Comment on “Formation of Large Voids in the Amorphous Phase-Change Memory Ge2Sb2Te5 Alloy”

Sun et al. [1] have performed ab initio molecular dynamics simulations on amorphous and liquid Ge2Sb2Te5 (GST) and proposed that very large voids play an important role in the fast reversible phase transition process in this material. Their structures are in sharp contradiction to information found in experiment [2–4] and in more extensive simulations of GST [5,6]. In [6] we suggested that (much smaller) vacancies are essential for the rapid phase transition in GST and in Ge/Te alloys [7].

The findings of [1] are artifacts of the details of the simulation, particularly the cooling rate and the shape of the simulation cell (the authors give no dimensions, but two appear to be $\leq 13 \text{ Å}$). Periodic boundary conditions (PBC) mean that the “void” shown is, in fact, a periodically repeated infinite “gap” in a layered structure. PBC combined with small cell parameters also rule out a decyclically repeated infinite “gap” in a layered structure. PBC (PBC) mean that the “void” shown is, in fact, a periodically repeated void of infinite extent, whose parameters bear no relationship to those found in more extensive simulations and experiments on the same material. The connections between cavities, Te atoms, and the phase transition in GST are not new [6].

Table I. Partial coordination numbers (as defined in Ref. [1]) and first minimum of PDF ($r_1$, used as integration cutoff).

<table>
<thead>
<tr>
<th></th>
<th>Sun et al.</th>
<th>Akola and Jones</th>
<th>Akola and Jones</th>
<th>$r_1$ (Å) Akola and Jones</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ge-Ge</td>
<td>0.68</td>
<td>0.36</td>
<td>0.48</td>
<td>3.2</td>
</tr>
<tr>
<td>Ge-Sb, Sb-Ge</td>
<td>0.72, 0.76</td>
<td>0.14, 0.40</td>
<td>3.0</td>
<td></td>
</tr>
<tr>
<td>Sb-Sb</td>
<td>0.82</td>
<td>0.82</td>
<td>0.97</td>
<td>3.6</td>
</tr>
<tr>
<td>Te-Te</td>
<td>0.77</td>
<td>0.30</td>
<td>0.62</td>
<td>3.2</td>
</tr>
</tbody>
</table>

The numbers in [1] are generally higher than in the rapid quench to 0 K of a well-equilibrated sample at 900 K [9]. Reduced quenching rate. More relevant results would have been obtained with the same number of atoms by using a cubic cell. The present structure is an unphysical, periodically repeated void of infinite extent, whose parameters bear no relationship to those found in more extensive simulations and experiments on the same material. The connections between cavities, Te atoms, and the phase transition in GST are not new [6].

There are unclear points of detail, such as the meaning of a “[111] crystallographic direction” in an amorphous material, but we focus on the use of an elongated cell with two small dimensions in conjunction with PBC and on the quenching rate. More relevant results would have been obtained with the same number of atoms by using a cubic cell. The present structure is an unphysical, periodically repeated void of infinite extent, whose parameters bear no relationship to those found in more extensive simulations and experiments on the same material. The connections between cavities, Te atoms, and the phase transition in GST are not new [6].

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[9] The sample [6] was quenched after cooling from 3000 K to 900 K (42 ps) and data collection (21 ps).