High-Resolution Spectroscopy Using A New All-Solid-State VUV Laser System

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Introduction

+ High-resolution electronic spectroscopy demands narrow-band VUV radiation (< 200 MHz, ns pulses).

+ Resolution of VUV laser sources is limited by

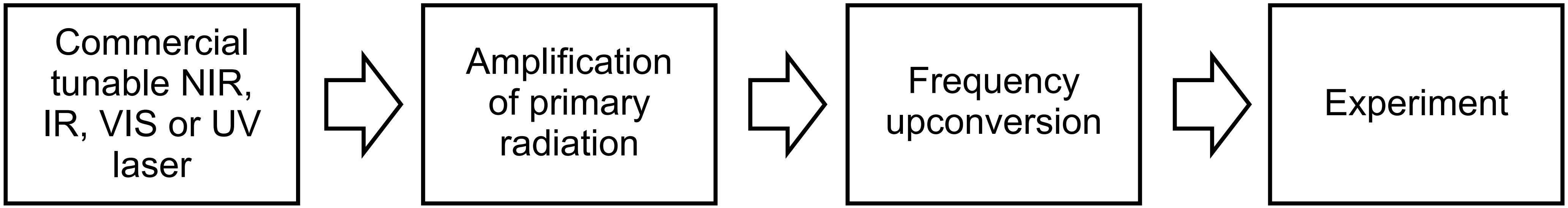
  a) bandwidth of primary sources in the NIR, IR, VIS or UV.

  b) FT-limit of VUV pulses, $_t$ = 4ln(2)/ (10 ns 88 MHz).

+ An all-solid-state VUV laser system with unprecedented resolution:

  - broad and easy tunability.

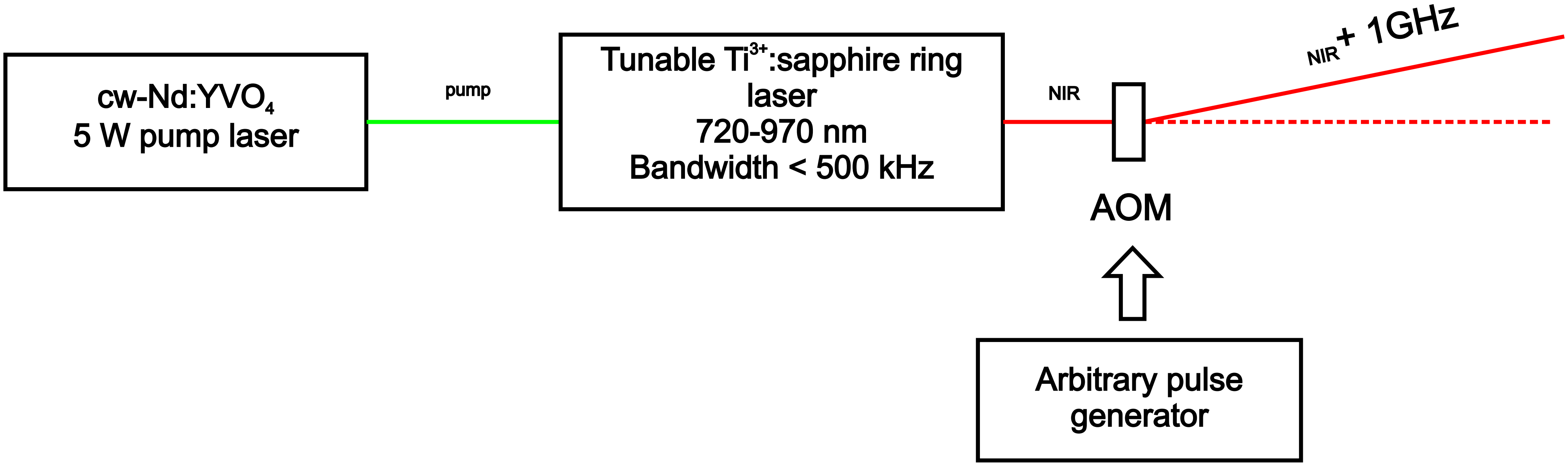
The route to VUV



Bandwidth - is intrinsically restricted by the FT-limit

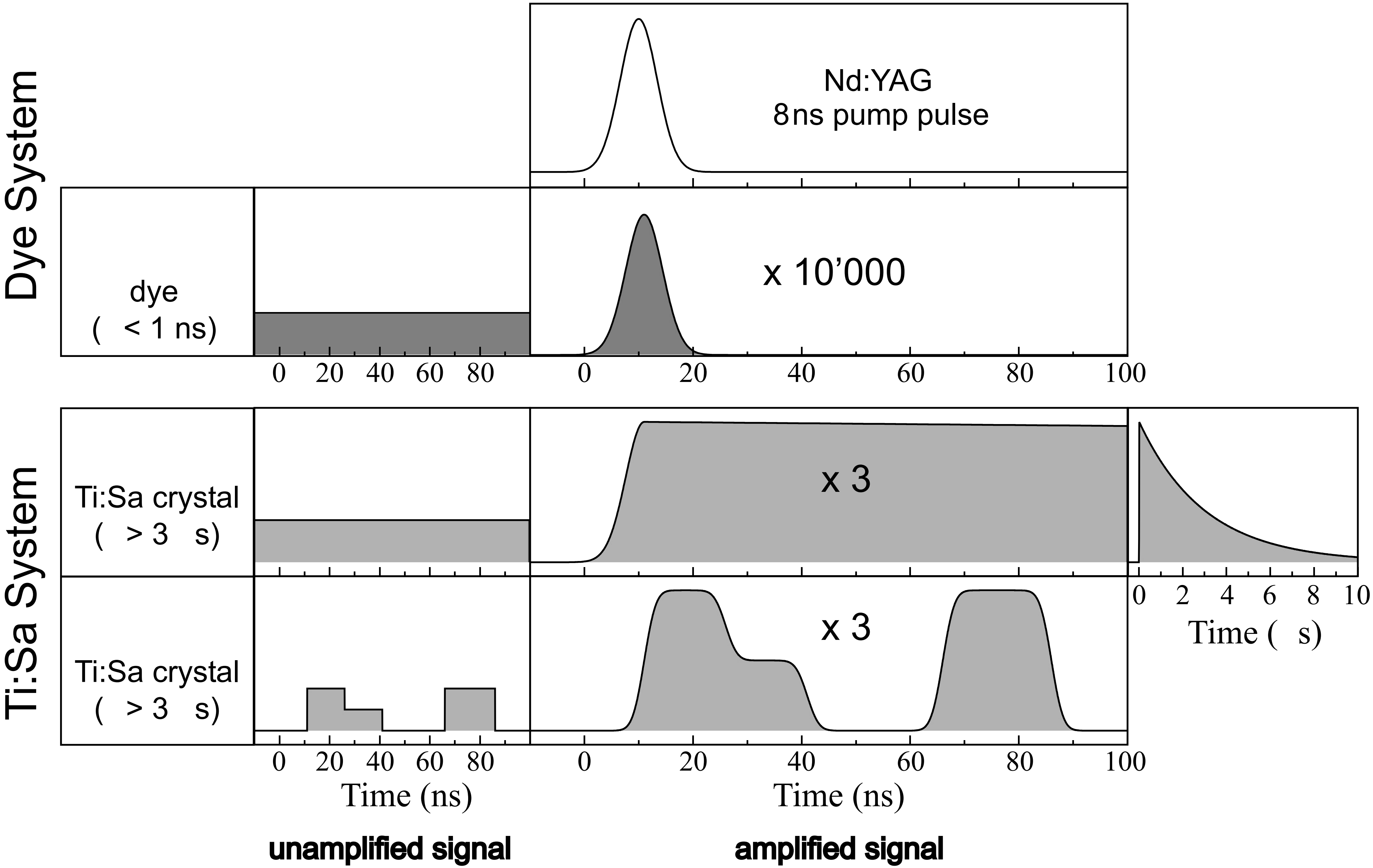
- may be augmented by adopting long pulses (ns -   s).

