

Coherent control with a short-wavelength Free Electron Laser

K. C. Prince^{1,2,3,*}, E. Allaria¹, C. Callegari¹, R. Cucini¹, G. De Ninno¹, S. Di Mitri¹, B. Diviacco¹, E. Ferrari¹, P. Finetti¹, D. Gauthier¹, L. Giannessi^{1,4}, N. Mahne¹, G. Penco¹, O. Plekan¹, L. Raimondi¹, P. Rebernik¹, E. Roussel¹, C. Svetina^{1,5}, M. Trovò¹, M. Zangrando^{1,3}, M. Negro⁶, P. Carpeggiani⁶, M. Reduzzi⁶, G. Sansone⁶, A. N. Grum-Grzhimailo⁷, E.V. Gryzlova⁷, S.I. Strakhova⁷, K. Bartschat⁸, N. Douguet⁸, J. Venzke⁸, D. Iablonskyi⁹, Y. Kumagai⁹, T. Takanashi⁹, K. Ueda⁹, A. Fischer¹⁰, M. Coreno¹¹, F. Stienkemeier¹², E. Ovcharenko¹³, T. Mazza¹⁴, M. Meyer¹⁴,

¹ Elettra-Sincrotrone Trieste, 34149 Basovizza, Trieste, Italy, ² Molecular Model Discovery Laboratory, Department of Chemistry and Biotechnology, Swinburne University of Technology, Melbourne, 3122, Australia, ³ Istituto Officina dei Materiali, Consiglio Nazionale delle Ricerche, 34149 Basovizza, Italy, ⁴ ENEA C.R. Frascati, 00044 Frascati, Rome, Italy, ⁵ University of Trieste, Graduate School of Nanotechnology, 34127 Trieste, Italy, ⁶ Dipartimento di Fisica, CNR-IFN, Politecnico di Milano, Piazza Leonardo da Vinci, 32, 20133 Milan, Italy, ⁷ Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, Moscow, 119991 Russia, ⁸ Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA, ⁹ Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan, ¹⁰ Max Planck Institute for Nuclear Physics, Heidelberg, 69117 Germany, ¹¹ ISM, Consiglio Nazionale delle Ricerche, 34149 Basovizza, Italy, ¹² Physikalisches Institut, Universität Freiburg, 79106 Freiburg, Germany, ¹³ Institut für Optik und Atomare Physik, TU Berlin, Berlin, Germany, ¹⁴ European XFEL, Albert-Einstein-Ring 19, 22761 Hamburg, Germany.

XUV 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 utilised for many experiments previously possible at long wavelengths only: 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 coherent^{8,9}, 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 the asymmetry of the photoelectron angular distribution^{10,11} was controlled by adjusting the phase, with temporal resolution 3 attoseconds. 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 $M_1 e^{i\theta_1}$ and $M_2 e^{i\theta_2}$, and can be written as

$$I \sim |M_1 e^{i\theta_1} + M_2 e^{i\theta_2}|^2 = |M_1|^2 + |M_2|^2 + 2|M_1||M_2| \cos(\theta_1 - \theta_2) \quad (1)$$

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 (i.e. temporal) phase correlation must first be demonstrated and manipulated. There are many ways to do this with optical lasers^{12,13}, e.g. using bichromatic light, i.e. two overlapping commensurate wavelengths^{11,13}. 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, e.g. by nonlinear birefringent crystals or third-harmonic generation in gases. In the XUV 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 XUV region, such methods become difficult or impossible, as all gases absorb too strongly to function efficiently, while 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 nanometer and microradian precision in an instrument several meters long. A recent state-of-the-art XUV delay line has a time resolution of 210 attoseconds¹⁸, insufficient for coherent control at short wavelengths, with a much shorter period. Higher performance (40 attoseconds) 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 at 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 of FELs. Coherent control using trains of attosecond pulses and synchronized infrared (IR) fields has been demonstrated^{22,23} 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 XUV harmonics. Bichromatic multi-photon ionization has also been reported²⁴ with phase control, but again the phase of optical photons was controlled, not that of XUV light.

Here we demonstrate and exploit the longitudinal coherence of two-colour XUV 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 described⁸, and here we summarise the salient points of the machine, Fig. 1. Six APPLE-type undulators²⁵ can be set independently to produce polarised light at harmonics of the seed wavelength: we used horizontal linear polarisation. Between each pair of undulators, an electron delay line or phase shifter²⁶ lengthens the path of the electrons by nm scale increments, thus allowing tuning of the relative phase between the bunched electron beam and the co-propagating photon beam (see Methods). This is the key to our approach: n undulators are set to the first harmonic, $6-n$ are set to the second harmonic, and the phase shifters are used to adjust the phase difference between the harmonics. The temporal and phase profiles were theoretically simulated and both ~ 100 fs pulses overlap well (see Methods and Supplementary figure 1.)

