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·综述·

重复频率脉冲流注放电演变现象与机制研究进展*

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摘 要： 重复频率脉冲流注放电是低温等离子体前沿应用的关键使能因子，然而，高重复频率脉冲作用下流注放电呈现复杂的不稳定和记忆效应现象，放电基础演变机理和调控方法尚不完善，极大影响应用的安全性和放电特性调控的有效性。综述了重复频率脉冲流注放电演变现象与机制的研究进展。首先归纳了重复频率脉冲流注放电的强非线性和渐进式演变特征，然后分析不同类型放电记忆效应因子对后续流注起始和传播的作用机制，最后总结了脉冲波形参数对重复频率脉冲流注放电的影响规律。凝练了重复频率脉冲流注放电演变机制研究的若干挑战，对脉冲放电等离子体机理研究具有一定的借鉴作用。

关键词： 脉冲放电； 重复频率； 流注； 记忆效应； 放电演变

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Research progress on evolution phenomena and mechanisms of repetitively pulsed streamer discharge

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Abstract: The repetitively pulsed streamer discharge is a critical enabling factor in many advanced low-temperature plasma applications. However, the streamer discharge exhibits complex instabilities and memory effect phenomena under high-frequency repetitive pulses. Fundamental discharge evolution mechanisms and regulation methods are not thoroughly understood, which significantly affects the application safety and regulation efficiency of discharge properties. In this paper, evolution phenomena and mechanisms of repetitively pulsed streamer discharge are reviewed, strong nonlinearity and progressive evolution features are summarized for repetitively pulsed streamer discharge, different memory effect agents and their influential mechanisms on initiation and propagation of subsequent streamers are discussed, effects of pulse waveform parameters on repetitively pulsed streamer discharge are outlined, and several research challenges are proposed regarding evolution mechanisms of repetitively pulsed streamer discharge, which would be helpful for revealing mechanisms of pulsed discharge plasmas.

Key words: pulsed discharge； repetitive frequency； streamer； memory effect； discharge evolution

流注放电是一种重要低温等离子体，相较于交直流驱动方式，重复频率脉冲驱动的流注放电具有高约化场强、电子能量大、活性高、体积大、稳定性好等显著优点[1]，在低温等离子体前沿应用中展现了独特优势，如辅助燃烧与流动控制[2]、辅助催化[3]、污染物降解[4]、等离子体活化水制备[5]、肿瘤细胞凋亡诱导[6]等，脉冲宽度覆盖 ns 至 μs 范围，脉冲频率可达数十 kHz[7-9]。提高电压幅值和脉冲频率有利于增强流注放电等离子体物理、化学、生物效应强度[7]。然而，高重复频率脉冲流注放电呈现复杂不稳定和记忆效应现象[10-11]，工作参数（如电压、重复频率、气压）固定时仍可能在一定数量脉冲作用后出现宏观和微观放电特性显著变化（如通道形态、发展模式、放电类型、放电机理等[11-15]），流注放电可能演变为有害的火花放电。重复频率脉冲流注放电演变机理和调控方法尚不完善，极大影响应用的安全性和放电特性调控的有效性，严重制约脉冲放电等离子体实际应用效果。本文综述重复频率

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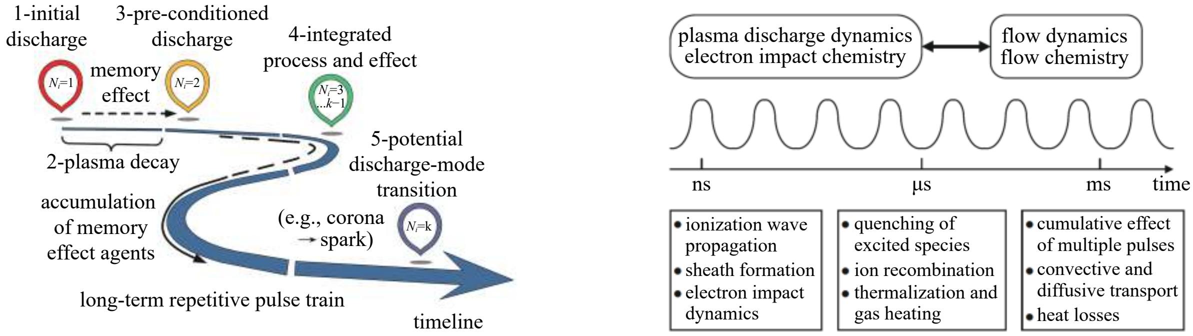
脉冲流注放电演变现象与机制研究进展，包括重复频率脉冲流注放电演变现象、重复频率脉冲流注放电记忆效应机制、脉冲波形参数的影响规律三个方面，并提出若干发展展望。

1 重复频率脉冲流注放电演变现象

重复频率脉冲流注放电演变现象复杂，是典型的多时空尺度和多物理场耦合问题。重复频率脉冲流注放电演变的基本环节如图 1 所示，主要包括以下阶段：



(a) evolution concept and different stages of repetitively pulsed discharge

(b) multiple timescales and physical phenomena in repetitively pulsed discharge

Fig. 1 Fundamental evolution stages and multi-timescale physical processes in repetitively pulsed streamer discharge[21-22]

