



An Exhaustive Survey on Raman Spectroscopy instrumentation and its Applications

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Abstract

The analytical technology of Raman spectroscopy has an almost 100-year history. During this period, many modifications and developments happened in the method like discovery of laser, improvements in optical elements and sensitivity of spectrometer and also more advanced light detection systems. Many types of the innovative techniques appeared (e.g. Transmittance Raman spectroscopy, Coherent Raman Scattering microscopy, Surface-Enhanced Raman scattering and Confocal Raman spectroscopy/microscopy). This review article gives a short description about these different Raman techniques and their possible applications. Then, a short statistical part is coming about the appearance of Raman spectroscopy in the scientific literature from the beginnings to these days. The third part of the paper shows the main application options of the technique (especially confocal Raman spectroscopy) in skin research, including skin composition analysis, drug penetration monitoring and analysis, diagnostic utilizations in dermatology and cosmeto-scientific applications. At the end, the possible role of artificial intelligence in Raman data analysis and the regulatory aspect of these techniques in dermatology are briefly summarized. For the future of Raman Spectroscopy, increasing clinical relevance and in vivo applications can be predicted with spreading of non-destructive methods and appearance with the most advanced instruments with rapid analysis time.

Keywords: Artificial intelligence, Cosmetoscience, Raman spectroscopy, Skin composition, Skin diagnostics, Skin research, Topical drug penetration

Introduction

Non-destructive spectroscopic techniques represent the top-choice for any kind of process monitoring. Among all of the available techniques, Raman spectroscopy is one of the most solid and versatile tools to analyze several materials, both in lab and on-field conditions. Raman spectroscopy was first independently developed in the first half of the 20th century by the Nobel laureate Chandrasekhara Venkata Raman and Grigorij Samuilovic Landsberg, but it was established after the implementation of laser light equipped spectrometers in the second half of the century. The establishment of Raman spectroscopy opened the path to a more detailed knowledge about materials, with a particular emphasis on carbonaceous materials such as graphite.

Throughout the years, Raman analysis has grown, reaching several industrial sectors such the food and textiles sectors. Raman spectroscopy displays several advantageous features over other techniques like infrared spectroscopy. For example, the quality of the signal collected is barely affected by the presence of water, allowing for use in plenty of applications where infrared analyses are not reliable. A representative case study is the in-situ monitoring of a fermentative process where Raman techniques outperformed any other spectroscopic approach. Nonetheless, Raman analysis suffers from some difficulties such as the challenge of developing quantitative robust and trustworthy methods of data analysis. Furthermore, the presence of highly active Raman species such as carbon particles could mask the presence of other species. Several studies have been devoted to overcoming these drawbacks. The Raman spectroscopy is based on an inelastic light scattering by molecules (the Raman effect).

In the Raman scattering process, a photon interacts momentarily with a molecule and is then scattered into surroundings in all directions. During the brief interaction with molecule, photon loses or gains energy which is then detected and analyzed. The Raman effect occurs when electromagnetic radiation impinges on a molecule and interacts with the polarizable electron density and the bonds of the molecule in the phase (solid, liquid or gaseous) and environment in which the molecule finds itself. For the spontaneous Raman effect, which is a form of inelastic light scattering, a photon (electromagnetic radiation of a specific wavelength) excites (interacts with) the molecule in either the ground rovibronic state or an excited rovibronic state. This results in the molecule being in a so-called virtual energy state for a short period of time before an inelastically scattered photon results. The resulting inelastically scattered photon which is "emitted"/"scattered" can be either of higher (anti-Stokes) or lower (Stokes) energy than the incoming photon.

In Raman scattering the resulting rovibronic state of the molecule is a different rotational or vibrational state than the one in which the molecule was originally, before interacting with the incoming photon (electromagnetic radiation). The difference in energy between the original rovibronic state and this resulting rovibronic state leads to a shift in the emitted photon's frequency away from the excitation wavelength, the so-called Rayleigh line. The Raman effect is due to inelastic scattering and should not be confused with emission (fluorescence or phosphorescence) where a molecule in an excited electronic state emits a photon of

energy and returns to the ground electronic state, in many cases to a vibrationally excited state on the ground electronic state potential energy surface.

TYPES OF RAMAN SPECTROSCOPY

There are a number of advanced types of Raman spectroscopy, including surface-enhanced Raman, resonance Raman, tip-enhanced Raman, stimulated Raman (analogous to stimulated emission), transmission Raman, spatially offset Raman, and hyper Raman.

