

Medium-range structure of amorphous silicon studied by the Voronoi-Delaunay method

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Structural inhomogeneities and their effect on the dynamics are investigated for a molecular dynamics model of quenched amorphous silicon. The structure of the model is analysed with the help of the Voronoi-Delaunay approach, which is a convenient tool for this purpose. The silicon structure is found to be rather homogeneous. Only localized defects are found, and no medium-range regions of 'imperfect' structure as were observed in a Lennard-Jones glass by V. A. Luchnikov, N. N. Medvedev, Yu. I. Naberukhin and V. N. Novikov (1995, *Phys. Rev. B*, **51**, 15 569). It is assumed that the inhomogeneity in glasses is the result of competition between the tendency of atoms to pack locally into the most favourable arrangements and the necessity to realize space filling structures. For spherical atoms a regular tetrahedron of four atoms is the densest local configuration; however, such units cannot cover the whole space. As a result, Lennard-Jones glasses have regions of structural inhomogeneity with an extension of 3-5 interatomic distances. Amorphous silicon is not governed by close packing: its structure is a tetrahedral bond network. The aforementioned conflict is not relevant here, because the disordered tetrahedral network can fill the space with minor distortions of bond lengths and tetrahedral angles.

1. Introduction

New concepts for the description of the structure of non-crystalline states have been developed during the past few years. Traditionally, amorphous phases are considered as homogeneous systems with some average local or 'short-range' order, preferentially described by pair distribution functions. But this approach does not appear to be well suited to the explanation of various experimental facts. The anomalously high heat capacity [1, 2] at low temperatures, as well as the excess density of vibrational states in comparison with the Debye distribution ('boson' peak), observed in glasses by low-frequency Raman scattering [3, 4], far-infrared absorption [5], and inelastic neutron scattering [6] indicate the existence of mesoscopic inhomogeneities, caused by regions of tens or hundreds of molecules [7-11]. Thus, the idea of structural inhomogeneities of nanometric size and the problem of 'medium-range order' has become relevant [12, 13]. On the other hand, computer simulation techniques can be employed successfully to study directly the structure of amorphous phases. This means that we are restricted no longer to simplified structural models of the amorphous state. Molecular dynamics or Monte Carlo simulations give us the coordinates of all atoms of a system. All of the information on the mutual arrangement of atoms can be extracted from these data. Although the route from the set of atomic

coordinates to suitable structural properties has its own difficulties, the development of methods for solving this problem is progressing rapidly [14–19].

It has been assumed in our recent papers [20, 21] that the universality of some characteristics observed in glasses may be related to the common geometric nature. The inhomogeneities result from competition between the tendency of atoms to pack locally into the most favourable arrangements and the necessity for realizing a space filling structure. The nature of this conflict is quite understandable for spherical atoms. Indeed, the regular tetrahedral arrangement of four atoms is locally the most dense and energetically favourable, but it is known that these arrangements fail to be space filling. Therefore, in close-packed crystalline structures tetrahedral holes alternate with octahedral. Dense liquids contain a fraction of local arrangements that are similar to the regular tetrahedral ones [22–25]. Being energetically most favourable, these are less subject to restructuring during fast freezing. As a result, a non-crystalline phase arises because the initial arrangement of tetrahedral units in liquids has no translational symmetry. Besides, these units usually form aggregates that are not impossible for crystals (branching clusters and five-membered rings) [14, 22, 26].

According to a detailed study of the structural inhomogeneities in vitreous Lennard-Jones systems, one can distinguish easily regions of ‘perfect’ and ‘imperfect’ structure [20]. The environment of atoms situated in regions with perfect structure can be described by local structural elements (Delaunay simplices) with shapes similar to the shapes formed by regular tetrahedra or quatoctahedra, i.e., similar to the shapes of perfect local structures in crystals. However, the mutual arrangement of these perfect structural elements is different from the infinite repetition in crystals, and limited to associates on the scales of 3–5 interatomic distances. Between these regions the arrangement of atoms is imperfect: the shape of local arrangements differs from the given perfect shapes.

