The Puzzling Behavior of Normal, Supercooled, and Glassy Water

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Recent experimental and theoretical investigations highlight the central importance of supercooled and glassy states for understanding liquid water, and are providing a body of knowledge from which a coherent interpretation of its properties is beginning to emerge [1]. We will review some of the highlights of this recent work in a style suitable for all specialties [2]. We will also discuss some recent work [3], such as attempts to understand cooperative phenomena in water by investigating the spatially heterogeneous dynamics in the SPC/E model of water by using molecular dynamics simulations [4]. We relate the average mass of mobile particle clusters to the diffusion constant and the configurational entropy. Hence, can be interpreted as the mass of the "cooperatively rearranging regions" that form the basis of the Adam-Gibbs theory of the dynamics of supercooled liquids. Finally, we examine the time and temperature dependence of these transient clusters.

This work was done in collaboration with C. A. Angell, S. V. Buldyrev, G. Franzese, N. Giovambattista, E. La Nave, S. Mossa, S. Sastry, A. Scala, F. Sciortino, A. Skibinsky, F. W. Starr, and M. Yamada.

[1] See, e.g., P. G. Debenedetti and H. E. Stanley, "The Physics of Supercooled and Glassy Water" Physics Today 56[issue 6], 40 (2003)---and the inspirational cover of the 1 June issue; see also the mini-review O. Mishima and H. E. Stanley, "The relationship between liquid, supercooled and glassy water" Nature 396, 329 (1998).

[2] Recent experimental work is described in: O. Mishima, "Liquid-Liquid Critical Point in Heavy Water," Phys. Rev. Lett. 85, 334--336 (2000); Y. Katayama et al, "A First-Order Liquid-Liquid Phase Transition in Phosphorus," Nature 403, 170--173 (2000); A. K. Soper and M. A. Ricci, "Structures of High-Density and Low-Density Water," Phys. Rev. Lett. 84, 2881--2884 (2000); O. Mishima and H. E. Stanley, "Decompression-Induced Melting of Ice IV and the Liquid-Liquid Transition in Water," Nature 392, 164 (1998). Recent calculations include A. Scala, F. W. Starr, E. La Nave, H. E. Stanley, and F. Sciortino, "The Free Energy Surface of Supercooled Water" Phys. Rev. E 62, 8016 (2000); G. Franzese, G. Malescio, A. Skibinsky, S. V. Buldyrev, and H. E. Stanley, "Generic mechanism for generating a liquid-liquid phase transition" Nature 409, 692-695 (2001).

[3] See, e.g., the recent work: A.Scala, F. W. Starr, F. Sciortino, E. La Nave, and H. E. Stanley, "Configurational Entropy and Diffusivity of Supercooled Water" Nature 406, 166-169 (2000); E. La Nave, A. Scala, F. W. Starr, H. E. Stanley and F. Sciortino, "Dynamics of Supercooled Water in Configuration Space," Phys. Rev. E 64, 036102-1 -- 036102-10 (2001); F. W. Starr, S. Sastry, E. La Nave, A. Scala, H. E. Stanley and F. Sciortino, "Thermodynamic and Structural Aspects of the Potential Energy Surface of Simulated Water," Phys. Rev. E 63 041201-1 -- 041201-10 (2001); E. La Nave, H. E. Stanley and F. Sciortino, "Configuration Space Connectivity across the Fragile to Strong Transition in Silica" Phys. Rev. Letters 88, 035501-1 to 035501-4 (2002) cond-mat/0108546; M. Yamada, S. Mossa, H. E. Stanley, F. Sciortino, "Interplay Between Time-Temperature-Transformation and the Liquid-Liquid Phase Transition in Water" Phys. Rev. Letters 88, 195701 (2002); cond-mat/0202094; M. Yamada, F. Sciortino, and H. E. Stanley, "Equation of State of Supercooled Water from the Sedimentation Profile," Phys. Rev. E Rapid Communications 67, 010202 (2003).

[4] N. Giovambattista, F. Starr, S. V. Buldyrev, and H. E. Stanley, "Connection between Adam-Gibbs Theory and Spatially Heterogeneous Dynamics" Phys. Rev. Letters 90, 085506 (2003). Cond-mat/0209395; N. Giovambattista, F. W. Starr, F. Sciortino, S. V. Buldyrev, and H. E. Stanley, "Transitions between Inherent Structures in Water," Phys. Rev. E 65, 041502-1 -- 041502-6 (2002) cond-mat/0201028.