Laser Cooling and Slowing of Diatomic Molecules

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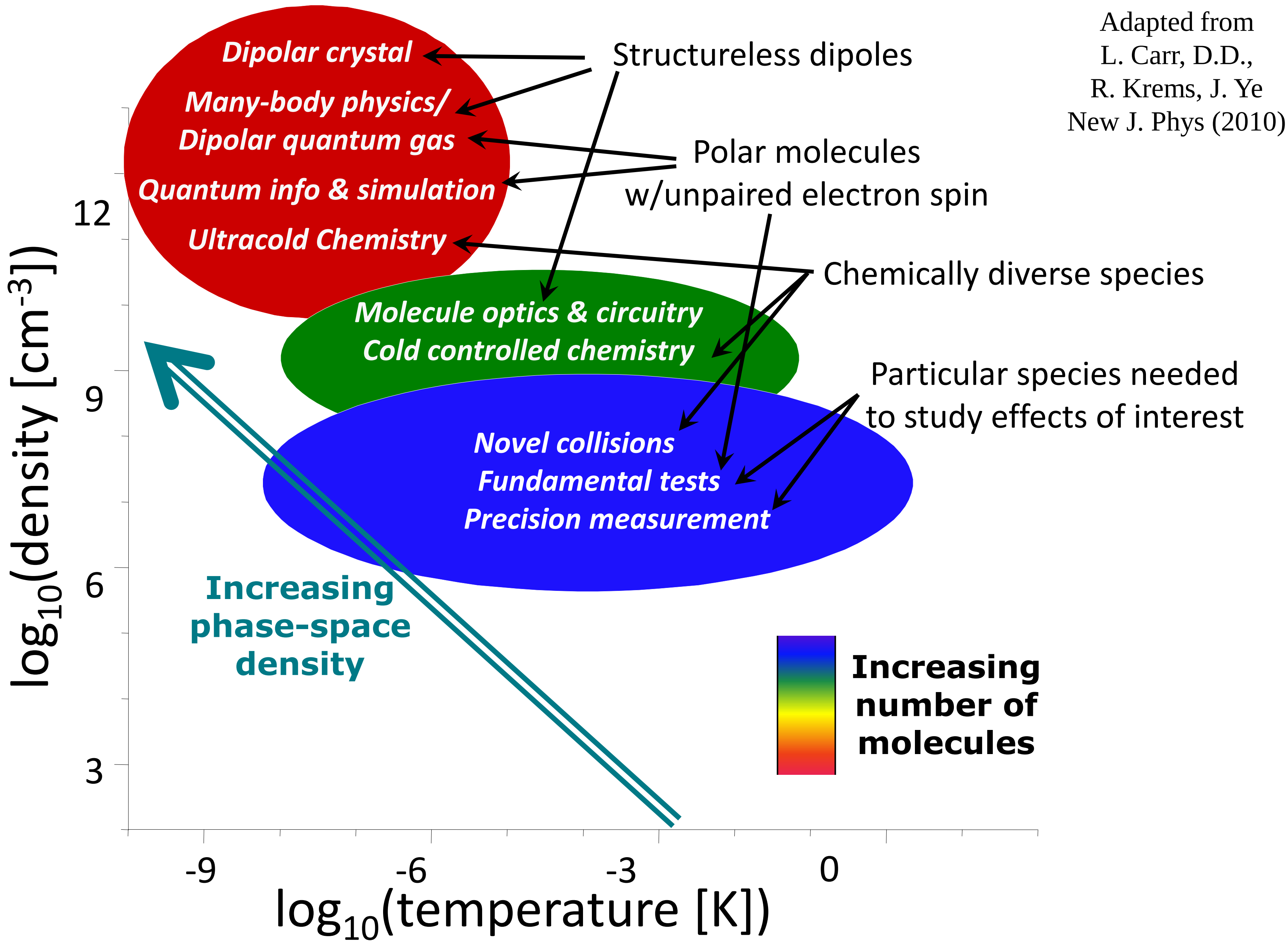
- Overview & motivations
- Transverse laser cooling of a molecular beam
- Longitudinal slowing of a molecular beam
- Towards trapping

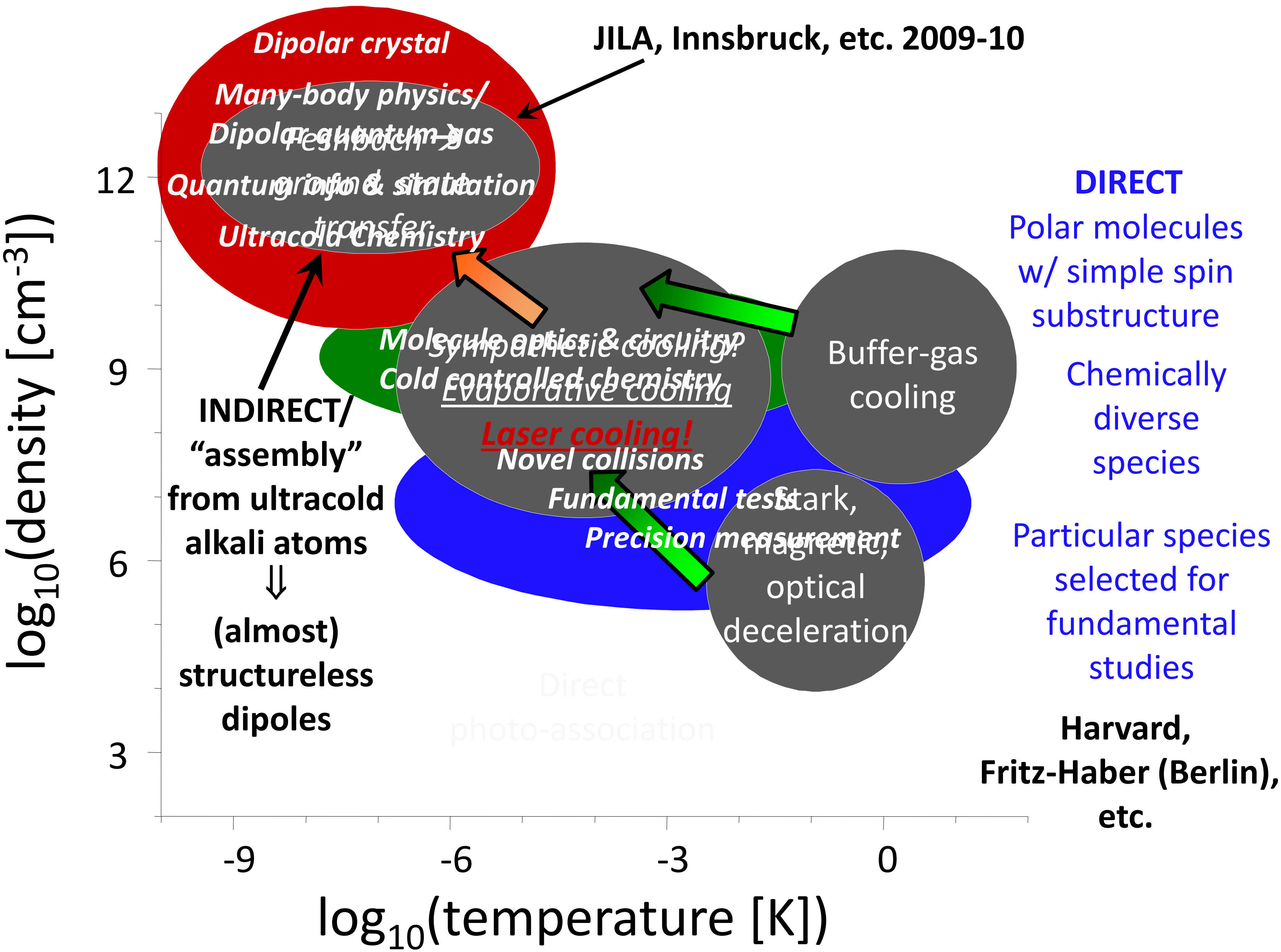
DeMille Group

Funding
NSF, ARO, AFOSR

Diverse physics goals w/cold & ultracold molecules: different conditions & types of molecules desired

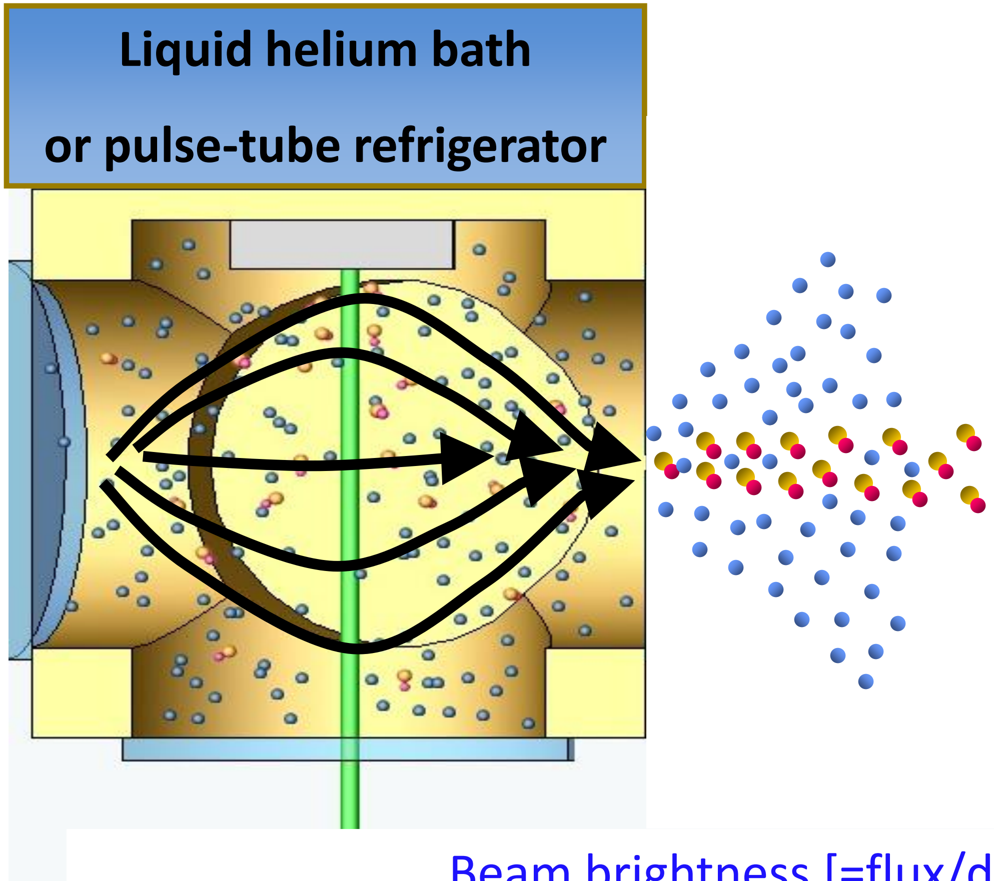


Different techniques address different needs



Starting point= new molecular beam technology: ***hydrodynamically enhanced*** *cryogenic beams*

[Maxwell *et al.* PRL 2005; Patterson & Doyle J Chem Phys 2007; Barry *et al.* PCCP 2011; Hutzler *et al.* PCCP 2011]



- Inject hot molecules (e.g. via laser ablation)
- Cool w/cryogenic buffer gas *@high density*
- Efficient extraction to beam via "wind" in cell: $10^{-4} \rightarrow$ 10%-40%
- "Self-collimated" by extraction dynamics
- Rotational cooling in expansion: T ~ 1 - 4K
- Moderately slow: v ~ 130-180 m/s
- BUT: spatially and temporally extended