Of interest is the fact that these regions are not only geometric characteristics of the system but also display distinct physical properties. In a recent paper [21], the separate velocity autocorrelation functions were determined and used to calculate the corresponding vibrational densities of states. It turns out that low-frequency vibrations are more characteristic for the regions with the imperfect structure. This confirms the concept that nanometric scale structural inhomogeneities give rise to the excess of low-frequency modes, compared with crystals.

Particles with more complex interaction potentials will have other preferred local arrangements. One can imagine that the more difficult it is to fill space by a given structural element, the easier it is for the system to have a structural inhomogeneity and to turn into a glass. On the other hand, if the local arrangements can fill the space without conflicts, such systems may be more homogeneous, and the structure should transfer to the crystalline state more easily. Silicon provides an example of such a system. The optimal local arrangement of silicon atoms is four neighbours tetrahedrally surrounding a central atom. This configuration is an element of crystalline structures, particularly of the diamond lattice into which silicon crystallizes. Indeed, it is not possible to vitrify liquid silicon by melt quenching: amorphous Si is obtained by precipitation from vapour. Non-crystalline silicon exists as a disordered tetrahedral network, filling the space with minor distortions of bond lengths and tetrahedral angles [27, 28]. This tetrahedral network of Si—Si bonds is a generally accepted model of amorphous silicon. It is known, however, that amorphous Si usually has defects, observed particularly in the deviation of the average number of nearest neighbours of atoms from four.

In this paper we try to study the structure of amorphous silicon on an intermediate scale to find structural inhomogeneities and their dynamic peculiarities. However, we do not rely on the concept of a network of Si—Si bonds, the construction of which for molecular dynamics models always bears some arbitrariness, but have used a purely geometric approach.

2. Model

A model of amorphous silicon, containing 1000 atoms in a cube with periodic boundary conditions, has been produced for structural analysis by molecular dynamics. The equilibrium conditions correspond to $T = 300$ K and density $\rho = 2.294$ g cm⁻³. The interatomic potential allowing for both pair and three-particle interactions has been taken as proposed by Stillinger and Weber [29]:

$$V = V_2 + V_3, \quad (1)$$

$$V_2(r_{ij}) = A(Br_{ij}^{-4} - 1) \exp\left(\frac{1}{r_{ij} - a}\right), \quad (2)$$

$$V_3(r_{ij}, r_{ik}, r_{jk}) = h(r_i, r_j, r_k) + h(r_j, r_i, r_k) + h(r_i, r_k, r_j), \quad (3)$$

$$h(r_i, r_j, r_k) = \lambda \exp\left(\frac{\gamma}{r_{ij} - a} + \frac{\gamma}{r_{jk} - a}\right) (\cos \theta_{ijk} + \frac{1}{3})^2. \quad (4)$$

The following values have been used: $A = 0.70495$; $B = 0.60222$; $a = 1.80$; $\lambda = 21.0$; $\gamma = 1.20$. Energy is given in units of $\epsilon = 3.4723 \times 10^{-19}$ J; distance r is in units of $\sigma = 2.0951$ Å. This effective potential certainly is not designed to reproduce the direct interaction of two atoms, but reproduces the prevalent local structure in condensed matter by the fact that three atoms forming a tetrahedral angle have a minimal potential energy. The equations of motion have been integrated in terms of the algorithm given in [30] with a time step of 0.4 fs. The molecular dynamics program uses a temperature and pressure scaling procedure [31] to attain preset values. This allows us to perform a gradual relaxation of the system to a given thermodynamic state.

To prepare the amorphous state of silicon, we start from the liquid state at a temperature of 1700 K and zero pressure, yielding the density $\rho = 2.46$ g cm⁻³ [32]. This state had been equilibrated for 40 ps. The process of vitrification and further simulation of the equilibrium amorphous state involved a modification of the expression for the three-particle interaction $h(r_i, r_j, r_k)$ proposed by Dodson [33], according to which the cosine in expression (4) is substituted by $(\cos \theta_{ijk} + \frac{1}{3})^2 + \eta(\cos \theta_{ijk} + \frac{1}{3})^4$. Dodson studied the homoepitaxial growth of an Si crystal on the (111) surface using the Monte Carlo method and found that the original Stillinger–Weber potential is inadequate for describing this process. In our calculations we used the value $\eta = 0.3$. In test runs we had also used the original potential, but in this case a much slower cooling rate is necessary to avoid an exceedingly large fraction of atoms with five neighbours, in accord with experimental pair distributions functions [34–37].

