# Carrier envelope phase effects on polarization gated attosecond spectra

Mahendra Man Shakya, S.Gilbertson, Hiroki Mashiko, C.Nakamura,C. Li, E.Moon, Z.Duan, Jason Tackett, and Zenghu Chang[a]
J.R.Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, KS 66502

## ABSTRACT

Polarization gated high harmonic generation in argon gas was phase matched to produce a single or double pulses with $10^4$ photons. It was accomplished by optimizing the argon gas pressure. The spectrum interference of the two pulses is affected by the carrier-envelope phase like in Young's experiments. The XUV flux is sufficient for measuring the single shot XUV spectrum in the 33eV-55eV photon energy range. The spectral profile was a super-continuum for some shots and showed discrete high harmonics peaks for other shots. The carrier-envelope phase of pulses from grating-based chirped pulse amplification was also varied smoothly to cover a $2\pi$ range by controlling the grating separation. It is demonstrated that XUV spectra measures both the absolute value of the phase and the stability of the phase by measuring the phase with an *f-to-2f* setup and by the variation of XUV spectra from polarization gated high harmonics generation.

**Keywords:** Polarization gating, Carrier-envelope phase, harmonics generation, chirp pulse amplification

## 1. INTRODUCTION

Development of light sources that produce a single attosecond pulse per laser shot is very important for studying electron dynamics in atoms and molecules using the pump-probe technique [1, 2]. Comparing to the generation of 250 attosecond pulses at the cutoff region of the high harmonic spectrum with few-cycle pulses, gating the harmonic generation process by a laser field with a time dependent ellipticity may produces pulses with a duration approaching one atomic unit (25 attosecond) [3-5]. A single attosecond in time domain correspond to a extreme ultraviolet (XUV) super-continuum in the spectrum domain. The super-continuum has been observed experimentally when the gating pulses were formed with few-cycle laser pulses [6]. In this case, the linearly polarized portion of the driving laser field is less than one optical cycle and the XUV spectra are sensitive to the shot to shot variations in the carrier envelope (CE) phase of the laser pulses.

Previous numeric simulation predicted that the change of the CE phase of the driving laser pulse can result in either single or double electron ion re-collision in the harmonic generation process [7]. Single re-collision produces an XUV super-continuum that resembles a single slit diffraction pattern while double re-collision generates a modulated XUV spectrum that resembles a two slit diffraction in space. The dependence of the polarization gated high harmonic spectra on the carrier envelope phase of driving laser pulse can be easily understood in the time domain. The laser pulse with a time dependent ellipticity can be decomposed into two orthogonally polarized components. The electric field of one component is shown in Fig.1. This is the component that contributes to the harmonic generation. The amplitude of the other component is very small around time zero as indicated by the value of the ellipticity. Here we assume that the duration of the circularly polarized pulses is 5 fs and the time separation between them is 7.5 fs. The center wavelength of the laser pulse is 750 nm that corresponding to a optical cycle of 2.5 fs. The carrier envelope phase of the laser pulse is defined as the offset of the electric field with respect to the time when the ellipticity is zero.

The time range within the two vertical dot lines is the so called polarization gate, where the ellipticity of the laser pulses is small (< 0.3) that allows efficient harmonic generation. In experiments, the gas target can be placed after the laser focus to select the short trajectory, which is the trajectory considered here. When the CE phase is 90 degree as in Fig.1 (a),

[a]chang@phys.ksu.edu

Ultrafast X-Ray Sources and Detectors, edited by Zenghu Chang, George A. Kyrala, Jean-Claude Kieffer,
Proc. of SPIE Vol. 6703, 67030H, (2007) · 0277-786X/07/$18 · doi: 10.1117/12.737600

only electrons released between t = -0.125 fs to t = 0 can return within the gate as indicated by arrow. As a result, one attosecond pulse is generated that corresponds to a super-continuum. As a contrary, for CE phase equals 0 degree as shown in Fig 1 (b), two groups of electrons with short trajectories are released within the gate, one returns within the gate, the other returns outside the gate but the ellipticity is still small. In this case, two attosecond pulses are produced and the second one is weaker. The interference between the two pulses yields modulated harmonic spectra. As the CE phase changes from zero to 90 degree, the amplitude of the second pulses decreases. Thus the spectral modulation depth also reduces till the spectrum evolves into a super-continuum.

