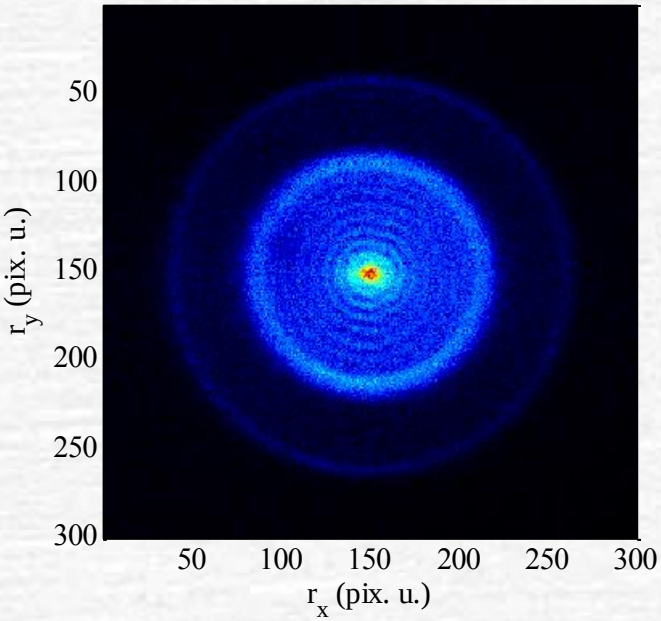
Alternative probes of photoionization time delays



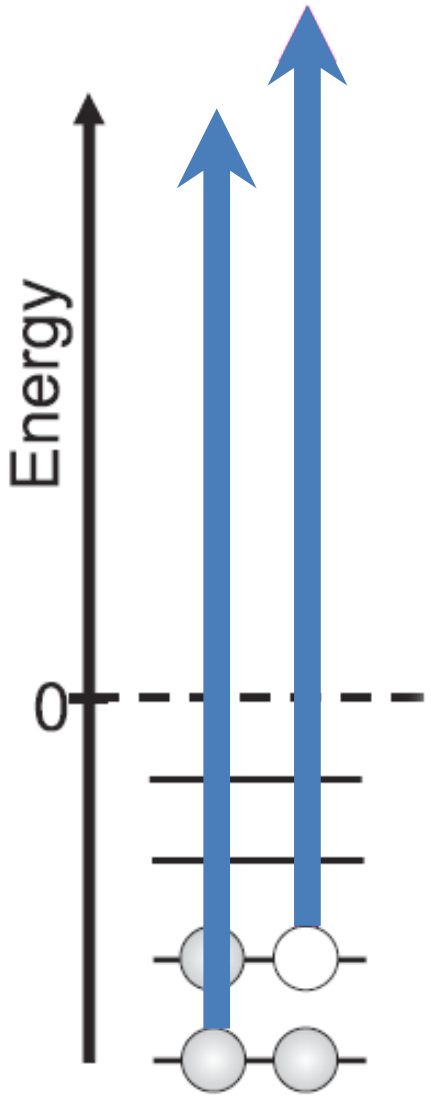
Marc Vrakking

Max-Born-Institut (MBI), Berlin, DE

Santa Barbara, 28-8-2014

Max-Born-Institut

Delay in photoemission



Questions: do two electrons that originate from different orbitals ionize at the same time or is there a delay between the two?

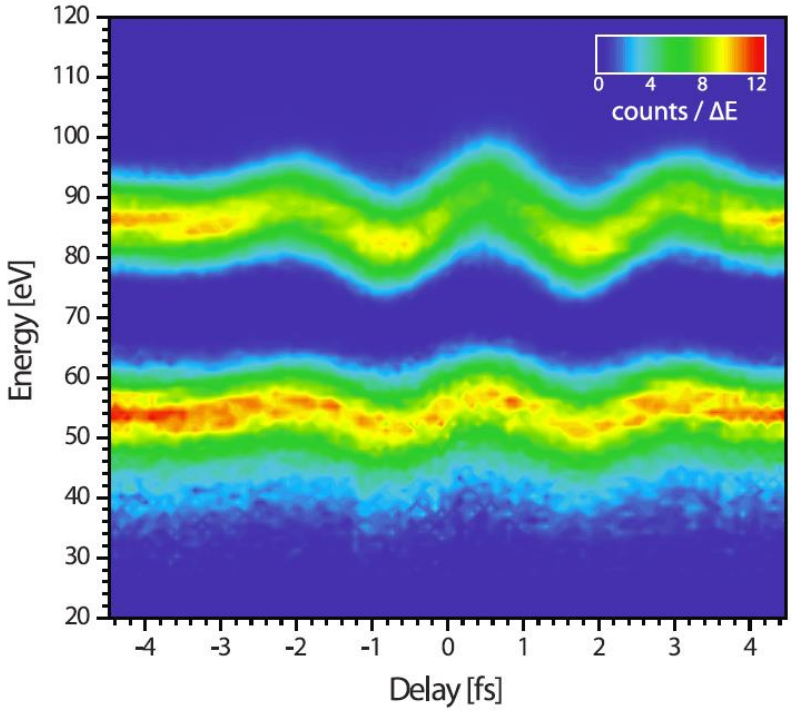
Two experimental approaches:

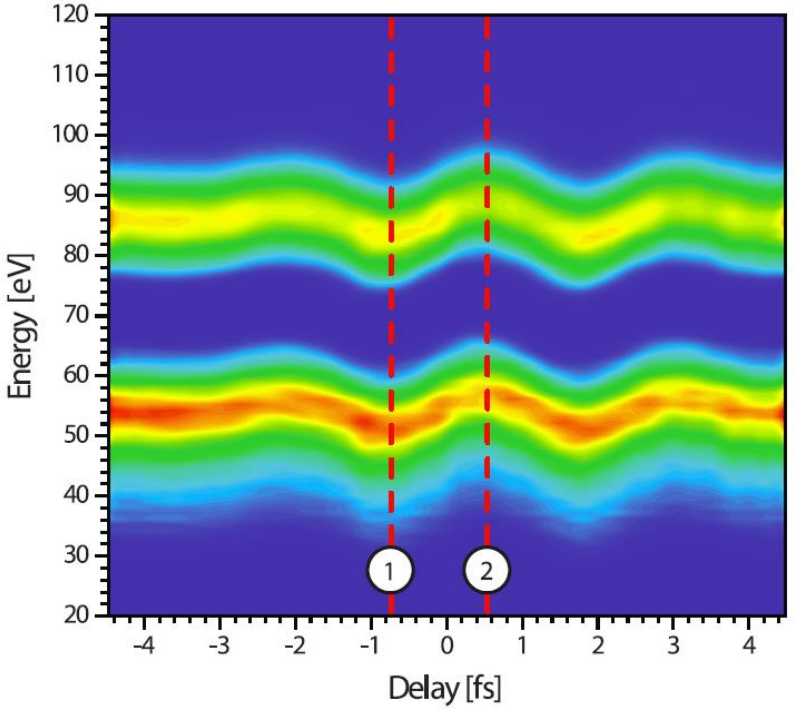
- Ionization by an isolated attosecond pulse (IAP) in combination with a streaking measurement

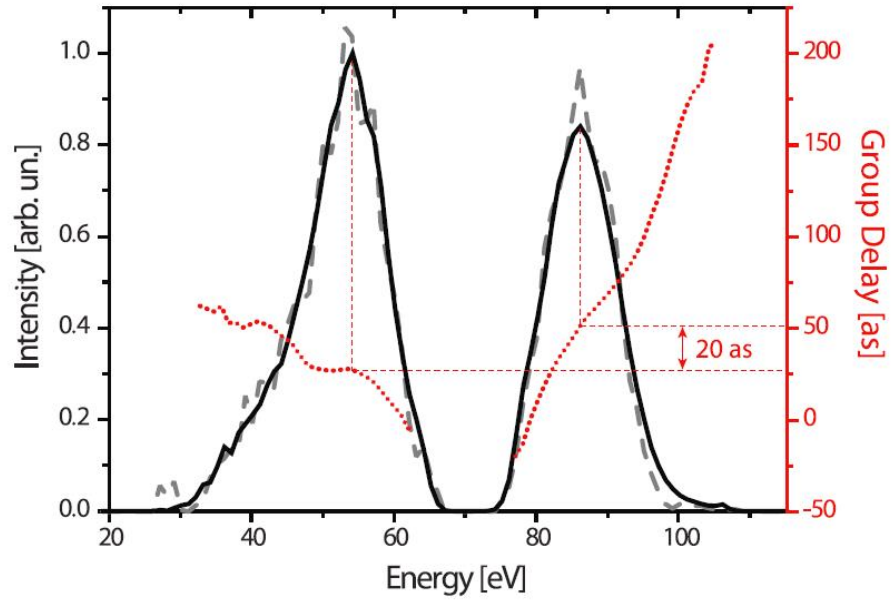
Schultze et al, Science 328, 1658 (2010)

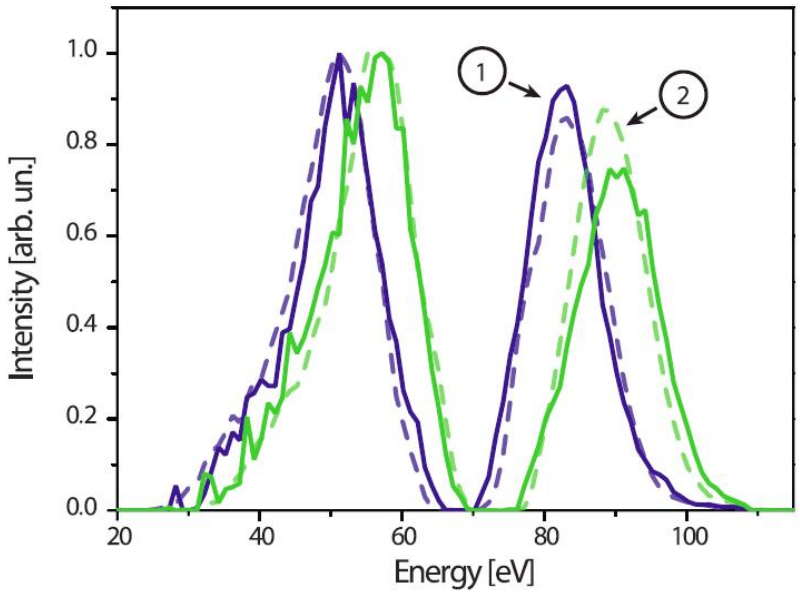
- Ionization by a train of attosecond pulse (APT) in combination with a RABBITT measurement

Kluender et al, Phys. Rev. Lett. 106, 143002 (2012)





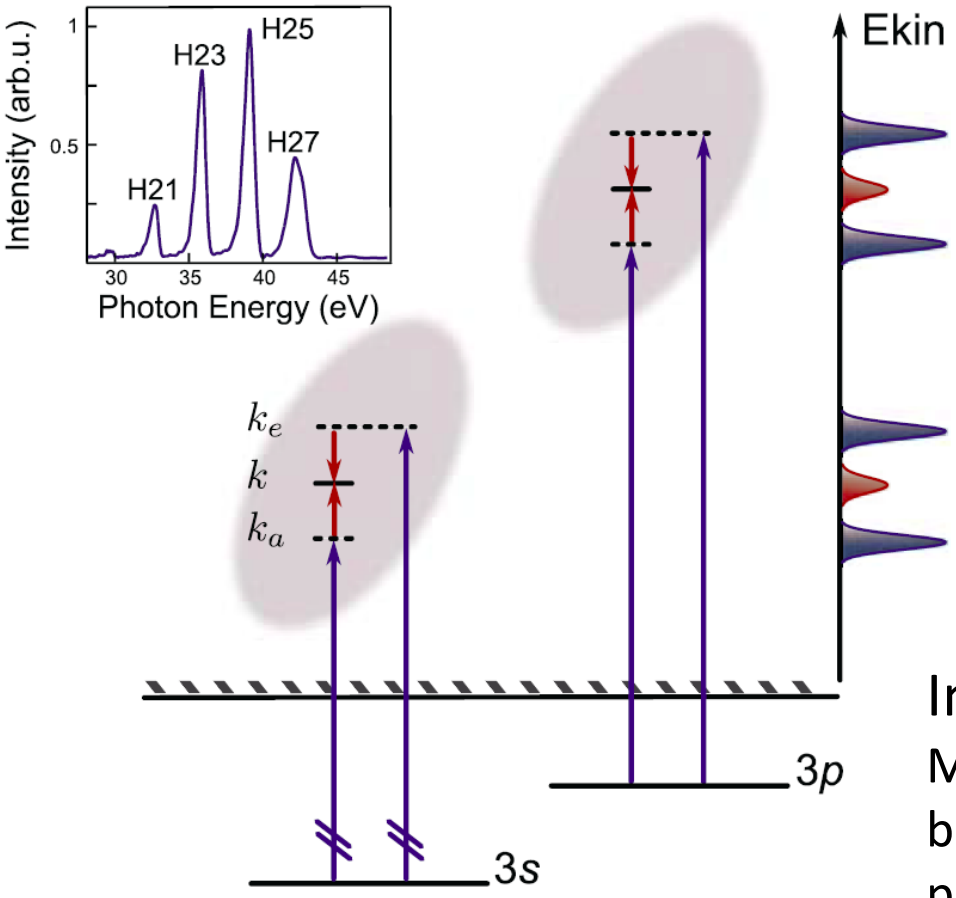


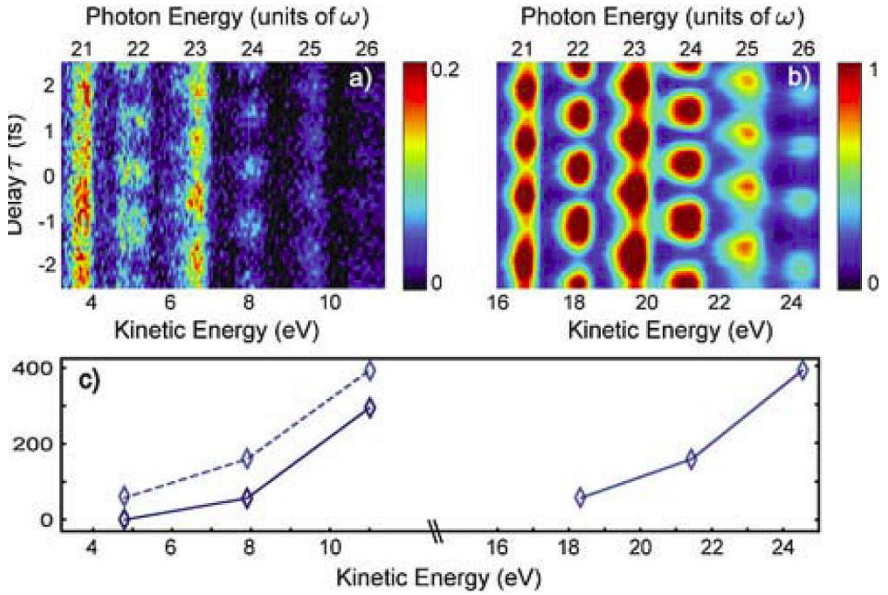


Schultze et al, Science 328, 1658 (2010)

Delay in photoemission

$$S(\tau) = \alpha + \beta \cos[2\omega(\tau - \tau_A - \tau_I)],$$



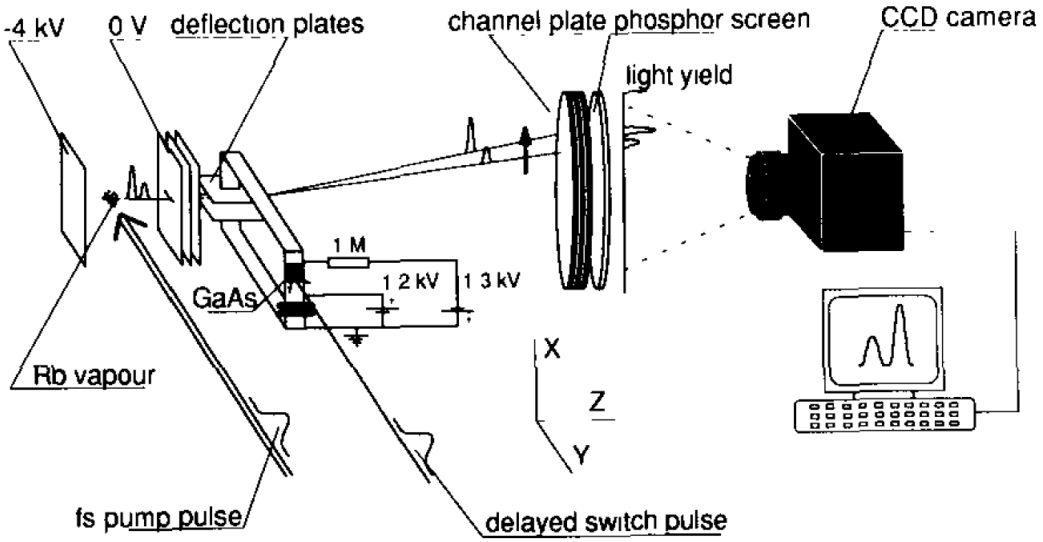


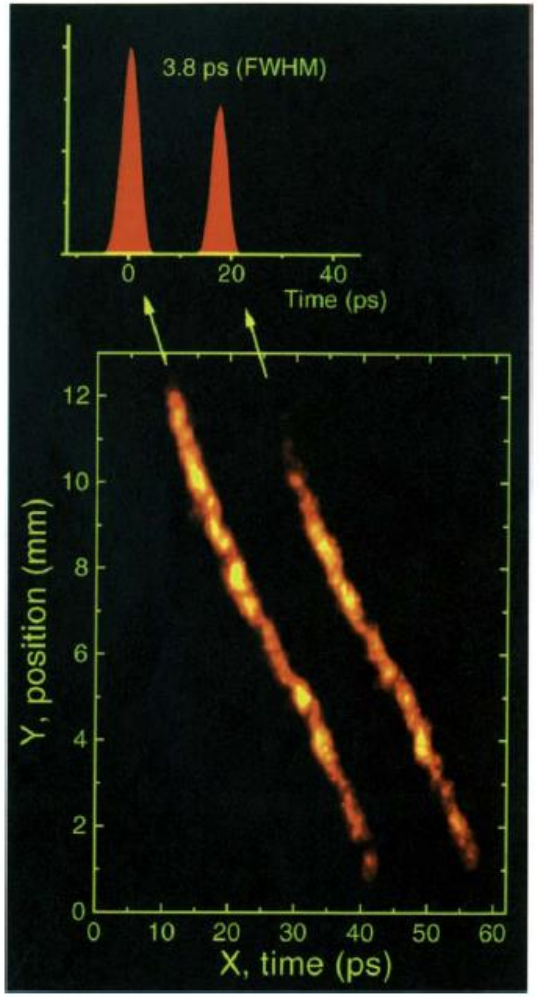
Important:

