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On the problem of the correct interpretation of phason elasticity in quasicrystals

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(today)

Recently Francoual *et al.* [1] claimed to have observed the dynamics of long-wavelength phason fluctuations in *i*-AlPdMn quasicrystals. We will show that the data reported call for a more detailed development of the elasticity theory of Jarić and Nelsson [2] in order to determine the nature of small phonon-like atomic displacements with a symmetry that follows the phason elastic constants.

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I. INTRODUCTION

Recently Francoual *et al.* [1] claimed to have observed the dynamics of long-wavelength phason fluctuations in *i*-AlPdMn quasicrystals. These claims were based on the observation of very long relaxation times in the so-called speckle patterns of the diffuse scattering measured with coherent X-rays. There are problems with these claims that we try to tackle in the present paper.

Let us resume some important points of the paper:

(1) There is no proof that the (kinetics of the) diffuse scattering observed by the authors is due to tile disorder produced by atomic jumps. Alternative interpretations for the data are possible and have not been ruled out. Chemical disorder and a field of small atomic displacements that are not phason jumps can lead to very similar signals.

(2) Atomic jumps are in principle not correlated over long distances, especially when they participate in atomic self-diffusion. Experimental data never code correlations between atomic jumps. They correspond to correlations between atomic positions.

(3) Identifying the diffuse scattering with jumps is in contradiction with quasielastic neutron scattering data which clearly demonstrate that the number of jumps increases with T, while the diffuse scattering shows exactly the opposite behaviour.

(4) Introducing a concept of a phason as a displacive mode of atomic jumps in a QC is problematic. An ondulating cut ("sine wave") does not produce a physical wave in physical space. The displacement field defined by the "sine wave" is non-periodic, and the concept of such a mode introduces notions that are unphysical, such as spooky long-distance correlations between isolated phason jumps. The "sine wave" is in this respect not any better than a rigid translation of the cut.

(5) The authors confuse coherent scattering signals with evidence for collective effects, Fourier components with physical waves, the Fourier transform of the QC with the Fourier transform of a field of atomic jumps.

(6) The authors overinterpret Lubensky's statement [3] that phasons are diffusive.

(7) The data of the authors are not specific enough to warrant claims that they would present evidence for the random tiling model.

(8) Contrary to statements by several authors, the terminology "phason jumps" is by all standards a correct terminology.

Several of these problems are quite subtle and require a detailed discussion. The task is rendered difficult by interferences between the problems listed above.

The paper is organized as follows. In the second Section we prove that the "sine wave" proposed by the authors is not a proper wave, and that it is not periodic. We take the occasion to show that the interpretation of the data is not unique. In the third Section we show why the coherence of the data does not imply that there would be correlations between atomic jumps. In the fourth Section we explain why the neutron data rule out the scenario of tile flips proposed by the authors. In Section V we discuss a number of problems for the understanding of the elasticity theory. In Section VI we reexamine the evidence presented in favour of the random tiling interpretation. In Section VII we discuss the validity of the claim that "phason jumps" would not be a correct terminology. And in Section VIII we conclude.

II. PERIODIC DELUSIONS

A. The function $u_\perp \sin(q_\parallel x_\parallel)$ is not a wave

Various authors [4] have proposed "phason waves" of the type $u_\perp \sin(q_\parallel x_\parallel)$, which propagate with wavelength $\lambda = 2\pi/q_\parallel$ along parallel space $E_\parallel$, and are polarized with an amplitude $u_\perp$along perpendicular space $E_\perp$. But it is an obvious mathematical fact that this does not define a periodic displacement wave in parallel space $E_\parallel$. To see

this, it suffices to inspect the displacement pattern the "sine wave" brings about on the drawing that defines the cut-and-projection algorithm in $E = E_\parallel \oplus E_\perp = \mathbb{R}^2$ that generates the Fibonacci sequence to convince oneself of this. The very misleading notation as a perfect sine wave $u_\perp \sin(q_\parallel x_\parallel)$ creates the illusion that there is a periodicity by definition. Certainly, the values this function takes in $E_\perp$ are periodic. But this is not the end of the story, as we have to inspect how this is translated by the algorithm into a displacement field in $E_\parallel$. With respect to this issue, the value the sine function takes in perpendicular space is not the only variable that comes into play. What also comes into play at a given value of $x_\parallel \in E_\parallel$ is the position of the atomic surface W at $(x_\parallel, x_\perp)$ with respect to the cut. This position will define the minimum value $t(x_\parallel)$ of perpendicular space ondulation it takes to induce a jump. Let us call the end points of W, A and B. We take A to be the point with the higher $x_\perp$-value. The minimum value $t(x_\parallel)$ is the distance between the point where parallel space cuts the atomic surface and one of the endpoints A or B of that atomic surface. It is the comparison between $t(x_\parallel)$ and $u_\perp \sin(q_\parallel x_\parallel)$, rather than the value $u_\perp \sin(q_\parallel x_\parallel)$ alone that will determine if there is a jump or otherwise. The function $t(x_\parallel)$ is quasiperiodic. Let us note the disordered QC generated by the sine wave $u_\perp \sin(q_\parallel x_\parallel)$ as $\mathbb{QC}^*$, and the perfect QC as $\mathbb{QC}$. To give $\mathbb{QC}^*$ as a whole a periodic description through a superspace embedding one needs to lift it to dimension 3. This is obvious from the fact that the Fourier spectrum of $\mathbb{QC}^*$ is spanned by three reciprocal lattice vectors, viz. the two reciprocal lattice vectors that span the Fourier spectrum of the Fibonacci lattice $\mathbb{QC}$ and the vector $q_\parallel$. Note that when we calculate the Fourier spectrum of $\mathbb{QC}^*$ to better approximations, by taking in higher order terms of the Taylor expansion that is used in the calculation (see below), the harmonics with wave vectors $2q_\parallel$, $3q_\parallel$, etc... will enter, such that the statement that $q_\parallel$ is a third generator of the Fourier spectrum is perfectly accurate (see below).

The Fourier spectrum we are talking about here, is the one of the modified QC, i.e. $\mathbb{QC}^*$, which is of course different from the Fourier spectrum of the displacement field $\mathbb{D}$ of phason jumps. The Fourier spectrum of the jump field $\mathbb{D}$ itself is not an experimentally measured quantity. The field $\mathbb{D}$ is not at all periodic, as we show now on the example of the Fibonacci chain.

The cut in the superspace description of the Fibonacci chain can pass either through zero or through two endpoints of atomic surfaces. In fact, if there is at least one, it means that the cut is exactly at the position of inducing a jump. The other end point of that jump will then also be an endpoint of an atomic surface, viz. of that atomic surface one is swapping to. When the endpoint of the first atomic surface is of the type A, then the endpoint on the second atomic surface will be of the type B, because all this together defines a single jump. There will be no endpoint of any other atomic surface on the cut, because else the slope of the cut would be rational. This shows that the number of atomic surface endpoints can only be 0 or 2, and that when there are two of them, they are separated by a jump distance, because they define a single jump and there can be only one jump at a time in the cut. It is therefore possible to take a point O on the cut, such that, to the right of it, the cut does not contain a single endpoint of an atomic surface. Let us take O as the origin of the $x_\parallel$ coordinates.

Choose an arbitrary large positive integer number $K \in \mathbb{N}$. Consider now the interval $[0, K\lambda]$ on parallel space. Consider the atomic positions x_j in this interval.

In each point x_j, it will take a minimal *non-zero* translation $t_j \neq 0$ of the cut along $E_\perp$ to induce an atomic jump, because, by our choice of $[0, K\lambda]$, we know that in x_j the cut does not pass through the end point of an atomic surface. For all the atomic positions x_j within $[0, K\lambda]$ we can define such a minimal non-zero translation t_j. The finite set of strictly positive numbers $|t_j|, j \geq 0$ does not contain 0, such that it has a strictly positive minimum value. Call a this minimum, and take $0 < |u_\perp| < a$. Then $u_\perp \sin(q_\parallel x_\parallel)$ will not induce a single atomic jump over the whole interval $[0, K\lambda]$. By taking increasinging values of K we can make this situation arbitrarily bad. In fact, a functions as a density parameter for the jump field. Nevertheless, the "sine wave" does define a non-zero displacement field, because the Fourier spectrum of $\mathbb{QC}^*$ is different from the Fourier spectrum of $\mathbb{QC}$.

This argument clearly indicates where the logical error lies in the reasoning of people who think that $u_\perp \sin(q_\parallel x_\parallel)$ would define a periodic displacement field. They have focused on the blind spot of the formal appearance of the "sine wave", wihout thinking about the way it cuts through the quasiperiodic pattern of the atomic surfaces.

It is obvious from the previous that λ has not the meaning of a wavelength for the displacement field $\mathbb{QC}$ (see also below). The value a is a measure for the density of jumps on parallel space. Of course, the density is inversely proportional to the average distance between two next neighbour jumps. As the jumps all involve the same jump distance $(\tau - 1)/\sqrt{2+\tau}$, this value defines the maxima of the displacement field defined by the sine wave. The value of this maximum amplitude $(\tau - 1)/\sqrt{2+\tau}$ cannot be changed by varying $u_\perp$. Hence we see that varying the amplitude of the "sine wave" changes the average distance between the maxima of the displacement field, rather than its amplitude. That is certainly not a wave: When we change the amplitude of a true wave, it is exactly the opposite that happens, viz. the distance between its maxima remains the same, while the amplitude of its maxima varies! In fact, $u_\perp$ does not act as an amplitude of the displacement field. It rather defines its density (or the average distance between displacements). The fact that the maximum amplitude of the physical displacement field is forcedly $(\tau - 1)/\sqrt{2+\tau}$ is already sufficient to provide us with a handwaving argument for the fact that $u_\perp$ acts as a density parameter. It is obvious that when $u_\perp$ increases, the effects must become stronger. As the only two possible local

values for the displacement field are 0 and $(\tau-1)/\sqrt{2+\tau}$ only can act on the number of jumps, not on the maximum amplitude of the field.

