

Explore New Physics Beyond the Standard Model by Precision Spectroscopy of Heavy Molecules

Yan Zhou

University of Nevada, Las Vegas

ESNT Workshop, Saclay

2025.6.27



Acknowledgement

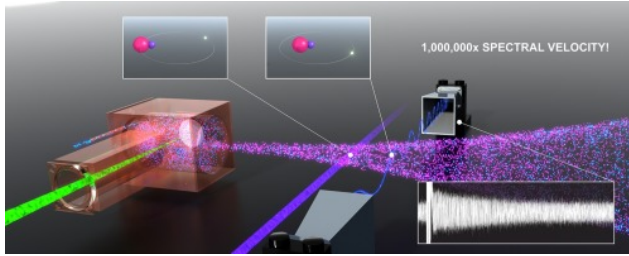
- Profs. Garcia-Ruiz and Field (MIT)
- NIST ion storage group
- Prof. Grau (ODU)
- Prof. Jayich (UCSB)
- Students at UNLV
 - Rodrigo Fernandez – PhD
 - Jose Mosquera Ojeda – PhD
 - Xuanyi Wu – PhD
 - Govinda Bhandari – PhD
 - Stephanie Letourneau – PhD
 - Bernardo Gutierrez – MS
 - Jiaqi Li – visiting PhD



If you plan to visit Las Vegas, please let me know.

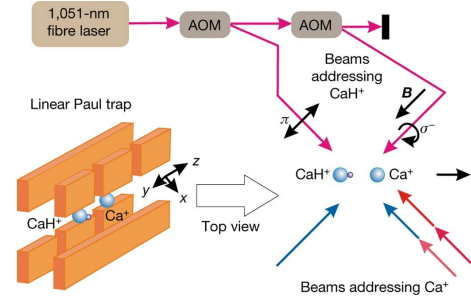
My research experience

Rydberg spectroscopy of BaF



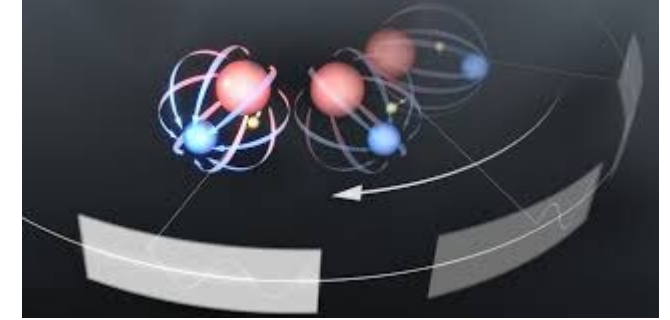
Field's group at MIT

Quantum Logic Spectroscopy



NIST Ion storage group

eEDM measurements

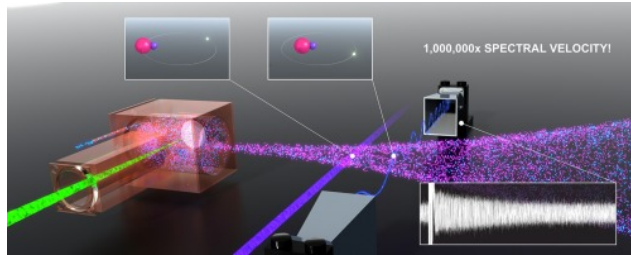


JILA, CU Boulder

- Spectroscopy of small molecules
- Experimental AMO physics
- Numerical modeling in precision metrology

Current research topics

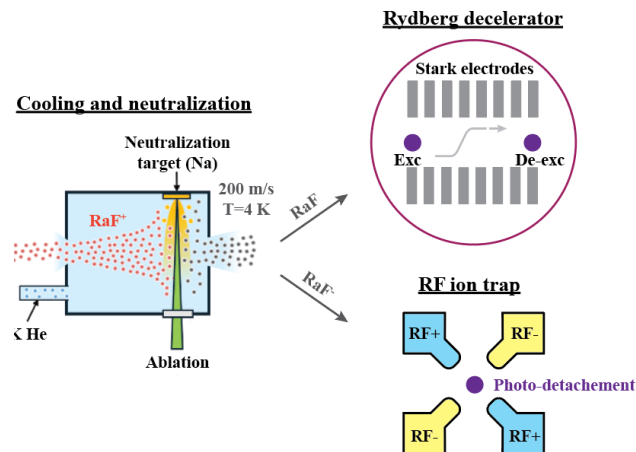
Rydberg spectroscopy of BaF



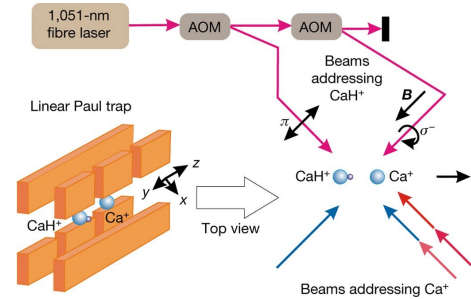
Field's group at MIT



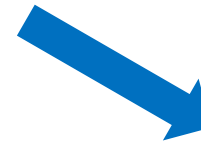
Deceleration of BaF/RaF



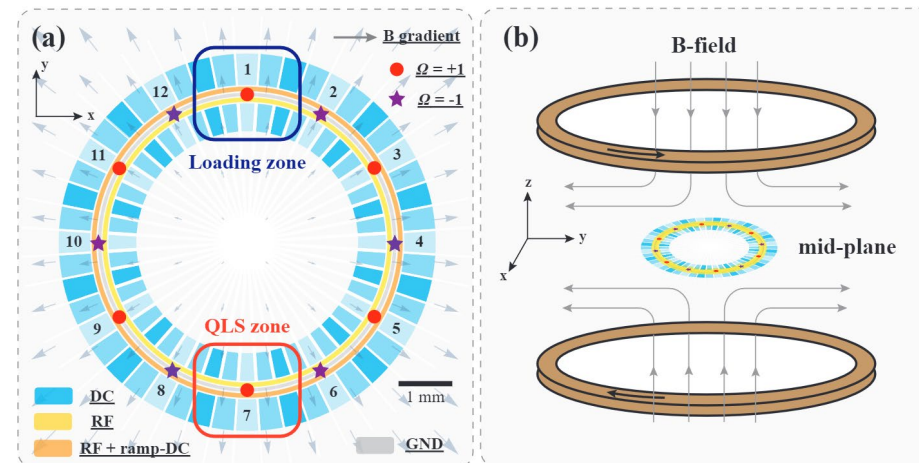
Quantum Logic Spectroscopy



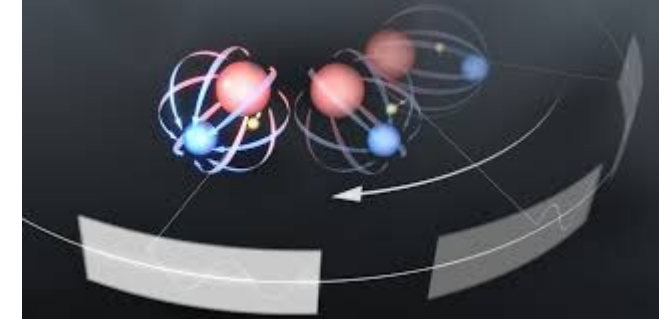
NIST Ion storage group



Molecular ions



eEDM measurements



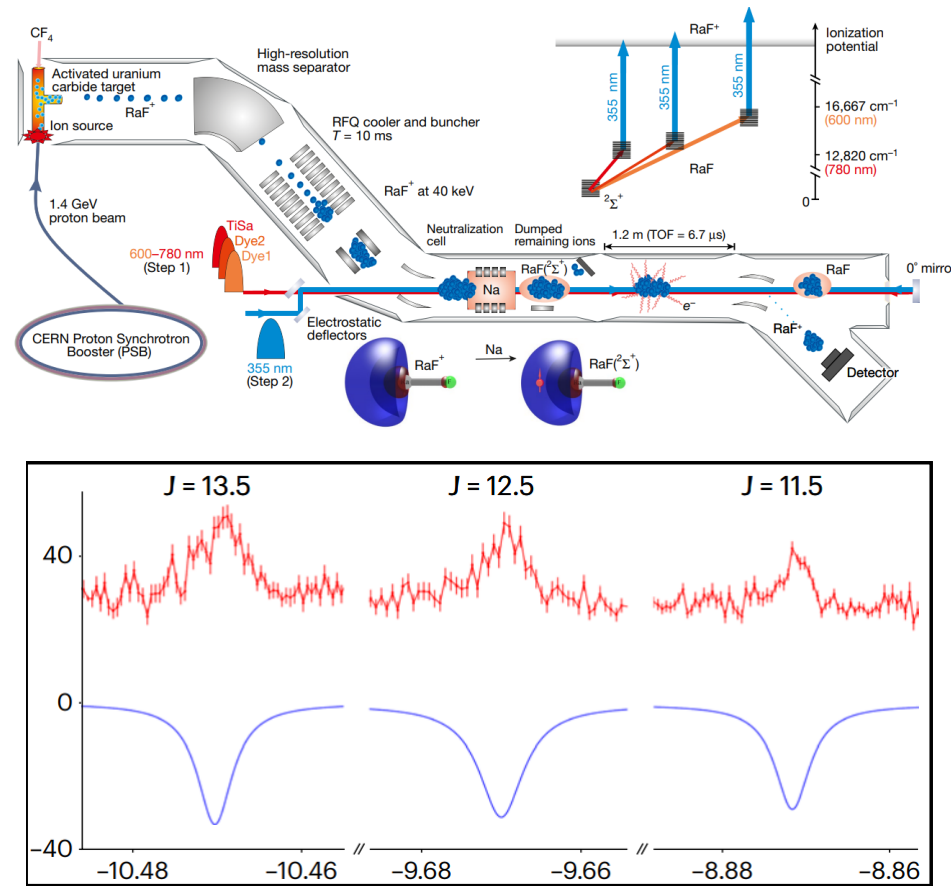
JILA, CU Boulder

Deceleration of BaF/RaF

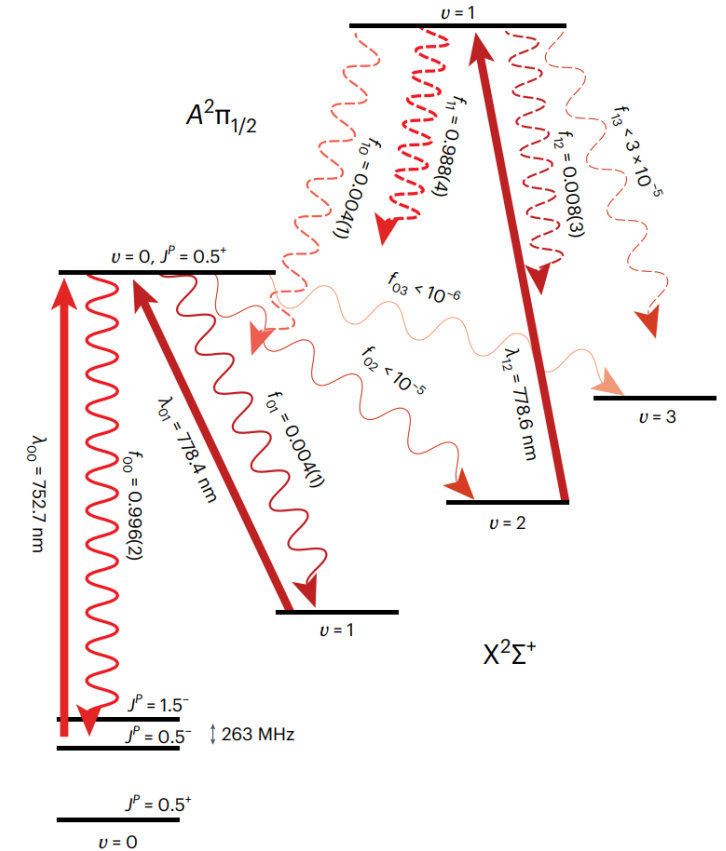
Pear-shape nuclear deformation



High-resolution spectroscopy



Laser cooling and trapping

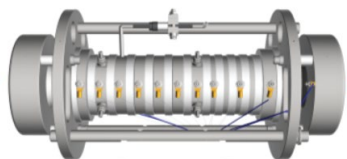


S. M. Udrescu, et., al. Nature Physics, 2024

Deceleration of BaF/RaF

Nuclear physics facilities

RIB Facilities
production



Cooler and
buncher

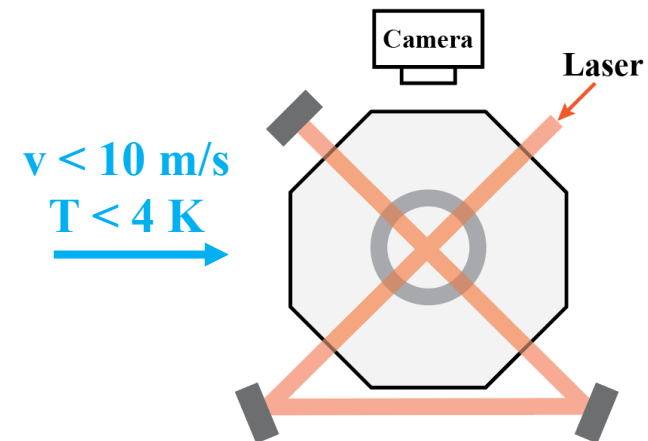
$v = 1000 \text{ m/s}$

$T = 300 \text{ K}$



Ra-225, $\tau \sim 15 \text{ days}$

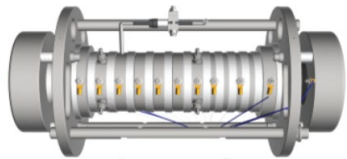
Table-top



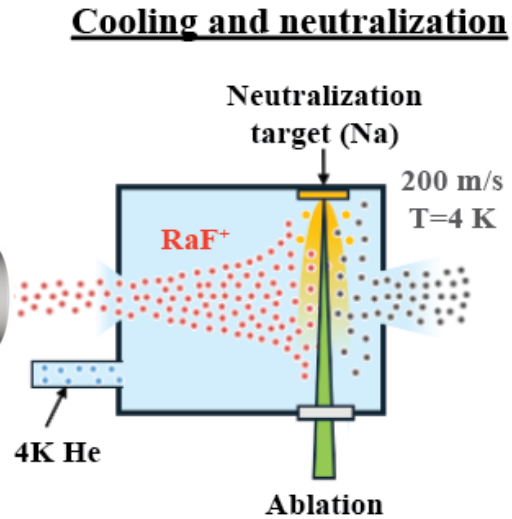
Deceleration of BaF/RaF

Nuclear physics facilities

RIB Facilities
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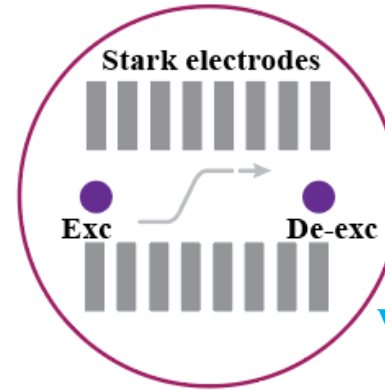


Cooler and
buncher



Ra-225 , $\tau \sim 15$ days

Rydberg decelerator



$v < 10\text{ m/s}$
 $T < 4\text{ K}$

RF ion trap

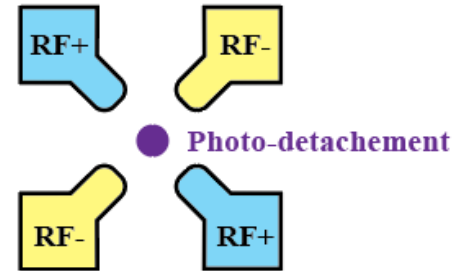
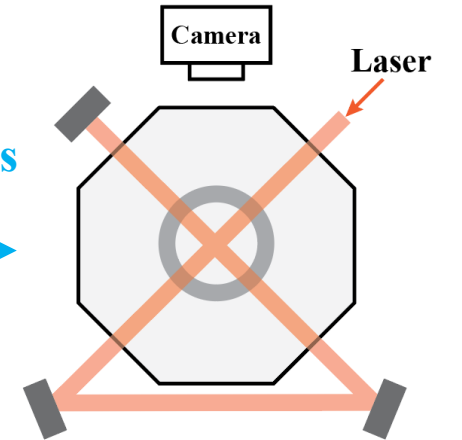


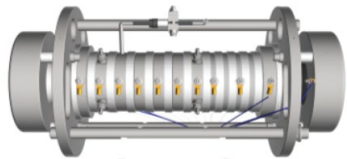
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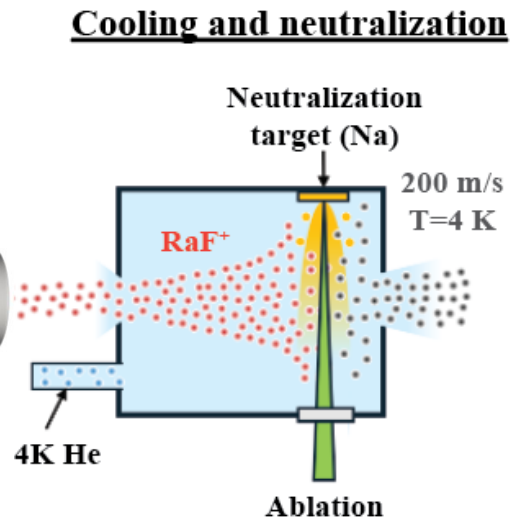
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Nuclear physics facilities

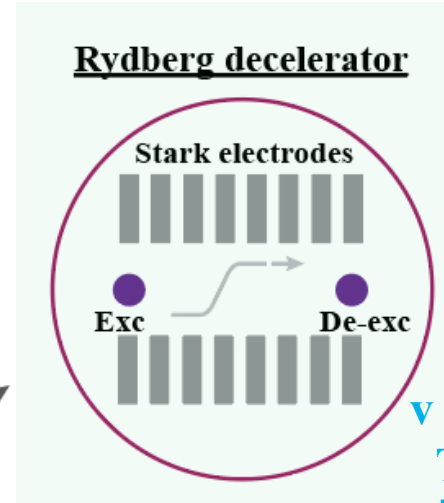
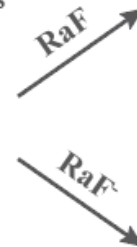
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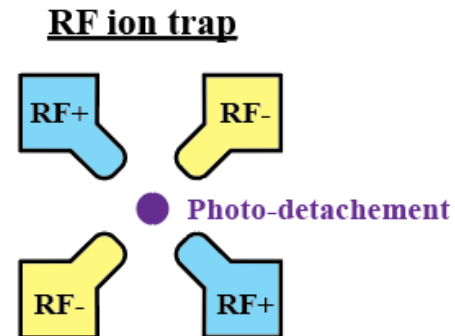
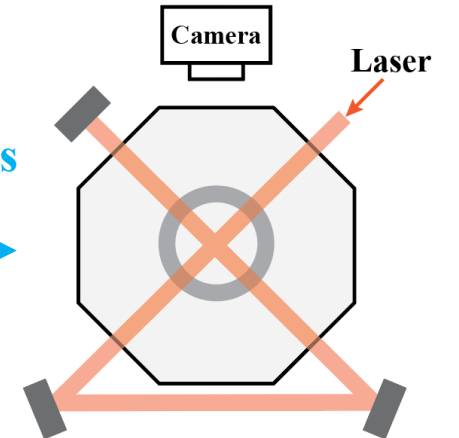


