Supplementary Information

Raman Imaging Spectroscopy: History, Fundamentals and Current Scenario of the Technique

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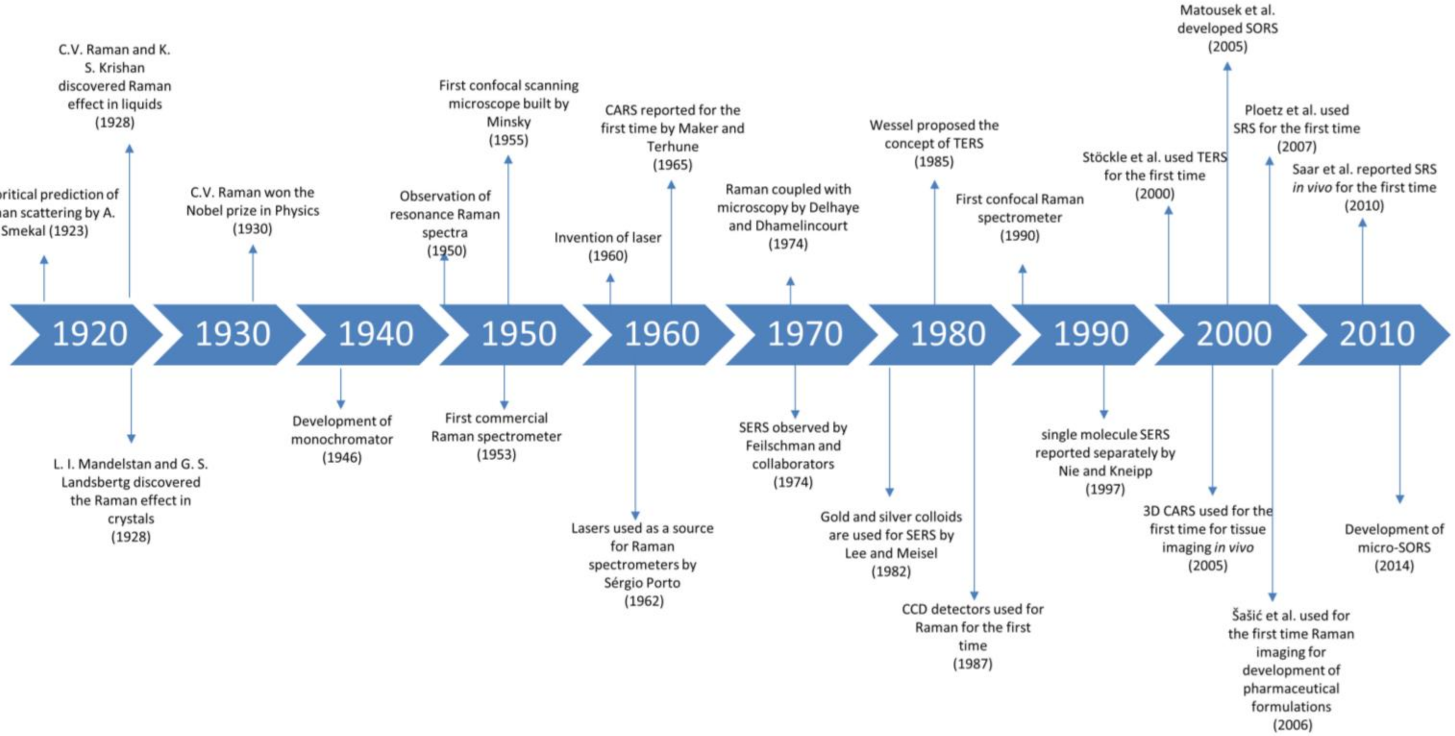

Figure S1. Timeline of events for Raman imaging spectroscopy (adapted from Smith *et al*.[1]).