Realizing long seed pulses



+ NIR radiation bursts are generated by the pulsed diffraction side-band of an acousto-optic-modulator.

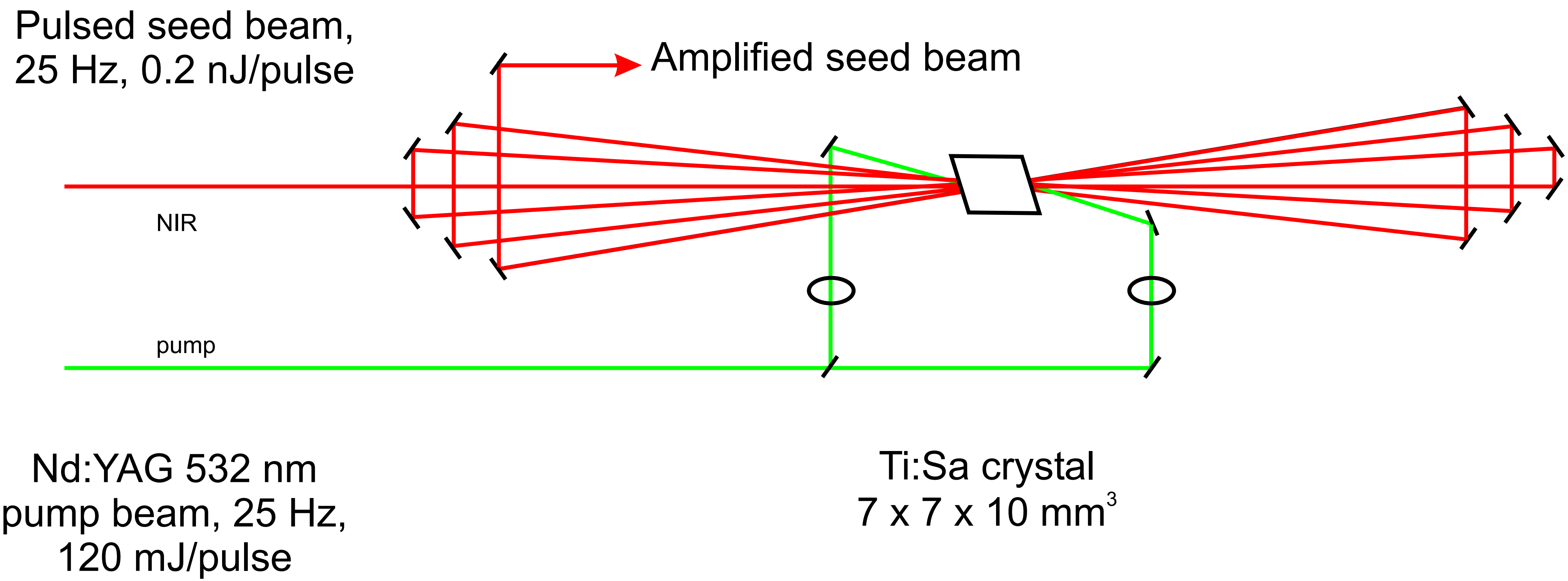
Amplifying pulses: Dye vs. Ti:Sa



