

Ionization rate and plasma dynamics at 3.9 micron femtosecond photoionization of airAdam Patel,¹ Claudia Gollner,² Rokas Jutas^{1,2},,² Valentina Shumakova^{1,2},,² Mikhail N. Shneider,³ Audrius Pugzlys^{1,2},,² Andrius Baltuska,² and Alexey Shashurin^{1,*}¹*School of Aeronautics and Astronautics, Purdue University, West Lafayette, Indiana 47907, USA*²*Photonics Institute, Vienna University of Technology, 1040 Vienna, Austria*³*Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544, USA*

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The introduction of mid-IR optical parametric chirped pulse amplifiers has catalyzed interest in multimillijoule, infrared femtosecond pulse-based filamentation. As tunneling ionization is a fundamental first stage in these high-intensity laser-matter interactions, characterizing the process is critical to understand derivative topical studies on femtosecond filamentation and self-focusing. Here, we report direct nonintrusive measurements of total electron count and electron number densities generated at 3.9 μm femtosecond midinfrared tunneling ionization of atmospheric air using constructive-elastic microwave scattering. Subsequently, we determine photoionization rates to be in the range 5.0×10^8 – $6.1 \times 10^9 \text{ s}^{-1}$ for radiation intensities of 1.3×10^{13} – $1.9 \times 10^{14} \text{ W/cm}^2$, respectively. The proposed approach paves the way to precisely tabulate photoionization rates in mid-IR for a broad range of intensities and gas types and to study plasma dynamics at mid-IR filamentation.

DOI: [10.1103/PhysRevE.106.055210](https://doi.org/10.1103/PhysRevE.106.055210)**I. INTRODUCTION**

Strong field ionization of gases by intense femtosecond laser pulses enables a plethora of frontier fundamental research fields in nonlinear optics, such as high-order harmonic generation (HHG) and attosecond science [1], femtosecond filamentation [2,3], standoff nonlinear spectroscopy of atmosphere [4], etc. In contrast to optical breakdown triggered by picosecond (and longer) laser pulses or microwave discharges (where Joule heating of photoelectrons and avalanche ionization play a major role in evolution of the plasma density, temperature, and energy or momentum distribution of photoelectrons), plasmas generated by femtosecond pulses in atmospheric pressure gases are fully determined by the fundamental quantum process of strong field ionization. Very recent progress in the development of high-energy femtosecond laser systems in the midinfrared (mid-IR) spectral range has opened new regimes and perspectives in strong field physics and femtosecond filamentation [5–9]. Specifically, these refer to the increased propagation range of the mid-IR laser radiation through the atmosphere in comparison to their traditional near-IR counterparts and, therefore, enable a variety of applications including remote sensing of the atmosphere, free-space communications, directed energy applications, and laser propulsion for deep-space travel [4–8,10,11].

Accurate knowledge of the plasma properties, generated by intense ultrashort mid-IR laser pulses, and their dependence on laser pulse parameters has critical importance for understanding physics and the precision of numerical simulations of HHG and generation of attosecond pulses, laser wake-field acceleration [12], femtosecond filamentation, and

related phenomena like standoff terahertz emission generation [13], laser-controlled weather phenomena [14], standoff lasing [4,15], etc. The dependence of ionization rate on laser intensity and wavelength for atomic gases was theoretically described in seminal works [16–18]. In the latest development of these theoretical models, it was shown that the ionization probability as well as the momentum and energy distributions of photoelectrons are strongly affected by the laser wavelength and polarization [19]. However, even for atomic gases, there are no absolute measurements of the plasma parameters and ionization rates generated by the laser pulse in mid-IR, and experiments conducted thus far are mostly in a narrow spectral range 0.8–1 μm with near-IR lasers, providing semiempirical agreement with theoretical predictions [20,21]. The status quo in molecular gases, demonstrating additional strong dependence of ionization on molecular orientation and internal ultrafast dynamics [22,23], is even worse and, despite extensive theoretical efforts to extend atomic strong field ionization models to molecules [24], simulations and experiments currently rely on empirical dependencies retrieved for a limited range of intensities and for particular laser wavelengths [25–27].

