Ultrafast direct imaging using a single high harmonic burst

Kyoung Hwan Lee,1,2 Seung Beom Park,1,2 Himanshu Singhal,1,2,3 and Chang Hee Nam1,2,4,*

1Department of Physics, KAIST, Daejeon 305-701, South Korea
2Center for Relativistic Laser Science, Institute for Basic Science, Gwangju 500-712, South Korea
3Laser Plasma Division, Raja Ramanna Centre for Advanced Technology, Indore 452-013, India
4Department of Physics and Photon Science, GIST, Gwangju 500-712, South Korea
*Corresponding author: chnam@kaist.ac.kr

Received February 6, 2013; revised March 14, 2013; accepted March 15, 2013; posted March 15, 2013 (Doc. ID 184747); published April 4, 2013

We have demonstrated ultrafast direct soft x-ray microscopy imaging. Microscopy images were acquired using an objective zone plate and a single strong high harmonic burst generated from He in a two-color laser field. This zone-plate-based microscopy system delivered real-space images directly without any data processing. The spatial resolution of the microscope was estimated to be about 140 nm from the image of nanoscale grating patterns. © 2013 Optical Society of America

OCIS codes: 180.7460, 320.7080, 140.7240.

Realization of microscopes with femtosecond temporal resolution and nanometer spatial resolution is an important task for investigating the ultrafast dynamics of microscopic objects. High-resolution soft x-ray microscopes coupled with ultrashort, strong illuminating sources are promising tools to achieve this goal. In the respect of spatial resolution, there have been intensive efforts to improve it. Fabrication of a sophisticated Fresnel zone plate (FZP) based on electron beam lithography showed the spatial resolution of ~10 nm and multiple enhancements to the ability of the FZP was shown by the high-order focusing of the FZP [1,2]. As different approaches, lensless imaging methods, such as coherent diffraction imaging and Fourier-transform holography, were proposed to overcome the limitations of lens-based microscopes [3,4]. In principle, the lensless imaging methods, applied to the far-field diffraction patterns of a compact, isolated object, can achieve a spatial resolution comparable to the wavelength of a light source. However, difficulty in forming a reference signal or time-consuming data processing in the phase recovery for image reconstruction still makes zone-plate-based microscopes attractive to soft x-ray microscopy imaging.

Single-shot imaging using an ultrashort and brilliant light source makes it possible to observe ultrafast dynamics. With the various types of x-ray sources, e.g., a plasma discharge laser, an x-ray free electron laser, and high-order harmonics, single-shot imaging has been performed [5–8]. In particular, high harmonics have great potential due to its subfemtosecond pulse duration, coherent nature, and broad spectral range [9–11]. Furthermore, high harmonics as a table-top soft x-ray source have advantages in accessibility and applicability compared to large facility x-ray sources. In spite of these advantages high-resolution single-shot direct imaging using soft x-ray from high harmonic generation (HHG) has not been achieved due to the low photon count of high harmonics and low photon efficiency of an x-ray microscopy system. Thus, ultrafast real-space imaging might be realized with improved conversion efficiency of HHG and improved photon efficiency of the microscope.

In this Letter, we present ultrafast direct imaging by adopting a strong high harmonic source with femtosecond pulse duration and an efficiency-enhanced zone plate. In order to generate brilliant high harmonics, HHG in the two-color laser field, consisting of the fundamental and second harmonic of a femtosecond laser pulse, was employed, allowing drastic conversion efficiency improvement over that using a single-color laser [12,13]. Strong high harmonics from the 34th order to 42nd order could be generated from helium driven by the two-color laser field. In this study, the 38th harmonic at 21.6 nm was selected as the illumination source. The low focusing efficiency of zone plates, being one of the primary obstacles for single-shot imaging, could be improved by adopting a phase-reversal zone plate (PRZP) having phase-shift zones instead of opaque zones used in the conventional FZP [14].

For single-shot imaging a very strong illumination source is required. Since HHG in the two-color laser field has been proven to be very efficient [13], it could be suitable for the illumination of single-shot imaging. The two-color laser field was generated by placing a 200 μm-thick beta barium borate crystal in the beam path of focused 26 fs, 3 mJ Ti:sapphire laser pulses at 820 nm. Strong HHG was achieved in a 6 nm He gas jet placed 10 mm before the focal position of a concave mirror with 60 cm focal length. Among generated harmonics, the 34th and 38th harmonics were the strongest. The 38th harmonic at 21 nm, adopted as the illumination source of the microscope, contained about $1 \times 10^{10}$ photons with a relative bandwidth ($\Delta \lambda / \lambda$) of 170, where $\lambda$ and $\Delta \lambda$ are the wavelength and bandwidth of the light source, respectively.

