# Celebrating the 90<sup>th</sup> Anniversary of the Raman Effect

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#### Abstract

In 1928, at the Indian Association for the Cultivation of Science, Kolkata (IACS), C V Raman and his associates showed that when monochromatic light is scattered by transparent media, the scattered light contains not only the original colour, but also other colours. This effect was named as Raman effect. It is well-known that K S Krishnan played a significant role in the discovery. The present article suggests that Raman's student, S Venkateswaran's contribution to the discovery had also been ignored.

In French, German, Russian and Indian scientific literature, the Raman effect also appears under other names. The causes for it are explored. In the end a short summary over the sharing of the Nobel Prize among different discoverers is given.

Key words: A Smekal, C V Raman, G S Landsberg, J Cabannes, K S Krishnan, L I Mandelstam, Nobel Prize, Raman effect, S Venkateswaran.

#### **1.** INTRODUCTION

Ninety years ago, the foundation of the Raman spectroscopy was laid by C V Raman in Calcutta. In 1928, shortly after Raman published his results, physicists from other countries, in particular Russia claimed to have discovered the same effect. In the following years, Russian historians felt that their countrymen were ignored by the Physics Nobel Committee. In contrast, the claim of the Austrians and French men of science was not so loud. Raman's major coordinator K S Krishnan (Fig. 1 and 2) so long he lived, never claimed any credit. After his death, in some circles it was felt that he did not get the honour he deserved. For instance, A R Verma, who was the Director of the National Physical Laboratory, New Delhi, wrote that Krishnan, "strongly felt that he had been treated unjustly and that he was denied the credit by not sharing the Nobel prize" (Singh, 2004 pp. 74-75).

Raman being India's only Nobel Laureate in 'natural sciences', remains a legendary figure.

Not surprisingly, a number of articles and books had been written on him, which deal various aspects of his life (Krishnamurti, 1938; Keswani, 1980; Pisharoty, 1982; Sen, 1988; Ramaseshan and Rao, 1988; Venkataraman, 1995; Venkataraman, 1994; Jayaraman, 1989; Parmeswaran, 2011; Singh, 2004; Singh, 2005; Biswas and Raman, 2013; Banerjee, 2014). To the best of my knowledge, the above aspects has not been discussed in detail. The present article intends to fulfil the gap.

# 2. THE RAMAN-KRISHNAN EFFECT OR THE RAMAN-VENKATESWARAN-KRISHNAN EFFECT

The remarkable observation made by S Venkateswaran whose observation was the first step, which motivated Raman to recheck the previous results. He asked Krishnan to reconfirm experimental work (Fig. 1 & 2). However Venkateswaran did not get any credit. His name is almost unknown in Indian circles. He was a part

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time worker at the Indian Association for the Cultivation of Science, as he worked as a Chemical Assistant, at the Government Test House, Alipore. In 1924, he studied: 'scattering of light by solutions of gases in dust-free water'<sup>1</sup>. A year later in the Annual Reports of the IACS his research work was noted under 'light-scattering by electrolytes and chromatic emulsions'<sup>2</sup>. After Raman 'abolished' the Chemistry Department at the IACS; the only Research worker doing research in chemistry was Venkateswaran. In 1928, out of 32 scholars who worked under Raman, he was one of the 17 persons, who published articles<sup>3</sup>.



**Fig. 1.** K S Krishnan and S Venkateswaran (Credit: Indian Academy of Sciences, Bangalore)

Here it will be worth to quote from Raman's lecture on the discovery:

The chemical importance of the subject led to Mr. S Venkateswaran attempting to make a fuller study of it in the summer of 1925, but without any special success. The research was discontinued at the time but was resumed by him later in the current year (January 1928). **The remarkable observation** (emphasis added) was made that the visible radiation which is excited in pure dry glycerine by ultraviolet radiation (sunlight filtered through



**Fig. 2.** C V Raman and some of his scholars at the IACS. Sitting (left to right): A S Ganesan, L A Ramdas, K S Krishnan, C V Raman, K R Ramanathan, S Venkateswaran, and S S M Rao. Standing (left to right): C Ramaswamy, S Bhagavantam, S Paramasivan, S Rao, N S Nagendranath, A Ananthakrishnan and C S Venkateswaran. (Courtesy: IACS.)

Corning glass G. 586) is **strongly polarised** (emphasis in original) (Raman, 1927-1928, pp 387-398).

Unfortunately for Venkateswaran who could not devote whole time for researches; Raman asked K S Krishnan to reconfirm Venkateswaran's observations. Krishnan not only reconfirmed, but also observed the effect in other liquids and vapours at the suggestion of his Professor.

