Comment on "Fractons and the Fractal Structure of Proteins"

In a recent Letter, Helman, Coniglio, and Tsallis[1] suggest that the inclusion of crosslinking bonds between nonneighboring monomers of a biopolymer chain would remove the current inconsistency between theory and experiment involving the fractal model[2,3] of electron-spin relaxation in low-spin ferric proteins. The problem in the model concerns the fracton dimensionality,[4] $d_{\rm fr}$, which relates the vibrational density of states $\rho(\omega)$ to the frequency ω as $\rho(\omega)\propto\omega^{d_{\rm fr}-1}$. With no cross-linking $d_{\rm fr}=1$ and the temperature dependence of the Raman electron-spin relaxation rate is predicted to follow a temperature dependence $T^{3+2d_{\rm fr}}=T^5$.[2,3,5] Experimentally, the data suggest a temperature dependence T^{3+2d_c}, where d_c is the chain fractal dimension[6] that defines the scaling exponent of the contour length (Nl_0) with respect to the end-to-end length (R) of the bipolymer chain, $N\propto(R/l_0)^{d_c}$.

Helman, Coniglio, and Tsallis[1] point out that theoretically[4] $d_{\rm fr}=2d/d_w$, where d is the fractal dimension of the structure and d_w is the fractal dimensionality of a self-intersecting random walk on the fractal structure. They further show that $d_w\rightarrow 2$ in the limit of a highly cross-linked structure. The purpose of this Comment is to note that, even in an appreciably cross-linked protein, an inconsistency could remain. It should be noted that cross-linking bonds in proteins are generally an order of magnitude weaker than those along the backbone. This presumably would alter the hopping frequencies across various bonds and influence the number of cross-linkages required to drive d_w to the appropriate limit of 2. It is further speculated that in this limit the concept of a protein backbone would fade and, along with it, the chain fractal dimension as defined above. The chain fractal dimension, d_c, might then be replaced by a reentrant fractal dimension,[6] d_r, relating the total number of bonds (contiguous or reentrant) lying within a sphere of radius R ($N_{\rm tot}\propto R^{d_r}$). The reentrant and chain fractal dimensions are numerically very different for proteins. In myoglobin, for example, $d_c\approx 1.54$ and $d_r\approx 1.91$. The experimentally observed fracton dimension[2,3] in low-spin myoglobin complexes is 1.61 ± 0.05 in metmyoglobin hydroxide (MbOH), and 1.55 ± 0.05 in myoglobin azide (MbN_3). The approximate values of d_w required to satisfy the experimentally observed relationship $d_{\rm fr}\approx d_c$ for the three heme proteins myoglobin, cytochrome C, and cytochrome C551 with the condition that $d_{\rm fr}=2d_r/d_w$ are[6] 2.48, 2.32, and 2.58, respectively.

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