SOFT X-RAY COHERENT CONTROL AT THE ATTOSECOND TIME SCALE

K. C. Prince

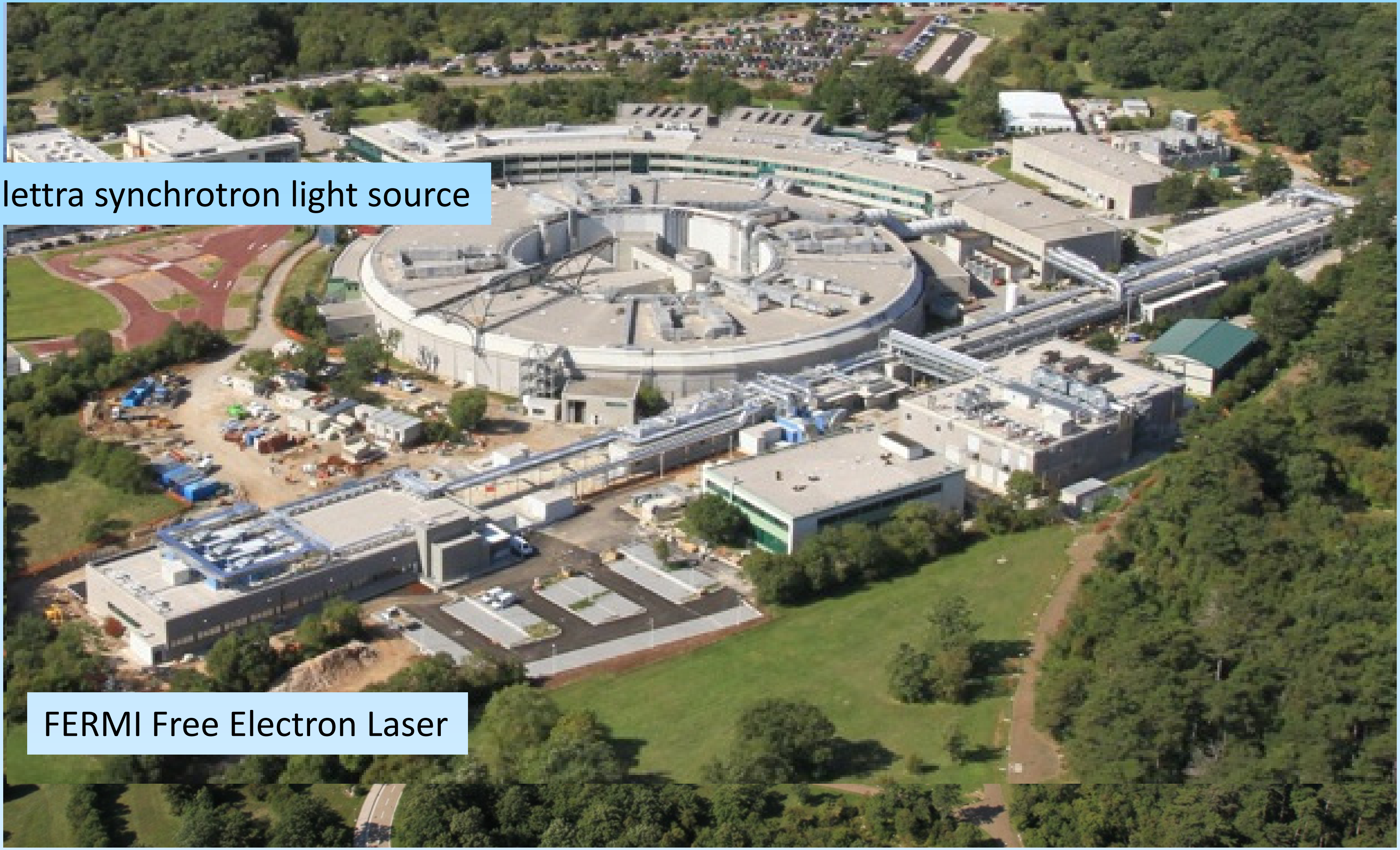
1. Introduction. Laboratory laser experiments
2. Background: Fundamental plus third/second harmonic.
3. How can we adapt this to the XUV spectral range?
4. Experimental set up.
5. Experimental results.
6. Prospects for further development.
7. Conclusions.

1. Introduction.

Elettra Sincrotrone Trieste

1. Introduction.

Elettra Sincrotrone Trieste



1. Introduction.

Elettra Sincrotrone Trieste

Pulsed optical lasers are characterised by transverse and longitudinal coherence, high intensity and ultrashort pulses.

Free Electron lasers (other than FERMI) are characterised by transverse coherence, high intensity and short pulses, but **low longitudinal coherence** for SASE machines.

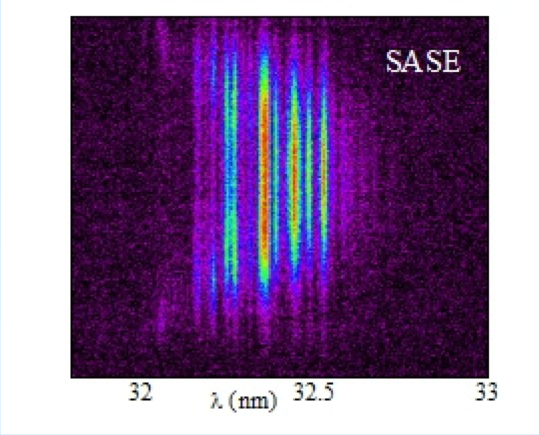
FERMI has longitudinal coherence: this is known from calculations, and from separate measurements of the pulse length and line width. The coherence of light coming from different undulators has been demonstrated (interference and polarization effects. Allaria et al, Phys. Rev. X **4** (2014) 041040.)

However there had been no direct demonstration of the longitudinal coherence to control an experiment, especially for multiple wavelengths.

1. Introduction.

Why is a seeded FEL better than a SASE FEL?

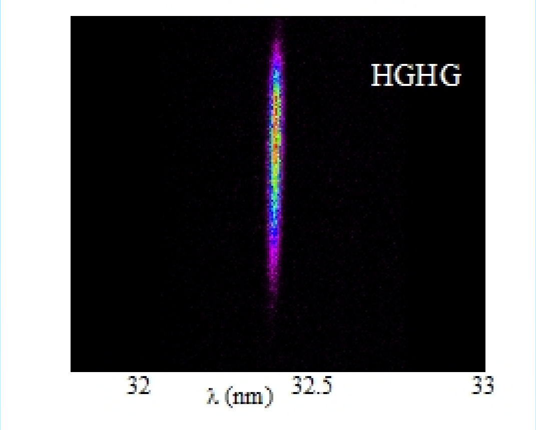
vertical angle



wavelength

FERMI SASE.

vertical angle



wavelength

FERMI seeded.

