**Interaction of Ultrashort and Ultra-intense Laser Pulses with Aerosols and Clouds**

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Non-linear spectroscopy using ultra-short and ultra-intense laser pulses has recently appeared as a very promising method to probe the atmosphere[1]. Particularly attractive applications are related to non-linear interactions with atmospheric aerosols such as, for example, remote detection and identification of bioaerosols or highly efficient data transmission through fog and clouds.

We investigated both theoretically and experimentally multi-photon induced processes in aerosol particles using femtosecond laser pulses. More specifically, we demonstrated that both multi-photon (n = 1, 2 and 3 photons) induced fluorescence (MPEF) and laser induced breakdown (LIB) emissions are strongly enhanced in the backward direction[2,3]. This remarkable behavior is of particular relevance for remote sensing techniques such as Lidar (Light Detection and Ranging). The backward enhancement increases from 1.8 to 35 (emission ratio between the backward direction and 90°) with increasing non-linear process order n. MPEF and LIB are efficient methods to analyze the composition of the aerosol particles, and in particular to detect the presence of biological signatures from vitamins (e.g. riboflavin) and amino acids (tryptophan, tyrosin). We demonstrated the first range-resolved detection and identification of biological aerosols in the air by non-linear Lidar[4]. Ultrashort laser pulses from the Teramobile[1] have been used to induce two-photon-excited fluorescence (2PEF) in riboflavin containing particles at a remote location. We show that, in case of amino acids detection, 2PEF-Lidar should be more efficient than 1PEF-Lidar beyond a typical distance of 2 km, because it takes advantage of the higher atmospheric transmission at longer excitation wavelengths.

2PEF-Lidar might moreover provide simultaneous composition and size measurement by pump-probe schemes, which would help distinguish between bioaerosols and organics (soot). An important feature of femtosecond pulses is indeed very high temporal resolution (50 fs corresponds to the roundtrip time of a 1,8 um sized water droplet). Ballistic trajectories of optical wavepackets within microdroplets have been measured with a 2-photon 2 colors excitation scheme. The time delay dependence of the resulting 2PEF allowed a precise measurement of the particle size and distinguish between whispering gallery modes and rainbow-type trajectories[5].

At higher power levels, laser beams undergo filamentation as they propagate in air. An important question arises about the stability of filamentary structures as they interact with cloud droplets. We demonstrated that light filaments survive their interaction with water droplets as large as 95 μm, and that they are transmitted through clouds having an optical thickness as high as 3.2 (transmission 5 %)[6]. This remarkable transmission through turbid media results from a dynamic energy balance between the quasi-solitonic structure and the surrounding laser photon bath, which acts as an energy reservoir. Implications for free space laser communications, remote sensing and telemetry are discussed.

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