1. Surface-enhanced Raman spectroscopy (SERS) – Normally done in a silver or gold colloid or a substrate containing silver or gold. Surface plasmons of silver and gold are excited by the laser, resulting in an increase in the electric fields surrounding the metal. Given that Raman intensities are proportional to the electric field, there is large increase in the measured signal (by up to 10¹¹). This effect was originally observed by Martin Fleischmann but the prevailing explanation was proposed by Van Duyne in 1977.

2. Resonance Raman spectroscopy – The excitation wavelength is matched to an electronic transition of the molecule or crystal, so that vibrational modes associated with the excited electronic state are greatly enhanced. This is useful for studying large molecules such as polypeptides, which might show hundreds of bands in "conventional" Raman spectra. It is also useful for associating normal modes with their observed frequency shifts.

3. Hyper Raman – A non-linear effect in which the vibrational modes interact with the second harmonic of the excitation beam. This requires very high power, but allows the observation of vibrational modes that are normally "silent". It frequently relies on SERS-type enhancement to boost the sensitivity.

4. Tip-enhanced Raman spectroscopy (TERS) – Uses a metallic (usually silver-/gold-coated AFM or STM) tip to enhance the Raman signals of molecules situated in its vicinity. The spatial resolution is approximately the size of the tip apex (20–30 nm). TERS has been shown to have sensitivity down to the single molecule level and holds some promise for bioanalysis applications.

5. Stimulated Raman spectroscopy – A spatially coincident, two color pulse (with polarization either parallel or perpendicular) transfers the population from ground to a rovibrationally excited state, if the difference in energy corresponds to an allowed Raman transition, and if neither frequency corresponds to an electronic resonance. Two photon UV ionization, applied after the population transfer but before relaxation, allows the intra-molecular or intermolecular Raman spectrum of a gas or molecular cluster (indeed, a given conformation of molecular cluster) to be collected. This is a useful molecular dynamics technique.

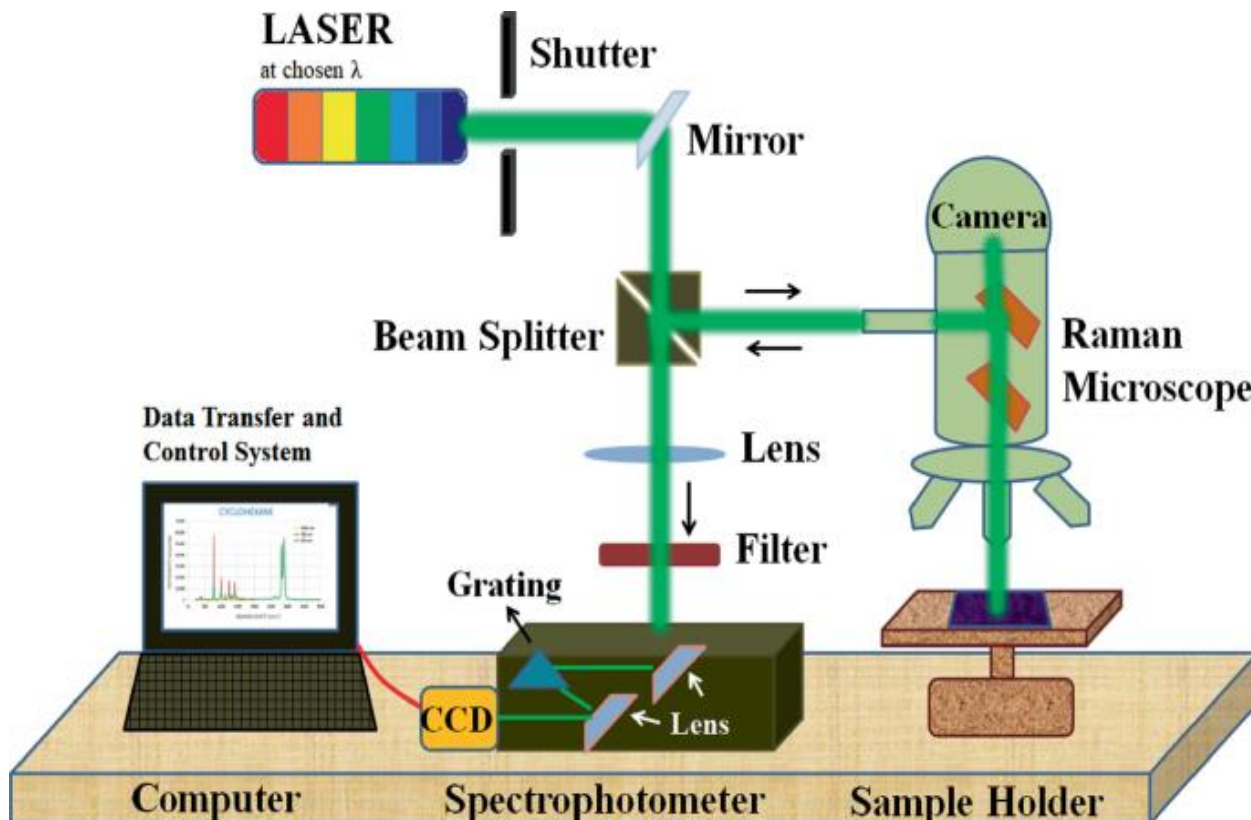
6. Transmission Raman – Allows probing of a significant bulk of a turbid material, such as powders, capsules, living tissue, etc. It was largely ignored following investigations in the late 1960s (Schrader and Bergmann, 1967) but was rediscovered in 2006 as a means of rapid assay of pharmaceutical dosage forms. There are also medical diagnostic applications.

