Perturbations In The Spectra of High Rydberg States: Channel Interactions, Stark and Zeeman Effects

Christa Haase, Martin Schäfer, Stephen D. Hogan and Frédéric Merkt,

Laboratorium für physikalische Chemie, ETH Zürich.

Overview

Motivation

Study the role of nuclear spin in photoionization by highly accurate (precision < 1 MHz) spectroscopy of high Rydberg states.

Experimental Considerations and Results

VUV/sub-millimeter wave double resonance experiments coupled with state-selective ionization to study the different Xe isotopes separately. Experimental results on s/d interaction in ^{129}Xe.

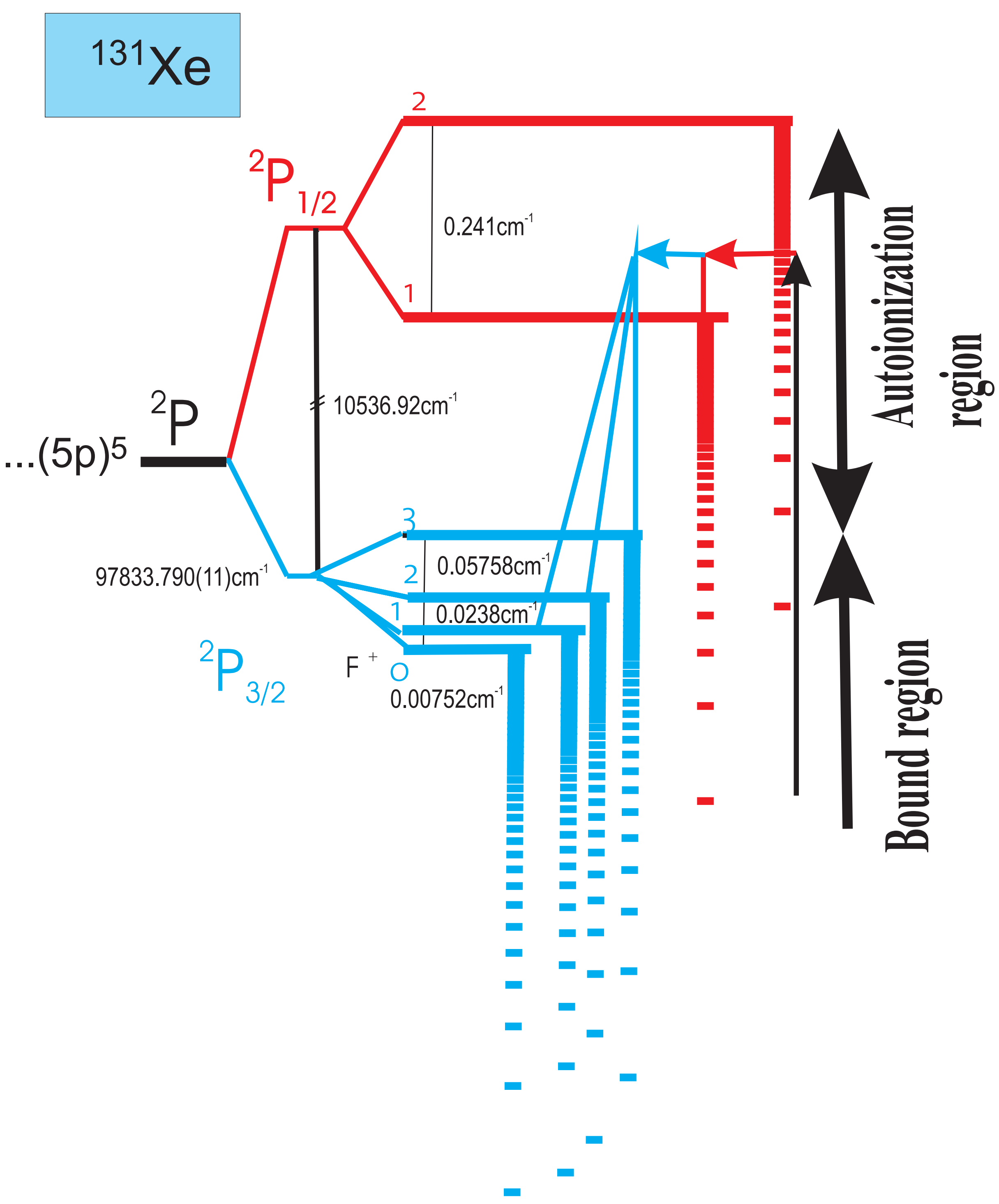
Perturbing Stray Electric and Magnetic Fields

