

# Final Report on Basic Technology Project ‘Attosecond Technology – Light Sources, Metrology and Applications’ GR/S22400/01

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## 1. Introduction

This is the Final Report for an RCUK/EPSCRC funded Basic Technology Project (2004-8) that was focused on developing *Attosecond Technology*. This project was a collaboration between Imperial College London (hub), the universities of Oxford, Birmingham and Reading, University College London and RAL (CLF). Dr John Tisch (IC) was the overall Project Manager for the project, and Prof Jon Marangos (IC) was the coordinator.

Attosecond Technology has arisen in this project from the synergy of a range of technologies (high-power femtosecond lasers, ultrafast and XUV optics, electron imaging, etc.) and through increased understanding of the coherent interaction of atomic and molecular systems with both intense (‘few-cycle’) femtosecond laser pulses and synchronised bursts of XUV radiation of attosecond duration.

Although we could not have predicted all the outcomes at the start, as a team we feel that the project has been a great success. We have succeeded in developing new technology and carrying out world-class new science. We have built up essentially from scratch significant infrastructure and expertise in this exciting new field to serve a growing UK community, and have made huge strides in our understanding of how to generate, characterise and control attosecond pulses. We have also learned how to make attosecond resolution measurements in atomic, molecular and surface science systems, a notable highlight being the fastest ever measurement (100 as resolution) of molecular dynamics that we made in 2006<sup>1</sup>.

The research carried out in this project will provide the tools to enable scientists to observe directly the dynamics of atoms and molecules on their natural length (Ångstrom) and time (sub-femtosecond) scales. Electronic motion on this timescale underpins many microscopic natural phenomena, such as charge transfer within molecules and at surfaces. Therefore, the research promises to have impact and application across the fields of physics, chemistry and eventually biology, as well as in nano-science and engineering.

The project has generated 49 papers in peer reviewed journals, including high impact publications in *Science*, *Nature Physics* and *Physical Review Letters* and more than 50 invited talks at leading conferences. Twelve PhDs will have directly arisen from the project.

We detail the major project achievements below.

## 2. Major Project Achievements

### i. First high-power few-cycle laser source in UK

One of the most important deliverables of the current Basic Technology project has been the development of the high-power, CEP-stabilised, few-cycle laser system<sup>2</sup> that is used as the driver for attosecond pulse generation via high harmonic generation (HHG). This system uses the process of hollow-fibre pulse compression (HFPC) which allows us to generate pulses of >0.5mJ at pulse durations of ~6fs (~100 GW peak power) at 1 kHz repetition rates. This was the first such system in the UK and is one of only a handful in the world capable of achieving these pulse parameters. This few-cycle technology was transferred to user facilities at the Central Laser Facility, RAL – see ‘Beneficiaries’.

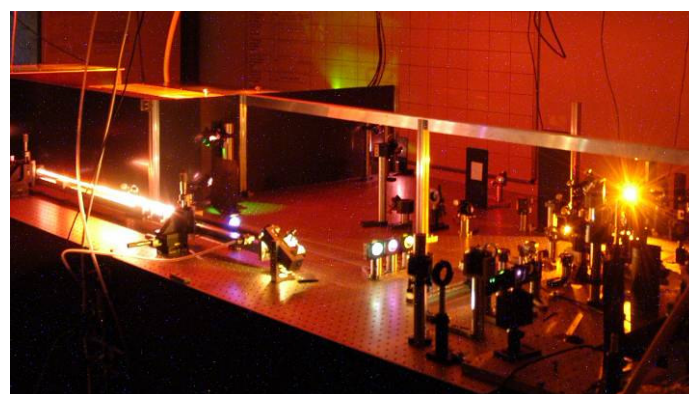


Figure 1 We developed a state-of-the-art few-cycle laser system.

### ii. High-energy few-cycle laser OPCPA system

Significant progress has been made towards the extremely challenging and adventurous goal of developing an optical parametric chirped pulse amplification (OPCPA) laser system to produce few-mJ, carrier envelope phase-stabilised few-cycle pulses at repetition rates up to 1 kHz. This system could provide a world-leading source for attosecond pulse generation as well as a range of other ultrafast experiments.

