References

ALEXANDER, L. E. & SMITH, G. S. (1962). *Acta Cryst.* **15**, 983-1004.
ALLISON, S. K. & WILLIAMS, J. H. (1930). *Phys. Rev.* **35**, 149-154.
CALVERT, L. D., KILLEAN, R. C. G. & MATHIESON, A. MCL. (1975). *Acta Cryst.* A**31**, 855-856.
CALVERT, L. D., KILLEAN, R. C. G. & MATHIESON, A. MCL. (1976). *Acta Cryst.* A**32**, 648-652.
COMPTON, A. H. & ALLISON, S. K. (1935). *X-rays in Theory and Experiment.* New York: Van Nostrand.
COOPER, M. J. & NATHANS, R. (1967). *Acta Cryst.* **23**, 357-367.
COOPER, M. J. & NATHANS, R. (1968). *Acta Cryst.* A**24**, 481-484; 619-624.
DUISENBERG, A. J. M. (1983). *Acta Cryst.* A**39**, 211-216.
LADELL, J. & SPIELBERG, N. (1966). *Acta Cryst.* **21**, 103-118.
MATHIESON, A. MCL. (1968). *Rev. Sci. Instrum.* **39**, 1834-1837.
MATHIESON, A. MCL. (1978). *Acta Cryst.* A**34**, 404-406.
MATHIESON, A. MCL. (1982). *Acta Cryst.* A**38**, 378-387.
MATHIESON, A. MCL. (1983). *J. Appl. Cryst.* **16**, 572-573.
MATHIESON, A. MCL. (1984*a*). *Acta Cryst.* A**40**, 355-363.
MATHIESON, A. MCL. (1984*b*). *Aust. J. Phys.* **37**, 55-61.
SCHOENBORN, B. P. (1983). *Acta Cryst.* A**39**, 315-321.
WERNER, S. A. (1971). *Acta Cryst.* A**27**, 665-669.
WERNER, S. A. (1972). *Acta Cryst.* A**28**, 143-151.
WILLIS, B. T. M. (1960). *Acta Cryst.* **13**, 763-766.

Acta Cryst. (1985). A**41**, 316-320

Lattice-Dynamical Calculation of Second-Order Thermal Diffuse Scattering in Molecular Crystals

BY A. CRIADO,* A. CONDE AND R. MÁRQUEZ

Departamento de Optica y Sección de Física del Centro Coordinado del CSIC, Universidad de Sevilla, Sevilla, Spain

(*Received* 27 *June* 1984; *accepted* 30 *November* 1984)

Abstract

A computer procedure has been developed to calculate second-order thermal diffuse scattering (TDS) intensity for molecular crystals from lattice-dynamical calculations with an atom–atom potential in the Born–von Kármán formalism. It is applied to monoclinic phenothiazine and different contributions to second-order TDS intensity, acoustic–acoustic, acoustic–optic and optic–optic, are compared. Calculations are also performed in the long-wave approximation allowing for dispersion (LWD) and correction factors of Bragg intensities due to TDS contribution in the LWD approximation are, generally but not always, lower than lattice-dynamical ones; the ratio between LWD and 'exact' factors ranges from 0·4 to 1·4 for reflections considered.

Introduction

In a previous paper (Criado, Conde & Márquez, 1985) we reported a computational procedure to calculate first-order thermal diffuse scattering (TDS) intensity from a lattice-dynamical point of view, using the external Born–von Kármán formalism within the harmonic approximation and a potential function in a pairwise form, where each pair contribution adopts the form: $V(r) = -A/r^6 + B\exp(-Cr)$; A, B and C are constants empirically adjustable. This method was applied to monoclinic phenothiazine, where a proposed potential function model had been successful for calculating the thermal crystallographic parameters (Criado, Conde & Márquez, 1984) and the contribution of inelastic TDS intensity to Bragg reflections measured on a diffractometer by calculating the first-order correction factors for different reflections. Further calculations revealed the influence of first-order TDS intensity over electronic density maps and structural parameters obtained in least-squares refinements. The long-wave limit (Born & Huang, 1968) allowing for dispersion of the acoustic mode frequencies was found to be quite suitable when calculating correction factors because inside the small volumes used to scan the Bragg intensity around a reciprocal-lattice point this limit is usually a good approximation.