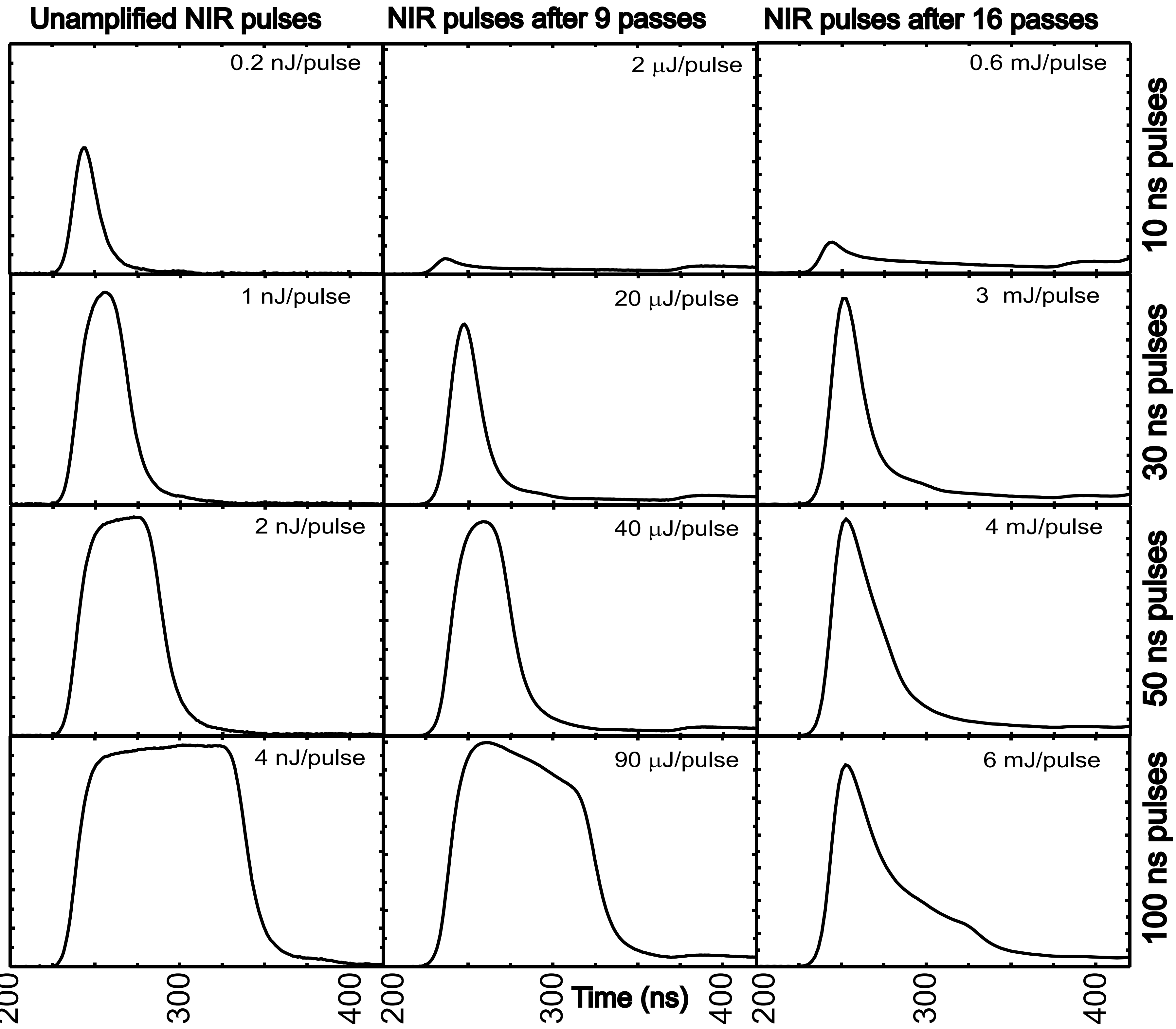
Amplifying long pulses

+ High pulse energies are required.

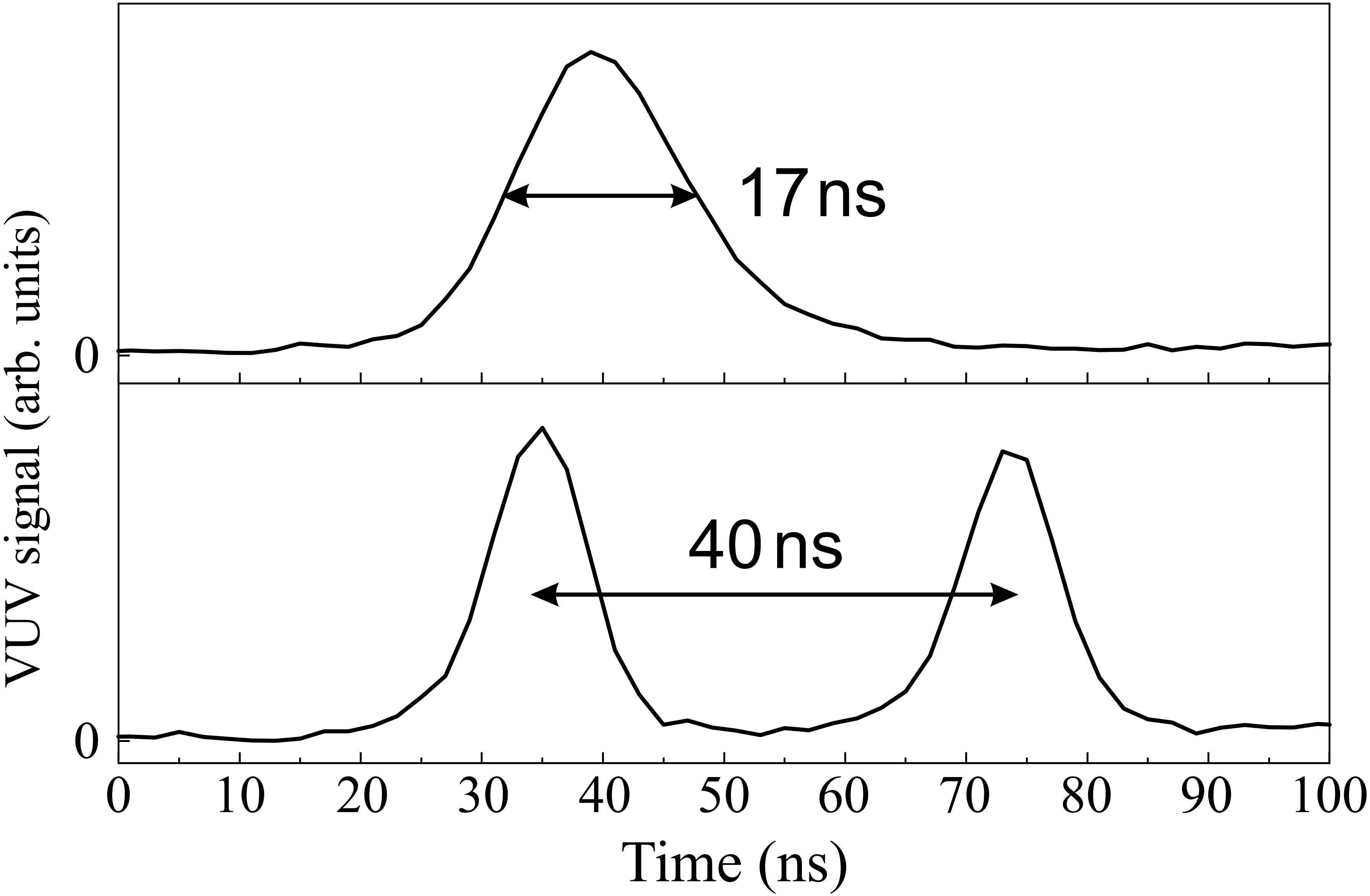
+ Long lifetime of population inversion implies low amplification factors.



