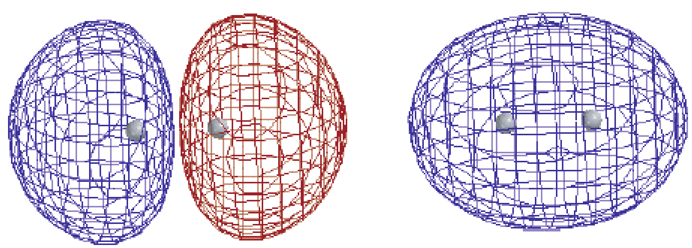
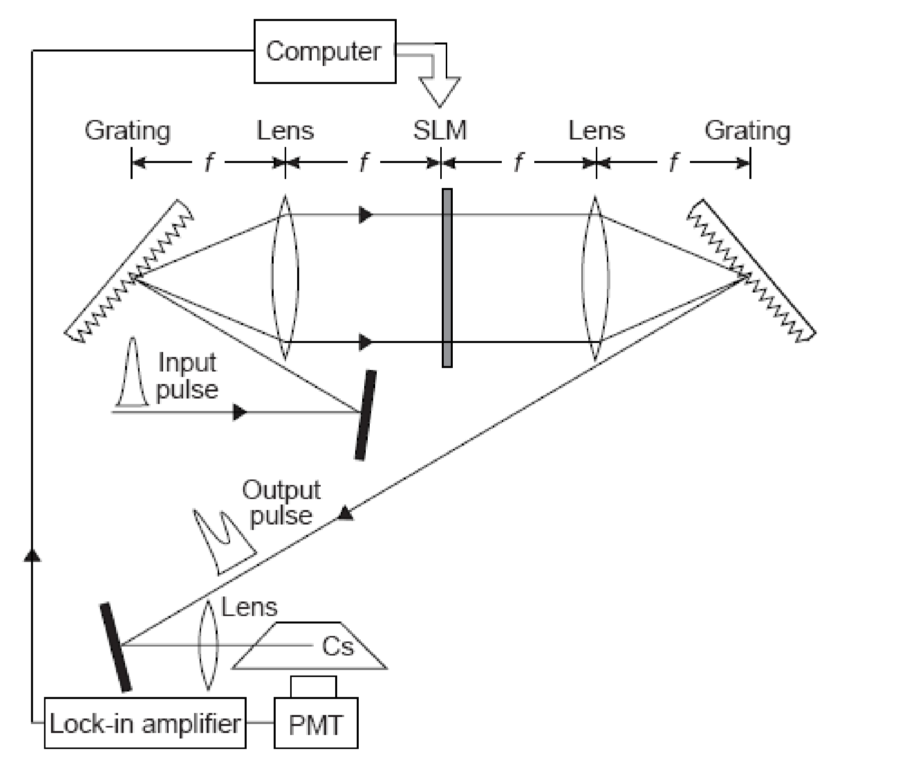
Complex Dissociation of Simple Molecules

Strong field photodissociation of H_2^+



Vaibhav S. Prabhudesai, Adi Natan, Uri Lev, Barry Bruner, Adi Diner, Oded Heber, Daniel Strasser, Dirk Schwalm, Itzik Ben-Itzhak, J. J. Hua, B. Esry
Daniel Zajfman and Yaron Silberberg

Simple control…



Site
ials,
om
with
arge
etal,
ups
ous
but
ili-
hey
matrix[28]. Carbonaceous chondrites
t of observed meteorite falls[29], but
onent of the asteroid belt, as most of
have characteristics typical of CM

l, H. V. Extraterrestrial cause for the Cretaceous-Tertiary
possible Cretaceous/Tertiary boundary impact crater on

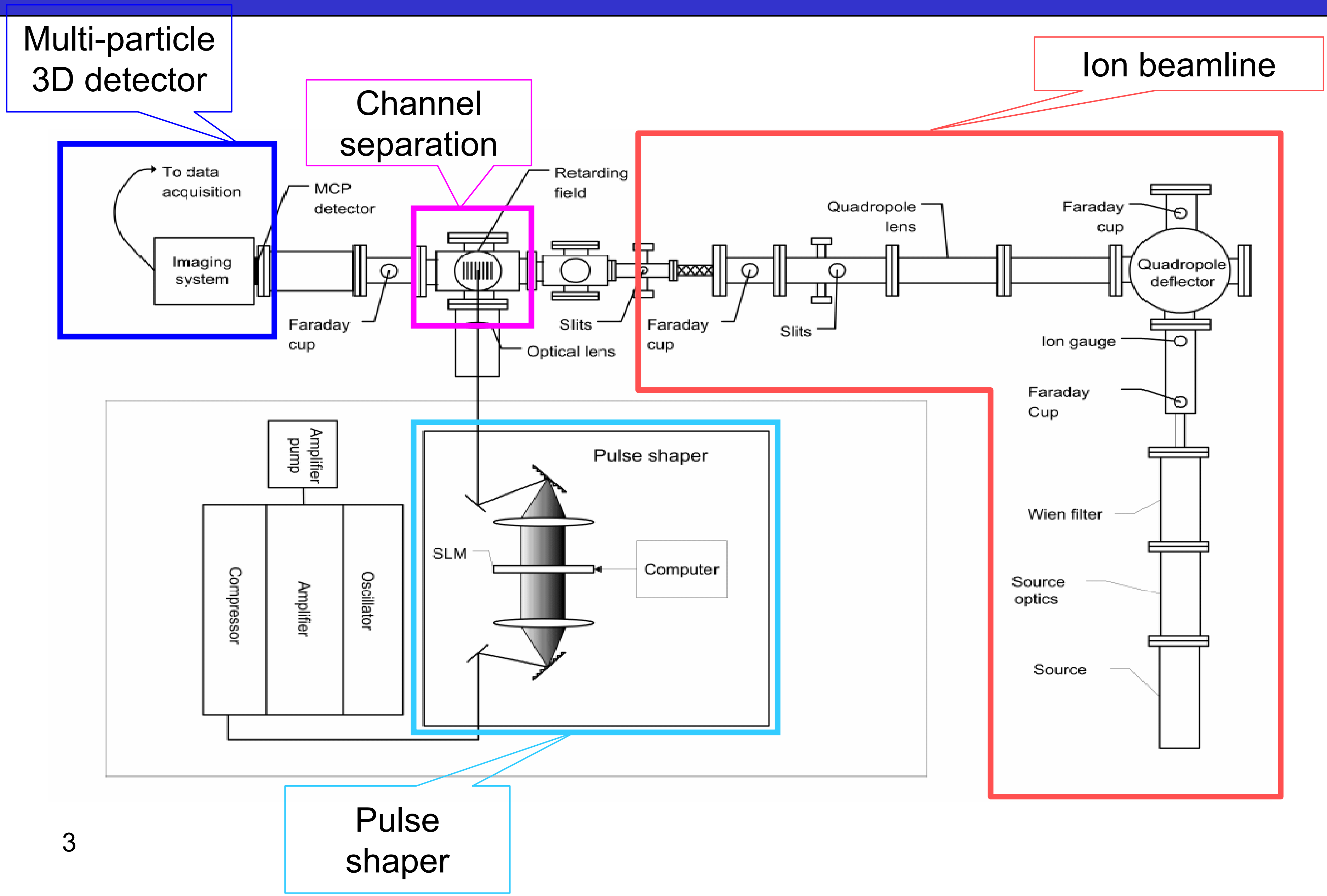
Coherent quantum control of two-photon transitions by a femtosecond laser pulse

Doron Meshulach & Yaron Silberberg

Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel

Coherent quantum control[1–3] has attracted interest as a means to influence the outcome of a quantum-mechanical interaction. In principle, the quantum system can be steered towards a desired state by its interaction with light. For example, in photoinduced transitions between atomic energy levels, quantum interference effects can lead to enhancement or cancellation of the total transition probability. The interference depends on the spectral

Experimental setup



Motivation: why photodissociation of H_2^+?

- H_2^+ is the simplest molecular system, still it reveals complex dynamical characteristics in strong fields.

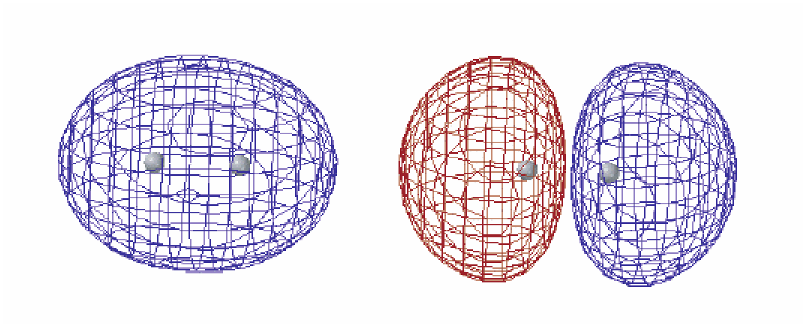
- How pulse shaping affects photodissociation?

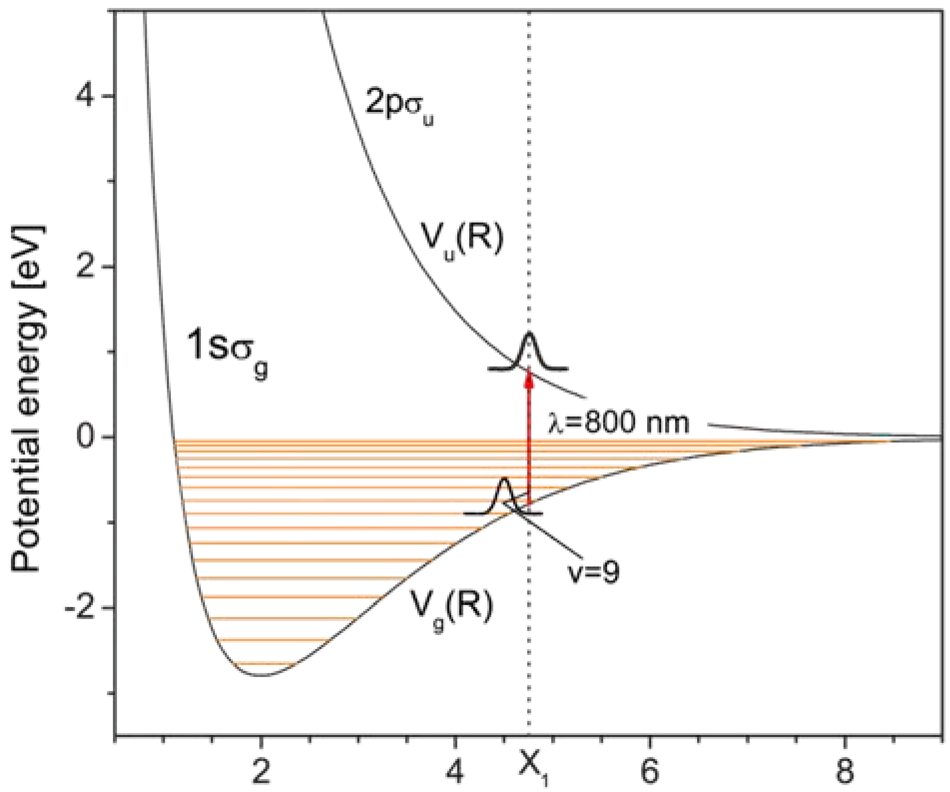
K. Sändig, H. Figger, and T.W. Hänsch
Physical Review Letters, **85**, 4876-4879 (2000)

