RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 31, NUMBER 11 1 JUNE 1985

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Coherent phonon scattering and the neutron Kikuchi effect

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(Received 4 March 1985)

The dynamical structure factor is derived for Bloch neutrons, the eigenstates of neutrons in the static lattice, and gives rise to corrections to the thermal diffuse scattering. For neutrons, the lines of secondary Bragg diffraction render the dynamics of the crystal lattice and can be obtained directly from sections through the scattering surfaces. The shape and the observability of the lines are analyzed for the Laue and Bragg cases.

A line pattern should arise if thermal neutrons, scattered by perfect crystals, undergo subsequent dynamical Bragg diffraction. Following Cowley,[1] it seems reasonable to call this line pattern a K-line pattern for neutrons as well as for electrons, x rays, and γ rays.

For elastic incoherent scattering of neutrons the K lines[2] should resemble the Kossel lines of x rays. The corresponding lines of inelastic coherent scattering of neutrons are the Kikuchi lines of electrons, insofar as they are caused by thermal diffuse scattering[3] (TDS). Even for x rays such lines arise from TDS.[4,5] Dips, found for several reflections in the TDS profiles of γ rays, are believed to be caused by secondary Bragg scattering (BS),[6] although their complete interpretation is still open.[7]

It is our aim to embed the effects of primary and secondary dynamical diffraction in the frame of the conventional theory of neutron scattering, and to consider the geometrical aspects of the K-line pattern in a more general way than had been done by Wilkins.[8]

Our interest will be focused on acoustic phonons and, for reasons of a more concise description, we consider only Bravais lattices. In perfect crystals the neutrons are described by the Bloch waves[2]

$$\phi_{\mathbf{k}}(\mathbf{x})=\sum_{i,\mathbf{G}} u_i(\mathbf{G})\exp[i(\mathbf{K}_i+\mathbf{G})\mathbf{x}]$$

of the dynamical theory of diffraction, rather than by plane waves. $u_i(\mathbf{G})$ is the Bloch coefficient belonging to the reciprocal lattice vector $\mathbf{G}$ and wave field i, excited by the incident wave $\mathbf{k}$ (the notation of Ref. 9 is used). The corresponding quantities of the scattered neutron are primed.

The coherent scattering function is defined as

$$S_c(\mathbf{k},\mathbf{k}',\omega)=\frac{1}{2\pi\hbar N}\int dt\, e^{i\omega t}\sum_{l,l'}[\langle\phi^*_{\mathbf{k}}(\mathbf{x}_l(t))\phi_{\mathbf{k}'}(\mathbf{x}_l(t))\phi^*_{\mathbf{k}'}(\mathbf{x}_{l'}(0))\phi_{\mathbf{k}}(\mathbf{x}_{l'}(0))\rangle-\langle\phi^*_{\mathbf{k}}(\mathbf{x}_l)\phi_{\mathbf{k}'}(\mathbf{x}_l)\rangle\langle\phi^*_{\mathbf{k}'}(\mathbf{x}_{l'})\phi_{\mathbf{k}}(\mathbf{x}_{l'})\rangle]\ , \tag{1}$$

where N is the number of unit cells and $\mathbf{x}_l(t)$ is the actual position of the nucleus l. Since the lattice periodic part of the interaction is added to the unperturbed Hamiltonian, Eq. (1) does not contribute to the elastic scattering. As a first approach attenuation effects and multiphonon scattering processes are neglected. Then the coherent scattering function for one-phonon creation (annihilation) processes is

$$S_c^{\pm1}(\mathbf{k},\mathbf{k}';\omega)=\sum_{\mathbf{q}\lambda}\frac{n_{\mathbf{q}\lambda}+\frac{1}{2}\pm\frac{1}{2}}{2M\omega_{\mathbf{q}\lambda}}\delta(\omega\mp\omega_{\mathbf{q}\lambda})P_{\mathbf{q}\lambda}(\mathbf{k},\mathbf{k}')\ , \tag{2}$$

with

$$P_{\mathbf{q}\lambda}(\mathbf{k},\mathbf{k}')=\left|\frac{1}{N}\sum_{\substack{\mathbf{G},\mathbf{G}'\\ i,j}} u_i^*(\mathbf{G})u_j'(\mathbf{G}')\exp[-W(\boldsymbol{\kappa}_{ij}+\mathbf{G}-\mathbf{G}')]\Delta(\boldsymbol{\kappa}_{ij}-\mathbf{q})(\boldsymbol{\kappa}_{ij}+\mathbf{G}-\mathbf{G}')\boldsymbol{\epsilon}(\mathbf{q}\lambda)\right|^2\ . \tag{3}$$

