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FOCUSING CONDITIONS IN NEUTRON INELASTIC SCATTERING EXPERIMENTS WITH THE IPEN TRIPLE-AXIS SPECTROMETER*

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ABSTRACT

The study of dynamic behaviour of the crystalline structure is one of the most important applications of neutron inelastic scattering technique. As part of the neutron inelastic scattering program of IPEN Nuclear Physics Division, a triple axis crystal neutron spectrometer was built, and, in order to verify operational conditions and performance of the instrument, measurements of the known dispersion relations of copper ~~were~~ performed. In connection with these recent improvements, ~~the present work~~ deals with focusing conditions in coherent neutron inelastic process in which a single quantum of vibrational energy (phonon) is exchanged between the neutron and the crystal. For the instrument focusing conditions attainment, mathematical expressions were deduced taking into account the finite collimation, incident and scattered wave vectors, and interplanar spacing of the monochromator and the analyser crystals. The focusing conditions were graphically represented in order to have a practical and adequate routine procedure for the focusing of the spectrometer. An experimental test was made by measuring acoustic phonon peaks propagating in the $[00\xi]$ symmetry direction of a copper crystal. The measurements were performed in focusing conditions such as: optimum, good and defocused; the agreement between experimental results and theoretical predictions shows the usefulness of the adopted graphical method.

I – INTRODUCTION

The observation of the scattering of radiation from a solid is the most powerful technique for studies of the behaviour of certain basic excitations such as phonons, excitons, magnons etc., which are related to many properties of solids. In general, one attempts to measure the changes in energy and momentum suffered by the radiation as it passes through the solid and interacts in various ways with the basic excitations[1].

The present work is concerning with the use of thermal neutrons emerging from a nuclear reactor as probe particles to study in particular phonons in a solid, which are the quanta associated with the vibrations of the atoms in a crystal lattice.

The neutron technique is based on the fact that energies and momenta associated with thermal neutrons are of the same order of those associated with thermal motions of atoms in condensed matter. The dynamical states of the system in study can be easily observed from the neutron scattering process by a scatterer system. The energy and momentum transfers from the neutron to the system, or vice-versa, lead directly to the knowledge of the corresponding quantities associated with the basic excitations, such as phonons in a crystal.

The study of dynamic behaviour of the crystalline structures is one of the most important applications of neutron inelastic scattering technique. In a crystalline solid the field of force present is of periodic nature. Thus, for a given momentum transfer, it is possible only to occur a very limited number of values for energy transfer. On the other hand, from the knowledge of the crystalline sample

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orientation with respect to the scattering vector and from the attainment of experimental data corresponding to momentum and energy transfers – which are associated with the "neutron groups", a significant intensity of scattered neutrons – it is possible to obtain directly the dispersion relations, between frequency and wave vector, for the normal modes of vibration of crystalline lattice. From the complementary analysis of neutron intensity data, at a given temperature, one can identify correctly the branch, polarization and population factor of phonons associated with a certain normal mode of crystal vibration. These important pieces of information can be used in the derivation of interatomic force constants, range of these forces within the crystal and other important dynamical properties of crystalline matter, such as lattice heat capacity, that depend upon the atomic vibrations.

The triple axis neutron spectrometer is the most convenient apparatus for experimental determination of dispersion relations for the lattice vibrations with wave vectors along the symmetry directions in a crystal; the spectrometer operation is based on the coherent inelastic scattering of thermal neutrons by a crystalline sample.

As part of the neutron inelastic scattering program of IPEN Nuclear Physics Division, a triple axis neutron spectrometer was built, details of which project, construction and main characteristics were object of previous paper[4]. Measurements of the known dispersion relations of copper were also performed[3], in order to verify operational conditions and performance of the instrument.

In connection with these recent improvements, the present work deals with focusing conditions in coherent neutron inelastic scattering process in which a single quantum of vibrational energy (phonon) is exchanged between the neutron and the crystal. No discusion of crystal spectrometer would be complete without some mention of its focusing properties and a description of why these properties arise, besides how they may be calculated and used to good advantage in actual experiments[2].

II – FOCUSING

A focusing technique to improve resolution in measurements on one phonon resonances in the inelastic scattering of neutrons from crystals, even approximate, is well worthwhile in almost any measurement. Focusing has been indispensable in the observation of various small effects, such as frequency widths of phonons in aluminum at 80K, frequency shifts between 80 and 300K in aluminum and lead, and Kohn anomalies in the phonon dispersion relations for aluminum and lead[6].

In measuring phonon energy-momentum relationships by the triple-axis spectrometer method the experimenter has to choose from a large number of possible spectrometer conditions. One of the factors that affects this choice is the focusing effects that arises from the linear terms in the expressions for finite instrumental resolution and the phonon dispersion relation. An experimental result can be used to confirm theoretical predictions that in certain cases the ratio of focused to defocused peak height can exceed 10. Even in less favourable cases, valuable gains can be obtained by correct focusing[5].

Following Peckham[5], the present paper is concerned to a graphical method that provides a rapid means of determining approximate conditions for focusing. In practice, it has been found that the graphical method is an adequate routine procedure for operating a triple-axis spectrometer.

