Electric field in an AC dielectric barrier discharge overlapped with a nanosecond pulse discharge

Benjamin M Goldberg, Ivan Shkurenkov, Igor V Adamovich and Walter R Lempert

Michael A Chaszeyka Nonequilibrium Thermodynamics Laboratory, Department of Mechanical and Aerospace Engineering, The Ohio State University, Columbus, OH 43210, USA

E-mail: b.goldberg@princeton.edu

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Abstract
The effect of ns discharge pulses on the AC barrier discharge in hydrogen in plane-to-plane geometry is studied using time-resolved measurements of the electric field in the plasma. The AC discharge was operated at a pressure of 300 Torr at frequencies of 500 and 1750 Hz, with ns pulses generated when the AC voltage was near zero. The electric field vector is measured by ps four-wave mixing technique, which generates coherent IR signal proportional to the square of electric field. Absolute calibration was done using an electrostatic (sub-breakdown) field applied to the discharge electrodes, when no plasma was generated. The results are compared with one-dimensional kinetic modeling of the AC discharge and the nanosecond pulse discharge, predicting behavior of both individual micro-discharges and their cumulative effect on the electric field distribution in the electrode gap, using stochastic averaging based on the experimental micro-discharge temporal probability distribution during the AC period. Time evolution of the electric field in the AC discharge without ns pulses, controlled by a superposition of random micro-discharges, exhibits a nearly ‘flat top’ distribution with the maximum near breakdown threshold, reproduced quite well by kinetic modeling. Adding ns pulse discharges on top of the AC voltage waveform changes the AC discharge behavior in a dramatic way, inducing transition from random micro-discharges to a more regular, near-1D discharge. In this case, reproducible volumetric AC breakdown is produced at a well-defined moment after each ns pulse discharge. During the reproducible AC breakdown, the electric field in the plasma exhibits a sudden drop, which coincides in time with a well-defined current pulse. This trend is also predicted by the kinetic model. Analysis of kinetic modeling predictions shows that this effect is caused by large-volume ionization and neutralization of surface charges on the dielectrics by ns discharge pulses. The present work demonstrates that this effect may be used to control the phase of AC barrier discharges, triggering reproducible breakdowns at well-defined moments.

Keywords: nanosecond pulse discharge, AC dielectric barrier discharge, electric field measurements, four-wave mixing, kinetic modeling

(Some figures may appear in colour only in the online journal)

1. Introduction

Over the last two decades, high pressure dielectric barrier discharges (DBD) have been studied extensively due to their significant potential for plasma flow control [1–3], ozone generation [4], removal of pollutants from air and flue gas [4], material surface treatment [4], plasma assisted combustion [5], plasma generation near a liquid-vapor interface [6], and biomedical applications [7]. In most of these applications, the discharge is sustained by an AC electric field, generating the
plasma near dielectric surfaces or across short gaps between dielectric-covered electrodes, although near-surface and volumetric barrier discharges sustained by short duration (ns and sub-μs), high peak voltage pulses are becoming more common. In AC barrier discharges, quasi-diffuse plasma typically consists of a large number of spatially and temporally random ‘micro-discharge’ streamers, self-terminating due to surface charge accumulation on the dielectric and plasma self-shielding, which limits the electric field in micro-discharges [8]. Plasma self-organization and instability development at high pressures, high AC frequencies, and high peak voltages result in formation of filamentary structure of the discharge [9, 10]. Spatially (but not temporally) random streamers have also been detected in ns pulse barrier discharges, although in this case rapid voltage rise and residual ionization from previous pulses may generate streamer-free diffuse volumetric plasmas [11] and nearly uniform large-surface area plasmas [12] at high pressures. Similar to AC barrier discharges, ns pulse discharges develop instabilities at high pressures, high pulse repetition rates, and high peak voltages, resulting in formation of filamentary structure of the plasma [13, 14], although in this case the instability onset is suppressed due to a very low duty cycle.

Both in AC and in ns pulse barrier discharges, charging of dielectric surfaces is a critical process limiting the electric field and terminating electron impact ionization in the plasma (in a ‘micro-discharge’ streamer, surface ionization wave, or volumetric ionization wave). In volumetric ns pulse discharges, residual surface charge density on the time scale greatly exceeding pulse duration may be quite low [15]. In this case, surface charges are readily neutralized by transport of charged species from the plasma volume when the applied voltage is rapidly reduced, due to high peak electron/ion density in the diffuse plasma [15]. However, in surface ns pulse discharges exhibiting strongly filamentary structure, especially with long isolated filaments, surface charge accumulation may be quite significant [16]. In AC barrier discharges, residual surface charge accumulation is also a significant factor, since charged species transport in the self-shielded, decaying plasma in micro-discharge streamers, without rapid changes in the applied electric field, is obstructed. In this case, surface charge accumulation, on a much longer time scale compared to the AC period, may significantly affect discharge parameters [17]. Specifically, this effect may limit considerably the electric field in the near-surface plasma and the electrostatic body force in AC DBD surface plasma actuators used for plasma flow control [18]. This effect may also reduce the energy coupled to the plasma in AC barrier discharges used for gas phase plasma chemistry, surface treatment, and biomedical applications.

To mitigate this effect, several approaches have been used, such as a ‘diode’ dielectric surface (with electrical conductivity in only one direction) [19], or a weakly conducting (e.g. liquid layer covered) surface [16], to slowly ‘bleed’ the surface charge to the ground. One of the approaches was to combine the high-voltage AC waveform with a series of high-voltage, ns duration pulses in a surface plasma actuator operating in quiescent room air [18]. Schlieren flow visualization demonstrated that this technique resulted in significant improvement of actuator performance (such as an increase of entrained flow velocity), which was attributed to reducing surface charge accumulation in the AC discharge by ns discharge pulses and enhancing electrostatic body force. It was also suggested that using alternating polarity of ns voltage pulses may enhance this effect. Since the main focus of [18] was on flow diagnostics, and plasma diagnostics capabilities were limited, the effect of ns pulses on the electric field in the AC barrier discharge has not been quantified.

