

Non-perturbative 2D spatial measurements of electric fields within a plasma sheath

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We introduce an all-optical quantum-enhanced diagnostic for electric fields in low-temperature plasmas. Trace amounts of rubidium vapor, added to argon plasma, allow us to produce spectrally narrow electric field-sensitive optical resonances via quantum optical effect of Rydberg electromagnetically induced transparency, and to non-invasively measure electric field in plasma with sensitivity exceeding 1 V/cm. By collecting fluorescence from the illuminated region of interest, we reconstruct a 2D spatial profile of the electric field magnitude with 30 μm resolution. As a proof-of-principle demonstration, we measured the changes in electric field within the plasma sheath surrounding a biased Langmuir probe tip. This method holds significant potential for studying sheath structures in low-temperature plasmas.

Electric fields are omnipresent in plasmas and play an important role in plasma-material interactions [1], dusty plasmas [2], waves, instabilities and turbulence. Precise measurements of electric fields in plasmas to uncover dynamics at the scale of the sheath and pre-sheath for plasma-material interactions requires non-perturbative measurements that can achieve high spatial resolution and very small electric fields [3]. Existing techniques involving laser-induced fluorescence dip (LIF-dip) [4, 5], electric-field-induced coherent anti-Stokes Raman scattering (E-CARS) [6] and electric field induced second harmonic generation (E-FISH) [7] focus on high temporal resolution of large electric fields relevant for atmospheric plasmas. At low pressures, dust particles suspended in plasma using optical tweezers have been employed to measure the electric field [8]. However, none of these techniques have been able to fully visualize the plasma sheath and pre-sheath with Debye-length precision in low pressure plasmas.

Our electric field measurement using quantum atom-based optical sensors techniques provide a 30 μm spatial resolution of the electric field to values below 1 V/cm. Currently, direct measurements of the electric field in low pressure plasmas is obtained with collecting and emissive probes [3, 9, 10]. While these probes have a small radius ($\sim 0.25\text{--}0.5\text{mm}$), they perturb the plasma with their own sheath and pre-sheath characteristics, limiting the resolution with which they can identify the electric field in the sheath. Our method solves these problems, as we use neutral atoms as sensitive probes of their electrostatic environment, and we use lasers to both prepare the atoms in the maximally sensitive quantum states and to accurately measure the spectroscopic changes due to the local electric field.

We insert trace amounts of neutral rubidium (Rb) vapor into an argon (Ar) plasma, and use a two-photon non-linear process called electromagnetically induced trans-

parency (EIT) [11, 12], that links changes in optical transmission with the minute variations in atomic energy levels driven by external fields, as shown in Fig.1(a). This enables accurate monitoring of the spatial and temporal variations of the impact of the plasma's electric field on highly-excited (Rydberg) quantum states of Rb atoms. This technique, using lasers interacting with alkali metal vapors contained in a sealed glass cell, has been originally explored as a way to accurately measure radiofrequency and static electric fields, in an SI-traceable way, with minimal perturbation [13]. In the past few years, interest in using such vapor cell-based measurement units to replace metal antenna and receiver systems has exploded [14–22]. Rydberg EIT was also used to measure electric fields in ion plasmas prepared from laser-cooled atom clouds [23, 24], but not inside more traditional low density ion-electron laboratory plasmas with room temperature ions and electron temperatures, $T_e < 10\text{ eV}$.

The successful development and deployment of this technique can enable a broad range of high precision plasma measurements in low-temperature, low-pressure plasmas. Beyond the aforementioned applications to sheath measurements, in dusty plasmas, these techniques may enable measurements of ion wakes around dust particles [25–27], sheath structures formed by dust clouds [28, 29], or potential structures formed in magnetized plasmas [30, 31].

The concept of the proposed measurement arrangement is shown in Fig.1(b). Two counter-propagating laser fields illuminate the region of interest inside plasma. When sum laser frequencies match the frequency difference between the ground and excited (Rydberg) states, Rb atoms are prepared in a so-called “dark” superposition of ground and excited quantum states, characterized by reduced fluorescence. In the presence of an electric field E , the $nD_{5/2}$ level splits into three due to Stark effect, according the their $|m_j|$ value. In the limit of

