Coherent control with a short-wavelength free-electron laser

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Extreme ultraviolet and X-ray free-electron lasers (FELs) produce short-wavelength pulses with high intensity, ultrashort duration, well-defined polarization and transverse coherence, and have been utilized for many experiments previously possible only at long wavelengths: multiphoton ionization¹, pumping an atomic laser² and four-wave mixing spectroscopy³. However, one important optical technique, coherent control, has not yet been demonstrated, because self-amplified spontaneous emission FELs have limited longitudinal coherence⁴–⁷. Single-colour pulses from the FERMI seeded FEL are longitudinally coherent⁸,⁹, and two-colour emission is predicted to be coherent. Here, we demonstrate the phase correlation of two colours, and manipulate it to control an experiment. Light of wavelengths 63.0 and 31.5 nm ionized neon, and we controlled the asymmetry of the photoelectron angular distribution¹⁰ by adjusting the phase, with a temporal resolution of 3 as. This opens the door to new short-wavelength coherent control experiments with ultrahigh time resolution and chemical sensitivity.

Coherent control with lasers involves steering a quantum system along two or more pathways to the same final state, and manipulating the phase and wavelength of light to favour this state. This technique represents a major achievement in the quest to understand and control the quantum world. In some cases it is a simple interference effect between transition matrix elements, say \(M_1e^{i\theta_1}\) and \(M_2e^{i\theta_2}\), and can be written as

\[
I \sim |M_1e^{i\theta_1} + M_2e^{i\theta_2}|^2 = |M_1|^2 + |M_2|^2 + 2|M_1||M_2|\cos(\theta_1 - \theta_2)
\]

(1)

where \(I\) denotes intensity and \(\theta_1 - \theta_2\) the relative phase. The amplitudes of the matrix elements must have similar absolute values to produce significant interference: otherwise the greater of the terms \(|M_1|^2\) or \(|M_2|^2\) dominates.

To achieve coherent control, longitudinal (that is, temporal) phase correlation must first be demonstrated and manipulated. There are many ways to do this with optical lasers¹²,¹³, for example using bichromatic light (two overlapping commensurate wavelengths¹⁴,¹⁵). In one implementation, ionization by two first-harmonic photons and one second-harmonic photon is measured, and the anisotropy of the photoelectron angular distribution is observed as a function of the phase difference between the two temporally overlapping wavelengths¹⁴–¹⁷. At optical wavelengths, harmonics are easily generated, for example, by nonlinear birefringent crystals or third-harmonic generation in gases. In the extreme ultraviolet (EUV) region, frequency doubling or tripling is impractical due to the lack of efficient media. The phase difference is easily tuned in the optical region by gas cells or mechanical delay lines. In the EUV region, such methods become difficult or impossible, as all gases absorb too strongly to function efficiently, whereas mechanical delay lines require extreme precision in path length differences. Soft X-ray delay lines are usually constructed with grazing incidence optics, and the required resolution and stability is beyond present technology. In particular, it is very difficult to maintain nanometre and microradian precision in an instrument several metres long. A recent state-of-the-art EUV delay line has a time resolution of 210 as (ref. 18), insufficient for coherent control at short wavelengths, with a much shorter period. Higher performance (40 as) is possible using normal incidence, split-mirrors¹⁹. This geometry functions at long wavelengths or over narrow ranges at short wavelengths with special coatings such as multilayers and filters working in restricted ranges, and so transmission is limited.

High-harmonic generation (HHG) sources produce ultrafast pulses of soft X-ray light as a comb of harmonics of the fundamental radiation²⁰. Although the coherence of the spectral components of the comb has been verified in several experiments, and is the basis of the attosecond temporal structure²¹, a straightforward and widely applicable method to control the relative phase of two harmonics has not been demonstrated. Also, harmonics generated by HHG do not have the high pulse energy and continuous tunability.

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of FELs. Coherent control using trains of attosecond pulses and synchronized infrared (IR) fields has been demonstrated, where the control parameter is the relative timing between the attosecond bursts and the phase of the IR field, rather than the relative phase of the EUV harmonics. Bichromatic multi-photon ionization has also been reported with phase control, but again the phase of optical photons was controlled, not that of EUV light.

Here we demonstrate and exploit the longitudinal coherence of two-colour EUV light from FERMI by adopting a radically different approach to tuning the phase: instead of generating the light and manipulating the phase subsequently, two colours are generated by the FEL with a variable phase difference. An electron delay line controls the phase of the light, which is adjusted by varying the phase of the electron bunch relative to that of the first colour. The delayed electrons then generate the second colour with a delayed phase. The carrier wave phase and pulse envelope are shifted, but for long pulses (~100 fs), the envelope shift is unimportant.

