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Generation and measurement of isolated attosecond pulses with enhanced flux using a two colour synthesized laser field

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Abstract: We have generated isolated attosecond pulses and performed attosecond streaking measurements using a two-colour synthesized laser field consisting of a strong near-infrared few-cycle pulse and a weaker multi-cycle pulse centred at 400 nm. An actively stabilized interferometer was used to coherently combine the two pulses. Using attosecond streaking we characterised the electric fields of the two pulses and accurately retrieved the spectrum of the multi-cycle pulse. We demonstrated a two-fold increase in the flux of isolated attosecond pulses produced and show that their duration was minimally affected by the presence of the weaker field due to spectral filtering by a multilayer mirror.

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1. Introduction

Over the last two decades, isolated attosecond pulses, produced in gas phase high harmonic generation (HHG) have become an invaluable tool for the study of electron dynamics in gases [1], molecules [2] and solids [3]. HHG is a remarkable process that has been shown to be extremely sensitive to the waveform of the driving field both experimentally [4] and theoretically with classical [5,6] and quantum mechanical [7] models. With typical HHG conversion efficiencies being only 10^{-5} or lower [8], there is significant interest in tailoring the waveform of the driving field to optimise the process [9,10]. Through enhancing ionization rates at particular times and by altering electron trajectories, engineered waveforms can be used to increase both flux and cutoff energies [11–16] and also provide a method for enhanced gating of isolated attosecond pulses and trajectory selection [17–21]. In recent years, the sub-cycle engineering of a single half-cycle of a pulse has been explored as it has potential to generate shaped isolated attosecond pulses. This can be done by coherent pulse synthesis of several few-cycle [22] or multi-cycle [23], or by a combination of few- and multi-cycle pulses [24].

A key challenge in the use of these synthesized fields is their full amplitude and phase characterization. Most synthesized fields of utility for controlling HHG contain spectral components spanning one octave or more. While conventional measurement techniques such as FROG [25], SPIDER [26] and d-scan [27] have been developed that can characterise ultra-broadband pulses, they typically fall short of being able to characterize a synthesized field because they are insensitive to the phase difference between the component pulses. The dynamic range of the measurement techniques must also be very high as even a very weak additional field can significantly affect HHG [28]. Field sensitive characterisation techniques have developed

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significantly in the last decade, with all optical [29,30] and field ionization [31] based techniques permitting complete characterisation of arbitrary waveforms. The most well established field-resolved characterisation technique available is attosecond streaking, where an isolated attosecond pulse is used to ionize gaseous atoms in the presence of a strong laser field [32,33]. The strong field modulates the momentum of the emitted photoelectrons, thus encoding the optical waveform into the energy shift of the photoelectron spectrum. The key advantage of attosecond streaking over the other methods is that, through use of an iterative algorithm [34], it can simultaneously retrieve the electric field of the optical waveform and the attosecond pulse. This is important since the tailored waveforms used in field synthesis alter the electron trajectories in HHG, potentially increasing the duration of isolated attosecond pulses generated and hence degrading the available time resolution for applications such as attosecond spectroscopy.

In this paper we report results from HHG experiments with a two-colour laser field consisting of a few-cycle (FC) pulse centred at 775 nm and a multi-cycle second harmonic (SH) pulse. We reproduce the previously recorded two-fold flux enhancement of the isolated attosecond pulse [24] and show that it is possible to characterise the generated attosecond pulse and synthesized field using attosecond streaking. Importantly we show that the duration of the attosecond pulse is not increased in this case. We also accurately retrieve the spectrum of the SH pulse, despite it being a factor of 300 less intense than the FC pulse.

2. Experimental setup

We constructed an actively stabilized two-colour interferometer, a diagram of which is shown in Fig. 1. Near-infrared (NIR) pulses with an energy of 1.5 mJ from a commercial, carrier-envelope phase stable, 800 nm, 30 fs, titanium sapphire amplifier (Femtopower HE CEP, Femtolasers) was split using an 80:20 beamsplitter. The reflected 80% of the pulse energy was focussed into a 350 µm diameter, 1 m long hollow core fibre (HCF) which was differentially pumped such that there was 3-3.5 bar of neon gas at the exit and < 2 mbar at the entrance. The output pulse was collimated and compressed with double angle chirped mirrors (Ultrafast Innovations) to generate a sub-4 fs FC pulse. A home-built f-2f interferometer was used to stabilize slow CEP drifts from the amplifier and fibre system, with a residual single-shot fluctuation RMS of better than 250 mrad.

