The same 23 structures were later processed by *SIR* using one-phase (according to Cascarano, Giacovazzo, Calabrese, Burla, Nunzi, Polidori & Viterbo, 1984) and two-phase seminvariants in addition to conditions (*a*) and (*b*). In Table 3 for each structure the number of correct solutions is given (column PROT 2) together with the number of trials (in parentheses).

Only four structures remain unsolved by *SIR* [however, it cannot be concluded that *SIR* is unable to solve those structures using non-default conditions; see for example, Burla, Giacovazzo & Polidori (1987)]. Very often the ratio (number of correct solutions/number of trials) is larger in PROT 2 than in PROT 1. Thus Table 3 provides clear evidence of how relevant phase seminvariants may be for the success of direct phasing procedures.

Two further observations can be made.

(1) The search of seminvariant pairs is made among the largest NRIF reflections commonly used for the $\sum_2$ list, while **H** varies over the complete subset of one-phase seminvariants. Limiting the cross term **H** to the strongest reflexions reduces both the computing time of the procedure and (dramatically) the number of available two-phase seminvariants. A large subset of reliable two-phase seminvariants can be more easily found when **H** is unrestricted.

(2) The factor SC used to rescale α_2 on the triplet reliability parameter α_3 ($\alpha_3 = 2|E_{\mathbf{h}}E_{\mathbf{k}}E_{\mathbf{h}+\mathbf{k}}|/N^{1/2}$) was empirically chosen so as to satisfy

$$0{\cdot}6 \times \mathrm{SC} \times \sum \alpha_3 = \sum \alpha_2,$$

where the summations are over the most reliable 400 triplets and two-phase seminvariants respectively. The scaling scheme is unsatisfactory from the theoretical point of view, even if it works quite well in practice for all our test structures. It is hoped that future probability distributions will make available more efficient formulas for estimating seminvariants.

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Phase Dependence of Kikuchi Patterns. I. Theory

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Abstract

Thermal diffuse scattering in high-energy electron diffraction is analysed using the Einstein model of lattice vibrations. An expression for the intensity distribution in the Kikuchi pattern is obtained which includes thickness-dependent terms (*i.e.* a dependent-Bloch-wave theory is used) and the effects of a general crystal structure. The corresponding two-beam limit is shown to consist of four distinct terms, two of which depend on the phase of the structure factor. One of these is found to be non-zero only for non-centrosymmetric crystals and for relatively thin crystals. It leads to an asymmetric Kikuchi band, even in a symmetrical scattering geometry. This asymmetry may be used to determine the polarity of non-centrosymmetric crystals.

1. Introduction

The theory of thermal diffuse scattering in high-energy electron diffraction and the associated process of Kikuchi pattern formation have been extensively studied over the last thirty years. Many of the earlier

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papers discussed two-beam theory (*e.g.* Kainuma, 1955; Fukuhara, 1963; Fujimoto & Kainuma, 1963; Ishida, 1970, 1971; Okamoto, Ichinokawa & Ohtsuki, 1971; Chukhovskii, Alexanjan & Pinsker, 1973), this giving way to more general single scattering theories (Gjønnes, 1966; Gjønnes & Høier, 1971; Rez, Humphreys & Whelan, 1977; Rossouw, 1985; Rossouw & Bursill, 1985) and analysis of the relationship between single and multiple diffuse scattering (Høier, 1973; Yamamoto, 1980). In two recent papers Taftø (1983, 1987) has revived interest in two-beam theory by showing that structural and even phase information is retained in two-beam inelastic scattering. This is particularly interesting because of the well known result that phase information is lost in two-beam *elastic* scattering; at least three beams are required in this case [*e.g.* Bird, James & Preston (1987); see § 3 for more discussion on this point]. In his papers Taftø (1983, 1987) considered characteristic X-ray emission and energy-loss spectra from crystals with the non-centrosymmetric GaAs structure. By analysing the wave fields that are set up in two-beam diffraction he explained a strong asymmetry that was observed between orientations close to the Bragg condition for $+\mathbf{g}$ and $-\mathbf{g}$ reflections. This only occurs if the total intensity of the wave field is considered, rather than a sum of intensities from individual Bloch states. In other words, a dependent-Bloch-wave model must be used. Here we show that similar effects are present in two-beam thermal diffuse scattering. In non-centrosymmetric crystals this gives rise to an asymmetric Kikuchi band, even in a symmetrical scattering geometry where the incident orientation lies exactly in the centre of the band. In principle, this may be used to determine the polarity of any non-centrosymmetric crystal (Bird & Wright, 1988, 1989). Perhaps surprisingly, none of the papers referenced above examine the non-centrosymmetric case in detail. Fujimoto & Kainuma (1963) give a general expression for the diffusely scattered intensity but their discussion concentrates on centrosymmetric structures. Gjønnes (1966) also gives general two-beam expressions but does not make the difference between centro- and non-centrosymmetric structures explicit.

