

Kaoru Yamanouchi  
Louis F. DiMauro  
Wendell T. Hill, III *Editors*

# Progress in Ultrafast Intense Laser Science XVII

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Kaoru Yamanouchi · Louis F. DiMauro ·  
Wendell T. Hill, III  
Editors

# Progress in Ultrafast Intense Laser Science XVII



*Editors*

Kaoru Yamanouchi  
Institute for Attosecond Laser Facility  
The University of Tokyo  
Tokyo, Japan

Louis F. DiMauro  
Department of Physics  
The Ohio State University  
Columbus, OH, USA

Wendell T. Hill, III  
Institute for Physical Science  
and Technology  
University of Maryland  
College Park, MD, USA

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# Preface

We are pleased to present the 17th volume of Progress in Ultrafast Intense Laser Science. As the frontiers of ultrafast intense laser science rapidly expand ever outward, there continues to be a growing demand for an introduction to this interdisciplinary research field that is at once widely accessible and capable of delivering cutting-edge developments. Our series aims to respond to this call by providing a compilation of concise review-style articles written by researchers at the forefront of this research field so that researchers with different backgrounds as well as graduate students can easily grasp the essential aspects.

As in the previous volumes, each chapter of this book begins with an introductory part, in which a clear and concise overview of the topic and its significance is given, and moves onto a description of the authors' most recent research results. All chapters are peer-reviewed. The articles of this 17th volume cover a diverse range of the interdisciplinary research field, and the topics may be grouped into three categories: applications of attosecond and femtosecond laser pulses (Chaps. 1–4), coherence and dynamics in quantum systems (Chaps. 5–7), and applications of super-intense laser fields (Chaps. 8 and 9).

From the third volume, the PUILS series has been edited in liaison with the activities of the Center for Ultrafast Intense Laser Science at the University of Tokyo, which has also been responsible for sponsoring the series and making the regular publication of its volumes possible. From the 5th to the 16th volumes, the Consortium on Education and Research on Advanced Laser Science, the University of Tokyo, has joined this publication activity as one of the sponsoring programs. From this volume, the Institute for Attosecond Laser Facility will succeed in sponsoring the publication of the series. The series, designed to stimulate interdisciplinary discussion at the forefront of ultrafast intense laser science, has also collaborated since its inception with the annual symposium series of ISUILS (<http://www.isuils.jp/>), sponsored by JILS (Japan Intense Light Field Science Society).

We would like to take this opportunity to thank all of the authors who have kindly contributed to the PUILS series by describing their most recent work at the frontiers of ultrafast intense laser science. We also thank the reviewers who have read the

submitted manuscripts carefully. One of the co-editors (KY) thanks Ms. Mihoshi Abe for her help with the editing processes.

We hope this volume will convey the excitement of ultrafast intense laser science to the readers and stimulate interdisciplinary interactions among researchers, thus paving the way to explorations of new frontiers.

Tokyo, Japan  
Columbus, USA  
College Park, USA

Kaoru Yamanouchi  
Louis F. DiMauro  
Wendell T. Hill, III

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# Contributors

**Ioannis Lontos** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece

**Jon I. Apiñaniz** Centro de Láseres Pulsados (CLPU), Villamayor, Salamanca, Spain

**E. Appi** Department of Physics, Lund University, Lund, Sweden

**D. Charalambidis** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece;  
Department of Physics, University of Crete, Heraklion (Crete), Greece;  
ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**T. Csizmadia** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**Z. Diveki** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**A. Emmanouilidou** Department of Physics and Astronomy, University College London, London, UK

**P. Eng-Johnsson** Department of Physics, Lund University, Lund, Sweden

**B. Farkas** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**Robert Fedosejevs** Electrical and Computer Engineering, University of Alberta, Edmonton, AB, Canada

**Ke Feng** State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics (SIOM), Chinese Academy of Sciences (CAS), Shanghai, People's Republic of China

**Calvin Z. He** Joint Quantum Institute, University of Maryland, College Park, MD, USA;  
Institute for Physical Science and Technology, University of Maryland, College Park, MD, USA

**Wendell T. Hill III** Joint Quantum Institute, University of Maryland, College Park, MD, USA;  
Institute for Physical Science and Technology, University of Maryland, College Park, MD, USA;  
Department of Physics, University of Maryland, College Park, MD, USA