Measurements of absolute electron numbers generated in gases by intense femtosecond laser pulses and corresponding photoionization rates are very challenging. For relatively high plasma densities, $n_e \geq 10^{16} \text{ cm}^{-3}$ methods of optical interferometry or shadowgraphy can be employed [28,29]. However, the ionization rate is predicted to drop with increasing laser wavelength [5], and methods for electron density measurements in mid-IR filaments below the limit of $n_e \leq 10^{16} \text{ cm}^{-3}$ are required. A number of semiempirical and/or indirect methods was proposed, including ion current measurement by time-of-flight mass spectrometer, scattering of THz radiation, measurements of capacitive response times, picosecond

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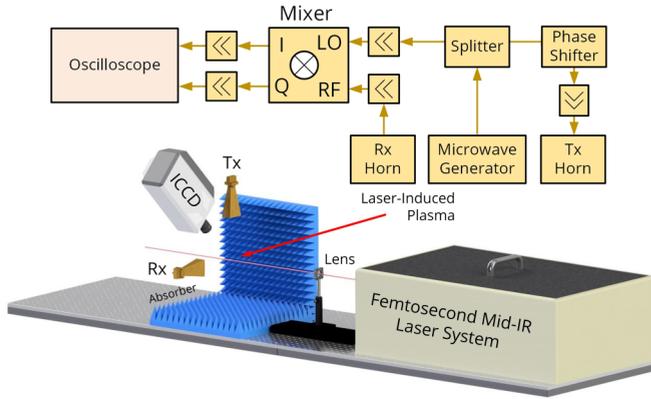


FIG. 1. Schematics of the experimental setup containing the 3.9- μm femtosecond laser and coherent microwave scattering system.

avalanche electron multiplication, and attenuation of the TE_{10} mode in a rectangular waveguide; however, these methods yield relative and/or indirect measurements associated with ambiguous interpretation [9,25,28–33]. Direct counting of photoionized electrons based on picosecond avalanche amplification of individual electrons was reported recently and the method is well suited for very low ionization degrees $\leq 10^{-10}$ [34].

Recently, coherent microwave scattering (CMS) was demonstrated as an effective diagnostic technique for the measurement of photoionization rates and temporally resolved electron number densities at laser-induced photoionization in UV and near-IR spectral ranges [35–37]. State-of-the-art CMS systems are associated with high sensitivity and a minimal detectable number of electrons in microplasma volume $\sim 10^8$. In this work, we expand CMS application to measure the total electron count and electron number density produced by focusing a 3.9- μm femtosecond laser pulse in air and subsequently determine tunneling ionization rates of ambient air in the mid-IR spectral band.

II. EXPERIMENTAL DETAILS AND METHODOLOGY

The experiments were conducted at the Ultrafast Laser Group of the Photonics Institute at TU Wien (Austria) with the unique 3.9- μm 30-mJ mid-IR femtosecond optical parametric chirped pulse amplifiers (OPCPAs) [7,38,39]. The coherent microwave scattering (CMS) measurement system was provided by Purdue University (USA) to conduct these experiments. The experimental setup including the femtosecond midinfrared laser and CMS system used in this work is schematically shown in Fig. 1.

