Attosecond Pulse Production From Excited Molecules

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Motivation

Mechanisms of attosecond pulse excitation have been understood:

Nonlinear response of atoms and molecules at ionization by superstrong femtosecond pulse

Pulse duration obtained:
- 250 as (R. Kienberger et al., *Nature*, 2004) – single pulse
- 170 as (R. Lopez-Martens et al., *PRL*, 2005) – pulse train

Next step: Search for ways to control efficiency and spectra in attosecond pulse production (especially single attosecond pulse)

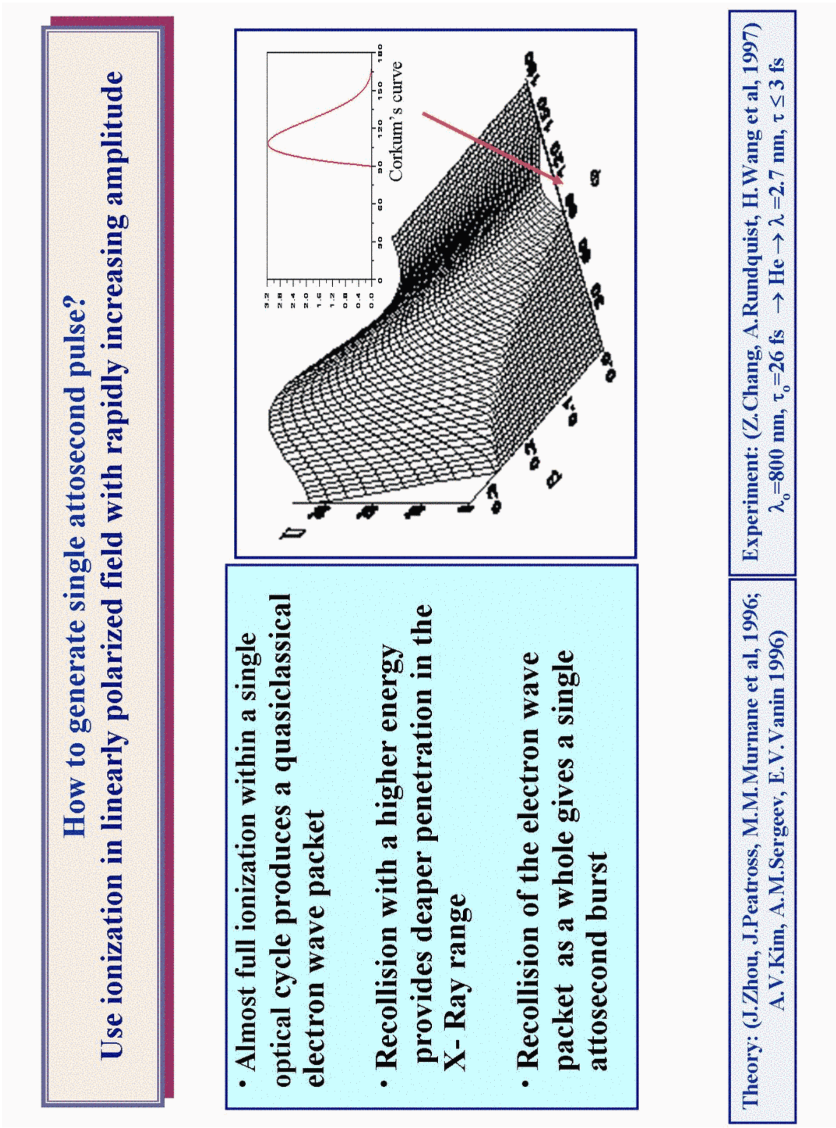
One of the ways:
To use excited atoms and molecules for preparation and control (“Engineering”) of electronic wave packets providing the maximum efficiency and flexibility of bremstrahlung at electron-ion recollision

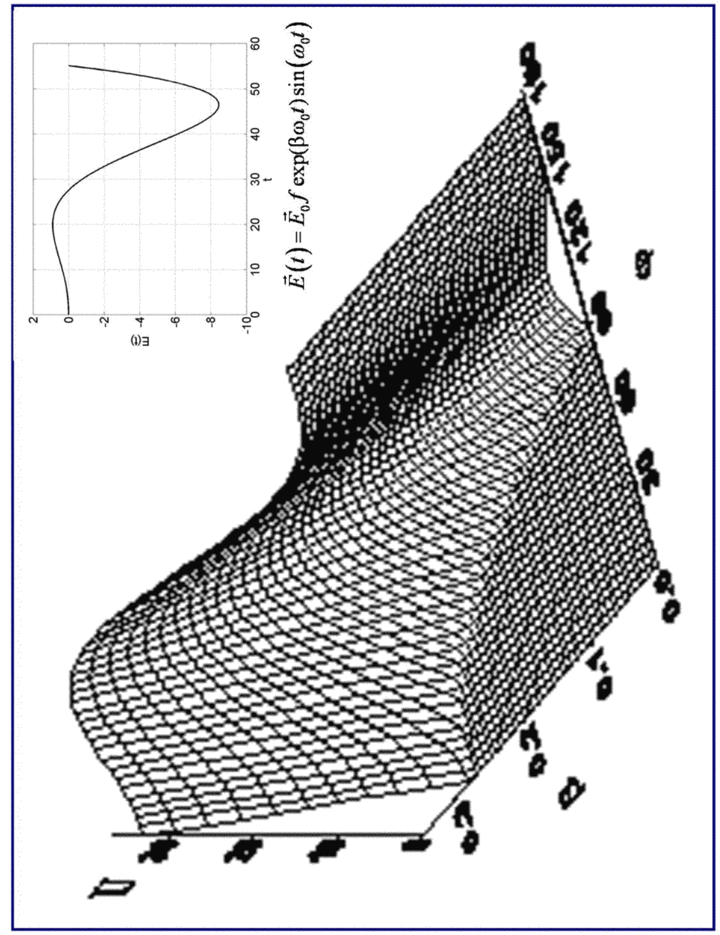
On this road, more classical than quantum-mechanical intuition may sometimes be helpful

To maximize the amount of electrons contributing to bremstrahlung we engage

- Fast single cycle ionization
- Electrons with large collision radii rather than Corkum's electrons
- Initial electronic excitation in atoms
- Vibrational excitation in molecules
- Orientation of molecule against laser field
- Orientation of molecule against direction of pulse propagation
- All together

Fast single cycle ionization


How to generate single attosecond pulse?
Use ionization in linearly polarized field with rapidly increasing amplitude
Corkum's curve
• Almost full ionization within a single optical cycle produces a quasiclassical electron wave packet
• Recollision with a higher energy provides deaper penetration in the X- Ray range
• Recollision of the electron wave packet as a whole gives a single attosecond burst
Theory: (J.Zhou, J.Peatross, M.M.Murnane et al, 1996; A.V.Kim, A.M.Sergeev, E.V.Vanin 1996)
Experiment: (Z.Chang, A.Rundquist, H.Wang et al, 1997) λo=800 nm, τo=26 fs → He → λ =2.7 nm, τ ≤ 3 fs


E(t)
t
$\vec{E}(t)=\vec{E}_0 f\exp(\beta\omega_0 t)\sin(\omega_0 t)$

Electrons with large collision radii

Full 3D modeling – theory and computer experiment

Schroedinger equation

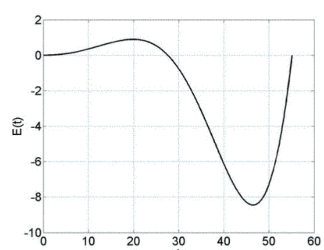
$$i\frac{\partial\Psi}{\partial t} = H\Psi = \frac{1}{2}\left(\vec{p} + \frac{\vec{A}(t)}{c}\right)^2 \Psi + V(x,y,z)\Psi$$

Here

$$\vec{A}(t) = c\int_{-\infty}^{t} \vec{E}(\tau)d\tau; \qquad \vec{E}(t) = \vec{E}_0 f(t)\sin(\omega_0 t)$$

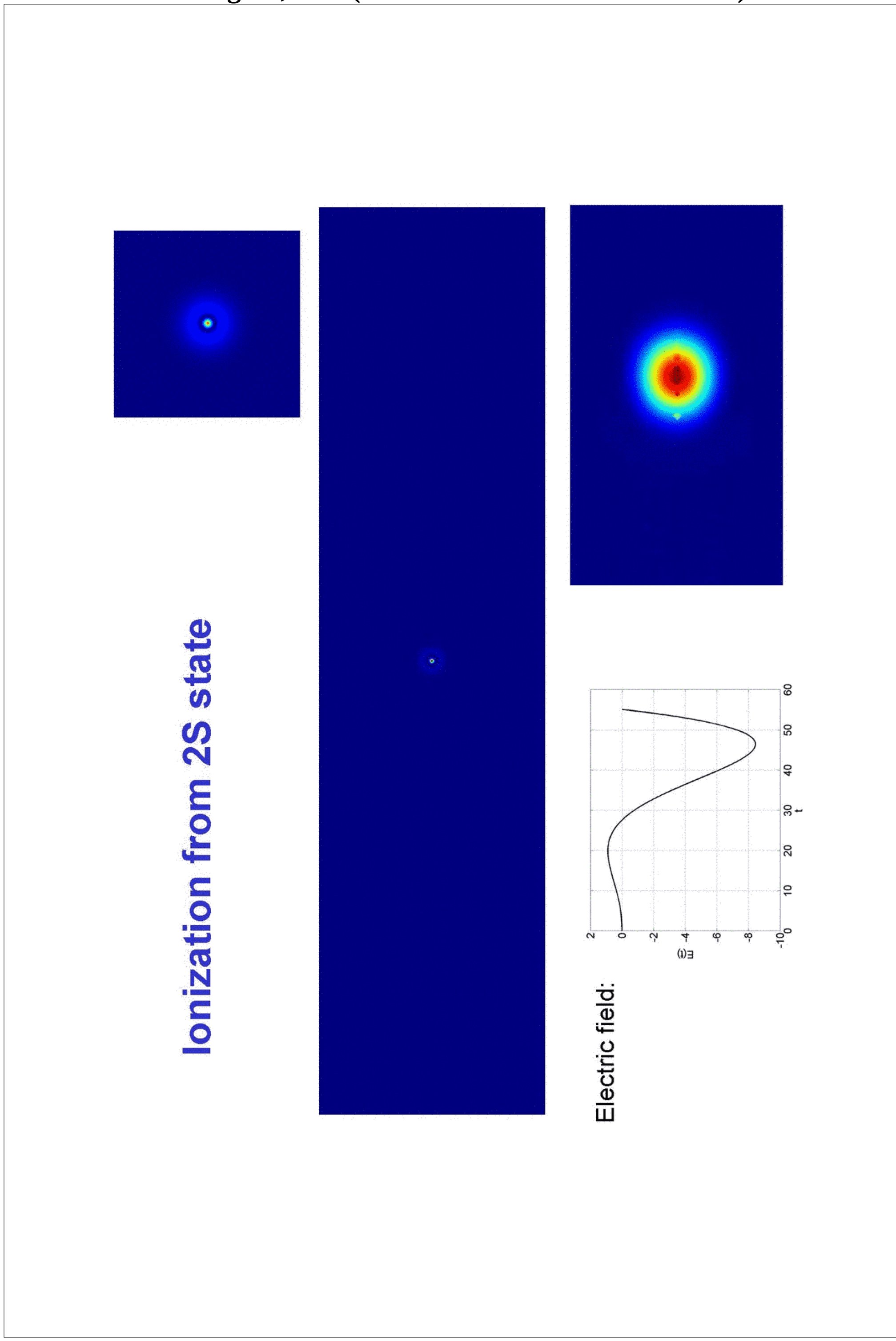
$$f(t) = \exp(2\omega_0 t/\pi); \qquad E_0 = 0{,}36; \qquad \omega_0 = 0{,}114$$

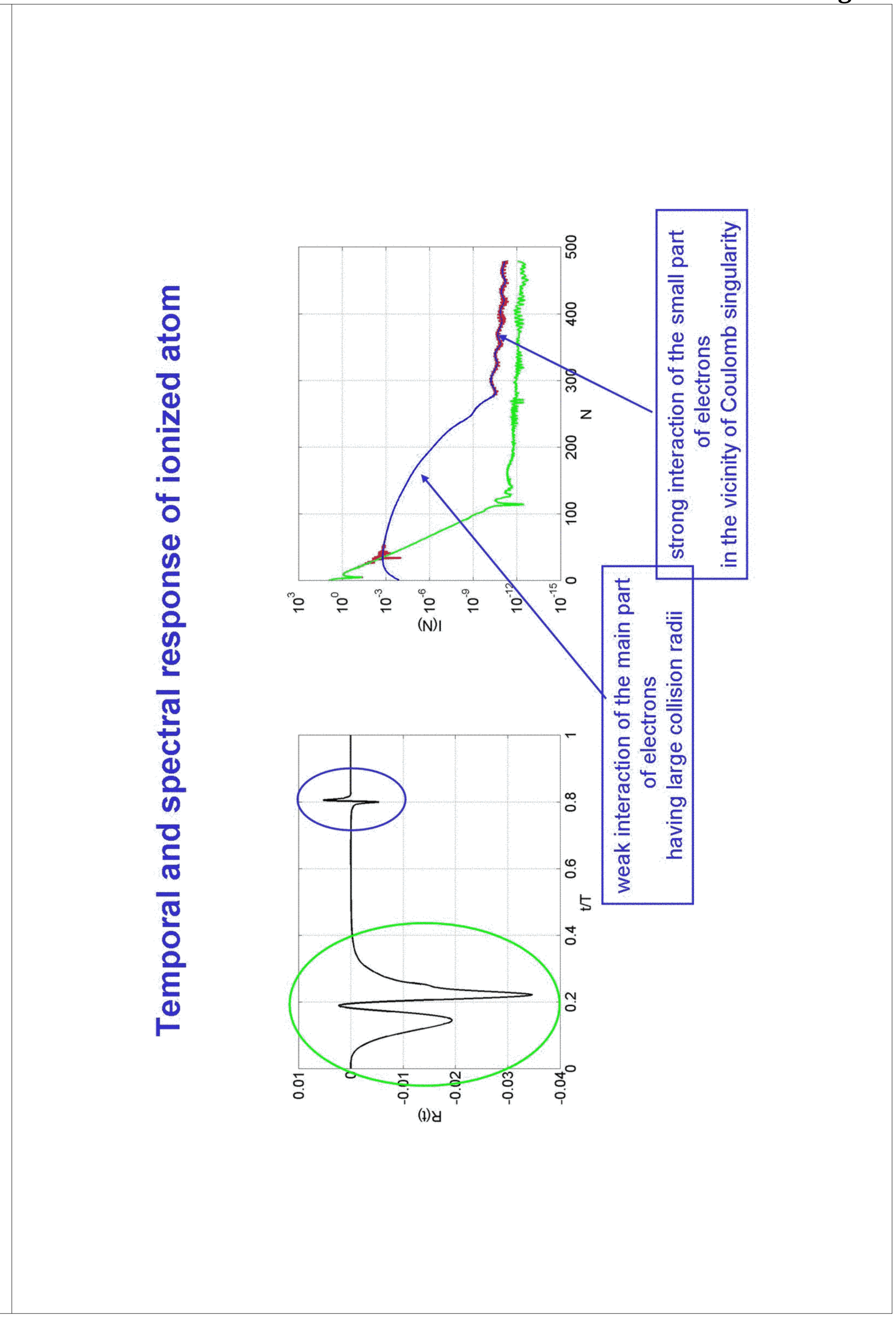
Electric field:



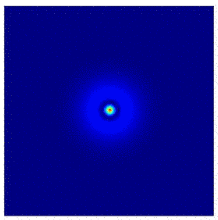
Potential:

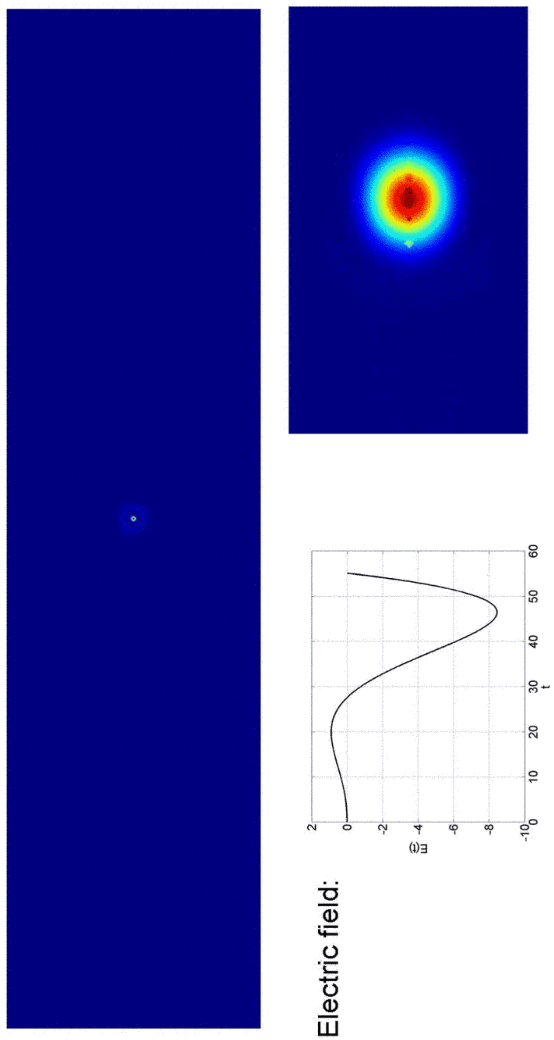
$$V(x,y,z) = -\left(x^2 + y^2 + z^2\right)^{-1/2}$$


Ionization from 2S state
Electric field:
E(t)
t


Temporal and spectral response of ionized atom
R(t)
t/T
N
weak interaction of the main part
of electrons
having large collision radii
strong interaction of the small part
of electrons
in the vicinity of Coulomb singularity

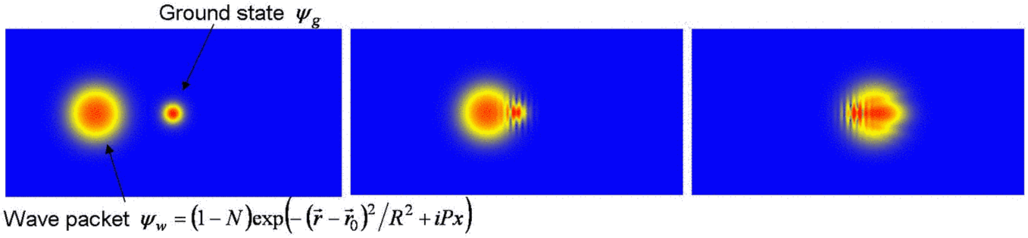
Ionization from 2S state



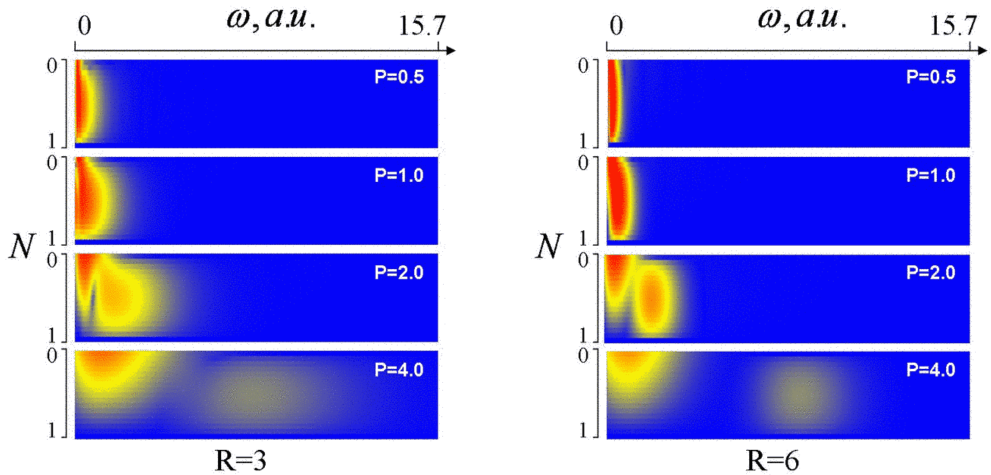


Electrons with large collision radii
versus
Corkum's electrons ?

Also, A.Pukhov et al, 2003

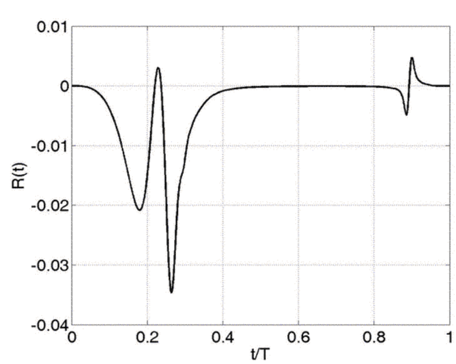


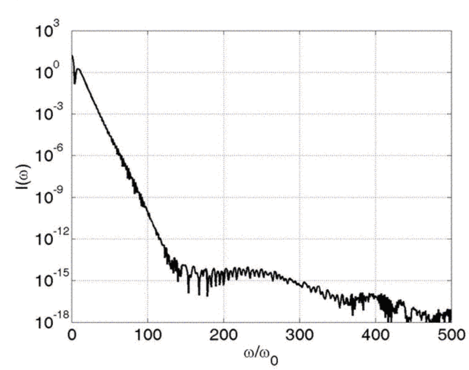
Radiation spectrum for $\left|N\psi_g + (1-N)\psi_w\right|^2$

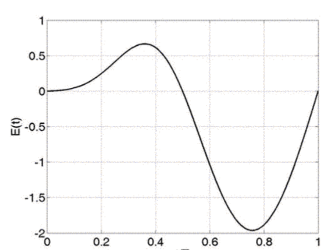


Temporal and spectral response of ionized atom

$$E(t) = E_0 \exp\left[-5\cdot\left(\frac{\omega_0 t}{2\pi} - 1\right)^4\right]\sin(\omega_0 t) \qquad E_0 = 2$$

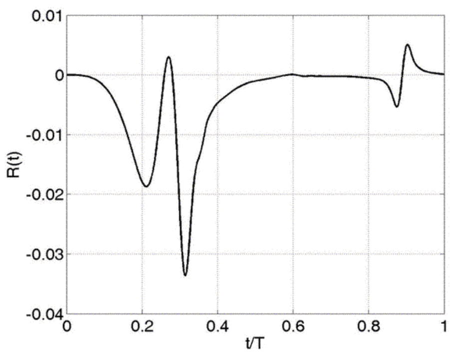


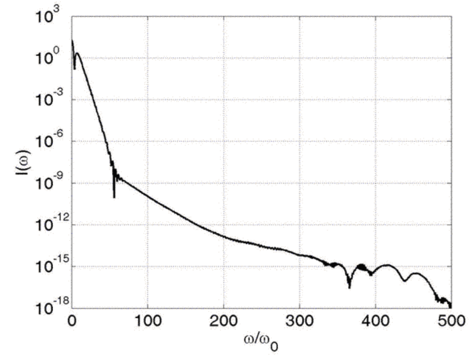


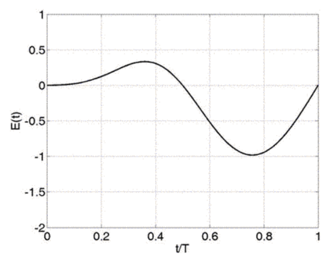


Temporal and spectral response of ionized atom

$$E(t) = E_0 \exp\left[-5\cdot\left(\frac{\omega_0 t}{2\pi}-1\right)^4\right]\sin(\omega_0 t) \qquad E_0 = 1$$

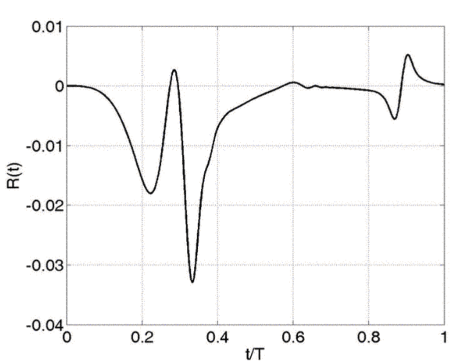


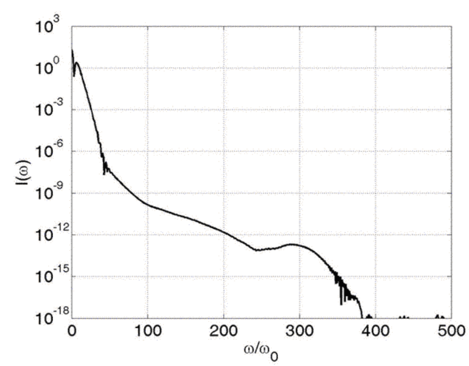


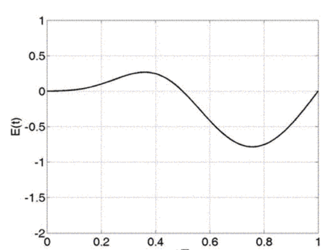


Temporal and spectral response of ionized atom

$$E(t) = E_0 \exp\left[-5\cdot\left(\frac{\omega_0 t}{2\pi}-1\right)^4\right]\sin(\omega_0 t) \qquad E_0 = 0.8$$

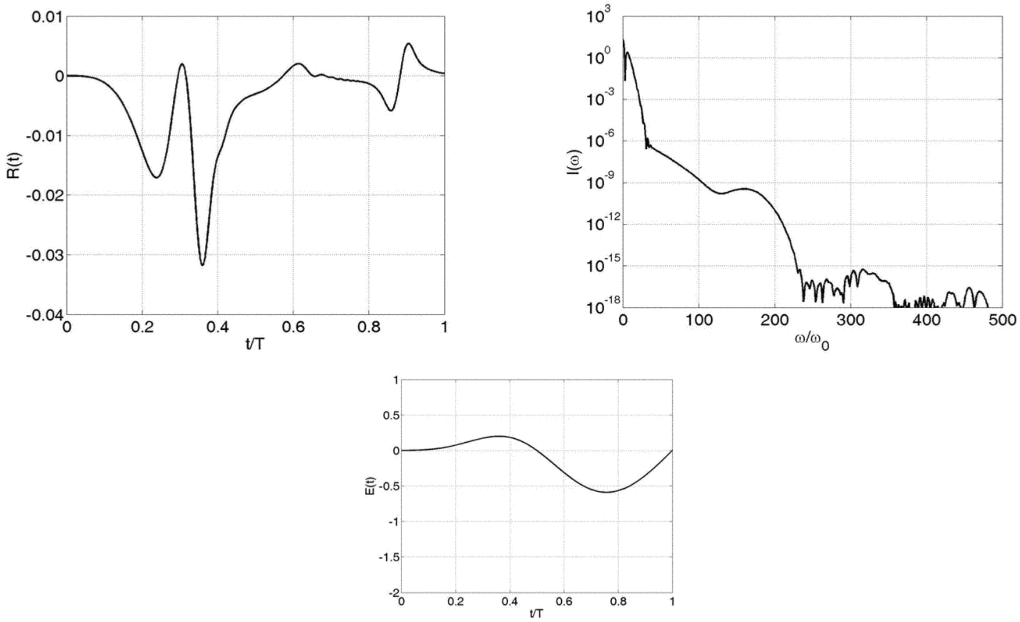






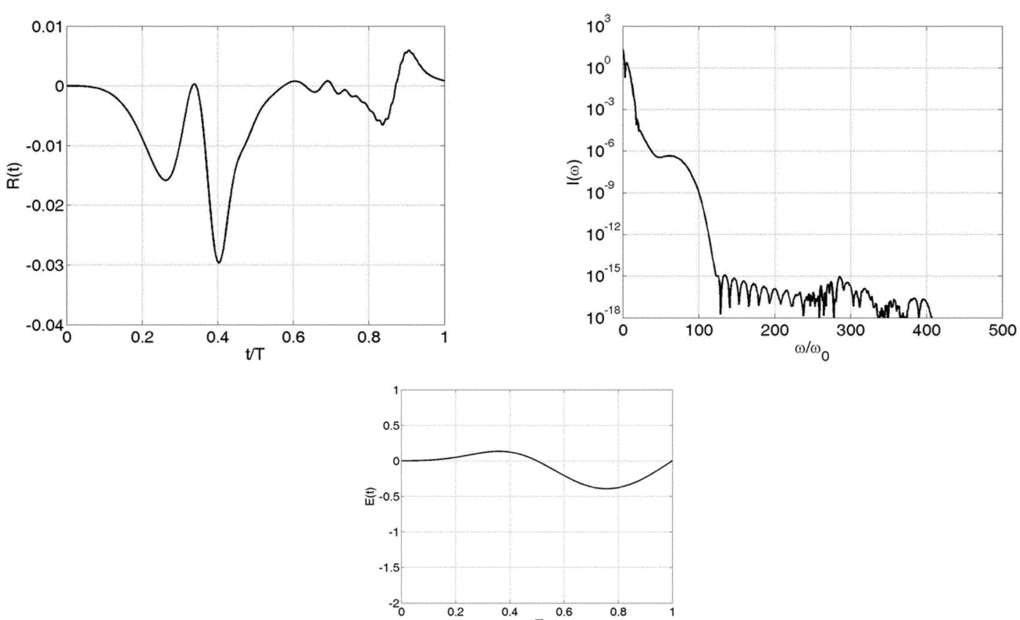
Temporal and spectral response of ionized atom

$$E(t) = E_0 \exp\left[-5\cdot\left(\frac{\omega_0 t}{2\pi}-1\right)^4\right]\sin(\omega_0 t) \qquad E_0 = 0.6$$



Temporal and spectral response of ionized atom

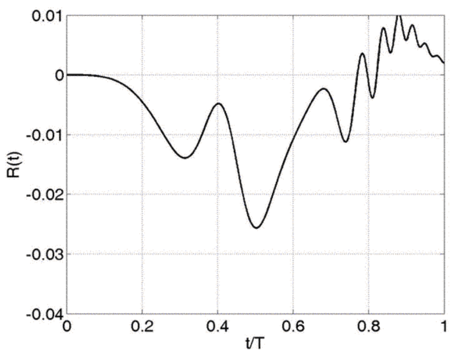
$$E(t) = E_0 \exp\left[-5\cdot\left(\frac{\omega_0 t}{2\pi}-1\right)^4\right]\sin(\omega_0 t) \qquad E_0 = 0.4$$

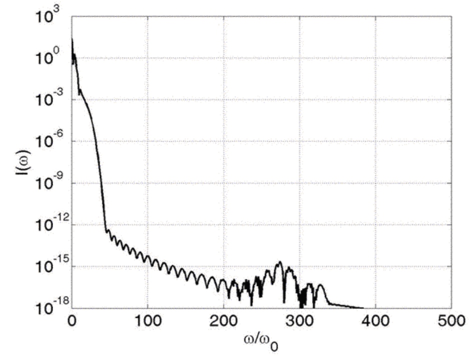


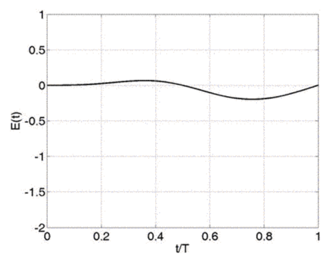
Temporal and spectral response of ionized atom

$$E(t) = E_0 \exp\left[-5\cdot\left(\frac{\omega_0 t}{2\pi}-1\right)^4\right]\sin(\omega_0 t)$$