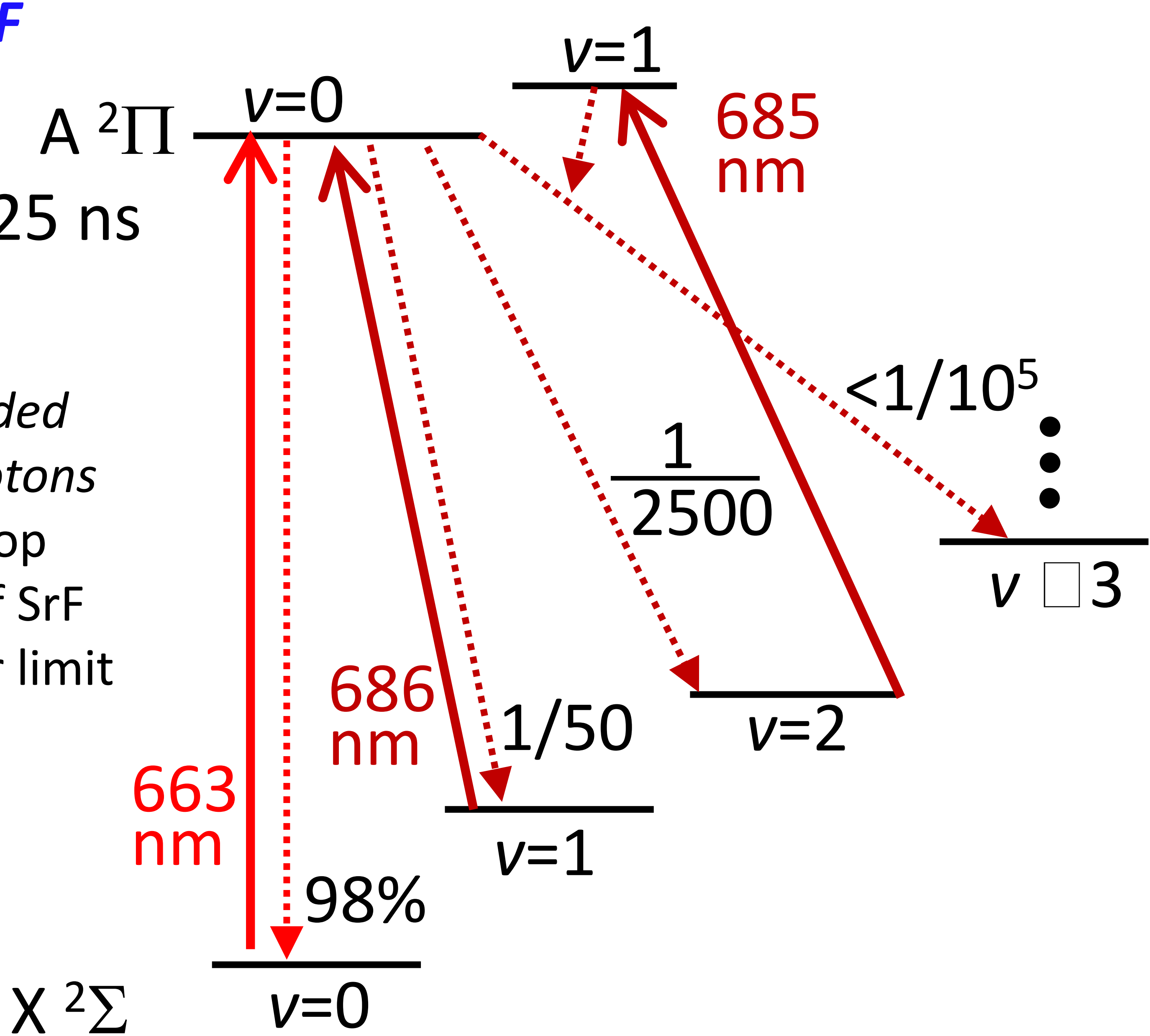
Beam brightness [=flux/divergence] ~ ***$10^3 \times$ larger***
than other sources for refractory/free radical species:
Typically ~ 2×10^{11} mol/sr/state/pulse @ □10 Hz rep. rate

Basic method used for SrO, **ThO**, **<u>SrF</u>**, BaF, O_2, NH_3, Yb, …

Beating vibrational branching by choice of species

Large handful of “easy” cases w/ favorable Franck-Condon factors [DiRosa, EPJD 2004]

Our case: ***SrF***



Only 3 lasers needed to scatter >10^5 photons

→sufficient to stop cryogenic beam of SrF and cool to Doppler limit

Beating rotational branching: general strategies

•Closed rotational transition (with dark Zeeman sublevels) using lowest P- OR Q-branch transition

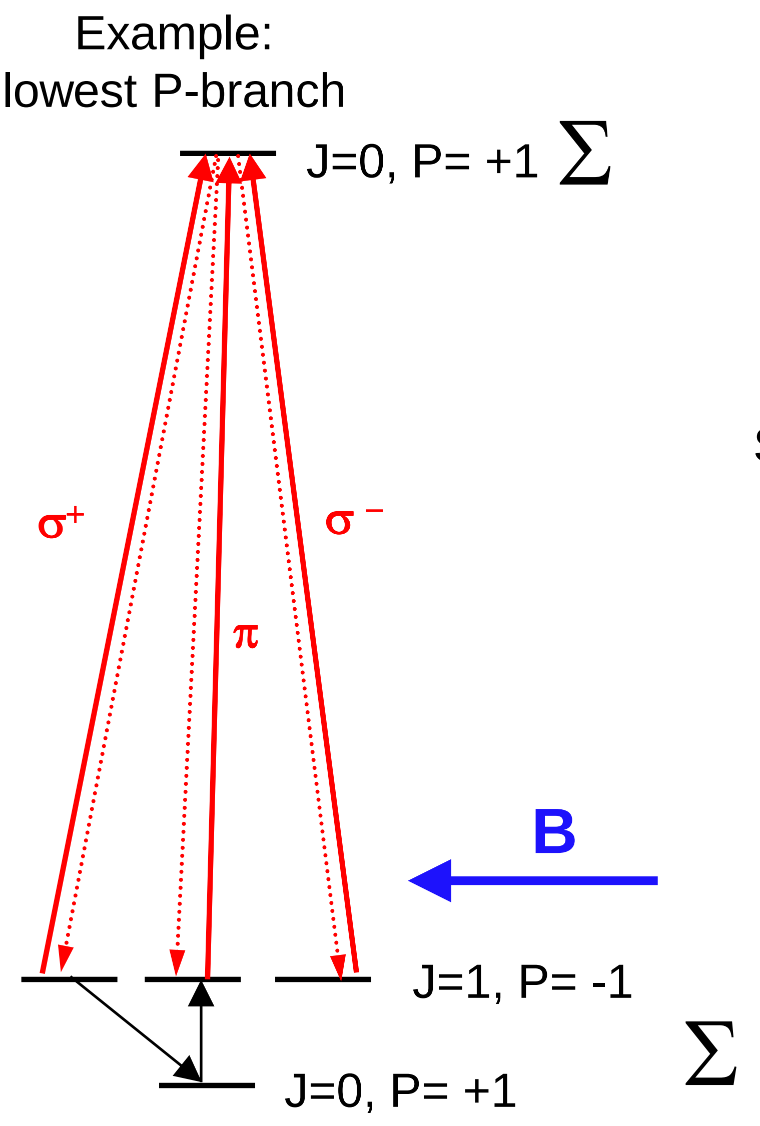
Dark Zeeman sublevels must be remixed into cycle

at least 3 routes for “typical” diatomics

1. Transverse B-field
   +Larmor precession

2. Rapidly switch
   laser polarization

3. Resonant E-field
   (microwave or DC)
   repumping



Inspired by
Stuhl *et al.* (Ye/JILA)
PRL 2009
proposal

“Extra” levels □ reduced scattering rate

Population fractions for saturated transitions determine maximum scattering rate: $\Gamma^{Max} = P_{exc}/\tau$

1+1 = 2-level system:

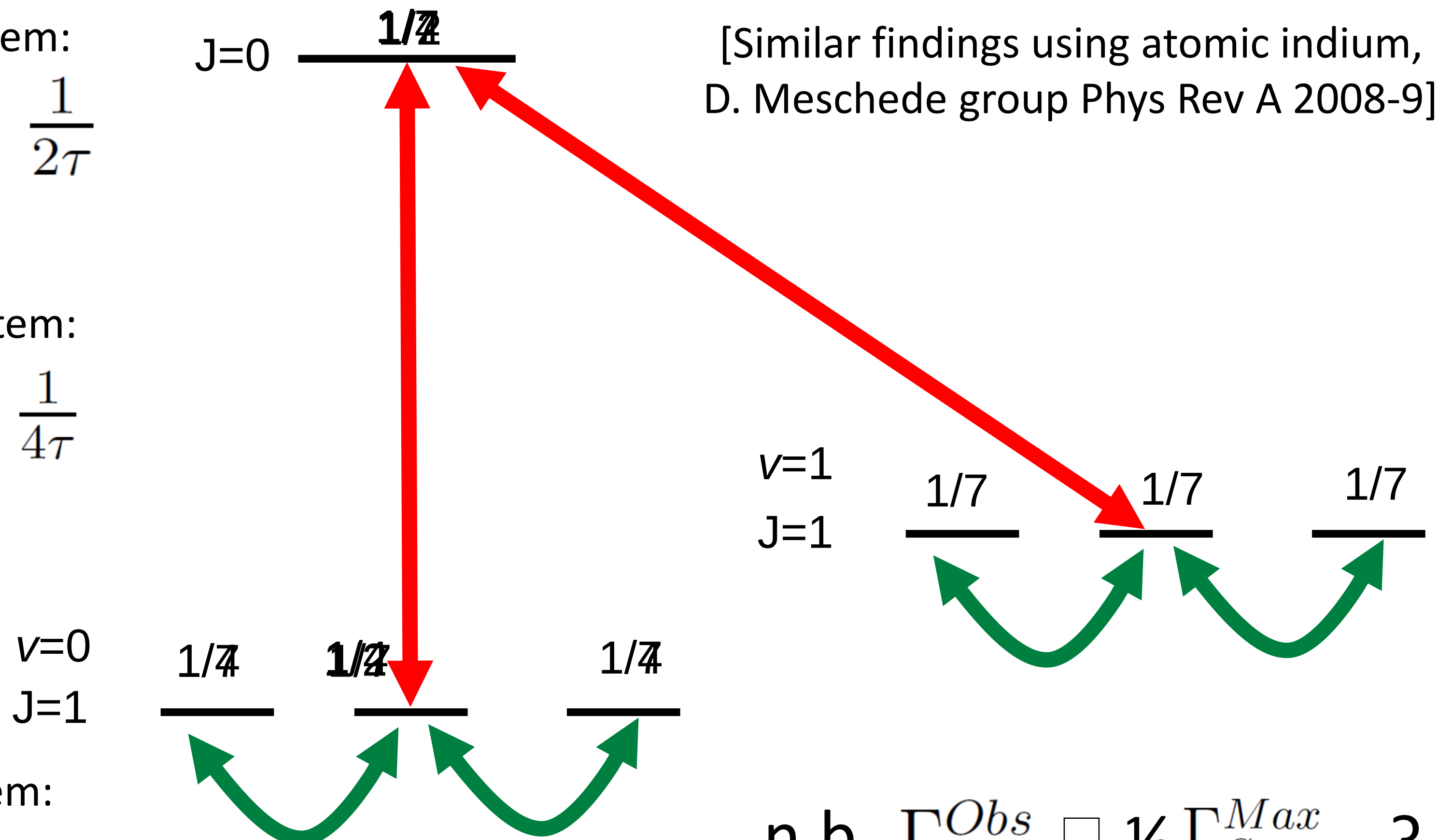
$$\Gamma^{Max}_{Scat} = \frac{1}{2\tau}$$

