Quadrupole moment of ^{223}Ra from molecular spectroscopy of ^{223}RaF
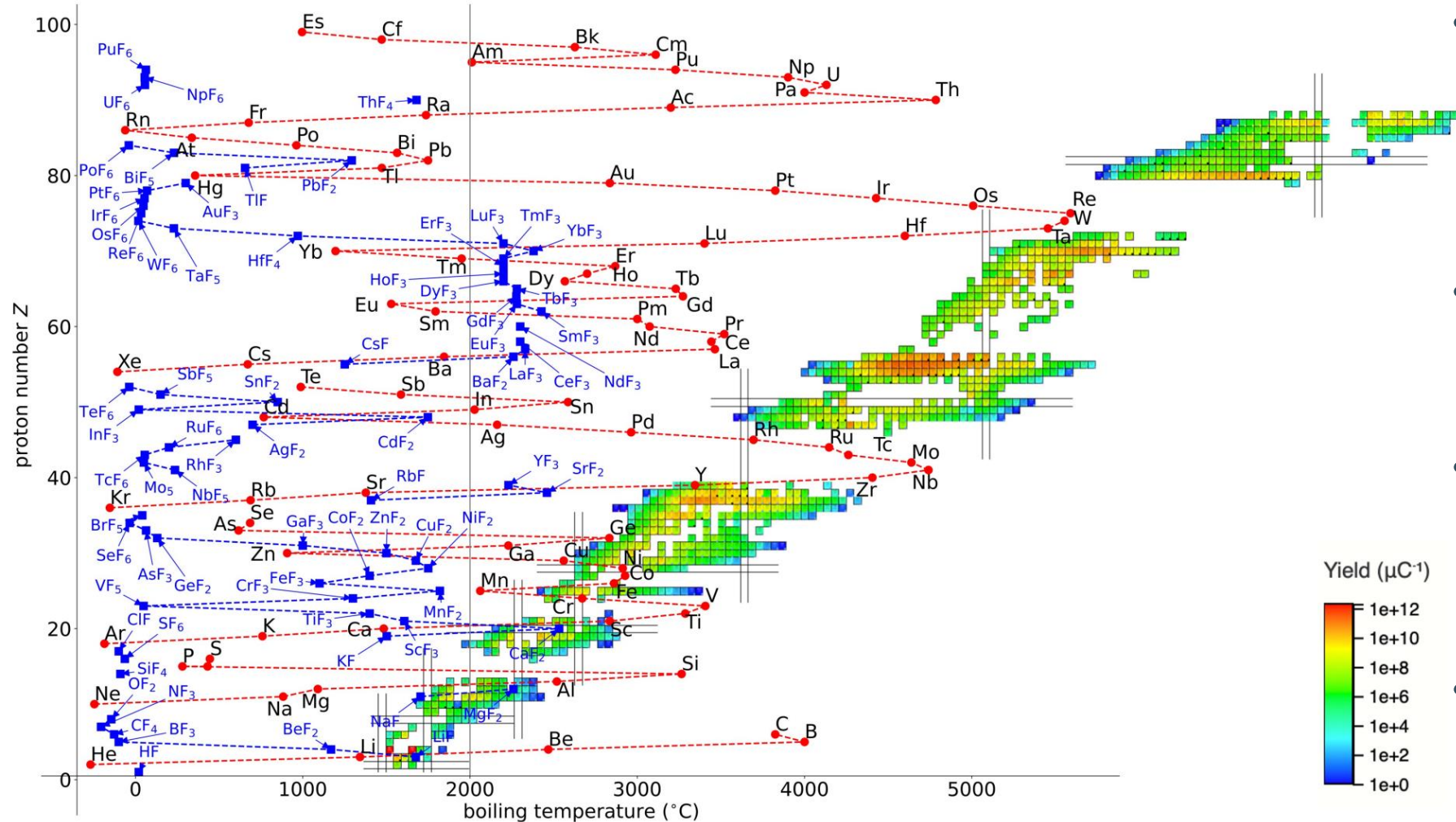
4 of September 2024

PhD student: Carlos M. Fajardo-Zambrano

Production of radioactive nuclei at ISOLDE and RaF

Produced isotopes at ISOLDE

- Molecules will have different chemical properties than its atomic counterpart ➔ different release and ionization properties.

- Molecules could be used to study poorly produced elements (Ac from AcF 2022)

- Mainly studied to provide high-purity beams in regions with high isobaric contamination.

- Molecules can be more sensitive to new observables (RaF).

[1] Au, M. (2023). Production of actinide atomic and molecular ion beams at CERN-ISOLDE (No. CERN-THESIS-2023-228).

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Why proof the existence of the electric dipole moment (EDM)?

experimentally excluded — unconstrained

Legend:
- Standard Model
- SUSY variants
- generic models

Models shown on chart: Multi-Higgs, Left-Right Symmetric, Extended Tecnicolor, Lepton Flavor Changing, Split SUSY, SO(10) GUT, Aligment, Seesaw Neutrino Yukawa Coupling, Accidental Cancellation, Approx CP, Approx Universality, Naive SUSY, Heavy sFermions, Standard Model, Imperial YbF fountain

Axis: d_e(e.cm) from 10^{-25} to 10^{-35}

Higher sensitivity in MOLECULES (and perspectives) !

- To answer unresolved problems, such as the Baryogenesis problem (matter/anti-matter asymmetry in the universe).

- **Polar molecules** containing heavy nuclei are most sensitive probes !

  - 2011: YbF $< 2.10^{-28}$ e.cm
  - 2013: ThO $< 9.10^{-29}$ e.cm
  - 2023: HfF$^+$ $< 4.10^{-30}$ e.cm

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Fluorine molecules – laser coolable

Best EDM measurements use laser cooling on **stable** fluorine molecules.

2010: SrF   Nature 467
2011: YbF   Nature 473

…

2017: SrF   Nature Physics 13
2018: YbF   PRL 120, 123201

2023: HfF$^+$ Science, 381(6653).

- **Octupole deformed nuclei** further enhance the sensitivity to EDM ! (N. Auerbach, et al., Phys. Rev. 4316 (1996)).

- Some **Ra isotopes are octupole deformed** ! (Gaffney et al., Nature 497 (2013)).

RaF: ideal molecule, but RADIOACTIVE !

- Heavy nuclei
- Octupole deform nuclei
- Polar molecule

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Laser spectroscopy and CRIS

Collinear Resonance Ionization Spectroscopy (CRIS)

Laser beams are overlapped with an accelerated ion beam, reducing doppler broadening.

Resonance ionization spectroscopy is very efficient and selective: background reduction by deflecting the resonant ions from the neutrals ➔ beam intensities of ~10-100 ions per second.

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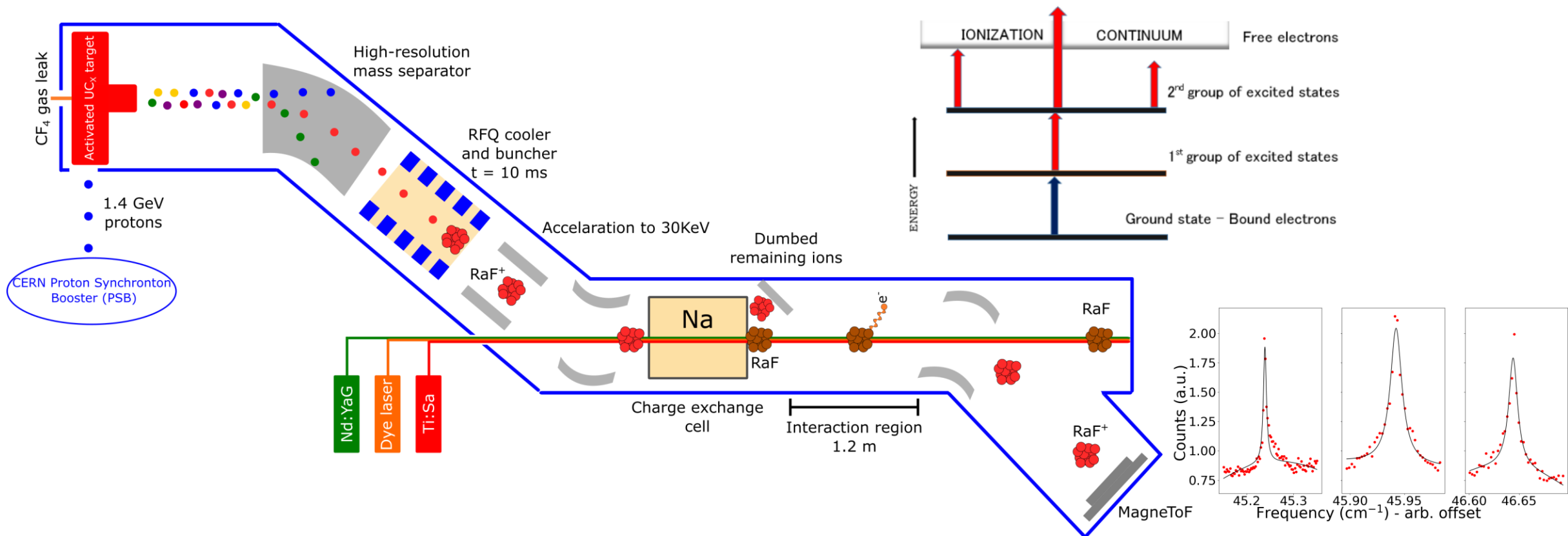
High resolution laser spectroscopy observables