**Kunio Ishida** School of Engineering and Center for Optical Research and Education, Utsunomiya University, Utsunomiya, Tochigi, Japan

**S. Kahaly** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary;  
Institute of Physics, University of Szeged, Szeged, Hungary

**A. L'Huillier** Department of Physics, Lund University, Lund, Sweden

**Th. Lamprou** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece;  
Department of Physics, University of Crete, Heraklion (Crete), Greece

**Roberto Lera** Centro de Láseres Pulsados (CLPU), Villamayor, Salamanca, Spain

**Ruxin Li** State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics (SIOM), Chinese Academy of Sciences (CAS), Shanghai, People's Republic of China;  
School of Physical Science and Technology, ShanghaiTech University, Shanghai, People's Republic of China

**Andrew Longman** Lawrence Livermore National Laboratory, Livermore, CA, USA

**Erik Lötstedt** Department of Chemistry, School of Science, The University of Tokyo, Bunkyo-ku, Tokyo, Japan

**S. Madas** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary;  
Institute of Physics, University of Szeged, Szeged, Hungary

**Rohan Mahnot** Joint Quantum Institute, University of Maryland, College Park, MD, USA;  
Institute for Physical Science and Technology, University of Maryland, College Park, MD, USA

**I. Makos** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece;  
Department of Physics, University of Crete, Heraklion (Crete), Greece;  
Physikalisches Institut, Albert-Ludwigs-Universität, Freiburg, Germany

**Vicent Mateu** Departamento de Física Fundamental and IUFFyM, Universidad de Salamanca, Salamanca, Spain

**Katsumi Midorikawa** Attosecond Science Research Team, RIKEN Center for Advanced Photonics, Wako, Saitama, Japan

**S. Mukhopadhyay** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**B. Nagyillés** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**Kazutaka G. Nakamura** Laboratory for Materials and Structures, Institute of Innovative Research, Tokyo Institute of Technology, Yokohama, Japan

**A. Nayak** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary;  
Institute of Physics, University of Szeged, Szeged, Hungary

**L. A. A. Nikolopoulos** School of Physical Sciences, Dublin City University, Dublin 9, Ireland

**Hideki Ohmura** National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki, Japan

**Tomoya Okino** Attosecond Science Research Team, RIKEN Center for Advanced Photonics, Wako, Saitama, Japan

**I. Orfanos** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece;  
Department of Physics, University of Crete, Heraklion (Crete), Greece

**José Antonio Pérez-Hernández** Centro de Láseres Pulsados (CLPU), Villamayor, Salamanca, Spain

**D. Rajak** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**Smrithan Ravichandran** Joint Quantum Institute, University of Maryland, College Park, MD, USA;  
Institute for Physical Science and Technology, University of Maryland, College Park, MD, USA

**Luis Roso** Departamento de Física Aplicada, Universidad de Salamanca, Salamanca, Spain

**G. Sansone** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary;  
Physikalisches Institut, Albert-Ludwigs-Universität, Freiburg, Germany

**Taro Sekikawa** Department of Applied Physics, Hokkaido University, Sapporo, Japan

**E. Skantzakis** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece

**Futa Sunaga** Graduate School of Regional Development and Creativity, Utsunomiya University, Utsunomiya, Tochigi, Japan

**Itsuki Takagi** Laboratory for Materials and Structures, Institute of Innovative Research, Tokyo Institute of Technology, Yokohama, Japan

**V. Tsafas** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece

**P. Tzallas** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece;  
ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**M. Upadhyay Kahaly** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary

**K. Varju** ELI-ALPS, ELI-Hu Non-Profit Ltd, Szeged, Hungary;  
Department of Optics and Quantum Electronics, University of Szeged, Szeged, Hungary

**E. Vassakis** Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, Heraklion (Crete), Greece

**Wentao Wang** State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics (SIOM), Chinese Academy of Sciences (CAS), Shanghai, People's Republic of China

