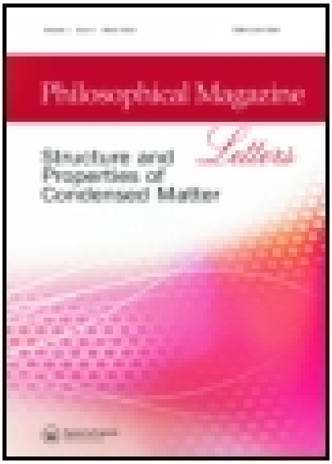

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Elastic stability and diffuse scattering in icosahedral quasicrystals

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Elastic stability and diffuse scattering in icosahedral quasicrystals

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ABSTRACT

Five elastic constants define the stiffness of coupled phonons and phasons in icosahedral quasicrystals. Requirements of elastic stability impose constraints (inequalities) on combinations of these elastic constants. This paper derives two sets of inequalities, one against the onset of spatially uniform strains, and a weaker set against the onset of spatially varying strains. When the elastic constants approach a violation of this weaker set of inequalities, Bragg peak intensities vanish continuously, while diffuse scattering intensities grow. Depending on which combination of elastic constants violates its stability condition, the diffuse scattering patterns peak in directions corresponding to icosahedron vertices, faces, or edges. Qualitative examination of diffuse scattering patterns therefore may yield an insight into the mechanism driving quasicrystal-to-crystal phase transitions, while quantitative studies may yield numerical values of the elastic constants.

§ 1. INTRODUCTION

Recently discovered icosahedral quasicrystals (Tsai, Inoue and Masumoto 1987), displaying long-range translational order (Guryan *et al.* 1989, Bancel 1989) appear to have solved sample quality problems (Heiney *et al.* 1987) and may allow the experimental study of intrinsic physical properties of quasicrystalline materials. The elasticity tensor is influenced by both icosahedral symmetry and quasiperiodic translational order, and relates directly to X-ray and neutron scattering experiments. Elasticity is especially interesting because two leading quasicrystal models make qualitatively different predictions for the temperature dependence of the elastic constants. And one model, the 'random tiling model' (Henley 1988, Widom, Deng and Henley 1989, Strandburg, Tang and Jaric 1989) predicts a phase transition driven either by instability in the phason modes (Henley 1989), or by coupling between phonons and phasons (Widom 1990, Hatwalne and Ramaswamy 1990). At low-temperatures the quasicrystal transforms into a crystal state which may (Biham, Mukamel and Shtrikman 1988, Ishii 1989, 1990a, b) correspond to a phason strained quasicrystal. Experiments (Bancel 1989, Audier and Guyot 1990) appear to verify both the phase transition and the crystalline nature of the low-temperature phase.

If one restricts attention to phonon modes, icosahedral symmetry is sufficiently close to full spatial isotropy that there exist only two independent elastic constants, which we take as the Lamé constants λ and μ. In this case the elastic free energy may be expressed as:

$$F_{\text{phonon}} = \int d\mathbf{r}\, [\tfrac{1}{2}\lambda u_{ii}^2 + \mu u_{ij} u_{ij}], \qquad (1)$$

where $u_{ij} \equiv [(\partial u_i/\partial r_j)+(\partial u_j/\partial r_i)]/2$ are components of the phonon strain tensor and $\mathbf{u}(\mathbf{r})$ is the phonon displacement as a function of position. But there also exist 'phason' displacements $\mathbf{v}(\mathbf{r})$ as a result of the quasiperiodic translational order (Bak and Goldman 1988). Associated with the phason modes is an additional component of elastic free energy (Lubensky 1988, Jaric 1987, Jaric and Mohanty 1988, Jaric and Nelson 1988, Tang 1990, Shaw, Elser and Henley 1991)

$$F_{\text{phason}}=\frac{1}{2}\int d\mathbf{r}\,[K_1 v_{ij}v_{ij} + K_2\{v_{kk}^2-\tfrac{4}{3}v_{ij}v_{ij}+[(\tau v_{12}+\tau^{-1}v_{21})^2+\text{cyclic permutations}]\}]. \quad (2)$$

