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Using 100 TW laser pulses, we demonstrate that laser-induced nanometric particle generation in air increases much faster than the beam-averaged incident intensity. This increase is due to a contribution from the photon bath, which adds up with the previously identified one from the filaments and becomes dominant above 550 GW/cm^2. It appears related to ozone formation via multiphoton dissociation of the oxygen molecules and demonstrates the critical need for further increasing the laser energy in view of macroscopic effects in laser-induced condensation. © *2011 American Institute of Physics*. [doi:10.1063/1.3646397]

During the last decades, efforts have been dedicated to seed clouds with small particles of carbonic ice, AgI, or salts.[1–3] We recently proposed[4] an alternative approach relying on self-guided filaments[5–9] generated by ultrashort laser pulses. Laser filaments result from a dynamic balance between Kerr self-focusing and defocusing by the self-generated plasma[5–9] and/or negative higher-order Kerr terms.[10,11] They convey a typical intensity of 5×10^{13} W/cm^2 at kilometer-range distances,[12] generating large amounts of oxidized species like O_3, NO, and NO_2, which subsequently generate hygroscopic HNO_3.[13] The latter allows binary HNO_3–H_2O condensation well below 100% relative humidity (RH),[14] in a similar manner to the well-known H_2SO_4–H_2O binary condensation.[15–18]

However, work up to now was restricted to moderate laser energies (some hundreds of mJ or less) and powers (a few TW), which are unable to initiate macroscopic effects on large atmospheric volumes. Here, we investigate the effect of a jump by more than one order of magnitude in the laser energy and power. We show that at such level, the production of nanoparticles increases much faster than the beam-averaged incident laser intensity due to the atmospheric activation not only within the filament volume, but also in the much wider volume of the photon bath, i.e., the beam portion surrounding filaments, which conveys a substantial amount of the beam energy. This result illustrates the critical need for ultra-high power atmospheric lasers to induce macroscopic amounts of condensed particles.

Experiments were performed with the DRACO laser of Forschungszentrum Dresden-Rossendorf, a Ti:Sa chirped pulse amplification (CPA) chain providing up to 3 J, 100 TW pulses of 30 fs duration, at a repetition rate of 10 Hz and a central wavelength of 800 nm. The pulse energy was adjusted by rotating a half-waveplate associated with a polarizer, placed before the grating compressor, while its duration was controlled by tailoring the pulse using a Dazzler located at the exit of the pulse stretcher and/or by detuning the grating compressor. The beam was launched into air as bursts of several minutes, collimated with a diameter of ~10 cm. The beam-averaged incident intensity was evaluated from the measured pulse energy, duration, and diameter. The number of filaments in each experimental condition was characterized by single-shot burns on photosensitive paper (Kodak Linagraph 1895).

After ~7.5 m of propagation, up to ~900 filaments were generated.[19] From this location, the filamenting beam propagated through an open diffusion chamber ($110 \times 40 \times 40$ cm inner dimensions)[20] filled with ambient air. The temperature and RH in the chamber were controlled by a heated water reservoir at its top, and a fluid circulator at a temperature of −15 °C on its bottom. The RH and temperature were permanently monitored by two independent thermocouples and capacitance hygrometers, which yielded consistent results within 0.1%. During the measurements, the RH ranged within 75%–95%, at a local temperature of 8–12 °C.

The aerosol generation was characterized by a nanoparticle sensor (Grimm Nanocheck 1.320), which counts and evaluates the median diameter of nanoparticles between 25 and 300 nm. This device was sampling at 2 cm distance from the laser beam. The size distribution and number density of larger particles was controlled with an aerosol spectrometer (Grimm 1.107). Measurement cycles without laser provided control conditions, while the room background particle concentration was monitored outside of the chamber by a second set of identical devices.

The photon bath contribution to condensation was further investigated by recording the production of ozone from the Helvetera platform, a Ti:Sa CPA chain providing 24 mJ pulses in 62 fs, centred at 800 nm, at a repetition rate of 100 Hz. Ozone is indeed a key component in the generation of hygroscopic HNO_3 at the root of laser-induced condensation. The beam, with initial diameter of 2.5 cm, was slightly focused with an $f = 3$ m lens. The pulses were chirped up to 7.2 ps duration to ensure that no filaments were generated. A

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6.5-cm long metal cell of 5 cm diameter, with an air inlet of 7 cm^2 on its side and open windows at its two ends (typically <5 mm diameter, adapted to the beam size), was inserted at the beam focus, where a beam cross-section of 1.06 mm^2 was evaluated from impacts on photosensitive paper. Opposite to the air inlet, the air was continuously pumped through a polyethylene tube by an ozone analyzer (Horiba APOA-350 E). When the laser was operating, the ozone concentrations increased by several parts per billion in volume (ppb) to several tens of ppb above the few-ppb background. However, these values include a dilution factor of the generated ozone in our setup, which could not be determined precisely, so that in the following, ozone generation is discussed in arbitrary units. The intensity and fluence at the cell location were adjusted by varying the incident energy and the pulse chirp.

Launching the DRACO 100 TW laser beam into the diffusion chamber results in a steep rise of the nanoparticle density of 50–60 nm median diameter. Conversely, the concentration of particles in the 250 nm–32 μm range varies little, so that laser-induced fragmentation of larger pre-existing particles or the re-condensation of matter released by their laser-induced vaporization can only account for 5%–10% of the increase of the nanoparticle concentration of most of the presented data. This figure never exceeds 30%, ensuring a net condensation of matter in the process of nanoparticle formation.

