

RAMAN AND CRYSTAL DYNAMICS *

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I DEEM it a privilege and honour to be able to participate in this session honouring the memory of late Professor Raman. Raman belonged to that rapidly dwindling species of scientists known as natural philosophers, and it is not surprising therefore that his interests were wide and far flung. One area to which he devoted considerable attention was the physics of crystals, in particular their dynamics. In this he was in distinguished company since several other giants like Einstein, Debye and Born were also attracted at one time to the same subject. In this talk I propose to review certain aspects of crystal dynamics and in the process set Raman's contribution to this subject in its proper perspective. Hopefully, my talk will not only bring back to you nostalgic memories of those years when Raman was vigorously propounding his ideas, but also offer you glimpses of recent developments of which I can speak with some familiarity having personally witnessed them.

In order to appreciate Raman's work in the proper historical context, it is necessary to go back to the turn of the century. At that time there existed a law called the Dulong-Petit law based on classical physics which stated that the specific heat of a solid must be temperature independent. However, this was contradicted by experiments which showed that specific heat diminished with temperature and approached zero as $T \rightarrow 0^\circ \text{K}$. The failure of the Dulong-

Petit law remained a puzzle for many years until Einstein appeared on the scene. Einstein¹ assumed as is done in the derivation of the Dulong-Petit law that all atoms in the solid vibrate with the same frequency and independent of each other. The crystal was thus viewed as an assembly of isotropic, independent, harmonic oscillators. Where Einstein departed from classical theory was in asserting that the average energy of these oscillators is determined by Planck's law rather than by the equipartition law of classical statistical mechanics. By introducing the quantum hypothesis, Einstein was able to show that indeed the specific heat of a solid decreased with temperature and approached zero. While eminently successful as a theory for specific heats, Einstein's picture of the frequency spectrum of the solid was clearly too naive. It is difficult to believe that all the atoms vibrate with just one unique frequency and that too independent of each other. Einstein himself realized these limitations but did not bother to remove them. Soon afterwards, it was noticed particularly by Nernst and Lindemann, that Einstein's theory while qualitatively successful showed certain quantitative deficiencies especially at very low temperatures. This led Debye² to speculate around 1912 that the deficiencies lay in the frequency spectrum assumed by Einstein. Debye reasoned that the vibrations of different atoms must be coupled and that therefore the frequency spectrum could not consist merely of a single δ -function. Following Rayleigh's earlier work on a

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one-dimensional atomic chain, Debye tried to construct a mathematical model for the vibrations of three-dimensional lattices, in collaboration with the mathematician Haar. The problem proved very complicated and so Debye made the ansatz that as a result of the coupling of the motion of adjacent atoms, the vibrations are propagated as waves. For long wavelengths, these waves of atomic motion are the same as the acoustic waves that could be set up by externally coupling the crystal to an ultrasonic source. Debye further asserted that the waves of short wavelength were also propagated in the same fashion as the acoustic waves, i.e., were characterised by the same velocity as sound. The crystalline lattice was thus swept away and its properties replaced by that of an elastic continuum. This resulted in a continuous frequency spectrum of the famous ν^2 type. The different elastic type vibrational waves were now regarded by Debye as independent and as equivalent to oscillators. The enumeration of their energy levels was then done according to quantum theory à la Einstein, and Debye was finally able to obtain the well-known specific heat formula which was in better agreement with experiment than that of Einstein.

About the same time when Debye was trying to improve Einstein's frequency spectrum for a solid, Born and von Kármán³ were also independently tackling the problem of vibrations of a crystal lattice. They started with a one-dimensional chain of atoms and later generalised the results to three-dimensional lattices. According to these authors, the normal modes of vibrations of an infinite crystal can be described in terms of travelling waves. In this respect there is a certain amount of commonality with the Debye theory. Where the Born

theory differs is in that while Debye analysed these waves by assuming the solid to be an elastic continuum, Born and von Kármán took explicit account of the lattice structure. One important feature of the Born-von Kármán theory is that it applies to an infinite crystal. In the formal manipulations the infinite crystal is replaced by a finite crystal which is subjected to special boundary conditions called cyclic conditions. These are similar to the box normalization procedures that are employed for momentum eigenfunctions in quantum mechanics. In reality, of course, one does not deal with infinite crystals or cyclic crystals, but rather with finite crystals often with arbitrary surfaces. However, Born argued that for many purposes, for example, the calculation of the frequency spectrum and thence the specific heat, the finite crystal could be visualized as a cyclic crystal.

Raman appears to have become actively interested in the vibrations of crystals in the early forties. Critically evaluating the preceding theories, he was convinced that all of them were deficient. He particularly rejected the notion that the normal modes could be classified in terms of travelling waves, and on this score objected to the theories of Debye and Born. A further objection he raised in regard to the Debye theory was that short wavelength waves could not have the same character as elastic waves, which is a very valid objection. With regard to Born's theory, he disputed particularly the cyclic boundary conditions which in fact were the source of travelling-wave solutions for the normal modes. He expressed himself strongly against the theories of Debye and Born. For instance, at one place he remarks, "One of the basic objections to their method of approaching the specific heat problem is that since wave motions in-

volve progressive changes of phase along the direction of propagation and may have any frequency assigned to them, they can neither be treated as normal modes nor enumerated. The theories of Debye and Born seek to escape this difficulty by postulating that the number of wave motions is identical with the number of degrees of freedom of the system, while the choice of wavelengths is determined by still another postulate, e.g., the so-called postulate of the cyclic lattice which is claimed to represent the effect of the external boundary of the crystal. Since it is obviously impossible to formulate any boundary conditions at the surface of a crystal, the procedure is clearly artificial"⁴. He adds in another place, "It follows that these theories (*viz.*, of Born and Debye) are fundamentally misconceived and must therefore fail together with all their consequences"⁴.

While rejecting the theories of Born and Debye, Raman found himself attracted to the Einstein theory, particularly its singular frequency distribution. He argued that the frequency spectrum of a solid was not continuous but discrete, with, however, more than the single frequency permitted by Einstein. To quote his own words, "Einstein's approach to the problem is fundamentally correct.... His view of a crystal as an assembly of immense numbers of quantized oscillators having a common set of vibration frequencies is not only the logical and correct view of the matter but also proves itself when fully developed to be an eminently successful view. It gives a deep and quantitative insight into the thermal behaviour of solids"⁵.

To deduce the nature of the frequency spectrum it is first necessary to describe the normal modes and here Raman proceeded as follows⁶. He asserted that in

a normal mode equivalent atoms in adjacent cells have the same amplitude and must either vibrate in phase or out of phase. Next, to enumerate these modes, he considered a part of the crystal called the super cell which is obtained by translating the unit cell along its three principal edges and subsequently completing the parallelepiped. The super cell thus has eight times the volume of the unit cell. If p be the number of atoms in the unit cell then clearly the super cell will have $8p$ atoms and therefore $24p$ degrees of freedom associated with it. Following the analogy of molecules, Raman argued that three out of the $24p$ degrees of freedom are associated with translations of the super cell as a whole. The remaining $(24p - 3)$ degrees were associated with the normal modes of vibration which are such that equivalent atoms in adjacent cells are either in phase or out of phase. Thus, according to Raman, the frequency spectrum of a solid must essentially be discrete as in Einstein's theory, and must consist of $(24p - 3)$ frequencies. In addition, there is a ν^2 -type spectrum in the low-frequency end associated with the centre of mass motions of the super cells in the crystal. Now not all the $(24p - 3)$ frequencies are distinct. Crystal symmetry can and does introduce degeneracies. Thus in diamond there are only 9 distinct frequencies, and Raman showed these are associated with the vibrations of the octahedral and cubic planes.

It must be pointed out that both in the Raman theory and in the Born theory, there are two aspects. The first is the description and enumeration of the normal modes and the second is the actual numerical calculation of normal mode frequencies. The former problem is essentially one of formalism while the

latter depends on the details of the inter-atomic forces. Here we are mainly concerned with the former.

In seeking to answer the question as to which of these theories is correct, one would naturally turn to experiments. Indeed Raman himself took a step in this direction and initiated several investigations in the scattering of light by crystals. Figure 1 shows some typical Raman spectra of diamond—one of Raman's favourite subjects—obtained here at Bangalore by Raman and his associates⁷. The characteristic feature is the presence of sharp lines in the scattered spectrum. Raman was successfully able to account for the observed features by starting with a set of 9 basic frequencies and interpreting the observed lines as due to fundamentals, overtones, etc., subject, of course, to selection rules.

Another technique which has often been employed for the study of vibration spectra is X-ray scattering. We are aware that X-ray scattering by perfect crystals, in which all atoms are stationary, leads to Bragg reflections. If, however, the atoms are harmonically vibrating, then owing to the periodic modulation of the lattice, one expects extra Bragg reflections rather akin to the ghosts produced by gratings with ruling defects. The additional feature of these extra Bragg reflections is that the frequency of the scattered wave is Doppler shifted owing to the fact that the reflecting planes of atoms are moving. By observing these extra Bragg reflections and by measuring their associated Doppler shifts, it should, in principle, be possible to get information about the vibrational spectra of solids. The extra scattering of X-rays consequent to the thermal agitation of solids has been investigated by several workers including Raman himself. The chief practical difficulty associated with

these experiments pertains to the measurement of the Doppler shift of the scattered rays. This is mainly an experimental difficulty and is connected with the fact that the shifts are of the order of 1 in 10^5 , a spectral resolution impossible to achieve in the X-ray region by currently known techniques.

In the fifties, thanks to the availability of intense neutron beams from high flux reactors, it became possible to achieve with neutrons what X-rays failed to do. Now thermal reactors produce copious supply of neutrons with wavelengths $\sim 1 \text{ \AA}$ (which is of the order of inter-atomic distances in solids). Neutron beams can therefore be diffracted from thermal agitations to produce extra Bragg reflections in exactly the same manner as X-rays, with the difference that the associated Doppler shift may be conveniently measured since the energy shifts suffered by the neutron upon scattering are often $\sim 10\text{--}100\%$ (unlike in the case of X-rays). This is the wave interpretation of the scattering of neutrons by lattice vibrations. In another sense, it can be viewed as the neutronic analogue of Raman scattering. The incident neutron exchanges energy and momentum with the quanta of vibrations of the lattice and is then scattered. As in all radiation scattering experiments, the scattering is subject to conservation of energy and momentum.

Figure 2 shows a schematic diagram of a typical neutron spectrometer. It consists essentially of two crystal spectrometers, one to produce a well-defined beam of monochromatic neutrons, and another to energy analyse the scattered beam. At Trombay we have several neutron spectrometers for carrying out such investigations. They are all indigenously built, and fully automated to ensure round-the-clock operation, a necessary