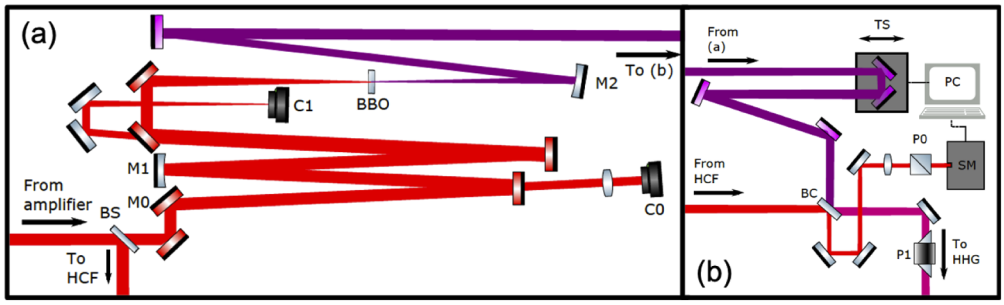

Fig. 1. Schematic diagram of the phase stable two-colour source. A description of the HCF arm can be found in [35]. BS, 80:20 (R:T) beamsplitter; C0 and M0, camera and mirror for beam position stabilization; C1 and M1, camera and mirror for beam pointing stabilization, M1 is also the focusing mirror; BBO, β-Barium Borate crystal; M2, collimating mirror; TS, translation stage assembly; BC, beam combiner; P0, polarizer for spectral interferometry; SM, spectrometer; PC, timing stabilization system; P1, broadband polarizer. Colour of mirrors indicates whether they are protected silver (silver), NIR dielectric (red) or NIR/SH dual band dielectric (purple).

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The transmitted 20% of the initial laser energy was sent through a delay line consisting of NIR dielectric mirrors. The beam alignment was actively stabilised by monitoring the leakage through the mirrors on a CCD camera and using a feedback loop to control motorised mirror mounts (Picomotor, Newport). A concave silver mirror with a focal length of 2 m was used to focus the pulses into a 200 µm thick β-Barium Borate crystal (BBO), cut for Type-I phasematching, to generate the SH pulse. The BBO crystal was placed after the focus and its position was adjusted to achieve a maximum conversion efficiency of 20%, corresponding to 50 µJ at generation. The orthogonally polarised SH pulse had a 9 nm bandwidth, corresponding to a 33 fs Fourier transform limited duration. It was recollimated with an 80 cm focal length concave silver mirror so that the collimated beam size matched that of the FC pulse. The SH pulse and remaining parent NIR pulse were transported using dual band dielectric mirrors to a broadband beam combiner (Layertec) that reflected the SH pulse and transmitted the NIR and FC pulses. In this way, the SH pulse was collinearly combined with the FC pulse, while the residual NIR was transmitted in a direction orthogonal to SH and FC beams.

Spectral interferometry was performed at 800 nm between the two arms using the residual NIR and the ~2% reflection of the FC pulse from the beam combiner. A polariser preceding the spectrometer projected the two pulses onto a common polarisation axis, allowing balancing of the two arms to ensure high spectral fringe contrast for phase-locking. The fringes were recorded with a spectrometer (Qmini Wide VIS, Broadcom) and the extracted phase was used in a feedback loop to control a piezo-actuated delay stage with 38 µm range and 0.25 nm resolution to actively stabilise the relative phase. A phase stability of less than 150 mrad shot-to-shot RMS was readily achieved at the NIR frequency, corresponding to a timing jitter of less than 100 as. For coarse adjustment of the delay between the two arms, the piezo-stage was mounted on top of an additonal motorised stage with 25 mm travel and 1 µm resolution.