In § 2 a general expression for the intensity distribution in the Kikuchi pattern is discussed. In order to keep the analysis as simple as possible an Einstein model of lattice vibrations is used. The equivalent two-beam expression is shown to consist of four distinct terms of which, as is discussed in § 3, one is particularly interesting because it is non-zero only for non-centrosymmetric and relatively thin crystals. It is this term which retains phase information and leads to an asymmetric Kikuchi pattern. When combined, the four terms show how the Kikuchi pattern emerges as the crystal thickness is increased up to the eventual thick-crystal independent-Bloch-wave limit (Cherns, Howie & Jacobs, 1973; Rossouw & Bursill, 1985). The thickness cannot be too large, however, as only single diffuse scattering is considered. [Multiple scattering effects are well known to reverse the black/white Kikuchi line contrast in thick crystals (Høier, 1973)]. In a second paper (Bird & Wright, 1989) the special but important case of III–V and II–VI compounds with the GaAs structure is discussed. Computational and experimental results will be presented which illustrate the theory discussed here.

2. Theory

The quantity we aim to calculate is the intensity at a point $\mathbf{K}'$ in the Kikuchi pattern which arises from an incident electron at $\mathbf{K}$, where $\mathbf{K}$ and $\mathbf{K}'$ are wave-vector components parallel to the surface and describe the incident and outgoing electron orientations respectively. In a single-scattering approximation, and using the Einstein model, this becomes (Rez, Humphreys & Whelan, 1977; Rossouw & Bursill, 1985; Rossouw, 1985)

$$I(\mathbf{K}') \propto t \sum_{jj'} \sum_{ff'} \sum_{\mathbf{g}\mathbf{g}'} \sum_{\mathbf{h}\mathbf{h}'} C_{\mathbf{0}}^{j*}(\mathbf{K}) C_{\mathbf{g}}^{j}(\mathbf{K}) C_{\mathbf{0}}^{j'}(\mathbf{K}) \times C_{\mathbf{g}'}^{j'*}(\mathbf{K}) C_{\mathbf{0}}^{f}(\mathbf{K}') C_{\mathbf{h}}^{f*}(\mathbf{K}') C_{\mathbf{0}}^{f'*}(\mathbf{K}') C_{\mathbf{h}'}^{f'}(\mathbf{K}') \times \left\{ \frac{\exp[i(k_z^f - k_z^{f'})t] - \exp[i(k_z^j - k_z^{j'})t]}{i(k_z^f - k_z^{f'} - k_z^j + k_z^{j'})t} \right\} \times \sum_k \exp[i(\mathbf{g} - \mathbf{g}' - \mathbf{h} + \mathbf{h}').\mathbf{r}_\kappa] S^{(\kappa)}_{\mathbf{h}-\mathbf{g},\,\mathbf{h}'-\mathbf{g}'}(\mathbf{q}) \tag{1a}$$

where

$$S^{(\kappa)}_{\mathbf{g},\mathbf{g}'}(\mathbf{q}) = v_\kappa(\mathbf{q}+\mathbf{g}) v_\kappa(\mathbf{q}+\mathbf{g}') (\exp[-M_\kappa(\mathbf{g}-\mathbf{g}')^2] - \exp\{-M_\kappa[(\mathbf{q}+\mathbf{g})^2 + (\mathbf{q}+\mathbf{g}')^2]\}). \tag{1b}$$