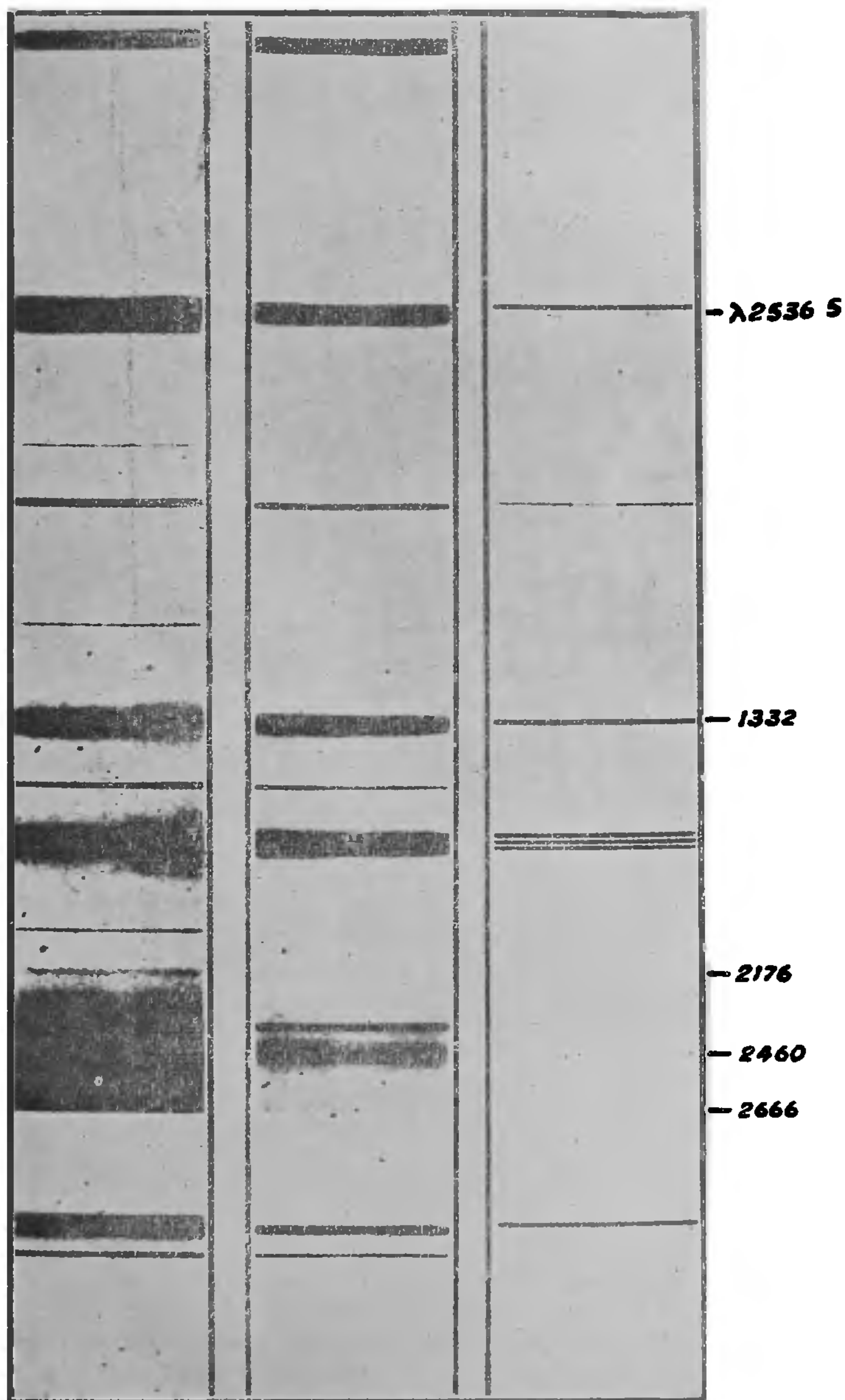


FIG. 1. Raman spectrum of diamond (After Raman reference 7),

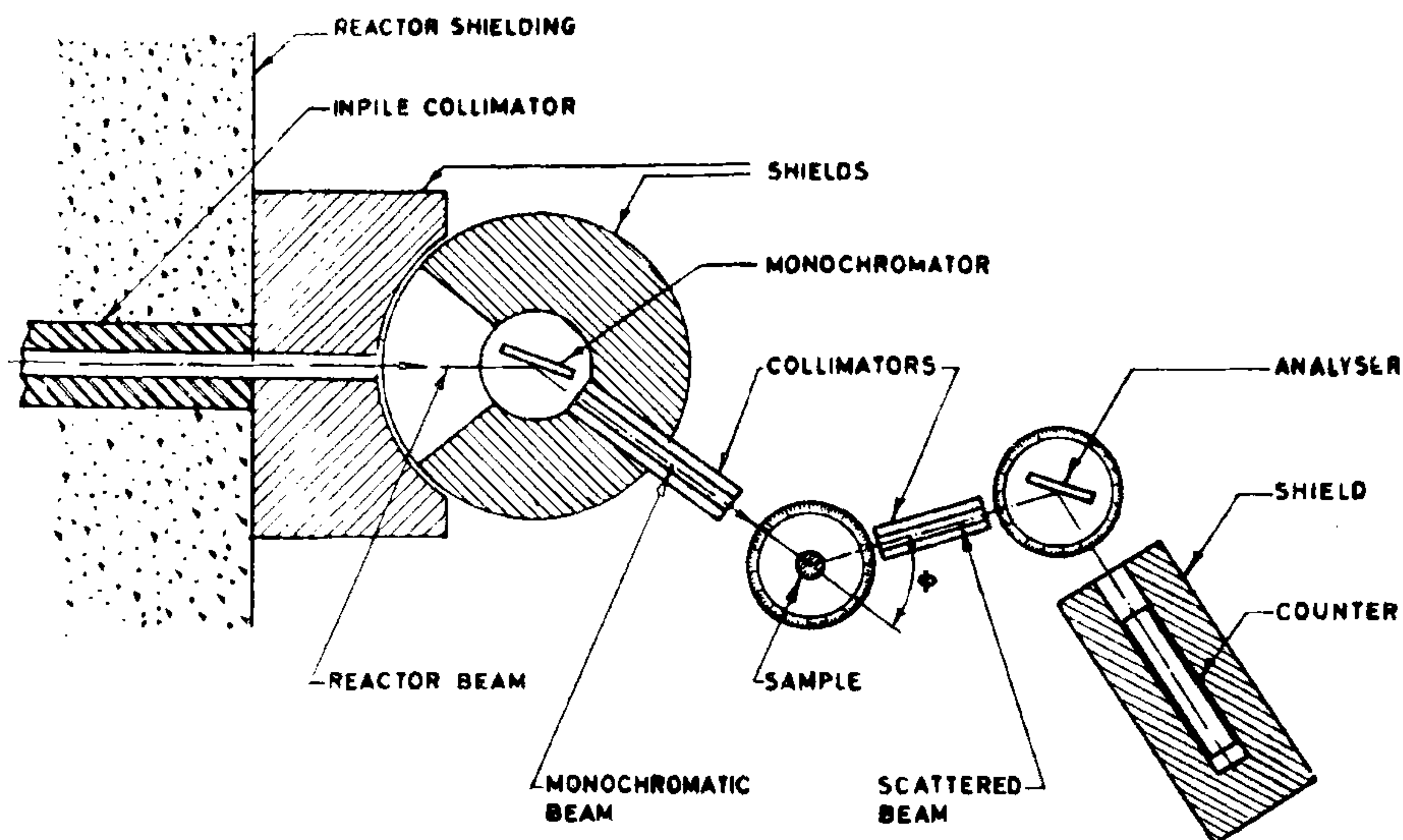


FIG. 2. Schematic drawing of a typical neutron spectrometer.

requirement since data collection is a slow and time-consuming process. Figure 3 shows typical phonon-induced Bragg peaks observed using the Trombay instruments. Figures 4, 5 and 6 show the dispersion curves of some of the substances investigated at Trombay^{8,9,10}. Similar plots for a number of other crystals have been obtained in laboratories elsewhere, and to date dispersion data is available for over 100 different crystals of various types ranging from insulators to metals, and from rare-gas solids to complex polymers. It would have been noticed in Figs. 4, 5 and 6 that curves are shown based on theoretical calculations made according to the Born theory. The good agreement between theory and experiment coupled with the fact that the spectrum of frequencies is not discrete as Raman propounded immediately raises two questions: (i) Does it mean that Born's theory is correct and Raman's theory is wrong? (ii) If so then

how does one explain the line spectrum obtained by Raman and coworkers in the light scattering experiments?

The short answer to the first question is that actually none of the theories is strictly correct but Born's theory comes closest to describing the actual situation, subject to some limitations to be discussed later. For the present let us assume the correctness of Born's theory and try to understand its relationship to that of Raman since both are successful in their own way in explaining certain experimental results.

To begin, let us consider the frequency spectrum of a simple solid like argon which has the fcc structure. Figure 7 shows the Brillouin Zone (BZ) which is a sort of symmetric unit cell of the reciprocal space associated with the fcc lattice. One could also view the BZ as a cell in (crystal) momentum space. According to Raman, the $(24p - 3)$ frequencies reduce to 4 distinct frequencies

