Ps four-wave mixing measurements of electric field in a ns pulse discharge in a hydrogen diffusion flame

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Abstract

Time-resolved electric field in ns pulse discharge plasmas generated in room air and in an atmospheric pressure hydrogen diffusion flame has been measured by ps four-wave mixing, for plane-to-plane electrode geometry. Electric field is put on the absolute scale using the Laplacian field measured before breakdown. The results show that peak electric field during breakdown in the flame, approximately 40 kV/cm, is significantly lower compared to that in room air, 75 kV/cm, due to higher temperature of combustion products. In both cases, peak electric field is higher compared to DC breakdown field. Both in air and in the flame, the electric field follows the applied voltage before breakdown and decreases rapidly after breakdown, due to charge separation and plasma self-shielding. The electric field in air is compared with the predictions of an analytic model of ns pulse breakdown, showing good agreement between the predicted and the measured breakdown field. The model also predicts earlier breakdown as well as breakdown voltage reduction as the temperature is increased, in qualitative agreement with the experimental data. The use of the present ps four-wave mixing diagnostics for measurements of electric fields below ~20 kV/cm in atmospheric pressure flames is challenging, due to low signal-to-noise. The sensitivity of the present diagnostics is controlled by the high temperature and low N₂ fraction in the combustion product mixture, as well as by the limited bandwidth of the Stokes beam generated by the stimulated Raman cell, which provides access only to several rotational levels of nitrogen molecules. The present diagnostics will have much better sensitivity in high-pressure flames, since the four-wave mixing signal scales as the squared number density of nitrogen.

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1. Introduction

Electric field measurements in flames and in plasmas sustained in fuel-air mixtures are essential for understanding the effect of strong electric fields on flame dynamics, kinetics of radicals generated in nonequilibrium plasmas, and for development

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of combustion control methods and plasma assisted combustion applications [1–3]. In flames, temporal variation and spatial distribution of the electric field control the ion wind and affect ion reaction chemistry [3], which may strongly influence flame stability [4], flashback [5], soot generation [6], and flow field in the reaction zone [7,8]. In pulsed electric discharges in fuel-air mixtures, spatio-temporal electric field distribution affects discharge energy partition among internal energy modes of molecules and atoms [1], which is the dominant effect controlling the number densities of excited species and radicals generated in the plasma, as well as the rates of plasma chemical reactions [2]. In these environments, predicting the electric field distribution by solving the Poisson equation for the electric potential may well be inaccurate, due to the strong sensitivity of the space charge distribution to rates of ionization, recombination, ion-molecule reactions, electron emission from electrodes, and transport coefficients of electrons and ions. Quantitative insight into these processes requires electric field measurements using non-intrusive experimental techniques.

Recently, significant progress has been made in electric field measurements in air plasmas by optical emission spectroscopy [9–11] and by four-wave mixing [12–15]. Time resolution of these measurements is limited by time response of a camera or a detector (in emission spectroscopy), laser pulse duration (in four-wave mixing), and discharge time jitter (in both techniques). Both methods have limitations on spatial resolution. In emission spectroscopy, the plasma region over which the signal is collected may have significant uncertainty, complicating the data analysis [9]. In four-wave mixing, spatial resolution along the laser beams, determined by the Rayleigh range of the beams and the coherence length, may be up to several cm [15]. Finally, optical emission spectroscopy can be used only for electric field measurements in plasmas during and after breakdown, such that it does not allow measurements in flames below breakdown or absolute calibration using the known electrostatic field.

In this work, we employ ps four-wave mixing diagnostics, using nitrogen as a probe species. This method is similar to Coherent Anti-Stokes Raman Scattering (CARS) and is described in detail in Refs. [12–15]. In this technique, the probe beam used in CARS is replaced by an externally applied electric field, which acts as a “zero-frequency” probe wave, generating an infrared coherent signal beam with the wavelength determined by the energy difference between the two lowest vibrational states of the probe species molecules. The intensity of the IR beam scales quadratically with the electric field and quadratically with pressure, similar to CARS, making this approach effective for electric field measurements in high-pressure flames and high peak voltage pulsed discharges. Absolute calibration is obtained by the Laplacian electric field measured in the same gas mixture and at the same laser operating conditions. The use of a picosecond pump laser provides the temporal resolution of the measurements of ~1–2 ns [15], limited by the high-voltage pulse generator jitter. The Stokes beam for the four-wave mixing is generated in a high-pressure stimulated Raman shifting cell, collinear to the pump beam. The time resolution can be improved to 0.2 ns [16,17] if signal waveforms are saved for every laser shot and subsequently post-processed, instead of averaging them over multiple laser shots during the experiment.

