Ultrashort Phase Locked Laser Pulses for Asymmetric Electric Field Studies of Molecular Dynamics

Kelsie Betsch
University of Virginia
Department of Physics
AMO/Fourth Year Seminar
April 13, 2009

Overarching Goal

- Application of asymmetric electric fields to molecular dynamics studies
    - Two color fields
    - Ultrashort Carrier-Envelope (CE) phase stabilized pulses
        - Synthesis of CEP stabilized pulses

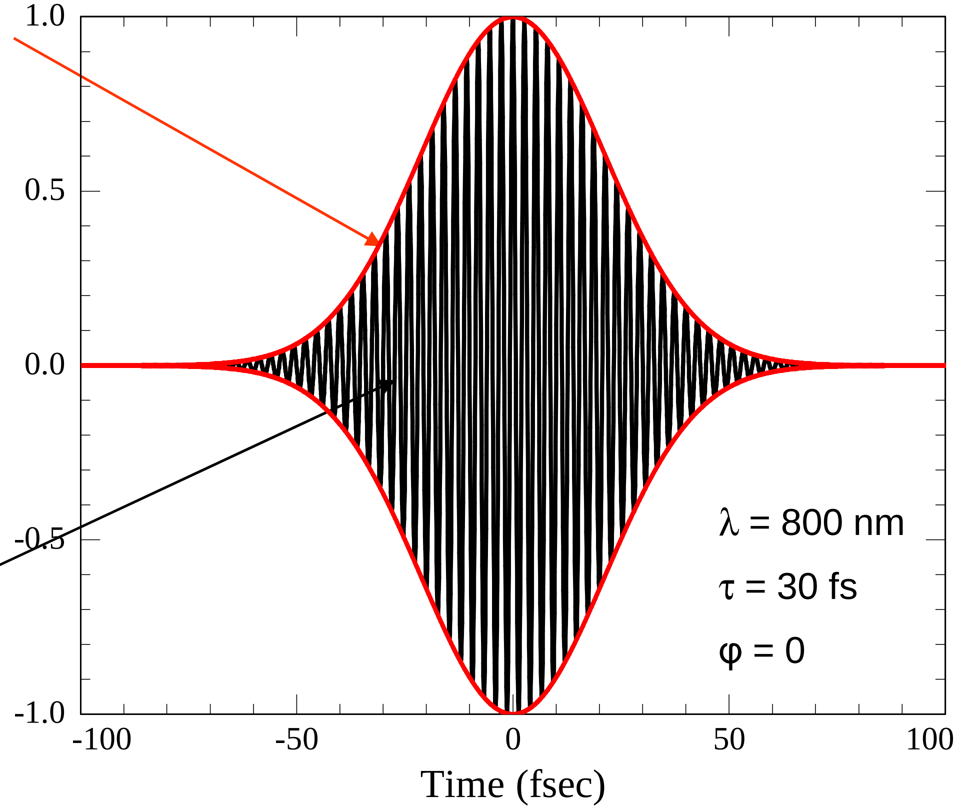
A Bit about Laser Pulses

$E_0(t)=\exp(-(t/\tau)^2)$
envelope

$$E(t) = E_0(t)\cos(\omega t+\varphi)$$

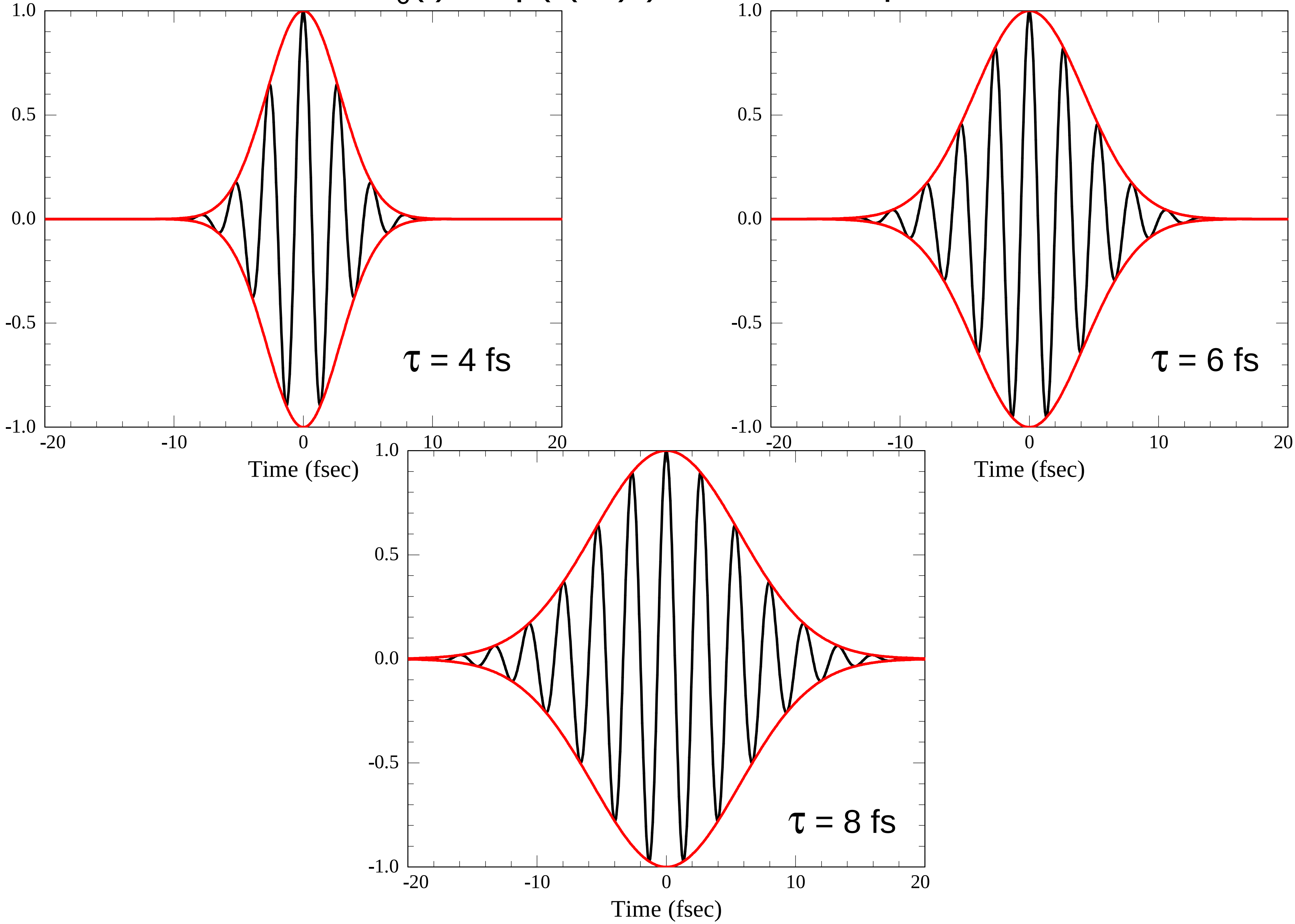
Carrier
envelope phase

carrier



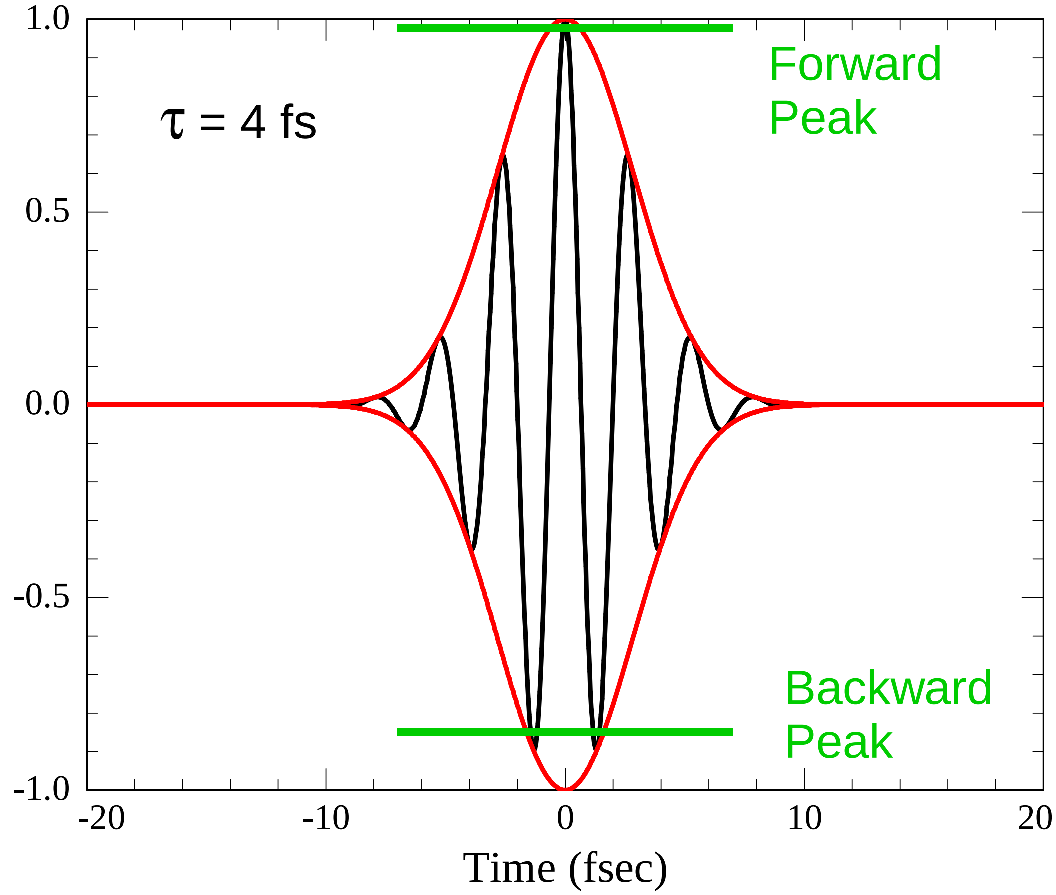
$$E(t) = E_0(t)\cos(\omega t+\varphi)$$

$$E_0(t)=\exp(-(t/\tau)^2);\ \lambda=800\text{nm};\ \varphi=0$$



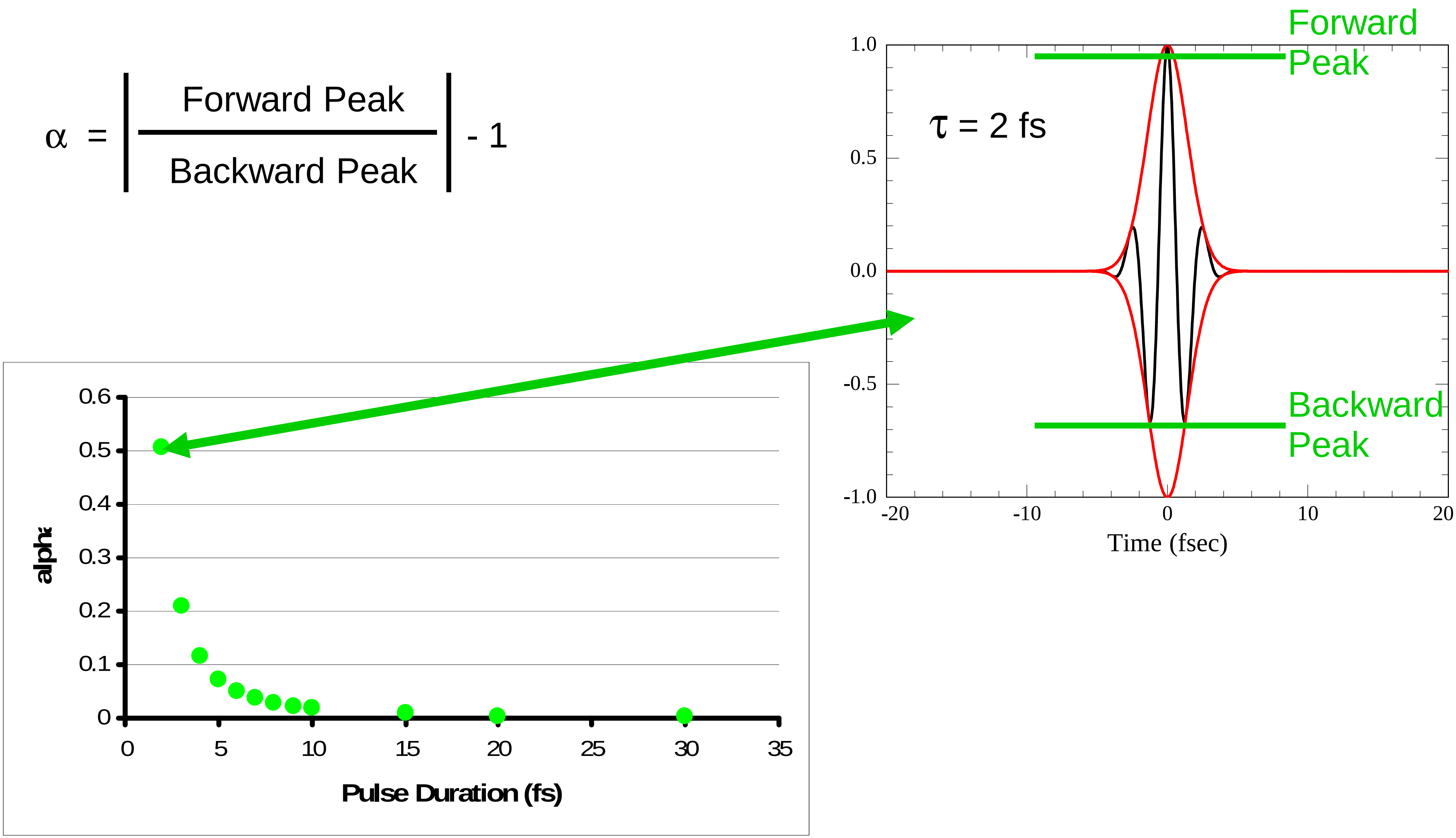
Electric Field Asymmetry vs Pulse Duration