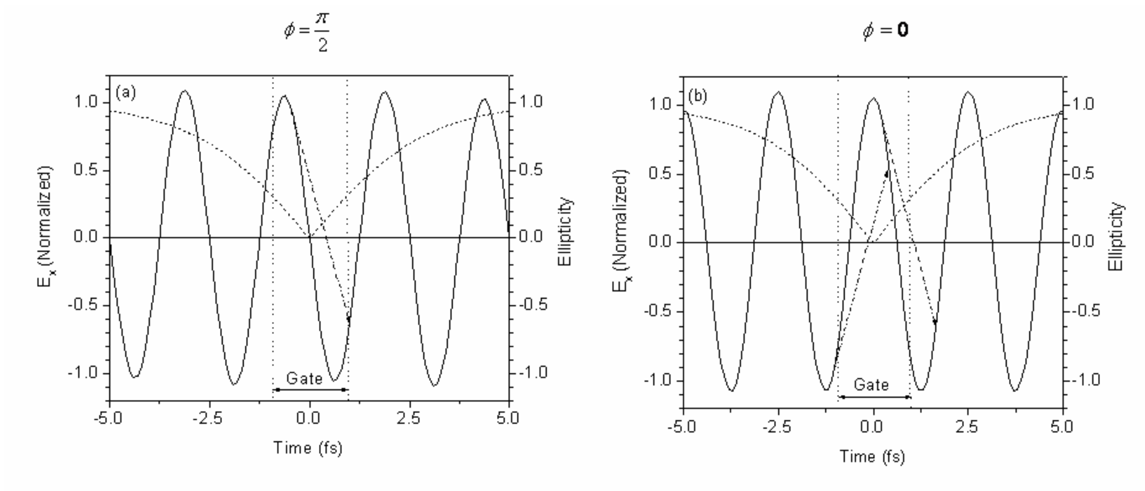


Fig.1 The effects of the carrier-envelope phase of the driving laser pulse on the returning of the ionizing electron within the polarization gate. The carrier envelope phase is 90 degree in (a) and is 0 degree in (b). The solid line is the electric field of one component of the laser pulse. The dashed line is the ellipticity. The range within the two vertical dotted line is the polarization gate.

If the XUV spectrum can be measured and analyzed for every laser shot, then it is possible to determine the shot to shot variation of the CE phase. The measurement of the single shot CE phase is important for studying CE phase effects in many experiments when the CE phase of the few-cycle laser pulses are not locked. Previously, CE phase has been measured by *f-to-2f* interferometers and phase meters based on above-threshold ionization [8]. The former only provides the relative phase value while the later can not measure phase in a single shot. In this work, we study the effect of CE phase on the spectra of the polarization gated high harmonics in order to develop an all optical scheme for determining the carrier envelope phase of laser pulses. Furthermore, the phase-matching in the gating process was studied in an attempt to boost the number of XUV photons for single shot CE phase measurement.

## 2. EXPERIMENTAL SETUP

### 2.1 Setup 1: To study shot-to-shot effect of CE phase on polarization gated XUV super-continuum

In order to record shot to shot effect of CE phase on XUV spectrum, experimental set up should be designed in such a way that it can generate sufficient extreme ultraviolet (XUV) photons. As the numbers of photons that can be produced are limited, the next step for the better measurement of single shot CE phase should be finding a ways to prevents photons from loosing during experiment. Although harmonics energy scales with increase in the intensity $(q_{cutoff} = I_p + 3.2I\lambda^2)$, highest laser energy that makes sense to use is the saturation intensity because at this intensity all the neutral atoms are used up by ionization. It also does not lead to the efficient harmonics by ionizing all the neutral atoms. One reason for this is the plasma induced defocusing because the plasma has negative refractive index. The other reason is the phase matching is very difficult at higher intensity. Hence, these are the critical parameters which one should essentially be aware of while designing the experimental setup to study the single shot measurement of absolute CE phase from the XUV spectrum.

In order to generate harmonics, argon atom was taken as the microscopic nonlinear medium in this entire work. Reason for making this choice was requirement of lower energy for generating harmonics, which polarization gating with thicker quartz plate can deliver. The experimental arrangement is shown in the Fig.2. Briefly, to avoid the dispersion and

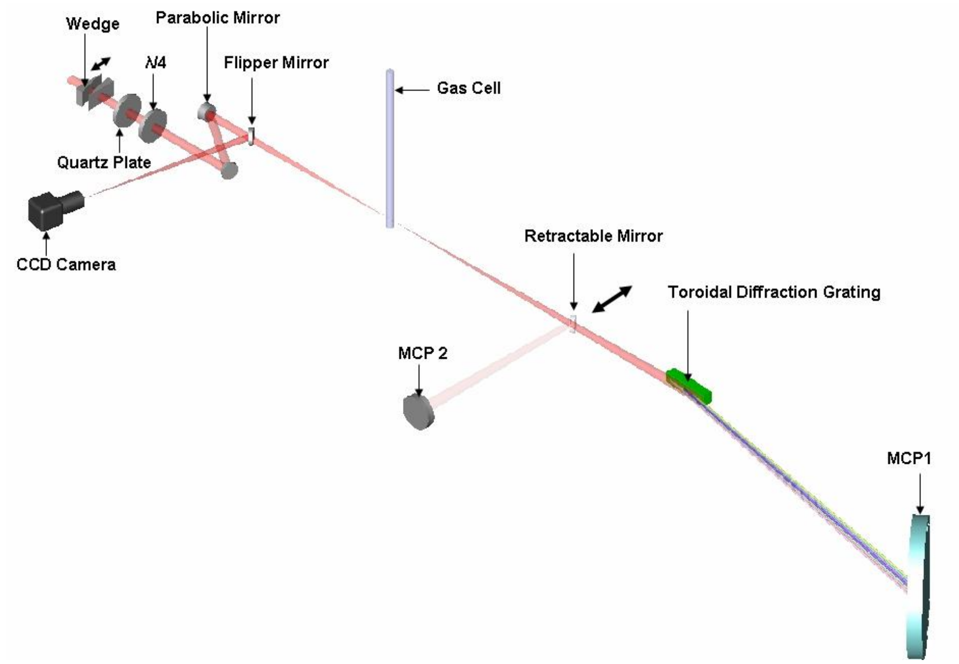


Fig.2 Experimental setup to study effect of shot-to-shot change in CE phase of driving pulse on XUV super continuum generation.

