

Direct Combinational Measurements of the Electron Density and Electric Field in Secondary Streamer Discharge under Atmospheric-Pressure Air

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The electron density and electric field govern physical and chemical reactions in a secondary streamer discharge under atmospheric-pressure air. We present direct combinational measurements of these essential quantities and demonstrate their consistency by solving the continuity equation for electrons. The measurements enabled validity examination of previous computational studies and clarified contradictions; the prominence was a near-cathode phenomenon measured during primary-to-secondary transition, which caused unpredicted ionization inside the positive secondary discharge. The thorough comparison revealed that a fundamental governing mechanism to be established for the secondary discharge is current continuity.

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In nature, a streamer discharge in air exists as a sprite above thunderclouds [1,2] and a precursor of lightning [3]. An air streamer discharge is also ubiquitous in practical applications such as plasma medicine [4], plasma agriculture [5], environmental pollution control [6], and energy efficiency improvements [7]. Consequently, the air streamer discharge has been intensely studied for decades.

Fundamental studies have investigated a 10-mm-scale pin-to-plane gap exposed to a sufficiently high, positive voltage applied to the pin anode under atmospheric-pressure air. Typically, an air streamer discharge develops in two phases [8]. First, a thin, luminous layer initially propagates from the pin anode to the plane cathode, which is called a primary streamer discharge or an ionizing wave of the potential gradient [9]. Next, a new bright zone

develops about halfway to the cathode once the primary discharge reaches the cathode. This is known as the secondary streamer discharge.

A systematic understanding and controlling of an air streamer discharge require direct combinational measurements of an electron density N_e [cm^{-3}] and reduced electric field E/N [Td] because these two quantities govern physical and chemical reactions occurring in the streamer discharge [10]. However, direct combinational measurements of N_e and E/N have been prevented for decades [11], due to two main reasons. First, an air streamer discharge has an irreproducible, complex branching structure [9]. Second, the candidates of applicable techniques are insufficiently resolved spatially [12], complex [13], or indirect based on modeled electron kinetics [14]. Therefore, these essential quantities could only be predicted through physical modeling [15] and numerical simulations [16]. Furthermore, investigations have mainly been focusing on the primary phase in specific contexts (e.g., morphological pattern analysis [17], branching factor identification [18], and streamer channel interaction [19]). However, investigations on the secondary phase are quite limited.

Limited numbers of studies on models [20,21] and numerical simulations [22,23] have predicted that the inside of a secondary streamer discharge should show a

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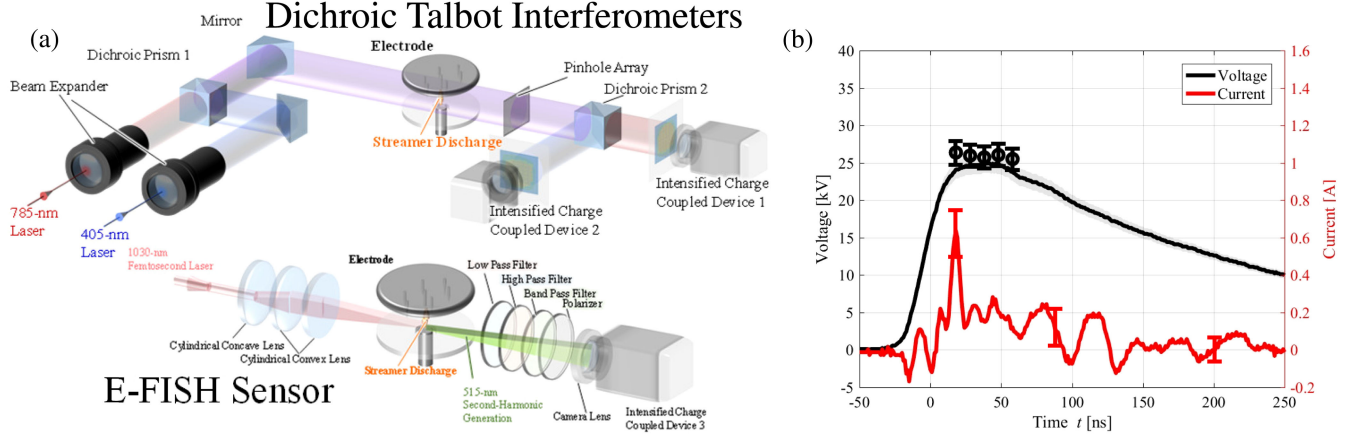


FIG. 1. (a) Optical configuration of dichroic Talbot interferometers for 2D N_e measurement and E-FISH sensor for 1D E/N measurement. (b) Voltage and current waveforms, where superimposed plots demonstrate $V_S = \int_{0 \text{ mm}}^{13 \text{ mm}} E_y(y, r = 0) dy + 1.5 \text{ kV}$ during secondary phase and accompanying error bars represent measurement accuracy of E-FISH sensor. Shaded area for voltage waveform and error bars for current waveform show shot-to-shot variations for ten different discharges.

lower N_e and a higher E/N uniformly distributed along the propagation direction. Additionally, they have predicted the opposite distributions along the propagation direction outside the secondary discharge (Supplemental Material (SM) [24], Fig. S1). However, direct measurements have yet to validate previous modeling and simulation studies. This critical missing of the validity examination undermines confidence in these computational predictions, which strongly motivates a decisive experimental benchmark of the essential quantities. Such a reliable, experimental anchor of N_e and E/N reveals the contradictions between experimental and computational studies. Reasonable explanation for the fundamental contradictions refines computational studies, makes them genuinely predictive, and accelerates advances in both science and applications.

In this Letter, we report the first, direct combinational measurements of N_e and E/N for the secondary phase and demonstrate that a higher N_e and lower E/N do exist inside the secondary streamer discharge. Such unpredicted structures of N_e and E/N were caused by a not so large E/N difference at the secondary streamer initiation between the present experiment and the previous computational studies. This moderate difference originated from a near-cathode phenomenon measured during the primary-to-secondary transition.

