Laser Spectroscopy of HeH^+

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2011 AMO TALK

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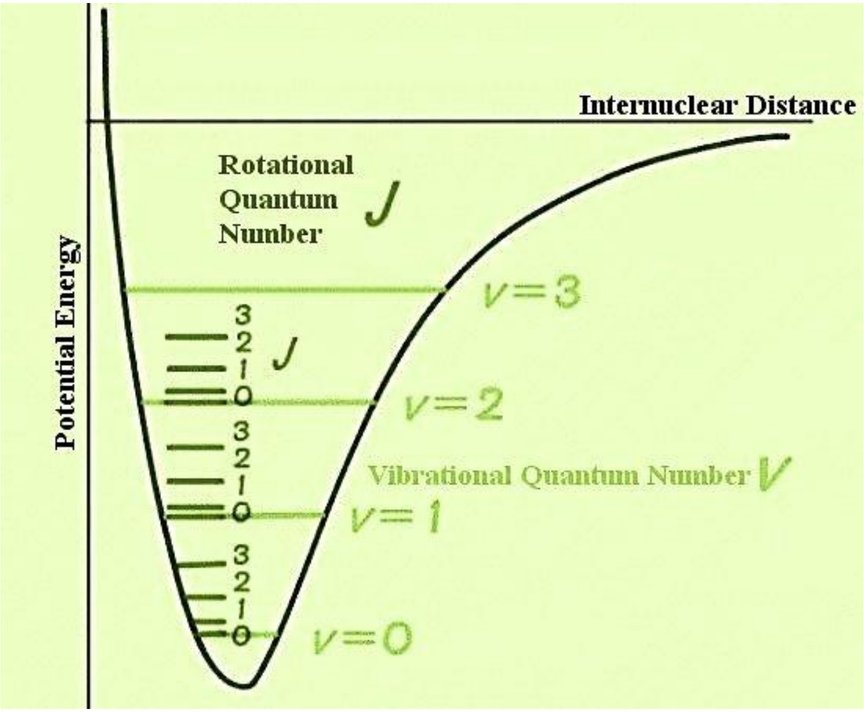
Outline

- Introduction
- Previous experimental results
- Saturation spectroscopy
- Conclusions and future works

Diatomic Molecules

- Total energy=electronic energy + vibrational energy + rotational energy

$$E_{tot} = E_{elec} + E_{vib} + E_{rot}$$



E_{elec} ~ *few eV*

$E_{vib} \sim E_{elec}/100$

$E_{rot} \sim E_{vib}/100$

Rovibronic transition Vis, UV

Rotation-vibrational IR

Rotational μwave, THz

Simple Diatomic Molecules

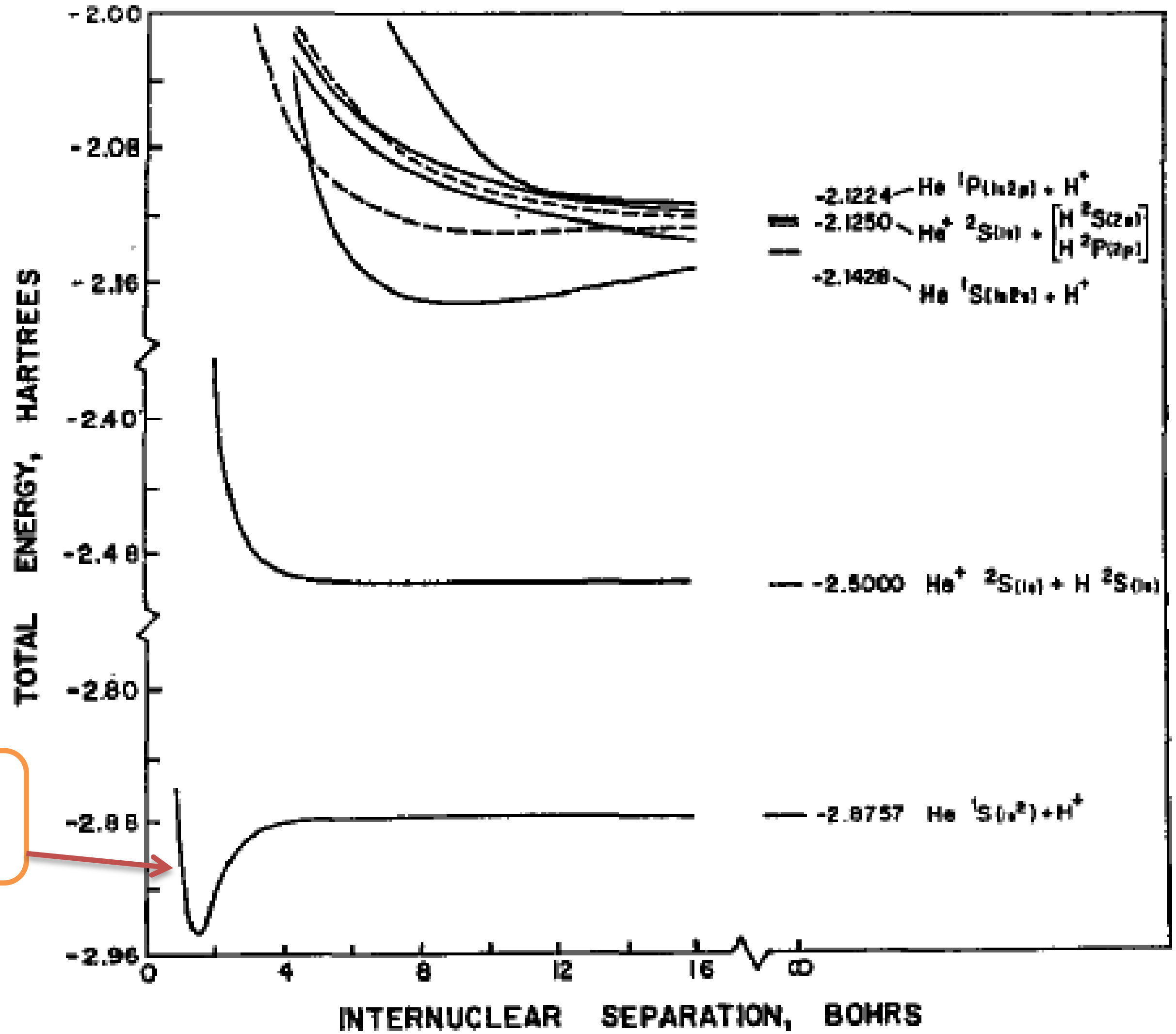
- One-electron: H_2^+, HD^+, D_2^+
- Two-electron: H_2, HD, D_2, HeH^+

HD^+, HeH^+: electric dipole moment for rotation-vibration transitions

Why HeH^+?

- One of the simplest two-electron diatomic molecules: benchmark of theoretical calculation
- It is formed by hydrogen and helium, the two most abundant elements in the universe. It is the first molecule in the universe.
- HeH^+ had been suggested presenting in the astronomical objects such as nebula, super nova, white dwarfs and quasi-stellar object (QSO)envelop.

Potential curves of HeH^+



Ground electronic level

H.H. Michels, J. Chem. Phys., 44, 3834 (1966).

