

## EXPLOSIVE CRYSTALLIZATION OF AMORPHOUS SILICON: TRIGGERING AND PROPAGATION

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Amorphous silicon may be transformed into crystalline silicon via a self-sustained process driven by the latent heat released upon crystallization. This is called explosive crystallization and is found to occur under rapid-heating conditions such as laser annealing. In this paper we compare different kinds of explosive crystallization, with emphasis on pulsed-laser induced explosive crystallization of ion-implanted amorphous silicon. It is shown that explosive crystallization of amorphous surface layers yields randomly oriented fine-grain polycrystalline silicon while explosive crystallization of amorphous layers buried beneath a crystalline top layer results in epitaxially aligned single-crystal silicon. The results are used to discuss ultra-rapid crystal nucleation and growth.

### 1. Introduction

In many materials the amorphous phase is metastable with respect to the crystalline phase. That is, the amorphous phase has a higher free energy than the crystalline phase and will therefore tend to crystallize under the release of latent heat. The rate of crystallization is strongly temperature dependent and may be negligible at room temperature. In order to fully crystallize amorphous material it is therefore necessary to heat during the time required for crystal nucleation and/or growth. Generally this heat is supplied by an external source such as a furnace or an energy beam. Under favorable conditions, however, it is possible to use the energy released by the crystallization itself to heat nearby amorphous material and to cause further crystallization. If the heat production by crystallization approximately balances the heat losses by conduction into the surroundings the crystallization process is self-sustaining and needs only be initiated. This is called explosive crystallization (EC). EC was probably first described in 1855 by Gore [1], who found that films of electrodeposited antimony “exploded” upon gentle striking or rubbing. Since then, EC has been observed in different substances but a systematic study has only been made

for amorphous silicon (a-Si) [2–44] and to a smaller extent, amorphous germanium (a-Ge) [45–57].

In principle EC can occur in 4 different modes [7], schematically depicted in figs. 1a–1d. Crystallization can either be seeded (epitaxial, figs. 1a and 1c) or unseeded (random, figs. 1b and 1d) and the transition from the amorphous to the crystalline phase can either be direct (figs. 1a and 1b) or mediated by melting (figs. 1c and 1d). It is an interesting question whether all modes can be realized or not and this is one of the subjects of this paper.

When the amorphous → crystal transition is direct, EC is simply sustained by the heat of crystallization. In the case of EC mediated by melting, the overall amorphous → liquid → crystal transition is exothermic, because crystallization from the liquid yields more latent heat than is required for melting of amorphous material. EC is then sustained by the heat which is effectively released upon melting and subsequent crystallization.

When EC occurs through seeded crystallization, the process is governed by solid- or liquid-phase crystal growth. Unseeded crystallization, on the other hand, relies upon spontaneous formation of crystallites in the amorphous matrix or the melt (nucleation). This process is extremely temperature-dependent and for that reason one often de-

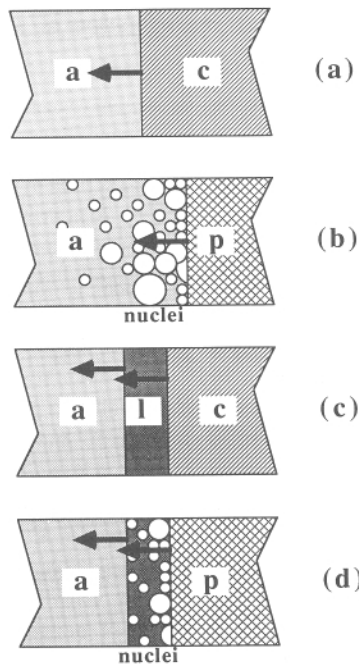


Fig. 1. Schematic representation of the different modes of explosive crystallization: (a) solid-phase explosive epitaxial crystallization (b) solid-phase explosive random crystallization, (c) liquid-phase explosive epitaxial crystallization, (d) liquid-phase explosive random crystallization (a = amorphous, c = monocrystalline, p = polycrystalline, l = liquid).

defines a nucleation temperature to distinguish between the temperature range where nucleation is fast on the scale of the experiment and the range where nucleation does not play a role. At a fixed temperature, nucleation is a transient process and characterized by a delay time during which very little nucleation occurs, followed by a steady-state regime of constant nucleation rate and crystallite growth [58]. Finally the volume fraction of crystalline material becomes significant and the effective nucleation rate drops again. The time-dependent behavior of nucleation marks a distinct difference with seeded crystallization, which is basically an instantaneous process and only temperature dependent.

