Active beamline stabilization for Attosecond Pump-Probe experiment

Attosecond Science Group

Srijoyee Datta, University of Eastern Finland, Finland

Supervisor: Prof. Francesca Calegari

September 5, 2019

Abstract

The interaction of light with molecules initiate electronic processes that affect the chemical reactivity as well as the biological functionalities of the molecule. The typical time scale of this process is ranging between few femtoseconds (1 fs = 10^{-15} s) down to the attosecond (1 as = 10^{-18} s) regime. Therefore, the investigation of this dynamics demands the use of time-resolved spectroscopy with attosecond temporal resolution. The discovery of attosecond pulses thus unfold the reason behind many complex phenomenon in nature. Time resolved attosecond experiments are implemented generally by combining extreme ultraviolet (XUV) attosecond pulses with few-fs near infrared (NIR) pulses in a configuration called pump-probe that helps to reveal the presence of charge density variations in molecule [7]. This approach has helped in tracking the charge migration process in real time. To perform this experiment, an attosecond beamline combining different pulses (sub-fs UV pulses with either attosecond XUV or few-fs NIR pulses) needs to achieve a good stability over time for the results to be more accurate. An interferometer including most of the beamline optics called "short interferometer" has been designed to actively stabilize the attosecond beamline. Another interferometer termed as "long interferometer" has been built up including all the optics of the beamline. A continious wave (CW) laser (He-Ne laser) of wavelength of 632.991 nm has been implemented for stabilization scheme. The goal was to observe the stability of the long interferometer while actively stabilizing the short interferometer. Initially, the drift of this short interferometer was 294 attosecond (as) and after stabilization, the drift has been minimized to 44 as, while in case of long interferometer drift has been minimized to 90 attosecond that potrays good stability of beamline.

Contents

1 Introduction

Ultrafast Science has been a great matter of interest to the scientist for last few decades. One of the branches of ultrafast science, i.e Attosecond science, allow to study the processes that occur on a time scale of a few attoseconds. The prime tool to study the attosecond science is the optical pulses from ultrafast laser in attosecond time scale. The photoionization process in molecules activate few phenomenon such as chemical bond formation or brekage that eventually leads towards the change of structure of a molecule [8]. The light interaction with molecule leads to an internal molecular rearrangement, happens in an ultrafast time scale. The femtosecond (10^{-15} s) laser pulses are capable to study the nuclei motion. Electronic correlation plays a keyrole in this molecular rearrangement mechanisms. Infact, the electron dynamics is most fundamental to initiate changes in chemical composition and functionality of biological molecules [7].The pulse duration of laser pulses to study the electronic motion must be shorter than the time scale in which the electronic motion is happening. Thus, the study of electronic motion demands much shorter time scale than femtosecond i.e. attosecond.

Remarkable developement of laser technology finally has provided to access the sub-femtosecond (sub-fs) time scale. The technology of pulse shaping has given birth of the train of attosecond pulses. The first train of attosecond pulses have been observed in 2001 [5]. This experimental achievement finally allowed to investigate the electron dynamics at molecular level.

The experiment with attosecond pulses allow to perform an "ultrahigh-speed photography" during the molecular rearrangement. Once the molecule has been triggered with a pulse, the process of rearrangement starts and this attosecond pulse helps to probe it, i.e to record the snapshots of molecule in motion. A timed series of different delay of probe pulses help to capture a full movie of the process. The working of such process demands molecular coherence and the motion in all molecules must be synchronized. To make this happen, an ultrashort laser pulse must work as a pump pulse to initiate the molecular process. The above mentioned approach is known as pump-probe configuration. The spectroscopy with pump-probe approach has been first proposed by Ahmed Zewail in 1999 in the field of femtochemistry [1]. Typically, the XUV pump radiation triggers the ultrafast electron dynamics by ionizing the molecule, then it is probed by absorbing NIR pulses of different time delay and inducing photo-fragmentation. This time resolved photo-fragmentation pattern and the localization of the charge in molecules are very sensitive to each other and therefore it helps to reveal the presence of charge density variations.
An interferometric approach has been built up in order to obtain the beamline for pump-probe experiment in which the compressed sub-fs laser pulse has been broken into pump and probe pulse through a beam splitter (70 percent reflected and 30 percent transmitted) where the weaker beam (probe) is used to monitor the stronger beam (pump) persuaded changes of electronic configuration of the molecules [2]. The stability of attosecond beamline is the most fundamental part to start with the pump-probe

experiment. If the beamline is not stable, the molecular movie won't be captured in a particular delay time. A time average around a particular time delay will degrade the quality of the snapshots of the molecular process. In result, a blurred snapshot of a particular time delay will be captured.