The direct combinational measurements were achieved by the application of three original instruments: a “dice-five” electrode configuration reproducibly achieving a nonbranching, straight single-filament streamer discharge [25] (SM [24], Fig. S2), dichroic Talbot interferometers visualizing the 2D N_e distribution from a single-shot recording [26] [Fig. 1(a)], and an electric-field induced second harmonic generation (E-FISH) sensor providing a 1D profile of the E/N vector with one-order-superior spatial resolution [30] [Fig. 1(a)]. These two diagnostic technologies are sufficiently resolved spatially and simple

(SM [24], Secs. 2 and 3). Furthermore, they do not require modeled electron kinetics. Therefore, direct combinational measurements of N_e and E/N are possible.

This Letter aims to directly measure the streamer discharge generated below the center needle (SM [24], Fig. S2). The influence of the surrounding discharges and their associated space charges on the central secondary streamer structure and dynamics can be assumed to be only slight (SM [24], Sec. 7.1). The vertical distance from the center needle tip is denoted by y [m], where the y axis corresponds to the center geometrical axis (SM [24], Fig. S2). The radial distance from the y axis is described by r [m]. The gap length between the center needle tip and the plane cathode was 13 mm. The electrode system was installed in the 1-bar dry air. The streamer discharges were generated with a repetition rate of 0.3 Hz; thus, they were single-pulse streamer discharges, not the repetitive ones generated at 1–100 kHz [53].

Figure 1(b) shows the applied voltage to the anode, where the voltage peak and rise rate were 25 kV and 0.83 kV/ns, respectively. The current waveform was obtained by subtracting the displacement current [25]. The initiation time of the primary streamer discharge was defined as $t = 0$ ns, which was calibrated using a previously established technique [25,55]. The primary streamer discharge arrived at the cathode surface at $t = 13$ ns. After the transition phase between the primary and secondary discharges in $13 \leq t \leq 16$ ns [30], a stable secondary streamer discharge was established in $t > 16$ ns [25].

The Talbot sensors (SM [24], Sec. 2) measured the 2D N_e distributions from single-shot recordings at the primary streamer contact with the cathode ($t = 13$ ns) and during the secondary phase ($t = 18, 28, 38$ ns). The 1D E-FISH method (SM [24], Sec. 3) measured the radial distributions of the reduced electric field components, E_y/N and E_r/N , at $y = 1, 2, 3, \dots, 11, 12$ mm during the transition phase

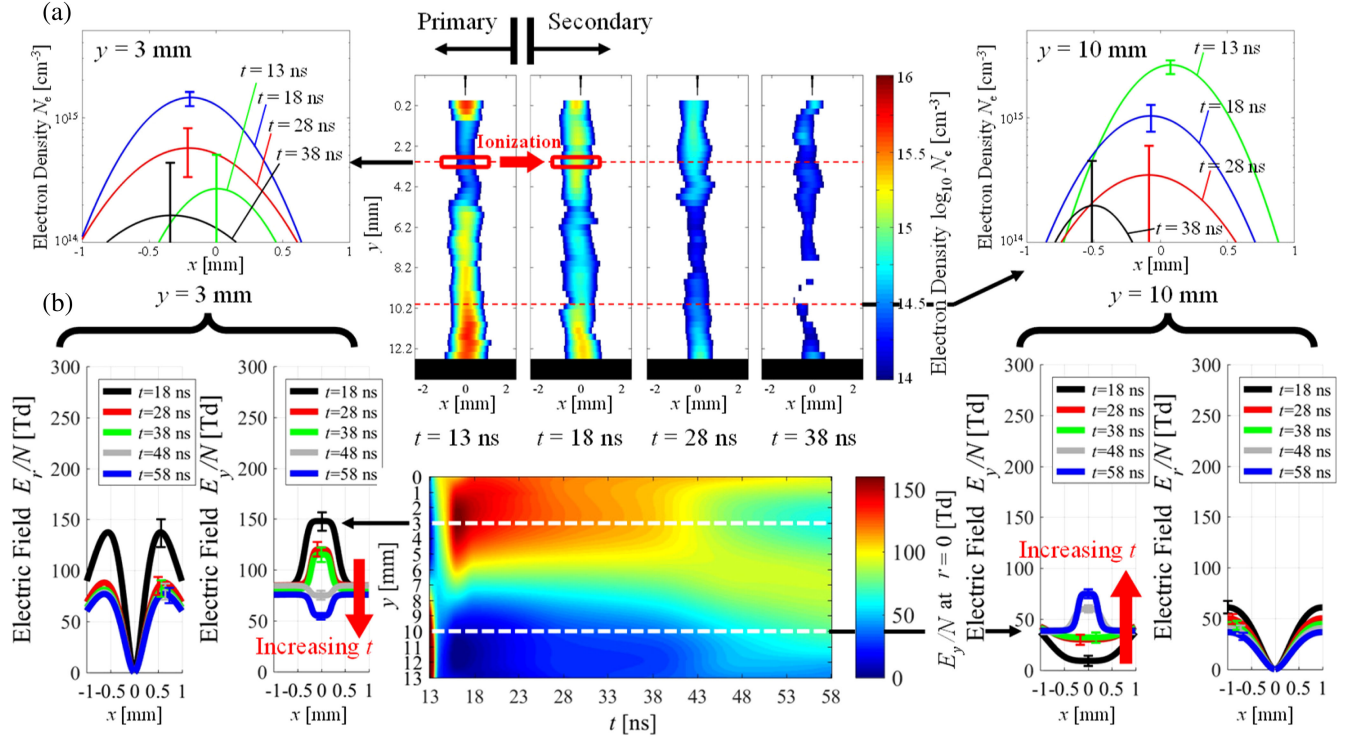


FIG. 2. (a) Spatiotemporal evolution of 2D N_e distribution. Error bars of 1D N_e distributions on $y = 3$ and 10 mm demonstrate error ranges (SM [24], Sec. 2.3.2). (b) Temporal evolution of 1D E/N distributions on $y = 3$ and 10 mm along with streak image of $E_y(y, r = 0)/N$. Error bars of 1D E/N distributions on $y = 3$ and 10 mm represent measurement accuracy (SM [24], Sec. 3.2.1).

