The universality of the Raman Effect

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Abstract

In this article which is dedicated to the memory of Sir C. V. Raman and his scientific genius we would like to focus the attention of the scientific community on the universality of the Raman effect in all the ranges of the electromagnetic spectrum from far infrared to gamma rays. After explaining the basic Raman effect the article discusses nuclear Raman effect in the gamma ray region, x-ray Raman effect involving the electronic states and the well known Raman process in the optical as well as infrared regions in which the valence electronic and the molecular states participate.

1. Introduction

The birth centenary year of Sir Chandrasekhar Venkata Raman was celebrated in the year 1987–88. The Raman effect which has brought glory to India is now more than sixty years old. A large number of articles have been written and many lectures and meetings have been organized in recent years to commemorate this occasion. Scientific, cultural and historical aspects of Raman’s personality and his work have been discussed at length. We would like to bring out in this article an aspect of the Raman effect which has not, perhaps, been so well recognised by the scientific community, namely the universality of the Raman effect. Raman himself probably had only a vague idea of this universality although he was well aware of the importance of the effect and the breadth of the applications of the effect discovered by him as can be seen from the following quotation from his Nobel lecture delivered in Stockholm on the 10th December 1930:

“The universality of the phenomenon, the convenience of the experimental technique, and the simplicity of the spectra obtained enable the effect to be used as an experimental aid to the solution of a wide range of problems in physics and chemistry. Indeed it may be said that it is this fact which constitutes the principal significance of the effect. The frequency differences determined from the spectra, the width and character of the lines appearing in them, and the intensity and the state of polarization of the scattered radiations enable us to obtain an insight into the ultimate structure of the scattering substances.”

The existence of the Raman effect in all kinds of scatterers, solids, liquids and gases, organic and inorganic materials, amorphous and crystalline as well as polymeric substances, has been rather well known. However, what is not so well known is that Raman scattering occurs in all the different wavelengths of the electromagnetic spectrum from infrared to gamma rays. The universality of the Raman effect that we want to bring out in this article is the existence of the Raman effect in the different parts of the electromagnetic spectrum and the different kinds of information about the scatterers revealed by such studies.

2. The basic Raman process

A general scattering process could be thought of as absorption of the incident particle and emission of the scattered particle by the scatterer. In the act of scattering, the scatterer may change its quantum state and these changes would be revealed in the state of the scattered particle. Many types of scattering processes are possible depending upon the nature of incident and scattered particles, the
states of the scattering system involved and the kinds of interactions responsible for the act of scattering.

The basic Raman process involves inelastic scattering of electromagnetic radiation with discrete changes in the frequency of radiation, either gains or losses, characteristic of the scattering system. These discrete frequency changes correspond to the quantum jumps between the energy levels of the scatterer:

\[ \Delta \nu = \nu - \nu' = (E_i - E_F)/h, \]  

where \( \nu \) and \( \nu' \) are the incident and scattered frequencies, \( E_i \) and \( E_F \) are the initial and final states of the scatterer and \( h \) is the Planck's constant. This equation can be readily understood as a consequence of energy conservation when one applies Planck's hypothesis that photon of frequency \( \nu \) carries a quantum of energy \( h\nu \) so that equation (1) is equivalent to

\[ h\nu + E_i = h\nu' + E_F. \]  

Different types of Raman processes could be distinguished by the detailed mechanisms involved in the scattering process. Simplest of these is the normal Raman process in which the scattering system goes to an excited state \( (E_F > E_i) \) and the scattered light contains the so called Stokes line of frequency \( (\nu' < \nu) \) lower than the incident frequency. The corresponding process, when the scattering system is initially in the excited state \( (E_i > E_F) \) and loses energy to the radiation so that the scattered photon is of higher frequency \( (\nu' > \nu) \) than the incident photon, gives rise to an anti-Stokes line. Fig. 1 shows the spectrum obtained by Raman by using a mercury arc lamp in the region of 4358.3 Å for carbon-tetrachloride using the photographic technique. The picture clearly shows the elastically scattered \( (\nu = \nu') \) Rayleigh line and on its both sides the Stokes and anti-Stokes Raman lines. Incidentally, this nomenclature of Stokes line is reminiscent of the Stokes law in fluorescence phenomenon according to which the frequency of fluorescent light is always lower or at most equal to that of the exciting light.