The main objective of the present work is to provide quantitative insight into the effect of ns barrier discharge pulses on the AC barrier discharge, using time-resolved measurements of the line-of-sight averaged electric field in the discharge gap and kinetic modeling. Since access of laser diagnostics to surface barrier discharge plasmas, such as studied in our previous work [20] is quite challenging, in the present work we are studying a volumetric barrier discharge sustained between two parallel, dielectric-covered electrodes. The electric field in the AC discharge between two parallel plate electrodes covered by dielectric plates may be inferred from the discharge voltage and current waveforms based on the equivalent circuit parameters, e.g. using a method developed in [21–23]. In the AC discharge enhanced by ns pulses, accurate determination of equivalent circuit parameters (i.e. capacitive and inductive) from voltage and current waveforms is challenging, both due to fairly complex external circuit used and due to significant difference between the AC and ns pulse currents (up to 4 orders of magnitude). Therefore in the present work, we apply a laser diagnostic technique known as electric field four-wave mixing, developed by Gavrilenko et al [24], similar to coherent anti-Stokes Raman scattering (CARS) [25]. Basically, in the four-wave mixing technique, the probe beam in a conventional CARS experiment is replaced with an externally applied field which induces a dipole moment in the normally non-polar molecules. A collinear, phase-matched pair of pump and Stokes beams are used to generate coherent excitation of the molecules with the induced dipole moment, allowing them to radiate coherently in the phase matching direction (collinear with the pump and Stokes beams), at the wavelength controlled by the energy difference between the excited state and the ground state. In the present work, H$_2$ molecules excited from the ground vibrational level, $v = 0$, to the vibrational level $v = 1$, radiate on the $v = 1 \rightarrow 0$ Q(1) line in the IR. The intensity of the coherent IR radiation scales proportionally to the square of the external electric field. To date, most of the electric field measurements using this technique were completed in hydrogen [26–30] due to its large Raman cross section, although recent work has been done in nitrogen as well [31–33]. The principal advantage of ps four-wave mixing method is the capability for measurements of spatially resolved electric field vector in transient discharges of complex geometry [15], as well as sub-ns time resolution [20].

2. Experimental

Figure 1 shows a schematic of the experimental setup used for electric field measurements by picosecond four-wave mixing, described in detail in our previous work [15, 20]. Key
benefits of picosecond four-wave mixing are much higher signal to noise, due to much higher ps laser intensity compared to ns lasers of comparable pulse energy, and sub-ns temporal resolution. Briefly, the second harmonic (532 nm) of an Ekspla PL2143A Nd:YAG laser, with a pulse duration of 30 ps and maximum pulse energy of ~22 mJ is focused using an f = 750 mm focal distance lens into a high pressure stimulated Raman shifting (SRS) cell filled with 7 Bar of hydrogen. In the cell, a Stokes beam (683 nm) and an anti-Stokes beam (435 nm) are generated collinearly with the residual pump beam. The use of a high-pressure SRS cell, rather than a dye laser, to generate the Stokes beam greatly simplifies the experimental setup and provides excellent alignment stability. The anti-Stokes beam is filtered out using a pair of blue reflecting dichroic mirrors, as shown in figure 1. A small amount of the pump and Stokes beams are reflected off a glass plate and separated using a Pellin–Broca prism, to monitor the pump beam intensity and to correct for the effect of shot-to-shot fluctuations of the laser beam energy on the IR signal.

The residual pump beam and the Stokes beams are recollimated (by an f = 500 mm lens) and then focused (by an f = 500 mm lens) into the gap between two plane circular electrodes (halfway between the electrodes, along the axis of symmetry), placed within the discharge cell filled with hydrogen at 300 Torr. As discussed in section 1, the collinear pump and Stokes beams generate coherent oscillating polarization of the H$_2$ molecules. In the presence of the induced dipole moment due to the externally applied electric field, the polarized molecules radiate coherently in the phase matching direction, collinear with the pump and Stokes beams, at the H$_2$(v = 1 → 0) Q(1) transition at 2.406 μm. An anti-Stokes beam is also generated in the discharge cell, collinearly with the pump and Stokes beams, and is used for calibration of the electric field measurements, as discussed below. The volume where the IR and the anti-Stokes CARS signals are generated, in collinear phase matching geometry, has been measured to be a 2 cm × 150 μm cylinder, located on the centerline of the discharge gap. After the discharge cell, the IR signal beam is spectrally separated from the collinear pump, Stokes, and anti-Stokes beams, using dichroic mirrors which efficiently transmit IR radiation while reflecting visible light, and a 1/3 m spectrometer (see figure 1). The anti-Stokes beam is also separated from the residual pump and Stokes beams, using additional dichroic mirrors, same as the mirrors placed after the SRS cell.

The pump beam intensity and the CARS beam intensity are monitored using standard PIN photodiodes. The IR signal intensity is monitored using a thermoelectrically cooled HgCdTe detector. Orthogonal field vector components are separated using a Glan–Thompson polarizer placed in front of the IR detector (see figure 1), as discussed in our previous work [20]. All three beam intensities are recorded by a LeCroy WaveRunner 104 MXi-A digital oscilloscope with 1 GHz bandwidth. The electric field is inferred from the intensities of the pump, anti-Stokes, and IR signal beams, as discussed below. The IR signal and CARS signal beam intensities are given as

$$I_{IR} \sim \chi_{II}^2 \frac{I_{Pump} I_{Stokes}}{E^2},$$

$$I_{CARS} \sim \chi_{CARS}^2 \frac{I_{Pump} I_{Stokes} I_{Probe}}{I_{CARS}^2 \chi_{Pump} I_{Stokes}},$$

where $E$ is the electric field, $I_{probe} = I_{pump}$ since the 532 nm pump beam also serves as the CARS probe beam, and $\chi_i$ are third order susceptibilities for four-wave mixing and CARS processes, respectively. From equations (1) and (2),

$$|E_{Em}| = \sqrt{\frac{A \cdot I_{IR} I_{Pump}}{I_{CARS}}},$$

where $A$ is a constant determined by calibration, by measuring the known electric field between the electrodes below the breakdown limit. Individual components of the electric field vector were determined by changing the polarization of the laser beams and of the IR signal collected by the HgCdTe detector.
detector by placing a Glan–Thompson polarizer in front of the detector, as discussed in greater detail in [20].