In this paper we will discuss and compare different modes of EC as they have been observed in a-Si and a-Ge. Emphasis will be put on pulsed-laser induced EC of a-Si layers formed by ion implantation of single-crystal Si (c-Si). This sys-

tem is well-defined and very suitable for detailed time-resolved and micro-structural analysis.

## 2. Lateral explosive crystallization of amorphous silicon and germanium

Pioneering work on EC of a-Ge has been done by Takamori et al. in 1972 [47] and Matsuda et al. in 1973 [45]. They employed films of vacuum deposited a-Ge on insulating substrates and observed spontaneous crystallization propagating in lateral direction. This process could be initiated by a variety of localized energy pulses. EC was found to occur in films held at room temperature but initiation became more easy when the films were heated to 50–100°C. Although the gross features of EC were already explained by Kikuchi et al. in 1974 [59], it was not before 1980 that a more detailed model was proposed by Gilmer and Leamy [60]. The essential part of their model was the prediction that for materials with distinctly different amorphous and liquid phases (such as Si and Ge), EC should be mediated by melting. It was argued that the velocity observed in EC of a-Ge (typically several m/s) is orders of magnitude higher than the maximum velocity for solid-phase epitaxial crystallization as obtained by extrapolation of low-temperature data and that for this reason solid-phase EC is unlikely.

Although the prediction of EC being mediated by melting was confirmed experimentally only a little later [46], the argument based on a comparison with solid-phase crystallization is false. This was implicitly shown by Auvert et al. [28,29,32,39,40], who claimed to observe a combination of liquid-phase EC and two types of solid-phase EC [61,62] in a-Si under scanning focused CW laser irradiation. Although at that time there was no data available on the solid-phase regrowth rate in a-Si at very high temperatures, an extrapolation of low-temperature data implied that also in a-Si the maximum rate was much lower than the apparent velocities in EC (up to 14 m/s for solid-phase EC). Due to extensive studies by Olson and Roth [63] we now know that the maximum solid-phase epitaxial regrowth rate is ~1 cm/s at temperatures approaching the melting temperature of c-Si.

Assuming that the rates determined by Olson on ion-implanted a-Si also apply to deposited a-Si, which is reasonable (ref. [63], p.23), it is clear that indeed solid-phase epitaxial regrowth rates are much lower than the apparent velocities in solid-phase EC. This, however, only shows that solid-phase EC can not proceed via layer-by-layer epitaxy, but only by growth of randomly distributed nuclei, which results in a three-dimensional filling of the amorphous layer with crystallized material. The apparent velocity of EC then reflects the filling rate rather than the growth rate at any a-Si/c-Si interface. This would be in agreement with the observation that material formed by solid-phase EC consists of small grains [29,33].

In later papers Auvert et al. note that one of the two types of solid-phase EC is really liquid-phase EC [33,34,38]. This kind of liquid-phase EC would be nucleation-controlled (unseeded EC in terms of this paper), while "normal" liquid-phase EC would be controlled by crystal growth (seeded EC; we use the term "seeded" when the process is locally and temporarily determined by crystal growth, even though the starting material may not contain a seed). Seeded crystallization is expected to yield large-grained dendritic crystalline material (with the typical dimensions in the experiment as a reference). This structure is frequently observed experimentally [7,33,35–37,50–52]. Analogous, unseeded crystallization yields fine-grained material. The second kind of solid-phase EC is claimed to be a true solid-phase process [38]. Its occurrence may be related to imperfections and strain in the a-Si film [43].

Nucleation in a-Si is generally assumed to play a key role in CW laser induced lateral EC. Part of the a-Si used in Auvert's experiments [28,29,32,38,40] was formed by plasma-enhanced chemical vapor deposition. This implies that the as-deposited material contains impurities and may already contain crystalline nuclei (except in cases where it was ion-implanted after deposition). One should therefore be careful in drawing general conclusions concerning processes which involve nucleation from experiments on this type of material. Nevertheless it is found that the overall features are similar for deposited a-Si and ion-implanted amorphized Si [39].