Our construction shows that λ is not a period of the displacement field. In fact, if X is the first point to the right of O where we have a jump, then at position $Y : |YX| = \lambda$ to the left of X, there is no jump, as it should be if λ were a period. In fact, in general there will even be no atomic position at Y, because in general λ will not correspond to a distance between two points of the Fibonacci sequence. The displacement field has no periodicity at all. In fact, restricting the *domain* of a periodic function of $\mathbb{R}$ to the aperiodic dicrete set of QC lattice points $\mathbb{QC}$ already destroys the periodicity *stricto sensu*, but the situation is rendered worse by restricting also its *range* in $\mathbb{R}$ to a set that only contains two values 0 and $(\tau-1)/\sqrt{2+\tau}$. The function is even no longer quasiperiodic with a two-dimensional $\mathbb{Z}$-module because its periodic embedding requires a superspace dimension of 3 rather than 2. It can thus *a priori* not be claimed on the basis of an argument of (wrongly) assumed periodicity that its Fourier transform would have non-zero components at $Q_B + q_\parallel$, where Q_B defines a Bragg peak of the QC, and $q_\parallel$ is defined by the relation $2\pi/\lambda = q_\parallel$. But we shall see below that this is nevertheless true. It is also *a priori* not obvious that the Fourier decomposition of the "sine wave" would yield $Q_B + q_\parallel$-values that all have the same value of $q_\parallel$. But again, this is true.

To save the periodicity, one might argue that one made indeed an error but that this just consisted in expressing one's ideas through the wrong formula. One might argue that a displacement wave of jumps of the type $u_\parallel \sin(q_\parallel x_\parallel)$ surely can exist. But this is also badly wrong: It is impossible to define any sine wave based on a unique jump distance on the Fibonacci chain, because it would just contradict quasiperiodicity (see our argument about restricting the domains of periodic functions). As we already stated, λ will only in very exceptional cases correspond to a distance between two atomic positions of the Fibonacci sequence. Hence the positions P were the atomic jumps PQ would have to take origin will in general fall somewhere in between the atomic positions of the quasicrystal. Moreover, the intensity of the diffuse pattern will not scale with $Q^2_{\perp,B}$, when the polarization is along $u_\parallel$.

Finally, we must make a remark about the fact that our approach has been one-dimensional. We can see that our objection can be avoided by taking the direction of propagation along the *periodic* direction of a "two-dimensional" quasicrystal (like a decagonal phase). In that case the "sine wave" does not run into the same conceptual difficulties. We may note that this reminds us of the electron microscopy observations of a "large phason jump" by Edagawa et al. [5], but these authors remain cautious about the interpretation of their data in terms of a "sine wave" [6]. Secondly, domain walls have been observed in icoshaedral phases by electron microscopy, with a high density of tile flips. Only a few orientations of such domain walls will be energetically allowed, and it would also require a lot of energy to move them as a whole. The Fourier transform of such a plane is a line. This represents then a correlation in the atomic positions where the flips occur, in the sense that they are all located in the same plane. But the $q_\parallel$ vectors that one could associate with the lines mentioned would in the "sine wave" picture correspond to a correlation in the direction normal to the plane, not within it. The domain wall can also not be of the "sine wave" form proposed by the authors, as the displacement field defined by a "sine wave" is not two-dimensional. And within the plane, relaxating phason jumps will not be any more correlated than along a Fibonacci chain (see also below).

In conclusion: There does not exist any true, periodic sine wave of atomic jumps in physical space on a QC.

B. Calculation of the diffraction pattern of the disordered QC

We must criticize the way a single Fourier component in the diffuse scattering pattern is given physical meaning by Francoual et al. Let us first point out that a Fourier componenent of the diffuse scattering corresponds to a density wave. In the theory of Jaric' and Nelsson, [2] the basic formalism is indeed based on density waves. These density waves are not restricted to the discrete set of atomic positions of the QC as de Boissieu et al. propose in their interpretation, but they are defined on the whole continuum of parallel space, where they are allowed to cancel mutually, etc... In fact this mutual canceling is what happens most of the time because the QC is a discrete density. The diffraction diagram as a whole, with the Bragg peaks and the diffuse scattering, describes thus a priori a field of *atomic positions*, not a field of *atomic jumps*.

Let us calculate the diffraction pattern of a Fibonacci chain, whose corresponding two-dimensional "supercrystal" has been modulated (in superspace) by the "sine wave" $u_\perp \sin(q_\parallel x_\parallel)$. After applying the "sine wave" the (non-decorated) two-dimensional lattice will be a truly modulated crystal, whose two-dimensional diffraction pattern will be given by Bragg peaks $\delta(\mathbf{Q}-\mathbf{Q}_B)$ and satellites $\delta(\mathbf{Q}-(\mathbf{Q}_B \pm \mathbf{q}_\parallel))$. Let us note the window function of the strip method as W. The points of this modulated lattice that fall into the strip $W \times E_\parallel$ project to the atomic postions of the disordered QC. Alternatively, we can decorate the atomic positions of the modulated "supercrystral" with atomic surfaces W and cut with $E_\parallel$. What we have to calculate is:

$$\int e^{\imath \mathbf{Q}\cdot\mathbf{r}} \sum_{(M,N)\in\mathbb{Z}^2} \delta(\mathbf{r}-\mathbf{r}_{M,N}-\mathbf{u}_{M,N}) * W(\mathbf{r})\, d\mathbf{r} = (\int e^{\imath \mathbf{Q}\cdot\mathbf{r}}\, W(\mathbf{r})\, d\mathbf{r}) \times (\sum_{(M,N)\in\mathbb{Z}^2} e^{\imath \mathbf{Q}\cdot(\mathbf{r}_{M,N}-\mathbf{u}_{M,N})}) \tag{1}$$

where $\mathbf{u}_{M,N} = \mathbf{u}_\perp \sin(\mathbf{q}_\parallel \cdot \mathbf{r}_{M,N})$, and where we have indexed the nodes of the "supercrystal" by (M,N). When $u_\perp$ is small we can develop $e^{\imath \mathbf{Q}\cdot\mathbf{u}_\perp \sin(\mathbf{q}_\parallel\cdot\mathbf{r}_{M,N})}$ in a Taylor expansion, such that we obtain for the second term of the product in the righthand side of Eq. (1):

$$\sum_{(M,N)\in\mathbb{Z}^2} (\, e^{\imath \mathbf{Q}\cdot\mathbf{r}_{M,N}} + (\frac{\mathbf{Q}\cdot\mathbf{u}_\perp}{2})\, (e^{\imath(\mathbf{Q}+\mathbf{q}_\parallel)\cdot\mathbf{r}_{M,N}} - e^{\imath(\mathbf{Q}-\mathbf{q}_\parallel)\cdot\mathbf{r}_{M,N}})\,) \tag{2}$$

The first exponential will lead to the set of Bragg peaks, the second and third exponentials will lead to Bragg peaks shifted by $\pm\mathbf{q}_\parallel$ and further weigthed with $\frac{1}{4}(Q_{\perp,B}u_\perp)^2$. We see thus that there will be satellites.

The situation is completely analogous to what we observe with a phonon modulation on a periodic crystal. E.g. a longitudinal phonon displacement field $u\sin(qx)$ on a periodic one-dimenionsal crystal will introduce satellites at $\pm q$ with respect to each Bragg peak Q_B. These satellites correspond to density waves with wavelength $2\pi/(q+Q_B)$. The function $u\sin(qx)$ is defined over the whole of $\mathbb{R}$. By using it to define a displacement wave, we restrict it to the discrete set of the lattice nodes of the crystal. As a corollary its Fourier transform changes from $\delta(Q\pm q)$ for the function whose domain is $\mathbb{R}$, to the infinite set of Dirac measures at Q_B+q. In other words, the price to pay for restricting a continous function to a lattice, is that one must convolute its Fourier transform with the reciprocal lattice. We see that $2\pi/(Q_B+q)$ corresponds to a density wave, not to a period, and that we need a whole set of them in order to render the intensity in between the atomic positions of the phonon-modulated crystal zero. The result for the QC follows the same philosphy, with a more complicated reciprocal lattice.

It may look surprizing and counterintuitive that we find that the position of these satellites for the QC is independent from the amplitude $u_\perp$, while we have shown that for small enough amplitudes there will be much longer distances between the jumps than given by $2\pi/q_\parallel$. But this is nevertheless correct (see below). This formalism corresponds to the paradigm de Boissieu's uses. It corresponds to the idea of Equations [42] and [43] in the paper by Janssen et al.

C. Criticism of the wave approach

There are very serious problems with the approach outlined above:

1. Topological problems

The phonon dynamics of the Fibonacci chain can in general not be obtained by just making a cut through the phonon dynamics of a two-dimensional lattice, as is suggested by the equation [42] in the paper of Janssen et al. The phonon problem of the Fibonacci is an unsolved problem of an horrendous difficulty. This has several origins:

(a) The topology of the problem is different from that of the phonon problem on an unbounded lattice. Neighbour interactions with atomic surfaces that are out of the acceptance window, i.e. out of the superspace strip, are just cut away. This gives rise to boundary conditions. This is the discrete analogon of a wave equation on a domain D. In the example of the Fibonacci chain with only first neighbour interactions, the boundary problem is quite extreme, as every atomic position is on the boundary ∂D of the domain D of the wave equation, such that D has an empty interior. In fact in $\mathbb{Z}^2$ each lattice node has 4 first neighbours, while on the Fibonacci chain each point only has 2 neighbours. This can be formulated as a condition that is the discrete analogon of a boundary condition of the type $\mathbf{e}_n \cdot \nabla\psi = 0$, on ∂D for the wave equation $\Delta\psi = 1/c^2\partial^2\psi/\partial t^2$; here $\mathbf{e}_n$ is the normal to the boundary. But in the discrete problem $\mathbf{e}_n$ does not vary smoothly. It takes different orientations, depending on the question if its first neigbour environment corresponds to $L\cdot L$, $L\cdot S$ or $S\cdot L$ (where L and S are the interatomic distances).

(b) Using another approach, the two-dimensional lattice must be considered to be decorated with atomic surfaces, which implies that there is an infinite number of "atoms" s in the unit cell (M,N). The dynamical matrix that defines all the interactions between pairs of atoms in first-neighbour cells $(M,N,s_1),(M+1,N,s_2)$, etc.., is infinite.

Postulating wavelike solutions as is done e.g. in Eq. [43] of the paper of Janssen *et al.* is, at least in general, very wrong. There is not the slightest proof that this is reasonable in general. Rigorous studies of the phonon problem on the Fibonacci chain through the transfer matrix method seem to indicate that such an *Ansatz* is just wrong. This in turn raises the question if there are phason "modes" (in the usual sense of a wave) all together.