$$\alpha = \left| \frac{\text{Forward Peak}}{\text{Backward Peak}} \right| - 1$$



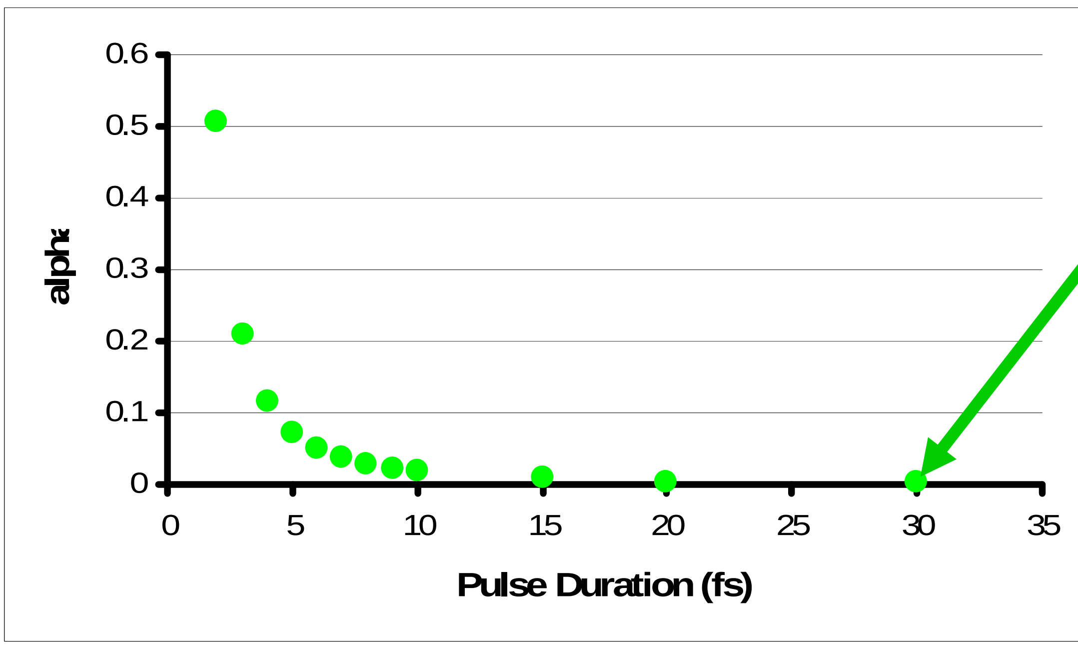
Electric Field Asymmetry vs Pulse Duration

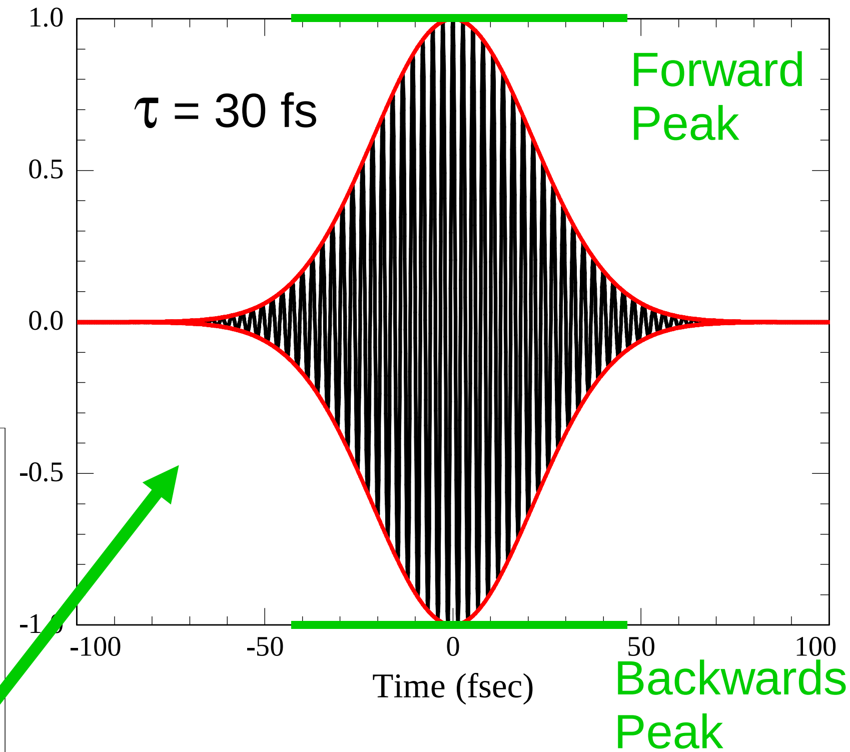
$$\alpha = \left| \frac{\text{Forward Peak}}{\text{Backward Peak}} \right| - 1$$



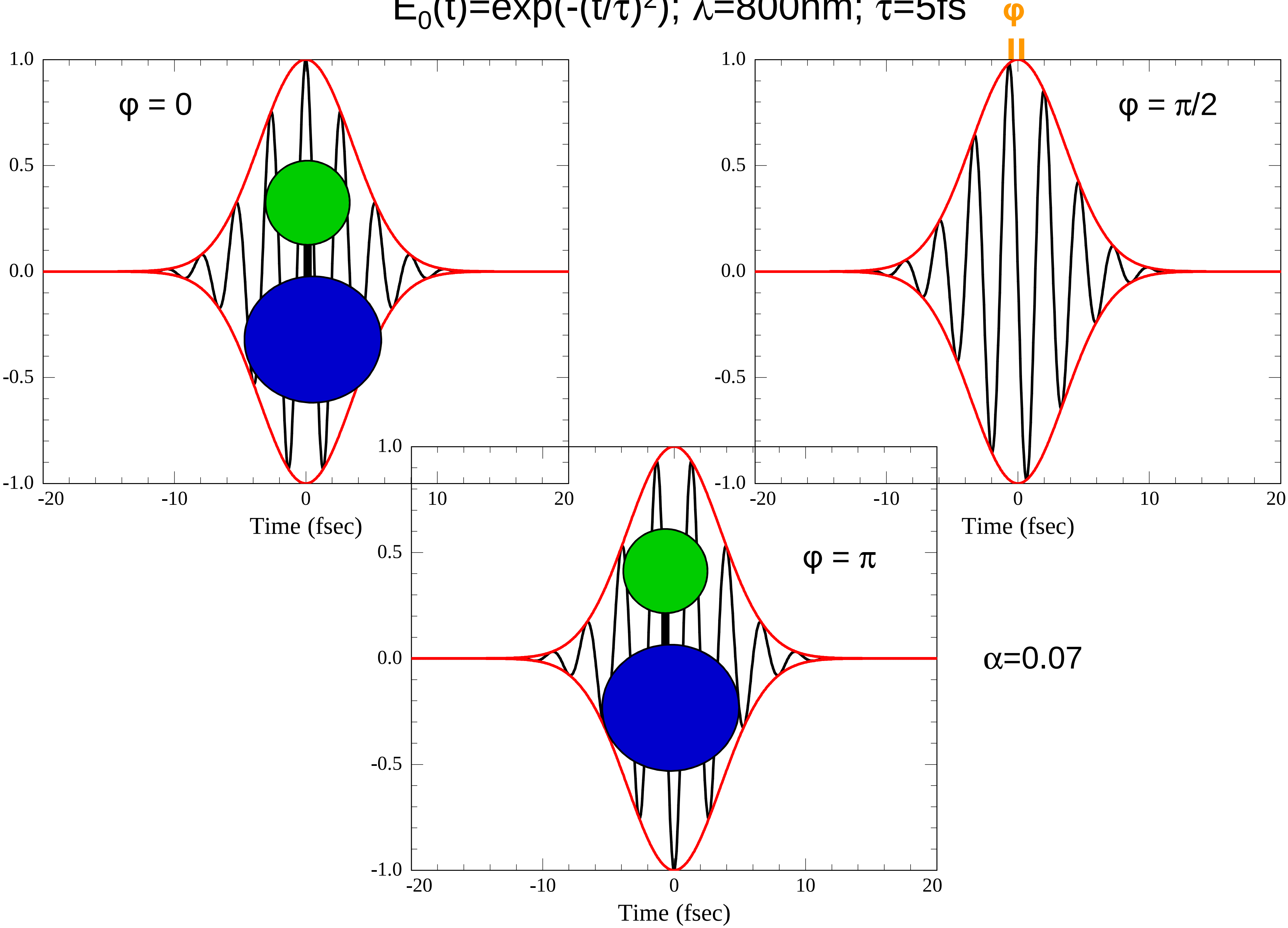
Electric Field Asymmetry vs Pulse Duration

$$\alpha = \left| \frac{\text{Forward Peak}}{\text{Backward Peak}} \right| - 1$$



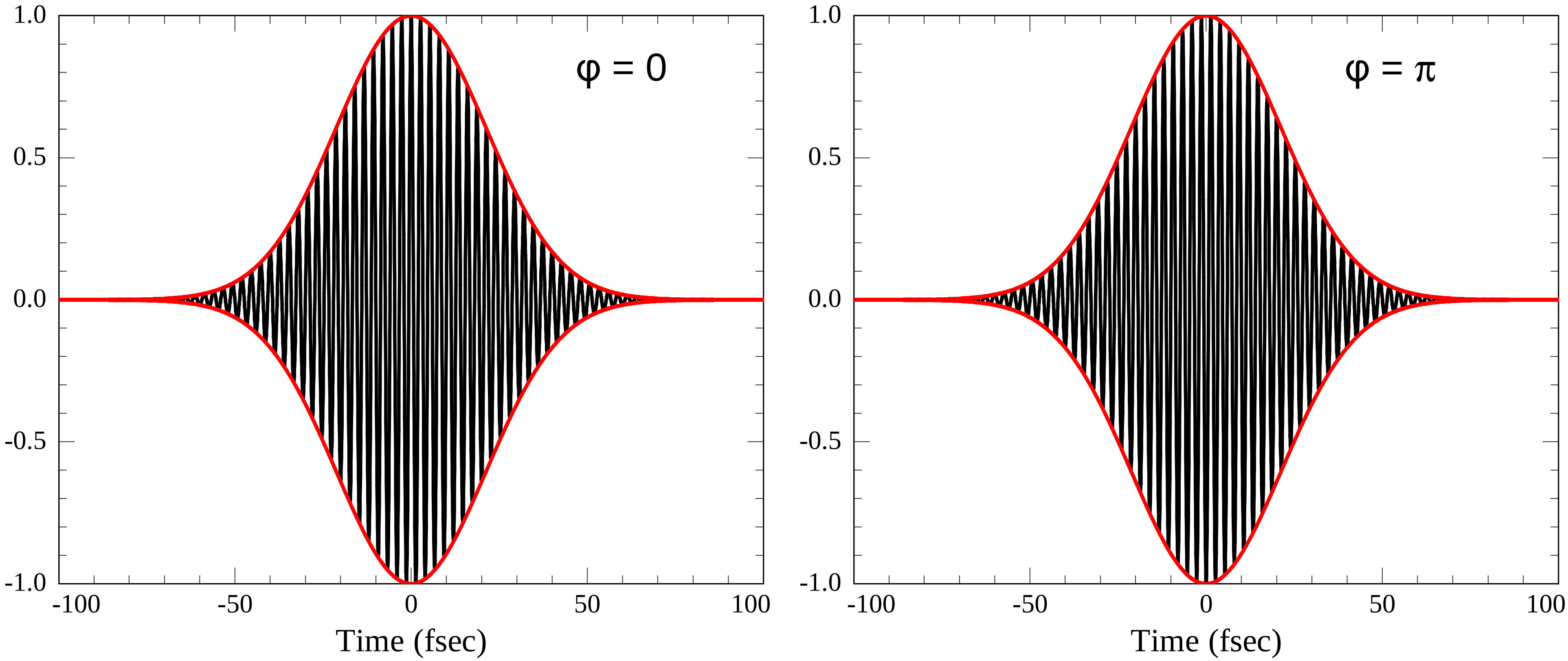


$$E(t) = E_0(t)\cos(\omega t+\varphi)$$
$$E_0(t)=\exp(-(t/\tau)^2);\ \lambda=800\text{nm};\ \tau=5\text{fs}$$



$\alpha=0.07$

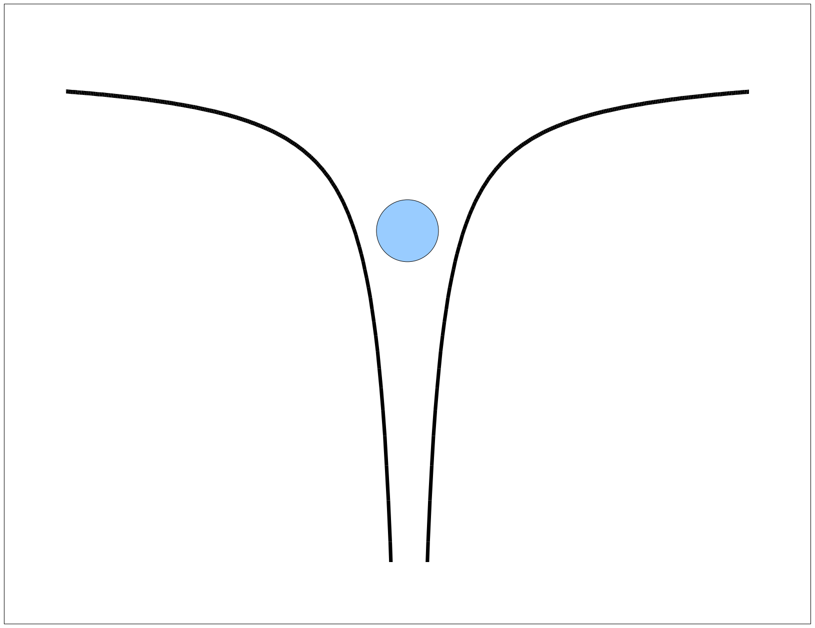
$$E(t) = E_0(t)\cos(\omega t+\varphi)$$
$$E_0(t)=\exp(-(t/\tau)^2);\ \lambda=800\text{nm};\ \tau=30\text{fs}$$