On February 5, 1928, K S Krishnan noted in his diary:

Recently Professor has been studying with Mr. Venkateswaran the fluorescence exhibited by many of the aromatic liquids in the near ultraviolet region present in sunlight and the fluorescence of some of the liquids was found to be polarised. However in view of the fact that fluorescence of anthracene vapour does not show any polarisation Professor asked me to verify again his observations on the polarisation in some of the liquids (emphasis added)<sup>4</sup>.

<sup>&</sup>lt;sup>1</sup> Annual Report-Indian Association for Cultivation of Science, 1924, pp 1-2.

<sup>&</sup>lt;sup>2</sup> Annual Report –Indian Association for Cultivation of Science, 1925, pp 124-126.

<sup>&</sup>lt;sup>3</sup> Annual Report –Indian Association for Cultivation of Science, 1928, pp 405-435.

<sup>&</sup>lt;sup>4</sup> The citations in this article are from "Personal Information File of K S Krishnan FRS, the Royal Society archive." Private Communication, July 6, 2017. Laura Outterside, Archivist Royal Society London. Prof. D.C.V. Mallik reproduced some pages from the original diary in "Notes and Records", see, Mallik, D.C.V. The Raman effect and Krishnan's diary, *Notes Rec. R. Soc. Lond.* 54(2000):67–83. Raman's student A. Jayaraman published extract of Krishnan's diary. See, Jayaraman, A. C.V. Raman – A memoir, Affiliated East-West Private Ltd., New Delhi, 1992, pp. 24-27.

In the following day, Krishnan studied the scattering of light in various liquids and vapours; and found that the scattered fluorescent light with weak intensity was polarized.

According to Krishnan's diary, on February 9, 1928, Professor Raman:

Told Mr. Venkateswaran about the discovery and was discussing the problem with us, in the course of which he said that the phenomenon should be called **the Raman-Krishnan-effect** (emphasis added).



**Fig. 3.** Title of Raman's article on the Krishnan effect. (Credit: The Mysore Times).

The author personally feels that instead of **Raman-Krishnan effect**, the new discovery deserved to be called the **Raman-Venkateswaran-Krishnan effect**. Krishnan only extended Venkateswaran's work, which was initiated under Raman's guidance.

It will be of interest to note that 'The Krishnan effect' appears in the literature (Fig. 3), but it is associated with the name of R S Krishnan, who was doing D.Sc. at the University of Madras.

The fact is that this caption never appeared in text books, even though it was recognized by Raman himself.

According to the Krishnan effect: "if the incident beam is polarised with the electric vibration horizontal, the light scattered transversely would exhibit a partial polarisation in which the horizontal vibration is more intense than the vertical" (Raman, 1941 pp 228-234)<sup>5</sup>. The term seems to be coined by H Müller in 1938, who stated:

In a series of recent investigations R. S. Krishnan (...) demonstrated the existence of a new effect which will be called the Krishnan effect. It relates to the state of polarization of the light scattered by certain liquid or solid media in directions normal to the incident beam (Meuller, 1938 pp 425-449).

# 3. THE RAMAN EFFECT

Raman himself did not see his first paper (with K S Krishnan) to '*Nature*' as the discovery article as his Russian opponents (detail below). In 1928 he was convinced that he made the discovery on 28<sup>th</sup> February, 1928 (Fig. 4).



**Fig. 4.** Announcement of the discovery by local press. The newspaper-cutting reproduced in 'C V Raman – A pictorial biography (Ramaseshan, 1988). The original does not contain underlining.

<sup>&</sup>lt;sup>5</sup> For biography of the discoverer of the Krishnan effect, see, Srinivasan, R. Rappal Sangameswara Krishnan (1911-1999), *Biog. Mem. Fellow INSA* 28(1999):63-82

Again in the late 1930s Raman fixed the date of his discovery as 28<sup>th</sup> February (Fig. 5).



**Fig. 5.** Diagram explaining the principle of Raman spectroscopy (Credit: Raman Research Institute, Bangalore).

As far as the coining of the term Raman effect is concerned, Raman's student, L A Ramdas was the first Indian to use the term, in an article, which he sent to '*Nature*' on May 6, 1928 (Ramdas, 1928 p. 57). However, before him the French physicists in April 1928, applied it, as is evident from the bibliography on Raman effect, which was prepared by A S Ganesan, Raman's student. It contains 160 entries, not 150 as the title suggests (Ganesan, 1929 pp. 281-346).

