Spectroscopy of RaF Molecules for Fundamental Physics

Silviu-Marian Udrescu

PhD. Student, MIT

June 30 2021

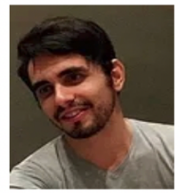
Thanks to…

Quantum chemistry:
R. Berger (U. Marburg, Germany),
T. Isaev (PNPI NRCKI, St. Petersburg)

***ISOLTRAP** (F. Wienholtz) , **RILIS** (S. Wilkins, K. Chrysalidis)*
Target group** (S. Rothe), **ISOLDE Technical group

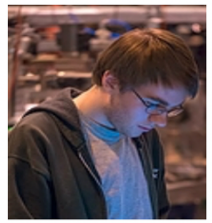
Graduate students

A. Brinson

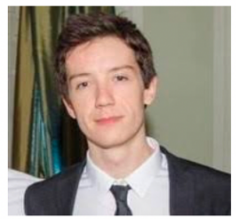


S. Udrescu

Postdocs



A. Vernon



S. Wilkins

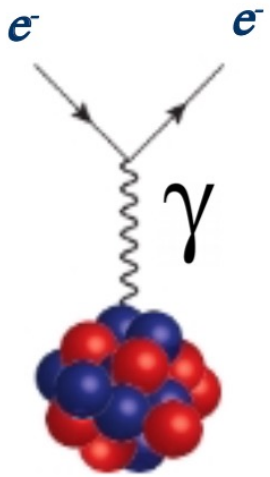
I. Belosevic

J. Karthein

Content

- Motivation
- Isotope shift in Radium Monofluoride
  - Results
  - Implications for nuclear physics and searches for new forces
  - What can we gain more compared to atoms?
- High resolution spectroscopy of $^{226}\mathrm{RaF}$
  - Preliminary results
  - How can we achieve a higher ground state population and reduced background?
  - How can we decelerate and trap RaF molecules?
- Conclusions and Outlook

Why short-lived radioactive molecules?



Low-energy SM tests

- **Nuclear matter**
- **Nuclear structure**
- **BSM searches**

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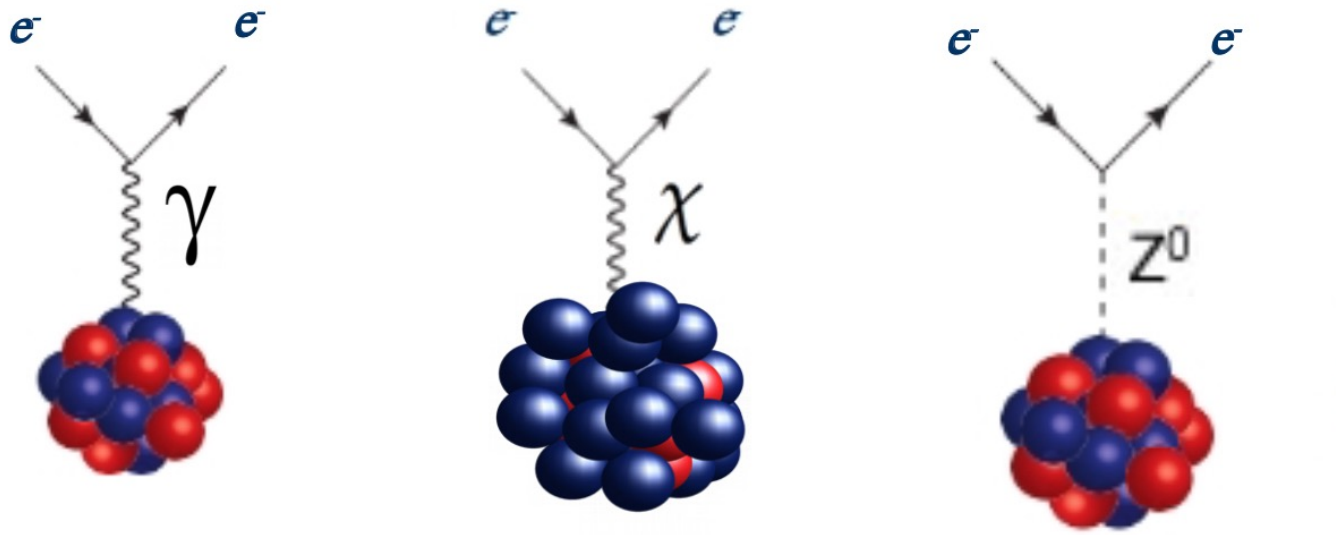

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- Dark Matter properties?
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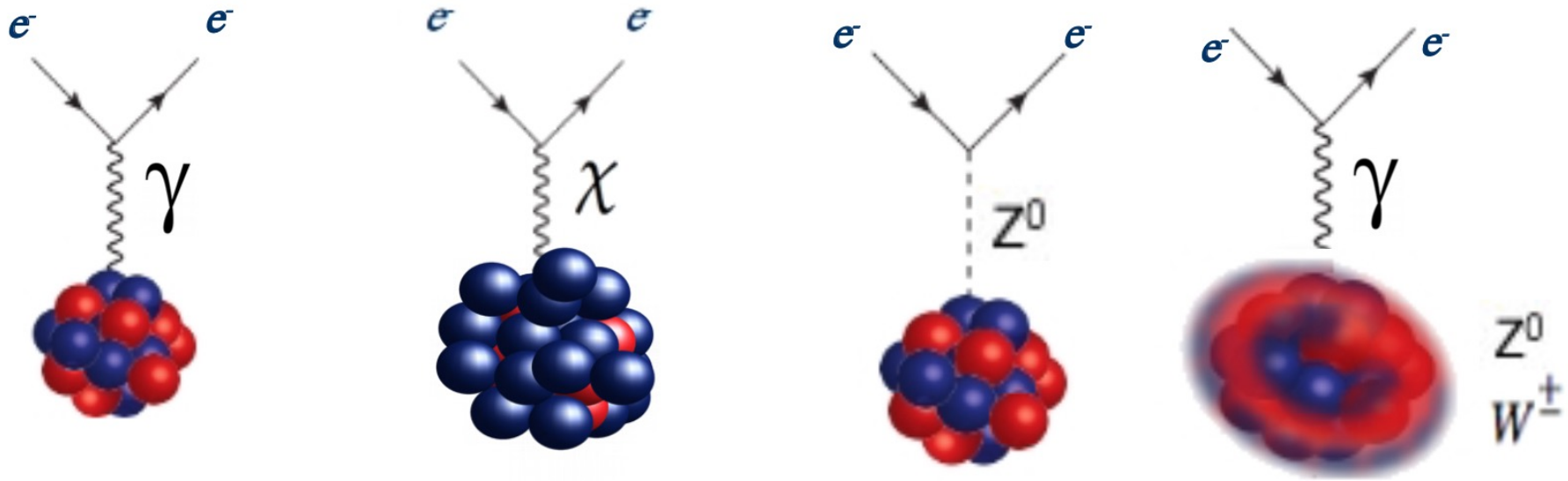

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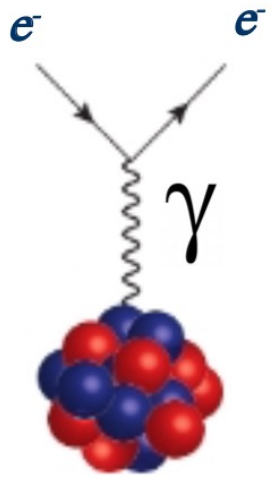

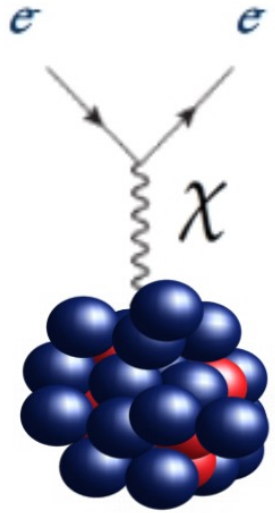

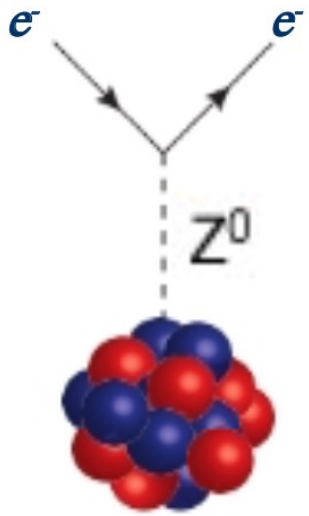

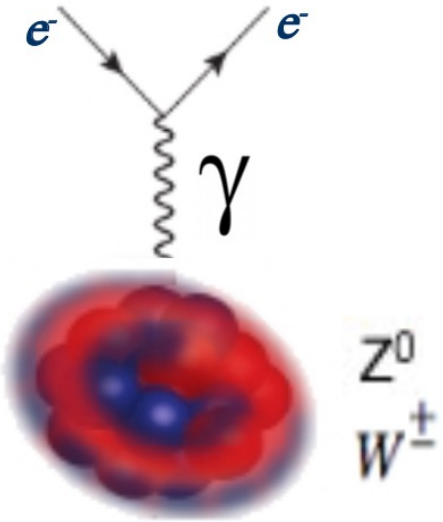

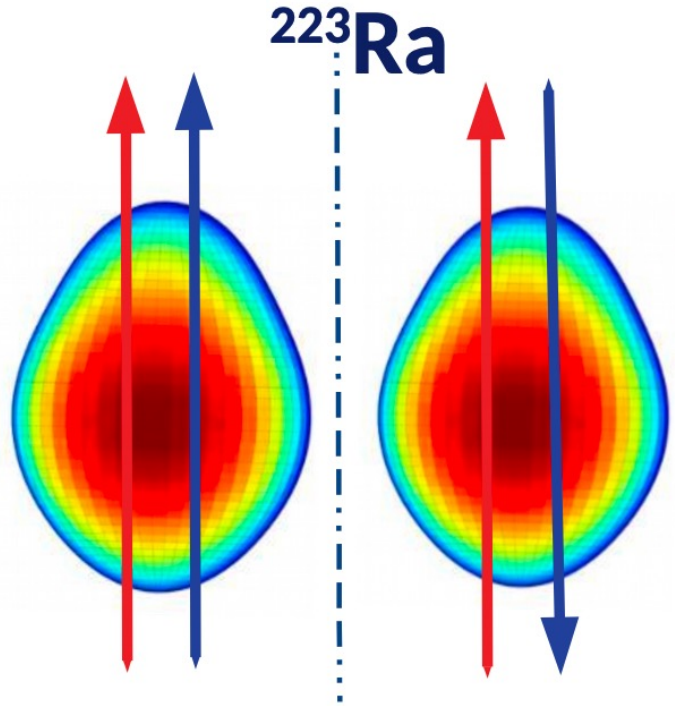

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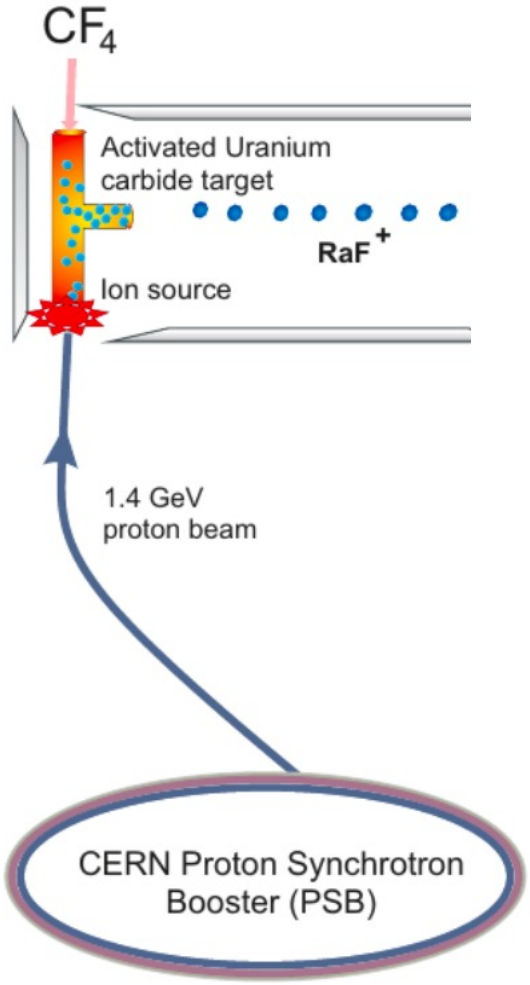
T-violation

- Baryogenesis

Experimental Setup



Garcia Ruiz, Berger et al. CERN-INTC-2018-017 (2018)

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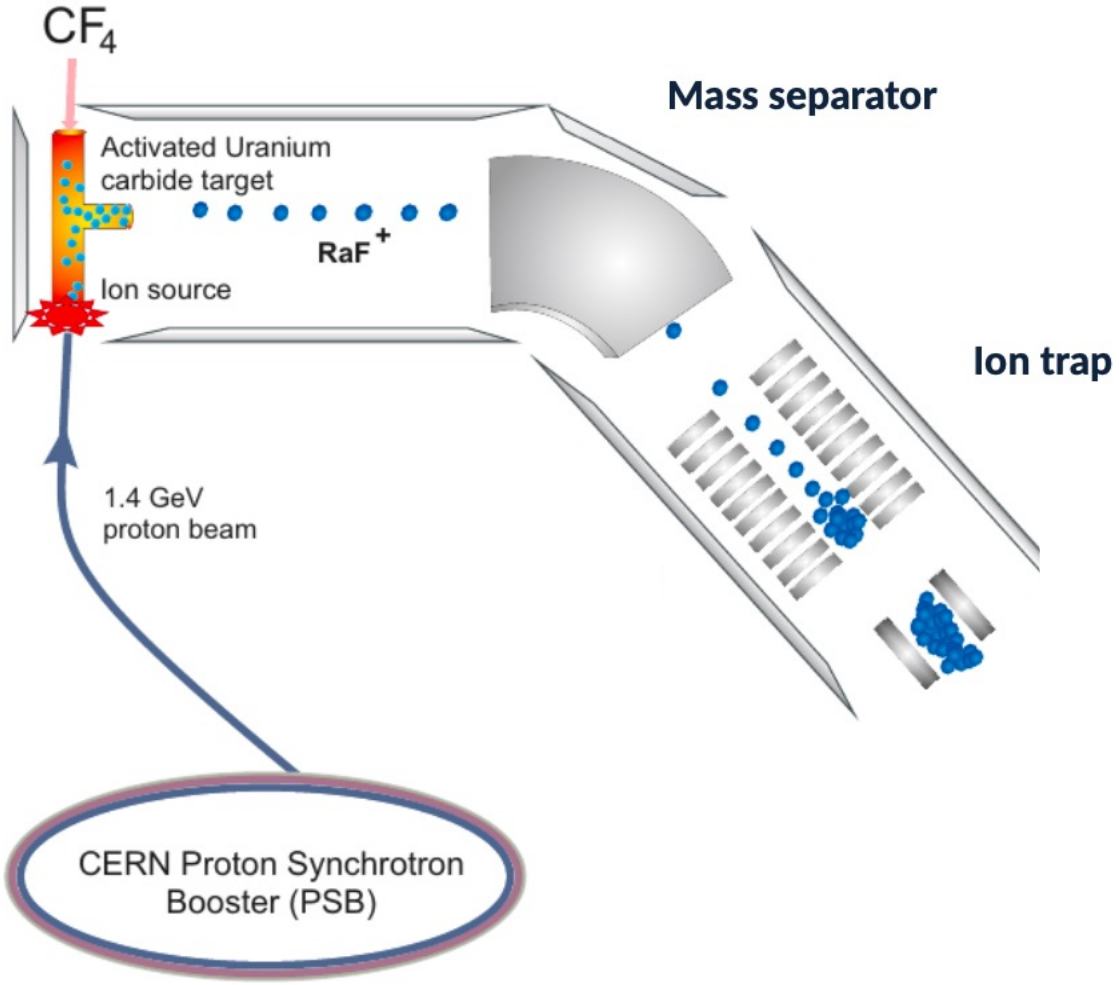

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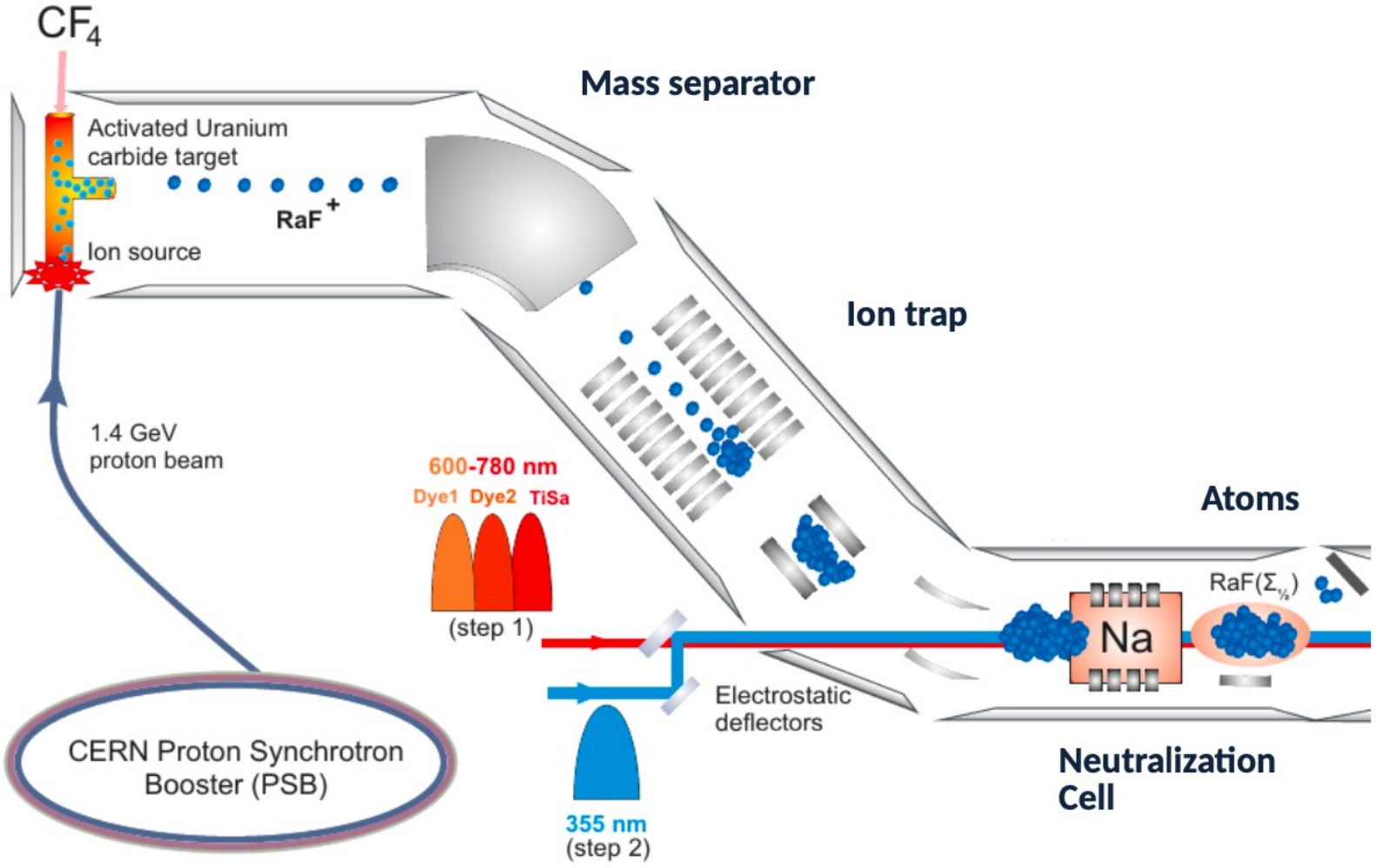