chromatic aberration, an off axis parabolic mirror of focal length 250 mm was used to focus the laser pulses to the target. Gas cell inside the chamber is a thin-wall tube which has two pinholes drilled in it by the laser itself, allowing the beam to pass through the gas inside, but confining the gas to a length about that of the tube internal diameter 1.44 mm. The diameter of the hole is comparable to the focal spot size, ~ 60 $\mu m$ diameter. The gas inside the tube can be varied from 0 Torr to 200 Torr with less than 200 mT in the background vacuum chamber when at the highest target pressure. At about 10 inch from the focus, an aluminum filter of 100 nm thick is mounted on the stainless steel gate valve (Mini UHV Gate Valve Series 010). At this distance the laser beam has diverged to a low intensity, and damage to the filter is very unlikely. Since, laser spot size at this distance is ~4.4 mm, which is quite smaller than the diameter (10 mm) of the filter, beam clipping therefore never occur for the well centered beam. The Al filter therefore never reduce throughput of the spectrometer by blocking photon from reaching the grating surface. On the contrary, aluminum filter blocks laser and low order harmonics (only high order can be used for polarization gating) and serves as a shield to prevent the gas flow from interaction chamber to the detector chamber. This is essential in order to maintain pressure as low as $10^{-8}$ Torr thereby preventing electrical break down of the MCP detector and reduce absorption of photons by gas.

The generated XUV beam is incident at an incident angle 86.262 degrees on the high efficiency, flat field XUV toroidal grating (JOBIN YVON HORIBA) placed at 23 inch away from the focus which serves as an image distance. The grating is 7.3 cm long and 1.3 cm wide with 384groves/mm provides the wavelength range from 11nm to 35nm (35 eV-110 eV). The vertical and horizontal acceptance angle of the grating from the focus is therefore 8.1mrads and 21.7 mrads respectively. In order to match this acceptance angle with the divergence angle of the laser, incident beam size on the focusing mirror was made 3.4 mm diameter which expands to 10 mm as it reaches the grating surface.

### 2.2 Measurement of number of photons per laser shot

XUV photons transmitted from aluminum filter can also be reflected at an angle of 45 degrees by a retractable bare gold mirror into the second microchannel plate (Fig.2). This allows us to estimate the divergence angle of the XUV beam. Fig.3 shows the far field image and the intensity profile of the harmonic spot image on the second microchannel plate (MCP2 in Fig.2) detector. The grid pattern in the image is from the mesh that supports the thin aluminum filter. The spot has a half angular width of 3.5 mrad at the $1/e^2$ criterion and was calculated from the divergence of the harmonic beam assuming a Gaussian intensity distribution. The divergence angle matches well with the acceptance angle of the diffraction

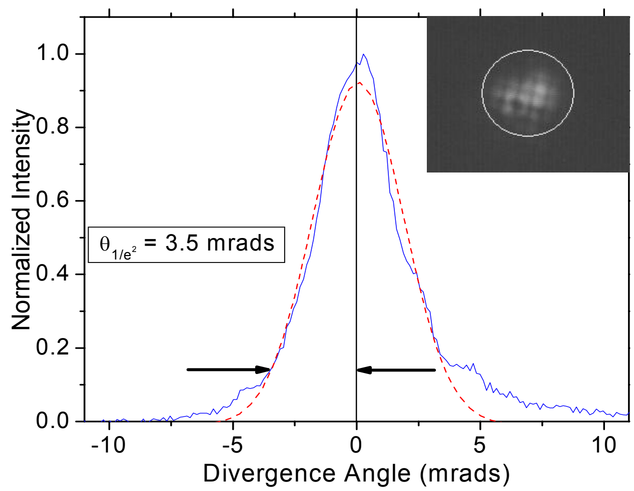


Fig.3 Far field image of XUV and its line out. Divergence angle at $1/e^2$ criterion is 3.5 mrad which matched with the acceptance angle of the grating used.

grating which is 8.1 mrad in the horizontal direction and 27.1 mrad in the vertical direction. Since, the number of photon per laser shot is low, it is important to match the angles to reduce the detection loss for suppressing the statistical noise of the measured single shot harmonic spectrum. The divergence angle of the harmonic beam is much smaller than that of the laser beam 8.1 mrad. The ratio is a useful number for designing beam lines that uses the harmonic pulses for pump probe experiments. The number of harmonic photons per laser shot can also be estimated by using the same microchannel plate. An ammeter which connects MCP rear and the ground reads the difference between the current with and without the presence of XUV on the MCP. If number of photons per laser shot generated is N, it can be written as,

$$N = \frac{\Delta I}{Ge f_{rep} R_{Al} Q T_{Al}} \tag{1}$$

Where $\Delta I$ is the difference in current with and without the presence of XUV on MCP. $G, e, f_{rep}, R_{Al}, Q$ and $T_{Al}$ are MCP gain, charge of an electron, repetition rate of laser, reflectivity of Al filter, quantum efficiency of MCP and transmissivity of the aluminum filter respectively. When MCP voltage was 1.8 kV, $\Delta I$ was 0.04 $\mu A$. The value of $G, e, f_{rep}, Q$ , $T_{Al}$ (for 27$^{th}$ harmonics) and $R_{Al}$ are $5.3\times10^6$, $1.6\times10^{19}$ C, 1000, 0.1, 0.8 and 0.02 respectively. With those values, expression (1) gives 30,000 photons per laser shot at 55 Torr target pressure.

