Space-time control of free induction decay in the extreme ultraviolet

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Ultrafast extreme-ultraviolet (XUV) and X-ray sources are revolutionizing our ability to follow femtosecond processes with ångström-scale resolution. The next frontier is to simultaneously control the direction, duration and timing of such radiation. Here, we demonstrate a fully functional opto-optical modulator for XUV light, similar to modulators available at infrared (IR) and visible wavelengths. It works by using an IR pulse to control the spatial and spectral phase of the free induction decay that results from using attosecond pulses to excite a gas. The modulator allows us to send the XUV light in a direction of our choosing at a time of our choosing. The inherent synchronization of the XUV emission to the control pulse will allow laser-pump/X-ray probe experiments with sub-femtosecond time resolution.

In this Article we report an extension of the temporal control of the XFID to the spatial domain thus demonstrating an opto-optical modulator for XUV light. We used an IR control pulse to manipulate both the spatial and spectral properties of resonant emission in the XUV, sending the emission in a direction of our choosing at a time of our choosing. By adding control over the spatial phase of the XFID emission we can both create narrow-band sources of XUV radiation tuned to specific frequencies and we can study resonant emission in a background-free measurement scheme. Our opto-optical modulator also opens a way towards synchronizing X-rays from XFELs and IR pulses at the sub-femtosecond level—an order of magnitude better than is currently possible. Our work also demonstrates the benefits of considering absorption and emission as field-driven processes in the time domain, which complements the more common picture of photon-driven processes in the frequency domain.

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**Experimental scheme**

To induce XFID we used a tunable XUV pulse produced by HHG7–11 to create a coherent superposition of the ground and excited states in gas-phase argon. After being coherently excited, the different atoms along the propagation direction emit light with a fixed phase relation. This leads to phase-matching of the dipole emission in the forward direction, resulting in an XFID signal (Fig. 1a–c). Although the XFID is emitted in the same direction as the excitation pulse, the temporal properties of the emission depend on the coherence time of the excited state. This results in an exponentially decaying XFID emission following the excitation pulse, which allows interaction between the excited atoms with an additional light pulse (Fig. 1d–g), enabling phase control without perturbing the excitation processes. When a moderately strong IR control pulse interacts with the excited atoms in the ensemble the energy levels will be a.c.-Stark shifted. If the laser pulse is not too strong (that is, not ionizing the medium), the energy levels will follow the field adiabatically and return to their initial energy after the pulse; the only difference being that the superposition has accumulated an additional phase shift $\Delta \varphi$. This phase shift depends on the intensity of the IR pulse:

$$\Delta \varphi(x, y, z) = \int_{t_m} \Delta E(x, y, z, t) \, dt$$  \hspace{1cm} (1)
Figure 1 | Schematic illustration of XFID radiation control. a, An ensemble of atoms excited by an ultrafast XUV pulse emit XFID radiation (purple) after the XUV pulse (blue) has passed. The atoms oscillate in phase orthogonal to the propagation direction of the excitation pulse (illustrated by the vertical purple lines). Phase matching creates a well-defined XFID beam. b, The temporal structure of the XFID emission decays over a long time. c, The phase relation between the excitation pulse and the XFID leads to destructive interference at the detector in the far-field, normally called absorption. d, An IR pulse (red) that co-propagates with the XUV pulse through the medium creates a spatially dependent phase shift of the dipoles via the a.c.-Stark effect. This phase shift depends on the integrated IR intensity for each atom and results in a rotation of the wavefronts after the IR pulse. e, This rotation redirects the XFID emission after the IR pulse. f, g, In the far-field this yields an off-axis emission component as well as an altered on-axis absorption.

where \( \Delta E \) is the intensity-dependent energy shift of the levels and \( T_{IR} \) is the duration of the IR pulse. If all the atoms are exposed to the same time-dependent IR intensity, the induced phase shift will lead to interference between the part of the XFID that comes before the IR pulse and the part that comes after, thereby modifying the spectral and temporal structure of the emission in the forward direction. By parallel shifting the IR beam, different atoms are exposed to different IR intensities, imposing a spatial phase variation across the gas. This allowed us to tailor the wavefront of the XFID, redirecting all the emission that comes after the control pulse (see Fig. 1d–g for a schematic illustration and Fig. 2 for an experimental demonstration).

