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Excited state population dynamics of a xenon ac discharge

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Abstract

We measure the time evolution of the $6s'[1/2]_{\pi} - 6p'[3/2]_{\pi}$ (834.68 nm, air) excited neutral xenon transition lineshape in a xenon 60 Hz oscillatory discharge by applying time-synchronized laser induced fluorescence (LIF) spectroscopy. Two different time-synchronized LIF techniques are demonstrated, yielding consistent results and revealing distinct features: a reduction of peak fluorescence intensity (representative of the $6s'[1/2]_{\pi}$ state density) is observed at high values of the discharge current, the maximum fluorescence intensity occurs at low values of the discharge current, and the excited state populations quench as the alternating current passes through zero. This behavior is reproduced and explained by collisional-radiative modeling, which highlights the role of collisional and radiative mixing between excited energy states throughout the current cycle.

Keywords: xenon ac discharge, time-synchronized laser induced fluorescence, collisional-radiative model, laser diagnostics

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

1. Introduction

This paper addresses the physics of an oscillatory 60 Hz xenon plasma capillary discharge with strongly time-dependent characteristics. In particular, we show the time evolution of the $6s'[1/2]_{\pi} - 6p'[3/2]_{\pi}$ transition lineshape at 834.68 nm (air) excited neutral xenon transition lineshape in a xenon 60 Hz oscillatory discharge by applying time-synchronized laser induced fluorescence (LIF) spectroscopy. Two different time-synchronized LIF techniques are demonstrated, yielding consistent results and revealing distinct features: a reduction of peak fluorescence intensity (representative of the $6s'[1/2]_{\pi}$ state density) is observed at high values of the discharge current, the maximum fluorescence intensity occurs at low values of the discharge current, and the excited state populations quench as the alternating current passes through zero. This behavior is reproduced and explained by collisional-radiative modeling, which highlights the role of collisional and radiative mixing between excited energy states throughout the current cycle.

Laser induced fluorescence (LIF) is a non-intrusive diagnostic method in which the quantum state densities of a species are perturbed through laser pumping and consequent changes in the spontaneous fluorescence emission are detected [1, 2]. The resulting neutral or ion transition lineshape provides a wide range of plasma parameters after applying proper deconvolution methods and spectroscopic models [3, 4]. The measurement achieves fine spatial resolution, as determined by the volume of intersection of the probe laser and collection optical path. Developing time-resolved LIF methods for characterizing time-varying plasma properties has been the subject of recent research effort [5–7] in order to capture the time-dependent characteristics in transient or oscillating plasmas, such as Hall thrusters [8], dielectric barrier discharges, magnetized $E \times B$ discharges [9, 10], plasma display panels [11, 12] and microdischarges [13].

In this study, two time-synchronized LIF techniques are implemented to track the evolution of the Xe I $6s'[1/2]_{\pi} - 6p'[3/2]_{\pi}$ transition lineshape at 834.68 nm (air) throughout a 60 Hz discharge current cycle. One approach relies on quickly switching the fluorescence signal; the second, implemented previously by MacDonald \textit{et al} [14], involves a sample-hold procedure. Both time-synchronization methods involve generating an acquisition gate triggered at a given phase of the oscillating cycle during which optical filtering and phase-sensitive homodyne detection extract the induced fluorescence signal from the bright background emission in the probed plasma volume. The sample-and-hold scheme was already applied to this 60 Hz discharge, although the previous study focused on demonstrating and validating the method rather than performing a detailed
physical investigation. In the present study, we validate the fast-switching time-synchronization scheme against the established sample-hold approach. We also extend the previous measurements on this discharge using a finer time resolution, revealing physical features not seen before: the maximum observed peak fluorescence intensity occurs at low values of the discharge current, although it sharply attenuates in a narrow time window at the current zero crossing. The peak intensity also decreases at the discharge current maxima. We implement a collisional-radiative model in order to identify the physical processes acting to produce the measured profile. A nine level scheme for xenon proposed by Sommerer and validated against experimental data on dc discharges [15] is modified to accommodate a 60 Hz alternating discharge current. This approach has been demonstrated to successfully capture the physics of positive columns in weakly ionized, relatively low pressure (few Torr) discharges [16–18]. Similar models have also been applied in power electronics engineering for predicting the impedances of fluorescent lamps [19, 20] and have been used to address the physics of plasma display panel cells [21].