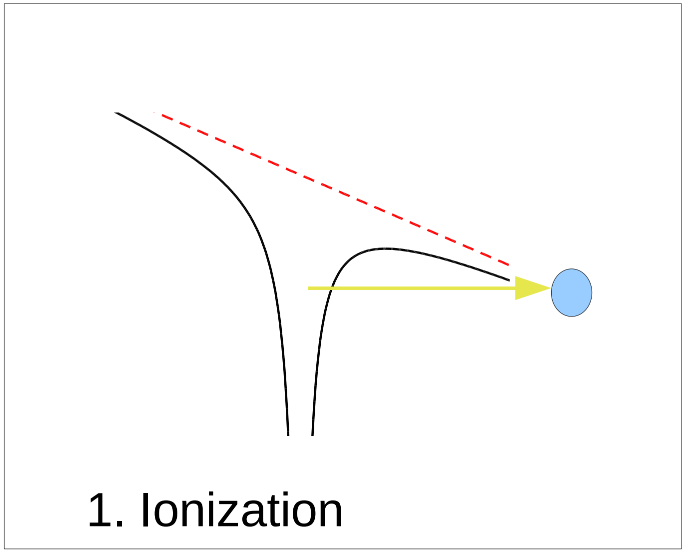
$\alpha=0.002$

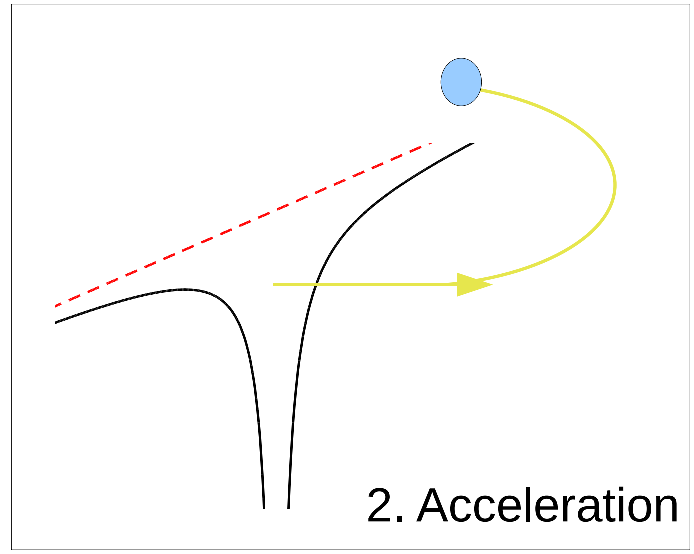
- Ultrashort laser pulses can look different
  - Pulse Duration
  - φ (Carrier Envelope Phase)
- Why would atoms or molecules care?

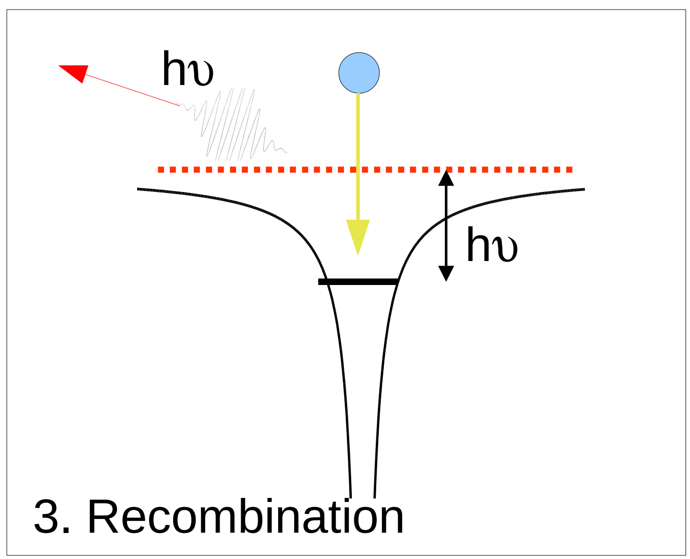
High Harmonic Generation



HHG: Three-Step Model







$E_{max}=I_p+3.17U_p$

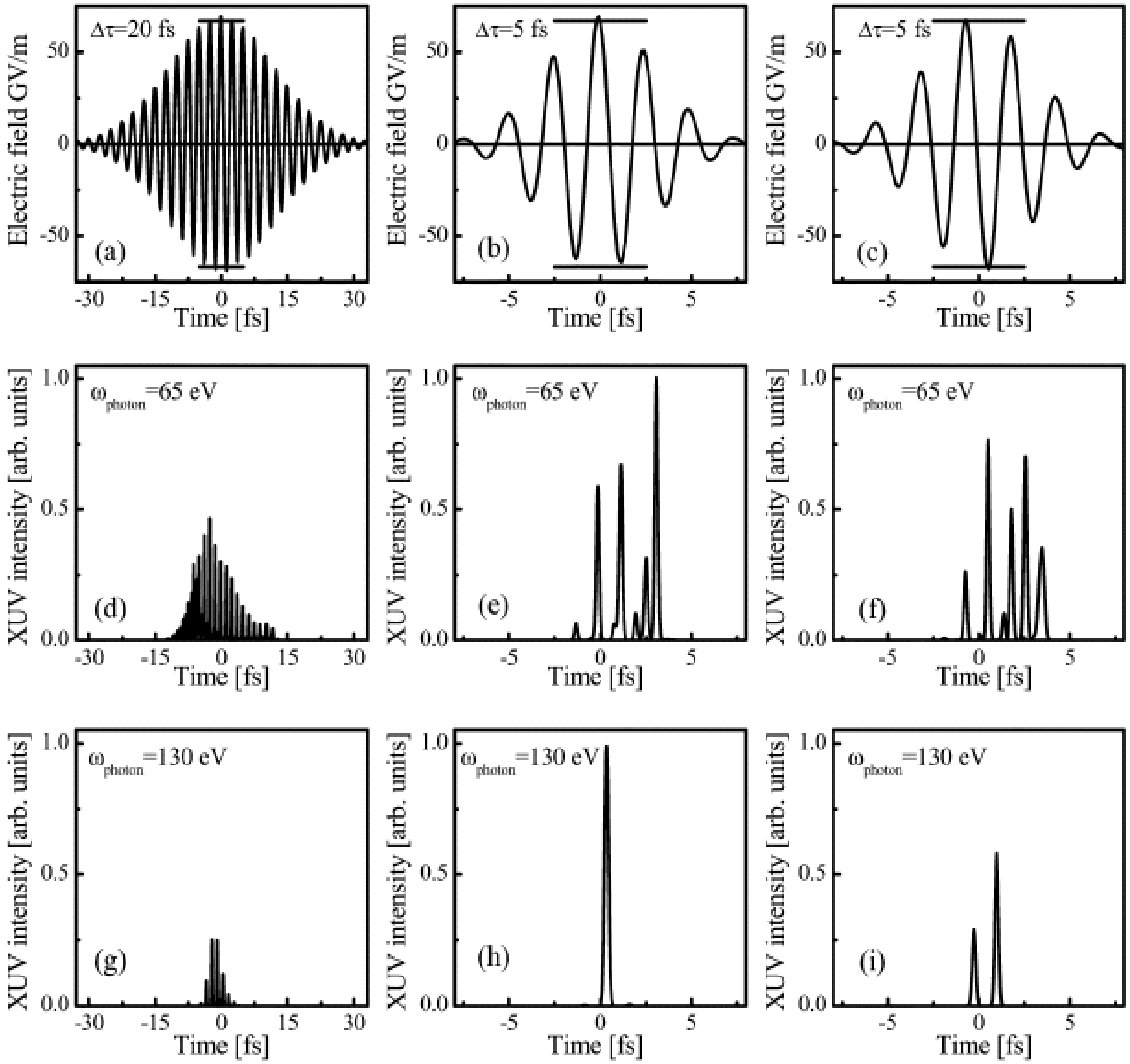
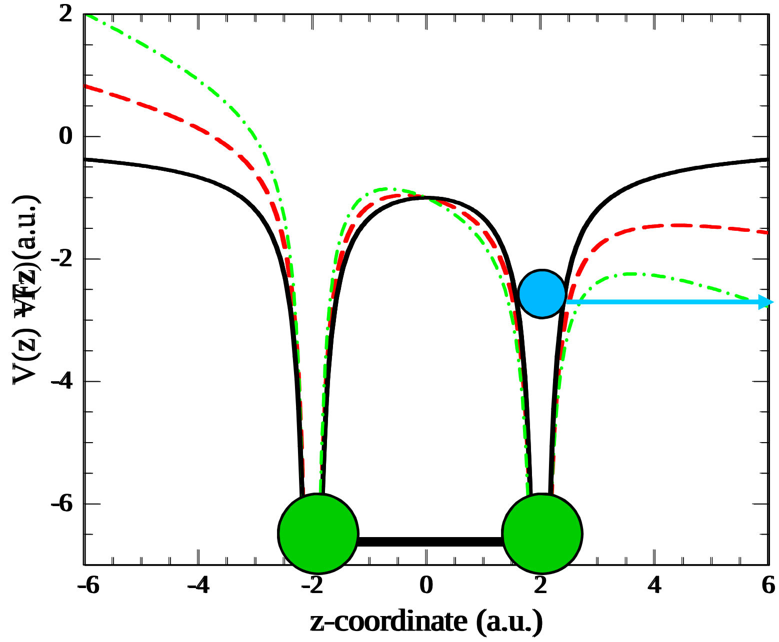


Fig. 1. Numerical simulation of laser-driven soft-X-ray emission from noble gas atoms. (a) Electric field of a 20-fs laser pulse. (b) Electric field of a 5-fs laser pulse optimized for generation of a single subfemtosecond X-ray pulse. (c) Electric field of a 5-fs pulse producing two highest energy X-ray pulses. (d)–(f) Time-domain structures of X-ray radiation emitted in a 10-eV bandwidth at half the cutoff energy. (g)–(i) Time-domain structures of X-ray radiation emitted in a 10-eV bandwidth at the cutoff energy. In this calculation, a neon gas target has the length of 2 mm and the pressure of 100 mbar. Pulse intensity was chosen to reach the cutoff frequency of 130 eV. Lower X-ray peak yield in (d) and (g) in comparison with (e), (f), (h), and (i) is the consequence of higher ionization by more numerous field peaks of the 20-fs pulse. In total, gas concentration loss due to ionization was 6.7% and 1.7% for the 20- and 5-fs pulses, respectively. Horizontal bars in (a)–(c) show peak intensity threshold required to yield cutoff-frequency X-ray radiation.

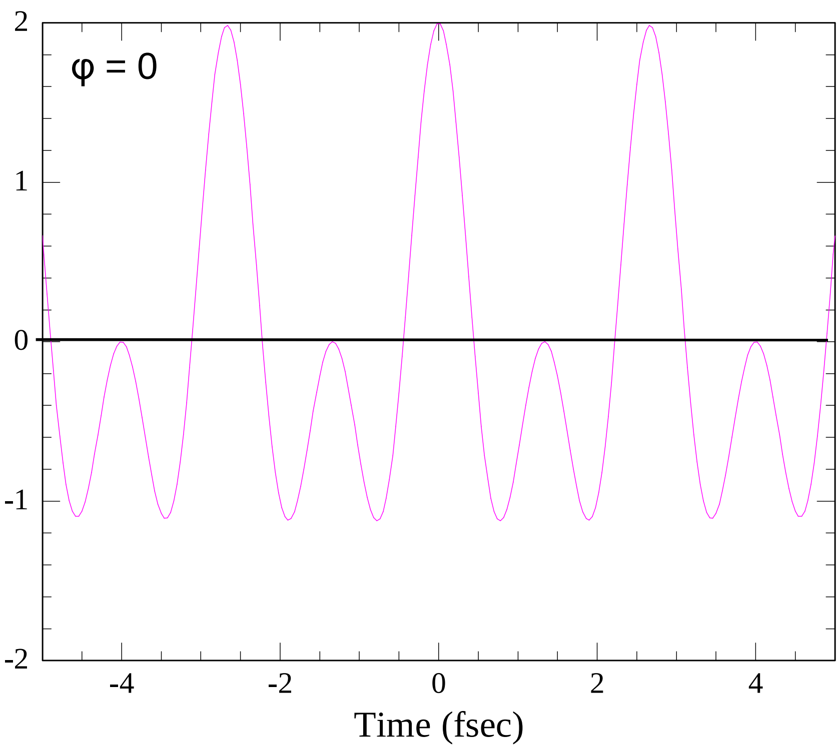
Baltuska et al, IEEE J. Sel. Top. Quant. Electron. **9**, 972 (2003).