where $v_{ij}\equiv\partial v_j/\partial r_i$ are components of the phason strain tensor. The phason elastic constants are defined so that K_1 and K_2 agree with Shaw (1991) ($K_1=0{\cdot}81\pm0{\cdot}01$ and $K_2=0{\cdot}495\pm0{\cdot}01$ for a three-dimensional random tiling model). Compared with Jaric and Nelson (1988) we have $K_1=m_3/3$ and $K_2=m_4/2\sqrt{5}$. Finally there is a contribution to the free energy which couples phonons and phasons

$$F_{\text{coupling}}=\int d\mathbf{r}\,K_3\{[v_{11}(u_{11}-\tau u_{22}+\tau^{-1}u_{33}) + 2u_{23}(\tau^{-1}v_{23}-\tau v_{32})]+\text{cyclic permutations}\}. \quad (3)$$

Compared with Jaric and Nelson (1988) we have $K_3=m_5/2\sqrt{15}$. Note that F_{phason} and F_{coupling} break spatial isotropy in a manner characteristic of icosahedral symmetry.

For an ordinary isotropic elastic medium without phason excitations the conditions of thermodynamic stability against a continuous phase transition may be expressed simply by requiring the positivity of bulk and shear moduli

$$\lambda+\tfrac{2}{3}\mu>0, \qquad \mu>0, \quad (4)$$

which implies that the free energy F_{phonon} is positive definite. One result of including the phason displacements is that the conditions (4) of elastic stability must be extended to include

$$K_1+\tfrac{5}{3}K_2>0, \qquad \mu(K_1-\tfrac{4}{3}K_2)>3K_3^2. \quad (5)$$

Note that Shaw's (1991) elastic constants obey these conditions. In this paper I ignore nonlinear terms in the free energy, so that (4) and (5) guarantee thermodynamic stability, and their violation results in a phase transition. Were higher order terms to be considered, including cubic terms of small magnitude, the continuous transition arising from violating these *local* stability criteria would be preempted by a weakly first-order transition.

In addition to thermodynamic stability one might enquire what are the effects of phonon and phason fluctuations on the structure factor. The Debye–Waller factor diminishes Bragg peaks from their values in the absence of fluctuations. The condition of a finite Debye–Waller factor imposes constraints on the elastic constants. For an ordinary isotropic elastic continuum without phasons these constraints are

$$\lambda+2\mu>0, \qquad \mu>0. \quad (6)$$

Note that (6) is less restrictive than (4). Including phasons we extend (6) by requiring

$$(\lambda+2\mu)(3K_1-4K_2)>12K_3^2,$$
$$\mu[27(K_1+K_2)-3\sqrt{5}(3K_1+5K_2)]>18K_3^2, \qquad (7)$$
$$3(\lambda+2\mu)(3K_1+4K_2)>4K_3^2.$$

Note that (7) is less restrictive than (5). One might call a violation of these latter inequalities a 'hydrodynamic' instability. Although one cannot in general reach the hydrodynamic instability in thermodynamic equilibrium, precursors such as a decline in Bragg peak intensities may be observed (Bancel 1989). Ishii (1991) has independently derived thermodynamic and hydrodynamic stability conditions equivalent to equations (5) and (7).

§ 2. Derivation of stability criteria

First consider the criterion of thermodynamic stability, equation (5). The condition for stability is that any uniform or fluctuating strain must increase the free energy. There are six independent uniform phonon strains $u_{ij}=u_{ji}$ and 9 independent uniform phason strains v_{ij}. Jaric and Mohanty (1988) express the elastic free-energy density as

$$f_{el}=\tfrac{1}{2}\,\mathbf{e}\cdot\mathbf{M}\cdot\mathbf{e}, \qquad (8)$$

where $\mathbf{e}$ is a 15-component vector of all uniform phonon and phason elastic strains and $\mathbf{M}$ is a 15×15 matrix including the phonon elasticity eqn. (1), phason elasticity eqn. (2) and coupling eqn. (3). The eigenvalues of $\mathbf{M}$ are the nondegenerate eigenvalue

