A new design of a fibre-optic probe system is presented which can be used to simultaneously detect scattering signals from multiple channels for spatially offset Raman spectroscopy and Raman tomography in reflection mode. The fibre setup contained 32 source fibres with an auto-controlled optical switch and 40 detector fibres, which were deployed in a large-area planar interface. Phantom experiments were conducted using a prototype of the Raman instrument, and have demonstrated the practical applicability of the system design. The presented area-detection instrument provides an efficient platform for various reflectance applications of Raman modalities over a large area. It could also be potentially adopted for other diffuse optical-based spectroscopic and tomographic applications.

Introduction: Raman spectroscopy (RS) has been extensively studied for in vitro and in vivo diagnosis of diseases and malignancies in various tissues via probing vibrational energies in molecules [1,2]. Recently, the penetration depth of RS was advanced up to several centimetres by spatially offset Raman spectroscopy (SORS) [3–5]. Furthermore, researchers have started to investigate Raman tomography (RT) by integrating Raman scattering into the diffuse optical tomography modality [6–8]. However, the majority of the current RT studies adopted an angular interface for data collection with a focus on transmission measurement. Such transmission designs can be powerful in imaging small limbs. However, in clinics when the body trunk is involved, performing SORS and RT in reflection mode will be inevitable.

In this Letter, we present a new custom-designed fibre-optic Raman probe system that is suitable for SORS and RT in reflection mode. This system contains 32 illumination and 40 detection units in a large planar interface. SORS and RT experiments were conducted to evaluate the system performance with phantoms, and have demonstrated the practical applicability of the instrument.

Materials and methods: Fig. 1 illustrates the Raman system, comprising a 785 nm continuous-wave laser, a Raman spectrograph, a CCD camera, a custom designed fibre-optic probe, and an auto-controlled 1 × 32 optical switch. The latter receives input from the laser source via a fibre, and couples to 32 output channels where the 32 source fibres are connected. This switch controls the sequential illumination of the source fibres, and ensures that only one source fibre is shooting at one time.

Fig. 1 Schematic of our Raman experimental setup

The prototype of the fibre-optic probing interface is shown in Fig. 2. It consists of a fibre probe head, a spectrometer fibre adaptor, 32 source fibres and 40 detector fibres. Multimode fibres with a 105 μm core were used for both illumination and collection. At the probe head, the proximal end of all fibres was bonded inside 1/16-inch-diameter aluminium tubings, which were fixed in even-spaced holes (Fig. 2c) of a 6 × 6 × 2 cm³ aluminium block. The spacing between adjacent holes was 5 mm. The holes were regularly deployed in rows and columns. The source (red columns) and detector (blue columns) fibres were arranged in alternating columns with 8 × 4 and 8 × 5 rectangle patterns, respectively. The minimum and maximum offsets between the source and detector fibres were 5 and ~50 mm, respectively. A 60 × 4 × 1 mm³ slot was made at each column, where multiple customised laser line filters (excitation) and long-pass edge filters (emission) were installed (Fig. 2a).

Fig. 2 Photographs of area-detecting fibre-optic probe prototype with customised excitation and emission filters (Fig. 2a); exemplary projected 40-spectral-band array on CCD camera, formed by signals emerging from 40 detector fibres (Fig. 2b), and its probe head (Fig. 2c)

Fig. 3 SORS spectra taken by fibres 9, 17, 25, 33 at the spatial offset Δs = 5, 15, 25, 35 mm, respectively, (upwards), pure Teflon (bottom) and pure HDPE (top) (Fig. 3a). Ratio of HDPE peak (1296 cm⁻¹) to Teflon peak (733 cm⁻¹) against Δs detected by fibres in row 2 (black curve) and by fibres in column D (blue curve), when the source fibre at (C, 2) shot (Fig. 3b). Calculated Δs distribution against position (column, row) of probe head (Fig. 3c). Measured ratio of HDPE peak (1296 cm⁻¹) to Teflon peak (733 cm⁻¹) against position (Fig. 3d)

The distal end of the 40 collection fibres was aligned into a customised spectrometer fibre adaptor (Fig. 2a) in a vertical line, and then installed at the entrance of a spectrometer to project their output signals into the spectrometer. Fig. 2b shows a photograph of an exemplary projected 40-spectral-band array on the CCD camera of the spectrometer, and the band array was formed by signals emerging from the 40 detector fibres. As such, signals from each detector fibre form a horizontal image on the CCD camera, leading to a specific spectrum for each detector fibre.

