Direct evidence of He-induced excitation process of H atoms in cooled laser plasma

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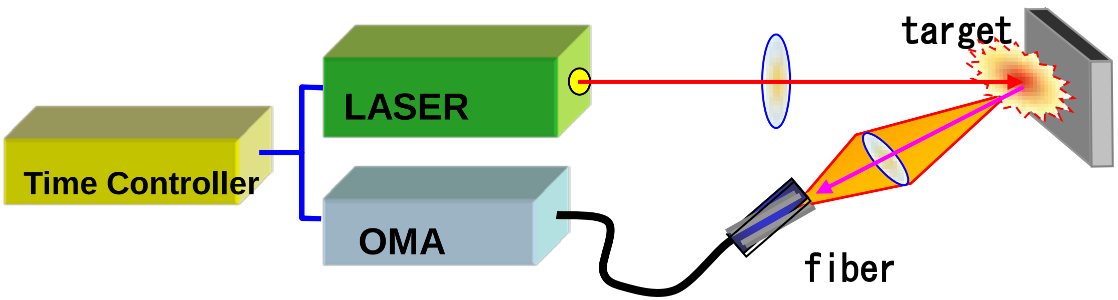
Laser Induce Plasma Spectroscopy

Advantage

- Metal and non metal
- Without pre-treatment
- In-situ analysis
- Micro-area analysis

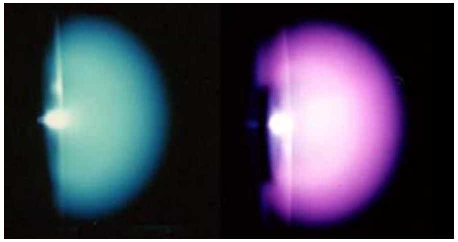
Mechanism



1 atm plasma

(Cremers & Radziemski's Group, 1981)

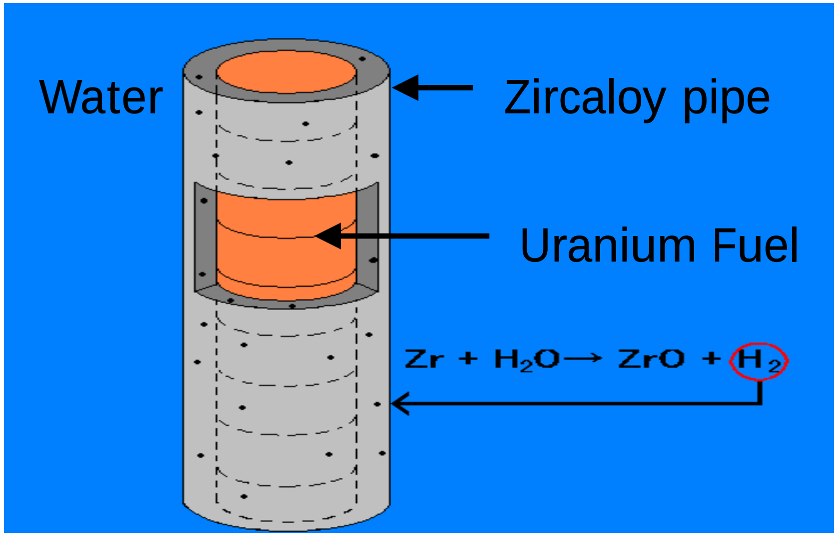
Low pressure plasma



He gas plasma

Kagawa & Kurniawan's Group

Practical Application of LIBS for Hydrogen Analysis In Nuclear Power Station



Zircaloy pipe is an important material used in a light water nuclear power station to contain uranium fuel

Problem:

Hydrogen accumulation in the pipe wall degrades the pipe's strength

Anticipation:

Periodical Inspection

Now in use Inspection Method:

Gas Chromatography

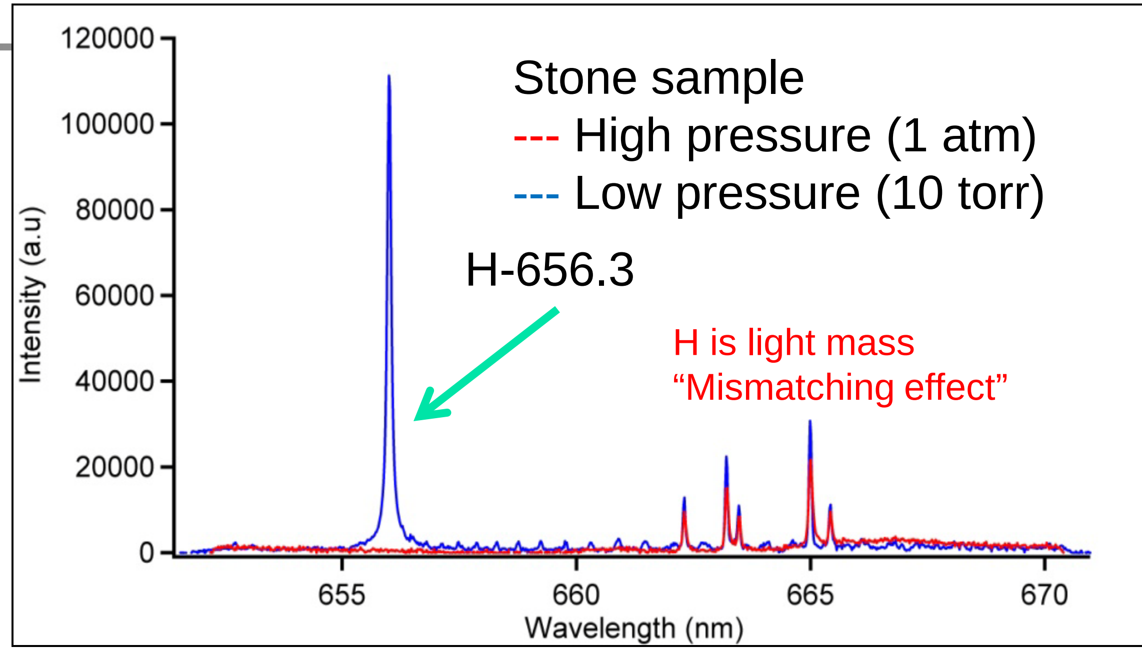
(sample and time consuming, hand touched radioactive and not in situ analysis)

New Method:

Laser Induced Plasma Spectroscopy (LIPS)