Table-top



BaF vs RaF

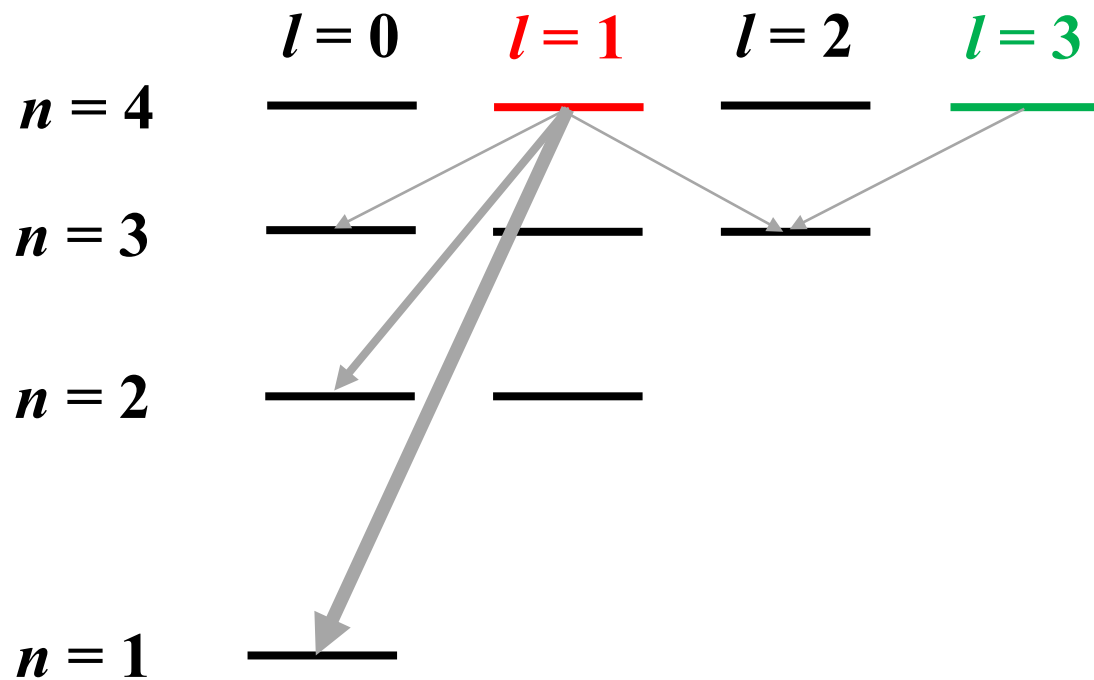
	BaF	RaF
Ground electronic state X	$^2\Sigma^+$	$^2\Sigma^+$
First excited electronic state A	$^2\Pi_{1/2}$	$^2\Pi_{1/2}$
A state lifetime	57.1(3) ns	35(1) ns
$A-X$ $\Delta v = 0$ Franck-Condon factor	~ 0.95	0.996(2)
Electric dipole moment	3.170(3) D	3.91 D
Ionization potential	38745 cm^{-1}	$<41453 \text{ cm}^{-1}$
Dissociation energy	48200 cm^{-1}	$\sim 45000 \text{ cm}^{-1}$
Electron affinity	$\sim 1 \text{ eV}$	$\sim 1 \text{ eV}$
Accessible isotopes	^{134}Ba ($I = 0$), ^{135}Ba ($I = 3/2$) ^{136}Ba ($I = 0$), ^{137}Ba ($I = 3/2$) ^{138}Ba ($I = 0$)	^{223}Ra ($I = 3/2$), ^{224}Ra ($I = 0$) ^{225}Ra ($I = 1/2$), ^{226}Ra ($I = 0$) ^{228}Ra ($I = 0$)
Nuclear deformation	Small	Large
Effective electric field for eEDM	6 GV/cm	50 GV/cm
Nuclear-type EDM	SM and MQM	SM and MQM

Rydberg deceleration

- Pros: large electric dipole moment
- Con: lifetimes, hundreds of states

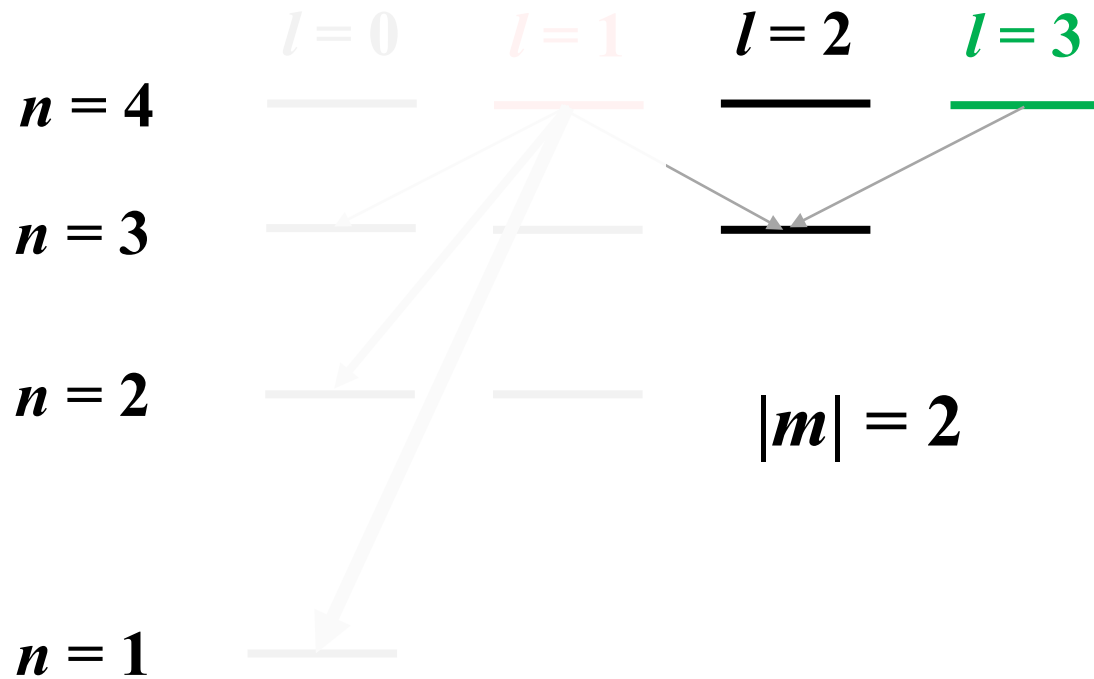
Near-circular states

- Pros: large electric dipole moment
- Con: **lifetimes**, hundreds of states
- Solution: near-circular high- l states (without E-field)



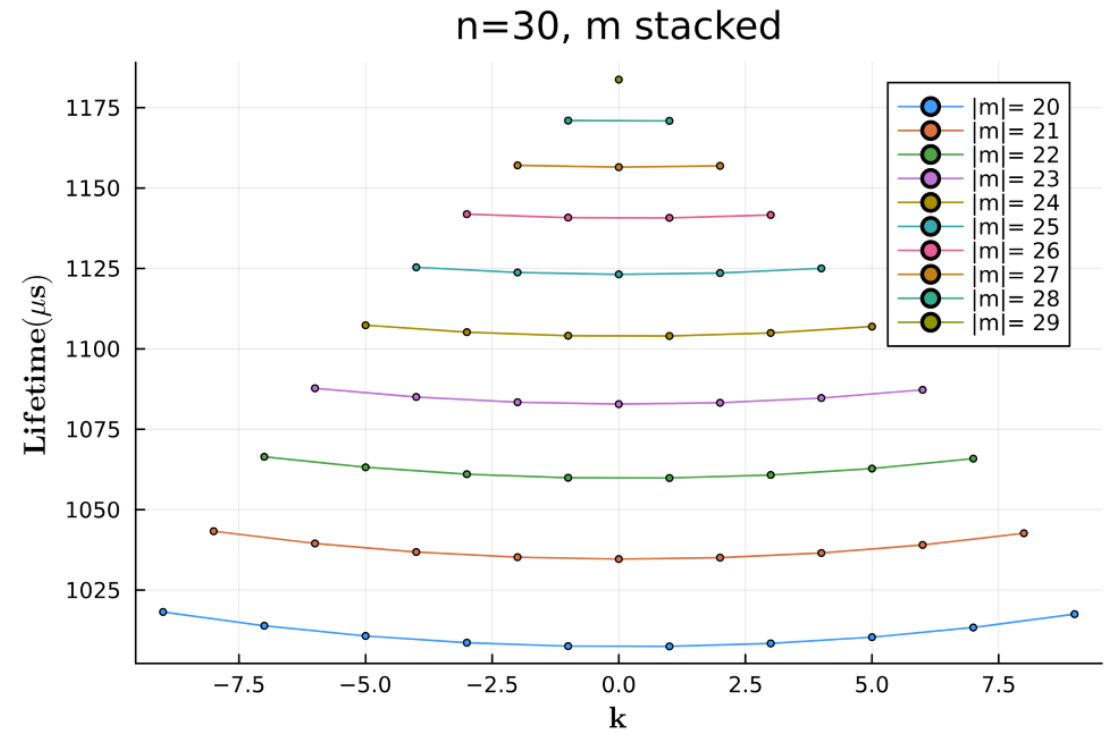
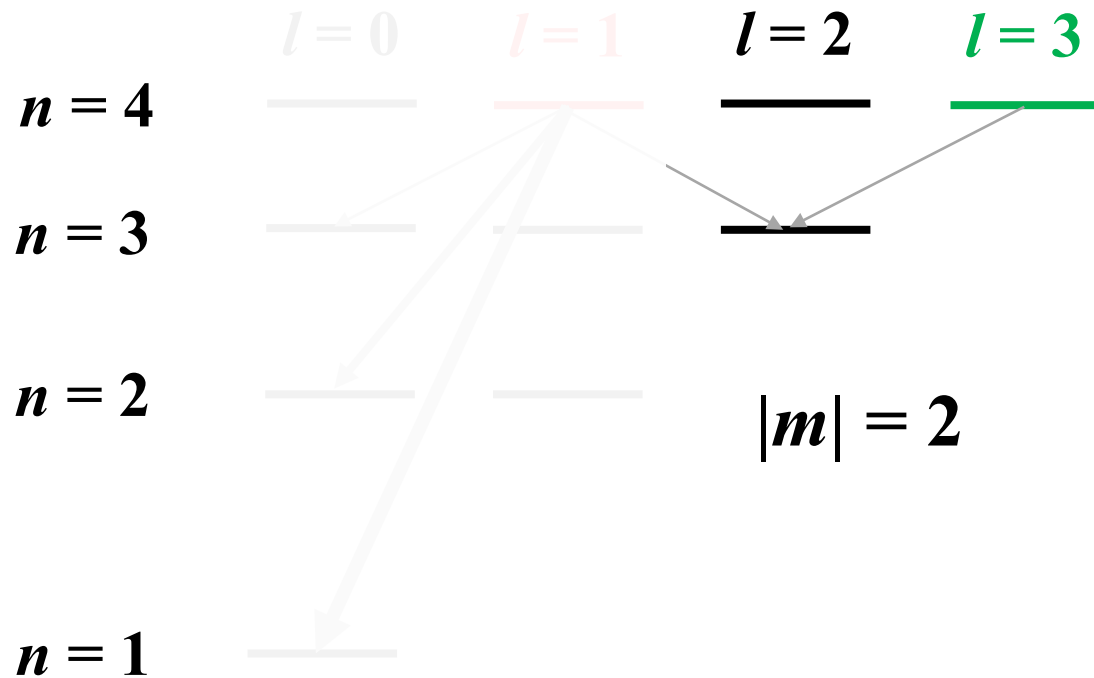
Near-circular states

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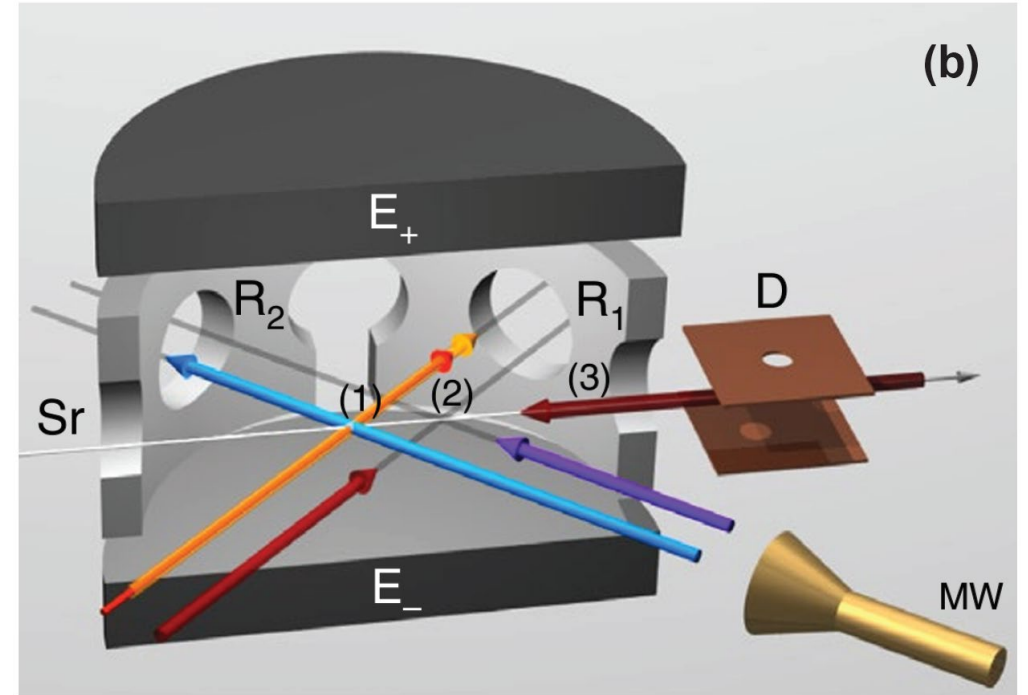
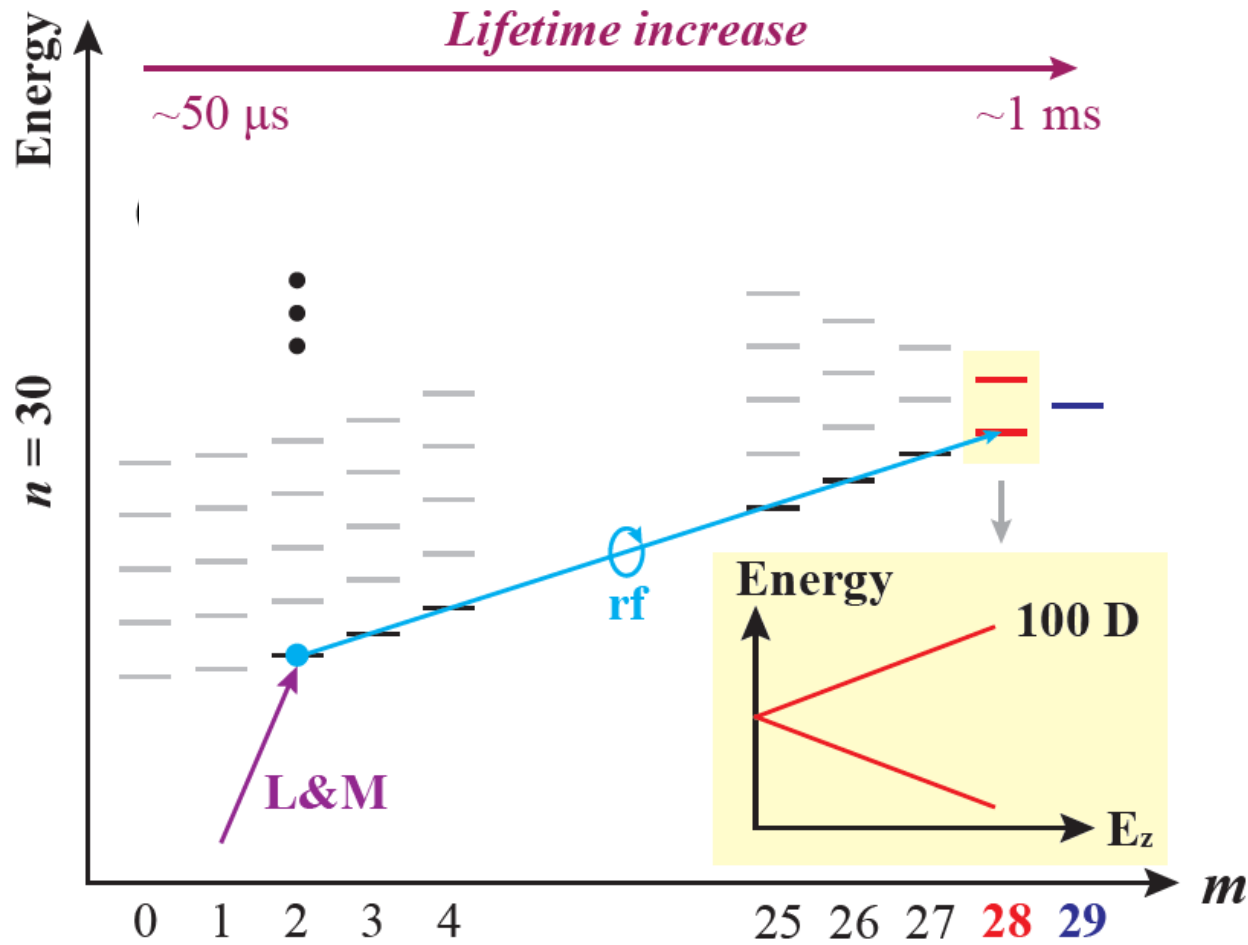
Near-circular states

- Pros: large electric dipole moment
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- Solution: near-circular high- $|m|$ states (with E-field)



