Seventy years ago – The discovery of the Raman effect as seen from German physicists

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The Raman effect was announced by Sir C. V. Raman at the Indian Association for the Cultivation of Science in Calcutta on 28 February 1928. After the discovery had been announced, some of the German physicists were sceptical about it, as the intensity of the spectral lines was supposed to be very low and similar experiments done in Munich were not successful. Some letters to and by Arnold Sommerfeld are analysed to support the above facts. In the German-speaking region, the effect was named Smekal–Raman Effekt, and the term is still being used.

In Calcutta, a local newspaper (Associated Press of India) announced a discovery under the headlines 'New Type of Radiation – Professor Raman's Discovery' on 29 February 1928 and also informed about a lecture by Raman on this topic on 16 March 1928 at the South Indian Science Association in Bangalore[1]. The lecture, 'A New Radiation' was delivered and the first spectra produced were presented there. Later it was published in the *Proc. Indian Assoc. Cultiv. Sci.*[2].

Outside India, the discovery of the Raman effect was disclosed by Raman and Krishnan in different articles which were sent to *Nature*[3–5]. From a historical point of view, the article 'A New Type of Secondary Radiation' is considered to be an important paper. As the earlier work on musical instruments by Raman was well known in Germany, the information about this new discovery was also taken notice of by Sommerfeld and his colleagues.