Measurements of the total electron count in the laser-induced plasma volume (N_e) and electron number density (n_e) were conducted using a homodyne-type CMS system operating at 11 GHz. Incident microwave radiation (~ 8 W, Tx horn) was linearly polarized along the plasma channel orientation, and the signal scattered off the plasma volume was received by a microwave horn (Rx horn), amplified, and detected using an I/Q mixer. All components of the microwave system operated in the linear regime to ensure that the measured response is proportional to the amplitude of the scattered signal. The temporal resolution of the CMS

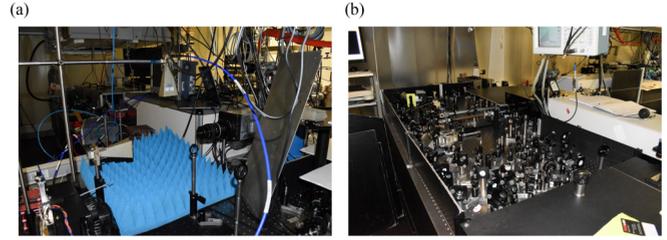


FIG. 2. (a) Photograph of CMS system aligned with the beam waist location. (b) Photograph of the 3.9- μm 30-mJ femtosecond laser system.

system was about 1 ns, limited by a 1-GHz bandwidth LeCroy WaveRunner 104Xi oscilloscope used in the circuit. The CMS system was calibrated using Teflon dielectric scatterers and attribution of the measured scattering signal U_S to N_e was done via the following equation:

$$U_S = \begin{cases} A \frac{e^2}{m v_m} N_e & \text{– laser – induced plasma} \\ A V_D \epsilon_0 (\epsilon_D - 1) \omega & \text{– dielectric scatterer} \end{cases}, \quad (1)$$

where A is the calibration coefficient of the CMS system (evaluated from the lower equation), V_D and ϵ_D are the volume and the dielectric constant of the Teflon dielectric scatterer, ϵ_0 is the dielectric permittivity of vacuum, v_m is the electron-gas collisional frequency, and ω is the circular frequency of the microwave radiation. The laser-induced plasma was imaged using an intensified charge-coupled device (ICCD) camera to observe its evolution with laser pulse energy and determine plasma volume V_p . The spatially averaged electron number density was determined as $n_e = \frac{N_e}{V_p}$. More details on the CMS technique can be found elsewhere [35,36,40].

III. RESULTS AND DISCUSSION

In the experiments, linearly polarized 3.9- μm pulses were focused in air by a 150-mm focal length CaF₂ lens (see Fig. 2). Femtosecond laser pulses were characterized by the second-harmonic generation frequency resolved optical-gating (SHG FROG) method, the beam waist was measured by a knife-edge technique (conducted with a strongly attenuated laser beam), and spatial distribution along the propagation was monitored with a Spiricon Pyrocam III beam profiler. The results of these measurements are shown in Fig. 3. SHG FROG measurement reveals spectral full width at half maximum (FWHM) of 302.9 nm and temporal FWHM of $\tau_{\text{FWHM}} = 117.7$ fs. Before focusing, the 3.9- μm beam was slightly elliptical with the beam diameters in the x and y directions at FWHM level being 3.98 and 4.64 mm, respectively. The beam waist was evaluated to be $w_0 = 95.85$ μm . These quantities can be regarded as invariant for the experimental conditions encountered in this work.

Representative line-of-sight ICCD images (gate width 100 ns) of laser-induced plasmas in ambient air and spatial beam profiling acquired 15 cm behind the focal plane are depicted in Fig. 4 for various laser pulse energies. As observed, and consistent with characteristics of the laser and focusing arrangement, encountered plasma channels are rather short, on the order of a millimeter. One can conclude that beam

