Reciprocal-space imaging of a real quasicrystal. A feasibility study with PILATUS 6M

Thomas Weber,^a Sofia Deloudi,^a Miroslav Kobas,^b Yoshihiko Yokoyama,^c
Akihisa Inoue^c and Walter Steurer^{a,d,*}

^aLaboratory of Crystallography, Department of Materials, ETH Zurich, CH-8093 Zurich, Switzerland, ^bDECTRIS Ltd, CH-5232, Villigen-PSI, Switzerland, ^cInstitute for Materials Research, Tohoku University, Katahira 2-1-1, Sendai City 980-8577, Japan, and ^dMNF, University of Zurich, CH-8057 Zurich, Switzerland. Correspondence e-mail: steurer@mat.ethz.ch

How many of the theoretically densely distributed Bragg reflections of a quasicrystal can be observed employing an area detector and synchrotron radiation? How does the reflection density of a real quasicrystal change as a function of exposure time, and what is the minimum distance between reflections? What does the distribution of diffuse scattering look like? To answer these questions, the Bragg reflection density of a perfect icosahedral quasicrystal with composition $\text{Al}_{64}\text{Cu}_{23}\text{Fe}_{13}$ was measured employing a novel type of single-photon-counting X-ray pixel detector, PILATUS 6M, which allows noise-free data collection with the extraordinarily large dynamic range of 20 bit. The reflection density was found to be two orders of magnitude lower than expected for a strictly quasiperiodic structure. Moreover, diffuse scattering reflects significant structural disorder, breaking six-dimensional F -lattice symmetry. These findings have some implications for the interpretation of physical properties.

© 2008 International Union of Crystallography
Printed in Singapore – all rights reserved

1. Introduction

More than 20 years of research have provided sometimes controversial but eventually converging answers to Per Bak's frequently quoted question 'Icosahedral crystals: Where are the atoms?' (Bak, 1986). Basically, the crystallography of quasicrystals is well understood today, and we know where most of the atoms are (see *e.g.* Steurer, 2004; Steurer & Deloudi, 2008, and references therein). However, fundamental questions concerning the factors governing the stability of quasicrystals, the role of clusters (Steurer, 2006) and, especially, of the configurational entropy (Henley *et al.*, 2006), are still not fully answered. Are quasicrystals high-temperature phases and is a minimum amount of structural disorder required for their stability, or does a quasiperiodic ground state exist? In other words, can real quasicrystals be described in terms of strictly quasiperiodic order or rather on the basis of a random tiling model (Elser, 1996; Henley *et al.*, 2006)? This is not an academic problem but is important for understanding and modeling structural-order-sensitive physical properties of quasicrystals. The Bragg reflection density and the amount of structural diffuse scattering considerably differs for these two cases.

With the availability of a novel type of area detector, PILATUS 6M (Kraft *et al.*, 2008), full reciprocal-space exploration with the required large intensity range recently became feasible. This detector is the significantly improved successor of the PILATUS 1M prototype (Broennimann *et al.*,

2006). For our reciprocal-space exploration with a focus on weak Bragg reflections and diffuse diffraction phenomena, we performed a special diffraction experiment on a high-quality icosahedral quasicrystal with composition $\text{Al}_{64}\text{Cu}_{23}\text{Fe}_{13}$, since its electric, magnetic and thermal properties have recently been studied by Dolinsek *et al.* (2007). Details about the growth of the crystal by the Czochralski method and its characterization as a perfect quasicrystal have been described by Yokoyama *et al.* (2002).

The degree of structural order can strongly influence physical properties such as electric or thermal conductivity. In most cases, disorder has not been taken into account for the modeling of physical properties. Usually, ideal quasiperiodic structure models have been used. Therefore, we wanted to compare our experimental observations with simulated diffraction patterns based on an ideal structure model. For that purpose, we used the sophisticated, strictly quasiperiodic structure model (QG model) of Quiquandon & Gratias (2006).

In the following, we call an actually existing crystal (*real crystal*), be it periodic or aperiodic, *perfect* if it only contains defects that are in thermodynamic equilibrium at given conditions. Point defects can exist in thermodynamic equilibrium. The atomic structure of a perfect crystal can be structurally ordered or disordered.