A shortened presentation of the subject neutron inelastic scattering from a single crystal, provides an useful introduction to the formulas given later.

II.1 – Inelastic Scattering of Neutrons from a Single Crystal

The motion of atoms in a crystalline solid may be expressed in terms of a noninteracting set of normal modes which have the form plane waves. They are characterized by a wave vector $\vec{q}$, frequency ω, and polarization direction $\vec{\epsilon}$. The frequency is a periodic function of $\vec{q}$, with $\omega(\vec{q}) = \omega(\vec{q}+\vec{G})$ for all reciprocal lattice vectors $\vec{G}$ of the crystal.

For any polarization $\vec{\epsilon}$, the angular frequency ω of a normal mode of a crystal for a predetermined value of wave vector $\vec{q}$ may be obtained by means of neutron scattering experiments. The coherent scattering for one phonon creation is governed by an interference condition:

$$\vec{Q} = \vec{k}_o - \vec{k}_1 = \vec{G} - \vec{q} \tag{1}$$

and by conservation of energy

$$\frac{\hbar^2}{2m}(k_o^2 - k_1^2) = \hbar\,\omega(\vec{q}) \tag{2}$$

where $\hbar\omega$ is the energy of a phonon, $\vec{k}_o$ and $\vec{k}_1$ are the ingoing and scattered neutron wave vectors and m is the neutron mass.

The neutron scattering experiment may be done by allowing neutrons of a particular energy to be incident on the specimen and observing the energy of those scattered through a particular angle. Observation of these energies enables the frequency and wave vector of the scattering phonons to be determined. As mentioned, a powerful instrument for making such observations is the triple-axis spectrometer. A full description of the IPEN triple-axis spectrometer was made elsewhere[4]. A schematic diagram is shown in Figure 1.

The polyenergetic neutron beam from the reactor passes through the first collimator (C_1), is incident on a single crystal monochromator (X_1) and reflected through the second collimator (C_2) to give a beam of neutrons of particular energy as determined by the monochromator angle, $2\theta_M$, the plane spacing of the crystal and Bragg's law; this defines $\vec{k}_o$. The energy of the neutrons scattered from the specimen (S) through an angle ϕ is likewise determined by reflection from the analysing single crystal (X_2) through an angle $2\theta_A$ after passing through the third collimator (C_3), into a ^{3}He detector; thus defining $\vec{k}_1$.

The advantage of a triple-axis crystal spectrometer lies in the control which can be exercised in the experiment. This is illustrated in Figure 2, where a possible experiment is shown by means of the appropriate vector diagram in reciprocal space. It is seen from this diagram that the coordinates of the momentum transfer are

$$Q_x = -|\vec{k}_o| \sin\psi + |\vec{k}_1| \sin(\phi+\psi) \tag{3}$$

$$Q_y = |\vec{k}_o| \cos\psi - |\vec{k}_1| \cos(\phi+\psi) \tag{4}$$

while the energy transfer is given by equation (2).

Since the IPEN spectrometer has the angle $2\theta_M$ fixed it is possible to chose any three angles $2\theta_A$, ϕ and ψ to satisfy equations (1) - (4) for particular Q_x, Q_y and ω, and so to obtain the intensity at any point in $(\vec{Q},\omega)$ space.

One particularly useful mode of operation is the "constant Q" method in which the momentum transfer is held fixed while an energy distribution is obtained. A peak in the scattering results when the frequency transfer is equal to the frequency of the phonon dispersion relation for that momentum transfer.

