Comment on "Elastic Instability in α-Quartz under Pressure"

In a recent Letter Binggeli and Chelikowsky [1] calculated the pressure variation of the elastic properties of α-quartz, and suggested that the crystal to amorphous transition at high pressure is due to a lattice shear instability. They obtained similar results using both a classical interatomic potential and a first-principles pseudopotential approach. We carried out [2] extensive lattice dynamical and molecular dynamics calculations using the same interatomic potential [3] as used in Ref. [1]. However, we obtain a somewhat different conclusion—we found that the amorphous transition is initiated by a soft optic phonon at the Brillouin zone boundary rather than the transverse acoustic mode near the zone center as suggested in Ref. [1]. In this Comment, we show that the difference is understood by considering the pressure variation of the phonon branches in the entire Brillouin zone.

From lattice dynamical calculations as a function of pressure, we identify the phonon branch showing instability to be along the $\Gamma\mathbf{K}$ direction. Figure 1 shows that the first phonon mode which becomes unstable is that at the zone boundary point **K** at 19 GPa. Above 19 GPa, the α-quartz structure is no longer stable with the present interatomic potential. However, if the lattice dynamical calculations are continued at pressures above 19 GPa retaining the α-quartz structure, the entire phonon branch from K to Γ becomes unstable at 22 GPa. The instability of the acoustic mode at Γ at 22 GPa is related to the anomaly of the elastic constant parameter B_3 as calculated in Ref. [1].

We found a similar zone boundary instability in $AlPO_4$ [2], which has a structure related to α-quartz, and which also shows the pressure induced amorphous transition. In both the cases of α-quartz and $AlPO_4$, the phonon mode initiates the instability of the parent crystal structure. Usually a soft phonon mode causes a second-order or nearly first-order phase transition involving small atomic displacements associated with the soft mode, and the transition leads to another stable crystal structure. In the present case, however, from molecular dynamics simulation we observe a transient superstructure associated with the zone boundary mode, followed by large atomic displacements accompanied by a volume collapse. In fact, the complete first-order transition involving volume collapse cannot be explained in terms of soft modes alone.

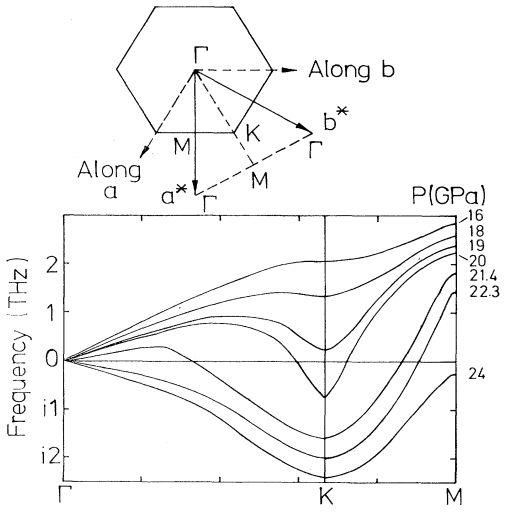

FIG. 1. Calculated pressure variation of the soft phonon branch for the α-quartz structure. The upper part shows the Brillouin zone in the hexagonal plane.

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