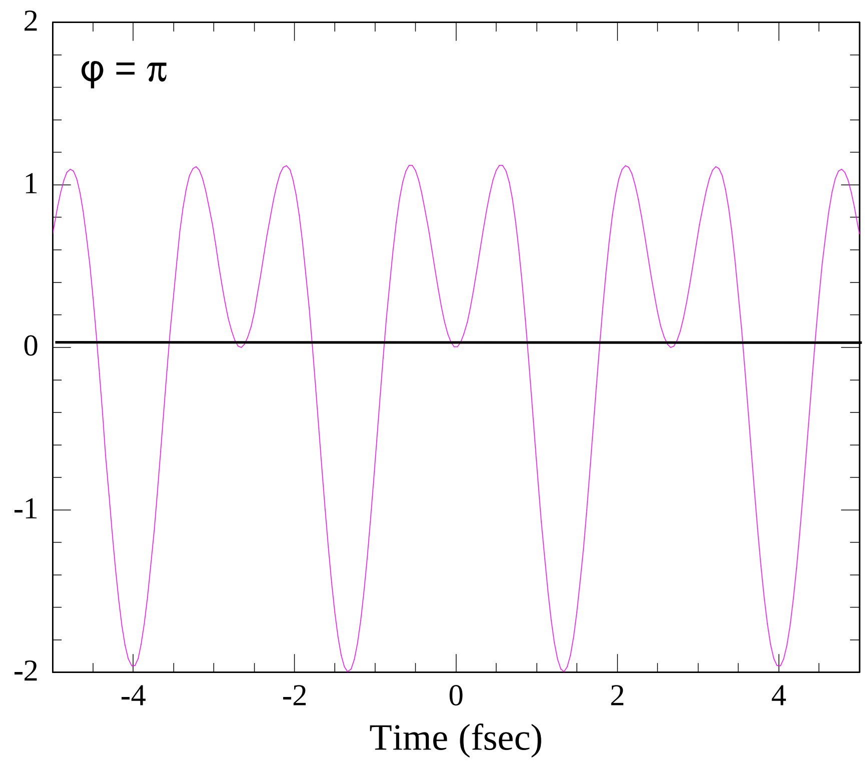
Two-Color Asymmetric Fields for N_2 Ionization

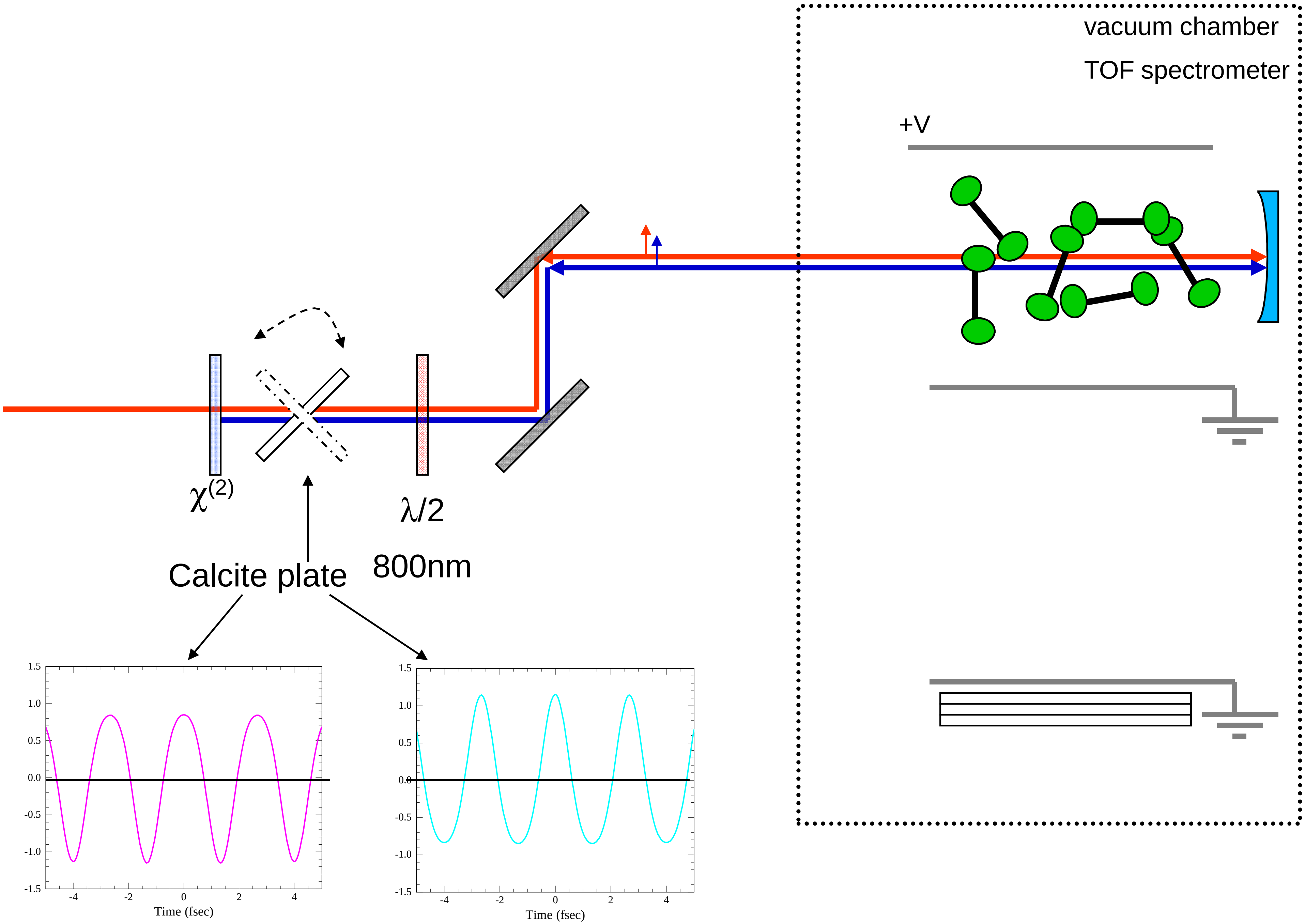
- N_2^+ tunnel ionization yield is sensitive to the electric field strength
  - Highly non-linear field dependence



$$E(t) = E_0(t)[\cos(\omega t) + \gamma \cos(2\omega t + \varphi)]$$
$$E_0(t) = \exp(-(t/\tau)^2);\ \lambda = 800\text{nm};\ \tau = 30\text{fs};\ \gamma = 1$$

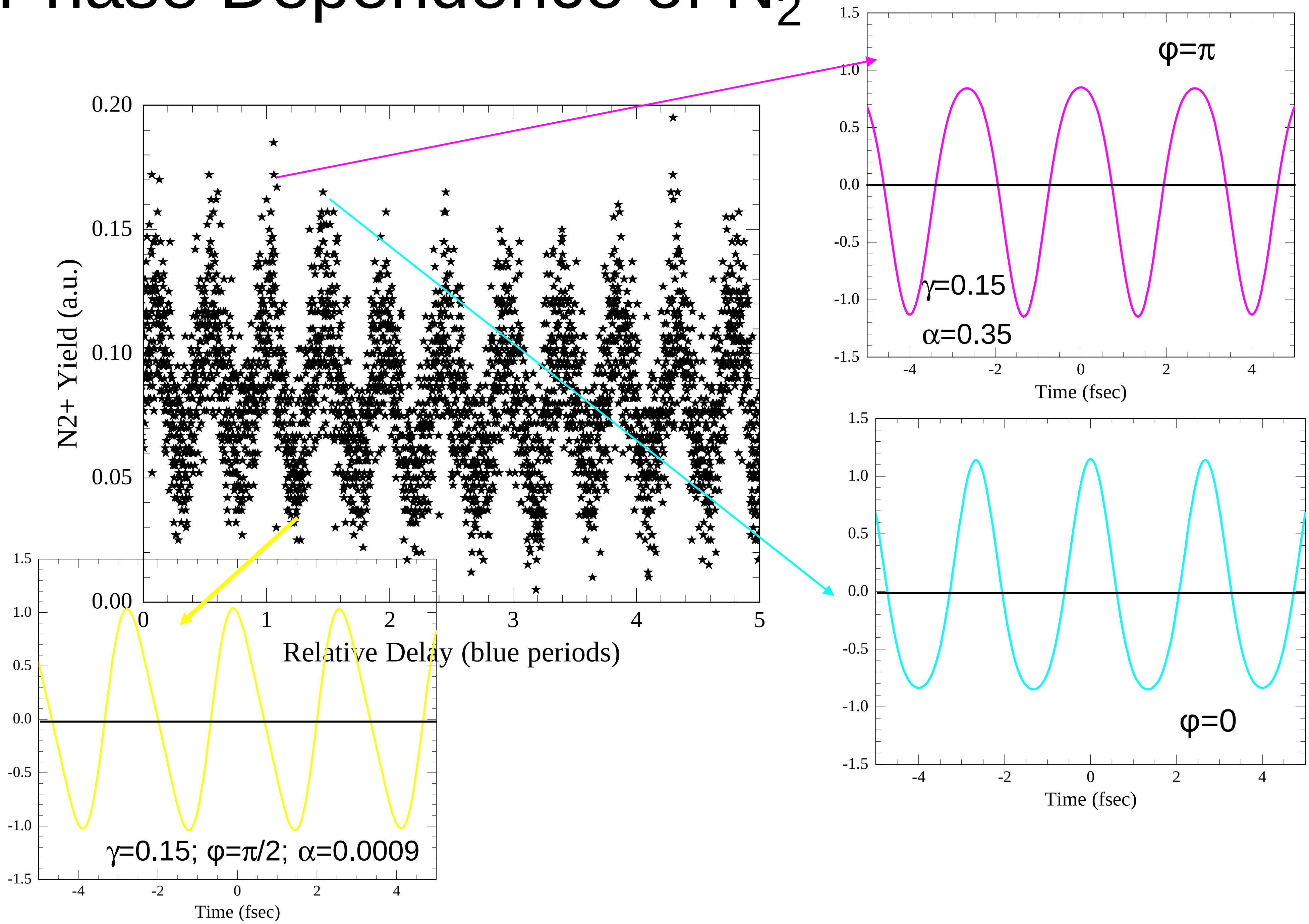






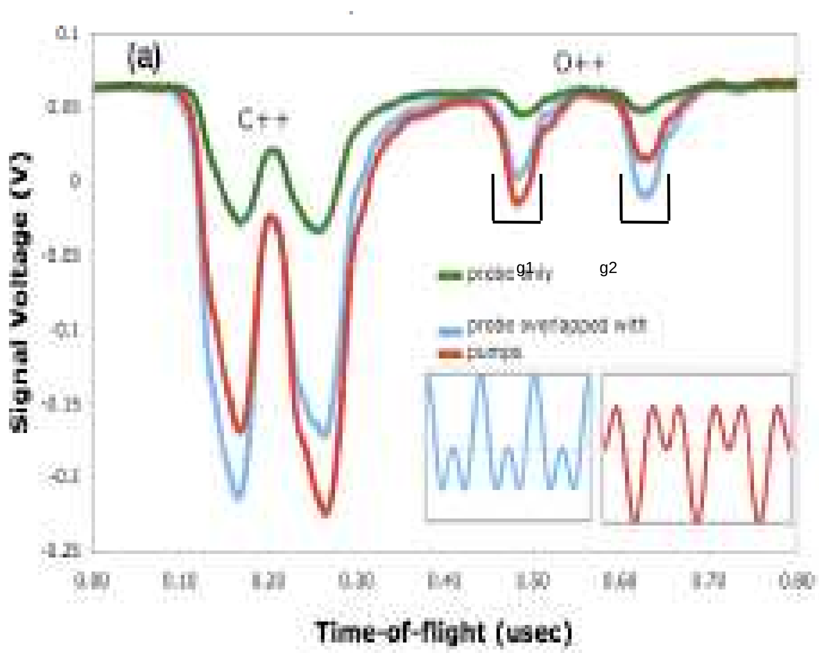
vacuum chamber
TOF spectrometer
+V
χ(2)
λ/2
800nm
Calcite plate
1.5
1.0
0.5
0.0
-0.5
-1.0
-1.5
-4
-2
0
2
4
Time (fsec)

