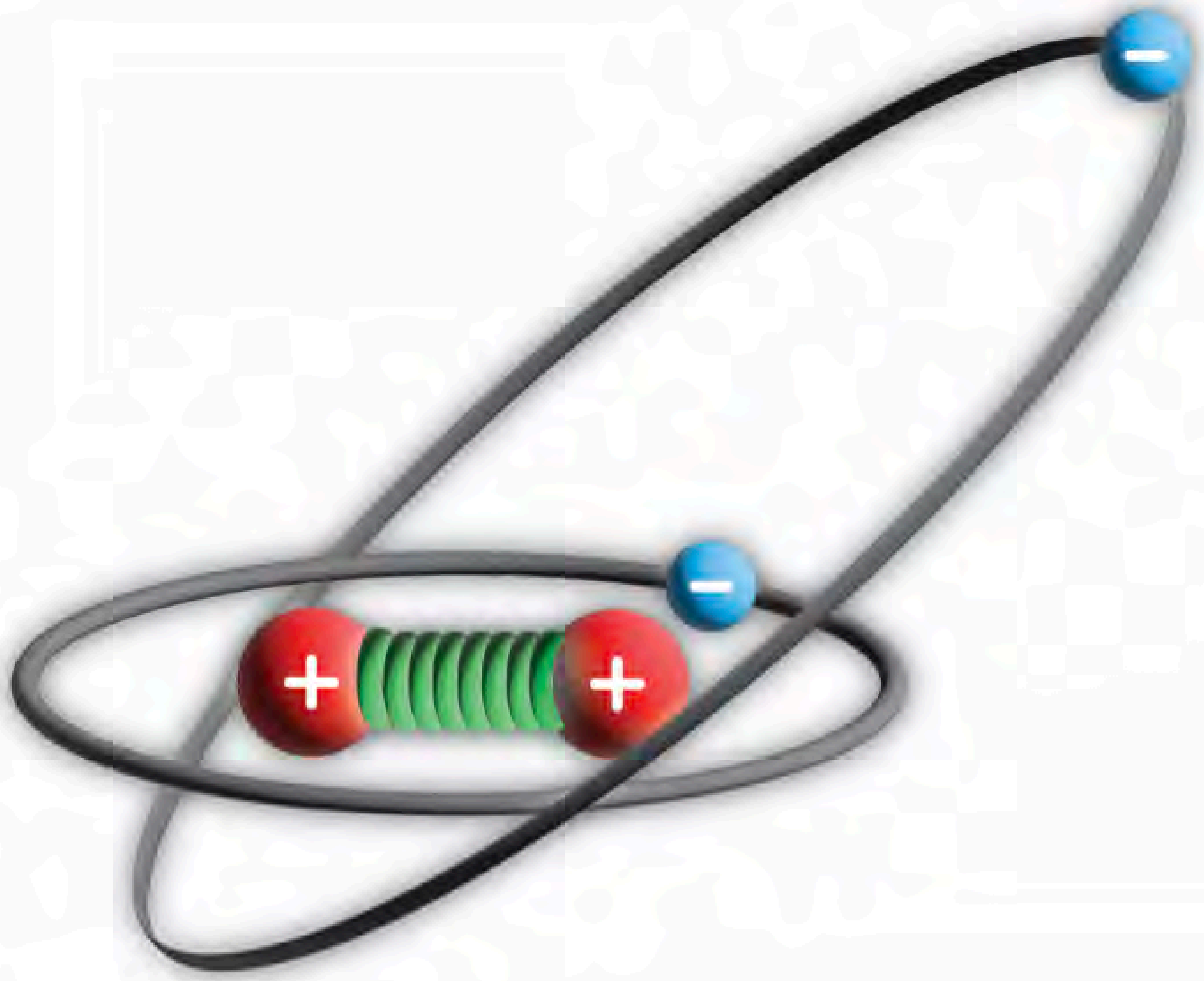
High-precision spectroscopy of hydrogen molecules



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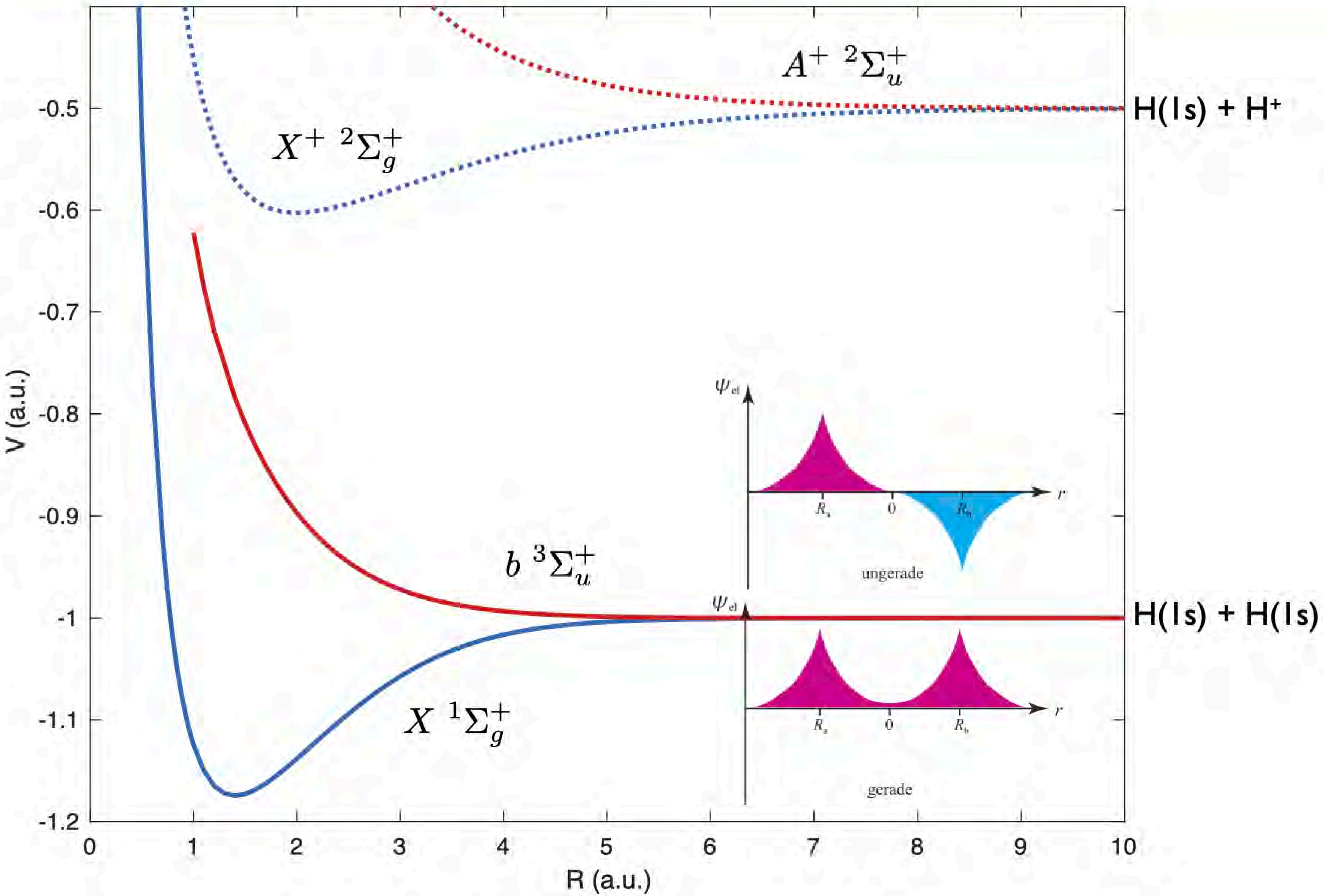
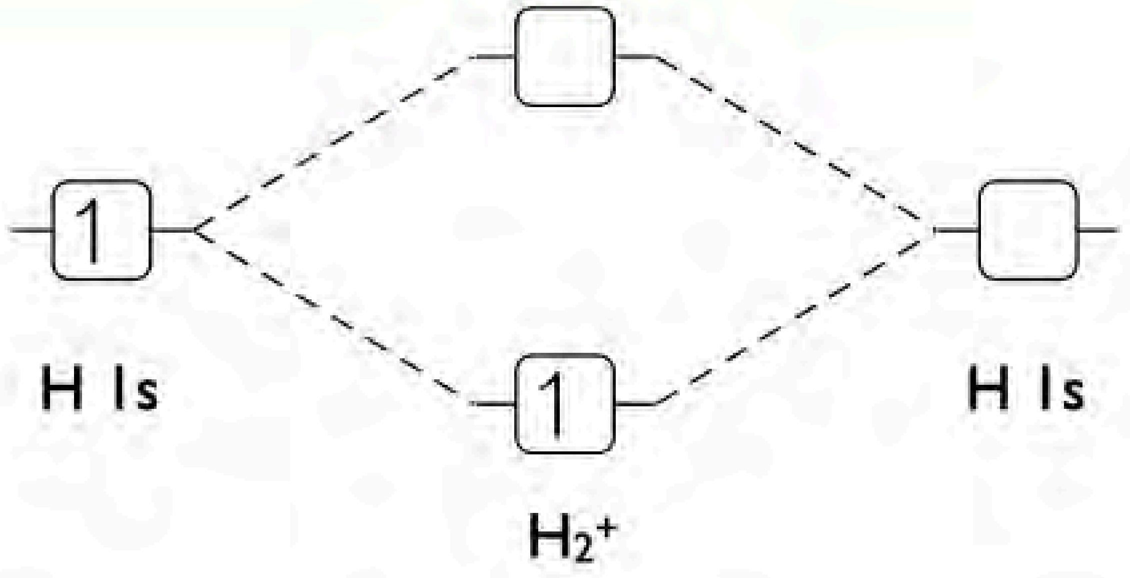
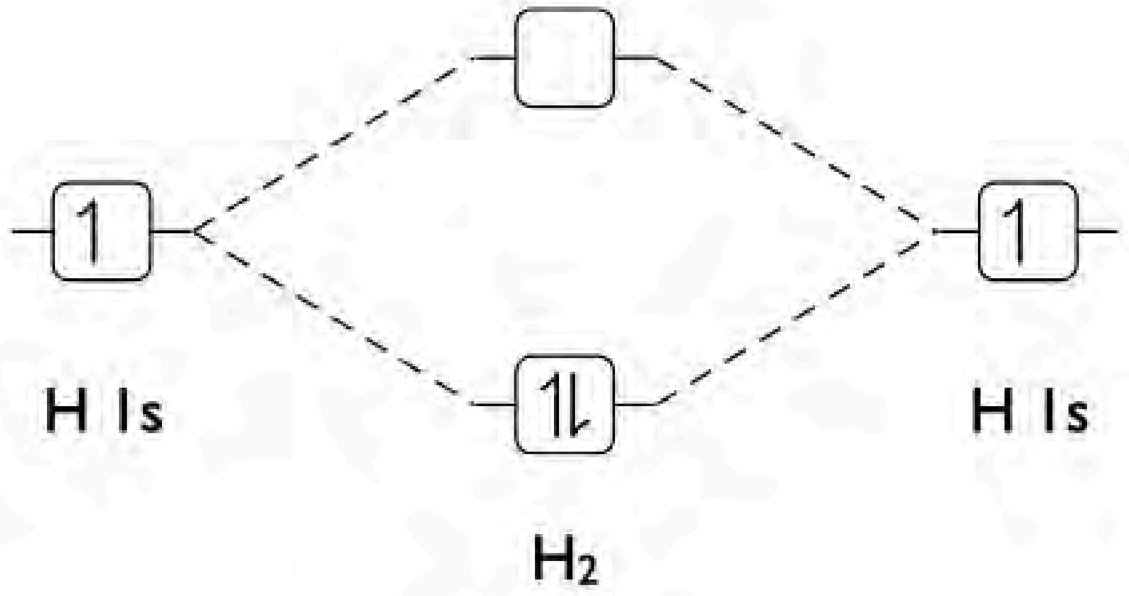
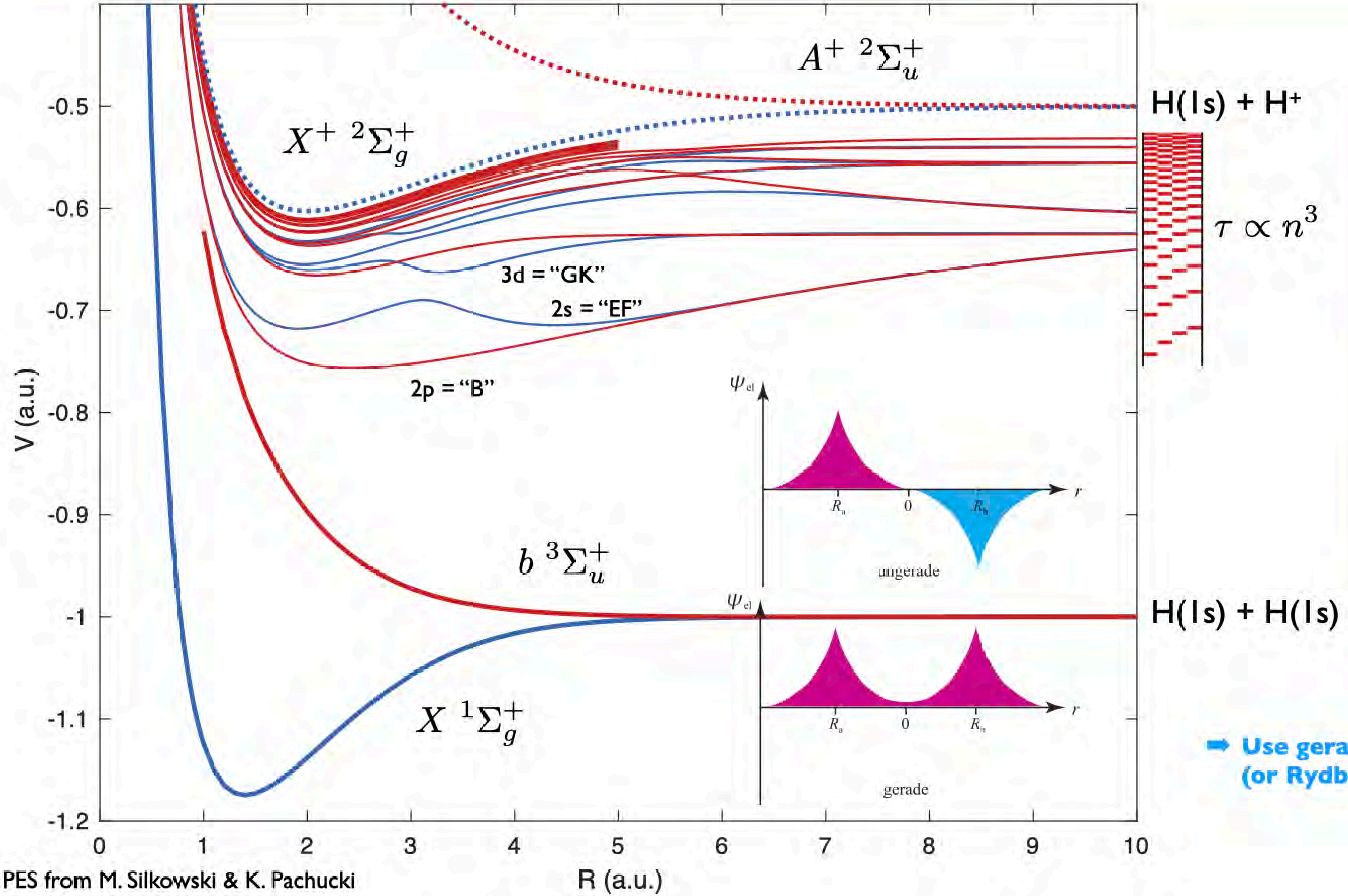
Molecular Hydrogen







Molecular Hydrogen



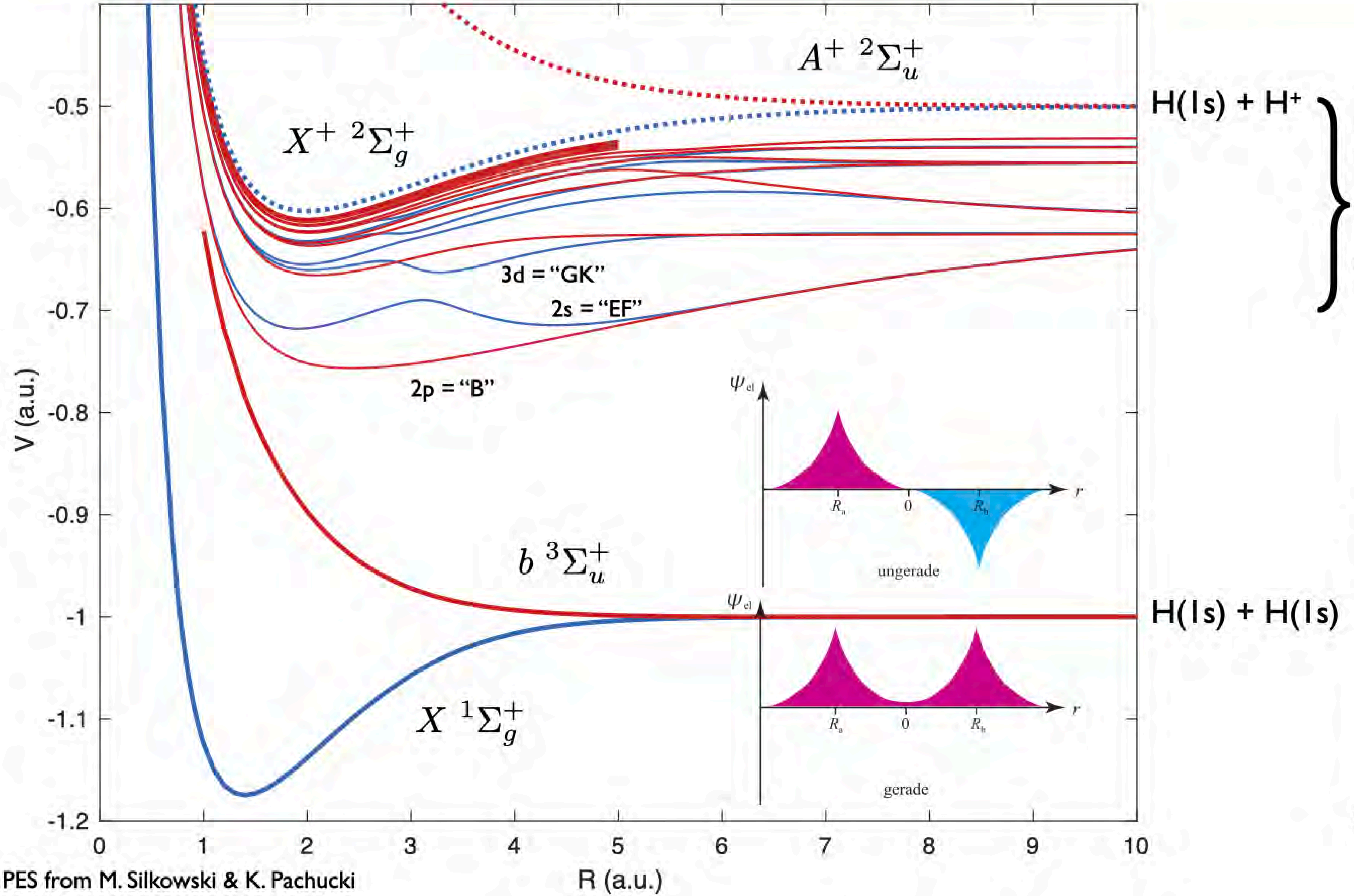
PES from M. Silkowski & K. Pachucki

$\tau\left(B\ ^1\Sigma_u^+\right) < 1$ ns	2pσ
$\tau\left(C\ ^1\Pi_u\right) < 1$ ns	2pπ
$\tau\left(B'\ ^1\Sigma_u^+\right) \sim 5$ ns	3pσ
$\tau\left(D\ ^1\Pi_u\right) \sim 3$ ns	3pπ
$\tau\left(EF\ ^1\Sigma_g^+\right) \sim 200$ ns	2sσ
$\tau\left(GK(v=0)\ ^1\Sigma_g^+\right) \sim 70$ ns	3dσ
$\tau\left(GK(v=1)\ ^1\Sigma_g^+\right) \sim 24$ ns	3dσ
$\tau\left(H\ ^1\Sigma_g^+\right) \sim 100$ ns	3sσ
$\tau\left(I\ ^1\Pi_g^+\right) \sim 20$ ns	3dπ
$\tau\left(O\ ^1\Sigma_g^+\right) \sim 170$ ns	4sσ

Astashkevich and Lavrov,
J. Phys. Chem. Ref. Data **44**, 023105-1 (2015)

➡ **Use gerade states for precision measurements (or Rydberg states)**

Molecular Hydrogen in this talk



PES from M. Silkowski & K. Pachucki

ETH Zürich (Merkt)

- Rydberg states

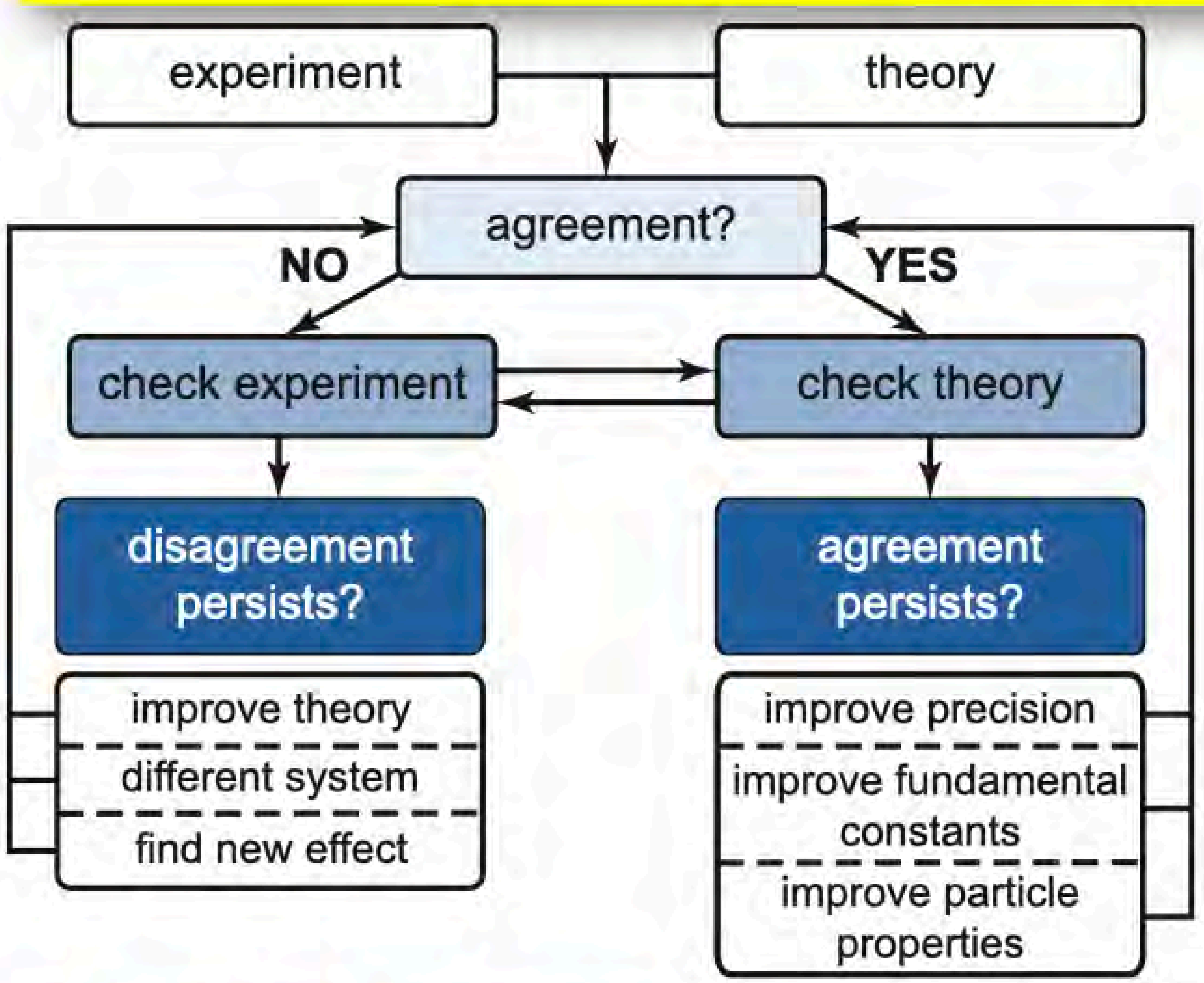
VU

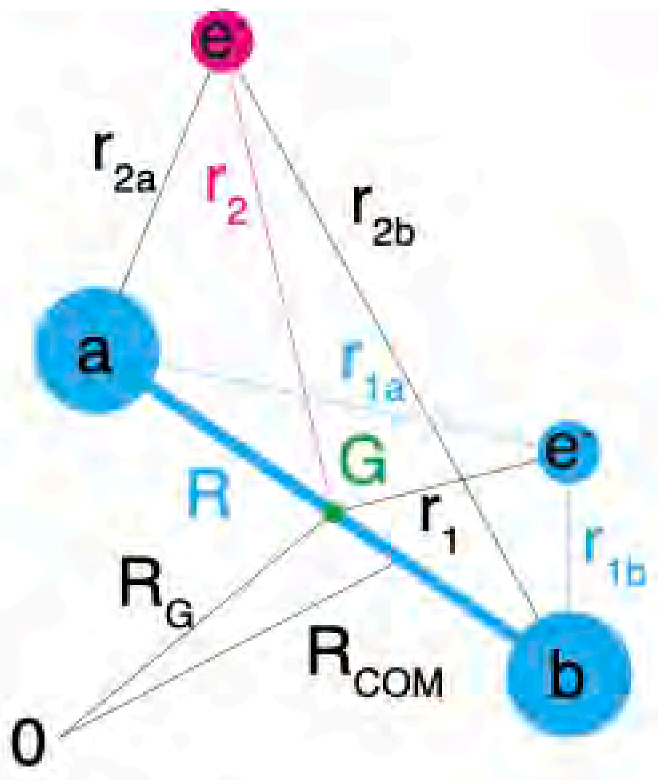
- weakly-bound states

VU Amsterdam (Ubachs, Eikema)

- X-GK (= 1s-3d)
- X-EF (= 1s-2s)

Theory





Goals

➡ **Provide benchmark to test/improve theory**

➡ **Extract values of fundamental constants (R, r_p, m_p/m_e)**

$$\mathcal{H}_{\text{rve}} = \underbrace{-\frac{\nabla_1^2}{2} - \frac{\nabla_2^2}{2} + \frac{1}{R} - \frac{1}{r_{1a}} - \frac{1}{r_{1b}} - \frac{1}{r_{2a}} - \frac{1}{r_{2b}} + \frac{1}{r_{12}}}_{H_{\text{cn}}}$$

$$\underbrace{-\frac{\nabla^2_{R\Theta\Phi}}{2\mu}}_{H_1'} \underbrace{-\frac{\nabla_1^2}{8\mu} - \frac{\nabla_2^2}{8\mu} - \frac{\nabla_1\cdot\nabla_2}{4\mu}}_{H_2'} \underbrace{-\frac{\nabla_{R\Theta\Phi}\cdot(\nabla_1+\nabla_2)}{2\mu_\alpha}}_{H_3'},$$

$$E_{\text{H}}^{\text{NA}} = \frac{m_{\text{p}}}{m_{\text{p}}+1}\left(-\frac{1}{2}\right) = \left(1+\frac{1}{m_{\text{p}}}\right)^{-1}\left(-\frac{1}{2}\right) = -\frac{1}{2} + \frac{1}{2m_{\text{p}}} - \frac{1}{2m_{\text{p}}^2+2m_{\text{p}}}$$

59.8 cm^{-1}
1.79 THz

- 0.0325 cm^{-1}
- 975 MHz

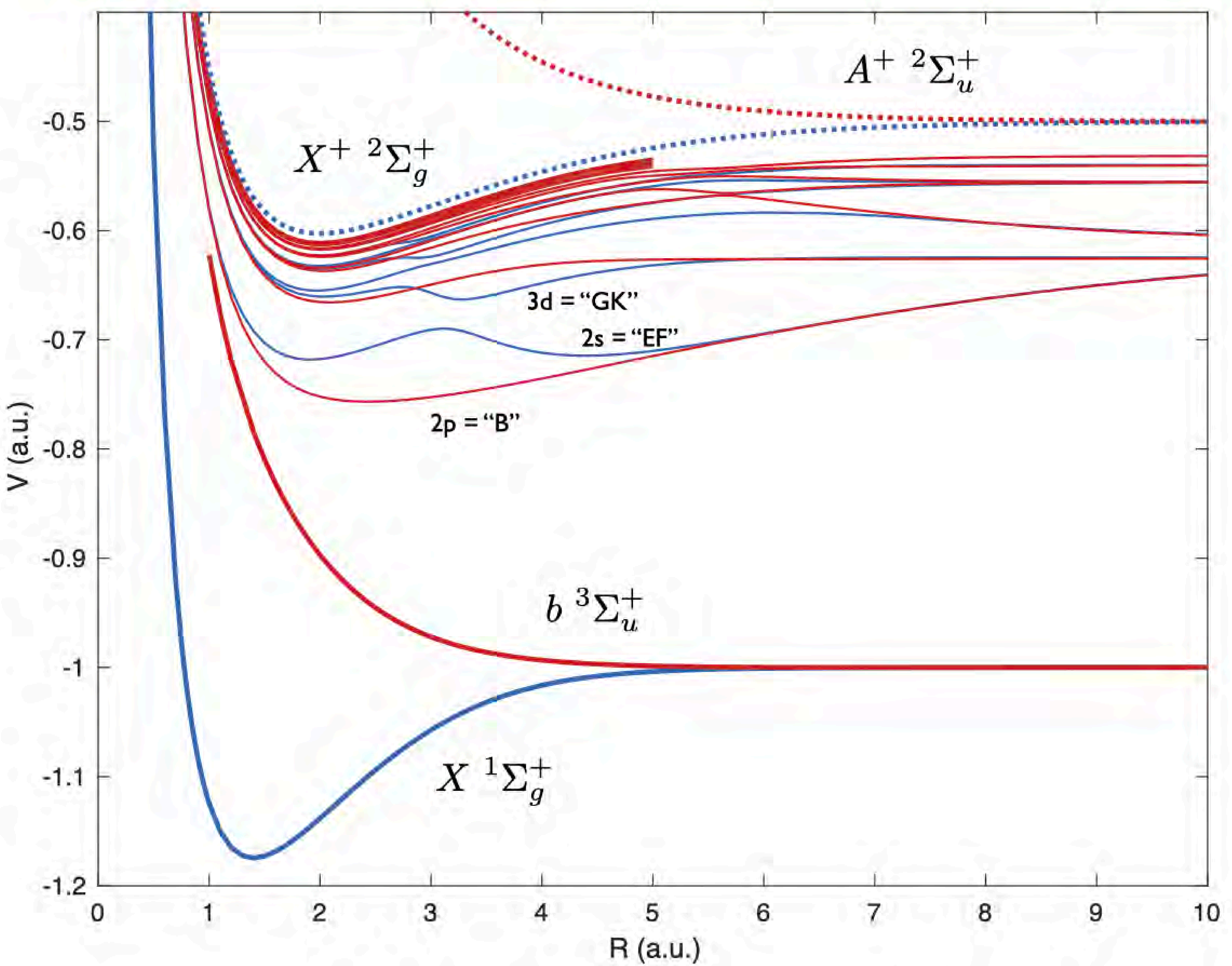
Born-Oppenheimer approximation (infinite nuclear mass)
➡ adiabatic correction (diagonal)
➡ nonadiabatic correction (off-diagonal)