In Germany, Peter Pringsheim, Berlin, repeated Raman's experiment successfully. In "Der Ramaneffekt, ein neuer von C V Raman entdeckter Strahlungseffekt" (The Raman effect – From C.V. Raman discovered a new radiation effect) (Pringsheim, 1928 pp. 597-606) he coined the terms 'the Raman effect' and 'Ramanlines'. One of Raman's students, S K Sircar recalled:

He [Raman] told us that the controversy about the priority of the discovery was settled when the effect was named after him alone. This was in reference to the discovery of some new lines in the spectra of light scattered by some transparent crystals made by [G S] Landsberg and [L I] Mandelstam" (Sirkar, 1988).

# 4. The Landsberg-Mandelstam effect, the Combination Scattering and Raman-Landsberg-Mandelstam effect

The Russian physicists L I Mandelstam and G S Landsberg (Fig. 6) started taking spectrum of a crystal (iceland spar) on 23<sup>rd</sup> and finished on 27<sup>th</sup> of February 1928 (Fig. 7). In 1978, and later in 1998, the Russian historian I.L. Fabelinskii, claimed, his countrymen discovered the effect nearly at the same time as Raman (Fabelinskii, 1978, 1998).



**Fig. 6.** L I Mandelstam (Credit: Russian Academy of Science), G S Landsberg (Credit: Prof. I L Fabelinskii)

However, much before I L Fabelinskii, O Chwolson, Leningrad, on January 6, 1930, nominated his countrymen for the Physics Nobel Prize and claimed that Raman observed the phenomenon on Feb. 16<sup>th</sup> and sent his results to '*Nature*'. This effect is known under the name 'Raman Effect.' Further:

"... Professors G.S. Landsberg and L.I. Mandelstam in Moscow have seen and explained the same phenomenon on February 21st. ... Unfortunately the Russian scientists did not hurry to make their discovery known. If they had written to *Nature* on February 21st, their communication would have been published earlier than Raman's; in that case there would not have been a 'Raman effect', but a **'Landsberg-Mandelstam-effect'** or **an effect designated to all the three names**" (emphasis added) (Chwolson, 1930).



**Fig. 7.** The first spectra taken by Landsburg and Mandelstam on 23<sup>rd</sup> to 27<sup>th</sup> Feb. 1928. Credit: Prof. I.L. Fabelinskii

About three decades later, Russian physicists applied the term as suggested by O. Chwolson. For instance, while discussing 'Rayleigh scattering in gases and liquids' the Russian authors L D Landau and F M Lifshitz wrote:

Two types of scattering can be distinguished, depending on the change in frequency of the light: (1) *combination scattering* (emphasis in original), which is the **Raman-Landsberg-Mandel'shtam effect** (emphasis added) and results in the appearance in the scattered light of lines whose frequency differs from that of the incident light, (2) Rayleigh scattering, in which the frequency is essentially unchanged" (Landau, 1960 p. 387).

G Venkataraman shows that the term 'the Smekal-Mandel'shtam-Raman scattering' had been also mentioned by the Russian authors L.D. Landau and F.M. Lifshitz (Venkataraman, 1994, p. 206).

# 5. The Smekal-Raman effect or Raman-Smekal effect

In 1925, the Austrian scientist Adolf Smekal (Fig. 8) by assuming that light has quantum structure, had shown that monochromatic light after scattering will have not only the original frequency but also frequencies of higher and lower wavelength (Smekal, 1925 pp. 241-244). However, the complete explanation of absorption,



Fig. 8. Adolf Smekal<sup>6</sup>

scattering and dispersion processes was given by H A Kramers and Werner Heisenberg (Kramers, 1925 pp 681-708).

In the year 1931 the term "Smekal-Raman-Effekt" was introduced by K W F Kohlrausch, Graz, Austria (Kohlrausch, 1931). Study of German literature shows that in books such as "Physik - Ein Lehrbuch für Studierende an den Universitäten und Technischen Hochschulen" [A text book for students at the Universities and Technical Colleges] (1963), and 'Geschichte der Physik' [History of Physics] (Hermann, 1972 p. 350) this term is to be found. Also it is mentioned in scientific journals like 'Physikalische Blaetter' (8, 364-365, 1951); and of course in different dictionaries from the years 1969, 1971 and 1972 (Singh and Riess, 1998 pp. 1112-1115).

