Collinear resonance ionization spectroscopy of RaF molecules

P-546

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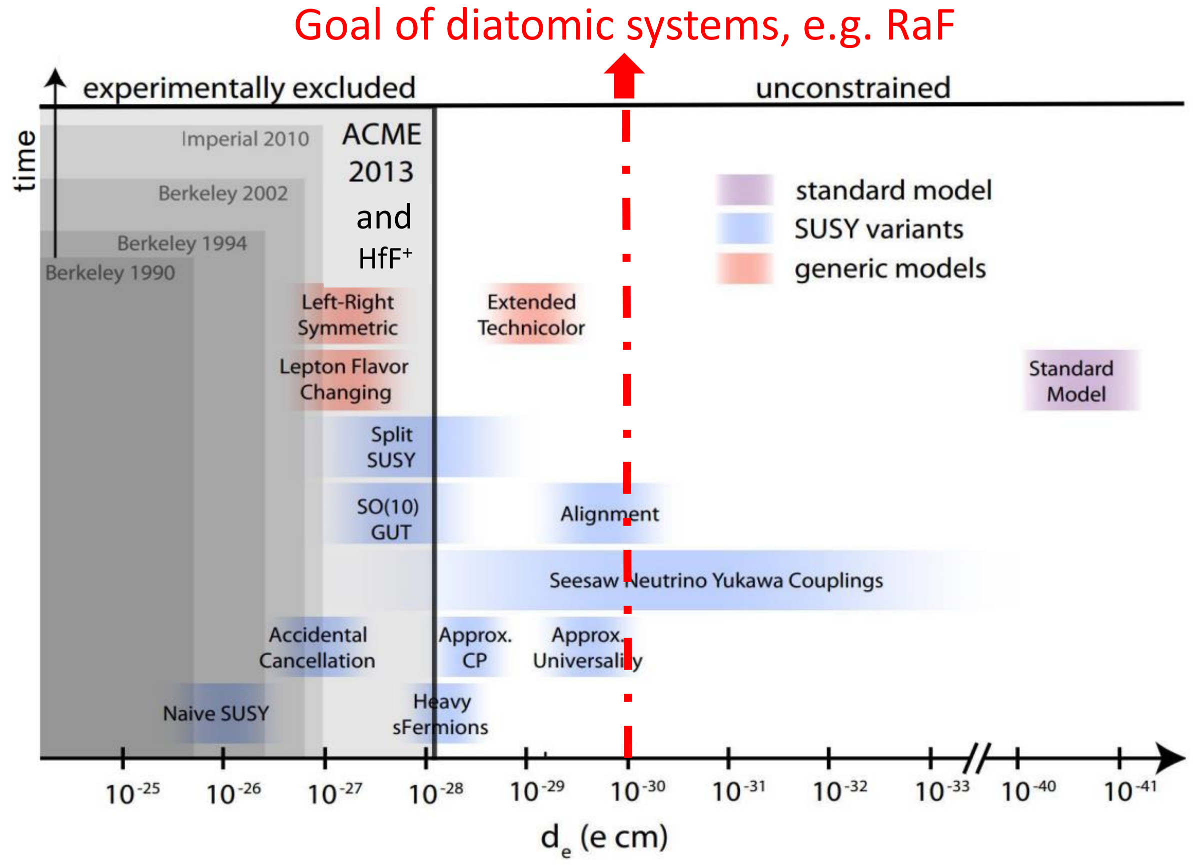
CRIS

Motivation: supporting the search for new physics

- Molecules provide compelling advantages for the search for P- and PT-odd interactions
  - For instance, the best limits on eEDM: molecular probes (2010: YbF, 2013: ThO, 2017: HfF^+)

- RaF is predicted to be an excellent candidate for future work



Adapted from B. Spaun, PhD thesis

Isaev, T. A. et al. R. **Laser-cooled RaF as a promising candidate to measure molecular parity violation.** PRA 82, 052521 (2010).
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- Molecules provide compelling advantages for the search for P- and PT-odd interactions
  - For instance, the best limits on eEDM: molecular probes (2010: YbF, 2013: ThO, 2017: HfF^+)
- RaF is predicted to be an excellent candidate for future work:
  - high sensitivity to electron EDM with a large E_{eff}
  - high sensitivity to other P- and P, T-parity-violating effects
  - Structure well suited to laser-cooling
  - …

	|Wa|/Hz (P-odd)	|Ws|/kHz (P,T-odd)	$\|E_{eff}\|$GV/cm (eEDM)
BaF	1.9×10^2	8.5	7.8
HfF^+	-	20	22.7
YbF	6.1×10^2	41	24.6
RaF	**2.1×10^3**	**150**	**57**

[1] Gaul & Berger J. of Chem. Phys 147, 014109(2017).
[2] Fleig. Phys. Rev. A 96, 040502 (2017)
[2] Kudashov et al. Phys. Rev. A 90, 052513 (2014).
[2] Kozlov et al. Phys. Rev. A 56, 3326 (1997).
[3] Mosyagin et al. JPB, 31, 763 (1998).

