5125
LA-UR-08-04111

Approved for public release;
distribution is unlimited.

Title: In-Line Holographic Imaging and Electron Density Extraction
of Ultrafast Ionized Air Filaments

Author(s): George Rodriguez (MPA-CINT)
Anthony R. Valenzuela (MPA-CINT)
Balakishore Yellampalle (MPA-CINT)
Ki-Yong Kim (MPA-CINT)
Mark J. Schmitt (X-1-PTA)

Intended for: Journal of the Optical Society of America B

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

In-Line Holographic Imaging and Electron Density Extraction of Ultrafast Ionized Air Filaments

George Rodriguez,^{1,*} Anthony R. Valenzuela,¹ Balakishore Yellampalle,^{1,2}

Mark J. Schmitt,³ and Ki-Yong Kim¹

¹*Material Physics and Applications Division, Los Alamos National Laboratory,*

Mail Stop K771, P.O. Box 1663, Los Alamos, NM 87545, USA

²*Currently with Luna Innovations Inc.,*

3157 State Street, Blacksburg, VA 24060, USA

³*Applied Physics Division, Los Alamos National Laboratory,*

Mail Stop F699, P.O. Box 1663, Los Alamos, NM 87545, USA

**Corresponding author: rodrigeo@lanl.gov*

In-line spatial holographic methods are used to image the optical diffraction patterns of ultrafast laser produced air filaments. Using femtosecond laser pump-probe methods, an ionized air channel filament is produced and subsequently probed by a time-delayed probe beam traversing the filament at a right angle. The resulting probe beam image is recorded as a holographic diffraction pattern on a CCD camera as the probe beam is diffracted due to the optical index change caused by the air plasma filament. Extraction of the electron density in the filament down to 10^{16} cm^{-3} is obtained by using the Helmholtz wave equation beam propagation model to simulate the data. By varying the pump-probe optical time delay, a plasma electron recombination time of approximately 250 picoseconds is measured and agrees with our rate equation population kinetics model. © 2008 Optical Society of America

OCIS codes: 100.0118, 110.1650, 120.2880, 260.5210, 280.5395.

1. Introduction

Capturing images of dynamically evolving femtosecond laser generated ionized air channels continue to receive interest due to their potential as conduits for guiding optical energy, remote sensing spectroscopy, LIDAR ranging, and trigger guiding lightning discharges for safety applications [1,2]. Ionized air channels also serve as media for studying complex non-linear optical phenomena resulting from the balance between Kerr self-focusing and plasma defocusing of the laser light inside the ionized filament. Consequently there is interest in

understanding the complex plasma dynamics contained in the channel. Previous experimental studies have used a variety of methods to extract the electron density from ionized air channels including: electrical conductivity measurements [3–6], interferometry [7–10], time-resolved fluorescence molecular spectroscopy [11–13], and optical diffractometry [14]. Of these methods, there has been conflicting reports and variation of the measured electron density in the filament by several orders of magnitude (10^{14} - 10^{18} cm $^{-3}$). Yet, robust and accurate methods for electron density measurements are still desired and are necessary for understanding complex plasma dynamics associated with generation (field-, multiphoton-, and avalanche-ionization) and recombination mechanisms across the range of laser intensities and time scales.

Of the filament electron density measurements made to date, the optical diffractometry method is explored here because of its inherent ability to make spatially- and temporally-resolved measurements. Spatial resolution is partially determined by the probe diameter dimension as it traverses the filament (see Fig. 1) in an orthogonal geometry. This allows for one to sample the electron density at several positions along the filament if desired. Temporal resolution is determined by the probe pulse duration, and allows for studying femtosecond time-gated plasma dynamics. Neither the electrical conductivity [3–6] nor spectroscopic methods [11–13] have the time resolution required for observing picosecond plasma recombination dynamics. However, because of the small filament diameter 50 - 200 μ m in the optical diffractometry method, the probe beam samples a short plasma pathlength and the over all phase change imparted to the probe beam is small, making a small electron density

induced index change difficult to measure. Nonetheless, successes of the optical diffractometry method in ionized air filaments [14, 15] and more recently in transparent condensed media [16, 17], lend support to the validity of the technique. Consequently, in this article we present an extended set of measurements with the aim at establishing a lower electron density limit of this technique than has previously been published. We also describe in detail our analysis method of in-line holographic reconstruction via our beam propagation code for simulating the recorded diffraction patterns.

Rooted in principle in early work by Gabor [18], the basis for understanding the interference pattern setup by in-line holographic interference of light was demonstrated. Since there are no separate signal and reference beams for the hologram generation, the technique is referred to as “in-line” as the light diffraction pattern being recorded is actually an interference pattern between the rays scattered from the filament object and the unscattered rays. The technique was recently demonstrated in condensed media experiments by Centurion *et al.* [16] including time-resolved holographically recorded movies of nonlinear Kerr dynamics in water and carbon disulfide liquids. In ultrafast air ionized filaments, the technique was demonstrated by Tzortzakis *et al.* [14] using a slightly different probe beam geometry, yet, no detailed explanation of the image reconstruction analysis was given. Moreover, measurements shown were for one laser pump energy, and it was not clear what the ultimate low electron density measurement limit using the technique was. Subsequent ultrafast ionized air filament measurements by Centurion *et al.* [15] using the holographic/optical diffractometry technique were also published, but at much higher electron densities, $n_e > 10^{18} \text{ cm}^{-3}$,

and it was also not clear if this method would be able to measure electron densities below $n_e = 10^{16} \text{ cm}^{-3}$. A sensitivity below $n_e = 10^{16} \text{ cm}^{-3}$ electron density is extremely desirable as it allows for measuring ultrafast ionized air filaments in conditions that are more closely related to naturally self-focused air filaments where electron re-attachment rates are thought to compete with recombination mechanisms. Furthermore, interest in characterizing plasma dynamics in the low electron density range also stems from long filament ranging and directed energy applications which require knowledge of some of these plasma mechanisms. Consequently, the aim of this paper is to demonstrate the technique and a detailed analysis in the low electron density regime, as well as to provide energy and time dependent studies where previous investigations have not concentrated on.

