

# Cooling with coherent pulse trains

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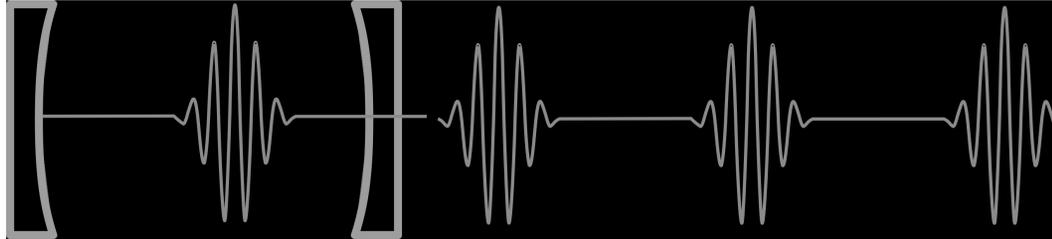


KITP program “Fundamental Science and Applications of Ultra-cold Polar Molecules” 2013

# Outline

- Frequency comb (FC) introduction
- Doppler cooling of two-level system
- See-saw protocol
- Stimulated cooling on multiple ro-vibrational transitions

# Time picture

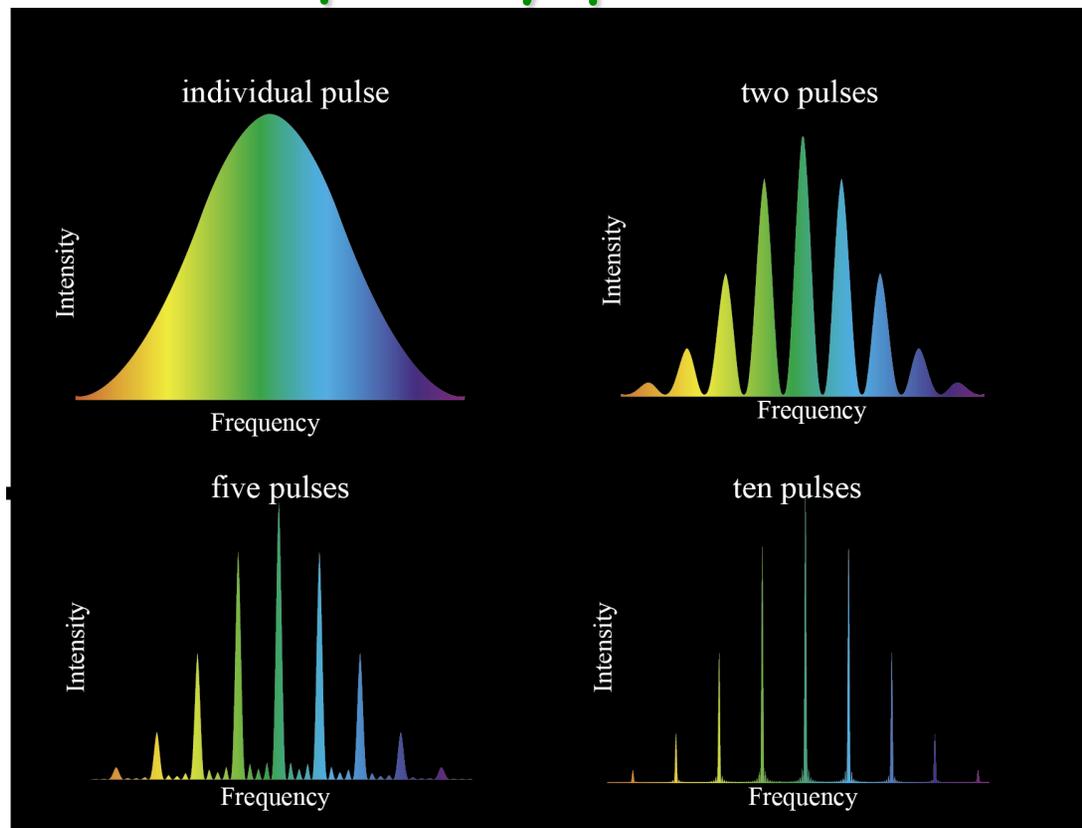


$T$ =repetition time = round-trip time inside the cavity

$$T = 1 - 10 \text{ ns} \quad \tau_p \sim 100 \text{ fs}$$

$$\mathbf{E}(t) = \hat{\mathbf{e}} E_p \sum_m \cos(\omega_c t - m\phi_{CEO}) g(t - mT)$$

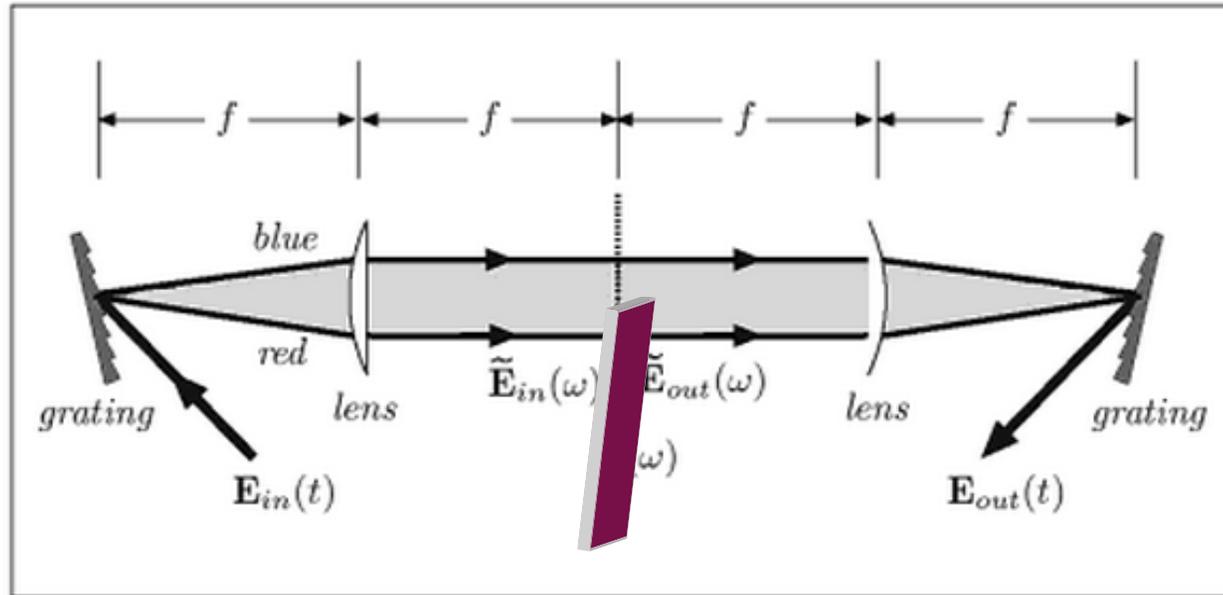
# Frequency picture



Infinite number of pulses = Dirac comb

$$\omega_n = \omega_c + \frac{2\pi}{T} n + \frac{\phi}{T}$$

# Controlling the train



- ❖ Both the amplitude and phase of teeth could be controlled
- ❖ Individual teeth could be controlled on the nanosecond time scale
- ❖ Simple example: mask

# Typical parameters

Repetition time/pulse duration:

$$T = 1 - 10 \text{ ns} \quad \tau_p \sim 100 \text{ fs}$$

Carrier frequencies:

infrared  $\rightarrow$  ultraviolet

Power:

100 mW (typical)  $\rightarrow$  10 W (demonstrated)  $\rightarrow$  10 kW (future)

$$\text{Intensity}(\propto E^2) = \frac{\text{power}}{\text{focus area}}$$

# Does a single tooth have enough power for cooling?

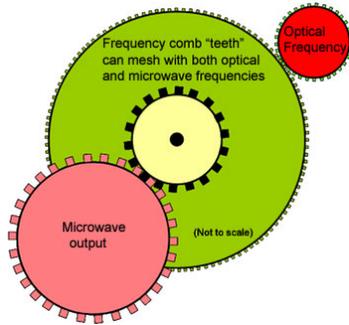
The relevant parameter is the saturation intensity.