Theoretical calculations

- Born-Oppenheimer (BO) approximation
- Non-adiabatic calculation
- Relativistic corrections
- QED corrections

Generation and destruction of HeH^+

- Generation

$$He^* + H_2 \rightarrow HeH^+ + H + e^-$$

$$H_2^+(v \geqq 3) + He \rightarrow HeH^+ + H$$

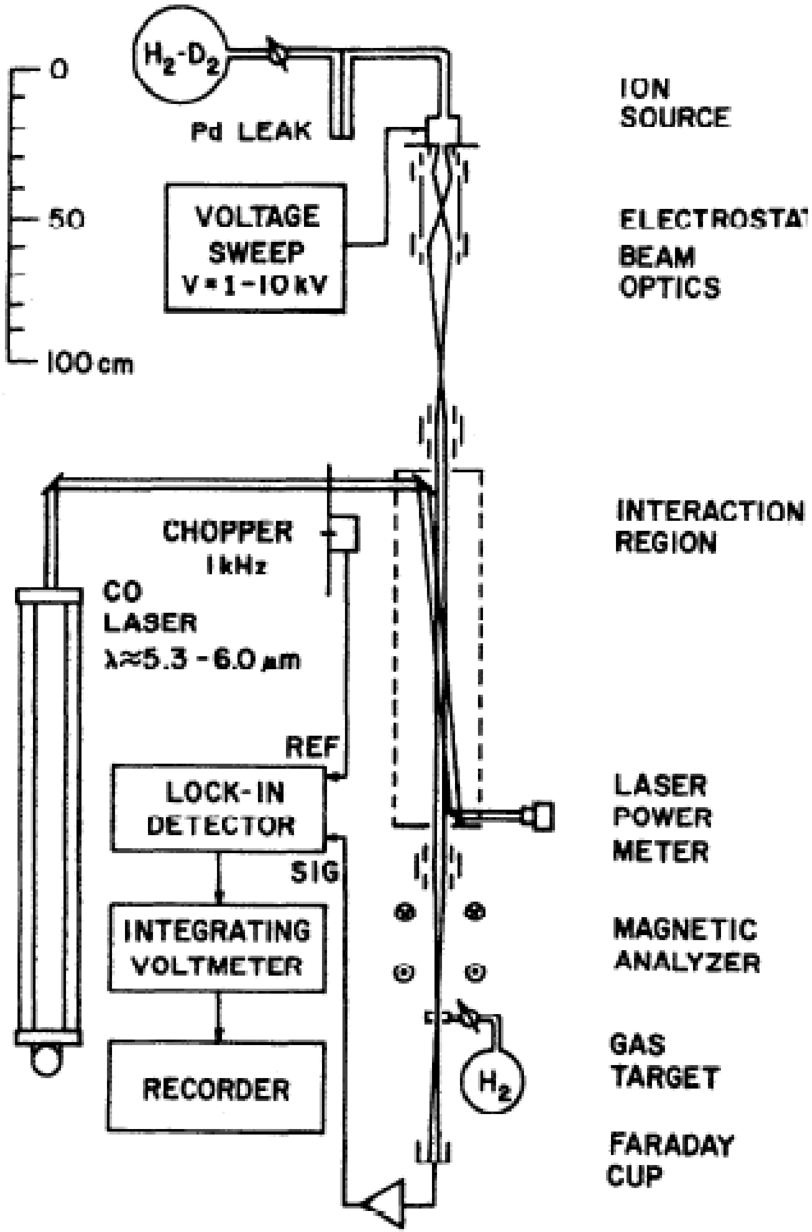
- Destruction

$$HeH^+ + H_2 \rightarrow He + H_3^+$$

Spectroscopic methods

- Doppler-tuned ion beam spectroscopy
- Laser absorption spectroscopy in discharge

Spectroscopy Using Ion Beam



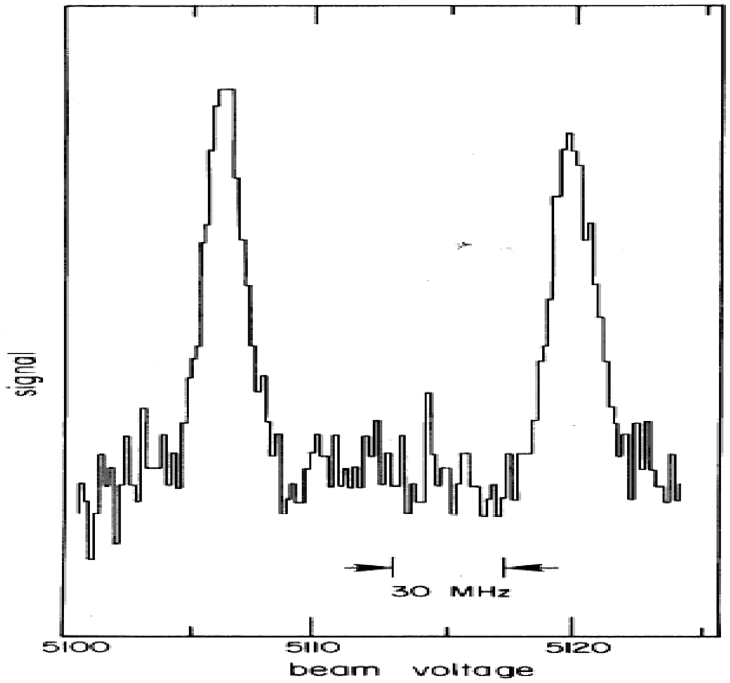
Willis Lamb (1913 -2008)
1955 Nobel prize
Mossbauer effect
Lamb shift
Laser physics
Lamb dip

William H. Wing
University of Arizona

Doppler tuning: tuning range depends on laser frequency, ion mass and accelerating voltage

High resolution: velocity compression
Linewidth ~ 10 MHz

Collision detection: low signal-to-noise ratio



Spectroscopy Using Discharge

Gerhard Herzberg (1904-1999)
1971 Nobel prize in chemistry

Takashi Oka
First observation of H_3^+ IR spectrum

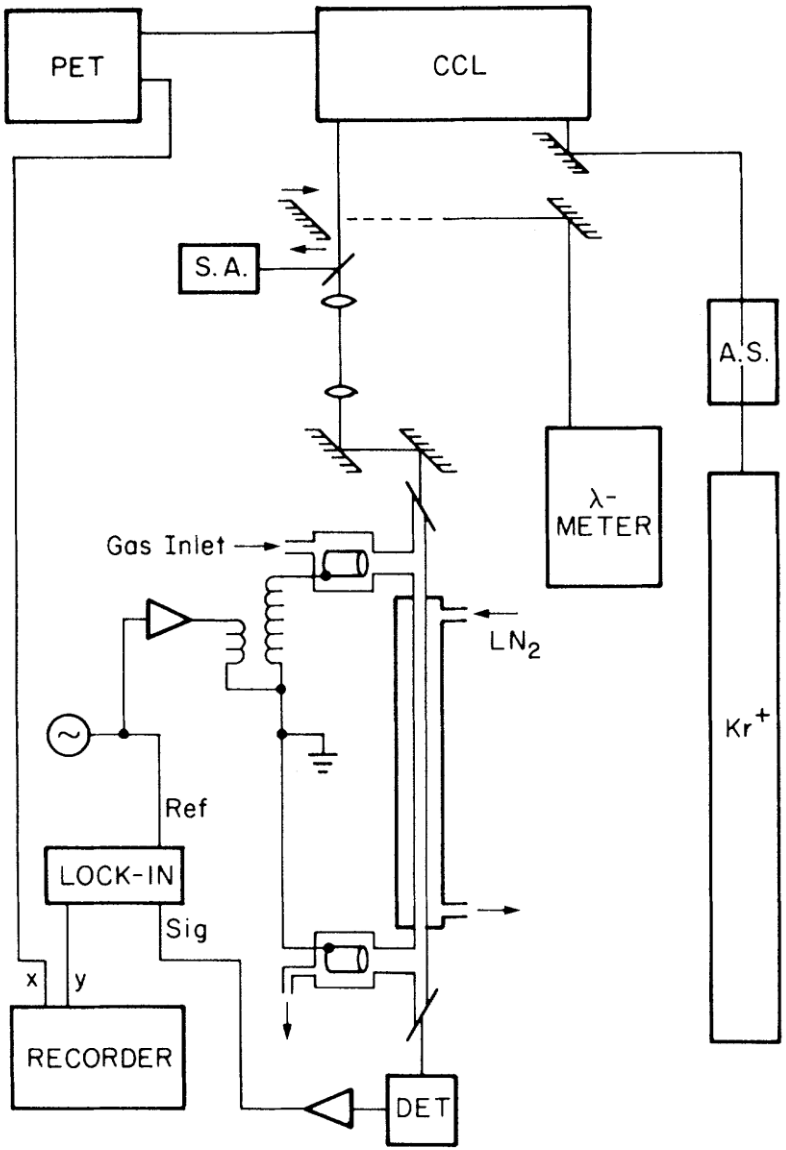
Observation of H_3^+ IR spectrum by Oka (1980)

- Tunable DFG source
- LN2 cooled hydrogen discharge
- Absorption spectrum with 32 m length