2. Experimental Setup

The experimental setup is shown in Fig. 1. An amplified Ti:sapphire laser system capable of delivering 815 nm, 50 fs, 30 mJ pulses at a 10 Hz repetition rate is used for the experiment. The laser pulse width is stretched to 100 fs (FWHM) to avoid nonlinear effects in the optics and air beam paths. Most of the available pulse energy in the laser beam is used as a pump beam to generate filaments in air. In order to maintain single filament conditions, a focusing lens (f_1) is placed in the pump beam path to produce optical field ionization. A soft focus condition is achieved with a 1 meter focal length lens, and an ionization spark length of approximately 30 cm is observed. Under hard focus conditions with a 25 cm focal length lens, an ionization spark length of approximately 5 mm is observed. Both conditions are

used to examine electron generation in the filament. However, the majority of the results in this work are under conditions of a soft focus, as we are interested in lower limit of measuring electron densities that may be near the conditions of self-focused generated filaments that lie near the detection limits of our technique. A small portion of the main laser beam is split off and sent to an optical delay stage before being sent across the filament at a right angle to interrogate the filament. This beam becomes the probe beam for the filament and is also spatially apertured with an iris to improve the spatial distribution of the beam profile. A second lens (f_2) is placed at a distance d before the filament to reduce the probe beam diameter size to nearly match the diameter of the filament. The lens f_2 has a focal length of 10 cm, and the length d typically is 8 cm. After focusing thru the filament, the probe beam is then allowed to expand naturally to be recorded on a charge coupled device (CCD) camera. The CCD camera is also precisely placed at a distance, L , 20 cm from the filament. As will be shown later, the holographic patterns recorded in the diffracted images of the probe beam, are sensitive to the distances d and L . The pump and probe beam energies are typically 15 mJ and 5 μ J, respectively. After traversing the filament, the diffracted probe beam is recorded on a Roper Scientific TE-cooled 16-bit 512 $\times$ 512 pixel (pixel size: 24 μ m $\times$ 24 μ m) camera operating in single shot mode with typical data set consisting of 100 images averaged at each pump-probe delay. One hundred images are recorded each with and without the filament. Subtraction of the averaged image without the filament from the averaged image with the filament is then done to reveal the diffracted probe beam image due to the index change produced by the electrons within the filament. Background free, or dark

field holography, is also part of our capability in these experiments (see Fig. 1). By placing a sharp edge stop near the probe focal plane to partially block the unperturbed probe beam, we can also examine if dark field recording improves our sensitivity. Finally, by blocking the probe beam entirely and mounting a near field imaging lens on our CCD camera, we can also make measurements of the filament diameter by imaging the light emitted from the filament, a measurement that aids us in our extraction of the radially dependent electron density across the diameter of the filament.

3. Results

In Figure 2 we show a series of images for two cases of filament generation, the (a) high and (b) low electron density cases. In the high electron density generation (case (a)), the filament is produced by using the hard focus $f_1 = 25$ cm focal length lens in the pump beam. In the low electron density (case (b)), the filament is produced using the soft focus $f_1 = 1$ m focal length lens in the pump beam. CCD probe beam images for both cases are shown with no filament, with a filament, and the difference image.

In the high electron density case, a clear modulation of the probe beam is observed in the diffracted probe beam image due to the abrupt index change (and the accumulated phase change) across the filament diameter. In the low electron density case, the modulation is much less obvious, but upon subtraction in the difference image, it becomes apparent that indeed electron density induced modulation is observed. Figure 3 shows lineout plots across the probe images shown in Fig. 2. In Figure 3 the lineout plots are from the (a) high and

(b) low electron density cases, respectively. The solid gray traces represent no filament cases, and the black dashed traces are lineout plots with a filament. Additionally, an inset trace is plotted to show a normalized lineout plot of the difference in each case. Notice that the vertical scale of the inset traces show that a peak-to-peak modulation of greater than 30 percent is observed for the (a) high electron density case and less than 1 percent for the (b) low electron density case. In addition to measuring the filament induced modulation on the probe beam, extraction of the electron density from Fig. 3 requires a precise estimate of the filament transverse size. We therefore also imaged the side-on fluorescence from the filament as well as took images of burn pattern spot images on thin pieces of fused silica windows to estimate the filament diameter size. Figure 4 shows an example data set of these measurements. Figure 4(a) shows a single-shot side-on image of a 2 mm segment the filament under the soft focusing ($f_1 = 1$ m) conditions at a 9.4 mJ pump pulse energy setting. Spatial calibration is done by imaging with a calibrated resolution target. The vertical lineout scan of Fig. 4(a) is shown in the inset (b), and a typical FWHM diameter of $85\text{ }\mu\text{m}$ is measured. Although the side-on fluorescence imaging provides a qualitative measure of the electron density profile, it assumes that photoemission from the imaged volume linearly correlates with the radially varying electron density. This may not be entirely correct due to the strong nonlinear interaction of the optical pulse and the air plus plasma. Furthermore, previous spectroscopic studies [19–21] have shown that filament fluorescence is attributed to ionic radiative decay in N_2^+ ($\text{B}^2\Sigma_u^+ \rightarrow \text{X}^2\Sigma_g^+$) and recombination fluorescence in excited state neutral N_2^* ($\text{C}^3\Pi_u \rightarrow \text{B}^3\Pi_g$). Therefore, side-on imaging of the filament fluorescence provides

a convoluted interpretation of the electron density radial distribution. Nevertheless, these images do represent volume regions of the filament that contain electrons and are a first order estimate of the electron density diameter size. Because of the interpretation issues associated with side-on imaging of the filament fluorescence, we also record burn spot pattern images of the filament on 1-mm thick fused silica slides. Figure 4(c) shows an optical image of the recorded burn spot. Using an optical microscope, a post shot diameter is taken from the burn spot. The diameter of the burn spot image of Fig. 4 is $120\text{ }\mu\text{m}$. Remarkably, a rough estimate using this technique is quite consistent with previously measured estimates using a variety of methods [2]. More recently, a sophisticated non-ionizing fluorescence diagnostic imaging method in helium gas has been demonstrated [22,23]. Under laser conditions similar to our experiments, Ting *et al.*, measured an air filament diameter of $\approx 130\text{ }\mu\text{m}$ to $170\text{ }\mu\text{m}$ at 15-20 mJ pulse energies and at a 50 fs pulse width using this method.

