High harmonic generation and multi-photon ionization of atomic hydrogen in intense laser fields: a computational perspective

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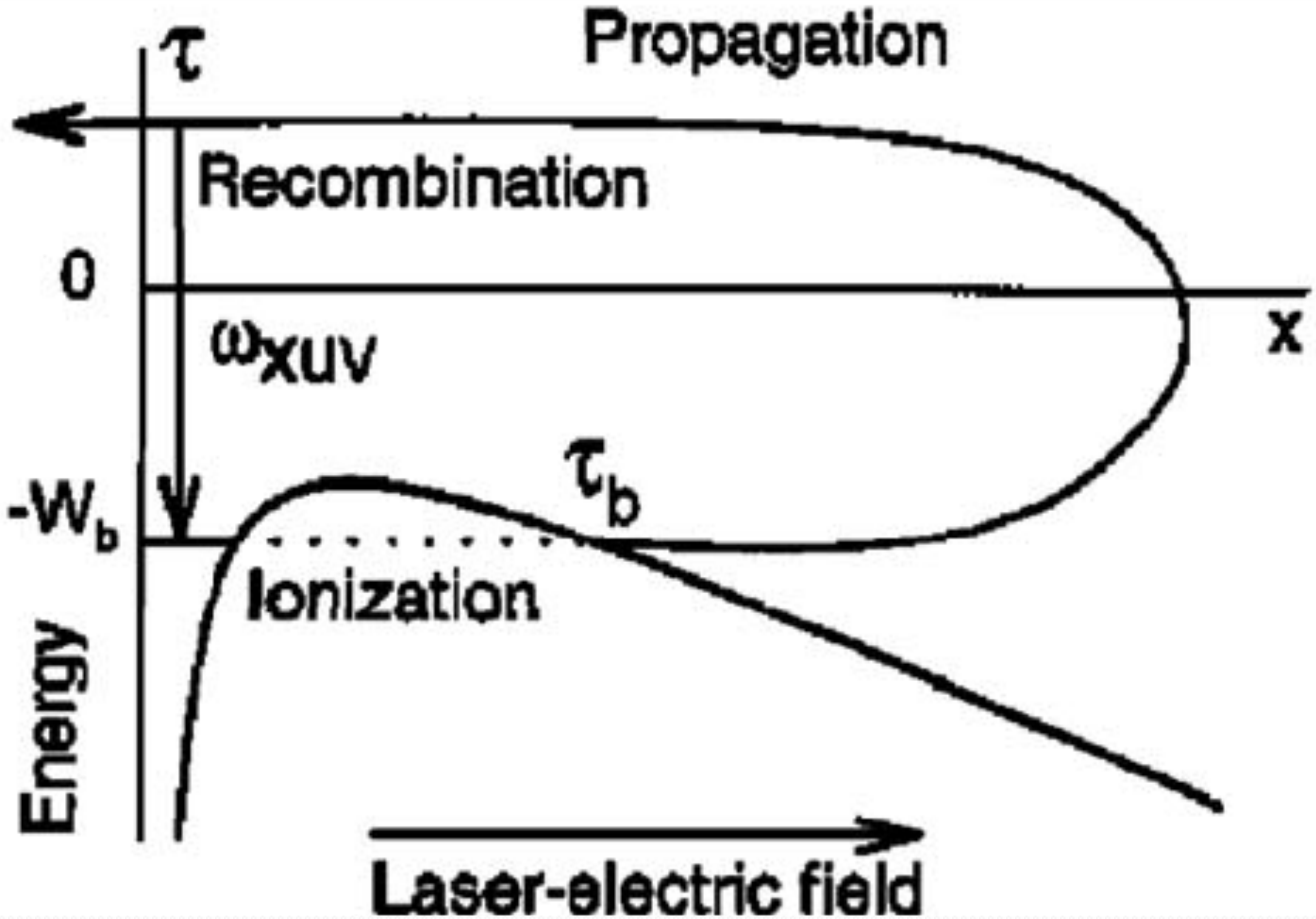
York Seminar: Sept 23, 2014

Outline

- Expt.: HHG with 2-color laser for $He(1s^2)$
- TDSE 3d: pseudospectral method for H(1s)

- Expt.: Attoclock measures tunneling time?
- TDSE 3d: dynamics of ionized wavepacket formation

Three-step model of HHG



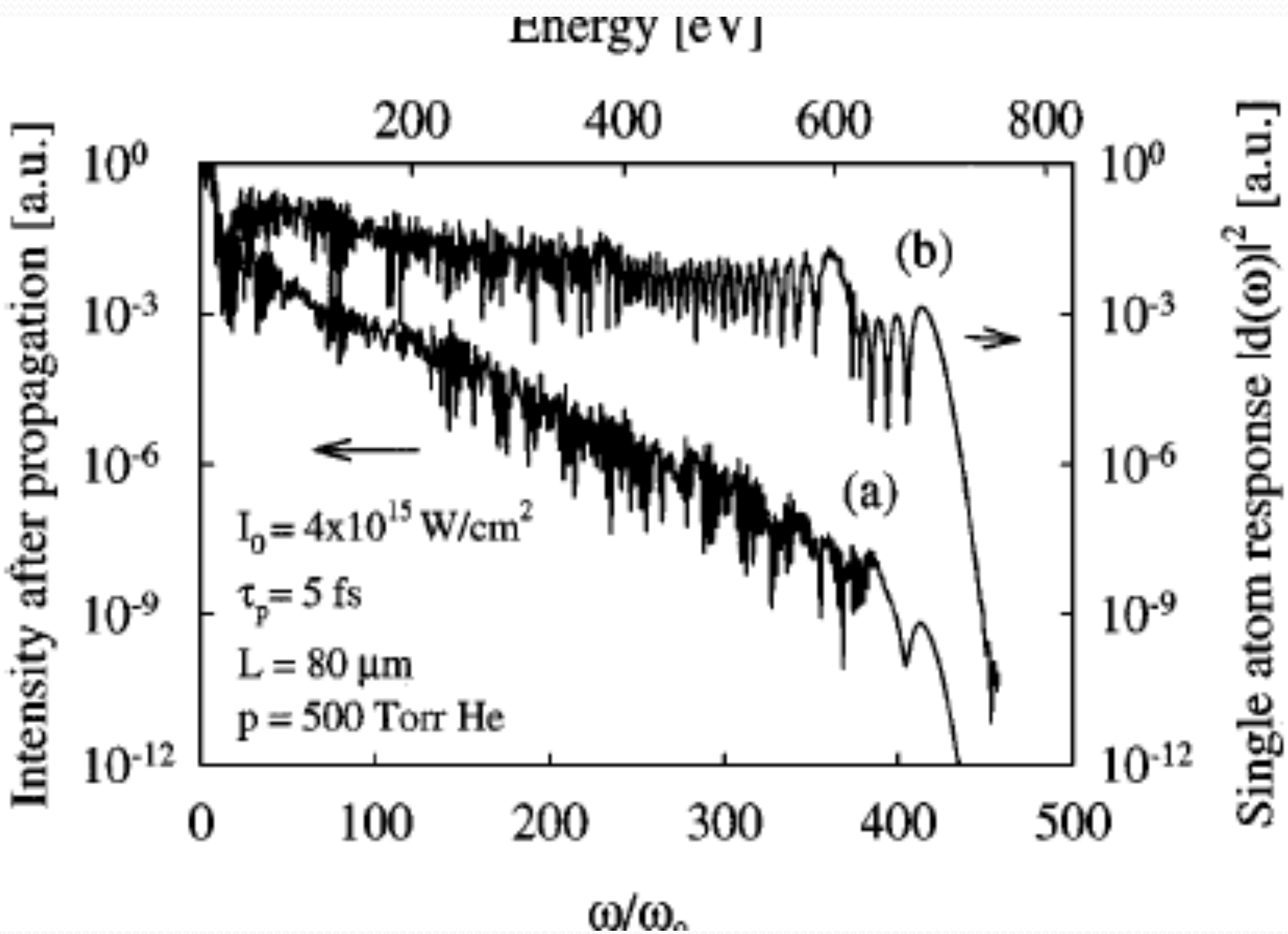
Ottawa Experiment (Corkum)

- HHG plateau cut off at max. return kinetic energy
- IR laser: odd-order harmonics can go up to ~400
- Lower for Ti:sapphire laser (ω=0.057 au, 800 nm)
- New idea: add a perpendicular, weaker laser at 2ω;
- How is recombination affected?
- Use thin gas jet target: no phase-matching calculation required, single-atom response only

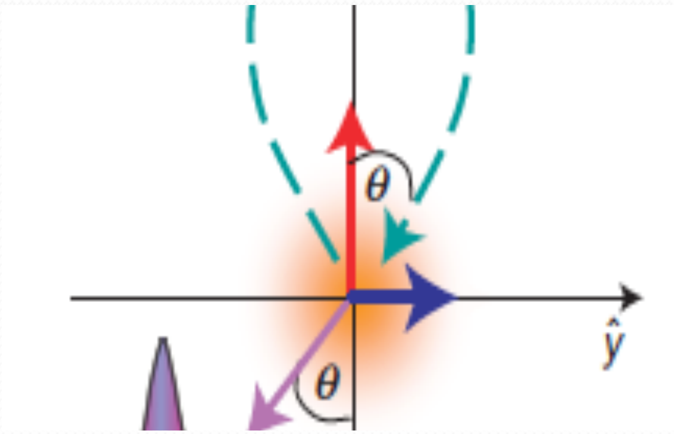
Thick target: propagation effects

PRL80, 3236 (F.Krausz et al., 750 nm laser):

theoretical simulation matched well by experiment



Shafir, Corkum, et al., Nature Ph.,2009



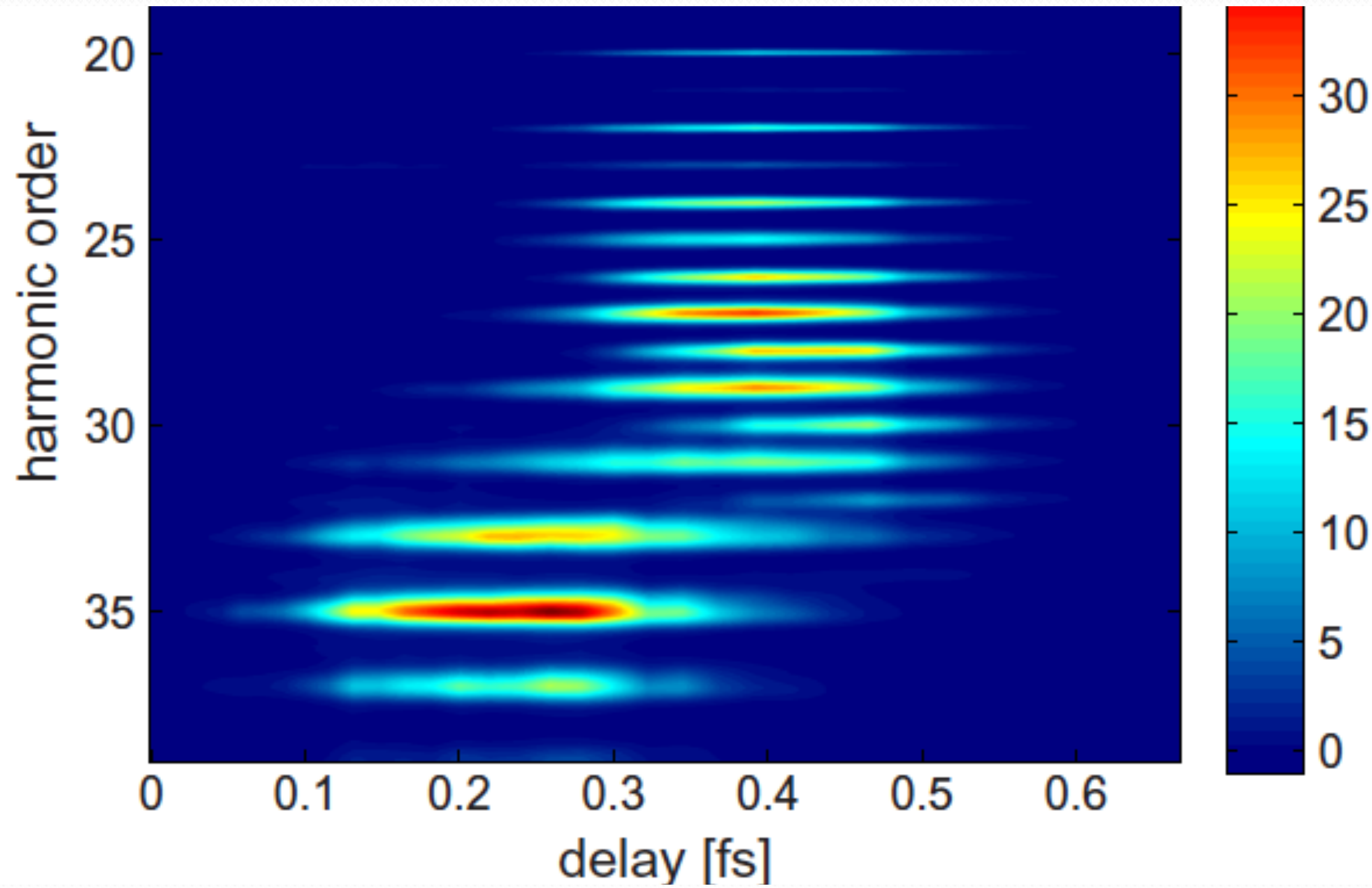
$$E_{\text{odd}} = E_x\hat{x} = E\cos(\theta)\hat{x} \qquad E_{\text{even}} = E_y\hat{y} = E\sin(\theta)\hat{y}$$

$$E_{n\omega_0} \propto E_x\hat{x} + E_y\hat{y} - e^{-in\pi}(E_x\hat{x} - E_y\hat{y})$$

HHG radiation is polarized along returning electron's **v**:
Resolve its components, know θ vs phase delay.

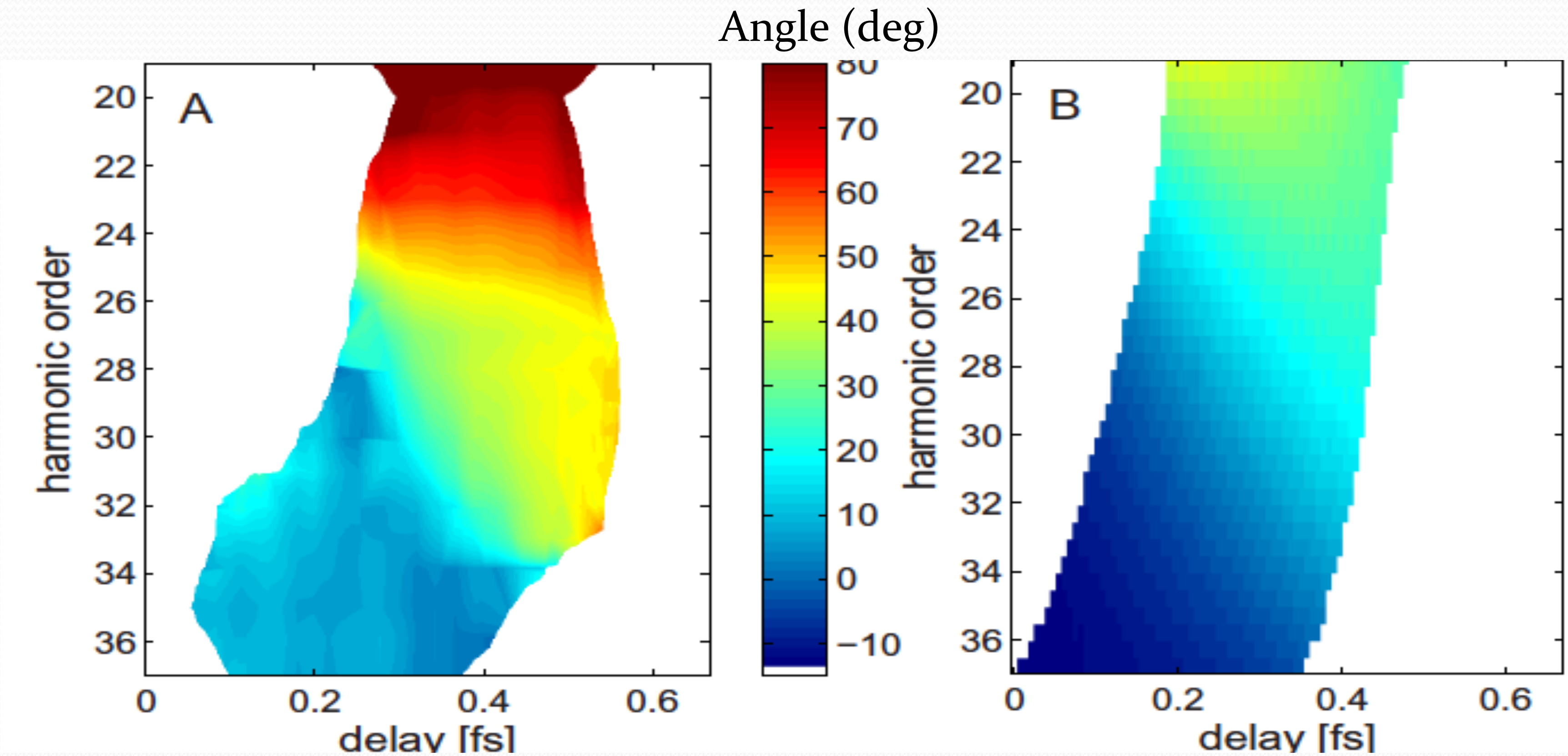
Harmonic E_x-components: odd orders (inversion symmetry)
E_y-components: only even orders (2ω driving field)

HHG as a function of delay $0 < \tau < T/4$



Higher harmonic order: odd orders only!
Intermediate orders: even and odd, even stronger than odd!

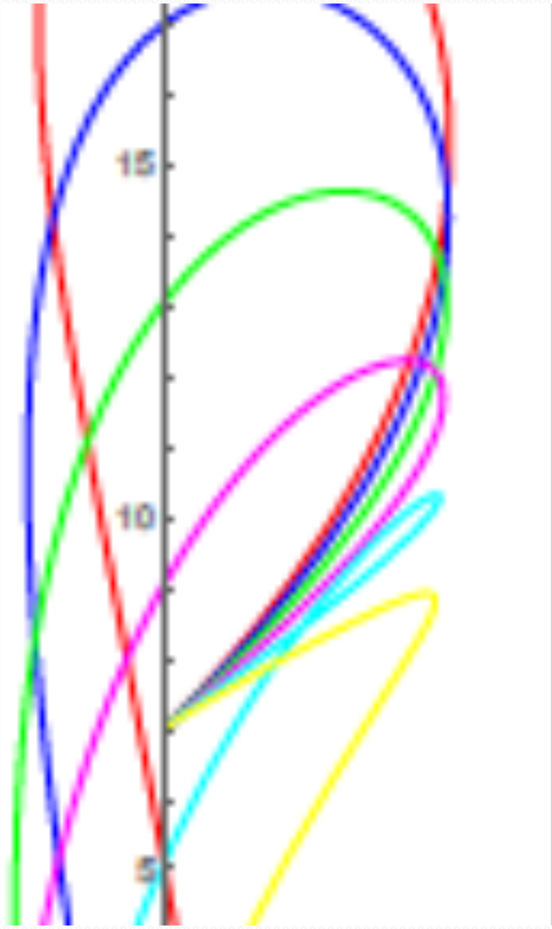
Recollision angles θ: Left=expt, right = class. simulation



Good: odd-only orders regime has near-zero angles θ (blue region, 0.1-0.3 fs delays)

Bad: simulation never gets larger angles than 45 degrees. (doesn't go into red!)