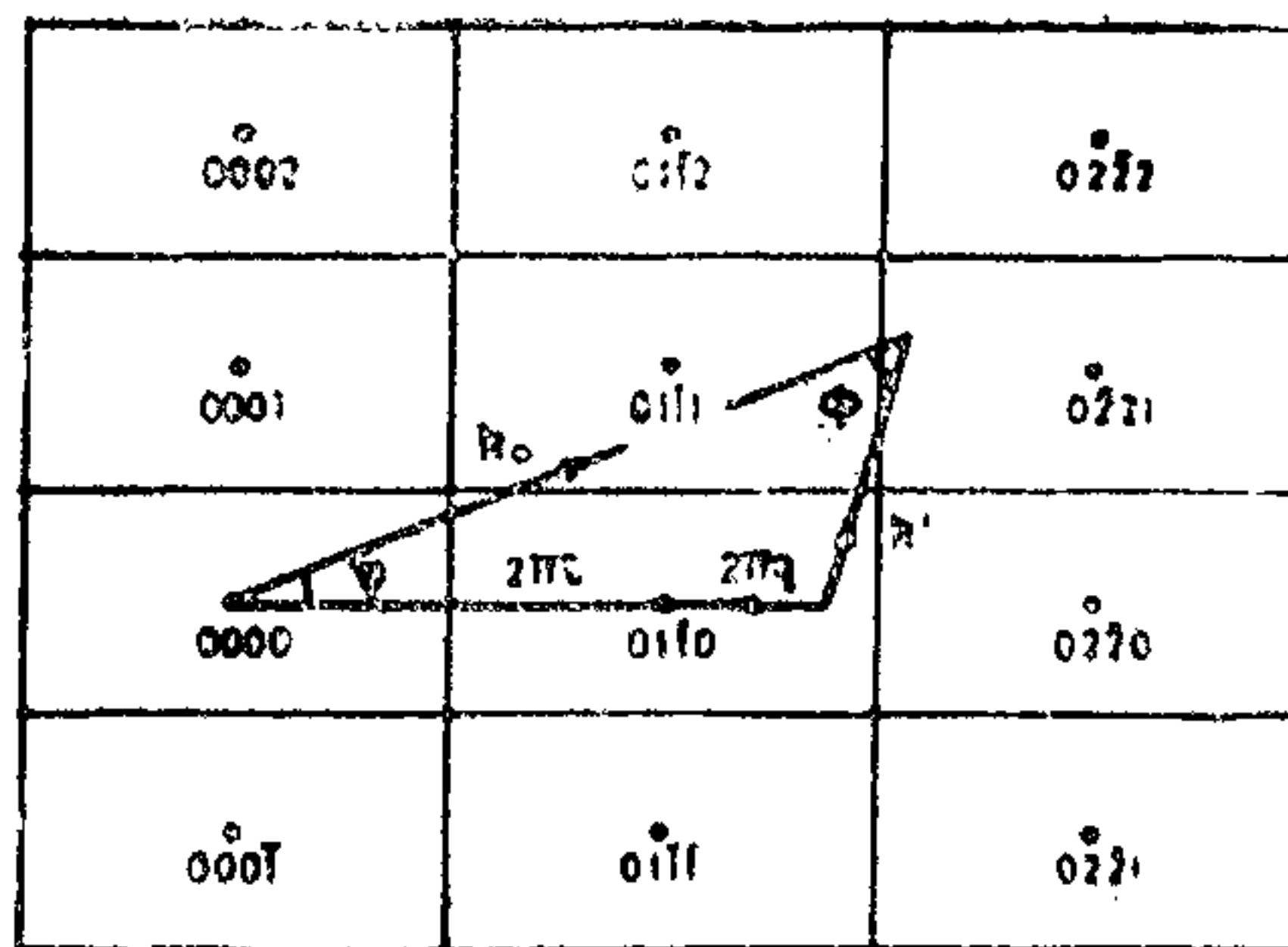
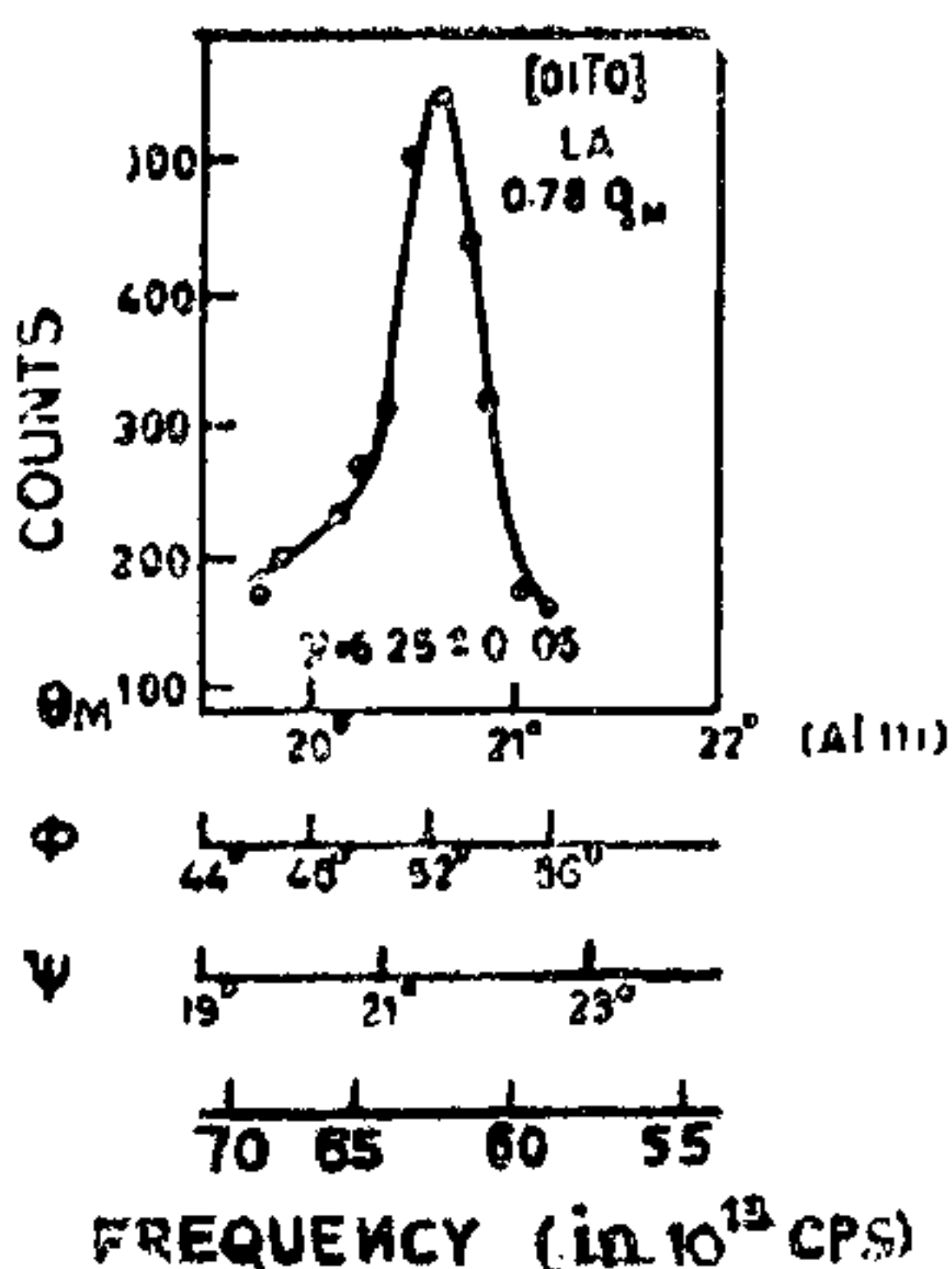
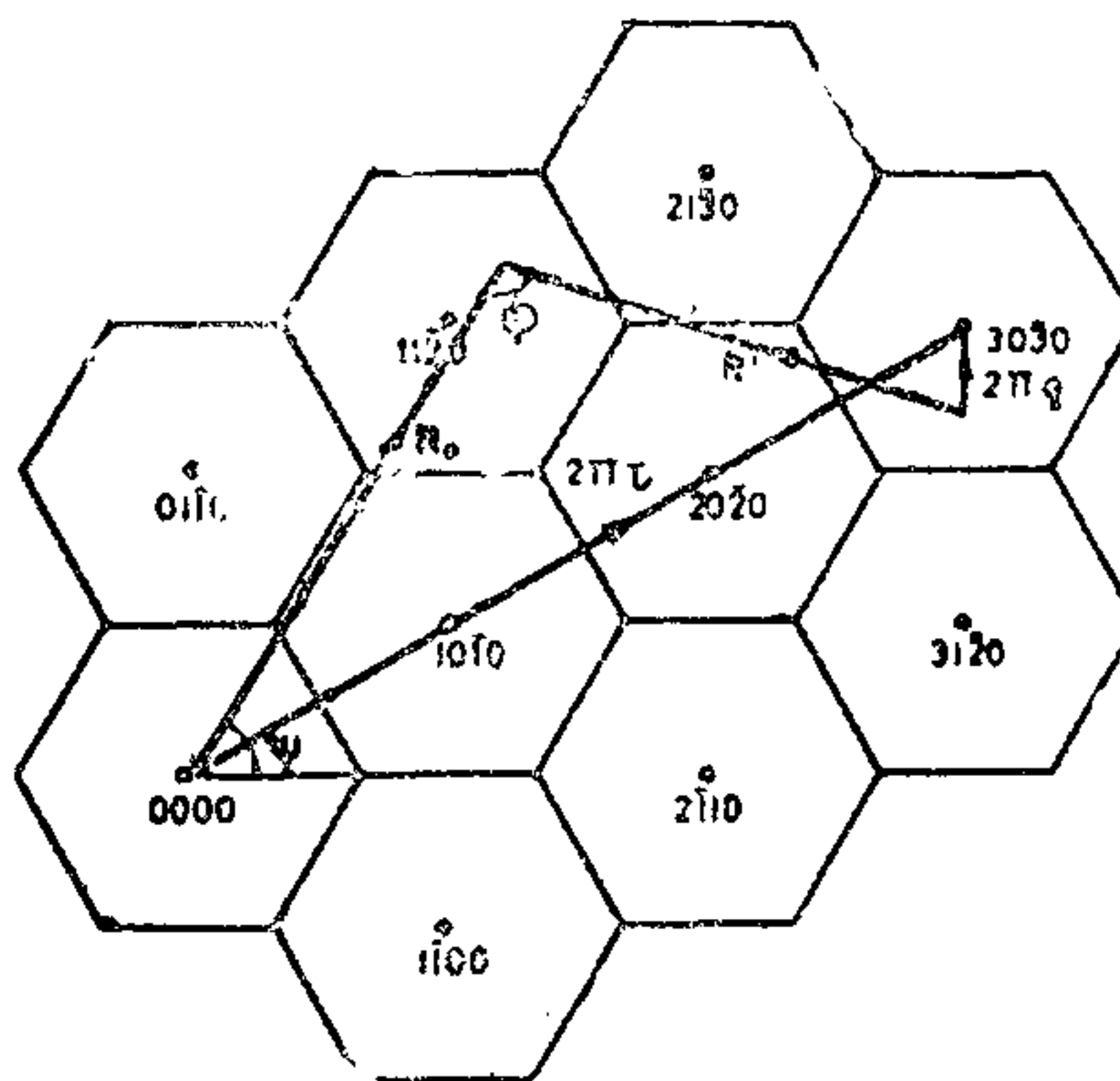
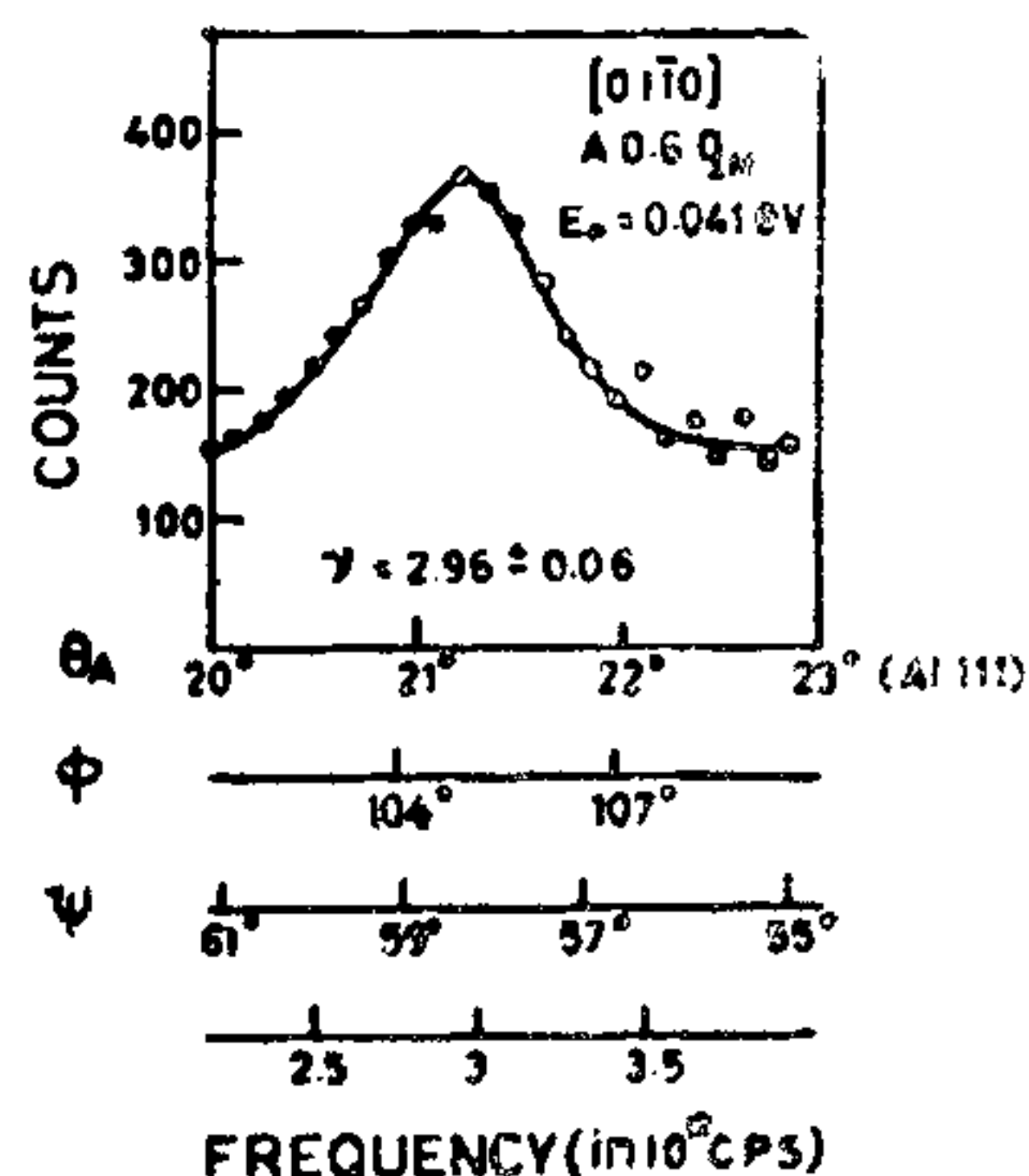
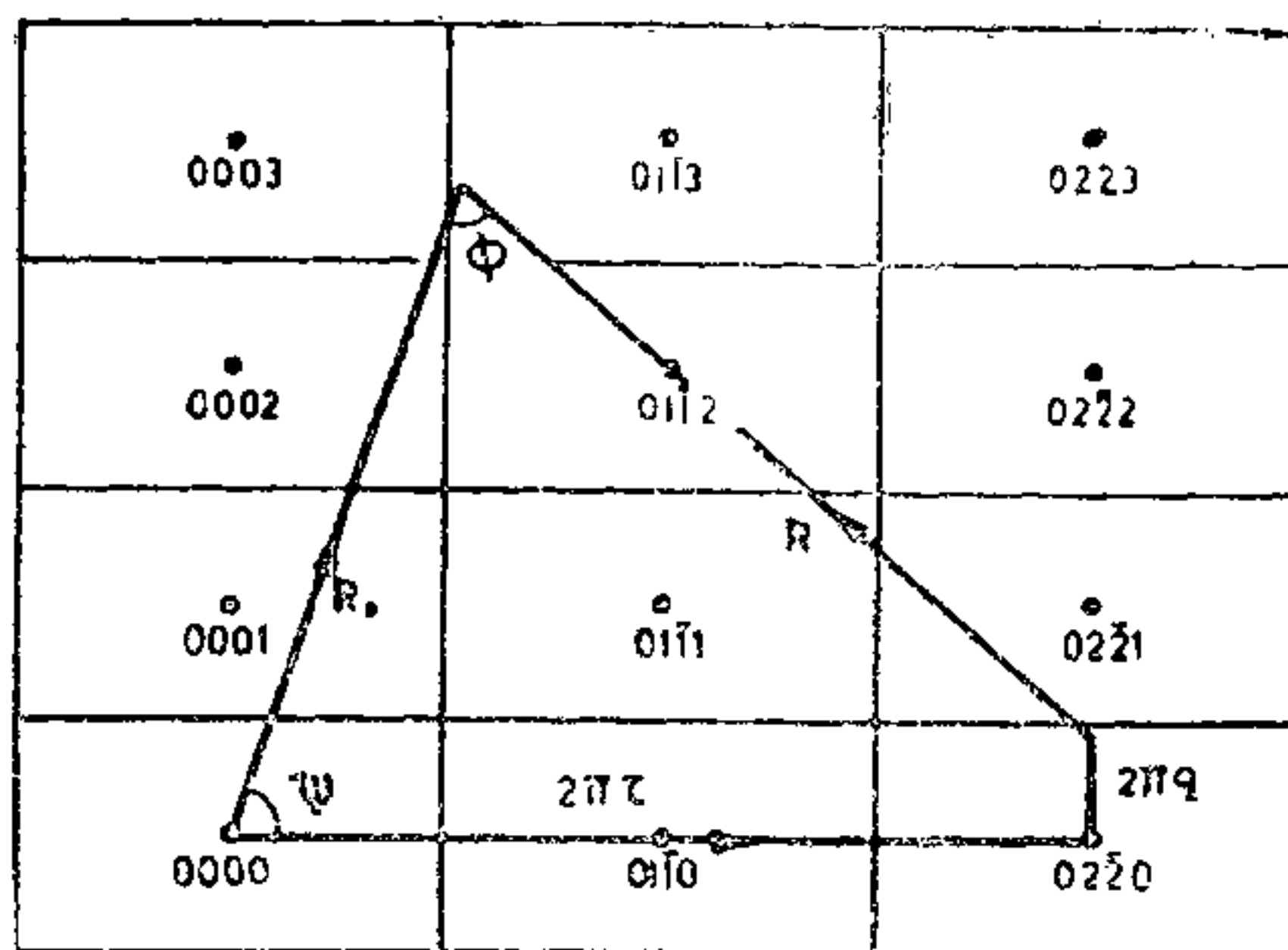
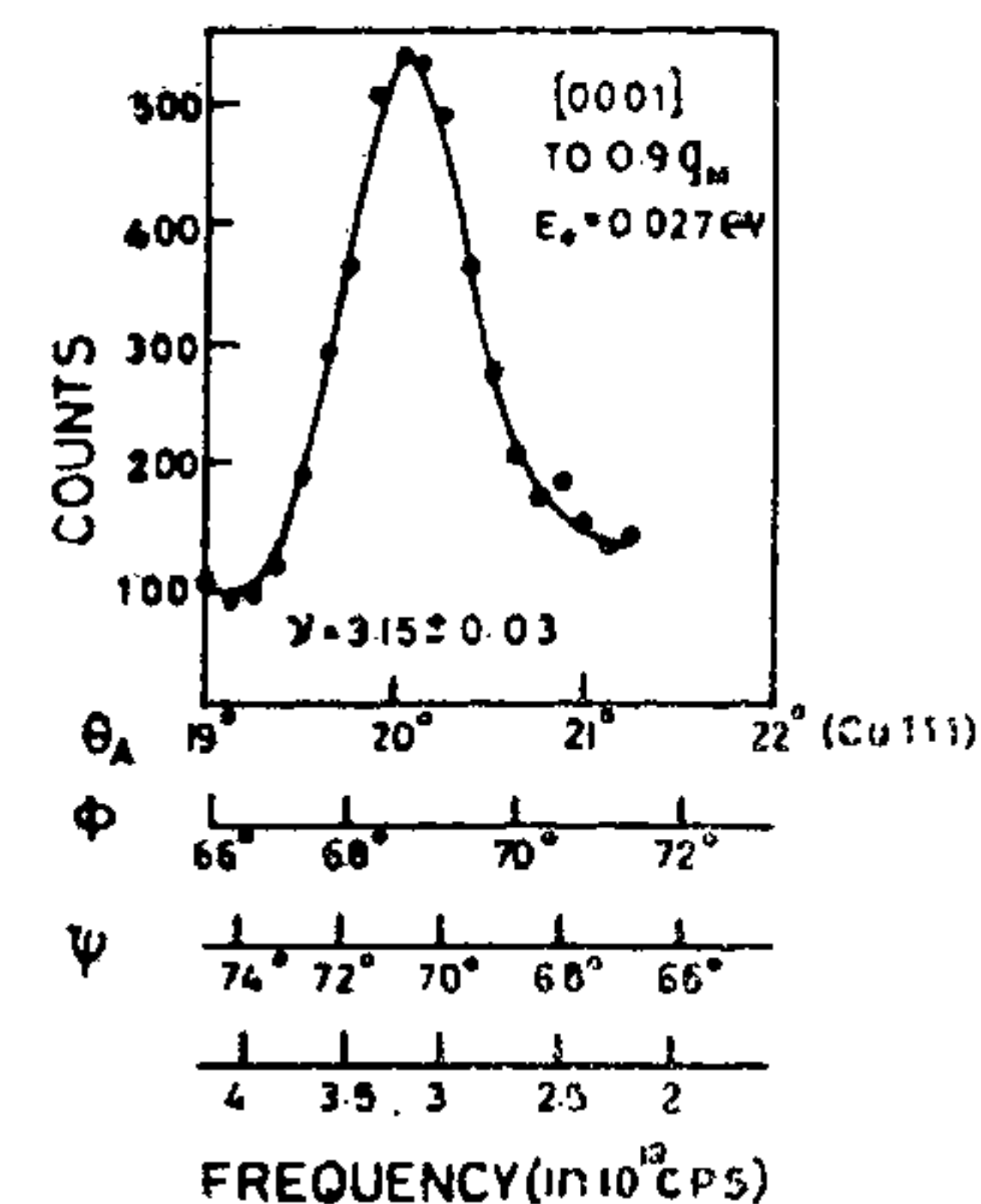