For the collinear phase-matching geometry used in the present work, the spatial resolution of the present diagnostics in the direction along the laser beams is approximately 2 cm, determined by traversing a thin glass plate (a microscope slide) along the focused laser beams and measuring the non-resonant background signal. The spatial resolution in the direction perpendicular to the laser beams is approximately 150 μm, measured by traversing a razor edge across the beam waist and measuring the intensity using a photodiode. The time resolution is limited by the coherence decay time in H₂ molecules, ~200 ps. However, the pump laser used in the present work (different from the laser used in [15, 20]) has significant jitter, several microseconds. For this reason, time-resolved electric field was measured by ‘binning’ and averaging the IR signals generated during individual laser shots into 3 or 10 μs bins, which is the limit of time resolution in the present work.

The discharge cell has three optical access windows, a BK-7 glass window for the incident pump and Stokes beams, a CaF₂ window for the output IR signal beam and CARS (anti-Stokes) signal beam, and a BK-7 glass window used for plasma emission imaging, at a 90° angle to the laser beams. Two circular copper electrodes 14 mm in diameter, each covered by a thin quartz plate (0.2 mm thick, 25 mm diameter) were placed in the cell parallel to each other and 3 mm apart. To preclude direct current path formation between two exposed metal surfaces, each electrode was covered with Dow Corning RTV3145 silicone adhesive/sealant (dielectric strength 19 kV mm⁻¹).

The pair of electrodes in the discharge cell are powered by the Trek 20/20 amplifier generating a high-voltage AC waveform, and by a custom-designed, high voltage ns pulse generator, also used in our previous work [15, 20], using the electric circuit shown in figure 2. Low-pass LC filter and high-pass LRC filter were used to prevent damage of the AC amplifier by the ns pulses and damage of the ns pulse generator by the AC voltage, respectively. The parameters of the filters were chosen to minimize the distortion of the voltage waveform generated by each power supply, while providing maximum attenuation of the other voltage waveform. Stray capacitance of the AC amplifier was used for the low-pass filter capacitor. The main constraint of this approach is that attenuation of the AC voltage waveform becomes more significant as the AC frequency is increased beyond ~2 kHz. Also, operation at high AC frequency and high peak AC voltage was limited by the peak output power of the AC amplifier. In the present work, all measurements were done at two AC frequencies, 500 Hz and 1750 Hz. The ns pulse discharge was operated at a pulse repetition rate twice that of the AC frequency, with pulses generated when the AC voltage was close to zero. The timing of the ns pulses was controlled using a Stanford Research System DG645 delay generator. The voltage between the discharge electrodes and the current in the external circuit were measured by a Tektronix P6015A high voltage probe and Pearson 2877 current monitor, as shown in figure 2. Plasma emission images were taken with a PI-MAX3 camera equipped with a Nikon UV-Nikkor 105 mm f/4.5 lens.

In the present work, the laser pulses were generated randomly during the AC period, and the intensity waveforms of the pump beam, CARS signal, and four-wave mixing IR signal were saved by the oscilloscope for each laser shot, to be analyzed during post-processing. The AC voltage during each laser shot and the voltage waveform over a subsequent 5 μs interval were also saved, to determine the AC phase and the timing of the laser shots/data points during the AC period. The AC period was divided into 200 ‘time bins’ (10 μs long for 500 Hz frequency and approximately 3 μs long for 1750 Hz frequency), and the intensity waveforms in each bin were averaged. Approximately 20 000 laser shots were collected for each run, resulting in an average of ~100 laser shots per bin. Finally, the averaged pump, CARS, and IR intensity waveforms in each bin were integrated over time, and the electric field was determined using equation (3). The calibration
constant in equation (3) was determined by using the same data post-processing procedure as described above, when sub-breakdown AC voltage was applied to the electrodes. In this case, the electric field between the electrodes during the laser pulse was determined as

$$E = \frac{U}{d + \Delta \xi},$$

(4)

where $U$ is the applied voltage measured by the high-voltage probe, $d = 3$ mm is the gap between the quartz plates, $\Delta = 0.2$ mm is the quartz plate thickness, and $\varepsilon = 4$ is the dielectric constant. By plotting the result of equation (3) versus known electric field, the calibration constant was determined from the slope of the calibration line.

3. Kinetic model

Kinetic modeling of AC and ns pulse barrier discharges sustained between two plane electrodes covered by dielectric plates was done using a quasi-1D model discussed in our previous work [15]. Briefly, the kinetic model incorporates (i) Poisson equation for the electric field, solved both in the plasma and in the dielectric plates ($\varepsilon = 4$); (ii) two-term expansion Boltzmann equation for the electron energy distribution function (EEDF), with superelastic collisions incorporated; (iii) equation for the electron temperature; and (iv) species concentration equations for electrons, ions (H$^+$, H$^+_2$, and H$^+_3$) and neutral species (H$_2$(X$^1\Sigma_g^+$), H$_2$(a$^3\Pi_u$), H$_2$(b$^3\Sigma_g^{-}$), H$_2$(c$^3\Pi_u$), H$_2$(B$^3\Sigma_u^+$), H$_2$(d$^3\Sigma_u^-$), H$_2$(e$^3\Sigma_u^+$), H$_2$(f$^3\Sigma_u^-$), and H($n = 1, 3$)), where H$^+_i$ is the sum of Rydberg excited electronic states. Electron impact cross sections are taken from [34] and rate coefficients for neutral species reactions and ion-molecule reactions were taken from [35–37]. It is assumed that charged species deposited on the dielectrics remain there until they are neutralized by the opposite sign charges arriving from the plasma. Secondary electron emission coefficient from the dielectric surface is assumed to be $\gamma = 0.02$ [15]. This represents an effective value, chosen to reproduce the measured electric field offset after a nanosecond pulse, dielectric barrier discharge in hydrogen in plane-to-plane geometry [15], at the conditions similar to the present experiments. The initial conditions assume zero surface charge density on the dielectrics and low electron/ion density in the gap, $10^3$ cm$^{-3}$.