A QC is not a continuum but a discrete lattice. The discrete approach prevails thus over the continuum approach. The continuum approach is obtained by taking the long-wavelength limit of the discrete problem. When we propose

thus a continuous phonon mode, it must be validated by checking if it makes sense by taking the limit of the discrete solutions. For continous phonon modes this makes sense. For phasons the situation is all together different:

(a) There are a number of famous papers by Levitov and others [7] that show that in general phasons in QCs crystals are not continuous.

(b) In contrast with what happens with the phonon modes, the displacement field of the postulated phason wave is not at all sinusoidal in parallel space. When make the discrete graph $u(x_\parallel)$ vs $x_\parallel$ of a long-wavelength phononlike sine wave diplacement field on the Fibonacci chain, and we look at this graph from very far, such that the atomic positions seem to fill a continuum, the picture of the sine wave will become clearly visible. It is this that gives sense to the continuum limit for phonons. When we do the same for the displacement field of the phasonlike "sine wave", we will not be able to make sense of it: In certain regions the graph will appear to consist of two horizontal lines $u(x_\parallel) = (\tau - 1)/\sqrt{2+\tau}$, and $u(x_\parallel) = 0$, as though it would correspond to a two-valued "function". The two lines will have grey shading , rather than being just black. In other regions, it will appear to be just $u(x_\parallel) = 0$ alone. It is thus no longer possible to keep telling accurately what is going on in the QC when we move out to inspect it from further away with a coarsened resolution. Taking the "long-wavelength limit" ceases to be a self-evident concept or method, because (1) one cannot define limits for discontinous functions, and (2) the displacement field cannot be described or regularized as the restriction of a continous wave to the QC.

(c) The postulated phason modes lead to notions of long distance coherence between atomic jumps that are completely unphysical. This will be developed below: The closer the jumps are in time, the farther away they must be in space! Also, in all experimental studies of atomic self-diffusion studies, the paradigm and the evidence is that atomic jumps are not correlated over long distances.

(d) We may add to this that the infinite lattice allows also for modes with an exponential decay in space. These are excluded in a real crystal because these exponential waves diverge, in one direction or another, but in the QC, divergence along the $x_\perp$ direction is harmless, due to the restriction to the strip.

2. Incompleteness of the data

While in a crystal the spectral reponse for a phonon modulation is rigorously the same at every Bragg peak, in a QC the intensity is different at every Bragg peak. It is thus no longer representative to study a single Fourier component at $Q_B \pm q_\parallel$ for a given $q_\parallel$-value at a choosen Bragg peak defined by Q_B. One has a whole set of satellites, with their respective intensities. It would be normal to check experimentally that these relative intensities at $Q_B \pm q_\parallel$ for various Bragg peaks are compatible with the calculation of the picture of a superspace phonon. It would also be normal to check experimentally if the same $q_\parallel$-value at different Bragg peaks Q_B yields identical relaxation times.

D. Solution of a paradox

We must now treat the apparent contradiction that $q_\parallel$ does not correspond to a period of the displacement field $\mathbb{D}$, while it appears in the diffraction spectrum.

1. An important distinction

Let us first note that we must make a difference between $\mathbb{QC}^*$, which is the whole disordered quasicrystal, i.e. a set of *atomic positions*, and the displacement field $\mathbb{D}$, which is a set of *atomic jumps* located at certain atomic positions, and which can have a very large average distance between the points where it is non-zero.

2. The disordered quasicrystal

It may look paradoxycal that the distance between successive jumps in the displacement field is much larger than the wavelength of the sine wave, while the wave vector $q_\parallel$ shows up in the Fourier spectrum of the disordered QC. It is nevertheless true. This is actually no more paradoxical than that the reciprocal lattice vectors of the the $\mathbb{Z}$-module of the superspace embedding (like $2\pi/L$ and $2\pi/S$ in the Fibonacci chain) appear (as generators) in the diffraction pattern of the QC, while they are not periods. In fact, we may consider the superspace lattice, decorated with its atomic surfaces as modulated by the sine wave $u_\perp \sin(q_\parallel\, x_\parallel)$. Following the method of Dewolff, Janssen and Janner, [9] we can approach such a modulation by lifting the reciprocal lattice to a three dimensional periodic lattice. This allows us to see that the Fourier module will now have the additional generator $q_\parallel$. And this does not depend on the amplitude $u_\perp$. We can also see that the whole spectrum of harmonics of $q_\parallel$ can in principle occur. It is just that they have too weak intensities when they are not observed. E.g. when $u_\perp$ becomes larger or if the modulation is not sinusoidal, they may show up. (E.g. if in our derivation of Eq. (2) one develops the exponential to second order, the harmonics $2q_\parallel$ come into play). This can perhaps even be used to find out how we can describe the system without

violating the conservation of atoms when the modulation is strong. We can see from this very clearly that λ is not a period, just like the interatomic distances L and S in the Fibonacci chain are not periods.

3. The displacement field

Let us now check how it is with the paradox for the proper displacement field $\mathbb{D}$. The Fourier spectrum of the displacement field is *not* the difference of the Fourier spectra of $\mathbb{QC}^*$ and $\mathbb{QC}$; for a displacement from x_1 to x_2, $e^{\imath Q(x_2-x_1)} \neq e^{\imath Q x_2} - e^{\imath Q x_1}$, also not in first order. But let us inspect what we obtain when we subtract the Fourier transforms of $\mathbb{QC}^*$ and $\mathbb{QC}$. It would yield the Fourier transform of a set of dipoles: There is a negative Dirac measure at each starting point x_1 of a jump, and a positive Dirac measure at each end point x_2 of a jump. Hence we have $\mathcal{F}[\sum(\,\delta(x-x_2)-\delta(x-x_1)\,)]$. What we would need to calculate is $\mathcal{F}[\sum(\tau-1)\,\mathbf{e}_\parallel\,\delta(x-x_1)\,/\sqrt{2+\tau}]$. In the superspace description the difference spectrum gives rise to just a subtraction of the densities of the atomic surfaces. At one end of an atomic surface the subtraction yields a positive density, in the region where the atomic surfaces do not overlap, at the other end a negative density. When the jump occurs to the right it is the upper end of a subtracted atomic surface where the positive density occurs, and the lower end of the neighbouring subtracted atomic surface where the negative density occurs. We certainly need these pieces of density to make a calculation of the Fourier transform of the displacement field, but we need a method to select only the negative pieces, and weighting it with $\pm(\tau-1)\,\mathbf{e}_\parallel/\sqrt{2+\tau}$ where the sign depends on the question if the jump occurs to the left or to the right.

To simpfly the problem it is better to consider the two displacement fields $\mathbb{D}_1$, defined by $u_\perp(1+\sin(q_\parallel\,x_\parallel)\,)$, and $\mathbb{D}_2$ defined by $-u_\perp$. The first one will only yield jumps to the right. The second one only jumps to the left. Subtracting the Fourier transform of the perfect QC from the Fourier transform of the QC modulated by $\mathbb{D}_1$, will correspond to the Fourier transform $\mathcal{F}_1 = \mathcal{F}(C_1)$ of a set of dipoles C_1, that are all oriented to the right. Call the set of origins of the jump vectors of $\mathbb{D}_1$, B_1, and a dipole oriented to the right d. Then $\mathcal{F}_1 = \mathcal{F}(B_1 * d) = \mathcal{F}(B_1)\mathcal{F}(d)$, from which $\mathcal{F}(B_1)$ can be calculated. (There is a problem with this, in that $\mathcal{F}(d) = e^{\imath Q(\tau-1)/\sqrt{2+\tau}} - 1$ contains zeros, but these do not occur on the satellites). Similarly we can calculate the Fourier transform of the set $\mathcal{F}(B_2)$ of origins of jump vectors defined by $\mathbb{D}_2$. If we take $\mathcal{F}(d)$ to deconvolute, the set B_2 will be negatively weighted. With this weighting $(\tau-1)\,(\,\mathcal{F}(B_1)+\mathcal{F}(B_2)\,)/\sqrt{2+\tau}$, will then be the Fourier transform of the displacement field $\mathbb{D}$. When in a given point both a jump to the right occurs within in $\mathbb{D}_1$ and a jump to the left within $\mathbb{D}_2$, then the two jumps will conveniently add up to zero. In all other points, the jumps will occur with the proper weighting. This point hinges of course on the fact that the amplitude of $u_\perp$ must be small enough. If the amplitude of the sine wave is too large, there will be successive jumps of a same atom, and the jump distances will not all be $(\tau-1)/\sqrt{2+\tau}$. In conclusion, also for $\mathbb{D}$ the solution of the paradox is that $q_\parallel$ does indeed occur in the Fourier spectrum.

4. Conclusion

We see that doubling the amplitude $u_\perp$ will *in first order* double the intensity of the satellites both in the Fourier spectrum of $\mathbb{QC}^*$, and in the Fourier spectrum of the displacement field $\mathbb{D}$. But of course, this does not imply that $\mathbb{QC}^*$ or $\mathbb{D}$ would be periodic, or that $\mathbb{D}$ would be physically meaninful (see below).

E. Clearly distinguishing the various fields

Let us resume the situation. A single Fourier component at some value $Q_B + q_\parallel$ in the diffuse scattering is a continous density wave and as such cannot be the Fourier transform of a field of phason jumps. Even a set of Fourier components $Q_B + q_\parallel$ with the same $q_\parallel$-values does not correspond to a periodic wave of jumps:

(a) It just corresponds to a disordered quasicrystal $\mathbb{QC}^*$, not to the field of jumps that would permit to go from the perfect quasicrystal $\mathbb{QC}$ to $\mathbb{QC}^*$. (Of course the "difference" between the pristine $\mathbb{QC}$ and the disordered $\mathbb{QC}^*$ corresponds to a field of jumps).

(b) We have proved that when a "sine wave" transforms the perfect $\mathbb{QC}$ into the disordered $\mathbb{QC}^*$ with such a $q_\parallel$-based set of Fourier components with wave vectors $Q_B + q_\parallel$, then this does not imply at all that the transformation from the perfect $\mathbb{QC}$ to the disordered $\mathbb{QC}^*$ would be be based on a *periodic* wave of atomic jumps at positions of $\mathbb{QC}$.

