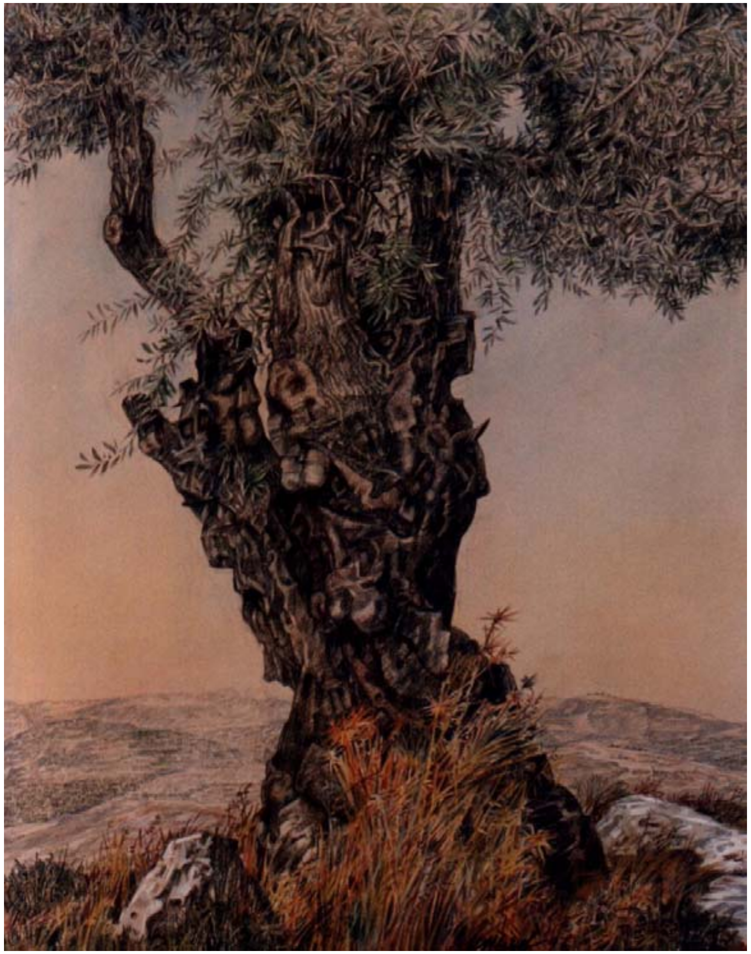
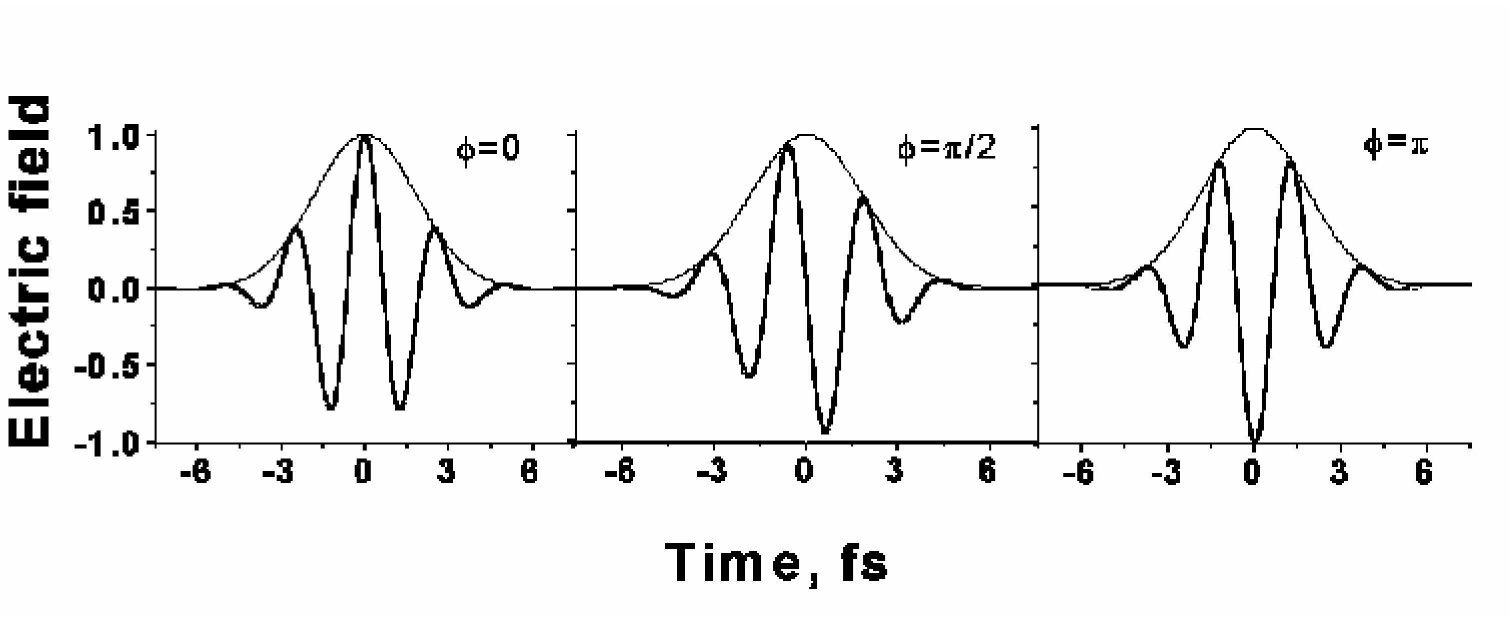
Attosecond measurements without attosecond pulses



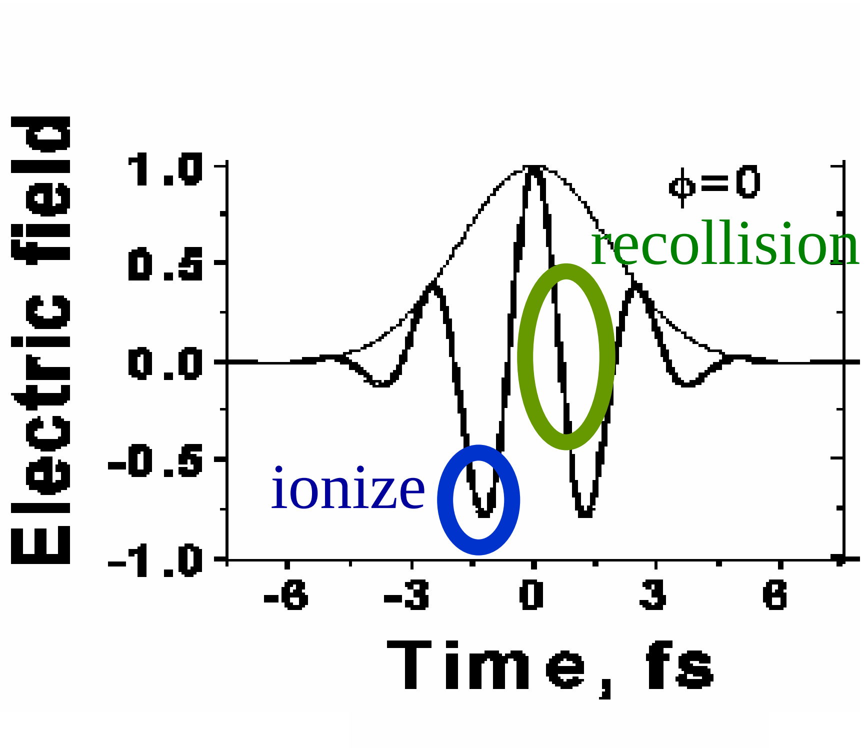
Phase-stabilized pulses

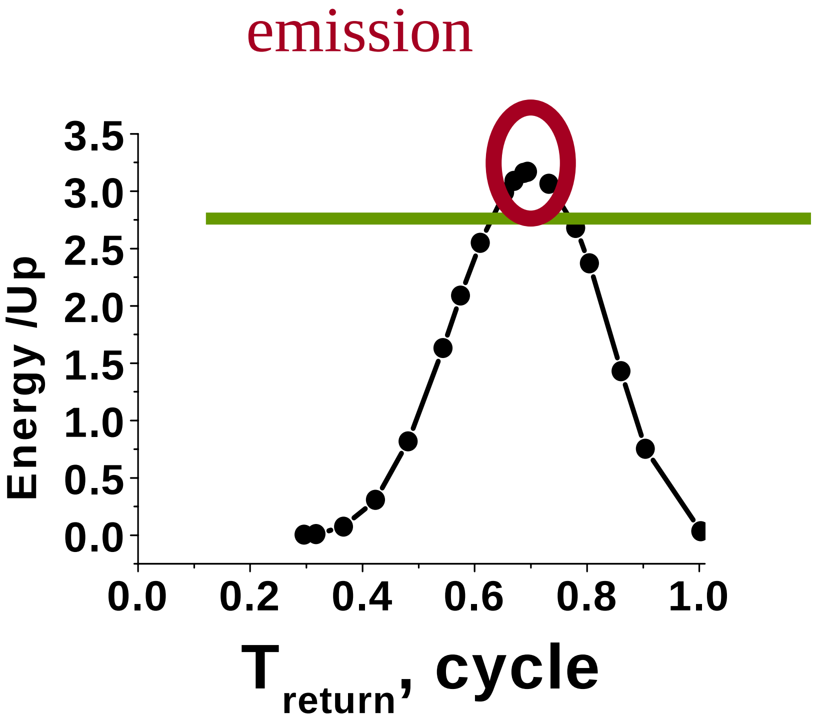


$\Delta\phi = 3^{o}$ per 10^{7} shots
$\Delta I = 3\%$ per 10^{7} shots
Shot-to-shot stability is few asec

This constitutes a built-in temporal ruler of incredible precision. We are trying to find a way to use it

Making asec XUV pulses





Temporal stability of asec XUV pulses is much better than its duration: 50-100 asec stability

In principle, we have better time resolution than the XUV pulse duration. Can we use it? How, when, what for?

Examples for attosecond spectroscopy

Two-electron dynamics induced by an XUV photon

Atom + 1 XUV = Atom $^{2+}$ + 2e^{-}
Removal of the first electron triggers the second

Processes:
- Auger,
- Coster-Kronig,
- Shake-off, etc

Effects:
- non-exponential decay due to non-flat continuum ,
- Interaction of many autoionizing states,
- Core rearrangements
- Zeno and Anti-Zeno stages of decay.

