Supporting Information for

A Rapidly Modulated Multifocal Detection Scheme for Parallel Acquisition of Raman Spectra from a 2-D Focal Array

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Figure S1

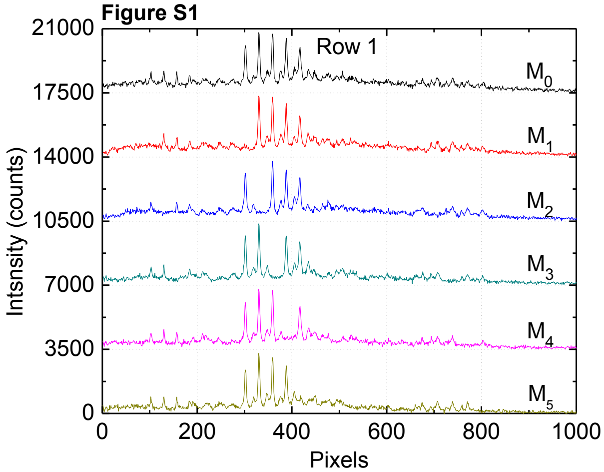


Figure S2

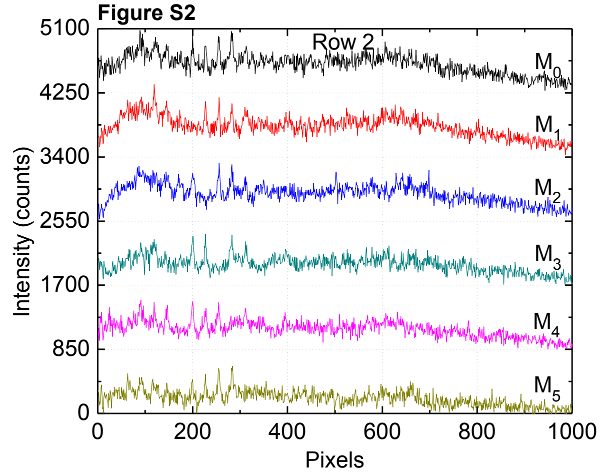


Figure S3

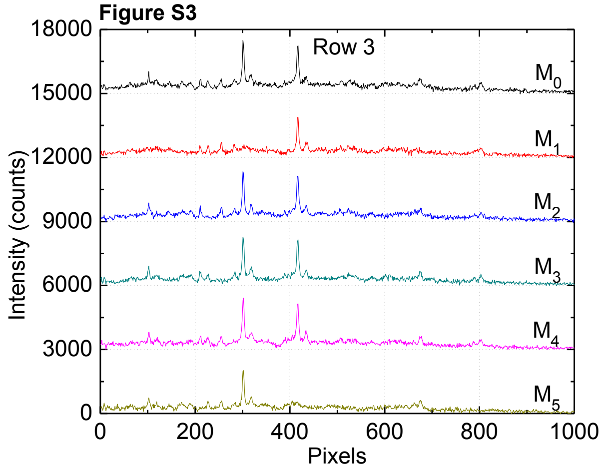


Figure S4

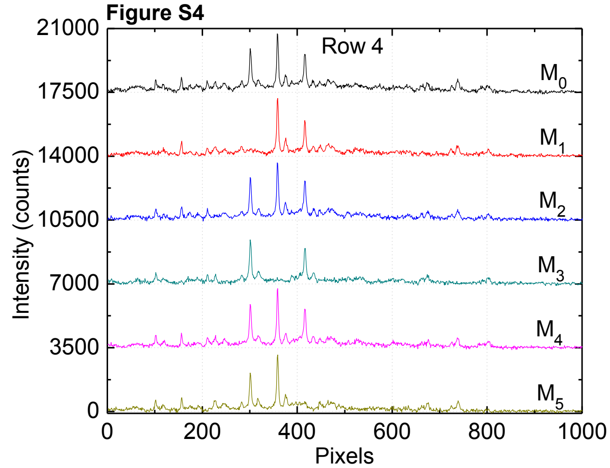
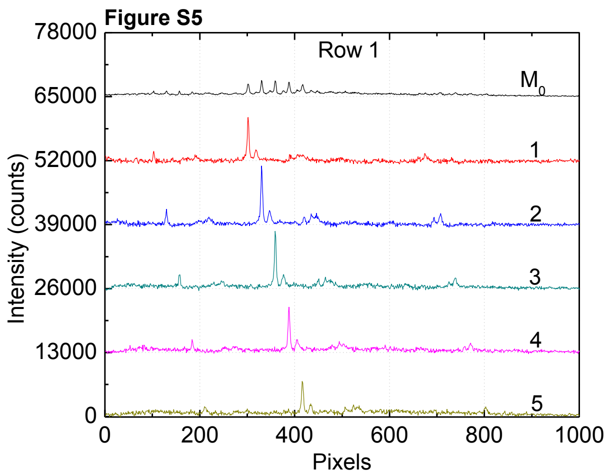


Figure S1 – S4: Superimposed Raman spectra of the different rows of trapped polystyrene and PMMA beads in the 4 × 5 laser tweezers array shown in **Figure 3(A)** for different matrix patterns. M_0 is the initial detection pattern where no columns are blocked. M_i (i = 1, … 5) are the five different Raman measurement patterns as shown in equation (1.2).



