

Outlooks for development of silicon nanoparticle memory cells

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Abstract

Phase change memory is based on changes in the optical, electrical or other properties of materials during phase transitions, e.g. an amorphous to crystalline transition. Currently existing and potential applications of this memory are primarily based on multicomponent alloys of metals and semiconductors. However single-component nanoparticles including Si ones are also of interest as promising nanosized memory cells. The potential for developing this type of memory cells is confirmed by the fact that the optical absorption index of bulk amorphous silicon is of the same order of magnitude as that of crystalline silicon. Certainly this phenomenon can hardly be implemented with a single nanoparticle the size of which is within light wavelength. Using molecular dynamics and the Stillinger-Weber potential we have studied the regularities of melting and the conditions of crystallization of silicon nanoparticles containing within 10^5 atoms. We have shown that cooling of nanosized silicon drops at a 0.2 TK/s rate or higher rates causes their amorphous transition whereas single-component nanosized metallic drops crystallize in molecular dynamics experiments even at a 1 TK/s rate. Further heating of amorphous silicon nanoparticles containing above $5 \cdot 10^4$ atoms causes their crystallization in a specific temperature range from 1300 to 1400 K. We have concluded that there is a possibility of developing phase change memory cells on the basis of the above phase transitions. An amorphous transition of a nanoparticle can be achieved by its melting and further cooling to room temperature at a 0.2 TK/s rate whereas a crystalline transition, by its heating to 1300–1400 K at a 0.2 TK/s rate followed by cooling. Results of molecular dynamics experiments suggest there is a minimum silicon nanoparticle size for which the development of phase change memory cells becomes theoretically impossible at a given temperature change rate. For a 0.2 TK/s temperature change rate this minimum size is 12.4 nm (number of atoms approx. $5 \cdot 10^4$).

Keywords

molecular dynamics, Stillinger-Weber potential, silicon nanoparticles, phase change memory cells

1. Introduction

Crystalline and amorphous silicon are the principal materials of modern electronics and change from bulk phases to nanosized particles greatly broadens the outlook of silicon for electronics applications [1]. Currently there is an urgent problem of developing and using random access

and non-volatile memory cells the operation of which is based on silicon nanoparticle phase change. (In line with earlier work [1] we will hereinafter use the notations c-Si and a-Si for crystalline and amorphous silicon, respectively, and nc-Si and na-Si for crystalline and amorphous Si nanoparticles, respectively). Phase change memory has for a long time been used in CD-RW and DVD-RW,

i.e., as external non-volatile memory. Practical applications of this memory have become possible because of a change in the reflective index of a crystalline silver, indium, antimony and tellurium alloy during a crystalline to amorphous transition [2]. However the promise of phase change memory is not limited to the above applications. One can make use of changes not only in optical properties of memory cell material but also changes in its electrical properties during heating [3] or some excitation processes of different origins [4–7]. Special interest has been paid to the promising $\text{Ge}_2\text{Sb}_2\text{Te}_5$ alloy (GST) [8]. For example, a transition from the amorphous to the metastable cubic crystalline phase of GST alloy increases not only its reflective index in the visible spectral region [9, 10] but also its electrical resistivity by three orders of magnitude [11]. Currently an overwhelming majority of works in this field deal with multicomponent alloys. However a hypothesis [12, 13] was put forward and proven, though only in a molecular dynamics experiment, that memory cells can be fabricated making use of phase change and stabilization in Ag nanoparticles. Molecular dynamics experiment [12, 13] showed that very rapid (at approx. 20 TK/s rate) cooling of a liquid metallic nanoparticle to room temperature makes it amorphous whereas relatively slow cooling (at about 1 TK/s or lower rates) causes a crystalline transition. However the authors [12, 13] did not suggest any idea how to practically achieve such a big difference in the cooling rates of nanoparticles located a few nanometers from one another. Furthermore providing such a high cooling rate requires nanosized metallic drops be located on a metallic substrate which might cause their crystallization upon contact within a few seconds or minutes even at room temperature. This requirement imposes strict limitations on the implementation of non-volatile memory cells with this technique. We suggest using silicon nanoparticles for which the critical cooling rate of amorphous transition is several orders of magnitude lower than that for metals. Silicon nanoparticles can be located, e.g. on an amorphous carbon substrate or on a metallic substrate the crystalline structure of which (e.g. FCC) differs largely from that of silicon (diamond), which will possibly avoid na-Si crystallization.