The electric field is produced by a ns pulse discharge sustained between two parallel plane electrodes encapsulated in dielectric sleeves, generating diffuse quasi-one-dimensional plasmas in room air and in a hydrogen diffusion flame. Ns duration voltage pulses are used to produce high peak electric field, while minimizing the ion wind effect on the flame, which would require significant impulse of the Coulomb force, typical for DC and AC fields. This discharge geometry lends itself to comparison with future kinetic modeling, to understand coupling between the applied electric field, flame dynamics, and plasma kinetics. The objectives of the present work are to demonstrate feasibility of electric field measurements in atmospheric pressure flames with high temporal resolution, to obtain insight into breakdown kinetics, and to provide data for assessment of predictive capability of kinetic models.

### 2. Experimental

Figure 1 shows a schematic of the four-wave mixing experimental apparatus [15]. A picosecond Nd:YAG laser (Ekspia SL333) producing output pulses ~150 ps long with energy of 60 mJ/pulse at 532 nm, pumps a high-pressure Raman cell to generate a collinear Stokes beam at 607 nm. The Raman cell is a stainless steel tube 3 m long and 5 cm in diameter, filled with a 50%–50% mixture of nitrogen and helium at a pressure of 15 bar. Helium is added to help dissipate heating caused by
laser beam absorption in the cell and to improve the output beam quality. The pump and Stokes beams linewidth, 0.1 cm$^{-1}$ and 0.4 cm$^{-1}$, respectively, are measured by a High Finesse Angstrom WS/6 wavemeter. Higher order Stokes and anti-Stokes output, also generated in the Raman cell, is filtered out by the dichroic mirrors transmitting the 607 nm first order Stokes beam and the 532 nm pump beam.

The collinear pump and Stokes beams are recollimated and focused in the region between the electrodes by a 50 cm focal distance lens, as shown in Fig. 1. After the test region, the pump and Stokes beams are recollimated again, and the four-wave mixing signal beam (at 4.3 μm) is separated from the pump, Stokes, and anti-Stokes ($N_2$ vibrational CARS) beams using a dichroic mirror which transmits in the infrared while reflecting in the visible. The CARS beam is separated from the pump and Stokes beams using dichroic mirrors such as placed after the Raman cell, and its intensity is measured by a photodiode. The IR four-wave mixing signal beam is separated from residual visible beams using a CaF$_2$ equilateral dispersion prism, and its intensity is measured by a liquid nitrogen cooled InSb detector with a matching preamplifier.

The four-wave mixing beam intensity is proportional to the squared electric field integrated along the length of the discharge electrodes. Previous measurements [15] show that at the present conditions, the four-wave mixing signal generated along the electrodes is distributed nearly uniformly, due to a large signal coherence length, 14 cm at room temperature and 40 cm at $T=1300$ K. IR, $N_2$ CARS, and pump laser signals, as well as voltage and current waveforms, are monitored by a LeCroy Waverunner MXi-A digital oscilloscope with a 1 GHz sampling rate. The waveforms are averaged over 300 laser shots.