The system of equations is solved self-consistently, using an experimental AC voltage waveform, and assuming a constant gas temperature of 300 K. This assumption is justified since the discharge energy loading per molecule, determined from experimental voltage and current traces, is relatively low, ~0.4 meV/molecule/pulse (which corresponds to temperature rise of $\Delta T \sim 1$ K/pulse). Since at the present conditions the characteristic time for diffusion/conduction is ~ ns, and the AC frequency/ns pulse repetition rate is of the order of ~1 kHz, the estimated temperature rise is of the order of a few degrees.

Although the 1D model is adequate for modeling large-volume ns pulse DBD, such as generated in our previous work [20], it is not applicable for simulation of AC barrier discharges, which consist of a large number of random micro-discharges, as discussed in section 1. High-fidelity kinetic modeling of high-pressure AC DBD plasmas is a formidable challenge, requiring development of a 3D model incorporating a wide range of spatial scales (from plasma sheath thickness to the electrode diameter) and time scales (from breakdown/plasma self-shielding time scale to the AC period). For this reason, in the present work a simpler stochastic approach was used, as described below.

First, a 1D kinetic model [15] was used to simulate the AC barrier discharge between two plane dielectric-covered electrodes over one AC period, and the results (in particular, the residual charge on the dielectric surfaces) were assumed to apply to an individual micro-discharge. This is a valid assumption since characteristic time scales for radial ambipolar diffusion in the micro-discharge streamer and for surface charge migration over the dielectric surface are much longer compared to the time scale of streamer propagation and charge separation/plasma self-shielding when the streamer reaches the surface. Second, the micro-discharges, kinetics of which was predicted by the 1D model, were assumed to be generated during the AC period according to the probability density obtained from multiple overlapped experimental AC micro-discharge current waveforms, and distributed randomly across the face of the electrodes. As an illustration, figure 3(a) shows a histogram with the distribution of the number of micro-discharges per the AC period, obtained from 68 experimental AC current waveforms. Also, figure 3(b) shows all 68 AC current waveforms overlapped and plotted together with the AC voltage waveform, illustrating the probability density of a micro-discharge generation versus AC phase. These results (the number of micro-discharges per period and the temporal probability distribution of their generation during the period) were combined with the residual local surface charge density predicted by the 1D model, and used as input parameters to the stochastic electrostatic model, predicting the ‘line-of-sight-averaged’, time-resolved root-mean-square electric field, $\overline{E^2}$, which is also measured by the four-wave mixing diagnostics. The only adjustable parameter in the stochastic model is the diameter of an individual micro-discharge, assumed to be 0.2 mm [8]. Varying the micro-discharge diameter within a factor of two had almost no effect on the predicted electric field, as discussed in section 4.

4. Results and discussion

4.1. Discharge waveforms and plasma images

Figure 4(a) shows voltage and current traces in the AC discharge in hydrogen at 300 Torr, at the AC frequency of 500 Hz and peak voltage of 4 kV. As expected, the current trace exhibits multiple peaks due to individual micro-discharges. Both the number of micro-discharges per AC period and their timing vary period-to-period, as shown in figure 3, which illustrates the micro-discharge statistics. Figure 4(b) plots voltage and current waveforms for the ns pulse discharge overlapped with the AC discharge and generated when the AC voltage is

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near zero. Pulse peak voltage and current are approximately 11 kV and 3 A, respectively, voltage pulse FWHM is about 30 ns, and the energy coupled to the plasma is about 0.6 mJ (time-averaged ns pulse discharge power of 0.3 W). This is comparable to the energy coupled by the AC discharge, 0.12 mJ/period, measured from charge-voltage characteristics. Figure 4(c) shows the repetitive ns pulse discharge voltage and current waveforms, overlapped with the AC voltage and

![Figure 3](image)

**Figure 3.** (a) Histogram showing distribution of the number of micro-discharge pulses during the AC period, obtained from 68 AC current experimental waveforms; (b) all 68 AC current waveforms overlapped and plotted together with the AC voltage waveform, illustrating the probability distribution of a micro-discharge generation during the AC period used in the stochastic model predicting the time- and spatially averaged effect of the micro-discharges. Hydrogen, $P = 300$ Torr, AC frequency 500 Hz.

![Figure 4](image)

**Figure 4.** Discharge voltage and current waveforms taken over a single AC voltage period: (a) AC discharge alone, showing random micro-discharge current pulses; (b) ns pulse discharge alone; (c) AC discharge overlapped with ns pulse discharges generated at the moments when the AC voltage is zero; (d) AC discharge overlapped with ns pulse discharges, with zoomed-in current waveform showing reproducible AC breakdown pulses (labeled by arrows). Hydrogen, $P = 300$ Torr, AC frequency 500 Hz.
current waveforms. In figure 4(c), ns discharge pulses are generated at a repetition rate of 1 kHz, i.e. twice the AC frequency, when the AC voltage was near zero. Since ns pulse discharge peak current, ~3 A, greatly exceeds the current during AC micro-discharges, ~50–100 mA, the latter are difficult to identify in figure 4(c). For this reason, figure 4(d) plots the combined AC + ns discharge current waveform on a different scale, such that the current peaks during the AC period can be resolved. The most significant difference between the baseline AC current waveform and the current in the AC discharge enhanced by ns pulses is that in the combined discharge the most prominent current peaks during the AC period (~200 μs after the ns discharge pulses) are no longer random and become quite reproducible (with period-to-period jitter not exceeding 10 μs). These current peaks are labeled with arrows in figure 4(d). It can also be seen that the current after the ns discharge pulses, ~1–10 mA, decays relatively slowly, on the time scale of the order of a few hundred μs. All waveforms shown in figure 4 are ‘single-shot’, i.e. taken during a single AC period.