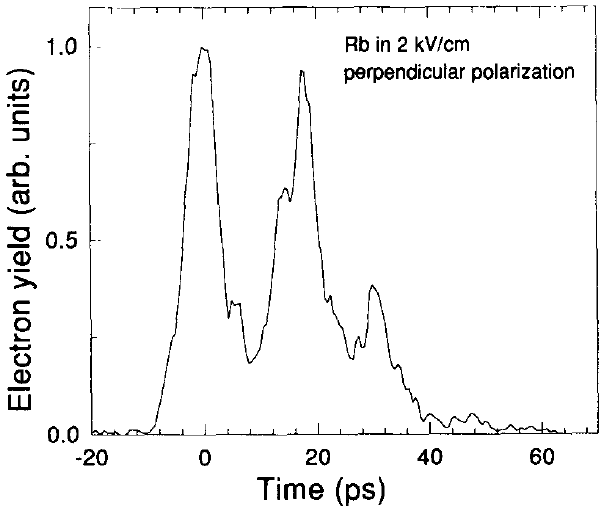
Measured time delays /phases accumulate both during ionization process and during propagation in the Coulomb+laser fields

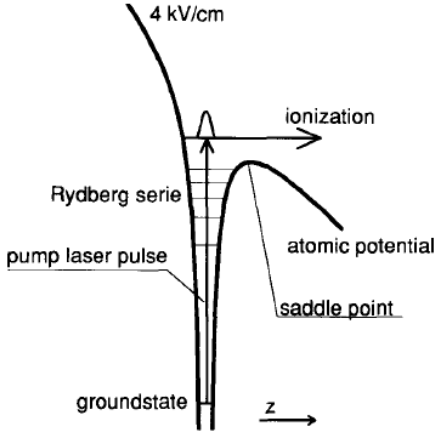
Kluender et al, Phys. Rev. Lett. 106, 143002 (2012)

Is this new?







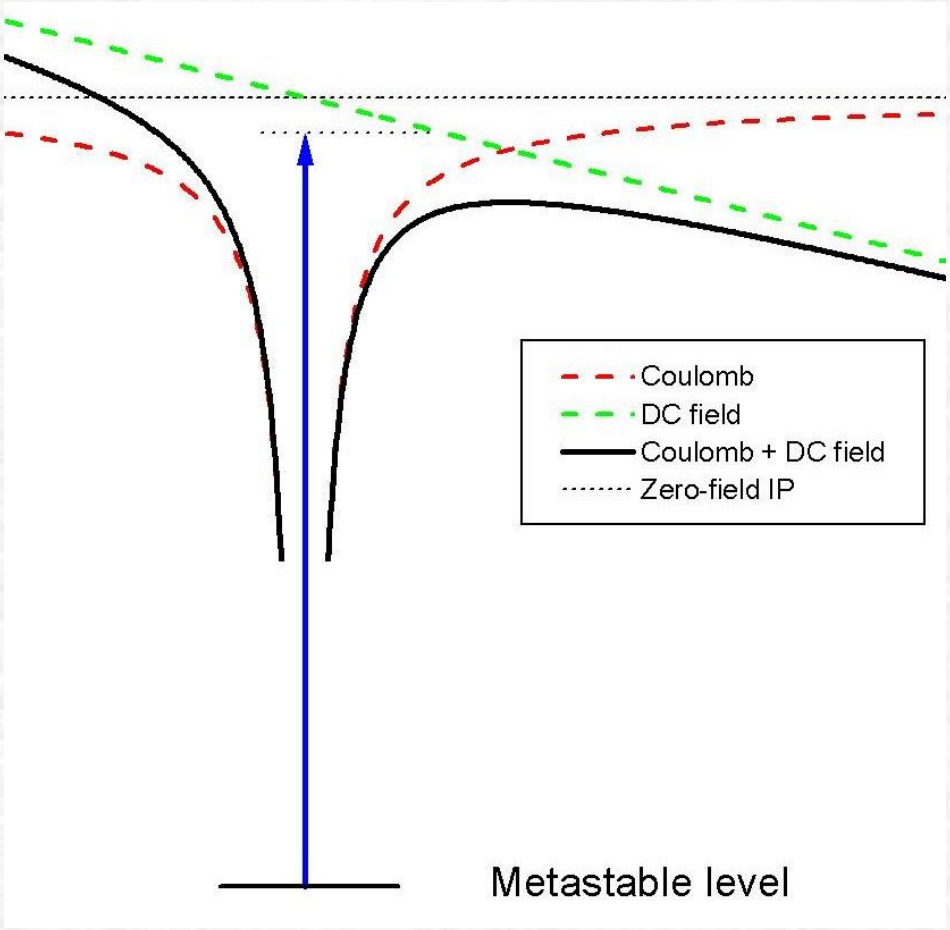


Lankhuijzen and Noordam, Opt. Comm. 129, 361 (1996)

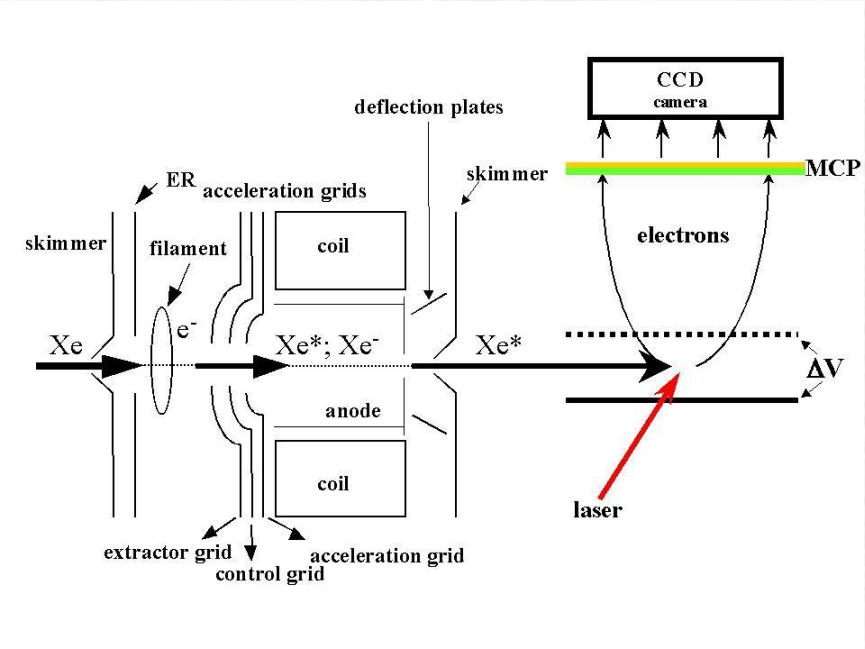
Part 1

Resonant Photoionization microscopy of Hydrogen atoms

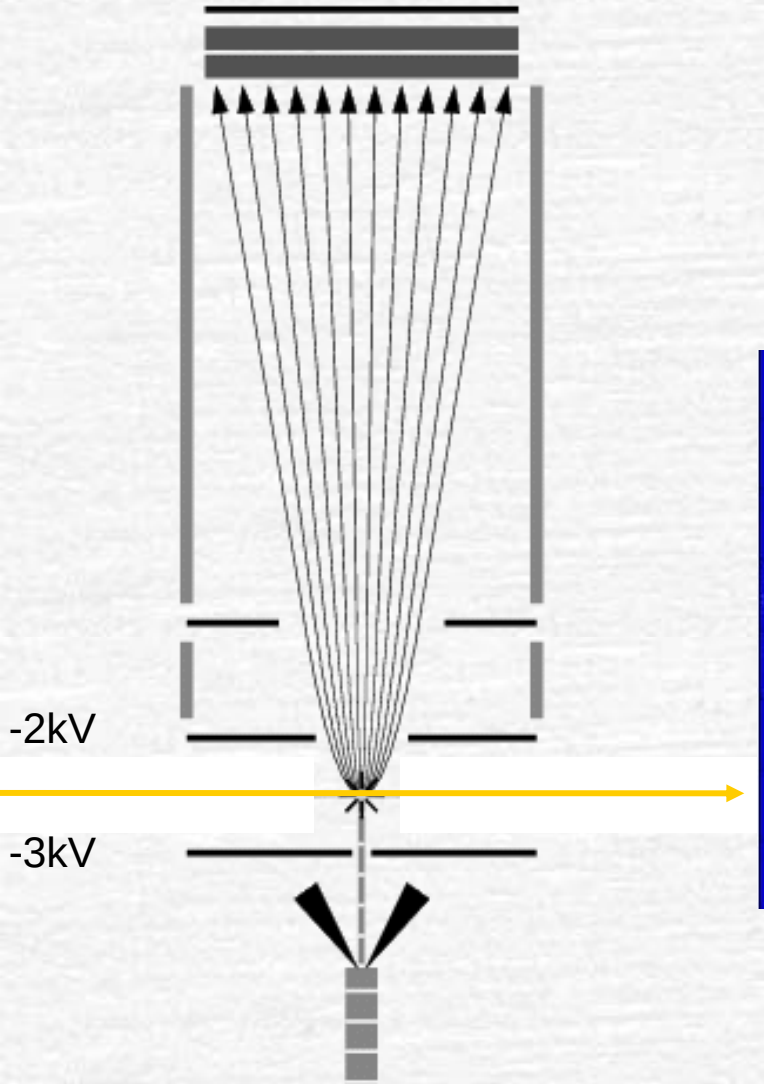
Ionization of metastable Xe atoms with a small excess kinetic energy, using a tunable ns laser



Excitation near the field-free ionisation limit (above the saddle-point in the combined DC field + Coulomb potential)



