Improvement of the Dissociation Energy of the Hydrogen Molecule (Part One)

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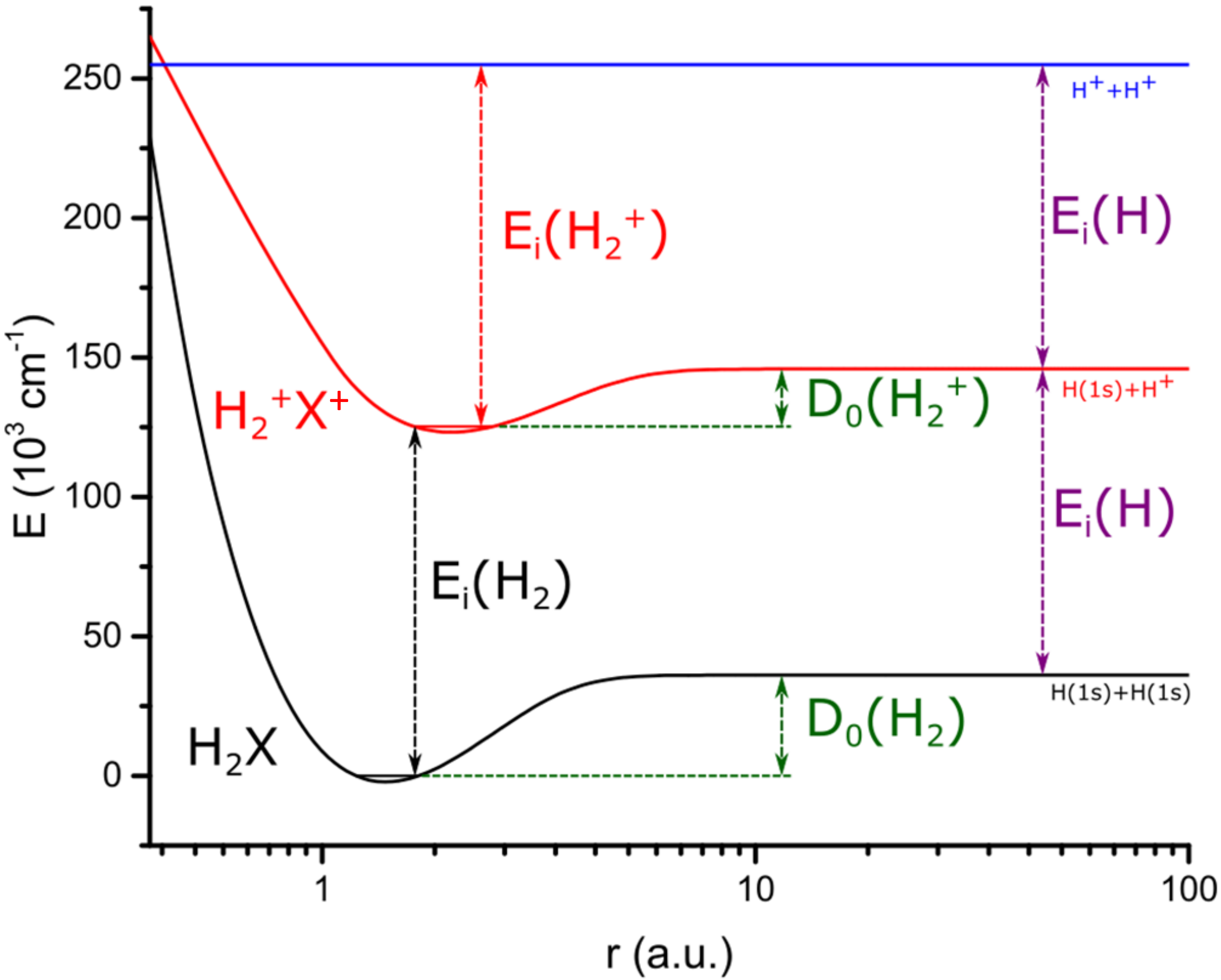
[b] Laboratorium für Physikalische Chemie, ETH Zürich

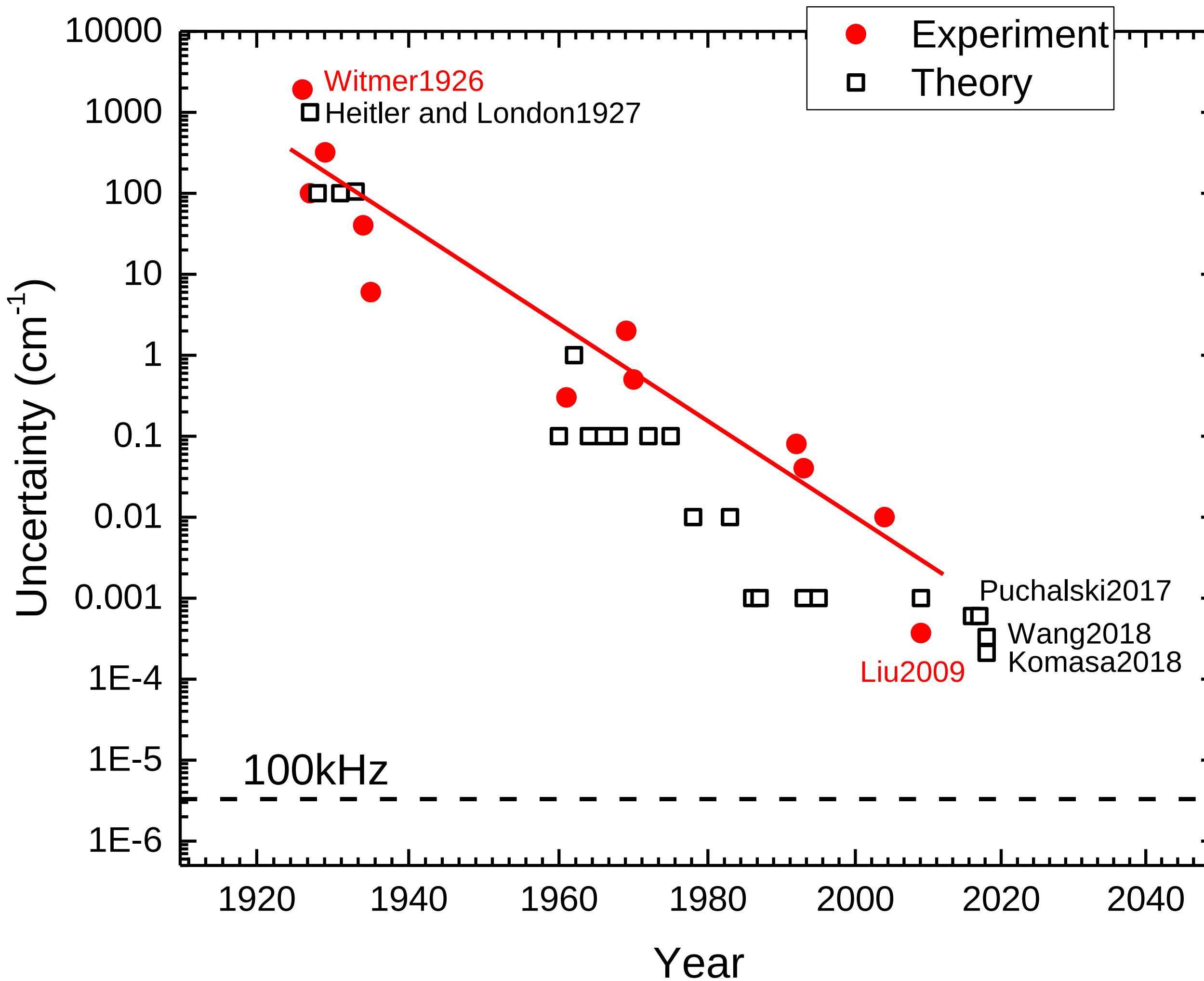
[c] Department of Chemical Physics, University of Science and Technology of China

[d] Laboratoire Aimé Cotton, CNRS, Orsay

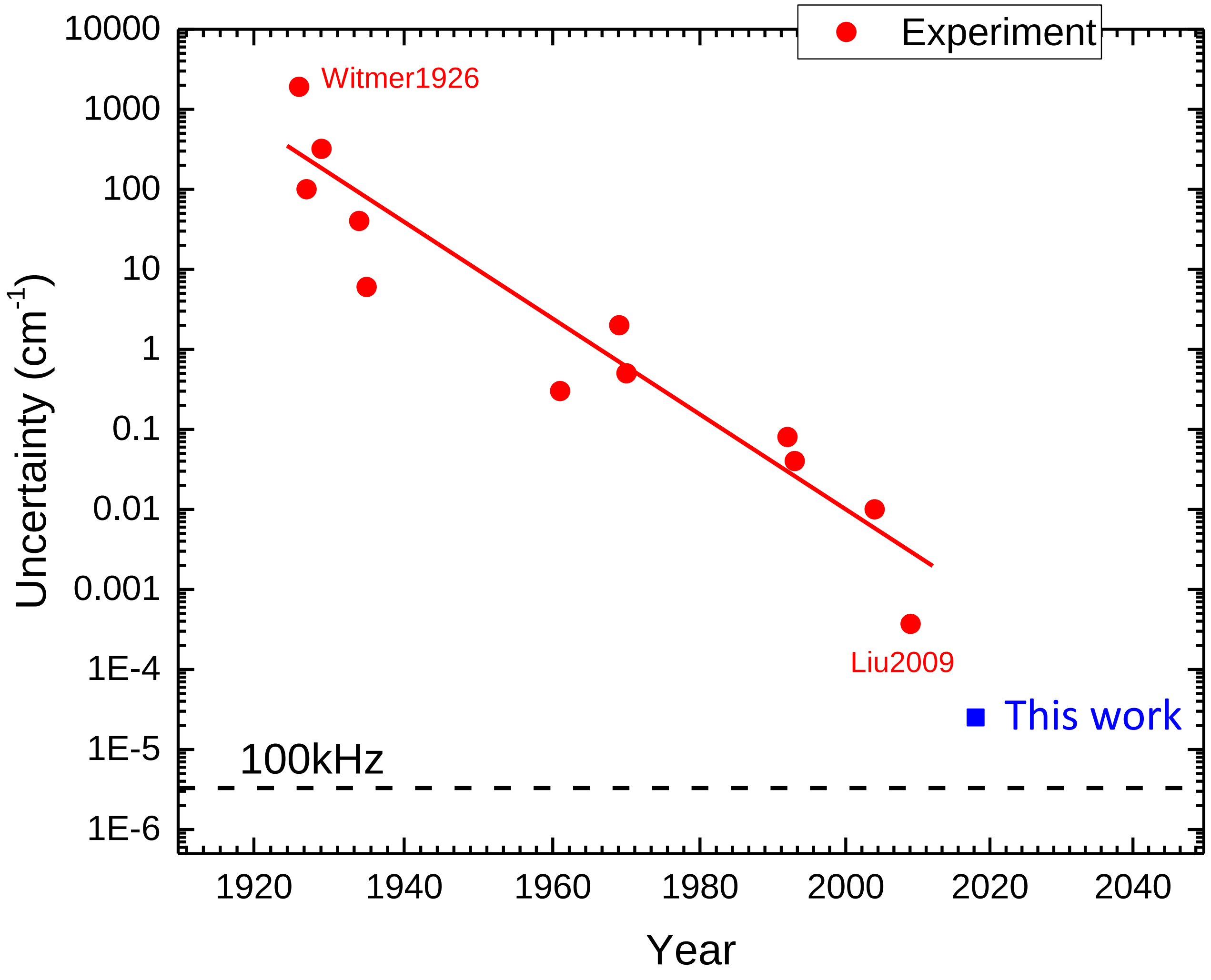
Determining D_0



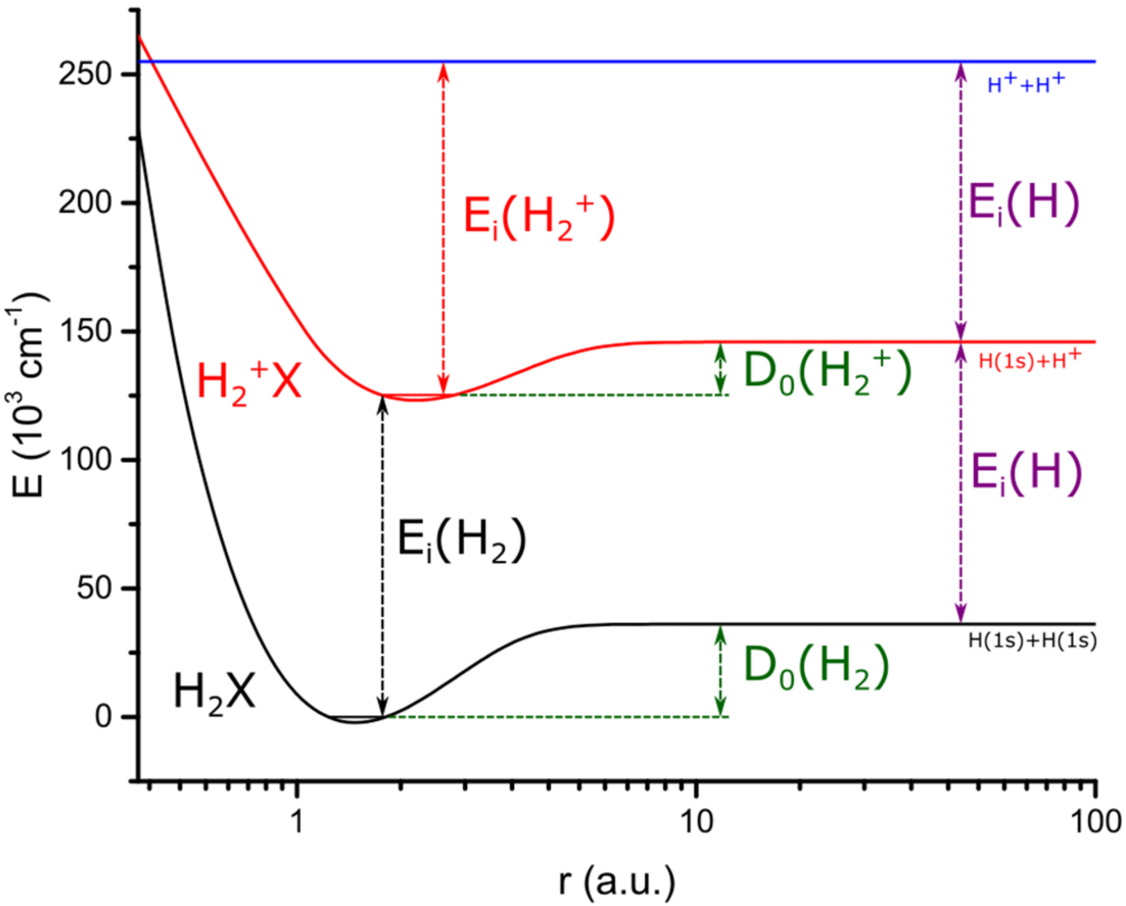
Test of molecular theory with D_0



Test of molecular theory with D_0



Determining D_0(ortho-H_2)



$$D_0(H_2) = E_i(H_2) + E_i(H_2^+) - 2E_i(H)$$

$E_i(H)$ =109678.77174307(10) cm-1

~3 kHz

$E_i(H_2+)$ =131058.1219937(6) cm-1

~18 kHz

$E_i(H_2)$ =124417.49113(37) cm-1

~11 MHz

$D_0(H_2)$ = 36118.0696(4) cm-1

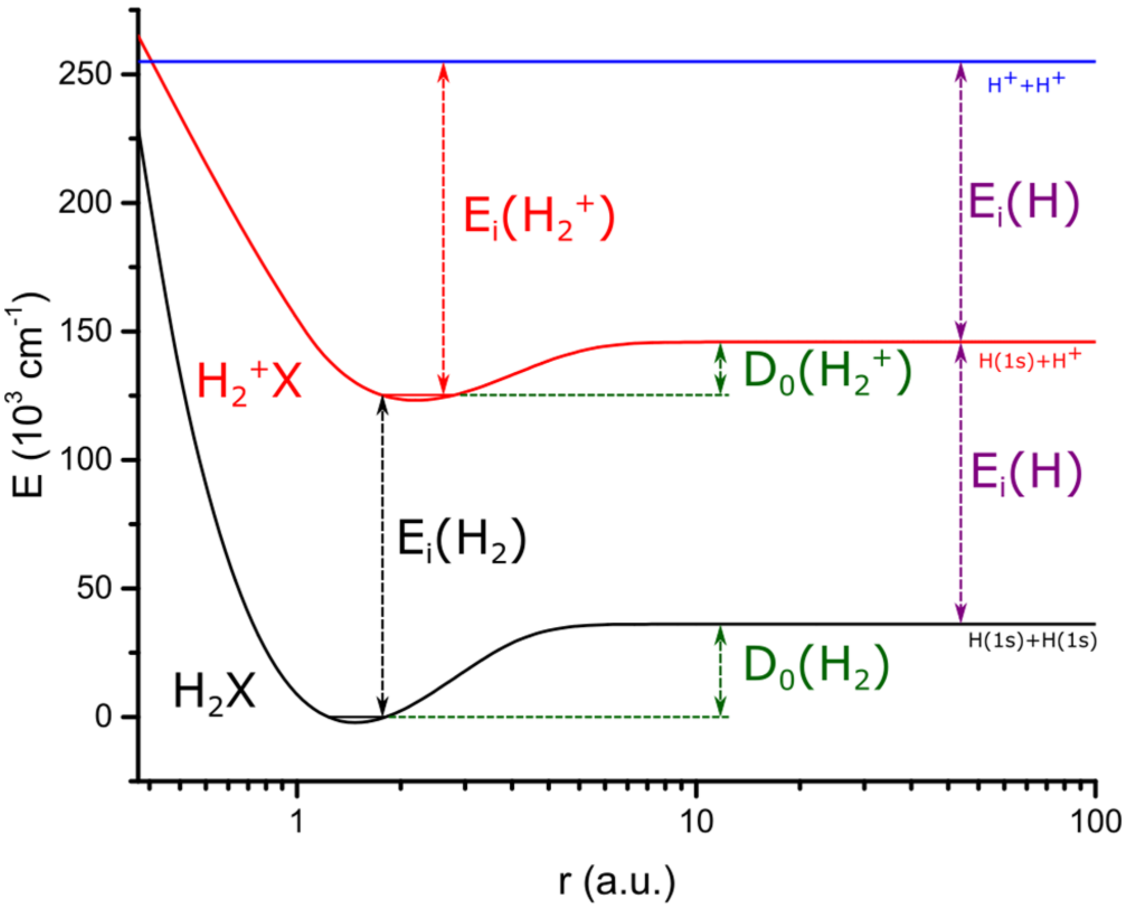
~11MHz

Korobov, PRL 2017

Mohr, RMP 2016

Liu, JCP 2009

Determining D_0(ortho-H_2)



$$D_0(H_2) = E_i(H_2) + E_i(H_2^+) - 2E_i(H)$$

Value	Uncertainty
$E_i(H)$	3 kHz
$E_i(H_2+)$	18 kHz
$E_i(H_2)$	11 MHz
$D_0(H_2)$	11 MHz