FIG. 3. Typical "Bragg peaks" due to "reflection" of neutrons by vibrational modulations of the lattice. The data pertains to Mg, and on the right are shown the Ewald constructions appropriate to these extra Bragg reflections (After Iyengar *et al.*, reference 8).

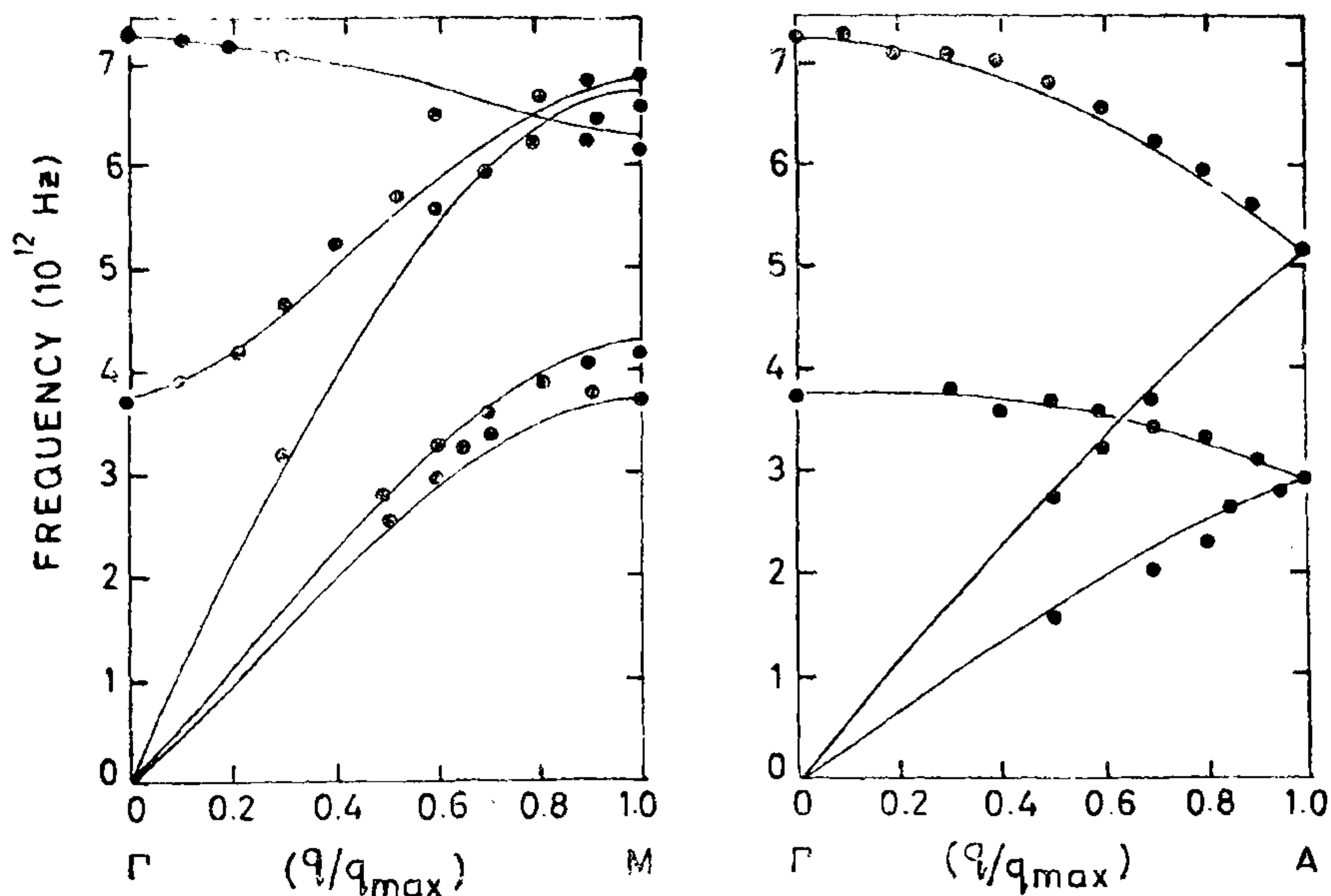


FIG. 4. Curves of frequency versus wavevector for normal modes in the hexagonal crystal Mg. Such curves are referred to as dispersion curves. ΓA refers to the (0001) direction and ΓM refers to the (0110) direction. The points are the experimentally measured data, and the solid lines are theoretically computed curves based on Born's model (After Iyengar *et al.*, reference 8).

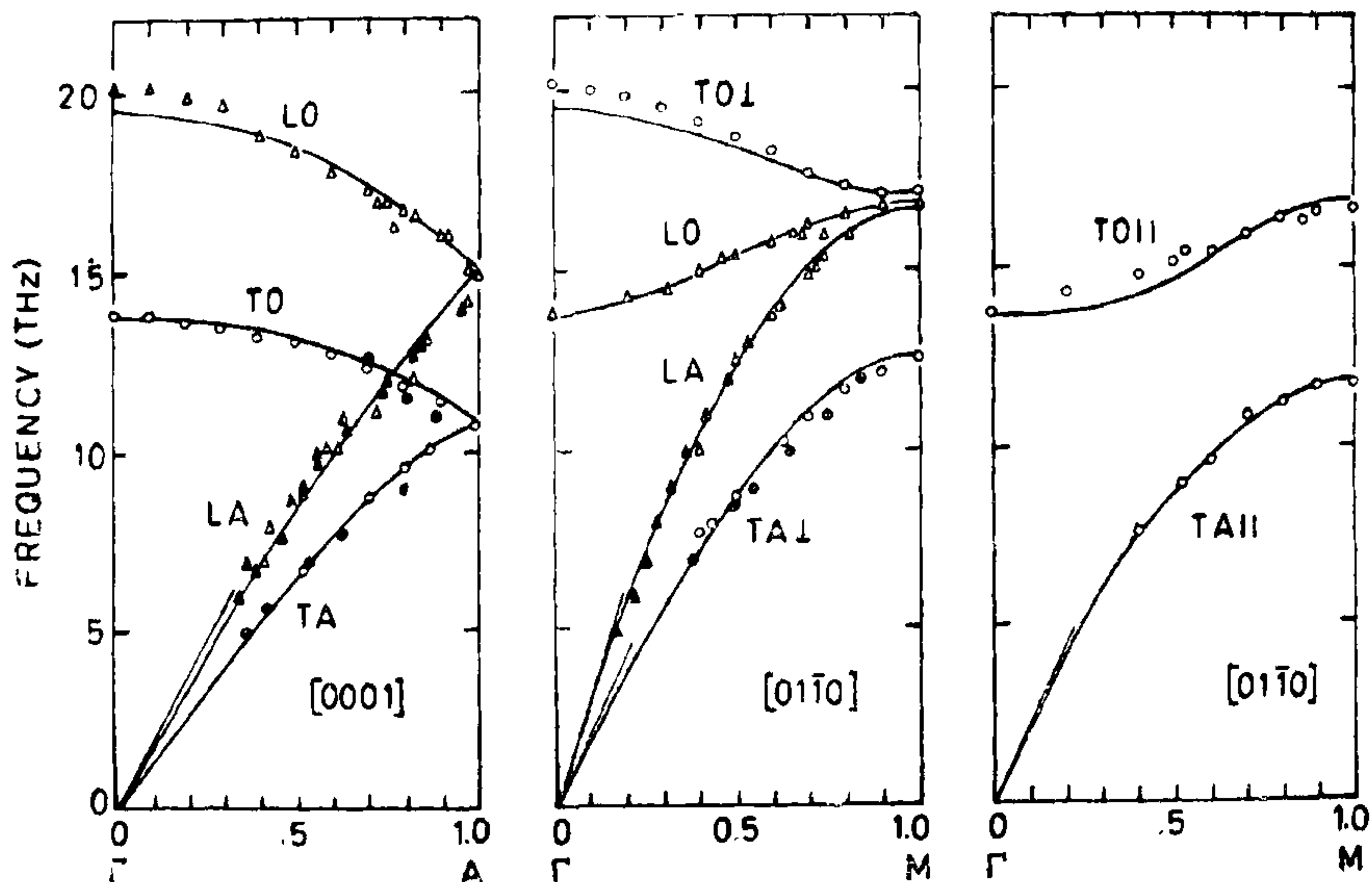


FIG. 5. Dispersion curves for Be (After Thaper *et al.*, reference 9).