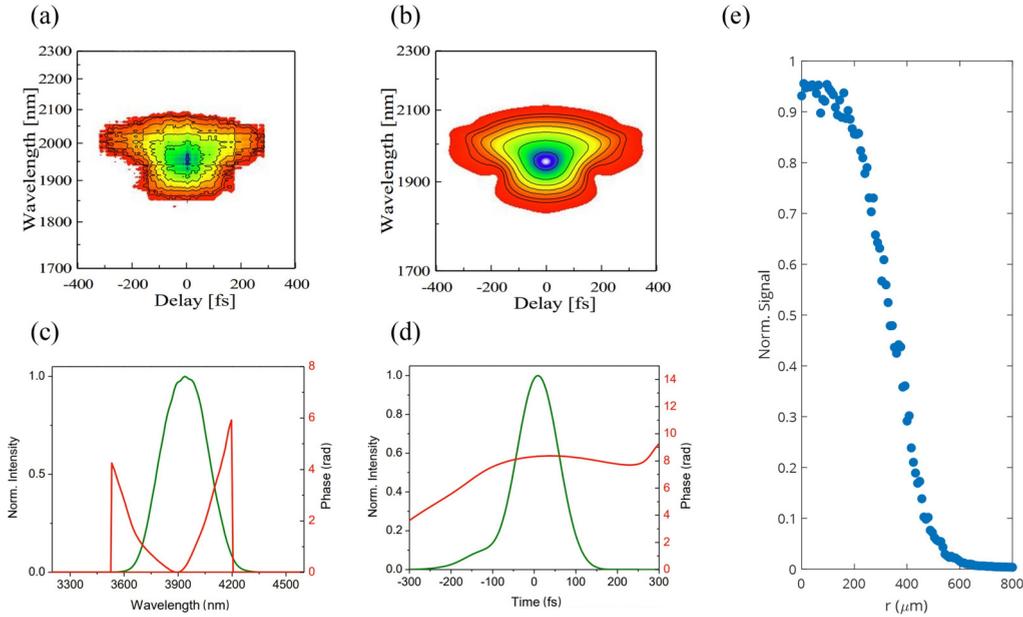


FIG. 3. Characteristics of the femtosecond pulse for $E_L = 0.2$ mJ. (a) Measured SHG FROG. (b) Retrieved SHG FROG. (c) Spectral composition and phase. (d) Temporal profile and phase. (e) Knife-edge beam waist measurements for $f = 150$ mm.

propagation for the tested irradiating energies E_L up to ~ 3.5 mJ can be regarded as nearly linear. This is justified by (1) relative invariance of the laser beam profile and (2) symmetry (longitudinal and transversal) of plasma geometric features up to $E_L \sim 2$ mJ as well as by a fairly weak shift of the focal point in the focusing lens direction. Slight onset of the nonlinear optical effects is encountered at the upper edge of tested energy range $E_L \sim 2\text{--}3.5$ mJ. Note that Kerr focus-

ing and plasma defocusing are expected to have comparable magnitude of contributions to the refractive index (n) for the range of tested laser intensities (I_L) and electron number (n_e) densities measured below; namely, $|\Delta n| \approx 5 \times 10^{-5}$ for $I_L = 10^{18}$ W/m² and $n_e = 10^{16}$ cm⁻³. Additionally, the plasma channel can be regarded as an oblate spheroid with semi-axis estimates of $a = b = 100$ μm (consistent with knife-edge measurements) and $c = 0.75$ mm. These dimensions suggest

