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Positional disorder in lattices: a model for the structural order responsible for the first sharp diffraction peak in glasses

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Abstract

Gaussian positional disorder has been introduced into the structure of various 2D and 3D crystal lattices. As the degree of positional disorder is increased, the weights of the highest- k peaks in the orientationally-averaged structure factor are decreased first, eventually leaving only a single peak having significant weight, that with the lowest k -value. The orientationally-averaged real-space pair-correlation function for lattices with such high levels of positional disorder exhibits a corresponding power-law-damped series of oscillations, with a single period equal to the separation between the furthest-separated, lowest- k lattice planes. This last surviving peak in the structure factor is at a k -value nearly identical to that of the experimentally-observed first sharp diffraction peak for the corresponding real amorphous phases of such lattices (e.g. Si, SiO₂). These results also have a bearing on the unsolved Gauss circle problem in mathematics.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Understanding the nature of the atomic structure of amorphous materials, or glasses, remains a perennial problem. Although the atomic structures of computer-simulated (e.g. molecular-dynamics) models of amorphous materials are of course completely determined, nevertheless uncertainties remain concerning the approximations used to generate empirical or *ab initio* interatomic potentials used in the simulations, and the uniqueness of the fits of calculated structural quantities (e.g. pair-correlation functions) with experimental structural data which, perforce, are only known in a statistical, averaged form. Due to the statistical nature of the experimental structural information for amorphous materials, available, for example, in the form of pair-correlation functions consisting of broad, strongly-overlapping peaks, only short-range order (SRO), e.g. nearest-neighbour bond lengths or coordination numbers, is known unambiguously [1]. Medium-range order (MRO), relating, for example, to the connection

of coordination polyhedra (e.g. SiO₄ tetrahedra in vitreous silica, v-SiO₂), is much more difficult to ascertain from experimental diffraction data [2]. One celebrated feature that has long been associated with MRO is the so-called first sharp diffraction peak (FSDP), or pre-peak, in the experimental structure factor, $S(k)$, where $k = |\mathbf{k}|$ is the magnitude of the wavevector transfer. An FSDP occurring at the value $k = k_1$, if interpreted as being an isolated pseudo-Bragg peak arising from remnant quasi-periodic atomic correlations, corresponds to a real-space, quasi-periodic oscillatory behaviour of the orientationally-averaged atom–atom pair-distribution function (PDF), $g(r)$, with a period D related to the FSDP position by [2]:

$$D = 2\pi/k_1. \quad (1)$$

The full width, half maximum of the FSDP, Δk_1 , is determined by the correlation length, l_1 , over which the quasi-periodic real-space atomic correlations persist:

$$l_1 = 2\pi/\Delta k_1. \quad (2)$$

The disorder in the structure of a non-crystalline material can be categorized in several ways: positional and topological, or even chemical, disorder may be present. Positional disorder refers to the atomic positions being displaced from the lattice-related sites of a crystalline structure. Topological disorder relates to a more profound departure from crystalline order; an example is the presence of non-crystalline ring sizes in amorphous network materials. The atomic positions in this case are not simply related to crystallographic sites, as they are for positional disorder. Chemical disorder refers to the breakdown of local chemical stoichiometry, e.g. the presence of homopolar covalent bonds in what would otherwise be a chemically-ordered structure having only heteropolar bonds, for a particular stoichiometric composition. Unfortunately, in real amorphous materials, the influence of each of these individual manifestations of disorder on particular physical properties, for example atomic-vibrational behaviour, often cannot easily be disentangled. However, it would be very interesting to explore to what relative extent positional or topological disorder, for example, controls the physical behaviour of amorphous materials. With this in mind, we have initiated a study of the effect of lattice-positional disorder on atomic-vibrational dynamics [3]. As a result of these investigations, we have had recourse to study in detail the atomic structure of positionally-disordered 2D and 3D lattices [4]. This paper further develops the work reported in [4], particularly with regard to the connection between this work and the Gauss circle problem. We also describe the evolution of $g(r)$ with increasing disorder in detail, and show results for the structure factor $S(k)$ calculated by Fourier-transforming $g(r)$.

2. Method

Positional disorder has been introduced into various 2D and 3D lattices or crystal structures by shifting the positions of the atoms away from their crystalline positions by an amount randomly chosen from a given probability distribution, centred on zero. For the most part, a Gaussian distribution has been used, the half-width, σ , of this distribution then being a measure of the amount of positional disorder introduced into the lattice. Other probability distributions have also been examined—e.g. based on the Lorentzian function—but they gave very similar numerical results to those of the Gaussian distribution, and so will not be discussed here. Random displacements, governed by the chosen probability distribution, were applied independently to x -, y - and z -components of the atomic positions. For simplicity, normally no restriction was placed on the closeness of approach of positionally-disordered atoms, although it was checked that the imposition of a minimum interatomic separation did not make any qualitative difference to the results presented here. Several types of lattice and