A 100 nm change in path length between pump and probe arms of the interferometer results in a delay offset by 330 as [4]. Therefore, in order to perform an efficient pump-probe experiment with attosecond temporal resolution,an active stabilization system has been built up with the help of a piezoelectric stage to compensate the variation of optical path length that might occur due to intrinsic thermal and mechanical drift. This piezoelectric stage faciliated the stability of optical path length in nanometer precision. A continious wave (CW) laser (He-Ne laser) of wavelength of 632.991 nm has been used along the optical paths of the interferometer in order to attain the attosecond beamline stabilization.

In this report, a brief discussion on pump-probe spectroscopy and how pump-probe experiment happen in molecules, and also active stabilization of the interferometer will be discussed in Theory portion. Measurement part will depict the set-up of the interferometers and how these has been used to stabilize the fringes. Lastly, the Conclusion part will depict the measured drift by two interferometers.

2 Theory

Pump-probe spectroscopy

In pump-probe spectroscopy, a higher energy pulse (short pulse duration) considered as pump pulse drives the sample into excited state. This excitation changes the optical properties (transmission, reflection or absorption) of the sample. The optical properties evolve with time as the sample relaxes to the ground state. A lower energy pulse (long pulse duration), termed as probe pulse with a given delay measures the optical properties. The variation of delay time between pump and probe pulses allow to measure the evolution of optical properties with time throughout the relaxation.

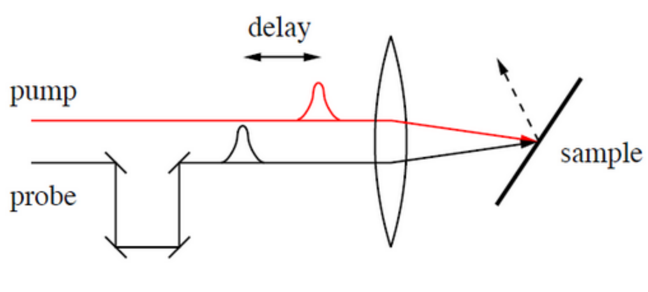

Figure 1: A schematic of pump-probe experiment

In case of molecules, as pump pulse hit the target under study it helps to create high concentration of transient species. These species permit their direct observation. The pulse duration should be significantly small than the lifetime of transient species. The system is then allowed to evolve for a certain time period during which the transient species decays. If the transient is an excited electronic state, it may decay to the ground state or it may transform into further transient species or reaction products. After this time of evolution, the system is probed with probe pulse that allows the information about the identity and concentration of the transient species under observation. A systemically variation of the time period between pump and probe pulse permits the study of kinetic behavior of the transient species [2].

Active stabilization of interferometer
Interferometry has been recognized as a very powerful tool to measure ultimately sensitive detection of the optical signals. In order to achieve very slow change in phase difference over extremely short timescale, active stabilization of interferometer has been achieved. Active stabilization system consist of a delay stage that help to stabilize the interference fringes. The piezo-electric delay stage with nanometer precision, consist of a feedback loop that helps to track the delay stage everytime it moves to a new position and stable them at that position. Active stabilization also helps to overcome the intrinsic thermal and mechanical drift due to various optical components of beamlime in the time of experiment.

3 Measurement

3.1 Experimental set-up

This section depicts the design of an attosecond beamline that has been used to perform the pump-probe experiment in molecules. An interferometric approach has been attained to realize the attosecond beamline which is schematically represented in Figure 2.