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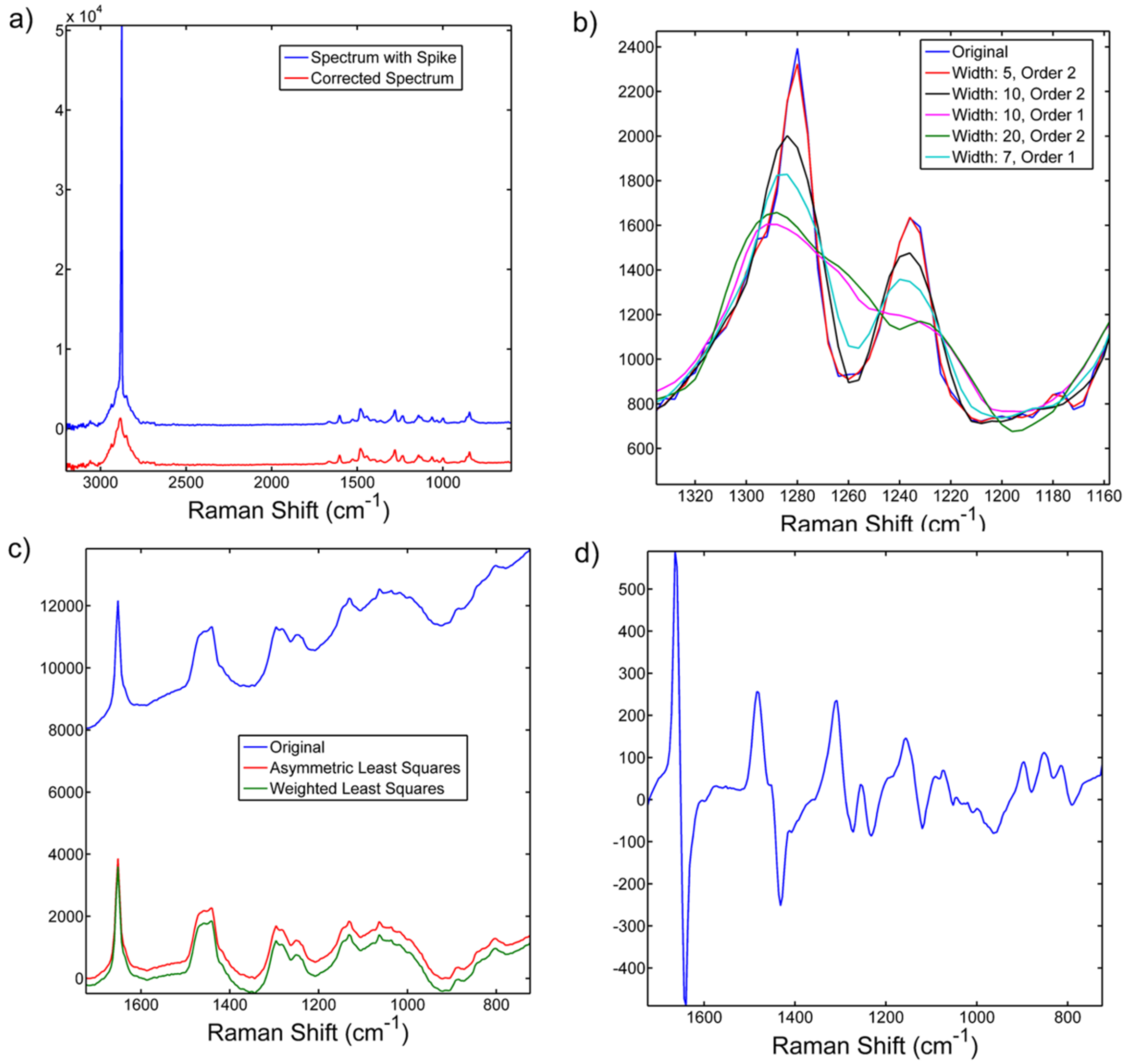

Figure S2. (a) Raman spectrum distortion caused by spike; (b) effects of different window sizes and polynomials on Raman spectrum smoothing; (c) spectrum with shift in the baseline corrected by asymmetric least squares or weighted least squares; (d) same spectrum shown in (c) corrected by the first derivative.

Table S1. Raman imaging applications in the areas of agriculture and biomass

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Methyl parathion (pesticide)	SERS	dispersive (633)	area	2
Amygdaline distribution in apricot seeds	spontaneous	dispersive (785)	intensity of peak and VCA	3
Organic matter in soils	spontaneous	dispersive (532.1)	intensity of peak	4
Carotenoids in internal maturation of tomatoes	SORS	dispersive (785)	intensity of peak	5
Characterization of cell wall for bioethanol production	spontaneous	dispersive (633)	area	6
Germination and growth of tomato seeds	spontaneous	dispersive (514.5)	area	7

SERS: surface enhanced Raman spectroscopy; SORS: spatially offset Raman spectroscopy; VCA: vertex component analysis.