### 3. In-depth explosive crystallization of amorphous silicon

#### 3.1. General

Most of the experiments described in the previous section were carried out on amorphous layers deposited onto insulating substrates or on free-standing films. EC was triggered locally and propagated primarily in lateral direction, that is, parallel to the sample surface. Study of these systems is complicated because the starting material is poorly defined and there are few experimental techniques available to perform accurate real-time probing of the process. As a result EC could only be described in rather general terms. In 1984 Thompson et al. [2] showed that EC does also occur in ion-implanted a-Si films under nanosecond pulsed-laser irradiation. This type of a-Si has the advantage of being clean and well-defined. In these experiments, coherent triggering of EC was achieved over a relatively large area and it was found to propagate at 10–20 m/s (later measurements yield 8–15 m/s [8,13]) in a direction perpendicular to the sample surface. It was shown that EC is mediated by a liquid layer with an effective thickness of less than 20 nm and that the resulting crystalline material consists of grains with a typical size of 5–10 nm. These findings were soon confirmed by various other groups [3,4,16]. "In-depth" EC of ion-implanted a-Si layers appeared to be a very suitable process for fundamental studies and has been explored intensively since then, using cross-section and planar-view transmission electron microscopy (TEM), impurity redistribution, time-resolved reflectivity and conductivity measurements [5,6,8–14,18,21,22,24–27,30] and computer modeling [21,30,64–66]. The general picture which evolves is shown in fig. 2. Upon pulsed-laser irradiation of an a-Si film, a surface layer is melted. This layer is highly undercooled with respect to the crystalline phase since the melting temperature of a-Si is approximately 200 K below that of c-Si [2]. Crystallites are formed in this melt and subsequently grow towards the surface to produce large-grain polycrystalline Si (lgp-Si). The latent heat released upon formation of polycrystalline Si causes the

embryo formation [58] than nucleation.) Unfortunately little information is available on the delay time and nucleation rate in a-Si at high temperatures [68] and this hampers accurate evaluation of the different suggestions. In addition, the surface and volume energies of small crystallites are not accurately known and this makes it very difficult to decide whether or not (sub-critical) nuclei formed in a-Si will be stable upon exposure to  $\ell$ -Si. The assumption of nucleation in the amorphous phase, however, is in agreement with the observations made in lateral EC, where solid-phase nucleation is observed in front of the scanned CW laser beam (though the time scales are generally different).

We now turn to the triggering of EC. In the situation depicted in fig. 2, nucleation leading to lgp-Si formation appears to be essential to start EC. Basically the set of nucleation mechanisms discussed above also applies here. (Note that the a-Si/ $\ell$ -Si interface velocity is small during a relatively long time when the surface melt is around its maximum depth.) However, there is an additional candidate which is related to the fact that the interface moves in the reverse direction after the maximum melt depth has been reached. It has been observed by Bruines [70] that polycrystalline Si can nucleate at the a-Si/ $\ell$ -Si interface during growth of a-Si from the liquid. The typical time required for nucleation is 10 (20) ns for growth velocities of  $< 0.5$  (1) m/s. This implies that polycrystalline Si may be nucleated at the a-Si/ $\ell$ -Si interface after the melt has reached its maximum depth. Once nuclei have been formed they grow out in the liquid and latent heat is produced. The structure of the material formed is determined by the density of nucleation sites and by the balance between heat production and heat loss into the underlying layers. Under the conditions described this results in formation of lgp-Si.

### 3.3. Epitaxial explosive crystallization

Recently, Polman and coworkers [71–73] have shown that EC of a-Si can also result in the formation of single-crystal Si (see fig. 3). They have formed a-Si layers buried beneath a crystalline top layer by high-energy ion implantation of