We have successfully developed a seed source for the OPCPA laser, achieving the key requirements of producing a CEP stable pulse of more than 400 nm bandwidth, stretching this to the appropriate duration, and accurately re-compressing it. The novel stretcher-compressor system includes transmission gratings and gives us unique access to multiple stretched pulse durations, enabling us to optimise the seed simultaneously for each of the amplifier stages.

The optical parametric amplifiers (OPAs) are pumped by a custom-built, high-peak and average power diode-pumped

<sup>1</sup> Baker *et al.* *Science* **312**, 424 (2006).

<sup>2</sup> J.S. Robinson *et al.* *Appl. Phys. B* **85**, 525 (2006).

Nd:YLF laser (developed with additional CLF support), producing 50 mJ, 40 ps, 523.5 nm pulses and capable of operating at 1 kHz. This system is extremely stable, with 0.8% pulse to pulse energy fluctuations, and recently we have begun to work towards the 1kHz repetition, with an increase from 10 to 40 Hz in the past month.

Testing of the amplification stages has shown a gain of  $10^5$  across a 240 nm bandwidth, sufficient for few-cycle pulse durations of  $<9$  fs. Work continues on this project, and we have recently reached energies of 0.9 mJ before compression, which is comparable to HFPC systems.

### iii. Development of a state-of-the-art attosecond XUV beamline

Another major deliverable of the current project is the XUV vacuum beamline that has been built at Imperial College. This is used for the generation, filtering, focusing and delivery of attosecond XUV and precisely synchronised IR pulses (20 as precision using a stabilised Michelson we constructed) to a range of experiments in several “end-stations”, including gas phase molecular and surface science studies. It also incorporates built-in diagnostics for measuring the spatially-resolved spectrum of the XUV pulses as well as their pulse duration using a variation on the atomic streak camera method<sup>3</sup>.

Great effort was spent in making the beamline as flexible and as versatile as possible for current and future experiments. Many technological challenges had to be met in its construction, from the development of novel gas targets for HHG capable of kHz repetition rates to the design and implementation of a vibration isolation system for optics in the vacuum chambers using concepts borrowed from gravity wave detection. The beamline and the few-cycle source together constitute the ‘system’ for doing Attosecond Science.

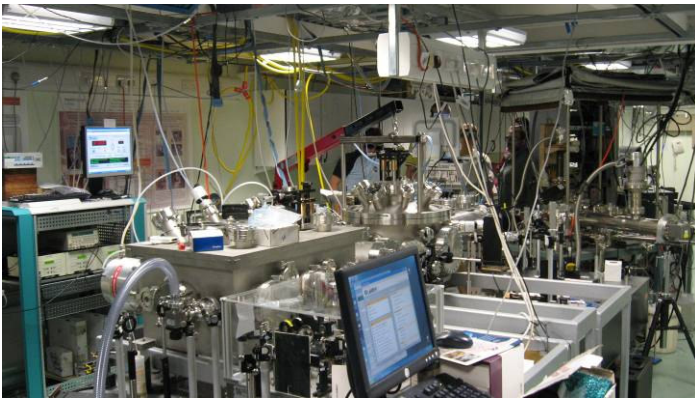


Figure 2 The XUV Attosecond beamline we have constructed provides a flexible system for doing Attosecond Science.

### iv. A new technique for the measurement of the absolute phase of few-cycle pulses

We made the first observation of ‘half-cycle cut-offs’ (or HCOs) during high harmonic generation (HHG). HCOs are the highest frequency bursts of radiation emitted each half-cycle of the laser pulse during HHG. They have been theoretically predicted but had not previously been measured. We showed that HCOs are extremely sensitive to the sub-cycle (ie. attosecond) details of the electric field evolution of the few-cycle driving laser pulse. In particular, the centre frequency of the HCOs was found to be a function of the CEP of the laser pulse, an essential parameter in the characterisation of few-

cycle pulses, but notoriously difficult to measure. We used this to develop a completely new measurement technique of the CEP that offers many advantages over the handful of existing measurement techniques. This result was published in *Nature Physics* in January 2007<sup>4</sup>, making the front cover of that issue, and the technique has already been picked up by other groups<sup>5</sup>.

