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## STATISTICAL TOPOLOGY OF GLASSES

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**Abstract** The structure and properties of glasses are investigated from a topological viewpoint (the only geometry surviving the absence of metric and the triviality of the space group), with two main results: The overall homogeneity is a gauge symmetry and the only structurally stable constituents (in the elastic continuum limit) are odd lines or  $2\pi$ -disclinations. The universal properties of glasses are immediate consequences of these results. Their structure corresponds to statistical equilibrium, i.e. to the most probable distribution of their constituents. Odd lines take up a semi-dilute distribution, and ideal space-filling cellular structures obey Lewis's law.

### Topological structure of glasses.

To gain a general, if superficial understanding of glasses, one needs to answer three questions: 1) What are the structural constituents of glasses? 2) How are these constituents arranged together? (structure of glasses and their symmetry) 3) What is the influence of the constituents on those physical properties which are universal and specific to the amorphous state (Vogel-Fulcher relaxation at high temperatures, anomalous excitations at low temperatures associated with tunneling modes /1/).

The structure of glass is represented by a network, continuous random network for covalent glasses, Voronoi froth for metallic glasses. In fact, amorphous materials, but also biological tissues, metallurgical aggregates, soap bubbles froths, geological jointings, are all random cellular structures which fill space. At a glance, one distinguishes their three main structural features: i) Non-collinearity of local reference frames, or variety of cell shapes. ii) Overall homogeneity. iii) Odd lines.

These three features are characteristic of the amorphous state, and can be taken as its structural definition. (If either i) or iii) are missing, one may still have a disordered structure, but without topological disorder). They can be formulated in mathematical terms:

Homogeneity as gauge invariance /2,3,4/ Homogeneity means that atoms are distinct, but not distinguishable. However, unlike in crystals, this homogeneity is not a generative symmetry, but the fact that any objective (physical) statement about one particular atom can also be made about any other, even though their environments (tetrapods attached to the Si atoms in a-Si or vitreous silica) are manifestly different and non-collinear. This is the kind of homogeneity experienced by getting lost in a forest. The automorphisms probing homogeneity are permutations of the atoms and their surroundings, effected by local rotations of the tetrapods. The physical properties are unchanged under local rotations, as long as connections with the rest of the system are modified covariantly, and homogeneity is a genuine, gauge symmetry.

The need for topology and its consequences Manifestly, amorphous structures have neither metric, nor unique, global reference frame orientation. The only invariant under automorphisms probing their overall homogeneity is their connectivity, and the relevant geometry is topology or rubber geometry, which replaces the metric geometry of ordinary crystallography. By the same token, the 232 (metric) space groups are replaced by homotopy groups which describe connectivity. k states and Bloch theorem give way to topological sectors, and to a theorem, curiously also formulated by Bloch for superconductors /5/, stating that the free energy is a periodic function of the flux triggering the gauge transformation /4/. The period corresponds to a large (non-trivial) gauge transformation. Consequences of this enforced retreat from metric to topology are that: a) Structural constituents cannot be distinguished by their sizes. b) A constituent exists, and is defined only if it is structurally stable, that is if it cannot be made to disappear under small deformations. c) Its only distinctive feature is its shape. Hence randomness, or topological disorder implies that cells have many different shapes. Glasses cannot be made of one single type of cell, as a result of "random avoidance of the niceties of adjustment" (i.e. 4 cells meeting along a mathematical line, or 5 or more at a mathematical point) (F.T.Lewis 1943). The problem of describing the structure is a statistical one, and statistical mechanics yields average features of the ideal cellular structure (Lewis's law).

Odd lines /6/ The presence of five- and seven-member rings in the structure is symptomatic of its non-crystallinity. They are incompatible with simultaneous translation and rotation (generative) symmetries. The surprising fact is that they cannot appear in isolation, but form uninterrupted lines which either close as loops or terminate at the surface of the material. This is a topological conservation law valid for any network. Besides the cells themselves, odd lines are therefore the structurally stable constituents of the amorphous state for which we were looking.

