Kaxiras Replies: In their Comment[1] Swift *et al.* argue that the models for Si_{33} and Si_{45} clusters proposed earlier[2] may not be the lowest-energy configurations. They introduce alternative structures for Si_{45}, obtained by classical potential simulations, and evaluate energy differences with a tight-binding model.

As discussed by Jones and Gunnarsson,[3] finding the lowest-energy configuration for a given size cluster by locating and comparing all the local energy minima is extremely difficult for sizes beyond ~ 10. This led me to pursue a physically motivated approach to the structure of "magic number" Si clusters containing 33 and 45 atoms. My Letter[2] pointed out that further "melting" and "recrystallization" studies are needed to support the validity of the models proposed there. A definitive answer will be provided only by extensive first-principle molecular-dynamics simulations. With this in mind, I shall argue that the claims of Swift *et al.* lack firm justification, both on computational and on physical grounds.

The choice of classical potentials by Swift *et al.* (Ref. 5 in their Comment) to verify cluster geometries is poor. These potentials were derived to reproduce bulk properties of Si and give an inaccurate description of *cluster and surface relative energies.* It is no surprise that they favor other structures (e.g., sixfold coordination) than geometries that are characteristic of the Si surface reconstructions. In fact, it is known that the Tersoff potential gives clusters that have energy per atom lower than bulk Si,[4] which is obviously unphysical. Thus, unconstrained simulations based on these potentials will most likely yield irrelevant geometries for clusters.

The use of the tight-binding model of Tománek and Schlüter[5] to calculate cluster energies is also known to be unreliable, favoring structures of high coordination by a large margin (several eV per atom).[6] The fact that it reproduces correctly the energy of the Si_{10} cluster, a structure of high coordination, does not guarantee that this model gives a reliable energy for the Si_{45} cluster. A good check of the applicability of the tight-binding model to the clusters in question would be a comparison of energies of the relevant Si(111) surface reconstructions to first-principles results. No such check is provided by Swift *et al.* It is doubtful that the structures obtained by the classical potential simulations are even local energy minima within the tight-binding model, a condition necessary for their stability.

Finally, the alternative models of Swift *et al.* are unconvincing on physical grounds. No explanation is provided, other than the high coordination of surface atoms, for their relative stability. If high coordination is the only stabilizing feature, any other size in the range of 40–50 atoms might also have exceptional stability; i.e., in this approach there is nothing special about Si_{45}. In contrast, the models of Ref. 2 were special as the only sizes that can accommodate *exactly* the stable surface reconstructions of Si. In the absence of a clearer description of the $T1$ and $T2$ structures of Swift *et al.* it is difficult to assess further their physicality.

Moreover, there is no attempt by Swift *et al.* to explain the observed low reactivity of Si_{45}.[7] If surface atoms are highly coordinated, as Swift *et al.* argue, addition or subtraction of a single atom should make little difference in the reactivity of the cluster, since the metallic character of the surface would be largely unaffected. This would contradict experimental observations that show a change of *several orders of magnitude* in reactivity between Si_{45} and its neighbors.[7] This effect had a natural explanation in my model for Si_{45} (see Ref. 2).

In short, the models of Swift *et al.* are based on unreliable calculations, they lack support on physical grounds, and they leave the experimentally observed dramatic variations in cluster reactivity unexplained. It may be that the models of Ref. 2 are not the lowest-energy configurations, but the preceding Comment can hardly be taken as evidence in support of this claim.

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