The reciprocal space of quasicrystals can best be described in terms of the higher-dimensional approach (see *e.g.* Yamamoto, 1996, and references therein). Accordingly, a three-

Figure 1

Log $I(\mathbf{H})$ versus $|\mathbf{H}^\perp|$ calculated (a) for the ideal QG structure model and (b) after applying a phason DW factor with $\langle u^2 \rangle = 0.12 \text{ \AA}^2$. Only Bragg reflections within a two-dimensional par-space section with $|\mathbf{H}^\parallel| \leq 0.8 \text{ \AA}^{-1}$ and $|h_i| \leq 40$, $i = 1, \dots, 6$, are shown. The reflections above the horizontal line are observable by standard synchrotron area-detector experiments. The curve, $I(\mathbf{H}) \propto |\mathbf{H}^\perp|^{-4}$, marks the general decrease of intensities with $|\mathbf{H}^\perp|$ without (a) and with (b) the phason DW applied.

dimensional quasiperiodic structure with icosahedral diffraction symmetry (icosahedral quasicrystal) can be considered as an irrational section of a periodic six-dimensional hypercrystal structure. The structural information is coded in so-called atomic surfaces (or occupation domains), which only extend in the perp(pendicular)-space. The three-dimensional physical or par(allel)-space $\mathbf{V}^\parallel$ and three-dimensional perp-space $\mathbf{V}^\perp$ are the two orthogonal subspaces of the six-dimensional Euclidean embedding space $\mathbf{V} = \mathbf{V}^\parallel \otimes \mathbf{V}^\perp$. The atomic surfaces of strictly ordered quasiperiodic structures are complex three-dimensional polytopes obeying the hard-core and the closeness conditions. This means that fully occupied atomic surfaces must not intersect each other and must be connected in such a way that the density remains constant upon shifting the par-space cut along $\mathbf{V}^\perp$.

Structural disorder affects the atomic surfaces in different ways. Random phonon fluctuations (thermal vibrations of the atoms, *i.e.* excitations in $\mathbf{V}^\parallel$) can be described by the well known Debye–Waller (DW) factor $T_k(\mathbf{H}^\parallel)$. Random phason fluctuations (*i.e.* excitations in $\mathbf{V}^\perp$), which correspond to par-space atomic jumps in double-well potentials (phason flips), can analogously be accounted for by the phason DW factor $T_k(\mathbf{H}^\perp)$. The diffuse scattering phenomena related to $T_k(\mathbf{H}^\parallel)$ and $T_k(\mathbf{H}^\perp)$ are thermal diffuse scattering (TDS) and phason diffuse scattering (PDS), respectively. TDS (PDS) is strong beneath and around strong Bragg reflections with a large par-space (perp-space) component of the six-dimensional diffraction vector $\mathbf{H}$ (see *e.g.* Kobas *et al.*, 2005*a,b*, and references therein).

The original motivation of our study was the characterization of the quasicrystal that has been used for the study of physical properties by Dolinsek *et al.* (2007). The availability of the novel pixel detector PILATUS 6M opened further possibilities, the first of which was the extension of the limits of reciprocal-space imaging of an icosahedral quasicrystal that

has been classified perfect by conventional diffraction techniques. An intensity range comparable to that of the present experiment has only previously been accessible with point detectors, which could not be used for quasi-continuous reciprocal-space imaging over large reciprocal space volumes with reasonable costs. A second possibility was to explore the suitability of PILATUS 6M for the detection of very weak intensities next to very strong Bragg reflections.

Since software for the integration of quasicrystal reflections for this detector does not yet exist, a quantitative structure refinement or a comparison of observed intensities versus calculated intensities was not possible. Therefore the reflection density has been chosen as a parameter to be measured. Furthermore, it has not been the goal of this study to show that a phason DW factor is needed to describe the experimental observations; this is known. The goal was to explore its effect on the observable reflection density and therewith on the information that is principally accessible for structure analysis. This has never been studied before in this way.