Three regimes are observed in the case of Fourier-limited pulses (Fig. 1). Below ~150 GW/cm^2, no filaments are generated at the chamber location, and the nanoparticle concentration is unaffected by the laser. Above this threshold, the nanoparticle density increases parallel to the filament number, as expected from a fixed condensation rate in filaments, due to the intensity clamping.[21,22] However, above ~500 GW/cm^2, the effect of the laser increases faster than linearly while the filament number saturates due to spatial constraints.[19] These diverging behaviours evidence a substantial contribution from the photon bath in the high-intensity regime. Such contribution of the photon bath to a non-linear process at extreme average incident intensity has indeed been observed in the case of white-light generation.[23]

The contribution of the photon bath to laser-induced condensation is further evidenced by considering the yield of nanoparticles when increasing the pulse duration at a fixed fluence (Fig. 1(b)). Substantial particle generation is observed even for pulses chirped up to 2.3 ps, i.e., for an intensity as low as 10 GW/cm^2 preventing filament formation in the chamber. In this regime, the particle generation depends on the fluence rather than on the intensity.

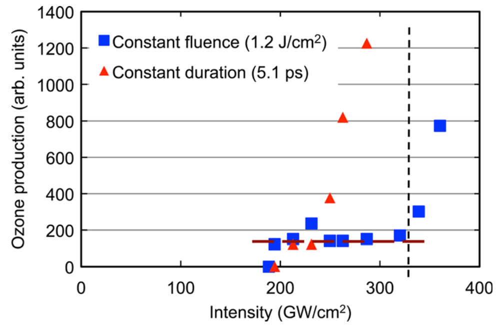

FIG. 2. (Color online) Ozone production by non-filamenting ultrashort laser pulses. The purple dashed line evidences the plateau observed at low intensity, corresponding to the linear, fluence-governed part of the photon bath contribution. The vertical black dashed line marks the threshold for detectable ionization.

Laser-induced condensation therefore appears to stem from three contributions. The first one originates from the filaments, while the photon bath offers both a non-linear, intensity dependent term and a linear, fluence-driven one at high fluence and low intensity, i.e., for long pulses. Owing to the limited number of experimental data points, the precise order of the non-linear contribution from the photon bath could not be achieved although it appears to lie between 5 and 8. Eighth and fifth order processes may respectively be identified as the ionization of oxygen and its multiphoton dissociation at the root of ozone production.[24,25]

The photon bath contribution to condensation is confirmed by the observation of ozone generation from filament-free beams at an intensity as low as 150 GW/cm^2. At this level, a sonometric setup[26] detects no ionization of the air. Ozone generation exhibits the same dual-component dependence on both fluence and intensity as observed for the generation of nanoparticles in the photon bath of the DRACO laser beam (Fig. 2). At low intensity, the ozone production is governed by the incident fluence. Conversely, at high intensity, it rises non-linearly with the incident intensity, as expected for multiphoton dissociation of oxygen molecules.[24]

The fluence-dependent contribution to particle and ozone production from the photon bath at low intensity may be due to mid-IR atmospheric photochemistry,[27] like the excitation of organic peroxyl radicals (RO_2)[28] or water-cluster mediated chemistry.[29] These radicals, originating from the volatile organic compounds present in the atmosphere, subsequently generate hydroxyl (OH) or hydroperoxyl (HO_2) radicals which are known to contribute to ozone formation. Note that the ppb-levels of ozone detected in the corresponding low-intensity conditions are compatible with this interpretation. Incoherent processes with time constants well

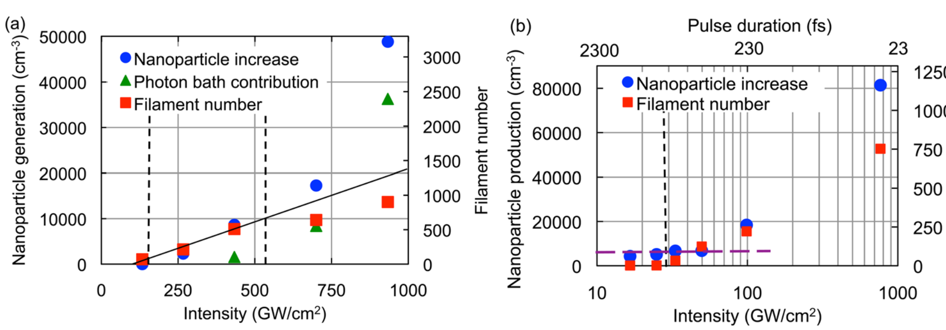

FIG. 1. (Color online) Dependence of laser-induced condensation on the beam-average incident intensity at 8–12 °C and 75%–90% RH. (a) Constant pulse duration of 30 fs. The solid line extrapolates a linear increase. (b) Constant incident fluence of 23 mJ/cm^2. The black dashed line displays the filamentation threshold under the considered conditions.

beyond the pulse duration, like thermal- or shockwave-related processes may also contribute. While a detailed study of these processes active at low intensities is beyond the scope of the present work, their identification will be crucial to anticipate the relevance of high-fluence, low-power, "long" pulses in the picosecond-range and potentially offers alternative ways to optimize the laser conditions for atmospheric experiments.

From a more general point of view, the efficient condensation induced by the photon bath provides an unexpected perspective on laser-assisted water vapour condensation over macroscopic scales. The active volume of the beam including the photon bath is typically 10^3–10^4 times larger than that of the filaments only. Such wider activated volume is favourable to an efficient use of the water vapour available in the atmosphere for condensing. It therefore demonstrates the critical need to use the highest possible laser power and energy available and improves the prospects for macroscopic effect of laser pulses on precipitation modulation.

As a conclusion, laser-induced nanometric particle generation occurs well below the filamentation threshold and increases much faster than the beam-averaged incident intensity due to the contribution of the photon bath, which adds up to the effect of the filaments. This contribution from the whole beam volume could offer a perspective to generate macroscopic effects in laser-induced condensation provided high energy lasers are used.

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