($t = 13, 14, 15, 16$ ns [30]) and secondary phase ($t = 18, 28, 38, 48, 58$ ns).

Figure 2(a) shows the spatiotemporal evolution of the 2D N_e distributions. Figure 2(b) depicts the temporal evolution of the 1D E/N distributions on $y = 3$ and 10 mm as well as the streak image of $E_y(y, r = 0)/N$. A polynomial fitting in the y direction and a shape-preserving piecewise cubic interpolation in the t direction were used to draw the streak image. An integrated $E_y(y, r = 0)$ over the gap length [$\int_0^{13 \text{ mm}} E_y(y, r = 0) dy$] can be compared with the applied voltage to the anode. Although the secondary streamer discharge includes strong E_y/N in a very thin cathode fall region, the E_y/N was not measured in the present study due to the mm-scale discrete sensing positions along the y direction. Assuming that 3000 Td existed in the cathode fall region with a thickness of 20 μm [56], 1.5 kV was added to the integrated $E_y(y, r = 0)$. Figure 1(b) plots $V_S = \int_0^{13 \text{ mm}} E_y(y, r = 0) dy + 1.5 \text{ kV}$ on the voltage waveform. The V_S and the shaded applied voltage are within the measurement accuracy of the present E-FISH method. Thus, the measured E_y/N in Fig. 2(b) is valid.

The validity of the measured N_e was examined by consistency analysis between the measured N_e and E/N . Specifically, the continuity equation for electrons [20] was solved on the y axis of a cylindrical coordinate system as

$$\begin{aligned} \frac{\partial N_e}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \{r V_r(E_r/N) N_e\} + \frac{\partial}{\partial y} \{V_y(E_y/N) N_e\} \\ = -\nu_{\text{att}}(E/N) N_e, \end{aligned} \quad (1)$$

where $\nu_{\text{att}}[\text{s}^{-1}]$ is the net electron-attachment rate, which includes the ionization rate, and $V_r[\text{m/s}]$ and $V_y[\text{m/s}]$ are electron drift speed in r and y directions, respectively. Electron diffusion was neglected because its coefficient is $\sim 0.1 \text{ m}^2/\text{s}$ [57,58] and has a negligible contribution to N_e decay on the 10-ns time scale [59]. The ν_{att} , V_r , and V_y are functions of E/N . By substituting the spatiotemporal evolution of the measured E_y/N and E_r/N into Eq. (1), the temporal variation of N_e was calculated.

Figure 3 shows the temporal variations of the calculated N_e (dashed lines) and measured N_e (circle plots) on $y = 3$ and 10 mm. Equation (1) was solved using the typical electric field dependence of V_r , V_y [41], and ν_{att} [26] (inset of Fig. 3). The analyzed N_e at $y = 3$ mm increases in $t \leq 18$ ns, where the measured E/N is beyond the critical ionization electric field $E/N_{\text{cri}} (\approx 120\text{--}135 \text{ Td})$ and the effective ionization coefficient is positive [57,58]. Conversely, it decreases in $t \gtrsim 28$ ns, where the measured E/N is comparable to or below E/N_{cri} and the effective ionization coefficient is approximately zero or negative [57,58]. At $y = 10$ mm, the analyzed N_e decreases

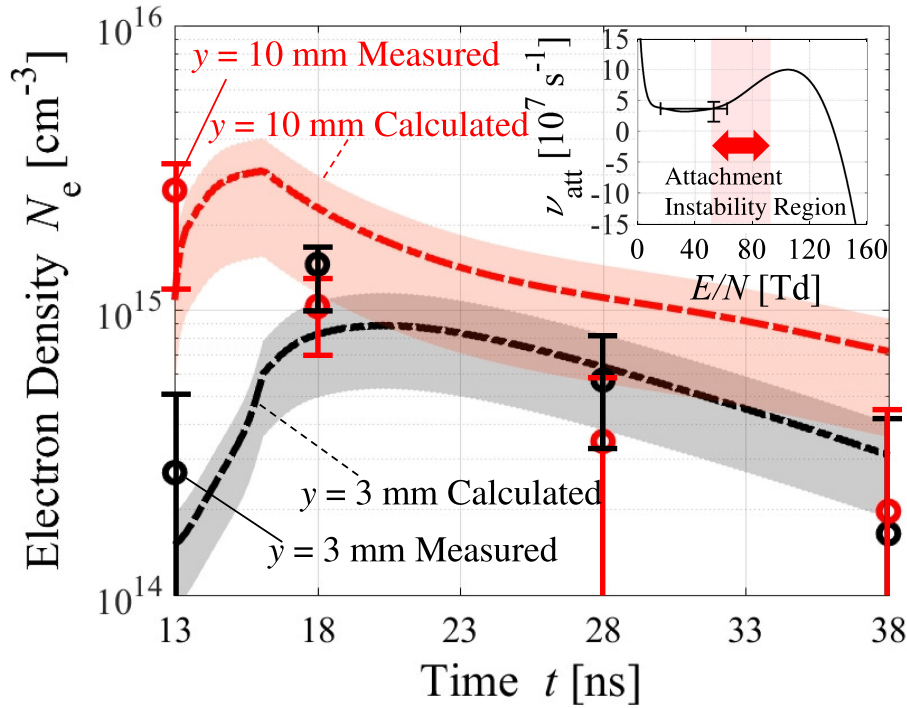


FIG. 3. Temporal evolution of calculated N_e (dashed lines) and measured N_e (circle plots, average of five different discharges) on $y = 3$ and 10 mm. Calculated and measured N_e indicate those at radial centers. Inset shows typical electric field dependence of ν_{att} [26]. Shaded areas for dashed lines are drawn based on measurement accuracy of E/N (SM [24], Sec. 3.2.1) and ν_{att} , and error bars for circle plots demonstrate error ranges (SM [24], Sec. 2.3.2) or shot-to-shot variations for five different discharges, whichever is larger.

monotonously in $t \geq 18$ ns because the measured E/N is below E/N_{cri} . These temporal variations of the analyzed N_e explain well those of the measured N_e within the error bars and shaded areas (Fig. 3). Hence, the measured N_e and E/N are consistent with each other.

