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ABSTRACT

The performance of atmospheric pressure dielectric barrier discharge (DBD) driven by dual-frequency sources of 200 kHz/13.56 MHz has been investigated numerically here in a one-dimensional fluid model. By varying the ratio of dual frequency excitation amplitudes, strong nonlinear coupling in the discharge is observed, with the electron density rising significantly due to the synergistic effect governing the ionization process. Moreover, it is shown that the phase shift between the dual frequencies also has an influence on the electron temperature and density. As to different high frequency components, the modulation of the electron temperature and density induced by the phase-shift also varies significantly. Those results then demonstrated the possibility of applying amplitude and/or phase modulations in a wide operation window to optimize the plasma parameters in dual-frequency DBDs with fixed frequencies.

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I. INTRODUCTION

In recent years, nonequilibrium cold atmospheric plasmas (CAPs) have been extensively investigated for various industrial applications, such as materials processing,^{1–6} environment engineering,⁷ plasma chemistry,⁸ and other emerging fields directing plasma treatment on biological samples, such as plasma medicine.^{9–11} Different requirements for plasma parameters are application-specific, according to the different natures of discharge parameters and the response of substrates to processing conditions. For materials processing in general, the goal is to pursue a high plasma density with controllable electron/ion energy and process uniformity over a large area. For biomedical applications particularly, adjustable higher plasma density and gas temperature are prerequisite to realize different purposes such as inactivation, modification, and sterilization.

In comparison with various configurations in generating CAPs, dielectric barrier discharges (DBDs) have shown unique features with higher plasma densities, lower gas temperatures, higher production rate of reactive chemical species, and more effective restriction on discharge currents to obtain stable discharges.^{12,13} Therefore, DBDs of different

configurations are widely studied in various aspects. In conventional DBD systems, a sinusoidal mono-excitation source is usually adopted with a selected frequency range from kilohertz to megahertz regimes.^{14,15} Under a single kilohertz frequency excitation, an increased input power causes a higher plasma density and higher concentration of reactive species. Meanwhile, nevertheless, the resulting higher gas temperature and filamentary discharge with significant spatial nonuniformity is inevitable, restricting its applications in some fields. While increasing the excitation frequency to the megahertz regime, as an effective method to improve the uniformity and increase the density of the plasma, it is usually accompanied by a high gas temperature and a low electron/ion energy, undesirable for certain applications. Thus, it is a great challenge to realize the independent control of plasma parameters by DBDs with the single frequency excitation sources. In order to solve the problem, a dual-frequency modulation facility has been proposed^{16–20} and preliminary works have indicated the feasibility for applications requiring precise independent control of plasma parameters. Such DBD systems operated with additional power supplies driven by different frequencies have attracted extensive attention as a mainstream plasma source for various applications.

In previous dual-frequency DBD system studies, the ratio of the two frequencies is of paramount importance in achieving control over plasma parameters. By means of simulation, Rauf and Kushner²¹ found that at large ratios of two frequencies ($\omega_h \gg \omega_l$, e.g., $\omega_h/\omega_l = 13.56 \text{ MHz}/100 \text{ kHz}$), the spectrum of the two discharge currents was a superposition of the spectra from the two single-frequency sources under certain conditions. On the contrary, when the two frequencies are comparatively different ($\omega_h/\omega_l = 13.56 \text{ MHz}/6.78 \text{ MHz}$), an obvious nonlinear effect was observed. This is indicated by the appearance of higher harmonics and sidebands in the spectrum. By means of experiments, Massines *et al.*²² found the sum and beat frequencies accordingly in the dual-frequency ($\omega_h/\omega_l = 13.56 \text{ MHz}/50 \text{ kHz}$) DBD system. In our previous work involving numerical simulations,^{23,24} we found a coupling sheath structure and sum/beat frequency under the condition of $\omega_h/\omega_l = 13.56 \text{ MHz}/2 \text{ MHz}$. The studies mentioned above positively indicate that the dual-frequency system can effectively influence the plasma parameters based on strong nonlinear effects. However, despite remarkable achievements of studies on the plasma parameter modulation by using appreciate frequency matching, the investigation of amplitudes and phase-shift of the dual frequency excitations on the plasma parameters modulation in such a complex system is still not clear.

