Neutron Kikuchi Effect: A New Method for Measuring Phonon Dispersion Curves

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Secondary Bragg scattering of neutrons which have already undergone inelastic scattering from a phonon will give rise to sharp changes in inelastic neutron scattering intensity near a Bragg peak. The effect should be observable, and the energy and momentum of the phonon can be found by a suitable experimental setup. Special attention is given to acoustic phonons and questions of resolution are discussed.

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Inelastic diffuse scattering of radiation in a crystal inevitably leads to the possibility of further Bragg (elastic) scattering in the same crystal. The effect was first observed by Kikuchi[1] for electrons and has come to be known as the Kikuchi (or K) effect (see, e.g., Cowley[2] for a recent discussion). The effect was observed much later with x rays,[3,4] although its possibility was actually considered earlier by Friedrich, Knipping, and Laue when they were devising an experiment to exhibit x-ray diffraction (see p. 8, Ref. 5). To my knowledge, the effect has not been knowingly observed or discussed for neutrons.

In a recent paper[6] (hereafter WCS), we explored some of the geometrical aspects of the x ray and γ ray K effects, with a view to explaining the origins of the "anomalously sharp dips" often observed in nonresonant Mössbauer measurements of thermal diffuse scattering (TDS) profiles.[7,8] Similar unexplained dips have also been observed for neutrons.[9]

The purpose of this Letter is to examine briefly the nature and experimental possibilities of the neutron Kikuchi effect and, in particular, to explore some of its geometrical aspects. Clearly, important differences between the neutron case and the electron and x ray cases will arise from the facts that (i) neutrons typically have thermal energies so that large relative changes in energy can occur on inelastic scattering by a phonon and (ii) the angular resolution in a neutron-scattering experiment is usually much lower. These factors might lead one to believe that the K effect would be extremely smeared out in the neutron case. However, our arguments below suggest that this will not be the case. Moreover, the present work has the following significant implications: (i) phonon dispersion curves may be measured on a two-axis diffractometer by using the crystal as both sample and analyzer (even for small crystals); (ii) doubt is cast on the validity of conventional methods for correcting neutron Bragg reflection data for TDS; and (iii) considerable simplication in treatment is achievable by using instrumental coordinates (Fig. 1).

For thermal-neutron scattering from a phonon, the neutron *conservation of momentum condition* (the triangle OCP in Fig. 2) reads

$$\vec{k} \equiv \vec{k}_s - \vec{k}_0 = \vec{G} + \vec{q}, \tag{1}$$

while the *conservation of energy condition* for one-phonon scattering (note, P no longer lies on the same energy surface as $\vec{k}_0$ in Fig. 2) is

$$\hbar^2(k_s{}^2 - k_0{}^2)/2m_n = -\epsilon\hbar\omega_p(\vec{q}), \tag{2}$$

where $\vec{k}_s$ and $\vec{k}_0$ are the wave vectors of the scattered and incident neutrons, respectively, $\vec{G}$ is a reciprocal-lattice vector, and m_n is the neutron mass; $\omega_p(\vec{q})$ is the phonon frequency for wave vector $\vec{q}$ if one assumes for simplicity in

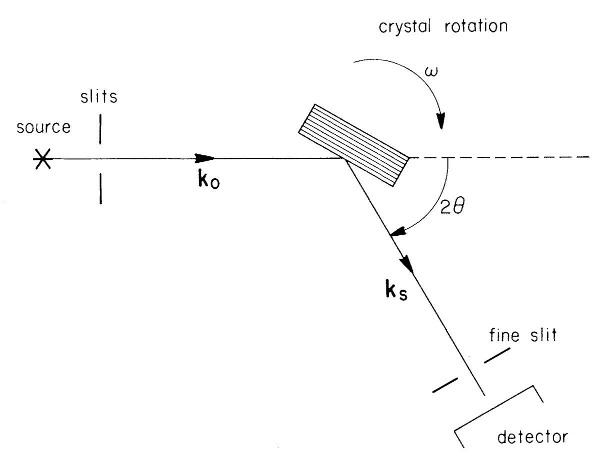

FIG. 1. Schematic diagram showing neutron-scattering geometry in the plane of diffraction, with ω denoting setting angle of the crystal and 2θ denoting setting angle of the detector.

