Contribution of water droplets to charge release by laser filaments in air

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We measured the electric charge release from single water microdroplets illuminated by ultrashort laser filaments in air. This charge is up to 600 times larger than from a comparable filament volume in air. In contrast, for atmospheric droplet concentrations and sizes, the volume-averaged overall droplet contribution to the charge is small as compared with that of the filaments along its whole propagation path. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3220066]

Self-guided filaments are generated by ultrashort laser pulses[1–4] through a dynamic balance between Kerr self-focusing and defocusing by the free electrons released from the propagation medium by the pulse itself. Filaments can propagate over distances beyond 100 m,[5] be initiated remotely[6] and propagate through fogs and clouds,[7,8] turbulence,[9] or reduced pressures.[10] Hence, they are ideally suited for atmospheric applications.[4,11] In particular, charges released by the filaments provide an electrically conducting path for high-voltage discharges[12] or lightning control,[13] and assist water nucleation[11] in the atmosphere in a similar way as cosmic rays or other ionizing particles do.[14]

In wet meteorological conditions or under rain, droplets hit by the laser ionize and contribute to the generation of electric charges in the atmosphere. On the timescale of the pulse duration, spherical droplets focus the beam onto a nanometric hot spot, where the ionization efficiency is strongly increased.[15] Then, the droplet explodes within a few microseconds[16] due to the energy released by the pulse. At this time, both the local charge release at the hot spot and the charge stabilization close to the particle surface yield an inhomogeneous charge distribution within the droplet. As a consequence, individual fragments resulting from droplet explosion bear a net charge, which can further ionize the surrounding atmosphere. Up to now, this charge had neither been estimated nor considered in models of filamentation in the atmosphere.

In this letter, we estimate the contribution of water droplets to the laser-induced electric charge release along laser filaments. We show that it is much higher than the charge released by a comparable volume of the filament in air. However at typical atmospheric droplet densities, their spatially averaged contribution is smaller than that of air. As a consequence, the charges released by the droplet either rapidly neutralize each other or can be considered as a secondary process in lightning control experiments using laser filaments.

The Helvetera platform[17] delivered laser pulses of up to 27.5 mJ energy and 65 fs Fourier-limited duration (420 GW peak power) at a wavelength of 800 nm and 100 Hz repetition rate. The slightly diverging beam ($f/D=1400$) had an initial diameter of 2×2.4 mm. In a first configuration (Fig. 1), the laser was slightly focused by an $f=2.8$ m lens. 1-m-long laser filaments started at the nonlinear focus $\sim$3.5 m downstream, between two planar electrodes of 1×1 cm that were swept along the beam. Alternatively, the beam was strongly focused between the electrodes by an $f=5$ cm lens.

One electrode was set to a potential of +2 kV, while the other one was grounded through a 27 kΩ resistor. The time-integrated voltage at the resistor, measured with 12.5 kHz bandwidth, yielded the total collected charge, assuming a constant dielectric permittivity.

A piezoelectric nozzle (Microdrop MD K 140, and MD E 201H driver) launched $\sim$100 μm diameter water droplets, synchronized so that each laser shot hit a droplet between the electrodes. The data acquisition was triggered by a sonometric detector[18] recording the acoustic shockwave of the droplet explosion to ensure its presence in the beam. The experiment was performed at atmospheric pressure, 20–22 °C temperature and relative humidity of 30–35%.

We detected charge only on the high-voltage (positive) electrode, excluding the detection of positive ions. The laser conditions had little influence on the time of flight (TOF) of the collected charges, excluding fragments which ejection speed strongly depends on the incident laser intensity.[16] Finally, we collected almost no free electrons, since their mobility (10^4 cm^2/s V in air[19]) would lead to a TOF of 25 ns to the positive electrode, much longer than the picosecond time scale of attachment. In contrast, the average TOF of the detected charge carriers amounts to 50–65 μs, consistent with the cm^2/s V range of the O_2^- ions mobility in a weakly ionized plasma.[19] Our relatively slow measurement therefore mostly focuses on the O_2^- ions, which are representative of the charge generated in the filaments.[20] More precisely, the electron density after the laser shot is governed by

