Reply to the comment of Angell and Borick

1999 J. Phys.: Condens. Matter 11 8167

(http://iopscience.iop.org/0953-8984/11/41/320)

J. Phys.: Condens. Matter **11** (1999) 8167–8168. Printed in the UK PII: S0953-8984(99)97058-3

Reply to the comment of Angell and Borick

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Received 28 August 1998, in final form 26 July 1999

We appreciate the kind comments of Angell and Borick [1] on our earlier report [2] and their bringing to the readers' attention the results for coordination numbers from molecular dynamics (MD) simulations of liquid Si with the Stillinger–Weber potential [3], which we were not aware of when we wrote our report.

As Angell and Borick point out, the temperature dependence of our measured coordination numbers is consistent with the MD results, although our measurements covered only a limited temperature range (287 K). There is a discrepancy in the absolute scale, with the MD values coming at the upper end of our experimental uncertainties. As we stated in our report, the uncertainties in the changes of the numbers with temperature are certainly less than those in the absolute values.

Interest in the temperature dependence of the coordination numbers arises from extrapolation to lower temperatures and the implications for the phase transition from the high-coordination metallic liquid to the tetrahedrally coordinated semiconducting disordered phase. There appear to be strong grounds for regarding this as a reversible first-order transition [4]. Experimental estimates of the transition temperature (T_{al} in the notation of [4]) range from 1345 K—the observed supercooling limit [2]—to 1340–1420 K from calorimetric studies [4] and 1480 $\pm$ 50 K from transient conductance measurements with laser heating [5]. If T_{al} is 1345 K or higher, Angell and co-workers [1, 3] argue that the tetrahedral disordered phase is liquid below T_{al}, so that this becomes a polymorphic liquid transition of the type proposed to occur generally in tetrahedrally coordinated liquids [6].

As shown in [1], extrapolation of our experimental values gives a coordination number of about 4.7 in the supercooled liquid just above T_{al}, close to the coordination number found in the MD simulation just above the transition (which occurs at a somewhat lower temperature than the experimental estimates). This implies a relatively small change in coordination at T_{al} itself, from 4.7 to approximately 4. (A recent x-ray measurement gives a coordination number of 3.88 in high-purity annealed amorphous silicon [7].)

Finally, we should point out that recent measurements of the density on the supercooled levitated liquid [8] extrapolate to a density maximum around 1200 K, considerably lower than the temperature at which it occurs in the simulation and in fact below the experimental estimate of T_{al}.

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0953-8984/99/418167+02$30.00

Acknowledgments

This work was supported by the US Department of Energy (DOE) Small Business Innovative Research and by the Division of Materials Science, Office of Basic Energy Sciences, DOE, under contract W-31-109-ENG-38.

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