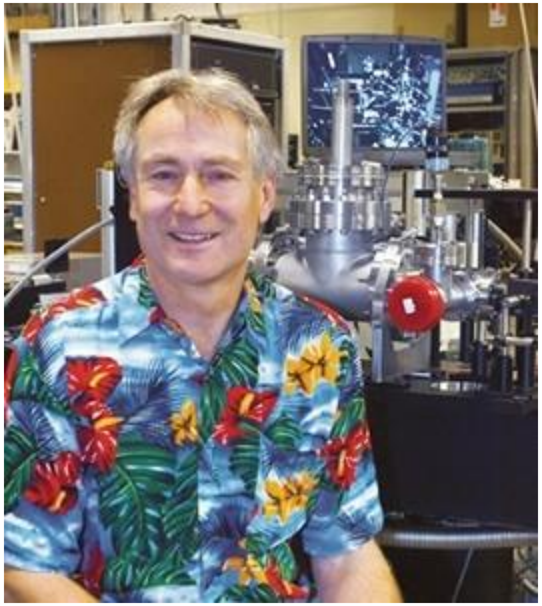
Observation of HeH^+ fundamental band spectrum by Bernath and Amano (1982)

4% absorption for R(1) transition in fundamental band

Velocity Modulation Spectroscopy



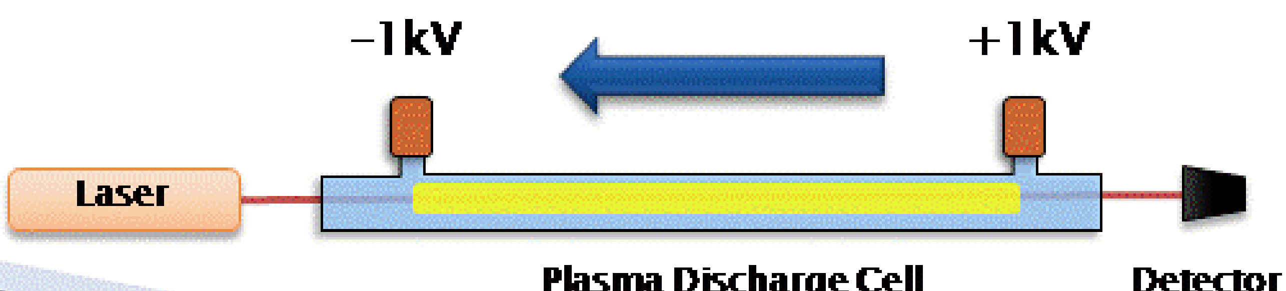
Sensitive to ions not to neutrals



Richard Saykally (UC Berkeley) 1983

Velocity Modulation Spectroscopy

- Positive column discharge cell
  - High ion density, rich chemistry
  - Cations move toward the cathode
  - Ions absorption profile is Doppler–shifted



Taken from Prof. B.J. McCall’s talk

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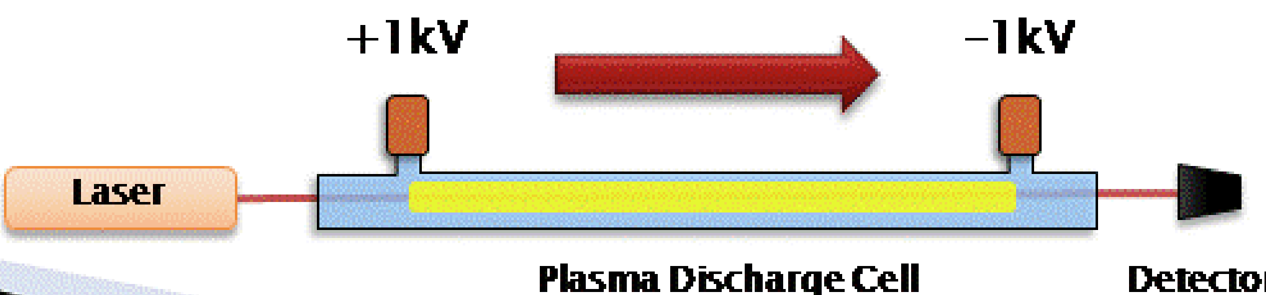

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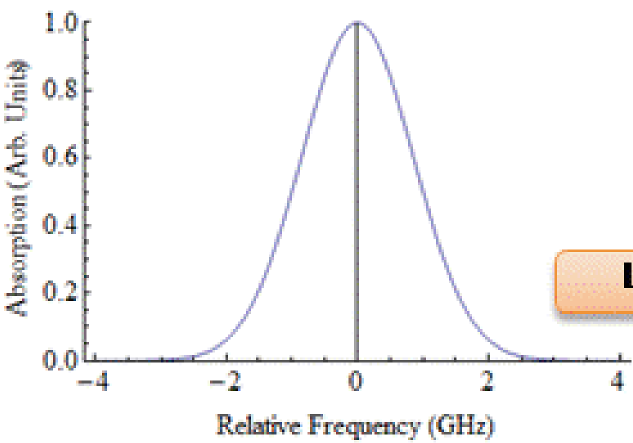
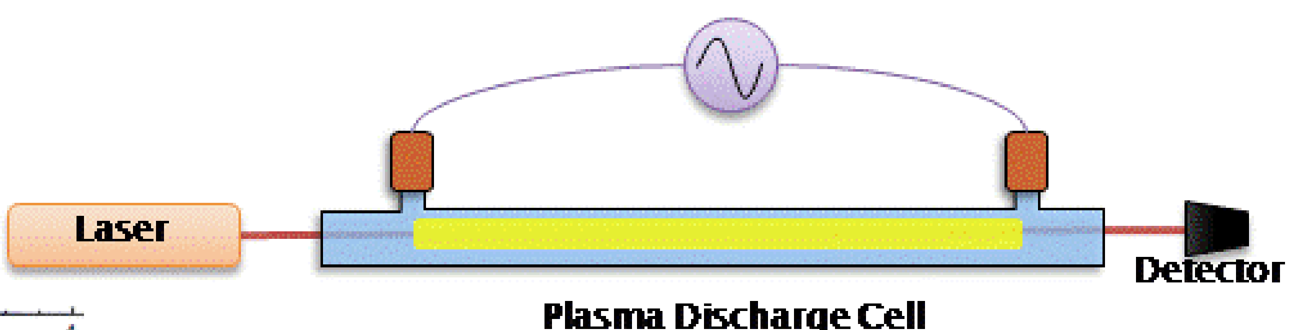
Velocity Modulation Spectroscopy

- Positive column discharge cell
  - High ion density, rich chemistry
  - Cations move toward the cathode
  - Ions absorption profile is Doppler-shifted
- Drive with AC voltage
  - Ion Doppler profile alternates red/blue shift
  - Laser at fixed wavelength
  - Demodulate detector signal at modulation frequency





Taken from Prof. B.J. McCall's talk

Laser Spectroscopy of $^{4}HeH^{+}$

Authors	Method	Measured Transitions	Accuracy (MHz)
[1979] D.E. Tolliver, G. A. Kyrala and W.H. Wing	Ion beam	Fundamental Band : P(12)-P(13) v = 2-1 : P(9)-P(11)	60
[1982] P. Bernath and T. Amano	Absorption	Fundamental Band : R(0)-R(4) and P(1)-P(4)	30~60
[1989] M.W. Crofton, R.S. Altman, N.N. Haese and T. Oka	VM	v=1-2 R(1)-R(5), R(8), P(1) and its isotopes	60
[1997] F. Matsushima, T. Oka and K. Takagi	VM	Low J pure rotation R(0) and R(1)	0.2
[1997] Z. Liu and P.B. Davies	VM	Quasi-bound to quasi-bond and bond to quasi-bond	90

HeH^+ Pure Rotation Transitions

- Tunable FIR source:

  two CO_2 lasers + microwave + MIM diode

  frequency up to 6 THz

  μW power

  LHe cooled bolometer

Oka and Takaji 1997

The lowest *J = 1 → 0 transition of* $^4HeH^+$ observed at 2010.1839(2) GHz will be an important future probe for detecting this species in space.

Saturation Spectroscopy of Molecular Ions (H_3^+ and HeH^+)

How to achieve?

> **Light Source – CW Optical Parametric Oscillator**

---- Enough power for saturation.