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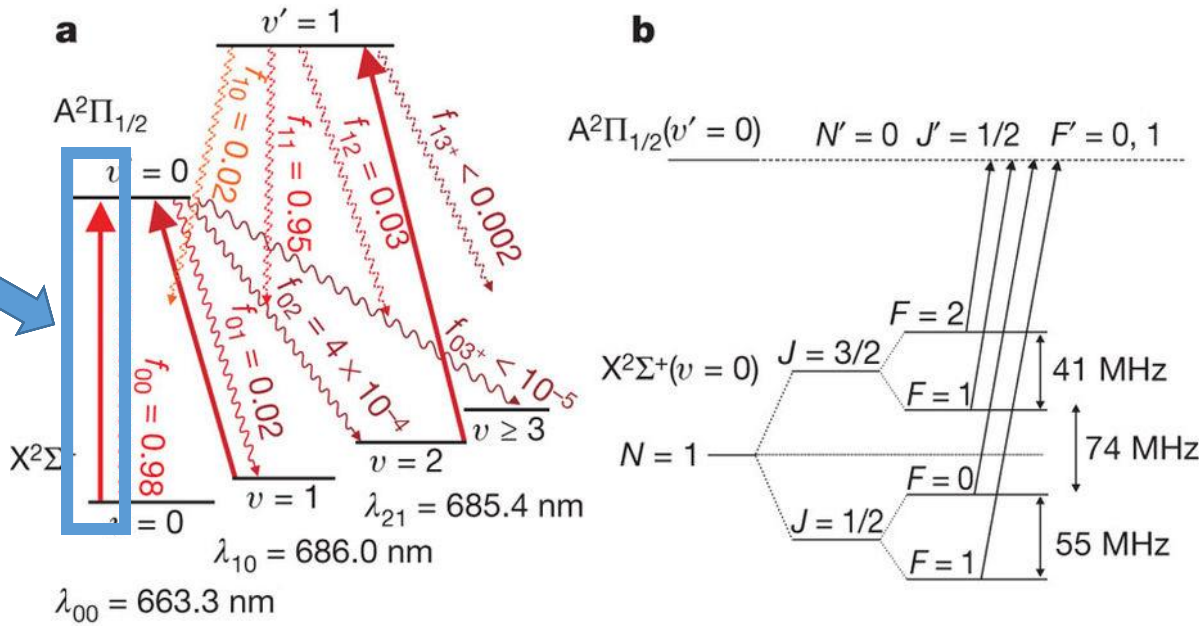
- BUT… no experimental data exists at all, **complicating the development of laser cooling schemes**

e.g.: What is the transition energy of the equivalent transition in RaF??



Cooling scheme for SrF, taken from Laser cooling of a diatomic molecule, Nature 467, 820–823 (2010)

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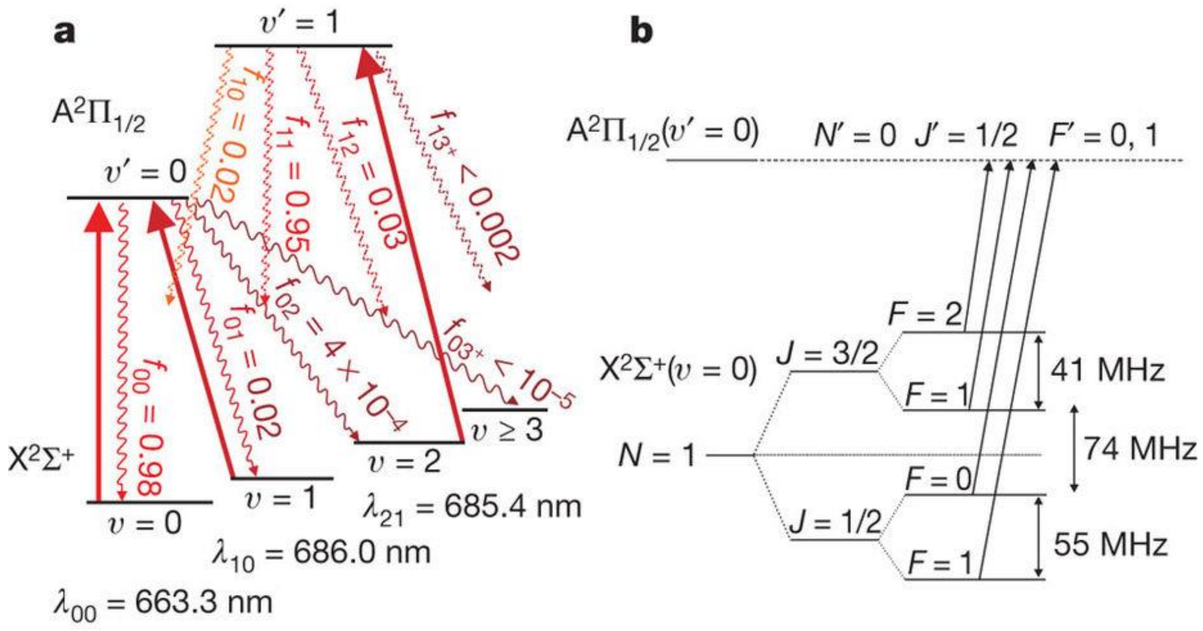
- AND… interpretation of experiments relies on quantum chemistry calculations, **which need to be benchmarked**



