two-color plasma generation pulse. In one of these reports, Minami et al. showed that a prepulse had the clear effect of suppressing the THz generation of the plasma generation pulse; however the THz yield when the prepulse and the plasma generation pulse were overlapped in time was not analyzed in detail [21]. Xie et al. offered some evidence that a prepulse may enhance THz emission, though, in their experiments they used an 800 nm pump and they did not use BBO for the more efficient two-color plasma generation of THz [20]. Furthermore, their observation of a long-lived enhancement seem to directly contradict the other works showing a suppression at longer prepulse delays [1,19,21]. Therefore, to build on the existing work it is expedient to investigate the enhancement of the more efficient and relevant two-color generation scheme using a longer pump wavelength and a BBO crystal to generate the second harmonic. In addition, a rigorous investigation should use an enhancement pulse that is not capable of efficiently generating THz radiation on its own, in order to avoid interference effects when the pulses are overlapped in time.

In this letter, we report on measurements showing that the THz emission of a two-color plasma is significantly enhanced, by as much as 30 times, when an additional 800-nm pulse is added to the plasma. Our experimental observations are corroborated by the well-known photocurrent model.

Terahertz radiation was generated using the standard two-color filamentation technique, as described previously [2] [8], and shown in Fig. 1. Briefly, the near-IR output of an OPA (centered at 1450 nm with a pulse duration of about 100 fs) is passed through a BBO frequency doubling crystal and focused to form a plasma in dry air (< 2% relative humidity). The emission of the
800 nm Probe

FIG. 1. A schematic of the experimental apparatus. The near IR output of an OPA is combined with the residual 800 nm pump of the OPA and focused in dry air to generate a plasma. The THz emission of the plasma is collected and its electric field is measured using EO sampling on GaP. The following optics are indicated: A. λ/2 waveplate, B. variable ND filter, C. polarizer pair, D. BBO crystal, E. GaP crystal, F. λ/4 waveplate, G. Polarizing beamsplitter, H. Balanced photodiode.

plasma is collected and passed through a Teflon filter, which rejects visible and near-IR frequencies and passes THz frequencies. The THz electric field is measured using electro-optic sampling in 100 μm GaP [23, 24].

Initially, the horizontally-polarized THz emission of the two-color plasma was optimized without the enhancement beam by adjusting the polarization of the two-color pulse and the BBO phase matching angle and angle of incidence (which effectively tunes the phase relationship between the fundamental and the second harmonic). The optimal polarization of the near-IR pulse was 30° from horizontal. The residual 800-nm beam from the second stage of a commercial OPA was used as the enhancement pulse, and we note that pumping the OPA results in a depleted beam with a non-gaussian spatial profile. This enhancement pulse was collinearly added to the two-color plasma and its alignment and polarization were optimized for maximum enhancement. The measured enhancements, as well as the optimum polarization of the enhancement beam were very sensitive to variations in alignment. The likely optimum polarization for the enhancement beam was, nevertheless, found to be horizontal. The enhancement pulse was set to a series of delays, relative to the two-color pulse, and a full measurement of the generated THz waveform was taken at each delay, averaging each point over 150 laser shots. This was done for several combinations of powers of the enhancement and two-color beams, as well as for two polarizations of the enhancement beam. THz yields for each measurement were calculated by integrating the absolute value of the complex Fourier transform of each time trace (note that the electric field strength scales as the spectral amplitude).