20 K, shielding BBR

Prepare near-circular states



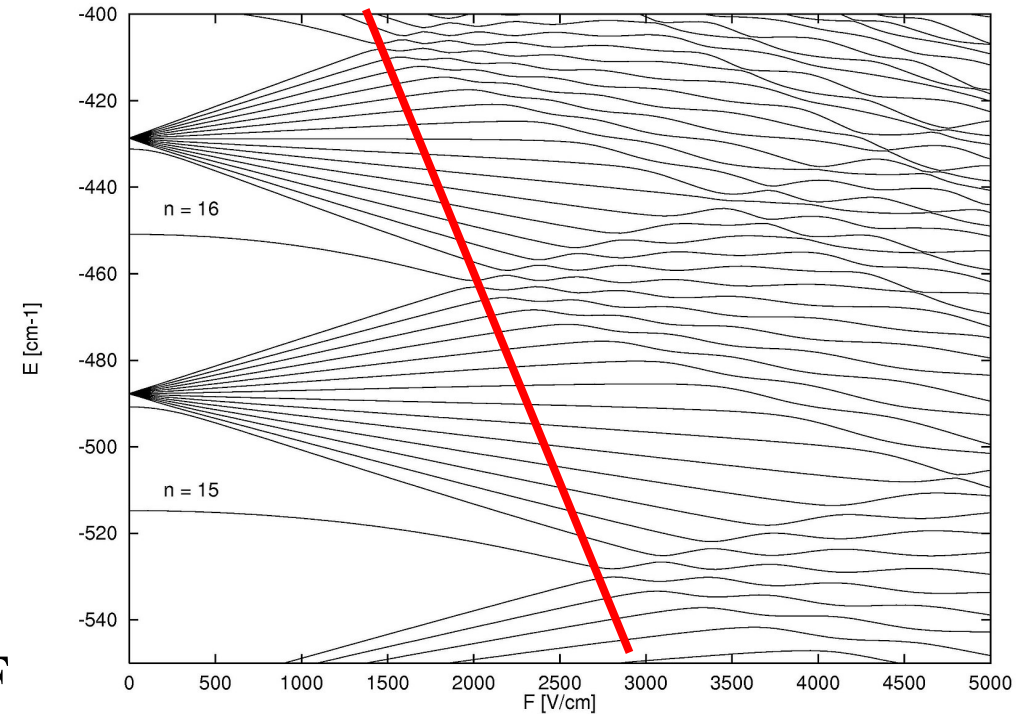
H. Wu, et., al. PRL, 2023

- Laboratoire Kastler Brossel
- Sr atoms
- 97% efficiency

Deceleration rate

- Initial velocity: 200 m/s
- Final velocity: <10 m/s
- Mass of RaF: 244 amu
- Largest electric field
 - Inglis-Teller limit: mixing $\Delta n = 1$
 - Ionization limit: ionizing Rydberg states
 - Typically, **Inglis-Teller ~ 0.1 * Ionization**
- Limited by Inglis-Teller limit, ~ 0.5 ms for RaF
- Limited by Ionization limit, <0.1 ms for RaF

Alkali atoms

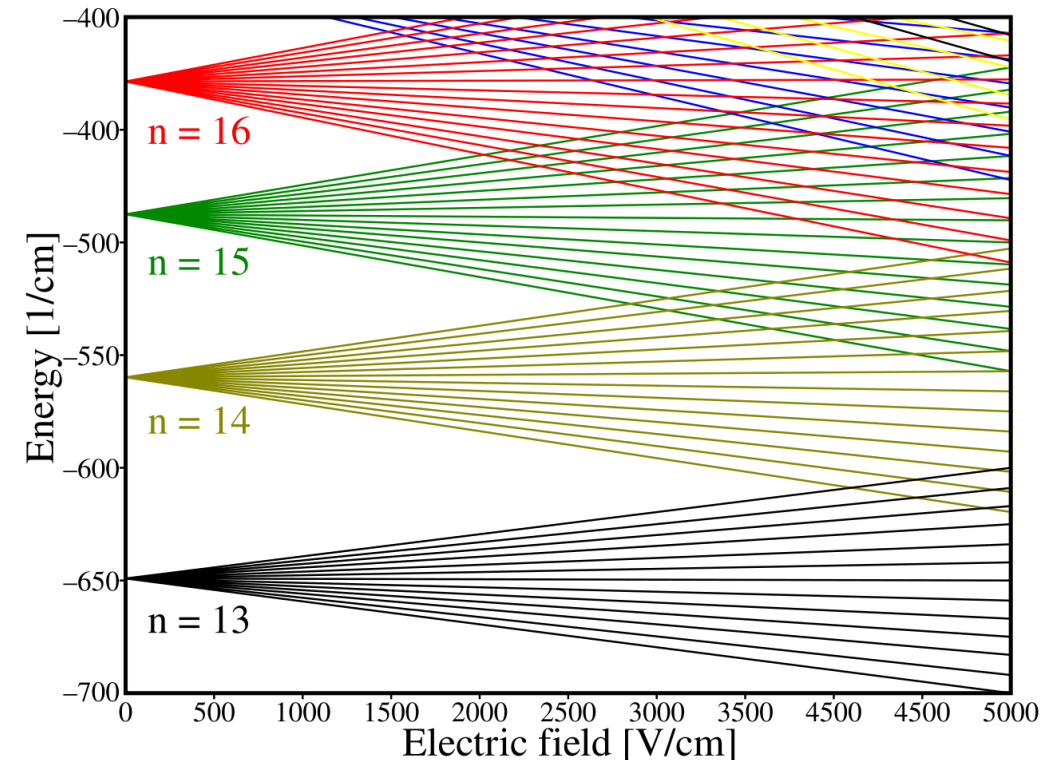


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No avoid crossing!

H atom



Zero quantum defects for all l

Core-penetrating vs. Core-nonpenetrating

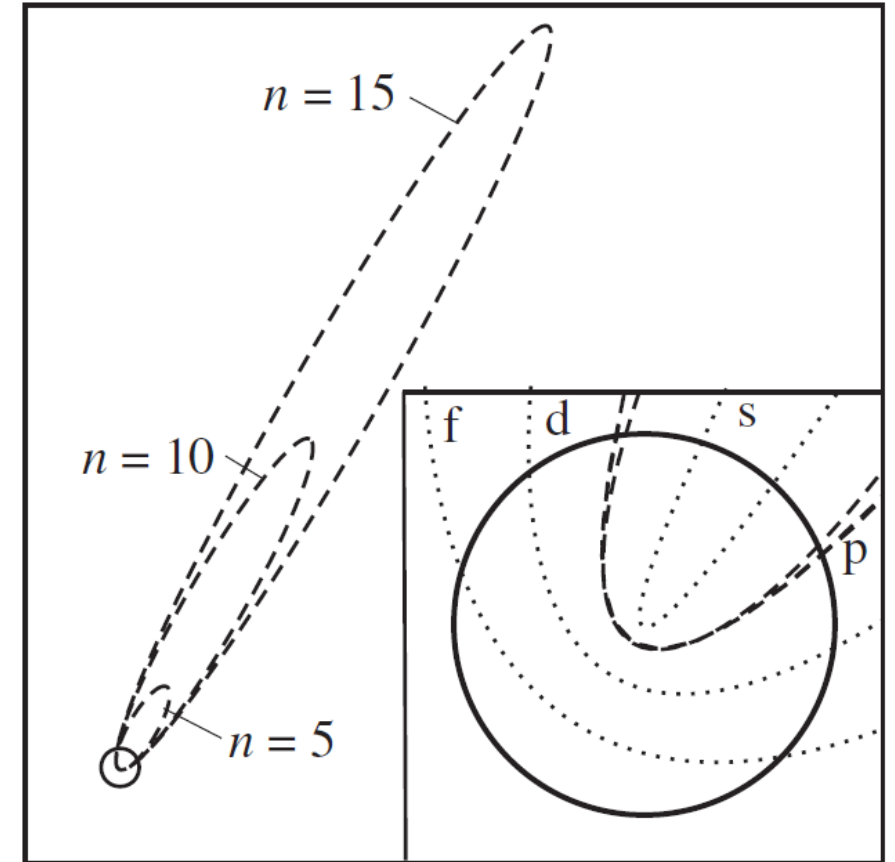
- Determined by the inner-turning point

$$V_l(r_-) = E_{nl}, V_l(r) = -\frac{2}{r} + \frac{l(l+1)}{r^2}, E_{nl} = -\frac{1}{n^2}$$



$$r_- = n^2 \left\{ 1 - \sqrt{1 - \frac{l(l+1)}{n^2}} \right\} \sim l^2$$

- If $r_- < r_{ion}$, core-penetrating (CP)
- If $r_- > r_{ion}$, core-nonpenetrating (CNP)
- Typically, all states with $l > 4$ are CNP
- Near circular states: $n = 30$, $|m| = 28$, l can only be 28 and 29

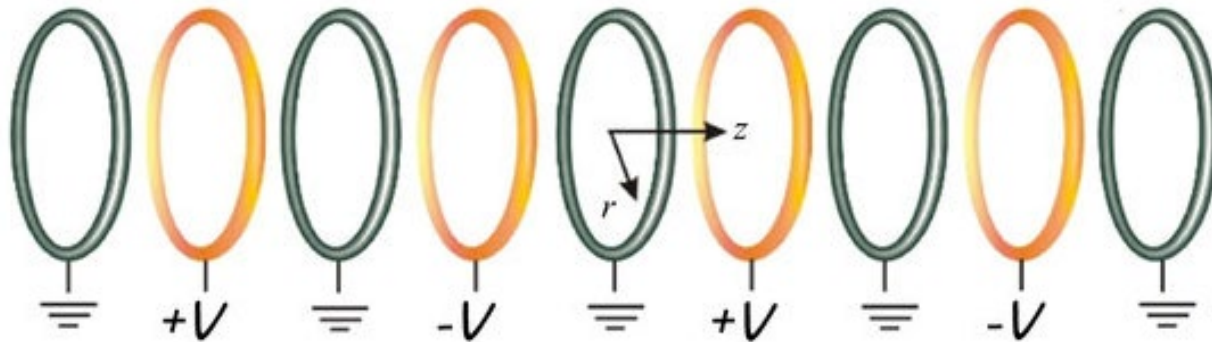


Core-nonpenetrating states

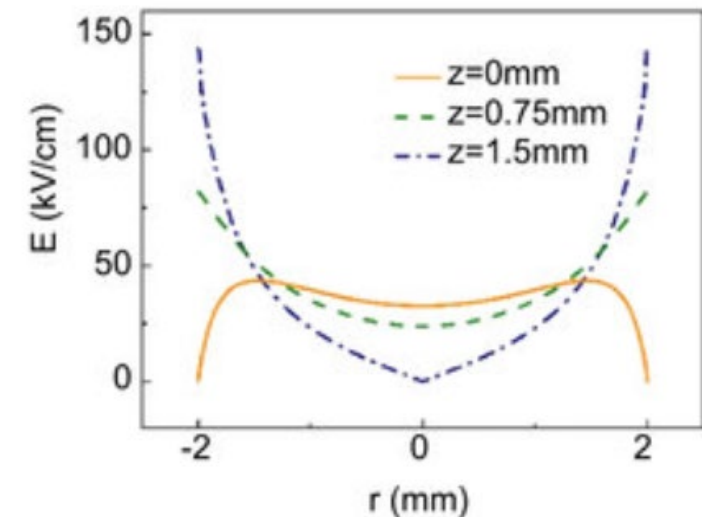
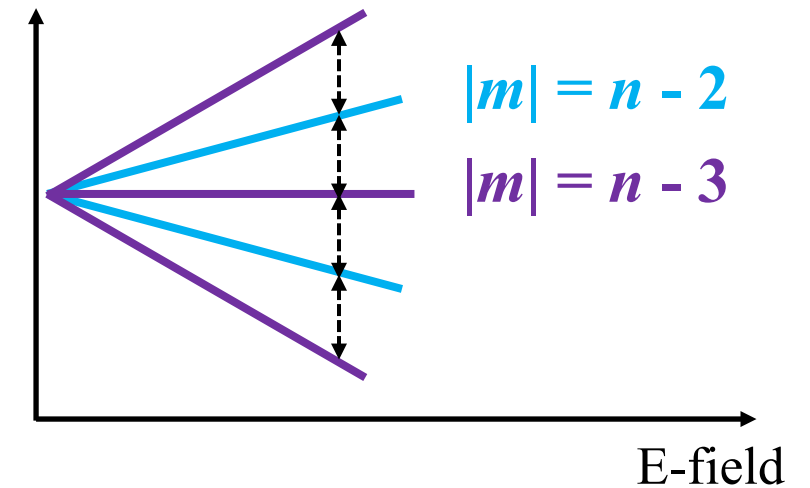
- Do CNP states have zero quantum defects?
 - **Very closed to zero**
 - **Long-range interaction** - multipole expansion and polarizability of the ion-core
 - Analytical expressions (existing)
 - Key parameters can be obtained from *ab-initio* calculations or high-resolution spectroscopy (under working)
- Connections between quantum defects and avoided crossings at Inglis-Teller limit (under working)

Decelerator design

- Minimization of transverse E-field
 - Transverse E-field can mix different $|m|$
 - Rydberg manifolds with different $|m|$ have different energy with large E-field
- Numerical model (under working)

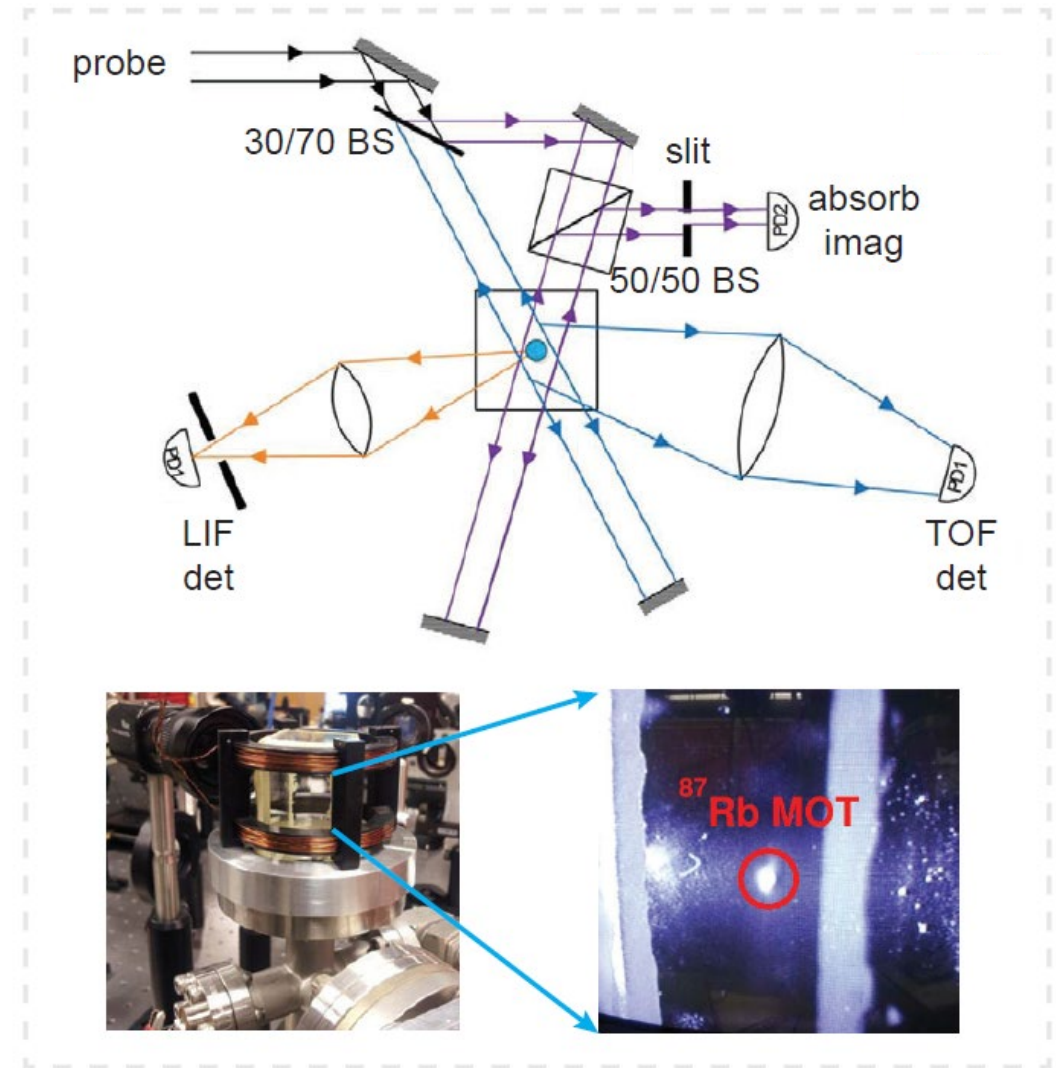


S. Hou, et., al. J. Phys. B: At. Mol. Opt. Phys. 2016



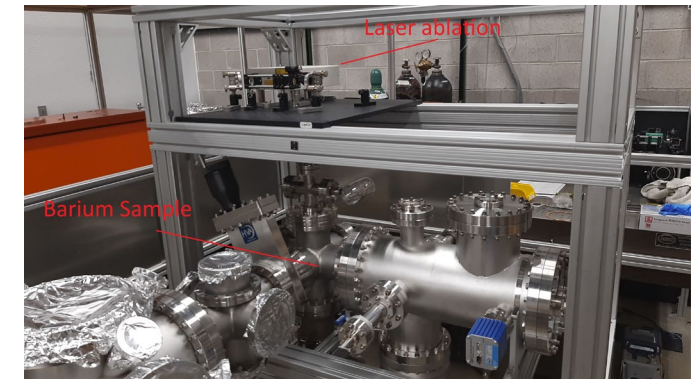
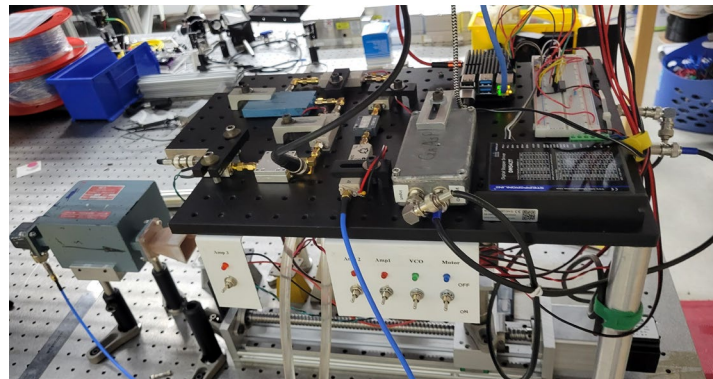
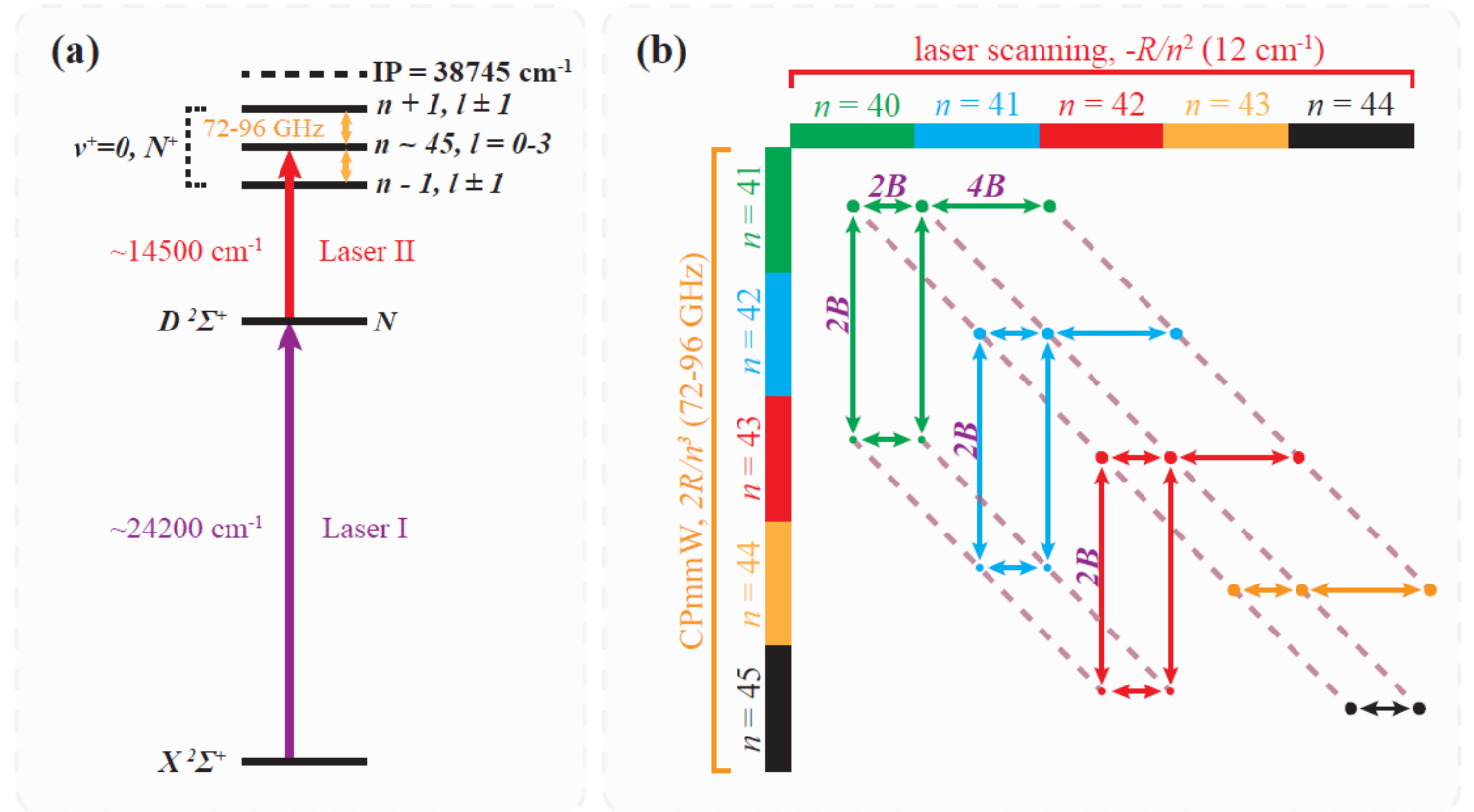
Rb experiments