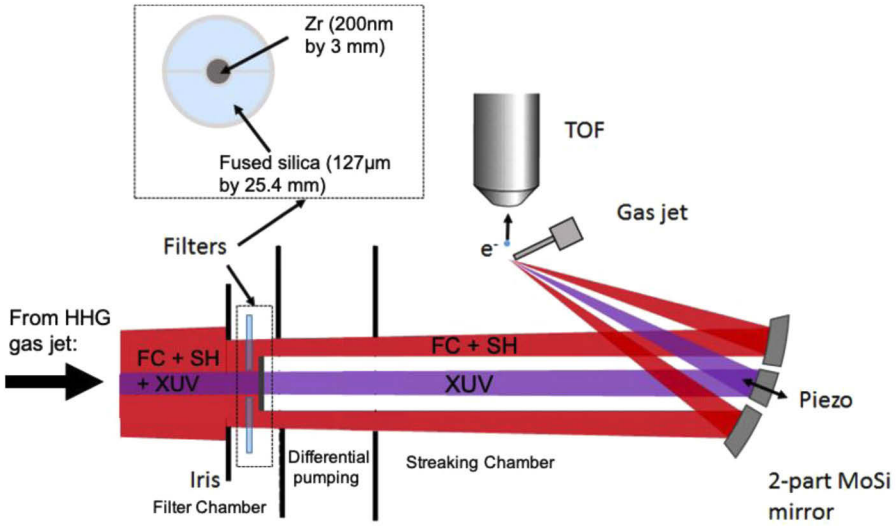

Fig. 2. The attosecond streaking setup. An annular filter consisting of a Zr inner part and fused silica outer part is used to separate the XUV from the FC/SH driving field. The Zr absorbs the optical radiation, transmitting the XUV, while the fused silica absorbs the XUV, leaving the FC/SH. An iris is used to adjust the intensity of the FC/SH. A piezo-actuated two-part MoSi mirror brings the pulses to a common focus in a Ne gas jet placed in the collection region of a electron time-of-flight (TOF) spectrometer. Inset: The annular filter cross section.

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The combined FC and SH pulses were sent through a broadband polarizer (FemtoOptics) to an attosecond beamline [36]. The SH pulse energy was 20 μJ entering the beamline, while the FC pulse energy was 200 μJ. In the beamline the pulses were focused into a 1.8 mm long, 100 mbar Ne gas jet with a 50 cm focal length concave silver mirror. The gas jet pressure and position relative to the focus were optimized for high harmonic generation intensity and short trajectory selection with the FC pulse alone, with the SH pulse blocked before the beam combiner. The spectrum of the harmonics were measured using an XUV flat-field spectrometer and temporally characterised by attosecond streaking. The streaking setup is shown in Fig. 2. The pulses were spatially separated using a two-part annular spectral filter (Fig. 2 inset) made from zirconium (Zr) and fused silica and focussed by a two-part piezo-actuated molybdenum/silicon multilayer (MoSi) mirror (Fraunhofer Inst.). The MoSi mirror acts as an additional 5 eV bandpass filter centred at 93 eV. The pulses were focussed into a second Ne gas jet emanating from a 100 μm inner diameter needle, the pressure of which was tuned to maximise spectrometer signal. A time of flight spectrometer (Stefan Kaesdorf) measured the energy of the streaked photoelectrons as the time delay between the pulses was scanned by the two part mirror.

3. Results and discussion

The two colour field was used for HHG in the extreme ultraviolet (XUV) region in the range 40-110 eV. Figure 3 shows the results of scanning the delay between the FC and SH pulses in the HHG gas jet using the piezo-stage. The modulations in the HHG cutoff and flux, taken at the peak of the MoSi mirror reflectivity curve, show a periodicity that is equal to the frequency of the SH. This flux is modulated by nearly a factor of two by scanning the relative delay, verifying previous results for two-colour HHG near saturation intensities [24]. The cutoff energy is seen to vary by up to 10 eV.