The compressed sub-fs incoming beam achieved through a hollow core fiber is split into reflected and transmitted part by a 1 mm thick beam splitter (BS) placed in chamber 1. The reflected beam (70 percent of incoming intensity)is used to initiate the Higher harmonic generation (HHG) process (in chamber 2 as shown in Figure 2) for generation of attosecond pulse, used as pump pulse for the time resolved experiment. After HHG, the generated attosecond pulse travels through a metallic filter that helps to remove the residual radiation from main laser. The higher the order of the harmonics, the rate of generation of attosecond pulses become lower and more stronger filter is needed to eliminate the residual radiation from attosecond pulse. After that, it travels to the drilled mirror (DM).
The transmitted beam (30 percent of the incoming intensity), passed through a piezo-electric stage, can act as probe pulse or it can be exploited to drive the Third harmonic

generation (THG) process for generation of UV pulses. The piezoelectric stage provides 1 nm spatial resolution (7 as in time domain), used to vary the relative delay between the two pulses during the time of pump-probe experiment, mentioned in Figure 2. After that, the beam then pass through spectral separators (SS), that helps to send a portion of beam to chamber 4 (experiment chamber). During the experiment spectral separators (SS) in chamber 5 are used to eliminate the residual IR radiation (main incoming beam) from the generatd UV light and send the UV beam to the chamber 4.

The UV beam would pass through another couple of spectral separators (SS), and then directly propagates to the chamber 4, where it joins with the beam coming through the hole of drilled mirror (DM). Now to stabilize this beamline, i.e to check with the drift of phases of interference fringes, the recording of the interfernce fringes could be done with the help of last mirror in chamber 4, at the outside of the chamber by camera 2 (C2). The problem with this scheme is, this last mirror could not be at that position during experiment, otherwise the main beam will be blocked to reach the experiment chamber that leads to the failure of the performance of experiment.
To avoid this problem and to stabilize the beamline too, an interferometer called Short interferometer have been realized with the help of this recombining beam splitter (BS) of chamber 3. An auxiliary continious wave (CW) laser, propagating collinearly with the main laser along the beampath, has been used to obtain the fringes. The low output power of this CW laser helps it to co-propagate with main laser beam without affecting it properties. During experiment, this CW laser is unable to propagate through the spectral separators (SS) to the chamber 4 since SS only allow to send the UV beam. The transmitted part of CW laser from SS in chamber 5 reflect from the mirror placed at outside of chamber 5 and propagates to the recombining BS in chamber 3. A portion of the beam from the outer part of the hole drilled mirror have been reflected by a mirror in and propagate to the recombining beam splitter (BS) in chamber. These two beams helped to make interference fringes and at this point fringes has been recorded, outside the chamber by camera 1 (C1) and the drift of phases has been analyzed from the captured fringes. This "short interferometer" covers almost 70 percent path of the beamline, therefore in principle, it could be possible to stabilize the attosecond beamline with stabilizing this short interferometer. In this experiment, a He-Ne laser of wavelgth of 632.991 nm has been used as CW laser. After the collimation of the output of the He-Ne laser with a telescopic lens set up, the beam passed through several optical components before travelling to the beam splitter (BS). The telescopic lens set up have been realized with a concave (helps to diverge the beam) and plano-convex lens (helps to collimate the beam). This beam collimation actually helps to focus the beam at infinity. Therefore as the distance between beam and source increases, the wavefront doesn't diverge anymore, it becomes flat. This plane wavefronts helps to achive an interference fringes of good contrast.

To check how well the short interferometer is working to stabilize the attosecond beamline, from the spectral separator part, a portion of this CW laser propagate to the chamber 4 and joins with the beam coming through the hole of DM. From experiment chamber, with the help of placing the last mirror, it is possible to record the interference fringes at the outside of the chamber 4 by camera 2 (C2). This second set-up has been named as "Long interferometer". This interferometer includes the optical components of the the experiment chamber too. In figure 2, the thicker red line represents the path followed by the short one, while the comparative thinner red line represent the path followed by long interferometer. The goal was to stabilize this long interferometer with the help of the short interferometer, since it covers most of the paths of the long one. This approach helped to stabilize the attosecond beamline in an efficient manner.

A scheme of this Short and Long interferometer has been shown seperately in Figure 3 and Figure 4.