Sample trajectories:



Start particles at the outer tunneling point

Vary the phase delay (different colors)

Trajectories become meaningless near nucleus

Re-collision angles tend to be small !

Coulomb interaction (in plane) is included

Quantum calculation: PRA88, 063419

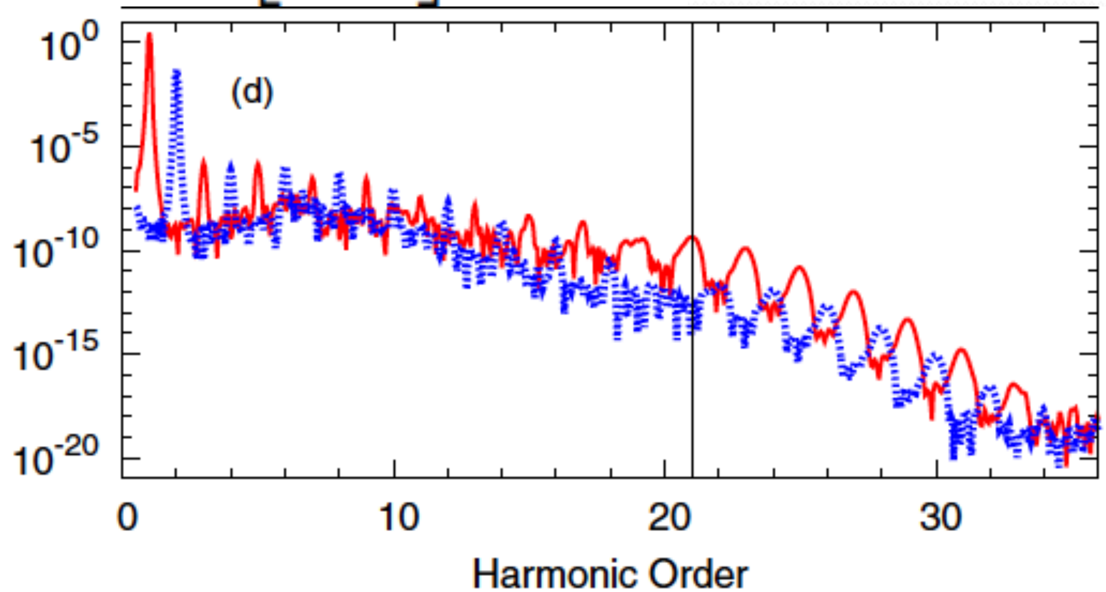
will it get larger recollision angles?
(i.e., strong even vs odd harmonics at low order)

- Do H(1s), not He (more stable numerically)
- Solve TDSE, calculate <a_x>(t), <a_y>(t), Fourier an.

$$a_x = -[\mathcal{H},[\mathcal{H},x]] = \left[\mathcal{H},\frac{\partial}{\partial r}\right]\sin\theta\cos\phi$$

- Power spectra:
- Red = x (odd),
- Blue = y (even)

One delay shown



TDSE H(1s) results for recollision θ

Expt is for $0<\Delta\phi<\pi/2$ only

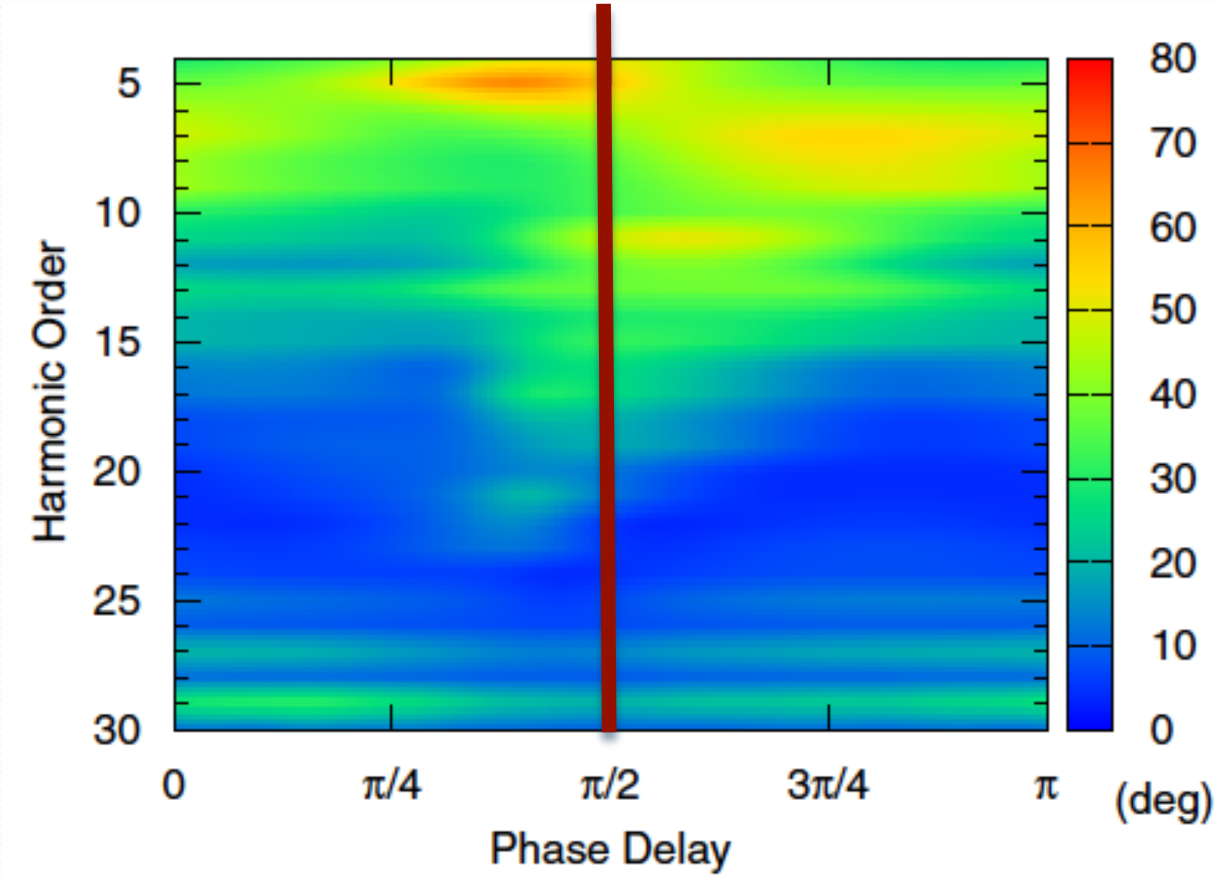
QM: do get θ up to 75 deg

Some of the pattern is OK

Absolute values of HHG orders are lower in H(1s)

Should push the QM calc. to He in SAE model.

Expt should explore longer delays.



QM does better, as the complete wavepacket is taken into account?

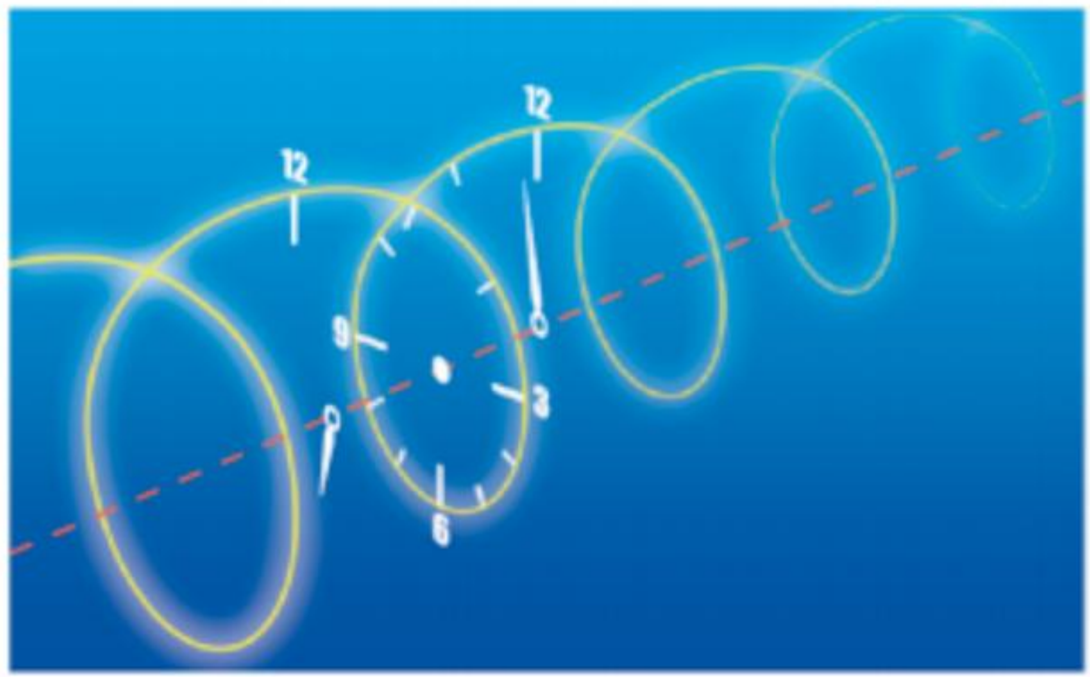
Attoclock: elliptically polarized laser

Rotating electric field vector provides a time reference on the sub-fsec scale.
Ursula Keller et al, ETH Zűrich, Nature Physics, 7, 371 (2011).

Tunnel ionization at the peak of the multi-cycle laser pulse;
follow classical trajectories for ionized electrons;
Detect electrons and parent ions in two dimensions;
Reconstruct: when was the electron released. Time delay?

Rotating E field vector =
Clock handle

Attosecond resolution



TDSE simulations

- Q1: is it really tunneling ionization?
- Photoelectron momentum distributions show multi-photon absorption (MPI) features!
- Q2: Which state does MPI start from? (1s or excited)
- Q3: How does the ionized wavepacket build up?
- 'time delay' = result of prolonged packet formation?

- Idea: monitor the time evolution of *m*-level population
- Classically: freed electron at some distance picks up angular momentum due to torque exerted by rotating electric field.

TDSE method: 3d extension of PRA74, 031405 (XM Tong et al)

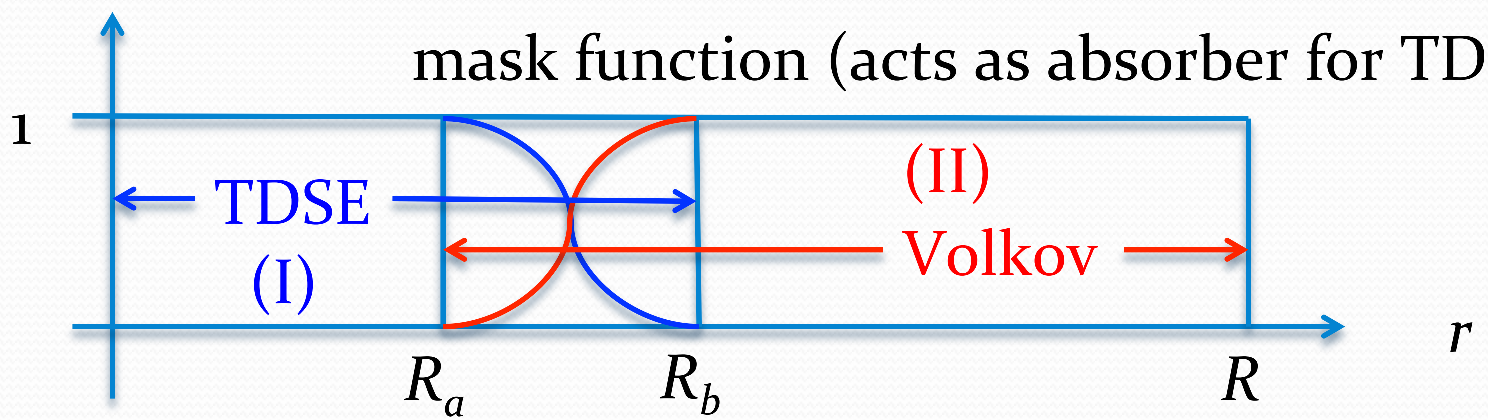
$$i\partial_t \Psi(\vec{r},t) = \left[-\frac{\nabla^2}{2} - \frac{1}{r} + f(t)\vec{E}(t)\bullet\vec{r} \right] \Psi(\vec{r},t) \qquad f(t) = \text{envelope fn}$$

$$\Psi(\vec{r},t) = \sum R_{lm}(r,t) Y_{lm}(\Omega) \qquad l = 0..128,\ m = -l..l$$

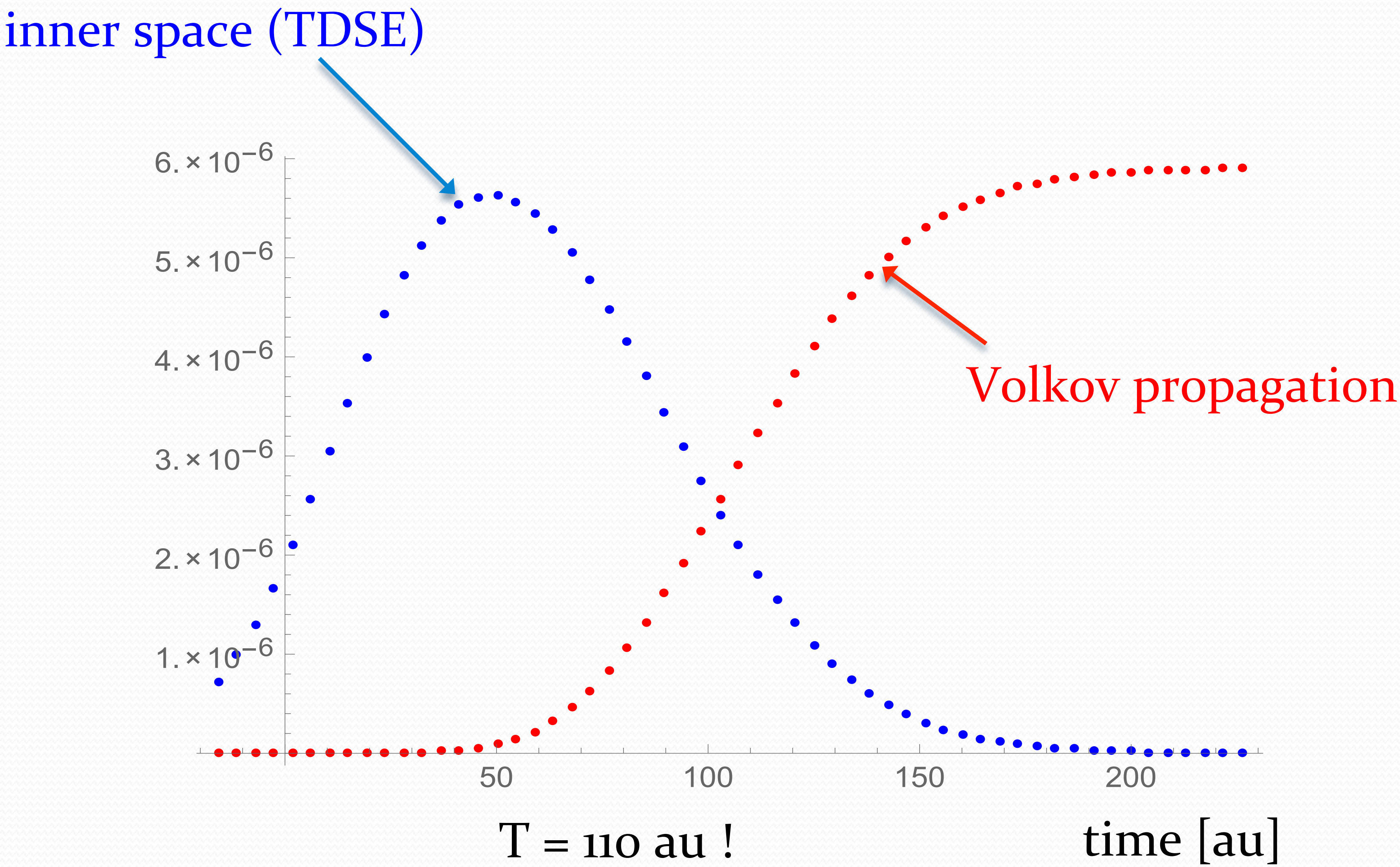
Pseudo-spectral method:
clever matrix representation, uses Ψ at grid points
Grid in SPC (r, θ, φ) acts as collocation mesh

Avoidance of reflection: radial space is split into (I) and (II).
Regions (I) and (II) overlap with coherent wavefunction transfer.
In (II) a Volkov propagator is used (momentum space, VG)

mask function (acts as absorber for TDSE)

