the respective procedure is to be applied. For either of the triples, one kind of quadruple corresponding to $b^{\to}d\ b^{\to}d$ and one kind of quadruple corresponding to $b^{\to}d\ b^{\leftarrow}d$ result, each leading to one only P/MDO polytype with $m = 2$ and $m = 4$, respectively.

It is remarkable that only three of the four MDO polytypes are amongst the frequently occurring polytypes listed in Table 2, but also two of the four only P/MDO polytypes (namely those with $m = 2$).

Concluding remarks

Although classification of polytypes of any family as MDO, only P/MDO or not P/MDO and deduction of all MDO polytypes and all only P/MDO polytypes is not too difficult, the explanation of the corresponding procedures may seem fairly involved. The reason is that by far the most OD crystals contain layers of not more than two different kinds, and for such families there is obviously only one kind of packet. Furthermore, by far the most OD crystals with $M > 1$ belong to category IV or category I – and treated as category III – there is thus only one position of d^1 relative to b^1 and for category IV also only one position of d^M relative to b^M, namely those leading together with b^1 or b^M to A^1 and A^M, respectively.

Although, as exemplified in Table 2, MDO and only P/MDO polytypes are not the only polytypes occurring in nature, they certainly seem to occur more frequently than others. In a number of polytype families they are indeed the only polytypes so far observed. Their knowledge may help for identifying the polytypes present (or mainly present) in a sample consisting of a multitude of crystals too small for single-crystal work, from powder diffraction diagrams. In this way they have already been used for the identification of vermiculite polytypes (Weiss & Ďurovič, 1980; Weiss, 1976).

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Dynamic Structure Factors for Excitations in Modulated Structures

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Abstract

A phenomenological Landau theory yields new excitations in incommensurate structures corresponding to a phase and amplitude fluctuation of the modulating function. The dynamic structure factors for the new excitation modes are calculated in harmonic approximation and the influence of phase and amplitude fluctuations is discussed.

1. Introduction

In recent years structures with a static periodical displacement of the atoms from their perfect lattice

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positions have been studied with rising interest. Since the wave vector $\mathbf{q}_0$ of the modulation cannot be represented by a simple rational fraction of a reciprocal-lattice vector, these structures are called incommensurate structures (for a review see Axe, 1976).

Overhauser (1971) proposed new excitations for these structures, so-called phasons, corresponding to a phase fluctuation of the static modulation wave. Owing to the incommensurability a uniform phase shift of the modulation wave is possible without any energy consumption, and therefore it is expected that the frequency of this new vibrational branch approaches zero at the site of the satellites. Quite recently, new excitations have been found in biphenyl (Cailleau, Moussa, Zeyen & Bouillot, 1980) and were explained by the authors by phasons, since they observed a linear dispersion relation originating from the satellite reflections.

A simple explanation for the new excitations within an incommensurate structure may be given with a phenomenological Landau theory (McMillan, 1975, 1976; Axe, 1976; Cowley & Bruce, 1978). Anharmonic terms in the free energy mix normal modes whose wave vectors differ by $\pm 2\mathbf{q}_0$, giving rise to new normal modes. They correspond to a fluctuation of the phase and amplitude of the static modulation wave:

$$\mathbf{u}_l = \mathbf{A} \sin(\mathbf{q}_0 \mathbf{r}_l + \Phi_0).$$

($\mathbf{r}_l$ is the position vector of the unit cell in a primitive Bravais lattice). The fluctuations $\delta\Phi_l$ and $\delta\mathbf{A}_l$ of phase and amplitude cause further atomic displacements:

$$\delta\mathbf{u}_l = \delta\mathbf{A}_l \sin(\mathbf{q}_0 \mathbf{r}_l + \Phi_0) + \mathbf{A}\,\delta\Phi_l \cos(\mathbf{q}_0 \mathbf{r}_l + \Phi_0). \quad (1)$$

Another *Ansatz* for the atomic displacements originating from the static modulation wave and the phase fluctuations was introduced by Overhauser (1971):

$$\mathbf{u}_l = \mathbf{A} \sin(\mathbf{q}_0 \mathbf{r}_l + \Phi_0 + \delta\Phi_l). \quad (2)$$

The displacements of the phase fluctuations in (1) and (2) coincide only for small fluctuations $\delta\Phi_l \ll 1$. The phenomenological Landau theory yields displacements corresponding only approximately to phase fluctuations of the static modulation wave. There are also consequences for the scattering effects. Whereas (2) leads to a rather simple **Q**-independent Debye–Waller factor for satellite reflections (Overhauser, 1971), the atomic displacements of (1) yield a Debye–Waller factor which is a complicated function of the scattering vector **Q** and the fluctuations $\delta\mathbf{A}_l$ and $\delta\Phi_l$ (Axe, 1980).