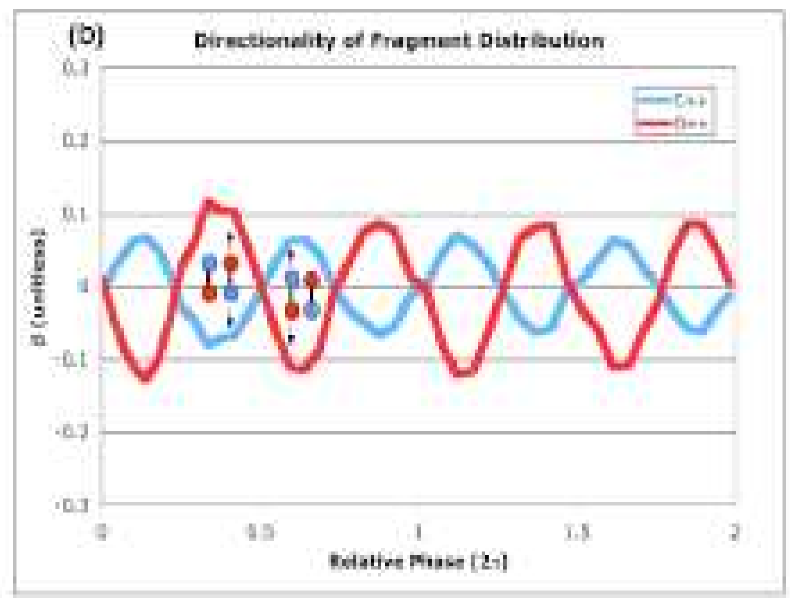
Phase Dependence of N_2^+



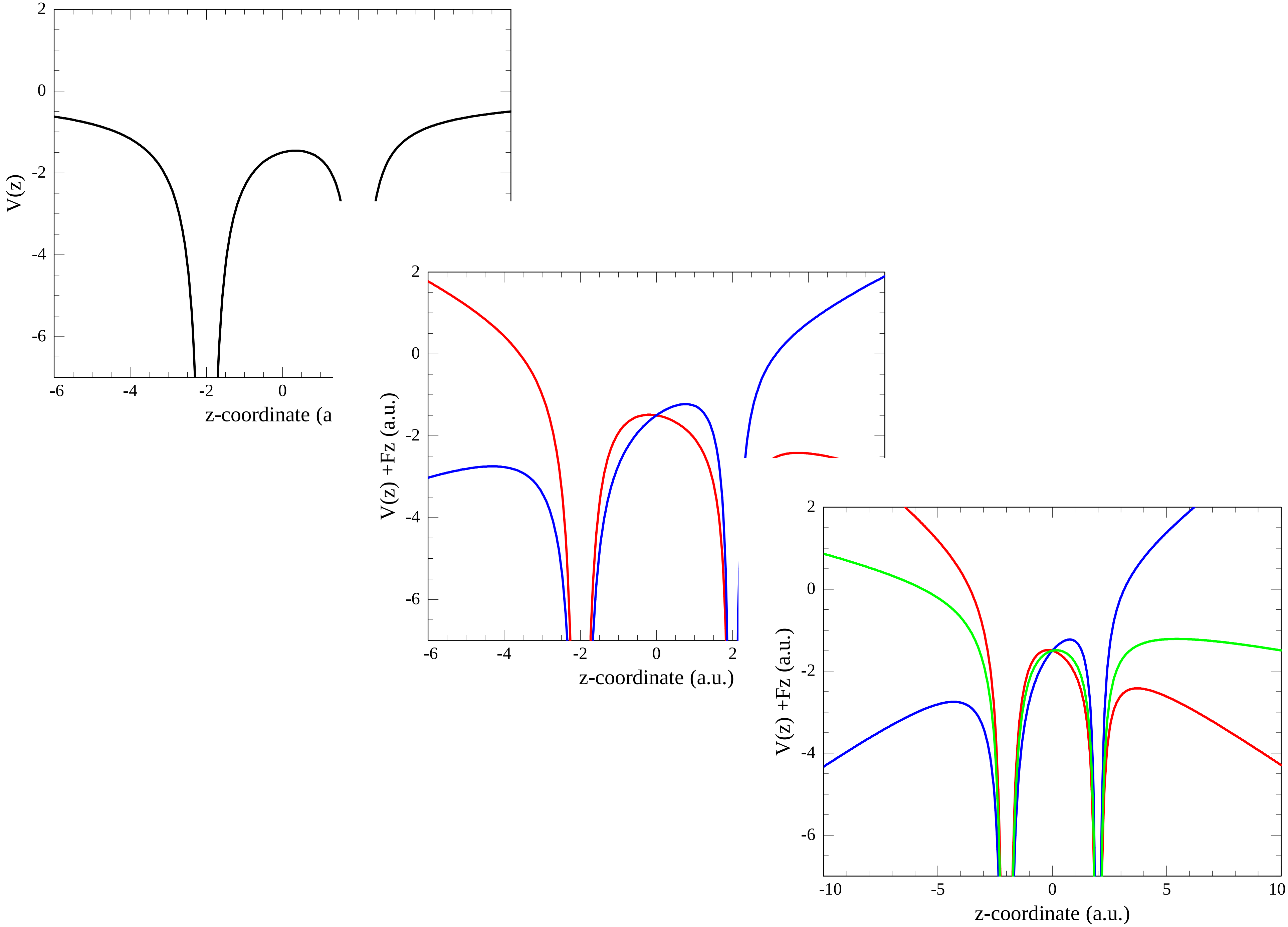
Asymmetric Molecular Dissociation

- Asymmetric dissociation
  - Attributed to enhanced ionization at R_c
  - 2% field asymmetry





from Dan Pinkham's dissertation


V(z)
z-coordinate (a
V(z) +Fz (a.u.)
z-coordinate (a.u.)
V(z) +Fz (a.u.)
z-coordinate (a.u.)

CEP in D_2^+ Dissociation

- $D_2 \rightarrow D^+ + D$
- Control forward/backward ejection of D+ by controlling φ
  - shows optical control of electron localization

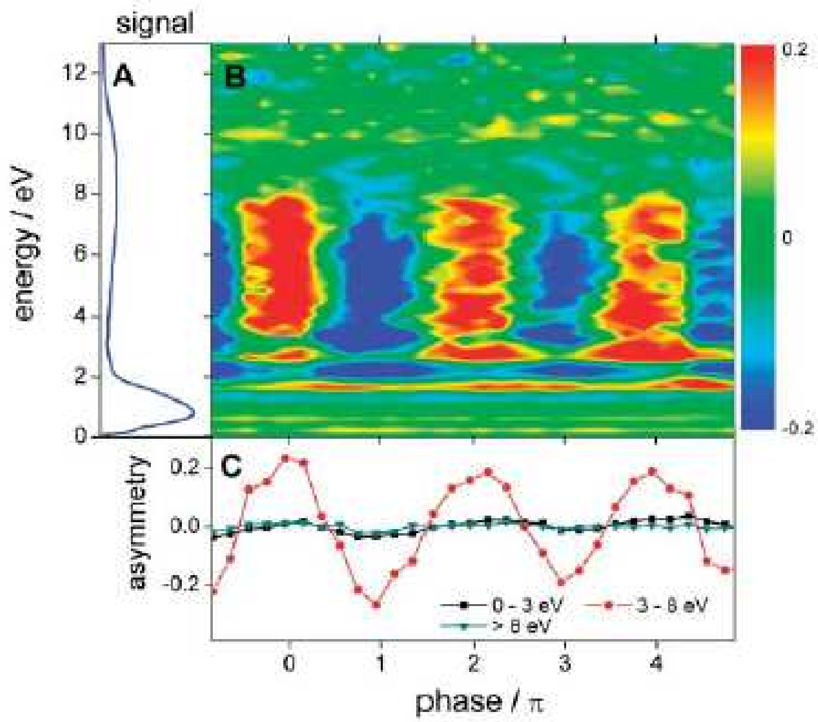


Fig. 3. (**A**) D^+ kinetic energy spectrum for D_2 dissociation with 5-fs, 1×10^{14} W cm^{-2} laser pulses without phase stabilization. (**B**) Map of asymmetry parameter $A(W,\varphi)$ as a function of the D^+ kinetic energy and carrier envelope phase φ (measured over a range of 6π with a step size of $\Delta\varphi = 0.1\pi$). (**C**) Integrated asymmetry over several energy ranges versus carrier envelope phase φ.

Kling et al, Science 312, 246 (2006).

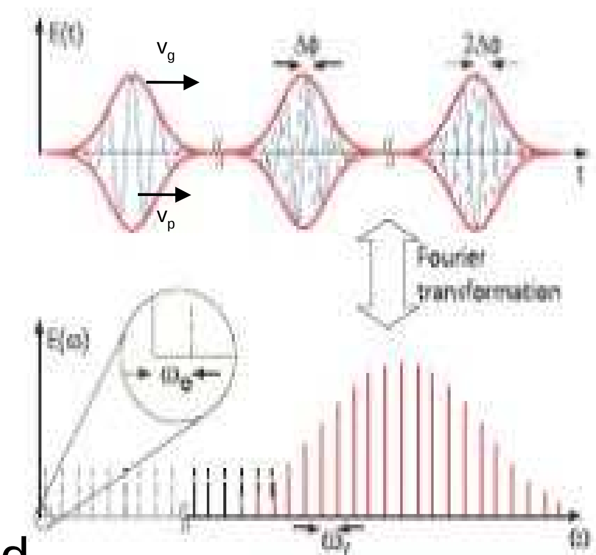
Towards Carrier-Envelope Phase Stabilized Pulses

- Origin of phase stabilization
- Laser Reconstruction process

Frequency Comb Theory

Ideal case: Pulse circulating in cavity of length L with carrier frequency ω_c

- Time Domain
  - Output is a sequence of nearly identical pulses separated by the round trip time $T=2L/v_g$ where is the mean group velocity
  - But $v_g \neq v_p$ => carrier/envelope phase shift $\Delta\varphi$ per pulse
- Frequency Domain
  - Spectrum is a comb of laser modes spaced by ω_r centered at ω_c
  - Continuous shift => offset frequency $\omega_0=\Delta\varphi/T$ from being exact harmonics of the repetition frequency



www.menlosystems.com

$$\omega_n=n\omega_r+\omega_0$$

Frequency Comb Theory

ω_r easy to measure
ω_0 need more than one optical octave:

$\omega_n = n\omega_r + \omega_0$

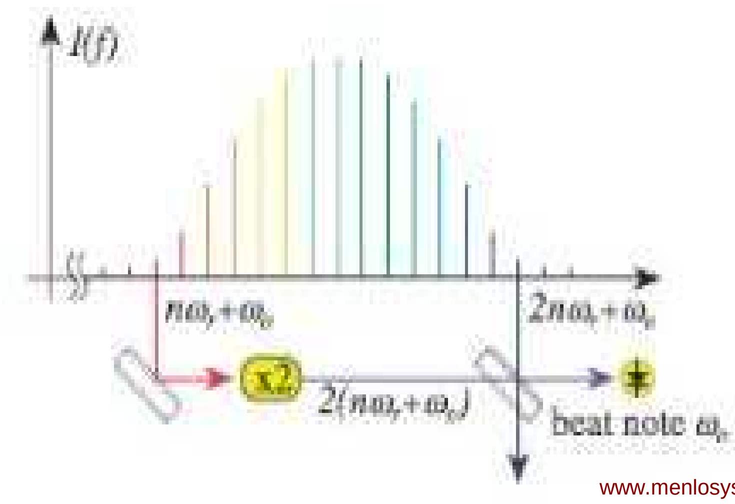
Red side, mode number n

$$\omega_n = n\omega_r + \omega_0$$

Blue side, mode number 2n

$$\omega_{2n} = 2n\omega_r + \omega_0$$

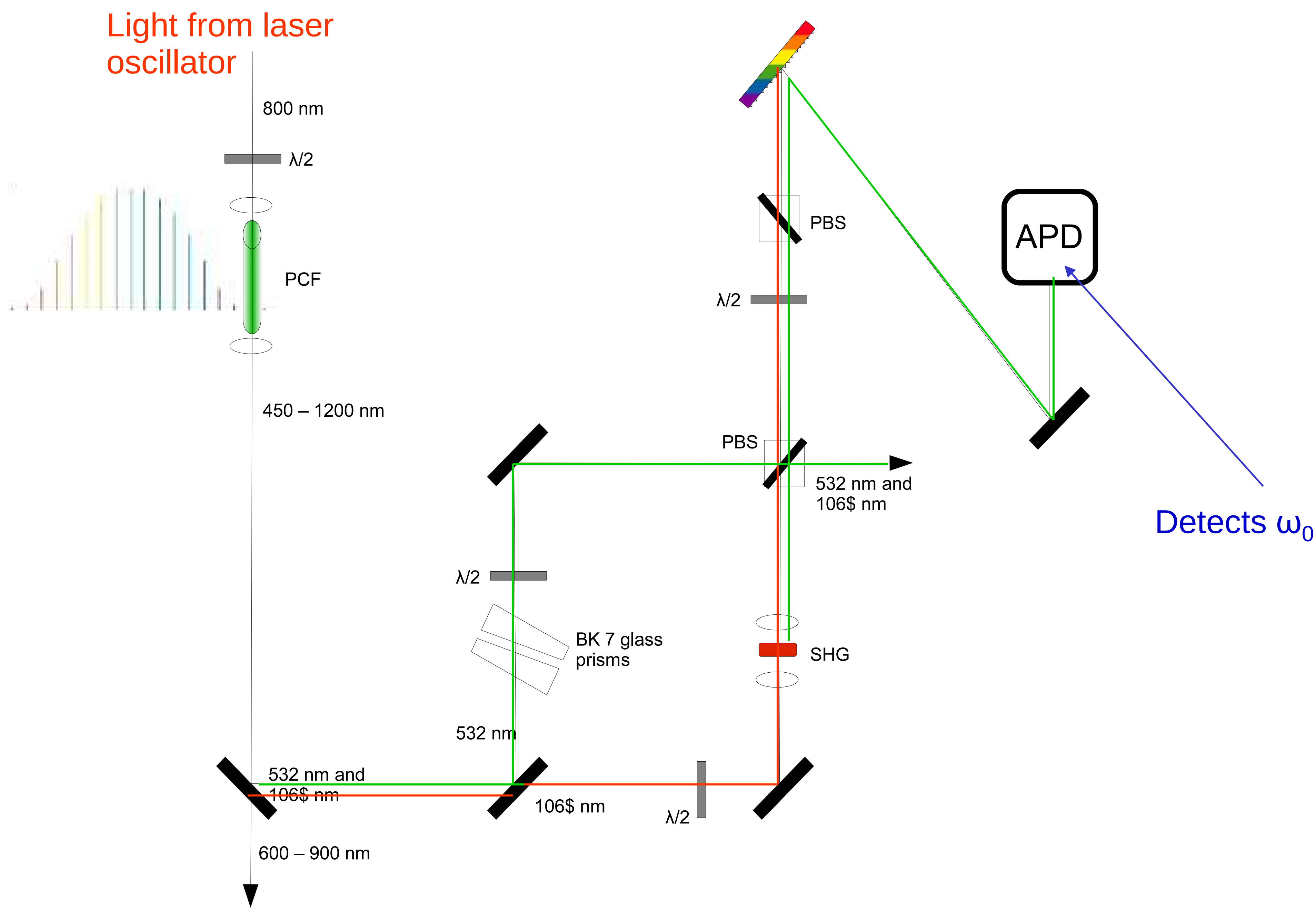
Beat them together:



www.menlosystems.com

$$2\omega_n - \omega_{2n} = 2(n\omega_r + \omega_0) - 2n\omega_r + \omega_0 = \omega_0$$

