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Discharge similarity in capacitive radio frequency carbon tetrafluoride plasmas

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ABSTRACT

Although similarity laws for breakdown have been demonstrated for molecular gases (e.g., N_2 and NF_3), their application to rf plasmas in electronegative gases remains unexplored. In this Letter, we report the discharge similarities in steady-state CF_4 rf plasmas via fully kinetic particle-in-cell simulations. The results show that the rf discharges of different dimensional scales obey the similarity law in both the striation mode and drift-ambipolar mode when the similarity conditions are fulfilled with the scaling factor k being relatively small (e.g., $k \leqslant 2$). As the scaling factor k increases, violations caused by "nonlinear collisions" become important, and deviations from the similarity are observed. The nondimensional parameters, η_ω and η_n, for characterizing the formation of the striation mode are evaluated for understanding the mode transitions under similar discharge conditions.

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Low-pressure capacitive radio frequency (rf) discharges have emerged as a fundamental technology in the field of semiconductor manufacturing, particularly in critical processes such as plasma etching and thin-film deposition.[1,2] Precise control and accurate prediction of plasma parameters are crucial for optimizing these manufacturing processes and ensuring product quality in practical applications. The detailed insights into electron kinetics and discharge modes can contribute to a comprehensive understanding of the underlying physics of rf plasmas, especially the mode transitions and the prediction of plasma parameters.[3–5] The development of chip fabrication tends to require larger-area plasma sources to improve the product yield and quality.[6] Comprehensively understanding the plasma behaviors of rf discharges across different dimensional or pressure scales is essential for the development and optimization of plasma equipment. It has been demonstrated that the similarity law for gas discharges offers a theoretical framework to correlate and predict discharge parameters at various scales,[7,8] which can help guide the design and optimization of plasma sources under diverse conditions.

The similarity principles in rf discharges have been proven as a useful tool to generalize plasma behaviors under different operating conditions. By scaling key parameters simultaneously (including gas pressure p, gap dimension d, and driving frequency f) (keeping the combined parameters pd and f/p constant), similarity in the physical properties of the compared rf discharge systems can be achieved. On this basis, plasma properties in similar rf discharges can be correlated across various dimensions without the requirement of additional simulations or experiments. The similarity principle has been demonstrated for electropositive (e.g., argon) rf plasmas by both experiments and through first-principle particle-in-cell (PIC) simulations.[9–17] In previous studies, spatiotemporal evolutions of plasma parameters under

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similar discharge conditions have been confirmed for single-frequency,[9–11] dual-frequency,[12] magnetized capacitive rf plasmas,[13,14] and inductive rf plasmas.[15,16] More recently, the similarity laws for capacitive rf plasmas have been experimentally demonstrated via the phase-resolved optical emission spectroscopy technique.[17] Based on the similarity theory, scaling networks, which can extensively correlate the discharge states [i.e., the initial, similarity states, and their intermediate when one or two control parameter(s) is/are tuned], have been established, enabling an effective strategy for determining scaling relations with fewer experiments.[17,18] However, the application of the similarity laws to electronegative rf plasmas has been rarely explored and remains ambiguous.

Electronegative gases (e.g., CF_4) are widely used in plasma etching processes.[19–21] Lisovskiy experimentally demonstrated the breakdown similarities for molecular gases (e.g., N_2 and NF_3).[22] Franklin developed a model for recombination dominated electronegative plasmas, which indicates that the electron temperature (T_e) is related to the combined parameter pd (d denotes the discharge dimension).[23] The combined parameter pd also affects the discharge regimes and their transitions in electronegative discharge.[24] However, the applicability of similarity laws to strongly electronegative rf discharges at the steady state is still unclear. In electronegative gas discharges, the presence of negative ions may have an essential effect on the spatial distribution of parameters and the discharge structure.[25] In glow steady-state discharges, the production and drift velocity of negative ions can affect the behavior of the positive column,[25] while in rf discharges, the modulation on temporal and spatial scales further complicates the discharge structure. Particularly, rf discharges in electronegative gases present unique discharge modes that differ from electropositive gas discharges, such as the drift–ambipolar (DA) mode[26,27] and the striation mode.[28,29] The DA mode is induced by a strong drift electric field due to the low conductivity in the bulk and an ambipolar field caused by local maxima of the electron density at the sheath edge.[26,27] On the basis of the DA mode, the striation mode is generated when the driving frequency and the eigenfrequency of the ion–ion plasma achieve resonance.[28,29] Previous studies have confirmed that the similarity laws hold for rf discharges in both $\alpha-$ and $\gamma-$ modes as well as their transition regimes.[30] However, whether or not the striation mode and the DA mode in electronegative rf plasmas obey the similarity laws is not yet answered.