3+1-level system:

$$\Gamma^{Max}_{Scat} = \frac{1}{4\tau}$$

3+3+1-level system:

$$\Gamma^{Max}_{Scat} = \frac{1}{7\tau}$$

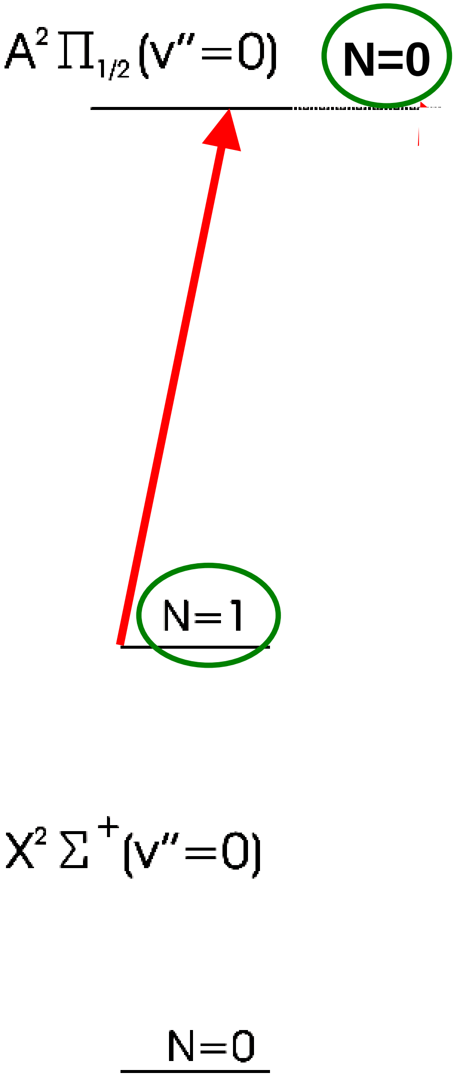


[Similar findings using atomic indium, D. Meschede group Phys Rev A 2008-9]

n.b. Γ^{Obs}_{Scat} □ ½ Γ^{Max}_{Scat} …?

Force = □k□$_{Scat}$ is reduced vs. atoms

Level scheme for optical cycling in SrF [with spin-rotation + hyperfine substructure]



Closed
rotational transition
due to selection rules

+ remixing of
dark sublevels
w/added fields

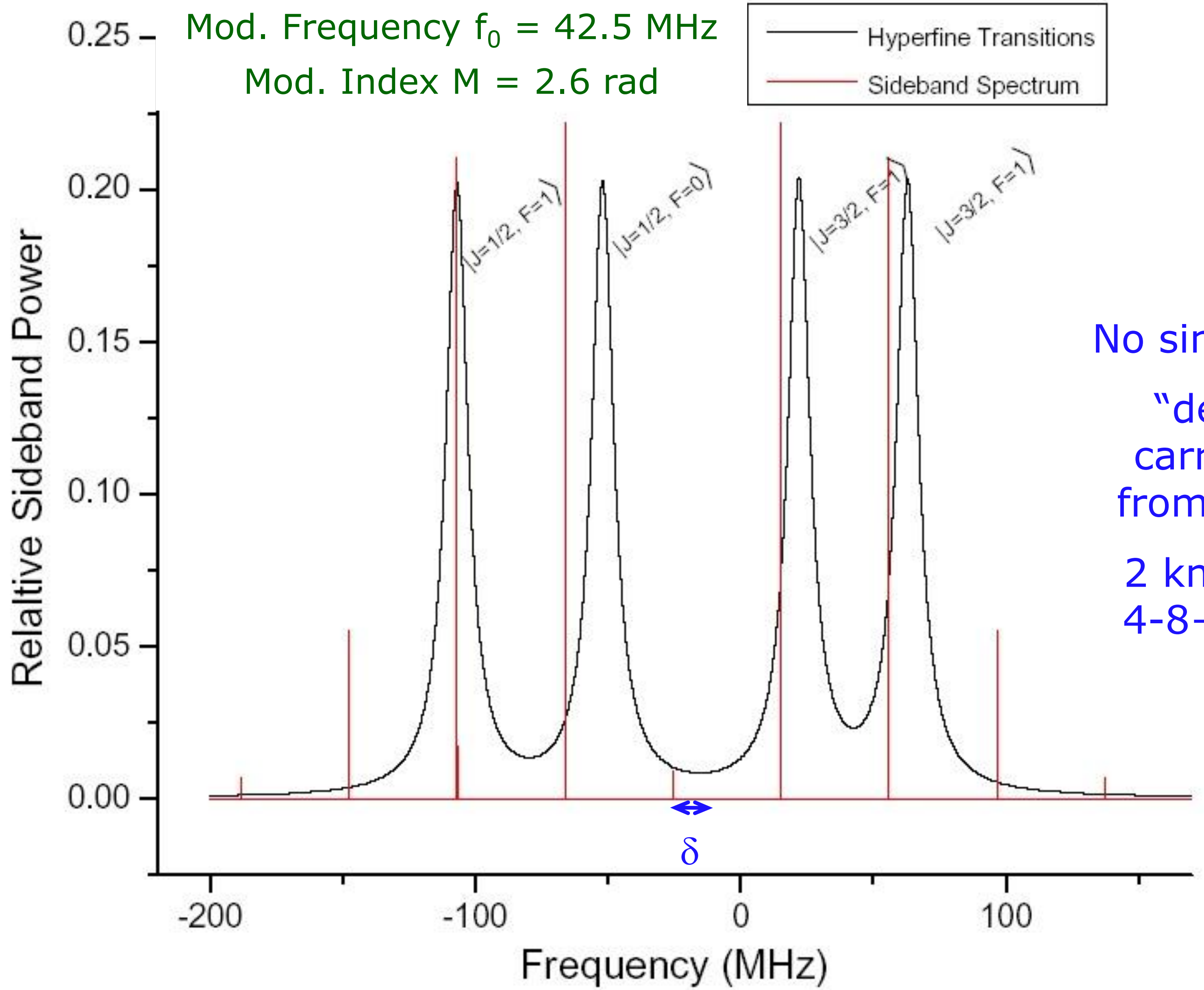
So far with SrF:
laser sidebands
for hyperfine repumping
+transverse B-field
r Zeeman sublevel remixing

Single-frequency modulation for SrF hfs repumping

Example with

Mod. Frequency f_0 = 42.5 MHz

Mod. Index M = 2.6 rad



NOTE:

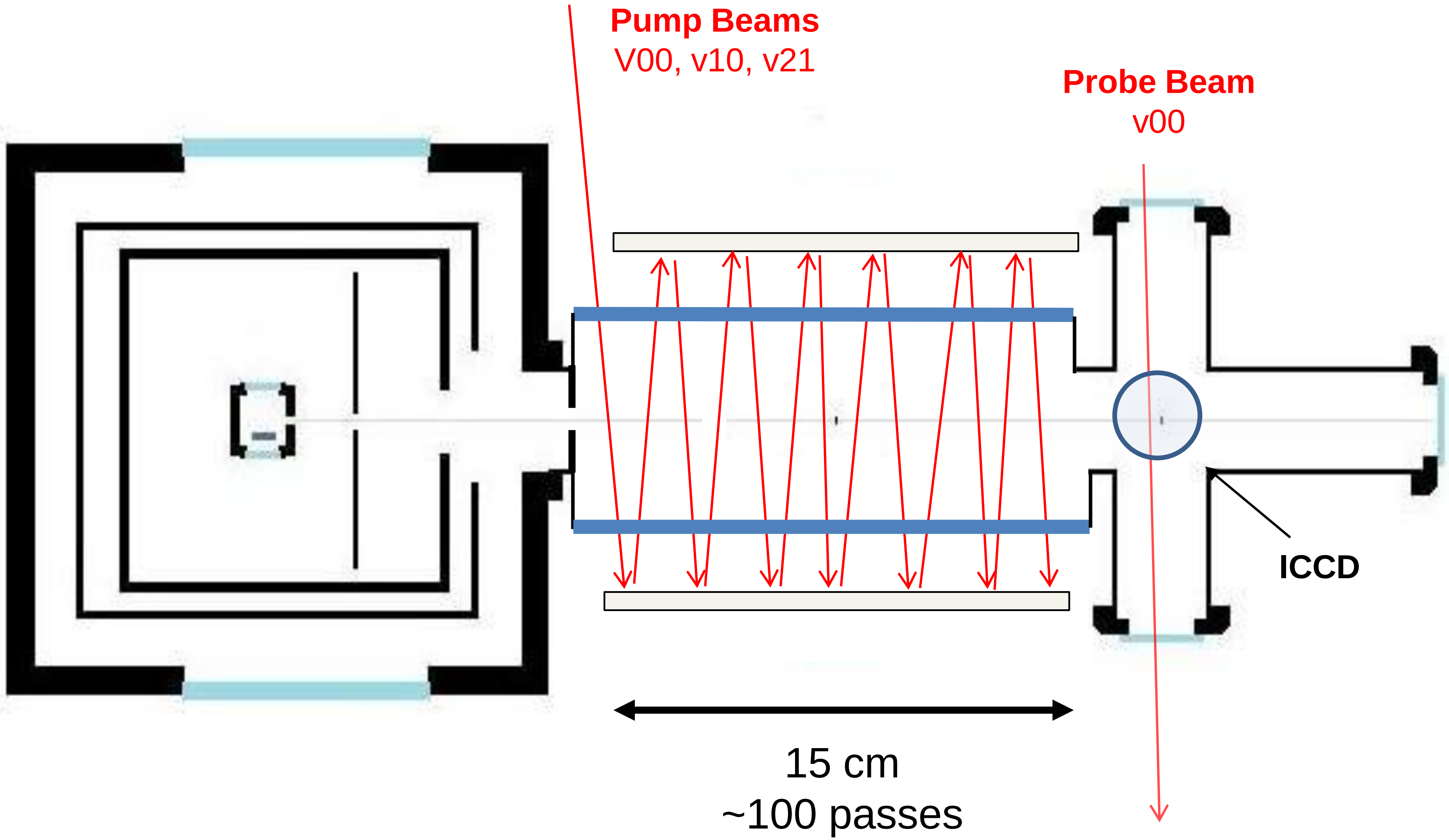
No single detuning

"detuning" ≡ carrier offset δ from max signal

2 knobs control 4-8+ detunings

1D transverse cooling of an SrF beam

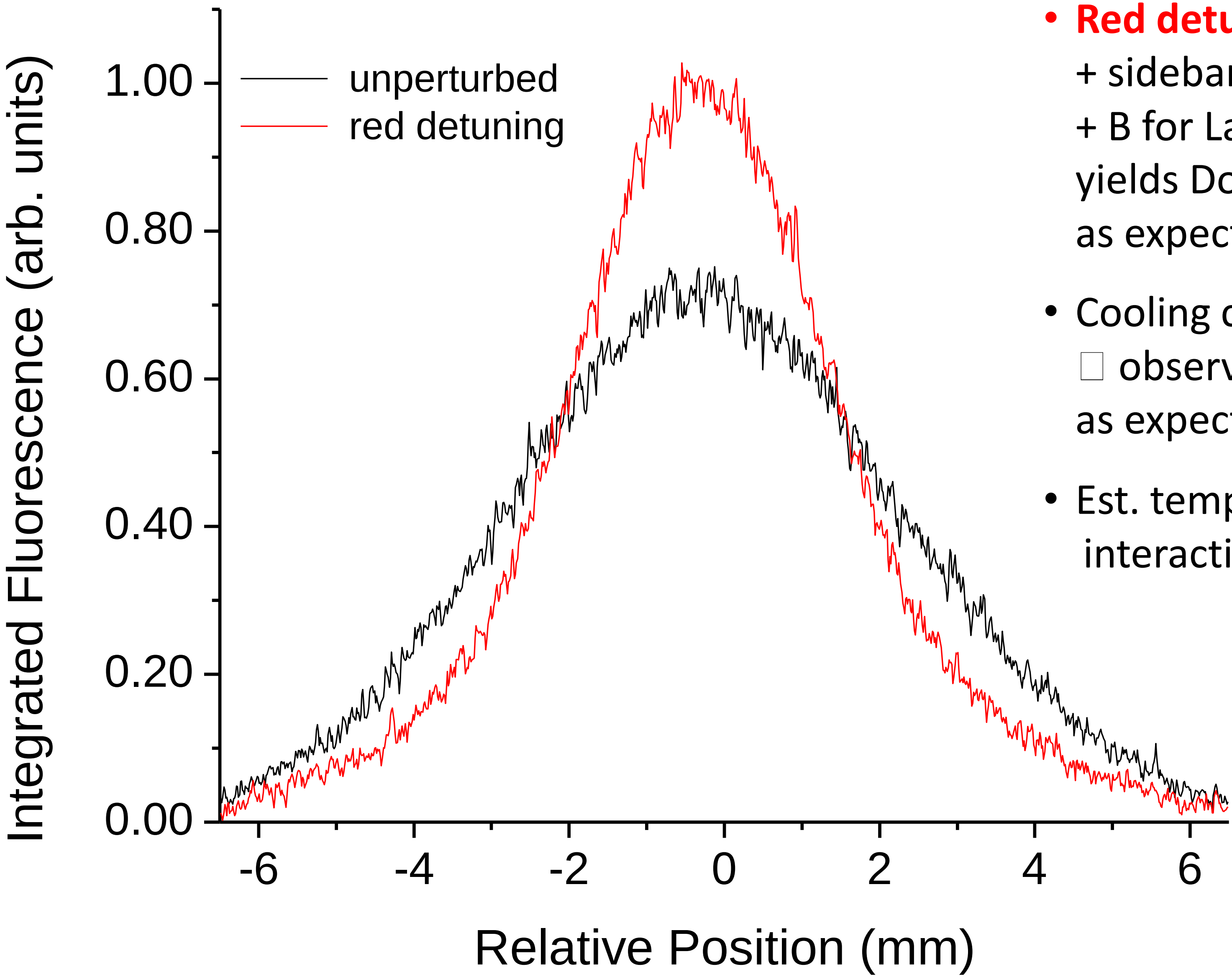
[E.S. Shuman *et al.* Nature **467**, 820 (2010)]



Long interaction region necessary
for cooling with reduced scattering rate
(due to “extra” ground-state sublevels)

Doppler cooling of SrF

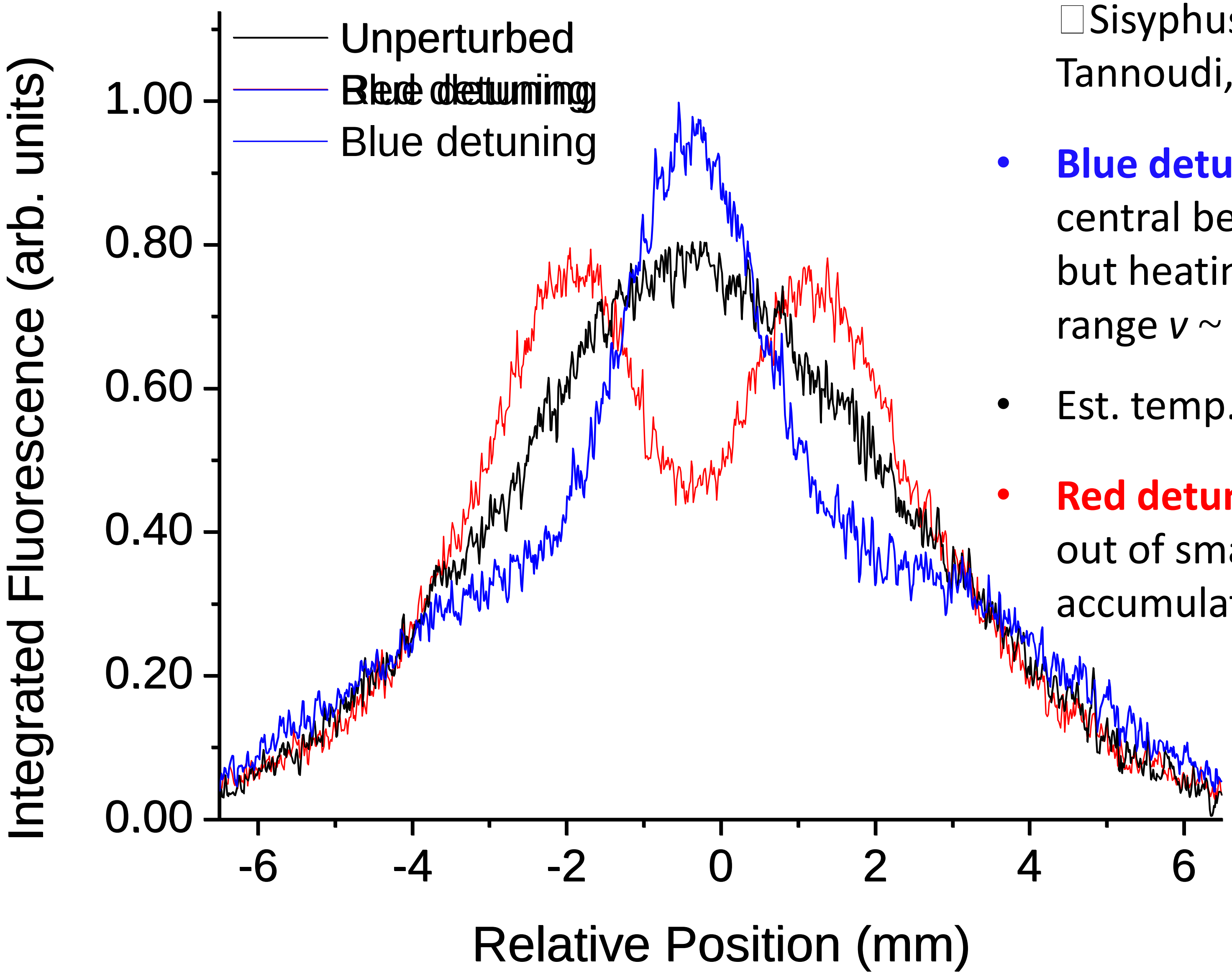
[E.S. Shuman *et al.* Nature **467**, 820 (2010)]



- **Red detuning** by ~Γ_{nat}
  + sideband freq. 46.4 MHz
  + B for Larmor freq. ω_B~Γ_{nat}
  yields Doppler Cooling
  as expected
- Cooling capture range
  □ observed width (~4 m/s)
  as expected
- Est. temp. T_{Dop} ~ 5 mK
  interaction time-limited

Sisyphus cooling (or heating) in SrF

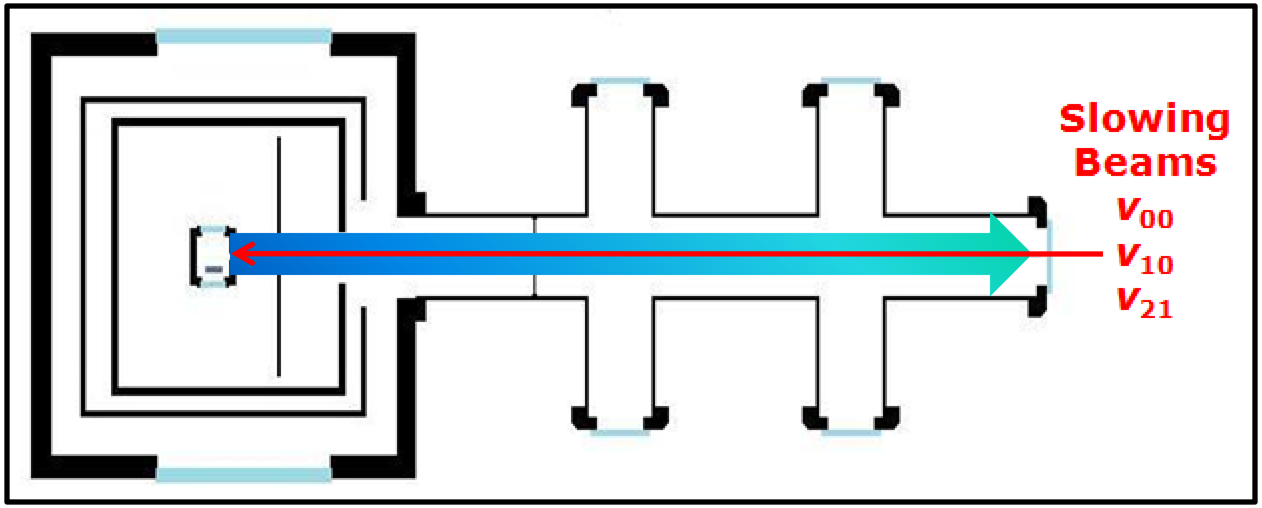
[Shuman *et al.* Nature **467**, 820 (2010)]



- Laser standing waves + Small B: $\omega_B \sim \Gamma_{nat}/10$ □Sisyphus effect [Cohen-Tannoudi, Metcalf,etc.]
- **Blue detuning** yields colder central beam than Doppler, but heating outside capture range $v \sim 1$ m/s
- Est. temp. $T_{Sis} \sim 300\ \mu K$
- **Red detuning** gives heating out of small velocities, accumulation at non-zero v

Laser slowing of a molecular beam: experimental setup

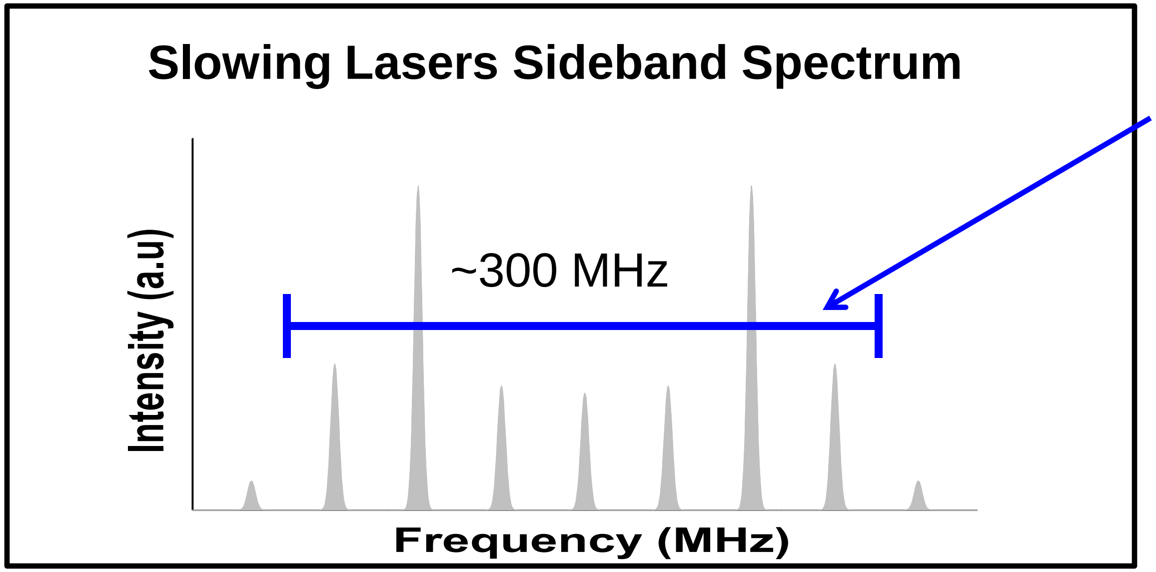
[J.F. Barry *et al.*, PRL **108**, 103002 (2012)]