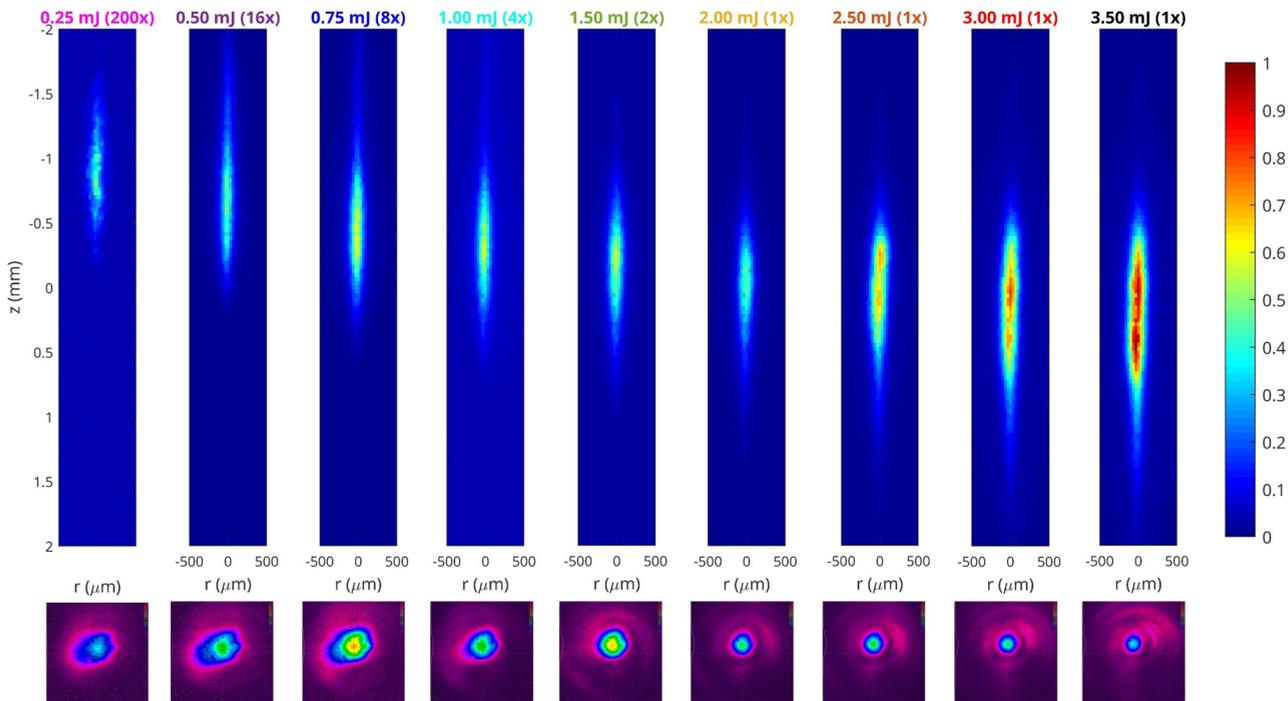


FIG. 4. ICCD images (exposure time 100 ns) and spatial beam profiling (acquired 15 cm behind the focal plane) associated with femtosecond midinfrared photoionization in atmospheric air for irradiating energies E_L up to 3.5 mJ and $f = 150$ mm lens.

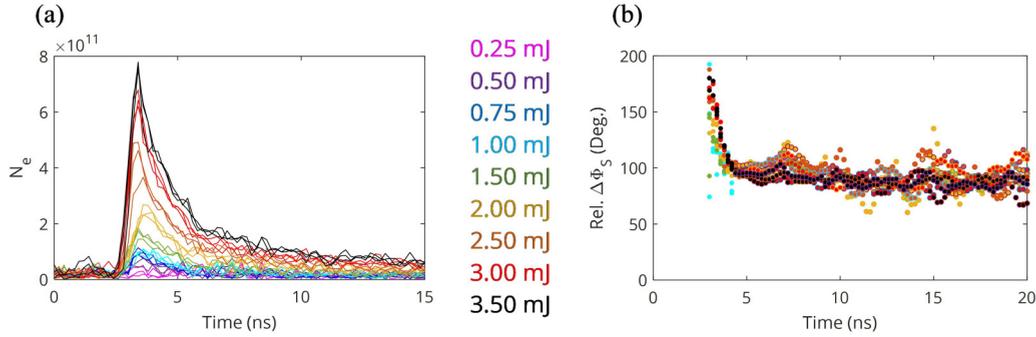


FIG. 5. (a) Temporal decay of N_e measured via CMS for various E_L . Line coloring corresponds to the headers in Fig. 4. (b) Associated relative phase wave forms.

plasma volume $V_p = 3.14 \times 10^{-11} \text{ m}^{-3}$ and somewhat large depolarization factors (governing magnitude of the microwave field screening inside the plasma spheroid due to accumulation of charges on the spheroid ends) on the order of $\xi \approx 0.03$ [35,36]. This, coupled with skin-depth considerations, largely restricts the measurable n_e up to about 10^{22} m^{-3} before a deviation from ideal collisional coherent microwave scattering behavior (and still below the threshold for significant electron-ion Coulomb contributions to ν_m). Note that longitudinal and transversal plasma boundaries determined using ICCD imaging were associated with intensities of about 10% and about 90% of the peak intensity, respectively (for the Gaussian beam with $w_0 = 95.85 \text{ }\mu\text{m}$ and Rayleigh length $z_R = 4.1 \text{ mm}$). (Even though this result is consistent with our previous work at near IR, it should be specifically considered and interpreted in the future) [36].