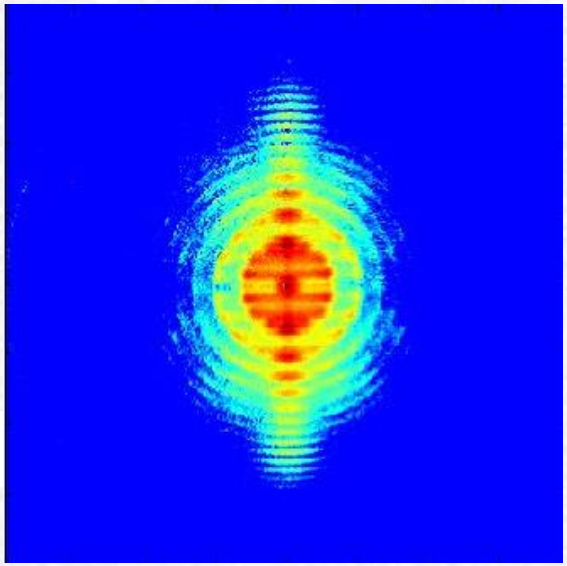
Velocity Map Imaging (VMI) Spectrometry

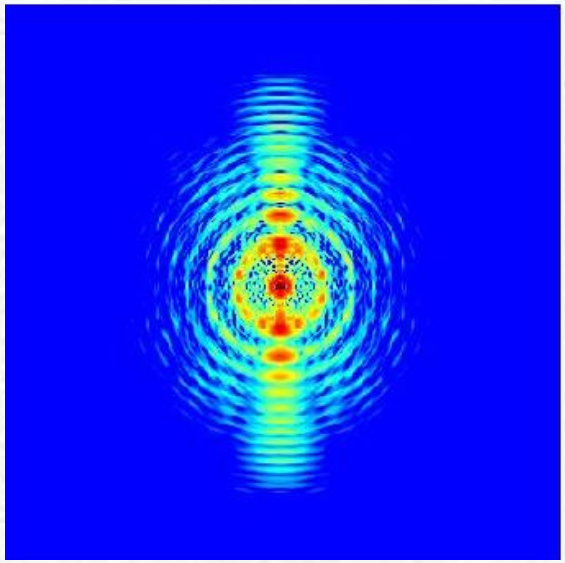


Abel inversion

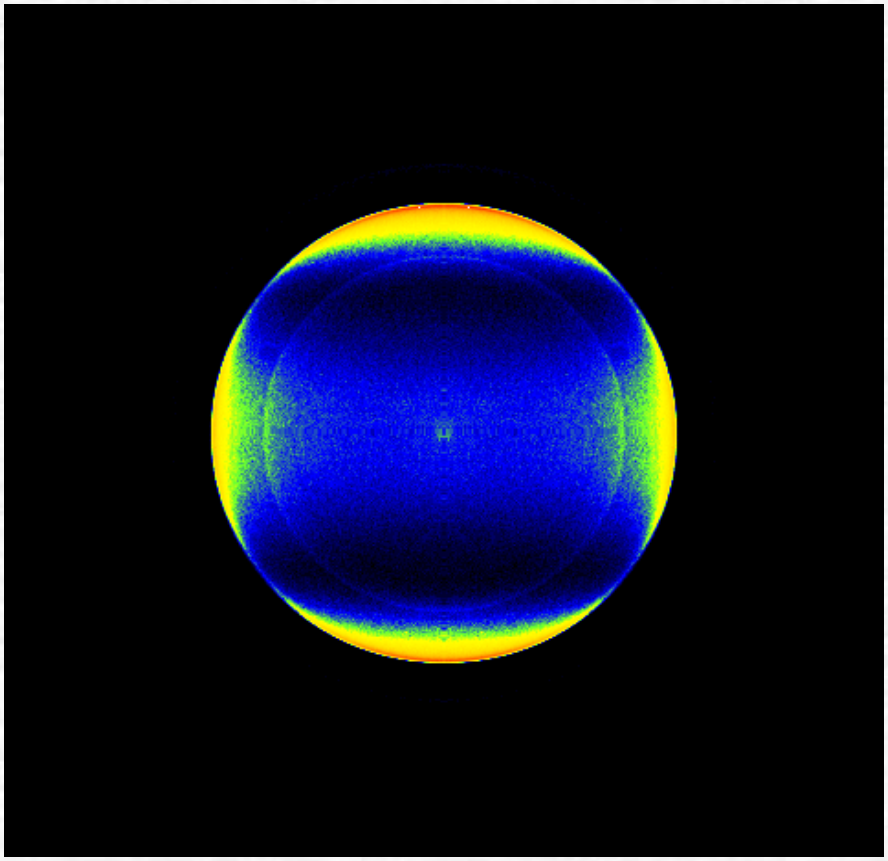
Velocity map image

Slice through the 3D distribution

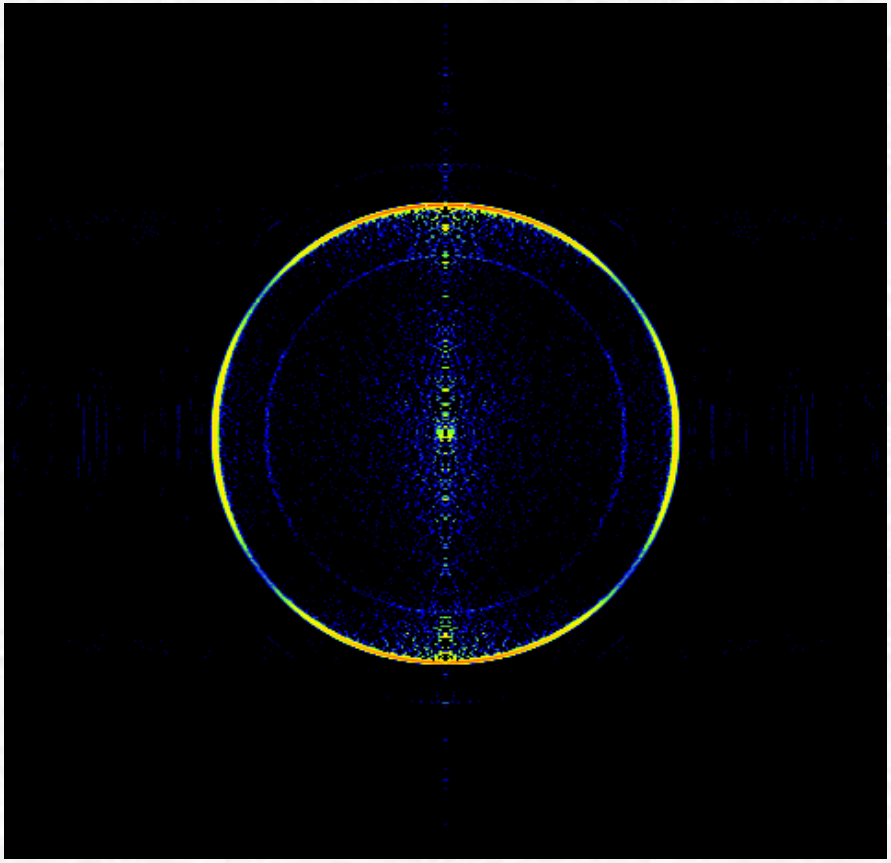




Extraction of the energy and angular distribution using an iterative procedure



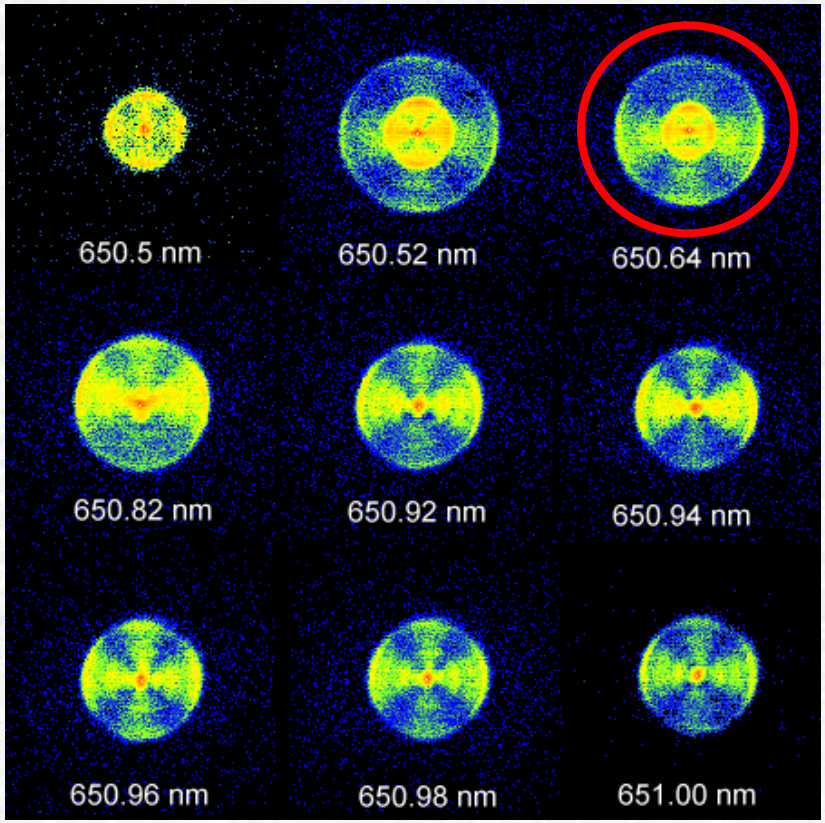
Raw image for 2-photon ionisation of Ar by 532 nm light

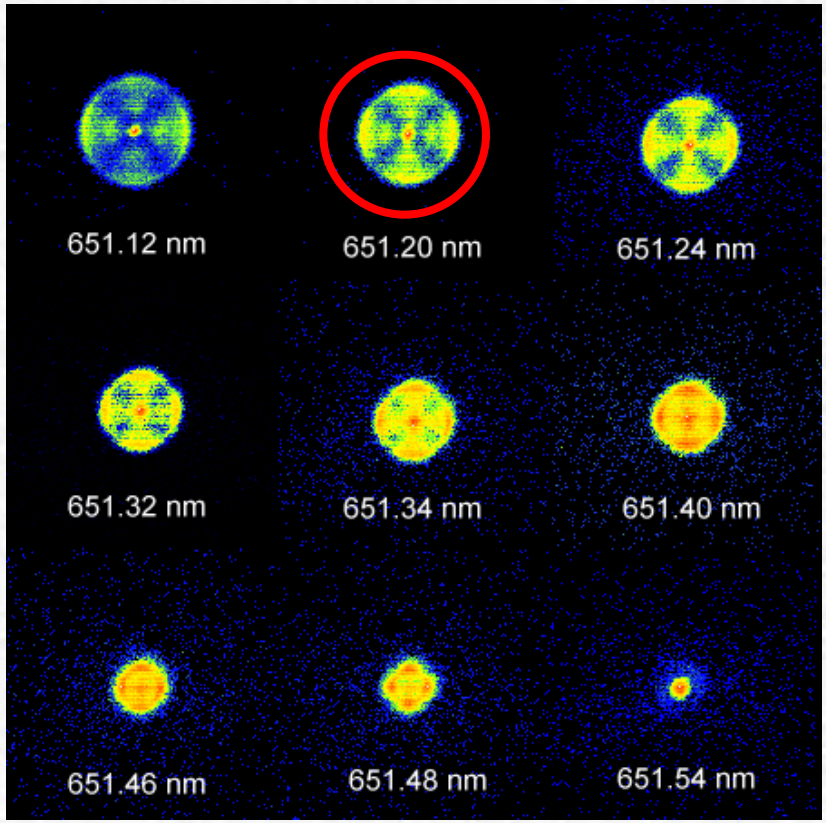


Slice through the 3D velocity distribution, obtained by Abel inversion of the image $\Delta v/v = 1\%$

Slow photoelectron imaging

2-photon ionization of Xe*(6s[3/2]$_2$) in a field of 170 V/cm



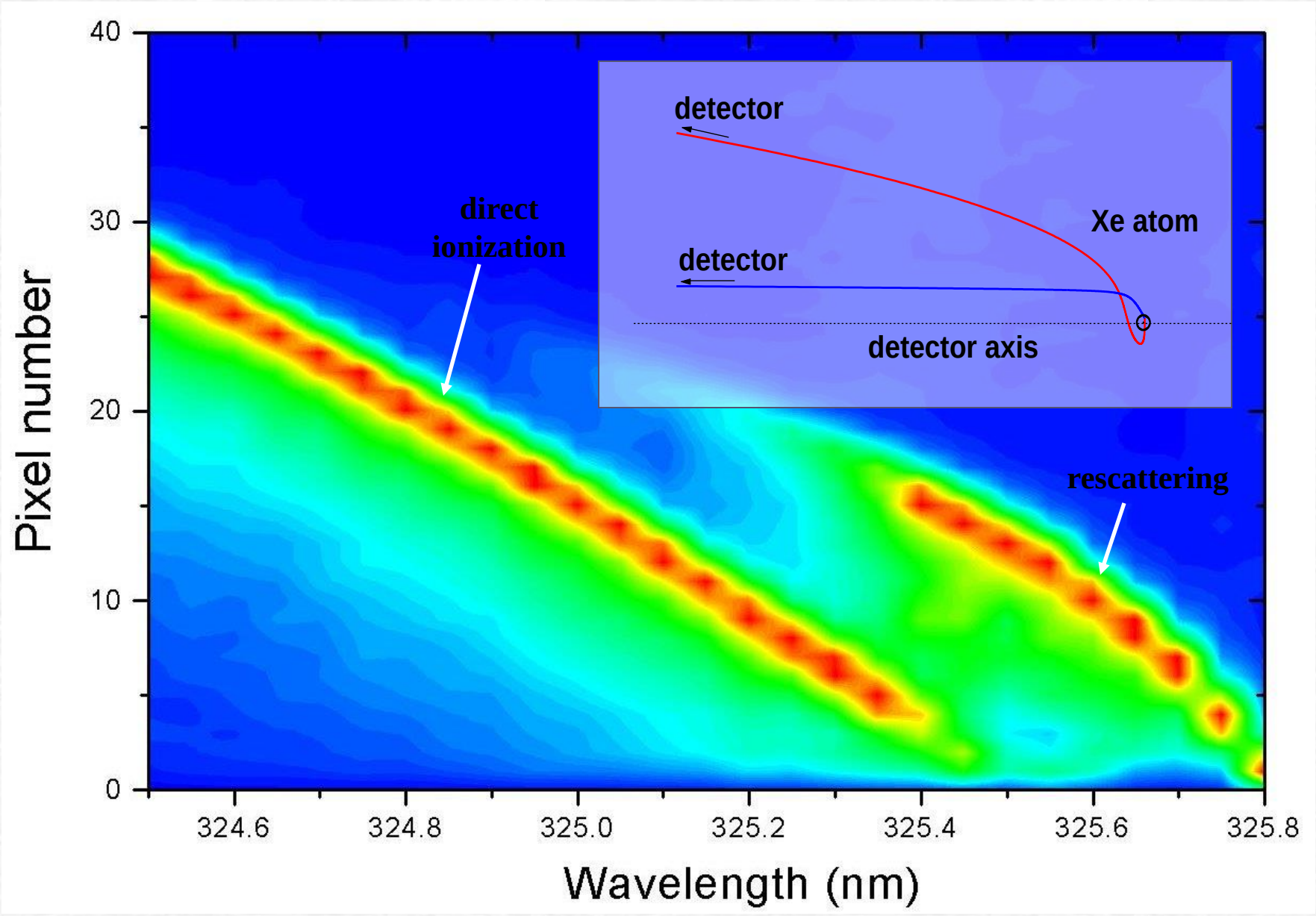


Switching of intensity from outer to inner ring!!

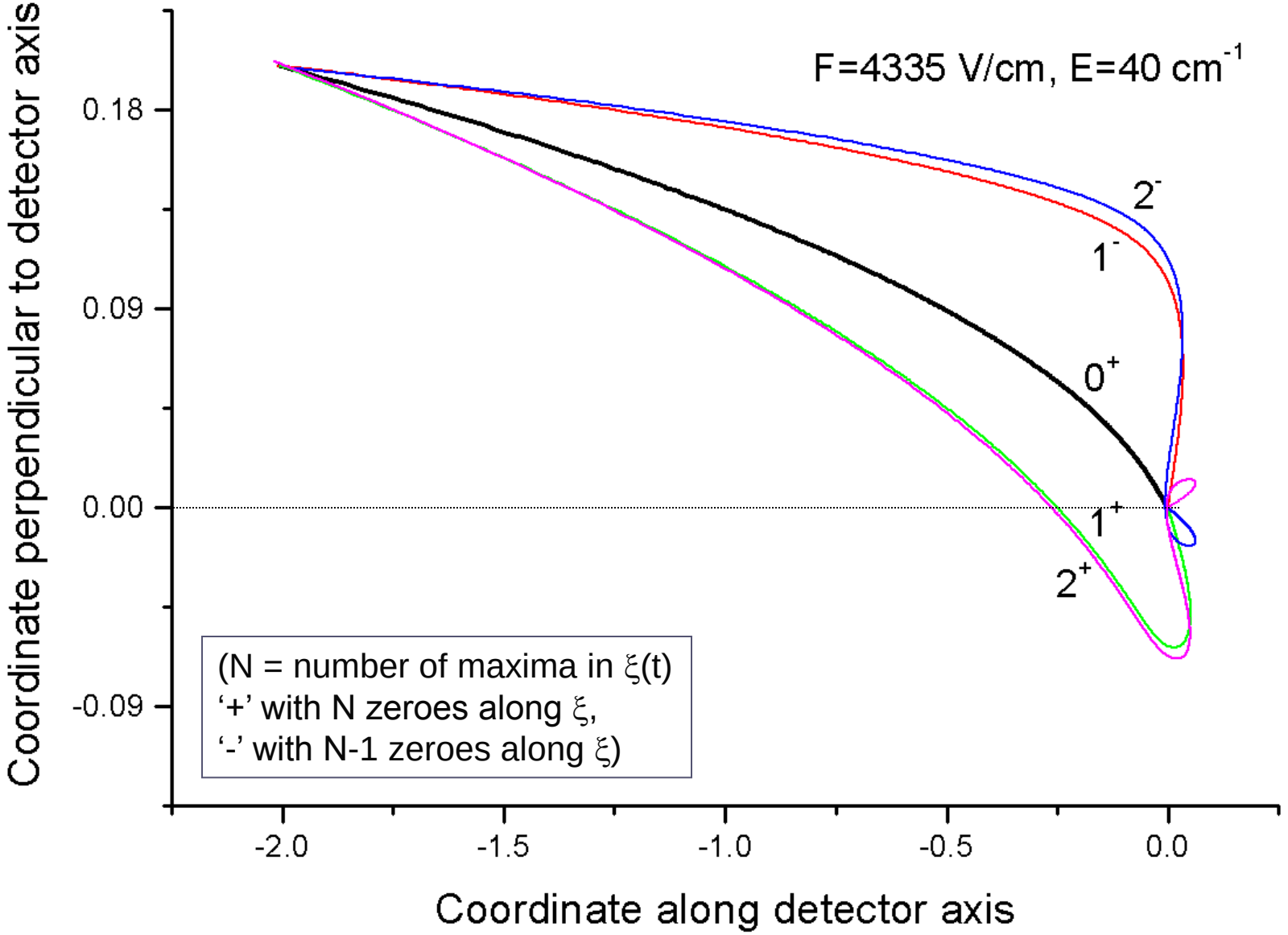
Images are not a single ring!!

Nicole et.al., PRL 85, 4024 (2000)

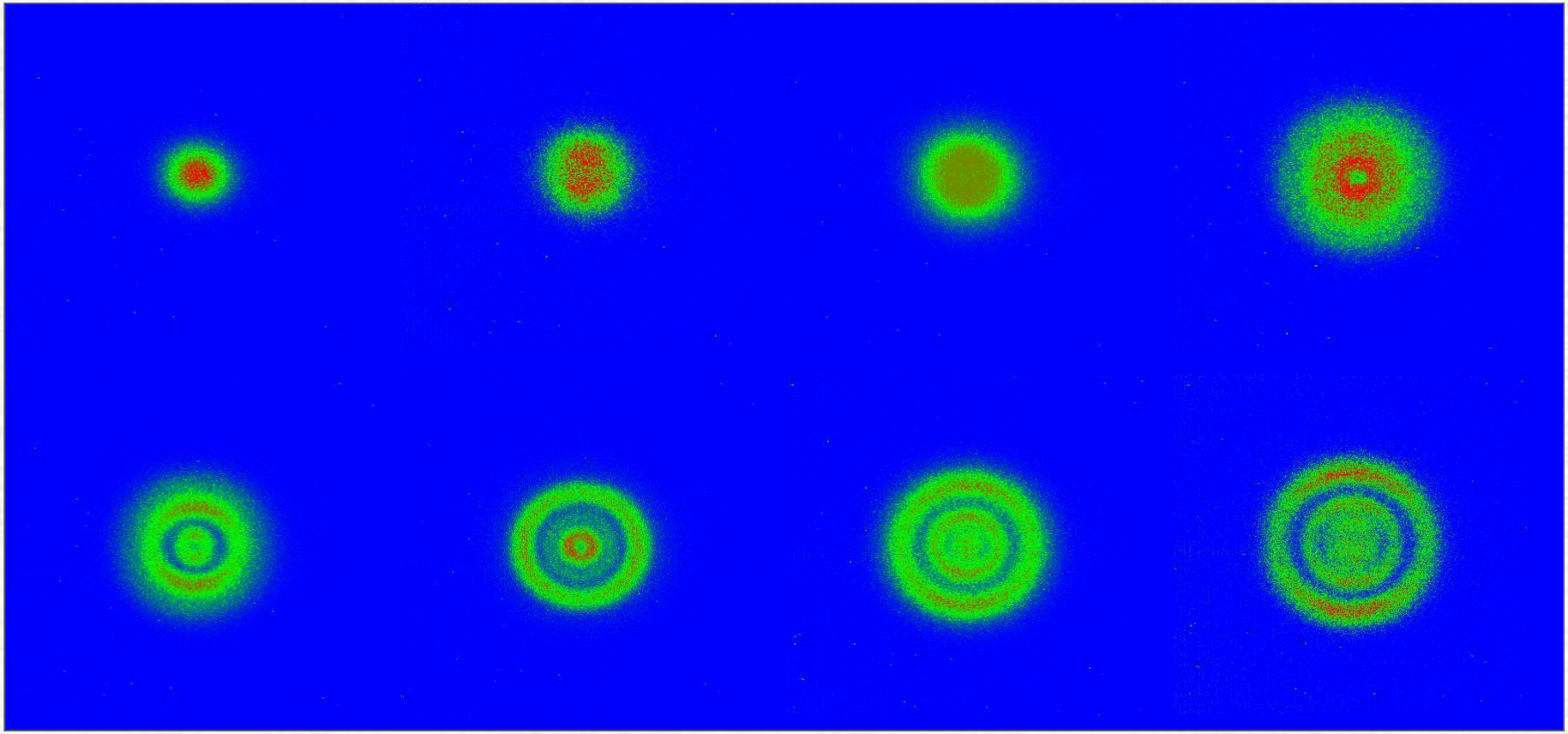
Classical interpretation of electron dynamics



