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Dynamic model of super-Arrhenius relaxation rates in glassy materials¹

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The principal theme of this talk is that anomalously slow, super-Arrhenius relaxation processes in glassy materials may be activated by chains of atomic displacements. The entropy of critically long excitation chains can enable them to grow without bound, thus stabilizing thermal fluctuations in the local density or atomic coordination of the material. I argue that the intrinsic atomic-scale disorder in a glass plays an essential role in determining the activation rate for such chains, and show that the resulting rate formula is essentially the same as the Vogel-Fulcher law. A key feature of this theory is that the spatial extent of critically long excitation chains diverges at the Vogel-Fulcher temperature. I speculate that this diverging length scale implies that ergodicity is partially broken in the super-Arrhenius region above that temperature, and point out how this partially broken ergodicity may explain the vanishing entropy at that point and other observed relations between dynamics and thermodynamics of the glass transition.

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