$E_0 = 0.2$







$E_0 = 0.4$

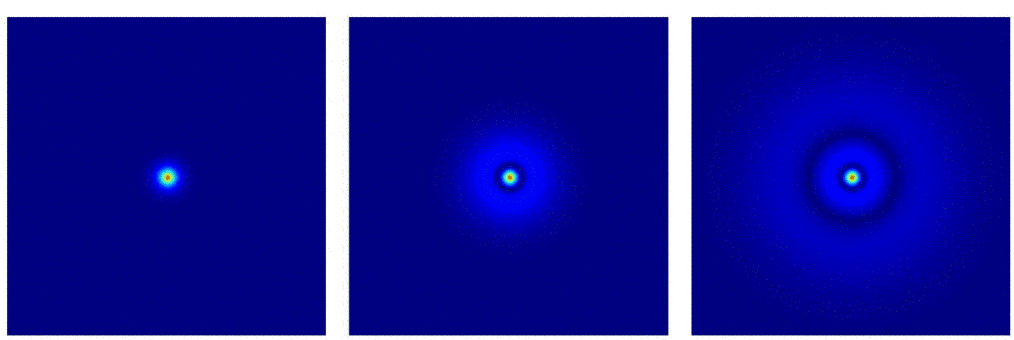
$E_0 = 2$

Electronic excitation in atoms

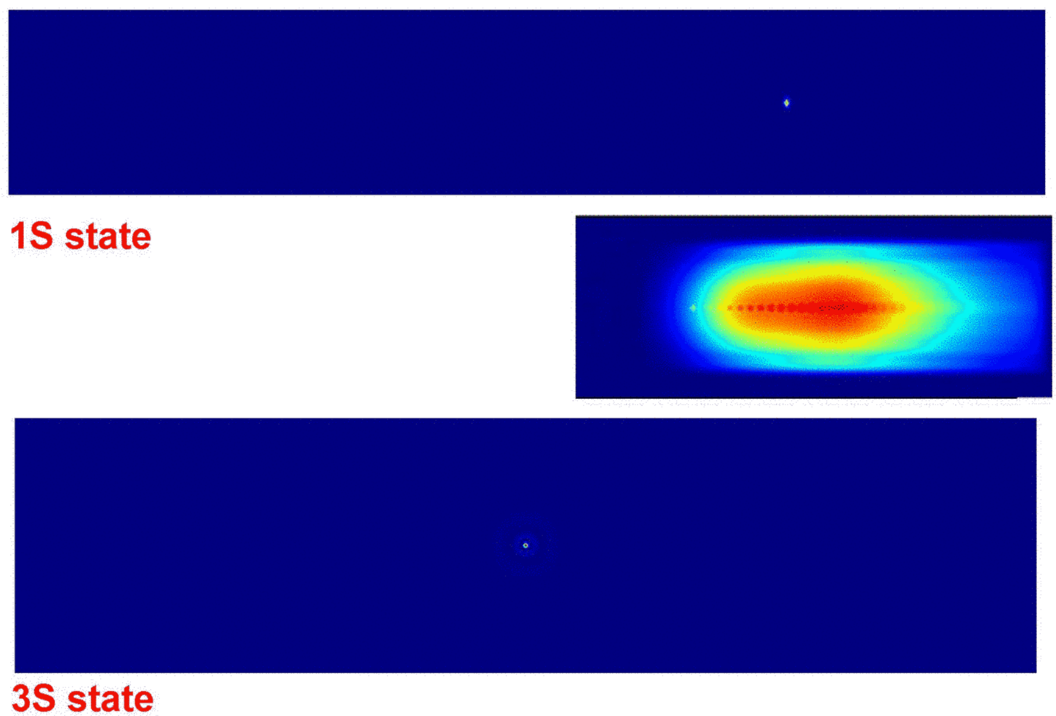
Ionization from initially excited s-states of H

$$\Psi_n(r, t=0) = \frac{1}{n\sqrt{\pi n}} \exp(-r/n) F\left(1-n\,,\,2\,,\,\frac{2}{n}r\right)\;; \qquad P = |\Psi|^2$$

Initial electron wave packet



$n = 1$ $n = 2$ $n = 3$



Three-step strong-field model
for the whole electronic wave packet:

1) Emerges from the atom being almost undisturbed
2) Is accelerated by the strong optical field and spreads
3) Recollides with ion and emits attosecond burst

The profile of attosecond burst can be found analytically for electrons weakly interacting with Coulomb center

M.Yu.Emelin et al., Europhysics Letters, 2005 V.69, N6.
M.Yu. Emelin et al., Laser Physics, 2005 V.15, N6.

Schroedinger equation

$$i\frac{\partial \Psi}{\partial t} = -\frac{1}{2}\frac{\partial^2 \Psi}{\partial r^2} - \frac{1}{r}\frac{\partial \Psi}{\partial r}$$

$$\Psi = \varphi / r$$

$$\frac{\partial \varphi}{\partial t} = \frac{i}{2}\frac{\partial^2 \varphi}{\partial r^2}$$

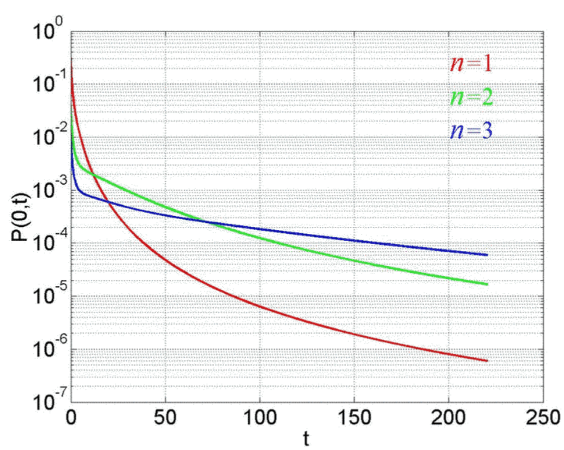
Solution in integral form

$$\varphi(r,t) = \frac{1}{\sqrt{2\pi i t}}\int_0^\infty \varphi(x,0)\left[\exp\left(\frac{i}{2t}(x-r)^2\right) - \exp\left(\frac{i}{2t}(x+r)^2\right)\right]dx$$

$$\Psi_n(r,t) = \frac{1}{r\sqrt{2\pi i t}}\int_0^\infty x\Psi_n(x,0)\left[\exp\left(\frac{i}{2t}(x-r)^2\right) - \exp\left(\frac{i}{2t}(x+r)^2\right)\right]dx$$

Electron probability density at the center of the packet

$$\Psi_n(0,t) = A\sum_{m=0}^{n-1} C_m\left[B_1 F\left(-n, m-n+\tfrac{3}{2}, z\right) + B_2(-z)^{n-m-\frac{1}{2}} F\left(-m-\tfrac{1}{2}, n-m+\tfrac{1}{2}, z\right)\right]$$



$$A = -\frac{\exp(-z)}{\sqrt{\pi^3 n^3}}$$

$$B_1 = \frac{\Gamma\left(m+\frac{3}{2}\right)\Gamma\left(n-m-\frac{1}{2}\right)}{n!}$$

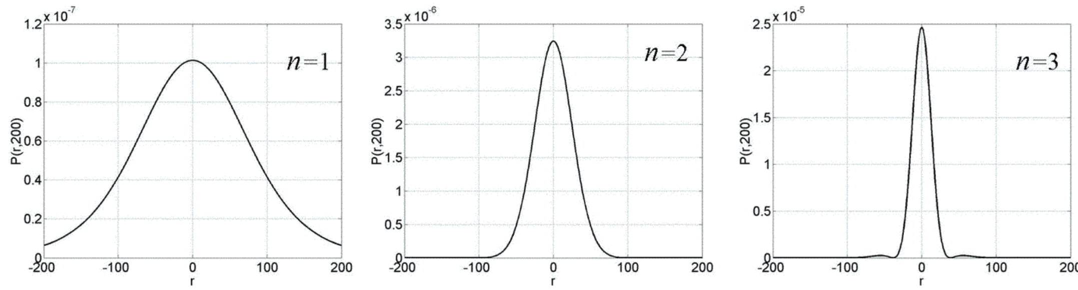
$$B_2 = \Gamma\left(m-n+\tfrac{1}{2}\right)$$

$$C_m = (-1)^{n-m} C_{2n}^{2m+1}$$

$$z = -\frac{it}{2n^2}$$

Asymptotic shape of the packet at t → ∞

$$\left|\Psi_n(r, t\to\infty)\right|^2 = \frac{2n^3 t^5}{\pi^2 \left(t^2 + n^2 r^2\right)^{2n+2}} \left[\sum_{m=0}^{n-1} C_{2n}^{2m+1} (-1)^m \left(t^2\right)^{n-m-1} \left(n^2 r^2\right)^m \right]^2$$



Analytical solution for attosecond atomic response

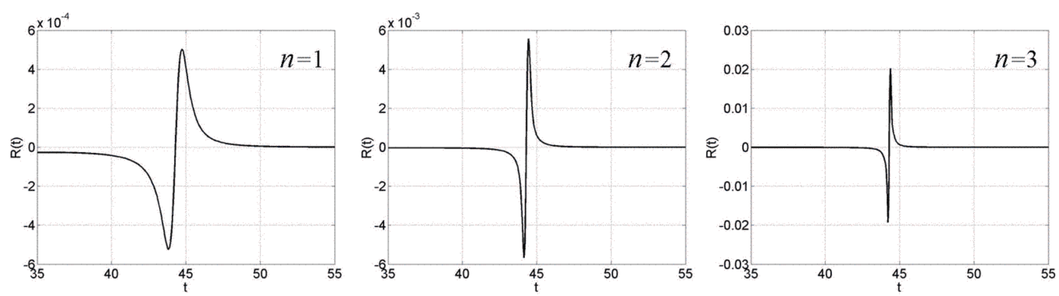
$$d_n''(t) = E(t) - R(t) \quad , \qquad R(t) = \iiint \left|\Psi(\vec{r}, t)\right|^2 \frac{\partial V}{\partial z} dx dy dz$$

$$R_n(t) = -\frac{2}{\pi z^2}\left[arctg(\lambda) + \frac{\lambda K_n(\lambda)}{n\left(4n^2-1\right)\left(1+\lambda^2\right)^{2n+1}} \right] \qquad \lambda = \frac{nz(t)}{t}$$

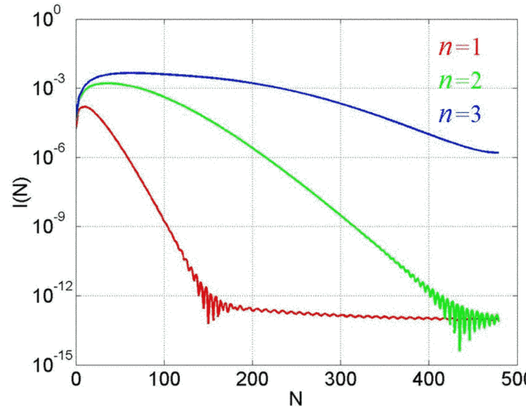
$$K_1(\lambda) = 3\lambda^4 + 8\lambda^2 - 3$$

$$K_2(\lambda) = 30\lambda^8 + 140\lambda^6 - 128\lambda^4 + 500\lambda^2 - 30$$

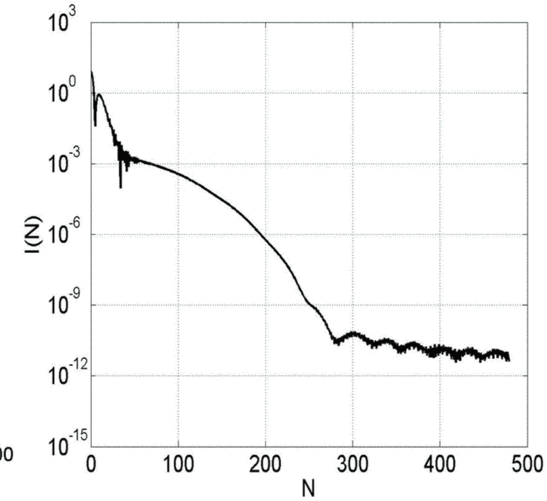
$$K_3(\lambda) = 105\lambda^{12} + 700\lambda^{10} - 1043\lambda^8 + 13584\lambda^6 - 11053\lambda^4 + 4340\lambda^2 - 105$$



Spectra of atomic nonlinear response

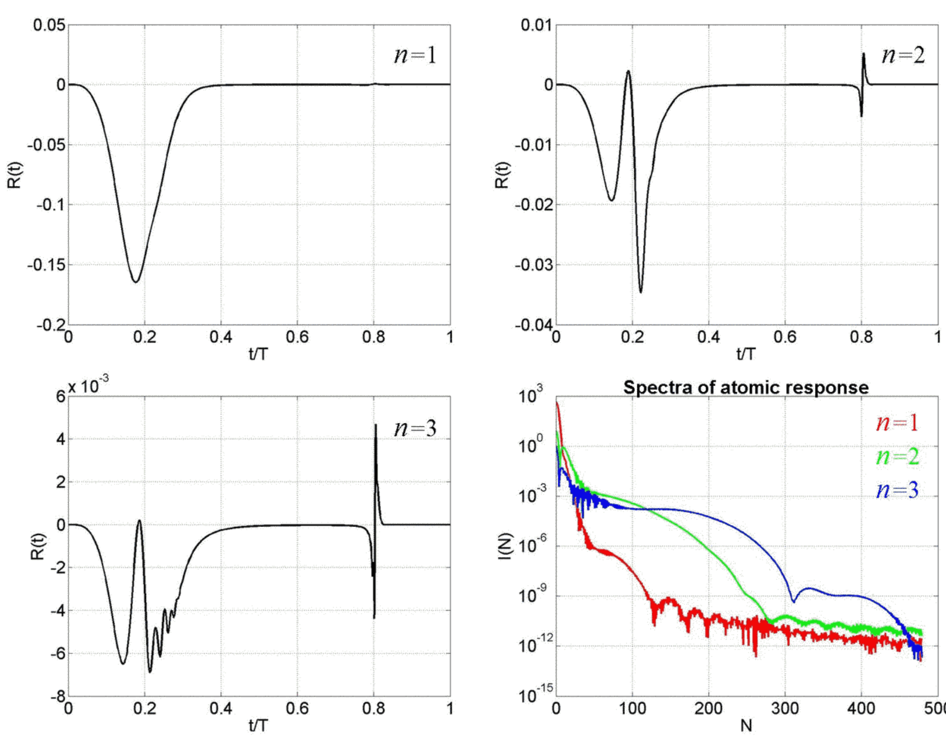


Analytical result

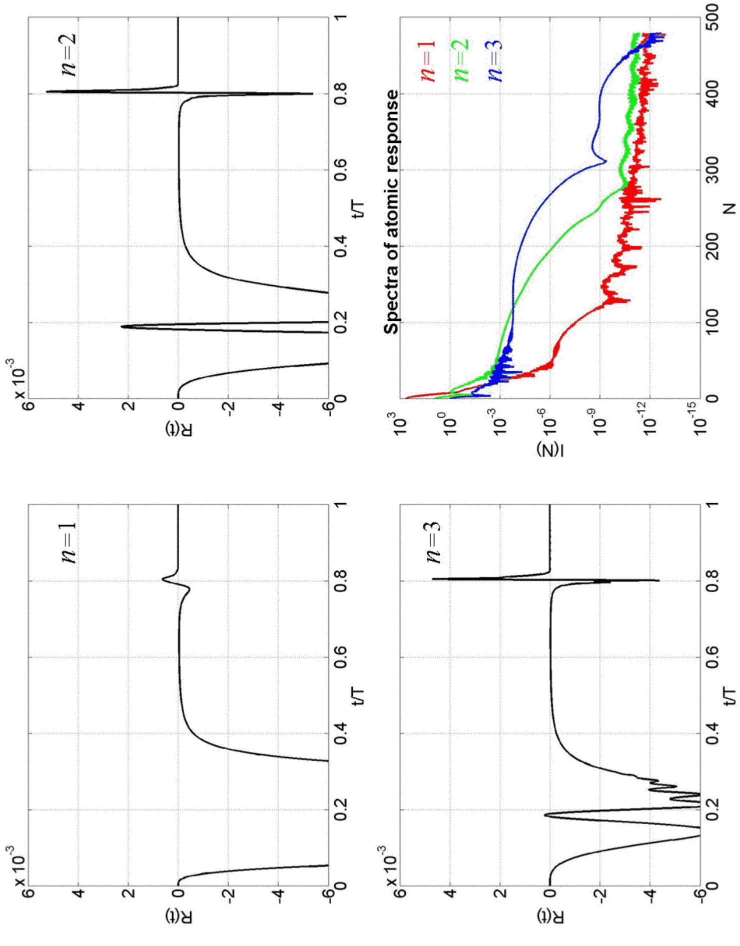


Numerical result (n=2)