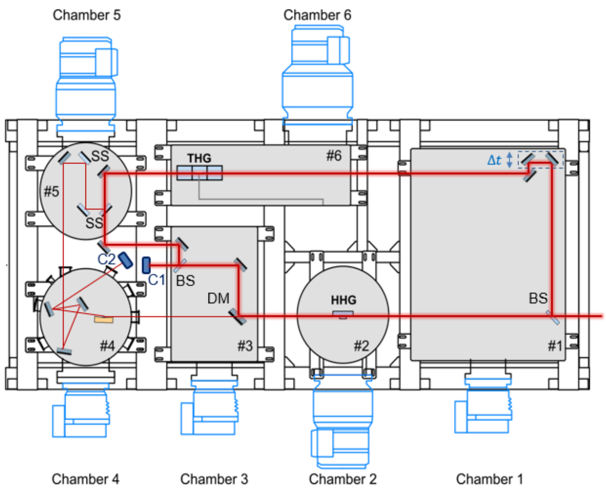

Figure 2: A schematic of short interferometer. Red line represents the beampath. BS:Beam splitter; HHG:Higher harmonic generation; THG:Third harmonic generation; SS: Spectral separator; C2: Camera 2; C1: Camera 1; DM: Drilled mirror

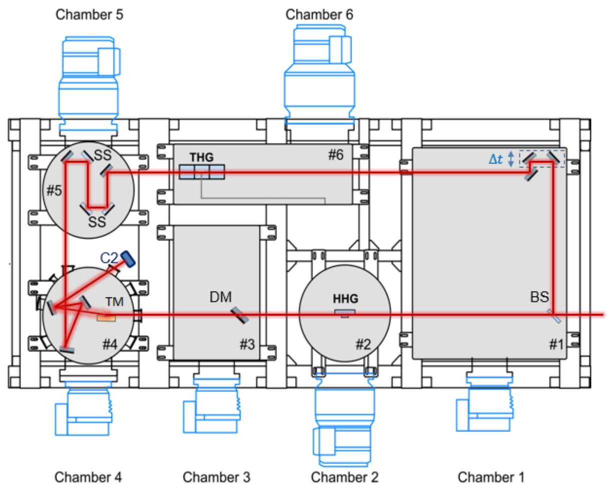

Figure 3: Schematic of the long interferometer.Red line shows the beam path. BS: Beam splitter; HHG: Higher harmonic generation; DM: Drilled mirror; SS: Spectral separator; THG: Third harmonic generation; TM: Toroidal mirror; C2: Camera 2.

3.2 Experiment

A scan has been done with different configurations for 30 minutes in each interferometer to verify the nature and amount of drift induced by two interferometers.

a) **Piezo stage off**
This has been done to get aware of the intrinsic drift by the interferometers due local variations of temperature in optical components without any difference between the path length. This ensured the amount of turbulent in the local environment of the interferometer.

b) **Piezo stage on**
To check with the amount of noise further caused by the piezoelectric stage, how accurate it is working and how much it is affecting the fringe stability, two more procedures have been followed under this configuration.
i) Piezo stage on but with no feedback (Open loop configuration).
ii) Piezo stage on and with the feedback loop (Closed loop configuration).

Finally, the fringes from both interferometers have been tried to stabilize with a stabilization system.

c)**Piezo stage on with stabilization on**
The stabilization software has been previously built up in LabVIEW, that helps to stabilize the fringes with respect to a reference one by controlling the proportional error.

In each cases, several snapshots of fringes has been taken and stored through a program previously written in LabVIEW. These images have been used for the analysis purpose.

To ensure if the short interferometer has been working well or not, the nature of drift has been first observed in both interferometers. The same trend of drift ensures that it could be possible to stabilize the beamline using this short interferometer. After that the amount of drift of this short interferometer has been tried to minimize in order to achieve better stability of beam line. Once the short interferometer shows lower drift, it also ensure smaller drift for the long one, that means for the whole attosecond beamline. The picture of fringes captured by the Long and short interferometer has been displayed in Figure 5 and Figure 6.

