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OPTICAL SPECTROSCOPY AND DEVELOPMENT OF QUANTUM ELECTRONICS

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1. INTRODUCTION

WITHIN the limited time at our disposal, it will be impossible to do justice in talking about even a small part of the research work done by Professor C. V. Raman, who was the greatest experimental physicist India has produced so far. In spite of this constraint we will make an effort to discuss the impact of some of the pioneering work in optics done by him and his students on the modern developments in Quantum Electronics.

Raman worked in many fields in physics including acoustics, magnetism and X-rays; but the study of optical properties of various materials fascinated him most. In fact, he was so much enchanted by "light" and "colour" from the very beginning (Raman, 1915) that he spent the later part of his research

career in a massive and detailed experimental investigation of diverse aspects of the physiology of vision (Raman, 1968). As a young man when Raman started doing physics, optical spectroscopy was one of the most powerful fields of investigation. It was rapidly producing new and unexpected results, and thereby introducing fundamental changes in the understanding of physics. It was, therefore, no accident that Raman decided to enrich this exciting field with all his vigour. It is perhaps significant to add that he brought with him to optics his earlier experience of working on the propagation of sound waves in matter. His original contributions to spectroscopy between the year 1917, when he became the Palit Professor of Physics in the Calcutta University, and the year 1948, when he retired from the Indian Institute of Science in Bangalore, were phenomenal by any standard.

In the fifties, spectroscopy and electronics were two highly developed disciplines. However, apart from industrial applications in chemical analysis, the fruits of the optical spectroscopic studies had remained almost entirely in the realm of basic physics. There was no direct electronic technology growing out of these investigations. The invention of lasers (Light Amplification by Stimulated Emission of Radiation) in the early sixties suddenly changed this picture. It gave rise to the new field, quantum electronics, which lies at the interface of spectroscopy and electronics. The present-day activities in quantum electronics should be considered to be at the developmental stage of the electronic technology growing out of the research on resonance spectroscopy involving discrete quantum energy levels of individual atoms or molecules in a material. In this sense it differs in an essential way from the electronics of vacuum tubes which grew out of the basic research in the last century on the discharge of electricity through rarefied gases, and from the electronics of transistors which grew out of the work on the electronic motion in a periodic crystal. Contrary to the case of lasers, the electronic energy excitations due to external electromagnetic fields in a vacuum tube or in a transistor are nonresonant and essentially continuous. Also, since the quantum of energy of the optical photon is much larger than the thermal energy kT at ordinary temperatures, the quantum approach becomes necessary to understand the behaviour of Opto-electronic devices.

In everyday electronics we are perhaps familiar with the role of variety of practical devices such as coherent amplifiers, oscillators, modulators, nonreciprocal circuits, nonlinear elements and detectors. A spectroscopist may now

claim that one can fabricate all these familiar circuit elements working at a much higher frequency by making use of optical resonances in a material medium.

Before we discuss any application of a particular study made by Raman and his collaborators to quantum electronics, let us first classify some of the relevant work done by the group.

2. OPTICAL SPECTROSCOPY FROM RAMAN'S SCHOOL

Raman and other members of his group worked in almost every branch of spectroscopy to study molecular and solid state physics. In fact, while most of the other physicists shifted to nuclear and high-energy physics in the thirties and the forties, Raman's group remained one of the strongest groups which continued doing fundamental work in optical spectroscopy and crystal physics. The type of studies undertaken by this group may roughly be divided, for our purpose, under the following headings:

- (a) Luminescence and Absorption.
- (b) Light Scattering.
- (c) Electro-optic, Elasto-optic and Magneto-optic effects.

In what follows, we will discuss the relevance of each of these investigations separately.

3. LUMINESCENCE AND ABSORPTION

Luminescence denotes the phenomenon of real absorption of energy in matter, and its re-emission as optical radiation. The initial excitation may be by light, X-rays, electrons, subatomic particles, electrical discharges or chemical reactions. The frequency of the radiated wave depends solely on the nature of the quantum states of the material. The intensity of the exciting agent, however, governs the intensity of the radiated wave, which also depends on the nature

of the excitation mechanism. Almost all solid materials which are luminescent, with a high efficiency for conversion of various forms of energy into optical radiation, consist of a host crystal containing a very small quantity of impurity atoms, called the activator. For example, bright red emission is obtained from ruby, which consists of an aluminium oxide host crystal containing a very small percentage of chromium oxide activator in which the luminescence takes place.