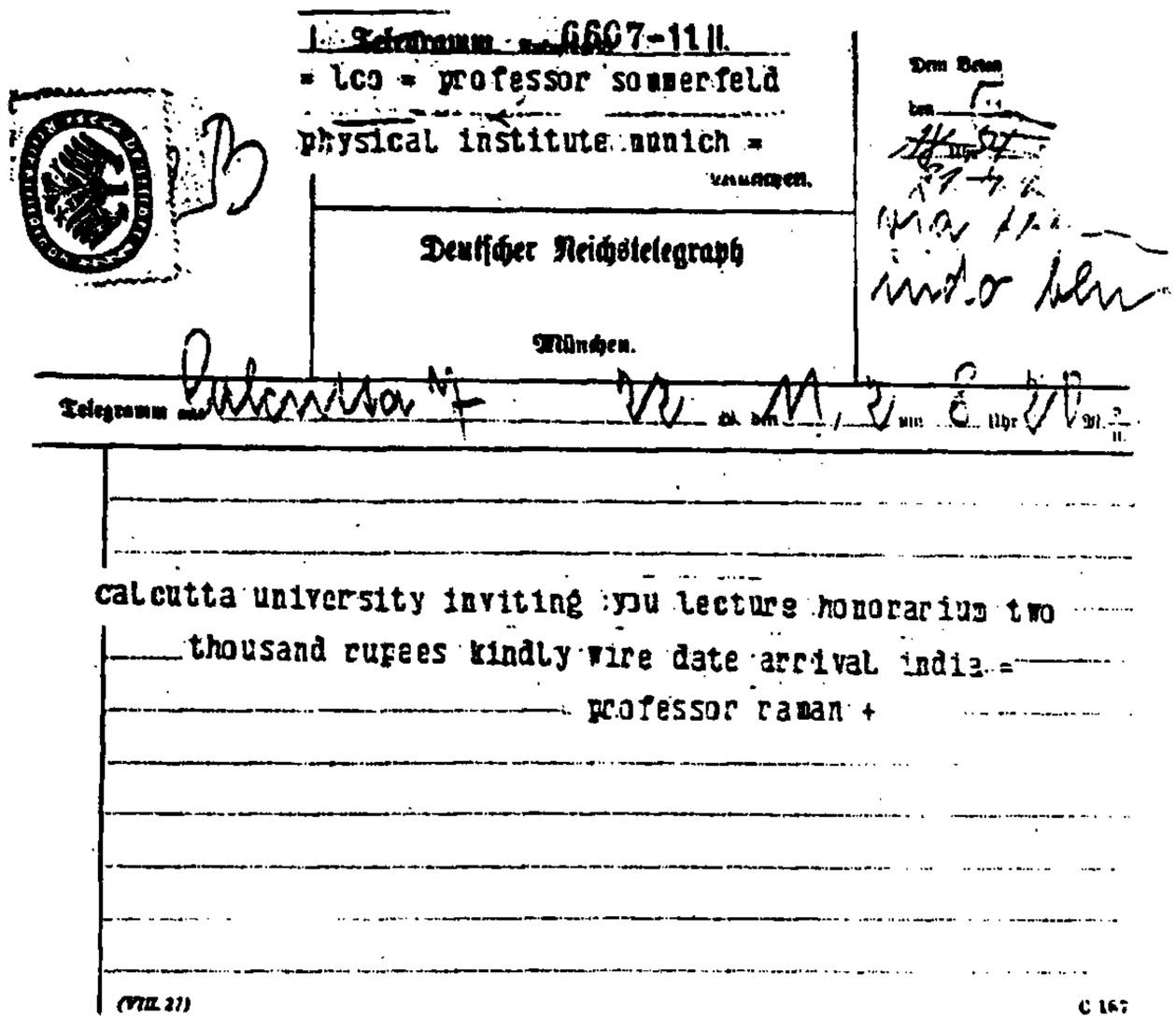
Arnold Sommerfeld and C. V. Raman

Arnold Sommerfeld (1868–1951) is considered as one of the founding fathers of modern theoretical physics. He contributed to the fields of atomic structure, quantum physics and wave mechanics. Some of his students, Pauli, Bethe, Debye and Heisenberg, were well-known and acquired fame in their respective fields. Sommerfeld's first direct contact with Indian physicists was due to the famous Indian astrophysicist, Meghnad Saha (1893–1956), a professor at the University of Allahabad in India. The latter visited Germany during 1920–1921 and worked in the laboratory of W. Nernst (1864–1942), a professor of physical chemistry at the university of Berlin. Saha sent a copy of his paper on *stellar spectra* to Sommerfeld who at once invited him to Munich to deliver a seminar[6]. Sommerfeld was well aware of Raman's work and he appreciated Raman. However, due to an Indian named Ray, who was working in Munich on the Raman's theory of musical instruments and whose results were not in accordance with the above theory, it led to questions on the validity of Raman's work. But Sommerfeld defended Raman in a letter he sent to Otto Blumenthal, a mathematician at the University of Goettingen. Sommerfeld wrote: *I have the firm impression that Ray's work is rubbish. Raman's observations of, who is a good physicist, are by all means correct; but their treatment by Ray seems to be invalid* (translated from a German letter dated 19 February 1927 to Blumenthal).

Raman came to know in early 1928 that Sommerfeld was going to USA via Japan and he sent a telegram to Sommerfeld inviting him to India (Figure 1). This correspondence between Raman and Sommerfeld took place just a few days before the famous discovery of the Raman effect. The text of Sommerfeld's reply to Raman's letter is given in Figure 2. Raman and Saha sent separate letters informing Sommerfeld about the fees, lecture themes and the planned programme of the visit (Raman to Sommerfeld, letter dated 24 March 1928 and Saha to Sommerfeld, letter not dated, 1928). The correspondence between Raman and Sommerfeld was of formal nature. Only once did Raman write about his work to Sommerfeld (Figure 3).

In September 1928, Sommerfeld came to Bangalore and stayed with Metcalfe, principal of Central College in Bangalore. Unfortunately, Sommerfeld fell ill. When Raman, who was in Calcutta at the time, came to know about the health of his guest, he sent Sommerfeld a letter asking him to take care of his health. Raman wrote: *I have been deeply distressed to hear of your illness. It is some satisfaction to learn from Prof. Metcalfe's telegram that it is not very serious. Every one in Indian Universities is of course eager to see and hear you. But even if it involves the cancellation of some of your engagements in Northern India, it will be the desire of every one that you should not endanger your health by travelling until you are quite well again* (Raman to Sommerfeld, letter dated 26 September 1928).