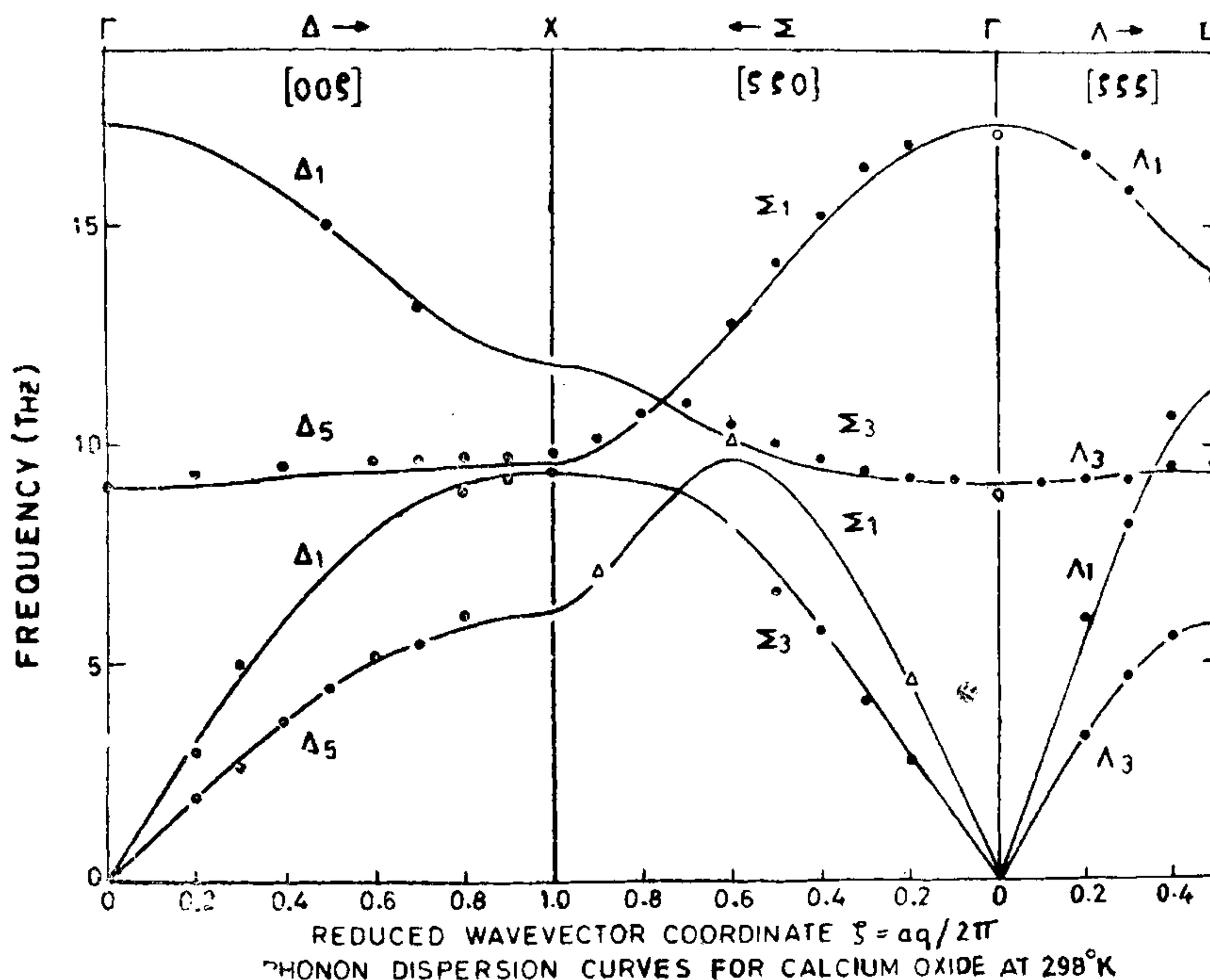


FIG. 6. Dispersion curves for CaO (After P. R. Vijayaraghavan, reference 10).

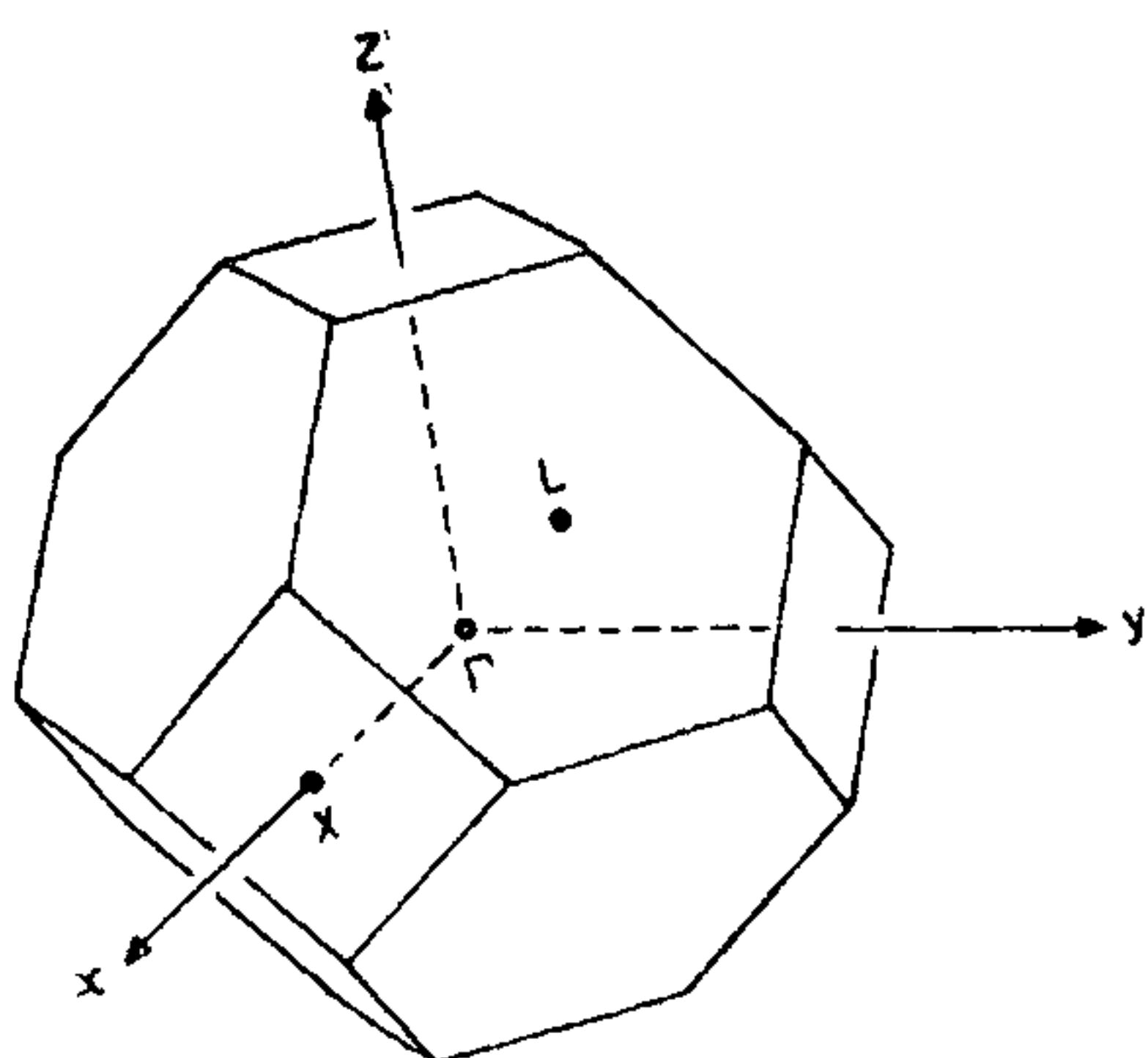


FIG. 7. Brillouin zone for a fcc lattice.

associated with the vibrations (longitudinal and transverse) of the octahedral planes and of the cubic planes. The group velocity associated with these vibrations would of course be zero. In

Born's theory where one visualizes the normal modes in terms of waves, the distinct wave vectors possible are distributed inside the BZ. Of these, waves associated with L and X would have vanishing group velocity and would in fact correspond to the vibrations of the type Raman visualized. In other words, out of the large number of wave-like normal modes permitted in the Born theory, Raman's theory focusses attention on a selected subset which are primarily determined by the translational symmetry of the lattice and are characterized by vanishing group velocity. Interestingly, this fact was recognized by Raman himself¹¹.

With these remarks let us now examine the frequency spectrum for

solid argon as determined by the two theories. Figure 8 shows this comparison.

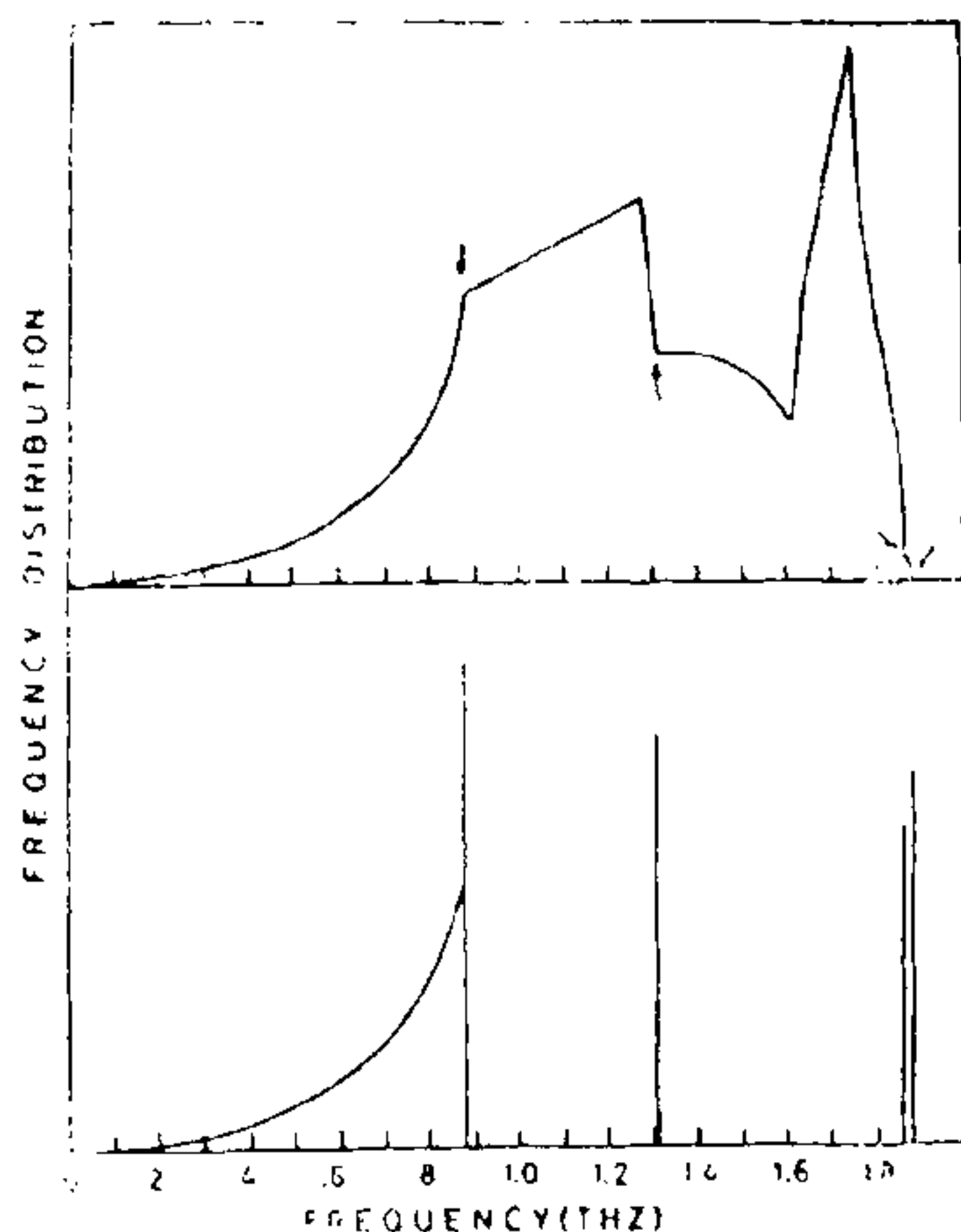


FIG. 8. Frequency spectrum of solid argon. In the top half is shown the spectrum calculated¹² according to Born's theory. The arrows indicate the critical frequencies associated with vanishing group velocity. In the bottom half is sketched the frequency spectrum expected in Raman's theory.