$$\Lambda_1=3\lambda+2\mu, \qquad (9)$$

the fourfold degenerate eigenvalue

$$\Lambda_4=K_1+\tfrac{5}{3}K_2, \qquad (10)$$

and two fivefold degenerate eigenvalues

$$\Lambda_{5\pm}=\frac{(\lambda_u+\lambda_v)\pm[(\lambda_u-\lambda_v)^2+4\lambda_c^2]^{1/2}}{2}, \qquad (11)$$

where $\lambda_u=2\mu$, $\lambda_v=K_1-\tfrac{4}{3}K_2$, and $\lambda_c=\sqrt{6}K_3$. The condition for stability is then the positivity of all four distinct eigenvalues, which leads to the four conditions in equations (4) and (5).

Now we examine the effects of phonon and phason fluctuations of hydrodynamic stability. Consider the effects of nonlinear spatial variation in $\mathbf{w}(\mathbf{r})\equiv(\mathbf{u}(\mathbf{r}),\mathbf{v}(\mathbf{r}))$. Local variations in $\mathbf{w}$ lead to local phonon and phason strains which affect the local elastic free energy density as shown in equation (8). These position-dependent strains affect the total free energy through the integrals in eqns. (1–3). Fourier transforming the displacement and inserting into eqns. (1–3) we obtain the total free energy

$$F=\frac{1}{2}\int\frac{d\mathbf{p}}{(2\pi)^3}\,\mathbf{w}(-\mathbf{p})\cdot\mathbf{C}(\mathbf{p})\cdot\mathbf{w}(\mathbf{p}). \qquad (12)$$

The six-dimensional 'hydrodynamic' matrix $\mathbf{C}(\mathbf{p})$ is given by Jaric and Nelson (1988).

The Debye–Waller factor and diffuse scattering will ultimately be expressed in terms of eigenvalues and eigenvectors of the matrix $\mathbf{C}(\mathbf{p})$. One principal criterion for having a Bragg peak with finite amplitude turns out to be that all eigenvalues must be positive. I term the vanishing of a Bragg peak an *instability*, although it must be remembered that true thermodynamic stability is in general a stronger condition. When parameters such as the elastic constants are varied, the onset of an instability corresponds to a vanishing of the *minimum* of an eigenvalue. These minima should occur in directions of high symmetry. Thus we calculate the determinants of $\mathbf{C}(\mathbf{p})$ for $\mathbf{p}$ in the directions of icosahedron vertices, faces, and edges. Then we deduce stability criteria from the zeros of these determinants.

We only consider instabilities in which phasons play a significant role so that we assume $\lambda+2\mu>0$ and $\mu>0$. Making the further assumption that an instability is driven by either a large value of K_2 or else by a large value of K_3, but not both simultaneously, leads to simplifications. The point is that we need not concern ourselves with *all* zeros of the determinants, but only with those which occur *first* as K_2 or K_3 become non-negligible. Consider first the case of small K_3. As K_2 grows from zero to finite positive values, the first zero of any of the three determinants occurs in the vertex direction when

$$K_2=\frac{3}{4}K_1-\frac{3}{\lambda+2\mu}K_3^2. \tag{13}$$

If instead K_2 becomes negative, the first zero occurs in the face direction when

$$K_2=\frac{-3}{4}K_1+\frac{1}{3(\lambda+2\mu)}K_3^2. \tag{14}$$

Next consider the case of small K_2. As K_3 becomes large (either positive or negative), the first zero occurs in either the vertex direction when

$$K_3^2=\frac{(\lambda+2\mu)(3K_1-4K_2)}{12}, \tag{15}$$

or in the edge direction when

$$K_3^2=\frac{\mu}{6}(9(K_1+K_2)-\sqrt{5}(3K_1+5K_2)). \tag{16}$$

Noting the equivalence of equations (13) and (15) yields the hydrodynamic stability conditions of equation (7).