In this paper, we report the numerical investigation of plasma electron temperature and electron density control in 200 kHz/13.56 MHz dual-frequency DBD system by applying amplitude modulation. On the basis, an additional modulation of the above key plasma parameters caused by phase-shift effect is explored. The investigation predicts a possible route to realize the plasma parameters modulation with a large operation window in the dual-frequency DBD system with fixed frequencies. In Sec. II, the simulation model is described. Then the results and associated analysis are presented in Sec. III, particularly with regard to the nonlinear coupling effect. The paper is concluded in Sec. IV with summary.

II. MODEL DESCRIPTION

A. Model equations

In this paper, the plasma is generated and sustained between two parallel-plates. Both electrodes are covered by an insulating dielectric layer, and the detailed schematic of this dual-frequency system is shown in Fig. 1. Argon is used as the working gas, and only direct

FIG. 1. Schematic setup of the dual-frequency DBD system modeled in this work.

impact ionization and recombination reactions are considered in the model. Under the atmospheric pressure, collisions between particles are frequent, and, thus, the non-local effect is not prominent.

Hence, a fluid model^{25,26} with semikinetik treatment of electrons^{27,28} is used to describe an atmospheric pressure dual-frequency glowlike DBD system, which provides a decent description of typical nonlinear phenomenon for atmospheric pressure discharge processes in DBD systems, such as bifurcation and chaos.^{29–31} Spatiotemporal distributions of particle species and electric field are obtained by solving the continuity and Poisson's equations as

$$\frac{\partial n_e(x, t)}{\partial t} + \nabla \cdot \Gamma_e = S_e, \quad (1)$$

$$\frac{\partial n_i(x, t)}{\partial t} + \nabla \cdot \Gamma_i = S_i, \quad (2)$$

$$\nabla \cdot (\epsilon E) = e(n_i - n_e) \quad (3)$$

with the divergence of particle fluxes

$$\nabla \cdot \Gamma_e = -n_e \mu_e E - D_e \nabla n_e, \quad (4)$$

$$\nabla \cdot \Gamma_i = n_i \mu_i E - D_i \nabla n_i, \quad (5)$$

where n_e and n_i are the number density of the electron and the i th kind of species, respectively, Γ_e and Γ_i are the flux of the electron and the i th kind of species, E is the electric field, and S_e and S_i are the source terms, and can be expressed as

$$S(x, t) = \mp \alpha \mu_{e,i} E(x, t) n_{e,i}(x, t) - \beta n_{e,i}(x, t). \quad (6)$$

The reaction of electron impact ionization and recombination are taken into account, ϵ is the dielectric permittivity, and e is the elementary charge. The reduced ionization rate α/p and the mobility of charged particles, μ_e and μ_i , are functions of the local reduced electric field E/p ,^{32–35} which are obtained by the Boltzmann equation solver BOLSIG.³⁶ The diffusion coefficients D_e and D_i are obtained from the Einstein relation.³⁷ At the surface of dielectric, the surface charge density is calculated by integrating the charged particle flux density toward the surface during the discharge development as follows:

$$\sigma(t) = e \int ((1 + \gamma_{\text{sec}}) \Gamma_i - \Gamma_e) dt, \quad (7)$$

where γ_{sec} is the secondary electron emission coefficient of the dielectric material. Hence, the flux toward a dielectric surface is written as follows:³³

$$\Gamma_e = -n_e \mu_e E + n_e v_{e,th}/4 - \gamma_{\text{sec}} \Gamma_i, \quad (8)$$