Starting with the configuration of the liquid, the sample was quenched to 300 K within a time period of $t_{\text{quench}} = 100$ ps. During this cooling process the temperature was varied with time according to $T = 1700 - 1400(t/t_{\text{quench}})^3$ K, keeping zero pressure. The state obtained is considered to be amorphous silicon (see following). The subsequent molecular dynamics evolution of the sample has been performed at constant temperature and pressure.

'Vibrationally averaged' configurations (V structure) of amorphous Si were

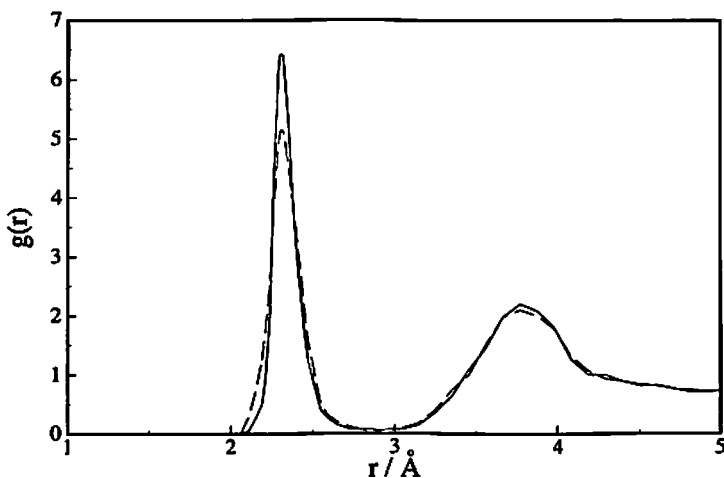


Figure 1. The pair correlation functions for vibrationally averaged (solid curve) and instantaneous (dashed curve) configurations of amorphous Si.

constructed to demonstrate the structural motives in the sample. To this end, the coordinates of each atom were averaged over 50 successive configurations of the original molecular dynamics run. This interval has been chosen as the optimal one for reaching the maximum narrowing of the first peak of $g(r)$ (see figure 1).

3. Local structure

The first maximum of $g(r)$ is separated from the second by a deep minimum at $r_1 = 1.3\sigma$ (figure 1). This allows the first coordination shell of the atoms to be defined clearly. The corresponding coordination number is 4.04. The minor excess over four is due to the fact that about 6.1% of the atoms have five neighbours. Simultaneously, 2.2% of the atoms have less than four. Independent of the number of particles in the first coordination sphere, the angles between the four nearest neighbours were measured for each atom. The mean value of the angle is $\alpha = 109.5^\circ$, with a mean-square deviation $\delta = 10.4^\circ$. These values are very close to those obtained by Polk [27] for the disordered tetrahedral network model.

Thus, in terms of conventional criteria, our model of amorphous silicon can be considered as a well defined tetrahedral network. However, in this network there are a few topological defects. If the Si—Si bonds are defined as existing between the central atom and its neighbours in the first coordination shell, there are some atoms with extra and some with missing bonds. If we take exactly four bonds per atom, being formed with the four nearest atoms, we get a small percentage of 'one-sided' bonds. This arises when a first atom is connected with a second, but the second atom does not include the first one in its four nearest neighbours.