Most of the existing programs that correct measured Bragg intensities for thermal diffuse scattering effects (Helmholdt & Vos, 1977; Walker & Chipman, 1970) use the long-wave (LW) approximation taking as the starting point the elastic constants of the crystal and calculating the frequencies and polarization vectors from them, and only very recently (Helmholdt, Braam & Vos, 1983) has the dispersive character of the acoustic modes been taken into account (LWD approximation) using acoustic frequencies obtained from lattice-dynamical calculations.

For the second-order TDS contribution, less effort has been devoted to it, mainly because of the huge amount of computational time required. The programs that consider it adopt the LW approximation (Stevens, 1974) and, in order to reduce the computing

* This work forms part of the Doctoral Thesis.

0108-7673/85/040316-05$01.50

time, a second approximation upon the first consisting of a theoretical evaluation of the Brillouin zone integral (Ramachandran & Wooster, 1951). The question of the reliability of the LW approximation in the calculation of the second-order TDS intensity remains open and, if we consider that second-order TDS intensity increases its relative importance with respect to first order for high-order reflections, its contribution may be important when measuring high-order data, necessary in high-resolution density studies, and therefore an exact evaluation of second-order intensity is desirable.

We have tried in this work to perform a lattice-dynamical calculation of second-order TDS intensity in order to compare it with that obtained in the long-wave approximation, and to study the changes introduced in Bragg correction factors when we adopt this approximation.

Basic theory

The expression of the second-order TDS intensity at a point of the reciprocal space $\mathbf{S}=\mathbf{G}-\mathbf{q}$, where $\mathbf{G}$ is a reciprocal-lattice point (Cochran, 1963; Maradudin, Montroll, Weiss & Ipatova, 1971) is given by

$$\mathrm{d}I_2(\mathbf{S}=\mathbf{G}-\mathbf{q})/\mathrm{d}\mathbf{q} = \frac{S^2}{2}\sum_{\mathbf{q}'}\sum_{j'}\sum_{j''}\frac{E_{j'}(\mathbf{q}')E_{j''}(\mathbf{q}'')}{\omega_{j'}^2(\mathbf{q}')\omega_{j''}^2(\mathbf{q}'')}|f_2(\mathbf{S},\mathbf{q}'j'\mathbf{q}''j'')|^2,$$

where $\mathbf{S}$ is the dispersion vector, $\mathbf{q}=\mathbf{q}'+\mathbf{q}''$, $\omega_j(\mathbf{q})$ is the angular frequency of mode $(\mathbf{q}j)$ and j labels the different modes with the same wave vector $\mathbf{q}$. $E_j(\mathbf{q})$ is the energy of mode $(\mathbf{q}j)$, proportional to K_BT for high temperatures, $\mathbf{q}'$ runs over all allowed wave vectors inside the Brillouin zone, and $F_2(\mathbf{S},\mathbf{q}'j'\mathbf{q}''\mathbf{j}'')$ is the second-order structure factor, which, for a molecular crystal with six degrees of freedom per molecule, and neglecting the internal mode contribution, takes the form

$$\begin{aligned}F_2(\mathbf{S},\mathbf{q}'j'\mathbf{q}''j'') &= \sum_k\sum_i\{f_{ki}(\mathbf{S})T_{ki}(\mathbf{S})\mathbf{s}\,.\,[\mathbf{e}^t(\mathbf{q}'|kj')+\mathbf{e}^r(\mathbf{q}'|kj')\times\mathbf{x}(ki)]\\ &\times\mathbf{s}\,.\,[\mathbf{e}^t(\mathbf{q}''|kj'')+\mathbf{e}^r(\mathbf{q}''|kj'')\times\mathbf{x}(ki)]\\ &\times\exp[i\mathbf{S}\,.\,\mathbf{x}(ki)]\exp[i\mathbf{G}\,.\,\mathbf{x}(k)]\},\end{aligned}$$