- Answer most of questions using a simple atomic system
 - Circular Rydberg states preparation
 - Lifetime measurements
 - Dipole moment measurements
 - Quantum control above Inglis-Teller limit
- Current status
 - Rb MOT
 - Ionization detections
 - Circular RF electrodes
 - Cryogenic system (under working)
 - Prototype decelerator (near future)



BaF spectroscopy

- CPmmW spectroscopy
- Spectroscopic pattern recognition
- Long-range model of CNP states – quantum defects
- Intermediate CP states for accessing the CNP states
- A model for evaluating feasibility of Rydberg Stark deceleration

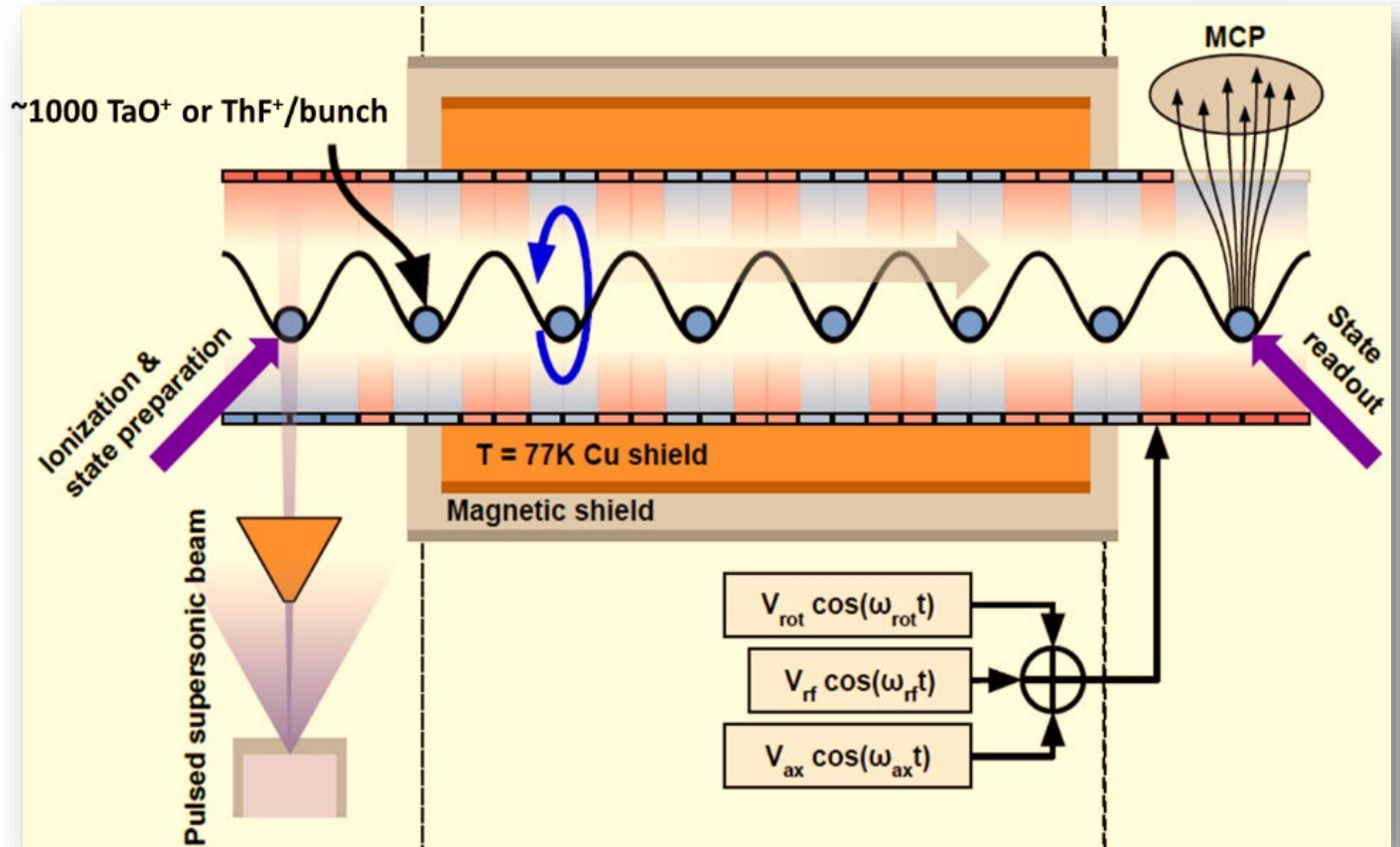


Molecular ions

- **Advantages of molecular ions**
 - Easy to trap for a long time
 - Low sample consumption
 - Sympathetic cooling
- **Challenges of molecular ions**
 - Low density
 - Ion-ion collisions
 - Biased electric field

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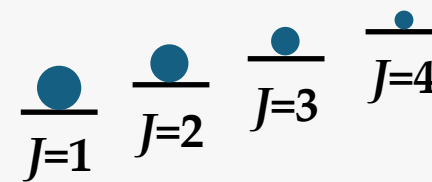
JILA, TRIUMP, Harvard

Opposite direction – single molecules

- Concentrate 100% population to a single quantum state from **hundreds of initial states**
- An efficient state readout with 100% fidelity
- Extremely long coherence time – beyond a minute
- **Universal platform** for different molecular species
- Rare isotopes – minimum amount of sample
- Repeat measurements of a single ion
- Chip-scale of the experimental device – microfabricated ion trap
- Scalability – highly parallel measurements
- Minimum systematics – inherit reference, quantum sensors