The critical finding of these measurements is that the enhancement pulse, at a specific delay relative to the two-color pulse, enhances the THz emission of the filament by as much as 30 times. Our measurements also confirm that for significant negative delays (the enhancement pulse arrives before the two-color pulse) the THz yield is suppressed significantly, as demonstrated previously for the case where both the two-color pulse and the enhancement pulse were centered at 800 nm [1, 19, 21]. Both of these findings are illustrated by plotting the THz yield of the filament as a function of the delay of the enhancement pulse relative to the two-color pulse. For convenience, the THz yields can be considered relative to the yield in the absence of the enhancement pulse - we will call this the electric field enhancement factor. Fig. 2 shows plots of the electric field enhancement factor as a function of enhancement pulse delay for several two-color and enhancement pulse energies. Each panel in Fig. 2 shows a different two-color pulse energy, while the different shades within a single panel represent varying enhancement pulse energies. Experimental measurements (represented by square symbols) are shown alongside model calculations (solid line) that are described in detail in the supplemental material [27]. It is emphasized that each experimental point on these traces represents a full measurement of a THz waveform, i.e. for a given enhancement pulse delay: the full THz waveform is measured, the Fourier transform is calculated and a THz yield is calculated as the integral of these spectra. Finally this THz yield is divided by the THz yield in the absence of the enhancement pulse to give a single point on the plot of electric field enhancement factors. Representative traces of the actual measured THz waveforms and their Fourier transforms are contained in the supplemental material [27].

Negative enhancement pulse delays in Fig. 2 correspond to the enhancement pulse arriving and generating the plasma before the two-color pulse. It is easily seen that at significant negative delays, the THz yield is substantially lower, or suppressed, relative to the yield of the two-color pulse alone. On the other hand, at significant positive delays, the enhancement pulse arrives at the plasma after the two-color pulse and the THz yields are equal to the yield of the two-color pulses alone. For some combinations of input pulse energies, a significant increase of the THz yield is also seen in Fig. 2 at a small positive delay around 75 fs.

A few important observations can be made concerning the electric field enhancement measurements: 1) At the lowest two-color pulse energy we studied, the THz emission is enhanced by as much as 30 times. 2) This enhancement decreases with increasing two-color pulse energy, asymptotically approaching a value of 1 (corresponding to no enhancement) at two-color pulse energies around 600 μJ. 3) For any given two-color pulse energy, the enhancement appears to saturate at high enhancement pulse energies. Furthermore, the onset of this satu-
Suppression of the THz yield at large negative delays and no significant increase in THz yield is observed. 4) Saturation of the plasma itself since the number density of the gas is finite.

In order to explain these observations, we turn to the transient photocurrent model, which is presented in detail in Refs. 2 and 13 and has been met with considerable success in describing the emission of two-color plasmas [3, 22, 23]. The essence of the model is to consider the photo-acceleration of electrons liberated by tunneling ionization. Details regarding our implementation of this model can be found in the supplemental materials [27]. The results of our calculations are shown alongside the measurements in Fig. 2.

Our results agree well with earlier reports showing a long-lived suppression of THz emission when the two-color filament is delayed with respect to a prepulse. These measurements suggest that this suppression is roughly linear with respect to prepulse energy (across a large range of experimentally relevant pulse energies) and irrespective of the two-color pulse energy. The transient photocurrent model appears to completely capture this suppression and its dependencies on pulse energies, as can be seen at large negative delays in Fig. 2. Considering the need for larger THz electric fields when the two-color filament is delayed with respect to a prepulse, we find it more interesting to note that the model also captures the maximum enhancement expected for a given combination of prepulse and two-color pulse energies. This can be seen for each of the curves in Fig. 2 around a delay of 75 fs, where the maxima of the experiment and the model agree well. The experiment and model also show that the dependence of the maximum enhancement on prepulse energy appears linear up to a point where it then saturates and no further increase in THz emission can be achieved. The model suggests that this is due to saturation of the plasma itself since the number density of the gas is finite.

