PHYSICAL REVIEW B VOLUME 28, NUMBER 10 15 NOVEMBER 1983

Phenomenological Lagrangian for the amorphous solid state

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(Received 7 December 1982; revised manuscript received 1 March 1983)

We construct a model which in a simple way takes into account the influence of the long-range correlations in an amorphous solid. With the Lagrangian which we propose, we are able to explain the existence of local minima which do not correspond to the crystalline state. A two-dimensional lattice is considered in some detail. Some relations with the experimental data are briefly discussed.

There is a common opinion[1-3] that in order to describe and understand the amorphous solid state such as silicates and glasses we have to take into account the long-range correlations, i.e., the influence of atoms placed beyond the closest neighbors of a given atom. As a matter of fact, if we consider atoms with valence m, and if the contribution to the energy comes only from their bonds with their m closest neighbors, it is difficult to conceive how a single atom would "know" whether it is placed in a regular crystalline lattice, or in a random m-coordinated lattice. If there is any significant difference, it comes from the structure of the neighborhood containing more than that atom's immediate neighbors.

We shall construct a single phenomenological model and apply it to the simplest case of a tricoordinated lattice in two dimensions. As we shall see, the model displays quite interesting properties, and enables us to explain why in some cases a random lattice is energetically preferable as compared with the regular one.

There exist only four regular tricoordinated lattices on the plane containing only perfect equilateral polygons (Fig. 1); we shall denote them by respective symbols (6,6,6), (4,8,8), (4,6,12), and (3,12,12); these symbols have the following meaning: As each atom belongs to the three adjacent polygons, the three numbers in the symbol define their number of sides. In each of these cases the three angles between the bonds linking the atom to its immediate neighbors are $\alpha_1=\alpha_2=\alpha_3=2\pi/3$; $\alpha_1=\pi/2$, $\alpha_2=\alpha_3=3\pi/2$; $\alpha_1=\pi/2$, $\alpha_2=2\pi/3$, $\alpha_3=5\pi/6$; finally $\alpha_1=\pi/3$, $\alpha_2=\alpha_3=5\pi/6$.

Let us now consider a random lattice, in which all the polygons are convex and equilateral, but not necessarily perfect (i.e., with equal angles). It may contain triangles, rhombs, pentagons, etc., in some unknown proportions; one should not however expect a lot of n-gons with a very large value of n, which would resemble a bubble in the midst of the lattice.

Consider an elementary tripod, i.e., an atom together with its three closest neighbors linked to each other via the covalent bonds. If it could be left free, by removing all the rest of the surrounding lattice, it would have taken on the most symmetric configuration in which $\alpha_1=\alpha_2=\alpha_3=2\pi/3$. The simplest Lagrangian displaying an absolute minimum in this configuration is obviously given by

$$L_0=(\alpha_1-\alpha_2)^2+(\alpha_2-\alpha_3)^2+(\alpha_3-\alpha_1)^2\ , \qquad (1)$$

where $\alpha_3=2\pi-\alpha_1-\alpha_2$; so, up to an additive constant,

$$L_0=6\alpha_1^2+6\alpha_2^2+6\alpha_1\alpha_2-12\pi\alpha_1-12\pi\alpha_2\ . \qquad (2)$$

On the other hand, if we consider an n-sided equilateral polygon found in the lattice, it would also take on the regular symmetric shape with all its angles equal, $\beta_1=\beta_2=\cdots=\beta_n$, if the rest of the surrounding lattice disappeared. Let us note that this shape gives a maximum for the polygon's surface. Therefore, the Lagrangian displaying a *maximum* for this configuration can be made proportional to the surface.