Next, validity of the previous studies on the modeling [20] and numerical simulations [22,23] is examined in comparison with the measured N_e and E/N . As shown in Fig. 2(a), the measured N_e at $y = 3$ mm increased from $t = 13$ to 18 ns, which means that ionization occurred inside the initial secondary discharge (SM [24], Secs. 4 and 5). As a result of the ionization, the N_e at $t = 18$ ns was higher for the inside of the secondary discharge (i.e., $y = 3$ mm) than the outside (i.e., $y = 10$ mm). Such ionization and the resulting higher N_e inside the secondary discharge contradict the previous studies involving the model [20] and numerical simulations [22,23]. The experimentally observed ionization was caused by the fact that the measured E/N at $y = 3$ mm reached $E/N = 150$ Td at $t = 16$ and 18 ns, which exceeded E/N_{cri} .

Particularly in the simulation studies for pulsed voltage application comparable with this research, E/N inside the secondary streamer discharge is $E/N \approx 100 - 120$ Td, which is below E/N_{cri} [22,23]. Consequently, the simulated N_e decreases as time increases from the primary to secondary phases [22,23]. The relative difference of the simulated E/N with respect to the measured one is

$\approx 20\% - 30\%$ at most. Considering the measurement accuracy of the present E-FISH method (SM [24], Sec. 3.2.1) and model-to-model variations of numerical simulations [60], the relative difference may seem acceptable. However, this contradiction has non-negligible consequences because the presence or absence of ionization significantly impacts the generation processes of the reactive species and their energy allocation.

In addition, the transition phase preceding the secondary phase demonstrates three major contradictions: (i) At the primary streamer contact with the cathode ($t = 13$ ns), the measured N_e distribution along the y axis was nonuniform [Fig. 2(a)]. In contrast, the numerically simulated one is almost uniform [22,23]. (ii) At $t = 13$ ns, the measured length of the high electric field region with $E/N \geq 150$ Td was 4–5 mm [$8 \lesssim y \lesssim 13$ mm, Fig. 2(b)]. However, it is within 1 mm in the numerical simulations [22,23]. (iii) The temporal variation of the measured E/N was considerably faster than that expected from the numerical simulation [23].

In particular, contradiction (ii) could decide the difference in the secondary phase (SM [24], Sec. 4). The measured high electric field region was ionized at $t = 13$ ns and the resulting high electron density of 10^{15} cm⁻³ was sustained until $t = 18$ ns [Fig. 2(a)]. Since such a region above the cathode was electrically conductive, the gap length for the present experiment was effectively shorter than the numerical simulation by $\approx 20\% - 30\%$ during $13 < t \leq 18$ ns [23].

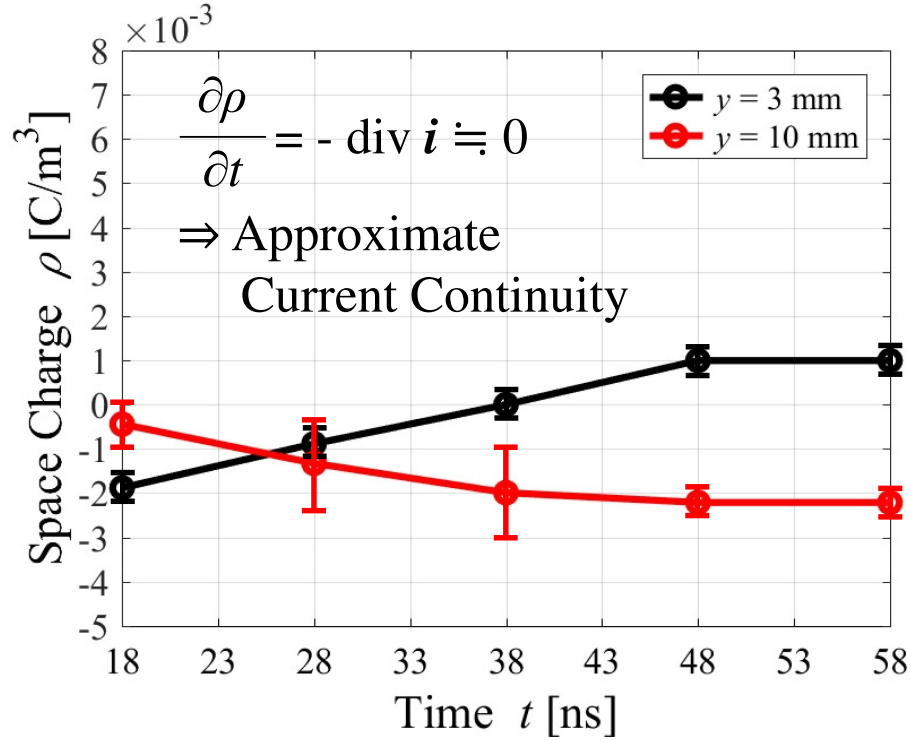


FIG. 4. Temporal evolution of space charge ρ on y axis. Error bars are drawn based on measurement accuracy of E_y/N and E_r/N (SM [24], Sec. 3.2.1).