Thus in a diffuse scattering pattern that uniquely corresponds to the disorder generated by tile flips (e.g. in the Monte Carlo simulations of Tang et al. [10,11]), the intensity at $Q_B + q_\parallel$ can never have physical meaning, as it corresponds to a continous sine wave by definition. The diffuse scattering intensity $Q_B + q_\parallel$ in the disordered-tiling model is only a Fourier component, without any real physical meaning. It corresponds to a density wave for the disordered quasicrystal $\mathbb{QC}^*$. To obtain a physical meaning we must combine all Fourier components $Q_B + q_\parallel$ at all

Bragg peaks. And in a given point of space, these Fourier components will most of the time just cancel mutually to yield zero density, as the atomic positions are a discrete set. By combining all contributions with wave vectors $Q_B \pm q_\parallel$, corresponding to a single $q_\parallel$ linearly with their appropriate intensities, we may reconstruct the wole disordered tiling $\mathbb{QC}^*$, produced by the "sine wave" with wavelength $2\pi/q_\parallel$ from the perfect quasicrystal $\mathbb{QC}$.

We see thus that we have the following scheme:

$$
\begin{array}{ccc}
Bragg\ peaks\ \{\, Q_{M,N} \parallel (M,N) \in \mathbb{Z}^2 \,\} & \underset{\pm\, q_\parallel}{\longrightarrow} & Bragg\ peaks\ \{\, Q_{M,N} \pm q_\parallel, 0 \parallel (M,N) \in \mathbb{Z}^2 \,\} \\
\downarrow \mathcal{F} & & \downarrow \mathcal{F} \\
perfect\ \mathbb{QC} & \underset{aperiodic\ field\ of\ phason\ jumps\ \mathbb{D}}{\longrightarrow} & disordered\ \mathbb{QC}^* \\
\downarrow lift & & \downarrow lift \\
W * \mathbb{Z}^2 & \underset{periodic\ modulation\ wave\ q_\parallel}{\longrightarrow} & W * modulated\ \mathbb{Z}^2
\end{array}
$$

Here only the last line in the diagram corresponds to superspace quantities. The two other lines refer to parallel spaces: The first line to reciprocal space, the second line to direct space.

It is important to realize that the periodic modulation wave is only the Fourier transform of $q_\parallel$ in the superspace sense, and that the Fourier transform of $q_\parallel$ in $E_\parallel$ has no physical meaning, e.g. it does not correspond to the aperiodic field of phason jumps $\mathbb{D}$. The periodicity of the modulation in superspace does not imply that the field of phason jumps $\mathbb{D}$ would be periodic. This field is definitely aperiodic. The field of the phason jumps is not directly measured, and its Fourier transform is not a single value $\delta(Q - (Q_B \pm q_\parallel))$. The only quantities that can be directly measured are the ones on the first line of the diagram.

F. Further criticism of the "sine wave" interpretation

The way the authors analyse the isolated diffuse intensities at $Q_B + q_\parallel$ by drawing the relaxation time as a function of $q_\parallel$ to prove that it would correspond to a diffusive mode of jumps is approximate and incomplete. As we have seen it corresponds to a physically meaningless density wave. To obtain meaningful jumps in the case of a sine wave modulation one would have to study a whole set of $Q_B + q_\parallel$-values simultaneously, as only a whole set will yield the disordered QC, created by a field of jumps. There is an experimental problem that we do not know exactly how the corresponding amplitudes are to be combined . This is the very same problem as for reconstructing the perfect QC from the intensities of its Bragg peaks. Even if we leave this practical problem aside, there remain conceptual problems.

1. The postulated diplacement field is unphysical

We have seen that when $u_\perp$ is sufficiently small, $q_\parallel$ can correspond to a field that induces very few atomic jumps that are very far away from each other, with no jumps at all in between. If we imagine $u_\perp$ as built up from infinitesimal contributions $\delta u_\perp$, we can have the sine wave continuously growing from 0 amplitude to the small amplitude $u_\perp$. In a given large patch of the QC, we will see then the tile flips occuring consecutively, one by one, and the consective atomic jumps will be separated by very large distances. The closer the phason jumps are in time, the further away they will be in space! That does not evoke an image of a coherent process with correlated jumps! For correlated jumps, we would like to see them not too far one from another, such that they can interact by a force. Of course, we can repeat the same argument for any increase of $u_\perp$ from a finite reasonable value $u_\perp^{(1)}$ to another finite reasonable value $u_\perp^{(2)}$. We have not proved such an argument rigorously for the case 0 is not one of these two values, but we doubt that anyone would claim the opposite to be true. Now what one sees in a speckle experiment at a single value of $Q_B + q_\parallel$ is exactly a change of intensity (in the form of an exponential decay) between two values close to a main value, that is interpreted to correspond to such a change of amplitude of the "sine wave". This does not correspond to any normal interpretation of the concept of a *mode*.

2. Lack of uniqueness

There exist other modulations than of the phonon type, e.g. compositional modulation, or magnetic modulation. They can be calculated by exactly the same formalism as for a displacive modulation and will yield satellites at

the very same positions in reciprocal space. The information is thus *unspecific*. We cannot claim without further justification that we have a displacement modulation.

When a specific interpretation for an observation is claimed, and when the interpretation is *a priori* not unique, like it is the case with satellites in a Fourier spectrum, the choice of the specific interpretation must be motivated. It can happen that the motivation given for the claim is not sufficient to make away with all possible ambiguities. This means that the charge of proof for the claim has not been met appropriately and that there are loopholes in the justification. When this is the case, this will in general give rise to objections. A good way to point out that the uniqueness of an interpretation has not been proved, is giving the possibility of a counter example. It is pointless and not appropriate to reply to the suggestion of such possibilities that one would have to prove them, since that would amount to a reversal of the charge of proof. E.g. when one wants to point out a loophole in a proof of a mathematical theorem, this does not imply that one would have to prove that the *theorem* is wrong. Pointing out a possible loophole shows that the proof is incomplete, and as such that the *proof* of the theorem is wrong. In an inductive science like physics, there is no parade against infinite scepticism. Therefore, the objections must remain reasonable. All possible *reasonable* objections must be duly incorporated in the discussion of the interpretation of the data, and it must be clarified how bad they could be. We want to raise the reasonable objection that the interpretation of the diffuse scattering data is in bad lack of proof of uniqueness. The complicated structure of the present paper, with its many subdivisions in sections, subsections, etc... is probably enough to show the profound confusion this has produced.

1. Chemical Disorder. The authors have speculated several times [6,12] about the possibility that chemical disorder could be a kind of phason defect. In reference [6] it is e.g. stated that de Boissieu "wonders if the perp-space Debye-Waller factor component is just another way of accomodating chemical disorder of atoms". It is also stated that "it is in agreement with the diffuse scattering". It is thus obvious that this possibility must be ruled out, before one can claim that the modulation is displacive. As pointed out above, chemical modulation can be treated by exactly the same formalism as displacive modulation. In that case, it is the complicated decoration of the atomic surface that interacts with the "sine wave". The change of the type of atom is then possible by varying the amplitude of the "sine wave", and the variation can occur without necessarily implying a phason jump. In other words, in this case, the "reason" for the change of type of atom is not given by the superspace description *in se*. We may finally add that in AlMnPd, a strong component of diffuse scattering has been observed at small angles [13], that is flat with Q and has an intensity that corresponds to what one would obtain from a calculation assuming complete chemical disorder. This seems to indicate that the presence of at least some chemical disorder has to be taken as a very serious possibility. Introducing a chemical modulation "sine wave" can be done without any reference to elasticity. This implies that when the data are due to chemical disorder, they are irrelevant for the *elastic* stability issues raised by the random tiling model, as Widom's theory is based on displacive modulation.

2. An alternative type of displacive modulation. But even if we stick to a displacement modulation, there is an alternative. Imagine a QC $\mathbb{QC}^{(\alpha)}$ obtained by replacing the atomic surfaces W of $\mathbb{QC}$ by other atomic surfaces $W^{(\alpha)}$ that are tilted by a small angle α with respect to W, and whose lengths are adjusted such as to keep the condition of conservation of atoms satisfied. The whole derivation of the calculation of the Bragg peaks of $\mathbb{QC}^{(\alpha)}$ remains the same. Hence, the positions of the Bragg peaks are the the same in $\mathbb{QC}^{(\alpha)}$ and $\mathbb{QC}$. Only the intensities of the Bragg peaks are changed: The value of $Q_\perp$ of the Bragg peak that occurs in the expressions of the intensities $(\sin Q_\perp \ell)/Q_\perp \ell$, must be replaced by the oblique coordinate of the Bragg peak $Q_\perp/\cos\alpha$, but this remains an expression that only depends on the $Q_\perp$-value of the Bragg peak, not on its $Q_\parallel$-value). As the length ℓ of the atomic surface also enters the expressions, it must also be properly modified to the new value.

Again we modulate the positions of the undecorated "supercrystal" with a sine wave $u_\perp \sin(q_\parallel x_\parallel)$. Note that this is slightly different from cutting the tilted atomic surfaces with the "sine wave", because in the latter, the value where the amplitude of the "sine wave" must be calculated would be slightly shifted along $E_\parallel$. Call the resulting QC $\mathbb{QC}^{(\alpha)*}$. As rigorously the same kind of calculation comes into play for the satellites as for the Bragg peaks, we see that also the satellites of $\mathbb{QC}^{(\alpha)*}$ will be in exactly the same positions as in $\mathbb{QC}^*$ and that only their intensities will be different. Note that, in the end, the factor $\mathbf{Q}\cdot\mathbf{u}_\perp$ can only lead to a $\mathbf{Q}_\perp$-dependence, due to the scalar product with $\mathbf{u}_\perp$. Hence, there is no qualitative difference between the diffraction diagrams of $\mathbb{QC}^{(\alpha)*}$ and $\mathbb{QC}^*$. It may disturb at first sight that the modulation produces displacements along $E_\parallel$, but this not conceptually different from the situation with phason jumps. It is thus a possible alternative.

This alternative is more physical. In $\mathbb{QC}^{(\alpha)*}$ there will be small atomic displacements in every atom of the QC, even when $u_\perp$ is very small. Nobody will find it difficult to believe from such a picture that the atoms are elastically coupled from neighbour to neighbour. There is thus no postulate of spooky correlations over large distances between two isolated jumps, while the whole intermediate region remains unaffected, as in $\mathbb{QC}^*$. There is no such problem that the closer two jumps are following each other in time, the further away they must be in space. Moreover, when

$u_\perp$ is very small, these small atomic displacements will outnumber the very scarce atomic jumps.