The sums j, j', f, f' are over branches of the dispersion surface, j and j' represent incident states at orientation $\mathbf{K}$ and f and f' represent outgoing states at $\mathbf{K}'$. $C_{\mathbf{g}}^{j}$ and k_z^j are the corresponding Bloch-wave coefficients and z wave-vector components respectively and t is the crystal thickness. The sums $\mathbf{g}$, $\mathbf{g}'$, $\mathbf{h}$, $\mathbf{h}'$ are over reciprocal-lattice vectors; the projection approximation and a symmetric Laue geometry are used throughout the paper so all these vectors lie in the (x, y) plane, parallel to the crystal surfaces. κ represents the atoms within the unit cell. Each atom has a position coordinate $\mathbf{r}_\kappa$, an atomic form factor $v_\kappa(\mathbf{q})$ and a Debye–Waller factor M_κ. $\mathbf{q}$ represents the scattering vector and is defined as $\mathbf{q} = \mathbf{k}' - \mathbf{k}$, where $\mathbf{k}$ and $\mathbf{k}'$ are the wave vectors of the incident and outgoing plane waves. The component of $\mathbf{q}$ parallel to the surface is $\mathbf{K}' - \mathbf{K}$ and its z component is approximately $(\mathbf{K}^2 - \mathbf{K}'^2)/2k$, where k is the magnitude of the wavevector. This ignores the small differences in q_z due to the interactions between different Bloch states

in $S(\mathbf{q})$, but these effects are retained in the t-dependent term in $(1a)$. Absorption has not been included explicitly but could be taken into account by adding imaginary parts $i\lambda^j$ to the wave vectors. A uniform absorption $i\lambda$ acting on all the Bloch states simply leads to an overall $t\exp(-2\lambda t)$ factor (instead of t) multiplying $(1a)$. Only anomalous absorption therefore matters and since we shall be interested in relatively weak diffraction situations, this is not expected to be a major effect. Equation (1) includes the full thickness dependence of the diffuse scattering process (*i.e.* it represents a dependent-Bloch-wave result) and is valid for a general crystal structure. Rossouw (1985) also considers both these effects, but does not analyse the corresponding two-beam limit.

As it stands, (1) includes the effects of diffraction on both the incident and outgoing electrons. However, since the Kikuchi pattern is formed by the final electron states, the diffraction of these must be most significant. The simplest approximation is to assume that the incident wave is undiffracted, *i.e.* it remains a plane wave. In this case $(1a)$ becomes

$$I(\mathbf{K}')\propto t\sum_{ff'}\sum_{\mathbf{h}\mathbf{h}'} C_{\mathbf{0}}^{f}(\mathbf{K}')C_{\mathbf{h}}^{f}{}^{*}(\mathbf{K}')C_{\mathbf{0}}^{f'}{}^{*}(\mathbf{K}')C_{\mathbf{h}'}^{f'}(\mathbf{K}')$$
$$\times\exp(i\Delta^{ff'}t/2)[\sin(\Delta^{ff'}t/2)/\Delta^{ff'}t/2]$$
$$\times\sum_{\kappa}\exp[i(\mathbf{h}'-\mathbf{h}).\mathbf{r}_\kappa]S_{\mathbf{h},\mathbf{h}'}^{(\kappa)}(\mathbf{q}) \qquad (2)$$

where $\Delta^{ff'}=k_z^f-k_z^{f'}$ and is related to the extinction distance $\xi^{ff'}$ by $\Delta^{ff'}=2\pi/\xi^{ff'}$. Equation (2) contains only four sums over f's and $\mathbf{h}$'s and is therefore much simpler to compute than $(1a)$, although the time will still scale as N^4 in an N-beam calculation. In two-beam theory explicit expressions are available for the Bloch-wave coefficients and $\Delta^{ff'}$. We write the relevant reciprocal-lattice vector as $\mathbf{H}$ and the corresponding structure factor $U_{\mathbf{H}}$ as $-U\exp(i\varphi)$. The minus sign is included because electron form factors are negative – the interaction between electrons and atoms is basically attractive. The deviation parameter W is written $W=\mathbf{H}.\delta\mathbf{K}'$, where $\delta\mathbf{K}'$ is the deviation away from the exact Bragg condition, $-\mathbf{H}/2$. For a non-centrosymmetric crystal the standard two-beam theory gives