} or full NA calculation (pre-BO)

$$E(\alpha) = m\alpha^2 E^{(2)} + m\alpha^4 E^{(4)} + m\alpha^5 E^{(5)} + m\alpha^6 E^{(6)} + m\alpha^7 E^{(7)} \ldots \quad \text{with}$$

$$E^{(i)} = E^{(i,0)} + \frac{m}{M}E^{(i,1)} + \left(\frac{m}{M}\right)^2 E^{(i,2)} + \ldots$$

Status of the theory

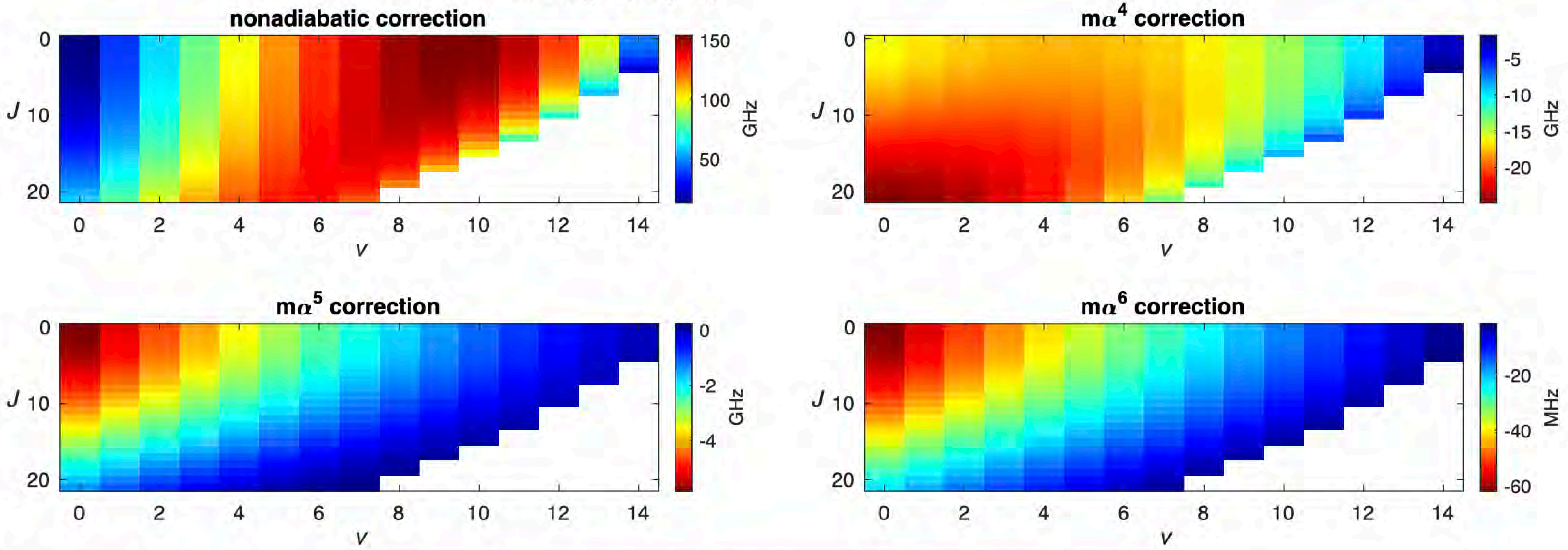


X^+, A^+	~ 3 MHz	preBO, ma^5	Moss (1993)
X^+ (v < 10)	~ 10 kHz	preBO, ma^8	Korobov, Karr, Hilico (2017)
Rydberg states (n > 40)	~ 300 kHz	MQDT	Jungen, Beyer, Merkt
EF, GK, …	~ 1 MHz	BO	Silkowsk, Pachucki
EF	~ 150 MHz	preBO, ma^7	Matyus (2019)
X	~ 0.78 MHz	preBO, ma^7	Pachucki + coworkers (2019)

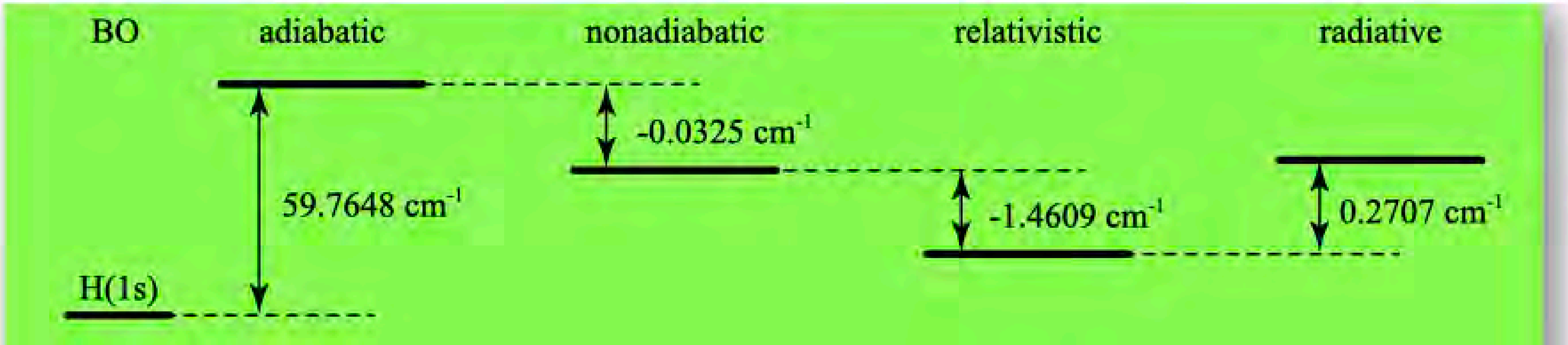
Corrections for the electronic ground state of H_2

Nonadiabatic corection $\Delta E_{\rm na} = -C \langle T \rangle_v$

Van Vleck, *JCP* **4**, 327 (1936)



Komasa et al., *PRA* **100**, 032519 (2019)
Lai et al., *PRL* **127**, 183001 (2021)



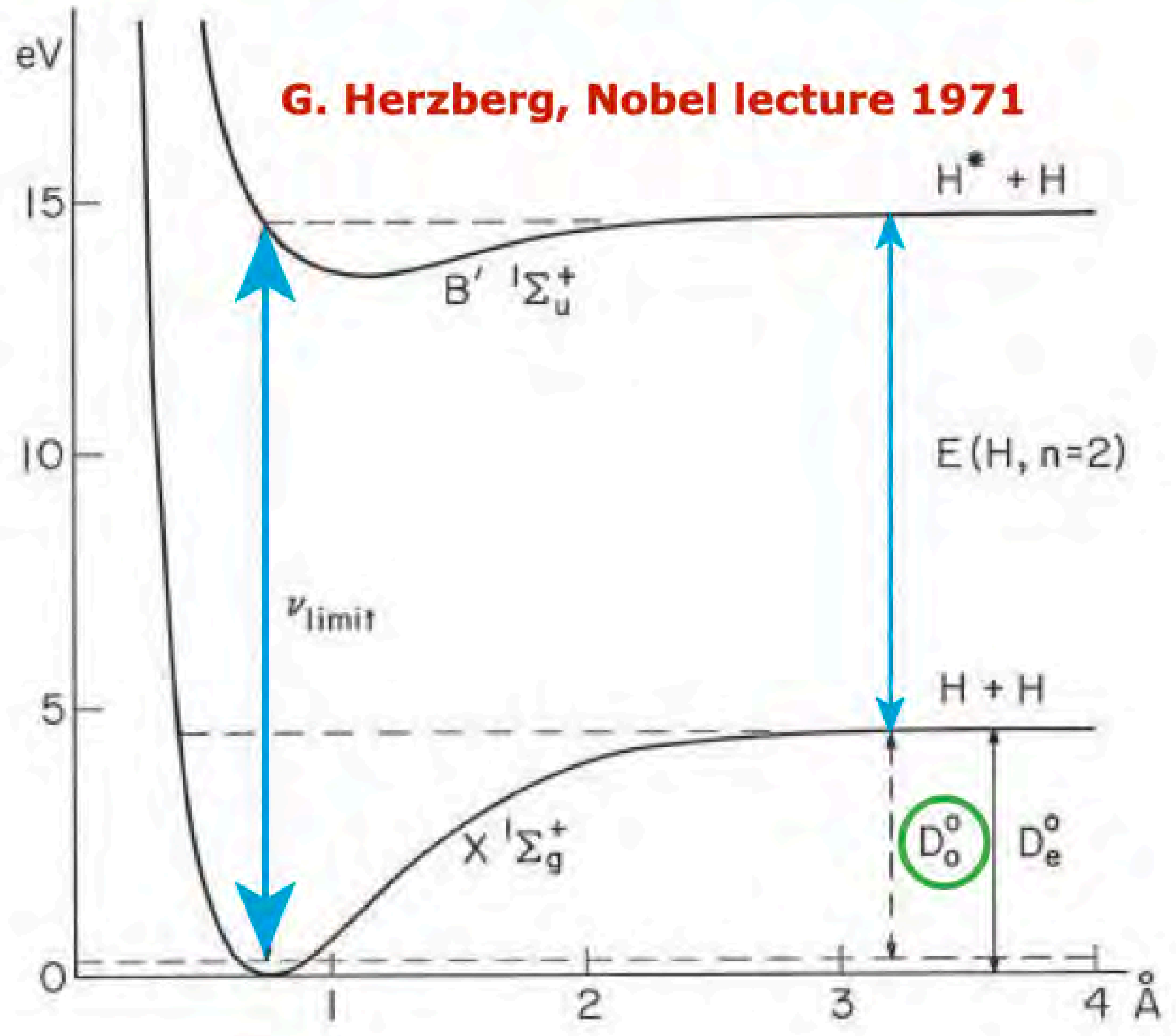
Measurement of the dissociation energy

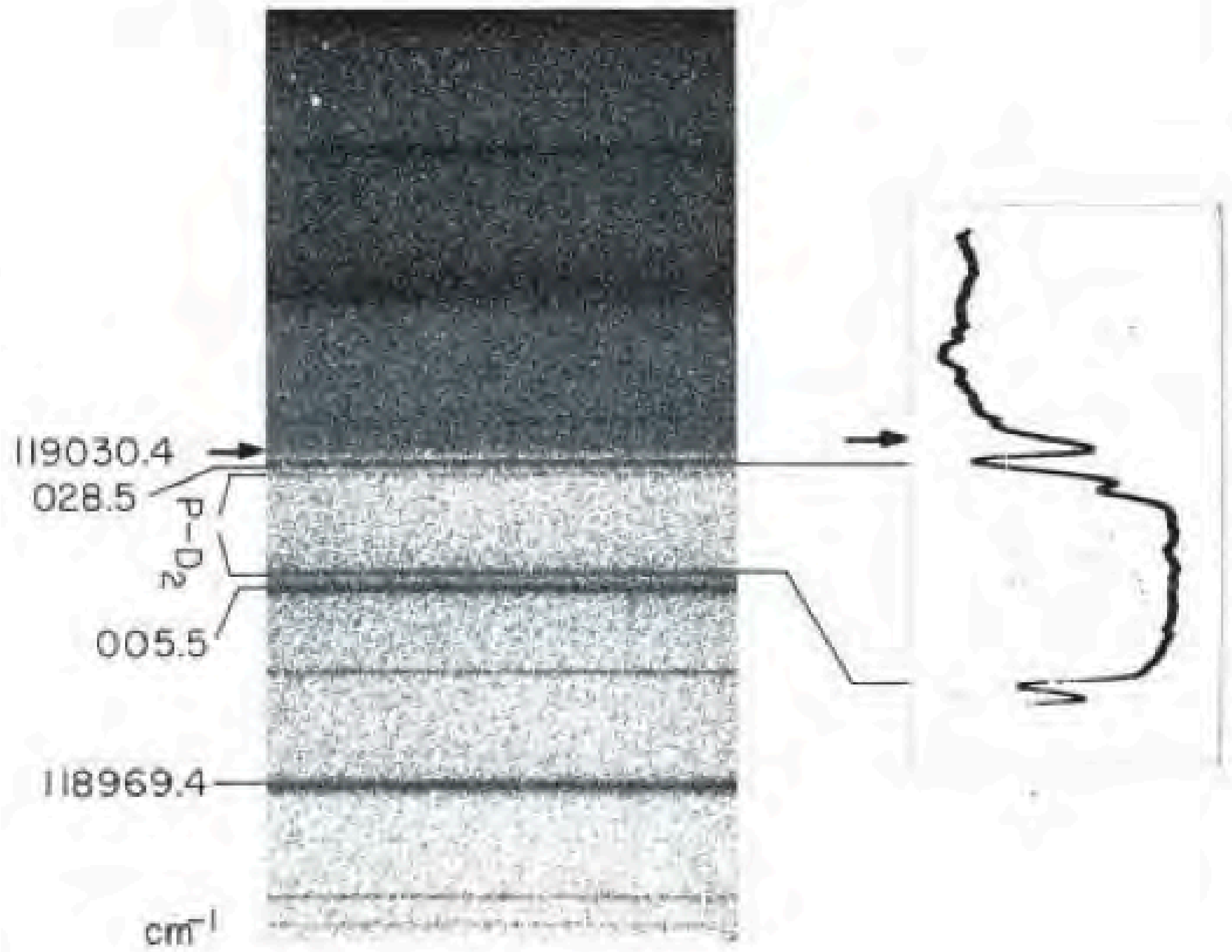
$$\left\langle \Phi'_{\rm elec}\Phi'_{\rm vib}\Phi'_{\rm rot} \left| \sum_\alpha \lambda_{\alpha Z}\hat{\mu}_\alpha \right| \Phi''_{\rm elec}\Phi''_{\rm vib}\Phi''_{\rm rot} \right\rangle = \sum_\alpha \langle \Phi'_{\rm rot} | \lambda_{\alpha Z} | \Phi''_{\rm rot} \rangle \langle \Phi'_{\rm elec}\Phi'_{\rm vib} | \hat{\mu}_\alpha | \Phi''_{\rm elec}\Phi''_{\rm vib} \rangle$$

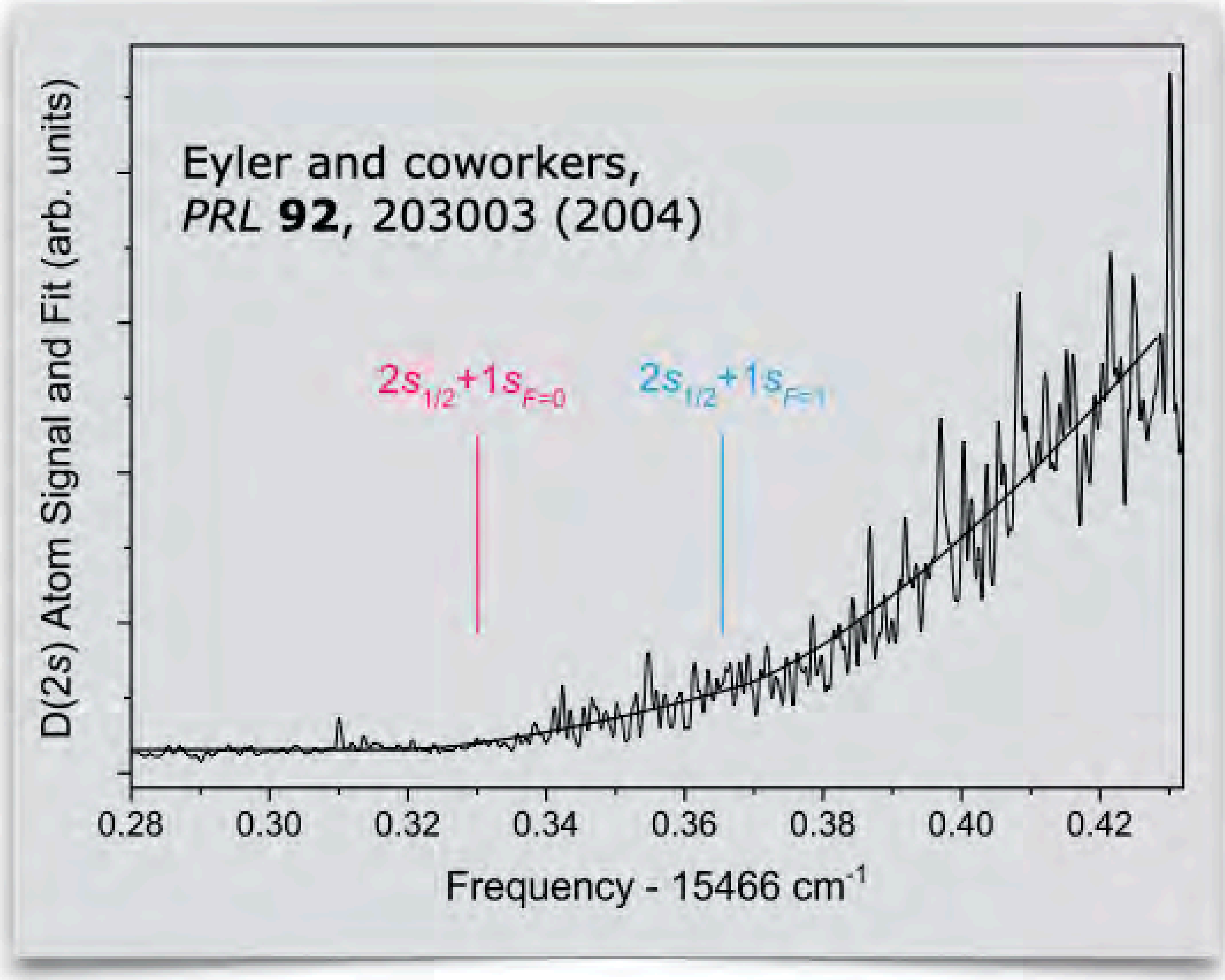
$$\langle \Phi'_{\rm vib} [\langle \Phi'_{\rm elec} | \hat{\mu}_\alpha | \Phi''_{\rm elec} \rangle_e] \Phi''_{\rm vib} \rangle = \langle \Phi'_{\rm vib} | \mu^e_\alpha(Q) | \Phi''_{\rm vib} \rangle$$

➡ **Zero permanent dipole moment for H_2, D_2**

➡ **No E1 rovibrational spectrum**

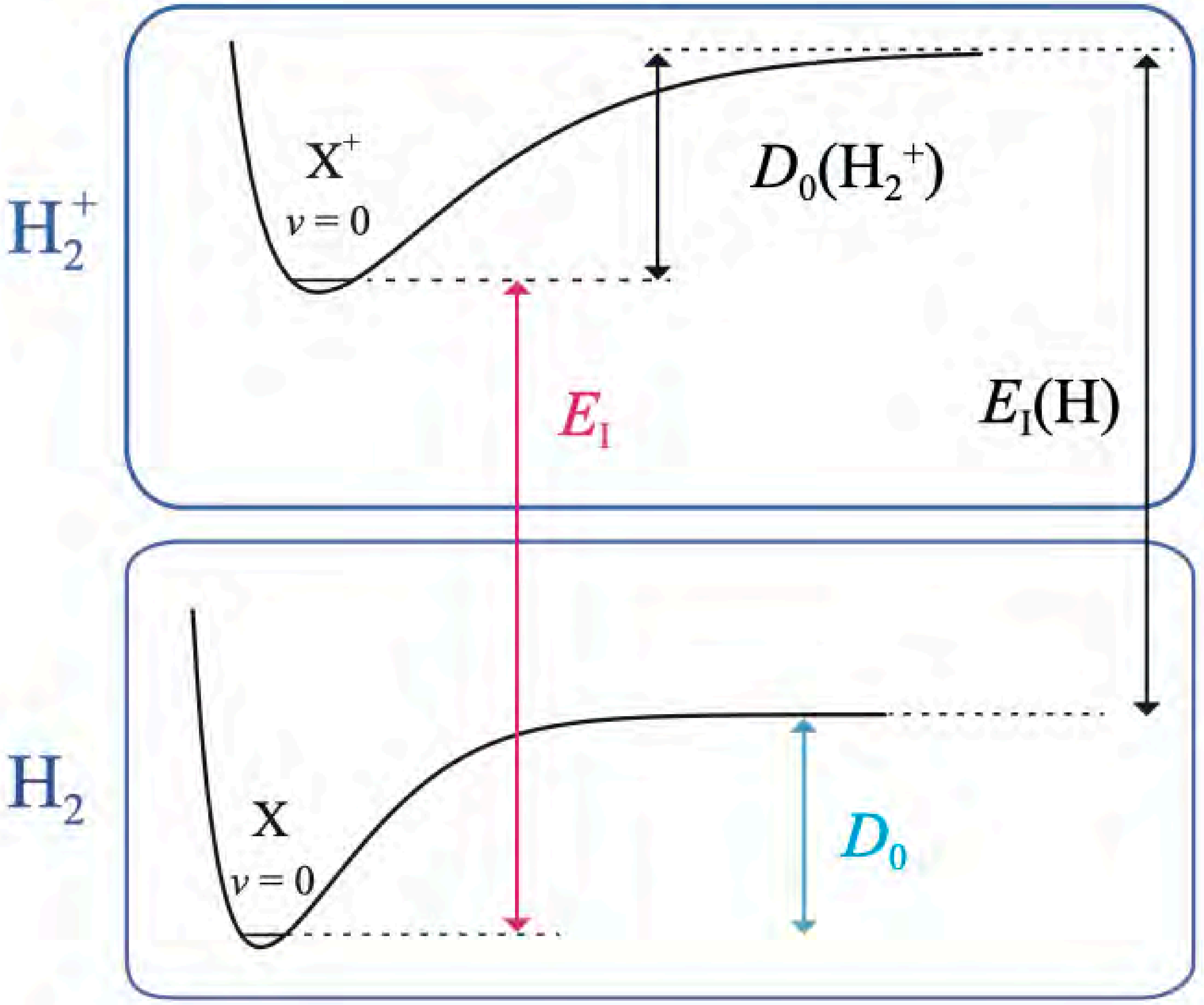


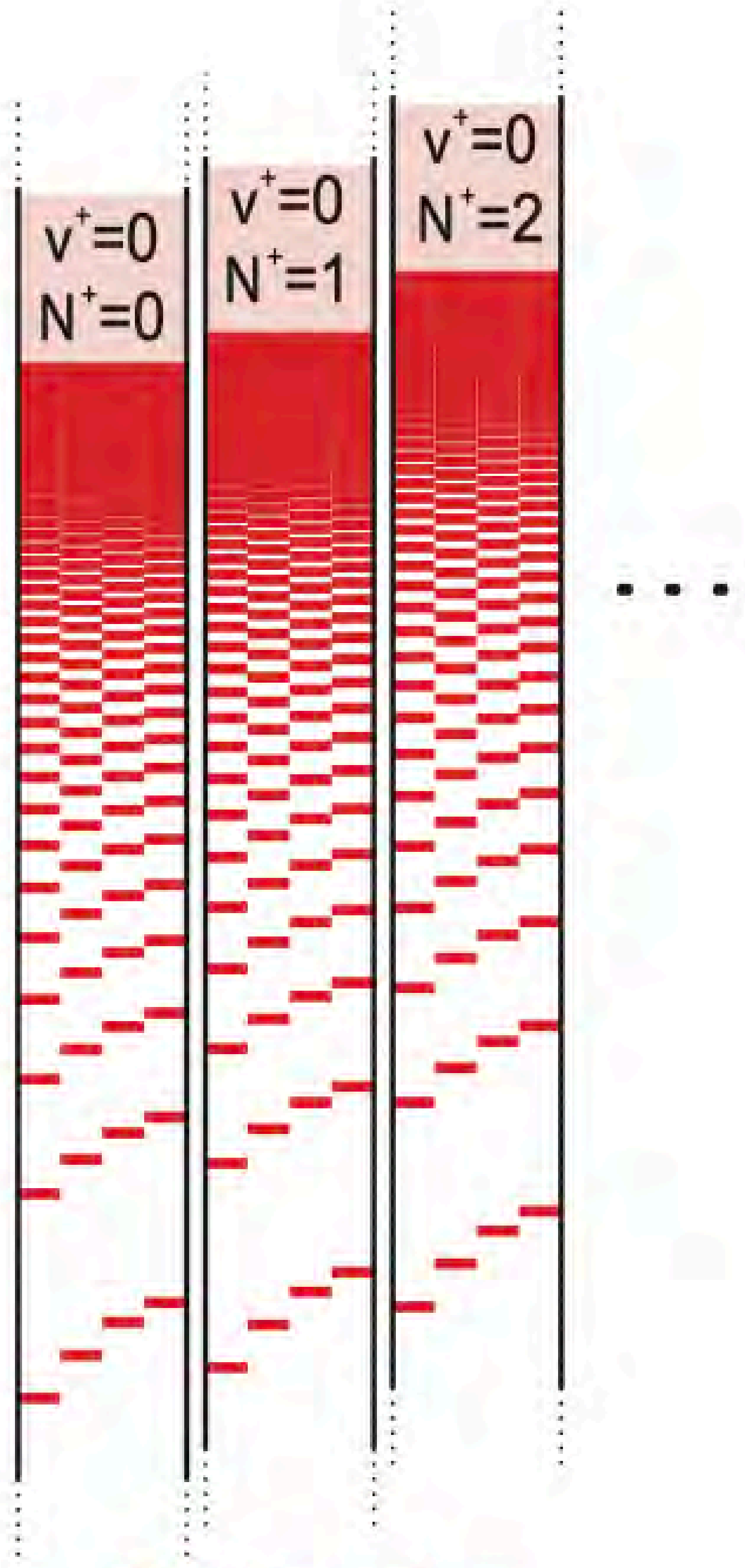




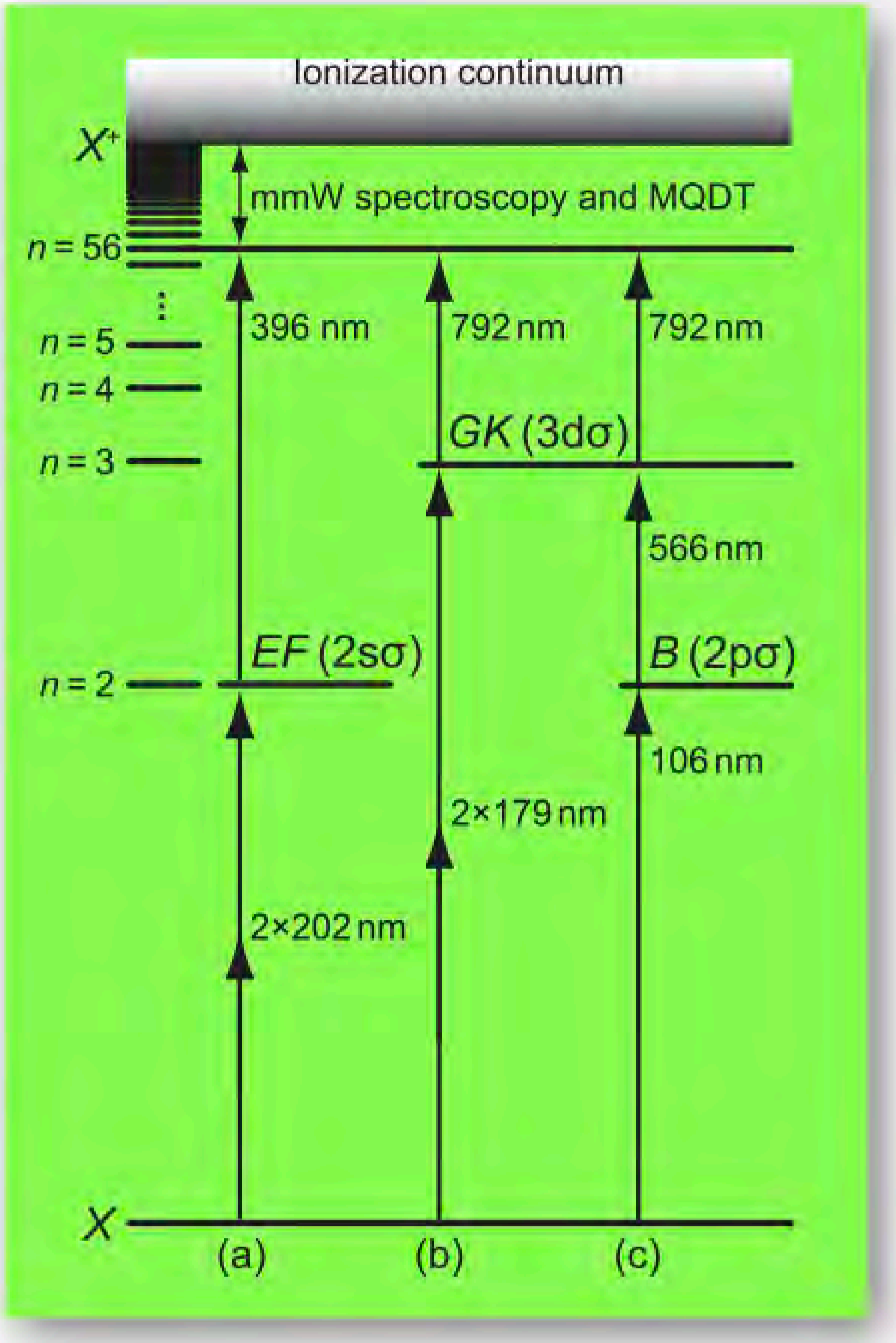
➡ **Can we measure a sharp feature, instead of an onset of dissociation?**