Figure 5(a) shows an image of the electrode/dielectric plate assembly used in the experiments. Figure 5 also shows ICCD images of the AC discharge (figure 5(b), taken with a long camera gate, 2 ms), the ns pulse discharge (figures 5(c), 10 μs gate), and the discharge generated by reproducible breakdown during the AC voltage rise ~200 μs after the ns pulse (figure 5(c), 10 μs gate), also labeled in figure 4(d). All ICCD images shown are accumulations over 100 pulses. From the time-averaged image of the AC discharge (see figure 5(b)), it can be seen that the plasma emission occupies nearly the entire region between the electrodes. However, this emission is generated by multiple micro-discharges, such as can be identified from current peaks in figure 4(a), which are randomly distributed in both time and space during the AC voltage rise (see figure 3(b)). The spatial distribution of individual micro-discharges, which could not be detected directly from emission, appears to be random, thus generating a diffuse emission image. The ns pulse discharge plasma fills the entire region between the electrodes and extends to the outside surfaces of the dielectric plates (see figure 5(c)). Finally, the diffuse plasma generated by the reproducible breakdown during the AC voltage rise after the ns pulse (figure 5(d)) fills approximately half the volume between the electrodes, in the region where the distance between the quartz plates is somewhat shorter, due to the plates not being perfectly parallel to one another.

4.2. Electric field calibration
As discussed in section 2, individual pump, CARS, and IR intensity signals were collected during the AC period, placed into different time bins, and averaged. Figure 6(a) plots typical IR signals measured by the HgCdTe IR detector for several values of the sub-breakdown electric field, each averaged over 150 laser shots. Note that although the signal decay is controlled by the IR detector time constant, the actual time resolution of the present measurements, controlled by the coherence decay time in H₂ molecules, is below 1 ns. The same data are also plotted on a semi-log scale (figure 6(b)), to compare the signal at the low value of the electric field, 400 V cm$^{-1}$, with the noise level. The averaged IR signal, CARS signal, and pump intensities were integrated over time, and equation (3) was used to plot the data versus known sub-breakdown electric field to determine the linear calibration line. Although this technique can be used to determine only the magnitude of the electric field but not its sign, the use of the AC sine wave voltage allowed for efficient calibration of both positive and negative electric field values.

Figure 7 shows both positive (a) and negative (b) electric field calibration lines. Each of the data points shown represents an average of ~100 laser shots at the given field value. The dashed lines above and below the calibration lines represent a 95% confidence interval. It can be seen, as expected, that a linear fit is obtained in both cases, with a near zero
intercept. From the data shown, the sensitivity of the present technique is approximately $\sim 400 \text{ V cm}^{-1}$, which is consistent with our previous measurements [15, 20]. Figure 7(c) shows the electric field values determined using the calibration lines, plotted along with the voltage-to-gap ratio (see equation (4)). The error bars indicate a 95% confidence interval for each measurement value. It can be seen that electric field values determined by the calibrated four-wave mixing technique closely match the high-voltage probe measurements.

4.3. Electric field in the AC barrier discharge

Figure 8 plots vertical and horizontal electric field components measured in the AC barrier discharge in hydrogen at 300 Torr,
at the frequency of 500 Hz, plotted together with applied voltage to gap ratio. Note that the probability of a microdischarge, such as indicated in figure 4, occurring in the path of the laser beam, and overlapping with the laser pulse in time, is very low. Thus, the present technique is measuring the line-of-sight-averaged electric field in the electrode gap, created by the superposition of the applied voltage and charges accumulated on the surface of the dielectric plates. As expected, the line-of-sight-averaged vertical field in the gas discharge, \( \langle E^v \rangle^{1/2} \), is distorted by the surface charge accumulation on the dielectric, such that it deviates significantly from the applied field and its peak value is significantly lower (see figure 8). It can also be seen that the vertical electric field is much higher compared to the horizontal field, which remains below the sensitivity limit of the present measurements, \(-400\text{V cm}^{-1}\), during the entire AC period. Note that since the four-wave mixing diagnostics measures the line-of-sight RMS values of the vertical and the horizontal electric field components, very low value of the horizontal field measured, \( \langle E^h \rangle^{1/2} \), cannot be explained by field cancellation due to the sign change.

Figure 9 shows the averaged vertical electric field in the AC discharge at two different frequencies, 500 Hz (a) and 1750 Hz (b), plotted together with the applied voltage-to-gap ratio and discharge current waveforms. It can be seen that in both cases, the electric field waveform has a nearly ‘flat top’ shape, with peak electric field of approximately 6.0 kV cm\(^{-1}\) at 500 Hz (see figure 9(a)) and 6.7 kV cm\(^{-1}\) at 1750 Hz (see figure 9(b)), exhibiting a modest rise, likely due to the electrodes not being perfectly parallel to one another, when the applied field increases from zero to the peak value of \(\sim 13\text{ kV cm}^{-1}\). The peak electric field approaches, but does not reach DC breakdown field predicted by Paschen law at these conditions (\(\sim 7.5\text{ kV cm}^{-1}\), see figure 9), and is likely limited by microdischarges occurring when the field between the electrodes exceeds the local breakdown threshold, which increase the surface charge density on the dielectrics and shield the plasma during further voltage rise.

As the applied voltage begins to decrease, microdischarges stop occurring, until the field in the plasma, which is a superposition of the applied field and the field created by the surface charges, changes polarity and increases to near breakdown threshold again. At this point, micro-discharges begin to occur again and the electric field rise slows down, resulting in a well-defined ‘kink’ in the electric field waveform, observed for both applied field polarities and at both AC frequencies (labeled with arrows in figure 9).