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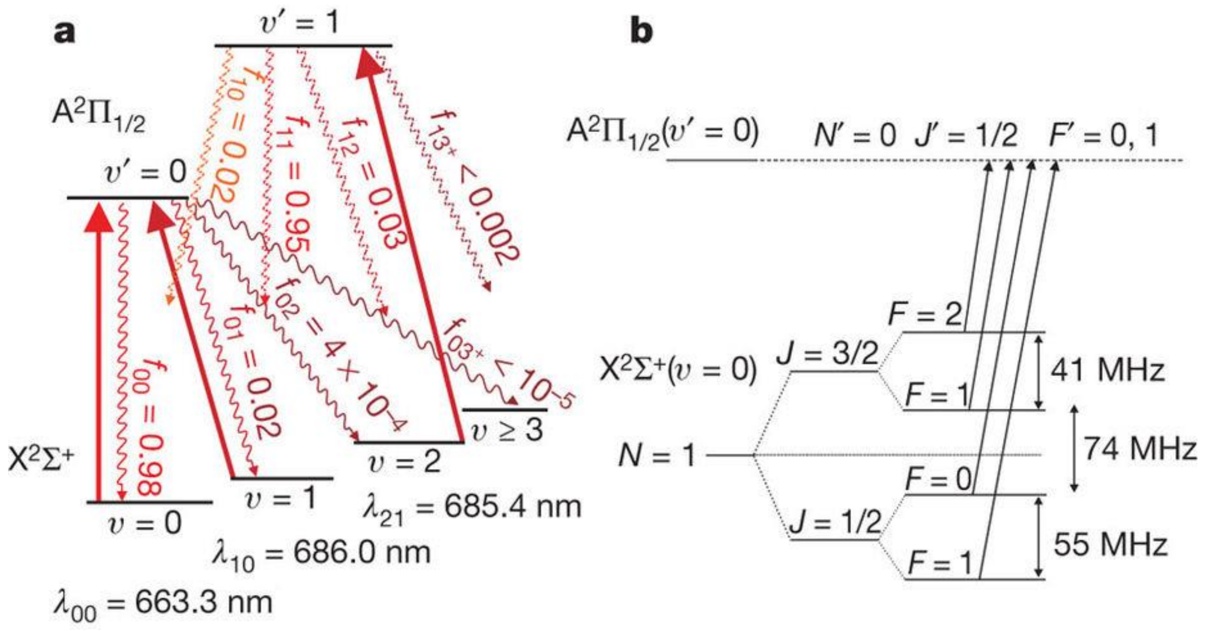
Motivation: supporting the search for new physics

- BUT… no experimental data exists at all, **complicating the development of laser cooling schemes**
- AND… interpretation of experiments relies on quantum chemistry calculations, **which need to be benchmarked**
- So, we propose to measure the following spectroscopic properties of RaF:
  - excitation energy of low-lying levels
  - ionization potential
  - Hyperfine structure of ^{225}RaF -> probes E_{eff} for eEDM!



Cooling scheme for SrF, taken from Laser cooling of a diatomic molecule, Nature 467, 820–823 (2010)

