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Neutron Kossel effect

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Incoherently scattered neutrons undergoing subsequent Bragg scattering give rise to a line pattern, which should be observable in crystals with pronounced coherent and incoherent scattering. The shape of these lines is obtained from the elastic, incoherent differential cross section. Both Laue and Bragg cases are considered.

Line patterns produced by divergent radiation appear in the scattering of x rays or electrons under a great variety of experimental conditions. It is a somewhat surprising fact that, for neutrons, these line patterns have been neither investigated thoroughly nor observed experimentally. Kossel lines arise from x rays generated within the crystal, whereas the to-some-extent similar Kikuchi lines are caused by inelastic scattering of electrons. In the past years, lines have been detected in diffraction patterns of x rays which are believed to be caused by thermal diffuse scattering.[1]

In large, perfect crystals the neutrons are described by Bloch waves rather than by plane waves. Hence, the dynamic structure factor is modified. Coherent neutron scattering should be responsible for a diffraction pattern analogous to the Kikuchi pattern of electrons,[2,3] whereas the line pattern of incoherent neturon scattering should look more like the Kossel pattern of x rays. Following Cowley[4] both types of lines are denoted as K lines.

The Hamiltonian for the Bloch neutron is given by

$$H_n=\frac{\mathbf{p}^2}{2m}+\frac{2\pi\hbar^2}{m}b_c\sum_l\langle\delta[\mathbf{x}-\mathbf{x}_l(t)]\rangle \ , \tag{1}$$

where the first term on the right-hand side describes the kinetic energy of the neutron and the second term the interaction with the static lattice. b_c is the coherent scattering length and $\mathbf{x}_l(t)$ the actual position of the nucleus l. The eigenfunctions

$$\phi_{\mathbf{k}}(\mathbf{x})=\sum_{i,\mathbf{G}}u_i(\mathbf{G})\exp[i(\mathbf{K}_i+\mathbf{G})\mathbf{x}] \tag{2}$$

are known from the dynamical theory of diffraction (see Ref. 5). $u_i(\mathbf{G})$ are the Bloch coefficients, where i labels the wave fields which are excited by the incident wave $\mathbf{k}$.

The incoherent scattering function is for Bravais lattices defined as

$$S_i(\mathbf{k},\mathbf{k}',\omega)=\frac{1}{2\pi\hbar N}\int dt\, e^{i\omega t}\sum_l\langle\phi^*_{\mathbf{k}}(\mathbf{x}_l(t))\phi_{\mathbf{k}'}(\mathbf{x}_l(t))\phi^*_{\mathbf{k}'}(\mathbf{x}_l(0))\phi_{\mathbf{k}}(\mathbf{x}_l(0))\rangle \ . \tag{3}$$

N is the number of unit cells and $\mathbf{k}$ and $\mathbf{k}'$ the wave vectors of the incident and the scattered neutrons, respectively.

For the elastic part of the incoherent scattering function one obtains from Eqs. (2) and (3)

$$S_i^0(\mathbf{k},\mathbf{k}',\omega)=\delta(\hbar\omega)\frac{1}{N}\sum_l\left|\sum_{\substack{\mathbf{G},\mathbf{G}'\\ i,j}}u_i^*(\mathbf{G})u_j'(\mathbf{G}')\exp[-W(\boldsymbol{\kappa}_{ij}+\mathbf{G}-\mathbf{G}')-i\boldsymbol{\kappa}_{ij}\mathbf{a}_l]\right|^2 \ , \tag{4}$$

where W is the Debye-Waller factor, $\boldsymbol{\kappa}_{ij}=\mathbf{K}_i-\mathbf{K}_j'$, and $\mathbf{a}_l$ is the equilibrium position of the nucleus l. If the thermal motion is negligible, then Eq. (4) is given by the probability density of the incident and scattered neutrons at the lattice points l:

$$S_i^0(\mathbf{k},\mathbf{k}',\omega)=\delta(\hbar\omega)\frac{1}{N}\sum_l|\phi_{\mathbf{k}}(\mathbf{a}_l)|^2|\phi_{\mathbf{k}'}(\mathbf{a}_l)|^2 \ . \tag{5}$$

If the incident and the scattered neutrons are far from any Bragg condition, Eq. (5) gives the familiar isotropic incoherent scattering.