Typical value (sodium atom) of saturation intensity is  $6.4 \text{ mW/cm}^2$ .

Typical FC parameters (100 fs pulse,  $T = 1 \text{ ns}$ ) and average power 1 W translate into the power per tooth of 0.1 mW.

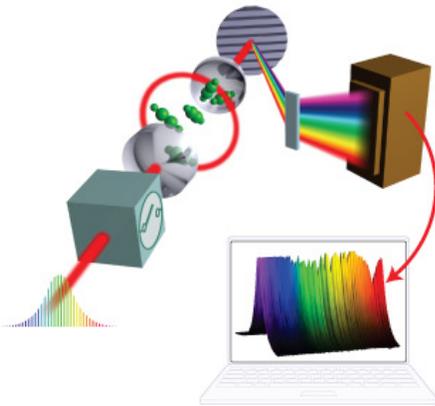
=> Saturation intensity can be attained by focusing the laser output to a spot of 1.4 mm diameter.

# Applications of FCs



## Atomic clocks

- Comparison of optical and microwave (Cs) clocks
- Direct link to electronic counters



## Molecular fingerprinting

- Spectroscopy done in parallel

# Initial motivation for laser cooling with FC

- ❑ Optical lattice clocks (ultrastable clocks)
    - 7 lasers required
    - FC is an essential element
  - ❑ ESA is evaluating the feasibility of sending lattice clocks (Sr and Yb) into space
    - Space Optical Clocks (SOC) mission
  - ❑ Can we reduce the number of required lasers by using FC for cooling/slowing atoms?
- 

Can we use quantum control ideas combined with FC to cool molecules?

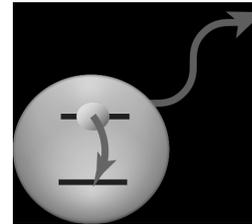
# FC summary

- ❖ Coherent pulse train in time-domain = comb in the frequency space
- ❖ Individual teeth phase and intensity and the CEO phase can be controlled
- ❖ FC + quantum control = new applications

**(Complexity)<sup>2</sup>**

# Doppler cooling vs stimulated cooling

Doppler

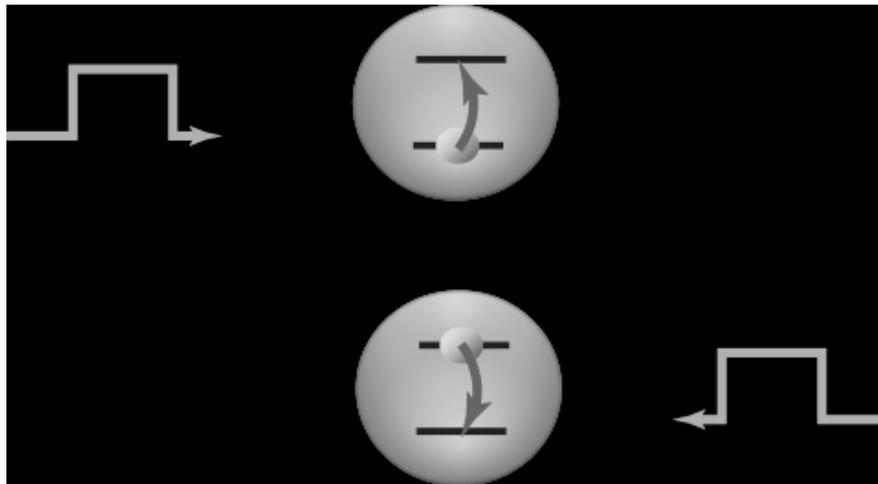


$$\Delta p = \hbar k_c$$

Need to wait for spontaneous emission event

Stimulated

$\pi$ -pulse



pulse

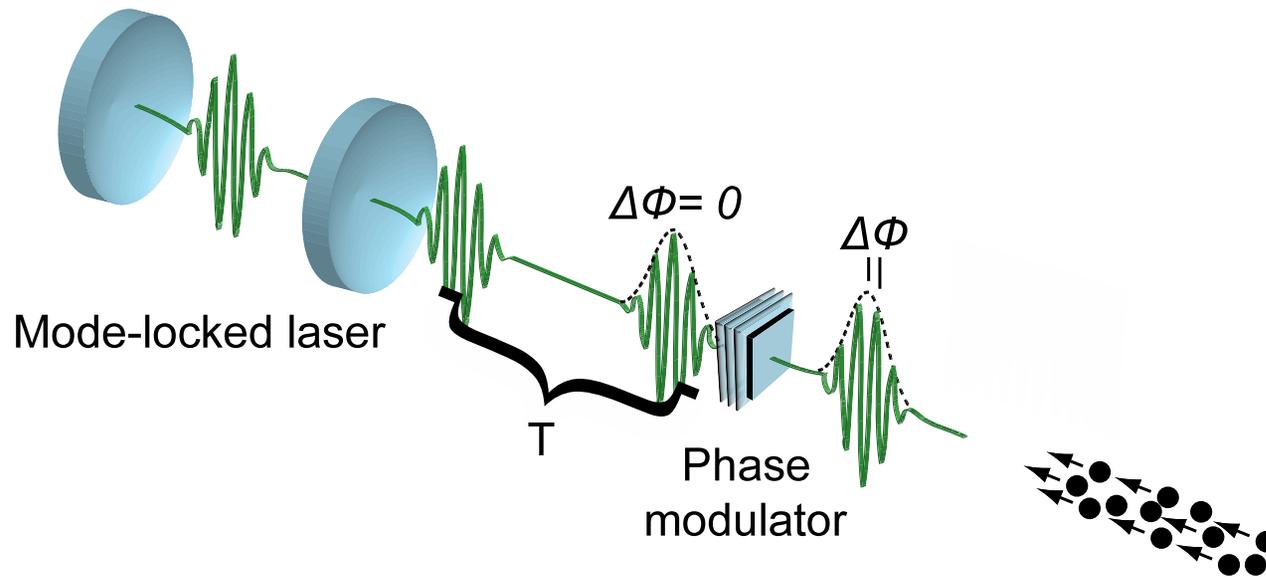
$$\Delta p = 2\hbar k_c$$

Can be very fast. Could work for long-lived transitions. Is it cooling?

# Doppler cooling of two-level system with coherent pulse trains

E. Ilinova, M. Ahmad, and A. Derevianko, PRA **84**, 033421 (2011)

# Setup



# Setup

Train E-field

$$\mathbf{E}(t) = \hat{\varepsilon} E_p \sum_m \cos(\omega_c t - \phi_m) g(t - mT)$$

Rabi frequency

$$\Omega_{ge}(z, t) = \Omega_p \sum_{m=0}^{N-1} g(t + z/c - mT) e^{i\phi_m}$$

Peak value

$$\Omega_p = \frac{E_p}{\hbar} \langle e | \mathbf{D} \cdot \hat{\varepsilon} | g \rangle$$