Narrow, stable bandwidth.
Nearly transform limited pulse.
Well-defined time structure: SASE pulses are spiky in wavelength and time.

1. Introduction. Laboratory laser experiments

Elettra Sincrotrone Trieste

What experiments can we do to (1) observe longitudinal coherence, and (2) control an experiment using this property?

What has been done with optical lasers?

"Bichromatic" or two colour experiments (not pump probe) have been done with optical lasers, starting from the 1990s. Brumer–Shapiro experiments.

First experiments combined the fundamental and second or third harmonic, with a controlled amplitude and phase relationship.

Other variants include different harmonics, pulses of light separated in time, but with a controlled **phase** relationship, and the whole field of coherent control.

2. Background: Fundamental plus third harmonic.

Elettra Sincrotrone Trieste

Two colours, first and third harmonic.
Chen, Yin, Elliott, PRL 64 (1990) 507.

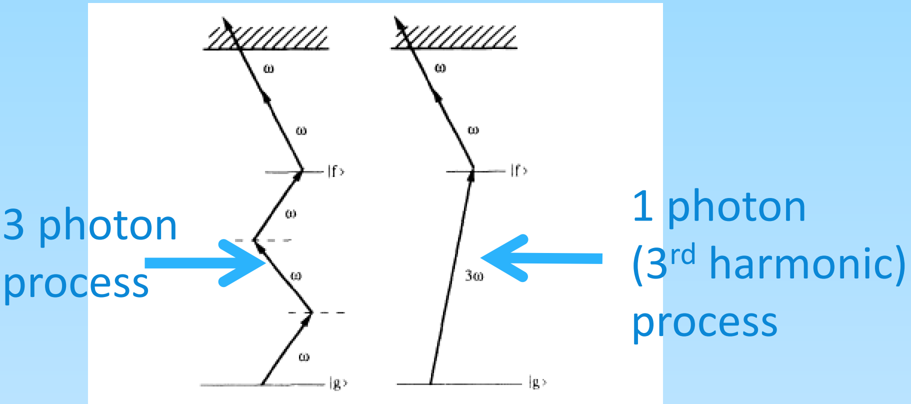


FIG. 1. The two processes which interfere in this observation. The transition $|g\rangle \rightarrow |f\rangle$ is three- and one-phonon allowed, as shown in (a) and (b).

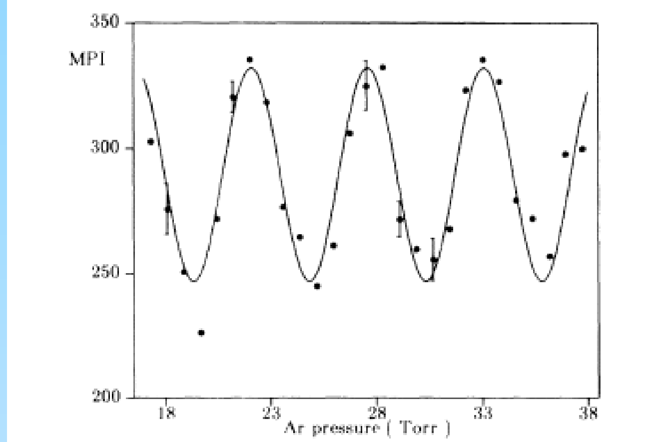


FIG. 3. Ionization signal measured as a function of argon pressure in chamber 2. Solid line indicates a best fit to the data. Error bars showing 1 standard deviation of the mean are shown for a few data points.

(Argon pressure tunes the phase.)
Total yield changes as a function of phase.

Can be understood as an interference effect:
Two paths to the same quantum state.
Thus the intensity depends on φ.
Also angular distribution depends on φ.

2. Background: Fundamental plus second harmonic.

Elettra Sincrotrone Trieste

First and second harmonic:
ionization of atomic rubidium.

Yin et al, PRL 69 (1992) 2353.

The figure shows the electron yield from detectors at different angles of emission.

In contrast to first + third harmonic, the total cross-section does not change.
Only the angular distribution changes.

0° detector

90° detector

180° detector

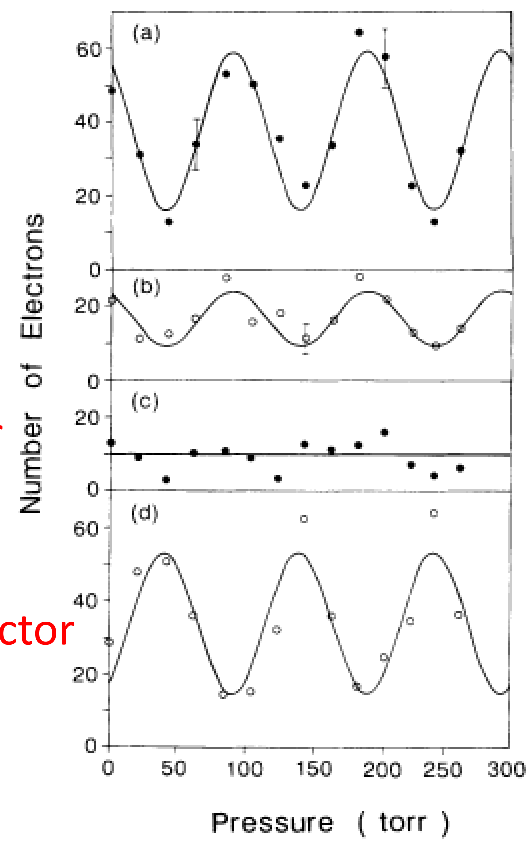
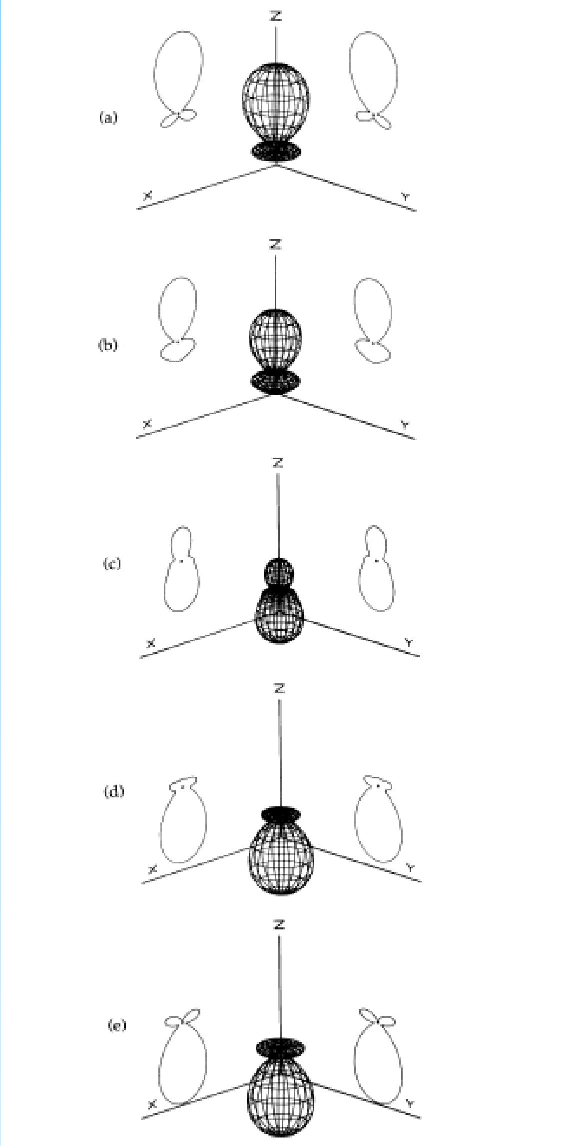