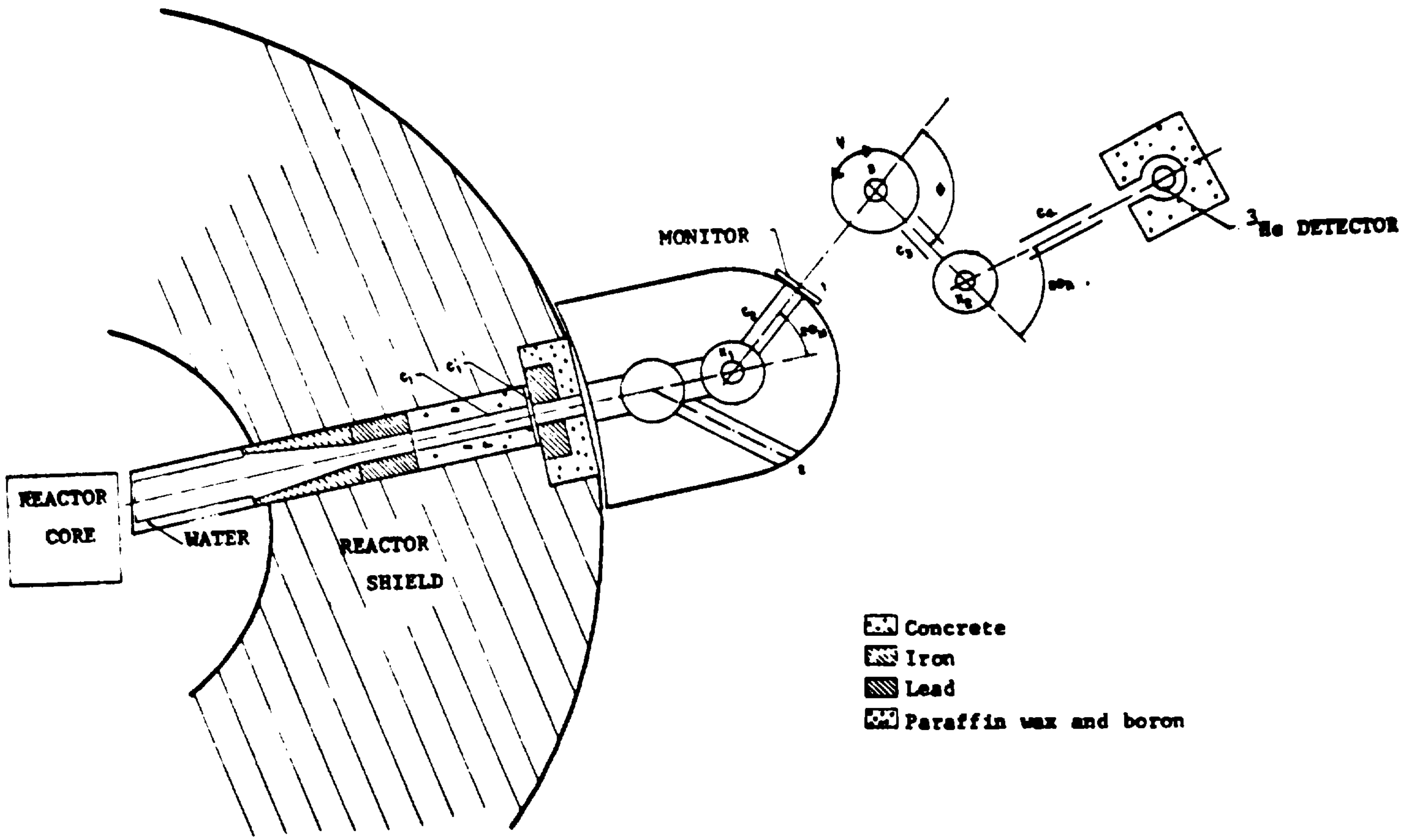

Figure 1 – Schematic diagram of the IPEN Triple-Axis Spectrometer

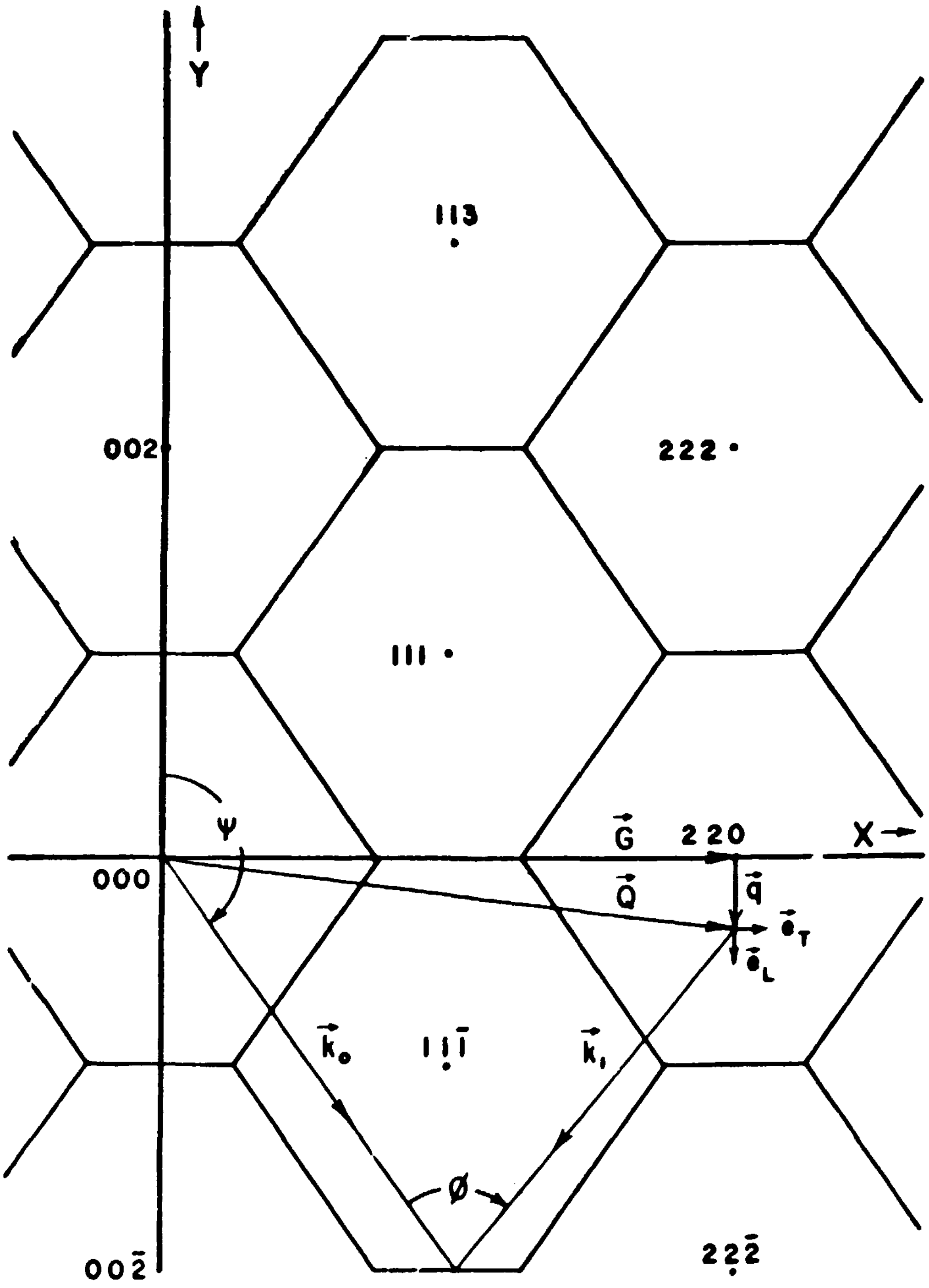

Figure 2 – The $(1\bar{1}0)$ plane of copper reciprocal lattice showing a typical inelastic scattering process

In the constant Q method the wave vector transfer vector $\vec{Q}$ is preselected, in magnitude and direction, corresponding to a particular position in the reciprocal lattice, and the frequency is scanned over a preselected range. A vector $\vec{Q}$ associated with a phonon wave vector $\vec{q}$, is indicated by the detection of a neutron group centered at the phonon frequency. Because of the periodicity of the dispersion relation, the phonon wave vector can always be referred to the first Brillouin zone by subtracting the appropriate reciprocal lattice vector $\vec{G}$ from $\vec{Q}$ to yield the first-zone wave vector $\vec{q}$. For example, Figure 2 shows a typical inelastic scattering process in the $(1\bar{1}0)$ plane of the copper reciprocal lattice. The vector construction can be seen to satisfy the wave vector conservation condition of equation (1). The wave vector $\vec{q}$ of the phonon has been referred to the (220) zone and is propagating in the $[00\xi]$ direction.

A typical neutron group for copper corresponding to a transverse acoustic phonon propagating in the $[00\xi]$ direction is shown in Figure 3.

II.2 – Graphical Focusing Method

A neutron group measurement with a triple-axis crystal spectrometer is affected by the contributions to the resolution from each of the collimators and from the mosaic spread of monochromator and analyzer crystal. The small but finite spread in direction of the neutrons falling on the monochromator crystal and the mosaic structure of the crystal