图 1 重复频率脉冲流注放电演变的基本环节和多时间尺度物理过程[21-22]

初始放电。预电离水平较低，仅包含各种外界射线形成的背景电离，通常电子密度为 10^3～10^4 cm^{-3} 量级[16]。由于预电离水平较低，在 cm 级别气体间隙中，初始放电的统计时延很可能大于放电形成时延。

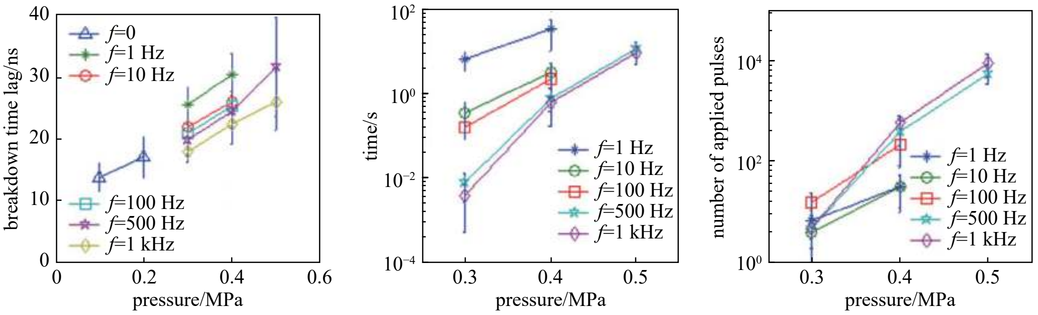
脉冲间隔期间的等离子体衰减过程。粒子衰减包含扩散、复合、退激励等过程[17-18]。若发生火花放电，还会发生温度梯度驱动下的显著气体对流过程[19]。

后续放电。后续放电可能表现出不同的起始和发展特性，变化程度与脉冲间隔时间、残余粒子作用机理、残余粒子空间分布等有关，可定性表述为存在“放电记忆效应”[20]。

长期积分效应。在一定数量脉冲连续作用后，残余粒子可能发生输运、聚集、转化等复杂过程。

脉冲放电显著转变。在重复频率脉冲长期作用下，可能出现如“流注-火花”转变等放电类型变化。

重复频率脉冲气体间隙和气固沿面的绝缘强度通常随频率增大而显著降低，初始流注放电可能演变为火花击穿[20, 23-24]。M. Pejovic 发现了低气压氩气、氮气放电时延随脉冲间隔时间呈非线性增大趋势[25-26]。中国科学院电工研究所邵涛系统性研究了重复频率纳秒脉冲气体间隙击穿特性，如图 2 所示，提出了包含击穿电压/场强、击穿时延、重复频率耐受时间、脉冲击穿数量等击穿特性表征体系，提出初始电子和亚稳态粒子密度增大是气体间隙击穿时延减小的主要原因[20, 27]。D. Pai 发现在 1～30 kHz 范围内脉冲频率对火花形成所需脉冲数量的影响较小[28]，但高重复频率下形成火花放电的最小脉冲电压显著降低，如图 3 所示。

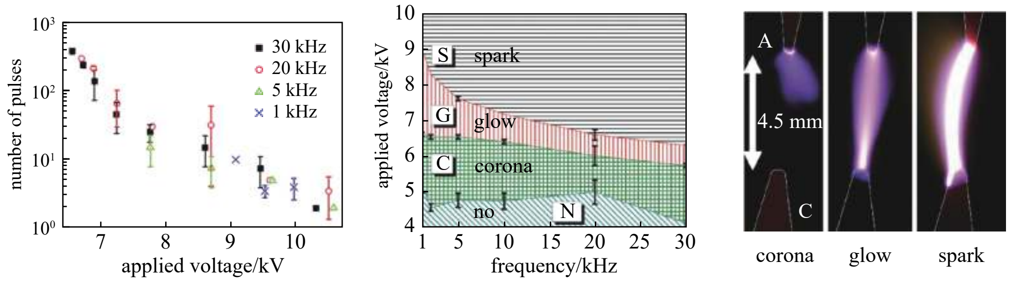


(a) breakdown time lag (b) repetitive stressing time (c) number of applied pulses

Fig. 2 Breakdown characteristics of plate-plate electrode and tip-plate electrode under repetitive nanosecond pulses[29]

图 2 重复频率纳秒脉冲下板-板电极和尖-板电极击穿特性[29]

重复频率脉冲流注放电通常具有渐进式演变特征，演变过程与电压幅值和频率有关。G. Naidis 仿真了重复频率纳秒脉冲针-板电极火花放电的“低电流加热-初始火花建立-稳态火花形成”三阶段形成过程[31]，放电电流、通道温度、电子密度等关键参数演变过程如图 4 所示。S. Nagaraja 基于自洽的重复频率纳秒脉冲介质阻挡放电（DBD）