$^2P_{1/2}$

$F = 0$

$F = 1$

$^2S_{1/2}$

^{89}Y (I=1/2)

$F = 0$

$F = 1$

$F = |J + I|$
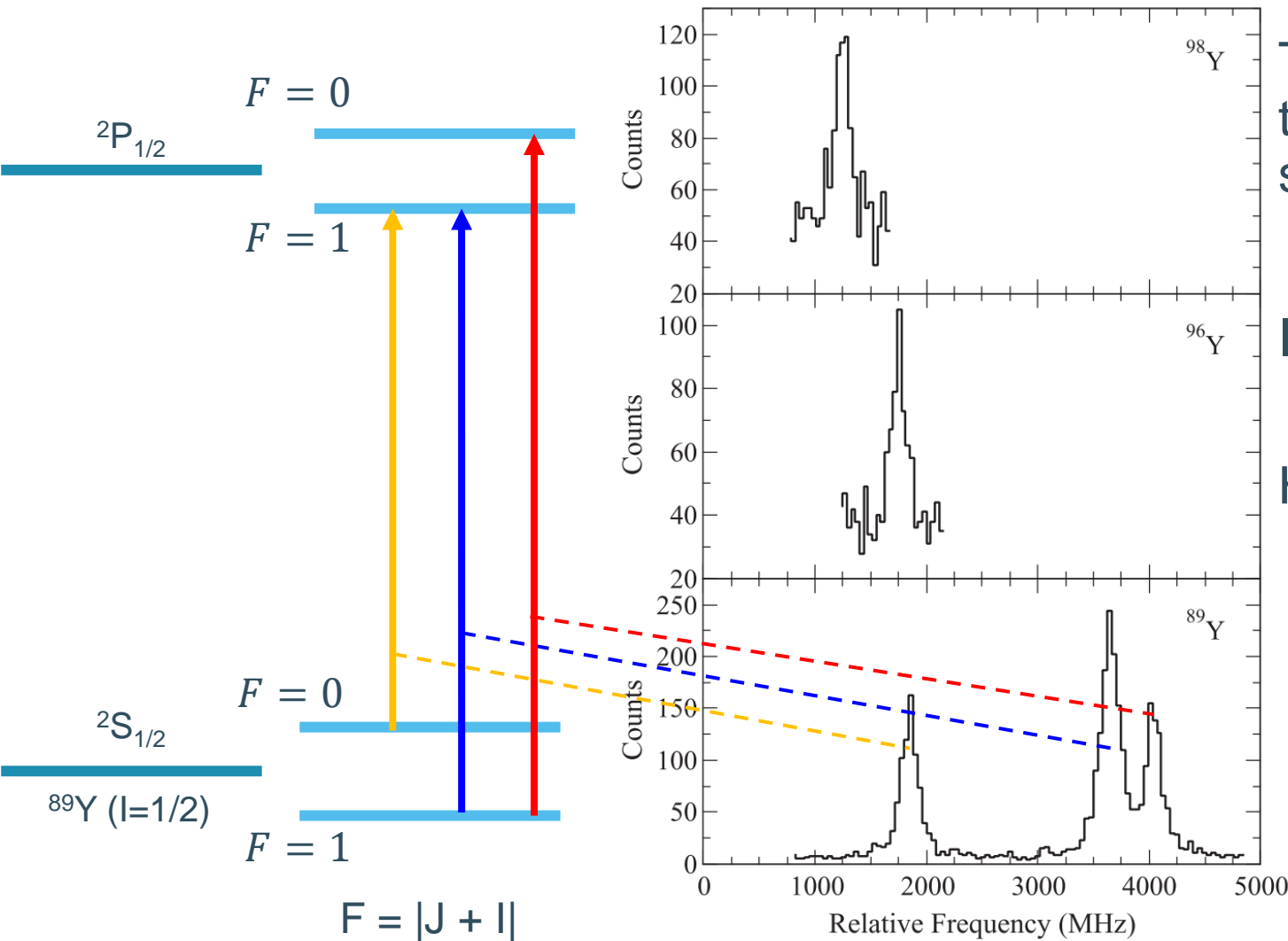
The electron-nuclear interaction leads to the appearance of the Hyperfine structure (H_{hfs}).

Isotope shifts
- Change in rms charge radii.

Hyperfine structure
- Nuclear spin.
- Magnetic dipole moment (I>0).
- Electric quadrupole moment (I>1/2)

[6] Vormawah, L. J., et al (2018). Isotope shifts from collinear laser spectroscopy of doubly charged yttrium isotopes. Physical Review A, 97(4), 042504.

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Laser spectroscopy on molecules