where k and i label different molecules in the unit cell and different atoms in each molecule, $f_{ki}(\mathbf{S})$ is the atomic scattering factor of atom ki, $T_{ki}(\mathbf{S})$ is the temperature factor, $\mathbf{s}$ is a unit vector along the direction of $\mathbf{S}$, $\mathbf{x}(k)$ is the position vector of the centre of mass of molecule k and $\mathbf{x}(ki)$ is the position vector of atom i belonging to molecule k, with respect to the principal inertia axes. $\mathbf{e}^t(\mathbf{q}|kj)$ and $\mathbf{e}^r(\mathbf{q}|kj)$ are the translational and rotational components, respectively, relative to molecule k, of the polarization vectors of the mode $(\mathbf{q}j)$ and can be obtained by diagonalization of the dynamical matrix $D(\mathbf{q})$ (Born & Huang, 1968), calculated using mass-weighted external coordinates and principal inertia axes, and a further mass-unweighting of eigenvectors.

If we adopt the LW approximation for the acoustic modes, neglecting the contribution of the optic modes, the expression of second-order TDS intensity takes the following form:

$$\frac{\mathrm{d}I_2(\mathbf{S}=\mathbf{G}-\mathbf{q})}{\mathrm{d}\mathbf{q}} = \frac{S^4}{2}|F(\mathbf{G})|^2\sum_{\mathbf{q}'}\sum_{j'ac}\sum_{j''ac}\left\{\frac{E_{j'}(\mathbf{q})E_{j''}(\mathbf{q}'')}{\omega_{j'}^2(\mathbf{q}')\omega_{j''}^2(\mathbf{q}'')}\times[\mathbf{s}\,.\,\mathbf{e}^{\mathrm{LW}}(\mathbf{q}'j')\mathbf{s}\,.\,\mathbf{e}^{\mathrm{LW}}(\mathbf{q}''j'')]^2\right\}$$

and second-order TDS intensity turns out to be proportional to $|F(\mathbf{G})|^2$, where $F(\mathbf{G})$ is the Bragg structure factor.

Method of calculation

The computer program reported by us to calculate first-order TDS intensity (Criado, Conde & Márquez, 1985) has been improved in order to allow the calculation of second-order TDS intensity. The calculation of the intensity at a point $\mathbf{S}=\mathbf{G}-\mathbf{q}$ of the reciprocal space requires a sum over all pairs of modes $(\mathbf{q}'j')$ and $(\mathbf{q}''j'')$ such that $\mathbf{q}=\mathbf{q}'+\mathbf{q}''$, but the expression for $I_2(\mathbf{S}=\mathbf{G}-\mathbf{q})$ is symmetric in $\mathbf{q}'$ and $\mathbf{q}''$, therefore we may save computing time if we calculate only once the contributions arising from pairs $\mathbf{q}'$ and $\mathbf{q}''$. For a given $\mathbf{q}$, contributions arising from pairs $\mathbf{q}'$ and $\mathbf{q}''$ such that $\mathbf{q}'\simeq 0$ and $\mathbf{q}''\simeq\mathbf{q}$ present the same problems as those encountered in the calculation of thermal crystallographic parameters: $\omega_{j'}(\mathbf{q}')$ tends to zero rapidly and the numerical integration fails. To calculate this contribution, where the rest of the factors are roughly constant as $\mathbf{q}'\to 0$, we have utilized the method of Kroon & Vos (1978) consisting of an analytical evaluation of this contribution, also used by us in the calculation of thermal parameters in phenothiazine (Criado, Conde & Márquez, 1984), where further details can be found.

We have also programmed the LW approximation allowing for dispersion, neglecting the rotational components of the polarization vectors and using the frequency values given by lattice-dynamical calculations.