itself give rise to a finite spread in the energy of the neutrons in the monochromatic beam. The ends of the wave vectors of neutrons in the beam occupy a thin disk in energy-momentum space. A similar disk is defined by the ends of the wave vectors of neutrons accepted by the analyser. The broadening of the observed phonon peak due to the resolution of the apparatus can be minimized by adjusting the orientation of these two disks relative to the dispersion surface. The conditions necessary to achieve this are known as focusing conditions[5]. The matching may be performed by focusing individually the monochromator and analyzer.

Considering only the finite collimation, with the effects of mosaic spread neglected, the incident monochromatic beam that reach the sample has momentum between $\vec{k}_o$ and $\vec{k}_o + \delta\vec{k}_o$.

The Bragg condition for the monochromator is:

$$\vec{k}_o \cdot \vec{d}_o = \text{const.} \tag{5}$$

where $\vec{d}_o$ is a vector normal to reflecting planes with magnitude equal to the plane spacing. By differentiation of equation (5),

$$d_o \cdot \delta k_o = 0 \tag{6}$$

it is seen that the disk defined by the ends of $\vec{k}_o + \delta\vec{k}_o$ is normal to $\vec{d}_o$, i.e., parallel to the monochromating planes (Figure 4).

The monochromator is focused when, of all the possible incident wave vectors between $\vec{k}_o$ and $\vec{k}_o + \delta\vec{k}_o$, a maximum number can give rise to the same final wave vector $\vec{k}_1$ after scattering from the sample. By differentiating equations (1) and (2) with respect to $\vec{k}_o$, keeping $\vec{k}_1$ fixed, the restriction on $\delta\vec{k}_o$ such that all neutrons scattered by the sample in a particular direction should have the same energy, can be obtained:

$$\delta\vec{k}_o = \delta\vec{q} \tag{7}$$

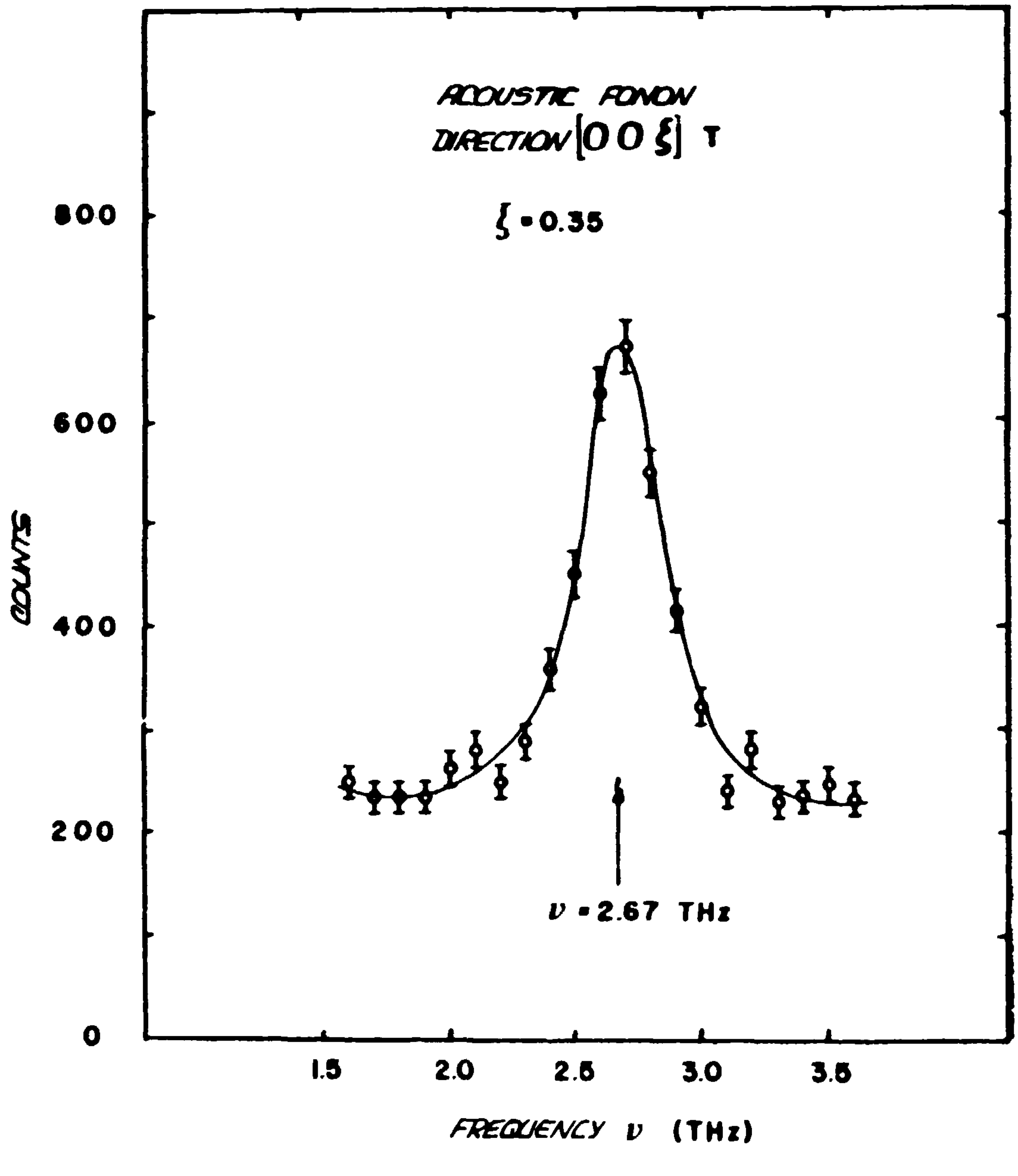

Figure 3 – A typical neutron group for copper, obtained by "constant Q" method, corresponding to a transverse acoustic phonon propagating in the [00ξ] direction

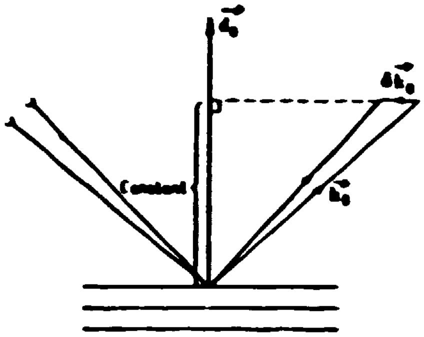

Figure 4 – Spread on the wave vector of the monochromatic beam caused by the finite collimation

and

$$\frac{\hbar}{m}\,\vec{k}_o\,.\,\delta\vec{k}_o \;=\; \nabla\omega\,.\,\delta\vec{k}_o \tag{8}$$

Defining $\vec{g} = \dfrac{m}{\hbar}\,\nabla\omega$, one obtains

$$(\vec{g} - \vec{k}_o)\,.\,\delta\vec{k}_o \;=\; 0 \tag{9}$$

Since by equation (6), $\delta\vec{k}_o$ is normal to $\vec{d}_o$, follows that $(\vec{g} - \vec{k}_o)$ is parallel to $\vec{d}_o$. This is the focusing condition for the monochromator, i.e, when the equations (6) and (9) are satisfied simultaneously for all $\delta\vec{k}_o$. If equations (1) and (2) are differentiated with respect to $\vec{k}_1$, keeping $\vec{k}_o$ fixed, a similar argument leads to the conclusion that the analyser will be focused when $(\vec{g} - \vec{k}_1)$ is parallel to $\vec{d}_1$.

The magnitude of $\vec{g}$ vector is calculated from equation (9):

$$\vec{g}\,.\,\delta\vec{k}_o \;-\; \vec{k}_o\,.\,\delta\vec{k}_o \;=\; 0$$

$$g\,\delta k_o \cos A \;=\; k_o\,\delta k_o \cos B$$

where A and B are the angles between $\vec{g}$ and $\delta\vec{k}_o$, and $\vec{k}_o$ and $\delta\vec{k}_o$, respectively

$$g \;=\; k_o\,\frac{\cos B}{\cos A} \tag{10}$$

The focusing conditions may be represented graphically on diagrams. The point D is the intersection of $\vec{d}_o$ with $\vec{d}_1$ and the point G is the end of $\vec{g}$ vector. The focusing conditions on monochromator, $\vec{k}_o$, and analyser, $\vec{k}_1$, can only be satisfied simultaneously if G coincides with D. Figures 5a, b, c illustrate schematically three different focusing conditions: optimum, good and defocused, respectively.