Now we assume that only the incident wave fulfills the Bragg condition. To evaluate the lattice sum in Eq. (4) it is necessary to fix the boundary conditions. For a crystal slab (thickness D) and Laue arrangement one obtains

$$S_i^0(\mathbf{k},\mathbf{k}',\omega)=\delta(\hbar\omega)e^{-2W(\boldsymbol{\kappa})}\left[1+\frac{\beta^2-1-2y\beta}{1+y^2}\left(1-\frac{\sin[A(1+y^2)^{1/2}]}{A(1+y^2)^{1/2}}\right)\right] \ . \tag{6}$$

y is the *Selektionsfehler* (deviation of $\mathbf{k}$ from the Bragg angle, see Ref. 5), $A=\pi D/\Delta_0$ is the thickness in units of the Pendellösung length Δ_0, $\boldsymbol{\kappa}=\mathbf{k}-\mathbf{k}'$ is the scattering vector, and

$$\beta=(|\cos\gamma/\cos\gamma_G|)^{1/2}\exp[-W(\boldsymbol{\kappa}+\mathbf{G})+W(\boldsymbol{\kappa})] \tag{7}$$

is a predominantly geometrical expression (see Fig. 1), defined for $b_c>0$ and centrosymmetrical reflections and changes the sign for $b_c<0$. Neglecting the Pendellösung oscillations, the differential cross section is

$$\left(\frac{d\sigma}{d\Omega}\right)_1^0=\frac{N\sigma_i}{4\pi}e^{-2W(\boldsymbol{\kappa})}\left(1+\frac{\beta^2-1-2y\beta}{1+y^2}\right) \ , \tag{8}$$

where σ_i is the total incoherent cross section for a single nucleus. It should be kept in mind that the incoherent scattered intensity is almost isotropic, but its value depends strongly on the angle of incidence. The intensity profile shown in Fig. 1 is obtained by rotating the crystal through the Bragg orientation. A similar profile arises, if one measures—instead of the incoherently scattered intensity—the transmitted Bragg intensity, attenutated by incoherent scattering. Such measurements have been performed on potassium diphosphate (KDP) by Sippel and Eichhorn.[6]

This suggests that the verification of primary dynamical scattering is not beyond the experimental feasibility.

The scattering function is in the Bragg case

$$S_i^0(\mathbf{k},\mathbf{k}',\omega) = \frac{\delta(\hbar\omega)e^{-2W(\boldsymbol{\kappa})}}{2(y^2-\cos^2\alpha)}\left[2(y^2-1)+(\beta^2+1+2y\beta)\left(1-\frac{\sin 2\alpha}{2\alpha}\right)\right] , \tag{9}$$

where $\alpha=A(y^2-1)^{1/2}$ is imaginary for $|y|<1$. By the neglection of the terms with Δ_0/D, the differential cross section is given by

$$\left(\frac{d\sigma}{d\Omega}\right)_i^0 = \frac{N\sigma_i}{4\pi}e^{-2W(\boldsymbol{\kappa})}\left(1+\frac{\beta^2+2y\beta}{2y^2-1}\right)\Theta(|y|-1) , \tag{10}$$

as shown in Fig. 1. Θ is the Heaviside step function. In the range of total reflection only a layer of a thickness of the order of the penetration depth Δ_0 contributes to the incoherent scattering. Therefore, the intensity [Eq. (9)] remains nearly unchanged in the range of total reflection and for $D/\Delta_0>2$. If the crystals are thinner than $D/\Delta_0<0.2$ the intensity variations are smoothed for Laue and Bragg cases. Attenuation effects, which will change the results considerably, are not taken into account.

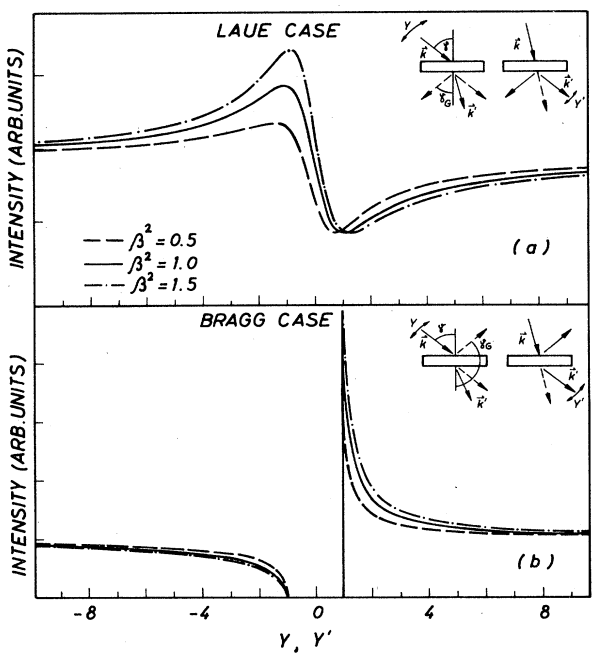

FIG. 1. Intensity of elastic, incoherent scattering in thick-crystal slabs. Attenuation effects are not accounted for. At $T=0$ β^2 [see Eq. (7)] is given by the ratio of the direction cosines: $\beta^2=|\cos\gamma/\cos\gamma_G|$. y is a parameter for the deviation of $\mathbf{k}$ from the Bragg angle (order of seconds of arc, see Ref. 5). For primary Bragg scattering y and β belong to the incident neutron, whereas for secondary Bragg scattering they have to be replaced by y' and β' of the scattered neutron and determine then the shape of the K lines.