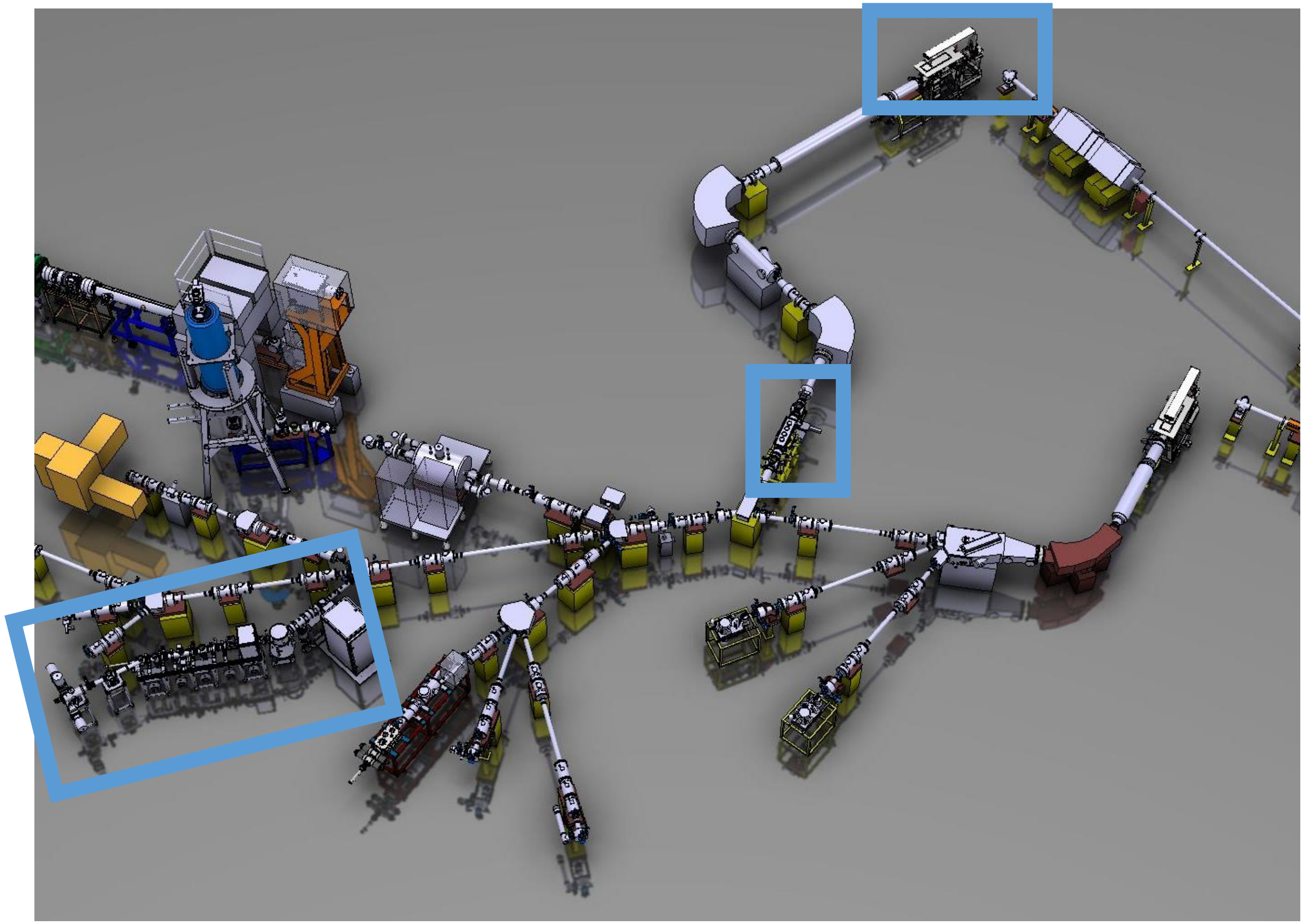
Test validity of (ab-initio) quantum chemistry calculations

Significantly boost the development of laser cooling schemes of RaF **required** for future high-precision measurements

Experimental method

- Produce ^{138}BaF and ^{225,226}RaF ions at HRS
  - Irradiated target required, but
  - No protons required during experiment itself
- Cool and bunch, inject into the CRIS beamline

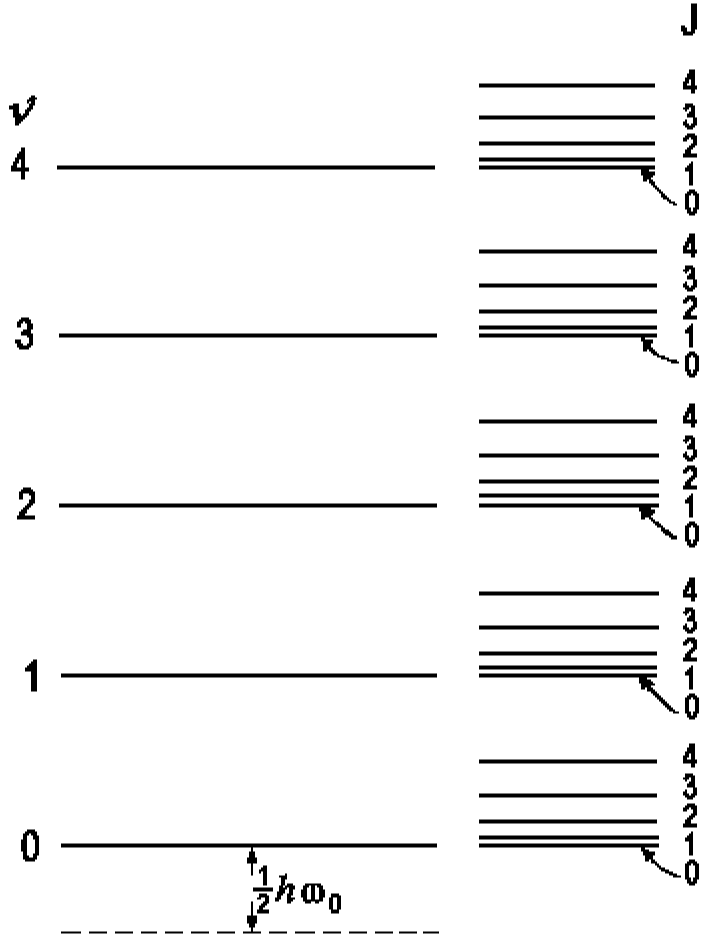
Molecule	Half-life	Yield
$^{138}BaF^{+}$	Stable	$> 10^6$
$^{225}RaF^{+}$	15 d	$> 10^5$
$^{226}RaF^{+}$	1600 y	$> 10^6$