**R. Weissenbilder** Department of Physics, Lund University, Lund, Sweden

**Kaoru Yamanouchi** Department of Chemistry, School of Science, The University of Tokyo, Bunkyo-ku, Tokyo, Japan;  
Institute for Attosecond Laser Facility, The University of Tokyo, Bunkyo-ku, Tokyo, Japan

# Chapter 1

## Non-linear Extreme Ultraviolet Applications with Attosecond Pulses



**E. Skantzakis, I. Orfanos, A. Nayak, I. Makos, Ioannis Lontos, E. Vassakis, Th. Lamprou, V. Tsafas, T. Csizmadia, Z. Diveki, B. Nagyillés, B. Farkas, S. Mukhopadhyay, D. Rajak, S. Madas, M. Upadhyay Kahaly, S. Kahaly, R. Weissenbilder, P. Eng-Johnsson, E. Appi, A. L'Huillier, G. Sansone, K. Varju, L. A. A. Nikolopoulos, A. Emmanouilidou, P. Tzallas, and D. Charalambidis**

**Abstract** In recent years laser driven attosecond sources based on loose geometry high order harmonic generation reach focused intensities as high as to induce multi-photon multiple ionization or even strong -field effects in the extreme ultraviolet

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The original version of the chapter has been revised: Author names have been updated. A correction to this chapter can be found at  
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E. Skantzakis · I. Orfanos · I. Makos · I. Lontos · E. Vassakis · Th. Lamprou · V. Tsafas · P. Tzallas · D. Charalambidis (✉)  
Foundation for Research and Technology—Hellas, Institute of Electronic Structure & Laser, PO Box 1527, GR71110 Heraklion (Crete), Greece  
e-mail: [chara@iesl.forth.gr](mailto:chara@iesl.forth.gr)

I. Lontos  
e-mail: [ilontos@iesl.forth.gr](mailto:ilontos@iesl.forth.gr)

I. Orfanos · I. Makos · Th. Lamprou · D. Charalambidis  
Department of Physics, University of Crete, PO Box 2208, GR71003 Heraklion (Crete), Greece

A. Nayak · T. Csizmadia · Z. Diveki · B. Nagyillés · B. Farkas · S. Mukhopadhyay · D. Rajak · S. Madas · M. Upadhyay Kahaly · S. Kahaly · G. Sansone · K. Varju · P. Tzallas · D. Charalambidis  
ELI-ALPS, ELI-Hu Non-Profit Ltd, Dugonics tér 13, H-6720 Szeged, Hungary

A. Nayak · S. Madas · S. Kahaly  
Institute of Physics, University of Szeged, Dom tér 9, 6720 Szeged, Hungary

R. Weissenbilder · P. Eng-Johnsson · E. Appi · A. L'Huillier  
Department of Physics, Lund University, SE-221 00 Lund, Sweden

I. Makos · G. Sansone  
Physikalisches Institut, Albert-Ludwigs-Universität, Stefan-Meier-Straße 19, 79104 Freiburg, Germany

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spectral range. In this chapter, we review four such sources developed through collaborative efforts between FORTH, ELI-ALPS and the University of Lund together with recent results obtained in the above mentioned topics utilizing these sources.

## 1.1 Introduction

Since the first demonstration of laser generated attosecond pulses, around the change of the century, most of the related applications are based on linear extreme ultra-violet (EUV) processes and EUV-infrared (IR) cross-correlation and pump-probe approaches. This is due to the commonly low energies of the generated EUV pulses. Such approaches, refined over the years, have led to a large number of noteworthy novel results. However, in specific cases they are associated with important complications [1–3]. Approaches based solely on EUV pulses, utilizing non-linear EUV processes, bypass such complications [1–3]. Indeed, almost since the advent of attosecond pulses targeted efforts achieved high enough EUV intensities as to induce two-EUV-photon processes [4]. It should be noted here, that prior to the arrival of two-EUV-photon processes induced by attosecond pulses, rich pioneering work has been implemented on non-linear EUV-processes induced by individual harmonics mainly by Japanese teams [5–8]. This early achievement, followed by systematic developments and improvements by a few groups worldwide led to significant advances in the inaugurated era of non-linear EUV processes. To mention some of the early works, multi-EUV-photon ionization by attosecond pulse trains (APT) was demonstrated in the first decade of this century [4, 9, 10]. The first applications of the non-linear EUV processes were in the temporal characterization of APTs through the 2<sup>nd</sup> order intensity volume autocorrelation (2<sup>nd</sup> IVAC) technique [11]. Later on, the technique was advanced to 2<sup>nd</sup> order interferometric autocorrelation [12] and energy resolved autocorrelation [13], towards EUV Frequency Resolved Optical Gating [FROG].