According to the similarity theory,[10,31] a physical parameter G at the corresponding spatiotemporal point (x, t) can be transformed between the original ($k = 1$) and similar discharge system via

$$G(x_1, t_1) = k^{\alpha[G]} G(x_k, t_k), \tag{1}$$

where k is the scaling factor and $\alpha[G]$ denotes the similarity factor for parameter G. It has been determined that $\alpha[x] = \alpha[t] = 1$ for length and time, $\alpha[p] = \alpha[f] = -1$ for pressure and frequency, and $\alpha[n_e] = \alpha[\mathbf{J}_e] = -2$ for electron density and electron current density in previous studies.[10,31] The similarity factor for all the physical parameters can be derived based on the power-law relation in Eq. (1). Furthermore, parameters having $\alpha[G] = 0$ (e.g., the combined parameters pd and f/p) or some dimensionless parameters (e.g., Knudsen number λ/d) are termed similarity invariants.[31] Similar rf discharge systems can be established by simultaneously tuning the gas pressure p, gap distance d, and frequency f and keeping pd and f/p constant, thus the discharge condition parameters are scaled as $k = p_k/p_1 = d_1/d_k = f_k/f_1$.

According to the similarity theory, the similarity factor for the electron density growth rate can be determined as $\alpha[\partial n_e/\partial t] = \alpha[\partial n_e] - \alpha[\partial t] = -3$, where $\alpha[\partial n_e] = \alpha[n_e] = -2$ and $\alpha[\partial t] = \alpha[t] = 1$. Processes, for which in terms of the similarity laws, the growth rate of each newly produced charged species density n_s in the compared discharge follows

$$\left(\frac{\partial n_s}{\partial t}\right)_1 = k^{-3}\left(\frac{\partial n_s}{\partial t}\right)_k, \tag{2}$$

are classified as *linear collision processes*. A collision process is classified as *nonlinear* if Eq. (2) is not satisfied.[32] The presence of nonlinear collision processes can cause a violation of the discharge similarity.

The validity of similarity has been confirmed in argon rf discharges, in which the linear processes dominate the discharge and the nonlinear collisions (e.g., stepwise ionization processes) play a minor role.[9] However, in the electronegative CF_4 rf discharges, the nonlinear collisions (e.g., recombination processes) are of significant importance, and their impact on the validity of similarity laws has not yet been evaluated. A deeper understanding of CF_4 rf plasmas under similar discharge conditions is still missing, which may aid in correlating discharge properties in cross-comparison systems.

In this work, fully kinetic one-dimensional (1d3v) particle-in-cell/Monte Carlo collision (PIC/MCC) simulations are conducted to investigate the discharge similarity in CF_4 rf plasmas. Details of the employed simulation model can be found in previous studies,[27,33,34] which has been benchmarked and validated against experimental measurements.[28,34,35] The theoretical basis for understanding the invariance of the striation mode is analyzed. The simulations are conducted for CF_4 rf discharges, including electrons, CF_3^+, F^-, CF_3^-, and CF_2^+ ions as the traced charged particles. The related cross sections and reaction coefficients for electron-neutral particle collisions and ion-neutral particle collisions can be found in Refs. 33 and 34. The simulations incorporate recombination between positive and negative ions and the recombination between electrons and CF_3^+ or CF_2^+. The recombination rate coefficients employed in the simulations are consistent with those in Refs. 34, 36, and 37. The background gas temperature is maintained at $T_g = 350$ K.[34,38] To ensure numerical stability and accuracy, the time step is $\Delta t = T_{rf}/8000$ with T_{rf} being the rf period, and the space step is $\Delta x = d/1000$ with d being the gap distance.

The rf plasmas are generated between two parallel-plate electrodes, where the powered electrode ($x = 0$) is driven by $V_{rf}(t) = \phi_0 \cos(\omega t)$, while the other electrode is grounded. A constant electron reflection coefficient of $R = 0.7$ and a positive ion-induced secondary electron emission coefficient of $\gamma = 0.01$ are assumed for the powered and grounded electrodes.[34,39] In the simulations, the voltage amplitude ϕ_0 is varied within the range of 100–300 V. The discharge conditions for the base case (with $k = 1$) at different voltage amplitudes are $[p_1, d_1, f_1] = [30\ \text{Pa}, 30\ \text{mm}, 6.78\ \text{MHz}]$. For each case, simulations are performed with scaling factor values of $k = 1$, 2, 4, and 10, where the scaled control parameters $[p_k, d_k, f_k]$ are given by $k = p_k/p_1 = d_1/d_k = f_k/f_1$. By sweeping the scaling factor k, the scale invariance of the striation and DA modes is examined.