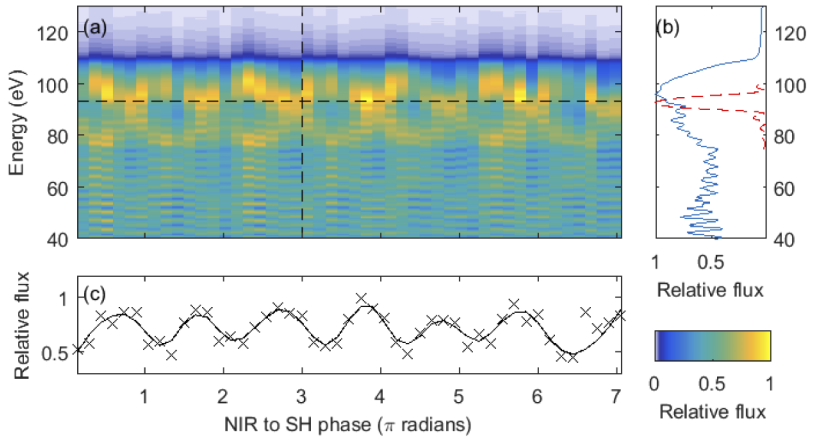

Fig. 3. (a) HHG spectrum generated in Ne with a a two colour field as a function of relative phase between the FC and SH pulses, showing HHG cutoff modulations. (b) Harmonic spectra at phase indicated by dashed black vertical line in (a), with normalized MoSi mirror reflectivity shown in red (c) HHG flux at the 93 eV, the peak reflectivity of the MoSi mirror, showing clear π phase oscillations. (a) and (c) share the same horizontal axis.

The streaked photo-electrons from the Ne 2p state are shown in Fig. 4. In Fig. 4(a) the SH pulse is present, spatially and temporally overlapped with the FC pulse in both the HHG and streaking foci, while it was absent from both the HHG and streaking in 4(c) with the SH interferometer arm blocked before the beam combiner. Figure 4(b) and (d) are the reconstructed traces corresponding to (a) and (c) respectively after FROG-CRAB retrieval using PCGPA [37].

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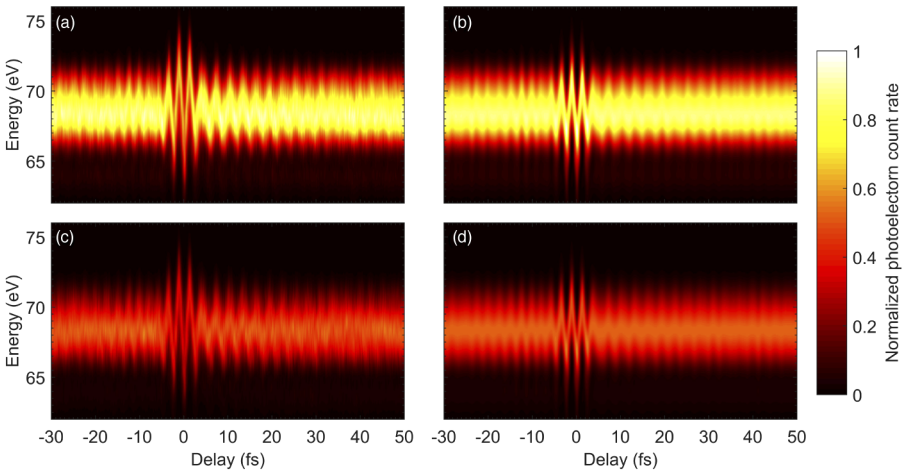

Fig. 4. Experimental streaking traces for XUV pulse generation with the SH pulse present (a) and absent (c) and their corresponding reconstructed traces (b) and (d) respectively.

For Fig. 4(a), the relative phase between the SH and FC pulse was 7.1π radian, corresponding to the next maximum inferred from the periodicity and phase of the scan in Fig. 3. Comparing Fig. 4(a-b) and 4(c-d), a significant change in overall count rate is observed, which is attributable to increased XUV flux from the presence of the second harmonic field, which is consistent with the flux enhancement shown in Fig. 3(c).