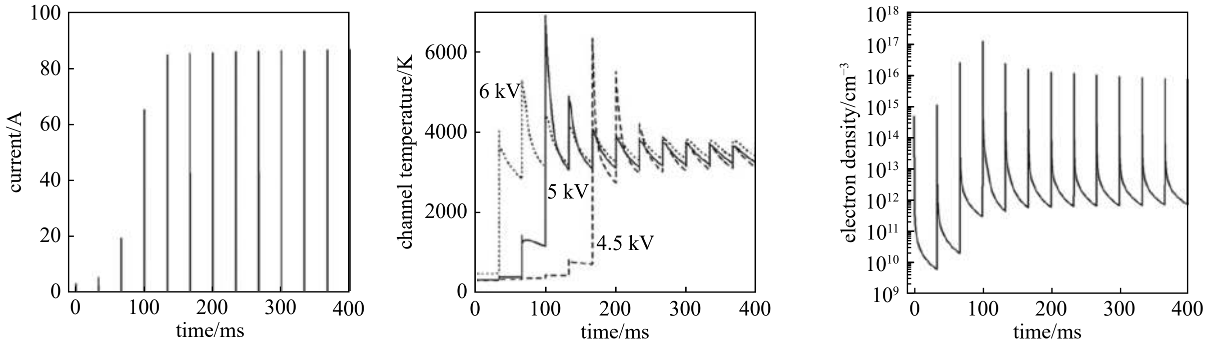
(a) number of pulses required to generate spark discharge

(b) relationship between discharge regime and frequency

(c) three types of discharge images

Fig. 3　Number of nanosecond pulses required for conversion to spark discharge and relationship between discharge interval and repetition frequency[12, 30]

图 3　重复频率纳秒脉冲火花转变所需脉冲数量及放电区间与脉冲频率的关系[12, 30]



(a) time evolution of current

(b) time evolution of channel temperature

(c) time evolution of electron density

Fig. 4　Three-stage formation process of spark discharge under repetitive nanosecond pulses[31]

图 4　重复频率纳秒脉冲针-板电极火花放电的三阶段形成过程[31]

仿真模型，获得了连续 100 个脉冲放电的活性粒子、约化场强、电子密度等关键参数演变过程，重复频率纳秒脉冲放电产生了空间均匀分布的氧原子，与脉冲放电阶段电子碰撞解离过程及余辉阶段激发态氮分子猝灭过程有关[21]。

2　重复频率脉冲流注放电演变的记忆效应机制

2.1　流注放电记忆效应因子类型及其作用机制

“前序放电显著影响后续放电”的记忆效应是重复频率脉冲流注放电演变的关键[32-33]。国内外研究提出多种记忆效应因子，包括亚稳态粒子、自由电子、正负离子、热量积累、表面电子脱陷等。不同因子对放电起始和发展的影响机制存在显著差异，因子类型与电压波形、气体压力、气体组分、前序放电类型、电场不均匀系数、脉冲间隔时间等因素有关。

亚稳态粒子寿命可达 s 级，通过超弹性碰撞过程、提供额外能量、促进阴极表面电子发射的途径促进后续流注发展，有利于流注向火花转变[17, 20, 34-36]。

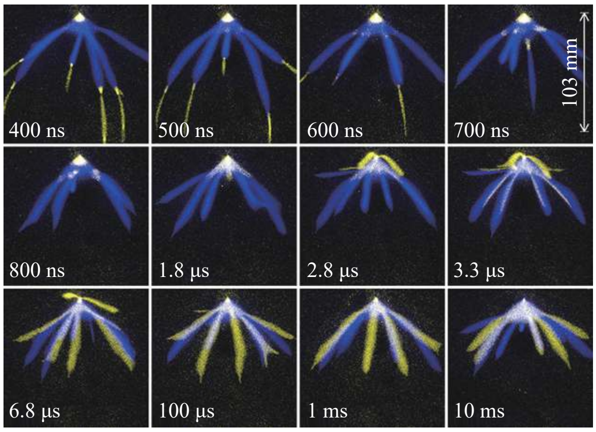


Fig. 5　Effect of the pulse delay time on variations of positive streamer channels (gas pressure: 13.3 kPa, voltage amplitude: 13.6 kV, pulse width: 200 ns). Images were created by superimposing two streamer discharge channels. Areas that only emitted during the first pulse are blue, areas that only emitted during the second pulse are yellow, and areas that emitted during both two pulses are white

图 5　脉冲间隔时间对正流注通道变化的影响（空气 13.3 kPa、幅值 13.6 kV、脉宽 200 ns）[18]。图中为两次流注放电图像的叠加，首次流注独有区域为蓝色，第二次流注独有区域为黄色，两次流注重叠部分为白色

空间自由电子显著影响后续正流注的起始和发展过程[37]。S. Nijdam 和 Y. Li 基于“双脉冲法”获得了脉冲延时对低气压针-板电极正流注放电的影响[18, 38]，发现了流注发展的暂停现象和流注演变的分阶段特性。如图 5 所示，随脉冲间隔时间增大，第二次正流注形态演变具有四个阶段：脉冲间隔时间为 100 ns 时，后续流注继续沿前序通道发展；脉冲间隔时间为 100 ns 至数 μs 时，后续流注仅沿前序通道的边沿发展；脉冲间隔时间为数 μs 至数 ms 时，后