2. Time-synchronized laser induced fluorescence measurements

2.1. Experimental Setup

The plasma source consists of an enclosed glass tube filled with xenon. There is a central capillary region 10 cm long with a diameter of 1.5 mm, and two wider hollow electrodes at each end that are powered at 60 Hz. Each electrode acts alternately as an anode or cathode, supporting a sinusoidal discharge current profile with an amplitude of 15 mA. The fill pressure estimated through a Voigt fit of the 834.68 nm (air) spectral lineshape is 7 Torr [22]. In this study we probe a plasma volume at the center of the capillary region. Further details regarding these discharges may be found in [14, 23, 24].

We apply a non-resonant laser induced fluorescence scheme. Atoms in the Xe I excited 6s′[3/2] state are optically pumped to the excited 6p′[3/2] state at higher energy (transition at 834.68 nm, air). As shown in figure 1, the excitation source is a New Focus TA-7600 semiconductor tapered amplifier seeded with a New-Focus TLB-6017 external cavity diode laser through a polarization-maintaining optical fiber. This system has a <300 kHz linewidth and a 50 GHz mode hop-free tuning range. We use 15 mW of laser power to perform the LIF measurements.

The instantaneous wavelength is visually monitored with a Burleigh WA-1500 wavemeter and the precise detuning is determined from the interference peaks of a Thorlabs SA-200 Fabry–Perot interferometer with a free spectral range of 1.5 GHz and a finesse of 200. A xenon hollow cathode lamp with optogalvanic detection is used as an additional reference for identifying line center. The main laser beam is amplitude-modulated with a mechanical chopper and focused on the discharge with a 25 mm diameter, 25 mm focal length lens. The fluorescence emitted during radiative decay from the 6s[3/2] → 6p′[3/2] transition at 473.4 nm (air) is detected with a photomultiplier tube (PMT) after passing through a 750 nm cut-off filter centered at 470 nm. A 2 mm diameter iris restricts the target collection volume to ~1 mm³, rejecting background emission from the remaining plasma. The PMT current signal is converted to a voltage through a 5 kΩ shunt resistor and processed through the analog time-synchronization circuits. Phase-sensitive homodyne detection using a Stanford Research Systems Model 850 lock-in amplifier discriminates the induced fluorescence signal from natural background emission from the same transition.

2.2. Time-synchronized LIF methods

Block diagrams of the time-synchronization methods are shown in figure 2. Both approaches achieve time resolution by collecting LIF signal only during an acquisition gate locked at a given phase of the discharge current cycle. The 60 Hz current oscillation is monitored with a 2878 Pearson probe.
and processed through a LM399 voltage comparator chip. The comparator generates a high-level voltage output for positive input signals and a low-level output for negative input signals (illustrated in solid red in figure 2), identifying when the current crosses zero in the positive direction. A Stanford Research Systems DG535 pulse-delay generator produces a TTL gate (dotted blue) of width $\tau$ and a delay $\Delta t$ from the ‘zero phase’ trigger received from the comparator. The LIF signal collected from the PMT is sampled inside the acquisition gate according to the two different methods, effectively rejecting all other signal originating outside the desired phase in the current cycle. The sampling methods used are as follows:

(i) **Sample-and-Hold Method.** The fluorescence signal collected inside each acquisition gate is averaged and held until the following gate, at which point it is updated with the new value. The optical chopping frequency used is lower than the characteristic frequency of the plasma oscillation so that multiple sample-and-hold signal updates occur within one laser modulation. This provides enough fluorescence photons to accurately represent the true LIF signal while applying the chopper modulation for the lock-in amplifier. For the 60 Hz xenon discharge, we use a laser chopping frequency of 4.6 kHz.