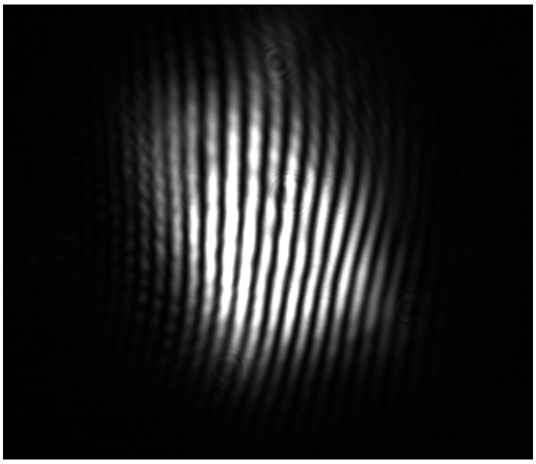

Figure 4: Fringes captured by the long interferometer

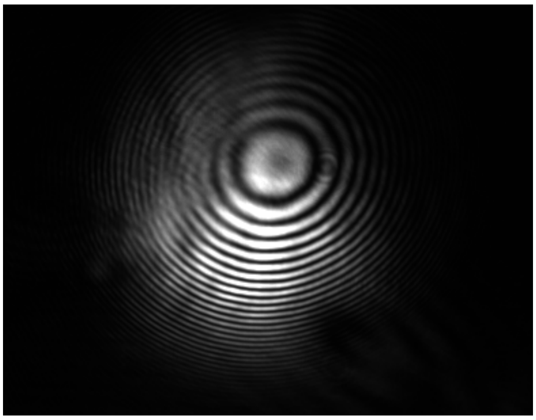

Figure 5: Fringes captured by the short interferometer

3.3 Analysis

In Figure 4 and Figure 5, any changes of the position between the bright and dark fringes will induce phase drift. The drift of phases over time for the long and short interferometer has been tried to compare and minimize. A Matlab code has been written to analyze the drift over time. A region of high contrast has been chosen from the images as shown in Figure 6.

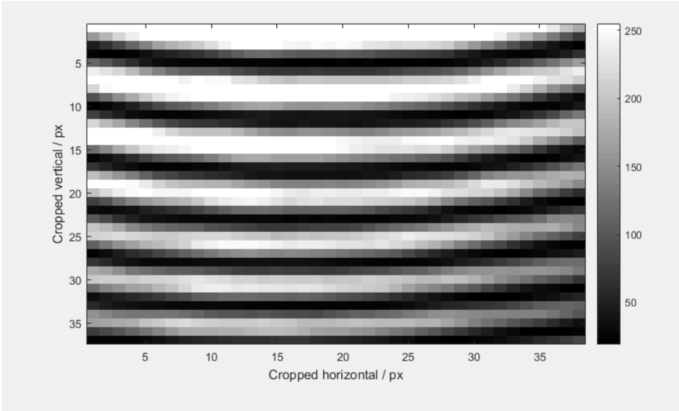

Figure 6: Cropped portion of the high fringe contrast region

An intensity plot has been derived by summing up this cropped image in vertical direction. The intensity plot has been shown in Figure 7.

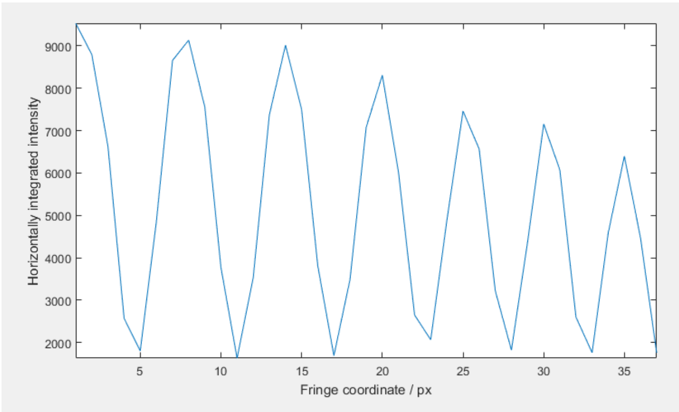

Figure 7: Intensity plot of the cropped region

After that, Fourier transform has been performed to find the frequency component from the intensity plot as shown in Figure 8. From figure, it is shown that fourier transform provides two peaks of spatial frequency and by selecting a single spatial frequency, standard deviation of phases have been tracked for that particular spatial frequency, and this standard deviation for last 100 images have been plotted. After that, this standard deviation (drift of phases) of phases have been realized in attosecond time scale by finding the delay time between two oscillations. The evaluation of drift in time have been shown in Figure 9 and in further figures in Results and Discussion section. Unwrapping of phase has been performed to overcome the limitation in phase plotting, (i.e. −pi to +pi) by Matlab and thus the accurate trends of the drift of phases has been observed over time.

