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Supplementary Material

Spatiotemporal Characterization of Cerium Monoxide in Laser Ablation Plasmas using Spectrally-Resolved Fast-Gated Imaging

Emily H. Kwapis and Kyle C. Hartig

Nuclear Engineering Program, Department of Materials Science and Engineering, University of Florida, Gainesville, FL 32611

Corresponding Author E-mail: ekwapis@ufl.edu

Images of the plasma emission in false color are provided for all atmospheric conditions tested in this work for time delays between 1 ns and 75 μs.

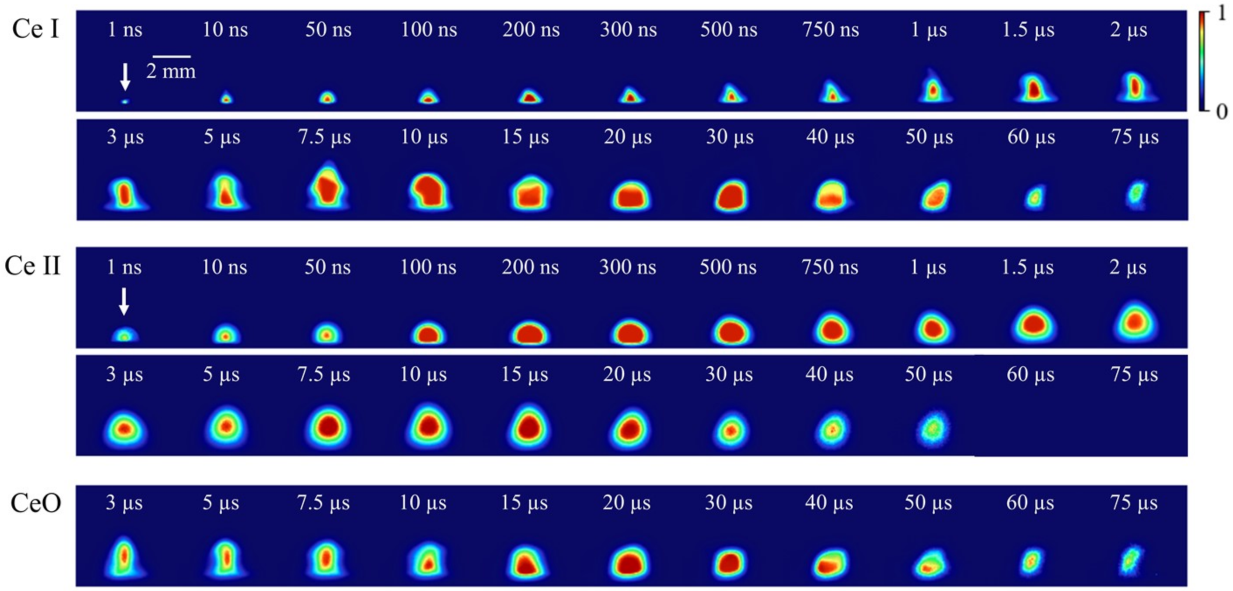

Figure S1. Time-resolved fast-gated images of Ce LPP plasma emission in air at atmospheric pressure. Each individual frame is normalized to its minimum and maximum pixel intensities.

