Abstract

Spontaneous Raman scattering is a physical process that provides a unique knowledge of materials at the molecular level. Its high chemical specificity with no labels motivates its use in many different fields, ranging from biomedical research to industrial quality control. Nevertheless, the efficiency of this simple process is limited by its extremely weak cross-section.

Typically, the Raman scattered light is dispersed and collected onto an array detector, for several spatial positions of the sample, resulting in a hyperspectral image. Yet, this leads to the generation of overwhelmingly large data sets and to lengthy acquisitions. In situations where hyperspectral measurements simply aim to map the spatial distribution of molecules, the spectral data is unmixed in a postprocessing step, in order to detect molecular species and/or estimate their concentrations. In those cases, acquiring a complete vibrational spectrum per spatial pixel may be inefficient, and a massive speed-up can be achieved by encompassing compressive techniques in the acquisition process. Some strategies, including compressive Raman technology (CRT), use spectral *a priori* information to integrate chemometric analysis directly into the spectrometer hardware: the measurement is designed to directly probe quantities of interest to be estimated (e.g., molecular concentrations), rather than deducing them from complete hyperspectral measurements. In CRT, this is made possible by replacing the array detector by a single-pixel-detector, combined with a programmable optical filter. Based on the *a priori* known spectra of pure molecular species contained in the sample, these filters select accurately chosen spectral components and combine them into the detector.

This thesis develops some theoretical and technological aspects of CRT and applies it to several concrete applications. In a first part of the work, we investigate the estimation precision achieved by CRT, show that our method of estimation is efficient, and experimentally validate this analysis. In a second part of the work, we compare CRT, to some extent, to commercial state-of-the-art instrumentation. We find some clear advantages in terms of acquisition speed and limit of detection. We also show some preliminary results that suggest its usefulness for fields related to biomedical imaging, pharmaceutical industry and the environment. Last, we take further advantage of the single-pixel architecture of CRT to perform multiplexed line-scan imaging. We quantify the potential gain of this approach in terms of signal-to-noise ratio, when the measurements are shot-noise limited.