The retrieved XUV photoelectron spectra can be seen in Fig. 5. The retrieved XUV pulse durations are 420 ± 20 as and 410 ± 20 as, with and without the SH respectively. The error is based on the estimated 5% retrieval error by Wang et al. for PCGPA for > 50 counts per pixel [38]. This shows that the XUV pulse duration does not change, within the experimental uncertainty, as a result of adding the SH pulse. We attribute this to the HHG cutoff being significantly more broadband in energy than the mirror bandwidth, meaning that the mirror bandwidth is what limits the pulse duration. For a high harmonic field, $\tilde{E}(\omega)$, and a MoSi reflectivity function, $\tilde{H}(\omega)$, the field of the attosecond pulse, $E(t)$, is

$$E(t) = \int_{-\infty}^{\infty} \tilde{H}(\omega)\tilde{E}(\omega)\exp(i\omega t)d\omega. \tag{1}$$

For a broad enough cutoff, such that the phase and amplitude of $\tilde{E}(\omega)$ are approximately constant across $\tilde{H}(\omega)$, $E(t)$ becomes

$$E(t) = \tilde{E}_0 \int_{-\infty}^{\infty} \tilde{H}(\omega)\exp(i\omega t)d\omega, \tag{2}$$

where $\tilde{E_0}$ is directly proportional to the XUV flux in the cutoff region. The pulse duration is therefore determined by the mirror reflectivity. This is valid as long as the modulations in the cutoff are not large enough that the high energy tail of the cutoff shifts down enough to overlap with $\tilde{H}(\omega)$ or shifts up enough that the modulated plateau region overlaps with $\tilde{H}(\omega)$. These conditions were satisfied in the experiment. Hence, the result of adding the SH field is an increased XUV flux with no significant change to the XUV waveform. This is advantageous for applications where a flux increase is desirable without any shift in the pulse central photon energy or pulse duration. For example, this flux increase at 90 eV will enhance the utility of our attosecond beamline for the study of surface electron dynamics using pump-probe techniques [39].

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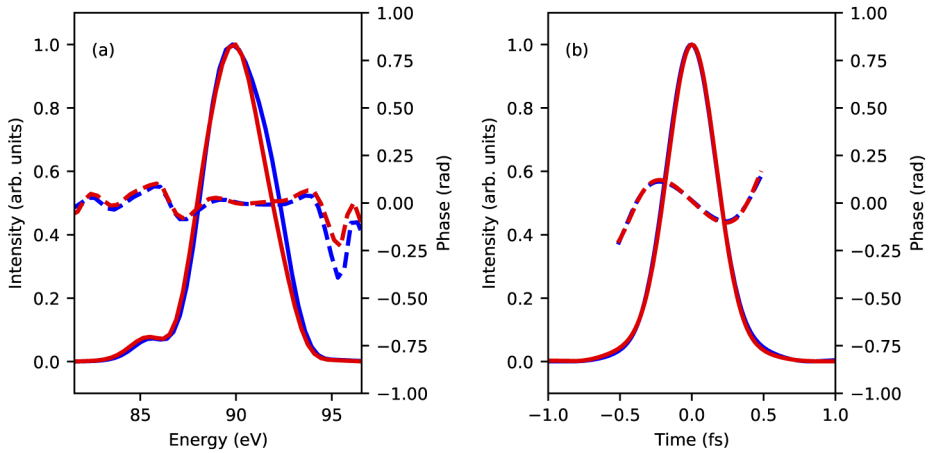

Fig. 5. Spectral profile (a) and temporal profile (b) of the XUV pulse with second harmonic field present (blue) and absent (red).

The retrieved streaking fields for FC+SH and FC alone are shown in Fig. 6. The retrieved pulse durations of 4.9 ± 0.5 fs for FC+SH and 5.1 ± 0.5 fs for FC alone are consistent with the pulse broadening expected of a ~4 fs pulse propagating through the 127 μm fused silica filter. From the spectrum of the retrieved streaking field shown in Fig. 6(a), a small 400 nm spectral component can be seen which is not present in Fig. 6(b). Both retrievals were seeded by a 5 fs Gaussian FC pulse and a 100 as Gaussian XUV pulse. No assumption about the presence of the SH pulse was made in either case. The low intensity wings in Fig. 6(c) and (d) are an artefact due to FC pulse intensity fluctuations. The SH pulse is too weak to be seen by eye in the time domain, but can be clearly observed in the frequency domain due to its spectral separation from the FC pulse.