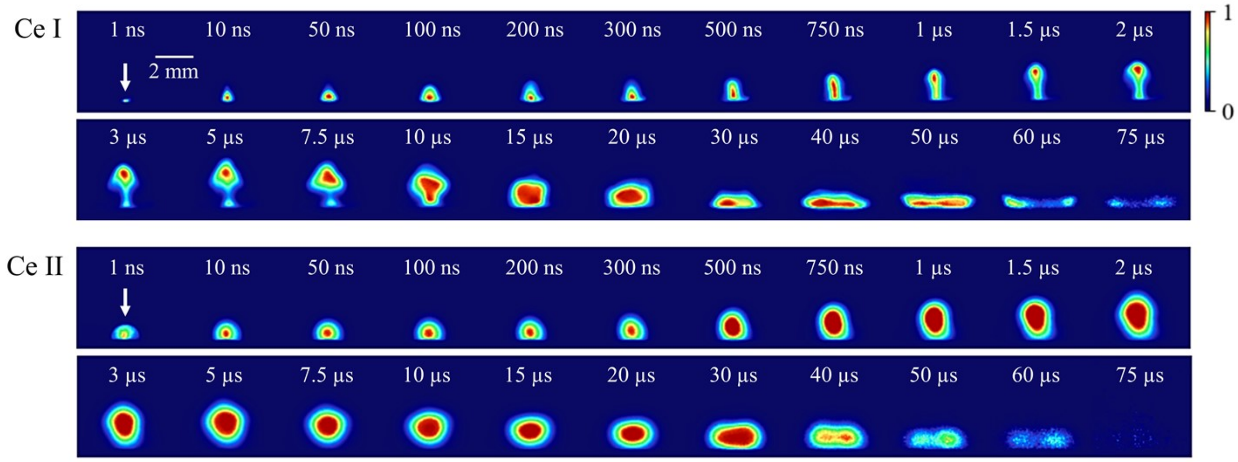

Figure S2. Time-resolved fast-gated images of Ce LPP plasma emission in argon at atmospheric pressure. Each individual frame is normalized to its minimum and maximum pixel intensities.

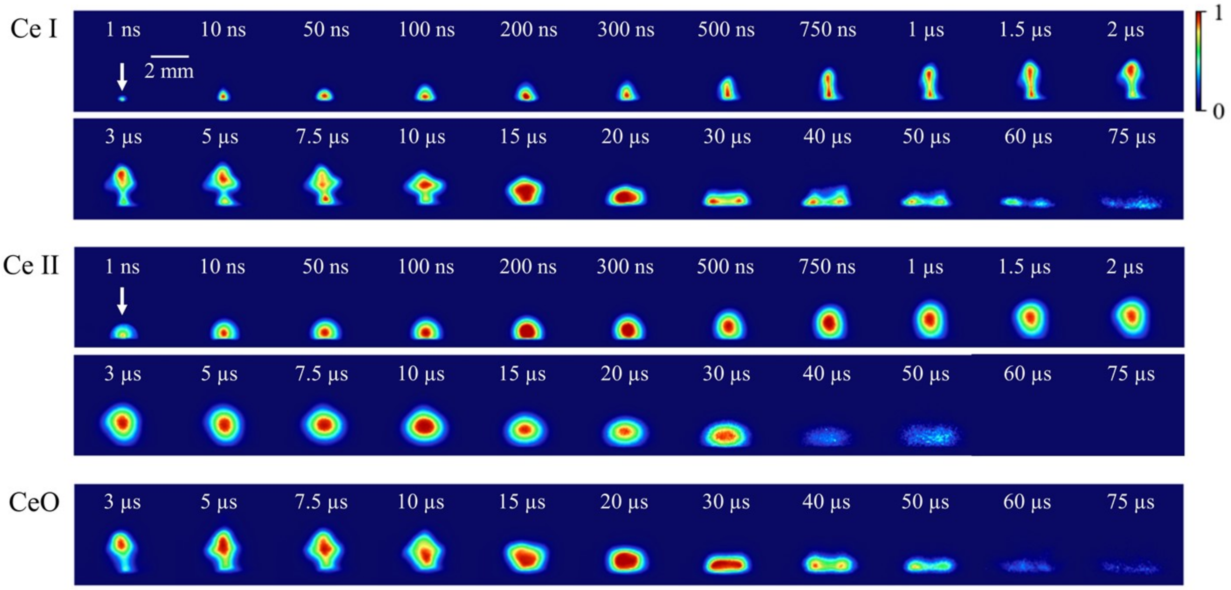

Figure S3. Time-resolved fast-gated images of Ce LPP plasma emission in argon with Y_{O_2} = 0.05 at atmospheric pressure. Each individual frame is normalized to its minimum and maximum pixel intensities.