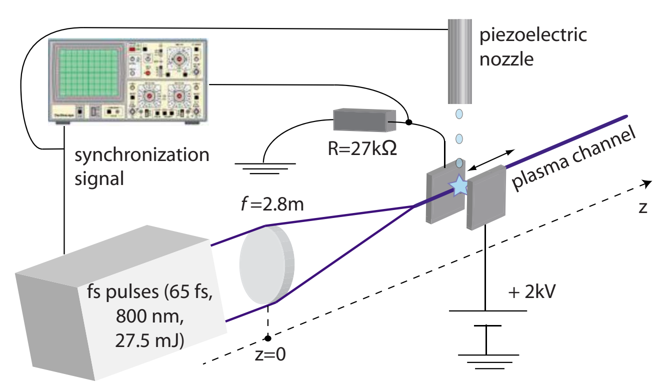

FIG. 1. (Color online) Experimental setup.

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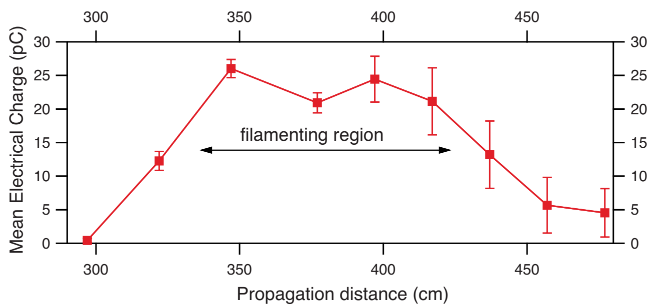

FIG. 2. (Color online) Longitudinal dependence of the charge collected from laser filaments in air, following the typical charge distribution in filaments. Error bars: one standard deviation over 150 measurements.

$dN_e/dt=-\eta N_e-\beta N_e^2$, where η and β, respectively, represent the attachment to O_2 molecules (yielding O_2^- ions) and the recombination with positive ions. In air at atmospheric pressure, $\eta=7.5\times10^6$ s^{-1} and $\beta=3.9\times10^{-8}$ cm^3 s^{-1},[21] so that 5.8% of the electrons undergo attachment and generate O_2^- ions.

The longitudinal dependence of the charge collected along a bundle of 3–5 filaments in air, for 22 mJ pulses of 65 fs Fourier-limited duration (Fig. 2) follows the typical plasma density in filaments.[18] This confirms that our measurement is representative of the charge generated in the filament and validates the detection technique. Considering three plasma channels with a typical electron density of 10^{16} cm^{-3} in the 10 μm diameter core of the filaments, the 1 cm long filament section located between the electrodes bear ~3.8 nC. We therefore collect almost 0.5% of the initial electron density, i.e., almost 9% of the generated O_2^- ions. This limited collection efficiency is due to a partial neutralization of the O_2^- ions by recombination during their TOF to the electrodes.[22]

Inserting a 100 μm droplet in the middle of the filament bundle ($z=375$ cm) increases the collected charge by several pC [Fig. 3(a)]. The droplet contribution steeply increases from 8 to 24 pC above 120 GW incident power, i.e., when the pulse intensity reaches several hundreds of GW/cm^2, sufficient to fragment the droplets[16] even if they are not hit by the filament itself. Below 120 GW, the charge release by the droplet is almost constant, consistent with the intensity clamping within the filaments.[23] The threefold increase in the charge released by the droplet, as compared with the 1 cm long section of the filaments bundle, corresponds to a local 600-fold increase for a comparable volume.

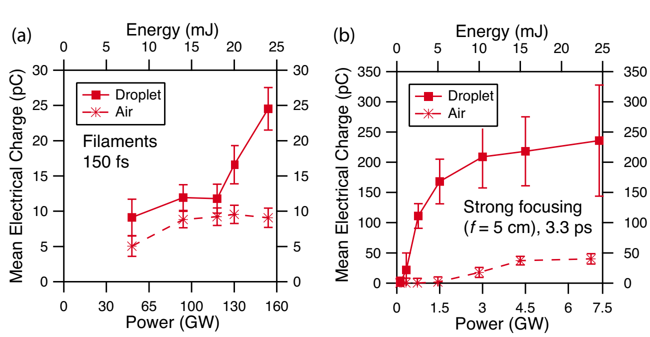

FIG. 3. (Color online) Power dependence of the charge collected in air with or without laser-droplet interaction. (a) Self-guided filaments generated by a slightly chirped pulse. (b) Focused beam ($f=50$ mm). Error bars: one standard deviation over 500 measurements.