State preparation and detection

$^{232}\text{ThF}^+$

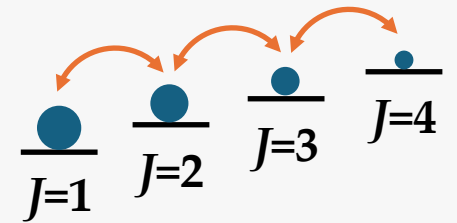


$^3\Delta_1, v=0$

State preparation and detection

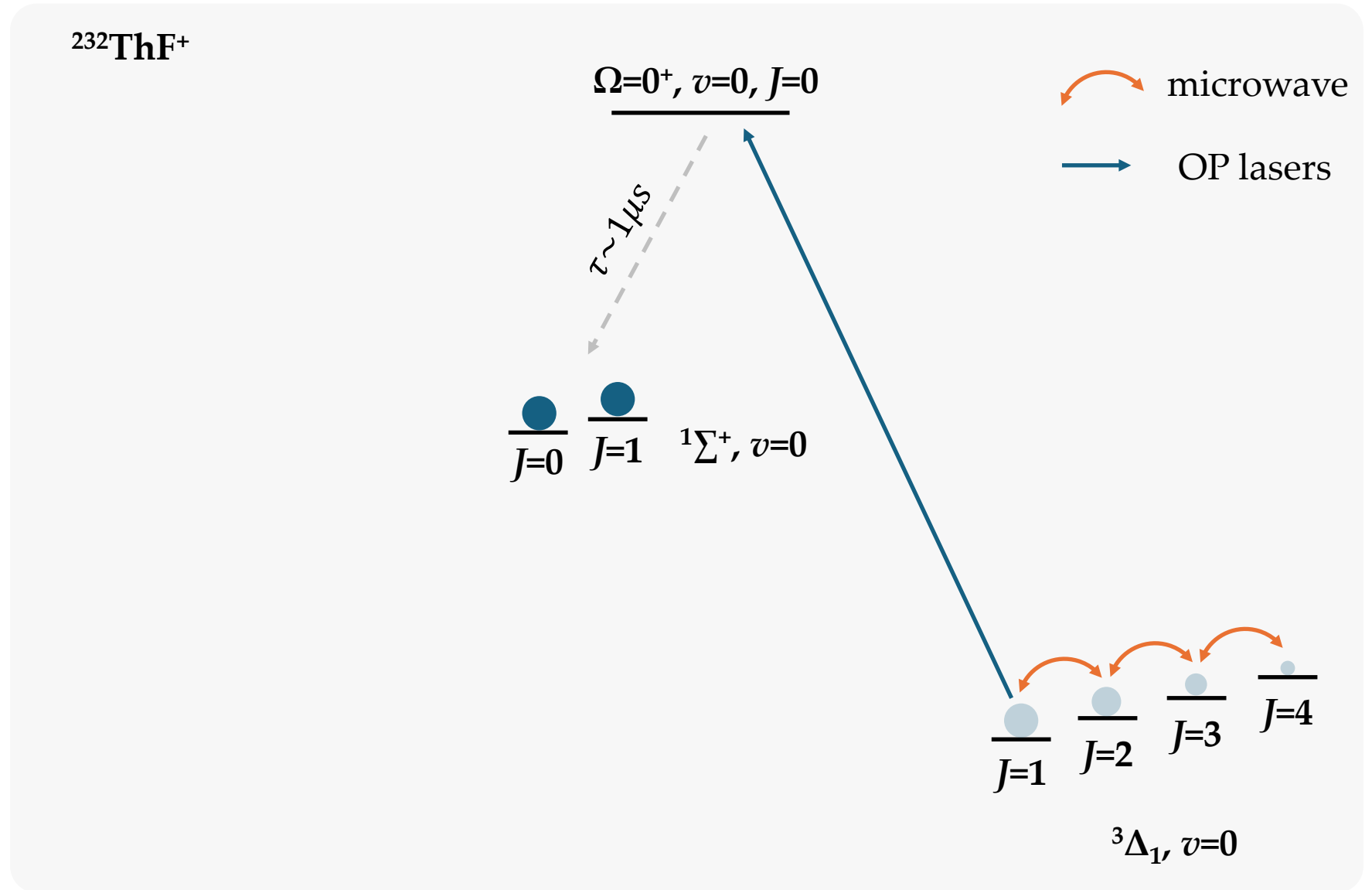
$^{232}\text{ThF}^+$

 microwave

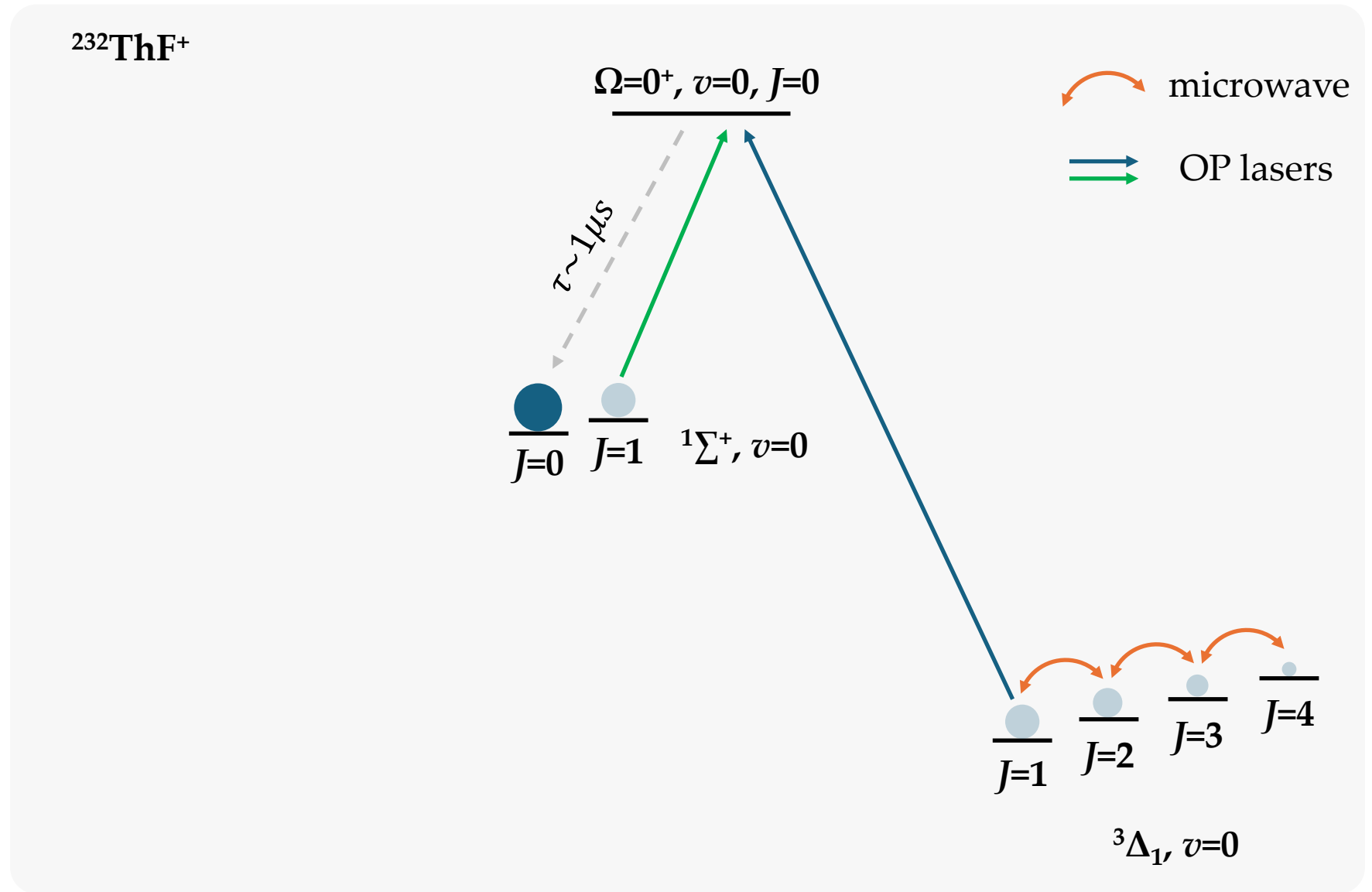


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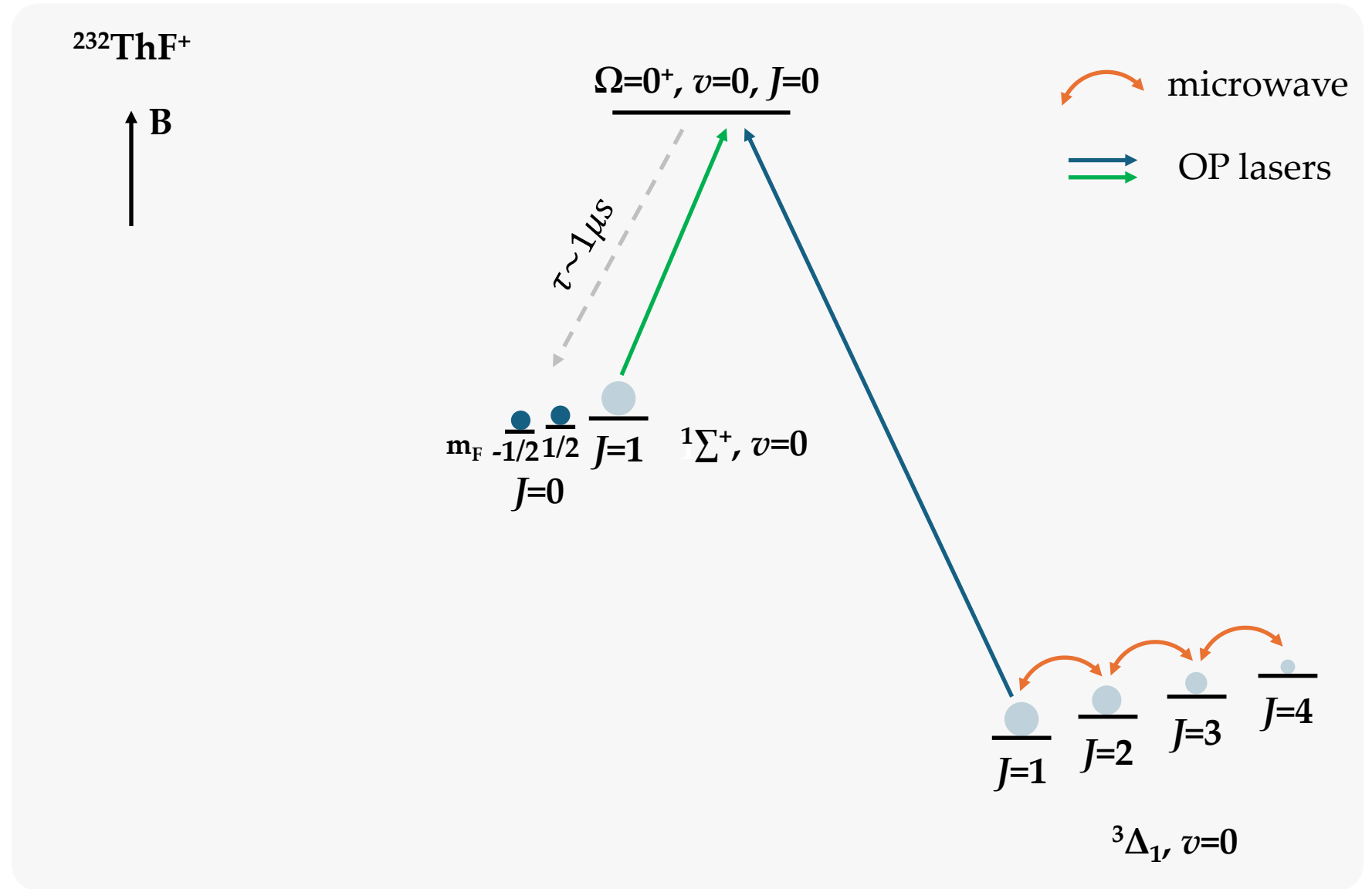
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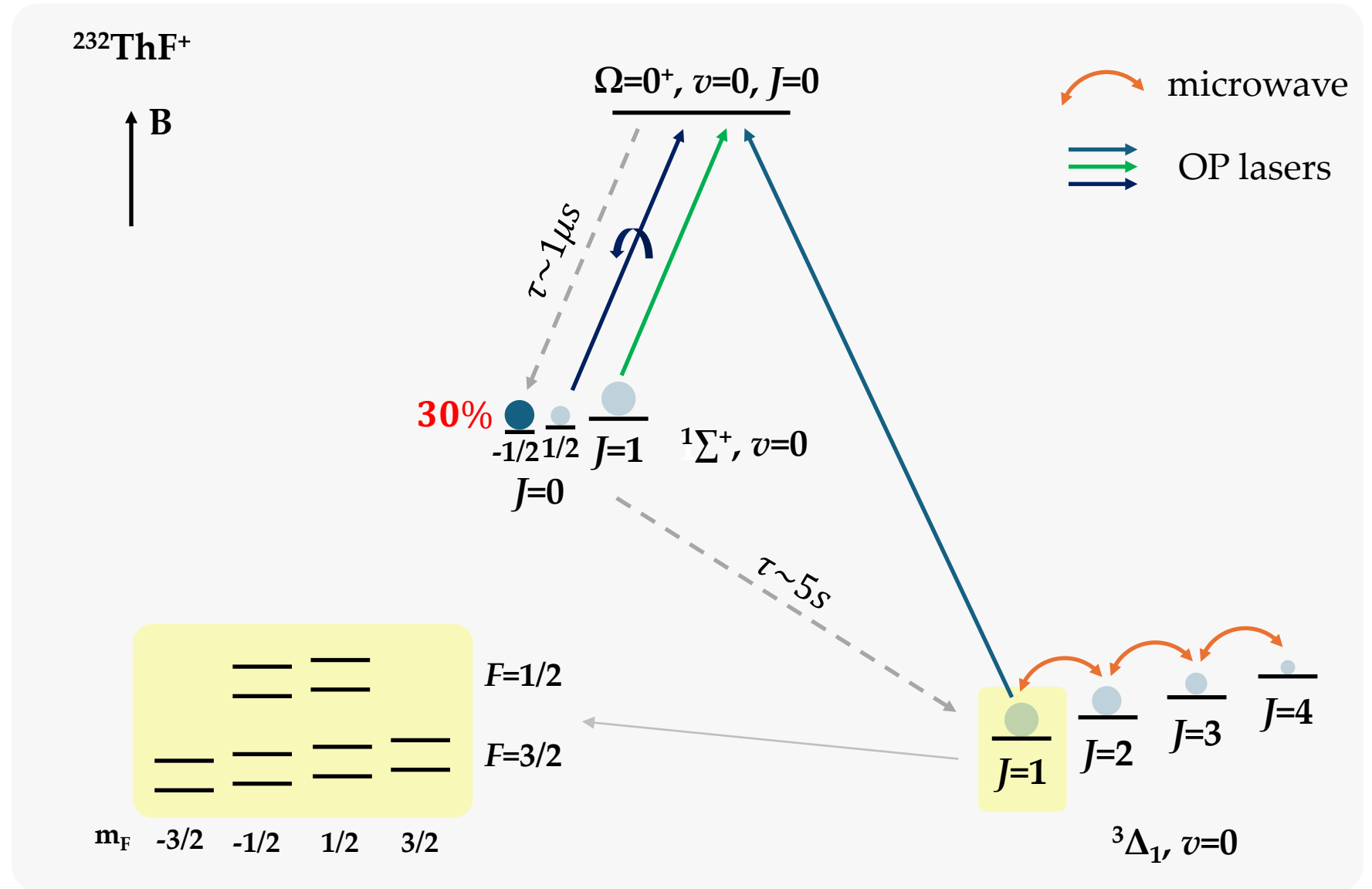
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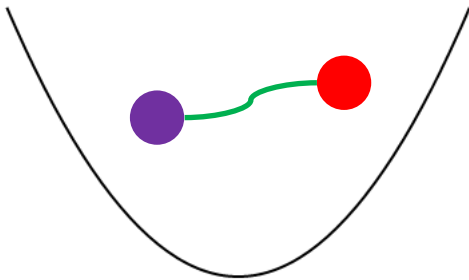


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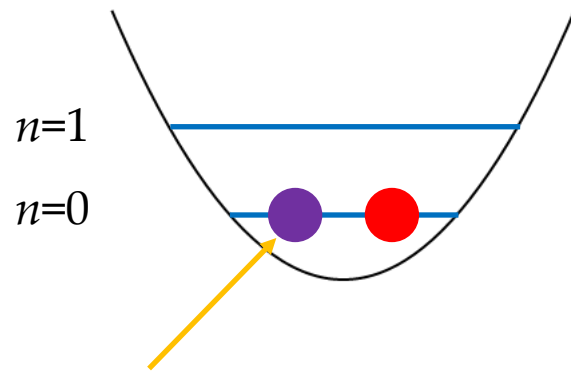
State preparation and detection

- We have a harmonic ion trap
- Load one Yb^+ and one ThF^+ in the trap
- Two ion species are linked by the Coulomb force



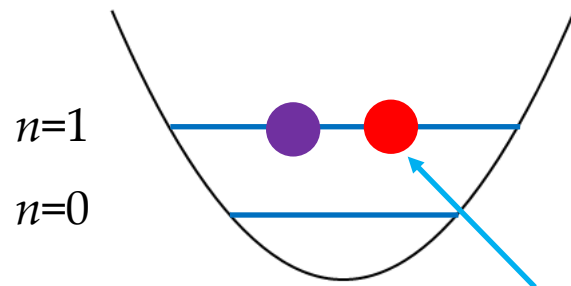
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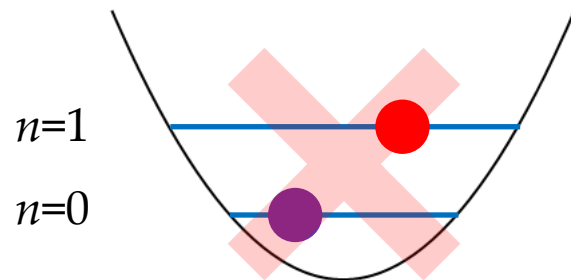
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- If ThF^+ is excited to the motional excited state, Yb^+ is in the motional excited state as well



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- ThF^+ is sympathetically cooled to the ground motional state
- If ThF^+ is excited to the motional excited state, Yb^+ is in the motional excited state as well
- This state does not exist



State preparation and detection

$^{171}\text{Yb}^+$

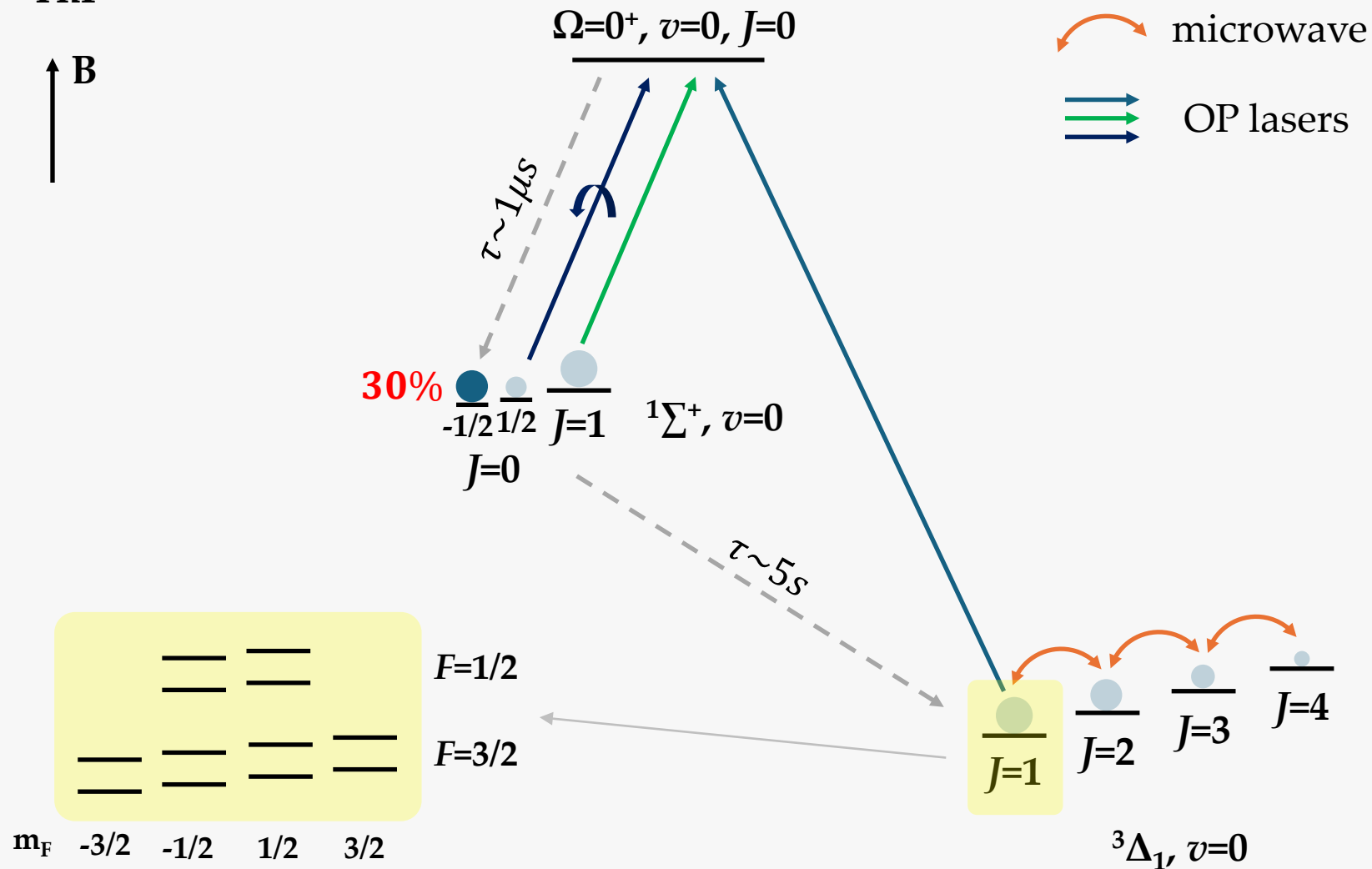
— $^2\text{P}_{1/2}, F=0$

$^2\text{D}_{5/2}, F=2$
—

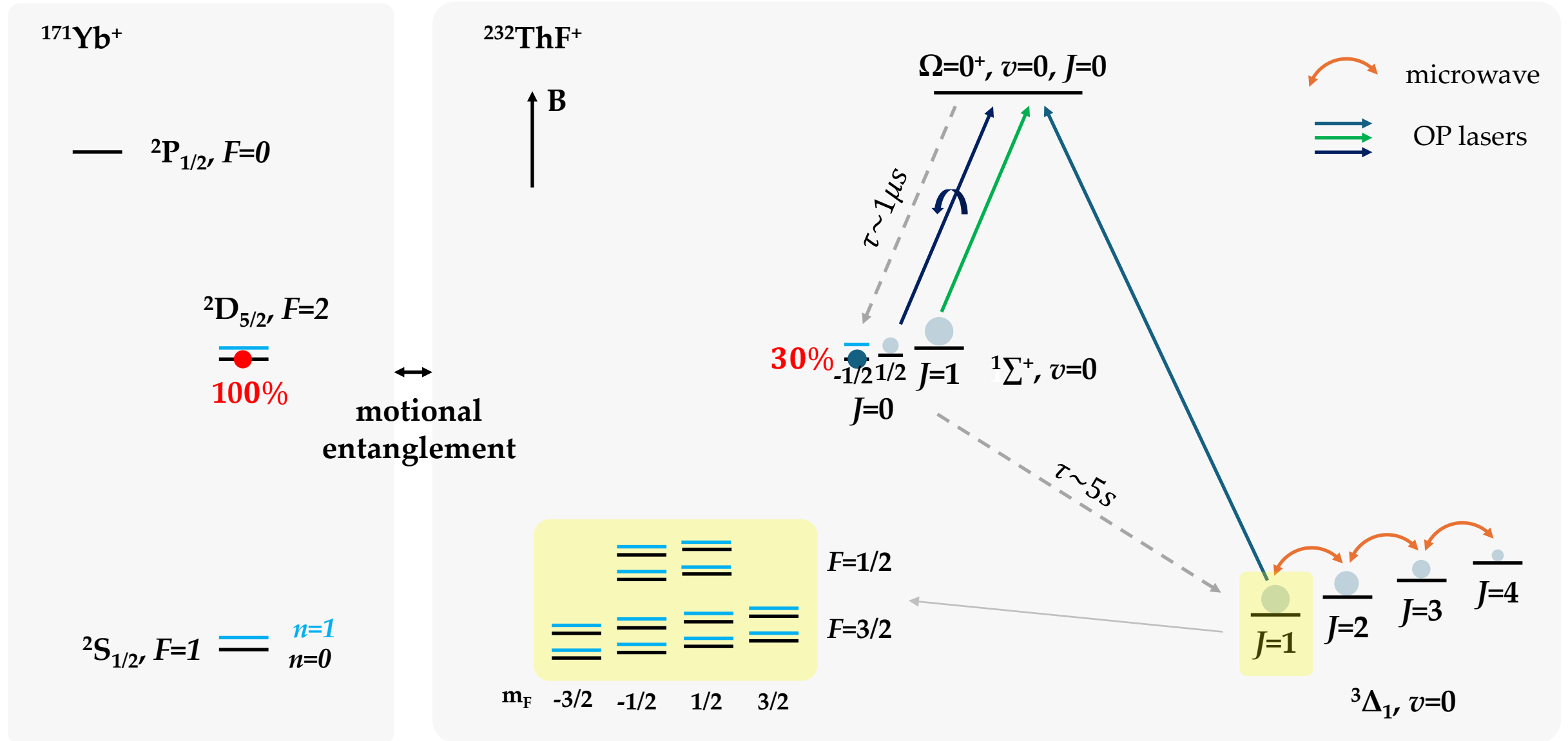
$^2\text{S}_{1/2}, F=1$ —

$^{232}\text{ThF}^+$

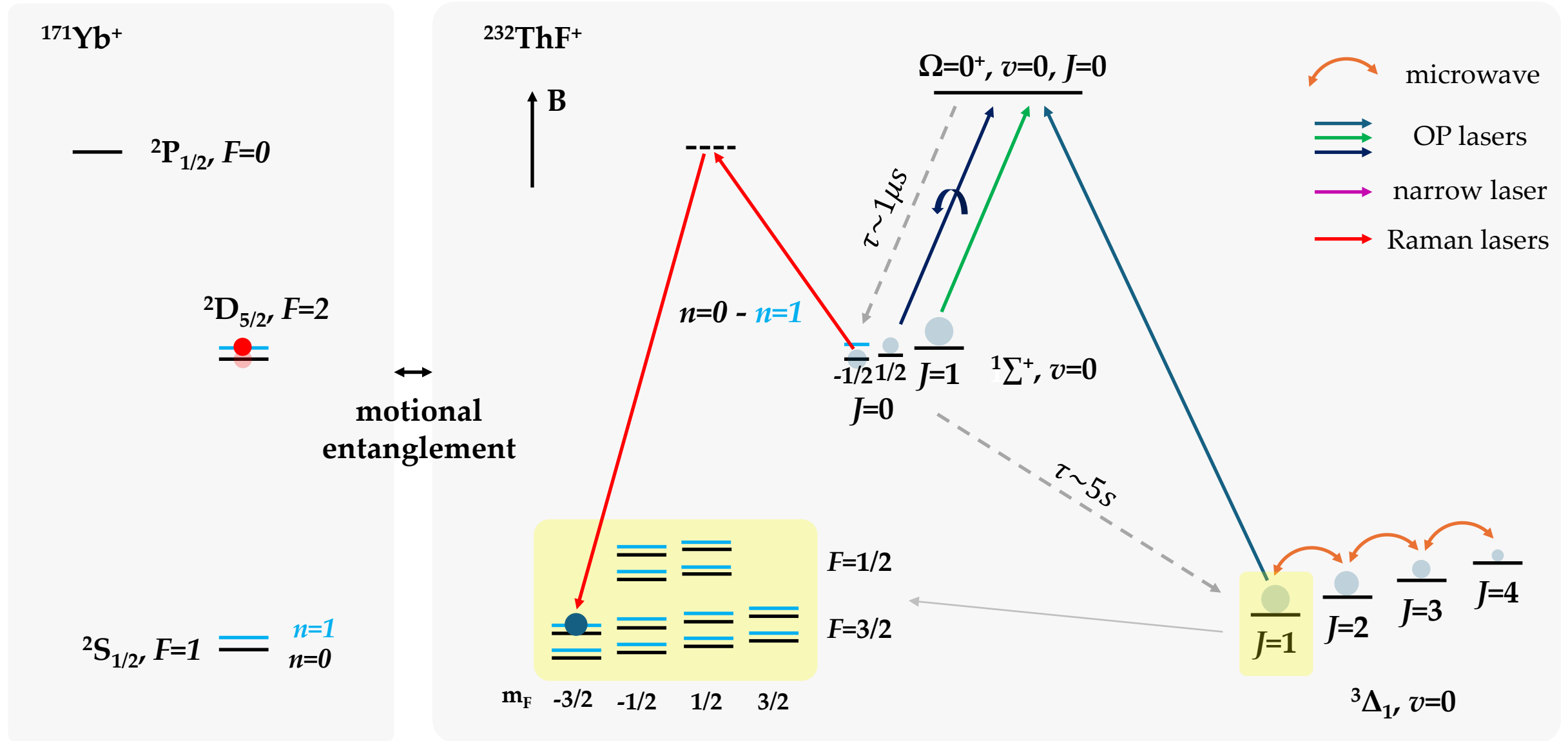
↑ B

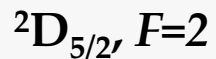
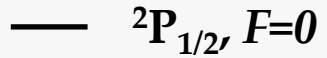
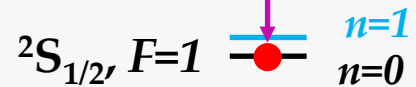


