

From quasicrystals to icosahedral glass

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In the present work, a model of glass is constructed by relating the order in metallic glasses to that of an ideal structure, the icosahedral quasicrystal. The Landau theory of this model leads to a ground state which is an ordered array of solitons. There are many stationary states corresponding to different soliton networks. It is argued that topological constraints lead to finite barriers separating these states. The system can therefore be effectively frozen into one of these metastable states. It is suggested that the excitations in one of these metastable states are related to the orientational degree of freedom of the order parameter. The model provides an attractive framework for understanding the physics of metallic glasses.

The dominant theme in the theoretical study of metallic glasses has been the understanding of the connection between local icosahedral order and the occurrence of a glassy phase.¹⁻⁴ The recent observation of icosahedral quasicrystals⁵ has raised the obvious question of the relationship between such phases and icosahedral or metallic glass. In fact, the concept of an icosahedral quasicrystal arose in connection with the effort to construct a reference structure for glass.⁶ However, most of the subsequent activity has been concentrated on understanding the nature of the quasicrystalline phase.

The purpose of this paper is to present a model of glass based on a generalization of the density-wave description of quasicrystals. In this picture, the quasicrystal is viewed as a particular three-dimensional (3D) cut of a periodic structure in six dimensions (6D). The extended icosahedral order of the quasicrystal can then be defined in terms of order parameters which correspond to the Fourier transforms of the periodic 6D density. Modeling glass as an imperfect icosahedral quasicrystal, the order in glass can be described by generalizing the above description to define local order parameters. A Landau theory of glass can then be formulated in terms of these local order parameters. At low temperatures, the important degree of freedom is expected to be associated with translations in 6D space. The model free-energy functional then leads to the appearance of planar soliton configurations in the stationary states.

The appearance of solitons is a common feature of models like the Frenkel-Kontorova (FK) model which describe incommensurate structures.⁷ In a one-dimensional FK model, the solitons are the spatial analogs of a classical particle moving in a one-dimensional periodic potential. The motion being one where the particle sits in one potential minimum for a relatively long time and then makes a swift move to a neighboring potential minimum. The description of a one-dimensional quasicrystal as a cut through a two-dimensional lattice leads to a similar model, except that the solitons correspond to paths connecting neighboring minima on a square lattice. There can, therefore, be two types of solitons corresponding to the

two primitive lattice vectors. Similarly, the solitons in the icosahedral crystal correspond to paths on a 6D lattice. The quasicrystal models are, therefore, similar to FK models with more than one set of periodic potentials.

A useful and convenient way of classifying these solitons is by using topological arguments. This classification scheme can then be used to analyze the nature of the various stationary states which correspond to different arrangements of the solitons. It can be shown that, in icosahedral media, there can be many topologically distinct soliton networks. It can also be argued that these networks are separated from each other by finite energy barriers. This provides an appealing picture of glass; each distinct network being thought of as one of the metastable states that the system can freeze into.

A complete description of the model requires the formulation of a free-energy functional which includes both the amplitude variations and the orientational degrees of freedom of the order parameter. The construction of such a generalized functional is briefly discussed in this work. A gauge-invariance property, arising from the embedding in 6D space, leads to an interesting structure of the functional. This functional is expected to become relevant when defects are present in the system.

Most of the existing work on metallic glasses has been based on the notion that icosahedral glass can be viewed as a mapping into flat space of a perfect icosahedral tiling in curve space.¹⁻⁴ Mappings from spaces of both positive and negative curvature have been considered.¹ The mapping that has received most attention is that of a 12-coordinated lattice of icosahedra on the surface of a four-dimensional sphere. Mapping from curved to flat space can be accomplished only by the introduction of defects like disclination lines which decurve space. These defects are, therefore, an essential ingredient of the curved-space description of glasses.¹⁻⁴ A Landau description of this model of glass is suggestive of a ground state consisting of an ordered array of disclination lines.² It has been argued that strong topological constraints render this ground state inaccessible to a rapidly cooled system like metallic glass, and glass is there-

fore characterized by a disordered network of disclination lines.^{2,3}

The present model is similar to the curved space description of glasses in describing the ground state by an ordered array of defects. The defects here are the solitons, in contrast to line defects of the curved space model. It will, however, be shown later that the two are related closely and that most networks of solitons are also forced to have line defects. Metastable states exist in the present model as a consequence of topological barriers which separate states with different soliton patterns. This is reminiscent of the line-defect entanglement phenomenon.^{2,3}