续流注出现新的通道，并随脉冲间隔时间延长，新放电通道数目逐渐增多；脉冲间隔时间为 10 ms 量级时，两次流注通道无明显的相关性。后续流注仅在前序通道边沿发展的现象可能与残余高电导率通道对外电场的屏蔽作用以及前序通道边沿较高电子密度有关[18]。

空间残余离子迁移率低，可能影响空间电场分布。M. Kazemi 研究了线-筒同轴电极“双脉冲”放电特性，发现后续电晕电流幅值显著低于首次电晕，随双脉冲间隔时间增大，后续电晕电流逐渐增大并接近首次电晕，可能与残余正离子对线电极附近电场的削弱作用有关[39]。空间残余负离子可通过碰撞脱附过程释放自由电子，促进后续放电发展，特别是对于含电负性气体的情况[40-41]。

通道温度和密度变化是重复频率脉冲流注放电演变的重要因素，特别是对于有水分存在[42] 和形成火花放电情况[43]。若脉冲间隔时间小于气体密度恢复时间，后续放电约化场强增大，增强了后续脉冲下的碰撞电离过程，可能出现如图 6 所示的重复频率纳秒脉冲火花放电通道“自收缩”现象[43]。Y. Zhu 采用 2D-0D 耦合模型研究了脉冲电压下“流注-火花”转变过程，提出板电极附近场强高于电离阈值是火花形成的重要条件[44]。

表 1 总结了重复频率脉冲气体间隙和气-固沿面放电的部分关键记忆效应因子类型、影响机制和衰减途径。

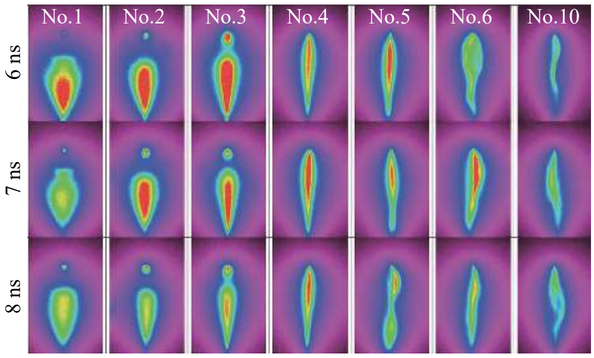


Fig. 6 The “self-focusing” phenomenon of spark discharge channel under repetitive pulses[43]

图 6 重复频率纳秒脉冲火花放电通道的“自收缩”现象[43]

表 1 重复频率脉冲气体间隙和气-固沿面放电中部分记忆效应因子类型、影响机制和衰减途径

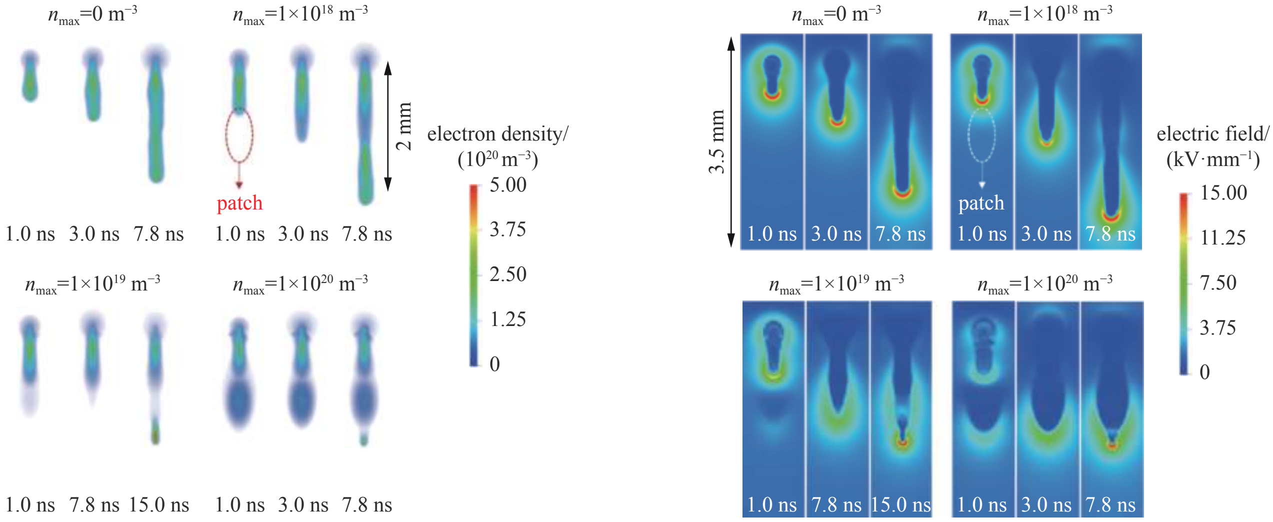
Table 1 Memory effect agents, influence mechanism and decay path in gas and gas-solid surface discharge under repetitive pulses