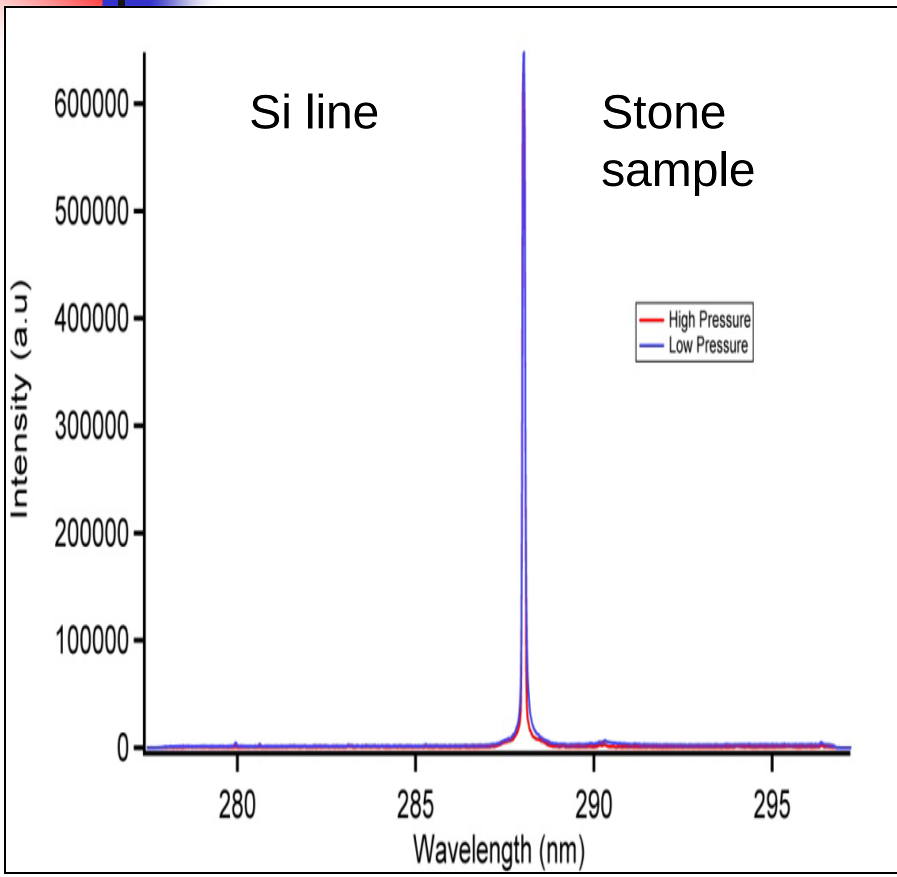
Hydrogen Analysis Using Laser Plasma Method has not been Carried Out Before

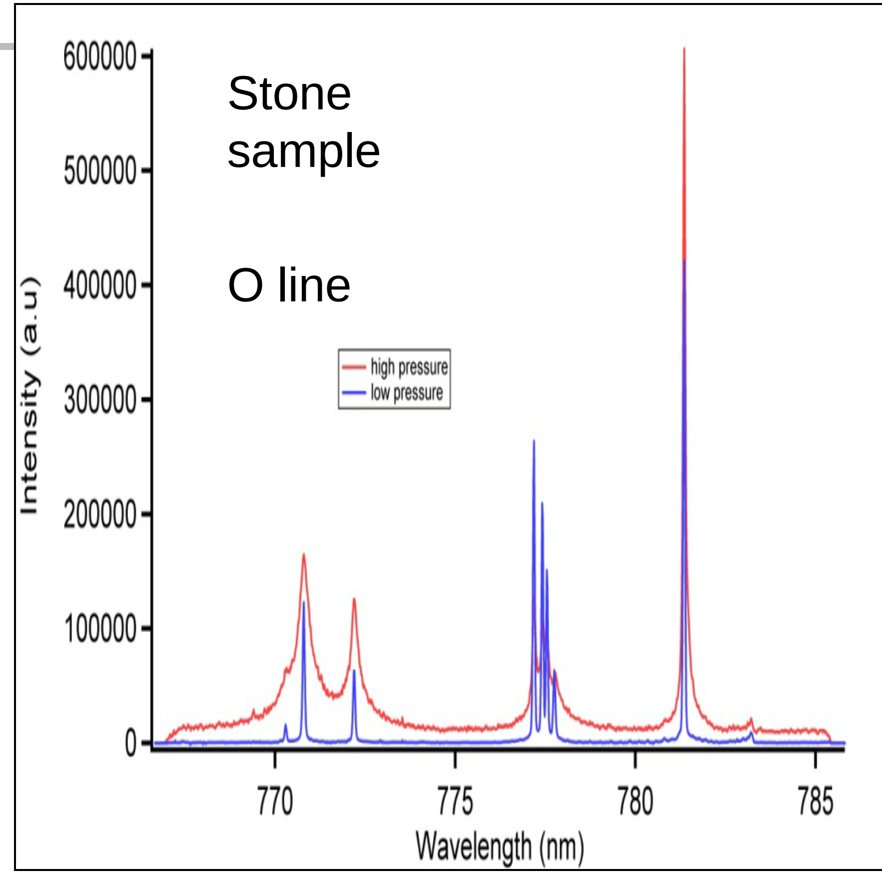
Why



- **Stark broadening effect happens when plasma is produced at 1 atm**
- **Emission efficiency is very low for H atom at 1 atm due to the "mismatching effect"**
- **Disturbance of H_2O**

No Mismatching Effect for Other Elements

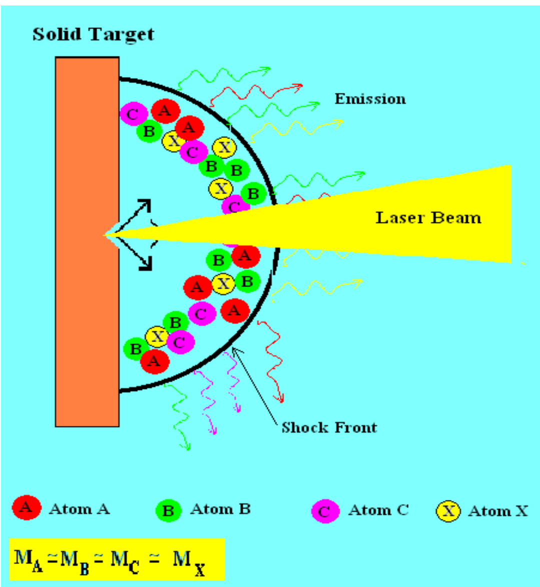




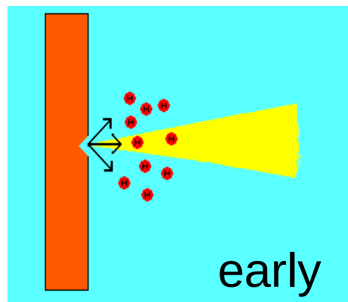
- Red line – High Pressure (1 atm)
- Blue line – Low Pressure (10 torr)