The starting point of the Landau theory is the density-wave description of quasicrystals.⁸⁻¹⁰ The quasicrystal is described by the density

$$\rho(\mathbf{r}) = \sum_{\{\mathbf{g}\}} \rho_{\mathbf{g}} \exp(i\mathbf{g} \cdot \mathbf{r}),$$

$$\mathbf{g} = \sum_i m_i \mathbf{q}_i. \quad (1)$$

The set $\{\mathbf{q}_i\}$ is the incommensurate set made up of the six vertex vectors of an icosahedron. They are linearly independent on the space of integers: $\sum_i m_i \mathbf{q}_i \neq \mathbf{0}$ for any set of integers $\{m_i\}$. The density function $\rho(\mathbf{r})$ is a cut through a function which is periodic in 6D space, and the 3D cut is defined by the three 6D vectors⁸⁻¹⁰ $\vec{Q}_\alpha = (q_{1\alpha}, \dots, q_{6\alpha})$, $\alpha \equiv x, y, z$. The set $\{\rho_{\mathbf{g}}\}$ figures as the set of order parameters in the Landau theory of freezing from a liquid into an icosahedral quasicrystal (IQ).⁸⁻¹⁰ The minimal set which can describe the symmetry of the IQ is $\{\rho_i\}$ corresponding to the set of vectors $\{\mathbf{q}_i\}$.

Viewing glass as an imperfect IQ, the order in glass can be described by the local order parameters $\{\rho_i(\mathbf{r})\}$. In order to be able to identify the important degrees of freedom of the order parameter, it is convenient to think in terms of the 6D periodic structure. This periodic density can be written as

$$\rho(\vec{\mathbf{R}}) = \sum_{\{\vec{\mathbf{G}}\}} \rho_{\vec{\mathbf{G}}} \exp(i\vec{\mathbf{G}} \cdot \vec{\mathbf{R}}). \quad (2)$$

Here, $\{\vec{\mathbf{G}}\}$ is the set of 6D reciprocal lattice vectors. Comparing Eq. (2) to Eq. (1), it is clear that $\{\rho_i\}$ can be identified with $\{\rho_{\vec{\mathbf{G}}}\}$ corresponding to the six smallest 6D reciprocal lattice vectors.

The free energy that one would like to construct from the $\rho_{\vec{\mathbf{G}}}(\mathbf{r})$ should be invariant under all translations and a subset of rotations in 6D space. The subset of rotations is defined by the requirement that they leave the orientation of the 3D cut with respect to the 6D lattice invariant. The latter restriction applies because changing the orientation of the cut leads to a completely different physical system whose free energy can be very different. The important degrees of freedom are therefore the order-parameter amplitude and the order-parameter phases corresponding to 6D translations and rotations. At low temperatures, the amplitude variation can be neglected and the orientational degree of freedom gets locked to the translational freedom.² An adequate description of the

low-temperature properties can then be given by a set of slowly varying displacement variables, $\vec{\mathbf{U}}(\mathbf{r})$. The order parameters can be written as

$$\rho_{\vec{\mathbf{G}}}(\mathbf{r}) = \rho_{\vec{\mathbf{G}}}^0 \exp[i\vec{\mathbf{G}} \cdot \vec{\mathbf{U}}(\mathbf{r})],$$

$$\rho_i(\mathbf{r}) = \rho_0 \exp[iU_i(\mathbf{r})]. \quad (3)$$

Under these conditions, the free-energy functional becomes identical to the one which has been used to analyze the local stability of the IQ.^{11,12} Measured with respect to the energy of the perfect IQ, this functional is

$$f^0 = \sum_i |\nabla U_i|^2 - \sum_M A_M \rho_0^M \cos \left[\sum_i m_i (\mathbf{q}_i \cdot \mathbf{r} + U_i) \right]$$

$$= |\nabla \vec{\mathbf{U}}|^2 - \sum_{\{\vec{\mathbf{G}}\}} A_M \rho_0^M \cos \left[\vec{\mathbf{G}} \cdot \left[\sum_\alpha \vec{Q}_\alpha r_\alpha \right] + \vec{\mathbf{G}} \cdot \vec{\mathbf{U}} \right]. \quad (4)$$

The sum of the moduli of the components of $\vec{\mathbf{G}}$ defines M , the order of a local term.