### 2.3 Measurement of phase matching pressure

As photons can easily be lost by several different processes mentioned above, careful investigation for finding the way to prevent the loss is a crucial part of this experiment. Every possible effort to boost the number of photon has been carefully made when the apparatus was designed and built. Since, our design has XUV divergence angle matched with the acceptance angle of the grating; throughput of this spectrometer cannot be poorer than any design claim to have the best throughput, which does not have those two angles (divergence angle and acceptance angle) matched. Besides, possibility of photon loss through absorption by gas has almost been completely eliminated by keeping the pressure as low as $10^{-7}$ torr in the rest of the chamber.

Although, it is necessary to have design capable of preventing photon loss, it is not sufficient to expect good output signal from the single shot experiment unless sufficient number of photons are produced. It has been shown experimentally that number of photons can be enhanced quadratically by increasing pressure. However, we should not assume that harmonic efficiency can be increased as high as we want since the quadratic variation in intensity continues only up to certain value of pressure. As this value exceeds, harmonic efficiency degrades due to phase mismatching. It might therefore seem extremely important to find out the maximum pressure that can be used without losing the phase matching condition. As our design provides knob to tune pressure at the target, we took that advantage to measure intensity of harmonics at different target pressure. This leads us to the identification of the maximum pressure, which does not effect negatively on the phase matching condition. Fig. 4 is the measurement of the polarization gated harmonic

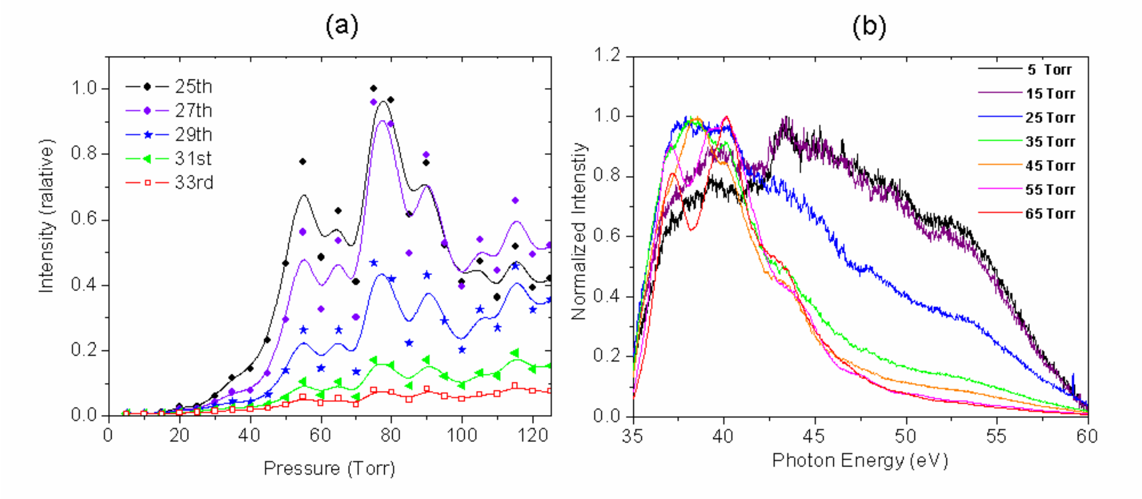


Fig.4 (a) the dependence of the harmonic intensity on the target gas pressure. (b) The polarization gated high harmonic spectra at different pressures. Each spectrum was accumulated over 500 laser shots.

intensity with pressure. All the harmonics intensity scaled quadratically over the increase in pressure up to 55 torr. Beyond this value variation is no longer quadratic. We believe that at this optimized pressure, the phase matching is achieved by the cancellation of the phase mismatch due to the plasma dispersion and the Guoy phase shift by the variation of the harmonic phase due to decrease of the laser intensity and the electron density in the laser propagation direction. Fig. 4(b) is the haimonic intensity as a function of photon energy at different target pressures. Decrease in the flat continuum portion with the increase in the pressure can be accounted for the decrease in intensity due to the plasma defocusing.