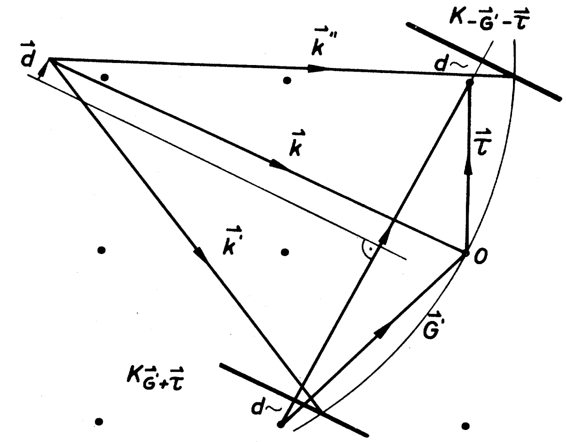

FIG. 2. Conditions for secondary Bragg scattering are fulfilled on the intersecting curves of the scattering surface (Ewald sphere) with the planes $K_{\pm G'\pm\tau}$, which are perpendicular to the plane of drawing.

Now, let us consider the situation where only the scattered neutron excites a Bragg reflection. The conditions for secondary Bragg scattering are fulfilled on the intersecting curves of the scattering surface (Ewald sphere) with planes $K_{\pm G'\pm\tau}$ parallel to the excited Bragg plane, as explained in Fig. 2. The characteristic distance is

$$\mathbf{d}=(\mathbf{G}'+\boldsymbol{\tau})\frac{G'^2-\tau^2-2\mathbf{k}(\mathbf{G}'+\boldsymbol{\tau})}{2(\mathbf{G}'+\boldsymbol{\tau})^2} .$$

The K lines are the projections of these intersecting curves onto the film. For the derivation of the shape of the K lines the large formal analogy between primary and secondary Bragg scattering enables us to take over the formulas of primary Bragg scattering by replacing $y,\beta,\boldsymbol{\kappa},\mathbf{G}$ by the corresponding quantities $y',\beta',-\boldsymbol{\kappa},\mathbf{G}'$ of the scattered neutron. Now, the curves in Fig. 1 represent sharp lines with a width of some seconds of arc. The intensity is enhanced or reduced relative to the background whether β is larger or smaller than one and, thus, gives rise to excess or defect lines. For $\beta'=1$ the profile is antisymmetic and may be referred to as K band.

Since primary and secondary Bragg scattering are of the same order of magnitude, the confirmation of the effect of primary Bragg scattering indicates the possibility of the verification of secondary Bragg scattering.

Generally, the effects of primary (or secondary) Bragg scattering will be smoothed by the wavelength spread of the incident beam. Thus, one may restrict the width of the incident beam to the range of acceptance of dynamical diffraction or one may enlarge this range, i.e., by back scattering.[3] Analogous to x rays,[4] the strength of the K lines is enlarged by a small mosaicity, but the lines become broad and can be explained from kinematical considerations.

For x rays, the divergent radiation can be generated outside the crystal and the resulting lines are known as pseudo-Kossel lines. Also for neutrons it seems to be possible to produce such a pseudo-Kossel pattern with incident divergent radiation. At least it may be mentioned that γ rays generated from neutrons in absorbing crystals are also subject to effects of primary and secondary Bragg scattering. Knowles[7] observed the variation of the γ-ray intensity from primary Bragg scattering in a weakly absorbing calcite crystal.

It has been shown that the Kossel lines should also appear in neutron scattering and their detection should be easier than the detection of the Kikuchi lines from thermal diffuse scattering. Under certain conditions they could give rise to additional corrections to the neutron structure factor and an experimental test of enhanced incoherent scattering under the Bragg peak would be interesting. As shown by Zeilinger and Shull[8] the incoherent (and inelastic) scattered neutrons can be measured in the Bragg case directly, without the necessity of having to use computations involving the small differences between large numbers.

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[1]Y. Kashiwashe and Y. Kainuma, J. Phys. Soc. Jpn. **51**, 2379 (1982).
[2]S. Wilkins, Phys. Rev. Lett. **50**, 1862 (1983).
[3]D. Petrascheck (unpublished).
[4]J. M. Cowley, *Diffraction Physics* (North-Holland, Amsterdam, 1975).
[5]H. Rauch and D. Petrascheck, in *Neutron Diffraction,* edited by H. Dachs, Topics in Current Physics, Vol. 6 (Springer, Berlin, 1978).
[6]D. Sippel and F. Eichhorn, Acta Crystallogr. Sec. A **24**, 237 (1968).
[7]J. W. Knowles, Acta Crystallogr. **9**, 61 (1956).
[8]A. Zeilinger and C. G. Shull, presented at the conference on "The Neutron and its Applications," Cambridge 1982 (unpublished).