memory effect	memory effect agents	typical examples	major influential mechanisms	decay processes
volume memory effect	(1)positive ions	N_2^+, N_4^+, O_2^+, O_4^+ (dependenton gas composition)	(1)distort spatial E-field[39] (2)possibly shield E-field cooperatively with negative ions[18]	diffusion /recombination /drift
	(2)negative ions	O_2^-, O_4^-, SF_6^-, (dependenton gas composition)	(1)distort spatial E-field (2)possibly shield E-field cooperatively with positive ion [45] (3)provide seed electrons through detachment process[41]	diffusion /recombination /drift
	(3)electrons	free electrons	facilitate the initiation and guiding the propagation of next streamer (dependent on the spatial distribution) [18, 46]	diffusion /recombination /attachment/drift /‘clearing effect’
	(4)remaining conductivity	remaining streamer channel of a certain conductivity	inhibit the formation of a streamer (shielding effect on E-field)[18, 38]	diffusion /recombination /drift
	(5)metastable and excited species	$N_2(A^3\Sigma_u^+)$, $N(^2D)$, (dependent on gas composition)	(1)super elastic collisions[20, 34-35] (2)extra energy gain[20, 34-35] (3)reaction with dielectric[47-48]	diffusion /decay /loss on wall
	(6)variation of gas density	cylindrical shock wave nearly with the local sound speed	(1)affect the distribution of memory effect agents (2)affect the reduced E-field[37]	gas kinetics
	(7)gas heat accumulation	heat released from the discharge energy	affect the reduced E-field[14]	thermal diffusivity
surface memory effect	(1)surface trapped charges	trapped holes and electrons	(1)distort the surface E-field[49-50] (2)guide volume charge carrier drift and motion (3)released by disturbances and involved in the next streamer [33, 51, 52]	detrapping /surface conductivity /surface hopping /recombination
	(2)surface destructive aging	carbonization and surface roughness	(1)high surface conductivity[53, 54] (2)facilitate the initiation and propagation of surface streamer	roughly permanent
	(3)surface heat accumulation	heat from discharge energy	(1)surface property degradation [54-55] (2)decrease local gas pressure	thermal diffusivity

2.2　空间预电离“等离子体云”对流注放电的影响机制

后续流注放电如何与空间残余粒子相互作用是理解重复频率脉冲流注放电演变机制的关键，代表性工作是研究典型空间预电离“等离子体云”对流注放电起始和传播的影响，包括“电子-离子云”和“正离子云”两种情况。若脉冲间隔时间非常短，电荷吸附和复合反应不充分，前序脉冲放电通道内部残余的大量电子-正离子对形成“电子-离子云”。正流注通道内部呈现准电中性、通道外围带正电，整体放电区域正电荷密度高于负电荷密度，可能在放电结束后形成“正离子云”。此外，若脉冲电压下降时间较长，空间电荷沿放电通道的输运过程也可能形成净正电荷区，形成电场阶跃现象[56-57]。

S. Nijdam 和 J. Teunissen 采用三维流体模型研究了弱光电离率气体中的“电子-离子云”对后续流注放电的影响，发现“电子-离子云”具有引导流注放电发展的作用[58]。孙安邦基于 3D PIC-MCC 程序 Pamdi3D，系统研究了均匀电场下正流注与“电子-离子云”相互作用[59]，发现“电子-离子云”对后续流注放电发展速度产生显著影响。如图 7 所示，正流注进入“电子-离子云”前，云团中向上漂移的电子为正流注的起始与发展提供了种子电子，加速正流注发展；正流注进入“电子-离子云”后，正流注头部场强被极大地削弱，发展速度减缓；高密度“电子-离子云”可以完全屏蔽正流注头部场强，使得正流注发展停滞。但在背景电场作用下，云团内电子不断向上漂移，同时尾部场强逐渐增加，最终导致流注重新起始。



(a) electron density　　(b) electric field

Fig. 7　The electron density and electric field distribution: electron-ion plasma cloud interacting with the streamer

图 7　正流注与“电子-离子云”相互作用过程

N. Babaeva 采用二维流体模型发现流注放电在靠近高密度正离子云时发生分叉[60]。作者采用粒子-全局耦合模拟，研究了棒—板电极负极性纳秒脉冲放电的空间电荷分离现象[61]。如图 8 所示，电子倍增过程结束后，剩余脉冲电压作用下出现正负空间电荷分离现象，逐步在棒电极周围形成以正离子为主导的空间电荷区，即“正离子云”。正空间电荷区所产生的反向空间电场会阻碍后续放电中初始电离云向流注的转变，使得放电通道的发展接近于均匀电离的辉光放电模式。

3　脉冲波形对重复频率脉冲流注放电特性的影响规律

重复频率脉冲流注放电特性很大程度受脉冲波形参数（前沿、宽度、频率等）调控，包括过电压倍数、电子崩发展过程、流注通道形态、预电离分布、击穿电压幅值、放电不稳定性、快速加热机制等。