Korobov, PRL 2017
Mohr, RMP 2016
Liu, JCP 2009

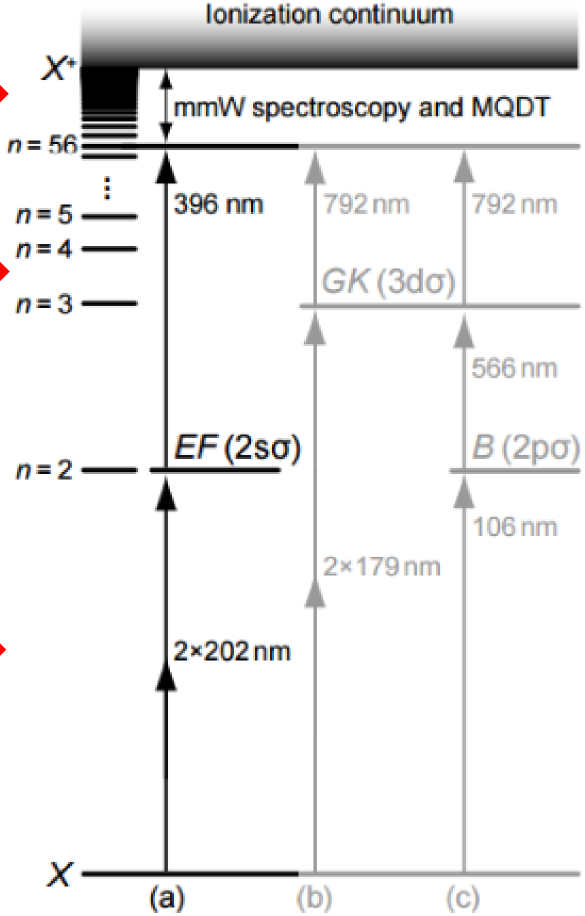
Status of the ionization energy

Liu, JCP 2009
~ 11 MHz

Δf = 100 kHz

Δf = 9 MHz

Δf = 5 MHz



D. Sprecher et al. Faraday Discuss. Chem. Soc. 150, 51 (2011)

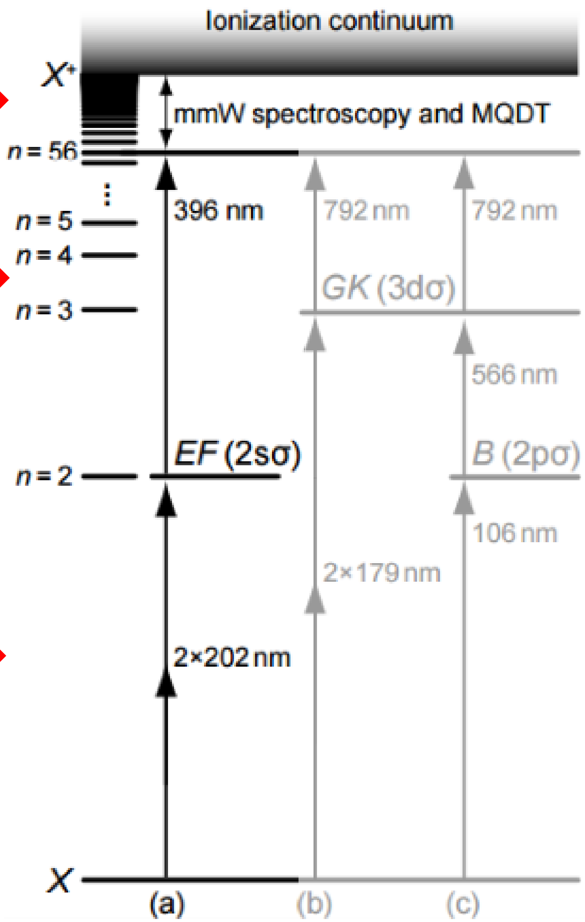
Status of the ionization energy

Altmann, PRL 2018
~ 9 MHz

Δf = 100 kHz

Δf = 9 MHz

Δf = 73 kHz
Δf = 5 MHz



D. Sprecher et al. Faraday Discuss. Chem. Soc. 150, 51 (2011)

Status of the ionization energy

Δf = 100 kHz

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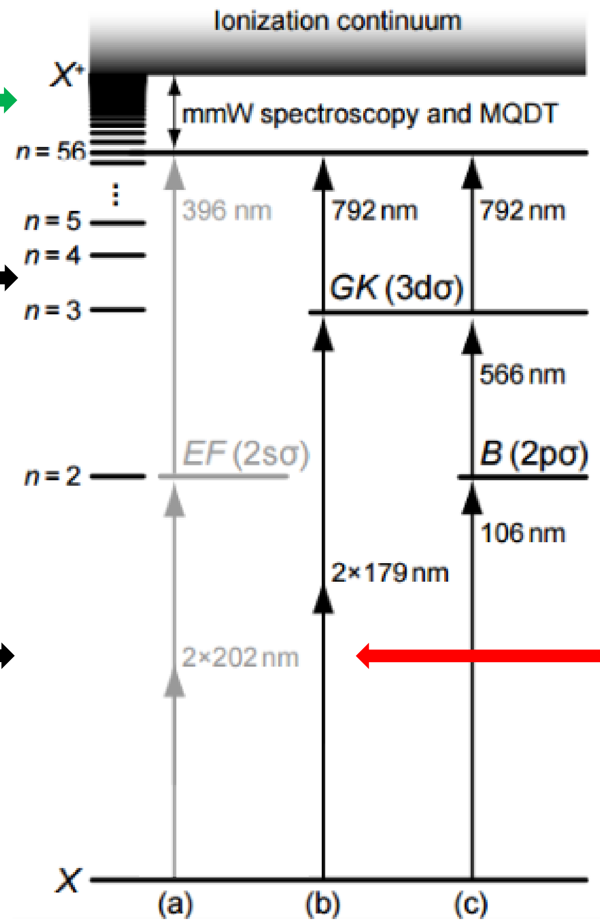


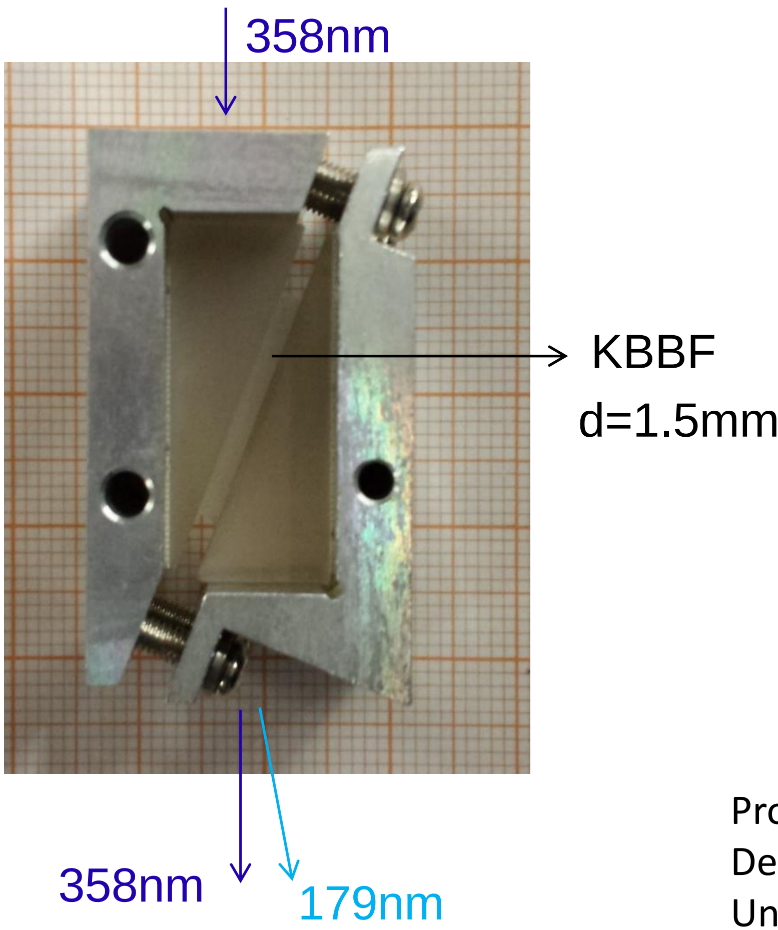
Δf = 1.2 MHz
Sprecher, MP 2013

Δf < 1 MHz?
D_0 < 1 MHz?
179 nm!

D. Sprecher et al. Faraday Discuss. Chem. Soc. 150, 51 (2011)

VUV generation: KBBF

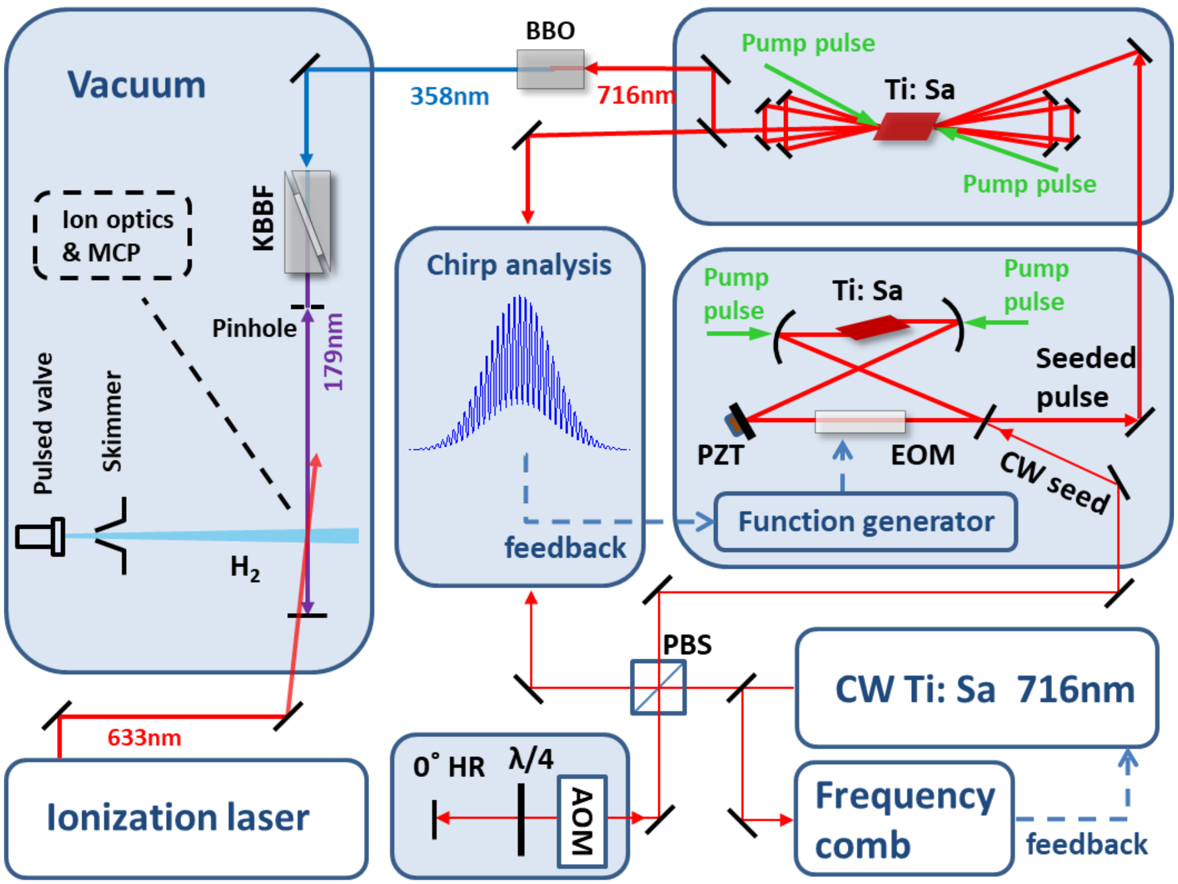
2nd harmonic from 716 nm laser

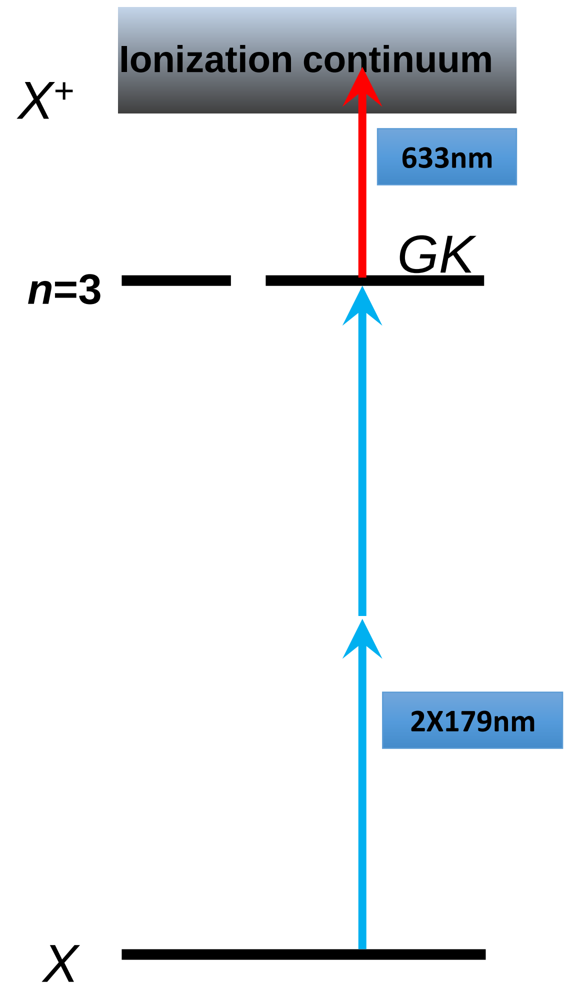


- $KBe_2BO_3F_2$
- Used in Vacuum
- Low efficiency (0.01% to 0.1%)
- Low damage threshold

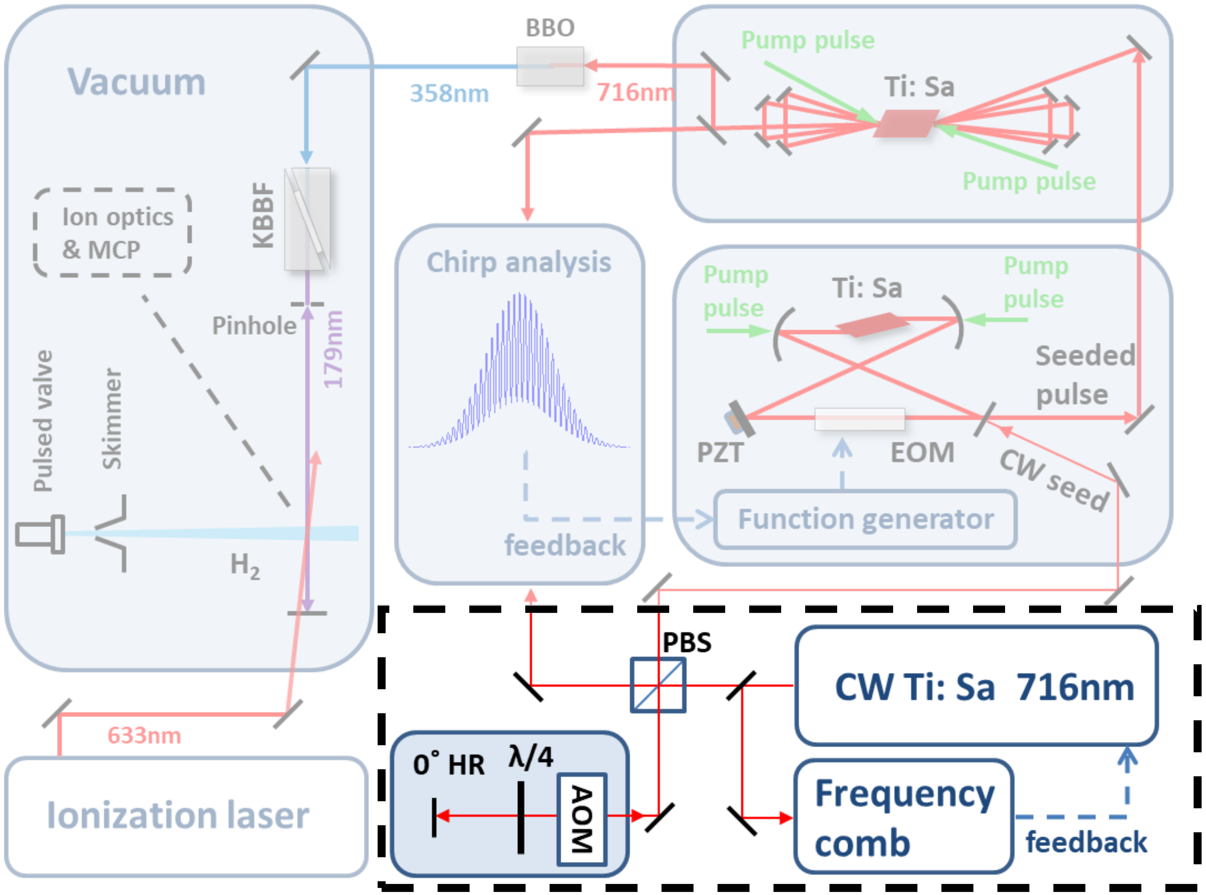
Provided by prof. dr. S. Hu,
Department of Chemical Physics,
University of Science and Technology of China