7. Spatially offset Raman spectroscopy (SORS) – The Raman scattering beneath an obscuring surface is retrieved from a scaled subtraction of two spectra taken at two spatially offset points.

8. Surface plasmon polariton enhanced Raman scattering (SPPERS) – This approach exploits apertureless metallic conical tips for near field excitation of molecules. This technique differs from the TERS approach due to its inherent capability of suppressing the background field. In fact, when an appropriate laser source impinges on the base of the cone, a TM₀ mode (polaritonic mode) can be locally created, namely far away from the excitation spot (apex of the tip). The mode can propagate along the tip without producing any radiation field up to the tip apex where it interacts with the molecule. In this way, the focal plane is separated from the excitation plane by a distance given by the tip length, and no background plays any role in the Raman excitation of the molecule.

Instrumentation

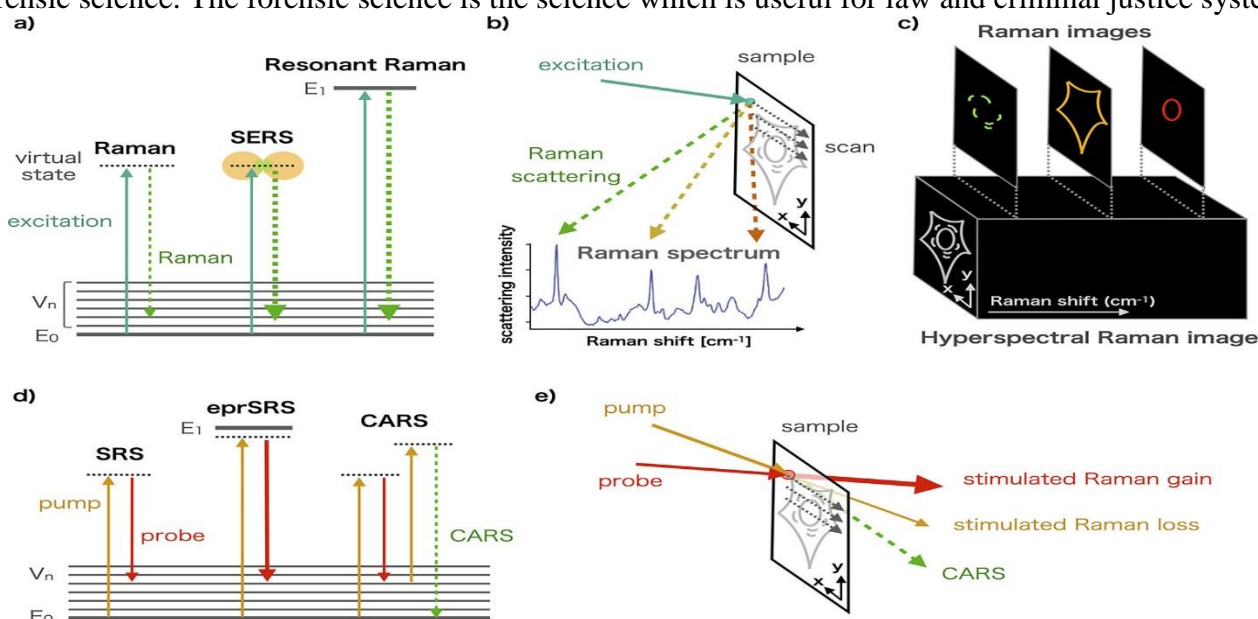
A source of monochromatic light, generally from a laser in a visible area, near infrared, or near UV radiation, although X-rays may not be used. Laser light interacts with vibrations of cells, pads, or other sensors in the system, causing the laser photon energy to be transferred up or down. Power switch provides information about vibration modes in the system. Infrared spectroscopy usually produces the same yet consistent information.