The purpose of this study is to show the influence of phase and amplitude fluctuations of the static displacement wave and the influence of 'normal' phonons on static and dynamic structure factors. It is anticipated that the fluctuations can be expanded into normal modes being all statistically independent from one another. The *Ansatz* for the atomic displacements is taken from the result of the phenomenological Landau theory [equation (1)]. We limit the calculation of the scattered intensities to the elastic structure factor and one-phonon processes. Multi-phonon processes are neglected.

2. Structure factors

In order to calculate the structure factors we first consider the scattered amplitude $F(\mathbf{Q})$, *i.e.* the Fourier transform of the electron density function. The equilibrium position $\mathbf{r}_{\kappa l}$ of the atom κ in the unit cell l is defined by the position vector $\mathbf{r}_l$ of the unit cell and the vector $\mathbf{r}_\kappa$, where $\mathbf{r}_{\kappa l} = \mathbf{r}_l + \mathbf{r}_\kappa$. The total displacement $\mathbf{u}_{\kappa l}$ from this equilibrium position consists of the fluctuation $\delta\mathbf{U}_{\kappa l}$ caused by normal phonons, the modulation wave

$$\mathbf{A}_\kappa \sin(\mathbf{q}_0 \mathbf{r}_{\kappa l} + \Phi_{\kappa 0}),$$

the fluctuation in phase

$$\delta\Phi_{\kappa l}\, \mathbf{A}_\kappa \cos(\mathbf{q}_0 \mathbf{r}_{\kappa l} + \Phi_{\kappa 0}),$$

and amplitude

$$\delta\mathbf{A}_{\kappa l} \sin(\mathbf{q}_0 \mathbf{r}_{\kappa l} + \Phi_{\kappa 0})$$

of the static modulation wave.

The instantaneous position of the atom κ within the unit cell l is therefore given by

$$\mathbf{r}_{\kappa l} + \mathbf{u}_{\kappa l} = \mathbf{r}_\kappa + \mathbf{r}_l + \delta\mathbf{U}_{\kappa l} + \mathbf{A}_\kappa \sin\theta_{\kappa l} + \delta\Phi_{\kappa l}\, \mathbf{A}_\kappa \cos\theta_{\kappa l} + \delta\mathbf{A}_{\kappa l} \sin\theta_{\kappa l}, \quad (3)$$

where $\theta_{\kappa l} = \mathbf{q}_0 \mathbf{r}_{\kappa l} + \Phi_{\kappa 0}$.

Introducing a Fourier expansion for the fluctuations we get

$$\delta\mathbf{U}_{\kappa l} = \sum_q \mathbf{U}_{q\kappa} \sin(\mathbf{q}\mathbf{r}_{\kappa l} + \chi_{q\kappa})$$

$$\delta\Phi_{\kappa l} = \sum_q \Phi_{q\kappa} \sin(\mathbf{q}\mathbf{r}_{\kappa l} + \varphi_{q\kappa})$$

$$\delta\mathbf{A}_{\kappa l} = \sum_q \mathbf{a}_{q\kappa} \sin(\mathbf{q}\mathbf{r}_{\kappa l} + \psi_{q\kappa}). \quad (4)$$

$\mathbf{U}_{q\kappa}$ is the amplitude of a normal phonon, $\Phi_{q\kappa}$ and $\mathbf{a}_{q\kappa}$ are the amplitudes of a phase modulation ('phason') and an amplitude modulation phonon ('amplitudon') respectively. For the following calculation it is anticipated that these modes are statistically independent.

The Fourier transform of the electron density function is

$$F(\mathbf{Q}) = \sum_l \sum_\kappa f_\kappa \exp[i\mathbf{Q}(\mathbf{r}_{\kappa l} + \mathbf{u}_{\kappa l}), \quad (5)$$

where f_κ = form factor of the κth atom.

In the following we consider the exponential term for the various contributions in (3).

2.1. *Elastic structure factor*

To calculate $F(\mathbf{Q})$ at $\mathbf{Q} = \mathbf{G} - m\mathbf{q}_0$ ($m = 0, \pm 1, \pm 2, \ldots, \{G\}$ = the total set of reciprocal-lattice vectors) we first transform the term $\exp[i\mathbf{Q}\mathbf{A}_\kappa \sin(\mathbf{q}_0\mathbf{r}_{\kappa l} + \Phi_{\kappa 0})]$ with the help of the Jakobi–Anger generating function for Bessel functions:

$$\exp(iz\sin\varphi) = \sum_{m=-\infty}^{+\infty} \exp(im\varphi) J_m(z).$$

Accordingly, we get

$$\exp[i\mathbf{Q}\mathbf{A}_\kappa \sin(\mathbf{q}_0\mathbf{r}_{\kappa l} + \Phi_{\kappa 0})] = \sum_m \exp[im(\mathbf{q}_0\mathbf{r}_{\kappa l} + \Phi_{\kappa 0})] \times J_m(\mathbf{Q}\mathbf{A}_\kappa). \quad (6)$$