Experimental setup





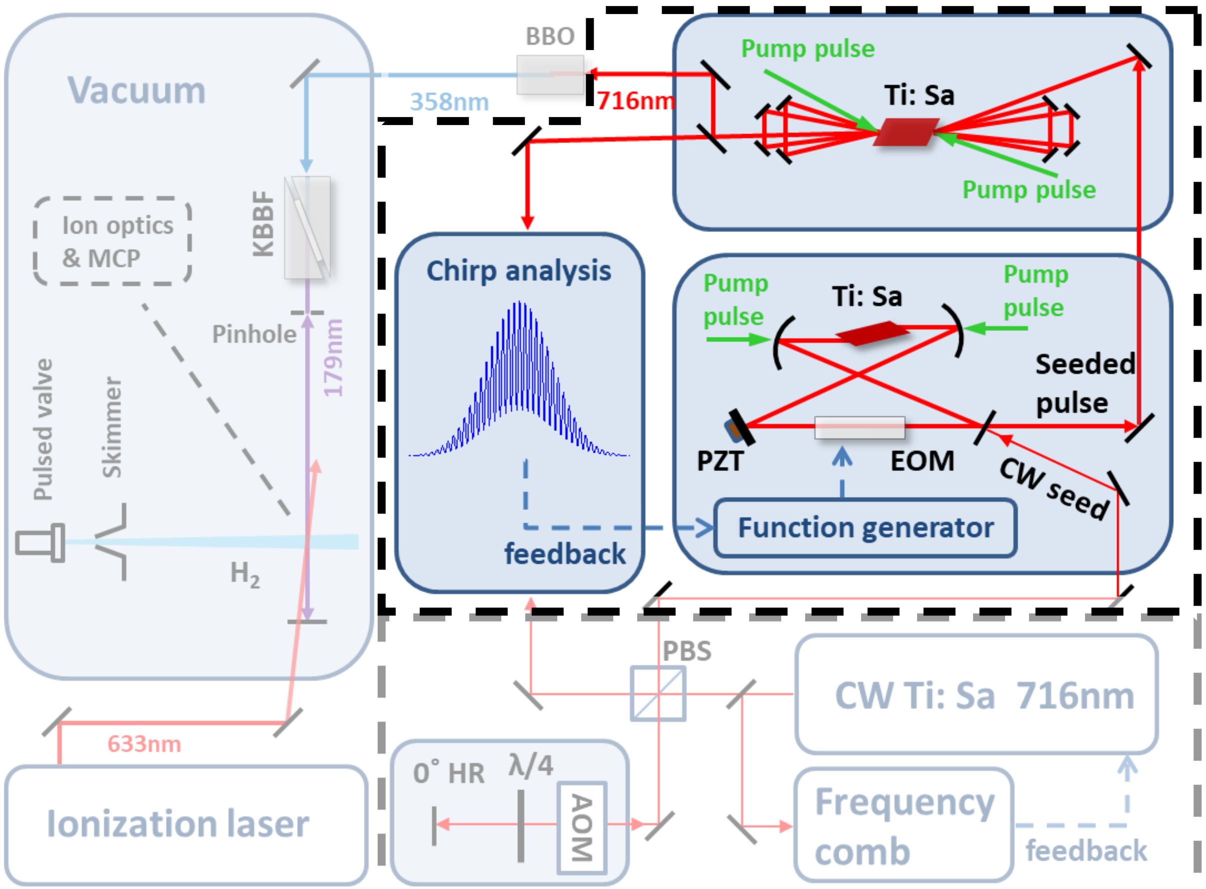
Experimental setup



Frequency accuracy < 10^{-12}

Experimental setup

High power, high accuracy



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Experimental setup

VUV generation

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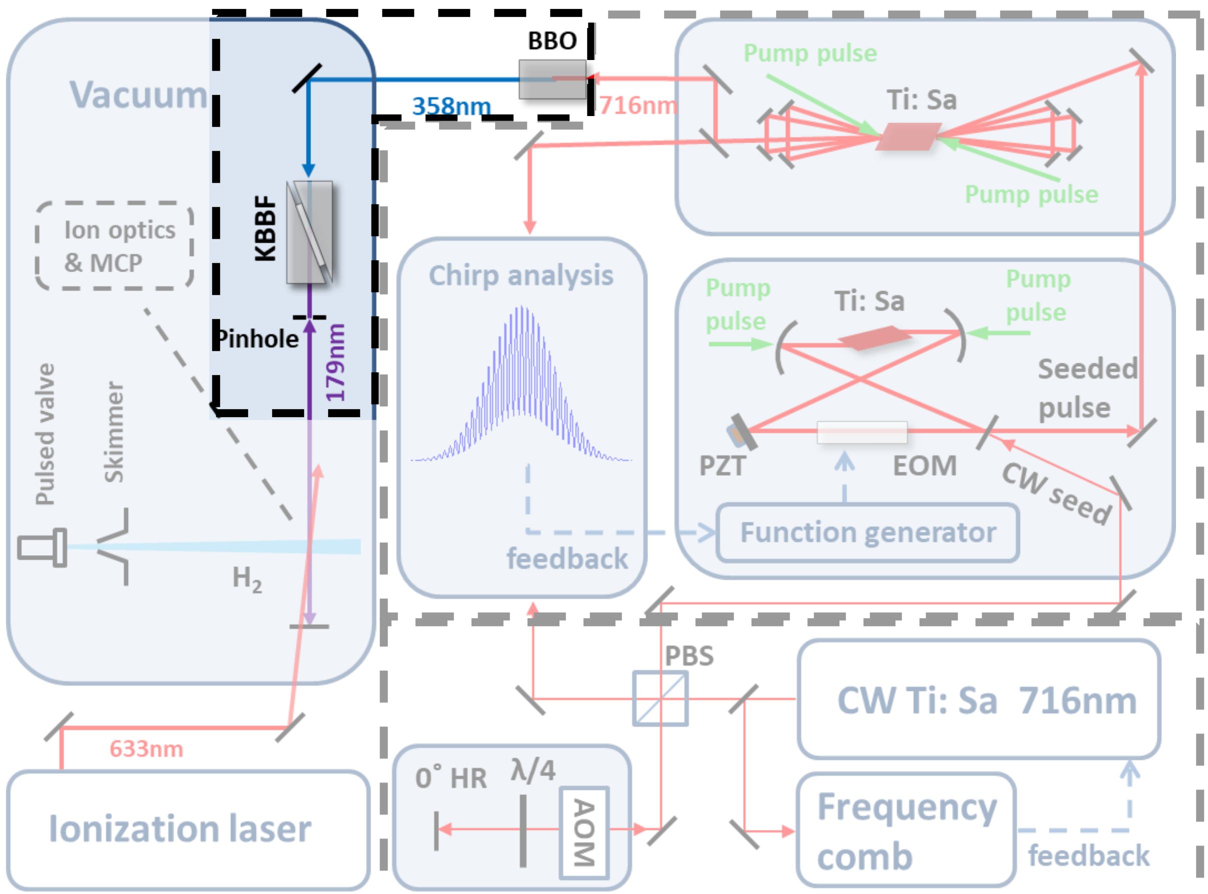
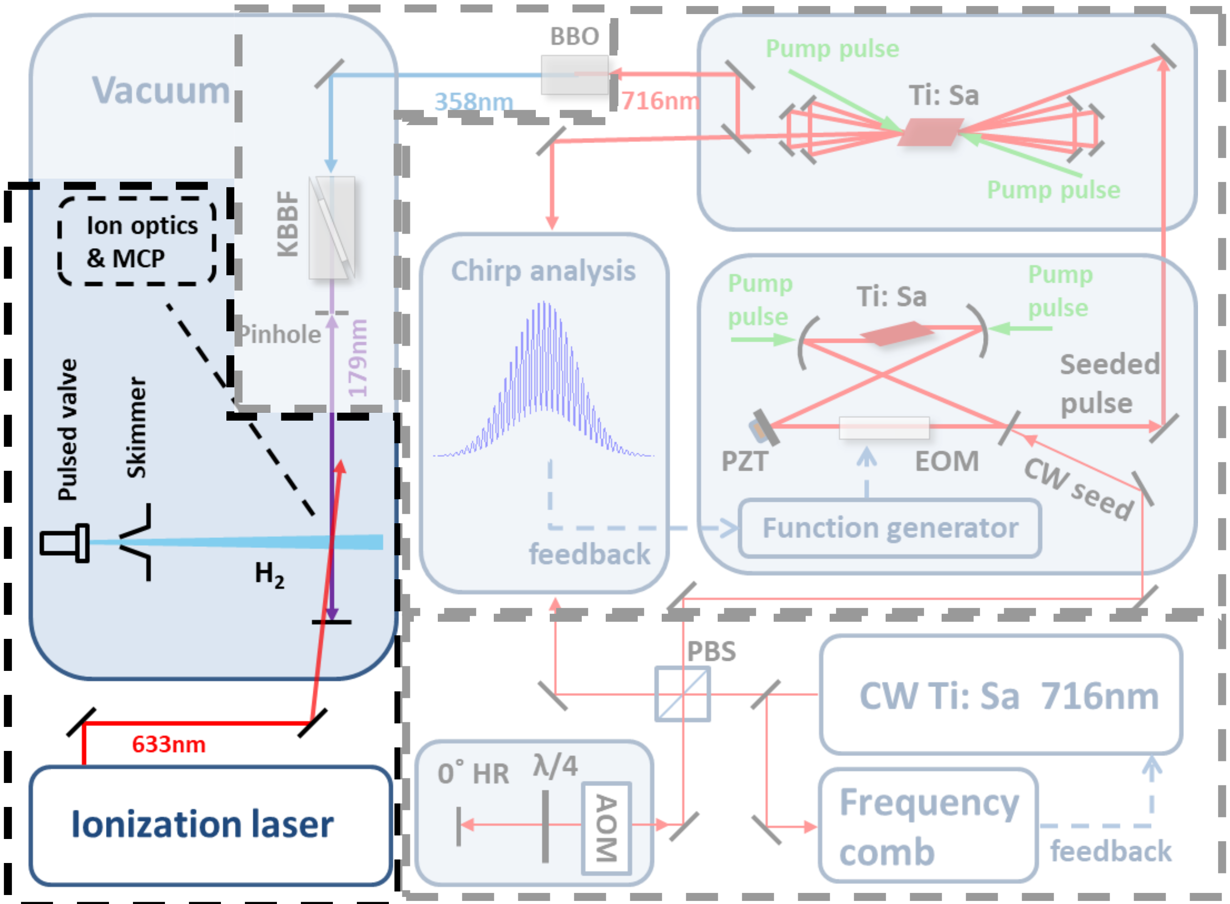


Frequency accuracy < 10^{-12}

Experimental setup

VUV generation

High power, high accuracy

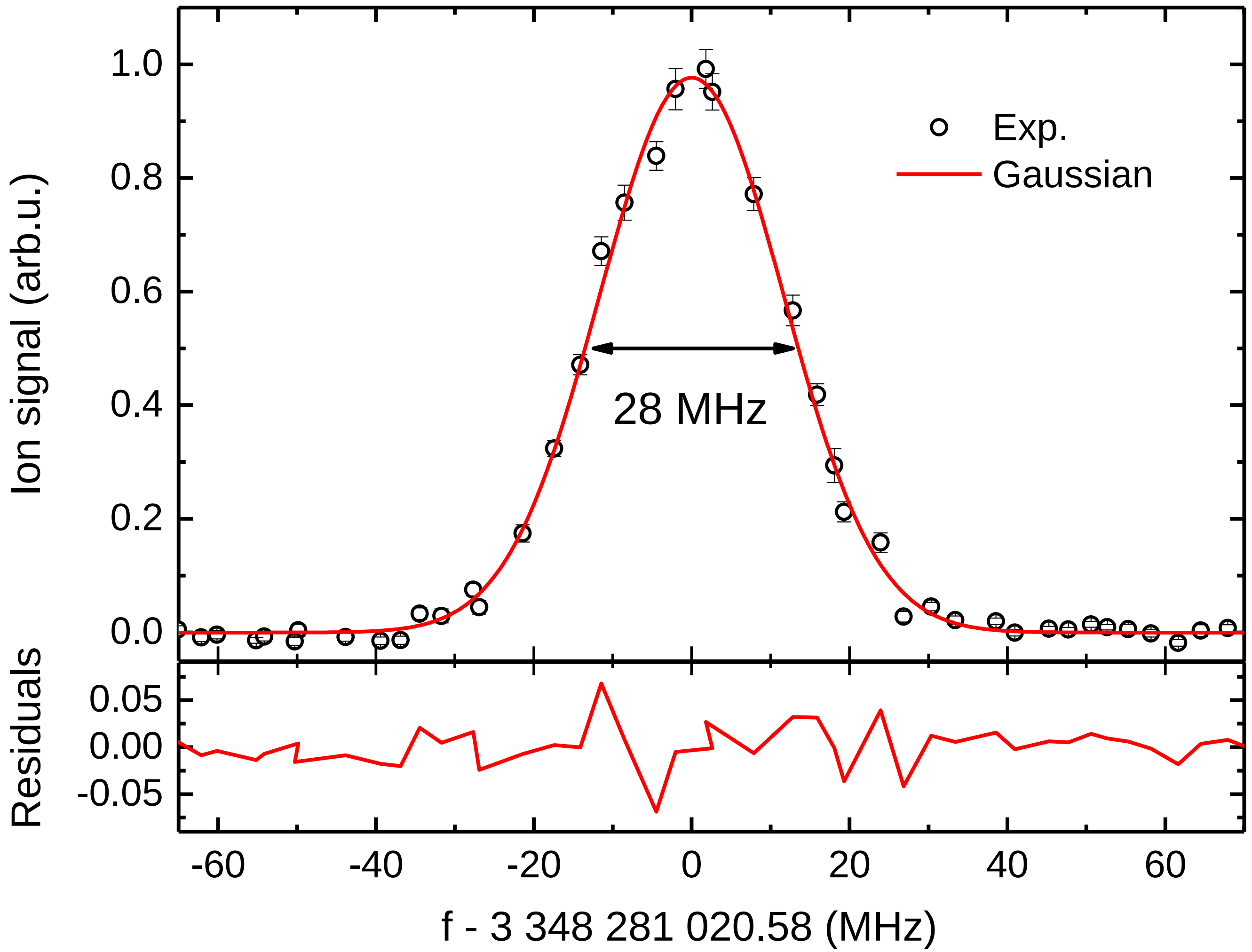


Ionize and detect

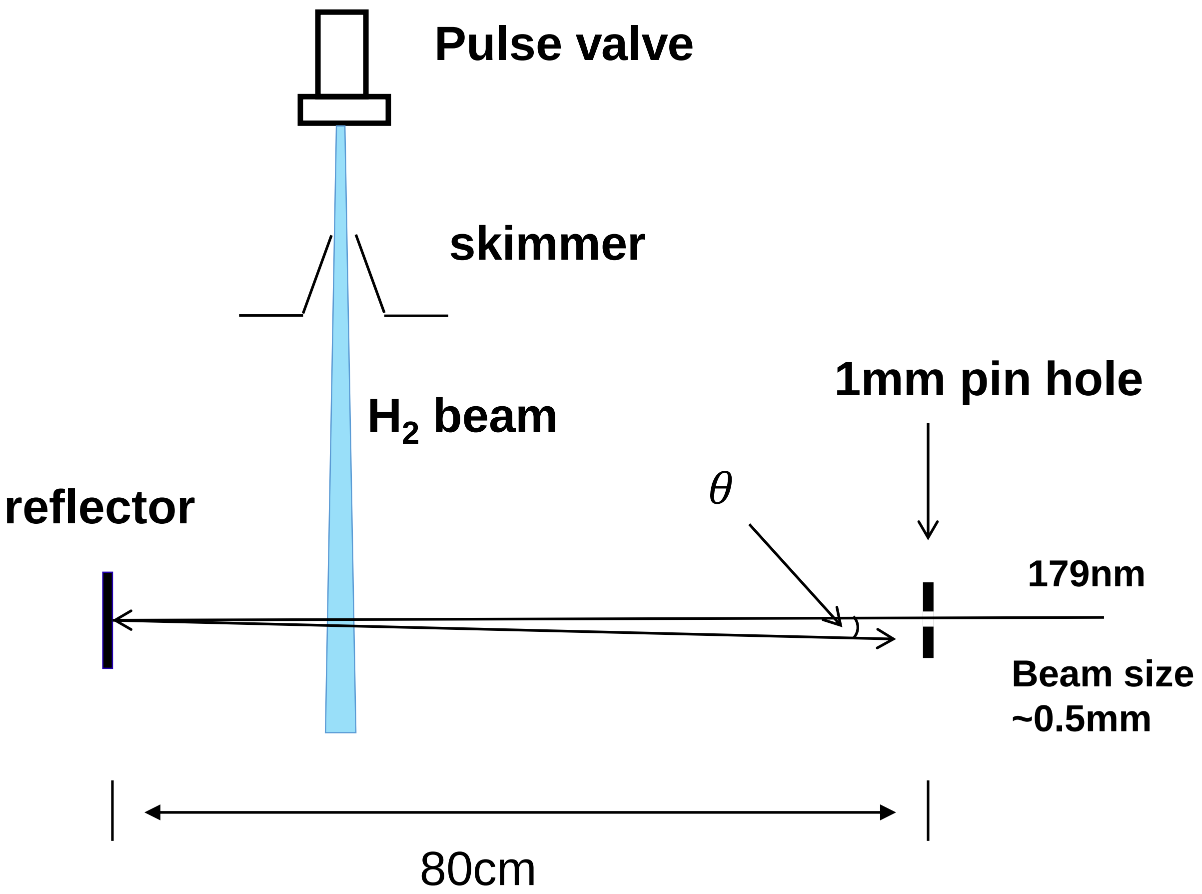
Frequency accuracy < 10^{-12}

Scan of GK-X Q(1) in H_2

GK(v=1, N=1) - X(v=0, N=1), chirp compensated shot-by-shot.