$$C_{\mathbf{0}}^{1}=2^{-1/2}[1+W/(U^2+W^2)^{1/2}]^{1/2}$$
$$C_{\mathbf{0}}^{2}=2^{-1/2}[1-W/(U^2+W^2)^{1/2}]^{1/2} \qquad (3a)$$
$$C_{\mathbf{H}}^{1}=\exp(i\varphi)C_{\mathbf{0}}^{2}$$
$$C_{\mathbf{H}}^{2}=-\exp(i\varphi)C_{\mathbf{0}}^{1}$$

and

$$\Delta^{1,2}=\Delta=(U^2+W^2)^{1/2}/k. \qquad (3b)$$

When (3) is substituted into (2) we eventually obtain

$$I\propto t\left[1-\tfrac{1}{2}\frac{U^2}{U^2+W^2}\left(1-\frac{\sin(\Delta t)}{\Delta t}\right)\right]\sum_{\kappa}S_{\mathbf{0},\mathbf{0}}^{(\kappa)}(\mathbf{q})$$
$$+t\left[\tfrac{1}{2}\frac{U^2}{U^2+W^2}\left(1-\frac{\sin(\Delta t)}{\Delta t}\right)\right]\sum_{\kappa}S_{\mathbf{H},\mathbf{H}}^{(\kappa)}(\mathbf{q})$$
$$+t\left[\frac{UW}{U^2+W^2}\left(1-\frac{\sin(\Delta t)}{\Delta t}\right)\right]$$
$$\times\sum_{\kappa}\cos(\varphi+\mathbf{H}.\mathbf{r}_\kappa)S_{\mathbf{0},\mathbf{H}}^{(\kappa)}(\mathbf{q})$$
$$+t\left[\frac{U}{(U^2+W^2)^{1/2}}\left(\frac{\sin^2(\Delta t/2)}{\Delta t/2}\right)\right]$$
$$\times\sum_{\kappa}\sin(\varphi+\mathbf{H}.\mathbf{r}_\kappa)S_{\mathbf{0},\mathbf{H}}^{(\kappa)}(\mathbf{q}). \qquad (4)$$

Equation (4) represents the basic result of this paper; it will be discussed in detail in the following section.

To conclude the present section we examine the approximation leading to (2) and (4), namely the assumption that the incident wave remains undiffracted. This should be a reasonably good approximation if $\mathbf{K}$ lies away from a region of strong diffraction, for example, near a high-index weak zone axis. There is no inconsistency between ignoring diffraction on the incoming states and including it on the outgoing states because $\mathbf{K}$ and $\mathbf{K}'$ are independent of each other. For any given $\mathbf{K}$ there will be a complete Kikuchi pattern formed by scattering into all possible $\mathbf{K}'$. Obviously, diffraction on the outgoing states must be kept because this is the origin of the pattern. The analysis can be extended a little further by looking at large-angle scattering in $(1a)$. In this case $\mathbf{q}$ is large compared with any $\mathbf{g}$ which contributes significantly, so $v_\kappa(\mathbf{q}+\mathbf{g})\simeq v_\kappa(\mathbf{q})$ and $\exp[-M_\kappa(\mathbf{q}+\mathbf{g})^2]$ is small. If we also assume that for the relevant $\mathbf{g}$'s $M_\kappa\mathbf{g}^2$ is small, $S^{(\kappa)}$ loses its $\mathbf{g}$ dependence and becomes simply $|v_\kappa(\mathbf{q})|^2$. This limit corresponds to having an interaction which is perfectly localized about each atomic site (*e.g.* Rossouw & Maslen, 1984). The only coupling between the incoming and outgoing states now acts through the thickness-dependent term in $(1a)$. This can be removed if we make an independent-Bloch-wave approximation for the initial states by setting $j=j'$. As usual, the justification for this is that terms with $j=j'$ are of order unity, while those with $j\neq j'$ fall off as $1/(k_z^j-k_z^{j'})t$. When the thickness exceeds the j,j' extinction distance, the terms with $j=j'$ begin to dominate. Although it might seem strange to have the incident states being independent and the final states dependent, this too can be justified by the independence of $\mathbf{K}$ and $\mathbf{K}'$. If we consider a relatively weak Kikuchi line that is well described by two-beam theory, the relevant extinction distance is fairly large, corresponding to a small splitting on the dispersion surface. However, if the incident orientation lies away from any weak two-beam point (for example, close