2. Reciprocal-space simulation

The structure factor of a Bragg reflection of an icosahedral quasicrystal with diffraction vector $\mathbf{H} = (\mathbf{H}^\parallel, \mathbf{H}^\perp)$ is

$$F(\mathbf{H}) = \sum_{k=1}^N f_k(|\mathbf{H}^\parallel|) g_k(\mathbf{H}^\perp) T_k(\mathbf{H}) \exp(2\pi i \mathbf{H} \cdot \mathbf{r}_k). \quad (1)$$

$f_k(|\mathbf{H}^\parallel|)$ is the usual atomic form factor; $g_k(\mathbf{H}^\perp)$ denotes the geometrical form factor, which is the Fourier transform of the k th atomic surface in the six-dimensional unit cell with position vector $\mathbf{r}_k$. The projection of the six-dimensional intensity-weighted reciprocal lattice onto the three-dimensional par-space gives the dense set

$$M^* = \left\{ I(\mathbf{H}^\parallel) = \sum_{i=1}^6 h_i a_i^*, \quad h_i \in \mathbf{Z} \right\} \quad (2)$$

of Bragg reflections with intensities $I(\mathbf{H}^\parallel) = I(\mathbf{H}) = F(\mathbf{H}) F^*(\mathbf{H})$ and their diffraction vectors $\mathbf{H}^\parallel$, which form a $\mathbf{Z}$ module of rank 6. In our case a_i^* equals $0.05599 \text{ \AA}^{-1}$. The six-dimensional unit cell is hypercubic face centered, with lattice parameter $a^{6D} = 12.6292 \text{ \AA}$, *i.e.* two times the frequently used lattice parameter, $6.3146 \text{ \AA}$, of the primitive six-dimensional unit cell. The F centering is mainly due to chemical ordering, leading to different atomic surfaces at even and odd nodes of the six-dimensional lattice. The centering translations are of the type $(\frac{1}{2} \frac{1}{2} 0 0 0 0)$, yielding, together with the position $(0 0 0 0 0 0)$, a total of 16 symmetrically equivalent positions and the integral reflection condition

$$h_1 h_2 h_3 h_4 h_5 h_6 : h_i + h_j = 2n \forall i \neq j = 1, \dots, 6. \quad (3)$$

The three-dimensional periodic average structure of such a six-dimensional face-centered cubic structure is of the NaCl type (Steurer & Deloudi, 2008).

Although the Bragg reflections are densely distributed in three-dimensional reciprocal space, the diffraction patterns of icosahedral quasicrystals appear discrete. Since reflection intensities fall off rapidly with increasing $|\mathbf{H}^\perp|$, $I(\mathbf{H}) \propto |\mathbf{H}^\perp|^{-4}$

for approximately spherical atomic surfaces, observable reflections have rather small perp-space components $|\mathbf{H}^\perp|$. Because of the way in which the six-dimensional reciprocal space is projected onto three dimensions, it follows that the closer neighboring reflections are to each other in par-space the farther away they are in perp-space. Consequently, their intensities differ by many orders of magnitude (Fig. 1). Accordingly, the experimentally observable reflection density depends on the dynamic range and the resulting peak/background ratio of a diffraction experiment. Random phason fluctuations, entering the structure factor as a phason DW factor, can further strongly decrease the observable Bragg intensities compared with those of a strictly quasiperiodic structure.

In the case of the ideal QG model we expect 74 725 reflections within a two-dimensional par-space section with $|\mathbf{H}^\parallel| \leq 0.8 \text{ \AA}^{-1}$, $|h_i| \leq 40$, $i = 1, \dots, 6$, and a dynamic range of nine orders of magnitude, which is the detection range of our experiment. The number of expected observable reflections is drastically reduced to 8357 when we apply an experimentally derived single isotropic phason DW factor with $\langle u^2 \rangle = 0.12 \text{ \AA}^2$. The DW factor was determined by fitting the calculated reflection density to the observed one under the constraint that all observed reflections must also have calculated counterparts of comparable intensity. Within the dynamic range of $\sim 10^5$ for a standard area-detector-based synchrotron data collection, only the reflections above the line in Fig. 1 would be strong enough to be detected. The determination of a reliable phason DW factor from a structure refinement would be difficult based only on such a small set of reflections. Strong correlations between the phason DW factor and occupation factors would bias the results.