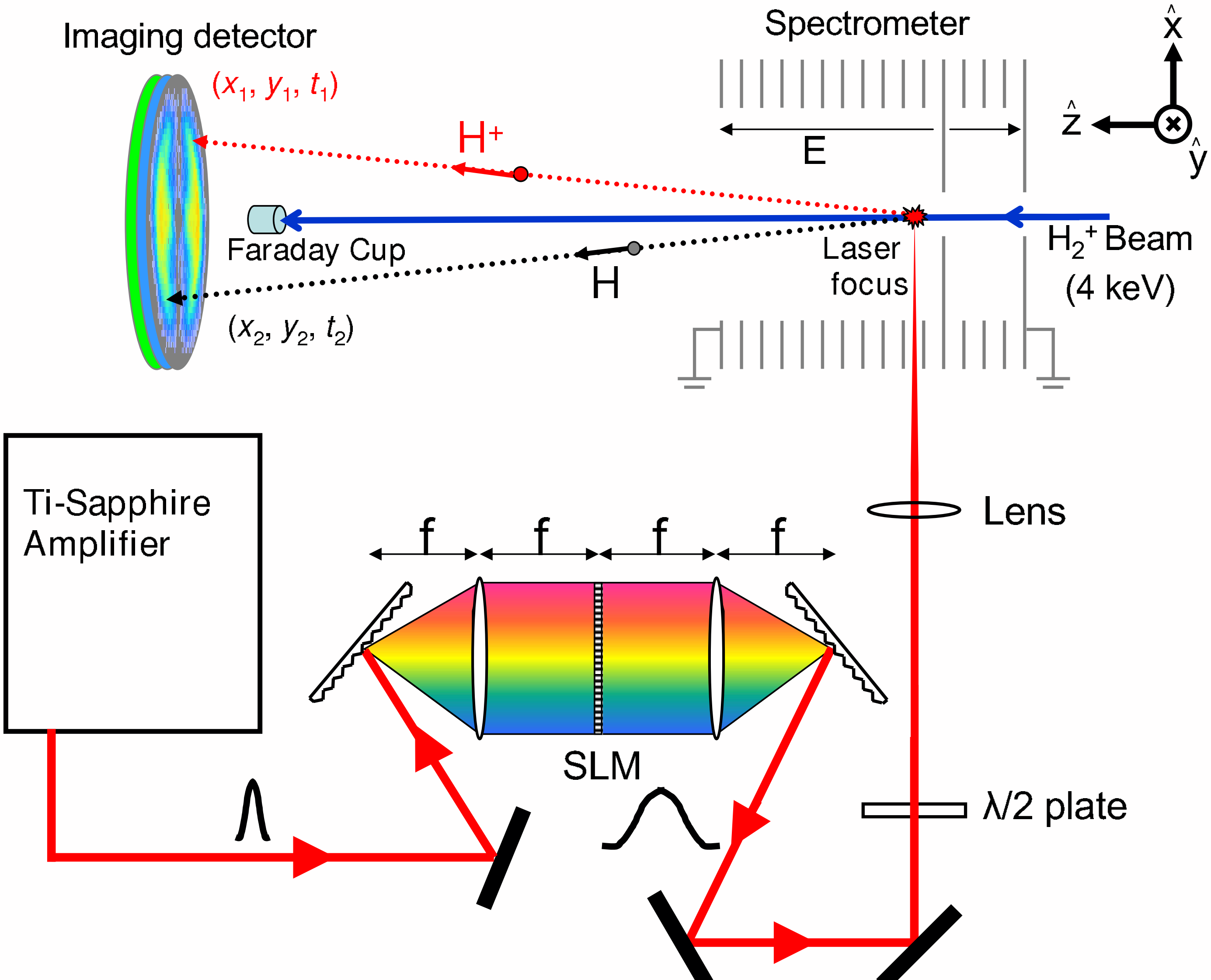
D. Pavicic, A. Kiess, T. W. Hänsch, and H. Figger
Physical Review Letters, **94,** 163002 (2005)

P. Q. Wang, A. M. Sayler, K. D. Carnes, J. F. Xia, M. A. Smith, B. D. Esry, I. Ben-Itzhak
Phys. Rev. A **74**, 43411 (2006)

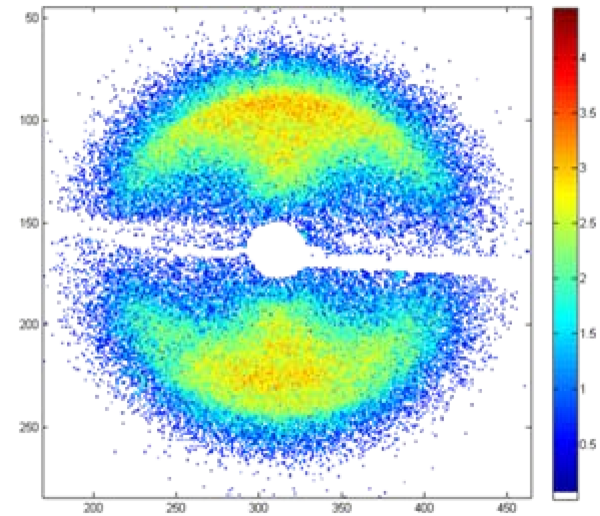
Photodissociation of H_2^+



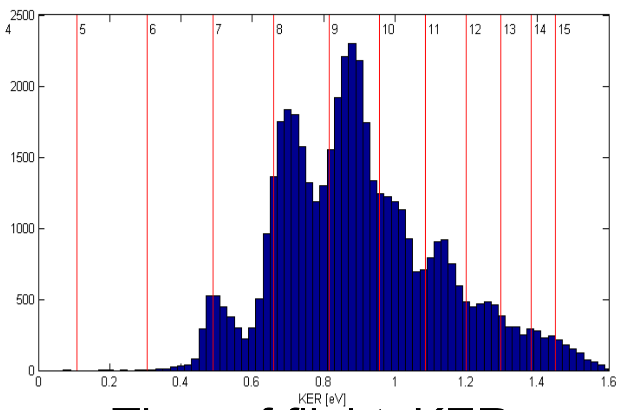



Imaging detector
(x1, y1, t1)
H+
Spectrometer
E
x
z
y
Faraday Cup
Laser focus
H2+ Beam (4 keV)
H
(x2, y2, t2)
Ti-Sapphire Amplifier
f f f f
Lens
SLM
λ/2 plate