For the moment we neglect all fluctuations and after summing over l we obtain from (5) and (6) the structure factor for the main and satellite reflections at $\mathbf{Q} = \mathbf{G} - m\mathbf{q}_0$:

$$F_m(\mathbf{Q}) = \delta(\mathbf{Q} + m\mathbf{q}_0 - \mathbf{G}) \times \sum_\kappa f_\kappa \exp[i(\mathbf{G}\mathbf{r}_\kappa + m\Phi_{\kappa 0})] J_m(\mathbf{Q}\mathbf{A}_\kappa).$$

In order to determine the influence of the fluctuations on the elastic structure factor we consider first the phonon term

$$\exp(i\mathbf{Q}\delta\mathbf{U}_{\kappa l}) = \exp\left\{i\mathbf{Q}\left[\sum_q \mathbf{U}_{q\kappa}\sin(\mathbf{q}\mathbf{r}_{\kappa l} + \chi_{q\kappa})\right]\right\}.$$

Using again the Jakobi–Anger function we may write

$$\exp(i\mathbf{Q}\delta\mathbf{U}_{\kappa l}) = \prod_q \left\{\sum_n J_n(\mathbf{Q}\mathbf{U}_{q\kappa}) \exp[in(\mathbf{q}\mathbf{r}_{\kappa l} + \chi_{q\kappa})]\right\}. \quad (7)$$

Since we are interested in the scattering at the sites $\mathbf{G} - m\mathbf{q}_0$ we may only use Bessel functions of zero order in (7), because in that case the term $\exp[in(\mathbf{q}\mathbf{r}_{\kappa l} + \chi_{q\kappa})] = 1$. For infinitesimal displacements $\mathbf{U}_{q\kappa}$ the function $J_0(z)$ may be developed logarithmically and we obtain

$$\ln J_0(z) \simeq \ln(1 - \tfrac{1}{4}z^2) \simeq -\tfrac{1}{4}z^2.$$

In this way (7) transforms to the well-known Debye–Waller factor for the atom κ:

$$\prod_q J_0(\mathbf{Q}\mathbf{U}_{q\kappa}) = \exp(-W_\kappa) \quad (8)$$

with

$$W_\kappa = \tfrac{1}{4}\sum_q (\mathbf{Q}\mathbf{U}_{q\kappa})^2.$$

Next we study the influence of the phase fluctuations of the modulation wave. With the help of the Jakobi–Anger function we obtain

$$\exp(i\mathbf{Q}\mathbf{A}_\kappa\,\delta\Phi_{\kappa l}\cos\theta_{\kappa l}) = \prod_q \left\{\sum_n \exp[in(\mathbf{q}\mathbf{r}_{\kappa l} + \varphi_{q\kappa})] \times J_n(\Phi_{q\kappa}\mathbf{Q}\mathbf{A}_\kappa\cos\theta_{\kappa l})\right\}. \quad (9)$$

In this case the Fourier expansion of $\delta\Phi_{\kappa l}$ was used according to (4). As for the phonons, we consider again only Bessel functions of zero order for the elastic structure factor. After a logarithmic expansion in analogy to (8), (9) can be transformed into

$$\exp\left\{-\tfrac{1}{4}\left[(\mathbf{Q}\mathbf{A}_\kappa)^2\sum_q \Phi_{q\kappa}^2\right]\cos^2\theta_{\kappa l}\right\} = \exp(-W_\kappa^\Phi)\exp(-W_\kappa^\Phi\cos 2\theta_{\kappa l}), \quad (10)$$

with

$$W_\kappa^\Phi = \tfrac{1}{8}(\mathbf{Q}\mathbf{A}_\kappa)^2\sum_q \Phi_{q\kappa}^2.$$

From (10) we see that the phase fluctuations cause a spatially modulated contribution with a wave vector $2\mathbf{q}_0$. As already thoroughly discussed by Axe (1980) this leads to a superposition of Bessel functions $J_m(\mathbf{Q}\mathbf{A}_\kappa)$ and $J_{m\pm 2}(\mathbf{Q}\mathbf{A}_\kappa)$ within the elastic structure factor.

The influence of the amplitude fluctuations of the modulation wave is given by the term

$$\exp(i\mathbf{Q}\delta\mathbf{A}_{\kappa l}\sin\theta_{\kappa l}) = \prod_q \left\{\sum_n \exp[in(\mathbf{q}\mathbf{r}_{\kappa l} + \psi_{q\kappa})] \times J_n(\mathbf{Q}\mathbf{a}_{q\kappa}\sin\theta_{\kappa l})\right\}. \quad (11)$$

In analogy to the phase fluctuations (11) becomes

$$\exp(-W_\kappa^a)\exp(W_\kappa^a\cos 2\theta_{\kappa l}), \quad (12)$$

with

$$W_\kappa^a = \tfrac{1}{8}\sum_q (\mathbf{Q}\mathbf{a}_{q\kappa})^2.$$

Owing to the similar spatial modulation we combine the phase and amplitude fluctuations and after multiplication of (10) and (12) we find

$$\exp(-w_\kappa^+)\exp(-w_\kappa^-\cos 2\theta_{\kappa l}), \quad (13)$$

where

$$w_\kappa^+ = W_\kappa^\Phi + W_\kappa^a \quad \text{and} \quad w_\kappa^- = W_\kappa^\Phi - W_\kappa^a.$$