Table S2. Raman imaging applications in the area of foodstuffs

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Carotenoids in tomato varieties	spontaneous	dispersive (785)	MCR	8
Polysaccharides on the cell wall of tomatoes	spontaneous	dispersive (532)	PCA and MCR	9
Surface and internal evaluation of pork and carrots	spontaneous and SORS	dispersive (785)	intensity of peak	10
Contamination of powdered foods	spontaneous	dispersive (785)	intensity of peak	11,12
Study about cheese and its components	spontaneous	dispersive (532)	intensity of peak and PCA	13
Barley composition and microstructure	spontaneous	dispersive (532)	BTEM	14
Cane sugar packed in a yellow plastic container	SORS	dispersive (785)	SMA	15

SORS: spatially offset Raman spectroscopy; MCR: multivariate curve resolution; PCA: principal component analysis; BTEM: band-target entropy minimization; SMA: self-modeling mixture analysis.

Table S3. Raman imaging applications in the environmental area

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Characterization of atmospheric particles	spontaneous	dispersive (632.8)	MCR	16-20
$CdSO_4$ and $CaCO_3$ particles in water	spontaneous	dispersive (632.8)	MCR-ALS and SIMPLISMA	21
Aquatic environment monitoring	spontaneous	dispersive (445)	intensity of peak	22
Carotenoids in lichens *Lecanoraceae*	RRS	dispersive (514)	intensity of peak	23
Study of lead contamination and transfer	spontaneous	dispersive (632.8)	intensity of peak	24
Detection of persistent organic pollutants	SERS	dispersive (532)	intensity of peak	25

RRS: resonance Raman spectroscopy; SERS: surface enhanced Raman spectroscopy; MCR: multivariate curve resolution; ALS: alternating least square; SIMPLISMA: simple-to use interactive self-modeling mixture analysis.

Table S4. Raman imaging applications in biological and medical area

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Differentiation between normal and carcinogenic cells	spontaneous	dispersive (514)	area; PCA; KMCA	26
Detection of leukaemia and lymphoma cells	SERS	dispersive (638)	CLS	27
Lipid structures	surface enhanced CARS	FT-Raman (1064)	intensity of peak	28
Image of colon tissue cells	spontaneous and CARS	dispersive (785)	intensity of peak; KMCA	29
Characterization of caries	SRS	(1064)	intensity of peak and PCA	30
Image of living cells	spontaneous and SERS	dispersive (532)	intensity of peak	31
Mapping of normal and cancerous oral mucosa	spontaneous	dispersive (532)	KMCA; PCA	32
Determination and location of vitamin E in biological simples	spontaneous	dispersive (633)	PLS; PLS-DA; PCA	33

SERS: surface enhanced Raman spectroscopy; CARS: coherent anti-Stokes Raman spectroscopy; SRS: stimulated Raman spectroscopy; PCA: principal component analysis; KMCA: k-means clustering analysis; CLS: classical least squares; PLS: partial least squares; DA: discriminant analysis.

Table S5. Raman imaging applications in instrumentation development

Objective	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Line-scan equipment for SORS	SORS	785	SMA	34
TERS mapping with AFM uplighting	TERS	633	intensity of peak	35
FT-CARS detected with wide-field	CARS	800	intensity of peak	36
Fast ICCD time filters to avoid fluorescence	spontaneous	266	intensity of peak	37
CARS under room light	CARS	798 and 1040	MCR	38
Shape smoothness restriction in MCR-ALS	spontaneous	dispersive (633)	MCR-ALS	39
Material optimization and tip shape for TERS	TERS	363.8	intensity of peak	40
Stand off Raman in remote samples	stand-off Raman	dispersive (532)	area	41

SORS: spatially offset Raman spectroscopy; TERS: tip enhanced Raman spectroscopy; AFM: atomic force microscopy; ICCD: intensified charge-coupled device; FT: Fourier transform; CARS: coherent anti-Stokes Raman spectroscopy; SMA: self-modeling mixture analysis; MCR: multivariate curve resolution; ALS: alternating least square.

Table S6. Raman imaging applications in geology/composition of planets and celestial bodies

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Aragonite in rocks in Corsica	spontaneous	dispersive (514.5)	PCA and correlation index	42
Mineral phase distribution in meteorite	spontaneous	dispersive (532, 632.8 and 785.14)	intensity of peak and ratio of peaks	43,44
Effect of grain size distribution on Raman analyses	spontaneous	dispersive (532)	intensity of peak	45
Effect of auto-fluorescence on Raman image	spontaneous	dispersive (514.5)	intensity of peak	46
Formation of aqueous solutions on Mars	spontaneous	dispersive (532)	intensity of peak	47
Gemology and mineralogy on mountain	spontaneous	dispersive (532)	intensity of peak and K-means	48-51

PCA: principal component analysis.