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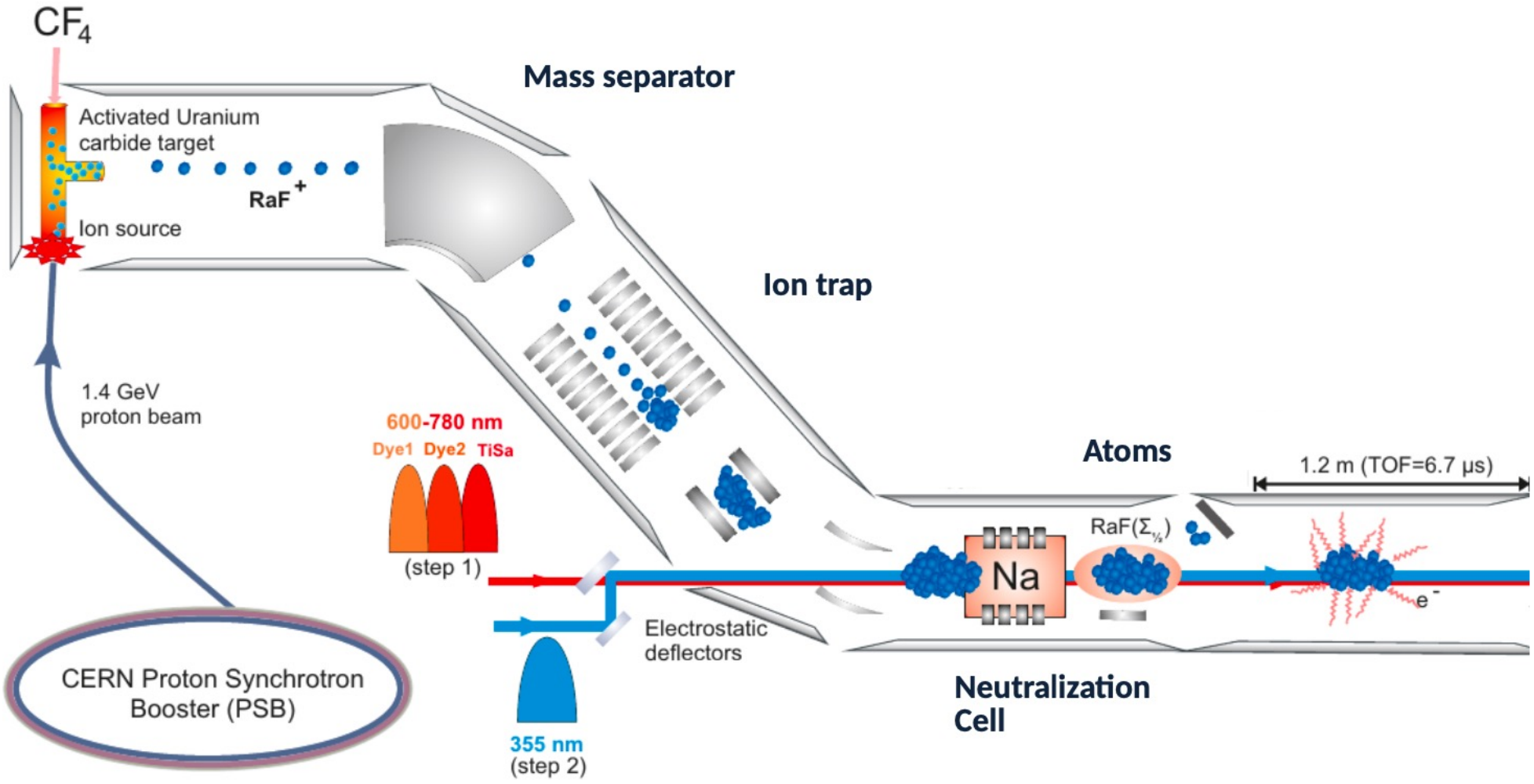

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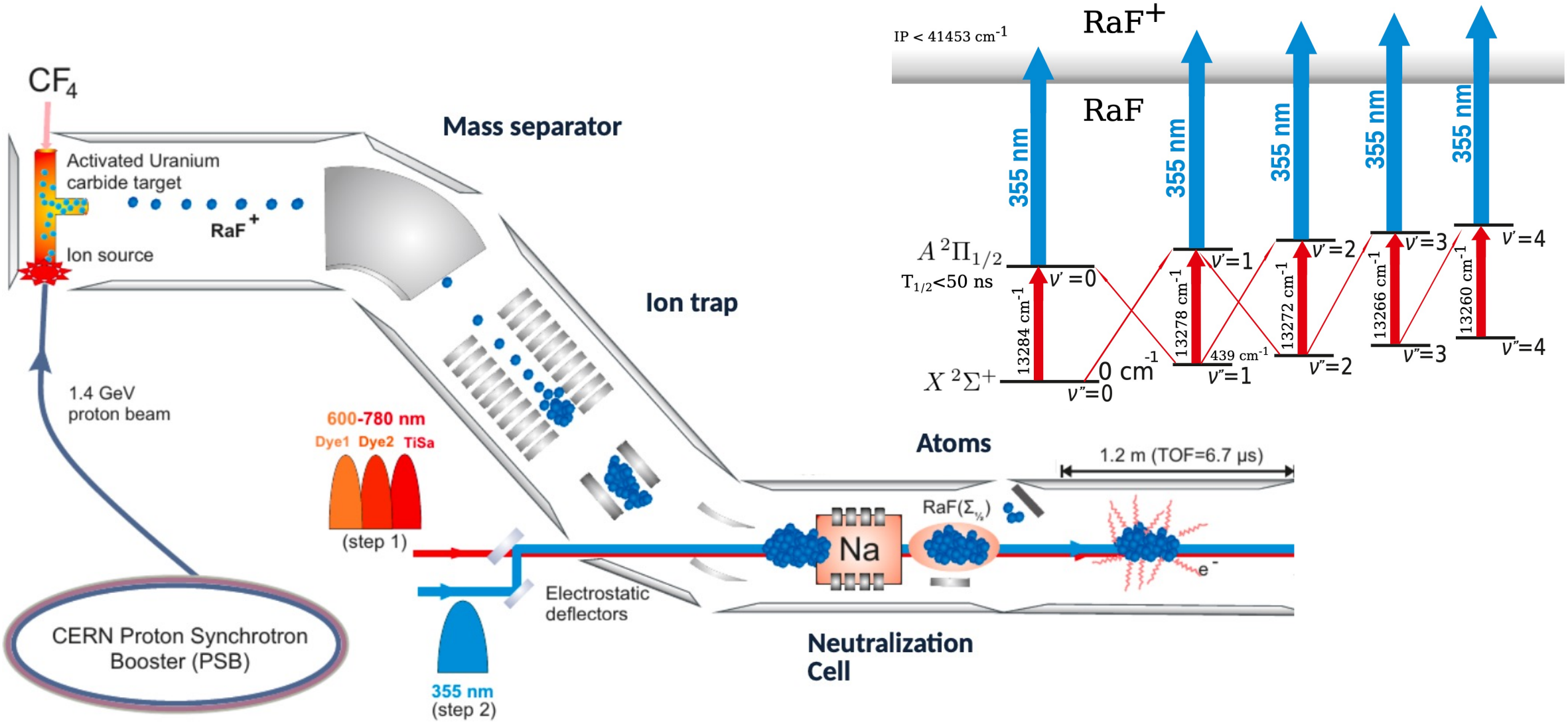

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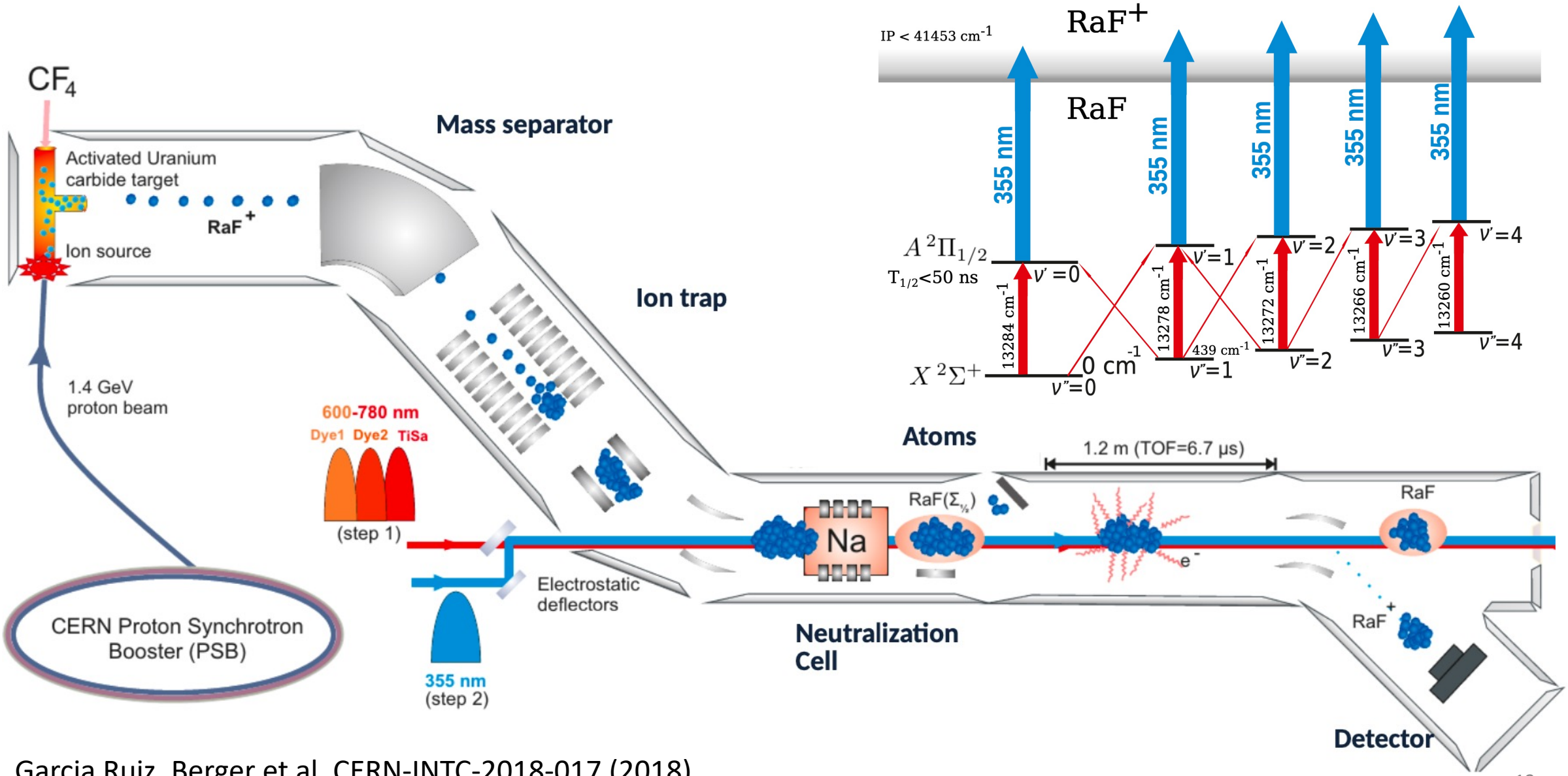

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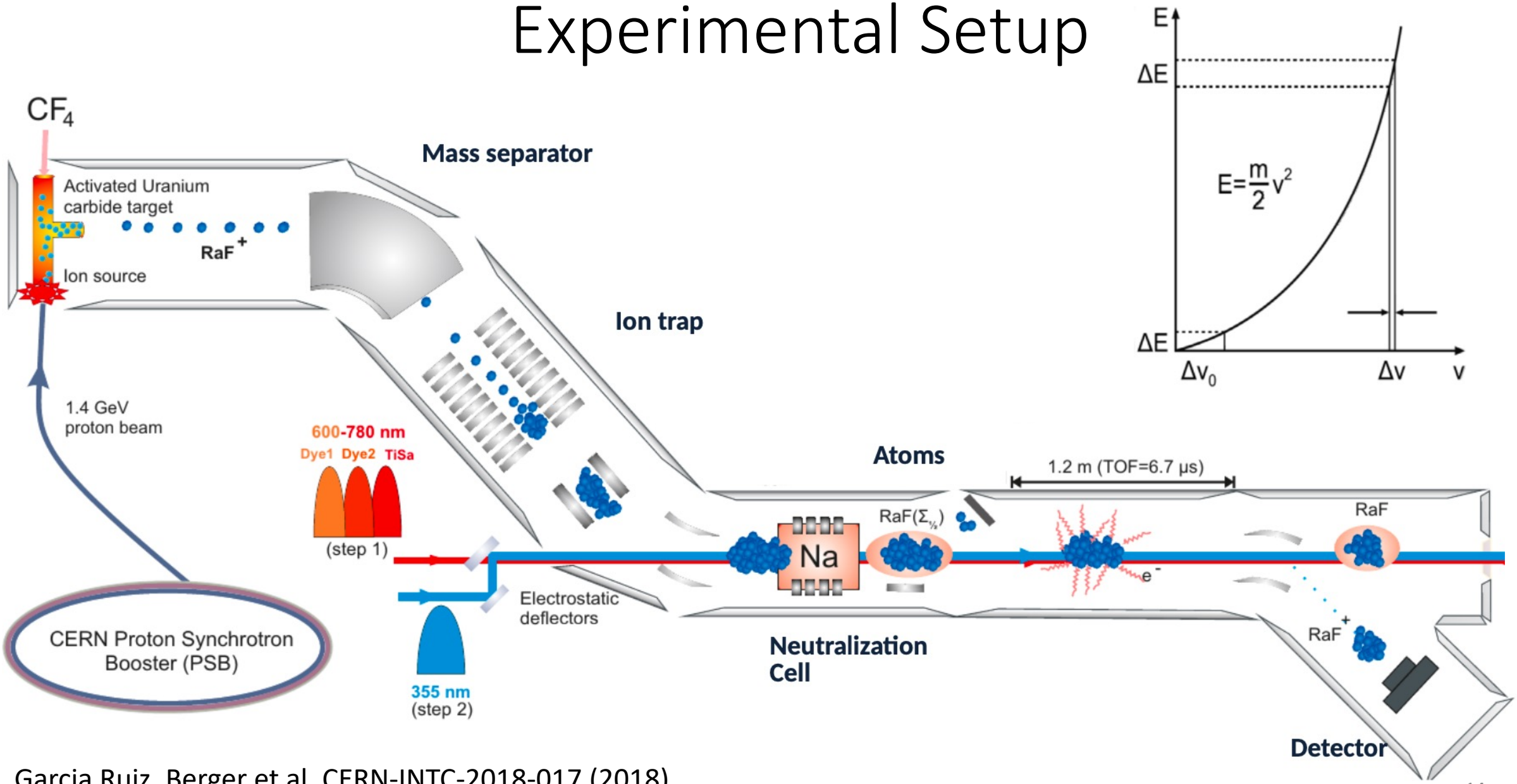