The spectrum of a rovibrational transition is divided in three different branches:
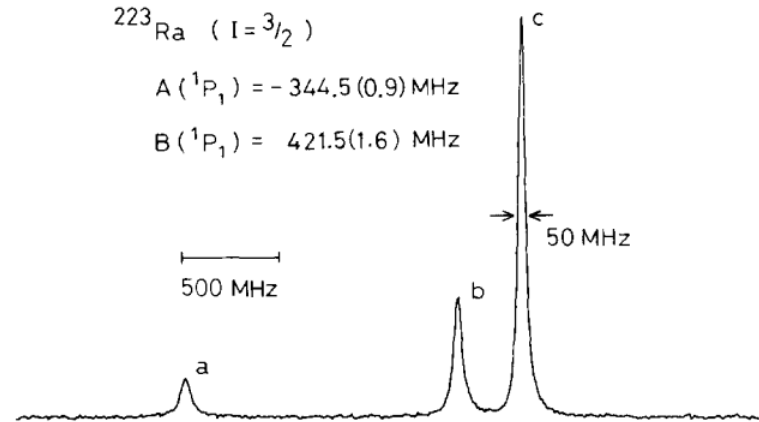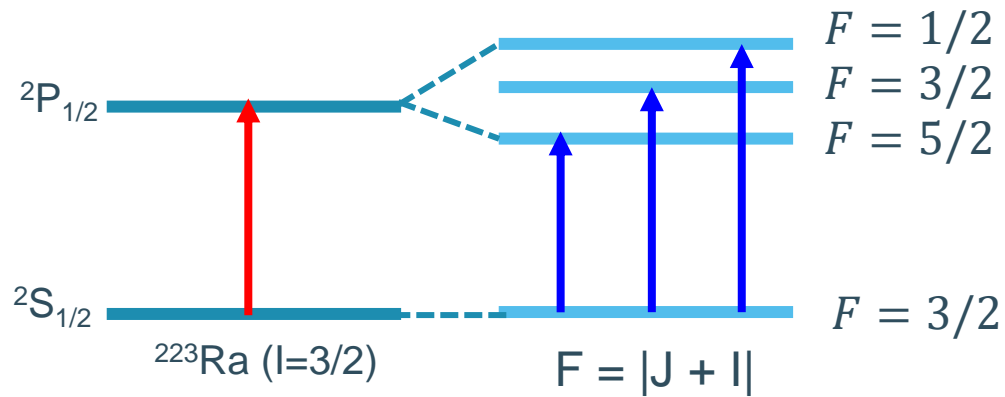R ($\Delta J = +1$), Q ($\Delta J = 0$), P ($\Delta J = -1$).
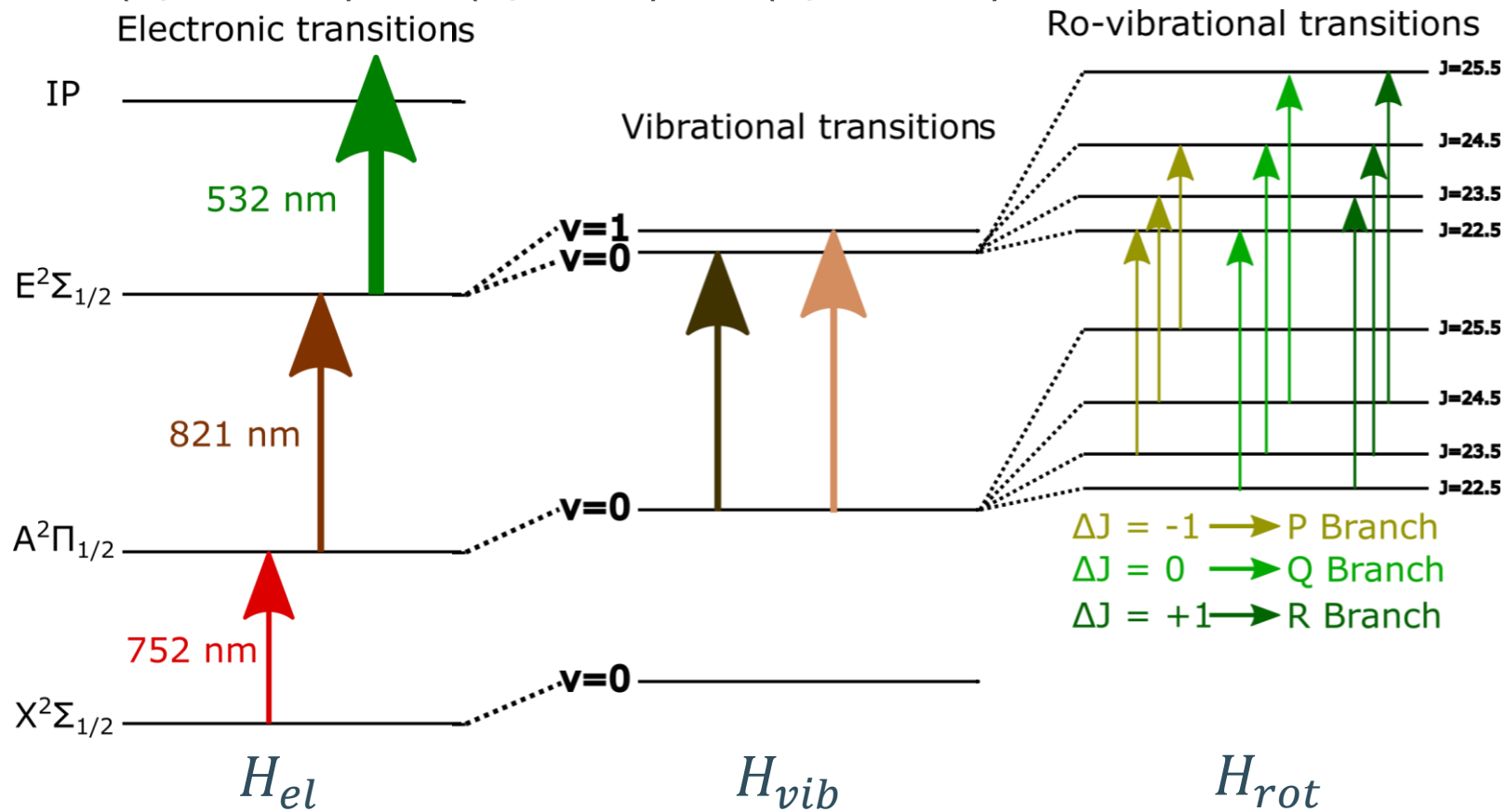
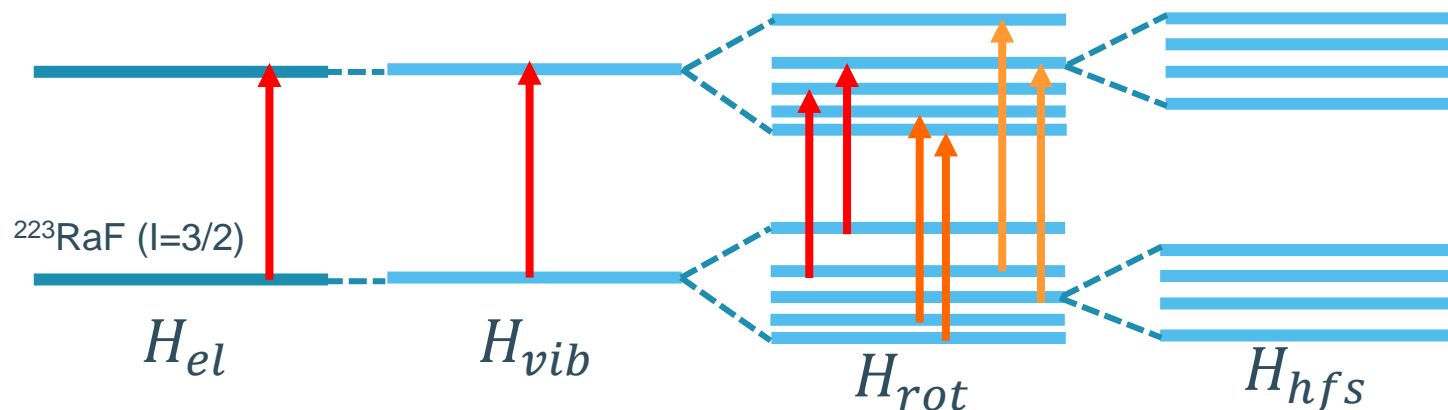Each rotational line is further split due to the electron-nuclear interaction (H_{hfs}).

ΔJ = -1 ➝ P Branch
ΔJ = 0 ➝ Q Branch
ΔJ = +1 ➝ R Branch

Typical textbook spectrum for HCl

 [7] Atkins, Physical chemistry, P. Atkins, P. W., & de Paula, J. (2014).   Institute for Nuclear and Radiation Physics, Nuclear Moments Group   KU LEUVEN
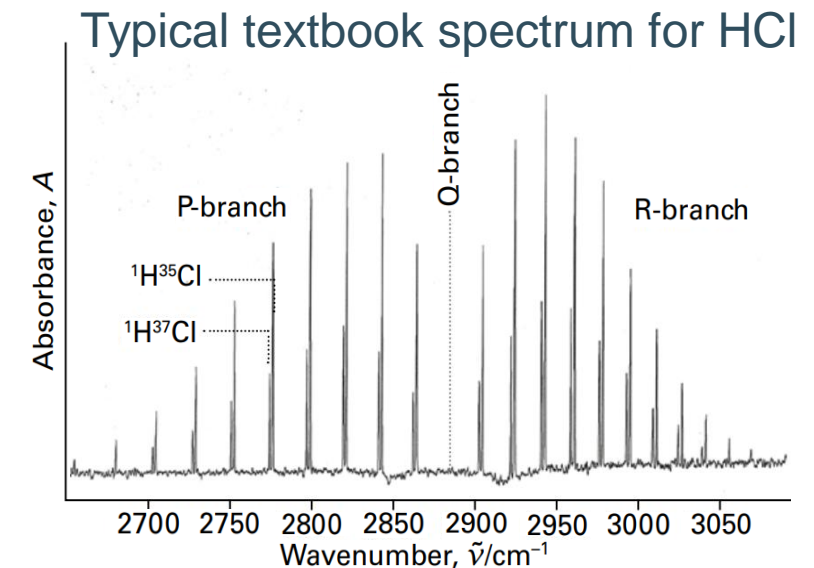
High-resolution in atoms and molecules

$$H_{hfs} = H_m + H_Q = A(I,J) + B(I,J)$$

$^2P_{1/2}$

$F = 1/2$
$F = 3/2$
$F = 5/2$

$^2S_{1/2}$

^{223}Ra (I=3/2)

$F = |J + I|$

$F = 3/2$

^{223}Ra (I = $^3/_2$)

A ($^1P_1$) = $-$ 344.5 (0.9) MHz
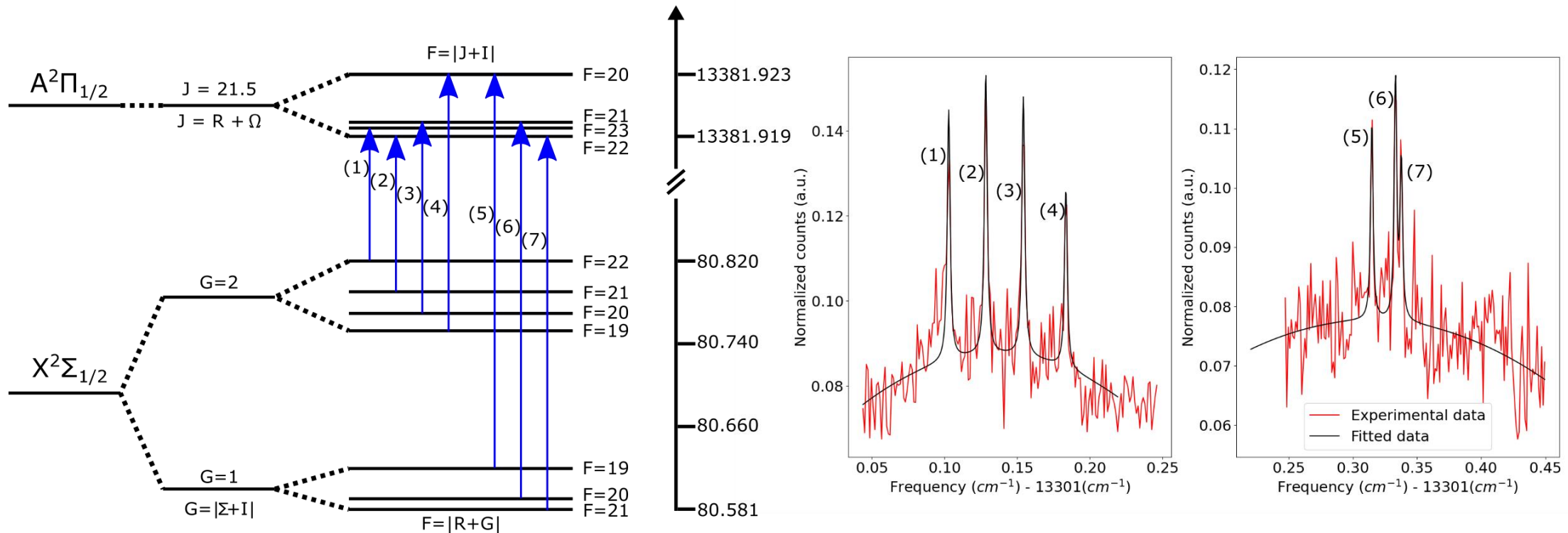
B ($^1P_1$) = 421.5(1.6) MHz