An essential point is that the real-space atomic diplacements created by the fluctuation of the cut are much larger and not harmonic in the model of the authors. An atomic jump can only occur in an double-well potential, and the function that describes such a potential is at least of the fourth degree. The potential is thus not harmonic. We think that our alternative leads to a less unphysical interpretation of the data, without a real necessity to invoke tile flips as the basic ingredient. The displacement fields are just like classical phonon displacement fields, except for the fact that they are parameterized by other, perpendicular space coordinates. We also want to stress that the procedure of tilting the atomic surfaces is only a first-order approximation to illustrate the idea. In reality, we should introduce a kind of devil's stair case in order to account for far-away changes of configurations. Furthermore, we should modify the atomic surfaces in such a way that it does preserve the symmetry. Duneau [14] has shown that in an admittedly wrong, polynomial approach for icosahedral symmetry, the minimal degree of the polynomials that comply with this condition is three. The idea of modulating the atomic surfaces finds its confirmation in numerical calculations of the dynamics, where it has to be introduced in order to relax the initial system. The idea of modulating the atomic surfaces is also present in a work of Steurer, [15] who calls it the IMS setting (as opposed to the QC setting).

It remains to explain how the diffuse scattering resulting from our alternative model could decrease when the temperature is raised, but there are many possibilities to do this. Widom's instability [16], with tile-flip phason elasticity replaced by a phason elasticity based on small atomic displacements, would already to the job. But it is even not necessary to claim that the QC would not be stable. A mere softening of the elastic constants would do. There are many examples known of elastic constants or phonon modes that soften in a given temperature range, without triggering any phase transition at all.

It is even not necessary to invoke a softening of the elastic constants. It could just be that the system acquires supplementary possibilities to reduce the strain. One possibility is e.g. that thermal vacancies contribute to the relaxation of the observed strain fields. It is quite plausible that their number becomes significant at the temperatures where the fluctuations observed by the authors set in. Also fast phason hopping between two positions could help in relaxing strain fields.

We may add a note about the fact that the diffuse scattering seems to follow predominantly the phason elastic constants. The diffuse scattering reveals "frozen" phasons rather than "frozen" phonons. Perhaps one thinks this is surprizing, and perhaps one thinks it must be full of highly significant information. But a frozen phonon would stipulate an amplitude for an atomic displacement field in a certain place, just on the basis of its position coordinate, without any hindsight on its environment. The phonon wave could be just totally out of phase with respect to the reality of the local environment. In sharp contrast with this, the phason variable is a parameter that refers to the local environment. In order to know the environment of a point x in a crystal, it suffices to take the non-integer part of x/a, where a is the lattice parameter. Hence x is a good quantity to define a phonon displacement field in a crystal, as it contains the necessary information about the local environment. In a QC, $x_\parallel$ is not a good parameter to define a displacement field in this respect. The parameter $x_\perp$ is much better suited to define the local environment. We think that this indicates that a displacement field in a QC can only reasonably expected to be of the frozen phason type.

3. Other possibilities? We cannot pretend that the list of possible alternatives has been exhausted with these two counter examples. The whole problem with the interpretation of the diffuse scattering is that it is hopelessly difficult, due to the large amount of diffuse scattering scenarios in general, combined with the complexity of the structure and of quasiperiodicity, more specifically. It is for this reason that it is impossible to propose with certainty an alternative interpretation of the diffuse scattering data, and that we were obliged in the previous lines to limit ourselves to suggesting the existence of alternative possibilities.

We should also warn the reader against a play of words. As the authors have rightly pointed out, the word "phason" has been used with many different meanings. This was also pointed out by Janssen et al [4] and, much earlier, in reference [17]. Therefore it is all the more surprizing that the authors rely on the mere fact that their data can be described in terms of "phason elasticity" to identify them without any further discussion with phason jumps, as though this would be self-evident. Clearly concepts cannot be identified on the mere basis that they are homonyms. The two examples of chemical disorder and small non-phason-jump small atomic displacements clearly may have physical realizations that could be described by a "wave" with perpendicular-space polarization, even if they both can have also other physical realizations that contain a parallel-space component in the polarization.

3. A possible ambiguity

There is not a single $q_\parallel$-value but a whole distribution of them. It is obvious that there is an ambiguity that has not been addressed: The perpendicular-space amplitude $h_\perp$ of the ondulation is not unambiguously defined: At each atomic position $h_\perp$ can be varied at will as long as it does not lead to a swap of atomic surface. In between the

atomic positions $h_\perp$ can also be varied. The physical situation in $E_\parallel$ allows thus for more than one decomposition in "sinosoidal phason waves", it i.e. in sets $Q_B + q_\parallel$. This is perhaps connected to the fact that $Q_B + q_\parallel$ can also be written as $Q_B^* + q_\parallel^*$, where Q_B^* is another Bragg peak. The information at $q_\parallel$ is thus not unambiguous. It will contain a strong contribution from a strong Bragg peak and other, weaker contributions. We can reduce the importance of the latter problem by making the speckle measurements in the vicinity of a strong Bragg peak Q_B, such that the decomposition $Q_B + q_\parallel$ will dominate over all other ones.

4. How is a non-sinusoidal ondulation of the cut Fourier-decomposed?

While the Fourier transform of a sine-wave modulation will produce a set of $Q_B + q_\parallel$-values and corresponding intensities that allow to reconstruct a field of pure phason jumps, it is not granted that the Fourier decomposition of the whole ondulation of the cut can be kept stepwise, putting each atomic jump as a whole into one set of $Q_B + q_\parallel$-values that reconstruct a sine wave that will generate this jump. Two jumps in succession of the same atom, that do not result in a zero displacement, will not forcedly lead to a Fourier decomposition that contains pure phason jumps in the $Q_B + q_\parallel$-sets. We see that this raises the question of how large the amplitude of the sine wave can be without ceasing to be sufficiently small for the validity of the approximation that is claimed to justify the formalism. In our development above, we have more or less acted as though the amplitude $u_\perp$ of the "sine wave" were less than the amplitude of W, in order to remain within the limit of small amplitudes, and to stay away from problems like violating the criterium of the conservation of the number of atoms. This is not an important restriction, as what we want to deal with in general are small variations of the amplitude, and the only thing that really matters is that the total ondulation of the cut conserves the number of atoms. In any case, it may thus well be that a set of measured $Q_B + q_\parallel$-values with its intensities defines fractional jumps. What will remain of the interpretation of the diffusion constant analysis in terms of a "sine wave" of jumps if this is the case? The data from $Q_B + q_\parallel$-sets may well reflect small atomic displacements that are not whole phason jumps. A field of such small atomic displacements cannot be distinguished from a field where the displacements are not the result of only phason jumps. This underlines once more our argument that it cannot possibly be claimed on the basis of the data, that what one observes is due to disorder induced by tile flips.

G. Summary

In conclusion of this Section, we have shown that the interpretation of an intensity at $Q_B + q_\parallel$ in terms of a *periodic* displacement field with wavelength $2\pi/q_\parallel$ of correlated jumps is wrong and unphysical. It contains several tacit, misleading conjectures, whose truth is just taken for granted, while they are obviously fallacies.

III. COHERENT DELUSIONS

A. Important Warning

Coherent scattering signals collect contributions from all particles of the system that have a non-zero coherent scattering amplitude. It is thus a many-particle signal. Despite all possible folk lore, this should not be overinterpreted. That the scattering is coherent does not prove that it corresponds to a "collective" signal or to a signal of some correlations, as the latter implies that the particles are no longer independent and move in a concerted, correlated fashion, due to some coupling or interaction, as is e.g. the case for phonons. That can very well be the case, but it is not a necessity. The oppositions independent vs. correlated, coherent vs. incoherent, single-particle vs. many particle cannot be amalgamated.

(1) Coherent scattering with a certain structure can also occur in a system wherein the dynamics of all particles are totally independent. A clear example of this are the Monte Carlo simulations of Tang et al. [10,11] The tile flips are here completely random and independent, but they lead to a clear coherent signal that is actually very similar in its reciprocal-space properties to the one observed by the authors (It is only the temperature dependence that permits to invalidate the pristine random tiling model used in this simulation). The same conclusions have been reached by Naumis et al. [18] They stated that in the long-wavelength limit one cannot draw conclusions as to the presence of correlations, because also totally uncorrelated jumps will give rise to a coherent signal.

(2) Coherent scattering signals can be obtained from the dynamics of a single particle, provided it is a coherent scatterer. This is independent by definition.

(3) Conversely, many-particle systems can give rise to incoherent signals, even if the dynamics are strongly correlated. It just suffices that the particles are incoherent scatterers.

The reader who wants to come to terms with these issues is refered to reference [19]. We will discuss how one calculates coherent scattering signals below.

B. Atomic jumps in self-diffusion problems are not correlated

It is generally admitted that phason jumps can give rise to long-range atomic self-diffusion. The idea has been pushed to the extreme in the model of Kalugin and Katz [20], which predicted extremely fast diffusion, but no evidence has been found in favour of this prediction. What has been observed is extremely fast phason hopping. We think that there is no absolute proof for the possibility that phason jumps lead to diffusion, due to the simultaneous presence of vacancy diffusion. But this is nevertheless generally believed to be true, because it is hard to see an obstacle against it. In any case, Francoual et al. make an analysis of their data in terms of a macroscopic diffusion constant. But in all atomic self-diffusion studies, the prevailing paradigm is that atomic jumps are not correlated over large distances. This adds up to our previous arguments that postulating correlations between atomic jumps over long distances is unphysical.

Such an objection cannot be rebutted by stating that it would be a mere speculation. First of all, the objection just formulates the prevailing paradigm. In this respect it is the postulate of correlations between the jumps that appears as a speculation. Secondly, it would consist in a reversal of the charge of proof.

