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Crystalline-Amorphous Interface: Molecular Dynamics Simulation of Thermal Conductivity

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ABSTRACT

Effect of a crystalline-amorphous interface on heat conduction has been studied using atomistic simulations of a silicon system. System with amorphous silicon was created using the bond-switching Monte Carlo simulation method and heat conduction near room temperature was studied by molecular dynamics simulations of this system.

INTRODUCTION

As the sizes of electronic devices decrease an increasing amount of heat has to be dissipated by ever decreasing volume of the device material. The details of device structures, namely interfaces, surfaces and defects in them affect the heat conduction, which can give rise to behavior that differs drastically from behavior in bulk materials [1, 2].

In addition to the interest in improving the heat dissipation from electronic components, different nanostructures can be utilized to decrease heat conduction where it is not desired. One example of this are the new thermoelectric devices where thermal conduction is reduced by ultra-short-period superlattices [3]. When the dimensions of nanostructures become comparable to the phonon mean free path in the material the Fourier law describing thermal conduction in macroscopic systems becomes inapplicable. Heat conduction is influenced – among other things – by phonon scattering from interfaces, by phonon interference and the modification of the phonon dispersion relation due to small dimensions of the device. These events can be included in kinetic theories of heat conduction [4] in an approximate way. However, in order to get a reliable estimate how the *atomic* level structure of the device influences the heat conduction, atomic level studies are needed.

In this work we study heat conduction through an interface between crystalline (c) and amorphous (a) material. We use molecular dynamics (MD) method to study the effect of the crystalline silicon and amorphous silicon (c-Si/a-Si) interface on the heat conduction near room temperature. We have chosen this particular system with only structural (not chemical) differences in order to study the basic effects of the interface. This study – which is interesting on own right – serves as a necessary but yet insufficiently explored precursor for investigating perhaps the most important interface in electronics; namely an interface between crystalline silicon and amorphous silicon dioxide.

COMPUTATIONAL DETAILS

Our simulation system consisted of a layer of a-Si between slabs of c-Si (see Fig. 3). In order to avoid complication with open surfaces periodic boundary conditions were applied in all three dimensions. However, this means that the system is effectively a superlattice. Three systems (denoted **A**, **B**, and **C**) with different sizes were studied (see Fig. 3):

A: $L_c = 296 \text{ \AA}$, $L_a = 100 \text{ \AA}$, $d = 32 \text{ \AA}$, 20736 atoms

B: $L_c = 187 \text{ \AA}$, $L_a = 38 \text{ \AA}$, $d = 16 \text{ \AA}$, 2952 atoms

C: $L_c = 187 \text{ \AA}$, $L_a = 38 \text{ \AA}$, $d = 32 \text{ \AA}$, 11808 atoms

Initially the amorphous part of the system was prepared by using the bond-switching Monte Carlo algorithm [5, 6]. In this method a continuous random network (CRN) of atoms is constructed starting from a perfect diamond lattice by introducing disorder through switching bonds between neighboring atoms (see Fig. 1). In this way, four-fold coordination of each atom is preserved but long-range order is destroyed. In the beginning of the simulation the system was mixed by applying bond-switching moves without rejection on the average two times for each atom in the system. After this bond switching moves were continued but this time the move was accepted by using the standard Metropolis algorithm. Moreover, all the atomic positions were relaxed in order to calculate the potential energy of the proposed state. This relaxation was performed by simulated annealing method, which is known to give very good 'optimization' results, quite slowly. The system with a c-Si/a-Si interface was created by applying the bond switches only to the center part of the system that was required to be amorphous.

The bond-switching method requires a potential model that uses explicit bonding information. Therefore, we chose a Keating-type valence force field (VFF) potential model [7, 8] for Si:

$$E = \sum_i k_b (b_i - b_0)^2 + \frac{1}{2} \sum_{i \neq j} k_\theta (\cos \theta_{ij} - \cos \theta_0)^2 \quad (1)$$

where the sums run over the *bonds* in the system and b_i is the length of the bond i and $\cos \theta_{ij}$ is the angle between bonds i and j originating from the same atom. Values for the constants k_b , b_0 , k_θ , and $\cos \theta_0$ were taken from Ref. [8]. In addition to the VFF potential a repulsive potential was applied between those atoms that were not the nearest or next nearest neighbors according to the bonding information used to calculate the VFF potential. This prevented atoms that were not bonded to come too near to each other.