Because we are interested in the low limit electron density ($< 10^{16}\text{ cm}^{-3}$) measurement capability of this diagnostic technique, the remainder of the results (and analysis discussion) we present is for the soft pump focusing ($f_1 = 1\text{ m}$) case. Figure 5 is a plot of the measured probe peak-to-peak modulation (red squares) as a function of pump energy. As the pump energy is varied from 15 mJ down to 2.75 mJ (using various neutral density filters in the pump beam path), the normalized peak-to-peak probe modulation decreases from a maximum of near 5 percent to a minimum detectable limit of approximately 0.2 percent modulation. For reference, the recorded side-on fluorescence measurements (black dots) are plotted in Fig. 5. The transverse filament FWHM size is also seen to follow the same trend as the peak-to-peak

modulation. That is, as the energy is decreased from 15 mJ to 2.75 mJ, the filament size diameter decreases from a maximum of 230 μm down to 55 μm . Since it is important to understand electron recombination dynamics in the filament, we also vary the pump-probe time delay. In Figure 6 is a plot of the measured probe beam peak-to-peak modulation as a function of pump-probe time delay for two separate pump energies, 15.2 mJ (red squares) and 8.2 mJ (black dots). In both cases, a rapid decrease in the peak-to-peak modulation is observed as the time delay is increased. This temporal behavior is attributed to loss of electron density from plasma recombination and not plasma expansion as the time scale is too short for significant hydrodynamic expansion. A single exponential decay fit to the 15.2 mJ data reveals a $1/e$ lifetime of 224 ps.

In order to see if the lower limit sensitivity could be improved, dark field imaging was also attempted. In Figure 1 we illustrate that by placing a razor blade edge in the probe focal plane, we can capture “background free” images with and without the filament. Results using this technique are shown in Fig. 7 at a low pump energy of only 2.75 mJ per pulse. In Fig. 7 we show “background free” CCD images for two cases: (a) no filament and (b) with filament. In each case there still is enough probe light scattering around the razor blade edge. However, upon subtraction of the images in Figs. 7(a) and 7(b), a clear modulation due to the filament is observed in Fig. 7(c). A lineout plot of the modulation is shown in Fig. 7(d), and we demonstrate that a peak-to-peak modulation below 0.2 percent is clearly evident. However, this is roughly the same sensitivity we observed in the “non-background free” mode.

4. Analysis

Until now we have primarily been representing the filament electron density in terms of a dimensionless quantity referred to as the filament induced probe beam peak-to-peak modulation. This quantity has been plotted as difference of two probe beam CCD images (with and without the filament) normalized by the peak intensity without the filament, *i.e.*, $\Delta I/I_o$. In this section we describe our model of forward beam propagation approach to compare the predicted CCD recorded patterns with the data. The beam propagation code is used to simulate the gaussian probe beam as it propagates through a focusing lens and traverses the filament containing a plasma index profile before being recorded as an intensity diffraction pattern at the CCD recording plane. Figure 8(a) shows the geometry we use for the beam propagation simulations. The plasma index is assumed to be radially varying and is comprised of a filament-sized volume bundle of electrons. The plasma index variation across the filament diameter is given by:

$$n(r) = (1 - \omega_p^2/\omega_o^2)^{1/2}, \quad (1)$$

$$\omega_o = 2\pi c/\lambda, \quad (2)$$

$$\omega_p = (n_e(r)e^2/m_e\epsilon_o)^{1/2}, \quad (3)$$

where, $n(r)$ is the radial plasma index; ω_p is the plasma frequency; ω_o is the probe beam laser frequency; λ is the probe beam laser wavelength; $n_e(r)$ is the radial electron density profile. Calculation of the phase change, $\Delta\phi(x, y)$, imparted to the traversing probe beam

after passing the plasma is given by:

$$\Delta\phi(x, y) = \frac{2\pi}{\lambda} \int_0^{L_p} n(r) dz, \quad (4)$$

where, L_p represents the radial spatial extent of the plasma filament. The co-ordinate system used is shown in Fig. 8(b). In the above equation, we have assumed that the phase change due to the filament is concentrated at a single transverse plane, similar to a thin lens. This is valid because the diffraction length due to the imparted phase is significantly large compared to the plasma diameter or the probe travel length in the plasma.

The beam propagation simulations require solving the linear Helmholtz equation:

$$\frac{\partial^2 A}{\partial z^2} + \nabla_{\perp}^2 A + k^2 A = 0, \quad (5)$$

where A is the probe laser field envelope. The measured intensity at the CCD plane is given by the following convolution:

$$I_{CCD} = |A_0(x, y)[1 + S_0(x, y)] \otimes h_{0 \rightarrow CCD}|^2, \quad (6)$$

where, A_0 and S_0 are the probe field and the plasma phase in the object plane (plasma), and $h_{0 \rightarrow CCD}$ is the transfer function from the object plane to the CCD plane. Rearranging terms in Eq. 6, we can express the intensity at the CCD as:

$$I_{CCD} = |A_1|^2 \left[1 + \frac{m_1^* A_1 + m_1 A_1^*}{|A_1|^2} \right], \quad (7)$$

where $A_1 = A_0 \otimes h_{0 \rightarrow CCD}$ and $m_1 = A_0 S_0 \otimes h_{0 \rightarrow CCD}$ represent convolution operations between the probe (and probe-modulated) field and transfer function. The free space propagation

transfer function $h_{0 \rightarrow CCD}$ is given by:

$$h_{0 \rightarrow CCD} = \frac{\exp(jk_z z)}{j\lambda z} \exp\left[\frac{jk_z}{2z}(x^2 + y^2)\right]. \quad (8)$$

The fringe contrast in this technique is determined by the modulation in m_1 , diffracted component of the field at the CCD plane. We can expect this quantity to vary with the distance between the object and CCD. This is because A_0 and $A_0 S_0$ have different spatial phase and consequently the distance dependent convolution ($h_{0 \rightarrow CCD}$) results in different rates of diffraction for the two fields. Therefore, the modulation depth on the CCD can be maximized by choosing the distances optimally. Rather than calculating the convolutions in Eq. 7 directly in the space domain, the convolutions are numerically evaluated in the Fourier domain using Fast Fourier Transforms where convolutions are evaluated by simple multiplication of the field with the appropriate Fourier domain transfer function. For example, two dimensional free space propagation along z is given by:

$$\tilde{A}(k_x, k_y, z) = \tilde{A}(k_x, k_y, 0) e^{j\frac{z}{2k_z}(k_x^2 + k_y^2)}. \quad (9)$$