FERMI has been described elsewhere, and here we summarize the salient points of the machine (Fig. 1). Six APPLE-type undulators can be set independently to produce polarized light at harmonics of the seed wavelength: we used horizontal linear polarization. The 2s to 2p resonance of Ne at 62.97 nm (hereafter 4s resonance) was selected and the first five undulators were set to it (see Methods). The sixth undulator was set to radiate at the second harmonic, 31.5 nm, and the electron delay line between the fifth and sixth undulators controlled the relative phase. We checked for spurious effects (see Methods and Supplementary Fig. 2). The overlapping beams were then transported to the experimental chamber via the PADReS system and focused to a measured spot size of 7–10 µm (see Methods and Supplementary Fig. 3).

The scheme of the experiment is shown in Fig. 3a. 2p electrons from neon can be emitted by two quantum paths: by a single photon (frequency ω) as an s- or d-wave; or by two photons (frequency 2ω) as a p- or f-wave. The weak second-harmonic field ionizes by a first-order process, whereas the intense first-harmonic field ionizes by a second-order process, the ionization rates were adjusted to similar values by varying the intensities of the two wavelengths. Choosing the 4s resonance enhances the cross-section for the two-photon process and selects an outgoing p-wave, without a significant f-wave contribution. Owing to the non-linear nature of the process and different parity of the outgoing electronic wave packets generated by the two wavelengths, symmetry breaking occurs in the photoelectron angular distribution with respect to the plane perpendicular to the electric vector of the light, see Fig. 1 of ref. 16. The asymmetry depends strongly on the relative phase of the two fields, and gives rise to an oscillatory term similar to that in equation (1). If the temporal lag between the two harmonics is Δt, the relevant parameter is the delay-induced phase difference Δφ = 2ωΔt.

The photoelectron angular distributions were measured using the VMI (velocity map imaging) spectrometer of the Low Density Matter (LDM) end-station, see Methods. The ‘left–right’ asymmetry
was quantified by the parameter $A_{LR}$

$$A_{LR} = \frac{I_L - I_R}{I_L + I_R}$$

(2)

where $I_L$ and $I_R$ are the integrated intensities on the left and right of the image.

Figure 3b shows the asymmetry parameter $A_{LR}$ as a function of $\Delta \phi$. Clear oscillations are present, with a period $2\pi$ rad or 105 as, the second harmonic period. The measurement steps were approximately 10 as, but a subsequent scan over a limited range, with steps of 900 zs, indicated a resolution of 3.1 as (Fig. 3c).

To understand the asymmetry in detail, the angular distribution was fitted with Legendre polynomials$^{29}$, each characterized by a $\beta$ parameter. The even ($\beta_2$, $\beta_4$ and so on) and odd ($\beta_1$, $\beta_3$ and so on) numbered parameters describe the symmetric and antisymmetric parts of the distribution respectively. The fits for $\beta_2$ and $\beta_4$ in Fig. 3b are consistent with the calculated errors, whereas $\beta_1$ shows a larger deviation, possibly indicating systematic errors. Comparing the data qualitatively with the calculated spectra of a simpler system, atomic hydrogen$^{30}$, we find the key characteristics are reproduced: $\beta_2$ is constant whereas $\beta_1$ and $\beta_3$ oscillate with a phase lag, in this case 1.06 rad. The non-zero lag already follows approximately 10 as, but a subsequent scan over a limited range, with steps of 900 zs, indicated a resolution of 3.1 as (Fig. 3c).

The present result demonstrates phase control at the attosecond level with FERMI, and opens the way for unique experiments in the gas phase, but it may be adapted to condensed matter to manipulate electron wave-packet motion in processes important in catalysis, photosynthesis and solar energy production. In addition, the generation of attosecond pulses and pulse trains is based on coherent control of harmonics, thus the way is now open to developing such pulse sculpting techniques with FELs, as more than two harmonics can be generated at FERMI by appropriate settings of the undulators. Furthermore, this method may be applicable at SASE FELs if operated in single spike or self-seeding modes, greatly extending the wavelength range beyond that presently available.