By use of the modified Bessel function

$$I_m(z) = i^{-m} J_m(iz),$$

(13) may be expanded to

$$\exp(-w_\kappa^+)\sum_s (-1)^s I_s(w_\kappa^-)\exp(2is\theta_{\kappa l}).$$

Combining this result with (6) and (8) we obtain

$$\exp[i\mathbf{Q}(\mathbf{r}_{\kappa l}+\mathbf{u}_{\kappa l})]=\exp(-W_\kappa)\times\exp(-w_\kappa^+)\,B(\theta_{\kappa l})\,C(\theta_{\kappa l}),\quad(14)$$

with

$$B(\theta_{\kappa l})=\sum_m \exp(im\theta_{\kappa l})\,J_m(\mathbf{QA}_\kappa)$$

and

$$C(\theta_{\kappa l})=\sum_s(-1)^s I_s(w_\kappa^-)\exp(2is\theta_{\kappa l}).$$

If we rearrange the sums $B(\theta_{\kappa l})$ and $C(\theta_{\kappa l})$ and perform the total Fourier transform the elastic structure factor considering all fluctuations is at $\mathbf{Q}=\mathbf{G}-m\mathbf{q}_0$

$$F_m(\mathbf{Q})=\delta(\mathbf{Q}+m\mathbf{q}_0-\mathbf{G})\sum_\kappa f_\kappa\exp(-W_\kappa)\,T_m(\mathbf{QA}_\kappa)\times\exp[i(\mathbf{Gr}_\kappa+m\Phi_{\kappa 0})].\quad(15)$$

Compared to the non-modulated structure we have an additional 'temperature factor'

$$T_m(\mathbf{QA}_\kappa)=\exp(-w_\kappa^+)\sum_s(-1)^s I_s(w_\kappa^-)\,J_{m-2s}(\mathbf{QA}_\kappa)$$

describing the influence of the modulation wave as well as the phase and amplitude fluctuations.

2.2. *Dynamic structure factor*

In the following we calculate the intensity of the diffuse scattering caused by the fluctuations (4).

Phonons. For the structure factor of a given phonon with the wave vector $\mathbf{q}$, we use the corresponding first-order Bessel function from (7)

$$J_1(\mathbf{QU}_{q\kappa})\simeq\tfrac{1}{2}(\mathbf{QU}_{q\kappa}),$$

and taking into account the fluctuations of all other phonons we obtain for (7):

$$\tfrac{1}{2}(\mathbf{QU}_{q\kappa})\exp[i(\mathbf{qr}_{\kappa l}+\chi_{q\kappa})]\exp(-W_\kappa).$$

In the Fourier transform of (5) we must consider the additional phase term $(\mathbf{qr}_{\kappa l}+\chi_{q\kappa})$ and with (15) the one-phonon structure factor at the site of $\mathbf{Q}=\mathbf{G}-m\mathbf{q}_0-\mathbf{q}$ is found to be

$$F_m^U(\mathbf{Q})=\delta(\mathbf{Q}+\mathbf{q}+m\mathbf{q}_0-\mathbf{G})\times\sum_\kappa f_\kappa\exp(-W_\kappa)\,T_m^U(\mathbf{QA}_\kappa)\times\frac{1}{2}\left(\frac{\bar U_{q\kappa}}{\bar A_\kappa}\right)\exp[i(\mathbf{Gr}_\kappa+m\Phi_{\kappa 0}+\chi_{q\kappa})].\quad(16)$$

$\bar U_{q\kappa}$ and $\bar A_\kappa$ are the components of $\mathbf{U}_{q\kappa}$ and $\mathbf{A}_\kappa$ along the scattering vector $\mathbf{Q}$, and using (15) the function T_m^U is given by

$$T_m^U=(\mathbf{QA}_\kappa)\,T_m.$$

Phasons. Similar to the phonons we examine in (9) a phase modulation phonon with a wave vector $\mathbf{q}$. Again we use the first-order Bessel function for the given phason and the phase fluctuation caused by all other phasons is taken into account by the Debye–Waller factor given in (10). Then the appropriate term for the one-phason process is

$$J_1(\Phi_{q\kappa}\mathbf{QA}_\kappa\cos\theta_{\kappa l})\exp[-W_\kappa^\Phi(1+\cos 2\theta_{\kappa l})]\times\exp[i(\mathbf{qr}_{\kappa l}+\varphi_{q\kappa})].\quad(17)$$

Compared to the phonons also the argument of the Bessel function has a spatial modulation. For small $\Phi_{q\kappa}$, (17) is transformed into

$$D(\theta_{\kappa l})\exp(-W_\kappa^\Phi)\exp(-W_\kappa^\Phi\cos 2\theta_{\kappa l}),$$

where

$$D(\theta_{\kappa l})=\tfrac{1}{2}\Phi_{q\kappa}\exp[i(\mathbf{qr}_{\kappa l}+\varphi_{q\kappa})]\,(\mathbf{QA}_\kappa/2)\times[\exp(i\theta_{\kappa l})+\exp(-i\theta_{\kappa l})].\quad(18)$$

In comparison to (10) we obtain an additional factor $D(\theta_{\kappa l})$, that is to say, in order to calculate the phason structure factor, the right-hand side of (14) must simply be multiplied by $D(\theta_{\kappa l})$.