(ii) **Fast Switching Method.** The signal collected inside each acquisition gate is sent directly to the lock-in amplifier with no additional processing, while the remaining signal is discarded. Typical optical chopping frequencies are higher than the characteristic frequency of the plasma oscillation so that several laser modulations occur within an acquisition gate and the lock-in amplifier has a clear frequency component to lock onto when receiving LIF signal. This procedure has the added benefit of operating in a higher frequency regime where the background spectral noise density is usually lower. For the 60 Hz discharge, we use a laser chopping frequency of 4.6 kHz.

For both techniques, the acquisition gate is parked at the desired phase of the oscillation period and a full laser wavelength scan is performed, reconstructing the LIF transition lineshape at one specific time delay $\Delta t$. The time evolution of the LIF lineshape is then obtained by repeating laser scans at various delay times along the full current cycle. Note these measurement schemes may be extended up to frequencies of oscillation in the tens of kilohertz range, as recently demonstrated using a 20 kHz Hall thruster breathing mode [25].

2.3. Time-synchronized LIF measurements

We track the time-varying Xe I 834.68 nm (air) transition lineshape in the 60 Hz discharge using both time-synchronization methods described in the previous section. First, LIF traces are collected in 1 ms increments using a 1 ms acquisition gate width for a total of 16 time points over the 16.7 ms discharge period. Additional scans at higher resolution are then performed between 6–11.5 ms where steep changes in the acquired LIF signal are observed as the discharge current crosses zero at $\Delta t = 8.35$ ms. The second measurement group uses a 0.5 ms gate width with a variable time step between 0.125–0.5 ms to resolve these features. Scans take roughly 5–10 min to complete and are obtained with a lock-in time constant of 3 s and sensitivity of 200–500 $\mu$V.
LIF traces acquired with both time-synchronization methods are presented in figure 3 as contour plots. Each trace at a given delay time $\Delta t$ (horizontal axis) constitutes a vertical slice in one of the panels as a function of the laser detuning frequency from line center (vertical axis). The traces are normalized by the maximum intensity value found in a single panel. Top panels (a1) and (a2) are obtained with the sample-and-hold method, while (b1) and (b2) are obtained using fast switching of the fluorescence signal. In each case, the panel labeled (1) displays the results of the coarse resolution, while panel (2) shows the finer resolution scans. The dotted black boxes indicate the temporal range where finer resolution scans are taken. The experimentally measured discharge current traces at the bottom of the figure orient the time axis. In post-processing we correct a small relative offset in the time axes between measurement groups due to daily drift in the monitored discharge current and ‘zero phase’ time marker.

Figure 3 shows a variety of interesting physical features that are reproduced by both methods at both time resolutions. The maximum observed peak fluorescence intensity occurs at low (absolute) values of the discharge current and peak fluorescence intensity decreases by roughly half near the discharge current extrema. The finer acquisitions (right panels) reveal an almost total attenuation of the LIF signal at the current zero crossing, a feature that appears distorted in the rougher acquisitions (left panels). This quenching of the signal as the current changes sign occurs in less than 1 ms, and thus cannot be fully resolved by the coarse measurement gate. Rather, the first signal maximum (while current is positive) dominates the 7–8 ms gate in panels (a1) and (b1), producing the observed peak intensity feature. However, the subsequent 8–9 ms gate samples both the strong signal dip and part of the second signal maximum (while current is negative), leading to a perceived reduction in signal intensity to $\sim 0.7$ during this time interval. The true features are only revealed by decreasing the measurement gate width, as shown in panels (a2) and (b2). By combining the results of the coarse and fine resolution scans, we construct the complete time evolution of the LIF peak intensity during a single discharge current cycle in figure 4(a). The plot is normalized to the maximum intensity in each case and highlights the consistency of the measurements based on the sample-and-hold and fast switching methods.