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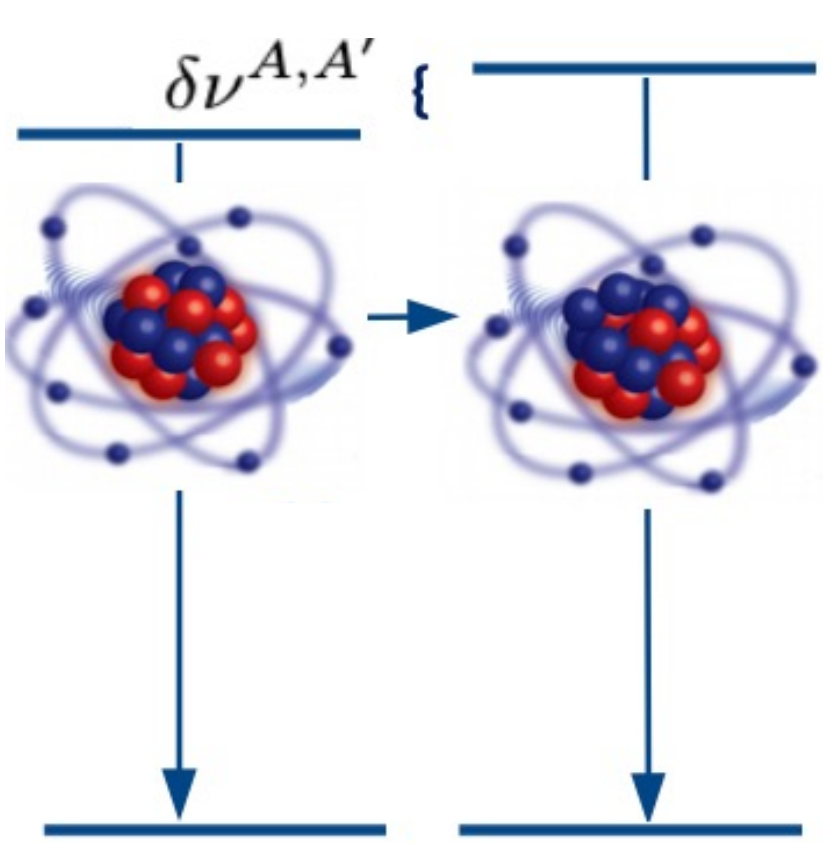
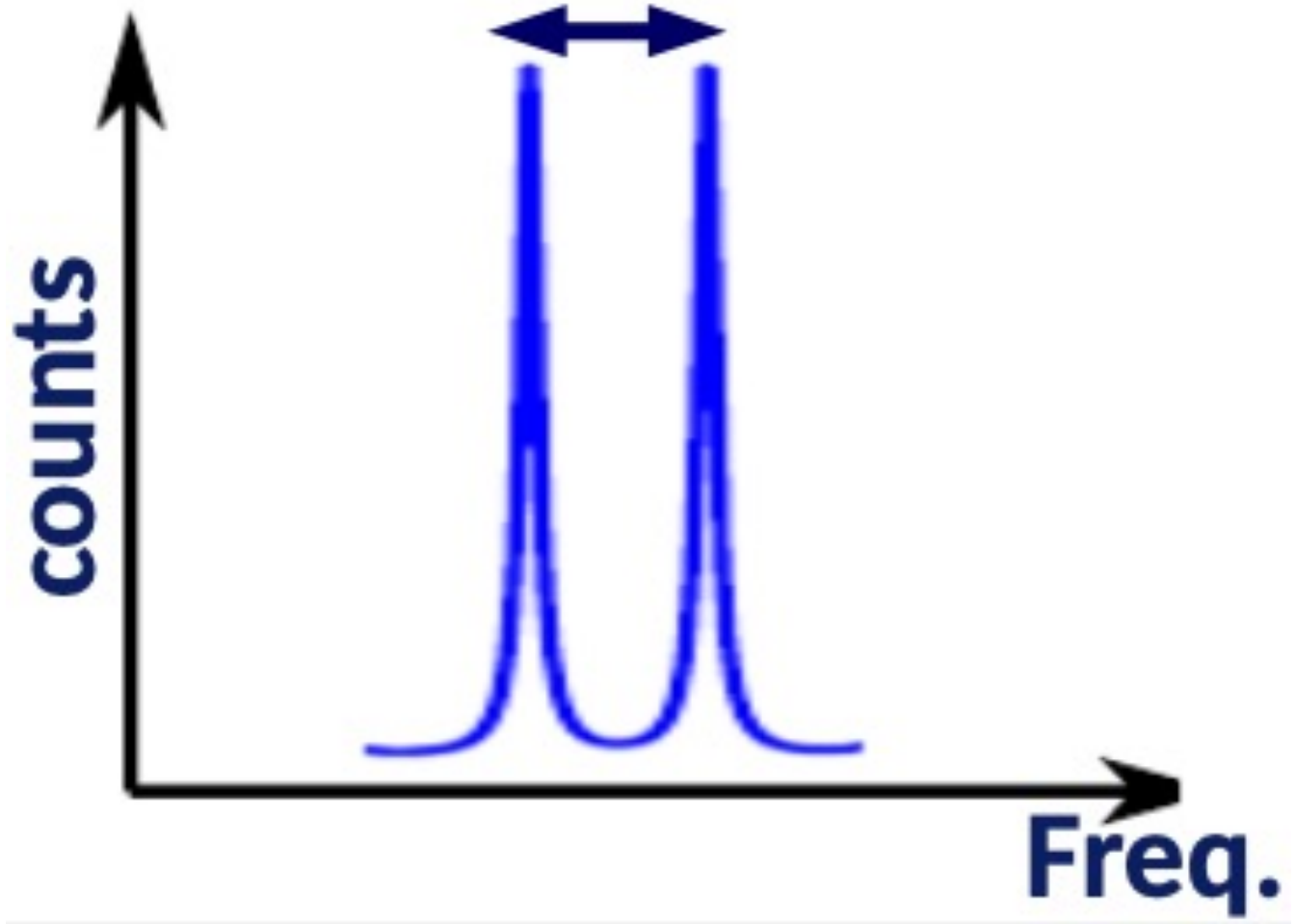
Isotope Shift



$\Rightarrow$



$$\delta\nu_{IS}^{AA'} = K\left(\frac{M_A - M'_A}{M_A M'_A}\right) + F\delta\left\langle r_c^2\right\rangle^{AA'}$$

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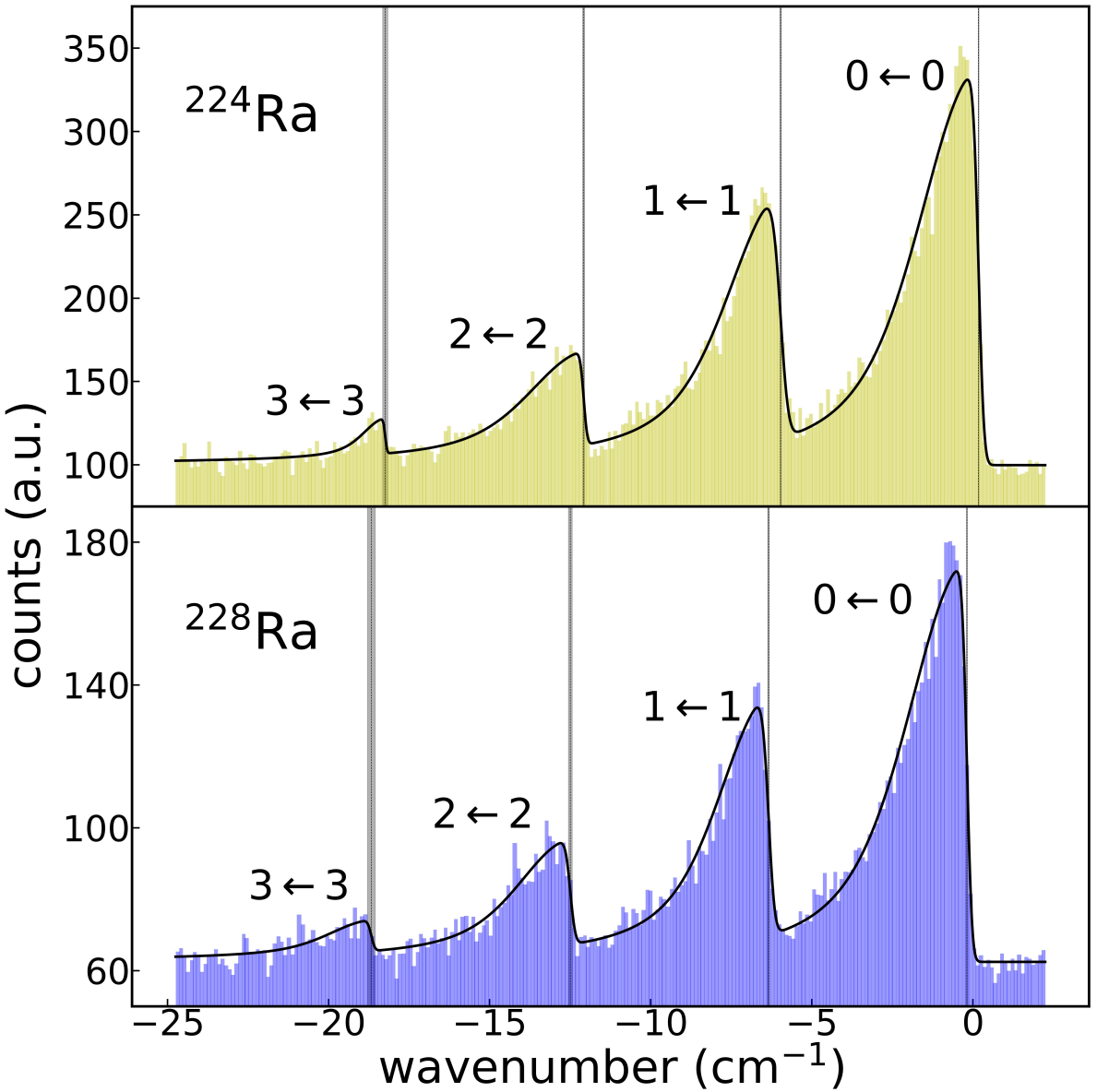
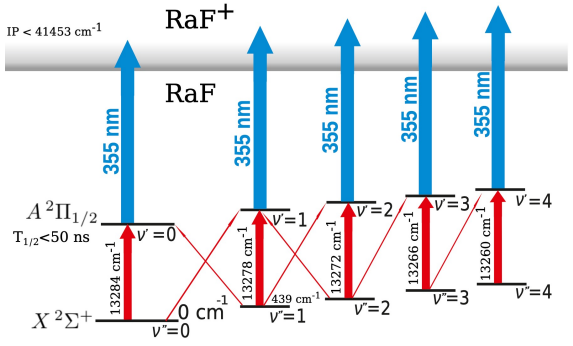
Isotope Shift of RaF - Results





S.M. Udrescu et al., "Isotope Shifts of Radium Monofluoride Molecules", Physical Review Letters, Accepted.

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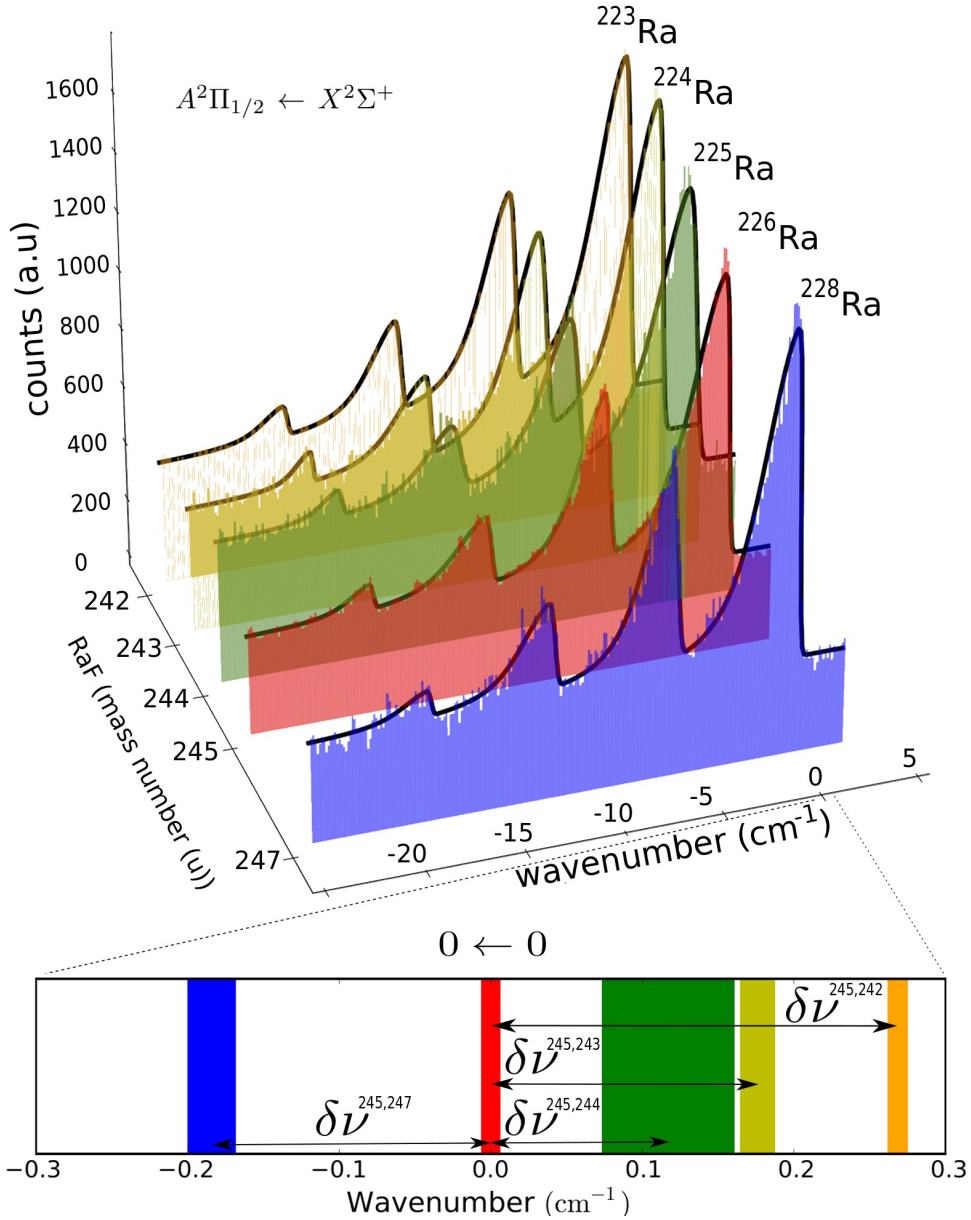

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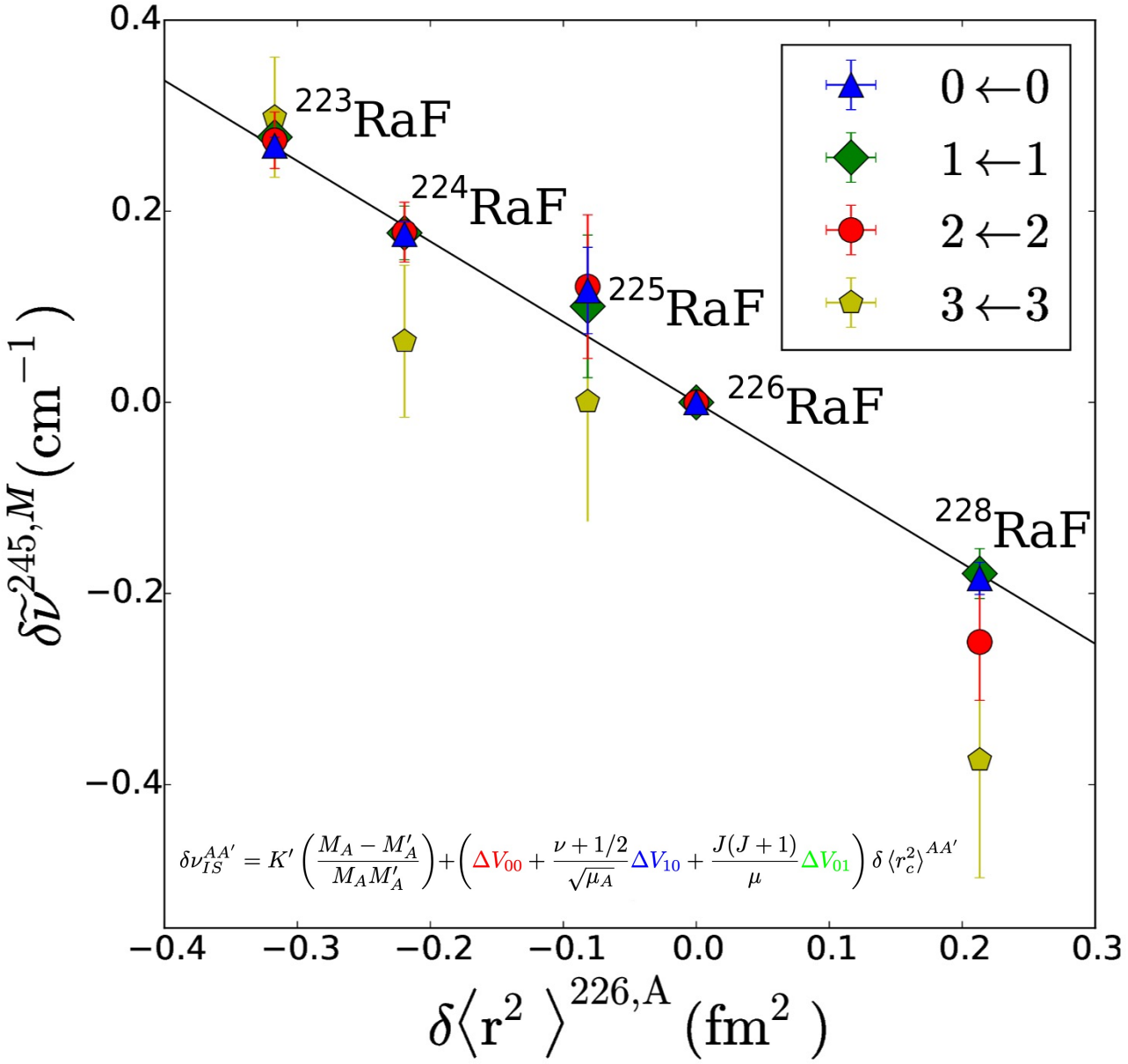

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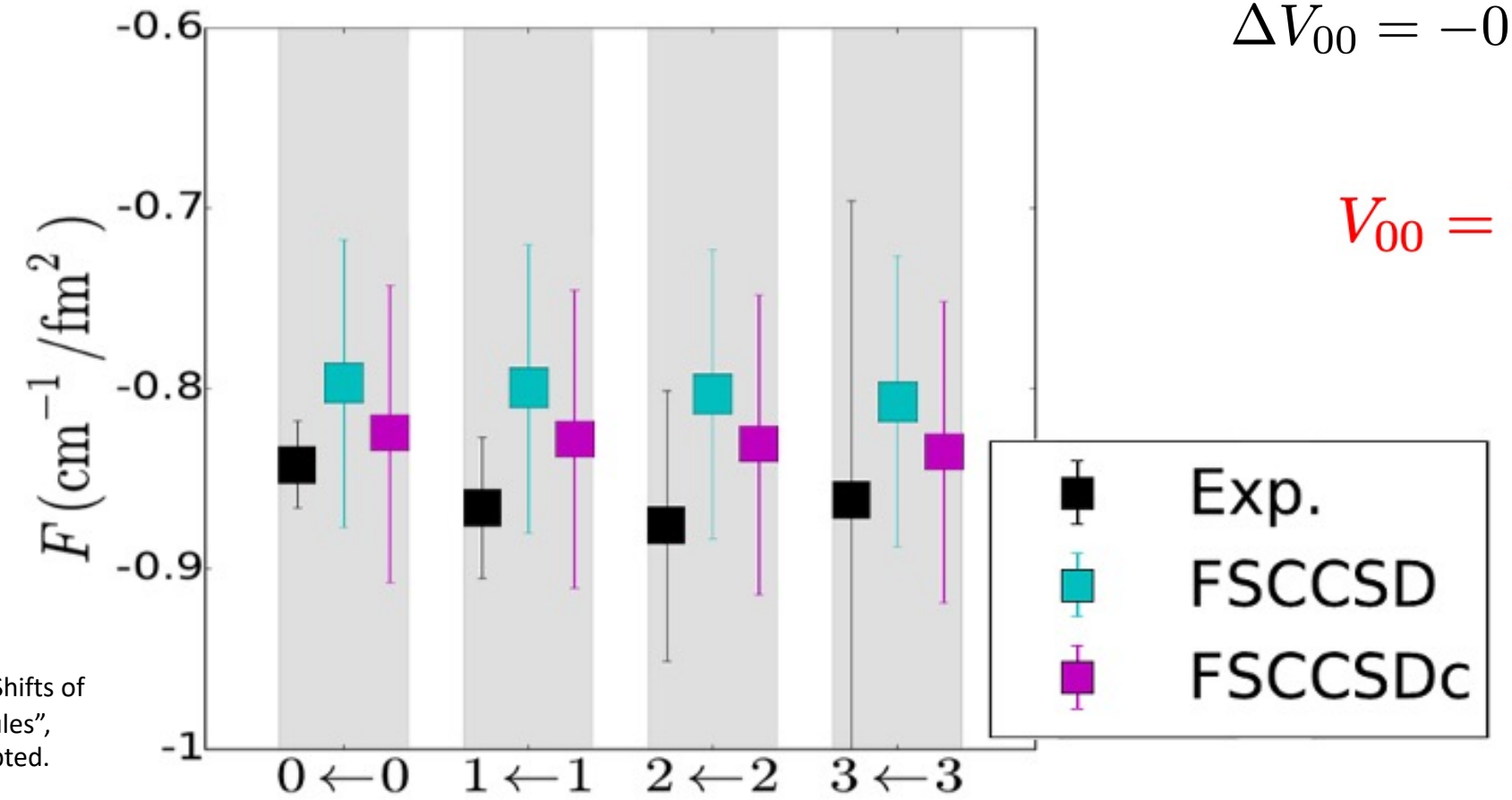

$$\Delta V_{00} = -0.839(33)\frac{\mathrm{cm}^{-1}}{\mathrm{fm}^2}$$

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Future Isotope Shift Measurements

