Scientific session of the Division of General Physics and Astronomy, Academy of Sciences of the USSR (29-30 March, 1978)

Usp. Fiz. Nauk 126, 673–689 (December 1978)

PACS numbers: 01.10.Fv

A session of the Division of General Physics and Astronomy of the Academy of Sciences of the USSR was held on 29 and 30 March, 1978 at the conference hall of the P. N. Lebedev Physics Institute. The session was dedicated to the 50th anniversary of the discovery of Raman scattering. The following papers were presented:

1. *A. M. Prokhorov,* Introductory remarks.

2. *M. A. Leontovich,* Historical aspects of the discovery of Raman scattering.

3. *M. M. Sushchinskiĭ,* Raman scattering of light in phase transitions on crystals.

4. *V. M. Agranovich,* Surface electromagnetic waves and Raman scattering by surface polaritons.

5. *V. T. Aleksanyan,* Certain applications of Raman spectra in chemistry.

6. *Yu. N. Denisyuk,* Present state and prospects for development of holography with recording in three-dimensional media

7. *Sh. D. Kakichashvili,* Polarization holography.

8. *B. Ya. Zel'dovich and V. V. Ragul'skiĭ,* Wave-front inversion in induced light scattering.

9. *V. G. Sidorovich,* The mode theory of the three-dimensional hologram.

10. *V. I. Bespalov, A. A. Betin, and G. A. Pasmanik,* Reproduction of the light-beam wave front in induced scattering.

Brief contents of eight of the papers are published below.

M. A. Leontovich. *Historical aspects of the discovery of Raman scattering.* Of those who witnessed the discovery of Raman scattering, there are today perhaps only two survivors: Frida Solomonovna Landsberg and I. Her ability to recall minute details has been of great assistance to me; it would appear that her memory is much better preserved than mine.

I can testify as a witness, of course, only to the work of L. I. Mandel'shtam and G. S. Landsberg. As to that of Raman and Krishnan, I can add nothing to what has already been published. I shall therefore limit myself to brief remarks on their work at the end of my report.

Research on the molecular scattering of light was begun at Moscow University as soon as the Department of Theoretical Physics was organized and placed under Mandel'shtam's direction. This took place in 1925, and the work on light scattering dates from that time. As far as I known—and this may perhaps explain a great deal of the subsequent history of this work—Mandel'shtam had been away from optical research for a long time, since he was working at the Central Radio Laboratory at Leningrad. Those were, as we all remember, difficult times for scientific activity. And that is why Mandel'shtam's transfer to Moscow University was essentially a return to his scientific career in physics or, more precisely, in optics.

At the very outset, in 1926, Mandel'shtam published a theory of the phenomenon that we now know as the Mandel'shtam-Brillouin doublet. This paper was actually the stimulus to further development of research on light scattering. The ultimate objective came to be the observation of the doublet in the spectrum of scattered light that was predicted by Mandel'shtam's formula

$$\frac{\delta\nu}{\nu}=\pm 2\frac{a}{c}\sin\frac{\theta}{2},$$

where θ is the scattering angle, a is the velocity of sound, c is the velocity of light in the medium, and $\delta\nu$ is the deviation of the frequency of the scattered light from the frequency ν of the incident light.

The experiments started with Landsberg's work on the scattering of light in crystals. Why were crystals chosen even though it was known that molecular scattering is weaker in crystals than in liquids of similar density? Because it was feared that the doublet splitting of lines in the scattered-light spectrum in liquids would be indistinguishable owing to the strong absorption of the sound. At that time, no data were available on the absorption of sound at very high frequencies. Recourse was therefore taken to study of light scattering in crystals.

In 1926, Landsberg began his work on measurement of the total intensity of light scattered by quartz crystals. It was necessary first of all to establish that molecular scattering does indeed dominate in good quartz crystals and that scattering of light due to imperfections of the crystal lattice is insignificant. The temperature dependence of scattered-light intensity was taken as a criterion of this. For molecular scattering, the effect should be proportional to temperature. Landsberg demonstrated as a result of about two years of investigation that the scattering in good quartz specimens does indeed depend on temperature, and that the admixture of nonmolecular scattering amounts to about 20% in the best specimens.

Preparation were begun to detect, in these superior quartz specimens, the effect that had been predicted by Mandel'shtam's theory. It was clear that the shift of the spectral lines would be very small and could not be detected with conventional prism spectrographs. Because of the smallness of the effect, it was necessary to use interference spectroscopy, but the laboratory owned only one Lümmer-Gehrcke plate, and it was not even of quartz, but of glass. It was decided first to obtain a scattered-light spectrum on a crude instrument and then to study it further for finer features. Systematic recording of spectra was begun with the only quartz spectrograph in the laboratory.

Worries began very soon after the first spectrograms were produced: either the spectrograph was a poor one or something in the crystal was somehow reflecting and producing false lines. I remember these conversations very clearly. The terminology usually used was "false light coming from somewhere." What we were actually observing on the spectrograms were Raman satellites. An experiment with mercury vapor was decisive in making this clear. We investigated the scattering of the 2536 Å mercury resonance line in the quartz. A small quartz flask containing mercury vapor was placed in front of the spectrograph slit. It was clear that the resonance line should be absorbed completely in the mercury vapor if its concentration and the thickness of the flask were properly adjusted. One of the very first spectrograms obtained by this procedure showed that the 2536 Å line was, as expected, absorbed. The secondary lines were not absorbed. This indicated that they had other wavelengths, i.e., were definitely not "false light" with the same wavelength that had somehow strayed into the spectrograph. The psychological effect of this experiment was decisive. It would have been sufficient, of course, to pick up scattered-light spectra from several different spectrographs. "False light" would have produced different patterns on different spectrographs. The fact that only the one spectrograph was available forced the clever recourse to mercury vapor. This experiment was, naturally, more convincing than the multiple-spectrograph experiment.