Therefore, during $13 < t \leq 18$ ns in the present experiment, an electric field imposed on the other region, e.g., inside the secondary discharge ($y = 3$ mm), was higher than the previous numerical simulation by $\approx 20\% - 30\%$ (Fig. 5), which agreed well with the aforementioned relative difference. Consequently, the electron density increase due to ionization in the early secondary phase could occur for the experiment, but not for the numerical simulation.

Hence, simulation studies may be modified so as to resolve the contradictions in the preceding transition phase especially near the cathode and subsequent secondary phase. This Letter shows the ability of combinational N_e and E/N measurements to reveal contradictions between the experiments and numerical simulations and to demonstrate a guide to improve simulation studies in the future.

One theory that proposes the formation model of the secondary streamer discharge is attachment instability [20]. This theory predicts that E/N inside the secondary discharge (e.g., $y = 3$ mm) remains higher than the upper end of the attachment instability region ($50 \lesssim E/N \lesssim 90$ Td), while E/N outside (e.g., $y = 10$ mm) stays lower than the lower end. Furthermore, it also predicts that ionization does not occur inside the secondary discharge. Previous numerical simulation reproduced these predictions [23]. However, as demonstrated in Fig. 2, ionization did occur in the measured transition phase ($t = 16$ ns) and initial secondary phase ($t = 18$ ns). Consequently, the spatiotemporal

evolution of the measured E/N in $t > 18$ ns would differ from the prediction. In actuality, Fig. 2(b) shows that, although the measured E/N at $y = 3$ and 10 mm were already higher and lower than the attachment instability region at $t = 18$ ns, respectively, they did not stay outside the attachment instability region. Instead, they started invading the attachment instability region after $t = 38$ ns. In the attachment instability region, the measured E/N at $y = 3$ mm decreased, while that at $y = 10$ mm increased. Finally, at $t = 58$ ns, the measured E/N at $y = 3$ mm was comparable or lower than that at $y = 10$ mm. Such a comparable or lower electric field inside the secondary streamer discharge also contradicts the previous theory [20] and numerical simulation [23].

To identify the mechanism driving the unpredicted spatiotemporal evolution of the measured E/N in $t > 18$ ns, $\text{div } \mathbf{i}$ was calculated on the y axis by substituting the measured E_y/N and E_r/N into the following equation:

$$\rho = \epsilon_0 \text{div } \mathbf{E} = \epsilon_0 \left\{ \frac{1}{r} \frac{\partial}{\partial r} (r E_r) + \frac{\partial E_y}{\partial y} \right\}, \quad (2)$$

$$\text{div } \mathbf{i} = -\frac{\partial \rho}{\partial t}, \quad (3)$$

where $\rho [\text{Cm}^{-3}]$ is the space charge and $\epsilon_0 [\text{Fm}^{-1}]$ is the permittivity in vacuum. From the numerical evaluation of Eq. (3), the temporal evolution of ρ was calculated (Fig. 4).

The absolute values of ρ were $|\rho| \sim 10^{-3} \text{ Cm}^{-3}$ at $y = 3$ and 10 mm, which indicated that the space-charge number density ρ_e was $\rho_e = (|\rho|/e) \sim (10^{-3} \text{ Cm}^{-3}/1.6 \times 10^{-19} \text{ C}) = 10^{10} \text{ cm}^{-3}$. Considering that the electron density in the secondary discharge was as high as $N_e = 10^{14-15} \text{ cm}^{-3}$, the space-charge ratio with respect to the electron density, ρ_e/N_e , was only slight ($\rho_e/N_e = 0.001\% - 0.01\%$). The temporal evolution of ρ can be caused by factors such as electron production and consumption processes (SM [24], Sec. 5), nonuniform distributions of N_e , V_r , and V_y (Fig. 2), and negative charge accumulation compensating the positive charge developed in the primary phase [55]. The ρ at $y = 3$ (10 mm) initially increased (decreased) with time, but became nearly stable with time, demonstrating that these factors became almost inactive and/or balanced.

From Eq. (3), this temporally nearly stable ρ means that the approximate current continuity, $\text{div } \mathbf{i} \doteq 0$, held. Hence, E/N for the ionized secondary discharge developed to gradually satisfy current continuity. Such a gradual satisfaction of the current continuity has also been reported in the numerical simulation [23] and modeling study [55] for the “nonionized” secondary discharge, indicating different spatiotemporal evolution of E/N and N_e from this Letter. Therefore, the current continuity is a fundamental governing mechanism to be established, regardless of whether ionization occurs or not inside the secondary streamer discharge.

In summary, the direct combinational measurements of electron densities and electric fields provided the first detailed examination of previous physical models and numerical simulations. The spatiotemporal evolution of the measured electron densities and electric fields completely differed from the predictions made by these previous computational studies. Such difference was attributed to a longer high electric field region with more than 150 Td measured above the cathode during the primary-to-secondary transition, which caused unpredicted ionization inside the initial secondary discharge. The next steps include coupling diagnostics with laser Thomson scattering to determine the electron energy distribution functions, examine Boltzmann equation schemes, and calibrate the electron-impact collisional cross sections. Other steps involve combinational measurement with (two-photon absorption) laser-induced fluorescence and/or laser absorption to quantify temporal evolution of radical number densities, assess the electron-impact reaction rates, and identify the radical production or consumption pathways. The present experimental results can serve as a benchmark for examining and refining the theoretical and numerical models. Resolving the contradictions between these modeling studies and the experimental benchmark will substantially advance fundamental understanding of physics and chemistry inside the streamer discharge. Consequently, this Letter will improve predictive capabilities of the models and controllability of the highly transient streamer discharge,

which is valuable for diverse plasma research fields ranging from lightning physics to plasma application technologies in medicine, agriculture, environment, and energy.