The degree of tetrahedrity of the nearest environment of the silicon atoms can also be estimated using the measure T (tetrahedrity) determined from the formula [38]:

$$T = \frac{\sum_{i>j} (l_i - l_j)^2}{15\bar{l}^2}, \quad (5)$$

where l_i is the length of the i th tetrahedron edge (the distance between vertices), and \bar{l} is the mean edge length of the given tetrahedron. For a perfect tetrahedral

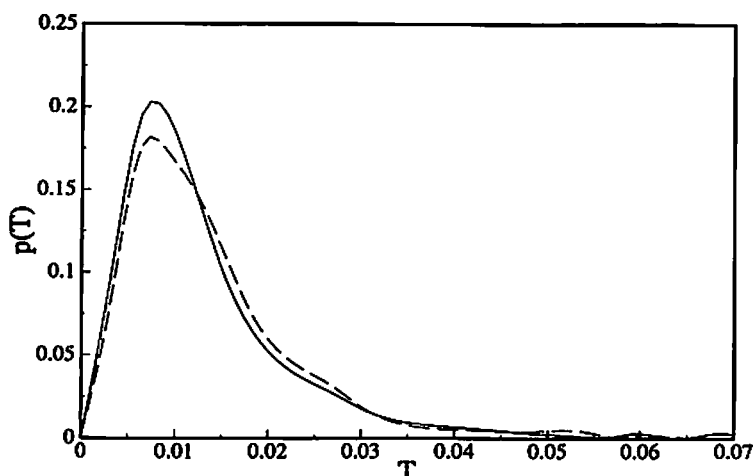


Figure 2. Frequency distribution of perfectness T (tetrahedrity) of the nearest environment for the atoms of amorphous Si in vibrationally averaged (solid curve) and instantaneous (dashed curve) structures of amorphous Si.

configuration the value of the measure T evidently is equal to zero. For minor distortions of the shape the values of the measure are also small. The larger the shift of the atoms from a regular tetrahedron, the larger is the measure. The tetrahedrity was proposed for studying the shapes of the Delaunay simplices in dense non-crystalline packings. In this case, however, we determine the tetrahedra of the four nearest neighbours of the silicon atoms. Figure 2 demonstrates the distribution of T for our amorphous Si model. It is seen that the local order of the majority of atoms is close to the regular tetrahedron. The mean value of T is 0.0125 with a mean-square deviation of 0.0107. For the atoms having exactly four neighbours in the first coordination sphere the mean value of T is 0.0106. For the 8.3% of atoms remaining the mean T is 0.0331 which is twice as much as the 'boundary' value $t_b = 0.018$ used in the previous papers for 'good' tetrahedra [15]. Note that the distribution of the measure T is almost the same for the V structure as for the instantaneous configurations (I structures). This means that the existing variety of structural distortions in amorphous silicon is of an intrinsic structural nature rather than a result of thermal fluctuations.

We have calculated also the Voronoi polyhedra for all atoms of our system. The mean-square deviation of its volume is $0.112\sigma^3$, which is only 5.1% of the mean value $V = 2.212\sigma^3$. This points to the absence of noticeable density fluctuations in our model. Note that a weak anticorrelation can be observed between the tetrahedrity and the volume of the Voronoi polyhedra. The better the tetrahedrity of a local environment, the larger the volume that is assigned to the given atom. The same anticorrelation between the mean density and the tetrahedral order in the system has been observed when analysing water models [39]. This is attributed to the nature of a regular tetrahedron with four atoms surrounding the central one.

4. Structural motives of the Delaunay network

The regularities of a medium-range structure can be described conveniently with the help of a geometric Voronoi–Delaunay approach [14, 15]. It does not need any physical bonds between atoms and therefore is free of the problems connected with