$$\Gamma_i = n_i \mu_i E + n_i v_{i,th}/4, \quad (9)$$

where $v_{e,th}$ and $v_{i,th}$ are the mean thermal speed of electron and Ar^+ . The applied voltage in this paper is adopted by superposing two sinusoidal voltages, $V = V_h \sin(\omega_h t + \phi_h) + V_l \sin(\omega_l t + \phi_l)$. The equations above are then solved by a semi-implicit Scharfetter–Gummel scheme, similar to the methods employed in Ref. 38. It should be noted that the application of 1D model and assumption of discharge mode in this paper are based on the recent experimental investigation,^{20,50} where it was found that the atmospheric pressure DBD system driven by dual-frequency can generate diffuse discharge in argon plasma and exhibit the diffuse form of the plasma in a relatively large domains of applied

TABLE I. The species and associated transport coefficients used in this model.

Species	ε_{th} (eV)	$\mu_p N$	$D_p N$ ($m^{-1} s^{-1}$)	Ref.
e	0	$\mu_p N(\varepsilon)$	Einstein	39
Ar^*	11.55	No charge	6.45×10^{20}	40
Ar^+	15.76	$\mu_{Ar^+} N(E/N)$	Einstein	41
Ar_2^+	14.5	$\mu_{Ar_2^+} N(E/N)$	Einstein	41, 42

parameters. Therefore, in such a situation, the 1D model can be a proper approximation to describe the discharge characteristics and electron dynamic behavior.⁵¹

B. Species and reactions

In the simulation, we adopt argon as the working gas for simplifying operations. Here, four species (e , Ar^* , Ar^+ , Ar_2^+) are considered in the description of the argon chemical kinetics of dual-frequency DBD discharge; the detailed information is listed in Table I.

The total set of reactions taken into account and the rate coefficients are given in Table II, including excitation (R1) and de-excitation (R2), stepwise (R3) and direct ionization (R4), metastable-metastable ionizations [both regular (R5) and associative (R6)], atomic to molecular (R7) and molecular to atomic (R8) ion conversions, dissociative recombination (R9), three-body recombination (R10, R11), and radiative decay (R12).

III. RESULTS AND DISCUSSIONS

For simulations carried out in this work, the driven frequencies are fixed at $\omega_h = 13.56$ MHz and $\omega_l = 200$ kHz at the atmospheric pressure of 760 Torr. The peak voltage at a constant value of $V_{total} = 2000$ V is set, and the amplitude ratio between the two frequency components is varied to study the modulation effect of the amplitudes of the dual frequency excitation on DBD plasma.

The thickness of each dielectric layer is 1 mm with a dielectric constant of 3.4, and the gas gap between the two dielectrically covered electrodes is fixed at 2 mm, where the secondary electron emission

coefficient of the dielectric material is set as 0.01. Initial densities of ions and electrons are assumed to be uniformly distributed in the discharge space at a level of $10^{17} cm^{-3}$. To avoid overheating and make the generated plasma available for the applications sensitive to high temperature, electrode cooling or working gas cooling is commonly used in experiments. And thus, a significant enhancement of electron density would not lead to the increase in gas temperature, which makes it available in the fields of surface modification and synthesis of high-quality functional thin films. Accordingly, the gas temperature of dielectric layer boundary here is fixed at $T_g = 450$ K.^{37,51}