Expect multiple interfering pathways



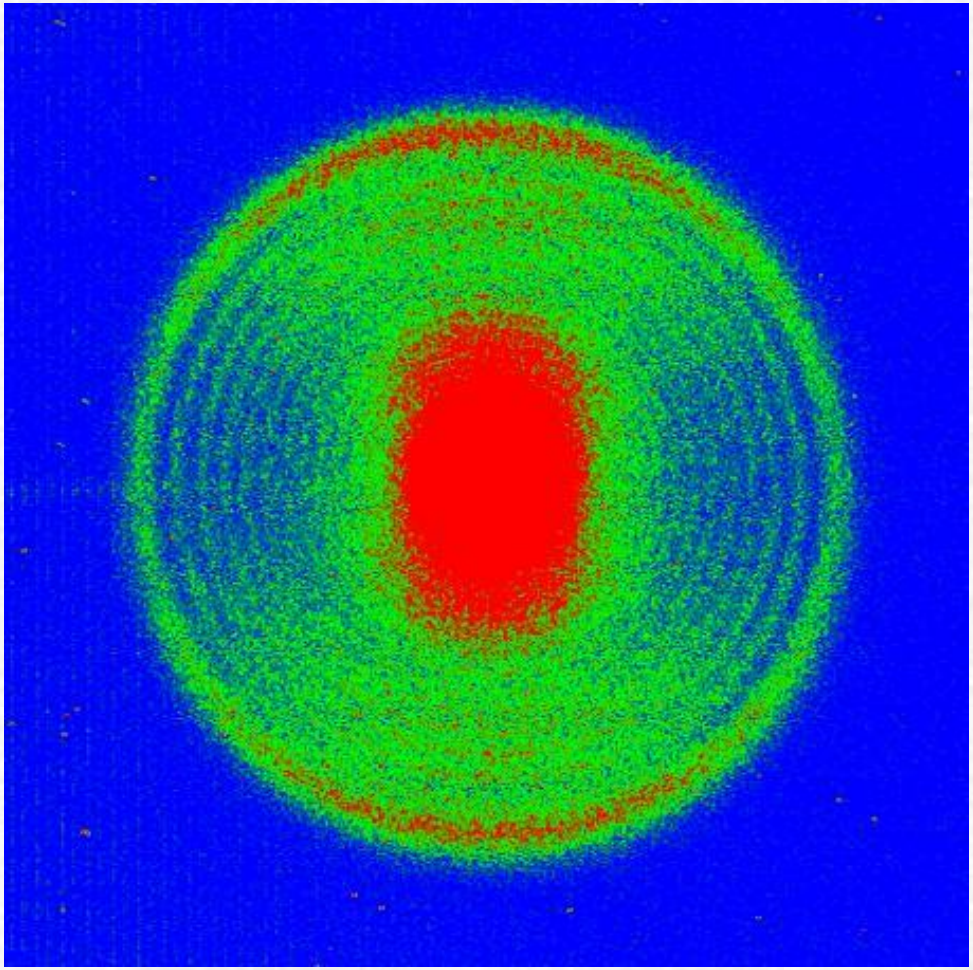
Observation of multi-slit electron interference



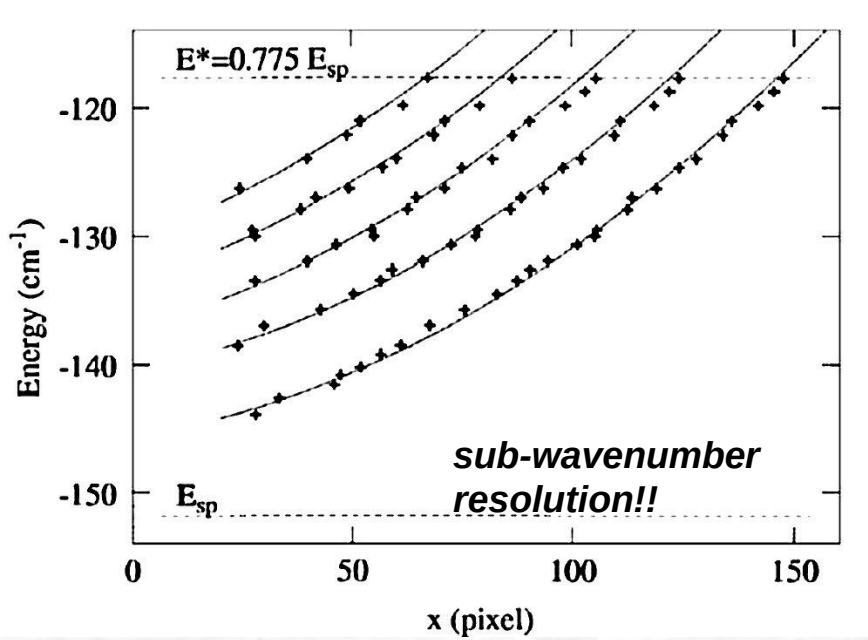
1-photon ionization of Xe*(6s[3/2]$_2$) in a field of 170 V/cm,
Increasing photon energy

Nicole et.al., PRL 88, 133001 (2002)

Observation of multi-slit electron interference



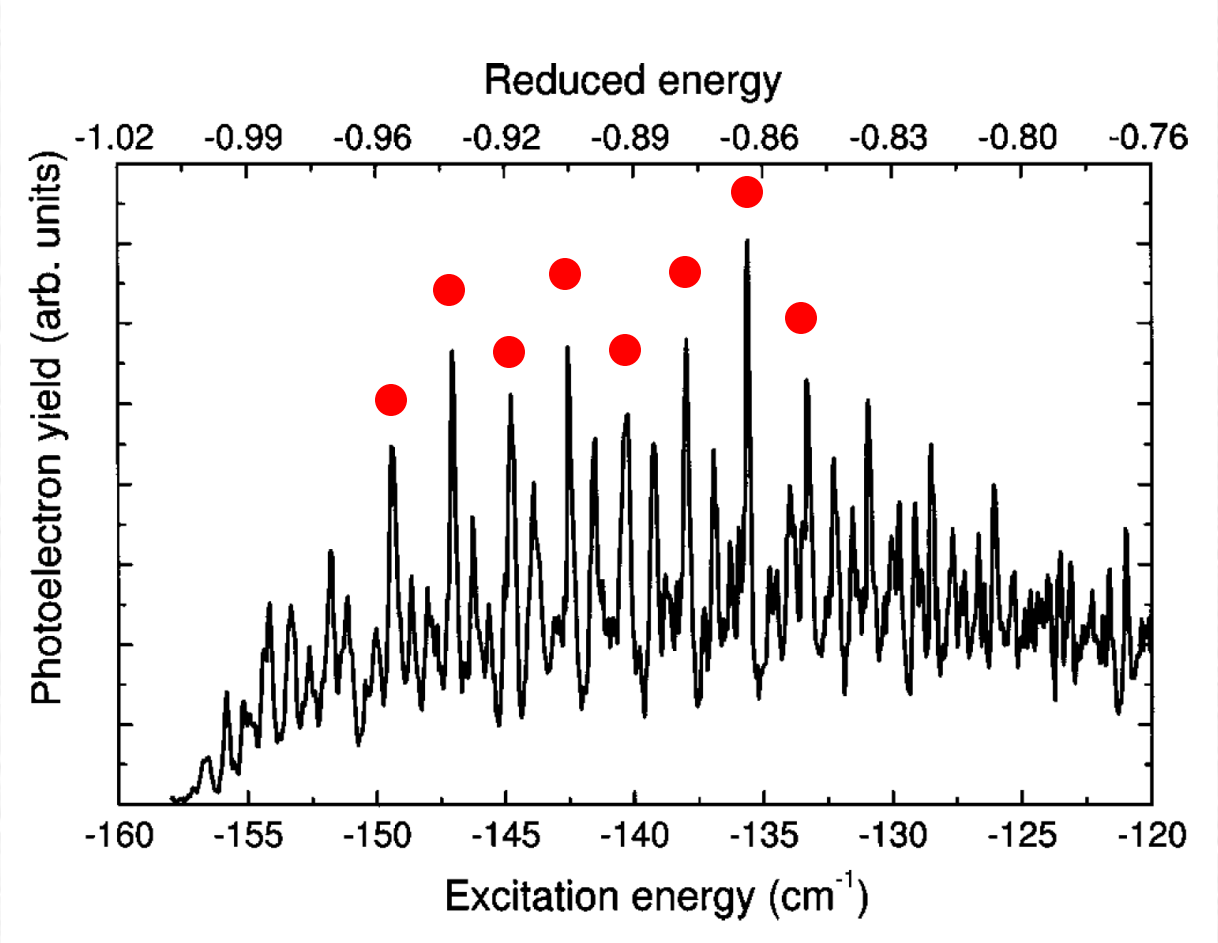
1-photon ionization of Xe*(6s[3/2]$_2$) in a field of 170 V/cm



The number of fringes increases as a function of the energy above the saddlepoint

Nicole et.al., PRL 88, 133001 (2002)

However, this results was also very disappointing…



Interference patterns in Xe are only governed by photoelectron energy and not at all by nature of Stark state…..

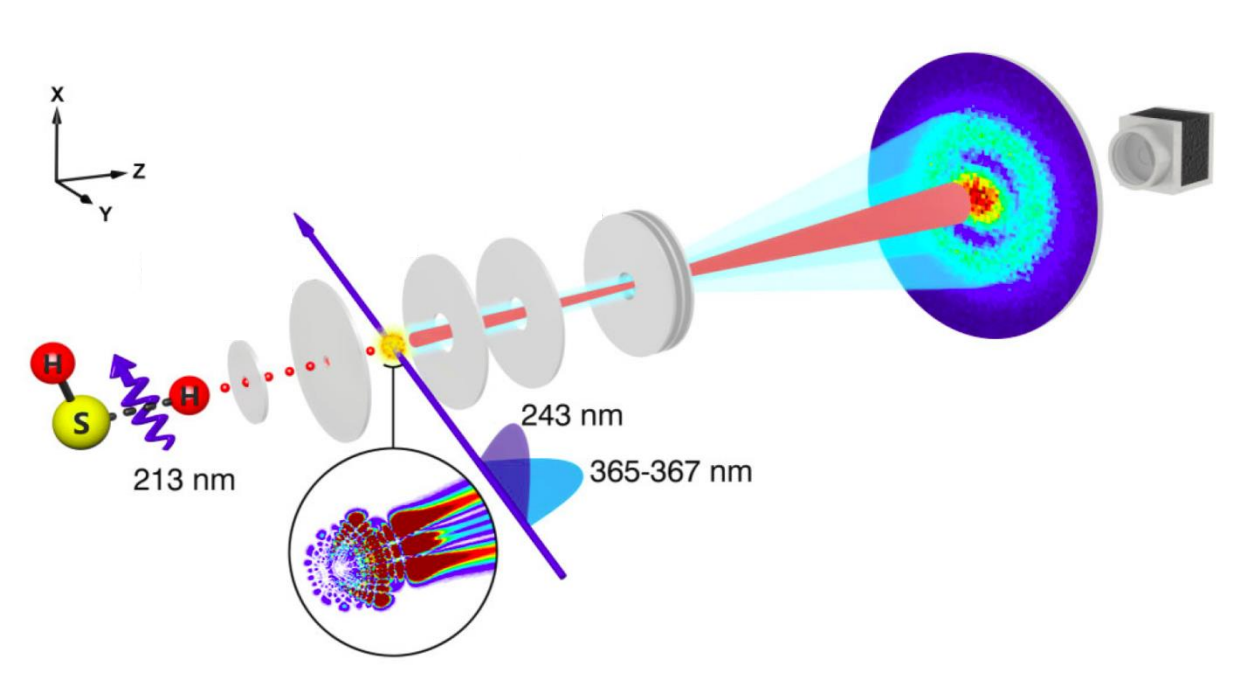
Solution: perform the experiment for H atoms!

The Stark Hamiltonian is separable in parabolic coordinates:

$$\Psi(\xi, \eta, \varphi) = (2\pi\eta\xi)^{-1/2}\chi_1(\xi)\chi_2(\eta)e^{im\varphi}$$

$$\eta = r - z$$
$$\xi = r + z$$

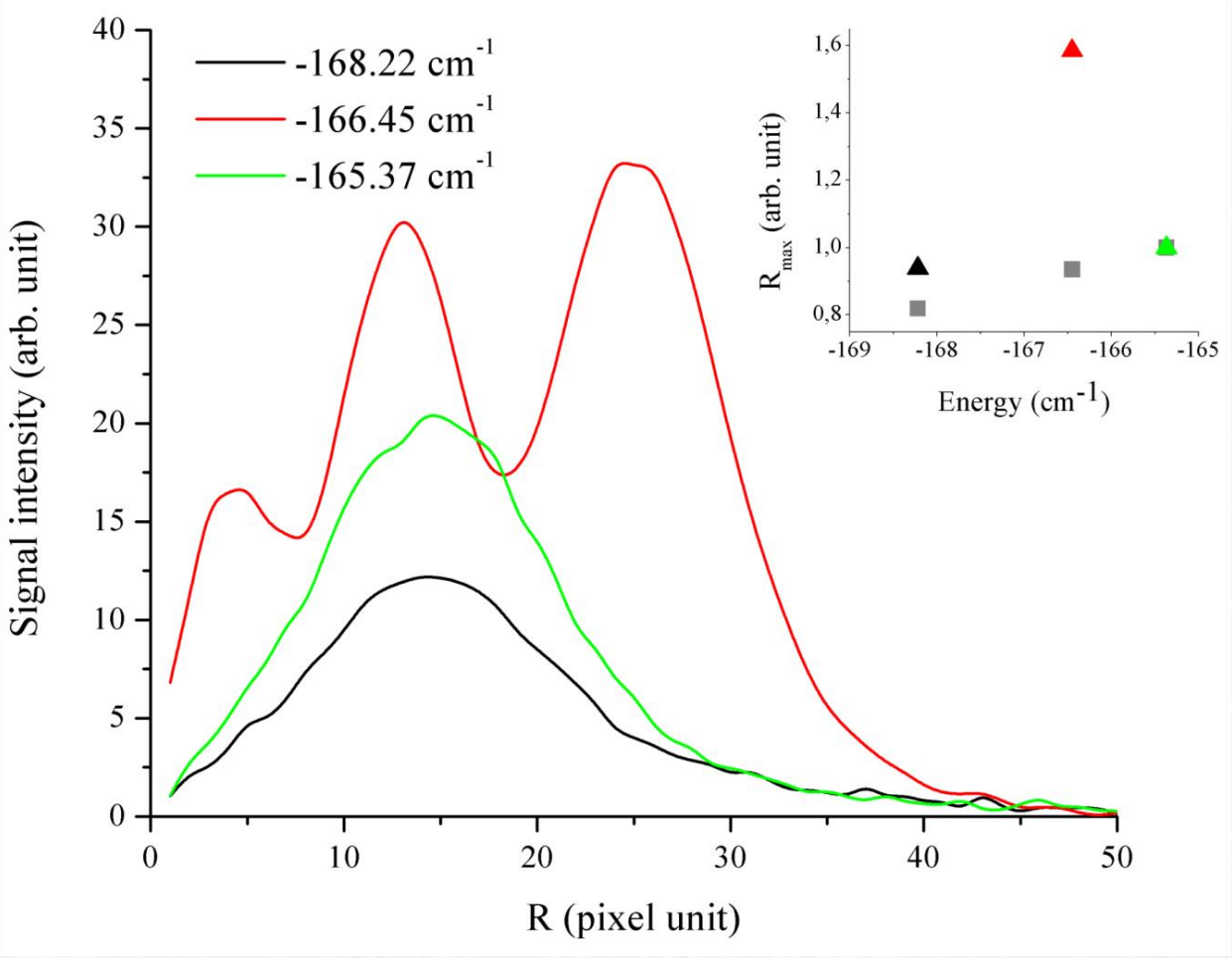
This separation in parabolic coordinates is *independent of the applied electric field* → persists from the interaction region (F ≈ 500 V/cm) to the two-dimensional detector (F = 0 V/cm)