A la Carte

- **Attosecond metrology for 1 electron:**
  - **FROG**
  - **Asec streak camera**

  - **SPIDER**
  - **SPIDER-like**

    streak-camera for long XUV

- **Down to 0.01 fsec time resolution:**

  Correlated Atto-Second

  Two-electron Optical

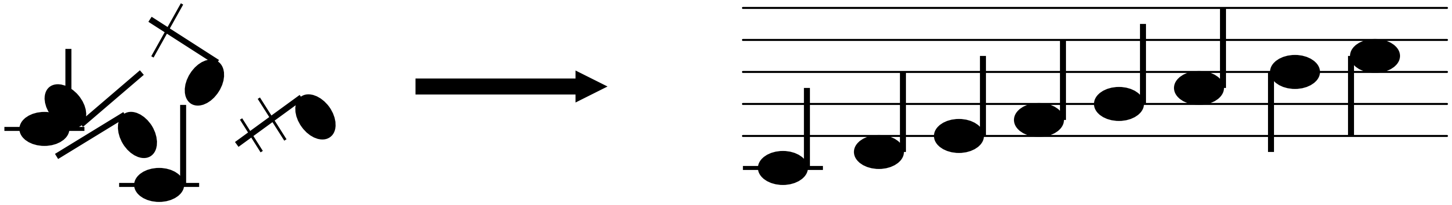
  Reconstruction

CASTOR

FROG

(Frequency-Resolved Optical Gating)

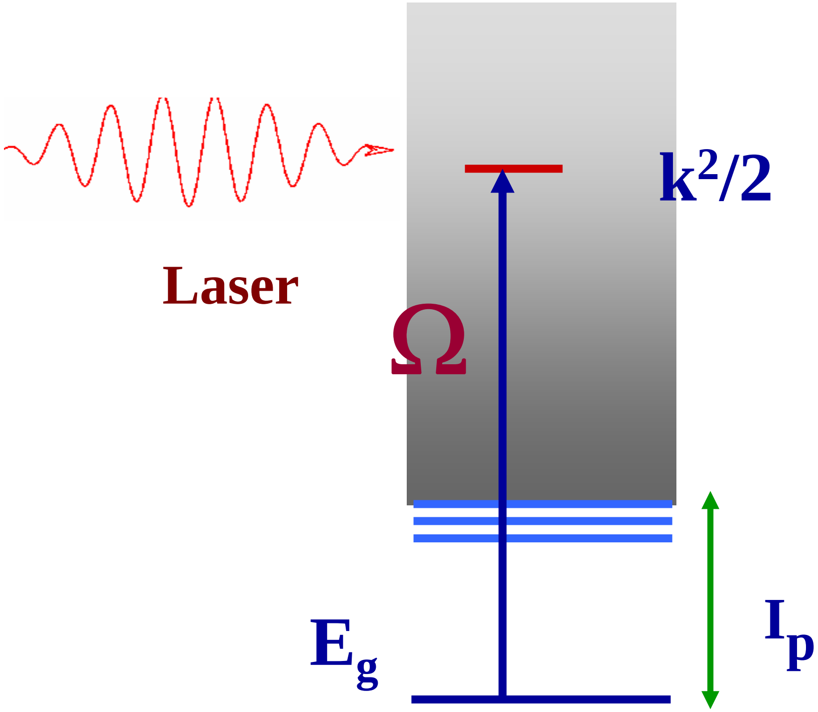
$$S(\omega,\tau) = \left| \int E(t)G(t-\tau)e^{-i\omega t}dt \right|^2$$



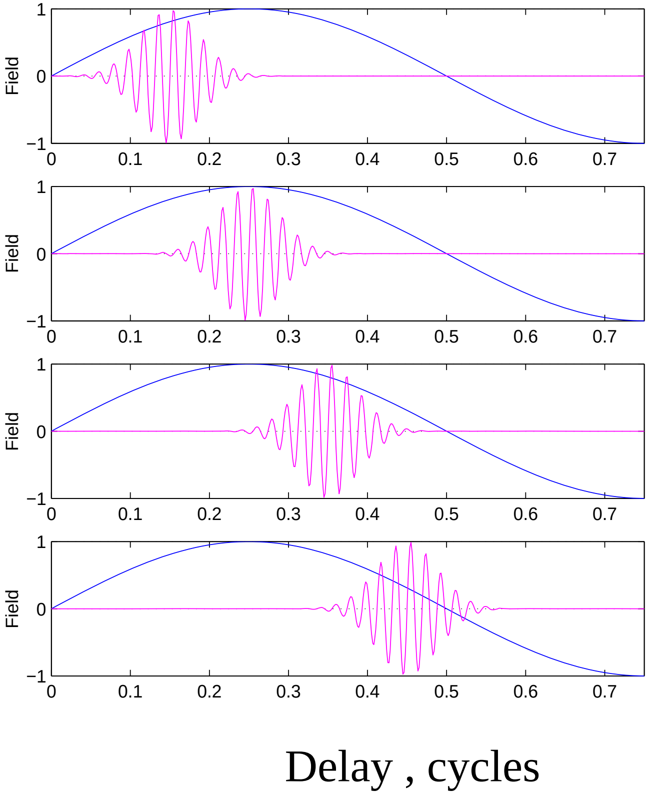
Spectrum

Spectrogram

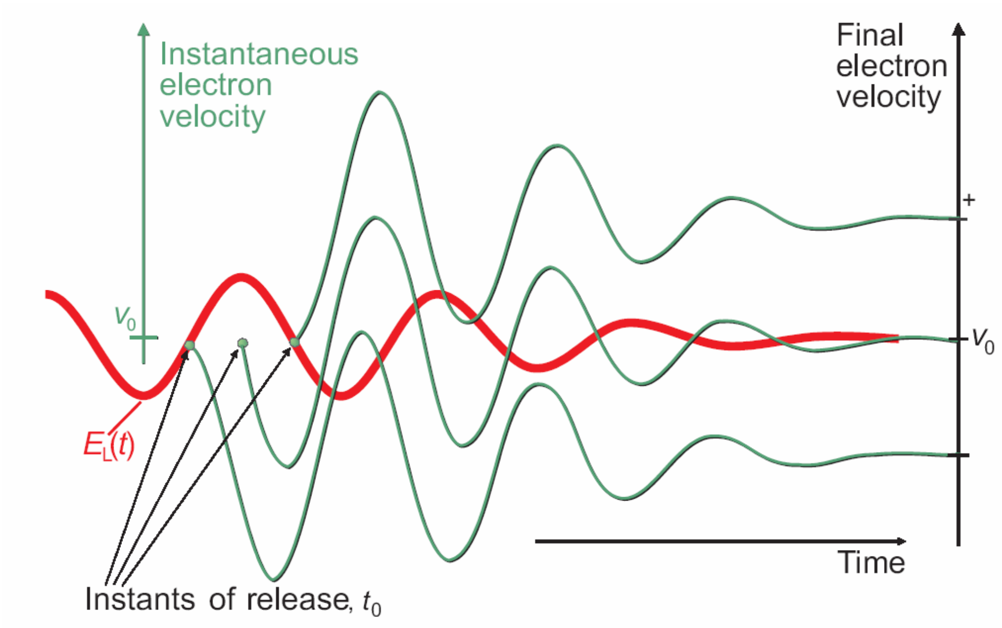
Attosecond streak camera for one electron