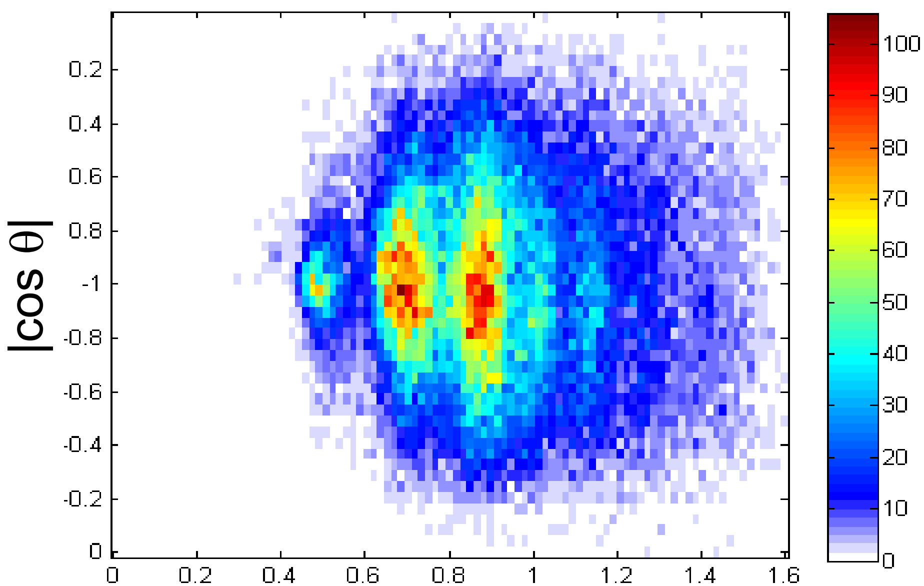
Typical Experimental Data



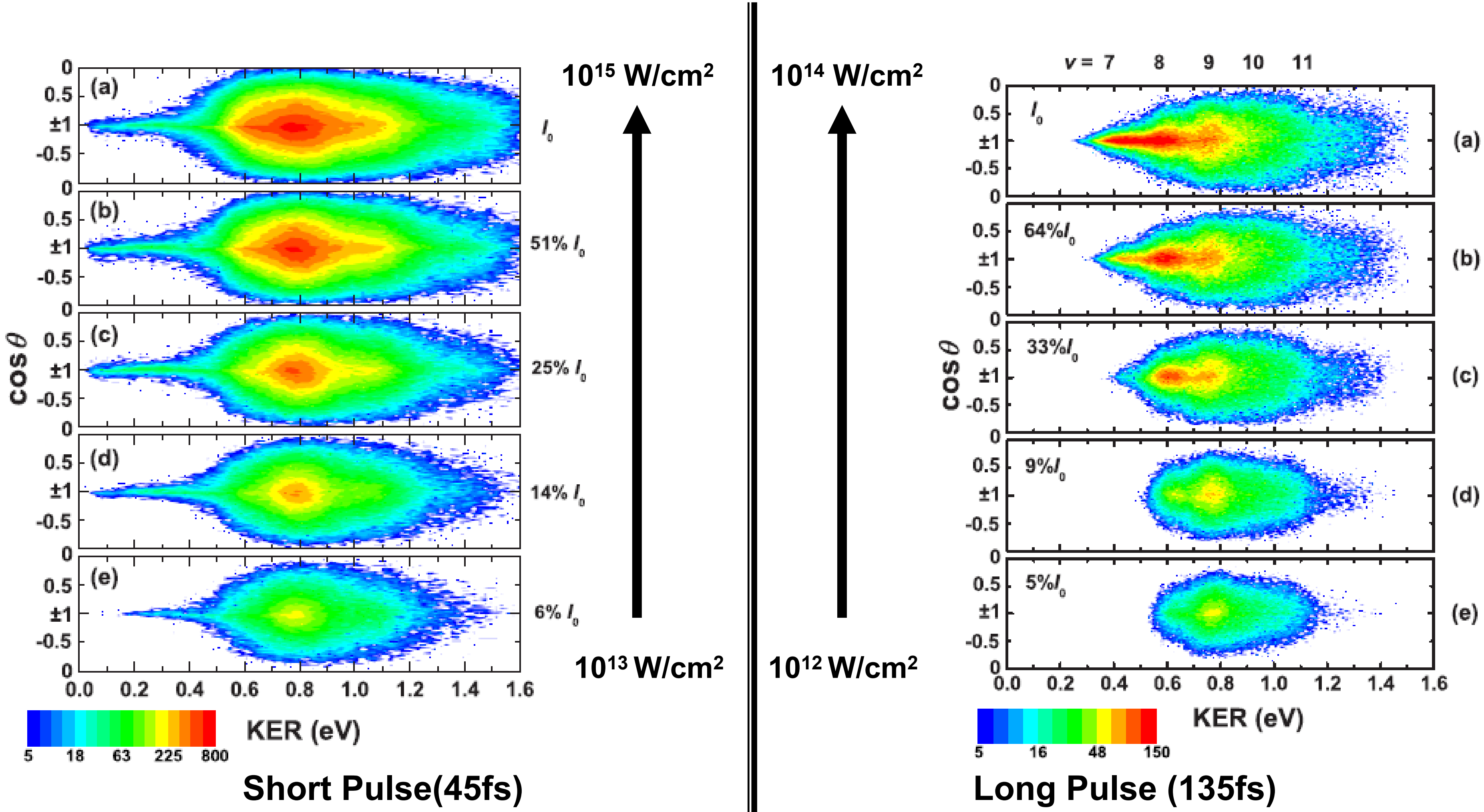
CCD: 2D momentum map



Time of flight: KER

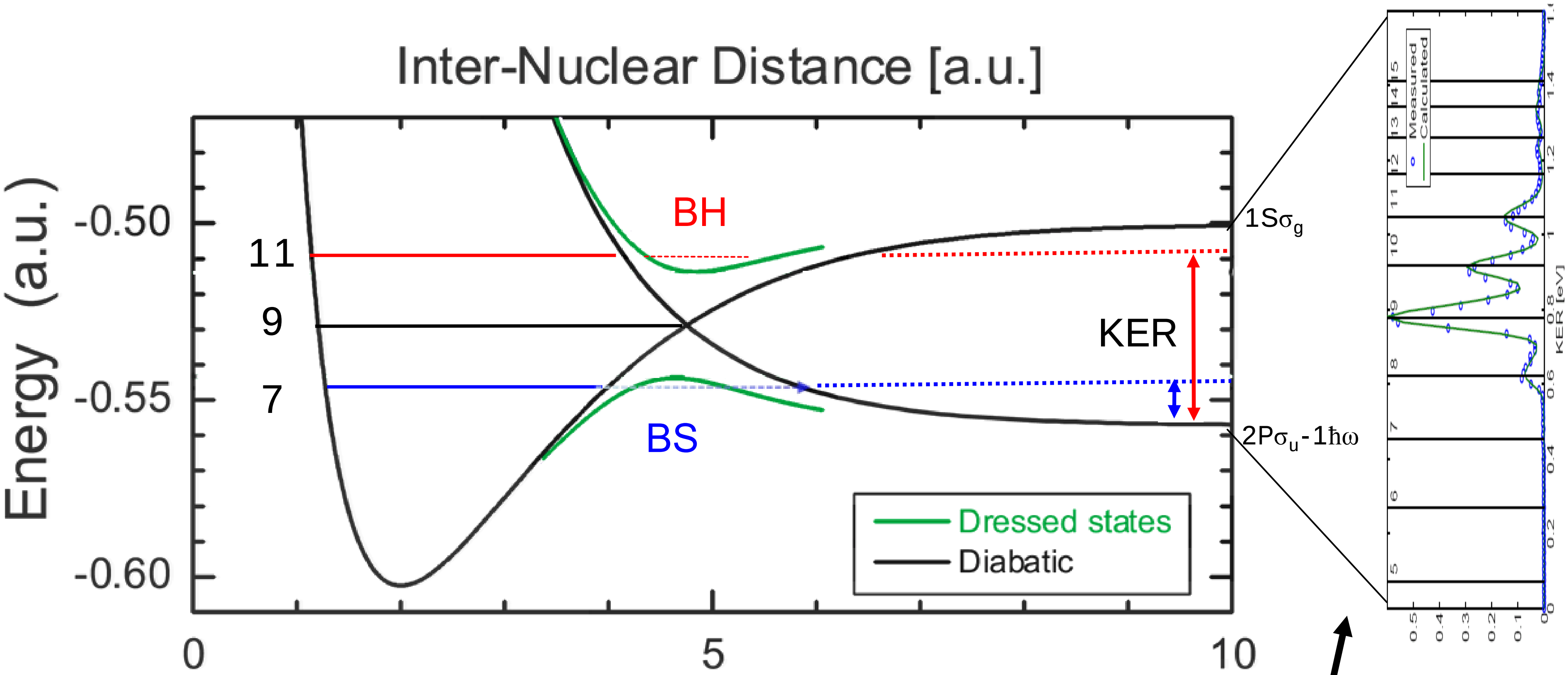


Previous studies…



WANG et al. Phys Rev A 74, 043411 (2006)

Strong field photodissociation dynamics in H_2^+

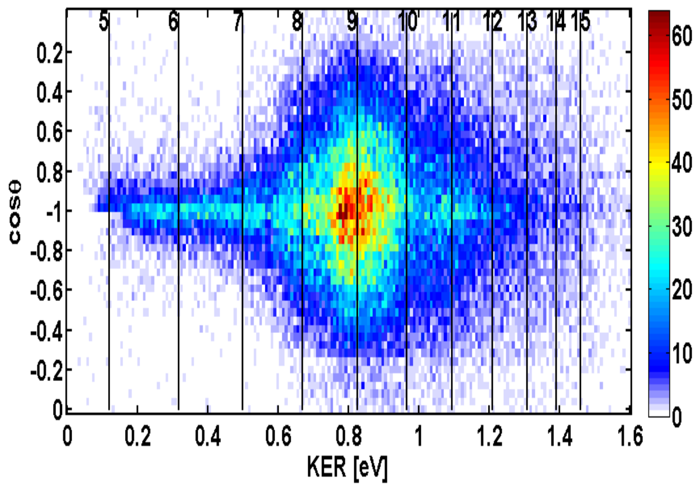


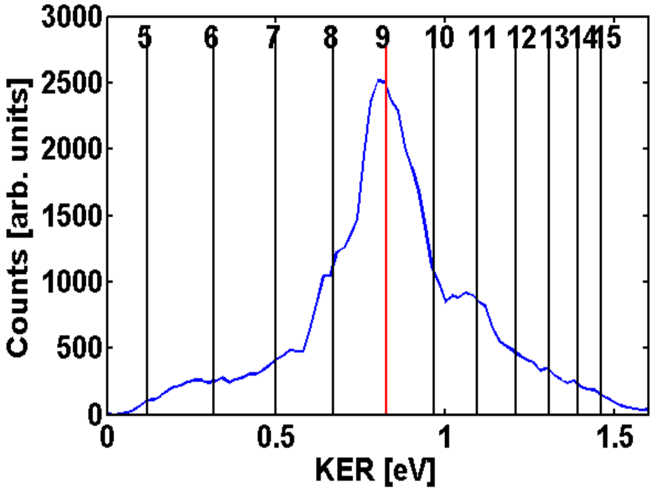
- Bond Softening (BS)
- Bond Hardening (BH)

