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We report on the use of pairs of 10-cycle extreme ultraviolet wave packets with attosecond-controlled spacing emitted by individual relativistic electrons within an electron bunch passing through a tandem undulator. Based on the temporal coherent control technique with circular polarization, we succeeded in controlling the excited state alignment in the photoexcitation of helium atoms, which we verified through the observation of oscillation in fluorescence yield depending on the attosecond-controlled delay time. Our work demonstrates the potential of undulator radiation for the generation of longitudinally coherent wave packets suitable for attosecond coherent control, an application which has hitherto been hidden in the incoherent nature of the radiation pulse emitted by a bunch of electrons.

Undulators are key components in most synchrotron light sources for producing energy-tunable brilliant photon beams over a wide spectral range, from the vacuum ultraviolet to hard x-rays. Since the pioneering work by Halbach [1], a considerable amount of effort has been exerted to develop novel undulators to enable unique investigations in the field of synchrotron radiation research [2]. One such development using novel undulator technology is the capability of polarization control, which is now widely used for spectroscopic studies at modern synchrotron light sources. In particular, the advent of helical undulators [3–6] has facilitated precise circular dichroism spectroscopy, used for detailed studies on the electronic and magnetic structure of materials [7] as well as for the determination of structures in biomolecules [8].

In this letter, we report on a new application of the circularly polarized radiation provided by a helical undulator at a synchrotron light source - the coherent control of excited state alignment. Our method is based on the use of undulator radiation to create longitudinally coherent wave packets with attosecond-controlled spacing. This potential use of undulator radiation has to date been hidden in the incoherent nature of the radiation pulse emitted by a bunch of electrons [9]. In our approach, we use pairs of left- and right- circularly polarized 10-cycle wave packets emitted by individual relativistic electrons passing through a tandem undulator, which consists of two identical helical undulators in series. The performance of our scheme is demonstrated by the photoexcitation of helium atoms in the extreme ultraviolet (XUV) wavelength region by using the temporal coherent control technique [10–13]. We demonstrate the creation of an atomic superposition state, with an alignment that can be readily controlled by tuning the time delay between the two wave packets at the attosecond level. We probe the alignment by observing oscillation of the fluorescence yield dependent on the delay time. Note that we use the term “wave packet” throughout this letter to refer to the emission from individual electrons [14-16] and to distinguish it from the radiation pulse emitted by a bunch of electrons.

The experiment was carried out at the 750-MeV UVSOR-III storage ring [17]. Figure 1(a) shows the experimental setup. The tandem undulator consisted of twin APPLE-II type variable polarization devices which were operated in the helical mode. The number of magnetic periods, and the period length of the undulators

are 10, and 88 mm. We set the upstream and downstream undulators to provide left-circular polarization (LCP) and right-circular polarization (RCP) at the interaction point, respectively. The central photon energy of the fundamental radiation was adjusted to ~24 eV, at which the period of the radiation is ~171 as. The spectral width of the radiation pulse was about 10%, which translates to about 10 periods in the time domain. The electron beam current was 10 mA, which corresponds to ~10^9 electrons per bunch. Each relativistic electron in the bunch which passes though the tandem undulator emits a pair of wave packets whose waveforms are well-characterized as time-separated 10-cycle oscillations, with longitudinal coherence between them. The spacing between the wave packets was tuned with attosecond precision by the phase shifter magnet which controls the electron path length between the undulators, a similar technique to that used in the optical klystron [18,19] or the crossed undulator system [20–22].