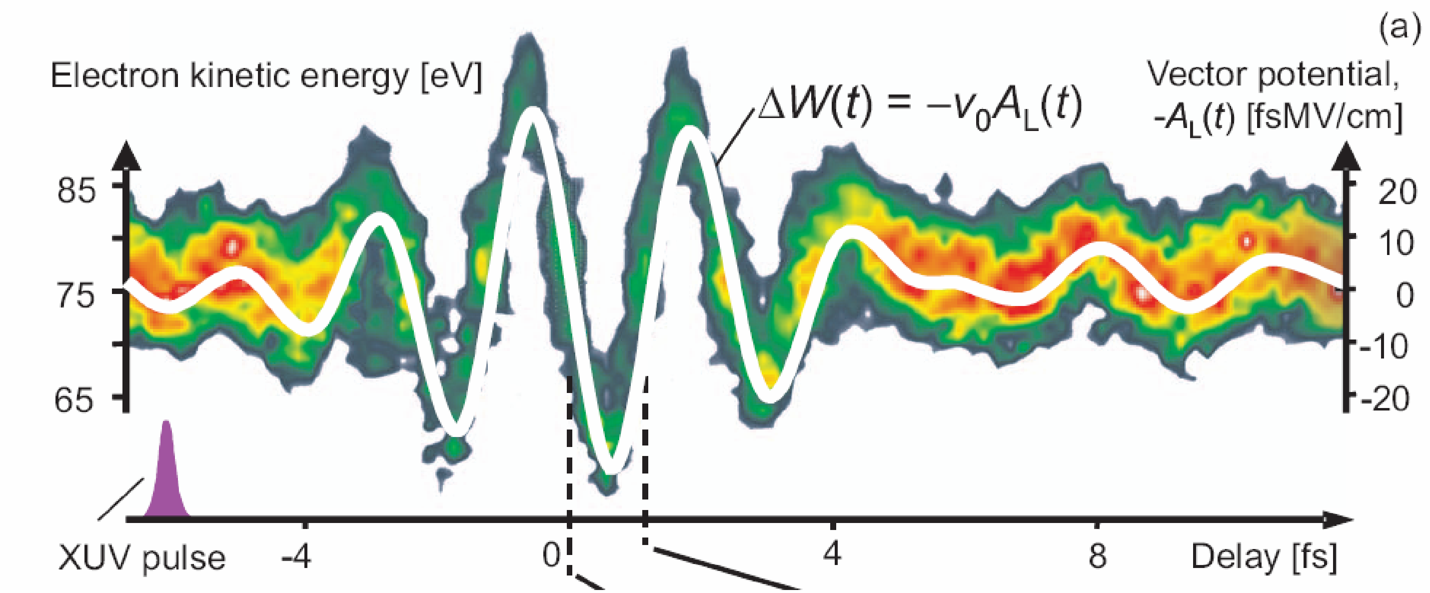
$k_f = k + A(t_0)$



Attosecond streak camera for one electron

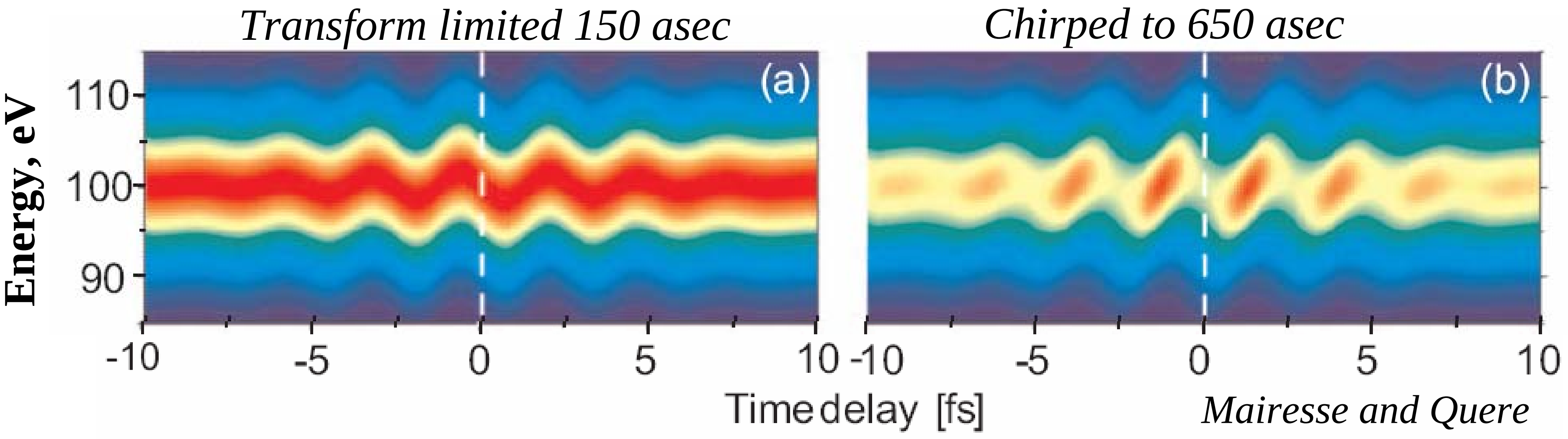


$$k_f = k + A(t_0)$$



Goulemakis et al

Attosecond streak camera is a FROG



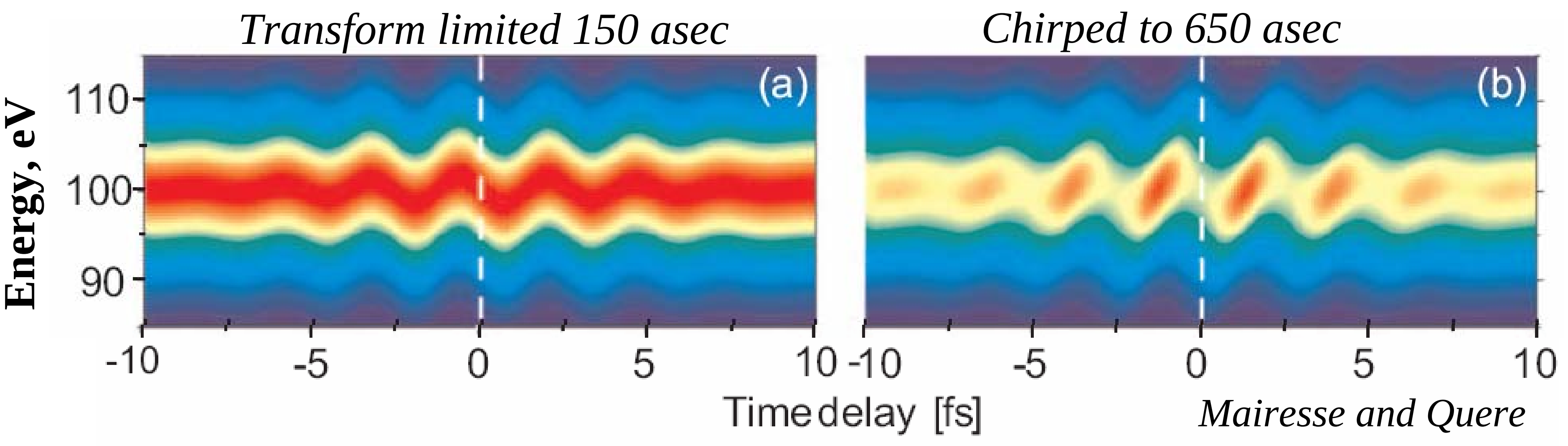
Transform limited 150 asec

Chirped to 650 asec

Mairesse and Quere

$$\sigma_A(v,\tau) = |a_A(v,\tau)|^2 = \left| \int_{-\infty}^{\infty} \underbrace{\boxed{\chi(v,t)}}_{\text{XUV-absorption amplitude}} \underbrace{\boxed{G(t-\tau)}}_{\text{Effect of the IR}} e^{i(v^2/2)t} dt \right|^2,$$

Attosecond streak camera is a FROG


Transform limited 150 asec

Chirped to 650 asec

Mairesse and Quere

$$\sigma_A(\upsilon,\tau) = |a_A(\upsilon,\tau)|^2 = \left| \int_{-\infty}^{\infty} \chi(\upsilon,t) G(t-\tau) e^{i(\upsilon^2/2)t} dt \right|^2,$$

$$G(t-\tau) = e^{i\Theta(t-\tau)}.$$

$$\Theta(t) = -\upsilon \int_t^{\infty} A_L(t')dt' - \frac{1}{2}\int_t^{\infty} A_L^2(t')dt'$$

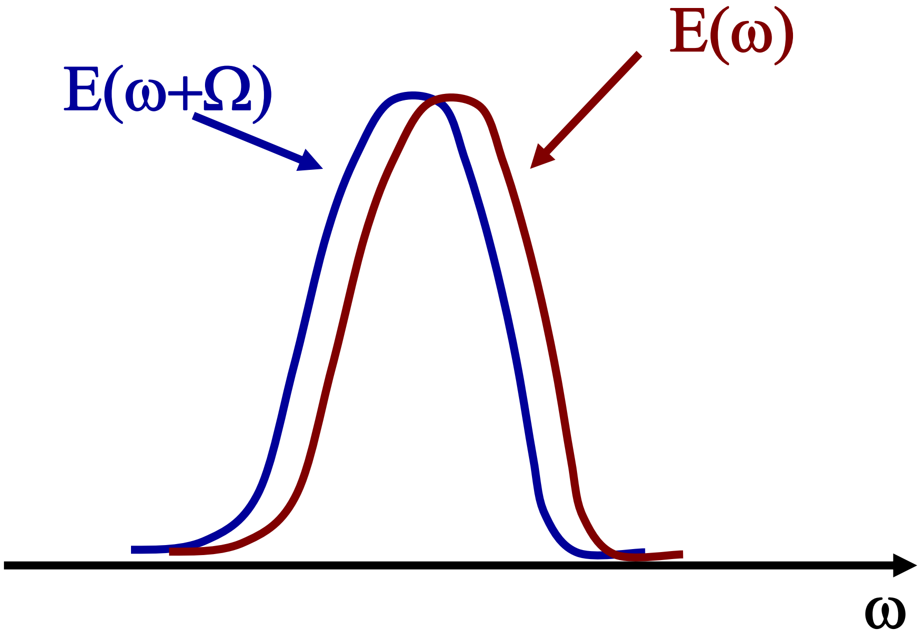
SPIDER

SPectral shearing Interfereometry for Direct Electric field Reconstruction

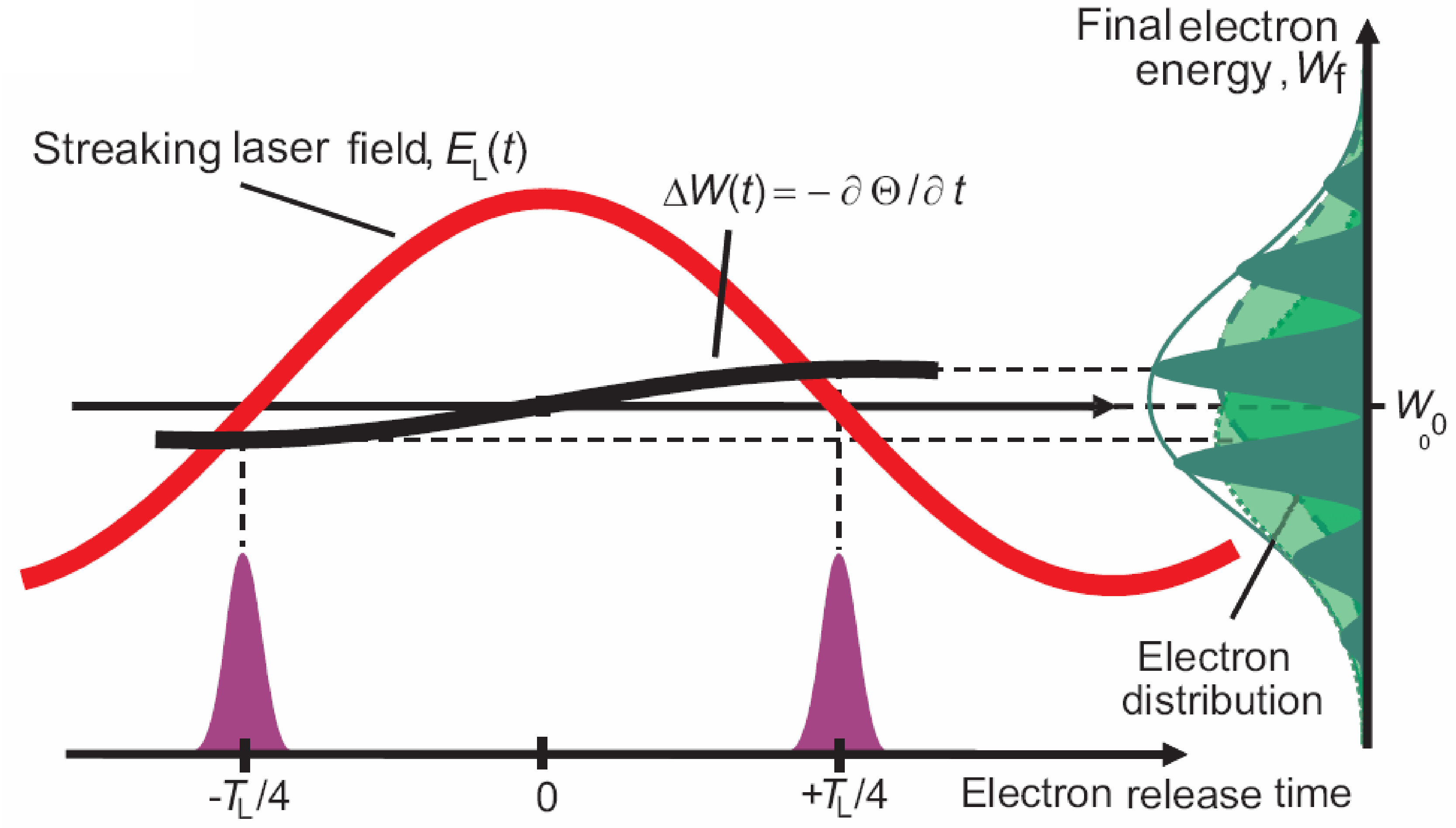
$$E(\omega) = |E(\omega)| \quad e^{i\varphi(\omega)}$$

$$S(\omega,\tau) = \left|E(\omega) + e^{i\omega\tau}E(\omega+\Omega)\right|^2 =$$