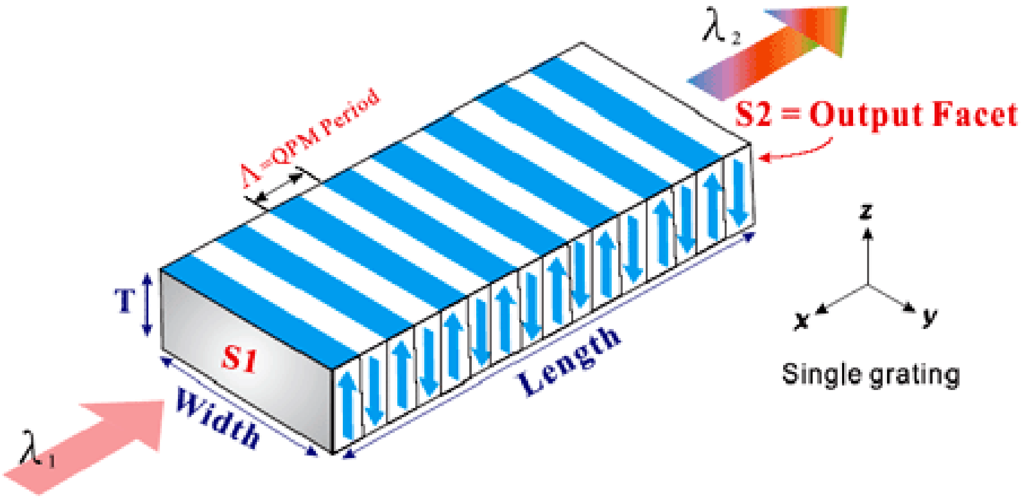
> **Extended Negative Glow Discharge Tube**

---- Low pressure, high concentration of ion.

> **Optical Frequency Comb**

---- Frequency ruler for precision measurement.

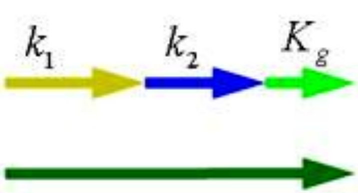
PPLN (periodically poled $LiNbO_3$)



Quasi-phase matching

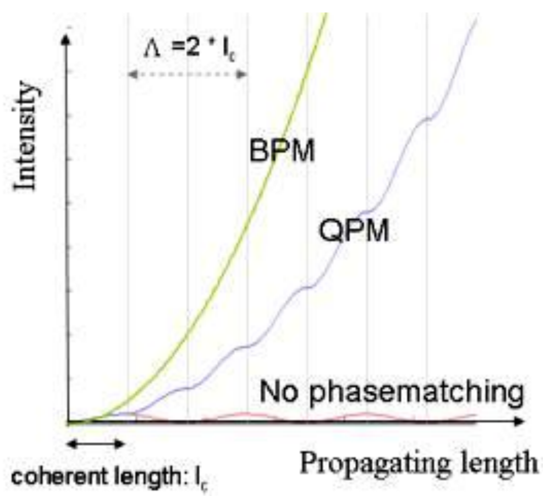
High nonlinear efficiency

over 0.4 to 5 μm



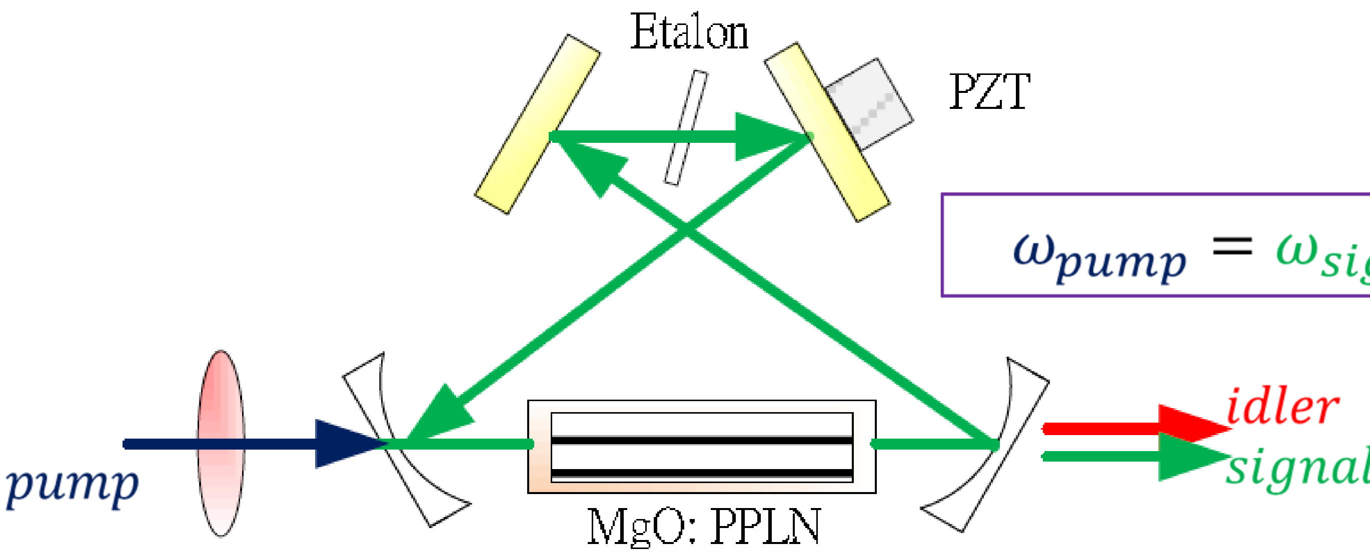
$$k_3 = k_1 + k_2 + K_g$$

$$K_g = \frac{2\pi}{\Lambda}$$



Taken from HC Photonics

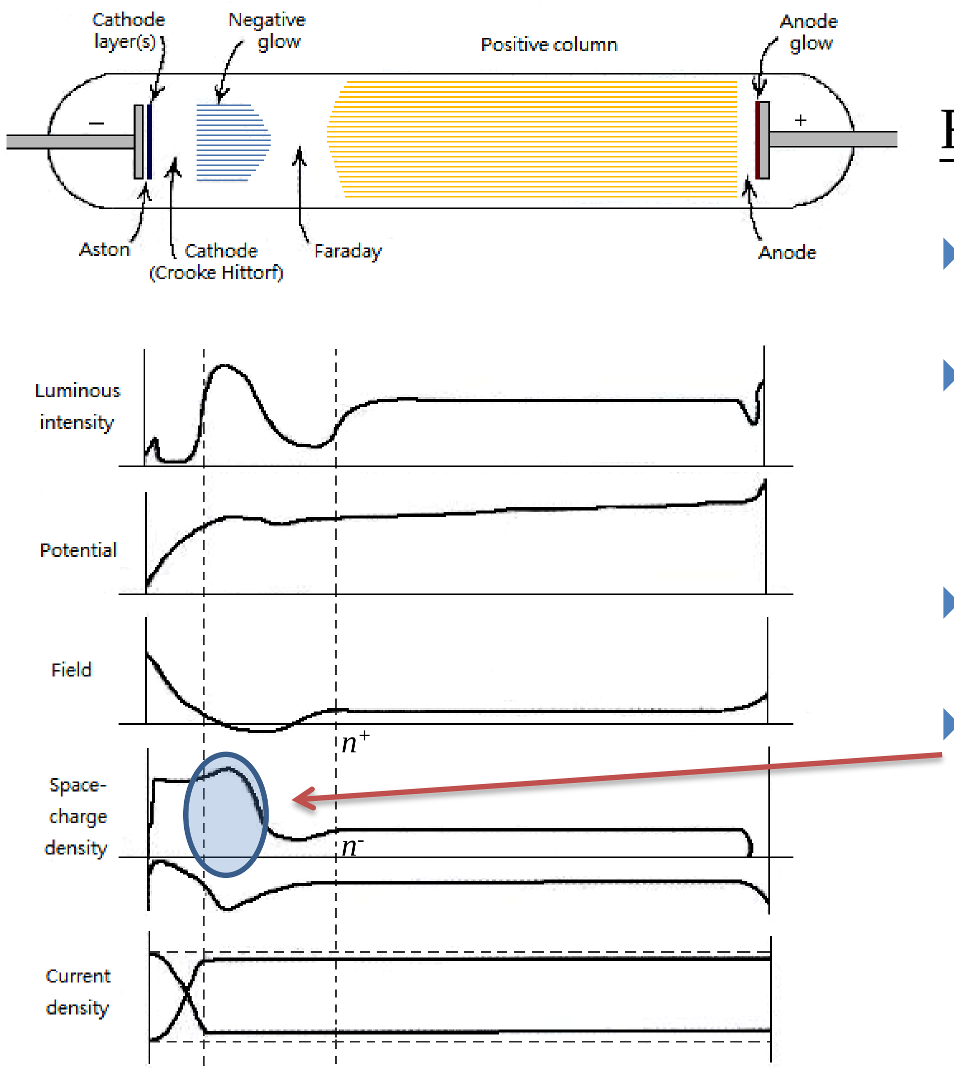
Singly Resonant PPLN OPO



$$\omega_{pump} = \omega_{signal} + \omega_{idler}$$

- Unlike other MIR light source, OPO provides larger power which is benefit for the saturation spectroscopy of molecular ion.
- Idler Wavelength: **2.7 – 3.9 μm**
- Average Power: > **300 mW**
- Frequency Tuning: **> 40 GHz (Mode-Hop-Free)**
- Free-Running Stability: **< 500 MHz @ 8 hours**
- Pump: **1062 nm alpha-DFB laser boosted to 6 W by a YDFA**