C. Atomic jumps in order-disorder transitions

The objection can also not be rebutted by invoking an order-disorder transition. First of all, invoking an order-disorder transition completely changes the context. It is difficult to see how an order-disorder transition could tell us something about atomic jumps in a self-diffusion process. Secondly, atomic jumps do not become correlated in an order-disorder transition. Once again, it is only the atomic positions, not the atomic jumps, that become correlated in an order-disorder transition. What one can e.g. do to illustrate this point in the situation close to the order-disorder phase transition is considering a periodic lattice of asymmetric double-well potentialss. E.g. the minima of the left wells lie much lower than the minima of the right wells. In each double well, an atom is allowed to jump. When the temperature rises the difference between the two wells becomes smaller, and at high temperature the wells are completely symmetric, while at the transition temperature they are highly asymmetric. At all temperatures the jumps in the different double-wells are stochastic, but when the temperature decreases, the jumping atoms will have a much longer residence time in the lower left well minima than in the higher right well minima. While at high temperatures the residence times will be equal, because the two wells are equally deep. The asymmetry will develop when the temeprature is decreased. In the asymmetrical double-well the jump rates left-right and right-left are different, and their ratio is governed by a Boltzmann factor. Such models have been used to describe the phenomenology of order-disorder transitions. This clearly shows that the jumps remain independent, while it is the most probable positions of the atoms (within the left well minimum in between the jumps) that become correlated.

D. Not every reciprocal lattice vector is a wavelength

In general, there is no guarantee for the attribution of a wavelength to the quantity $q_{\|}$ as the authors do. The authors believe that the QC is not stable and that its zero temperature ground state is a crystal. The low-temperature phase is not observed because the kinetics become frozen, such that the phase is not reached. They want to interpret $q_{\|}$ as a wavelength associated with the phase transition towards the low-temperature periodic, crystalline phase. It may well be that the random tiling scenario predicts a diffuse scattering as observed by Francoual et al. But the "wave lengths" of this scenario are in superspace, and do not have the real meaning of a wavelength in physical space, as we have already shown. We can also give some physical reasons within the context used by the authors.

As there is no conclusive interpretation of the diffuse scattering, considerations about the nature of the *ad hoc* stipulated purely hypothetical transition (order-disorder, first or second order) cannot play a role at this stage. Consider thus the clear analogon of a (second-order) antiferromagnetic phase transition.

At approaching the Néel temperature from above, larger and larger antiferromagnetically ordered domains (or clusters) will occur that will take longer and longer times τ to decay. This will show up as diffuse scattering intensity centered at the antiferromagnetic Bragg position, e.g. at $\mathbf{Q} = [\frac{1}{2}, 0, 0]\frac{2\pi}{a}$ of the future low-temperature phase, where a is the lattice parameter of the high-temperature phase. The intensity at $\mathbf{Q} + \mathbf{q}$ will have a characteristic decay time τ, which is a measure of how long an antiferromagnetically ordered cluster of size $2\pi/q$ will persist in time without being disrupted by the spin flip dynamics. We see that it is $2\pi/Q$ rather than $2\pi/q$ that characterizes the wavelength $2a$ of the spin wave that is being built up. The quantity $2\pi/q$ is not a long wavelength of some spin wave, but an instantaneous domain size, a coherence length of the short wavelength spin wave. The time τ is not characteristic

of the spin flips themselves (which are local), but of the absence of spin flips within a domain of size $2\pi/q$. These domain sizes increase when the spin flip dynamics slow down on approaching T_N.

In this discussion, we use the phase transition only to illustrate a possibility of an interpretation. In the context outlined above, this possibility will remain valid in its general ideas, even if there is no phase transition at stake at all: q refers to a domain size, rather than to a wavelength. By analogy, we see that it does not go without justification in the quasicrystal, to associate $2\pi/q_{\|}$ with some hypothetical long wavelength phason wave. Moreover, if there were some wavelength λ in the phason dynamics, diffuse scattering should eventually build maxima at *new* Bragg peak positions at $\mathbf{Q}$ with $Q = 2\pi/\lambda$, or at satellites positions at $\mathbf{Q} = \mathbf{Q_B} + \mathbf{q}$ rather than remaining smeared out over a continuum of positions $\mathbf{Q_B} + \mathbf{q}$ in a distribution centered on $\mathbf{Q_B}$, with $q = 2\pi/\lambda$ (if it is the signature of the mechanism behind the transition). As the diffuse scattering in QCs remains centered at the Bragg peaks of the high temperature regime and its maxima do not define new Bragg or satellite positions, the *ad hoc* interpretation in terms of critical scattering announcing a phase transition that would not be reached due to the slowing down of the phason dynamics, lacks proof. Note that the slowing down of the spin flips is what triggers the antiferromagnetic phase transition rather than impeding it!

E. Experimental data never contain direct information about correlations between atomic jumps

We have seen above that the whole discussion about correlations between the jumps originates in a confusion that has been made between correlations between *atomic positions* and correlations between *atomic jumps*. We will see below that this confusion has been amplified by overinterpreting the coherence of the signal. The confusion manifests itself already in the apparent paradox that λ is not a period of the displacement field, while $2\pi/\lambda$ occurs in the Fourier spectrum of the disordered quasicrystal. Correlations between atomic jumps can just not be directly observed in an experiment. We would like to stress this point further on the basis of the Van Hove formalism for coherent neutron scattering.

The Van Hove formalism says that what you measure in coherent neutron scattering is the Fourier transform of the probability of having a particle j at position x at time t if the same or another particle k (whereby $k = j$ is thus allowed) was present at position 0 at time 0 we deal with in a measurement are correlations between positions and not correlations between jumps. The very same formalism applies, *mutatis mutandis*, for X ray intensities.

F. Conflicting time scales

We have seen that a Fourier component at a single value of $Q_B + q_{\|}$ corresponds to a density wave for the disordered QC, and that a set of them with the same value of $q_{\|}$ would define a displacement field that is not periodic. The time constant for the relaxation of this field is claimed to be of the order of minutes, on the basis of the speckle experiments. Let us first of all point out that it is difficult to claim that two phason jumps separated by a long distance, could by correlated in time at such a time scale of the order of minutes. In the mean time, the corresponding atoms will have jumped independently a huge number (at least of the order 10^{12}) of times, as the relaxation time for individual jumps measured by neutron scattering and Mössbauer spectroscopy lies in the range from a few picoseconds to a few nanoseconds. What permits us to say that phason jump number n at a given position x will be the one phason flip that correlates through the "sine wave" with a flip, at position x', hundreds of Angstroms away, a few minutes earlier? Of course it can be argued that one has to filter out the fast components. It has been stated [6] that "In this case of the long-wavelength phason fluctuation, individual phason flips, which are local rearrangements of underlying tiles or atomic jumps, correlate at long distance because the same Fourier mode is responsible for them". We would like to point that this contains a kind of reversal of the causality that is by no means warranted by the information content of the data.

To point out why, it is sufficient to refer to the pristine random tiling model Monte Carlo calculations by Tang et al. [10,11] This model will yield the very same type of diffuse scattering around the Bragg peaks, as in a model wherein the kind of correlation evoked above is stipulated. This point was also made by Naumis et al. [18] (see above). And this happens despite the fact that all the jumps are totally uncorrelated by definition: What one does in the model is to flip tiles at random by Monte Carlo. The only experimentally detectable qualitative difference between the model simulated by Tang et al. [10,11] and Widom's model [16] is the T dependence (which is opposite). The model of Tang et al. [10,11] will yield similar time decays of the speckle, because any wavelength that might have been built up in the fluctuation pattern is entirely random and must therefore decay away again with time. Of course, these chance Fourier components are also building up with time following the very same time constants. They come and go. In the model of Tang et al., [10,11] there is no correlation whatsoever between the jumps, but of course the random configurations of the system will yield the Fourier components with their time decay. These are chance Fourier components of random correlations. The authors invert the logical order of cause and effect by attributing

the origin of the correlation to this Fourier component, such that it would be the Fourier component that causes the fluctuations. In reality the fluctuations are random, and the Fourier analysis of these fluctuations yields the chance Fourier component. There is not the slightest element in the experimental data that permits to justify this kind of reversal of cause and effect that is present in the claims of the authors. Such a reversal might be necessary to make the conceptual change from the model of Tang et al. [10,11] to a reading of Widom's model [16] in terms of tile flips (rather than small atomic displacements), but there is nothing in the experimental data that can be claimed to be evidence for it.

Certainly, in the model of Tang et al. [10,11] it can occur that locally, a tile flip only becomes possible after another one, and one could build a chain of such possibilities over a long distance. But many other tile flips disrupt this chain all the time at a very high rate, as explained above. The origin of the coherence of the signal is not the presence of such marginal chains. What is overinterpreted in the data is the fact that the coherence of the signal is not due to correlations between atomic jumps, but to correlations between atomic positions. In fact, the totally uncorrelated jumps in this model will nevertheless give rise to strongly structured coherent signals, but these are due to the correlations between the atomic positions. The paradox is thus, once gain, due to a confusion between correlations of atomic positions and correlations of jumps.

Altough it is not relevant for the data, its is perhaps interesting to end this subsection on a remark about random correlations between jumps. For the atomic jumps themselves one should not confuse the *constraints* of the random tiling model with some correlations or lack of independence in the tile flips. The jumps are totally uncorrelated within the given set of constraints dictated by the random tiling model. To give an analogon: For two completely independent walkers in a city where all streets run only eighter North-South or East-West, like New York (without Broadway), we might find a mysterious correlation in that they are found to walk always only in mutually perpendicular or parallel directions. This is not a mysterious correlation between the two independent walkers, but a constraint imposed by the city map of New York. Hence, yes it is true that the directions of the jumps in the random tiling model are strongly "correlated", but nevertheless the jumps are totally independent by the very construction of the Monte Carlo simulation.

IV. THE TEMPERATURE DEPENDENCE

A. What it is all about

The diffuse scattering data cannot correspond to disorder generated by phason jumps. Up to now we have analyzed the diffuse scattering in the assumption that it would be due to tile flip disorder. We have already pointed out alternative possibilities, e.g. chemical disorder. Here we show that the data simply cannot correspond to quasicrystal disorder that would be created by tile flips.

The quasielastic neutron scattering signal that corresponds to tile flips has been studied in great detail. The intensities of the quasielastic signals in these data show that the number of phason jumps increases when the temperature is raised. This is model-independent factual information. If the diffuse scattering observed by the authors were to correspond to tile flip kinetics it should thus follow the same temperature behaviour as in the neutron data. In reality it follows the opposite one: the diffuse scattering diminishes when the temperature is raised. This contradiction is unassailable. Perhaps, this requires a more detailed discussion.