This free-energy density describes a frustrated system. The frustration arises from the competition between the gradient terms favoring a uniform $\vec{\mathbf{U}}$ and the local terms favoring a spatially varying $\vec{\mathbf{U}}$. The frustration can be relieved only by the introduction of defects. In order to analyze the nature of these defects, it is helpful to define a new displacement vector

$$\vec{\mathbf{W}}(\mathbf{r}) = \vec{\mathbf{U}}(\mathbf{r}) + \sum_\alpha \vec{Q}_\alpha r_\alpha.$$

It is seen from Eq. (4) that the $\vec{\mathbf{W}}$ field feels a periodic potential defined on a lattice given by the vectors $\vec{\mathbf{G}}$

$$f^0 = \left| \left[\nabla \vec{\mathbf{W}} - \sum_\alpha \vec{Q}_\alpha e_\alpha \right] \right|^2 - \sum_{\{\vec{\mathbf{G}}\}} V_{\vec{\mathbf{G}}} \cos(\vec{\mathbf{G}} \cdot \vec{\mathbf{W}}). \quad (5)$$

Here e_α denotes a unit vector along the direction α . The stationary states of the system can be obtained by functional minimization of the above free-energy density. The appearance of solitons is a general feature of all such model free-energy functionals,⁷ and can be understood in terms of an analogy with particle motion in a periodic potential.¹³ The analogy is not exact because here one is dealing with three spatial dimensions as opposed to one time dimension in the particle problem, and therefore can be used only as a quantitative tool. The model of a 1D quasicrystal would map onto the particle problem, and the local minima of f^0 would correspond to paths which satisfy Newton's equation for a particle moving on a square lattice. There are many such minima, each corresponding to an event in which the particle sits at one lattice site for a long time and then swiftly moves into a neighboring lattice site. In the 3D problem, the spatial configurations of the $\vec{\mathbf{W}}$ field which minimize f^0 are planar solitons. These are singularity free configurations which are subject to the constraint that sufficiently far away from a given plane, the field is uniform and corresponds to a minimum of the potential given in Eq. (5).

It is to be noted that the vectors \vec{Q}_α do not appear in the equations of motion.⁷ The stationary states of the system are therefore made up of a superposition of solitons. The ground state being given by that particular ar-

rangement of solitons which minimizes the total free energy. It is not possible, in general, to obtain explicit solutions for the stationary states. An explicit solution has been worked out in Ref. 12 assuming that only one set of local terms, with a given value of M is important. This analysis shows that the ground state is a network of solitons which has quasicrystalline icosahedral order. The free-energy functional has other stationary states which are characterized by soliton patterns differing from the ground state pattern. In order to understand the physics of this model of glass, it is therefore necessary to analyze the properties of these solitons. The most convenient framework for this analysis is the topological theory of defects.¹⁴ Although, a more physically intuitive treatment is desirable, the nature of the icosahedral phase makes this prohibitively difficult. The topological analysis serves as an efficient tool for gaining some insight into the properties of the model, and provides a starting point for more detailed studies.

Topological defects or textures such as planar solitons have a one-to-one correspondence with spatial variation of the order parameter and can be described by mappings of the relevant physical space into the order-parameter space.^{14,15} Given a free energy, its symmetry group G , acting on a prototype order parameter, generates the complete set of order parameters. Symmetry breaking implies that a subgroup H of G leaves the order parameter invariant, and the order-parameter space is identified with the coset space G/H . Definition of this coset space for the IQ requires identification of the symmetry group of f^0 and the group of invariance of the density function $\rho(\mathbf{r})$. An analogous procedure based on the projection technique¹⁶⁻¹⁸ has been employed for classifying line defects in IQ.¹⁹ In the absence of phase-locking terms in the free energy, the two procedures are equivalent.