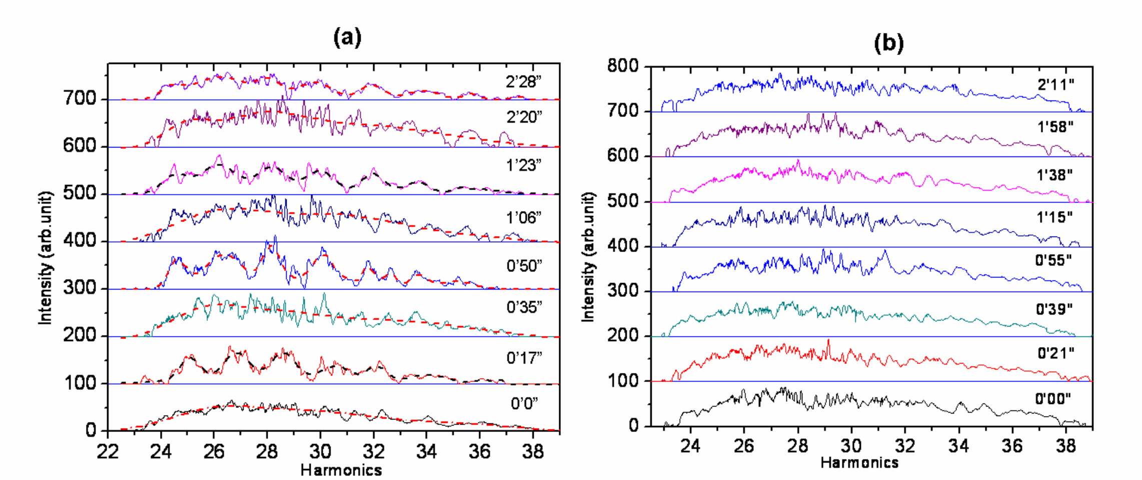


Fig.5 The single shot polarization gated high harmonic spectra measured at the time indicated by each lineout. The carrier envelope phase is not locked in (a) and is locked in (b).

The effects of the CE phase of the laser field on the XUV spectra can be seen from the single shot spectra at the maximum phase matching pressure (55 Torr) shown in Fig.4, which were taken over a 2 minute period. When the CE phase is not locked (Fig.5 (a)), the spectrum's profile changes significantly from shot to shot. Some spectra exhibit discrete harmonics orders corresponding to two electron re-collisions while others exhibit the super-continuum as a result of a single re-collision. As a contrary, when the CE phase is locked (Fig.5 (b)), the spectrum does not change much from shot-to-shot. The fine structures on the XUV spectra are likely from the statistical noise of the photons and the noise of the MCP gain. To the best of our knowledge, this is the first time that the effects of CE phase on the single shot, polarization gated harmonic spectra were observed.

### 2.4 Setup 2: To study effect of CE phase on multi-shot polarization gated XUV super-continuum

In order to observe effect of carrier shift with respect to envelope, there is no alternative method than to record pulse to pulse evolution of XUV spectrum generated by single or double electron ion re-collision unless CE phase of the driving laser field can be locked for certain period of time. If the spectrum were produced by acquiring data over many pulses, it would not provide any information than average effect of interaction with matter due to randomly varying CE phase with respect to envelope (Fig.7). Problem of large statistical noise in the single shot experiment and averaging effect in many cycle acquisitions can both be eliminated if CE phase of the driving laser can be locked for sufficiently long interval of time such that it allows to control over CE phase of the driving field by some mean such as by introducing dispersive material on the beam path or by changing the grating separation of the compressor[9-12] To make the scheme practicable it therefore requires co-ordination between two systems with one system stabilizing CE phase the other generates and records the high order harmonics. Schematic of the experimental set up to study attosecond two slits controlled by the carrier envelope is shown in the Fig.5. The CE phase stabilized intense output pulses of the KLS

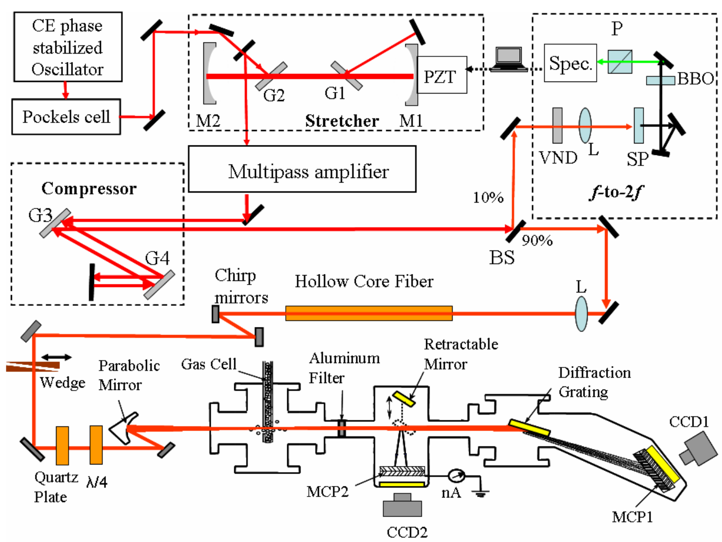


Fig.6 Schematics of CE phase stabilizing system of KLS and HHG spectrometer to study effect of CE phase on polarization gated XUV super-continuum.