At the hyperspace reciprocal lattice point $\mathbf{Q}$, the structure factor $S(\mathbf{Q})$ is diminished by the Debye–Waller factor

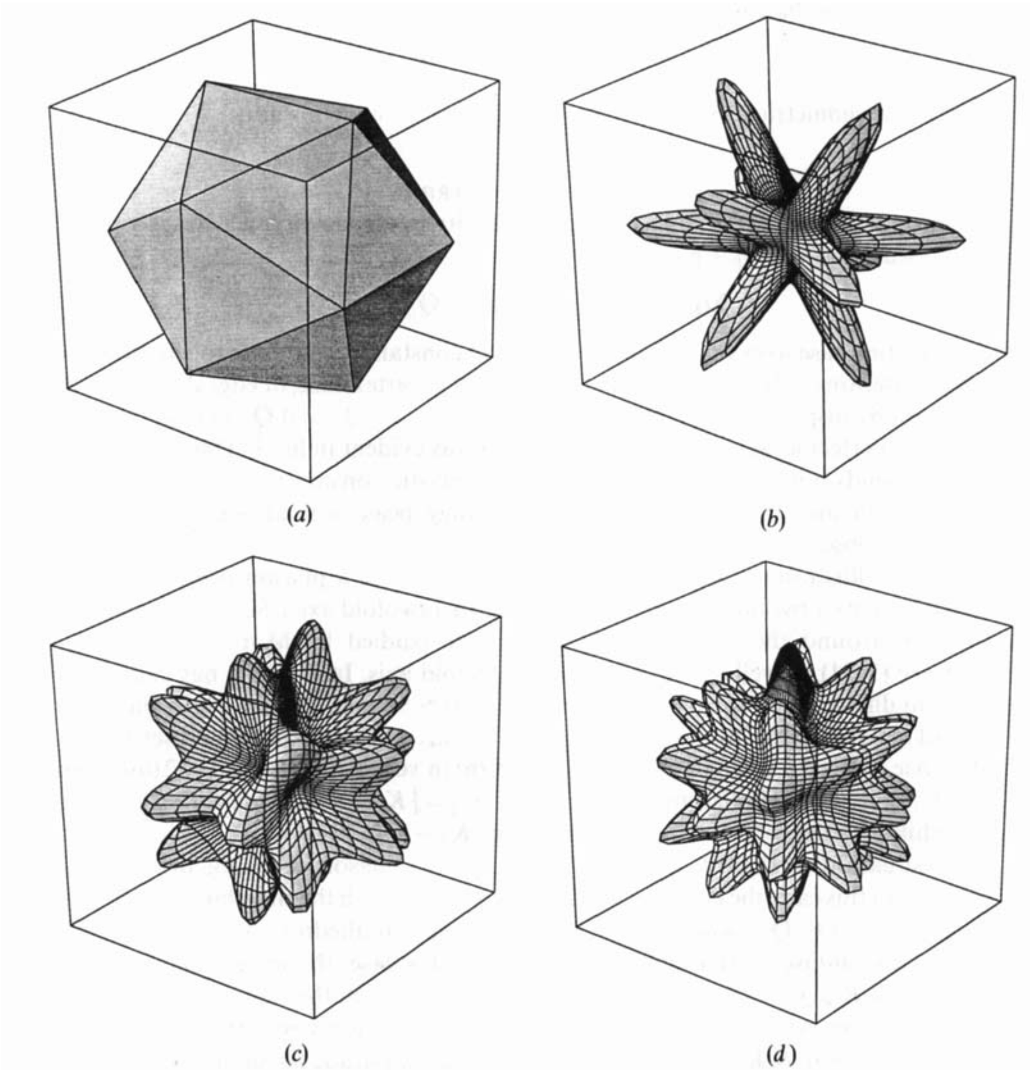
$$f(\mathbf{Q})=f_{\parallel}(\mathbf{Q}_{\perp})f_{\perp}(\mathbf{Q}_{\parallel}), \tag{17}$$

where $f_{\perp}$ and $f_{\parallel}$ are given by Jaric and Nelson (1988)

$$f_{\parallel(\perp)}(\mathbf{Q}_{\parallel(\perp)})=\exp\left\{-T\left(\frac{\mathbf{Q}^2_{\parallel(\perp)}}{6}\right)\int\frac{d^3\mathbf{p}}{(2\pi)^3}\operatorname{tr}[\mathbf{C}^{-1}(\mathbf{p})]_{\parallel,\parallel(\perp,\perp)}\right\}. \tag{18}$$