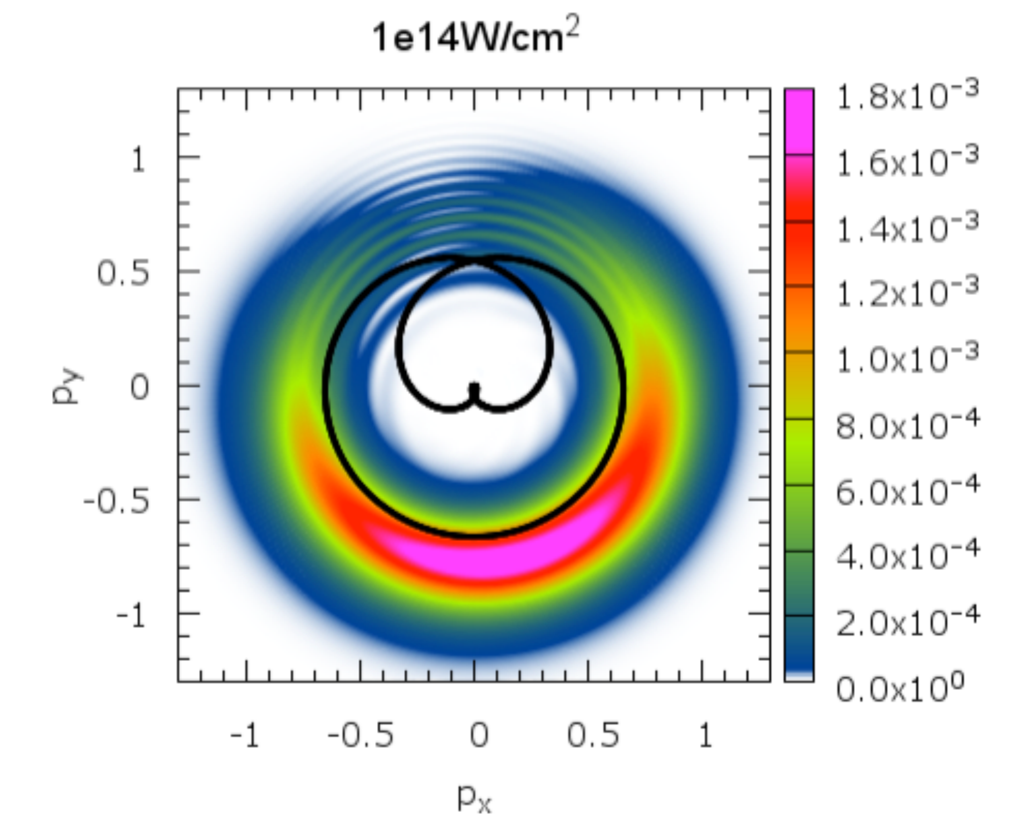


Ionization probability grows over time



Electron spectrum: *x-y* plane; 3-cyc. CP

p_y

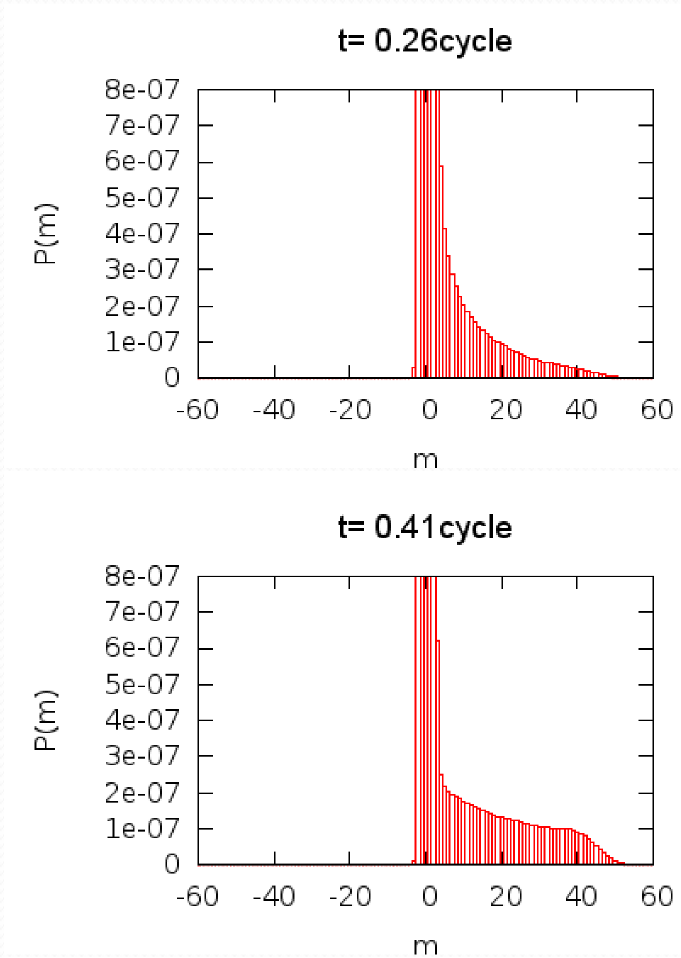


p_x (au)

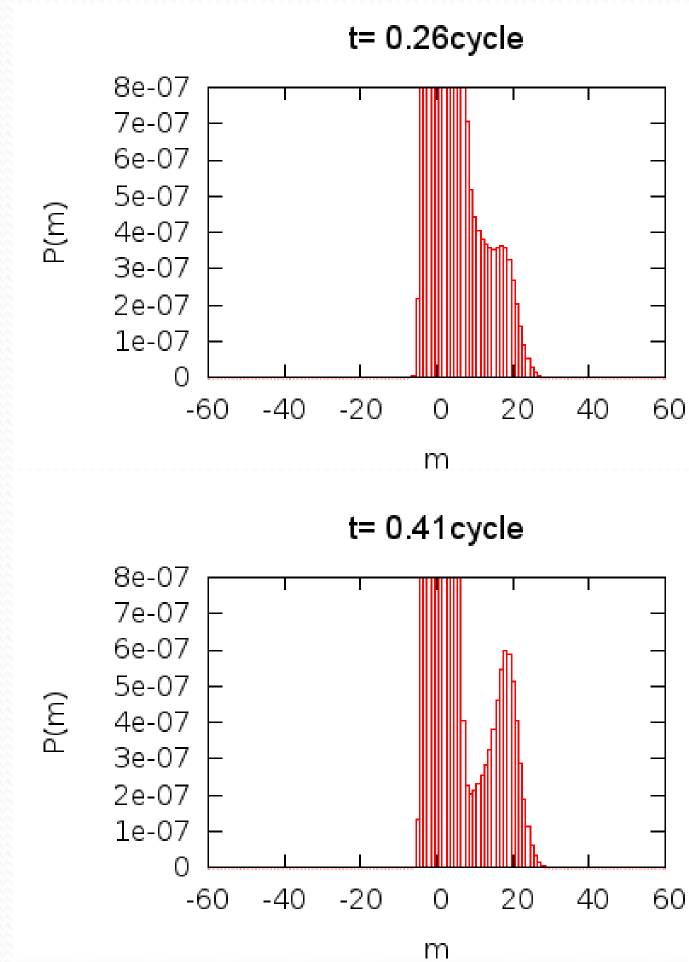
Field E_x pushes e$^-$ out to the left (tunneling?)
Then the field E_y imparts momentum on free e$^-$
Classical naïve result: [$-A_x(t)$, $-A_y(t)$] parametric plot:
$\varphi_p = 270°$ and $\mathbf{p} = [-A_x(0), -A_y(0)]$

m-level population over time

Length gauge (calc.)



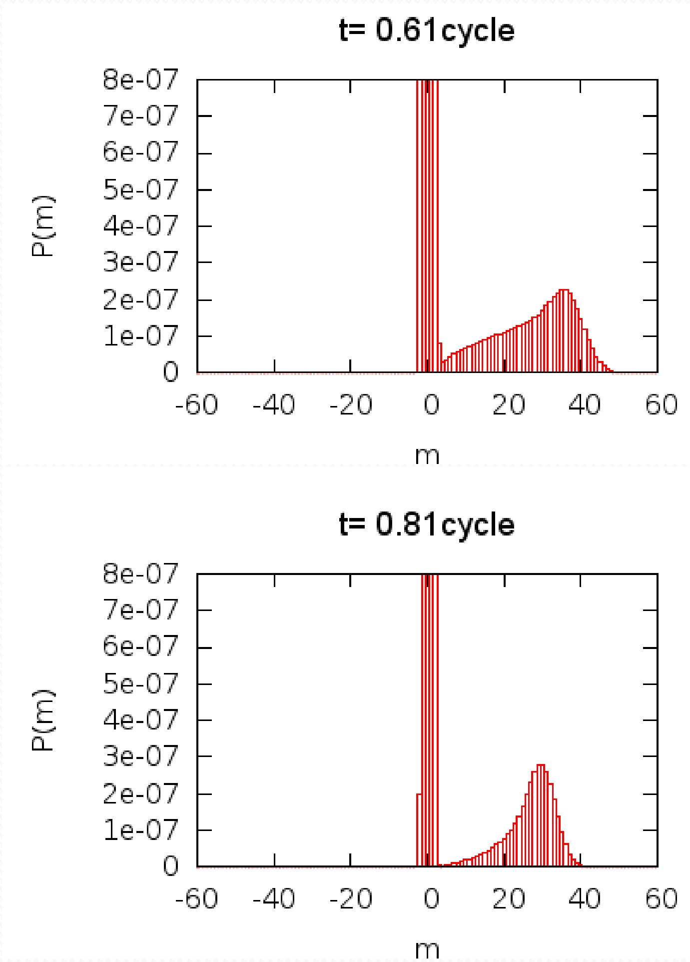
Transformed to velocity gauge



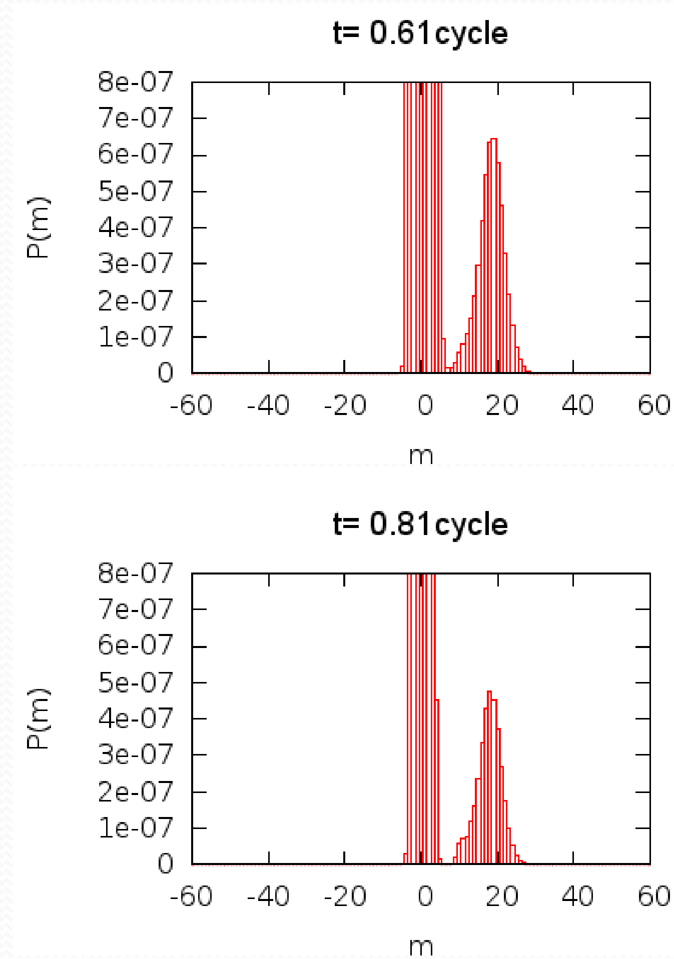
Evolution of $\langle L_z \rangle$ is a gauge-dependent result

m-level population over time

Length gauge (calc.)



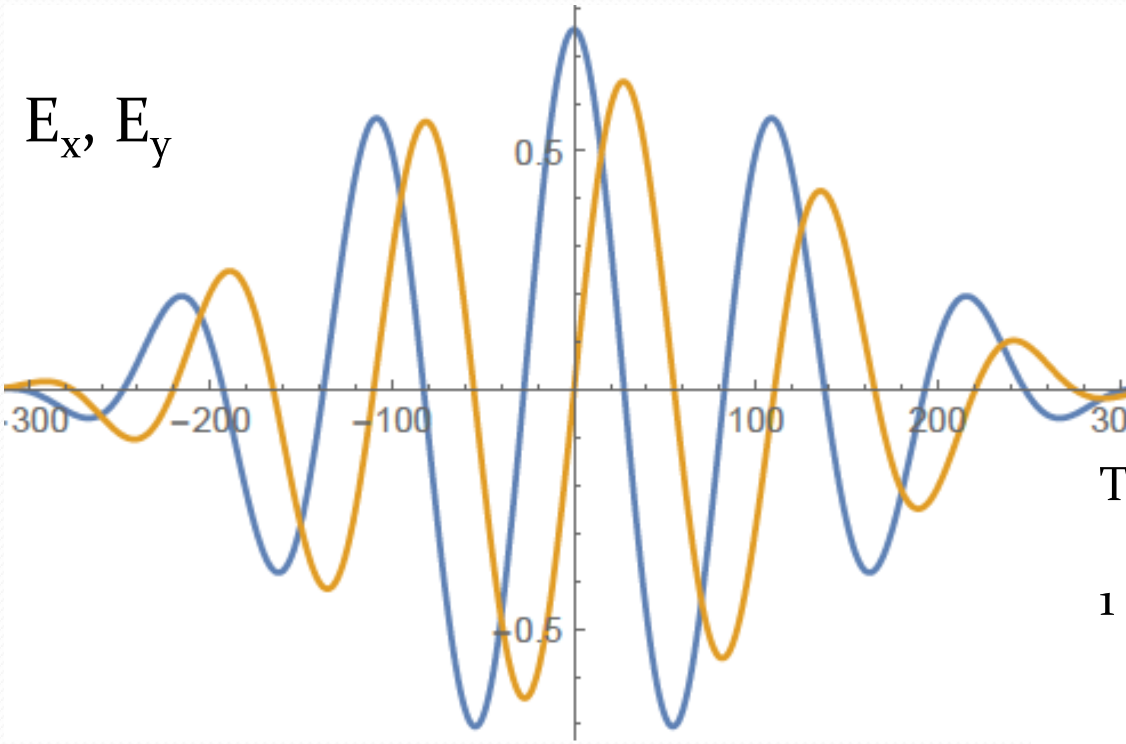
Transformed to velocity gauge



CP laser exerts torque on free electron -> angular momentum L_z

QM: electron is launched into high-***A*** region: 'ang. mom. potential'

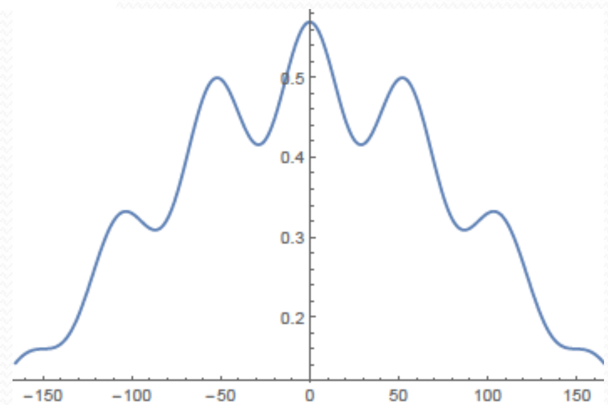
6-cycle pulse, Elliptic Polarization



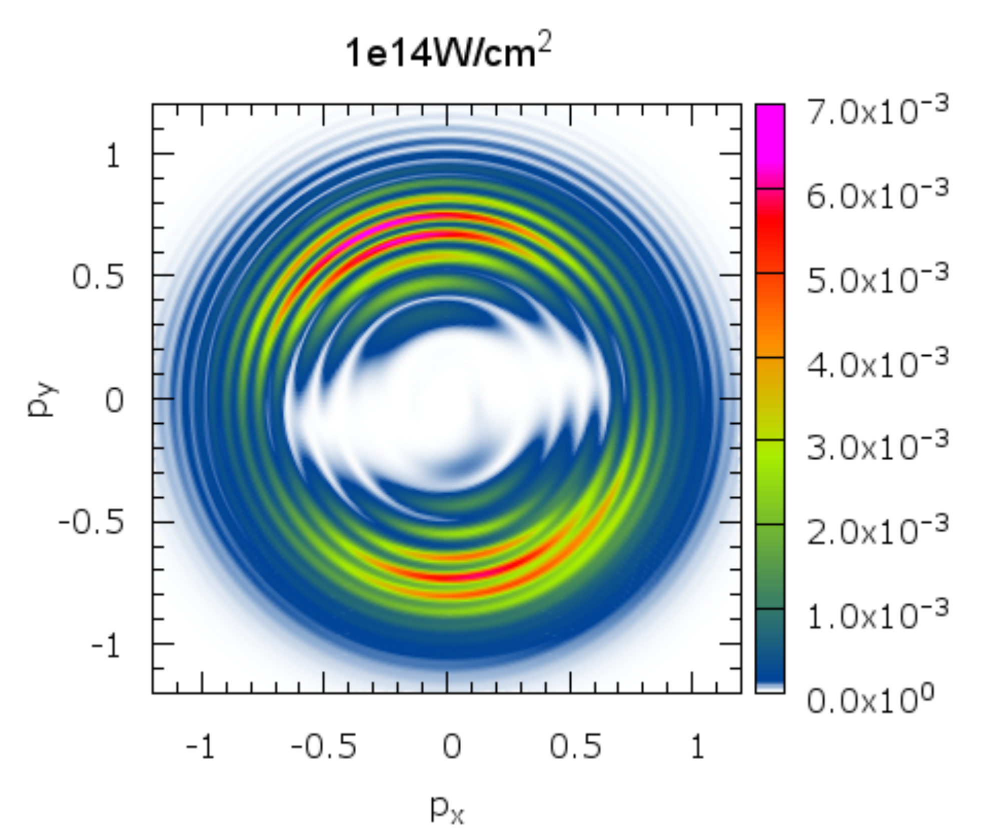
Time (au)

1 au = 24.2 attosec

Small ellipticity, ω=0.057 au, (800 nm)
Innermost cycle matters !
Central + 2 side peaks in intensity ->
(caused by ellipticity)
Intensity range: 10^{13}-10^{14} Wcm^{-2}
E_o < 0.1 au



6-cycle EP result $I_0 = 10^{14}$ Wcm^{-2}



Ring structure = multi-photon ionization (MPI) ?
Why two structures on opposite sides ?
Three times higher peak electron yields compared to 3cyc CP