In the problem depicted in Fig. 8, two transfer functions, $\tilde{h}_d$, and $\tilde{h}_L$, are defined as we propagate the probe field from left to right.

$$\tilde{h}_z = e^{j\frac{(k_x^2 + k_y^2)z}{2k_z}}, \quad (10)$$

before the CCD plane is reached. The phase due to the lens, T_f , and the filament, T_p , are accounted by multiplication in the spatial domain with the probe profile $A(x, y, z)$ at the phase location, z .

$$T_f = e^{j\frac{k_z(x^2 + y^2)}{2f}}, \quad (11)$$

and

$$T_p = e^{-j\Delta\phi(x,y)}. \quad (12)$$

Here L_p is the full-width at half-maximum of the plasma filament. The plasma index phase change $\Delta\phi$ is assumed to have a Gaussian distribution across the filament diameter,

$$\Delta\phi(x, y) = \sqrt{2\pi}wk_z\Delta N(x), \quad (13)$$

and w and $\Delta N(x)$ are given by:

$$w = \frac{L_p}{2\sqrt{2\ln 2}}, \quad (14)$$

$$\Delta N(x) = \frac{n_{eo}e^2/m_e}{\epsilon_o\omega_o^2}e^{-\frac{x^2}{2w^2}}. \quad (15)$$

Here n_{eo} is the electron density at the center of the filament and the transverse variation of index, $\Delta N(x)$, is derived using,

$$\Delta n(r) = n(r) - 1 \approx \frac{1}{2}(\omega_p^2/\omega_o^2) \propto n_e(r), \quad (16)$$

The radial electron density profile, $n_e(r)$, is assumed to be,

$$n_e(r) = n_{eo}e^{-\frac{r^2}{2w^2}}. \quad (17)$$

After propagation through the plasma filament, free space propagation of distance L (see Fig. 8) is then computed to compute the field at the CCD image plane. Final intensity at the image plane is computed from $|\tilde{A}\tilde{A}^*|$. Ultimately, the input experimental parameters that determine the diffraction pattern are λ , f , L_p , d , L , and n_{eo} , in addition to the spatial calibration of the CCD individual pixel and array sizes. The beam propagation method

outlined above can be used to obtain optimal distances that result in maximum contrast values in our experiments. The contrast as a function of L and d , when all other parameters are held constant is shown in Fig 9. For these simulations, $n_{eo} = 1 \times 10^{17} \text{ cm}^{-3}$, $f = 150 \text{ mm}$, and $L_p = 150 \text{ }\mu\text{m}$ are used. We notice that contrast is maximum along one of the marked dotted lines. Our experimental values of $L = 20 \text{ cm}$ and $d = 8 \text{ cm}$ result in a contrast of approximately 3% which is reasonably close to the maximum.

In Figure 10 we demonstrate calculated fits to our data from Figs. 2 and 3. The extracted filament diameter and electron density for the hard pump focus and high electron density case (a) is $L_p = 145 \text{ }\mu\text{m}$ and $n_{eo} = 2 \times 10^{18} \text{ cm}^{-3}$, respectively. Similarly, the extracted filament diameter and electron density for the soft pump focus and low electron density case (b) is $L_p = 100 \text{ }\mu\text{m}$ and $n_{eo} = 7 \times 10^{16} \text{ cm}^{-3}$, respectively. The simulated results for these two cases are consistent with the trend in Fig. 5 that shows a decreasing filament diameter size and peak-to-peak modulation depth in the diffracted pattern image as the pump laser energy is decreased.

In a similar manner as in the two cases for Fig. 2 we proceed to extract the electron density from the lineout data used to generate the pump-probe time-resolved plot of Fig. 6. The corresponding data for the electron density versus pump-probe time delay plot is shown in Fig. 11. For the case of $E = 15.2 \text{ mJ}$ pump energy (squares), an initial peak electron density of $n_{eo} = 1.75 \times 10^{17} \text{ cm}^{-3}$ is extracted at $\tau = 0$ pump-probe time delay. By $\tau = 0.53 \text{ ns}$, the peak electron density has dropped over an order of magnitude to $n_{eo} = 1.6 \times 10^{16} \text{ cm}^{-3}$. For the case of $E = 8.2 \text{ mJ}$ pump energy (dots), and initial peak electron density of $n_{eo} = 9.5 \times 10^{16} \text{ cm}^{-3}$ is

extracted at $\tau=0$ pump-probe time delay. At $\tau \geq 0.4$ ns, the peak electron density has dropped below $n_{eo} = 2 \times 10^{16} \text{ cm}^{-3}$. Another feature in Fig. 11 is the similarity in the long tail decay rate between the two cases despite having different initial ($\tau=0$) electron density values and radial distributions. After approximately 150 ps, the decay rates in each case show a similar trend; yet, clearly early time dynamics (<100 ps) are dissimilar. The result is significant in that it suggests that early recombination dynamics show an intensity dependent component that can not be extracted from consideration of the peak-to-peak modulation depth alone, such as the data and exponential decay data fits shown in Fig. 6. Folded into the data interpretation and extraction is the fact that the filament diameter, L_p is dependent on the pulse energy fluence. The inset graph in Fig. 11 shows the normalized radial electron density profile used in the extraction of the electron density results. For the $E=15.2$ mJ data, a filament diameter of $L_p=175 \text{ } \mu\text{m}$ was used, and for the $E=8.2$ mJ data, a filament diameter of $L_p=100 \text{ } \mu\text{m}$ was used. These differences in the filament diameter, are clearly evident in the measured probe beam diffraction pattern recorded by the CCD camera. We observe that as the pump energy decreases and the filament diameter decreases, the oscillatory period in the CCD recorded diffraction pattern also decreases, consistent with decreasing scale object size effects on expected diffraction pattern frequencies.