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Data availability—The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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- [1] R. C. Franz, R. J. Nemzek, and J. R. Winckler, *Science* **249**, 48 (1990).
 - [2] V. P. Pasko, M. A. Stanley, J. D. Mathews, U. S. Inan, and T. G. Wood, *Nature (London)* **416**, 152 (2002).
 - [3] V. A. Rakov and M. A. Uman, *Lightning: Physics and Effects* (Cambridge University Press, Cambridge, United Kingdom, 2003), Chap. 4.3.
 - [4] K. Mizuno, Y. Shirakawa, T. Sakamoto, H. Ishizaki, Y. Nishijima, and R. Ono, *IEEE Trans. Radiat. Plasma Med. Sci.* **2**, 353 (2019).
 - [5] C. Bradu, K. Kutasi, M. Magureanu, N. Puač, and S. Živković, *J. Phys. D* **53**, 223001 (2020).
 - [6] J. S. Chang, P. A. Lawless, and T. Yamamoto, *IEEE Trans. Plasma Sci.* **19**, 1152 (1991).
 - [7] S. M. Starikovskaia, *J. Phys. D* **39**, R265 (2006).
 - [8] L. B. Loeb, *Electrical Coronas* (Cambridge University Press, Cambridge, United Kingdom, 1965).
 - [9] L. B. Loeb, *Science* **148**, 1417 (1965).
 - [10] Y. P. Raizer, *Gas Discharge Physics* (Springer, Berlin, 1991).
 - [11] I. Adamovich *et al.*, *J. Phys. D* **50**, 323001 (2017).
 - [12] M. S. Simeni, B. M. Goldberg, C. Zhang, K. Frederickson, W. R. Lempert, and I. V. Adamovich, *J. Phys. D* **50**, 184002 (2017).
 - [13] K. Tomita, Y. Inada, A. Komuro, X. Zhang, K. Uchino, and R. Ono, *J. Phys. D* **53**, 08LT01 (2020).
 - [14] T. Hoder, M. Černák, J. Paillol, D. Loffhagen, and R. Brandenburg, *Phys. Rev. E* **86**, 055401(R) (2012).
 - [15] G. V. Naidis, *Phys. Rev. E* **79**, 057401 (2009).
 - [16] S. Pancheshnyi, M. Nudnova, and A. Starikovskii, *Phys. Rev. E* **71**, 016407 (2005).
 - [17] V. P. Pasko, U. S. Inan, and T. F. Bell, *Geophys. Res. Lett.* **27**, 497 (2000).
 - [18] A. Luque and U. Ebert, *Phys. Rev. E* **84**, 046411 (2011).
 - [19] A. Luque, U. Ebert, and W. Hundsdorfer, *Phys. Rev. Lett.* **101**, 075005 (2008).