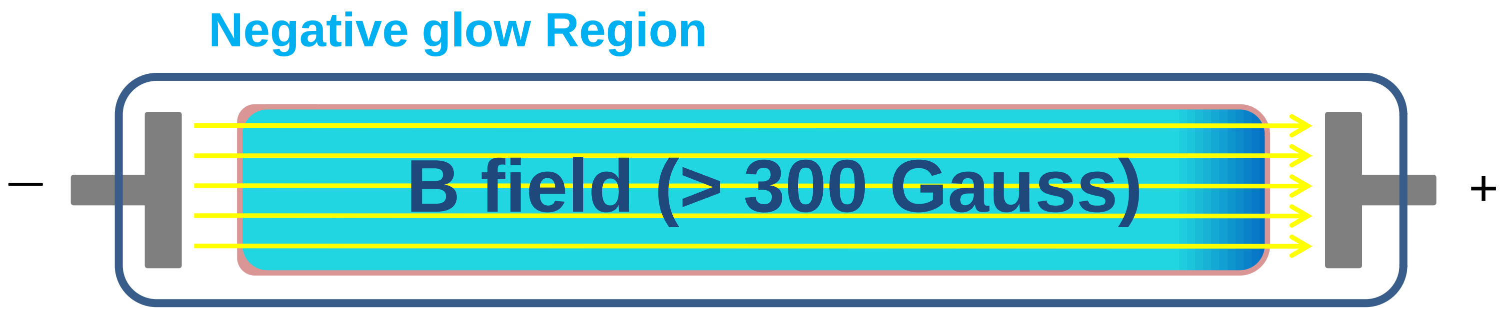
Glow Discharge



Features of negative glow region:

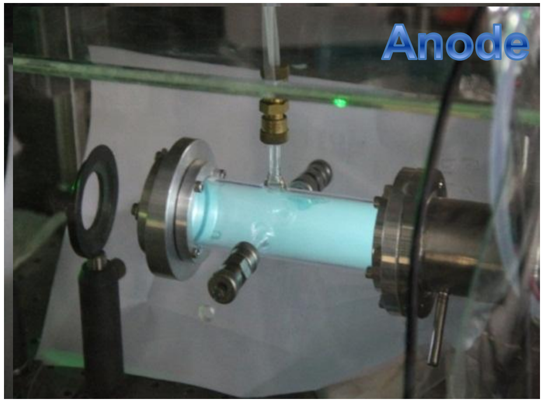
- Largest glow intensity
- Highest concentration of positive ions
- Nearly field free
- **The region is relatively short**

Extended Negative Glow Discharge



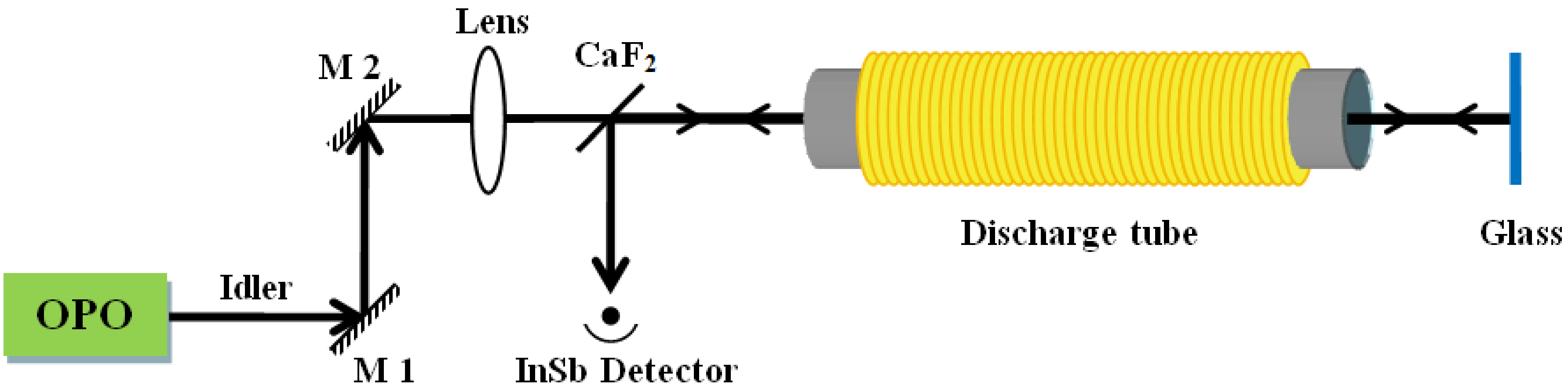
Benefits of extended negative glow :

- **Lower gas pressure (~100 mtorr)**
- **Narrower linewidth for precision measurement**



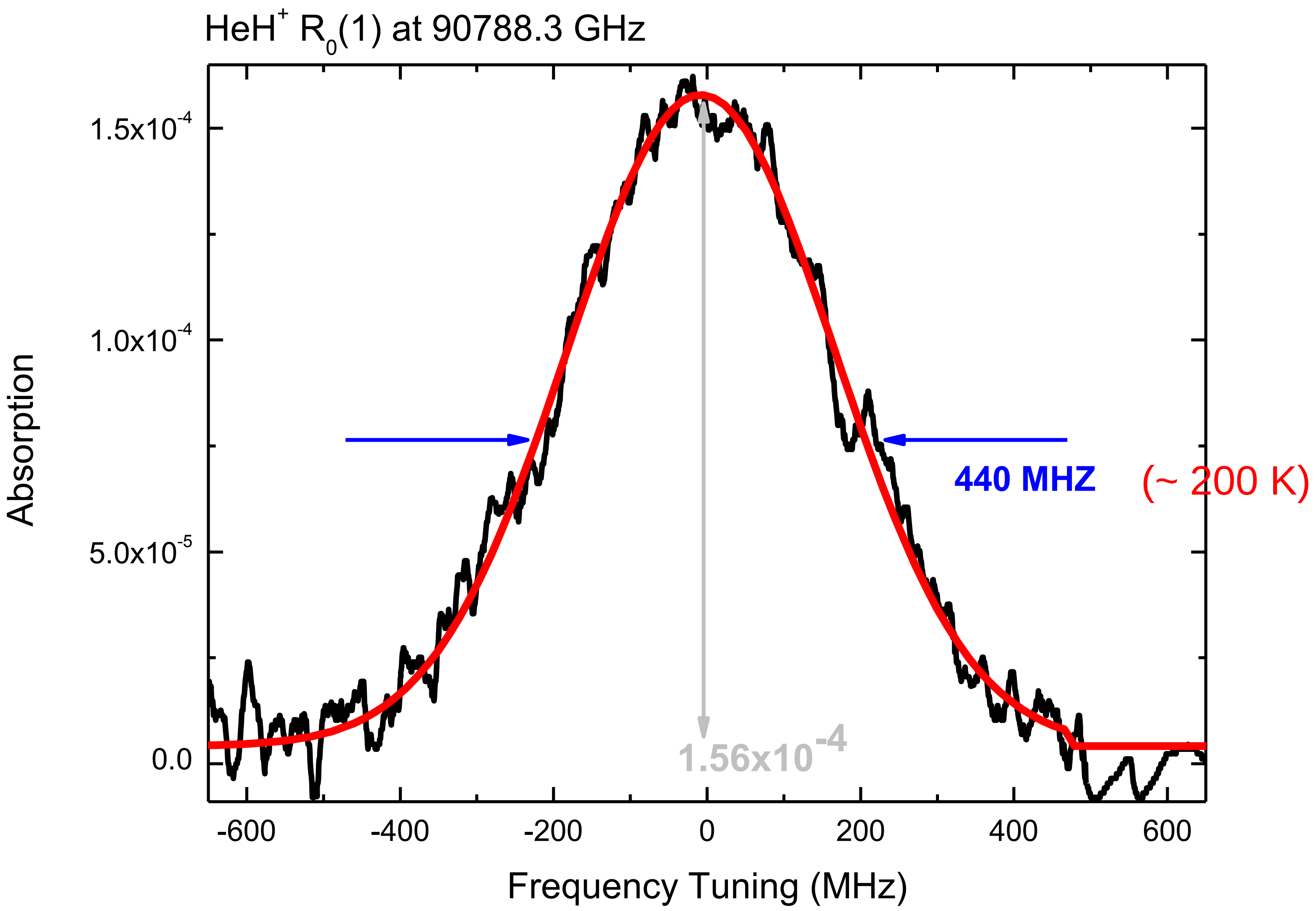
- Flow mixture of He:H_2 = 98 : 2 (120 mtorr)
- Discharge Current : 16 mA
- Ethanol cooled at -70 °C
- Discahrge length = 150 cm