The two-EUV-photon processes induced by APTs, i.e., harmonic combs, were followed up by two-EUV-photon processes induced by coherent EUV quasi continua, supporting isolated attosecond pulses. This has led to the first EUV-pump-EUV-probe experiments addressing one fs scale dynamics in atomic [14] and molecular [15] systems. The non-linear processes utilized in these experiments were two-EUV-photon direct double ionization [16] and two-EUV-photon dissociative ionization respectively. Due to the lack of carrier envelope phase (CEP) stabilized laser pulses,

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K. Varju

Department of Optics and Quantum Electronics, University of Szeged, Dom tér 9, 6720 Szeged, Hungary

L. A. A. Nikolopoulos

School of Physical Sciences, Dublin City University, Collins Ave, Dublin 9, Ireland

A. Emmanouilidou

Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

these experiments could not be proven being attosecond resolved EUV-pump-EUV-probe experiments, despite the fact that they most probably are.

In all the above experiments, the key factor is the EUV focused intensity. The main limitations in the EUV intensity are the depletion of the generating medium and the reabsorption of the generated EUV radiation by the generating medium. These limitations can be partially overcome using high peak power laser pulses and large geometrical interaction cross sections achieved either through loose focusing or off-focus harmonic generation. Applying loose focusing geometries and quasi-phase-matching in a dual gas-jet set-up, the highest laser induced EUV pulse peak power (20 GW) has been recently demonstrated in the spectral region  $33 \pm 15$  eV [17]. Two years later it was verified that the EUV radiation of this source was emitted in the form of an attosecond pulse train [18]. Utilizing short driving wavelengths and off-focus harmonic generation high EUV intensities have been achieved at 22 eV photon energy [19]. Triggered by the high peak powers achieved using loose focusing, two attosecond beam lines have been developed and installed at the European Research Infrastructure Extreme Light Infrastructure—Attosecond Pulse Light Source (ELI-ALPS) [20]. Indeed, non-linear-EUV processes at  $40 \pm 5$  eV photon energies have been very recently established in one of the beam-lines of ELI-ALPS [21]. Loose focusing conditions led to high EUV pulse energies also in set-ups emitting highly elliptically polarized harmonics [22].

Although strong field effects at short wavelengths are not expected due to the small ponderomotive energy, EUV intensities have reached levels that allow observation of such effects at photon energies  $20 \pm 3$  eV prior to depletion of the medium [23].

In this chapter we review the recent advancements in high peak power laser-driven EUV sources at FORTH and ELI-ALPS and their use in non-linear and strong field applications. Those include multi-EUV-photon multiple ionization measurements, direct multiple ionization processes and ponderomotive shifts in photoelectron spectra.

## 1.2 High EUV Peak Power Beamlines

In this section, we give an overview of four attosecond beam-lines. Two of them are located at FORTH and the other two at ELI-ALPS.

### 1.2.1 *The FORTH High EUV Peak Power Beamlines*

At the attosecond science and technology laboratory of FORTH is operating for five years a 20 GW attosecond beam line, shown in Fig. 1.1. It is a 18 m long beamline driven by a 10 Hz repetition rate Ti:sapphire laser system, which emits 20 fs long pulses at 800 nm central wavelength and pulse energy up to  $\approx 400$  mJ/pulse. The attosecond pulse emission is based on high-order harmonic generation in gaseous



**Fig. 1.1** The 18 m long 20GW attosecond beamline of the attosecond science and technology laboratory of FORTH. Attosecond emission is based on high harmonic generation in noble gas media introduced through a dual-pulsed-jet configuration

media. A fraction of the maximum laser pulse energy of 25–45 mJ is focused in gas jets with a spherical mirror of 9 m focal length placed in an optical set-up allowing almost normal incidence on the mirror to minimize astigmatic deformations at the focus. The gaseous non-linear medium is introduced by a dual-pulsed-jet configuration operated with noble gases (usually Ar or Xe). Single-jet operation is also possible.