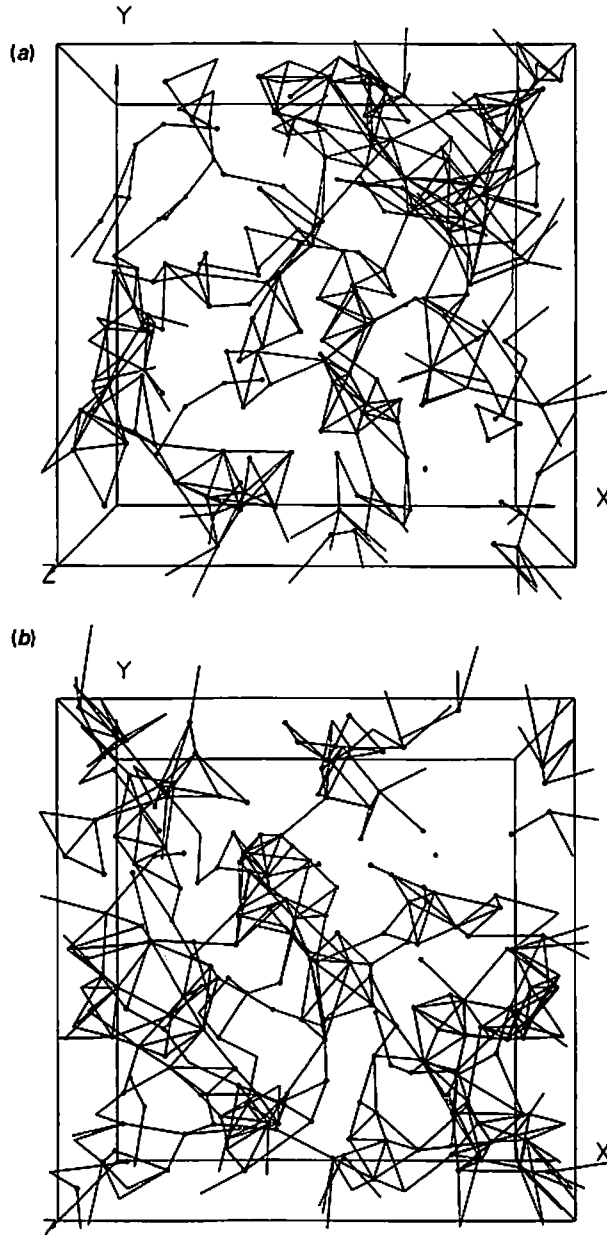


Figure 3. Clusters of atoms with (a) more regular and (b) more disordered structural surroundings. 20% of all atoms are shown in each.

their definition and determination. This approach employs two geometric networks: Voronoi and Delaunay. One can choose between them according to the concrete problem. The Delaunay network seems quite suitable for the present case.

The Delaunay network can be obtained by constructing the Voronoi polyhedra around each atom of the system. The sites of the Delaunay network coincide with the atomic centres, and the edges (the bonds of the network) connect 'geometric neighbour sites' (which share a common face of their Voronoi polyhedra). Note that according

to this definition in our model every atom has, on average, 18.5 geometric neighbours. They include first, second, and in part, third neighbours of the network of 'physical' Si—Si bonds.

In order to detect the mutual arrangement of atoms with the most regular tetrahedral local order one should label (colour) those atoms (sites on the Delaunay network) which have the smallest values of the measure T . Similarly, in order to find aggregates of atoms with a strongly distorted environment one should colour only the sites which correspond to the largest values of T . For a pictorial illustration of such aggregates we have taken one V structure configuration of our model. Figure 3(a) shows the 20% of atoms with the best tetrahedrality, and figure 3(b) the same number with the worst one. In the first case the value of T for any atom is less than 0.005, and greater than 0.018 in the second case. The lines depicted are the bonds of the Delaunay network connecting 'coloured' geometric neighbours (not only 'covalently bonded' Si atoms), therefore three-membered rings can be recorded in the figures. Note that the aggregates obtained are not at all compact regions. This distinguishes amorphous silicon from argon in which a separation between aggregates of atoms with different degrees of 'perfection of the nearest environment' has been quite obvious ([21], figures 1 and 2). In silicon these aggregates are likely to have a more random nature. For comparison, figure 4 shows clusters on the silicon Delaunay network obtained by a random colouring of 20% of the sites.

