Experimental analysis on the nonlinear behavior of DC barrier discharge plasmas

Dogan MANSUROGLU1,2,3 and Ilker Umit UZUN-KAYMAK2

1 Physics Department, CanakkaleOnsekiz Mart University, Canakkale 17100, Turkey
2 Physics Department, Middle East Technical University, Ankara 06800, Turkey
E-mail: mansurdogan@gmail.com and mansuroglu@comu.edu.tr

Received 24 May 2016
Accepted for publication 7 September 2016
Published 23 November 2016

Abstract
Nonlinear behavior of glow discharge plasmas is experimentally investigated. The glow is generated between a barrier semiconductor electrode, Chromium doped namely Gallium Arsenide (GaAs:Cr), as a cathode and an Indium–Tin Oxide (ITO) coated glass electrode as an anode, in reverse bias. The planar nature of electrodes provides symmetry in spatial geometry. The discharge behaves oscillatory in the time domain, with single and sometimes multi-periodicities in plasma current and voltage characteristics. In this paper, harmonic frequency generation and transition to chaotic behavior is investigated. The observed current–voltage characteristics of the discharge are discussed in detail.

Keywords: DC glow discharges, nonlinear behavior, digital signal analysis

1. Introduction
The glow discharge plasmas attract a great deal of attention in many areas such as plasma processing and deposition, glow discharge detectors, display panels, plasma diodes etc [1–4]. Laboratory plasmas are also interesting to study nonlinearity and chaos since they may include periodically linear characteristics in equilibrium at first, followed by a disruption of the equilibrium, which may cause the discharge to present nonlinear characteristics. The simplest and the most convenient experimental setup to study nonlinearity would be a semiconductor barrier discharge system which is discussed in this paper. A barrier semiconductor discharge system also resembles a photographic system, in which one of the electrodes is used as a semiconductor photodetector. Some patterns are produced as a result of electrically unstable interactions and non-uniform ionization between the electrodes [5–7].

The barrier glow discharges are thoroughly investigated for various semiconductors used for electrodes, for example, doped and undoped versions of Gallium Arsenide (GaAs:Cr, GaAs), Silicon doped with materials such as Zinc, Platinum, Gold (Si:Zn, Si:Pt, Si:Au), and Germanium (Ge) [5–10]. However, these investigations are often limited to the characterization of the materials used. Due to its reactive nature, employing a semiconductor as a barrier creates a time constant on the discharge [11]. The barrier discharge is also discussed for different gases such as Neon [12], Helium [13, 14], and Nitrogen [14, 15]. It is found that nitrogen is an attractive gas by providing a discharge at a lower current value compared to other inert gases at higher pressure [15]. It should also be noted that, in a barrier discharge, the characteristics of the discharge significantly depend on both the thickness of the semiconductor as well as the thickness of the plasma. On the other hand, the dynamics of the ionization play an important role in the formation of these oscillations, so called current instabilities causing nonlinear behavior [8]. In this study, the emphasis of the research is mainly on the frequency domain analysis of the oscillatory current and voltage characteristics.

In this work, we focus on the experimental analysis of the nonlinear behavior formed in a dc barrier glow discharge using a semi-insulating (SI) GaAs:Cr material as the barrier. The other electrode is a glass plate coated by conductive Indium–Tin Oxide (ITO). The GaAs is a promising material with the properties of high carrier mobility, wide band sensitivity and large absorption coefficient [16]. The features of
the discharge are explored using plasma current \((I)\)–voltage \((V)\) characteristics by acquiring long time series data using a fast oscilloscope. The discharge current behavior is investigated under a parametric scan of applied voltage. The results show that the nonlinear behavior of the discharge is caused by instabilities forming multi oscillations in the discharge current. Power spectral analysis of the current oscillations shows the formation of sharp peaks and sometimes multi frequency oscillations whereas in a chaotic regime these peaks are replaced with higher frequency broadband structures.