Spatial control of XFID emission

Our experimental technique (see Methods and Supplementary information) utilizes an attosecond transient-absorption scheme in a non-concentric geometry together with a high-resolution, flat-field imaging spectrometer. In the first set of measurements that we will discuss, the laser wavelength is 780 nm so that the 9th harmonic is centred at 14 eV and the broadband XUV light spectrally covers both the \( 3s^23p^6 \rightarrow 3s^23p^5[2^3P_{3/2}]3s(J = 1) \) (14.09 eV) and \( 3s^23p^6 \rightarrow 3s^23p^5[2^3P_{3/2}]3d(J = 1) \) (14.15 eV) transitions. The lifetimes of these states are in the nanosecond range, 10.1 and 3.48 ns, respectively. Although the coherence times are shorter than the lifetimes, they are still orders of magnitude longer than the femtosecond pulses used. Figure 2 shows experimental results for different IR intensities. When no IR control pulse is used (Fig. 2a) the XFID signal consists of narrow spectral lines that are slightly more divergent than the original pulse. This increased divergence depends on the coherence time of the states. For states with shorter coherence times than the ones presented here, such as autoionizing states presented later in this Article, the divergence of the XFID emission is the same as that of the excitation pulse. Figure 2b–d show the results when the IR control pulse is 200 fs after the excitation pulse. As the control pulse interacts with the medium it induces a spatial phase variation across the ensemble of excited atoms. This terminates the on-axis component of the XFID field, and the subsequent XFID signal is redirected, creating the observed off-axis signal. With increasing intensity, the spatial phase variation increases, resulting in an increase in the redirection angle.

The efficiency of the process can largely be divided into the sum of three effects. When the XUV pulse passes through the ensemble of atoms the amount of absorption, and therefore also emitted FID signal, is set by the gas pressure, medium length and oscillator strength. The second effect depends on how much the control pulse is delayed. For a long delay the polarization of the ensemble will have decayed resulting in less energy in the redirected XFID. For short delays most of the XFID signal may, in principle, be redirected. The third effect comes from the control pulse interacting with the ensemble, for example, how well the phase of the ensemble is shaped to emit light in one direction, or how much the energy is reduced through coupling to other states or ionizing. In the experimental conditions leading to Fig. 2d, approximately 50% of the incident light (Fig. 2a) is redirected.

The on- and off-axis components of the XFID also display different spectral characteristics. The total on-axis signal results from coherent interference of the incoming field and the generated XFID field in the forward direction. Since the XFID emission is \( \pi \) phase-shifted with respect to the excitation pulse, the two fields interfere destructively. This results in a spectral hole at the transition energies, normally called absorption. The width of the spectral hole
relates to the lifetime of the emission and by terminating the on-axis component of XFID with a control pulse the effective lifetime is shortened and the spectral hole is widened (compare Fig. 1c and g). Changing the delay between the pulses thus affects the spectral width of the absorption profile. In contrast to this, the spectral width of the redirected XFID signal is determined by the decay time of the emission, which is independent of the delay. The width of the redirected signal does, however, depend on the gas pressure due to propagation effects\(^\text{36}\). Propagation effects may also induce a spatial chirp on the redirected XFID emission.

Figure 3 shows the on-axis versus off-axis spectral features as a function of the relative delay ($\tau$) between the pump and control pulses.
pulses. The intensity of the control pulse and the gas pressure are reduced compared with that in Fig. 2 to minimize the perturbation of the excitation and propagation processes, in particular when the two pulses overlap temporally. In this experiment we directed the XFID emission upwards. Figure 3 compares the spectral width of the on-axis absorption and the XFID off-axis emission, and shows that the spectral width of the off-axis emission is not affected by the delay, while the total signal is reduced. In contrast, the spectral width of the on-axis absorption shows an inverse relation with the time delay, in agreement with our description of the process. In the off-axis emission the two states are clearly resolved as emission peaks for all delays, whereas they can only be resolved as two absorption dips in the on-axis emission for delays longer than 500 fs.