State preparation and detection



State preparation and detection



$^{171}\text{Yb}^+$ 
$$n=1 - n=0$$


$n=1$
 $n=0$

motional entanglement



B

$$\Omega=0^+, v=0, J=0$$

$\tau \sim 1 \mu s$

$$J=0 \quad J=1 \quad {}^1\Sigma^+, v=0$$
 $\tau \sim 5s$
$$F=1/2$$
 $F=3/2$
$$m_F \quad -3/2 \quad -1/2 \quad 1/2 \quad 3/2$$

 microwave

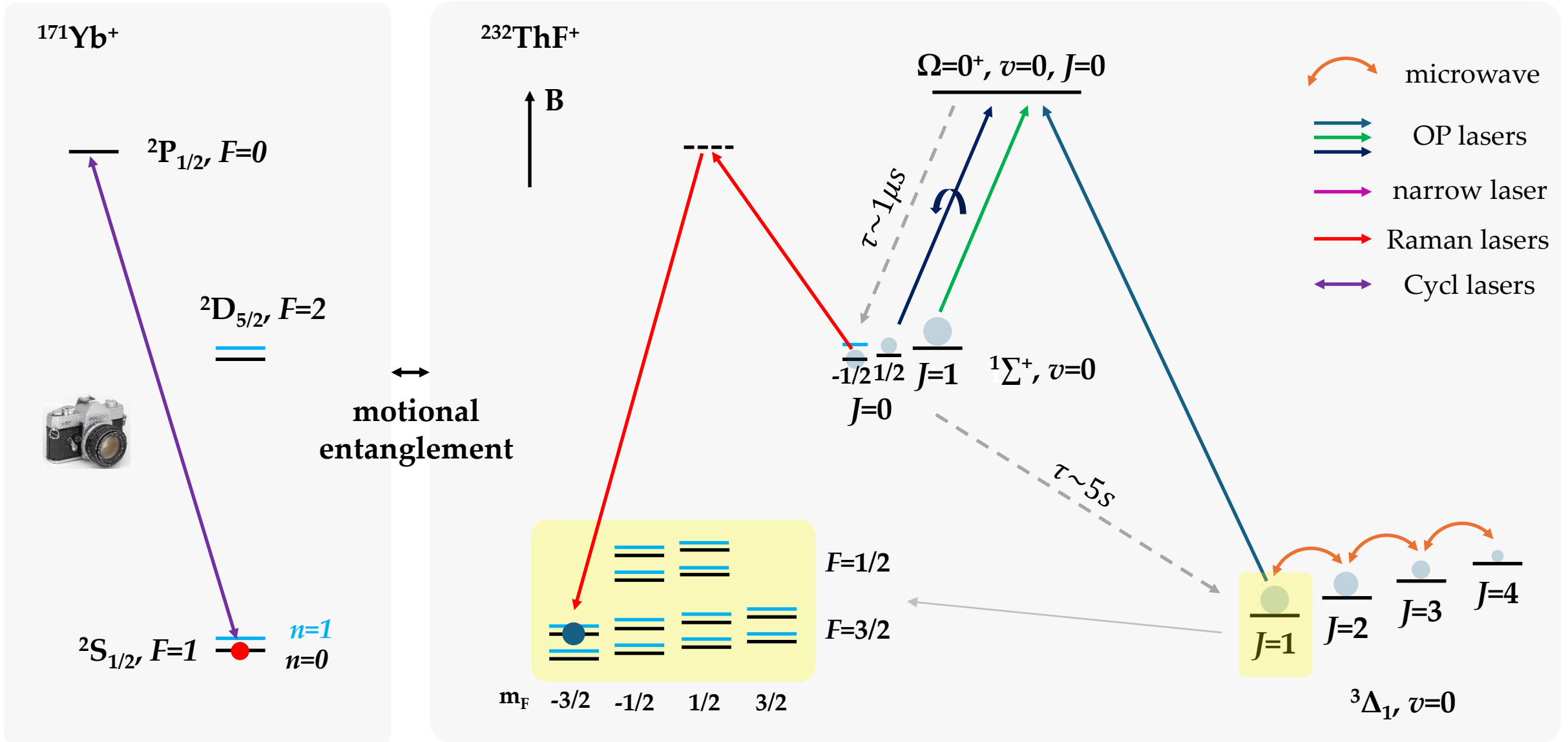
OP lasers

→ narrow laser

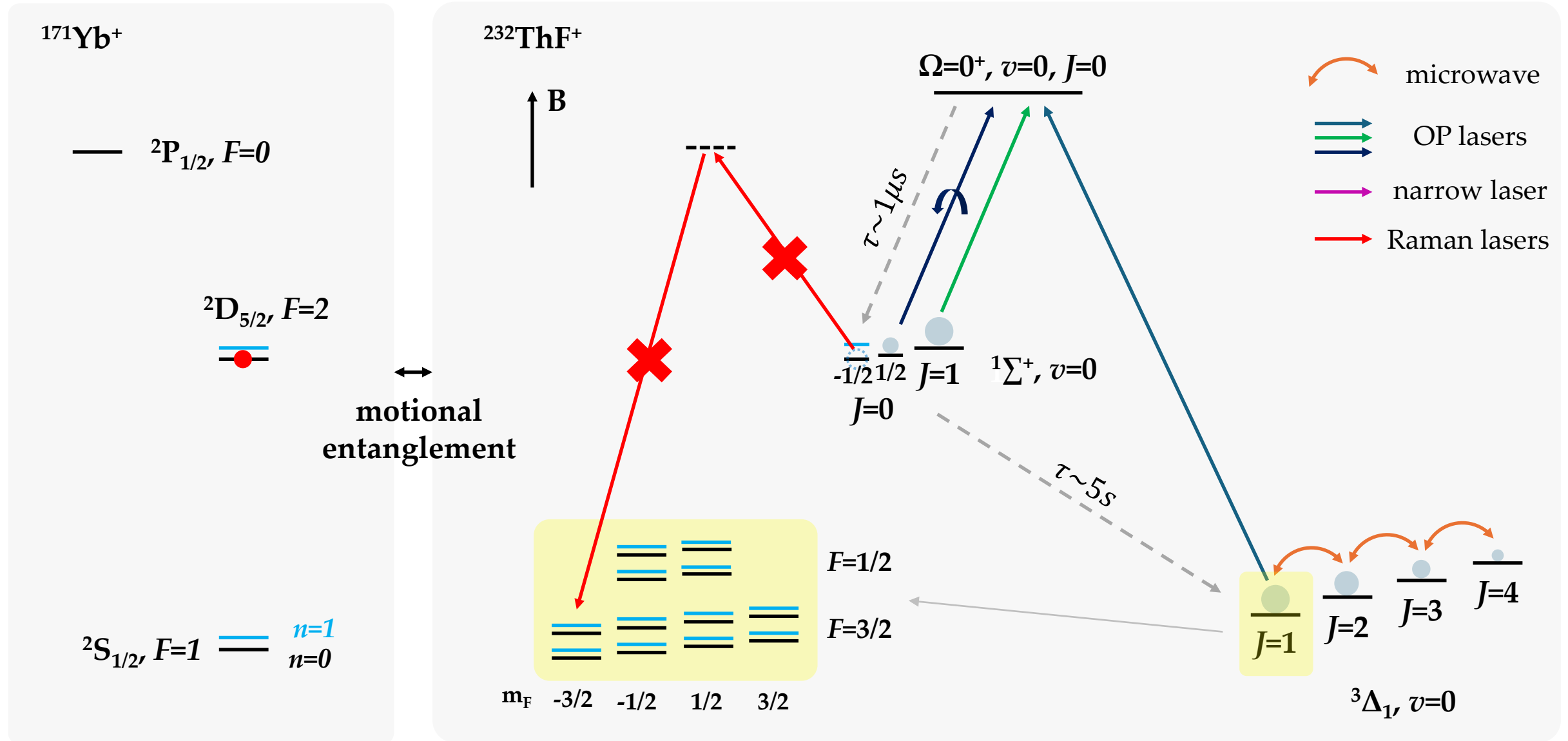
→ Raman lasers

 ${}^3\Delta_1, v=0$

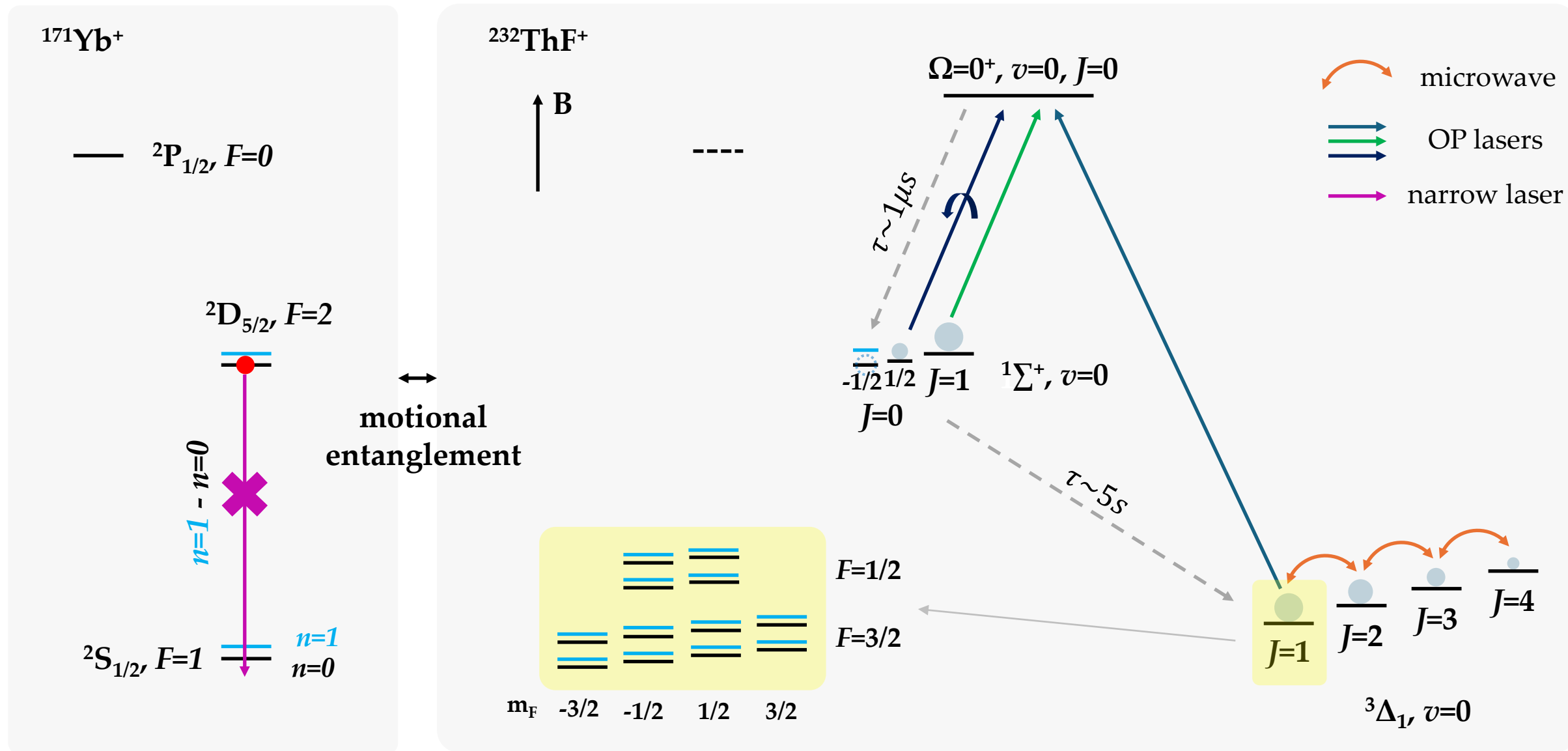
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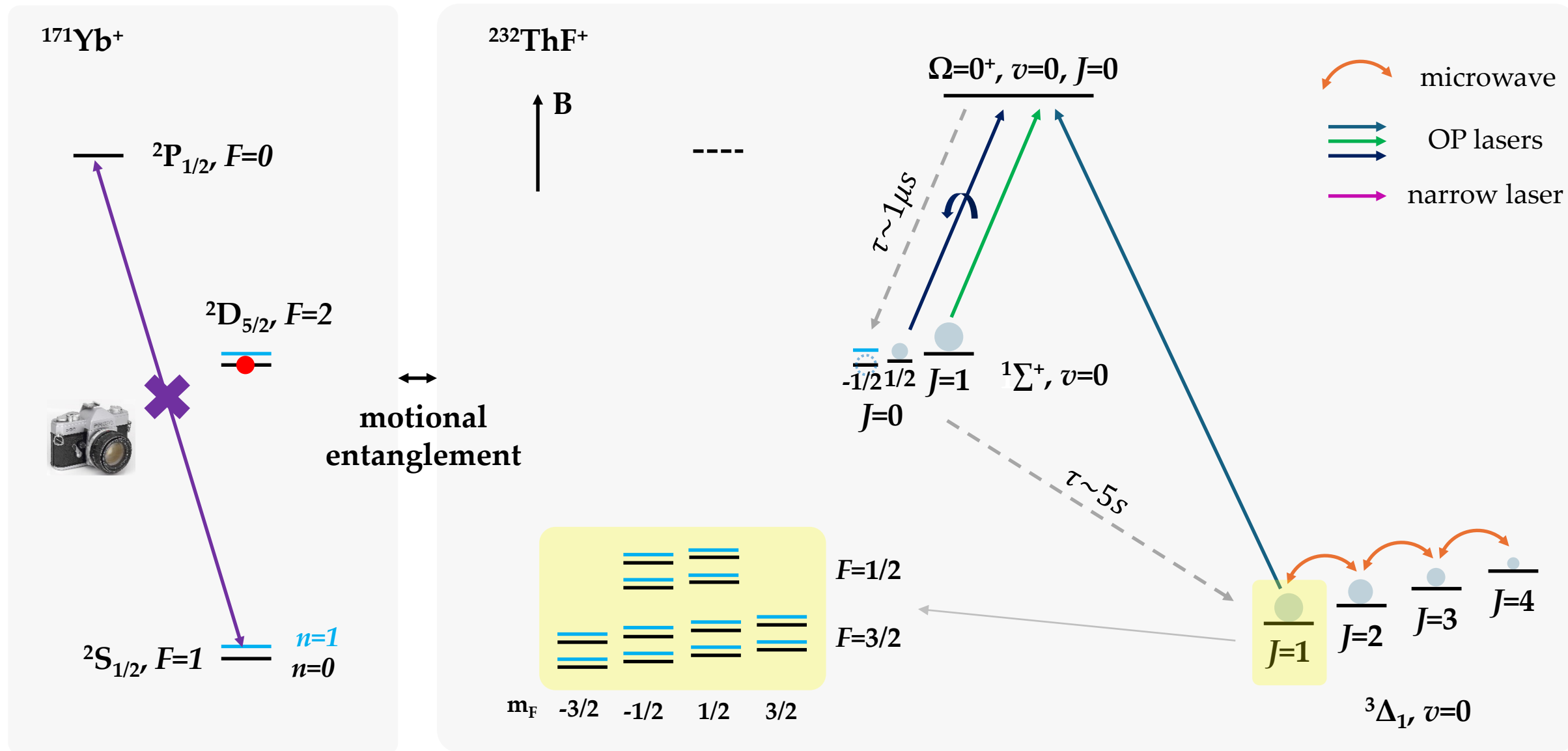
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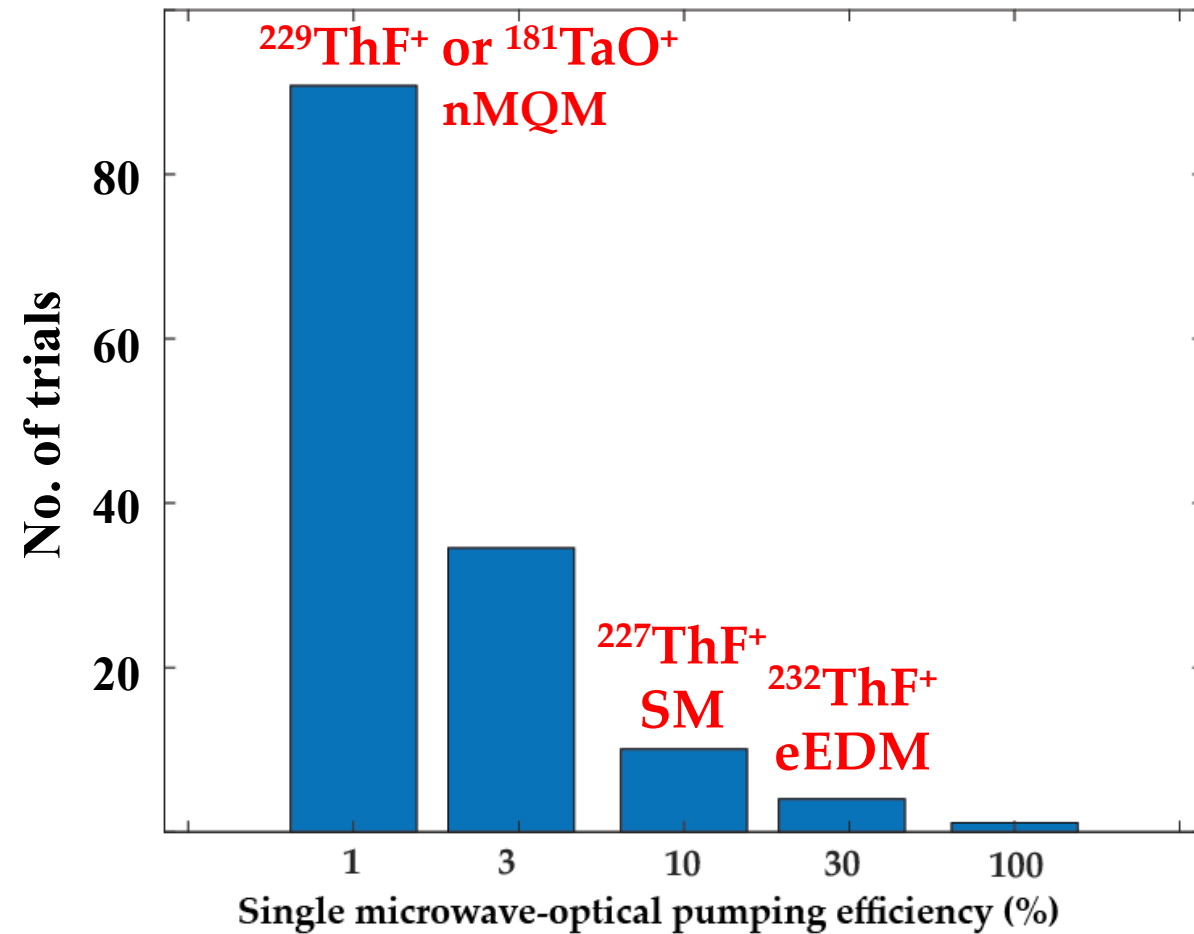
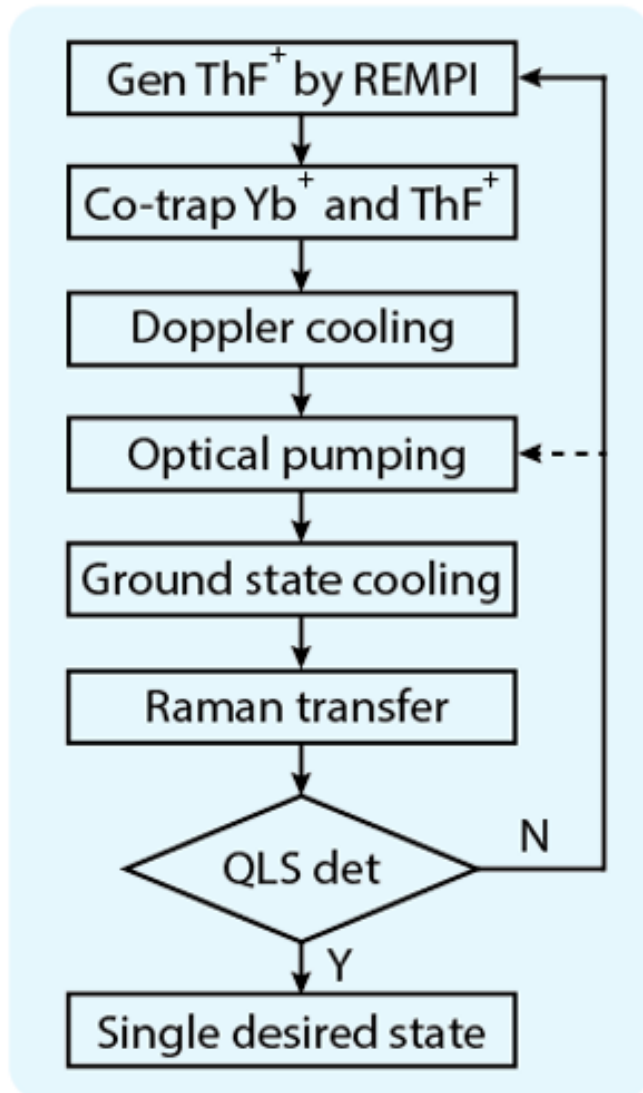
State preparation and detection