Experimental Setup for saturation spectroscopy

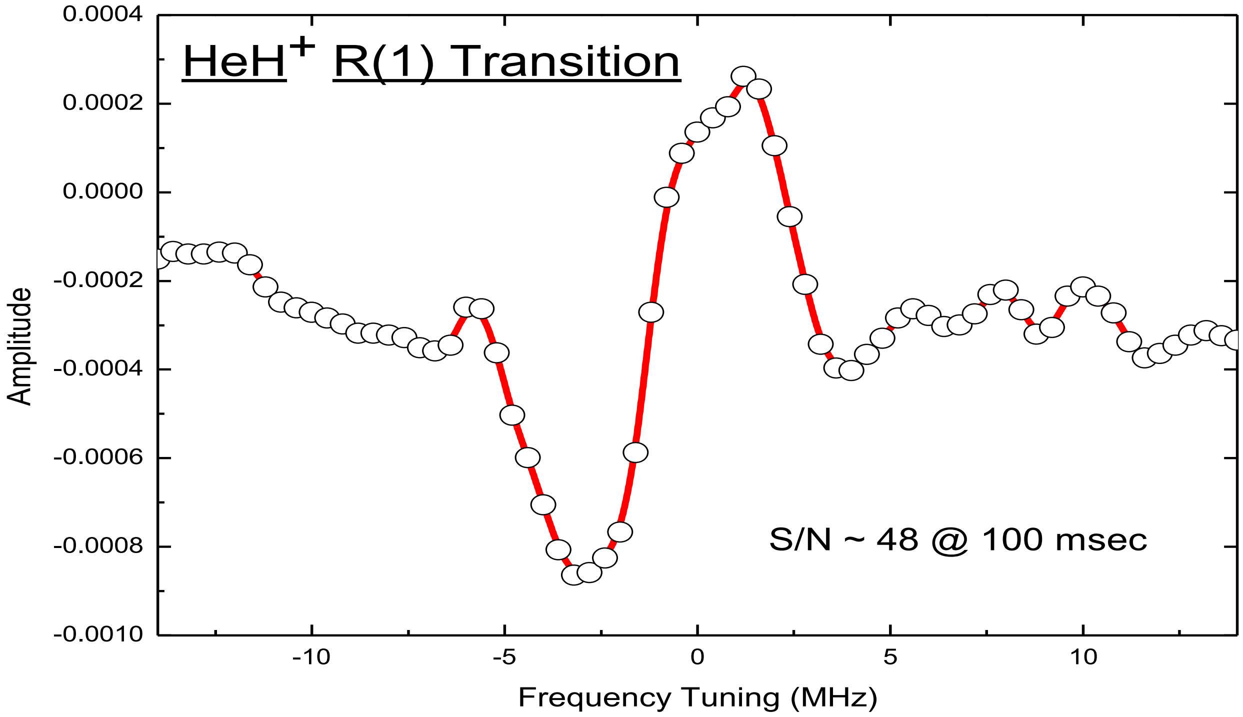


Pump Power: ~110 mW
Beam Size: 1.75 mm (Diameter, $1/e^2$)
Probe beam ~ 10 mW
The intensity is ~ 40 kW/m^2

Doppler Broadened Spectrum

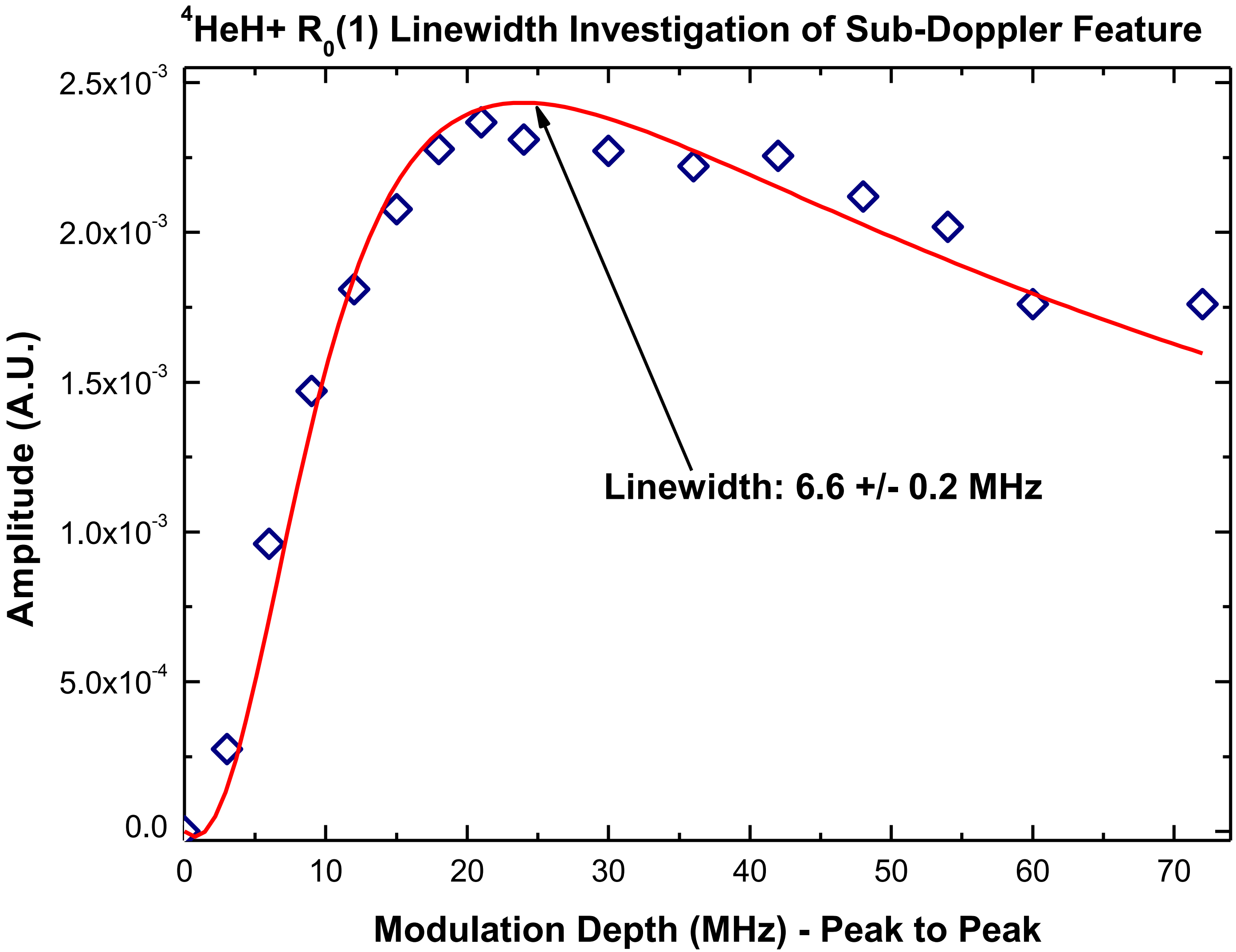


3rd Derivative Spectrum



Frequency Modulation Method (by modulating the pump frequency)
Modulation Frequency : 31 kHz
Modulation Depth : 7.6 MHz
Laser Intensity : ~ 40 kW/m^2 (or 40 mW/mm^2)

