X-Rays and the Eigen-Vibrations of Crystal Structures

In evaluating the effect of the atomic movements in a crystal on the propagation of a beam of X-rays through it, such of these movements as may be identified with the elastic modes of vibration of the solid may be taken up first for consideration. Each of these modes presents a distinctive pattern of nodal surfaces, the frequency of the oscillation being the same everywhere and the phase being opposite in every pair of adjacent cells in the pattern. The configuration of the nodal surfaces in any given mode is determined by various *macroscopic* factors, namely, the form of the crystal, the external boundary conditions and the elastic anisotropy, besides being dependent on the frequency of the mode. Hence, it would bear no particular relation to the atomic architecture of the crystal and would also be different for each of the numerous possible modes. These circumstances have important consequences in relation to the optical aspects of the problem. Each atom in the track of the X-ray beam may be considered as a source of secondary radiations of frequencies ν, $\nu + \nu^*$ and $\nu - \nu^*$, ν being the frequency of the X-radiation and ν^* that of the elastic vibration under consideration, and their effects have to be summed up over the illuminated volume. Since the amplitudes and phases of the radiations of modified frequency depend on the amplitudes and phases of the atomic movements, they would vary from atom to atom in a manner determined by the configuration of the nodal pattern, and hence would be different for each different ν^* and in no case would be definitely correlated with the internal architecture of the crystal. It follows that these scattered radiations cannot possibly give rise to any geometric diffraction pattern exhibiting an observable relationship to the structure of the crystal.

It remains to consider the possible vibrations in the solid which cannot be described as elastic modes, but which, on the other hand, are specifically related to the structure of the crystal and may therefore be designated as its eigen-vibrations. That such vibrations exist is proved by spectroscopic investigations, and their number and nature have been investigated by me[1] with results which have found experimental confirmation in various cases[2]. Since these eigen-vibrations are specifically related to the structure of the crystal, the secondary radiations of modified frequency resulting therefrom should be capable of giving rise to geometric diffraction patterns having a recognizable relationship to the internal architecture of the crystal. Theoretical considerations thus lead us to recognize the fundamental connexion between the eigen-vibrations of crystal structures and the dynamic X-ray reflexions exhibited by them. The existence of such a relationship and the possibility of the eigen-vibrations being excited even in the absence of thermal agitation first become apparent from the experimental facts discovered in the case of diamond by the present writer and Dr. P. Nilakantan[3].

C. V. Raman

Indian Institute of Science,
Bangalore.
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[1] *Proc. Ind. Acad. Sci.,* A, **18,** 237 (1943).
[2] *Proc. Ind. Acad. Sci.,* A, **26,** 339 (1947).
[3] *Current Science,* **9,** 165 (1940).