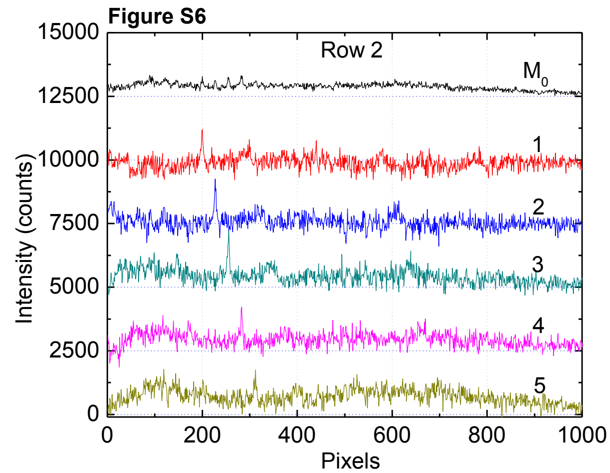


Figure S5 and S6: Superimposed and reconstructed Raman spectra of rows 1 and 2 of trapped polystyrene and PMMA beads in the 4 × 5 laser tweezers array shown in **Figure 3(A).** M_0 is the initial detection pattern where no columns are blocked. Raman spectra labeled as n = 1, … 5 are the reconstructed individual spectra as shown in **Figures 3(C) and 3(D)**.

Table S1

Intensity of Raman peak (counts)		1	2	3	4	5
Row 1: polystyrene	Measured spectra	2025	2734	2650	2421	1944
	Reconstructed spectra	8782	11934	11329	8741	6546
Row 2: PMMA	Measured spectra	384	289	399	423	211
	Reconstructed spectra	1445	1743	1824	1376	1181

Signal-to-noise ratio		1	2	3	4	5
Row 1: polystyrene	Measured spectra	35.2	47.5	46.0	42.0	33.8
	Reconstructed spectra	47.8	79.6	74.7	56.7	44.7
Row 2: PMMA	Measured spectra	6.8	5.2	7.1	7.5	3.8
	Reconstructed spectra	9.0	9.9	10.8	9.0	6.4

Table S1: Raman peak intensities (1001 cm^{-1} of polystyrene and 813 cm^{-1} of PMMA) and signal- to-noise ratios of Raman spectra in **Figure S5** and **S6** for trapped particles 1 to 5. Measured spectra: Raman spectra (2 second acquisition) shown in the top of **Figure S5** and **S6;** they are the measured spectra with initial detection pattern where no columns are blocked. Reconstructed spectra: Reconstructed individual Raman spectra (8 second acquisition) in rows 1 and 2 after the post data processing using equation (1.4). The signal- to- noise ratio was estimated by using the peak intensity (at 1001 cm^{-1} for polystyrene or at

813 cm^{-1} for PMMA) divided by the peak-to-peak noise of the background (standard deviation of the background between 1800 to 1900 cm^{-1}).

Detection Patterns

The five simple mask patterns (matrices in (1.2)) presented in the main text are just one example of the modulated detection that can be used for the 4 × 5 ($m \times n$) laser tweezers array. The following five mask patterns are another example:

$$M_1 = \begin{pmatrix} 0 & 1 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 & 1 \end{pmatrix}, M_2 = \begin{pmatrix} 1 & 0 & 1 & 0 & 0 \\ 1 & 0 & 1 & 0 & 0 \\ 1 & 0 & 1 & 0 & 0 \\ 1 & 0 & 1 & 0 & 0 \end{pmatrix}, M_3 = \begin{pmatrix} 0 & 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 & 0 \end{pmatrix},$$

$$M_4 = \begin{pmatrix} 0 & 0 & 1 & 0 & 1 \\ 0 & 0 & 1 & 0 & 1 \\ 0 & 0 & 1 & 0 & 1 \\ 0 & 0 & 1 & 0 & 1 \end{pmatrix}, M_5 = \begin{pmatrix} 1 & 0 & 0 & 1 & 0 \\ 1 & 0 & 0 & 1 & 0 \\ 1 & 0 & 0 & 1 & 0 \\ 1 & 0 & 0 & 1 & 0 \end{pmatrix}; \quad \text{(S1. 1)}$$

The individual Raman spectra of the $m'th$ row can be reconstructed by the following linear equations:

$$\begin{pmatrix} 0 & 1 & 0 & 0 & 1 \\ 1 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 & 1 \\ 1 & 0 & 0 & 1 & 0 \end{pmatrix} \begin{pmatrix} I_{m1} \\ I_{m2} \\ I_{m3} \\ I_{m4} \\ I_{m5} \end{pmatrix} = \begin{pmatrix} I_1^m \\ I_2^m \\ I_3^m \\ I_4^m \\ I_5^m \end{pmatrix}; \quad \text{(S1. 2)}$$

The solutions for these equations are:

$$\begin{aligned} 2(I_{m1}) &= I_1^m + I_2^m - I_3^m - I_4^m + I_5^m; \\ 2(I_{m2}) &= I_1^m + I_2^m + I_3^m - I_4^m - I_5^m; \\ 2(I_{m3}) &= -I_1^m + I_2^m + I_3^m + I_4^m - I_5^m; \\ 2(I_{m4}) &= -I_1^m - I_2^m + I_3^m + I_4^m + I_5^m; \\ 2(I_{m5}) &= I_1^m - I_2^m - I_3^m + I_4^m + I_5^m; \end{aligned} \quad \text{(S1. 3)}$$

For a specific $m \times n$ laser tweezers array, the best signal to noise ratio of the reconstructed spectra can be realized by choosing the appropriate/optimal detection patterns.