

# Time-gated fourier-transform raman imaging achieves high temporal and spectral resolution with SPAD arrays

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## Abstract

Fourier-transform time-gated Raman imaging is proposed and demonstrated. By developing a detection arm utilizing a SPAD array and a high performing interferometer, the system can achieve both high temporal and spectral resolution and strong separation between Raman and fluorescence signals.

**Keywords:** Time-gated Raman, Imaging, SPAD, Fourier transform

Raman spectroscopy is a powerful optical technique used across the physical and life sciences to determine the chemical components and structure of a sample. Raman is attractive as it is both label-free and chemically specific. However, Raman scattering is a physically inefficient process, with only approximately 1 in  $10^7$  photons Raman scattered and is thus often dwarfed by autofluorescent background signals which may be multiple orders of magnitude higher. To mitigate this, researchers have attempted to exploit the distinct temporal dynamics of these physical processes to separate Raman and autofluorescent signals in a technique known as time-gated Raman spectroscopy<sup>1</sup>. This is possible as Raman scattering occurs effectively instantaneously, whereas fluorescence is excited and emitted at nanosecond timescales.

In recent years, time-gated Raman systems based on single-photon avalanche diodes (SPAD) arrays have emerged as promising candidates to achieve high quality time-gated Raman imaging<sup>2,3</sup>. They exhibit extremely high time resolution (approx. 100 ps), are sensitive to single photon events and are inherently parallelizable. Typical

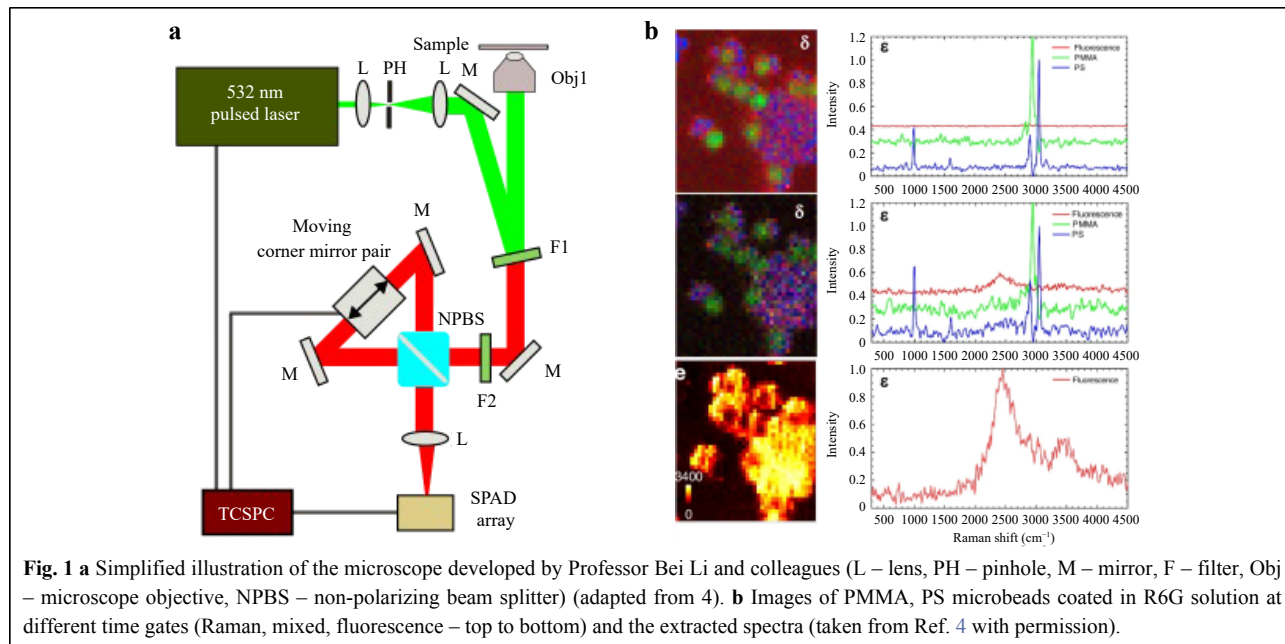
systems utilize a pulsed laser temporally synchronized with a SPAD array with the collected Raman scattering dispersed on the detector chip with a grating. In Ref. 4, published in *Light: Science and Applications*, Professor Bei Li from the Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences and colleagues developed a new time gated Raman imaging system utilizing a Fourier transform detection scheme. This system can achieve temporal resolution on the order of hundreds of picoseconds and broad spectral coverage from  $-1,000$  to  $10,000\text{ cm}^{-1}$ , with spectral resolution of  $0.05\text{ cm}^{-1}$ . The principle of the optical design is shown in Fig. 1a. A pulsed visible laser is expanded and reflected into a microscope objective. The resulting Raman scattering is then collected by the objective and transmitted through the filter. This beam is then split by a non-polarizing beam splitter to form each arm of the interferometer which are then reflected back toward each other. Between each arm, two corner mirrors mounted on a displacement stage are used to reflect each path back through the system and onto the SPAD array. During the acquisition, the displacement stage is moved at a constant velocity and precise photon arrival time (relative to the last pulse of the laser) and coarse photon arrival time (relative to start of acquisition) are

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acquired and stored within an array. To accurately recover the spectral information, the position of the stage was monitored in real time at nanometric accuracy. The utility of the system was tested to image PMMA and PS microbeads that were mixed with R6G to induce a background fluorescent signal. The extracted images are shown in Fig. 1b at different temporal windows (Raman, Raman/fluorescence mix and fluorescence). The component spectra were then extracted by applying a multivariate technique demonstrating the robust separation of Raman and fluorescence achieved by the authors optical technique.

The techniques developed in this work will potentially find applications in many areas of Raman imaging. Time-gated Raman has recently been used to Raman image strongly autofluorescent tissue samples using visible light<sup>5</sup> with potential for integration into clinics under ambient conditions<sup>6</sup>. In addition, the instrument will be of interest in studies of crystallinity and gas-phase Raman which typically require higher spectral resolution than conventional Raman systems. These developments will be supported by ongoing improvements in SPAD array development supporting higher temporal resolution, and sensitivity<sup>7</sup>. Such advances pave the way for compact, high-throughput, and fluorescence-free Raman imaging systems suitable for both research and clinical diagnostics.

#### Data availability

All data are available from the corresponding authors upon reasonable request.

#### Conflict of interest

The author declares no conflicts of interest.

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