Typically, Laser beam is used to illuminate sample. Electromagnetic radiation from the illuminated area is collected through the lens and transmitted by a monochromator. Scattering radiation with elastic at wavelength along the laser line (Rayleigh scattering) is filtered by a notch filter, edge pass filter, or band pass filter, while other collected light is dispersed in the detector Raman Spectroscopy is a biological medium and inorganic ready for Raman analysis. This can be solids, liquids, polymers or vapour. In Raman spectroscopy doesn't need the sample preparation. Very clear as a fingerprint chemical. Samples can be analysed by glass or polymer packaging. a region from 4000 cm^{-1} to 50 cm^{-1} can be covered with a single recording Raman spectra can be collected in very small volume.

Application of Raman spectroscopy

The many spectroscopy techniques used in analysis examination and identification purpose in many fields. The Raman spectroscopy is also used for the identification examination and analysis purpose. This Raman spectroscopy is not only use in physics, chemistry they can also use in Forensic Science, medical profession. In this article we give comprehensive review on many several applications of Raman spectroscopy in forensic science. The forensic science is the science which is useful for law and criminal justice system.



In forensic science they have many field/ divisions in it such as chemistry toxicology, Biology, DNA, serology, Ballistic, Question Document, Physics etc. The following review on application of Raman spectroscopy. In the study of the documents in question, it is generally not appropriate to destroy a writing sample in order to perform chemical analysis, as may be required for mass spectrometry. Without the necessity for sample alteration or compromising the value of documentary evidence. Raman spectroscopy enables for chemical analysis of contentious writing samples.

This Raman spectroscopy application is used for the pen and printing ink analysis; but this technique shows the drawbacks, some limitation like the fluorescence signals exhibited on the paper. But this spectroscopic method is applicable for examination of inks and ink materials. The determine source of origin of unknown printed document based on their classification into laser or other inkjet photocopier devices. They

or complied Raman spectroscopy with chemo metrics as well as principle of component analysis. By using or applying Raman Spectroscopy can be measure the security features of currency banks notes. The Raman micro spectrometry were used for the differentiate ink colour of banknotes by showing the Raman spectra of that sample. Also, can find or detect the sample was genuine or fake on the basis of colour combination. In the Biology examination, identification of the body fluids like saliva sweat, urine, semen.

The Raman Spectroscopy is used for the determine heterogeneous single sample of dried saliva as well as examine the qualitative variations in saliva. Using this saliva sample create unique spectroscopic signature for body fluids. The NIR RS was also used for the measure the spectra of dried blood sample of human for these using various mapping software and get the statistical analysis spectra of dry blood sample. Raman Spectroscopy beneficial for the give the difference between peripheral Blood and menstria 1 blood. The peripheral Blood and menstrual blood are same but spectra of Raman spectroscopy show the difference. The Raman spectroscopy is also b useful for serological evidence analysis. Blood tests have been impressively included in Raman's analysis, investigating whether it is possible to use Raman techniques to detect the presence of something in the blood and to try to age the blood sample. The study concluded that Raman spectroscopy has been able to detect the presence of blood, which is usually found in the bloodstream, especially haemoglobin. This was achieved even when the blood samples were thoroughly purified. In addition, studies have investigated the possibility of using Raman spectroscopy to detect abnormal blood age, based on differences in peak distribution between pure and dried blood samples. With the possibility of portable Raman technology, the ability to detect blood using this process can allow analysis rapid identification of controllable substances and the ability to analyse samples without sample preparation or destruction. In physical evidence can be analysed by Raman spectroscopy with various methods.

The Raman spectroscopy concerned with analysis of pigments or paints for differentiate and is examination of forgeries, in paint. The fibre cases analysis by Raman spectroscopy with the certain characteristics of Fibre dyes can be subsequently identified by an unambiguously by applying Raman spectroscopy. The Raman spectroscopy gives the more in detailed infor mation about molecular structure of fabric and without distiches sample. The analysis of soil they have many techniques to analyse The Morphologically Directed Raman spectroscopy gives the in detail morphological characteristic of soil. The Glass is also physical evidence. The Raman spectroscopy is a useful for the analysis of glass surface and a difference between artifacts. In chemistry the Raman spectroscopy is used to determine element structure, Nature of chemical bond as well as the studying the chemical combination. The Raman spectroscopy on-destructive technique.