### 6. The Cabannes-Daure Effect

Within Europe, in France there were different physicists who were working on light scattering in gases. J Cabannes (Fig. 9) was one of them, who started his researches under Charles Fabry. It is worth to mention that Fabry was wellknown in Indian circles. For instance, Raman's student, LA Ramdas, at the University of Calcutta, submitted his D.Sc. thesis: 'The scattering of light by liquid surfaces and other related phenomena'. It was evaluated by a Board of Examiners, with members: Lord Rayleigh, Prof. C Fabry (Sorbonne) and Prof. C G Darwin, FRS (Edinburgh).<sup>7</sup>

<sup>&</sup>lt;sup>6</sup> https://www.flickr.com/photos/alessandrovolta1745/6008329901, Oct. 16, 2017

<sup>&</sup>lt;sup>7</sup> Minutes of syndicate 1926 - III, Calcutta University Press, Calcutta, 1926, p. 2058



Fig. 9. Jean Cabannes<sup>8</sup>

In '*Comptes Rendus*' 186, 1712-1713, June 18, 1928, an article: 'Quelques précisions au sujet des effets Raman et Cabannes-Daurer' ('Some details about the Raman and Cabannes-Daure effects') was written by the French A Bogros and Y Rocard. It was summarised in '*Science Abstract*' 31, 772, 1928, as follows:

Only the coherent radiation scattered by a transparent body undergoes marked increase in intensity near the critical point and produces opalescence. An experimental study of the light scattered by the opalescence mixture of water and phenol indicates that the Raman rays are incoherent. Broadening and displacement of scattered radiation lines in opalescence (Cabannes-Daure effect) are not found for this mixture, and an explanation is advanced based on the distinction between coherent and incoherent radiation.

According to Wikipedia, in 1928, J Cabannes, P Daure and Y Rocard discovered the change in the wavelength of monochromatic light after scattering by gases. This effect was named as the Cabannes-Daure effect. In '*Comptes Rendus*' 186, 186, 1928; on April 23<sup>rd</sup>, 1928, Y. Rocard reported under the title 'Les nouvelles radiations diffuses', about Raman's discovery which was published in the '*Indian Journal of Physics*' 2, 1928. He told that theoretically he had predicted the phenomenon discovered by Raman. However, he had to postpone the publication of his results, which he presents now. He also suggested: 'that the diffuseness of the lines appearing in the spectrograph of benzene scattering reproduced in Raman's paper might be due to disturbing causes, such as intermolecular fields, polymerisation, etc (Ganesan, 1929, pp. 281-346).' In '*Comptes Rendus*' 186, 1201, 30<sup>th</sup> April, 1928, J Cabannes, while discussing the Raman effect suggested 'that the new radiation can be explained by the phenomenon of 'optical beats' predicted by the author in 1924' etc (Ganesan, 1929, pp. 281-346).

# 7. Indian Newspapers reporting the European Reaction to Raman's discovery

After the discovery by Raman, Smekal wrote a letter to the discoverer. On May 31, 1928, Raman told to a journalist:

Professor Smekal of Vienna, a distinguished physicist and authority on the quantum theory has written conveying congratulations ... on his [Raman's] 'wonderful and fundamental discovery' and emphasising that it opens out an entirely new territory of research.

On May 31, 1928, 'The Basumati' entitled an article 'Prof. Raman's Remarkable Discovery – New kind of radiation of light', therein it was written:

Professor Raman's discovery of a new kind of radiation is attracting world-wide attention. A 'Free Press' representative who called on Professor Raman found him in a communicative mood and naturally rather pleased with the evidences of the appreciation of his work by scientific men abroad. Smekal of Vienna, a distinguished physicist and authority on the quantum theory, had written conveying congratulations to our Professor on his 'wonderful and fundamental discovery' and emphasising that it opens out an entirely new territory of research.

#### And further:

The French school of Physicists under Professor Fabry and Brillowin [Léon Brillouin] are very excited about the discovery and within a week of

<sup>&</sup>lt;sup>8</sup> https://d1k5w7mbrh6vq5.cloudfront.net/images/cache/89/5a/f7/895af782ee81e60e8c6a22edc2b3704b.png, Oct. 20, 2017

the news reaching Europe, two papers have been published by the Paris Academy of Sciences, in its 'Comptes Rendus' commenting on it as an 'extremely remarkable discovery'.

As we see from A Ganesan article, within a year of the discovery 160 articles appeared in international journals. The main reasons for that were the practical application of the effect as well as the availability of the instruments required of the work on light scattering in different laboratories. Within a short period of one year, Raman and French physicist J Cabannes were nominated for the Noble Prize. Shortly after that followed other nominations. However, only Raman got the Physics Nobel Prize (detail below).