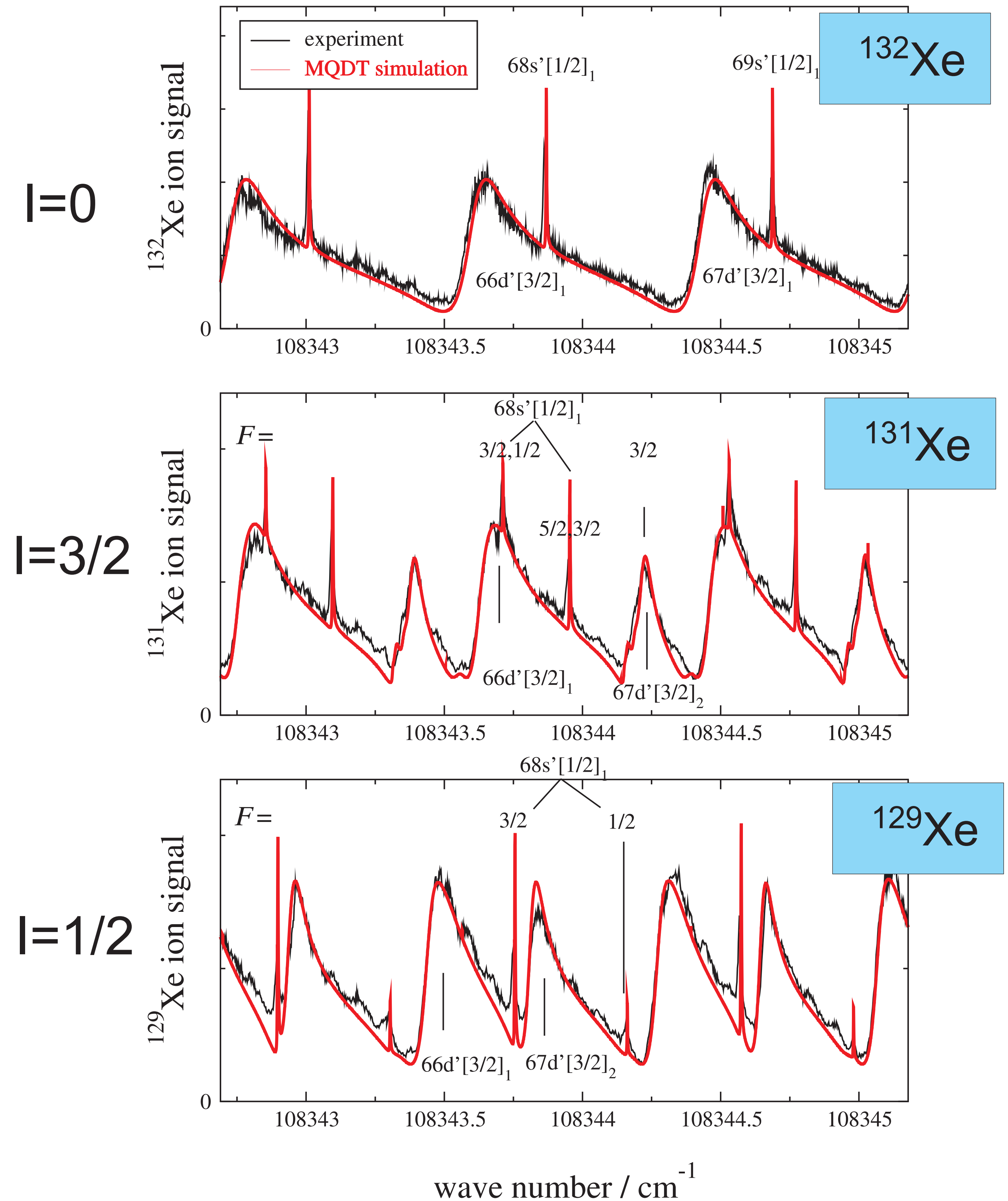
Stark shift

Zeeman splitting

Conclusions

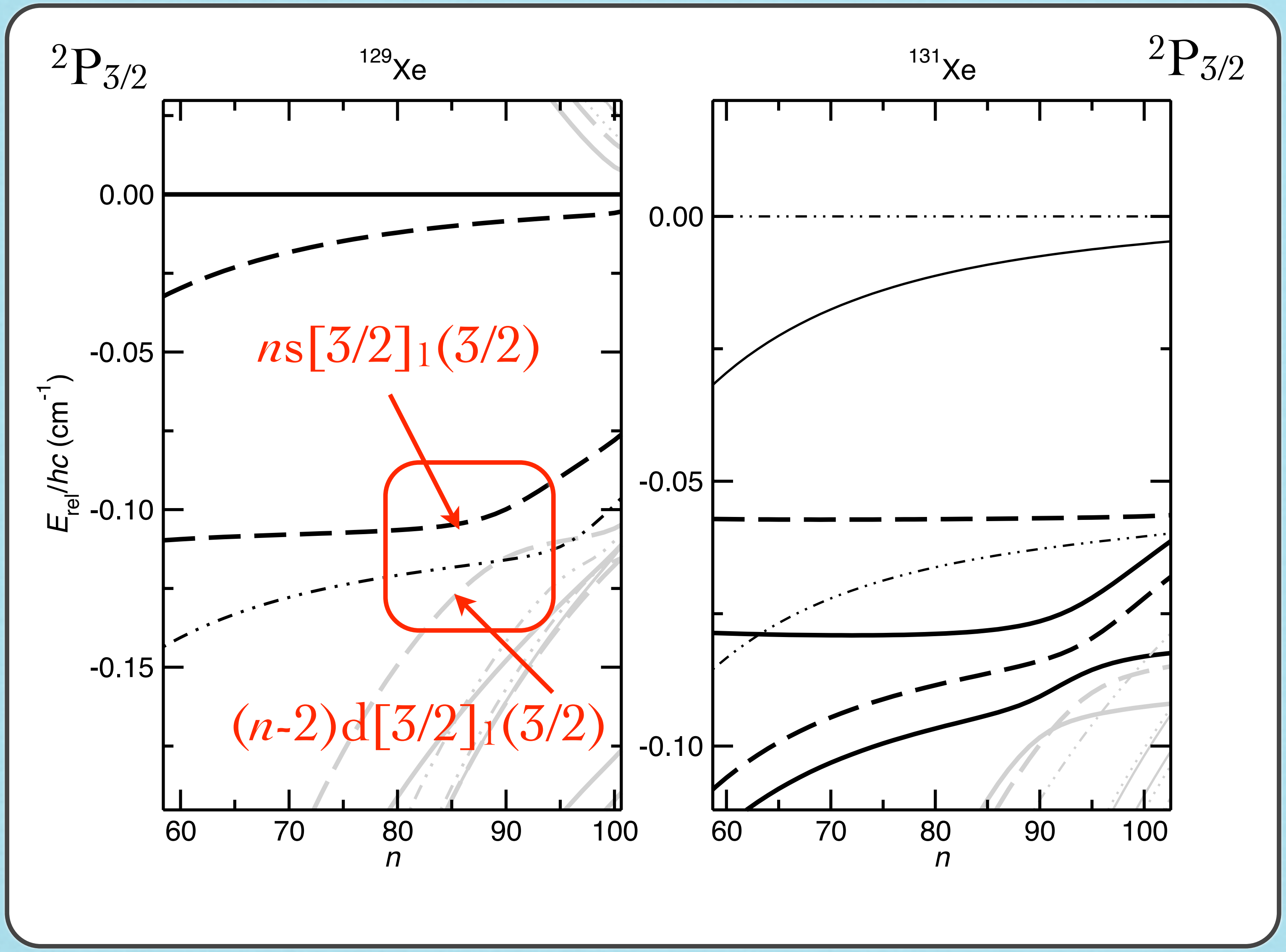
Autoionization dynamics in ^{131}Xe (I=3/2)





Wörner et al. Phys. Rev. A 71, 054501 (2005)

Previous Calculations [1]



Accurate MQDT calculations done on hyperfine structure of ^{129}Xe and ^{131}Xe up to n=100 in [1] that predict several s/d interactions around and above n=90.

No experimental studies in this region thus far.

[1] M. Schäfer et al., Phys. Rev. A. 81 (2010) 032514:1-17.

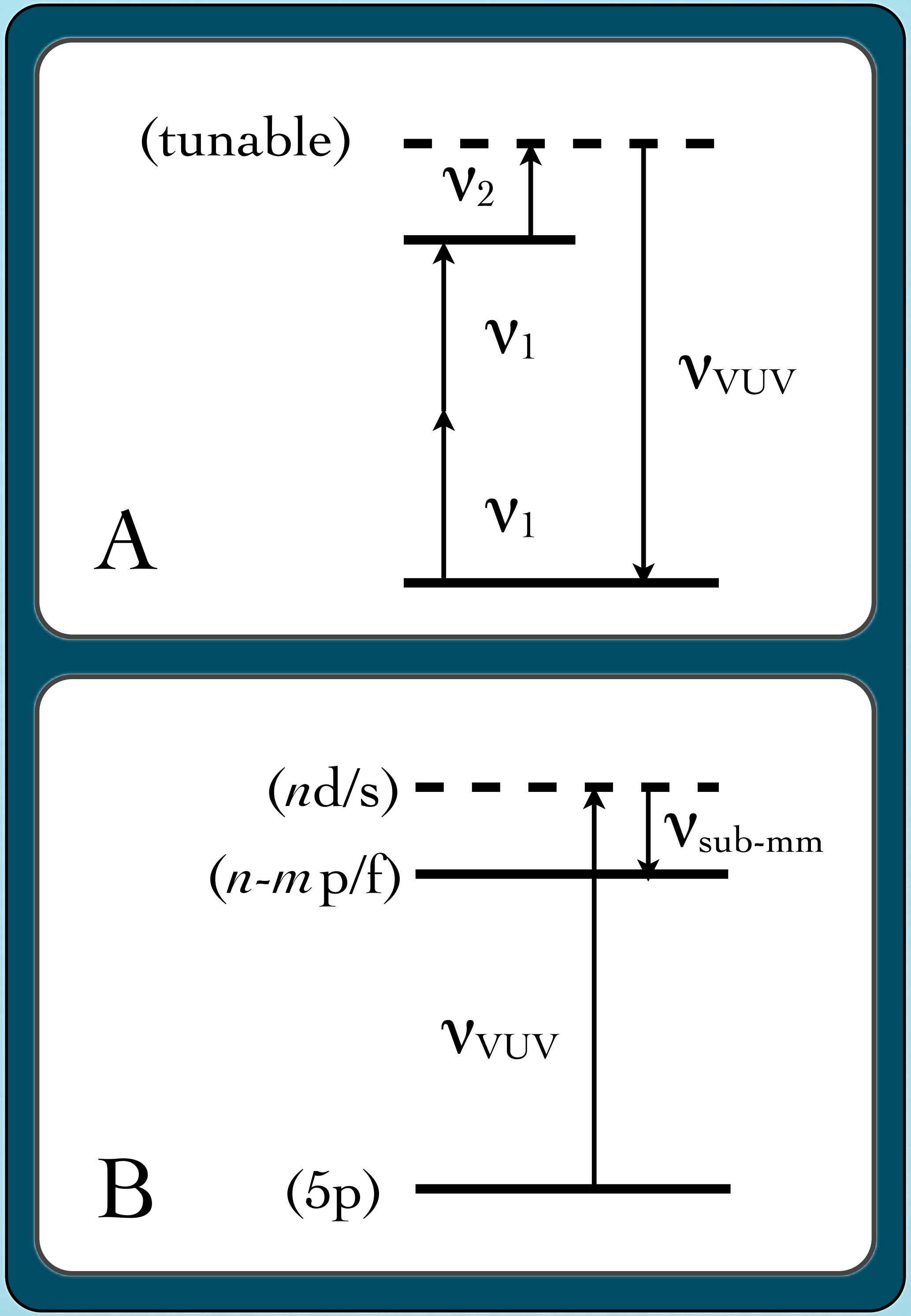
Excitation Scheme

Excitation of an atomic beam of Xe to high Rydberg states with VUV radiation, transition to lower-lying Rydberg states with sub-mm wave radiation, selective field ionisation (see next slide), ion or electron detection.