Measurement of the ionization energy





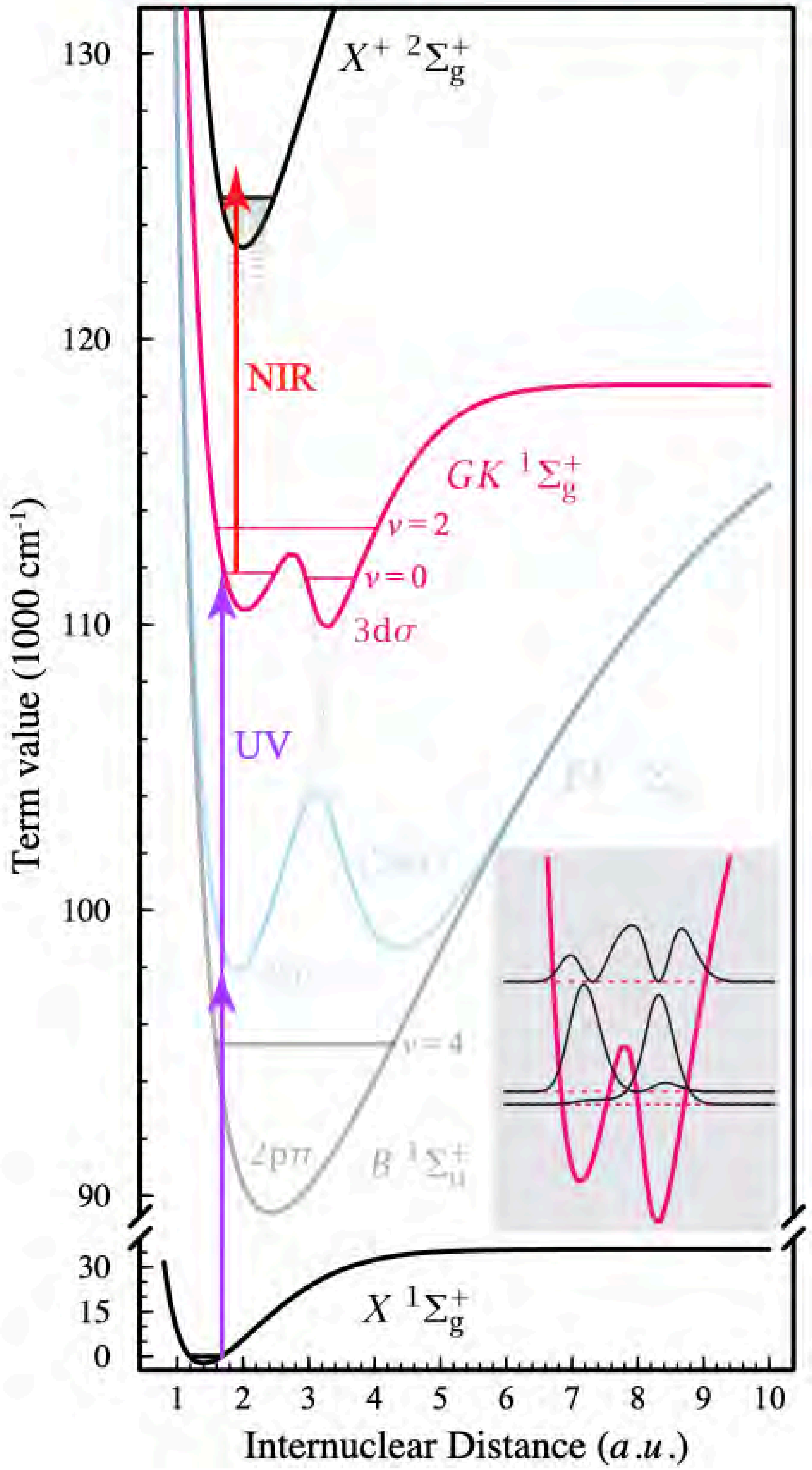
Molecules

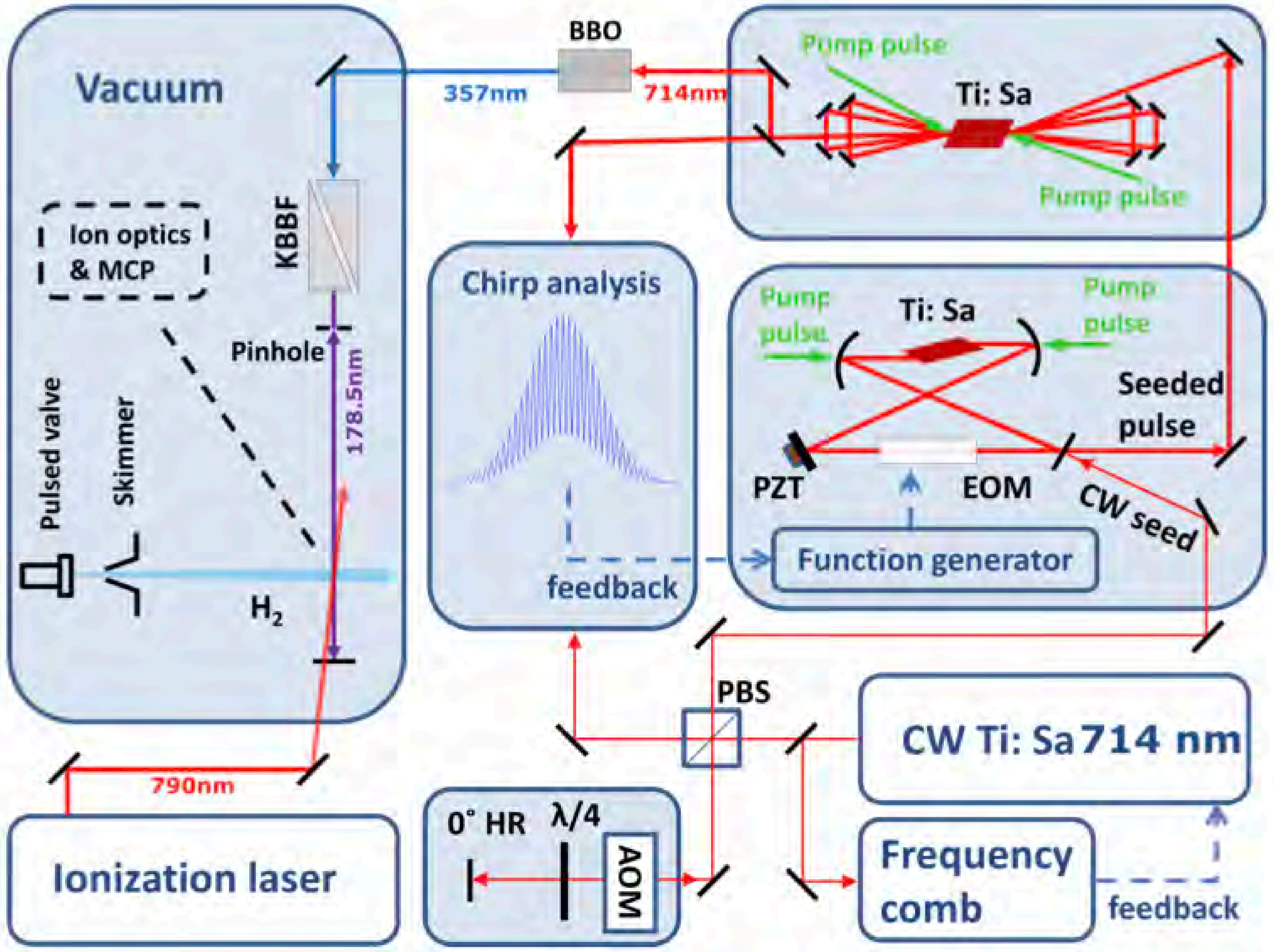


➡ **Measure transitions from gerade intermediate states to long-living Rydberg states**

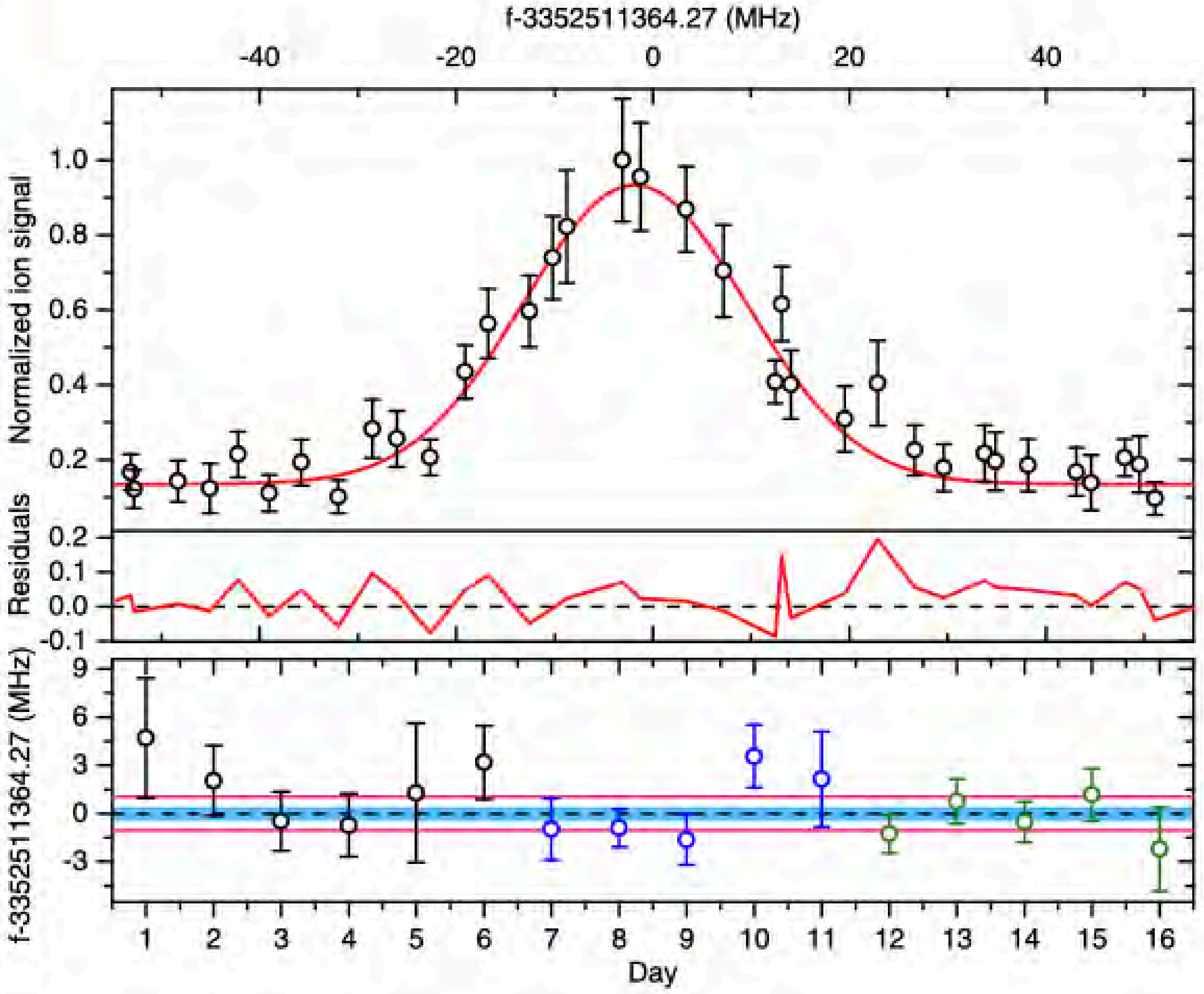
➡ **Extrapolate Rydberg series to the ionization limit using multichannel quantum-defect theory (MQDT)**

VU: two-photon deep-UV spectroscopy for X-GK interval





J. Hussels, C. Cheng, E. J. Salumbides, and W. Ubachs, Opt. Lett. 45, 5909-5912 (2020)



Dominating systematic uncertainties:

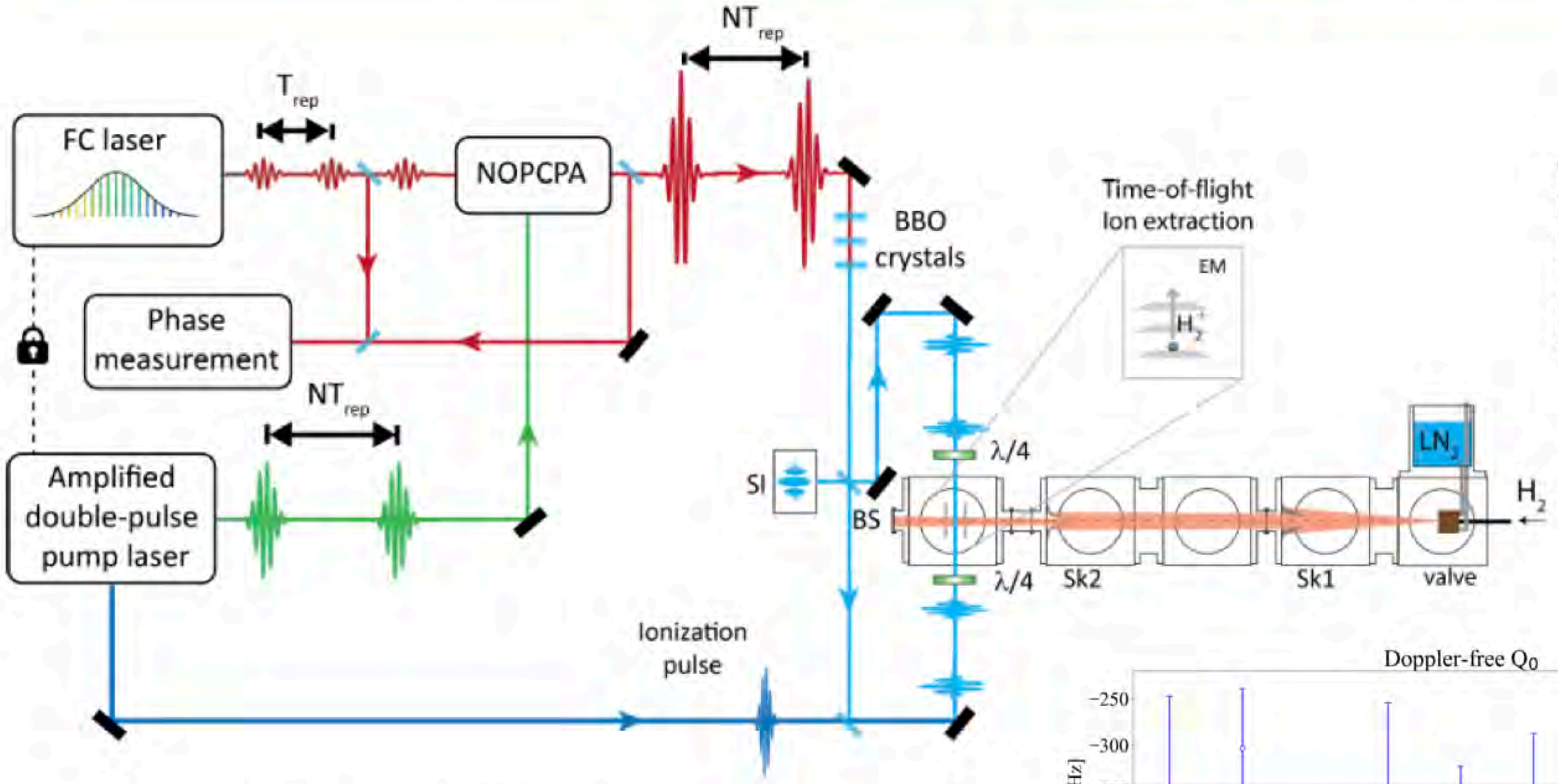
i) Residual first-order Doppler shift

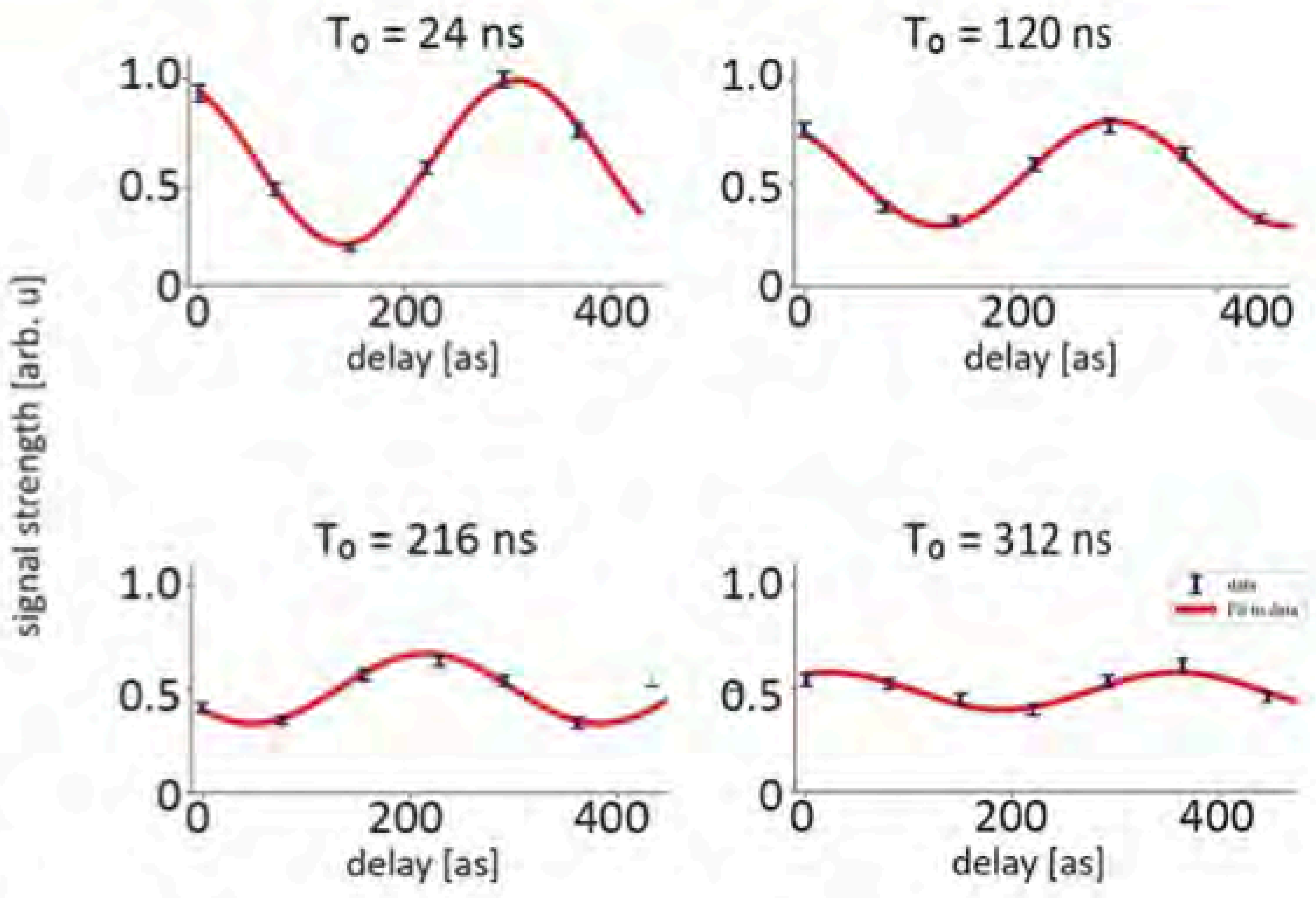
ii) ac-Stark effect from the two lasers

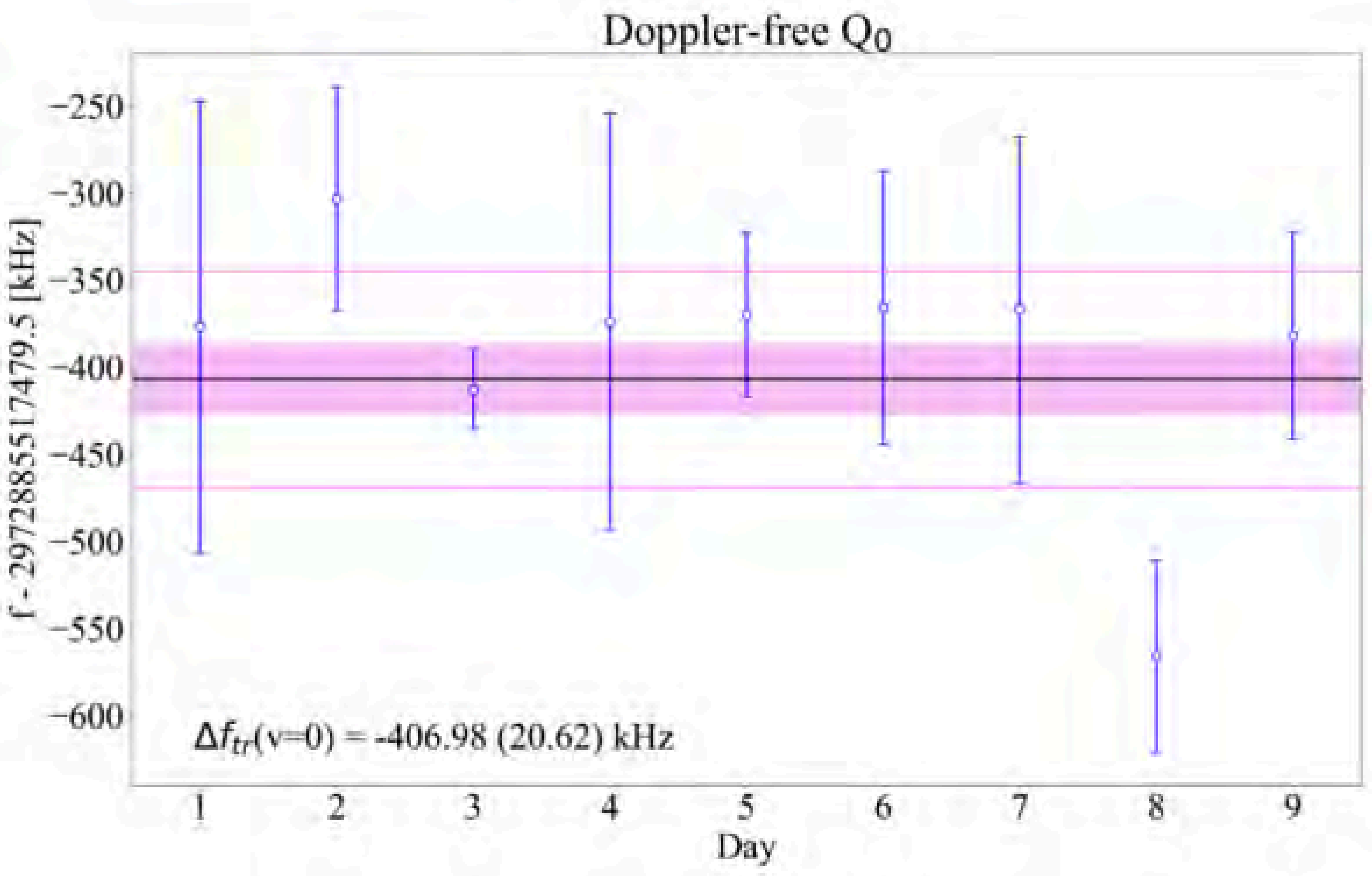
Work by:

J. Hussels, C. Cheng, E. Salumbides, W. Ubachs

VU: Ramsey-comb spectroscopy for X-EF interval







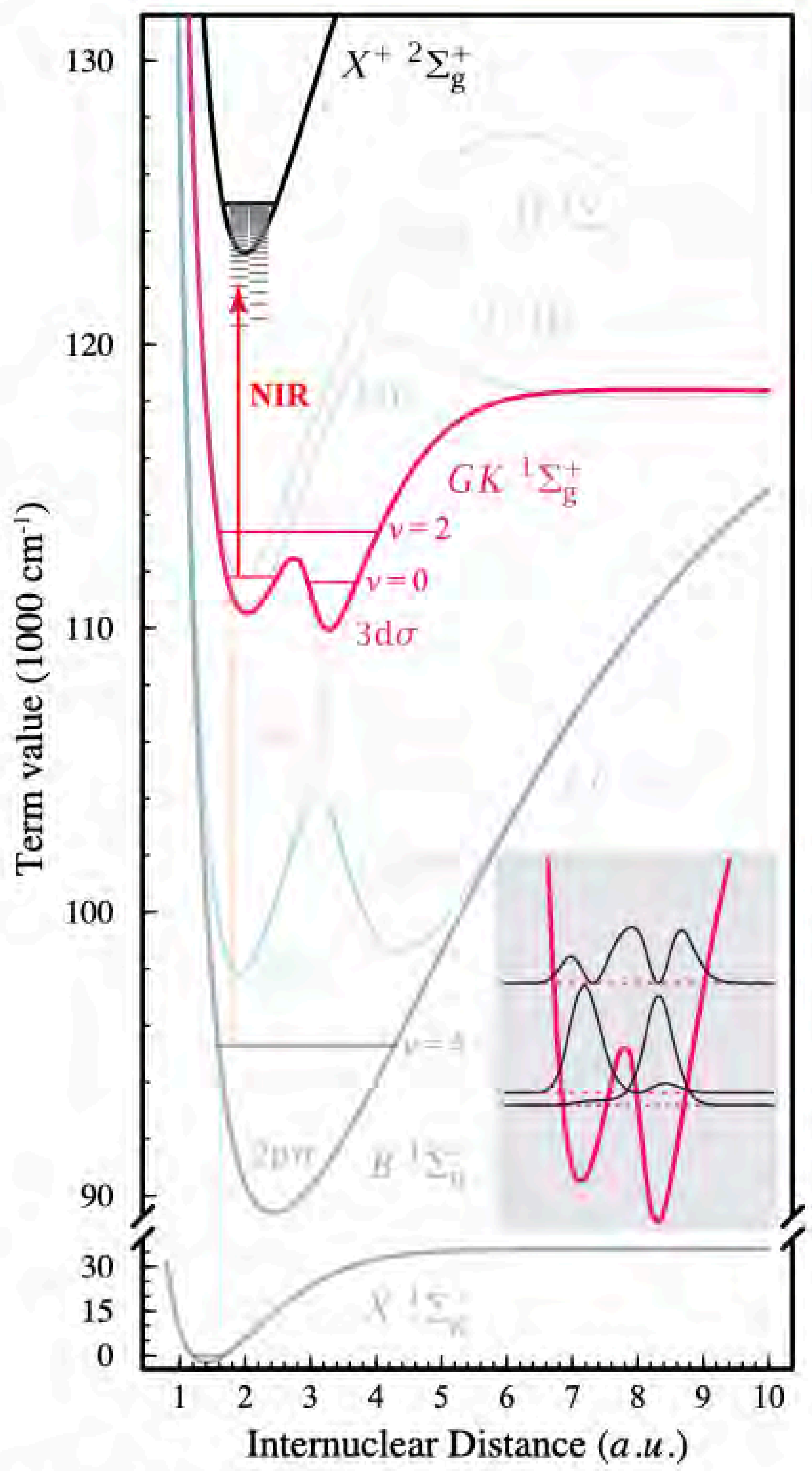
Dominating systematic uncertainties:

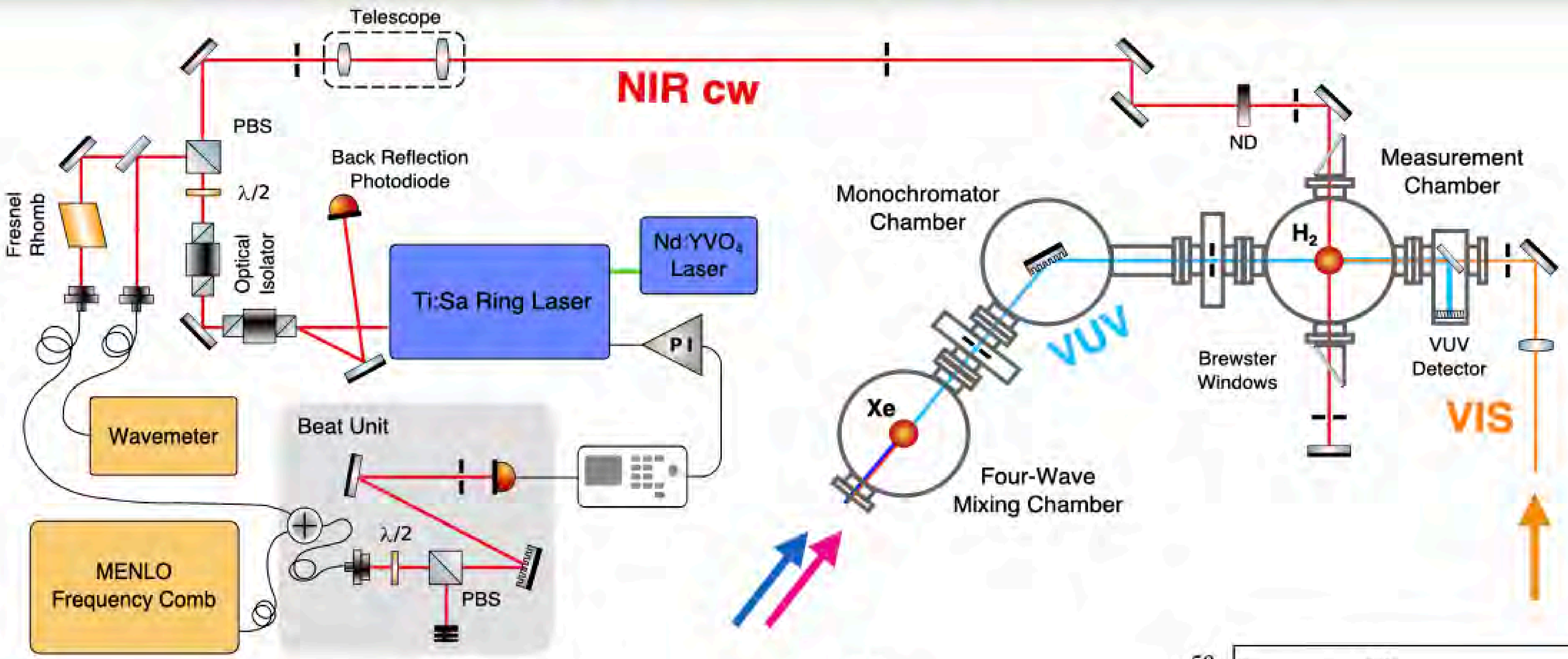
i) ac-Stark effect

Work by:

Ch. Roth, A. Martínez de Velasco, K. Eikema

ETH: Rydberg spectroscopy

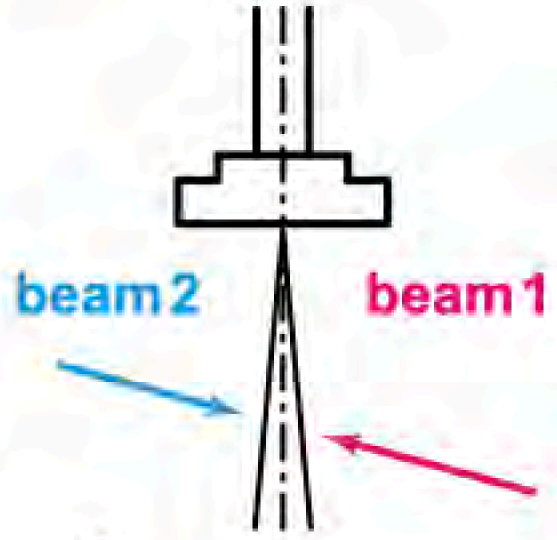


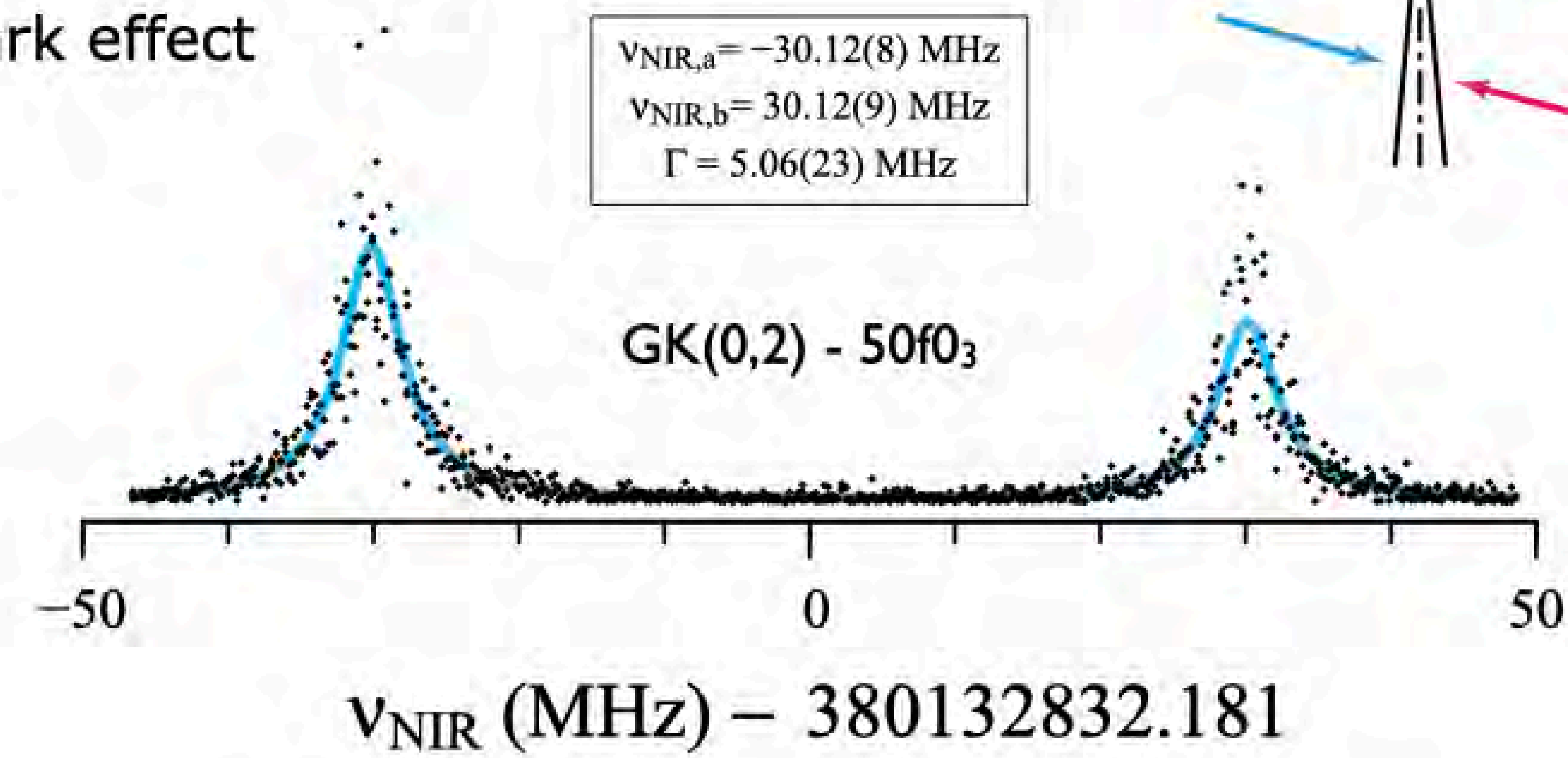


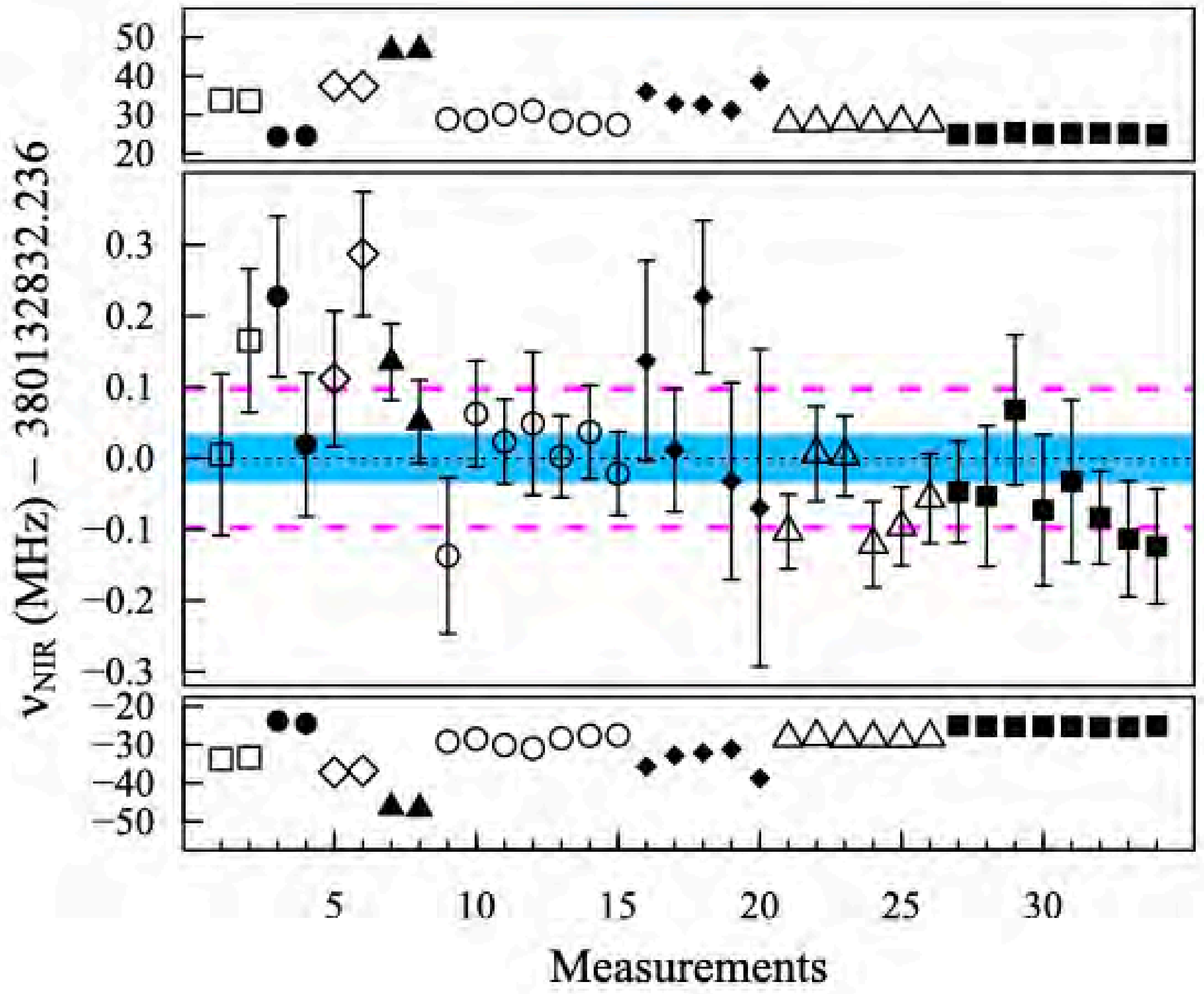
Dominating uncertainties:

i) Residual first-order Doppler shift

ii) dc-Stark effect







Work by:

MB, N. Hölsch, F. Merkt

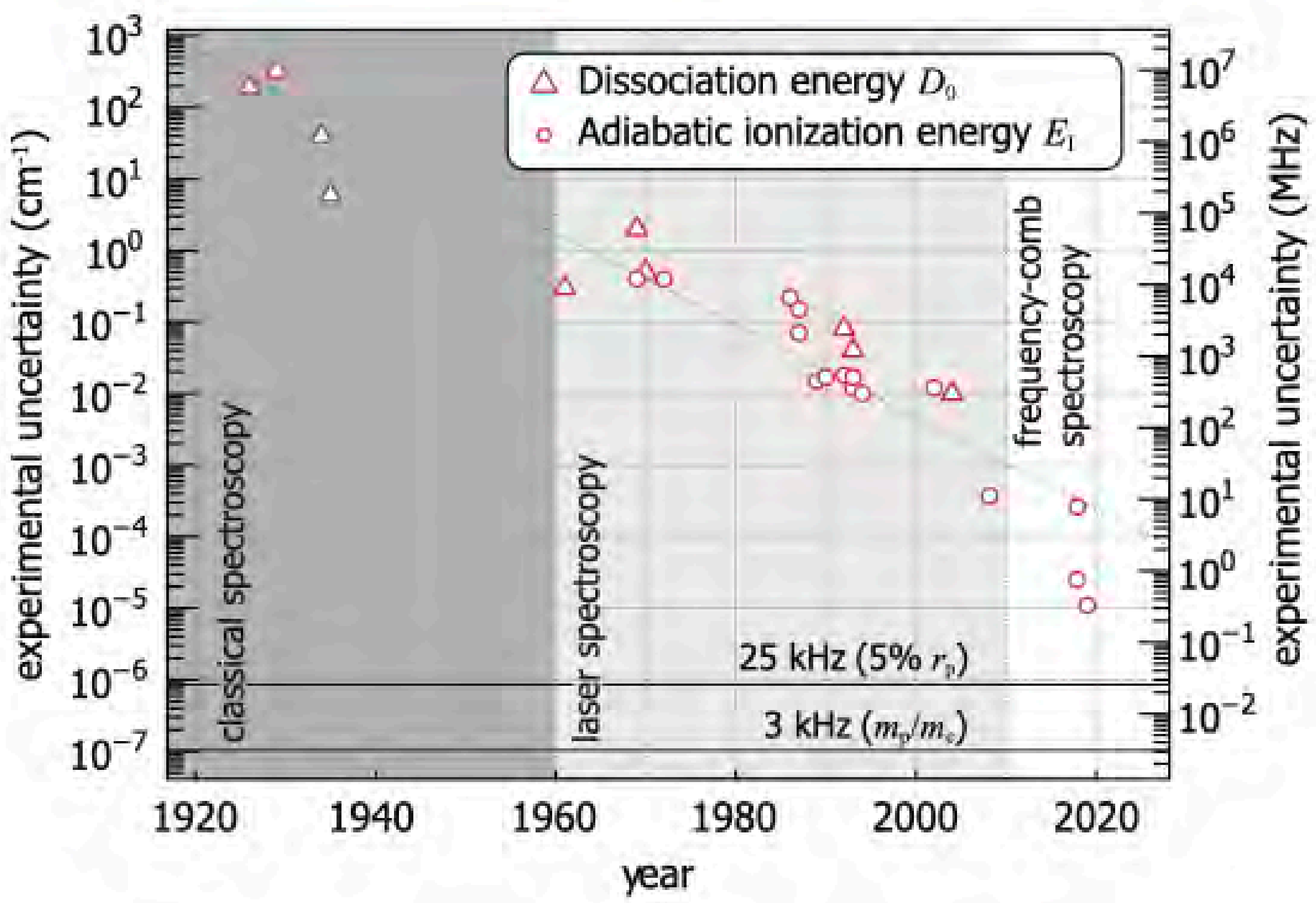
Measured ionization and dissociation energies

Values in cm^{-1}

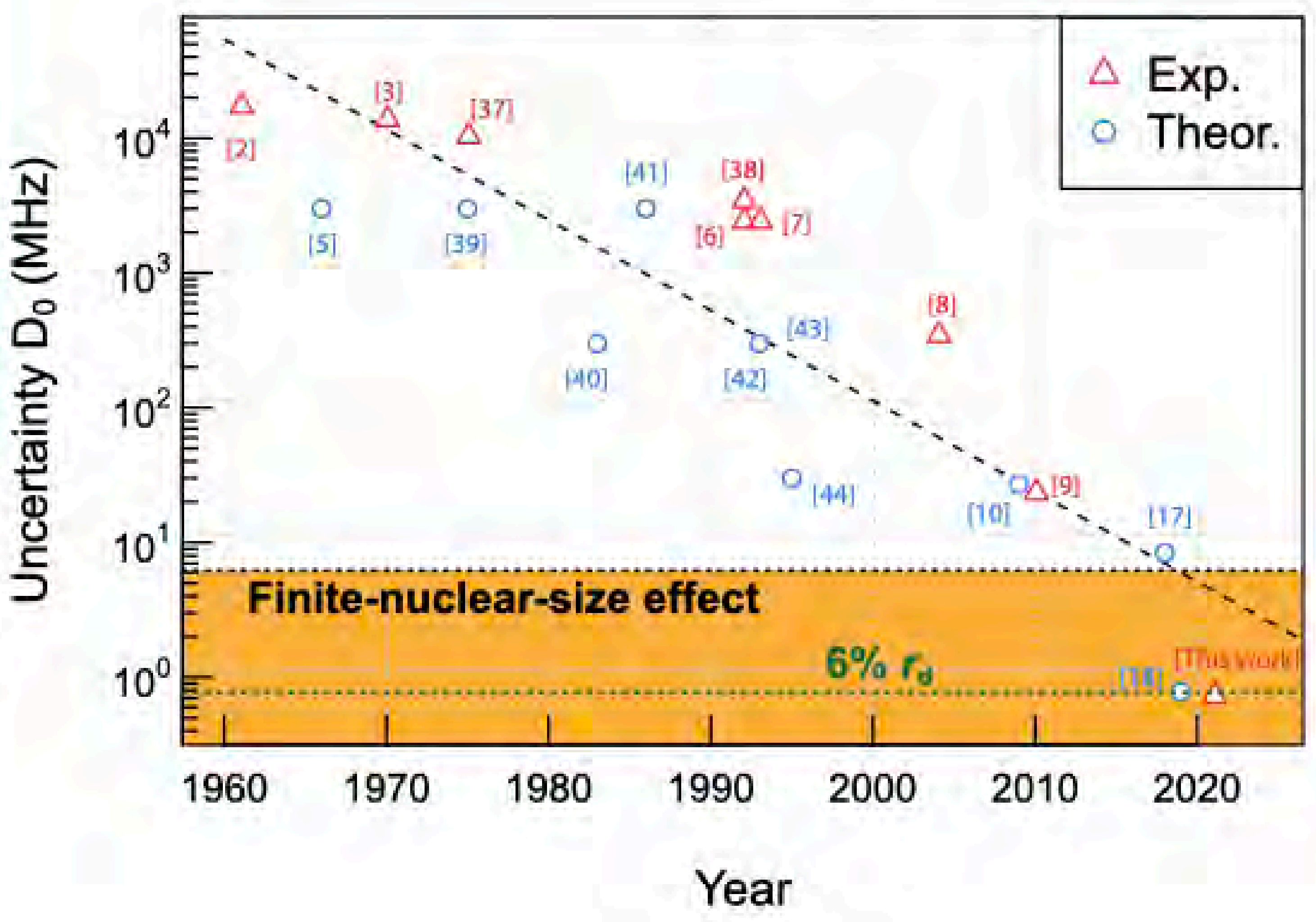
	H_2			D_2	HD
	para (N = 0)	ortho (N = 1)		ortho (N = 0)	N = 0
X-GK/EF	111 827.741 986(21) 630 kHz	111 686.632 836(22) 660 kHz	99 109.731 204 9(24) 73 kHz	112 099.087 712(15) 450 kHz	400 kHz
GK/EF-Rydberg	12 723.757 461(23) 700 kHz	12 635.724 114(12) 360 KHz	25 209.997 785(10) 300 kHz	12 979.344 802(20) 600 kHz	…
Rydberg extrapolation		150 kHz	150 kHz		…
IP	124 417.491 098(31) 940 kHz	124 357.238 062(25) 750 kHz (2E-10)	124 357.238 003(11) 340 kHz (9E-11)	124 745.393 739(26) 780 kHz	…
D_0	36 118.069 605(31) 940 kHz	35 999.582 894(25) 750 kHz (7E-10)	35 999.582 834(11) 340 kHz (3E-10)	36 748.362 282(26) 780 kHz	[36 405.783 66(36)]
D_0 (theory)	36 118.069 632(26) 780 kHz		35 999.582 820(26) 780 kHz	36 748.362 342(26) 780 kHz	36 405.782 477(26) 780 kHz

H_2 ortho: PRL **121**, 013001 (2018), PRL **122**, 103002 (2019),
H_2 para: PRL **123**, 163002 (2019),
D_2 ortho: PRA **105**, 022820 (2022)
Theory: PRL **122**, 103003 (2019), PRA **100**, 020503(R) (2019),

ortho H_2



ortho D_2



New results for X-EF in H_2 (para) and D_2 (ortho)

Contribution	Value [kHz]	Uncertainty [kHz]
Transition frequency, 1st and 2nd order Doppler-	2 972 885 517 058	21
AC-Stark shift correction	28	23
Phase measurement setup correction	-8	1
Zeeman shift correction	0	1
DC-Stark shift correction	1	1
Clock	0	1
Total	2 972 885 517 079	31

H_2

Contribution	Value [kHz]	Uncertainty [kHz]
Transition frequency, 1st and 2nd order Doppler-	2 981 779 227 576	13
AC-Stark shift correction	9	14
Phase measurement setup correction	-6	2
Zeeman shift correction	0	1
DC-Stark shift correction	0	1
Clock	0	1
Total	2 981 779 227 579	19

D_2

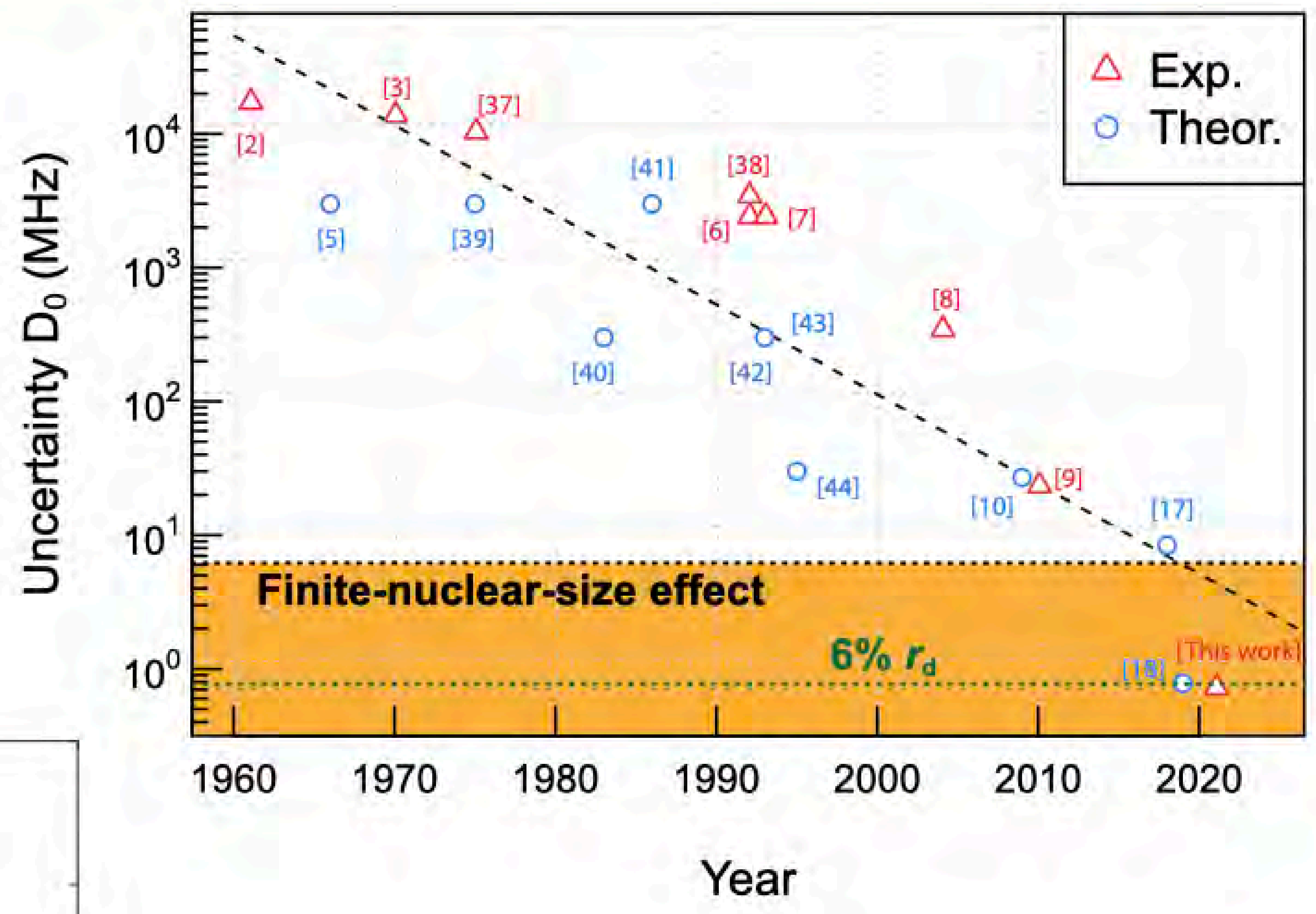
Work by:

Ch. Roth, A. Martínez de Velasco, K. Eikema

Effect of charge radius: D_0 vs X-EF vs IP

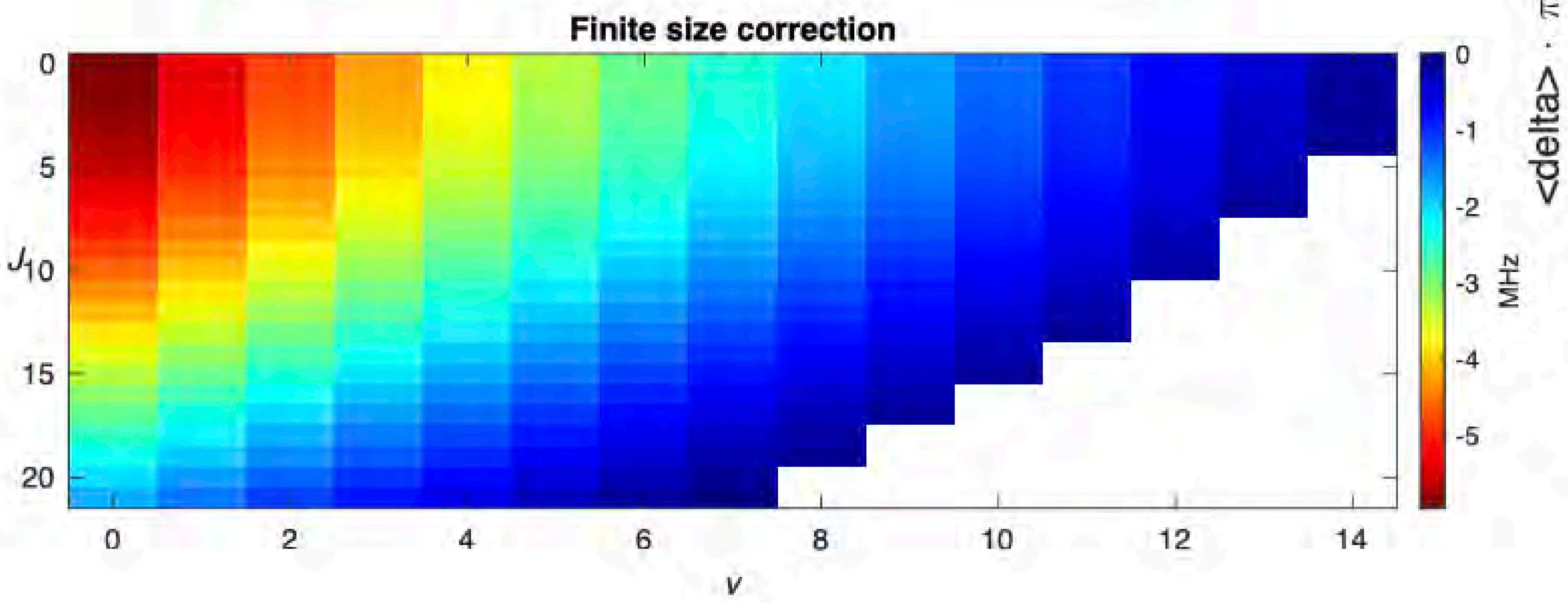
Contribution	H_2	D_2	HD
$E^{(2)}$	36 118.797 746 10(3)	36 749.090 990 00(1)	36 406.510 890 07(1)
$E^{(4)}$	−0.531 215 6(5)	−0.528 206 05(9)	−0.529 887 5(2)
$E^{(5)}$	−0.194 910 43(15)	−0.198 256(3)	−0.196 441(4)
$E^{(6)}$	−0.002 067(6)	−0.002 096(6)	−0.002 080(6)
$E^{(6)}_{\mathrm{sec}}$	0.000 009 2	0.000 009 4	0.000 009 3
$E^{(7)}$	0.000 101(25)	0.000 103(25)	0.000 102(25)
$E^{(4)}_{\mathrm{FS}}$	−0.000 031	−0.000 202	−0.000 116
Total	36 118.069 632(26)	36 748.362 342(26)	36 405.782 477(26)

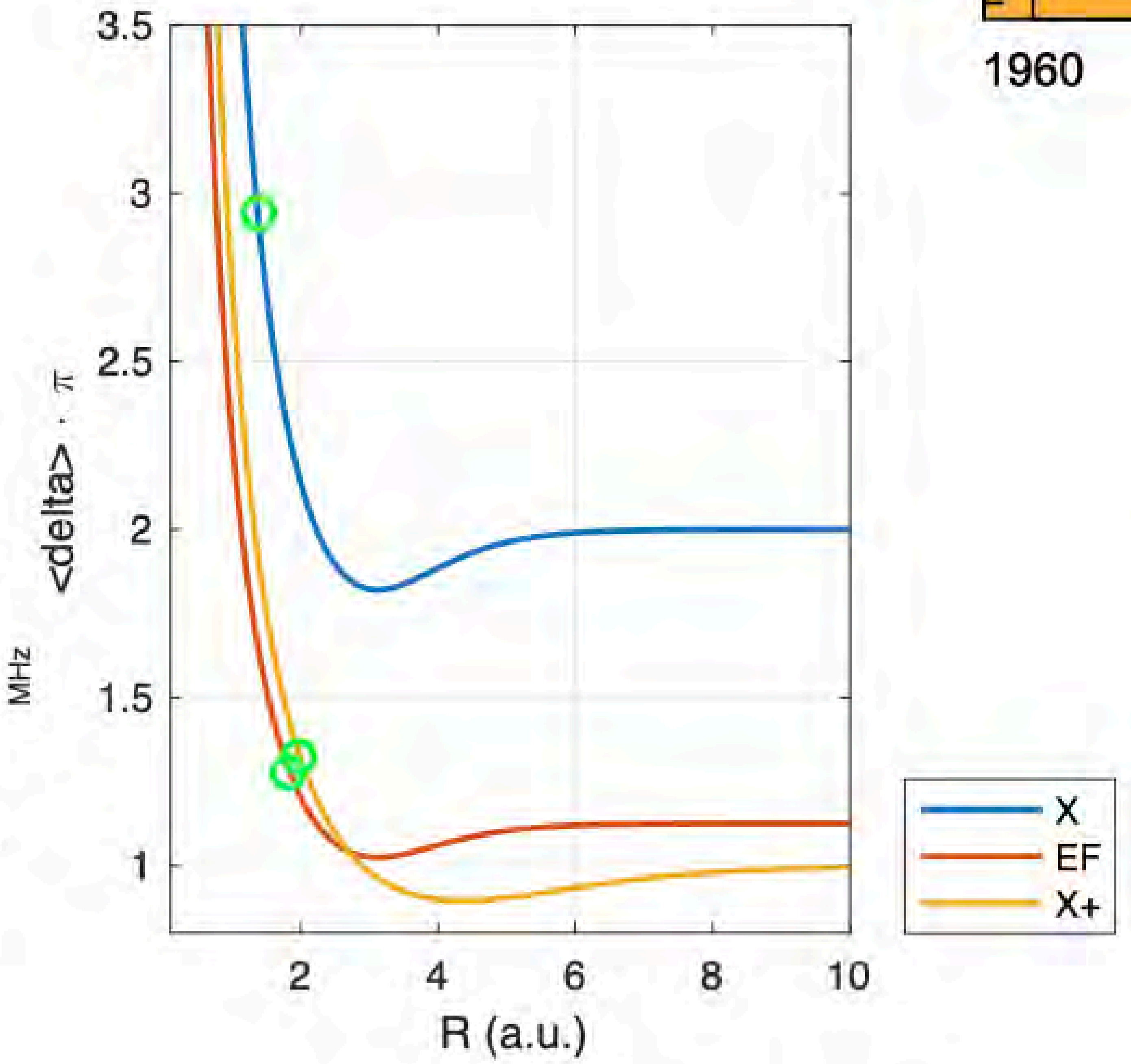
PRL **122**, 103003 (2019), PRA **100**, 020503(R) (2019)



$$\mathcal{E}^{(4)}_{\mathrm{FS}}(R) = \frac{2\pi}{3}\,\langle\phi_{\mathrm{el}}|\sum_{a,X}\delta^3(r_{aX})|\phi_{\mathrm{el}}\rangle_{\mathrm{el}}\,\frac{(r^2_{C,A}+r^2_{C,B})}{2\lambda\!\!\!^{-}{}^2}$$