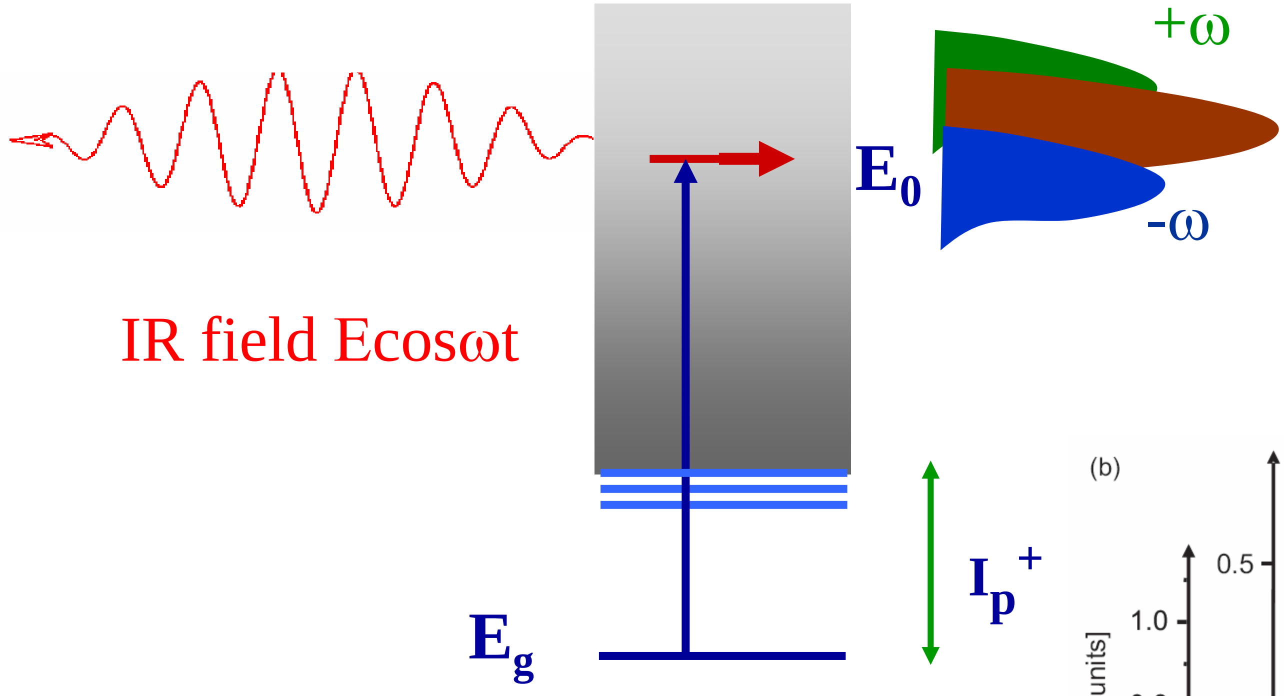
$$= \left|E(\omega)\right|^2 + \left|E(\omega+\Omega)\right|^2 + \left|E(\omega)\right|\left|E(\omega+\Omega)\right|\cos\left(\omega\tau + \underbrace{\varphi(\omega+\Omega) - \varphi(\omega)}_{\theta(\omega)}\right)$$

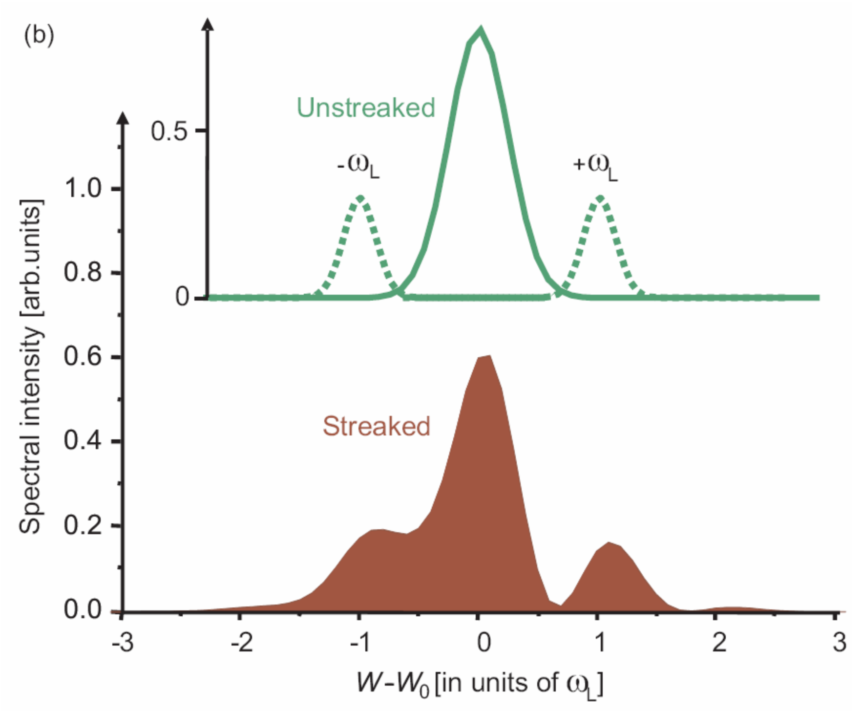


Attosecond SPIDER



Streak-Camera for LONG XUV: *SPIDER-like*





A la Carte

- Attosecond metrology for 1 electron:
  - FROG
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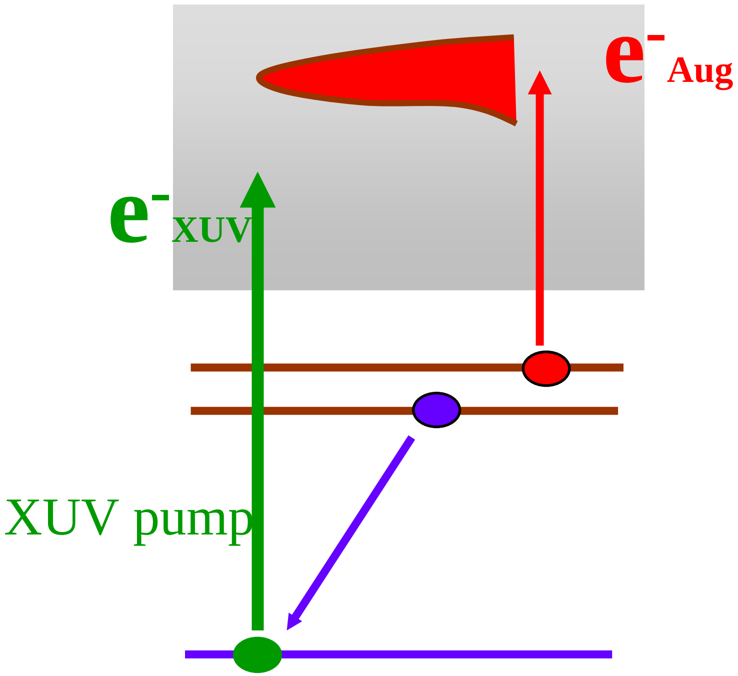
    streak-camera for long XUV

CASTOR

- **Down to 0.01 fsec time resolution:**

  Correlated Atto-Second
  Two-electron Optical
  Reconstruction

Photo-induced Auger decay



“Conventional” pump-probe approach

Short pump – sets t=0 by removing the green electron

Attosecond streak-camera measures the red electron

M. Drescher et al, Nature

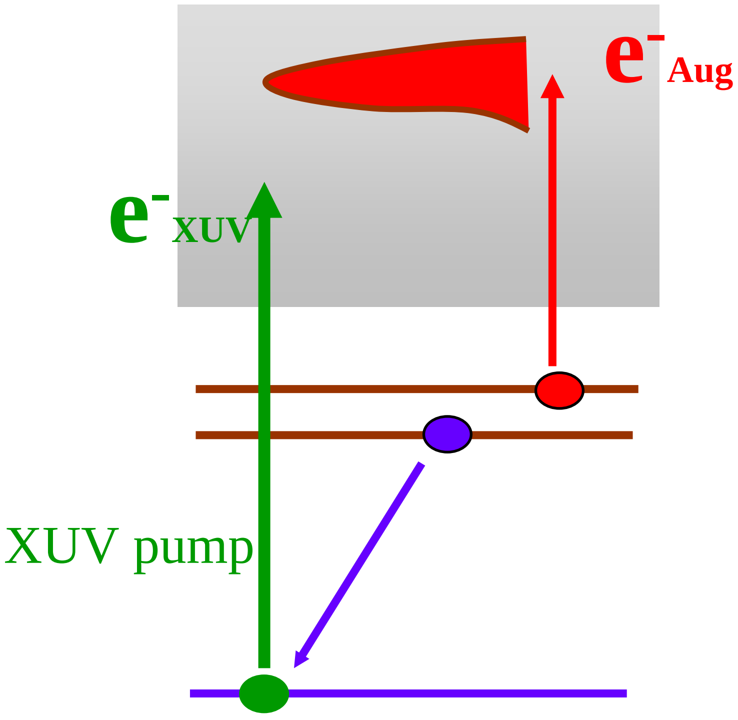
Conventional approach: Time resolution is limited by pump duration and by time resolution of the attosecond streak-camera

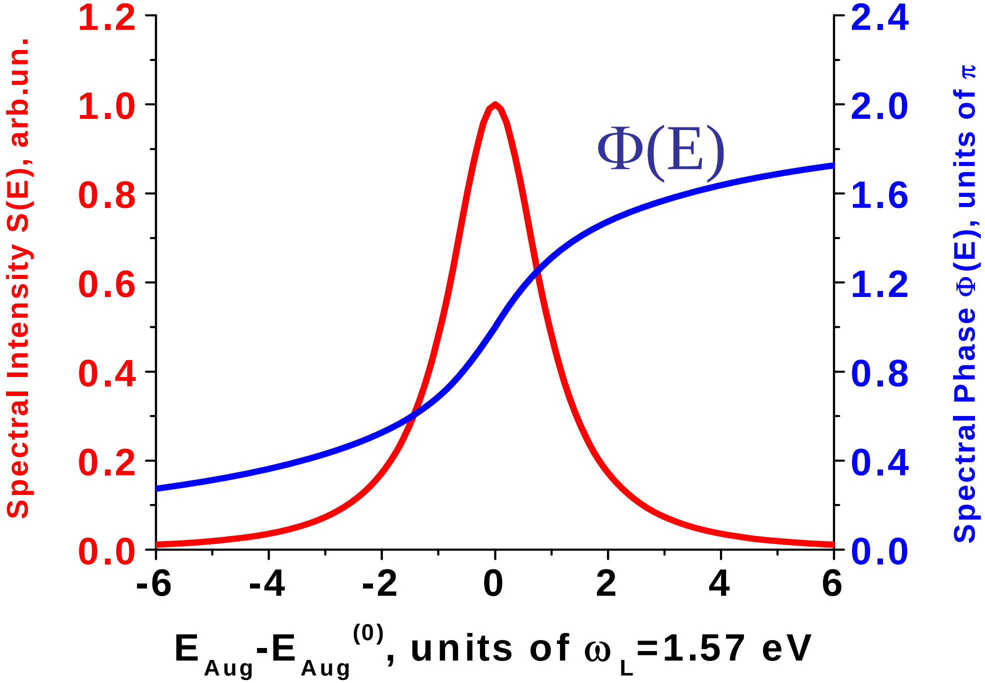
We propose: “long” pump – “long” probe + correlated measurement