Further agreement between the methods is illustrated in figure 4(b), in which representative LIF lineshapes obtained by both methods at $t = 8.5, 9,$ and $10$ ms in the fine resolution scans are plotted together. Each has been normalized by the respective maximum intensity used in figure 3. The traces match well with some expected variation due to background noise. Figure 4(c) shows that the lineshape has a nearly constant width in time, suggesting that the pre-dominant broadening mechanism is due to pressure and not other mechanisms such as Doppler broadening. Neglecting
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the sample-hold outlier at 8.3 ms (due to reduced signal-to-noise ratio near the minimum intensity point), the mean and standard deviations ($\mu$, $\sigma$) of the lineshape full widths at half maximum in GHz are (1.19, 0.059) and (1.23, 0.057) for the sample-hold and fast switching methods, respectively. Both metrics agree within 3%.

The peak intensity of each LIF trace provides a relative measure of the $^{6s'}^{6s}[1/2];$ xenon neutral excited state population at a given time interval. From figures 3 and 4, it is apparent that this state depopulates at high discharge current, increases in density at lower values of the discharge current and rapidly quenches when the current crosses zero. Several physical mechanisms at work in the discharge, including electron-impact excitation and de-excitation, radiative decay, heavy particle collisions and radiation trapping all combine to generate the particular time history measured here. The details of these processes are further investigated numerically with a collisional-radiative model.

3. Physical interpretation of LIF measurements

We implement a zero-dimensional dynamic collisional-radiative (CR) model of the weakly-ionized xenon discharge to further examine the physics underlying the acquired LIF data. Using an appropriate set of atomic energy levels and coupled rate equations, the model tracks the time variation of the ground and exited state population densities, $n_g$ and free electron density, $n_e$, in the positive column of the discharge.

The implemented model incorporates simplifying assumptions when compared with state-of-the-art modeling of excited state populations in weakly-ionized plasmas, including a simplified treatment of radiation transport. As extensively described by Golubovskii et al [16], a rigorous accounting of radiation trapping, in addition to solving the various transport equations in multiple dimensions, is necessary for obtaining exact values for the full spatial and temporal distribution of excited state population densities in a discharge. However, as shown in this section, such assumptions do not preclude the present model from achieving qualitative agreement with the LIF measurements and providing a simple physical interpretation of the observed experimental profiles.

3.1. Energy levels and cross sections

The model is based on the set of xenon atomic energy levels initially proposed and validated by Sommerer [15] for analyzing the positive column of an axially symmetric dc plasma discharge. The levels are summarized in table 1 and illustrated schematically in figure 5, where we have chosen to neglect the $8p$ level. The figure also specifies the physical processes considered for transitioning between levels in the model: electron-impact excitation and de-excitation, electron-impact ionization, radiative decay, and heavy particle collisions between excited xenon atoms. The proper electron impact cross sections are identified from the literature and used to compute accurate electron impact rate coefficients with the Boltzmann solver Bolsig$^+$ [26]. Xenon cross sections for electron elastic collisions, electron impact excitation and ionization from the ground state are retrieved from online databases [27, 28]. The remaining excitation and ionization cross sections, heavy particle collision rates and radiative decay rates included in the calculation are taken from [29–31], as originally identified by Sommerer [15]. Electron impact de-excitation rates are computed by applying detailed balance with the corresponding excitation transition.

Figure 4. (a) Time profiles of the peak LIF lineshape intensity (Xe $^{6s'}^{6s}[1/2];$ transition, 834.68 nm air) obtained with the two time synchronization methods. The profiles are constructed by polynomial fits to the rough and fine time resolution data for each case (see figure 3) and are normalized to the respective maximum intensities. (b) Comparison of raw LIF lineshapes taken from the high time resolution scans at $t = 8.5, 9,$ and 10 ms. (c) Full widths at half maximum of the fluorescence lineshapes from all scans showing a fairly constant distribution.
Adopting a conventional approach often used in similar models, we represent the radiative decay rates through effective lifetimes according to Holstein’s theory of radiation trapping [32] to include the effects of cascade absorption and reemission of photons from surrounding atoms. A complete representation of this process would require the solution of the full integral radiation transport equation, as demonstrated by Golubovskii et al [33] in a model of the spot mode in a 7 torr He-Xe discharge.