5. Simulations

In order to simulate the time-resolved electron density decay rates observed in Fig. 11, a zero-dimensional rate-equation based model has been applied to ultrafast ionization of air for a

laser intensity in the $I = 10^{13}$ W/cm² to $I = 10^{15}$ W/cm² range [25]. This model includes the effects of (a) multi-photon and tunneling ionization by the ultrashort laser pulse; (b) further electron by avalanche and impact ionization; (c) attachment of electrons to form negative molecular oxygen ions, O₂⁻; (d) recombination between electrons and positive ions; (e) recombination between negative and positive ions; and (f) depletion of the neutral gas.

We consider an ultra-short pulse laser with intensity I applied into a neutral gas of air. The pulse length of the laser, τ_L , is 100 fs, and a gaussian pulse shape. The rate equations for electrons (n_e), positive ions (n_+), negative ions (n_-), and neutrals (n) are:

$$\frac{dn_e}{dt} = Z + \alpha n_e - \eta n_e - \beta_{e+} n_e n_+ \quad (18)$$

$$\frac{dn_+}{dt} = Z + \alpha n_e - \beta_{e+} n_e n_+ - \beta_{+-} n_+ n_- \quad (19)$$

$$\frac{dn_-}{dt} = \eta n_e - \beta_{+-} n_+ n_- \quad (20)$$

$$\frac{dn}{dt} = -Z - \alpha n_e - \eta n_e + \beta_{e+} n_e n_+ + \beta_{+-} n_+ n_- \quad (21)$$

where $Z, \alpha, \eta, \beta_{e+}$, and β_{+-} , are rate coefficients adopted from various references [24–26].

Z is the optical field ionization rate; α is the collisional ionization rate; η is the attachment rate; β_{e+} is the electron-positive ion recombination rate coefficient; β_{+-} is the ion-ion recombination rate.

The ionization source term, $Z = W \times n$ is comprised of two terms associate with the optical ionization rates (multi-photon and tunneling) of the constituent air molecules, molecular O₂ and N₂, where $Z = W_{O_2} \times n_{O_2} + W_{N_2} \times n_{N_2}$. The optical ionization rate coefficients W_{O_2} and W_{N_2} are exponentially-modified power law fits to the semi-empirical

Perelomov, Popov and Terent'ev (PPT) field ionization model [27], which can be written $W = W_o(I/I_o)^a \exp[-0.71(I/I_o)]$, where $I_o = 10^{13} \text{ W/cm}^2$. W_o and a are, respectively, $2.8 \times 10^6 \text{ s}^{-1}$ and 6.5 (for oxygen), and $2.5 \times 10^4 \text{ s}^{-1}$ and 7.5 (for nitrogen), obtained by fitting with experimental results [25, 28]. Note this fitting is only valid for laser intensity up to $I = 10^{15} \text{ W/cm}^2$ for a Ti:sapphire laser with a central wavelength of 800 nm. The densities of oxygen and nitrogen are assumed to be $n_{O_2} = 0.21 \times n$ and $n_{N_2} = 0.79 \times n$, respectively, where n is the atmospheric air density at Los Alamos, New Mexico, USA (590 Torr).

At atmospheric densities without external fields, and for femtosecond laser fields, the collisional ionization rate (α) is negligible. However, at atmospheric air densities, liberated electrons do have a tendency to quickly attach to oxygen molecules, creating negative molecular ions such as O_2^- through a 3-body reaction with rate (η_3). Other negative ions like O^- are formed through a 2-body reaction with rate (η_2). Thus, the total electron attachment rate (assuming these two mechanisms dominate) is $\eta = \eta_2 + \eta_3$ and is taken to be $2.5 \times 10^7 \text{ s}^{-1}$ [10, 24].

For time scale longer than the laser pulse length, the recombination rate between electron, negative ion and positive ions will become important. Here, we consider electron-positive ion recombination characterized by the rate β_{e+} and ion-ion recombination characterized by the rate β_{+-} . These rates are defined as (see Ref. [24], Eq. 49 and 50, or Ref. [26], Eqs. 40, 38 and Eq. 1 of section 2.7),

$$\beta_{e+}[\text{cm}^3/\text{s}] = \beta_{e+}[O_2^+] + \beta_{e+}[N_2^+] \quad (22)$$

$$\beta_{e+}[O_2^+] = 2 \times 10^{-7} (300/T_e) \quad (23)$$

$$\beta_{e+}[N_2^+] = 2.8 \times 10^{-7} (300/T_e)^{1/2} \quad (24)$$

and,

$$\beta_{+-} = 2 \times 10^{-7} (300/T_e)^{1/2} \quad (25)$$

where T_e is the electron temperature in units of Kelvin.

In solving the equations, the initial conditions (at $t=0$) of $n = n_o$, $n_e = n_- = n_+ = 0$ are assumed, and the gaussian laser pulse is turned “on” only during the time interval of the laser pulse length from $t=0$ to $t=\tau_L=100$ fs. The solid curves in the main plot are of Fig. 11 represent the best simulation fits to the experimental data for both cases, pulse energies of $E=15.2$ mJ (squares) and $E=8.2$ mJ (dots). In each case a transverse averaged laser intensity of $I = 7.6 \times 10^{13}$ W/cm² and 6.8×10^{13} W/cm² are used to fit the data for $E=15.2$ mJ and $E=8.2$ mJ, respectively. An electron temperature of $T_e=1500$ K is used to best fit the tail in the decay of the data in Fig. 11. This yields a set of recombination coefficients: $\beta_{e+} = 1.7 \times 10^{-7}$ cm³/s and $\beta_{+-} = 8.9 \times 10^{-8}$ cm³/s. These recombination rate coefficient numbers are slightly lower than have been previously published [10,14,24] ($\beta_{e+} = \beta_{+-} = 2.2 \times 10^{-7}$ cm³/s), but this is because in previous studies the electron temperature had been assumed to be $T_e=300$ K, where heating of the electrons and gas had been ignored. This is evident in the paper by Tzortzakakis *et.al.* [14] where empirically lowering both, β_{e+} and β_{+-} , to 1.2×10^{-7} cm³/s better fit their results at an initial electron density of $n_{eo} = 2 \times 10^{17}$ cm⁻³.