脉冲前沿和脉冲上升率影响脉冲放电的过电压倍数、放电通道形态、电子能量和密度。过电压倍数通常随脉冲前沿缩短而提高，高过电压倍数下气体放电的电子能量显著提高，甚至可能出现逃逸电子现象[62]，然而，F. Iza 提出平均电子能量随脉冲上升时间（1～40 ns）增大而增大，可能与准中性等离子体产生时延随脉冲前沿缩短而减小有关[63]。T. Ito 采用激光探针测量了重复频率脉冲 DBD 预击穿阶段空间电场，如图 9 所示，若外施脉冲上升时间远小于正离子迁移经过整个间隙的时间，会在阴极附近产生正离子积聚，导致阴极附近电场显著增强[64]。A. Komuro 仿真研究了脉冲上升率（0.11～0.52 kV·ns^{-1}）对大气压针—板流注放电发展速度和通道直径的影响，发现提高脉冲上升率会增大流注通道直径和传播速度，但流注头部约化场强基本不变[65]。

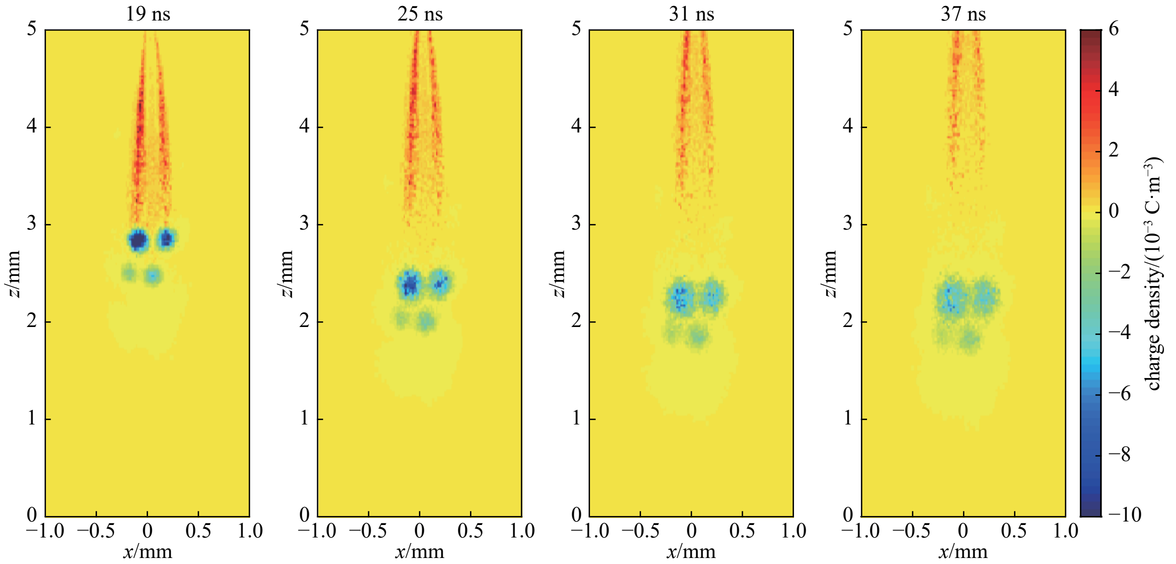


Fig. 8　Space charge separation under residual voltage[61]

图 8　剩余电压下空间电荷分离现象[61]

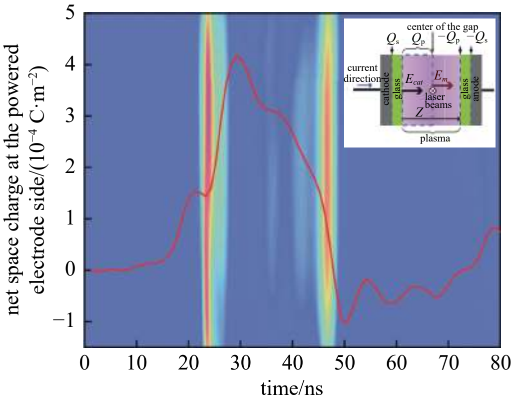
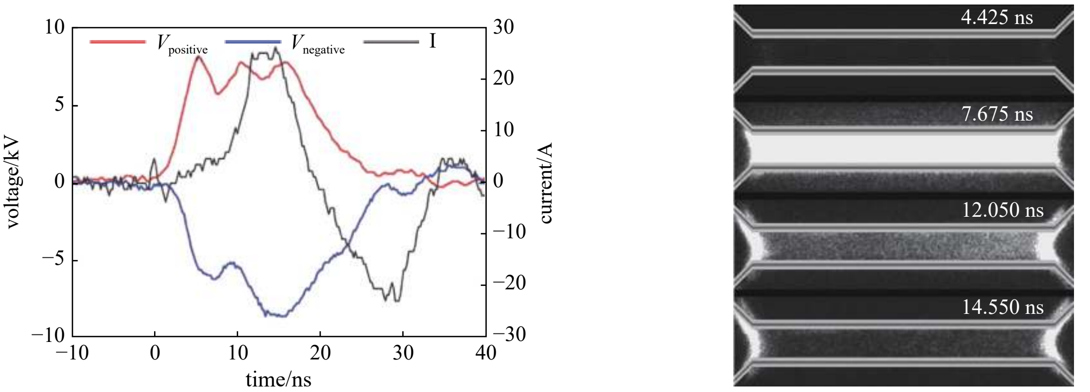


