

Lecture Notes: March 2003-- C.D. Lin  
Attosecond X-ray pulses issues:

1. Generation: Need short pulses (less than 7 fs) to generate HHG  
HHG in the frequency domain  
HHG in the time domain  
Issues of attosecond pulse trains and of single attosecond pulses
2. How to measure attosecond pulses? How to measure absolute phase?  
Measure photoelectrons in combining x-ray pulses with lasers
  - (a) The nature of electron spectra with respect to the x-ray temporal width
  - (b) measurements of electrons in a direction perpendicular to the laser polarization
  - (c) measuring the electrons in the laser polarization direction
3. What can be done with the attosecond pulses?

References:

1. RMP article: T. Brabec and F. Krausz, RMP72, 545 (2000) – good overview, went over the issues, but not the solutions
2. F096. Kienberger et al Science 297, 1144 (2002)—steering attosecond electron wave packets with light
3. F090. Drescher et al, Science 291, 1923 (2001)—first measurement of the x-ray pulse duration
4. Baltuska et al Nature 421, 611 (2003)—stabilized short laser pulses achieved, absolute phase can be fixed.

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## 1. Generation of x-ray pulses by HHG

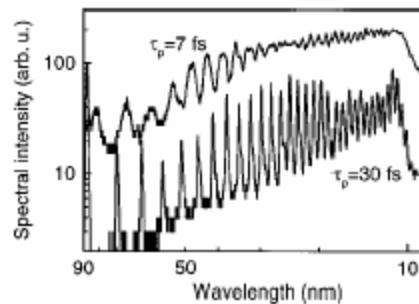


Fig. 1

Fig.35(RMP) **Short pulses are more efficient in producing HHG** since the atoms are less ionized. At intensity of  $3 \times 10^{14} \text{ W/cm}^2$ , only 1% of Ne is ionized for the 7fs pulse but 10% for the 30fs pulse. The peaks from the 7fs pulse is less pronounced since they are emitted from a few cycles so the interference is less complete.

As shown in the early lectures, the temporal profile of such a harmonic spectra is a pulse train, each pulse has a width of the order of  $T/N$ , and the pulse repeats at every  $T/2$ , where  $T$  is the optical cycle.

The HHG can be calculated approximately using the **Lewenstein model** and Chang has the code. The temporal profiles of HHG can be analyzed with the **wavelet theory** (Tong).

For a short pulse laser, the electric field is written as  $E(t) = E_L(t) \cos(\omega t + \mathbf{f})$  where  $\mathbf{f}$  is called the carrier envelope (C-E) phase, or the **absolute phase**. For few-cycle pulses, the absolute phase is important. However, the absolute phase from each laser pulse in general varies randomly. In combination with the optical frequency comb technique, only recently (Feb 2003) the group from Vienna have been able to stabilize the absolute phase. From their recent paper (Ref4), they have succeeded in producing stabilized few-cycle pulses.

**2. HHG by few-cycle pulses**

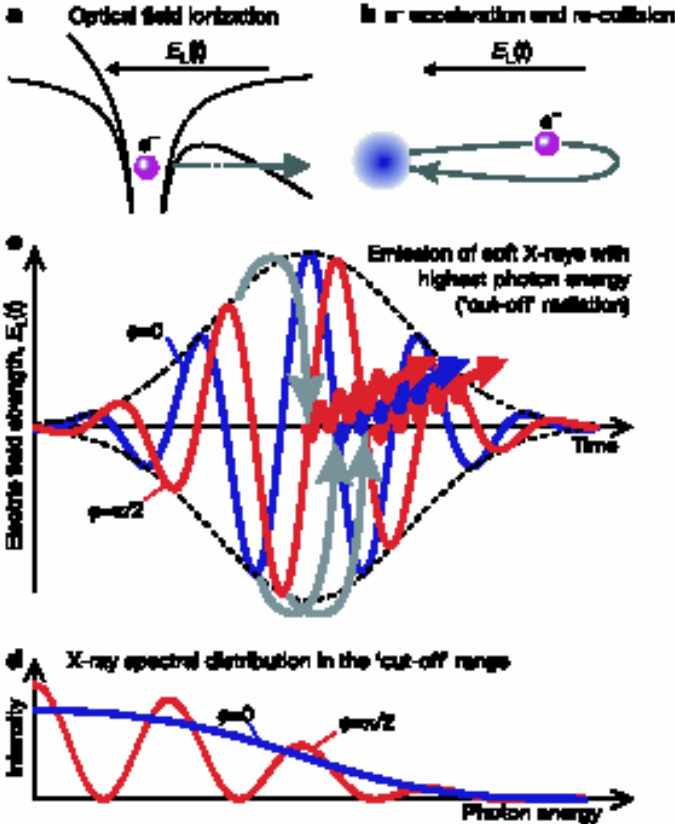


Fig. 2

Fig.2 illustrates that the HHG near the cutoff region is very sensitive to the absolute phase. For the cosine pulse ( $\mathbf{f}=0$ ) there is only one time that the electron can be ionized and return to rescatter with the ion to generate HHG in the cutoff region. For the sine pulse, there are two times that the electron can be ionized and return to form HHG in the cutoff region. The last frame indicates that for the cosine pulse the x-ray spectra is smooth while for the sine pulse, there is modulation from the two different ionization events. The spectra from actual numerical simulations confirm this qualitative interpretation. See Fig. 3 below.