In addition to capturing the suppression and enhancement of THz emission and their dependencies on both prepulse and two-color pulse energies, the model also captures some finer details observed experimentally. For example the asymmetry in the plots corresponding to 160 µJ and 240 µJ in Fig. 2 appears to agree quite well between model and experiment. Two features observed experimentally that are not captured well by the model are the onset of suppression before the enhancement (especially seen for higher two-color pulse energies in the bottom of Fig. 2) and the shape of the enhancement plots for the higher two-color pulse energies in the range: 0 < ∆ < 150 fs. The experimental measurements show a small level of suppression or enhancement before the main features captured by the model - one likely explanation for this discrepancy is that there may be undesired pulses present in the experimental pulse train due to double reflections within the optical setup. The ex-

![Graph](image-url)
cess suppression observed at small negative delays also has a possible explanation. Our method of performing an equally weighted average over a full cycle of relative prepulse phases in the model calculations is roughly the equivalent of assuming a phase instability > 2π in our experimental apparatus on the time scale of ~1 sec. This is perhaps an overestimate of the instability and the model does show pronounced sensitivity to prepulse phase in that exact range of prepulse delays [27].

Having established the veracity of the model calculations described, we can now use it to present a more complete and continuous picture of the observed THz enhancement. Fig. 3 shows the maximum expected THz yield, based on the model, as a function of two-color pulse energy for several different enhancement pulse energies. It is clearly seen how the addition of the 800 nm enhancement pulse can significantly enhance the THz emission of a filament, especially in cases where the fluence of the two-color pulse is relatively low. A saturation of the THz output is also clearly seen where no further increases are obtained beyond about 600 µJ of prepulse energy. An important conclusion drawn from our results is that this saturation level depends on the number density of the neutral gas and the total optical fluence (the fluence of the two-color pulse plus that of the enhancement pulse).

In our current experimental configuration, the saturation prevents us from achieving substantial improvements in the absolute THz yield. Bulk plasma effects such as the plasma opacity and dispersion are not included in our model, nor have we scaled the absolute THz yield with the volume of plasma. We anticipate that considering the complex interplay between optical fluence, plasma volume and the THz emission, there is likely a range of pulse energies and focal spot size that would allow for a significant increase in absolute THz yields.

The critical conclusion from our measurements is the demonstrated ability to enhance THz emission from a two-color laser filament by as much as 30 times using traditionally wasted 800-nm light from an OPA. Plasma generation of THz radiation has emerged as an important tabletop THz source, and we anticipate that this simple means of enhancing that emission represents a fundamental advancement of the technique. We emphasize that the enhancement of plasma emission is demonstrated here for the relevant case of a two-color plasma where the fundamental is a longer near-IR wavelength (1450-nm) in order to take advantage of the wavelength scaling of plasma generation [9]. In light of recent work exploring the plasma generation of fractional harmonic frequencies [25, 28], in addition to our own results demonstrating THz generation using non-commensurate wavelengths [14], it is especially interesting that this enhancement does not require a commensurate (or harmonic) frequency of the two-color pulse and is seemingly adaptable to a broad range of plasma generation frequency configurations. A likely best practice for researchers desiring to generate large THz electric fields is to first convert to longer wavelengths using an OPA, in which case a large amount of optical power is typically wasted in the residual OPA pump beam. We now understand, however, that this beam can be used in a very straightforward manner, with minimal cost, to further increase THz output.

Importantly, the enhancement factor saturates at high optical fluences and oftentimes plasma generation of THz radiation is carried out in a regime of fluences where further enhancement can not be achieved. Our results suggest that reducing the optical fluences of the two-color beam and the enhancement beam by increasing their spot sizes in the focal plane (essentially moving to the left in Fig 3) is a promising route to increased THz outputs. This idea is bolstered by recent work showing the scaling up of plasma THz emission by increasing the plasma size [15]. We also suspect that further gains can likely be made by increasing the density of the plasma supporting medium [10]. Further work will be directed to these ends.

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Supplemental Material for:
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THZ WAVEFORM MEASUREMENTS

In order to emphasize the actual measurements made, Fig. S1 shows a selection of representative, measured THz waveforms and their Fourier transforms, with the enhancement beam set to 1930 µJ and the two-color beam set to 240 µJ. Note that the spectra are consistent and no obvious spectral variations are introduced by the overlapping enhancement pulse. Enhancement factors are measured relative to the THz emission of the plasma in the absence of the enhancement pulse. Each square on the plot represents a full measurement of the THz electric field.