- [20] R. S. Sigmond, *J. Appl. Phys.* **56**, 1355 (1984).
- [21] F. Bastien and E. Marode, *J. Phys. D* **18**, 377 (1985).
- [22] O. Eichwald, O. Ducasse, D. Dubois, A. Abahazem, N. Merbahi, M. Benhenni, and M. Yousfi, *J. Phys. D* **41**, 234002 (2008).
- [23] A. Komuro, K. Suzuki, K. Yoshida, and A. Ando, *Jpn. J. Appl. Phys.* **59**, SAAB03 (2020).
- [24] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/5sv6-28qp> for the supplemental figures of the secondary streamer structure, streamer generation setup, and axial distributions of electron densities and electric fields including Refs. [20,21,25], the measurement principles of the dichroic Talbot method including Refs. [23,26–29] and the 1D E-FISH method including Refs. [23,30–41], influences of the transition phase on the secondary phase including Refs. [22,23,42], short-life state-to-state chemistry including Refs. [43–47], missing physical processes in existing models including Refs. [20,21], and the electrode configuration including Refs. [43,44,48–54].
- [25] R. Ono and A. Komuro, *J. Phys. D* **53**, 035202 (2020).
- [26] Y. Inada, A. Komuro, R. Ono, A. Kumada, K. Hidaka, and M. Maeyama, *J. Phys. D* **52**, 185204 (2019).
- [27] R. H. Huddleston and S. L. Leonard, in *Plasma Diagnostic Techniques* (Academic Press, London, 1965), pp. 439–441.
- [28] P. E. Ciddor, *Appl. Opt.* **35**, 1566 (1996).
- [29] R. A. Alpher and D. R. White, *Phys. Fluids* **2**, 162 (1959).
- [30] Y. Inada, T. Shioda, R. Nakamura, M. Maeyama, A. Kumada, S. Nakamura, and R. Ono, *J. Phys. D* **55**, 385201 (2022).
- [31] A. Yariv and P. Yeh, in *Photonics: Optical Electronics in Modern Communications* (Oxford University Press, New York, 2007), 6th ed., Chap. Ch 2.11.
- [32] D. Oron and Y. Silberberg, *J. Opt. Soc. Am. B* **21**, 1964 (2004).
- [33] R. W. Boyd, in *Nonlinear Optics* (Academic, New York, 2020), 4th ed., Chap. Ch 2.10.
- [34] M. S. Simeni, Y. Tang, K. Frederickson, and I. V. Adamovich, *Plasma Sources Sci. Technol.* **27**, 104001 (2018).
- [35] N. D. Lepikhin, D. Luggenhölscher, and U. Czarnetzki, *J. Phys. D* **54**, 055201 (2021).
- [36] E. Marode, F. Bastien, and M. Bakker, *J. Appl. Phys.* **50**, 140 (1979).
- [37] S. Rostami, J. Diels, and L. Arissian, *Opt. Express* **23**, 3299 (2015).
- [38] S. Nakamura, T. Fujii, M. Sato, A. Kumada, and Y. Oishi, *Opt. Lett.* **49**, 726 (2024).
- [39] J. Kasparian, R. Sauerbrey, and S. L. Chin, *Appl. Phys. B* **71**, 877 (2000).
- [40] T. Darny, J. M. Pouvesle, V. Puech, C. Douat, S. Dozias, and E. Robert, *Plasma Sources Sci. Technol.* **26**, 045008 (2017).
- [41] J. Dutton, *J. Phys. Chem. Ref. Data* **4**, 577 (1975).
- [42] A. Komuro, K. Takahashi, and A. Ando, *J. Phys. D* **48**, 215203 (2015).
- [43] X. Chen, Y. Zhu, and Y. Wu, *Plasma Sources Sci. Technol.* **29**, 095006 (2020).
- [44] B. Zhang, Y. Zhu, X. Zhang, N. Popov, T. Orriere, D. Z. Pai, and S. M. Starikovskaia, *Plasma Sources Sci. Technol.* **32**, 115014 (2023).
- [45] A. Komuro and R. Ono, *IEEE Trans. Plasma Sci.* **42**, 2406 (2014).
- [46] A. Komuro, R. Ono, and T. Oda, *J. Phys. D* **45**, 265201 (2012).
- [47] S. Pancheshnyi, *J. Phys. D* **46**, 155201 (2013).
- [48] Z. Wei, R. Ono, and A. Komuro, *Plasma Sources Sci. Technol.* **33**, 075001 (2024).
- [49] Y. Kashiwagi and H. Ito, *J. Phys. D* **39**, 113 (2006).
- [50] Y. Li, E. M. van Veldhuizen, G. J. Zhang, U. Ebert, and S. Nijdam, *Plasma Sources Sci. Technol.* **27**, 125003 (2018).
- [51] M. Janda, V. Martišovits, and Z. Machala, *Plasma Sources Sci. Technol.* **20**, 035015 (2011).
- [52] R. Ono and T. Oda, *J. Phys. D* **36**, 1952 (2003).
- [53] D. Z. Pai, D. A. Lacoste, and C. O. Laux, *Plasma Sources Sci. Technol.* **19**, 065015 (2010).
- [54] F. Tholin and A. Bourdon, *J. Phys. D* **46**, 365205 (2013).
- [55] E. Marode, *J. Appl. Phys.* **46**, 2005 (1975).
- [56] V. I. Gibalov and G. J. Pietsch, *J. Phys. D* **33**, 2618 (2000).
- [57] G. J. M. Hagelaar and L. C. Pitchford, *Plasma Sources Sci. Technol.* **14**, 722 (2005).
- [58] PHELPS and Morgan databases, <http://lxcatalaplace.univ-tlse.fr> Accessed: 31 January, 2017.
- [59] H. Hazama, M. Fujiwara, and M. Tanimoto, *Chem. Phys. Lett.* **323**, 542 (2000).
- [60] B. Bagheri, J. Teunissen, U. Ebert, M. M. Becker, S. Chen, O. Ducasse, O. Eichwald, D. Loffhagen, A. Luque, D. Mihailova, J. M. Plewa, J. Dijk, and M. Yousfi, *Plasma Sources Sci. Technol.* **27**, 095002 (2018).

End Matter

Appendix A: Summary of electron density diagnostics—2D electron density distributions were measured in single-shot recordings by dichroic Talbot interferometers (SM [24], Sec. 2), visualizing Talbot images of circular pinhole arrays in square lattice formation simultaneously at two wavelengths of continuous-wave diode lasers ($\lambda_1 = 785$ nm; $\lambda_2 = 405$ nm), as shown in Fig. 1(a). The Talbot images were recorded by two intensified charge-coupled devices (ICCDs) specific for each laser wavelength. In this method, plasma refractive indices integrated along the laser paths were converted into the spot shifts of the Talbot images $T_k(\lambda)$. Exploiting the λ

dependence of the plasma refractive indices, the two-dimensional line-integrated electron distributions were obtained by subtraction of the two images [$T_k(\lambda_1) - T_k(\lambda_2)$]. The horizontal distributions of the line integrals at each axial position were fitted with Gaussian forms and converted into radial electron density profiles by inverse Abel transformation under the axial symmetry of the plasma. The error ranges of the radial electron density profiles were caused by the measurement uncertainty of $T_k(\lambda)$ that could be mainly induced by statistical errors and systematic errors. The error range was determined by quantifying the statistical errors, synthesizing random

TABLE I. Representative specification of electron density diagnostics.

Parameter	Value	Determinant factor
Temporal resolution	2 ns	Exposure time of ICCDs
Spatial resolution	300 μm	Pitch of pinhole arrays
Single-shot sensitivity	$1 \times 10^{14} \text{ cm}^{-3}$	Talbot length ($\sim 1.8 \text{ m}$)

noise under the assumption that the statistical errors follow normal distributions, adding the synthesized noise to the measured line-integrated electron density distribution, fitting the noisy line-integrated electron density distribution with the Gaussian function, conducting the inverse Abel transformation to the Gaussian function, repeating this process 10 000 times, assessing 68% confidence intervals of the 10 000 radial electron density profiles, and, finally, accumulating the systematic errors (+3%) to the intervals. Representative specifications of the electron density diagnostics are listed in Table I.

*Appendix B: Summary of electric field diagnostics—*1D distributions of electric field vectors (E_y/N and

TABLE II. Representative specification of electric field diagnostics.