In the process of luminescence one had found a method to convert one form of energy or energy from a wave at one frequency to the energy at a different frequency. However, most of the optical emission under normal conditions is spontaneous and incoherent. There is only a very small number of atoms in the upper level which are stimulated by a spontaneously emitted photon to emit photons which are synchronized to the inducing photon. Under normal thermal equilibrium conditions, this is of course completely offset by very large absorption since there are many more atoms in the lower state than the upper state. From the work of Einstein and that of Bose, this was well known to every spectroscopist including Raman. Obviously weak and incoherent luminescence was of not much use to an electronic engineer. It was, therefore, clear that the ratio of the stimulated to spontaneous emissions had to be greatly increased, if stimulated emission was to be used in a practical device. In other words, the signal has to be stronger than the noise. A population inversion between the upper and lower levels thus becomes one of the fundamental requirements in making an amplifier. The process of achieving this is called "pumping" in quantum electronics. This, however, is not the only requirement. The other require-

ment for efficient amplification is that the stimulated emissions that go to make up the laser beam must have a high gain in only one direction and in a narrow frequency region within the spontaneous linewidth. The resonator which accomplishes this in the optical case consists simply of mirrors kept at each end of the material.

An efficient pumping to obtain the population inversion by an amount greater than various types of loss in the material may be done by an appropriate method of exciting luminescence. To most of the spectroscopists in the thirties and the forties, the chances of obtaining, in time shorter than 10^{-6} sec, a very large non-equilibrium situation that is required at optical frequencies seemed to be very remote. But as it turned out later, it was not such a difficult task after all (Kastler, 1954). If one is considering photo-luminescence, one has simply to choose a material which has a broad absorption band but narrow emission lines. In this way a large number of atoms can be transferred to an upper level. Ruby is a good example of a solid that has broad absorption bands centered around 5500 Å and two very narrow emission lines R_1 and R_2 at approximately 6943 Å and 6929 Å, respectively. Using Xenon flash tube for pumping, the ruby laser at 6943 Å was in fact the first laser device made by Maiman (1960) after the first full theoretical treatment of laser action was published by Schawlow and Townes (1958) and by Prokhorov (1958).

Luminescence of various crystals including ruby, sapphire, emerald, uranyl salts and diamond was actively studied by Raman and his students. We specially mention their extremely important and detailed work on ruby. The luminescence of ruby was investigated by C. S. Venkateswaran in 1935, B. V.

Thosar in 1938, A. Mani in 1942 and R. S. Krishnan in 1947. Thosar's interpretations of the results established important and useful details of the microscopic processes involved in the luminescence. Many quantitative data obtained by A. Mani, including her observation that the line R_1 was stronger than R_2 , can be shown to be very close to more exact values.

Like the transition metal ion Cr^{+++} in ruby, activator rare-earth ions have also narrow emission lines. But many of the rare-earths have narrow absorption bands as well, so that photo-excitation becomes inefficient. For these, other methods of excitation have to be used. However, Neodymium, Europium, Ytterbium and some other rare-earth ions have broad absorption bands and narrow emission lines—just what is needed for efficient luminescence and laser action. In these rare-earths, one of the 4f electrons moves into the unfilled and unscreened 5d subshell when the atom absorbs energy. The absorption band is quite broad as a result of this transition to an unscreened energy level. When the excited energy level changes as a result of the rearrangement of the atoms, the electron moves back into the 4f subshell. It is the return of this well-shielded 4f electron to the ground state that produces the very narrow emission line (Thosar, 1944). Many different types of lasers using several of these rare-earths in glass, Yttrium aluminum garnet (YAG) or calcium fluoride host have been fabricated.

