The novel crystallization pathway of rapid cooling SiGe alloy

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The details of simulation and analysis methods are described as follows.

1. Melting point (T_m) of this model

The melting point (T_m) of $Si_{50}Ge_{50}$ alloy is first identified by equilibrating the initial configuration under NVT (constant number of atoms, constant volume and temperature of the system) assembles under zero pressure (see Fig.S1), and T_m is about 2552 K under our present model. Fig.S1 shows the energy evolution of the system with the decrease of temperature. It is easy to find that the system energy will increase or decrease rapidly to respective stable values (about -3.42 eV and -3.90 eV) after a short time of relaxation, and the boundary temperature is about 2552 K.

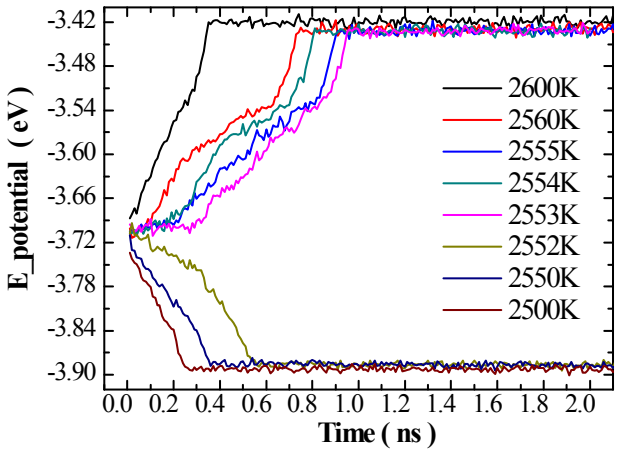


Fig.S1. The energy evolution of the initial configuration during relaxation.

This melting point T_m~2552K is between 2564K (Si) and 2447K (Ge) that are also obtained by Tersoff potential [1]. Although different from the experimental value, this T_m is acceptable, because it is consistent with the general fact that the melting point of SiGe alloy is between those of the crystals of pure Si and Ge[2], and the structure of liquid, amorphous and crystalline SiGe alloy can be well reproduced with this potential.

2. MD Simulation details

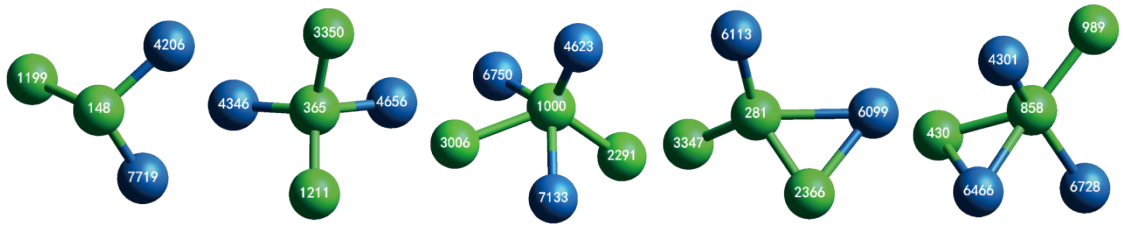
Our simulations are performed under the *NPT* ensemble at zero pressure and the time step is 2.0 fs. All samples contain 8000 atoms (Si:Ge=1:1) in a tetragonal box subjected to the three dimensional (3D) periodic boundary conditions and the initial cell (at T=0 K) arranged as a perfect diamond (DIA) structure (a=5.5373Å) with the mass density of 3.91g/cm^3. To get well equilibrated liquid, the initial configurations first run for 10 ns at 4000K that is much higher than the melting point of this model (T_m~2552 K). The system is subsequently cooled down to 200 K with temperature linearly decreased at R=1.0~8.0×10^9 K/s. The atomic positions and other relevant data of the system are recorded at an interval of 10 K for further structural analysis by several methods.