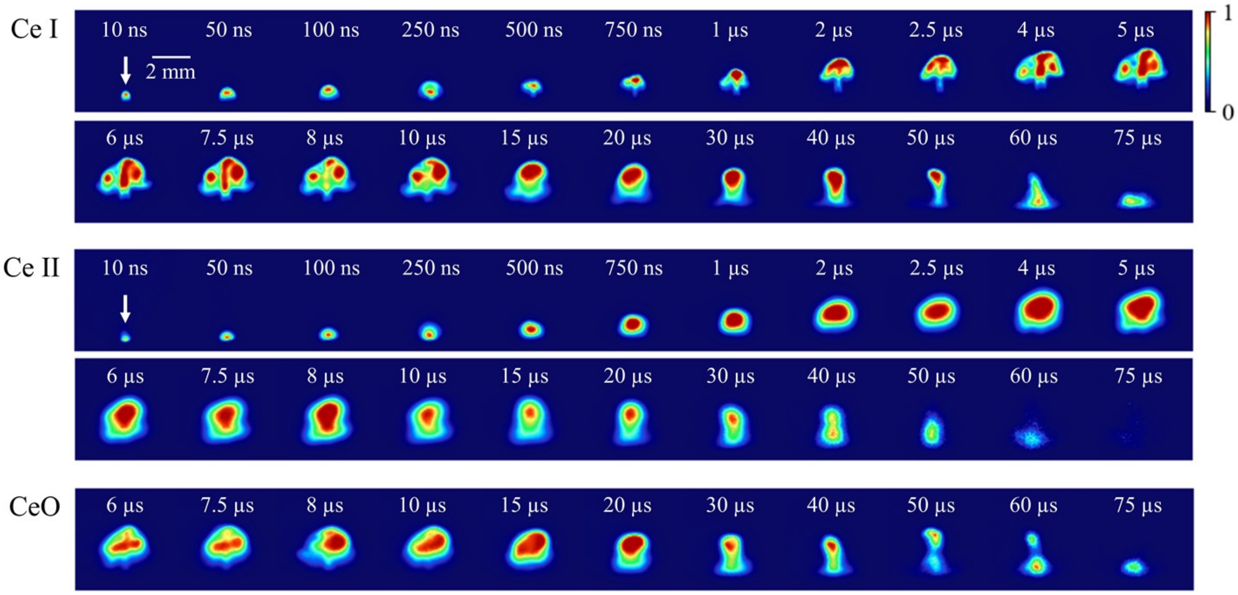

Figure S4. Time-resolved fast-gated images of Ce LPP plasma emission in argon with Y_{O_2} = 0.10 at atmospheric pressure. Each individual frame is normalized to its minimum and maximum pixel intensities.

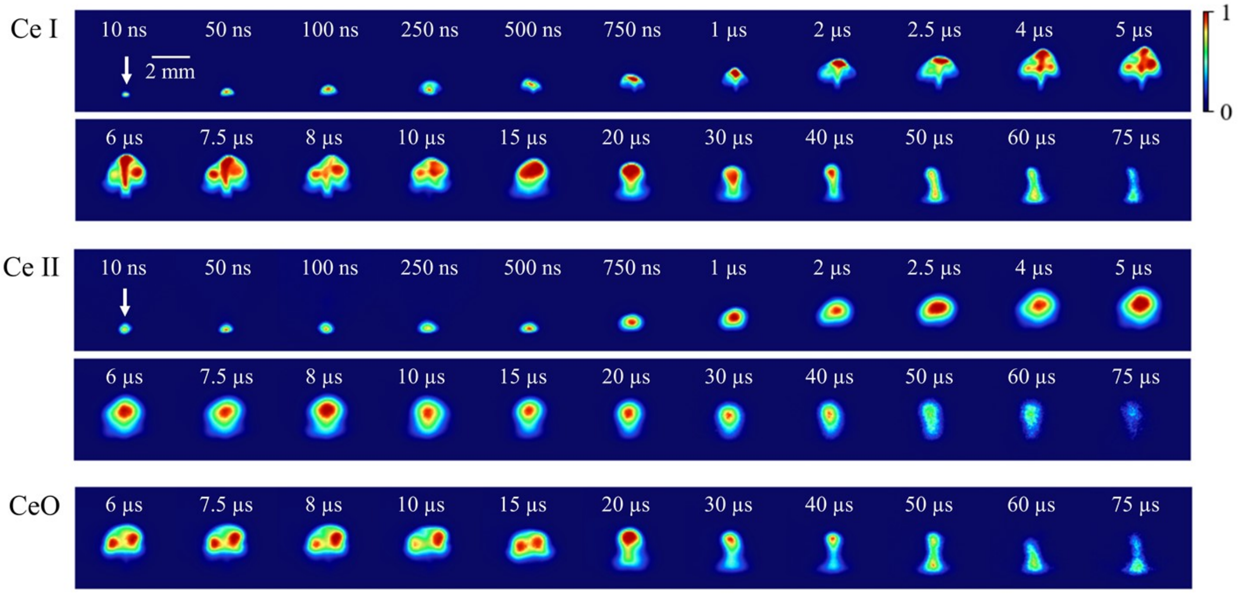

Figure S5. Time-resolved fast-gated images of Ce LPP plasma emission in argon with $Y_{O_2} = 0.20$ at atmospheric pressure. Each individual frame is normalized to its minimum and maximum pixel intensities.