A double dielectric barrier discharge in room air and in a hydrogen diffusion flame is sustained between two parallel plate stainless steel electrodes placed inside quartz sleeves 10 cm long each, as shown in Fig. 2. The distance between the sleeves is $d = 1.3-1.7$ mm, the electrode dimensions are $W = 18$ mm and $L = 45$ mm, and the thickness of the sleeve walls (dielectric constant $\varepsilon = 3.8$) is $\Delta = 1.3$ mm. The clearance between the electrodes and the channels in the sleeves is approximately $\delta = 0.1$ mm, such that the effective electrode gap is $d^* = d + 2\delta + 2\Delta/\varepsilon = 2.2-2.6$ mm, with the uncertainty of ±0.2 mm. The electrodes are powered by a Mega Impulse pulse generator producing pulses with peak voltage of up to 26 kV and pulse duration of 5–10 ns FWHM, operated at pulse repetition rate of 10 Hz and triggered by a delay generator, which also triggers the flash lamps of the laser. A second delay generator triggers the Q-switch of the laser and controls the delay time between the discharge pulse and the laser pulse, varied from $t = −10$ ns (10 ns before breakdown between the electrodes) to $t = 20$ ns. The pulse voltage and current are measured by Tektronix P-6015 high voltage probe and Princeton 2877 current probe. Comparison of pulse voltage waveforms measured by the Tektronix probe and by a custom-made, high bandwidth voltage probe [18] shows that peak voltage and pulse width measured by both probes agree within several percent. The voltage falls below 100 V (≈0.05-0.1% of the peak) within ~1 μs after each pulse, such that the applied voltage duty cycle is extremely low, ~10$^{-5}$. Plasma emission images are taken by Princeton Instruments PI-Max 3 ICCD camera.

The electric field is determined from time-integrated IR, pump, and CARS signal intensities, $E = \alpha \sqrt{I_{\text{IR}} I_{\text{pump}}/I_{\text{CARS}}}$, where $\alpha$ is the calibration constant [15,16]. For absolute calibration, the signal is scaled to match the voltage waveform at $t < 0$ (before breakdown), when the electric field in the gap remains Laplacian, $E = U/d^*$, where $d^*$ is the effective electrode gap. The knowledge of the combustion product mixture composition (specifically the number density of the probe species, $N_2$) is not needed for the inference of the electric field. The uncertainty of the electric field is determined by the uncertainty of the electrode gap, ±10%. The detection limit at room temperature is approximately 3–4 kV/cm.

![Fig. 2. Top: schematic of the discharge electrode assembly, bottom: photograph of a hydrogen electrode flame (gap between the electrode sleeves 4 mm).](image-url)
electrode sleeves are placed approximately 20 mm above the burner, as illustrated in Fig. 2. The flame temperature is measured by the broadband, collinear geometry ns CARS diagnostics [19], using an injected-seeded Nd:YAG laser (pump beam linewidth 0.015 cm⁻¹) and a custom-build broadband dye laser (Stokes beam FWHM 3 nm, centered at 605 nm). The CARS measurement region length is determined from the non-resonant background signal distribution from a thin glass plate, with 90% of the signal generated over a distance of 4 mm. The flame temperature is inferred from the best fit CARSFT synthetic spectra [20].

3. Results and discussion

Figure 3 shows a ns broadband N₂ CARS spectrum taken in a hydrogen diffusion flame 5 mm above the burner, without the electrode sleeves in place, and compared with the CARSFT spectrum. The flame temperature inferred from the synthetic spectrum is $T = 1300 \pm 50$ K. Adding the electrode sleeves has a significant effect on the CARS spectrum, which in this case cannot be represented by a single temperature synthetic spectrum, most likely due to incomplete mixing of air with high-temperature combustion products in a relatively narrow “slot” between the electrode sleeves. This effect becomes more pronounced as the distance between the electrode sleeves is reduced from $d = 10$ mm to 4 mm. At $d < 4$ mm, pump and Stokes beams, focused by a 15 cm focal distance lens, could not pass between the electrode sleeves without clipping them, and CARS data were not taken.

Comparison of N₂ vibrational CARS spectra in room air taken using the broadband ns CARS system [19], and using a ps pump laser beam and a narrowband ps Stokes beam generated in the Raman cell (see Fig. 3) shows a significant difference. As expected, the resolution of the ps CARS spectrum employed in four-wave mixing is much lower compared to that of the ns CARS spectrum, due to the significant difference in the pump beam linewidth (0.1 cm⁻¹ for ps Nd:YAG laser compared to 0.015 cm⁻¹ for injection-seeded ns Nd:YAG laser). Also, the linewidth of the Stokes beam generated in the Raman cell is very narrow, 0.4 cm⁻¹ (compared to ~80 cm⁻¹ Stokes beam FWHM for the broadband dye laser) such that both ps CARS and ps four-wave mixing have access only to a small number of N₂ rotational states (the ones populated in the Raman cell). The estimated bandwidth of ps four-wave mixing spectra is ~ 5 cm⁻¹ Raman shift. Although this is comparable with the bandwidth of the broadband ns CARS spectrum in room air (see Fig. 3), in the flame, when N₂ molecules are distributed over a larger number of rotational states (at least ~30 cm⁻¹ Raman shift, see Fig. 3), the limited bandwidth of the ps diagnostics results in significant four-wave mixing signal reduction. In addition, since the four-wave mixing signal scales as N₂ number density squared, higher temperature in the flame reduces the signal further.