Let us finally consider an elementary cell of the lattice, containing an elementary tripod and the three adjacent polygons, with numbers of sides respectively n_1, n_2, and n_3 (altogether $n_1+n_2+n_3-5$ atoms) (Fig. 2). The shape of such a cell is fully determined by fixing the three numbers n_1, n_2, and n_3, and all the angles in the polygons. It is easy to see that only $(n_1+n_2+n_3-10)$ of these angles are independent. Let us call the angles in the tripod α_1, α_2, and α_3 (with $\alpha_3=2\pi-\alpha_1-\alpha_2$), and the angles of the polygons $\beta_{(1)1},\beta_{(1)2},\ldots,\beta_{(1)n_1};\beta_{(2)1},\beta_{(2)2},\ldots,\beta_{(2)n_2};\beta_{(3)1},\beta_{(3)2},\ldots,\beta_{(3)n_3}$; of course in each set of β's one angle is identical with one of the α's.

Following the suggestions formulated above, we postu-

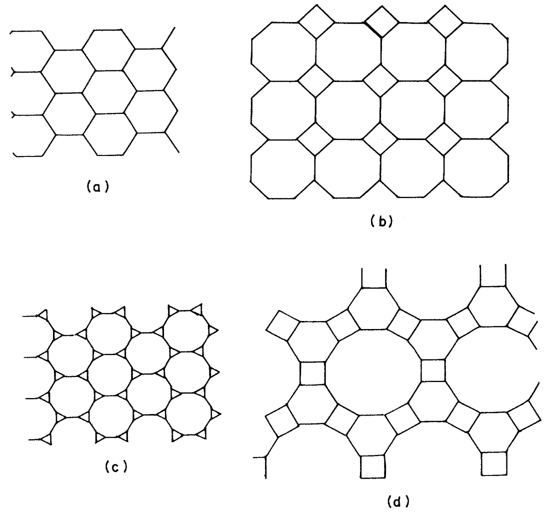

FIG. 1. Four tricoordinate regular homogeneous lattices on the plane: (a) (6,6,6), (b) (4,8,8), (c) (3,12,12), and (d) (4,6,12).

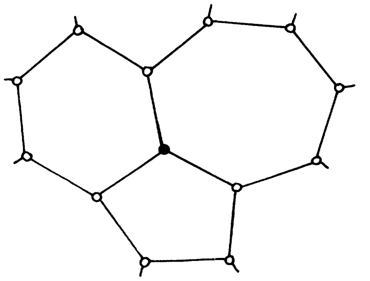

FIG. 2. Elementary cell in the amorphous lattice. $(n_1,n_2,n_3)=(5,6,7)$.

late the Lagrangian of the cell to be equal (up to a constant) to

$$L=6\alpha_1^2+6\alpha_2^2+6\alpha_1\alpha_2-12\pi\alpha_1-12\pi\alpha_2 +\lambda(S_{n_1}+S_{n_2}+S_{n_3}) \tag{3}$$

with S_{n_i} denoting the surface of the ith polygon $(i=1,2,3)$, and λ some constant describing the relative weight of the two contributions to the energy.

The formulas giving S_n for different values of n and for arbitrary angles are quite cumbersome; for example, the surface S_5 of an equilateral pentagon with angles $\beta_1,\beta_2,\ldots,\beta_5$ is equal to

$$S_5=\tfrac{1}{2}\sin\beta_1+\tfrac{1}{2}\sin\beta_3 -2\sin\frac{\beta_1}{2}\sin\frac{\beta_3}{2}\sin\left[\beta_2+\frac{\beta_1}{2}+\frac{\beta_3}{2}\right] \tag{4a}$$

while the surface S_6 of a hexagon is given by

$$S_6=\tfrac{1}{2}\sin\beta_1+\tfrac{1}{2}\sin\beta_3+\tfrac{1}{2}\sin\beta_5 -2\sin\frac{\beta_1}{2}\sin\frac{\beta_3}{2}\sin\left[\beta_2+\frac{\beta_1}{2}+\frac{\beta_3}{2}\right] \tag{4b}$$

and so on.