- New opportunities for nuclear studies of heavy elements (e.g. ThO, PaO)

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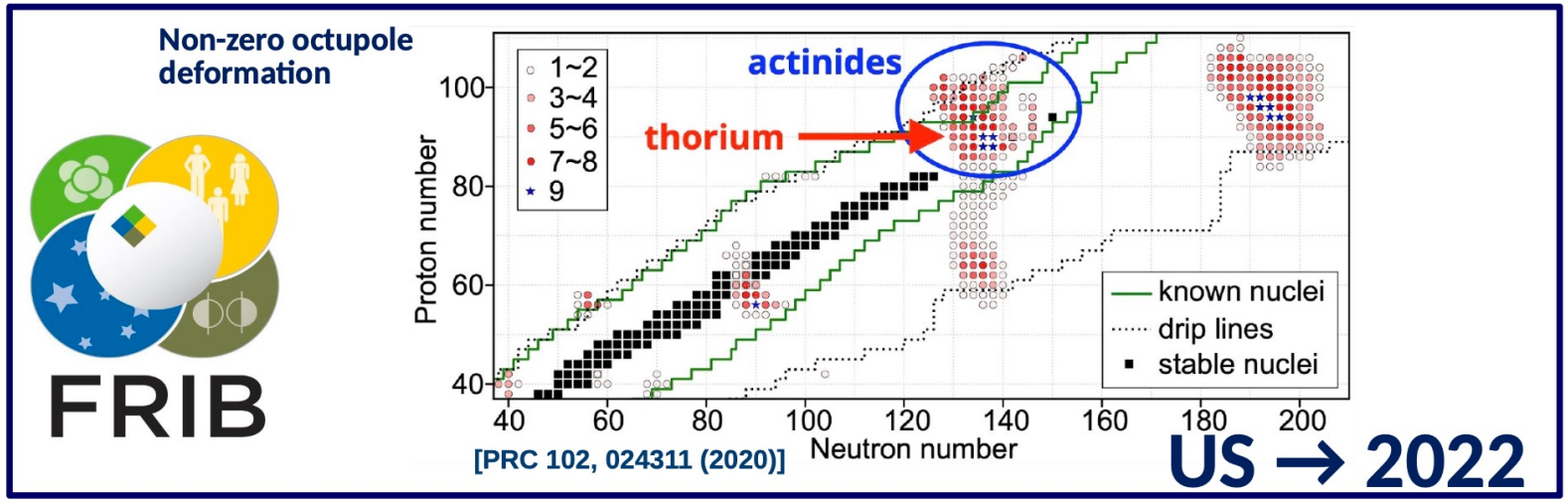

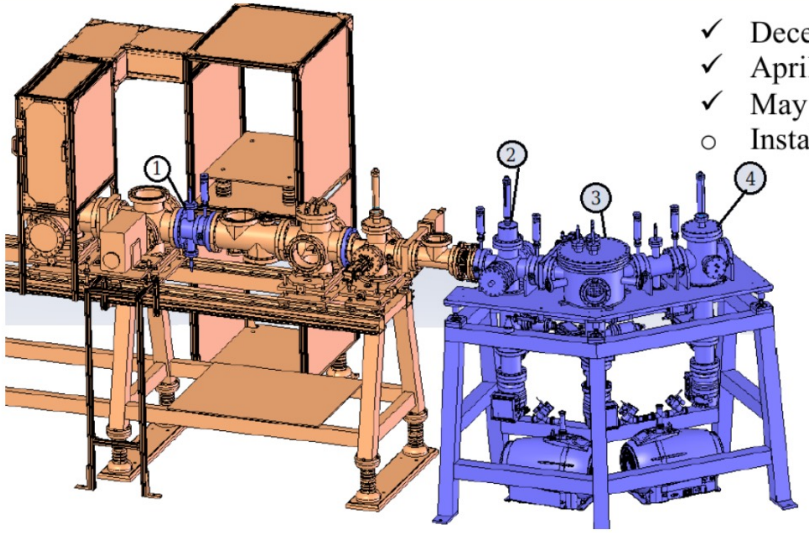

- ✓ December 2020: CRIS project started Dec 2020.
- ✓ April 2021: Beamline design completed.
- ✓ May 2021: Experimental parts are being purchased.
- ○ Installation at FRIB expected in March 2022.

FRIB

Isotope Shift - Open questions

- What elements can be more easily studied in molecular form?

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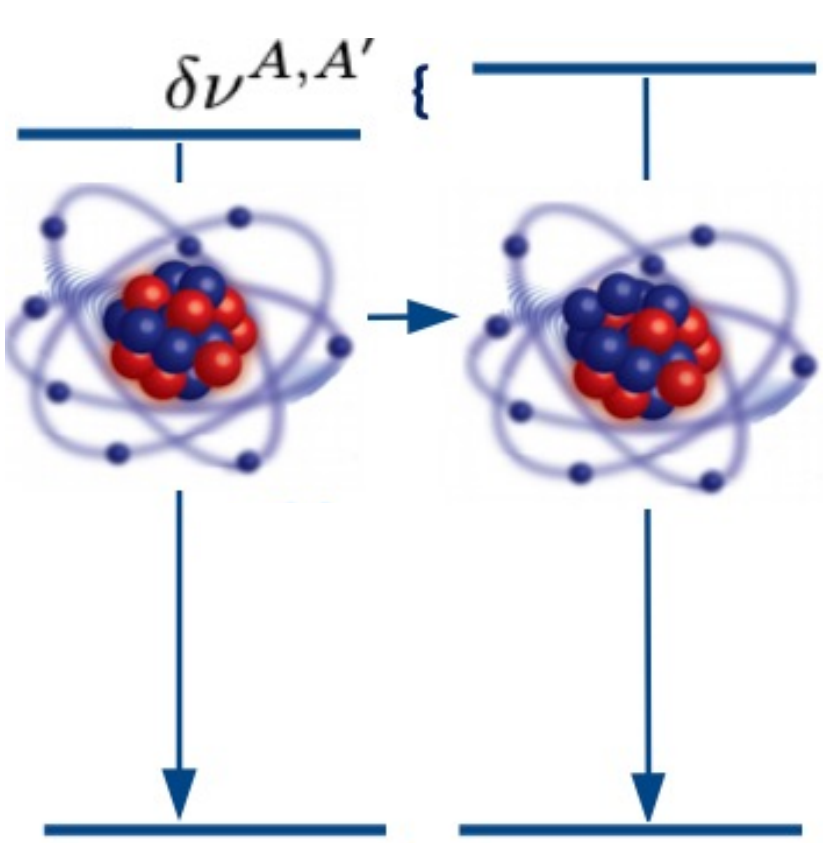
- Can we do systematic studies of spin dependent effects along isotopic chains?
- Can we use isotope shift in molecules to search for new forces and particles (e.g. King plot non-linearities)? What are the advantages compared to atoms?
  - More transitions
  - More sensitivity to lighter mediators

Isotope Shift – King plot nonlinearity



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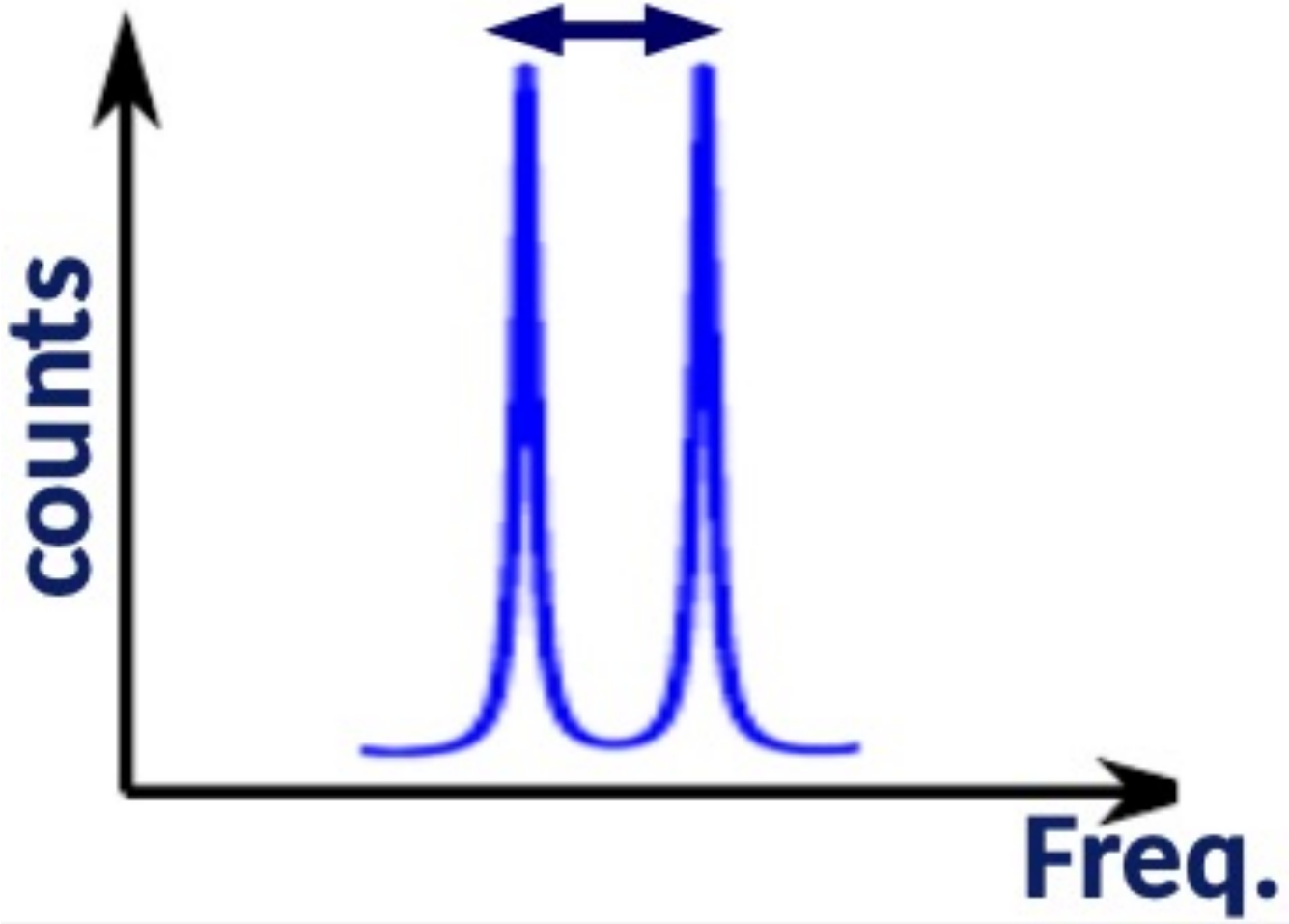

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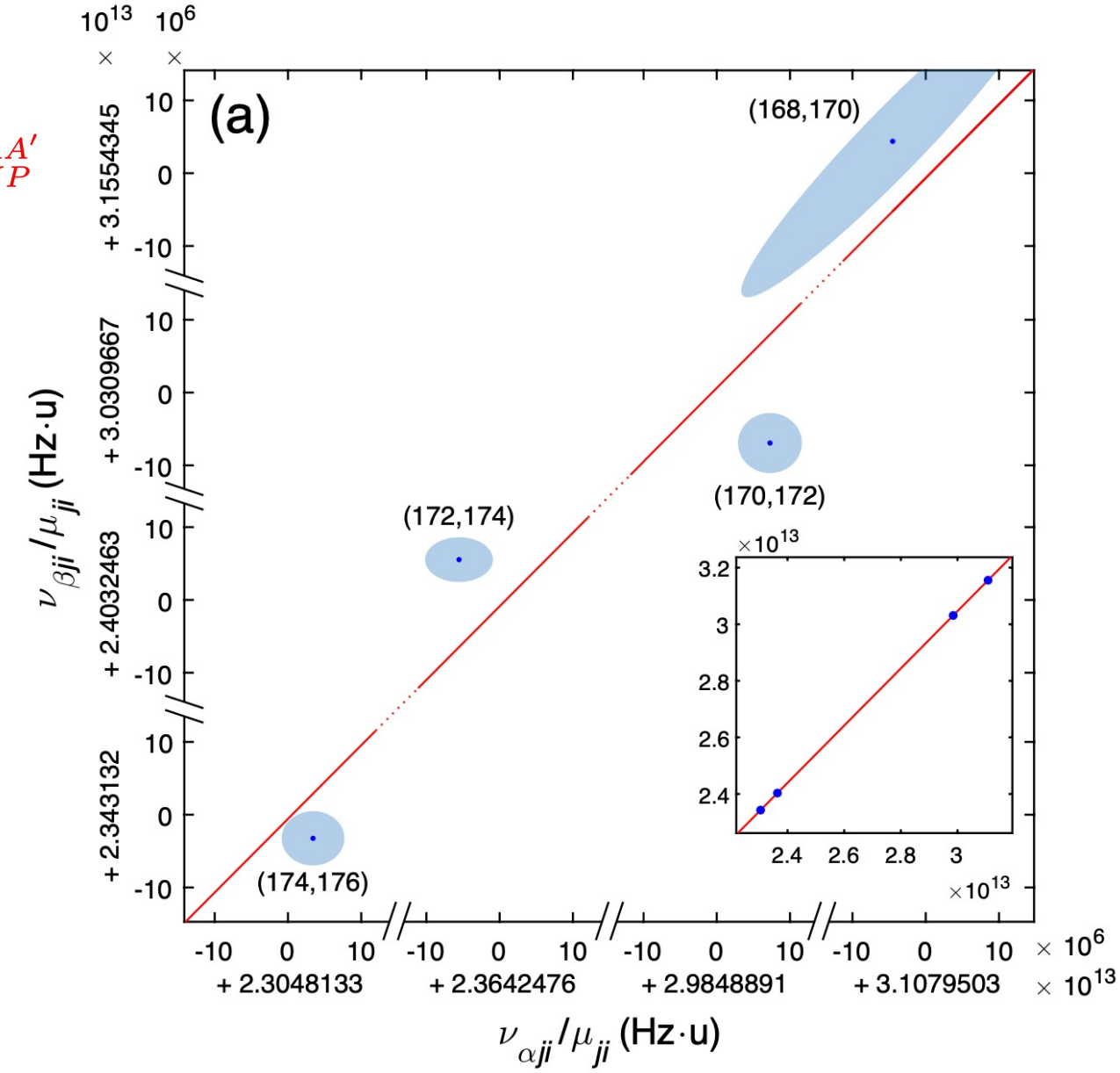
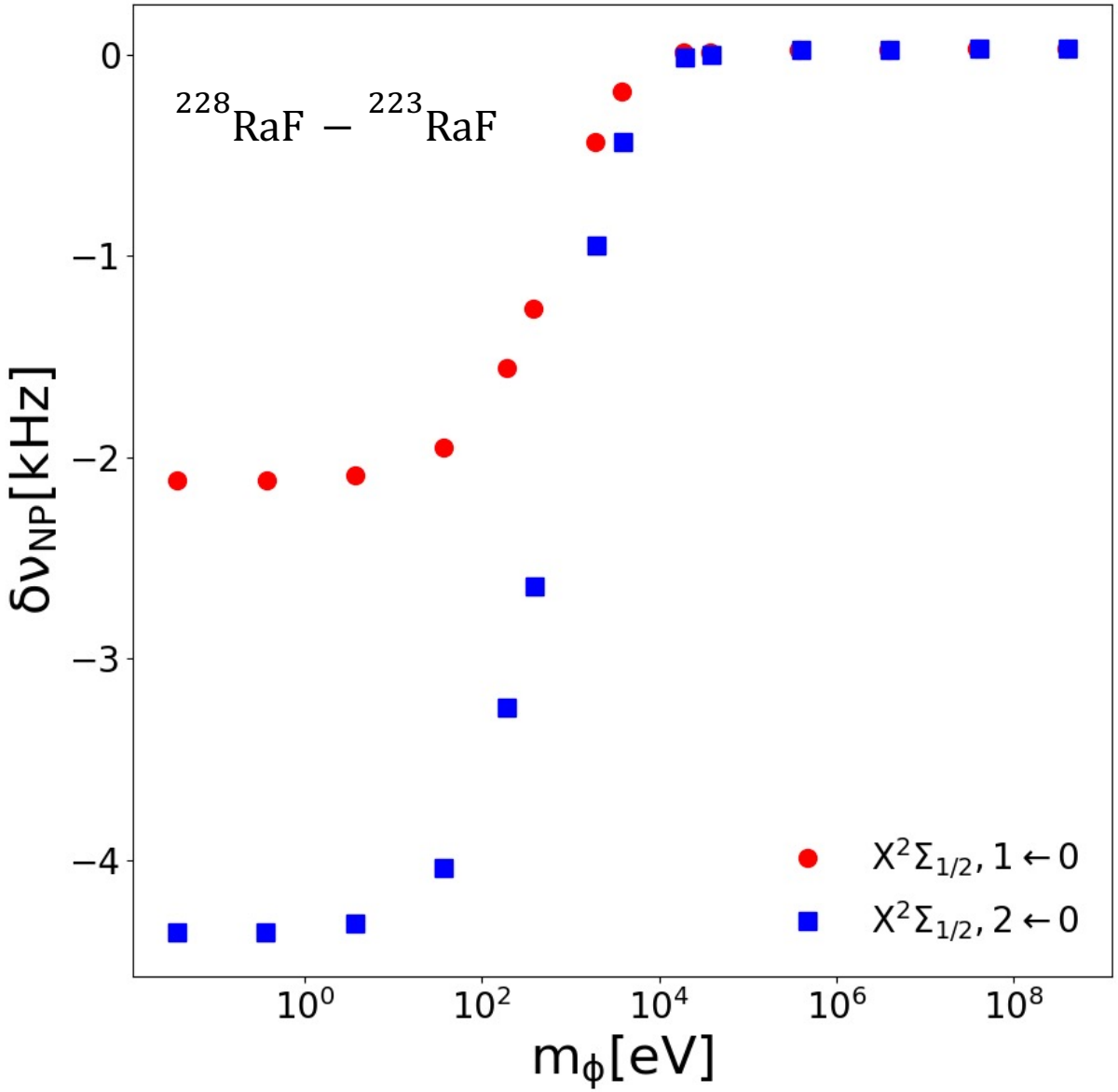