Experimental method

- Neutralize molecular ions using standard charge exchange cell
  - Theoretically estimated cross sections are sufficiently large for efficient neutralization
  - However, only a fraction of the neutral molecules are in any given rovibrational J, ν-state

  ➔ efficiency of CEC into the state of interest estimated at 1:1000

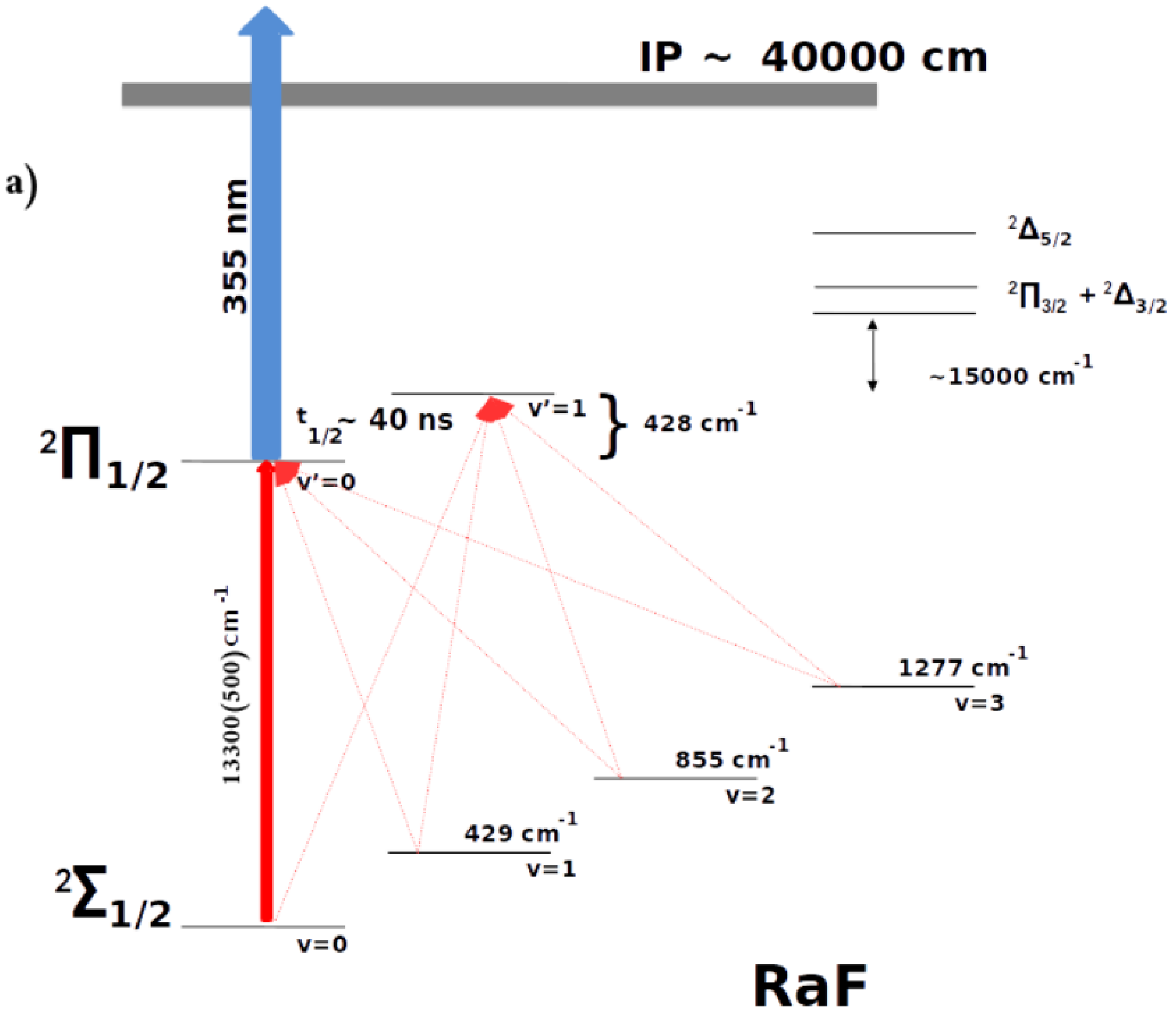


Schematic illustration of vibrational and rotational energy levels of a diatomic molecule

Isaev, T. A. et al**. Ion neutralisation mass-spectrometry route to radium monofluoride (RaF)**. arXiv:1310.1511v3 (2013).