State preparation and detection

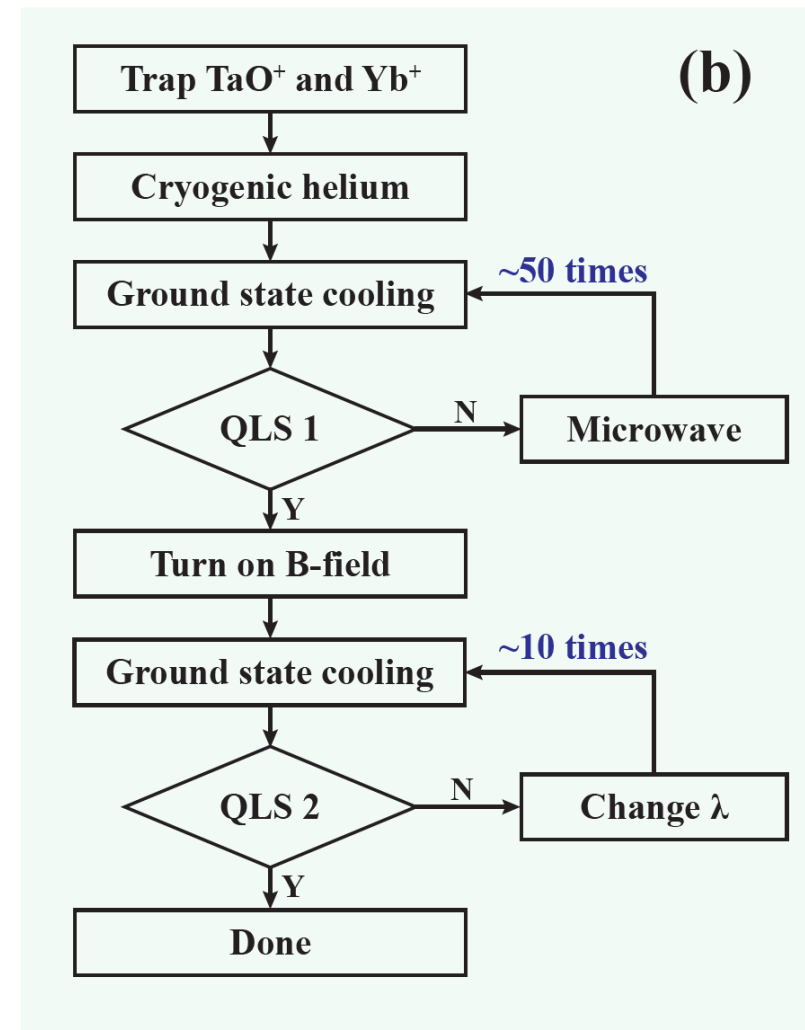
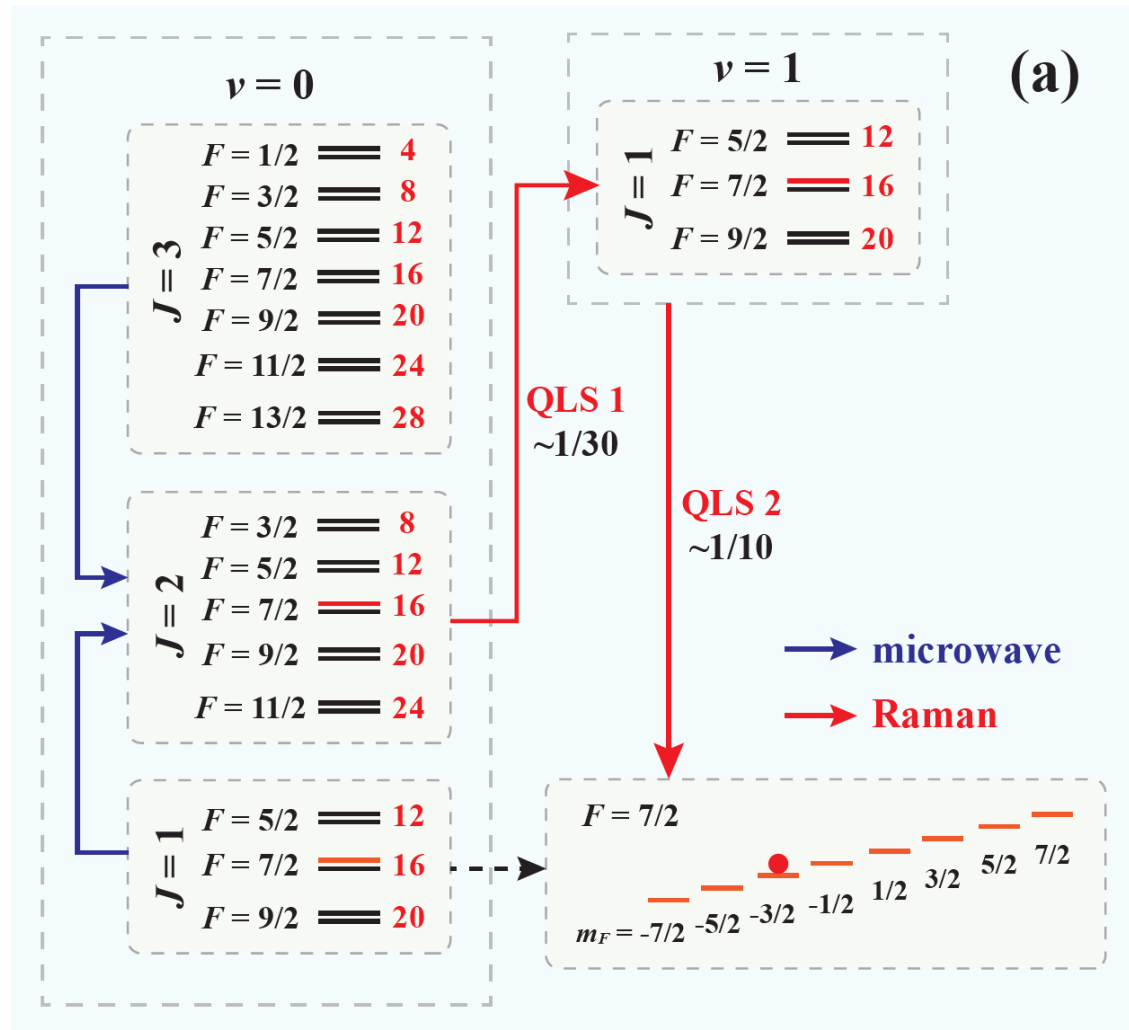


Repetitive trials



- QLS Fidelity ~95%

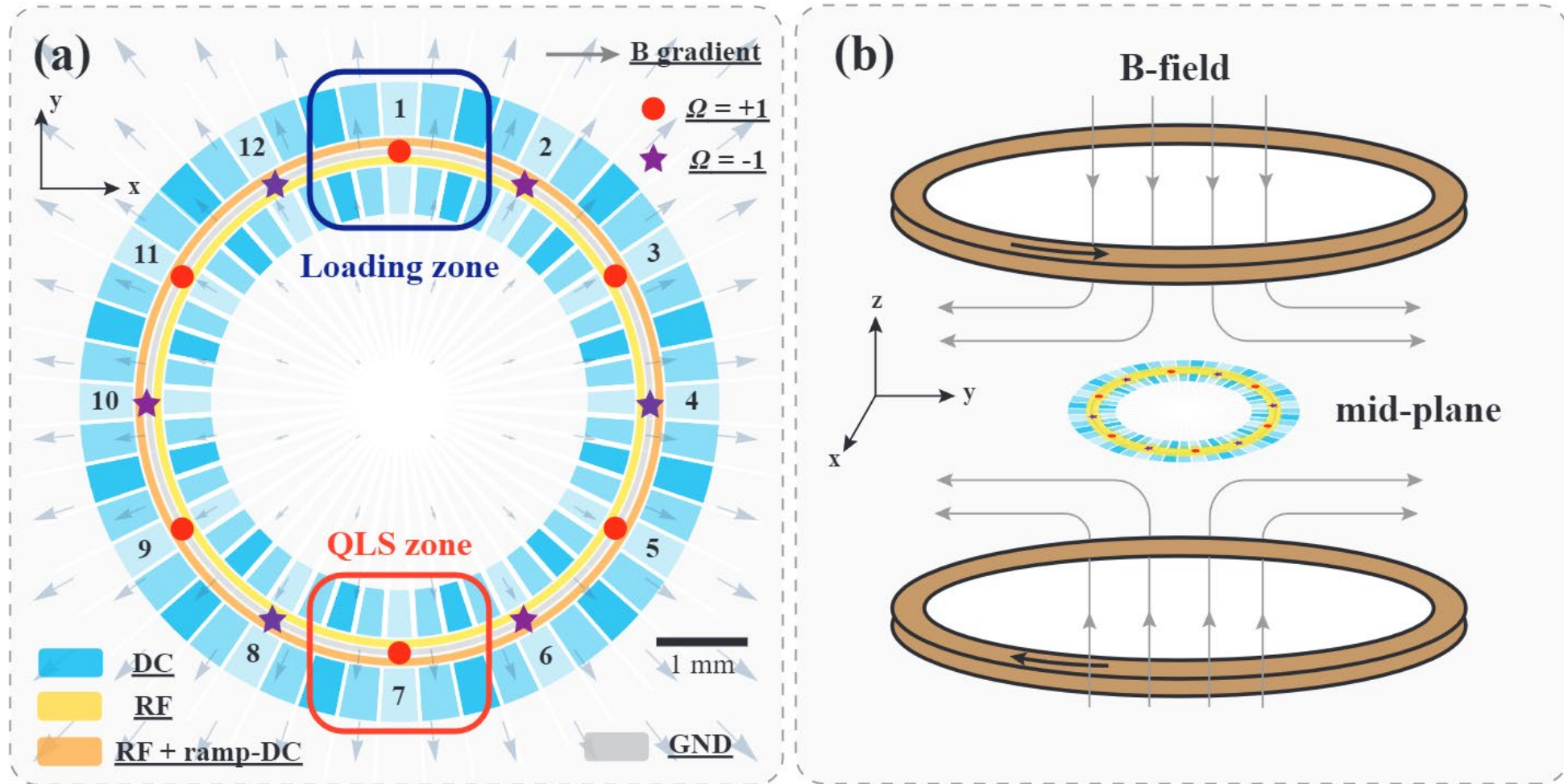
Degenerate QLS



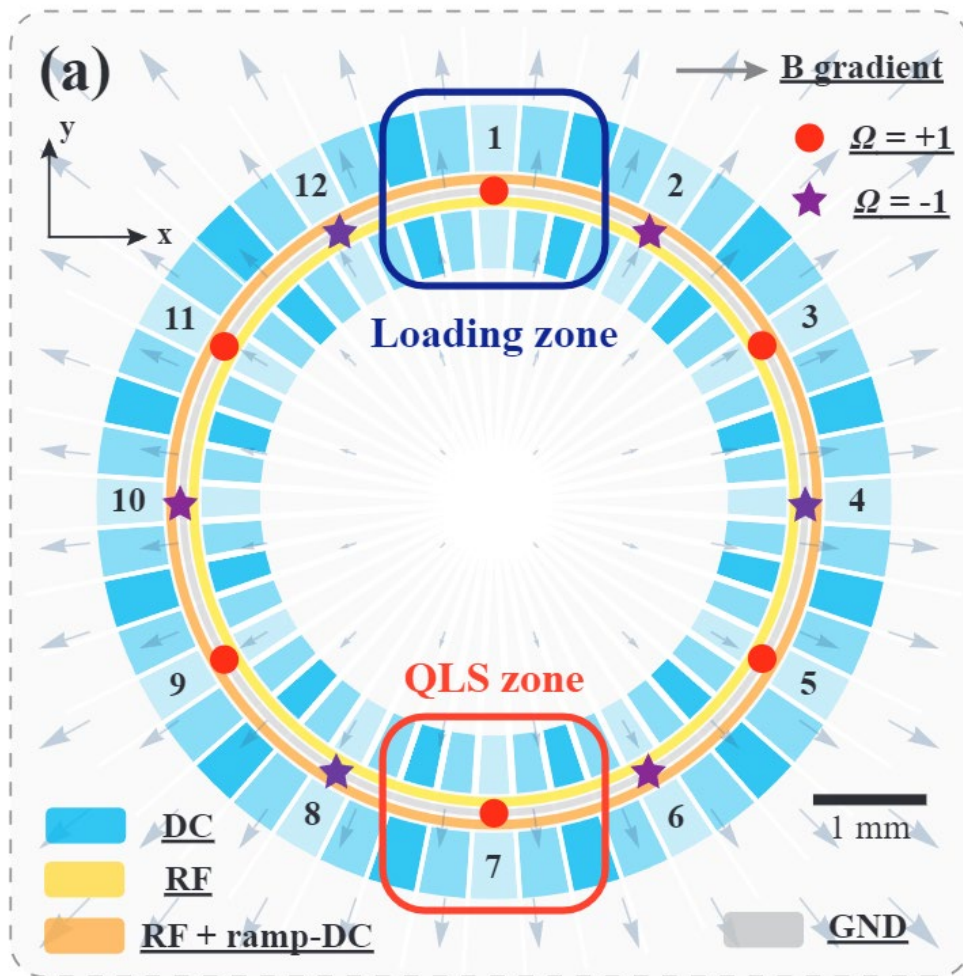
Electric and magnetic fields

- We can apply DC magnetic fields for spin-polarization
- We cannot apply DC electric fields for molecular ions
- No biased electric field, no eEDM sensitivity
- JILA eEDM group applies ~ 10 V/cm rotating external field to polarize molecules
- Rotating E-field is incompatible with QLS
 - Send laser beams into a rotating frame
 - **Excess heating**
- Solution
 - **Separate QLS (static frame) and spin-precession (rotating frame)**
 - A smooth transition between these frames

Ring ion trap



Ring ion trap

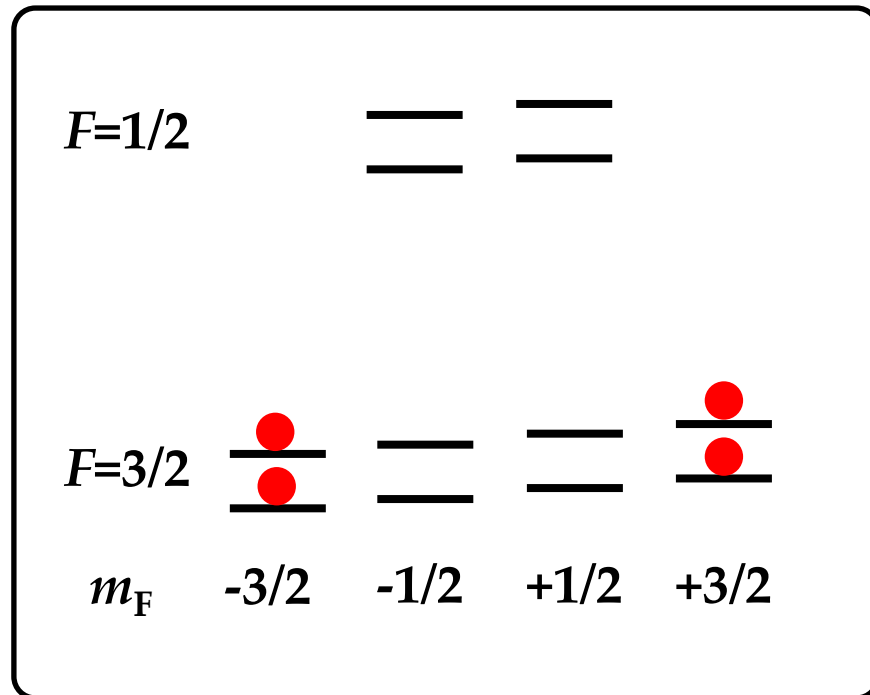


Property	Value
Radial Freq	2.5 MHz
Axial Freq	1 MHz
Rotation Freq	0-100 kHz
Rotation radius	3 mm
E-field	0-32 V/cm