FIG. 3. Experimental data. The total electron count as a function of pressure of N_2 gas in the phase delay cell for the four detectors positioned at (a) 0°, (b) 45°, (c) 90°, and (d) 180°. The solid line is the result of a least-squares fit of a sinusoidally varying curve to the data.

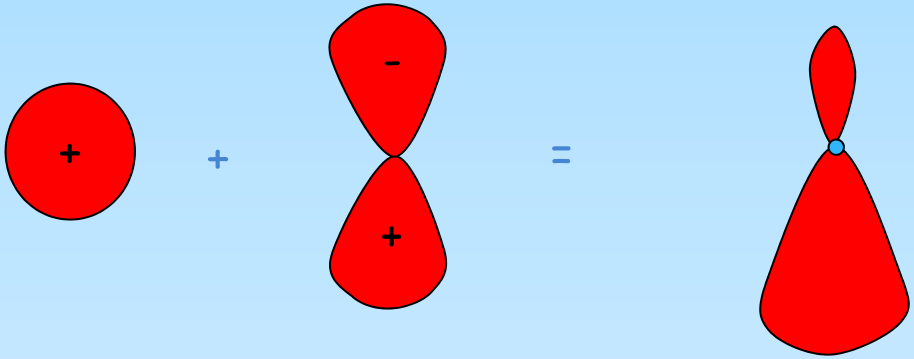
2. Background: Fundamental plus second harmonic.



Yin et al, PRL 69 (1992) 2353.

Calculated angular distributions.

Polarization is vertical (along z axis).



s wave p wave asymmetric wave

3. How can we adapt this to the XUV spectral range?

Elettra Sincrotrone Trieste

Differences with respect to optical lasers:

- ionization directly into the continuum, rather than via intermediate states (choose He or Ne so that the fundamental photon does not ionize)

- shorter wavelength => greater time/path length resolution is required

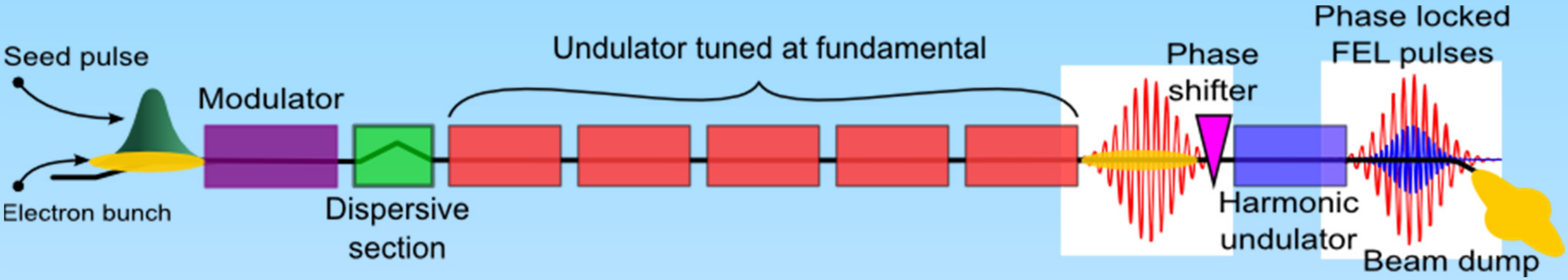
- many optical techniques do not work well, or do not work at all, e.g. mechanical delay lines at grazing incidence (210 as, Sorgenfrei et al. Rev. Sci. Instrum. 81 (2010) 043107), normal incidence beamsplitters (40 as, Tzallas et al, Nature 426 (2003) 268; 37 as, Midorikawa), phase tuning via gas cells. Requires special coatings, multilayers, filters, etc and so is limited.

How can we tune the phase?

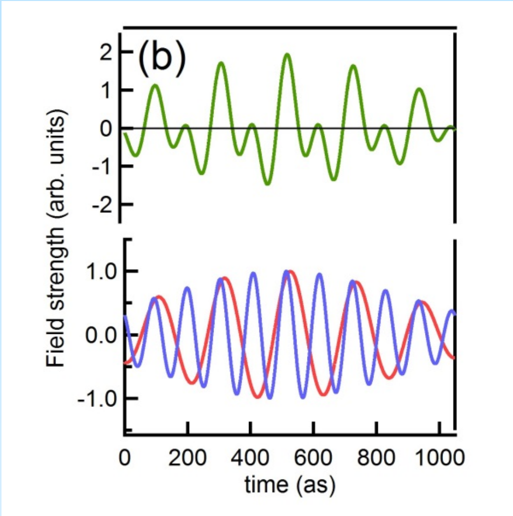
Best solution: give the problem to some-one else, i.e. FERMI machine people.

Phase control can be achieved by tuning 5 undulators to the first harmonic, and the last undulator to the higher harmonic, then using the ***electron*** **delay line** of FERMI. This provides a strong fundamental (for multi-photon excitation) and a weak harmonic.

