LETTERS TO THE EDITORS

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The Raman Effect in Rocksalt

THE theory of the Raman effect in crystals, as found in the literature, is rather unsatisfactory. Apart from some general symmetry considerations, the only attempt known to us of establishing definite formulæ is that by Fermi and Rasetti[1] who gave the correct expressions in a general way for explaining the observations on rocksalt. These observations made by Rasetti[2] show clearly a continuous spectrum with small superimposed peaks. The theory mentioned explains this general aspect satisfactorily; but Fermi and Rasetti find the formulæ too complicated for quantitative discussion.

We have taken up the same problem again because these same observations have been used by Raman and his pupils as an argument against lattice dynamics and in favour of their own theory of lattice vibrations. The latter contends that a crystal lattice has a vibration spectrum of a small number of frequencies, in contradiction to elementary laws of classical or quantum mechanics, according to which the number of vibrations of a system of N particles is $3N$, hence quasi-continuous for a crystal of finite dimensions (N large).

Krishnan[3] interprets Rasetti's published reproduction of the Raman spectrum as a line spectrum of nine Stokes lines (and nine anti-Stokes lines), neglecting the very strong background effect. This is rather strange, since the publication of the Italian authors also contains a photomicrograph of the density distribution, which shows that the continuous background is an essential feature. The original picture is reproduced herewith (Fig. 1).

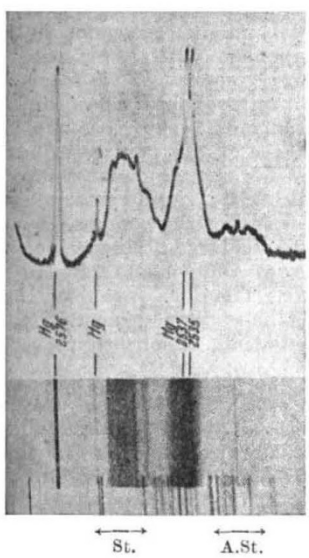

Fig. 1. LOWER: PHOTOGRAPH OF RAMAN SPECTRUM SHOWING FAINT LINES ON BACKGROUND. ST., STOKES LINES. A.ST., ANTI-STOKES LINES. UPPER: PHOTOMICROMETRIC CURVE OF INTENSITY DISTRIBUTION, SHOWING THAT THE APPARENT LINES ARE SMALL PEAKS ON A STRONG BACKGROUND.

Krishnan further states that the number of lines (nine) is in agreement with Raman's theory; but he makes no attempt to compute the frequencies and intensities of these nine 'lines'.

In these circumstances we thought it necessary to work out the complete theory of the Raman effect in crystals and apply it to the case of rocksalt. This substance is particularly suitable, as Kellermann[4] has worked out the vibration spectrum in detail. His numerical results are independent of any arbitrary constants and are based only on the measured values of the lattice constant and compressibility. Using these results, we have derived the Raman spectrum,

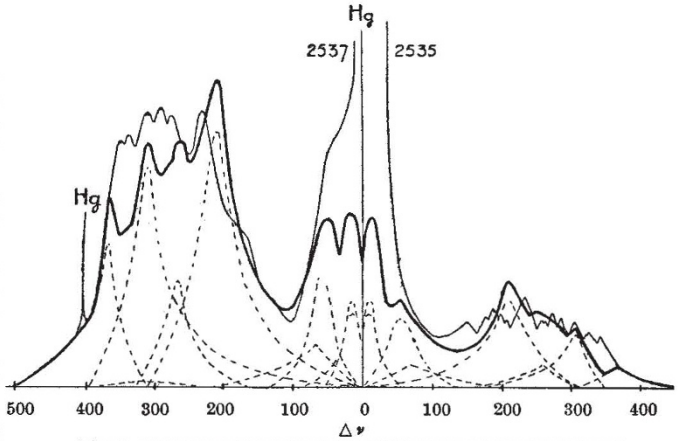

Fig. 2. FINE LINE REPRESENTS FERMI-RASETTI PHOTOMICROGRAPH (FIG. 1). BROKEN LINES ARE APPROXIMATE THEORETICAL CONTRIBUTIONS FROM PAIRS OF FREQUENCY BRANCHES. THICK LINE IS THEORETICAL CURVE FOUND BY SUPERPOSITION OF THESE.

which is in this case a second-order effect with respect to the amplitudes of vibration, and we present the results in Fig. 2.

It shows that the general features of the observations (continuous background with small peaks) come out very well from the theory. The range of the continuum and the positions of the maxima are obtained without any arbitrary assumptions, while the intensities are fitted to the experiment by adjusting some coupling constants. The formulæ for the intensities are so involved that we have picked out only the main terms. It is quite possible that further careful study would improve the agreement.

The Indian physicists have published new and valuable observations of a similar kind (on Raman effect and other optical phenomena in crystals) and have interpreted them as evidence for their theory. We are convinced that ordinary lattice dynamics will be able to explain these facts in a satisfactory way, though the calculations may be involved.

MAX BORN.
MARY BRADBURN.
Department of Mathematical Physics,
University, Edinburgh.
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[1] Fermi, E., and Rasetti, F., *Z. Phys.*, **71**, 689 (1931).
[2] Rasetti, F., *Nature*, **127**, 626 (1931).
[3] Krishnan, R. S., *Proc. Ind. Acad. Sci.*, A, **18**, 298 (1943).
[4] Kellermann, E. W., *Phil. Trans.*, A, **238**, 513 (1940).

The Editors, on receiving our communication, sent us the proofs of a new note by Prof. Krishnan on the same subject published in *Nature* of September 1.