FIG. S1. Bottom: The measured (square symbols) and calculated (solid line) enhancement factor of the electric field strength of the plasma-generated THz as a function of delay of the 800 nm enhancement pulse relative to the two-color pulse. The measured waveform, for the indicated points, along with their calculated Fourier amplitudes are shown above.

ENERGY DEPENDENCE OF MAXIMUM ENHANCEMENT

Fig. S2 shows the maximum enhancement factor measured for each of the two-color and enhancement pulse energies. Plotted this way, the saturation at higher total fluences may be easier to visualize.
ALIGNMENT SENSITIVITY

We observed that the electric field enhancements were very dependent on alignment. Despite best efforts to optimally align the enhancement beam for different sets of measurements, there remained some variability in the measured enhancement factors. Along with this variability in achievable enhancement, was a concomitant variability in the optimal polarization of the enhancement beam. Fig. S3 shows the maximum electric field enhancement factor as a function of enhancement pulse energy for three different sets of measurements taken on different days for nominally the same two-color pulse energy of 160 µJ. The solid red line is the same data shown in the main text and the measurements were taken with a horizontally polarized enhancement beam. The two dashed lines were both taken with the enhancement beam polarized 25° relative to the horizontal axis. In all three cases, the pointing and the polarization of the enhancement beam were adjusted to optimize the enhancement factor. Furthermore, in all three cases the main conclusions presented in the main text hold true concerning the magnitudes of the enhancements.
FIG. S3. Measured electric field enhancement factors for nominally the same two-color pulse energy of 160 µJ on different days and presumably with slightly different beam alignment.

POLARIZATION DEPENDENCE OF THE ENHANCED TERAHERTZ RADIATION

As explained above, the measured enhancements, as well as the optimum polarization of the enhancement beam were very sensitive to variations in alignment, however the likely optimum polarization was found to be horizontal. All the measurements shown in the main text were taken using the optimal enhancement beam polarization. Electric field enhancement measurements were additionally taken with the enhancement beam polarization oriented perpendicular to the optimal polarization for a select number of the pulse energy combinations. A representative comparison of the electric field enhancement for the optimal and the perpendicular polarization of the enhancement pulse is shown in Fig. S4. Again, the symbols represent experimental data and the solid lines show model calculations.

FIG. S4. Electric field enhancement factors are shown for two perpendicular polarizations. The measured (squares) and the calculated (lines) are in good agreement. When the enhancement pulse is polarized perpendicular to the two-color field, no enhancement is observed.

We hypothesized that the THz yield for the measurements made with the perpendicular enhancement pulse polarization could be simulated by allowing the prepulse to assist in generating carriers but not accelerate them. In other words, the field driving the tunneling ionization in the perpendicular polarization simulations is identical to that used in the main text including the fundamental, its second harmonic and the 800-nm pulses, but the field used in accelerating the electrons omits the 800-nm pulse contribution. The results of such a calculation are shown in Fig. S4 along with the experimentally measured electric field enhancement factors. The important observations here are
that the enhancement is practically eliminated when the prepulse polarization is rotated to be perpendicular to the experimentally determined optimal polarization. It can also be seen that the suppression of the THz yield at larger negative delays appear to be identical between the two polarizations.

**DETAILS ON MODELING**

In our calculations, we have assumed that the number of free electrons satisfies the equation

$$\frac{dN(t)}{dt} = w(t) \left[ N_g - N(t) \right] - r_r N(t), \quad (S1)$$

where $N(t)$ is the total number of electrons at time $t$, $N_g$ is the number density of the neutral gas, $r_r = 8 \times 10^9 \, \text{s}^{-1}$ is the rate of recombination estimated from Ref. [S1] and $w(t)$ is the tunneling rate. We used the static tunneling rate

$$w(t) = \frac{\alpha}{E_L(t)} \exp \left( -\frac{\beta}{E_L(t)} \right), \quad (S2)$$