Linewidth analysis



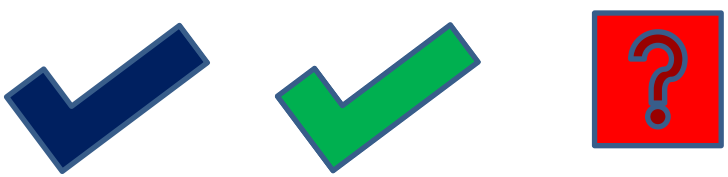
Fitting Function : HM Fang et. al., Opt. Comm., **257**, 1, 76-83 (2006)

ITRI Fiber OFC

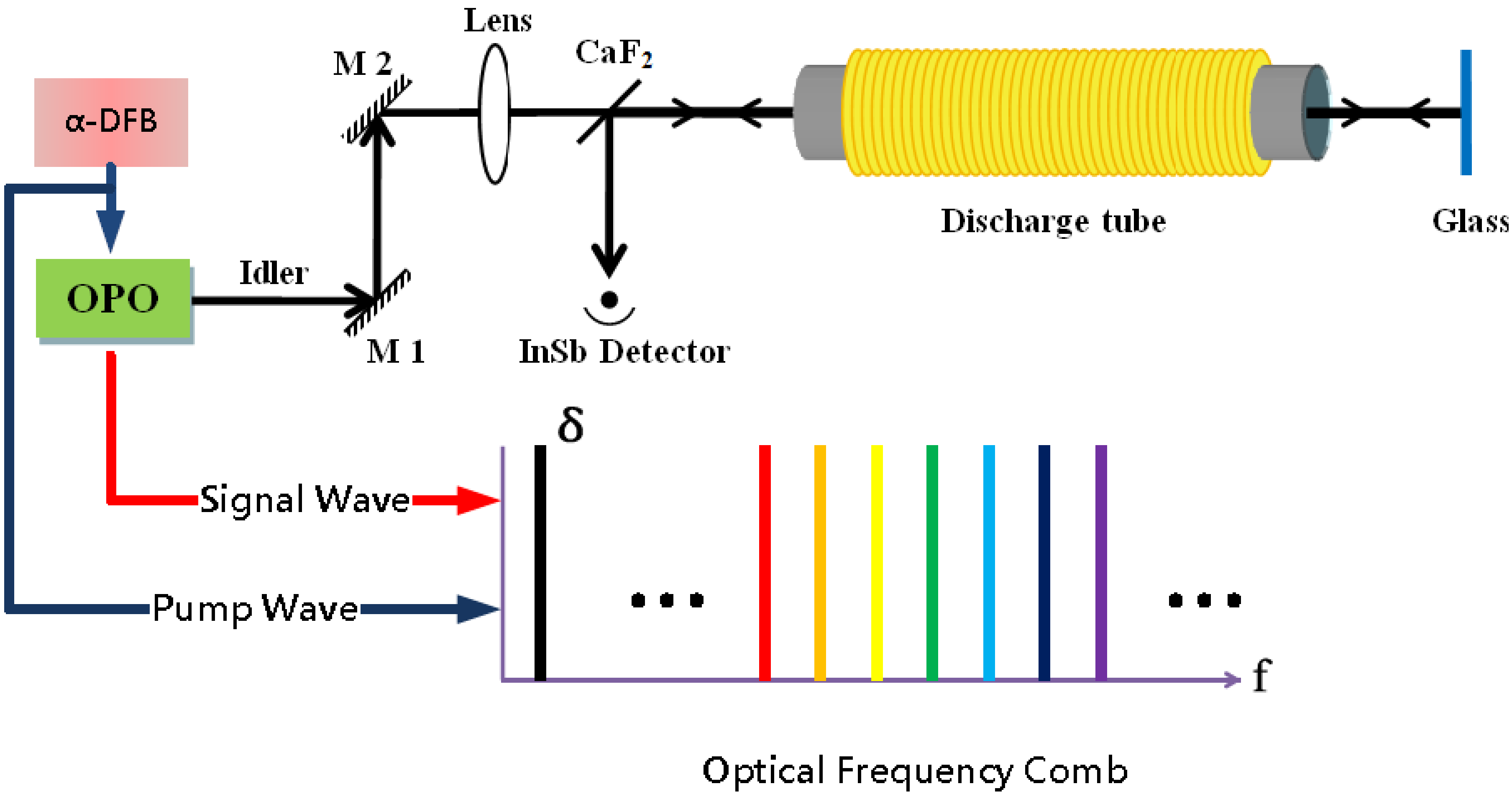
- Repetition Rate : 250 MHz
- Supercontiuum : 1030 ~ 2200 nm
- RF Reference : GPS-locked Rubidium clock
- Accuracy : < 10^{-11} @ 1000 sec (~few kHz in the MIR region)

1.062 μm 1.4-1.8 μm 2.7-3.9 μm

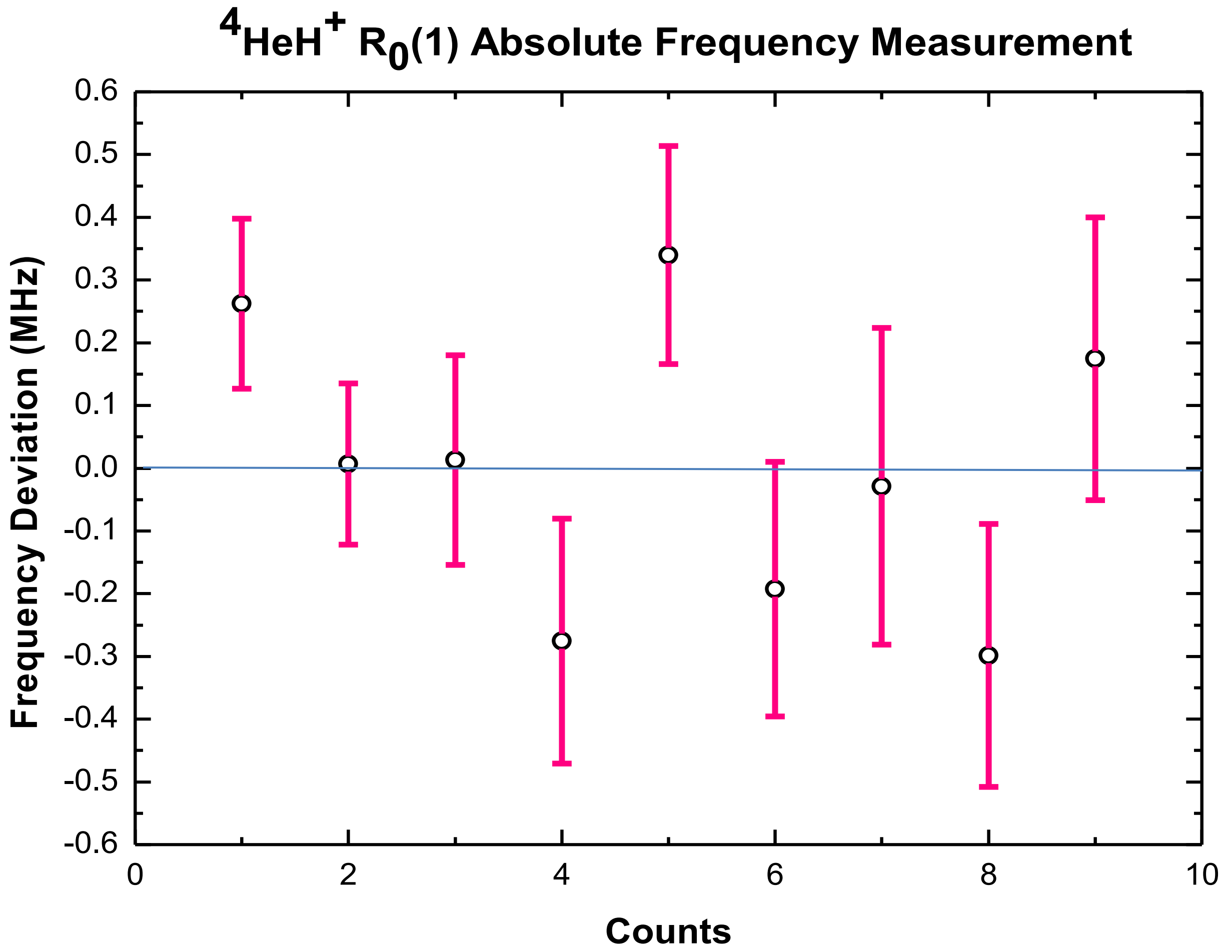
$$\omega_{pump} = \omega_{signal} + \omega_{idler}$$