The free energy, in the absence of phase-locking terms, is invariant under all translations in 6D space (R^6) and under those $SO(6)$ rotations which leave the orientation of the 3D cut with respect to the 6D lattice invariant. The phase-locking terms are important in the region where the phases are uniform, and can be neglected in the regions close to the planes of the solitons where the gradient term dominates, and the phases vary rapidly. In these regions of nonuniformity, the symmetry group G is the group $\{SO(3) \otimes SO(3)^{\perp}\} \wedge R^6$. The group G consists of all 6D translations and those rotations which can be written as a product of two $SO(3)$ rotations, one in the 3D space defined by $\{\bar{Q}_\alpha\}$, and the other in the 3D space defined by the orthogonal vectors $\{\bar{Q}_\alpha^\perp\}$.¹⁹ The subgroup H of G which leaves $\rho(\mathbf{r})$ invariant, consists of discrete translations and operations of the icosahedral group $A(5)$.^{9,20} In the present work, effects related to the breaking of translational symmetry will not be considered. It is expected that the inclusion of these effects would enrich the present model but would not alter its essential features. Consequently, only the rotational parts of the groups will be retained. The order-parameter space in the region close to the planes of the solitons is, therefore,

$$R \equiv G/H = \frac{SO(3) \otimes SO(3)^\perp}{A(5)}. \quad (6)$$

The phase-locking terms dominate sufficiently far away from the regions of nonuniformity. These terms are invariant under the symmetry group which leaves the lattice vectors $\bar{\mathbf{m}}$ invariant; the 6D hyperoctahedral group. The subgroup of the hyperoctahedral group included in G is the icosahedral group $A(5)$.^{19,20} The action of this group on $\rho(\mathbf{r})$ leaves it invariant. In the uniform regions, the order-parameter space is therefore restricted to a single point in R , and the solitons are characterized by the constraint that the order parameter approaches a single value far away from a given plane. Thus, a line drawn between the two uniform regions of a soliton determines a closed loop in the order-parameter space. The solitons can therefore be classified by the homotopy classes of loops in order-parameter space, and are characterized by the elements of the fundamental group of the order-parameter space $\Pi_1(R)$.^{14,15} Since the group H is discrete, Π_1 is isomorphic to the lift of H into the cover group of G . The cover group of $SO(3)$ is $SU(2)$ and, therefore

$$\Pi_1(G/H) = \bar{A}(5). \quad (7)$$

Here, $\bar{A}(5)$ is the lift of the icosahedral group into $SU(2)$. The same group describes disclination line defects in icosahedral media and has been the subject of extensive investigation.² Because of this relationship between line defects and solitons, the solitons can end in these line defects¹⁵ and also be generated by the motion of these line defects.¹⁴

The different classes of Π_1 correspond to different classes of $SU(2)$ rotations. A soliton introduces a shift in the cut through the higher dimensional space without changing the relative orientation of the cut with respect to the lattice. The classes of solitons correspond to the distinct ways in which this can be accomplished. In the physical space, a soliton leads to local changes in the arrangements of the icosahedral building blocks. This can also be visualized as a change in the relative phases of the six density waves describing the icosahedral structure. The analogy with particle motion on a lattice can be used as a tool for visualizing the topological classification scheme. A particle at a given lattice site can jump to any of the neighboring sites. These sites are related by different symmetry operations of the point group to the original site and the possible paths from this site can therefore be labeled by the possible symmetry operations. Identifying the solitons with paths connecting two neighboring points on a lattice with icosahedral point group symmetry, in the 6D space, leads to a classification scheme which is identical to the one in terms of the elements of Π_1 .

The group Π_1 is nonabelian; the individual group elements do not commute. The properties of media with nonabelian Π_1 are quite different from those of abelian media¹⁴ and this has important implications for the physics of glass.² The properties relevant to the present work are summarized later.