Concept of Mismatching Effect

Ideal Shockwave Plasma Formation



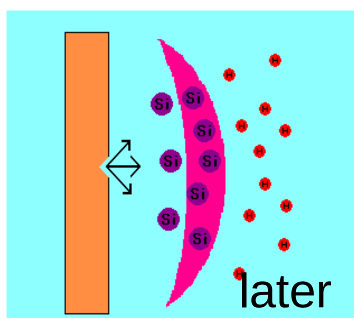
Hydrogen Atom Case



Hydrogen atoms gush out faster

M_H=1

M_{Si}=28



Shock-wave generation

Mismatching Effect

↓

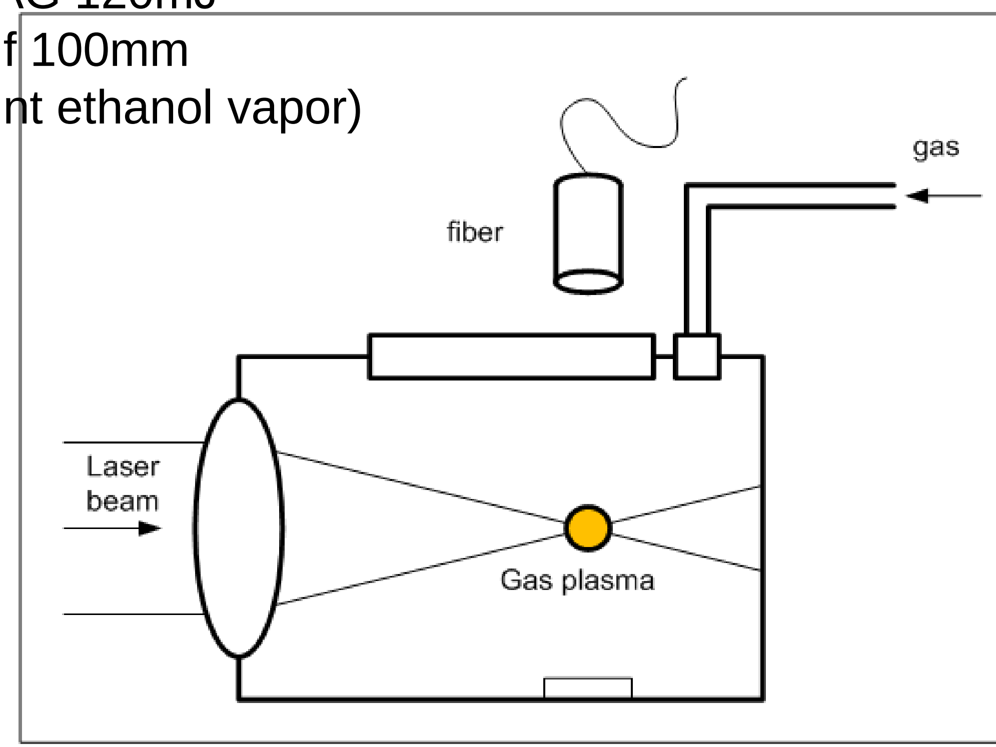
Time difference between gushing out of H atoms and shock-wave generation

Mismatching Effect will significant at higher pressure, or at low power density, because shockwave generation is somewhat delayed

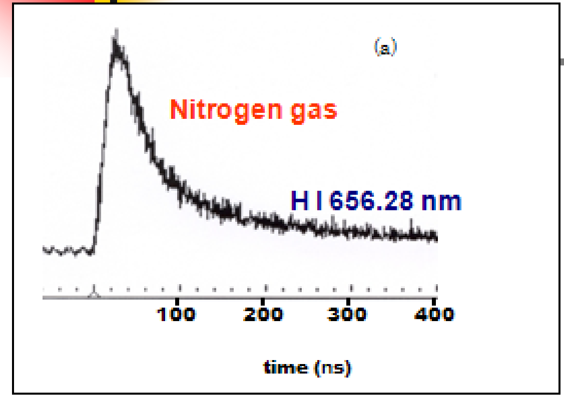
Gas Plasma Induced by Focusing Nd-YAG laser in the Gas

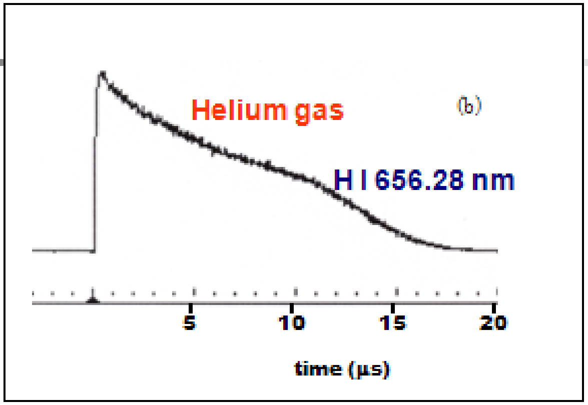
Nd-YAG 120mJ
Lens f 100mm
(in faint ethanol vapor)

