

Abstract: Studies of interaction of high-power ultrashort laser pulses with matter are not only of fundamental scientific interest, but are also highly relevant to applications in the domain of remote sensing. Here, we discuss the extension of our recent work on optimizing the coupling between femtosecond laser filaments and solid targets to explore the effect of laser wavelength on the emission signal intensity and background. We use a pulsed Ti:sapphire laser at 0.8 μm and techniques of second-harmonic generation and optical parametric amplification to generate energetic 0.4 μm and 2 μm pulses, respectively. This allows for investigation of the wavelength-dependent filament ablation mechanisms, as well as filament-induced plasma dynamics and its thermodynamic parameters. The results of this study may offer a path to maximize the signal to background in filament-based laser-induced breakdown spectroscopy.

OPTICAL (REMOTE) SENSING FOR NONPROLIFERATION, SAFEGUARDS, AND VERIFICATION

Conventional sample preparation

In-situ LIBS

Laboratory LIBS

FILAMENTATION LASER-INDUCED BREAKDOWN SPECTROSCOPY (F-LIBS)

Performing LIBS at large standoff distances leads to two main challenges associated with beam delivery:

- Prohibitively large optics
- Absorption and atmospheric turbulence

Spot size (diffraction limited): $w_0 \geq 1.22 \lambda f/D$

Self-focusing: $n = n_0 + n_2 I$

Ionization defocusing: $\Delta n_{\text{plasma}} = -\frac{e^2 N_e(t)}{2m_e \epsilon_0 \omega_0^2}$

Balance of lensing/defocusing results to intensity clamped propagation: $n_2 I = \Delta n_{\text{Kerr}} = \Delta n_{\text{plasma}} \cong \frac{n_e}{2 n_{\text{crit}}}$

GENERATION OF ADDITIONAL EXCITATION WAVELENGTHS

Generating 0.4 μm and 2.0 μm laser radiation

Fundamental harmonic of the laser is set at $\sim 800 \text{ nm}$;

The second harmonic (0.4 μm) is created within BBO crystal ($\beta\text{-BaB}_2\text{O}_4$);

2.0 μm radiation is produced via Optical Parametric Amplification (OPA).

Pulse duration vs BBO thickness

Exp. setup: M – mirror, SM – spherical mirror, DM – dichroic mirror, L – lens, BBO – beta barium borate, OPA – Optical Parametric Amplification

PRODUCTION OF ULTRAFAST MID-IR RADIATION

Optical Parametric Amplification

$$P(t) = \epsilon_0 (\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots)$$

Output energy: 2.2-mJ, (~ 6 -optical-cycles);

Output pulse duration $\tau_{\text{out}} \sim 42 \text{ fs}$;

Experimental setup of the two-stage OPA: Blue, 800nm pump beam; green, white light continuum (WLC); red, 2.0 μm signal beam; and orange, 1.3 μm idler beam. BS, beamsplitter; WP, $\lambda/2$ waveplate; DM, dichroic mirror; S1, sapphire plate; S2, ZnSe plate; LPF, long-pass filter; C1, type I BBO crystal; and C2, type II BBO crystal.

PLASMA DYNAMICS AND MORPHOLOGY AT FUNDAMENTAL LASER WAVELENGTH

In order to have reliable analysis and diagnostics, spatio-temporal intensity mapping of emitted species was performed.

Normalized emission intensity vs Pump laser energy E_p (mJ)

3D surface plots of emission intensity vs Axial position (mm) and Delay (ns) for 1.9 mJ and 3.8 mJ.

CONCLUSIONS, CHALLENGES AND FUTURE WORK

UO₂F₂ Fluorescence Spectrum

UO Molecular Emission Spectrum

- Filament-produced plasma spectroscopy can be successfully utilized to discriminate emission from natural and weapons-grade uranium and develop practical methods for their detection;
- Detailed fundamental studies performed on solid targets serve as a basis to investigating relevant thermodynamic parameters and plasma emission properties of nuclear materials;
- Different laser wavelengths will be used to optimize signal-to-background ratio of signature emission features highly relevant to remote sensing applications;
- The former approach will be applied for studying the character of UO_xF_x fluorescence, and molecular emission of UO.

References

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