The measured KER

Pulse length: 35fs

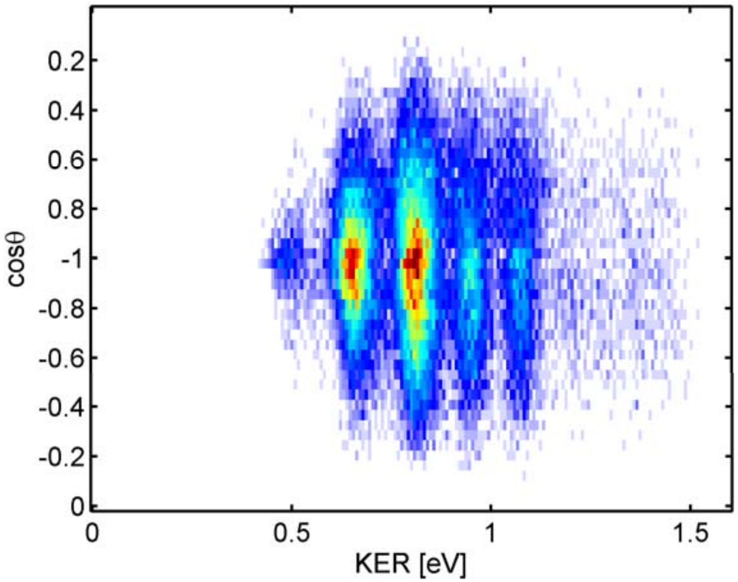
Peak Power: 10^{14} W/cm^2

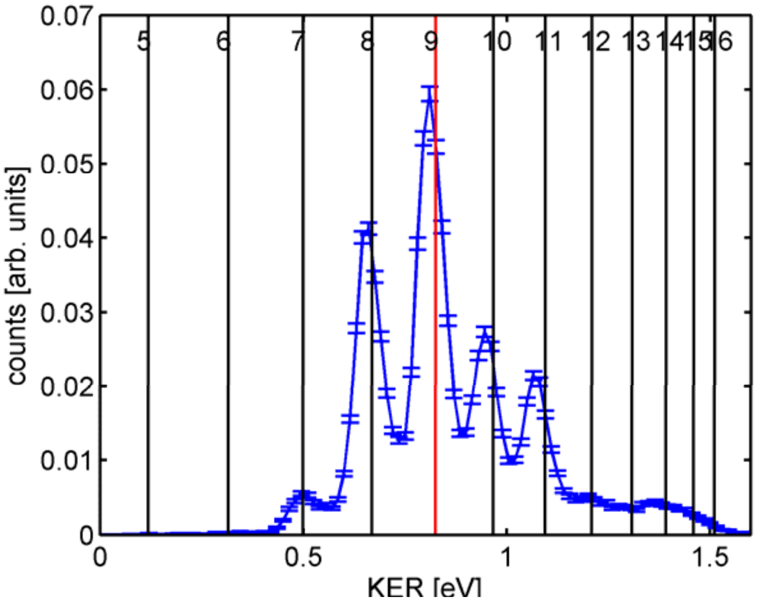




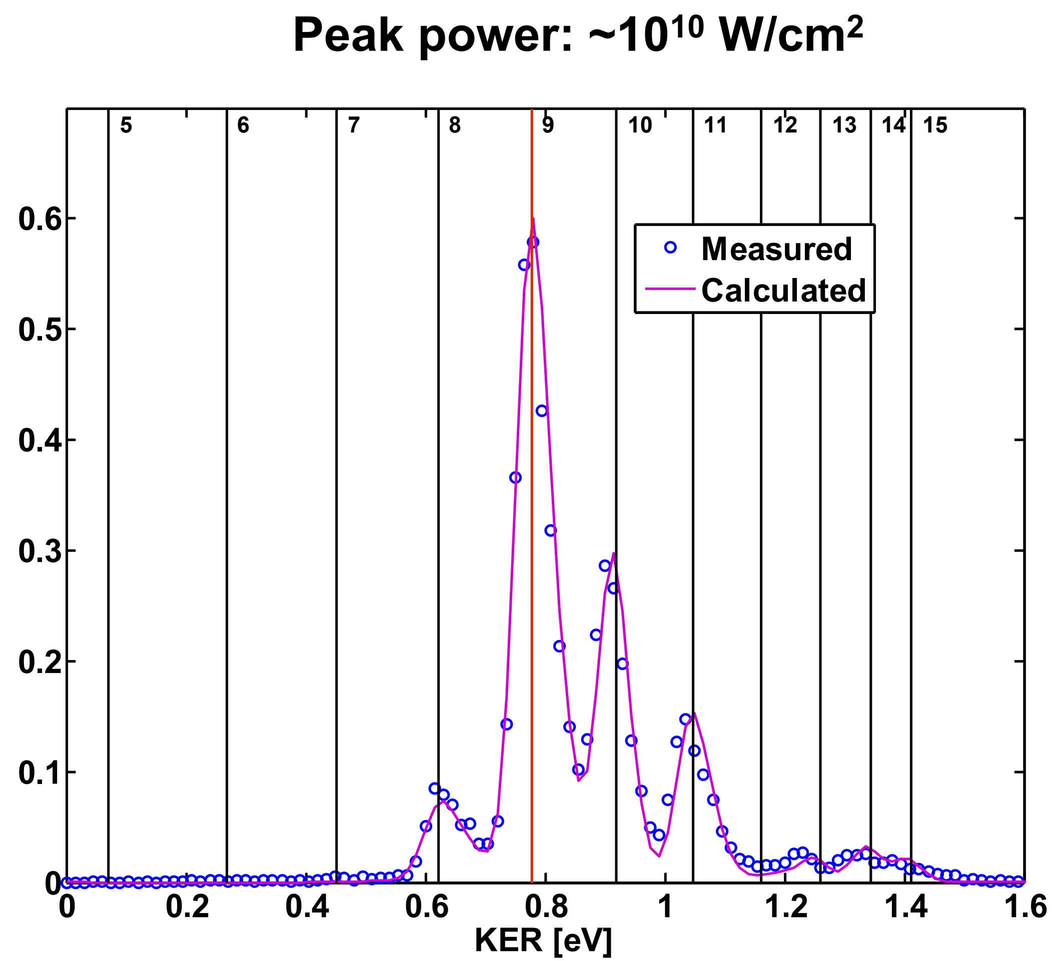
KER spectrum for a narrow band strong field

Peak power: 3×10^{12} W/cm^2
Pulse length: 120fs (transform limited)





Weak-field KER spectrum



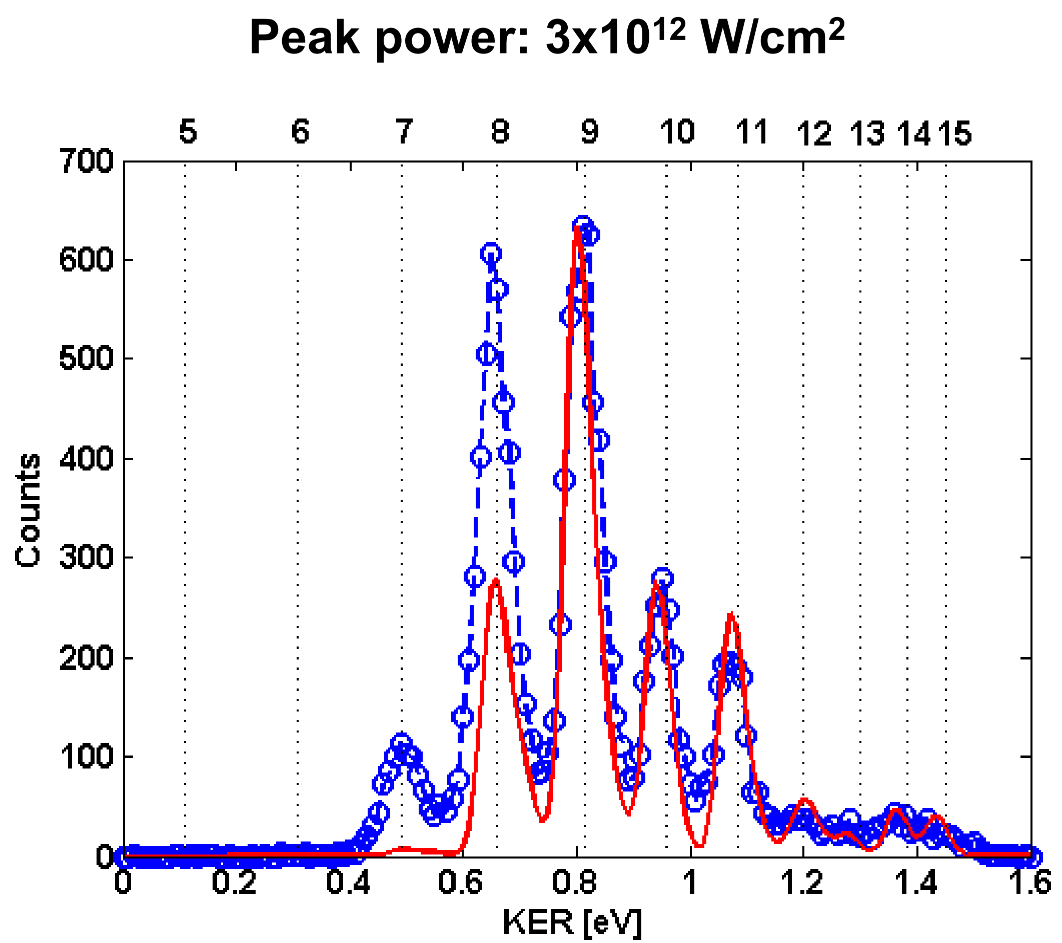
“weak field” model

We sum over all the contributions for all the frequencies sequentially for each (v,j) state with the constraint of a finite population.

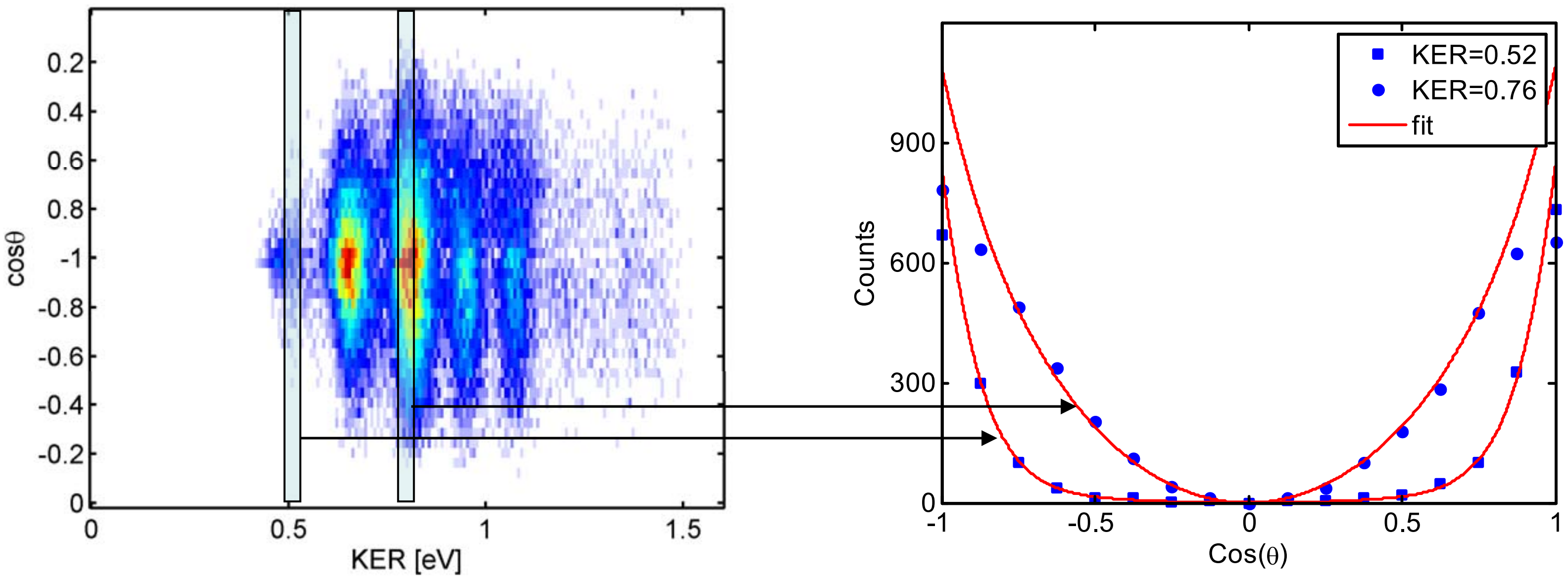
$$R_{measured}(\lambda_i, v, j) = N_{photons/pulse}(\lambda) \cdot \sigma(v, j) \cdot \rho \cdot L \cdot P(v, j) \left[1 - \frac{\sum_{\lambda < \lambda_i} R(\lambda, v, j)}{\rho \cdot V_{\text{int}} \cdot P(v, j)} \right]$$

The calculation parameters are as close to the experimental parameters as possible.