Besides a low Bragg reflection density, there is another indicator for the lack of strictly quasiperiodic order: structural diffuse scattering. While purely substitutional disorder changes neither the shape nor the size of atomic surfaces, displacive disorder leads to partially overlapping blurred atomic surfaces, enhancing the decrease of Bragg intensities. However, substitutional disorder is usually coupled with displacive disorder due to structural relaxation effects. Thus, Bragg and diffuse intensity data not only provide information on the atomic positions but can also give clues about the amount of disorder necessary for the stabilization of quasiperiodic order. While phason diffuse scattering is strongest beneath and around strong Bragg reflections with large $\mathbf{H}^\perp$, other types of structural diffuse scattering may be found elsewhere.

3. Experimental data collection

Despite the significant progress made in the development of X-ray sources and area detectors in the past two decades, measurement of very weak scattering intensities adjacent to strong ones has always been a challenging task. Area detectors based on charge-coupled devices (CCDs) or imaging plates (IPs) are well suited for the purposes of standard X-ray structure determination. However, their restricted dynamic

range (≤ 17 bit) with relatively high intrinsic background, the long read-out time (seconds to minutes) and particularly the lack of energy discrimination still hamper experiments where most of the desired information is in the weak diffraction intensities. Long exposure times lead to artifacts such as blooming effects of saturated CCD pixels or to non-erasable overexposures in IP detectors. Multiple exposures can partly overcome these problems. However, the fact that parasitic scattering (extrinsic background) and read-out noise are accumulated at the same time sets the experimental limit for the accessible signal-to-noise ratio.

The situation is different for the novel PILATUS 6M detector, developed at the Swiss Light Source (SLS), Villigen, Switzerland. PILATUS detectors are based on CMOS hybrid pixel technology and operate in single-photon-counting mode (Kraft *et al.*, 2008). X-rays are directly transformed into electric charge in the silicon sensor. The signals are amplified and processed in the CMOS readout chips. This technology allows the collection of noise-free data with a maximum dynamic range of 20 bit. An adjustable lower energy threshold permits energy discrimination. Pixels are physically distinct in so far as an overexposure of a pixel does not influence the neighboring ones. The framing frequency of the PILATUS 6M detector is up to 10 Hz with 5 ms read-out time. Its 60 modules (487×195 pixels each, with a pixel size of $172 \mu\text{m}$) are separated by gaps of 7 pixels vertically and 17 pixels horizontally (Fig. 2).

Our measurements were performed at the protein crystallography beamline X06SA at the SLS, with a flux (at 12.4 keV) greater than 10^{12} photons s^{-1} and a beam divergence of $450 \mu\text{rad}$ horizontally and $60 \mu\text{rad}$ vertically. The sample was mounted on top of a glass capillary. In order to avoid parasitic scattering from glass and glue, only the crystal tip was irradiated by the beam. There was no need for 'bathing' the crystal in the beam since a quantitative evaluation of intensities was not possible anyway owing to the lack of integration software.

In a first exposure the energy threshold of the detector was set to count only radiation with energy $E > 8 \text{ keV}$ ($E_{\text{source}} = 16.02 \text{ keV}$, $\lambda = 0.765 \text{ \AA}$), *i.e.* scattered photons and fluorescence radiation from Cu were accepted, while that from Al and Fe was filtered (Fig. 2*a*). The experiment was then repeated with the energy threshold increased to 10 keV, completely eliminating fluorescence radiation (Fig. 2*b*). The remaining background amounts to $\ll 10$ counts per pixel. A comparison of Figs. 2*a*) and 2*b*) clearly shows that in the first experiment weak Bragg and diffuse intensities are almost completely hidden in the background because of fluorescence from Cu, although the incident beam radiation energy is far from the Cu absorption edge. For energy-integrating detectors such as CCDs or IPs, the observed background would be even more pronounced because Fe and Al fluorescence would be detected as well.