4. Experimental set-up. The machine.



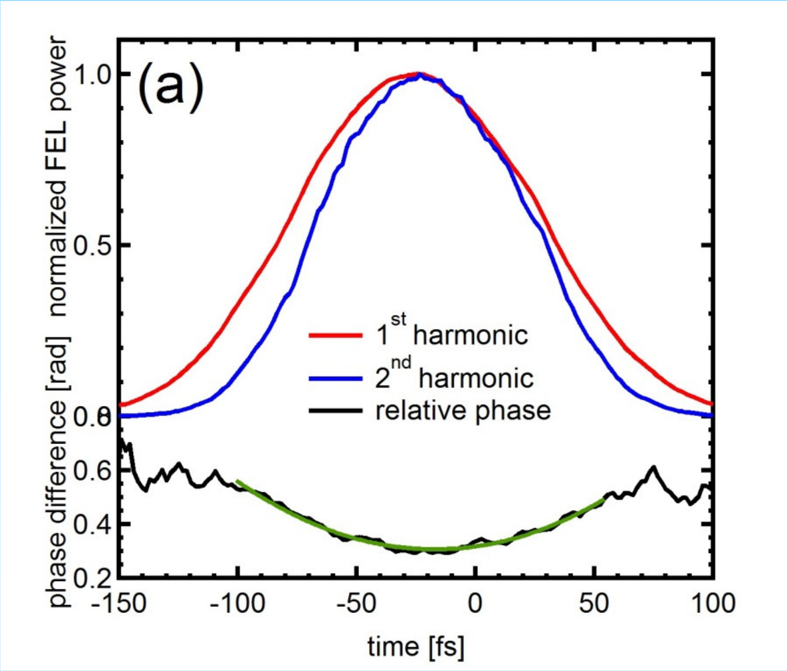
The phase of the first harmonic is locked
to the electron bunch phase.
The electron buch is delayed by the phase
shifter.
The phase of the second harmonic is locked
to the electron bunch, and so to the first
harmonic, with an adjustable phase.

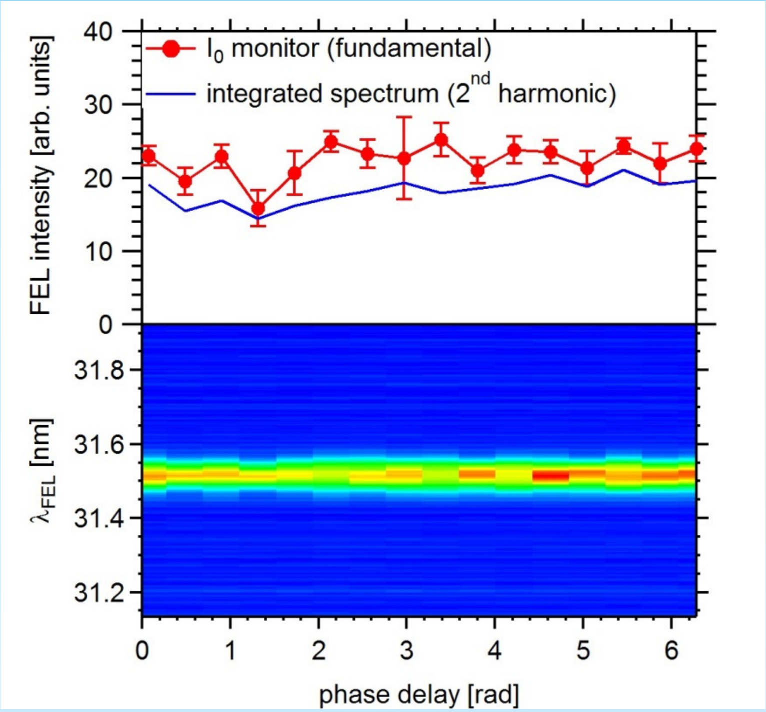


4. Experimental set-up. The machine.

Elettra Sincrotrone Trieste

First plus second harmonic, Ne target.





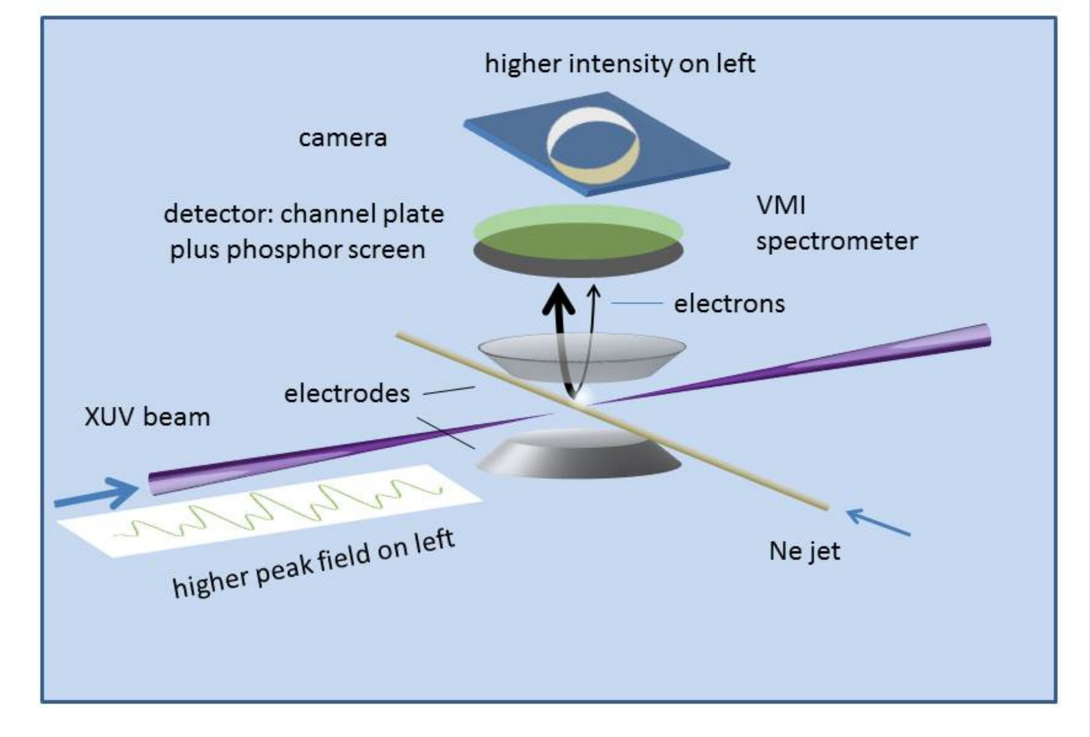
First harmonic: 63.0
second harmonic: 31.5 nm
temporal
evolution and phase relation.
Calculated phase error corresponds to a few attoseconds.