Experimental results and discussion: First, SORS experiments were performed using the setup. SORS allows differentiation of Raman signals originating from layers at different depths. The fibre probe was used to test a two-layer sample containing one 1 mm-thick 15 × 15 cm Teflon sheet atop one 60 mm-thick 15 × 15 cm high density polyethylene (HDPE) block. The fibre probe was placed directly on top of the sample, contacting the Teflon surface. The excitation power was set at 150 mW for all experiments. The source fibre at the position (column, row) = (C, 2) was used to shot the sample. Fig. 3 shows the results. We measured the peak ratio of 1296 cm⁻¹ (HDPE) to 733 cm⁻¹ (Teflon) against the spatial offset Δs between the source and detector.
fibres. Fig. 3a shows the Raman spectra of pure Teflon and pure HDPE, and four SORS spectra taken by fibres 9, 17, 25 and 33 at Δz = 5, 15, 25 and 35 mm, respectively. It indicates that the Raman spectrum at Δz = 5 mm contained a substantial contribution from the top Teflon layer. As Δz increased, HDPE’s contribution (e.g. peak 1296 cm⁻¹) gradually increased with respect to Teflon (e.g. peak 733 cm⁻¹). Hence, the ratio of HDPE to Teflon increased as Δz increased. It suggests this probe can be used for deep-layer Raman detection while suppressing Raman perturbation from the surface layers. Fig. 3b shows ratios of the HDPE peak (1296 cm⁻¹) to the Teflon peak (733 cm⁻¹) against Δz for SORS spectra taken by fibres in row 2 (fibres 9, 17, 25, 33, black curve) and by fibres in column D (fibres 9–16, blue curve). We note that the black and blue curves nearly overlapped with each other. Hence, it shows that the detector fibres in both row and column can provide reliable SORS measurement results in terms of the spatial offset.

Fig. 4 Raman spectrum of SaAuNPs (Fig. 4a). 2D Raman images of SaAuNPs–gel cylinder (Fig. 4b). (i)-(iii) Simulation results. (i) X−Y section at Z = 9.5 mm; (ii) X−Z section at Y = 0 mm; (iii) Y−Z section at X = 0 mm. (iv)–(vi) Experimental results. (iv) X−Y section at Z = 9.5 mm; (v) X−Z section at Y = −2 mm; (vi) Y−Z section at X = 2.2 mm. (ii)–(iv) Image size is 20 × 20 mm². 3D RT image (image cube size 20 × 20 × 20 mm³) (Fig. 4c).

Figs. 3c and d show the calculated spatial offset Δz for all 40 detector fibres and the measured ratio of the HDPE peak (1296 cm⁻¹) to the Teflon peak (733 cm⁻¹) when the source fibre shot at (C, 2) (red star). The Δz and ratio were deployed against the position (column, row). The results show that the ratio of HDPE to Teflon increased with Δz, and the trend of the ratio variation agreed well with that of the Δz variation. It further indicates that this probe can provide a reliable SORS measurement.

We then tested this system on a tissue phantom to demonstrate its capability in RT imaging. A cylinder hole of 5 mm diameter by 5 mm height filled with gelatine gel (10% w/v) containing 8 pM gold nanoparticles with surface enhanced Raman scattering (SERS) signals. Afterwards, a piece of 10 mm-thick lean pork tissue (named top tissue) and four SORS spectra taken by SaAuNPs and four SORS spectra taken by fibres in both row and column can provide reliable SORS measurement results in terms of the spatial offset.

Fig. 4b illustrates the simulation (i)-(iii) and experimental (iv)-(vi) results from RT imaging. The dimension variance of the inversion results was ∼5%. The centre of the RT image was not exactly located at the origin and slightly shifted to (2.2, −2.0, 0 mm). The misplacement could be caused by the incorrect positioning of the probe centre. The 3D RT image (Fig. 4c) showed a narrowed end in the deep (Z) instead of a perfect cylinder shape, which agrees with the drill tip. These results indicate this Raman system can efficiently work for RT imaging.

Conclusion: We have custom designed a new area-detection fibre system and demonstrated its practical applicability for SORS and RT in reflection mode. This reflection-mode system made new system designs with a brand-new probe layout with alternating source and detector fibres and multiple excitation and emission filters. It also provided a deployment of an optical switch for auto-controlled illumination from multiple source fibres that results in an adequate amount of illumination/collection patterns with pre-set locations and thus ensures the imaging resolution. This instrument offers an efficient platform for various reflectance applications of Raman modalities over a large area. This Raman system could offer new avenues for diagnostic spectroscopy, imaging and image-guided treatment. It could also be potentially adopted for other diffuse optical-based spectroscopic and tomographic applications.

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