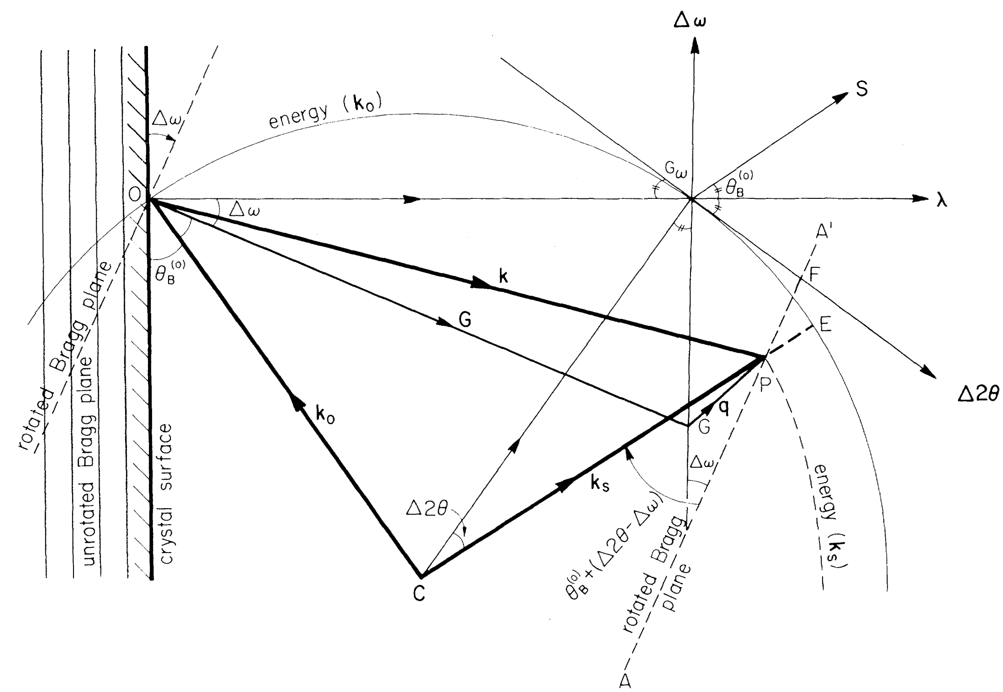

FIG. 2. Scattering geometry in reciprocal space for an incident neutron of wave vector $\vec{k}_0$, and showing the wave vectors for (i) scattering ($\vec{k}$), (ii) the scattered neutron ($\vec{k}_s$), (iii) the reciprocal-lattice point ($\vec{G}$), and (iv) the scattering phonon ($\vec{q}$), all taken when the crystal has been rotated by $\Delta\omega$ from the Bragg condition and the detector has been rotated by $\Delta 2\theta$. Also shown are constant energy surfaces corresponding to $\vec{k}_0$ and $\vec{k}_s$, and directions in reciprocal space corresponding to (i) wavelength variation (λ), (ii) crystal mosaicity or rotation ($\Delta\omega$), (iii) angular divergence of the source (S), and (iv) detector motion ($\Delta 2\theta$). The rotated Bragg plane (AA') through P gives the angle $\vec{k}_s$ which leads to the K condition.

notation that only a single phonon branch is present. The quantity ϵ is taken to be $+1$ or -1 accordingly as phonon creation or annihilation occurs. Equations (1) and (2) determine the topology of reciprocal-space scattering surfaces for neutrons, which for acoustic-phonon branches with $\omega_p(\vec{q}) = v_s q$ are conic sections of revolution.[10,11]

In order to discuss the neutron K effect, it is helpful to first introduce local coordinates ($\Delta 2\theta$, $\Delta\omega$) for angular deviations of the detector (2θ axis) and crystal (ω axis) both taken in the plane of diffraction (see Fig. 1) and such that the exact Bragg condition is ($\Delta 2\theta = 0$, $\Delta\omega = 0$). It is these instrumental coordinates of a diffractometer which have recently been used by Mathieson[12] to analyze the various instrumental and crystal factors affecting the distribution of scattering near a Bragg reflection (see also WCS[6]).