The practical way of performing Brillouin sums is as follows: for a given $\mathbf{q}$, $\mathbf{q}'$ is allowed to run through the Brillouin zone (BZ) and $\mathbf{q}''$ is calculated as $\mathbf{q}''=\mathbf{q}-\mathbf{q}'$. If $\mathbf{q}''$ lies outside the BZ, it is reduced by adding a suitable vector $\mathbf{G}$ so as to lie inside, and a selection procedure prevents the contribution of a reduced couple $\mathbf{q}'$, $\mathbf{q}''$ lying inside the BZ from being summed twice.

This method is equivalent to that used frequently, consisting of a calculation of BZ overlapping integrals

related to different reciprocal-lattice points, and furnishes all the possible couples of phonons contributing to the second-order TDS intensity.

Results

When we calculated first-order intensity, the Brillouin zone was divided into volume elements (25 divisions along each reciprocal basic vector) and the corresponding eigenvalues and eigenvectors obtained by diagonalization of the dynamical matrix were stored and used for all further calculations. Nevertheless, this density of sampling turned out to be impracticable when we proceeded to calculate second-order TDS intensity owing to the large amount of time. Because of this we have chosen a thicker mesh consisting of 13 divisions along each reciprocal vector, based on the fact that a similar sampling density has been sufficient when performing Brillouin sums in order to calculate thermodynamic properties and thermal crystallographic parameters (Criado, Conde & Márquez, 1984) and on the smoother character of the second-order TDS with respect to the first-order one (Pawley, 1969). In Fig. 1 we show thermal diffuse scattering intensity along $\mathbf{b}^*$ calculated for monoclinic phenothiazine at 300 K with this sampling density where we observe divergent behaviour of the intensity over the reciprocal-lattice points owing to the contribution of the acoustic modes, except for the systematic absences $(0k0)$: $k=2n+1$), where the second-order TDS structure factor corresponding to acoustic pairs such that $\mathbf{q}'\simeq-\mathbf{q}''$ and $|\mathbf{q}'|\ll$, $|\mathbf{q}''|\ll$ tends to zero cancelling the divergence produced by $\omega_{j'}^{-2}(\mathbf{q}')\omega_{j''}^{-2}(\mathbf{q}'')$. If we compare Fig. 1 with Fig. 2, showing the calculated first-order TDS intensity (Criado, Conde & Márquez, 1985), we observe that, in the range represented, the fall of the intensity with the scattering angle is smaller because the factor S^4 cancels in part this effect and, for high-angle values, second-order contribution equals first-order one.

When we compare the 'exact' (EX) lattice-dynamical calculations, performed with all the dispersion branches, with the long-wave approximation allowing for dispersion (LWD), where only three acoustic branches have been taken into account, we can observe that for points far from the reciprocal-lattice points the LWD approximation gives a systematic low contribution, undoubtedly due to the optic branches that have not been taken into account in this approximation. For points close to the reciprocal-lattice points the LWD approximation tends to fit the 'exact' values, indicating that optical contribution is less important because of the difference in frequency values and, on the other hand, that LWD approximation of acoustic modes is more accurate.

Nevertheless, even when the behaviour of both calculations close to the reciprocal-lattice points are very similar, the agreement is not so good as in the first-order case, where values coincide perfectly for points near reciprocal points. The reason for this discrepancy is obvious: when we calculate first-order TDS intensity at a point $\mathbf{S}=\mathbf{G}-\mathbf{q}$, only modes with wave vector $\mathbf{q}$ contribute, and if $\mathbf{q}$ is sufficiently small so that the long-wave limit is valid, both calculations will give identical results. On the contrary, when we calculate second-order TDS intensity, all pairs of vectors $\mathbf{q}'$ and $\mathbf{q}''$ such that $\mathbf{q}=\mathbf{q}'+\mathbf{q}''$ contribute and, even when $\mathbf{q}$ is small, contributions from modes through all the Brillouin zone will be present, and although those with small wave vector, where the LW approximation is valid, will contribute the most, those near the zone boundary will also have their contribution.