The system created using the bond-switching method was relaxed with the Stillinger-Weber [9] potential and the heat conduction through the interface was studied by using this potential and the method proposed in Ref. [10]. This is particularly suitable to study heat conduction through interfaces, because it does not require a homogeneous system. Moreover, it is compatible with periodic boundary conditions and a rapidly convergent quantity

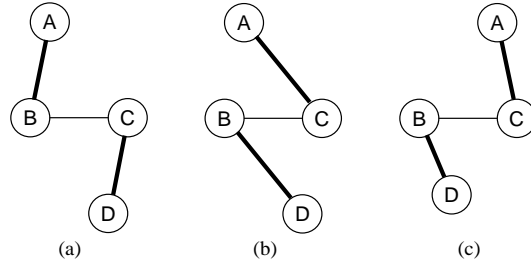


Figure 1. Principle of the bond-switching algorithm. (a) Initial configuration. (b) Configuration after bond switching. (c) Configuration after energy minimization.

– i.e. temperature gradient – is calculated. In this method the desired heat flux \mathbf{J} is generated into the system by exchanging velocities in the 'hot' and 'cool' parts of the system. This creates a temperature gradient ∇T . The thermal conductivity k is calculated based on the macroscopic definition

$$\mathbf{J} = -k\nabla T. \quad (2)$$

In the simulations a slab with width of 8 Å at the center of the amorphous region was assigned as the hot part and slabs with widths of 4 Å at the both ends of the system were assigned as cool parts (see Fig. 3). In this case temperature in the system depends only on one coordinate, namely x , and the problem reduces to one-dimensional: $J = -kdT/dx$. In a homogeneous system the temperature profile should be linear and the thermal conductivity can be obtained by fitting a line to the profile. Conductivities in the amorphous and crystalline parts of the system were calculated this way. Temperature profiles were calculated as averages of data obtained from simulation times between 200–500 ps.

The amount of heat flux can be adjusted by varying the interval between velocity exchanges. In our simulations we used a flux of $J = 3 \times 10^9$ W/m².

RESULTS AND DISCUSSION

In Fig. 2 we show the radial distribution function of the amorphous part of the system along with the recent experimental data [11]. The simulated curve is convoluted with a Gaussian in order to make it comparable with the experimental data. The width of the Gaussian was chosen such that the width of the first peak in $T(r)$ is the same for the experimental and simulated curves. As can be seen, results of the current simulation are in agreement with the experimental data. Also shown in Fig. 2 is the bond angle distribution which is in reasonable agreement with other simulation studies [12]. The final potential energy – given by the VFF potential – of the amorphous part of the system was 0.74 eV/atom. Note that the VFF potential has zero potential energy for the equilibrium diamond structure of silicon. In Fig. 3 we show an example of the configuration created using the bond-switching simulation. It should be noted that not all the crystalline material is shown: some additional crystalline material are added on both sides of the system.

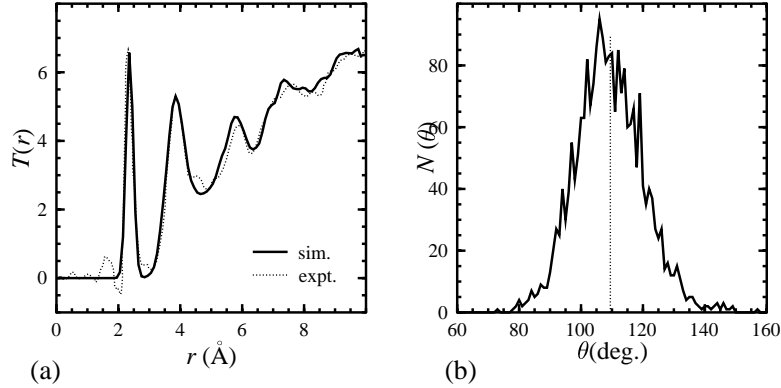


Figure 2. (a) Radial distribution function of amorphous silicon. Solid line is the result of the bond-switching simulation (convoluted with a Gaussian) and dotted line is experimental data from Ref. [11]. (b) Bond angle distribution of simulated amorphous silicon. Dashed line denotes the tetrahedral angle of the perfect diamond lattice.

Simulations of the interface systems yielded the following values for the thermal conductivity in the amorphous (k_a) and crystalline (k_c) regions: system **A**, $k_a = 0.93$ W/mK, $k_c = 13$ W/mK; system **B**, $k_a = 0.85$ W/mK, $k_c = 9$ W/mK; system **C**, $k_a = 0.80$ W/mK, $k_c = 15$ W/mK. Uncertainties due to fitting procedure are approximately 5% and 20% for the amorphous and crystalline values, respectively. For an amorphous silicon system without crystalline parts and with a size of $215 \times 42 \times 46 \text{ \AA}^3$ (20480 atoms) thermal conductivity of $k = 1.11 \pm 0.04$ W/mK was obtained.