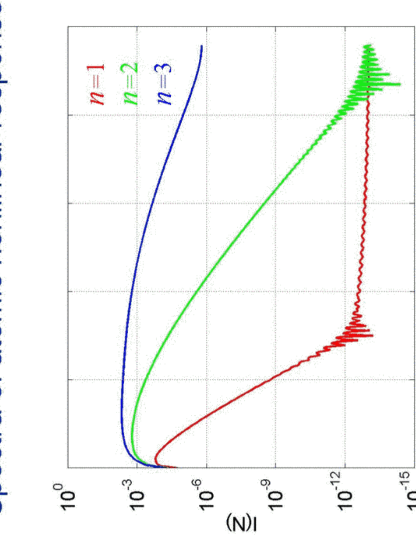
Atomic nonlinear response

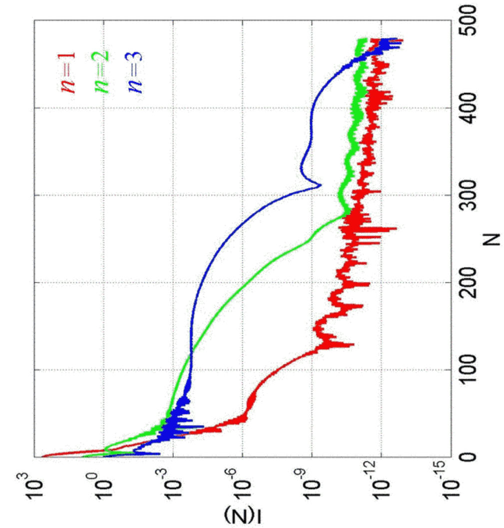


Atomic nonlinear response



Spectra of atomic nonlinear response





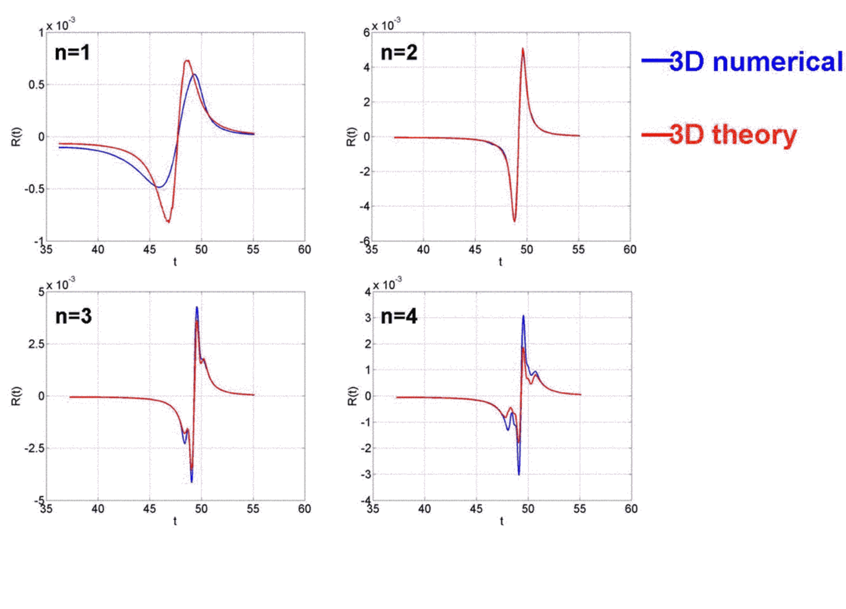
$W = \int R(t)^2 dt$ $\quad W_2/W_1 \approx 35$ $\quad I_2(100)/I_1(100) \approx 10^4$

How short an attosecond pulse can be?

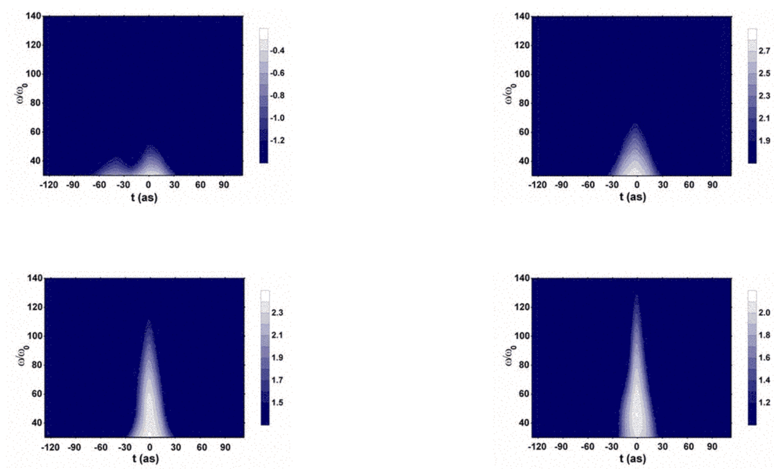
Besides the increased efficiency, the attosecond pulse is shorter for excited states. What is the shortest pulse that can be produced by this mechanism?

Optimal excitation state n ≈ 4-5
Maximum collision velocity V ≈ 25-30 (v/c ≈ 1/5)

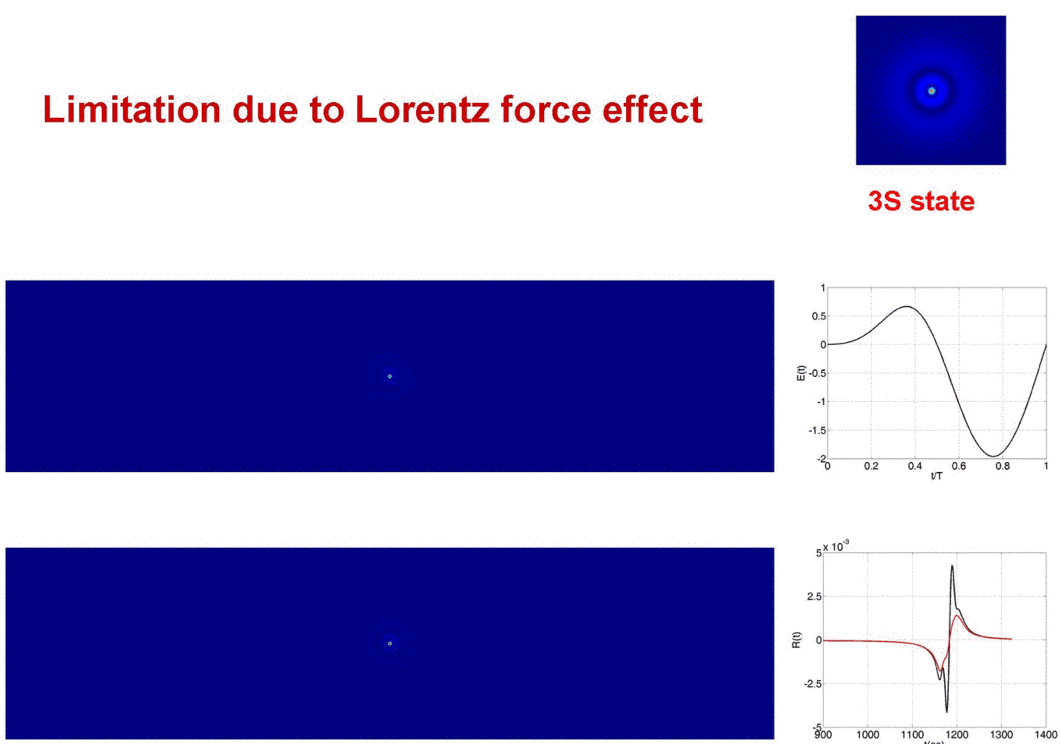
$t_{min} \approx 10$ as



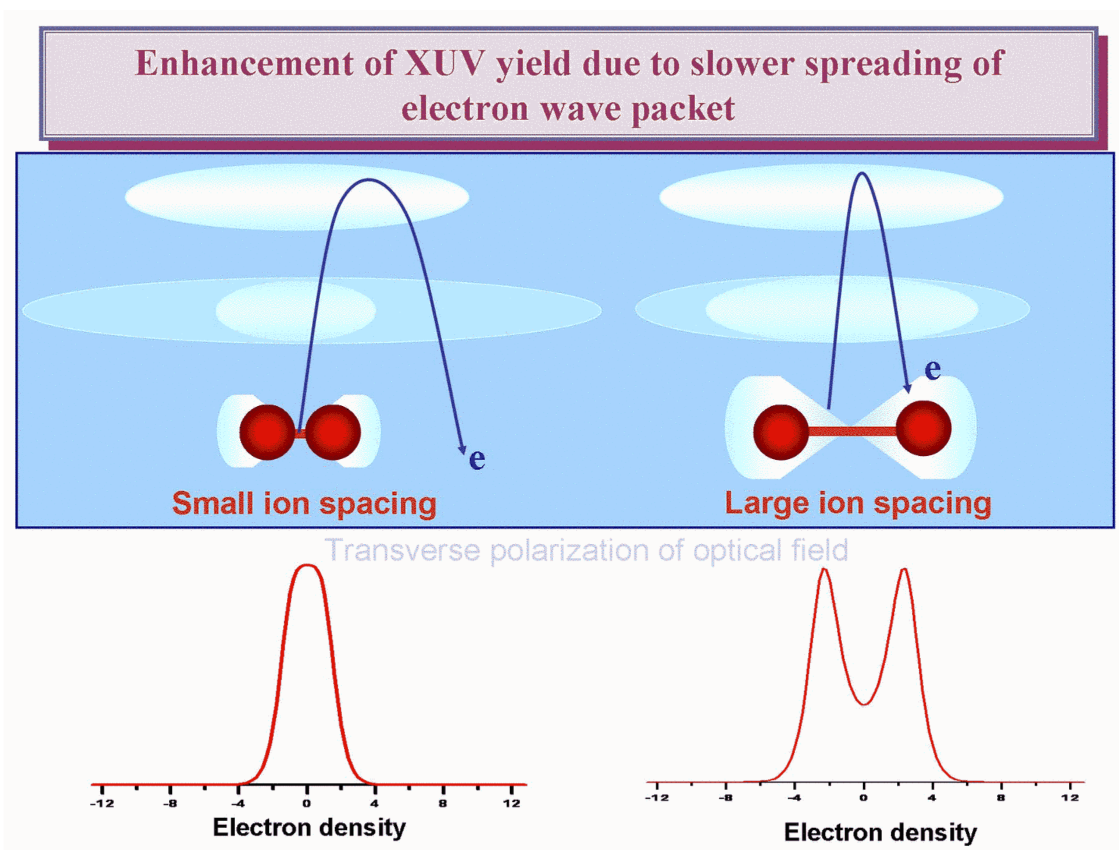
Time-frequency spectra of attosecond bursts for n=1, 2, 3, 4 electronic states



Limitation due to Lorentz force effect

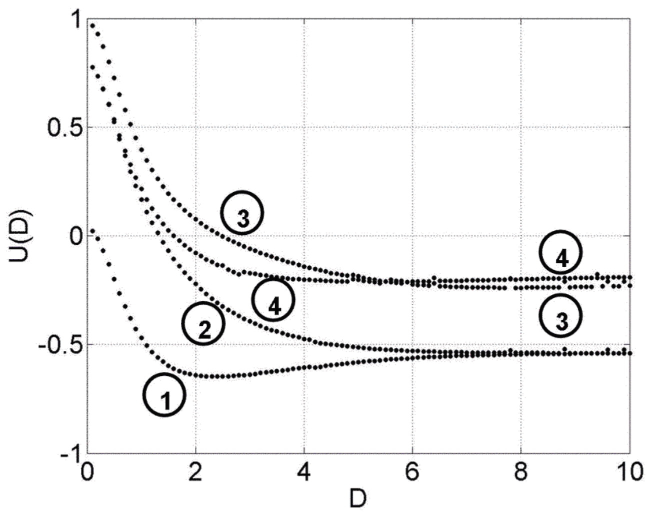


Vibrational excitation in molecules

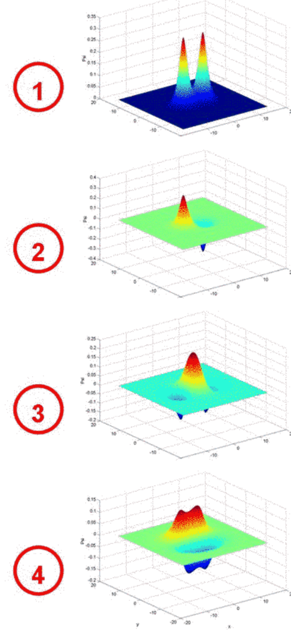


Attosecond burst production in high intensity regime (H_2^+, ionization from excited states)

Potential curves



Eigenfunctions



D=7.5

HHG in stretched molecules

P. Moreno, L. Plaja, and L. Roso, *PRA* **55**, R1593 (1997).

A. Bandrauk and H. Yu, *PRA* **59**, 539 (1999).

M.Yu. Emelin, M.Yu. Ryabikin, A.M. Sergeev, M.D. Chernobrovtseva, T. Pfeifer, D. Walter, and G. Gerber, *JETP Letters* **77**, 212 (2003).

Pump-probe scheme for HHG in molecules

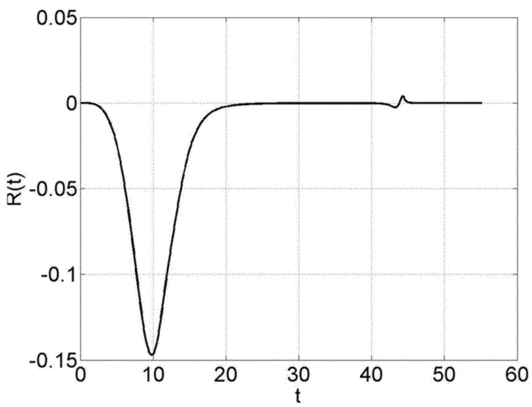
R. Numico, P. Moreno, L. Plaja, and L. Roso, *J. Phys. B* **31**, 4163 (1998).