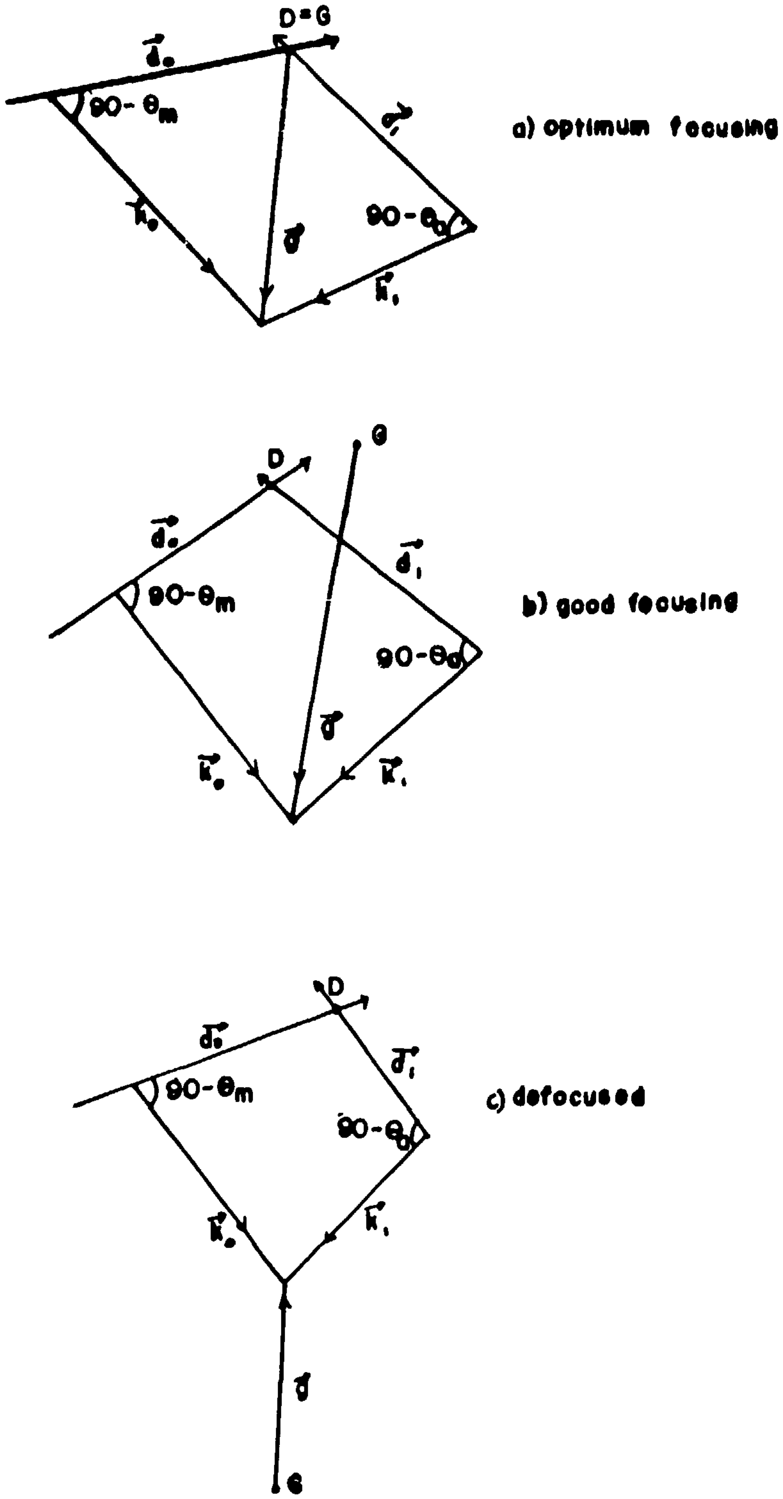

Figure 5 – Schematic diagram of three different focusing conditions: optimum, good and defocused

III – EXPERIMENTAL MEASUREMENT APPLYING FOCUSING CONDITIONS

In order to verify the predictions of the diagrams, shown in Figure 5, measurements of neutron groups in four positions (corresponding to reduced wave vectors $\xi = \pm 0.35$ and $\xi = \pm 0.5$) of the copper reciprocal lattice, related to transverse acoustic phonons propagating in the $|00\xi|$ directtion were performed with the IPEN triple-axis crystal spectrometer at the IEA-R1 2 Mw swimming pool research reactor. In the spectrometer the monochromator Bragg angle is fixed ($2\theta_M = 40.22°$), so for the Cu(111) monochromator planes the incident wave vector k_o is constant and equal to 2.519 $Å^{-1}$. An analyzer single crystal of pyrolytic graphite (2d = 6.708 Å) of high reflectivity, was used to determine the energy of the neutrons scattered from the sample.