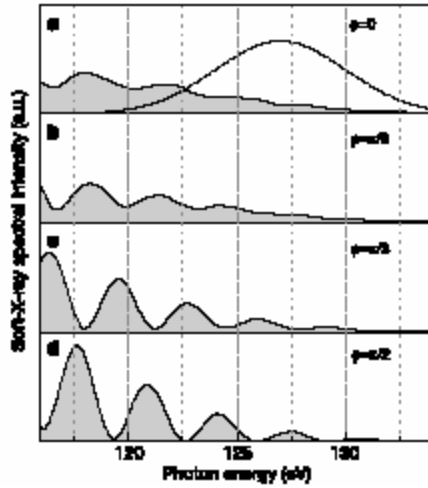


Fig. 3 HHG spectra near the cutoff from numerical simulations

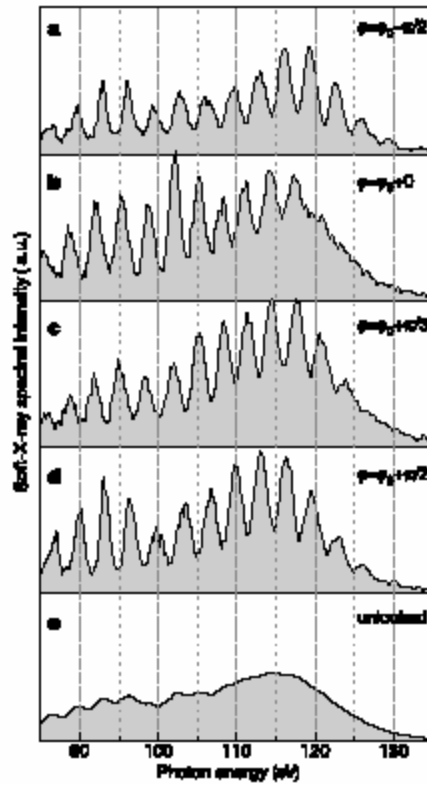


Fig. 4 experimental HHG from few-cycle pulses after the absolute phase is stabilized compared to the random phases at the bottom frame.

### 3. Generation of attosecond pulses in the x-ray region

A closer look at the HHG spectra (From F096). The absolute phase is not stabilized in this experiment. Fig. 5 show the photon energy spectra vs the laser intensity.

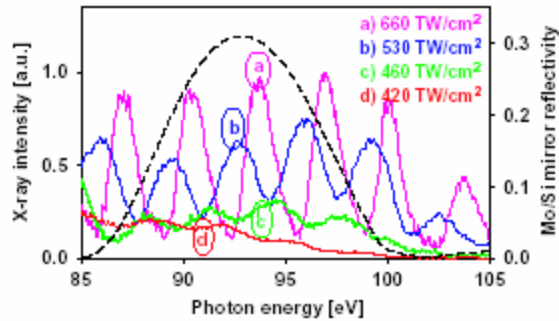


Fig. 5

From Fig. 3 of F096, the HHG near 90 eV from 7fs pulse on Ne at different laser intensity. At higher intensity the individual HHG peaks are clearly seen at odd harmonics. For lower intensity near the cutoff region, the spectrum is a smooth continuum. Using a broadband multilayer mirror to select the frequency domain allows the production of a single pulse attosecond x-ray.

#### 4. How to measure the attosecond pulses?

F096—steering attosecond electron wavepackets with laser light

**From the simulations:** If an x-ray is superimposed with a laser pulse at the instant where the laser's electric field is zero (at the maximum drift velocity), the photoelectron energy can be modified significantly. In the setup, the x-ray and the laser are polarized along the same direction and electrons are measured at a small cone in that direction.