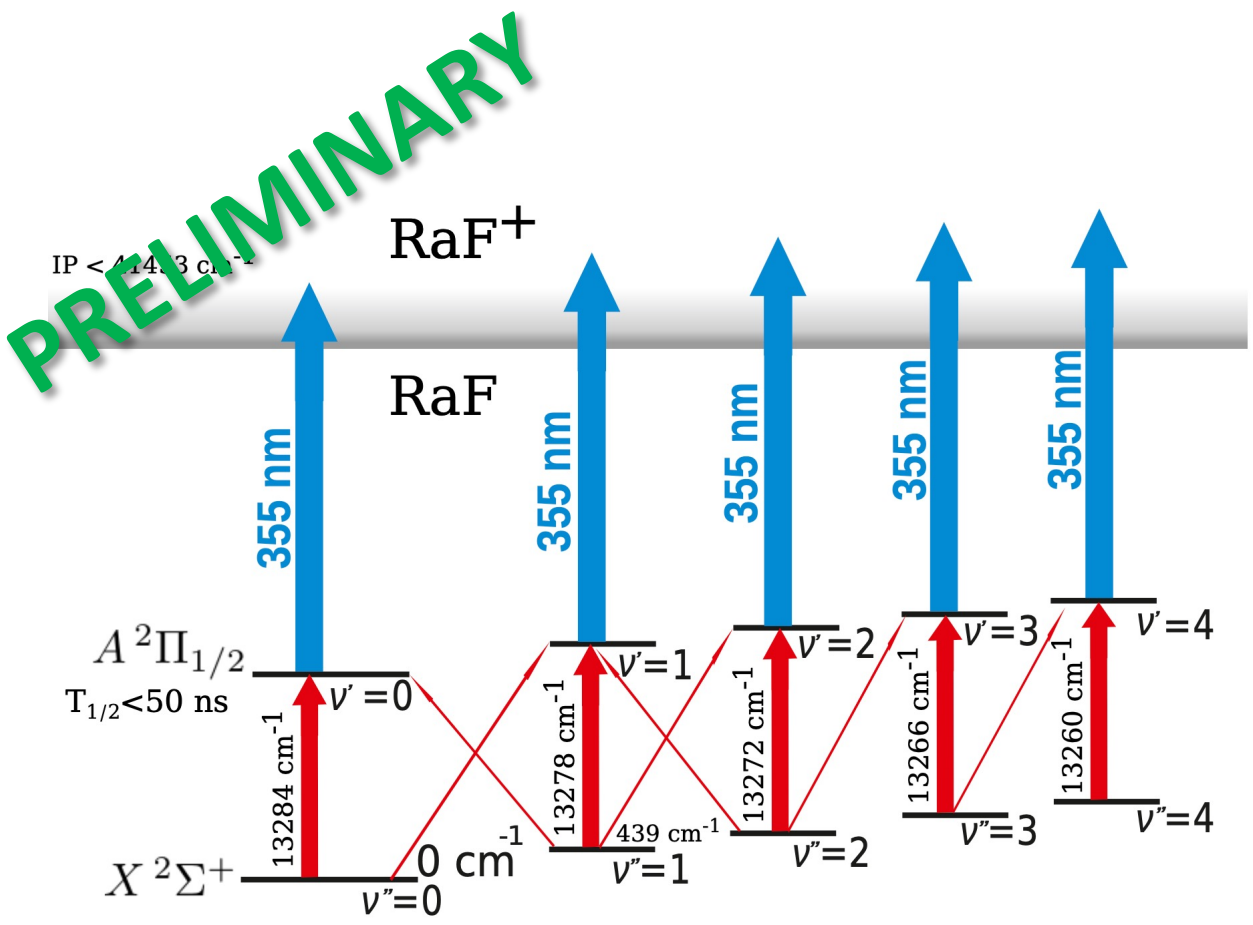
I. Counts, et al., "Evidence for nonlinear isotope shift in Yb+ search for new boson," Phys. Rev. Lett. 125, 123002 (2020).

Isotope Shift – King plot nonlinearity



K. Gaul, R. Berger

High Resolution spectroscopy of ^{226}RaF



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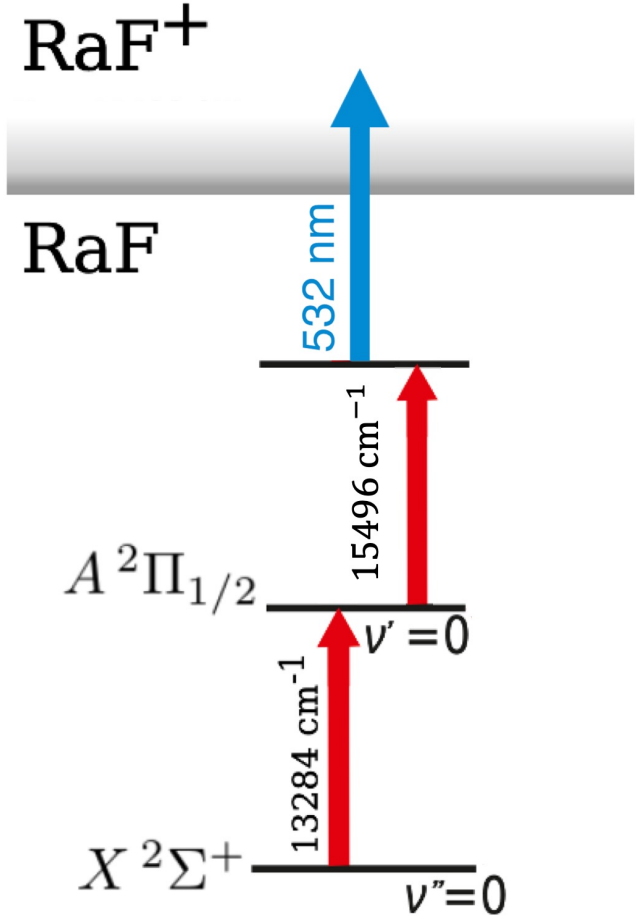
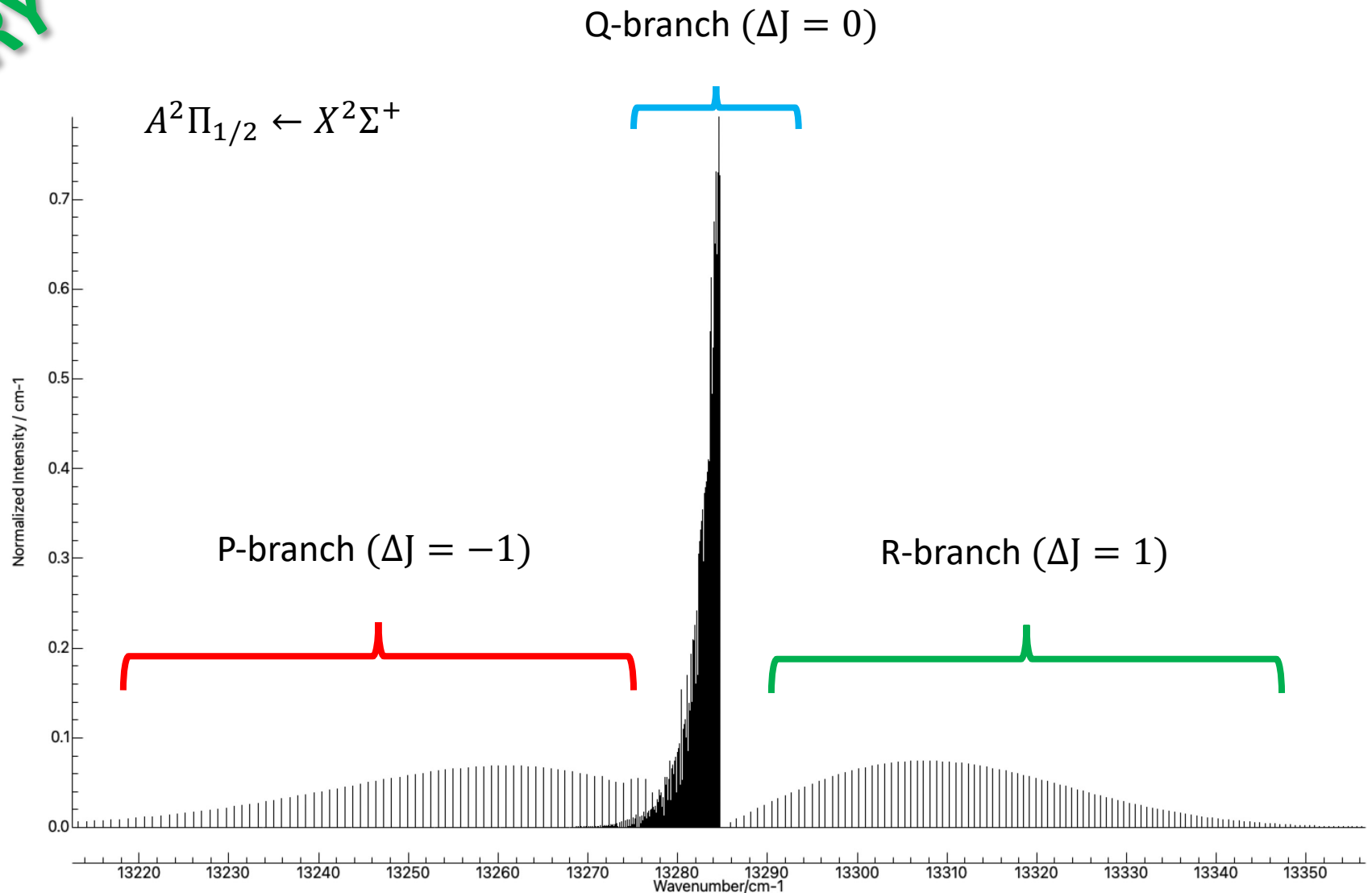


10 GHZ

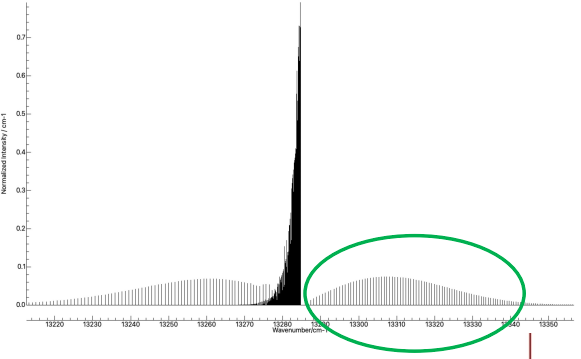
50 MHZ

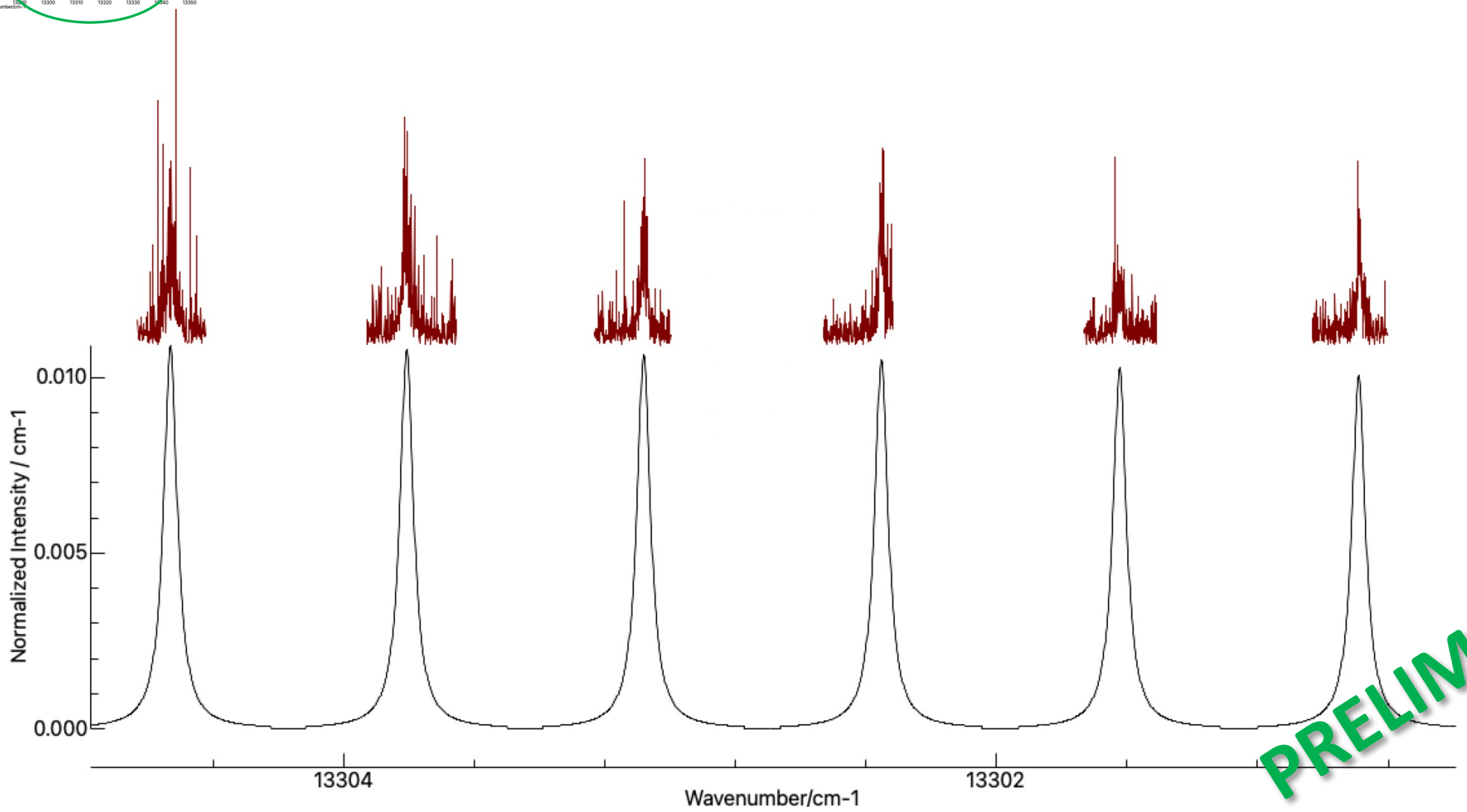
High Resolution spectroscopy of ^{226}RaF

PRELIMINARY

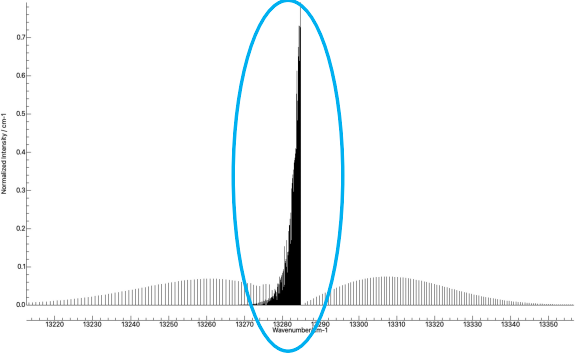


Results (R-branch)

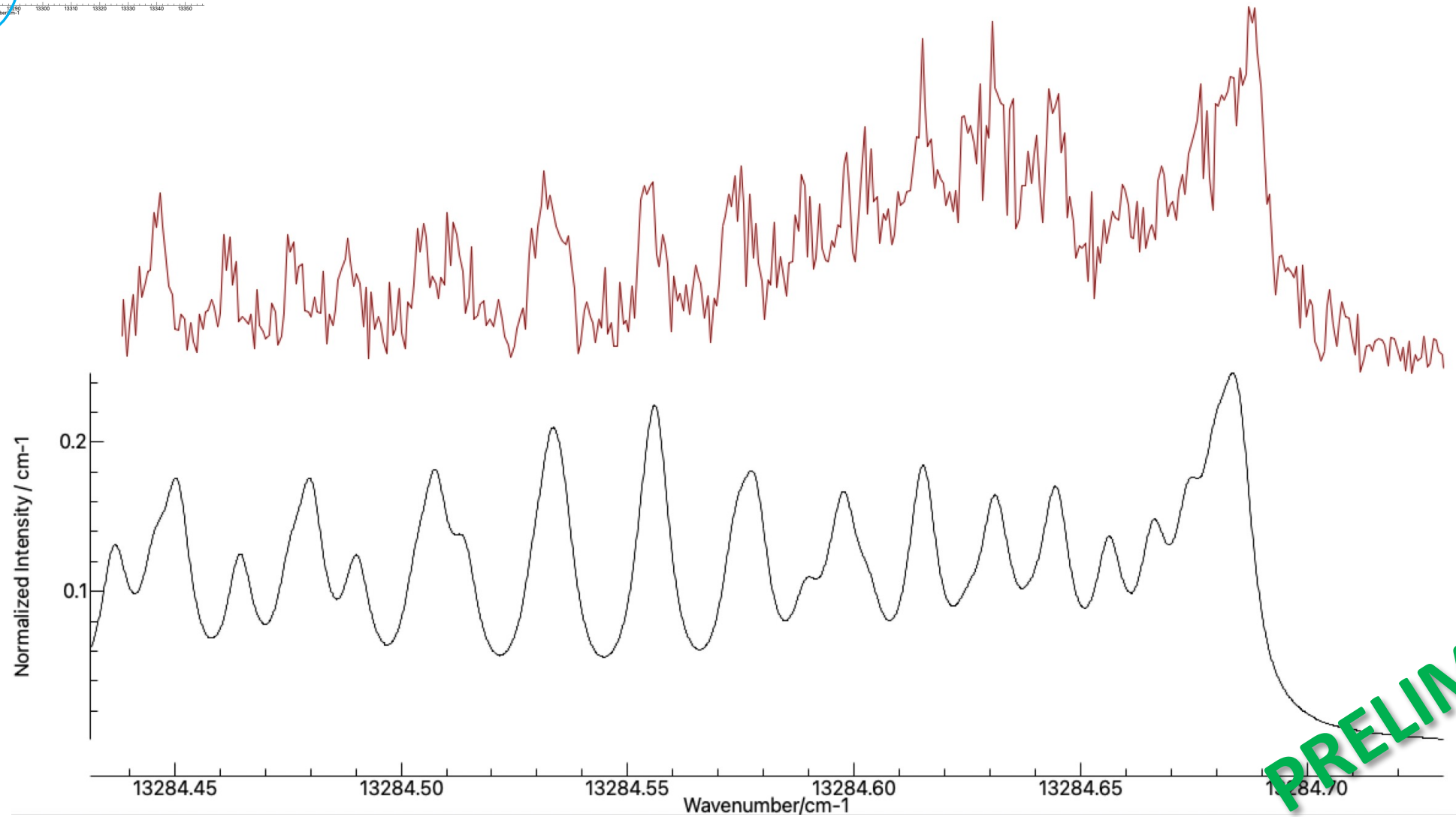




PRELIMINARY



Results (Q-branch)