where $\alpha = 4\omega_a^5/2 \cdot \beta = (2/3)r_H^3/2$, $\omega_a = 4.134 \times 10^{16} \, \text{s}^{-1}$ is the atomic frequency unit and $r_H = U_{N_2}/U_H$ is the ionization potential of nitrogen gas (15.6 eV) relative to that of atomic hydrogen (13.6 eV). We also define $E_L(t) = |E(t)|/E_a$ as the absolute value of the electric field of the laser in atomic units ($E_a = 5.14 \times 10^{11} \, \text{V/m}$). We treat the electric field $E(t)$ as a linear combination of Gaussian pulses whose carrier frequencies and pulse widths are chosen to match the experimental conditions. We define the phases and delays relative to the 1450 nm beam, and we assume the phase of the second harmonic, $\theta_{SH} = \pi/2$ in order to optimize THz yields in the simulations [S2]. Tuning the angle of incidence of the BBO in the experiment to optimize the THz output is necessary in order to bring $\theta_{SH}$ close to this optimal value. The amplitude of the electric field envelope for each of the three pulses was calculated based on the experimentally measured powers $P$

$$E_{\text{max}} = \frac{4}{d} \sqrt{\frac{P \sqrt{\ln 2}}{\nu \pi^{3/2} \tau \epsilon_0}}, \quad (S3)$$

where $d$ is the focal spot diameter, $\nu = 1 \, \text{kHz}$ is the repetition rate of our laser, and $\tau = 75 \, \text{fs}$ is the pulse duration. In light of reports showing variability in the shape of a two-color plasma with changing pulse energies [S3, S4] we varied the spot diameters in the calculations. The delay $\Delta$ between the enhancement pulse and the two-color pulse pair is determined experimentally and the phase of the enhancement pulse $\theta_{\text{enh}}$ was averaged in the simulations considering the distinct lack of phase stability in the experiments (further discussion on the effects of $\theta_{\text{enh}}$ can be found below).

Assuming that there are no free electrons in the gas before the pulses arrive (i.e. $N(t_0) = 0$), then the solution to Eq. (S1) can be calculated as

$$N(t) = \exp \left[ -r_r (t - t_0) - \int_{t_0}^{t} dt' w(t') \right]$$

$$\times N_g \int_{t_0}^{t} dt' w(t') \exp \left[ r_r (t' - t_0) + \int_{t_0}^{t'} dt'' w(t'') \right] \quad (S4)$$

The photo-emission of the plasma $E_{\text{emit}}(t)$, was then calculated as

$$E_{\text{emit}} \propto \frac{e^2}{m_e} E(t) N(t), \quad (S5)$$

where $e$ and $m_e$ are the electron charge and mass, respectively. As was done for the experimental measurements, THz yields were calculated from the simulated electric field time traces as the integral (from 0.1 to 10 THz) of the absolute value of the Fourier transform. The results of these calculations are shown alongside the experimental measurements for all the power combinations in Fig. 2 of the main text.
SPOT SIZE VARIATION

The beam diameter (and correspondingly, the diameter of the plasma) was expected to be close to 100 µm for all of the beams, however it was left as an adjustable parameter in light of the observation that filament size is distinctly dependent on the energy density of the laser pulses [S3, S4]. In order to approximate the diameter of the plasma we performed a number of THz yield calculations using the plasma current model discussed in the paper over a range of enhancement and two-color beam fluences. Since our model does not account for bulk plasma characteristics, the THz spot size only enters the model in calculating the electric field magnitudes from the pulse energies. We can therefore use fluences in the model and avoid the explicit inclusion of the spot size.

The calculations showed that the THz yield at significant negative enhancement pulse delays (where there is pronounced suppression of the THz yield) was independent of the two-color fluence. This is corroborated by the experimental observations that are summarized in Fig. S5 that shows the measured dependence of the suppression of THz emission as a function of enhancement pulse delay for the various two-color pulse energies. We could plot a calculated curve of maximum suppression (or in other words minimum enhancement) vs. enhancement beam fluence to serve as a calibration curve where we look up the experimentally observed maximum suppression and find the corresponding enhancement beam fluence. This procedure using the suppression was critical in allowing us to separate the two pulse energies in correlating the calculations to the measurements. The procedure is illustrated in Fig. S6.