Test MPI hypothesis

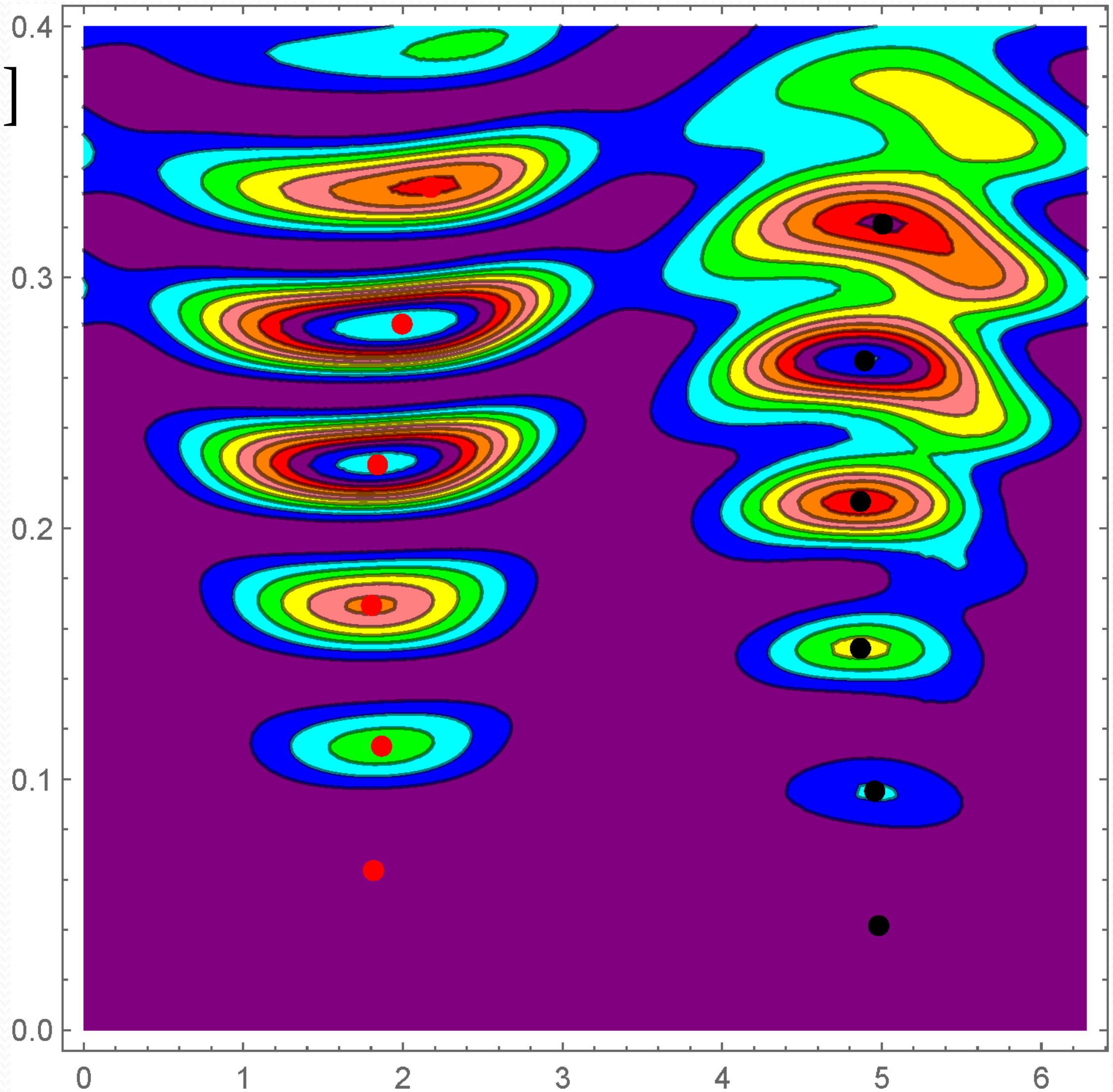
Low orders suppressed

Spaced by ħω

φ_p≈270 peaks shifted in energy vs the φ_p≈90

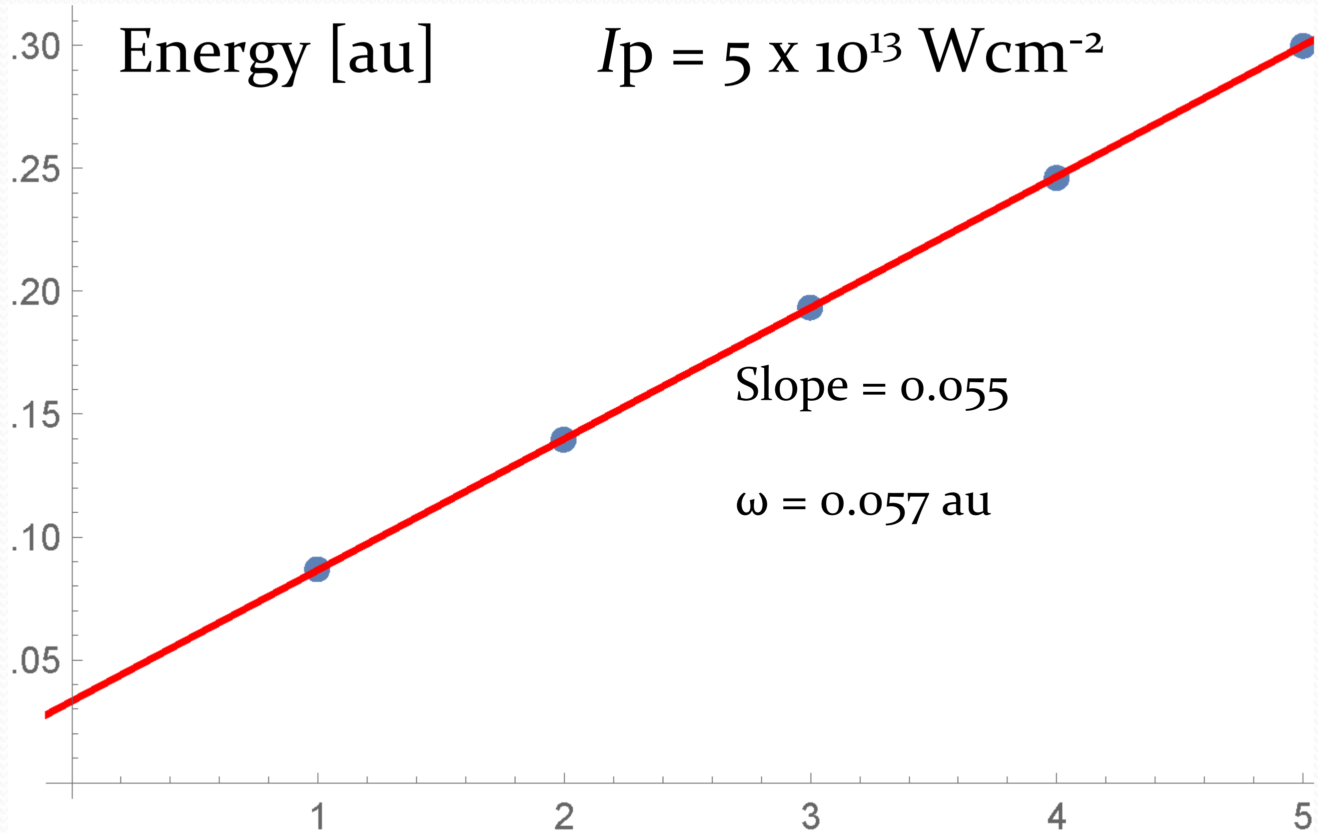
Idea: φ_p≈270 comes from t=0; φ_p≈90 from two neighbor intensity peaks

$p^2/2$ [au]



φ_p [rad]

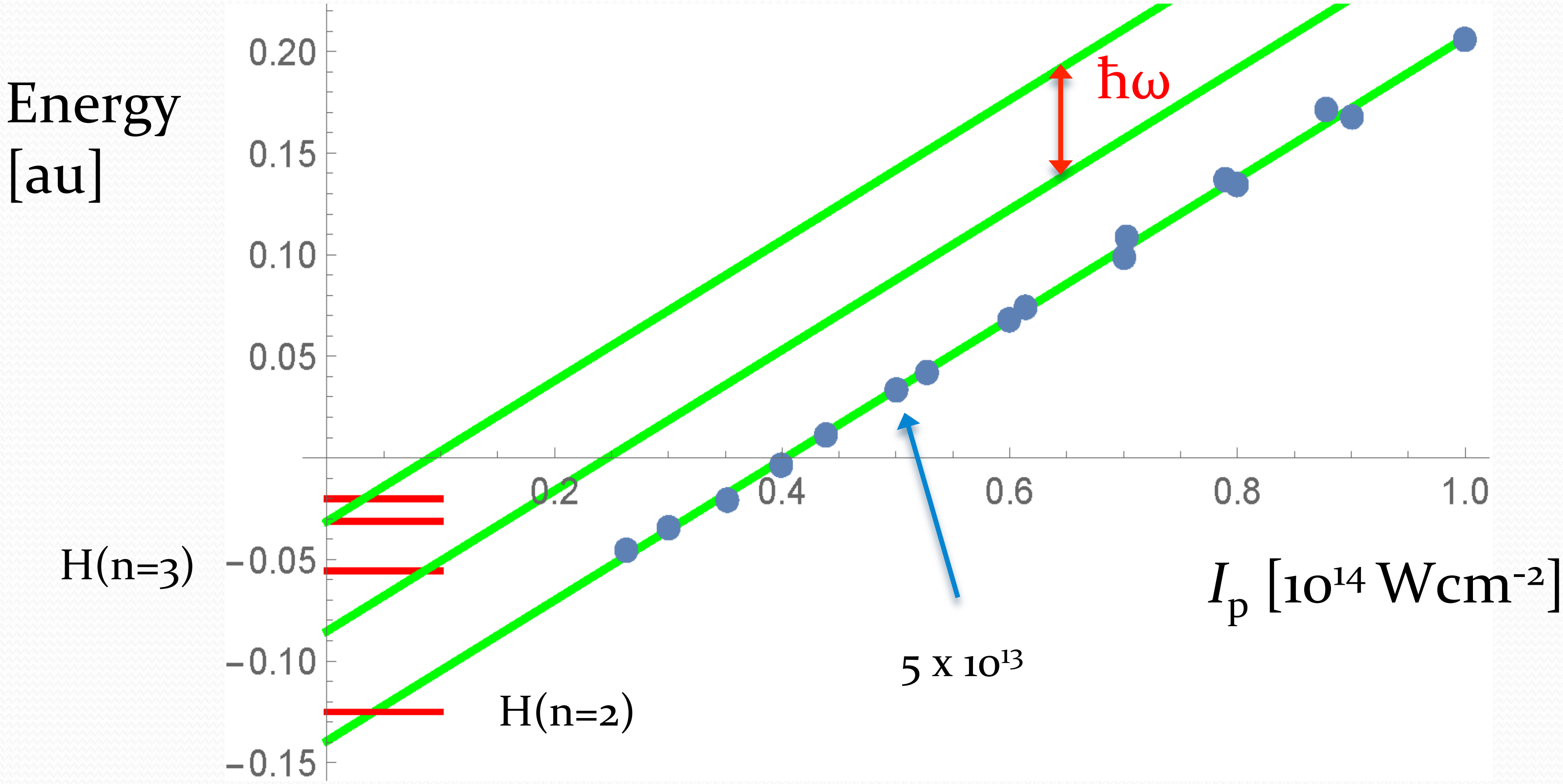
Fitting the peaks



photon order

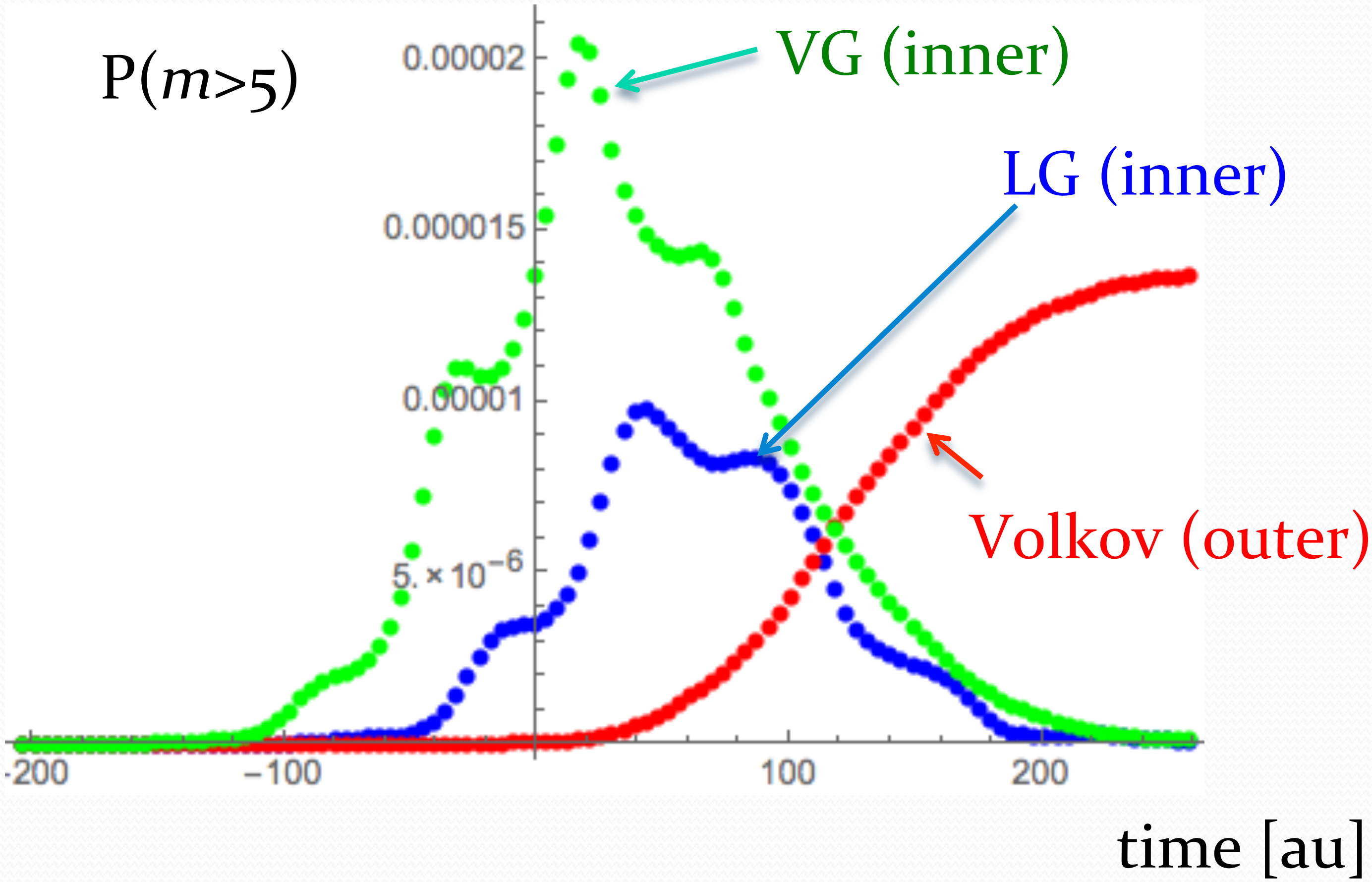
Record energy for order zero (ambiguous) vs peak intensity I_p
Understand ponderomotive shifts
Which *n*-level did the MPI electron come from?

Ponderomotive shift is ~ I_p or E_p^2



I_p is the peak intensity for t=0, or side peaks (16 data; 8 I_o values)
Data: intercepts of MPI peak fits, absolute photon nr. is unknown

Time delay = tunneling time ??

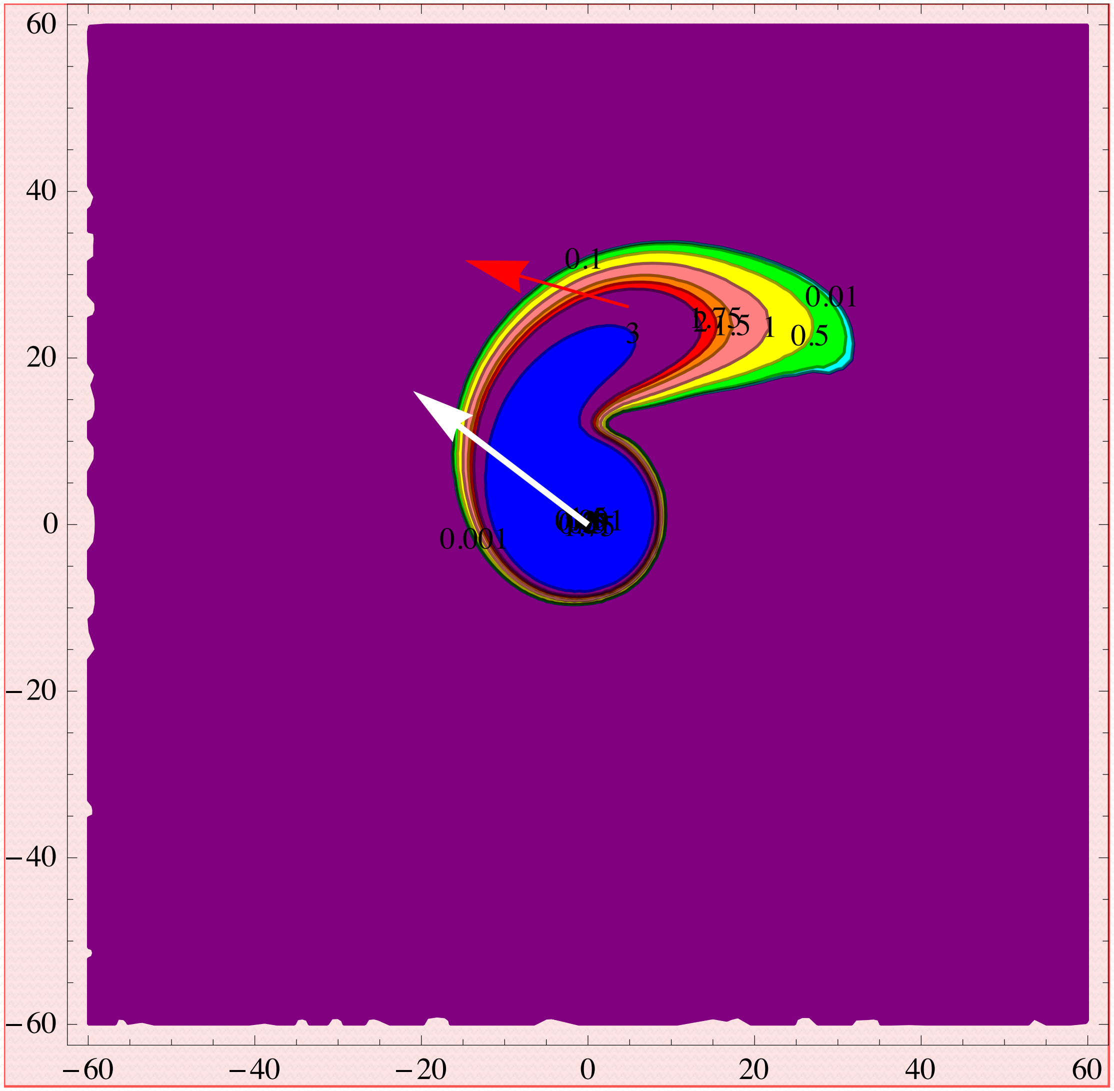


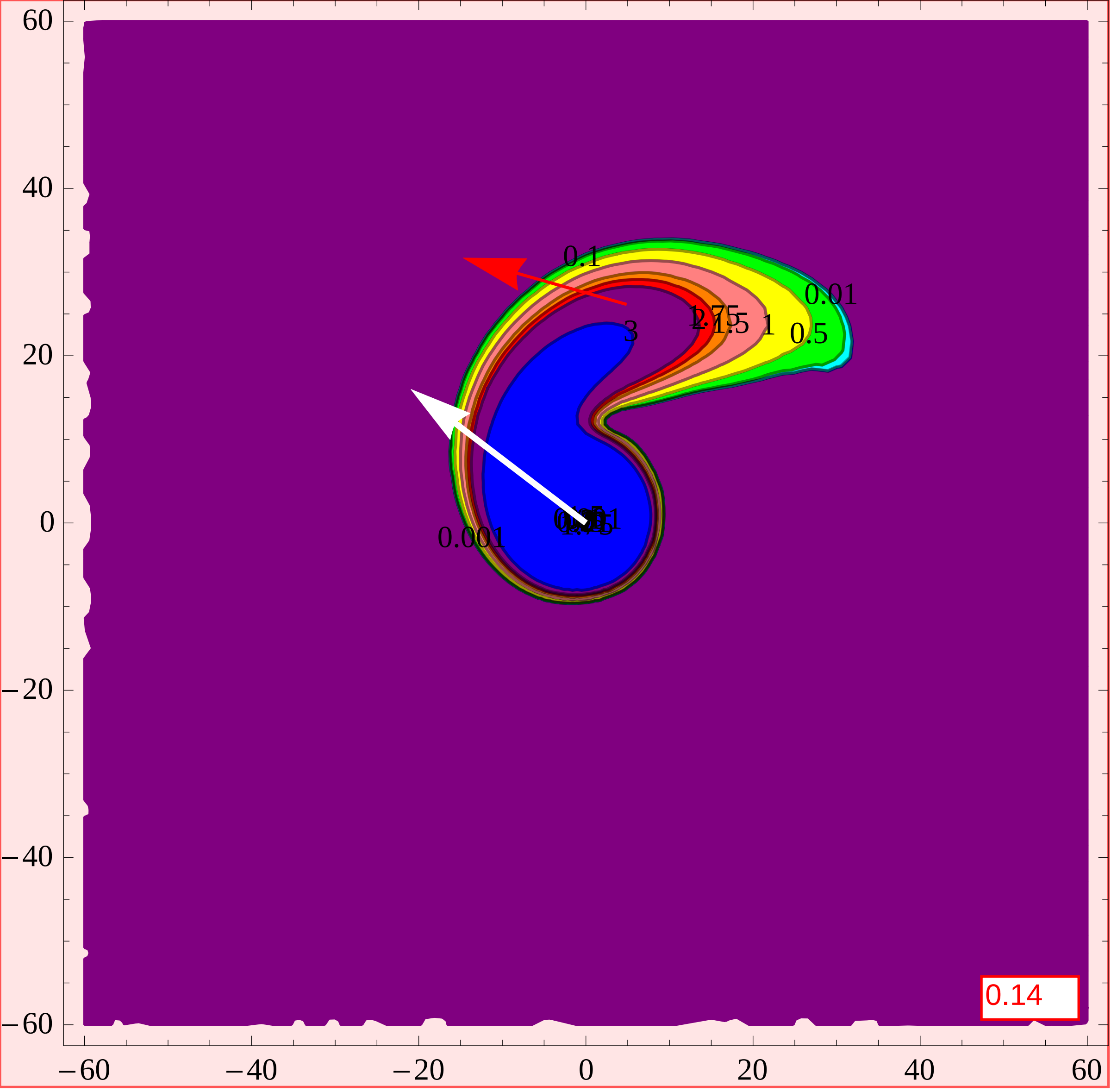
In LG $m>5$: definitely ionization (not temporary bound population)
In VG bound states acquire high m values (oscillate with field)
This is NOT tunneling from the H(1s) state!