Since in a gas there are no broad absorption bands suitable for optical pumping, usually electrical discharge is used for excitation. He-Ne (A. Javan *et al.*, 1961) and CO_2 -N₂ (Patel, 1964) lasers are examples of gas lasers using this principle for pumping. In semiconductor

p-n junction lasers, e.g., in GaAs lasers, high-current injection with a forward bias voltage nearly equal to the direct gap voltage E_g/e is the usual method for pumping (Dumke, 1962). These lasers are becoming more and more popular for technological applications because of already highly developed semiconductor technology. The high energy density that is released in exothermic chemical reactions is used in producing chemical lasers, whereas synchronized luminescence is obtained in many dye solutions (Sorokin *et al.*, 1966) by pumping either with another laser or with long flashes from high intensity lamps. Dye-lasers are particularly useful because it may be tuned easily by changing the dye concentration.

4. LIGHT SCATTERING

Light scattering has been the most fruitful field of research pursued by Raman's group from the very beginning. An essay by Raman entitled "Molecular Diffraction of Light" was published by the Calcutta University in February 1922. The concluding chapter of this essay on "Molecular Diffraction and the Quantum Theory of Light" envisaged the possibility of inelastic scattering of light due to an exchange of energy between the incident light quanta and the vibrations of the structural unit of the material. The history of the discovery of the Raman effect in 1928 in both liquids and solids is well known to everyone (Ramanathan, 1970), and we need not repeat it here. We will, however, like to stress the point that Raman and his group had to wait for several years before announcing the first observation of the inelastic process. They wanted to make sure that what they were observing was not just the photo-luminescence, discussed earlier. Whereas the photo-luminescence is a real

two-step process in which a photon is absorbed and then emitted in a time-scale of the order of 10^{-8} sec or more, the Raman effect is the true inelastic atomic process with a time scale of the order of 10^{-10} sec. In the present-day usage, indeed any inelastic light scattering process is called "Raman scattering."

To show how modern were the ideas of Raman's research group in the twenties, let us quote a few sentences from a paper by C. V. Raman and K. S. Krishnan (1928). Writing on "A Theory of Light Scattering in Liquids" they remark: "If we follow our individual molecule in a liquid over a sufficiently long time, it can take up all possible positions and orientations, and thus the conditions approximate to those of a gas. If, however, we consider molecules in relation to its immediate neighbours the analogy between the solid and liquid states is closer. We are not justified in treating the space distribution as random". This approach lies at the heart of the modern theory of the liquid state.

With the advent of lasers and better photon-detection techniques, Raman Spectroscopy has become one of the most useful fields of research to investigate any type of long wavelength excitations of a medium (Jha, 1971). This is providing fundamental inputs for the theory of liquid and solid states, and for the theory of phase transitions. We will not discuss this aspect either. What we will like to consider here is the fact that Raman scattering is a nonlinear process (Bloembergen, 1967), and one can exploit this property to convert energy from one frequency to another (Raman laser).

The transition probability for Raman scattering from any excitation, whether electronic or ionic, is proportional to $n_i(n_i + 1)$, where n_i and n_s are the number of photons in the incident and

scattered modes, respectively. The number 1 in $n_s + 1$ gives the spontaneous scattering, whereas n_s in $n_s + 1$ refers to the stimulated scattering. In order to predict the temperature dependence of the scattered intensity, Raman (1928) considered both these processes to derive the correct formula. If $\Delta\omega$ is the linewidth of the spontaneous line, assumed to be Lorentzian, there are $(V\omega^2\Delta\omega/2\pi^3c^3)$ modes of a given polarization in volume V for spontaneous scattering in a unit solid angle. This immediately gives the well-known relation between the spontaneous cross-section and the stimulated cross-section for a single mode:

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{stim.}} = n_s \left(\frac{d\sigma}{d\Omega}\right)_{\text{spont.}} \left(\frac{8\pi^3 c^3}{V\omega_s^2 \Delta\omega_s}\right).$$