3.2. Model basic equations and computation

The modeled discharge is defined to have a fill pressure $P = 7$ torr, wall temperature $T_w = 300$ K, and plasma column radius $R = 0.75$ mm. We apply a sinusoidal discharge current with an amplitude $I_0 = 15$ mA and a frequency $f = 60$ Hz:

$$I(t) = I_0 \sin(2\pi ft).$$

Table 1. Xenon atomic energy levels included in the collisional-radiative model as identified by Sommerer [15].

<table>
<thead>
<tr>
<th>Level</th>
<th>Sommerer’s notation</th>
<th>Racah notation</th>
<th>Energy (eV)</th>
<th>Multiplicity</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>Xe($^1S_0$)</td>
<td></td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>Xe($6s_2$)</td>
<td>6s($3/2</td>
<td>2$)</td>
<td>8.31</td>
</tr>
<tr>
<td>2</td>
<td>Xe($6s_1$)</td>
<td>6s($3/2</td>
<td>1$)</td>
<td>8.43</td>
</tr>
<tr>
<td>3</td>
<td>Xe($6s'_0$)</td>
<td>6s($1/2</td>
<td>0$)</td>
<td>9.44</td>
</tr>
<tr>
<td>4</td>
<td>Xe($6s'_1$)</td>
<td>6s($1/2</td>
<td>1$)</td>
<td>9.57</td>
</tr>
<tr>
<td>5</td>
<td>Xe($6p$)</td>
<td>6p($1/2</td>
<td>1$, 6p($5/2</td>
<td>2$, 6p($5/2</td>
</tr>
<tr>
<td>6</td>
<td>Xe($7p$)</td>
<td>7p($1/2</td>
<td>1$, 7p($5/2</td>
<td>2$, 7p($5/2</td>
</tr>
<tr>
<td>7</td>
<td>Xe($6p'$)</td>
<td>6p'($3/2</td>
<td>2$, 6p'($3/2</td>
<td>1$,</td>
</tr>
<tr>
<td>8</td>
<td>Xe$^+$</td>
<td></td>
<td>12.13</td>
<td>2</td>
</tr>
</tbody>
</table>

Note: Level 0 is the ground state, level 4 is probed with LIF, and level 8 is the continuum (singly-ionized xenon).

Adopting a conventional approach often used in similar models, we represent the radiative decay rates through effective lifetimes according to Holstein’s theory of radiation trapping [32] to include the effects of cascade absorption and reemission of photons from surrounding atoms. A complete representation of this process would require the solution of the full integral radiation transport equation, as demonstrated by Golubovskii et al [33] in a model of the spot mode in a 7 torr He-Xe discharge.

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(i) Rate Coefficients. First, Bolsig+ solves the zero-dimensional Boltzmann equation using the given cross sections and creates lookup tables for the rate coefficients $K$ and mean electron energy $\langle \epsilon_e \rangle$ for several values of the reduced electric field $E/n_0$ (where $n_0$ is the ground state density). The mean electron energy is derived from the electron energy distribution function (EEDF), which we assume is established primarily by collisions with the ground state. Bolsig+ also estimates the average electron mobility as $\mu_e = 2.22 \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ for the conditions of interest. The Boltzmann equation is solved at each time step assuming a steady-state, stationary electric field, given the low oscillation frequency of the plasma. The reduced electric field, $E(t)/n_0(t)$, thus acts as the time variable. A similar approach was used by Meunier in a one-dimensional discharge model of an ac plasma display panel cell [21].