Theoretical model

To model these experimental results we used the theoretical framework outlined in ref. 43. We numerically solved the coupled time-dependent Schrödinger equation (TDSE), in the single active electron approximation (SAE), and the Maxwell wave equation (MWE). While the experiment considers a non-coaxial geometry for the two pulses, to simplify the computations we assumed a coaxial geometry in our simulations. The primary difference between these two geometries is that the non-coaxial geometry redirects the resonant XUV emission in a single direction, whereas the coaxial geometry directs the resonant XUV emission into a halo around the shared axis. For the SAE-TDSE calculations we used a pseudopotential that accurately reproduces the singly excited energy levels of helium. The IR pulse had a central wavelength of 770 nm, a full-width at half-maximum (FWHM) duration of 30.8 fs and a peak intensity of 0.5 TW cm$^{-2}$, whereas the XUV pulse had a central energy of 21.1 eV, a FWHM duration of 20.5 fs and a peak intensity of 10$^{10}$ W cm$^{-2}$. The central energy of the XUV pulse was chosen to match the $1s^2 - 1s2p$ excitation energy in our helium potential. We imposed a 360 fs dephasing time on the dipole, which, although it is much shorter than the true dephasing
time, is much longer than the timescale set by the IR pulse and the relative XUV–IR delays used. The confocal parameters of the two beams were chosen such that they had similar beam waists, with a $b_{\text{IR}} = 1 \text{ cm}$ and $b_{\text{XUV}} = 25 \text{ cm}$. This allowed the atoms that interact with the XUV field at the centre of the IR beam to experience a Stark shift considerably different from the atoms that interact with the XUV field at the edge. To further reduce the required computational time, we restricted the fields to interact with only a single plane of atoms, with a width of $0.001 \text{ cm}$ and a high atomic density of $5 \times 10^{18} \text{ atoms cm}^{-3}$. The calculated near-field macroscopic electric field was transformed to the far field, and the azimuthally integrated spectral intensity was plotted.

The results are shown in Fig. 4a for the case of an IR pulse arriving 13 fs after the XUV pulse. Here, we see that close to the common propagation axis, for example, at $0.02 \text{ cm}$, the XUV spectrum shows absorption at the resonant frequency, whereas off-axis, for example, at $0.08 \text{ cm}$, there is an emission feature at the resonant frequency. In our calculations, this off-axis resonant emission only occurs when the IR pulse overlaps with the XUV pulse in time or arrives after the XUV pulse, which agrees well with the experimental observations. We also find (not shown) that we can reduce the radial extent of this off-axis emission by reducing the peak intensity of the IR pulse, in agreement with the experimental observations illustrated in Fig. 2b–d.

To compare our theoretical calculations to these experimental results, we plotted lineouts of the azimuthally integrated spectral intensity at different delays for the off-axis and on-axis cases in Fig. 4b and c, respectively. These off-axis and on-axis results agree well with the observed experimental results in Fig. 3b and c. For the off-axis case, we find that increasing the IR–XUV delay reduces the intensity of the resonant emission feature, approaching the results for the case of no IR pulse at delays close to the dephasing time of the dipole. The off-axis emission feature maintains a Lorentzian lineshape with an almost constant spectral width for all delays. In contrast, the on-axis case shows much less intensity modulation with increasing delay, however, the dispersive lineshape

Figure 5 | Delay dependence of XFID emission. a–c, Spatial-spectral profile of XUV light transmitted through the argon gas for three different delays between the IR and XUV pulse. Negative delays correspond to the IR pulse preceding the XUV pulse. d,e, Lineouts showing how the emission direction changes as a function of delay from the different excited states (7p and 8p). The signal is normalized to the case when the IR pulse precedes the XUV pulse. The off-axis signal is integrated around $\pm 2 \text{ mrad}$ and shown as a white line, normalized to the final value.
evolves significantly as we vary the IR–XUV delay. The on-axis absorption linshape becomes narrower with increasing delay, approaching the on-axis results for the case of no IR pulse at long delays.

Temporal dynamics of XFID emission
Next we experimentally investigated the delay dependence of the XFID emission while also showing that our technique can be extended to shorter XUV wavelengths. To this end, we excited several autoionizing states in argon. The $3d^33p^6np$ series of autoionizing states in argon have much higher excitation energies and shorter lifetimes than the states previously described. We focused on the control of states with principal quantum numbers $n \geq 6$ since these states can be expected to have lifetimes longer than the duration of our control pulse\textsuperscript{33,44}. The laser system was tuned to a carrier wavelength of 820 nm, resulting in harmonic 19 spectrally overlapping the $n = \{6,7,8\}$ states (the vertical lines in Fig. 5a).