50 MHz

500 MHz

a

b

c

$$H_{hfs,R} = H_m + H_Q = A_\parallel(I_z, J_z) + A_\perp(I_{(x,y)}, J_{(x,y)}) + eq_0 Q(I_z, J_z) - eq_2 Q(I_{(x,y)}, J_{(x,y)})$$


^{223}RaF (I=3/2)

H_{el}

H_{vib}

H_{rot}

H_{hfs}

Each rotational transition is one HFS spectrum of ^{223}RaF like the one above

 [8] Ahmad, S. A., et al. Physics Letters B 133.1-2 (1983): 47-52.   Institute for Nuclear and Radiation Physics, Nuclear Moments Group   KU LEUVEN

High resolution ^{223}RaF (I =3/2) spectrum (R-branch)

- The HFS of ^{223}RaF was initially fitted using SATLAS2 to include the background on the spectra.

- The obtained centroids were used for the final fit with PGOPHER.

Fitting of the ^{223}RaF spectra (PGOPHER).
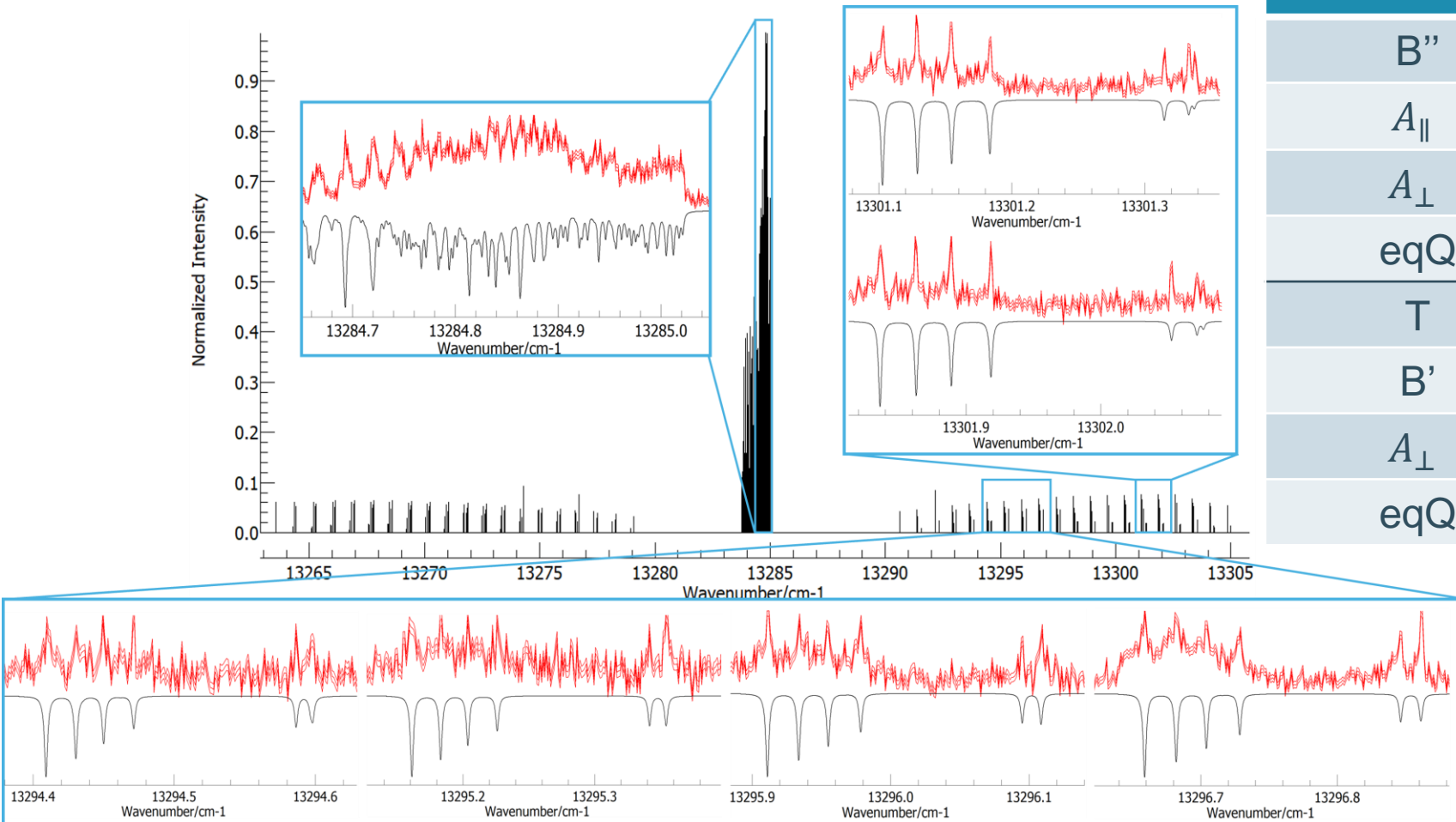
Parameter	This work	Theory
B"	5762.32(3)	5730
$A_\parallel$	2084(7)	2095
$A_\perp$	2025(2)	2016
eqQ	-1480(29)	-1473
T	13284.6959(1)	-
B'	5732.56(3)	5709
$A_\perp$	255(2)	272
eqQ	-934(23)	-912

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Extracting the nuclear moments

The ab initio magnetic dipole $(A_\parallel, A_\perp)$ and electric quadrupole coupling (eqQ) constants are defined as,

Magnetic dipole moment

Magnetic field at the nucleus

Electric quadrupole moment

EFG at the nucleus

$$A_\parallel(A) = \frac{\mu}{I} f(r_{iA}, \Sigma_z) \qquad A_\perp(A) = \frac{\mu}{I} f(r_{iA}, \Sigma_{x/y}) \qquad eqQ(A) = Q(A)0.2349647 \cdot q(A)$$

Extract the magnetic dipole and electric quadrupole moment **using the known moments of an isotope (^{225}RaF)**

Benchmark the theoretical predicted field, e.g., electric field gradient (EFG)

$$\mu_2(A) = \frac{I_2 \, A_{2(\parallel,\perp)}}{I_1 \, A_{1(\parallel,\perp)}} \mu_1(A)$$

$$q(A) = \frac{eqQ(223RaF)}{Q(^{223}Ra)0.2349647}$$

$\mu(^{223}\text{RaF}) = 0.27095(85) \, \mu_n$

$\mu(^{223}\text{Ra}) = 0.2703(6) \, \mu_n$

$q(^{223}\text{RaF})_{exp} = 5.02(2)$ a.u.

$q(^{223}\text{RaF})_{theo} = 5.02(2)$ a.u.

Only statistical error bars

[9] Wilkins, S., et al. (2023). arXiv preprint arXiv:2311.04121.
[10] Lynch, K. M., et al. (2018). Physical Review C, 97(2), 024309.
[11] Petrov, A. N., & Skripnikov, L. V. (2020). Physical Review A, 102(6), 062801.

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Reference moments from molecules

The measurement of nuclear moments using molecules is not new.

Elements highlighted have their current reference moments from molecular measurements.

[12] Stone, N. (2016). Atomic Data and Nuclear Data Tables, 111:1–28.
[13] Stone, N. (2019). Technical report, International Atomic Energy Agency.

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Discussion

- Given its different chemical properties, molecules can be used for the study of isotopes that are poorly produce or are in a region with high isobaric contamination.

- Molecules are the most sensitive system to the presence of an eEDM.

- The magnetic dipole and electric quadrupole moments of ^{223}RaF has been successfully extracted and are in great agreement with their atomic counterpart (work in progress).

- Molecular spectroscopy is a promising tool to study unknown nuclear moments for nuclear and beyond the standard model physics.

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- Au, M. (2023). Production of actinide atomic and molecular ion beams at CERN-ISOLDE. PhD thesis, Johannes Gutenberg-Universitat, Germany.

- Vormawah, L. J., Vilén, M., Beerwerth, R., Campbell, P., Cheal, B., Dicker, A., ... & Voss, A. (2018). Isotope shifts from collinear laser spectroscopy of doubly charged yttrium isotopes. Physical Review A, 97(4), 042504.

- Atkins, P. W., De Paula, J., & Keeler, J. (2023). Atkins' physical chemistry. Oxford university press.

- Wilkins, S. G., Udrescu, S. M., Athanasakis-Kaklamanakis, M., Ruiz, R. F., Au, M., Belošević, I., ... & Zülch, C. (2023). Observation of the distribution of nuclear magnetization in a molecule. arXiv preprint arXiv:2311.04121.