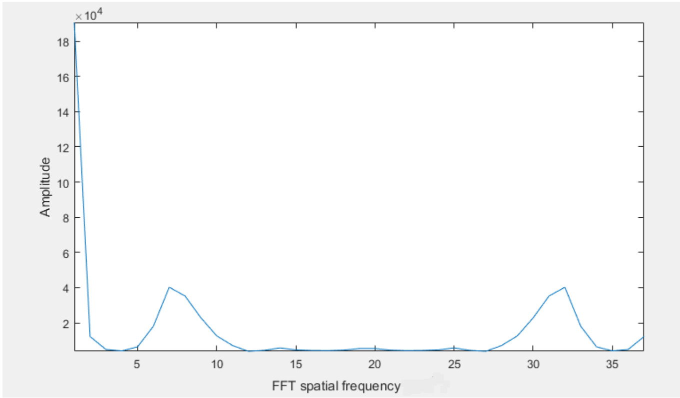

Figure 8: Fourier transform of the Intensity plot

3.4 Results and Discussion

The results has been obtained for different configurations mentioned in the Experiment paragraph.

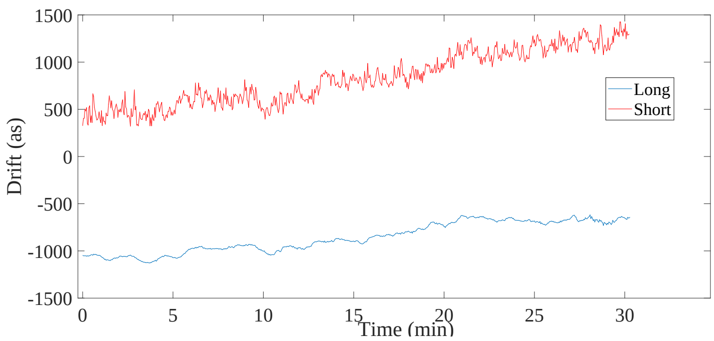

Figure 9: Drift in attosecond with Piezostage off configuration for last 100 images. Red curves represent nature of drift in short interferometer. Blue curve depicts for long (main) interferometer.

Figure 9 shows the nature of drift is same for both interferometers over time when the piezo stage was off. A drift of 174 as for long one and 294 as for the small one has been observed. It could be inferred that the intrinsic stability of the interferometers behave in same way for two interferometers and thus stabilizing the short interferometer will allow to stabilize the main interferometer. The figure also depicts larger drift for small interferometer than the main one. This problem has been reduced by replacing the mirror mount outside the spectral separator chamber.

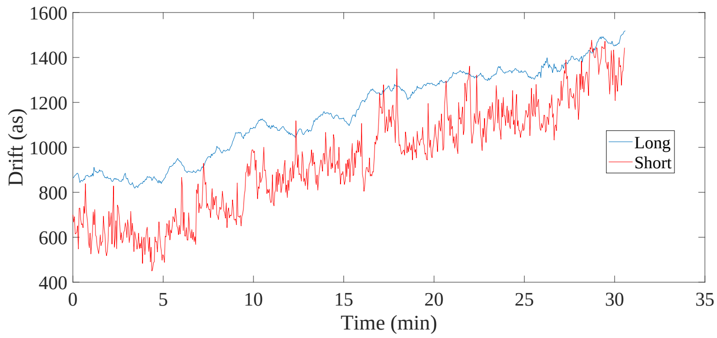

Figure 10: Drift in attosecond with Open loop configuration of Piezostage for last 100 images. Red curves represent nature of drift in short interferometer. Blue curve depicts for long (main) interferometer.

Figure 10 shows the trend of drift in fringes over time when the piezo stage was on in open loop configuration.In open loop configuration, the standard deviation in phases (drift) of 171 as has been perceived for long interferometer and the short one indicates a drift of 322 as. The replacement of the mirror mount helped the long interferometer to be a bit more stable but it left the short one more unstable. Therefore the mirror mount on a new breadboard have been installed to get better stability. The same nature of both the plot again ensures that short interferometer could be an efficient tool to control the delay in the pump-probe experiment.

The same conclusion can be depicted from the Figure 11 in closed loop configuration. The feedback loop was required to control the relative delay between two arms.In this configuration, a drift of 173 as for the long interferometer and a drift of 176 as for the short interferometer has been achieved which seems quite better result than all other previous configuration. The intrinsic stability of the interferometers observed was quite good and the drift for the short interferometer has also been reduced in an efficient manner.

Figure 12 shows the drift in long interferometer when the piezo and stabilization both were on. A scan of 30 miniutes results in a drift of 90 as. The result of this scan ensures a good intrinsic stability that is actually helpful to perform the pump-probe experiment.