Parameter	Value	Determinant factor
Temporal resolution	190 fs	Laser pulse width
Spatial resolution	20 μm	ICCD pixel size, visual field
Accuracy for $E_y(r)/N$	$\pm 6\%$	Shot accumulation (~ 500)
Accuracy for $E_r(r)/N$	$\pm 10\%$	Shot accumulation (~ 500)

E_r/N) were measured by an E-FISH sensor (SM [24], Sec. 3), employing a femtosecond laser beam focused along the horizontal direction ($\lambda = 1030 \text{ nm}$; pulse energy = 1 mJ), as shown in Fig. 1(a). The E-FISH signal is induced by a third-order, nonlinear optical effect of the gaseous plasma and was recorded by an ICCD. The measured electric field component was selected by the polarization direction of the laser beam. The E-FISH signal calibration for the electric field was conducted using uniform Laplacian fields generated by 10-mm-diameter plate-to-plate electrodes for dc and pulsed voltage waveforms; both calibration constants were found consistent with each other. Furthermore, the calibration was conducted beyond the critical ionization

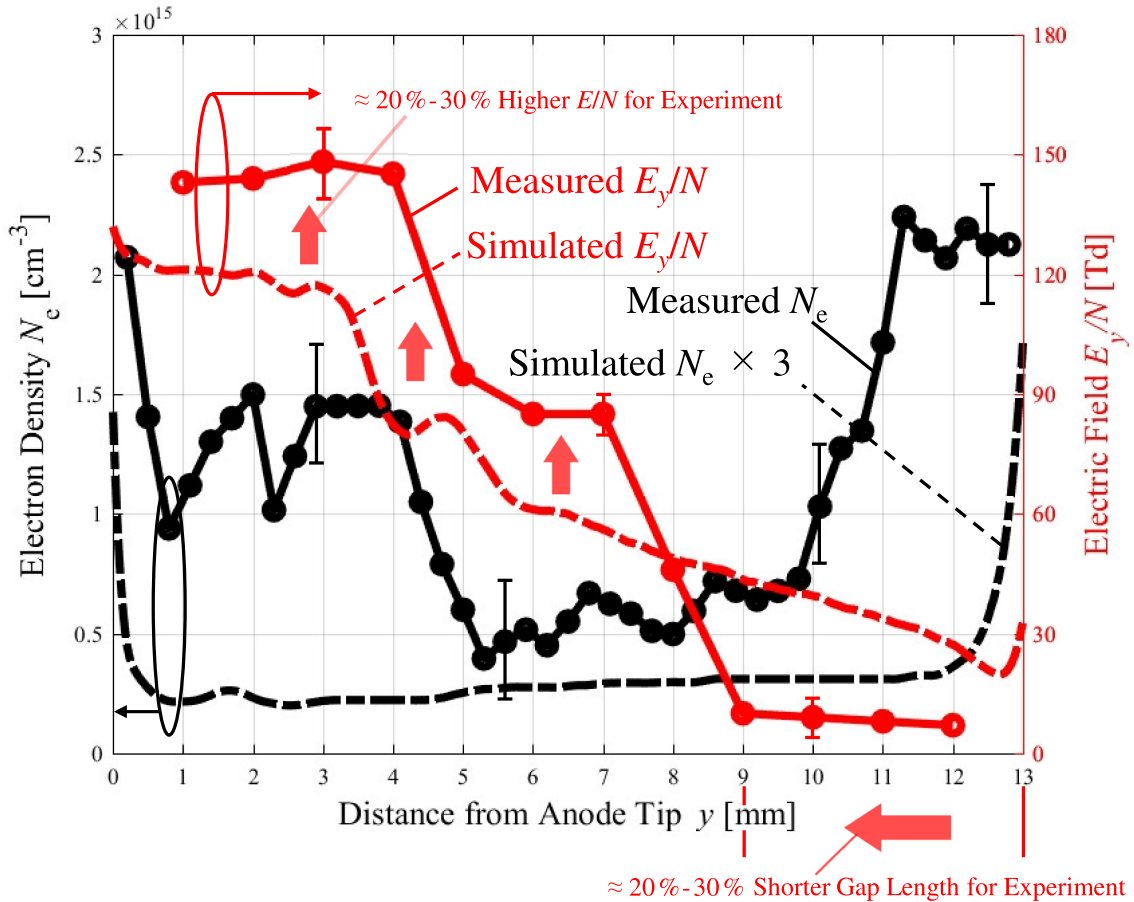


FIG. 5. Comparison between measurement and numerical simulation [23] in early secondary phase.

electric field as well as below it, using equivalency between the electric field and laser intensity.

The measured E-FISH signal distributions along the horizontal axis were converted into radial electric field distributions by a forward-model inversion: assumed cylindrically symmetric model profiles for E_y/N and E_r/N , computed the horizontal E-FISH signal distributions for the assumed profiles, transformed the computed E-FISH signal distributions into analytic electric fields via the analytic calibration constant, and iteratively adjusted the model profiles until the analytic electric fields matched the measured corresponding ones within preset mean absolute percentage errors (5% for E_y/N , 15% for E_r/N). The measurement uncertainty of the radial E_y/N and E_r/N profiles was caused by statistical errors (shot noise of the ICCD) contaminating the measured E-FISH signal. The measurement uncertainty (accuracy) was assessed by determining the model profiles for the upper and lower ends of the statistical errors (95% confidence intervals under assumption of normal distributions). Representative

specifications of the electric field diagnostics are listed in Table II.

Appendix C: Comparison between measurement and numerical simulation—Figure 5 shows comparison of the axial distributions of the electron densities at the radial centers and electric fields E_y/N on the y axis between the present measurement and previous numerical simulation in the early secondary phase (at 5 ns after primary streamer contact with the cathode for the measurement; at 7 ns after primary streamer contact with the cathode for the numerical simulation [23]). For convenience of terminology, “transition phase” for the numerical simulation [23] is until 3 ns after primary streamer contact with the cathode, following the present experimental results. The error bars of the measured electron densities indicate the error ranges described in Appendix A (SM [24], Sec. 2.3.2). The error bars of the measured electric fields E_y/N indicate the measurement accuracy described in Appendix B (SM [24], Sec. 3.2.1). Notably, numerically simulated electron densities demonstrated here are multiplied by 3.