3. The largest standard cluster analysis (LSCA)

Any kinds of 3D structures can be readily characterized by the largest standard cluster analysis (LSCA) in terms of a topological criterion without any pre-set parameters. In LSCA, a local structure composed of a central atom and its all neighbours in a spherical local space is called a cluster, and any two atoms are bonded if they depart less than the spherical radius. In such a cluster, one neighbour and the central comprise a reference-pair, and a common-neighbour-subcluster (CNS) is composed of a reference-pair and their common near neighbours (CNNs). The topology of a CNS can be systematically described by a CNS index Sijk, where i is the number of CNNs,

j is the number of all bonds between CNNs, and *k* is the number of bonds in the longest CNN string formed by part or all of the *j* bonds. If all CNSs can be denoted by CNS indexes effectively, the cluster is called a standard cluster; and around a given atom, the largest standard cluster (LSC) is unique. The details about the topological criterion and an implementing algorithm can be found in Ref. 23.

For convenience, Fig.S2 demonstrates the major LSCs that will be further discussed in the following sections. In the LSCs of [3/000], [4/000], and [5/000], each neighbour comprises a reference-pair with the central; but there are no CNNs, so they all are S000 CNSs. In the LSCs of [2/000, 2/100] and [3/000, 2/100], two neighbours are bonded with each other and result in two S100. In particular, in [3/000, 2/100] two reference-pairs composed of 858-430 and 858-6466 with only one CNN, comprise two S100 CNSs; other three neighbours comprise three S000 CNSs together with the central.



[3/000] [4/000] [5/000] [2/000,2/100] [3/000,2/100]

Fig.S2 the major LSCs in $Si_{50}Ge_{50}$ alloy.

4. The DIA atom and DIA cluster

The stable crystalline phase of SiGe alloy is diamond (DIA) crystal, where the smallest repeat unit is composed of a centre and its 4 neighbours, in which the angle formed by each pair of neighbours with respect to the centre (called bond-angle) is just 109°28′. Therefore a [4/000] is not necessary a DIA unit because the bond angles may be much deviate from 109°28'. If all bond-angles are within 109°±8°, a 5-atom LSC [4/000] is defined as a DIA core; all the 5 atoms are referred to DIA atoms, and a bond between two DIA atoms is called a DIA bond. As shown in Fig.S3 (a), the

central (blue) and the 1st neighbours (four red atoms) comprise a DIA core. Thus there are at least 5 DIA atoms in a system, if any.

With such a DIA core, a DIA lattice can be constructed; if an atom locates on a grid point (within a certain deviation) of such a DIA lattice and has a path to the DIA core via DIA bonds in the same lattice, it is also a DIA atom; then by means of this criterion, a DIA cluster can be identified (the size of any DIA cluster is no less than 5). For example, Fig.S3 (b) shows a 31-atom DIA cluster (N=31), although its second and third neighbour shells are incomplete.

On the other hand, if two neighbouring DIA atoms belong to different lattices, they are in different DIA clusters. For example, as shown in (see Fig.S3 (b)) all the 20 DIA atoms are bonded but they belong to four DIA clusters centred at atoms 489, 1915, 3496 and 7201 respectively. In particular, the clusters around 489 and 3496 are both 5-atom DIA clusters; and those around 1915 and 7201 are 6-atom ones; and three atoms 3349, 3874 and 6518 are shared by such 4 DIA clusters.

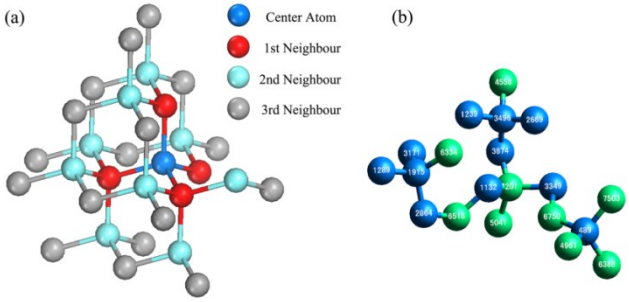


Fig.S3 the demonstration for DIA atoms and DIA clusters: (a) a 31-atom DIA cluster with the incomplete 2nd and 3rd neighbour shells; (b) 4 DIA clusters bonded together via shared atoms selected from the simulation system. The blue and green balls respectively represent Si and Ge atoms in (b).

References

[1] Y. Xiao, T. Motooka, R. Teranishi, S. Munetoh. Nucleation of Si and Ge by rapid cooling using molecular-dynamics simulation, Journal of Crystal Growth 362 (2013) 103-105.

[2] E. Kasper, K. Lyutovich. Properties of silicon germanium and SiGe : carbon, INSPEC, Institution of Electrical Engineers, 2000.