Single-shot ICCD images of the plasma generated by the discharge pulse in the hydrogen flame are shown in Fig. 4(a). These images are taken using a 20 ns camera gate, comprising the entire voltage pulse with peak voltage of 13 kV and FWHM of 10 ns. The plasma appears diffuse, without well-pronounced filaments, with good pulse-to-pulse reproducibility of the images. To illustrate evolution of the plasma emission intensity during the discharge pulse, Fig. 4(b) shows a sequence of 100-pulse average images taken with a camera gate of 1 ns. The emission decays approximately 6 ns after
breakdown, except for two regions near the ends of the electrodes. Plasma emission images taken in room air for the same discharge gap, with pulse peak voltage of 18 kV and FWHM of 10 ns, exhibit similar behavior.

Figure 5 shows four-wave mixing IR signal waveforms measured for peak electric field in three different cases: (a) room air with the gap between the electrode sleeves of \( d = 5 \) mm, when no breakdown is produced; (b) ns pulse discharge in room air for \( d = 1.3 \) mm; and (c) ns pulse discharge in the hydrogen flame, for the same gap of \( d = 1.3 \) mm. As discussed in Ref. [15], the duration of the IR waveforms, \(~ 10-40 \) \( \mu \)s, is controlled by the time constant of the IR detector preamplifier, and is much longer than the duration of the four-wave mixing signal, determined by the coherence decay time of \( N_2 \) molecules, a few hundred ps. For electric field measurements, the IR waveforms are integrated over \( t = 0-40 \) \( \mu \)s.

From Fig. 5, it is evident that peak electric field in the discharge in room air, proportional to the square root of the time-integrated IR signal intensity, is significantly higher compared to the peak electrostatic (no-breakdown) field between the electrodes, \( E_{\text{peak}} = U_{\text{peak}}/d' = 44 \pm 1.5 \) kV/cm, where \( U_{\text{peak}} = 26 \) kV and \( d' = d + 2\Delta d/s = 5.9 \pm 0.2 \) mm. Therefore extrapolating a calibration line obtained from the electrostatic field measurements toward higher electric fields achieved in the discharge may result in significant uncertainty. Instead, in this work the electric field in the discharge is inferred from comparison of the IR signal with the applied voltage pulse waveform before breakdown, when the electric field remains electrostatic. At the present conditions, this assumption is valid, since the electric field offset before the discharge pulse, due to residual charge accumulation on the electrode sleeves from the previous pulse, was found to be negligible.

Comparing the IR signals measured in ns pulse discharges in room air and in the hydrogen flame (see Fig. 5), it can be seen that the four-wave mixing signal in the flame is over two orders of magnitude lower compared to that in air. This occurs due to the significantly higher temperature (i.e., lower number density) in the flame, lower \( N_2 \) mole fraction in the combustion products compared to air, and limited bandwidth of the present diagnostics, which can access only several rotational transitions of \( N_2(v = 0\rightarrow 1) \) vibrational band, as discussed above. The background, obtained when the IR detector was blocked, shows that the signal-to-noise in the flame is approximately 10:1 and the effect of electromagnetic interference from the pulser on the IR detector is insignificant. Figure 6 compares the square root of the time-integrated IR signal in air, measured when the electrode sleeves are separated by \( d = 5 \) mm (at no-breakdown conditions), with the pulse voltage divided by the effective discharge gap, \( d^* = 5.9 \) mm. As expected, the two sets of data match well, demonstrating the validity of calibrating the electric field by the electrostatic voltage measured before the breakdown.