B. Coherent and incoherent scattering in many-particle problems

1. Method of Calculation

First of all the reader should check in Reference [19] how a coherent quasielastic signal is calculated. Such a coherent quasielastic signal is the temporal Fourier transform of the speckle signal whose intensity decays exponentially with time. We cannot possibly reproduce the whole development of that paper. Perhaps it suffices to say that one first has to map out the whole set of configurations $\mathcal{C}_\mu$ that the system can take. Each configuration $\mathcal{C}_\mu$ can be represented as a point in a higher-dimenional space. When an atomic jump with relaxation time τ takes the system from configuration $\mathcal{C}_1$ to configuration $\mathcal{C}_2$, we connect the corresponding points by a line that we label with τ. This way the whole dynamics can be mapped in the terms of a graph in higher-dimensional space. We can then say that the whole system is an abstract, single particle that diffuses through first-neighbour jumps on the higher-dimensional graph. This high-dimensional diffusion problem can then be expressed in terms of a set of coupled linear differential equations with a jump matrix, just as the normal, less abstract diffusion problem of a single particle in physical space.

We may note that not only coherent scattering, but also incoherent scattering for a many-particle system can be calculated following this method. Contrary to folk lore, this leads to a more complicated and structured neutron signal than coherent scattering. In coherent scattering, it is not relevant where the individual particles are within

a configuration. All configurations obtained by particle exchange operations are in the same equivalence class. All that counts in coherent scattering is the configuration. But, in incoherent scattering one must keep track of a single particle. In incoherent scattering, two identical configurations, where an individual particle finds itself in different positions, are therefore no longer equivalent. The configuration space to be considered is therefore much larger in the case of incoherent scattering. The reader can verify this on the calculation of the model in reference [19].

There is still one step missing if we want to compare this with the speckle data, which are obtained with X-rays. In general the scattering formalisms for various techniques are quite analogous. They can be just transcribed *mutatis mutandis* from one probing particle to another one. But it is only with the advent of synchrotron sources that the study of dynamics has been opened to X-ray studies. In fact, X rays do not have the necessary energy resolution (which lies at the very best in the eV range) to separate even the broadest quasielastic scattering (which occurs in the μeV or meV range) from purely elastic scattering. The diffuse scattering observed is thus an energy-integrated whole of all quasielastic and elastic signals. That means that one has to add up all quasielastic form factors from the calculation described above in order to obtain the diffuse scattering signal. A diffuse signal at a paricular Q-value will in general arise from many contributions with different relaxation times. But in the case of the speckle data, there is a time dependence, such that the separation between the quasielastic scattering and the elastic scattering is obtained in the time domain. It also appears that, at least in a first approximation, at a given Q-value, one measures only one, dominant value of the many relaxation times that are probably present.

C. Applying it to quasicrystals

If one were able to diagonalize the huge jump matrix that describes the whole of the phason dynamics, along the methods of calculation descibed above, one would find a (very large) number of characteristic times, each leading to a Lorentzian signal with a dynamical structure factor. Such a jump matrix exists both for the coherent and for the incoherent scattering case. There is a coherent and an incoherent signal even for the case of completely independent jumps. Such a theoretical treatment is beyond reach however, due to the sheer size and complexity of configuration space. The observed neutron scattering data correspond to the sum of coherent and incoherent scattering (contrary to the statements of reference [1] that the neutron scattering signals would be incoherent). The long-time signals attributed to phason dynamics in reference [1] would just correspond to some of these Lorentzians, with very long relaxation times.

These long relaxation times are not elementary parameters of the jump model, but functions of more elementary jump times. These functions pop up as the inverses of the eigenvalues $\lambda_j = f_j(\tau_1, \cdots \tau_n)$ of the jump matrix that has been defined in terms of the few more elementary jump times, $\tau_1, \cdots \tau_n$, which are much faster. The Q-dependence of the intensities of all Lorentzians Λ_j is given by the corresponding structure factors. Any temperature dependence enters into the model through the temperature dependence of the elementary jump times in terms of activation energies. Through the functional dependence $\lambda_j = f_j(\tau_1, \cdots \tau_n)$ evoked, the temperature dependence of all Lorentzians is thus dictated by the temperature dependence of the elementary jump times. According to crude criteria such as increasing or decreasing of jump times or intensities, the long time dynamics should thus have the same temperature dependence as the short time dynamics, and any person who wants to formulate a claim that they could show opposite behaviour will have to work very hard to gain credibility for it.

But the experimental observation (from quasi-elastic neutron scattering) that the intensity, rather than the width of the fast signals increases with temperature is unusual. In a first approach, one might argue that it could be due to the finite energy resolution of the neutron scattering experiments: At low temperatures the dynamics are too slow to be resolved and appear as elastic. At higher temperatures they become resolved leading to the illusion that the intensity of the elastic peak decreases and the intensity of the quasielastic signals increases accordingly. But this is not what happens: The width of the quasielastic signals cannot be detected to change with temperature, while their intensities change drastically in a way that cannot be attributed to some broadening. One needs to introduce an assistance scanario to explain this very unusual behaviour.

But what one observes in the diffuse scattering and has been called the effect of an inverse Debye-Waller factor, is that the intensities (i.e. structure factors) of the very slow signals *decrease* while the temperature is raised, *with the intensities being transfered to the Bragg peaks!* If there were no assistance scenario in the jump dynamics, the intensity and its Q-dependences for a given Lorentzian Λ_j would remain the same at all temperatures. In fact, the structure factor of the Lorentzian Λ_j associated with λ_j is not changed by a speeding up of the dynamics (allowance made for the phonon Debye-waller factor). At the very best, its intensity at a given Q-value would appear to be associated with a *faster* relaxation time, through the functional relationship $\lambda_j = f_j(\tau_1, \cdots \tau_n)$. If one wants to change f_j or the structure factors, rather than just $\tau_1, \cdots \tau_n$, one has to introduce special assumptions. The observation that the quasielastic neutron scattering intensity increases with temperature forced us to introduce such an assumption in the form of an assistance scenario, as even the elastic intensity is determined by the jump model: It corresponds

to the eigenvalue 0 of the jump matrix, and hence its intensity or structure factor should normally not change with temperature. In other words: Allowing for the effect of the Debye-Waller factor due to the phonons, the ratios of the various structure factors, including the elastic one, should have remained the same. In the assistance scenario, the elastic intensity decreases with temperature, because one introduces long-lived excited states, whose population is governed by a Boltzmann factor with a large activation energy. What kind of most extraordinary *ad hoc* assumptions would have to be introduced into the jump model in order to obtain a *decreasing* diffuse intensity as observed by the authors, that could be attributed to phason jumps despite the fact that the quasielastic intensity corresponding to fast phason jumps has been observed to *increase* by neutron scattering?

To resume the situation: The number of tiles that flip increases when the temperature is raised. Therefore, the intensity of the signal that should betray the presence of these tile flips, e.g. off-Bragg-peak diffuse scattering claimed to correspond to the structural disorder produced by the flips, should also increase when the temperature is raised. Such a temperature dependence runs contrary to what the authors observed.

The authors [1] then decided to proceed by adopting an "alternative random tiling model", proposed by Widom, wherein the diffuse scattering intensity decreases when the temperature is raised. This is certainly important for comparing different versions of the random tiling model, that differ in their predictions about the temperature dependence of the data, but it sidetracks the attention with respect to the important issue we want to address here: Such a change of model does not change a iota to the fact that the diffuse scattering intensity cannot be attributed to structural disorder produced by tile flips as it has the wrong temperature behaviour. It follows that the diffuse scattering, which they call the "phason fluctuations" of the alternative random tiling model, must be dissociated from tiling disorder, i.e. the "phason fluctuations" are not tile flip kinetics.

This point cannot be rebutted by arguing that this would not be contradictory because it is only the pristine random tiling model, as used in the simulations of Tang et al. [10,11] that would ask for an intensity that increases with rising temperature, while Widom's more elaborate model [16] can also incorporate other temperature dependences. That would be a totally pointless misrepresentation of the issues. *We are not opposing the temperature dependence data to the predictions of the random tiling model* (although it is interesting to point out that the temperature dependence discards the pristine model in favour of Widom's model [16]) *we are opposing the temperature dependence to the information given by quasielastic neutron scattering data*!

V. WHAT IS PHASON ELASTICITY ON THE MICROSCOPIC LEVEL?

By formulating their claims in a macroscopic language of phason elasticity, the authors [1] created a language barrier: Nobody understands how the macroscopic phason elasticity is supposed to relate to the microscopic-level tile flips, if it does at all. And therefore, nobody knows how the previously mentioned microscopic objections about the temperature dependence should be translated across this language barrier. In Widom's paper [16] it is stated that on lowering the temperature the QC moves away from the ideal random tiling conditions and that this drives an elastic instability. It is not told how this should be described on the microscopic level. The elastic instability could e.g. correspond to a distortion of the tiles rather than to their mere flips. Widom's paper [16] talks about an inverse Debye-Waller effect on the *elastic* intensity.

A. Debye-Waller factors

What the authors call the inverse Debye-Waller effect is the fact that when the temperature increases the diffuse scattering decreases, and the intensity is transfered to the Bragg peak, such that simultaneously the intensity of the Bragg peak increases.

A true Debye-Waller diminishes all the elastic intensity at the profit of the inelastic intensity. There is a sum rule between the elastic and the inelastic intensity. The Bragg peaks and the diffuse scattering intensities are *both elastic* intensities, such that they should be both affected the same way. What is the solution of this apparent contradiction?

What Widom calls the Debye-Waller factor in his paper [16] is not the true Debye-Waller factor, but a theoretical auxiliary quantity that consists on integrating on only the very long-wavelength modes (i.e. on only an infinitesimal domain of q-vectors). It thus excludes the whole phonon density of states (as e.g. measured by Suck), except that infinitesimal part. Similarly, it excludes all the phason dynamics measured by TOF neutron-scattering, except an infinitesimal part. Within this long-wavelenth approximation Widom then calculates his auxiliary quantity that indeed applies to the transfer of intensity between the Bragg peak which is elastic and the diffuse scattering which becomes inelastic in this approach. This leads to the interesting possibility that the true Debye-Waller factor, obtained by including the rest of the full q-range, could again invert the tendencies described by Widom, [16] i.e. some Bragg peaks could show the inverse effect, while others could show the normal effect.