Figure 13 shows the deviation of phase in time for the short interferometer with piezo

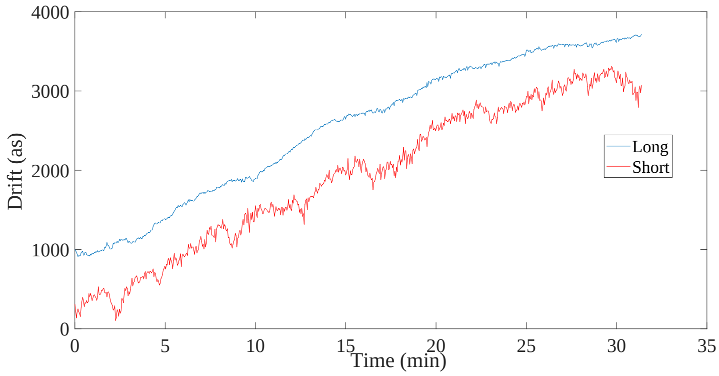

Figure 11: Drift in attosecond with Closed loop configuration of Piezostage for last 100 images. Red curves represent nature of drift in short interferometer. Blue curve depicts for long (main) interferometer.

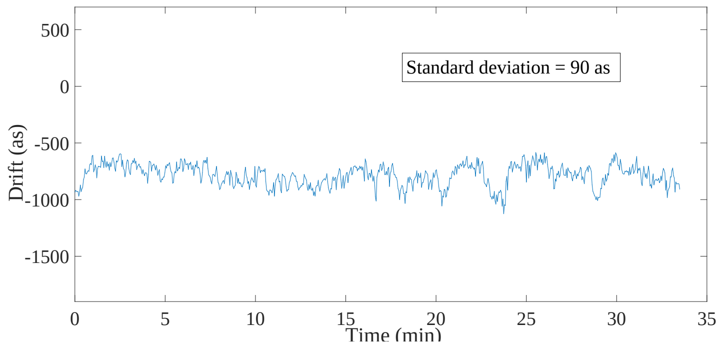

Figure 12: Drift in attosecond with Closed loop configuration along with stabilization for last 100 images of long interferometer.

and stabilization both on configuration. A drift of 44 as has been attained which potrays very good stability for the short interferometer that is going to stabilize the long one during experiment.

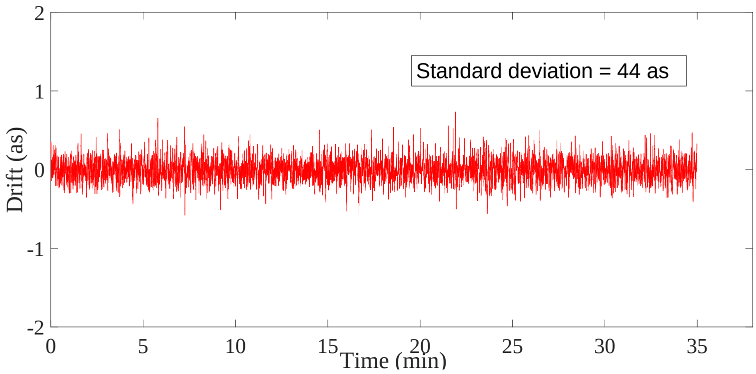

Figure 13: Deviation of Phase over time with Closed loop configuration along with stabilization for last 100 images of short interferometer.