The process measures itself

Time resolution is limited only by statistics and pump-probe jitter

Photo-induced Auger decay



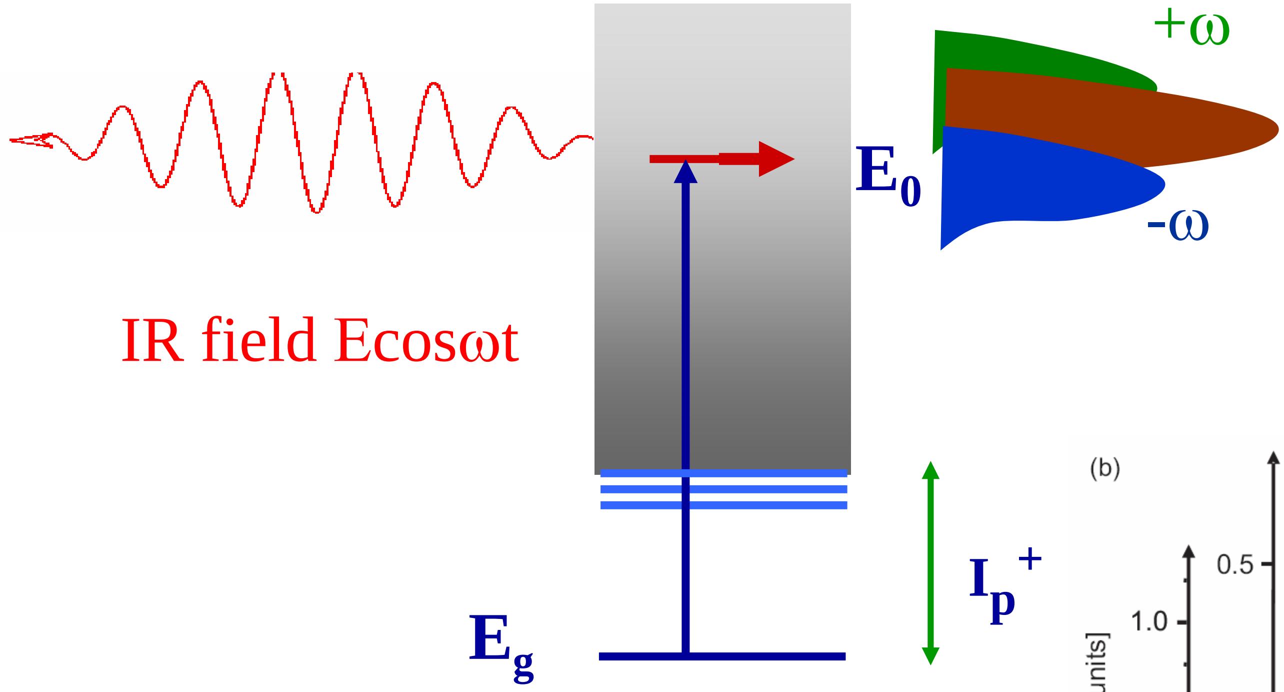


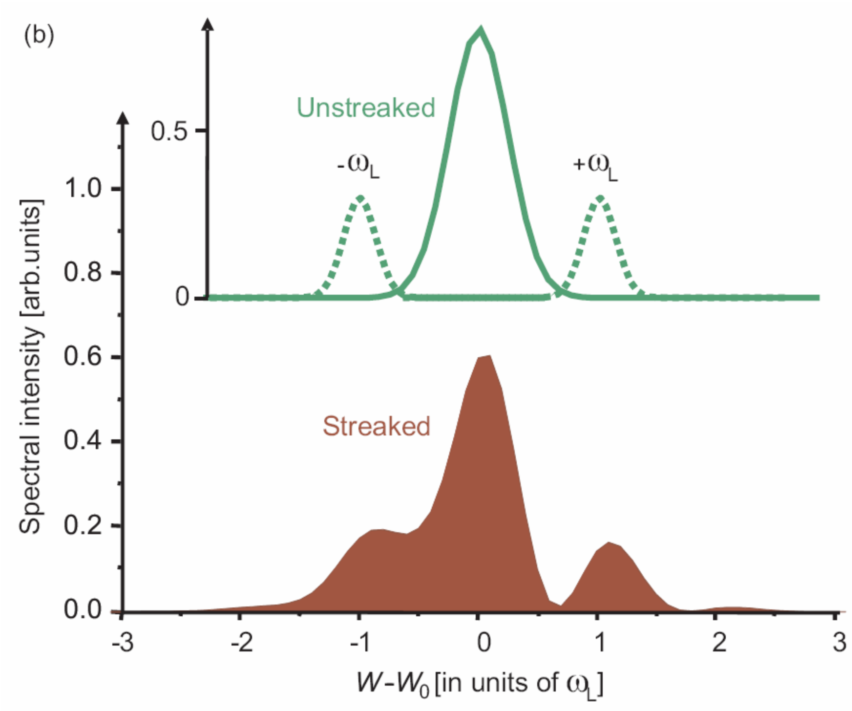
To get dynamics of the decay, one needs

EITHER direct time domain measurement OR the spectral phase Φ(E)

- We use spectral approach
- We need complete (correlated) two-electron spectrum

SPIDER-like Streak-Camera for LONG XUV





- •The process measures itself
- •There is no fundamental limit to time-resolution