**The pulse duration of the x-ray  $t_x$  vs.  $T_0$  of the laser is very important:**

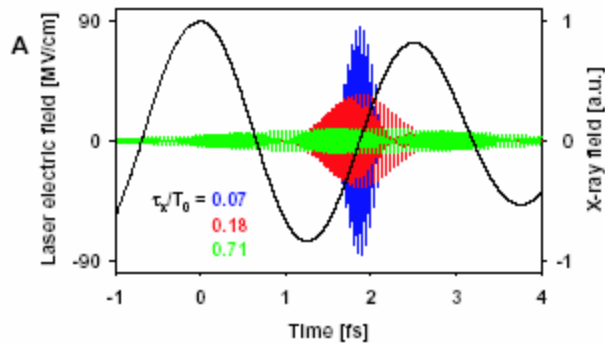


Fig. 6

This figure shows the overlap of x-rays vs. the 7fs laser pulse where the x-ray is of different duration. The simulated photoelectronspectra are shown in this figure 7.

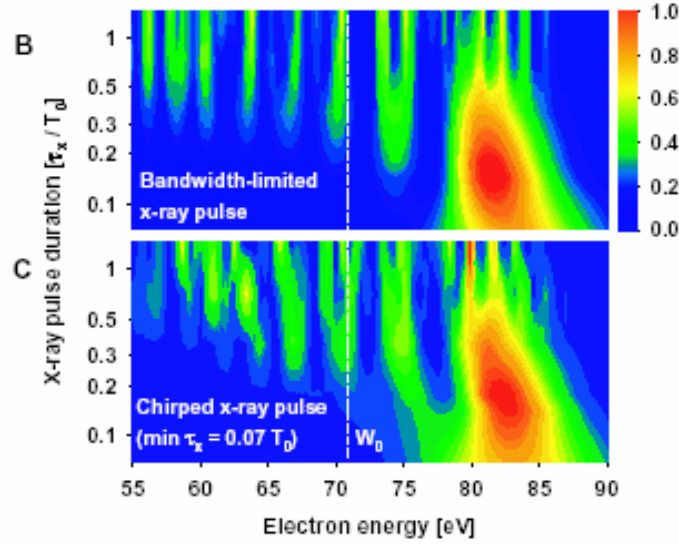


Fig.7

In this figure the dashed line is the mean peak position of the photoelectron if there is no laser present. For large  $\tau_x$ , we can see the formation of sidebands. For short pulses, the photoelectron is shifted significantly.

#### 4.1. Electron spectra(x-ray+laser) --simple classical theory

Let us try to understand these results from simulations qualitatively. For a long x-ray pulse, it is easier to think of the pulse in terms of photons with well defined energies. Without the lasers, the photoelectron energy is given by  $W_0$  as in the figure. The sidebands are due to the emission or absorption of additional laser photons.

If the x-ray is a short pulse. One can think that the photoelectron is created at a short time with initial kinetic energy  $W_0$ . If it is created at the instant where the laser electric field is zero, see Fig.6, the photoelectron will acquire a large drift velocity. Let the electron be created at time  $t_d$ , then the photoelectron energy is given by

$$W_f = W_0 - U_p(t_d) + U_p(t_d) \cos 2\omega t_d + 4U_p(t_d) \cos^2 \theta \sin^2 \omega t_d + \sqrt{8W_0 U_p(t_d)} \cos \theta \sin \omega t_d \quad (1)$$

where  $\theta$  is the angle of the photoelectron with respect to the laser polarization axis. In such experiments, one would like to avoid ionization from the lasers thus  $U_p \ll W_0$ . From Eq. (1), it is clear that for  $\theta=90$ , there is little shift of the photoelectron spectra. In other words, if the photoelectron was detected in the direction perpendicular to the laser polarization the spread due to the laser will be small. For the different  $t_d$ , the centroid is shifted by  $U_p(t_d)$ . This method was used in earlier experiments to determine the temporary profile of the attosecond pulses.

For example, in F090, x-ray of mean photon energy of 90 eV was used to examine the photoelectron spectra from Kr (4p) state. Fig. 8 below is the electron spectra measured in the perpendicular direction at different time delays. The measurement takes advantage of the fact that the angular distribution of Kr(4p) at around 90 eV is almost isotropic since the ionization is mainly from 4p  $\rightarrow$  es. Note that the polarizations of the x-ray and of the laser are in the same direction.