The conviction that the satellites were real began to grow after these experiments, and it was established at once that many of the differences between the satellite frequencies and the frequency of the exciting line were equal to quartz frequencies in the infrared. This relation did not hold for all of the satellites. Some of the frequency differences disagreed with frequencies in the absorption bands of quartz in the infrared.

Thus, the actual existence of frequency-shifted lines or satellites in the scattered light was reliably established. This was followed surprisingly quickly by understanding of the phenomenon. Surprising or not, it is a fact that Mandel'shtam, who had meditated long on the Debye theory, knew nothing of its development by Born at that time. This is why the experiments that he devised were, if you wish, concerned with testing of conclusions from the Debye theory of solids, which gave only a very small shift of the spectral lines on light scattering. I had long found myself unable to believe this, but Mme. Landsberg still confirms it. Why it happened this way can easily be understood. Mandel'shtam had been taken away from the range of interests that had occupied him during the Strasbourg period of his life. At Strasbourg, he had worked on both optics and radio. He later "went over" into radioengineering research of a more technical nature. Later still when the foreign scientific journals had begun to appear and inferences could be drawn as to the progress of physics, he was not abreast of its over-all development. Afterward, however, understanding of the phenomenon that he and Landsberg had discovered came very quickly. The relation to Born's theory was established within a couple of weeks, and it was also understood why there are lattice vibration frequencies that do not appear in infrared spectra: these are the frequencies of types of vibrations in which the electric moment does not change and which are therefore not accompanied by emission.

Thus, the complete picture of the effect emerged very quickly. It became clear that while a search was being made for scattered-light frequency shifts due to acoustic vibrations of the lattice much larger shifts governed by the optical branches of the lattice vibrations were found.

It very soon became clear that the changed frequencies were already present in the quantum theory of dispersion created in 1925 by Kramers and Heisenberg. They had been, so to speak, predicted even for single molecules, and not only for crystals or complex systems. As I remember it, I. K. Tamm had a very important role in the discussions and clarifications of these problems. He had already then a good grasp of the quantum mechanics of the day and knew Heisenberg and Kramers' work, and it was he who carried quantum concepts over into light-scattering theory.

What happended then? The search for the doublet continued further, although, of course, attention had been diverted to the study of the newly discovered phenomenon, Raman scattering. The equipment in Landsberg's laboratory turned out to be totally unsuitable for study of the doublet predicted by Mandel'shtam's theory. An agreement was therefore reached with D. S. Rozhdestvenskiĭ concerning performance of this work in the Optical Institute of Leningrad. Rozhdestvenskiĭ placed E. F. Gross in charge of the research. In 1930, after two or two and a half years, the Mandel'shtam-Brillouin satellites were observed in an experiment. Here we should note a historically or perhaps psychologically interesting point: Gross first detected this phenomenon in a liquid. Naturally, the experimentor makes his investigations where it is simplest to experiment. The scattering intensity is much higher in liquids than in crystals, and it is simpler to experiment with liquids. Both I and Mme. Landsberg in particular remember how this astonished Mandel'shtam, because the appearance of the doublet in a liquid did not conform to the then prevailing conceptions of sound absorption in liquids.

Now a few words on Raman are in order. It is striking how people working from totally different ideas can arrive at the same discovery. Raman's first paper states quite insistently that there should be a phenomenon in optics analogous to Compton scattering of x-rays. This was the basic idea that motivated his search. His first publication describes an experiment in which sunlight was used in a study of light scattering. He used a strong light source of unlimited availability at Calcutta. The strongly focused solar radiation was not decomposed spectrally. He used a crossed-filter technique with one red and one green filter. In the idea behind it, this is the mercury-vapor experiment in a crude form. I must say that I cannot understand how Raman could have expected any success with these filters. No filter exists that is capable of separating scattered-light components that are frequency-shifted by tens or even hundreds of cm^{-1}. But this did not discourage him, and his experiment somehow convinced him of the reality of the phenomenon. In later experiments, he used a mercury lamp and a spectrograph and the entire procedure was more reliable. It would seem to me that the analogy with the Compton effect would have come up naturally five years later, when Ya. I. Frenkel' introduced the phonon concept. Nevertheless, the fact remains that the same phenomenon was discovered by totally different routes.

I can still remember how we took our turns at the mercury lamp. Recording went on for weeks, and it was necessary to come and "trim" the mercury lamp to make it burn steadily. The filmholder had to be loaded and the films developed in total darkness. Here I had a terrible time with Landsberg. He was a very polite fellow. When he wished to show displeasure, he would display exaggerated politeness and delicacy, and I found this harder to bear than the most violent scolding. Such was the penalty that I drew on one occasion when, in loading a plate for a week-long exposure, I put it in backward. This, of course, produced two or three pitiful lines of the mercury spectrum...

Now a few words on the history of publication of Landsberg and Mandel'shtam's work. Why was publication of the discovery held up? Chiefly because of the mercury-vapor experiment. It had to be performed very precisely, the thickness of the quartz flask had to be adjusted, and so forth. These were not simple matters, and this took up at least one and a half to two months. The results were obtained in the fall, but the paper was not written until February or March.