Pulse shapes at different stages in the amplification process:



VUV pulses



17 ns
40 ns
VUV signal (arb. units)
0
0
0
10
20
30
40
50
60
70
80
90
100
Time (ns)

Assessment of possible chirp

- Frequency evolution during pulse:

$$= \quad {}_0 + \quad (t)$$

Cause:

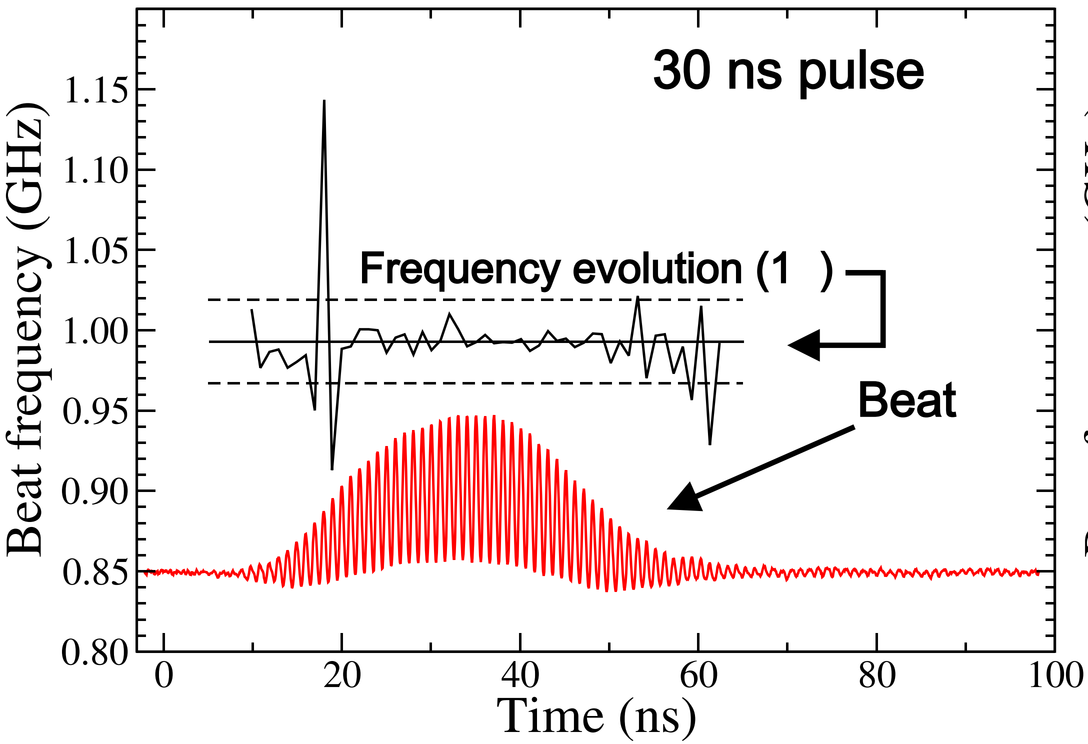
- Time dependant changes in refractive index:

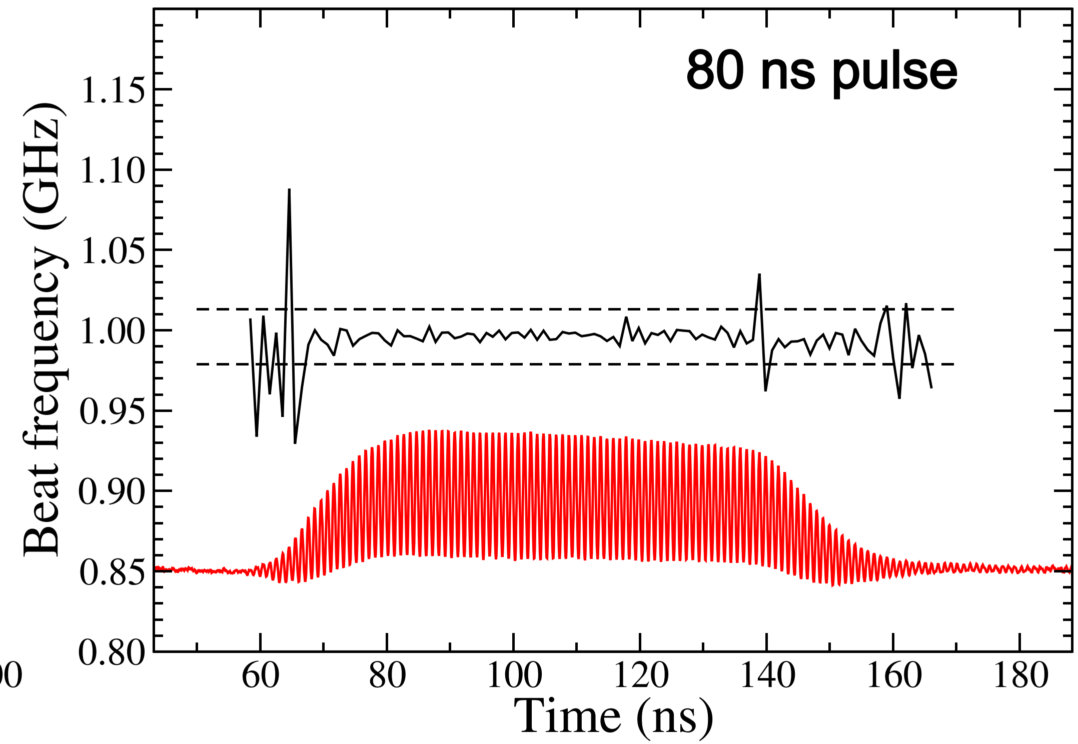
$$n(\quad ,I) = n_0(\quad) + n_1 I(t)$$

- Extracting chirp from beat pattern with a slightly frequency detuned cw reference beam:

$$I(t) = I_{cw} + I_p(t) + 2\sqrt{I_{cw} I_p(t)}\,\sin(\quad t)$$

Chirp measurement





The frequency evolution can be reconstructed from

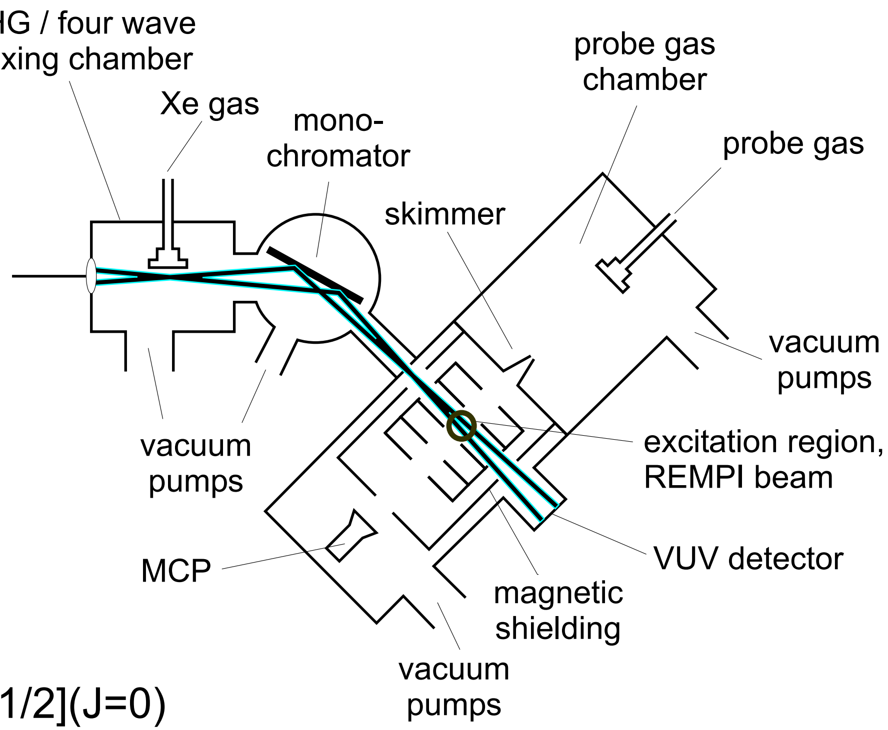
- phase information obtained from frequency filtering the FT of the heterodyne signal and subsequent inverse FFT.

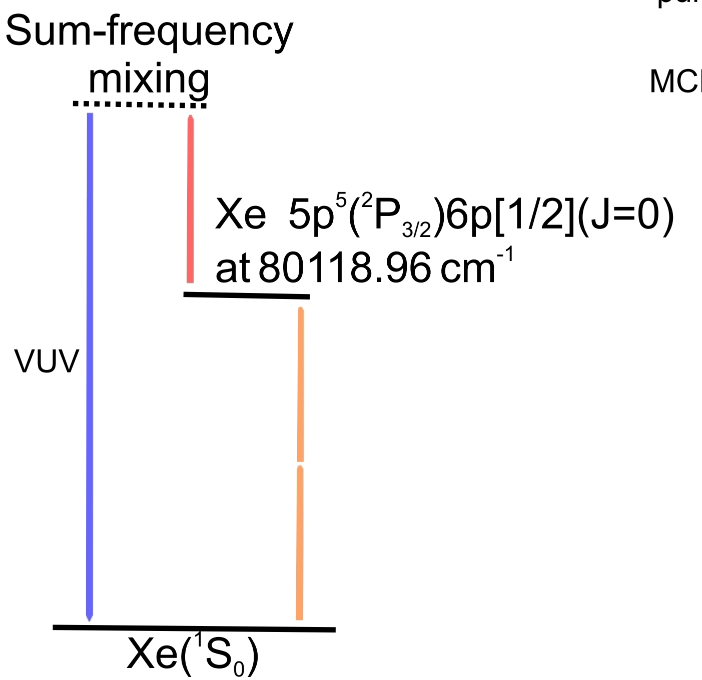
\+ No measurable frequency chirp and a constant frequency shift of (-5 ± 2) MHz exists.

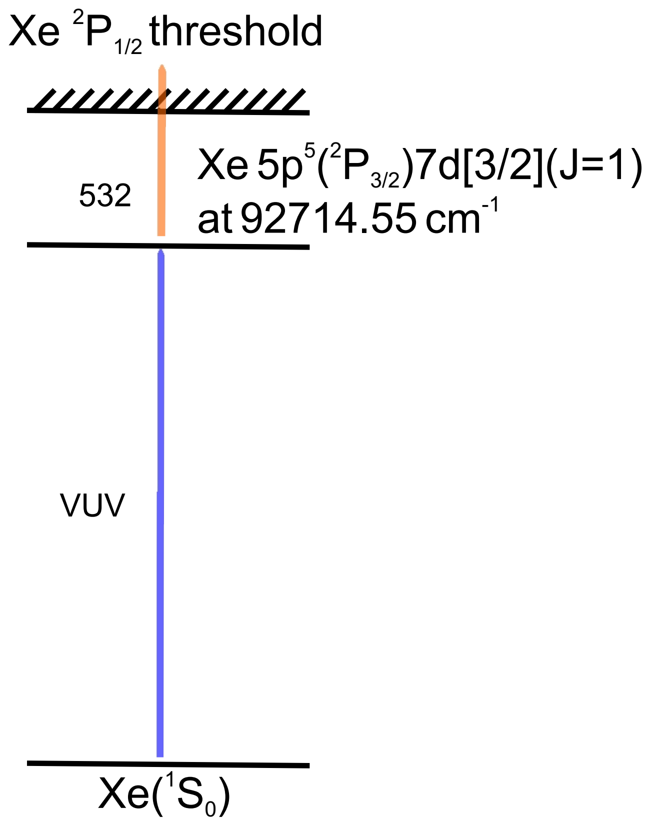
\+ VUV pulses of 17 ns 50 MHz bandwidth.