To estimate quantitatively the correlation between the chosen atoms, a correlation coefficient for the neighbours on the Delaunay network has been introduced. For the first neighbours on the Delaunay network it is

$$k_1 = \frac{\overline{T_i T_j} - \bar{T}^2}{\bar{T}^2}, \quad (6)$$

where T_i and T_j are the values of the measure T for the i th and j th atom, which are geometric neighbours. It appeared that for our silicon model this coefficient of correlation is rather small: $k_1 = 0.12$. For comparison, the same correlation coefficient for the 'perfection' of the local structure in amorphous argon [21] was determined to be much larger: $k_1 = 0.37$. Similarly, the correlation coefficient k_2 can be introduced for the second neighbours on the Delaunay network. In amorphous silicon this turned out to be almost zero, and in argon $k_2 = 0.109$. With an independent random colouring of the sites the values of k_1 and k_2 are in the range ± 0.008 . This scattering can be considered as a measure of the accuracy of the correlation coefficients.

Similar investigations have been performed for the volumes of the Voronoi polyhedra which give a measure of the local density. Figure 5(a) labels the 20% of atoms with the smallest volumes of the Voronoi polyhedra and figure 5(b) shows those with the largest. The correlation coefficients of the volumes of the Voronoi polyhedra are somewhat higher than for the measure T : $k_1 = 0.16$ and $k_2 = 0.005$. However, this is attributed partially to a trivial geometric fact, namely, that atoms participating in dense (or loose) local aggregates are geometric neighbours automatically. Note that the density of the clusters is similar because, as mentioned earlier, the width of the polyhedra volume distribution is rather narrow.

5. Partial velocity autocorrelation functions and the density of vibrational states

It has been demonstrated [21] that in amorphous argon the dynamic behaviour of the atoms in the regions of perfect structure differs from that of the atoms in the

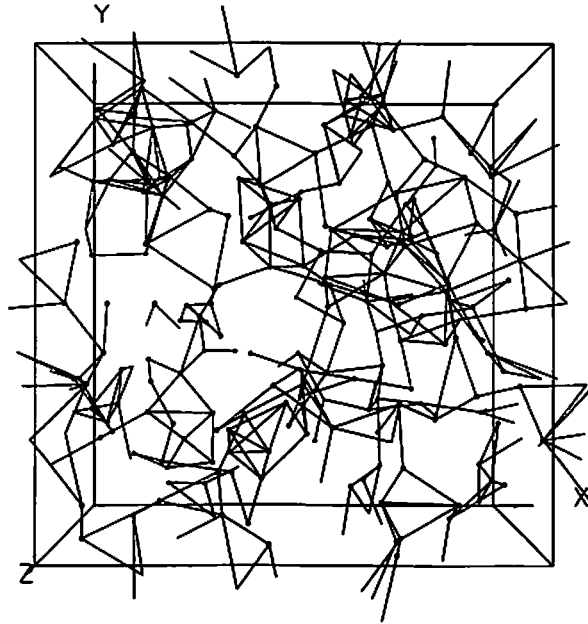


Figure 4. Clusters of randomly chosen atoms on the silicon Delaunay network. 20% of all atoms are shown.

regions of imperfect structure. This was obtained from an analysis of the partial velocity autocorrelation functions. These functions are calculated as

$$K(\tau) = \langle v(t)v(t+\tau) \rangle, \quad (7)$$

where the averaging is performed over time and the given types of atom.

Figure 6 (dashed curves) shows the partial velocity autocorrelation functions for the 20% of atoms with the best and, accordingly, the worst tetrahedrality of the nearest environment. The corresponding aggregates of atoms were shown in figure 3. The solid curves denotes the function $K(\tau)$ averaged over all the atoms of the system. Its overall behaviour is typical for amorphous silicon [40, 41]. The amplitude of the oscillations of the partial $K(\tau)$ for the atoms with a high degree of tetrahedrality is larger than on average. In contrast, the group of atoms with a low degree of tetrahedrality demonstrates a lower amplitude.

The same method has been used to calculate the partial velocity autocorrelation functions for the 20% of atoms distinguished by the largest and smallest volumes of the Voronoi polyhedra. However, these appeared to be almost identical, which is interesting because with this colouring of the Delaunay network more strongly correlated clusters than in the case of the measure T have been observed (compare figures 3 and 5). This means that the symmetry of the nearest surrounding is more important for the dynamic behaviour of atoms than minor variations of local density.