In CF_4 rf discharges, the dominant linear and nonlinear collision processes, including ionization, attachment, and recombination

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TABLE I. Dominant linear and nonlinear collisions in CF_4 rf discharges.

No.	Collision processes	Collision type
R1	$e + CF_4 \rightarrow 2e + CF_3^+ + F$	Linear collision
R2	$e + CF_4 \rightarrow 2e + CF_2^+ + 2F$	Linear collision
R3	$e + CF_4 \rightarrow F^- + CF_3$	Linear collision
R4	$e + CF_4 \rightarrow F + CF_3^-$	Linear collision
R5	$e + CF_3^+ \rightarrow CF_3$	Nonlinear collision
R6	$F^- + CF_3^+ \rightarrow F + CF_3$	Nonlinear collision
R7	$CF_3^- + CF_3^+ \rightarrow CF_3 + CF_3$	Nonlinear collision

processes, are shown in Table I. According to Eq. (2), the similarity factor for the reaction rate of linear collisions is expected to be −3. More specifically, the similarity factors for the reaction rate of ionization (R1, R2) and dissociative attachment (R3, R4) processes are expressed as follows:

$$\alpha[R_r(j)] = \alpha[K_r(j)] + \alpha[n_e] + \alpha[n_g] = -3, \quad j = 1, 2, 3, 4, \tag{3}$$

where $\alpha[n_g] = \alpha[p] = -1$ and $K_r(j)$ is the corresponding rate coefficient for the jth reaction, which is a similarity invariant, i.e., $\alpha[K_r(j)] = 0$.[10] However, for recombination processes, the similarity factors are given by

$$\alpha[R_r(j)] = \alpha[K_r(j)] + \alpha[n_{s1}] + \alpha[n_{s2}] = -4, \quad j = 5, 6, 7, \tag{4}$$

where $\alpha[n_{s1}] = \alpha[n_{s2}] = -2$ for the charged particles involved. The recombination processes are, thus, classified as nonlinear collisions, which may lead to a violation of similarity laws.

Figure 1 shows the spatiotemporal evolutions of the ionization rate (R1) under similar discharge conditions with the voltage amplitudes ϕ_0 of 100, 200, and 300 V. The scaling factor is set to k of 1 (base case), 2, 4, and 10 for each voltage amplitude. It can be observed that higher voltages are more likely to induce the development of the striation mode. For example, in the case $k = 2$, no striation occurs for $\phi_0 = 100$ V, whereas distinct striation structures can be observed at $\phi_0 = 200$ and 300 V. At 300 V for cases with $k = 1$ and $k = 2$, the striation structures are rather alike. Likewise, for cases $k = 4$ and $k = 10$ (corresponding to a dimension magnification of 2.5), the ionization rate in the DA mode also exhibits similar spatiotemporal distributions. Although the spatiotemporal profiles of the ionization rates show similarity under different k conditions, the striation structures become less pronounced and even diminish as k increases. The striation structure becomes unrecognizable, showing a transition to the DA mode as k increases to 4 and 10. The discharge similarity no longer exists with a larger k factor, since the discharge mode is changed in the systems under comparison. The results confirm that the discharge similarity of the striation mode and the DA mode exists when the scaling factor is relatively small to maintain the compared discharges in the same mode.

According to the similarity theory, charged particle densities should satisfy the transformation relation $n_{s,1} = k^{-2} n_{s,k}$.[31] Figure 2(a) shows the spatiotemporally averaged charged particle density scaling with the scaling factor k at a voltage amplitude of 300 V. The charged particle densities demonstrate a power law as a function of k. The particle density increases as k increases, which is consistent with the