Systematic check: Residual Doppler



$$\frac{\delta f}{f} = \frac{v}{c}\sin\frac{\theta}{2}$$

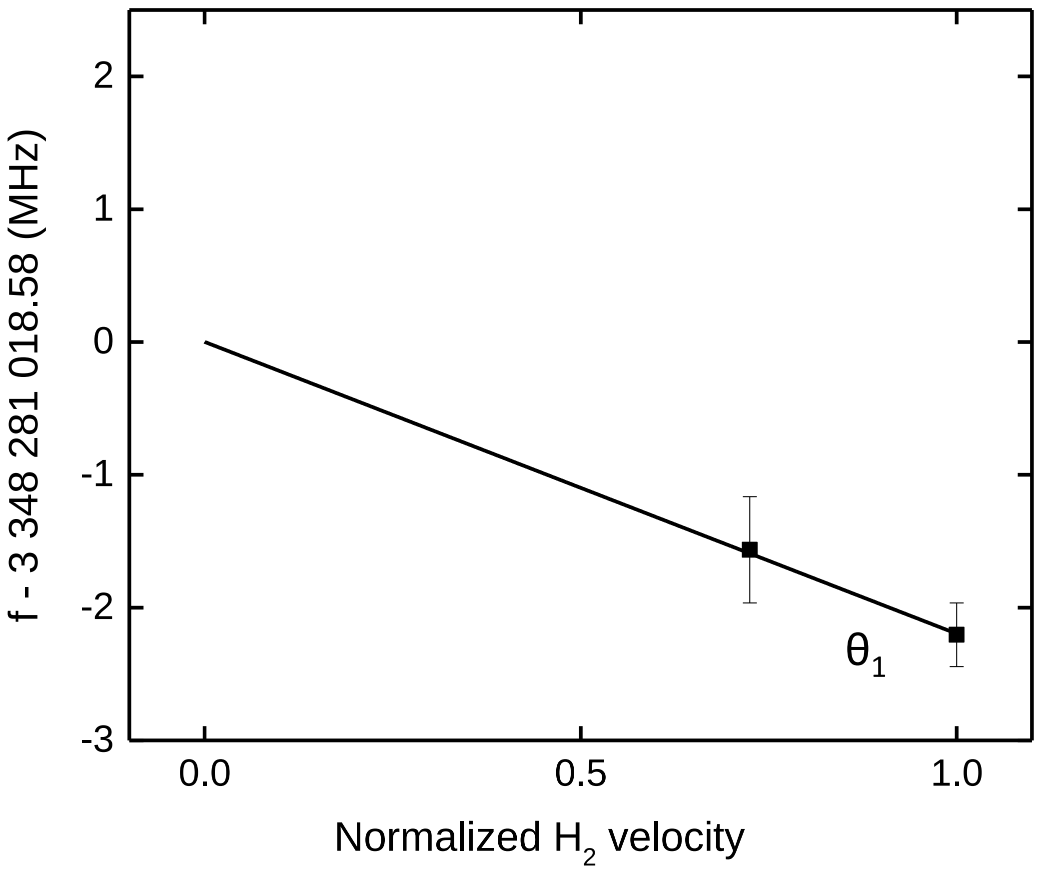
v = 2900(300) m/s

θ < 0.3mrad

→ δf < 3MHz

Systematic check: Residual Doppler

$$v_{norm} = \frac{v_{mix}}{v_{pure}} = \sqrt{\frac{m_{H_2}}{n_{H_2} \cdot m_{H_2} + n_{Ne} \cdot m_{Ne}}}$$



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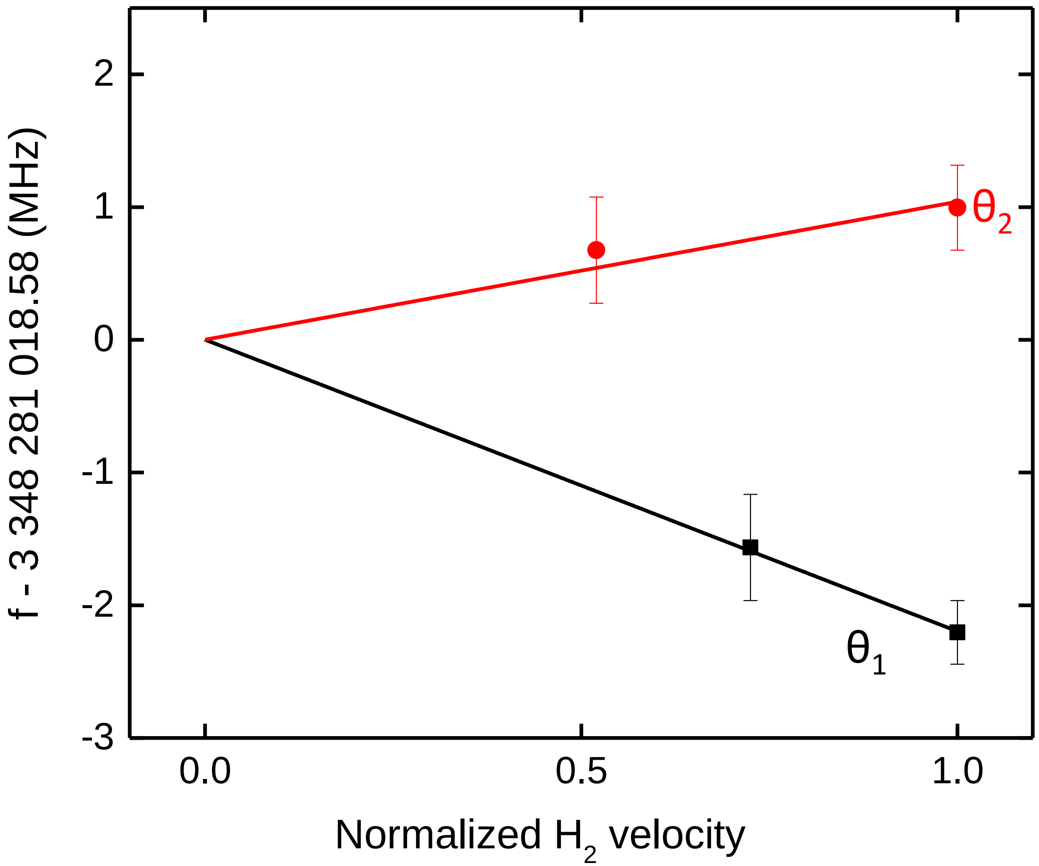
Changing θ



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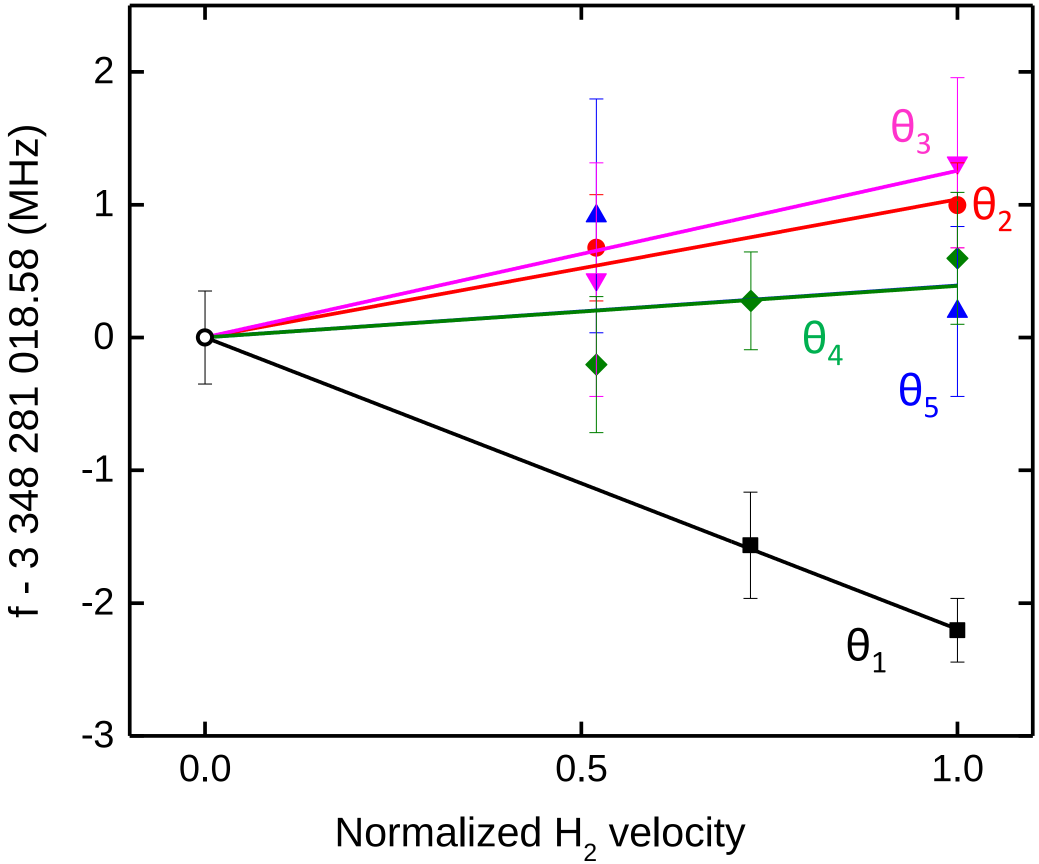
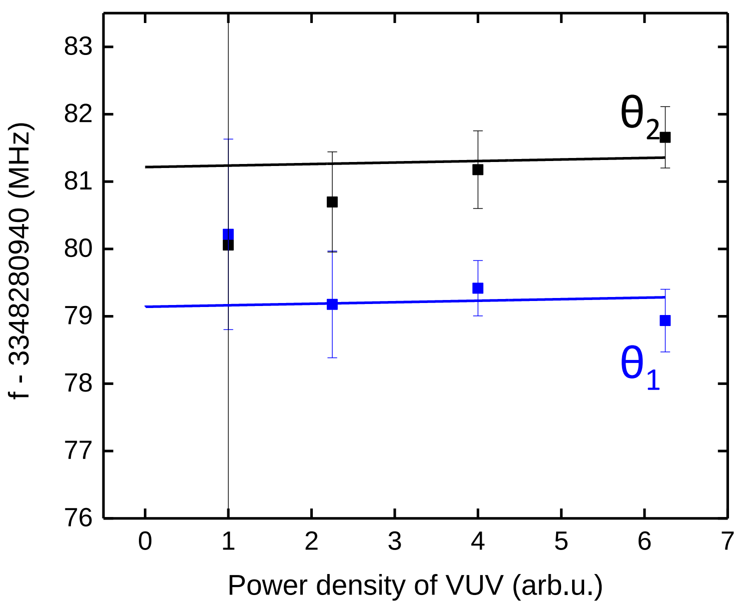
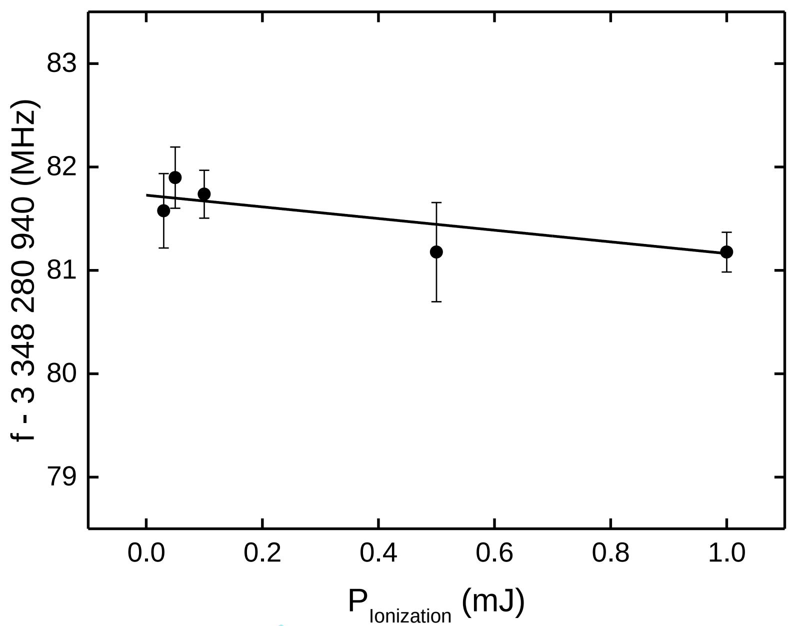
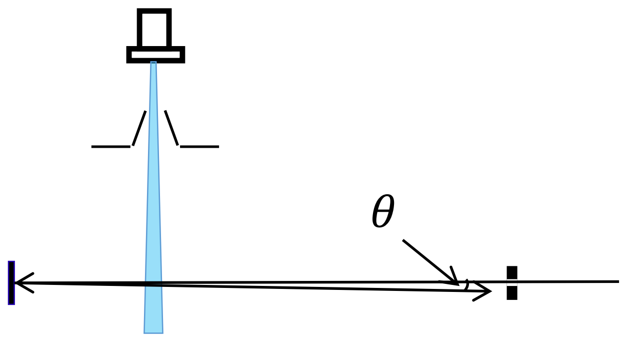
Changing θ

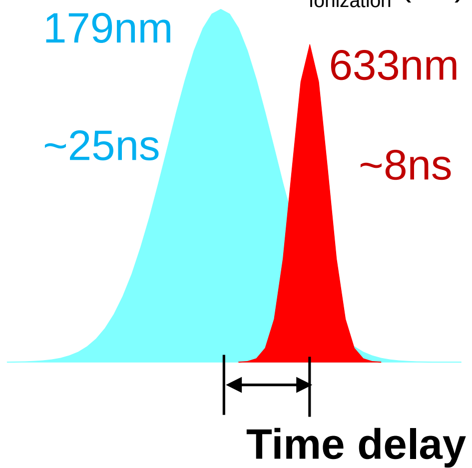


Systematic check: AC Stark

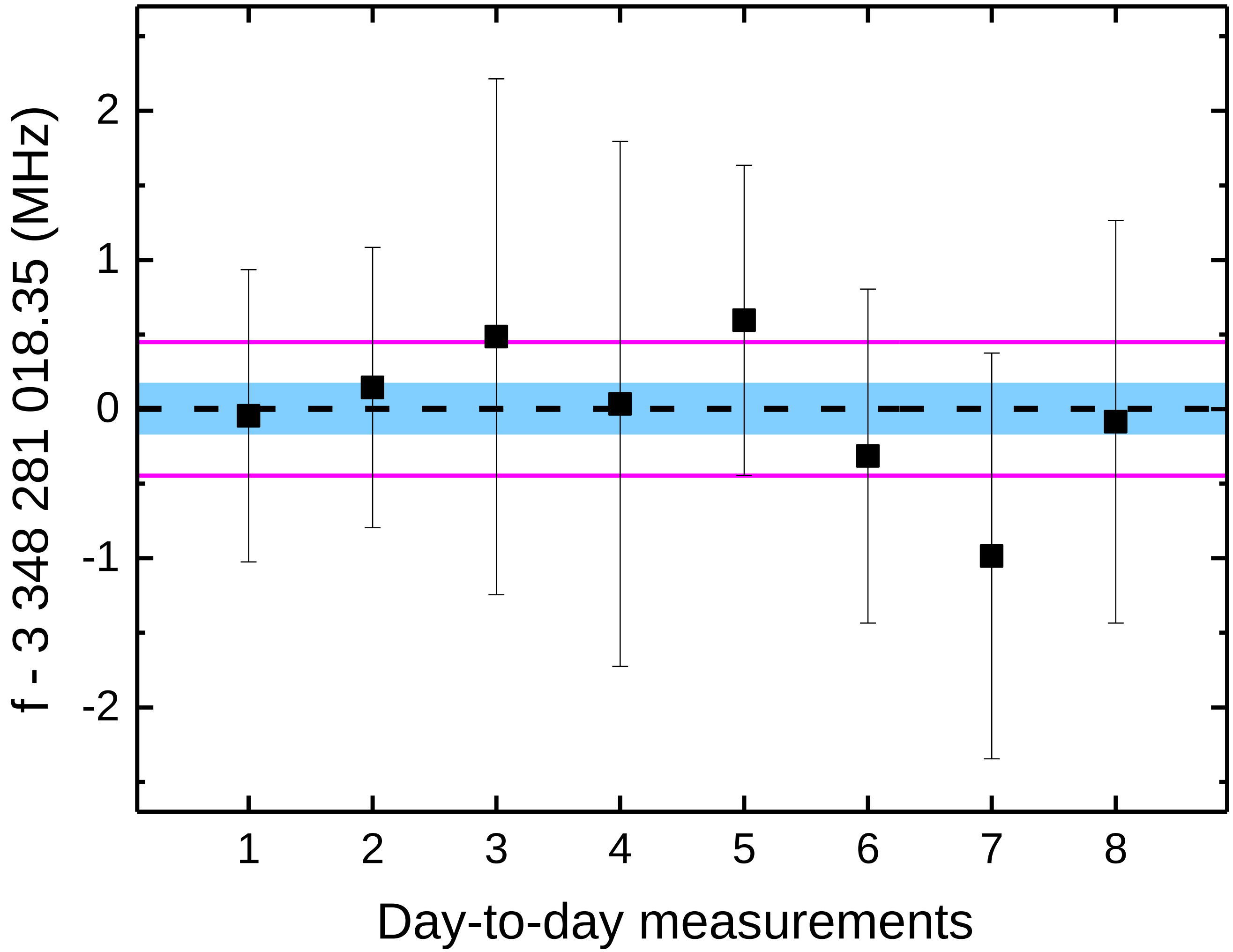








Statistical uncertainty



Error budget

For GK(v=1, N=1) - X(v=0, N=1) (ortho-H_2)