Figure 2(a) shows the experimental set-up and Figure 2(b) a typical spectrometer image.

The $2s^22p^5(^2P^{\circ}_{3/2})4s$ 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, 31.49 nm, while 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 system²⁷ 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. 3(a). $2p$ electrons from neon can be emitted by two quantum paths: by a single photon (frequency 2ω) as an s - or d -wave; or by two photons (frequency ω) 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. Due to the non-linear nature of the process²⁸ 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 like that in equation (1). If the temporal lag between the two harmonics is Δt , the relevant parameter is the *delay-induced phase difference* $\Delta\phi=2\omega\Delta t$.

The photoelectron angular distributions were measured using the VMI spectrometer of the Low Density Matter end-station, see Methods. The 'left-right' asymmetry was quantified by the parameter A_{LR} ,

$$A_{LR} = \frac{I_L - I_R}{I_L + I_R} \quad (2)$$

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

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

To understand the asymmetry in detail, the angular distribution was fitted with Legendre polynomials²⁹, each characterised by a β parameter. The even (β_2, β_4 etc.) and odd (β_1, β_3 etc.) numbered parameters describe the symmetric and antisymmetric parts of the distribution respectively. The fits for β_1 and β_3 in Fig. 3(b) are consistent with the calculated errors, while β_2 shows a larger deviation, possibly indicating systematic errors. Comparing the data qualitatively to the calculated spectra of a simpler system, atomic hydrogen³⁰, we find the key characteristics are

reproduced: β_2 is constant while β_1 and β_3 oscillate, with a phase lag, in this case 1.06 rad. The non-zero lag already follows from lowest-order perturbation theory. For infinite pulses and neglecting non-resonant two-photon transitions (Fig. 3(a)), the lag is derived as $\arg(\frac{2\sqrt{2}}{5} - \frac{D_s}{D_d})$, where D_s/D_d is the ratio of the complex first-order ionization amplitudes into the s - and d -channels (red arrows in Fig. 3(a)). Frozen-core Hartree-Fock calculations of D_s and D_d predict a lag of approximately 0.55 rad for neon. Thus this theory, whose weak point is the single intermediate state approximation, provides only qualitative predictions for our experimental conditions.

The present result demonstrates phase control at the attosecond level with FERMI, and opens the way for unique experiments in the XUV and soft X-ray region, with complete control of the wavelength, polarization, phase and intensity. FERMI produces light with wavelengths down to 4 nm, providing access to core levels, and thus chemical specificity in coherent control experiments. This is impossible with optical lasers. The extreme time resolution may allow the study of ultrafast phenomena: two colour coherent control experiments have been used to study chemical reactions by manipulating the nuclear wave-packet, but now it is possible to shape the electron wave-packet. The present experiment was performed 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. As well, the generation of attosecond pulses and pulse trains is based on the coherent control of harmonics, and 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.

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Figure legends.

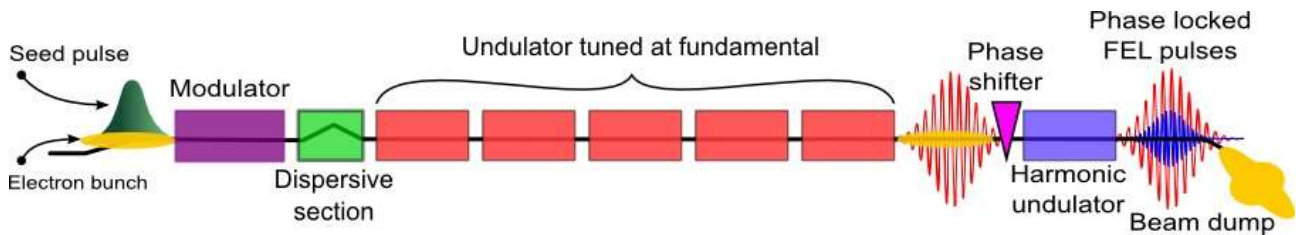


Figure 1. Scheme used in the present study. Red waves indicate schematically the first-harmonic radiation, and blue waves the second-harmonic radiation.