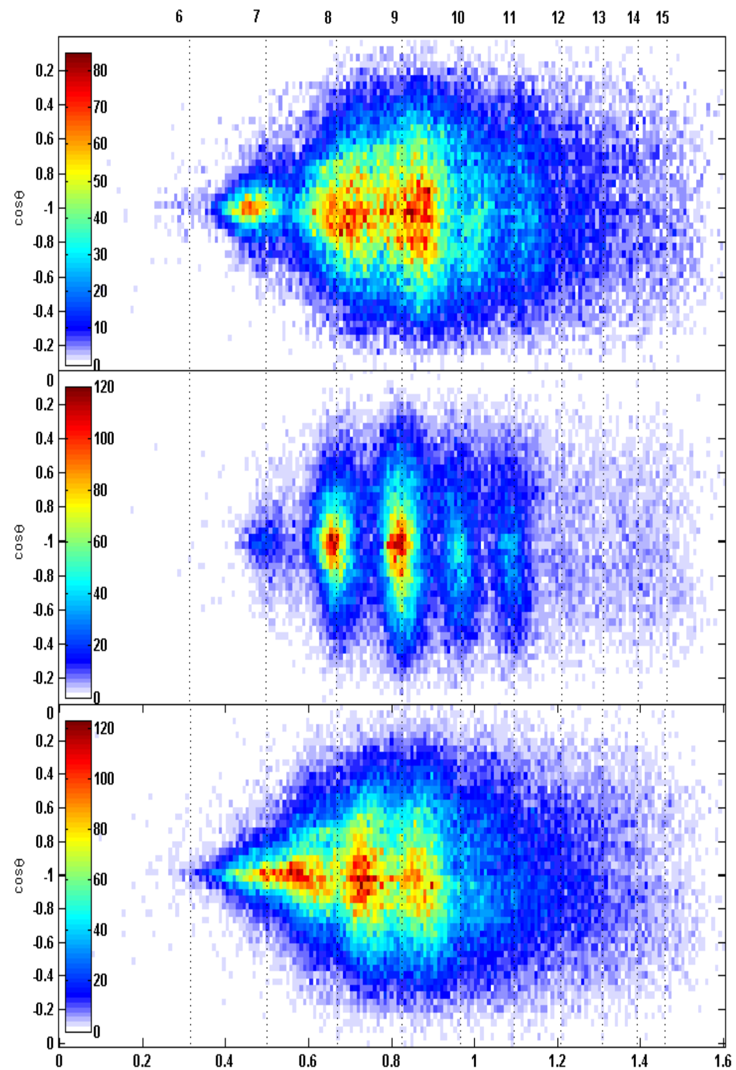
Intermediate-field KER spectrum



Angular distributions



The effect of chirp – 120 fs pulses



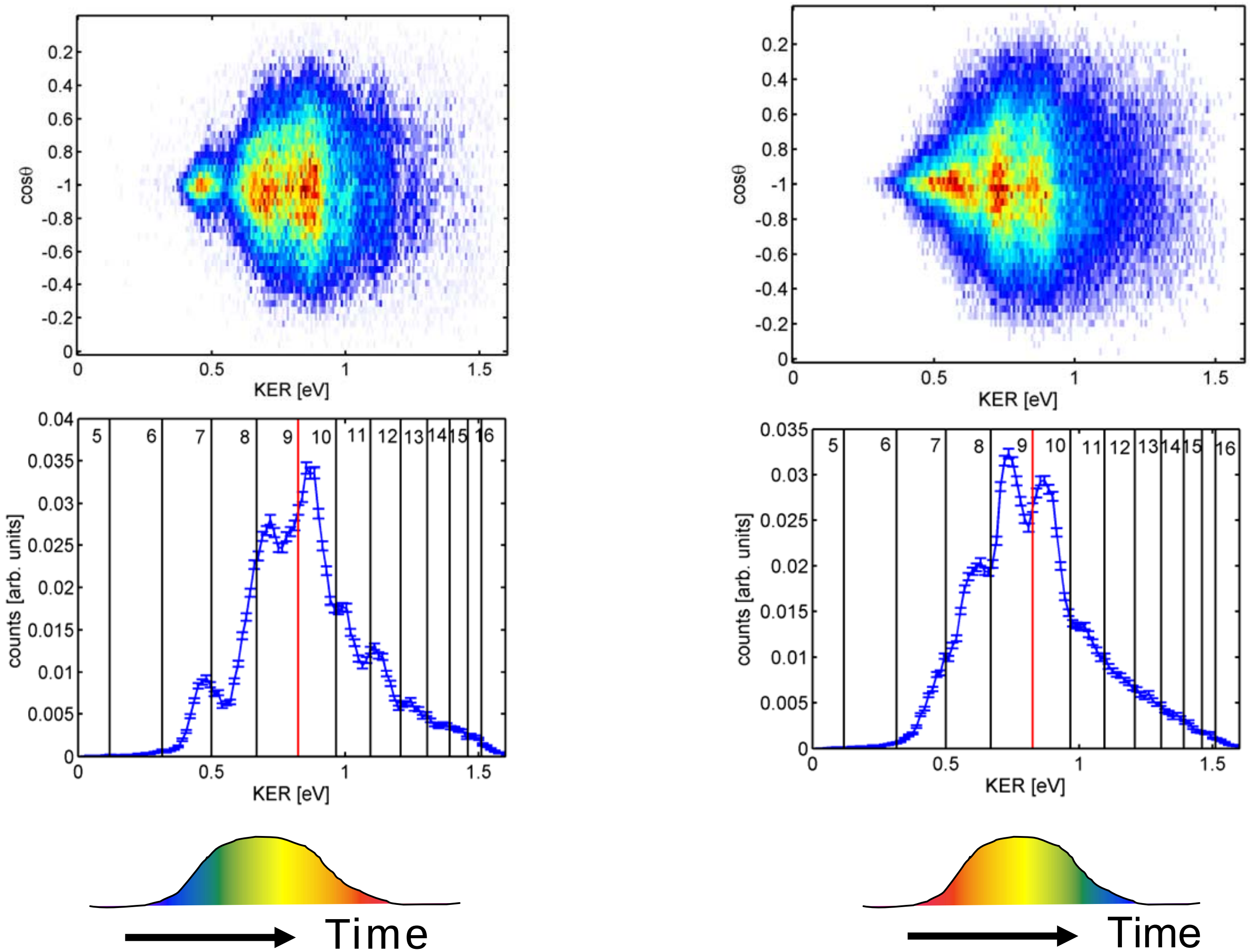
Negative Chirp

No Chirp (Narrow band)

Positive Chirp

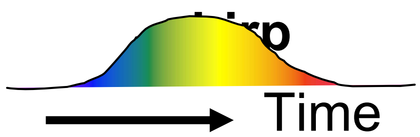
Negative v/s positive chirp

Peak Power: 8 x 10^{12} W/cm^2
Pulse length: 120fs

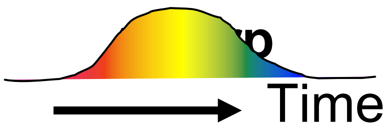


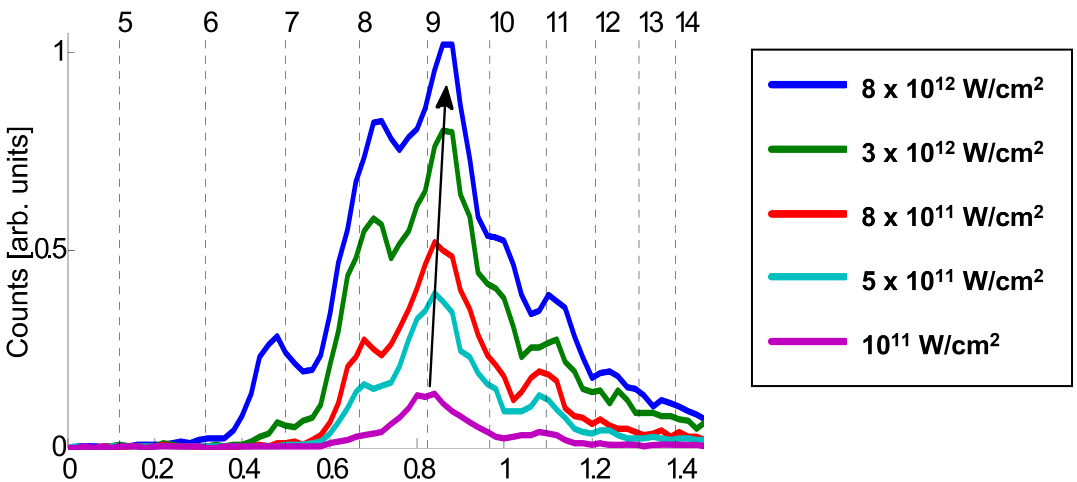
chirped pulses levels shift with intensity