For a cell (n_1,n_2,n_3) the Lagrangian depends on $(n_1+n_2+n_3-10)$ independent parameters. It is obviously out of the question, for technical reasons, to minimize the Lagrangians of this type for all the elementary cells in the random lattice; therefore, we should simplify the problem by introducing some mean value parameter instead of the multitude of real angles of the lattice.

First of all, as the maximum of S_n is obtained when all the angles are equal, we shall replace the complicated expressions in (4) by a simple function of *one* variable, displaying the maximum at the same time as the corresponding S_n; supposing that the real angles in a random lattice take on the values not very far from the perfect polygons' values, the error will not be very great.[4] A suitable simple substitution we choose is

$$S_n\rightarrow A_n\{(n-1)\sin\alpha+\sin[(n-2)\pi-(n-1)\alpha]\} \tag{5}$$

with the normalizing coefficients A_n chosen in such a way, that for the value of α corresponding to the angle of a perfect polygon (i.e., $[(n-2)/n]\pi$ for the n-gon) our expression will take on the exact value of S_n. One can easily compute, for example, $A_5=0.3618$, $A_6=0.5$, $A_7=0.664$, etc.

We also have to fix the precise meaning of the variable α. We think that the *geometrical mean value* is a good choice; that is, we shall put for a cell with the three adjacent polygons n_1,n_2, and n_3,

$$\alpha=(\beta_{(1)1}\beta_{(1)2}\cdots\beta_{(1)n_1}\beta_{(2)1}\cdots\beta_{(2)n_2}\beta_{(3)1}\cdots\beta_{(3)n_3})^{1/(n_1+n_2+n_3)} \tag{6}$$

rather than the *arithmetic mean value* over the three polygons. Such a choice becomes convincing if we look at the plot in Fig. 3, which shows the relation between the inverse surface density and α for the eleven regular homogeneous lattices existing on plane. The four tricoordinate lattices are found beyond the angle $2\pi/3$; the three tetracoordinate ones are found near $\pi/2$; then we have two pentacoordinate lattices and only one hexacoordinate (tiling the plane with the perfect triangles). The numerical values for the tricoordinate lattices on plane are given in Table I (see also Ref. 4).

Although not *exact,* the inverse density D^{-1} of the lattice seems to be proportional to the geometrical mean value of the angles in an elementary cell; at least on the qualitative level the fits are quite satisfying (if we take the arithmetical mean value instead, the fits are less close to straight lines).

With this in mind we can analyze in more detail the Lagrangian (3), modified as follows:

$$L=3\alpha^2-4\pi\alpha +\lambda\sum_{k=3}^{N}P_kA_k\{(k-1)\sin\alpha +\sin[(k-2)-(k-1)\alpha]\}\ , \tag{7}$$

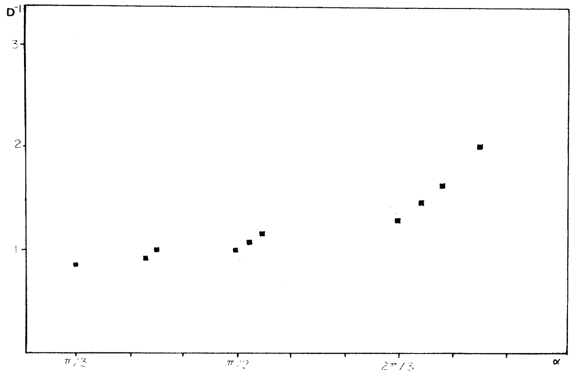

FIG. 3. Inverse density D^{-1} (in arbitrary units cm^{-2}) as function of the mean geometrical value α of angles in an elementary cell. The 11 regular lattices fall into four groups, one hexacoordinate, two pentacoordinate, three tetracoordinate, and four tricoordinate. The dependence is close to linear. The extrapolation to the straight lines passing through these groups of points enables us to evaluate D^{-1} for any α corresponding to the amorphous states.

TABLE I. The numerical values for the tricoordinate lattices on plane.