Conclusions

- TDSE simulations support the idea of MPI in few-cycle CP or EP laser pulses after pre-excitation
- The idea of pre-excitation was proposed on a classical level by P.Corkum (years ago)
- Ponderomotive shifts (slope in the intercept energy vs I_p line fit) are in line with steady-state formula
- HHG spectra for H(1s) are not as strong as the experimental He($1s^2$) data, but on the right track
- Simulations coupled with analysis provide insights into electron dynamics
- MPI idea extends PRA89, 021402 (2014).

Wavepacket formation

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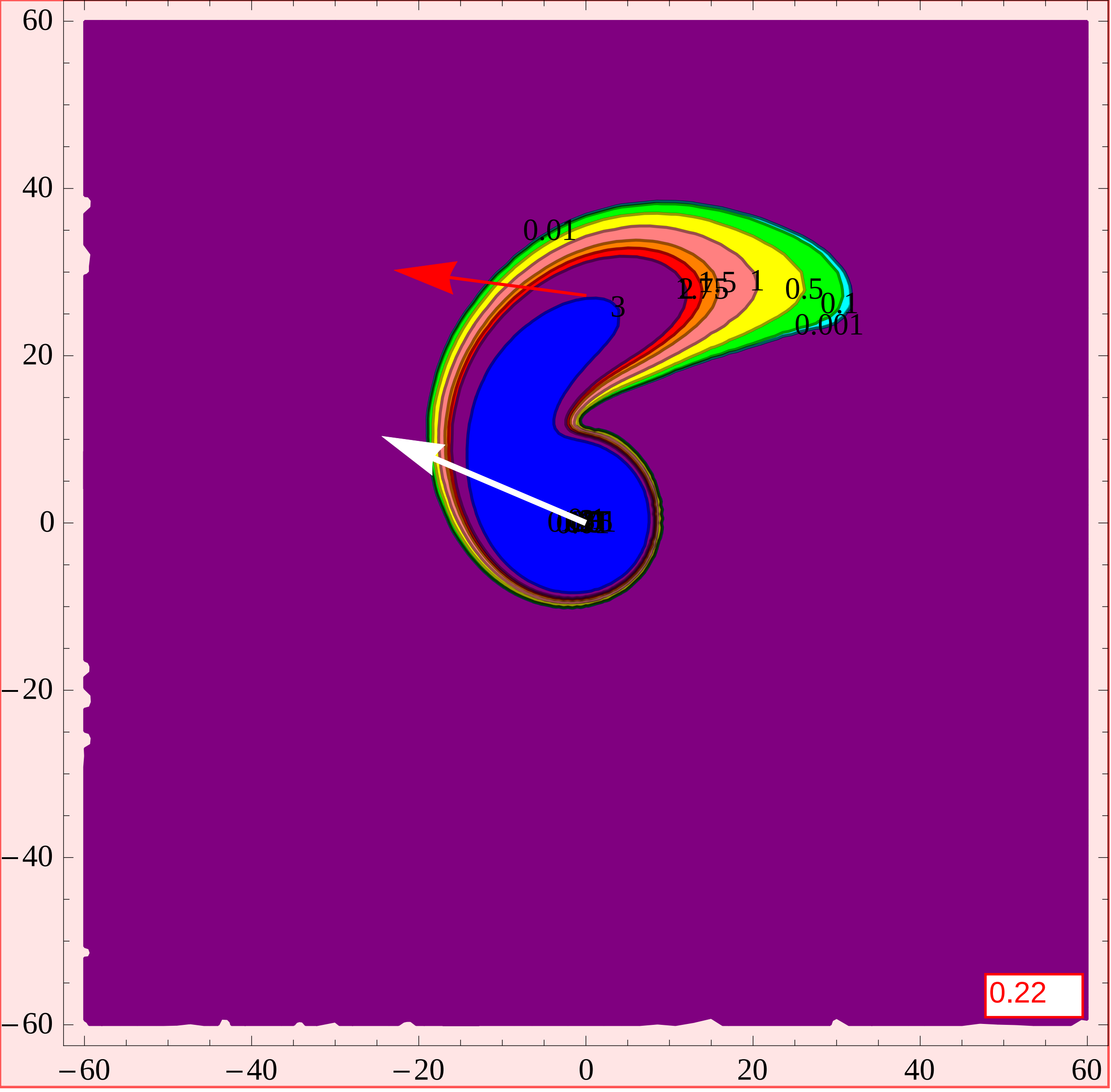
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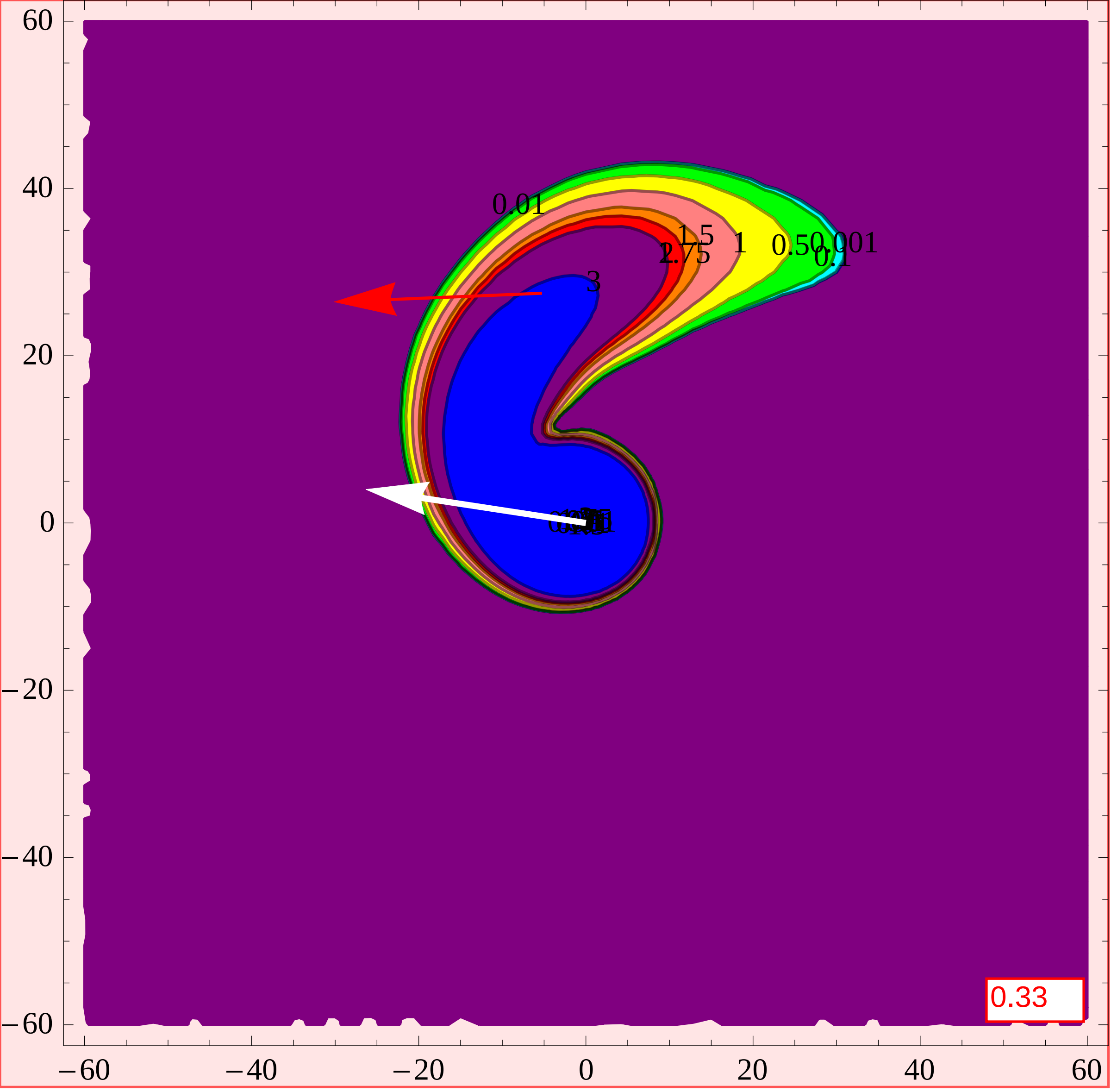

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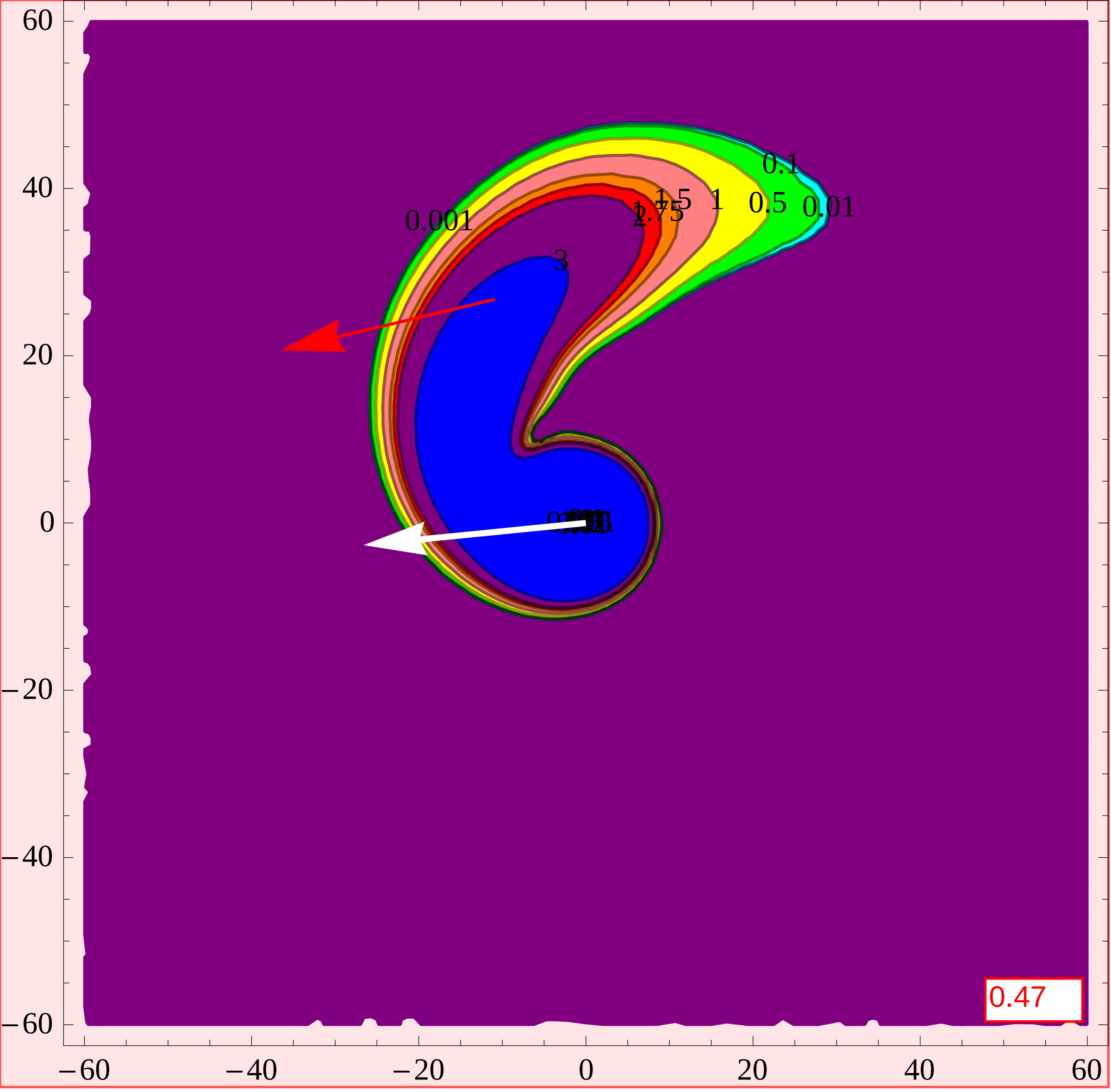

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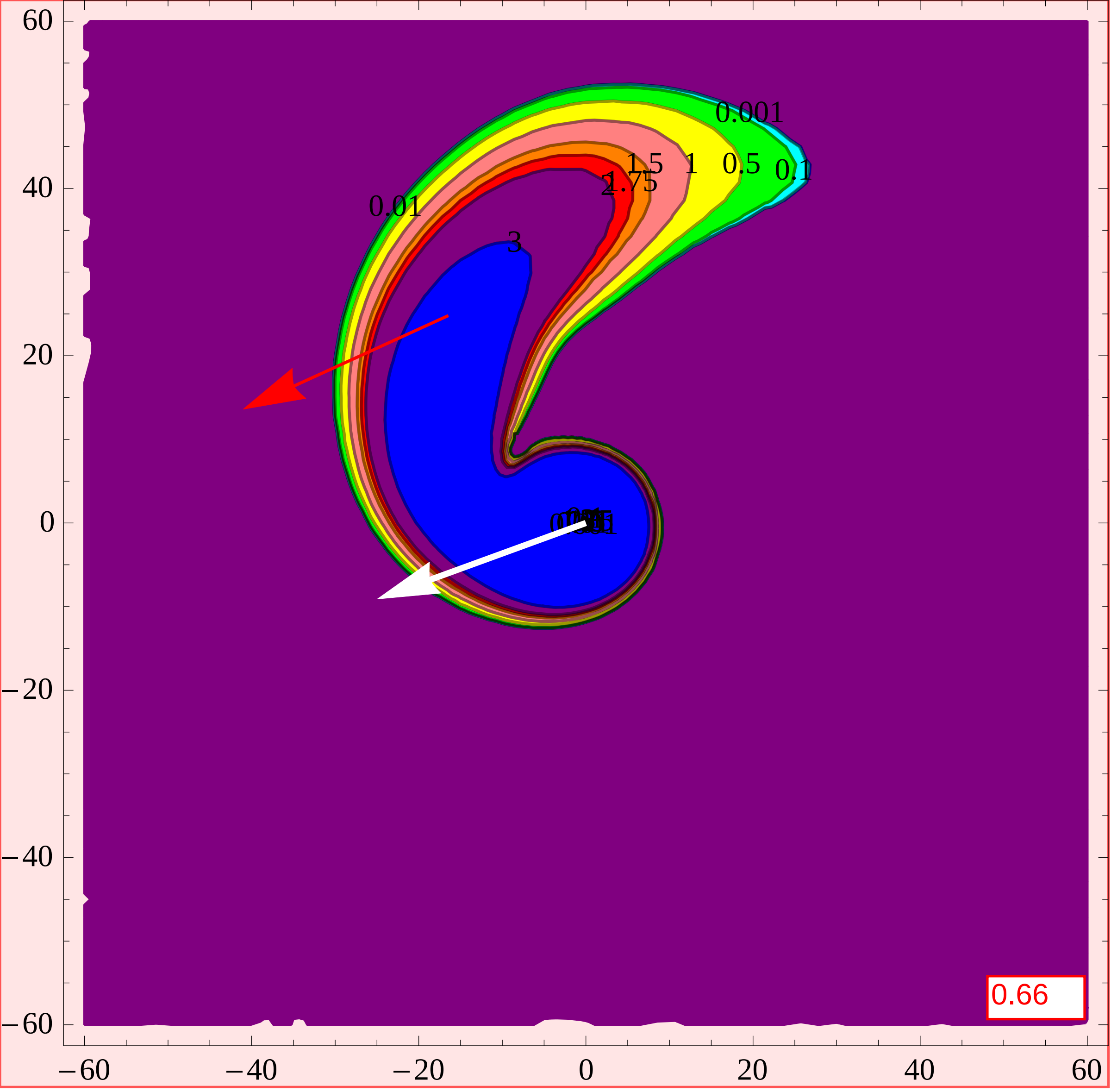