In lowest order of perturbation, the scattering of light by a non-relativistic system can take place either in a single elementary process involving two photons (the seagull diagram) or in two elementary processes involving a single photon in each, as shown in Fig. 2. When two elementary processes of photon absorption and emission are involved, an intermediate state of the scattering system plays a role in the scattering mechanism besides its initial and final states. It should be emphasised here that, since this intermediate state exists only for a very short time, energy conservation is not relevant.
for the intermediate state. However, when the situation is such that energy is conserved for a particular intermediate state, photon absorption cross section increases tremendously and we get a resonance phenomenon accompanying the Raman scattering. The different scattering processes and fluorescence are illustrated schematically in Fig. 3.

Photons have spin one and they obey the Bose-Einstein statistics. Thus for each spontaneous process involving photons a corresponding stimulated process is possible. With the advent of tunable dye lasers it is now possible to get an intense monochromatic coherent light beam over a wide frequency range. Thus one can have stimulated Raman scattering when the incident photon beam also contains photons of energy equal to that of the Raman scattered radiation. The cross section of the Raman process could be increased by many orders of magnitude by using this mechanism of stimulated Raman scattering. However, only one Raman line and not the whole Raman spectrum can be stimulated at a time. With the use of very high intensity laser sources for the incident light, higher order electromagnetic processes involving the Raman transitions can also be studied. These processes involve, for example, simultaneous absorption and emission.

**Neutron Raman Scattering**

Just as inelastic light scattering is an important tool for studying various energy levels in solids, liquids, and gases, thermal neutrons from a reactor (energy ~10 meV–100 meV) can also be used as a powerful probe for investigating dynamics of condensed matter. In analogy to Stokes and anti-Stokes processes in light scattering, neutrons can exchange energy with the scattering system. The cross-sections are given by similar quantum mechanical expression. An important difference which may be noted here is that whereas wavelength of visible light is several thousand angstroms, that of thermal neutrons is less by about three orders of magnitude. In other words, light couples with long-wavelength fluctuations and neutrons interact with comparatively short wavelength excitations in the medium. The two techniques are thus complementary for probing the spectrum of elementary excitations in condensed systems. Neutrons also couple to the atomic magnetic moments and hence give information on spin dynamics also.

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Feynman diagrams

In 1949, in a series of very interesting papers, Richard Feynman introduced a novel way of analyzing interactions between charged particles and electromagnetic fields. His approach was to "deal directly with the solution of the basic differential equations involved rather than with these equations themselves". Using his ideas he was successful in resolving certain fundamental difficulties that had plagued quantum electrodynamics for a long time and eventually the work won him the 1965 Nobel prize in physics (shared with Schwinger and Tomonaga).

Feynman’s method primarily introduced a geometric way of looking at complicated algebraic terms and physically interpreting them in terms of simple intuitive processes in space-time. He developed a consistent way of organizing these processes by looking at their geometric representations, and by employing the topological properties of the ensuing figures he could deal with infinitely many of them in a relatively easy manner. In some sense this heralded a re-entry of a geometrical approach in the hitherto algebra-dominated field of theoretical physics. It is incidentally interesting to remark that although Newton developed the mathematical branch of calculus (which is algebraic in character) in his famous *Principia* he presented almost all his arguments in a very beautiful geometric form. By now Feynman diagrams have permeated the entire field of theoretical physics in the quantum realm—be it atomic, molecular, the solid state or the nuclear and particle physics.

A simple example of geometric manipulation can be seen in summing the series

\[ S = 1 + 1/2 + 1/4 + 1/8 + \ldots \text{ad inf.} \]

Representing 1 by a unit square the series could be written as

A more general series (viz. for Green’s function) that often appears in quantum mechanics is of the form

\[ G = G_0 + G_0VG_0 + G_0VG_0VG_0 + \ldots \text{ad inf.} \]

\[ G_0 \text{ and } V \text{ are in general integral operators; e.g. the second term, in coordinate representation, is given as} \]

\[ \int G_0(r_t; r''t'')V(r'')G_0(r'')dt'' \]

Let us represent this series pictorially by using a straight line joining points \( r_t, r''t'' \) as a picture of \( G_0(r_t, r''t'') \), a shaded small blob to denote the interaction \( V \) with the integral understood. The second term of the series then looks like

\[ \text{Put the different figures together as shown below} \]

A simple example of geometric manipulation can be seen in summing the series

\[ S = 1 + 1/2 + 1/4 + 1/8 + \ldots \text{ad inf.} \]

Further, the sum of the series is obtained without any mathematical manipulations.