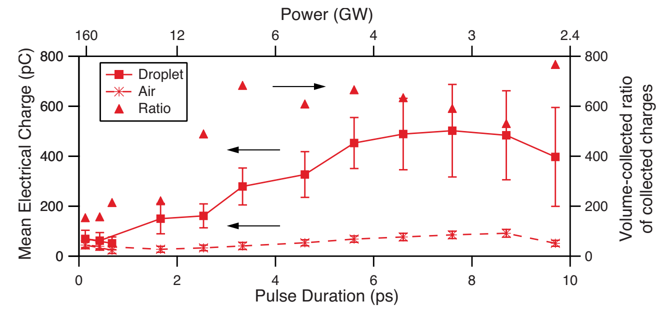

FIG. 4. (Color online) Chirp dependence of the charge emission by air and water droplets under illumination by strongly focused laser pulses of 24 mJ energy. Right scale: relative increase in the collected charge due to the laser-droplet interaction.

In contrast, the charge emitted by the droplet in a strongly focused incident beam [Fig. 3(b)] continuously varies with the incident power until 3 GW (2×10^{14} W/cm^2 at the 80 μm diameter waist) and then saturates, reaching a value about ten times the charge released in air in similar focusing conditions. Such enhancement corresponds to a factor of 900 when comparing with an equivalent volume of air illuminated by a strongly focused beam. The continuous increase in the emitted charge is due to the fact that the intensity at the focus is proportional to the incident power, contrary to the clamped intensity of the filaments.

Although the *local* charge release by the droplets is strong as compared to that released by the filaments in the air, the *spatially averaged* contribution is moderate. A typical cloud density of 1 $droplet/mm^3$ (Ref. 24) corresponds to 10 droplets per meter along a 100 μm diameter filament. The droplets therefore cover only 0.1% of the filament volume, which averages their 600-fold local enhancement in the charge release to only 60% of the total generated charge. Typical atmospheric cloud particles are however 10 to 100 times smaller than in our experiments, and therefore release much less charges. As a consequence, the contribution of atmospheric droplets to the charge release is marginal in the action of ultrashort laser pulses in the atmosphere, e.g., in the context of lightning triggering. However, they could locally contribute to the droplet growth through electrostatic collapse resulting in larger, more stable droplets.

Both the absolute charge emitted by a droplet and its relative contribution to the total generated charge are higher for longer, chirped pulses up to 3.5 ps (Fig. 4). Longer pulses ionize water more efficiently by allowing cascade ionization, contrary to subpicosecond pulses. The charge release in the air depends less on the pulse duration, since the contribution of avalanche ionization for durations below 10 ps keeps moderate (e.g., 12% for $I=10^{12}$ W/cm^2). As a consequence, self-compression in the filaments[25] is expected to limit the charge release efficiency of the droplets illuminated by filaments, and therefore to reduce the contribution of atmospheric aerosols to the ionization. Moreover, since the pulse duration inside the filaments depends little on that of the incident pulses, the initial chirp of the pulses does not influence much the charge released by the filaments in air.

As a conclusion, we have characterized the contribution of individual water droplets to the electric charge generated in air by filamenting as well as strongly focused ultrashort laser pulses. Droplets of 100 μm diameter significantly enhance the local charge generation under filament illumination, up to a factor of 600 at the droplet scale. However, actual atmospheric aerosols have a negligible space-averaged contribution to the atmospheric ionization and their influence on the action of laser filaments on the electric activity of thunderclouds is negligible. The electrostatic collapse of droplets of opposite charge may however contribute to droplet growth in subsaturated atmospheres[11]

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