Excitation Scheme

A: VUV generation by four-wave mixing

B: Excitation of Xe

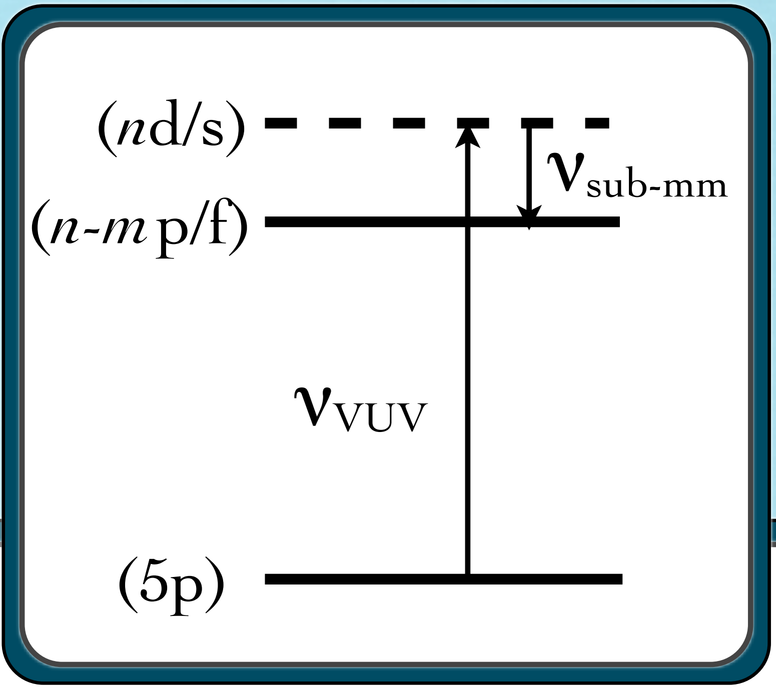


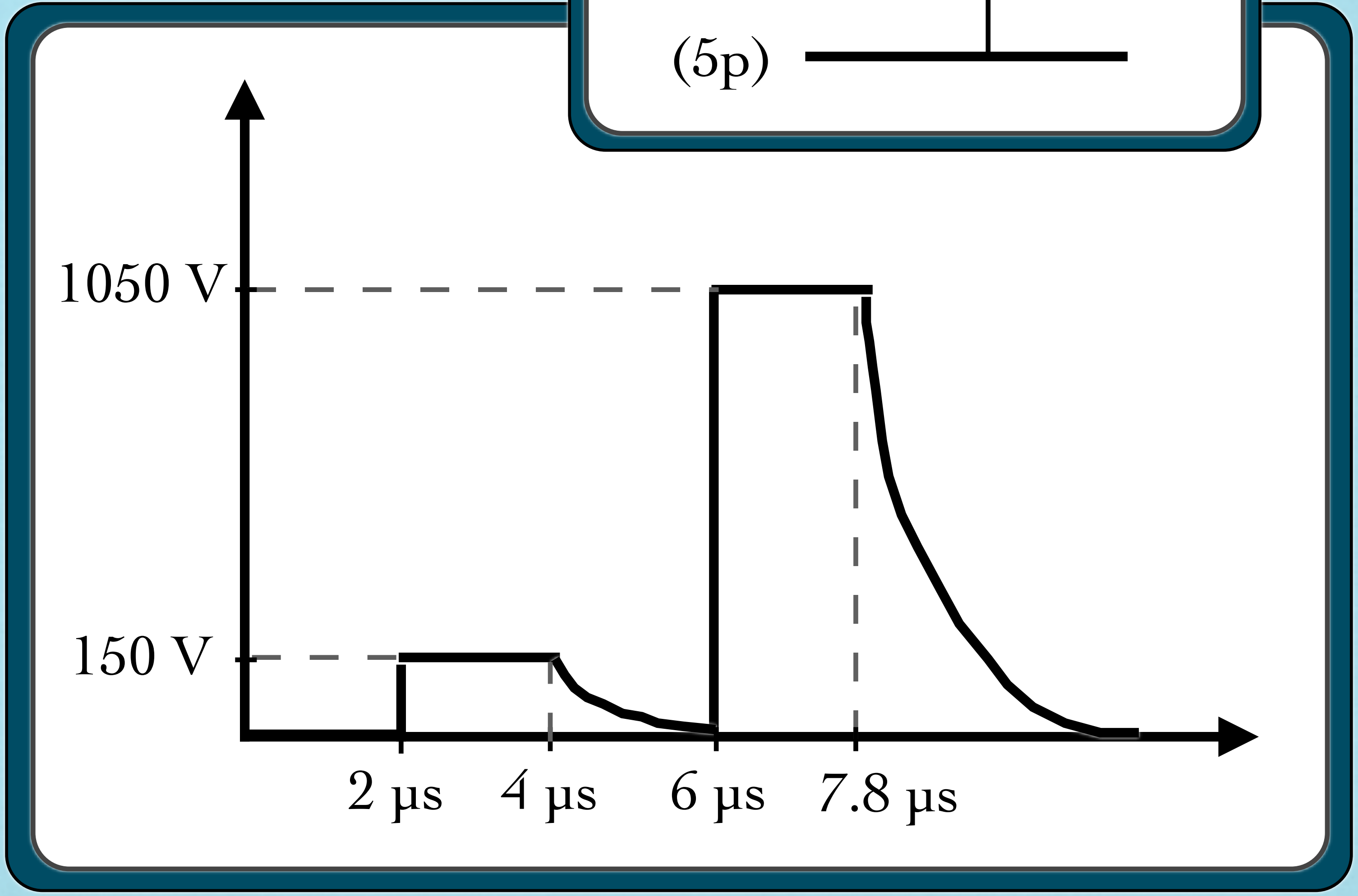
Rydberg-State Selective Ionisation

Lower voltage pulse (150V) used to ionize intermediate state (highest energy state).

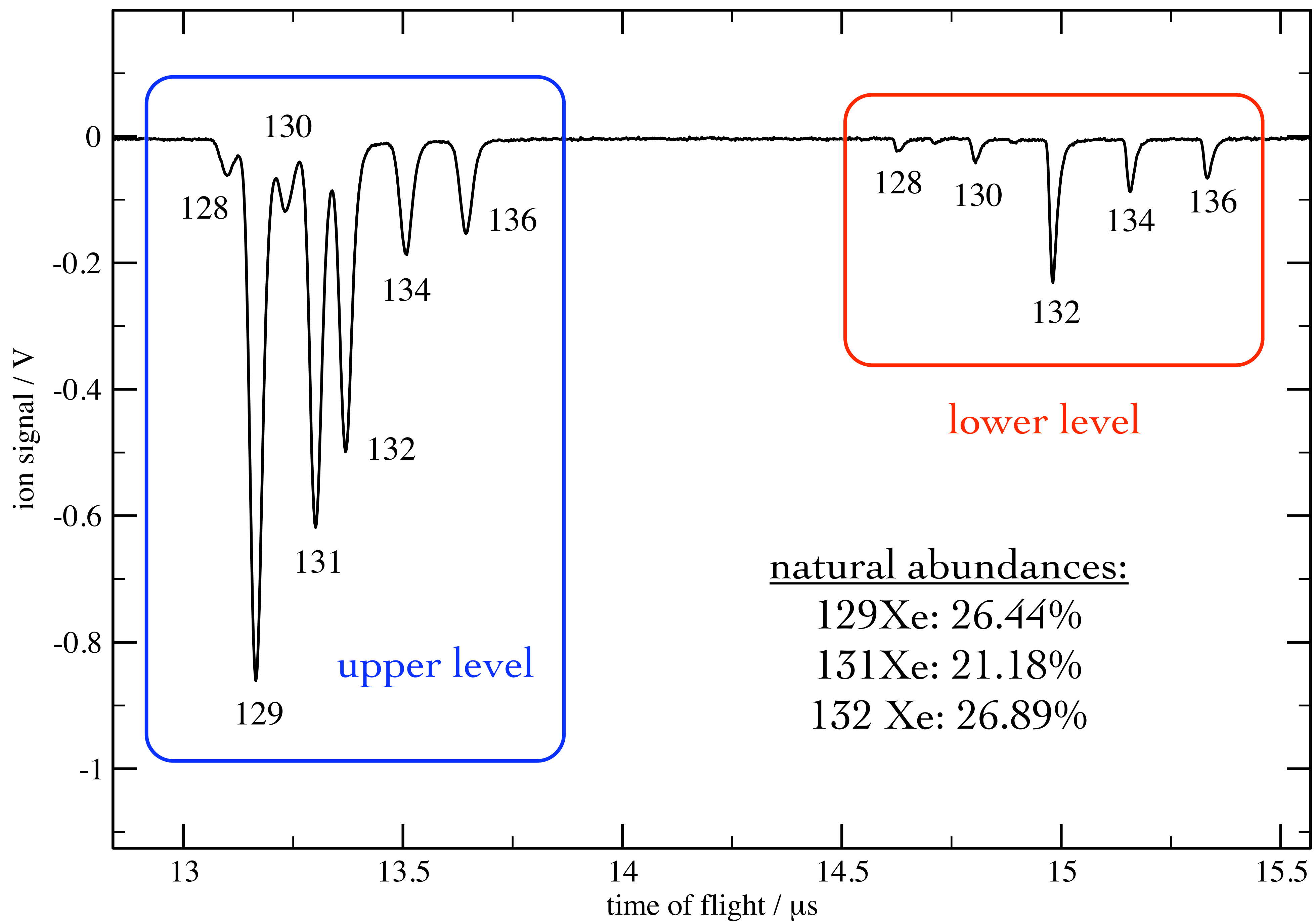
High voltage pulse used to ionize final (lower-lying) state.

Delay between pulses makes it possible to separate all of the Xe isotopes of the upper state from all of the isotopes of the lower state due to their time of flight difference and to record them simultaneously.