### v. New all-optical techniques for attosecond metrology

The production of attosecond XUV pulses by means of high harmonic generation is predicated on the ability to measure the pulses in a simple and robust manner<sup>6</sup>. We have contributed to this by developing a method to measure the electric field of such pulses directly from the XUV radiation itself. This approach differs from the more common methods based on photoelectron spectroscopy both because it makes use of very different physics, and because it offers the possibility of application on a wider range of systems, with single shot capability. We have also contributed to the development of underpinning technologies that are crucial for generating the few-cycle infrared optical pulses that are used to generate attosecond XUV pulses in HHG. Our approach is based on the extension of spectral shearing interferometry to these extreme wavelengths and pulse durations.

The means for characterizing the XUV radiation involves the interference of light from independent sources driven by two separate drive pulses. This interferogram provides a direct measurement of the dipole phase of the radiating atoms, and thus to the electron wavepacket dynamics in the strong drive field. Careful shaping of the drive pulses enables the complete spatio-temporal field of the XUV pulses to be measured<sup>6</sup>. We have also proposed the idea of spectral interference for photoelectron spectroscopy<sup>7</sup> realising that the electron wavefunction is a faithful map of the XUV radiation spectrum. This has been analysed primarily for pulse characterisation, but may also be used for determining the structure of the initial electronic quantum state, which itself reflects the structure of the atom or molecule from which it was ionised.

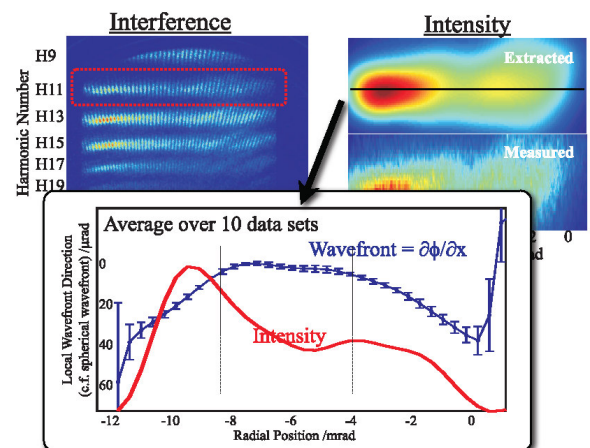


Figure 3 Lateral spatial shearing interferometry of the 11th harmonic in Ar driven by an optical pulse at 800 nm. The harmonic interferogram is shown in the upper left, with the extracted spatio-spectral intensity and phase in the upper right. The lower figure shows the spectral dependence of the wavefront tilt across the beam.

As a first step to implementing a full XUV characterization using spectral shearing interferometry, we have developed an

<sup>3</sup> R. Kienberger *et al.* *Nature* **427** 817 (2004).

<sup>4</sup> C.A. Haworth *et al.* *Nature Physics* **3** 52 (2007)

<sup>5</sup> T. Pfeifer *et al.* *Optics Express* **15** 17120 (2007)

<sup>6</sup> E. Cormier *et al.* *Phys. Rev. Lett.* **94** 033905 (2005)

<sup>7</sup> E.M. Kosik *J.Mod. Opt.* 20361 (2005)

interferometer for the generation of two spectrally sheared few-cycle infrared pulses which are controllable temporally and spatially. The two pulses can independently be spectrally sheared and delayed in time as well as spatially sheared and tilted. This device has been employed to characterize space-time-coupling of high harmonics generated in argon. Figure 1 shows the complete spatio-temporal phase of the 11th harmonic of an 800 nm pulse converted in Ar.

We have also developed a robust and reliable method for characterising the space-time field of the infrared drive pulses for such sources with the minimum amount of data (SEA-SPIDER<sup>8</sup>) in collaboration with the Max-Born Institute.

#### vi. Fastest ever measurement of molecular dynamics

In photochemistry, it is customary to apply the Franck-Condon principle which assumes that the atoms (nuclei) in a molecule are stationary while an electron makes a transition. However, in the case of protons – the lightest nuclear components in molecules – significant motion may occur on the timescale of electronic motion. This motion is extremely important because it can be the precursor to chemical transformation. We pioneered a new technique known as PACER<sup>9</sup> that allowed us to observe this motion for the first time. We tracked the movement of the protons in the H<sub>2</sub> molecule in a ~1 fs time window following its photoionisation by an intense, few-cycle laser pulse. Within this 1 fs window, we achieved an unprecedented time-resolution of ~100 as, allowing us to effectively ‘make a movie’ of the H<sub>2</sub><sup>+</sup> molecular ion after an electron had been removed. We also applied the PACER technique to the more complex molecule methane (CH<sub>4</sub>) and found evidence for a significant structural transformation occurring on the ~1 fs time scale. This result has stimulated considerable interest from quantum chemists keen to interpret this surprising result. Our data in H<sub>2</sub> and CH<sub>4</sub> represent the fastest ever measurement of dynamics in molecules. This work was published in *Science* in 2006<sup>1</sup> (cited 65 times so far) and received considerable press coverage, including from the BBC.

