Progress in the study of Raman scattering by crystals is reviewed. The observed phenomena are related to the theory of longwavelength lattice vibrations, particularly for noncentrosymmetric crystals. The possible observation of light scattering by solidstate excitations other than phonons is discussed. [The SCI® indicates that this paper has been cited over 570 times since 1964.]

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July 23, 1979

"It was obvious to most workers in Raman spectroscopy in the early 1960s that the laser should provide an ideal source for light scattering experiments. Raman’s discovery of his eponymous effect in 1928 stimulated a large amount of experimental and theoretical work on crystals during the following 20 years. This effort had, however, largely died down by the 1950s because most of the crystals available in sufficiently high optical quality and transparent to the mercury lamps then used as light sources had already been thoroughly studied. A relatively small amount of useful work continued, mainly in India, France, Russia, and Canada.

"On the other hand, the theory of excitations in crystals had become well developed by 1960, partly as a result of the growth of interest in solidstate devices. However, because of the rundown on the experimental side, nobody had thought of applying this theory to the Raman effect. My own work began as an extension of some calculations of the nonlinear optical properties of crystals, then a rapidly blossoming research area. This led to a comprehensive microscopic theory of light scattering by crystal vibrations which could in principle predict all the measurable features of the Raman spectra.

"In addition to vibrations, most other kinds of crystal excitation produce some Raman scattering, although none of these were seen with mercury sources. It was possible to calculate scattering cross sections for many excitations, including spin waves, electronic transitions, and polaritons.

"The article gathered together my own and other people’s work in these areas. Its appearance coincided with the first experimental papers on laserexcited Raman spectra of crystals—indeed these were published between submission and the proof stage of my article. Its largely theoretical material has been widely applied in the ensuing renaissance of solidstate Raman spectroscopy. Almost all the predicted phenomena have been found, and have even become commonplace. Of course the article now shows its age with many interesting aspects of subsequent discovery omitted. I considered updating it for a review journal, but the field has become too big to be covered in a mere article, and the outcome is a book Scattering of Light by Crystals written in collaboration with William Hayes and published by Wiley in 1978, appropriately the 50th anniversary of Raman’s original observation."

"Looking over my article today, the comment, ‘It seems likely that the Raman spectra of many crystals will be measured for the first time in the next few years’ may seem unduly cautious, but the laser was already four years old and experimental spectra were slow to appear in print."