To predict the AC barrier discharge dynamics in a single micro-discharge, we used a 1D kinetic model described in section 3. Figure 10(a) plots the voltage-to-gap ratio obtained from the experimental voltage waveform used as an input to the model, together with the predicted electric field and current waveforms in an individual micro-discharge, for a single AC period. It can be seen that the model predicts generation of two separate micro-discharges during each half-period, which occurs each time when the electric field in the plasma exceeds breakdown threshold of approximately 7 kV cm\(^{-1}\). During each micro-discharge, the electric field in the plasma halfway between the electrodes is predicted to drop to zero within \(\sim 0.1\mu s\), due to charge accumulation on the dielectric surfaces in 1D approximation and complete plasma shielding (see figure 10(a)). This behavior is illustrated in figure 10(b), which plots the predicted surface charge density and electron density halfway between the electrodes. It is apparent that when the micro-discharges occur, electron density jumps by nearly three orders of magnitude (see figure 10(b)), until ionization is terminated due to electric field reduction. Electron drift toward the positive electrode continues even after the ionization stops, resulting in further charge separation and complete plasma shielding. This effect is well known in kinetic modeling of DBD and is also predicted by 2D micro-discharge models [38, 39]. If 2D effects are incorporated, the electric field predicted in the micro-discharge is no longer zero, but it remains very low [39]. After the electric field is reduced below threshold for electron impact ionization, the plasma begins to decay by electron–ion recombination, as shown in figure 10(b).

After each micro-discharge breakdown, the electric field in the plasma remains near zero for a few tens of microseconds (see figure 10(a)), until the plasma density decays considerably due to electron–ion recombination (see figure 10(b)), such that the surface charge density can no longer increase as the applied voltage is increased further. After this, the field in the decaying plasma starts increasing following the applied voltage waveform (see figure 10(a)), with an offset caused by the surface charges accumulated on the dielectrics (see figure 10(b)), until the breakdown threshold is reached again and the next micro-discharge occurs. When the applied voltage begins to decrease and reaches zero, the surface charges enhance the field in the plasma until it reaches the breakdown threshold after the voltage changes polarity. First micro-discharge breakdown after the voltage polarity change also changes the sign of the surface charge density (see figure 10(b)).
To obtain insight into the cumulative effect of multiple micro-discharges generated between the electrodes during the AC period on the electric field, we used a stochastic model based on the measured average number of micro-discharges per period and their temporal probability distribution (see figure 3), described in section 3. Figure 11 compares the vertical electric field in the AC discharge measured by the four-wave mixing technique (also plotted in figure 9(a)) with the line-of-sight averaged RMS electric field, $\sqrt{\frac{1}{T} \int_0^T E(t)^2 dt}$, predicted by the stochastic model. It can be seen that the experimental results and the modeling predictions are in good agreement, including a nearly ‘flat top’ shape and a well-defined ‘kink’ in the field waveform. The measured peak electric field, approximately $6.0 \text{ kV cm}^{-1}$, is somewhat higher compared to the predicted peak field of $5.0 \text{ kV cm}^{-1}$. This is most likely due to the use of a 1D model of a micro-discharge, which assumes that it is radially uniform and generates uniform surface charge density. In reality, radial non-uniformity of micro-discharge filaments results in non-uniform distribution of surface charge on the dielectric, incomplete plasma shielding, and consequently non-zero vertical field in the filament. Therefore the stochastic model, based on the 1D micro-discharge model predictions, somewhat underestimates the line-of-sight averaged electric field measured by the present four-wave mixing technique.

4.4. Electric field in the AC barrier discharge enhanced by ns pulses

For electric field measurements in the AC barrier discharge overlapped with the ns pulse discharge, the high voltage ns pulses were generated at the repetition rate twice that of the AC frequency, at the moments when the AC voltage was near zero, as shown in figure 4(c). Since the ns pulse generator used in the present work produces alternating polarity pulse trains, the polarity of the pulses was chosen to be the same as the polarity of the AC voltage following the pulse, as shown in figure 4(c). During the experiment, data from laser shots occurring within $\pm 5 \mu s$ of the ns pulses were not collected, since they could be erroneously assigned to incorrect time.
operated alone (see figure 8), the horizontal electric field components measured in the AC discharge operated at 500 Hz, enhanced by ns pulses. Similar to the AC discharge with and without ns pulses, plotted together with applied voltage to gap ratio. Hydrogen, $P = 300$ Torr, AC frequency 500 Hz.

Figure 11. Comparison of four-wave mixing electric field measurements in the AC discharge with the electric field predicted by the 1D kinetic model, after stochastic averaging. Hydrogen, $P = 300$ Torr, AC frequency 500 Hz.

Figure 12. Time resolved electric field vector components measured in the AC dielectric barrier discharge overlapped with ns discharge pulses, plotted together with applied voltage to gap ratio. Hydrogen, $P = 300$ Torr, AC frequency 500 Hz, ns pulses are generated when the AC voltage is zero.

Figure 13. Comparison of the experimental data with the predictions of a 1D kinetic model provides insight into the kinetics of the AC barrier discharge enhanced by high voltage ns pulses. Figure 14(a) plots the voltage-to-gap ratio for the experimental voltage waveform (AC overlapped with two ns pulses), used as an input to the model, together with the electric field and current waveforms for a single AC voltage period, predicted by the model. Figure 14(b) shows predicted electron density halfway between the electrodes and the surface charge density. It can be seen that during the ns pulse discharge, the electron density rapidly increases, up to $n_e \approx 7 \cdot 10^{13} \text{cm}^{-3}$, producing a high-amplitude current pulse (peak current approximately 3 A, off scale in figure 14). After the ns discharge pulse, residual surface charge density is very close to zero, since surface charges are neutralized by charge transport from the volume of the plasma [15].

As the AC voltage gradually increases from zero, the electrons drift toward the positive electrode and increase the surface charge on the dielectric plate covering the electrode, which shields the plasma and keeps the field in the plasma near zero (see figure 14(a)). After the electron density in the plasma decreases considerably due to electron–ion recombination, to $n_e \approx 3 \cdot 10^{11} \text{cm}^{-3}$ (over $\approx 100 \mu s$ after the ns discharge pulse, see figure 14(b)), further increase of the surface charge cannot keep up with the AC voltage rise, such that the field in the plasma begins to increase, following the applied voltage with an offset due to residual surface charge (see figure 14(a)). The field continues to rise until it reaches breakdown threshold, at which point breakdown occurs in the electrode gap, electron density constant in time, except for a less pronounced but detectable kink/drop observed approximately 160 $\mu s$ after each ns pulse, more reproducible during the negative half-periods (see figure 12).