A soft x-ray microscope for ultrafast imaging was constructed using the high harmonic at 21 nm and the PRZP, as shown in Fig. 1. The copropagating two-color laser field was effectively suppressed using a 150 nm thick Al filter and a 2 mm aperture. The Al filter and its mount, entirely covering the entrance of the microscopy chamber, blocked any stray light coming from the HHG chamber. Among the high harmonics the 38th harmonic was selectively reflected using a pair of Mo/Si multilayer...
mirrors. The second Mo/Si multilayer mirror was a concave mirror with \( f = 20 \) cm, working as a condenser to focus the 38th harmonic into a sample. The light propagated through the sample was imaged by the PRZP on a soft x-ray CCD with 2048 × 2048 pixels. We chose polymethyl methacrylate (PMMA), a popular photoresist material used in electron beam lithography, as the PRZP material because of its focusing efficiency (18%), stability, and simplicity in fabrication [15]. The PMMA PRZP consisted of 400 zones with 740 μm focal length and 100 nm outermost zone widths. The constructed microscope had a magnification of 340.

In order to measure the spatial resolution of the microscope, periodic test samples were prepared. Freestanding gratings, having half-periods from 100 nm to 1 μm, were fabricated on a 100 nm thick Si3N4 membrane coated with 20 nm Pt. The portion of Pt layer corresponding to the transmission line was removed using focused ion beam milling. Figure 1 also contains a scanning electron microscope (SEM) image of the fabricated grating with a 1 μm half-period, and its magnified view is shown in the inset of Fig. 2(a). The black, dark gray, and bright gray regions in the inset image are the hole, Si3N4, and Pt areas, respectively. The dark gray area is the portion of the bar pattern in which the Pt coating was partially erased by the ion beam. The partially erased area makes the transmissive linewidth look thicker than the opaque line in the SEM image. However, in the soft x-ray imaging, it is considered opaque, since the photon absorption is almost 90%. Thus, the grating worked properly as a test sample with a one-to-one duty period.

The single-shot capability of the microscope was examined while changing the number of integrated shots on CCD. In the experiment, a 2 × 2 binning of CCD pixels, corresponding to an effective pixel size of 80 nm at the sample plane, was used in order to boost the measured signal level per pixel. Using a single-shot exposure as well as multi-shot, images with high contrast were successfully acquired. Figures 2(a) and 2(b) show microscopy images of the 1 μm half-period grating acquired with three-shot and single-shot acquisitions, respectively. Though it showed clear periodicity, in three-shot acquisition, the grating image was slightly blurred, possibly due to mechanical vibration causing a slight movement of the sample and the zone plate with respect to the harmonic light between each shot. In contrast, the single-shot image precisely showed the grating patterns with the an effective field of view of 6 μm in diameter.

We evaluated the spatial resolutions in two cases by using intensity profiles, as shown in Fig. 2(c). A red curve (circles) and a blue curve (triangles) were obtained along the white lines in Figs. 2(a) and 2(b), respectively. Since the Rayleigh criterion corresponds to a 10%–90% intensity variation along a well-defined edge, the spatial resolutions were determined as 140 and 300 nm for the single-shot and three-shot acquisitions, respectively. To clarify the measured resolution of the single-shot imaging, two gratings with 100 nm and 200 nm half-periods were imaged with single-shot exposures. In the case of 100 nm half-period grating, which had a finer structure than the spatial resolution, each transmissive line could not be identified, as shown in Fig. 3(a). On the other hand, the 200 nm half-period grating, showed clear grating images, as shown in Fig. 3(b). Therefore, the expected resolution, better than 200 nm, is consistent with the resolution of 140 nm previously measured by the edge transition.

The most valuable advantage of single-shot imaging is in its temporal resolution. In the case of harmonics generated from Ar with 30 fs laser pulses, the envelope duration of the harmonic pulse, measured with the complete reconstruction of attosecond burst technique, was as short as 7 fs [16]. We expect that the temporal resolution obtained with the two-color harmonic light source case would be comparable, being much shorter than the laser pulse duration. With the single-shot imaging, the dynamic change of a sample can be traced with
femtosecond resolution. For example, it can be applied to the dynamic observation of large complex samples, such as proteins and bacteria, that are difficult to observe with lensless imaging methods without degradation of spatial resolution or contrast. Another example would be the measurement of ultrafast magnetization dynamics by making use of the large magnetic circular dipole effect of 3d transition metals. Thus, this full-field soft x-ray microscope coupled with a table-top high harmonic light source will be a powerful tool for the dynamic measurement of biomolecular and magnetic samples.

In summary, direct single-shot soft x-ray imaging with a femtosecond temporal resolution was demonstrated using a high harmonic light source. This device was constructed using a bright high harmonic at 21 nm, generated from He in the two-color laser field, and a PMMA PRZP. A clearly resolved grating image was obtained with 140 nm spatial resolution in a single-shot acquisition. Since the single-shot soft x-ray microscope can provide images of an ultrafast event with femtosecond resolution, the dynamic change of ultrafast phenomena can be revealed. Consequently, the single-shot direct soft x-ray imaging can open a new horizon in nanoscience requiring femtosecond resolution.

This work was supported by the Ministry of Education, Science and Technology of Korea through the National Research Foundation and through the Institute for Basic Science.

References