Application to atomic VUV spectroscopy

$(5p)^6\,{}^1S_0$ $5p^5({}^2P_{3/2})7d[3/2](J=1)$ resonance by $(1_{VUV} + 1_{VIS})$ REMPI:



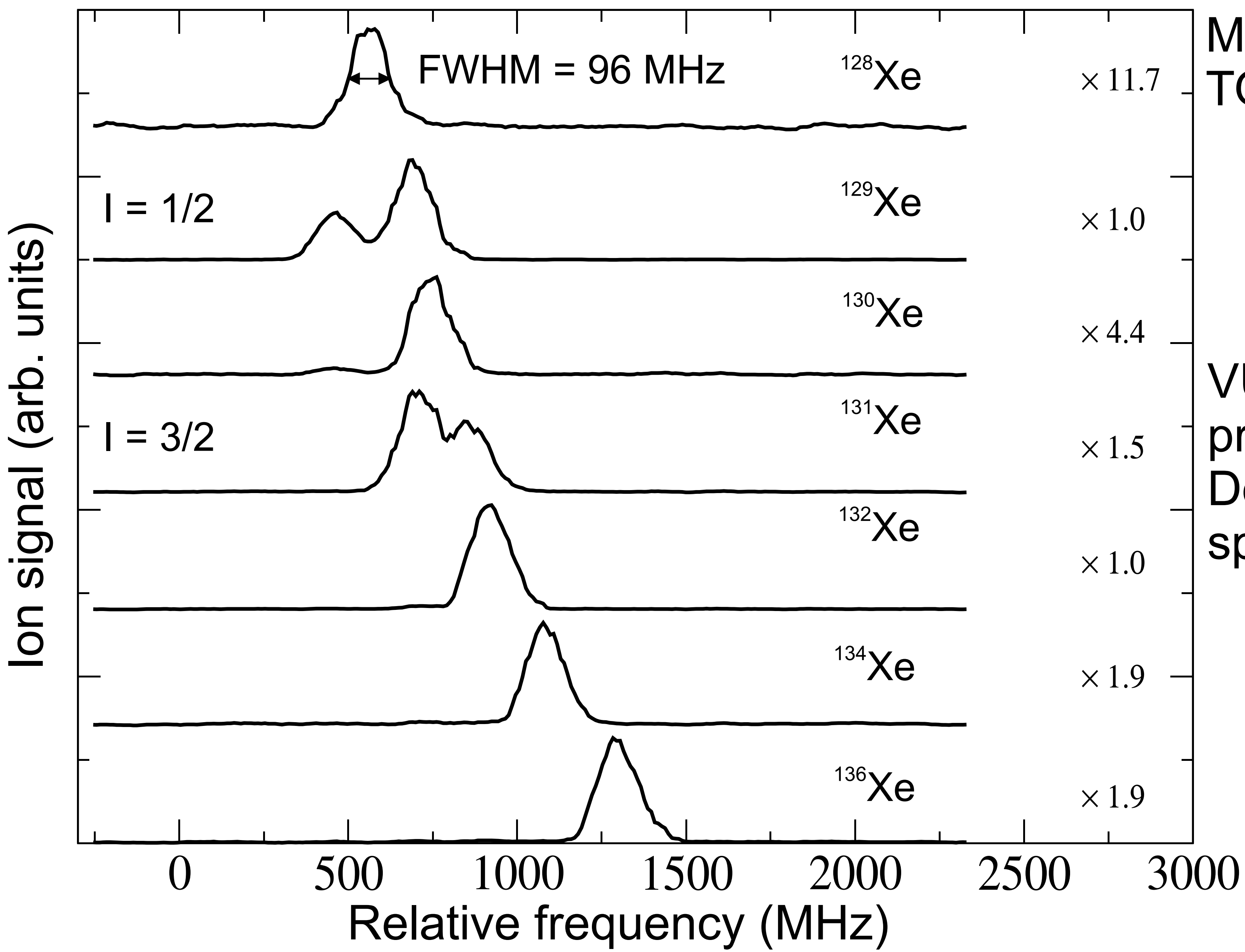




$_1$: fixed frequency

$_2$: tunable frequency

Xe $(5p)^6\ ^1S_0$ $5p^5(^2P_{3/2})7d[3/2](J=1)$ at 92714.55 cm^{-1}



Mass resolution of TOF:

m/ m > 300

VUV bandwidth predicted from Doppler-free UV spectroscopy:

50 MHz

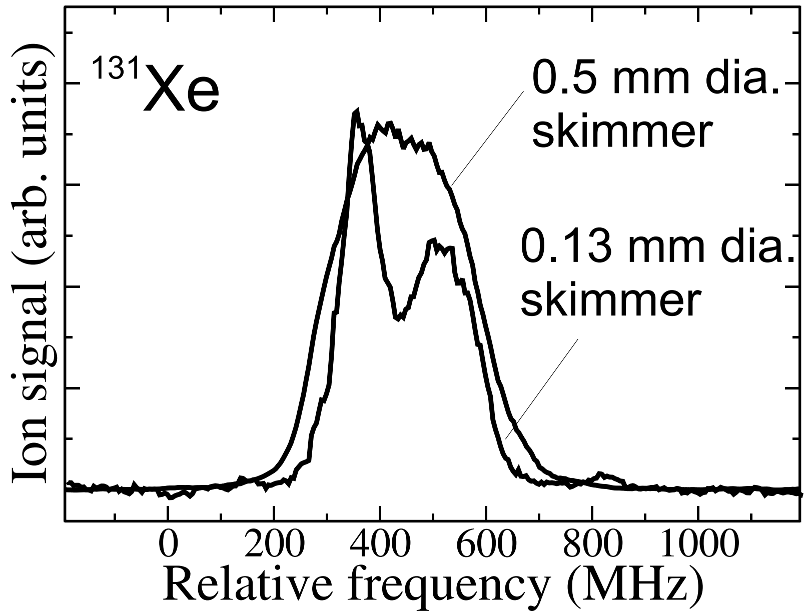
Approaching the FT-limit

Reducing Doppler-broadening by:

- use of small skimmer and nozzle.