When the hydrodynamic matrix $\mathbf{C}(\mathbf{p})$ loses stability, one or both of the matrices $[\mathbf{C}^{-1}(\mathbf{p})]_{\parallel,\parallel(\perp,\perp)}$ diverge. But by (18), such a divergence leads to a vanishing of the corresponding Bragg peak intensity. This is a signature of a continuous phase transition out of the quasicrystal phase. Figure 1 plots the trace of $[\mathbf{C}^{-1}(\mathbf{p})]_{\perp,\perp}$ close (fig. 1 (*b*) and (*c*)) to the pure phason instabilities $K_2\approx\pm3K_1/4$ and close (fig. 1 (*d*)) to a phonon–phason coupling instability. Clearly these traces diverge in directions of high symmetry at the points of instability. Divergences in directions of fivefold symmetry (vertex directions) arise from positive values of K_2, while divergences in directions of

Fig. 1



(*a*) Icosahedron in its standard orientation with twofold axes along the *x*, *y*, and *z* coordinates. Integrand tr $[\mathbf{C}^{-1}(\mathbf{p})]_{\perp,\perp}$ of the phason Debye–Waller factor near (*b*) vertex instability, $K_1=1$, $K_2=+0{\cdot}7$, $K_3=0$. (*c*) Face instability, $K_1=1$, $K_2=-0{\cdot}7$, $K_3=0$. (*d*) Edge instability, $\lambda=\mu=K_1=1$, $K_2=0$, $K_3=0{\cdot}57$.

threefold symmetry (face directions) arise from negative values of K_2. Divergences in directions of twofold symmetry (edge directions) reveal the role of phonon–phason coupling in eqn. (16).

The integral in eqn. (18) is dominated by these divergences. Rather than evaluate the Debye–Waller factors in closed form, we study their behaviour approximately in the vicinity of the instabilities. When the distance ΔK from the instability vanishes, tr $[\mathbf{C}^{-1}(\mathbf{p})]_{\|,\|(\perp,\perp)}$ diverges like ΔK^{-1} for $\mathbf{p}$ in the appropriate symmetry direction, but varies quadratically with finite Gaussian curvature Γ for $\mathbf{p}$ slightly off the symmetry direction. Evaluating the integral in (18) we find

$$f_{\perp}(\mathbf{Q}_{\perp}) \sim (\Delta K)^{(\alpha T/\Gamma^{1/2})\mathbf{Q}_{\perp}^2}, \tag{19}$$

where α is a geometrical parameter independent of elastic constants.

§ 3. Diffuse scattering

Contours of constant diffuse scattering intensity in the vicinity of a Bragg peak at $\mathbf{Q}_{\parallel}$ are located at $\mathbf{q} = \mathbf{Q}_{\parallel} + \mathbf{p}$ where $|\mathbf{p}|^2$ is proportional to

$$\Sigma(\mathbf{p}) \equiv (\mathbf{Q}_{\parallel}, \mathbf{Q}_{\perp}) \cdot \mathbf{C}^{-1}(\mathbf{p}) \cdot (\mathbf{Q}_{\parallel}, \mathbf{Q}_{\perp}). \tag{20}$$

The interesting case to examine is when the elastic constants come close to violating the stability criterion (7). In such a case $\mathbf{C}^{-1}(\mathbf{p})$ diverges in vertex, face, or edge directions, as can be seen by inspecting fig. 1. But the dot products with $\mathbf{Q}_{\parallel}$ and $\mathbf{Q}_{\perp}$ in equation (20) remove the perfect icosahedral symmetry which was evident in fig. 1. Thus the precise pattern depends not only on the combination of elastic constants responsible for the instability, but also on the symmetry of the Bragg peak around which the diffuse scattering is observed.