At a finite temperature τ , the rate of increase of the number of scattered photon is, therefore, given by

$$\frac{dn_s}{dt} + \frac{n_s}{\tau_s} = G n_i n_s,$$

where τ_s is the life-time of the scattered photon in the medium and where the gain coefficient G , per unit time, per incident photon, is given by

$$G = c \left(\frac{1}{V} \frac{d\sigma}{d\Omega}\right)_{\text{spont.}} \left(\frac{1}{V} \frac{8\pi^3 c^3}{\omega_s^2 \Delta\omega_s}\right) \times \left[1 - \exp\left(-\frac{\hbar\omega}{kT}\right)\right],$$

$$\omega_i - \omega_s = \omega.$$

The threshold for stimulated scattering is obtained by equating the total gain coefficient $G n_i$ to the loss coefficient $1/\tau_s$. The threshold powers (per cm^2) for the incident beam, therefore, becomes

$$(P_{\text{inc.}})_{\text{thresh.}} = \frac{n_i \hbar \omega_i}{V} = \frac{c \hbar \omega_i}{\tau_s V G}.$$

For low threshold and high gain, the spontaneous cross-section has to be large, and the linewidth small. It has to be noted that no population inversion is involved in stimulated Raman scattering. However, in every case, the incident threshold power comes out to be so high

that one has to use another laser as the incident beam.

The Raman effect now provides coherent beams in parts of the spectrum where they cannot be produced directly from a laser. Using a large number of liquids like Benzene, Nitrobenzene, Carbon tetrachloride, etc., and solids like diamond, calcite, naphthalene, etc., Raman lasers have been produced in a wide range of frequencies. It is well known that spontaneous data for most of these materials (Sirkar and Chakravorthy, 1935) were obtained by Raman's group. We specially mention the detailed work of P. G. Nayar (1942) on the Raman effect in diamond which confirmed the earlier results of Bhagavantam (1930), and the investigations of R. Ananthakrishnan, C. S. Venkateswaran, T. M. K. Nedungadi and R. S. Krishnan.

Stimulated scattering from highly dispersive polariton modes, which are coupled optical phonon-photon modes in polar crystals like LiNbO_3 (Yarborough *et al.*, 1969), and from acoustic phonon modes in solids like quartz, have been used to obtain tunable Raman lasers. Using electronic spin-flip Raman scattering in InSb tunable Raman lasers have been obtained in the infra-red (Patel and Shaw, 1970).

That the Raman effect could be used to make oscillators and amplifiers at optical frequencies is not new. As early as 1929, R. V. L. Hartley compared the generation of Stokes frequency to the three-wave parametric oscillator; coupling in this case being between the incident light wave, the Stokes wave and the molecular vibration *via* the Raman susceptibility. Electro-mechanical parametric oscillators working at very low frequencies were well known to most of the classical physicists including Rayleigh. The actual realization of the Raman

amplifier was not possible only because of the high incident power threshold condition.

5. ELECTRO-OPTIC, ACOUSTO-OPTIC AND MAGNETO-OPTIC EFFECTS

In the preceding sections we were discussing the development of oscillators and amplifiers at optical frequencies. Next we must turn to the method of controlling a laser beam so that it can carry information. Of the several different ways of impressing information on a laser beam, the most important one is the amplitude modulation technique. Frequency modulation, phase modulation or phase-code modulation may also be used.

In many liquids and crystals there is a large variation of the index of refraction when an electric field is applied. This nonlinearity may be expressed by writing the change in the refractive index μ as

$$\Delta \left(\frac{1}{\mu^2} \right)_i = r_{ij} E_j + h_{ijk} E_j E_k$$

where r is the linear electro-optic coefficient and h is the coefficient associated with the quadratic effect ($i = 1, 2, \dots, 6$, Voigt notation; $j, k = 1, 2, 3$). In solids the linear variation is known as Pockels effect while the variation arising from the quadratic term is called the Kerr effect. It has to be noted that Pockels effect can exist only for crystals which lack center of inversion. A crystal that undergoes such an index of refraction change in a particular direction is called birefringent, and can be used in what is called a retardation type of modulator. The incident beam in this case is split in two and each travels at a different velocity in the crystal, which results in the phase difference between the two parts of the output by an amount that depends upon the applied modulating voltage and

the length of the electro-optic crystal. The mixed response of an analyser is thus modulated by the applied voltage.