Raman scattering thus basically involves transitions between discrete states. In general, the corresponding wave functions are localized in space and momentum is not a good quantum number for these states. The incident and the scattered photons have well defined momenta \( \hbar k \) and \( \hbar k' \), with \( k = 2\pi \nu/c, k' = 2\pi \nu'/c \), \( c \) being speed of light in vacuo. The scattering probability involves the modulus square of the \( (k - k') \)-th Fourier component of the interaction potential and momentum is not conserved in the act of scattering. In other words, the scattering probability is not sensitive to the amount of momentum \( (\hbar k - \hbar k') \) transferred by the photon in the act of scattering. Geometrically, this momentum transfer is predominantly determined by the scattering angle \( \theta \). When

\[ |\nu - \nu'| \ll \nu, \nu', \]

the momentum transfer is given by

\[ \hbar |k - k'| = 2\hbar k \sin(\theta/2). \]

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$G_0 \ (r, r')$ physically represents the propagation of a noninteracting particle from a point $r$ to a point $r'$ and this is indicated by putting an arrow on the line. The whole series for $G$ is then given in terms of Feynman diagram (by contracting the space axis for simplicity) as

\[
G = (1 - G_0 V)^{-1} G_0
\]

with a formal solution

where $I$ is the unit or identity operator.

Let us now consider electron-photon interaction and represent free electron propagation by a straight line and free photon propagator by a wiggly line. A diagram like

\[\text{Diagram 1}\]

represents emission of a photon at $a$ and its reabsorption at $b$. A couple of higher order similar process could be shown by

\[\text{Diagram 2}\]

A similar geometrical expansion for free energy or energy can also be made. We can use the arguments similar to those used in summing the series for $G$ to evaluate the influence of such terms on the electron and photon propagators. In most instances the Feynman graphs are drawn in momentum space rather than in position space to exploit the translational invariance properties of the system under investigations.

Further reading

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Equation (3) is satisfied in many common situations and the Raman scattering cross section is relatively independent of the scattering angle. This fact is often used in identifying the Raman lines from the competing scattering processes in complicated situations.

In the Compton scattering, the scatterer is a free electron, supposed to be initially at rest. The quantum mechanical states of the free electron form a continuum rather than a discrete set of states and thus one gets a distinction between the Raman and the Compton scattering mechanisms, the cross section for the latter being sensitive to the scattering angle, as the scatterer states are then also momentum eigenstates. When the electron momentum distribution in the scatterer is ignored, the Compton modified line is independent of the scatterer but dependent upon the scattering angle. In contrast, the Raman modified line is independent of the scattering angle but is dependent upon the specific scatterer.

3. Different excitations—different frequency ranges

Quantum states of physical systems appear over a
Figure 2. Schematics of a scattering process. The wiggly lines represent the photons and the straight lines represent the scatterer. The subscripts i and s denote the incident and the scattered photon states. I, F and n stand for the initial, final and the intermediate states of the scatterer.

very wide range of energy values from millionths of an electron volt to millions of electron volt. Typical physical systems and the corresponding ranges of energy levels are summarised in Table 1. We have nuclear levels with energies in the range of a few tens to hundreds of MeV; atomic electronic levels in keV range and valence electronic-molecular orbital levels in the eV arena. The molecular rotational and vibrational levels and phonon spectra in solids range from meV to a fraction of an electron volt. The nuclear and electronic spin-flip states have even lower energies. Since electromagnetic interactions play a part in all these physical systems, these levels can, in principle, couple to the incident electromagnetic radiation and participate in the Raman scattering process.

