Multiphoton population transfer in a kicked Rydberg atom

adiabatic rapid passage by separatrix crossing

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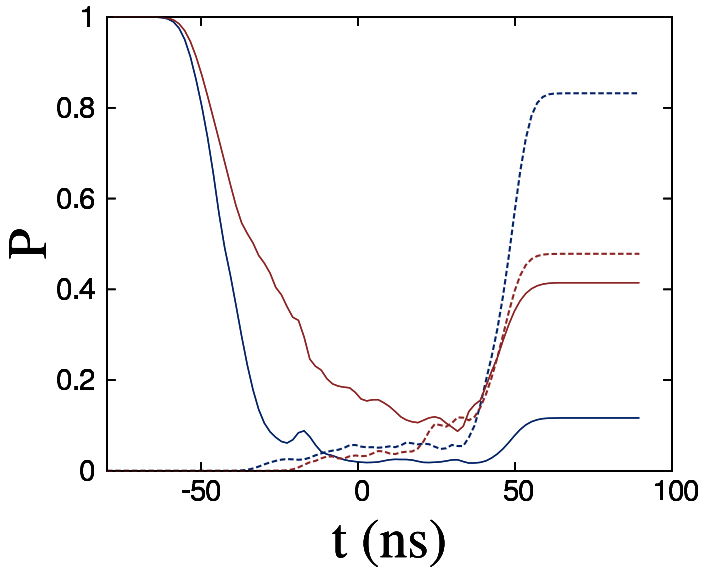
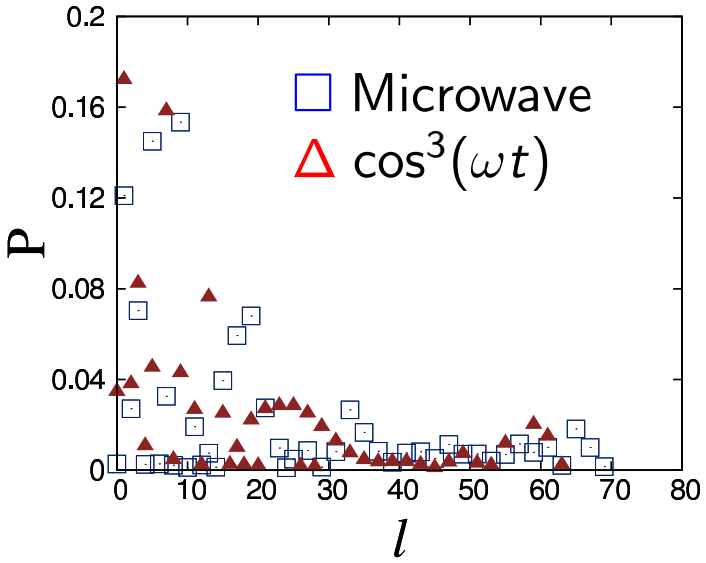
Previous work with microwaves (mARP)

* Earlier experimental work demonstrated 10γ transition in Li between states with $n = 72$ and $n{\sim}82$ using microwaves **(Maeda** *et al.***)**
* Classical and quantum simulations showed substantial population transfer in Li and H atoms **(Topcu** *et al.***)**
* Final ℓ-distributions suggested that many photons absorbed/emitted during transition
* Mechanism can be alternatively looked upon as a classical transition in phase space.
* Also relevant for transferring highly excited Rydberg wave packets $n > 300$ in phase space for wave packet engineering **(Rice group)**