crystalline structure were investigated, both two-dimensional (2D) and three-dimensional (3D) in character. The 2D square lattice was positionally-disordered, as were the 3D simple-cubic, face-centred-cubic (FCC), body-centred-cubic (BCC) and diamond-cubic lattices. In addition, the low-temperature, tetragonal α -phase cristobalite structure, a crystalline counterpart to vitreous silica (v-SiO₂), was also positionally disordered. Very large models, consisting of $\sim 10^5$ sites and having periodic boundary conditions (PBCs), were used. The use of such very large models is essential for two reasons. The use of PBCs limits the allowed \mathbf{k} -vectors (necessary in the calculation of the structure factor) to the form $\mathbf{k} = \frac{2\pi}{L}\{l, m, n\}$, where L is the cubic box size, and l, m and n are integers. Evidently, it is necessary to use very large models (large L) in order to increase the number of allowed \mathbf{k} -vectors, particularly at low k -values ($k = |\mathbf{k}|$). Periodic boundary conditions also imply that the pair-distribution function, $g(r)$, is unreliable for distances beyond half of the box size; we do not show, therefore, $g(r)$ data for values of $r > L/2$.

3. Theory

The most common experimental technique for determining the structure of either crystalline or amorphous solids is by diffraction: the diffraction pattern of a single crystal consists of a (generally anisotropic) 3D array of points (Bragg peaks), the wavevector transfer, \mathbf{k} , for each Bragg peak in this case being equal, by the Laue condition, to the difference between two lattice points in the reciprocal lattice of the real-space crystal lattice [5]. The modulus of the wavevector transfer is related to the scattering angle, 2θ , and wavelength of the diffracting radiation, λ , by [5]:

$$k = 4\pi(\sin \theta)/\lambda. \quad (3)$$

The measured diffraction intensity can be reduced to the structure factor, $S(\mathbf{k})$, which is simply a sum over phase factors for waves diffracted from pairs of atoms i and j [5]:

$$S(\mathbf{k}) = 1 + \frac{1}{N} \sum_i \sum_{j \neq i} \exp(i\mathbf{k} \cdot \mathbf{r}_{ij}), \quad (4)$$

where N is the number of atoms in the sample, and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ is the vector between the positions of atoms i and j . In the case of polycrystalline, or amorphous, samples, which have spherically-averaged symmetry, the 3D anisotropic information inherent in $S(\mathbf{k})$ for a single crystal is reduced to a 1D representation characterized by the function $S(k)$ (the powder pattern, in the case of polycrystalline materials).

If the atomic positions in the structure of a material are known, e.g. in a structural model, the structural information can be characterized by the atom–atom pair-distribution function, $g(\mathbf{r})$, given by [5]:

$$g(\mathbf{r}) = \frac{1}{N\rho_0} \sum_i \sum_j \delta(\mathbf{r} - \mathbf{r}_{ij}), \quad (5)$$

where ρ_0 is the average atomic density of the sample. The orientationally-averaged 1D functions $g(r)$ and $S(k)$ are related via a Fourier transform [1]:

$$S(k) = 1 + \frac{4\pi}{k} \rho_0 \int_0^\infty r [g(r) - 1] \sin kr \, dr. \quad (6)$$

Gaussian positional disorder in lattices, as studied here, has a real physical analogue: it corresponds to thermally-induced motion of atoms about their crystalline-lattice positions [6, 7]. In this work, the displacements of atoms from their equilibrium crystalline

(lattice) sites that are considered can far exceed those that occur thermally in a real crystal, but the mathematical formalism describing the two situations is nevertheless identical. The effect of thermal motion on the structure factor (equation (4)) is well-known: the weights of the single-crystalline Bragg peaks in $S(\mathbf{k})$ are reduced by (thermal) positional disorder by the exponential Debye–Waller-like factor, $e^{-k^2\sigma^2}$, without a corresponding broadening of the peaks, and the scattering intensity so lost is transferred into a low-amplitude background diffuse-scattering intensity. In terms of the (orientationally-averaged) crystalline structure factor, $S(k)^{\text{cryst}}$, the structure factor for the case when Gaussian positional disorder is present is given by [7, 8]

$$S(k) = 1 + e^{-k^2\sigma^2} [S(k)^{\text{cryst}} - 1]. \quad (7)$$

Given the Fourier-transform relationship between $S(k)$ and $g(r)$ (equation (6)), the effect of Gaussian positional disorder is to convolve the δ -function peaks in the crystalline $g(r)$ (equation (5)) with the Fourier transform of the Debye–Waller factor, i.e. giving:

$$g(r) = \frac{1}{(2\sqrt{\pi})^d \sigma^d N \rho_0} \sum_i \sum_{j \neq i} \exp\left\{-\frac{(r - r_{ij})^2}{4\sigma^2}\right\}, \quad (8)$$

where d is the dimensionality of the system. In principle, $g(r)$ can be calculated by evaluating the summations in equation (8) over all crystalline interatomic distances for a given lattice, and then orientationally averaging the result.