Figure 2 shows the typical temporal evolution profiles of the electrical characteristics for discharges with various voltage ratios. Conventionally, the waveform of a DBD driven by a single kilohertz frequency is presented as one or multiple short pulses that can be observed every half period. As the driving frequency increases, the current pulses are observed to undergo significant distortion and exhibit a sinusoidal waveform. As operating in the dual-frequency configuration, the waveform is superimposed by two frequencies which are dependent on the frequencies between the two interacting excitations and the ratio of their amplitudes, as shown in Figs. 2(a)–2(f). The 3D limit surfaces in the phase space of the current (I), gas voltage (V), and space charge (Q) are illustrated in Figs. 2(g)–2(i). The 3D limit surface is a very useful tool to enable one to characterize and evaluate the nonlinear phenomenon in DBD systems.⁴⁸ Under the single-frequency condition, the 3D limit surface presents a profile of a space-enclosed curve. As to a 2D Q - V cross-section, the surface resembles typical Lissajous profiles. Correspondingly, limit cycle oscillations can be observed in the 2D I - V cross-section of the 3D limit surface. As to the dual-frequency discharge system, the electrical characteristics are more complex. As shown in Figs. 2(g)–2(i), in spite of the individual components with similar waveforms, the 3D limit surface presents vastly different structures with the variation of the ratio of V_h/V_l . This indicates that the characteristics of the plasma parameters have been effectively influenced by means of modulating the ratio.

Figure 3 shows the spatiotemporal averaged variation of the electron density and temperature for different fractions of the high frequency component on the total dual frequency voltage. It is observed

TABLE II. The reactions taken into account and the rate coefficients.

Index	Reaction	Rate coefficient	Ref.
R1	$e + Ar \rightarrow Ar^* + e$	$k_1(\varepsilon)$	39, 43
R2	$e + Ar^* \rightarrow Ar + e$	$k_2(\varepsilon)$	39, 43
R3	$e + Ar^* \rightarrow Ar^+ + e + e$	$k_3(\varepsilon)$	39, 43
R4	$e + Ar \rightarrow Ar^+ + e + e$	$k_4(\varepsilon)$	39, 43
R5	$Ar^* + Ar^* \rightarrow Ar^+ + e + Ar$	$5.16 \times 10^{-6} m^3 \cdot s^{-1}$	44, 45
R6	$Ar^* + Ar^* \rightarrow Ar_2^+ + e$	$4.64 \times 10^{-16} m^3 \cdot s^{-1}$	44, 45
R7	$Ar^+ + Ar + Ar \rightarrow Ar_2^+ + Ar$	$2.5 \times 10^{-43} [300/T(K)]^{3/2} m^6 \cdot s^{-1}$	46
R8	$Ar_2^+ + Ar \rightarrow Ar^+ + Ar + Ar$	$5.22 \times 10^{-16} T(eV)^{-1} \exp(-1.304/T[eV]) m^3$	46
R9	$e + Ar_2^+ \rightarrow Ar^* + Ar$	$7 \times 10^{-13} \cdot [300/T_e(K)]^{1/2} m^3 \cdot s^{-1}$	46
R10	$e + Ar^+ + e \rightarrow Ar + e$	$8.75 \times 10^{-39} \cdot T_e(eV)^{-9/2} m^6 \cdot s^{-1}$	46
R11	$e + Ar^+ + Ar \rightarrow Ar + Ar$	$1.5 \times 10^{-40} \cdot [300/T(K)]^{2.5} m^6 \cdot s^{-1}$	47
R12	$Ar^* \rightarrow Ar + h\nu$	$6.24 \times 10^5 s^{-1}$	42

FIG. 2. The waveform versus different ratios of the voltage for high and low frequencies, with (a)–(c) for the current density; (d)–(f) for the gas voltage; and (g)–(i) for 3D limit surfaces in the phase space of gas voltage, current density and charge density. And each column corresponds to the condition listed on the top.

that the averaged electron density is relatively low when either the high frequency excitation or the low frequency excitation is solely applied. Then, the averaged electron density exhibits a significant enhancement when the contributions of the two applied frequencies are comparable, and the maximum electron density appears where the percentage of the high frequency component is about 70%. In previous studies,^{23,24} it was found that in contrast to the single frequency excitation, several expansion and collapse structures with varying intensity can be observed in the low frequency cycle for the dual-RF DBD. The intensive spots are not simply superimposed of the essential frequencies, but they present a nonlinear enhancement with the local efficient excitation extremes. Thus, besides the driven frequencies of f_l or f_h , there are clearly other frequencies due to a synergistic effect of nonlinear frequency coupling. Compared to the single high frequency driven DBD, the dual-frequency driven discharge is able to tailor the electron