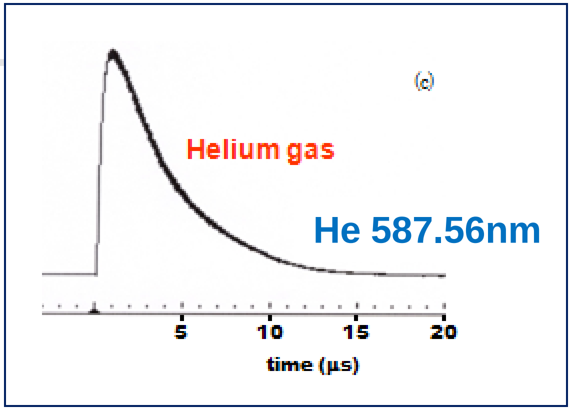
To OMA

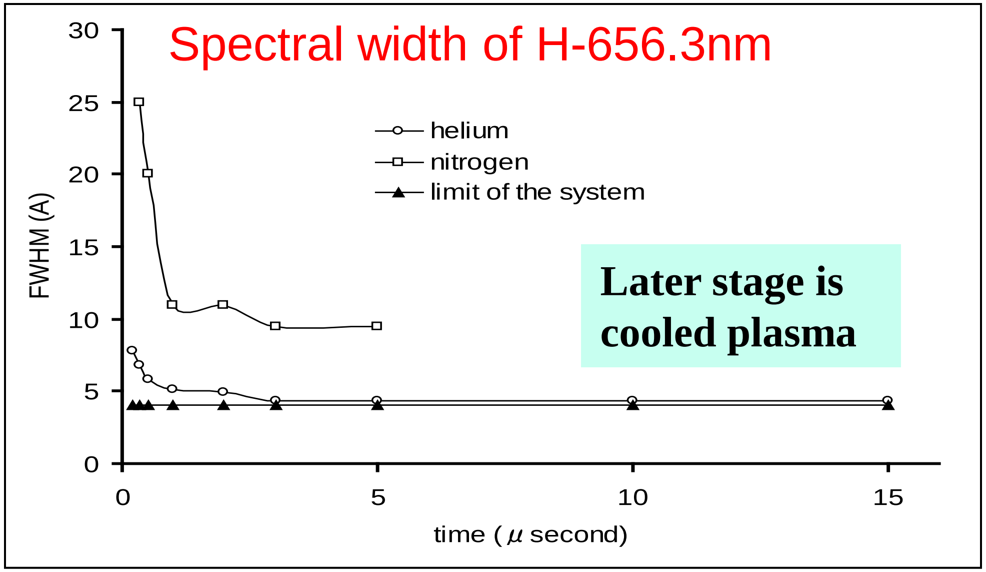


Time Profile of Laser Induced Gas Plasma Emission









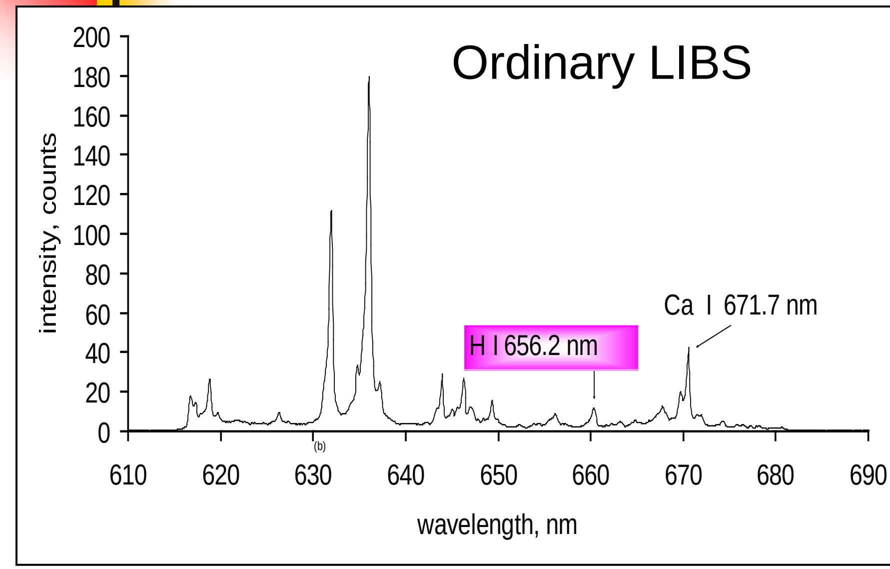
He as host gas:

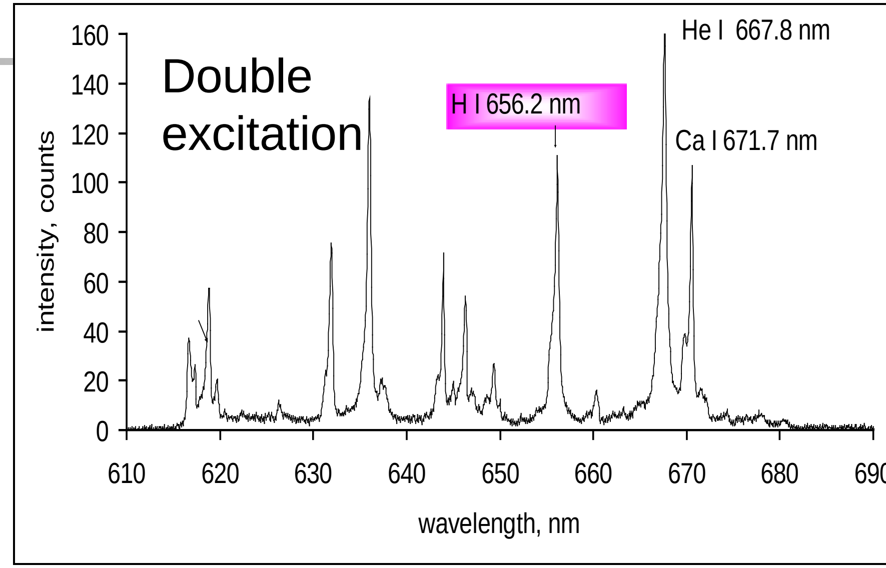
- **Long Life Emission**
- **Very narrow spectral width at later stage**

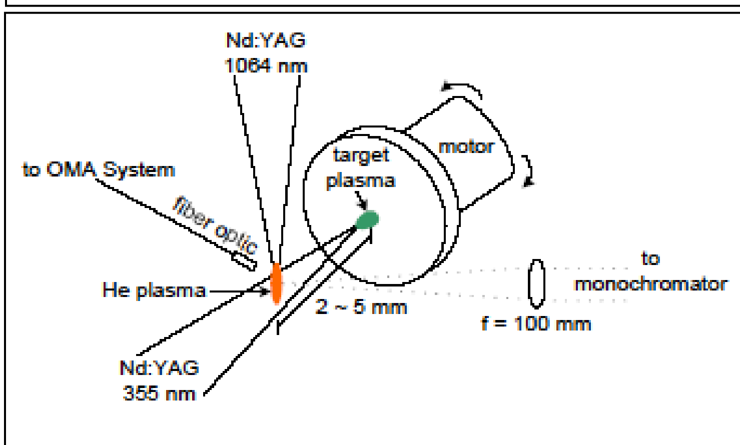
N_2 as host gas:

- **Short Life Emission**
- **Rather broad spectral width**

Double Excitation Scheme for H analysis under He 1 Atm



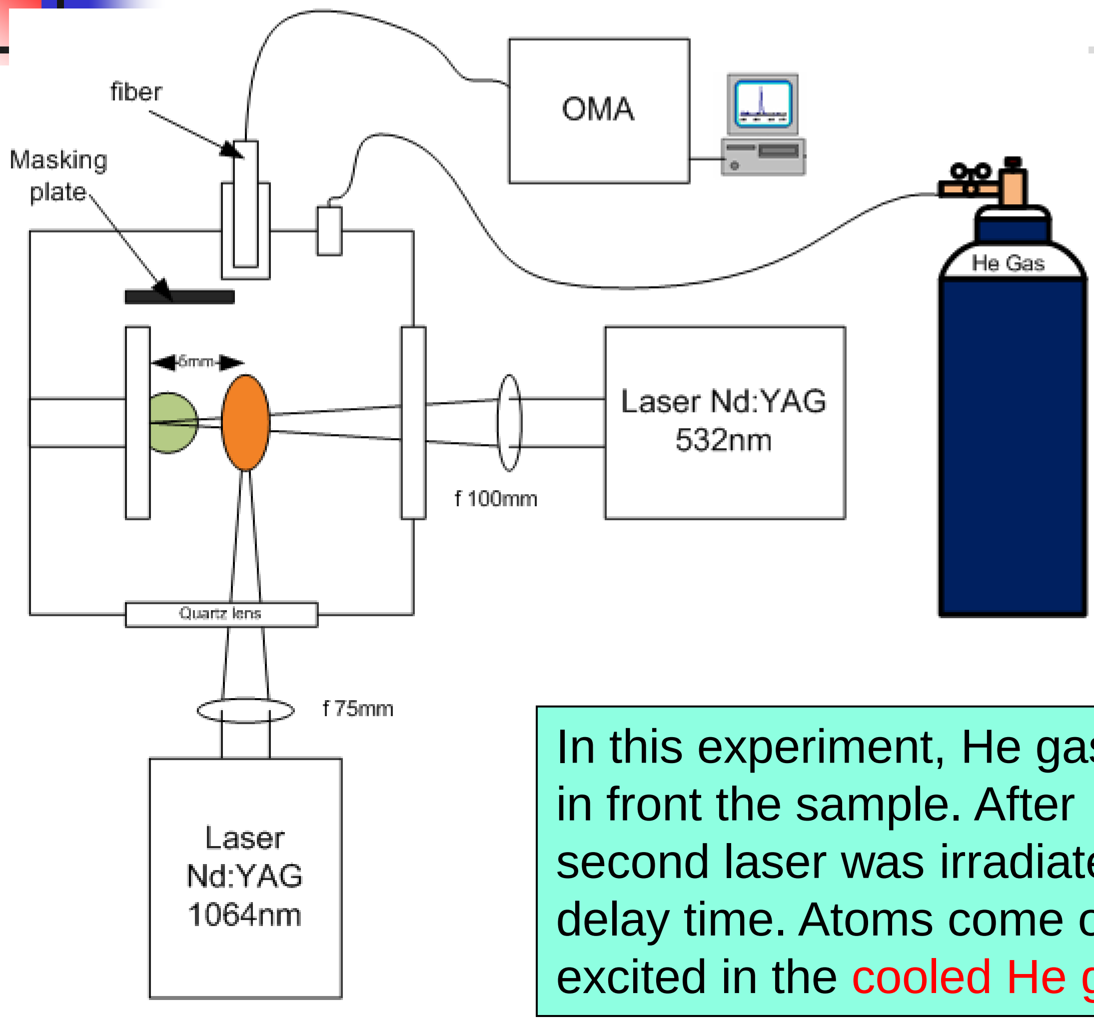




Sample: Slide glass

Hydrogen Emission can be detected with high efficiency and very narrow spectral width

Experiment Setup for Proving Excitation in Cooled Plasma



- Stone Sample

For gas Plasma:
Nd-YAG (1064nm), 163mJ

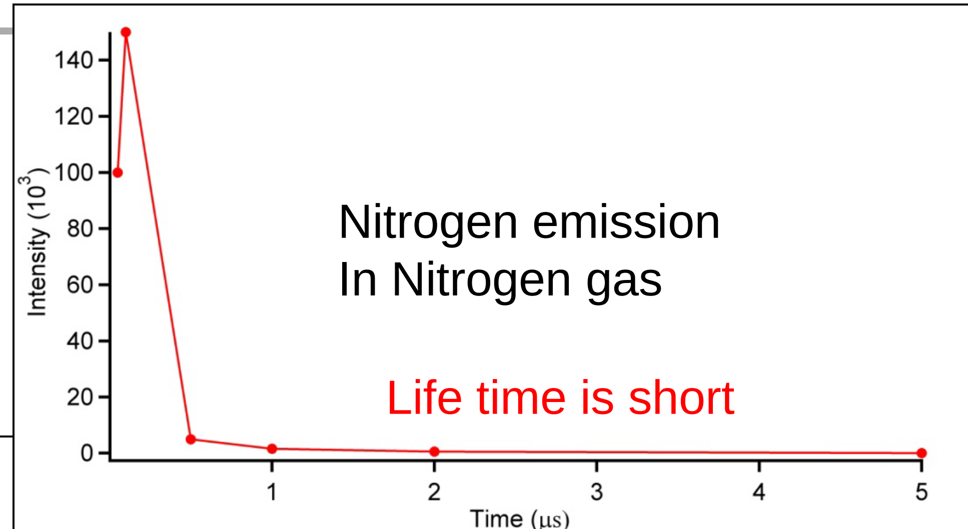
For ablate sample:
Nd-YAG (532nm), 74mJ

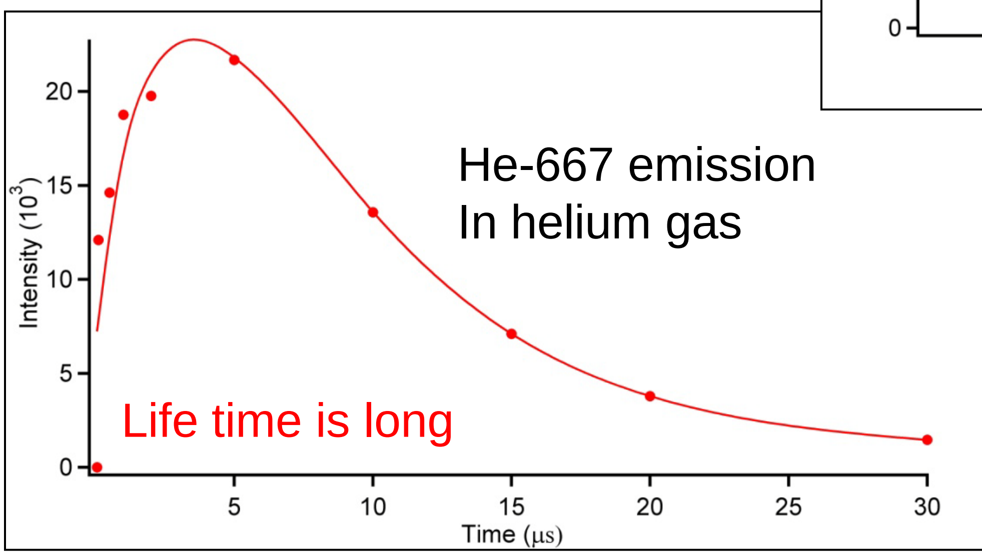
Delay Time between two laser: 10us

In this experiment, He gas plasma was made at 5mm in front the sample. After the gas plasma formation, second laser was irradiated for ablation with 10us delay time. Atoms come out from the sample and excited in the cooled He gas plasma.