to a zone axis) the branch splittings are considerably larger and typically correspond to extinction distances of order 100 Å. Clearly cases could arise where the diffraction effects on the incoming and outgoing states are similar in size and this argument fails (Gjønnes, 1966). However, such situations can be avoided in experiments and the decoupling becomes a good approximation.

With $S^{(\kappa)}$ losing its **g** dependence and considering only terms with $j=j'$, the **K**-dependent terms in (1*a*) can be taken out to give

$$\sum_j \left| C_0^{j*}(\mathbf{K}) \sum_{\mathbf{g}} C_{\mathbf{g}}^j(\mathbf{K}) \exp(i\mathbf{g}.\mathbf{r}_\kappa) \right|^2 = \sum_j |\varepsilon^j \psi^j(\mathbf{K}, \mathbf{r}_\kappa)|^2 \quad (5)$$

where ψ^j is the jth Bloch wave

$$\psi^j(\mathbf{K}, \mathbf{r}) = \sum_{\mathbf{g}} C_{\mathbf{g}}^j(\mathbf{K}) \exp[i(\mathbf{K}+\mathbf{g}).\mathbf{r}] \exp(ik_z^j z) \quad (6)$$

with excitation amplitude $\varepsilon^j = C_0^{j*}(\mathbf{K})$. The sum of Bloch-wave intensities (*i.e.* independent Bloch waves) on each atomic site in (5) simply has the effect of modifying the $S^{(\kappa)}$ factors in (2) and (4), *without changing the basic structure of these equations.* Although this has been derived in a large-angle scattering limit a similar conclusion is expected to hold for smaller scattering angles where the localization of the interaction is not complete. It is also worth pointing out that in the same approximation, where $S^{(\kappa)}(\mathbf{q})$ is independent of **h** and **h'** (but retaining $f \neq f'$ terms), (2) can be written

$$I(\mathbf{K}') \propto \sum_\kappa S^{(\kappa)}(\mathbf{q}) \int_0^t dz \left| \sum_f \varepsilon^f \psi^f(\mathbf{K}', \mathbf{r}_\kappa) \right|^2 \quad (7)$$

where now $\varepsilon^f = C_0^{f*}(\mathbf{K}') \exp(-ik_z^f t)$ to match to plane waves below the crystal. This represents a dependent-Bloch-wave result, with the *total* intensity on each site being integrated over the crystal thickness. The difference between dependent and independent models can be clearly seen by comparison of (5) and (7). It is only in the dependent model that the thickness-dependent terms of (4) arise, and it is these terms that carry the interesting phase information.