N. Hay, R. Velotta, M. B. Mason, M. Castillejo, and J.P. Marangos, *J. Phys. B* **35**, 1051 (2002).

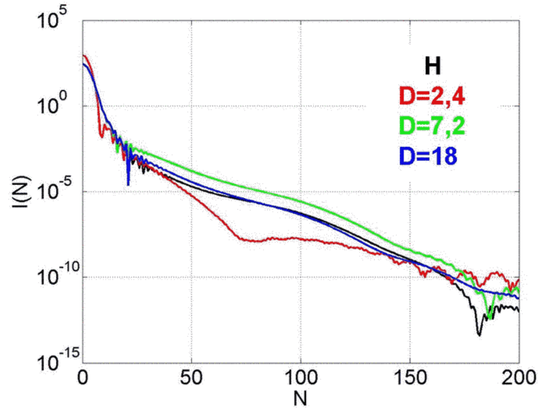
T. Pfeifer, D. Walter, G. Gerber, M.Yu. Emelin, M.Yu. Ryabikin, M.D. Chernobrovtseva, and A.M. Sergeev, *PRA* **70** (2004)

Enhanced attosecond burst production in a stretched molecule

Molecular nonlinear response



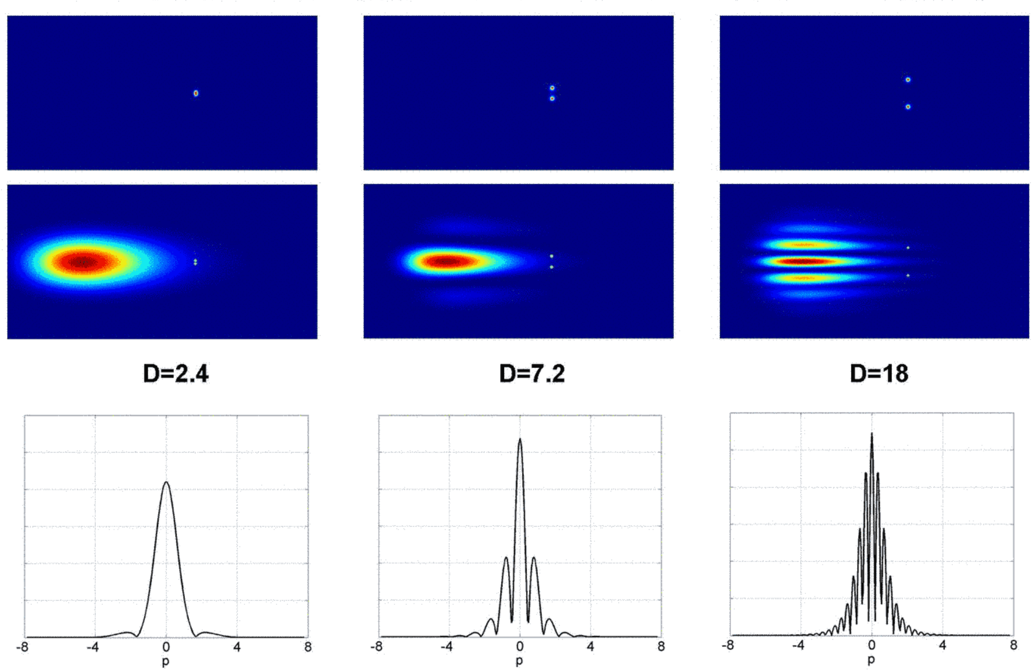
Spectra of molecular response



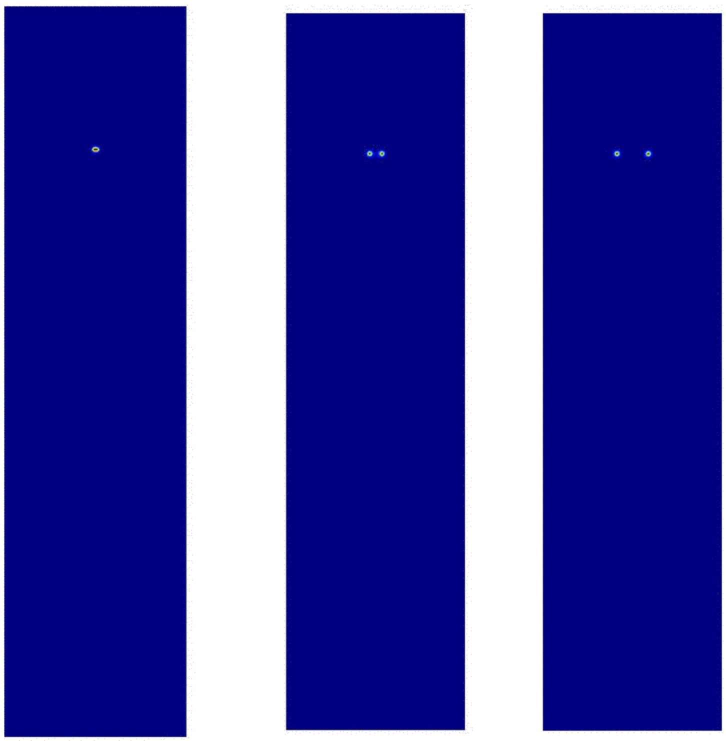
$$R(t) = \iint |\Psi(\vec{r},t)|^2 \frac{\partial V}{\partial y} dxdy$$

M.Yu. Emelin, M.Yu. Ryabikin, A.M. Sergeev, M.D. Chernobrovtseva, T. Pfeifer, D. Walter, and G. Gerber, *JETP Letters* **77**, 212 (2003).

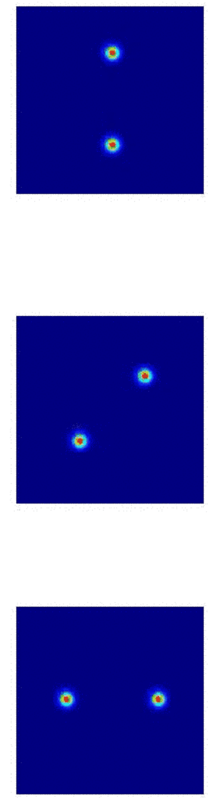
Wave packets: initial (up) and before recollision with ion core (down)



Initial electron transverse momentum distribution



3. Orientation of molecule against laser field



Alignment dependence of HHG in molecules

Theory

D.G. Lappas and J.P. Marangos, *J. Phys. B.* **33**, 4679 (2000).

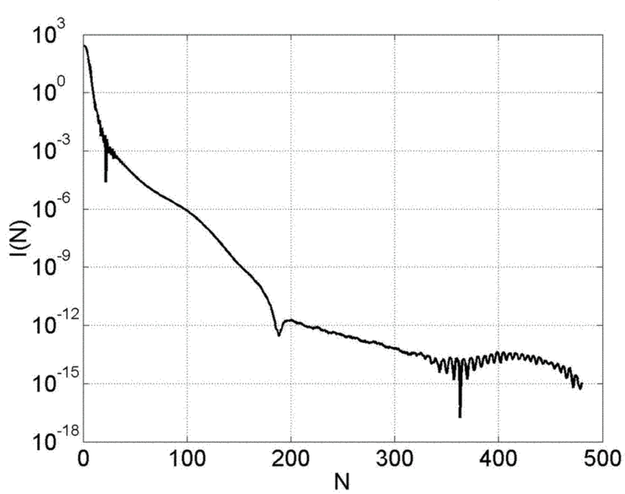
M. Lein, N. Hay, R. Velotta, J.P. Marangos, and P.L. Knight, *PRL* **88**, 183903 (2002); *PRA* **66**, 023805 (2002).

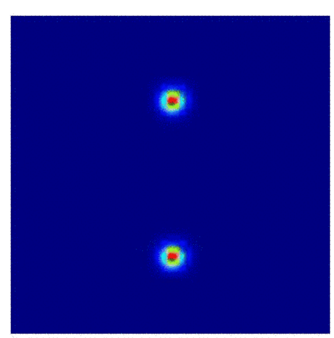
Experiment

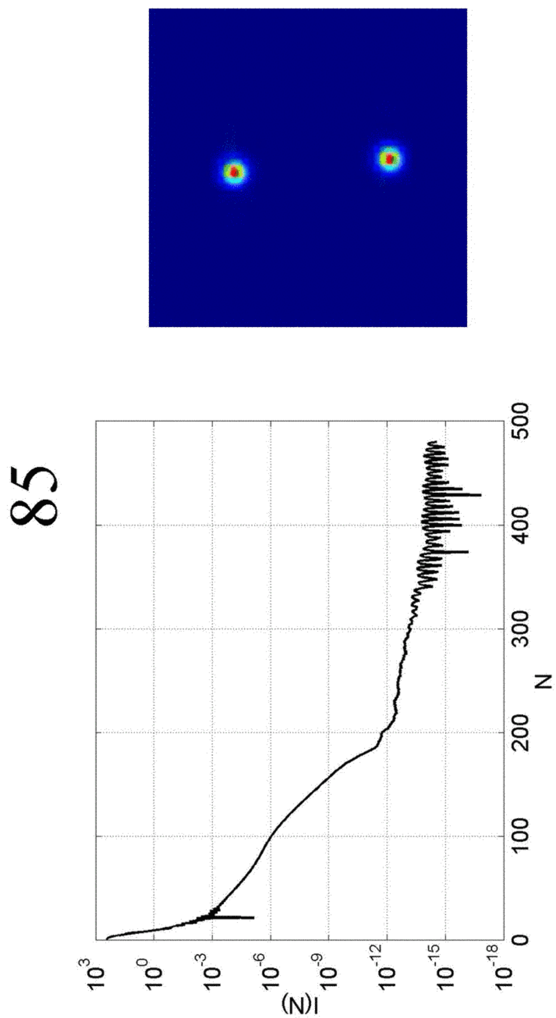
R. Velotta, N. Hay, M.B. Mason, M. Castillejo, and J.P. Marangos, *PRL* **87**, 183901 (2002); N. Hay, R. Velotta, M. Lein, R. de Nalda, E. Heesel, M. Castillejo, and J.P. Marangos, *PRA* **65**, 053805 (2002).

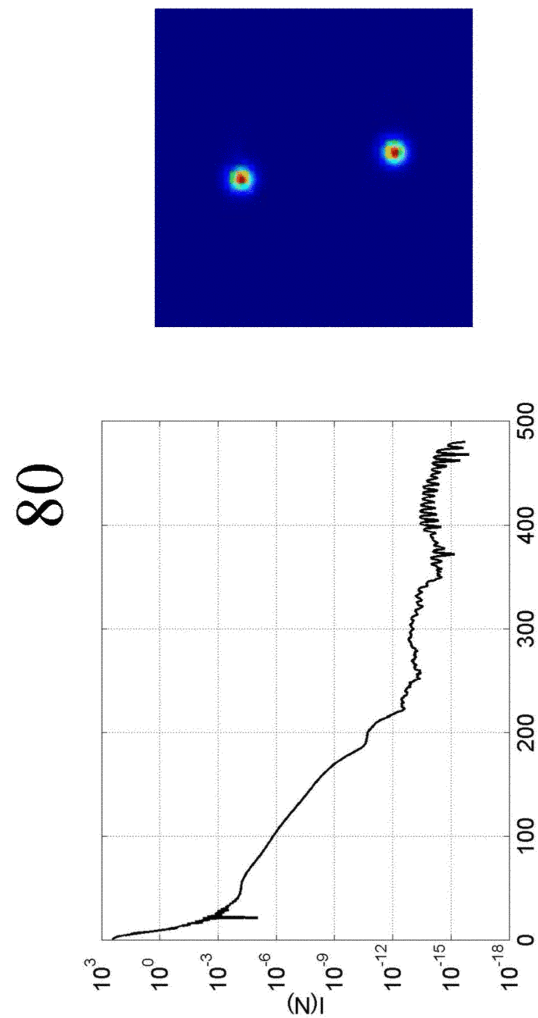
D. Zeidler, J. Levesque, J. Itatani, K. Lee, P. Dooley, I. Litvinyuk, D.M. Villeneuve, and P.B. Corkum, in: *Springer Series in Optical Sciences.* V. 95. *Ultrafast Optics IV* **(**Ed. by F. Krausz, G. Korn, P. Corkum, and I.A. Walmsley). Springer-Verlag, 2004.