Figure 6 shows two measuremetns for each value of ξ, of a transverse acoustic phonon in the $|00\xi|$ direction in copper. The measurements were made at two equivalent points P and R, in the reciprocal lattice for neutron energy loss. Corresponding focusing diagrams are shown in Figure 7.

A narrow and intense peak, corresponding to the optimum focusing condition, is shown in Figure 6a; the measurement was made at the position P of reciprocal lattice ($\xi = -0.35$) that provides the best-focused condition where point G is coincident with point D in the diagram 7a. Measurements performed in the equivalent position R, where $\xi = 0.35$, shows that the peak has almost disappeared and this is in agreement with the prediction of defocused condition corresponding to diagram 7b.

In Figure 6c it can be seen a well shaped peak measured at position P($\xi = -0.5$); the peak is not to much intense since it corresponds to the good, but not optimum, focusing condition showed in diagram 7c, where the point G is almost, but not exactly, coincident with point D. Figure 6d shows the measure made at the equivalent position R($\xi = 0.5$), in the defocused condition of diagram 7d, where the points G and D are widely separated.

From the simple comparison of the intensities and shapes of the phonon peaks measured under the three different focusing conditions presented in this work, it is possible to conclude about the importance of focusing in the routine procedure for operating a triple-axis spectrometer.