There M denotes the mass of the nucleus, W the Debye Waller factor, and $\boldsymbol{\kappa}_{ij}=\mathbf{K}_i-\mathbf{K}_j'$. $\omega_{\mathbf{q}\lambda}$ is the frequency of a phonon of branch λ, polarization vector $\boldsymbol{\epsilon}$, and thermal weight $n_{\mathbf{q}\lambda}$. In thin crystals, where the thickness D is of the order of the Pendellösung length Δ_0, the lattice sum (lattice vector $\mathbf{a}_l$)

$$\Delta(\boldsymbol{\kappa}_{ij}-\mathbf{q})=\sum_{l}\exp[-i(\boldsymbol{\kappa}_{ij}-\mathbf{q})\mathbf{a}_l] \tag{4}$$

describes the interference phenomena, i.e., the "Pendellösung" oscillations,[10] which become less important with increasing thickness. We restrict ourselves to thick crystals for reasons of intensity. Then, Eq. (4) expresses the pseudomomentum conservation only. Replacing κ_{ij} by the scattering vector $\kappa = \mathbf{k} - \mathbf{k}'$, one obtains

$$P_{q\lambda}(\mathbf{k},\mathbf{k}') = \sum_{\tau} \delta_{\kappa,q-\tau} \sum_{ij} \left| \sum_{\mathbf{G},\mathbf{G}'} u_i^*(\mathbf{G}) u_j'(\mathbf{G}') \exp[-W(\kappa+\mathbf{G}-\mathbf{G}')](\kappa+\mathbf{G}-\mathbf{G}')\epsilon(q\lambda) \right|^2 , \tag{5}$$

where τ is a reciprocal lattice vector. Keeping in mind that $\mathbf{q} = \kappa + \tau$, the sum over τ and the Kronecker δ are omitted in Eqs. (6), (7), and (9). If neither the incident nor the scattered neutron fulfill any Bragg condition, then

$$P = e^{-2W(\kappa)} |\kappa\epsilon|^2 \tag{6}$$

is just the result of the usual theory of neutron scattering.[11]

If only the primary wave excites a Bragg reflection, i.e., $\mathbf{G}$, one obtains, in the Laue case

$$P = \tfrac{1}{2} e^{-2W(\kappa)} \left[|\kappa\epsilon|^2 \left(1 + \frac{y^2}{1+y^2} \right) + \frac{|\beta(\kappa+\mathbf{G})\epsilon|^2}{1+y^2} - \frac{2y(\kappa\epsilon)[\beta(\kappa+\mathbf{G})\epsilon]}{1+y^2} \right] , \tag{7}$$

where

$$\beta = (\sqrt{|\cos\gamma/\cos\gamma_G|})^{1/2} \exp[-W(\kappa+\mathbf{G}) + W(\kappa)] \tag{8}$$

is an almost geometrical expression.[2] γ and γ_G are shown in Fig. 1. y measures the deviation of the incident wave from the Bragg condition and is, especially for electron diffraction, referred to as "Selektionsfehler."[9] In the limit $y \to \pm\infty$, P approaches the expression for the one-phonon scattering far from any Bragg reflection, i.e., Eq. (6). Whether the TDS is enhanced or reduced depends on whether $(\kappa\epsilon)^2$ is smaller or larger than $[\beta(\kappa+\mathbf{G})\epsilon]^2$, as shown in Fig. 1.