Bragg intensity second-order correction factors

From the results obtained above it must be expected that the LWD approximation will give correction factors for Bragg intensities $\alpha_2(\mathbf{G})$ that will reproduce the order of magnitude but not the correct values. To verify this conclusion we have calculated second-order TDS contribution to Bragg intensities using a constant symmetric volume for all reflections in a parallelepipedic form with edges equal to 3/13 of the corresponding basic vectors, and a volume of one more division along $\mathbf{a}^*$ to obtain the background contribution. Even with this low sampling density, the full lattice-dynamical calculation of correction

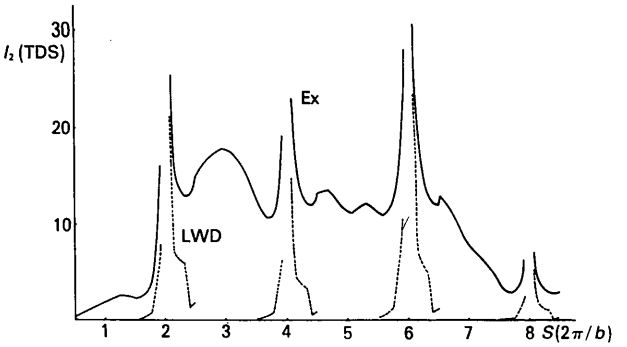

Fig. 1. Calculated second-order TDS intensity along $\mathbf{b}^*$.

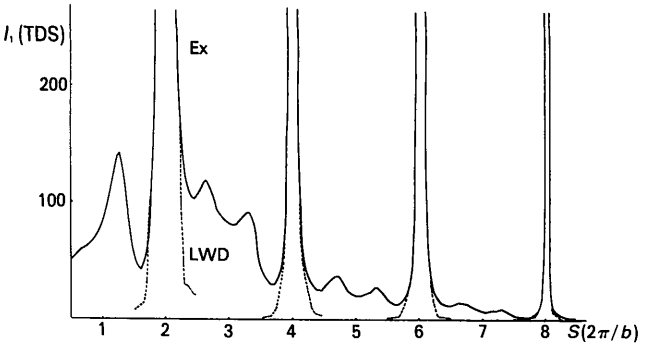

Fig. 2. Calculated first-order TDS intensity along $\mathbf{b}^*$.

factors $\alpha_2(\mathbf{G})$ for a given reflection takes an excessively long time, and for this reason we have tried to separate different contributions to second-order TDS intensity. The intensity can be divided into three parts: acoustic-acoustic, coming from phonon pairs $\mathbf{q}'$ and $\mathbf{q}''$, both acoustic; acoustic-optic, arising from acoustic and optic phonons; and optic-optic, which comes from two optic phonons. The three contributions to the second-order TDS intensity through a given reflection along $\mathbf{b}^*$ can be seen in Fig. 3, together with the result obtained in the LWD approximation and it may be observed that both acoustic-optic and optic-optic profiles are practically uniform through the diffraction peak and their contribution will cancel with background correction but, on the contrary, the acoustic-acoustic contribution shows non-uniform behaviour around the reciprocal-lattice points. Therefore, values for $\alpha_2(\mathbf{G})$ can be approximated taking into account only the three acoustic branches, both in the exact calculation and in the LWD approximation, reducing the computing time considerably.

In order to calculate the contribution of the volume element centred on the reciprocal-lattice points we have used again the method of Kroon & Vos (1978), calculating it as a function of the contributions of the surrounding elements, but now the functional form of second-order TDS intensity near the reciprocal points is smoother than the first-order one, it varies roughly as q^{-1} (Ramanchandran & Wooster, 1951) and the expression for the contribution of the $q=0$ element must be adjusted to this functional form. This contribution is

$$I(q=0)=\sum_a 0{\cdot}055I(a)+\sum_b 0{\cdot}09I(b)+\sum_c 0{\cdot}11I(c),$$

where a, b and c represent the different kinds of volume elements surrounding the $q=0$ element

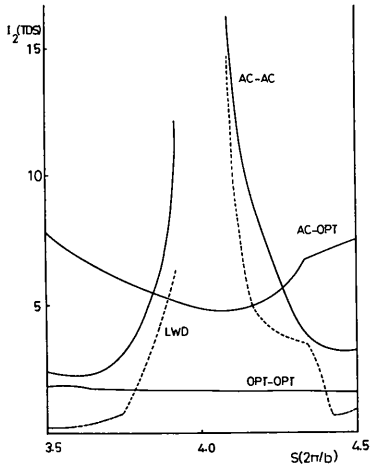

Fig. 3. Different contributions of second-order TDS intensity to a reflection along $\mathbf{b}^*$.