For example, for the case of the Gaussian-positionally-disordered 2D square lattice (with lattice positions $r_i = n_x \hat{x} + n_y \hat{y}$ where \hat{x} and \hat{y} are unit vectors), the orientationally-averaged expression for the PDF is

$$g(r) = \frac{1}{4\pi\sigma^2\rho_0} \sum_{n_x, n_y = -\infty}^{\infty} \exp[-(r^2 + n_x^2 + n_y^2)/4\sigma^2] I_0\left(\frac{r\sqrt{n_x^2 + n_y^2}}{2\sigma^2}\right), \quad (9)$$

where $I_0(z)$ is a modified Bessel function of the first kind. For the 3D equivalent, the simple-cubic lattice, the corresponding expression is

$$g(r) = \frac{1}{8\pi^{3/2}\sigma\rho_0 r} \sum_{n_x, n_y, n_z = -\infty}^{\infty} \frac{[e^{-(r-n)^2/4\sigma^2} - e^{-(r+n)^2/4\sigma^2}]}{n}, \quad (10)$$

where $n = \sqrt{n_x^2 + n_y^2 + n_z^2}$, and the sums in equations (9) and (10) both exclude the case where all the indices n_i ($i = x, y, z$) are zero simultaneously.

It has not been possible to evaluate the sums in equations (9) and (10) analytically. In retrospect, this failure to evaluate exact expressions for $g(r)$ is unsurprising. More than 150 years ago, the German mathematician C F Gauss posed what is now known as the ‘Gauss circle problem’ [9]: given a regular square lattice of points with spacing r_1 , and a circle of radius r centred on one of the lattice points, how many lattice points lie within the circle? This celebrated mathematical problem is still unsolved (as is the corresponding problem for a lattice and hypersphere in $d > 2$ dimensions). The answer for the number of contained lattice points, for the 2D problem, can be written as [10]:

$$N(r) = \pi(r/r_1)^2 + E(r), \quad (11)$$

where the error term, $E(r)$, is unknown, despite much attention [11–13]. However, Huxley [11] has shown that the error term satisfies the inequality:

$$|E(r)| \leq Cr^\theta, \quad (12)$$

where C is a constant and the exponent θ lies in the range $\frac{1}{2} < \theta \leq \frac{46}{73}$, the lower limit having been obtained independently by Hardy [12] and Landau [13]. It should be noted that, in our formalism, an exact expression for $g(r)$ (equation (9)), with a width $\sigma = 0$, would solve the Gauss circle problem.

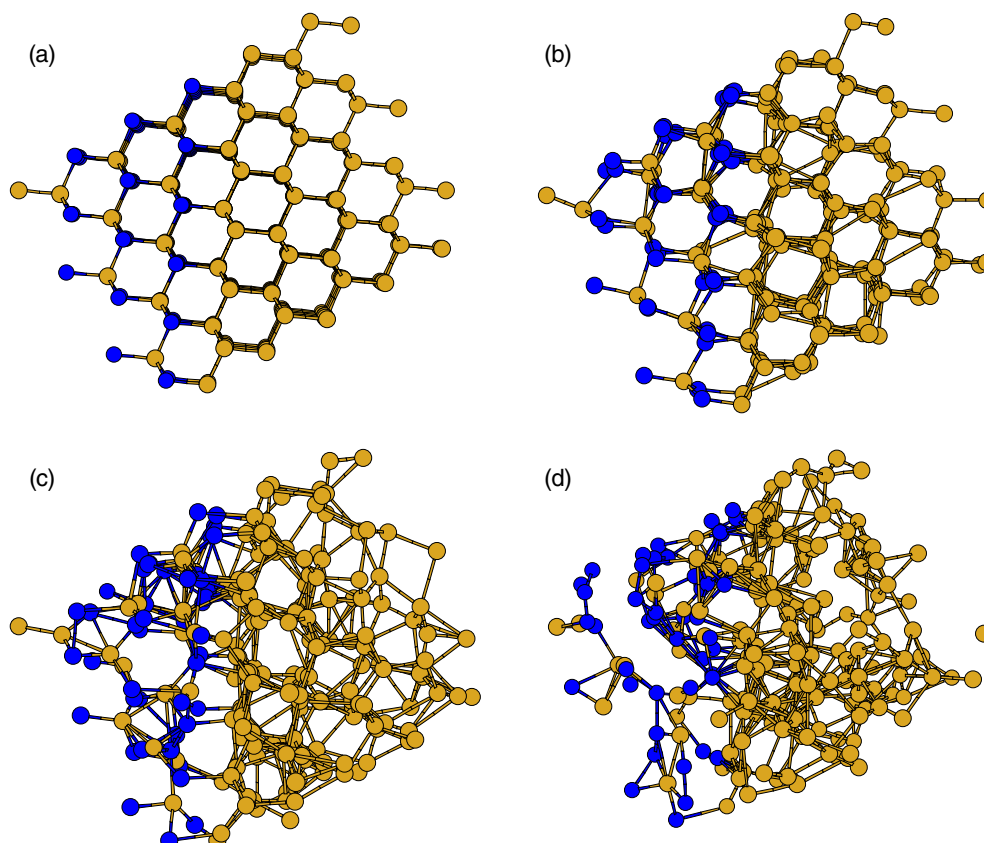


Figure 1. Visualization of Gaussian positional disorder in the diamond-cubic lattice for different values of disorder parameter σ : (a) 0 (perfect crystalline lattice); (b) $0.1r_1$; (c) $0.2r_1$; (d) $0.3r_1$, where r_1 is the nearest-neighbour distance. All models have the same orientation. Atoms in three adjacent (111) lattice planes perpendicular to the page are darkened with respect to the others to show the gradual destruction of the lattice planes with increasing disorder.