Since we deal with phase change memory based on silicon nanoparticles we preliminarily ran a molecular dynamics experiment for revealing the size dependence of the melting point of Si nanoparticles containing from 1015 to 100153 atoms, i.e., over a sufficiently wide range of sizes from 3.2 to 15.6 nm. This allows rectifying earlier results [14, 15]. Initially we also planned studying the size dependence of the crystallization temperature but it turned out that unlike metallic nanoclusters Si nanoparticles do not crystallize upon cooling even at minimum rates available in molecular dynamics experiments. However bulk phases and thin films are known to have other crystallization mechanisms. In earlier experiments [16, 17] amorphous silicon films [16] and silicon nanoparticles in a matrix [17] crystallized as a result of pulse femtosecond laser heating. Obviously this crystallization method

can be used in phase change memory cells consisting of silicon nanoparticle arrays. Below we will confirm this possibility by a molecular dynamics computer simulation.

Structural changes in Si nanoparticles and practical synthesis of nc-Si have been attracting extensive interest since 1990s. For example one research team [1] following earlier data [18] developed two techniques of synthesizing Si nanocrystals: “top to bottom” (crushing microscopic sized single crystals) and “bottom to top” (self-association of atoms or smaller size nanoclusters). The following synthesis methods were described [19]:

- an electrochemical process;
- nc-Si formation in an a-Si matrix;
- nc-Si synthesis from porous c-Si;
- synthesis of Si nanocrystals in SiO_2 by ion implantation.

In a relatively recent publication [20] nc-Si and na-Si were obtained by decomposing silicon monoxide SiO . Seemingly the authors did not aspire to synthesize nc-Si by na-Si crystallization, yet discussing the possibility of na-Si crystallization. Transmission electron microscopy showed [21] that annealing of amorphous Si particles at 573 to 873 K for 1 h did not cause nanocrystal formation whereas annealing at $T = 1173$ K almost completely crystallized the amorphous particles. However the possibility and probability (reproducibility) of this transition under rapid (pulse) heating remain unclear.

Below we will analyze the possibility of the above-mentioned structural transformations in Si nanoparticles in short characteristic times (up to 100 ns) which can be achieved in molecular dynamics experiments and study the regularities and mechanisms of these transitions including the contributions of the factors revealed in our study. Also we will analyze the general lower limitations upon the sizes of Si nanoparticles and metallic nanoparticles which can be used for the fabrication of phase change memory cells.

2. Molecular dynamics simulation of melting and crystallization of Si nanoparticles

Molecular dynamics simulation of melting and crystallization of Si nanoparticles was carried out using the well-known LAMMPS software [22] and the Stillinger-Weber potential [23]. This proven simulation tool is customized for the simulation of a diamond structure which is the most probable structural modification for Si nanoparticles containing more than 100 atoms [1]. Clearly this potential can only roughly approximate surface reconstruction. Thermostating was by Nose-Hoover method [24]. A nanoparticle for structural transition simulation was chosen to be a sphere cut out of a single crystal material. The initial temperature in the simulating cell was $T = 300$ K. The particle was exposed to a heating and cooling cycle.

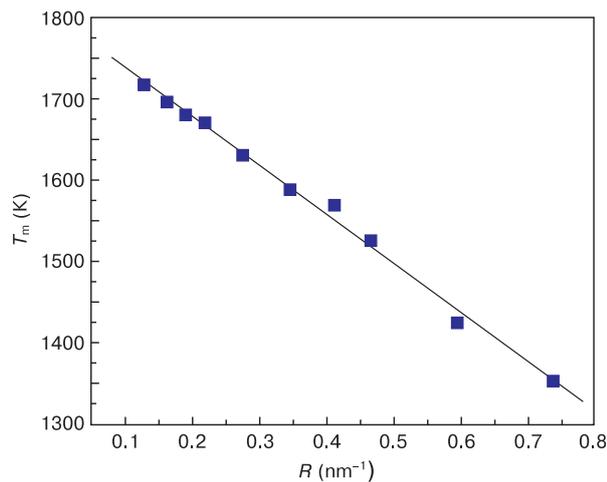


Figure 1. Size dependence of Si nanoparticle melting point T_m obtained from molecular dynamics experiments.