72p $\longrightarrow$ $n = 80$ transition through 8γ resonance

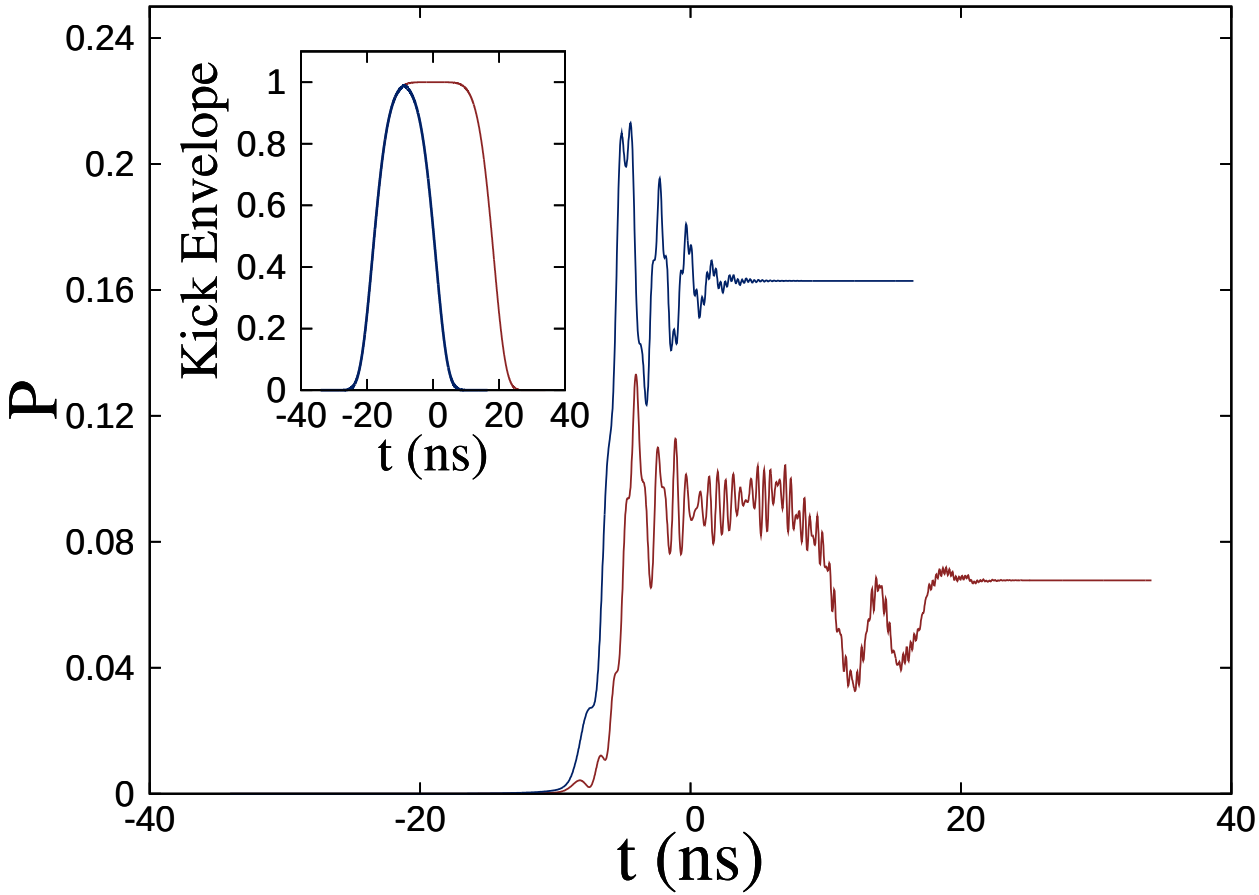




* For fixed peak field strength $F = 2.5$ V/cm
  - $\sim$83% Microwave driven
  - $\sim$44% $\cos^3(\omega t)$ driven
* Time scales and chirp
  - $\tau_R = 56.3$ ps for $n = 72$
  - $\tau_R = 77.2$ ps for $n = 80$
  - Pulse duration $\sim$97 ns at FWHM
  - Linear chirp range of $\pm\sim$1% of ω
* Only odd-ℓ are populated in both microwave and $\cos^3(\omega t)$ driving
* Population spread out to entire ℓ-range
* $\cos^5(\omega t)$, $\cos^9(\omega t)$, and higher order powers result in transfers at $\sim$10% level and below