Time Profile of Emission of the Gas Plasma

Due to the thermal excitation →





← Due to the He meta-stable excitation

He emission proceeds in cooled plasma

Two possible mechanism

- He meta - He meta collision

$$He^{*} + He^{*} \rightarrow He + (He^{+} + e^{-})$$

$$He^{+} + e^{-} \rightarrow He^{**} \rightarrow He + h\nu$$

- Thermal excitation from He meta

$$He^{*} + thermal \rightarrow He^{**} \rightarrow He + h\nu$$

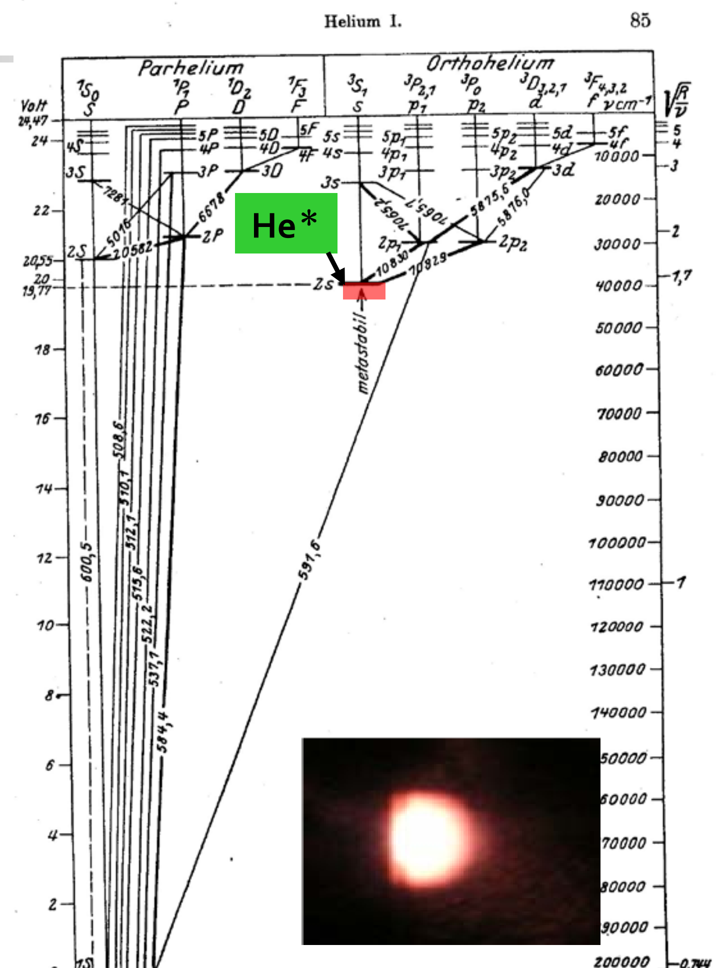
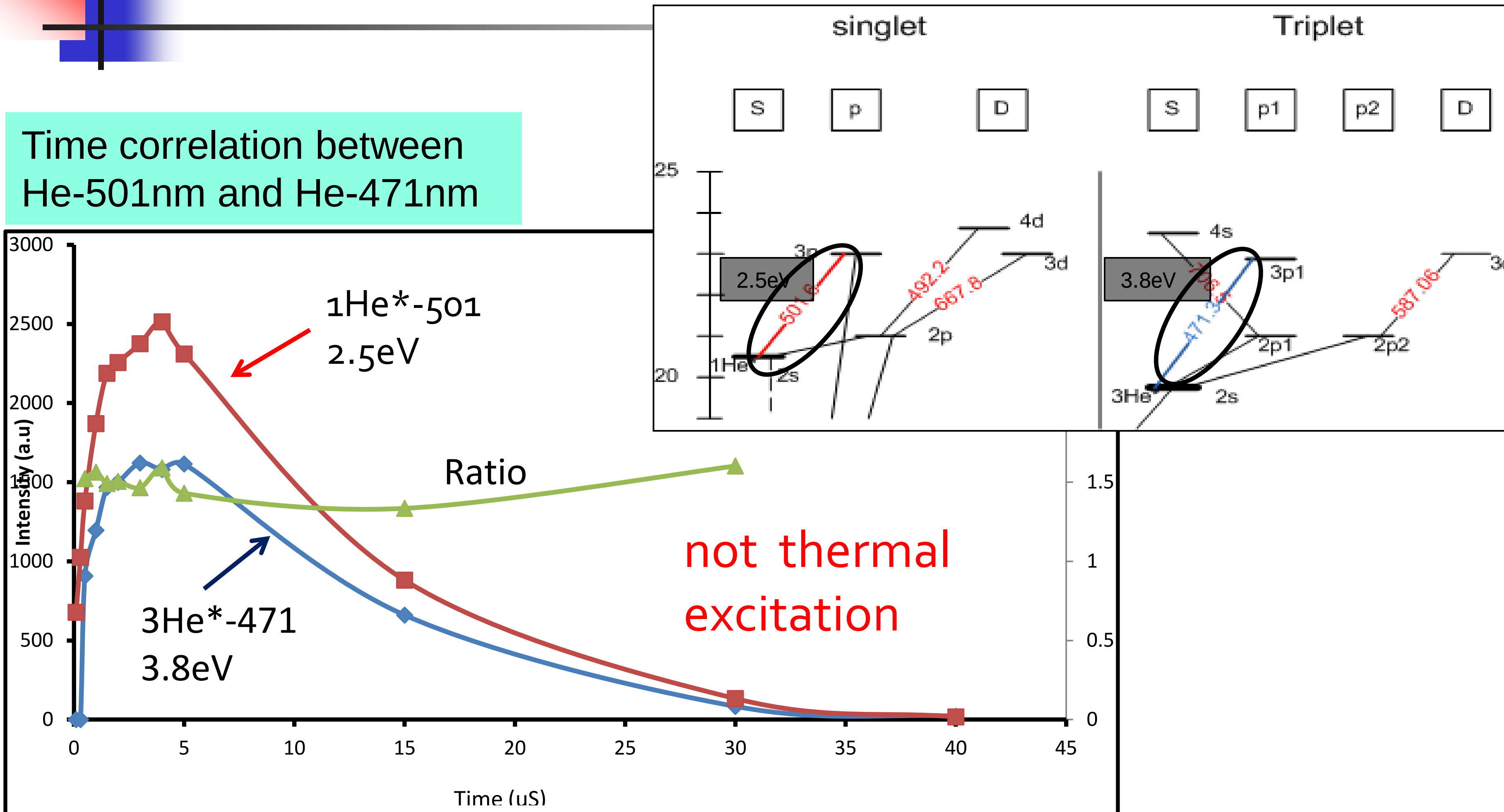


Fig. 77, II, Text S. 116. Niveauschema des Helium I.

