

Adam G & Gibbs J H. On the temperature dependence of cooperative relaxation properties in glass-forming liquids. *J. Chem. Phys.* 43:139-46, 1965.  
[Brown University, Providence, RI]

This paper gives a theoretical description of the temperature dependence of kinetic properties of glass-forming liquids centering on the configurational requirements of molecular packing. Its application to the numerical description of different types of experiments allows for a quantitative correlation between thermodynamic and kinetic properties of these amorphous states of matter. [The SCI® indicates that this paper has been cited over 375 times since 1965.]

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"The thermodynamical status of the 'glassy state' was recognized very early as a nonequilibrium state.<sup>1</sup> However, the thermodynamic and statistico-mechanical<sup>2</sup> analysis of the supercooled liquid phase pointed to the importance of the configurational restraints of molecular packing for a deeper understanding of the glassy state and eventually gave rise to the prediction of a second-order transition underlying the experimental glass temperature as a lower limit to be approached only at an infinite time scale.<sup>2</sup> Clearly there was the need for a reconciliation of these two apparently diverging views.

"My doctoral dissertation was on the molecular kinetic description of vol-

ume- and enthalpy-relaxation of amorphous polymers<sup>3</sup> and clearly was in the spirit of the early German contributions of Simon<sup>1</sup> and Tammann (cited in reference one) to the kinetic interpretation of the glassy state. During this work, I became aware of and was greatly impressed by the neat application by Julian H. Gibbs and E.A. DiMarzio of a lattice-theoretical model to the description of the equilibrium properties of polymer melts.<sup>2</sup> I was lucky to obtain a postdoctoral fellowship for one year's work with Gibbs at the chemistry department of Brown University, which eventually led to the paper cited above. This postdoctoral stay abroad was a very stimulating and rewarding experience for me, both personally, by way of encountering a fairly different way of life (as compared to a small German university town), and scientifically (in a chemistry department with a strong physics component deriving largely from its longtime chairman R.H. Cole).

"The reason for the number of citations to the paper may be found in 1) the fascination arising from the phenomenon studied, i.e., the glass transformation; 2) the intellectual satisfaction arising from seeing the reconciliation of apparently diverging concepts; 3) the applicability of the theory for description of phenomena originally not anticipated;<sup>4</sup> and 4) its being one of the very early papers on the statistico-mechanical description of the kinetics of cooperative systems, a topic coming into full attention only in the early-1970s.

"Soon after this work I moved into biophysics and now find myself fascinated by exploring the role of cooperative phenomena in basic processes in biology, in particular in biological membranes."

1. Simon F. Über den Zustand der unterkühlten Flüssigkeiten und Gläser. *Z. Anorg. Allg. Chem.* 203:219-27, 1931.
2. Gibbs J H & DiMarzio E A. Nature of the glass transition and the glassy state. *J. Chem. Phys.* 28:373-83, 1958.
3. Adam G. Zur molekularkinetischen Theorie der Volumen- und Enthalpielaxation im Einfrierbereich organischer Gläser. *Kolloid Z.* 195:1-8, 1964.
4. Goldstein M. Statistical thermodynamics of configurational properties. *Ann. NY Acad. Sci.* 279:68-77, 1976.