1 Introduction

Infrared and Raman spectroscopy are two of the most widely used techniques in the physical and natural sciences today. In 1800 Sir William Herschel, while studying the heating effect produced by various portions of the solar spectrum, established that it contained some form of radiant energy which could not be seen (Herschel 1800). A few years later, in 1840, his son Sir John Herschel was able to demonstrate the existence of infrared absorption and transmission bands by noting variations in the rate of evaporation of alcohol from blackened paper upon which the solar spectrum was projected (Kruse et al. 1962). By utilizing detectors, the science of infrared moved steadily ahead and the idea that infrared radiation was quite similar to visible light began to be accepted. The utility of infrared spectroscopy as a tool for identification of molecules and functional groups was realized by chemists in the late 1920s. Modern infrared spectroscopy started in the 1940s and 1950s with tremendous improvements in instrumentation, which put the technique at the head of physical and chemical research (Kruse et al. 1962).

The Raman effect also allows the observation of vibrational spectra providing information which complements those obtained by infrared spectroscopy. This effect had been repeatedly predicted. Lommel (Lommel 1878) described certain anomalies of fluorescence, the color of which is dependent on the nature of the sample and the frequency of the exciting radiation. Smekal (Smekal 1923), Kramers and Heisenberg (Kramers and Heisenberg 1925), Schrödinger (Schrödinger 1926), and Dirac (Dirac 1927) predicted the Raman effect by applying quantum mechanics to molecules. Raman was looking for the optical analogue of the Compton effect, when his co-workers Krishnan and Venkateswaran observed "modified scattering" of sunlight, which Raman identified as the Kramers-Heisenberg effect. A short paper titled "A New Type of Secondary Radiation" by Raman and Krishnan was submitted to Nature on February 16th, 1928 (Raman and Krishnan 1928). As mentioned above, the basic theory of the Raman effect was developed before its discovery. However, at this time, numerical calculations of the intensity of Raman lines were impossible, because this requires information on all eigenstates of a scattering system. Placzek (Placzek 1934) introduced a "semi-

classical" approach in the form of his polarizability theory. This provided a basis for many other theoretical and experimental studies. The most important stimulus to the development of the Raman spectroscopy has been the laser, invented by Maiman in 1960 (Maiman 1960). During a short period the mercury arcs were replaced by really monochromatic and powerful light sources. At the same time, the photographic plates were replaced by photomultipliers, and scanning grating spectrometers replaced the prism spectrographs. Also, the introduction of double and triple monochromators, an elaborate sample technique (Kiefer 1977), and later the introduction of diode arrays and charge-coupled devices (CCDs) contributed considerably to the development of Raman spectroscopy. However, until about 1950, Raman spectroscopy was applied more often than infrared spectroscopy. After 1950, when automatically recording infrared spectrometers were introduced to the market, infrared spectroscopy became widely used in routine analysis.

Due to its non-destructive character, Raman spectroscopy represents, together with infrared absorption spectroscopy, one of the most useful tools for obtaining information about the structure and properties of molecules from their vibrational transitions, despite the fact that the direct assignment of the infrared or Raman bands of relatively complex species is rather complicated (Nakamoto 1997, Nafie 2001). Theoretical simulations can certainly assist in obtaining a deeper understanding of the vibrational spectra of complicated molecules. Recently, it was shown that density functional theory (DFT) methods are a powerful computational alternative to the conventional quantum chemical methods, since they are much less computationally demanding and take account of the effects of electron correlation (Parr and Yang 1989, Seminario and Politzer 1995).

However, the application of conventional Raman spectroscopy is limited by the weak intensity of the Raman scattered light and the appearance of fluorescence. One way to overcome these disadvantages is the use of surface-enhanced Raman spectroscopy (SERS) (Moskovits 1985, Vo-Dinh 1988, Campion and Kambhampati 1988). The existence of SERS was recognized thirty years ago and since then has been demonstrated to be a powerful analytical tool for the sensitive and selective detection of molecules adsorbed on nanostructured (i. e., roughened), coinage metal surfaces. SERS was first discovered by Fleischmann and coworkers in 1974, when they observed the strong Raman scattering of the pyridine molecules adsorbed on electrochemically roughened silver electrodes (Fleischmann et al. 1974). The scientific groups of Jeanmaire and Van Duyne (Jeanmaire and Van Duyne 1977) and Albrecht and Creighton (Albrecht and Creighton 1977) confirmed this enhancement phenomenon (up to 10^6) and attributed the effect to complex surface enhancement processes. As in the case of other scientific fields, the development of SERS with regard to detailed understanding, extensive application, and widespread acceptance was not continuous; to a certain extent, SERS has experienced its up and down periods.

The first progress period of SERS started immediately after its discovery and lasted until the mid 1980s. Throughout this time interval the research on SERS was largely populated by representatives of the condensed matter physics and chemical physics communities. The research activity was mainly focused on get-

ting a mechanistic understanding of the 10^6 fold intensity enhancement observed for normal Raman scattering. It was stated (Moskovits 1985, Moskovits 1982, Gao et al. 1990) that the enhancement of the Raman signal can be considered as the product of two main contributions: an electromagnetic enhancement mechanism and a chemical or charge-transfer enhancement mechanism. The contribution of the electromagnetic mechanism to the total enhancement is of the order of 10^4, while the chemical mechanism participation is in the range of 10^2. Surface selection rules were also presented in this time. In their simplest form, and assuming no specific symmetry selection rules, the most intense bands are predicted as those from vibrations, which induce a polarization of the adsorbate electron cloud perpendicular to the metal surface (Creighton 1988, Moskovits and Suh 1984). This information can be used qualitatively to find out details about the angle formed between the adsorbed molecule and the metal surface. One should also emphasize that surface-enhanced resonance Raman scattering (SERRS) with combined SERS and RRS enhancement factors in the 10^9–10^{10} range was already known (Jeanmaire and Van Duyne 1977, Sequaris and Koglin 1985).