Heating onset caused a relaxation of the initial crystalline structure and the particle was heated to the preset temperature $T = 2300$ K which is *a priori* above the bulk melting point of the material $T_0 = 1688$ K [25] yet not high enough to cause particle material evaporation and destabilization. Further cooling brought the particle back to the initial temperature $T = 300$ K, i.e., *a priori* below the bulk melting point. Under certain heating conditions the particle melting point was preceded by particle crystallization in a certain temperature range. As in earlier works [26–28] we determined the melting point by a discontinuity in the temperature function of potential (cohesive) component of specific internal energy (per atom) (Fig. 1). Metallic nanoparticles also exhibited a similar discontinuity during cooling at the crystallization point $T_c < T_m$. Thus metallic nanoclusters exhibited a melting/crystallization hysteresis. However Si drops did not crystallize but changed to an amorphous state even at a cooling rate of 0.2 TK/s which is one order of magnitude lower than the temperature change rate used for the simulation of melting and crystallization of metallic nanoclusters.

3. Results and discussion

Figure 1 shows size dependence of silicon melting point obtained by molecular dynamics simulation. Figure 2 shows heating and cooling curves for Si nanoparticles containing 100153 atoms for 0.2 TK/s heating and cooling rates. The heating curve of the initial crystalline Si nanoparticle has the melting point related to discontinuity at $T = 1640$ K. Cooling of the molten nanosized drop does not cause crystallization (Fig. 2a). Further heating of the amorphous Si nanoparticle causes its crystallization at $T = 1200$ – 1400 K (Fig. 2b) and melting at higher temperatures. Heating of amorphous Si nanoparticles containing 1000–5000 atoms does not cause their crystallization. Crystallization was observed in several molecular dynamics experiments for nanoparticles containing $5 \cdot 10^4$ or

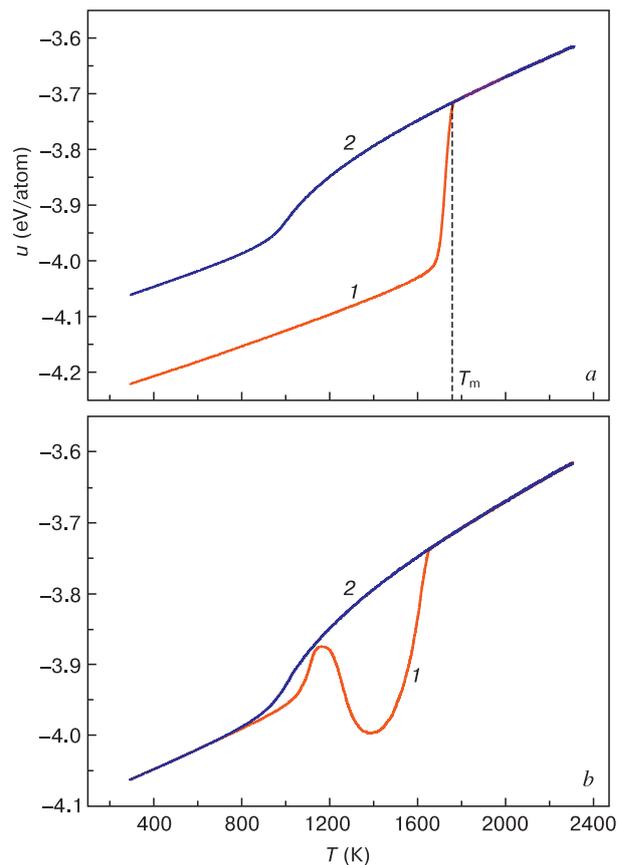


Figure 2. (1) Heating and (2) cooling curves of Si nanoparticles consisting of 100153 atoms: (a) first heating and cooling cycle and (b) further heating and cooling cycles. Melting point T_m is marked by a discontinuity in Curve 1.