impact excitation structure via nonlinear coupling effect, and the modulation effect is determined by the matching of these dual components. With an appropriate application of the amplitude of the dual components, the electron density can be upraised significantly. On the other hand, an inverse trend is observed for the averaged electron temperature. The results demonstrate that the spatiotemporal averaged electron density and temperature can be modulated by varying the amplitude ratio of the high and low frequency voltages. The technique may provide a possible approach to precisely tailor the electron density/temperature of discharge⁴⁹ of the DBD system.

Figure 4 shows the spatiotemporal profiles of the electron power absorption ($P_{e,abs} = J_e \cdot E$) in four low-frequency periods under the same parameters. The similar result has been observed in experiments under comparable discharge conditions.⁵⁰ It is seen that the amplitude variation of dual frequency components results in different electron

FIG. 3. Spatiotemporal averaged electron density and temperature as a function of the high frequency contribution fraction with $V_{\text{total}} = 2000$ V.

power absorption structures. As shown in Figs. 4(a) and 4(b), most of the electric field power dissipates in the plasma bulk. And the power absorbed by electron increases significantly with a stronger high frequency component, as shown in Fig. 4(a), since the high frequency power is more efficient to be coupled into electrons. Besides, the profile of the electron power absorption also changes, indicating the electron heating mechanism undergoing a variation since the electron kinetic behavior governing the discharge does not remain the same. Moreover, it is worth mentioning that in both of these two cases in Fig. 4, the electron power absorption distribution in each low frequency period is not the same, showing a very slow modulation, with a characteristic time scale longer than the period, on the power absorption pattern due to the non-linear coupling.

The detailed high frequency oscillation cannot be seen in Fig. 4. In order to provide further insights into the phenomena, we in Fig. 5 show the electric field power absorbed by electrons within a single low-frequency period, together with the conducting current density,

gas voltage, and excitation intensity. In Fig. 5(a), the high frequency component is dominant. It is observed that the intensity of major power absorption structures is strengthened apparently. And the conducting current is continuous with a profile similar to that of the gas voltage due to the fast oscillating high frequency electric field, representing classical high frequency characteristics. On the other hand, in Fig. 5(b) where the low frequency component plays a dominant role, the major power absorption pattern is discrete and clearly separated from each other. Moreover, the profile of the conducting current presents typical low-frequency DBD characteristics since the “memory effect” plays an important role in the discharge propagation for the low frequency dominated system. Also, the multipeak structures are observed, accompanied by the synchronous discharge emission. Correspondingly, by tailoring the averaged electron power absorbance, the electron density is modulated effectively, as illustrated in Fig. 3.

In order to further study the nonlinear coupling phenomena in dual-frequency DBDs, the temporal evolution of the total discharge current density J under different ratios of voltage amplitudes are analyzed by fast Fourier transform (FFT), shown in Fig. 6.

For the high frequency component to be dominant, the characteristic spectrum is shown in Fig. 6(a). And several main peaks can be observed within the frequency range. Besides the eigenfrequency of the two excitation sources, the rest of them in the spectrum are located at about $\omega_1 = 27.15$ MHz, $\omega_2 = 40.7$ MHz, $\omega_3 = 54.26$ MHz, and $\omega_4 = 67.8$ MHz, respectively. They are in fact the higher harmonics of $\omega_h = 13.56$ MHz ($2\omega_h = 27.12$, $3\omega_h = 40.68$, $4\omega_h = 54.24$, and $5\omega_h = 67.80$ MHz precisely), as also be observed in experiments.²² Besides, the spectrum of the low frequency range is modulated with broad sidebands around $\omega_l = 200$ kHz, a phenomenon of nonlinear coupling also reported in atmospheric pressure plasma jet experiments based on DBDs. In comparison, Fig. 6(b) shows the spectrum in a low frequency dominated process. It is found that only two peaks at $\omega_h = 13.56$ MHz and $\omega_l = 200$ kHz appear in the FFT spectrum, without the higher harmonics of ω_h , showing a weak nonlinearity in the high frequency band. However, the sidebands in the low frequency regime around 200 kHz still exist. Combining the analysis of electron density, power absorption characteristics described above, and the comparison with other relevant report,⁵¹ it has been shown