4. Results

A visualization of the results of introducing (Gaussian) positional disorder into, as a representative example, the diamond-cubic lattice is shown in figure 1. In this, atoms in three (111) lattice planes have been darkened compared to the rest, as a guide for the eye, to show the progressive destruction of the crystalline structure (figure 1(a)) with increasing disorder (i.e. increasing σ). It can be seen that for small disorder, say $\sigma = 0.1r_1$, in this case, the positionally-disordered structure is still rather like the original crystal (figure 1(b)); if the disorder is large enough, say $\sigma = 0.3r_1$, the atomic positions are located more or less randomly, and only faint vestiges of the original crystal structure persist (figure 1(d)).

The effect of positional disorder on the orientationally-averaged structure factor, $S(k)$, is large and striking: the Debye–Waller factor decreases the weight of the high- k peaks in $S(k)$ the fastest as the degree of positional disorder (σ) increases (figure 2). It is clear from figure 2 that, as the disorder increases, there will be a range of values of σ for which the weights of all peaks in $S(k)$ but one are exponentially small (figure 3); the last remaining peak will therefore be the crystalline peak occurring at the lowest k -value. The peak in $S(k)$ at the lowest value of k corresponds to the set of Bragg planes in the crystalline lattice that are furthest separated in

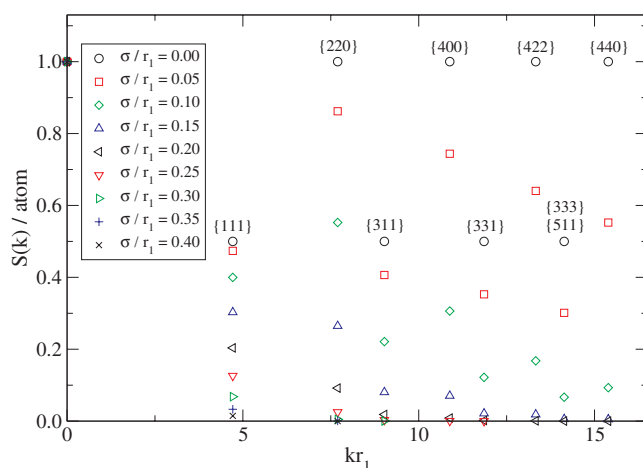


Figure 2. The weights of Bragg peaks in the orientationally-averaged structure factor, $S(k)$ (normalized per atom), for Gaussian-positionally-disordered diamond lattices for different values of disorder parameter, σ . Here k is normalized by the inverse of the nearest-neighbour crystalline distance, r_1 . Only the lowest- k peak (at $kr_1 = 4.71$) survives with significant weight for $\sigma \geq 0.3r_1$. The low-amplitude diffuse scattering is not shown.

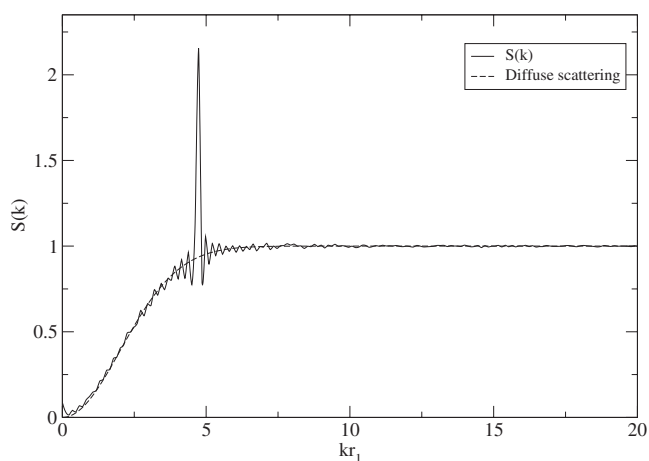


Figure 3. The orientationally-averaged structure factor, $S(k)$, for a Gaussian-positionally-disordered diamond lattice with disorder parameter $\sigma = 0.35r_1$, where r_1 is the nearest-neighbour distance. Here, $S(k)$ has been calculated by taking the Fourier transform of the pair-distribution function $g(r)$, as described in the text. The $\{111\}$ Bragg peak standing on the diffuse background is the only one surviving this degree of positional disorder. The theoretical diffuse background of the structure factor, given by $S(k)^{\text{diff}} = 1 - e^{-k^2 \sigma^2}$, is shown as the dashed curve. The finite width of the Bragg peak, and the sinc-like oscillations on either side of it, are the result of taking the Fourier transform of $g(r)$ for a finite range of r (i.e. half the simulation-box size).

real space; these are also the set of planes with the highest areal atomic density. For the case of the diamond-cubic lattice, for example, for $\sigma \geq 0.3r_1$, only the $\{111\}$ peak at $kr_1 = 4.71$ still has significant weight (figures 2 and 3).