# Interference (perturbative solution)

$$|\Psi\rangle = c_e(t) e^{-iE_e t} |e\rangle + c_g(t) e^{-iE_g t} |g\rangle$$

$$c_e(t) = c_e^1 + c_e^2 + \dots + c_e^N \qquad c_g(t) \approx 1$$

$$c_e^1 = \frac{1}{2i} \Omega \int_{-\infty}^T g(t) e^{i\delta t} dt \approx \frac{1}{2i} \Omega \int_{-\infty}^{\infty} g(t) e^{i\delta t} dt = \frac{1}{2i} \sqrt{2\pi} \Omega \tilde{g}(\delta)$$

$$c_e^2 = \frac{1}{2i} \Omega \int_{T/2}^{3T/2} g(t-T) e^{i\delta t} dt = e^{i(\phi+\delta T)} c_e^1$$

$$c_e^k = \left[ e^{i(\phi+\delta T)} \right]^{k-1} c_e^{k-1}$$

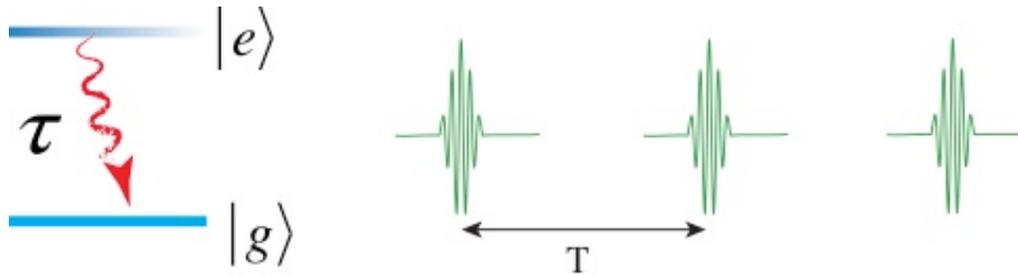
$$c_e(t) = \sum \text{pulses} c_e^k$$

← Interference b/w amplitudes

$$c_e^N = c_e^1 \left( e^{i\frac{1}{2}(\phi+\delta T)} \right) \frac{\sin\left(\frac{N}{2}(\phi+\delta T)\right)}{\sin\left(\frac{1}{2}(\phi+\delta T)\right)}$$

← Comb structure

# Essential parameters



Pulse area

$$\theta = \Omega_p \int_{-\infty}^{\infty} g(t) dt$$

$$\theta = \pi/2: \quad |g\rangle \rightarrow \frac{1}{\sqrt{2}}(|g\rangle + |e\rangle)$$

$$\theta = \pi: \quad |g\rangle \rightarrow |e\rangle$$

Persistence of memory

$$\gamma T = \frac{T}{\tau}$$

$$\tau \gg T$$

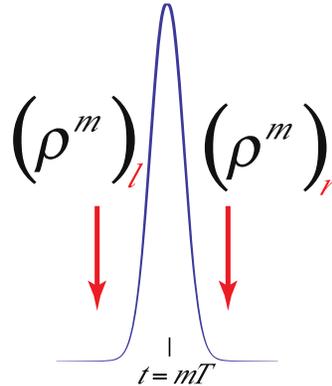
Interference of probability amplitudes  
due to subsequent pulses

Doppler-shifted phase

$$\eta = (\omega - \omega_0 + \mathbf{k}_c \cdot \mathbf{v})T - \phi$$

# Exact solution for ultrashort pulses

Across  
the  $m^{\text{th}}$  pulse



$$(\rho^m)_r = e^{i\theta/2 \sigma_m} (\rho^m)_l e^{-i\theta/2 \sigma_m}$$

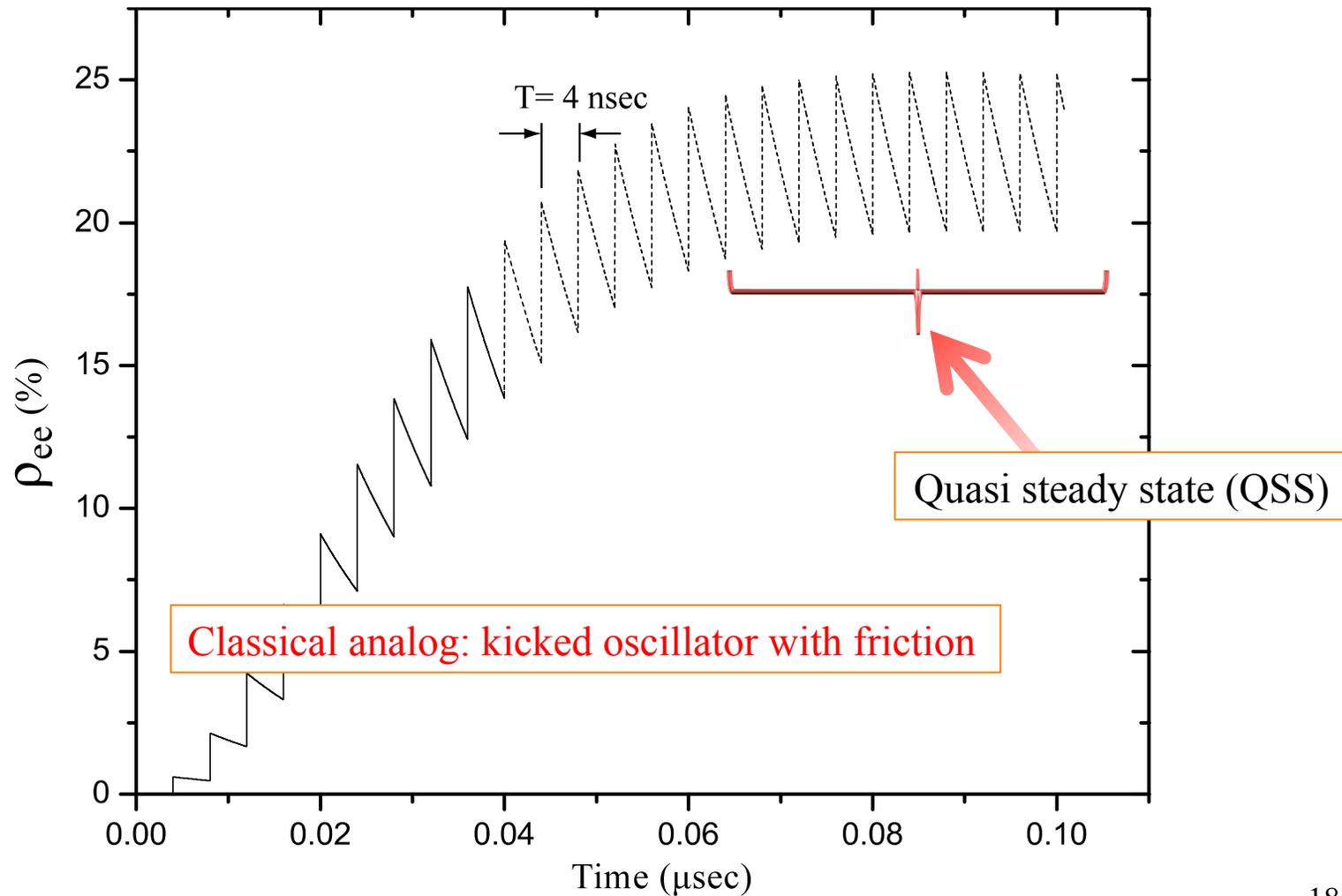
$$\sigma_m = \cos\phi \sigma_x - \sin\phi \sigma_y$$