The spatially central part of the undulator radiation was selected using a 0.4-mm-diameter pinhole, and focused by a toroidal mirror to a rms size of 120 μm (horizontal) × 10 μm (vertical). A 75-nm-thick Al filter was inserted in front of the interaction chamber to eliminate visible light from the tandem undulator and bending magnets. It should be noted that the undulator radiation was not monochromatized, as for ordinary synchrotron radiation experiments. The focused undulator radiation interacted with an effusive helium gas beam. The photon flux at the focal point was around 1×10^{11} photons/s. Since the undulator radiation had a broad bandwidth, helium atoms were excited into coherent superposition states made up from both bound and continuum states (see Fig. 1(b)). We selected the 1s6p state from the coherently excited bound states by detecting fluorescence photons of 345-nm-wavelength using a photomultiplier tube equipped with a band pass filter and polarizer. The photomultiplier tube was arranged to detect fluorescence photons emitted parallel to the *y*-axis (as defined in Fig. 1(a)). The polarization axis of the polarizer was set to parallel to the *x*-axis.

Figure 2(a) illustrates the helium atoms interacting with the radiation pulses from the tandem undulator. The duration of each wave packet pair is a few femtoseconds. The pairs are randomly distributed within the overall pulse length of 300 ps (fwhm), which arises from the electron bunch length [9]. We note that the waveform shapes and the time delay τ are common for all wave packet pairs emitted from each individual electron. Therefore the individual light-atom interaction should be equivalent to that in the phase-locked double pulse scheme which has been used for coherent control of physical and chemical processes using optical lasers [13]. This implied possibility of the coherent control of atomic states using tandem undulator radiation has until now been unrecognized, due to the incoherent nature of the undulator radiation emitted by a bunch of randomly distributed electrons.

The scheme for coherent control using pairs of cross circularly polarized wave packets is shown in Fig. 2(b). The quantization axis is chosen to be parallel to the direction of light propagation. As a result of the sequential interaction with the wave packets, helium atoms are excited to the 1s6p coherent superposition state (transition frequency of ω), which consists of the M_j=±1 magnetic substates associated with the first (LCP) and second (RCP) parts of the wave packet pair, following the selection rules for dipole transitions. In an orbital picture, the $6p_{\pm}$ orbitals are coherently superposed. The phase difference ωτ between the magnetic substates can be precisely controlled by varying the time delay between the wave packets of opposite polarization.

Figure 3 shows the fluorescence yield measured as a function of this delay time, revealing a clear sinusoidal modulation. Note that, for obtaining the absolute delay time τ between the radiation wave packets, the minimum delay time $\tau_0 \sim 2.4$ fs, resulting from the slippage in the undulator section and the electron travel in the drift space, has to be added to the horizontal axis of Fig. 3. We interpret this oscillation in terms of the alignment of the 6*p* orbital controlled by the sequential interaction between the pair of wave packets and the helium atom. Initially the LCP wave packet prepares the $6p_+$ orbital, which rotates counter-clockwise around the *z*-axis. The delayed RCP wave packet then couples the $6p_-$ orbital, which rotates clockwise and overlaps coherently with the previously prepared $6p_+$ orbital. The coherent superposition of $6p_{\pm}$ orbitals forms a 6*p* orbital aligned in the *xy*-plane, with an angular distribution which can be written as

$$\left|-Y_{1+1}(\theta,\phi)+e^{+i\omega\tau}Y_{1-1}(\theta,\phi)\right|^2 \propto \sin^2\theta\cos^2\left(\phi-\frac{\omega\tau}{2}\right), \quad (1)$$

where $Y_{lm}(\theta,\phi)$ are the spherical harmonics. Therefore the orbital alignment is controlled by the delay time,

where the tilt angle is set to ωτ/2 with respect to the *x*-axis. The detector setup allows us to determine the alignment of the 6*p* orbital because the fluorescence photons are preferentially emitted perpendicular to the alignment axis. Reflecting the spatial distribution of the dipole emission as shown in the top panel of Fig. 3, we can expect the signal intensity of the fluorescence detection to obey $I(\tau,\omega) \propto \cos^2\left(\omega\tau/2\right)$, and oscillate at the resonant frequency of the 1s6p excited state depending on the delay time. The observed oscillation shows high visibility and is well reproduced by a sinusoidal curve. This supports the applicability of circularly polarized undulator radiation to control excited state alignment based on the temporal coherent control technique.