In order to check whether increasing the exposure time leads to the increasing reflection density expected for a strictly quasiperiodic crystal, we collected 753 data sets: 0.1° scans over a total of 10° were repeated 251 times, each at three

different detector positions to fill the gaps between the single detector modules. Images were taken with a frequency of six frames per second, so that the measurement of the 75 300 frames took less than four hours. While the counting range increases by a factor of 753 to 7.53×10^8 , the range of observable reflections is only increased by a factor of $753^{1/2}$ to 2.74×10^7 according to Poisson statistics. In the line scan (Fig. 3c), the improvement of the signal-to-noise ratio can be clearly seen. Two reconstructed twofold zero-layer reciprocal-space sections with minimum and maximum exposure time, respectively, are shown in Fig. 3. A 753 times longer exposure leads to only $\sim 20\%$ more reflections, much fewer than expected for a quasicrystal with strictly quasiperiodic structure.

Figure 2

Single-frame images (one oscillation around 0.1° each) recorded at 16 keV beam energy and with energy thresholds of (a) 8 keV and (b) 10 keV. The white lines result from the gaps between the detector modules.

4. Discussion

The observed reflection density can be best described by the QG model after applying an isotropic phason DW factor

$$T_k(\mathbf{H}^\perp) = \exp(-2\pi^2 |\mathbf{H}^\perp|^2 \langle u^2 \rangle), \quad (4)$$

with $\langle u^2 \rangle = 0.12 \text{ \AA}^2$ (see Fig. 1). This factor reduces the number of observable reflections expected in the reciprocal-space section shown in Fig. 3 from 2466 to 276. However, experimentally, we can only observe approximately 50 reflections, some of them with rather large perpendicular-space components $|\mathbf{H}^\perp|$ of the diffraction vector. Since no broadening of reflections as a function of $|\mathbf{H}^\perp|$ has been observed for this crystal (Yokoyama *et al.*, 2002), random phason strain cannot

Figure 3

Reconstructed twofold reciprocal-space sections (horizontally $0\text{--}0.33 \text{ \AA}^{-1}$, vertically $0.4\text{--}0.6 \text{ \AA}^{-1}$ from the origin) based on (a) a single exposure and (b) 753 exposures. The white stripes in (a) result from the gaps between the detector modules, which are filled by multiple exposures at shifted detector positions in (b). Arrows indicate diffuse maxima breaking six-dimensional F -lattice symmetry; the circle marks a contribution from a second grain and the brace the shortest distance between Bragg reflections. Indices and perp-space components ($\text{\AA}^{-1}$) of numbered reflections: 1 004242 0.046, 2 $\bar{1}151\bar{1}$ 0.342, 3 $\bar{1}133\bar{3}$ 0.149, 4 004240 0.157, 5 222424 0.335, 6 $611333\bar{1}$ 0.040. Line scans through (a) and (b) along the thin line in (a) are shown in (c). For clarity, the upper curve is shifted upwards by two counts. The absolute number of counts is given.

be the reason for non-observability of so many reflections. One reason may be shortcomings of the idealized model structure. Another, perhaps even more important, cause may be the influence of disorder on the intensity of Bragg reflections, which cannot be described properly by an isotropic overall phason DW factor. This would be the case if the different atomic species Al, Cu and Fe have different phason DW factors or for disorder of non-phasonic origin.

These results underline the importance of including not only all observed but also all unobserved (*sic*) reflections in a quasicrystal structure analysis, as is standard for periodic crystals. However, since ideal quasicrystals have infinitely many reflections in a given par-space range, not all unobserved reflections can be included. Only those that have calculated intensities within the dynamic range of the diffraction experiment should be considered. To the best of the authors' knowledge, this has never previously been done.

The (static) mean-square perp-space fluctuation amplitude, $\langle u^2 \rangle^{1/2} = 0.35 \text{ \AA}$, corresponds to 7.2% of the diameter of the largest atomic surface and to 19% of the smallest one. In the structure model, such a fluctuation creates a considerable amount of phasonic (including chemical) disorder. For instance, about 20% of the atoms created by the largest atomic surface and 47% of the atoms originating from the smallest atomic surface are involved. Indeed, the large amount of chemical disorder, mostly Al/Cu substitution, found for the 1/1 approximant (Simonet *et al.*, 2005) indicates the possibility for a similar kind of disorder for the icosahedral phase.