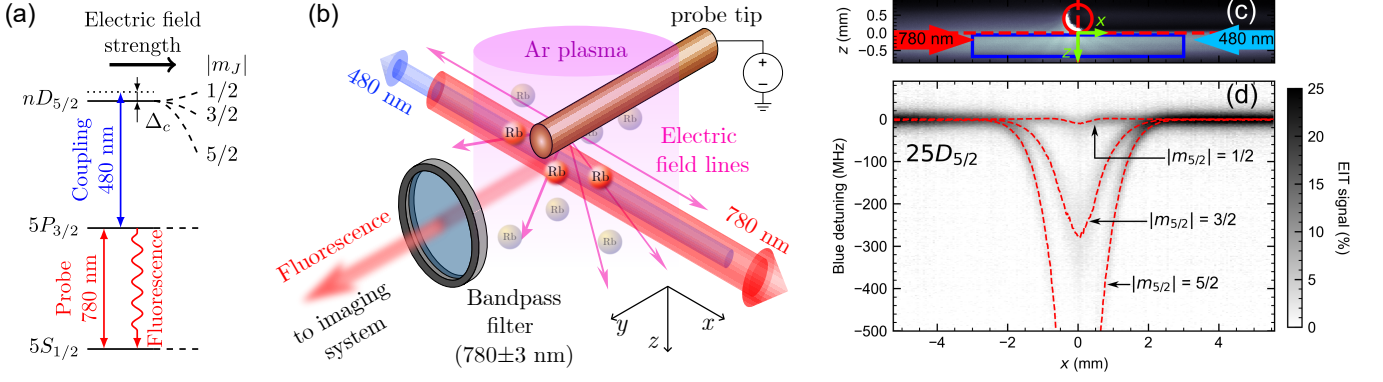


FIG. 1. **(a)** Interaction scheme for Rb Rydberg EIT, showing Stark splitting of the excited Rydberg state $nD_{5/2}$ in electric field. Δ_c is the detuning of the coupling laser frequency from the unperturbed $5P_{3/2} \rightarrow nD_{5/2}$ transition. **(b)** Conceptual experimental schematic: counter-propagating red probe (780 nm) and blue coupling (480 nm) lasers are weakly focused just beneath the Langmuir probe tip within the argon plasma column. The thin arrows depict electrostatic field lines originating from the biased probe tip. Thick red and blue arrows indicating the 780 nm and 480 nm laser propagation directions. **(c)** The fluorescence image with the probe tip marked as a red circle. The xz coordinate system orientation is shown with green arrows. The blue rectangle indicates the area shown in Fig. 3(a,b). **(d)** Rydberg EIT ($25D$) resonances in plasma with electron density $n_e = 6 \times 10^{14} \text{ m}^{-3}$ and electron temperature $T_e = 0.4 \text{ eV}$ at 14 mTorr Ar pressure, the probe is biased at +6.0 V relative to the grounded chamber.

weak fields the frequency shift of each sublevel depends quadratically on the amplitude of the electric field E at the atom's location:

$$f_{|m_j|} = -\frac{1}{2}\alpha_{|m_j|}E^2, \quad (1)$$

where $\alpha_{|m_j|}$ (in units of $\text{MHz}\cdot\text{cm}^2/\text{V}^2$) is the polarizability of the m_j sublevel. We can experimentally measure these Stark shifts and thus reconstruct local electric field values from direct measurements of the frequencies corresponding to the three dips in laser fluorescence [32, 33].

Our experimental apparatus uses low energy electrons (accelerated by $< 36 \text{ V}$) to generate an Ar plasma by impact ionization (ionization threshold: $E_{i,Ar} \approx 15.7 \text{ eV}$). This method ensures operation at low plasma densities and temperatures [34], while avoiding radio-frequency fields that can impact atomic spectroscopy. In addition, the plasma can be biased by an external electric field. The plasma is generated within an octagonal vacuum chamber with eight separate ports used for optical access, injection of Rb vapor, a pressure gauge and retractable Langmuir probe (“L-probe” hereafter) for plasma temperature and density measurements. The same probe is used to generate the source of the electric field and sheath subsequently measured by our Rydberg EIT. During each experiment run, the Ar gas pressure was maintained constant within the range of $p = 13 - 16 \text{ mTorr}$ ($\pm 15\%$), which are sufficiently low not to cause significant collisional broadening for Rydberg EIT resonances [35]. We use a so-called “hot filament”, dc plasma source. Here, a four filaments wires of 0.125 mm diameter tungsten wire are heated to a point of thermionic emission with a dc power supply that provides a heating current of 10.0 A

at a voltage of 2.4 V. The filaments are negatively biased with respect to the grounded chamber walls with values from 0 to 36 V. This configuration leads to plasma densities spanning $n_e = 4 \times 10^{14}$ to $1 \times 10^{16} \text{ m}^{-3}$ and electron temperatures $T_e = 0.1 - 0.7 \text{ eV}$ measured by the L-probe.

An oven, holding a Rb metal ampule, is attached to one of the plasma chamber viewports and heated to approximately 80°C for all measurements. This introduces sufficient Rb vapor into the plasma region, and we estimate Rb number density to be about $5 \cdot 10^{14} \text{ m}^{-3}$ using the Doppler-broadened absorption profile of the probe laser at the ^{85}Rb D_2 line.

