Fast compressive Raman microspectroscopy

Fernando Soldevila[1], Jonathan Dong[2,3], Benneth Sturm[4], Enrique Tajahuerce[1], Sylvain Gigan[2], Hilton B. de Aguiar[4]

1. GROC·UJI, Institute of New Imaging Technologies (INIT), Universitat Jaume I, E12071Castellò, Spain.

2. Laboratoire Kastler Brossel, CNRS UMR 8552, Ecole Normale Supérieure, PSL Research University Sorbonne Universities & Université Pierre et Marie Curie Paris 06, F-75005, Paris, France

3. Laboratoire de Physique Statistique, CNRS, PSL Universités & Ecole Normale Supérieure, Sorbonne Universités et Université Pierre et Marie Curie, 75005, Paris, France

4. Département de Physique, Ecole Normale Supérieure / PSL Research University, CNRS, 24 rue Lhomond, 75005 Paris, France

E-mail: h.aguiar@phys.ens.fr

KEYWORDS: spontaneous Raman, microspectroscopy, compressive sensing, matrix completion

Spontaneous Raman microspectroscopy allows imaging a chemically heterogeneous system with high spatial resolution associated with superb molecular selectivity. Nevertheless, acquiring a large field-of-view hyperspectrum is slow and requires large data storage space. Lately, compressive sensing strategies have been used to speed up Raman microscopy by under-sampling the hyperspectrum, followed by computational reconstruction using efficient algorithms [1-6]. Most of these approaches are based on the use of inexpensive Digital Micromirror Device (DMD) that combines selected wavelengths in a single-pixel detector (typically faster and less noisy than conventional CCD-based hyperspectroscopy [3]). However, compressive spectrometer layouts have prohibitive losses for low-light levels applications, such as in the spontaneous Raman effect.

In this contribution, we present a new high-throughput programmable spectrometer layout with large spectral coupling bandwidths (>1500 cm^{-1}) [7] and its usage for fast Raman using the new matrix completion framework [8]. We exploit this spectrometer to implement a fast random wavelength selection sampling scheme of the hyperspectrum, a prerequisite necessary for using matrix completion algorithms. We then demonstrate 8X high-fidelity hyperspectral compression, with 5 ms effective pixel dwell times. By using a priori information of the hyperspectrum provided by the matrix completion algorithm, we show fast chemical imaging of biological specimens in a few seconds [9] (Fig. 1).

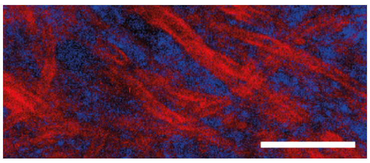

Figure 1. Representative results of the compressive Raman framework applied for imaging of biological specimens (brain tissues). The red stripes are the response of the lipids and blue represent protein-rich regions (red= lipids, blue=protein).

[1] P. Berto, C. Scotte, F. Galland, H. Rigneault, H. B. de Aguiar. "Programmable single-pixel-based broadband stimulated Raman scattering". Opt. Lett. **42,** 1696, 2017.
[2] F. Refregier, C. Scotte, H. B. de Aguiar, H. Rigneault, F. Galland. "Precision of proportion estimation with binary compressed Raman spectrum". J. Opt. Soc. Am. A **35**, 125, 2018.
[3] C. Scotte, H. B. de Aguiar, D. Marguet, E. Green, P. Bouzy, S. Vergnole, P. Winlove, N. Stone, H. Rigneault. "Assessment of compressive Raman versus hyperspectral Raman for microcalcification chemical imaging". Anal. Chem. **90**, 7197, 2018.
[4] D. S. Wilcox, Gregery T. Buzzard, B. J. Lucier, O. G. Rehrauer, P. Wang, D. Ben-Amotz, "Digital compressive chemical quantitation and hyperspectral imaging", Analyst **138**, 4982, 2013.
[5] D. F. Galvis-Carreño, Y. H. Mejía-Melgarejo, and H. Arguello-Fuentes, "Efficient reconstruction of Raman spectroscopy imaging based on compressive sensing". Dyna 81, 116, 2014.
[6] J. V. Thompson, J. N. Bixler, B. H. Hokr, G. D. Noojin, M. O. Scully, V. V. Yakovlev, "Single-shot chemical detection and identification with compressed hyperspectral Raman imaging", Opt. Lett. **42**, 2169, 2017.
[7] B. Sturm, F. Soldevila, E. Tajahuerce, S. Gigan, H. Rigneault, H. B. de Aguiar, "High-sensitivity high-speed compressive spectrometer for Raman imaging", arXiv:1811.06954, 2018.
[8] E. J. Candes and B. Recht, "Exact matrix completion via convex optimization," Foundations of Computational Mathematics, vol. 9, no. 6, pp. 717–772, 2009.
[9] F. Soldevila, J. Dong, E. Tajahuerce, S. Gigan, H. B. de Aguiar, "Fast compressive Raman bio-imaging via matrix completion", arXiv:1811.12389v1, 2018.