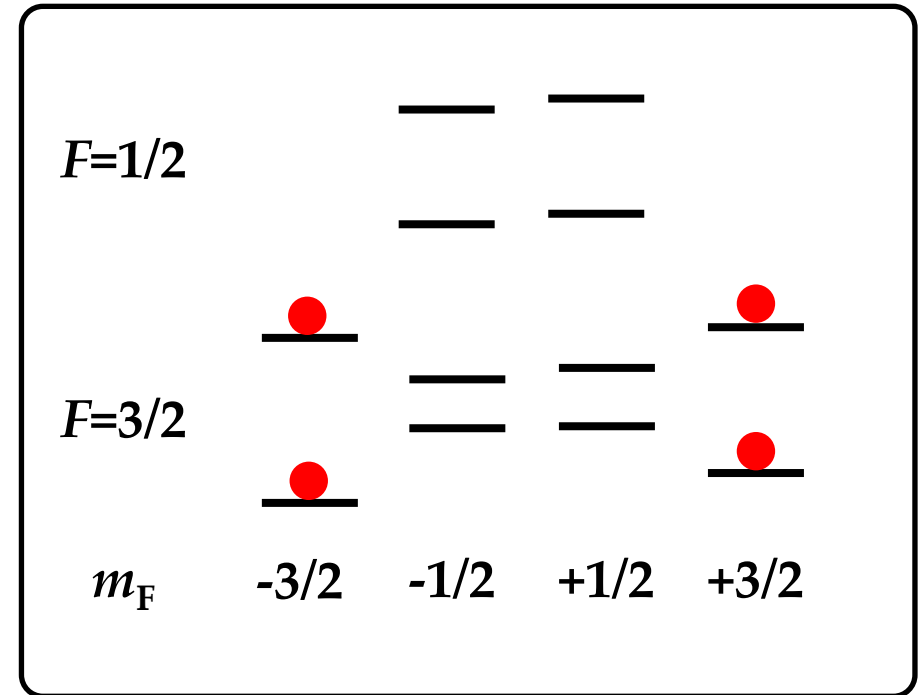
- ☐ QLS in the static frame
- ☐ Precision measurement in the rotating frame

Transition from static to rotating frames

Static frame

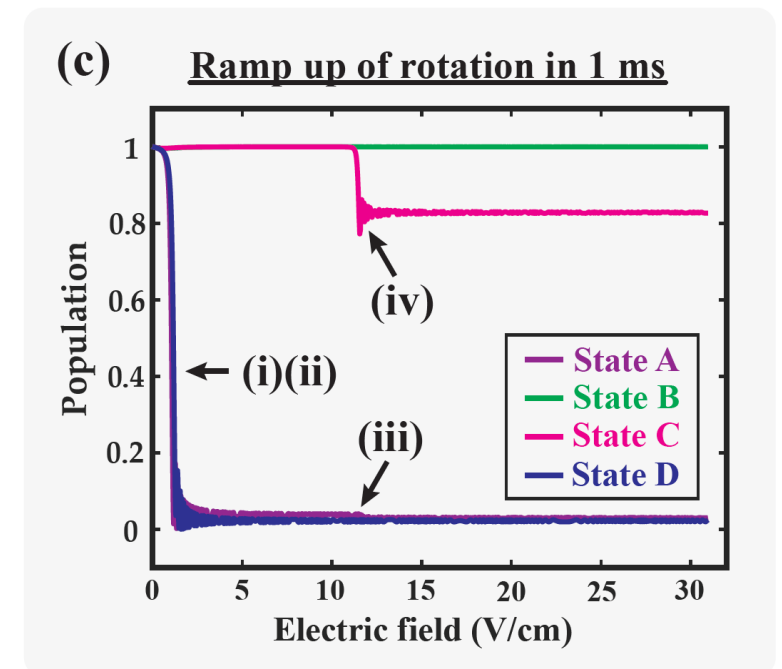
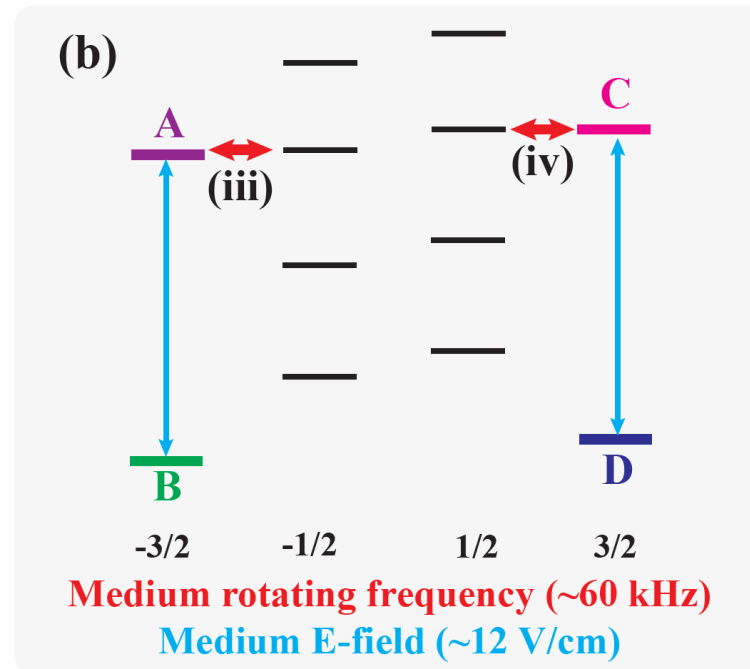
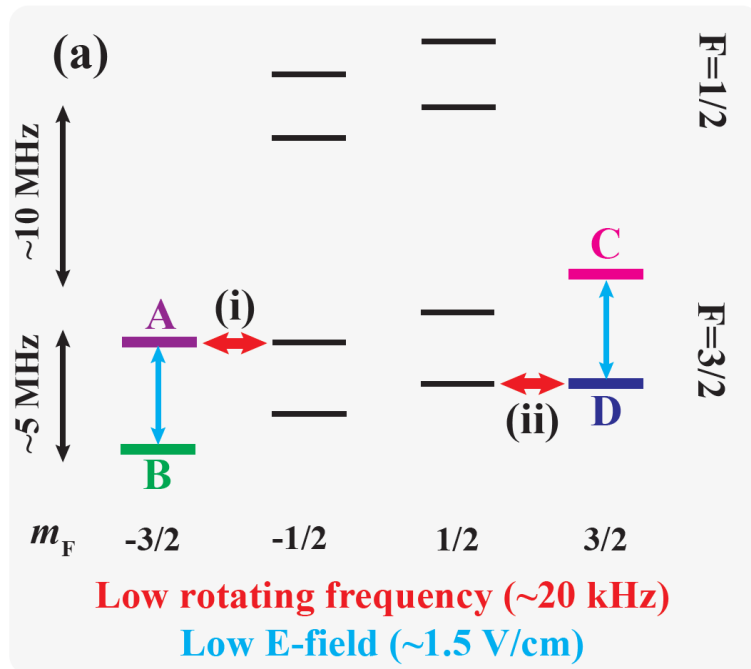


Rotating frame



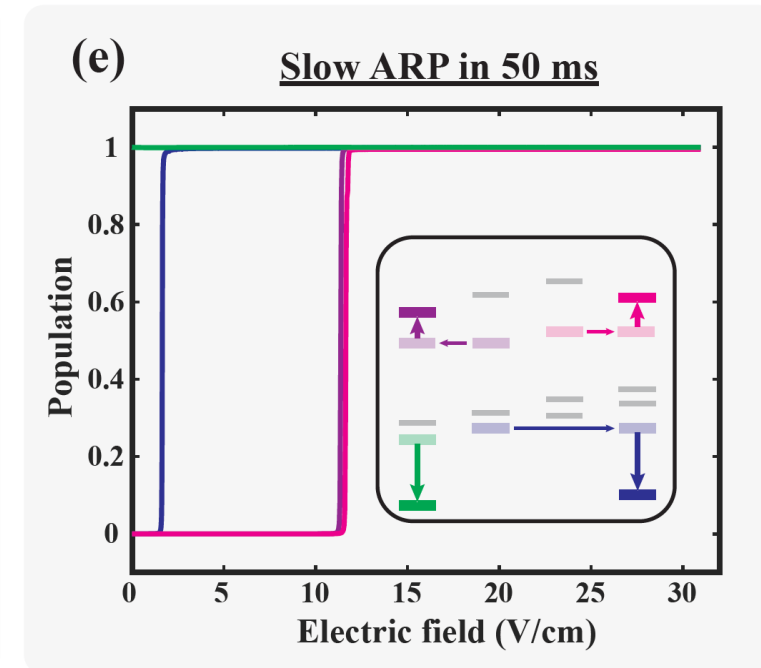
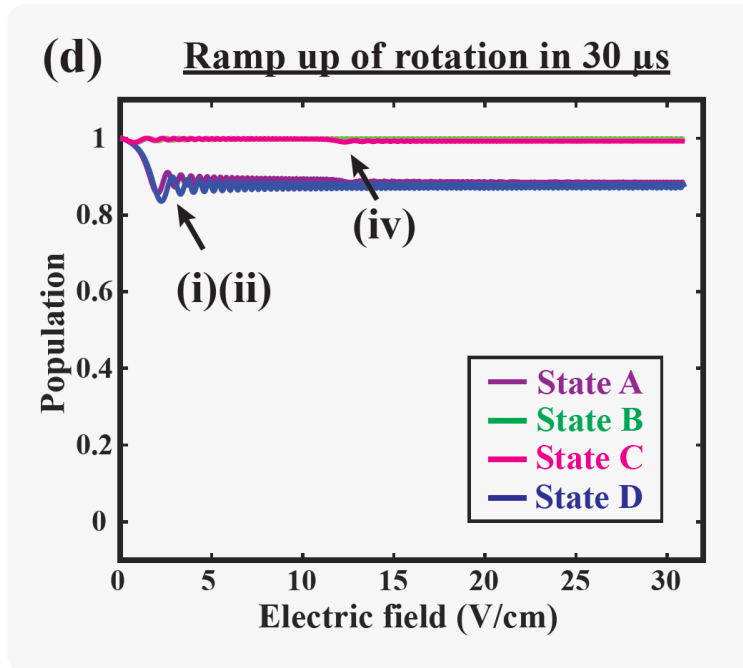
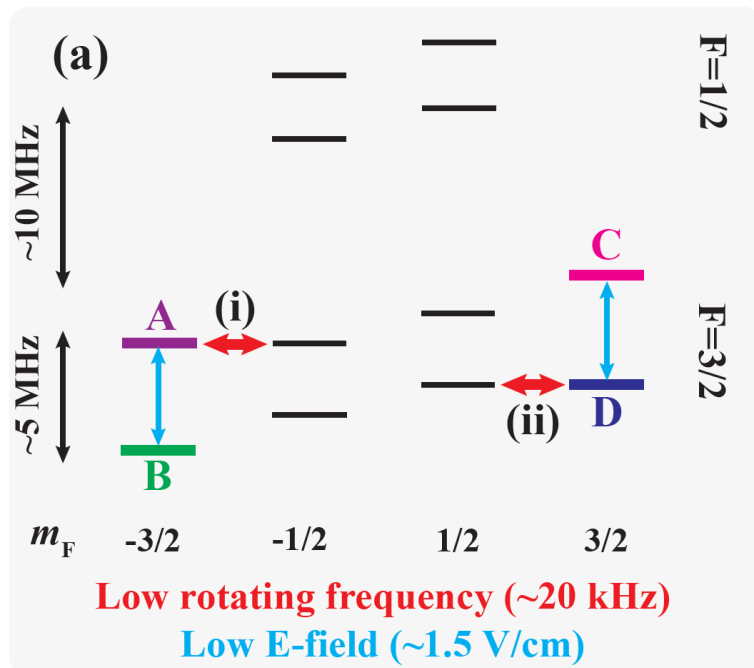
Transition from static to rotating frames

- What happens when the ions are rotating faster and faster?
 - Rotating E-field repel A and B, C and D
 - Rotating coupling interacts $\Delta m_F = \pm 1$
 - Rotating B-field is constant – Zeeman shifts do not change

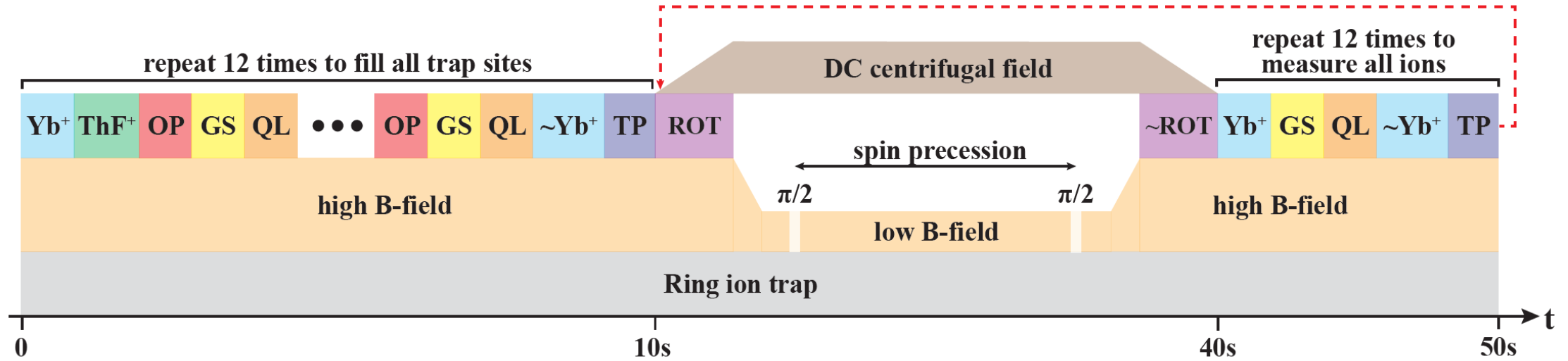


Transition from static to rotating frames

- ❑ Solution 1: increase the ramp rate
- ❑ **Solution 2: adiabatic population transfer**



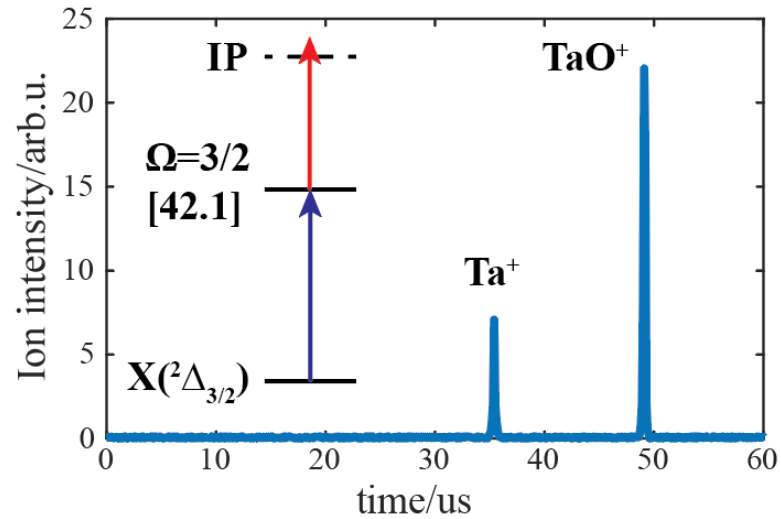
Experimental sequence



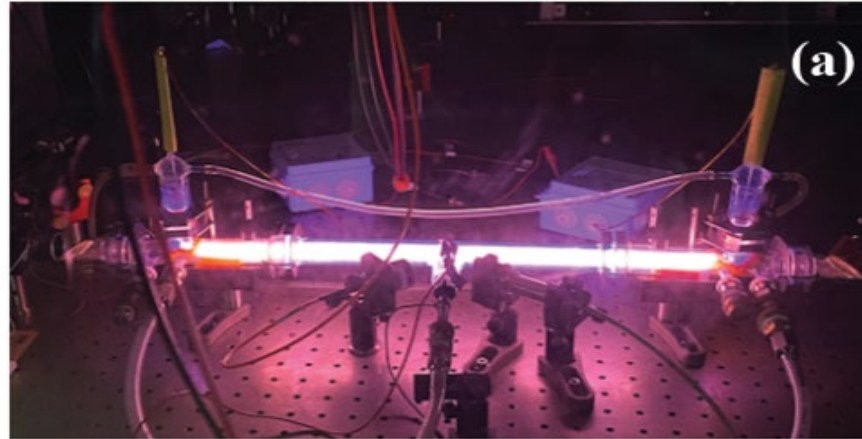
- ❑ State preparation and detection is performed for each ions one-by-one
- ❑ Spin-precession is for all ions
- ❑ State readout of one measurement is the state preparation of the next measurement

Current progress

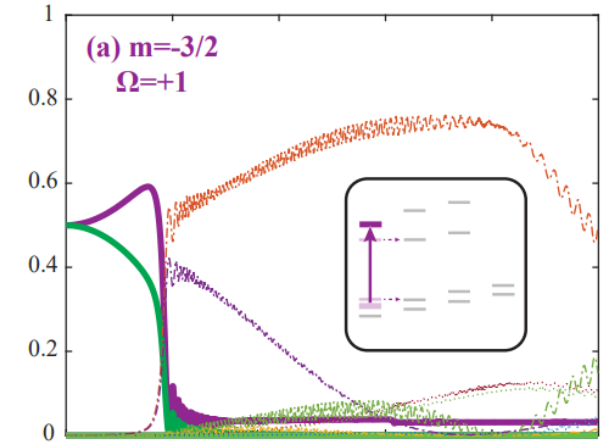
TaO spectroscopy



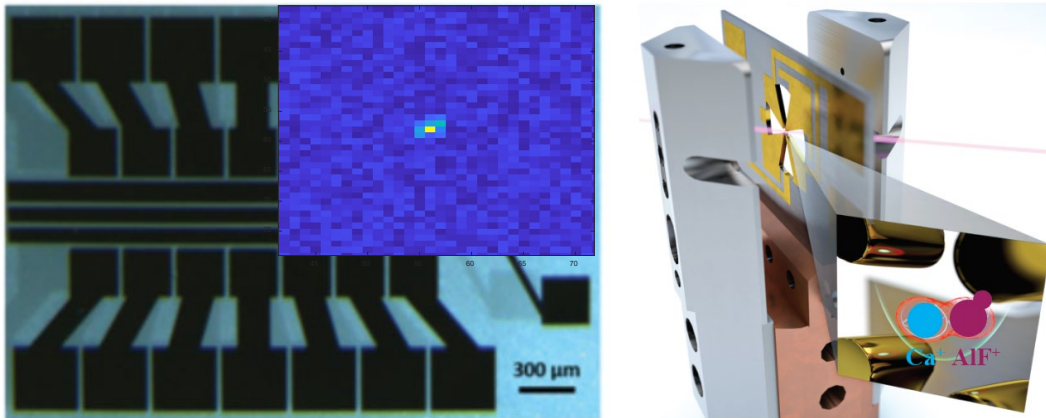
VMS spectroscopy of TaO⁺



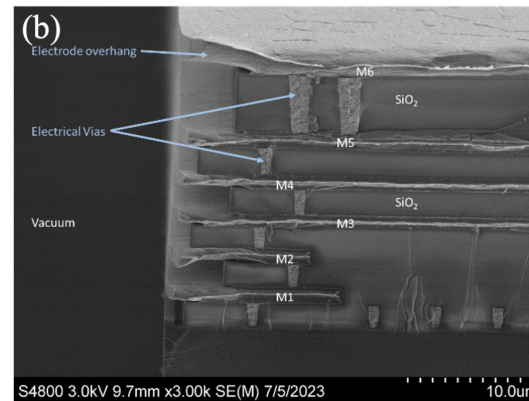
Numerical model



Quantum logic spectroscopy



Trap fabrication



Sandia National Laboratories

Collaborations