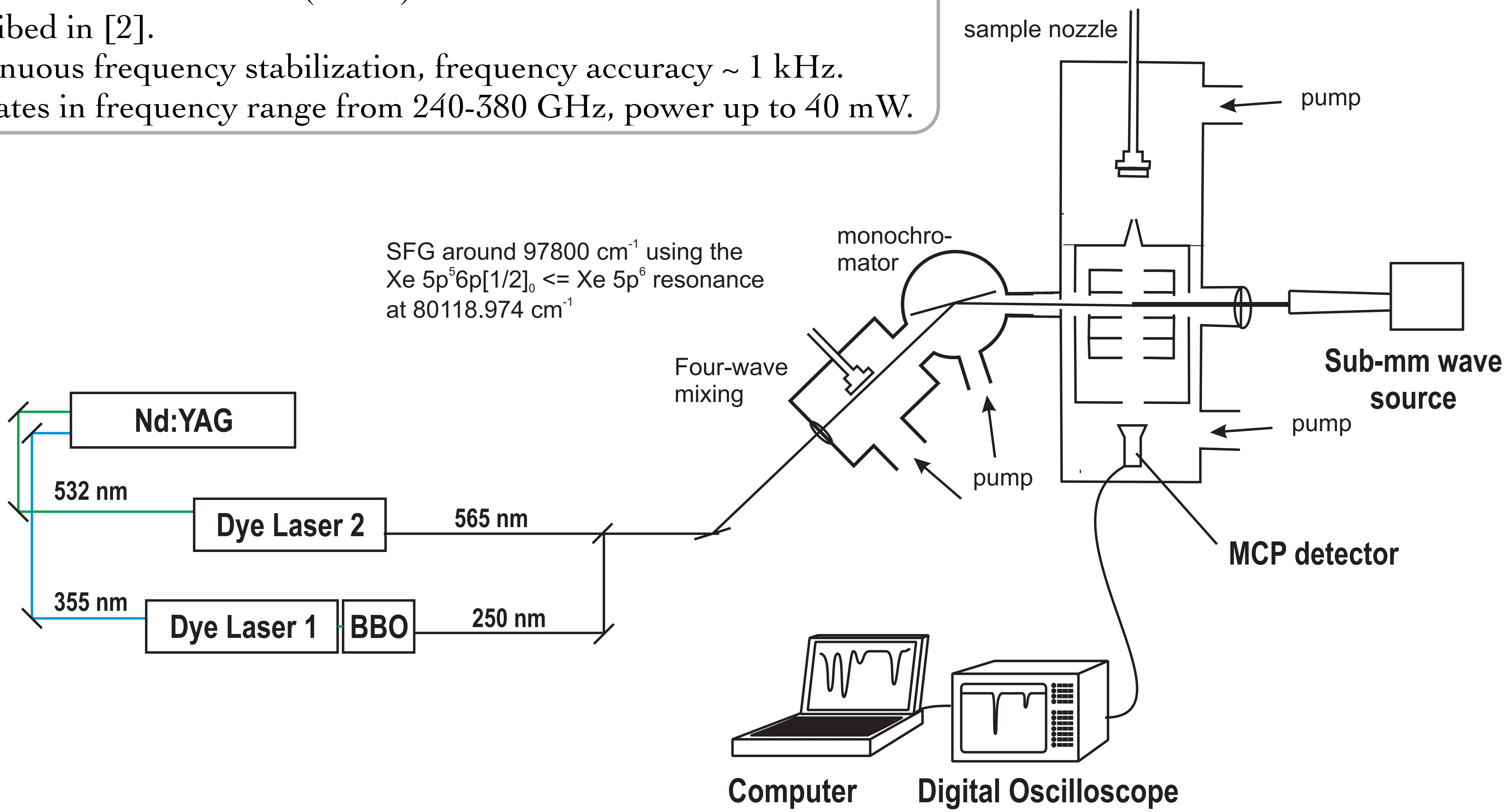


Time-of-Flight Trace: Resonant Excitation of even Xe isotopes

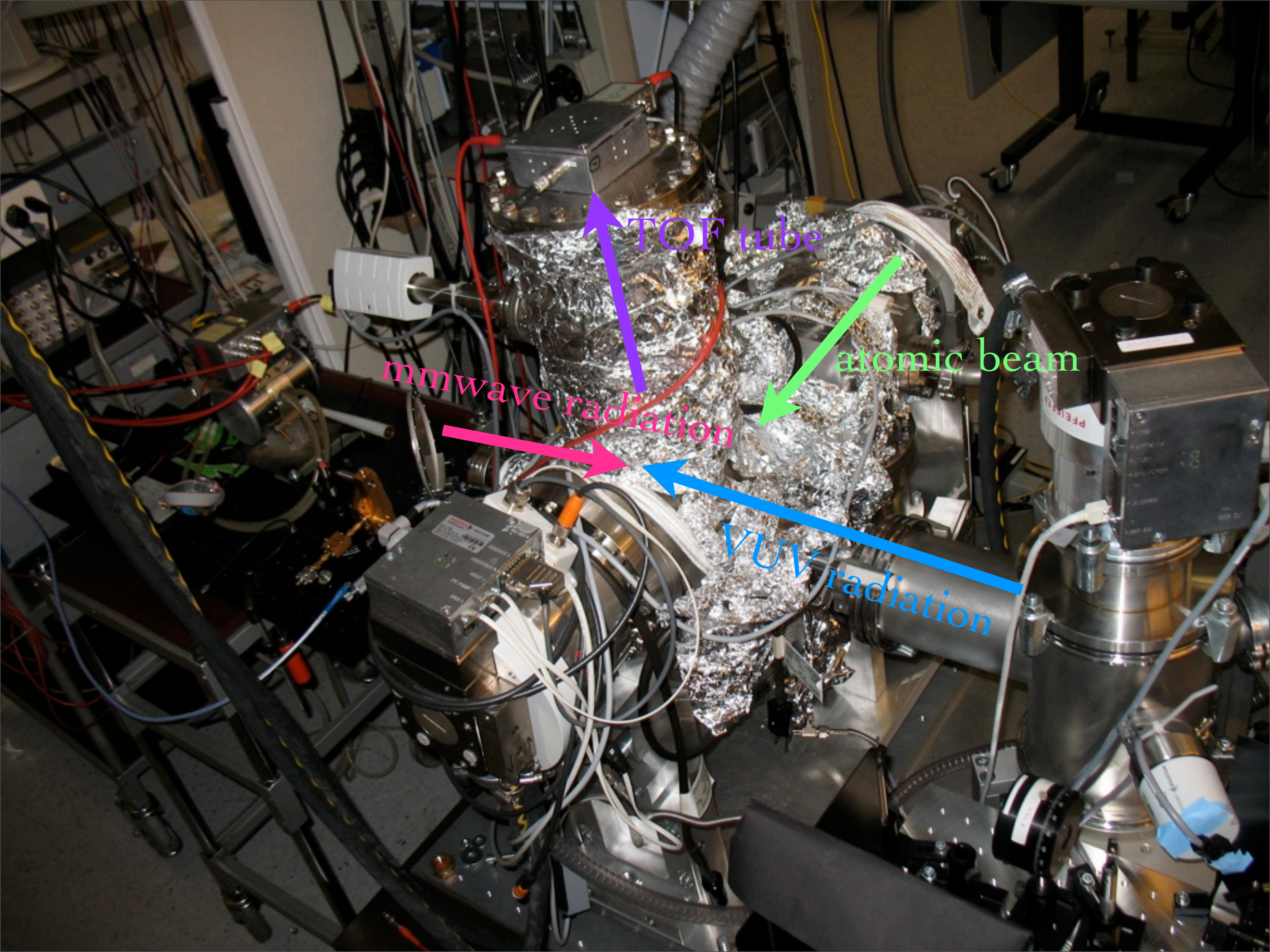


Experimental Configuration

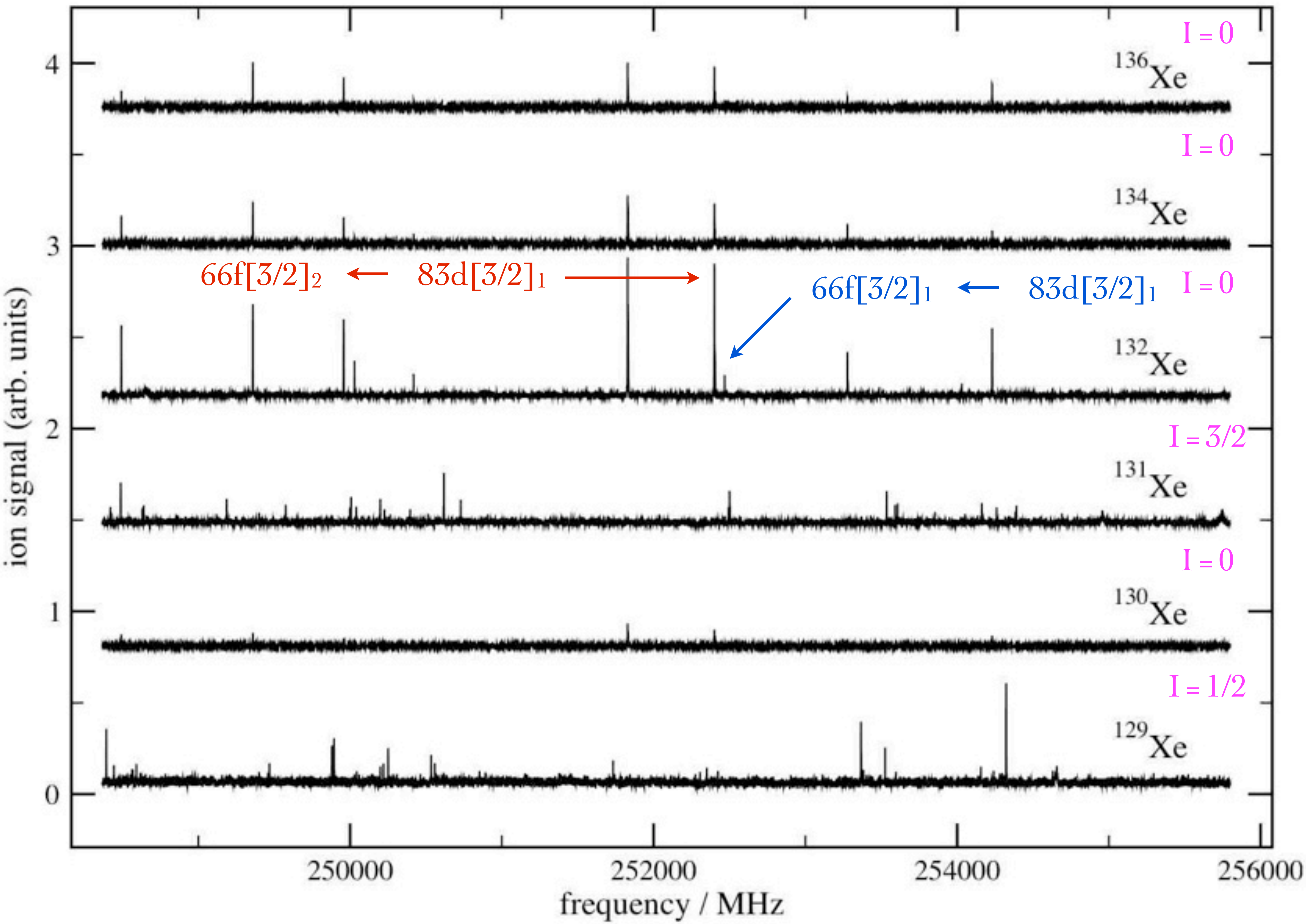
Backward-wave oscillator (BWO) based sub-millimeter wave source described in [2].