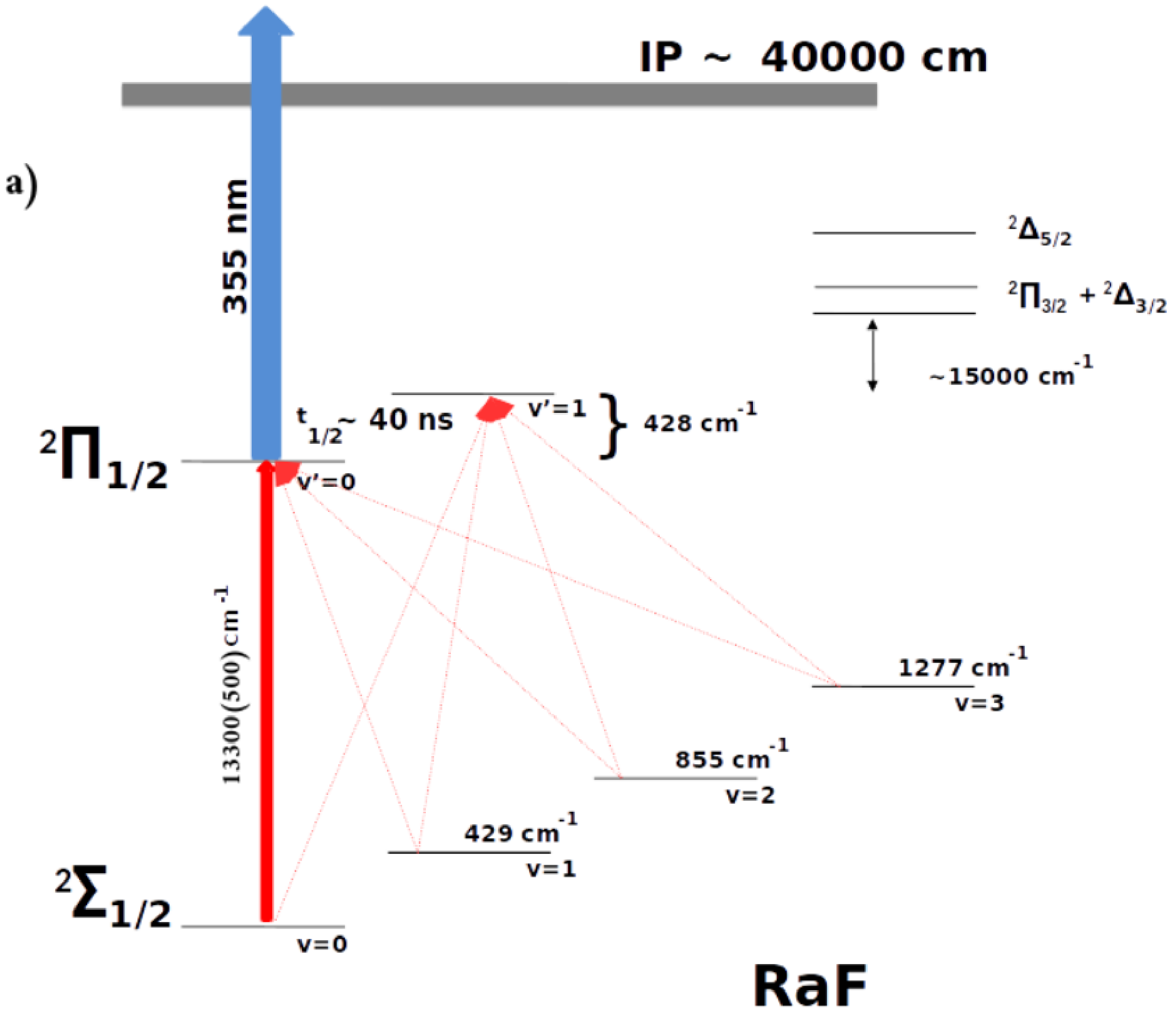
Experimental method

- Neutralize molecular ions using standard charge exchange cell
- Laser-ionize RaF using a two-colour laser scheme
  - Theoretical calculations of transition wavenumber: 12800-13800 cm^{-1}
  - Two broadband laser systems to minimize scanning time
  - Once resonance located, scan with narrowband system



Experimental method

- Neutralize molecular ions using standard charge exchange cell
- Laser-ionize RaF using a two-colour laser scheme
- Detect laser-ionized molecules with efficient charged-particle detector (MCP)
  - Background due to collisional ionization of RaF estimated at $1{:}10^4$-10^5 of total neutrals



Beamtime estimate

Note: All shifts are without protons!

- **1 shift** for setup and initial optimization with BaF from ISOLDE target
  - Known ionization scheme, so no looking for the first resonance
  - Required to ensure shifts on RaF are used optimally

Beamtime estimate

Note: All shifts are without protons!