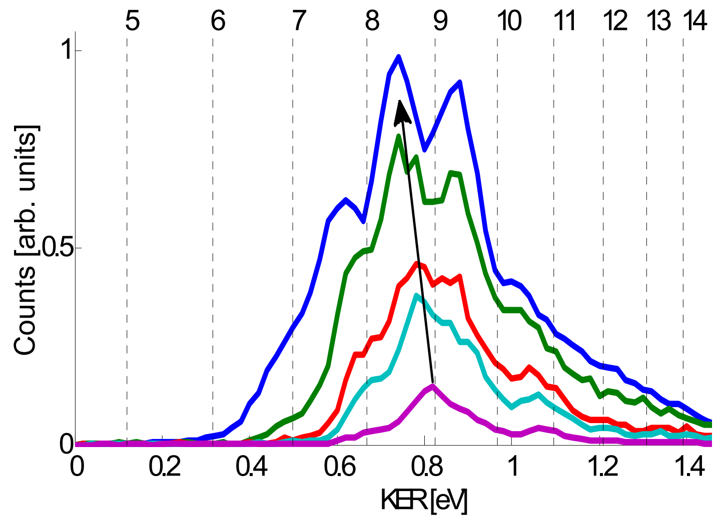
120 fs Negative



120 fs Positive

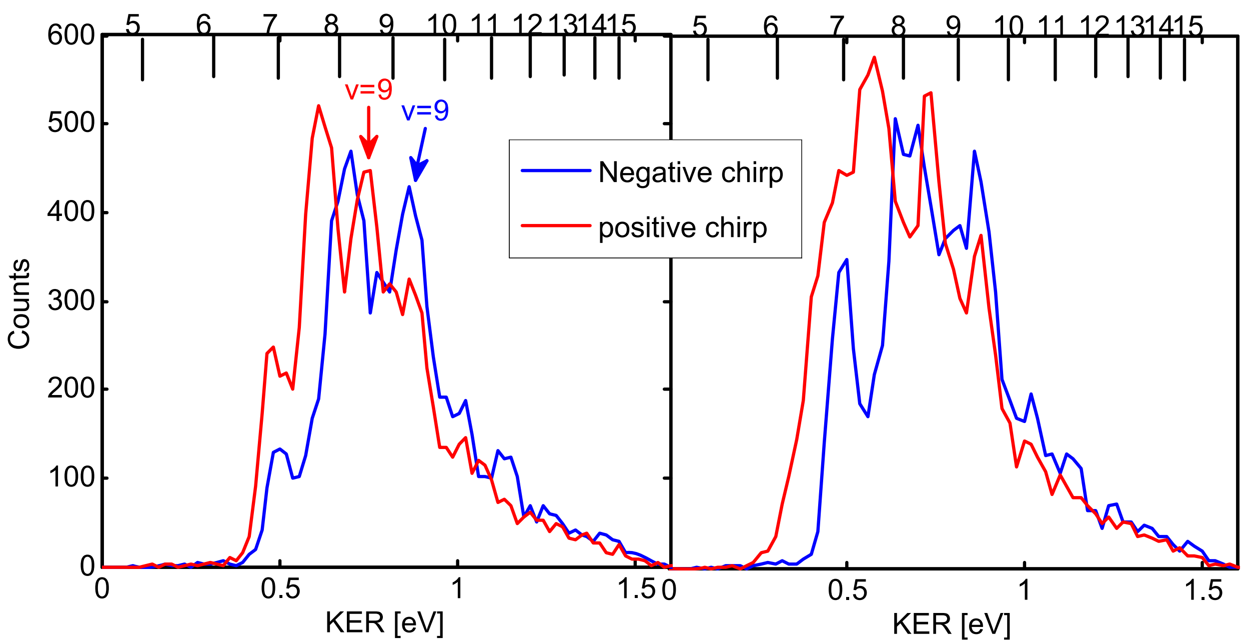




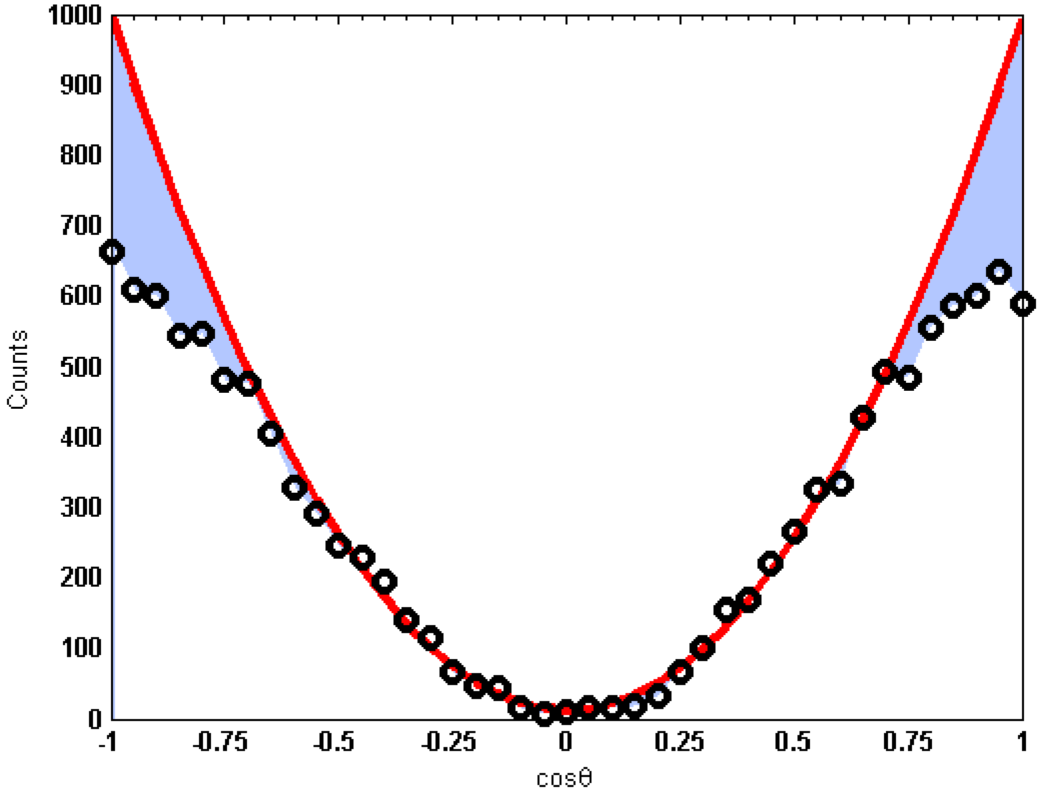


We've seen this also in higher chirp rates…

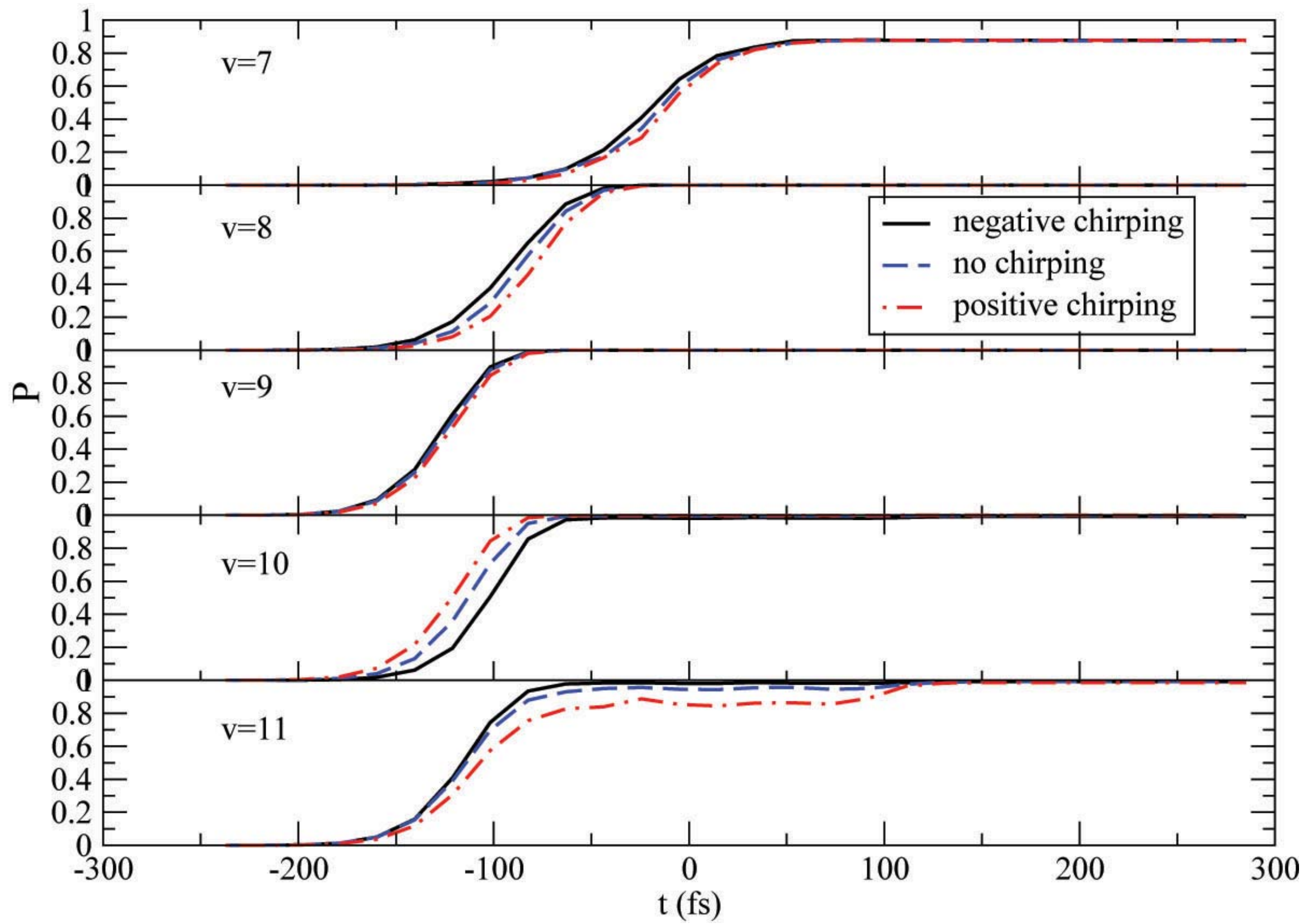
260fs vs 120fs chirp



Angular distribution in 1-photon region

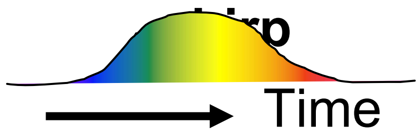


1D TDSE calculation confirms shift mechanism

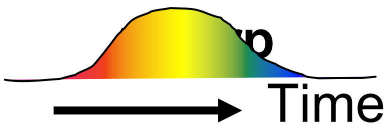


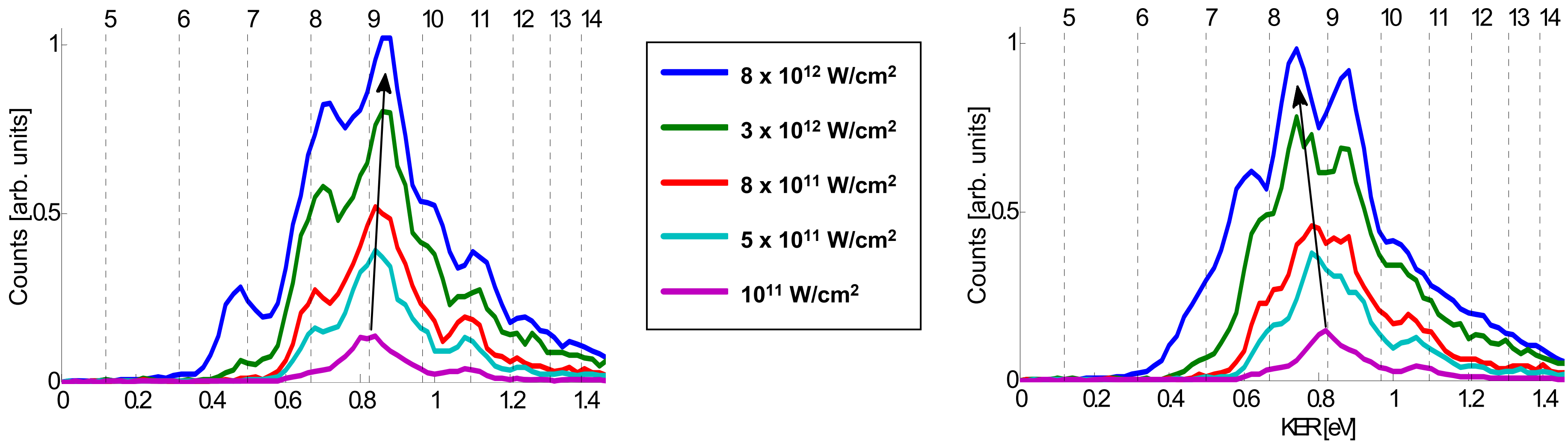
chirped pulses levels shift with intensity