Check that any residual second harmonic from the first 5 undulators does not interfere with the 6th undulator harmonic.
Upper curves: intensity of 2nd harmonic
Lower panel: spectra.

4. Experimental set-up. The end-station.

Elettra Sincrotrone Trieste

VMI spectrometer.
Pulsed Ne jet.
Wavefront sensor behind chamber (not shown) for focusing diagnostics.

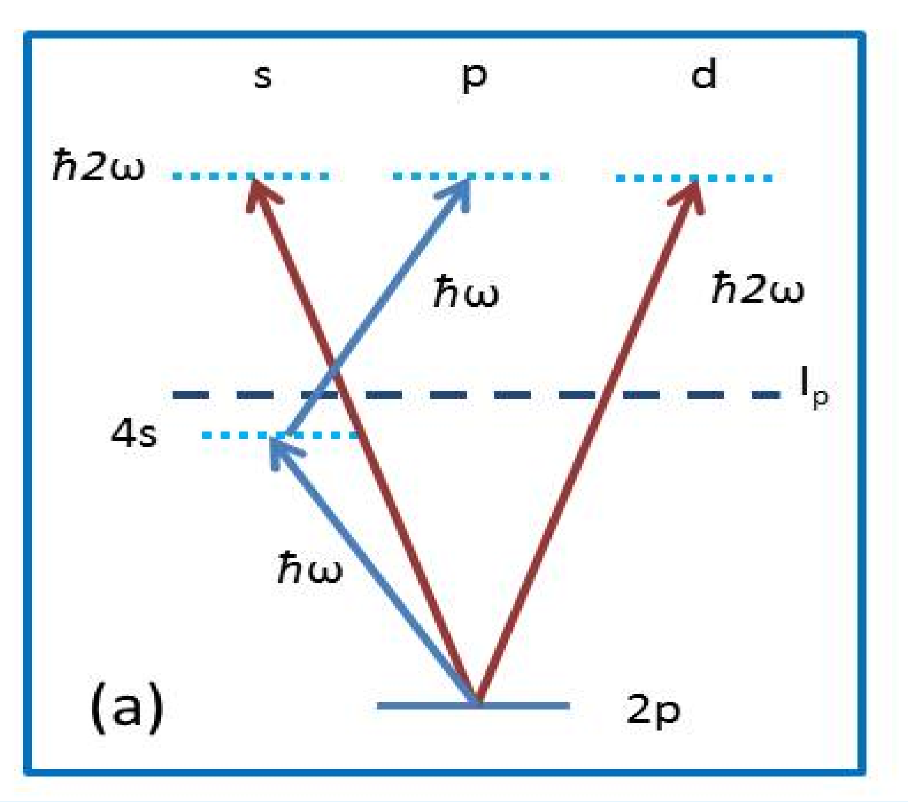


LDM end-station @ FERMI.
Lyamaev et al, J. Phys. B: At. Mol. Opt. Phys. 46 (2013) 164007.

5. Experimental results.

Elettra Sincrotrone Trieste

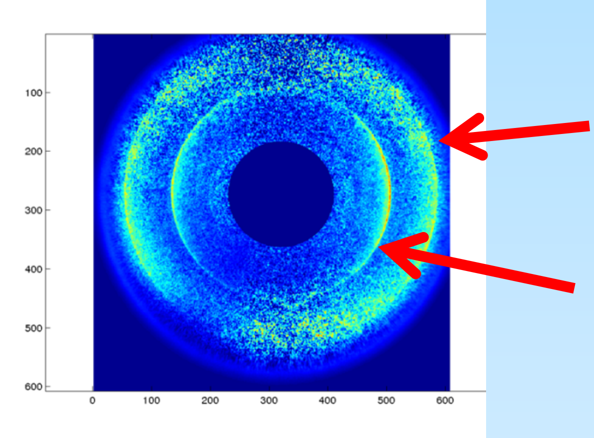
Scheme: two-photon, first harmonic PLUS one-photon, second harmonic ionization of Ne.



By choosing the Ne $2p^5 4s$ resonance, 62.97 nm, we aim to avoid outgoing *f* waves.
Second harmonic: 31.48 nm

5. Experimental results.

Step one, find the Ne resonance.
First 5 undulators scanned over resonance energy.
Undulator 6 open (to be used for second harmonic.)



VMI image at the wavelength of the Ne $2p^5 4s$ resonance.

Outer ring: a sharp line due to third harmonic radiation, plus a broad distribution due to fluorescence (the fluorescence excites photoelectrons which then impinge on the detector).

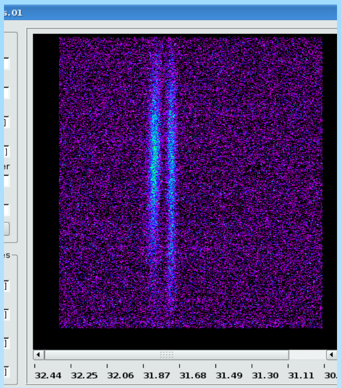
Inner sharp ring: electrons ionized by two photons (or second harmonic radiation as contamination).

The centre part of the image has been blanked.

Wavelength set with an accuracy of 0.04 nm – 10 meV.

5. Experimental results.

Having found the resonance
set 5 undulators for first harmonic, high power,
1 undulator for second harmonic, low power.



32.4 30.9
wavelength (nm)

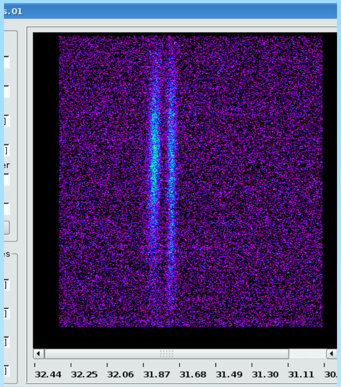
First harmonic has single stable line shape.
PADReS spectrometer image of second harmonic:
Split lines, ugly shape due to high power.

62.97/31.48 nm

5. Experimental results.

Elettra Sincrotrone Trieste

Having found the resonance
set 5 undulators for first harmonic, high power,
1 undulator for second harmonic, low power.