The laser beams are focused to a 0.8 mm beam diameter, with power levels for the 780 and 480 nm wavelengths maintained at 0.4 and 51 mW, respectively. The IR probe 780 nm laser is locked at the $F = 3 \rightarrow F' = 4$ cross-over resonance of the $5S_{1/2} - 5P_{3/2}$ transition of ^{85}Rb , and the blue coupling 480 nm laser frequency is scanned across the fine structure of the $25D$ Rydberg level. The fine structure splitting between the $25D_{5/2}$ and $25D_{3/2}$ states of 802.215 MHz is used for frequency calibration, with overall accuracy better than 1 MHz.

In our experiment a simple imaging system collects the Rb fluorescence $I(x, z)$ from the laser beams passing immediately below the L-probe, shown as a red circle in Fig. 1(c). Biasing the L-probe creates a non-uniform electric field we use to study the sheath formation. To locate the spectral positions of three EIT resonances at each location, we slowly scan the coupling laser frequency at a rate of 10 MHz, and record $1536 \times 128 \text{ px}$ fluorescence images with 13.7 Hz frame rate for 683 values of the coupling laser detuning Δ_c . Fig. 1(d) shows the variation of relative change in fluorescence S as a function of the

position along the laser beam (horizontal x -axis) and the coupling laser detuning Δ_c (vertical axis). The EIT signal defined as $S = 1 - \frac{I}{I_{bg}}$, where I_{bg} is the background fluorescence signal away from EIT resonances.

The EIT resonances persist even when Ar plasma is ignited. Away from the L-probe, where electric field is zero, we observe a single EIT peak. Near the probe, three resonances are clearly resolved, and their separations are largest immediately below the L-probe, where electric field is the strongest. The resonances are noticeably broadened as a result of E-field inhomogeneities across the laser beams cross-section, which is perpendicular to the direction of the laser propagation. Slicing the fluorescence images along the z -axis decreases this broadening and produces 2D electric field distributions.

To reconstruct the magnitude of the local electric field E we model each individual $|m_j|$ Stark component using area-normalized lineshapes $L_{\sigma_i}(f)$, where σ_i is the linewidth of the $j = (2i - 1)/2$ sublevel. In the presence of plasma, the EIT peak shape is best described using Holtsmark distribution [36]. Then we can extract the value of the electric field E by fitting the total fluorescence signal S by the sum of all Stark components:

$$S = A \sum_i a_i L_{\sigma_i}(f - f_i(E)), \quad (2)$$

where A represents the overall amplitude, a_i is the scaling factor for each of three $|m_j|$ resonances, and $f_i(E)$ are the frequency positions of the peaks, given by Eq.(1) for weak E and calculated numerically for arbitrary field strengths using the Alkali Rydberg Calculator [37]. We also account for electric field gradients by introducing an additional resonance shift $(df_i/dE)\delta E_{\text{eff}}$, where δE_{eff} represents the deviation of the electric field from the average field within the beam. Collectively these shifts increase the EIT resonances linewidth as $\sigma_i(E) = \sigma_0 - \frac{df_i}{dE}\delta E_{\text{eff}}$, where σ_0 corresponds to the unperturbed EIT linewidth at $E = 0$.

The examples of the electric field reconstructions in three different vertical locations below the L-probe ($x = 0$) are shown in Fig. 2(a), where we analyze the fluorescence spectrum integrated over the bins of $28 \mu\text{m}$ along z -axis. As expected, the electric field is stronger closer to the probe surface (for smaller values of z), resulting in more pronounced resonance shifts in the Stark maps. By repeating this analysis for each point across the laser beams, as shown in Fig. 2(b), we can construct a 2D map of the electric field.

To better illustrate the effect of plasma on the electric field, we set the probe bias to $+6.0$ V and continuously increase the plasma density by raising the negative filaments' bias voltage from the breakdown voltage of -13.0 V to -30.0 V at an Ar gas pressure of 14 mTorr. Fig. 3(a,b) shows examples of the 2D electric field distribution around the L-probe below and above the plasma

