Investigation of fundamental mechanisms related to ambient gas heating and hydrodynamics of laser-induced plasmas

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Laser-induced plasmas (LIP)

• Remote sensing applications [1-2]:
  – Nuclear safeguards
  – Space exploration
  – Biological/geological forensics

• Intense pulsed laser focused onto a target generates plasma
  – Plasma consists of excited atoms, ions, molecules, nano- and micro-particles
  – Plasma cools emitting electromagnetic radiation
    • Emission useful as diagnostic tool through spectroscopy

Optical sensing techniques

1. Laser-ablation (LA) coupled with optical emission spectroscopy (OES)
2. LA molecular isotopic spectrometry (LAMIS)
3. LA coupled with laser absorption spectroscopy (LAS)
4. Various imaging techniques (i.e. spectral mapping, shadowgraphy)
# LIP for nuclear material sensing

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![Graph of U spectra with intensity vs. wavelength](image)

Skrodzki et al. (2016)
LIP for nuclear material sensing

Vast parametric space for optimization

↑Precision; Isotope distinction

Hartig et al. (2013)
Skrodzki et al. (2016)
Cremers et al. (2012)
Doucet et al. (2011)
Recent LIP applications in U sensing

- Dual-pulse (DP) OES enhances standard single-pulse (SP) signal
- Initial pulse ablates target
- Secondary pulse reheats plasma → more emission
- Also increases background and noise

Recent LIP applications in U sensing

• Recent comparison of U emission spectra from two solid targets [4]:
  – Kopp glass containing 1.3% natural U by mass
  – Depleted U metal
• U oxide bands prevalent among several U I features in metal
• Matrix effects mitigate U signal in glass

Laser-induced sparks & impetus

- Aforementioned studies include primarily solid targets
- Gaseous targets generate *sparks* which have various applications in ignition, machining, further nuclear material sensing
- Uranyl fluoride (UO$_2$F$_2$) is relevant to enrichment process and may be an indicator of enrichment facilities

Kemp (2006)
Impetus

1. Employ optical sensing and imaging techniques to understand spark morphology
2. Identify physical phenomena associated with expansion and collapse of sparks
3. Optimize spectroscopic viewing windows (spatial and temporal) in sparks for latter applications in UO$_2$F$_2$ sensing
Expansion & collapse of sparks

Previous literature shows heating (excitation and ionization) of the gas surrounding the spark [5]:

1. Prompt electrons:
   - Originate from interaction between laser pulse and target
   - ~10^1-10^2 ns

2. Radiative heating:
   - Intense ultraviolet (UV) radiation from plasma
   - Early emission (~10^1-10^3 ns) relative to plasma lifetime (~ms)
   - Instantaneous interaction with surrounding gas

3. Detonative heating
   - Pressure/density gradient from LPP generates shock
   - Shock expands detaching from plasma (~10^1-10^2 µs)

Shock Radius

- Horizontal Axis
- Vertical Axis

Time after breakdown [ns]

0 mm
1.25 mm
2.5 mm

Spectral Image of Spark

0.5 mm

Air (1.0 atm)

Time after ablation: 10 ns
Air

10 ns

100 ns

250 ns

500 ns

1.0 mm

1000 ns

2000 ns

3000 ns

4000 ns

5000 ns

6000 ns

7000 ns

8000 ns
Appearance of O I emission features at ~300-400 ns at 2.5-mm distance from kernel

- Shock only reaches 2.5-mm distance after 4500 ns
- Profound late-time features following arrival of plasma
Modeling Details

Model

• Open source Computational Fluid Dynamics (CFD) software package OpenFOAM
• 2D numerical simulation of laser-induced electrical breakdown of air
• Computational domain: 5-mm (x-axis) x 10-mm (y-axis) discretized into 250 x 500 cell mesh, respectively
• Left-side y-boundary considered a symmetry axis while outflow boundaries are placed sufficiently far from region of interest in flow field

Parameter Space

• Ambient: Ar; pressure 101,325 kPa; temperature 300 K
• Initial plasma specified as ellipse with 50-µm (x-axis) x 150-µm (y-axis) major axis lengths
• Initial plasma: air; pressure 25 MPa; temperature 70,000 K
• Equation of state: Ideal Gas Law
• Duration: 10 µs following onset of laser pulse
Pressure Contour
5 µs

Modeling Scale:
0.5 mm
Shadowgraphy Scale:
0.5 mm
Conclusion & future work

Experiment

• Different gases exhibit unique shock morphologies related to laser absorption parameters
• Observed time-dependent emission features unique to each gas
  – Broad mixing at early times, ionic emission, neutral emission, then molecular emission
• Radiative heating proves dominant mechanism; detonative heating negligible

Model

• The shockwave pressure is ~20 times greater than the atmospheric pressure at 100 ns and then rapidly decreases as the spark decays
• The shock front becomes increasingly symmetric in the shape with time
• The temperature of the plasma has severely decreased from 70,000 K to ~20,000 K during the first 100 ns

Future Work

• Expanding optical techniques to sparks containing UO$_2$F$_2$
Experiment Details

Breakdown

• Generate gaseous spark in four gases (air, argon, nitrogen, helium) at atmospheric pressure (~760 Torr)
• 55 mJ energy, 1064 nm Nd:YAG (8 ns FWHM) focused to ~100 µm spot diameter – 90 GW cm⁻²

Time-resolved Shadowgraphy

• Pressure/density difference along shock-front has different refractive index
• Observe shock by shining backlight laser through spark onto CCD camera
• ~5 mJ, 532 nm Nd:YAG (4 ns FWHM) expanded to ~1 cm spot diameter as backlight

Time-resolved Spectroscopy

• Observe emission at three horizontal positions with respect to plasma core (kernel): kernel (0 mm), 1.25 mm, and 2.5 mm
Argon

10 ns  100 ns  250 ns  500 ns

1.0 mm

1000 ns  2000 ns  3000 ns  4000 ns

5000 ns  6000 ns  7000 ns  8000 ns
Argon

10 ns

100 ns

250 ns

500 ns

1000 ns

2000 ns

3000 ns

4000 ns

5000 ns

6000 ns

7000 ns

8000 ns

0.5 mm
Time after ablation: 10 ns

Shock Radius

- Horizontal Axis
  - $R_{\text{hor}} = 0.12 \text{ mm}$
  - $R_{\text{ver}} = 0.48 \text{ mm}$
- Vertical Axis

Spectral Image of Spark

- 0 mm
- 1.25 mm
- 2.5 mm

Nitrogen (1.0 atm)
Nitrogen
Nitrogen

N$_2$

10 ns  100 ns  250 ns  500 ns

0.5 mm

1000 ns  2000 ns  3000 ns  4000 ns

5000 ns  6000 ns  7000 ns  8000 ns
Helium
Helium

He

10 ns

600 ns

1400 ns

100 ns

800 ns

1600 ns

200 ns

1000 ns

1800 ns

400 ns

1200 ns

2000 ns

0.5 mm