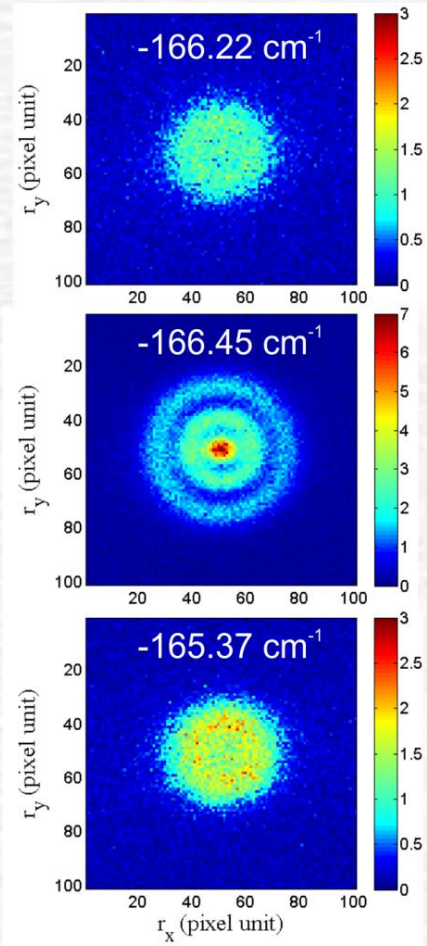


PRL 110, 213001 (2013)
Selected for a Viewpoint in *Physics*
PHYSICAL REVIEW LETTERS
week ending
24 MAY 2013

Hydrogen Atoms under Magnification: Direct Observation of the Nodal Structure of Stark States

A. S. Stodolna,[1,*] A. Rouzée,[1,2] F. Lépine,[3] S. Cohen,[4] F. Robicheaux,[5]
A. Gijsbertsen,[1] J. H. Jungmann,[1] C. Bordas,[3] and M. J. J. Vrakking[1,2,*]





→ can recognize n_1, related to quantization in ξ coordinate

PRL 110, 213001 (2013)

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PHYSICAL REVIEW LETTERS

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Hydrogen Atoms under Magnification: Direct Observation of the Nodal Structure of Stark States

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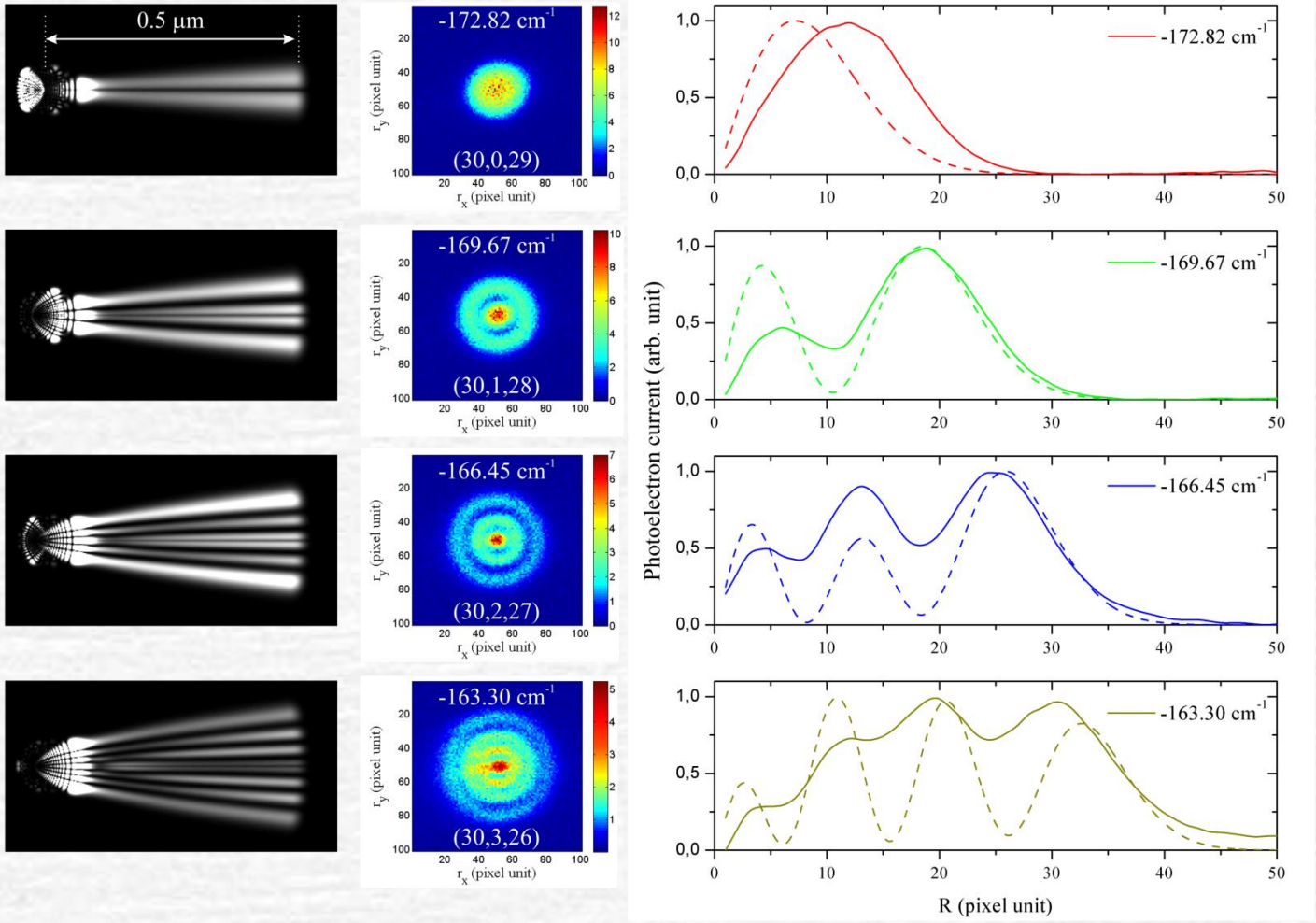


And then came Facebook…

Qhathryn Sedai **This is amazing! I finally have a good answer for my Jr. High students that always ask "how do we really know an atom goes like that?" I have a real picture to show them!**

Michal Kowalski **Did hydrogen atom ever expected to have it's photo posted on Facebook? Too late my little original friend, your face is out and we all see you for what you are**

Itzael Tamayo **I can't even take a pic of a flying bird and they take pics of H atoms? WTF!?!?!?**

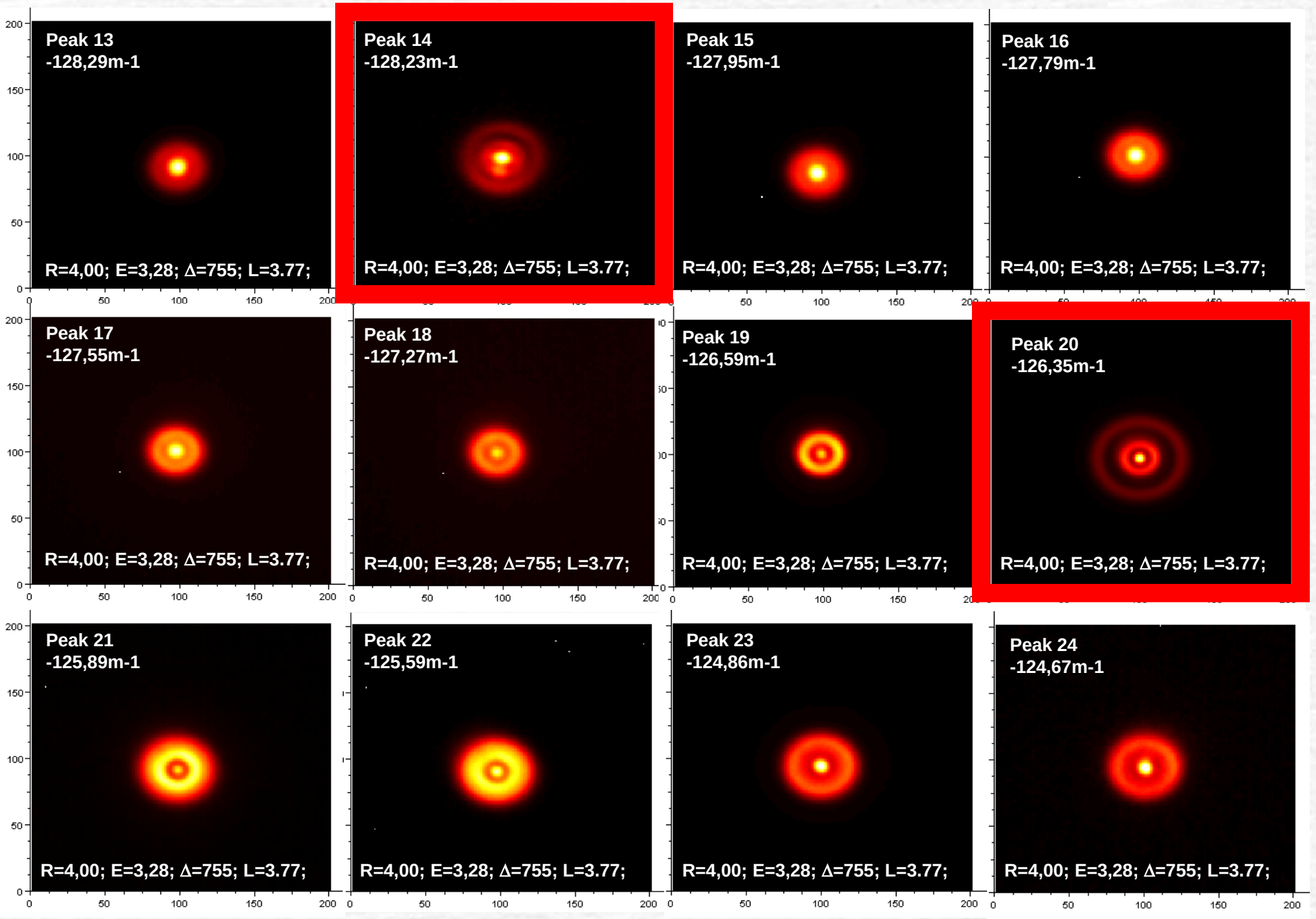
Jenna Burns **This is really awesome, but shit like that is going to eventually open a portal to hell and then we're all lost.**

Dawn Doxey **Quantum Microscope! I would like to know if it could be used to investigate the mutation of cells, and in particular, if any foreign like cancerous or infectious - causing were detected, then they would be graciously escorted out of the human body (go find another place to " party "). This Quantum Microscope seems to be that. Powerful!**

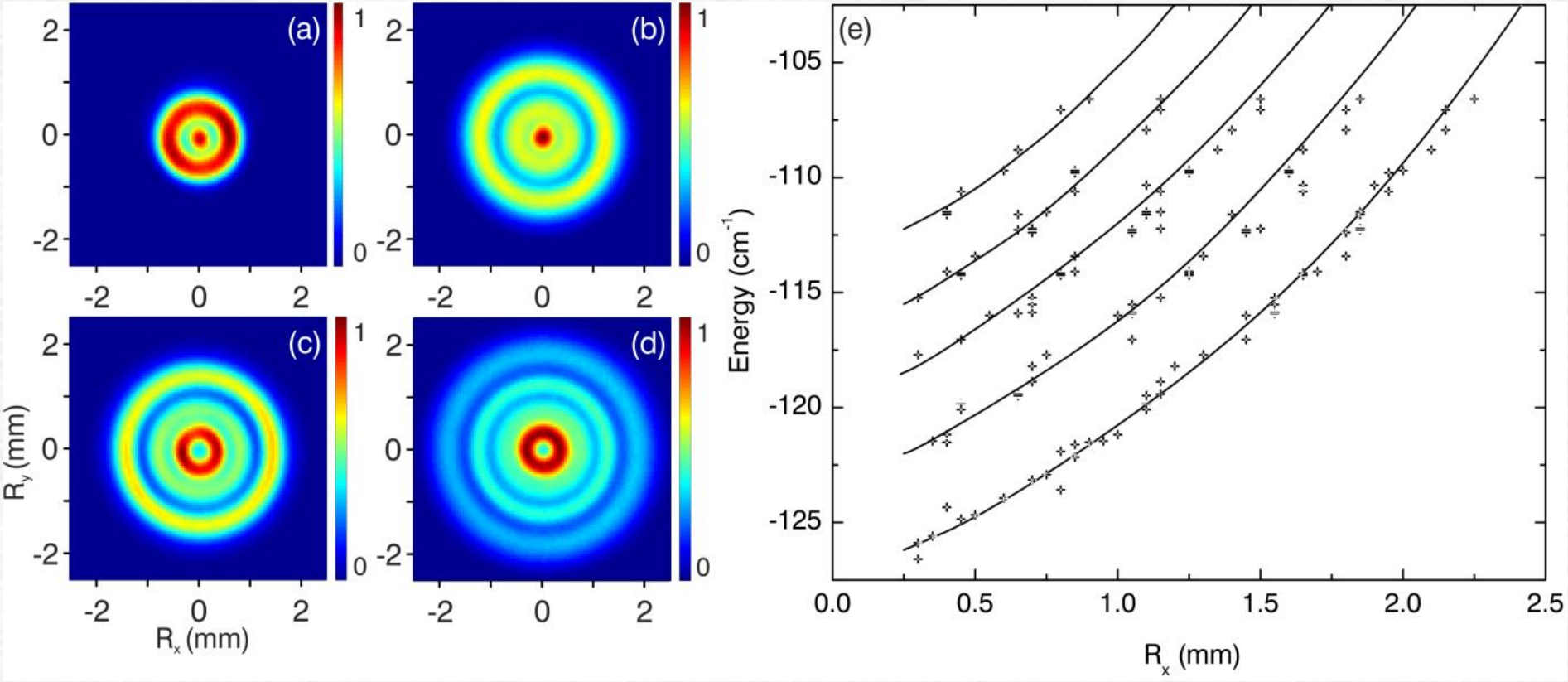
Christopher Atanasopulo **imagine if in a few decades we will be buying our kids science kits featuring working toy quantum microscopes...it would be so cool to be a kid that day! lol**

Part 2

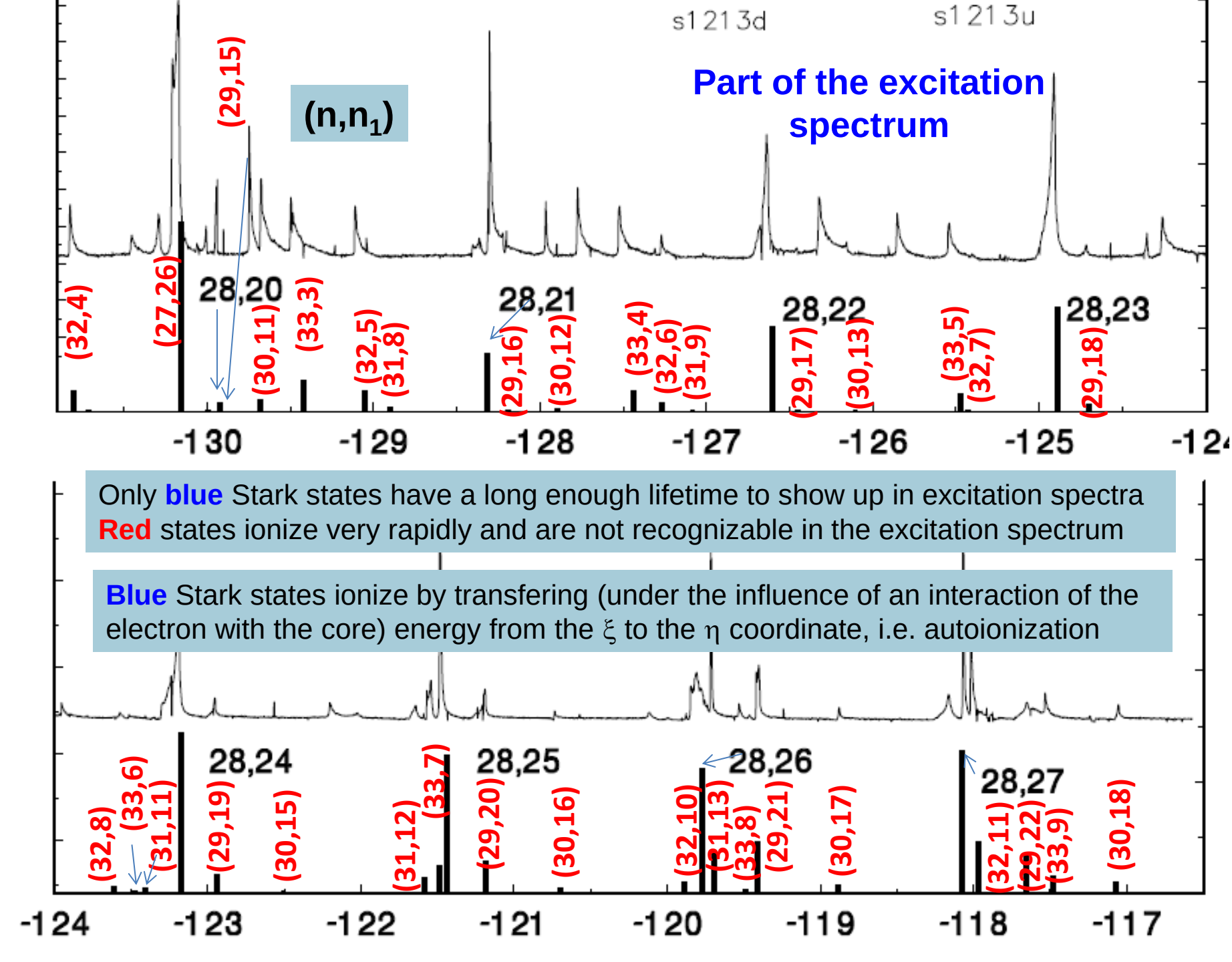
Resonant Photoionization microscopy of He atoms