Table 1. *Bragg intensity correction factors in the 'exact' and in the LWD calculations defined as* $\alpha_2(\mathbf{G})=I_2(\mathbf{G})/I_{\text{Bragg}}(\mathbf{G})$

h	k	l	$\sin\theta/\lambda$(Å^{-1})	$\lvert F(\mathbf{G})\rvert^2$	$\alpha_2(\mathbf{G})$(%)	$\alpha_2^{\text{LWD}}(\mathbf{G})$(%)
1	1	0	0·1074	266·2	0·2	0·1
2	2	1	0·2282	39·31	3·4	1·4
−3	2	1	0·2555	147·3	5·1	5·7
0	3	1	0·2576	436·6	4·1	3·3
3	2	1	0·2756	60·91	5·8	3·3
4	0	1	0·2833	577·4	5·7	6·9
2	4	−4	0·3937	149·2	23·6	19·0
3	3	4	0·4037	61·61	21·4	13·4
6	1	1	0·4236	12·41	39·2	29·4
5	5	5	0·6260	0·05	141·6	74·3
−2	6	8	0·6296	1·331	175·5	139·5
−5	6	−3	0·6438	5·618	154·5	176·8
5	6	5	0·6857	1·251	187·0	105·0
6	6	6	0·7513	0·624	273·3	153·0
12	0	0	0·7981	0·0841	313·5	451·8
−8	7	7	0·8047	0·9212	467·4	568·0
7	7	7	0·8765	0·3885	476·1	282·2
7	8	6	0·9119	0·1041	618·4	328·2
0	11	7	0·9879	0·0189	930·4	677·8

(Kroon & Vos, 1978). As in the first-order case, an empirical parameter η that multiplies the $q=0$ contribution must be introduced to correct errors in numerical integration and it can be adjusted performing a numerical integration of the q^{-1} function over a sphere and calculating η in order to obtain the analytical value, yielding a value of $\eta=1{\cdot}78$.

In this way we have calculated second-order correction factors at 300 K for a set of reflections both in the 'exact' calculation and in the LWD approximation, which can be seen in Table 1. These values of $\alpha_2(\mathbf{G})$ must be understood only in order to compare the LWD approximation with the lattice-dynamical results, since the low sampling density used to scan the measuring volume produces numerical values considerably higher than their exact value, but, as the calculation conditions have been the same for both approximations, it may be considered as indicative of the reliability of the LWD approximation. It is our aim in future work to obtain an estimation of the real amount of second-order TDS contribution.

Discussion

The LWD approximation reproduces the order of magnitude but not the exact values as was the case for first-order correction factors and there is no systematic disagreement between the two methods. The differences can be considered random and depend on the different effects that neglecting rotational components of polarization vectors introduces, especially near the zone boundary. Because of this, in order to perform accurate measurements of high-order reflections it is advisable to adopt conditions (low temperature, principally) where second-order contributions may be minimized, since an exact TDS correction would involve a full lattice-dynamical treatment, difficult to perform, in general.

For the LW approximation, usually utilized in correction of Bragg intensities, it must be expected that its results will be worse than those of LWD. The LW approximation tends to increase the frequency mode values with respect to the real ones when we come out of the long-wave limit and this effect will be more important when calculating the background contribution, where the LW approximation is less valid, and therefore the tendency will be an overestimation of the net intensity calculated for the scanned volume with respect to LWD values, similar to the first-order case (Kroon & Vos, 1979).