The orientationally-averaged pair-distribution functions, $g(r)$, of the positionally-disordered lattices were computed from the atomic positions in the models. The results, for

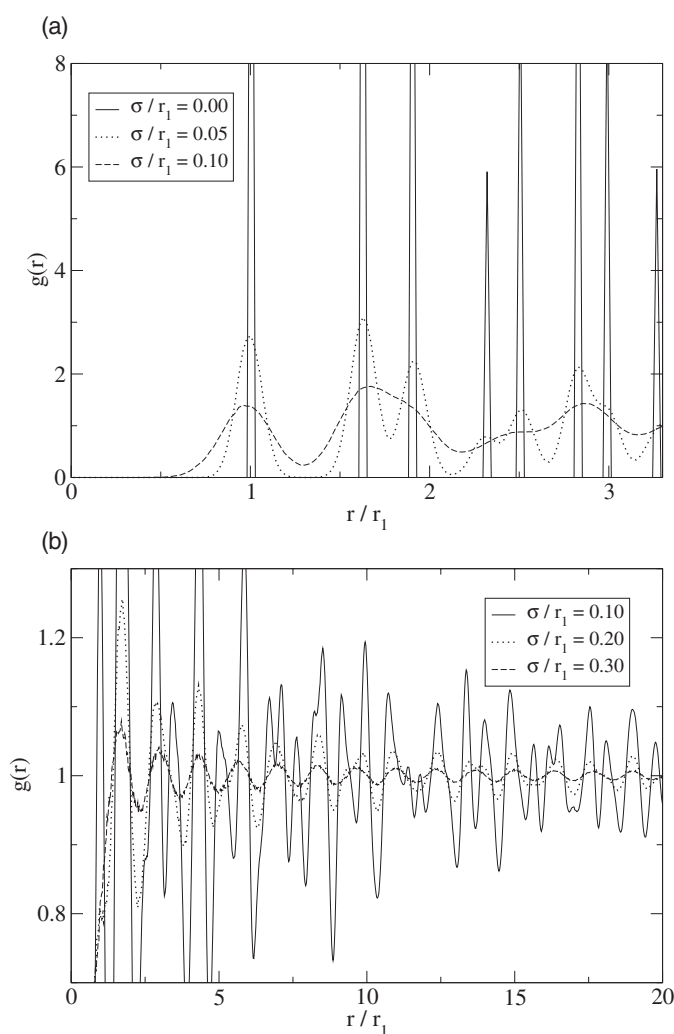


Figure 4. Evolution of the real-space pair-distribution function $g(r)$, with increasing positional disorder in the diamond-cubic lattice for values of disorder parameter σ , in the range (a) $0 < \sigma/r_1 < 0.1$; (b) $0.1 < \sigma/r_1 < 0.3$.

the representative case of the diamond-cubic lattice, are shown in figure 4. For small amounts of disorder ($\sigma \leq 0.1r_1$), the result of positional disorder on $g(r)$ is simply to broaden the delta-function peaks characteristic of the perfect crystal ($\sigma = 0$)—see figure 4(a). However, for larger disorder, $\sigma \geq 0.1r_1$, the broadened peaks start to merge together to give a complicated pattern, and this process continues with increasing σ until, for a sufficiently large amount of disorder ($\sigma \sim 0.3r_1$ in the case of the diamond lattice), remarkably just a *single*-period, damped oscillatory behaviour of $g(r)$ emerges (figure 4(b)). At yet larger levels of positional disorder, there is no further qualitative change in the form of $g(r)$; the amplitude of the single-period extended-range oscillations just decreases with increasing σ . Such damped single-period oscillations in $g(r)$ at sufficiently high levels of disorder have been observed in all the positionally-disordered lattices (2D and 3D) that we have examined. An example of such oscillatory behaviour for the 3D diamond-cubic lattice is shown in figure 5(a), together with