During Sommerfeld's six-weeks stay in India, he got invitations from different universities such as Hyderabad, Patna, Bombay, Benares and Dacca. He stayed in Calcutta from 4 October 1928 to 26 October 1928 where he delivered his lectures. He attended some of Raman's lectures and saw the experimental work being done in Calcutta. As he was impressed by Raman's work, he dropped somebody a hint that he intended to recommend him for the Nobel Prize. When Raman came to know about the proposed suggestion – almost one year later – he wrote to Sommerfeld: *Mr. Rajam from Madras was recently at Calcutta, and he told me that you have proposed my name for the Nobel Prize. I do not know how to adequately express my gratitude for this act of kindness. The literature of the new effect is growing at a great pace. I hope the Nobel Committee may give a favourable*



Telegramm 6667-11 ll.
= lco = professor sommerfeld
physical institute munich =
Deutscher Reichstelegraph
München.
calcutta university inviting you lecture honorarium two
thousand rupees kindly wire date arrival india =
professor raman +

Figure 1. The telegram sent by Raman to Sommerfeld. Two thousand rupees was a big amount of money at the time; in comparison the salary of Prof. Raman was 600 rupees per month[7].

München,,
February 28, 1928

Dear Professor Raman:

I thank you sincerely for your kind cable: I shall be very glad to spend some time with you in Calcutta and to give some lectures there. Please write me how long you expect me to stay at your university. I made, at Como, a provisional arrangement with Prof. Saha to come for some days to Alahabad. I hope you may be willing to put in order my program for India together with Prof. Saha. Therefore I am enclosing my letter for Saha, and beg you to read it and to send it on. I shall have altogether something like four weeks to spend in India.

Very faithfully yours,

Figure 2. Script of letter, written by Sommerfeld to Raman, dated 28 February 1928.

decision in December next if my name is put before them on this occasion also (Raman to Sommerfeld, letter dated 23 January 1929). But it is doubtful whether Raman's name was really recommended by Sommerfeld as there are no written documents to prove this. However, *Raman was proposed for the Nobel Prize for Physics in 1929 by Bohr N. and Fabry C. and in 1930 by Bloch E., Bohr N., deBroglie L. and N. Khvolson, Perrin J., Pfeiffer R., Rutherford E. Stark J. and Wilson C.T.R. Sommerfeld's name does not appear in the list of nominators. However, it is likely that he wrote to the Committee and the recommendation did not get into the official record*[8]. After receiving the Nobel Prize, Raman visited Munich where he was welcomed by Sommerfeld.

The Raman effect and German scientists

Before Sommerfeld left for India, the discovery of the effect had been known among the community of the German scientists, some of whom were sceptical of it. G. Joos who worked on optics, para- and diamagnetism wrote to Sommerfeld from Jena: *Do you think that Raman's work on the optical Compton effect in liquids is reliable? To repeat the experiment is not a big task and most probably we are going to do it. The sharpness of the scattered lines in liquids seems doubtful to me* (translated from a German letter dated 14 May 1928). Further, though the experiments carried out to verify the Raman effect in Munich were not successful, Sommerfeld was sure that the results of Raman were correct. On 9 June 1928 he wrote to Joos: *In my opinion Raman is correct and important. He writes to me, that the difference between the lines is exactly equal to the infra-red frequencies of the molecules under consideration* (translated from a German letter dated 9 June 1928).