The class multiplication table usually fails to provide a unique product class. This implies, for example, that the product of the combination of two line defects depends on the path followed in bringing them together.¹⁴ In

connection to soliton networks, this implies that a given set of solitons can give rise to soliton networks belonging to different topological classes. This follows from the observation that a soliton network can be characterized by the sequence of steps followed in its construction. Each step in the sequence being given by the product of the classes characterizing the soliton being added, and that of the existing network. Since the different networks belong to different topological classes, they cannot be continuously deformed into each other. Again, drawing upon the rough analogy with particle motion, a given soliton configuration can be identified with a path on the lattice going through a particular sequence of lattice points. Any two paths which go through exactly the same sequence of lattice points can be deformed into each other since the paths are pinned only at the lattice sites. But, because of the discreteness of the lattice, paths going through different sequences cannot be continuously deformed into each other.

The crossing of two solitons, characterized by noncommuting elements of Π_1 , necessarily give rise to a line defect at their line of intersection. This can be seen by drawing a contour surrounding this line. The element of Π_1 characterizing the line would be given by $\alpha\beta\alpha^{-1}\beta^{-1}$, where α and β are the elements characterizing the individual solitons. This belongs to the identity class only if α and β commute. This implies that most soliton networks will also contain a network of line defects. These line defects are disclination lines in the sense that the 6D vector \vec{W} , associated with each point in 3D space, does not return to its original value on going around a closed loop around the line defect. This observation establishes a closer correspondence between the present model and the curved space model of glass.^{2,3}

The possibility of constructing many distinct topological networks from a given set of solitons is indicative of the occurrence of metastable states in the system. These states being separated by barriers arising from topological constraints. The height of such barriers can be deduced only from a detailed study of the energetics involved in creating and destroying solitons. A detailed analysis of the energetics has not been carried out so far. However, such a study has been carried out for planar solitons in He³, and it has been shown that the barrier for destroying a soliton which can end at a line defect is related to the energy necessary for creating a closed singular line with ring-radius comparable to the soliton width.¹⁵ Since solitons in icosahedral media can also end at line defects, a similar result is expected to hold here, implying that the topological barriers are not infinite.

It would be interesting to see if the barriers separating form a hierarchical structure. Classification of the states by the sequence of steps on a lattice does suggest such a structure. Two paths of N steps which differ only in the way the N th step was added can be transformed into each other by an activation process involving the creation and destruction of one step (soliton). Similarly, if the paths branched off at the stage of adding the $(N-1)$ th step, two activation processes would be required to transform them into each other. The various N -soliton states can therefore be grouped according to the number of activa-

tion processes separating them. This defines a distance between the states, and these distances form an ultrametric space.²¹ Defining a triangle by specifying its vertices to three possible N -soliton states, it can be seen that its sides, which correspond to the number of activation processes separating the vertices, have to be either all equal or two of them have to be equal and greater than the third.

A hierarchical structure of the space of states would have important consequences for the relaxation process in glasses. As discussed in Ref. 21, random walk in an ultrametric space leads to anomalous relaxation laws. In the present model, the distance between states is related to the number of activation processes, and therefore appears in the entropy contribution to the free energy barrier separating the states. In the absence of any other distance-dependent contribution, the barrier is therefore a logarithmic function of the distance. If the space is ultrametric, then it has been shown that random walk in such a space leads to the stretched exponential law for relaxations.²¹

The free-energy functional, discussed so far, is frustrated, and has been shown to lead to the appearance of defects in the stationary states. Under these circumstances, it is not clear whether the original assumption of the locking of orientational degree of freedom to the translational degree of freedom is valid. The locking follows from the minimization of a generalized functional including both orientational and translational degrees of freedom.^{22,23} To include the orientational order, a field of 6D rotation matrices have to be introduced, the free-energy density has to be generalized to include terms involving the gradients of the rotation matrices and has to be made invariant under the following transformations:

$$\begin{aligned}\hat{\Omega}(\mathbf{r}) &\rightarrow \hat{\Omega}_0 \hat{\Omega}(\mathbf{r}) \hat{\Omega}_0^{-1}, \\ \rho_{\vec{G}} &\rightarrow \rho_{\vec{G}} \exp[i\vec{G} \cdot (\hat{\Omega}_0 \vec{R} - \vec{R})].\end{aligned}\quad (8)$$