amplifier with 1kHz repetition rate have an energy of 2.5 mJ and the duration 30 fs which is too long to assume few cycle pulses (sub-ten femtosecond pulses). So, about 1.2 mJ of pulses were passed through hollow-fiber and then compressed by a chirp mirrors which delivers 0.6 mJ with 7 fs duration. A couple of wedge plates mounted on a motorized stage after the hollow-fiber allow us to change the CE phase of the stabilized (Wedge in Fig.6) pulses. Introducing 58 μm of the thickness on the beam path gives rise to the change in CE phase of $2\pi$ radian, which corresponds to the 1.15 mm horizontal shift in the motorized stage. Quartz plate and the quarter wave plate generate the time dependent ellipticity as stated earlier. By optimizing pressure to maximum phase matching, XUV spectrum was generated by a compressed short pulse from hollow fiber with CE phase of the amplifier and oscillator locked.[9, 11] CE phase of the driving pulse on the gas target was gradually changed by varying the thickness of the fused silica wedge pair on the laser beam path. The measured dependence of the polarization-gated spectra on the CE phase is shown in Fig 7. The CE phase was scanned over $5\pi$, while one each phase value the spectrum were integrated over 50 laser shots.

From the width of the polarization gate it was estimated that there are only two attosecond pulses emitted in each laser shot. The modulation of the spectra is the result of the interference between the two pulses. The change of the CE phase causes a shift of the spectra. Spectra repeat for every π CE phase shift, as expected. Effect of CE phase on the XUV spectrum was first seen immediately after the polarization gate width was made less than one optical cycle Fig.7 (a). Successive continuum and discrete harmonics spectra with periodicity of π/2 were also observed with the change in CE phase of the laser pulse as the width of the linear portion of time dependent ellipticity approaches the value less than half the optical cycle (Fig7(c)) similar to the result reported in [13], which is consistent with the theoretical predictions [14, 15].

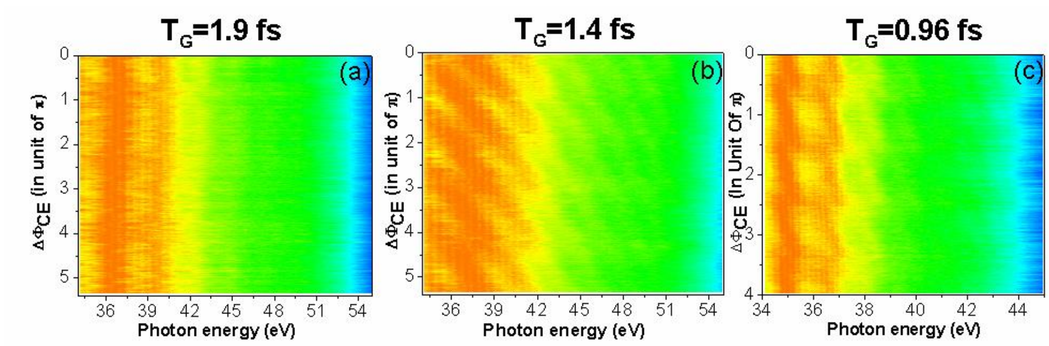


Fig.7 Effect of Carrier envelope phase on XUV spectrum at different polarization gate width. CE phase of the amplifier and the oscillator were locked. CE phase of the driving field is changed by changing thickness of the fused silica plates on the beam path

Further evidence for its sensitivity to the CE phase of the driving field has been depicted in Fig.8 (b). Shift in HHG spectrum was observed until CE phase of the amplifier and oscillator were locked and CE phase of the driving laser was changed by introducing fused silica plates on the beam path. The shift no longer observed as CE phase of the amplifier and oscillator were unlocked. As random shift in the CE phase of the driving laser causes HHG spectrum to shift in the random direction giving rise to the averaging effect. This effect is pictorially observed as a washed out continuum in the intensity map of XUV spectrum.

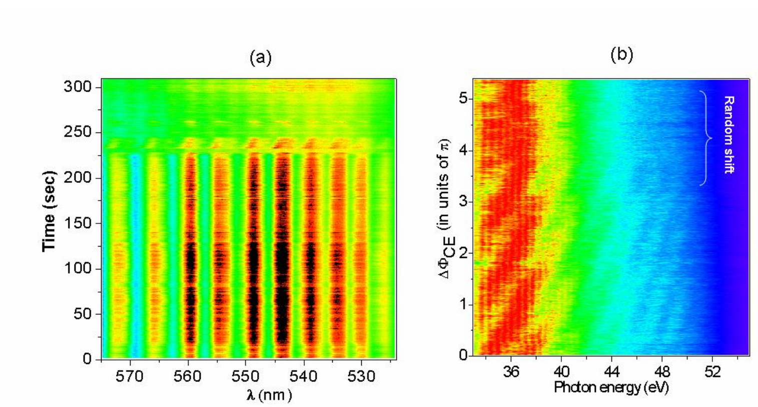


Fig.8 (a) f-to-2f interferrogram .Vertical strips represent the CE phase locked amplifier and the oscillator and washed portion indicates CE phase were unlocked (b) XUV spectrum when CE phase was locked and unlocked.