Measured frequency	3348281018.58(49) MHz	
Effect	Correction	Uncertainty
Chirp		$(<490\text{kHz})_{stat}$
Residual 1^{st} order Doppler		350kHz
2^{nd} order Doppler		<30kHz
AC Stark red	-40kHz	90kHz
AC Stark VUV	-190kHz	200kHz
Hyperfine structure		<100kHz
Total systematic uncertainty		426kHz
Final frequency	$3348281018.35(49)_{stat}(43)_{sys}$ MHz	

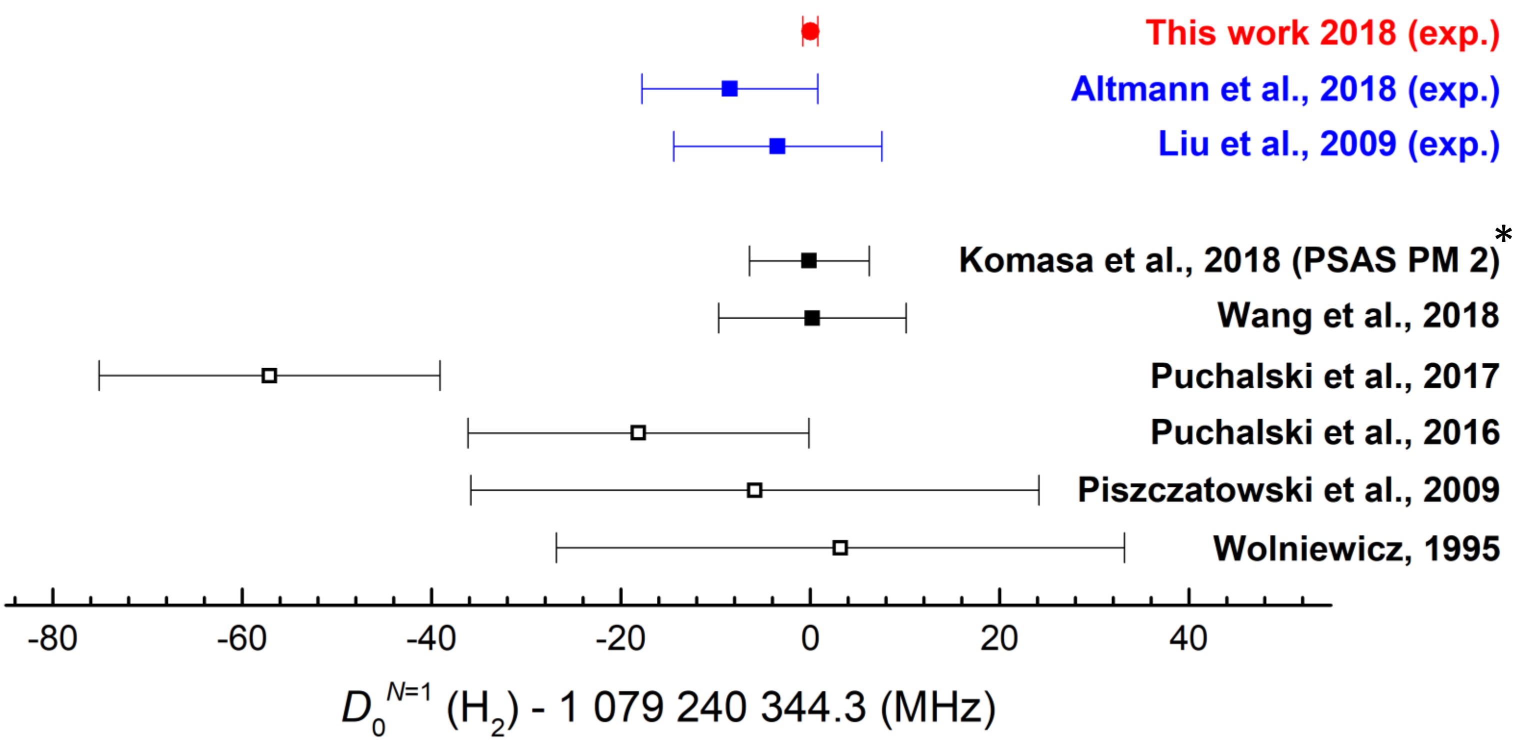
Error budget

For GK(v=1, N=1) - X(v=0, N=1) (ortho-H_2)

Measured frequency	3348281018.58(49) MHz	
Effect	Correction	Uncertainty
Chirp		(<490kHz)
Total systematic uncertainty		426kHz
Final frequency	$3348281018.35(49)_{stat}(43)_{sys}$ MHz	

Total accuracy is 650 kHz: 2×10^{-10} for GK-X

Dissociation Energy of ortho-H_2



This work:

"Dissociation energy of the hydrogen molecule at 10^{-9} accuracy"

C. Cheng, J. Hussels, ..., N. Hölsch, et al. accepted *Phys. Rev. Lett.* (2018), arXiv: 1804.11143v1.

Conclusion & Outlook

- GK-X Q(1) has been measured directly for the first time. An accuracy of 0.65 MHz was achieved.
- D_0(ortho-H_2) determination has been improved by an order of magnitude to 0.8 MHz

- Now: measure GK-X S(0) to improve D_0(para-H_2)
- Future: Similar measurement for HD and D_2

Thanks to:

LaserLaB, VU Amsterdam:

Dr. Cunfeng Cheng
Dr. Ming Li Niu
Dr. Edcel Salumbides
Dr. Rick Bethlem
Prof. dr. Kjeld Eikema
Prof. dr. Wim Ubachs

Rob Kortekaas (Technician)

ETH, Zürich:

Maximilian Beyer
Nicolas Hölsch (Next speaker)
Prof. dr. Frédéric Merkt

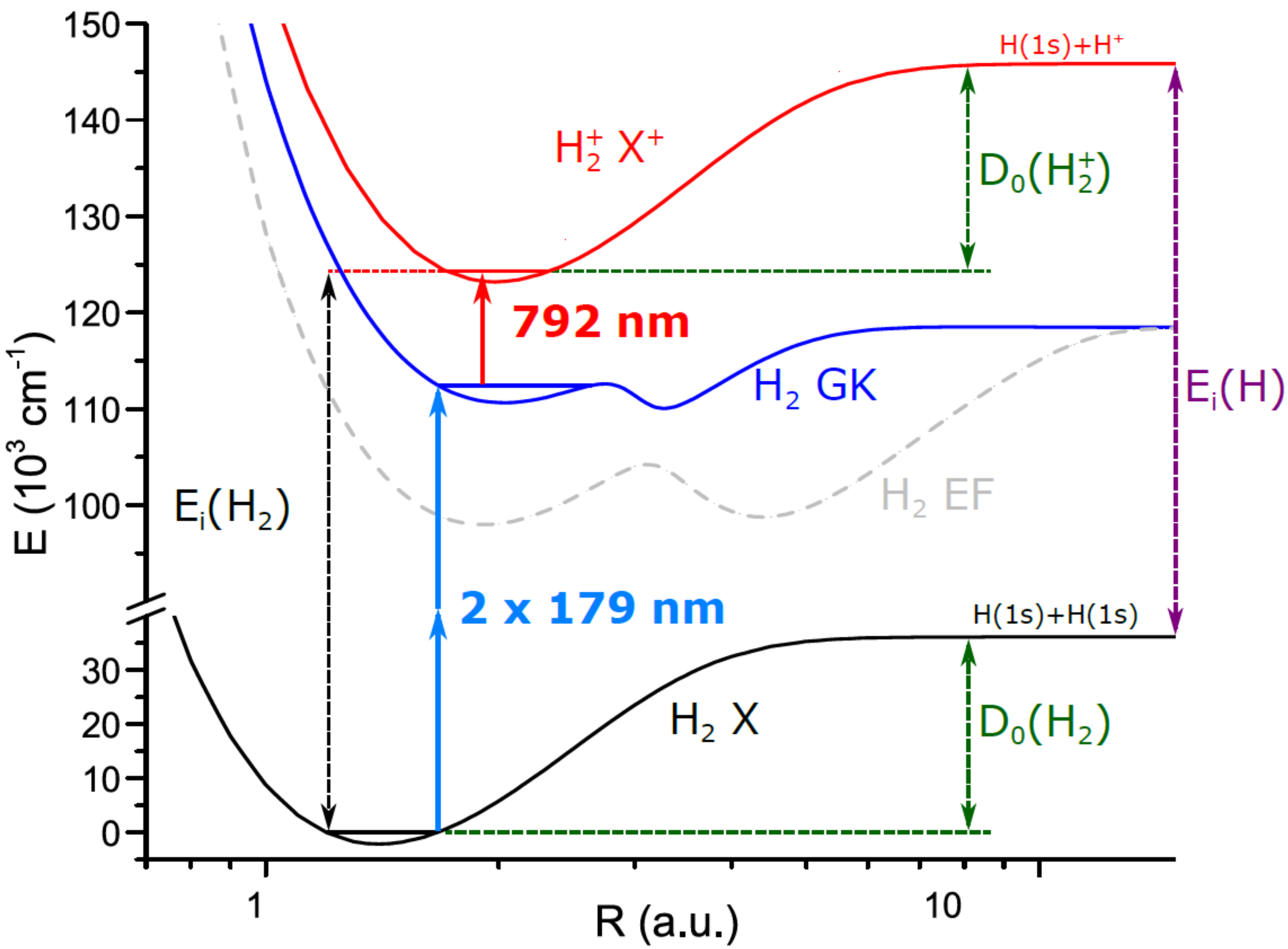
CNRS, Orsay:

Prof. dr. Christian Jungen

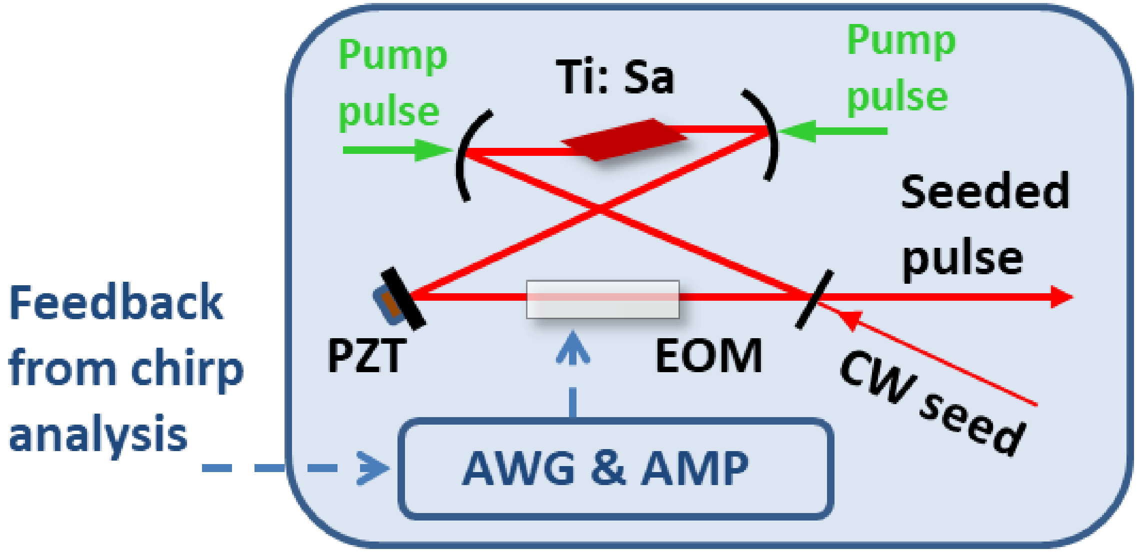
USTC, Hefei:

Lei-Gang Tao
Prof. dr. Shui-Ming Hu

H_2 Potentials



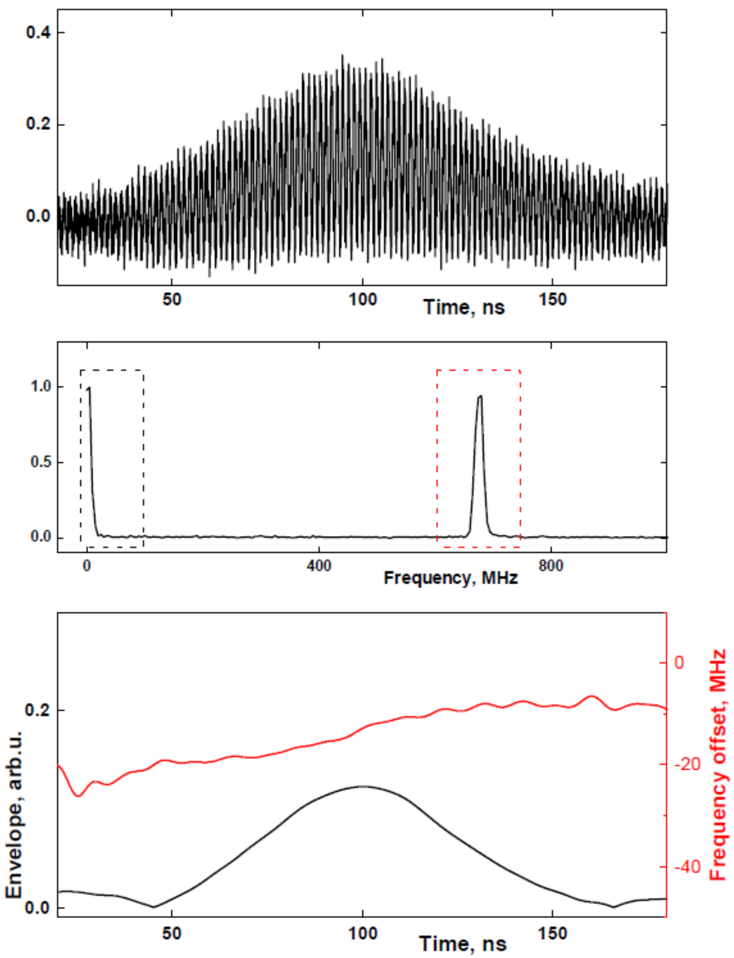
Anti-Chirp



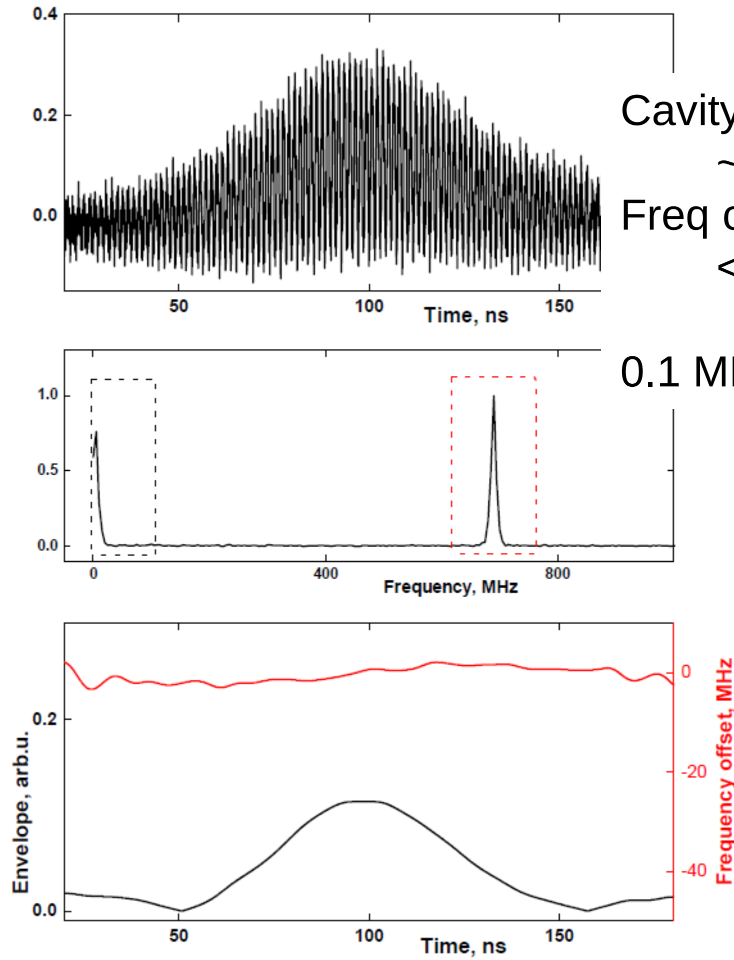
- Sensitive to temperature and stress
- Arbitary function generator to drive
- Amplified to max 150V