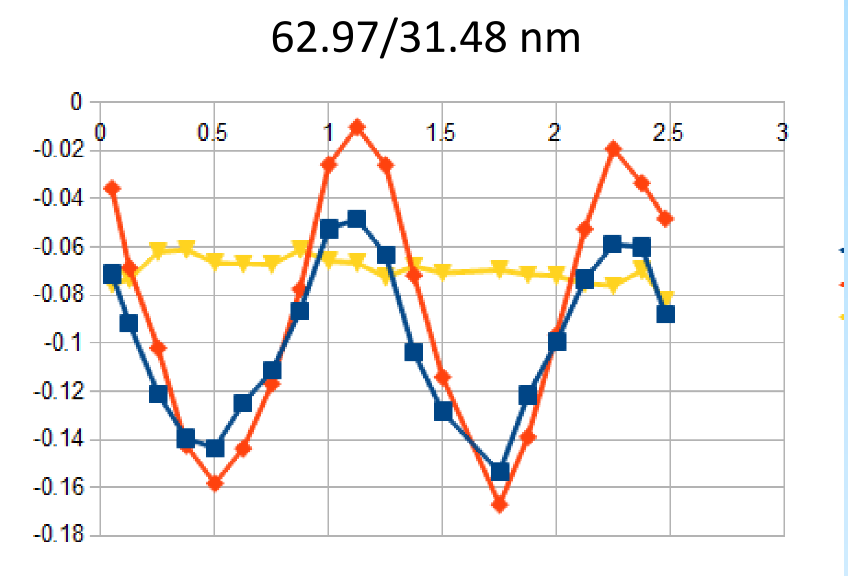


32.4 30.9
wavelength (nm)

Phase scan works!

First harmonic has single stable line shape.
PADReS spectrometer image of second harmonic:
Split lines, ugly shape due to high power.

$$A_{LR} = \frac{I_L - I_R}{I_L + I_R}$$



We verified temporal stability of phase control, change of phase on tuning around a resonance, etc.

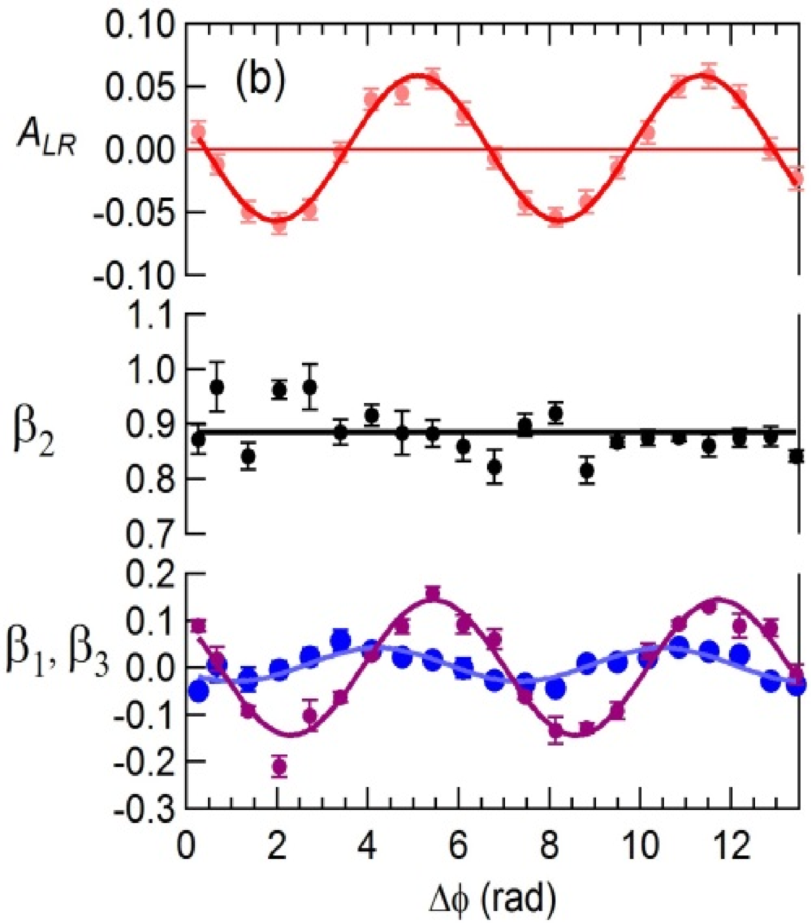
5. Experimental results.

The angular distributions were fitted with Legendre polynomials, each with a β_n parameter.

The even (β_2, β_4 etc.) and odd (β_1, β_3 etc.) parameters describe the symmetric and antisymmetric parts of the distribution respectively

5. Experimental results.

Elettra Sincrotrone Trieste



Periodic oscillation of β_1 (blue) and β_3 (magenta).

Phase offset between β_1 and β_3.

β_2 almost constant.

Data also taken at low power: single line in shot-by-shot photon spectra.
Lower statistics, higher contrast.

5. Experimental results cf. calculations for H atom (Alexei).

Elettra Sincrotrone Trieste

PHYSICAL REVIEW A 91, 063418 (2015)

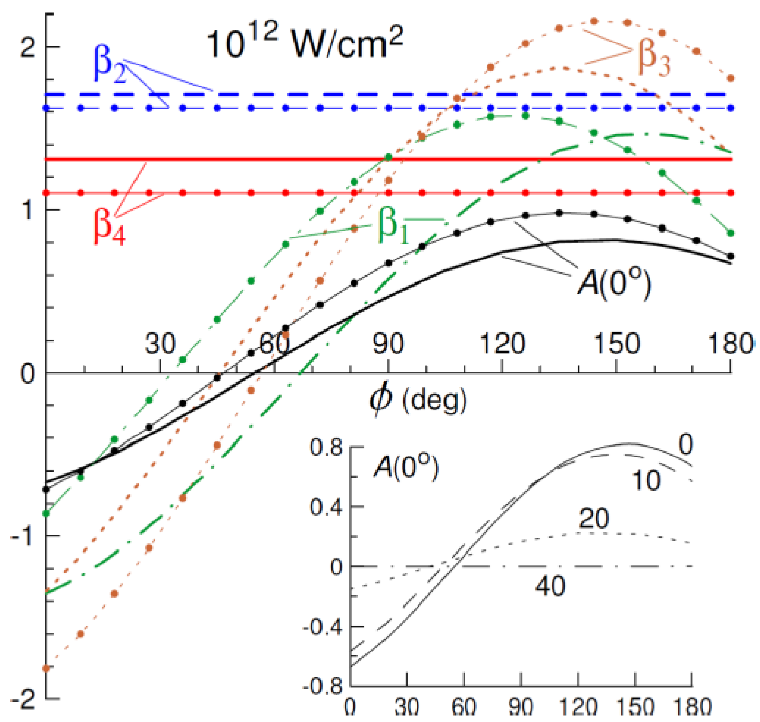


FIG. 5. (Color online) Asymmetry parameters β_k and asymmetry $A(0°)$ for the resonance photon energy of 0.375 a.u. and $\eta = 0.225$ as a function of the relative phase between the harmonics ($N = 40$, 10^{12} W/cm²). The results were calculated in PT (curves with dots) and by solving the TDSE (curves without dots). The parameters are averaged over the photoelectron line. The inset shows the asymmetry $A(0°)$ for different time delays τ between the fundamental and the second harmonic pulses, calculated by solving the TDSE. Value of τ measured in multiples of optical cycles is indicated for each curve.