Natural birefringence was actively studied by Raman himself in the twenties. Potassium dihydrogen phosphate (KDP) and ammonium dihydrogen phosphate (ADP) are two crystals most commonly used as electro-optic modulators these days. It should be emphasized here that the linear electro-optic modulation of the dielectric function is one of the mechanisms contributing to Raman scattering from longitudinal optical phonons in polar crystals. The early Raman work on KDP was done by C. S. Venkateswaran (1936) whereas the work on ADP was done by Narayanan (1948).

Ultrasonic waves propagating through a material can scatter the radiation passing through, and thus the modulation can occur under proper conditions. The intensity of the scattered wave is governed by the acousto-optic (photo-elastic) coefficient. A comprehensive theory of such a coherence process was developed by Raman and Nath (1936) in the early thirties. In effect a diffraction grating is set up in the material by the moving ultrasonic wave, which gives out the modulated output. For high-frequency modulation acousto-optic modulation should prove to be highly useful.

In order to show that almost any familiar circuit element can be fabricated at optical frequencies, let us consider the example of a non-reciprocal element like an "isolator". To prevent feedback between different amplifier sections one needs a device called an isolator. The purpose of an optical isolator is to allow a light beam to be transmitted in only one direction through it. In the optical case this nonreciprocal element works by using the Faraday effect. The Faraday effect is the rotation of the plane of

polarization by a magnetic field parallel to the light path, and is described by the equation.

$$\theta = VLB$$

where θ is the angle of rotation, L is the path length in the material, B is the magnetic field in the direction of the light path, and V is the Verdet constant. The operation of this device is surprisingly simple. The device consists of polarizer and analyser prisms between which the Faraday rotator (crystal) is kept. The length of the crystal is fixed in such a way that the angle of rotation of the polarization of the light wave passing through it is 45° . The analyser is set at 45° to the polarizer so that the beam coming from the direction of the polarizer is allowed to pass. Any reflected beam from the direction of the analyser, however, suffers a 45° rotation in the wrong direction, and thus escapes the polarizer.

In Raman's group magneto-optical effects were studied mainly by Raman, Bhagavantam and Ramaseshan (1948).

We may go on and on to describe many other types of recent optical devices. Instead, we feel it would be enough to assert here that the physical principles on which all these devices work were indeed the essence of extensive investigations in optics by Raman and his group.

6. CONCLUSIONS

Raman and his group, while working in the fields of luminescence, light scattering and nonlinear optics, were entirely motivated by the intrinsic physical importance of these subjects. They did not have in their minds any technical applications of their work. We trust we have conveyed in this talk the profound impact his work now proves to have on the new field of engineering, called quantum electronics. Time and again

this seems to be the case in the history of physics. Any basic work of physical importance ultimately finds practical applications.

We conclude by emphasizing that optical spectroscopy has finally grown to become a part of the new rapidly expanding technology, and we hope our tradition in optics established by Professor C. V. Raman would play a leading role in developing opto-electronics in India.

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COLOURS OF THIN BLACK NICKEL DEPOSITS*

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1. INTRODUCTION

BRIGHT lustrous metallic coatings of immense decorative appeal generally result from electroplating. But there are instances where coloured deposits are formed by deposition. Black nickel—obtained by the electrolysis of a solution containing nickel sulphate, zinc sulphate and ammonium thiocyanate—is one such example. Poor (1943) noticed that these deposits exhibited vivid colours in thin layers and were black in thick layers. He attributed the colours of black nickel to be due to interference in thin films. This hypothesis appears, at the outset, to be incongruent with some of the salient

features of black nickel that emerged from our studies. Prof. C. V. Raman's work on the colours of many materials considerably influenced us and paved the way for understanding the phenomenon of colours of black nickel. I shall endeavour now to discuss this in some detail.

2. EXPERIMENTAL OBSERVATIONS ON BLACK NICKEL

The deposits are definitely metallic as they evolved hydrogen on being reacted with acids. They show appreciable electronic conductivity. The colour changes sequentially, i.e., in the order yellow, red, blue, green, yellow, etc., with thickness which is reminiscent of interference phenomenon.

* Talk delivered at the 37th Annual Meeting of the Indian Academy of Sciences.