Lattice type	Density D	Inverse density D^{-1}	Arithmetic mean value	Geometric mean value
(6,6,6)	0.7698	1.2990	120°	120°
(4,8,8)	0.6863	1.4571	126°	124°30′
(4,6,12)	0.6188	1.6160	130°55′	128°37′
(3,12,12)	0.4974	2.0104	140°	135°30′

where λ as before is the overall ratio of the two contributions (tripods versus polygons), A_k are the normalizing constants introduced above, and P_k is the relative frequency of the k-sided polygons in the lattice. Note that if we call p_k the probability of finding a k-sided polygon among the three polygons of an elementary cell chosen at random, then obviously

$$p_k/p_j = kP_k/jP_j \tag{8}$$

and of course, in both cases

$$\sum p_k = 1, \quad \sum P_k = 1 \tag{9}$$

which gives

$$p_k = \frac{kP_k}{\sum kP_k} \ . \tag{10}$$

The fact that we are using the probabilities P_k comes from the way we have constructed our mean value Lagrangian; this is an important point which needs to be underlined.

The first term $3\alpha^2 - 4\pi\alpha$, no matter how it is parametrized, is obtained as follows: We consider all the tripods of the lattice whose number is exactly the number of atoms present (we neglect, of course, the boundary which is supposed to be at infinity); for each of them we take the corresponding contribution given by (2), add them up and divide by the number of atoms.

The second term is obtained by considering all the polygons of the lattice, taking the contributions proportional to their surfaces S_k, adding them up together, and dividing by the number of all polygons present in the lattice; this will yield the relative frequencies P_k in front of each "mean valued" surface S_k.

Now comes the subtle point: How should we add up the two contributions? Of course, there is the unknown relative strength of these two types of energy visualized by the constant λ in formula (7), but this is not enough. We have to translate, so to speak, the two different ways of averaging, the averaging "per atom" and the averaging "per polygon." To do so, it seems reasonable to admit that the surfacial energy term of a k-sided polygon will equally contribute to the energy of the k adjacent tripods; in other words, when we add up the two kinds of contributions, the terms P_kS_k should be taken *with the relative weight* k^{-1}. Therefore, the final version of the Lagrangian is

$$L = 3\alpha^2 - 4\pi\alpha + \lambda \sum_{k=3}^{N} \frac{1}{k} P_k A_k \{ (k-1)\sin\alpha + \sin[(k-2)\pi - (k-1)\alpha] \} \ . \tag{11}$$

Let us carry on with a simplified model in which the lattice is supposed to contain only five-, six-, and seven-sided polygons. This means that all the P_k's vanish except P_5, P_6, and P_7; of course $P_5 + P_6 + P_7 = 1$. There is one more constraint on P_5 and P_7, namely $P_5 = P_7$, which is of purely geometrical character (cf. Ref. 5). If P_6 is close to 1 and P_5 and P_7 quite small, we can argue as follows. The presence of a pentagon in a hexagonal lattice creates a defect in the mean angle value, as would happen when there is a *local positive curvature;* the presence of a heptagon creates an excess in the mean angle value, corresponding to a *local negative curvature.* If our lattice has to remain globally flat, there should be as many pentagons as heptagons present; the five defects of pentagon angles will then annihilate the seven smaller excesses of a heptagon. That is why $P_5 = P_7$ is a rigorous equality in flat two dimensions, and we have $P_5 = P_7 = (1 - P_6)/2$. Now our Lagrangian depends on one variable α and on two essential parameters, λ and P_6:

$$L = 3\alpha^2 - 4\pi\alpha + \lambda\{ 0.0362(1-P_6)[4\sin\alpha + \sin(3\pi - 4\alpha)] + 0.0833P_6[5\sin\alpha + \sin(4\pi - 5\alpha)] + 0.0474(1-P_6)[6\sin\alpha + \sin(5\pi - 6\alpha)] \} \ . \tag{12}$$