more atoms. It is noteworthy that $T = 1200$ K is quite close to 1173 K at which na-Si crystallizes almost completely during annealing [21]. Molecular dynamics simulation data suggesting that larger amorphous Si nanoparticles crystallize more readily agree with earlier thermodynamic data [29]. It was concluded [29] that there is a stability threshold of nc-Si size which is about 3 nm. Smaller nc-Si particles (diameter $D < 3$ nm and number of atoms $N < 700$) transform to na-Si. However these estimates were made for equilibrium thermodynamics and hence implied an equilibrium state. Nonequilibrium pulse heating conditions should increase the stability threshold size which depends on heating rate according to our molecular dynamics experiment. If na-Si particles are heated at a 0.2 TK/s rate the stability threshold of crystalline nanoparticles is $5 \cdot 10^4$ atoms while at a lower heating rate this threshold decreases by one order of magnitude, i.e., to 5000 atoms. Obviously the treatment of the term *stability threshold* in our work differs from that used earlier [29] and hence only a general comparison can be made. Unlike the situation described in Fig. 2a the hysteresis loop for metallic nanoparticles is closed, i.e., metallic nanoparticles always crystallize during cooling at rates of below 10–20 TK/s. This is in agreement with the well-known experimental observation: bulk single-component metals and island

metallic films are not prone to amorphization [30–32]. Si nanoparticles are a contrary case: heating Si nanoparticles of 10^5 atoms to 1800–1900 K and further cooling to 300 K at a 0.2 TK/s rate makes them amorphous whereas amorphous Si nanoparticles crystallize upon heating up to a definite temperature. Figure 3 shows the temperature dependence of the crystallinity degree x of the silicon particles of this size for the abovementioned heating rate. Practically this heating of nanoparticles to the required temperature can be achieved with a pulse laser.

As noted earlier [12, 13] conventional memory types based on electric charge storage have quantum mechanics limitations upon the size of the transistors. If the size of a memory cell is comparable with half of De Broglie wavelength (in silicon it is 3.4 nm and 6.6 nm for the electron and hole conductivities, respectively) electrons or holes will be smeared over its area and the device will not operate as required. However our molecular dynamics simulation suggests that phase change memory cells also have physical limitations upon minimum size. First, for nanoparticles consisting of just a few hundreds of atoms one can only give quite an arbitrary definition of long range order and accordingly of a crystal-liquid transition causing its distortion. Secondly, crystallization may turn out to be impossible in amorphous nanoparticles containing several tens of thousands of atoms since crystallization takes place extremely slow at low temperatures, whereas for higher temperatures at which undercooling is small the critical precipitate size is greater than that of a nanoparticle and therefore an amorphous nanoparticle of this size cannot crystallize due to thermodynamic limitations. Our molecular dynamics experiments reliably confirmed crystallization of Si nanoparticles with sizes of above 12 nm ($N = 5 \cdot 10^4$ atoms). Currently chalcogenides or GST alloy with a typical memory cell size of approx. 100 nm [3] are used as phase change materials. Noteworthy the minimum memory cell size determined by De Broglie wave-

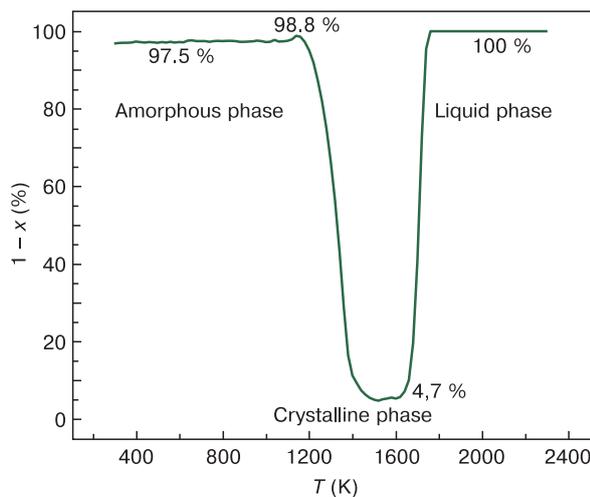


Figure 3. Temperature dependence of crystallinity degree x for heating of Si nanoparticles containing 100153 atoms. $(1 - x)$ is amorphization degree.

length is of the same order of magnitude as our estimate made on the basis of other assumptions.