Frequency measurement



Frequency measurement results



90 788 392.057 MHz

Frequency Measurement: 90788 392.057 $\pm$ 0.076 MHz

Conclusions and Future Works

- First observation of saturation spectrum of HeH^+
- Absolute frequency measurement better than 100 kHz (~1 ppb accuracy)

In future

- Saturation spectroscopy of H_3^+, HeD^+
- Velocity modulation spectrum of HeH^+ and HeD^+ using PPLN DFG (breakdown of BO approximation)

Acknowledgements

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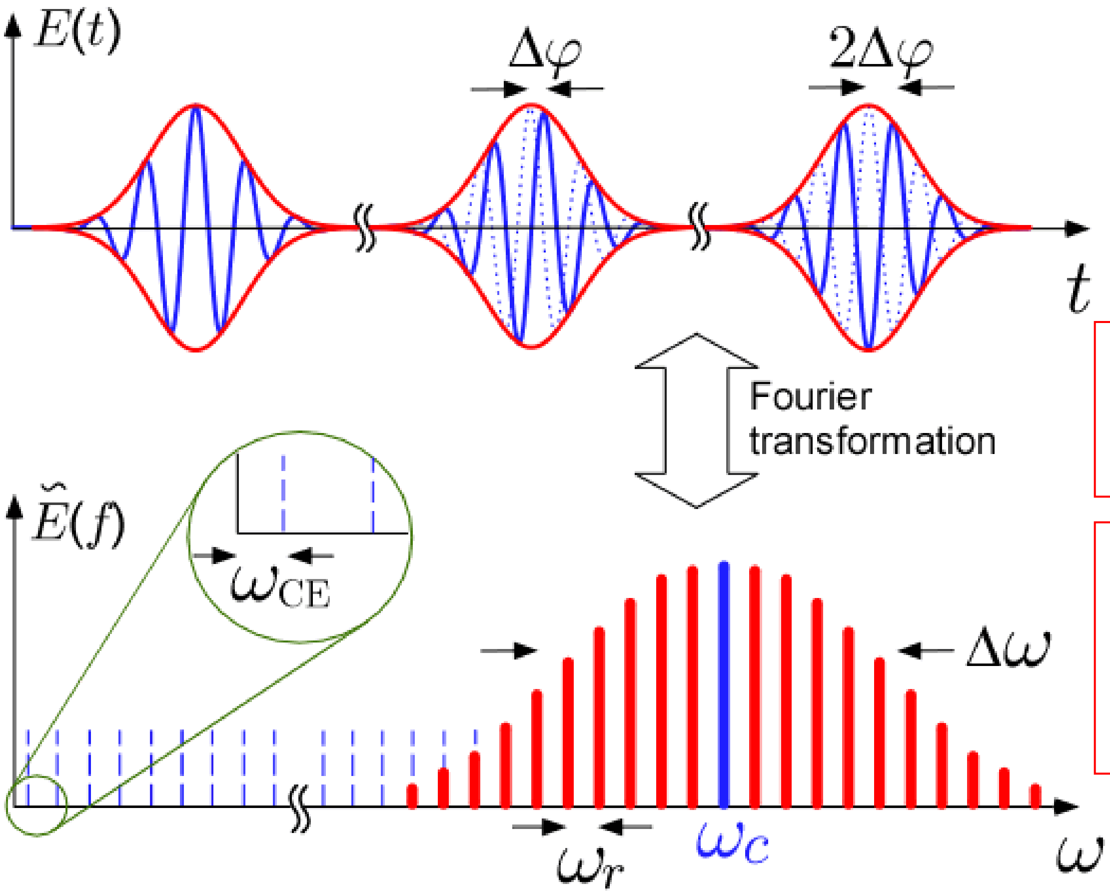
Thanks For your Attention!

Frequency Comb



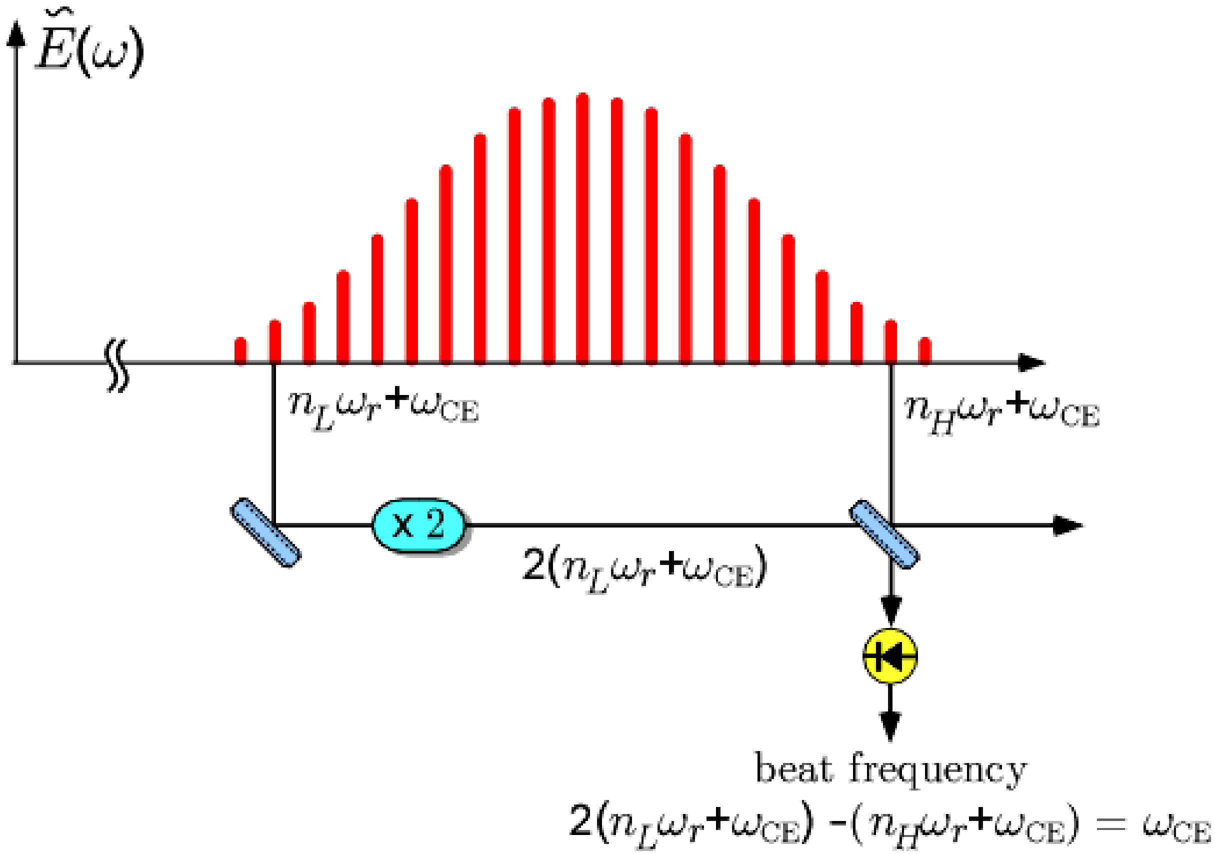
Ti:Saphire Kerr-lens mode locked laser

ω_r=1/T, T: repetition time
Controlled by cavity length

$\Delta\varphi$ pulse-to-pulse carrier envelope phase shift
Controlled by pulsed power

$$\omega_n = n\omega_r + \omega_{CE}$$

f-2f self-reference



$\Delta\nu/\nu = 1X\ 10^{-15}$ 上限 $\Delta\nu/\nu = 1X\ 10^{-18}$