

Liu *et al.* Reply: In our Letter [1], we have demonstrated the plausibility of a structure for β -cristobalite SiO_2 consisting of domains of $I42d$ symmetry, and provided strong evidence against other proposed models, based on first-principles total energy and lattice dynamics calculations. We are pleased that the authors of the preceding Comment [2] agree with us on the major findings of our study, namely, the incorrectness of previous models, and the fact that the structure is derived by rotating the tetrahedra about the average $Fd3m$ structure with dynamic orientational disorder.

Instead of a domain structure, they propose an alternative scenario in which the disorder takes the form of fluctuations of rigid unit modes (RUM's) [2-4]. We feel that the question of whether the structure is "best" described by transient domain formation or fluctuating RUM's is rather subtle, and we remain open-minded about it. However, we would like to make the following points.

First, we note that while a single RUM does not generate any distortion of the tetrahedral units, simultaneous excitation of more than one RUM generally does cause such a distortion. This incompatibility among different RUM's is expected to be an important source of anharmonicity which will give rise to strong correlations. These correlations would have the character of suppressing the excitation of multiple RUM's in favor of condensation of a single RUM in any given space-time region—that is to say, formation of transient domains. Although in principle any RUM may condense, our calculations indicate that domains with $I42d$ symmetry are most likely on energetic grounds. Therefore, to the extent that these anticorrelations are strong, one is naturally led back to precisely the same kind of domain model which we have discussed.

Thus, the question of whether a domain description is appropriate is a relative one, not an absolute one. It depends upon the degree of correlation in the RUM picture, or equivalently, upon the typical correlation length and correlation time (domain size and lifetime) in the domain picture. While Swainson and Dove have cited the absence of sharp peaks in the infrared and Raman spectra of β -cristobalite as evidence against the existence of domains, it is obvious that short domain dimensions and

lifetimes would smear out any such peaks. Thus, we would like to see this argument quantified: What upper bounds are placed upon the domain sizes and lifetimes by the spectroscopic experiments?

Finally, we note that the molecular dynamics simulations discussed in the Comment were carried out with an empirical two-body interatomic potential model [5]. It appears doubtful to us whether such a model can reliably answer so subtle a question as the degree of anticorrelation among RUM's in the high-temperature β -cristobalite structure. In fact, it has been pointed out that this model fails to correctly predict the phase transition of α -quartz and α -cristobalite under pressure; and introduce rather large errors in phonon frequencies in a lattice dynamics study of α -quartz [6]. Unfortunately, first-principles quantum molecular dynamics simulations will be very difficult to perform on unit cells large enough to settle this question directly. Thus, we believe that future work making clever use of first-principles approaches will be needed to unravel this important problem.

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