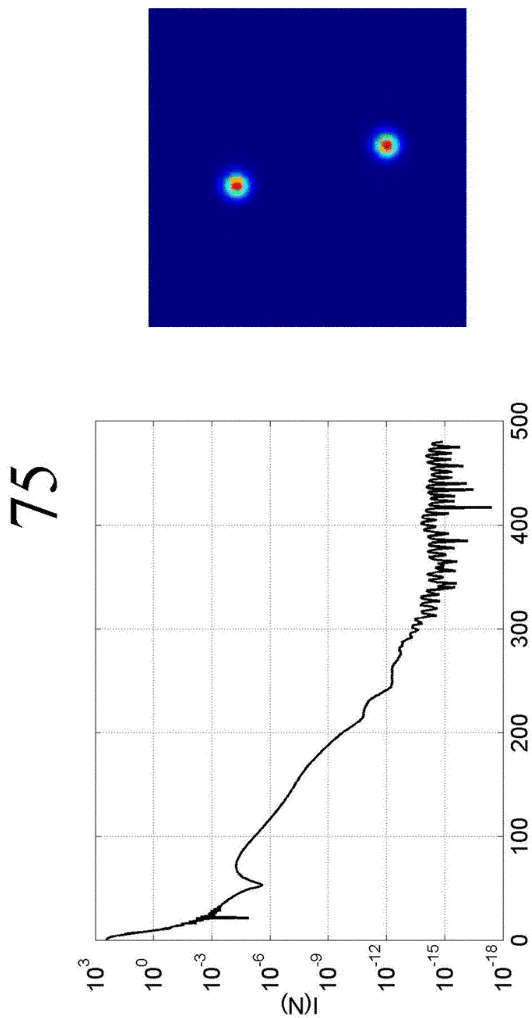
90

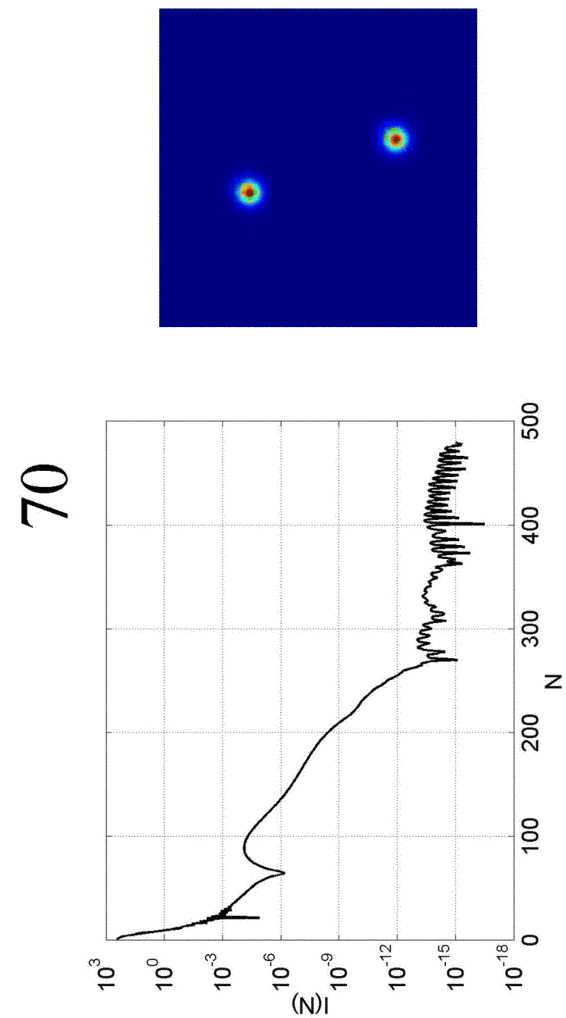


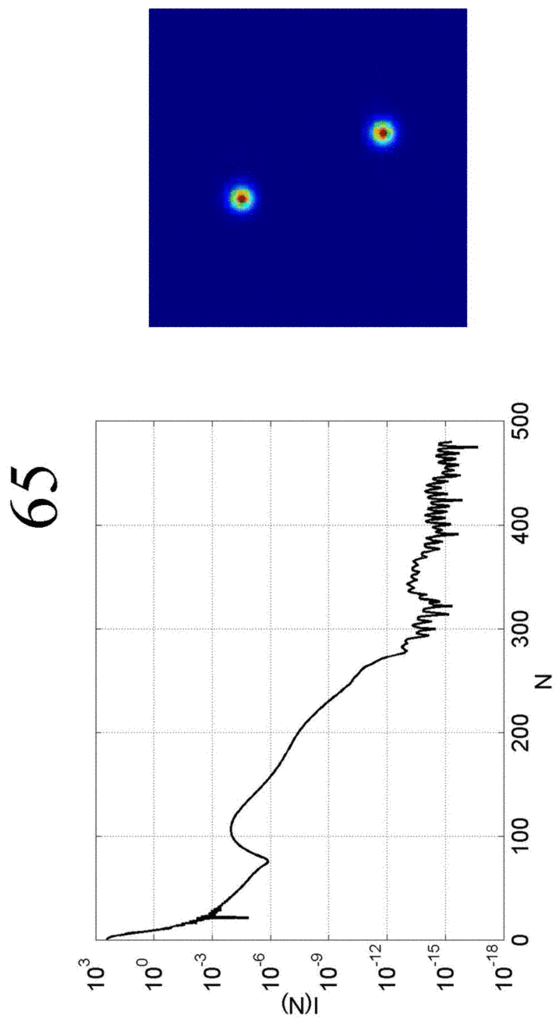


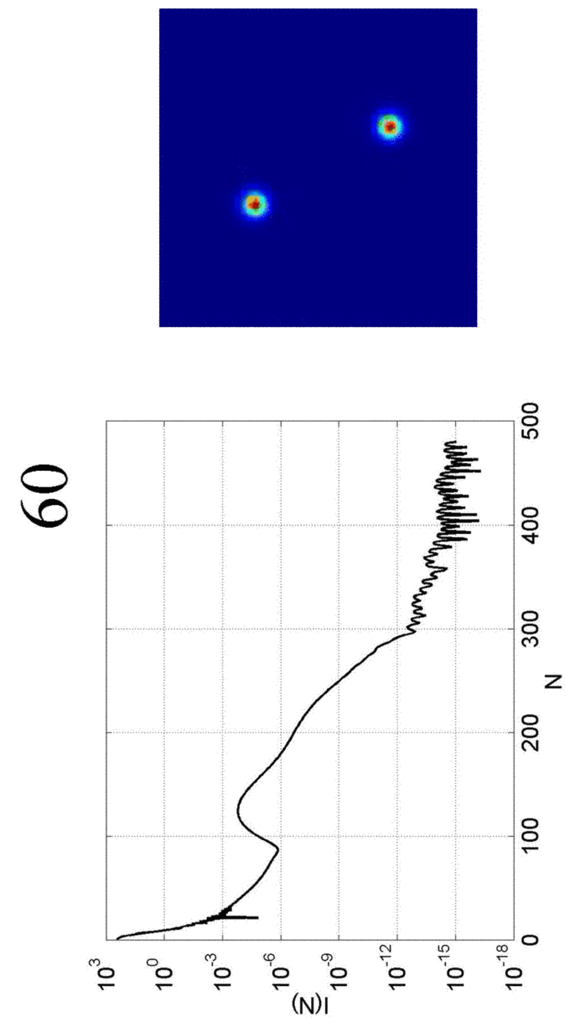

85
I(N)
N

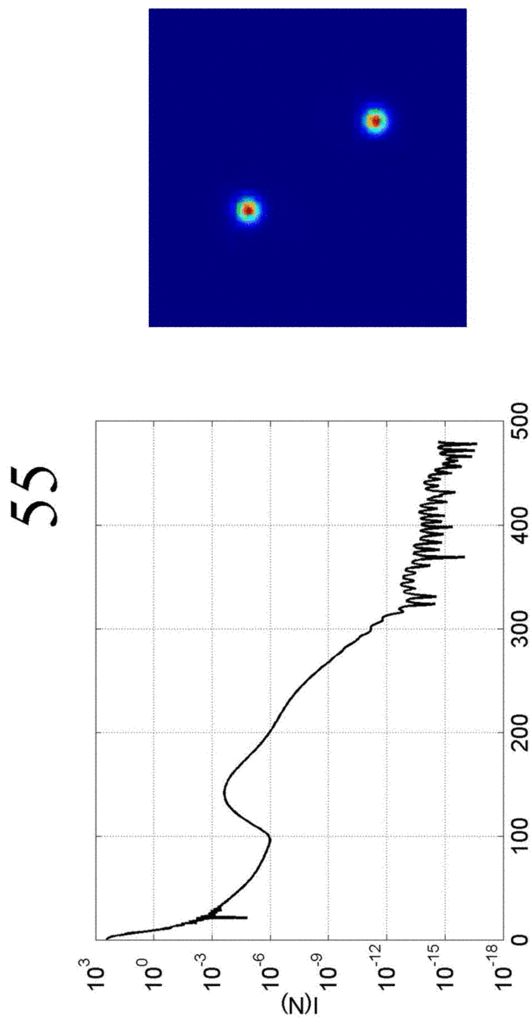

80
I(N)
N

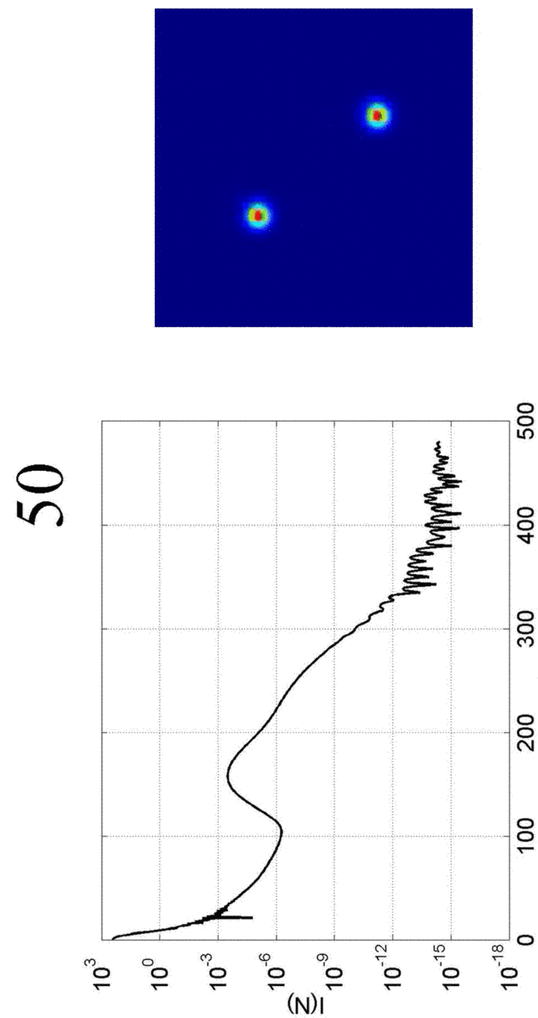

75
I(N)
N
10^3
10^0
10^-3
10^-6
10^-9
10^-12
10^-15
10^-18
0
100
200
300
400
500

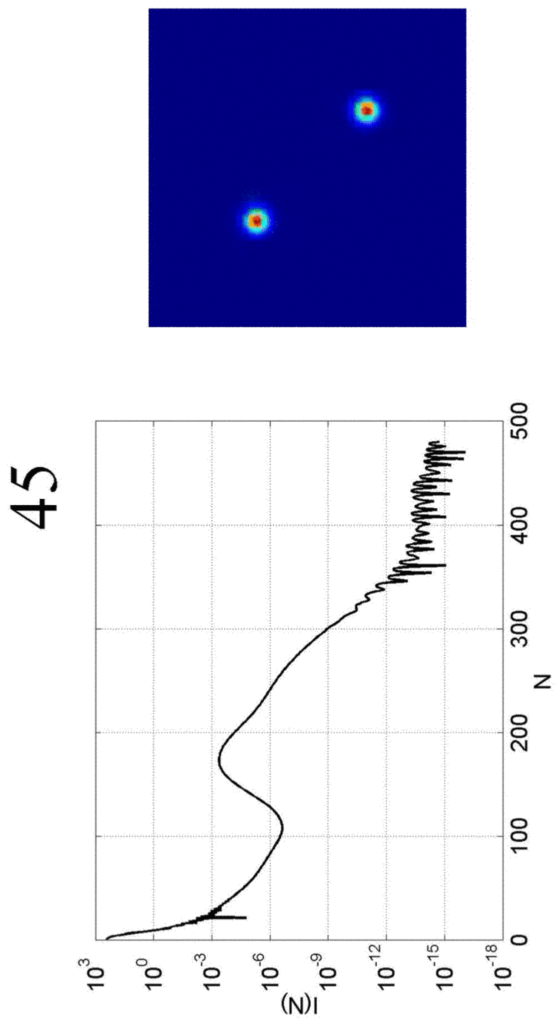

70
I(N)
N
10^3
10^0
10^-3
10^-6
10^-9
10^-12
10^-15
10^-18
0
100
200
300
400
500

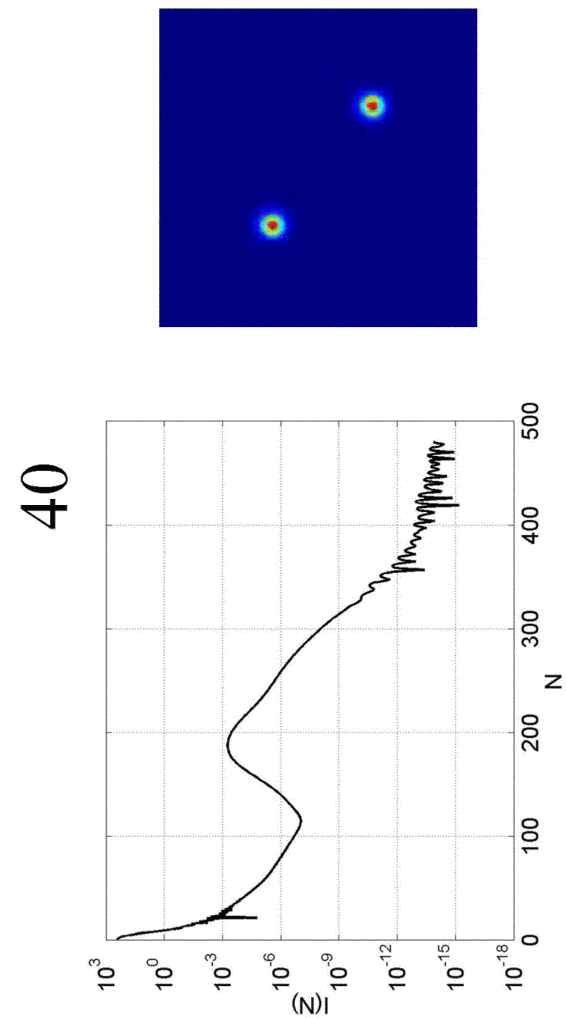

65
I(N)
N

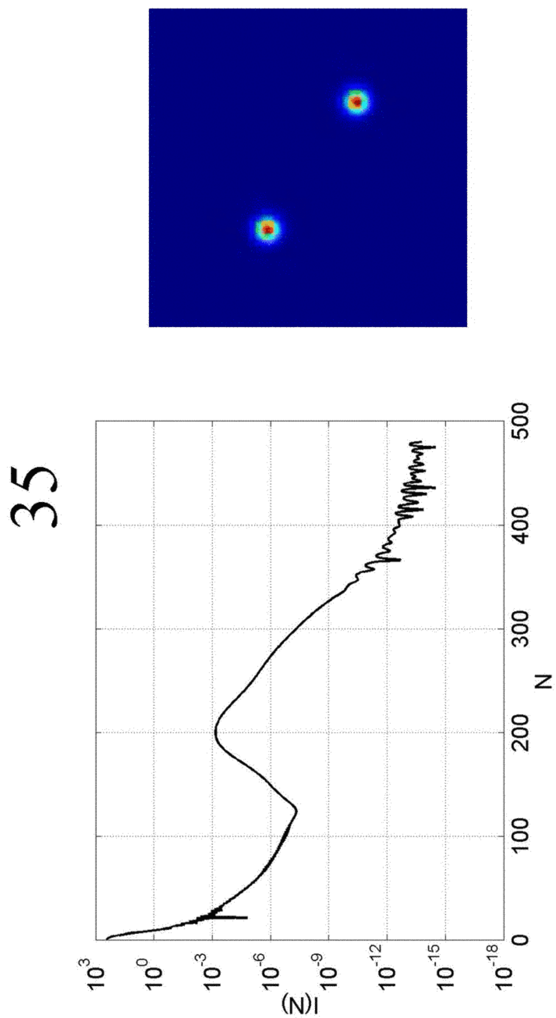

60
I(N)
N

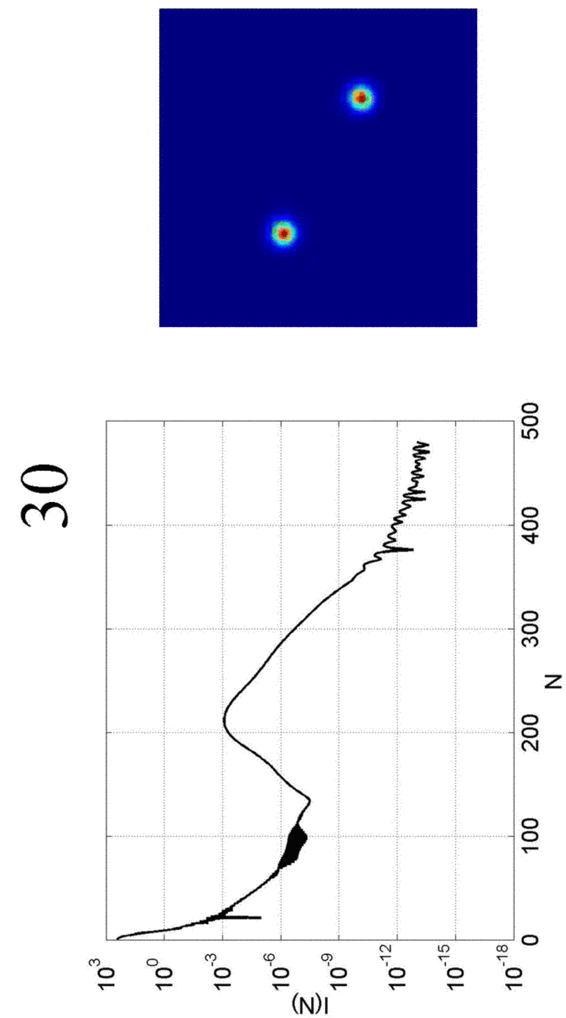

55
I(N)
N

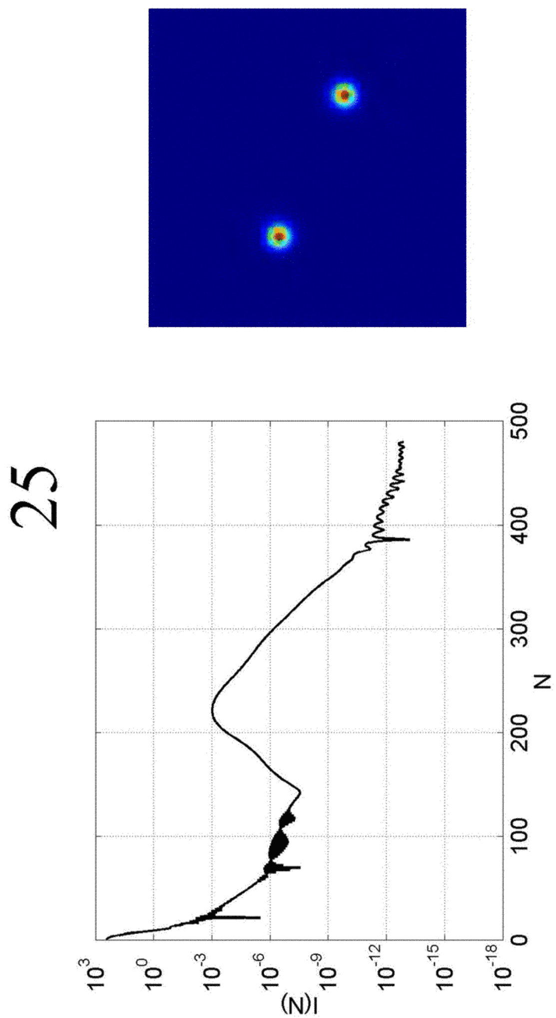

50
I(N)
N

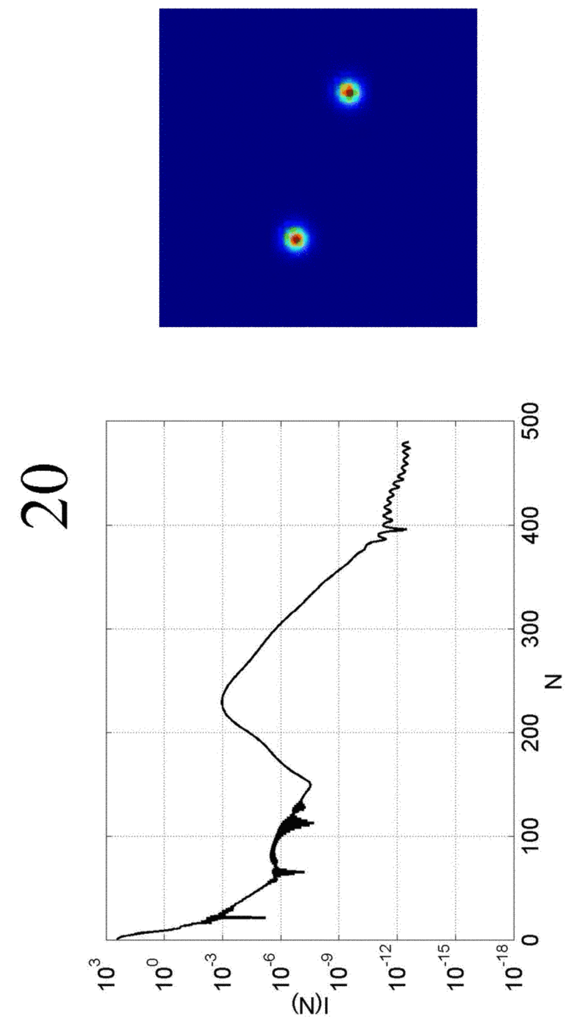

45
I(N)
N
0
100
200
300
400
500

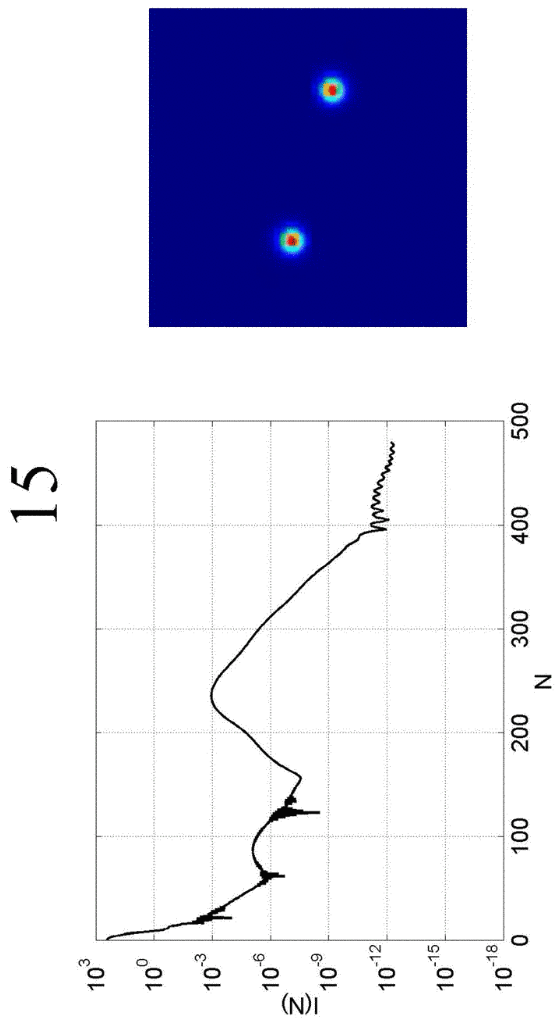

40
I(N)
N
0
100
200
300
400
500

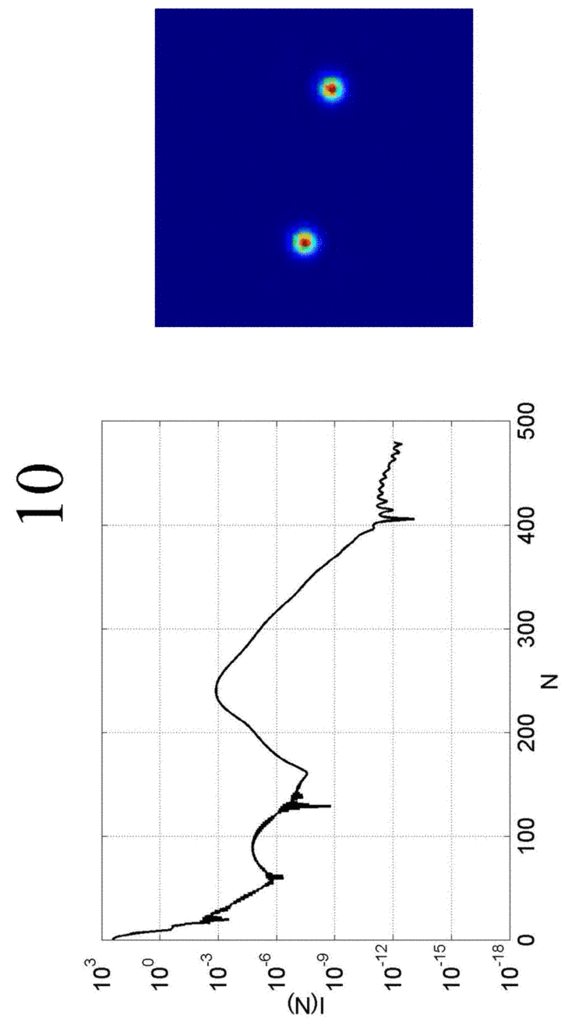

35
N
I(N)
0
100
200
300
400
500

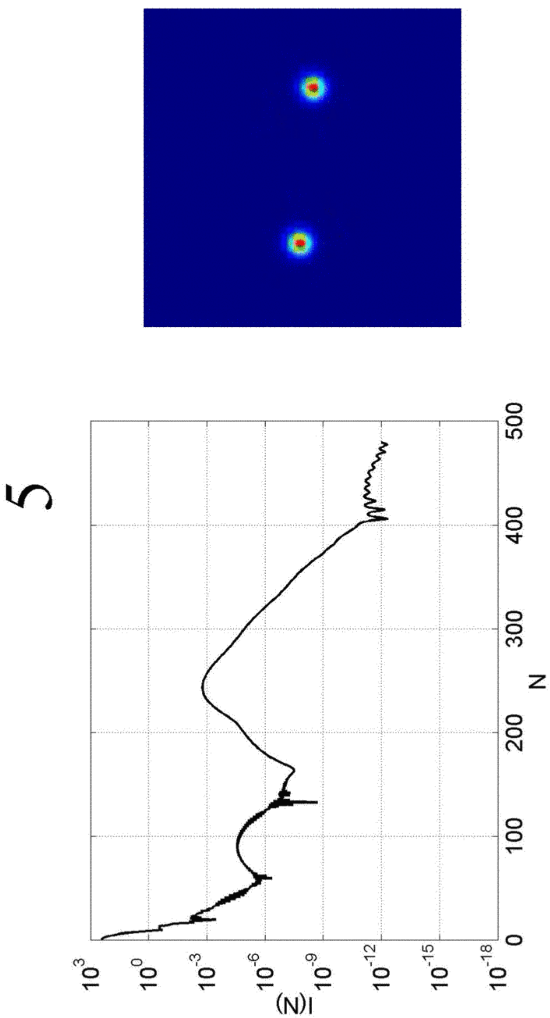

30
N
I(N)
0
100
200
300
400
500

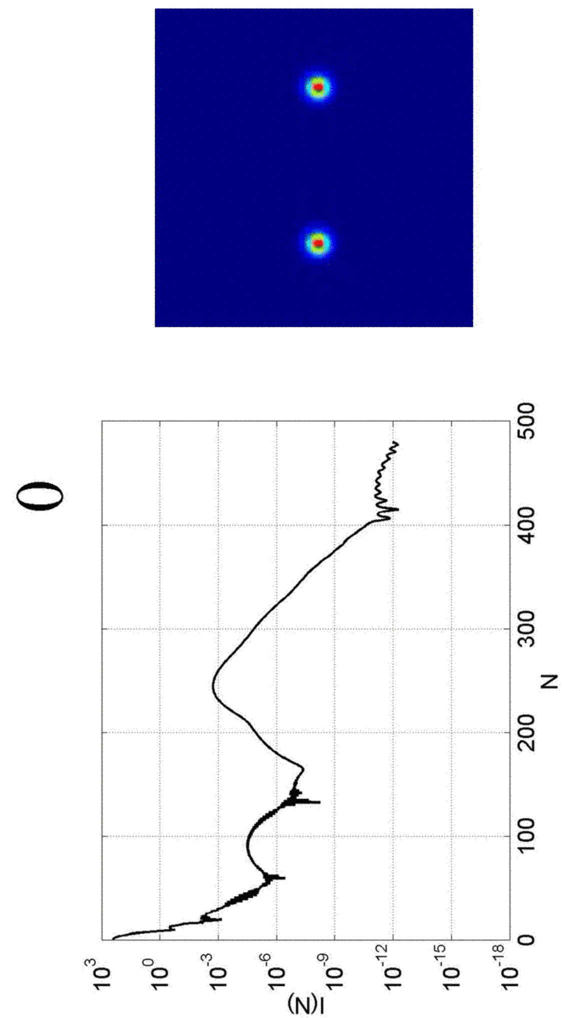

25
I(N)
N

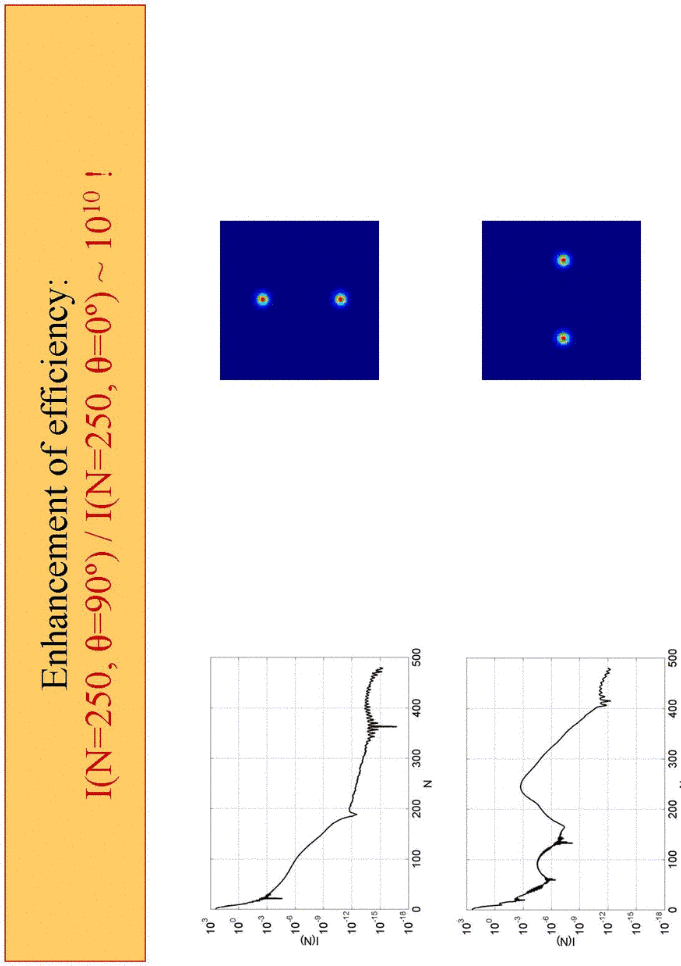

20
I(N)
N

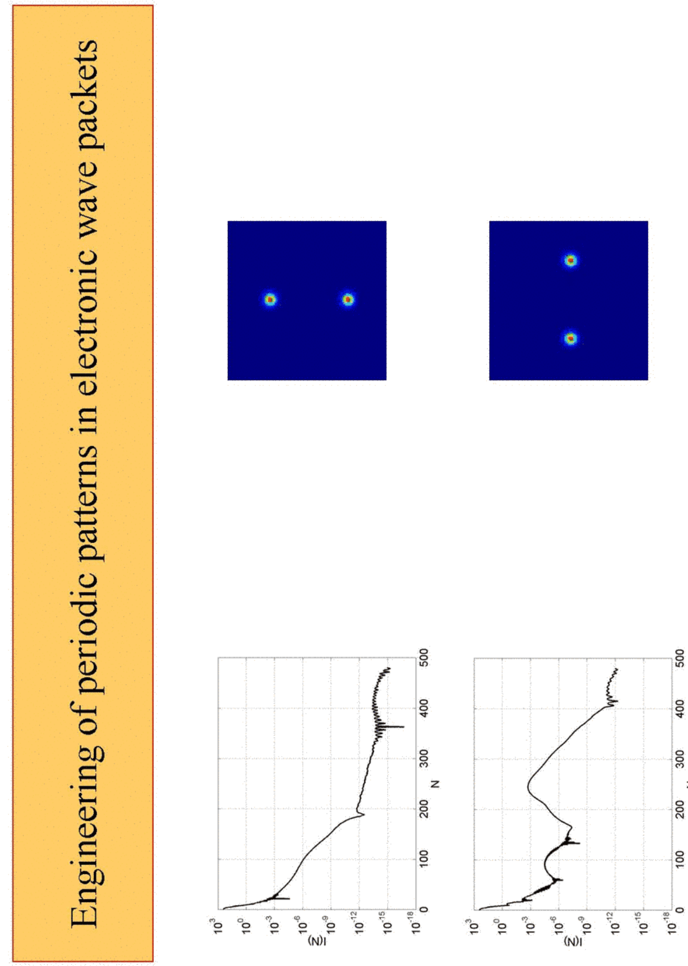

15
N
I(N)