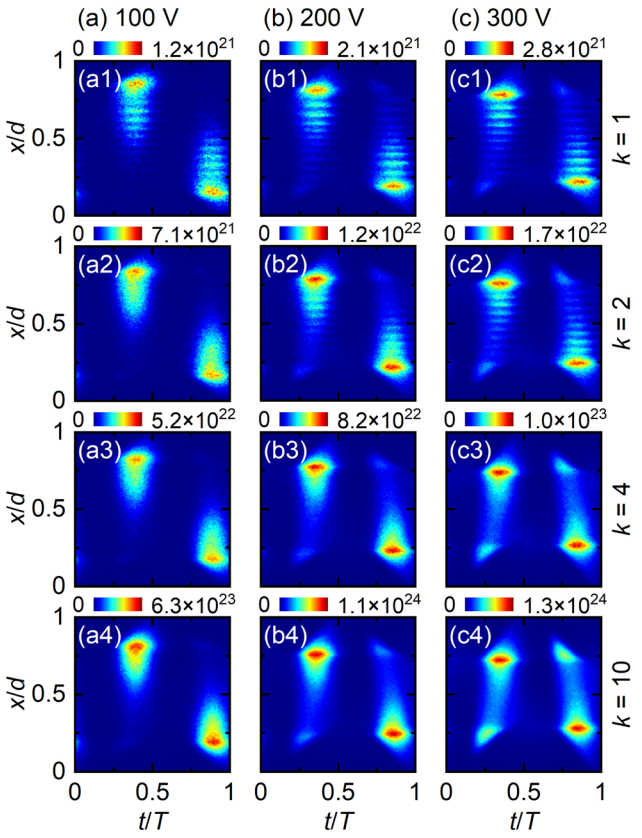


FIG. 1. Spatiotemporal evolutions of the ionization rate (unit in $m^{-3}s^{-1}$) under similar discharge conditions with different voltage amplitudes ϕ_0 of (a) 100 V, (b) 200 V, and (c) 300 V. The rf discharge condition parameters are $[p_1, d_1, f_1] = [30\ \text{Pa}, 30\ \text{mm}, 6.78\ \text{MHz}]$ for $k = 1$ (base case), while the discharge condition parameters for $k = 2$, 4, and 10 (compared cases) are scaled as $[p_k, d_k, f_k] = [kp_1, k^{-1}d_1, kf_1]$.

theoretical prediction. The electron density shows a scaling law of $n_e \propto k^{1.85}$, which slightly deviates from the theoretical scaling $n_s \propto k^2$. The ion densities, largely affected by the nonlinear processes (R5, R6, and R7), scale approximately as $n_i \propto k^{1.4}$, showing a relatively larger deviation. The scaling relations for charged particle densities are almost the same under other voltage amplitude conditions. This discrepancy from the similarity relation is caused by the influence of nonlinear collisions, which will be discussed later.

To quantify the deviation from the theoretical prediction, the relative deviation percentage of charged particle densities with different scaling factors is determined as

$$\delta n_s = \frac{k^{-2} n_{s,k} - n_{s,1}}{n_{s,1}}. \tag{5}$$

As shown in Fig. 2(b), the deviation δn_s is negative for all the cases. This indicates that the normalized densities $k^{-2} n_{s,k}$ in similar systems (at $k > 1$) are lower than those in the base case. The deviations are more pronounced for ions than for electrons and increase with k. The magnification of the dimensions for different generations of rf plasma chamber and wafer is typically by a factor of 1.5 or 2.[1] When the scaling factor is relatively small ($k \leqslant 2$), the violation of discharge

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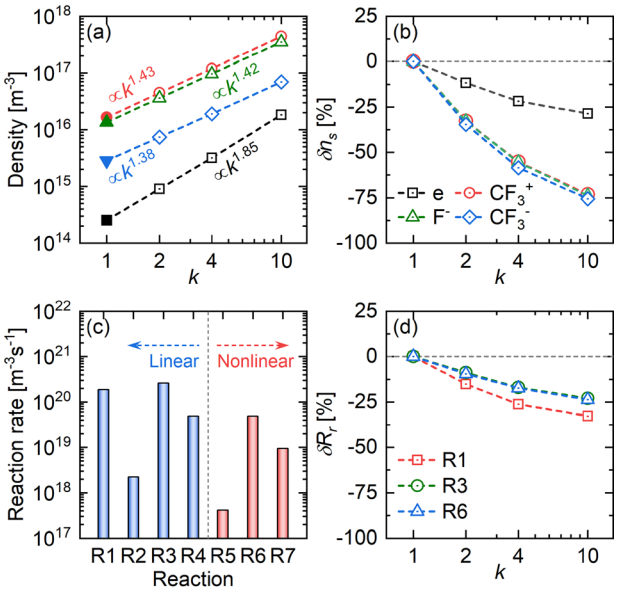