Between the pulses

$$\rho_{eg}(t) = (\rho_{eg}^m)_r \exp\left[-\left(\frac{\gamma}{2} - i\delta_{\text{eff}}\right)(t - mT)\right]$$

$$\rho_{ee}(t) = (\rho_{ee}^m)_r \exp[-\gamma(t - mT)].$$

# Population of the excited state



# Mechanical effects

Fractional momentum kick per pulse in QSS

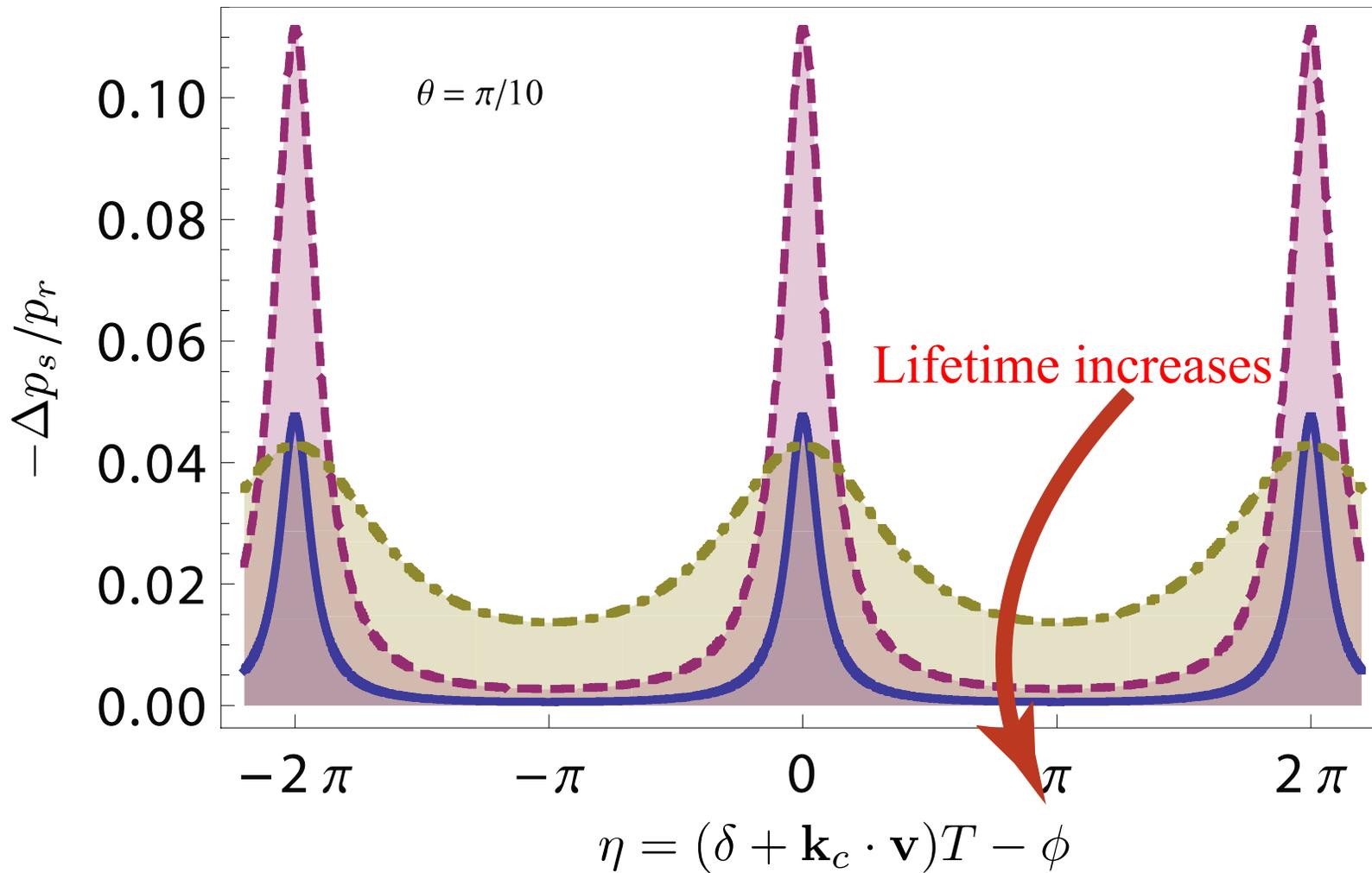
$$-\frac{\Delta p_s}{p_r} = \left( \rho_{ee}^s \right)_r \times \left( 1 - e^{-\gamma T} \right)$$

Post-pulse QSS excited-state population

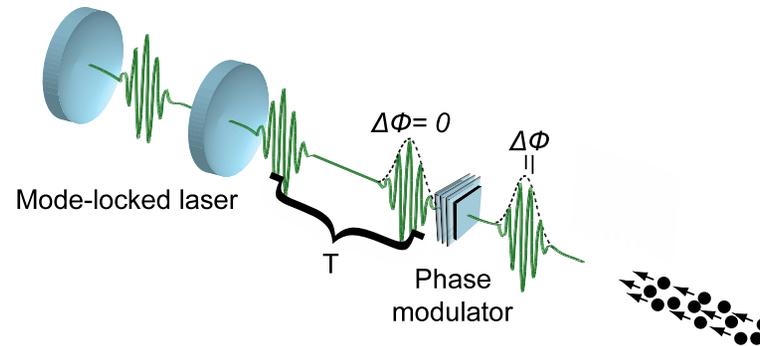
$$-\frac{\Delta p_s}{p_r} = \frac{\sin^2(\theta/2) \sinh(\gamma T/2)}{\cosh(\gamma T/2) - \cos^2(\theta/2) \cos \eta}$$