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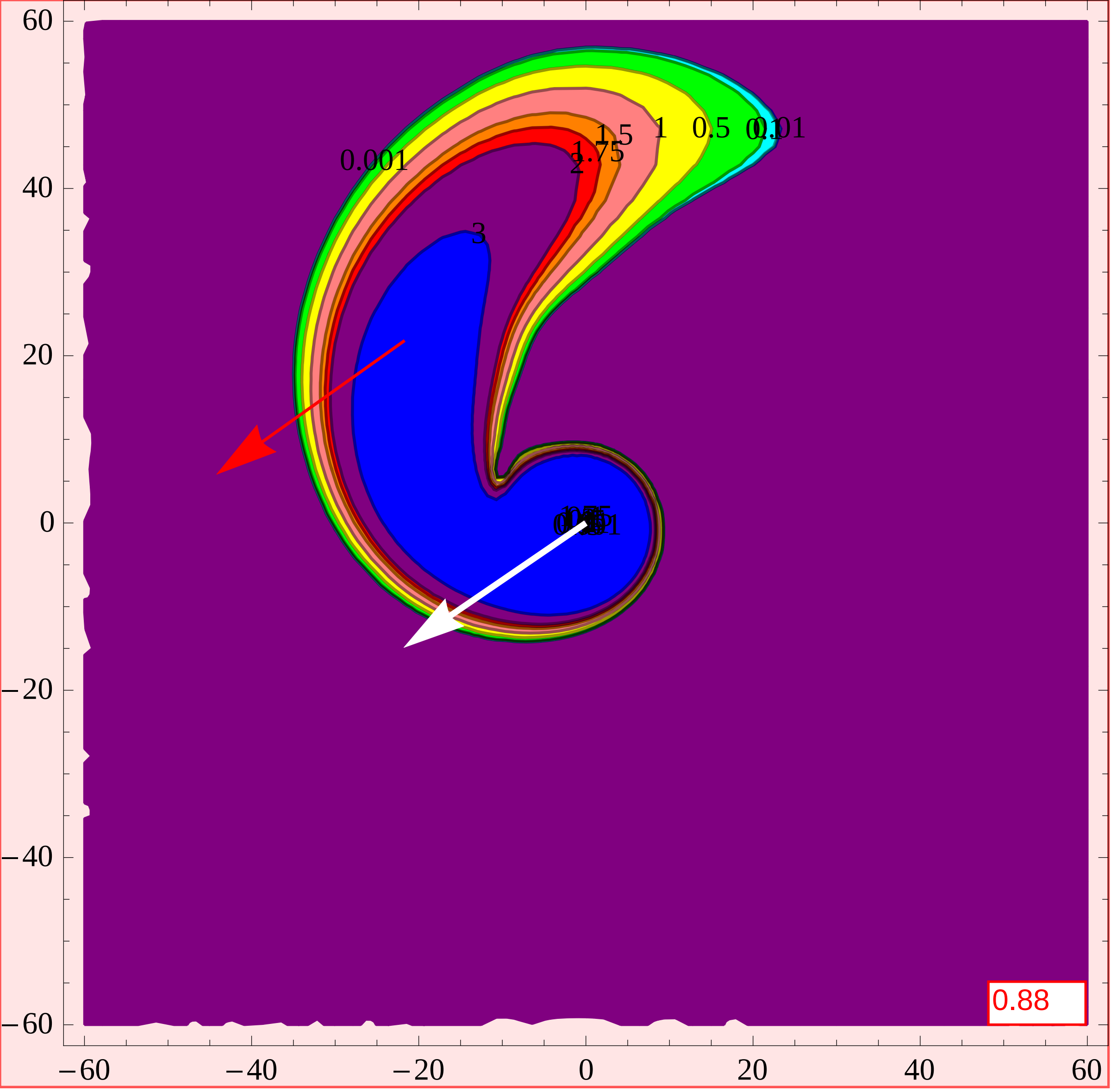

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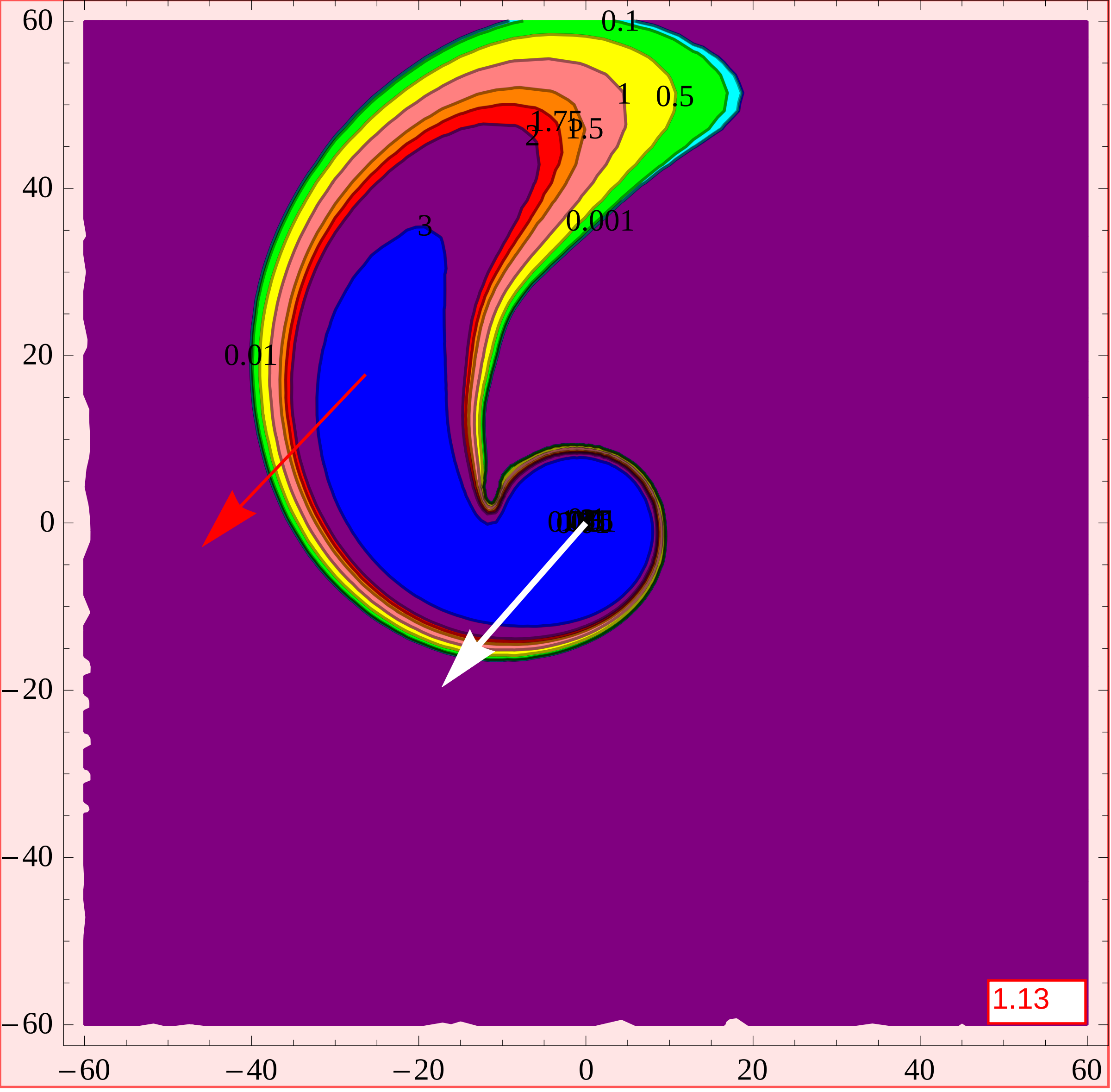

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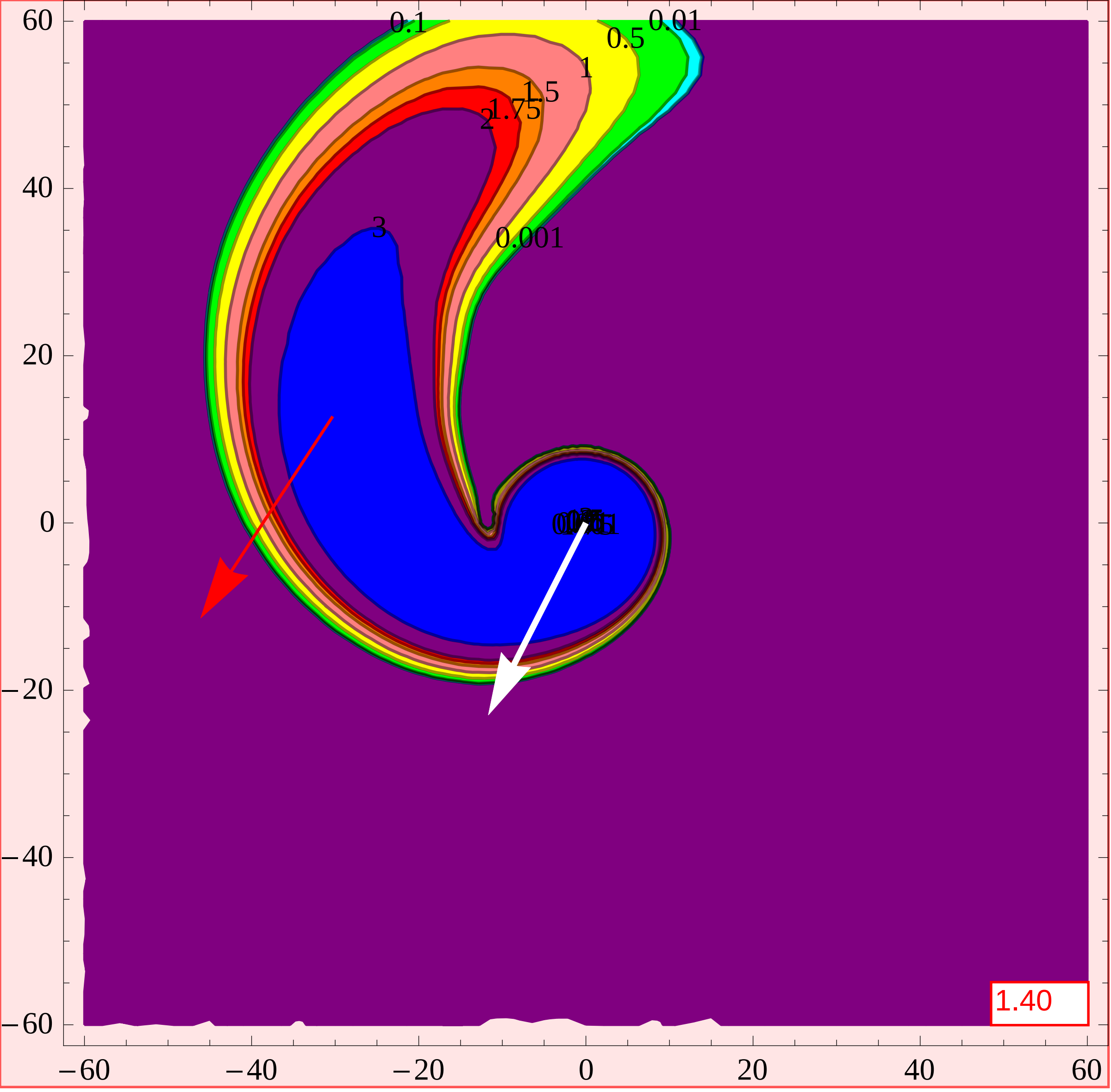
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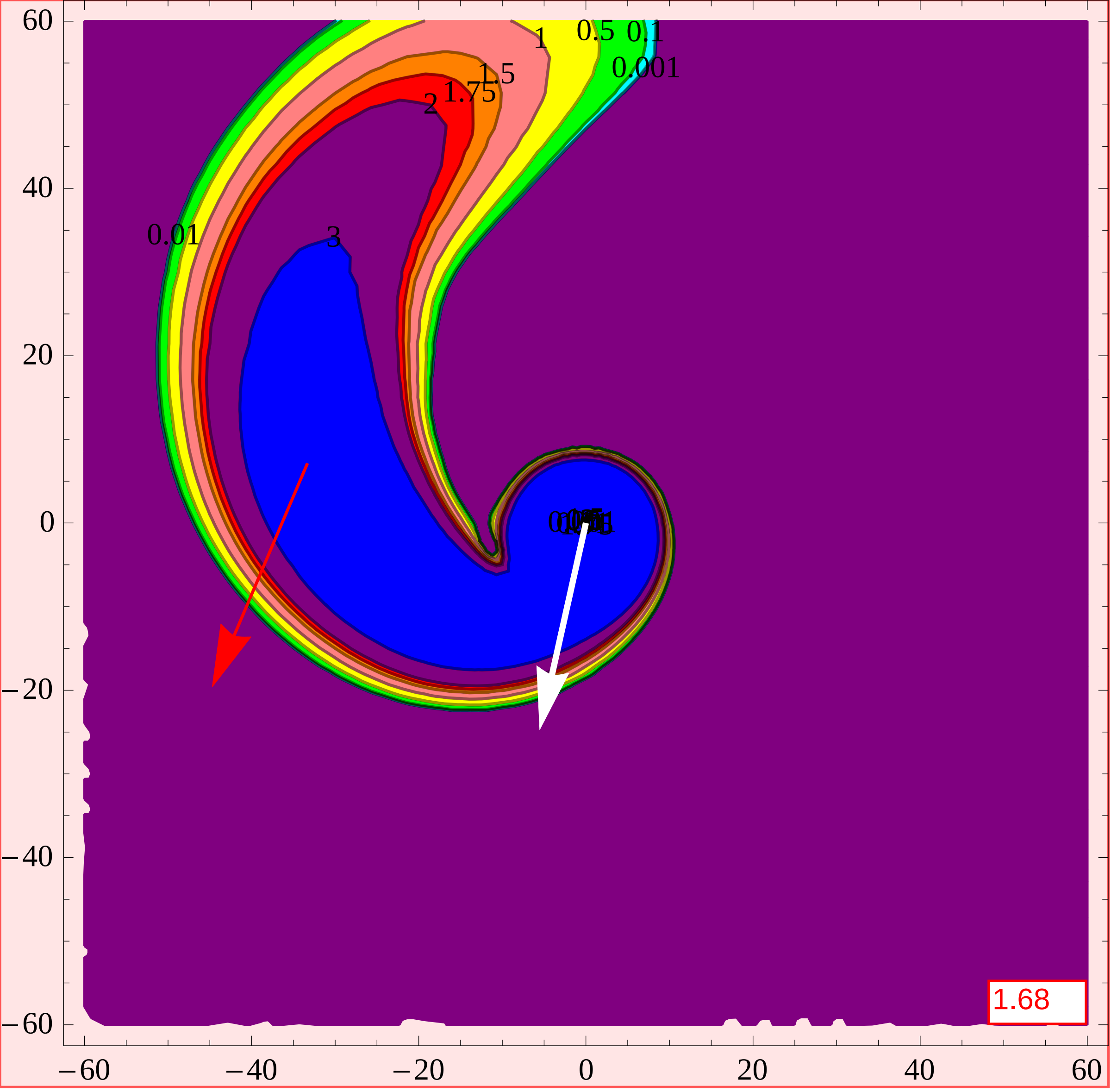

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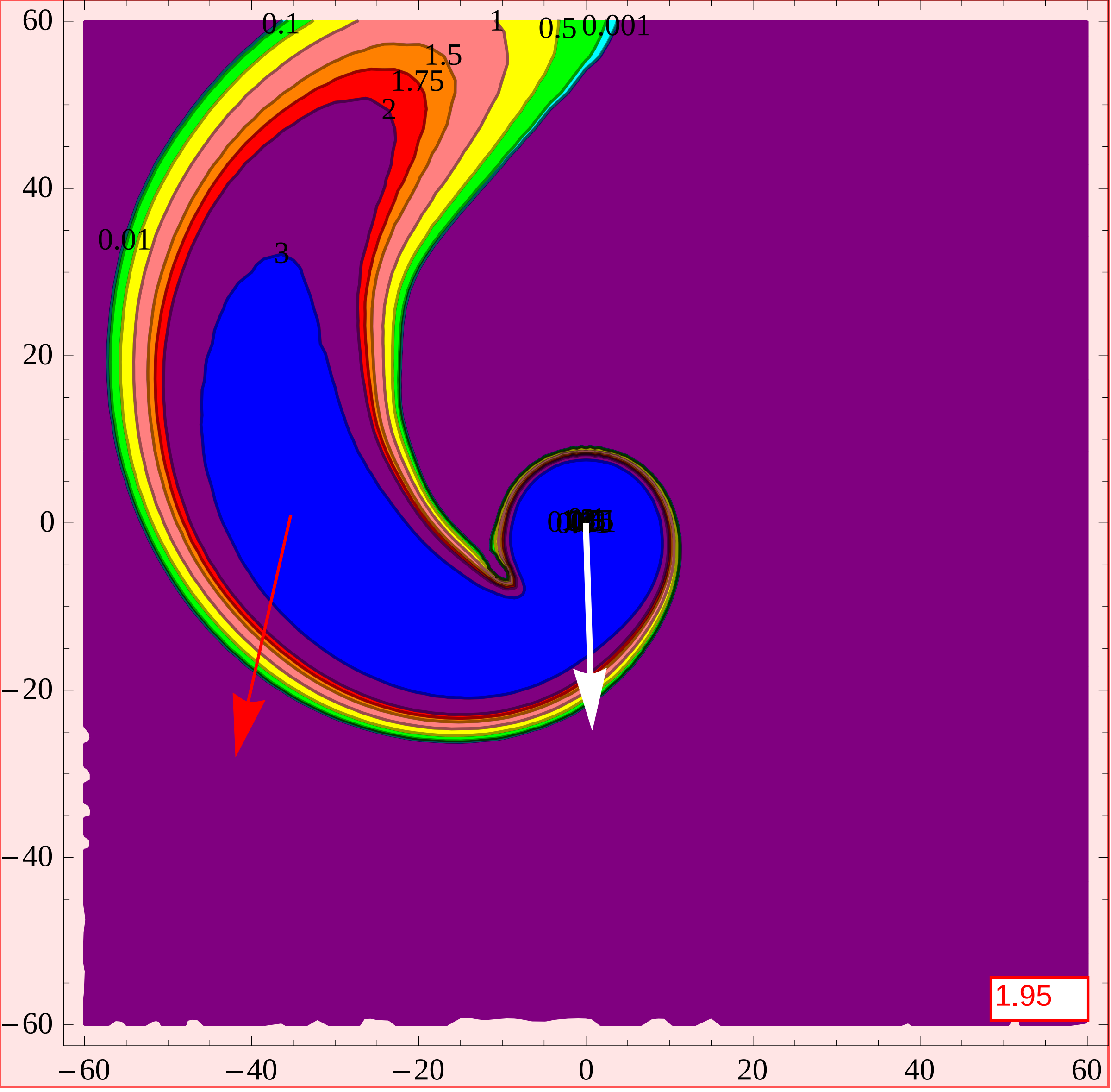