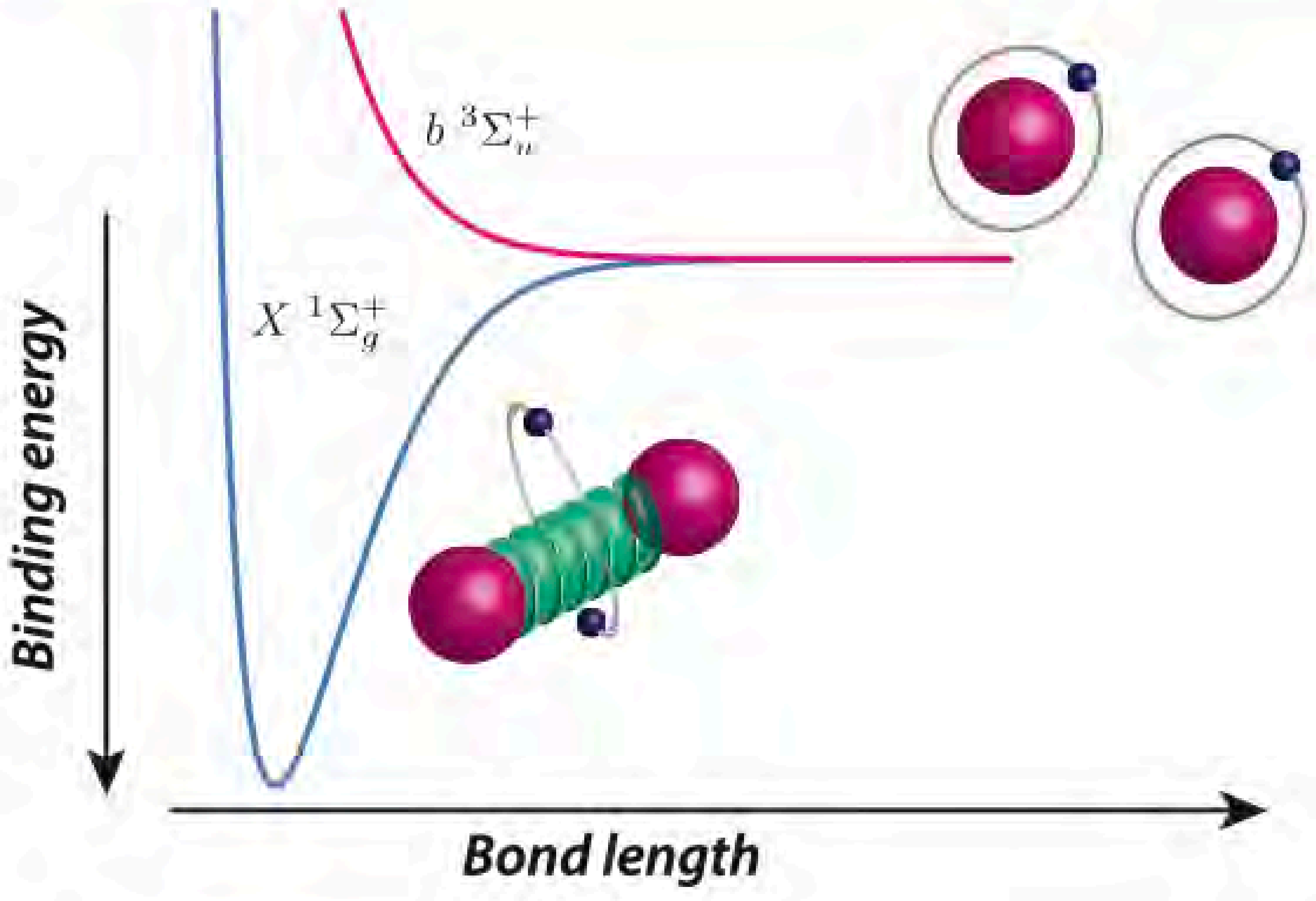
$$r_d = 2.12799(74)\ \mathrm{fm}$$





- ➡ Effect of FS in D_2 D_0: 6.056 MHz
- ➡ FS uncertainty on D_2 D_0: 4 kHz
- ➡ FS uncertainty on D_2 IP: 11 kHz
- ➡ FS uncertainty on D_2 X-EF: 12 kHz

Collisions of (ground state) Hydrogen atoms



- Frequency shift (H maser, 1s-2s spectroscopy, …)

VOLUME 81, NUMBER 18 PHYSICAL REVIEW LETTERS 2 NOVEMBER 1998

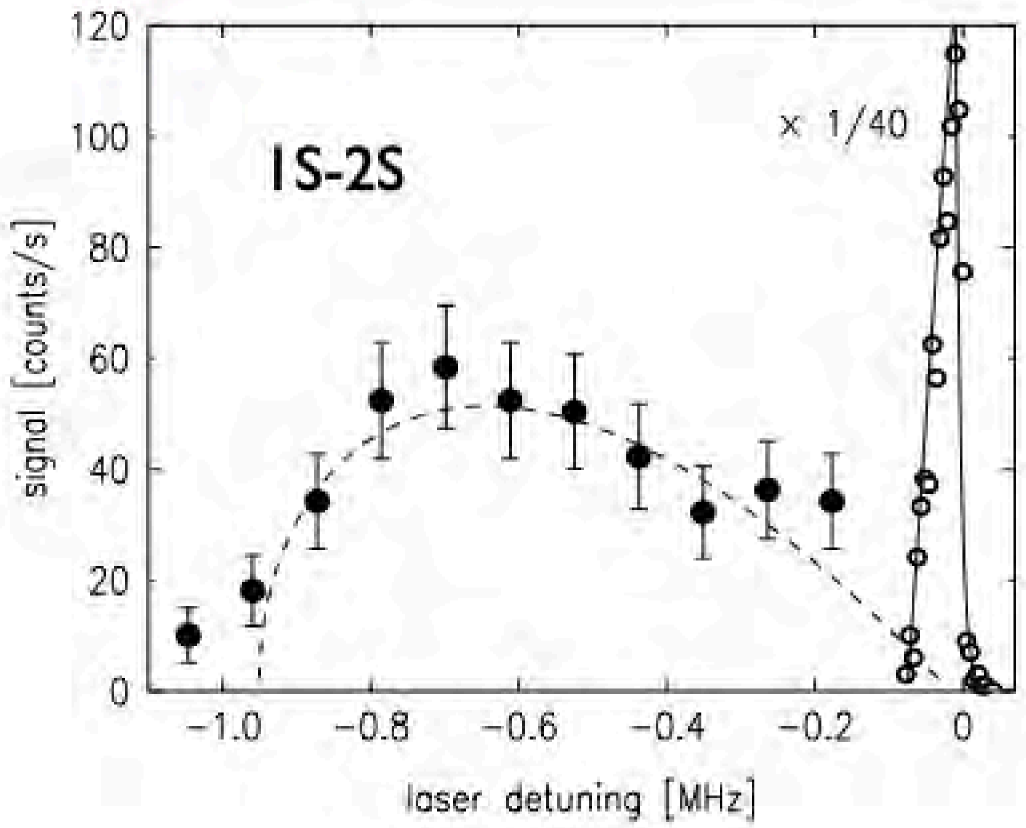
Bose-Einstein Condensation of Atomic Hydrogen

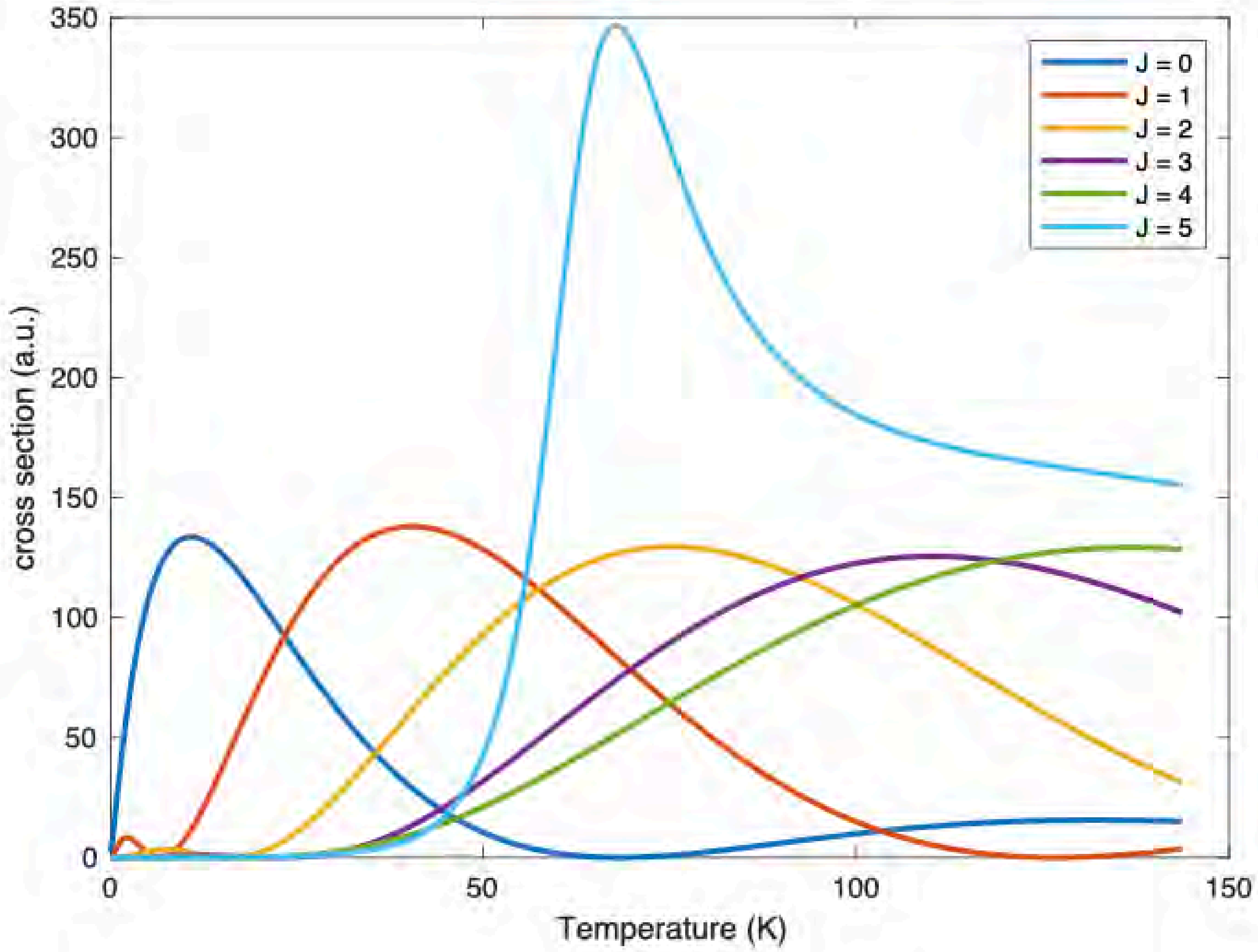
Dale G. Fried, Thomas C. Killian, Lorenz Willmann, David Landhuis, Stephen C. Moss, Daniel Kleppner, and Thomas J. Greytak

Department of Physics and Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 11 September 1998)

- BEC





- Three-body recombination

THE JOURNAL OF CHEMICAL PHYSICS VOLUME 50, NUMBER 12 15 JUNE 1969

Resonance Theory of Termolecular Recombination Kinetics*: $H+H+M \rightarrow H_2+M$

ROBERT E. ROBERTS,† R. B. BERNSTEIN, AND C. F. CURTISS

Theoretical Chemistry Institute and Chemistry Department, University of Wisconsin, Madison, Wisconsin 53706

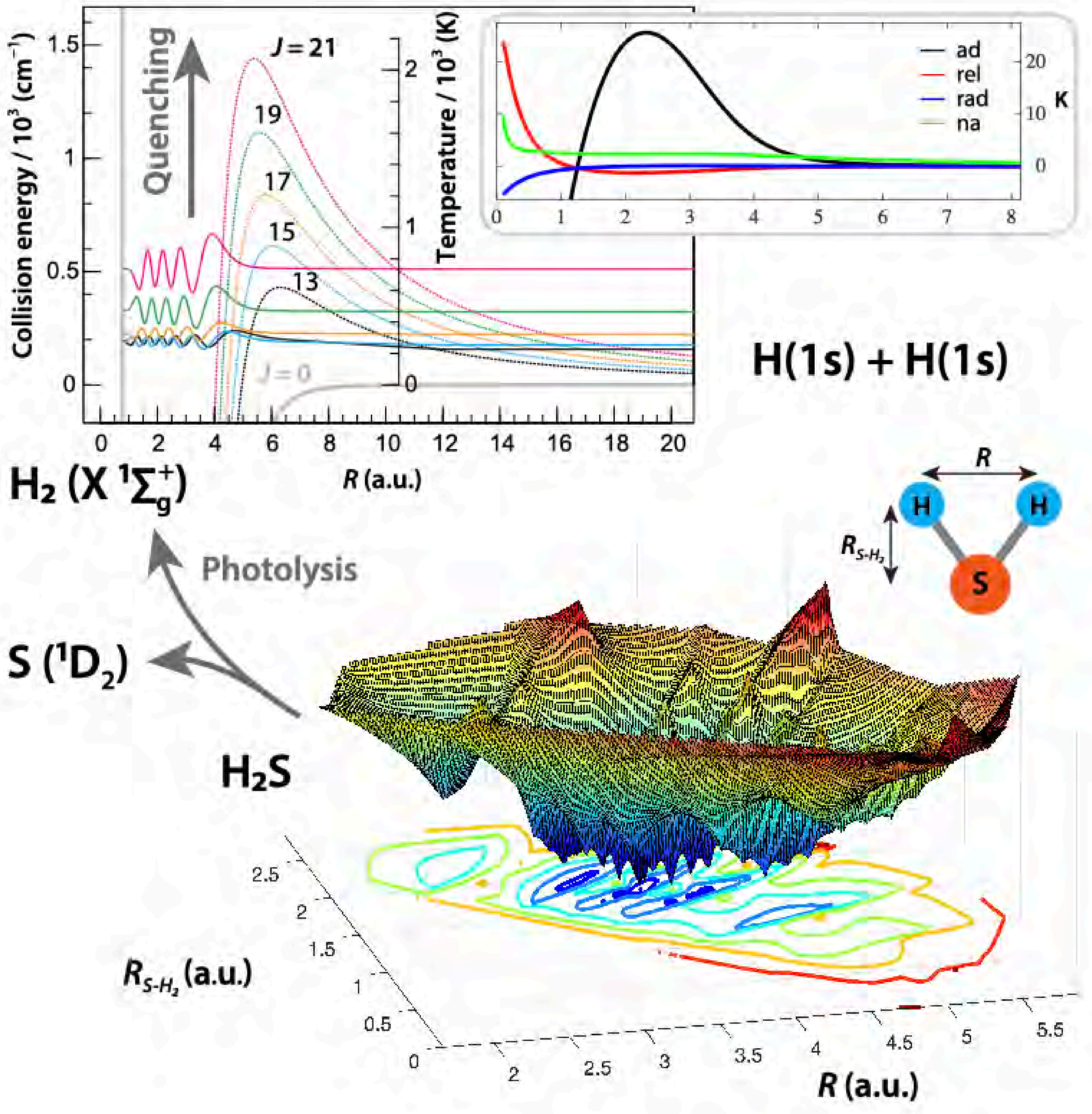
(Received 14 February 1969)

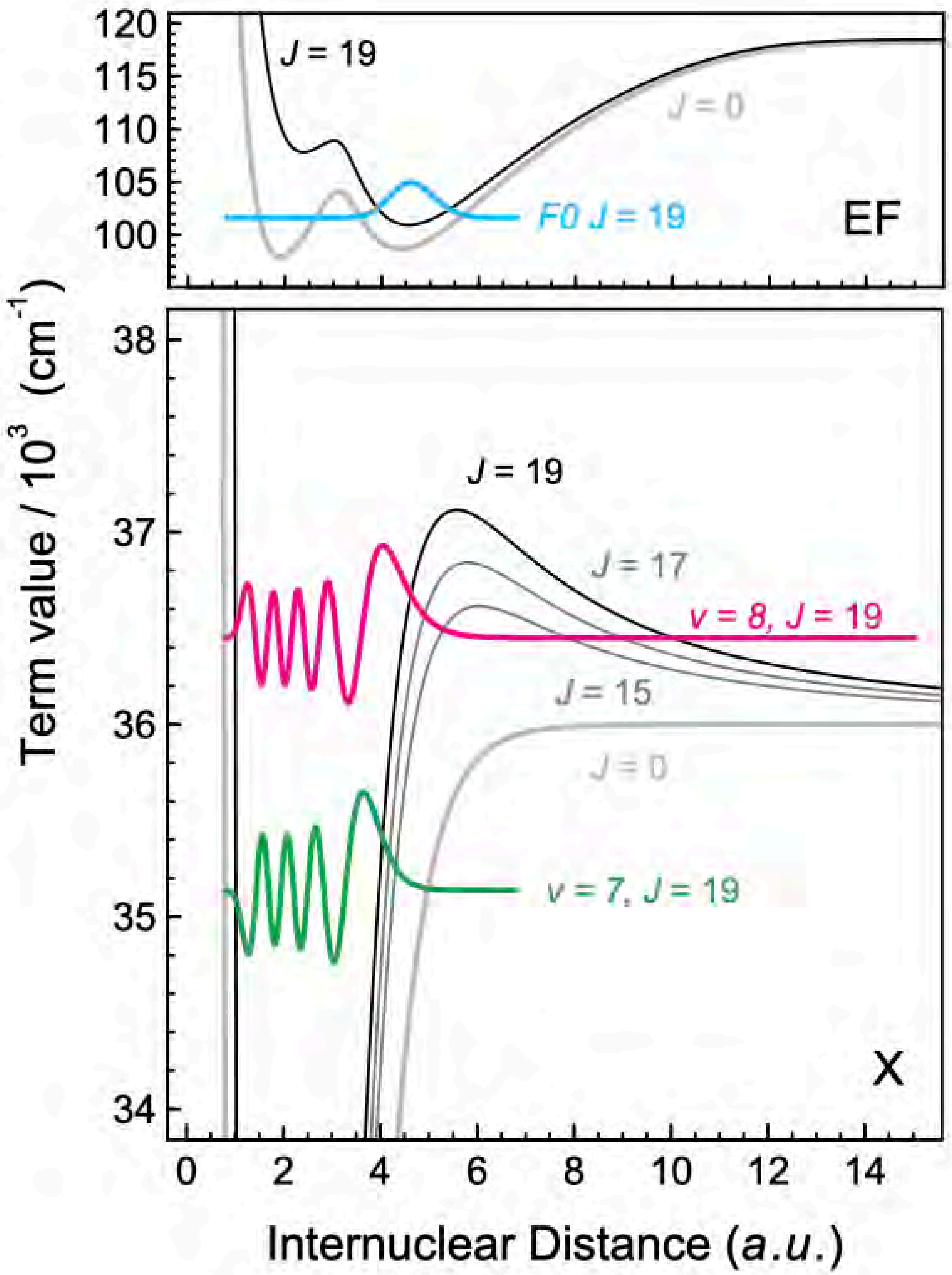
$$X+X \xrightarrow{k_f^i} X_2^i$$

$$X_2^i \xrightarrow{k_d^i} X+X,$$

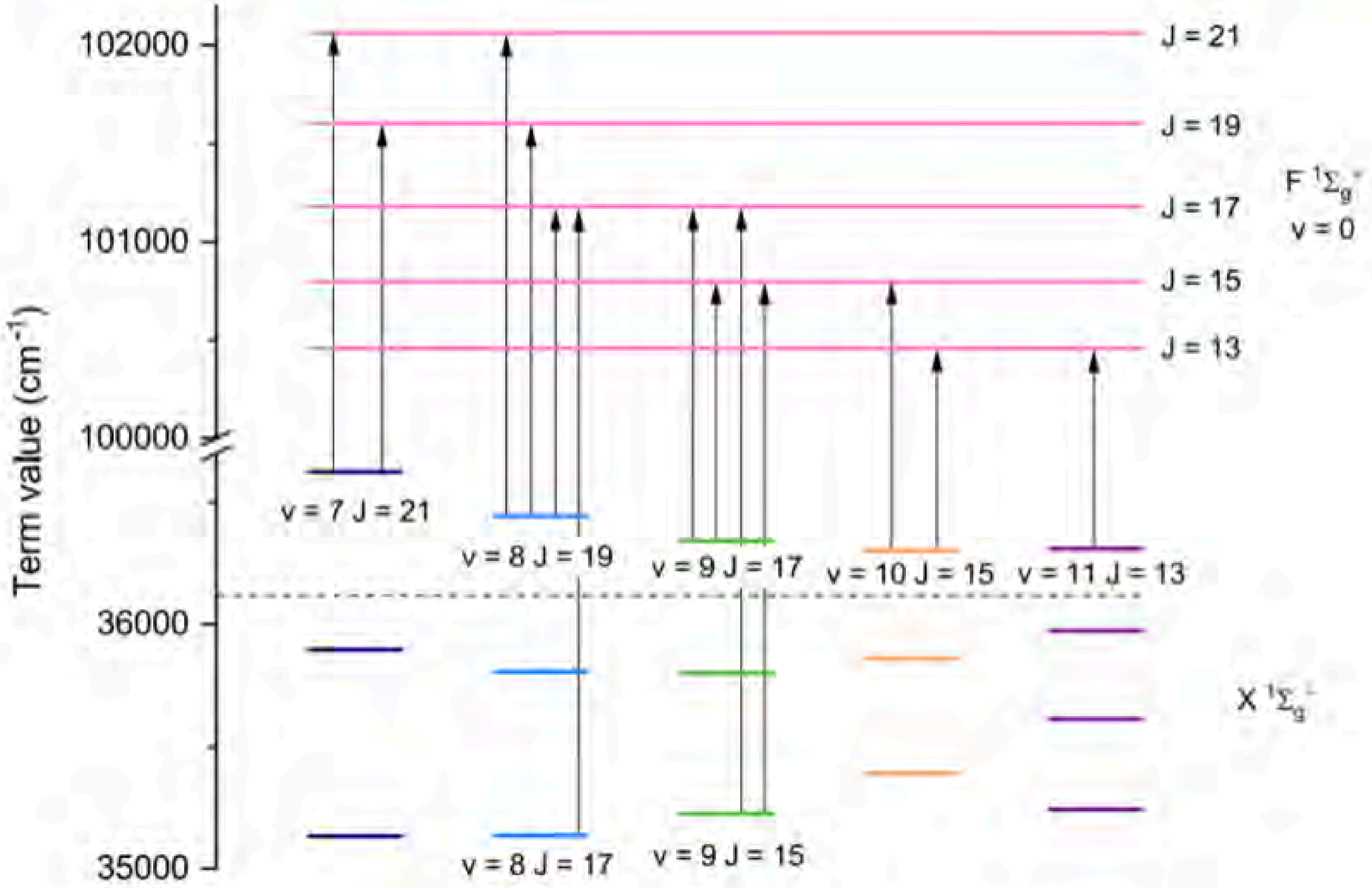
$$X_2^i+M \xrightarrow{k_f^{in}} X_2^n+M$$

Production of highly rovibrationally excited states





Dissociation energies of shape resonances

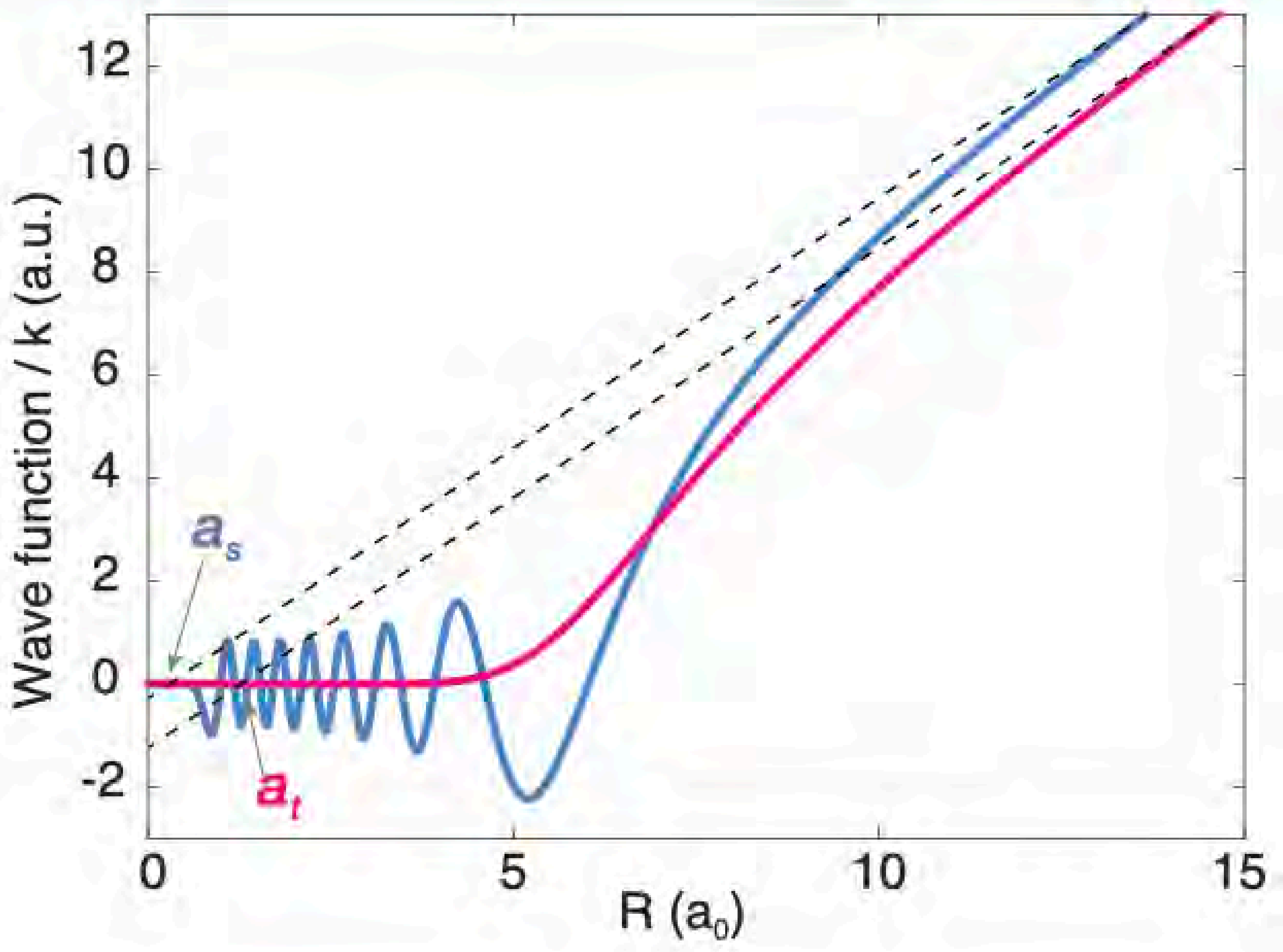


Contribution / (v, J)	(11,13)	(10,15)
$m\alpha^2$	−192.2683(29)	−186.1598(36)
adiabatic	3.1895	4.2857
nonadiabatic	2.6847	3.2666
$m\alpha^4$	−0.2254	−0.2945
BO	−0.2261	−0.2952
nonadiabatic	0.0007	0.0007
$m\alpha^5$	−0.0008	0.0002
$m\alpha^6$	−0.0001	−0.0001
Total	−192.4961(29)	−186.4555(36)
Γ_{FWHM} (kHz)	1×10^5	73.6
Lifetime (s)	1.59×10^{-9}	2.16×10^{-6}

(v, J)	$T_{\mathrm{rel}}^{\mathrm{exp}}$	$T_{\mathrm{rel}}^{\mathrm{calc}}$	ΔT
(7,21)*	319.4772 (28)	319.4768 (9)	0.0004 (29)
(8,17)	−1168.9200 (25)	−1168.9200 (12)	0.0000 (28)
(8,19)*	140.9748 (25)	140.9750 (7)	−0.0002 (26)
(9,15)	−1078.6659 (21)	−1078.6671 (11)	0.0012 (24)
(9,17)*	38.4872 (30)	38.4869 (4)	0.0003 (30)
(10,15)*	0	0	0
(11,13)*	6.041 (28)	6.0404 (6)	0.001 (28)

➡ Determination of ro-vibrational intervals

Determination of the scattering length



$$a = \begin{cases} 0.5699^{700}_{697}\ a_0 & \text{BO} \\ 0.4160^{61}_{58}\ a_0 & \text{AD} \\ 0.2572^{76}_{68}\ a_0 & \text{NA} \\ 0.2735^{39}_{31}\ a_0 & \text{NA}, m\alpha^4, m\alpha^5, m\alpha^6. \end{cases}$$

Using the atomic mass: $\mathbf{a_{NA} = 0.2651\ a_0}$ 3% difference

A calculation for the triplet PES gives: $\mathbf{a_t = 1.331\ a_0}$
using Wolniewicz, *PRA* **61**, 042705, 2000.

Calculation	Mass (m_e)	a
atomic mass [5]	1837.1527	0.3006
nuclear mass [5]	1836.1527	0.4503
Bunker et al. [7]	1837.0777	0.3121
new mass	1837.0318	0.3191
Williams & Julienne [4]		0.3159
variable mass [8]		0.4042
close coupling [2]		0.564

4. C.J. Williams, P.S. Julienne, Phys. Rev. A **47**, 1524 (1995)
5. M.J. Jamieson, A. Dalgarno, J. Phys. B **31**, L219 (1998)
7. P.R. Bunker, R.E. Moss, Molec. Phys. **33**, 417 (1977); P.R. Bunker, C.J. McLarnon, R.E. Moss, Molec. Phys. **33**, 425 (1977)
8. M. Aubert-Frécon, G. Hadinger, S.Ya. Umanskii, J. Phys. B **27**, 4453 (1994)

PRL **98**, 043004 (2007) PHYSICAL REVIEW LETTERS week ending 26 JANUARY 2007

Cold Collision Frequency Shift in Two-Dimensional Atomic Hydrogen

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(Received 2 October 2006; published 24 January 2007)

We report a measurement of the cold collision frequency shift in atomic hydrogen gas adsorbed on the surface of superfluid ^{4}He at $T \leq 90$ mK. Using two-photon electron and nuclear magnetic resonance in 4.6 T field we separate the resonance line shifts due to the dipolar and exchange interactions, both proportional to surface density σ. We find the clock shift $\Delta\nu_c = -1.0(1) \times 10^{-7}$ Hz cm$^{-2} \times \sigma$, which is about 100 times smaller than the value predicted by the mean field theory and known scattering lengths in the three-dimensional case.