Figure 5a–c presents the experimentally measured spatial–spectral profile for three different delays between the XUV and IR pulses. When the IR pulse arrives 50 fs after the XUV pulse (Fig. 5c), we observe that the on-axis light is almost completely redirected.

The temporal dynamics of the waveform rotation induced by the IR-imposed phase shift is explored further in Fig. 5d,e, where the temporal structure of the redirected XFID emission at the energies corresponding to the $n = \{7,8\}$ states is plotted as a function of delay. When the IR pulse precedes the XUV pulse the emission has the same direction and divergence as the pump pulse. In this case the coherence times of the states are short with no time for dephasing, and therefore no increase in the divergence of the XFID emission. When the two pulses start to overlap, the XFID emission begins to change direction. With the intensity gradient used in this experiment, the maximum waveform rotation speed is about 0.06 mrad fs$^{-1}$. For longer delays the direction no longer changes, and the decay can be followed by integrating the off-axis signal. The decay times for the XFID signals from the $7p$ and $8p$ states were found to be 264 ± 2 fs and 405 ± 9 fs, respectively. These numbers can be compared with the expected lifetimes for even more precise manipulation of the temporal structure of the XFID emission along a particular direction and thus precise temporal control over narrow-band XUV sources. The ability to control the XUV emission in both time and space will also make these light sources attractive to a wider variety of users, and there are many exciting opportunities ranging from ultrafast quantum information in the XUV to nanophotonics studies and femtochemistry. It is also possible to envision the seeding of free-electron lasers with controllable narrow-band XUV light to minimize temporal jitter effects\textsuperscript{45}. These same techniques may also help improve the synchronization between light from FEls and lasers by exciting the ensemble of atoms using the XUV and then producing an XUV pulse using opto-optical modulation. The resulting XUV pulse will then be synchronized with the laser pulses used to redirect the emission, at the femtosecond precision level.

Methods
Methods and any associated references are available in the online version of the paper.

Received 25 November 2016; accepted 7 February 2017; published online 20 March 2017

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Methods
The experimental set-up relies on an amplified titanium:sapphire laser system, which delivers 5 mJ pulses, with a carrier wavelength around 800 nm, at a repetition rate of 1 kHz. The laser system contains two acousto-optical programmable dispersive filters (Dazzler and Mazzler, Fastlite), capable of shaping the spectral phase and amplitude of the amplified pulses. The Mazzler is placed inside the cavity of a regenerative amplifier, where it is used to counteract the effects of gain narrowing by selectively diffracting the spectral components with the highest gain out of the cavity. The use of the Mazzler allows for a top-hat-shaped spectral gain profile of the laser system with a bandwidth of around 100 nm. The Dazzler is placed in the stretched oscillator beam and is used to pre-compensate any nonlinear phase accumulated by the pulse in the amplifier system to generate pulses with near transform-limited duration ($T_{\text{IR}}$ $\approx$ 20 fs) after a grating compressor. In addition to optimizing the compression of the pulses, the Dazzler is used to restrict the bandwidth of the seed pulses for the laser chain. The combination of the Dazzler and Mazzler allows for a tunability of the carrier wavelength of the laser system to roughly 50 nm (775–825 nm), while maintaining a bandwidth of 50 nm. The duration of the bandwidth-reduced pulses corresponds to $T_{\text{IR}}$ $\approx$ 30 fs. The pulses were directed into a balanced Mach–Zehnder interferometer actively stabilized by a co-propagating frequency-stable helium–neon laser. A detailed description of the experimental set-up can be found in the Supplementary Information.

The decay times for the XFID signals in Fig. 5d,e were calculated in the following way. Assuming a Gaussian temporal structure of the infrared laser pulse allowed us to derive a simple analytical expression for the time dependence of the redirected signal by utilizing equation (1) in the following way

$$S(\tau) = \frac{a}{2} \left[ 1 + \text{erf} \left( \frac{b - \tau}{\sqrt{4 \ln(2) T_{\text{IR}}}} \right) \right] \exp \left( -\frac{\tau - b}{T_{\text{FID}}} \right)$$

(2)

where $a$ denotes the amplitude, erf denotes the Gaussian error function, $b$ the position of temporal overlap and $T_{\text{FID}}$ the characteristic decay time for the FID signal. The uncertainty above denotes the 95% confidence interval of the fit.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.