In the next decade, SERS developed into quite a mature field. During this period, the attention of the representative researchers of the condensed matter physics was turned away from SERS to other subjects, and their place was taken by researchers interested mainly in applications of SERS to problems from electrochemistry, heterogeneous catalysis, polymer science, the biochemistry of surface immobilized proteins, and many others (Kneipp et al. 1999, Kneipp et al. 2002). However, this period was not so exciting as the earlier one and therefore can be considered as a down period.

In striking contrast, in the last ten years the interest in SERS was completely revived, mainly because of the remarkable discovery of a single molecule by SERS (Kneipp et al. 1997, Nie and Emory 1997) Moreover, the developments achieved in nanoscience and nanotechnology have determined a strong interest in SERS. Nowadays, it is difficult to find a paper on nanoscale optical properties which does not recommend SERS as the first example for applications. Today, there is an astonishing research interest concerning how to control, manipulate, and amplify light on the nanometer length scale using the properties of the collective electronic excitations in noble metal films or nanoparticles, known as surface plasmons. The interactions between adsorbed molecules and plasmonic nanostructures (Van Duyne 2004) may possibly have a considerable impact on many applications, such as localized surface plasmon resonance spectroscopy for chemical and biological sensing, sub-wavelength optical microscopy, and nanolithography, as well as SERS (Srituravanich et al. 2004, Anderson et al. 2005a, Anderson et al. 2005b, Haes and Van Duyne 2002, Riboh et al. 2003, Haes et al. 2004a, Haes et al. 2004b, Haes and Van Duyne 2004, Haes et al. 2005).

Although the theoretical understanding of the mechanism of surface enhancement is not definite and still evolving, the experimental data accumulated in the last years has demonstrated SERS to be a sufficiently sensitive spectroscopic method for surface science, analytical and environmental applications, biomedicine, biophysics, and biochemistry (Cotton et al. 1991, Baker and Moore 2005,

Haynes et al. 2005, Dieringer et al. 2006, Rosi and Mirkin 2005, Kneipp et al. 2002).

In this book, we present several of our recent results concerning SERS investigations on molecules of pharmaceutical interest (Iliescu et al. 1994, Iliescu et al. 1997, Iliescu et al. 2000, Iliescu et al. 2001, Iliescu et al. 2002a, Iliescu et al. 2002b, Iliescu et al. 2002c, Iliescu et al. 2003a, Iliescu et al. 2003b, Iliescu et al. 2003–2004, Iliescu et al. 2004a, Iliescu et al. 2004b, Iliescu et al. 2004c, Iliescu et al. 2006, Bolboaca et al. 2002, Bolboaca et al. 2003a, Bolboaca et al. 2004a, Baia et al. 2004, Baia and Baia 2005, Leopold et al. 2005) followed by a few studies related to the testing of the SERS efficiency of newly designed SERS substrates that would contribute to the improvement of the Raman enhancement of the adsorbed molecules, and to the enlargement of the investigation possibilities of the adsorption behavior of such molecules by SERS (Baia et al. 2006a, Toderas et al. 2004a, Toderas et al. 2004b, Baia et al. 2005a, Baia et al. 2006b, Toderas et al. 2006a, Toderas et al. 2006b, Astilean et al. 2006a, Toderas et al. 2007, Bolboaca et al. 2003c, Astilean et al. 2004a, Bolboaca et al. 2004b, Astilean et al. 2003, Astilean et al 2004b, Baia et al. 2005b, Astilean et al. 2005, Baia et al. 2006c, Baia et al. 2006d, Astilean et al. 2006b, Baia et al. 2006e).

Why SERS on pharmaceuticals? From pharmacological studies it is known that each drug is specific to a certain human organ on which it is adsorbed on some special centers. The adsorption of the molecules on a metal surface can be seen to mimic this adsorption process. In these investigations, the silver or gold surface can serve as an artificial biological interface (Dryhurst 1977). Moreover, for a complete understanding of the action of various drugs, such as the derivatives discussed in this book, it is very important to know if the structure of the adsorbed molecules is the same as that of the free species, and also to establish whether or not the molecule-substrate interaction may be dependent on the pH value of the environmental solution.

Besides this introduction section, the book consists of other eight chapters. Prior to presenting the experimental results, the fundamentals of infrared, Raman, and surface-enhanced Raman spectroscopy are highlighted. A few details about the experimental measurements are also specified. Since the assignment of the vibrational modes of most of the investigated species was performed with the help of theoretical simulations, a short presentation of the employed methods is also included in this chapter.

The SERS investigations illustrated in the next chapters are focused on different kind of drugs: tranquilizers and sedatives, anti-inflammatory drugs, vitamins, drugs with anti-bacterial properties, etc. Since there is an increased interest in designing highly effective and controllable SERS-active substrates, a few newly-developed substrates that could contribute to a deeper understanding and knowledge of the adsorption behavior of various types of molecules of pharmaceutical and medical interest are presented in the eighth chapter. The conclusions drawn from all these investigations are summarized in the last chapter.

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