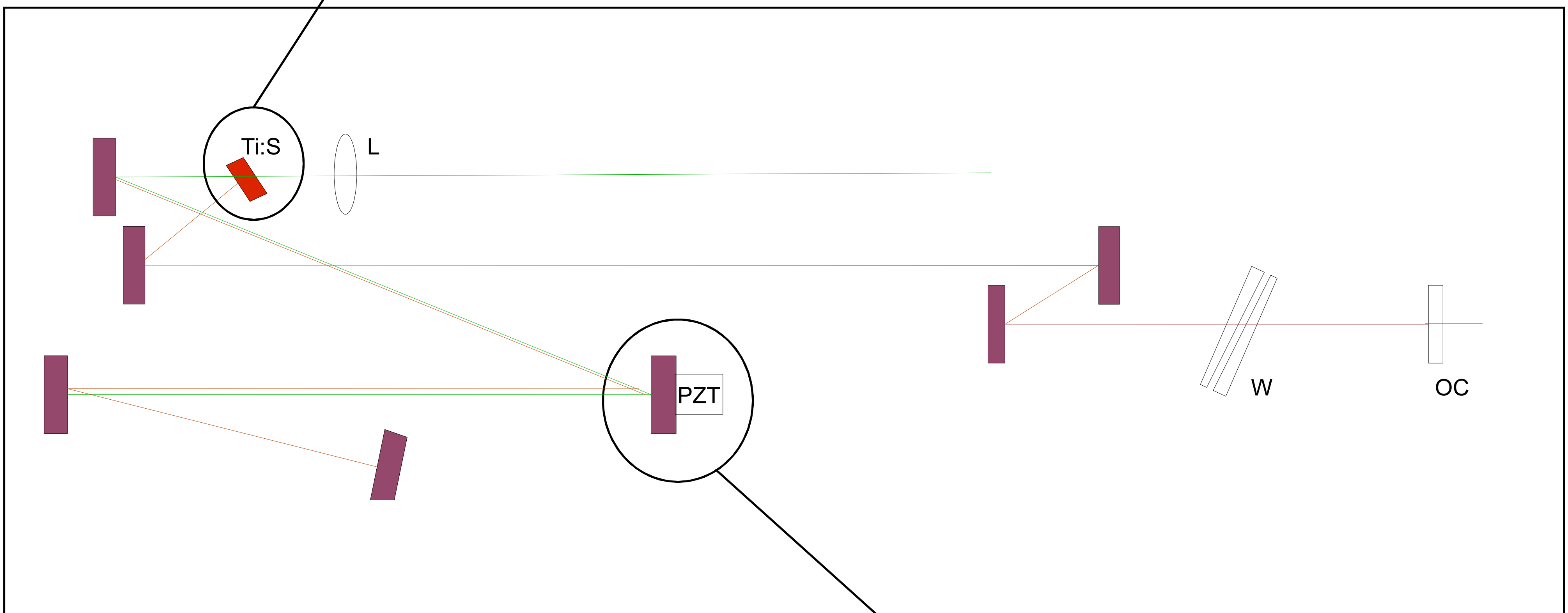
Stabilizing ω_r and ω_0 will stabilize frequency comb



Light from laser oscillator
800 nm
λ/2
PCF
450 – 1200 nm
600 – 900 nm
532 nm and 106$ nm
106$ nm
λ/2
532 nm
BK 7 glass prisms
λ/2
PBS
532 nm and 106$ nm
SHG
λ/2
PBS
APD
Detects ω0

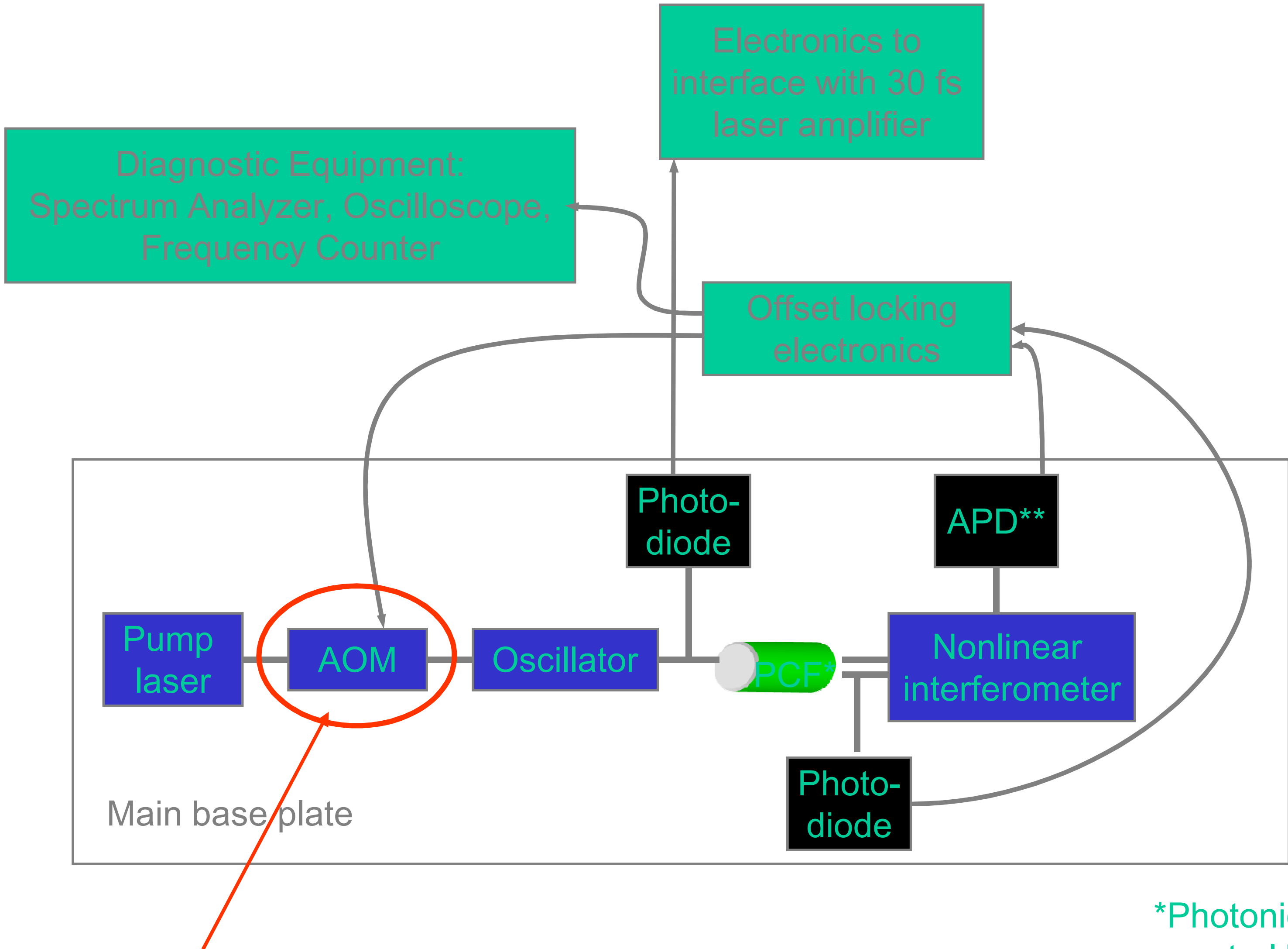
Stabilizing ω_0 and ω_r

$n=n(E) \Rightarrow$ control ω_0



control ω_r

Frequency Comb Block Diagram

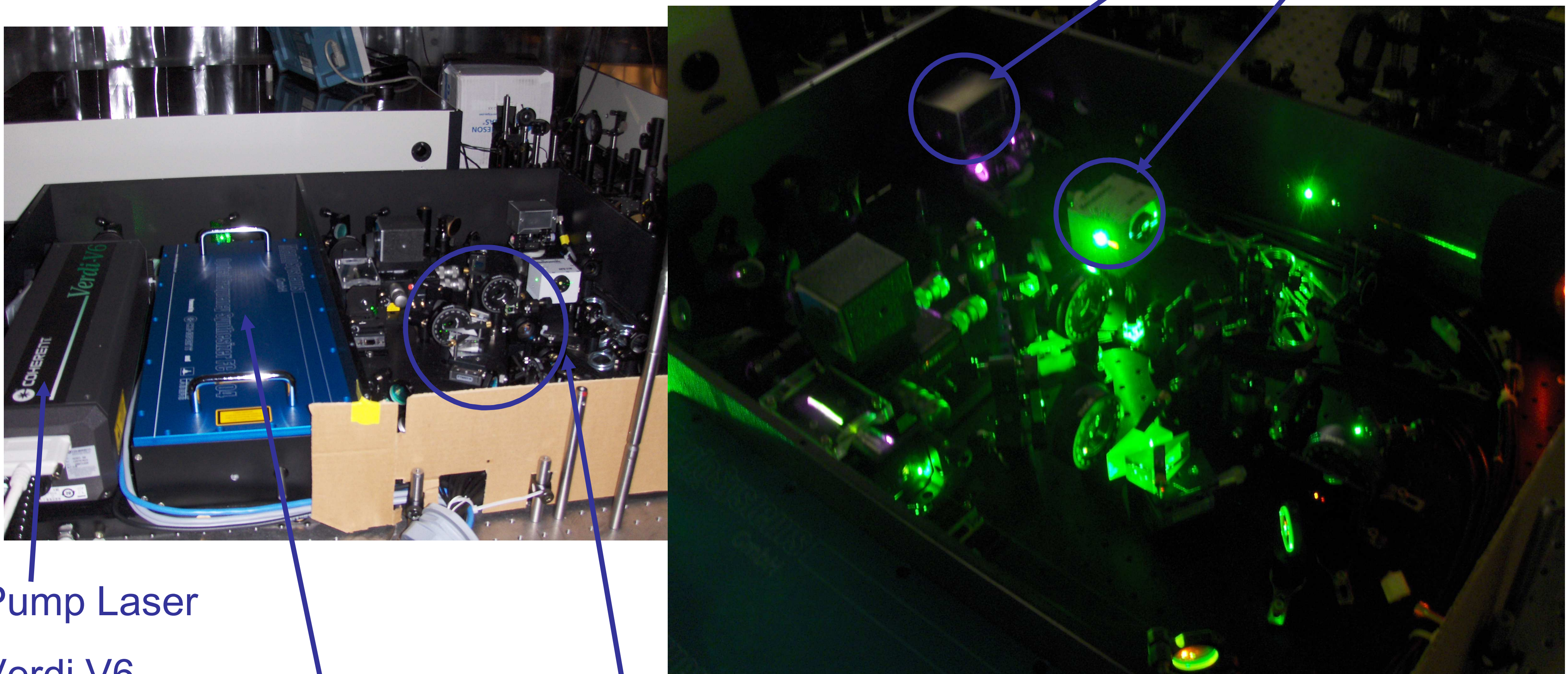


Sound wave creates “grating” and diffracts an amount of the light

*Photonic crystal fiber for spectral broadening

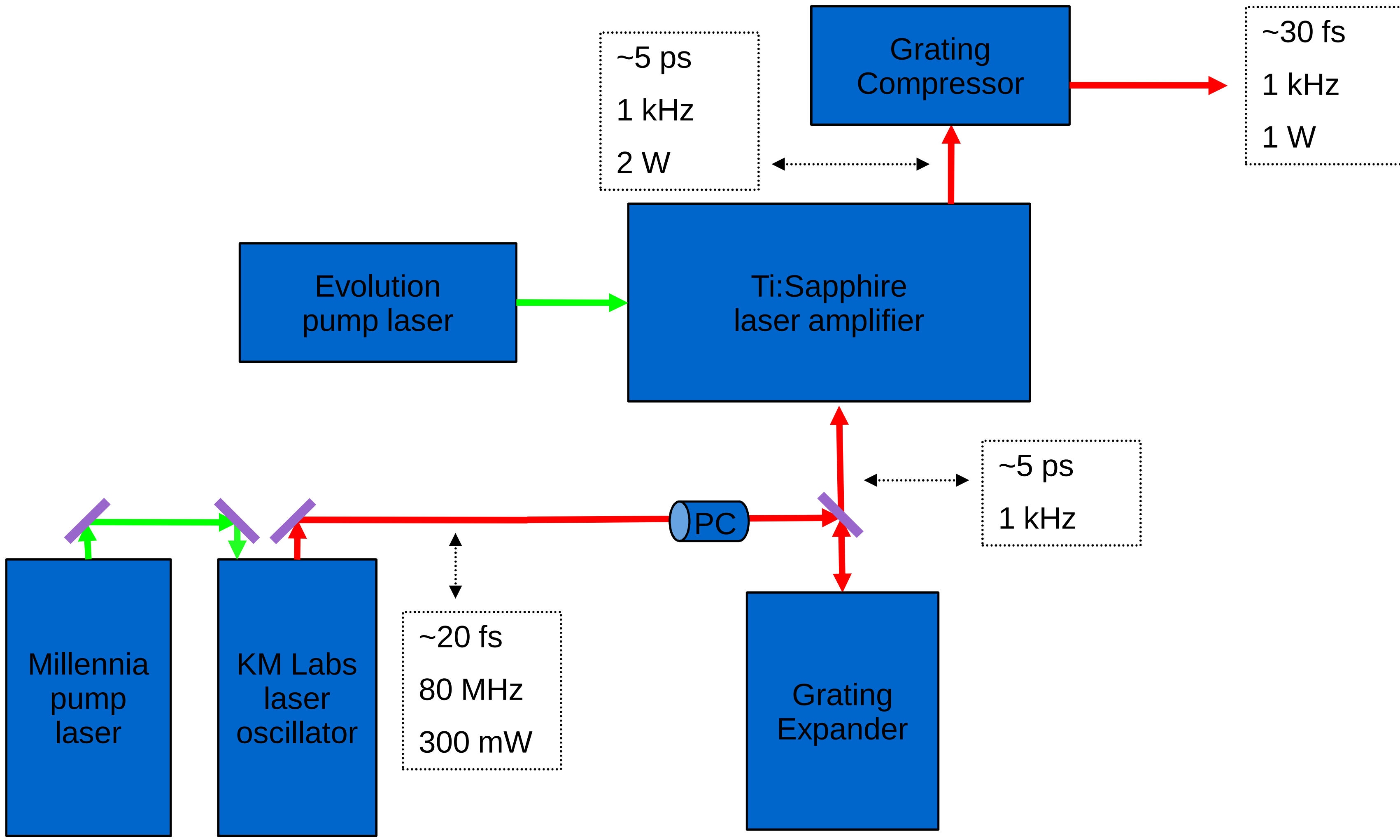
**Avalanche photodiode

FC8004 Optical Frequency Synthesizer (MenloSystems GmbH)