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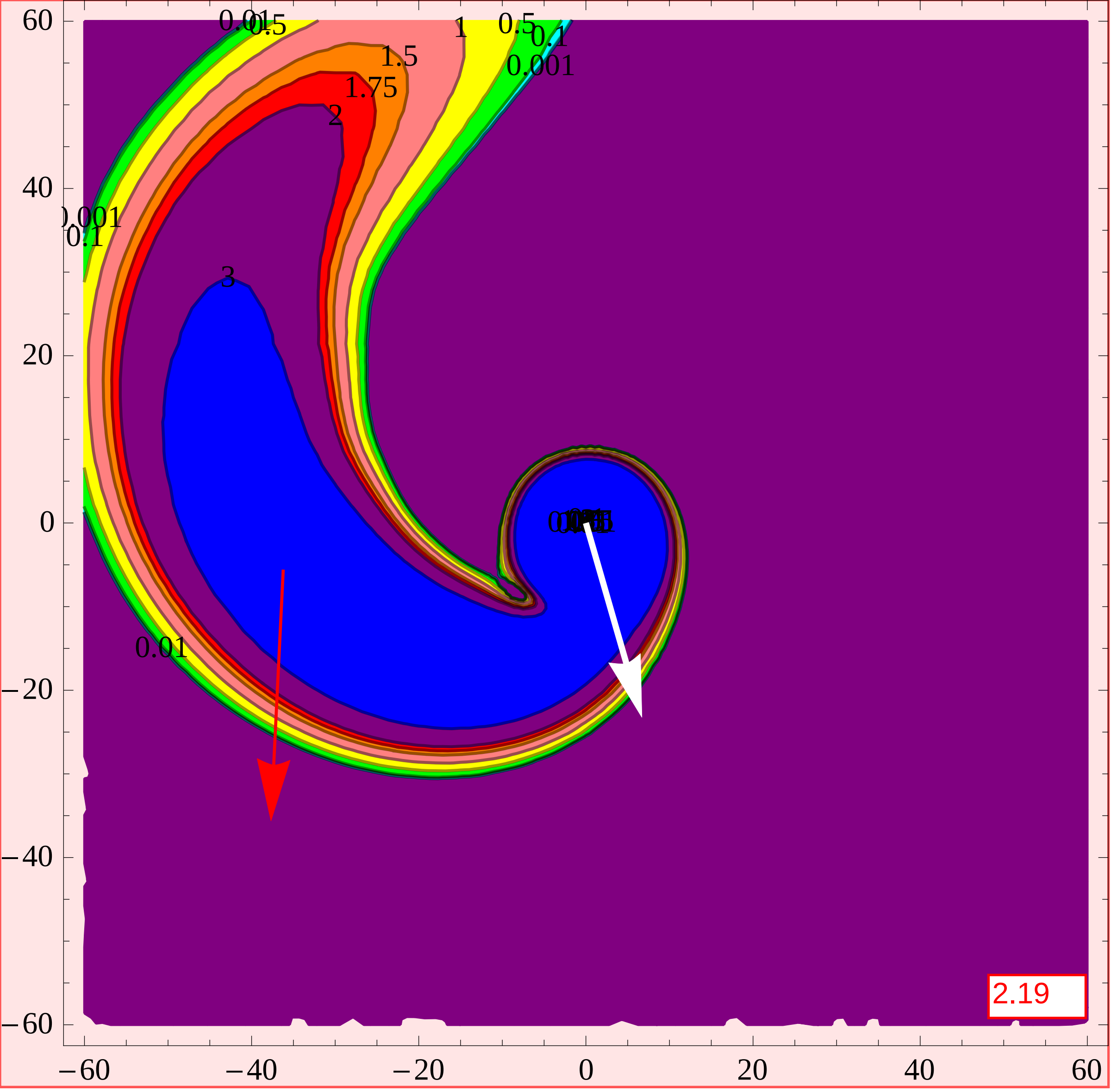

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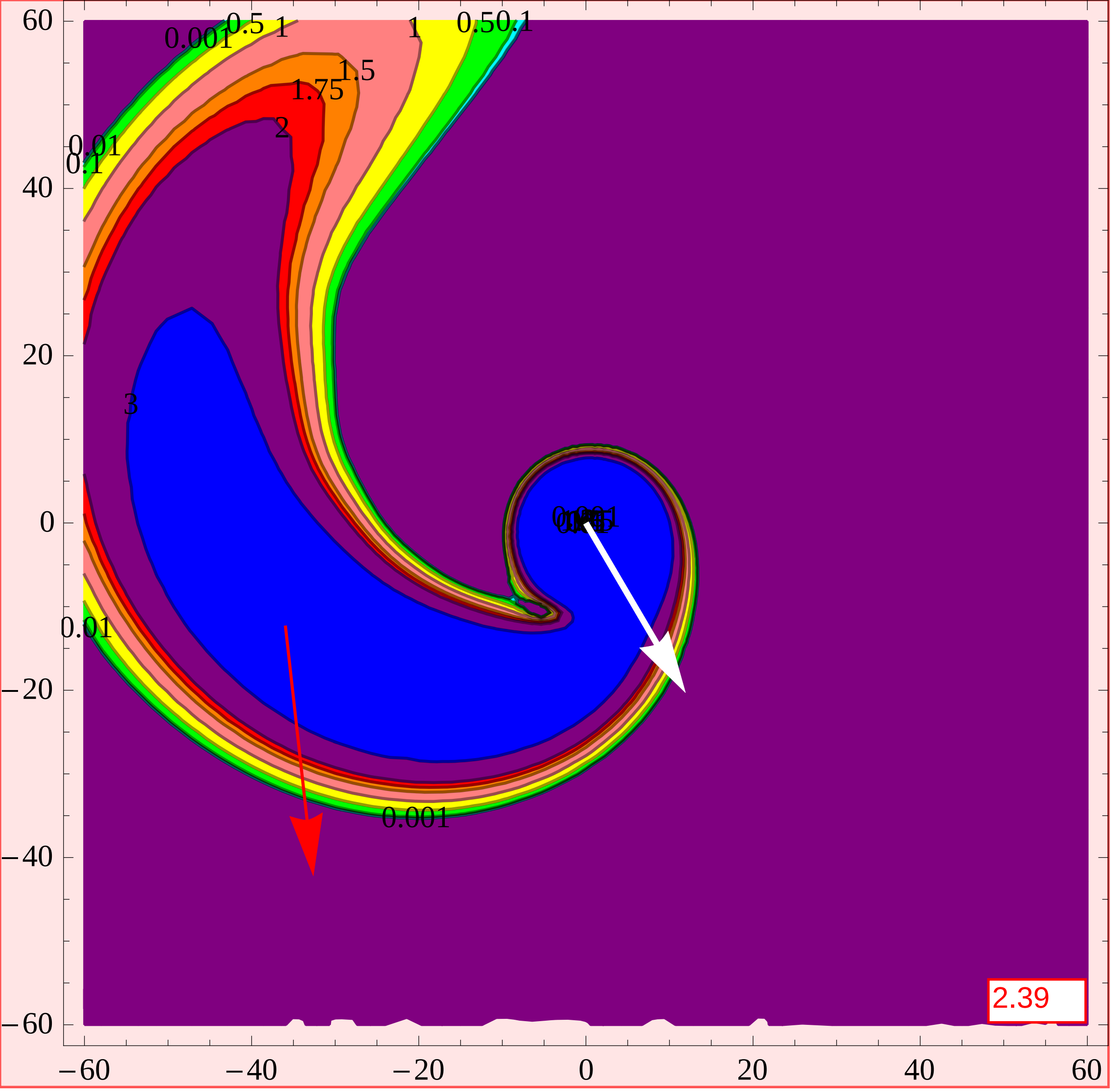
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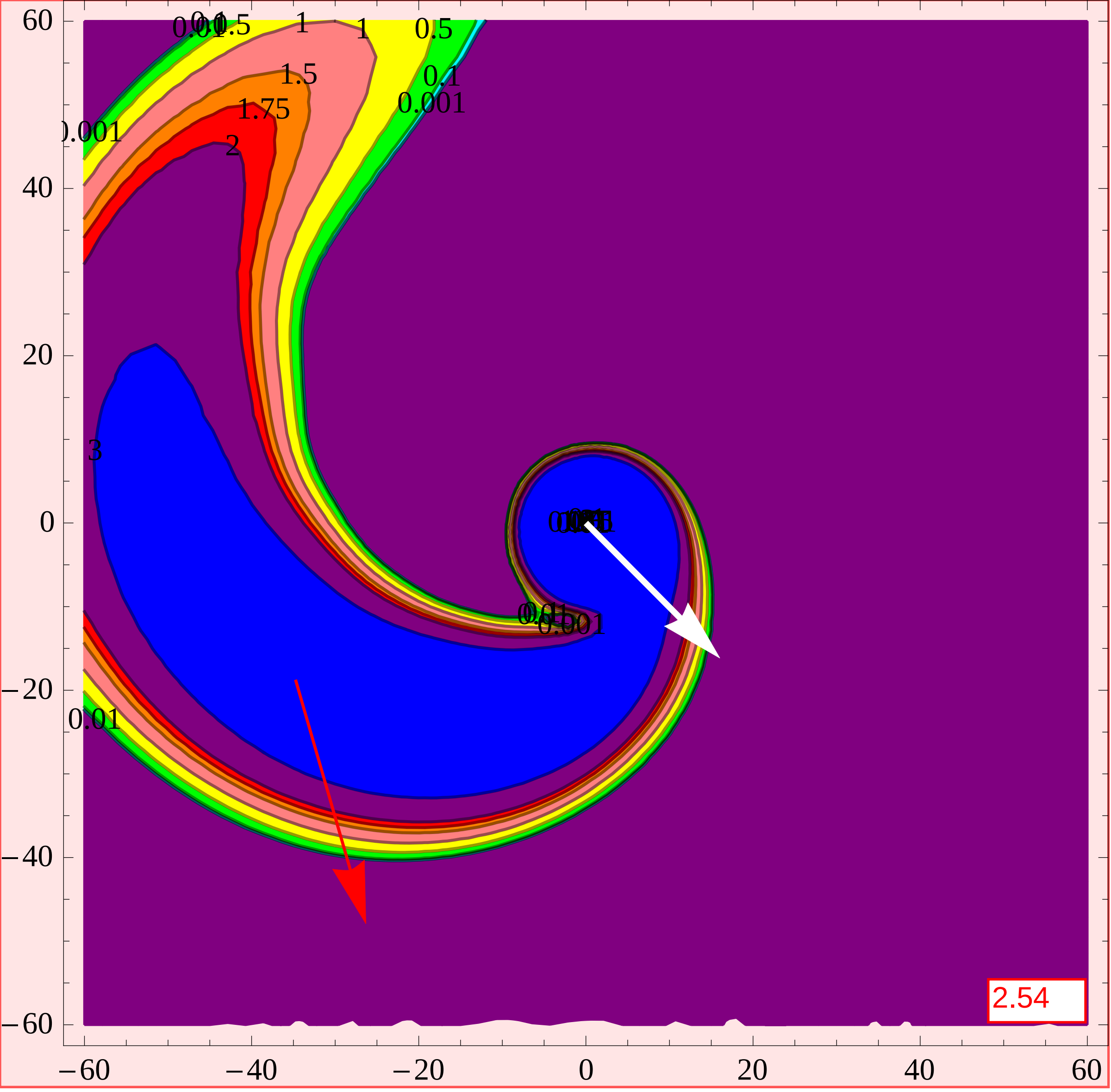

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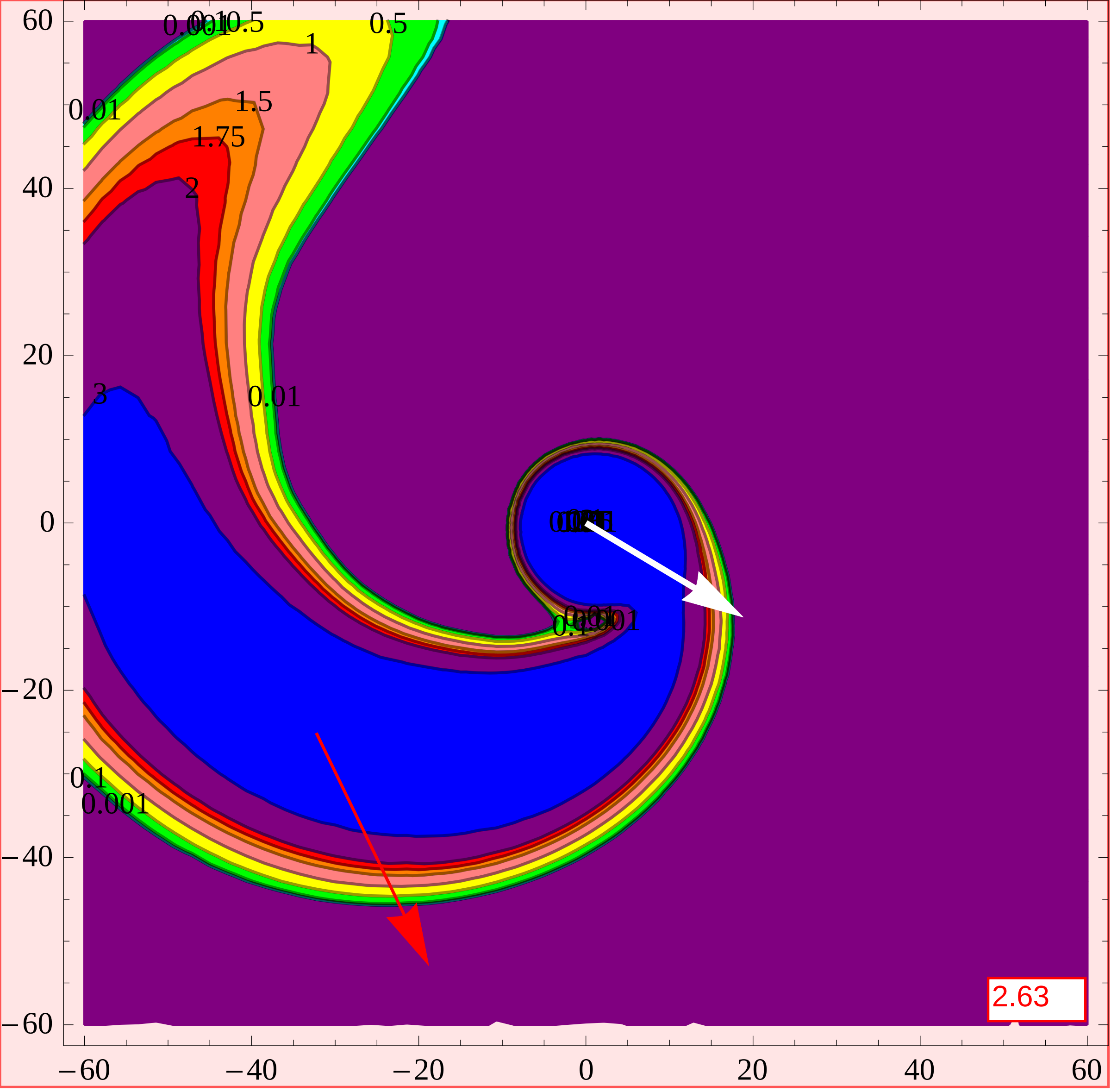

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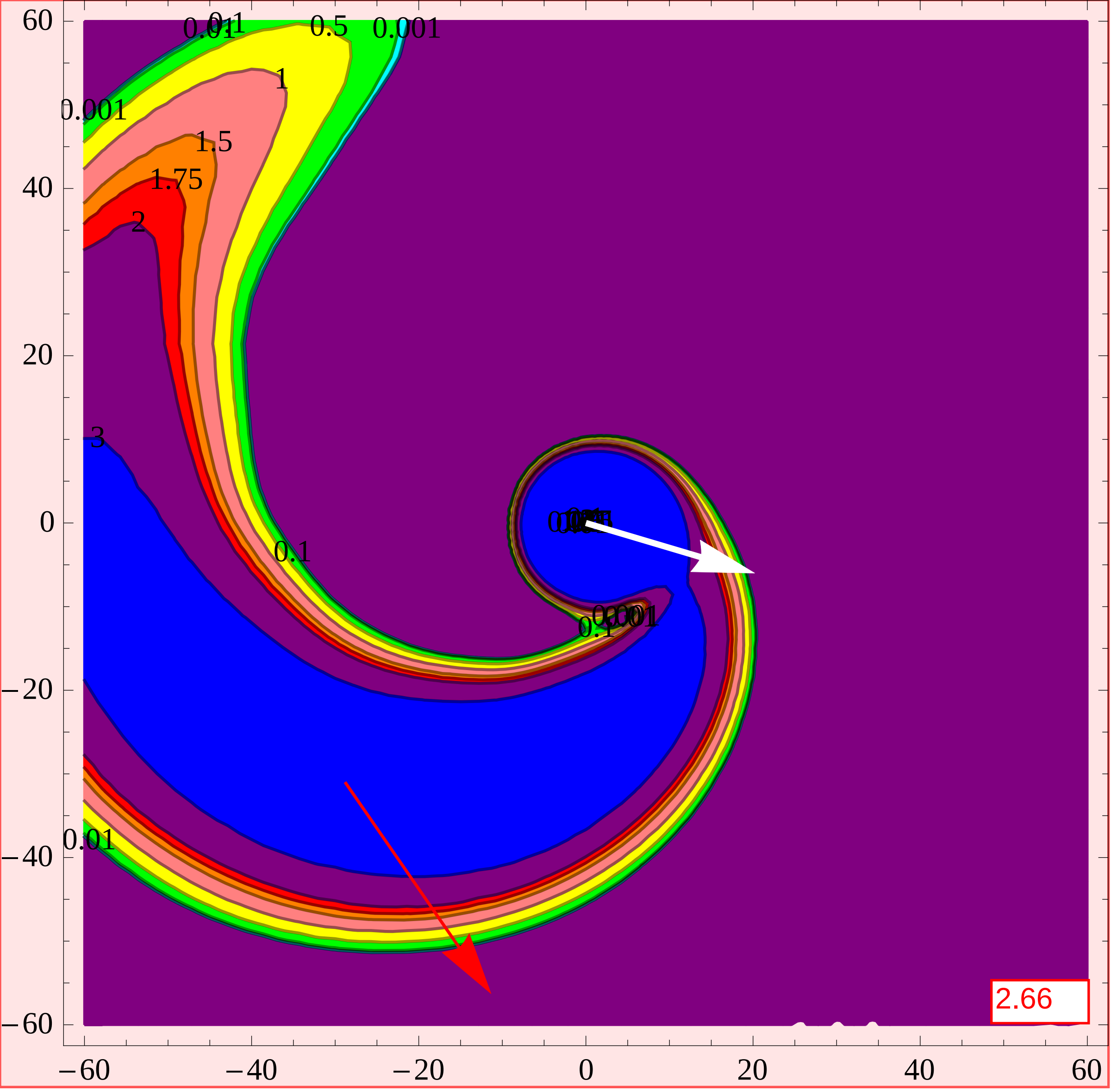