However, the scattering cross section for the Raman process involving a set of energy levels in a particular range is significant only when the incident radiation has energy $h\nu$ in the same range or a little higher. Experimentally, also, this is important since to demand a resolution $\Delta \nu = |\nu - \nu'|$ which is very small compared with $\nu$ may not be practically feasible, thus making experimental detection of the Raman scattered photons difficult when the states participating in the Raman process have energies very much lower than that of the incident photon. Therefore, for Raman processes involving excitations in different energy ranges we have to use the incident electromagnetic radiation in the corresponding frequency ranges. Historically molecular vibrational states in organic liquids were the first systems studied by this mechanism. Later on the rotational states of molecules, the vibrations of the atoms of the crystalline solids (lattice vibrations) were also
corresponding to different processes when radiation is incident on a scatterer: (a) absorption, (b) Rayleigh, (c) and (d) Raman Stokes and antiStokes, (e) resonance Raman, (f) fluorescence.

studied with great success. Establishing the Raman effect with participation of discrete electronic states of atoms (and molecules etc.) took a much longer time. The main reason for this was the considerable experimental difficulties involved in recording the weak scattered spectrum using incident x-rays. In the domain of nuclear states the Raman effect is more complicated due to other competing processes such as particle emission, Coulomb excitation and so on. Also the resolution in nuclear gamma ray scattering experiments is poor in comparison with the optical experiments. Thus the nuclear Raman scattering took even longer time to become an important field of study. However, with present day improved experimental techniques the nuclear Raman scattering has become a very sensitive tool for testing models which describe the decay modes of the giant dipole resonance.

It should be remarked here that quantum mechanically the Raman process is always accompanied by other competing processes. The relative ease with which the Raman process can be experimentally studied depends upon the strengths of these competing processes. Also the Raman process is essentially quantum mechanical, and as the wavelength of the incident electromagnetic radiation increases, the classical rather than the quantum mechanical effects tend to predominate, thus obscuring the Raman process in the long wavelength (low frequency) region.

In recent years there has been a considerable growth in high intensity radiation sources in different wavelength regions—masers, free electron lasers, optical high power lasers, tunable dye lasers, synchrotron radiation sources, rotating anode x-ray

![Figure 3. Energy level diagram showing transitions corresponding to different processes when radiation is incident on a scatterer: (a) absorption, (b) Rayleigh, (c) and (d) Raman Stokes and antiStokes, (e) resonance Raman, (f) fluorescence.](image)

![Figure 4. Energy level diagram showing transitions corresponding to (A) stimulated Raman process and (B) nonlinear or hyper Raman process. \( \omega_s = \omega_i - \Delta \) and \( \omega_s = 2\omega_i - \Delta \) are the frequencies of the incident and the scattered photons and \( \hbar \Delta \) is the energy difference between the states participating in the Raman process.](image)
Table 1: The Raman process in different physical systems

<table>
<thead>
<tr>
<th>Physical system</th>
<th>Participating states</th>
<th>Incident radiation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nuclei</td>
<td>Nuclear rotational and vibrational states, about 60 MeV from states participating in giant dipole resonance at an approximate energy loss of 20 MeV</td>
<td>γ-rays of about 60 MeV from nuclear reactions (No anti-Stokes line possible)</td>
</tr>
<tr>
<td>Atoms</td>
<td>discrete atomic electronic states, transition from discrete state to a continuum state corresponding to ionization in the keV energy range</td>
<td>x-rays of about 10–60 keV from rotating anode sources/synchrotron radiation (No anti-Stokes line)</td>
</tr>
<tr>
<td>Molecules</td>
<td>discrete rotational and vibrational levels of molecules in a few eV energy range</td>
<td>Ultraviolet and visible radiation preferably from laser sources</td>
</tr>
<tr>
<td>Solids</td>
<td>phonon and magnon states in the meV energy range</td>
<td>Free electron laser in the IR range</td>
</tr>
</tbody>
</table>

generators, intense γ-rays from nuclear reactors and so on. Also a large number of sensitive detection systems are now available in different frequency ranges. These developments have given a great impetus to the study of the Raman process in different wavelength regions to which we now turn.

### 4. Raman scattering in the optical region

Historically, the Raman effect was first observed in this wavelength range. Photon energies $h\nu$ in this region are about an electron-volt or so. The participating quantum states of the scattering system in this range are the rotational and vibrational levels of atoms and molecules. The early experiments had involved many organic liquids as scatterers. The existence of radiation in the scattered spectrum, which later on was recognised as Raman modified line, was noted in Raman’s laboratory as early as 1923. However, it was then thought of as some fluorescence due to impurities. When this radiation persisted inspite of many repeated purifications to remove the impurities, Raman suspected it to be more fundamental and careful experiments using a mercury arc and a spectrograph led to the discovery of the Raman effect in 1928.