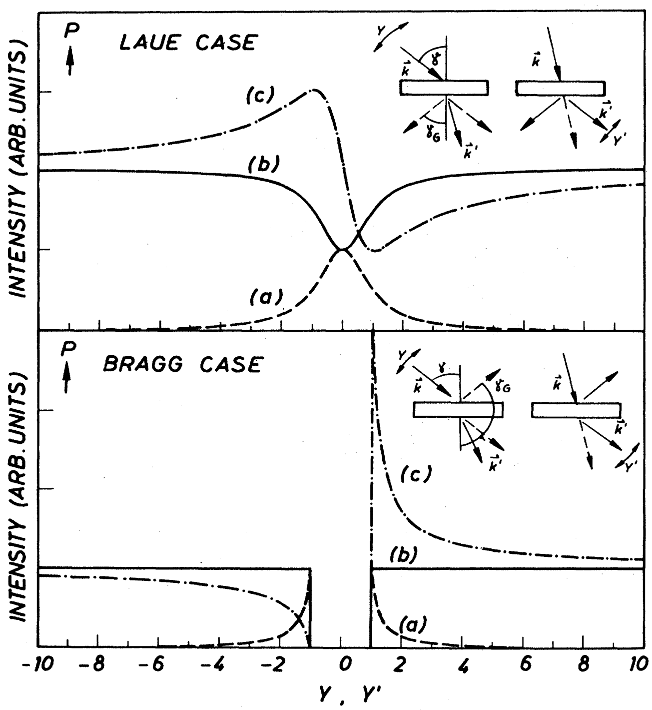

FIG. 1. Intensity for primary and secondary Bragg diffraction as a function of the setting angle of the crystal (y) and of the detector (y'), respectively. (a) Excess intensity $\kappa\epsilon = 0$. (b) Defect intensity $(\kappa+\mathbf{G})\epsilon = 0$. (c) $(\kappa\epsilon)^2 = [\beta(\kappa+\mathbf{G})\epsilon]^2$.

For the Bragg case, P may be approximated by

$$P = e^{-2W(\kappa)} \left(|\kappa\epsilon|^2 + \frac{|\beta(\kappa+\mathbf{G})\epsilon|^2}{2y^2-1} + \frac{2y(\kappa\epsilon)[\beta(\kappa+\mathbf{G})\epsilon]}{2y^2-1} \right) \Theta(|y|-1) . \tag{9}$$

In the range of total reflection only a layer of the thickness of the penetration depth, i.e., Δ_0, contributes to the TDS. These contributions to the scattering function are of the order $\Delta_0/D \ll 1$ and therefore are negligible. Although the profiles for the Laue and Bragg cases differ considerably, the conditions for enhanced or reduced TDS are quite similar if the profiles are understood as being integrated over the width of the incident beam. If noncentrosymmetrical reflections are considered, β will include a complex phase factor and, hence, the third, antisymmetric term of Eqs. (7) and (9) is modified.

The partial differential cross section is obtained from Eq. (2).[11] A subsequent integration over E', the energy of the scattered neutron, gives

$$\left(\frac{d\sigma}{d\Omega} \right)_c^{\pm 1} = N b_c^2 \frac{k'}{k} \sum_{q\lambda} \frac{\hbar}{2M\omega_{q\lambda}} \frac{n_{q\lambda} + \frac{1}{2} \pm \frac{1}{2}}{1 \pm \hbar \mathbf{k}' \nabla_q \omega_{q\lambda}/2E'} P_{q\lambda}(\mathbf{k},\mathbf{k}') , \tag{10}$$

where k' and E' have to be determined from energy and momentum conservation. b_c is the coherent scattering length.

The contributions to the TDS by primary dynamical scattering, derived above, appear in a typical range of some seconds of arc and will, therefore, generally be smeared out by the width of the incident beam. Thus, two possibilities arise: the restriction of the incident beam to the range of acceptance of dynamical diffraction and an enlargement of this range.

The former case may be realized with a two-crystal spectrometer. Keeping in mind that the scattered intensity is very sensitive only on the angular scale of the incident beam, the TDS should be changed by rotating the second crystal through the Bragg reflection. A considerable increase of the angular width for elastic scattering is obtained by backscattering. Qualitatively, an enhancement of these effects is expected for mosaic crystals, as explained by successive kinematical scattering.

Let us now turn to the case where only the scattered wave excites a Bragg reflection. The modifications of the one-phonon scattering treated up to now are also of interest for secondary Bragg scattering (BS), i.e., the K effect: (i) there is a notable analogy between primary and secondary BS if the relevant quantities are interchanged for the incident and the scattered neutron, respectively; (ii) the effects of primary and secondary BS are of the same order of magnitude. Thus, the evidence for TDS from primary Bragg diffraction can be used to estimate the demands for the evidence of the K effect.

For secondary dynamical BS, y, β, $\boldsymbol{\kappa}$, and $\mathbf{G}$ have to be replaced by y', β', $-\boldsymbol{\kappa}$, and $\mathbf{G}'$ in Eqs. (7)–(9), according to the rules mentioned above. y' denotes now the deviation of the scattered wave from the Bragg angle, and P characterizes the shape of the K pattern on an angular scale of some seconds of arc. The condition $(\boldsymbol{\kappa}\boldsymbol{\epsilon})^2 \ll [\beta(\boldsymbol{\kappa}-\mathbf{G}')\boldsymbol{\epsilon}]^2$ and its inverse inequality are connected with excess or defect lines. The antisymmetric third term of Eqs. (7) and (9), which becomes important if $(\boldsymbol{\kappa}\boldsymbol{\epsilon})^2 \sim [\beta(\boldsymbol{\kappa}-\mathbf{G}')\boldsymbol{\epsilon}]^2$, is known from electron diffraction to be responsible for the so-called K band. These profiles are shown in Fig. 1. It should be noted that anomalous attenuation changes this picture considerably.[12] The excess curve in Fig. 1 shows that (transverse) phonons with a vanishing one-phonon structure factor $\boldsymbol{\kappa}\boldsymbol{\epsilon}=0$ contribute to the intensity—although it is forbidden in the usual theory of neutron scattering [Eq. (6)]—via dynamical diffraction according to Eqs. (7) and (9).