FIG. S5. The Experimentally measured electric field enhancement factor at a significant negative delay as a function of the enhancement pulse energy. Of particular importance in the context of this section is that the shape and the magnitudes of the enhancement factors are the same for all of the two-color pulse energies. The 160 µJ trace is an obvious outlier, because at lower enhancement pulse energies the measurements of the THz waveforms are approaching the noise floor of the electro-optic sampling apparatus.
FIG. S6. Calculated enhancement factors at significant negative enhancement pulse delays were independent of the two-color fluence. The curve shown here is true for all possible two-color fluences (within an experimentally conceivable range). We were therefore able to match the measured suppression at significant negative delays and look those up on this curve to determine the enhancement beam fluence.

Having determined the enhancement beam fluence, we were then able to determine a two-color beam fluence by again looking up the experimentally observed maximum electric field enhancement factor on the calculated electric field enhancement factor surface. The maximum enhancement factor is a function of both the enhancement and two-color beam fluences, thus the calculated maximum enhancement is a two-dimensional surface. Once the enhancement beam fluence was determined from the suppression, the experimentally observed maximum enhancement could be matched to a single point on this surface and both the beam fluences independently determined. Fig. S7 may assist in visualizing this process of determining the beam fluences using the maximum enhancement factor.

FIG. S7. The maximum enhancement factor is a function of both the enhancement pulse and the two-color pulse energies. Knowing the enhancement pulse energy from the suppression (obtained from Fig. S6) allowed us to look on this surface, along the correct enhancement beam fluence, for the two-color beam fluence that would match the observed maximum enhancement.

Finally we are able to determine spot sizes for both the enhancement and two-color beams since we have both a fluence (from matching calculation to experiment) and a measured pulse energy. We find remarkable consistency of these calculated spot sizes across the multiple measurements we performed. The results presented in the letter include 5 measurements at each IR power and 6 measurements at each enhancement power. Using these multiple
measurements, we calculate a mean and standard deviation and plot the dependence of this theoretical spot size on pulse energy in Figs. S8 and S9. It should be mentioned that these spot sizes and the general shape of the spot size curves is consistent with those measured directly in Ref. [S4].

![Image](image_url)

**FIG. S8.** The spot size of the enhancement beam as a function of the enhancement pulse energy determined by matching the calculations to the measurements.

![Image](image_url)

**FIG. S9.** The spot size of the two-color beam as a function of the two-color pulse energy determined by matching the calculations to the measurements.

**PHASE AVERAGING**

Experimentally, the prepulse and two-color beams follow different paths and consequently the prepulse phase $\theta_{pre}$ is not expected to be stable relative to the two-color pulse. For this reason, in our modeling, the THz yield was calculated at 11 different phases between $-\frac{\pi}{2}$ and $\frac{\pi}{2}$ and the reported THz yield was taken as the average of those. This assumption of perfect phase instability may be a little too strong, which may explain the failure of the model calculations in exactly reproducing the measured enhancement factors at delays between 0 and 100 fs. Fig. S10 shows calculated enhancement factor traces, again as a function of delay, but rather than averaging the phase of the enhancement pulse, we have just used a single phase. It is seen how distinct oscillations appear in the plots in
the region of delays between 0 and 100 fs and the phase of these oscillations depends on the chosen phase of the enhancement beam.

FIG. S10. The calculated electric field enhancement factor as a function of delay without averaging the enhancement pulse’s phase. Each line corresponds to the enhancement factor for a single enhancement pulse phase as indicated in the legend. The phase of the second harmonic relative to the fundamental is still fixed at $\pi/2$. The calculations in the main text used the average of an even distribution of several enhancement pulse phases from $-\pi$ to $\pi$.

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