# 8. Why the Nobel Committee "ignored" A Smekal, K S Krishnan, J Cabannes and Russian physicists?

The details on the nomination and decision of the Nobel Committee had been discussed in different articles (Singh, 1999; Singh and Riess, 2001; Singh and Riess, 2004). To summarize the results: (I) K S Krishnan and A Smekal were not nominated. (II) As French scientists thought to have discovered the 'Raman effect' in gases; they claimed for the Nobel Prize. In 1929, C Fabry recommended J Cabannes and C V Raman for a shared Nobel Prize (Kueppers, Weingart and Ulitzk, 1982 p. 25). In the same year, N Bohr proposed - Either the Prize be awarded to the American R W Wood, USA.; or to be shared between Wood and C V Raman<sup>9</sup>. From the history of the Nobel Prizes we know that for the year 1929, the Prize went to the French Louis de Broglie "for his discovery of the wave nature of electrons." (III) For the Nobel Prize for the year 1930, N Papalexis nominated Mandelstam<sup>10</sup>; whereas O Chwolson suggested that half of the prize should

go to Raman and rest should be divided between the Russian physicists<sup>11</sup> (Chwolson, 1930). In Raman's favour there were 10 proposals (out of them 6 for unshared Prize) (Crawford, Heilbron and Ullrich, 1987 pp 120-123). The expert of the Nobel Committee, Erik Hulthen, Director of the Physical Institute concluded<sup>12</sup> rejected the proposals of Prof. Chwolson and Prof. Papalexis as he was of the opinion that the Russian physicists did not come to an independent interpretation as they referred to Raman's work. According to the judgement of the Nobel Committee Raman proved the universal nature of the phenomenon as he studied the effect in liquids, gases and crystals. Russian nominators had also claimed that Mandelstam predicated the effect theoretically. About it the Committee was of the view that A. Smekal in 1923, and H A Kramers and W Heisenberg in 1925, had already given this explanation.13

On September 20, 1930, the Committee asked 'the Academy to award the Nobel Prize for Physics for the year 1930 to Sir Chandrasekhra Venkata Raman, for his work on the scattering of light and for the effect named after him (Singh and Riess, 1998):1112-1115)." It is to be noted that the Nobel Committee can only recommend candidate/s. The Swedish Academy of Sciences is empowered to take the final decision. In the end the Swedish Academy of Sciences gave its decision in Raman's favour.

#### 9. CONCLUSIONS

In the beginning of the 20<sup>th</sup> century C V Raman initiated researches on the scattering of light. Under his guidance a number of scholars began research. His student K S Krishnan was one of them. Who later occupied important positions

<sup>&</sup>lt;sup>9</sup> Bohr, N. to the Nobel Committee, Jan. 29, 1929

<sup>&</sup>lt;sup>10</sup> Papalexi, N. to the Nobel Committee, Jan. 6, 1930.

<sup>&</sup>lt;sup>11</sup> Chwolson, O. to the Nobel Committee, Jan. 6, 1930.

<sup>&</sup>lt;sup>12</sup> The report of the expert's for the Nobel Committee, May 26, 1930.

<sup>&</sup>lt;sup>13</sup> The Report of the Nobel Committee, Sept. 30, 1930.

such as the Directorship of the National Physical Laboratory, Delhi. His successor brought to the notice that Krishnan "suffered" as he was not given proper credit. In contrast, until the date historians have ignored the role played by S Venkateswaran. It was his observation, which "forced" Raman to recheck the previously results. The reason for not giving credit to Venkateswaran is that he was not well-known as Krishnan. The moral of the story is, only those leave mark in the historical record, who possess high positions or have influential successors.

Russian men of science rightly give/gave credit to their countrymen for the simultaneous discovery. However, they ignore the cardinal fact in the field of researches, namely, the question of priority is settled on the bases of date of publication. Historical documents leave no doubt that they observed the effect, 'even before' Raman (who himself fixed the date of the discovery as 28th Feb. 1928); and their interpretation of the experimental results was correct. In contrast Raman's interpretations were wrong. He took time to come to the right conclusion. However, the fact is, he took risk and quickly published his results; which were improved in the proceeding publications. Raman's case shows the importance of quick publication and taking risk.

The claim of the Austrian men of science, to call the effect as 'the Smekal Raman effect' is less understandable. As shown in a different article, Raman was not aware of Smekal's work. It was the theory of dispersion given by Kramers and Heisenberg, which stood in the focus of Raman's work for interpreting his results (Singh and Riess, 1998, pp. 1112-1115).

The French men of science accepted the term 'Raman effect', but reduced Raman's discovery to liquids and vapours. In the case of scattering of light by gases, they preferred to coin their own term. How far this continued in the 20<sup>th</sup> century needs further research, as present author does not know French language.

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