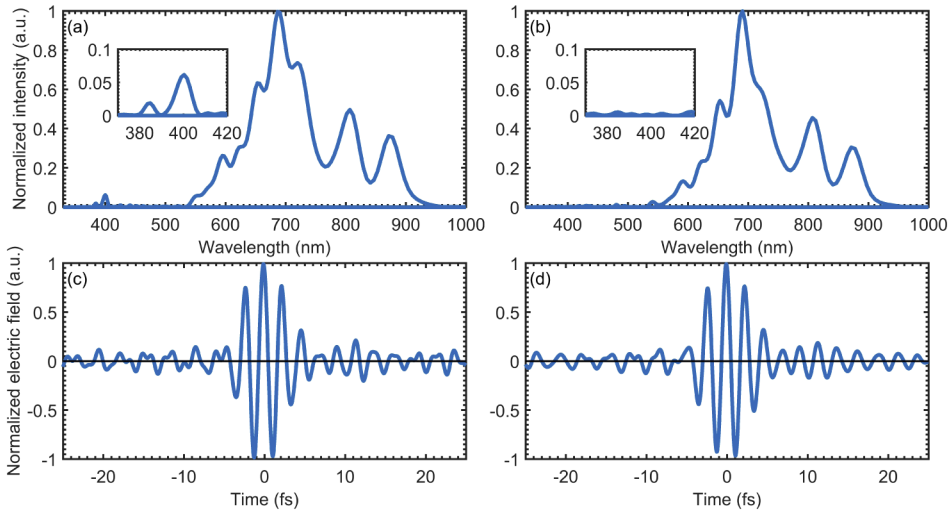

Fig. 6. Retrieved gate pulse spectrum and field with second harmonic field present (a) and (c), and absent (b) and (d).

From the peak streaking amplitude, the intensity of the combined fields is calculated to be 9×10^{10} W cm^{-2}, and from the ratio of peak field strengths between the two retrieved pulses, a

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peak SH intensity of 3×10^8 W cm^{-2} is calculated. The SH field strength is around 2 orders of magnitude lower than typically used for streaking, and on its own is too weak to be retrieved. However, with the FC field present the two fields form a coherent superposition which modulates the photoelectron momentum [17,30]. The effect of the SH pulse is therefore to cause shifts in momentum around the centre of momentum caused by the FC field. These modulations are occurring well above the noise floor of the streaking trace, allowing the modulations to be extracted by the FROG-CRAB retrieval. The FC:SH intensity ratio is ~300:1, corresponding to a field strength ratio of ~17:1.

It is important to note that that the phase of the SH field relative to the few-cycle field retrieved from the streaking measurement also includes the optical path difference of the annular fused silica filter that is only known to within an error of ±0.34 μm due to the thickness tolerance of this optic. Hence, the relative phase retrieved from the streaking is not indicative of the relative phase in the HHG gas jet. However we do not see this as a significant limitation, as the relative phase can be directly obtained from the XUV cutoff shift in Fig. 3 using the method described in [30].

4. Conclusion

We generated and measured isolated attosecond pulses using a synthesized two-colour field consisting a few-cycle near-infrared pulse and a weaker multi-cycle second harmonic pulse. We showed that a two-fold flux enhancement can be achieved with the correct choice of phase difference between the few-cycle and second harmonic pulses, consistent with previous measurement with plateau harmonics [24]. We found that the addition of the second harmonic field did not change the temporal profile of the attosecond XUV pulse, which we attribute to the spectral filtering of the cutoff by the MoSi mirror in the streaking setup. We retrieve the spectrum of the second harmonic pulse using a heterodyning arrangement with the few-cycle pulse, despite the second harmonic intensity being too weak for a streaking measurement to be performed on the pulse alone. This work not only affirms the sensitivity of the attosecond streaking technique and its utility for full characterisation of synthesised fields, but is also of importance to nonlinear XUV science where flux enhancement whilst maintaining the attosecond pulse duration is a critical enabling step toward attosecond XUV pump-probe spectroscopy [40].

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Disclosures

The authors declare no conflicts of interest.

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