For two-electron ionization we need two-electron spectrum

A la Carte

- **Down to 0.01 fsec time resolution:**
  - **CASTOR**

    Correlated

    Atto-Second

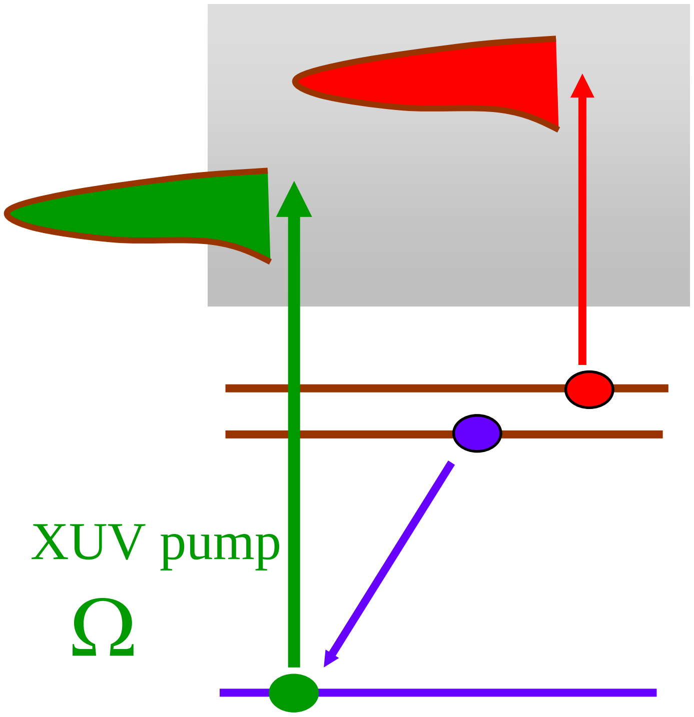
    Two-electron

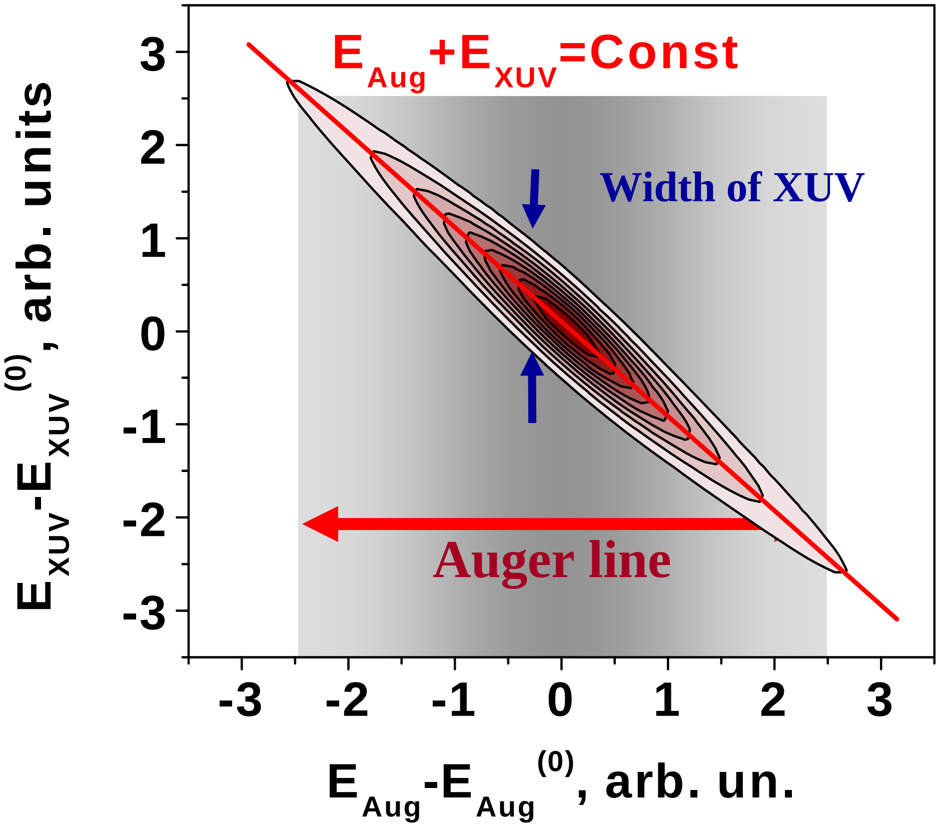
    Optical

    Reconstruction

Correlated two-electron spectra

$$E_{Aug} + E_{XUV} = \Omega - I_p^{++}$$

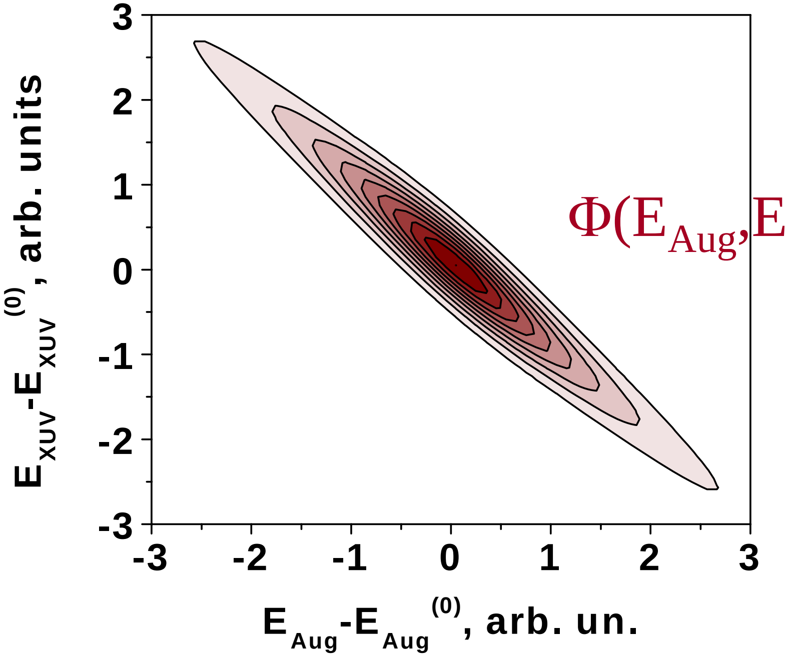




$$C \propto \tilde{E}_X\left[E_{XUV} + E_{Aug} - \Omega_{XUV} + I_p^{++}\right]\tilde{F}_A\left[E_{Aug} - E_h + I_p^{++}\right]$$

Auger decay: the spectral amplitude

$$C \propto \tilde{E}_X\left[E_{XUV} + E_{Aug} - \Omega_{XUV} + I_p^{++}\right]\underbrace{\tilde{F}_A\left[E_{Aug} - E_h + I_p^{++}\right]}_{\text{Decay amplitude}}$$



$\Phi(E_{Aug},E_{XUV}) = \Phi_{XUV} + \Phi_{Aug}$

How can we reconstruct the spectral phase of $F_A(E)$, Φ_{Aug}?

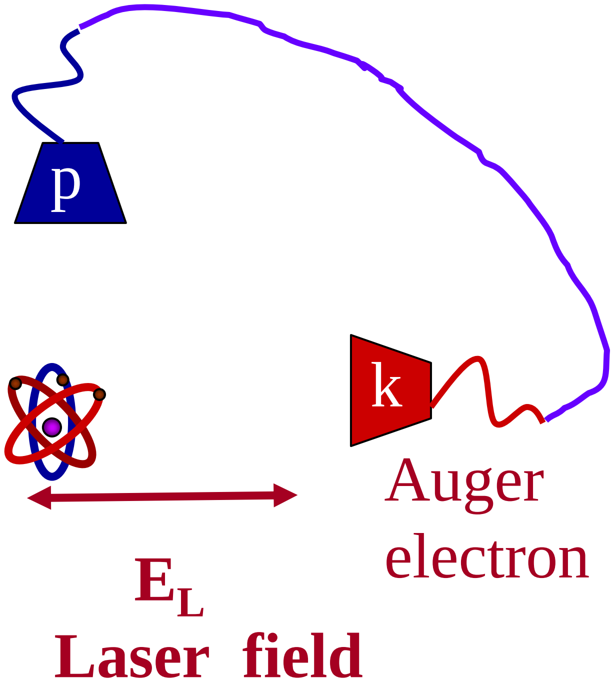
- Reconstruct the phase of $C[E_{XUV},E_{Aug}]$: $\Phi(E_{Aug},E_{XUV})$
- Use $\Phi(E_{Aug},E_{XUV}) = \Phi_{XUV} + \Phi_{Aug}$

The key point is to get $\Phi[E_{XUV},E_{Aug}]$. How?

Measurement geometry

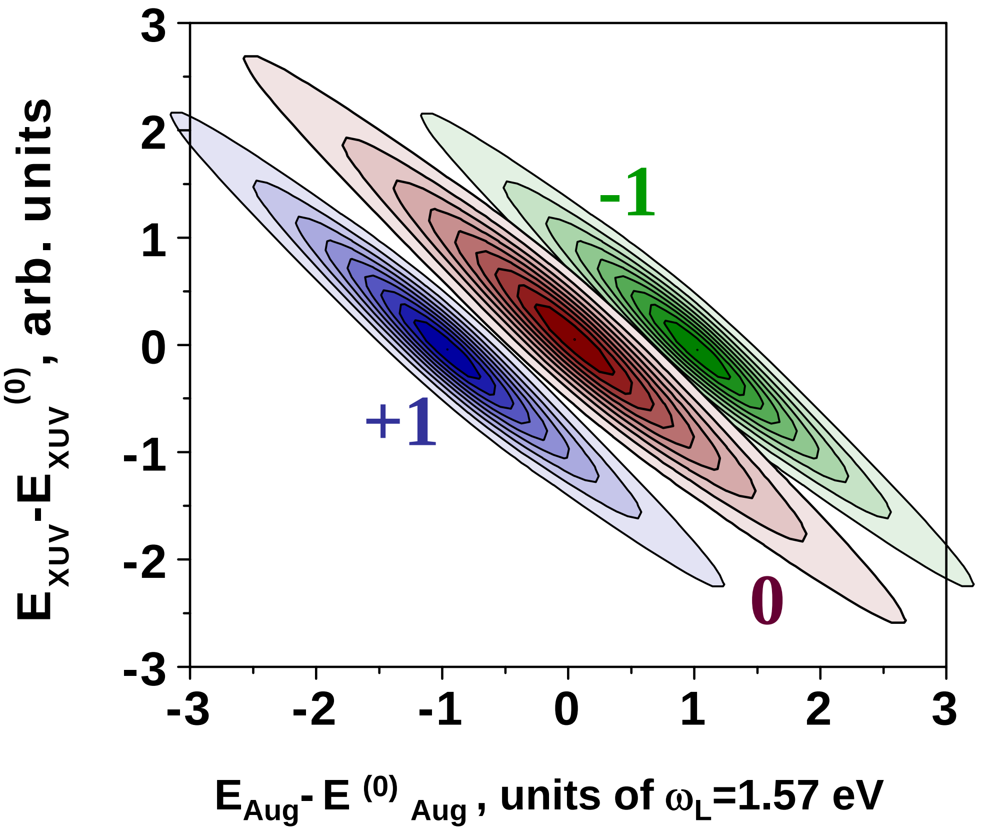
Detect $\mathbf{k}_{\mathrm{Aug}} \parallel \mathbf{E}_L$ to ensure sufficient streak by E_L

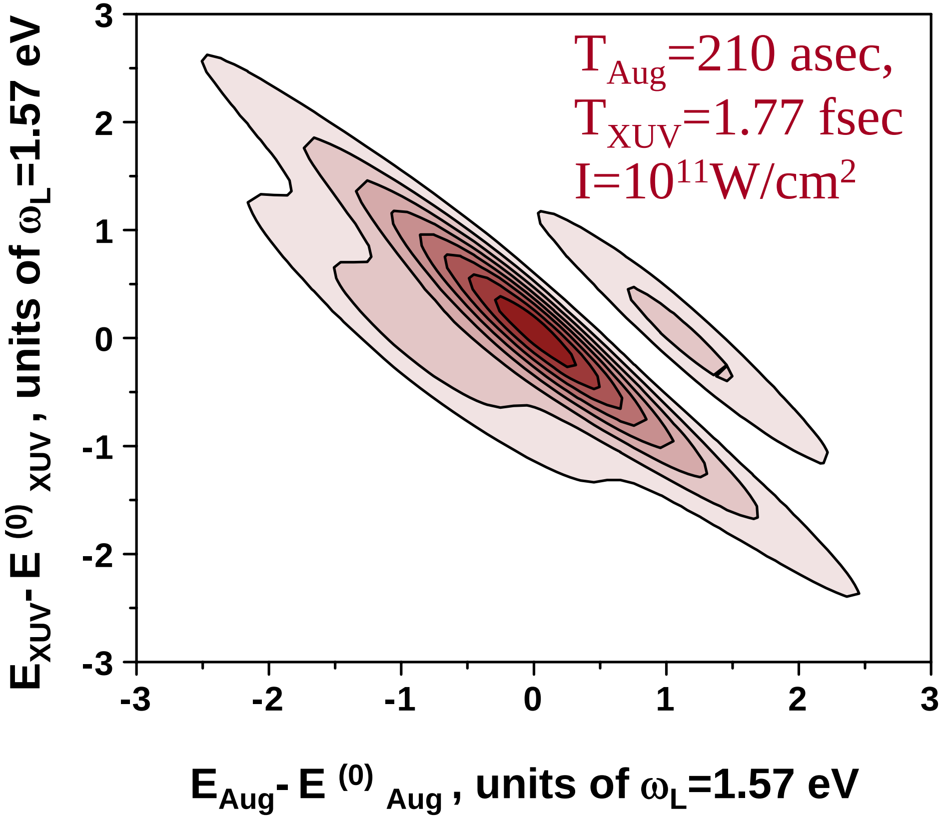
Detect $\mathbf{p}_{\mathrm{XUV}} \perp \mathbf{E}_L$ and reduce field so that XUV electron is not streaked