$$\eta = (\omega - \omega_0 + \mathbf{k}_c \cdot \mathbf{v})T - \phi$$

# Fractional momentum kick



# Tuning the CEO phase

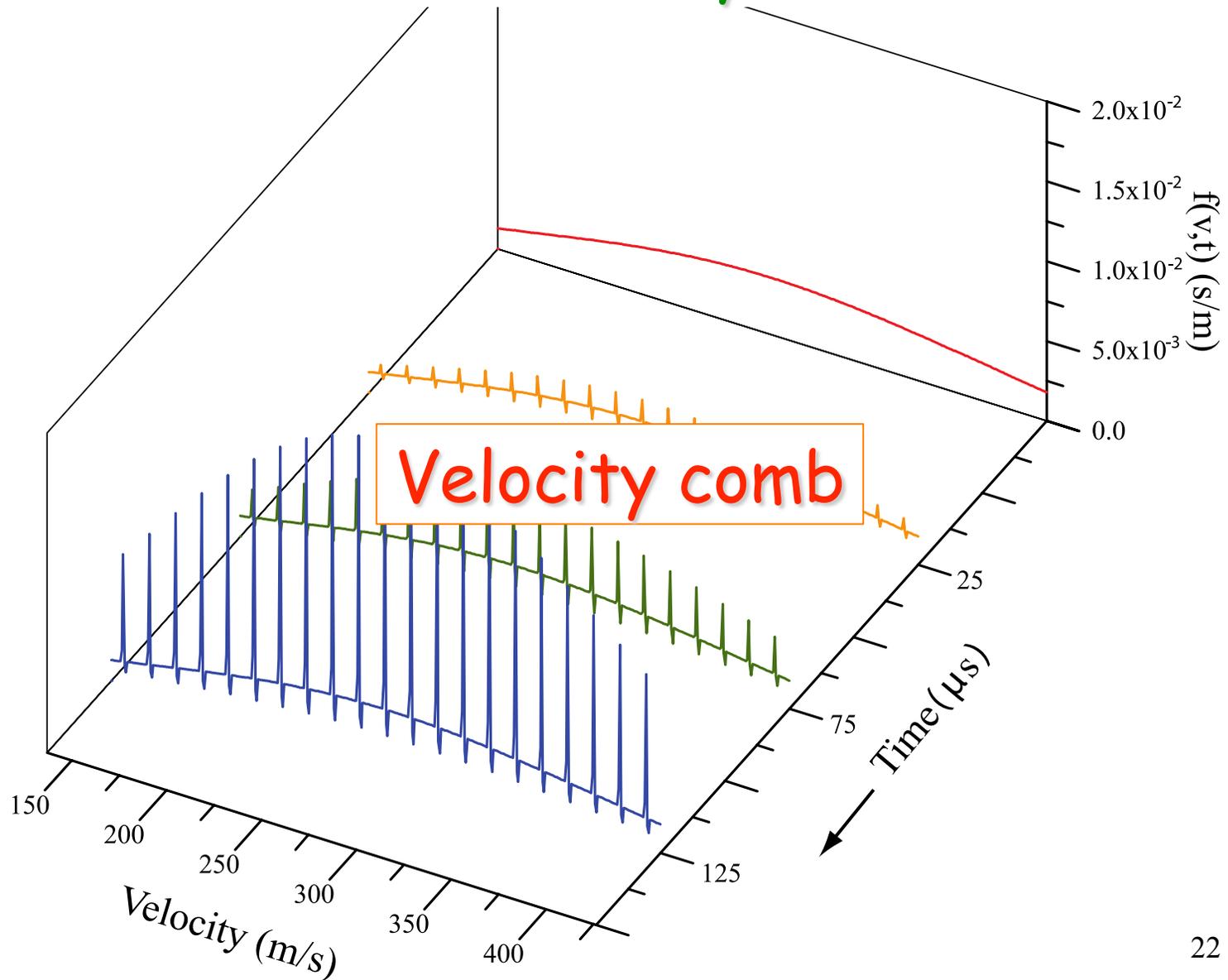


$$\eta = (\omega - \omega_0 + \mathbf{k}_c \cdot \mathbf{v})T - \phi$$

Deceleration  $\Rightarrow$  velocity decreases  $\Rightarrow$  the phase becomes smaller  $\Rightarrow$  need to keep atoms at the optimal value of the total phase (e.g. max )

Idea: tune the CEO phase through AOMs or EOMs so the total phase stays constant  $\Rightarrow$  alternative to Zeeman slower

# Time evolution of velocity distribution



# Velocity comb

Force maxima are @  $\eta_n = (\omega_c - \omega_0 + \mathbf{k}_c \cdot \mathbf{v})T - \phi = 2\pi n$

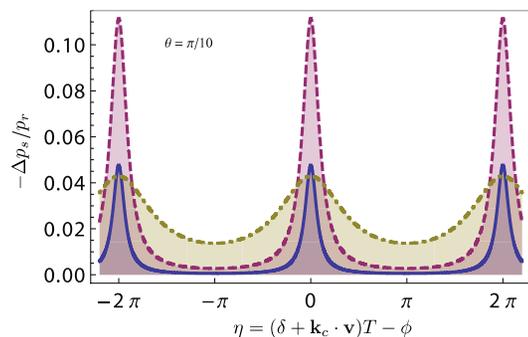
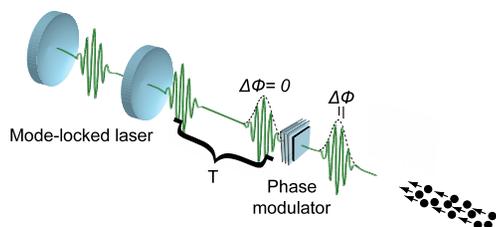
=> Velocity spikes are @

$$v_n = \frac{1}{k_c T} (2\pi n - T(\omega_c - \omega_0) + \phi)$$

separation 
$$v_{n+1} - v_n = \frac{\lambda_c}{T} = c \frac{\lambda_c}{2L_{\text{cavity}}}$$

As the teeth sweep through the velocity space, atomic  $v(t)$  trajectories are “snow-plowed” by teeth, ultimately leading to narrow velocity spikes collected on the teeth.

# Two-level Doppler cooling summary



- Problem can be solved exactly in a closed form
- Periodic dependence of the force reflects underlying FC structure
- The contrast of the force depends on the interplay between decoherence and interference
- CEO phase control = replacement for Zeeman slower
- FC could replace dedicated CW laser for lattice clock loading
- Periodic dependence of the force  $\Rightarrow$  Velocity comb
- Velocity comb = collision studies done in parallel?

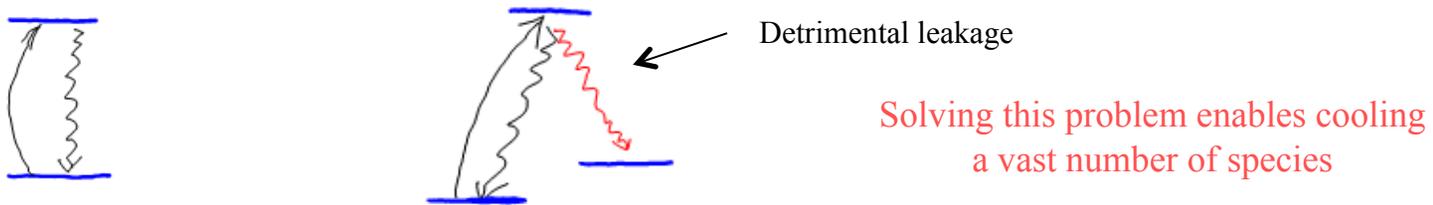
# Three-level $\Lambda$ -system

Dynamics of three-level  $\Lambda$ -type system driven by the trains of ultrashort laser pulses,  
E. Ilinova and A. Derevianko, Phys. Rev. A 86, 013423 (2012)

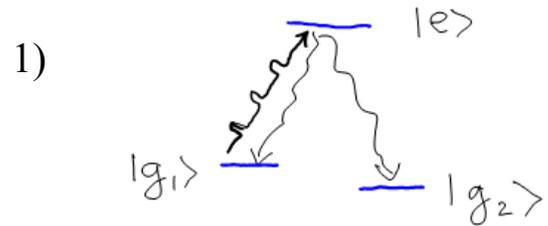
Doppler cooling of three-level  $\Lambda$ -systems by coherent pulse trains,  
E. Ilinova, A. Derevianko, Phys. Rev. A 86, 023417 (2012)

# See-saw protocol

Laser cooling requires cycling (closed) transitions



## See-saw protocol idea



Cool on the  $g_1$ - $e$  transition until most of the population accumulates in  $g_2$

2) Tune the comb in resonance with the  $g_2$ - $e$  transition (Change phase)  $\nu_n = \nu_c + n\nu_{rep} - \frac{1}{2\pi T_{rep}}\phi$

**Work in progress**



Cool on the  $g_2$ - $e$  transition until most of the population accumulates in  $g_1$

4) Tune the comb in resonance with the  $g_1$ - $e$  transition (Change phase)