$v_{fwd} \approx 150$ m/s

~20,000 photon recoils to stop SrF

$\Gamma_{scat}=3\times10^6\ s^{-1}$ ➔ ~70 cm to stop



Extra sidebands via EOM ➔ Larger capture range

~"white light slowing"

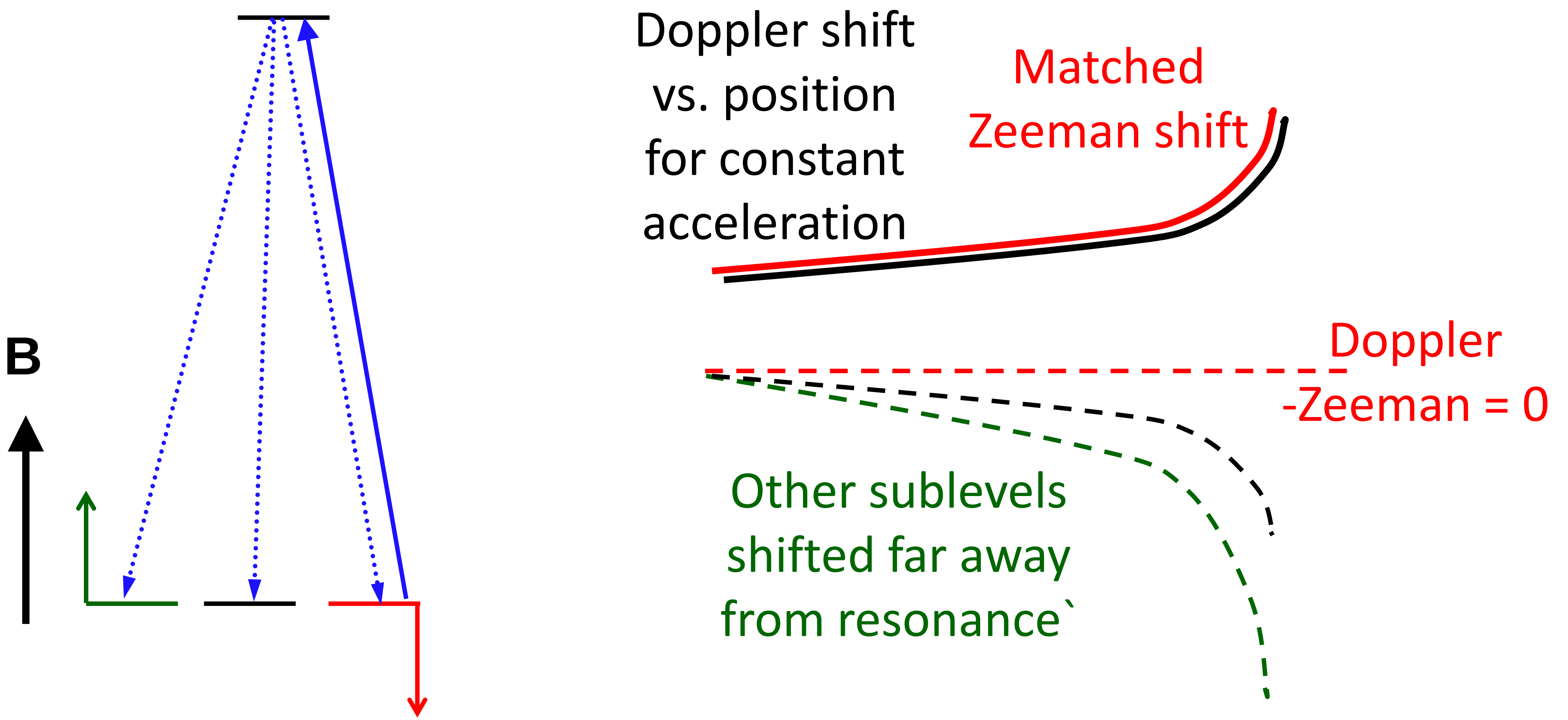
n.b.: Zeeman slowing of molecules is impractical

Cycling transition fast redistribution over Zeeman sublevels

BUT: shaped B-field such that

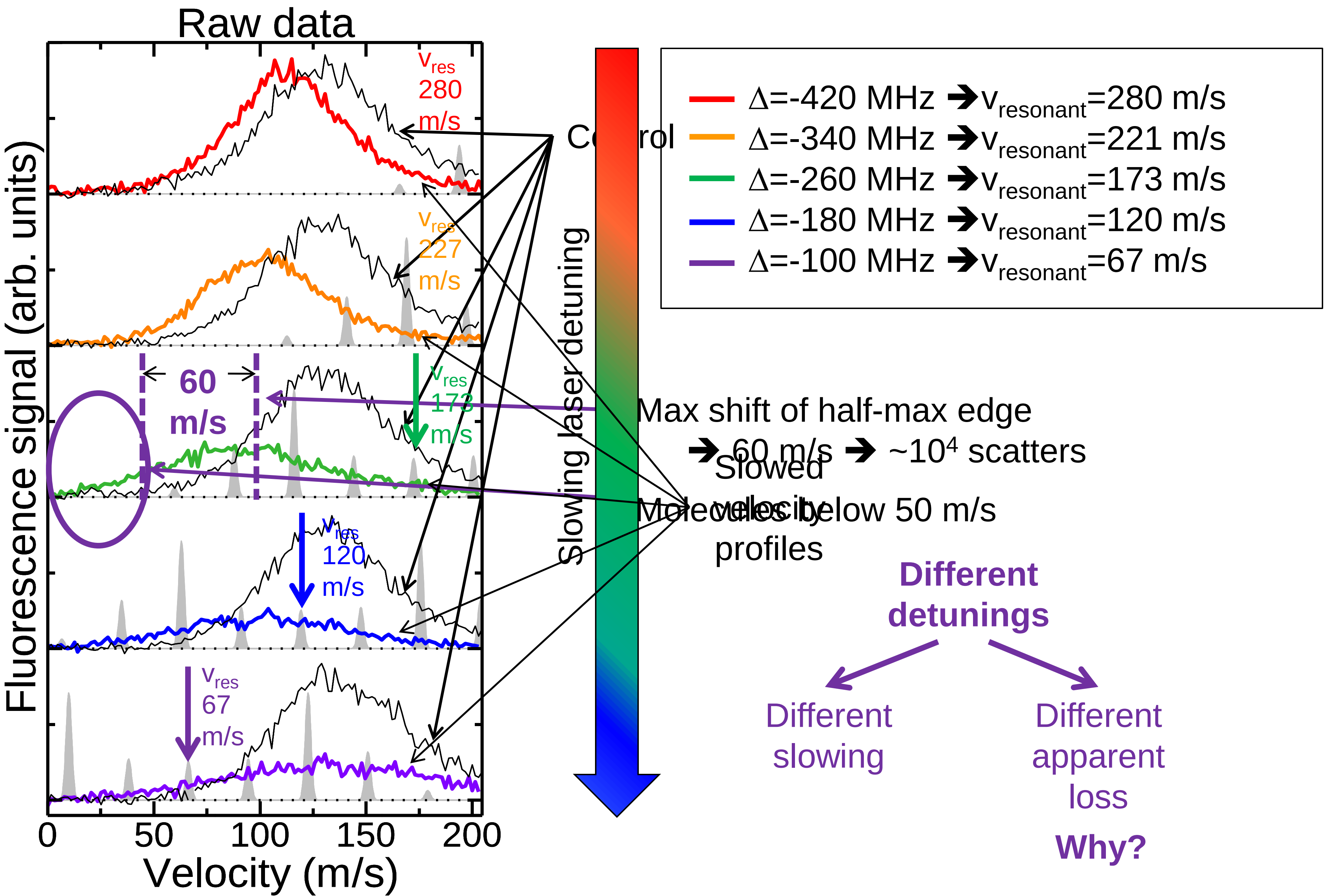
Zeeman shift = Doppler shift during slowing

possible for only 1 sublevel



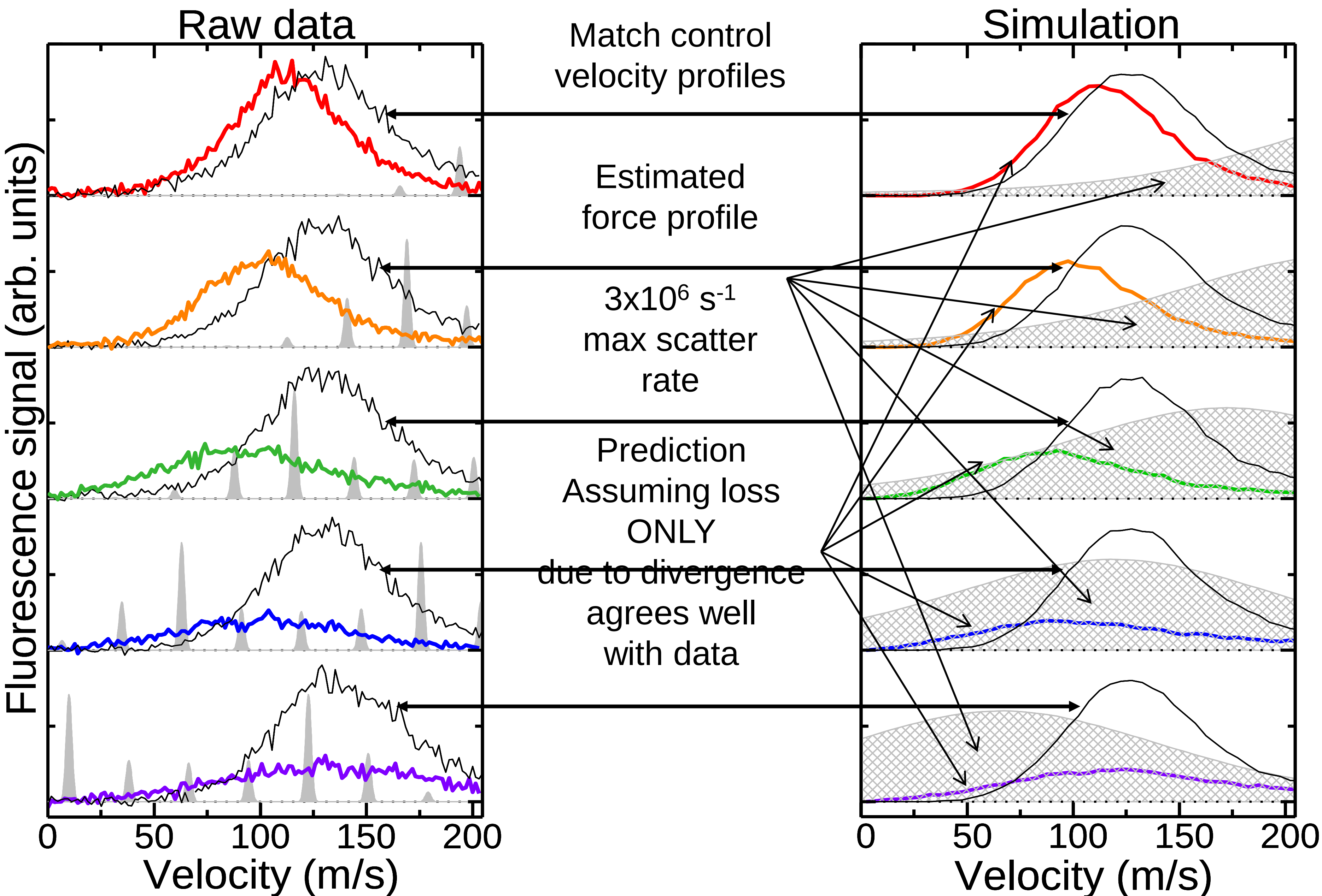
Raw Slowing Data

[Barry *et al.*, PRL **108**, 103002 (2012)]