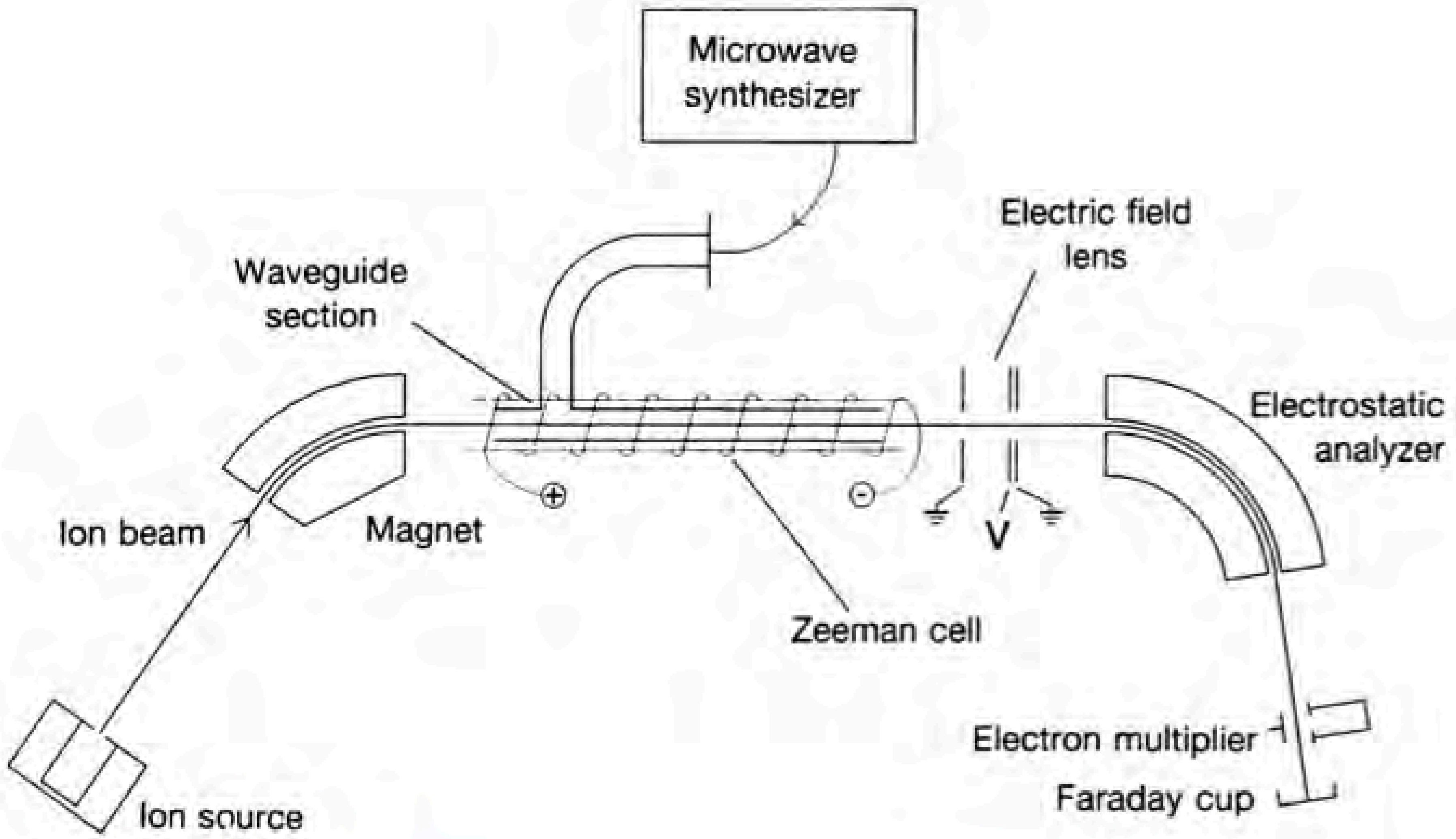
Weakly-bound states of hydrogen molecular ions

SCIENCE • VOL. 274 • 22 NOVEMBER 1996

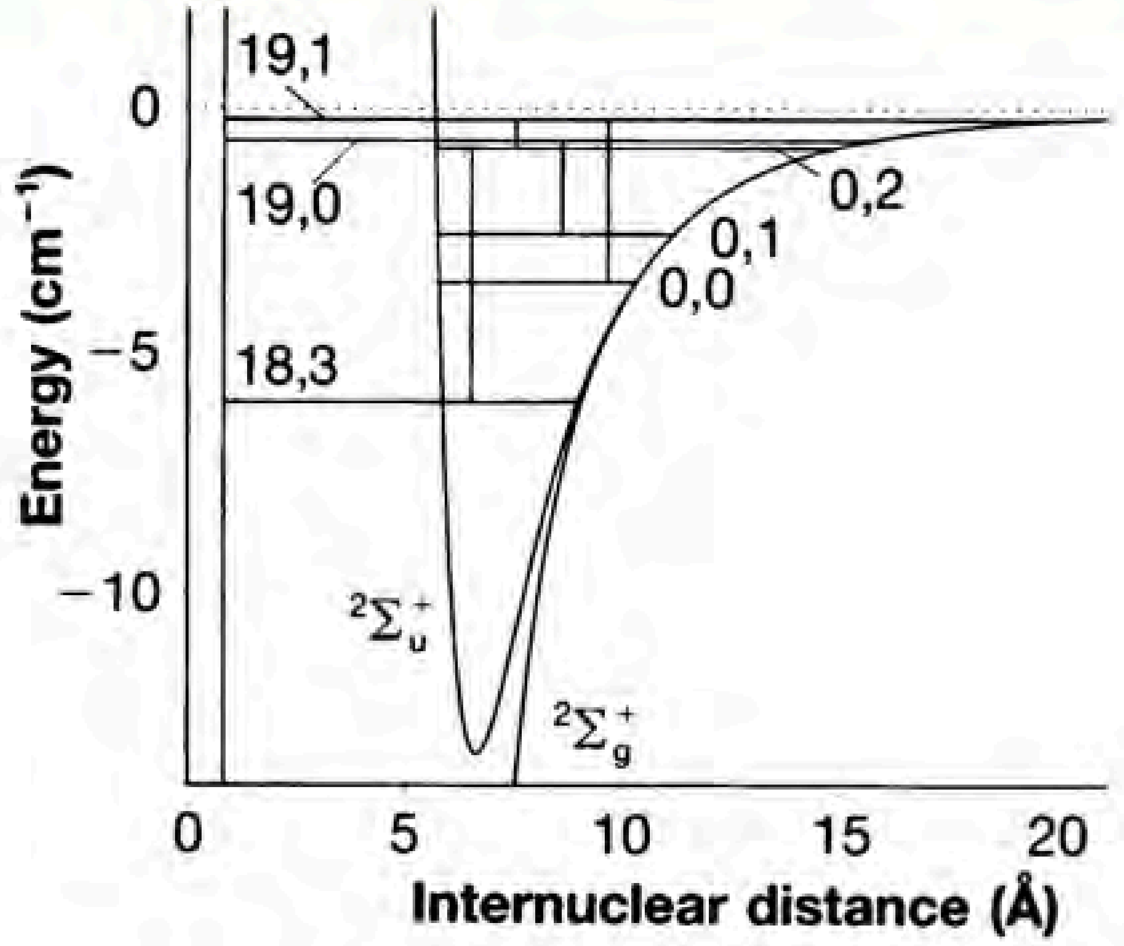
ARTICLE

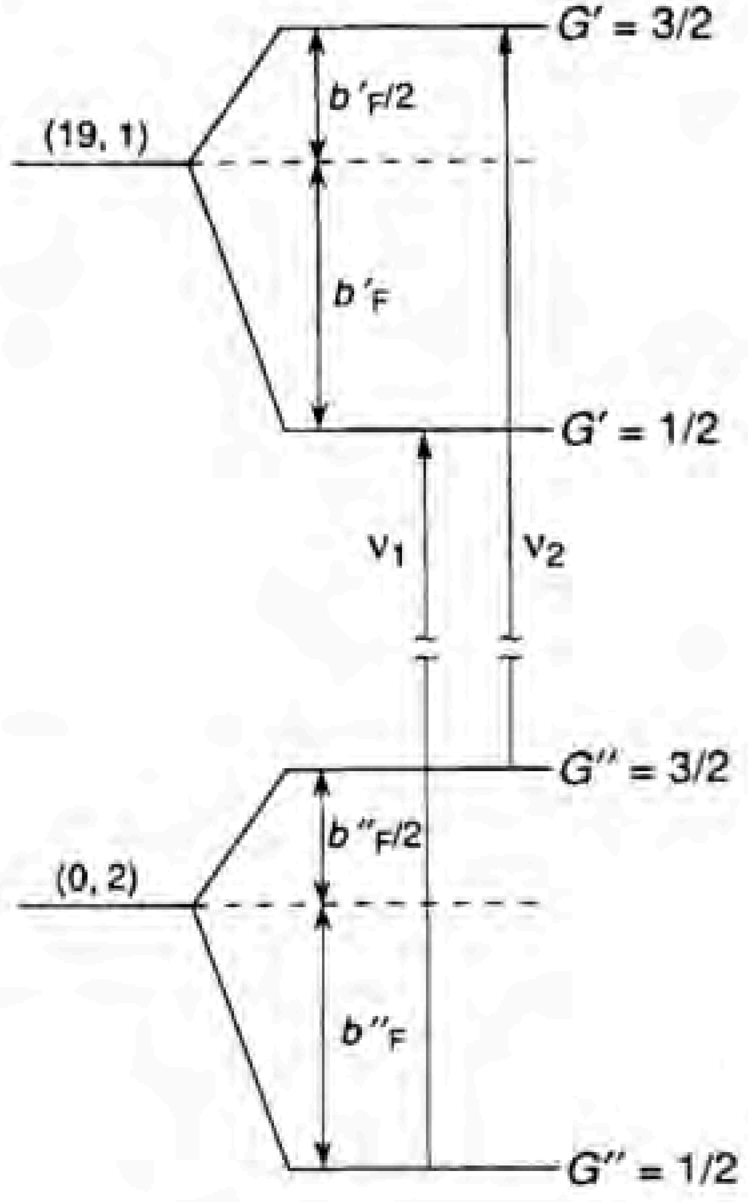
Microwave Spectroscopy at the Dissociation Limit

Alan Carrington



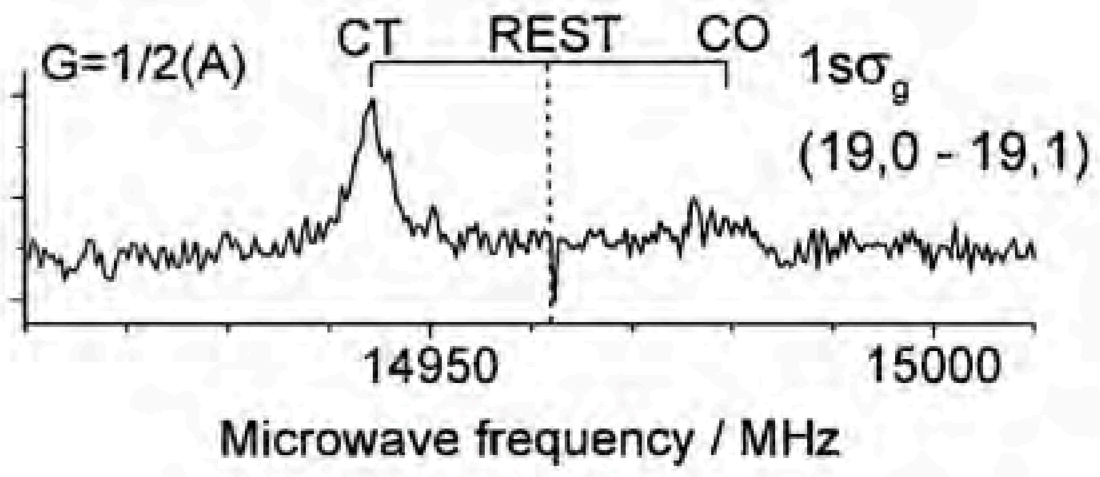
600 kHz linewidth due to transit time broadening



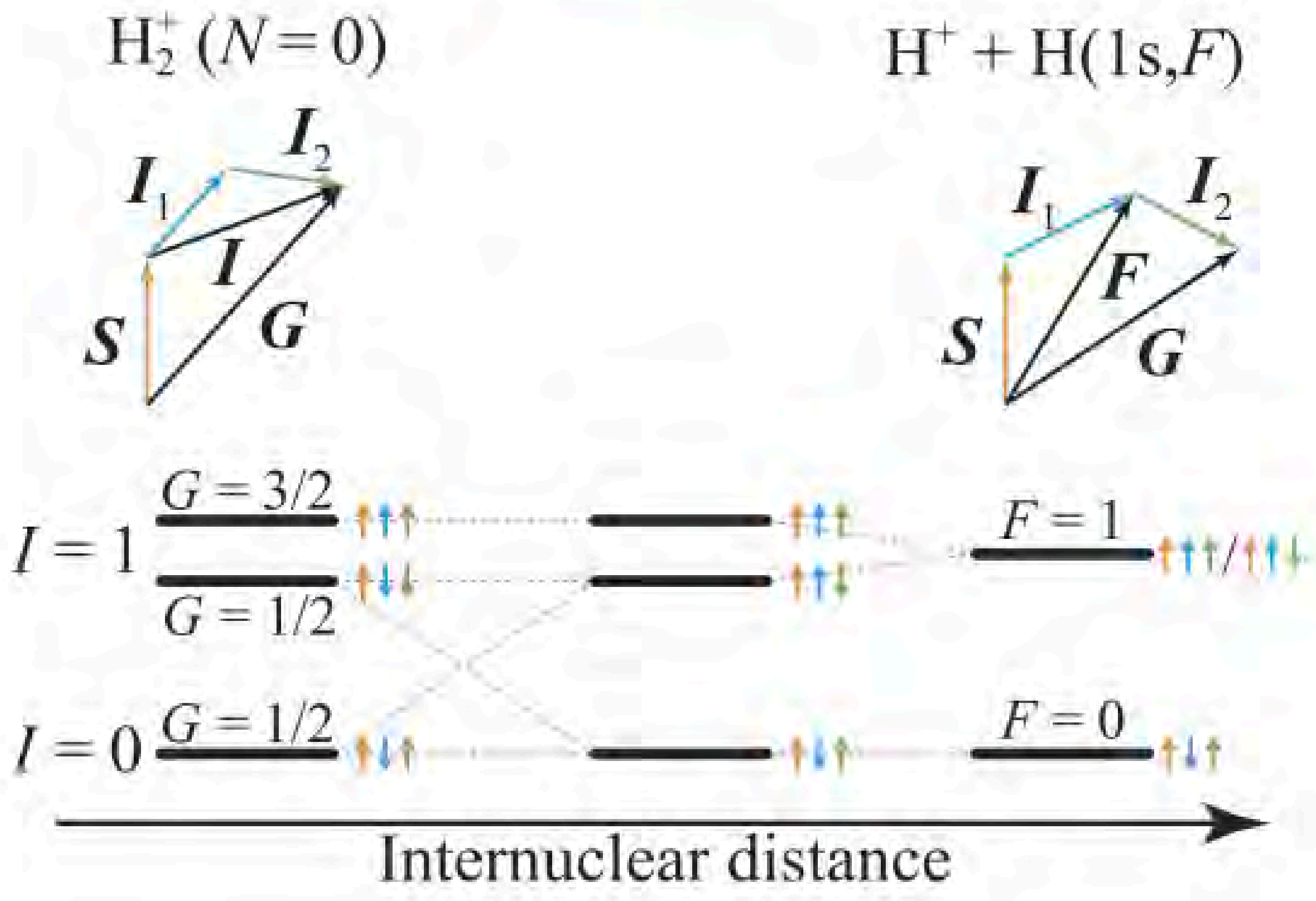


Mixing of ortho and para (= gerade and ungerade)!

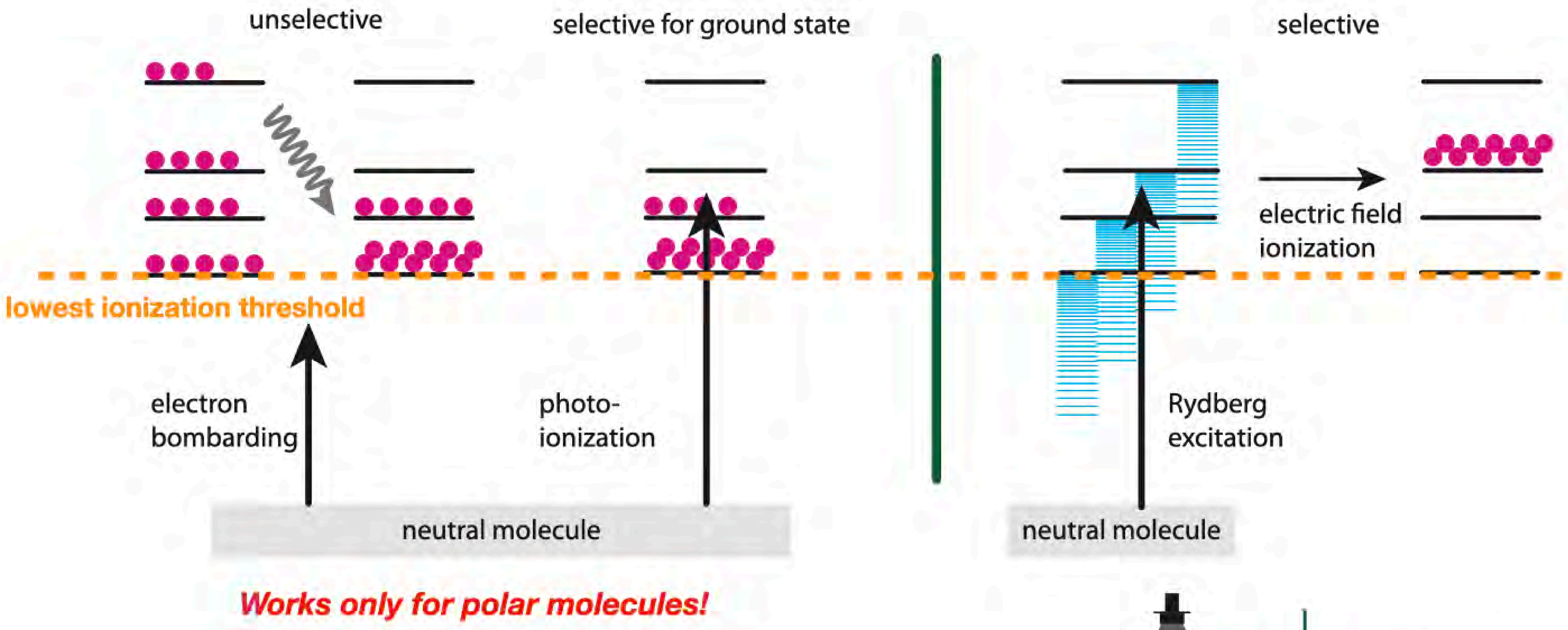
Critchley *et al.*, *PRL* **86**, 1725 (2001)

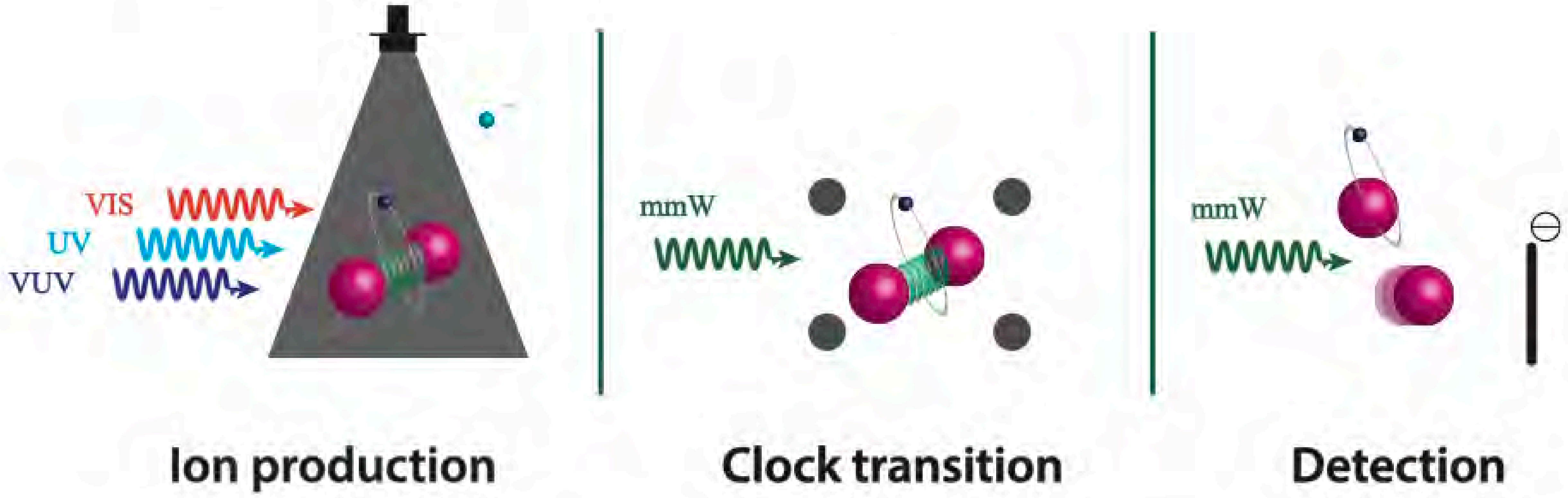


550 kHz



Weakly-bound states of hydrogen molecular ions

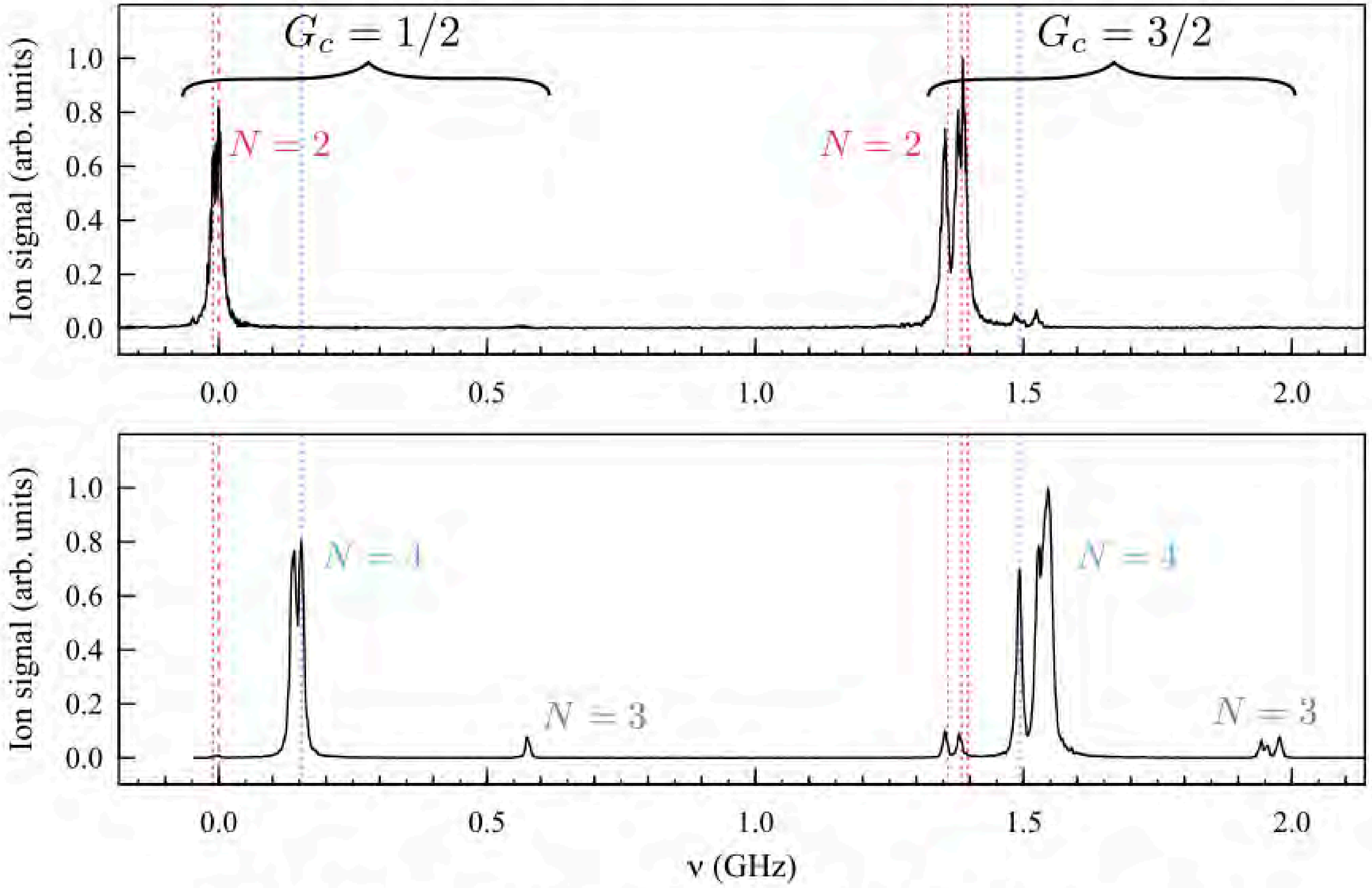




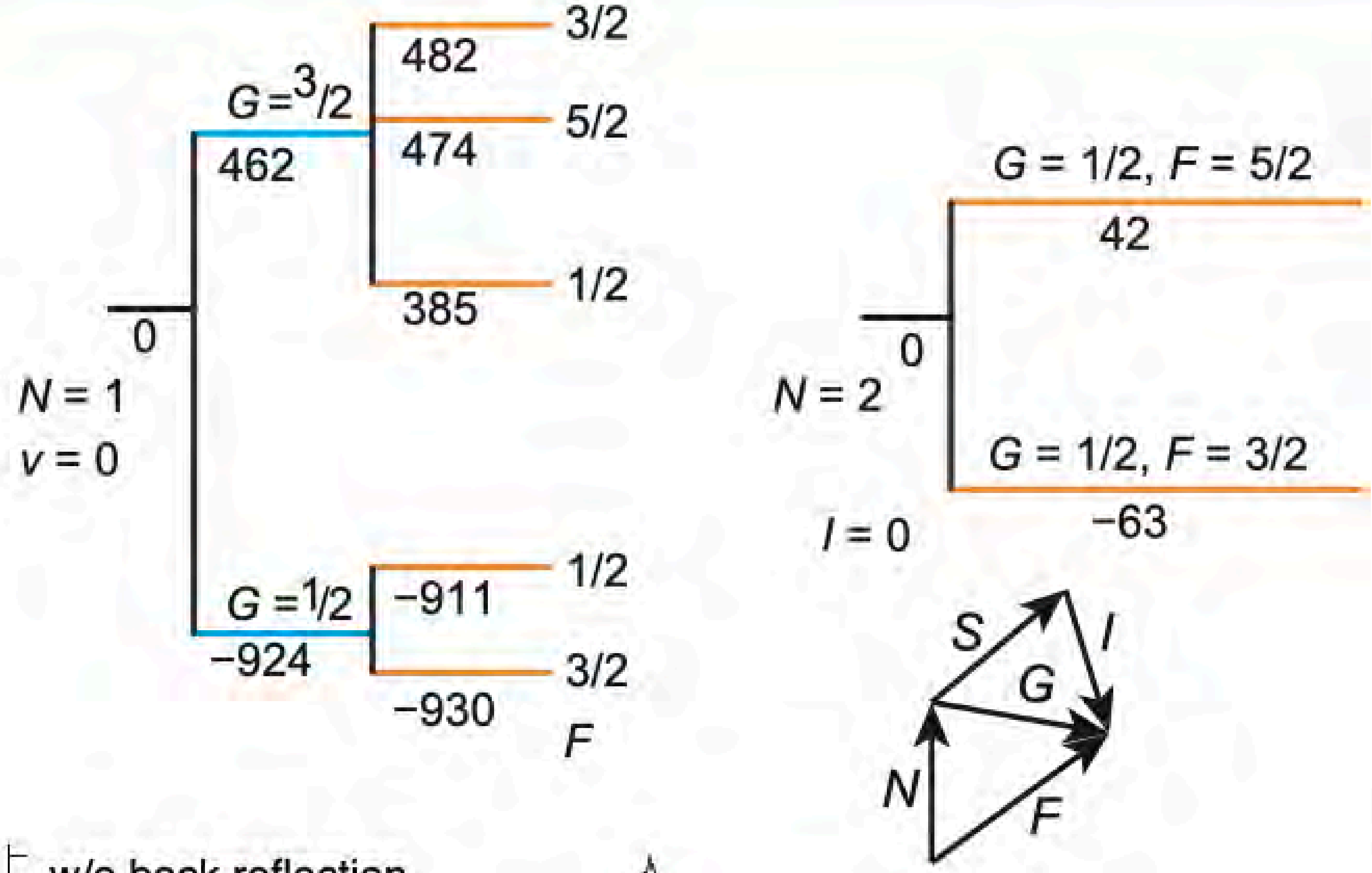
Molecular Rydberg states with resolved hfs