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- Increased statistics for the fine structure of $^{226}\mathrm{RaF}$ (+ higher lying electronic states)

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- Better multi-step excitation scheme development

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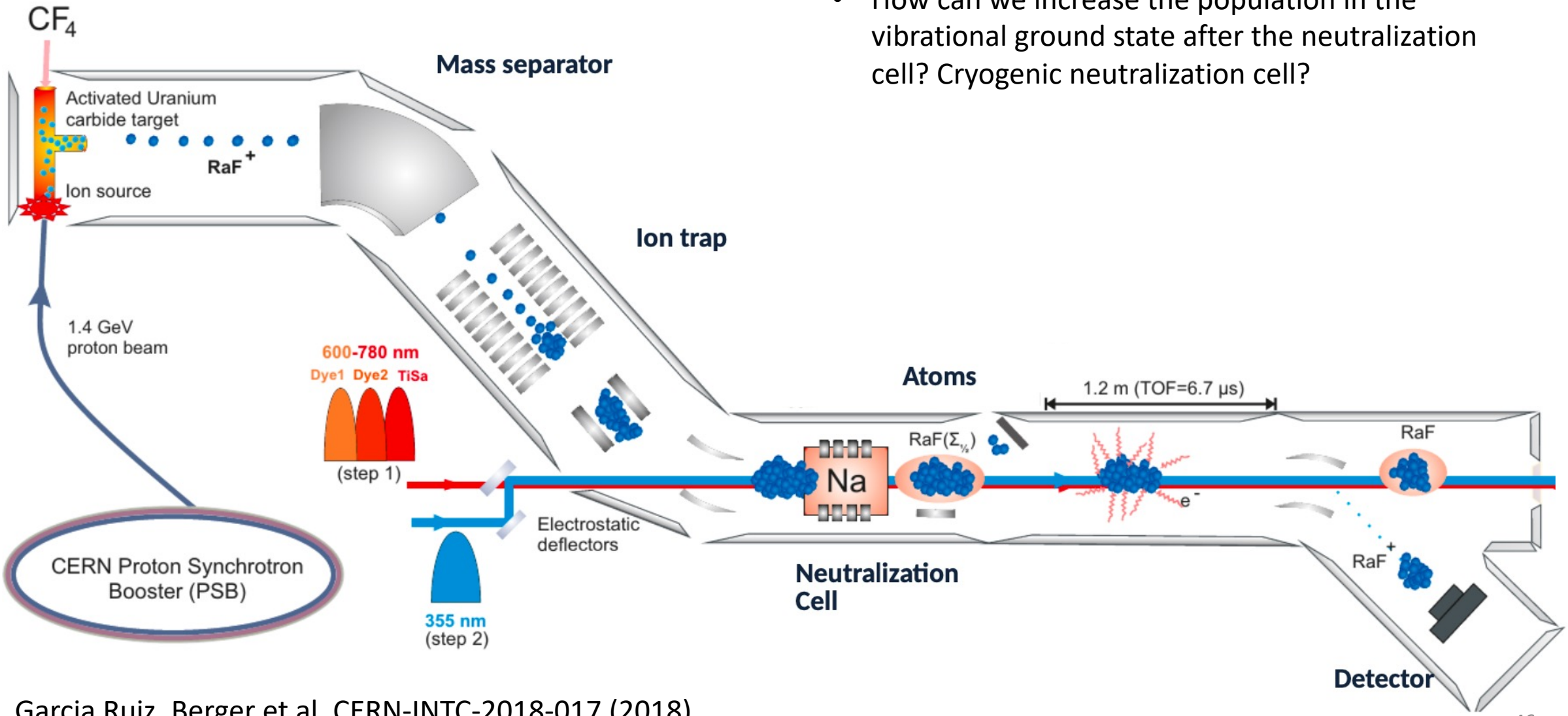
- Measurement of more RaF isotopologues at higher resolution
- Measurement of the RaF ionization potential
- Better multi-step excitation scheme development

Nov-Dec 2021
CRIS-ISOLDE,
CERN

Outlook (Long-term)

- How can we increase the population in the vibrational ground state after the neutralization cell? Cryogenic neutralization cell?



Garcia Ruiz, Berger et al. CERN-INTC-2018-017 (2018)

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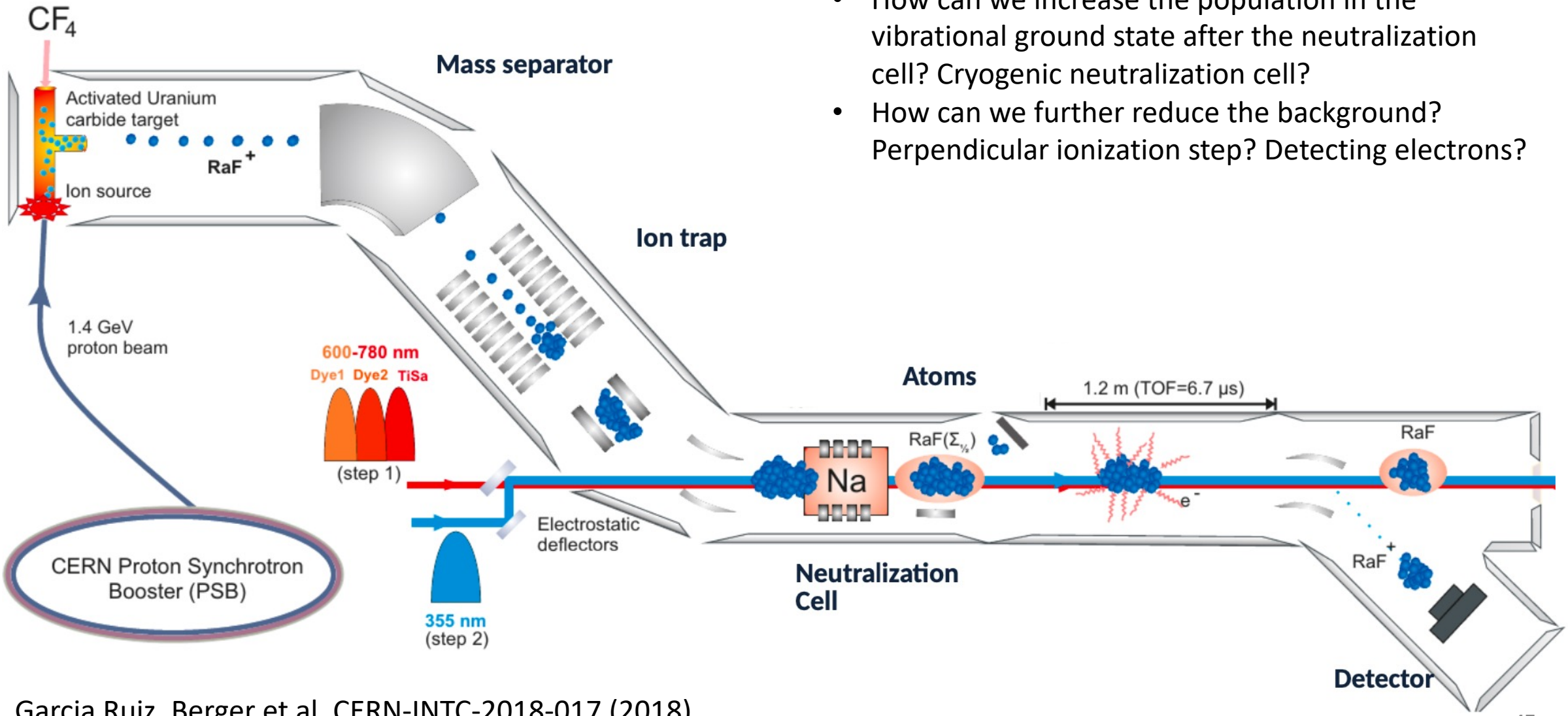
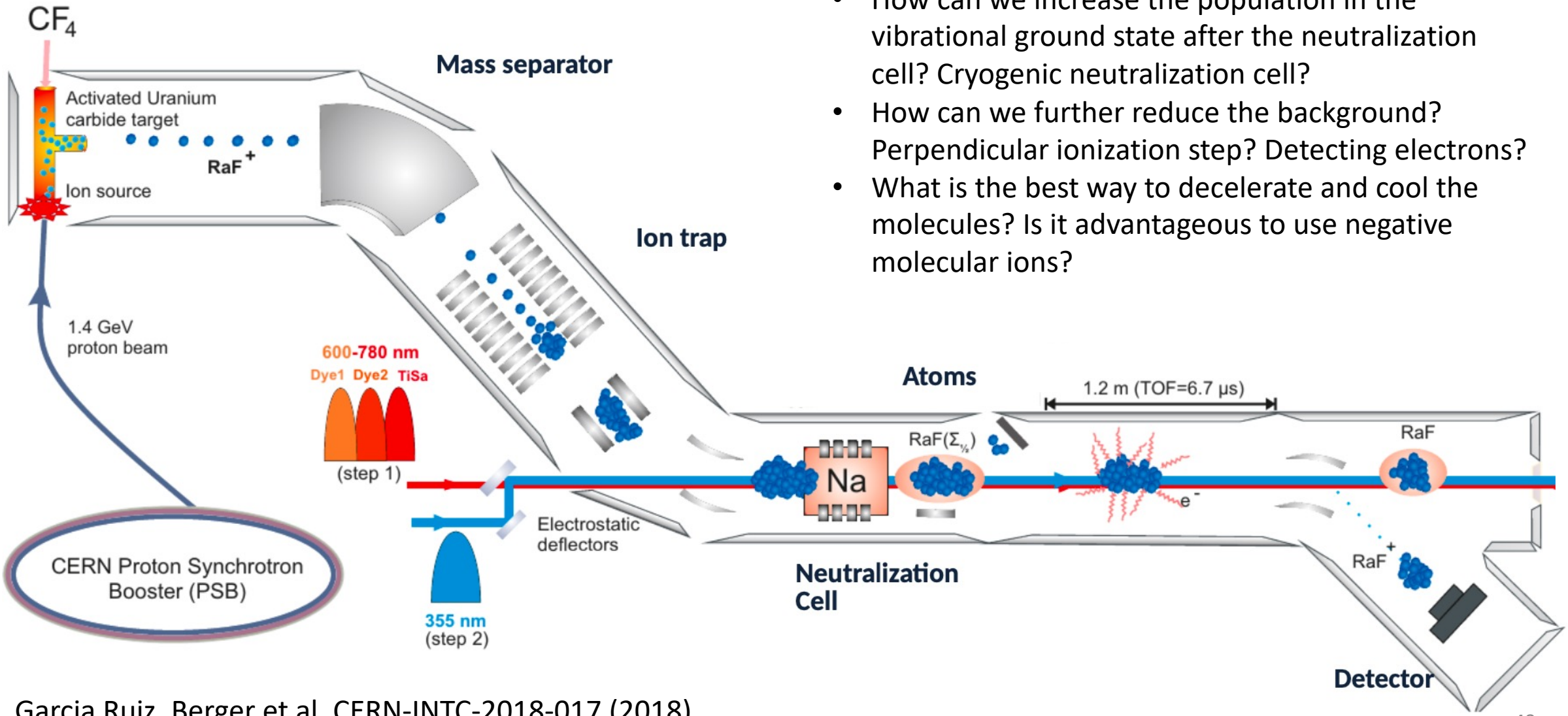


Garcia Ruiz, Berger et al. CERN-INTC-2018-017 (2018)

Outlook (Long-term)



- How can we increase the population in the vibrational ground state after the neutralization cell? Cryogenic neutralization cell?
- How can we further reduce the background? Perpendicular ionization step? Detecting electrons?
- What is the best way to decelerate and cool the molecules? Is it advantageous to use negative molecular ions?

Garcia Ruiz, Berger et al. CERN-INTC-2018-017 (2018)

Outlook (Long-term)

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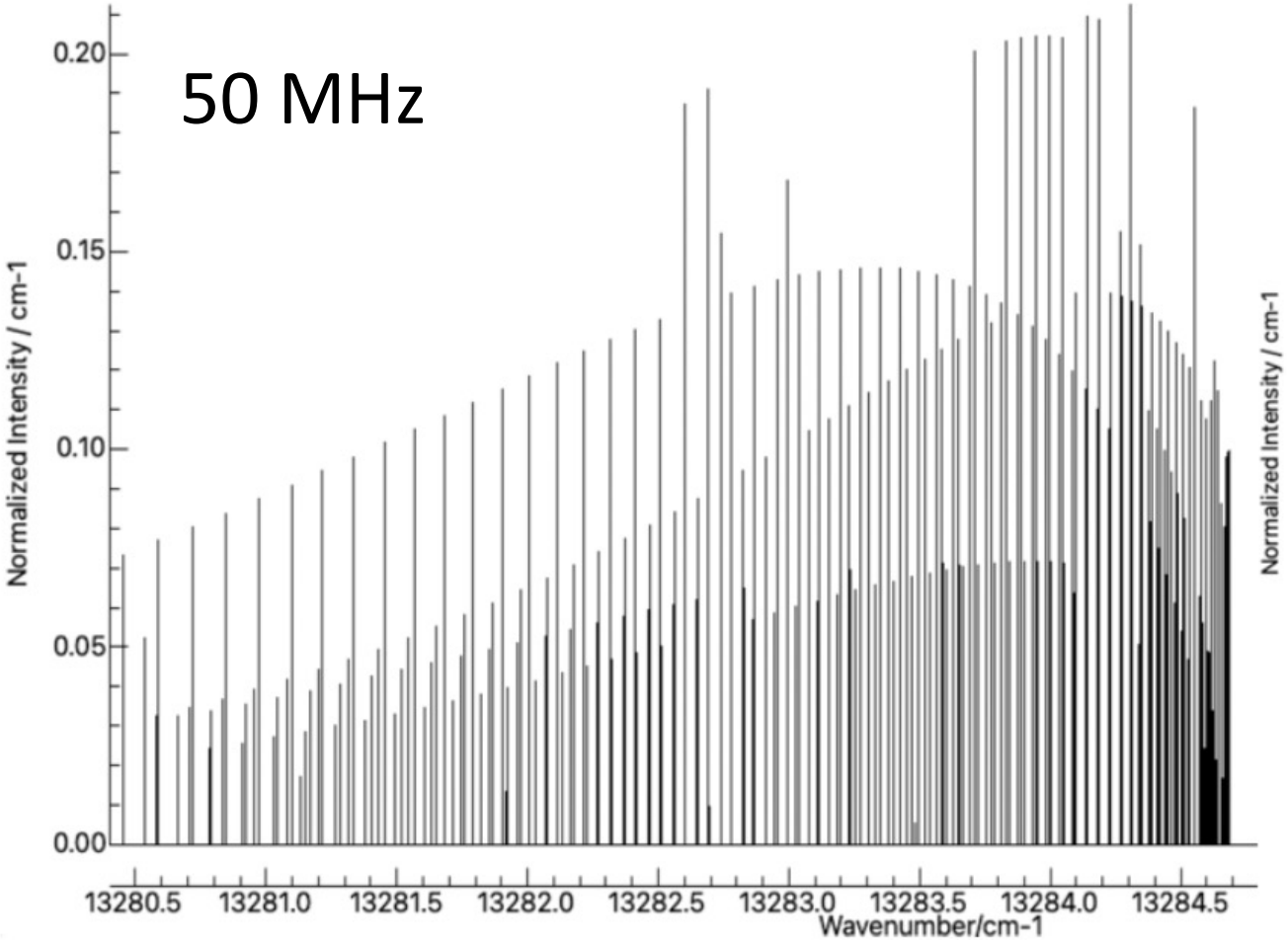
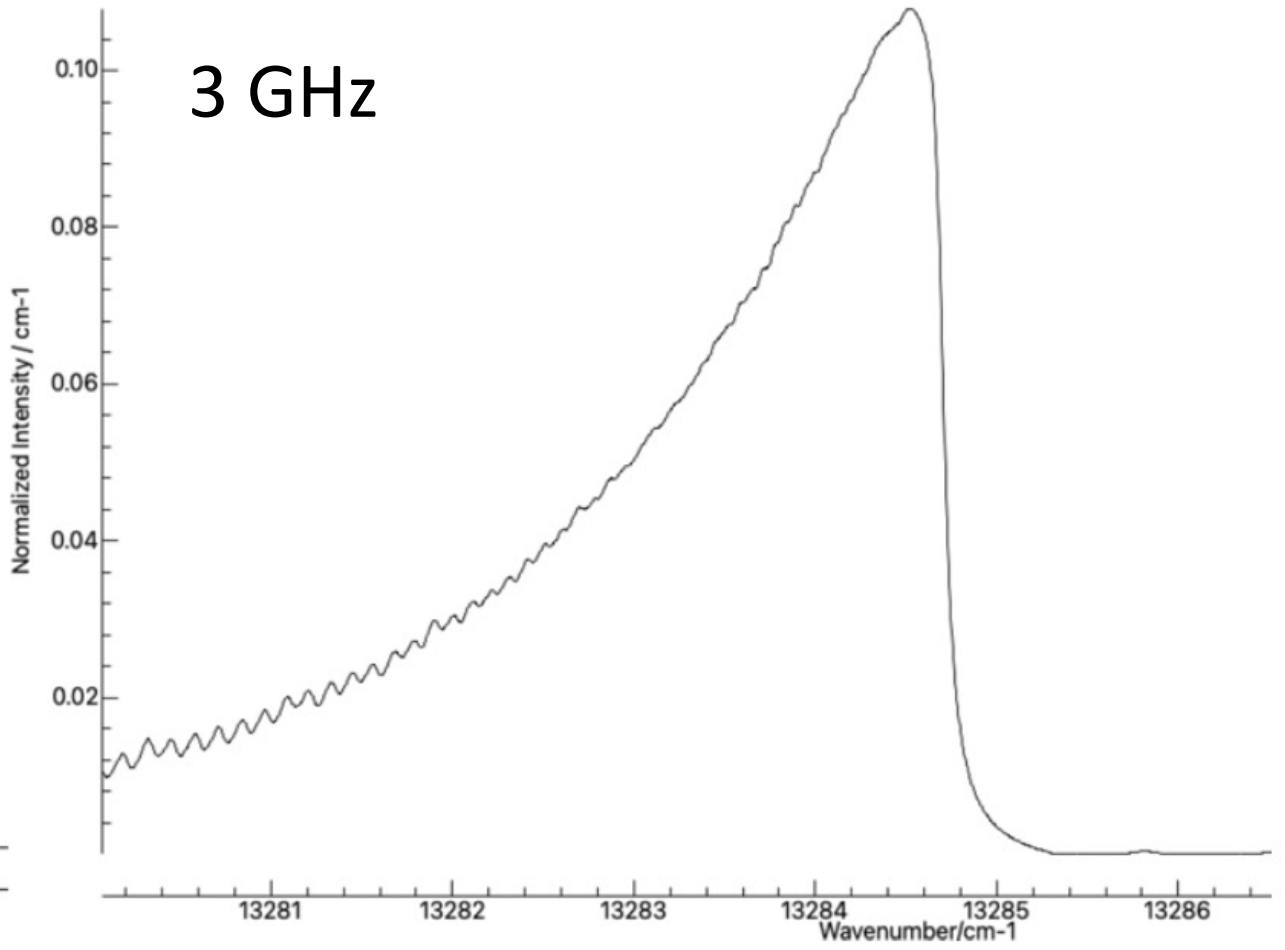
- What else?