(ii) Applied Electric Field. The time-varying electric field $E(t)$ is given by Ohm’s Law:

$$E(t) = \frac{1}{\mu_e \epsilon_0 n_0(t)} \cdot \frac{I(t)}{\pi R^2} \quad (1)$$

![Figure 5](image-url)
where \( e \) is the elementary charge, \( \mu_e \) is the electron mobility from Bolsig+, and \( n_e(t) \) is the time-dependent electron density. We assume a quasi-neutral plasma \( n_i = n_e \).

(iii) Rate Equations. Rate equations are written for each excited state energy level \( j \in [1, 7] \), incorporating the various physical mechanisms that produce allowed transitions between states (outlined in figure 5):

\[
\frac{dn_j}{dt} = \sum_{p=r} K_{pj} n_i n_p - \sum_{q=r} K_{pj} n_i n_q + \sum_{r>j} \frac{n_r}{\tau_{rj}} - \sum_{s<j} \frac{n_j}{\tau_{sj}} - K_{pj} \sum_{s=1}^7 n_s. \tag{2}
\]

\( K_{pj} \) denotes the rate coefficient of the transition from state \( p \) to state \( j \) due to electron impact excitation, ionization or de-excitation. \( \tau_{rj} \) are the characteristic lifetimes. \( K_h \) is the rate coefficient for ionizing collisions between (heavy) excited states, estimated to be \( 4.3 \times 10^{-18} \text{ m}^3 \text{ s}^{-1} \) [15]. The analogous equation for electrons (level 8) is:

\[
\frac{dn_e}{dt} = \sum_{p=0}^7 K_{pj} n_i n_p - 5.47 \frac{D_a}{R} n_e + K_h \sum_{q=1}^7 n_q n_r. \tag{3}
\]

\( D_a \) is the ambipolar diffusion coefficient. In these types of discharges, electron losses due to charged particle recombination in the bulk plasma are negligible compared with ambipolar diffusion to (and consequent recombination at) the walls [17, 20, 34]. The instantaneous radial distribution of electrons is approximated with a zero-order Bessel function, giving the overall diffusion flux that appears as the second term on the right-hand side of equation (3). The ambipolar diffusion coefficient is calculated as \( D_a \approx \frac{2}{3} \tau_{m_e} \) from the mean electron energy \( \epsilon_e \) and the ion mobility \( \mu_i \). In the latter equation we assume a Maxwellian distribution function and use the simplified electron temperature \( k_B T_e = \frac{2}{3} \epsilon_e \). Note that this assumption is applied only for calculating the ambipolar diffusion coefficient. The xenon ion mobility in a background of neutral xenon gas is estimated according to Piscitelli [35] as \( 7.54 \times 10^{-3} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1} \).

(vi) Time Integration. The rate equations are advanced in time using a stiff coupled ordinary differential equation solver. At each time step \( n \), the reduced electric field is first estimated using equation (1) for the instantaneous electric field \( E^n \) and the initial ground state density \( n_0 \).
is calculated from the ideal gas law $n_0 = p/K_B T$ using the wall temperature $T_w$. An initial guess for the mean electron energy $\epsilon_{\text{e}0}^{m}$ is then obtained from the tables generated by Bolsig+. This allows for the power deposition $P_e^{m}$ into the gas due to electron collisions and the current gas temperature $T^m$ to be estimated using [15, 19]:

$$P_e^{m} = \frac{2m_e}{m_i} \epsilon_{\text{e}0}^{m} \nu_e n_e$$

$$T^m = T_w + \frac{P_e^{m} R^2}{4K_{Xe}}$$

(4)

(5)

where $m_e$ and $m_i$ are the electron and xenon ion masses, respectively, $\nu_e = e/m_e j_{\text{e}i}$ is the electron momentum scattering rate (using the electron mobility from Bolsig+), and $K_{Xe} = 6 \times 10^{-3}$ W m$^{-1}$K$^{-1}$ is the xenon thermal conductivity. With the updated gas temperature, the current ground state population $n_0^m$ is calculated knowing the current population of all the excited state levels $n_j^m$ and electrons ($j = 8$):

$$n_0^m = \frac{P}{k_B T^m} - \sum_{j=1}^{8} n_j^m.$$ 

(6)