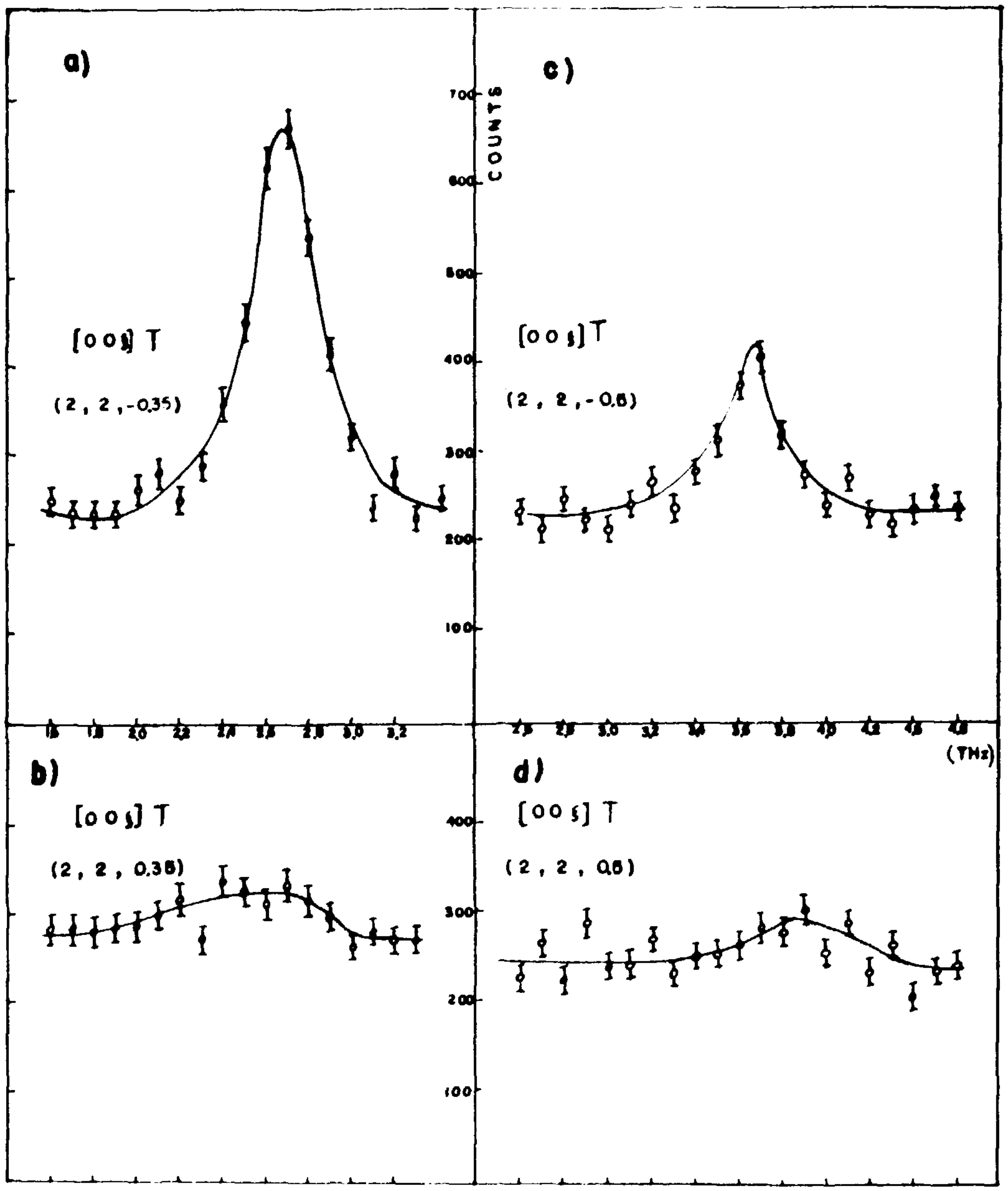

Figure 6 – Typical neutron groups measured in focused and defocused conditions

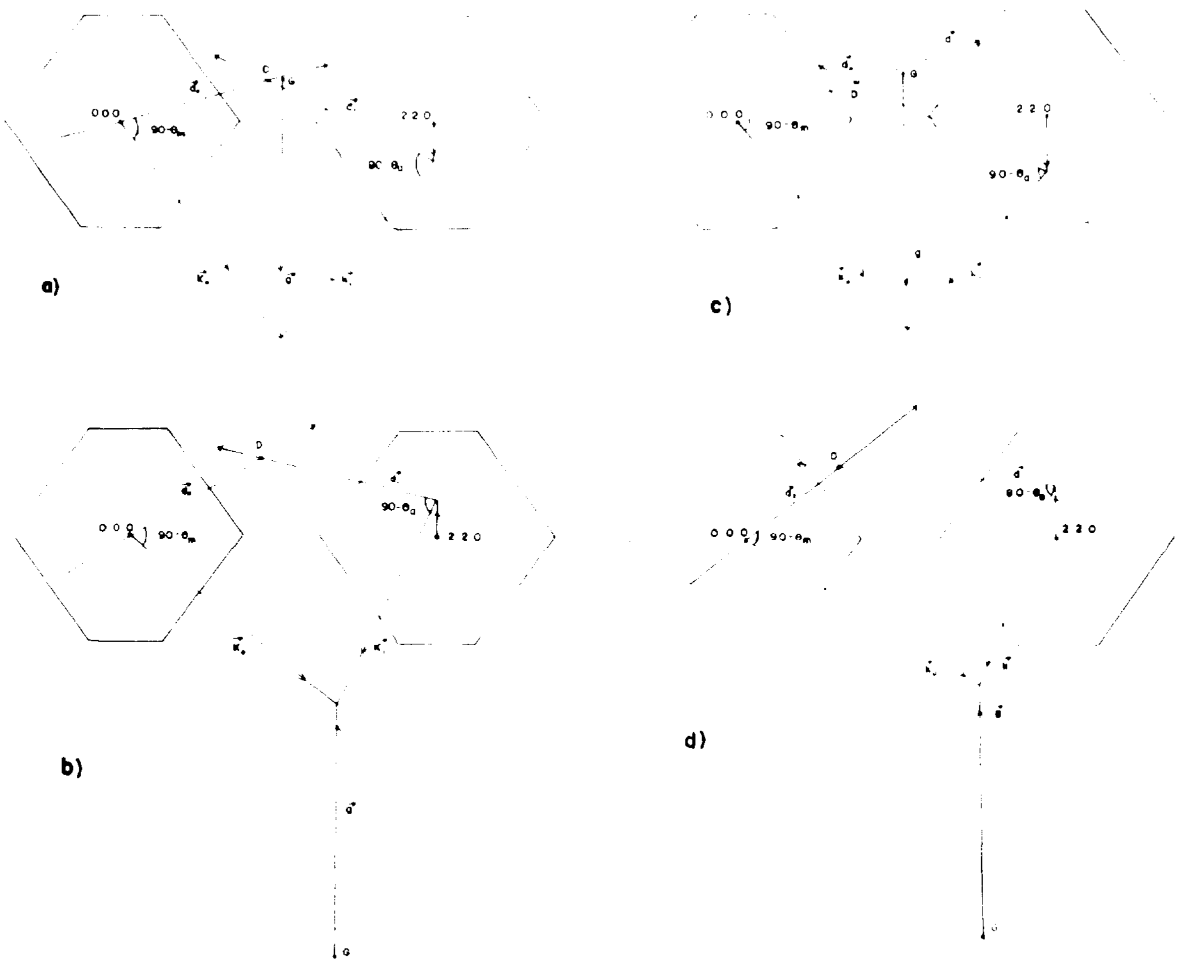

Figure 7 – Focusing diagrams corresponding to the measurements shown in Figure 6

RESUMO

O estudo do comportamento dinâmico de estruturas cristalinas é uma das mais importantes aplicações da técnica de espalhamento inelástico de nêutrons. Foi construído na Área de Física Nuclear do IPEN um Espectrômetro de Cristal de 3 Eixos, considerado o instrumento mais adequado para a realização de experiências sobre espalhamento coerente inelástico de nêutrons, onde ocorrem processos no qual um quantum de energia vibracional (fonon) é trocado entre o nêutron e uma amostra cristalina. O desempenho do aparelho foi verificado por meio de medida de relação de dispersão do cobre, usado como padrão. Em continuação a esses desenvolvimentos, foram deduzidas expressões para a obtenção das condições de focalização do instrumento, levando em conta o efeito da colimação finita, vetor de onda e ângulo dos feixes incidentes e espalhado, além das distâncias interplanares dos cristais monocromador e analisador. As condições de focalização foram representadas graficamente de maneira a se ter um prático e adequado procedimento de rotina para focalizar o espectrômetro. Para a verificação experimental foram medidos picos de fonons acústicos do cobre se propagando na direção (00ξ) de simetria do cristal. As medidas foram feitas em condições de ótima, boa e má focalização; a concordância dos resultados obtidos com as previsões teóricas mostra a utilidade do método gráfico adotado.

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