Plotting the measured electric field waveform together with the applied voltage and discharge current waveforms at 500 Hz and 1750 Hz AC frequencies (see figure 13), and comparing these data with plasma emission images (see figure 5) provides additional information on discharge dynamics. The main trends of the electric field behavior during the AC periods at both frequencies are very similar. Specifically, time lags between the applied voltage and the electric field after each ns pulse are similar, $\approx 30 \mu s$ and $\approx 10 \mu s$, respectively. Reproducible field drop occurs approximately at 7.5 kV cm$^{-1}$ at 500 Hz and at about 5 kV cm$^{-1}$ at 1750 Hz. It is apparent that the sudden electric field jumps after each ns pulse (by a factor of $\approx 2 - 3$, down to $\approx 2$ kV cm$^{-1}$) coincide with well-defined discharge current peaks (labeled by arrows in figure 13), reproducible period-to-period, which are observed during both positive and negative half-periods, at both AC frequencies. These current peaks also correspond to well-reproduced AC plasma emission generated at the same time delay after each ns discharge pulse (approximately 200 $\mu s$ at 500 Hz, see figure 4(d)). It can also be seen that the secondary kinks/drops in the electric field, better pronounced during the negative AC half-periods, correlate with lower amplitude current peaks, which are also reproducible period-to-period, although with more significant jitter (up to $30 \mu s$).

Comparison of the experimental data with the predictions of a 1D kinetic model provides insight into the kinetics of the AC barrier discharge enhanced by high voltage ns pulses. Figure 14(a) plots the voltage-to-gap ratio for the experimental voltage waveform (AC overlapped with two ns pulses), used as an input to the model, together with the electric field and current waveforms for a single AC voltage period, predicted by the model. Figure 14(b) shows predicted electron density halfway between the electrodes and the surface charge density. It can be seen that during the ns pulse discharge, the electron density rapidly increases, up to $n_e \approx 7 \cdot 10^{13} \text{cm}^{-3}$, producing a high-amplitude current pulse (peak current approximately 3 A, off scale in figure 14). After the ns discharge pulse, residual surface charge density is very close to zero, since surface charges are neutralized by charge transport from the volume of the plasma [15].

As the AC voltage gradually increases from zero, the electrons in the high-density plasma drift toward the positive electrode and increase the surface charge on the dielectric plate covering the electrode, which shields the plasma and keeps the field in the plasma near zero (see figure 14(a)). After the electron density in the plasma decreases considerably due to electron–ion recombination, to $n_e \approx 3 \cdot 10^{11} \text{cm}^{-3}$ (over $\approx 100 \mu s$ after the ns discharge pulse, see figure 14(b)), further increase of the surface charge cannot keep up with the AC voltage rise, such that the field in the plasma begins to increase, following the applied voltage with an offset due to residual surface charge (see figure 14(a)). The field continues to rise until it reaches breakdown threshold, at which point breakdown occurs in the electrode gap, electron density constant in time, except for a less pronounced but detectable kink/drop observed approximately 160 $\mu s$ after each ns pulse, more reproducible during the negative half-periods (see figure 12).

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increases, and the field drops to zero again, due to additional surface charge accumulation on the dielectrics. Note that peak electron density and peak current during this ‘AC’ breakdown are much lower compared to the ns pulse discharge, since the AC voltage at this moment is significantly lower compared to ns pulse peak voltage.

After the AC breakdown, the line-of-sight averaged electric field in the gap remains near zero until the plasma density decays due to recombination (over ~50 μs), when it begins to follow the applied electric field again, although with a more significant offset caused by additional surface charge accumulation (see figures 14(a) and (b)). The next AC breakdown occurs when the applied field decreases after reaching maximum, such that the field in the plasma reaches breakdown threshold due to the offset. The electric field variation after the second AC breakdown is very similar to that after the first breakdown. Finally, both the field in the plasma and the surface charge density on the dielectrics are reduced to zero after the next nanosecond discharge pulse, generated when the AC voltage is reduced to zero. The discharge behavior during the negative AC half-period is identical to that described above.

Figure 15 compares the measured electric field with the model predictions in the 500 Hz AC barrier discharge enhanced by ns pulses. It can be seen that the main trends in the electric field evolution during the AC period are reproduced by the model fairly well, justifying the use of the 1D model at these condition and providing insight into the plasma dynamics. Specifically, the model shows that residual surface charge density and residual electric field after ns discharge pulses are very low, due to neutralization of surface charge by transport of charge species from the high electron density plasma, the effect discussed in greater detail in our previous work [15]. The model also predicts the moment of reproducible AC breakdown detected both during the positive and negative AC half-periods, approximately 200 μs after the ns discharge pulses (see figure 15). Note that in the experiment, the electric field after the AC breakdown pulses does not drop to zero, as predicted by the
1D, most likely because the plasma is generated over only approximately half the volume between the electrodes, since the dielectric plates are not perfectly parallel to each other (see figure 5(d)). Because of this, the electric field measured after AC breakdown has a significant non-zero contribution from the region outside the plasma. For the same reason, the model overpredicts the surface charge density and the offset between the applied electric field and the field in the plasma, thus predicting the second AC breakdown when the field in the plasma changes direction, which is not detected in the experiment (see figure 15). Basically, lower surface charge density on the electrodes and consequently lower electric field offset in the experiment prevent the second AC breakdown pulse. Finally, both in the experiment and in the modeling calculations, field in the gap is reduced to near-zero after ns discharge pulses.

The other difference between the experimental results and the model predictions apparent from figure 15 is a significantly longer delay time before the field starts to increase after the nanosecond pulses. The model predicts this delay time to be \( \sim 100 \mu s \), controlled by electron–ion recombination (see figures 14(a) and (b)), compared to \( \sim 30 \mu s \) in the experiment. This difference is likely to be due to ns pulse discharge plasma also being generated outside of the discharge gap (see figure 5(c)), which may establish a direct current path between the exposed parts of the electrodes. This would result in additional conduction current through the decaying plasma in the discharge gap, when capacitors in the external surface are discharging after the ns discharge pulses. This ‘external current leak’ would reduce charge separation in the AC discharge plasma, limit surface charge accumulation, and prevent electric field shielding. Indeed, the experimental current waveform (see figures 13(a) and (b)) shows a slow decay after the ns pulse discharge over a few hundred \( \mu s \), both for positive and negative polarity pulses, while the predicted current, which does not take this effect into account, decays to zero within \( \sim 100 \) ns (see figure 14(b)).