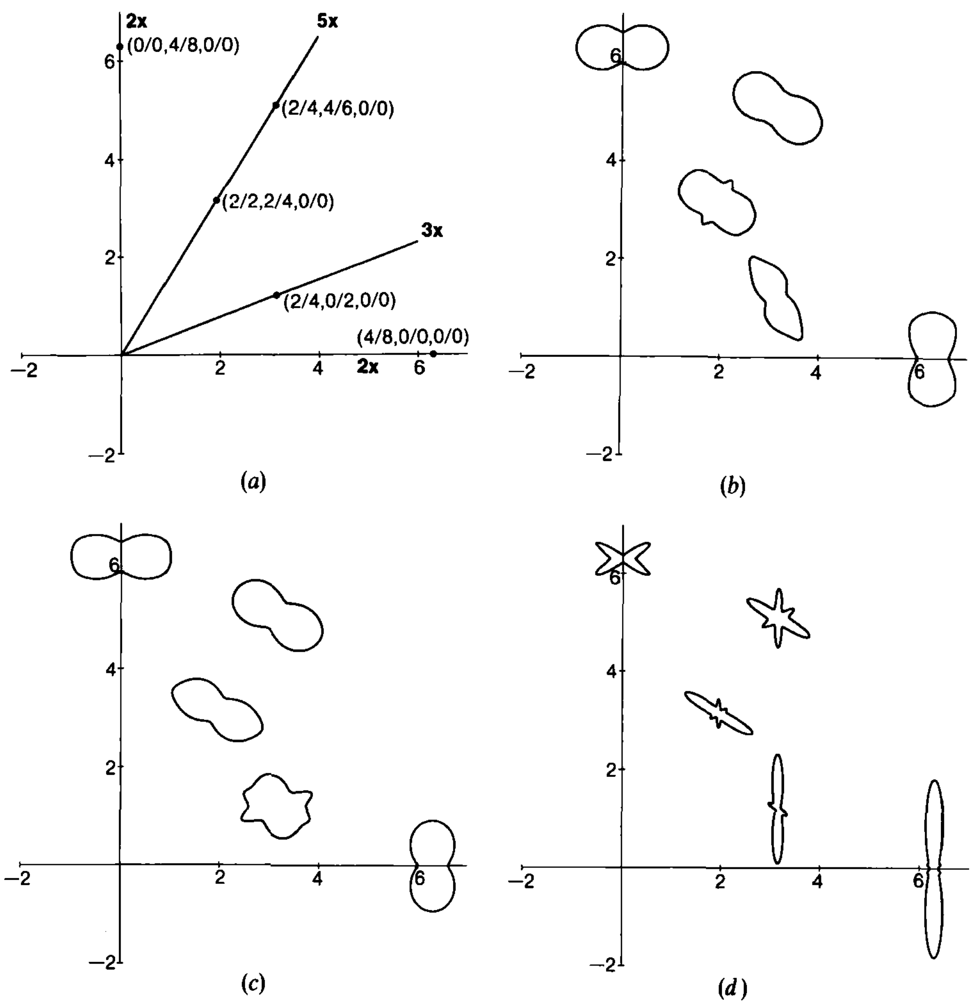
Figure 2 illustrates diffuse scattering patterns close to a phason instability. This figure represents a twofold plane (perpendicular to a twofold axis). Scattering patterns are shown around the same four peaks as were studied by Mori, Ishimasa and Kashiwase (1991) as well as one peak on a threefold axis. Ishii (1991) has calculated equivalent diffuse lineshapes and some in other planes as well. Consider now the likely modes of instability. We examine first those which are phason induced and set $K_3 = 0$. In this case we find a vanishing eigenvalue of $\mathbf{C}(\mathbf{p})$ in vertex directions (fig. 2 (*b*)) when $K_2 = \frac{3}{4} K_1$ and in face directions (fig. 2 (*c*)) when $K_2 = \frac{3}{4} K_1$. Diffuse intensity falls off as $|\mathbf{k}|^{-2}$, while at fixed $\mathbf{k}$ the intensity diverges as $|K_2 - K_2^*|^{-1}$.