For comparison with the experimental results, an ideal sinusoidal curve was convolved with a Gaussian function (σ=30 as) representing the effective temporal resolution in the delay time control. By removing the instrumental effect of the finite detection angle on the measurement, the actual temporal resolution was estimated to be 26 attoseconds. From the viewpoint of alignment control a temporal resolution of 26 attoseconds limits the angular resolution to approximately 26 degrees. We estimate a systematic error of roughly 4.5 degrees (rms) in the alignment angle, arising from the reflectivity of the focusing mirror which depends on the polarization state. The temporal resolution achieved is slightly worse than those obtained in other methods in the XUV spectral region, which include 15 attoseconds (rms) using a seeded FEL [24] and 7.5 attoseconds with a delay-line device [25].

In our approach the visibility of the oscillation is limited mainly by the properties of the electron beam, such as energy spread, angular divergence, and the stability of the phase shifter magnet. Using a similar approach to that used to evaluate the spectral broadening of optical klystron radiation [18,19] and the reduction of polarization in the crossed-undulator [20], we estimate that the actual temporal resolution, as limited by the light source conditions, was actually around 10 attoseconds in the present study. We can thus anticipate improvements in the temporal resolution of our scheme. A possible explanation for the discrepancy between the experimentally observed resolution and the light source limit could be a misalignment between the radiation emitted from the two undulators and the finite size of the pinhole, which would reduce the visibility of the oscillation. Assuming the Gaussian beam profiles, we can estimate that the transverse offset of the beams is lower than $1.3\sigma_{x,y}$ at the interaction point, where $\sigma_{x,y}$ are sizes of the focused beam.

We now discuss the applicability of our method at shorter wavelengths. Since an excited state of concern has to interact with a radiation wave packet pair, the total temporal duration of the wave packet pair should be shorter or at least comparable to the lifetime of the excited state. Such a situation for an K-shell or L-shell excitation in most elements can be reached using a 3-GeV class synchrotron light source [26]. For instance, while the natural width of the L_2 shell (binding energy of about 21 keV) in U (Z=92) is 70 attoseconds [27], the minimum temporal durations are estimated to be 28, 36 and 44 attoseconds in the fundamental, third and fifth harmonic radiation, respectively, assuming a pair of the 21-keV wave packets from a 3-GeV electron beam passing through 0.5-m separated 10-period undulators.

The excitation with a shorter wavelength requires a more precise control of the delay length for tuning the phase difference between the two radiation fields. Unlike the optical control with a delay-line device, we control the phase difference by a motion of a relativistic electron. The phase difference is determined by the ratio between the delay length and the radiation wavelength, and both are inversely proportional to the square of the beam energy. Therefore the accuracy in controlling the phase difference does not depend on the beam energy nor the radiation wavelength [18, 20].

Other technical constraints relate to the electron beam properties. The energy spread and emittance may smear the phase difference. To avoid this, as shown in previous works [18, 20], the energy spread should be lower than the inverse of the number of undulator periods. This condition holds even at high energy synchrotrons since the energy spreads are typically 10^{-3} or smaller and the number of undulator periods is usually designed to be around 100. On the other hand, the effect from beam emittance becomes more significant for shorter wavelength. It can be shown that this constraint is equivalent to so-called "diffraction limit" [28]. Therefore the diffraction limited light source based on ultimate storage rings or energy recovery linacs becomes the best candidate for performing the coherent control experiment in the hard x-ray regime. While at present such a low emittance synchrotron light source has not

been available in the hard x-ray region, a pinhole technique which has been widely used in the x-ray imaging for improving the spatial coherence of the light beam [29, 30] enables us to sufficiently suppress the emittance effect. We thus conceive that our method has the potential feasibility of coherent control even in the hard x-ray region.