Whether Joos really performed the experiment is still not known, but the experiments were indeed carried out in Munich in 1928 by experimental physicist, Wilhelm Wien and F. Kirchner. Sommerfeld's assistant, Heinrich Ott, probably also conducted these experiments, since Kirchner lectured in the summer term of 1928 on light quantums and electrons, and Ott on scattering of light and X-rays (Eckert, M., private commun.). However, it cannot really be ascertained by whom the experiments were actually carried out. Nevertheless, in a letter to Sommerfeld by Pringsheim (1881–1963), working in Berlin in the field of fluorescence, luminescence, photoelectric effect and scattering of light, Pringsheim wrote: *As I know that you are especially interested in the new Raman effect and as I have heard that the experiments done in Munich create doubt about the existence of the effect, I send you some of the spectra of some of the organic compounds, which can be taken using a mercury arc lamp as*

I am these days with my collatorators engaged chiefly in investigating the optical analogue of the Compton Effect which we have discovered. In several papers appearing in the Indian Journal of Physics, we have shown that the difference of frequencies between the incident and the scattered spectral lines is exactly identical with the several characteristic infra-red frequencies, We have an equal number of modified spectral lines. A large field of research has thus been opened up.

With best regards,

Yours sincerely,

C. V. Raman

Figure 3. Letter written by Raman to Sommerfeld on 14 May 1928.

scattering source. I have made still more pictures, these three I have chosen by keeping a definite principle in mind that two of them have very similar but characteristically different spectra (benzene and toluene) and another entirely different substance (CCl_4) to compare with... (translated from a German letter dated 20 June 1928). Thus Raman's results were verified by Pringsheim who published two articles in July under the titles 'Über den Ramaneffekt'[9] and 'Der Ramaneffekt, ein neuer von C.V. Raman entdeckter Strahlungseffekt'[10].

When the initial scepticism had been overcome, the Germans took deep interest in the Raman effect. Back *et al.*[11], Schaefer *et al.*[12] and Kohlrausch[13] wrote detailed articles on it and Kohlrausch's paper contained a bibliography listing 417 references. Moreover, all the abovementioned authors emphasized the application of the Raman effect in the field of chemistry. From 1928 to 1937, Germany held second place in the world as far as the production of papers on this topic was concerned[14]. Due to the importance of this effect, it was included in university text books within two years[15].

Raman effect or Smekal–Raman effect or Raman–Smekal effect?

In the beginning of the twentieth century, studies on light scattering were being pursued in several laboratories, e.g. by the younger Lord Rayleigh (England), R. W. Wood (USA), J. Cabannes (France), G. Landesberg *et al.* (USSR) and C. V. Raman (India). The French and the Russians were working on the scattering of light in gases and quartz respectively, Raman and his collaborators on liquids. As soon as Raman's group came to know that the effect was observable in all liquids, they experimented with gases and solids as well in order to establish the effect as a universal one. As the results of the three groups were published nearly at the same time, this led to a controversy with regard to the priority of this discovery among Russian and Indian scientists. A summary of the Indian and the Russian arguments in the dispute is given by Venkataraman[16] and Fabelinskii[17].

It took nearly half-a-year until the first article was published by Pringsheim 'Ueber den Ramaneffekt'[18], and so the term Raman-Effekt was coined by him in the German-speaking region, whereas L. A. Ramdas had sent a note to *Nature* on 29 May 1928 under the title 'The Raman Effect and the Spectrum of the Zodiacal Light'[19]. Thus he had the honour to use the English term for the first time.

In the year 1931, the term 'Smekal–Raman-Effekt' was introduced by Kohlrausch[20], a physicist at the technical institute of Graz, Austria. Before this time only the term 'Raman-Effekt' had been used; for example in the physics text book for universities and technical institutes by W. H. Westphal, written in the year 1930 (ref. 21). In the 7th and 8th editions[22] (1941) and in the 22nd–24th edition[23] (1963) the term 'Der Smekal–Raman-Effekt' was used. Some more examples for the usage of this term are provided by articles in *Naturwissenschaften*[24] (1935), *Physik griffbereit*[25] (1972), *Geschichte der Physik*[26] (1972) and an article in the journal of the German Physical Society *Physikalische Blaetter*[27] (1951). In many German scientific dictionaries[28–30] (1969, 1971, 1972) though the term Raman-Effekt is being used, at the same time Smekal–Raman-Effekt is given in parentheses. It seems most of the authors take for granted the use of the term Smekal–Raman Effekt.