Peak 13
-128,29m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 14
-128,23m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 15
-127,95m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 16
-127,79m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 17
-127,55m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 18
-127,27m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 19
-126,59m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 20
-126,35m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 21
-125,89m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 22
-125,59m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 23
-124,86m-1
R=4,00; E=3,28; Δ=755; L=3.77;
Peak 24
-124,67m-1
R=4,00; E=3,28; Δ=755; L=3.77;

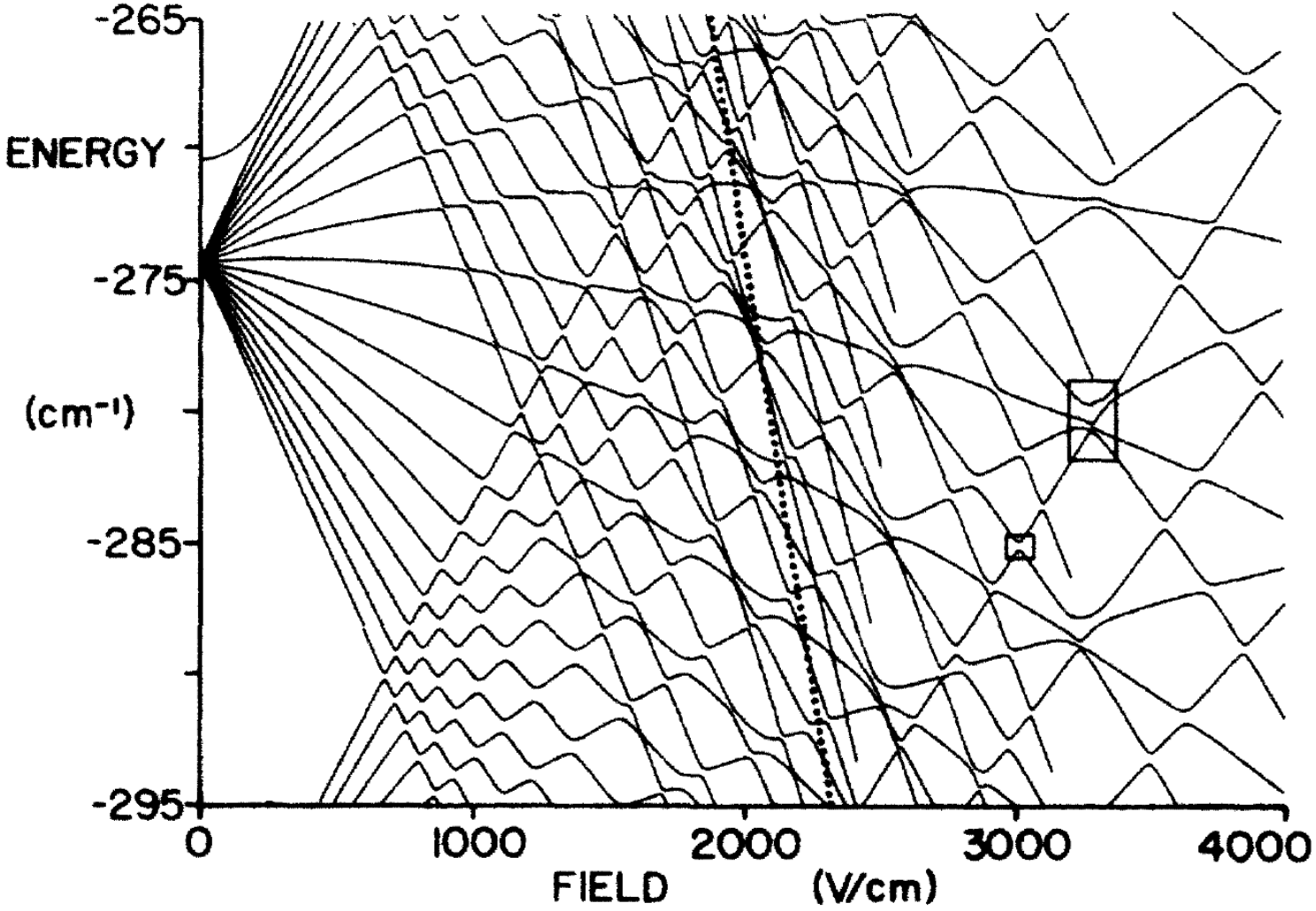
For most of the measurements Helium behaves like Xenon



But some measurements behave very differently...


s1213d
s1213u
(n,n1)
(29,15)
Part of the excitation spectrum
(32,4)
(27,26)
28,20
(30,11)
(33,3)
(32,5)
(31,8)
28,21
(29,16)
(30,12)
(33,4)
(32,6)
(31,9)
28,22
(29,17)
(30,13)
(33,5)
(32,7)
28,23
(29,18)
-130
-129
-128
-127
-126
-125
-124
Only blue Stark states have a long enough lifetime to show up in excitation spectra
Red states ionize very rapidly and are not recognizable in the excitation spectrum
Blue Stark states ionize by transfering (under the influence of an interaction of the electron with the core) energy from the ξ to the η coordinate, i.e. autoionization
(32,8)
(33,6)
(31,11)
28,24
(29,19)
(30,15)
(31,12)
(33,7)
28,25
(29,20)
(30,16)
(32,10)
28,26
(31,13)
(33,8)
(29,21)
(30,17)
28,27
(32,11)
(29,22)
(33,9)
(30,18)
-124
-123
-122
-121
-120
-119
-118
-117

Avoided crossings in the Starkmap of Helium



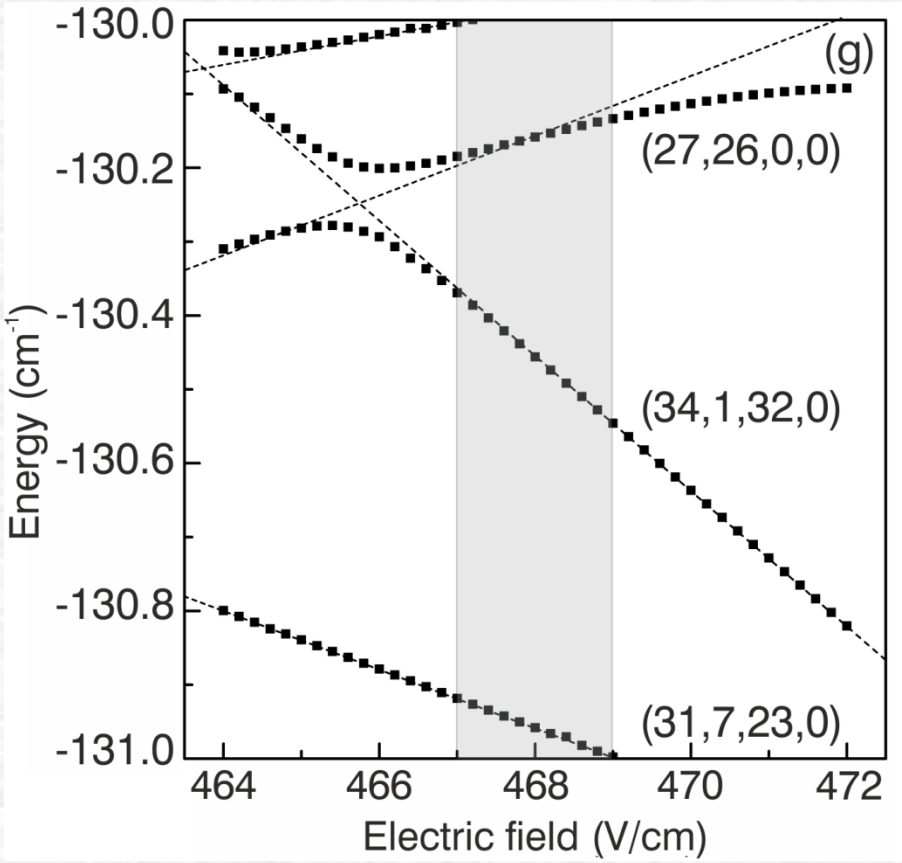
Interaction of red and blue Stark states near an avoided crossing

Near an avoided crossing, two new states emerge that can be written as

$$\Psi_1 = \cos\alpha(F)\,\Psi_{blue} + \sin\alpha(F)\,\Psi_{red}$$
$$\Psi_2 = -\sin\alpha(F)\,\Psi_{blue} + \cos\alpha(F)\,\Psi_{red}$$

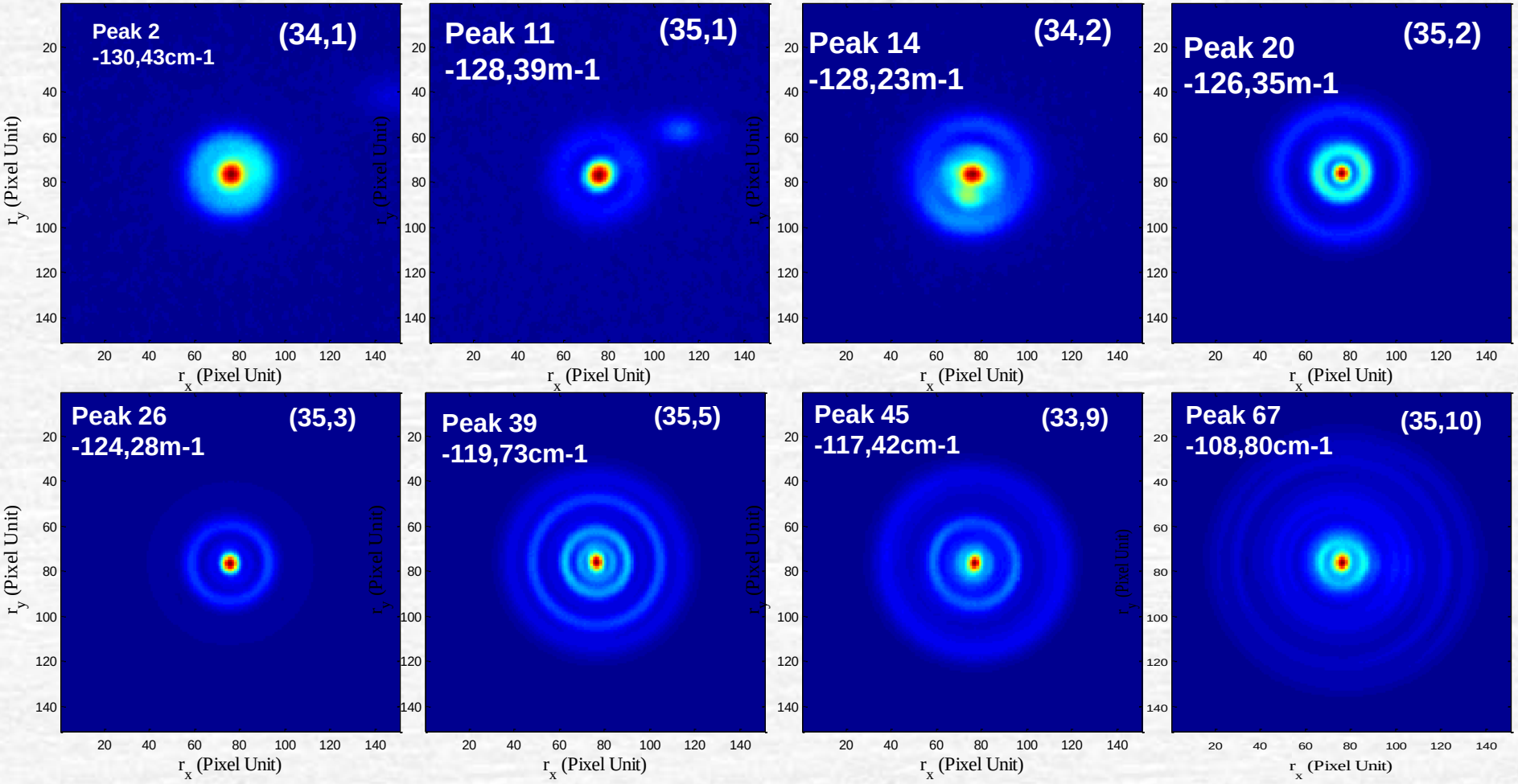
The decay of these states is given by

$$\Gamma = 2\pi \langle \Psi_{1,2} | V | \Psi_{cont} \rangle$$



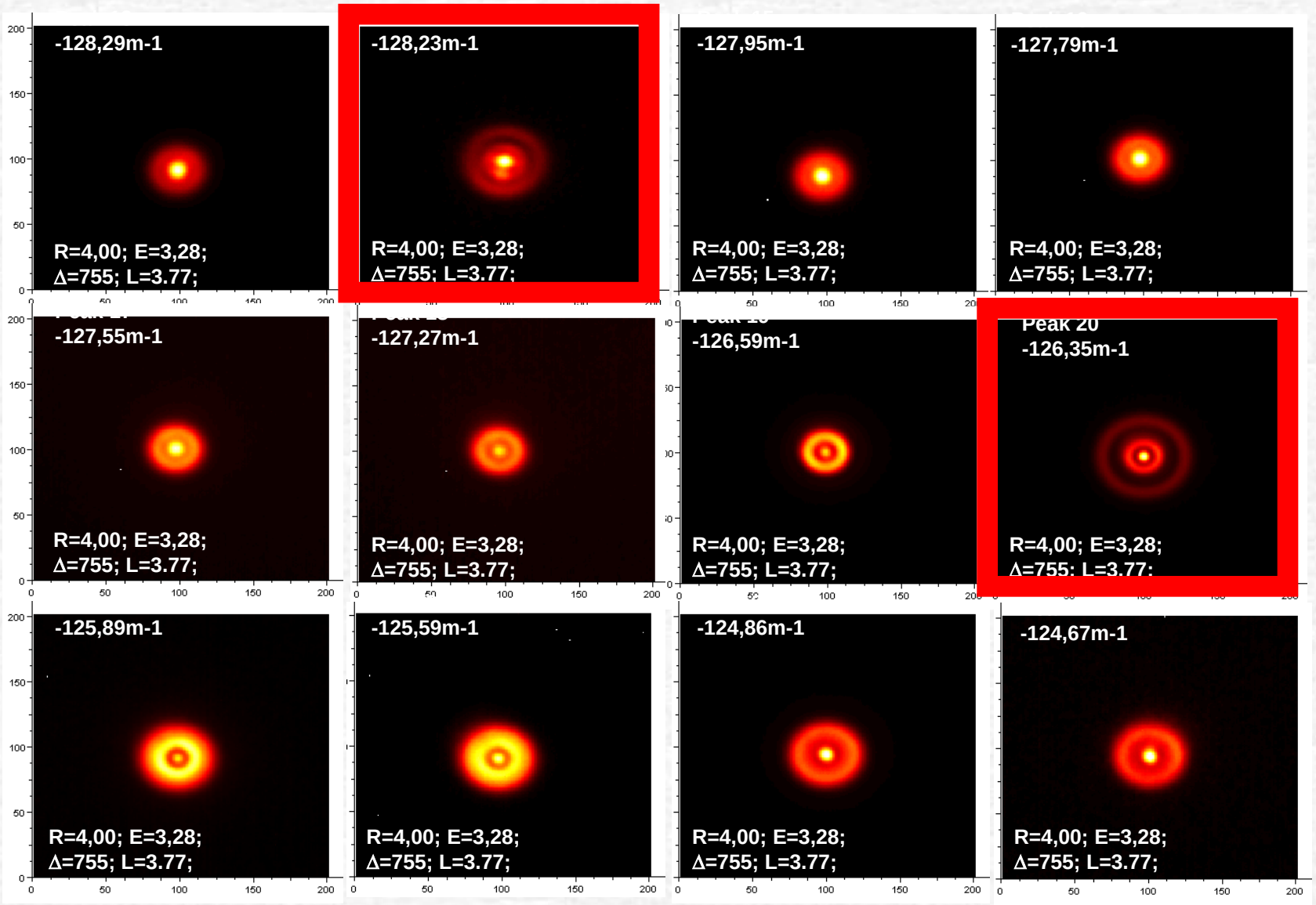
For some value of the fieldstrength the contributions of the red and blue state may cancel each other and the short-lived state acquires a long lifetime

“Hydrogen-like measurements in He: 8 “special” peaks (n,n_1)



A. Stodolna et al., Phys. Rev. Lett. (in press)

Red Stark states ionize by tunneling through the barrier in the V(η) potential energy curve