If $P_6 = 1$, the Lagrangian (11) has an extremum at $\alpha = 2\pi/3$ for *any* value of λ. However, if $P_6 \neq 1$, it is easy to see that when λ grows larger at some critical value λ_c the minimum at $2\pi/3$ gives way to a maximum, whereas other minima can be observed nearby [Figs. 4(a)—4(d)]. The critical value of λ is easily computed,

$$\lambda_c = \frac{6}{0.873 + 1.292P_6} \ . \tag{13}$$

The fact that there is always an extremum at $\alpha = 2\pi/3$ should be expected. As a matter of fact, we could have argued in a different way: Suppose that we have not imposed any constraints on P_5 and P_7, and introduced the Lagrange multipliers λ_5, λ_6, and λ_7 for the corresponding contributions

$$L = 3\alpha^2 - 4\pi\alpha + \lambda_5 P_5 A_5 S_5(\alpha) + \lambda_6 P_6 A_6 S_6(\alpha) + \lambda_7 P_7 A_7 S_7(\alpha) \tag{14}$$

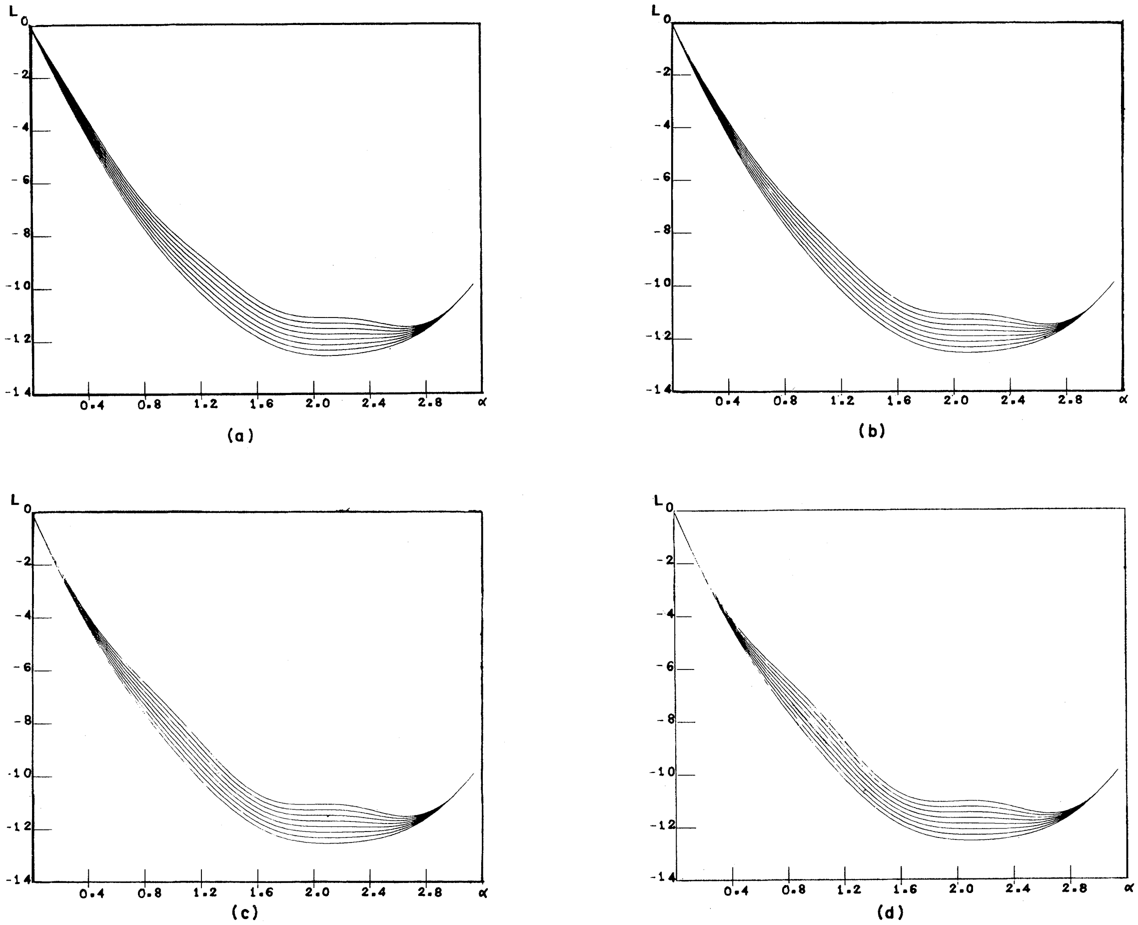

FIG. 4. Lagrangian plotted as function of α. The values of P_6 have been chosen as follows: (a) $P_6=0.25$, (b) $P_6=0.50$, (c) $P_6=0.75$, (d) $P_6=0.90$. On each plot we have traced eight curves corresponding to varying values of the parameter λ: $\lambda=0.5m$, with $m=3,4,5,\ldots,10$. The lowest curve corresponds always to the lowest value of λ. α is given in radians, energy units arbitrary.

with A_k,S_k defined as above. Then require an extremum at $\alpha=2\pi/3$. This yields, after simple calculus, the relation

$$\frac{\lambda_5 P_5}{\lambda_7 P_7}=\frac{1.99}{1.44}\approx 1.382 \approx \frac{7}{5} \tag{15}$$

which coincides up to 1.3% with the ratio of our previous weighting coefficients introduced via the geometrical reasoning. This is quite comforting if we recall the apparent roughness of our approximation.

Let us return to the curves displayed in Fig. 4. The minimum at $2\pi/3$ can be identified with the regular crystalline lattice (6,6,6) when $P_6=1$; for $P_6\neq 1$ we still have a minimum very close to the value $2\pi/3$, which we can interpret as stable amorphous configuration. If λ grows bigger, new minima are observed away from $\alpha=2\pi/3$; these should in principle correspond to new stable amorphous configurations; we think that in our simplified model only the minimum at $\alpha>2\pi/3$ can be taken seriously, because when