In the top half is presented the frequency spectrum for solid argon computed using Born's theory and using as input data force constants determined from the well-known Lennard-Jones potential for argon¹². (This frequency spectrum incidentally is in good agreement with that determined from neutron scattering data.) Particularly to be noticed in (a) are the kinks (shown by arrows) occurring at certain frequencies. These are known as van Hove singularities¹³ and are associated with the vanishing of group velocity for the associated modes. In the lower half of Fig. 8 is sketched the frequency spectrum as given by Raman's theory. This predicts a low frequency continuum with a ω^2 behaviour as in Born's theory, but in the high frequency region there are 4 discrete frequencies as mentioned earlier, which,

observe, coincide with the critical frequencies.

The fact that Raman intuitively focussed attention on the critical frequencies is rather interesting because these frequencies play an important role in many contexts including not only the familiar Raman and infrared spectra, but also superconducting tunnelling experiments. This aspect is illustrated in Fig. 9. Shown at the top are the dispersion curves for lead (a fcc lattice) as measured by neutron spectrometry¹⁴. A notable feature of these results is the somewhat unusual nature of the dispersion curves, including the presence of many modes of zero group velocity over and above those associated with the points X and L (which all fcc lattices share). These unusual features are related to the strong electron-phonon interactions in lead and are in fact responsible for its relatively high superconducting transition temperature. Figure 9b shows the frequency spectrum as computed using Born's theory, using as input parameters force constants deduced from a ¹⁵. The frequency spectrum so calculated exhibits several singularities especially as compared to argon. However, the spectrum is believed to be a poor one since the starting force constant model is not able to provide a good description of the effects of electron-phonon interactions as observed in the measured dispersion curves. Figure 9c shows the 'measured' frequency spectrum¹⁶. This was obtained by sampling the frequencies at a large number of points in the BZ using the technique of neutron spectrometry. Like Fig. 9b, this spectrum also shows considerable structure associated with the van Hove singularities. Figure 9d shows a quantity closely related to the frequency spectrum deduced from the tunnelling experiments¹⁷. The van Hove

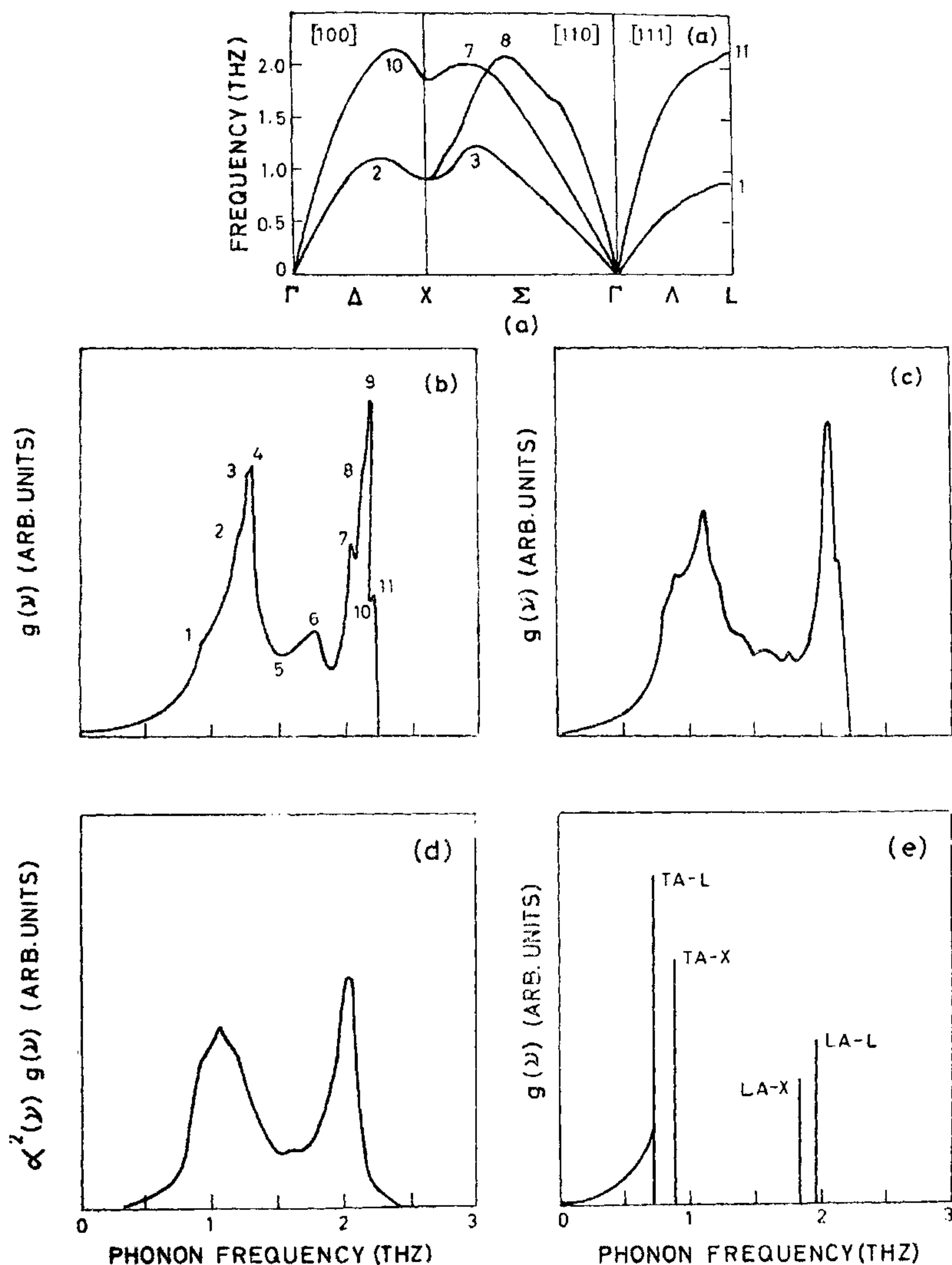


FIG. 9. (a) Dispersion curves for Pb as determined by neutron spectrometry¹⁴. (b) Frequency spectrum $g(\nu)$ computed on the Born model, with force constants deduced by fitting to data of (a)¹⁶. (c) Frequency spectrum directly determined by neutron spectrometry¹⁶. (d) Spectrum closely related to the frequency spectrum deduced from tunnelling experiments¹⁷. (e) Frequency spectrum expected in Raman's theory.

singularities deduced from the neutron scattering results are all visible in the spectrum obtained from the tunnelling experiment (— though this is not evident on the scale shown). Finally, Fig. 9e shows schematically the frequency spec-

trum as deduced from Raman's theory. The latter predicts only 4 distinct frequencies with vanishing group velocities, and these let us remember are determined by the symmetry of the structure. The fact that there are many more modes with zero group velocity is somewhat unusual and is related to the electron-phonon interaction strength as previously mentioned. These extra critical frequencies are not symmetry determined. It may be remarked that until about 10 years ago when Walter Kohn¹⁸ first drew attention to the relevance of the electron-phonon interaction to vibration spectra, there was no reason to expect the occurrence of such unusual features.

We now address ourselves to the second question, namely, "How can the line structure of the Raman spectrum be explained if the frequency spectrum is a continuum?" I shall seek to answer this question by considering two examples, namely, (i) ZnS and (ii) diamond. Figure 10 shows the structure of (cubic) ZnS. It is very similar to that of diamond except that the atoms

in the two sublattices are different. Figure 11 shows the phonon dispersion curves for ZnS obtained by neutron spectrometry by the University of Michigan group¹⁹. Also shown are critical frequencies of which there are 9 which are symmetry dictated and have vanishing group velocity. (This is also the number predicted by Raman's theory for this structure. One might actually count 10 frequencies in ZnS if one takes account of the LO-TO splitting at the

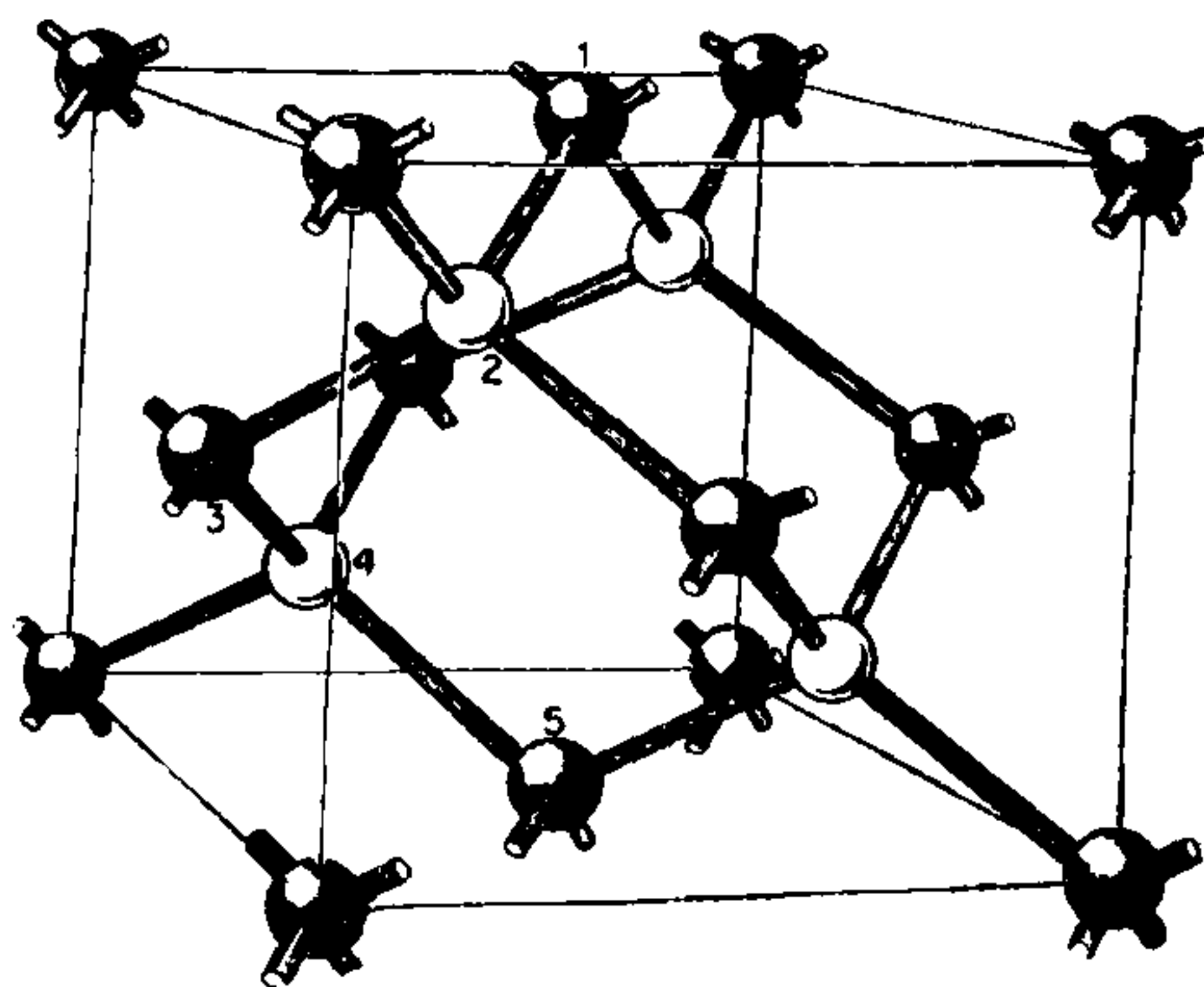


FIG. 10. Structure of zinc blende.