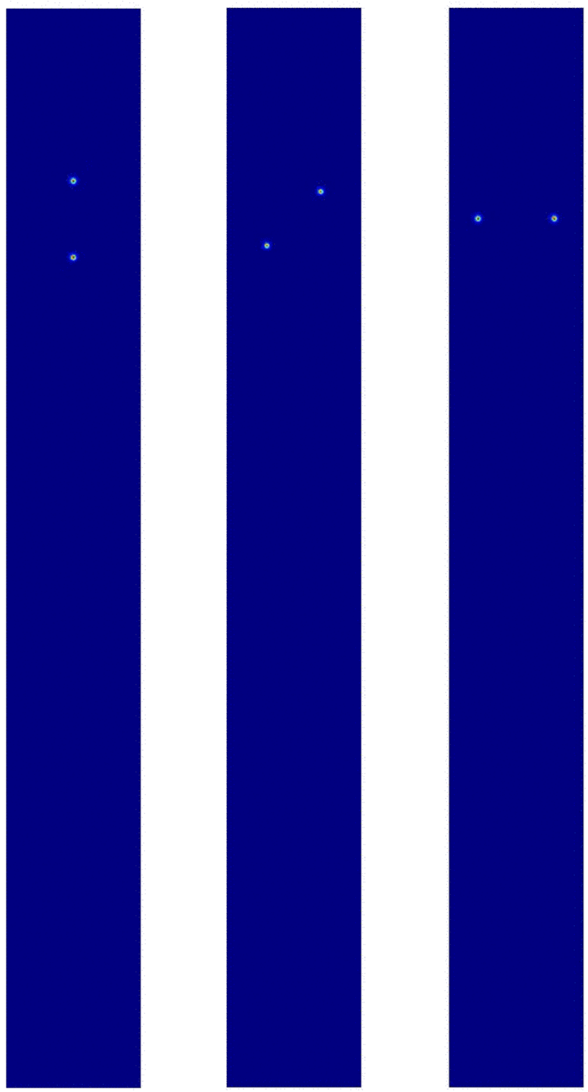

10
N
I(N)

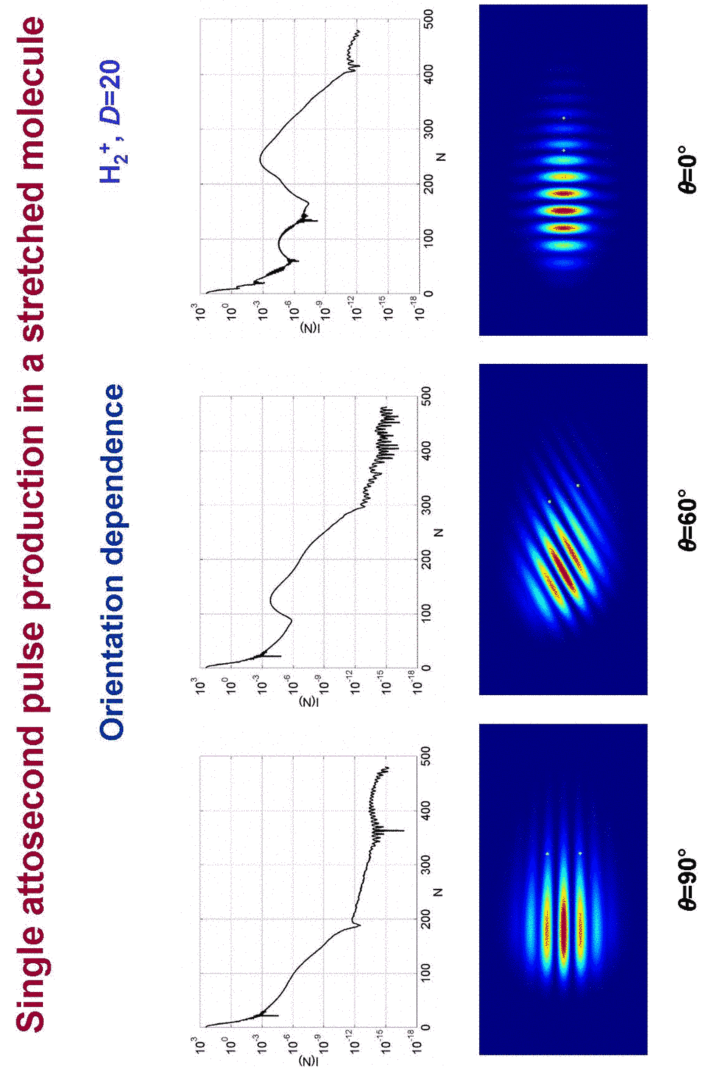

5
I(N)
N
0
100
200
300
400
500


0
I(N)
N
0
100
200
300
400
500


Enhancement of efficiency:
I(N=250, θ=90°) / I(N=250, θ=0°) ~ 10^10 !
I(N)
N

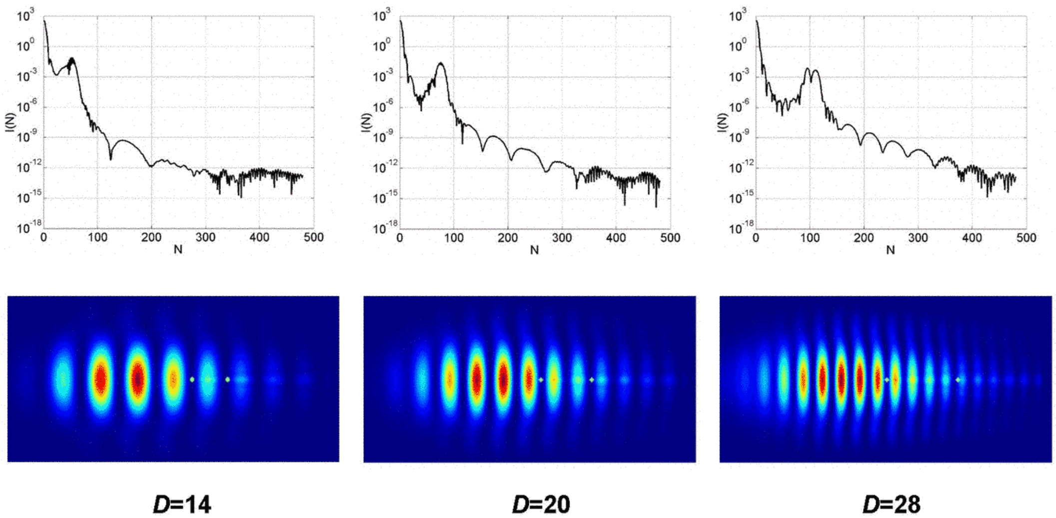

Engineering of periodic patterns in electronic wave packets
I(N)
N




Single attosecond pulse production in a stretched molecule
Orientation dependence
H_2^+, D=20
θ=0°
θ=60°
θ=90°
I(N)
N

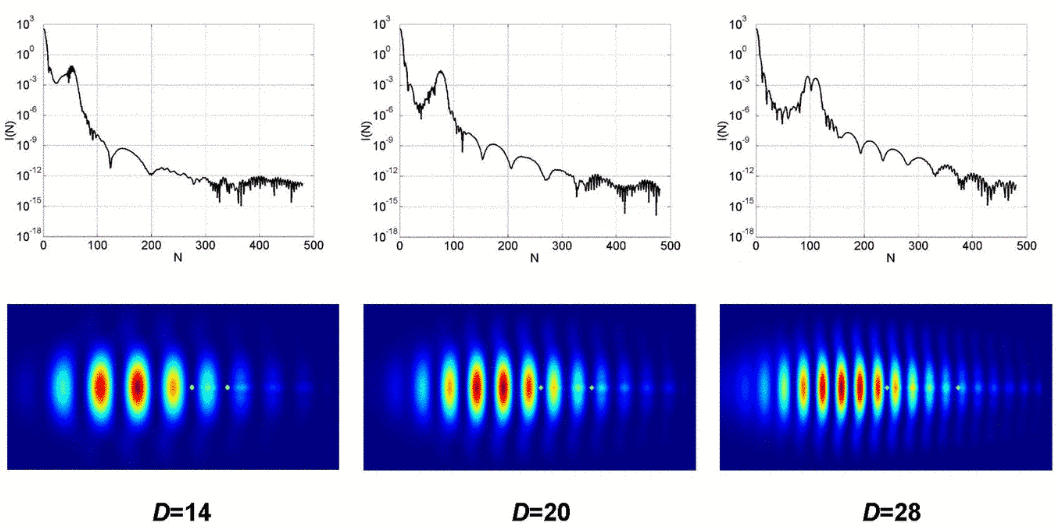
Single attosecond pulse production in a stretched molecule

Bond-length dependence H_2^+, θ=0°

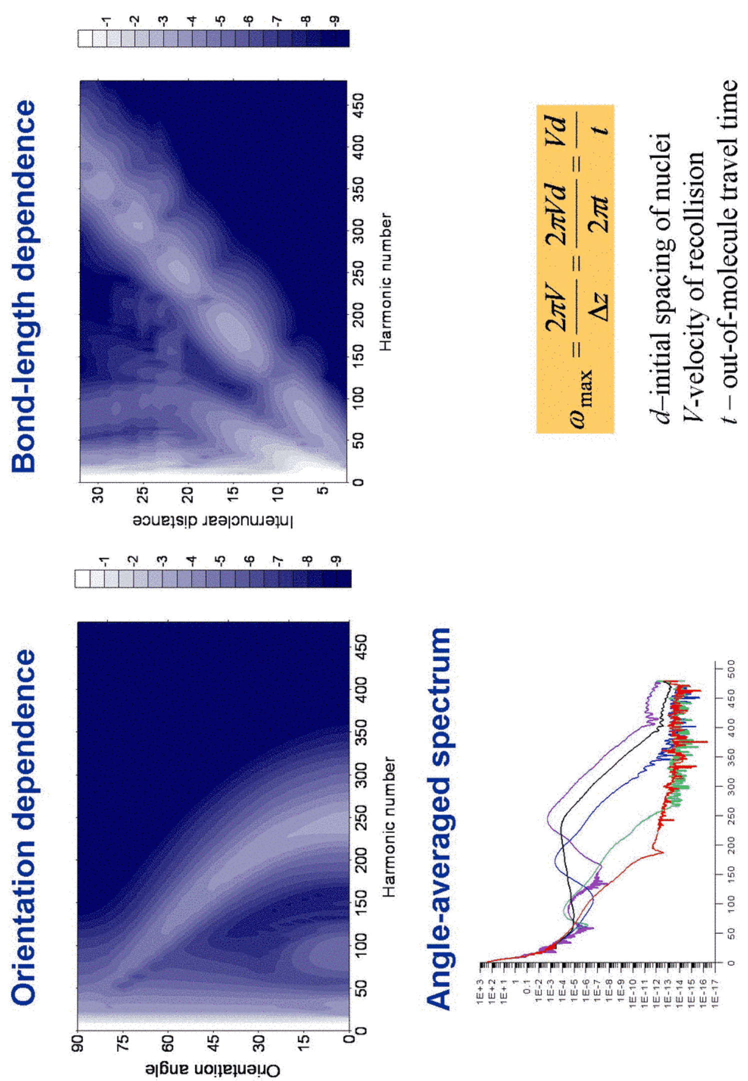


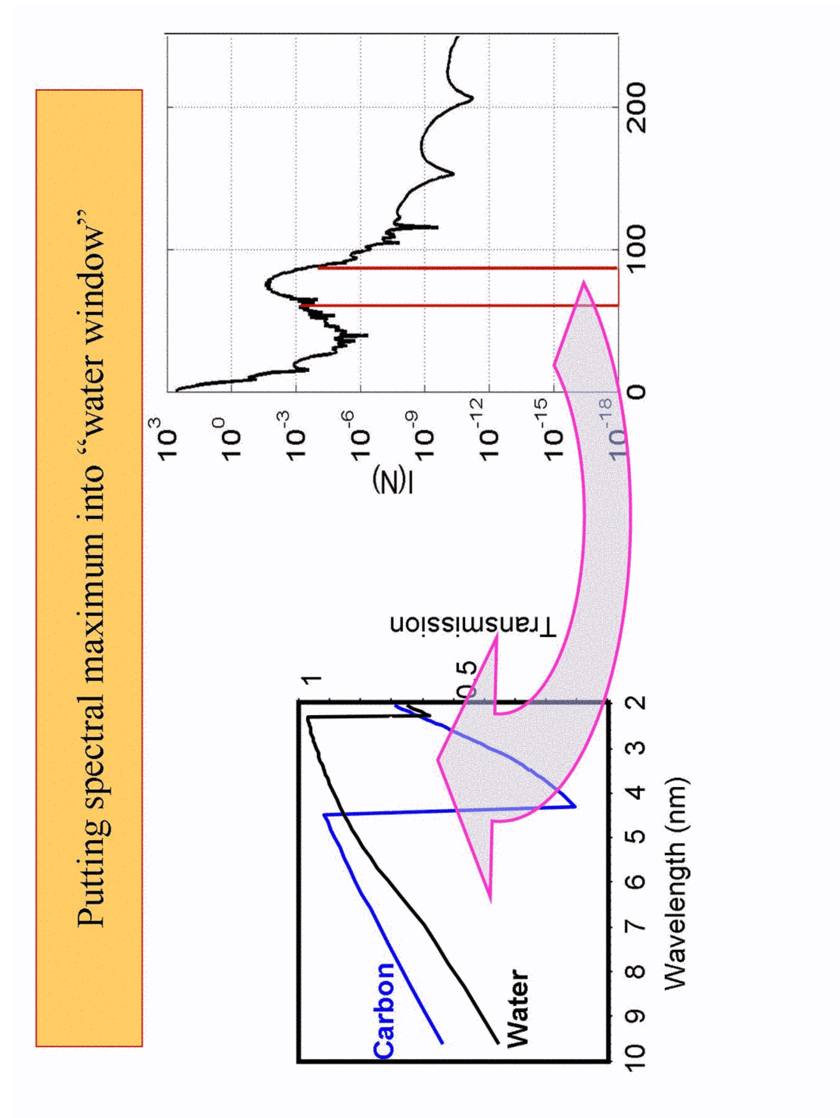
Single attosecond pulse production in a stretched molecule

Bond-length dependence H_2^+, θ=0°



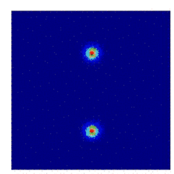
By recording attosecond pulse spectra, one can image molecule structure!


Orientation dependence
Orientation angle
Harmonic number
Bond-length dependence
Internuclear distance
Harmonic number
Angle-averaged spectrum
$\omega_{\max} = \frac{2\pi V}{\Delta z} = \frac{2\pi V d}{2\pi t} = \frac{V d}{t}$
d–initial spacing of nuclei