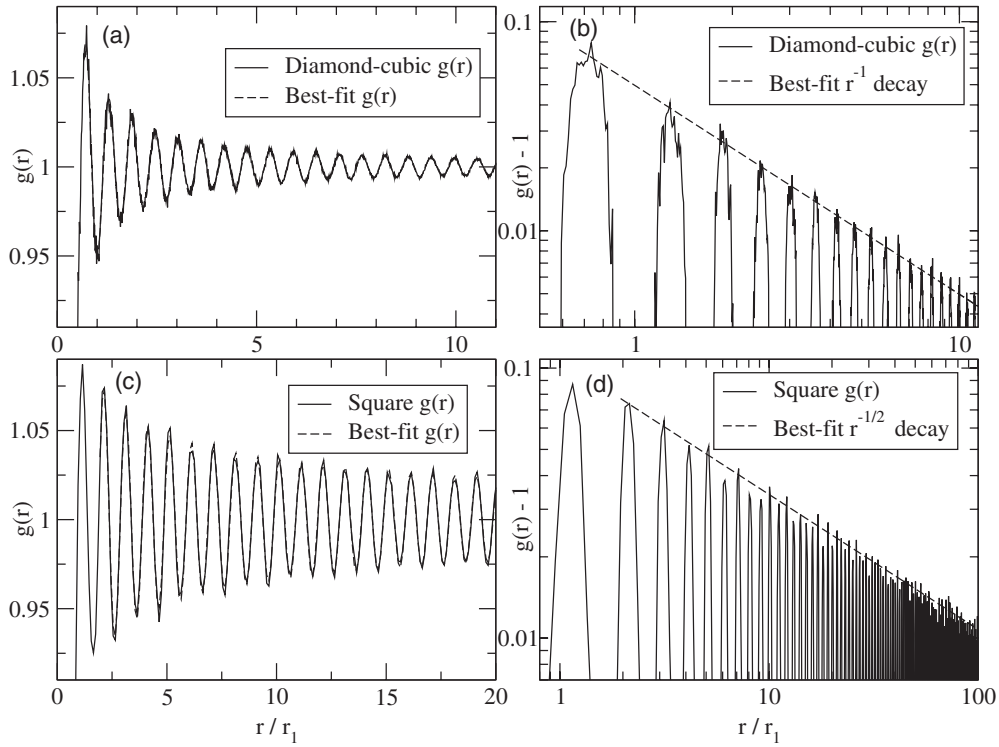


Figure 5. Single-period oscillatory behaviour of $g(r)$, and its spatial-decay characteristics, for Gaussian-positionally-disordered lattices: (a) 3D diamond-cubic lattice with $\sigma = 0.3r_1$ (solid curve) and the best-fit curve (dashed curve); (b) the same data plotted double-logarithmically to reveal the r^{-1} functional dependence (dashed line) of the spatial decay of the oscillations; (c) 2D square lattice with $\sigma = 0.25r_1$ (solid curve) and the best-fit curve (dashed curve); (d) the same data plotted double-logarithmically to reveal the $r^{-1/2}$ functional dependence (dashed line) of the spatial decay of the oscillations.

the results for the 2D square lattice (figure 5(c)), for comparison. From a double-logarithmic plot in both cases (figures 5(b), (d)), it can be seen that the functional dependence of the spatial decay of the single-period oscillations in $g(r)$ is a simple power law in both cases: it is found that the oscillations in $g(r)$ for all lattices in 3D decay as r^{-1} , while in 2D the decay is as $r^{-1/2}$.

The periods, D , of the single-frequency oscillations in $g(r)$ for the various positionally-disordered lattices were found by fitting the computed curves of $g(r)$ by the following empirical functions: in 3D $g(r) = 1 + A'r^{-1} \sin(k_1r + \phi')$, and $g(r) = 1 + A''r^{-1/2} \sin(k_1r + \phi'')$ in 2D where A' , A'' , ϕ' and ϕ'' are constants. A very good fit to these sinusoidal functional forms was found for all peaks in $g(r)$ except for the first couple of peaks at small r ; data for $r < 2r_1$ were therefore excluded from the fits. Values for the average periods resulting from this fitting procedure applied to 20 realizations in each case are given in table 1 for a number of 3D lattices (simple cubic, FCC, BCC, diamond) and also the 2D square lattice. In all cases, as can be seen from table 1, the periods of the single-period oscillations seen in $g(r)$ for sufficiently large values of disorder parameter, σ , are identical to the interplanar distance for the furthest-separated lattice planes in the crystal structure (e.g. the $\{111\}$ planes in the diamond-cubic structure). Evidently, this behaviour in $g(r)$ is consistent with what was found for $S(k)$ (figures 2 and 3) for positionally-disordered lattices with sufficiently large values

Table 1. Families of lattice planes in 3D and 2D lattices having the largest interplanar separation; r_1 is the nearest-neighbour distance in each case. For the values of disorder parameter, σ , given, $g(r)$ consists of single-period oscillations, the periods, D , of which were found from a best-fit procedure, as described in the text. (SC = simple cubic; FCC = face-centred cubic; BCC = body-centred cubic.)