3. Two-beam results

We now return to (4) and examine the details of the two-beam intensity. As it stands, (4) only refers to one of the pair of linked Kikuchi lines, but the intensity of the second line is simply obtained by setting $\mathbf{H} \rightarrow -\mathbf{H}$ and $\varphi \rightarrow -\varphi$. For both lines a positive value of W points towards the centre of the Kikuchi band (see Fig. 1). The variation of intensity with orientation comes from two factors, through W (both explicitly, and in Δ) and the **q** dependence of $S^{(\kappa)}$. For a relatively weak Kikuchi line the W dependence is most important since the diffraction is highly localized about the exact Bragg condition, $W=0$. We therefore ignore the **K'** dependence of the S term and calculate it with **K'** at the Bragg position. All the orientation dependence now lies in the factors in square brackets in (4). This part of the second, third and fourth terms is shown in Fig. 2 as a function of the dimensionless deviation parameter $w = W/U$ for thicknesses corresponding to $0{\cdot}1\xi$, $0{\cdot}25\xi$ and ξ, where $\xi = 2\pi k/U$ is the two-beam extinction distance at the Bragg position. The first term contribution is just 1 − (second term). Both the second and fourth terms are symmetrical about $W=0$ and give either a peak (bright line) or trough (dark line) depending on the sign of the term. The third term is antisymmetric about the Bragg position and gives a bright/dark line profile. In considering the $S^{(\kappa)}$ factors in (4) it is important to distinguish between a symmetrical and a general scattering geometry. In the symmetrical case the incident orientation lies exactly at the centre of the relevant Kikuchi band and $S_{\mathbf{0},\mathbf{0}}^{(\kappa)} = S_{\mathbf{H},\mathbf{H}}^{(\kappa)}$ because $|\mathbf{q}| = |\mathbf{q}+\mathbf{H}|$ (see Fig. 1). It follows that the first and second terms of (4) can be combined to give an orientation-independent term which simply represents the background diffuse scattering. The pair of Kikuchi lines are also linked because $S_{\mathbf{H},\mathbf{H}}^{(\kappa)} = S_{-\mathbf{H},-\mathbf{H}}^{(\kappa)}$ and $S_{\mathbf{0},\mathbf{H}}^{(\kappa)} = S_{\mathbf{0},-\mathbf{H}}^{(\kappa)}$. Similar relationships hold for pairs of incident orientations which lie on either side of centre of the band. This may be seen in Fig. 1(*a*) where $|\mathbf{Q}_1| = |\mathbf{Q}_2+\mathbf{H}|$ and $|\mathbf{Q}_2| = |\mathbf{Q}_1+\mathbf{H}|$. Equivalent results hold for Fig. 1(*b*), and between Figs. 1(*a*) and 1(*b*). The results derived below for a single symmetrical orientation therefore apply equally well to a convergent-

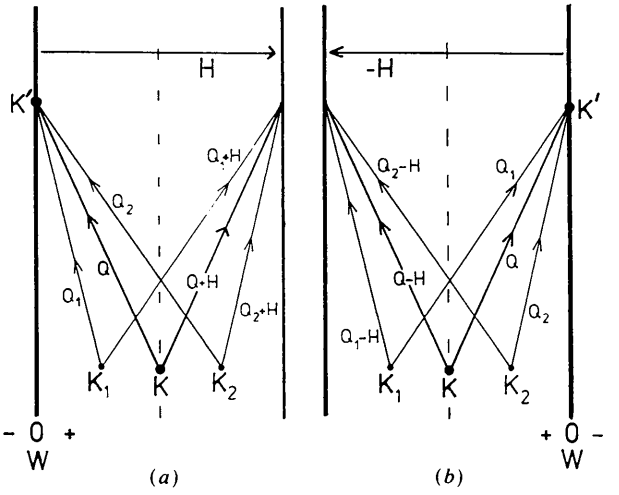

Fig. 1. Two-beam Kikuchi band geometry for reflections ±**H**. For clarity, the formation of the left-hand (*a*) and right-hand (*b*) lines are shown separately. **K'** is the final orientation (*i.e.* observation point). **K** represents a symmetrical incident orientation and $\mathbf{K}_1$ and $\mathbf{K}_2$ represent incident orientations on either side of the centre of the band (dashed line). The **Q**'s are parallel components of the scattering vector. See text for details.

beam probe which lies exactly in the centre of the band (*i.e.* is on-axis with respect to the band).

The thick crystal limit, $\Delta t \gg 1$ or $t \gg \xi$, shows how the familiar independent-Bloch-wave result for two-beam Kikuchi lines (*e.g.* Ohtsuki, 1983) is contained in (4). (We again emphasize that t cannot be so large that multiple scattering takes effect.) The fourth term goes to zero and the t-dependent parts of the first three tend towards 1 (Fig. 2). In an asymmetric geometry the first and second terms give rise to a bright-line/dark-line pair, while in the symmetric case only the third term remains and gives a bright/dark profile for each line (Fig. 2*b*), with a mirror symmetry linking the pair of lines. Whether the lines have their bright or dark part towards the centre of the band depends on the sign of the $\sum_\kappa \cos(\varphi + \mathbf{H}.\mathbf{r}_\kappa) S^{(\kappa)}_{0,\mathbf{H}}(\mathbf{q})$ factor. For large-angle scattering $S^{(\kappa)}$ is positive, and for a primitive lattice $(\varphi + \mathbf{H}.\mathbf{r}_\kappa) = 0$. In this case the Kikuchi line profile is given by $+UW/(U^2+W^2)$, which is bright towards the centre of the band.