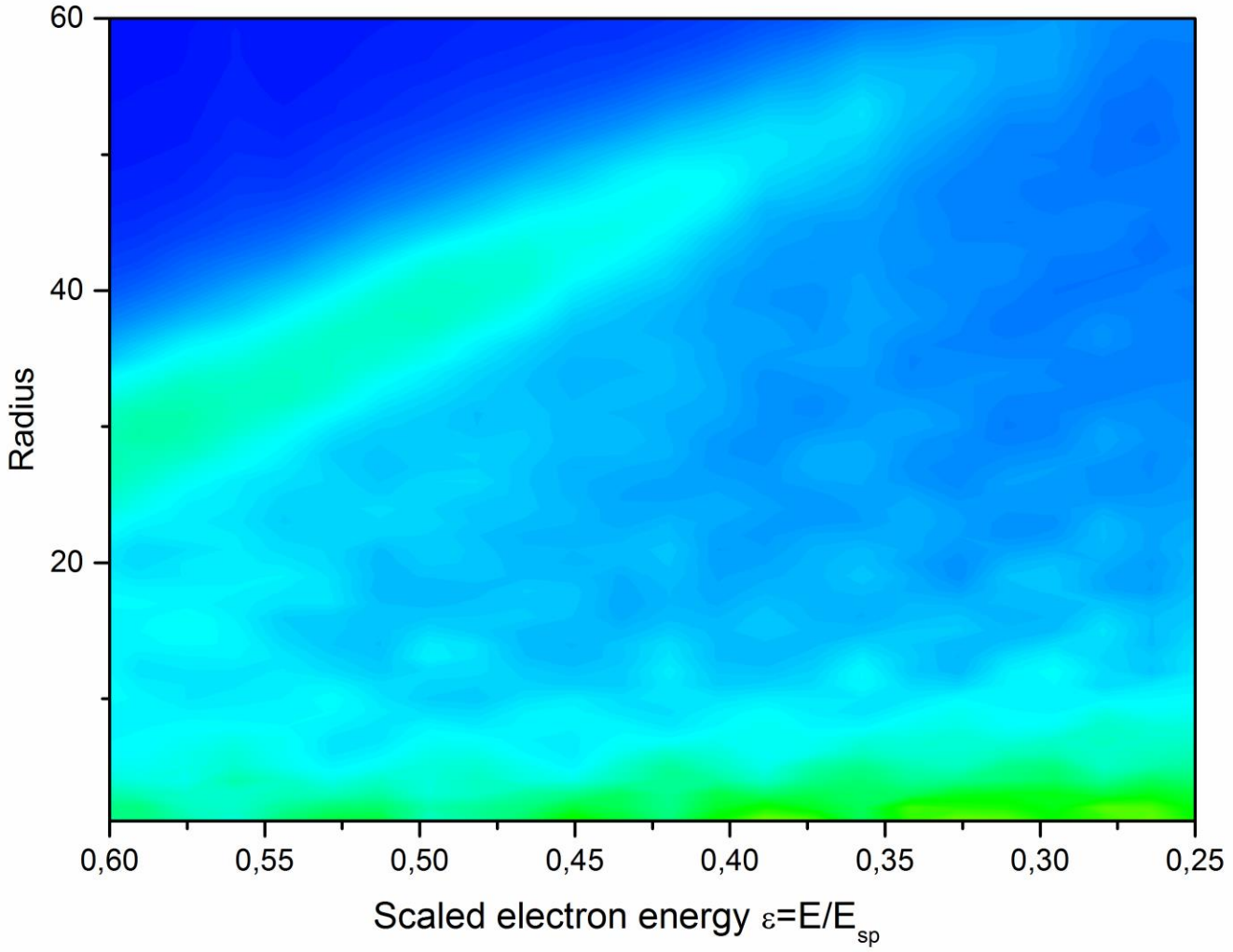


Blue Stark states ionize by transfering (under the influence of an interaction of the electron with the core) energy from the ξ to the η coordinate, i.e. autoionization

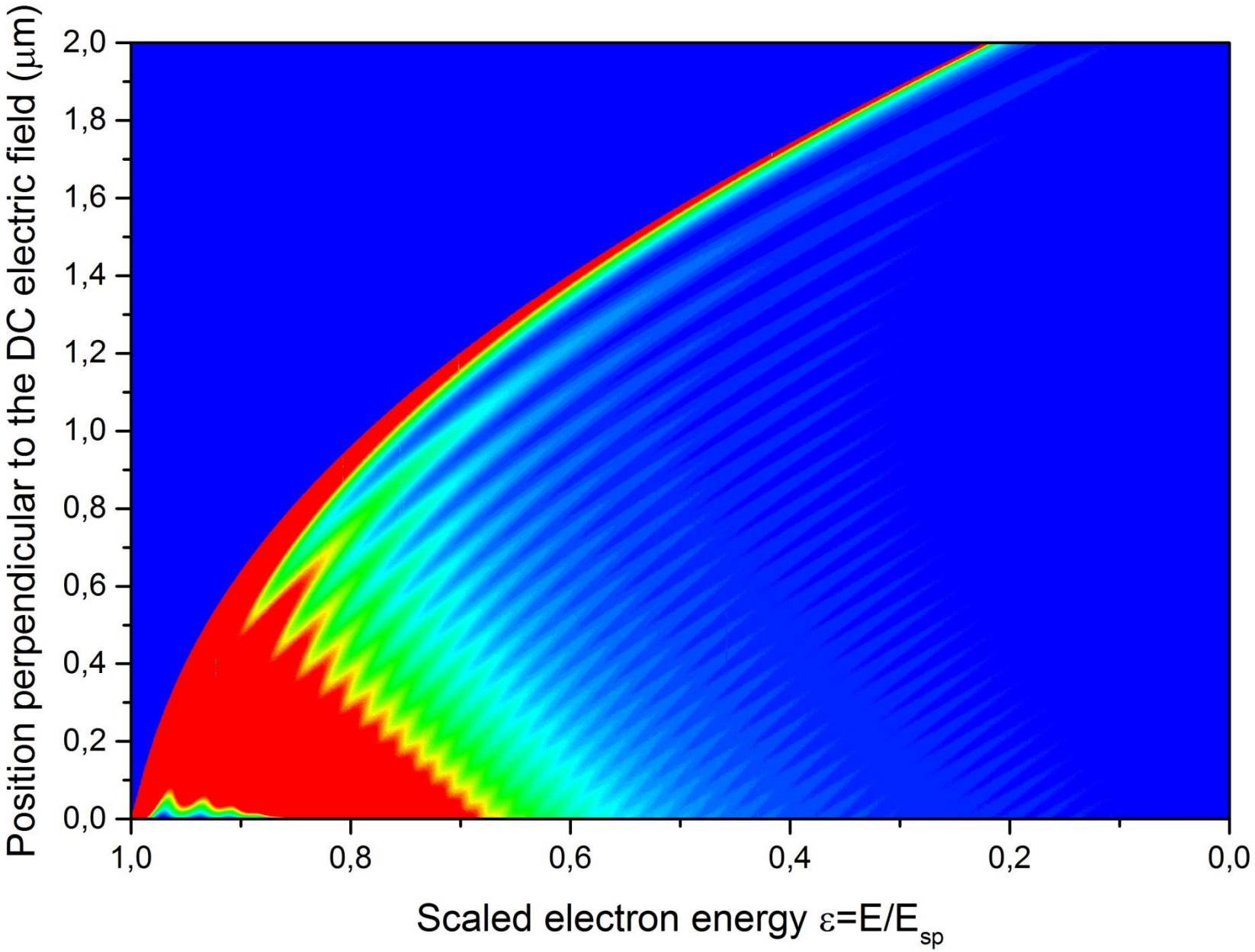
Part 3

Non-Resonant Photoionization microscopy of Hydrogen atoms

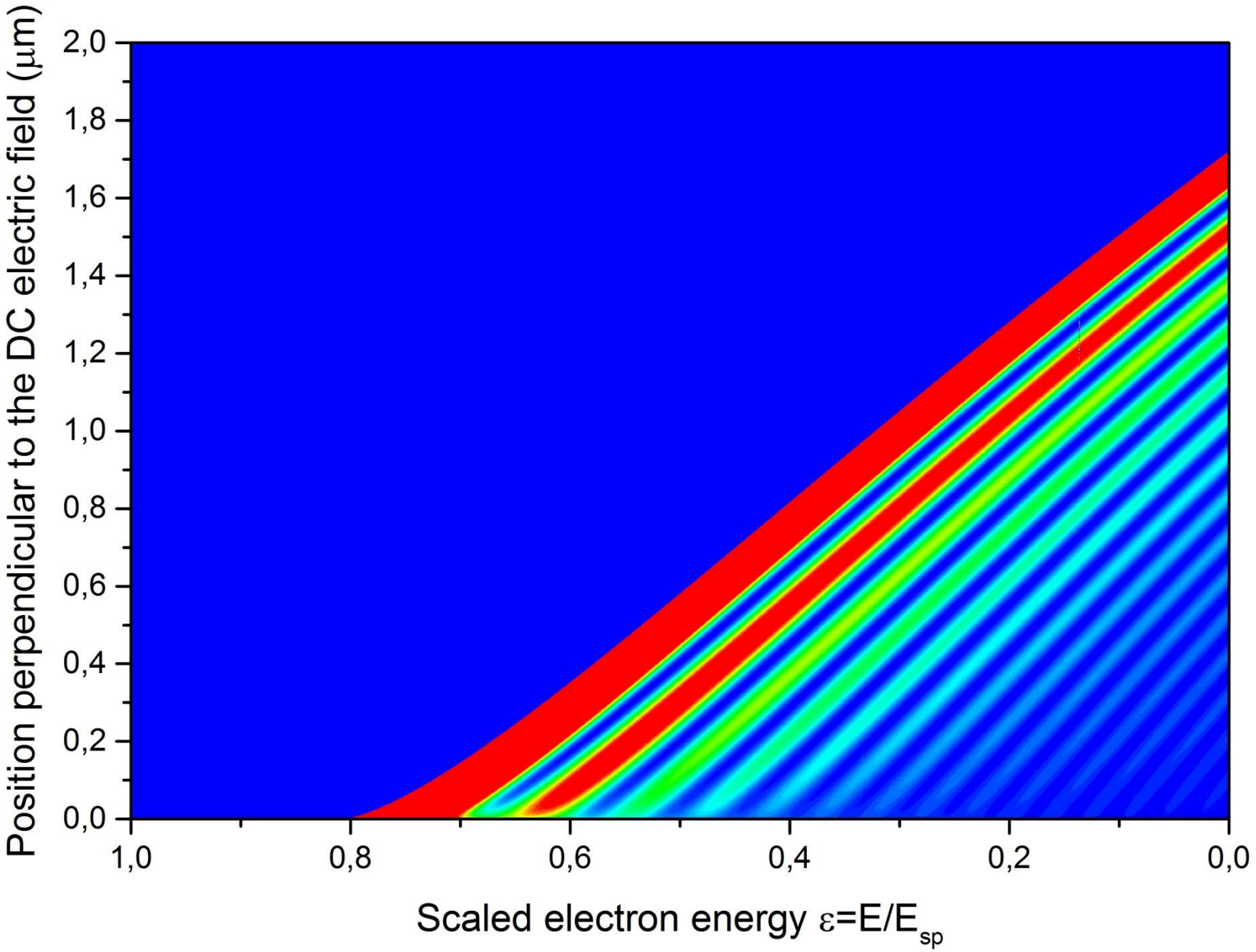
Back to hydrogen: Interference between direct and indirect trajectories



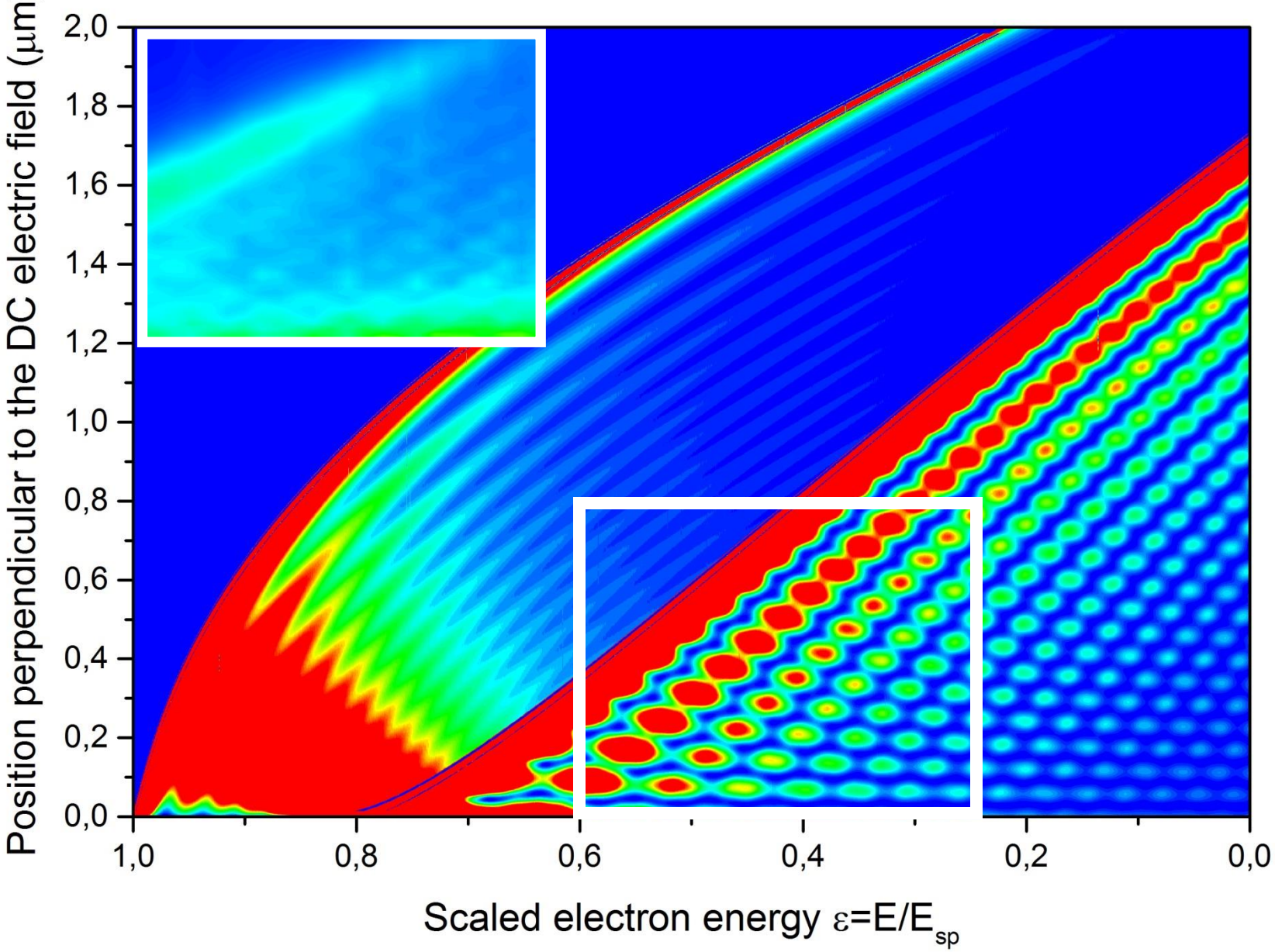
At each energy there are two dominent indirect trajectories