120 fs Negative

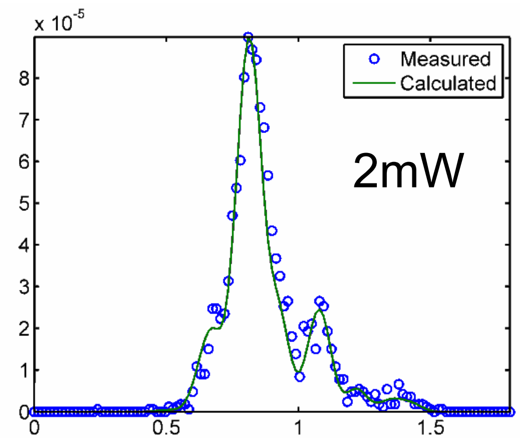


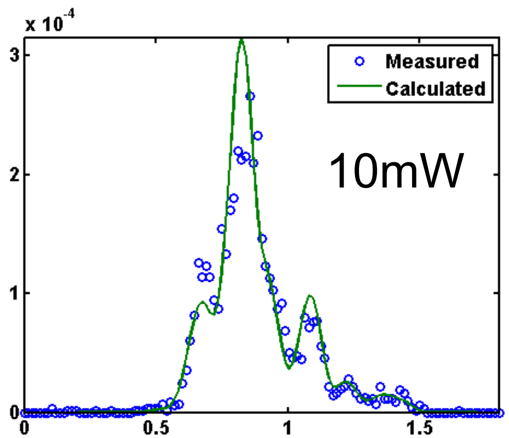
120 fs Positive

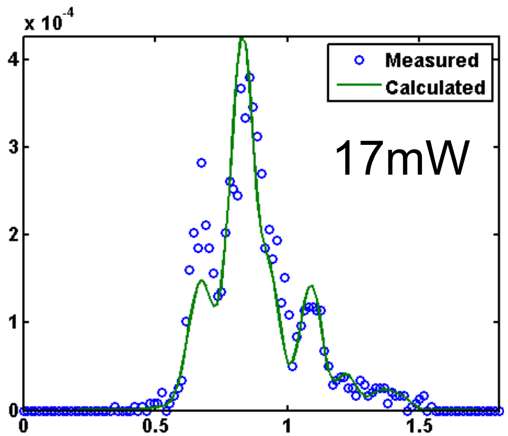


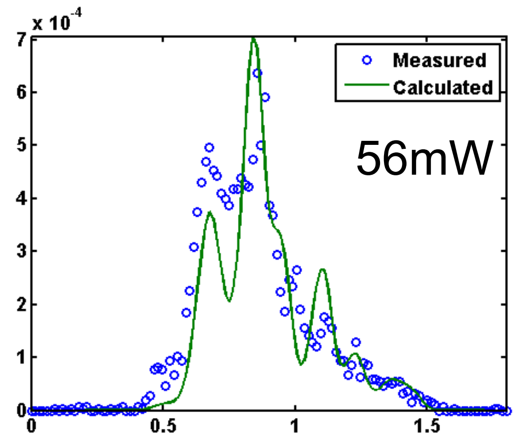


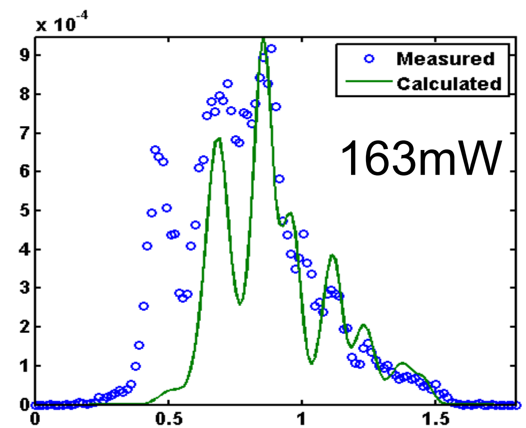
Negative chirp:
comparison with absolute normalized data along the polarization



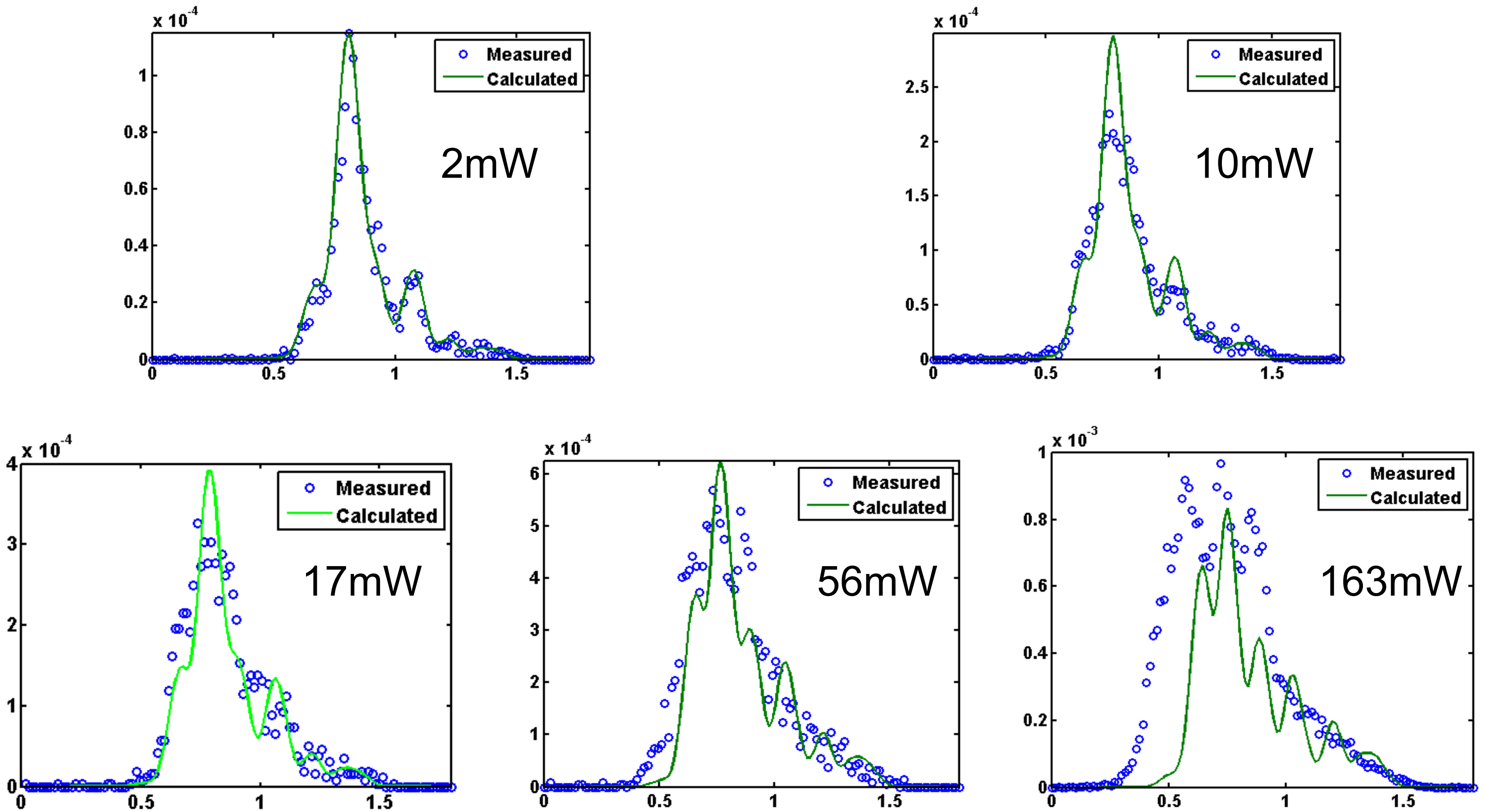








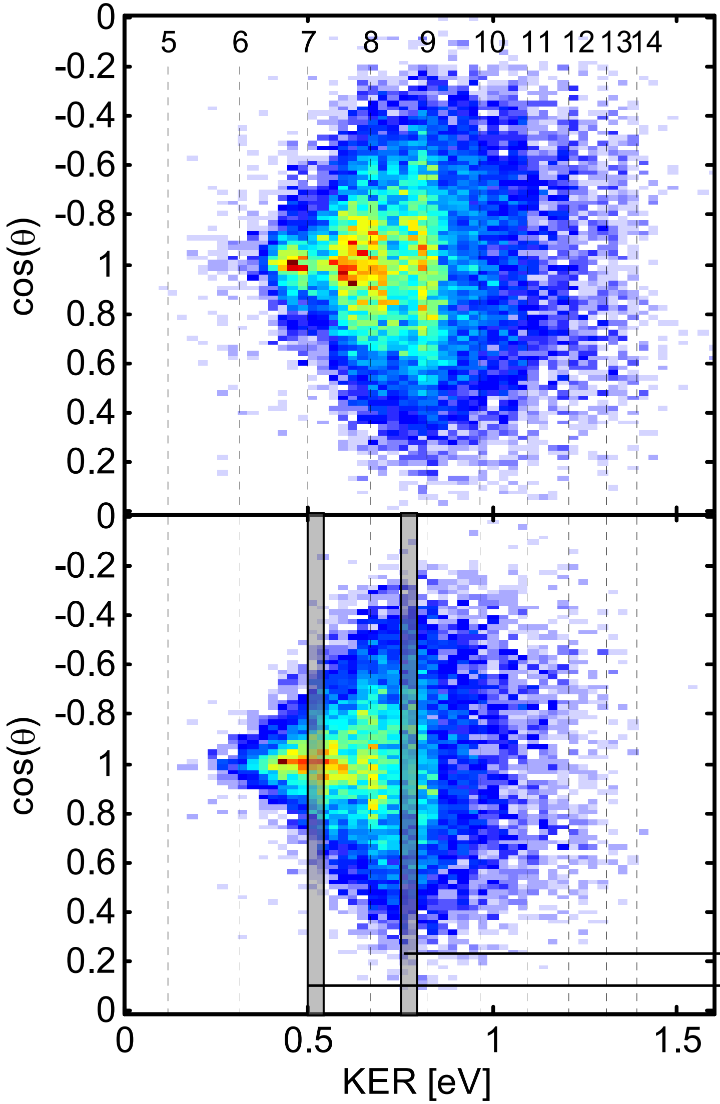
Positive chirp:
comparison with absolute normalized data along the polarization