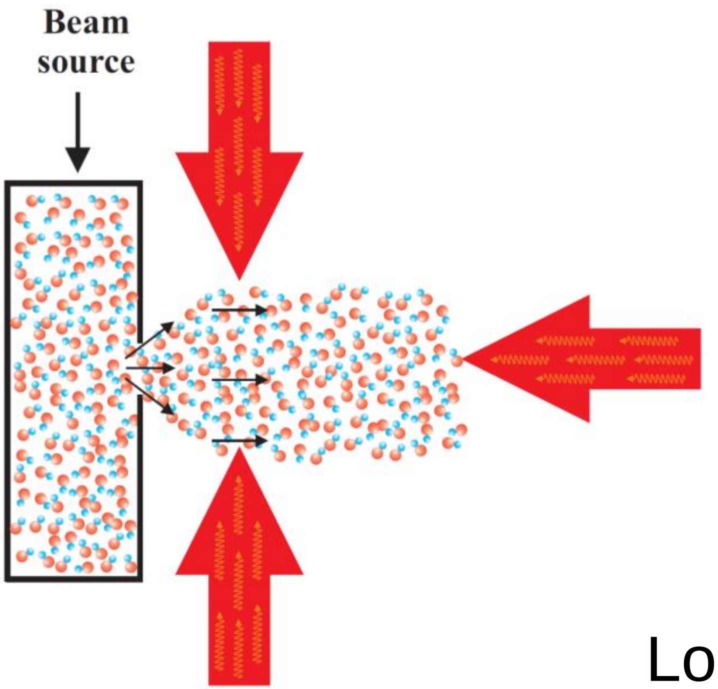


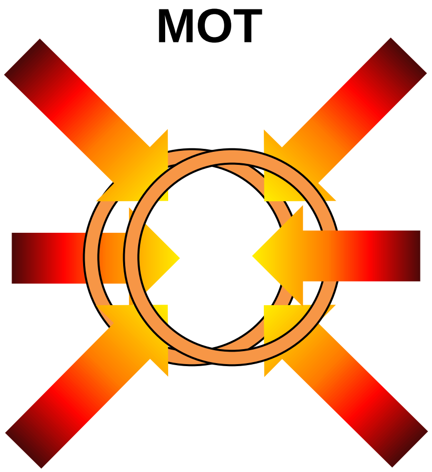
Loss due merely to divergence/transverse heating

[Barry *et al.*, PRL **108**, 103002 (2012)]



Towards trapping of SrF



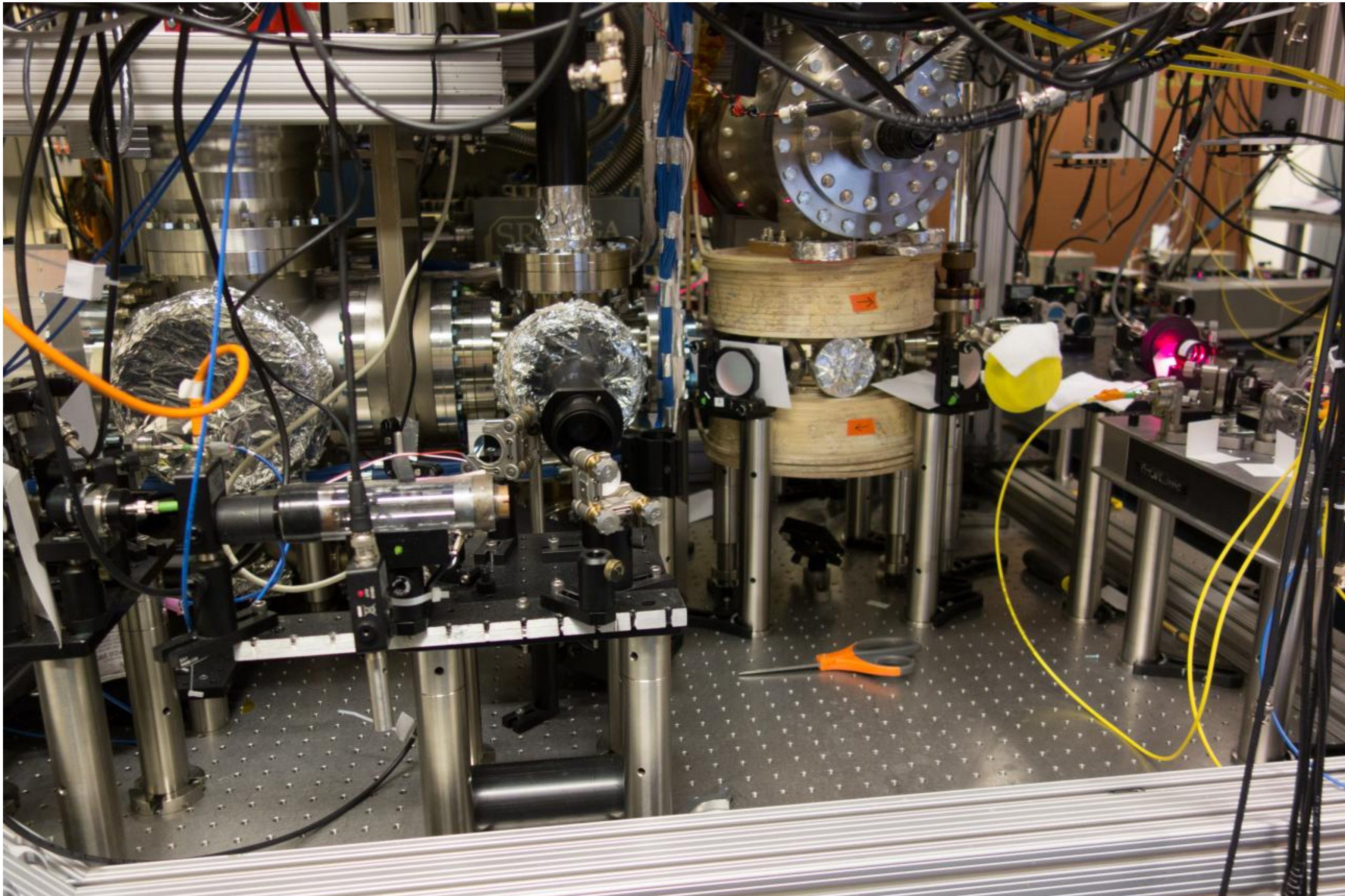


Load slowed beam into a trap

- MOT…?
- Conservative traps w/dissipative loading
  - Magnetic trap
  - Microwave trap?

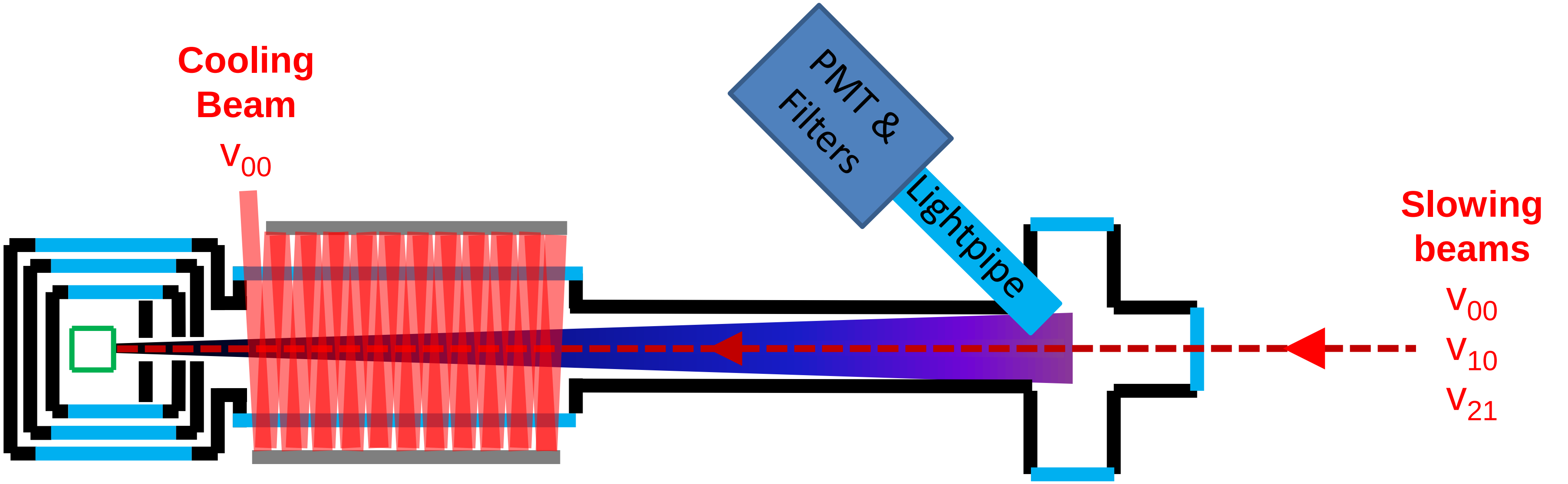
Increase slow flux
for trap loading
w/transverse cooling…?

Underway: attempting to load a 3-D MOT of SrF



How to deliver more slow molecules to a trap?

1. Simultaneous Slowing & Transverse Cooling



Cooling + Slowing = Simultaneously?