Accordingly, we obtain

$$\exp[i\mathbf{Q}(\mathbf{r}_{\kappa l}+\mathbf{u}_{\kappa l})]=\exp(-W_\kappa)\exp(-w_\kappa^+)\,B(\theta_{\kappa l})\times C(\theta_{\kappa l})\,D(\theta_{\kappa l}).$$

The sums $B(\theta_{\kappa l})$, $C(\theta_{\kappa l})$, and $D(\theta_{\kappa l})$ are rearranged relative to a uniform phase $\theta_{\kappa l}$. By summing over l we obtain the phason structure factor

$$F_m^\Phi(\mathbf{Q})=\delta(\mathbf{Q}+\mathbf{q}+m\mathbf{q}_0-\mathbf{G})\times\sum_\kappa f_\kappa\exp(-W_\kappa)\,T_m^\Phi(\mathbf{QA}_\kappa)\,\Phi_{q\kappa}/2\times\exp[i(\mathbf{Gr}_\kappa+m\Phi_{\kappa 0}+\varphi_{q\kappa})],\quad(19)$$

with

$$T_m^\Phi(\mathbf{QA}_\kappa)=\tfrac{1}{2}(\mathbf{QA}_\kappa)\exp(-w_\kappa^+)\times\sum_s(-1)^s I_s(w_\kappa^-)\{J_{m-2s-1}(\mathbf{QA}_\kappa)+J_{m-2s+1}(\mathbf{QA}_\kappa)\}.$$

Owing to the relation

$$J_{m-1}(z)+J_{m+1}(z)=(2m/z)\,J_m(z),$$

T_m^Φ can be transformed to

$$T_m^\Phi(\mathbf{QA}_\kappa)=\exp(-w_\kappa^+)\times\sum_s(-1)^s I_s(w_\kappa^-)(m-2s)J_{m-2s}(\mathbf{QA}_\kappa)$$

It is interesting to compare $F_m^\Phi(\mathbf{Q})$ with the elastic structure factor for the case $w_\kappa^- = 0 = W_\kappa^\Phi - W_\kappa^a$. Then we find

$$T_m^\Phi = mT_m. \quad (20)$$

In this case of equal amplitude and phase fluctuations, phasons cannot be observed at main reflections.

Amplitudons. To derive the structure factor for an amplitude modulation phonon an analogous way as in the foregoing section must be chosen.

For a given amplitude phonon with the wave vector $\mathbf{q}$, (11) becomes

$$\exp(i\mathbf{QA}_{\kappa l}\sin\theta_{\kappa l}) = E(\theta_{\kappa l})\exp(-W_\kappa^a) \times \exp(W_\kappa^a\cos 2\theta_{\kappa l}),$$

with

$$E(\theta_{\kappa l}) = \tfrac{1}{2}\mathbf{Qa}_{q\kappa}\exp[i(\mathbf{qr}_{\kappa l} + \psi_{q\kappa})] \times \frac{i}{2}[\exp(-i\theta_{\kappa l}) - \exp(i\theta_{\kappa l})],$$

and the structure factor for amplitude modes is

$$F_m^a(\mathbf{Q}) = \delta(\mathbf{Q} + \mathbf{q} + m\mathbf{q}_0 - \mathbf{G}) \times \sum_\kappa f_\kappa \exp(-W_\kappa)\, T_m^a(\mathbf{QA}_\kappa)\tfrac{1}{2}(\bar{a}_{q\kappa}/\bar{A}_\kappa) \times \exp[i(\mathbf{Gr}_\kappa + m\Phi_{\kappa 0} + \psi_{q\kappa})], \quad (21)$$

with

$$T_m^a(\mathbf{QA}_\kappa) = \frac{i}{2}(\mathbf{QA}_\kappa)\exp(-w_\kappa^+)\sum_s (-1)^s I_s(w_\kappa^-) \times \{J_{m-2s+1}(\mathbf{QA}_\kappa) - J_{m-2s-1}(\mathbf{QA}_\kappa)\}.$$

$\bar{a}_{q\kappa}$ is the component of $\mathbf{a}_{q\kappa}$ along $\mathbf{Q}$. Contrary to the phasons, amplitudons are observable also for $w_\kappa^- = 0$ at main reflections.

Scattering law. So far we have calculated the elastic structure factor $F(\mathbf{Q})$ and the scattered amplitudes for the various modes [equations (16), (19), and (21)], disregarding the time dependence of the modes in (4). This is possible for X-ray scattering owing to the energy integration of the scattered radiation.