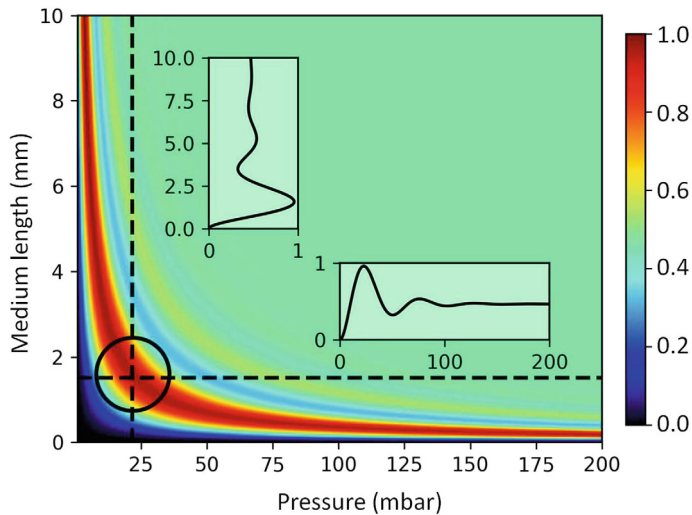
The two gas jets setup is used for establishing quasi-phase matching conditions [25, 26] thus maximizing the throughput of the EUV source. Quasi-phase matching is achieved by changing the distance between the two jets after optimization of the harmonic emission in each jet. Due to the long focal length used the jet distance can be adjusted with high accuracy. Optimization of the harmonic emission in each jet is performed by varying the gas pressure and the medium length. It is well established [24–26] that for coherence lengths  $L_{\text{coh}}$  much larger than the absorption length  $L_{\text{abs}}$  and the medium length  $L_{\text{med}}$  the EUV yield is proportional to  $(P \cdot L_{\text{med}})^2$ ,  $P$  being the generating gas pressure. The product  $P \cdot L_{\text{med}}$  can though not exceed a maximum value as above this value reabsorption of the EUV either due to the high pressure or propagation length reduces the yield. This leads to a phase matching gas pressure—medium length hyperbola (see Fig. 1.2). For the given beamline the conditions  $L_{\text{coh}} \gg L_{\text{abs}}$  and  $L_{\text{coh}} \gg L_{\text{med}}$  are fulfilled. The dependence of the harmonic yield on the pressure and medium length for an Ar gas at a laser intensity  $1.5 \times 10^{14} \text{ W cm}^{-2}$  is calculated numerically and is shown in Fig. 1.2.

In the experiment, the medium length can be, to some extent, changed by moving the jet along its axis. Optimization is occurring by mainly varying the gas pressure.

The optimized operation of the beamline with Ar and Xe as generating media and the setup with one or two gas jets has led to EUV pulse energies summarized in Table 1.1.

The pulse energies have been measured using a calibrated EUV photodiode. The two pulse energy values given for each case in Table 1.1 are deduced using two different calibration curves published in the documents of the manufacturing company (see also [18] where the energy determination procedure is presented in detail). Using a dual gas jet arrangement the emitted harmonic energy could be controlled by varying the distance between the two jets. This is shown for two Ar jets in Fig. 1.3.

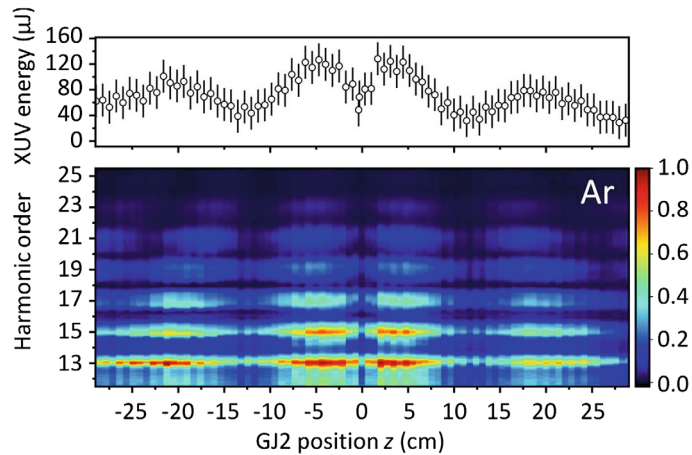




**Fig. 1.2** Harmonic yield calculated numerically for an Ar gas as generating medium as a function of the gas pressure and medium length. The laser intensity is  $I_L \approx 1.5 \times 10^{14} \text{ W cm}^{-2}$ . The black circle depicts the conditions used in the experiments. The insets show a line-out of the harmonic yield along the dashed lines. Figure reproduced from Ref. [17]