#### vii. Influential work on attosecond pulse trains

Though much attention has been given to the generation of isolated attosecond pulses (ie one per laser pulse), *trains* of attosecond pulses – typically one pulse per half cycle – offer some very interesting experimental possibilities, and can be generated with longer drive laser pulses (typically >20 fs). In collaboration with other European Attosecond groups (notably, the group of Pascal Salieres in Saclay, Paris), members of our team have made a number of significant contributions to the field of attosecond pulse trains. Perhaps the most important of these was the experimental confirmation<sup>10</sup> of an intrinsic frequency chirp (known as the ‘attosecond chirp’) on the attosecond pulses generated by HHG, as well as methods to control this chirp<sup>11,12</sup>. Ref 10 has been particularly influential, being cited already 149 times.

One of the challenges of pulse train work is the accurate reconstruction of the electric field of the train from IR-XUV pump-probe photoelectron data. A new method to do this was

proposed and verified experimentally<sup>13</sup> through the same collaboration. And recently, we contributed to the development of a new method for controlling the generation of attosecond pulse trains using interference effects in molecules<sup>14</sup>. This attosecond ‘pulse shaping’ capability will be important for future applications in the sub-femtosecond coherent control of atomic and molecular processes.

#### viii. New measurements of ultrafast electron dynamics at surfaces

During the project an ultra high vacuum (UHV) surface chamber equipped with surface preparation tools, a scanning tunnelling microscope (STM) and Time of Flight (ToF) mass spectrometer was successfully built and attached to the attosecond beamline at Imperial College.

Using this apparatus we have made measurements of ultrafast electron dynamics at the surface of highly-oriented pyrolytic graphite (HOPG) which shed new light on the involvement of surface plasmons in the electron emission process at surfaces.

ToF techniques were used to study photoelectrons produced at the HOPG surface when irradiated with 13fs pulses from the HFPC laser system (centre wavelength of ~800nm, vacuum intensity at surface ~10<sup>11</sup>Wcm<sup>-2</sup>). Electrons with kinetic energies significantly higher than expected from multiphoton transitions were detected<sup>15</sup>.

The process leading to emission of these electrons was found to have a highly nonlinear intensity-dependence, and their lifetime was studied via both IR/IR and IR/XUV pump-probe experiments. Strong evidence was found of surface plasmon involvement in the electron emission process, and thus high-energy electrons can be attributed to the enhancement of the laser field at the surface and subsequent tunnel ionisation. This is supported by photoelectron spectra taken from XUV irradiation of HOPG, which shows the structure of the valence band at the surface and bears great similarities to measurements made using IR wavelengths.

Silicon nanopillar-structured surfaces were also studied using IR/IR pump-probe measurements and XUV photoelectron spectra. Ongoing analysis suggests that similar effects are observed.

#### ix. Important contributions to the modelling of attosecond pulse generation and propagation

The use of sophisticated computer simulations of attosecond pulse generation and propagation have played a vital role in the success of this project. As well as a ‘standard’ code based on solution of the Time Dependent Schrödinger Equation, two innovative new computer codes were developed during this project: a) a robust and very fast, ‘quantum orbit’ code for simulating HHG and attosecond pulse generation in single atoms<sup>16</sup>, and b) a full propagation code<sup>17</sup> for simulating the macroscopic aspects of HHG, ie phase-matching in a gas target.