The evidence to prove He emission is not due to thermal excitation

Time correlation between He-501nm and He-471nm



not thermal excitation

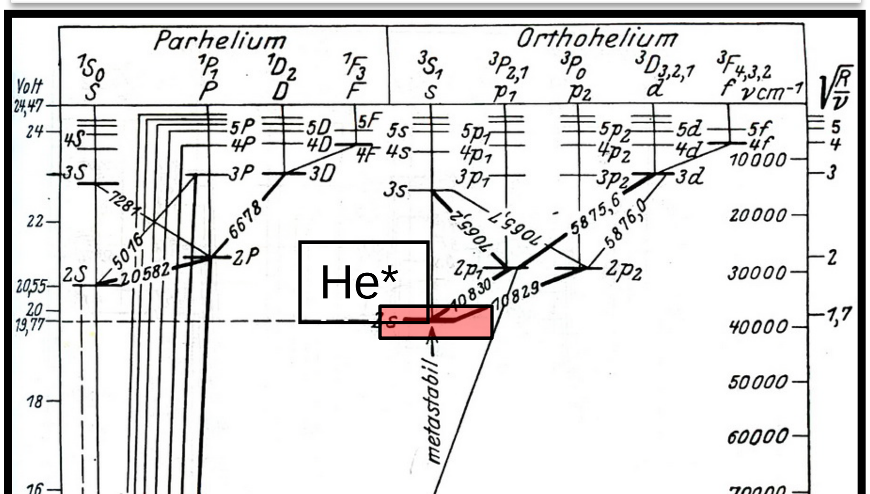
The Excitation Mechanism through Helium Meta-stable

Penning effect

$$He^{*} + X \rightarrow He + (X^{+} + e^{-})$$

$$X^{+} + e^{-} \rightarrow X^{**} \rightarrow X + h\nu$$

recombination



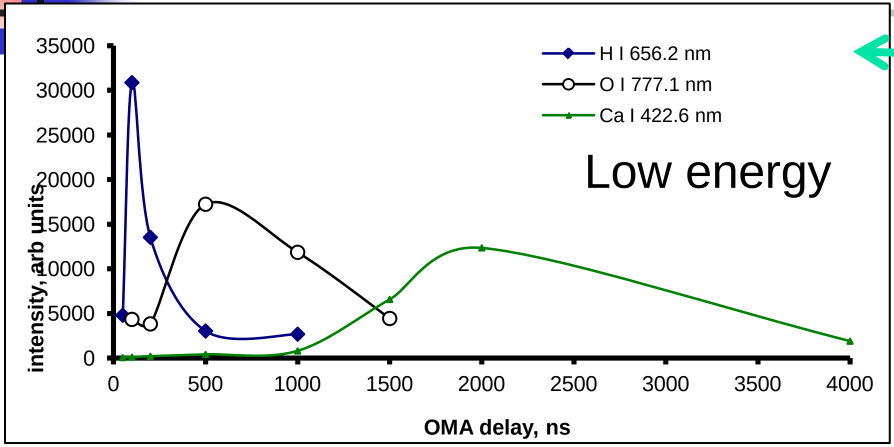
X atom collides with He metastable

X atom is ionized and Releases free electron

Electron recombines with X+ ion

Spectral emission of X atom

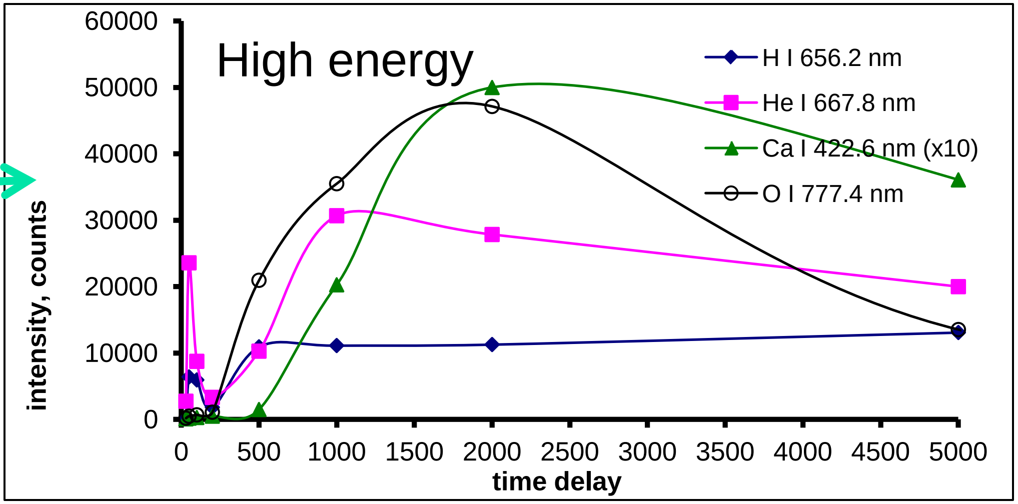
Time Profile of H, O, Ca emission in the cooled He gas plasma