Temporal evolution of total electron numbers measured by the CMS is plotted in Fig. 5(a) for various E_L (line colors refer to the headers in Fig. 4). N_e was extracted from the measured scattered signal U_s under the assumption of a constant collisional frequency $\nu_m = 2 \times 10^{12} \text{ s}^{-1}$ corresponding to an electron temperature of $T_e \sim 1 \text{ eV}$. This is consistent with quick T_e collapse from the range corresponding to quiver energy ($E_{\sim} = e^2 I_L / 2c\epsilon_0 m_e \omega_L^2 \approx 10\text{--}100 \text{ eV}$) to an approximately single eV/sub-eV range on the time scale of $\ll 1 \text{ ns}$ followed by fairly slow evolution during several next ns [28]. Note that during this quick T_e collapse phase the electrons can produce postpulse ionization of air and, therefore, reported N_e values might be slightly overestimated (about factor of 2–3). Additionally, observed CMS measurements unrealistically reflect (underestimate) the initial growth stage of N_e temporal dynamics due to limited temporal resolution of the detecting system of about 1 ns (oscilloscope with 1 GHz bandwidth was used). The decay is governed by dissociative recombination ($X_2^+ + e \rightarrow X + X$, where X is N or O) and three-body attachment to oxygen ($e + O_2 + X_2 \rightarrow O_2^- + X_2$, where X is N or O) and is adequately resolved in time by the utilized CMS system.

Temporal evolution of the relative phase of detected microwave radiation measured by the CMS system is plotted in Fig. 5(b). One can see that, for all tested pulse energies, the phase quickly collapsed to a constant value and remained unchanged during the subsequent electron decay which is indicative of a collisional scattering regime. This validates the mathematical formulation used to evaluate N_e from

the measured scattered signal U_s [upper Eq. (1)]. Note that pulse energies $E_L > 2 \text{ mJ}$ might indicate a mixed collisional-Rayleigh scattering regime during the first nanosecond after the laser pulse.

The spatially averaged electron number density (n_e) and photoionization rate (ν_{PI}) are plotted as a function of laser intensity in Fig. 6. n_e was determined as $n_e = \frac{N_e}{V_p}$ and ν_{PI} was subsequently calculated from the simple relation $n_e = n_g \nu_{PI} \tau_{FWHM}$, where n_g is the air number density ($n_g = 2.5 \times 10^{25} \text{ m}^{-3}$). Laser intensities indicated on the horizontal axis were calculated assuming Gaussian beam intensity distribution in spatial and temporal domains in vicinity of the waist using $I_L = \frac{E_L}{\frac{1}{2}\pi w_0^2 \tau_{FWHM} \sqrt{\pi}}$. Note that assumption of Gaussian intensity distribution is based on experimentally validated linear beam propagation with weak contribution of nonlinear optical effects as detailed above. It was observed that the total electron count N_e scaled with laser energy E_L as $N_e \propto E_L^{(1.5\text{--}1.7)}$; however, determination of effective plasma volume based on this scaling law (developed and utilized in our previous works) [36,37] was not pursued in this work as it was beyond the accuracy of the current dataset associated with limited size and substantial measurement errors.