This technique applicable for the detection of drug and uncontrolled substances from nails of drug abused person. The Raman spectroscopy is powerful technique in the come drug abuse examination. This Raman spectroscopy applicable for drug abuse due to the fluorescence and also using Surface Enhanced Raman scattering for drug abuse. Kevin Buckley and Pavel Matousek demonstrate the use of spatially affect Raman

spectroscopy for the detection of illicit Drug from the various materials like cloths, plastic bottles, these techniques use because the Raman spectroscopy is non-destructive technique.

The pharmaceutical analysis is included in chemistry. In the pharmaceutical industries checking and testing content of pharmaceutical substances like tablets, medically authorised drug as well as checking purity of substances. The Identification of Raw Material, Quantitative determination and analysis of pharmaceutical substances like drugs and tablets analysed by using Raman spectroscopy. The Kaho Kwok and Lynne S. Taylor he describes the care on to get information of characterising of genuine tablet. The detection and examine tablet were fake or not by applying Raman spectroscopy. In addition, Raman spectroscopy is used successfully in the analysis of Gunshot residue and explosives, soil samples, fibres, and aggressive drug analysis. Raman spectroscopy can provide information about the chemical structure and ownership of compounds.

The field of scientific research has benefited greatly from using this method because of its advantages over other scientific techniques. For example, Raman spectroscopy does not require careful sample adjustment that is required by other methods, such as infrared spectroscopy. Also, it can analyse a wide range of samples. Aqueous solutions can also be analysed in this way, which is the limit of many similar strategies. Raman spectroscopy also generally produces a cleaner spectrum than other similar techniques, and has greater variability. In addition, Raman spectroscopy is a non-invasive procedure, which is another benefit due to the need to analyse the material that can be considered as evidence in a court case. It always prefers to preserve the evidence in its original form. Since the data generated by forensic analysis is often used as evidence in court, it is very important that it is very accurate and reliable, for this reason, Raman scattering finds itself used in many areas of forensic science because of its intensity. Below, we discuss the many applications for Raman's dissolution in intelligence science. Advanced Raman Scattering (SERS) and the common Raman rely heavily on forensics labs to explore the vast amount of physical evidence collected in crime scenes. For example, it can be used to analyse dyes, paints, fibres, fabrics, and textiles even lipsticks and shoe polish smears.

Raman and SERS can compare samples with known samples, determine their identity, and provide important information about the smallest tracking evidence, such as the colour of the paint chip, trace analysis as well as toxic material the origin of the fabric and the way it is made. It can even be used to compare hair samples, helping to identify suspects. The remains of a firearm can be important evidence in criminal cases. While scanning electron microscopes with energy dispersive spectroscopy (SEM/EDS) is often used to analyse gun fragments. Raman scattering can help detect small traces in almost any type of area, without the need to change the evidence by any means Raman scattering and SERS. They have long been used to analyse illicit drugs. Although infrared spectroscopy has often been a common way to detect and detect illegal drugs, Raman spectroscopy has gained popularity due to their non-destructive nature.

As compare to forensic science the Raman Spectroscopy use in various areas such as the Geological, Mineralogy (Carbon Material), Life sciences, Art, Archaeology. In Geology the examination and identification of various types of stones, Gems and rocks. In mineralogy the Raman Spectroscopy is applicable for the detection of the defects in carbon materials, minerals can be identified in transparent matrices. In the life science the Raman spectroscopy plays important role because by using this spectroscopic method we can analyse the biomolecules, biocompatibility of materials and its features, also analyse the nucleic acids. The abovementioned theory is related to the specific application of Raman Spectroscopy in various fields' areas.

Conclusion

The Raman spectroscopy although there are some limitations, such as low sensitivity and fluorescence disorders, Raman spectroscopy has emerged as a non-invasive method, non- destructive technique. Also, the Raman spectroscopy technique is less time consuming. There are some automatic mapping techniques and many software like wire 2.0, GRAMS/Ai7.0, matlab7.4.0 toolbox used for make accurate results. The quantitative analysis and fit in a few settings with the new technological advances soon, Raman spectroscopy is expected to become a popular analytical tool for a growing number of applications.

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