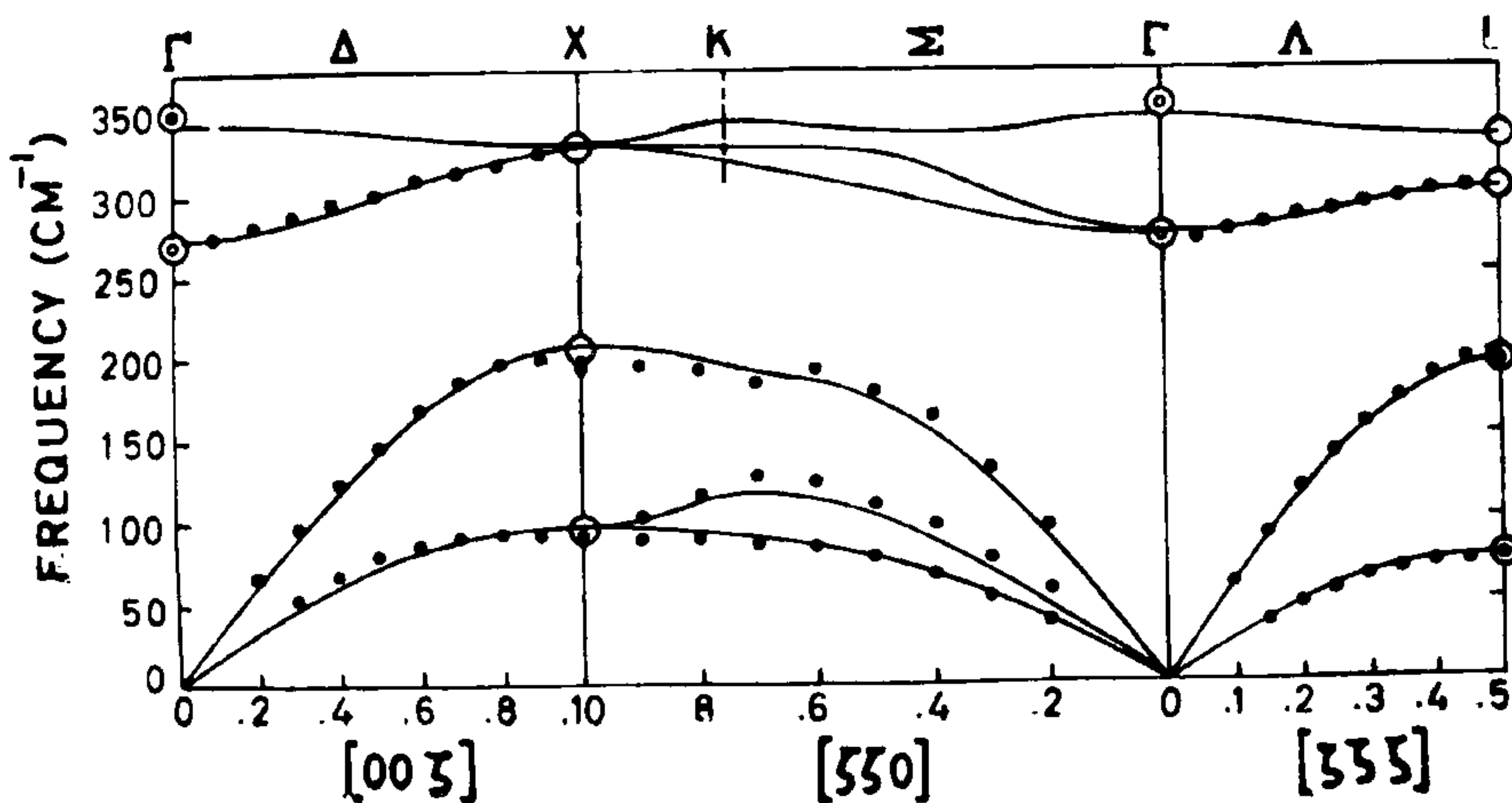


FIG. 11. Dispersion curves for ZnS (After Feldkamp *et al.*, reference 19).

zone centre brought about the macroscopic electric field. However if a careful study of the $q \rightarrow 0$ region is made taking into account retardation effects, then one finds only 9 critical frequencies in ZnS.) The Raman spectrum of ZnS is shown in Fig. 12. This was obtained at the Bell Telephone Laboratories by employing Laser Raman spectroscopy²⁰. The experiments have been performed under various conditions, and as in the early Bangalore experiments, one sees several lines. These could all in principle be explained in terms of the various critical frequencies (including the set of 9 mentioned above). This, of course, would be analogous to Raman's original explanation of the diamond spectrum. In practice such an interpretation needs care since ambiguities could arise due to close proximity of lines, etc.

Finally let us consider the case of diamond itself. Figure 13 shows the dispersion curves for diamond as measured by neutron spectrometry at Los Alamos²¹. There are 9 critical frequencies associated with the points Γ , X and L which are the frequencies mentioned by Raman also. Figure 14 shows a part of the Raman spectrum of diamond as obtained here; also shown is the associated intensity distribution⁶. The point worthy of note is that the spectrum is not entirely a line spectrum but rather a continuum with, however, strong singular features superposed. This aspect is better seen in a more recent Raman spectrum shown in Fig. 15 which was obtained by Solin and Ramdas²² using Laser Raman spectroscopy. Once again we observe there is a continuum superposed on which are the van Hove singularities associated with the combination spectrum. These include the singularities arising from the combination of the 9 critical frequencies associated with Γ , X and L, which are the

frequencies featuring in Raman's theory. Table I shows these 9 frequencies as measured by neutron spectrometry and

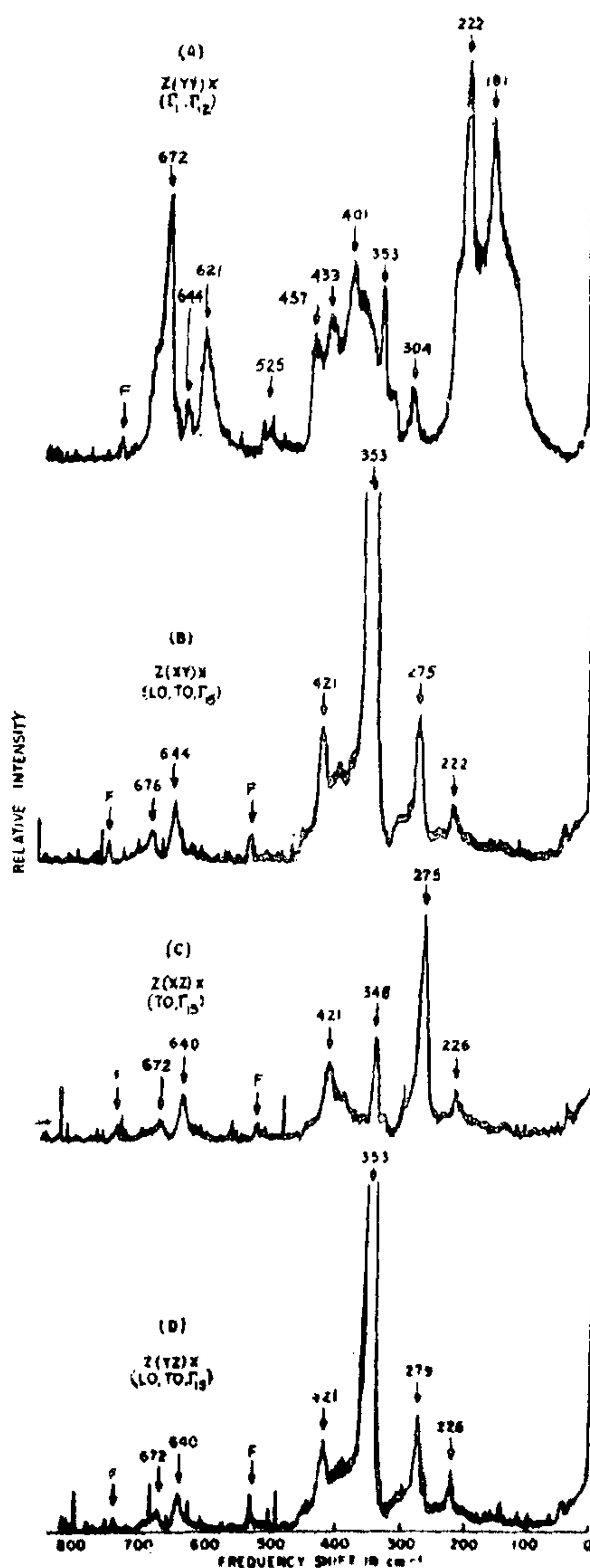


FIG. 12. Raman spectrum of ZnS (After Nilsen reference 20).

as deduced by Solin and Ramdas. (A similar set was also obtained by Raman himself but is not shown here. While Raman's values are in broad agreement

with those shown in Table I, the values of Solin and Ramdas have a higher accuracy obtained as they were with modern techniques.)

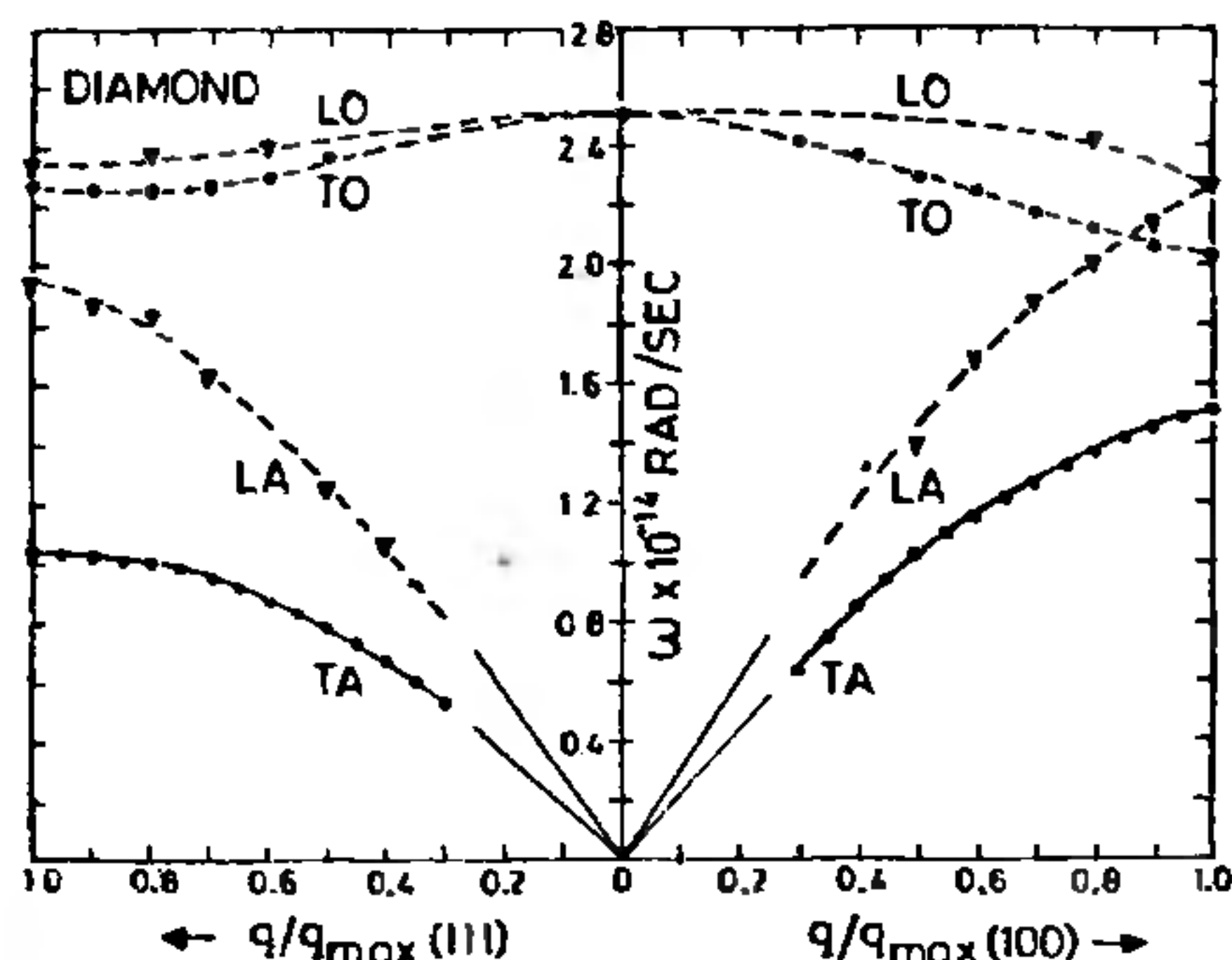


FIG. 13. Dispersion curves of diamond (After Warren *et al.*, reference 21).