The thin crystal limit, $t \ll \xi$, shows how the Kikuchi pattern emerges from the uniform diffuse background of kinematic theory. At the exact Bragg position the $1-[\sin(\Delta t)/\Delta t]$ factors in the first three terms of (4) go like $\frac{2}{3}(\pi t/\xi)^2 + O(t/\xi)^4$, while the fourth term goes as $\pi t/\xi - \frac{1}{3}(\pi t/\xi)^3 + O(t/\xi)^5$. The fourth term therefore makes a significant contribution in a dependent-Bloch-wave model *and is the leading orientation-dependent term in thin crystals* (see also Fig. 2). This term has two other interesting properties. First, it is non-zero only for non-centrosymmetric crystals. This follows because in the centrosymmetric case, with the origin at the inversion centre, all phases φ are either 0 or π and the contributions of the atoms at $\pm\mathbf{r}_\kappa$ cancel. Second, it gives rise to a bright-line/dark-line pair of Kikuchi lines, even in the symmetrical scattering geometry. This may be seen by putting $\mathbf{H} \to -\mathbf{H}$ and $\varphi \to -\varphi$ in which case the $\sin(\varphi + \mathbf{H}.\mathbf{r}_\kappa)$ term changes sign. The mirror linking the pair of Kikuchi lines is therefore broken – as it is in the case of X-ray production discussed by Taftø (1983).

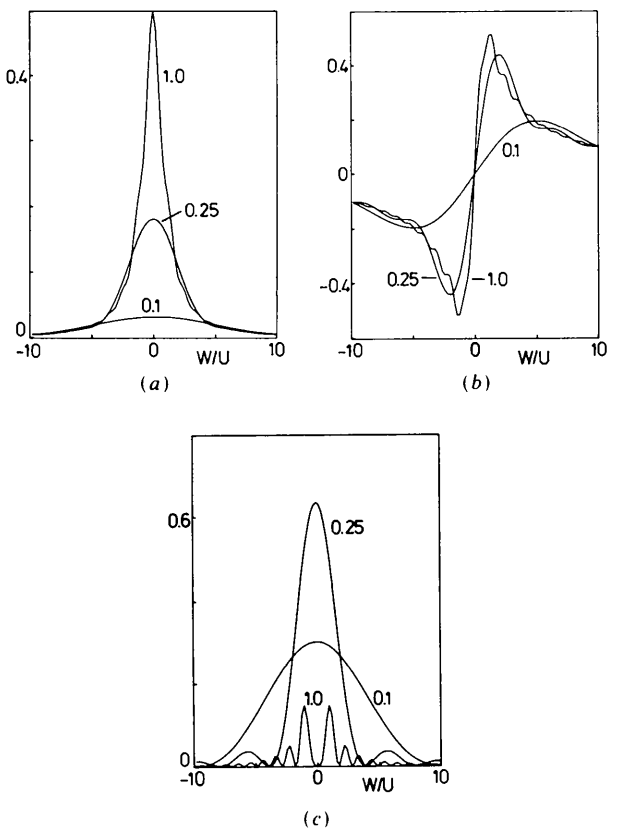

Fig. 2. The orientation dependent parts of the (*a*) second, (*b*) third and (*c*) fourth terms of equation (4), for thicknesses t/ξ = 0·1, 0·25 and 1·0. Note the different scales in (*a*), (*b*) and (*c*).