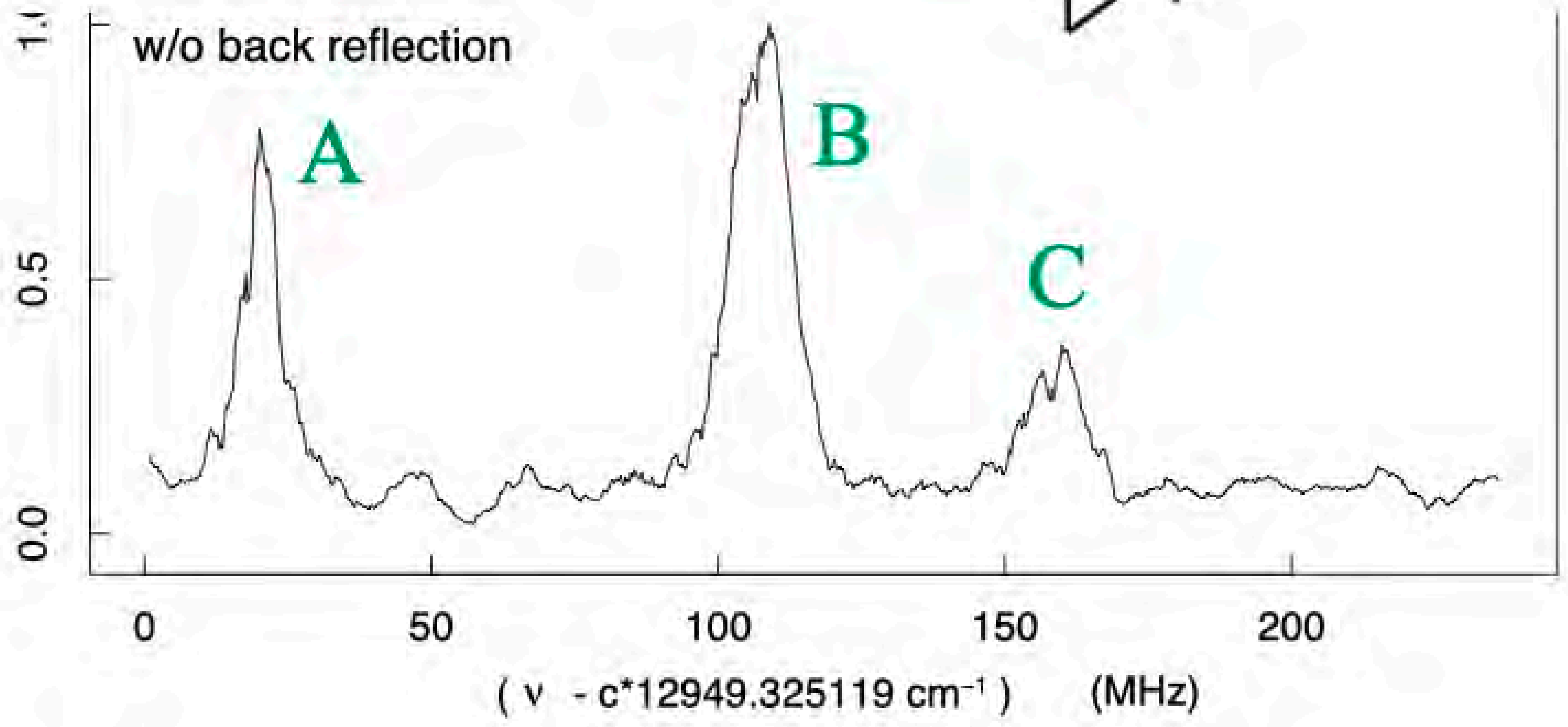
➡ **Excitation of f Rydberg states (small exchange interaction)**

$$\mathrm{GK}(v = 1, N = 1) \rightarrow 56\mathrm{f}1(v^+ = 0)$$



$$\mathrm{GK}(v = 0, N = 3) \rightarrow 56\mathrm{f}1(v^+ = 0)$$



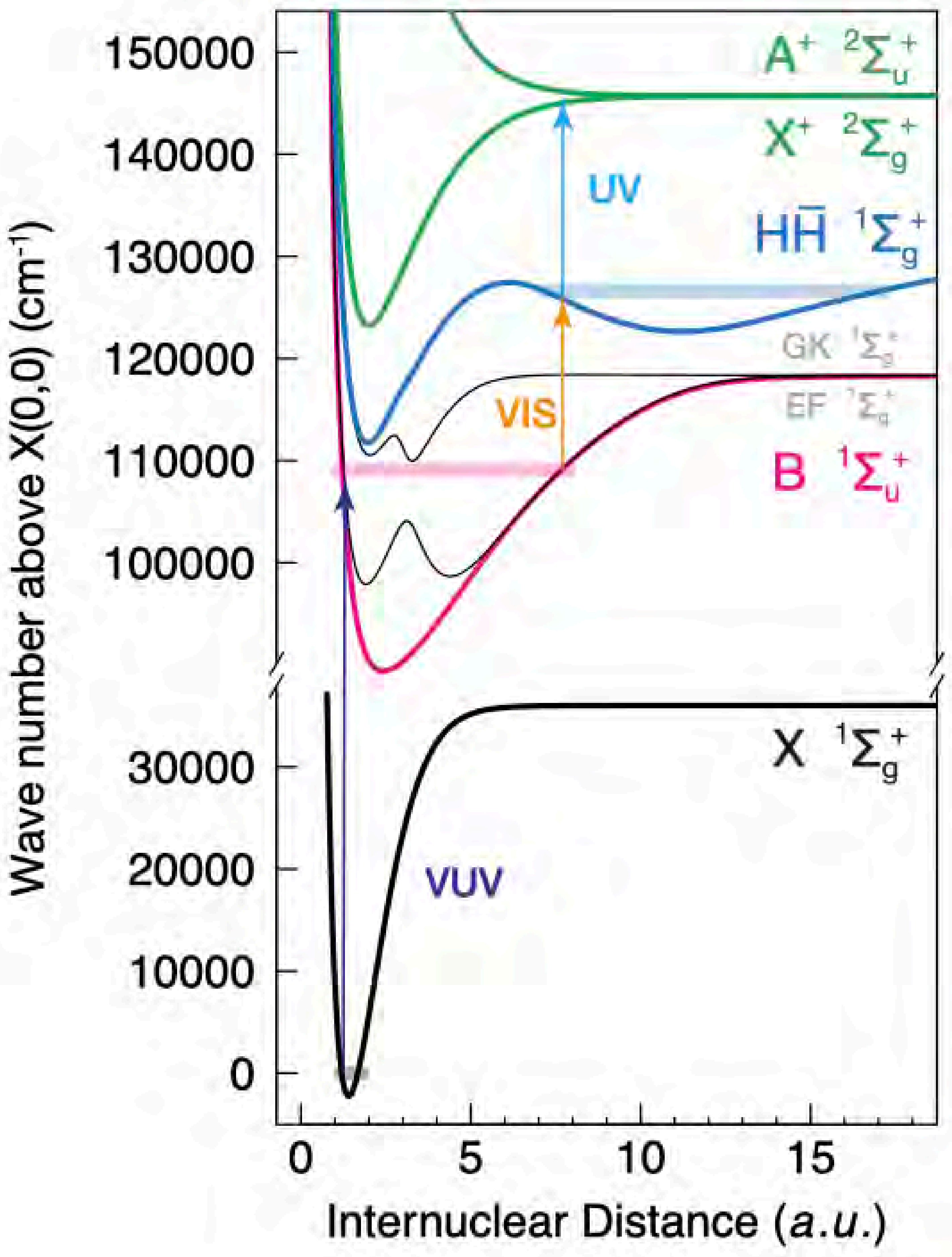


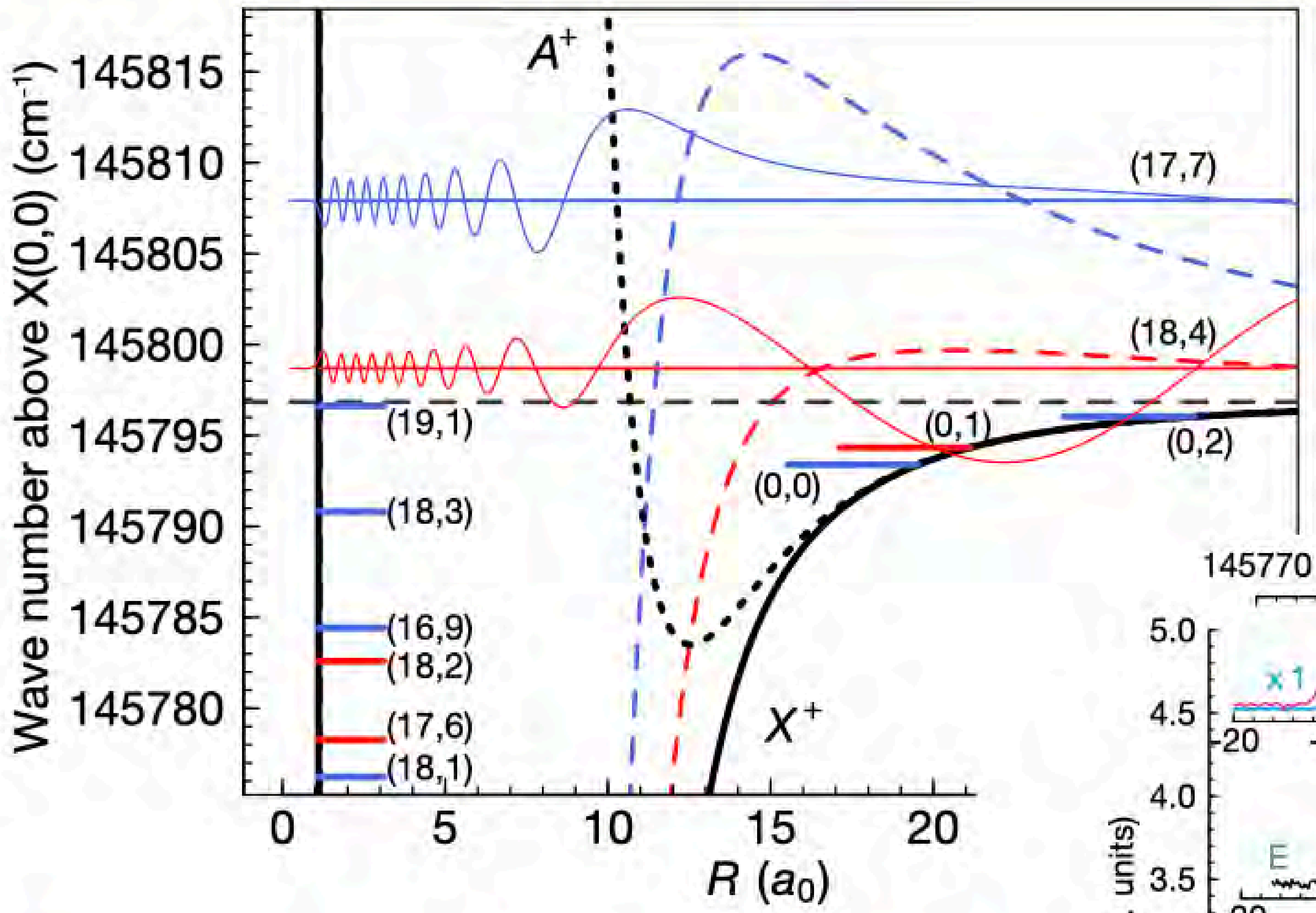
GK(0,0) -> 90f2 (J=1)

M. Beyer, PhD Thesis, ETH Zürich (2018)

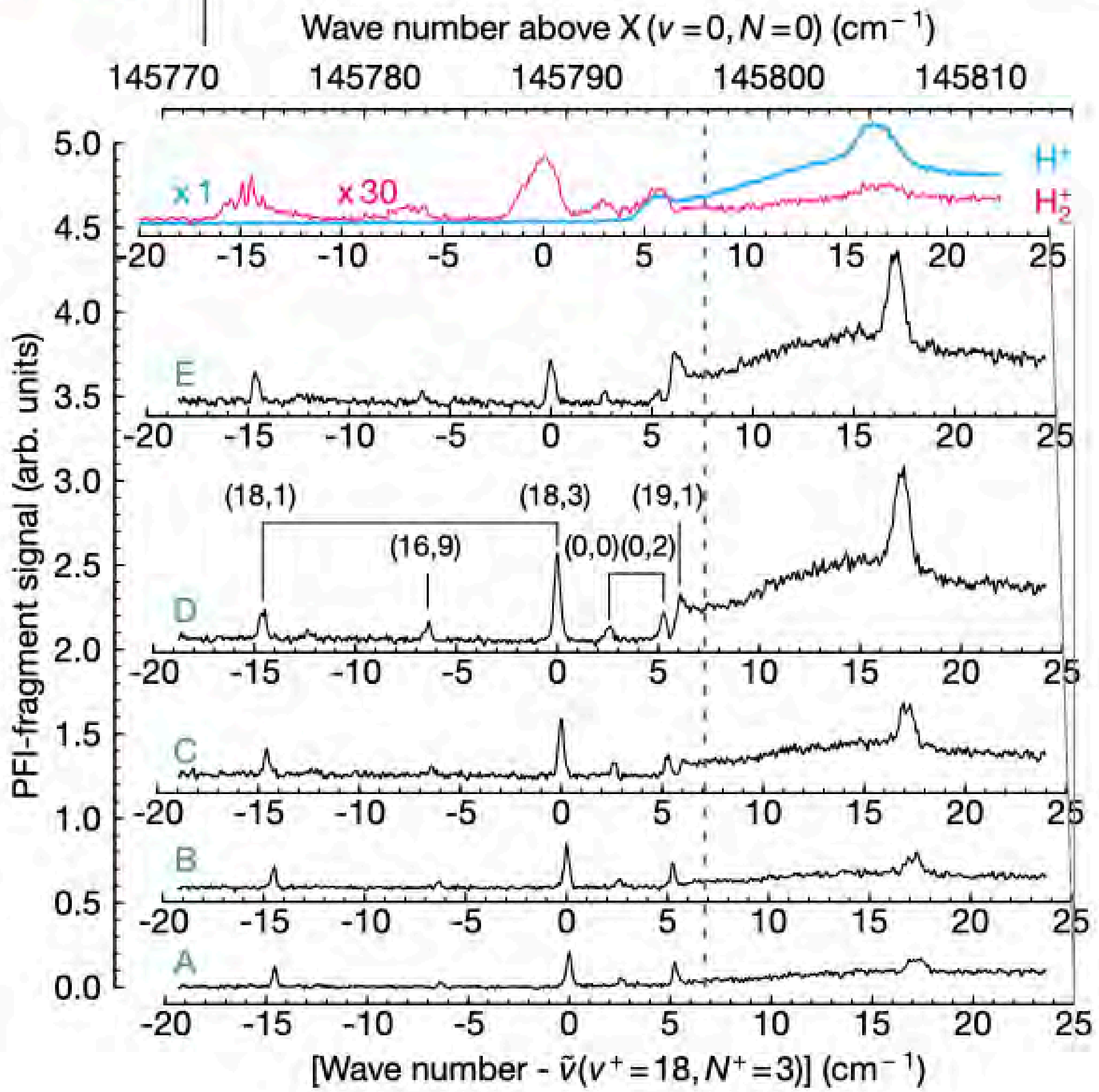
➡ **Selective generation of molecular ions.**

Weakly-bound states of hydrogen molecular ions



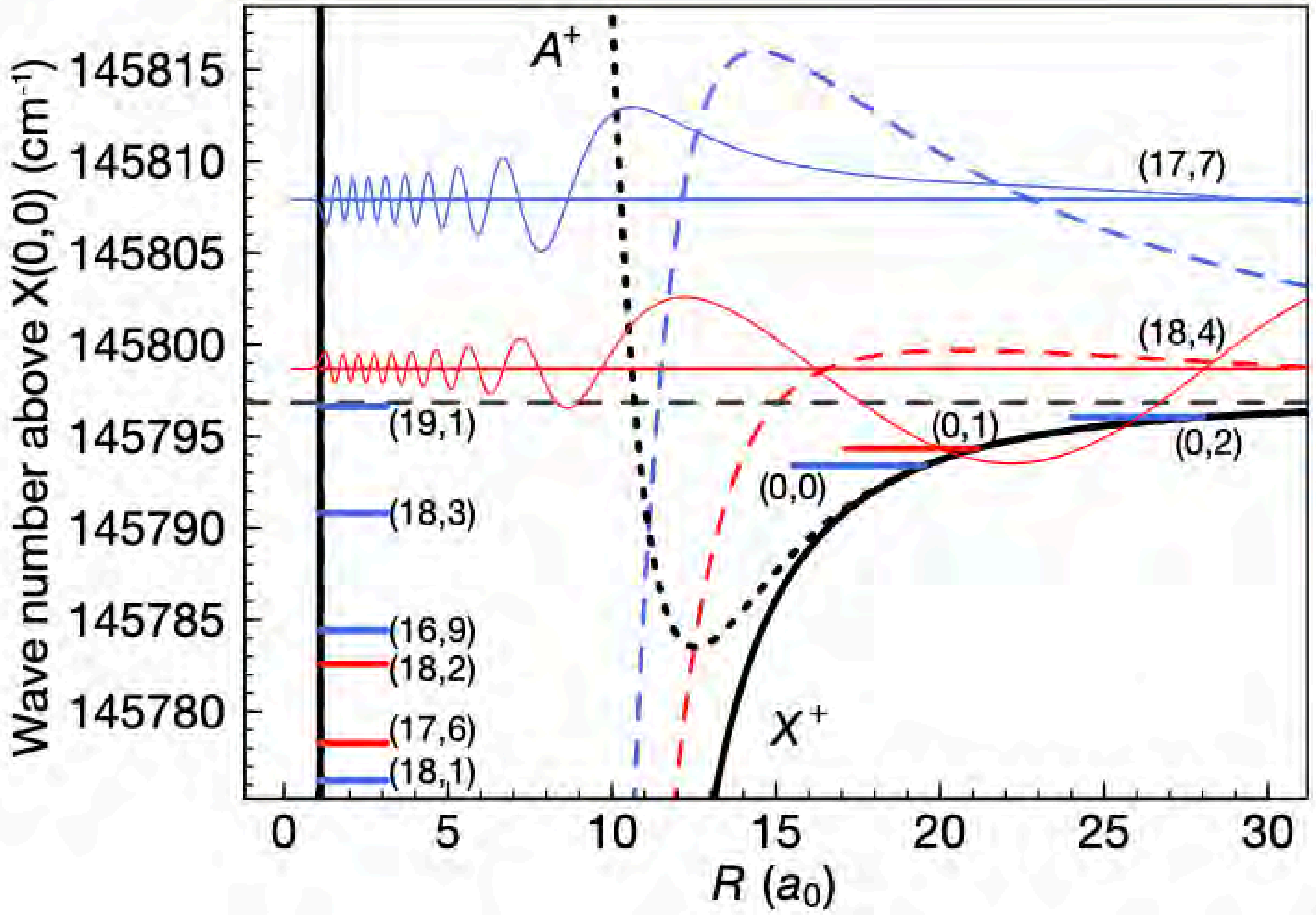


Beyer and Merkt, *Phys. Rev. Lett.* **116**, 093001 (2016)
Beyer and Merkt, *J. Mol. Spectrosc.* **330**, 147 (2016)

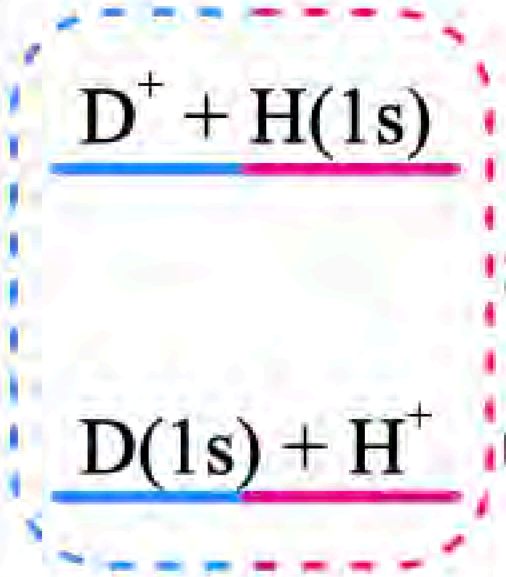


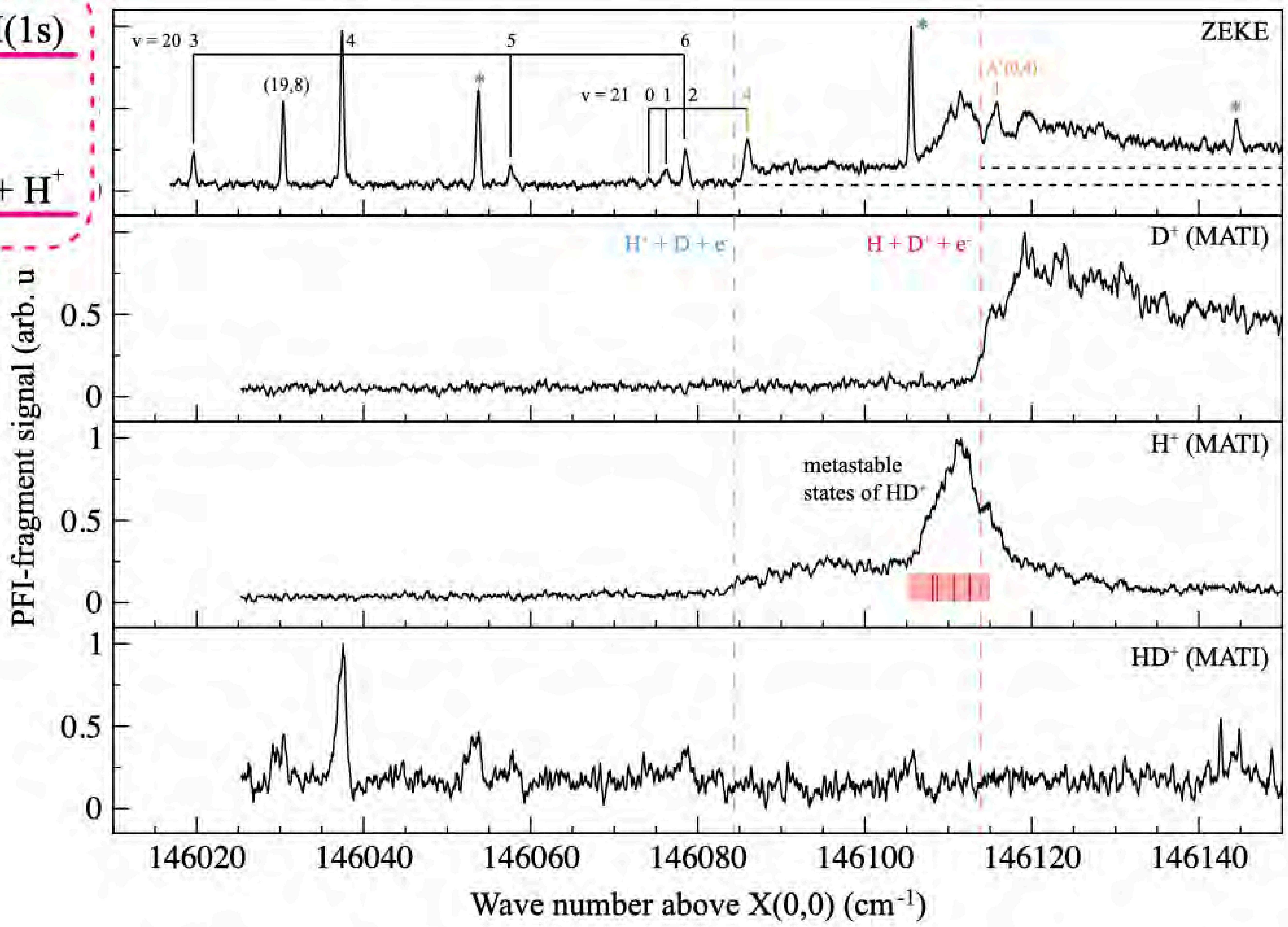
- ➡ **Enhanced sensitivity for m_p/m_e: K ~ 65**
- ➡ **Enhanced sensitivity for g/u-mixing (= ortho/para mixing)**
- ➡ **Measurement of pure rotational transitions**

Weakly-bound states and Feshbach resonances in HD^+

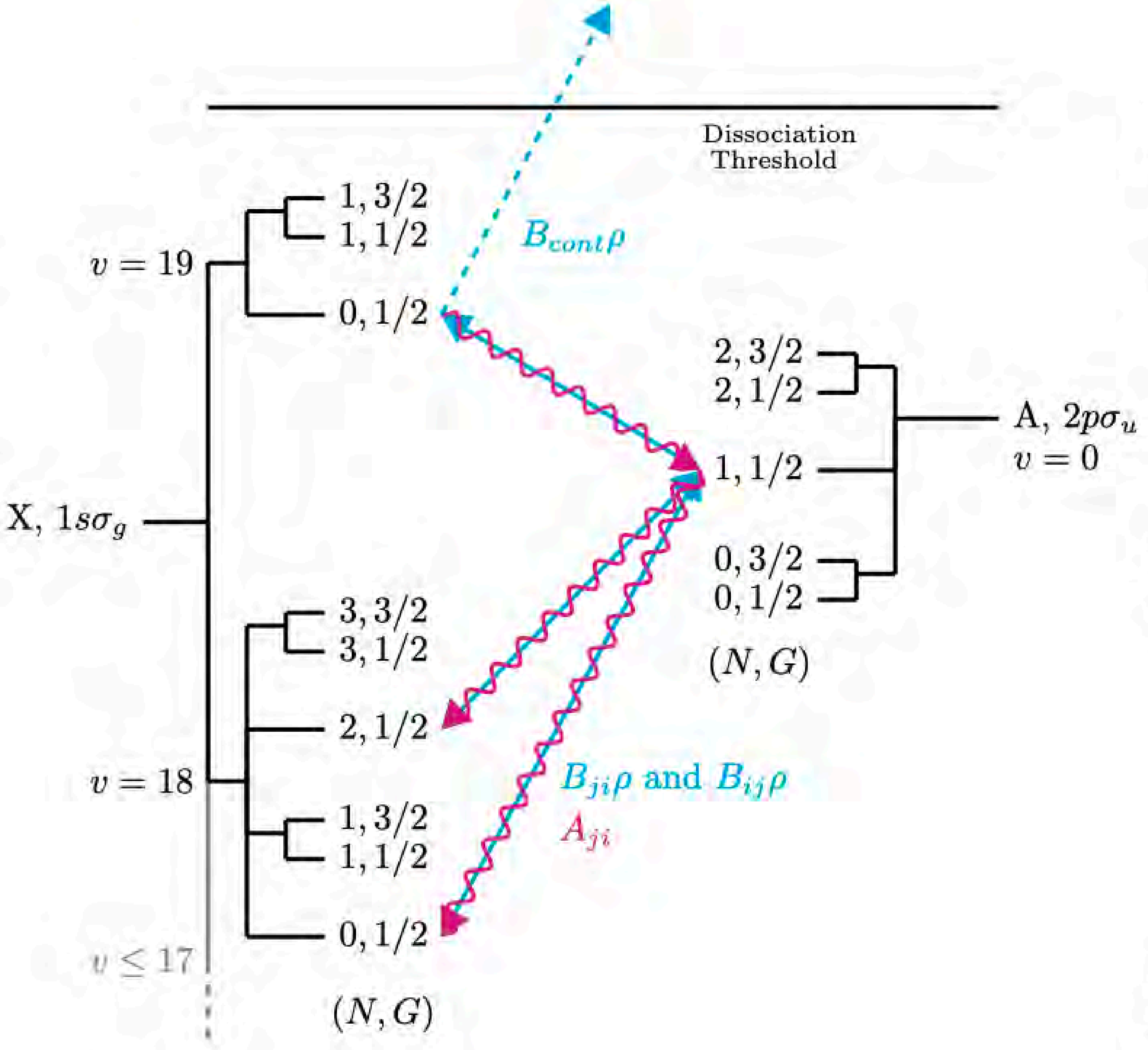


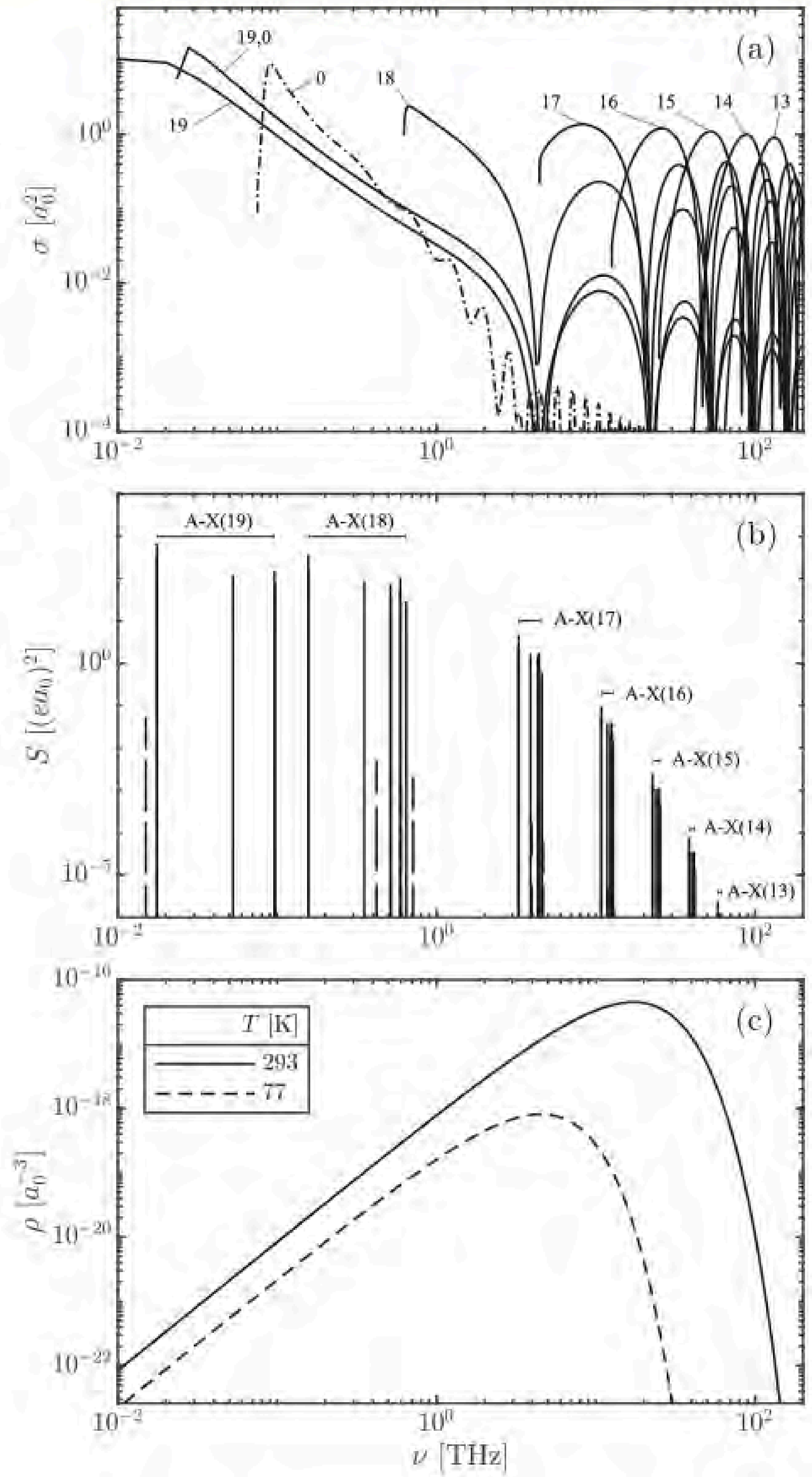
Beyer and Merkt, *Mol. Phys.* e2048108 (2022)

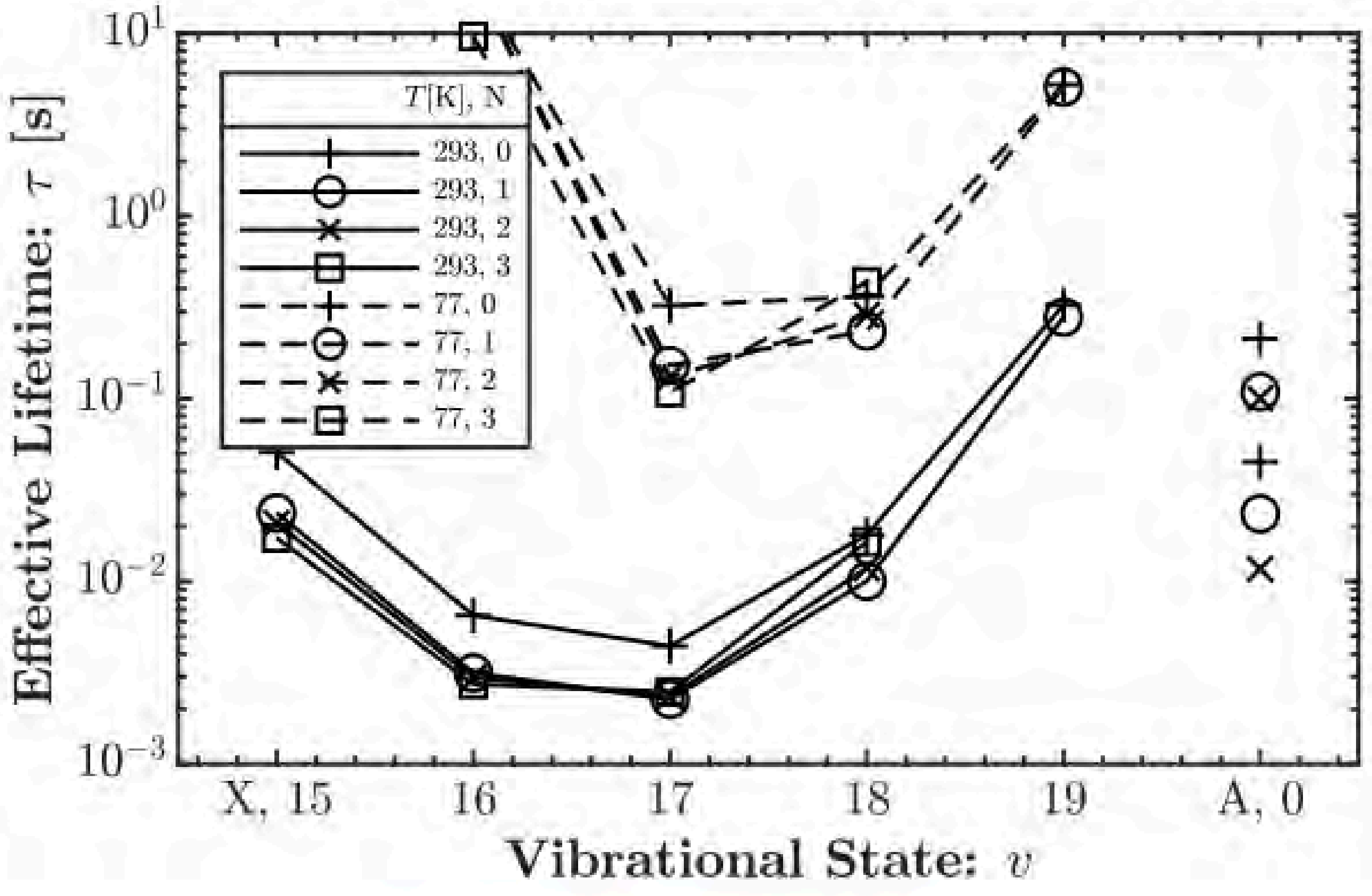




Weakly-bound states: blackbody redistribution







➡ >1 ms lifetime of

Work by:

D. Ochoa, MB

Summary

- *Most accurate determination of the dissociation energy of a molecule.*
- *Experimental and theoretical accuracy sufficient to see the effect of the charge radius.*
- *Experimental uncertainty for D_0 on the way towards ~ 100 kHz.*
- *Rydberg states can be used to create molecular ions selectively.*
- *Measurements of weakly-bound molecular ions as m_p/m_e and ortho/para mixing probe*

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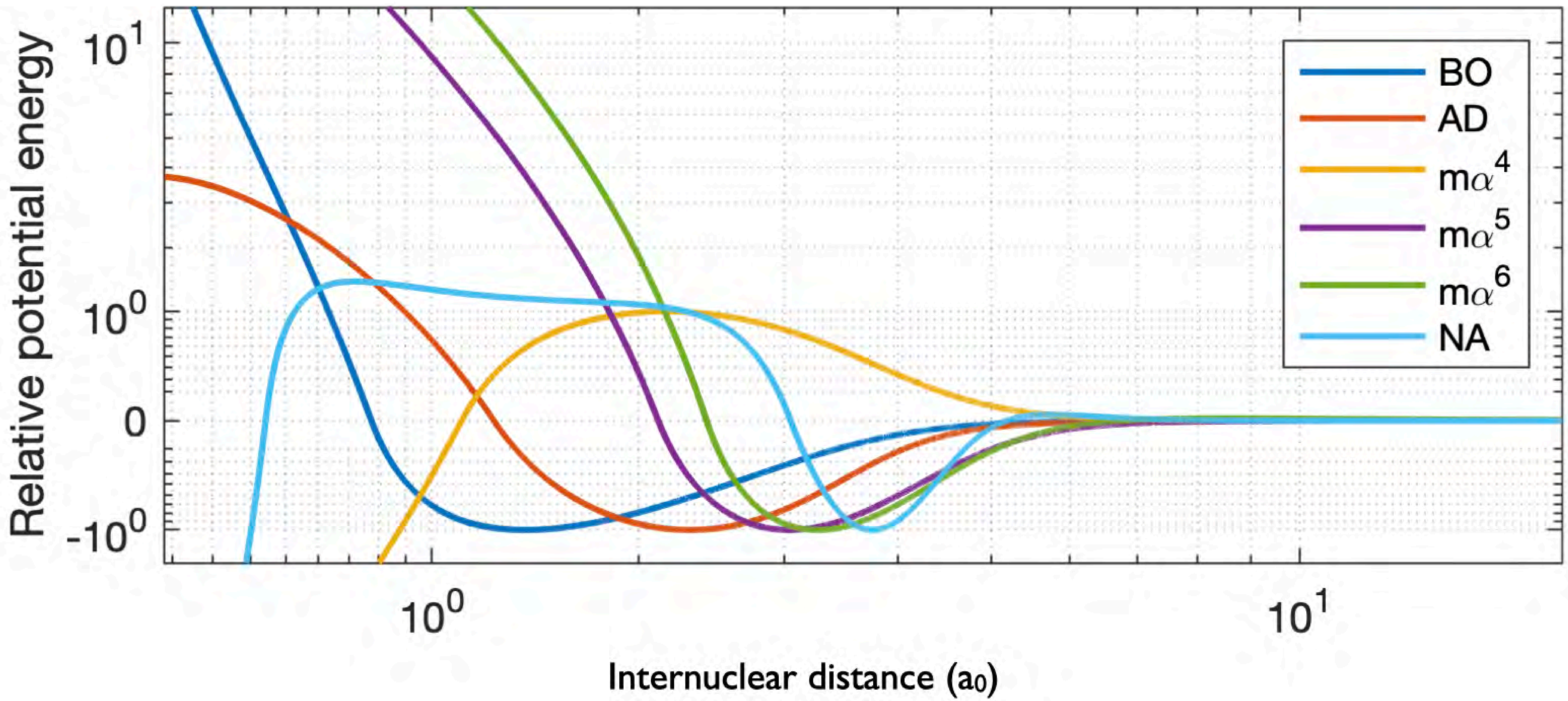
@ ETH

Ioanna Doran

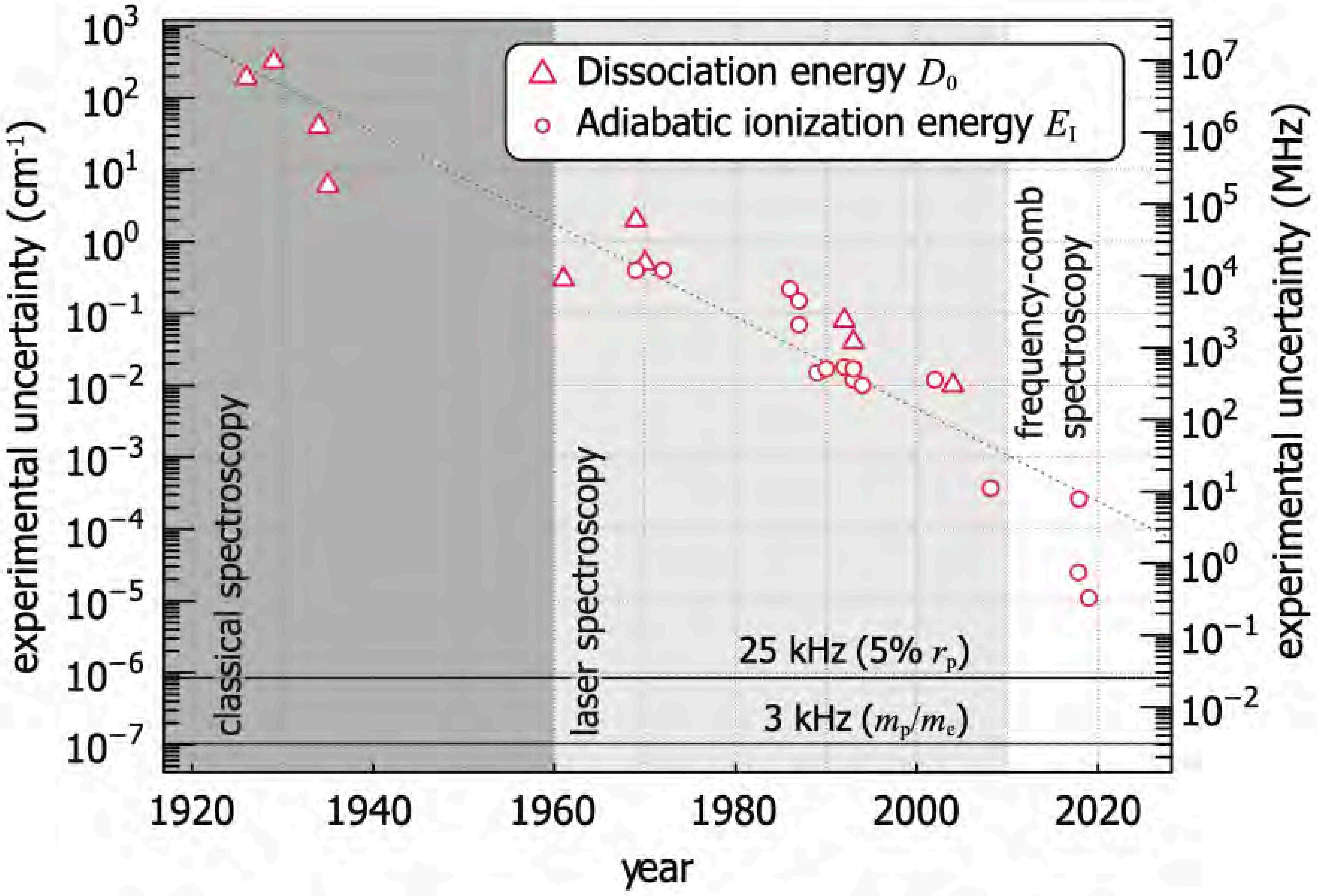
Josef Agner

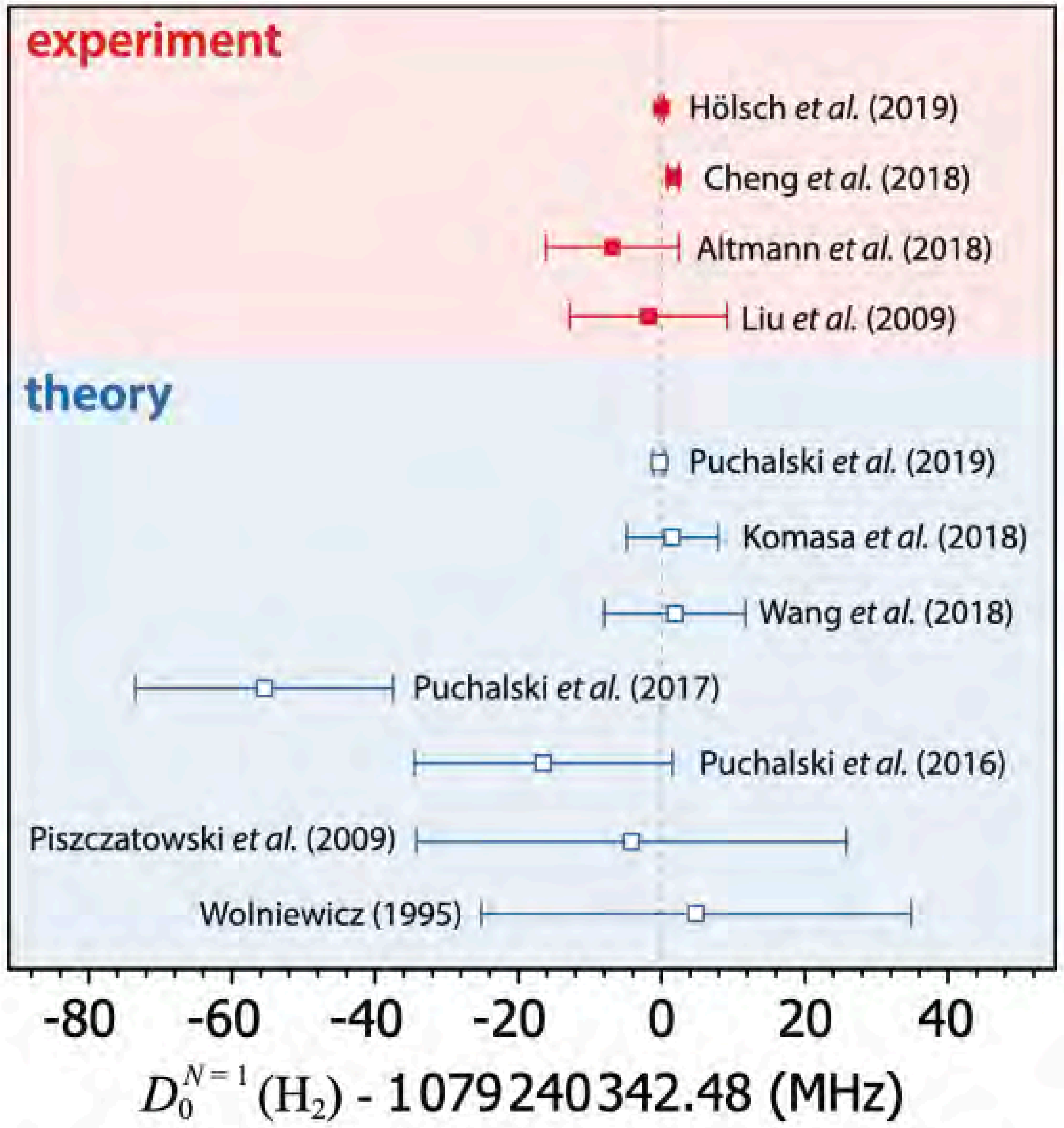
Hansjürg Schmutz

Electronic ground state of H_2 - corrections

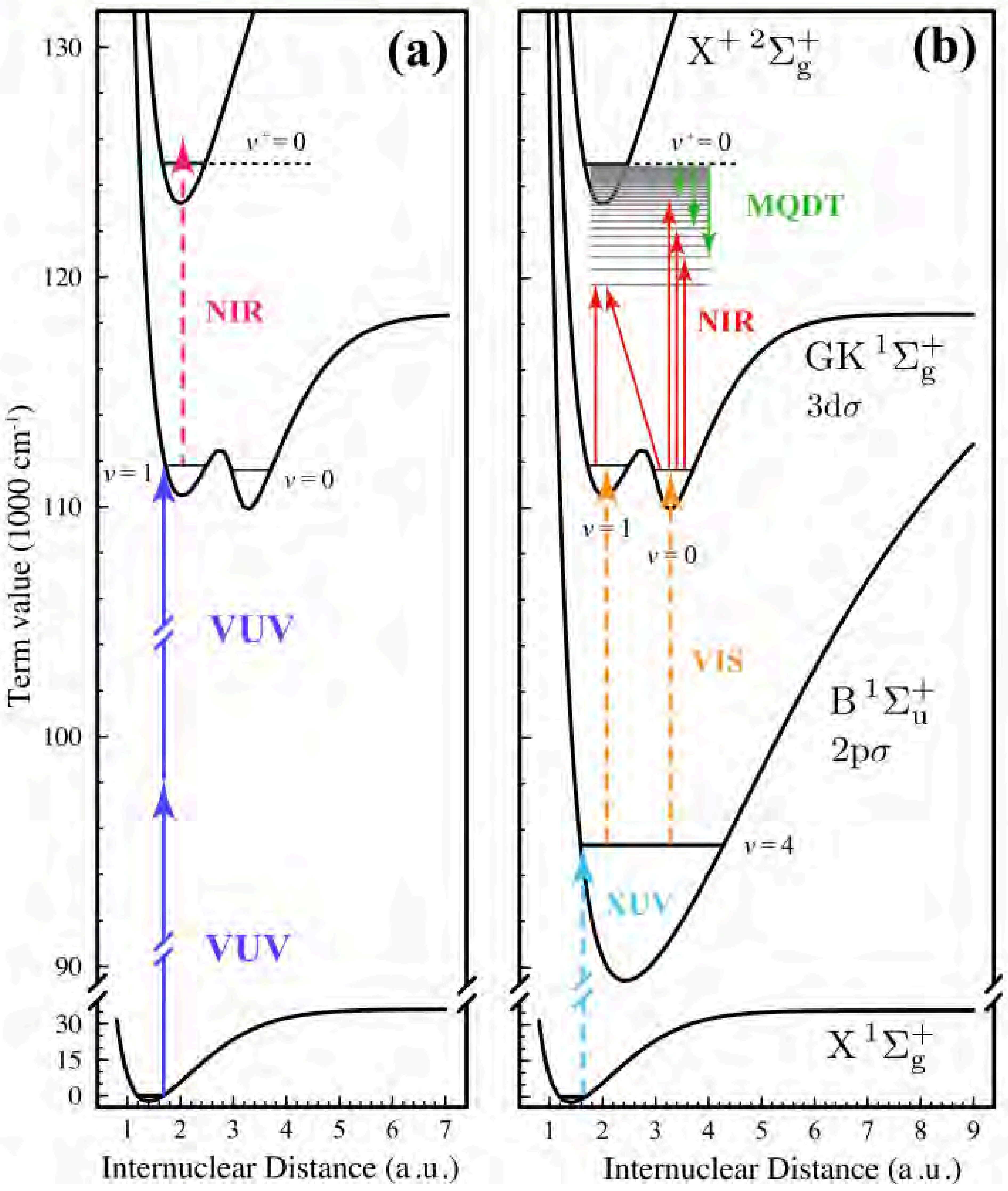


D₀ in H2 - theory vs. experiment





Ionization energy in D_2



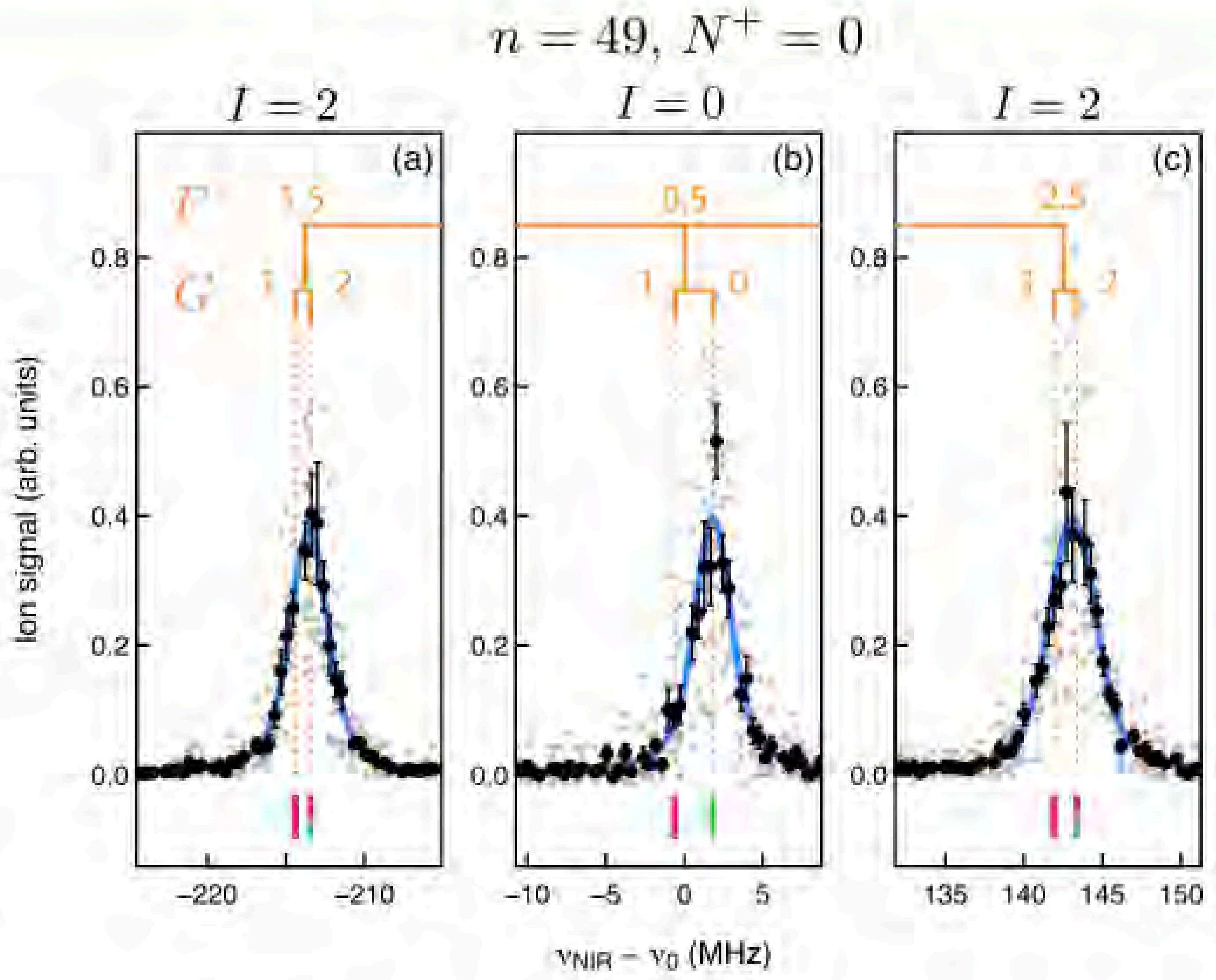


TABLE I. Error budget of the measurement of the GK $^1\Sigma_g^+(1,2) \leftarrow$ X $^1\Sigma_g^+(0,0)$ transition in D_2.

Measured frequency	3 360 646 104.46 MHz
Effect	Uncertainty
Residual first-order Doppler	$(350\text{ kHz})_{\text{stat}}$
Second-order Doppler	30 kHz
ac-Stark ionization laser	100 kHz
ac-Stark VUV laser	240 kHz
Hyperfine structure	100 kHz
Final frequency	3 360 646 104.46(45) MHz

TABLE II. Error budget for the transition between the GK(0,2) state of D_2 and the $62f0_3(I=0, S=0)$ Rydberg state, resulting from a series of independent measurements. All systematic uncertainties are the same for the other measured transitions except the uncertainty in the dc Stark shift which is n-dependent (see text).

Measured frequency	388 255 115.129(16) MHz	
	Correction	Uncertainty
dc Stark shift		60 kHz
ac Stark shift		~5 kHz
Zeeman shift		~10 kHz
Pressure shift		~1 kHz
Residual first-order Doppler shift		$(<200\text{ kHz})_{\text{stat}}$
Second-order Doppler shift	+2 kHz	0.5 kHz
Line-shape model		50 kHz
Photon-recoil shift	−83 kHz	
Systematic uncertainty		80 kHz
Final frequency	388 255 115.048(82) MHz	

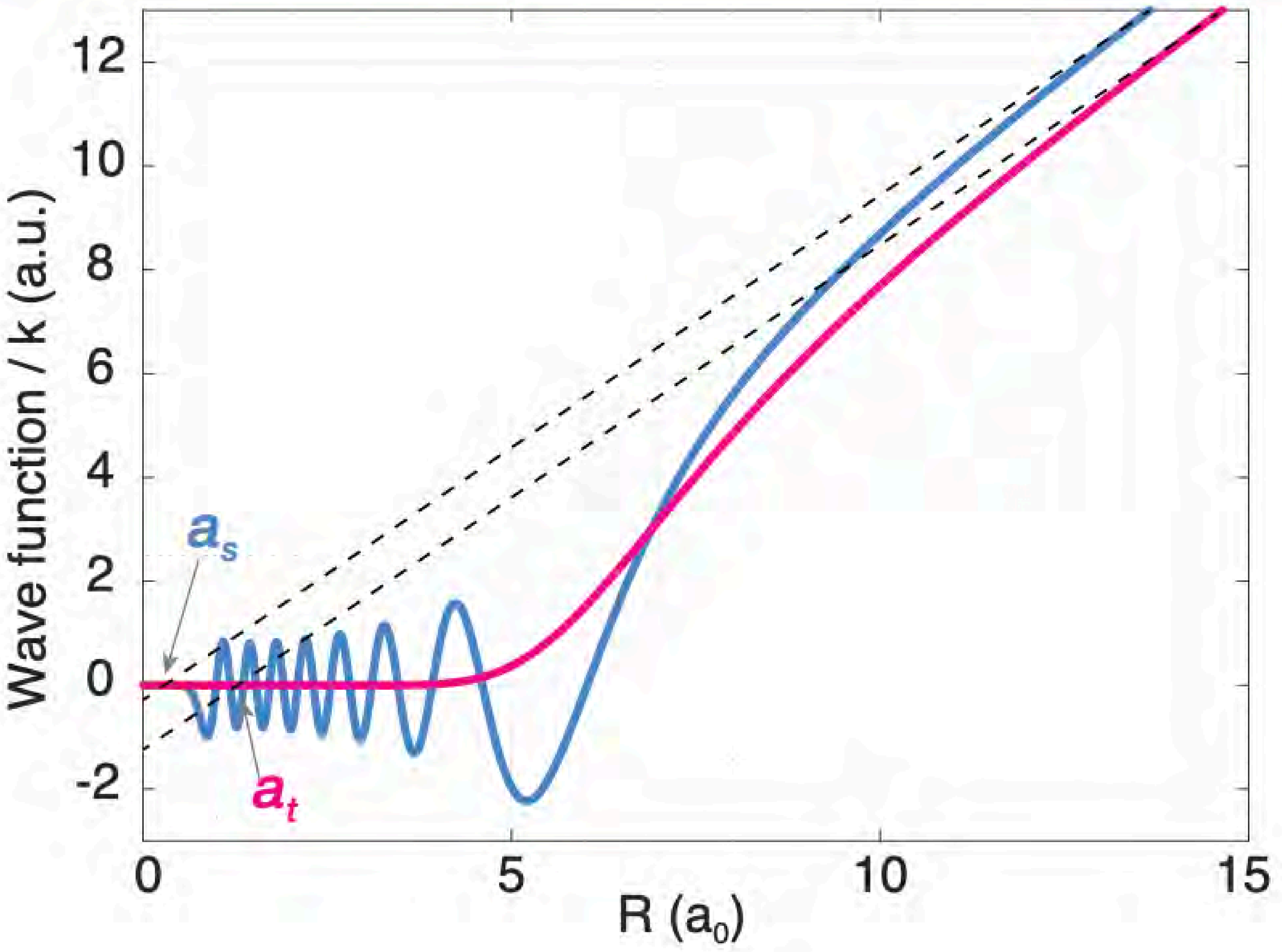
Rydberg spectroscopy - characteristics

	H_2
Initial state	$GK\ ^1\Sigma_g^+\ (3s/d)$
Final state	$50\ f0_3(S=0, v^+=0)^2\Sigma_g^+$
$\tilde{\nu}$ (cm^{-1})	12 700
α_{nl} [$MHz/(V/cm)^2$]	$5.47(11) \times 10^4$
Lifetime initial state (ns)	69(6)
Lifetime final state (μs)	>5
$\Gamma_{natural}$ (MHz)	2.31(20)
Density (particles/cm^3)	10^{11}
Sample volume (mm^3)	1
Mass (u)	2.0
Temperature (μK)	100[b]
Velocity (m/s)	1390[c]
Γ_{laser} (MHz)	1.2
$\Gamma_{Doppler}$ (MHz)	<2
Γ_{FWHM} (MHz)	5

Electric field (mV/cm)	<0.8
Magnetic field (mG)	≪40
Line-shape model (kHz)	<50
dc Stark shift (kHz)	18
Zeeman shift/broadening (kHz)	<10
ac Stark shift (kHz)	<4
Rydberg-Rydberg interaction (kHz)	≪1
Pressure shift (kHz)	<1
First-order Doppler shift (kHz)	<110[e]
Second-order Doppler shift (kHz)	4.1(5)
σ_{stat} (kHz)	91
σ_{sys} (kHz)	54

[a]Calculated using the approach described in Ref. [39].
[b]Corresponds to the width of the velocity distribution in the direction of the NIR laser beam.
[c]For a valve temperature of 88 K.
[d]ac Stark shift caused by residual light from the optical-dipole-trapping laser.
[e]The uncertainty averages out over several measurements and contributes to σ_{stat} (see text).

Determination of the scattering length

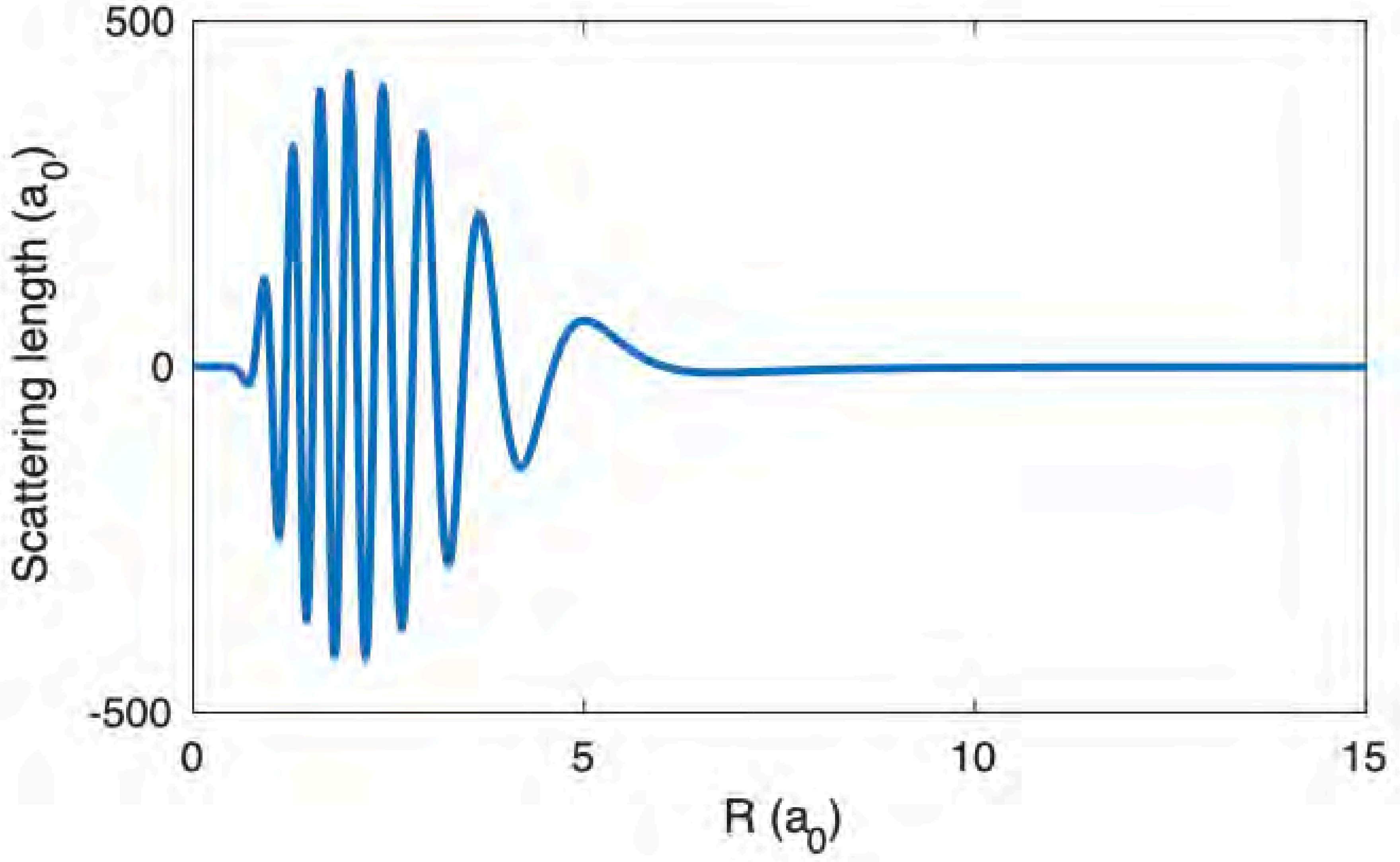


$$a = -\lim_{k\to 0} \frac{\tan \eta_{J=0}}{k}.$$

$$k = \sqrt{2\mu(E - \mathcal{V}(\infty))}$$

$$\lim_{k\to 0} \chi(R;k) \propto \sin(kR) + \tan\eta_{J=0}\cos(kR)$$
$$\propto k(R-a)$$

$$a = \frac{2\mu}{k^2}\int_0^\infty \sin(kR)\mathcal{V}(R)\chi(R;k=k_0)dR$$



Universal model: scattering length + C_6 ↔ resonance position + width

PHYSICAL REVIEW A **82**, 012510 (2010)

Shape resonances in ground-state diatomic molecules: General trends and the example of RbCs

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(Received 28 April 2010; published 29 July 2010)