2. Experimental setup

The barrier glow discharge system consists of two planar electrodes separated by a narrow gap. The electrodes are accurately positioned using a holder placed inside a vacuum chamber. The Si Chromium doped GaAs wafer of thickness 350 \(\mu\)m is used as the cathode. The resistivity of the cathode ranges in between \(1.5 \times 10^8\) and \(2.5 \times 10^8\) \(\Omega\) cm. One side of the GaAs wafer is coated with gold thin film to obtain a uniform electrical contact with the high voltage supply. The anode is a glass plate coated with ITO, which is transparent and conductive. The diameters of the cathode and anode are about 50 mm and 35 mm, respectively, and the gap between the electrodes is kept at 1 mm using a spacer. The spacer is made out of Teflon to prevent arcing in the vacuum region between the electrodes. The pressure in the discharge system is first dropped to a base pressure of 1.33 Pa via a rotary pump and then the discharge gap is filled with \(N_2\) gas at a fixed pressure of 4 kPa.

The electrodes are connected to a DC high voltage power supply with a load resistor \(R_0\) in series to limit the current of the discharge. Another resistor, \(R_1\) (about 100 \(\Omega\)), shown in figure 1, is connected in series between the electrodes and the HV source to record the current flowing through the circuit [17]. A voltage divider with a ratio of 1/1000 is implemented on the system to record voltage. Two serial resistors of \(R_2\) and \(R_3\) are connected parallel in the circuit as shown in figure 1, where \(R_1 \ll R_3\), and the voltage is recorded from resistor \(R_3\) by using a fast sampling oscilloscope. By applying a voltage difference between the electrodes, the glow discharge is initiated mainly due to the Townsend \(\alpha–\)ionization [11, 18]. The resistivity of the GaAs wafer is lowered as the material is irradiated. In literature, this process is known to excite electrons in GaAs, hence causing a transition from the valence band to the conduction band of the wafer, generating additional radiation in the gap [8]. This behavior of the wafer is often modeled as a reaction–diffusion system creating a time constant on the discharge [11, 19]. Therefore, the system can be reviewed as a primary oscillation where an increase in the plasma current is associated with a decrease in the voltage between the electrodes. As it decays and surpasses below a certain critical value, the current starts to decay as well, leading to an increase in the voltage, hence generating the glow back. Current density is expected to be proportional to the radiation intensity of the glow. Therefore, oscillations in plasma current can be considered as filament and pattern formations on the electrodes. These spatial formations can also be observed optically as well [8].

It is important to note that, various types of light sources, such as halogen lamps, LEDs operating at specific wavelengths etc are implemented as an additional irradiation source in the experimental setup. However, the observed oscillations in the current and voltage measurements seem to be unaffected by this type of additional irradiation. It is reasonable to suspect that the strong spectral lines of the neutral and single ionized nitrogen dominate the irradiation process [20]. The transmission of additional radiation sources may also be reduced due to thick bk7 windows used in the vacuum chamber.

During the next section, the long data sets acquired using a fast oscilloscope, i.e. LeCroy 6100 are analyzed in both the time-domain and the frequency domain. The voltage is scanned in small increments of 20 V starting from the breakdown voltage up to 1.3 kV. At each point in the voltage scan, the discharge current and voltage are recorded as time series data. Based on the resulting data sets, the discharge...
characteristics are discussed. The plasma current exhibits single frequency oscillations at first, however as the voltage increases multi-oscillatory behavior takes over. The coupling between various frequencies indicates that the instabilities in the discharge are driving the system. In earlier studies, the presence of the oscillatory instabilities has been shown in several gas discharge systems by investigating Turing- and Hopf-type bifurcations [8, 21]. In this paper we are investigating a similar system with the emphasis on signal processing to understand mode coupling and chaotic formation. The frequency and coherence spectra are analyzed in detail using the characterization of the cross power spectral density (CPSD), obtained for both single and multi-oscillation phases of the plasma.