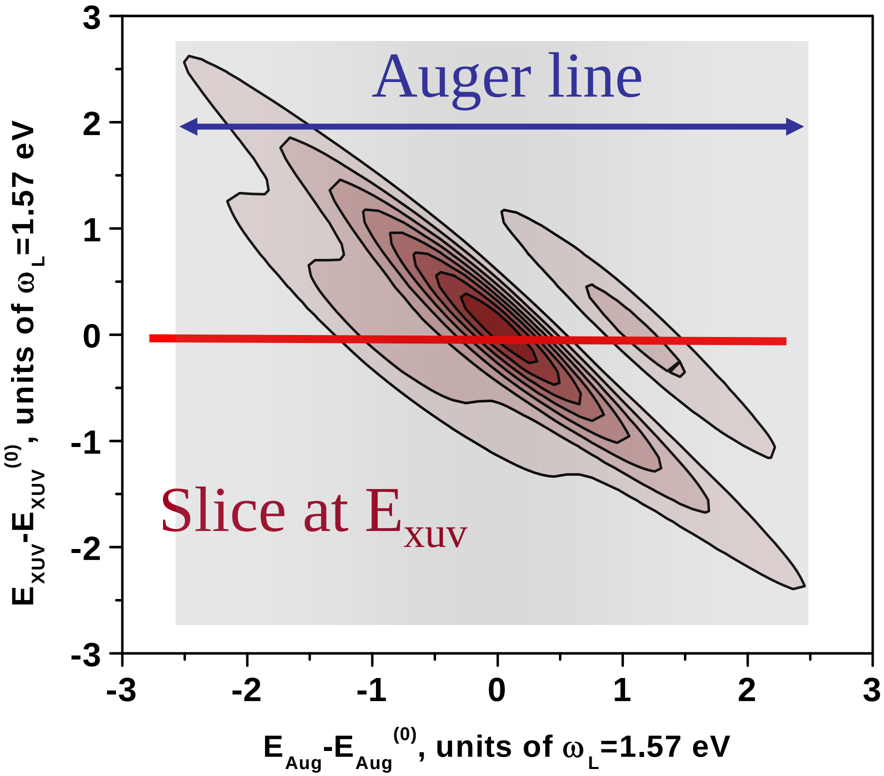
Streaked correlated spectra

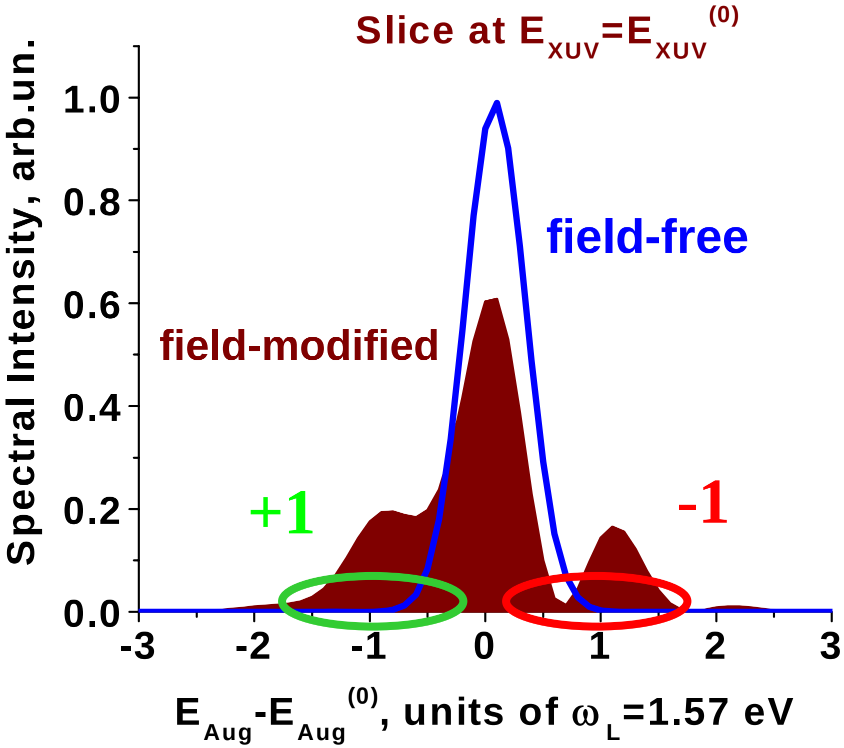
If there were no interference between sidebands:



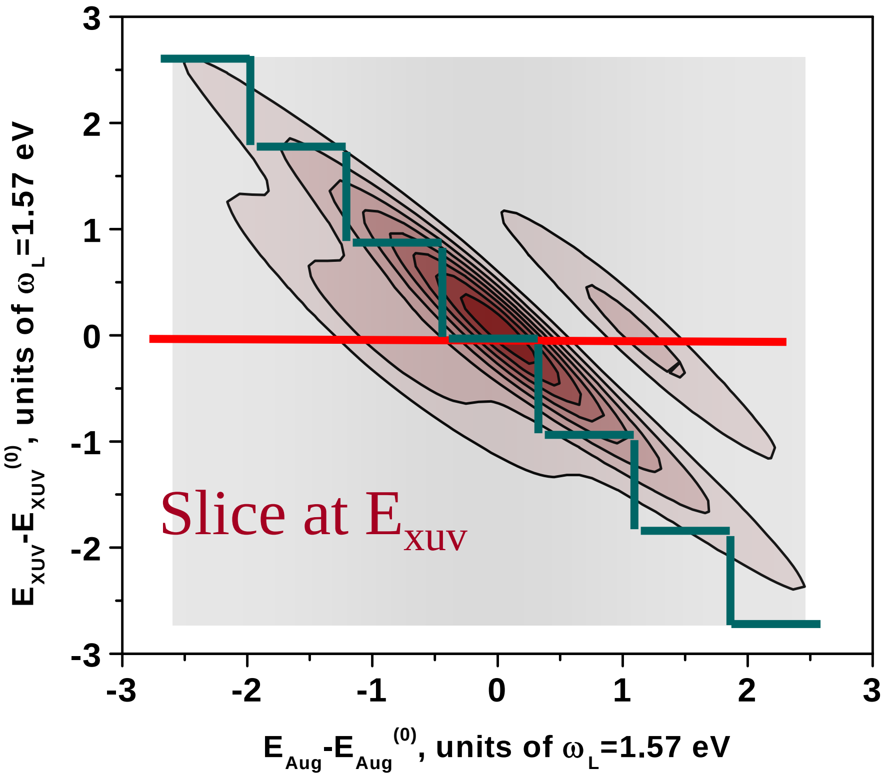


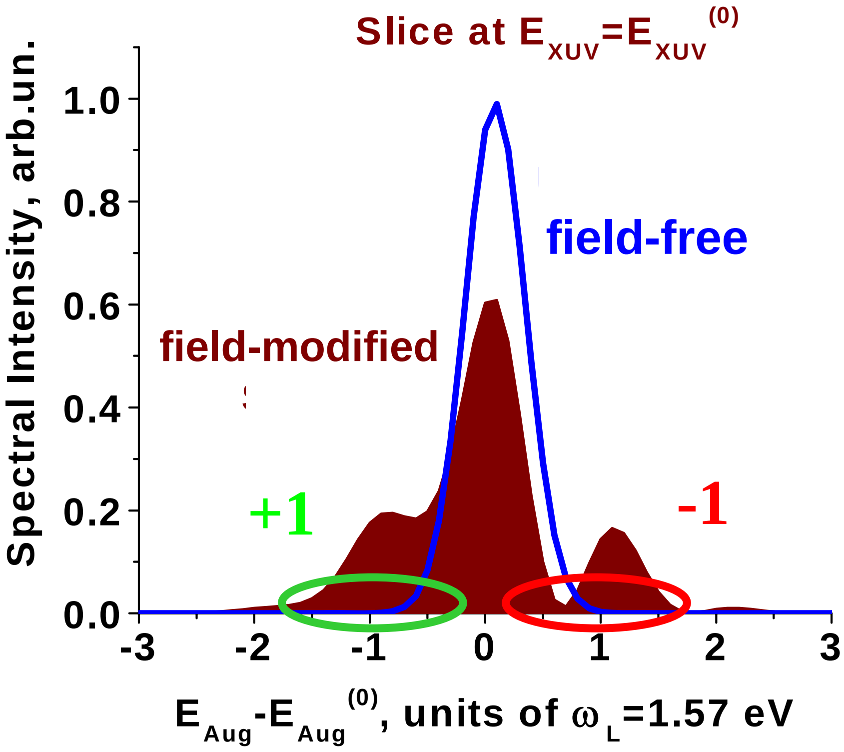
Effect of interference: individual slice





Succession of slices

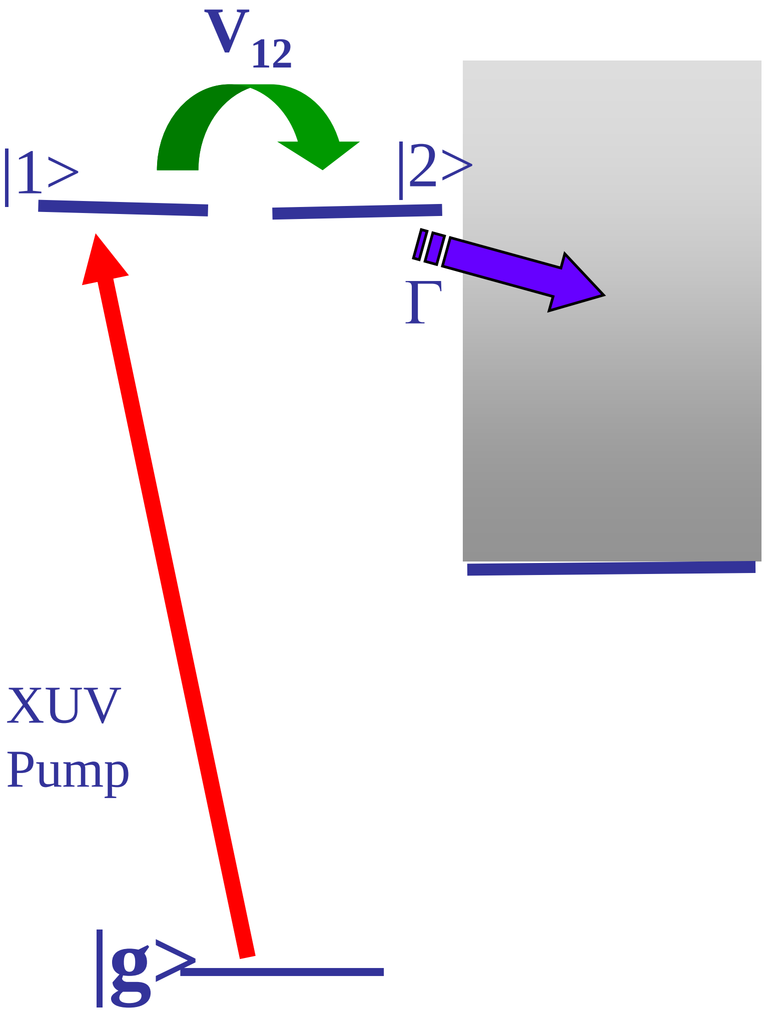




Using interference of 0, 1, and –1 sidebands, reconstruct spectral phase of $C_{\text{field-free}}\,[E_{xuv},E_{Aug}]$ **slice by slice**