- Lynch, K. M., Wilkins, S. G., Billowes, J., Binnersley, C. L., Bissell, M. L., Chrysalidis, K., ... & Yang, X. F. (2018). Laser-spectroscopy studies of the nuclear structure of neutron-rich radium. Physical Review C, 97(2), 024309.

- Petrov, A. N., & Skripnikov, L. V. (2020). Energy levels of radium monofluoride RaF in external electric and magnetic fields to search for P-and T, P-violation effects. Physical Review A, 102(6), 062801.

- Stone, N. J. (2016). Table of nuclear electric quadrupole moments. Atomic Data and Nuclear Data Tables, 111, 1-28.

- Stone, N. J. (2019). Table of recommended nuclear magnetic dipole moments (No. INDC (NDS)--0794). International Atomic Energy Agency.

- Köster, U., Arndt, O., Bouquerel, E., Fedoseyev, V. N., Frånberg, H., Joinet, A., ... & Targisol Collaboration. (2008). Progress in ISOL target–ion source systems. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 266(19-20), 4229-4239.

- Steimle, T. C., Ma, T., & Linton, C. (2007). The hyperfine interaction in the $A\Pi 1/2 2$ and $X\Sigma + 2$ states of ytterbium monofluoride. The Journal of chemical physics, 127(23).

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Thanks for your attention

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Backup slides

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Collinear geometry

- Ion beam is accelerated to 30-60 keV, reducing the velocity spread.

- Use of a gas-filled radiofrequency cooler-buncher to produce short (μs) bunches.

- Overlapped (anti-)collinearly with two or more lasers.
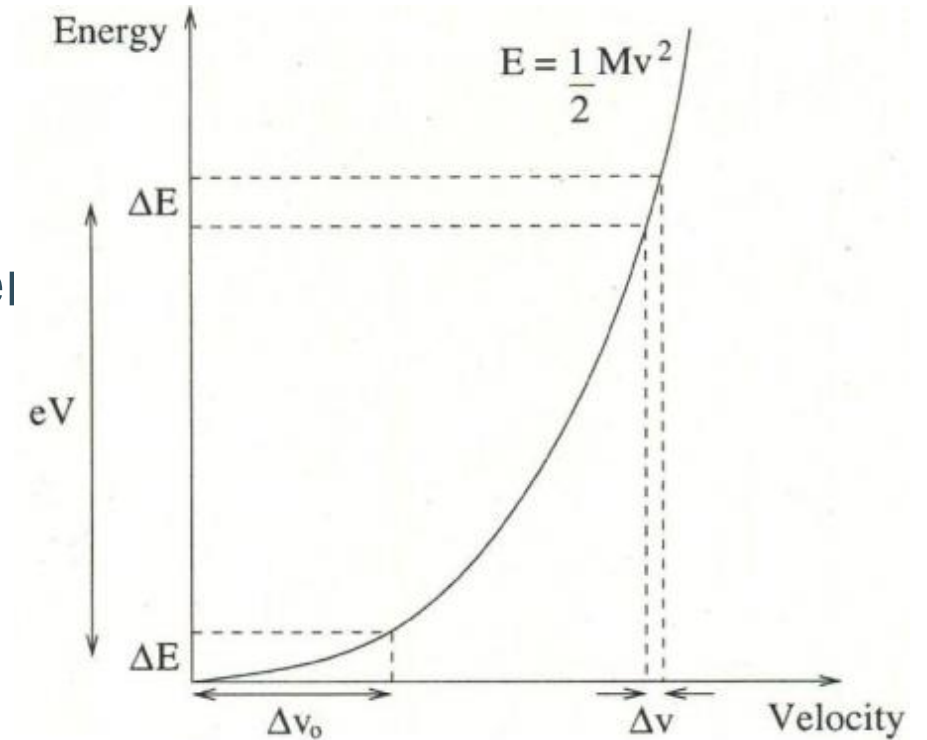
$$\nu - \nu_0 = \nu_0 \frac{\Delta E}{c \sqrt{2eV_{acc}m}}$$

Reduction in spectrum broadening from ~GHz to ~MHz!!

 [5] Campbell, P. et al. Progress in Particle and Nuclear Physics, 86:127–180.   Institute for Nuclear and Radiation Physics, Nuclear Moments Group

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Background present on the spectra

The line strength of a molecular dipole transition between HFS states is proportional to,

$$\langle N'SJ'IF' | \mu^{(1)} | NSJIF \rangle$$

$$\propto [(2J+1)(2J'+1)(2F+1)(2F'+1)]^{1/2} \begin{Bmatrix} N' & J' & S \\ J & N & 1 \end{Bmatrix} \begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix}$$

Where the terms in {} is the Wigner 6j symbols. For a transition between…(R-branch) the intensity of a $\Delta F = +1$ is ~18 and ~666 times stronger than a $\Delta F = 0$ and a $\Delta F = -1$ transition, respectively.

If the transition energies the $\Delta F = 0$ are calculated (based on the final fit), the position of the peaks are mainly within the quadruplet observed.

[2] Campbell, W. C. (2008), Doctoral dissertation, Harvard University.
[3] Breier, A. A. (2019). Doctoral dissertation, kassel university press GmbH.

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Error bars on the molecular constants of ^{223}RaF

- Each HFS peak was used to create a Gaussian distribution with centroid and width equal to the peak centroid and uncertainty.

- Re-fitting of the spectra with new peak centroids taken from the Gaussians.

- Store the fitted molecular constants and re-fit the spectra.

- The retrieved distribution from each constant gives its nominal value and uncertainty.

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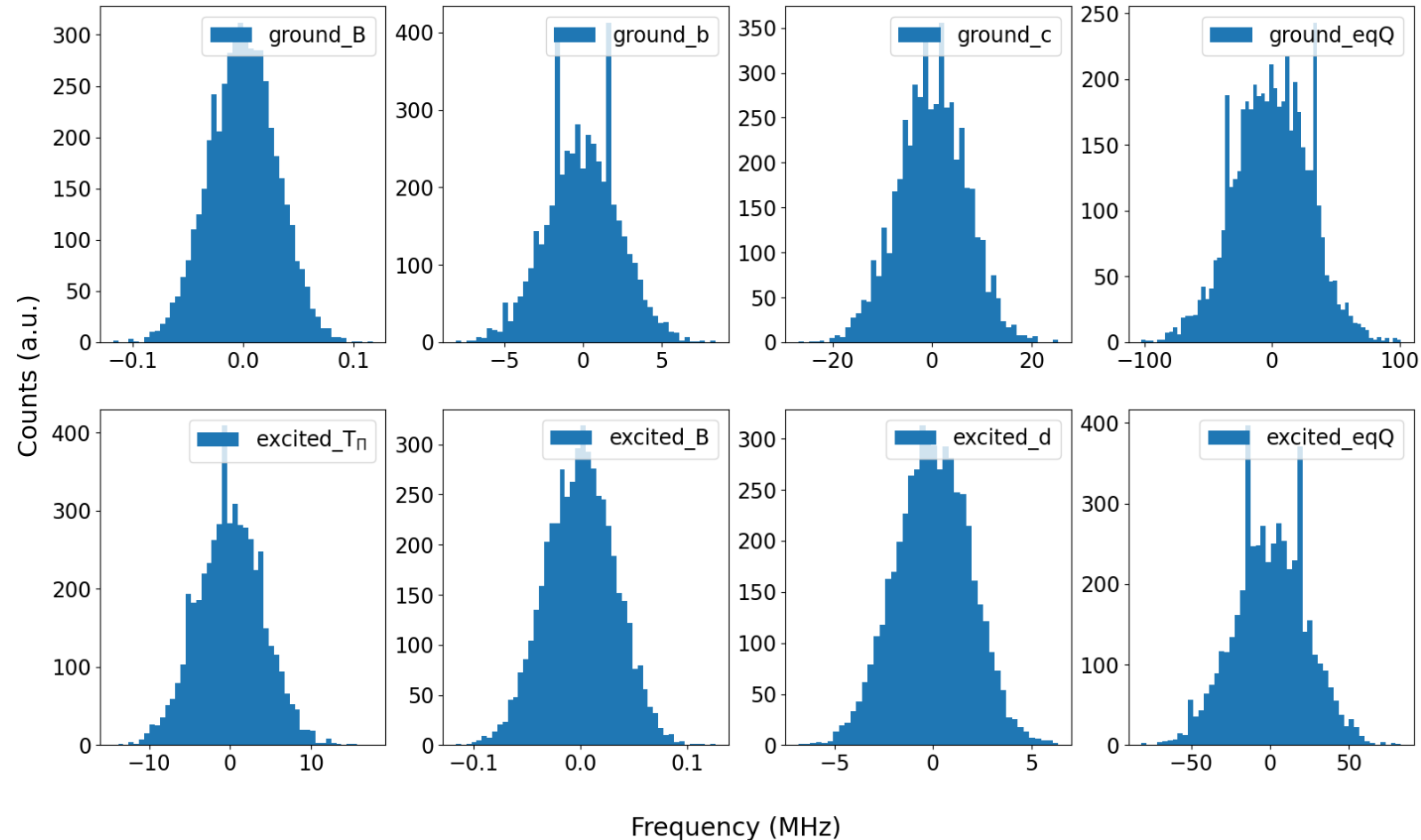
Electric dipole moment (EDM)