Fig. 9　Temporal evolution of the estimated charge in the half-space near the cathode, evaluated from the electric field and charge on the cathode (the background is the optical emission)[64]

图 9　基于空间电场和阴极电荷计算的阴极附近空间电荷密度随时间的变化（背景为放电光强分布）[64]

脉冲宽度影响流注放电的发展阶段和稳定性。纳秒脉冲流注放电过程通常包括初始流注、二次流注和类辉光等阶段[66-67]，“辉光-火花”转变因素包括热量累积[47]、分步电离[47]、阴极斑点形成[68]等，若脉冲宽度小于辉光放电不稳定性的发展时间，有可能维持高气压下辉光放电[69]。F. Qi 实现了重复频率纳秒脉冲大尺度辉光放电（电压幅值 16.2 kV，脉冲宽度约 20 ns，重复频率 40 kHz，气体间隙距离 7 mm），观察到放电在整个气体间隙同步发生的现象（时间精度为 0.125 ns），如图 10 所示。放电空间同步发生现象与传统流注放电的空间传播特性显著不同，可能的原因包括放电空间温度升高导致约化场强升高和较高预电离密度[70]。

脉冲频率直接影响预电离水平。B. Huang 研究了脉冲频率对重复频率纳秒脉冲大气压 Ne/空气微放电和低气压 He 快速电离波的影响[71-72]，如图 11 所示，随脉冲频率增大，快速电离波击穿前轴向电场强度峰值降低，击穿后轴向电场强度增加。作者提出了包括重复频率工作系数、击穿前耐受脉冲数量、电晕放电跟随系数、“序列-相位-强度-密度”分辨谱图在内的重复频率脉冲放电新表征参数体



(a) voltage and current characteristics　(b) spatiotemporal evolution of glow discharge

Fig. 10　Uniform DBD glow discharge under repetitive nanosecond pulses in atmospheric pressure air[70]

图 10　重复频率纳秒脉冲大气压空气 DBD 均匀辉光放电[70]

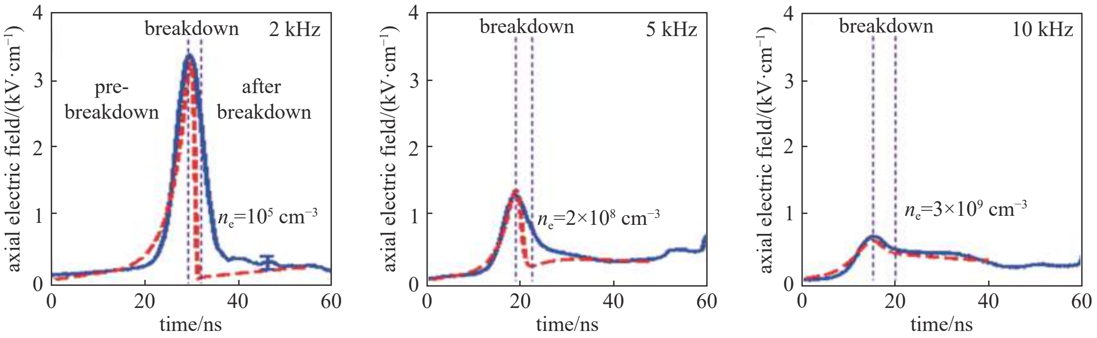


Fig. 11　Effect of pulse repetition frequency on pre-breakdown and post-breakdown axial electric field strengths of fast ionization wave[72]

图 11　脉冲频率对快速电离波击穿前和击穿后轴向电场强度的影响[72]

系，研究了脉冲频率对重复频率亚微秒脉冲氮气流注放电通道演变的影响[73]，如图 12 所示，脉冲频率为 500 Hz 时流注放电出现径向收缩、轴向增长现象，最终形成火花击穿，可能与频率较高时密集空间残余正电荷显著增强正流注前方电场有关。