In summary we have demonstrated a new application of circularly-polarized radiation from a synchrotron light source which is based on the hidden potential of undulator radiation as longitudinally coherent wave packets with attosecond-controlled spacing. By using pairs of cross-circularly polarized 10-cycle wave packets emitted by individual relativistic electrons within a bunch, we succeeded in controlling the alignment of the excited state in the photoexcitation of helium atoms at XUV wavelengths. Our method is a new route to attosecond coherent control in the XUV region [31–33], and is extendable to shorter wavelengths as far as hard x-ray. This new capability of undulator radiation will open the possibility of probing and controlling ultrafast phenomena in a wide range of atomic and molecular processes as well as in a variety of materials, taking the advantage of the short wavelength radiation such as the chemical selectivity in excitation processes [34]. In addition, from a view point of fundamental atomic and molecular physics, the description of short wavelength coherent control involving inner-shell electrons possesses theoretical challenge for many-electron approaches even in isolated atoms [34], and this method will provide benchmark outputs for the unexplored area.

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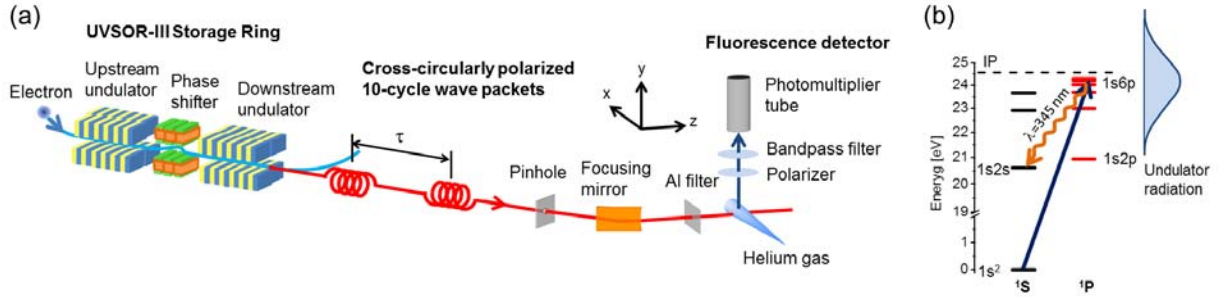

FIG. 1. (a) Experimental setup for temporal coherent control in the photoexcitation of helium atoms. The tandem undulator consists of two helical undulators and each relativistic electron in the bunch emits a pair of 10-cycle XUV cross-circularly polarized wave packets. The energy spread and beam divergence of the electron beam are $\Delta E/E \sim 5\times10^{-4}$, and $\sigma_{ex}' \sim 43$ μrad × $\sigma_{ey}' \sim 26$ μrad, respectively. The delay time between the wave packets is controlled by the phase shifter magnet between the two undulators. The stability of the magnetic field in the phase shifter is 0.1%. The pinhole and the focusing mirror are located, respectively, at 9 m and 10 m downstream the middle point of the two undulators. The incidence angle of the focusing mirror is 80 degrees and the focal point locates at 2 m downstream the mirror. The undulator radiation interacts with a helium gas beam at the focal point, and fluorescence photons are detected by the photomultiplier tube. (b) Helium atoms are coherently excited into an atomic superposition state. The fluorescence decay from the 1s6p state to the 1s2s state is selectively detected.