In summary, photoionization rates at $3.9 \text{ }\mu\text{m}$ wavelength were experimentally determined for the range of radiation intensities $1.3 \times 10^{13}\text{--}1.9 \times 10^{14} \text{ W/cm}^2$ using a $3.9\text{-}\mu\text{m}$ femtosecond laser and absolutely calibrated coherent microwave scattering technique. Several important modifications

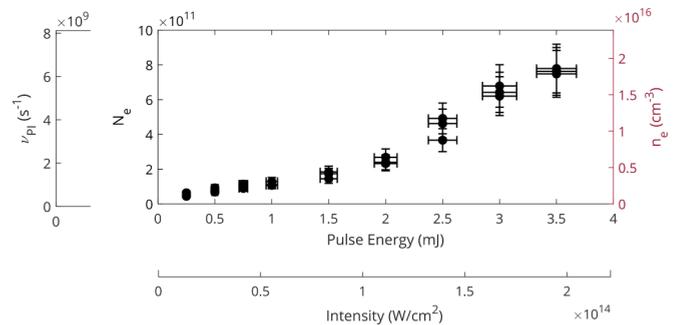


FIG. 6. Experimentally measured total electron numbers, electron number densities, and photoionization rates as a function of pulse energy or peak intensity.

can be considered in the future to improve the accuracy of ν_{PI} determination and extend the intensity range. First, a more rigorous procedure could identify effective plasma volume via deriving exact power-law scaling of N_e with E_L , extrapolating the same scaling locally on n_e vs I_L dependence, and conducting corresponding integrations similar to the methodology developed in our recent works [36,37]. This would further introduce a correction to the effective plasma volume V_p and, correspondingly, to the photoionization rate ν_{PI} . Second, experiments in pure gases should be conducted to identify individual contributions of various gaseous species contained in the atmospheric air (O_2 , N_2 , etc.) to the overall photoionization rate. Note that in the current work we calculated ν_{PI} based on the total air density $n_g = 2.5 \times 10^{25} \text{ m}^{-3}$ so that higher gas-specific photoionization rates (e.g., ν_{O_2} , ν_{N_2}) should be expected ($\nu_{PI} = \nu_{O_2}\chi_{O_2} + \nu_{N_2}\chi_{N_2}$, where χ_{O_2} and χ_{N_2} are mole fractions of oxygen and nitrogen in air). The photoionization rates measured in this work coincide with the lower bound of the corresponding range predicted by various theoretical models developed in the past (10^9 – 10^{13} s^{-1} for the radiation intensity $\sim 10^{14} \text{ W/cm}^2$ with intensity scaling varying in the range $I_L^{(1-5)}$) [25]. Note that we compare the photoionization rates measured in this work at $3.9 \mu\text{m}$ with that reported previously at 800 nm as similar rates in the tunneling regime are expected (largely governed by the amplitude of electric field rather than by the photon energy or wavelength determining the multiphoton regime). Third, conducting experiments in the collisionless ν_m -independent Thomson scattering regime at low pressures ($p \leq 1 \text{ Torr}$) would allow us to substantially extend the range of intensities

at which ν_{PI} can be evaluated by delaying the onset of nonlinear optical phenomena to higher laser intensities [35]. This refers to the fact that nonlinear optical phenomena including Kerr and plasma nonlinear terms in the refractive index reduce proportionally with a decrease in pressure ($n_2 \propto n_g \propto p$, $\omega_p^2 \propto n_e \propto n_g \propto p$). Fourth, expanding bandwidth of the CMS measurement system is recommended to accurately capture temporal dynamics of N_e and fully resolve the N_e peak value.

IV. CONCLUSIONS

In this work, total electron count, electron number densities, and photoionization rates were reported for midinfrared femtosecond tunneling photoionization at $3.9 \mu\text{m}$ in atmospheric air using the absolutely calibrated coherent microwave scattering technique. Utilization of atmospheric pressure limits the highest intensity at which a photoionization rate can be evaluated due to the onset of Kerr focusing. To extend this limit to higher intensities, the experiments at low pressures ($p \leq 1 \text{ Torr}$) in the collisionless Thomson scattering regime should be conducted.

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