40p $\longrightarrow$ $n = 46$ transition through 6γ resonance

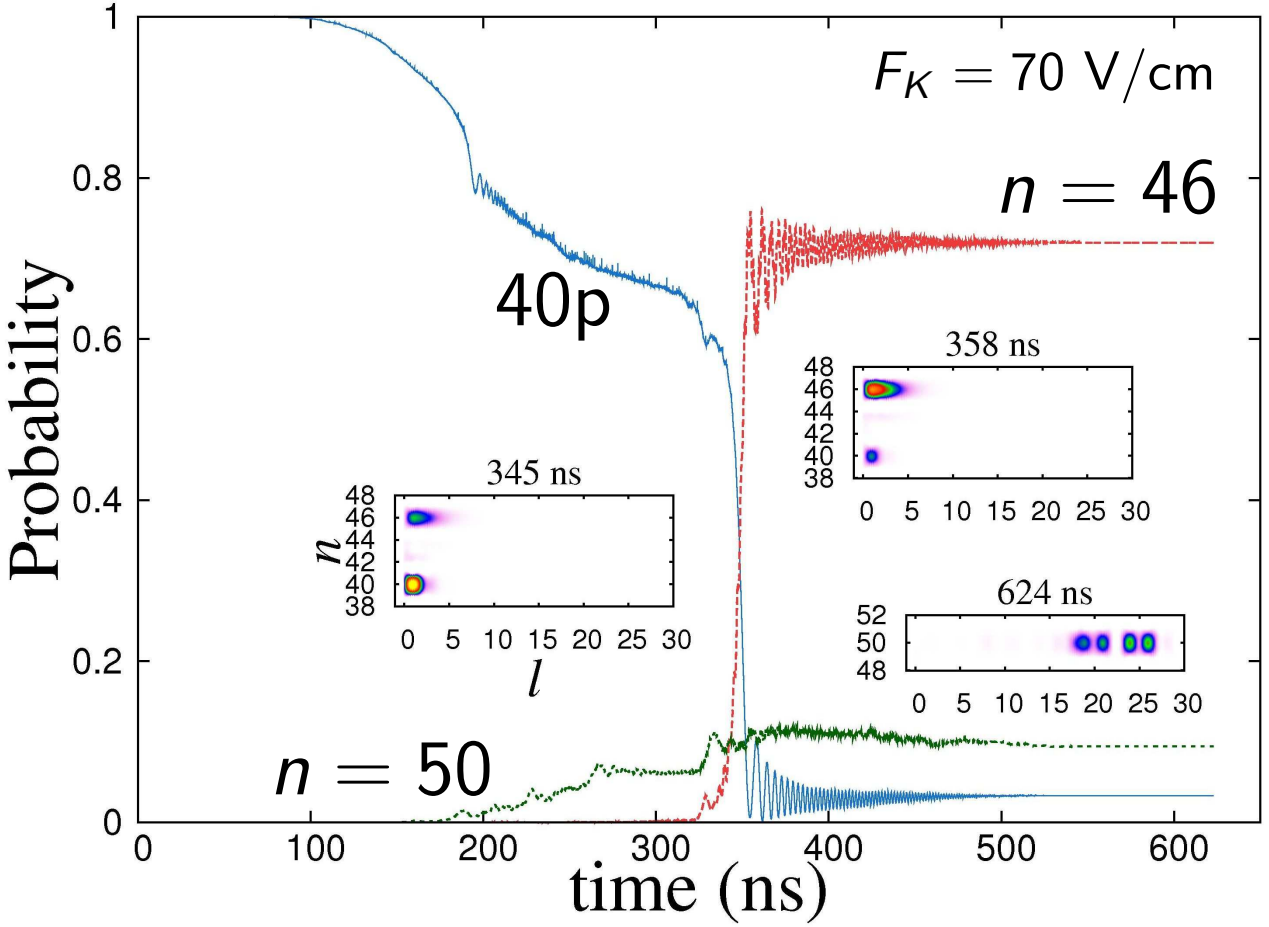
* Unidirectional kicks with flat-top carrier envelope
* Turn off kicks right after the kick strength peaks (shorter envelope)



* Shorter envelope let us kick harder
* 16% transfer into $n = 46$ compared to $\sim$8% with flat-top envelope

Shorter envelope in time

* **Unidirectional kicks** with shorter carrier envelope
* Pulse duration of ∼300 ns at FWHM



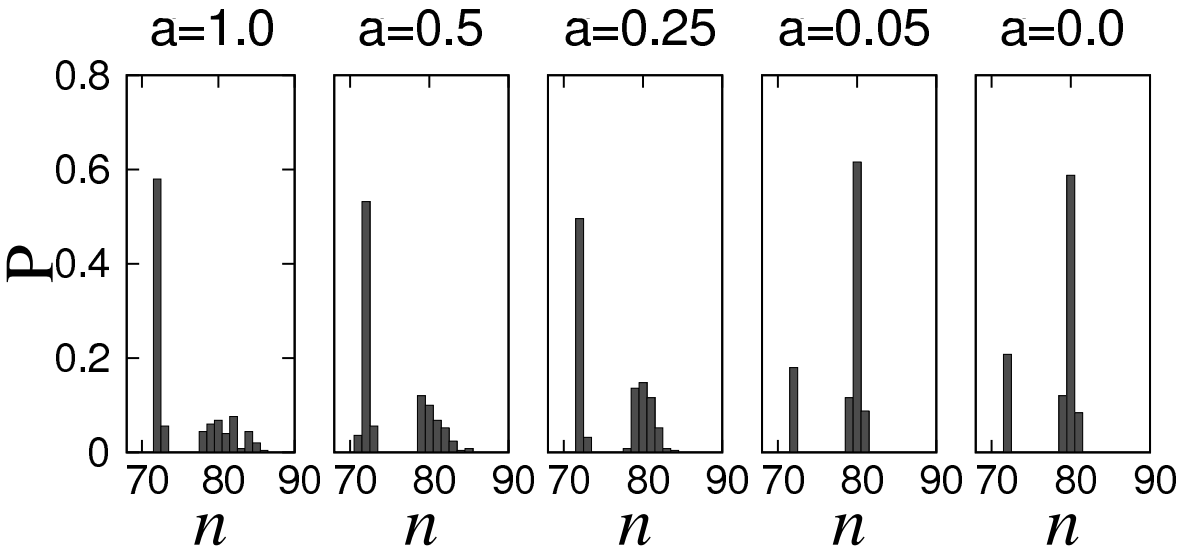
* ∼74% in $n = 46$
* $\ell < 5$
* Note that $\cos^3(\omega t)$ is $\frac{1}{4}\cos(3\omega t) + \frac{3}{4}\cos(\omega t)$
* **Accidental 3γ resonance**
  3ω differs from
  $\omega_{46\to49}$ by ∼26%
  $\omega_{46\to50}$ by ∼5%
  $\omega_{46\to51}$ by ∼16%