6. Discussion

The results of these studies can be put into perspective by comparison with previous ultra-fast laser generated air filaments and their corresponding attempts in measuring electron densities in these systems. Our studies were performed in the optical energy fluence regime of $F \leq 40 \text{ J/cm}^2$ (assuming a 15 mJ and 220 μm optical pump beam energy and FWHM diameter). Focal intensities approach $5 \times 10^{14} \text{ W/cm}^2$, and tunneling ionization is expected to significantly contribute to electron production in addition to multi-photon ionization. Our electron density measurements are remarkably consistent with previous optical pump-probe diffractometry measurements made by Tzortzakis *et al.* [14], where initial electron densities of $n_{eo} = 2 \times 10^{17} \text{ cm}^{-3}$ were measured for a 14 mJ, 120 fs, 800 nm beam focused in a air with a 2 meter lens. Similar electron population decay times were also observed. In those studies, however, the laser pump energy was not varied, and effects of varying plasma filament size on decay dynamics were not addressed. Later studies, by Yang *et al.* [10], at somewhat higher laser energies (45 mJ, 25 fs, 800 nm) using a several meter focusing optic, significantly higher densities were measured. In the studies by Yang, an inline Wollaston interferometer was used to detect the plasma induced fringe shift, and initial electron densities of $n_{eo} = 2.7 \times 10^{18} \text{ cm}^{-3}$. The higher electron density result than our studies can be accounted for by the fact that their focal laser intensities (assuming their measured optical diameter size of 120 μm) produces focal intensities $1.6 \times 10^{16} \text{ W/cm}^2$, similar to our hard pump focus condition results of Figs. 2,(a) 3(a) and 10(a) where we extract electron den-

ties of $\approx 2 \times 10^{18} \text{ cm}^{-3}$. Another comparison with our results can be made to experiments by Liu *et al.* [8] where they instead used the method of longitudinal diffractometry to extract electron density in the “low” limit where an initial electron density of $n_{eo} = 5 \times 10^{16} \text{ cm}^{-3}$ and low limit of $5 \times 10^{15} \text{ cm}^{-3}$ at a pump-probe time delay of 0.83 ns was measured. Their filament generation conditions consisted of a 800 nm, 0.6 mJ, 50 fs ultrafast laser beam focused by an 40 cm focal length lens in air. Their results are consistent with our results in both, n_{eo} and the $n_e(t)$. However, it is not obvious from their paper how one would easily implement this method for electron density extraction as one varies the pump energy since issues such as exact filament plasma length and diameter then become more difficult to account for in the experimental procedure and analysis. Nonetheless, the method in this paper of transverse diffractometry and the method of longitudinal diffractometry [8] both demonstrate the ability to measure electron densities below 10^{16} cm^{-3} .

Finally, we also note that consideration of effects due to refraction in the probe beam from the plasma index is deemed as negligible. We have only considered a purely phase disturbance (Eqs. 4 and 12) on the probe field from the filament channel. However, an estimate of the upper electron density limit where this estimate breaks down and at where refraction effects begin to dominate the fringe contrast ratio can be made. Typically, as one approaches electron densities near one percent level of critical density ($n_{ecrit} = 1.6 \times 10^{21} \text{ cm}^{-3}$ at $\lambda=800 \text{ nm}$), or plasmas with steep gradients, alternative techniques such as Schlieren or Moiré deflectometry must be used as the plasma index gradient begins to cause significant bending of the probe beam rays. None of our measurements here approach these densities as it would mean full

depletion of neutrals, assuming all the gas is singly-ionized ($n_e = 1.9 \times 10^{19} \text{ cm}^{-3}$ at 580 torr). For interferometry, Lisitsyn *et al.* [29] computed the maximum acceptable plasma index induced deflection that can occur before refraction renders interferometric techniques less robust. More recently Ruiz-Camacho *et al.* [30], made direct comparison of interferometric techniques with Moiré deflectometry and studied conditions where one method is preferable over the other.

7. Summary

An ultrafast time-resolved in-line holographic imaging method for extracting time-dependent electron densities in ultrafast ionized air filaments is demonstrated. We show that electron densities below 10^{16} cm^{-3} can be measured with this technique, and that the sensitivity of the technique is a strong function of the distances between the probe beam imaging lens and filament position (d), and the filament position to camera imaging plane (L) (see Fig. 9). We also show that the created plasma filament diameter is a strong function of pump laser input energy and should be taken into consideration when extracting the electron density from the analysis method presented. Our numerical simulations of the time dependent population dynamics reveal excellent agreement between experiment and theory for describing the plasma recombination dynamics.

8. Acknowledgments

This work is funded through a work for others contract for the US Army Armament Research, Development and Engineering Center (ARDEC) in Picatinny, NJ and in conjunction with Applied Energetics, Incorporated in Tucson, AZ. All work at Los Alamos National Laboratory is carried out under the auspices of the Department of Energy contract number DE-AC52-06NA25396.

References

1. S. L. Chin, S. A. Hosseini, W. Liu, Q. Luo, F. Théberge, N. Aközbek, A. Becker, V. P. Kandidov, O. G. Kosareva, and H. Schroeder, “The propagation of powerful femtosecond laser pulses in optical media: physics, applications, and new challenges”, *Can. J. Phys.* **83**, 863 (2005).
2. A. Couairon and A. Mysyrowicz, “Femtosecond filamentation in transparent media”, *Phys. Rep.* **441**, 47 (2007).
3. S. Tzortzakis, M. A. Franco, Y.-B. André, A. Chiron, B. Lamouroux, B. S. Prade, and A. Mysyrowicz, “Formation of a conducting channel in air by self-guided femtosecond laser pulses”, *Phys. Rev. E* **60**, R3505 (1999).
4. A. Ting, I. Alexeev, D. Gordon, R. Fischer, D. Kaganovich, T. Jones, E. Briscoe, J. Peñano, R. Hubbard, and P. Sprangle, “Measurements of intense femtosecond laser pulse propagation in air”, *Phys. Plasmas* **12**, 056705 (2005).
5. H. Schillinger and R. Sauerbrey, “Electrical conductivity of long plasma channels in air

