L.6: Attosecond pulse characterization

Pump probe methods are widely used to study the ultrafast processes in nature. To study the processes with ultrashort time scales, one needs ultrashort pump and probe pulses. With the advent of ultrashort laser technology, it is possible to produce the pump and probe pulses with duration as short as ~3 fs. It is well known that to generate shorter pulses, one needs larger spectral bandwidth ($\Delta t \sim 1/\Delta\lambda$) and due to this limitation, it is not possible to break the femtosecond (fs) barrier with IR laser pulses. In order to generate shorter duration pulses, one needs to generate coherent source with large bandwidth, and this can be easily achieved for shorter wavelengths. Generation of high order harmonics (HOH) of fs lasers in XUV region with its non-linear interaction with low density gases is a very promising source to generate sub-fs/attosecond (as) pulses. HOH of fs laser have the coherence properties of the laser beam and they are inherently phase locked, thus, overlapping of HOH generates attosecond pulses. The nonlinear interaction of fs laser pulse with gaseous medium generates only odd harmonics due to inversion symmetry in the medium. Overlap of these odd HOH generate attosecond pulse train separated by half of the laser cycle.

To measure the duration of attosecond pulses, one needs to measure the relative phase between different harmonic orders. This is achieved by the cross correlation between IR laser pulse and the HOH radiation. The technique used here is popularly known as RABITT (Reconstruction of Attosecond Beating By Interference of Two photon Transitions). Here, both HOH radiation and a part of harmonics generating fs IR pulse are focused in a low density gas sheath. Interaction of HOH with gas generates photo-electrons with energy

$$E_{ph\text{-}el} = qh\nu_L - IP \qquad (1)$$

where q is order of harmonic, ν_L is the frequency of laser, and IP is the ionization potential of the gas atom. A Magnetic Bottle Time of Flight (MBTOF) spectrograph is in-house developed and used to generate the electron spectrum (electron energy vs intensity) via electron counting. As the harmonic orders are separated by 2 orders, i.e., one will have $q, q+2, q+4, \ldots$ harmonics, and hence the electron spectrum will be separated by $2h\nu_L$. Interaction of q^{th} harmonic in the presence of laser photon generates the original photoelectron as well as side bands with energies $(q+1)h\nu_L\text{-}IP$, $(q\text{-}1)\ h\nu_L\text{-}IP$ corresponding to absorption of laser photon and emission of laser photon, respectively. Now, one can see that any side band can be generated from two channels, absorption of laser photon by lower harmonic and emission of laser photon by next harmonic order. If the field of laser photon, q-1th and q+1th harmonic (neglecting the relative amplitudes) can be written as:

$$\begin{aligned} E_L &= \exp[i\omega(t+\tau)] + CC \\ E_{q-1} &= \exp[i(q-1)\omega t + i\phi_{q-1}] + CC \\ E_{q+1} &= \exp[i(q+1)\omega t + i\phi_{q+1}] + CC \end{aligned} \qquad (2)$$

where, τ is the delay between laser and harmonic pulse and φ_q is phase of q^{th} harmonic. Then the field of side band can be written as

$$\begin{aligned} E_q &= \exp[iq\omega t + i\phi_{q-1} + i\omega\tau] + \exp[iq\omega t + i\phi_{q+1} - i\omega\tau] + CC \\ &= \exp(iq\omega t + i(\phi_{q+1} + \phi_{q-1})/2)(2\cos((\phi_{q+1} - \phi_{q-1})/2 + i\omega\tau)) \\ &\quad + CC \end{aligned} \qquad (3)$$

From above expression, we can see that the intensity of q^{th} side band oscillates with delay as $2\omega\tau$ and the peak position will give the relative phase between harmonic orders. Thus, if we can identify the peaks of all the side bands, we can measure the relative phases between all the harmonic orders and the attosecond pulse can be reconstructed as

$$E = \sum E_q \exp[iq\omega t + i\varphi_q] \qquad (4)$$

Figure L.6.1 shows the schematic of the experimental setup. Ti:sapphire laser (45 fs, 800 nm, 6 mJ, 1 kHz) was used to generate the HOH radiation. First this pulse is divided in two parts, first part is used to generate HOH radiation and second part was used for the cross correlation. These two beams are combined by a mirror (mirror with a hole at the centre, HM), then they are matched in space and time and pass through the gas cell. Generated harmonics are then focused in gas sheath using toroidal mirror (TM) and generated photoelectron signal is detected by MBTOF spectrograph. The delay between HOH and laser is changed by a piezoelectric transducer with a resolution of 10 nm (~66 attosecond). Corresponding photoelectron spectrum with side bands is shown in Figure L.6.2. If all the harmonics are in same phase then the line joining the peaks of the side bands should be vertical, but it is little tilted as shown in Figure L.6.2. This indicates the chirp in the harmonic spectrum. From this phase measurement, the duration of the one attosecond pulse is calculated between 300-350 attosecond using Eq. (4).

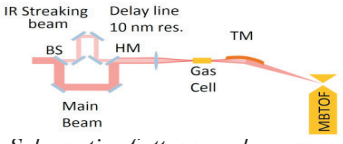

Fig. L.6. 1: Schematic of attosecond measurement setup.

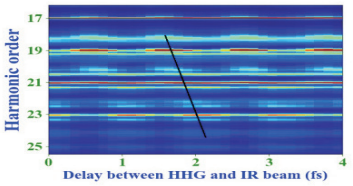

Fig. L.6.2: Intensity modulation of harmonics and side bands with delay between IR and HOH pulse.

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