The robustness and speed of the quantum-orbit code were essential for its use in simulations of the propagation of

<sup>8</sup> A. S. Wyatt et al. *Opt. Lett.* **31** 1914 (2006)

<sup>9</sup> Probing Attosecond dynamics using Chirped Electron Recollisions

<sup>10</sup> Y. Mairesse et al. *Science* **302** 1540 (2003)

<sup>11</sup> Y. Mairesse et al. *Phys. Rev. Lett.* **93** 163901 (2004)

<sup>12</sup> Y. Mairesse et al. *Laser Physics* **15** 863(2005)

<sup>13</sup> K. Varjú et al. *Phys. Rev. Lett.* **95** 243901(2005)

<sup>14</sup> W. Boutou et al. *Nature Physics* doi 10.1038/nphys964, Letters (04 May 2008)]

<sup>15</sup> E. Catton et al. “Electron tunnelling from graphite surfaces in short pulse and strong field laser regime” in preparation (May 2008).

<sup>16</sup> L.E.Chipperfield et al. *J. Mod. Opt.* **52** 243 (2005)

<sup>17</sup> L.E.Chipperfield et al. *Opt. Comm.* **264** 494 (2006)

attosecond pulses which require a very large number of single-atom calculations to be carried out for different space-time points in the interaction volume. This theoretical capability underpinned the HCO work (Section iv) by predicting the sensitivity of attosecond pulse production on the CEP of the driving laser pulse, and through the discovery of a new phase-matching regime that favours the formation of readily detected HCO in the HHG spectrum.

Increasing the maximum electron recollision energy is an important goal for optimising HHG as a source of attosecond XUV pulses. Recently we have investigated this problem<sup>18</sup> and found analytically the optimum waveform which maximises the electron recollision energy for a given average fluence and oscillation period. This waveform is a simple sawtooth with a DC offset. The maximum electron recollision energy produced by such a waveform is over three times greater than for a monochromatic field. However, it is not currently possible to produce this sawtooth waveform. Therefore, with the aid of a genetic algorithm, we have investigated more readily implemented waveforms that approximate the ideal case. We have found that the addition of a strong low frequency field along with some weak high frequency harmonics produces a waveform that increases the electron recollision energy by a factor of 2.5, compared to the single frequency case, without any loss in yield. This provides a route for producing shorter wavelength attosecond pulses, eg for attosecond probing of x-ray transitions in atoms.

#### x. Highly successful international workshop held in London in 2006

We held an international workshop ‘Ultrafast Dynamic Imaging’ at Imperial College in April 2006, supported by the BT grant, the ESF (who made a very significant financial contribution) and through commercial sponsorship. This was a great success, attracting more than 150 delegates, including twenty field-leading invited speakers in ultrafast imaging (femtosecond to attosecond) from the accelerator and laser communities. It provided a fantastic opportunity to highlight the UK’s growing international status in the attosecond field, and to showcase the Project through talks, posters and laboratory visits. The proceedings were published in a special issue of the *Journal of Modern Optics*<sup>19</sup>.

Recently, funding has been again secured from the ESF to hold a follow-up workshop in Naples in 2009, with the likelihood of this becoming a regular series in the future.

### 3. Beneficiaries

Attosecond Technology provides the tools to make real-time observation of the fundamental processes of chemistry, biology and materials science that are mediated by the electronic and nuclear motions of the constituent atoms on the attosecond time-scale. New measurements lead to increased understanding of these processes and ultimately to the ability to control them. The technology is opening up many new scientific opportunities.

In physical sciences, for instance, the direct measurement of electron dynamics in chemical reactions, the study of electronic motion in strong fields, the exploration of dynamics of inner shell excitations in matter is becoming possible. We

anticipate also wide applications in nano-scale and biological systems. However, the field is still in its infancy, and it is anticipated that it may take five to ten years for applications involving more complex (eg. biological) systems to be realised.

On the more immediate time scale, we have transferred the few-cycle technology (HFPC) to the Central Laser Facility at RAL in 2005 and 2008, providing them with a new high-power, sub-10 fs capability in TA1 and TA2 on the Astra Laser. In TA1 this has been used in a number of high-impact experiments<sup>20</sup> by university user groups. It was also the catalyst for the development of a major new user facility at RAL (Astra-Artemis) currently under construction to provide few-cycle and short-wavelength pulses. Some of our team (JPM, JWGT) are co-investigators on this project. Technology from the current project will continue to be fed into this development, especially in the areas of CEP stabilisation and HHG source technology and the XUV beamline.