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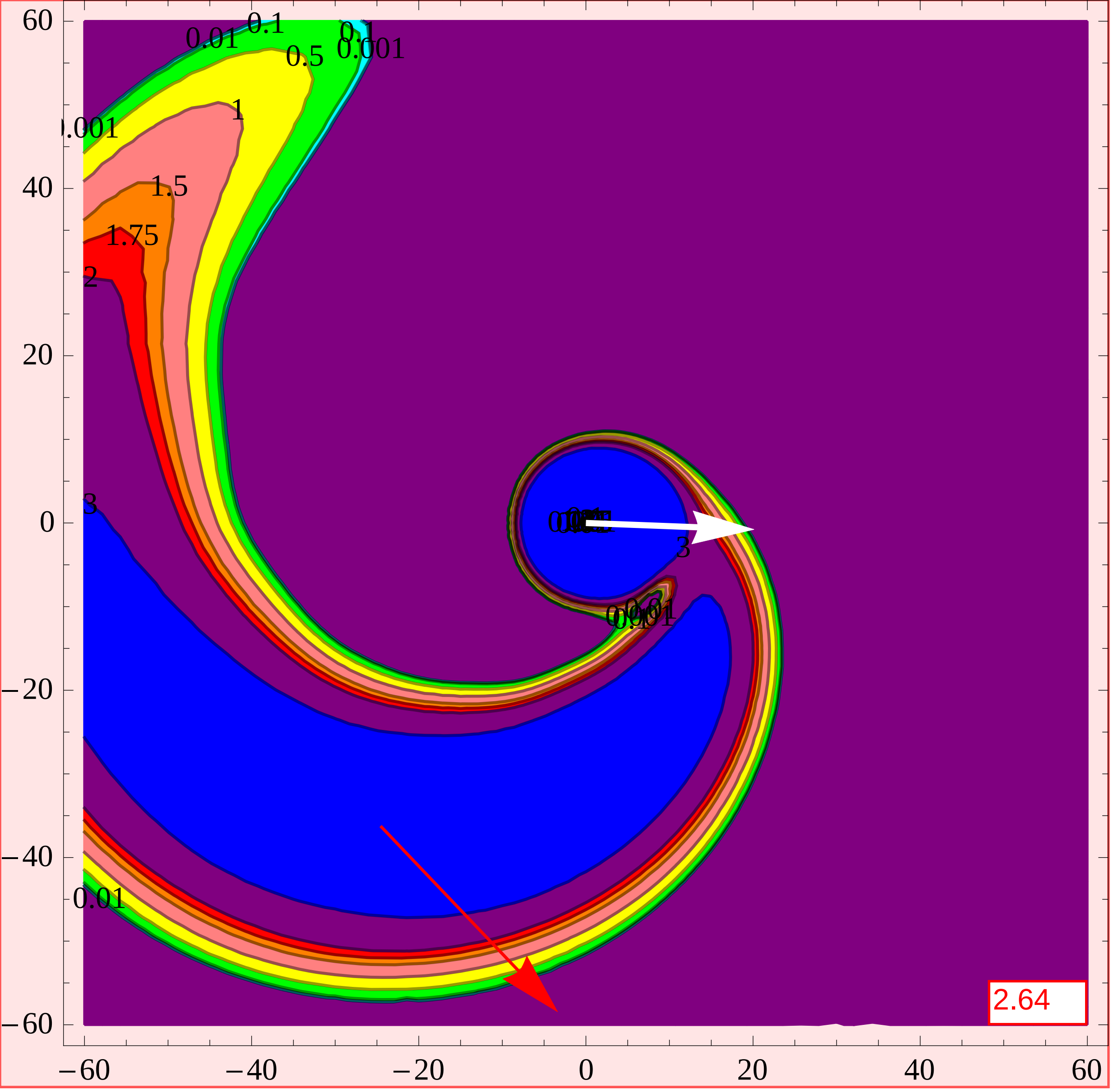

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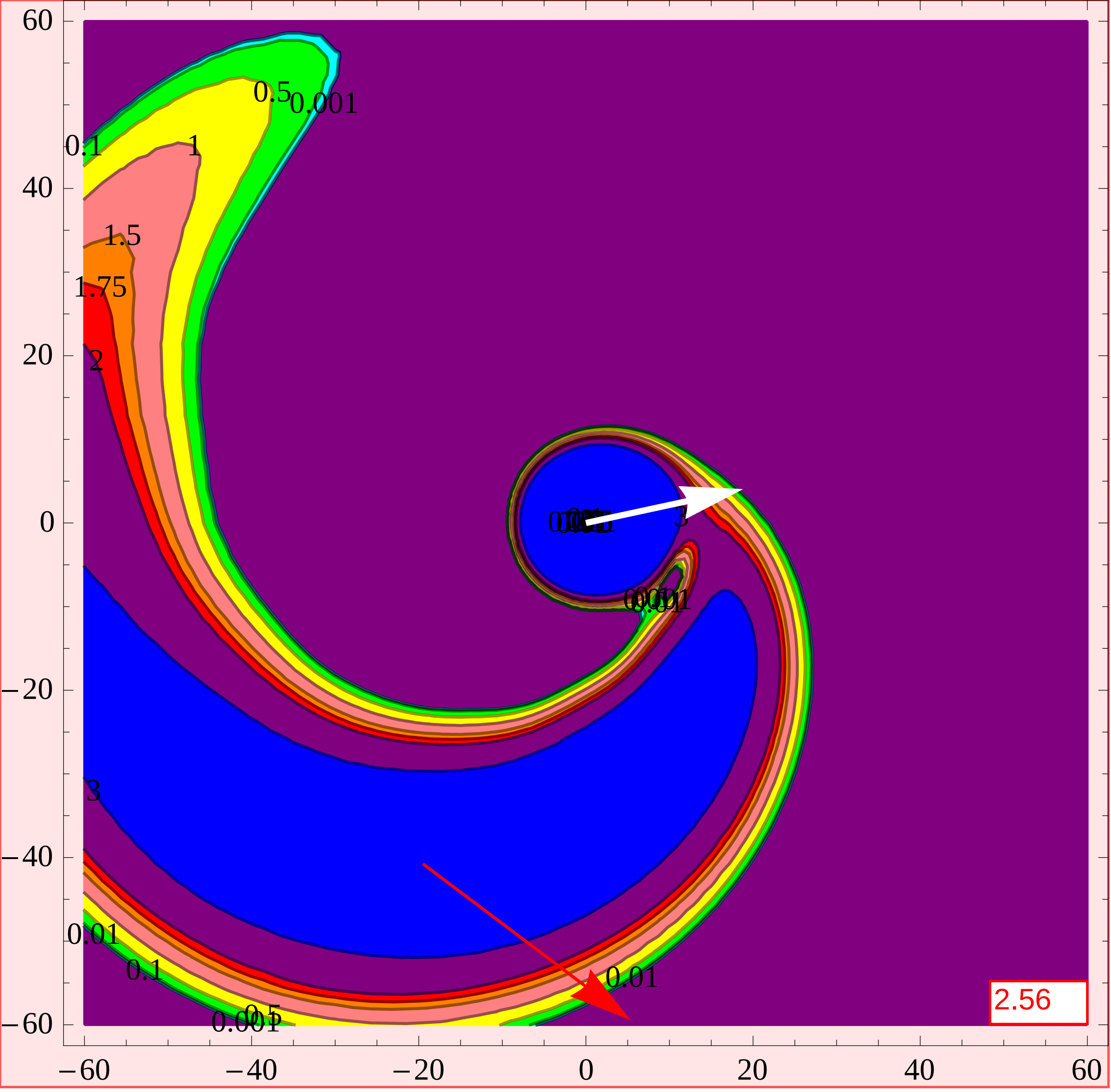

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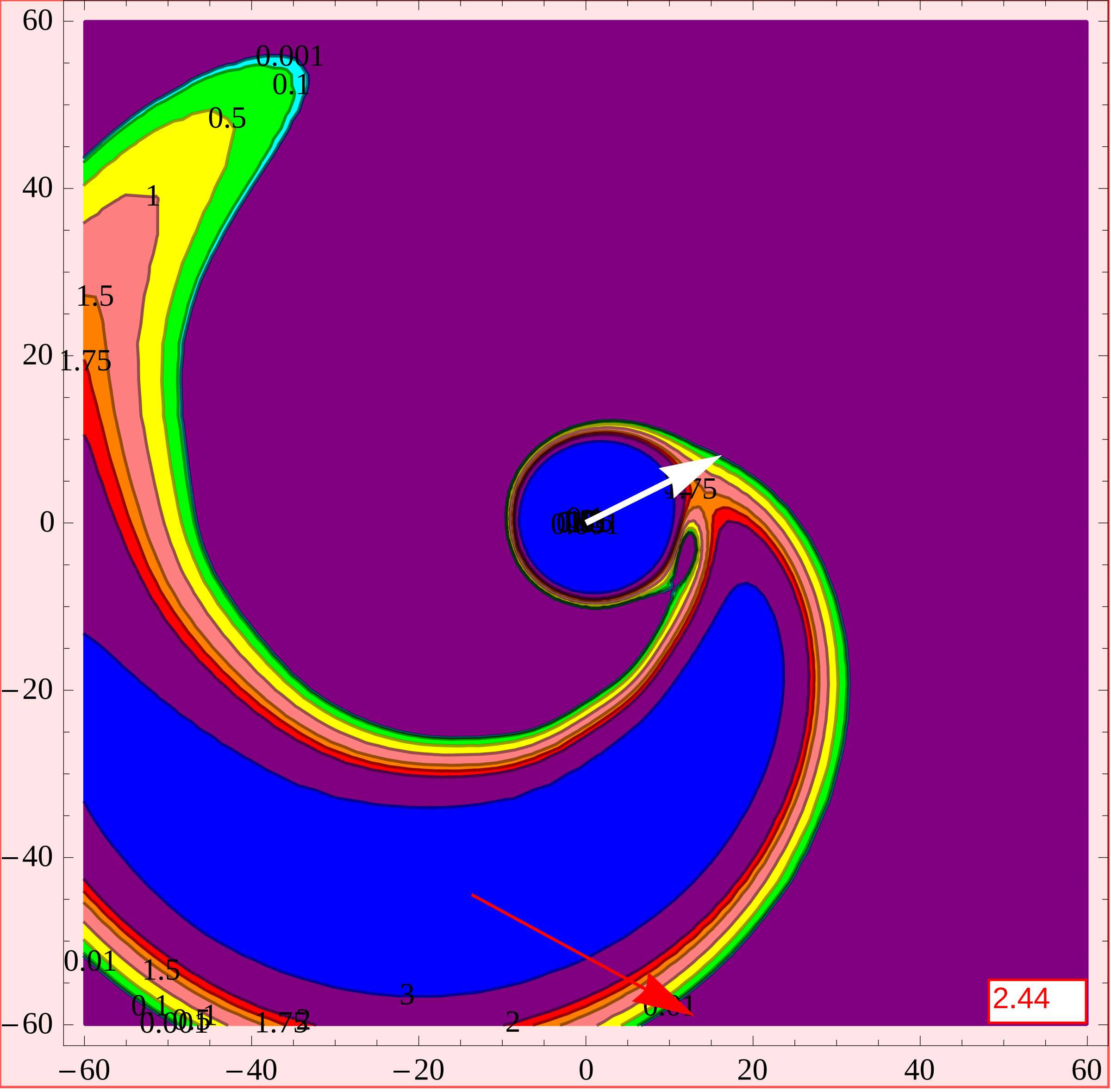

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1
1.5
1.75
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0.001
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1.75
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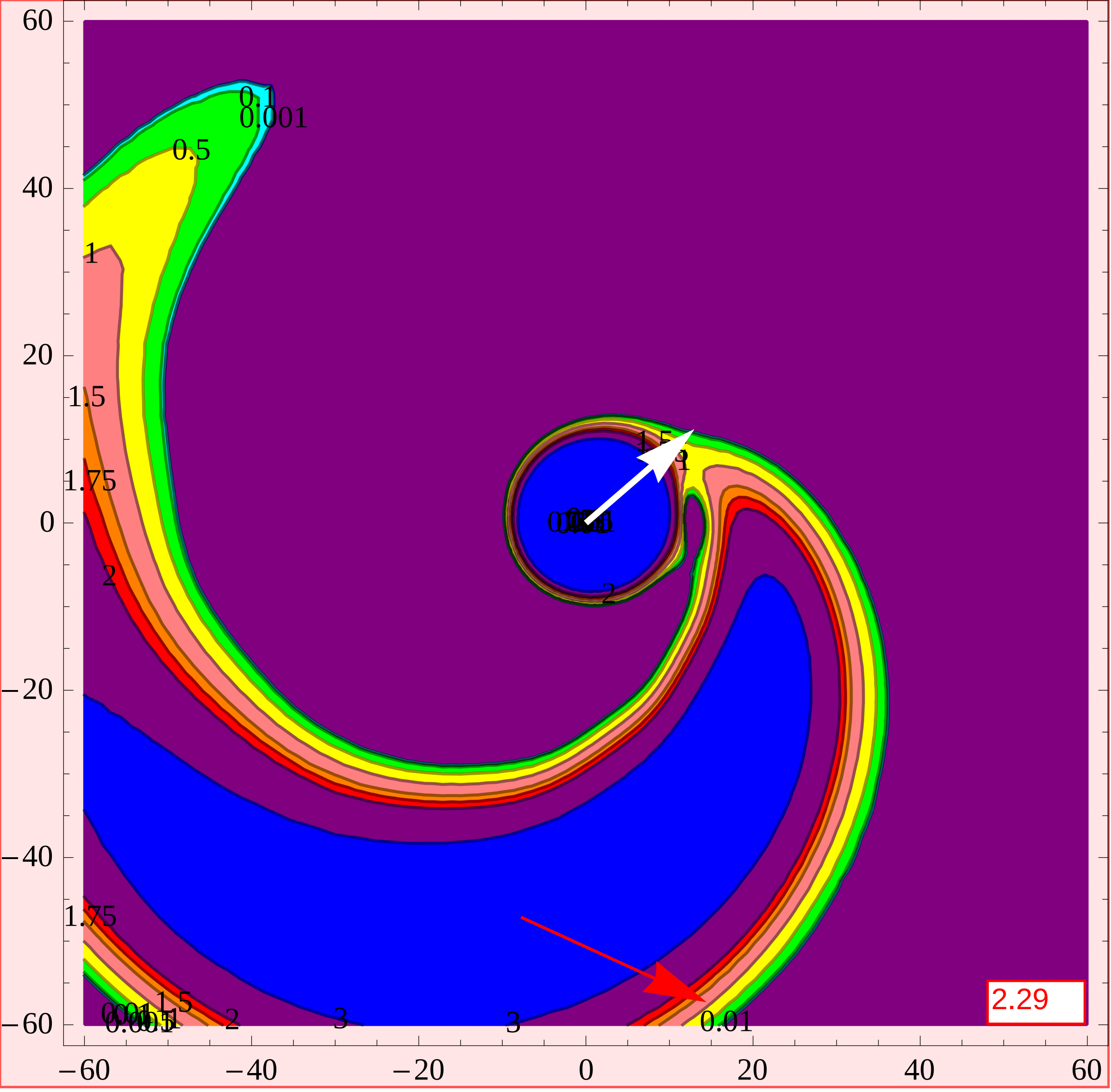
2.44



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0.5
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1.5
1.75
2
1.75
1.5
2
3
3
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−60
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−20
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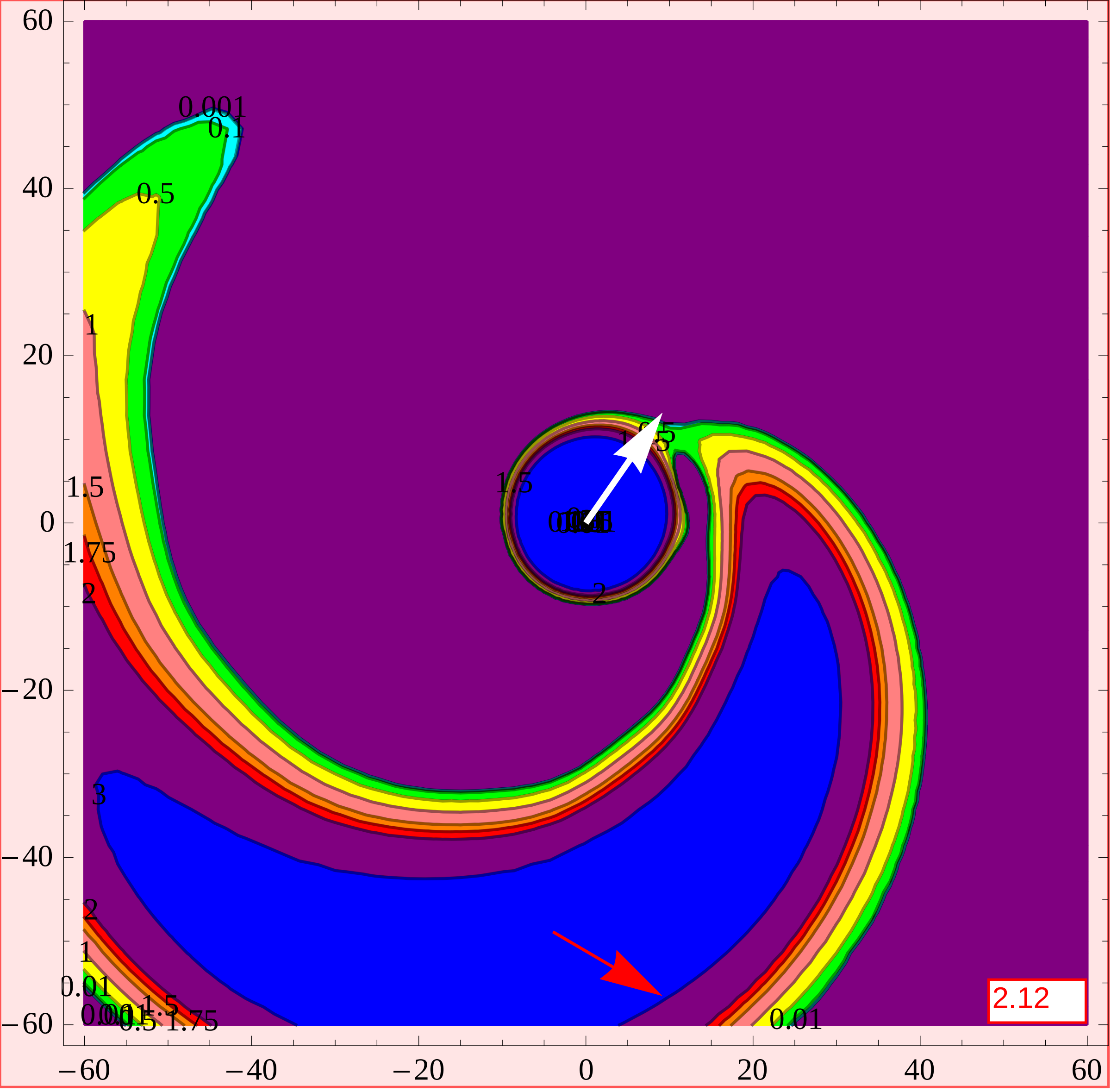

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0
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0.5
1
1.5
1.75
2
3
2
1
0.01
1.5
1.75
0.5
1.5
2
0.01
2.12