As for the further test to confirm the attosecond burst in the polarization gating, we scan CE phase over 5π and integrated the spectrum over 50 laser shots for each value of CE phase [10, 12] by changing the effective distance of the grating in the stretcher. The advantage of using this method over the method of introducing the fused silica on the beam path is that it never introduces material dispersion to the few cycle pulse as it does when wedge plates are used. Besides, it also eliminates problem of beam pointing instability at the interaction region caused by the addition of material thickness on the beam path. This is very important because pointing instability can fluctuate intensity in the interaction region which in turn can add similar kind of effect like change in CE phase does in XUV spectrum. Fig.9 (a) shows the interferrogram taken from *f*-to-*2f* interferometer when the effective grating separation was varied to change CE phase 0-π-2π and vice-versa. Corresponding XUV traces due to those changes are shown in Fig. 9 (b) Continuum spectrum are separated by π radian and the direction that high harmonic spectrum shifted were in consistent with that observed when wedge plates were used. Both the results of the measurement give better agreement in the periodicity with which, continuum appeared in the plot when the quartz plate of thickness 0.5 mm was used which corresponds to the gate width 0.96 fs.

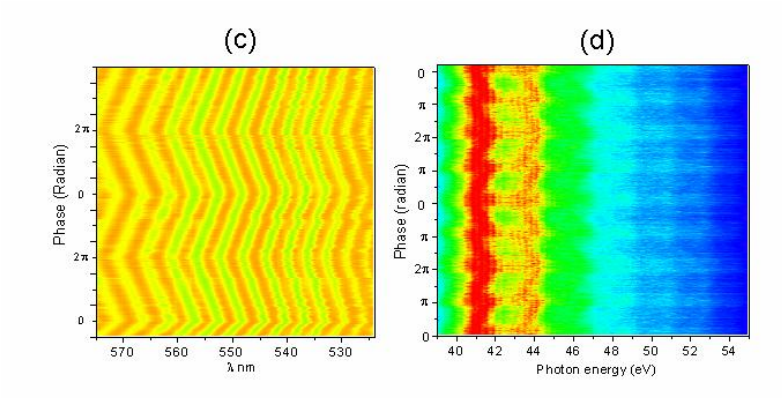


Fig.9 *f*-to-*2f* interferrogram on the left and corresponding effect on XUV spectrum on the right when effective grating separation was varied to change CE phase by 0-π-2 π and vice-versa.

Other sets of our investigation shows that sensitivity to the change in CE phases also depend on the relative position of the target from the focus. Our measured data shows, effect of CE phase appeared clearer as the target was moved closer to the focus which is shown in the Fig.10. Harmonic spectrums with alternative continuum stripes were observed when the target approach 1.8 mm away from the focus. We believe that the effect observed after the focus is due to the phase matching of the harmonics by the cancellation of the phase mismatch due to plasma dispersion and the Guoy phase shift by the variation of the harmonics phase due to a decrease of the laser intensity and the electron density in the laser propagation direction. Therefore, by the optimization of the four different parameters (gas pressure, polarization gate

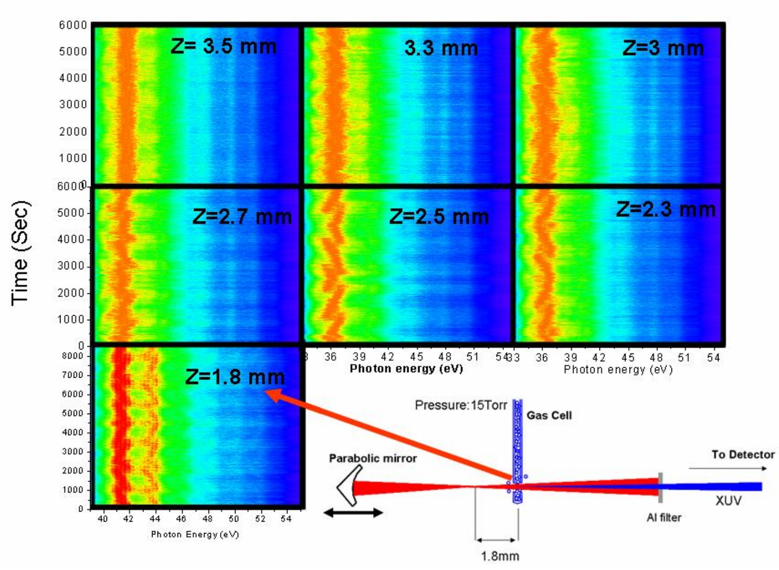


Fig. 10 Effect of relative position of the target from the focus in the measurement of CE phase effect on polarization gated XUV super-continuum.

width, rotation of the quartz plate and the focal position) sensitivity to the CE phase influence on XUV can be enhanced which is important in order to design the XUV phase meter. One neat example which might lead to a powerful phase meter has been depicted in Fig.11. In this case we varied CE phase by the known values by changing the effective distance between the grating pair of the stretcher and obtained the XUV traces under the best optimization of the