Finally, the updated ground state population $n_0^m$ is used to better estimate the reduced electric field $E^m/n_0^m$ at the current time step. This corrected reduced electric field is then used for interpolating the Bolsig+ tables to obtain the time-dependent rate coefficients $K_{ji}$ and the mean electron energy $\epsilon_{\text{e}1}^{m}$. 

### 3.3. Physical discussion of LIF and computational results

We track the time evolution of the various energy levels of the model throughout 20 ms of the 60 Hz discharge current oscillation. Time histories of five key excited state populations (in m$^{-3}$), along with the ionization fraction $(n_i/n_0)$ and simulated discharge current trace, appear in figure 6. The simulations highlight the differing periodic behavior of the lower energy $6s$ states (panels (a)–(c) in the figure; $6s'$ is probed with LIF) compared with the higher energy $6p$ states (panels (d) and (e) in the figure). The lower energy states deplete at higher (absolute) values of the discharge current and increase in density as the current decreases. On the contrary, the higher energy states achieve their maximum density along with the discharge current maxima. The ionization fraction is also greatest at the discharge current peak, as expected, due to the high energy input into the plasma. As the discharge current changes sign, population of all states ceases and radiative decay and electron-impact de-excitation mechanisms strongly quench the excited state and continuum densities.

Qualitative agreement between the experimental LIF measurements and the model solutions is demonstrated in figure 7. The experimental and numerical time evolution of the $6s'$ density throughout one current cycle (normalized to the maximum value in each case) is shown in figure 7(a). Figure 7(b) compares the simulated radiated emission from the $6s_1 - 6p'$ radiative transition at 473.5 nm as predicted by the CR model and detected through a photomultiplier tube with a 10 nm bandpass filter centered at 470 nm. The radiation intensity is representative of the upper state $6p'$ density. (c) Electric field calculated with the model and measured with high voltage probes, normalized to the mean value in the plateaus. All three parameters demonstrate the close agreement obtained between the model and experimental system.

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**Figure 7.** (a) Time evolution of the normalized $6s'$ excited state density predicted by the model and measured with time-synchronized LIF spectroscopy. The main features of the profile are labeled 1–3 and described further in the text. (b) Time-varying emission from the $6s_1 - 6p'$ radiative transition at 473.5 nm as predicted by the CR model and detected through a photomultiplier tube with a 10 nm bandpass filter centered at 470 nm. The radiation intensity is representative of the upper state $6p'$ density. (c) Electric field calculated with the model and measured with high voltage probes, normalized to the mean value in the plateaus. All three parameters demonstrate the close agreement obtained between the model and experimental system.
emission \(\eta_{j}/n_{j}\) has been normalized to the peak value, and the PMT voltage has been smoothed and normalized to the mean value in the high emission regions. Figure 7(c) compares the discharge electric field in the capillary region calculated by the model and measured by applying a high voltage probe to each electrode referenced to a common ground. The traces have been normalized to the mean value in the plateau regions and the measured electric field has been smoothed. The model clearly reproduces all the physical features experimentally observed.

We identify three characteristic regions in the oscillation period, labeled on figure 7(a), defined by the time-varying energy fluxes to and from the experimentally probed \(6s'_{1}\) state (outlined further in figure 8). At high values of the discharge current (phase 1), the dominant process leading to depopulation of the \(6s'_{1}\) state is electron-impact excitation to the upper levels. In contrast, for low values of the discharge current (phase 2), excitation from the lower levels plus radiative decay from the upper \(6p'\) state combine to overcome the depopulating processes, leading to the maximum observed \(6s'_{1}\) density. As the current changes sign (phase 3), however, the electric field no longer sustains the plasma discharge and all excitation processes halt—the \(6s'_{1}\) (and other) populations are quenched. The sequence then repeats in reverse for the next half period of the oscillation.