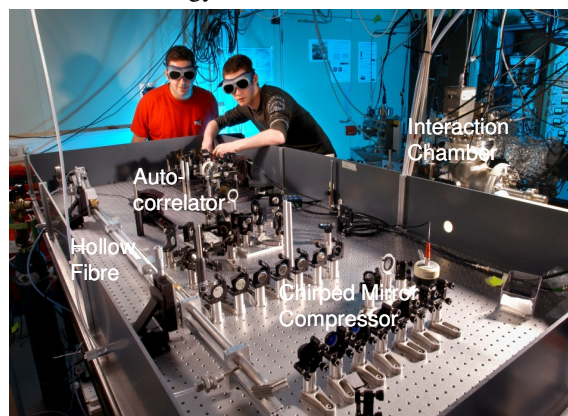


Figure 4 We transferred the few-cycle technology to RAL.

### 4. Training

This project has been instrumental in expanding the Attosecond Science & Technology base in the UK through training of a dozen PhD students. More than five other PhD students funded by other sources benefitted from their partial involvement with this project.

Through graduate lectures, workshops (one organized by themselves), conferences and summer schools, and through ‘hands-on’ instruction, these students have received extensive training in areas including ultrafast high-power laser systems notably CEP-stabilized and OPCPA systems, pulse compression techniques, nonlinear optics, strong-field physics in gas and condensed phase targets, attosecond pulse generation and characterization, and advanced theoretical analysis and numerical modelling of many of the above.

The students benefitted greatly from working together on collaborative experiments conducted on the Attosecond Beamline at Imperial College. As well as providing a valuable opportunity for scientific networking, this enabled the exchange of expertise, eg. the ‘source’ people learned about the intricacies of conducting surface-science experiments and vice versa.

We have also participated in a number of European Research Training Networks related to Attosecond Science & Technology.

<sup>18</sup> L.E. Chipperfield et al. ‘Optimising the energy of laser driven electron recollisions’ submitted to *Phys. Rev. Lett.* April (2008)

<sup>19</sup> M. Yu Ivanov and J.P. Marangos eds. *J. Mod. Opt.* **54**, nos.7-9 (2007).

<sup>20</sup> Eg. R. Torres et al. *Phys. Rev. Lett.* **98**, 203007 (2007), I.D. Williams et al. *Phys. Rev. Lett.* **99** 173002 (2007)



Figure 5 The PhD students in the Attosecond project at a one-day workshop that they organised themselves.

## 5. Public Engagement

The project website [www.attosecond.org](http://www.attosecond.org) has been an extremely useful means of publicising our activities. It is currently the first site displayed by Google when searching UK pages for the keyword 'attosecond' and the fourth when searching the web in general.

Members of our team have given more than 10 talks to school groups, (mostly 6th form students) on topics closely connected to this project, eg. 'Don't blink or you'll miss it – the physics of making attosecond pulses' at the PhysSoc 'Einstein Day' at Imperial College March 2006.

Also a group from our team has been selected to exhibit at the prestigious Royal Society Summer Exhibition 2008. This is the Society's flagship public engagement activity attracting thousands of visitors from the public, especially 16+ students and science teachers, and drawing considerable press interest. The exhibit is entitled 'Can we freeze time? Using lasers to film the secret lives of atoms – frame by frame' and is directly concerned with Attosecond Science & Technology.

## 6. Future Work

The field of Attosecond Science has moved rapidly over the last five years which fully vindicates the decision to invest in the development of new technology. We are in a very strong position to further develop the technology and carry out new science that will have a wide-ranging impact. We have recently been awarded an EPSRC Translation Grant to take strategically chosen Attosecond topics to the next level. Some of our future research goals include:

- To develop techniques for isolated attosecond pulse production based on propagation-filtering to the point where they are robust enough for widespread application.
- To construct a Raman attosecond source in the deep UV to VUV – a new wavelength range for attosecond pulses – with prospects for the fastest-ever pump-probe molecular measurements.
- To take Attosecond Metrology to the next level by pioneering new methods for probing attosecond dynamics and molecular tomography through XUV interferometry.

## 7. Research Outputs

49 publications in peer-reviewed journals have arisen to date from the project, with more submitted or in preparation. Publications have appeared in the following high-profile journals: *Science* (2), *Nature Physics* (2), *PRL* (8), *Optics Letters* (6).