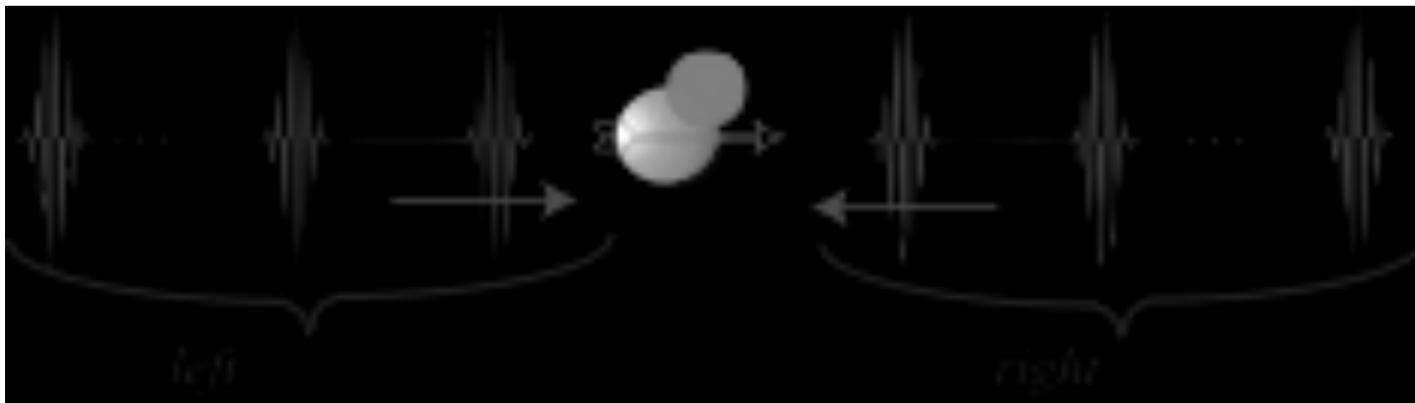
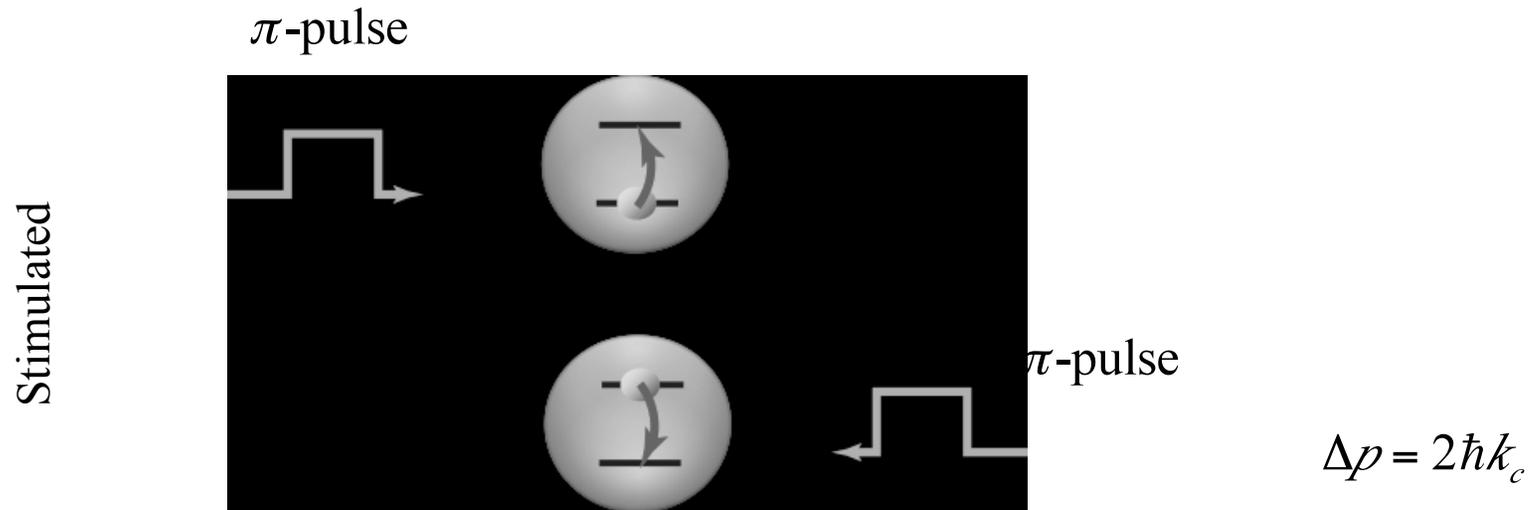
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Cooling is accomplished with a single laser source on a multi-level system

# Stimulated cooling of molecules on multiple rovibrational transitions with coherent pulse trains

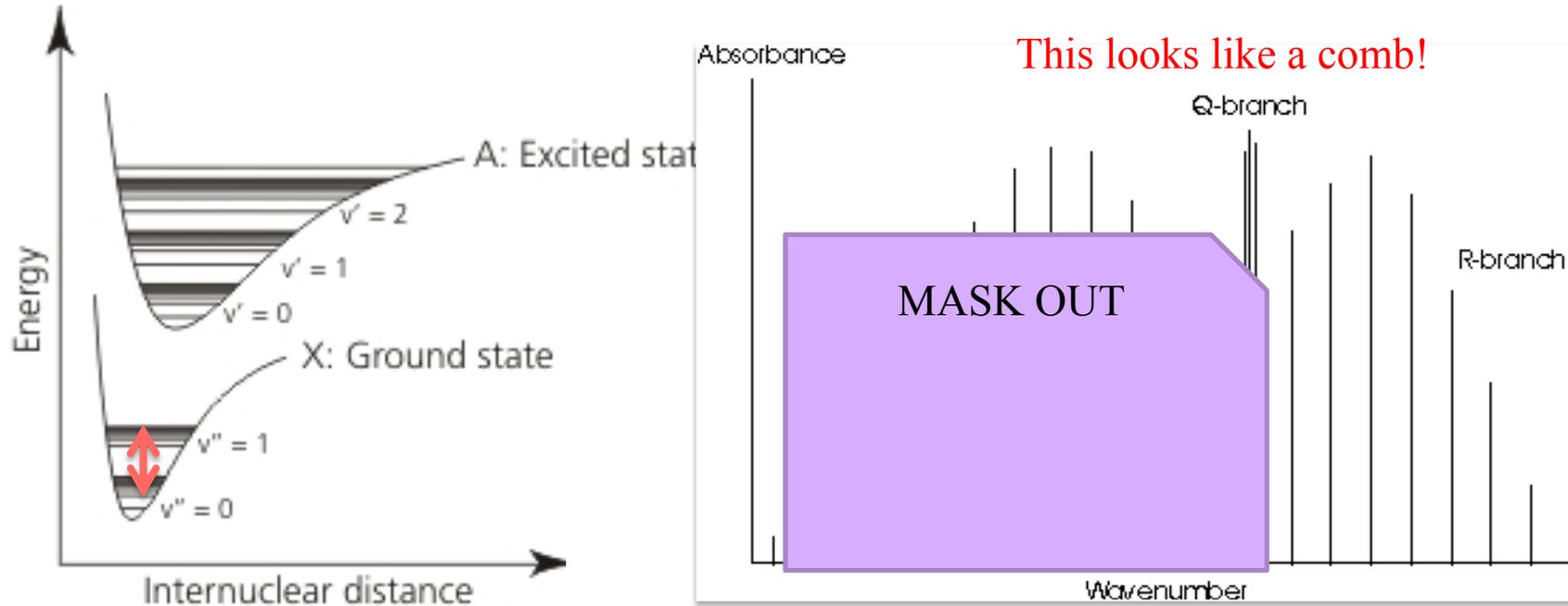
Stimulated cooling of molecules on multiple rovibrational transitions with coherent pulse trains, E. Ilinova, J. Weinstein, A. Derevianko, arXiv:1201.1015

# Stimulated cooling and Pi-trains



# Molecular ro-vibrational spectra

Focus on a vibrational transition inside the same electronic manifold



R-branch:  $J \rightarrow J+1$

$$\nu_{J,J+1}^{v,v'} \approx \nu_{0,1}^{v,v'} + (3B_{v'} - B_v)J$$

# Matching ro-vibrational and FC spectra

$$\nu_{J,J+1}^{v,v'} \approx \nu_{0,1}^{v,v'} + (3B_{v'} - B_v)J$$

$$\nu_n = \nu_c + \nu_{\text{rep}} \times n - \frac{\phi}{2\pi T}$$

Focus on the  $v=0 \rightarrow v=1$  transition

$$\nu_c = \nu_{0,1}^{0,1} \quad \nu_{\text{rep}} = (3B_1 - B_0) / n'$$

Example:

$$\text{LiCl: } \nu_c \approx 19.29 \text{ THz} \quad T = 0.95 \text{ ns} \quad n' = 40$$

$$\text{PbO: } \nu_c \approx 21.63 \text{ THz} \quad T = 0.98 \text{ ns} \quad n' = 15$$

# Back to the two-level system

Since we have masked out P-branch ( $J \rightarrow J-1$ ) and focused on the R-branch, the dynamics of levels ( $v=0, J$ )-( $v=1, J+1$ ) becomes uncoupled  
 = collection of independent two-level systems