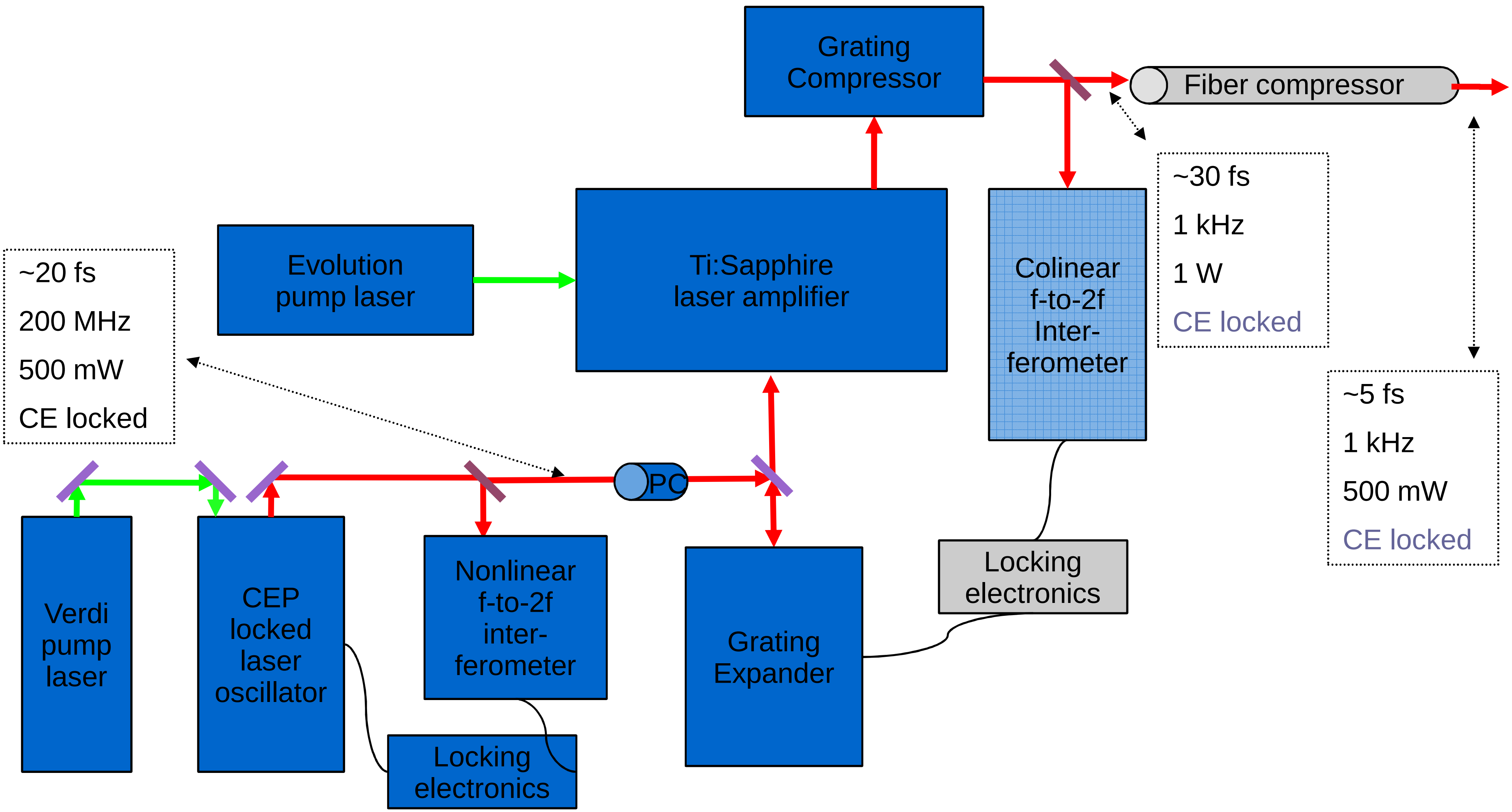


Conventional Laser Schematic

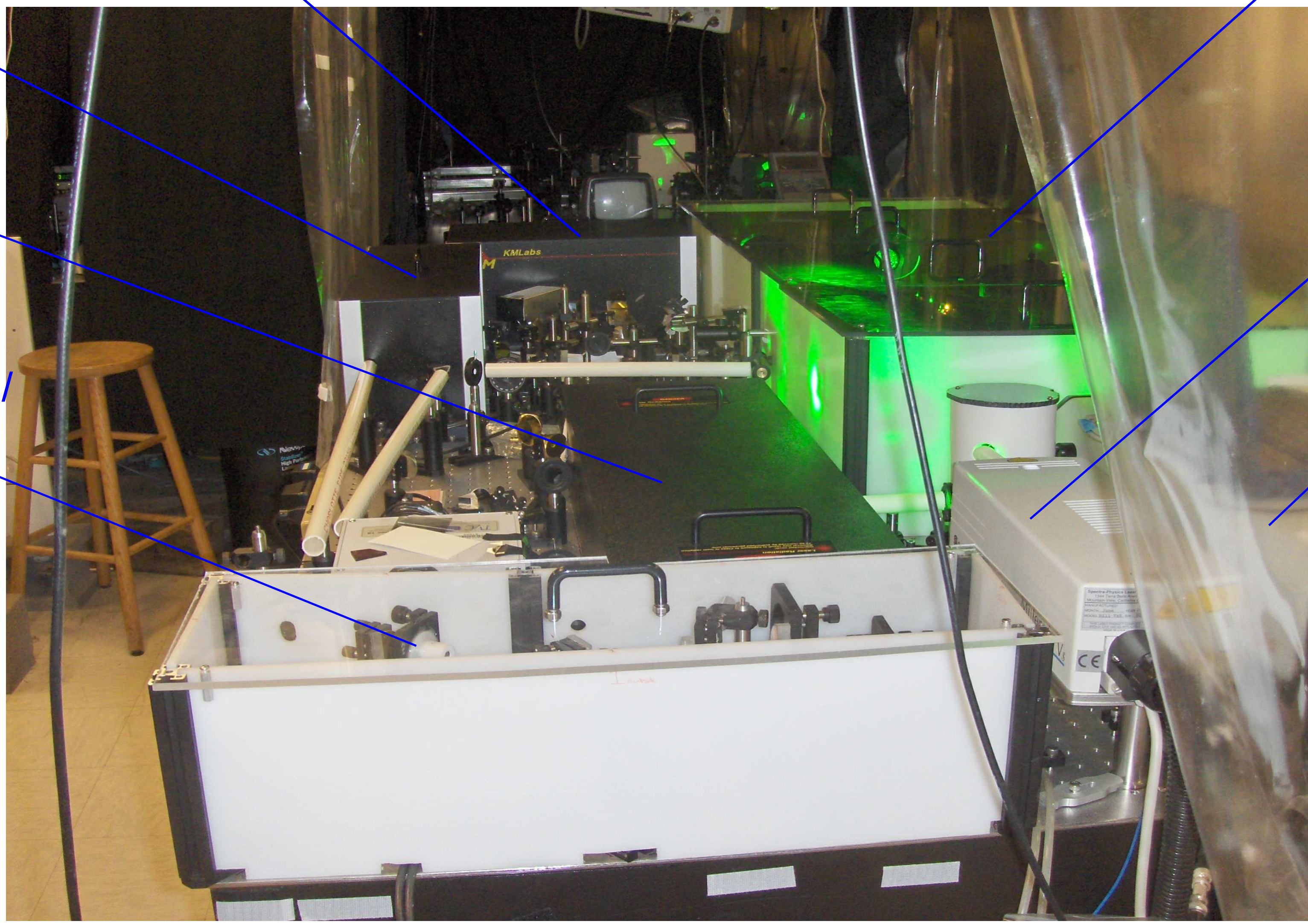
Chirped Pulse Amplification



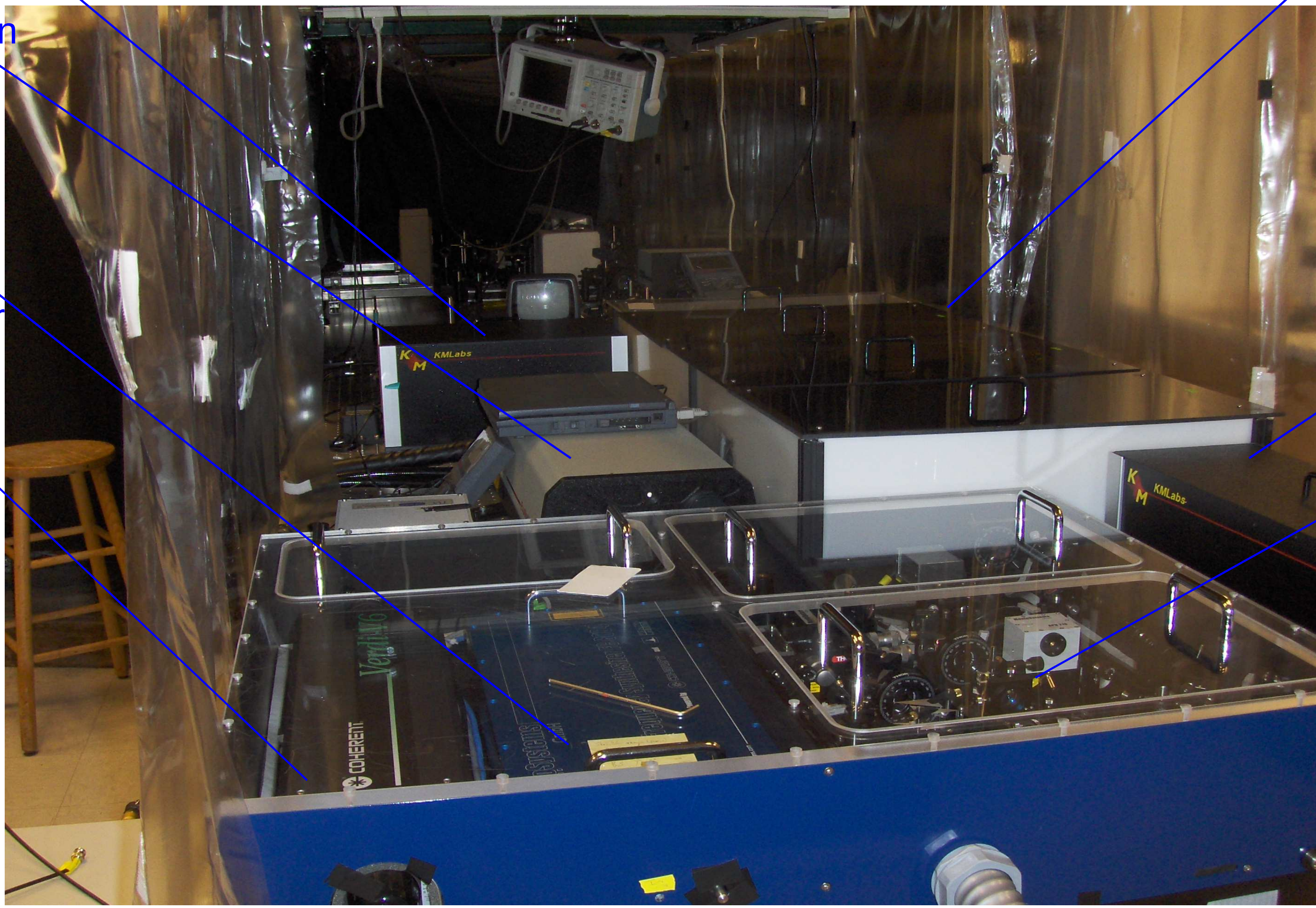
CE Phase-Locked Laser Schematic



Laser Rebuild – Former Setup



Laser Rebuild – New Setup

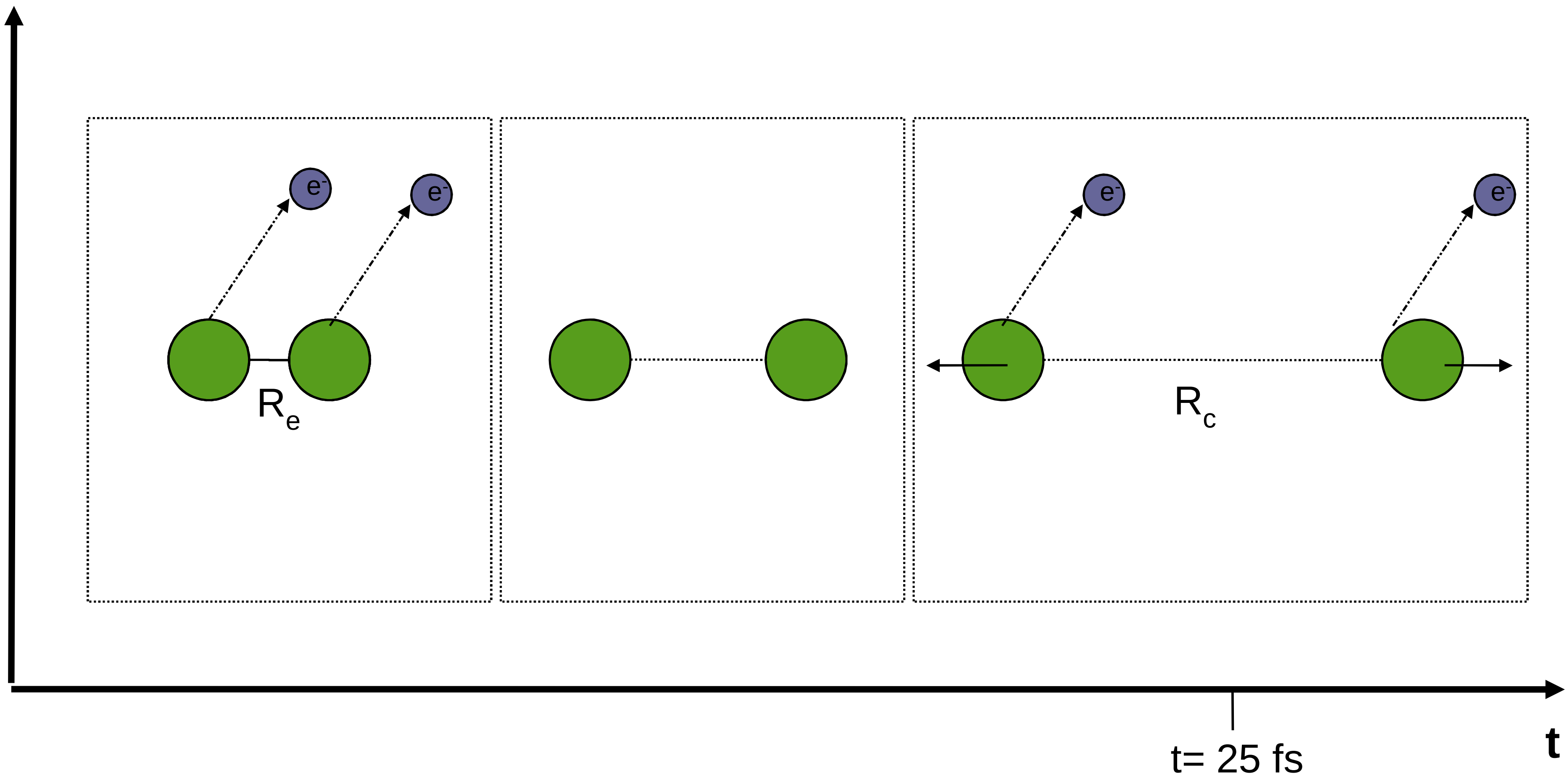


Current output

- Oscillator is phase locked
- ~1 W, 800 nm, 1kHz, ~30 fs pulse output (?)
- Amplifier is not yet phase locked
  - Working on colinear f-to-2f interferometer
    - Duration? Pulse shape?