Odd lines as sole structural constituents of amorphous elastic continua /7,8/ For some amorphous structures, e.g. polymer glasses, it is difficult or too complicated to construct a network. The simplest, and least specific description of any glass is as an elastic continuum whose symmetry is completely broken. Indeed, glasses do ring, and support phonons as long wavelength excitations. The only structural constituent surviving the transition from cellular network (simplicial decomposition of space) to continuum is the odd line, which takes then the form of a  $2\pi$ -disclination. This is because a local configuration must be returned to the same orientation after circumnavigation in space, during which it follows the orientation of the local environment (parallel transport), because the configuration determines the physical properties (strain, etc) of the system. In crystals, it is sufficient that the configuration returns to any orientation related to the original one by an element of the space group: Burgers' vectors of dislocations and disclinations are elements of the space group of the crystals. In glasses, the space group is trivial, yet there are still two possible transformations of the local configuration upon circumnavigation: A rotation by  $4\pi$  is homotopic (continuously deformable) to the identity, but rotation by  $2\pi$ entangles connections of the local configuration with the rest of the system. Thus, one remains with one single structurally stable constituent, a  $2\pi$ -disclination. It is universal and specific to the amorphous state. In terms of homotopy groups /9/,  $\pi_1(SO3)=\mathbb{Z}_2$ . Translations are homotopically trivial, so that dislocations of any kind are not structurally stable in amorphous elastic continua. Neither are point defects ( $\pi_2(SO3)=1$ ).

Note that the space in which the elastic matter is put, is punctured by the cores of the  $2\pi$ -disclinations. Odd lines have been promoted from a passive, geometrical role (odd rings) to a dynamical part as sources of non-collinearity. The mathematical proof of this identity between geometrical and dynamical approaches uses exterior differential calculus and de Rham's theorem

/4/. It is rigourous only for abelian gauge theories, as far as I know.

Odd lines and physical properties of glasses.

Constituents of glasses have now been reduced to one type only, the odd lines. We can now discuss their distribution in the glass, and their influence on its physical properties. It is elementary to calculate the entropy of loops of arbitrary sizes /10,11/. There are three possible types of loops distributions: dilute, semi-dilute and melt. The semi-dilute distribution maximizes the entropy and is the most probable one (it combines high density of odd lines with high entropy of mixing)/7,11/. It is characterized by one single length scale, the distance  $\zeta$  between two nearest but non-adjacent points on any odd lines. (In metallic glasses, the density of odd lines is high, but their number and mobility at high temperatures is restricted by the structural stability of the packing /12/).

Explicit theories of glass (free energy) have been proposed, which have the proper (S03) gauge invariance and contain the correct structural ingredients (odd lines). They are either a model, minimally coupled, S03 gauge theory /3/ of the type first suggested by Yang and Mills in a different context /13/, or simply the classical continuum theory of elasticity, allowing for disclinations /7,14/. Both are nonlinear, but can be linearized for a semi-dilute distribution of odd lines where they are identical. One has then a theory of glass with one single length scale  $\zeta$  and one energy scale  $kT_0$ , to be discussed below. This enables us to correlate low and high temperatures properties of glasses, and their behaviour under external forces.

Viscosity of supercooled liquids /10,11/ Odd lines can move as the structure is deformed. They expand or shrink, but remain uninterrupted (assuming that bonds are broken and reconstituted over a much shorter time scale than that associated with the fluidity). Viscosity, or any structural relaxation rate, is associated with diffusion of some "defect", in the crude but classic model of Glarum and Philips et al./1/. The "defect" must be of sufficient generality to account for the universality of the relaxation process and of sufficient stability to avoid disintegrating during diffusion. Odd lines, as the only structurally stable ingredients of all glasses, clearly fill the bill. They also yield the Vogel-Fulcher-WLF... law  $\eta = A \exp(B/(T-T_0))$ .

Glass transition /10,11/ There is a genuine phase transition at  $T_0$ , corresponding to freezing of the odd lines. A mean field calculation yields a density of free odd lines  $\rho_{\text{free}}(T) \propto 1/\eta$ , and a condensation similar to a Kosterlitz-Thouless transition, but in 3D.

Elastic energy of glass /7,14/ In crystals, disclinations have a prohibitively high strain energy (which increases with distance). In glasses, this energy is screened by elastic fluctuations, including dislocations which are not topologically stable and can be regarded as a thermal bath of local incompatibility fluctuations of average energy  $kT_o$ , the only energy scale in the glass. (This bath corresponds to the Somigliana dislocations introduced by Escaig /15/ in this context. Note that the energy scale is much lower than the bare shear strain energy of disclinations). The strain energy between two odd lines in glasses decreases accordingly as the inverse of their distance like the mutual inductance of two current loops. This result depends on a semi-dilute loop distribution which enables us to linearize the theory.