Members of our team have also given more than 50 invited talks at international conferences, a similar number of other conference and workshop presentations (contributed talks and

posters) and approximately 30 university seminars in the UK and abroad.

We note that some outputs have arisen from work partially supported by other grants in addition to the Basic Technology Grant. We gratefully acknowledge:

- EU-FP6 XTRA (MRTN-CT-2003-505138) Attosecond Network Attosecond Metrology, Attosecond Pulse Trains
- EPSRC GR/N11292 Raman work at IC
- EPSRC EP/C530764 Molecular Imaging & Attosecond Pulse Trains
- EPSRC EP/P028063 Electron Control

We also had support for collaboration with Italian groups from Royal Society 2002-2004 (joint project grant) and from Laser Lab Europe for experiments conducted in Milan.

### i. PhDs

1. Joseph Robinson (IC) "The generation and application of few-cycle laser pulses" (2006)
2. Charles Haworth (IC) "Development and Application of an Attosecond Light Source Based on High Harmonic Generation" (2007)
3. Philip Bates (RAL Project Student registered at IC) "Optical Parametric Chirped Pulse Amplification in the few-cycle regime" (2008)
4. Luke Chipperfield (IC) High Harmonic Generation with Few-Cycle Pulses (2007)
5. Justin Steele-Davies (Reading) "Attosecond Physics and Technology" submitted (2008)
6. Gavin Waters (Reading) "High-harmonic generation from noble gases exposed to intense laser fields" expected (2008)
7. Adam Wyatt (Ox) "Spectral interferometry for the complete characterisation of near infrared femtosecond and extreme ultraviolet attosecond pulses" (2007)
8. Phil Bustard (Ox) "Non-adiabatic molecular modulation for ultrabroadband pulse generation" expected (2008)
9. Tobias Witting (Ox) "Photoelectron chronocyclic tomography for attosecond pulse characterization" expected (2009).
10. Dane Austin (Ox) "Attosecond control of high-harmonic radiation" expected (2010)
11. Christophe Huchon: (B'ham) "Time of flight mass spectrometry of graphite: ultrafast laser ablation and photoelectron spectroscopy" submitted (2008)
12. Emma Catton (B'ham) "Electron dynamics at semiconductor surfaces in the short pulse, strong field regime" expected (2009)

### ii. Published in refereed journals

1. W. Bouto *et al.* "Coherent Control of attosecond emission from aligned molecules" *Nature Physics* doi 10.1038/nphys964, Letters (04 May 2008)
2. J.P. Marangos *et al.* "Dynamic Imaging of molecules using high order harmonic generation" *Physical Chemistry Chemical Physics* 10 35 (2008)
3. M.E. Anderson *et al.* "SPIDER: A decade of measuring ultrashort pulses" *Laser Phys. Lett.* 5 259 (2008)
4. M.E. Anderson *et al.* "Gold-spider: spectral phase interferometry for direct electric field reconstruction utilizing sum-frequency generation from a gold surface", *J. Opt. Soc. Am. B* 25 A13 (2008)
5. N. Kajumba *et al.* "Measurement of the electronic structure from high harmonic generation in non-adiabatically aligned polyatomic molecules" *New J. of Physics* 10 025008 (2008)
6. S. Baker *et al.* "Probing fast nuclear wavepackets in light molecules: monitoring structural rearrangements on an attosecond timescale" *J. Mod. Opt.* 54 1011 (2007)
7. A. Zair *et al.* "Quantum path interferences in high-order harmonic generation", *Phys. Rev. Lett.* 100 143902 (2007)
8. A. Kaplan *et al.* "Nonlinearity and time-resolved studies of ion emission in laser ablation of graphite" in press *Applied Physics A* (2007)
9. A. Kaplan *et al.* "Emission of ions and charged clusters due to impulsive Coulomb explosion in ultrafast laser ablation of graphite", *Phys. Rev. B* 76 073401 (2007)
10. S.-P. Gorza *et al.*, "Tailoring the phase matching function for ultrashort pulse characterization by spectral shearing interferometry", *J. Opt. Soc. Am. B* 24 2064 (2007)