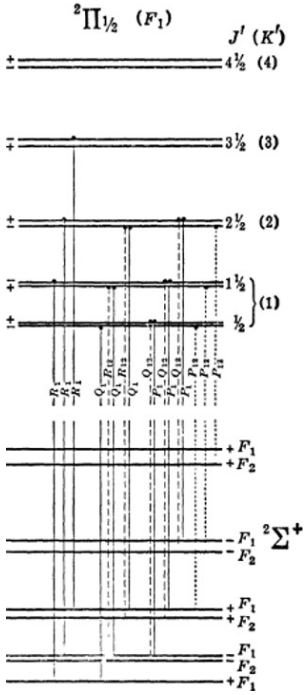
Thank you!

Backup slides


50 MHz
Normalized Intensity / cm-1
0.20
0.15
0.10
0.05
0.00
13280.5
13281.0
13281.5
13282.0
13282.5
13283.0
13283.5
13284.0
13284.5
Wavenumber/cm-1


3 GHz
Normalized Intensity / cm-1
0.10
0.08
0.06
0.04
0.02
13281
13282
13283
13284
13285
13286
Wavenumber/cm-1

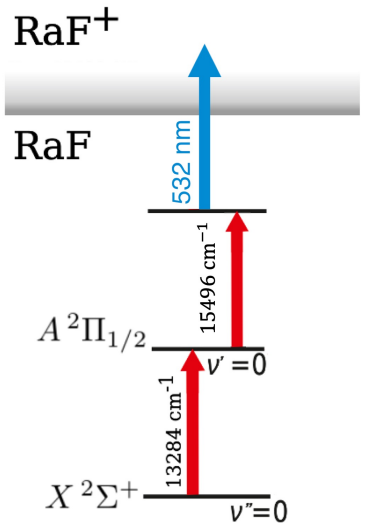
Predicted rotational spectrum



Gerhard Herzberg, The spectra and structures of simple free radicals: an introduction to molecular spectroscopy (Courier Corporation, 1988).

Measured Isotope Shift

M	$\delta\widetilde{\nu}^{245,M}_{0\leftarrow 0}$	$\delta\widetilde{\nu}^{245,M}_{1\leftarrow 1}$	$\delta\widetilde{\nu}^{245,M}_{2\leftarrow 2}$	$\delta\widetilde{\nu}^{245,M}_{3\leftarrow 3}$
242	0.269(9)	0.278(14)	0.274(30)	0.298(63)
243	0.176(13)	0.177(28)	0.178(31)	0.064(80)
244	0.117(46)	0.100(74)	0.121(75)	0.001(131)
245	0	0	0	0
247	−0.184(17)	−0.179(26)	−0.251(60)	−0.374(123)

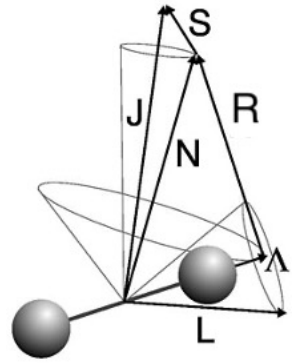


RaF Hamiltonian

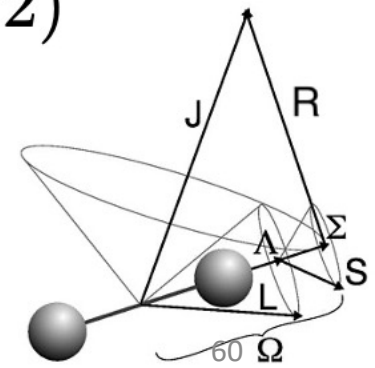
$$H = H_{rot} + H_{SO} + H_{SR} + H_{\Lambda-doubling}$$

$X^2\Sigma$ (Hund case b) $E_{X^2\Sigma}(J)$

$$\begin{cases} B_g(J+1/2)(J-1/2) + \frac{\gamma}{2}(J-1/2) \\ B_g(J+1/2)(J+3/2) - \frac{\gamma}{2}(J+3/2) \end{cases}$$



$A^2\Pi$ (Hund case a) $E_{A^2\Pi_{1/2}}(J) = T - \frac{A}{2} + B_e J(J+1) \pm \frac{p}{2}(J+1/2)$



Centrifugal corrections: $DJ^2(J+1)^2$

Fitting procedure

Parameters needed:

- Ground electronic state:
  - $\mathrm{B_g}$ – free parameter [1]
  - $\mathrm{D_g}$ – low resolution data
  - γ – theory
- Excited electronics state:
  - $\mathrm{B_e}$ – high resolution data
  - $\mathrm{D_e}$ – low resolution data
  - p – theory
  - T – low resolution data

Constraints:

$$\gamma = -2\mathrm{B_g}\Delta\mathrm{g}_\perp \text{ [1]}$$

$$p = -2B_e$$

$$D = \frac{4B^3}{\omega^2}$$

[1] TA Isaev and R Berger, “Lasercooled radium monofluoride: A molecular all-in-one probe for new physics,” arXiv preprint arXiv:1302.5682 (2013).

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  - T – low resolution data

Constraints:

$$\nu_{R(J+2)} - 2\nu_{R(J+1)} + \nu_{R(J)} = 2(B_e - B_g)$$

^{223}RaF

- Fine structure similar to ^{226}RaF → small isotope shift corrections

Can be ignored

$$\mathcal{H}_{\mathrm{hfs}} = \sum_{\alpha} \left\{ a_{\eta v}^{\alpha} \mathrm{T}_{q=0}^{1}(\boldsymbol{I}_{\alpha}) \mathrm{T}_{q=0}^{1}(\boldsymbol{L}) + b_{\mathrm{F}\eta v}^{\alpha} \mathrm{T}^{1}(\boldsymbol{I}_{\alpha}) \cdot \mathrm{T}^{1}(\boldsymbol{S}) + (\sqrt{6}/3) c_{\eta v}^{\alpha} \mathrm{T}_{q=0}^{2}(\boldsymbol{I}_{\alpha}, \boldsymbol{S}) + {\sum_{q=\pm 1}}' \exp(-2\mathrm{i}q\phi) d_{\eta v}^{\alpha} \mathrm{T}_{2q}^{2}(\boldsymbol{I}_{\alpha}, \boldsymbol{S}) \right\} \quad (7.191)$$

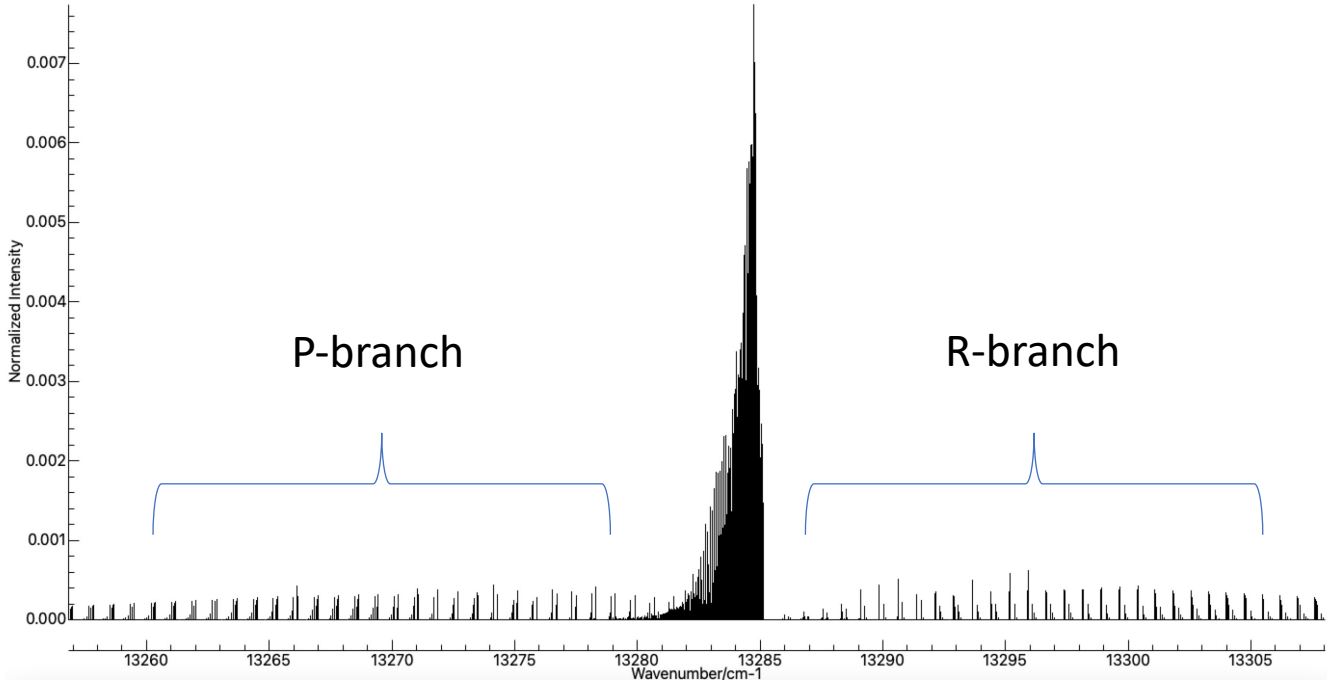
Non-zero only in $\Pi_{1/2}$ state

$$\mathcal{H}_{\mathrm{Q}} = \sum_{\alpha} \frac{eQ_{\alpha}}{4I_{\alpha}(2I_{\alpha}-1)} \left\{ \sqrt{6} q_{0}^{\alpha} \mathrm{T}_{q=0}^{2}(\boldsymbol{I}_{\alpha}, \boldsymbol{I}_{\alpha}) + {\sum_{q=\pm 1}}' \exp(-2\mathrm{i}q\phi) q_{2}^{\alpha} \mathrm{T}_{2q}^{2}(\boldsymbol{I}_{\alpha}, \boldsymbol{I}_{\alpha}) \right\}$$