Classical calculations in three-dimensions

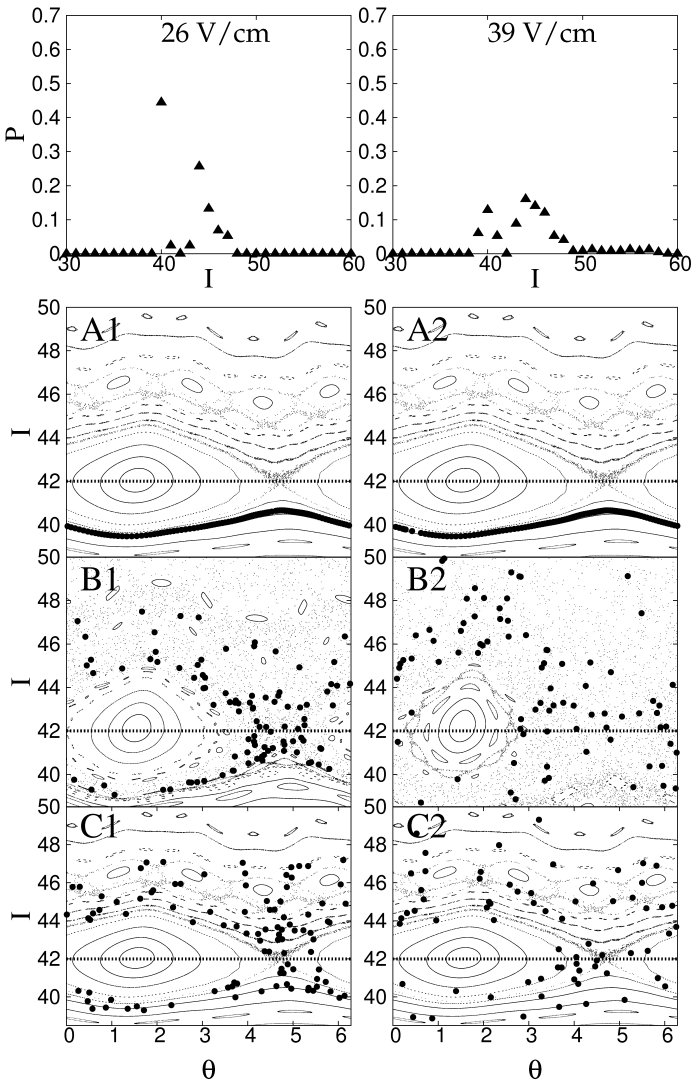
$n = 40 \rightarrow 46$ **via** 6γ **resonance condition**

* Population initially sits at $n = 40$ and then suddenly spreads over an n range when critical peak kick strength is reached for $\cos^3(\omega t)$ driving
* Progressing form $\cos^3(\omega t)$ to $\cos(\omega t)$, we go from $\sim$20% to $\sim$80% transfer without as much spreading in n

$$F(t) = F_K \exp[-(t/t_W)^6](\tfrac{a}{2}\cos(3\omega t) + \tfrac{4-a}{2}\cos(\omega t))$$



Classical calculations in one-dimension



$n = 40 \rightarrow 46$ **via** 6γ **resonance condition**

* Action-angle variables for hydrogen atom subject to $\sin^3(\omega t)$ driving
* $\sim$52% transfer for 26 V/cm into $43 < n < 47$
* $\sim$35% ionization for 39 V/cm
* **For 26 V/cm, edge of the regular surface is sticky**
* **By stronger driving, sticking can be overcome but now large chaotic sea yields to ionization**

Future directions

* Many driven systems actually fit this classical description, such as kicked/driven rotor, Morse potential, etc.
* We have observed **complete population transfer** between the vibrational levels $\nu = 0 \rightarrow 4$ of a diatomic molecule by chirping through 4γ resonance using IR pulses
* Effect of frequency profile with energy
  For Rydberg atoms and molecules: **frequency increases with energy**
  For kicked rotor: **frequency decreases with energy**
  What if **frequency increases and then decreases** with energy???

Thank you.