11. M.Lenner et al "Nanoscope Coulomb explosion in single-shot graphite ablation", *Appl. Phys. Lett.* 90, 153119 (2007)
  12. C.A.Haworth et al., "Half-cycle cutoffs in harmonic spectra and robust carrier-envelope phase retrieval", *Nature Physics*, 3, 52 (2007)
  13. J.Levesque et al. "High harmonic generation and the role of atomic orbital wave functions", *Phys. Rev. Lett.*, 98 183903 (2007)
  14. R.Torres et al., "Probing orbital structure of polyatomic molecules by high-order harmonic generation", *Phys. Rev. Lett.*, 98, 239903 (2007)
  15. I.N.Ross et al. "Contrast limitation due to pump noise in an optical parametric chirped pulse amplification system", *Opt. Comm.*, 273, 510 (2007)
  16. A.S.Radunsky et al. "Compact spectral shearing interferometer for ultrashort pulse characterization", *Opt. Lett.*, 32, 181 (2007)
  17. A.Wyatt et al., "Sub-10fs pulse measurement using spatially encoded SPIDER", *Opt. Lett.*, 31, 1914 (2006)
  18. G.Stibenz et al, "Advanced methods for the characterization of few-cycle light pulses: a comparison", *Appl. Phys. B*, 83, 511 (2006)
  19. A.Radunsky et al., "Simplified SPIDER apparatus using a thick nonlinear crystal", *Opt. Lett.*, 31, 1 (2006)
  20. A.Kaplan et al. "Efficient vacuum ultraviolet light frequency downconversion by thin films of CdSe quantum dots", *Appl. Phys. Lett.* 88, 24, 2006
  21. J.S.Robinson et al. "The generation of intense, transform-limited laser pulses with tunable duration from 6 to 30 fs in a differentially pumped hollow fibre", *Appl. Phys. B* 85, 525 (2006)
  22. H.Wabnitz et al. "Generation of attosecond pulses in molecular nitrogen" *Eur. Phys. J. D* 40 305 (2006)
  23. L.E.Chipperfield et al., "Tracking individual electron trajectories in a high harmonic spectrum", *Opt. Comm.*, 264, 494 (2006)
  24. S.Baker et al. "Probing proton dynamics in molecules on an attosecond time scale", *Science*, 312, 424 (2006)
  25. Y.Mairesse et al., "High-harmonics chirp and optimization of attosecond pulse trains", *Laser Physics*, 15, 863 (2005)
  26. K.Varjú et al. "Reconstruction of Attosecond Pulse Trains using an Adiabatic Phase Expansion", *Phys. Rev. Lett.* 95, 243901 (2005)
  27. C.Vozzi et al., "Controlling two-center interference in molecular high harmonic generation", *Phys. Rev. Lett.*, 95, 13902 (2005)
  28. S.Gundry et al. "Ultrashort-pulse modulation in adiabatically prepared Raman media", *Opt. Lett.*, 30, 180 (2005)
  29. R.Torres et al., "Dynamics of laser-induced molecular alignment in the impulsive and adiabatic regimes: A direct comparison", *Phys. Rev. A*, 72, 023420 (2005)
  30. E.Sali et al., "Behavior of high-order stimulated Raman scattering in a highly transient regime", *Phys. Rev. A*, 72, 013813 (2005)
  31. J.P.Marangos "Ultrafast science - Molecular structure in an instant", *Nature*, 435, 435 (2005)
  32. J.Yin et al., "Excited states of surfaces and nanostructures in the VUV", *Proc. of SPIE*, 5725 188 (2005)
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- iv. Book Chapters**
1. J.P. Marangos et al. "Probing molecular structure and dynamics by laser-driven electron recollisions." In *Strong Laser Field Physics* T. Brabec and H. Kapteyn eds, Springer-Verlag New York Inc.
- v. Patents**
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  3. J.W.G. Tisch et al. "A system for doing Attosecond Science"
  4. A.Kaplan et al. "Monitoring initial steps of fullerene formation from graphite by autocorrelation mapping method"
  5. T. Witting et al. "Two-dimensional spectral shearing interferometry for the complete characterisation of attosecond XUV pulses"
  6. T. Witting et al. "Reliable reconstruction of single attosecond pulses with simplified chronocyclic tomography using an arbitrarily shaped ultrashort streaking field"
  7. J.D. Steele-Davies et al. "Strong isotope effect in ionisation of molecular hydrogen exposed to intense laser fields"