Chirp measurement

w/o anti-chirp



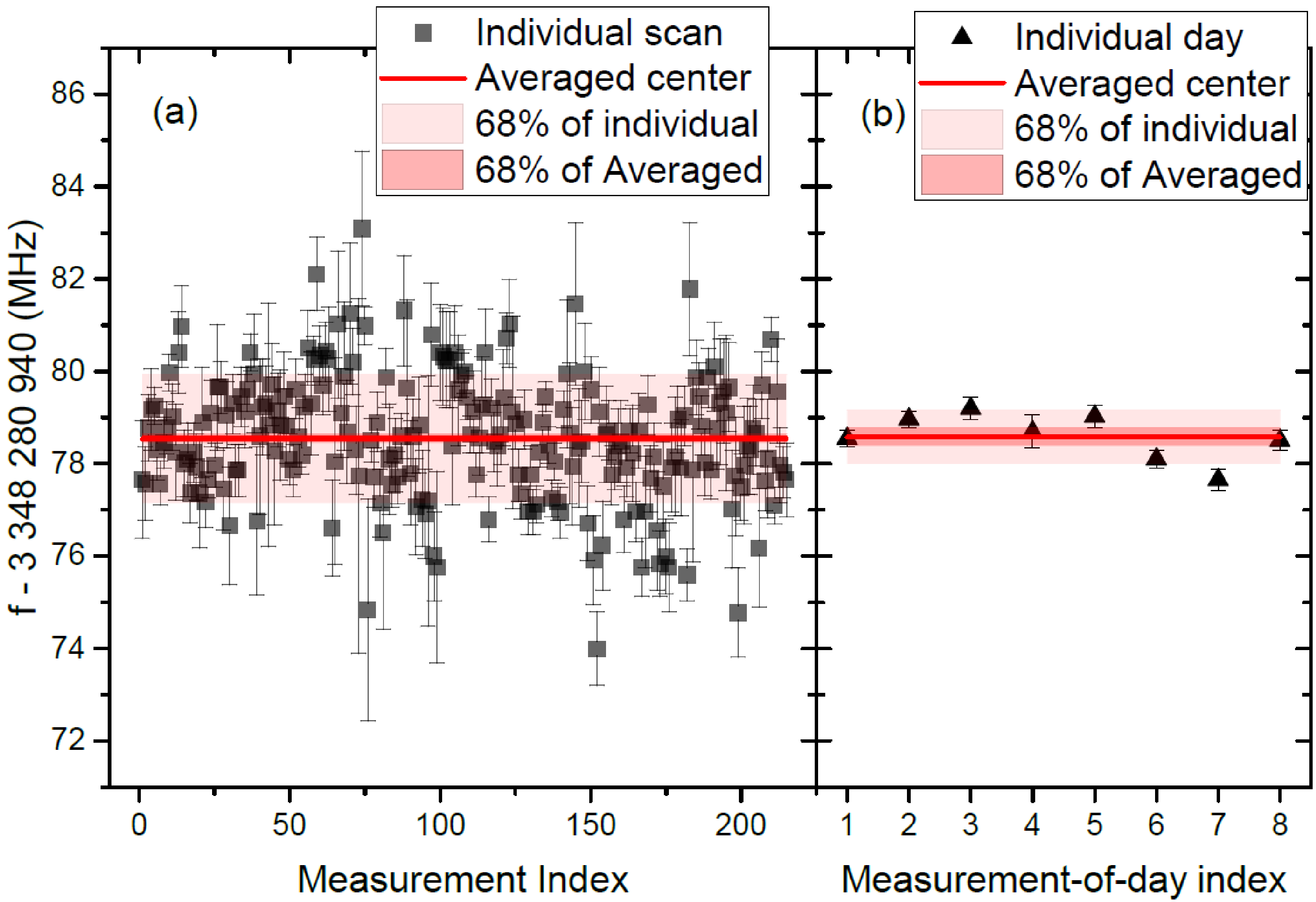
with anti-chirp



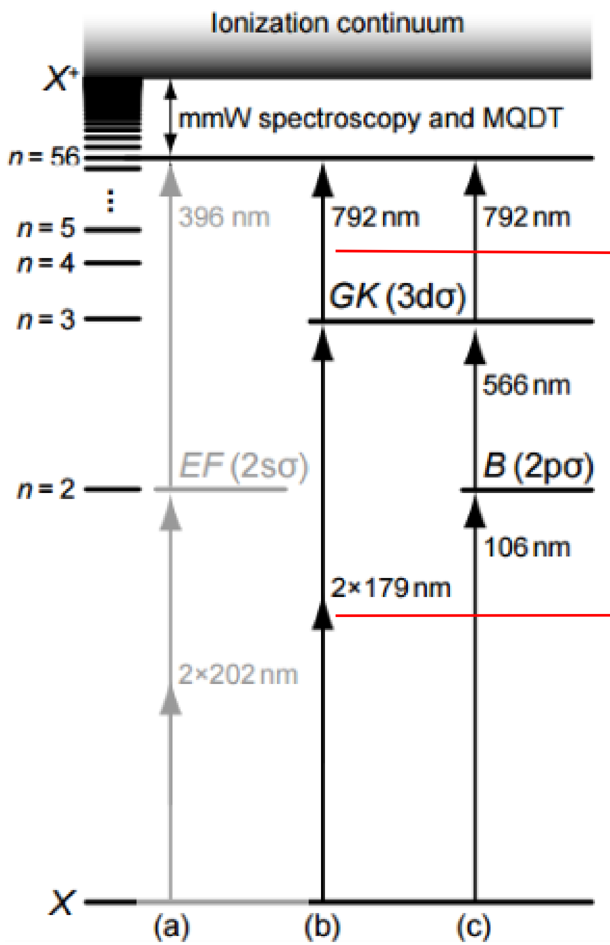
Cavity pulling:
~ 0 MHz
Freq chirp:
< 1.5 MHz/50 ns

0.1 MHz uncertainty

Residual systematical and statistical



Ionization energy of H_2 at 750 kHz



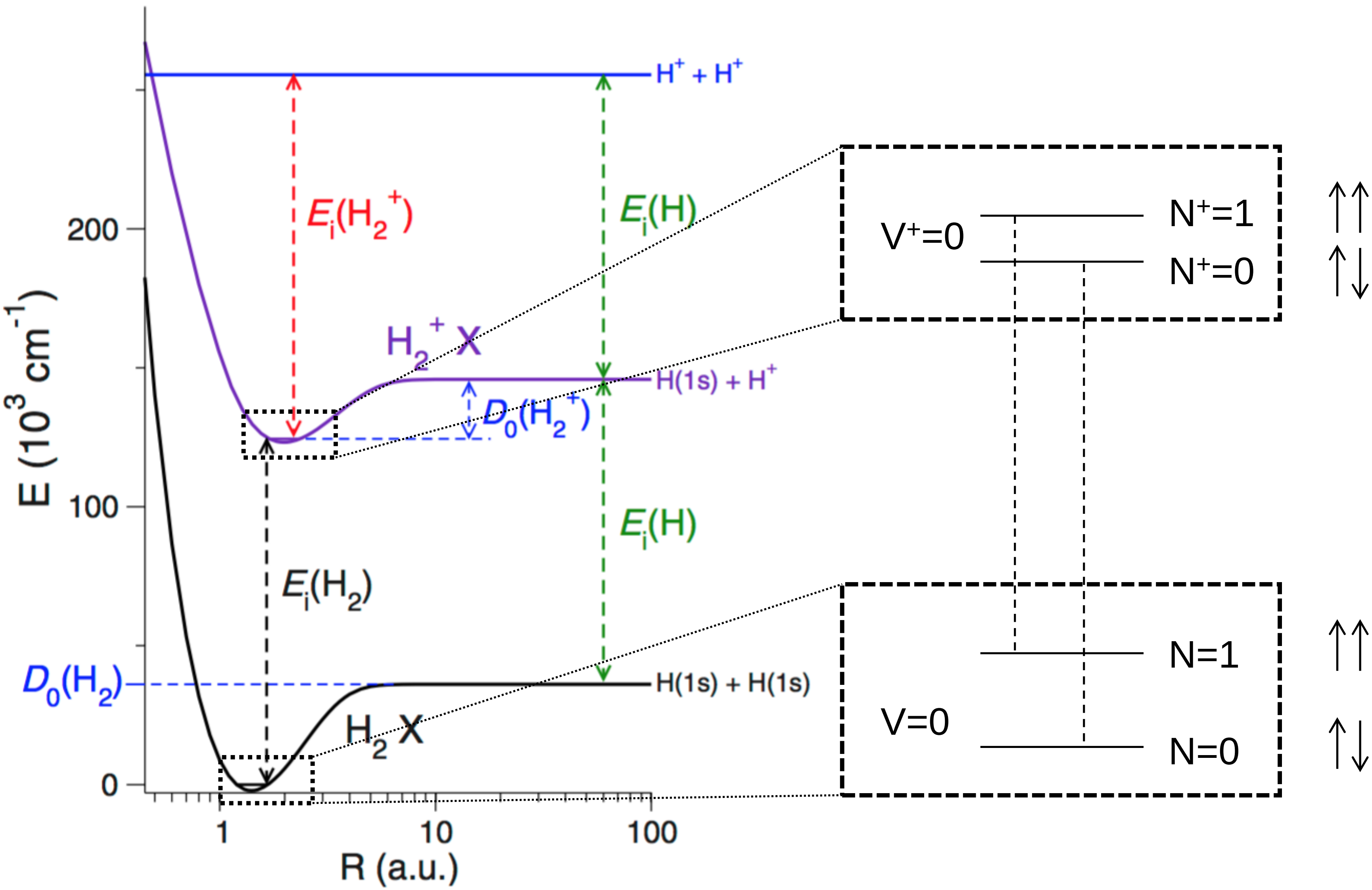
Δf = 100 kHz

360kHz

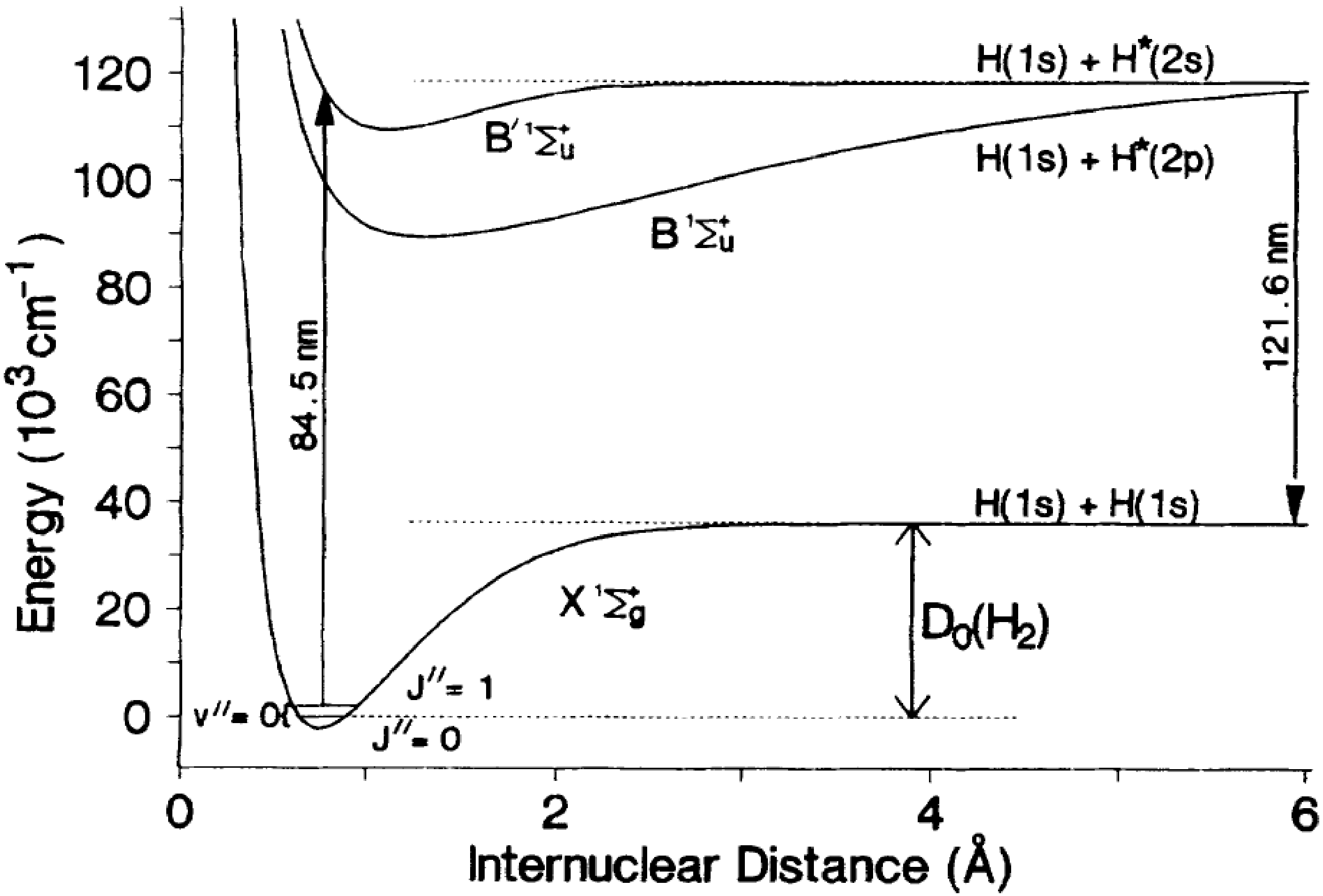
See next talk:
Improvement of the Dissociation Energy of the Hydrogen Molecule (Part Two) by N. Hölsch