The EDM changes sign with parity and time reversal transformations.
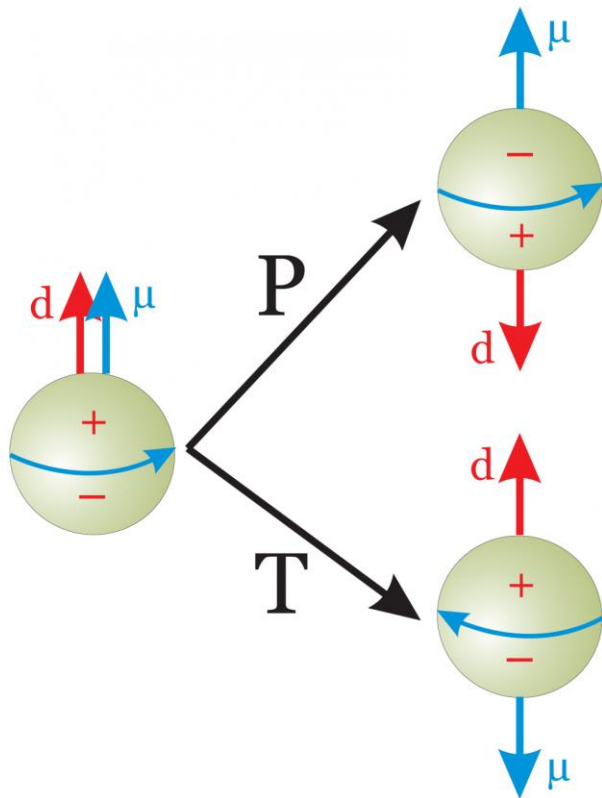
EDM is odd under parity (P) and time-reversal (T) transformation.

A permanent EDM implies violation P,T

EDM's are theoretically predicted to be extremely small,
therefore, not yet observed

[2] "Parity-and time-reversal tests in nuclear physics." 2013. 155-170.

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Intensity of a rotational-vibrational transition

This intensity increases with J, passing through a maximum and tailing off as J becomes even bigger.

- Population distribution given by the Boltzmann distribution.

- Degeneracy of each rotational level ($g_J = 2J + 1$).

A bigger population distribution is allowed with J. But the population of a state decreases exponentially as its energy increases.

$$N_J = g_J e^{-E_J/kT}$$

 [1] Atkins, Physical chemistry, P. Atkins, P. W., & de Paula, J. (2014). Institute for Nuclear and Radiation Physics, Nuclear Moments Group

KU LEUVEN
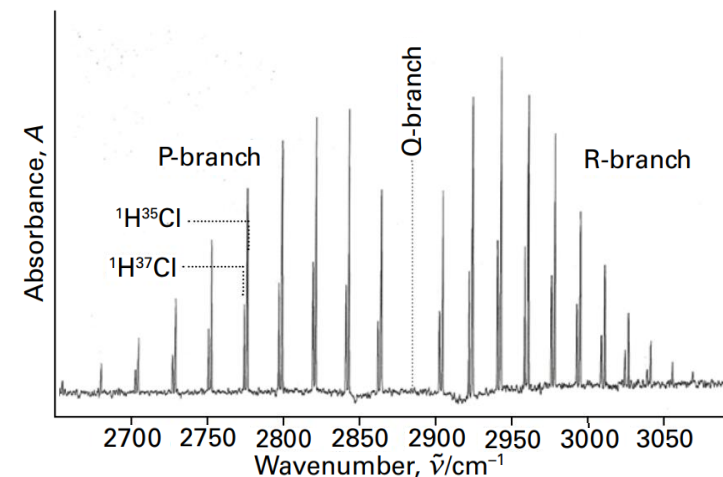
HFS precision limits with laser spectroscopy

Resolution:

- Laser bandwidth
- Lifetime (atomic and nuclear)
- Ion beam properties

Sensitivity to nuclear properties

Instrumentation for measurements

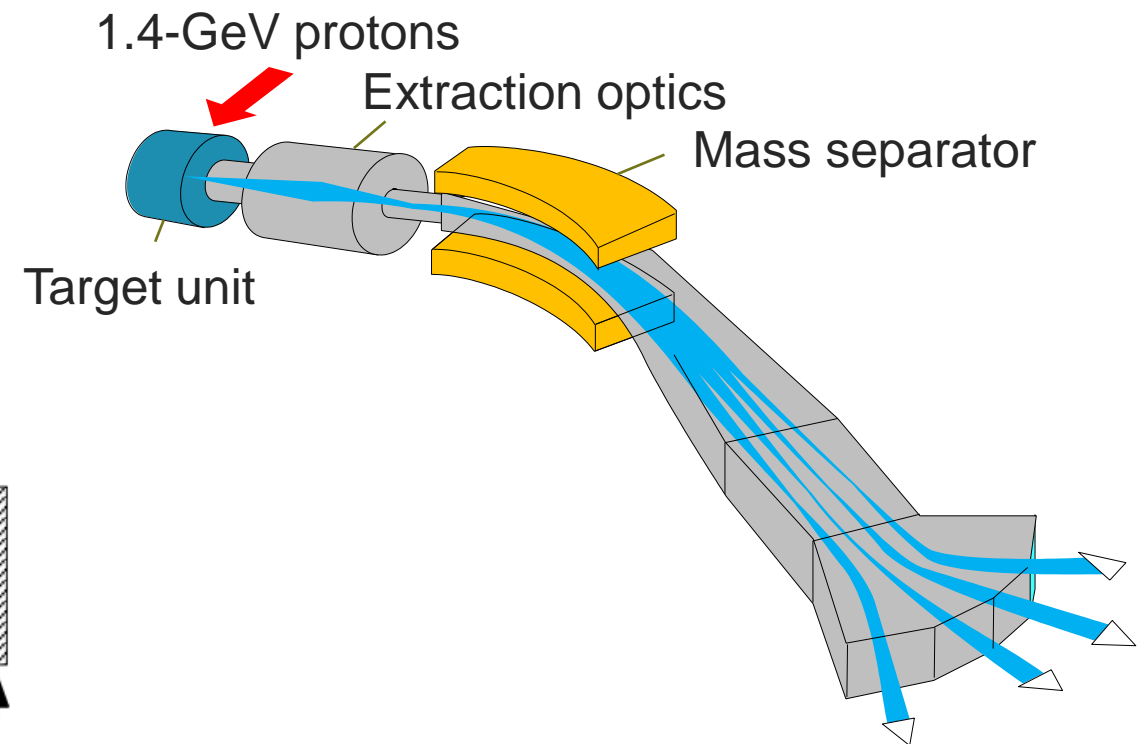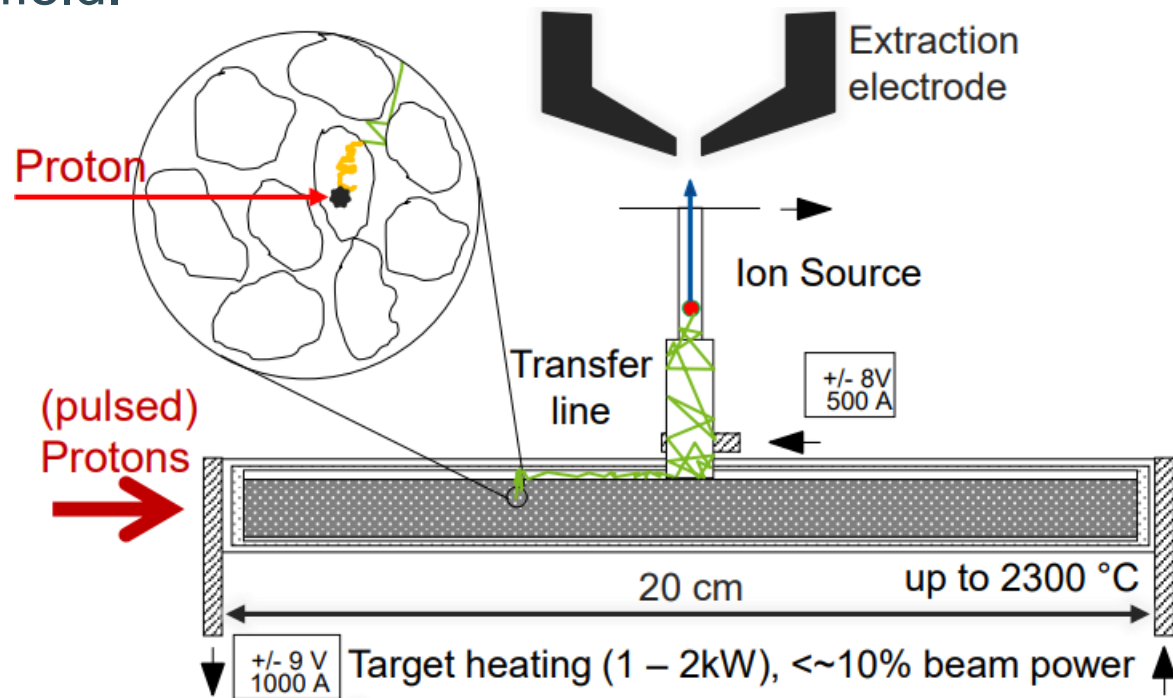- Wavelength meter
- Voltage/laser scanning

Statistics

[1] Taken from Ági Koszorús, LISA conference 2023.   Institute for Nuclear and Radiation Physics, Nuclear Moments Group   **KU LEUVEN**

EDM and why RaF has been studied?

