Perspectives of Remote Aerosol Analysis by Means of fs-LIDAR

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Non-linear spectroscopy using ultrashort laser pulses has recently appeared as a very powerful method to characterize molecules, liquids, solids and interfaces. We present recent results on multi-photon excited fluorescence (MPEF) from fluorophores (Coumarin, Tryptophan) imbedded in microparticles and white light generation in microdroplets. The results demonstrate the advantages of using ultrashort laser pulses, namely high peak power with low energy and high temporal/spatial resolution.

In particular, we demonstrated theoretically and experimentally that one-, two-, and three-photon excited fluorescence from dye molecules in spherical microcavities (microdroplets of 30 um radius) has an asymmetrical angular distribution and is enhanced in the backward direction [1]. This backward enhancement is particularly attractive for fs-LIDAR applications. Femtosecond excitation allowed to illuminate the microparticles at high intensity without shape deformation and competitive effects such as stimulated Brillouin scattering. The enhancement ratios (of intensities at 180° and 90°) is 9, 5, and 1.8 for three-, two-, and one-photon excitation, respectively. 3-PEF experiments in Tryptophan containing particles, which are typical simulants for bioaerosols, have also been carried out. As the 3PEF cross-section in Tryptophan is significantly weaker than in Coumarin dyes, higher incident intensities I_{inc} had to be used. At $I_{inc} > 10^{12}$ W/cm^2 we showed [2] that, in addition to 3-PEF, a highly localized nanometric plasma is generated within the microparticles, at its internal geometric focus. White light is emitted from the plasma, again with high directionality in the backward direction (enhancement factor 35). Theoretical interpretation of this behavior, based on non-linear Mie theory, showed that localization of the emitting source occurs both for MPEF and LIB due to the spherical shape of the particle and to non-linearity, and that light emitted from this region is, by reciprocity, re-directed towards the illuminating source.

The other attractive feature of femtosecond pulses is very high temporal resolution. This allowed us to measure for the first time the ballistic trajectories of optical wavepackets within a microdroplet, using a pump-probe arrangement. The fluorophores were excited with two femtosecond laser pulses of different wavelengths, in a 2-photon 2 colors excitation scheme. The resulting fluorescence could then be used as a measure of the coincidences between the two wavepackets within the droplet. In these experiments, ballistic motion of the wavepackets has been observed on whispering gallery modes and rainbow trajectories. Cavity lifetimes have been determined from the energy loss after each roundtrip, which constitutes the first cavity ringdown experiment performed in a micrometer size droplet. We plan to apply these results directly in the atmosphere, within the frame of the Teramobile [3] project. Pump-probe 2-PEF will be performed on bioaerosol simulants, in order to remotely measure both composition and size of the microparticles.

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[2] C. Favre, V. Boutou, S. C.Hill, W. Zimmer, M. Krenz, H. Lambrecht, J. Yu, R. K. Chang, L. Woeste, J.P. Wolf, *Phys.Rev.Lett*. **89(3)** 035002 (2002)
[3] http://www.teramobile.org