- **1 shift** for setup and initial optimization with BaF from ISOLDE target
- **14 shifts** Find excitation energy of $^2\Pi_{1/2}$ excited state in ^{226}RaF
  - Broadband
    - 100 cm^{-1} = 30000 GHz to scan
    - 65 seconds per 6 GHz means ~80 hrs of continuous scanning
    - + 5 hours for regular laser optimization
  - Narrowband
    - Scan range of only 30 GHz, but much slower scan speed; 28 hrs

Beamtime estimate

Note: All shifts are without protons!

- **1 shift:** Setup and initial optimization with BaF from ISOLDE target
- **14 shifts:** Find excitation energy of $^2\Pi_{1/2}$ excited state in ^{226}RaF
- **1 shift:** Determine Ionization Potential of ^{226}RaF
  - Fix first step laser to the newly discovered transition wavelength
  - Scan second step using broadband, tunable dye laser
- **2 shifts:** Determine Hyperfine structure of ^{225}RaF
  - Transition wavelength can be well predicted from results on ^{226}RaF
  - ~100 GHz scan range

In summary

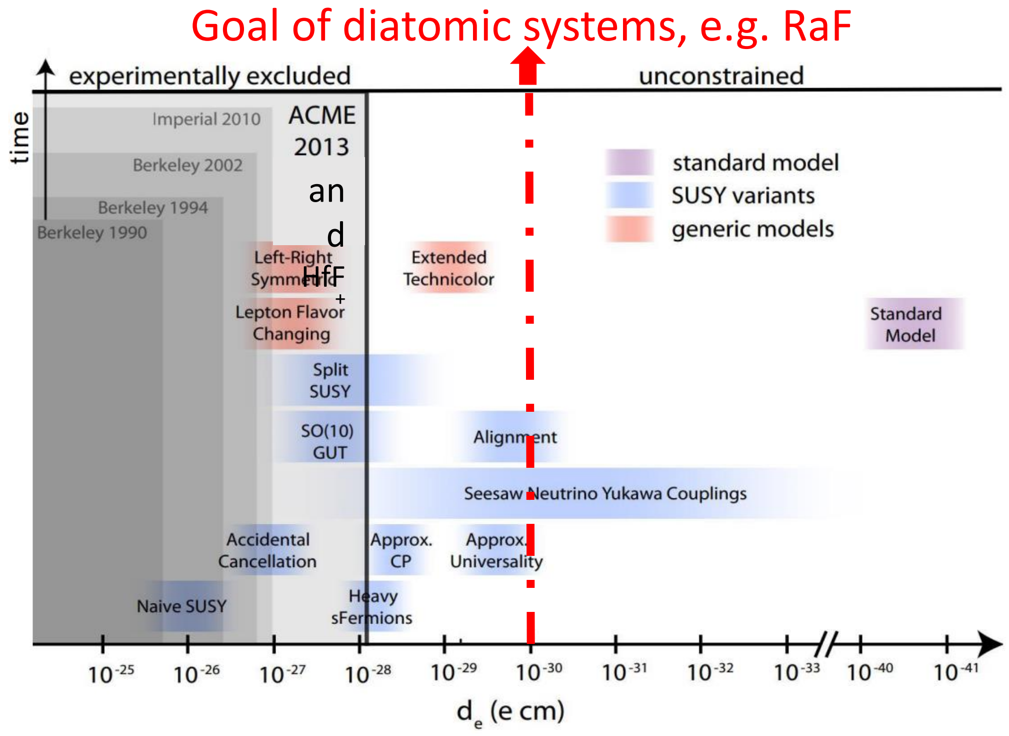
Goal: Measurement of spectroscopic properties of the diatomic molecule RaF

- excitation energy of low-lying levels
- ionization potential
- Hyperfine structure

Through these measurements, we can

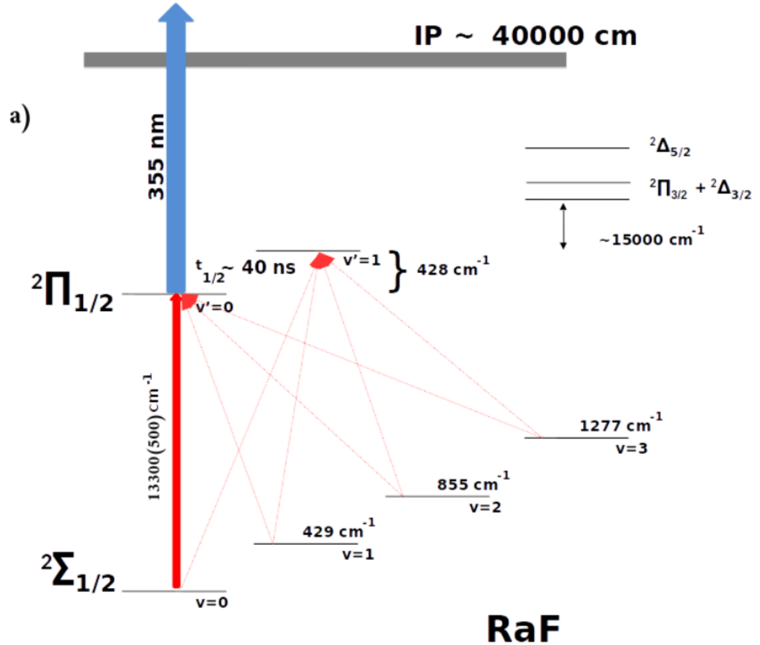
- Test validity of (ab-initio) quantum chemistry calculations
- Significantly boost the search for laser cooling schemes of RaF

These measurements will make a strong impact on future searches for physics beyond the standard model.