The situation is slightly different for a phonon–phason coupling driven edge instability. In this case, the eigenvector of $\mathbf{C}(\mathbf{p})$ associated with the singular eigenvalue is orthogonal to $(\mathbf{Q}_{\parallel}, \mathbf{Q}_{\perp})$ when $\mathbf{k}$ points towards an icosahedron edge. Figure 2(*d*) illustrates the diffuse scattering for $|K_3| < K_3^*$. In this case, the orthogonality is not complete. As $|K_3| \to K_3^*$ the intensity falls off precipitously in the radial direction. The reason, of course, is that the singular contribution to the diffuse scattering vanishes in the radial direction. A $|\mathbf{k}|^{-2}$ fall off remains, but with amplitude far smaller than in the transverse direction. In fact Mori, Ishimasa and Kashiwase (1991) report diffuse scattering lineshapes in Al–Cu–Fe with contours of constant intensity greatly elongated in the transverse direction. But their result appears to conflict with the present theoretical analysis in that they report the radial intensity falling off faster than $|\mathbf{k}|^{-2}$.

§ 4. Conclusions

Let us consider the implications of eqns. (18) and (19) on the temperature dependence of scattering peak intensities. In random tiling models of quasicrystals the phason elastic constants are determined entirely by entropy; thus $K_1(T) = \kappa_1 * T$ and $K_2(T) = \kappa_2 * T$. In addition, for rigid tiles $K_3 = 0$. In this approximation $[\mathbf{C}^{-1}]_{\perp,\perp}$ is inversely proportional to temperature, leading to phason Debye–Waller effects $f_{\perp}(\mathbf{Q}_{\perp})$ *independent* of temperature. As the temperature decreases, the random tiling approximation should become less appropriate. For instance, there may be temperature-

Fig. 2



Diffuse scattering line shapes in a plane perpendicular to a twofold axis. Reciprocal space units are $2\pi/a$ (Jaric and Nelson 1988). All contours reduced by scale factor $1/(10Q_{\perp}^2+Q_{\|}^2)$. Elastic constants set as in fig. 1. (*a*) Peaks are indexed in accordance with the Cahn indexing scheme as modified by Ishimasa, Fukano and Tsuchimori (1988). (*b*) Vertex instability; (*c*) face instability; (*d*) edge instability (contours reduce by additional factor of 1/4).

independent contributions to $K_1(T)$ and $K_2(T)$. If we hypothesize that these temperature-independent contributions violate the stability criteria (7), then the actual ground state is not a quasicrystal. The peak intensities thus vanish below a temperature T_0 at which $\mathbf{C}(\mathbf{p})$ becomes singular. Above this temperature the peak intensities grow as a power law (eqn. (19)) then level off until the onset of melting. (But remember that true thermodynamic stability sets in at $T_1 \geqslant T_0$ determined by the criteria (5) so the complete vanishing of peaks may not be observed.) This behaviour stands in contrast to the predictions of energetic stabilization mechanisms (Levine and Steinhardt 1984, 1986, Socolar and Steinhardt 1986) for which the phason contribution to the peak intensity should diminish with increasing temperature just as does the phonon contribution (Levine *et al.* 1985, Lubensky *et al.* 1986).

The calculations outlined here indicate a means of measuring combinations of elastic constants. It is hoped that study of the temperature dependence of the elastic constants may discriminate between the energetic and entropic mechanisms of quasicrystal stability. More important, perhaps, are the purely qualitative results, including the phase transition from a crystal phase to the quasicrystal as temperature rises, and a jump in peak intensity to a finite value followed by a further increase as the temperature rises. These predictions appear to agree with experiment (Bancel 1989), but the experimental evidence so far cannot be regarded as proof of the theory because *detailed* and *quantitative* comparisons have not been made. This calculation aims to correct that problem by showing how important quantitative information may be extracted from experiments, and by providing greater qualitative detail than was previously available. Experimental measurements of diffuse scattering lineshapes, and their dependence on equilibration temperature and Bragg peak symmetry, can test this theory and possibly provide a clue to the mechanism driving the quasicrystal instability.

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