Time-resolved electric field in the discharge in air and in the hydrogen flame, for the gap between the electrode sleeves of \( d = 1.3 \) mm, calibrated by the electrostatic voltage before breakdown, is plotted in Fig. 7(a). Breakdown occurs when the field in the discharge gap begins to deviate from the applied voltage waveform, also plotted in Fig. 7(a). Breakdown field in air, \( E_{\text{peak}} = 75 \pm 7.5 \) kV/cm, is much higher compared to DC breakdown threshold estimated from Paschen law, \( \approx 30 \) kV/cm, due to the rapid voltage increase rate, \( \approx 4 \) kV/ns. After breakdown, charge separation and plasma self-

![Fig. 5. IR signal waveforms at peak electric field: “no-breakdown” conditions in room air (5 mm gap between the sleeves), discharge in room air (1.3 mm gap), and discharge in hydrogen flame (1.3 mm gap).](image1)

![Fig. 6. Pulse waveform and square root of the four-wave mixing signal in room air at no-breakdown conditions, with the electrode sleeves separated by 5 mm.](image2)
shielding result in rapid electric field reduction, to 10–15 kV/cm over 3 ns, while the plasma emission decays. The electric field remains below breakdown threshold during the rest of the applied voltage pulse. Although time resolution of the present measurements is approximately 2 ns, these results are consistent with the higher temporal resolution measurements in air, 0.2 ns, at similar conditions [17].

In the flame, breakdown occurs well before the applied voltage peaks (see Fig. 7(a)). Again, the field in the discharge gap begins to deviate from the applied voltage waveform at the breakdown moment. Similar to the discharge in air, in the hydrogen flame the electric field after breakdown is reduced rapidly, over several ns, while the plasma emission decays (see Fig. 4). Accurate time-resolved field measurements significantly before and well after breakdown are challenging due to low signal-to-noise, with measurement uncertainty at 20 kV/cm increasing to approximately ±35%. Breakdown field in the flame, $E_{\text{peak}} = 40 \pm 4$ kV/cm, is considerably lower compared to that in air, for the same discharge gap. Figure 7(b), plotting electric field data and voltage waveforms in the discharge in air and in the flame for a larger gap between the electrode sleeves, $d = 1.7$ mm, exhibits very similar results to Fig. 7(a), including breakdown field values and electric field behavior before and after breakdown.

Comparing the results of electric field measurements in air and in the hydrogen flame, it is clear that peak electric field in the flame is significantly lower, approximately 40 kV/cm vs. 75 kV/cm. Although it is somewhat difficult to compare these values directly, since the pulse voltage waveforms and the gas mixture compositions for these two cases are different, the breakdown field reduction is almost certainly due to the higher temperature (i.e., lower number density, N) in the flame, which results in a higher reduced electric field, E/N, during the pulse. As an illustration, Fig. 8 compares the electric field measured during breakdown in room air, at the conditions of Fig. 7(a), with the analytic solution of coupled electron transport, ion transport, and Poisson equations, predicting time-dependent electric field and electron density in the air plasma during pulsed breakdown in a one-dimensional dielectric barrier discharge [21]. Analysis of the model predictions indicate that the electric field in the plasma during breakdown is controlled only by electron impact ionization and electron transport by the applied field, resulting in rapid charge separation and plasma self-shielding on a ns time scale. On a ~1 ns time scale, electron recombination and attachment are too slow to affect electron kinetics and transport.

From Fig. 8, it is evident that the breakdown field predicted in room temperature air,
$E_{br} = 73 \text{kV/cm}$, is in good agreement with the data. The analytic solution overpredicts the rate of electric field reduction during breakdown, since ionization coefficient dependence on $E/N$, used by the model, assumes instantaneous relaxation of the electron energy distribution, which overpredicts the rate of ionization. Also, the measured electric field does not decrease all the way to zero, as predicted by the analytic solution, reaching a minimum of $|E| \approx 10-13 \text{kV/cm}$ at $t \approx 3-12 \text{ns}$ (see Fig. 8). This may be caused by plasma asymmetry, generating non-zero electric field component in the direction parallel to the electrode sleeve surfaces (see Fig. 2), as well as by incomplete plasma self-shielding near the ends of the electrodes, resulting in non-zero electric field, as suggested by the plasma emission images in Fig. 4.