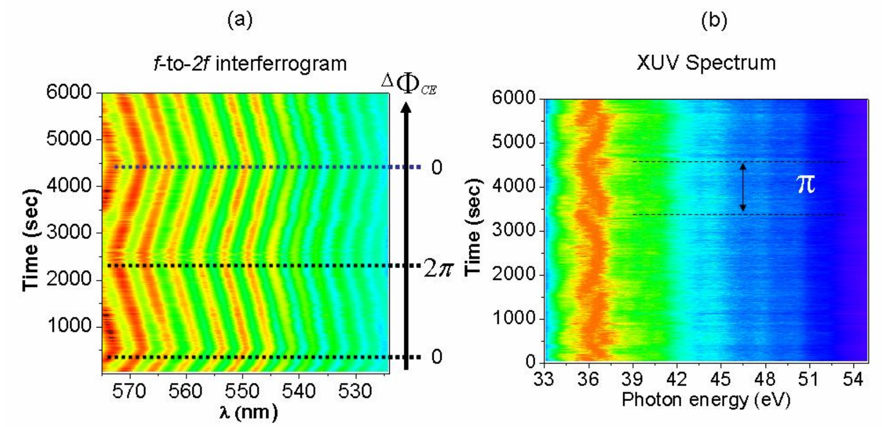


Fig.11 Measured CE phase effect on XUV by changing relative CE phase by known amount.

critical parameters. XUV spectrum appeared with the continuum with the separation of π radian. As continuum is the result of the laser field with CE phase π/2, this method therefore can be used to measure CE phase of the laser system.

# 3. CONCLUSION

The dependence of the CE phase on polarization-gated XUV spectra was investigated. It was found that the effect can only be seen when the width of the polarization gating is less than one optical cycle. We believe that the shift in the position of the spectral peak was caused by the change of the path of the electron returning as a result of the change in the CE phase. By generating ~$10^4$ photons per laser shot under the best phase-matching conditions, we were able to polarization-gate the single-shot XUV spectrum. This study can lead to a "phase meter" to measure the shot-to-shot change in CE phase of the laser pulse which would greatly benefit other CE phase dependent laser experiments using lasers whose CE phase is not locked. This work is funded by the United States Department of Energy and the National Science Foundation.

# REFERENCES


[1] **Brabec, T. and Krausz, F.**, Intense few-cycle laser fields: Frontiers of nonlinear optics. *Rev. Mod. Phys.* 2000. **72:** 591.

[2] **Krausz, F.**, *Physics World* Septemper 2001. p.41.

[3] **Salieres, P., LHuillier, A., Antoine, P. and Lewenstein, M.**, *Adv. at. Mol. Opt. Phys.* 1999. **41:** 83.

[4] **Hentschel, M., Kienberger, R., Spielmann, C., Reider, G. A., Milosevic, N., Brabec, T., Corkum, P., Heinzmann, U., Drescher, M. and Krausz, F.**, Attosecond metrology. 2001. **414:** 513.

[5] **Drescher, M., Hentschel, M., Kienberger, R., Uiberacker, M., Yakovlev, V., Scrinzi, A., Westerwalbesloh, T., Kleineberg, U., Heinzmann, U. and Krausz, F.**, Time-resolved atomic inner-shell spectroscopy. *Nature* 2002. **419:** 803-807.

[6] **Shan, B., Ghimire, S. and Chang, Z.**, Generation of the attosecond extreme ultraviolet supercontinuum by a polarization. *Journal of Modern Optics* 2005. **52:** 277-283.

[7] **Corkum, P. B., Burnett, N. H. and Ivanov, M. Y.**, Subfemtosecond pulses. *Opt. Lett.* 1994. **19:** 1870-1872.

[8] **Paulus, G. G., Lindner, F., Walther, H., Baltuska, A., Goulielmakis, E., Lezius, M. and Krausz, F.**, Measurement of the phase of few-cycle laser pulses. *Phys. Rev. Lett.* 2003. **91:** 253004.

[9] **Chengquan Li and Eric Moon, Hiroki Mashiko, Christopher M. Nakamura, Predrag Ranitovic, Chakra M. Maharjan, C. Lewis Cocke, Zenghu Chang, Gerhard G. Paulus**, Precision control of carrier-envelope phase in grating based chirped pulse amplifiers. 2006. **14:** 11468-11476.

[10] **Chengquan Li, Eric Moon and Zenghu Chang**, Carrier-envelope phase shift caused by variation of grating separation. 2006. **31:** 3113-3115.
[11] **Moon, E., Li, C. Q., Duan, Z. L., Tackett, J., Corwin, K. L., Washburn, B. R. and Chang, Z. H.**, Reduction of fast carrier-envelope phase jitter in femtosecond laser amplifiers. *Optics Express* 2006. **14:** 9758-9763.
[12] **Chang, Z.**, Carrier-envelope phase shift caused by grating-based stretchers and compressors. *Appl. Opt.* 2006. **45:** 8350-8353.
[13] **Sola, I. J., Mevel, E., Elouga, L., Constant, E., Strelkov, V., Poletto, L., Villoresi, P., Benedetti, E., Caumes, J. P., Stagira, S., et al**, Controlling attosecond electron dynamics by phase-stabilized polarization gating. *Nature Physics* 2006. **2:** 319-322.
[14] **Chang, Z.**, Single attosecond pulse and xuv supercontinuum in the high-order harmonic plateau. *Physical Review a* 2004. **70:** 043802.
[15] **Chang, Z.**, Chirp of the single attosecond pulse generated by a polarization gating. *Physical Review a* 2005. **71:** 023813.