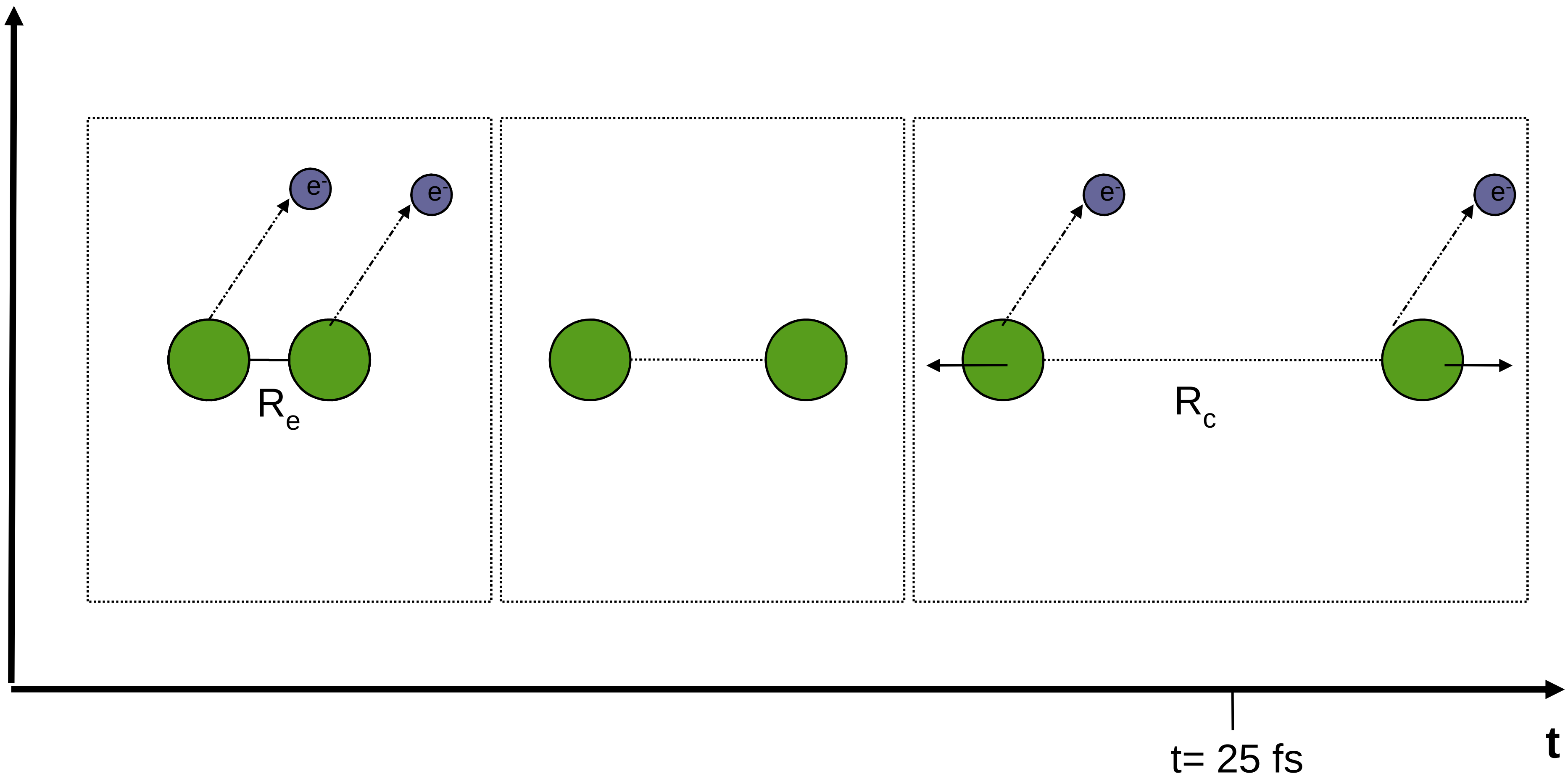
Plans for the Future

- Extend two-color studies
  - probe dissociation processes of O_2, CO_2
    - HOMO structure

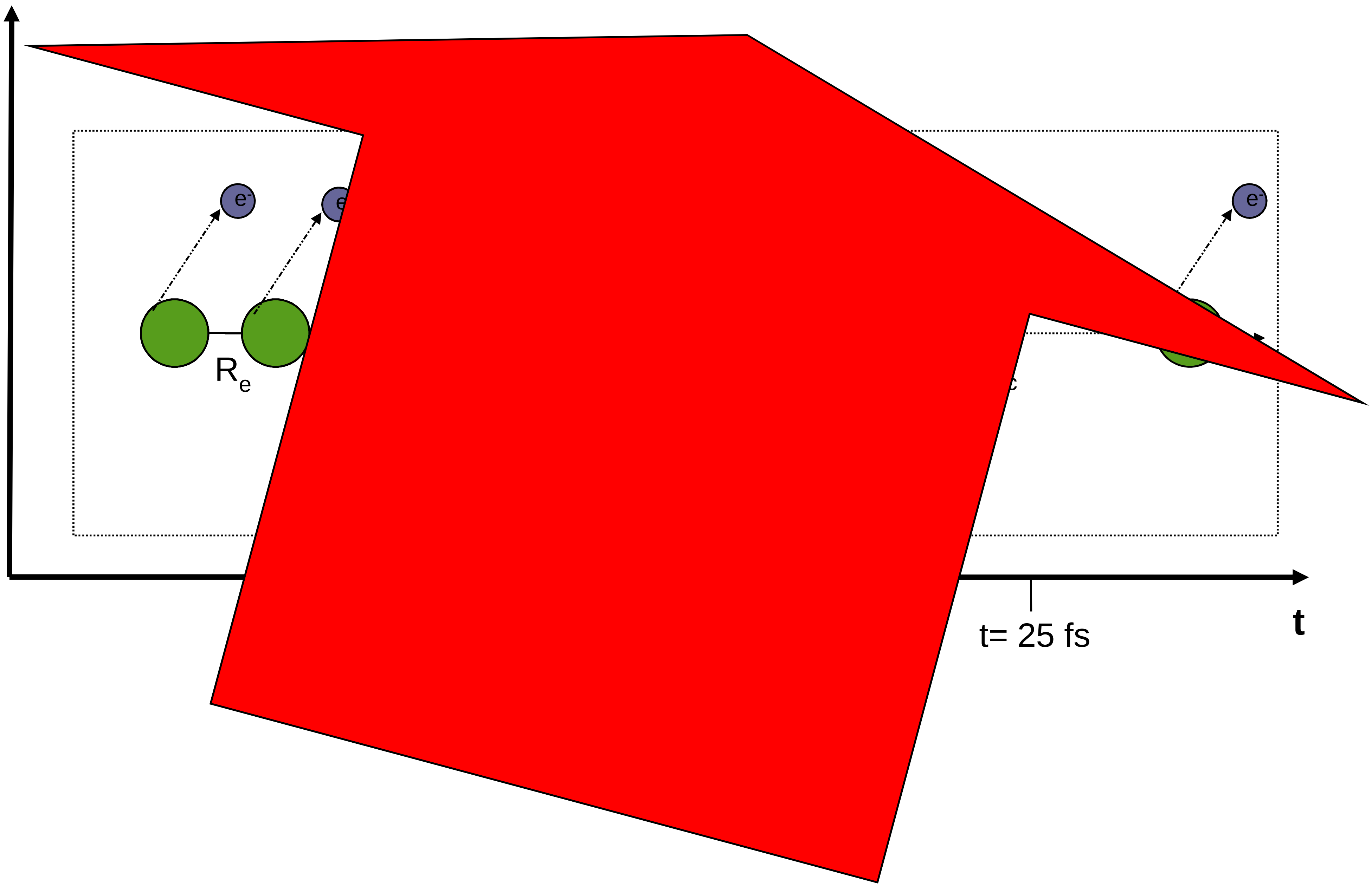

e-
e-
R
e
e-
e-
R
c
t= 25 fs
t

Plans for the Future

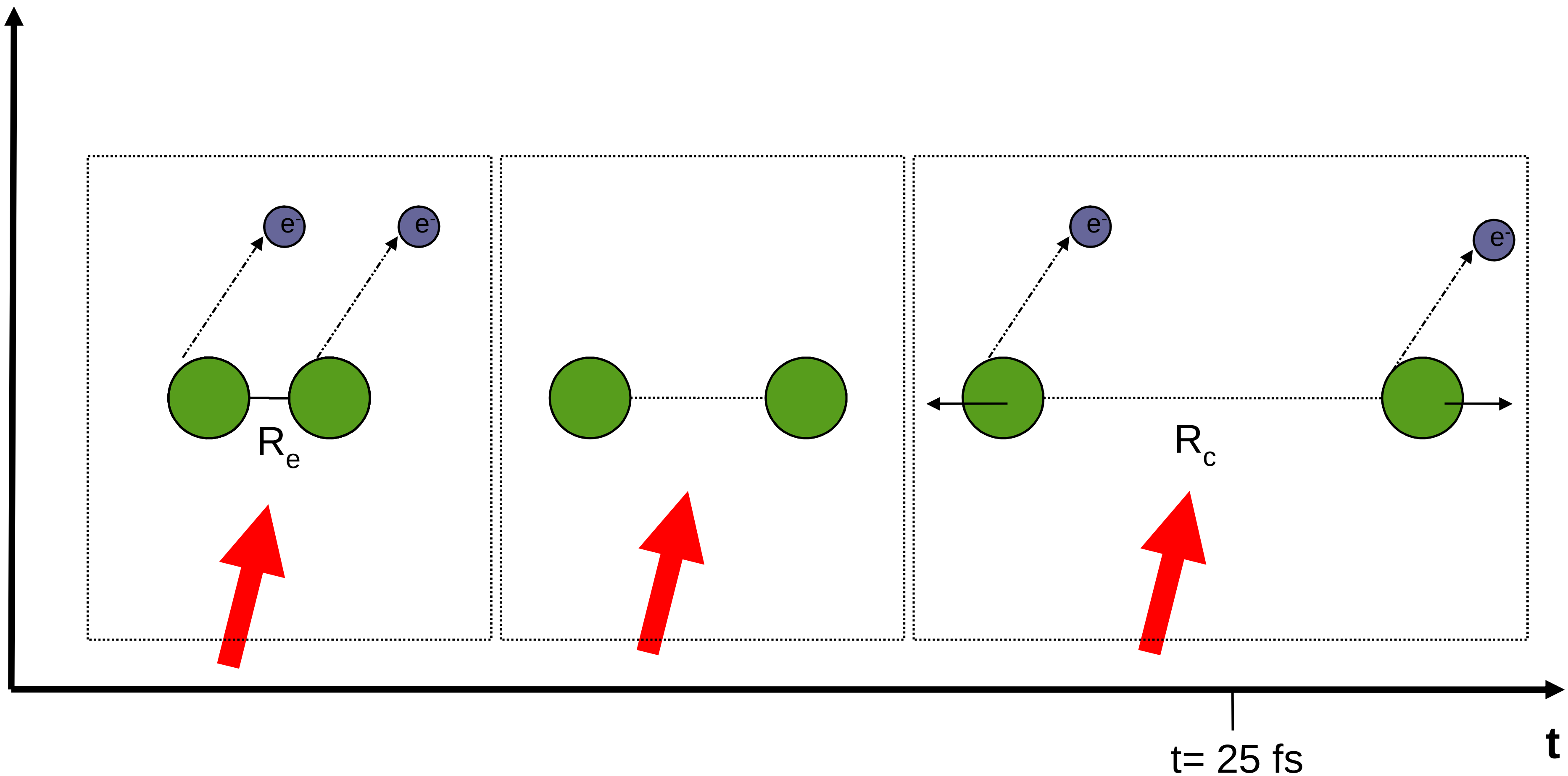
- Extend two-color studies
  - probe dissociation processes of O_2, CO_2
    - HOMO structure
- CEP to study same processes
  - Greater time resolution
  - Which step(s) is(are) phase-dependent
  - Way to determine CE phase



e-
e-
Re
e-
e-
Rc
t= 25 fs
t



e-
e-
e-
R
e
t= 25 fs
t


e-
e-
R$_e$
e-
e-
R$_c$
t= 25 fs
t

Acknowledgments

- Dr. Robert Jones
- Dr. Dan Pinkham
- Mary Kutteruf
- Dr. Brett Sickmiller