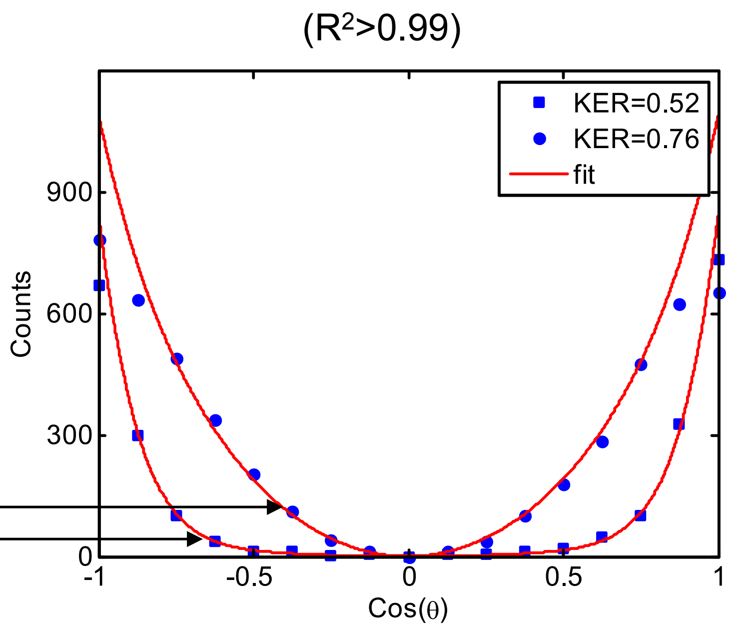
Angular distribution analysis

Negative chirp

Positive chirp



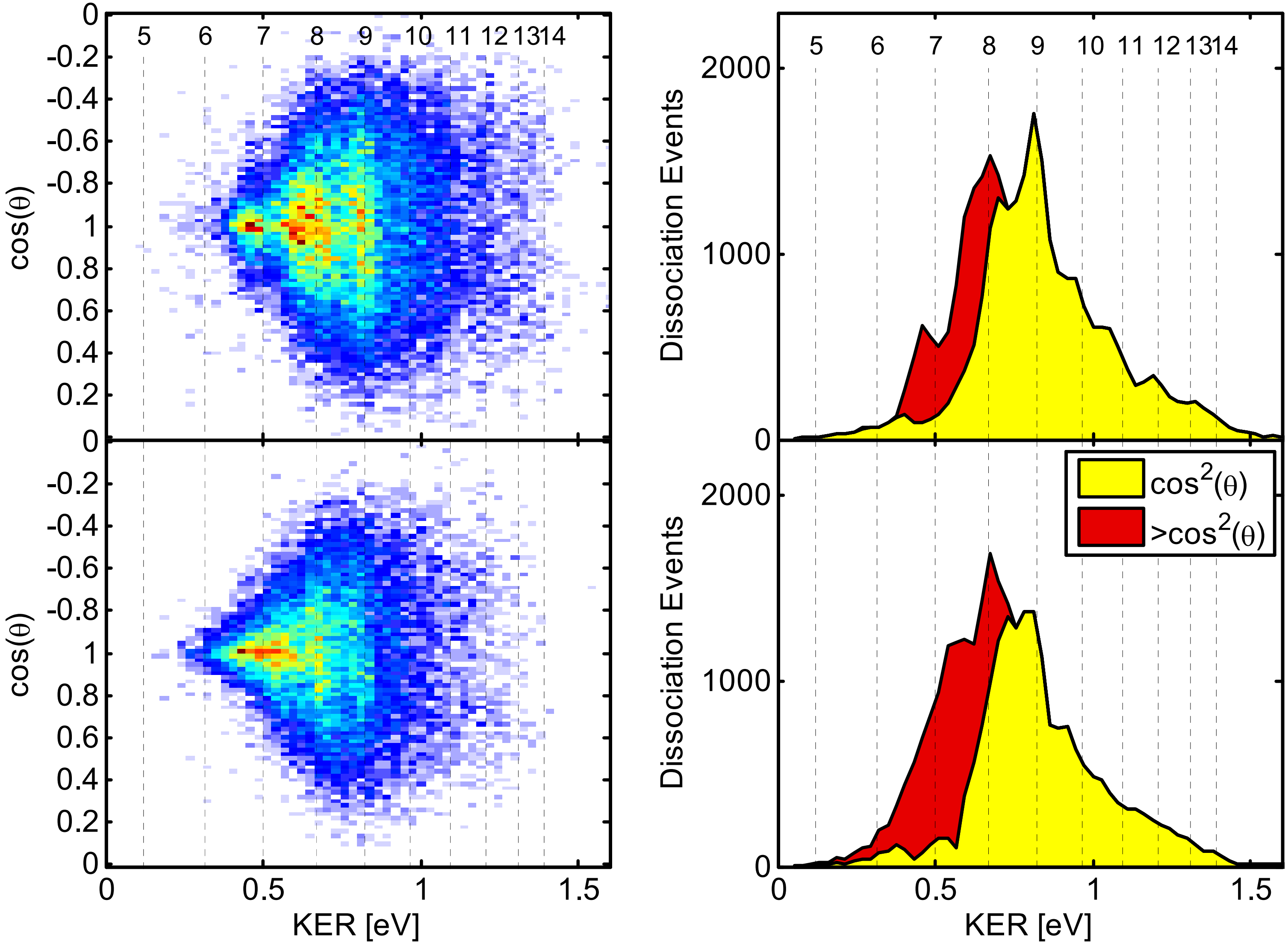
Strong field \ multiphoton processes have angular distributions larger than $\cos^2\theta$



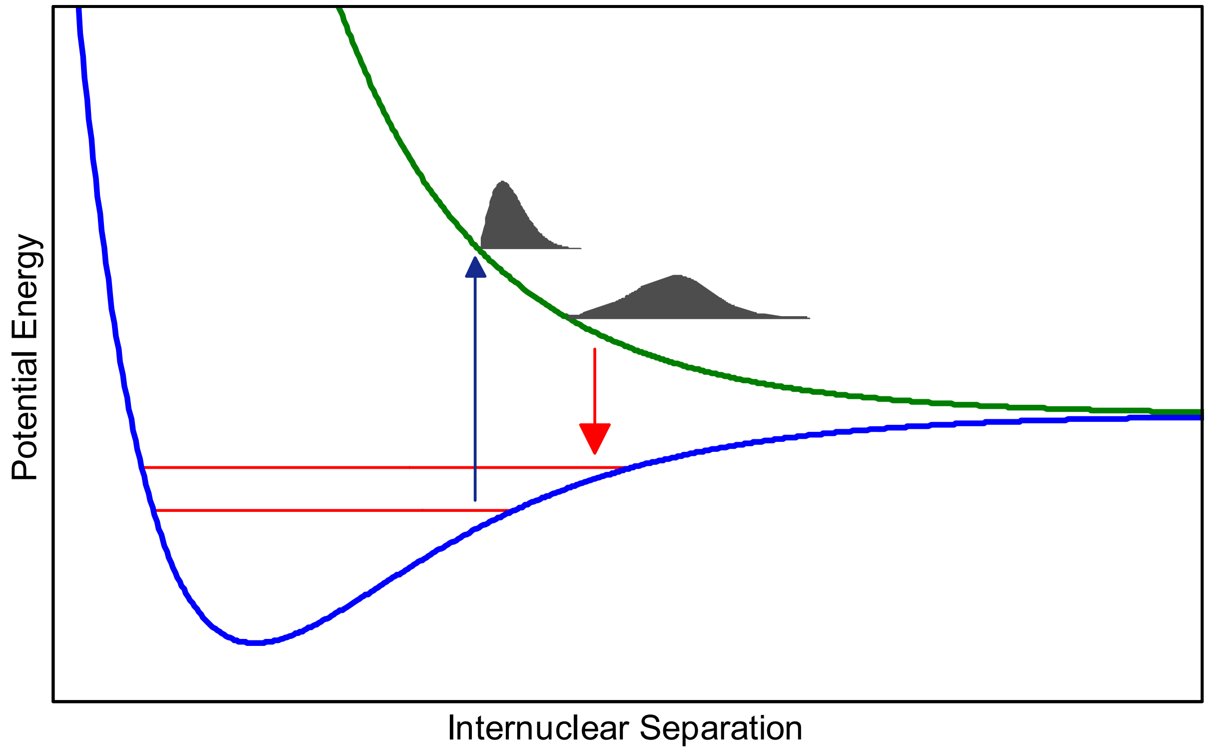
Angular distribution analysis

Negative chirp (90fs)

Positive chirp (90fs)



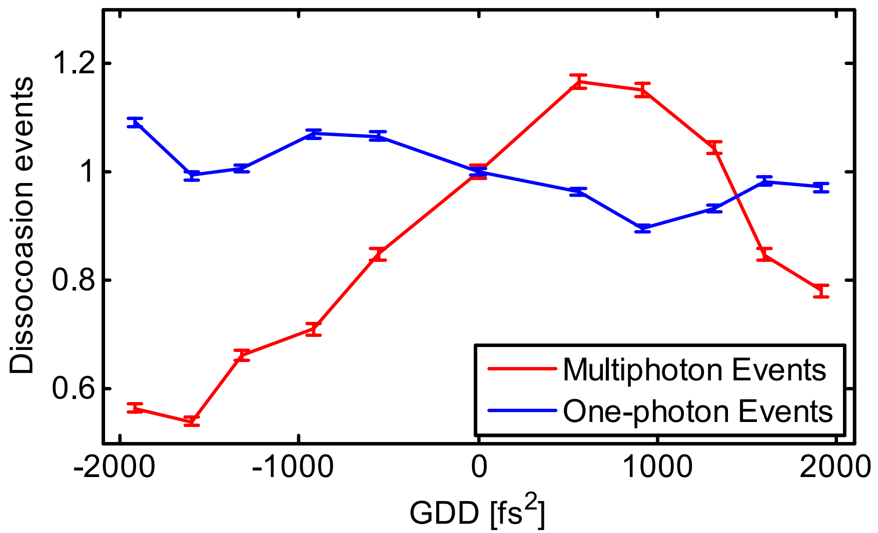
Negative chirp dumps better

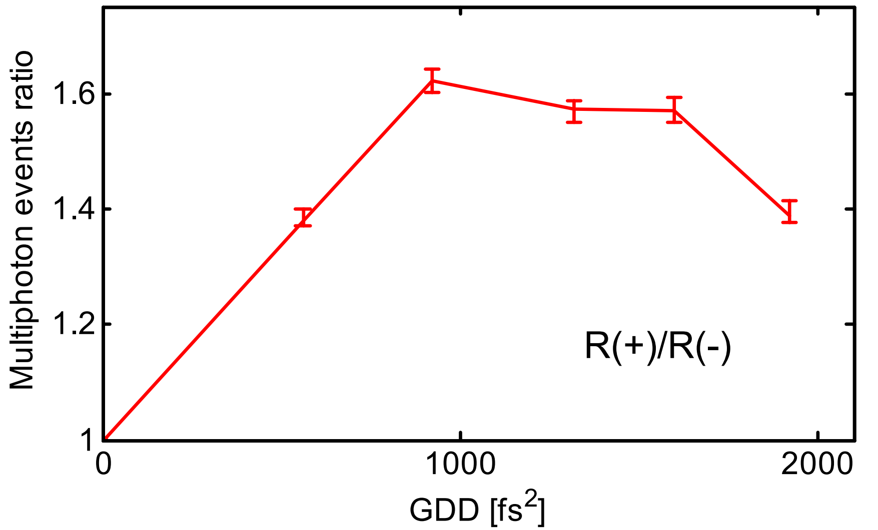


Chirp sign and rate controls dissociation efficiency

One photon events are barely affected

For multiphoton events, positive chirp is always more efficient





In conclusion…

- The simplest strong field experiment on molecular photodissociation has still some elaborate physics

- The frequency content of the pulse, given by the chirp rate, has a significant role in the overall dissociation mechanism, it affects:
  - Dissociation probability.
  - Angular distribution.
  - KER peak positions (levels shift).

Thank you