Here the rotation matrix $\hat{\Omega}_0$ has to belong to the subset of 6D rotations which leave the orientation of the lattice with respect to the cut invariant. The requirement of rotational invariance leads to a gradient term of the form

$$\left| \left[\nabla U_i - i \left[\hat{\Omega} \sum_{\alpha} \vec{Q}_{\alpha} \hat{\mathbf{e}}_{\alpha} \right]_i \right] \right|^2.$$

Minimizing this term with respect to $\hat{\Omega}$ gives a relationship between $\hat{\Omega}$ and the gradient of \vec{U} , and for a slowly varying field \vec{U} , justifies the neglect of the $\nabla \hat{\Omega}$ term. This leads to the locking of the orientational order to the translational order. In the presence of solitons this procedure is no longer justified, since there are regions of space where \vec{U} varies rapidly and $\nabla \hat{\Omega}$ is not small compared to

$$\left[\nabla U_i - i \left[\hat{\Omega} \sum_{\alpha} \vec{Q}_{\alpha} \mathbf{e}_{\alpha} \right]_i \right].$$

The system can therefore be adequately described only by treating the translational and orientational order independently.

In constructing this generalized free-energy functional, it is noticed that the global invariance implied by Eq. (8)

has to be generalized to a local gauge invariance property. This is due to the fact that the 3D system is defined by a particular cut of the 6D system. It is seen from the second equation of Eq. (8) that it is possible, by a simultaneous rotation of $\bar{\mathbf{R}}$ and the cut, to keep the projected density unchanged. This is understandable since the operation corresponds to the choice of an arbitrary frame of reference. This implies that the free-energy density has to be invariant under local transformations $\hat{\Omega}_0(\mathbf{r})$. The gauge fields which have to be introduced to enforce this correspond physically to rotation matrices defining the orientation of the 3D cut.

The construction of the complete free-energy functional and the statistical mechanics of the model will be the subject of a future publication. In this work we describe some of the general features of this model free-energy functional. The soliton configurations and the accompanying line-defect configurations are expected to be frozen for long time scales because of the topological barriers separating different configurations. Under these circumstances, the free-energy density can be assumed to have been minimized with respect to the field $\bar{\mathbf{U}}$, and the relevant part of the free-energy functional involves only the rotation matrices $\hat{\Omega}(\mathbf{r})$ and the gauge fields $\hat{A}_\alpha(\mathbf{r})$, $\alpha=x,y,z$. Over time scales short compared to the transition times involved in going from one soliton network to another, the behavior of the system is then expected to be defined by these rotational matrix fields. The partition function can be written as

$$Z = \int \mathcal{D}\hat{\Omega} \mathcal{D}\hat{A}_\alpha \left[\exp \left[-\beta \int d\mathbf{r} f(\mathbf{r}) \right] \right], \quad (9)$$

where f is the complete free-energy functional. The free energy of the system would be obtained by calculating the free energy corresponding to a fixed soliton network from the above partition function, and then averaging over all

such networks.

The ground-state configuration of the rotation fields can be obtained by minimizing the free-energy functional. If the variation in the amplitude of the order parameter can be neglected outside the immediate vicinity of line defects, and if the line-defect density is not too high, then one could think of defining a space over which the order-parameter amplitude is a constant. Such a space has punctures in it at the locations of the line defects. By imposing appropriate boundary conditions, one can then minimize the free-energy functional to obtain the stationary state configurations of $\hat{\Omega}$ and \hat{A}_α . It has been argued in Ref. 24 that the punctured space combined with the local gauge invariance of the system leads to the occurrence of two-level systems or tunneling states. It would be interesting to search for such states in the present free-energy functional.

In conclusion, a model of glass has been developed based on the concept of icosahedral glass being an imperfect icosahedral quasicrystal. The Landau theory has been shown to lead to stationary states which have defects in them. The topological characteristics of these defects have been used to deduce some properties of the model. There is scope for much further development of the model. For instance, the free-energy functional can be used to construct time-dependent Ginzburg Landau equations for describing the dynamics of the model. The nonlinearities of the functional are expected to lead to slow relaxation processes. This work is in a preliminary stage of development.

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