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Accelerating confocal Raman microscopy by capturing Essential Information in the Fourier Domain (EIFD)

Point-by-point Raman confocal hyperspectral imaging is a widespread technique through which both spectral and spatial information can readily be extracted, allowing one to have a global understanding of the samples under study. However, when the sample surface to analyze is large (i.e., for pharmaceutical specimens), its characterization may require from few hours to days, hampering specific analytical applications which require fast identification (i.e., falsified medicines) or can induce photodamage. To circumvent these issues, we propose an approach based on the assessment of Essential spectral Information in the Fourier Domain (EIFD) which permits to enhance confocal Raman imaging speed by 50-fold compared to classical point scanning [1]. The set of essential spectral profiles selected encompass the most linearly independent (dissimilar) spectra measured and the sole use of these spectra enables the reproduction of the collected data in a convex linear way [2-3]. The selection of EI has proven to be a very useful and reliable data reduction tool for linear spectral unmixing, it brings advantages in terms of speed and compression and allows challenging analytical issues such as the detection of very minor species to be tackled [4]. In EIFD, EI is evaluated from a convex hull analysis of the data point cloud in the 2D phasor plots displaying the Fourier coefficients estimated from the registered spectra for a few selected harmonics. One of the major assets of EIFD is that essentiality can be assessed independently for each spectral pixel and potentially as the data acquisition is ongoing. This paves the way to acquisition processes that would be based on the EI content of a spectral pixel and that would speed up the collection at those locations where information is not essential, considerably accelerating the whole imaging procedure. To illustrate our approach, we show outcomes obtained for several Raman hyperspectral imaging datasets resulting from the analysis of pharmaceutical samples of varying complexity (from handmade to commercial tablets) and outline a new data acquisition scheme relying on targeted sampling based on EIFD that could represent a real breakthrough in the domain of Raman confocal hyperspectral imaging. As an example, such a scheme permits to decrease the time required for the collection of a 101x101x293 Raman hyperspectral image from 14 hours to 28 min by identifying the essential information encoded in this image in the presence of noise and by increasing the spectral accumulation (and, thus, the spectral resolution) only when essential pixels are found. We show that in this way we are able to obtain very similar chemical information compared to when a standard analysis on the entire sample is conducted. Another advantage of EIFD is that it is broadly applicable since it is based on spectral frequencies and could be implemented to perform the acquisition of EI on-the-fly, opening the doors to the characterization of photosensitive biological systems.

References

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