Continuous frequency stabilization, frequency accuracy ~ 1 kHz.
Operates in frequency range from 240-380 GHz, power up to 40 mW.



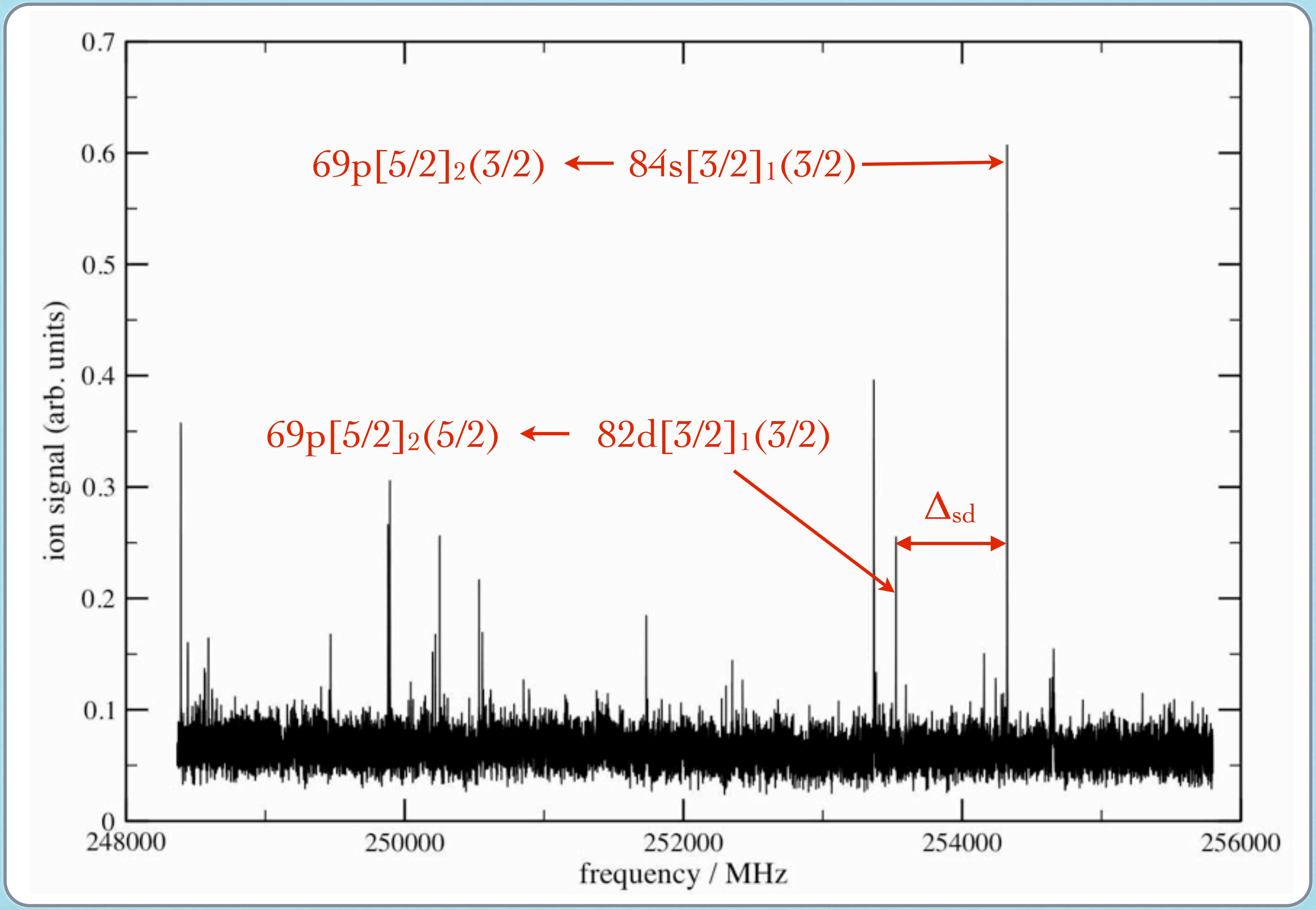
[2] M. Schäfer et al., J. Phys. B: Mol. Opt. Phys. **39** (2006) 831-845.