In Fig. 2 the scattering surfaces $S_\lambda^{\pm}$ for phonon emission (absorption) processes are displayed in the reciprocal lattice of Al (Ref. 13) for 1.10-Å neutrons. The ellipses belong to the longitudinal phonon branch, for which the velocity of sound c_l is slower than the velocity of the neutron v_n, whereas for the transverse phonon branch $v_n > c_t$. The deviation of $\mathbf{k}$ from the Bragg condition with respect to $-\mathbf{G}'$ is[2] $d_K=(G'^2-2\mathbf{k}\mathbf{G}')/2G'$. Therefore all wave vectors $\mathbf{k}'$, which have their end points on the planes $K_{\pm\mathbf{G}'}$ (see Fig. 2) displaced by d_K from the Bragg planes, fulfill the Bragg condition. Energy and momentum conservation restrict the conditions for secondary BS on the intersecting curves of $K_{\pm\mathbf{G}'}$ with the scattering surfaces $S_\lambda^{\pm}$. The K lines are the projections of these intersecting curves onto the film. The plane $K_{\mathbf{G}'}$ does not intersect the ellipsoid of the longitudinal phonon in Fig. 2, indicating that there is no K effect for slower-than-sound neutrons.[8] But, as shown in Fig. 2, if other reflections such as $\pm\mathbf{G}'\pm\boldsymbol{\tau}$ are considered, K lines appear even for slower-than-sound neutrons, and are far from the straight lines of electron diffraction, as demonstrated in the inset of Fig. 2. Investigating the conditions of an enhanced K effect, one perceives that for $G'^2=4k^2(1-v_n^2/c_\lambda^2)$, $K_{\pm\mathbf{G}'}$ and $S_\lambda^{\pm}$ are parallel.

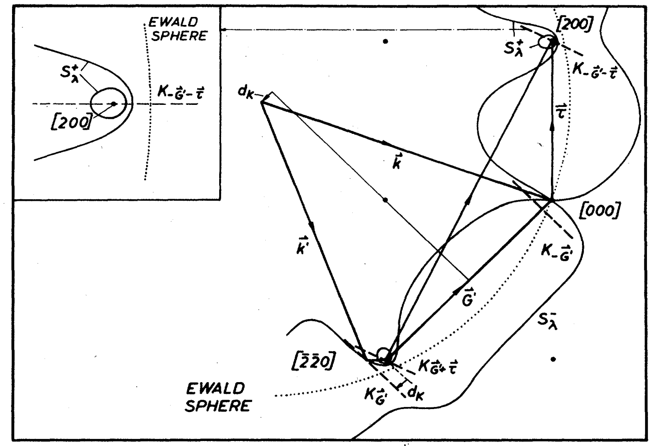

FIG. 2. Construction of the conditions for secondary Bragg scattering (BS) in the reciprocal lattice of Al. The scattering surfaces $S_\lambda^{\pm}$ determine the conditions for the one-phonon scattering (only one transverse phonon is shown). The planes $K_{\pm\mathbf{G}'}$ and $K_{\pm\mathbf{G}'\pm\boldsymbol{\tau}}$, which are perpendicular to the plane of drawing, determine the conditions for secondary BS. The intersecting curves of $S_\lambda^{\pm}$ with $K_{\pm\mathbf{G}'\pm\boldsymbol{\tau}}$ are the K lines as they are shown in the inset.

To estimate the requirements for the experimental evidence of the K effect, we can take over the considerations for primary BS by interchanging incident and scattered neutrons. As already mentioned, the effects of secondary dynamical diffraction are of the order of a few seconds of arc. Thus, generally, the K effect will be smoothed by the divergence of the source. Nevertheless, the experimental confirmation of the K effect seems to be realizable under sufficiently well-defined conditions.

Helpful discussions with R. Folk, H. Rauch, and A. Zeilinger are gratefully acknowledged. The work is supported by the Fonds zur Förderung der wissenschaftlichen Forschung, Project No. 4230.

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