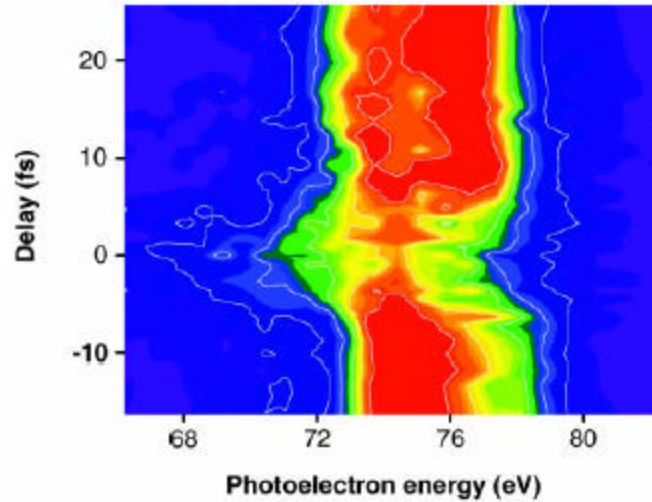


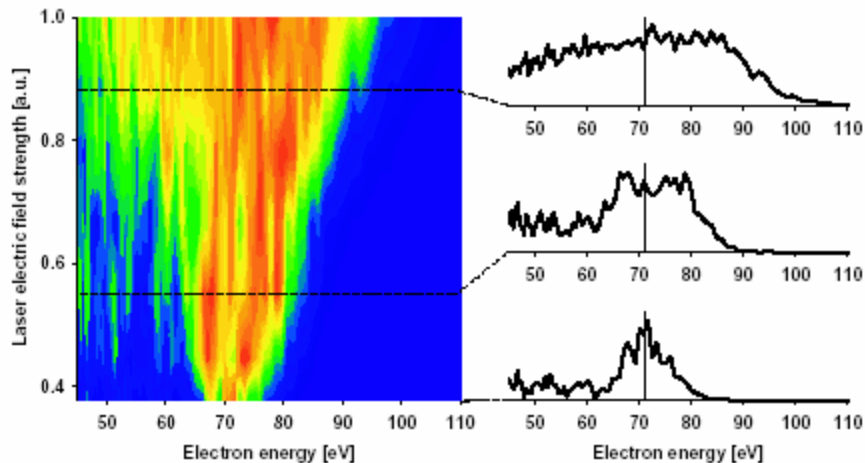
Fig.8

Since the x-ray has a finite pulse length, the analysis of Fig.8 from which to determine  $t_x$  is still not that straightforward. Eq. (1) is useful for the qualitative analysis but to extract the x-ray pulse width one needs to calculate the photoelectron spectra in the combined x-ray and laser fields. In F090, they extracted the x-ray pulse to have a duration of about 1.8fs.

The method used in F090 is not the best for attosecond pulses. **As shown from the simulation in Fig. 7, if the x-ray pulses are in the attosecond regime (in the limit that its duration is much less than the optical period), then the photoelectron spectra in the laser polarization direction is simple--mainly a shift in energy, with the shift proportional to the laser field strength (see eq.1).** The following data came from the work of F096.

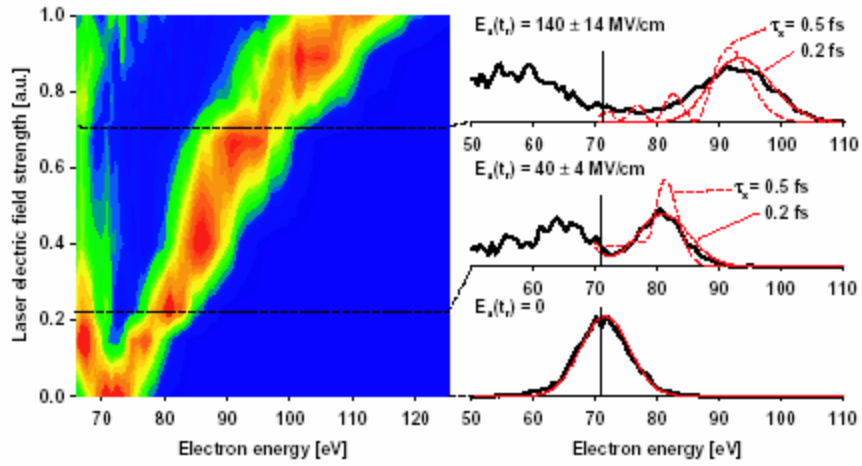
**From the experiment:**

- (a) *At higher intensities* of the input laser the HHG generated consists of discrete harmonics or temporarily **a series of pulse trains**. The observed photoelectron spectra look like this.  
(note: The laser field strength in the next two figures)



The laser intensity range for generating these spectra is 650-700 TW/cm<sup>2</sup>. Photons in the range of 85-100 eV were selected. Note the severe broadening at higher intensity. It is hard to extract information about the pulse trains from such measurements. For such pulse trains, the photoelectrons should be measured in the perpendicular direction. (This was first demonstrated in F091, see section 4)

(b) With *lower laser intensity* of 400-450 TW/cm<sup>2</sup>, the electron spectra look like this:



Using two models for the phases the analysis indicates that the x-ray pulse has width of 0.2 or 0.5 fs. Thus the authors have been able to demonstrate the HHG used in their experiment indeed is an attosecond pulse of duration of 0.2 or 0.5 fs.

(c) Measurement of pulses with durations longer than the optical cycles. (F091)--for the future