But it is not possible for neutron scattering, where we must analyse the coherent elastic cross section

$$\frac{d^2\sigma}{d\Omega\, d\omega} = \frac{k'}{2\pi k} S(\mathbf{Q}, \omega),$$

where k and k' are the initial and final wave vectors. The scattering law $S(\mathbf{Q},\omega)$ is defined by the Fourier transform of the correlation function $G(\mathbf{r},t)$ in space and time. As we have already performed the Fourier transform in space in the foregoing sections we obtain for

$$S(\mathbf{Q},\omega) = \int dt \exp(-i\omega t)\langle F(-\mathbf{Q},0)\,F(\mathbf{Q},t)\rangle_T. \quad (22)$$

The time dependence of the structure factor is easily introduced by substituting for all phases in (4)

$$\chi_{q\kappa} \to \chi_{q\kappa} + \omega_q t$$
$$\varphi_{q\kappa} \to \varphi_{q\kappa} + \omega_q' t$$
$$\psi_{q\kappa} \to \psi_{q\kappa} + \omega_q'' t.$$

Equation (19) is for instance transformed to

$$F_m^\Phi(\mathbf{Q},t) = F_m^\Phi(\mathbf{Q})\exp(i\omega_q' t).$$

The only time dependence results from the phase mode $\mathbf{q}$, since (19) contains no phases of other fluctuation modes. Accordingly, the scattering law for this example is

$$S(\mathbf{Q},\omega) = F_m^{\Phi *}(\mathbf{Q})\,F_m^\Phi(\mathbf{Q})\,\delta(\omega - \omega_q').$$

Consequently the results in (16), (19), and (21) may also be used for interpretation of neutron experiments.

3. Discussion

The elastic and dynamic structure factors given in (15), (16), (19), and (21) have been calculated within the

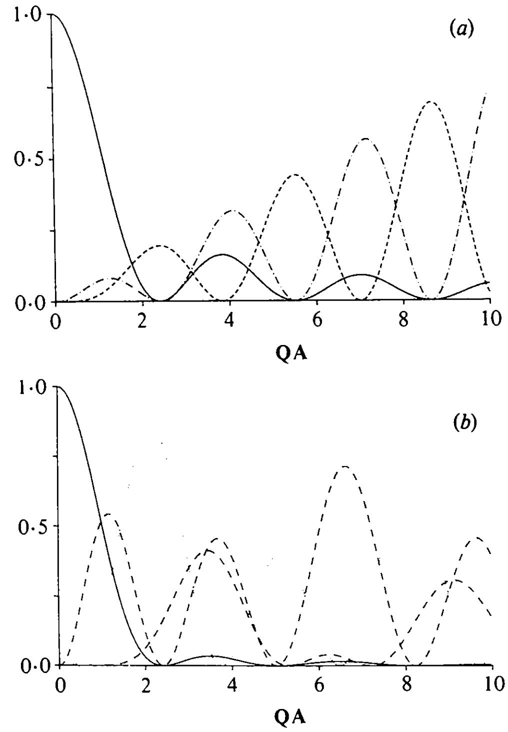

Fig. 1. Scattered intensity of main reflections and phasons, amplitudons and phonons around the main reflections ($m = 0$). The intensities are proportional to $(T_0)^2$ ——, $(T_0^\Phi)^2$ -----, $(T_0^a)^2$ ····· and $(T_0^U)^2$ -·-·-· [see equations (15), (16), (19) and (21)]. For better presentation different scale factors were chosen for the various functions. (*a*) $\langle\Phi^2\rangle$, $\langle a_{\rm rel}^2\rangle = 0$. Scale factors are 1 for $(T_0)^2$ and 8 for $(T_0^a)^2$ and $(T_0^U)^2$. (*b*) $\langle\Phi^2\rangle = 0{\cdot}25$, $\langle a_{\rm rel}^2\rangle = 0$; the same result is obtained for $\langle\Phi^2\rangle = 0$, $\langle a_{\rm rel}^2\rangle = 0{\cdot}25$. Scale factors are 1 for $(T_0)^2$, $(T_0^a)^2$ and $(T_0^U)^2$, and 0·3 for $(T_0^\Phi)^2$.

range $0 \le (\mathbf{QA}) \le 10$. Estimations of the phase fluctuations

$$\langle \Phi^2 \rangle = \tfrac{1}{2} \sum_q \Phi_q^2$$

indicate that, by assuming the mean-square atomic displacements caused by phonons to be comparable to acoustic phonon modes, $\langle \Phi^2 \rangle$ may reach values between 0·004 and 1 (Axe, 1980).