The velocity autocorrelation function is related to the spectral density of vibrational states via the Fourier transformation [42]

$$Z(\omega) = \frac{1}{\pi} \int_0^{\infty} \cos(\omega\tau) K(\tau) d\tau. \quad (8)$$

The spectral density of the vibrational states of amorphous silicon obtained by

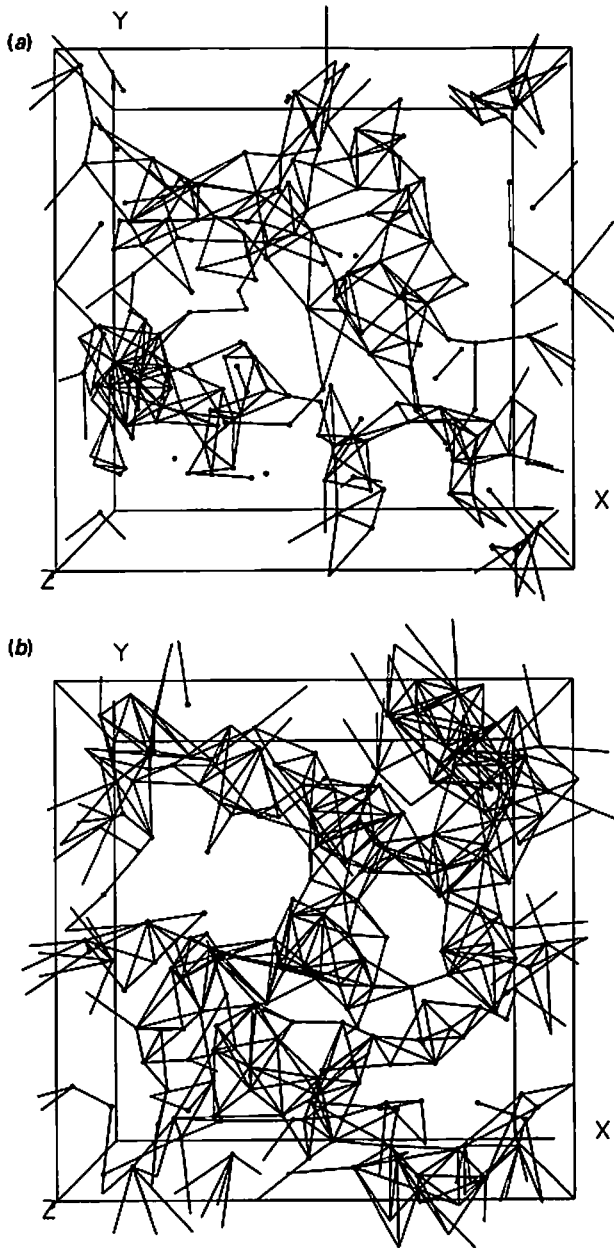


Figure 5. Clusters of atoms with (a) smallest and (b) largest volumes of Voronoi polyhedra. 20% of all atoms are shown in each.

transformation of the mean function $K(\tau)$ is depicted in figure 7 by a solid curve. The locations of the peaks at 20 meV and 63 meV coincide with the positions of the transverse acoustic (TA) branch and transverse optical (TO) branch of the spectrum of polycrystalline silicon. Two weak intermediate peaks at 38 meV and 47 meV correspond approximately to the maxima of the longitudinal acoustic (LA) branch and longitudinal optical (LO) branch of the spectrum [41]. The period of the dominant high frequency oscillations observed in the autocorrelation functions (figure 6) fairly