4 Conclusion

The attosecond technology is most proficient tool to study the electronic processes. In DNA base adenine, attosecond technology has been exploited to measure the characteristic time of the shake up process, typically happens in ultrafast time scale. Electron dynamics could be observed in biologically relevent molecules by performing pump-probe experiments. The probe pulses in the attosecond beamline must achieve good stability in order to track the electron dynamics over time. This requires a very good stability of the beamline. In order to achieve the stabilization, interferometric approach was most simple to implement for stabilization. An interferofeter covering most of the optics of the beamline, called "short interferometer" has been designed to control the delay stability of the beamline. Delay has been provided by a piezoelectric stage. In the experiment, different configuration (Open loop and closed loop) of the piezo stage has been performed to make sure that the delay line was working fine. The active stabilization enables to stabilize any local drifts caused by the optical components and mechanical stages inside the vacuum chambers. The piezoelectric stages usually suffer from hysteresis problem and therefore a continuous wave laser must be used as a reference for the stabilization and controlling the delay [4]. The feedback loop (piezo in closed loop configuration) of piezo-stage helps to control the delay line. As soon as the delay was set to a new value, the feedback loop scanned the piezo-stage at new position as demarcated by the interference fringes and stabilize the interferometer at that position. In order to check how well the short interferometer is capable to control the stability of beamline, another interferometer termed as long interferometer has been designed including all the optics of the beamline. The goal was to observe the stability of the long interferometer while actively stabilizing the short interferometer. A drift with same nature for both interferometers has ensured this stategy to stabilize the beamline could be a noble method for attosecond beamline stabilization. A drift of 44 as for short interferometer and 90 as for the main interferometer has been obtained in the end. Initially the target was to perform the experiment with a laser of wavelength of 473 nm (Blue laser) since it can easily pass through the optical components that has been chosen for ultraviolet beam, specially the SS in chamber 5. The blue laser was not available in the beginning, therefore the experiment has been performed with a He-Ne laser, wavelength of 632.991 nm. The spectral separators has been chosen to separate the wavelength of blue from the ultraviolet beam during experimet. In reality, it also helped to transmit a fraction of He-Ne wavelength and the fringes has been observed on the cameras. The stability in attosecond beamline could be considered as one of the most fundamental steps in order to begin with a pump-probe experiment that could open up important perspectives in photo-therapy and photo-biology. The precise stability in time scale ensures a focused image of the electron dynamics of molecules captured by the camera of Velocity map imaging (VMI) spectrometer. Thus it could be inferred that the active stabilization of attosecond beamline plays a crucial role in determining the ultrafast electron dynamics of molecules in an efficient manner.

5 Acknowledgement

I would like to express my gratitude to my supervisor, Prof. Francesca Calegari who ushered me on this exciting path of Ultrafast science. I felt immensely lucky and happy to work with all the members of the Attosecond science group who guided and helped me throughout the project and always responded to my queries promptly. Special thanks to Erik, Vincent, Krishna, Gaia and Lorenzo for being so helpful and motivating.

I would also like to express my appreciation for the team behind the summer student program at DESY, which is an invaluable experience for any student. Thanks for filling my days with sunshines.

Lastly, I want to thank my parents and few close friends for their love and support throughout the time.

References

[1] Zewail, A. H. (2000).”Femtochemistry:Atomic-scale dynamics of the chemical bond.” *The Journal of Physical Chemistry A*, 104(24), pp. 5660-5694.

[2] Long, D. A., ”An Introduction to Laser Spectroscopy.” Edited by David L. Andrews and Andrey A. Demidov. Kluwer Academic/Plenum Publisher, New York, Second Edition, *Journal of Raman Spectroscopy*, 2004.

[3] Calegari, Francesca and Sansone, Giuseppe and Stagira, Salvatore and Vozzi, Caterina and Nisoli, Mauro, ”Advances in attosecond science”. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 10.1088/0953-4075/49/6/062001, 2016.

[4] Chini, Michael and Mashiko, Hiroki and Wang, He and Chen, Shouyuan and Yun, Chenxia and Scott, Shane and Gilbertson, Steve and Chang, Zenghu, ”Delay control in attosecond pump-probe experiments.” *Optics Express*,10.1364/oe.17.021459,2009.

[5] Krausz, Ferenc, ”The birth of attosecond physics and its coming of age.” *Physica Scripta*,10.1088/0031-8949/91/6/063011, 2016.

[6] Schlaepfer, F. and Volkov, M. and Hartmann, N. and Niedermayr, A. and Schumacher, Z. and Gallmann, L. and Keller, U., ”Phase stabilization of an attosecond beamline combining two IR colors.” *Optics Express*, 10.1364/oe.27.022385, 2019

[7] Maansson, E. P. and De Camillis, S. and Castrovilli, M. C. and Galli, M. and Nisoli, M. and Calegari, F. and Greenwood, J. B., ”Ultrafast dynamics in the DNA building blocks thymidine and thymine initiated by ionizing radiation.” *Physical Chemistry Chemical Physics*, 10.1039/c7cp02803b, 2017

[8] Remacle, F. and Levine, R. D., ”An electronic time scale in chemistry.” *Proceedings of the National Academy of Sciences of the United States of America*, 10.1073/pnas.0601855103, 2006