TOF tube
atomic beam
mmwave radiation
VUV radiation

Sub-mm Wave Spectra for Xe



Exemplary Assignments for ^{129}Xe



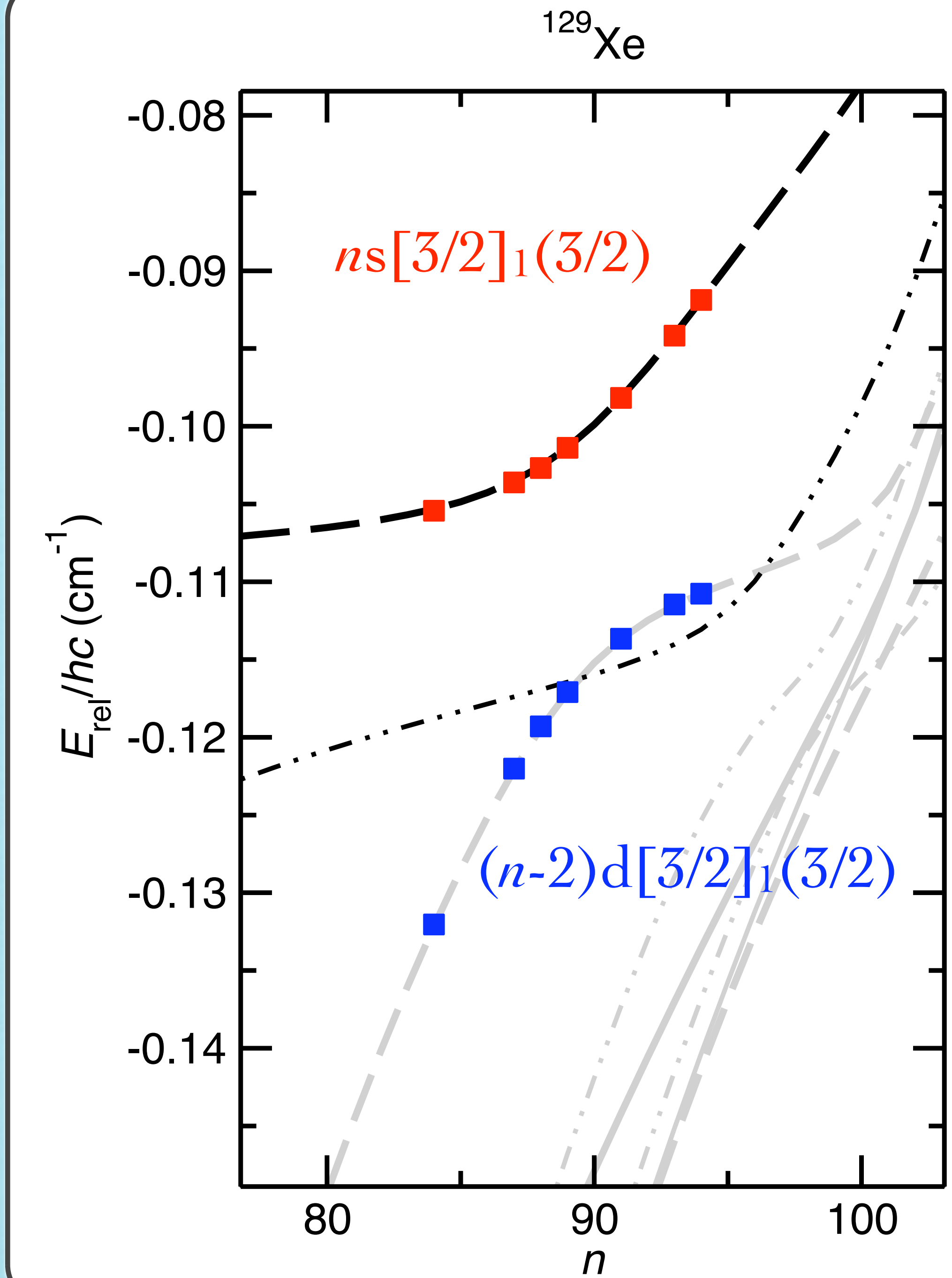
Preliminary Results for ^{129}Xe

Comparison of experimentally determined values (red and blue squares) and calculated values (adapted from [1]).

ns[3/2]$_1$(3/2) Rydberg states shown by black, dashed line, (n-2)d[3/2]$_1$(3/2) Rydberg states shown by the uppermost gray, dashed line.

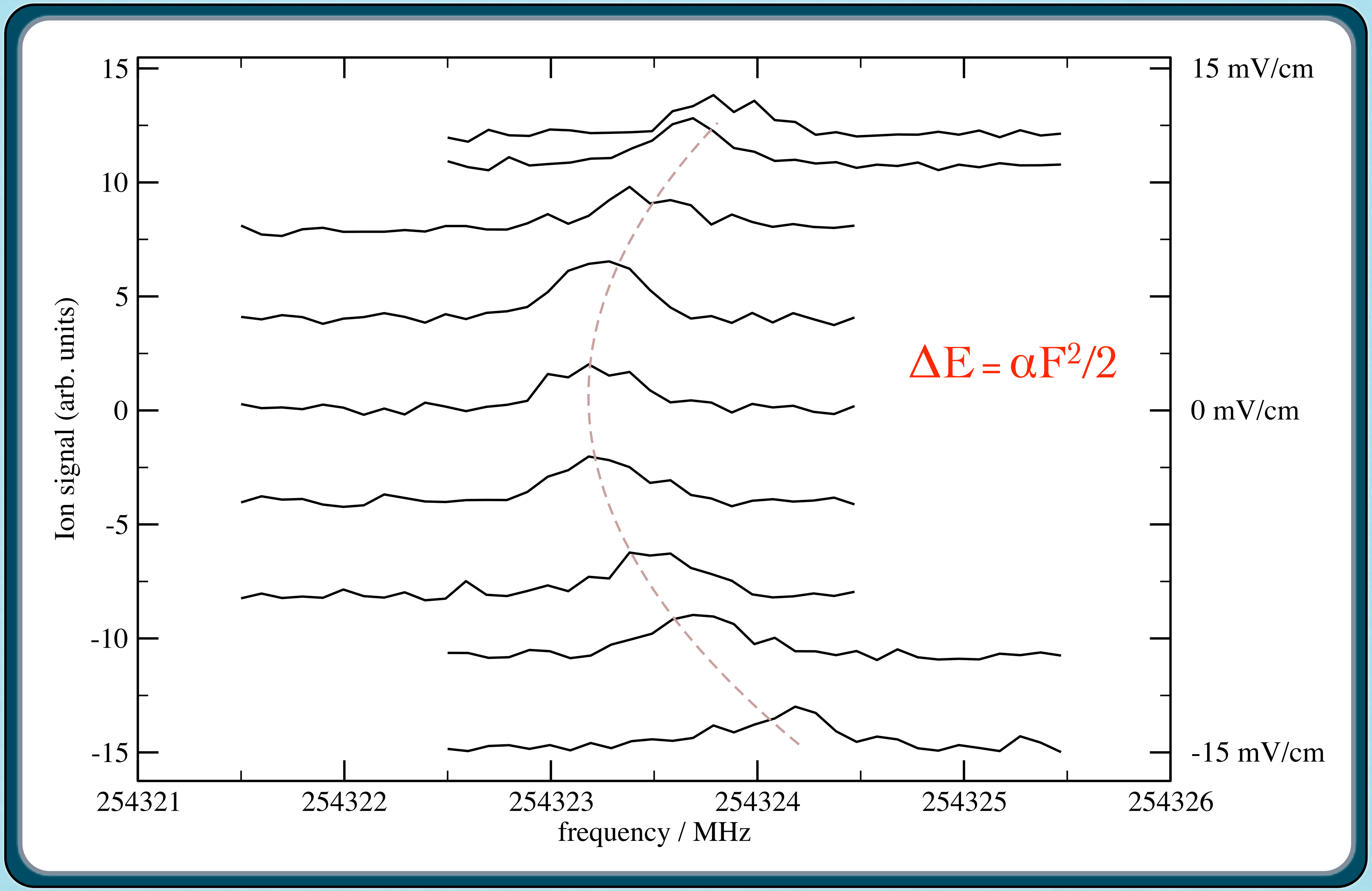
Agreement to within ~ 5 MHz.

Error on experimental results less than 1 MHz.



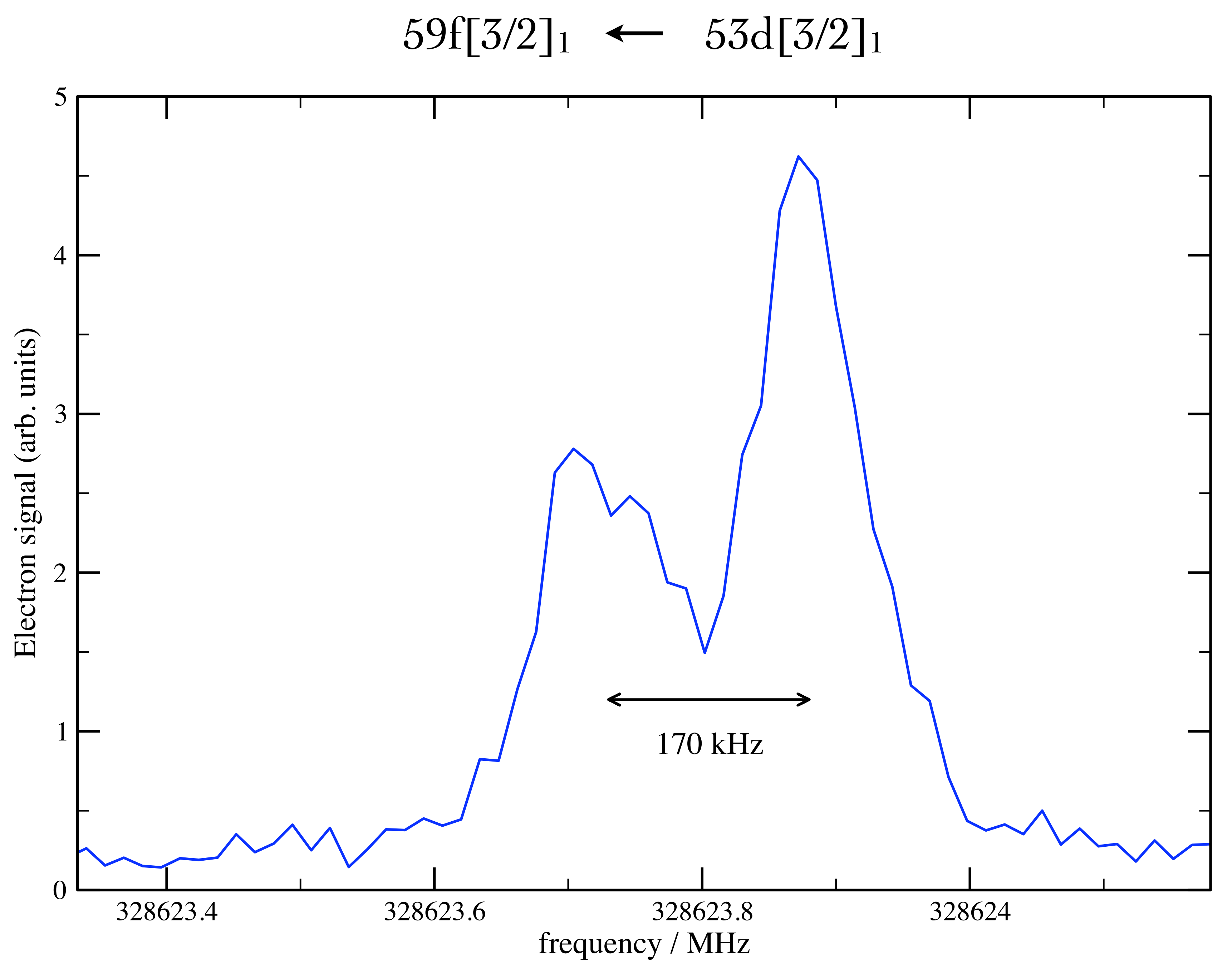
[1] M. Schäfer et al., Phys. Rev. A. 81 (2010) 032514:1-17.