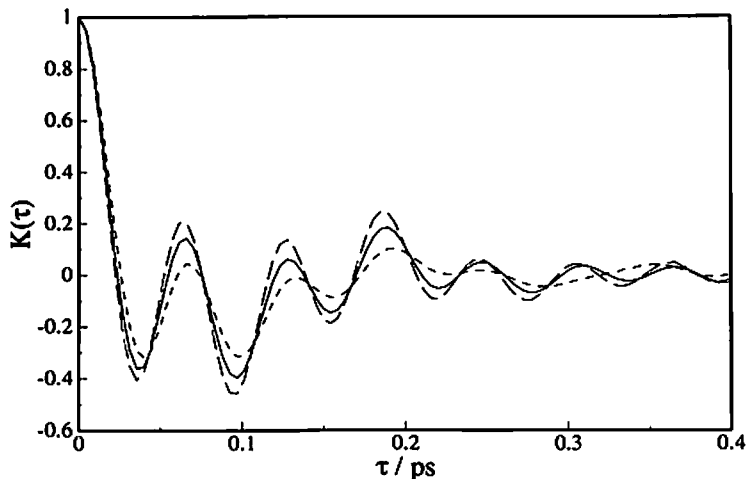


Figure 6. Velocity autocorrelation functions. The solid curve relates to the total system; the long-dashed curve to atoms with more regular (figure 3(a)) and the short-dashed curve to atoms with more disordered (figure 3(b)) structural surroundings.

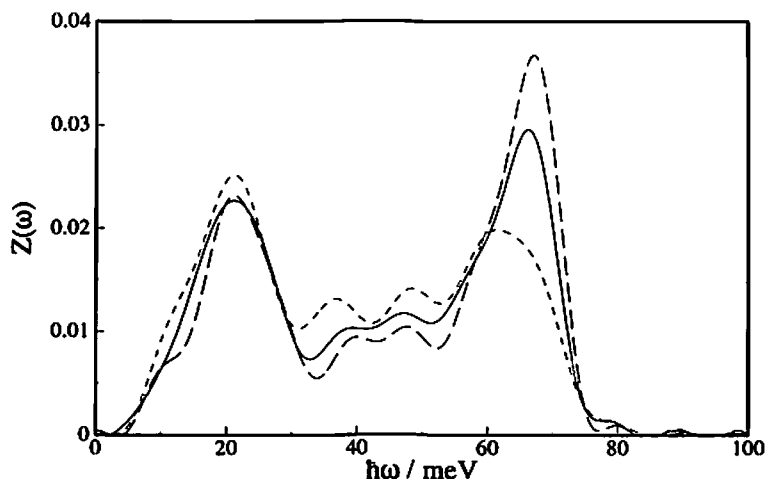


Figure 7. Fourier transforms of the velocity autocorrelation functions shown in figure 6.

coincides with the reverse frequency of TO vibrations. The dashed curves in the same figure depict the partial densities of states obtained by the Fourier transformation of the corresponding partial velocity autocorrelation functions. The main difference is observed in the region of the TO peak, where the density of vibrations for atoms with a perfect tetrahedral environment is almost twice the size of that for atoms with a non-tetrahedral one. In the crystalline solid state this vibrational branch corresponds to an anti-phase displacement of neighbouring atoms. Therefore the TO branch is more sensitive to the local structure than the TA branch, for which neighbours are moving with the same phase. In addition, a slight frequency shift of the TO peak is observed which is in accord with this interpretation: for large values of the measure T the peak is shifted to lower frequencies, because Si—Si-bonds are weakened by more distorted surroundings due to the influence of the three-body interaction term. We can see some excess density of vibrational states at low frequencies. However, compared with

amorphous argon [21] this excess is rather weak. In conclusion, one can say that amorphous silicon does not exhibit a medium-range 'imperfect' structure and, consequently, we do not observe excess low frequency vibrations in this model.

6. Discussion

The spatial distribution of atoms with different local structural environments are studied by a molecular dynamics model of amorphous silicon. The distribution of atoms with the most regular and with the most irregular tetrahedral surroundings differ slightly from a random distribution. However, there are no well separated regions of 'perfect' or 'imperfect' structure in amorphous silicon, as has been discussed recently for quenched argon. Analysis of the atomic dynamics reinforces this result. The partial spectra of the density of vibrational states of the atoms belonging to the different groups demonstrate a difference only for high frequency vibrations, which are sensitive to any local tetrahedral order of the atoms. An appreciable excess density of vibration states at low frequencies (like the 'boson' peak), which would result from medium-range structural inhomogeneities according to the present concepts, is not manifested.

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