We thank the Computing Centre of the University of Seville for facilities given to run these rather lengthy calculations and the Spanish Government, which has supported in part this work through the Comisión Asesora de Investigación Científica y Técnica.

References

BORN, M. & HUANG, K. (1968). *Dynamical Theory of Crystal Lattices.* Oxford: Clarendon.
COCHRAN, W. (1963). *Rep. Prog. Phys.* **26**, 1-45.
CRIADO, A., CONDE, A. & MÁRQUEZ, R. (1984) *Acta Cryst.* A**40**, 696-701.
CRIADO, A., CONDE, A. & MÁRQUEZ, R. (1985). *Acta Cryst.* A**41**, 158-163.
HELMHOLDT, R. B., BRAAM, A. W. M. & VOS, A. (1983). *Acta Cryst.* A**39**, 90-94.
HELMHOLDT, R. B. & VOS, A. (1977). *Acta Cryst.* A**33**, 38-45.
KROON, P. A. & VOS, A. (1978). *Acta Cryst.* A**34**, 823-824.
KROON, P. A. & VOS, A. (1979). *Acta Cryst.* A**35**, 675-684.
MARADUDIN, A. A., MONTROLL, E. W., WEISS, G. H. & IPATOVA, I. P. (1971). *Theory of Lattice Dynamics in the Harmonic Approximation.* New York: Academic Press.
PAWLEY, G. S. (1969). *Acta Cryst.* A**25**, 702-707.
RAMACHANDRAN, G. N. & WOOSTER, W. A. (1951). *Acta Cryst.* **4**, 335-344.
STEVENS, E. D. (1974). *Acta Cryst.* A**30**, 184-189.
WALKER, C. B. & CHIPMAN, D. R. (1970). *Acta Cryst.* A**26**, 447-455.

Acta Cryst. (1985). A**41**, 320-327

Interpretation of Dynamical Diffuse Scattering of Fast Electrons in Rutile

BY C. J. ROSSOUW

CSIRO Division of Chemical Physics, PO Box 160, *Clayton* 3168, *Australia*

AND L. A. BURSILL

School of Physics, University of Melbourne, Parkville 3052, *Australia*

(*Received* 15 *September* 1983; *accepted* 4 *December* 1984)

Abstract

An Einstein model for thermal diffuse scattering is extended to a fully dynamical *n*-beam Bloch-wave treatment, where explicit account is taken of the scattering site symmetry from individual atoms. Dynamical effects in this model are related to orientation-dependent fluctuations in current density on localized scattering centres within the crystal, yielding excess or deficit Kikuchi bands. Calculated diffuse scattering distributions are compared with experimental observations from rutile (TiO_2). The predicted diffuse distribution for scattering from oxygen sites correlates reasonably well with experiment, implying a relatively weak contribution for (localized) thermal diffuse scattering of fast electrons from titanium sites.

1. Introduction

The Einstein model for thermal diffuse scattering (TDS) lends itself to interpretation in terms of localized scattering centres within a unit cell. It has been shown that, for ionization events, a Bloch-wave formulation in describing the passage of a fast electron through a crystal can clearly predict the formation of excess or deficient bands in the inelastic beam, depending on (1) diffraction conditions for the elastic and inelastic beams, (2) scattering kinematics and (3) site of interaction within the crystal (Maslen & Rossouw, 1984; Rossouw & Maslen, 1984). In this paper we extend the Einstein model for TDS, developed by Hall & Hirsch (1965), to evaluate the scattering kinematics term in a dynamical *n*-beam Bloch-wave formulation. Computer simulations based on this theory are compared with the diffuse scattering observed from TiO_2 viewed down the *c* axis.

2. Theory

Hall & Hirsch (1965) derived a formula for the TDS intensity as a function of momentum transfer $\hbar\mathbf{q}$ to the crystal, using an Einstein model for uncorrelated thermal displacements of crystal atoms (here $\mathbf{q} = \mathbf{k} - \mathbf{k}'$, where $\mathbf{k}$ and $\mathbf{k}'$ are the wavevectors of the fast

0108-7673/85/040320-08$01.50