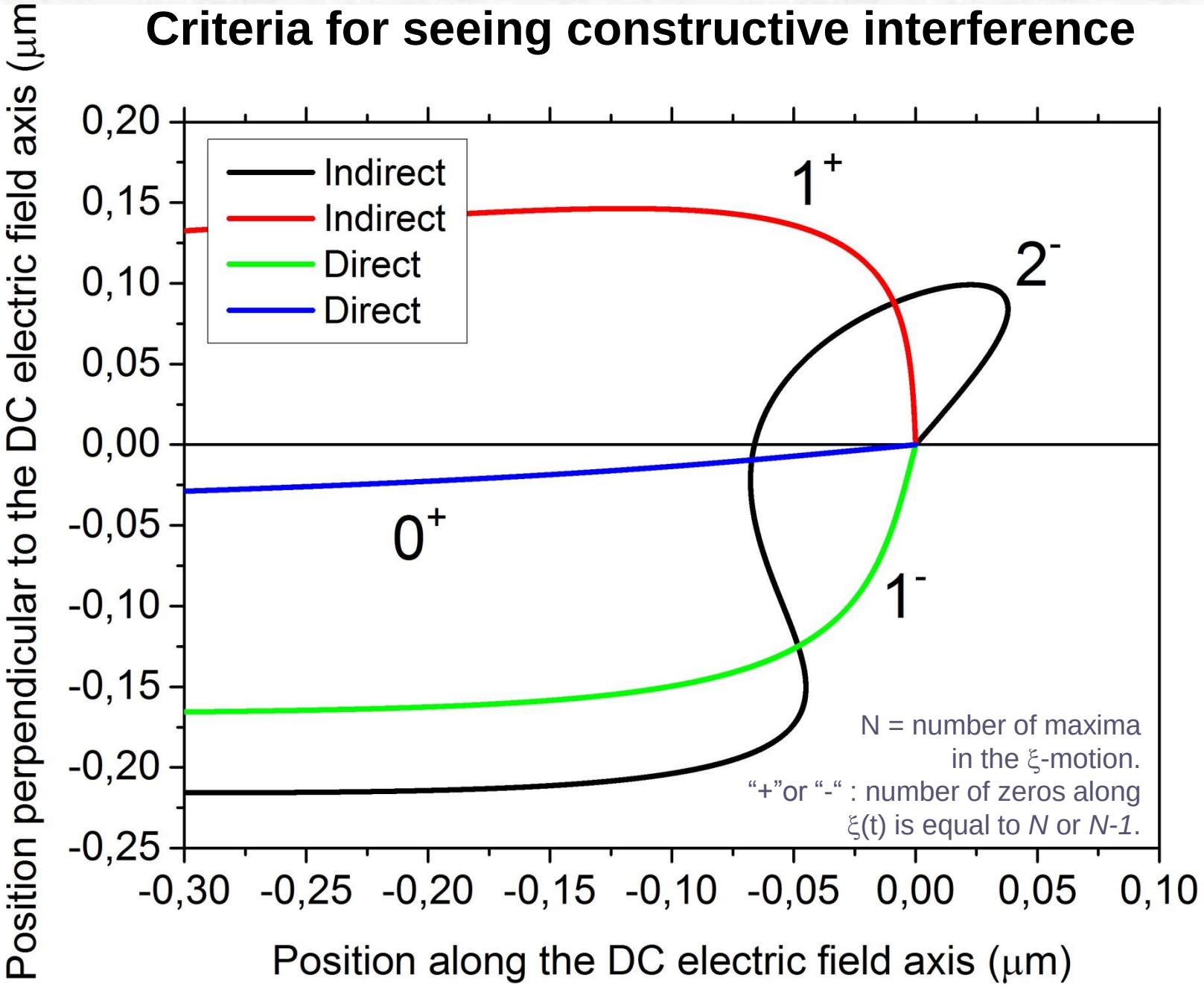
At each energy there are two dominent direct trajectories



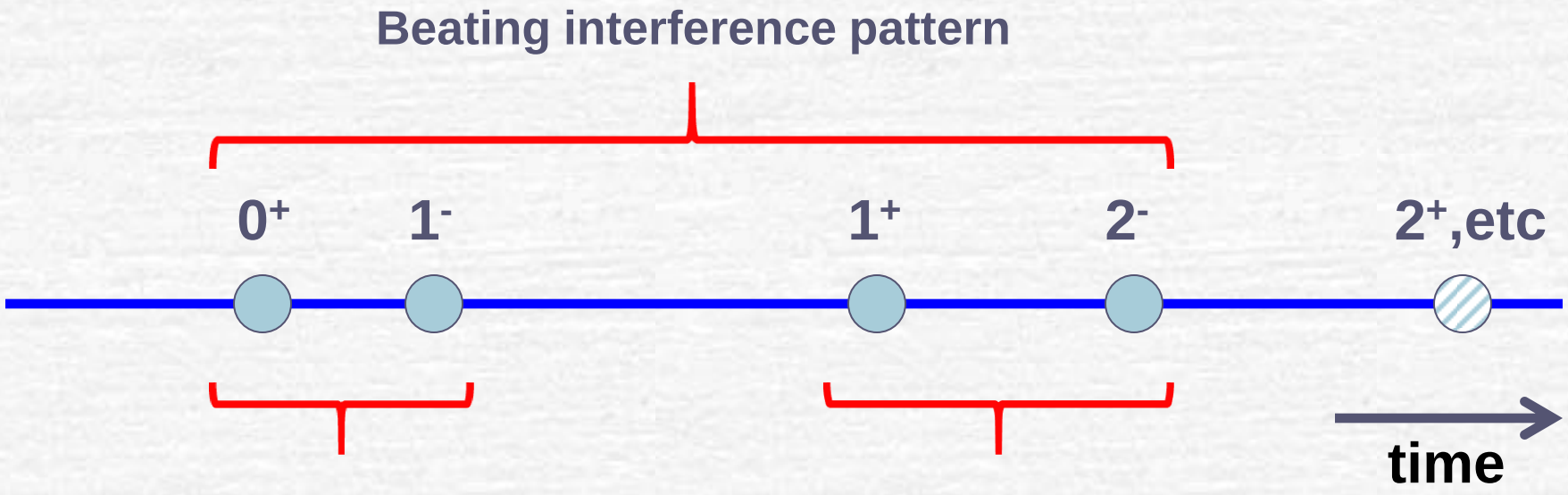
Beating between the interference patterns „measures“ the time delay between direct and indirect trajectories



Criteria for seeing constructive interference

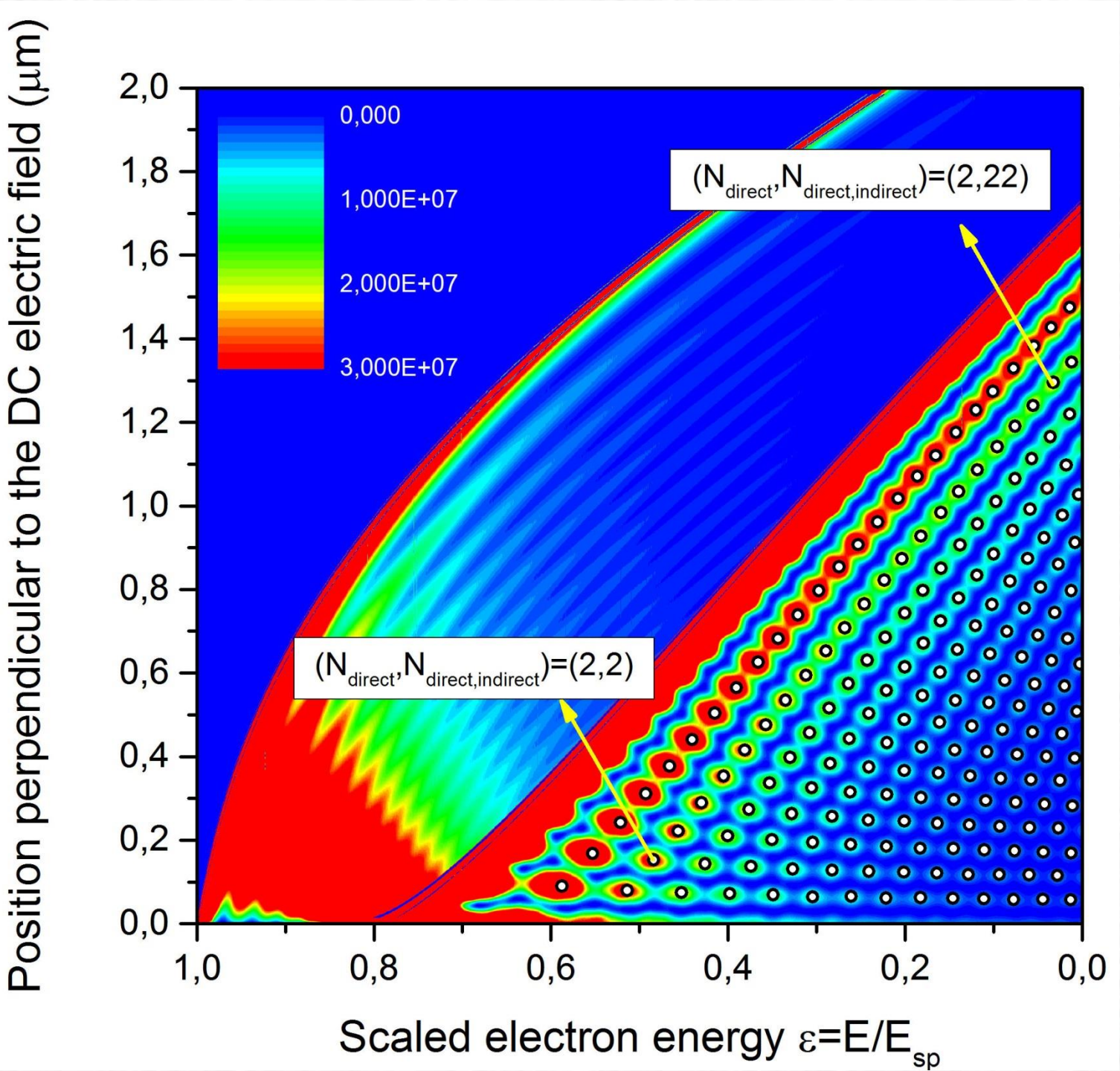


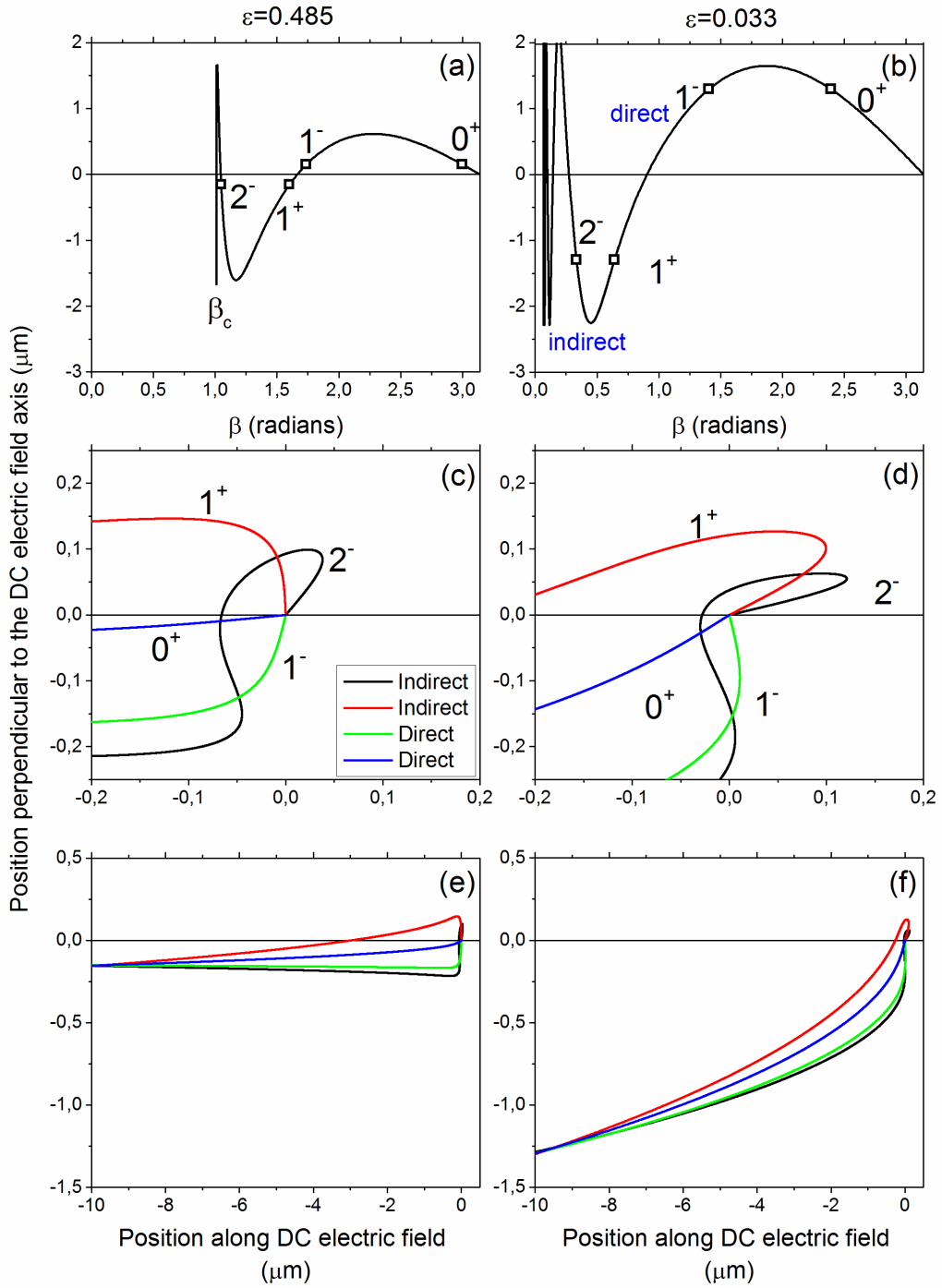
Criteria for seeing constructive interference



Question: with 4 phases (3 relative phases) what is the criterium for observing the beating pattern?

Answer: 0^+ and 1^- have to be in phase (direct fringe) + 1^+ and 1^- have to be in phase (direct-indirect)

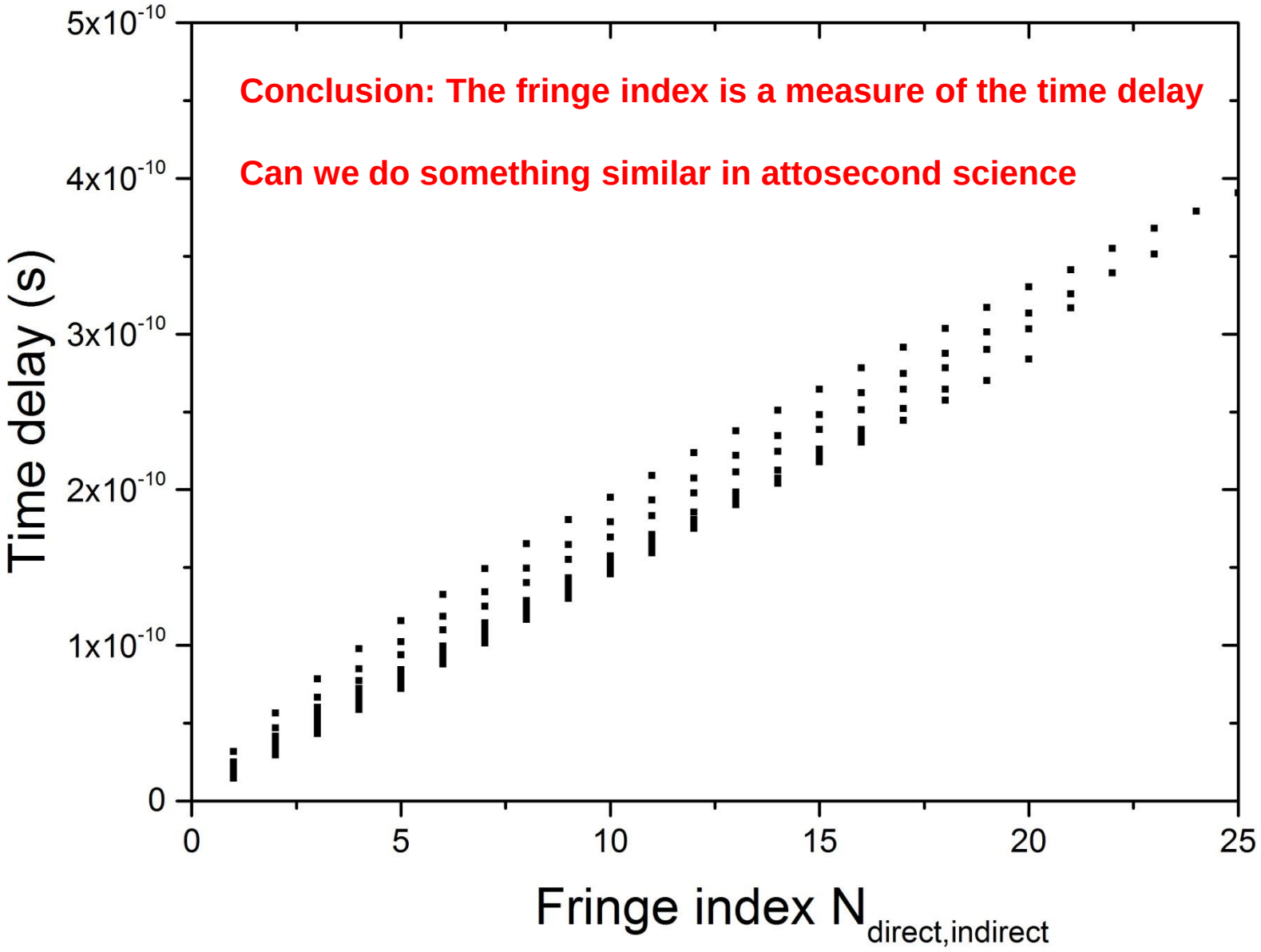

Position perpendicular to the DC electric field (μm)
2,0
1,8
1,6
1,4
1,2
1,0
0,8
0,6
0,4
0,2
0,0
0,000
1,000E+07
2,000E+07
3,000E+07
(N direct, N direct,indirect)=(2,22)
(N direct, N direct,indirect)=(2,2)
1,0
0,8
0,6
0,4
0,2
0,0
Scaled electron energy ε=E/E sp



At each energy there are two dominent direct and two dominent indirect trajectories

Beating between the interference patterns „measures“ the time delay between the 1^+ and the 1^- trajectory

Linear relation between fringe index and time delay



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