Minimization of Stray Electric Fields

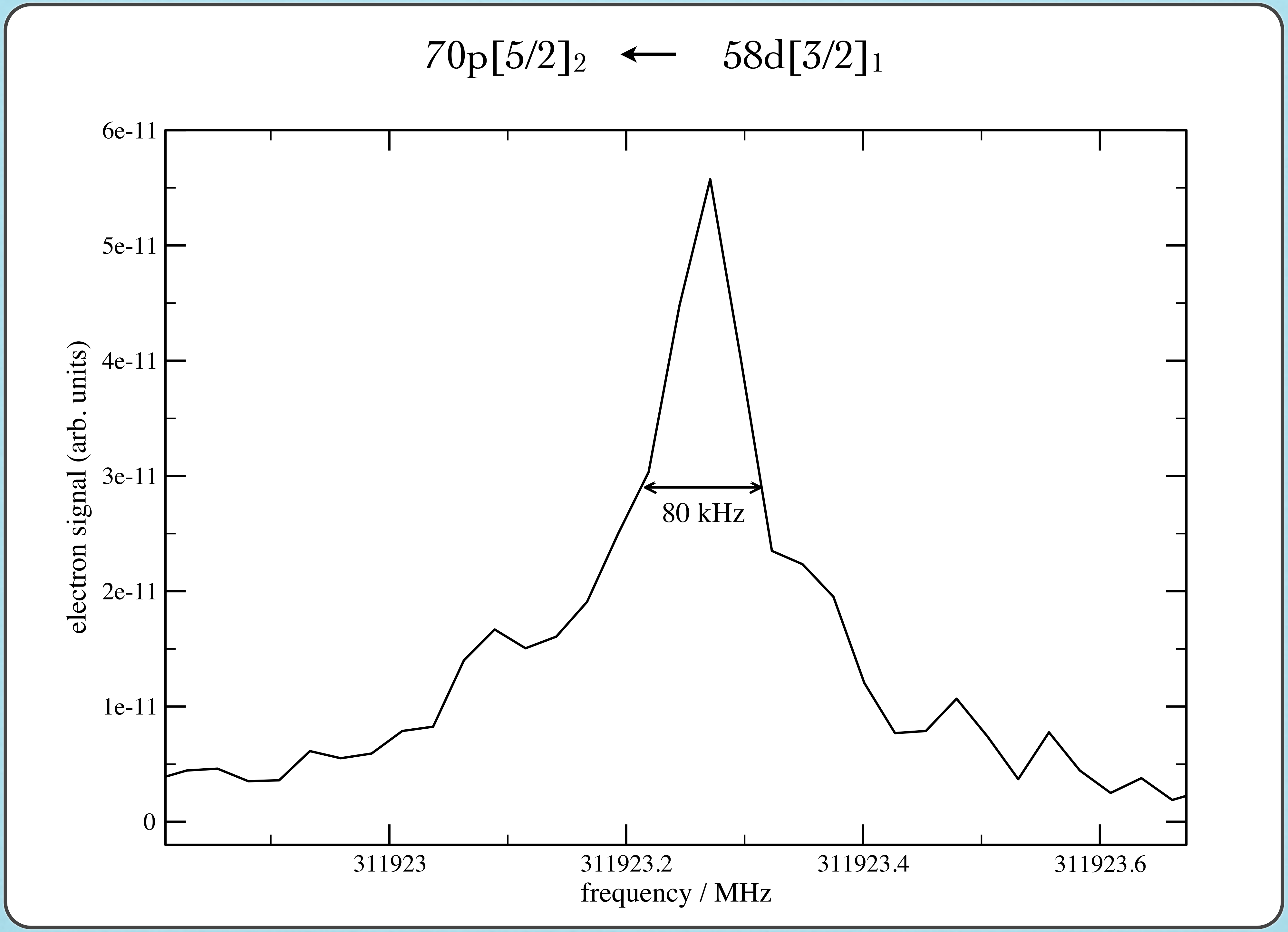


Millimeter-wave transition in the presence of different dc offset fields. The spectra are shifted vertically by an offset corresponding to the value of the total field in mV/cm (between -14.4 mV/cm and 11.8 mV/cm).

Stray Magnetic Field Effects



Transition after Stray Field Minimization



Conclusions and Outlook

Sub-millimeter wave transitions from high Rydberg states of Xe(129, 130, 131, 132, 134, 136) between n=84s/82d and n=100s/98d to various lower-lying states between n=69p and n=73p were recorded experimentally.

s/d interaction characterized experimentally for ns$[3/2]_1(3/2)$ and $(n\text{-}2)$d$[3/2]_1(3/2)$ states in ^{129}Xe in the region between 84s/82d and 94s/92d.

As preparation for these measurements, effects of perturbing stray electric and magnetic fields were studied and minimized.

Outlook: Characterize n-dependent s/d interaction for ^{131}Xe with experimental data and adjust MQDT parameters to incorporate experimental results for both ^{129}Xe and ^{131}Xe.







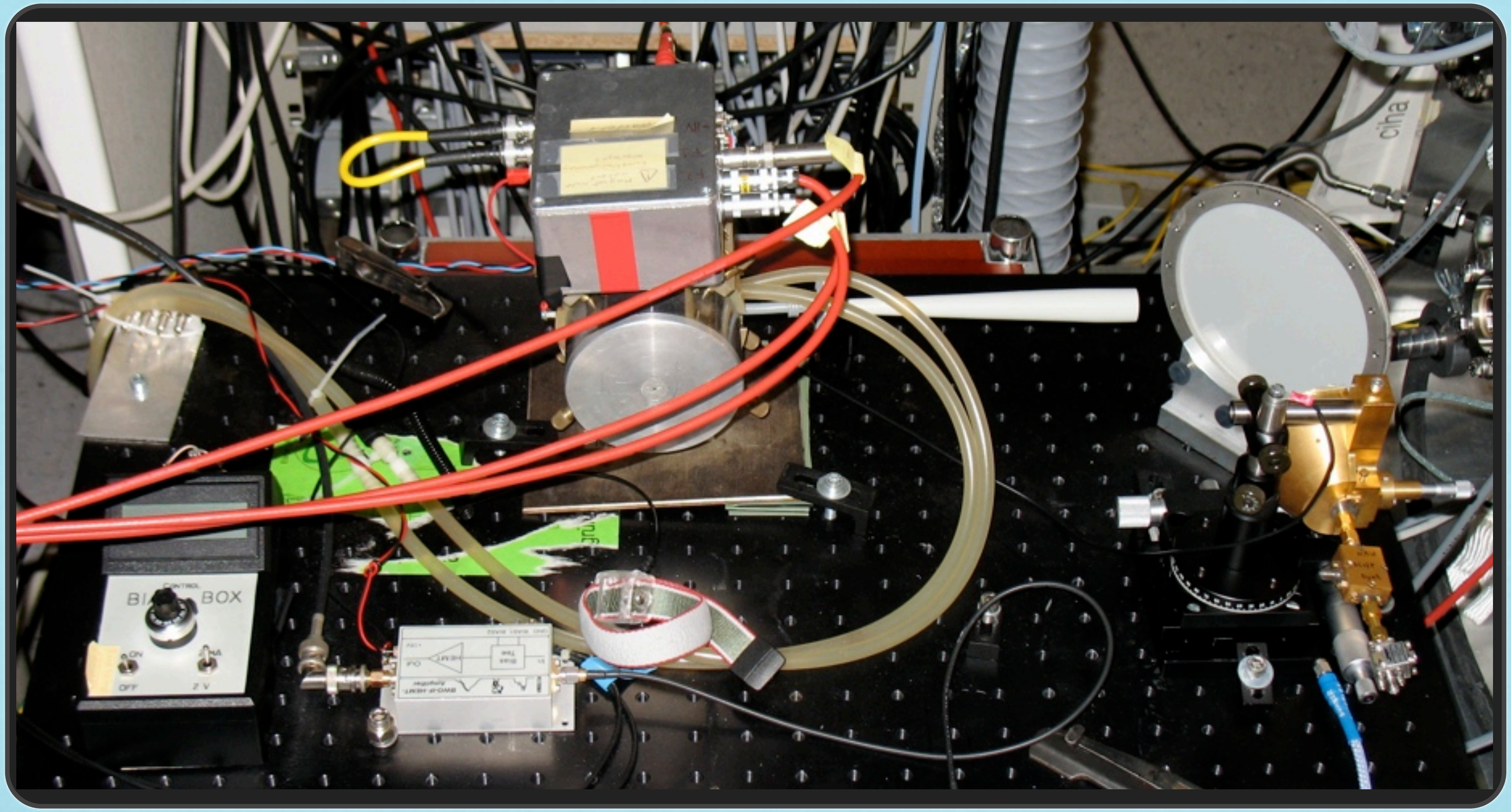



Sub-millimeter-Wave Source

Backward-wave oscillator (BWO) based source reported in [2].