- generated by self-guided femtosecond laser pulses”, *Appl. Phys. B* **68**, 753 (1999).
6. R. Fischer, A. Ting, D. F. Gordon, R.F. Fernsler, G. P. DiComo, and P. Sprangle, “Conductivity measurements of femtosecond laser-plasma filaments”, *IEEE Trans. Plasma Sci.* **35**, 1430 (2007).
 7. B. La Fontaine, F. Vidal, Z. Jiang, C. Y. Chien, D. Comtois, A. Desparois, T. W. Johnston, J.-C. Kieffer, H. Pépin, and H. P. Mecure, “Filamentation of ultrashort laser beams resulting from their propagation over long distance in air”, *Phys. Plasmas* **6**, 1615 (1999).
 8. J. Liu, Z. Duan, Z. Zeng, X. Xie, Y. Deng, R. Li, Z. Zu, and S. L. Chin, “Time-resolved investigation of low-density plasma channels produced by a kilohertz femtosecond laser in air”, *Phys. Rev. E* **72**, 026412 (2005).
 9. C. Y. Chien, B. LaFontaine, A. Desparois, Z. Jiang, T. W. Johnston, J. C. Kieffer, H. Pépin, F. Vidal and H. P. Mercure, “Single-shot chirped-pulse spectral interferometry used to measure femtosecond ionization dynamics in air”, *Opt. Lett.* **25**, 578 (2000).
 10. H. Yang, J. Zhang, Y. Li, J. Zhang, Y. Li, Z. Chen, H. Teng, Z. Weim, and Z. Sheng, “Characteristics of self-guided laser plasma channels generated by femtosecond laser pulses in air”, *Phys. Rev. E* **66**, 016406 (2002).
 11. J. Bernhardt, W. Liu, F. Thérberge, H. L. Xu, J. F. Daigle, M. Chateâuneuf, J. Dubois, and S. L. Chin, “Spectroscopic analysis of femtosecond laser plasma filament in air”, *Opt. Comm.*, (in press, 2008).

12. A. Talebpour, M. Abdel-Fattah, and S. L. Chin, "Focusing limits of intense laser pulses in a high pressure gas: road to new spectroscopic sources", *Opt. Comm.***183**, 479 (2000).
13. A. Couairon, G. Méchain, S. Tzortzakis, M. Franco, B. Lamouroux, B. Prade, and A. Mysyrowicz, "Propagation of twin pulses in air and concatenation of plasma strings produced by femtosecond infrared filaments", *Opt. Commun.* **225**, 177 (2003).
14. S. Tzortzakis, B. Prade, M. Franco, and A. Mysyrowicz, "Time-evolution of the plasma channel at the tail of a self-guided IR femtosecond laser pulse in air", *Opt. Commun.* **181**, 123 (2000).
15. M. Centurion, Y. Pu, and D. Psaltis, "Holographic capture of femtosecond pulse propagation", *J. Appl. Phys.*,**100**, 063104 (2006).
16. M. Centurion, Y. Pu, Z. Liu, D. Psaltis, and T. W. Hänsch, "Holographic recording of laser-induced plasma", *Opt. Lett.*,**29**, 772 (2004).
17. S. Minardi, A. Gopal, M. Tatarakis, A. Couairon, G. Tamošauskas, R. Piskarskas, A. Dubietis, and P. Di Trapani, "Time-resolved refractive index and absorption mapping of light-plasma filaments in water", *Opt. Lett.* **33**, 86 (2008).
18. D. Gabor, "A new microscopic principle", *Nature* **161**, 777 (1948).
19. Q. Luo, W. Lui, and S. L. Chin, "Lasing action in air induced by ultra-fast laser filamentation", *Appl. Phys. B* **76**, 337 (2003).
20. A. Becker, N. Aközbek, N. Vijayalakshmi, E. Orel, C. M. Bowden, and S. L. Chin, "Intensity clamping and re-focusing of intense femtosecond laser pulses in nitrogen molecular

- gas”, *Appl. Phys. B* **73**, 287 (2001).
21. A. Iwasaki, N. Aközbek, B. Ferland, Q. Luo, G. Roy, C. M. Bowden, and S. L. Chin, “A LIDAR technique to measure the filament length generated by a high-peak power femtosecond laser pulse in air”, *Appl. Phys. B* **76**, 231 (2003).
 22. A. Ting, D. Gordon, E. Briscoe, J. Peñano, and P. Sprangle, “Direct characterization of self-guided femtosecond laser filaments in air”, *Appl. Optics* **44**, 1474 (2005).
 23. D. Gordon, A. Ting, I. Alexeev, R. Fischer, and P. Sprangle, “Direct measurements of the dynamics of self-guided femtosecond laser filaments in air”, *IEEE Trans. on Plasma Sci.* **34**, 249 (2006).
 24. X. M. Zhao, J. C. Diels, C. Y. Wang, and J. M. Elizondo, “femtosecond ultraviolet laser pulse induced lightning discharges in gases”, *IEEE J. Quantum Electron.* **31**, 599 (1995).
 25. J. Kasparian, R. Sauerbrey, and S. L. Chin, “The critical laser intensity of self-guided light filaments in air”, *Appl. Phys. B* **71**, 877 (2000).
 26. I. A. Kossyi, A. Y. Kostinsky, A. A. Matveyev, and V. P. Silakov, “Kinetic scheme of the nonequilibrium discharge in nitrogen-oxygen mixtures”, *Plasma Sources Sci. Technol.* **1**, 207 (1992).
 27. A. M Perelomov, V. S. Popov, and M. V. Terent’ev, “Ionization of atoms in an alternating electric field”, *Sov. Phys. JETP* **23**, 924 (1966).
 28. A. Talebpour, J. Yang, and S. L. Chin, “Semi-empirical model for the rate of tunnel ionization of N-2 and O-2 molecule in an intense Ti:sapphire laser pulse”, *Opt. Commun.*

163, 29 (1999).

29. I. V. Lisitsyn, S. Kohno, S. Katsuki, and H. Akiyama, “Effect of laser beam deflection on the accuracy of interferometer measurements”, *Rev. Sci. Instrum.* **69**, 1584 (1998).
30. J. Ruiz-Camacho, F. N. Beg, and P. Lee, “Comparison of sensitivities of Moiré deflectionometry and interferometry to measure electron densities in z -pinch plasmas”, *J. Phys. D: Appl. Phys.*, **40**, 2026 (2007).