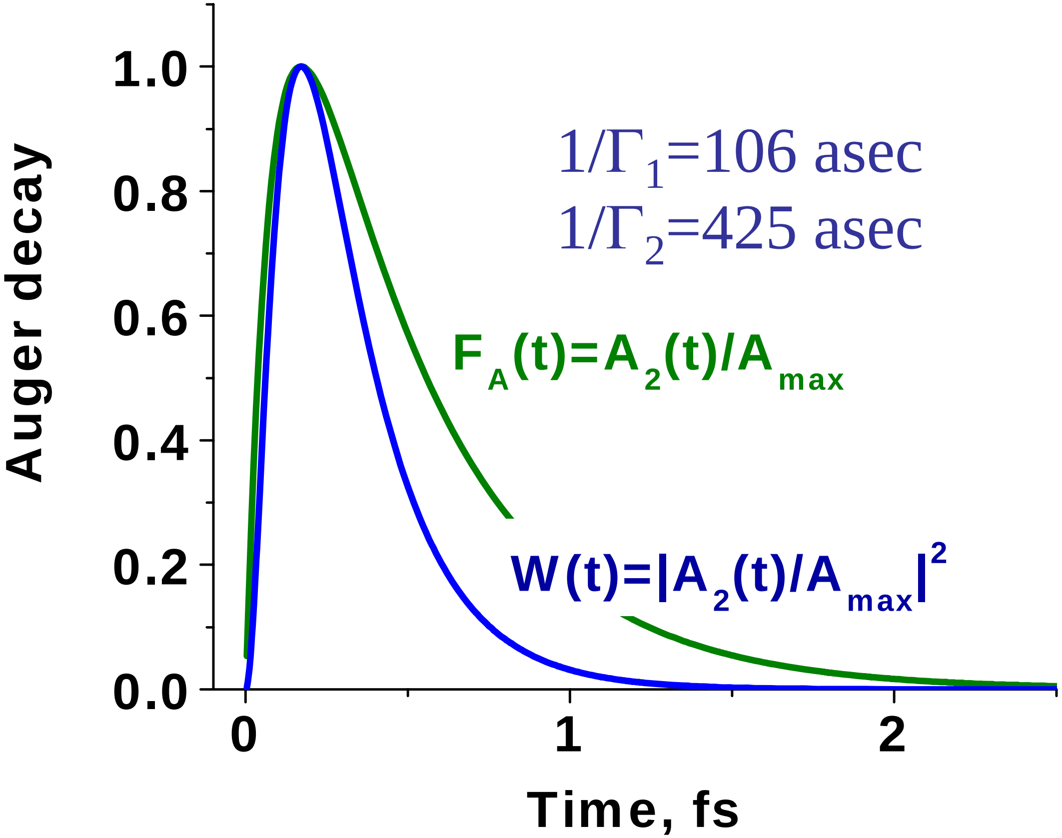
We are using correlated two-electron distribution to reconstruct its phase - *use entanglement to reconstruct the entangled wavefunction*

Example: decay after core rearrangement



$$F_A(t) = [1 - \exp(-\Gamma_1 t)]\exp(-\Gamma_2 t)$$

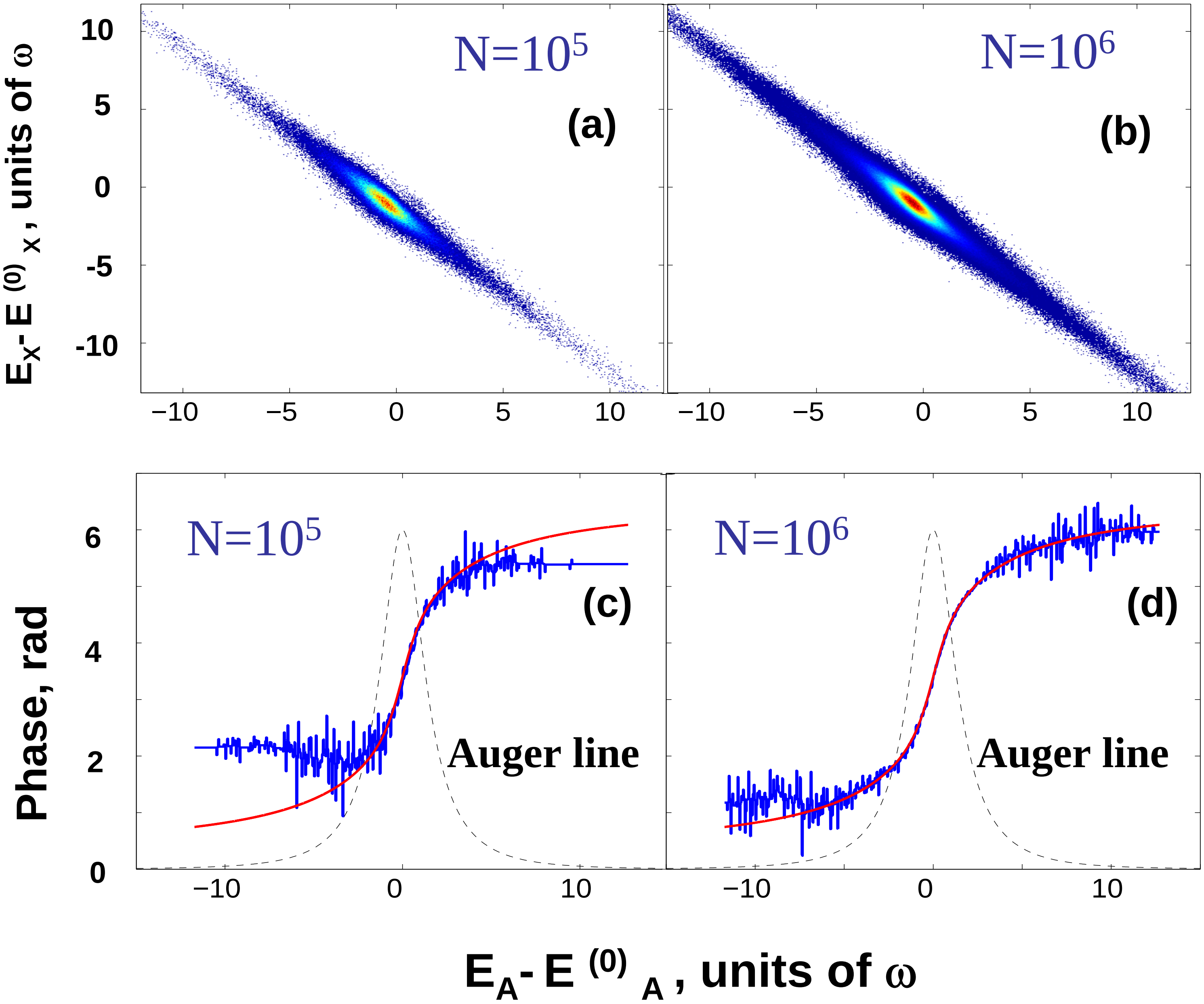
$$\Gamma_{1,2} = \Gamma\,\frac{1}{2}\left[1 \pm \sqrt{1 - \frac{4V_{12}^{\,2}}{\Gamma^2}}\right]$$



$1/\Gamma_1$=106 asec

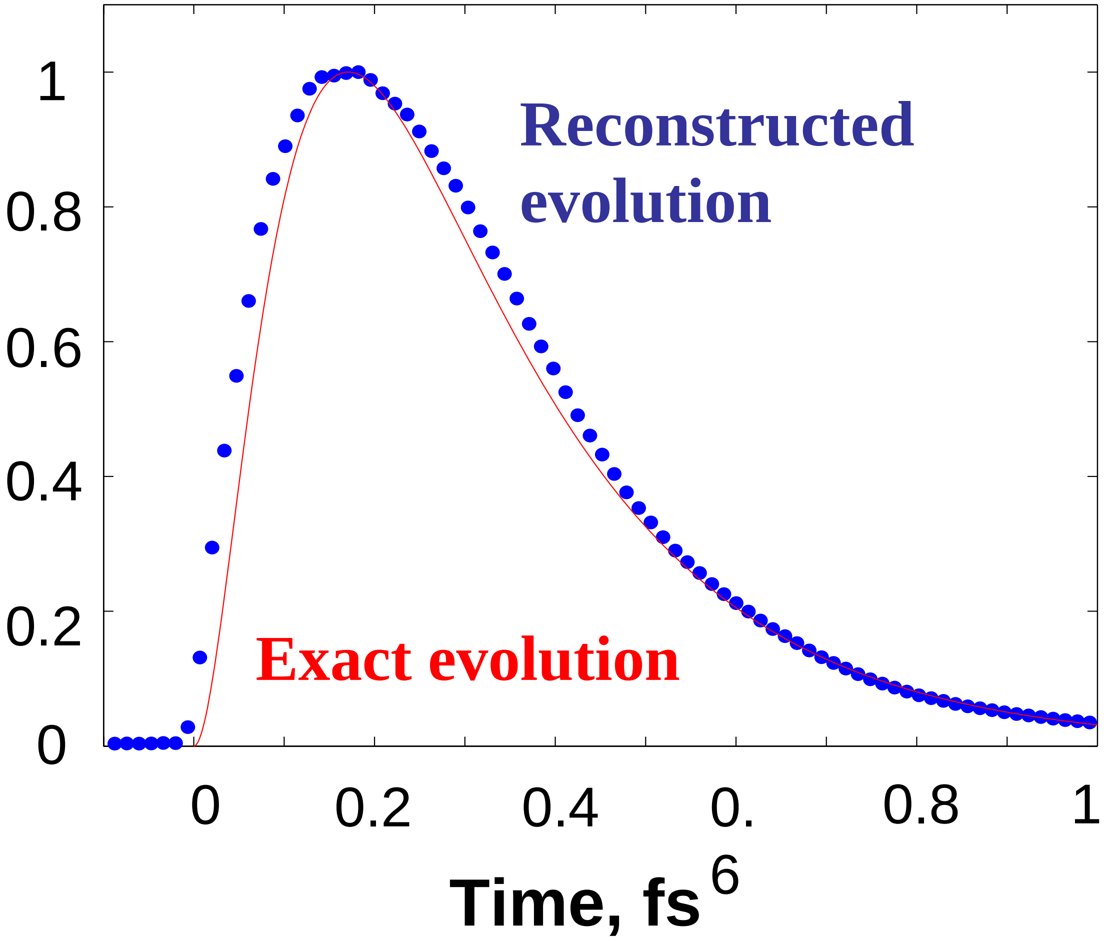
$1/\Gamma_2$=425 asec

Results of Reconstruction: Frequency domain



Results of Reconstruction: Accuracy

-XUV pulse: FWHM=1.2 fsec, I=10^{10} W/cm^2 ,
- 100 asec jitter, 160 meV energy resolution of electron spectrometer, 10^5 total counts



Accuracy: 10-20 asec

Conclusions

Correlated measurements enhance temporal resolution in pump-probe measurements

We are using correlated two-electron distribution to reconstruct its phase-

use entanglement to reconstruct entangled wavefunction

Time resolution comes from the fast process itself + temporal stability of XUV relative to IR streaking field