Spontaneous forces

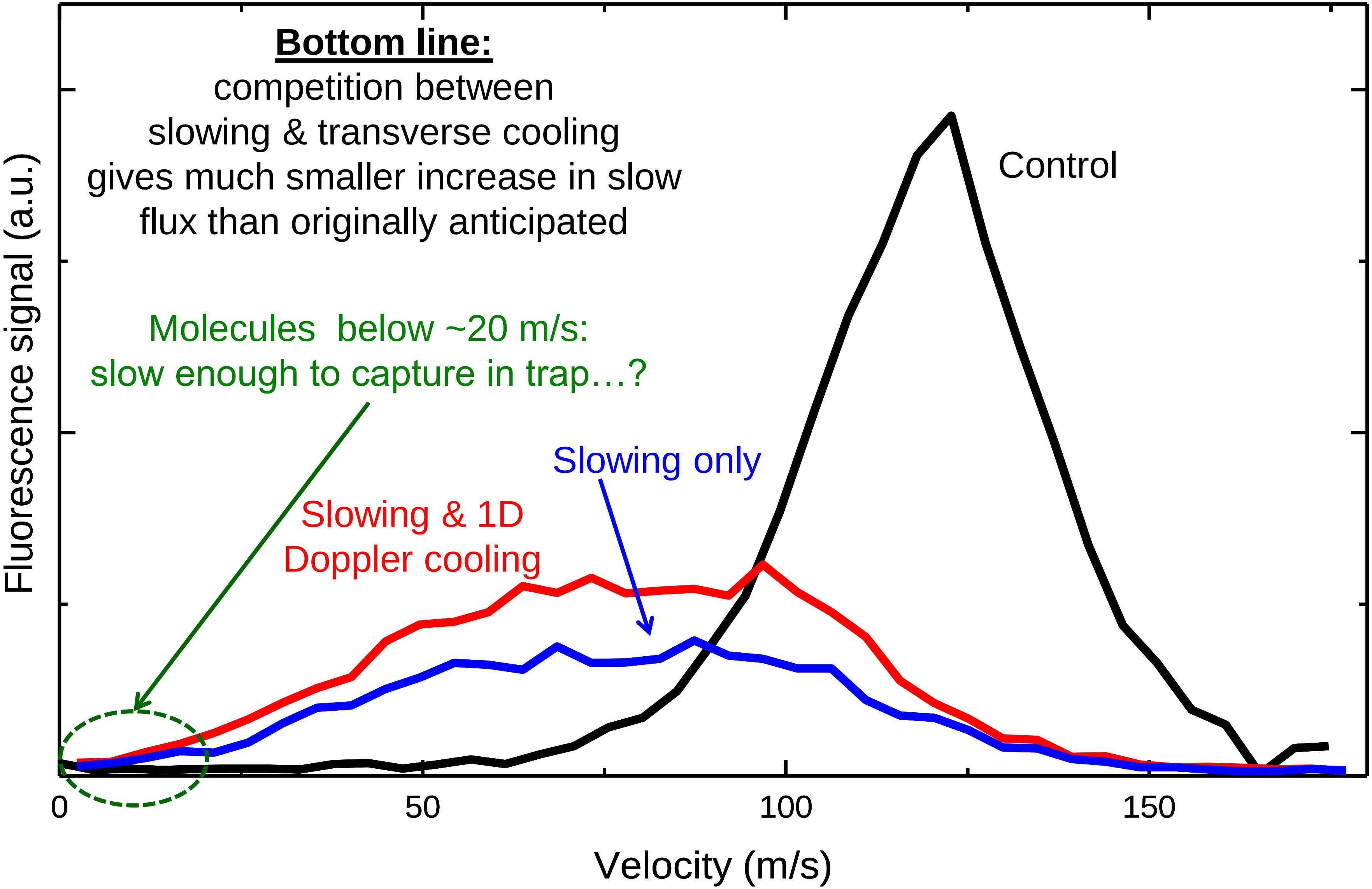
↓

Fixed total spontaneous scattering rate

↓

Forces compete

Preliminary data: Simultaneous slowing + transverse cooling (1D)



How to deliver more slow molecules to a trap?

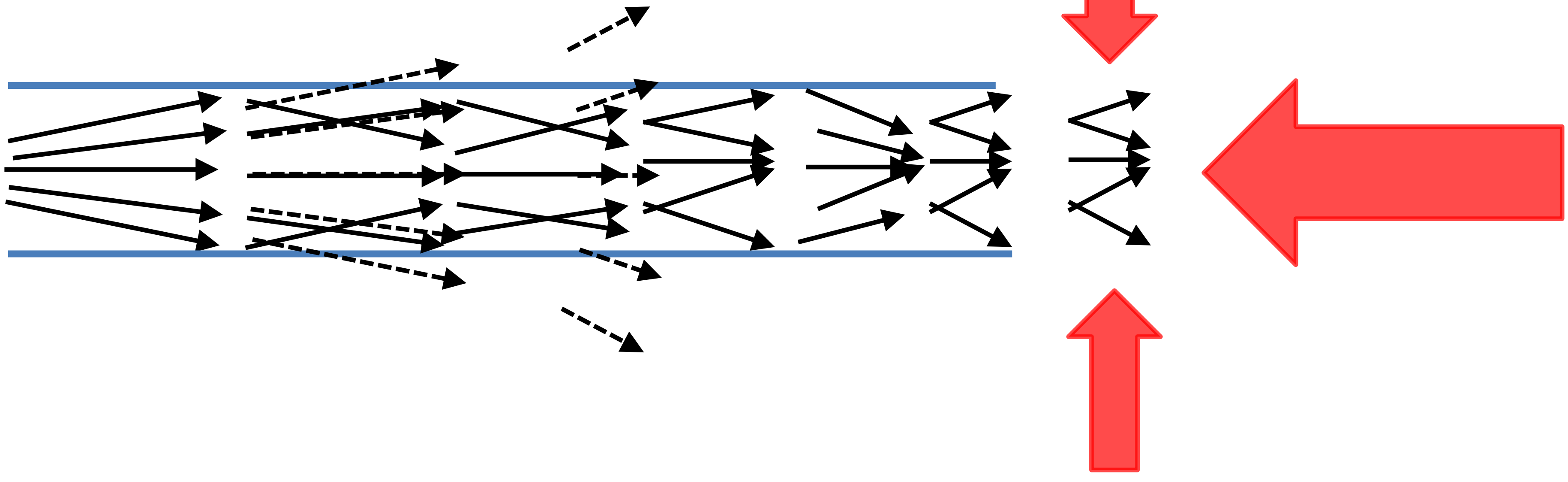
2. Simultaneous slowing & (conservative force) guiding

Problem: beam divergence during slowing

Basic approach to mitigate:

Transverse restoring force while slowing
can confine molecules without loss

THEN transverse cooling can be applied
more effectively with already-slow molecules

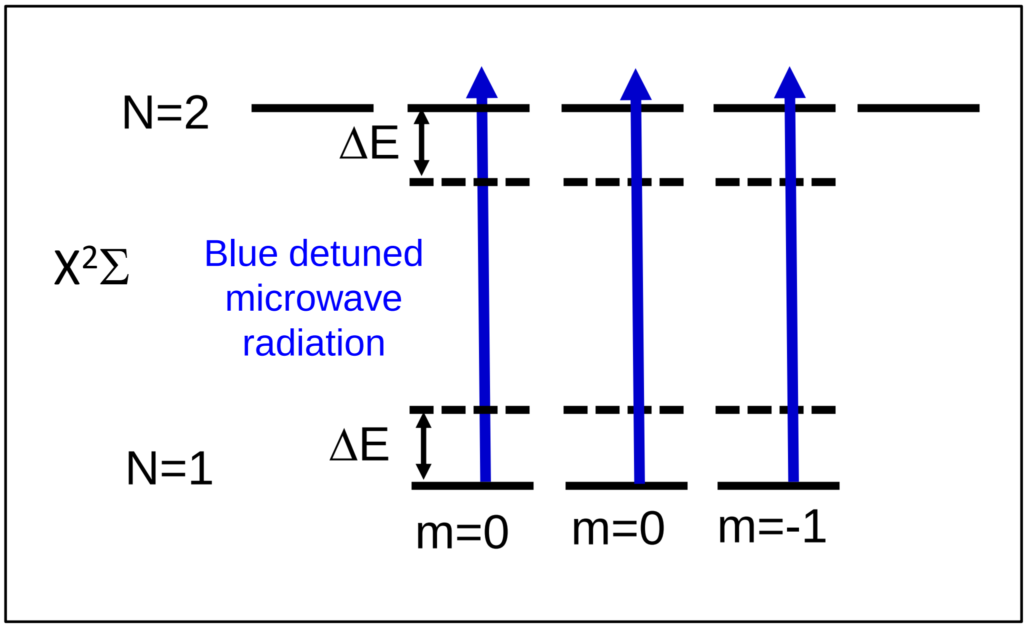


How to deliver more slow molecules to a trap?

2. Simultaneous slowing & (conservative force) guiding

Technical approach:

Microwave Guiding



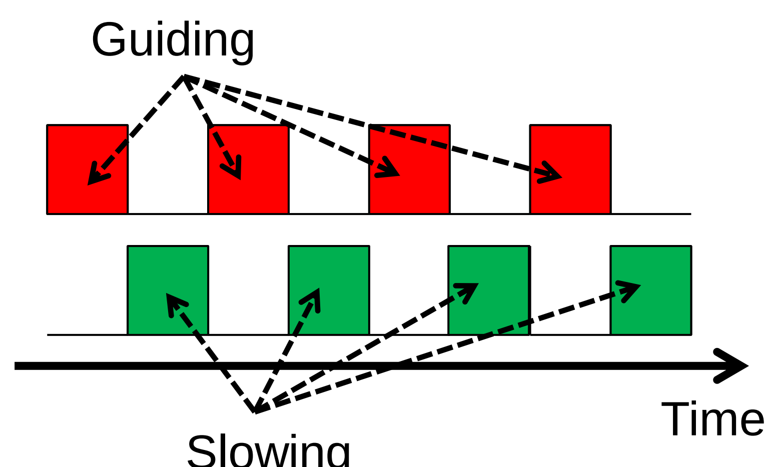
(Inspried by Meijer/Schnell *et al.* result w/ND_3: S. Merz *et al.* PRA 2012)

All N=1 sublevels guided into field minimum (N=1 states: low field seekers)

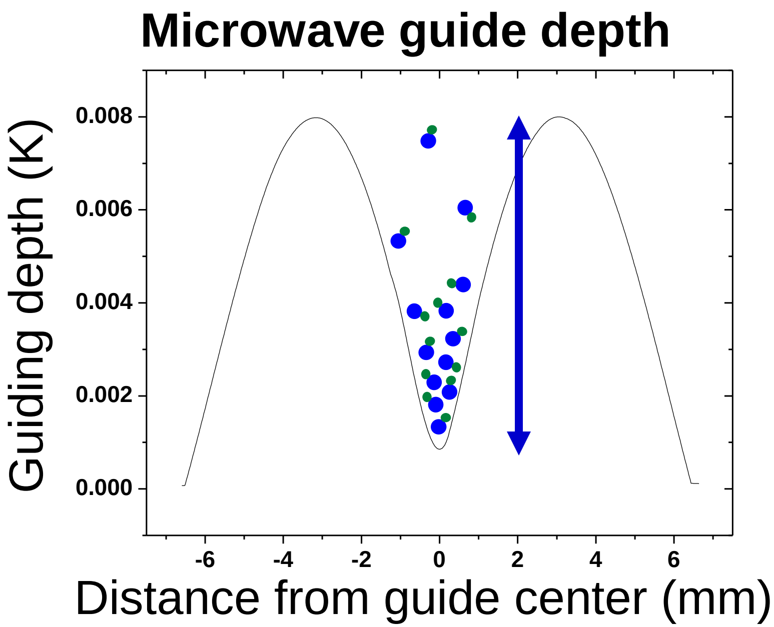
All sublevels feel ~equal potential depth

➔State redistribution from cycling ***no problem for guiding***

Slowing & microwave guiding to be applied alternately in time (guiding fields leaks in cycle)



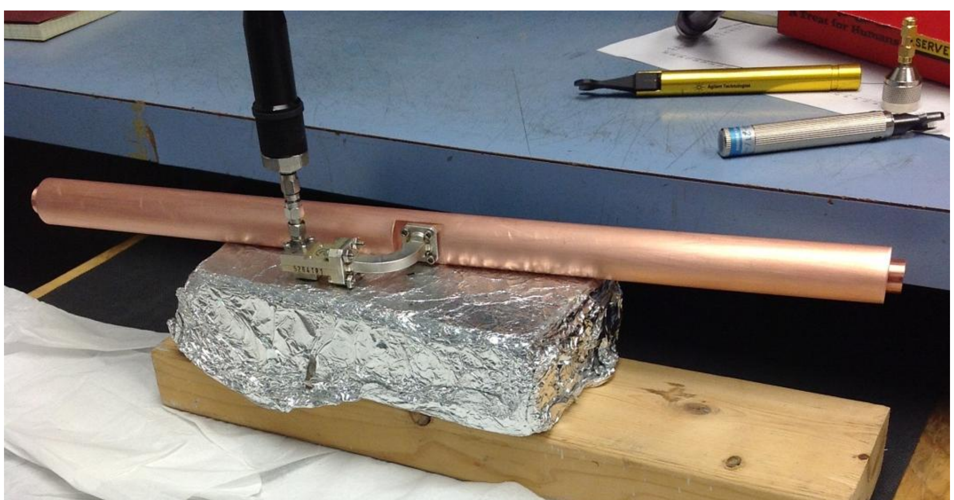
Details of the microwave guide design



Use TE_{01n} mode:
(zero field at guide center)