650 kHz

Para-H_2 and ortho-H_2



Direct measurement of D_0



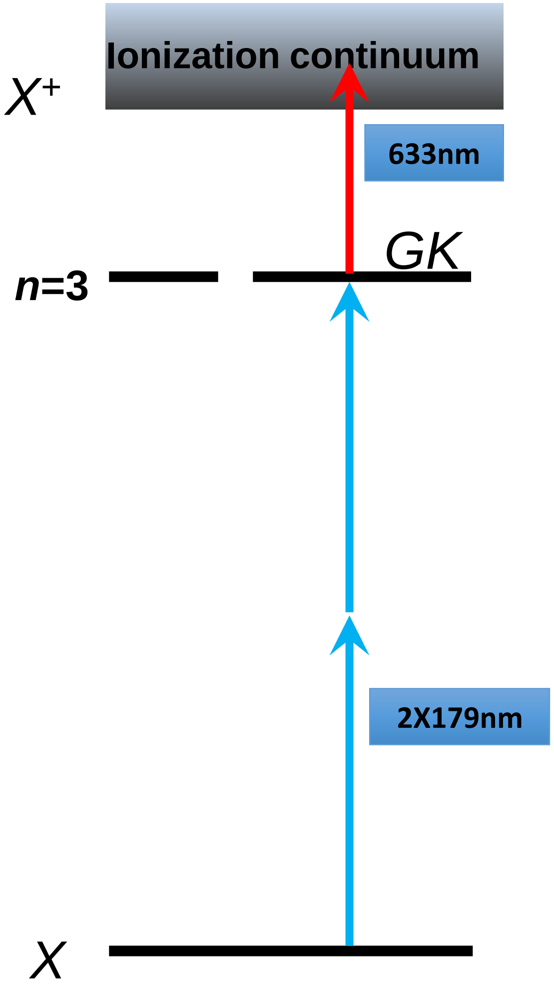
~0.01cm^{-1}

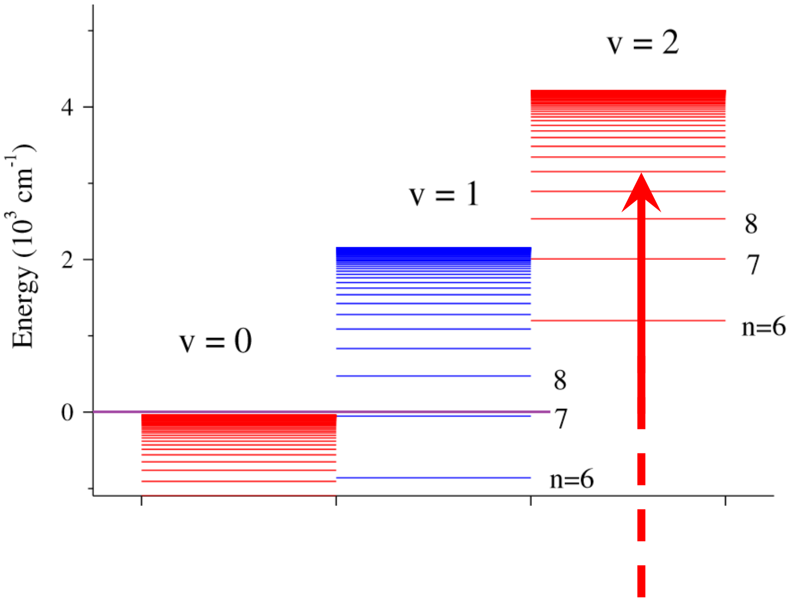
Zhang, PRL 2004

1 cm^{-1} = 1.24X10^{-4} eV= 30 GHz

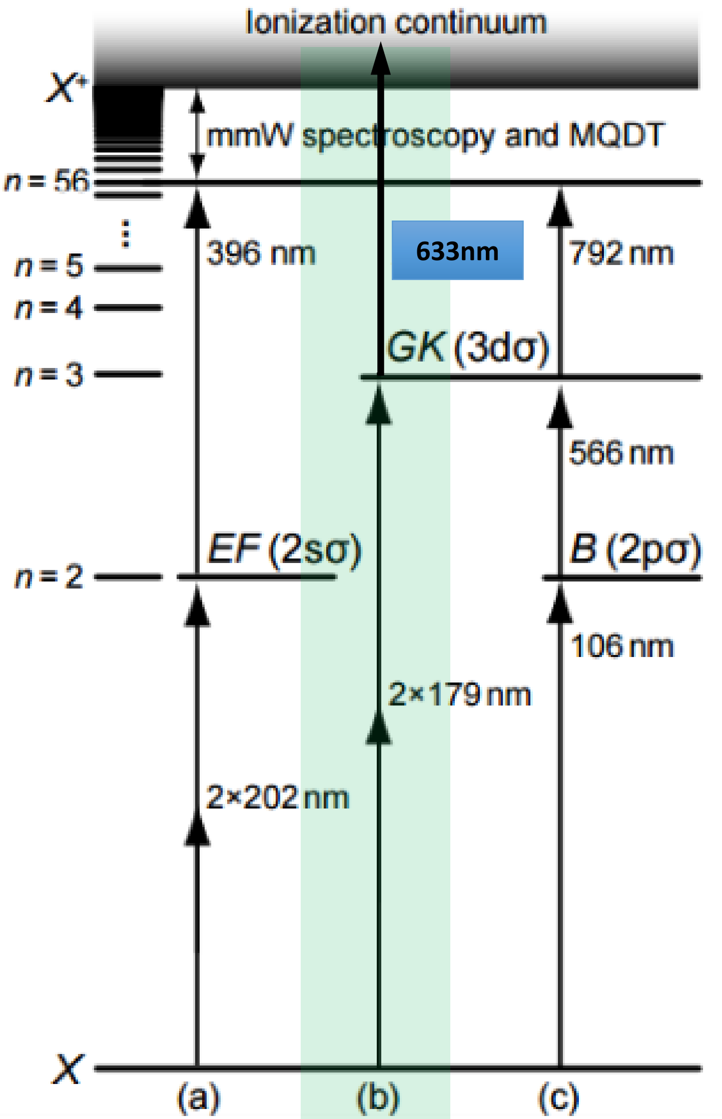
Balakrishnan, PRL 1992

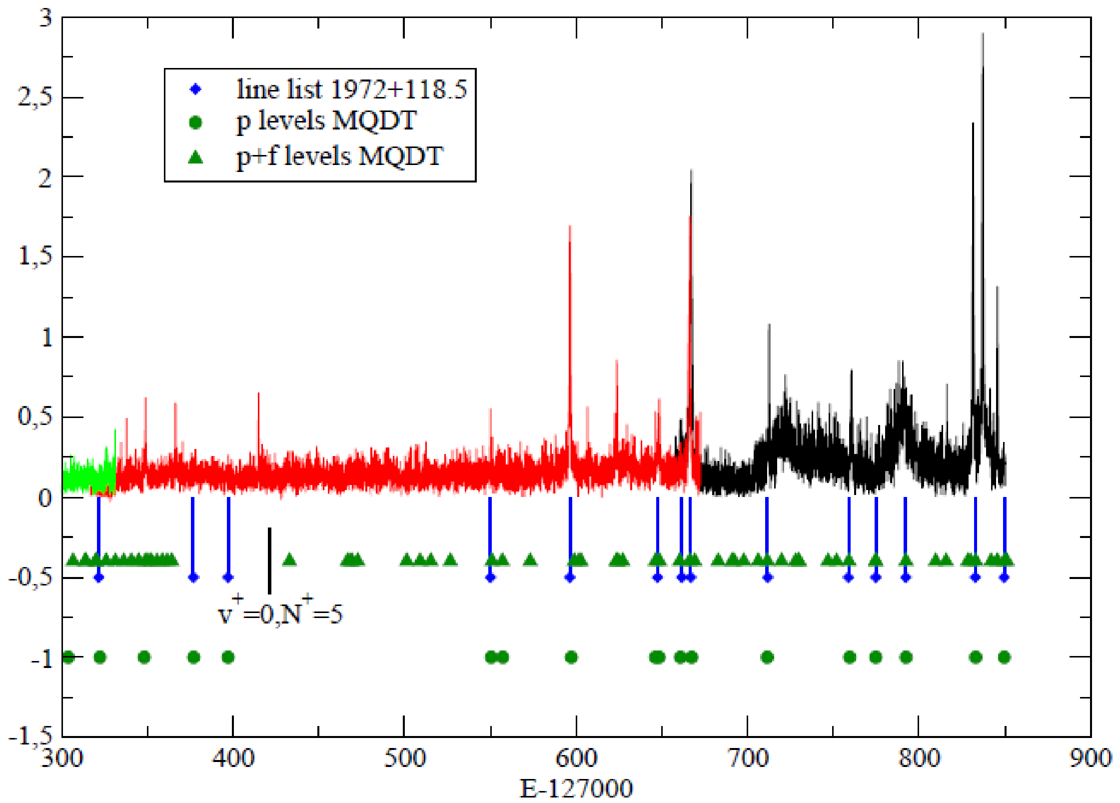
Measurement of GK-X



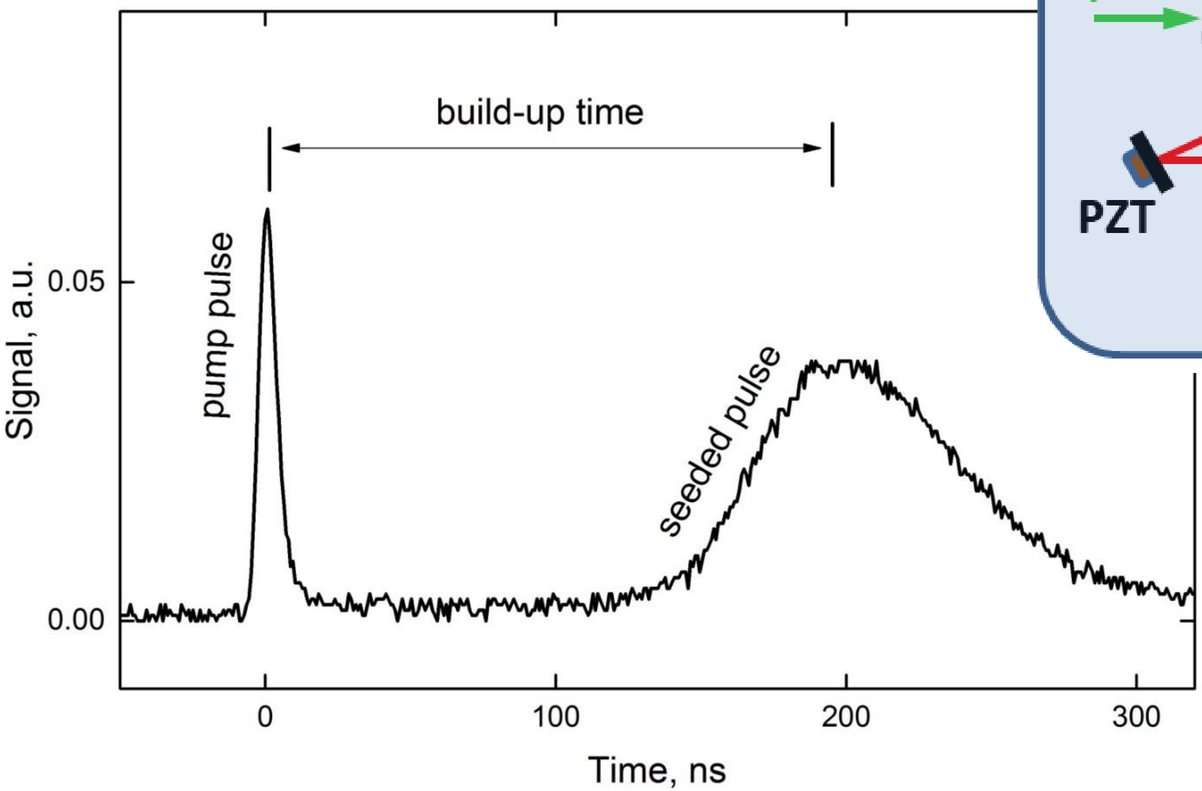


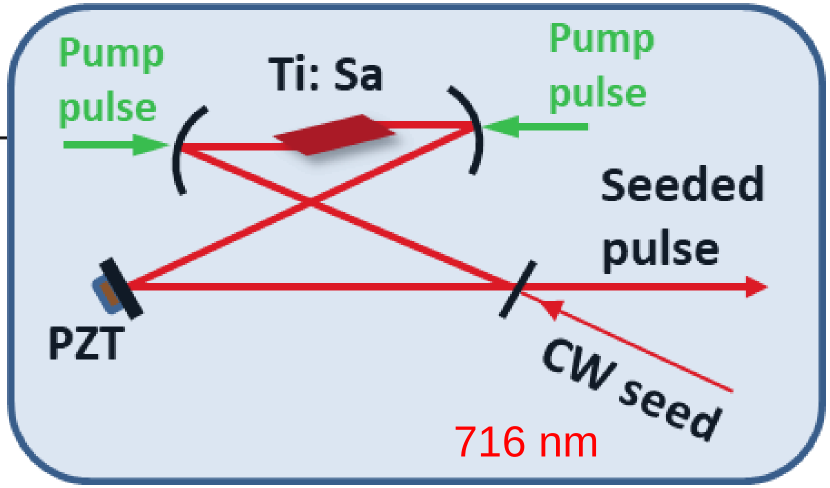
Transition measurement on H_2





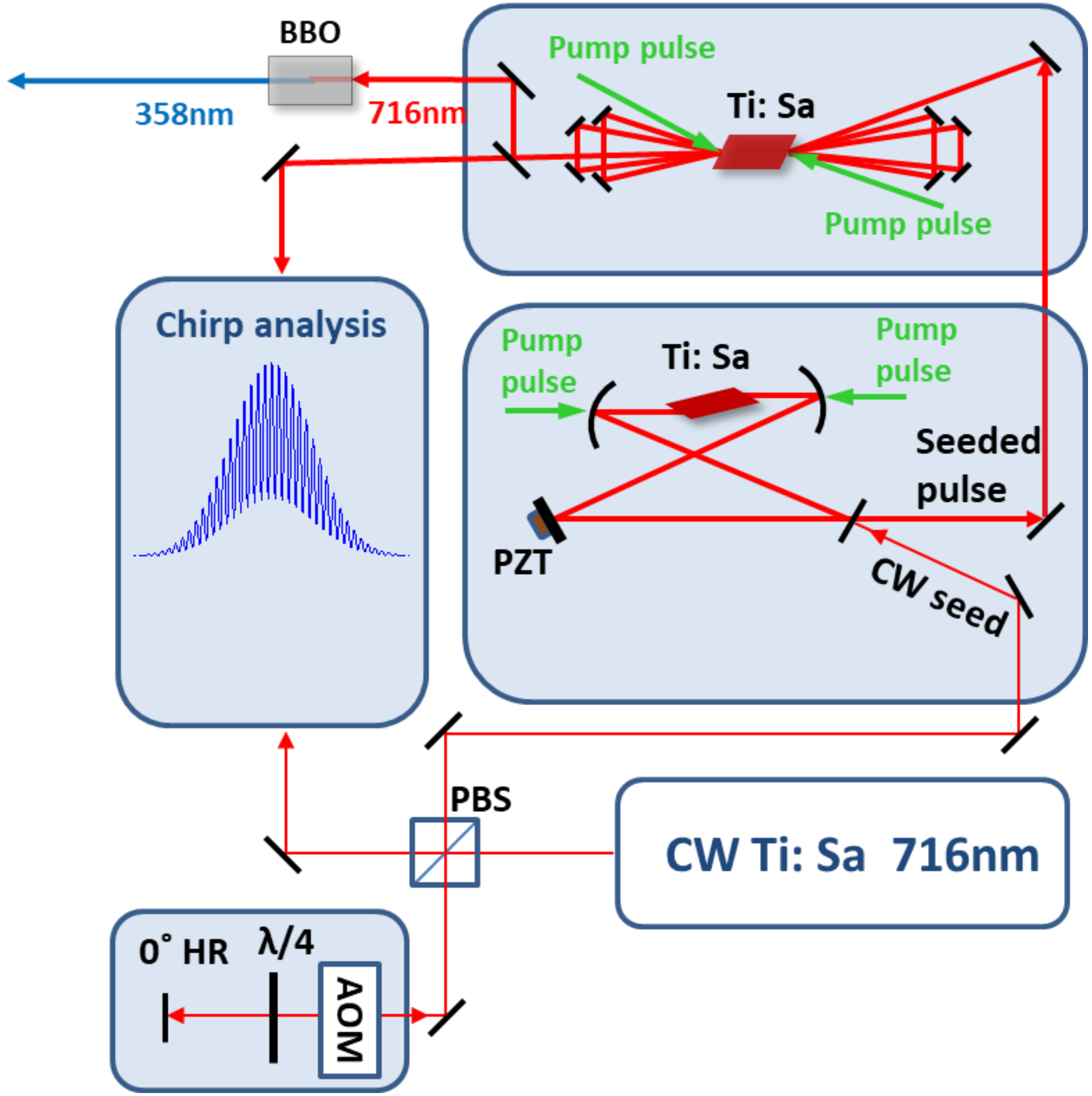
Pulse generation and metrology problem



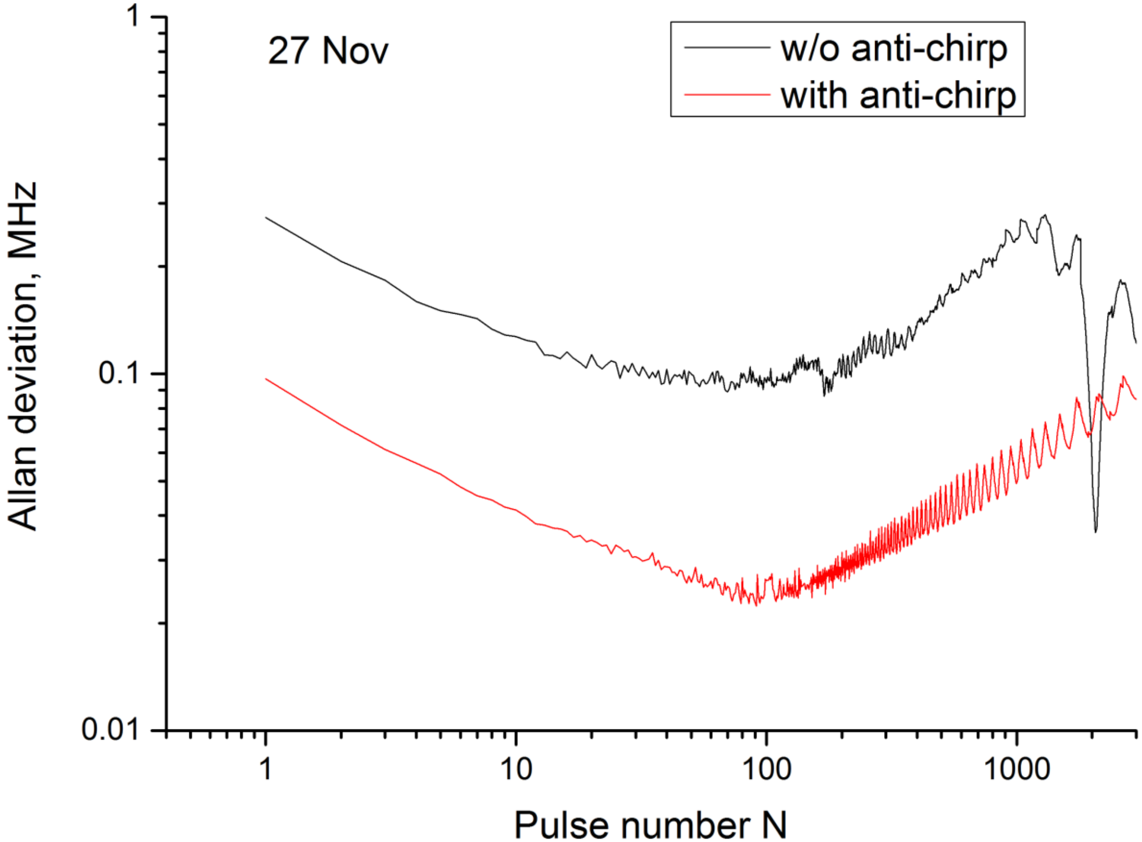


- Cavity Pulling
- Frequency chirp

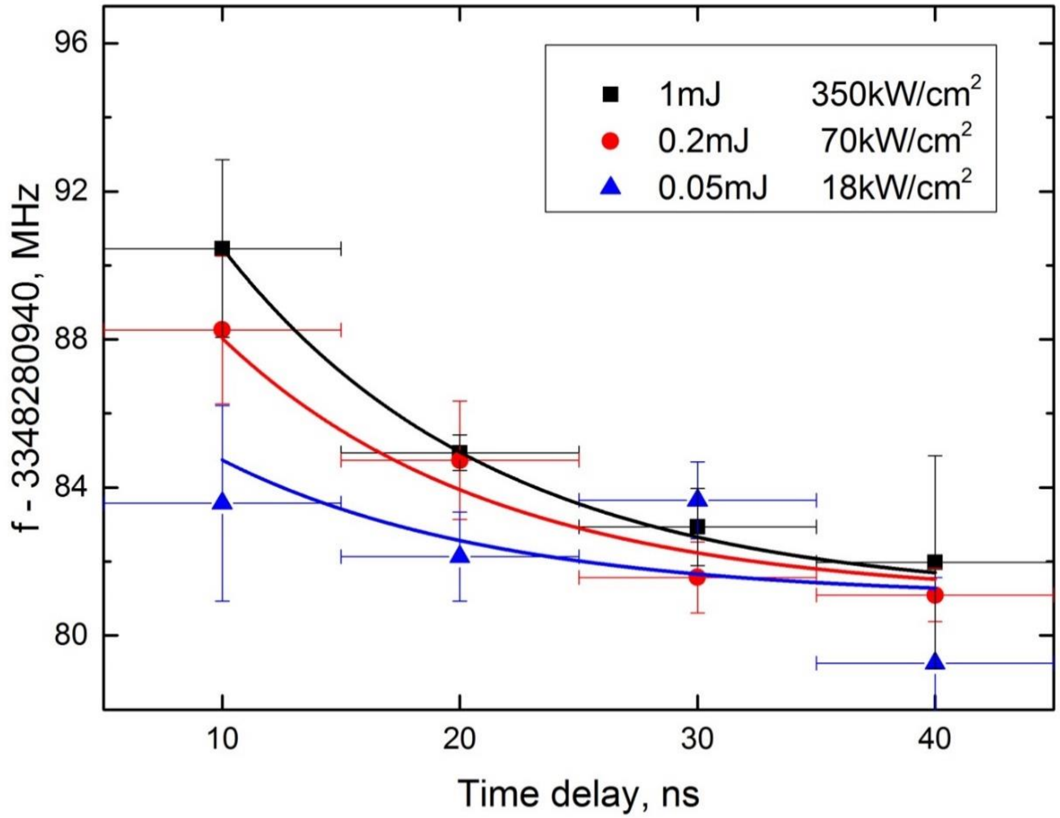
Chirp measurement

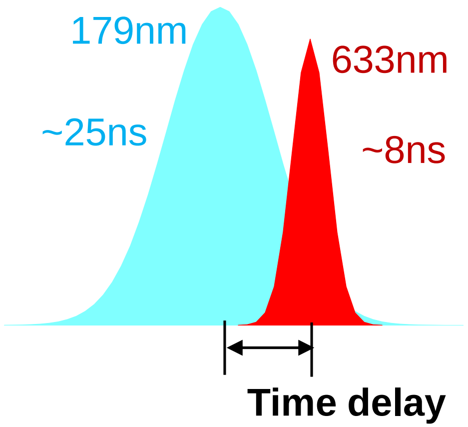


Development of anti-chirp



Systematic check: AC-stark of ionization light





Systematic check: AC Stark VUV

Power range:

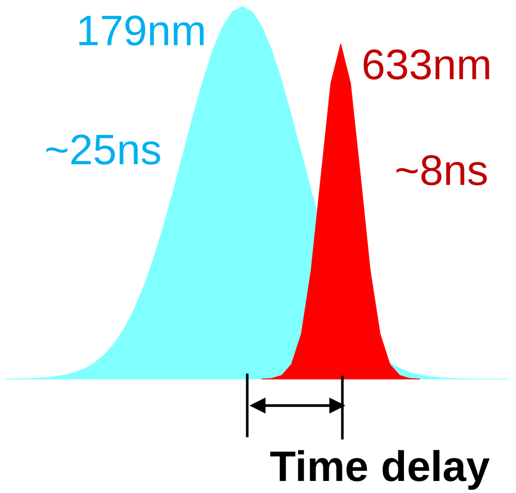
1 – 2.5mJ (358nm)

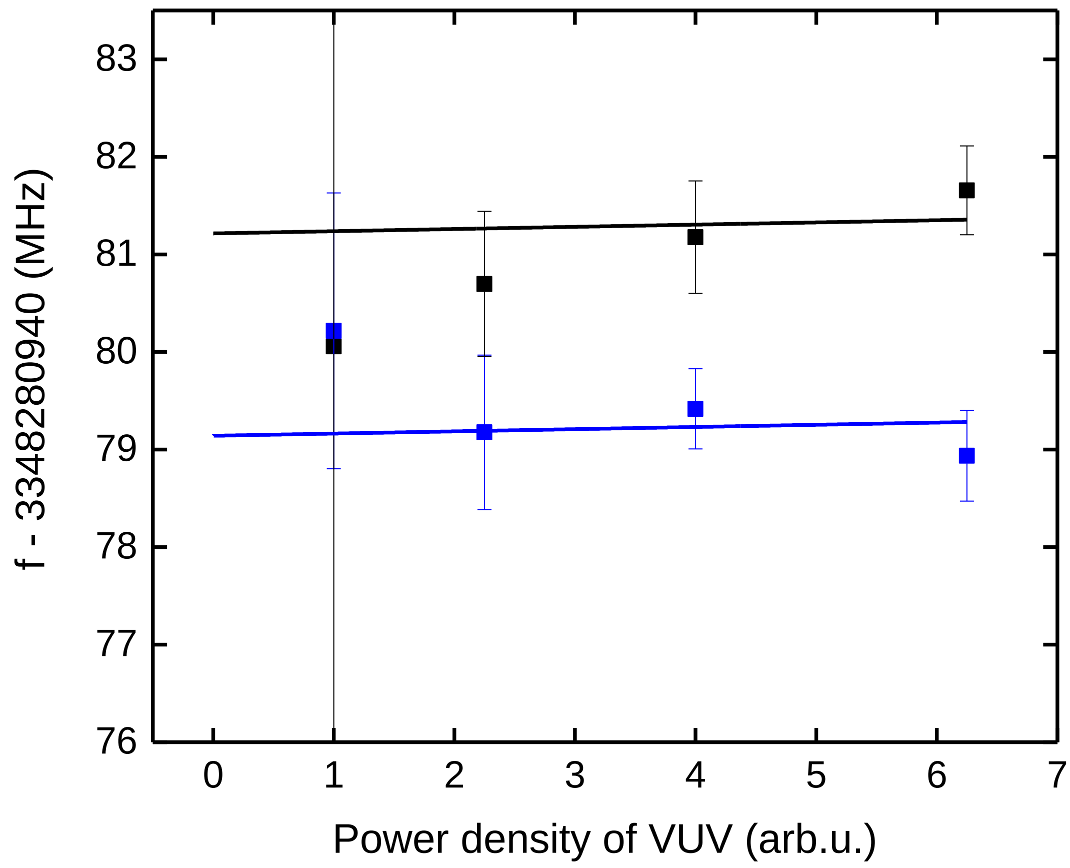
About

1 – 10uJ (179nm)

4 – 40 kW/cm²

Two different “Doppler” alignments





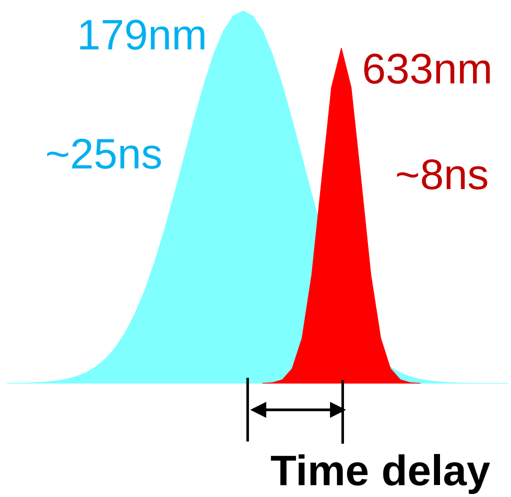
Systematic check: AC Stark ionization

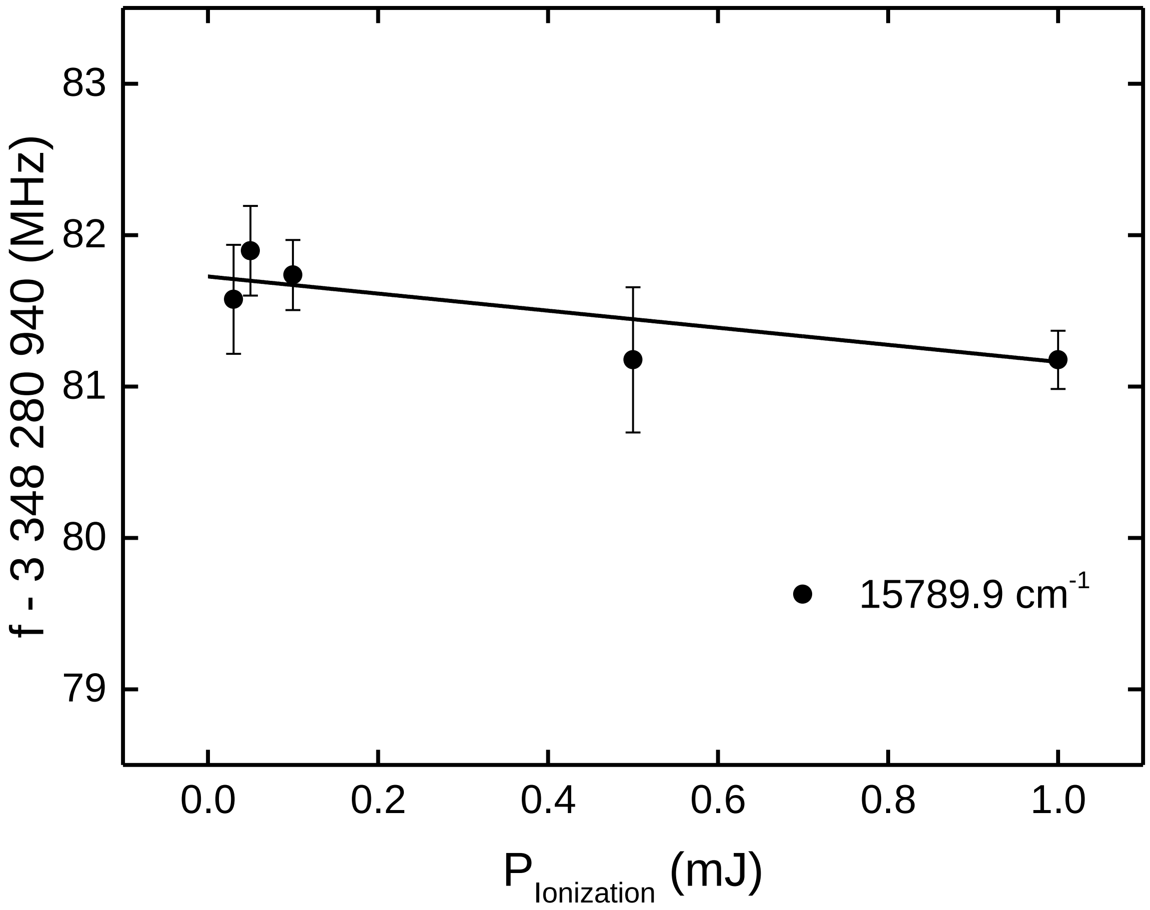
Power range:

0.05 – 1.0mJ (633nm)

About

18 – 350 kW/cm²





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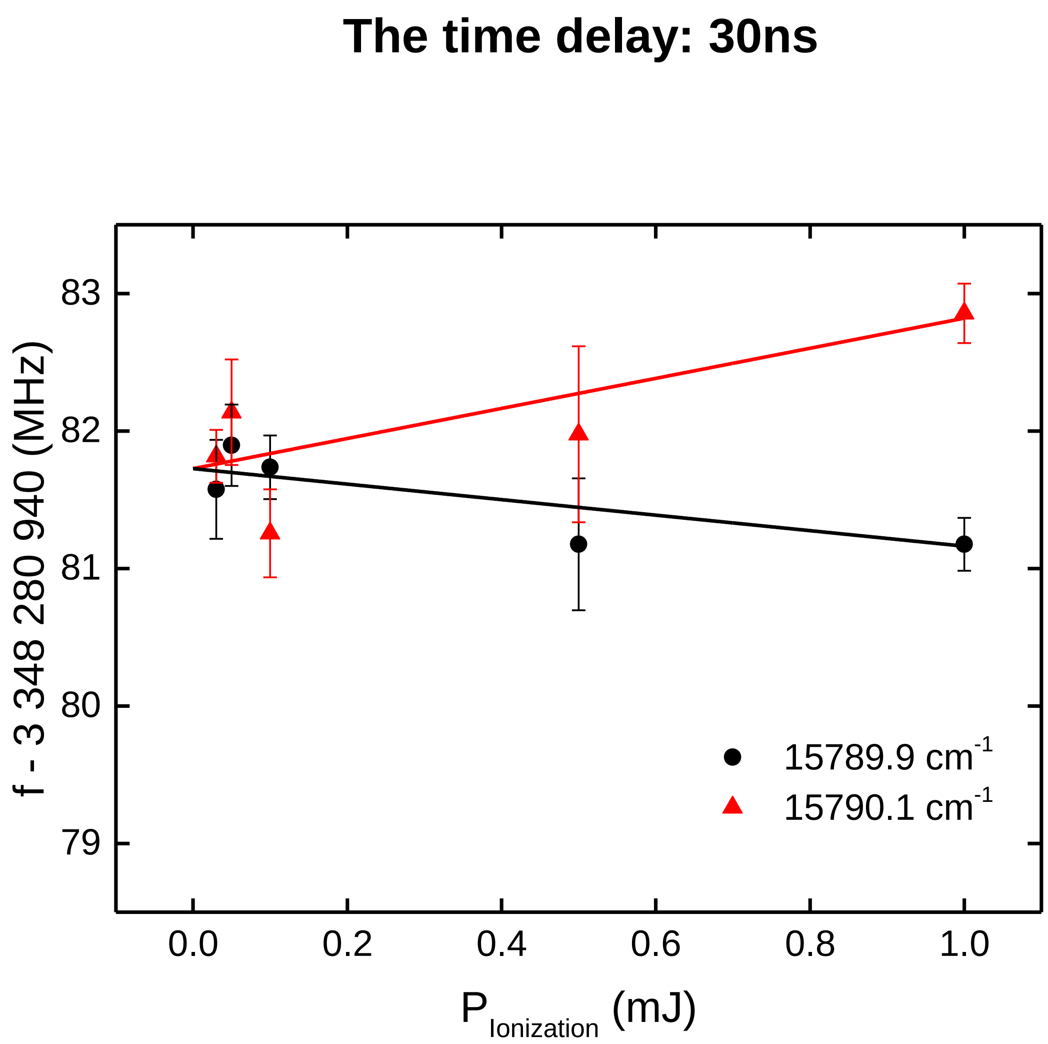

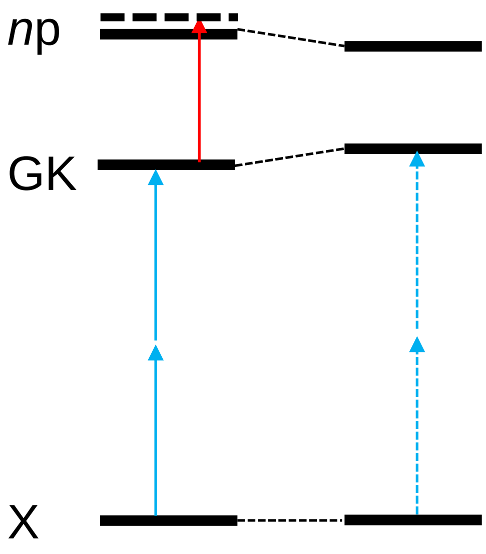

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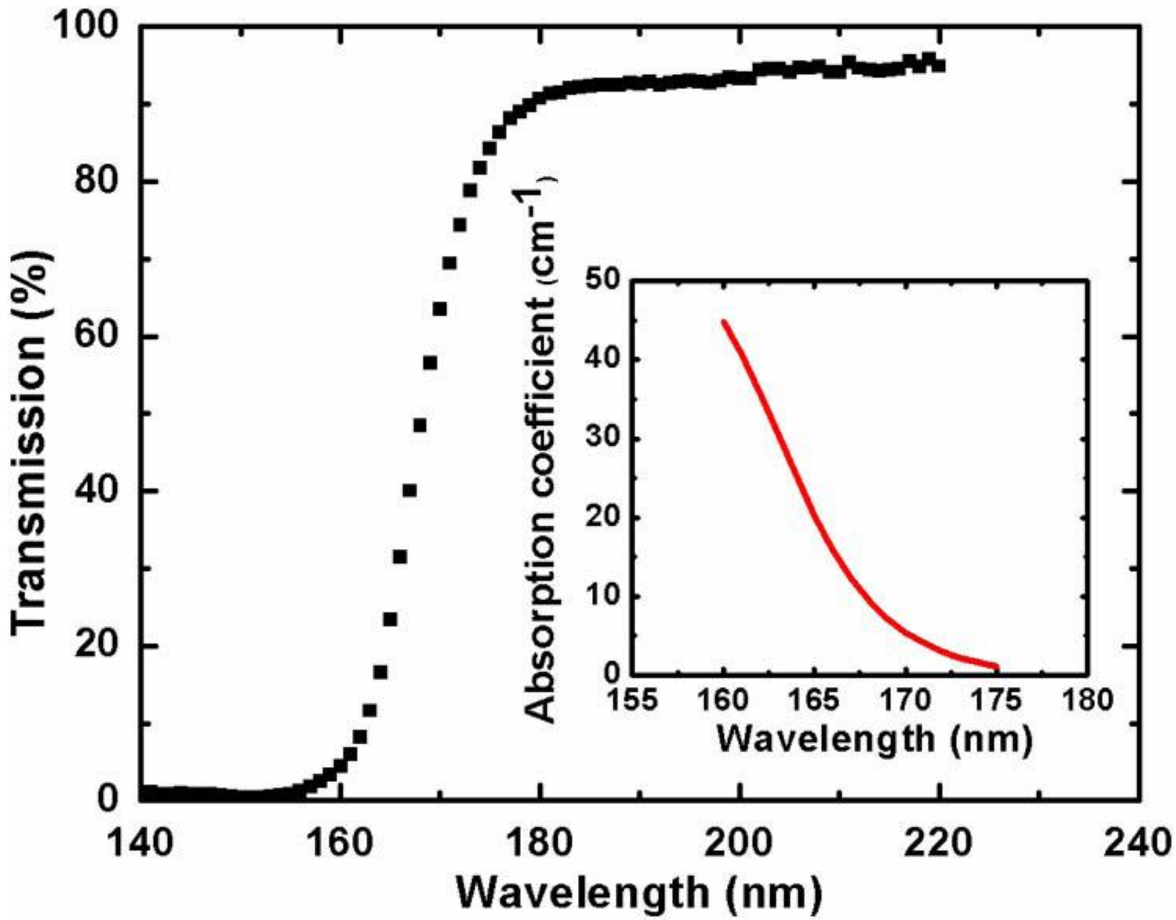
KBBF



100
80
60
40
20
0
Transmission (%)
140
160
180
200
220
240
Wavelength (nm)
Absorption coefficient (cm-1)
50
40
30
20
10
0
155
160
165
170
175
180
Wavelength (nm)

Test QED in H_2 system

Ground state $E_{level} = E_{BO} + E_{AD} + E_{NAD} + E_{REL} + E_{QED}$

Non- Relativistic

Relativistic

Born-Oppenheimer
$E_{BO} = E_{ELE} + (E_{VIB} + E_{ROT})$

Electron's self energy & vacuum field
~ 10^{-5} of BO

Adiabatic
finite nuclear mass correction
~ 10^{-4} of BO

Relativistic corrections for orbiting electron
~ 10^{-5} of BO

Non-Adiabatic
Couples electronic and nuclear motion
~10^{-4}-10^{-5} of BO

$1 cm^{-1}$~30GHz

- Current calculation accuracy 3~30MHz, including QED
- Proton size kicks in when accuracy reaches 0.01~0.1MHz

Jacek Komasa, Krzysztof Pachucki, *et.al.*

J. Chem. Theory Comput. 7, 3105–3115 (2011)