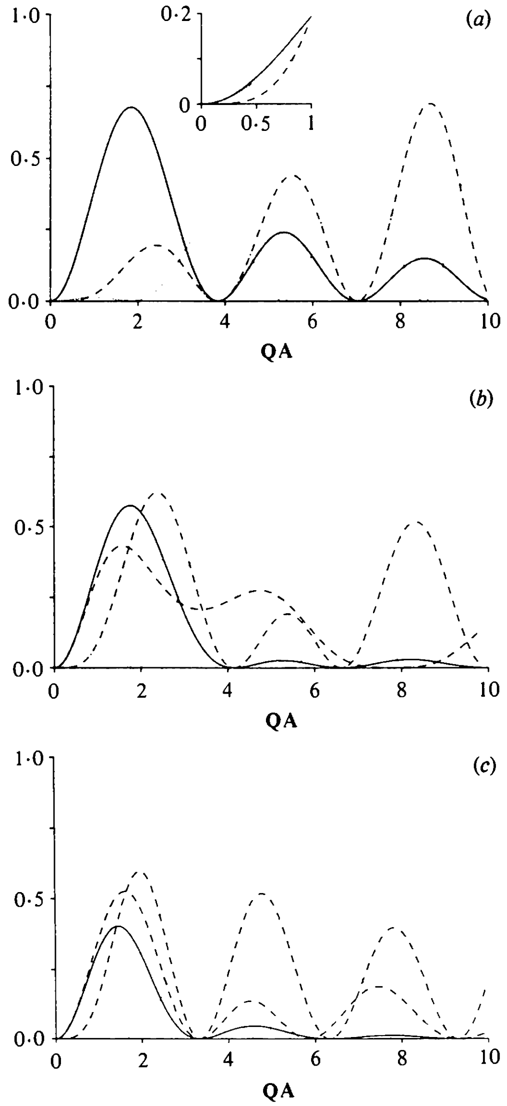

Fig. 2. The same functions as in Fig. 1 for $m = 1$ (first-order satellite reflections). (*a*) $\langle \Phi^2 \rangle$, $\langle a_{\rm rel}^2 \rangle = 0$. The scale factors are 0·5 for $(T_1)^2$, and 8 for $(T_1^a)^2$ and $(T_1^U)^2$. The insert shows the scattered intensities for small values (**QA**). All scale factors in the insert are 1. For the given values of $\langle \Phi^2 \rangle$ and $\langle a_{\rm rel}^2 \rangle$, $T_1^\Phi = T_1$ [equation (20)]. (*b*) $\langle \Phi^2 \rangle = 0{\cdot}25$, $\langle a_{\rm rel}^2 \rangle = 0$. Scale factors are 0·5 for $(T_1)^2$ and $(T_1^\Phi)^2$, and 2 for $(T_1^a)^2$ and $(T_1^U)^2$. (*c*) $\langle \Phi^2 \rangle = 0$, $\langle a_{\rm rel}^2 \rangle = 0{\cdot}25$. Scale factors are 0·5 for $(T_1)^2$ $(T_1^\Phi)^2$, and 1 for $(T_1^a)^2$ and $(T_1^U)^2$.

The phenomenological Landau theory explains the incommensurate structure by the freezing of a soft mode. In that case amplitude and phase fluctuations should be of approximately the same magnitude shortly below the phase transition. For this reason we have also examined the effect of amplitude fluctuations

$$\langle a_{\rm rel}^2 \rangle = \tfrac{1}{2} \sum_q (\bar{a}_q/\bar{A})^2.$$

Using this expression, the Debye–Waller factors

$$W^\Phi = \tfrac{1}{4}(\mathbf{QA})^2 \langle \Phi^2 \rangle \quad \text{and} \quad W^a \tfrac{1}{4}(\mathbf{QA})^2 \langle a_{\rm rel}^2 \rangle$$

may be directly compared.