On the theoretical side the possibility of frequency shifts in light scattering was discussed by Smekel in 1923 when he considered the interaction of light with an atomic system in the context of Bohr’s atomic model. Smekel’s analysis was elaborated by Kramers and Heisenberg in 1925 by applying the newly developed quantum mechanics and in 1927 Paul Dirac developed a fully quantum mechanical theory of interaction of radiation with matter. However, Raman was not aware of this theoretical work when he experimentally discovered the frequency shift of the scattered radiation and realized its basic significance. It was only after the pioneering work of Raman and his students that researchers all over the world took up this work and developed it further. Notable among these were Wood in USA, Menzies in England, Cabannes and Daure in France, Rasetti in Italy, Schaefer and Kohlrausch in Germany, Mizushima in Japan, McLennan in Canada, and Placzek and Gross and his collaborators in the Soviet Union. In the last sixty years considerable work has been done on the Raman effect in the optical range. Thousands of materials have been investigated and a very large number of research publications, general articles, review papers and books have appeared on the Raman effect. It has found applications in different kinds of materials: liquids, crystalline and amorphous solids as well as polymers and biological materials.
### Table 2: History of x-ray Raman scattering

<table>
<thead>
<tr>
<th>Authors(s)</th>
<th>Brief description of work</th>
</tr>
</thead>
<tbody>
<tr>
<td>Smekel (1923)</td>
<td>Theoretical prediction of x-ray Raman scattering</td>
</tr>
<tr>
<td>Raman (1928)</td>
<td>Independent theoretical prediction</td>
</tr>
<tr>
<td>Krishnan (1928)</td>
<td>First observation of x-ray Raman effect using MoK radiation scattered by graphite.</td>
</tr>
<tr>
<td>Sommerfeld (1936)</td>
<td>Theoretical analysis of discrete Raman lines in Compton scattered x-radiation</td>
</tr>
<tr>
<td>Dasgupta (1950, 1951, 1959, 1962, 1964)</td>
<td>Careful experiments leading to observations of x-ray Raman scattering of Mo and Cu\textsubscript{0.5}K rays by beryllium, boron and graphite.</td>
</tr>
<tr>
<td>Faessler and Muhle (1966)</td>
<td>Repeated Dasgupta's experiments and confirmed the phenomenon of x-ray Raman scattering</td>
</tr>
<tr>
<td>Cooper and Leak (1966)</td>
<td>Theory of x-ray Raman scattering</td>
</tr>
<tr>
<td>Mizuno and Ohmura (1967)</td>
<td></td>
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<tr>
<td>Pimpale and Mande (1984)</td>
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</table>

### 5. Raman scattering from lattice vibrations

A crystal could be thought of as a macroscopic molecule and the vibrational states of this 'molecule' could then be reflected in the Raman scattered spectrum. These states are analyzed by following the theory of lattice dynamics developed by Max Born who considered the crystal as a three-dimensional array of \( n \times n \times n \) Bravais unit cells, where \( n \) is a large integer, and applied to this problem his now famous periodic boundary conditions. The vibrational states then correspond to the quasi-continuum of phonon spectrum. Experimentally, such a continuum was not observed, rather a number of discrete Raman lines showed up in the inelastic scattered radiation. Historically, this observation triggered the Born-Raman controversy in the field of lattice dynamics. Raman (1943, 1961) considered only a 'supercell' consisting of \( 2 \times 2 \times 2 \) Bravais cells having eight times the volume of the unit cell. The vibrational states of this supercell could be easily analyzed taking into consideration the geometric symmetries of the atoms comprising it, and only a small number of discrete vibrational states result in qualitative conformity with experimental observations. It is now realised that the Born theory is essentially correct for getting the lattice dynamics, although the Raman formulation offers an insight based upon only the near-neighbour interaction. The experimental observations of one-phonon continuum in the Raman scattered spectrum could be attributed to weak intensity of the incident radiation so that only those peaks appear in the scattered spectrum which correspond to a rather large density of states. Van Hove (1953) has shown the existence of such large density of states regions in the phonon spectrum arising due to symmetry considerations. These are known as Van Hove singularities and it is these which are predominantly reflected in the Raman spectrum from crystals. The modern work on Raman scattering from lattice vibrations is done by using a laser beam as incident radiation. Some results obtained by using this technique are shown in Fig. 5.