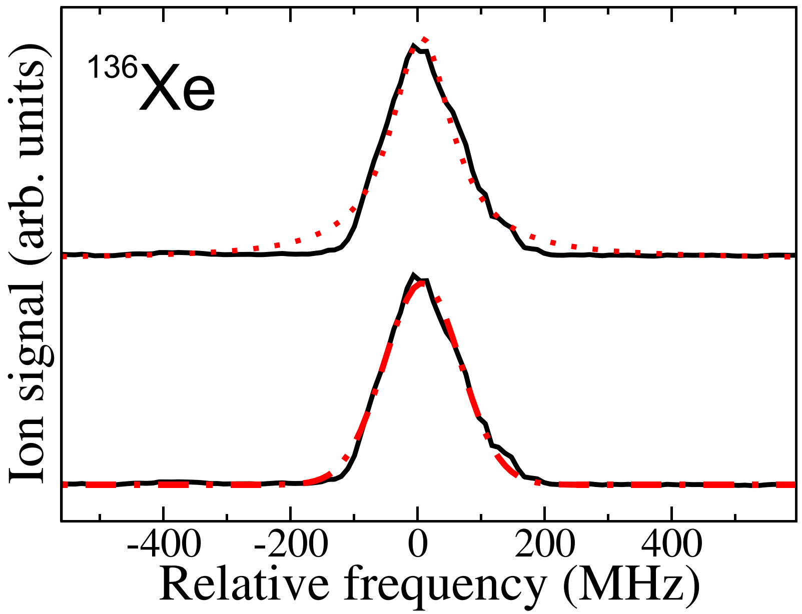
Reducing power broadening by:

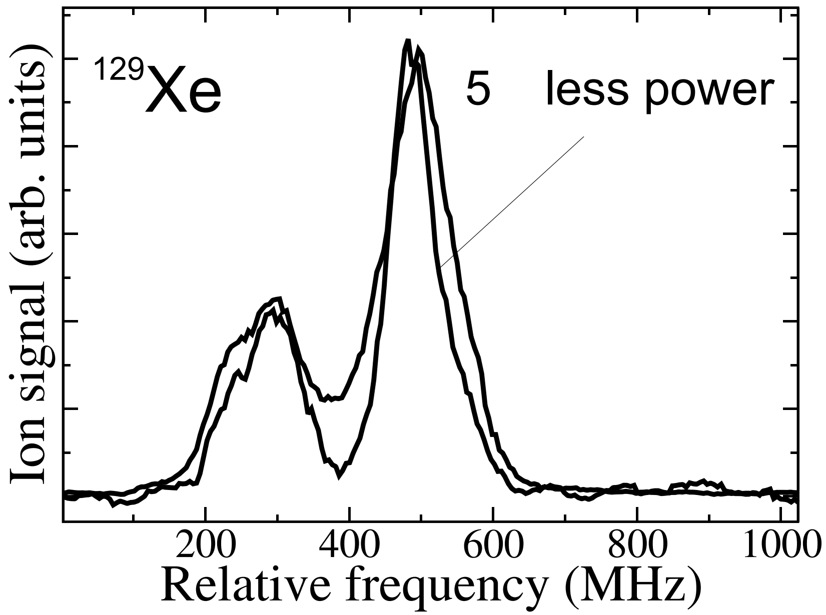
- moderate VUV intensities.