According to the model predictions, increasing the temperature of air from $T = 300 \text{ K}$ to $1500 \text{ K}$, while keeping the voltage pulse waveform the same, would result in earlier breakdown, by approximately $2.5 \text{ ns}$, and reduce the breakdown voltage by nearly a factor of two, from $E_{br} = 73 \text{kV/cm}$ to $40 \text{kV/cm}$. This is consistent with the experimental data taken in air and in the hydrogen flame (see Fig. 7). Although increasing the temperature (i.e., reducing the number density) results in a lower breakdown field, this effect is not nearly as strong as may be expected based on the exponential dependence of the ionization coefficient on the reduced electric field, $\alpha/N \sim A \exp[-B/(E/N)]$ [21]. The breakdown field decreases approximately proportional to the square root of the number density. This behavior is due to the lower ionization frequency and more rapid transport (charge separation) in the plasma during breakdown at lower densities, which results in plasma self-shielding, electric field reduction, and termination of electron impact ionization. Peak electron density generated by the discharge pulse at $T = 300-1500 \text{ K}$ varies within only a factor of 2, due to the same mitigating effect. Peak reduced electric field during breakdown increases as the temperature is increased from $T = 300 \text{ K}$ to $1500 \text{ K}$, from $E/N = 300 \text{Td}$ to $800 \text{Td}$ ($1 \text{Td} = 10^{-17} \text{Vcm}^2$). At these conditions, most of the discharge power in air and fuel-air mixtures goes to electronic excitation of nitrogen, dissociation of oxygen, and ionization, generating highly reactive species [1]. Note, however, that this model cannot be used for quantitative predictions of the electric field during breakdown in reacting flows and in flames, since it assumes that both the temperature and the chemical composition of the mixture are known.

4. Conclusions

In the present work, electric field during ns pulse discharge breakdown in room air and in an atmospheric pressure hydrogen diffusion flame has been measured by ps four-wave mixing. The measurements have been done in diffuse plasmas generated in plane-to-plane geometry. Electric field is put on the absolute scale using the Laplacian field measured before breakdown. The results show that peak electric field during breakdown in the flame, approximately $40 \text{kV/cm}$, is significantly lower compared to that in room air, $75 \text{kV/cm}$, due to the higher temperature (lower number density) of the combustion products. Both in air and in the flame, the electric field follows the applied voltage before breakdown and decreases rapidly after breakdown, due to charge separation and plasma self-shielding.

The electric field in air is compared with the predictions of an analytic model of ns pulse breakdown, showing good agreement between the predicted and the measured breakdown field. The model also predicts earlier breakdown and breakdown voltage reduction as the temperature is increased, in qualitative agreement with the experimental data. Peak reduced electric field predicted by the model varies from $E/N = 300 \text{Td}$ to $800 \text{Td}$, indicating that most of the discharge input energy goes to electronic excitation of nitrogen, dissociation of oxygen, and ionization, generating reactive species. A detailed kinetic model incorporating combustion chemistry, electron impact ionization, electron attachment, electron-ion and ion-ion recombination, electron and ion transport, charge accumulation on dielectric surfaces, as well as plasma chemical reactions is needed to obtain quantitative insight into the electric field dynamics during and after breakdown. Comparison with the present data can be used for validation of kinetic models and for assessment of their predictive capability.

Although the sensitivity of the present ps four-wave mixing diagnostics in room air is $3-4 \text{kV/cm}$, its use for measurements of electric fields below $\sim 20 \text{kV/cm}$ in atmospheric pressure flames is challenging, due to low signal-to-noise. One of the reasons for this, along with the higher temperature and lower $N_2$ fraction in the combustion product mixture, is the limited bandwidth of the Stokes beam generated by the stimulated Raman scattering process, which provides access only to several rotational levels of $N_2(v = 0 \rightarrow 1)$ vibrational band. At these conditions, the use of a ps broadband dye laser to generate the Stokes beam would be more effective. The present diagnostics will have much better sensitivity in high-pressure flames, since the four-wave mixing signal scales as the squared number density of nitrogen. The detection limit in flames maintained at pressures above $\sim 3 \text{ Bar}$ is estimated to improve to below $\sim 2 \text{kV/cm}$, far below breakdown threshold. The present method determines the electric field averaged over the four-wave mixing coherence length (several cm at the present conditions). The use of a shorter focal distance lens would result in a significantly higher spatial resolution, controlled by the Rayleigh range of the lens.
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