**Table 1.1** EUV pulse energies emitted from single and dual Ar and Xe gas jets

	Ar	Xe
Single gas jet	75/48 $\mu\text{J}$	135/88 $\mu\text{J}$
Dual gas jet	130/85 $\mu\text{J}$	230/150 $\mu\text{J}$



**Fig. 1.3** Dependence of the EUV energy generated in Ar gas on the distance of the two Ar gas jets. The error bars are one standard deviation from the mean. Figure reproduced from Ref. [17]

The temporal characterization of the superposition of harmonics 11, 13, 15, emitted by one Xe gas jet and transmitted through a Sn filter has been performed through 2nd order IVAC runs. As non-linear process the two photon double ionization of Ar was used. The beam splitter of the EUV delay line is a gold coated bisected spherical mirror [11]. The measured traces for the attosecond bursts of the APT and the envelope are shown in Fig. 1.4. The measured duration of the individual pulses in the APT is  $660 \pm 80$  as and that of the envelope  $9.8 \pm 0.9$  fs [18]. From the measured EUV pulse duration and focal spot size that was recorded using an ion microscope [27], a maximum achieved focused EUV intensity of  $\sim 7 \times 10^{15}$  W/cm<sup>2</sup> has been deduced. This value can be further increased using high throughput EUV optical elements such as EUV multilayer mirrors. These results establish the described beamline as the most intense laser driven attosecond beamline so far.

The beamline further hosts a compact-collinear polarization gating set-up with which broadband, coherent XUV quasi-continua have been generated by many-cycle infrared fields [28].

The set-up used is illustrated in Fig. 1.5 together with a single shot image of the spatial distribution of the quasi-continuum EUV radiation. The components shown are: ZO  $\lambda/2$ : Zero order half wave plate, MO  $\lambda/4$ : Multiple order quarter wave plate, ZO  $\lambda/4$ : Zero order quarter wave plate, PBS: polarizing beam splitter, FM: flat mirror, SM: spherical mirror (The FM and SM mirrors have been placed very close to the normal incidence with respect to the incoming beam). Xe GJ: pulsed-jet filled with Xenon, TS: translation stage, used to move in- and out the set-up from the beam path. The principle and operational details of the set-up are described elsewhere [28, 29].

EUV spectra generated in a Xe gas, with (blue line) and without (black line) polarization gating are shown in Fig. 1.6. The insets (a) and (b) show measured traces of the calibrated EUV photodiode that is used for the EUV pulse energy measurement. The spectra are averages of 150 shots. The switching from discrete to continuum spectra when the polarization gating is turned on is clear. The quasi-continuum spectrum spans the range 17–32 eV and has pulse energy of about 1  $\mu$ J. This pulse energy, when focused is sufficient in inducing non-linear EUV processes as described in Sect. 1.3.

Single-shot EUV spectra carry information about the CEP of the driving IR field. This is evidenced by the measured harmonic frequency shift  $\Delta\omega$  (black dot and red dashed-dot curves), along with the EUV spectrum exhibiting a continuum structure (blue curve) shown in Fig. 1.7. This effect has been previously used in measuring the absolute carrier-envelope phase of many-cycle laser fields. The insets (i) and (ii) show calculated Fourier Transform Limited (FTL) pulses resulted from the blue and the red dashed-dot curves respectively.

A second EUV attosecond source at the attosecond science and technology laboratory of FORTH, driven by the same laser and with a 3 m length focusing system has been operating for more than 15 years. Recently the source is equipped with a unit that allows the generation of highly elliptically polarized harmonics with controllable ellipticity and variable central wavelength. Briefly, the efficient generation and tunability of such harmonics in an Ar gas is realized by employing intense two-color