B. Diffusion constants

We may note that a linear relationship $\tau \propto 1/q^2$ certainly does not prove that the speckle data are produced by a diffusion mechanism. The relevant parameter for the diffuse scattering is $1/q^2$, such that the first terms of any Taylor expansion for the underlying physics will lead to $\tau = C_1 + C_2(1/q^2)$. The authors have previously considered the relationship $\tau = C_1 + C_2(1/q^2)$ and this resulted in a much better fit of their data. Such a relationship is totally unspecific. In general, fits of the type $\Gamma = 1/\tau = \Gamma_0 + Dq^2$ (where $\Gamma_0 \neq 0$ is an indication for confined motion) are used to analyze data when we already know that they are produced by a diffusion mechanism, not to present the data as supplementary evidence that a diffusion mechanism would be at work.

But let us admit that the analysis of the authors in terms of a diffusion constant is correct. We want to point out that the use that the authors make of Lubensky's statement [3] that "phason modes are diffusive" is misleading, as it creates the impression that the data analysis would contain proof for their interpretations, while this is not true.

E.g. Huang scattering in standard crystals is traditionally described as a "frozen phonon". (This has nothing to do with phonon dynamics: The kinetics of frozen phonons will lead to (very narrow) quasielastic scattering, while dynamical phonons correspond in general to non-zero frequencies). *The kinetics of frozen phonons shall be "diffusive" in many instances, although phonons are not qualified as diffusive by Lubensky.* [3] That the kinetics of the diffuse scattering is of the relaxational or even of the "diffusive" type, in the interpretation Francoual et al. want to give to it is thus unspecific.

There is a conceptual contradiction between wanting to describe a phenomenon as diffusive and taking simultaneously a "sine wave" *Ansatz* as the authors do. The "sine wave" makes a decay take simultaneously over the whole of space, while a diffusive phenomenon should progressively spread out in space. Diffusive "sine waves" are a *contradictio in terminis.*

We may finally note that time scales of the order of minutes put the interpretation of the data beyond any possible cross-checking with other spectroscopic techniques. This is a dangerous situation that demands for extreme caution. It is preferable to have a situation where we can follow the physics from time window to time window, with different techniques that have overlapping time scales if we want to link two observations in vastly different time domains to a same phenomenon as the authors do, by identifying diffuse scattering on the minute time scale with phason dynamics on the pico-second time scale.

C. Phason elasticity

It might be misleading that theoretical physicists also use the term elasticity for situations where there are no interactions between atoms. E.g. in the simulations of Tang et al., [10,11] the tile flips are totally independent, but still a "phason elasticity" can be defined to refer to an "entropic restoring force". If this is not appreciated properly, it can lead to more confusion in terms of "elastic wave" pictures that do not apply.

VI. LACK OF PROOF FOR THE RANDOM TILING MODEL

A. Introduction

In reference [21] the authors stated that the temperature dependence of the data are "in contradiction with the hypothesis of a simple random tiling model". They added that the tile flip interpretation can be maintained by introducing a more complex random tiling model. [16] The random tiling model and the tile flip scenario can thus be claimed on the basis completely opposite temperature behaviours. This is thus not very convincing evidence for the random tiling model. To cite Sir Karl Popper: *"Falsifiability should be the criterion of demarcation in science".* [22] Moreover, as already pointed out, all the temperature data might show is that there is some softening of the elastic constants.

What the temperature dependence settles *within* the context of the random tiling model, is that the pristine model should be abandoned in favour of Widom's model. [16]

The diffuse scattering data, without a mention of their temperature dependence, were eventually reported in reference [23]. Nonetheless, in the final discussion of that paper, the authors already introduce the arguments to thwart the criticism that can be formulated based on the temperature behaviour observed. As the reader had no access to the information about the temperature dependence, he could not possibly understand the issues at stake. Rather than the mention of a crucial problem raising serious doubts [24] about the validity of the interpretation, it looked like a very puzzling digression.

In that paper, the authors concluded that their data were "compatible" with the random tiling model. The problem with this statement does not reside in what it literally states, but in its tacit *tertium non datur.* It does not say that

the data may also be compatible with other models. Due to this formulation the reader may pick up the totally unjustified belief that the data would have conclusively proved the random tiling model. As a matter of fact, such overinterpretations have indeed made their way to the literature. [25]. This is of course very harmful. It has installed a strong oral tradition, that made it very difficult to point out the absence of convincing proof for the random tiling model. The unassailable character of the literal interpretation of the formulation may have contributed to this situation. It is not by qualifying this problem as an old story, that the validity of these claims can be settled.

B. The data claimed to be evidence are too unspecific

At this point it is perhaps good to review the evidence the authors proposed in favor of their interpretation in terms of tile flips.

(a) An $1/q_{\parallel}^2$ dependence of the intensity. This is unspecific and occurs in many other cases of Huang scattering in crystals. [26]

(b) A certain shape of the diffuse scattering intensity contours. Again, this is unspecific, and purely due to symmetry in analogy with the situation in conventional crystals. [26] Such symmetry-based arguments do not contain any information about the underlying mechanisms or interactions.

(c) The shapes of the contours depend only on the phason-phason elastic constants. This is the only feature that is not unspecific. As such, it may rule out a number of alternative models. But this does not mean that it rules out all possible alternative models. We have clearly shown that it is still not unique, even if it has been presented as self-evident that it would be unique.

As the temperature dependence of the neutron scattering data does not agree with an interpretation in terms of tile flips, we actually know that the correct model must be different.

We should insist on the fact that an interpretation in terms of tile flips is a *derived* application of the elasticity theory, which is formulated in terms of a continuum of small atomic displacements rather than on a discrete set devoid of infinitesimals. The very definition of an elastic constant cannot be written down if we cannot assume that the atomic displacements explore a continuum. The validity of the derived application is not obvious, as on the microscopic level tile flips do not explore a continuum of atomic displacements. This is very different from the continuum limit for phonons. There is thus *a priori* no good theoretical rationale to explain the results of Tang et al. [10,11] except the *post facto* observation that it works despite such theoretical objections.

To improve on this situation, Henley [27] has proposed an argument of coarse graining. Unfortunately, this may rather have contributed to the fact that it was not realized that the interpretation of the data is not unique. Without this argument of coarse graining, the results of Tang et al. [10,11] could have stood out as a clearly different alternative in the form of a discrete displacement field rather than a continuous one.

In any case, it remains a cracking pass to generalize the finding by Tang et al. [10,11] in the sense that one takes it for granted that the only exclusive way to obtain such a dependence on the phason elastic constants would be the tile flips that are so improper for the first-hand application of the theory. Similarly, in Widom's [16] Landau-type theory it is not granted that the phason elastic constants can only correspond to tile flips: Even if some elastic instability were observed in diffraction experiments that completely tallied with his calculations, it would not yet prove the random tiling scenario.

We may note that the phason elasticity constants obtained by the authors of their diffuse scattering data do not agree with calculations based on canonical cell tilings. [28]

Finally, we must point out that the alternative that the diffuse scattering could be due to chemical disorder has not been properly ruled out. If data really were due to chemical disorder, than they would not contain any proof for the random tiling model.

In conclusion, we think that the experimental evidence is too narrow to claim that it would be a proof for the validity of the random tiling model. It could be misleading to want to harness the observation of diffuse scattering into a stability issue in the form of an opposition between random tiling models and a perfect quasicristal model.

C. Absence of proof for a phase transition

We may note that the QC-crystal transition postulated by the random tiling model is of first order, while in the example of $NaNO_2$ cited in reference [1], as a paradigm for the observations, the transition is second order.

We may also note that there is no proof for the existence of the phase transition postulated by the random tiling model. In fact, one must introduce *ad hoc* assumptions in order to deny the clear experimental evidence that there is no phase transition. One might well invoke the superstructure reported by Ishimasa, [29] which shows both satellite peaks and diffuse scattering. However, for this exceptional observation there are many other ones where there is no phase transition at all. [30] But even in the favorable case reported by Ishimasa, the dominant diffuse scattering maxima are centered on the Bragg positions of the QC, not on the satellites. The scenario is thus not one of diffuse scattering progressively building up at the future satellite positions, and eventually turning into satellite Bragg peaks.

Let us finally point out that in a discussion about the presence of a phase transition or otherwise, only those who claim such a transition exists can solve the issue by providing the proof. It is in fact virtually impossible to prove that there is no phase transition: absence of proof can simply not be taken as proof of absence.

VII. "PHASON JUMPS" IS BY ALL CRITERIA AN APPROPRIATE TERMINOLOGY

It has been claimed that the terminology "phason jumps" would not be appropriate, and that the terminology "phason" should be reserved to the modes postulated in Eq. [42] in the paper by Janssen et al. [4] There are several problems with this restriction or change of definitions:

(1) Before defining "phason modes" as is done in reference [4], one could wish to wait until convincing proof of their existence has been given. The "phason modes" are just a conjecture. We have seen that they are introduced on the basis of an argument that is not rigorous. It has to be checked that the definition is meaningful. It rather seems as though this is not the case.

(2) To motivate the claim mentioned, it has been argued that phason jumps are not characteristic of quasicrystals because they also occur in 1/1 approximants. [31] This capitalizes on undue remarks in references [32] where it was stated that phason jumps are "not a special property for QCs". These remarks focused on the fact that extremely fast jumps also occur in a B2-based phase that can contain up to 12 % vacancies. But phason jumps are not only "special" in just being fast, and it is doubtful that the presence of atomic jumps in a very unusual sample with an exceptional amount of vacancies could shed light on such issues. What is obvious is, that if atomic jumps occur in a system, they will forcedly produce diffuse scattering. Diffuse scattering or speckle are also not characteristic of quasicrystals. And the modes that are supposed to replace the phason jumps as the unique observation of phason dynamics most probably just do not exist.

(3) The terminology "phason" was introduced by Overhauser [33] in the field of dynamics of charge-density waves. It did not rely on elasticity considerations. It is thus the terminology "phason elasticity" that is a derived concept, rather than the phason jumps.

(4) The argument that "phason jumps" is not a good terminology has been pushed to the extreme. One has even proposed a sarcastic, belittling terminology "flipon" for them, [6,34] but e.g. Edegawa et al. have clearly pointed out that the terminology is perfectly appropriate. [6]

VIII. CONCLUSION

In conclusion, we have shown that many claims of the authors concerning the random tiling model and phasons in QCs are not justified. The author would like to thank Michel Duneau for many helpful discussions.

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