The use of the term Smekal–Raman effect is questionable. The following will elaborate this further: Undoubtedly, Adolf Gustav Smekal (1895–1959) was the first person who predicted such an effect while trying to explain the dispersion of light in terms of quantum theory[31]. This rudimentary idea was extended further by Kramers *et al.* and the process of light dispersion was completely explained in terms of wave mechanics[32]. However, as the implications were not made obvious by the authors, experimental physicists did not take any notice of the article.

The motivation of Raman's work in Calcutta was not to prove Smekal's prediction. Raman was the first person to give an explanation for the blue colour of the sea. He was convinced that scattering of light can take place even in molecules. To give shape and direction to his research, he wrote a monograph about the molecular dispersion of light[33] (1922) in which he described his own work and the work done by other persons in the field of scattering of light. It included studies of the molecular diffraction of light in systems undergoing phase transitions and the research on the relationship of the light scattering to the chemical constitution. At the same time he discussed the drawbacks of the classical wave theory of light. He did not reject the idea to think of light as a quantum. *So it was somewhat bold of Raman to think in terms of a quantum theory of light scattering when leading lights like Bohr and Planck disapproved of even the idea of a light quantum*[34].

Raman's experimental work was directed to elaborate the process of light scattering. For this purpose, he and his collaborators started with the Rayleigh

theory to explain the blue colour of the sea. Afterwards they modified the famous Einstein–Smoluchowski equation to explain the strong scattering of light near a phase transition, which is referred to as critical opalescence. From the discovery of the Compton effect, Raman was convinced that if there is a change in frequency in the case of X-rays, it should similarly be possible in the case of ordinary light. In the year 1923 they found out some peculiar phenomenon, which they named 'feeble fluorescence' but they could not explain it. Again in 1925 they observed this phenomenon but rejected it on the basis that it might be due to presence of impurities in the liquids. Compton received a Nobel prize in 1927, which gave Raman new impetus. Once again, the liquids were purified more intensively and the experimental work was repeated by his group. In the end they found what they had been searching for was an analogue of the Compton effect for ordinary light. As the first paper sent to the journal *Nature* shows, this was their main motivation; *If we assume that the X-ray scattering of the unmodified type observed by Prof. Compton corresponds to the normal or average state of the atoms and molecules, while the 'modified' scattering of altered wavelength corresponds to their fluctuations from that state, it would follow that we should expect also in the case of ordinary light two types of scattering...* (ref. 35).

For the interpretation of their experimental results, Raman took the help of the Kramer's theory. An entry in the diary of Krishnan (1898–1961) confirms this. On 7 February 1928 Krishnan wrote: ...*When we went down, we found he was much excited and had come to tell me that what we had observed that morning must be the Kramers–Heisenberg Effect we had been looking for all these days. We, therefore, agreed to call the effect 'Modified scattering'. We were talking in front of our house for more than a quarter of an hour when he repeatedly emphasised the exciting nature of the discovery*[36].

Thus the experiments done on the molecular scattering of light were not directed to prove the (predicted) 'Smekal effect'. Furthermore, if a scientific effect is named after two persons generally the names are arranged in alphabetical order, e.g. Bose–Einstein statistics, Einstein–Smoluchowski equation and so on. Thus to call the effect Smekal–Raman effect is not correct. In case the credit has to be given to Smekal, the right term would have been Raman–Smekal-Effekt. Therefore, a correction in the naming of this effect seems to be justified in German literature on this subject.

Conclusion

The analysis of different letters written to Sommerfeld within Germany has shown that the relations between him and Raman were rather close, especially after Sommerfeld's visit to India. The scepticism about Raman's results among German physicists is understandable up to some extent as only few of them were working in the field of light scattering. Further, a certain underestimation of experimental work carried out by Raman exists among some German scientists largely due to the name 'Smekel–Raman-Effekt'. Therefore, a correction in the naming of this effect to 'Raman–Smekal-Effekt' is called for.

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