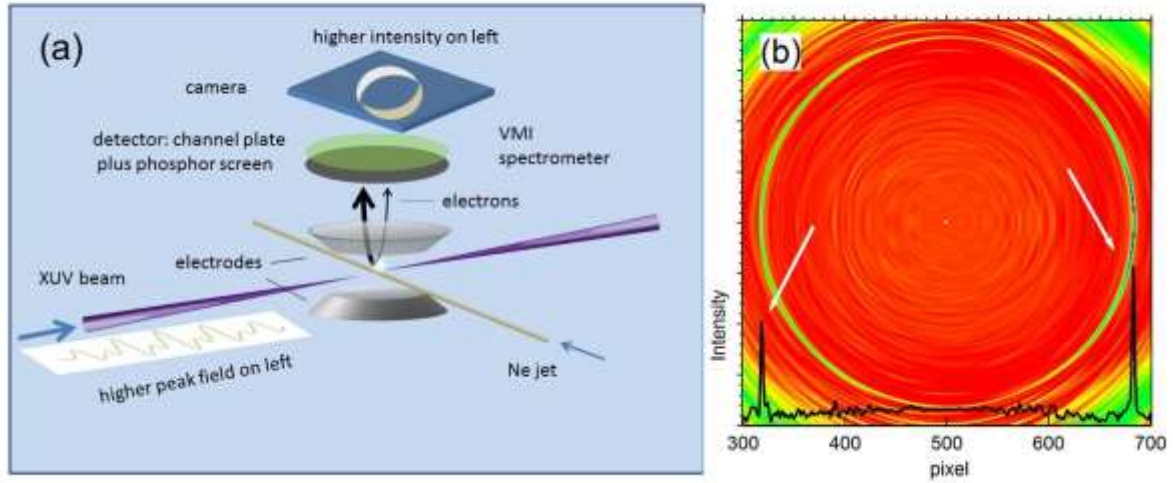


Figure 2. (a) Schematic set-up. The bichromatic light beam with fixed phase relation crosses the atomic jet of neon and ionizes the atoms. The Velocity Map Imaging (VMI) spectrometer measures the angular distribution of ejected electrons. The intensity is higher on the left or right, depending on the phase difference. (b) Typical inverted VMI image, 6000 shots. The strong, sharp ring is due to Ne $2p$ electrons, emitted by first- and second-harmonic light. A line profile across the centre of the image is shown (black line) at the bottom, demonstrating the left-right asymmetry (white arrows).

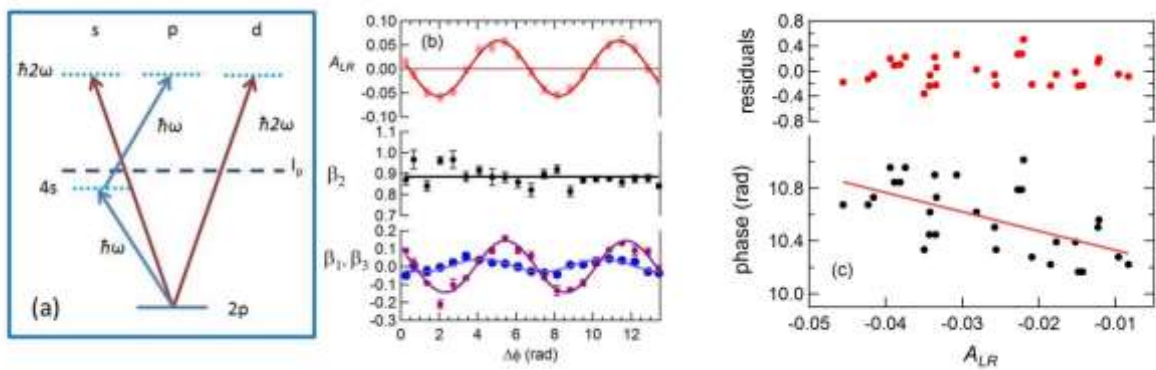


Figure 3. (a) Scheme of the present experiment. A photoelectron may be ejected as a p -wave by a two-photon process, or as an $(s+d)$ -wave by a one-photon process. (b) Asymmetry parameter A_{LR} as a function of $\Delta\phi$ (red curve), and β_1 (blue), β_3 (magenta) and β_2 (black) parameters as a function of phase. Markers: experimental data; lines: sinusoidal fits for β_1 and β_3 , linear fit for β_2 . (c) Phase

setting as a function of asymmetry parameter at the steepest part of the delay curve. Step size: 0.056 rad (900 zeptoseconds). Steps were not sequential, so the measurement includes possible errors due to hysteresis in the system, if present. The residuals (difference between the straight line fit and the data) have a standard deviation of the phase corresponding to 3.1 as.

Author contributions.

The experiment was conceived by KCP, GS, ANGG and KU, and the method of operating FERMI to carry it out was devised by EA and LG. The experiment was prepared and carried out by KCP, EA, CC, RC, GDN, SDM, BD, EF, PF, DG, LG, NM, GP, OP, LR, PR, ER, CS, MT, MZ, GS, PC, DI, YK, TT, KU, AF, FS, EO, TM, MN, MC and MM. Theoretical calculations (of machine properties or neon spectra) were performed by EA, LG, ANGG, EVG, SIS, KB, ND and JV. Detailed data analysis was performed by MR, PC and DI. The manuscript was drafted by KCP and completed in consultation with all authors.

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Author information

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