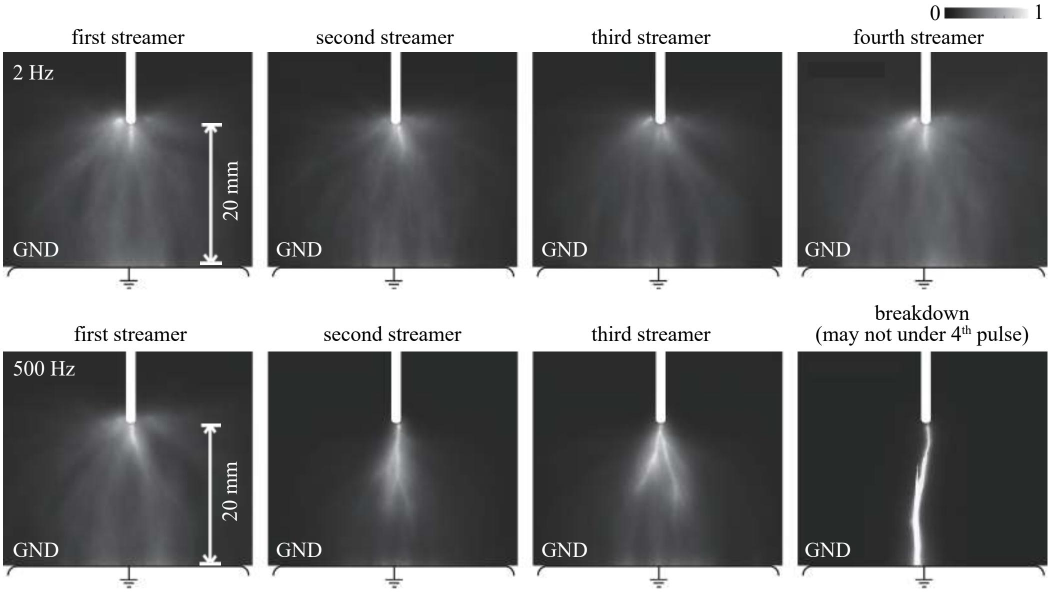


Fig. 12　Evolution tendency of repetitively sub-microsecond pulsed streamer channel in N_2 (gas pressure: 0.2 MPa)

图 12　重复频率亚微秒脉冲氮气流注放电通道的演变趋势（气压：0.2 MPa）

脉冲放电部分能量用于产生振动激励态分子和电子激励态原子和分子，产生快速加热效应[74-76]，加热时间尺度通常小于 100 ns。放电通道温度的快速上升可能引起弱冲击波[77, 78]。由于通道热扩散时间一般远大于脉冲间隔时间，前序放电沉积能量可能触发“通道温升-通道膨胀-气体密度下降-约化场强提高-电子密度增大”放电正反馈增强过程，是重复频率脉冲流注向火花转变的重要原因[14, 44]。

放电记忆效应与波形参数紧密相关，纳秒脉冲放电记忆效应现象和交流放电存在显著差异，由于纳秒脉冲持续时间远低于交流放电，空间电荷密度、电荷迁移程度（包括交流电压极性改变造成的空间电场增强效应等）、沉积能量、空间温度分布、放电通道分布、放电模式转变规律等存在显著差异，例如高频交流电压滑动弧放电通道通常跟随前次放电，而重复频率纳秒脉冲滑动弧放电通道较少跟随前次放电[13]。此外，电源功率限制、电源—等离子体负载匹配特性、放电频率等关键外部参数均会显著影响纳秒脉冲和交流放电记忆效应[13, 79]。

4　结　论

在先进低温等离子体应用迫切需求的驱动下，国内外已发现多种重复频率脉冲流注放电演变现象，对放电演变机制进行了一定探讨。重复频率脉冲流注放电特性呈强非线性特点，放电通常具有明显的渐进式演变特征，放电记忆效应是演变的关键机制，脉冲波形参数是调控重复频率脉冲放电的重要手段。然而，由于重复频率脉冲流注放电演变机制极为复杂，调控重复频率脉冲流注放电演变趋势仍存在较大困难，存在诸多挑战：

国内外研究通常基于单调促进放电发展的记忆效应机制解释重复频率脉冲下气体间隙和气固沿面绝缘强度下降现象。然而，诸多重复频率脉冲放电非线性现象与单调促进放电发展的记忆效应机制（如基于亚稳态粒子）相矛盾。后续研究需进一步考虑真实的“等离子体云”种类、密度、形状等参数，深入研究流注放电与“等离子体云”的相互作用机制。进一步研究重复频率脉冲放电数值模拟方法，解决脉冲施加期和脉冲间隔期物理、化学过程时空尺度差异大等难题。

空间残余电荷输运是重复频率脉冲流注演变的重要机制。目前研究主要基于间接光、电特征变化趋势或求解正负电荷均匀分布等理想情况电荷输运方程。然而，空间残余电荷的初始分布和脉冲间隔期输运行为与脉冲波形参数密切相关，可能偏离空间均匀分布假设，重复频率脉冲流注放电数值仿真缺乏关键诊断参数验证，需进一步基于电场致二次谐波产生等激光瞬态电场诊断手段揭示空间残余电荷输运机制。

重复频率纳秒脉冲放电是典型的过电压放电，约化场强高、物理现象丰富，可能产生高能电子等复杂现象。一方面，高能电子可能影响空间残余电荷、亚稳态粒子等关键记忆效应因子的分布，促进空间弥散放电的形成[80]；另一方面，重复频率脉冲放电导致的空间预电离条件变化可能会改变后续脉冲放电高能电子产生规律，相关耦合机制尚需进一步研究。

现有研究较多从重复频率脉冲放电演变现象和机制入手，较少涉及重复频率脉冲流注放电稳定性调控。约化场强是表征流注放电稳定性的基础性参数，需要进一步研究基于约化场强的重复频率脉冲流注放电不稳定性判据，并从脉冲波形定制、电源输出阻抗等方向研究重复频率脉冲流注放电稳定性调控方法。

目前重复频率脉冲流注放电演变研究较多针对干燥、单一气体种类等理想氛围，而实际低温等离子体应用中常出现潮湿环境、高速气流、气液界面等复杂条件，存在复杂的残余电荷输运和能量弛豫过程，需研究复杂环境中重复频率脉冲流注放电特性和演变机制。

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