Tunneling modes /3,4/ At low temperatures, odd lines are frozen. At any point  $\underline{x}$ , a ground state configuration can be labelled  $|0\rangle$  or  $|2\pi\rangle$ , according to whether it rotates by  $0 \sim 4\pi$  or  $2\pi$  when circumnavigating the nearest odd line. But a large gauge transformation at  $\underline{x}$  (rotation by  $2\pi$  at  $\underline{x}$ ) changes  $|0\rangle$  into  $|2\pi\rangle$  and vice-versa. Thus, neither are gauge invariant and qualify as energy eigenstates. The true, gauge invariant, low-energy states are the linear combinations  $|\pm\rangle = (1/2)(|0\rangle \pm |2\pi\rangle)$ , 2 per odd line. This constitutes a microscopic model for the tunneling states in glasses (cf./1/)

The energy splitting  $\Delta_o$  between the two levels  $|\pm\rangle$  is given by the tunneling rate of a "flux" of rotation  $2\pi$ , from the core of the odd line to a distance  $\zeta$  where memory of the configuration has been screened out. One obtains /4/  $\Delta_o = (kT_o/\pi) \exp(-D\zeta \ln \zeta/a)$ , where  $a$  is the interatomic spacing and  $D = 8\pi kT_o / (\hbar c \sqrt{a/2\gamma})$ . A distribution of loop sizes gives rise to a flat density  $1/kT_o$  of energy splitting with width  $kT_o$  in rough agreement with experiments. Tunneling is not utterly negligible, by contrast with superconductors, because odd lines are loops of finite size whereas superconducting vortices are usually straight and traverse the sample (length  $L$ ), and because in glasses, the length associated with fluctuations of the gauge field  $l$  is  $\gg \zeta$ , whereas the penetration depth  $l \ll \zeta$  in superconductors (where the exponent in the tunneling rate is  $2DL\zeta/l$ ).

Hall effect The cores of odd lines puncture the space available, and act therefore as a 2 slit setup for electron wave packets. The packet splits and interferes with itself, and the interference is modified by an external magnetic field. This setup, specific and universal in amorphous solids, should be responsible for the anomalous Hall effect in amorphous semiconductors ("two orders of magnitude less than might be expected..., almost invariably it has

the wrong sign"/16/).

The ideal space-filling random cellular structure /17,11,18/.

Assume that a random space-filling structure is in statistical equilibrium, i.e. that it corresponds to the most probable distribution of cell shapes. We have argued that topological disorder imposes the presence of a considerable diversity of cell shapes. In 2D, the shape of a cell is parametrized by  $n$ , the number of its sides, and the probability distribution ( $p_n$ ) must satisfy the three, constraints of normalization ( $\sum p_n = 1$ ), space-filling ( $\sum A_n p_n = A_o / F$  ( $A_n$ =average area of a  $n$ -sided cell,  $A_o$ =total area available for  $F$  cells)) and topology ( $\sum (6-n)p_n = 0$ ). There may be other constraints of physical origin which may lead to non-universal features of the structure if they are relevant.

The universal, most probable distribution is obtained whenever the above constraints, a system of linear, inhomogeneous relations between  $p_n$ , are linearly dependant. (Clearly, the dimensionality of the space of solutions is larger if that of the space of constraints is smaller, and the solution is, accordingly, more probable). Thus,  $A_n = (A_o / F) \lambda^{n-6+1} / \lambda$ , a relation discovered empirically by the botanist F.T.Lewis /19/, which is obeyed by random mosaics of various origins. Lewis's law is a criterion for the ideality of the structure, a trivial mathematical consequence of the representation of a concrete structure by topological polygons. It can, of course, be generalized to 3D. It has also general applicability (sections of 3D aggregates, though 2D mosaics, should not obey Lewis's law because the space-filling constraint is 3-dimensional).

If the tissue evolves sufficiently slowly for statistical equilibrium to be obeyed at all times, differentiation of Lewis's law yields the average rate of growth of a  $n$ -sided cell, a topological formula (this rate is independant of the size of the cell) first derived for 2D soap bubble froths by von Neumann /20/ from metric geometry and specific physical assumptions. The present derivation /18/ is topological, it holds for mosaics other than soap bubble froths, and, like Lewis's law, it is generalizable to 3D. One then finds that  $\lambda$  is an ageing parameter: it increases monotonically with time.

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