Table S7. Raman imaging applications in art, archeology and paleontology

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Evidence of amphora production site	spontaneous	dispersive (532)	intensity of peak	52
Treatment of conservation of cultural heritage	spontaneous	dispersive (785)	intensity of peak	53
Carotenoids in wall painting in the house of Marcus Lucretius (Pompeii)	spontaneous	dispersive (785 and 514)	intensity of peak	54
Raman image complementarity to micro XRF image	spontaneous	dispersive (785)	intensity of peak	55
Conservation status of 16th century paintings	spontaneous	dispersive (785 and 830)	intensity of peak and area	56-58
Glass painting	spontaneous	FT-Raman (1064)	intensity of peak	59
Mineral microbial structures in dinosaur fossils	spontaneous	dispersive (532)	intensity of peak; HCA	60

XRF: X-ray fluorescence; HCA: hierarchical cluster analysis.

Table S8. Raman imaging applications in material characterization

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
3D Raman image of rubber mixtures	spontaneous	dispersive (633)	intensity of peak	61
Corrosion study of materials	spontaneous	dispersive (532, 633 and 514)	intensity of peak area ratio	62-65
Ex and *in situ* studies Raman confocal of organic films and their transistors	spontaneous	dispersive (488)	intensity of peak	66
Ge nanofiber mapping	TERS	dispersive (532)	intensity of peak	67
Monitoring and manipulation of Si nanocrystals in Si-SiO_2 nanocomposites	RRS	dispersive (441.6, 488 and 514.5)	intensity of peak	68
Graphene doping and mapping	spontaneous	dispersive (514.5)	intensity of peak	69
Mapping of thin polymeric films	TERS	dispersive (633)	intensity of peak	70
Evolution of G and G' modes in graphene	spontaneous	dispersive (532)	intensity of peak	71

TERS: tip enhanced Raman spectroscopy; RRS: resonance Raman spectroscopy.

Table S9. Raman imaging applications in forensic analysis

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Shooting residues	spontaneous	dispersive (455 and 780)	intensity of peak; MCR; PLS-DA	72,73
Anthrax detection in correspondence	CARS	dispersive (532)	intensity of peak	74
Explosive in banknotes	spontaneous	dispersive (785)	ICA	75
Dynamite analysis	spontaneous	dispersive (532)	intensity of peak	76
SERS metafilm for fingerprints	SERS	dispersive (633)	intensity of peak and cls	77
Document ink analysis	spontaneous	dispersive (780)	intensity of peak	78
Fingerprint detection	spontaneous	dispersive (473 and 785)	intensity of peak;	79,80
Fingerprints contaminated with explosives	spontaneous	dispersive (532)	Pearson's correlation coefficient	81,82
Cocaine chloride on human nails	spontaneous	dispersive (785)	area	83
Detection of explosives	stand-off Raman	dispersive (229)	intensity of peak	84

SERS: surface enhanced Raman spectroscopy; CARS: coherent anti-Stokes Raman spectroscopy; MCR: multivariate curve resolution; PLS: partial least squares; DA: discriminant analysis; ICA: Independent components analysis.

Table S10. Raman imaging applications in the pharmaceutical area

Analyte	Type of scattering	Spectrometer (λ of laser / nm)	Data analysis	Reference
Homogeneity of powder mixture	spontaneous	dispersive (785)	CLS; PCA; MCR	85
Falsification of medicines	spontaneous	dispersive (783)	intensity of peak	86
Particle aggregation	spontaneous	dispersive (532)	PLS	87
Pharmaceutical films	spontaneous	dispersive (632.81)	PLS-DA	88
Crystalline and amorphous condition of drugs	spontaneous	dispersive (532.09)	CLS	89
Alteration of drug release	spontaneous	dispersive (785 and 532)	BTEM; area; CLS	90-92
Distribution of API and/or excipients	spontaneous	dispersive (785)	ICA; CLS	93,94
Drug-lipid interaction and molecular distribution	spontaneous	dispersive (632.8)	intensity of peak	95
Study of formulation production process	spontaneous	dispersive (785)	intensity of peak; CLS	96,97
Characterization of silica particles with drug	CARS	1064	intensity of peak	98
Homogeneity of formulation comparing chemometric methods	spontaneous	dispersive (532)	intensity of peak; CLS; MCR-ALS; PCR; PLS; iPLS; PLS-GA	99

API: active pharmaceutical ingredient; CLS: classical least squares; PCA: principal component analysis; MCR: multivariate curve resolution; iPLS: interval; partial least squares; DA: discriminant analysis; BTEM: band-target entropy minimization; ALS: alternating least square; GA: genetic algorithms.

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