V-velocity of recollision
t – out-of-molecule travel time

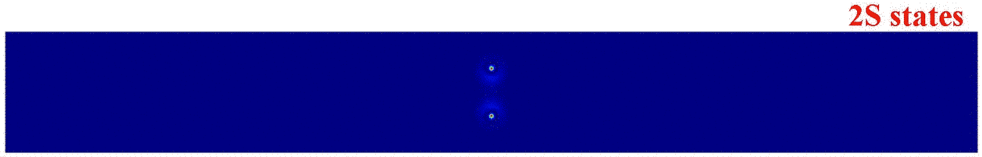

Putting spectral maximum into "water window"
|(N)|
Transmission
Carbon
Water
Wavelength (nm)

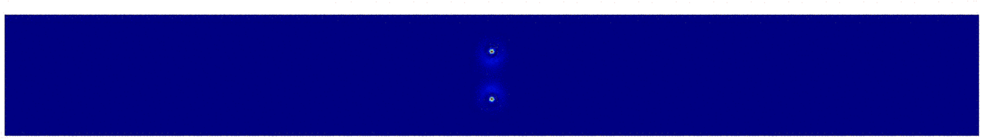
Orientation of molecule against direction of pulse propagation

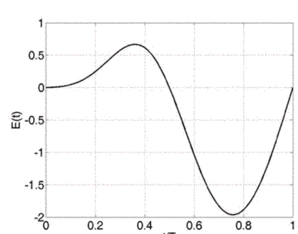
Optimization of electronic, vibronic excitations and Lorentz force effect

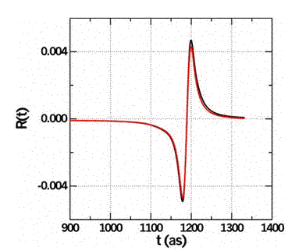


H2+, *D*=20
2S states









CONCLUSION

The choice of an initial state of atoms and molecules can have a significant effect on attosecond pulse production.

Optimal preparation of an excited molecule medium can lead to highly enhanced IR-XUV frequency conversion and provide efficient control of the spectral and temporal distributions of attosecond radiation.

Strong bond-length and orientation dependence of emission spectra can be used for continuously tunable coherent X-ray production and for probing molecular vibration-rotational dynamics.

Compromising excitation type, molecule orientation and Lorentz force effect can result in production of single pulse with ≈ 10 attosecond duration.