3. Measurements and signal analysis

The circuit of the system behaves like a resistor–capacitor (RC) circuit including a load resistor, where the discharge gap acts as the capacitor. In order to determine the impedance of the plasma, the current measurements are collected via scanning different $R_0$ and applied voltage values. From the measurements illustrated in figure 2, the impedance of the plasma is estimated on the order of 1 MΩ and the discharge current values are found in the range of milliamperes (mA), which are compatible with the values measured by Purwins et al [7]. When the scanning $R_0$ is smaller than 1000 kΩ, it is hard to sustain a stable current. For $R_0$ values greater than 8000 kΩ the plasma cannot be generated since all the power is now spent on the load resistor.

The applied voltage scan is started at 470 V, and both the plasma current and plasma voltage data are sampled at 1 MS$^{-1}$ and recorded for a duration of about 4 s. When the gas pressure is about 4 kPa and the electrode separation is at 1 mm, the plasma breakdown is observed at the applied voltage of 480 V, which is consistent with the Paschen curve for N$_2$ gas discharge [22]. At the lower end of the voltage scan, a faint glow is observed in the discharge gap. Due to the semiconductor barrier, the discharge current is observed to be oscillating which in return causes an oscillation in the plasma voltage. It appears that the fluctuations in plasma current cause the plasma voltage to drop, which in return cause a decay in the current fluctuations. The rise time of these current fluctuations is rather fast, about 1 μs, whereas the fall time is observed to be about 3 μs. The plasma exhibits single frequency oscillations for the applied DC voltage in the range of 480 V < $V_{\text{app}}$ < 640 V. Within this range of operations, the peak plasma current is increasing and the oscillations appear to be more frequent as the applied voltage increases, as shown in figures 3(a)–(c). This effect can also be observed optically in terms of an increase in radiation intensity [8]. In the range of 640 V < $V_{\text{app}}$ < 780 V, these observed homogeneous single oscillations evolve to higher frequencies, moreover, the plasma starts to exhibit oscillations at harmonic and sub-harmonic frequencies, as seen in figures 3(d) and (e). A second oscillation is obtained along the fundamental oscillation with smaller frequency ($f = f_0$/2) and the amplitude of this secondary oscillation is substantially smaller compared to the amplitude of the fundamental peak oscillating at $f = f_0$. For applied voltages above 780 V, the discharge exhibits oscillations at even higher frequencies. As the applied voltage increases, the fundamental frequency of oscillations increases and the total number of sub-harmonics also increases. In other words, the plasma presents multi-oscillations with the variable oscillation frequency and amplitude with different order, as illustrated in figure 3(f).

In addition, by imposing the time series data for both plasma current and voltage on the same graph, the phase diagram plot is obtained. In figure 4(a) the current–voltage phase diagram is plotted for single frequency oscillations. Figure 4(b) shows the current–voltage phase diagram with a thickened limit cycle line for an applied DC voltage of 700 V. This observation is consistent with the sub-harmonic
frequency oscillations observed at the time series data as shown in figure 3(d). Likewise, the current–voltage phase diagram plotted for 990 V of applied voltage shows multi loops as shown in figure 4(c), indicating some nonlinear formation in the system. These types of oscillations are also known as the mixed-mode oscillations and the nonlinear behavior is often explained in terms of current instabilities in the discharge depending on the ionization characteristics [23].

To investigate such oscillatory characteristics, the frequency domain analysis of the experimental data is conducted using the MATLAB® Signal Processing Toolbox. The time series data for both plasma current and plasma voltage are analyzed by calculating the CPSD and associated coherence levels. Frequency domain data is defined by applying the Fourier transform to the time series data for a given number of points, known as nfft [24]. Using the results for the current signal, \( I(f) \), and voltage signal, \( V(f) \), the CPSD is given by the formula:

\[
CP_{IV}(f) = \langle I(f) V^*(f) \rangle = \frac{1}{L} \sum_{k=1}^{L} I_k(f) V^*_k(f),
\]

where \( L \) is the number of partitions generated using a partition length nfft for a given length of time series data, whereas \( k \) represents the partition number. The CPSD calculation characterizes the cross-correlation between the plasma current and voltage thereby noise in the observed experimental data is eliminated almost completely due to extensive ensemble averaging. Coherence levels of the observed peaks in frequency domain can be investigated using the formula:

\[
coh_{IV}(f) = \frac{\langle I(f) V^*(f) \rangle}{\sqrt{\langle I(f)^2 \rangle \langle V^2(f) \rangle}}.
\]
The results of CPSD analysis and coherence are illustrated in figure 5 for different applied voltages. Figure 5(a) indicates only one oscillation with fundamental frequency around 8.1 kHz, and the coherence of the related spectrum is shown in figure 5(b). As the applied voltage increases the fundamental frequency of the oscillations also increases. Figure 5(c) shows the fundamental frequency rises to 19.8 kHz and also the sub-harmonic frequency, i.e. \( f_0/2 \), appears as a broad structure. Moreover, these frequency values shift even more for higher voltage ranges. The system exhibits the multi oscillatory states at different frequencies as seen in figure 5(e). Two intense oscillations are observed at 21.9 kHz and 43.8 kHz, and a weak oscillation at 32.8 kHz. Although previous studies show frequency peaks appearing at the order \( f_0, f_0/2, f_0/3 \) [8], in our experiments, such ordering is not observed. Instead, sub-harmonic formations, and shifting towards higher frequencies ride the system to a chaotic level. Moreover, this sub-harmonic formation is observed at frequencies \( f = nf_0/2 + mf_0/4 \) where \( n = 1, 2, \ldots \) and \( m = 0, 1, 2, \ldots \) are integers.

Interestingly, the coherence spectrum shows that the fundamental frequency and its harmonics are highly coherent, almost 95% and above. The sub-harmonic frequencies appear to have coherence levels at the noise floor at first, see figure 5(c), however, as the applied voltage increases, they will reach coherence levels near 85% and above, as seen in figure 5(e). The evolution of these modes and their coherence levels are linked to the increase in applied voltage. Therefore, it is suspected that as the voltage increases the current fluctuations are increasing in amplitude at first, until they become unstable, so they burst into many sub-harmonic modes meanwhile they remain highly coherent. To investigate these mode couplings, further analysis is needed. One approach to investigating nonlinear mode coupling would be to calculate higher order moments of the time series data [25].

4. Conclusion

In this study, the nonlinear harmonic generation is investigated by gradually scanning the applied voltage in a barrier glow discharge system utilizing a GaAs:Cr semiconductor as the barrier electrode. Long time series data of the discharge current and plasma voltage are collected using a fast oscilloscope at each applied voltage parameter. The discharge shows homogeneous single oscillatory peaks with their harmonics at low applied voltages. The fundamental oscillation amplitude also increases with increasing applied voltage.
However, as the voltage increases above a certain threshold, these oscillations evolve into sub-harmonic formations and multi-periodicities are observed.

The frequency spectrum of data is analyzed by calculating the CPSD using plasma current and plasma voltage time traces. Results show that the fundamental frequency is shifting towards higher frequencies with increasing applied voltage. Harmonics of the fundamental frequency ($f_0$) are ubiquitous in all data sets, however, the formation of sub-harmonic frequencies and combination of sub-harmonic frequencies formalized as $f = n f_0/2 + m f_0/4$ only appear by increasing the applied voltage above a certain threshold. Moreover, the power spectral density is used to calculate the coherence levels of the frequencies. These levels show that the harmonic frequencies are highly coherent while the sub-harmonic are obtained with lower levels, thus, it can be suggested that the existence of the energy transition in the system. And the nonlinearity occurring in the system can be explained by nonlinear frequency coupling among the oscillations. The cross power spectral estimates and coherence

Figure 5. For the applied voltages of (a)–(b) 580 V, (c)–(d) 700 V, and (e)–(f) 990 V; CPSD and coherency spectra are shown. (The calculations are conducted with the data length of 1MS, and the nfft is taken 2048).

However, as the voltage increases above a certain threshold, these oscillations evolve into sub-harmonic formations and multi-periodicities are observed.
calculations are only calculated using second order moments, in other words the linear part of the signal analysis. To pinpoint the nonlinearity, further studies are needed investigating more extensive data sets using the higher order moment analysis.

Acknowledgments

This research is supported by the Scientific Research Project Fund of Middle East Technical University, under project # BAP-08-11-2016-044.

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