The simpler hydrogen atom illustrates the main effects:

Periodic oscillation of β_1 and β_3

Phase offset between β_1 and β_3

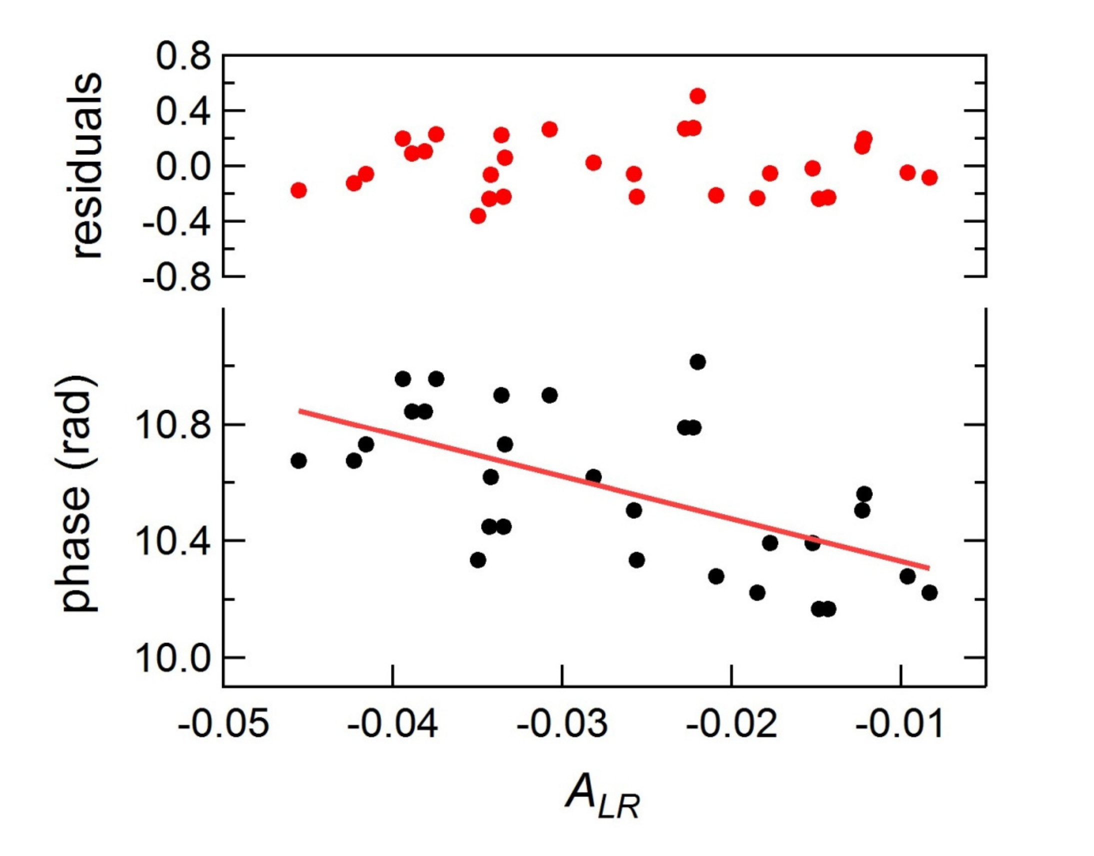
β_2 constant

Grum-Grzhimailo et al,
Phys. Rev. A 91 (2015) 063418.

Results: measurement of temporal resolution

Elettra Sincrotrone Trieste

Phase scan in steps of 0.9 as.
(900 zeptoseconds.)
Close to the maximum slope
of the oscillating curve.
Result: **rms deviation of 3.1 as.**



Bottom: asymmetry ratio, experimental data
Top: residuals after subtraction of straight line.

5. Experimental results.

Important properties of FERMI that we used:

narrow bandwidth,
stable wavelength,
high power in the first harmonic,
flexibility of undulator configuration,
longitudinal coherence of FERMI.

Surprises:
- The effect is stronger than we expected. Is it because the asymmetry parameter used excludes the incoherent part?
- It can work in spite of poor spectrum of the first harmonic at high power.

Enrico Allaria's calculations explain why: wavelength splitting occurs, but part of the light is still at the "right" wavelength.

6. Prospects for further development.

Elettra Sincrotrone Trieste

Continue experiments on fundamental plus harmonic. Beamtime approved for first plus third harmonic. Not only angular distribution, but also cross-section may change. Also, Fano parameter changes, if on resonance.

-> December 2015: experiment using first+third harmonic, helium resonance.
(We tried first plus third harmonic in December 2014, but there were technical issues.)

Can we measure fast phenomona, such as Wigner times?

The Wigner time is interpreted as the time between the absorption of a photon and the emission of an electron and is typically tens of attoseconds. It is equal to the energy derivative of the phase of the outgoing electron.

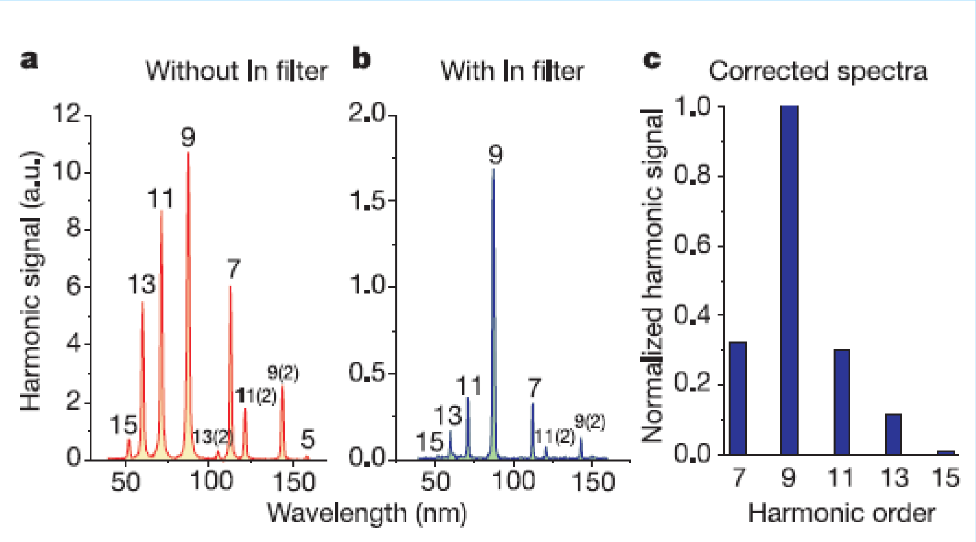
We are in discussions with theoreticians and machine physicists concerning a measurement.