Can be ignored

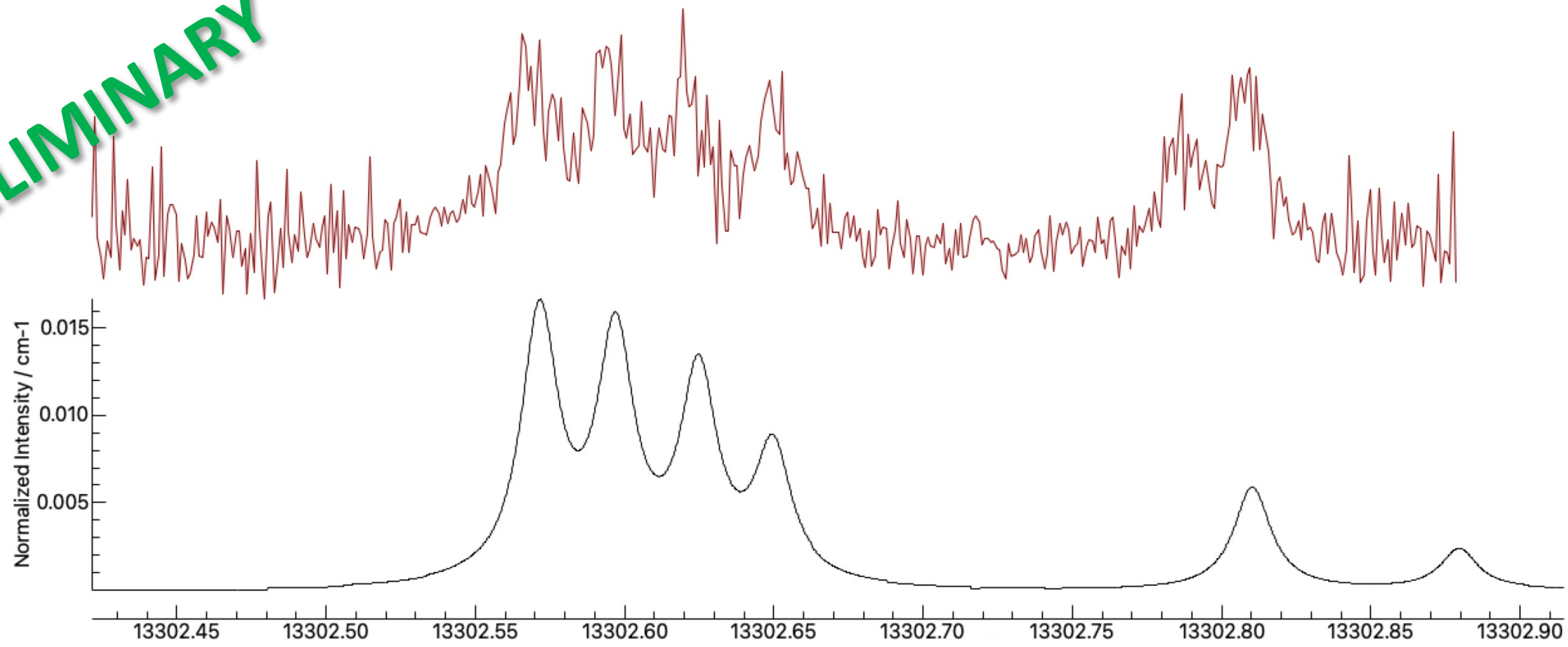
⇒ 8 parameters

2 shifts for Q-branch $1 - 2\ \mathrm{cm}^{-1}$



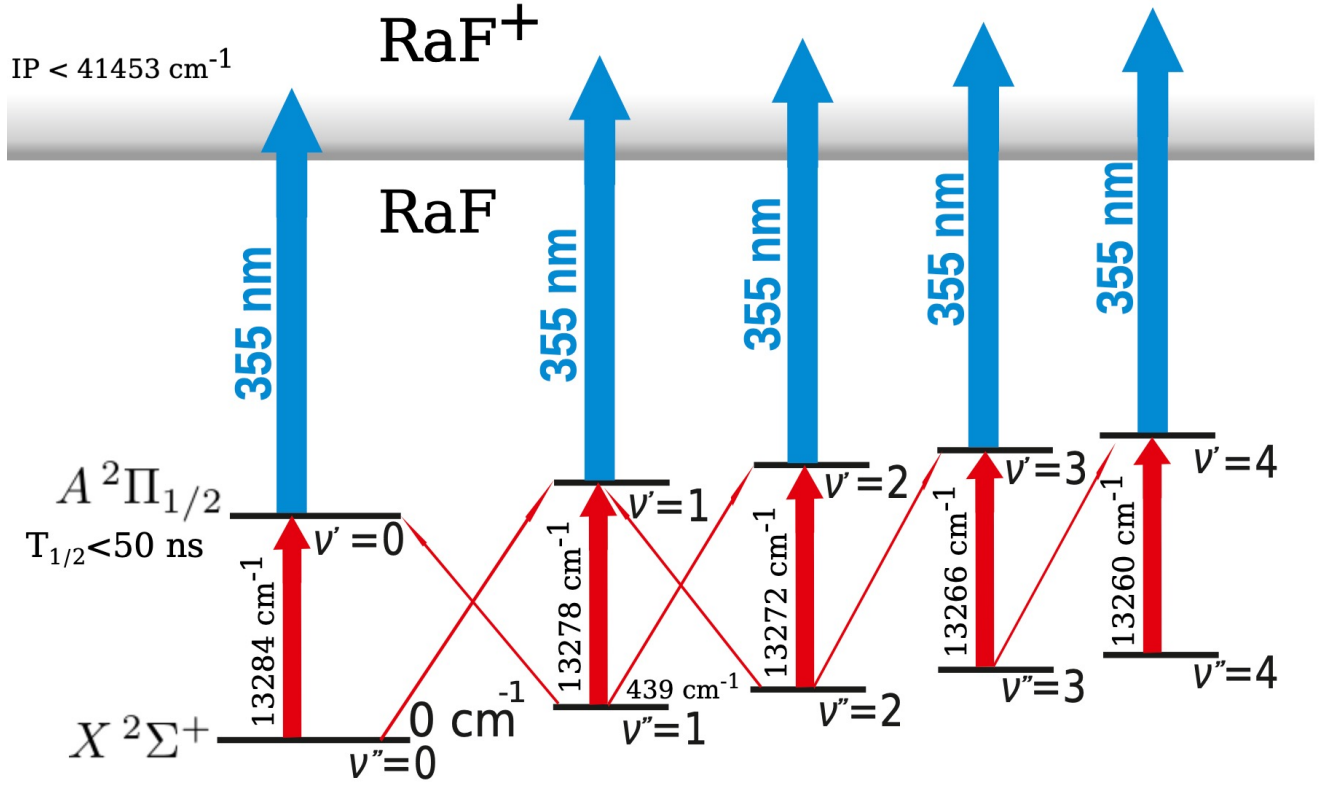
^{223}RaF

PRELIMINARY



Resonant ionization scheme

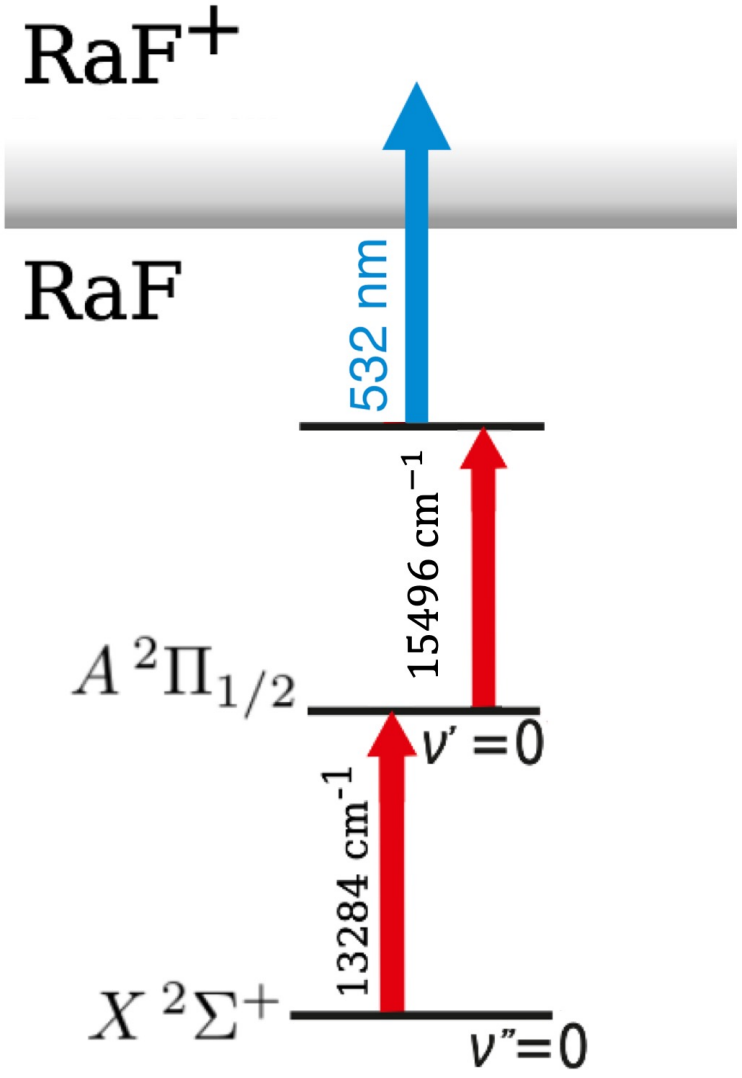
- Resonant laser:
  - Dye laser (pulsed)
  - 10 GHz (0.3 cm^{-1}) linewidth
  - 100 µJ
- Ionization laser:
  - Nd:YAG laser (3rd harmonic)
  - $\lambda = 355$ nm
  - 100 Hz repetition rate
  - 30 mJ



RF Garcia Ruiz, et al., "Spectroscopy of short-lived radioactive molecules," Nature 581, 396–400 (2020).

Resonant ionization scheme

- Resonant laser 1
  - Injection seeded
  - 50 MHz linewidth
  - 200 μJ
- Resonant laser 2
  - PDL
  - 30 GHz linewidth
  - 1 mJ
  - $\lambda = 15496\ \mathrm{cm}^{-1}$
- Ionization laser
  - Nd:YAG laser
  - $\lambda = 532\ \mathrm{nm}$
  - 30 mJ

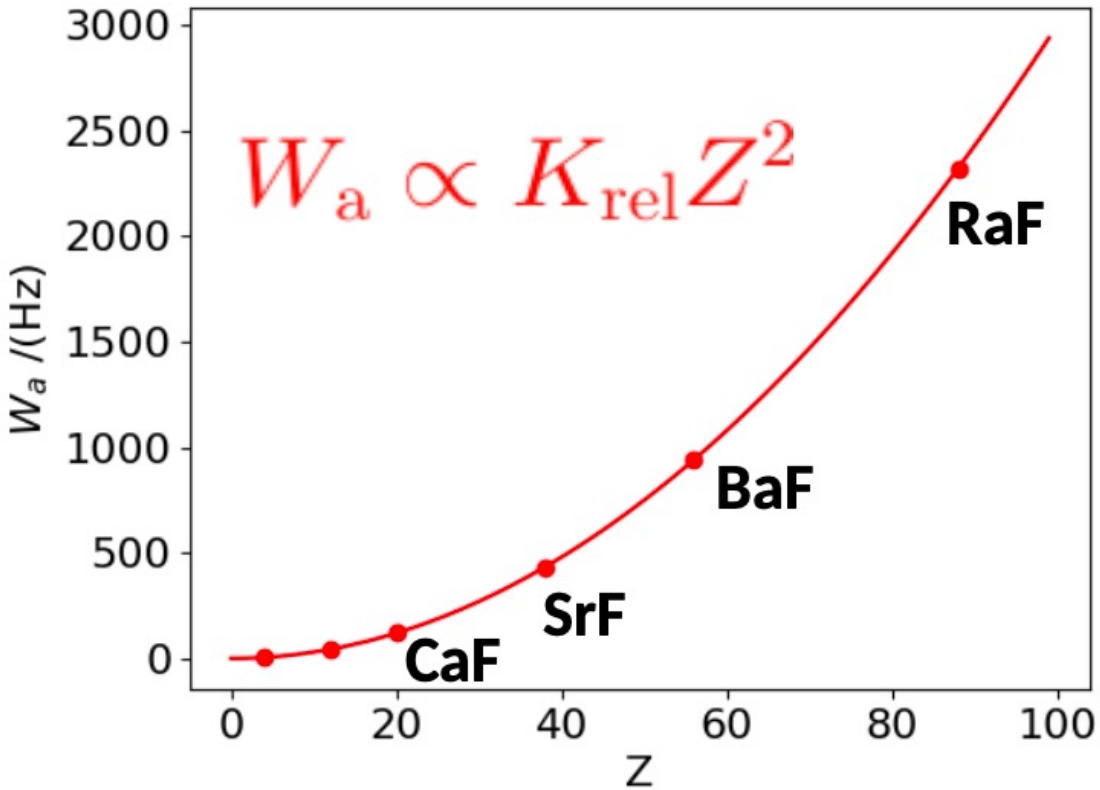


Why Radium Monofluoride (RaF)?

- Increased sensitivity to P-odd, T-even effects

$$\widehat{H}_{\rm sr} = B\vec{\mathcal{N}}^2 + \gamma\,\vec{\mathbf{S}}^{\rm eff}\cdot\vec{\mathcal{N}} + \vec{\mathbf{S}}^{\rm eff}\cdot\widehat{\mathbf{A}}\cdot\vec{\mathbf{I}} + \vec{\mathcal{N}}\cdot\widehat{\mathbf{C}}\cdot\vec{\mathbf{I}} + \cdots$$

$$+W_a(K_A/2)[\vec{\lambda}\times\vec{S}^{\rm eff}].\bar{I}$$



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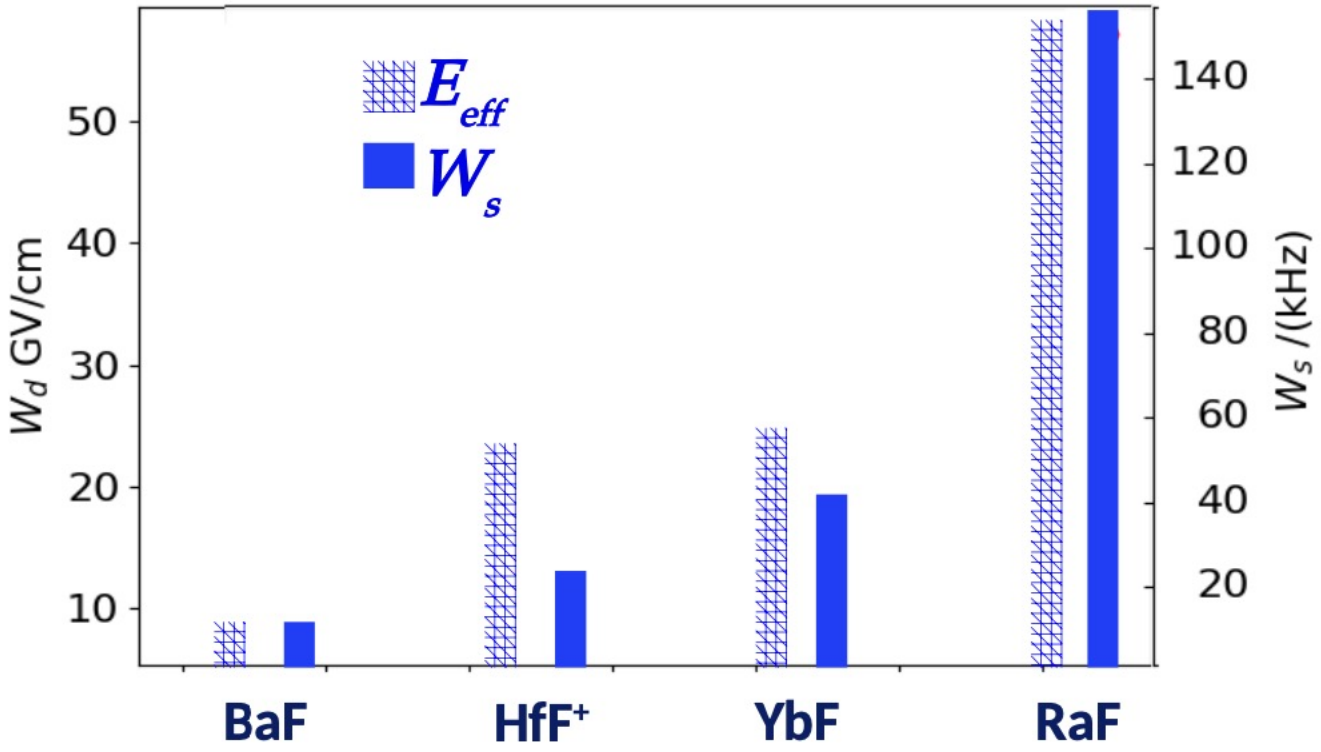
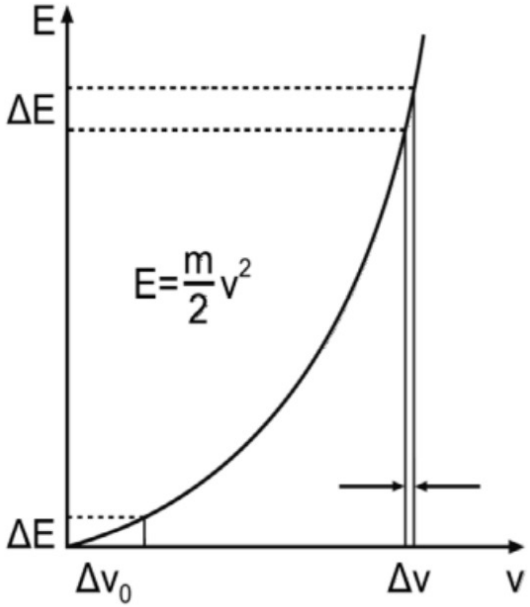
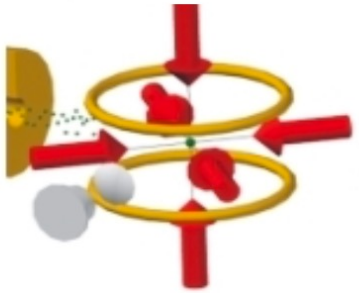
$$+W_a(K_A/2)[\vec{\lambda}\times\vec{S}^{\rm eff}].\vec{I} + (W_s k_s + E_{\rm eff} d_e)\vec{\lambda}.\vec{S}^{\rm eff}$$



Experimental Setup

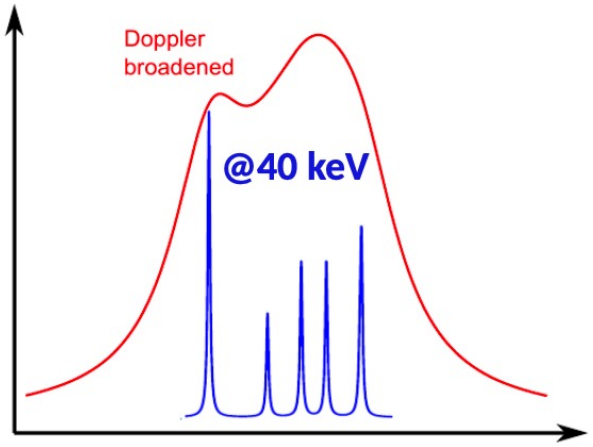
Hot molecules ⇒ Doppler broadening



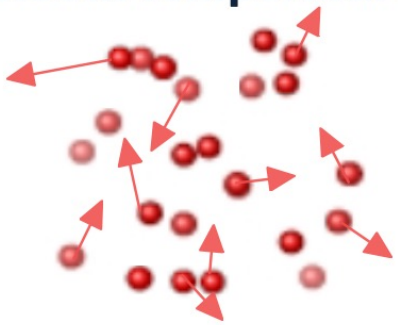


T < μK

Γ_D < MHz



Room temperature



300 K → 25 meV

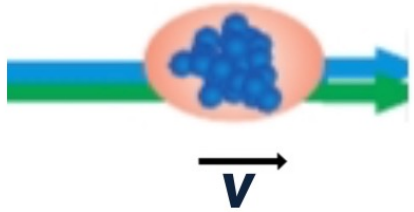
Γ_D > GHz

Energy spread

$$\Gamma_D = \nu_0 \frac{\delta E}{\sqrt{2eUmc^2}}$$

Ion beam energy

Fast beam



@ 40 keV

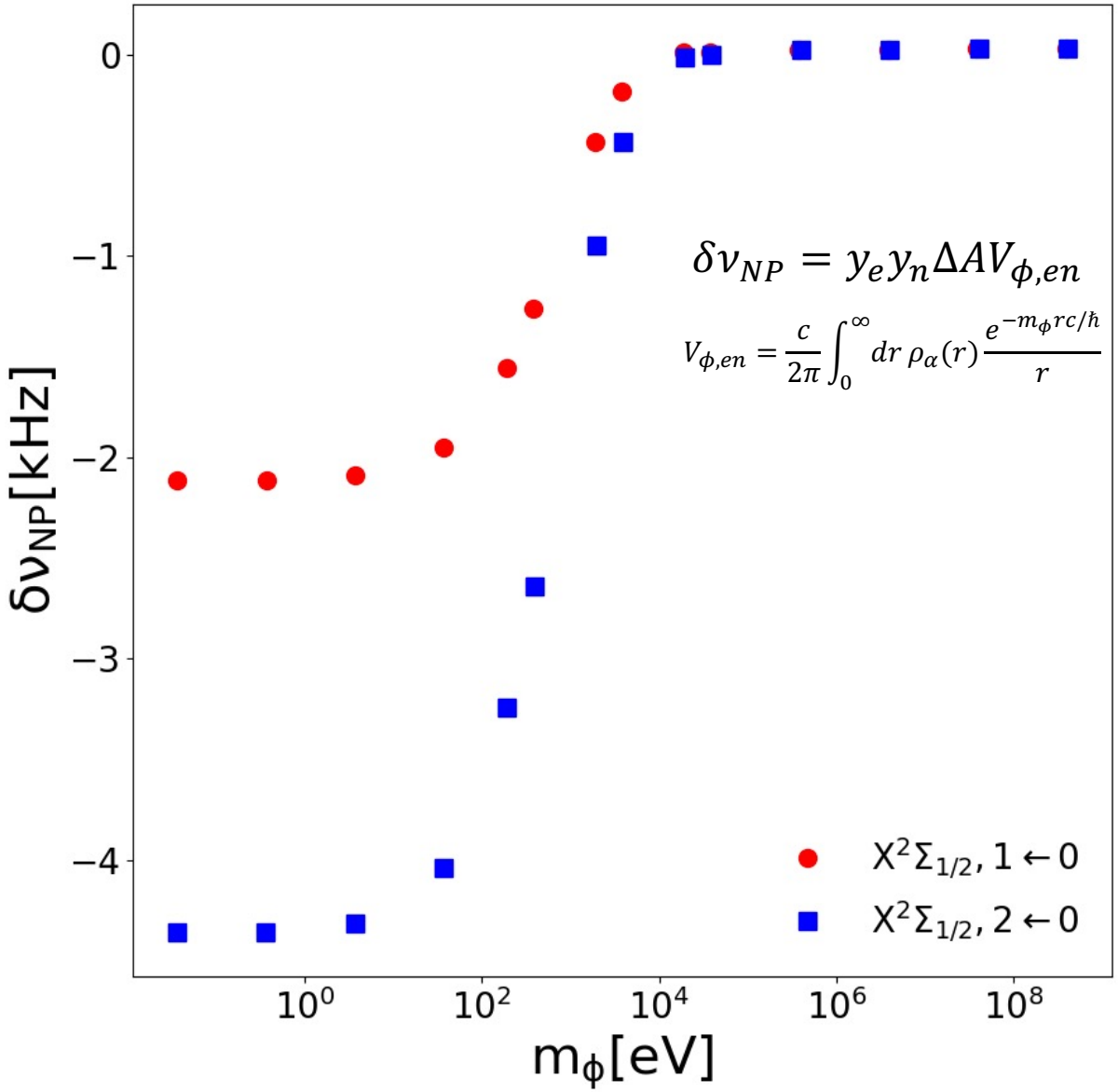
Γ_D ~ MHz

Isotope Shift of RaF - Results

$$\delta\nu_{IS}^{AA'} = K'\left(\frac{M_A - M'_A}{M_A M'_A}\right) + \left(\Delta V_{00} + \frac{\nu + 1/2}{\sqrt{\mu_A}}\Delta V_{10} + \frac{J(J+1)}{\mu}\Delta V_{01}\right)\delta\left\langle r_c^2\right\rangle^{AA'}$$

Method	$\Delta V_{00}^{A,\Pi-\Sigma}$	$\Delta V_{10}^{A,\Pi-\Sigma}$	$\Delta(\bar{\rho}_e^A)^{\Pi-\Sigma}$
exp.	-0.839(33)	-0.065(120)	392(15)
FSCCSD	-0.795(82)	-0.014(150)	371(38)
FSCCSDc	-0.823(85)	-0.014(155)	385(40)

Isotope Shift – King plot nonlinearity



K. Gaul, R. Berger