As for x rays[6,8] (but not electrons because of the more severe multiple scattering), the K effect for neutrons may reasonably be understood to arise from a predominant inelastic scattering process involving secondary Bragg (elastic) scattering of one-phonon scattered neutrons which peak in intensity near the given Bragg reflection. Clearly, any attempt to estimate K-line profiles for neutrons is still a multiple-scattering problem; however, the aim of the present Letter is essentially to examine only geometrical aspects of the K effect and the information which this can yield.

If the scattered wave vector, $\vec{k}_s$ [Eqs. (1) and (2)], satisfies the Bragg condition (elastic scattering) for the crystal rotated by $\Delta\omega$, then the K effect occurs in a direction $\Delta 2\theta$. It is our purpose to determine the curves in the ($\Delta 2\theta$, $\Delta\omega$) coordinate system for which this is possible. Each such curve is called a K locus. With use of geometry, as shown in Fig. 2, the following relation is found:

$$\theta_B(0) + (\Delta 2\theta^K - \Delta\omega^K) = \theta_B^{(s)}, \qquad (3)$$

where ($\Delta 2\theta^K$, $\Delta\omega^K$) is a point on a K locus, and $\theta_B^{(0)}$ and $\theta_B^{(s)}$ are the Bragg angles for the incident and scattered neutrons, respectively. When the neutron wave vector changes from $k_0 = 2\pi/\lambda_0$ to $k_s = 2\pi/\lambda_s$, the Bragg condition changes and the following approximate relation holds:

$$\frac{-(k_s - k_0)}{k_0} = \frac{\lambda_s - \lambda_0}{\lambda_0} = \cot\theta_B^{(0)}[\theta_B^{(s)} - \theta_B^{(0)}]. \quad (4)$$

The phonon energy $\hbar\omega_p(\vec{q})$ is now related to the coordinates of a point on a K locus through Eqs. (2)–(4):

$$\epsilon\hbar\omega_p(\vec{q}) = \frac{\hbar^2 k_0^2}{m_n}(1 - k_s/k_0)$$
$$= \frac{(\hbar k_0)^2}{m_n^2}\cot\theta_B^{(0)}(\Delta 2\theta^K - \Delta\omega^K), \quad (5)$$

with the assumption that $|k_s - k_0| \ll k_0$. The corresponding wave vector, $\vec{q}$, is also related to $(\Delta 2\theta^K, \Delta\omega^K)$. This relation is most easily found in a system with orthogonal axes λ and $\Delta\omega$ in Fig. 2 in which the coordinates of $\vec{q}$ are (q_λ, q_ω):

$$(2sck_0)\begin{pmatrix}\Delta 2\theta \\ \Delta\omega\end{pmatrix} = (k_0 - k_s)\begin{pmatrix}2s^2 \\ 1\end{pmatrix} + \begin{pmatrix}2s & 0 \\ s & c\end{pmatrix}\begin{pmatrix}q_\lambda \\ q_\omega\end{pmatrix}, \quad (6)$$

where $s = \sin\theta_B^{(0)}$ and $c = \cos\theta_B^{(0)}$. Substituting (5) in (6) yields

$$\begin{pmatrix}q_\lambda \\ q_\omega\end{pmatrix} = \frac{k_0}{s}\begin{pmatrix}0 & sc \\ -1 & 1+s^2\end{pmatrix}\begin{pmatrix}\Delta 2\theta^K \\ \Delta\omega^K\end{pmatrix} \quad (7)$$

for the phonon wave vector corresponding to the point $(\Delta 2\theta^K, \Delta\omega^K)$ on a K locus. Thus, both phonon energy and corresponding momentum may quite generally be derived from measured K loci by using (5) and (7).