Radiative branching is very slow  $\sim$  ms and can be ignored

We use our previous analytical propagators for ultrashort pulses

## Momentum kick

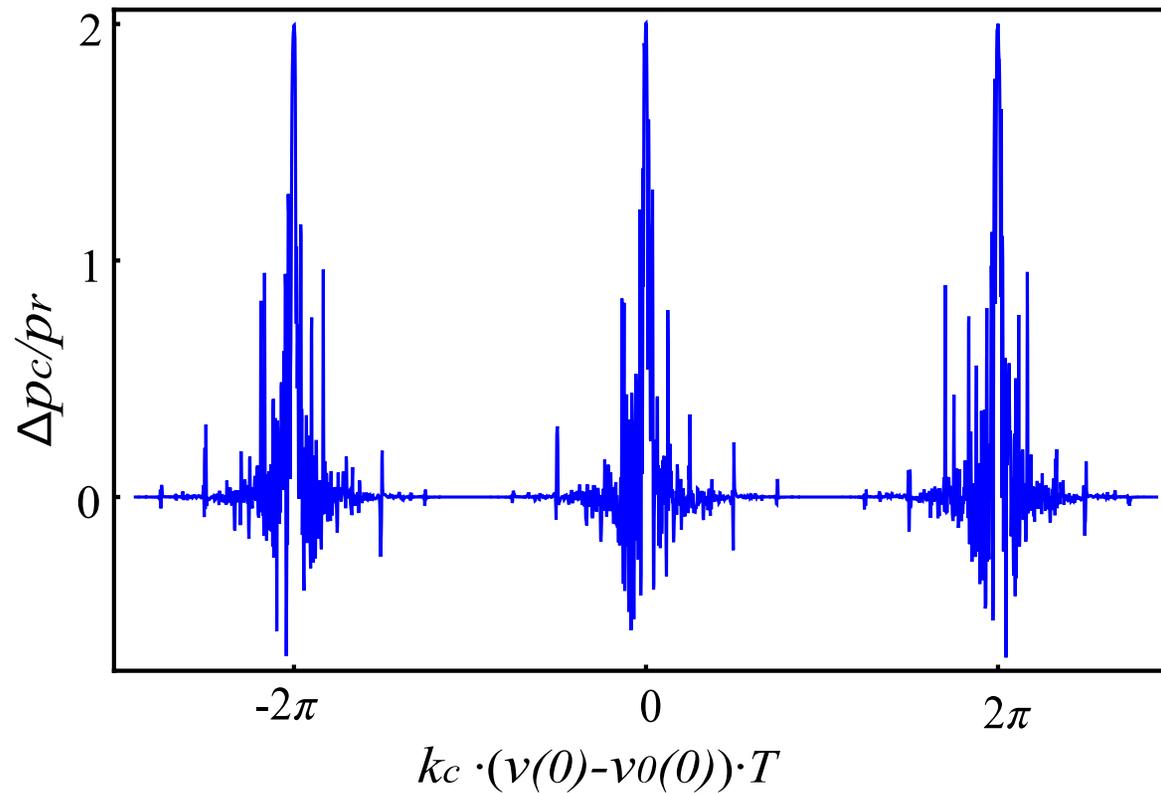
$$\frac{\Delta p_{\text{cycle}}}{p_r} = \rho_{ee}(t_0^+ + 2(N-1)T) + \rho_{ee}(t_0^-) - 2\rho_{ee}(t_0^+ + (N-1)T)$$

After the left pi-train
After the right pi-train =  
Before the left pi-train

Before the right pi-train

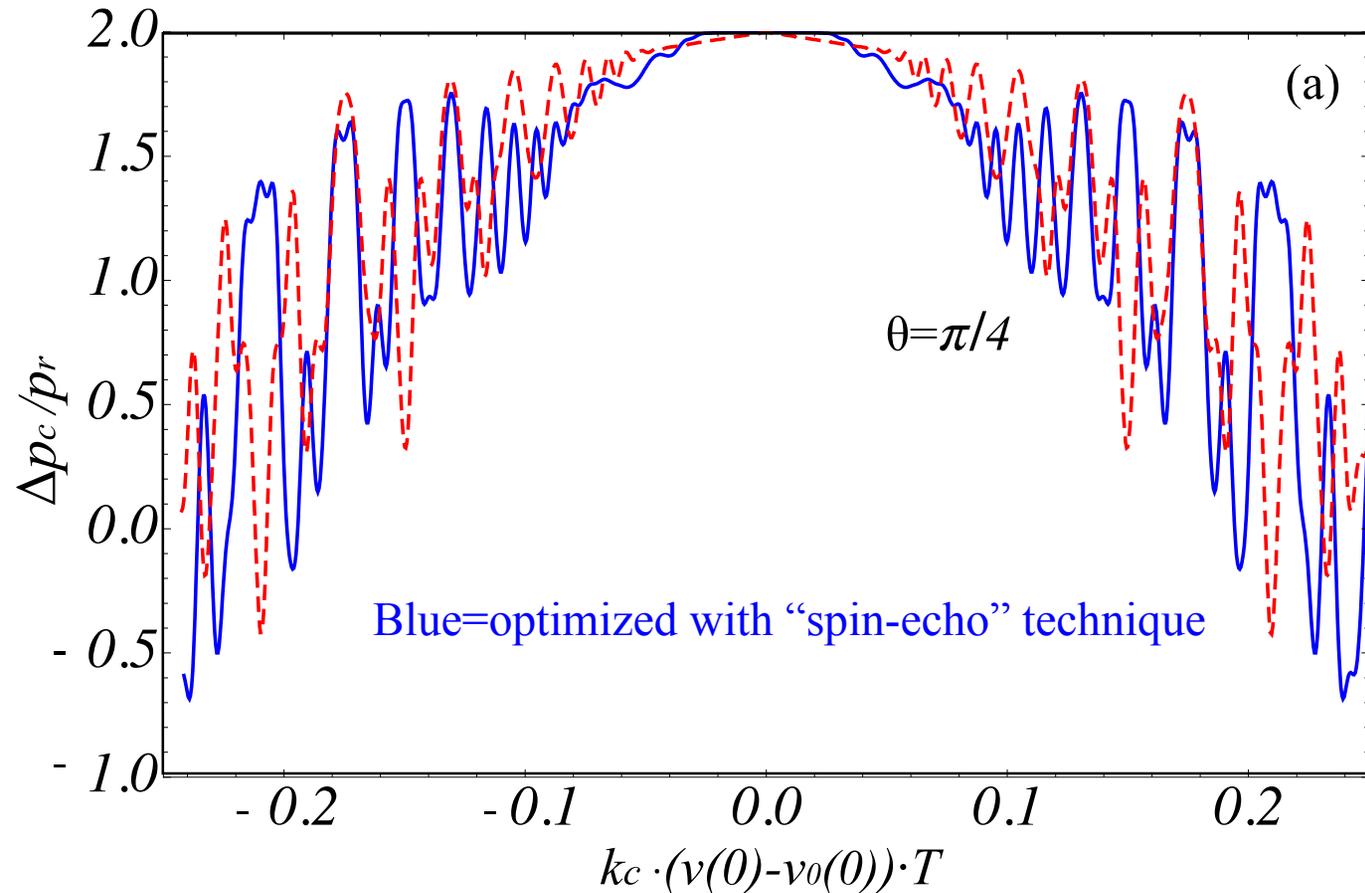
$$\left( \frac{\Delta p_{\text{cycle}}}{p_r} \right)_{\text{max}} = 2$$

# Average momentum kick



- Periodic function, reflecting the underlying periodicity of the FC spectrum
- At the peaks the system starts and ends in the ground state, with a full transfer of the population to the excited state by the first train (constructive interference)