Mean adjusted pixel intensities are extracted from the images provided in Figures S1-S5, and are provided and compared in Figures S6 and S7.

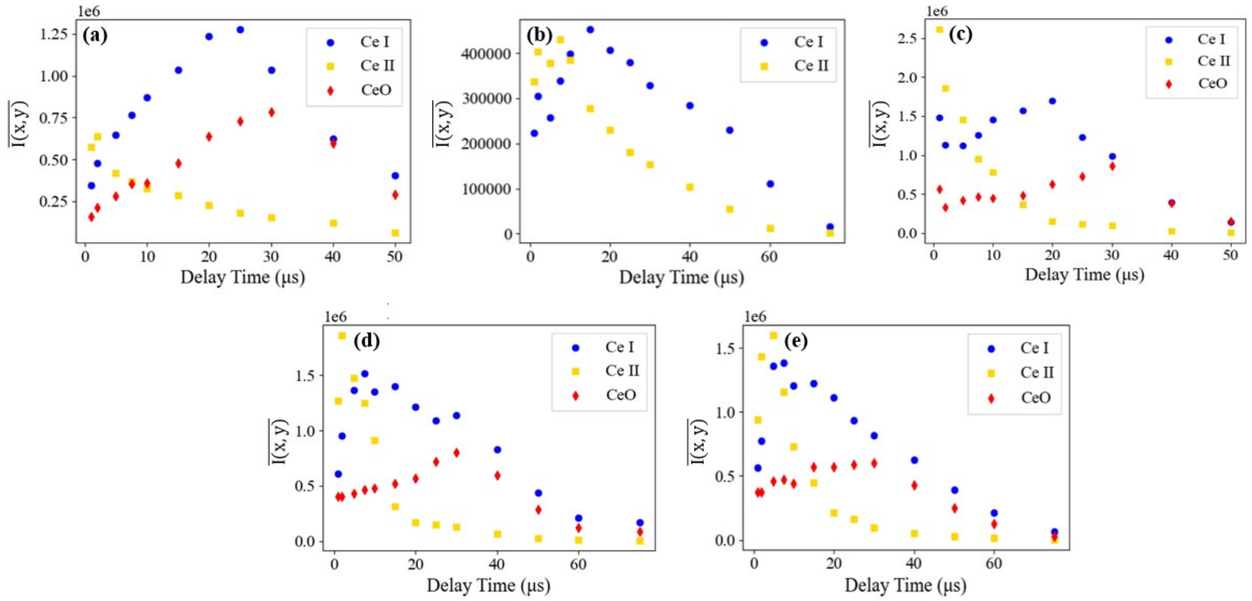

Figure S6. Mean adjusted pixel intensities for images collected at atmospheric pressure in (a) air, (b) argon, (c) argon with $Y_{O_2} = 0.05$, (d) argon with $Y_{O_2} = 0.10$, and (e) argon with $Y_{O_2} = 0.20$. Pixel intensities are not comparable between the separate subfigures.