The measurement of a permanent electric dipole moment (EDM) implies P,T violation ➜ to answer the baryogenesis problem and benchmark beyond the standard model theories.
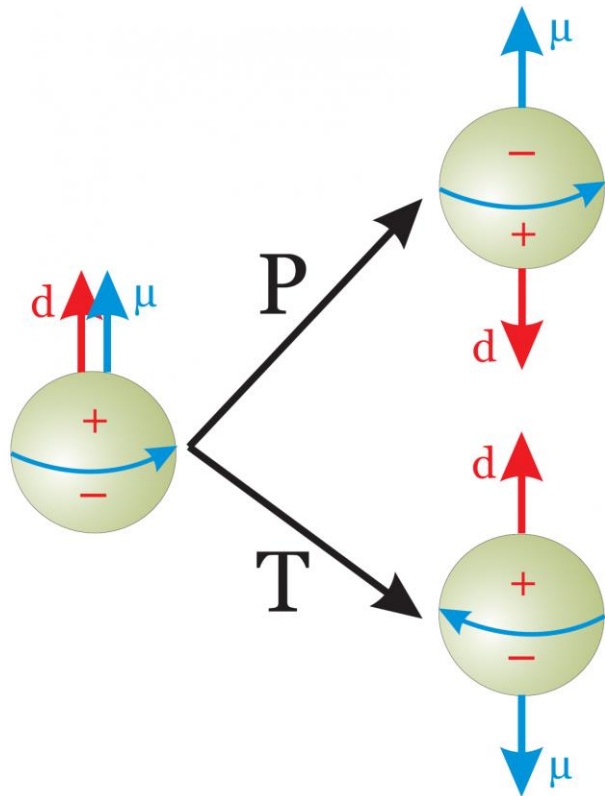
- Diatomic polar molecules have higher sensitivity to the EDM than atoms (Internal effective electric field). Current best measuring of eEDM is **three order** of magnitudes more sensitive than atoms.

- Heavy octupole deformed nuclei are more sensitive to a nuclear EDM, but they are all radioactive.

- RaF as the ideal case: diatomic polar molecule, with a laser cooling scheme and a heavy octupole deformed nuclei.

[5] "Parity-and time-reversal tests in nuclear physics." 2013. 155-170.

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The isotope separation on-line (ISOL) method

Use of thick targets allows a large total cross-section. Main reactions: spallation, fragmentation and fission. The produced atoms/molecules are ionized in an ion source. Ions of a specific mass (atomic or molecular) are separated from others using a magnetic field.
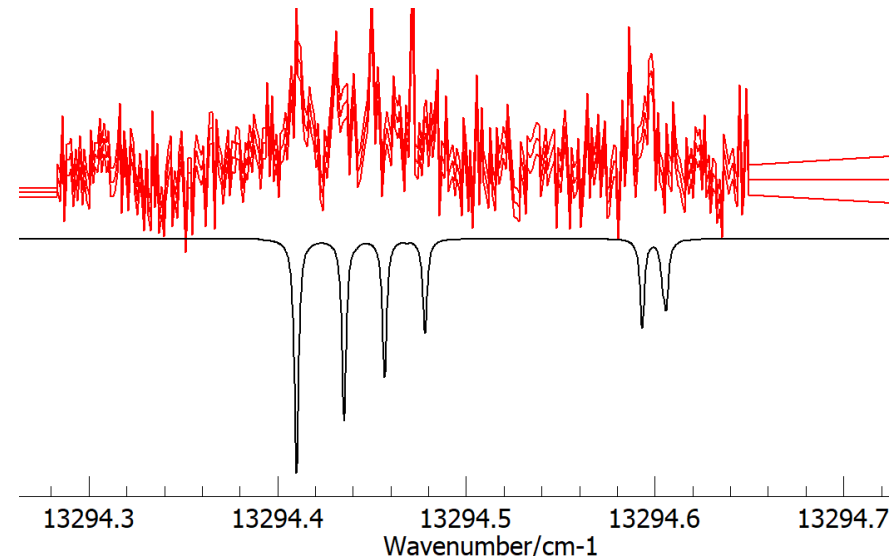
[1] J. P. Ramos | 17/09/2018 EMIS2018
[2] Catherall, R. et al. (2017) J. Phys. G. 44, 9

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Molecular HFS Hamiltonian

$$H_{hfs} = H_m + H_Q = a(I,L) + b_F(I,S) + c(I,S) - d(S_+I_+, S_-I_-) + eq_0Q(I,S) - eq_2Q(S_+I_+, S_-I_-)$$

- a, is the nuclear spin-electron orbit interaction, b_F is the Fermi contact interaction, c and d are the nuclear spin-electron spin axial and perpendicular dipole interaction

- eq_0Q and eq_2Q are the nuclear quadrupole axial and perpendicular coupling constants.

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SPFIT/SPCAT

- The fit does not include all constants. Error on the assignment of transitions.

- All the constants are "close" but not fully reproduce the spectra.

- The "quantum numbers" are arbitrary selected. They might be miss assigned.

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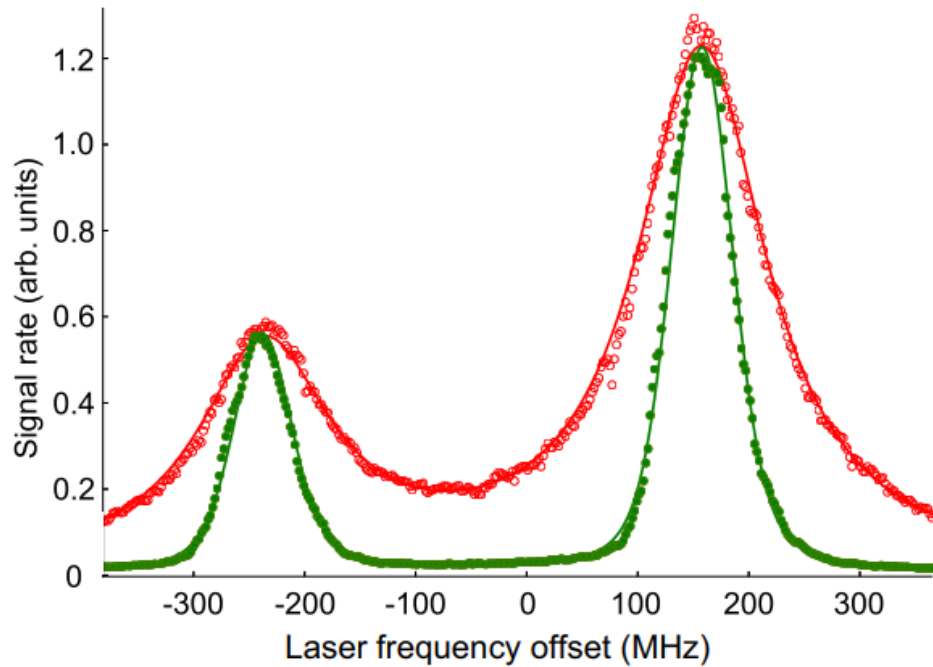KU LEUVEN

Background present on the spectra

The line strength of a molecular dipole transition between HFS states is proportional to,

$$\langle N'SJ'IF' | \mu^{(1)} | NSJIF \rangle$$

$$\propto [(2\,J+1)\,(2\,J'+1)\,(2\,F+1)\,(2\,F'+1)]^{1/2} \begin{Bmatrix} N' & J' & S \\ J & N & 1 \end{Bmatrix} \begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix}$$