Molecule	Half life	Yield (ions/s)	Target	Shifts
$^{222}RaF^+$	38 s	2×10^6	UC_x	–
$^{225}RaF^+$	15 d	$> 10^5$	UC_x	2
$^{226}RaF^+$	1600 y	$> 10^6$	UC_x	15
$^{138}BaF^+$	stable	$> 10^6$	UC_x	1

No shifts with protons required



Figures from S. Rothe et al, Nature Communications **4**, 1835 (2013)

IP determination

Allowed transitions to Rydberg states?

yes

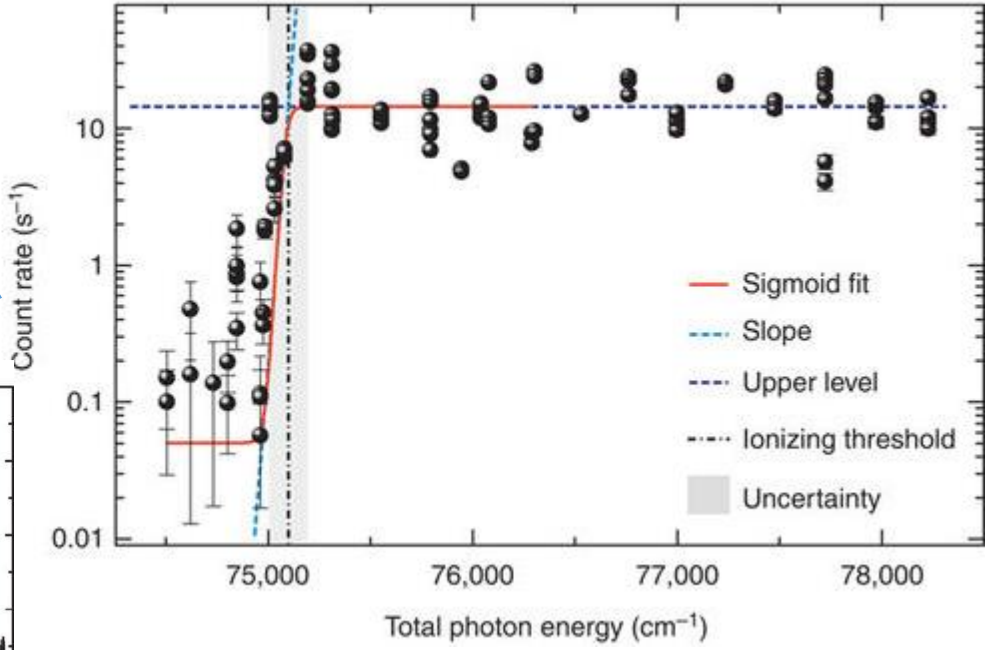
no

Laser ionized as well?

no

yes

c



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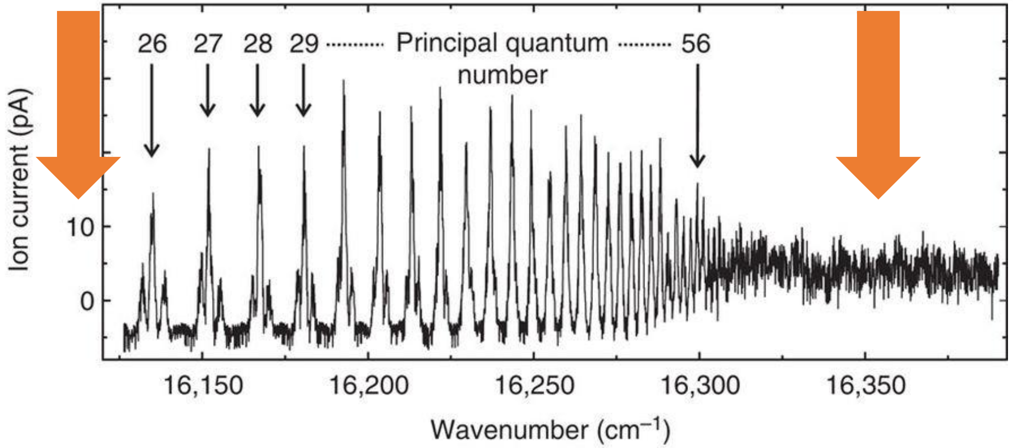

Coarse scan with only ~100 cm-1 precision, starting point for future studies of Rydberg series

The CRIS method