### 6. Raman scattering in the x-ray region: atomic electronic states

The history of Raman scattering in the x-ray region involving electronic transitions between discrete atomic states is rather chequered. The early experiments in this field were somewhat inconclusive and it led Compton and Allison to remark in their famous book, "The existence of
Figure 5. Polarized Raman spectra of naphthalene crystal in the lattice vibrational region at 20 °C, with 4880 Å excitation. It is seen that different vibrational modes are reflected in different polarizability components $aa$, $ab$ and so on [from Long (1977)].

such radiation as a part of scattered x-rays, though frequently suspected, has never been established. It is probably too weak to be detected.” Thanks to the careful experiments of Das Gupta in the nineteen fifties and sixties and later the Japanese work, x-ray Raman scattering is now firmly established. Table 2 summarizes the major work on x-ray Raman scattering for a ready reference. Some experimental results of Das Gupta are shown in Fig. 6.

An important application of x-ray Raman scattering is to get the term values for low atomic number materials. These usually lie in the very soft x-ray region and conventional techniques in this wavelength range are very complicated and are not so accurate. Using the x-ray Raman effect one can use medium x-rays and obtain the atomic core level energies accurately. Such work is particularly relevant in studying the chemical effects on the inner atomic levels in solids.

7. Nuclear Raman scattering of $\gamma$-rays
The first indirect observation of nuclear Raman scattering was reported by Fuller and Hayward (1962) who used a bremsstrahlung incident beam and a NaI spectrometer. The term nuclear Raman scattering involves incoherent scattering of $\gamma$-ray photons from the electric dipole giant resonance photoabsorption region. The discrete
levels participating in the Raman process could be nuclear rotational or vibrational collective states. The incident photon is absorbed in the giant dipole resonance mode and the scattered photon is emitted so that the nucleus is left in one of the low lying collective excited states analogous to the Raman Stokes line in the optical region. The Fuller-Hayward experiment involved the rotational Raman scattering. High resolution detailed measurements on several nuclei were made by Bar-Noy et al. (1974) and Jackson and Thomas (1974). These workers used neutron capture gamma rays as a radiation source and Ge(Li) detectors and rotational Raman scattering from different rare earth nuclei was studied.

Qualitatively, nuclear Raman scattering involving the rotational states of the nuclei is caused by the coupling between the dipole oscillations of the giant dipole resonance and the rotational motion of the nucleus. A simple rotator model is adequate to explain the nuclear rotational Raman scattering. Nuclear vibrational Raman scattering arises due to coupling between the giant dipole oscillations and low lying quadrupole surface vibrations of the nuclei, which can be described by the dynamical collective model. The experiments involving nuclear vibrational Raman process were done by Arehenovel and Maissen (1970) and Hayward et al. (1973, 1974). However, the agreement between theory and experiment is mostly qualitative in these studies. More successful experiments in this field were carried out by Argonne-Illinois group (Bowles et al. 1981) using a tagged photon beam for five spherical nuclei between $^{52}\text{Cr}$ and $^{46}\text{Mo}$. Some typical experimental results are shown in Fig. 7. Similar experiments for spherically deformed nuclei $^{166}\text{Er}$ and $^{238}\text{U}$ were carried out by Nathan and Moreh (1980).

8. Concluding remarks

In this article we have shown how the Raman effect, originally discovered in the optical region of the electromagnetic spectrum, occurs in all the spectral ranges from infrared to gamma rays (see Table 1). In these various frequency ranges the radiation couples appropriately to the qualitatively different energy levels of the scattering system, such as the nuclear levels, the atomic levels, the molecular orbitals and the continuum of states in the valence bands of the solids. We have not mentioned here the possible Raman interactions with microwaves and radio waves. To our knowledge such studies have not been reported so far, but it is quite likely that such investigations may provide information about dilute plasmas in the upper atmosphere having natural frequencies in the same region. It is thus seen that the...
Raman effect is a basic process of nature involving energy transfer between radiation and matter and it provides a fertile ground for pioneering basic and applied research. Finally, we would like to express our confidence that young and old researchers in our country would continue to work on the Raman effect for many years to come.

References