In this work we concentrated on the regularities and mechanisms of amorphous to crystalline transitions in Si nanoparticles and exploring the theoretical potential to use these phase transitions for random access and non-volatile memory cell fabrication. Studying optical, electrophysical or other phenomena caused by phase change in nanoparticles was initially beyond our scope. However the possibility to make use of changes in optical properties due to amorphous to crystalline transitions in Si nanoparticles is confirmed by the fact that the absorption index of bulk amorphous silicon is one order of magnitude higher than that of crystalline silicon [33]. This cannot be achieved in a single nanoparticle the size of which is within the wavelength of the radiation used. From this viewpoint the size of optical memory cells based on Si nanoparticles should be greater than 100 nm. Pure silicon has a low intrinsic conductivity. However, doping changes the chemical composition of silicon nanoparticles and the regularities of the above discussed phase transitions but slightly, yet greatly increasing the conductivity of silicon. Indeed amorphous and crystalline nanoparticles should have different conductivities and therefore one can develop memory cells making use of the difference in the conductivities of na-Si and nc-Si. The location and orientation of structural components within the neighborhood of an atom are variable in na-Si [1], and this causes the difference in conductivity. On the other hand [1] properties of c-Si and a-Si differ but a little. However [1] the intrinsic carrier mobilities in c-Si and a-Si differ by 3–4 orders of magnitude, i.e., greatly. It was also exemplified [1] that addition of only one impurity atom to nc-Si dramatically changes its electron structure and hence the optical and electron transport properties. For a nanocrystal with the diameter D of about 1 nm, representing its doping level through impurity concentration (or carrier concentration) is quite arbitrary and provides less information than specifying the number of doping impurity atoms. Indeed one impurity atom per 3 nm diameter Si nanocrystal (700 atoms) corresponds to a Si bulk concentration n of $7 \cdot 10^{19} \text{ cm}^{-3}$ [1]. At this doping level Si becomes a degenerate semiconductor, i.e., behaves like a metal. Accepting $n = 10^{19} \text{ cm}^{-3}$ one can easily evaluate the number of doping impurity atoms N_{dop} in a nanocrystal as a function of nanocrystal volume $V = (1/6)\pi D^3$. Results of this evaluation are shown below.

D , nm	N_{dop} , atoms
1	0
6	1
10	5
100	5000

Thus a nanoparticle with $D = 1$ nm will on the average contain no doping impurity atoms and hence particles of this size cannot be used as memory cells. However for

$D = 6$ nm we already have $N_{\text{dop}} = 1$ and N_{dop} increases rapidly with nanoparticle size because $N_{\text{dop}} \sim D^3$. Thus practical application of doped Si nanoparticles as memory cells employing difference between electrical conductivities in amorphous and crystalline silicon seems to be possible yet questionable. One problem to be faced on the way to practical implementation will be self-purification [1] which hinders controlled doping of Si nanoparticles.

4. Conclusion

It was suggested to use silicon nanoparticles as alternative nanosized phase change memory cells. Molecular dynamics experiments showed that nanosized silicon drops do not crystallize during cooling but change to an amorphous state. However heating at a controlled rate in a preset temperature range ΔT caused amorphous nanoparticles to crystallize and retain the crystalline state upon further cooling to room temperature. For silicon nanoparticles containing $N = 100000$ atoms, $\Delta T = 1200$ – 1400 K. We

estimated the characteristic switching time of a respective memory cell to be about 1 ns.

Crystallization was not achieved in our molecular dynamics experiments for $N \leq 10^4$ atoms whereas crystallization is probable for $10^4 < N < 5 \cdot 10^4$ atoms. Thus $N = 5 \cdot 10^4$ ($D = 12.4$ nm) should be accepted as the lower applicability limit of silicon nanoparticles including doped nanoparticles for memory cell fabrication. Detailed description of the physical properties of Si nanoparticles which can be used for making this memory type requires further research effort.

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