FIG. 2. (a) Spatiotemporally averaged plasma density and (b) deviation δn_s as a function of the scaling factor k at a voltage amplitude of $\phi_0 = 300$ V. The scaling law for plasma densities, which is obtained by fitting the data points from simulation results, deviates from the theoretical similarity relation $n_s \propto k^2$. (c) Spatiotemporally averaged reaction rate R_r (case $k = 1$) and (d) similarity deviation $\delta(R_r)$ of the dominant reactions (R1, R3, and R6) vs the scaling factor k at a voltage amplitude of $\phi_0 = 300$ V.

similarity is minor since the similarity prediction holds generally reasonable. Taking the case with $k = 2$ as an example, i.e., the dimensions of two rf plasma sources differ by a factor of two, the deviation is around 10% for electron density and approximately 35% for the ion densities compared to the base case.

Figure 2(c) shows the spatiotemporally averaged reaction rates of the main reactions in CF_4 rf discharges at a voltage amplitude of 300 V. The results indicate that R1 serves as the dominant ionization channel, while the attachment process is governed by reaction R3, and the recombination process is dominated by reaction R6. The other reactions exhibit relatively smaller rates in the respective reaction types. The key processes correspond to the production and destruction of the four main charged species (e, CF_3^+, F^-, and CF_3^-) in steady-state rf discharges. It can be seen that the percentage of nonlinear collisions is relatively high. Specifically, in the base case, the ratio of the averaged reaction rates between R6 (4.90×10^{19} m^{-3} s^{-1}) and R1 (1.87×10^{20} m^{-3} s^{-1}) is approximately 26%. The relatively high proportion of the recombination process indicates that the role of nonlinear collisions is significant, which is responsible for the deviations from the similarity relation.

Figure 2(d) presents the deviation of the reaction rates for the three dominant reactions, which are calculated by

$$\delta(R_r) = \frac{k^{-3}R_{r,k} - R_{r,1}}{R_{r,1}}, \tag{6}$$

where $\delta(R_{r,k})$ is the spatiotemporally averaged reaction rate in the compared case with the scaling factor k. The deviation percentage $\delta(R_r)$ generally increases as k increases. The deviation $\delta(R_r)$ is negative, which indicates that the normalized reaction rate is less than that in the base case. Thus, the nonlinear collision processes have a relatively minor impact on the similarity law when the scaling factor k is small. The deviations in the dominant reactions remain less than 15% for $k \leqslant 2$. However, as k increases, the effects of nonlinear processes become more pronounced, showing a marked violation of the similarity law, which is consistent with the density scaling [see Fig. 2(b)].

In electronegative gas rf discharges, the striation mode can form under specific conditions:[28,40] the bulk plasma region exhibits very low electron density and high electronegativity, similar to the DA mode. Additionally, the ion plasma frequency ω_{pi} approaches or exceeds the rf angular frequency ω_{rf} ($\omega_{pi} \geq \omega_{rf}$), allowing ions to oscillate in response to the rf electric field. The coupling between the motions of positive and negative ions in rf ion-ion plasmas has been analyzed to elucidate the underlying physics of striation formation.[28,40] Based on the momentum balance equations for ions, the resonance condition for an ion–ion plasma subjected to an rf electric field is given by

$$\omega_{cr} = \sqrt{\frac{e^2 n}{\varepsilon_0 \mu}} \geq \omega_{rf}, \tag{7}$$

where e is the elementary charge, $n = n_+ = n_-$ is the ion density, ε_0 is the vacuum permittivity, and $\mu = (m_+ m_-)/(m_+ + m_-)$ is the reduced mass of the positive and negative ions. This critical condition [Eq. (7)] reveals the fundamental mechanism governing the striation formation, determining the required critical ion density for resonance as

$$n_{cr} = \frac{\varepsilon_0 \mu \omega_{rf}^2}{e^2}, \tag{8}$$

where the striations may occur for $n \geq n_{cr}$.

According to the similarity transformation [Eq. (1)], the similarity factors for the resonance condition of the striations are expressed as $\alpha[\omega_{cr}] = \frac{1}{2}\alpha[n] = -1 = \alpha[f]$ and $\alpha[n_{cr}] = 2\alpha[\omega_{rf}] = -2 = \alpha[n_e]$. This indicates that the resonance frequency and critical density transformations also obey the similarity law. Therefore, the striation mode can be theoretically preserved under similar discharge conditions.