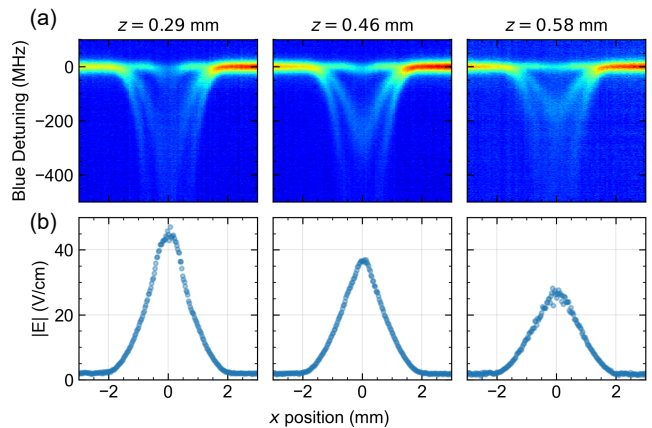


FIG. 2. (a) Maps (arbitrary colormap scale) showing Stark split EIT resonances evolution at increasing vertical distances (z -axis) from the Langmuir probe surface in plasma. The resonances correspond to the splitting of the $25D_{5/2}$ Rydberg level. The smearing of the resonances is attributed to the electric field gradient across the cross-section of the laser beam. (b) Magnitude profiles of the E-field, reconstructed from the Stark maps using Eq. (2).

threshold. The electric field without plasma clearly extends further from the probe. Plasma ignition results in curtailing the wings of this distribution and reducing the non-zero electric field region to about 2 mm around the probe wire tip. Figure 3(c) illustrates the changes in the reconstructed radial electric field distributions at the distance R from the L-probe wire tip as the plasma density n_e increases. These changes clearly demonstrate enhancement in electric field screening and a slight increase in the peak field at the L-probe position.

In the current experimental configuration we are not able to reconstruct the electric fields below 2 V/cm due to technical limitations, making it problematic to reliably study pre-sheath formation. At the same time, it is possible to increase the sensitivity by tuning the blue coupling laser to a higher n Rydberg state. In recent measurements we demonstrated ~ 0.02 V/cm electric field sensitivity using $n = 58$ Rydberg state [33]. It should be possible to reach similar sensitivity, enabling accurate pre-sheath measurements, though electric field fluctuations and gradients may somewhat worsen the signal to noise ratio in plasma.

In conclusion, we demonstrate a powerful method for monitoring electric field inside plasmas. By inserting trace amount of neutral alkali metal (in our case, Rb) we can take advantage of optical atom-based quantum sensing toolkit to image spatial distribution of electric field in plasma with high spatial resolution and high sensitivity. Specifically, we take advantage of high electrostatic sensitivity of highly excited Rydberg ($n > 20$) Rb states to accurately map local Stark shifts by monitoring fluorescence spectra. Moreover, by adjusting the principle

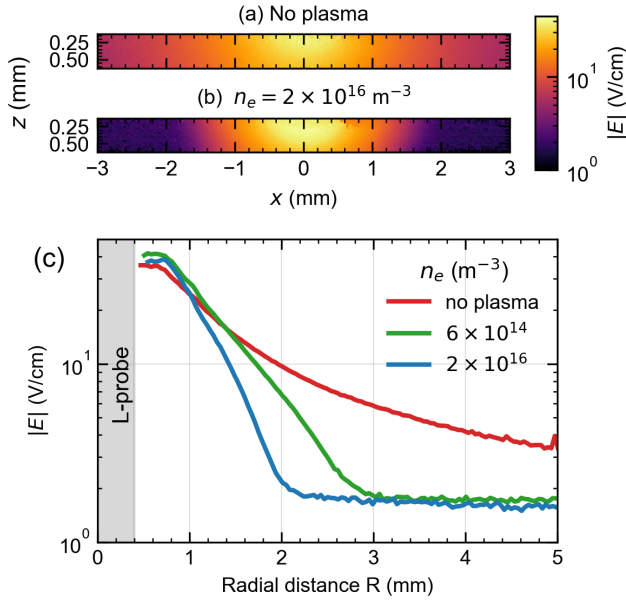


FIG. 3. (a) 2D reconstruction of the E-field magnitude surrounding the Langmuir probe tip, which is biased to +6.0 V in the absence of plasma. (b) 2D reconstruction with a plasma density of $n_e = 2 \times 10^{16} \text{ m}^{-3}$, an electron temperature of $T_e = 0.7 \text{ eV}$, and an Ar pressure of $p = 14 \text{ mTorr}$. Note that the aspect ratio is to scale. (c) The radial E-field profile within the plasma sheath around the Langmuir probe at a biasing voltage of +6.0 V. The evolution of the curve is caused by the increasing plasma density n_e via enhanced screening. The shaded area marks the probe tip wire. The profiles were derived from angular integration of the 2D reconstructions.

quantum number n we can select our sensitivity from a few V/m for $n = 25$ to a few tens of mV/m of $n \geq 45$. Spatial resolution of 2D electric field reconstruction is determined by optical resolution for fluorescence detection and signal to noise ratio of the detected images, and for current experiment is around $30 \mu\text{m}$. We demonstrate the power of this method by accurately monitoring the variation of electric field around biased Langmuir probe. We observe gradual increase in electric field screening as plasma density increases, as expected.

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