In the symmetrical geometry the overall form of two-beam Kikuchi lines is therefore controlled by a competition between the third and fourth terms of (4). In centrosymmetric crystals the fourth term is absent and the two lines on either side of the band are mirror related. In non-centrosymmetric crystals this mirror is broken. The degree of asymmetry depends on the relative sizes of the structure-dependent terms and on thickness because the effect of the fourth term decreases with increasing t. In order to observe the asymmetry it is therefore best to work with the weakest Kikuchi lines visible in a pattern. These have correspondingly large two-beam extinction distances and will satisfy the dependent-Bloch-wave criterion $t \ll \xi$. In principle this asymmetry may be used to determine the polarity of any non-centrosymmetric crystal because if a bright-line/dark-line pair is observed a calculation of the sign of $\sum_\kappa \sin(\varphi + \mathbf{H}.\mathbf{r}_\kappa) S^{(\kappa)}$ will enable $+\mathbf{H}$ to be distinguished from $-\mathbf{H}$. In practice, a number of other factors must be taken into account. First, it is important to avoid regions where the chosen Kikuchi lines intersect other lines. Although these might show pronounced asymmetries the two-beam model is inadequate to describe them and the simple expressions of (4) cannot reliably be used. Second, the incident beam must be accurately on-axis because the first two terms of (4) give a similar bright-line/dark-line asymmetry in the off-axis case. This effect should not be too important for the weakest lines because its leading term goes as $(t/\xi)^2$, while the structure-dependent asymmetry is of order (t/ξ). Finally, the effects of a tilted or wedged crystal have not been considered. These are difficult to include in the theory but it is expected that the usual result, that small tilts do not significantly affect diffraction calculations, will hold (*e.g.* Gjønnes & Gjønnes, 1985).

We have seen that the quantity $(\varphi + \mathbf{H}.\mathbf{r}_\kappa)$ plays a vital role in the theory. It is through this term that the phase dependence of the Kikuchi pattern arises. As in the usual elastic diffraction theory, phase information can only be carried in the form of phase-

invariant quantities which are independent of the choice of origin. $(\varphi+\mathbf{H}.\mathbf{r}_\kappa)$ satisfies this condition, but it takes a very different form from the invariants found in elastic diffraction, the simplest of which involves a sum of three phases (*e.g.* Bird, James & Preston, 1987). $(\varphi+\mathbf{H}.\mathbf{r}_\kappa)$ arises naturally in inelastic scattering theory because many such processes are localized about the atomic sites $\mathbf{r}_\kappa$. It follows that in addition to thermal diffuse scattering, X-ray production and energy loss spectroscopy, phase-dependent two-beam effects should also be present in, for example, backscattered and channelling patterns (Marthinsen & Høier, 1986; Marthinsen, Anisdahl & Høier, 1987). Throughout the paper we have referred to our analysis being a two-beam theory, and in this context $(\varphi+\mathbf{H}.\mathbf{r}_\kappa)$ might be called a two-beam phase invariant. This, however, may be a little misleading.* Without an incident beam there could be no 'two-beam' Kikuchi pattern formation, so in this sense ours is a three-beam theory (one incident and two scattered beams), even though the incident beam is treated on a very different footing from the scattered beams and plays no significant role in the final results. Looked at this way, our results do not break the standard rule from elastic diffraction theory, that at least three beams are required to produce phase-dependent quantities.

4. Concluding remarks

The basic results of this paper are (2) and (4) which give the intensity distribution in a Kikuchi pattern. Both expressions show how structural information is carried in the pattern, provided a dependent-Bloch-wave theory is used. Equation (2) is valid in a general diffraction situation [with possible corrections from (5) and for absorption], such as the central region of a strong band. The weakest lines in the pattern may be analysed using the two-beam result (4). In a second paper (Bird & Wright, 1989) computational results based on (2) and (4) are presented for crystals with the non-centrosymmetric GaAs structure and comparison is made between theory and experimental patterns.

* We are grateful to a referee for pointing this out.

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The Possible Translational Parts of $(3+d)$ Superspace Symmetry Operations

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Abstract

A necessary condition for possible translational parts of $(3+d)$ superspace symmetry operations is derived. The general conditions are discussed especially for $(3+1)$ superspace symmetry operations and some examples illustrate the application.

1. Introduction

An electron density function of incommensurate crystals with an internal dimensionality d can be described as a periodic function $\tilde{\rho}$ in $(3+d)$-dimensional space (de Wolff, 1974). The translational periodicity in $(3+d)$ superspace is characterized by

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