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

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Laser Induce Plasma Spectroscopy

Mechanism

Time Controller

LASER

OMA

fiber
target

Advantage

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

1 atm plasma

Low pressure plasma

He gas plasma

(Cremers & Radziemski’s Group, 1981)

Kagawa & Kurniawan’s Group

(Advantage)

(Cremers & Radziemski’s Group, 1981)
Practical Application of LIBS for Hydrogen Analysis In Nuclear Power Station

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)

Zircaloy pipe is an important material used in a light water nuclear power station to contain uranium fuel
Hydrogen Analysis Using Laser Plasma Method has not been Carried Out Before

- 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 H₂O

Why

**Graph**
- Stone sample
  - High pressure (1 atm)
  - Low pressure (10 torr)

**Spectral Data**
- H-656.3

H is light mass
"Mismatching effect"
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

- \( M_H = 1 \)
- \( M_{Si} = 28 \)

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

- 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

N2 Gas Plasma

He Gas Plasma
Time Profile of Laser Induced Gas Plasma Emission

He as host gas:
- Long Life Emission
- Very narrow spectral width at later stage

N₂ as host gas:
- Short Life Emission
- Rather broad spectral width
Double Excitation Scheme for H analysis under He 1 Atm

Hydrogen Emission can be detected with high efficiency and very narrow spectral width
Experiment Setup for Proving Excitation in Cooled Plasma

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

- **He-667 emission**
  - In helium gas
  - Life time is long

Due to the He meta-stable excitation

- **Nitrogen emission**
  - In Nitrogen gas
  - Life time is short
He emission proceeds in cooled plasma

- He meta - He meta collision
  
  \[ He^* + He^* \rightarrow He + (He^+ + e^-) \]
  
  \[ He^+ + e^- \rightarrow He^{**} \rightarrow He + hv \]

- Thermal excitation from He meta
  
  \[ He^* + \text{thermal} \rightarrow He^{**} \rightarrow He + hv \]
The evidence to prove He emission is not due to thermal excitation

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

1He*-501 2.5eV
3He*-471 3.8eV

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
He emission takes place through the collision of two Helium meta-stable atoms ($\text{He}^*$).

\[
\text{He}^* + \text{He}^* \rightarrow \text{He} + (\text{He}^+ + e^-) \rightarrow \text{He}^{**} \rightarrow \text{He}^* + \text{hv}
\]

In a cooled He plasma, Helium meta-stable atoms ($\text{He}^*$) excites atoms including Hydrogen

\[
\text{He}^* + X \rightarrow \text{He} + (X^+ + e^-) \rightarrow \text{He}^{**} \rightarrow X + \text{hv}
\]

Hydrogen atoms gushed out faster than other atoms, which proved our hypothesis, namely “mismatching effect”
Published work