we mix up pentagons, hexagons and heptagons so that $P_5=P_7$, the geometrical mean value α in an elementary cell will be more than $2\pi/3$. The possibility of having other crystalline states corresponding to these minima can be safely excluded, because no regular lattice displaying a translational symmetry can be produced out of five-, six-, and seven-sided polygons only.

The parameter λ has a well-determined physical meaning, and should vary from one chemical element to another. In principle, it can be evaluated by means of quantum mechanics (calculus of orbitals in elementary cells of the lattice). Let us note that in the case of carbon the formation of polygons occurs very easily; therefore, the energy captured in a polygon is not very high, which means that λ is small. In the case of silicon the formation of "trees" (polymerization tendency) is preferred and the polygons must be more difficult to create, which means that λ is large. According to our model, when λ grows larger, the amorphous state becomes energetically preferable; if we recall that silicon is a good glass former, whereas carbon is not, we have reasons to think that we are on the right track.

In principle, we can go further if in a more realistic model we include the contributions coming from other possible polygons also, appearing with the finite probabilities $P_3, P_4, \ldots, P_{12}$ (we can safely stop at 12). There will be plenty of new information, too. Of course, we shall still have $\sum P_i = 1$, but our generalized Lagrangian should present new minima corresponding to the regular tricoordinated lattices, i.e., we shall impose $\partial L/\partial\alpha = 0$ also at other values of α (given in Table I) and at the values of P_i defined by the regular lattice itself, e.g., $P_4 = \frac{1}{2}$, $P_8 = \frac{1}{2}$, other P_i's vanishing for the lattice (4,8,8), etc. It is quite easy to see that all such identities cannot be satisfied for one common value of λ, because in reality the energy of polygon forming is not just proportional to the surface of the polygon with the same scale constant factor independent of the number of sides; our model is oversimplified. In a more sophisticated version one should introduce λ_n different for each n.