Comparison of the electric field measurements and kinetic modeling calculations, shown in figure 11 for the AC barrier discharge and in figure 15 for the AC discharge enhanced by ns discharge pulses demonstrates that adding ns pulses changes the discharge behavior drastically. Most significantly, the AC discharge transitions from a superposition of random micro-discharges generated mainly during the AC voltage rise (see figure 3), and distributed in space fairly randomly over the discharge volume, as suggested by plasma emission in figure 5(b), to a far more regular, quasi-1D discharge. Basically, in the AC discharge enhanced by the ns pulses, breakdown has a tendency to occur regularly, at a well-defined time delay after each ns pulse, and over a large volume (e.g. see figure 5(d)). The number of random micro-discharges, on the other hand, is reduced significantly, especially at the lower frequency of 500 Hz (compare figures 9 and 13).

The experimental data and modeling calculations indicate that the main role of volumetric ns pulse discharge pulses is (i) to neutralize surface charges over the entire dielectric surface area, and consequently reduce residual electric field in the entire plasma volume to near zero (see figures 5(c) and 14(b)), and (ii) to generate significant diffuse ionization in the entire volume between the electrodes (see figures 5(c) and 14(b)). Both considerable reduction of surface charge density accumulated by random micro-discharges, which no longer generates the ‘grainy’ electric field distribution in the plasma, and generation of diffuse ‘initial’ ionization are critical for producing large-volume, well-reproduced AC breakdown pulses. A qualitatively similar effect has been detected previously in a ns pulse dielectric barrier discharge in plane-to-plane geometry (without the AC voltage), when increasing the pulse repetition rate from 10 Hz to \( \sim 10 \) kHz range transformed the ns pulse discharge from an ensemble of spatially random streamers to a uniform diffuse plasma [14].

In the present work adding ns high voltage pulses to the AC discharge waveform did not produce uniform AC breakdown in the entire volume between the electrodes, such that the enhanced discharge behavior still exhibits significant deviation from the 1D kinetic model. However, there is little doubt that a more precise parallel plate electrode arrangement, as well as placing the electrodes outside of the discharge cell to block conduction current from the external circuit would help generating AC breakdown over a larger volume.

5. Summary

In the present work, the effect of ns barrier discharge pulses on the AC barrier discharge in hydrogen in plane-to-plane geometry is studied using time-resolved measurements of the electric field in the plasma. The components of the electric field were measured by ps four-wave mixing technique similar to CARS, which generates coherent IR signal proportional to the square of electric field. Absolute calibration was done using a sub-breakdown field applied to the electrodes, when no plasma was generated. The sensitivity of the present measurements is approximately \( 400 \text{ V cm}^{-1} \). The results are compared with 1D kinetic modeling of the AC discharge and
nanosecond pulse discharge, predicting behavior of both individual micro-discharges and their cumulative effect on the electric field distribution in the electrode gap, using stochastic averaging based on the experimental micro-discharge temporal probability distribution during the AC period.

The results show that, as expected, the electric field in the AC barrier discharge is controlled by a superposition of random micro-discharges, produced mainly during the AC voltage rise and distributed in space over the discharge volume, generating quasi-diffuse plasma. The root-mean-square horizontal electric field component averaged over the line of sight is much smaller compared to the root-mean-square vertical field component, and is near detection limit of the present diagnostics. Time-resolved vertical field during the AC period exhibits a nearly ‘flat top’ distribution, with the maximum near breakdown threshold. Kinetic modeling predictions, based on the 1D micro-discharge model combined with stochastic averaging of micro-discharges during the AC period, are in good agreement with the electric field measurements.

Adding ns discharge pulses on top of the AC voltage waveform changes the AC discharge behavior in a fundamental way, inducing a transition from a superposition of random micro-discharges to a more regular, quasi-1D discharge. In this case, volumetric AC breakdown is produced with a well-defined delay after ns pulses, reproducible period-to-period. During the reproducible AC breakdown, the electric field in the plasma exhibits a sudden drop, which coincides in time with a well-defined current pulse. This experimentally observed trend is also predicted by kinetic modeling of the AC discharge enhanced by ns pulses, using a 1D model. Analysis of kinetic modeling predictions shows that this effect is due to large-volume ionization and neutralization of surface charges on the dielectrics by ns discharge pulses. The kinetic modeling predictions are in fairly good agreement with the trends detected from electric field and discharge current measurements. The differences between the experimental data and the modeling predictions, such as the magnitude of the electric field drop and the amplitude of the current pulse, are due to the AC breakdown being not quite 1D, with the plasma generated only over about half of the volume between the electrodes. The delay time after which the field in the plasma starts increasing following ns discharge pulses, much shorter in the experiment compared to the 1D model prediction, is likely due to the ‘external current leak’, which limits surface charge accumulation and prevents electric field shielding.

The present work provides quantitative insight into the effect of ns discharge pulses on the AC dielectric barrier discharge, and demonstrates that this approach may be used to control the phase of barrier discharges, triggering reproducible breakdowns at well-defined moments. Although the present electric field measurements were done with ns pulses generated when the AC voltage was near zero, the same approach may be used at other moments during the AC period, such that multiple AC breakdowns per period may be triggered during pre-determined moments. Neutralization of surface charges accumulated during the AC barrier discharge operation, produced by multiple ns discharge pulses, may significantly enhance energy coupled to the volumetric plasma by the AC voltage waveform, as well as electrostatic body force in surface AC DBD plasma actuators [18]. Further parametric studies of barrier discharges sustained by combined AC/ns pulse waveforms in different geometries, over a wide range of frequencies, pulse repetition rates, and phases are needed to quantify these effects.

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