Lorentz fit

Gauss fit



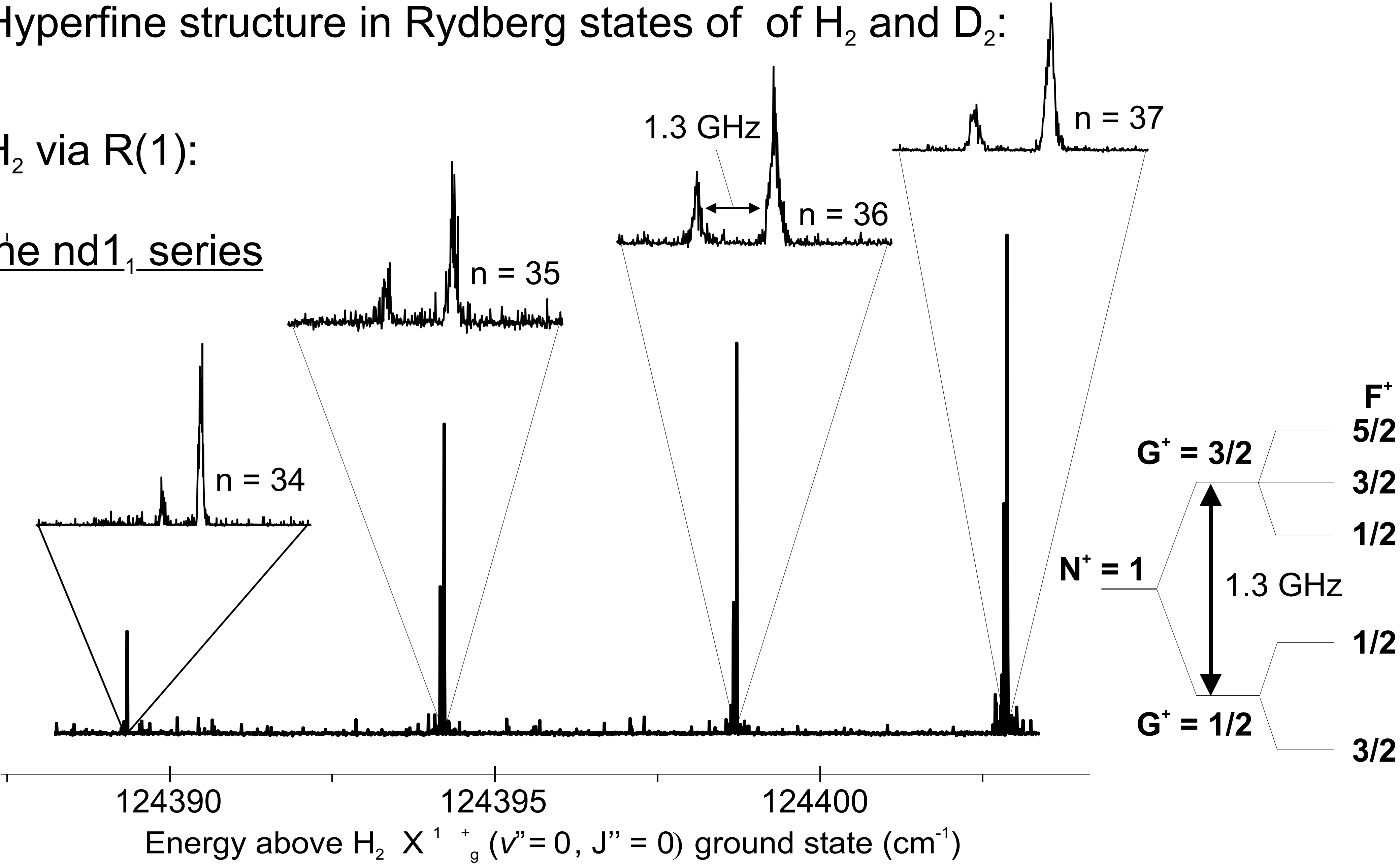


Application to molecular VUV spectroscopy

Hyperfine structure in Rydberg states of of H_2 and D_2:

H_2 via R(1):

the $nd1_1$ series



Conclusion

+ Realization, characterization and application of a new high resolution (0.003 cm^{-1} at 93000 cm^{-1}, Doppler-limited), tunable all-solid-state VUV system was presented.

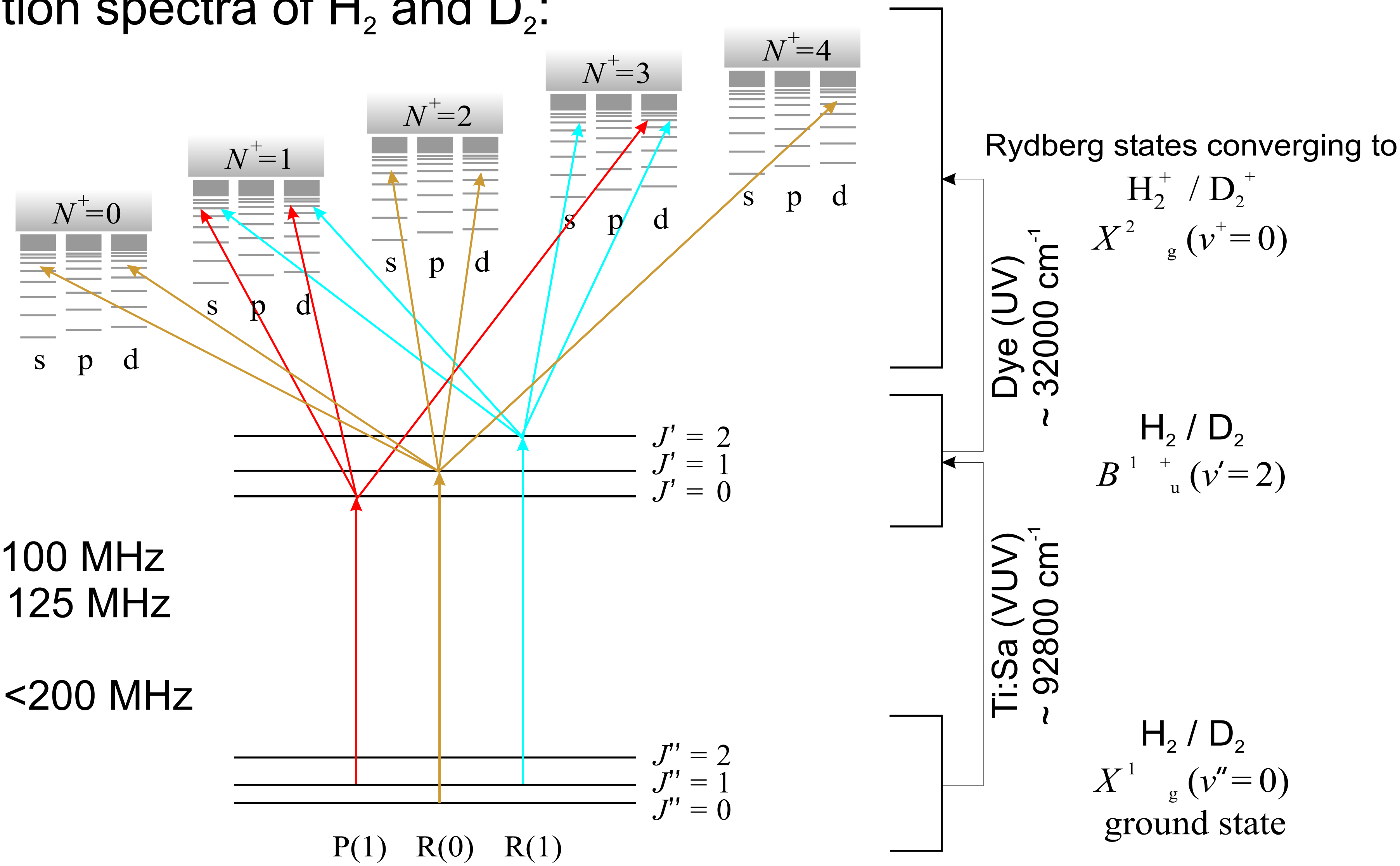
+ Measurements of the $^1S_0$ Xe $5p^5(^2P_{3/2})7d[3/2](J=1)$ resonance revealed, for the first time, the hyperfine splittings of the $I \neq 0$ isotopes.

+ Measurements of the high lying nd Rydberg series of H_2 and D_2 have been performed revealing hyperfine structure.

+ The narrow bandwidth and wide tunability attainable will enable the exploration of the finest details in VUV photochemistry and photophysics.

Applications

High-resolution spectra of H_2 and D_2:



Bandwidth:

Ti:Sa (VUV): 100 MHz
PDA (UV): 125 MHz

Combined: <200 MHz