Then, after satisfying all the identities for the known minima, most of the parameters λ_i and P_i can be eliminated; the remaining independent ones can be fixed by comparing the values of L for different lattices. As L describes the energy per atom bound in the lattice, these differences have a precise physical meaning and can be put into correspondence with, for example, the melting points of the given crystalline structures. This in turn can fix the overall scale of the Lagrangian, which is not defined yet.

Let us close by summarizing the features of the model which seem to be the most important ones. In spite of the very rough approximation we have used, it enables us to get the following insights.

(1) The ease or difficulty of glass formation can be expressed by one parameter λ (energy of polygon forming versus energy of a bare tripod). Its magnitude can be evaluated, in principle, for different chemical elements using the methods of atomic physics. We have seen on our curves that for λ above the critical value an infinity of amorphous states exist; glass is an amorphous state, but not every amorphous state has the properties of glass. Our guess is that among all the amorphous states observed as minima on our curves, the glassy state should correspond to the flexion point at which $(\partial L/\partial\alpha) = 0$ and $(\partial^2 L/\partial\alpha^2) = 0$, too. This fixes the amount of P_6 for a given λ in our bidimensional "glass."

(2) The choice of the geometrical mean angle value in an elementary cell enables us to take into account, in a very simple way, the medium-range correlations in the lattice; it enables us also to compare in a continuous manner the crystalline and the amorphous lattices. The extrapolation of the (roughly approximative) linear law of the dependence of D^{-1} on α gives us the density of any amorphous configuration. Then the coefficient of elasticity (at least the contribution coming from pure bond-bending deformations and neglecting the bond stretching) can be calculated as $\partial L/\partial(D^{-1})$.

(3) In a more realistic model the contributions of all possible polygons, i.e., triangles, rhombs, etc., have to be considered, too. It is easy to see that in this case more new minima will appear, because the Lagrangian shall contain more different harmonics. Such a phenomenon is really observed, because a denser glass can be obtained by thermal annealing and internal restructuration without a phase transition in a classical sense.

(4) The fact that the energy dependence on P_6 is very weak in the vicinity of the extremal point [a kind of "plateau" on the graph $L(P_6)$ for α fixed near the critical value] also suggests the possibility of a gentle restructuration, i.e., the quasicontinuous passage from one amorphous state to another, if the temperature is high enough to easen the bonds here and there, but not sufficient for melting the glass completely.

(5) The very small differences between the energy levels of different amorphous states (as compared to the difference in energy between these states and the crystalline state) make possible the existence of tunneling modes in glasses on the quantum level. The possibility of the tunneling in glasses, which might explain the Vogel-Fulcher law at low temperatures, has been discussed by Rivier,[6] Kléman,[7] and other authors, although in a completely different context, which considered the "defects" in glasses and their possible contribution to the energy of the lattice. We see that our formalism can take into account the existence of tunneling modes, giving also some clues to the order of magnitude of the potential barriers.

(6) In order to make this theory operational and to permit the verification by experiment, we have to generalize it to three dimensions. A tetracoordinate lattice should be then considered, and the parametrization of the problem has to undergo some important changes. Not only the geometrical mean value of the angles in the adjacent polygons has to be considered, but also the *dihedral angles* between the adjacent polygons are important, too; the solid angles can be then derived as the function of these two kinds of angles. The density as well as the energy depend on these quantities, and also on the new parameters absent in the two-dimensional case, such as the number of polygons per bond and the number of polygons per atom (which is not a simple function of the coordination number as before).