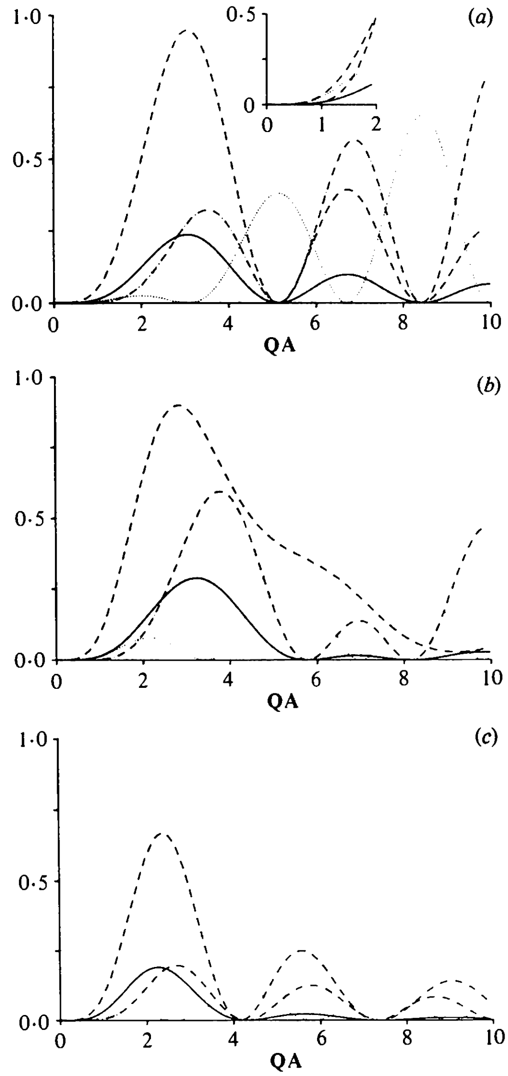

Fig. 3. The same functions as in Fig. 2 for $m = 2$ (second-order reflections). (*a*) $\langle \Phi^2 \rangle$, $\langle a_{\rm rel}^2 \rangle = 0$. The scale factors are 1 for $(T_2)^2$ and $(T_2^\Phi)^2$, and 8 for $(T_2^a)^2$ and $(T_2^U)^2$. In the insert all scale factors are 1, (*b*) $\langle \Phi^2 \rangle = 0{\cdot}25$, $\langle a_{\rm rel}^2 \rangle = 0$. The scale factors are 0·5 for $(T_2)^2$ and $(T_2^\Phi)^2$, and 3 for $(T_2^a)^2$ and $(T_2^U)^2$. (*c*) $\langle \Phi^2 \rangle = 0$, $\langle a_{\rm rel}^2 \rangle = 0{\cdot}25$. The same scale factors as in (*b*) are used.

The numerical calculation accuracy is better than 1%, entailing sometimes summations of Bessel functions up to 16th order. The given squared functions $(T_m)^2$ $(T_m^U)^2$, $(T_m^\Phi)^2$, and $(T_m^a)^2$ contain no phonon Debye–Waller factor, *i.e.* the actual structure factors may be considerably attenuated for high (**QA**), depending on the factor $\exp(-W_\kappa)$.

In Fig. 1(*a*) the results for $m = 0$ (main reflections) are summarized. For $\langle \Phi^2 \rangle$ and $\langle a_{\rm rel}^2 \rangle = 0$ (these ought to be taken as limiting cases for negligible phase and amplitude fluctuations, respectively) phasons cannot be observed. Yet, surprisingly enough, amplitudons may be well observed at the sites of weak main reflections. Anticipating comparable amplitudes $\mathbf{a}_q$ and $\mathbf{U}_q$, *i.e.* comparable mode frequencies, the amplitudons ought to be as well observable as phonons.

The results for $\langle \Phi^2 \rangle = 0{\cdot}25$ and $\langle a_{\rm rel}^2 \rangle = 0$ as well as $\langle a_{\rm rel}^2 \rangle = 0{\cdot}25$ and $\langle \Phi^2 \rangle = 0$, respectively, are shown in Fig. 1(*b*). Both cases yield the same results. The considerable attenuation of the functions T_0, T_0^U, and T_0^a above $(\mathbf{QA}) \simeq 2{\cdot}4$ are quite characteristic. Furthermore, phasons become observable, but only for $(\mathbf{QA}) \gtrsim 2$. Nevertheless, this result seems quite important for experiments, since it provides the possibility of estimating the magnitude of the phase and amplitude fluctuations by observation of the phasons near main reflections.

For first-order satellites and small (**QA**), the functions T_1, T_1^Φ, and T_1^a are almost identical (see insert in Fig. 2*a*). Whereas the functions T_1 and T_1^Φ become identical for $\langle \Phi^2 \rangle$ and $\langle a_{\rm rel}^2 \rangle = 0$ (see Fig. 2*a*), the antiphase behaviour of the satellite intensity relative to the amplitudons may be observed for all calculated fluctuations as well as for $m = 0$. At the maximum intensity of a first-order satellite no amplitudons may be observed (Figs. 2*b*,*c*). It is interesting to see that the fluctuations $\langle \Phi^2 \rangle$ and $\langle a_{\rm rel}^2 \rangle$ have quite a different effect on the function T_1^Φ. For phase fluctuations the node at $(\mathbf{QA}) \simeq 3{\cdot}8$ is displaced (Fig. 2*b*). The antiphase behaviour of the functions T_2 and T_2^a observed for $m = 2$ is similar to the cases $m = 0$ and $m = 1$. For small (**QA**) we can write

$$(T_2^\Phi)^2 \simeq (T_2^a)^2 \simeq 4(T_2)^2,$$

i.e. phasons and amplitudons increase relative to the satellite intensity. As for $m = 1$, phase and amplitude fluctuations affect the function T_2^Φ quite differently (see Figs. 3*b* and *c*).

4. Conclusion

In this report the phason and amplitudon structure factors have been derived and the influence of phase and amplitude fluctuations on the elastic as well as dynamic structure factor have been investigated. The main results are:

(i) Phasons are well observable near intense satellite reflections.

(ii) Owing to the influence of amplitude and phase fluctuations it is also possible to observe phasons near main reflections.

(iii) Amplitudons may be observed near main and satellite reflections. Apart from very small (**QA**) values, amplitudons are well observable when the main and satellite reflections are weak.

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