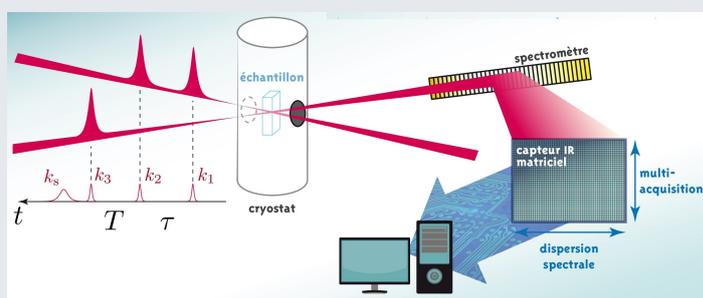


Towards the complete mapping of the vibrational dynamics of atmospheric acids by means of two-dimensional infrared spectroscopy

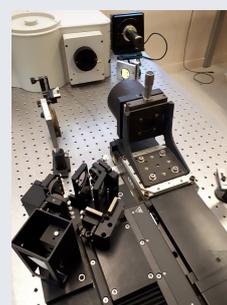
R. Thon (DR4), J.-P. Galaup (LAC), W. Chin, D. Chamma, C. Crépin (ISMO).

The advent of ultrafast laser sources in the infrared domain has enabled the study of a wide variety of systems through nonlinear four-wave mixing techniques, e.g. infrared photon echo and two-dimensional infrared (2D-IR) spectroscopy. The latter method possesses the unique property of giving access simultaneously to the structure and the dynamics of a system. It informs on the time-evolution of structural changes and on relaxation processes: loss of coherence, vibrational couplings, and energy transfers. Our project intends to provide a comprehensive description of the vibrational properties of clusters of small organic acids present in aerosols in order to better understand their role in larger aggregates formation. Dicarboxylic acids and related polar compounds are very abundant in aerosol particles so they can play an important role in the physical chemistry of aerosols. Although the key steps of the critical nucleus formation occur in the sub-2 nm size range, i.e. size of molecular clusters, crucial gaps remain and detailed information on small carboxylic acids is still lacking.

The experimental apparatus is under progress. The two-dimensional spectrometer is under test. The PALM financial support has enabled us to obtain funding from DEFINSTRUM and ANR to achieve the entire set-up. This has led to a new collaboration with ONERA for developing an innovative multipixel infrared detector based on 320x256 photovoltaic elements for improved resolution ($<1 \text{ cm}^{-1}$). Using 2D-IR spectroscopy and photo-reactivity experiments in controlled environments - especially in low temperature matrices - we will bring new insights into the physicochemical properties relevant to aerosol chemistry and in particular to particles formation.



2D-IR scheme based on a pump-probe geometry with nonlinear signal dispersed onto a grating



2D-IR setup and Mach-Zehnder interferometer

R. Thon, W. Chin, D. Chamma, J.-P. Galaup, A. Ouvrard, B. Bourguignon, and C. Crépin, *Vibrational spectroscopy and dynamics of $W(\text{CO})_6$ in solid methane as a probe of lattice properties*, The Journal of Chemical Physics 145, 214306 (2016)

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