~ 10 mK guide depth (@ 10 Watts circ. Power)
(~ 1 m/s trans. velocity)

Guide depth ➔ power limited



Prototype (short) cavity characteristics:

Unloaded Q ~ 18000

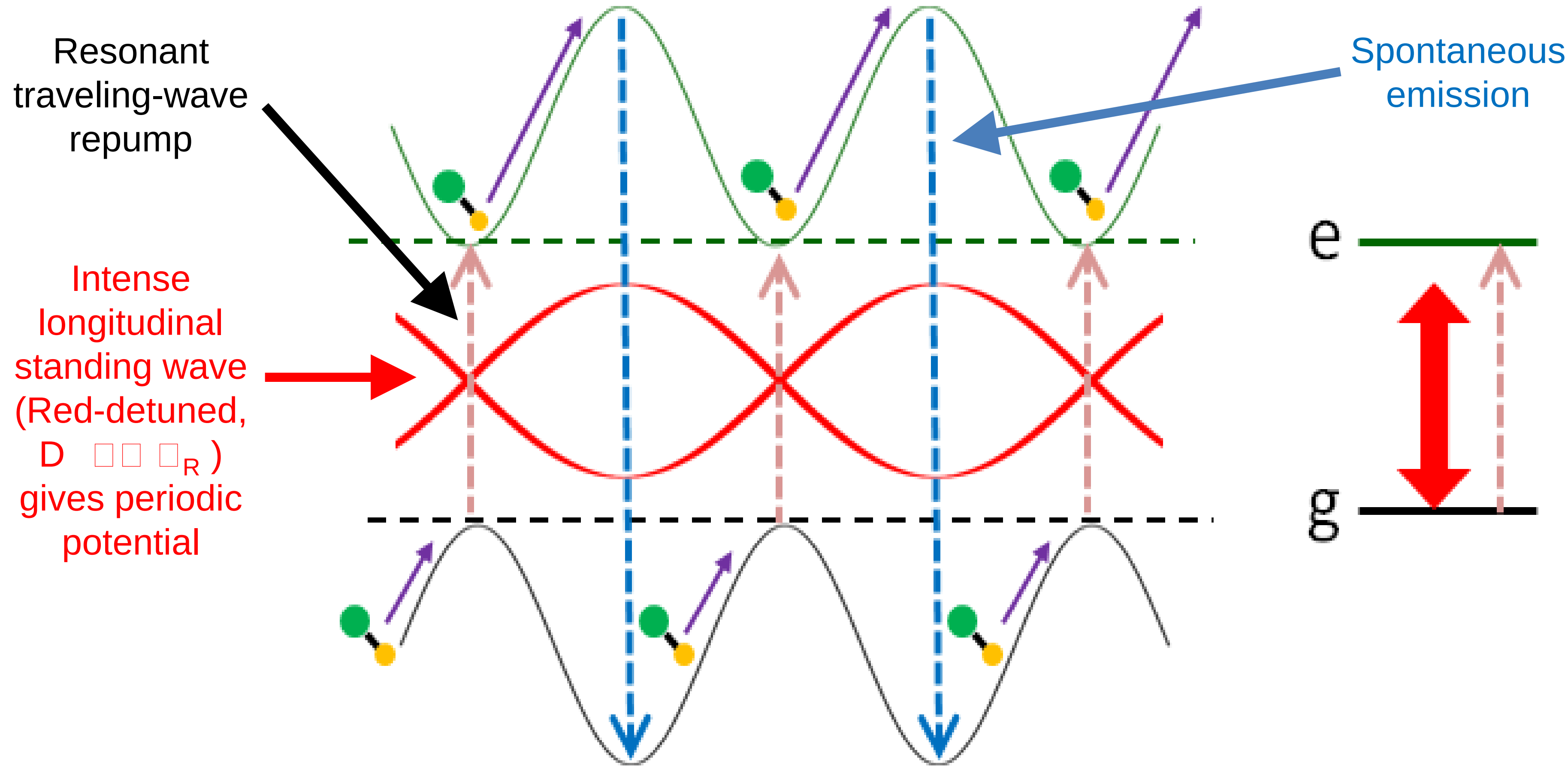
Frequency = 30.150 GHz (N=1→N=2)

Laser slowing w/reduced photon scatter?

Enhanced optical forces from stimulated/coherent processes:

Some demonstrated & more proposed e.g.

Bichromatic Force slowing [Grimm, Metcalf, Eyler]; Optical Stark deceleration [Barker]; "Forced Sisyphus slowing" [DD *et al*., in preparation]



BUT: *all* need high laser intensity over large area for efficient use of molecules

Laser slowing w/reduced photon scatter?

Scaling behavior for stimulated forces & use w/pulsed beams leads naturally to desire for a new laser technology:

- With optimal detuning D » W_R , Force F µ W_R µ $\sqrt{I}$
- Over time *T*, change in velocity dv µ $\sqrt{IT}$ µ $\sqrt{ET / A}$

where *E* = laser energy felt by molecules in beam of area *A* in time *T*

□ ***Optimal laser has long pulses w/high energy, large area***

Example: *E* = 100 mJ @ *T* = 100 ms, A = 1 cm^2 → F_{stim} » 100 ´ F_{spont}

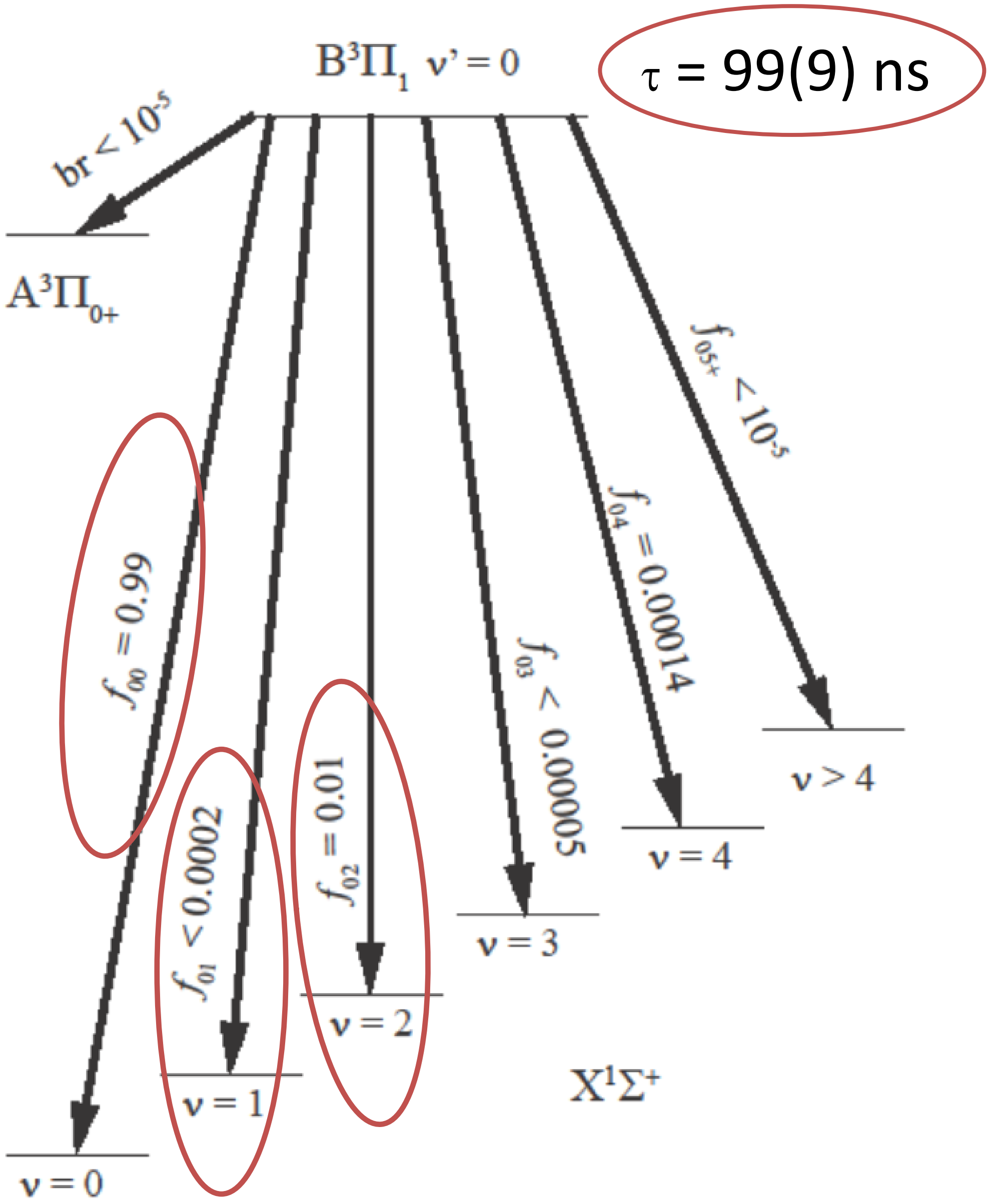
(n.b. instantaneous power is *P* = 1 kW)

□*Reduce required number of scattered photons by >100*□

Construction underway on novel
long-pulse, high pulse-energy tunable laser

A new candidate for laser cooling: TlF

w/Larry Hunter, Amherst College: L.R. Hunter *et al.*, PRA **85**, 012511 (2012)



- Favorable FC factors calculated & measured values in agreement for B-X
- Measured lifetime short enough to give substantial force
- Cycling laser + 1 repump enough for transverse cooling…
- *BUT* UV wavelengths (270-280 nm) needed
- 2-3 repumps sufficient for radiative slowing
- **Possible application to Schiff moment (□ ”nuclear EDM”) search**
- Chemically distinct from other laser-cooling species (NOT a free radical)

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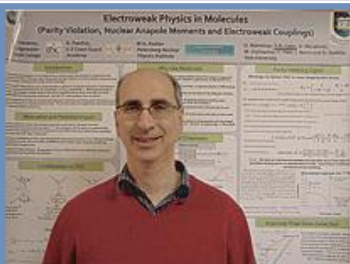

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Summary and Conclusions

– 1-D transverse laser cooling of SrF demonstrated; Doppler and Sisyphus effects observed; [Shuman *et al.*, Nature 2010] qualitative agreement w/expectations, T_D ~ 5 mK, T_S ~ 300 μK

– Possible application to bright molecular beams for precision measurements (electron EDM, parity violation)

– Longitudinal slowing demonstrated [Barry *et al.*, PRL (2012)]

– Beam divergence during slowing limits useful flux to trap: efforts underway to improve by transverse guiding (microwave)

– SrF has properties (unpaired electron spin, simple hfs, efficient cycling detection) useful for quantum information & simulation

– Basic method applicable to several other species

– Possibility to efficiently slow species with poorly-closed cycles, using stimulated forces?