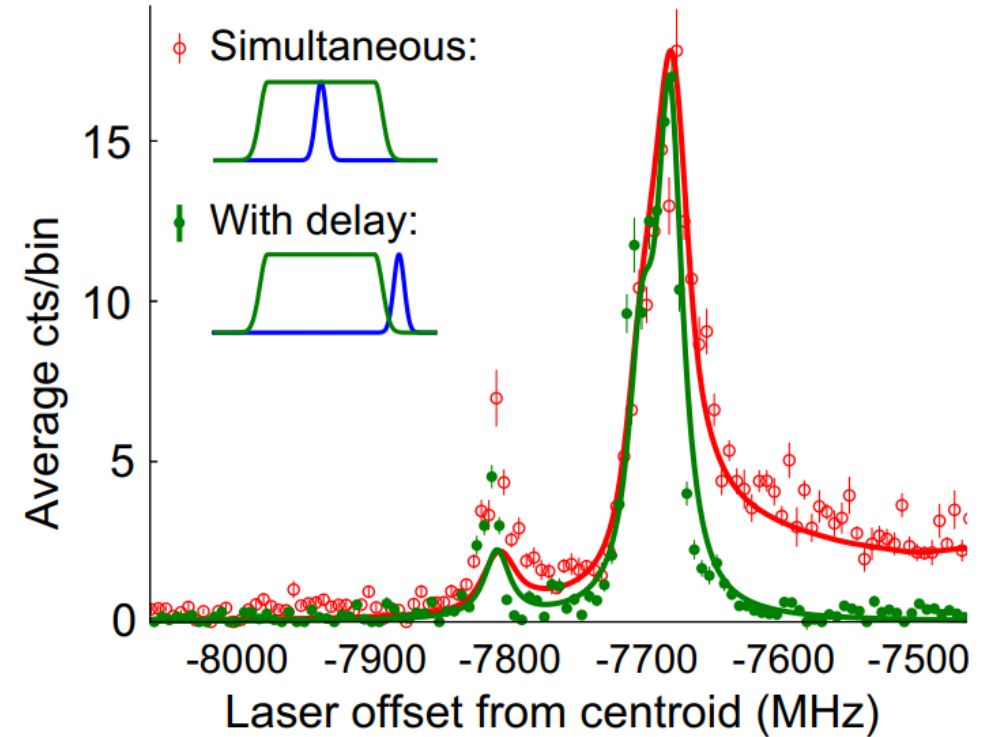Where the terms in {} is the Wigner 6j symbols. For a transition between…(R-branch) the intensity of a $\Delta F = +1$ is ~18 and ~666 times stronger than a $\Delta F = 0$ and a $\Delta F = -1$ transition, respectively.

If the transition energies the $\Delta F = 0$ are calculated (based on the final fit), the position of the peaks are mainly within the quadruplet observed.
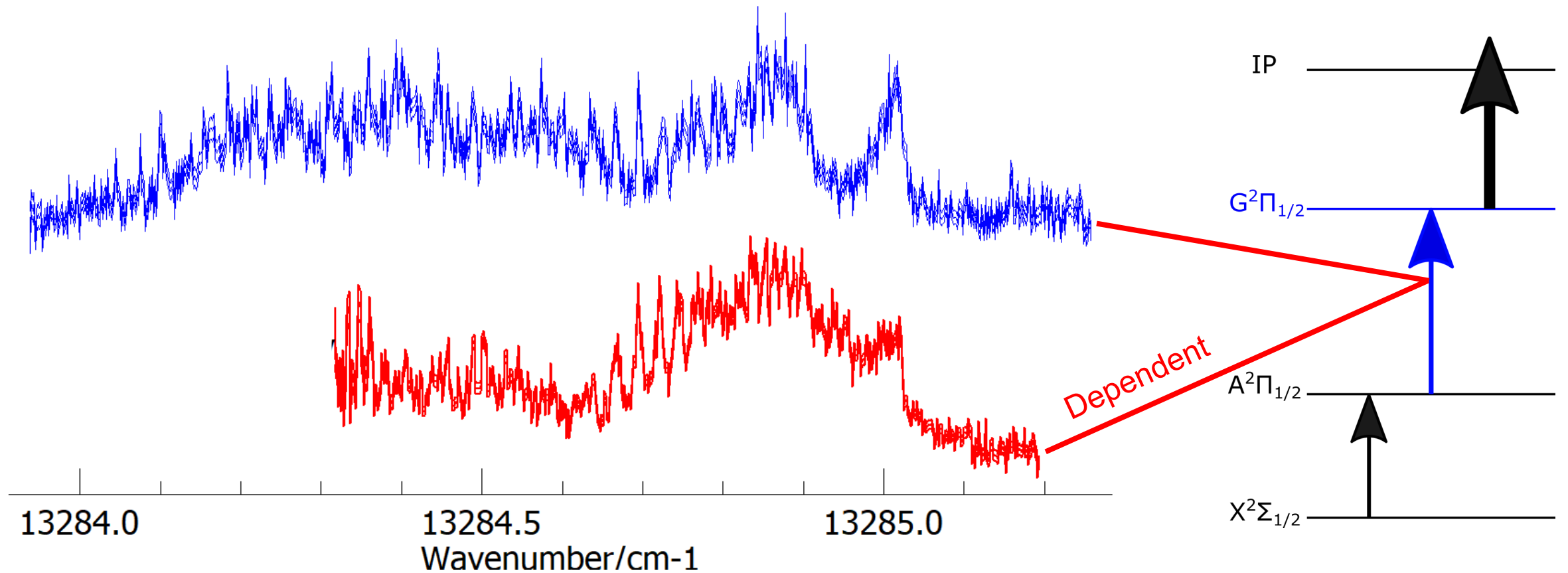
[2] Campbell, W. C. (2008), Doctoral dissertation, Harvard University.
[3] Breier, A. A. (2019). Doctoral dissertation, kassel university press GmbH.

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AC stark effect and power broadening

Power broadening (red)

AC stark effect (red)

 [1] De Groote, R. P., et al. (2017). Physical Review A, 95(3), 032502.   Institute for Nuclear and Radiation Physics, Nuclear Moments Group   KU LEUVEN
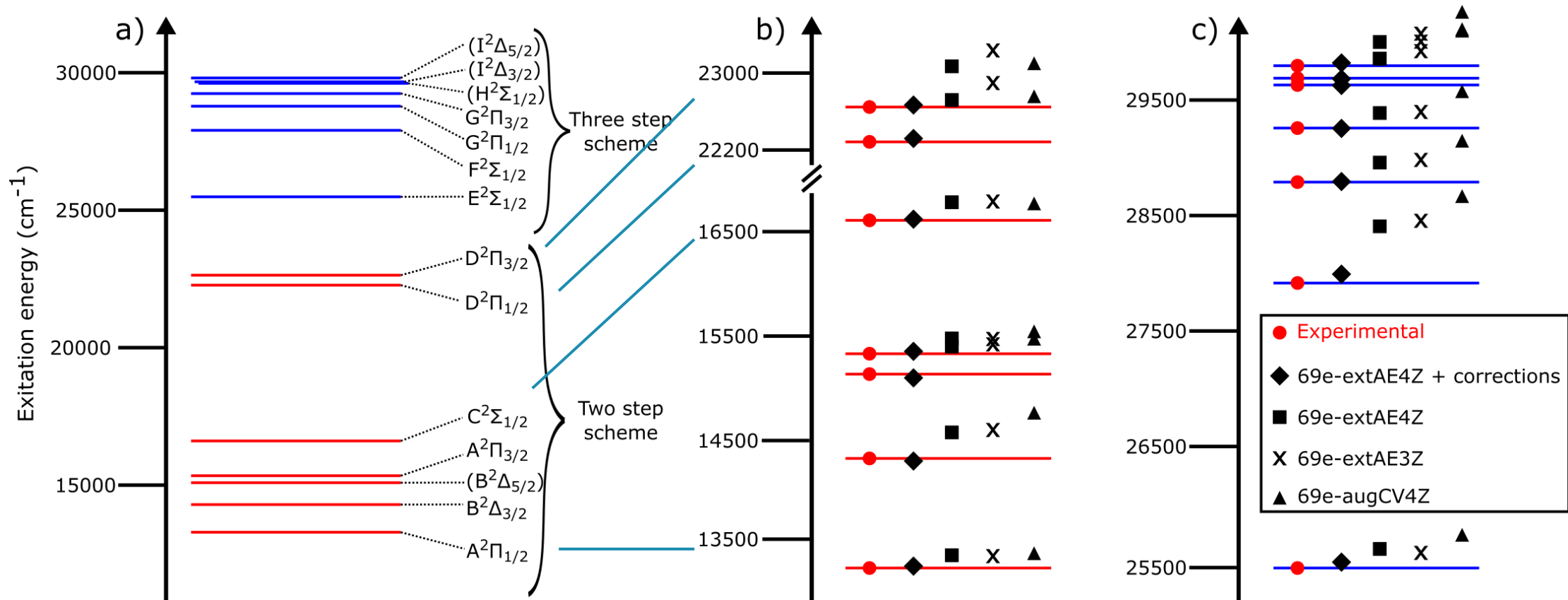
High resolution ^{223}RaF spectrum (Q-branch)

- Measured changing the wavelength of the second step (J-selectivity).
- Allows the access of different rotational lines in the Q-branch (e.g., low and high J)

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Final level scheme of RaF – all levels below 30.000 cm^{-1}

- All predicted electronic levels below 30,000 cm^{-1} have been measured and analyzed.

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Nuclear moments from molecules

The measurement of nuclear moments using molecules is not new, and several isotopes have their current reference moments from molecular measurements:

Magnetic dipole moments

- 35S

- 79Se

- 21Ne

Electric quadrupole moments

- 2H, HD

- 33S, CS and SiS

- 37Cl, BrCl

- 39-41K, KF and KCl, KBr

- 75As, AsP

- 79Br, HBr

- 115In?, InF and InI

- 121Sb, SbX, X=N,P,F,Cl

- 131Xe, Six molecules

- 139La, LaX, X=F,I

-

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