Lattice	Furthest planes	Inter-planar distance	Period of oscillations D
SC	{100}	r_1	$(1.0000 \pm 0.0003)r_1$ for $\sigma = 0.25r_1$
FCC	{111}	$\sqrt{\frac{2}{3}}r_1 = 0.8165r_1$	$(0.8167 \pm 0.0001)r_1$ for $\sigma = 0.25r_1$
BCC	{110}	$\sqrt{\frac{2}{3}}r_1 = 0.8165r_1$	$(0.8165 \pm 0.0001)r_1$ for $\sigma = 0.25r_1$
Diamond	{111}	$\frac{4}{3}r_1 = 1.3333r_1$	$(1.3333 \pm 0.0001)r_1$ for $\sigma = 0.35r_1$
Square	{10}	r_1	$(0.99993 \pm 0.00001)r_1$ for $\sigma = 0.3r_1$

of disorder parameter, σ : the single-period oscillations in $g(r)$ with period D correspond to the single remaining peak in $S(k)$ with appreciable weight, at a wavevector-transfer value k_1 , where $D = 2\pi/k_1$.

The reasons for the observation of damped, single-period oscillatory behaviour of $g(r)$ for sufficiently large disorder, σ , can be inferred from an examination of equations (9) and (10), even though it has not proved possible to obtain analytic expressions for the summations therein. As far as the spatial decay is concerned, the asymptotic behaviour of the modified Bessel function in the expression (equation (9)) for the 2D $g(r)$ has an $r^{-1/2}$ dependence, in accord with the behaviour found from numerical simulations (figure 5(d)). The r^{-1} spatial decay of oscillations in the 3D $g(r)$ (figure 5(b)) is also predicted theoretically by equation (10). A heuristic argument can be given for why single-period oscillations in $g(r)$, with a period equal to the distance between the furthest-separated lattice planes, occurs for sufficiently large values of σ , by analogy with the Rayleigh criterion for resolving overlapping broad peaks: i.e. two such peaks are just resolvable when the full width of the peaks is comparable to their separation. Equations (9) and (10) consist of sums of Gaussian functions, i.e. they can be regarded as consisting of overlapping broad peaks, with their width determined by the value of σ and their spatial separation (for sufficiently large distances from an arbitrary origin point) being set by the interplanar distance between various lattice planes. Thus, for sufficiently large σ , the Gaussian functions in the summations in equations (9) and (10) corresponding to closely-spaced lattice planes (the spacing for which has $d \ll \sigma$) sum to give a featureless (r -independent) function, whereas the Gaussians associated with the furthest-separated lattice planes give rise to a resolved structure, i.e. oscillations with a period $D = d$, as long as $d \geq \sigma$. Numerical solutions of equations (9) and (10) (see figure 6 for a 2D example) do indeed reveal single-period oscillatory behaviour for large enough σ , even though we have not been able to show this outcome analytically.

5. Relation to the FSDP

As mentioned previously, the first sharp diffraction peak (FSDP), or prepeak, observed in the structure factor of (primarily network) amorphous materials and glasses is generally regarded as a signature of MRO [2], although the structural origin of this feature is still hotly debated

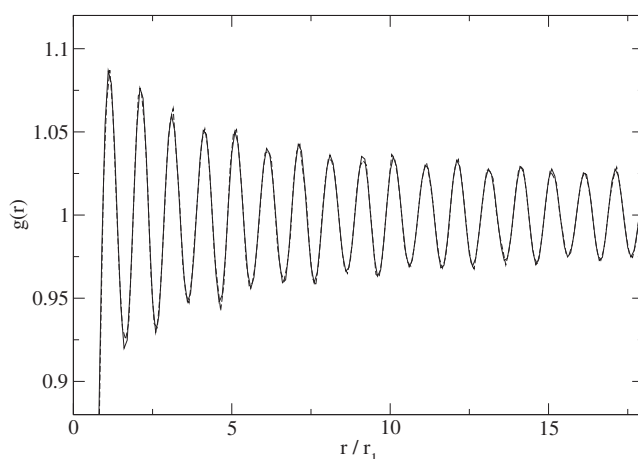


Figure 6. Single-period oscillatory behaviour in $g(r)$ for a positionally-disordered 2D square lattice with $\sigma = 0.25r_1$: the solid curve is $g(r)$ calculated numerically using equation (9), and the dashed curve is $g(r)$ calculated from the positions of the atoms in the disordered lattice.

[14–20]: explanations for the FSDP have been based for example on layer-like motifs [17, 20] or structurally-ordered interstitial voids [14, 18]. Gaskell and Wallis [20] have stated that, in many amorphous materials (although not all), the wavevector, k_1 , of the FSDP corresponds to the wavevector of a diffraction peak (the lowest in k) in the structure factor, $S(k)^{\text{cryst}}$, of a related crystalline phase, and thus, in real space, to the presence of a set of Bragg planes (with the largest interplanar spacing). They proposed that remnants of these lattice planes survive into the disordered phase, and that scattering from these planes is responsible for the FSDP. We have demonstrated here that the peak in $S(k)^{\text{cryst}}$ at the lowest value of k , corresponding to the set of lattice planes which are the furthest separated in real space and concomitantly have the largest areal atomic density, is the most resistant to the destructive effect of positional disorder.

A couple of positionally-disordered crystalline structures have been investigated, namely the diamond-cubic structure in which silicon crystallizes, and the low-temperature α -phase cristobalite structure of silica, which are crystalline analogues of the corresponding amorphous phases, a-Si and v-SiO₂, respectively, with a view to ascertaining the relationship of the extended-range order evident in strongly positionally-disordered crystalline variants of such structures and the FSDPs of the corresponding amorphous materials.