4.1. Disorder

Structural disorder is always reflected in diffuse scattering. Indeed, Fig. 3 shows almost omnipresent diffuse diffraction intensities. Fig. 4 shows a series of reciprocal-space sections, which differ by $0.005 \text{ \AA}^{-1}$ increments from image to image. This shows the three-dimensional intensity distribution of diffuse scattering, of PDS around Bragg reflections and of other types of structural diffuse scattering in between. The ring-like intensities, which move from right to left in Figs. 4(e)–4(g), originate from secondary air scattering of the strong reflection marked 1 in Fig. 4(h). This can be seen in the lowermost image, where the arc of diffuse scattering crosses the center of reflection 1.

Besides PDS, TDS and diffuse streaks, there are also diffuse maxima located at Bragg positions of the type

$$h_1 h_2 h_3 h_4 h_5 h_6 : h_i + h_j = 2n + 1 \forall i \neq j = 1, \dots, 6, \quad (5)$$

which are systematically extinct for a six-dimensional F lattice (see Fig. 3). From the full width at half-maximum of the diffuse maxima ($\sim 0.01 \text{ \AA}^{-1}$), the average correlation length of the symmetry-breaking ordering can be estimated as $\sim 100 \text{ \AA}$. This can be interpreted as a form of antiphase domain structure with a domain size of $\sim 100 \text{ \AA}$.

This means that the quasiperiodic structure shows strict long-range F centering only on a statistical basis, averaging over the structures of the antiphase domains. Thus, in each domain a subset of atoms must order in a way that breaks F

centering. Indeed, as mentioned above, considerable chemical disorder has been found on some sites in the 1/1 approximant (Simonet *et al.*, 2005); significant chemical disorder also results from the random phason fluctuations related to the phason DW factor needed to explain our observations.

4.2. Umweganregung

Umweganregung (multiple diffraction) is believed to be omnipresent during a diffraction experiment on a quasicrystal

Figure 4

Reconstructed twofold reciprocal space sections based on 753 exposures. Images (a)–(h) differ by incremental shifts of $0.005 \text{ \AA}^{-1}$ along the reciprocal-space direction perpendicular to the image plane and show a similar reciprocal-space section to Fig. 3(b), anticlockwise rotated by 90° . The locations of these images are shown in the lowermost image; this is a reciprocal-space section perpendicular to images (a)–(h) along the white horizontal hairline in (h), which has the same orientation as that in Fig. 3(a). Indices and perp-space components ($\text{\AA}^{-1}$) of numbered reflections are the same as in Fig. 3(b).

as a result of its dense set of Bragg reflections. Consequently, the poor fit of weak reflections in some quasicrystal structure analyses is frequently attributed to the enhancement of weak reflections by *Umweganregung*. Are there any indications for multiple scattering effects in our experiment? Multiple diffraction means that at least two Bragg reflections, $I(\mathbf{H})$ and $I(\mathbf{G})$, are excited simultaneously by the primary beam with wavevector $\mathbf{k}_0$. Then, the coupling reflection $I(\mathbf{H}-\mathbf{G})$ is also excited, with the reflected beam $\mathbf{k}_G$ acting as the (usually much weaker) primary beam. The reflected beams $\mathbf{k}_H$ and $\mathbf{k}_{H-G}$ point in the same direction and the resulting interference wave is detected instead of $I(\mathbf{H})$.

Fortunately, multiple diffraction only plays a role if $I(\mathbf{G})$, $I(\mathbf{H}-\mathbf{G}) \gg I(\mathbf{H})$. Strong reflections must have rather small values for the perp-space component of the diffraction vectors (see Fig. 1). If $\mathbf{G}^\perp$ and $\mathbf{H}^\perp - \mathbf{G}^\perp$ are both small, then $\mathbf{H}^\perp$ is also small. Therefore, the majority of very weak unobservable reflections, i.e. those with large values of $\mathbf{H}^\perp$, cannot be enhanced sufficiently by multiple diffraction to become observable. Indeed, we do not see more reflections with large $\mathbf{H}^\perp$ than expected without *Umweganregung*. Since our crystal was randomly oriented, symmetry-breaking reflections would be created; these have not been observed. Of course, this does not exclude the cases where intensities of observable reflections are merely modified by *Umweganregung*.

