Machine Learning aids Human Intelligence to understand glasses

1. **Finding two-level systems in glasses through machine learning**
   Authors: Simone Ciarella, Dmytro Khomenko, Ludovic Berthier, Felix C. Mocanu, David R. Reichman, Camille Scalliet, and Francesco Zamponi
   ArXiv:2212.05582

2. **Depletion of two-Level systems in ultrastable computer-generated glasses**
   Authors: Dmytro Khomenko , Camille Scalliet , Ludovic Berthier, David R. Reichman, and Francesco Zamponi
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Recommended with a Commentary by Chandra Varma, University of California

To assure oneself of the utility of machine learning appropriately used, as I needed to do, one must compare the first paper above, which uses machine learning, with the second. The same program for the investigation of the much investigated two level states (TLS) in glasses by advanced numerical methods is pursued in both, but the first takes about 10^{-5} lesser computer time than the second. So the statistics are vastly improved and hope exists for answering the even more mysterious issues about the glassy state which remain to be understood, if the right questions are asked and the machine is trained to answer them. The physics already discovered in these papers is also very interesting, although long anticipated, and some weak ideas about the nature of TLS states have been laid to rest.

In 1971 Zeller and Pohl [1] discovered that all glasses have a linear in T heat capacity at low temperatures besides Debye's T^3 required in all solids. Also that they have a thermal conductivity which is proportional at low temperatures to T^2 rather than the expected T^3 due to a finite mean-free path for heat carried by phonons due to impurities. Almost immediately it was proposed [2,3] that the metastability in glass configurations must allow local re-arrangements which may be described by two level systems with very weak interactions between them. A two level quantum-mechanical system may be represented by a Hamiltonian,

$$\Delta\sigma_z + t\sigma_x.$$

Δ is the energy difference of the two configurations and t is the tunneling matrix element between them. Further assume that the probability distribution of Δ is uniform on both sides of $\Delta = 0$ for at least small values, as might

be expected due to local metastability, and that the probability distribution of t is proportional to $1/t$. This happens for $t = \omega_0 e^{-\lambda}$; the distribution of ω_0 is unimportant compared to that of λ, the dimensionless integral over the most probable WKB path between the two configurations and $P(\lambda)$ is also assumed uniform, and cut-off at its upper limit by the measurement time of experiments. Then the density of energy differences $E = \sqrt{\Delta^2 + t^2}$ is uniform near $E = 0$ and the linear in T heat capacity follows. If one considers resonant transfer of energy between the phonons and the TLS, their mean-free path is $\propto 1/T$ and the thermal conductivity follows. The coupling to phonons has to be very small indeed or else the TLS would dissolve into them, and also to minimize the coupling between different TLS. In other words the TLS have to be nearly orthogonal to the phonons, harmonic or anharmonic. The measured dimensionless coupling constant through measurements of the phonon echos and ultrasound attenuation shows that the coupling constant was requisitely small, of $O(0.1)$.

The issues in discovering such states are thus easy to state but impossible to verify in microscopic structural experiments and very challenging in numerical searches as well. The zero point entropy of glasses is of $O(k_B)$ per molecular unit, signifying $O(2^N)$ configurations in an assembly of N atoms. The TLS are a quantum-mechanical leaking of a tiny part of these, judging from the fact that the extrapolated measured integrated entropy of tunneling states from near $T = 0$ to about $T = 10$ K is typically less than $10^{-4} k_B$ per molecular unit. Most of the random configurations are thus mutually inaccessible. This is not a problem which a mean-field theory of ultrametric configurations can address. Specific criteria of choosing the relevant configurations are required.

An ambitious program for numerical simulation of glasses using molecular dynamics to freeze random arrangements of atoms with various simple potentials and study the distribution of potentials and the quantum-mechanical tunneling between them at effectively zero temperature using advanced versions of Monte-carlo techniques and molecular dynamics was started many years ago by the authors above and their collaborators as well as several other groups referred to in the of the papers highlighted. 1500 atoms in random configurations are frozen in a 3 dimensional box at different effective temperatures and rapidly quenched with results obtained from averages over hundreds of random configurations. The distribution of potentials in as many as about a million configurations are examined to look first for pairs of configurations forming the double well structure and then isolating those with

large enough tunneling probability calculated from solving the Schrodinger equation over the most probable one dimensional path between them. The difference between the second and the first paper is the use of machine training and its subsequent use to reduce the computer time taken by about 10^{-5}, as already mentioned. This allows collection of very good statistics indeed. Not only is the distribution of states at low excitation energies found to be nearly a constant, as required for the TLS specific heat, the distribution of effective $\lambda' s$ defined above is also calculated and so are the distribution of distances between configurations and their mean-square fluctuations. The reader will have to read the papers above for the wealth of technical details of machine learning and other techniques employed as well as the details of the results. Importantly, the authors also provide a comparison of several results obtained by machine learning with those without, with very impressive success by the machine. Of the physical conclusions, I particularly liked the emphasis on drawing a distinction between finding double well potentials, which are much more numerous, and the relevant TLS.

One of the truly notable results is that the density of TLS goes down by more than an order of magnitude as the initial freezing temperature goes down by a factor of about 3/2. This is consistent with the fact discovered a few years ago that actual glasses made by vapor deposition have precisely this property [4,5].

On the obverse side, the density of the deduced TLS is over an order of magnitude larger than in experiments in typical glasses including those whose potentials are used in the calculations. My guess is that this has to do with the requirement of near orthogonality from phonons for the TLS mentioned above, which it is not clear is met in the numerical search. There are some ancient questions about the universality of the specific heat in glasses and more so the sound attenuation rate, as well as the transferability of parameters from one measurement to the other which would require much more numerical work to answer. The authors would have to estimate the coupling of the TLS to local displacements about equilibrium to begin to have a grip on these.

More generally, although several details about the TLS discovered are given, their nature needs more specification. This is important to address the even more interesting and long-enduring questions about the glassy state. There is more to the TLS than just the curious low temperature properties. For example, it is known [6] that the coefficient of the linear heat capacity at very low temperatures, in a given chemical species of glasses, is inversely

related to the melting temperature which is $O(10^2)$ times larger than the temperatures in which the linear heat capacity is observed. Not co-incidentally, the reduced low temperature vapor deposited glasses appear to develop a narrow but large specific heat peak near the freezing temperature [4] unlike the others which show the usual near jump downwards. Reducing the TLS thus appears to be connected to reducing the overall configurational entropy of glasses. It is known through other numerical work that plastic flow under shear in glasses starts at specific microscopic regions [7] where the response is non-affine. Do these regions house the TLS? Some suggestions about the properties of TLS in relation to such questions have been given [8], but other view points may be developed. With the advanced machine learning techniques it may be possible to answer such questions but machines need guidance, in the right direction, at this point at least, from humans.

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