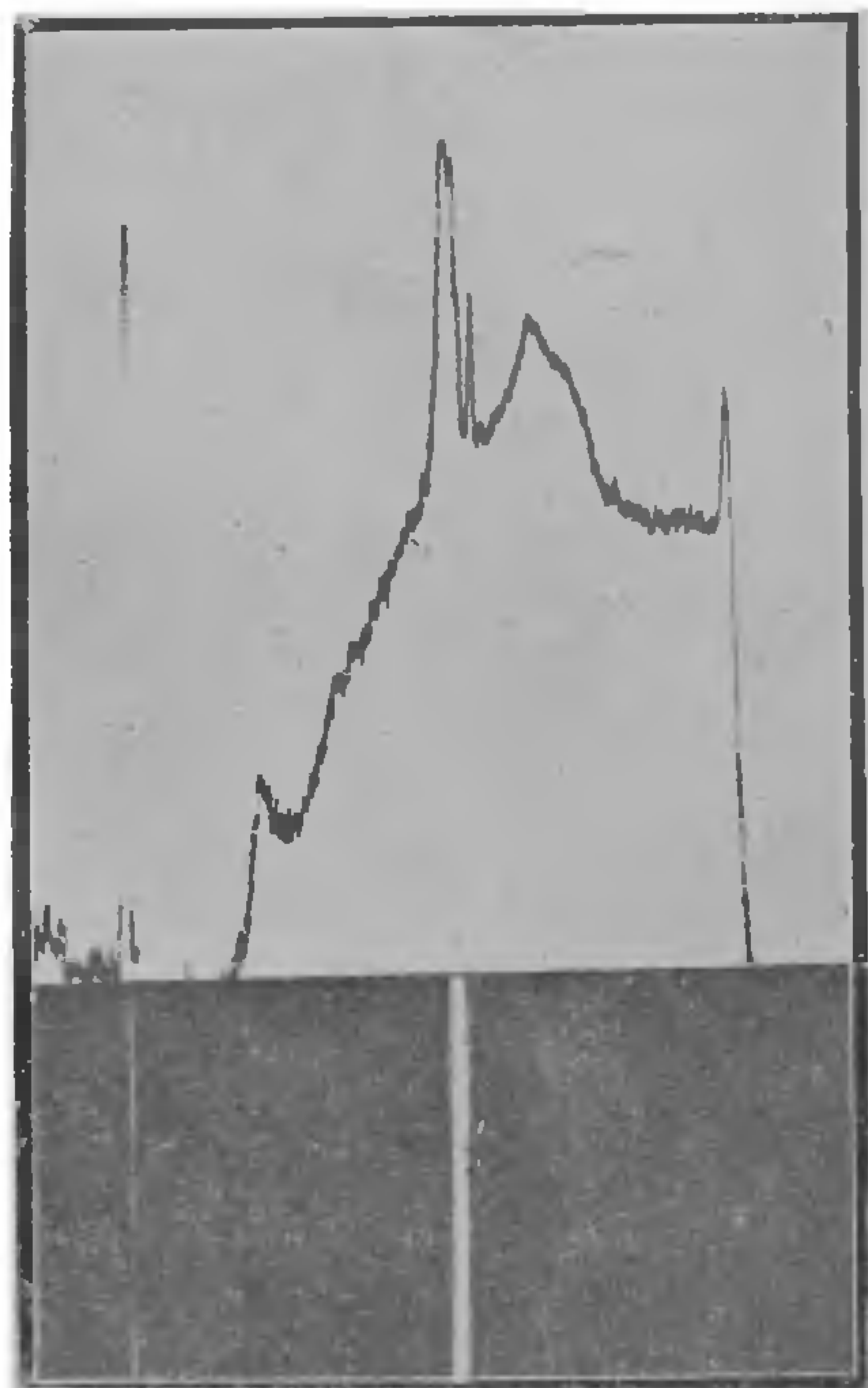


FIG. 14. Raman spectrum of diamond. (After Raman, reference 6).

TABLE I
Critical frequencies in diamond

Symmetry point	Phonon	Frequency (cm ⁻¹)	
		Neutron	Optical
Γ	O	..	1332±0.6
	TO	1072±26	1069±5
X	L	1184±21	1185±5
	TA	807±32	807±5
	TO	1210±37	1206±5
L	LO	1242±37	1252±5
	TA	552±16	563±5
	LA	1035±32	1006±5

The mystery of the sharp lines seen in earlier spectra is now clear. They were mainly the van Hove singularities of the combination spectra, showing up somewhat prominently above a continuous background, which is always present²³. Indeed a similar situation obtains with regard to second order infrared spectrum also.

Summarising, we see in retrospect that Raman with the intuitiveness of an experimenter tended to concentrate on the symmetry-determined critical frequencies associated with vanishing group

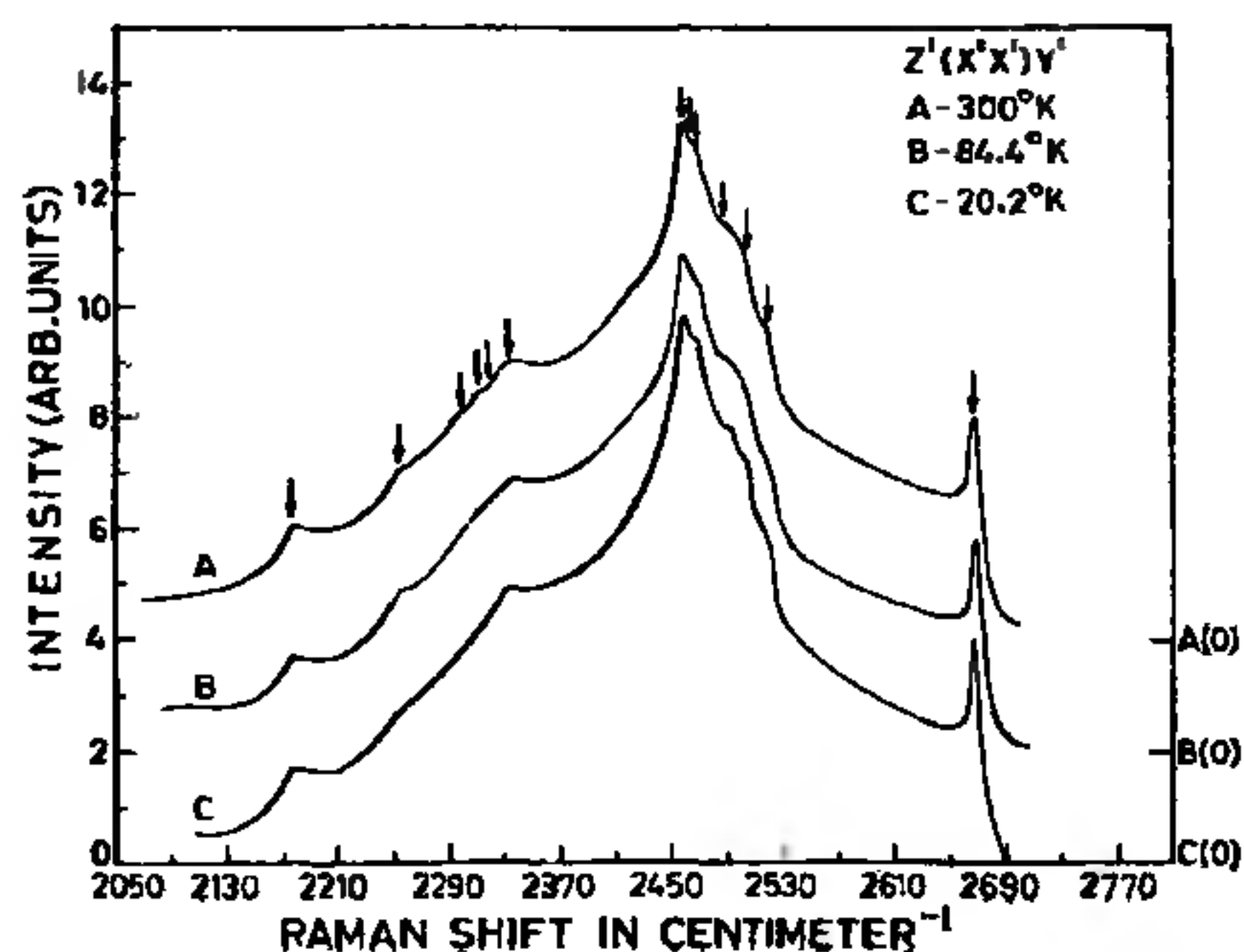


FIG. 15. Raman spectrum of diamond obtained by Laser Raman Spectroscopy. The arrows indicate some of the critical features (After Solin and Ramdas, reference 22).

velocity since these produced the most important features in the observed Raman spectra. These critical frequencies are important in several other contexts also, for example, in infrared absorption and in tunnelling experiments. Nevertheless the critical frequencies considered by Raman do not exhaust all the possible critical frequencies (in the modern sense of that term), and much less the frequency spectrum. That there are other frequencies is now clear from the neutron, tunnelling and indeed even the Raman scattering experiments. These other frequencies are accessible to calculation from Born's theory but not from Raman's theory. Even so, the doubts that Raman expressed concerning Born's method of enumerating the normal modes, and in particular concerning the application of the so-called cyclic boundary conditions have some historical significance. These objections coming as they did from a person of Raman's eminence could not be lightly brushed aside. The question was carefully examined by Lederman²⁴ and by Peirels²⁵ and it was concluded that while indeed the normal modes in a finite and in an infinite (or cyclic crystal) are not identical, the frequency spectra of the two are practically identical if the ratio of the number of atoms on the surface to those in the bulk is small. Thus except in special situations where surface effects are likely to be important, one could safely employ the simpler (but perhaps somewhat artificial) picture of a cyclic crystal for describing the normal modes and thereby enumerating the frequency spectrum.

It must be emphasized at this point that notwithstanding the progress made to date, our understanding of atomic motions in solids is far from complete. Undoubtedly the works of Einstein,

Debye, Born and Raman represent important landmarks. Einstein called attention to the importance of using a quantum description for the statistical mechanics of the oscillators. Debye pointed out that the vibrations of various atoms are coupled and that the waves (at least the low frequency ones) have an acoustic character and therefore lead to a ν^2 -type of frequency spectrum. Born showed the way to discussing the dynamics with due regard to the lattice structure while Raman focussed attention (particularly in the context of Raman spectra) on the symmetry determined set of critical frequencies associated with vanishing group velocities. Of the various theories, that of Born offers the most comprehensive scheme of discussing vibration spectra. However, it still leaves many things unexplained. For instance, it is well known that Born's theory if applied to solid helium totally fails in that it predicts imaginary frequencies! This is closely connected with the peculiar quantum features of solid helium which necessitate totally new methods²⁶. At a more mundane level, the phenomenon of diffusion which is known to occur in solids simply does not come out as a natural consequence of any of these theories. So also the problem of molecular reorientation, rotational diffusion, etc., in molecular crystals. Thus, in spite of the contributions of the giants of the past, our understanding of the mysteries of atomic motions in the solid state is still far from complete. There are many more secrets to unravel, and in the words of the poet Robert Frost,

"We dance around in a ring and suppose
But the secret sits in the middle and
knows."

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OPTICAL SPECTROSCOPY AND DEVELOPMENT OF QUANTUM ELECTRONICS

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1. INTRODUCTION

WITHIN the limited time at our disposal, it will be impossible to do justice in talking about even a small part of the research work done by Professor C. V. Raman, who was the greatest experimental physicist India has produced so far. In spite of this constraint we will make an effort to discuss the impact of some of the pioneering work in optics done by him and his students on the modern developments in Quantum Electronics.

Raman worked in many fields in physics including acoustics, magnetism and X-rays; but the study of optical properties of various materials fascinated him most. In fact, he was so much enchanted by "light" and "colour" from the very beginning (Raman, 1915) that he spent the later part of his research

career in a massive and detailed experimental investigation of diverse aspects of the physiology of vision (Raman, 1968). As a young man when Raman started doing physics, optical spectroscopy was one of the most powerful fields of investigation. It was rapidly producing new and unexpected results, and thereby introducing fundamental changes in the understanding of physics. It was, therefore, no accident that Raman decided to enrich this exciting field with all his vigour. It is perhaps significant to add that he brought with him to optics his earlier experience of working on the propagation of sound waves in matter. His original contributions to spectroscopy between the year 1917, when he became the Palit Professor of Physics in the Calcutta University, and the year 1948, when he retired from the Indian Institute of Science in Bangalore, were phenomenal by any standard.