Generally, the situation for quasicrystals is comparable to that of complex intermetallic phases with large unit cells where *Umweganregung* is usually no problem at all for structure analysis. This is particularly true for our icosahedral quasicrystal, for which only reflections related to the 13/8 approximant can be observed. Significant *Umweganregung* in quasicrystals mainly takes place for special diffraction geometries such as rotation around diffraction vectors (see e.g. Lee *et al.*, 1993).

5. Summary

By a qualitative approach we have shown that the icosahedral quasicrystal studied can be considered perfect only with respect to its average structure. The low Bragg reflection density and the diffuse scattering indicate that the local structure considerably differs from the quasiperiodic average structure. Although these results are no proof of the validity of the random-tiling-based structure model of Elser (1996), they are more supportive of this model than of the strictly quasiperiodic QG model, which is the 'very continuation of Elser's approach but keeping a fully deterministic point of view'

(Quiquandon & Gratias, 2006). However, to obtain more experimental evidence for the possible validity of Elser's model, temperature-dependent measurements would be needed. For the modeling of structure-sensitive properties, such as electrical or thermal conductivity, structural disorder has certainly to be taken into account. Ideal quasiperiodic structure models would not be able to fully describe the experimental findings. Ideal structure models can be very useful, however, in understanding fundamental structure/property relationships of quasicrystals. Finally, multiple diffraction does not seem to create greater problems for structure analysis than is the case for other complex metallic phases, since their observable reflection density is on a similar scale.

Support by the Swiss National Foundation under grant No. 200020-105158 is gratefully acknowledged. We thank both the staff of the protein crystallography beamline X06SA and the SLS detector group for the opportunity for and assistance with data collection.

References

- Bak, P. (1986). *Phys. Rev. Lett.* **56**, 861–864.
- Broennimann, Ch., Eikenberry, E. F., Henrich, B., Horisberger, R., Huelsen, G., Pohl, E., Schmitt, B., Schulze-Briesse, C., Suzuki, M., Tomizaki, T., Toyokawa, H. & Wagner, A. (2006). *J. Synchrotron Rad.* **13**, 120–130.
- Dolinsek, J., Vrtnik, S., Klanjek, M., Jaglicic, Z., Smontara, A., Smiljanic, I., Bilusic, A., Yokoyama, Y., Inoue, A. & Landauro, C. V. (2007). *Phys. Rev. B*, **76**, 054201.
- Elser, V. (1996). *Philos. Mag. B*, **73**, 641–656.
- Henley, C. L., de Boissieu, M. & Steurer, W. (2006). *Philos. Mag.* **86**, 1131–1151.
- Kobas, M., Weber, Th. & Steurer, W. (2005a). *Phys. Rev. B*, **71**, 224205.
- Kobas, M., Weber, Th. & Steurer, W. (2005b). *Phys. Rev. B*, **71**, 224206.
- Kraft, P., Henrich, B., Eikenberry, E. F. & Broennimann, C. (2008). *J. Synchrotron Rad.* **15**. In preparation.
- Lee, H., Colella, R. & Chapman, L. D. (1993). *Acta Cryst.* **A49**, 600–605.
- Quiquandon, M. & Gratias, D. (2006). *Phys. Rev. B*, **74**, 214205.
- Simonet, V., Hippert, F., Brand, R. A., Calvayrac, Y., Rodriguez-Carvajal, J. & Sadoc, A. (2005). *Phys. Rev. B*, **72**, 024214.
- Steurer, W. (2004). *J. Non-Cryst. Solids*, **334**, 137–142.
- Steurer, W. (2006). *Philos. Mag.* **86**, 1105–1113.
- Steurer, W. & Deloudi, S. (2008). *Acta Cryst.* **A64**, 1–11.
- Yamamoto, A. (1996). *Acta Cryst.* **A52**, 509–560.
- Yokoyama, Y., Matsuo, Y., Yamamoto, K. & Hiraga, K. (2002). *Mater. Trans.* **43**, 762–765.