Amorphous silicon exhibits a strong FSDP at $k_1 = 2.00 \text{ \AA}^{-1}$ [21]. From table 1, the period, D , of the last surviving extended-range oscillations for the (Gaussian) positionally-disordered diamond lattice is the apex–basal-height distance in an SiSi₄ tetrahedron, $d_{111} = 4r_1/3$, where r_1 is the nearest-neighbour distance. In crystalline silicon, $r_1 = 2.35 \text{ \AA}$ [22], corresponding to an oscillation with a period $D = 3.13 \text{ \AA}$, and hence $k_1 = 2\pi/3.13 \text{ \AA} = 2.01 \text{ \AA}^{-1}$, in excellent agreement with the experimental value [21].

The FSDP is often observed in AX₂-type glasses [2, 14], the best known example being vitreous silica, v-SiO₂. This material is known [1, 2] to consist of fairly well ordered SiO₄ tetrahedra, linked in a corner-sharing fashion to form a disordered network. In positionally disordering the cristobalite structure in order to mimic the medium-range structure of v-SiO₂, no attempt was made to keep the tetrahedra ordered; Gaussian disordering was applied to every Si or O atom without restriction. Hence, these positionally-disordered models are not expected to be particularly good representations of the structure of real v-SiO₂, especially

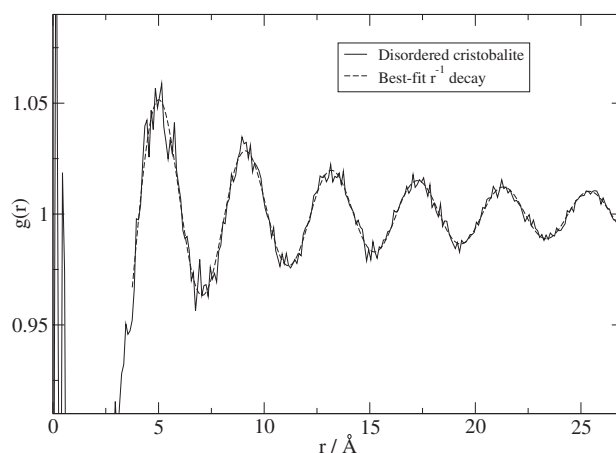


Figure 7. Pair distribution function, $g(r)$, calculated for a Gaussian-positionally-disordered model of α -cristobalite (with value of disorder parameter $\sigma = 0.8 \text{ \AA}$), together with a best-fit curve for the function $g(r) = 1 + Ar^{-1} \sin(k_1 r + \phi)$ used to obtain a value for the FSDP position $k_1 = 1.546 \text{ \AA}^{-1}$ and hence the spatial period of the extended-range oscillations, $D = 2\pi/k_1 = 4.066 \text{ \AA}$.

concerning the short-range order. Nevertheless, single-frequency, damped extended-range oscillations were also found in Gaussian-positionally-disordered structures of α -cristobalite for values of disorder parameter in the range $0.35 \leq \sigma/r_1 < 0.6$ (figure 7). The period of the last surviving oscillation in $g(r)$ was found to be $D = 4.07 \text{ \AA}$, corresponding to a peak in $S(k)$ at $k_1 \approx 1.54 \text{ \AA}^{-1}$, in very good agreement with the experimental value of the FSDP wavevector-transfer value of $k_1 = 1.52 \text{ \AA}^{-1}$ in $v\text{-SiO}_2$ [23].

Thus, it appears, at least for the cases of a-Si and $v\text{-SiO}_2$, that the wavevector-transfer value of the FSDP position for topologically-disordered real amorphous materials can be predicted from the position of the sole remaining peak in the structure factor with appreciable weight for positionally-disordered structures of the corresponding crystals, implying that, at sufficiently large distances from an arbitrary origin atom, the atomic structure of a topologically-disordered amorphous material appears to be similar to that of the corresponding positionally-disordered crystal.

6. Conclusions

Positional disorder, with atomic displacements taken from a Gaussian distribution, has been introduced into various 2D and 3D lattices. In all cases, it is found that, at sufficiently high degrees of positional disorder (as characterized by the width of the Gaussian distribution used), only a single Bragg peak with appreciable weight remains in the crystalline structure factor, that with the lowest value of wavevector transfer, corresponding to the lattice planes with the largest separation and highest areal density. Such structures also exhibit corresponding spatially power-law damped, single-period extended-range oscillations in the real-space, orientationally-averaged pair-distribution function, whose period is equal to the interplanar spacing of the furthest-separated lattice planes. The position of the sole remaining peak with significant weight in the structure factor for highly-positionally-disordered lattice structures is the same as that of the first sharp diffraction peak observed in the structure factor of amorphous materials, e.g. silicon or silica. This methodology also may offer the promise of a solution to the famous long-standing Gauss circle problem in pure mathematics.

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