Result for the amorphous system without interfaces are in agreement with theoretical and experimental data presented in Ref. [13]. Values of the thermal conductivity for the amorphous regions in the interface systems are not very far from that of the pure amorphous system. Moreover, the dependence on system size is not very strong, indicating that even a moderate size simulation systems are sufficient in describing thermal conduction realistically in amorphous materials. However, the values for the crystalline regions are at least order of magnitude lower than experimental values for bulk silicon (160 W/mK at room temperature) and the calculated values depend on the system size. This is expected since the mean free path of the heat carriers – i.e. phonons – in crystalline silicon at room temperature is of the order of 1000 Å. In addition, the statistical errors in the curves for crystalline regions in Fig. 4 are larger than for the amorphous parts. Finite size of the simulation system is manifested in the bending of the temperature profiles at the ends of the samples. This gives additional uncertainty to the results for the crystalline regions. In order to draw quantitative conclusions from the size dependence of the thermal conductivity more extensive simulations are needed.

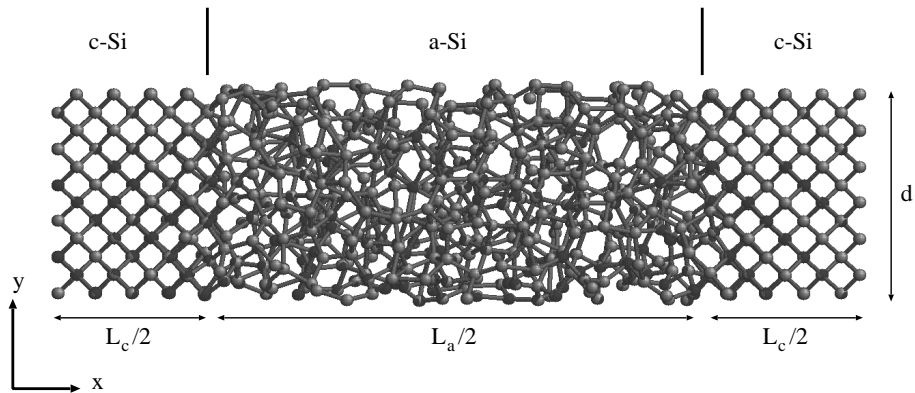


Figure 3. Example of a configuration of a simulation system containing crystalline-amorphous interface.

No thermal boundary resistance could be observed in the simulation data. This probably partly due to the statistical noise in the data and partly due to the fact that the two materials are not very different as far as phonon properties are concerned.

CONCLUSIONS

We have studied heat conduction through an interface between crystalline and amorphous silicon. The effect of the interface can be seen clearly in the temperature profiles. Results for the amorphous regions indicate that even for very thin layers conductivity values turned out to be near the bulk values. Values obtained for the crystalline regions show larger size dependence. Due to the statistical noise in the data and the fact that the two materials are not very different no thermal boundary resistance was observed. This study is currently being extended to investigate the thermal properties of an interface between crystalline silicon and amorphous silicon dioxide.

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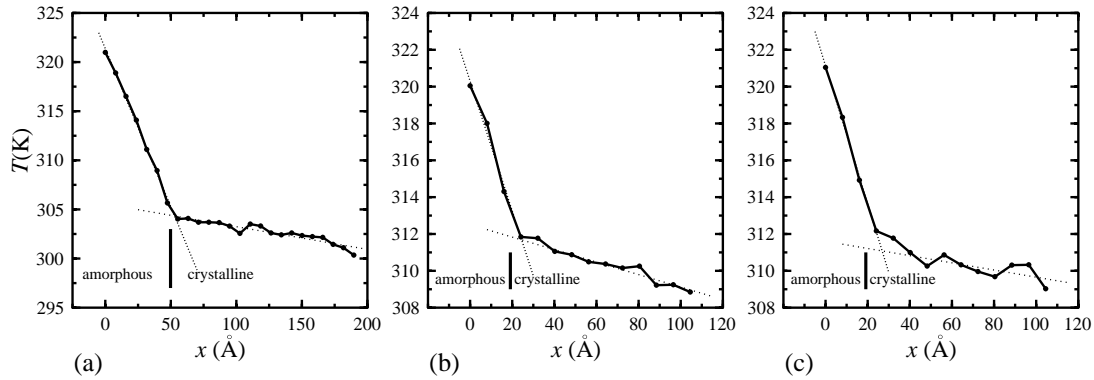


Figure 4. Temperature profiles for the simulation systems **A** (a), **B** (b), and **C** (c). Straight lines are fits to the amorphous and crystalline parts of the curves. Thick vertical lines show the position of the interface. Origin is in the center of the amorphous region (see Figure 3). Curves are averages of the profiles of the left and right parts of the system.

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