We obtain additional confirmation of this physical picture by comparing the radiative emission from the \(6s_{1} - 6p'\) transition predicted by the code and measured through the PMT in figure 7(b). Note that multiple other neutral and ion lines are present within the 10 nm bandwidth of the PMT filter; however, the contributions to the total emission from these lines are negligible. The other neutral transitions have much lower relative intensities compared to the target, and the low ionization fraction of the discharge (on the order of \(10^{-5}-10^{-4}\)) discounts significant contributions from the ion lines. The predicted and measured emission traces also agree, accurately capturing the quenching feature at the current zero crossing. Since the emission intensity is representative of the upper state density \(n_{j}\), this serves as experimental confirmation that the CR model predicts the behavior of the higher energy states as well.

An analysis of the energy fluxes to and from the probed \(6s'_{1}\) state in figure 8 reveals that this state populates both from direct electron-impact excitation from the ground state and from collisional-radiative cascading from the upper energy states. Similar processes have been observed in extensive studies of time-varying excited state populations in plasma display panel cells [11, 21, 36] that operate at different conditions than those treated in this paper. In such devices, UV light emitted from xenon resonance or molecular (excimer) states excites phosphors in the three fundamental colors. Of particular interest is the 147 nm UV emission originating from the \(6s_{1}\) resonance state, which we calculate to follow the same trend as the \(6s'_{1}\) state shown here (which emits at 130 nm). Population of resonance states through the cascading processes seen here represents an energy loss in PDPs since it implies higher energy investment per UV photon produced in comparison with direct excitation from the ground state. Some energy is lost in collisional-radiative mixing between close-lying energy states, which is also observed.

Ionization (see figure 6(f)) is seen to occur mainly as a multi-step process; direct ionization from the ground state is rare due to the low average electron energy. During periods of maximum ionization, ground state xenon atoms first experience two excitation steps: initially to the low energy \(6s\) states, and then to the higher energy \(6p\) and \(6p'\) states comprising the main reservoirs for ionization. Similar stepwise ionization through the resonance and metastable states is observed in other types of discharges, such as ac plasma display panel...
cells [12] and dc capillary devices [15]. In contrast, very near to the current zero crossing, the few ionized atoms originate mostly from the 6s states since the upper 6p and 6p′ levels are strongly depopulated and the majority of atoms lie in lower energy excited states.

4. Conclusion

We combine experimental and modeling approaches to characterize in detail the physics of a 60 Hz oscillating xenon discharge. Experimentally, we implement two time synchronization schemes for laser induced fluorescence spectroscopy: one, a method based on quickly switching the collected fluorescence signal, and the other, an established sample-and-hold technique previously applied to these types of discharges. By tracking the time evolution of the 6s′[1/2] − 6p′[3/2] spectral lineshape, we reveal how the excited 6s′[1/2] state population density changes throughout the 60 Hz discharge current cycle. This state appears to depopulate at high discharge current, reach a maximum density at low discharge current, and quench as the discharge current crosses zero. Both time synchronization methods capture this time-dependent behavior and yield consistent results.

A simple collisional radiative model delves further into the physical mechanisms behind the observed time variations. We simulate the evolving density of seven xenon excited states and the first ionization level. The model, derived from Sommerer’s validated xenon CR system [15], includes several mechanisms for transitioning between states, approximates radiation trapping and accounts for heavy particle collisions, and treats electron losses through ambipolar diffusion to the walls. Simulation results and experimental measurements demonstrate qualitative agreement. At high discharge current, the lower energy excited states depopulate through additional excitation to the upper states, while at low discharge current they achieve maximum density. The predicted increase in density of the higher energy states at high discharge current is also observed and validated through emission measurements from the 60 Hz discharge. Finally, when the current approaches zero, population halts and all the excited states quench through radiative and non-radiative decay processes.

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