(7) The generalization for the mixed lattice, with different coordination numbers appearing in the lattice simultaneously, can be obtained quite easily. In the same spirit as before, we should consider the coordination number as a continuous variable (mean value of N_c over the lattice). If we continued Fig. 3 in three dimensions, considering D^{-1} as a function of both α and N_c, our points would most probably stay on some surface in the space (D^{-1}, α, N_c). It would be worthwhile to look at the minima of the modified Lagrangian (in which the term $3\alpha^2$ is replaced by $N_c\alpha^2$) as functions of α, N_c, and P_i's. This should give some insights concerning such glass formers as $\mathrm{Ge}_x\mathrm{Se}_{1-x}$, etc., as has been suggested by Phillips.[8,9]

(8) Until now, we considered P_i's as independent parameters; if so, we are describing the elastic deformations of an amorphous solid (without considering the bond-stretching contributions). However, if both α and P_i's are supposed to vary, then we are in fact describing a structure which behaves rather as a liquid, or something that is happening during the glass transition or crystallization. In such a case, it seems reasonable to assume, as a first approximation, that the spontaneously formed polygons are very close to the perfect (equiangular) form; then a simple relation between D^{-1}, the probabilities P_k and α can be introduced as follows: According to our definition,

$$\alpha = \left[\prod_k \left[\frac{(k-2)\pi}{k} \right]^{kp_k} \right]^{(\sum_k k p_k)^{-1}} \tag{16}$$

or, as $p_k = kP_k / \sum_k kP_k$,

$$\ln\alpha = \frac{1}{\sum_j j^2 P_j} \sum_k k^2 P_k \ln\left[\frac{(k-2)\pi}{k}\right] . \tag{17}$$

As for D^{-1}, we recall that the surface of a perfect k-gon is given by

$$S_k = \frac{k}{4}\tan\left[\frac{(k-2)\pi}{2k}\right] \tag{18}$$

and each atom in the lattice is surrounded by three polygons with the probability of finding a k-gon equal to p_k; therefore

$$D^{-1} = \frac{3}{4}\sum_k \frac{kP_k}{\sum_j P_j}\tan\left[\frac{(k-2)\pi}{2k}\right] . \tag{19}$$

In the simplified model with $P_5 = P_7 = (1-P_6)/2$, other P_i's equal to zero, everything depends then on one parameter α only; e.g., one obtains easily the relation

$$P_6 = \frac{37\ln\alpha - 27.726}{\ln\alpha - 1.1128} . \tag{20}$$

Then we can replace P_6 by this expression in our Lagrangian (12). Let us note that now $\alpha = 2\pi/3$ is the *minimal* value of α, corresponding to $P_6 = 1$; the variation of α is very limited now, e.g., for $P_6 = 0.5$, $\alpha = 120°37'$. The inverse of density is also easily obtained now,

$$D^{-1} = 1.3386 - \frac{1.4622\ln\alpha - 1.0957}{\ln\alpha - 1.1128} . \tag{21}$$

Maximal density is reached for $P_6 = 1$, when $D^{-1} = 1.299$; minimal density corresponds to $P_6 = 0$ when $D^{-1} = 1.3386$ (the variation is of the order of 1.57%).

In order to have an extremum at $\alpha = 2\pi/3$ we have to impose now the condition (14). Similar phenomenon as before can be observed now, too: Below some critical value of the parameter λ we get $(\partial L/\partial\alpha) \geq 0$ for $\alpha \geq 2\pi/3$, and the "crystallization" will be energetically preferable; for λ growing larger, at some point $\partial L/\partial\alpha < 0$ for $\alpha \geq 2/3$, and the system will tend spontaneously to an amorphous configuration corresponding to $P_6 \neq 1$. Closer investigation of these phenomena, together with the eventual analysis of the time dependence $\alpha(t)$ will be the subject of our planned future papers.

Enlightening discussions with Professor M. Kléman, Professor L. Michel, Professor J. Mozrzymas, and Miss Dina M. Lucas Ferreira dos Santos are gratefully acknowledged. The author also expresses his thanks to Dr. Hubert Bonnin de Fraysseix for the computer programming of the curves.

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