To quantitatively determine the formation of the striation and its similarity, two dimensionless parameters are defined as follows:

$$\eta_\omega = \omega_{cr}/\omega_{rf} = \sqrt{\frac{e^2 n}{\varepsilon_0 \mu \omega_{rf}^2}}, \tag{9a}$$

$$\eta_n = n/n_{cr} = \frac{e^2 n}{\varepsilon_0 \mu \omega_{rf}^2}, \tag{9b}$$

where $\eta_\omega^2 = \eta_n$. Equations (9a) and (9b) imply that striations can occur when $\eta_\omega \geqslant 1$ or $\eta_n \geqslant 1$. Then, the deviation from the similarity law at larger k (see Fig. 2) can be indicated by the variation of η_ω and η_n values.

Figure 3 demonstrates the variation of η_ω and η_n with the scaling factor k for different voltage amplitudes. In our cases, CF_3^+ and F^- are dominant positive and negative ions, respectively. To ensure the validity of the results, the ion density is taken as the averaged density in the bulk region, while the sheath region is excluded. The results show that the conditions $\eta_\omega \geq 1$ and $\eta_n \geq 1$ are satisfied for $\phi = 100$ V with $k = 1$ [Fig. 1(c1)] as well as for $\phi = 200$ V [Figs. 1(b1) and 1(b2)] and

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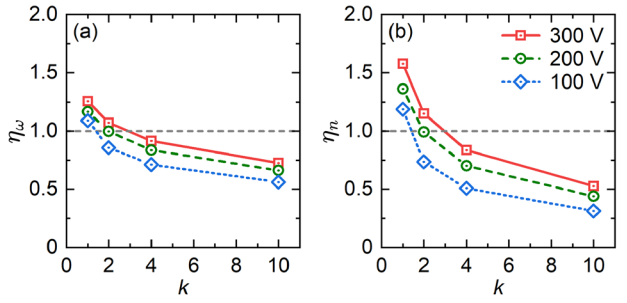


FIG. 3. Variation of (a) η_ω and (b) η_n vs the scaling factor k for different voltage amplitudes, which are indicators for the presence of the striation mode. Here, $\eta_\omega \geqslant 1$ or $\eta_n \geqslant 1$ indicates the presence of the striation mode.

300 V [Figs. 1(c1) and 1(c2)] with $k = 1$ and 2; striations are observed accordingly, which confirms the validity of the criteria. However, as k increases, the violation of similarity caused by the nonlinear collisions leads to a progressive reduction in the ion density, which is below the theoretical prediction (see Fig. 2). Thus, η_ω and η_n gradually decrease with an increased k. When η_ω or η_n becomes less than 1, the striations disappear. The prediction is consistent with the PIC results in Fig. 1 and aids in understanding the similarity of striation mode with a limited scaling factor.

In conclusion, the discharge similarity in CF_4 rf discharges is investigated through PIC simulations. The results show that the discharge in the striation and DA modes both follow similarity laws when the scaling factor is relatively small (e.g., $k \leqslant 2$). Theoretical analysis suggests that the striation mode should obey similarity principles; however, under practical discharge conditions, nonlinear collisions could cause the scaling relation of plasma density to progressively deviate from the theoretical predictions. As the scaling factor k increases, the discharge may transition from striation to the DA mode. The similarity invariants are proposed based on the analytical model for better understanding of the mode transition and striation formation. Under the critical condition for the striation formation, two similar discharge systems may exhibit different modes, one in the striation mode and the other in the DA mode, showing the mode transition. The mode transition corresponds to the deviations from similarity caused by nonlinear collisions. To maintain the striation mode invariance in similar discharge systems, the striation mode conditions should be simultaneously fulfilled in the compared systems. The results from this work extend the applicability of similarity laws to electronegative gas rf plasmas but with limited conditions, such as in the same discharge modes with relatively smaller scaling factors.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Dong Yang: Formal analysis (equal); Investigation (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Xiaokun Wang:** Formal analysis (equal); Investigation (equal); Methodology (equal); Resources (equal); Software (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Zhenhua Zhou:** Formal analysis (supporting); Investigation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Hanyang Li:** Formal analysis (supporting); Investigation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Wenjin Zhang:** Formal analysis (supporting); Investigation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Yongxin Liu:** Formal analysis (supporting); Investigation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Julian Schulze:** Formal analysis (supporting); Investigation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Peter Hartmann:** Formal analysis (supporting); Investigation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Zoltan Donko:** Formal analysis (supporting); Methodology (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Yangyang Fu:** Conceptualization (lead); Formal analysis (equal); Funding acquisition (lead); Investigation (equal); Project administration (lead); Supervision (lead); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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