# Average momentum kick (zooming in)

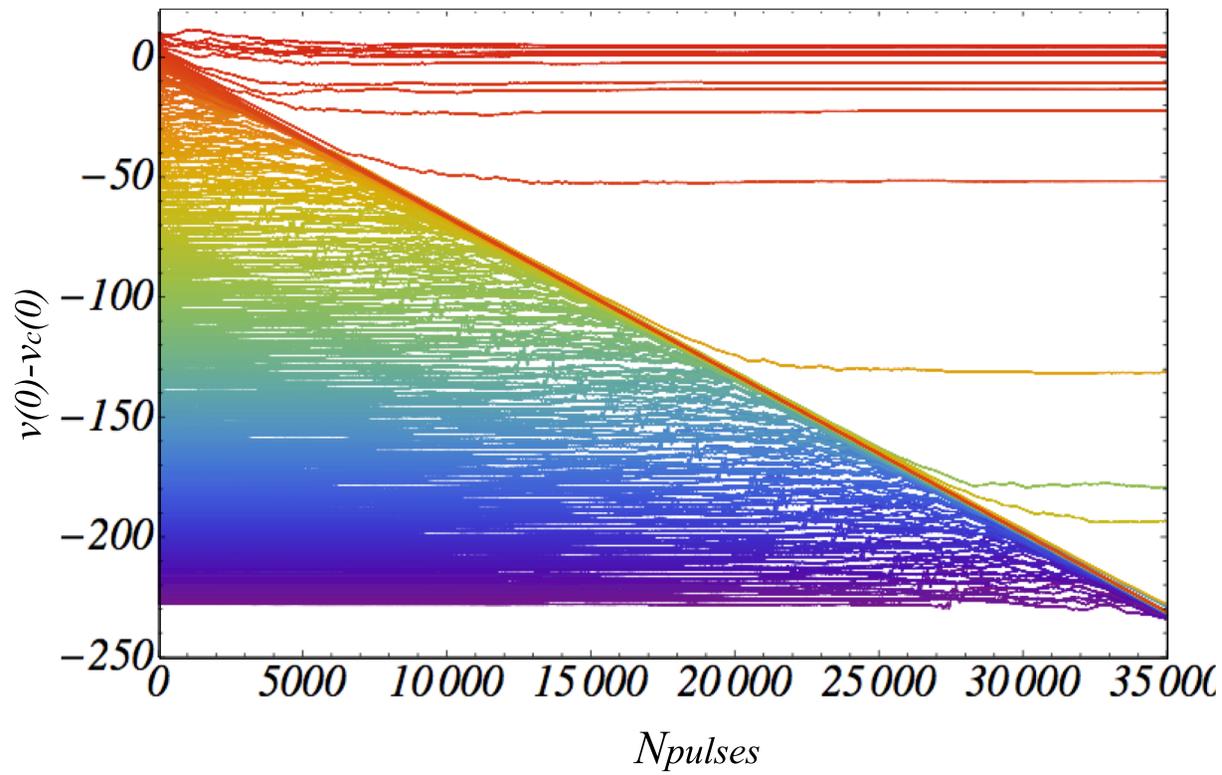


Complicated substructure resulting from intricate interferences of probability amplitudes driven by the multitude of pulses

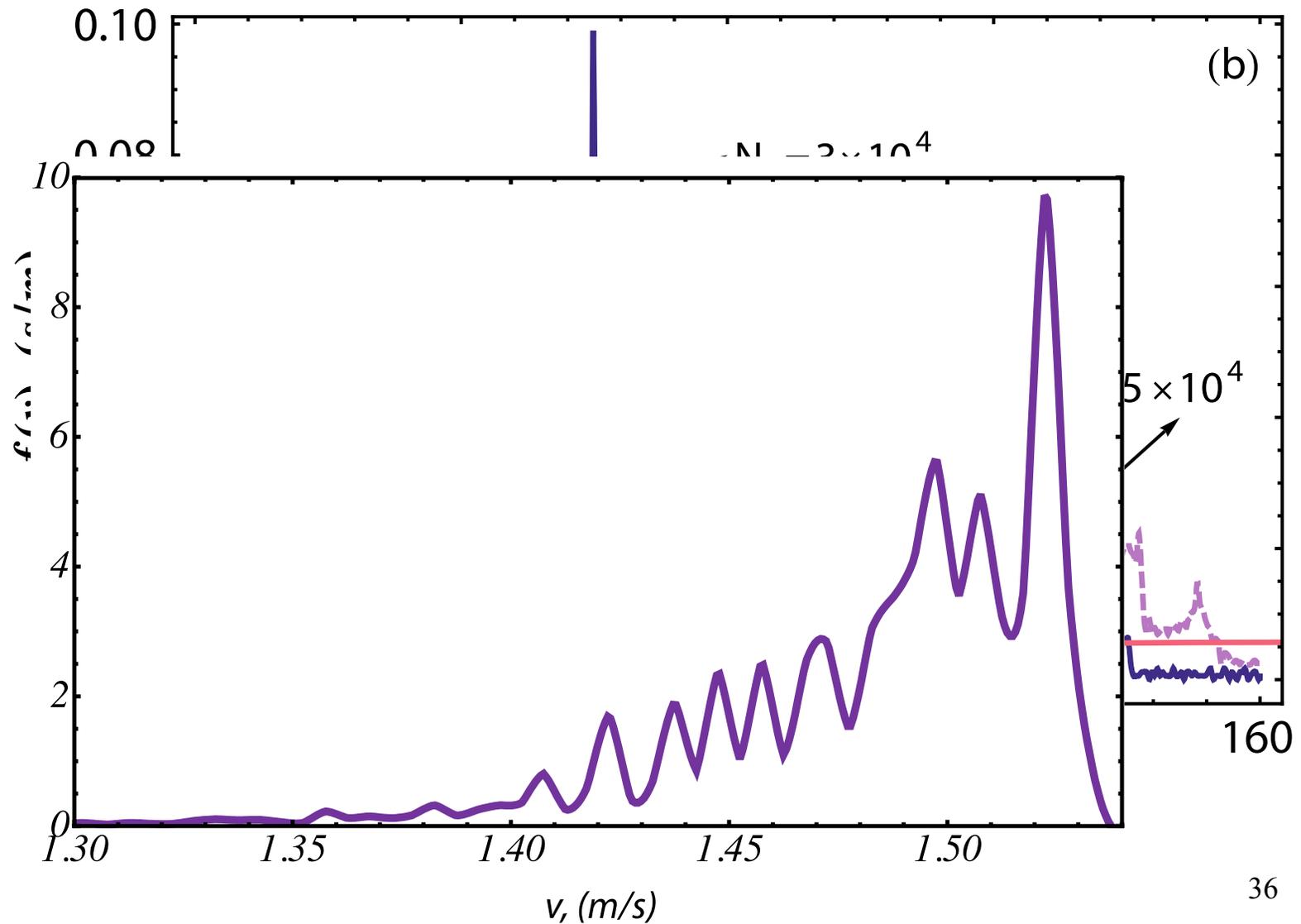
# Snow-plowing through the velocity space

As molecules slow down we tune the CEO phase, so the force tracks changing Doppler shift

$$d\phi / dt \approx p_r^2 / (\hbar M) / (N-1)$$



# Time evolution of velocity distribution



# Summary: stimulated optical force

- ❖ Spontaneous decay rates can be very slow –  
no problem as long as pi-train conditions are satisfied
- ❖ Much faster than Doppler cooling/slowing for lifetimes  $> 10$  ns
- ❖ Occasional photon emission is a plus: resets the population + carries away the entropy
- ❖ In molecules branching ratio problem can be fully avoided  
(if the spontaneous decay is slow)
- ❖ Frequency comb - using several transitions at once for exerting optical force