6. Prospects for further development.

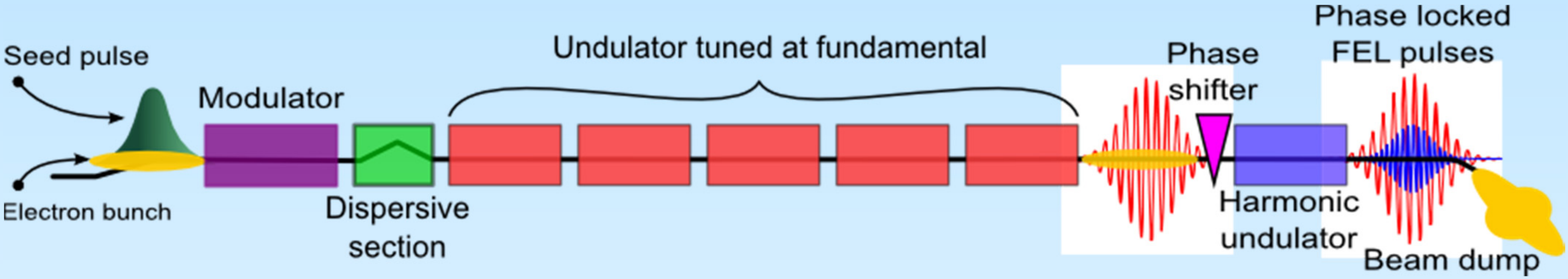
Elettra Sincrotrone Trieste

Pulse sculpting is possible.
e.g. Tzallas et al measured a train of pulses with width 780 as.

FERMI can produce multiple wavelengths



Tzallas et al, Nature 426 (2003) 427.



7. Conclusions

Elettra Sincrotrone Trieste

FERMI can produce two colour light that is phase coherent.

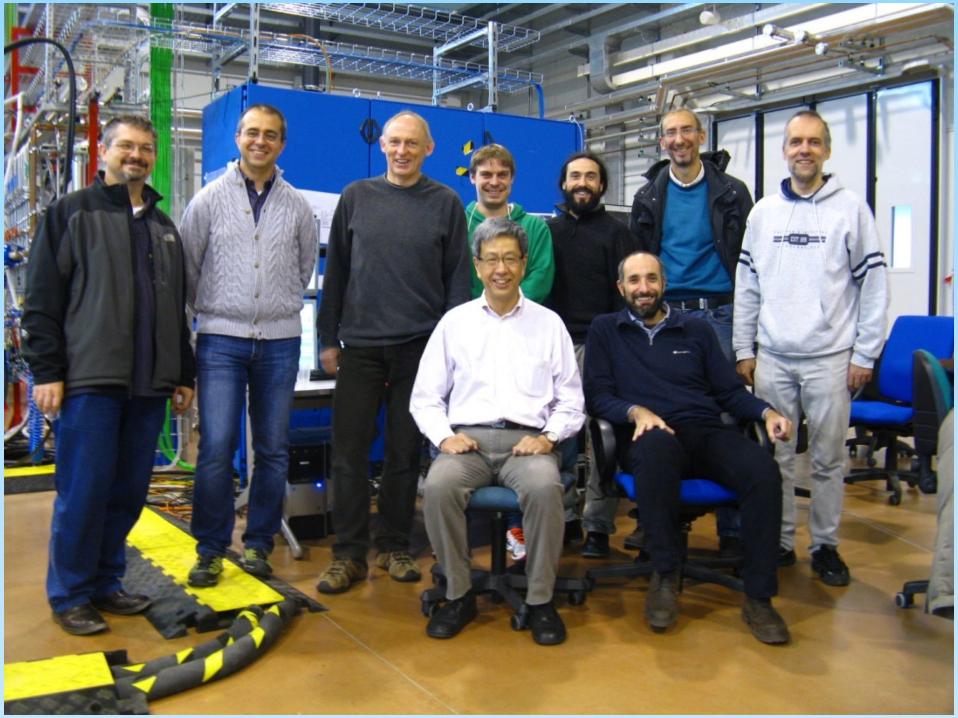
This development opens up new possibilities:

- manipulation of quantum systems, using multicolour pulses.

For example:

- first plus second harmonic experiments control the asymmetry and give information about the outgoing electron waves
- first plus third harmonic experiments change Angular Distribution, intensity, Fano parameter, etc.
- very high time (phase) resolution: 3 attoseconds, (or better in future)
- pulse sculpting is possible

This was only possible due to the extremely close collaboration between machine physicists, experimentalists and theoreticians.



Some people, not all...

7. Conclusions: participants

Elettra Sincrotrone Trieste

K. C. Prince, E. Allaria, C. Callegari, R. Cucini, G. De Ninno, S. Di Mitri, B. Diviacco, E. Ferrari, P. Finetti, D. Gauthier, L. Giannessi, N. Mahne, G. Penco, O. Plekan, L. Raimondi, P. Rebernik, E. Roussel, C. Svetina, M. Trovò, M. Zangrando, (Elettra-Sincrotrone Trieste)

G. Sansone, P. Carpeggiani, M. Negro, M. Reduzzi (Politecnico Milano)

K. Ueda, Yoshiaki Kumagai, T. Takanashi, D. Iablonskyi (IMRAM, Tohoku University)

M. Meyer, T. Mazza (Eurofel, Hamburg)

A. Grum-Grzhimailo, E.V. Gryzlova, S.I. Strakhova (Moscow State University)

K. Bartschat, N. Douguet, J. Venzke (Drake University, Iowa)

M. Coreno (ISM, CNR), F. Stienkemeier (Freiburg University), E. Ovcharenko (TU Berlin), A. Fischer, (Max Planck Institute, Heidelberg)

And lastly a word from our sponsor...

Elettra Sincrotrone Trieste

RESONANCE workshop: Multicolor FEL pulses and coherent control on the attosecond time scale opening new science perspectives
Trieste, Italy, December 10-11, 2015
http://www.elettra.eu/Conferences/2015/RESONANCE

Thank you for your attention.