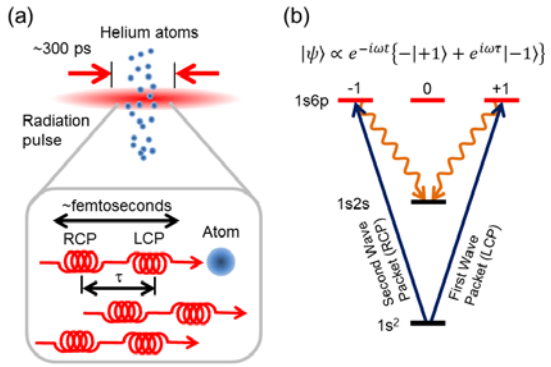

FIG. 2. (a) Interaction between the cross-circularly polarized wave packet pair and single atoms. While the radiation pulse consists of numerous wave packet pairs randomly populated in the radiation pulse, each atom interacts with one of the identical wave packet pairs. (b) The helium atom is excited into the 1s6p superposition state which consists of the M_j=±1 substates associated with the first (LCP) and second (RCP) parts of the wave packet pair.

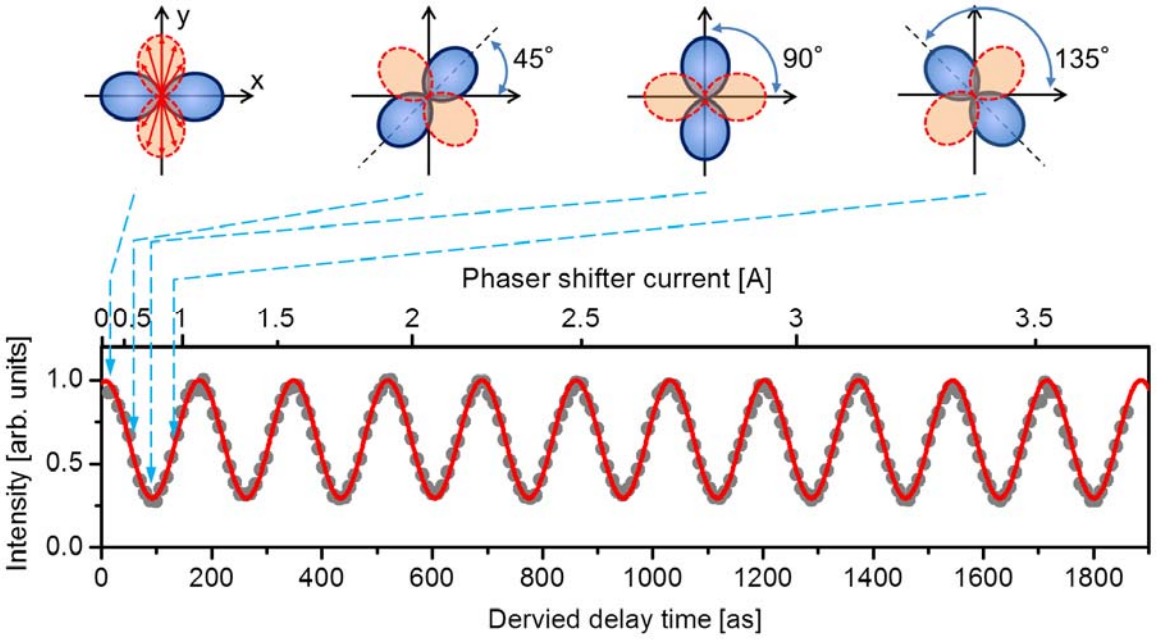

FIG. 3. Fluorescence yield as a function of wave packet delay in decays from the 1s6p to the 1s2s states. The gray circles are measured data and the red line shows the theoretical curve in which the 30 attosecond time resolution is taken into account. The derived delay time in the horizontal axis corresponds to the delay produced by the phase shifter magnet. The delay time produced by the phase shifter magnet was calibrated using the sinusoidal curve which oscillates at the resonant frequency of the 1s6p state [23], assuming that the delay is proportional to I^n where I is the magnet current. In the fitting, the initial phase of the oscillation was determined to reproduce the measurement and we obtained $n = 1.97$ which agree well with the theoretical value of $n = 2$ [18]. The current of the phase shifter magnet is shown in the top horizontal axis. Polar plots of the aligned 6*p* orbital (blue) and dipole emission pattern (red) in the *xy*-plane are also shown.