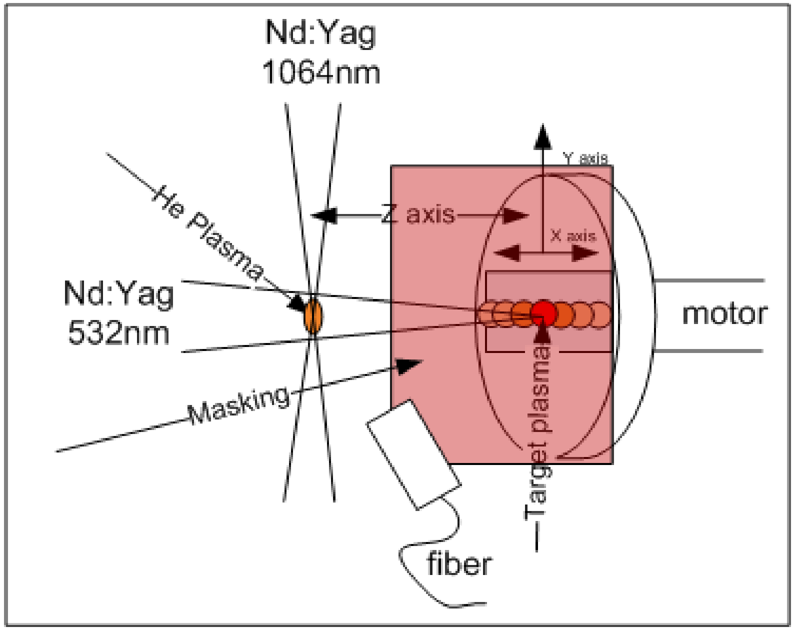
Due to Helium meta-stable excitation

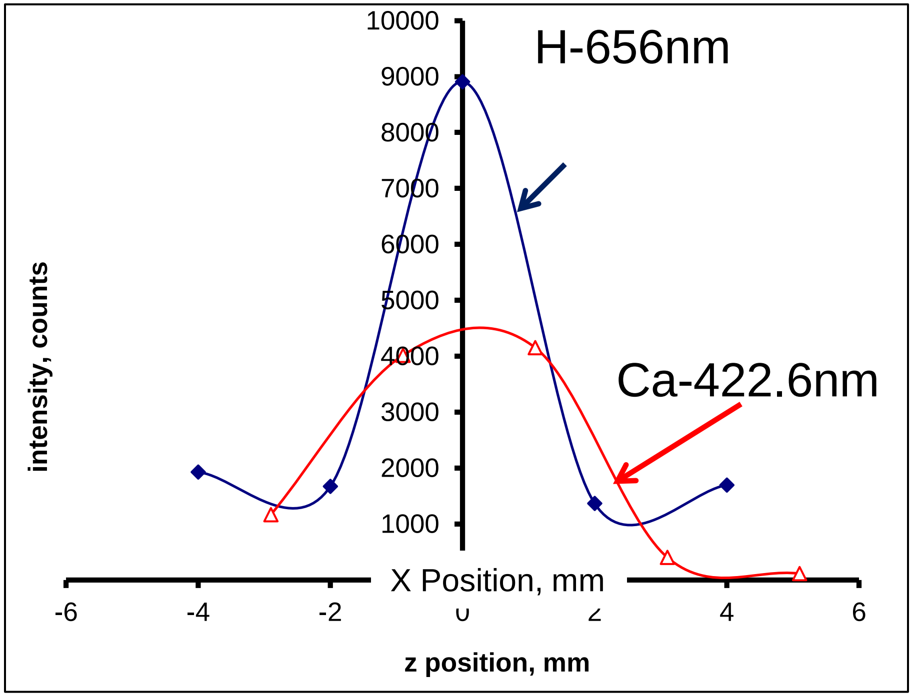
H atoms come out faster than other atoms, which proves the "mismatching effect"

Another mechanism (shock wave excitation) works for high energy pulse case



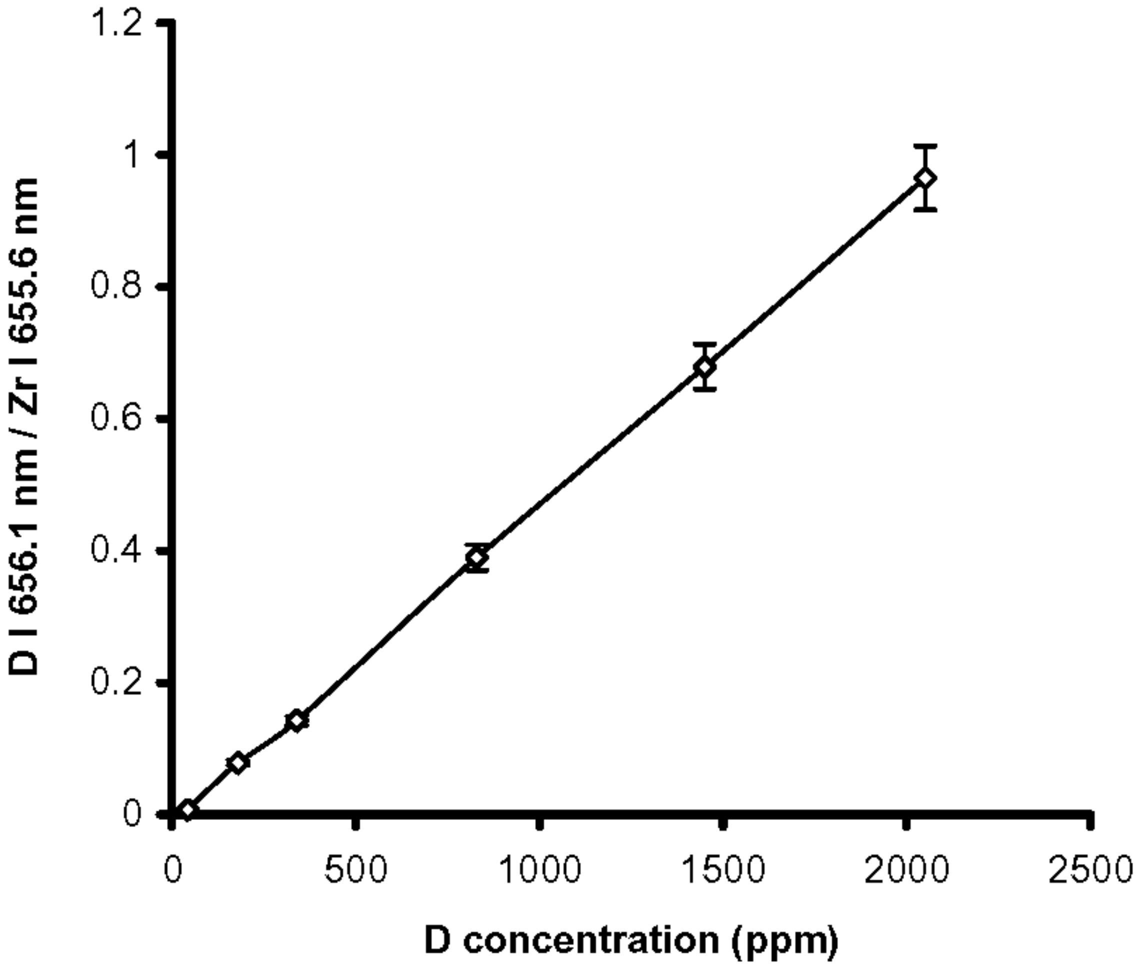
Direction of Gushing Atoms





Distribution of H atoms is more straight forwards compared to other atoms

Intensity Calibration Curve of the Deuterium Impurity in Zircaloy-4 Samples



Conclusion

- He emission takes place through the collision of two Helium meta-stable atoms (He*).

$$He^* + He^* \rightarrow He + (He^+ + e^-) \rightarrow He^{**} \rightarrow He^* + h\nu$$

- In a cooled He plasma, Helium meta-stable atoms (He*) excites atoms including Hydrogen

$$He^* + X \rightarrow He + (X^+ + e^-) \rightarrow He^{**} \rightarrow X + h\nu$$

- Hydrogen atoms gushed out faster than other atoms, which proved our hypothesis, namely "mismatching effect"

Published work

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- Z.S. Lie, M. Pardede, R. Hedwig, M.M. Suliyanti, E. Steven, Maliki, Koo H. Kurniawan, M. Ramli, S.N. Abdulmadjid, N. Idris, K. Lahna, K. Kagawa and M.O. Tjia, Intensity Distributions of Enhanced H Emission from Laser-Induced Low-Pressure He Plasma and a Suggested He-Assisted Excitation Mechanism, J. Appl. Phys., **106**, 3 (2009) pp. 043303 1-6
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- K.H. Kurniawan, M. Pardede, R. Hedwig, Z.S. Lie, T.J. Lie, D.P. Kurniawan, M. Ramli, K. Fukumoto, H. Niki, S.N. Abdulmadjid, N. Idris, T. Maruyama, K. Kagawa, M.O. Tjia, Quantitative Hydrogen Analysis of Zircaloy-4 Using Low-Pressure Laser Plasma Technique, Anal. Chem. **79**,7 (2007) pp. 2703-2707
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