In practice, most of the neutron TDS near a Bragg reflection comes from short-wave-vector acoustic modes (e.g., Ref. 13), for which a suitably simple case is

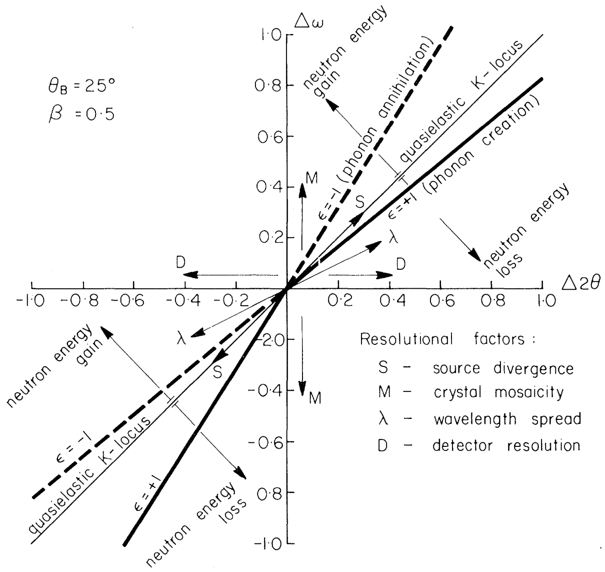

FIG. 3. Schematic illustration of TDS intensity in $(\Delta 2\theta, \Delta\omega)$ space showing loci dips (K loci) for an acoustic phonon branch with $\beta = v_s/v_n = 0.5$, which were obtained by solving (9). The thick solid line is the K locus for phonon creation, while the dashed solid line is for phonon annihilation.

$$\omega_p(\vec{q}) = v_s q, \quad (8)$$

where v_s is the sound velocity. Substitution of (8) into (5) and use of (7) for $\vec{q}$ yields the following equation for the K loci in $(\Delta 2\theta, \Delta\omega)$ space:

$$(\Delta 2\theta^K - \Delta\omega^K) = (\epsilon\beta/c)[(1+3s^2)(\Delta\omega^K)^2 - 2(1+s^2)\Delta 2\theta^K\Delta\omega^K + (\Delta 2\theta^K)^2]^{1/2}, \quad (9)$$

where $\beta = v_s/v_n$ and $v_n = \hbar k_0/m_n$. This leads to two broken straight lines (Fig. 3) for $\beta \leq 1$ and has no physical solution for $\beta > 1$. Significantly, the case $\beta = 1$ involves the changeover from hyperbolic ($\beta < 1$) to elliptic ($\beta > 1$) scattering surfaces about $\vec{G}$.[9,10] For $\beta < 1$, the locus above the quasielastic case ($\Delta\omega^K = \Delta 2\theta^K$, i.e., $\beta = 0$) corresponds to phonon annihilation ($\epsilon = -1$), while the locus below corresponds to phonon creation ($\epsilon = +1$), and the direction of dispersion [via (5)] is normal to the $\Delta\omega = \Delta 2\theta$ line (Fig. 3). As β is varied from 0 to 1, the K loci diverge from the quasielastic case and ultimately collapse onto the λ line (Fig. 3).

Although the scattering geometry for the K effect is quite complicated to depict in terms of conventional neutron-scattering surfaces in reciprocal space, a very simple picture emerges if one considers the K loci in instrumental $(\Delta 2\theta, \Delta\omega)$ space, which indicate the occurrence of dips in such TDS intensity plots. The K effect clearly leads to dispersion of neutrons having undergone one-phonon inelastic scattering in the crystal and so is akin to doing a triple-axis neutron spectrometer experiment, only using the same crystal as both sample and analyzer. For other than long-wavelength acoustic modes, the K loci will no longer be straight lines in general. Nonetheless, it is still true in general that measurement of the $(\Delta 2\theta, \Delta\omega)$ coordinates of a K line give $\vec{q}$ [via (7)] and $\omega_p(\vec{q})$ [via (5)]. Note also that not all values of $\vec{q}$ are accessible via the K effect.

Instrumental resolution and crystal characteristics will affect the observability of neutron K loci. The main factors affecting such observability are indicated on Fig. 3 by arrows signifying the principal direction in which the factor leads to smearing out of resolution in ($\Delta 2\theta$, $\Delta\omega$) space. Note, e.g., that both S and λ may lie nearly parallel to K loci and so have virtually no broadening effect (see also WCS[6]).

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