Continuous frequency stabilization, frequency accuracy ~1kHz.

Operates in frequency range from 240-380GHz, power up to 40mW.



[2] M. Schäfer et al., J. Phys. B: Mol. Opt. Phys. **39** (2006) 831-845.

Backward-Wave Oscillator

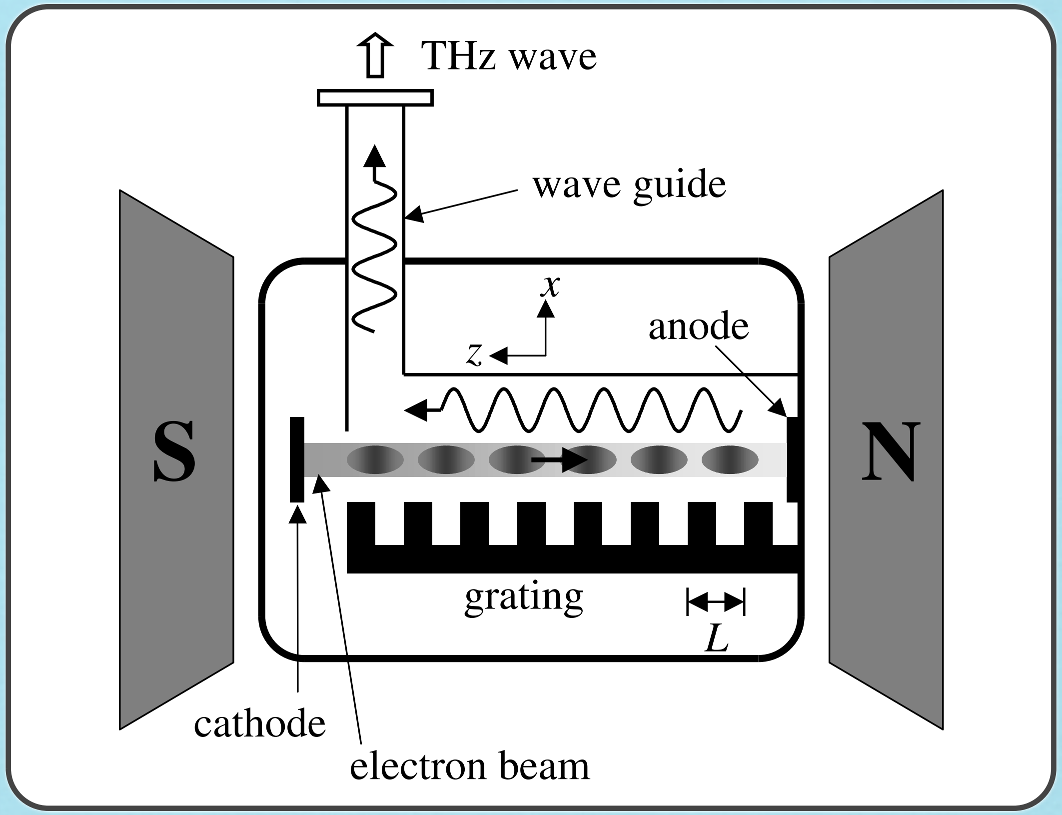
Vacuum tube in which electron beam propagates

Electron beam slowed by "comb slowwave structure", kinetic energy transferred to EM wave

Bias voltage determines the velocity of the electrons

Frequency of the backward wave dependent on velocity of the electrons

Magnetic field collimates electron beam



[3] Y.-S. Lee, *Principles of Terahertz Science and Technology* (Springer, New York, 2009).

Conclusions and Outlook

Measure all necessary transitions to quantify s/d splitting in the F=3/2 hyperfine states of ^{131}Xe and ^{129}Xe

Characterize/prepare system so that only a minimum of parameters need to be varied to do double resonance experiment in Xe with difference-frequency THz source

Study high-resolution vibrational spectra of ν_6 in $CHCl_3$/$CFCl_3$, although some difficulty with VUV generation for $CHCl_3$

Measure vibrational spectra of rare-gas clusters (although further experimental developments may be necessary for this)

Acknowledgements

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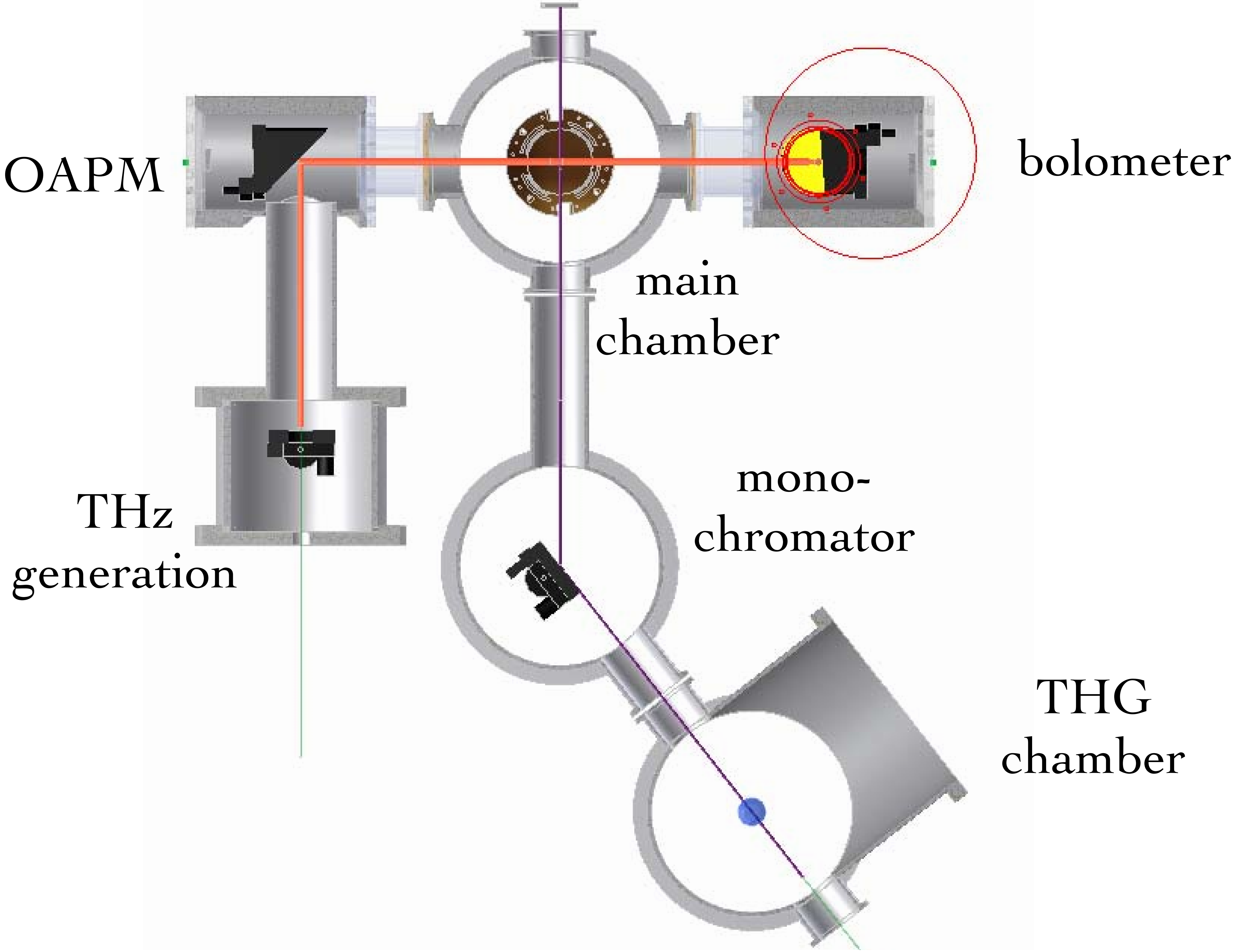
Irène, Urs (administrative, organizational)

Martin, Steven, Hansjürg, Seppi and all the other members of the XUV group for their help in various ways...!


OAPM
bolometer
main chamber
THz generation
mono-chromator
THG chamber

Advantages of the Planned Experimental Setup

Advantages of working in a molecular beam:

i) Low temperature (less spectral congestion)
ii) No pressure broadening
iii) Easier to measure rare-gas clusters later on

Advantages of detecting ions:

i) background-free
ii) detection of absorption (not transmission)
iii) isotopomer-selective (no need for isotopically pure sample)

Avoid problem of interfering water lines