Methods

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

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Author contributions
The experiment was conceived by K.C.P., G.S., A.N.G.G. and K.U., and the method of operating FERMI to carry it out was devised by E.A. and L.G. The experiment was prepared and carried out by K.C.P. E.A., C.C., R.C., G.D.N., S.D.M., B.D., E.F., P.F., D.G., L.G., N.M., G.P., O.P., L.R., P.R., E.R., C.S., M.T., M.Z., G.S., P.C., D.I., Y.K., T.T., K.U., A.F., F.S., E.O., T.M., M.N., M.C. and M.M. Theoretical calculations (of machine properties or neon spectra) were performed by E.A., L.G., A.N.G.G., E.V.G., S.I.S., K.B., N.D. and J.V. Detailed data analysis was performed by M.R., P.C. and D.I. The manuscript was drafted by K.C.P. and completed in consultation with all authors.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to K.C.P., G.S. and K.U.

Competing financial interests
The authors declare no competing financial interests.
Methods
The measurements were carried out at the LDM beamline, FERMI$^{31,32}$. The relative phase of the two wavelengths was adjusted by means of the electron delay line or phase shifter$^{29}$, which is a magnetic structure that deflects the electrons so that they follow a roughly sinusoidal path over two damped periods. The length of the electron trajectory can be increased in small steps from zero to one wavelength of the light (31.49 nm in the present case) or more, with a calibration depending on the electron beam energy and wavelength of the light. In principle the path length difference can be adjusted at the picometre scale, but before the present experiment there was no method available for precise determination of the resolution, which we have now measured, see below.

Supplementary Fig. 1 shows the calculated FEL output intensities for the first (63.0 nm, red) and second harmonics (31.5 nm, blue) as a function of time, and their phase difference (black curve), with a quadratic fit (green curve). The simulation was carried out with the FEL time-dependent code Ginger$^{29}$, using the undulator and seeding configuration of the experiment shown in Fig. 1. The two pulses have good temporal overlap, with calculated pulse durations of 118 and 102 fs. The phase difference has a mild longitudinal dependence on the intensity of the harmonic light, and that the second harmonic produced in the first undulator was set to a pulse energy of approximately 50 µJ per pulse, and the two-photon signal from the second undulator was present. Different behaviour was observed when they were tuned to the third harmonic, thereby demonstrating that this diagnostic is effective. The use of a helical undulator configuration has been shown to remove this contamination, allowing the flexibility of phase control of odd and even harmonics.

The optical focusing conditions were simulated for optimal curvature of the Kirkpatrick–Baez active optics$^{29}$, and verified experimentally by using a Hartmann wavefront sensor. Supplementary Fig. 3 shows the calculated focal spot shape and line profiles of the first and second harmonics, both calculated and measured from the reconstructed image measured by the wavefront sensor. The measured spot size was 7–10 µm (FWHM). The average pulse energy of the first harmonic was 50 µJ, and after correcting for the transmission efficiency of the beam transport optics, we estimate that the average intensity in the focal spot was $1 \times 10^{17}$ W cm$^{-2}$, for a pulse duration of 100 fs$^{36}$.

The atomic beam was produced by a supersonic expansion and defined by a skimmer and vertical slits. The length of the beam along the light propagation direction was approximately 1 mm. The data from the wavefront sensor was used to reconstruct the spot size at the centre and extremes of this excitation volume and compared with spot profiles calculated using the WISE code$^{32}$ and other software. The diameter at the extremes is approximately 2% larger than at the centre.

The data was analysed by firstly correcting the VM1 images for spatial variations of sensitivity of the detector. The spectrometer works by projecting the expanding sphere of photoelectrons of a given kinetic energy onto a plane surface, and to recover the original angular distribution, the projected images must be inverted. Cylindrical symmetry is assumed and we used the pBASEX software$^{29}$. The inverted (or reconstructed) angular distributions were integrated over the left and right sides of the image to give a value of the asymmetry parameter. An example of an image and a line profile is shown in Fig. 2b.

The error bars of $A_{2\beta}$ in Fig. 3c of the main text were estimated from the standard deviation of the fitting parameters for the photolines separately for the left and right sides of the VM1 images. The error bars for the $\beta_2$ parameters were determined by dividing the data into five subsets, and analysing them separately. Then the standard deviation was calculated for the whole data set. This procedure estimates statistical fluctuations, but does not take account of systematic errors. The $\beta_2$ curve was fitted by a constant straight line and $\beta_2$ and $\beta_3$ by sinusoidal functions, with the frequency of both curves constrained to be equal. A constant for $\beta_2$ and a sine function for odd $\beta$ values are the forms expected for the case of hydrogen$^{29}$.

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