FIG. 4. Spatiotemporal profiles of electron power absorption in four periods of the low frequency with (a) $V_h = 1600$ V, $V_l = 400$ V; and (b) $V_h = 400$ V, $V_l = 1600$ V.

FIG. 5. The electric field power absorbed by electrons within one low-frequency period, together with the conduction current density, gas voltage and excitation intensity. (a) $V_h = 1600$ V, $V_l = 400$; (b) $V_h = 400$ V, $V_l = 1600$ V.

preliminarily that the nonlinear coupling effect caused by the amplitude matching of dual frequency excitations plays a key role in FFT spectrum variation. In general, the nonlinear effect of plasma discharge process is an easily observable phenomenon, especially for DBD systems. Such typical dissipative systems with strong nonlinearity, i.e., period-doubling bifurcation, quasi-periodic/intermittent oscillations, and other chaotic behaviors, are considerably complicated and, thus, research focuses of great interest.

FIG. 6. FFT spectrum of the total discharge current density with different ratios of the dually applied amplitudes, with (a) $V_h = 1600$ V, $V_l = 400$; and (b) $V_h = 400$ V, $V_l = 1600$. The embedded part in panel (a) is the detailed information marked with red lines.

Figure 7 shows the greyscale contour of spatiotemporally averaged electron temperature and density as functions of the high frequency component and phase-shift of the two frequencies. The results preliminarily indicate that it is possible to control the electron temperature and density precisely in the dual-frequency DBD system by amplitude and phase modulations. From Figs. 7(a) and 7(b), it is found that with a fixed phase-shift, the corresponding highest electron temperature appears when either frequency excitation is solely applied (as seen in regions I and II), and a major change is observed when the fraction of the high frequency component is $\sim 50\%$ – 75% , with the lowest electron temperature occurring in region III. In general, the variation of the electron temperature presents an inverse tendency in comparison with the electron density. However, as to different high frequency components, the modulation of averaged electron temperature and density caused by phase-shift effect varies significantly, predicting a method for realization of a large operational window to optimize the plasma parameters by a particular high frequency component applied.

IV. CONCLUSIONS

In this paper, a self-consistent fluid model with semi-kinetic treatment of electrons has been used to study the amplitude and phase modulations of plasma parameters in dual-frequency atmospheric pressure DBDs. The conclusions can be summarized as follows:

- (1) By modulating the amplitude ratio of two excitation sources, the averaged electron density and temperature are effectively influenced. And there exists a significant nonlinear coupling effect during the discharge process due to the dual-frequency interaction, and with an appropriate application of the amplitude ratio of these dual components, the electron density can be upraised significantly.
- (2) The investigation of electric field power absorbed by electrons, together with the conducting current density, gas voltage, and excitation intensity, has shown that the electron density and temperature are modulated effectively by tailoring the averaged electron power absorbance.

FIG. 7. Contour plot of (a) the electron temperature and (b) density as functions of the high frequency component and phase shift of the dual frequencies.

- (3) The temporal evolution of the total discharge current density under different ratios of voltage amplitudes is analyzed by the FFT spectrum, and the nonlinear coupling effect plays a key role in leading to the spectrum variation.
- (4) On the basis, an extra modulation by means of phase-shift is explored. A tuning of the phase differences between the dual frequency excitation sources also has a direct influence on the averaged electron density and temperature, which enables one to optimize plasma parameters with a large operation window.

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