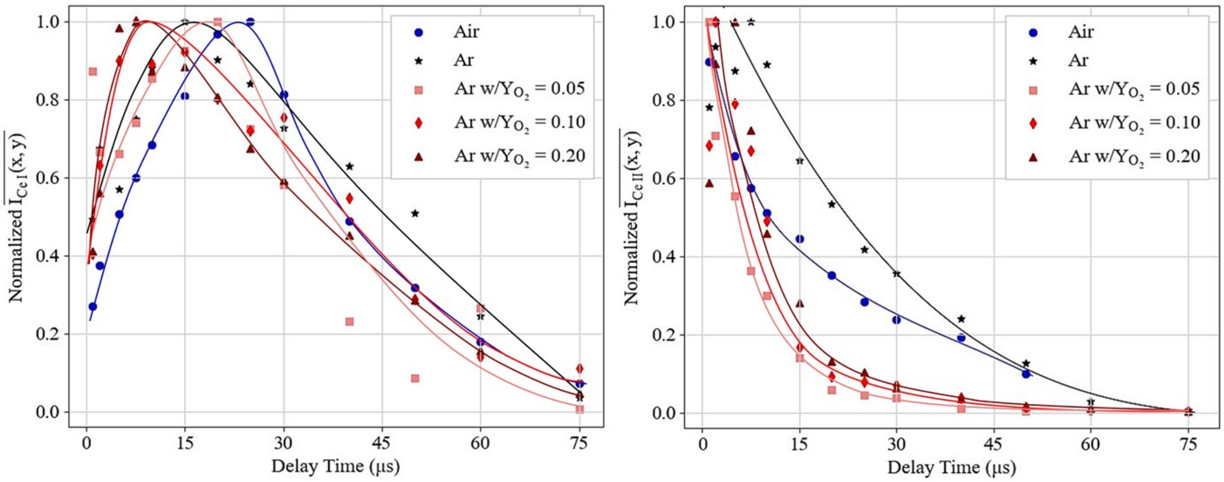

Figure S7. Comparison of normalized mean pixel intensities extracted from spectrally-integrated images of the plasma emission for the (left) Ce I filter centered at 560.23 nm and the (right) Ce II filter centered at 418.61 nm. Spline curves overlaid on the data points are drawn for visualization purposes only.

Observations on variables that impact the quality of time-resolved fast-gated images of LPPs generated from solid samples:

- *Imaging setup and resolution:* To resolve details in the images, it is best to select a magnification where the full-size plasma length covers at least 300-400 pixels. The resolution is sufficient at this magnification and there is still room to work with to image larger plasma plumes if desired.
- *Experimental parameters (i.e., gain and gate width):* Adjust the experimental parameters to acquire images that average 30,000 counts or less or the region of the plasma (i.e., draw a box around the plasma in software). Detail will be lost if the image is saturated, where saturation occurs around 60,000 counts.
- *Colormaps:* PI LightField software provides several colormaps to choose from to display the images. The ‘Jet’ colormap is useful for determining whether the image is saturated or close to saturation. Greyscale images will provide more detail on the stem and vortices of the plasma plume.
- *Laser beam quality:* Influences the energy deposited into the plasma during laser ablation as well as the crater morphology. A good, Gaussian laser beam tends to provide relatively symmetric and reproducible plasma plumes. Struggles with obtaining symmetric, reproducible plumes may be a result of poor beam quality. Additionally, random and abstract plasma shapes will result when using a deformed laser beam.
- *Laser energy:* Higher ablation energies will produce larger plasmas, although there may be greater shot-to-shot variations in the directionality and symmetry of the plasma plume. This behavior has also been observed with shadowgraphy. Aim to use a laser energy of less than 100 mJ if possible to mitigate this effect.
- *Crater morphology:* Tied to the laser beam quality; need a Gaussian laser beam to provide a circular, symmetric crater. Otherwise, random valleys and mountains that form in the crater during/after LA will affect the plasma shape and reproducibility of the measurements. The depth of the crater will also impact the morphology and emission intensity of the measured plasma.

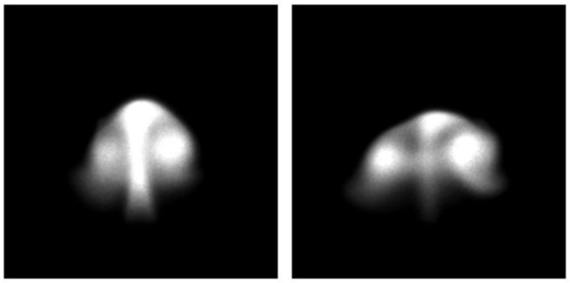

Figure S8. Images of Ce I emission collected at a time delay of 6 μs for different craters.

- *Spot size:* Affects the plasma plume morphology and behavior. Have observed pronounced plume splitting in measurements at later times (>10 μs); this behavior was eliminated by decreasing the spot size.
- *Filters:* The stem of the plasma may not be resolvable because of the filter being used (e.g., I've never been able to resolve the stem of the plasma when targeting Ce II emission). I highly recommend using narrowband filters (<1 nm FWHM) if possible, especially if targeting molecular emission that falls very close to and/or overlaps with atomic lines. The narrowband filters seem to help in resolving finer details of the plasma (e.g., the stem).