List of Figures

1	(Color online) Experimental layout for side-on holographic imaging of ultra-fast laser generated filaments. The distances d and L are adjusted to provide optimal diffraction contrast and CCD field of view coverage. In side-on filament imaging mode (dashed boxed), no probe beam is used and single-shot fluorescence images are captured by the CCD.	29
2	(Color online) Example probe beam CCD images captured under (a) hard pump beam focusing, $f_1 = 25$ cm and under (b) soft pump beam focusing, $f_1 = 1$ m. In each case the probe beam with no filament and with filament are shown, in addition to the probe beam difference images.	30
3	Example probe beam line out plots for the data in Fig. ??: (a) hard pump focus, $f_1 = 25$ cm and (b) soft pump beam focus, $f_1 = 1$ m. The solid gray traces are the probe beam line outs with no filament, and the dashed black traces are the probe beam line outs with the filament present. The inset figures are the intensity normalized difference line outs for each case.	31
4	(Color online) (a) Side-on fluorescence image of 2 mm segment of filament. (b) calibrated line out plot of (a). (c) Microscopic optical image of burn spot pattern on a piece of fused silica glass at the filament position (at pump focus).	32
5	(Color online)(a) Plot of the peak-to-peak modulation (squares) versus pump energy. (b) Plot of the filament side-on fluorescence imaging diameter (dots) versus pump energy.	33
6	(Color online) Plot of the peak-to-peak modulation versus pump-probe time delay for two pump energy settings: 15.2 mJ (squares) and 8.2 mJ (dots). Exponential fit to the data indicates a 224 ps (solid line fit to squares) and 257 ps (solid line fit to dots) plasma recombination time.	34
7	(Color online) Experimental results showing our background-free dark field imaging capability for (a) without filament and (b) with filament. The panel (c) shows a subregion where we extract the difference image showing the plasma filament induced modulation. (d) shows the intensity normalized difference line out for this case as below 0.2% peak-to-peak modulation. The pump pulse energy in this case is 2.75 mJ.	35
8	(a) Diagram showing relevant distances used in the two-dimensional beam propagation and image reconstruction model. Shown in (b) is the finite-sized diameter, L_p , of the filament.	36
9	Plot showing the fringe contrast as a function of L and d . By choosing L and d appropriately, the contrast can be maximized.	37
10	(Color online) Graphs of the intensity normalized difference lineouts from Fig. ?. The solid line plots are the experimental results and the dotted plots are the results extracted from the simulation fits for (a) hard pump focus case, $n_{eo} = 2 \times 10^{18} \text{ cm}^{-3}$, and (b) soft pump focus case, $n_{eo} = 7 \times 10^{16} \text{ cm}^{-3}$	38

- 11 (Color online) The symbols represent the analysis extracted electron densities n_{eo} versus pump-probe time delay for the 8.2 mJ (dots) and 15.2 mJ (squares) pump energy cases, respectively. The inset to the figure is a graph of the radial electron density profile used to extract the experimentally measured electron densities from the analysis at each pump energy setting. The solid curves in the main plot are the best simulation fits to each data set using the rate equation based model for the electron population dynamics. 39

Fig. 1. (Color online) Experimental layout for side-on holographic imaging of ultrafast laser generated filaments. The distances d and L are adjusted to provide optimal diffraction contrast and CCD field of view coverage. In side-on filament imaging mode (dashed boxed), no probe beam is used and single-shot fluorescence images are captured by the CCD.

(a) High electron density ($>10^{18} \text{ cm}^{-3}$) case

(b) Low electron density ($>10^{16} \text{ cm}^{-3}$) case

Fig. 2. (Color online) Example probe beam CCD images captured under (a) hard pump beam focusing, $f_1 = 25 \text{ cm}$ and under (b) soft pump beam focusing, $f_1 = 1 \text{ m}$. In each case the probe beam with no filament and with filament are shown, in addition to the probe beam difference images.

Fig. 3. Example probe beam line out plots for the data in Fig. 2: (a) hard pump focus, $f_1 = 25$ cm and (b) soft pump beam focus, $f_1 = 1$ m. The solid gray traces are the probe beam line outs with no filament, and the dashed black traces are the probe beam line outs with the filament present. The inset figures are the intensity normalized difference line outs for each case.

Fig. 4. (Color online) (a) Side-on fluorescence image of 2 mm segment of filament. (b) calibrated line out plot of (a). (c) Microscopic optical image of burn spot pattern on a piece of fused silica glass at the filament position (at pump focus).

Fig. 5. (Color online)(a) Plot of the peak-to-peak modulation (squares) versus pump energy.
(b) Plot of the filament side-on fluorescence imaging diameter (dots) versus pump energy.

Fig. 6. (Color online) Plot of the peak-to-peak modulation versus pump-probe time delay for two pump energy settings: 15.2 mJ (squares) and 8.2 mJ (dots). Exponential fit to the data indicates a 224 ps (solid line fit to squares) and 257 ps (solid line fit to dots) plasma recombination time.

Fig. 7. (Color online) Experimental results showing our background-free dark field imaging capability for (a) without filament and (b) with filament. The panel (c) shows a subregion where we extract the difference image showing the plasma filament induced modulation. (d) shows the intensity normalized difference line out for this case as below 0.2% peak-to-peak modulation. The pump pulse energy in this case is 2.75 mJ.

Fig. 8. (a) Diagram showing relevant distances used in the two-dimensional beam propagation and image reconstruction model. Shown in (b) is the finite-sized diameter, L_p , of the filament.

Fig. 9. Plot showing the fringe contrast as a function of L and d . By choosing L and d appropriately, the contrast can be maximized.

Fig. 10. (Color online) Graphs of the intensity normalized difference lineouts from Fig. 3. The solid line plots are the experimental results and the dotted plots are the results extracted from the simulation fits for (a) hard pump focus case, $n_{eo} = 2 \times 10^{18} \text{ cm}^{-3}$, and (b) soft pump focus case, $n_{eo} = 7 \times 10^{16} \text{ cm}^{-3}$.

Fig. 11. (Color online) The symbols represent the analysis extracted electron densities n_{eo} versus pump-probe time delay for the 8.2 mJ (dots) and 15.2 mJ (squares) pump energy cases, respectively. The inset to the figure is a graph of the radial electron density profile used to extract the experimentally measured electron densities from the analysis at each pump energy setting. The solid curves in the main plot are the best simulation fits to each data set using the rate equation based model for the electron population dynamics.