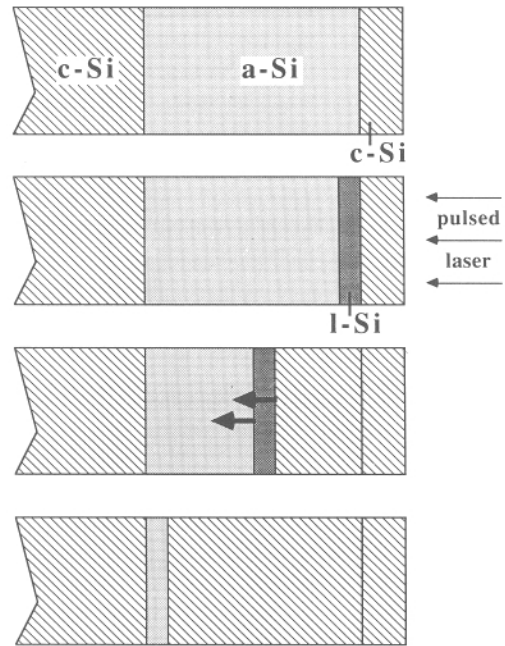


Fig. 3. Schematic representation of liquid-phase explosive epitaxial crystallization in ion-implanted buried a-Si layers.

c-Si and irradiated these structures with a pulsed laser under conditions comparable to those which are known to produce fgp-Si in a-Si surface layers. From time-resolved reflectivity and conductivity measurements, cross-section TEM, impurity redistribution and ion channeling it is concluded that in these sample structures melting is initiated at the buried a-Si/c-Si interface, after which EC is triggered by epitaxial crystal growth from the c-Si top layer. EC is again mediated by a thin  $\ell$ -Si layer and propagates at a velocity of  $\sim 15$  m/s for Si(100) and  $\sim 14$  m/s for Si(111). The material formed consists of c-Si epitaxially aligned with the c-Si surface layer but contains twin defects. An important characteristic of the layers formed in this way is the absence of any polycrystalline material. This is surprising since the irradiation conditions and general EC features are roughly similar to those which yield polycrystalline Si in a-Si top layers. There are three remarks to be made in this context.

(a) Triggering of EC in a-Si surface layers always appears to be delayed by  $\sim 10$ – $15$  ns in comparison to triggering of EC in buried a-Si

layers or a-Si layers containing crystalline nuclei at or near the surface ([73], see also ref. [21]). This delay time has been associated with random nucleation and growth of crystallites necessary to release sufficient latent heat to trigger EC. In the presence of a crystalline top layer or crystallites at or near the surface, the time required for nucleation (and growth of small nuclei) is eliminated and EC is triggered without delay. As a result, the a-Si underneath the  $\ell$ -Si top layer is heated for a longer time prior to EC in case of a-Si surface layers than in case of buried a-Si layers. If fgp-Si is nucleated in a-Si during the heating phase prior to melting, as has been suggested by Roorda [68], this difference in heating time may be crucial and explain why fgp-Si is nucleated in EC of a-Si surface layers but not in EC of buried a-Si layers. It is noted, however, that there is a steep thermal gradient in the a-Si at all times. This is due to the high thermal conductivity of the c-Si substrate and implies that deeper lying a-Si will only be heated to very high temperatures when the  $\ell$ -Si layer mediating EC is near. The effective heating time for this material is therefore primarily determined by the velocity of the liquid layer and not by the moment of onset of EC. This velocity is generally, but not always, slightly lower for EC of a-Si surface layers than for EC of buried a-Si layers.

(b) During EC, the latent heat produced by crystallization has to be transported through the thin  $\ell$ -Si layer to be available for heating and melting of a-Si. It has been argued [73] that the thermal gradient over the  $\ell$ -Si layer associated with this transport is different for formation of fgp-Si and c-Si. This can be understood from the fact that growth of crystallites in the melt constitutes a diffuse heat source with a relatively small thermal gradient while in epitaxial growth the heat source is localized at the plane of crystallization. In the latter case the temperature of the solidification interface can be up to 75 K [73] above that of the melt front, being approximately the melting temperature of a-Si. If the melting temperature of very small crystallites is significantly below that of bulk c-Si [74,75], these crystallites may be formed but remelted during epitaxial EC, whereas they survive in EC yielding fgp-Si. The temperature gradient in the  $\ell$ -Si layer

in the latter process is expected to take such a form that the number of surviving crystallites is just sufficient to sustain EC: a large thermal gradient results in remelting of nuclei, which in turn will cause a lowering of the gradient. This feedback mechanism is supported qualitatively by computer simulations.

(c) The thickness of the  $\ell$ -Si layer mediating EC may be different for epitaxial growth and for crystallite nucleation and growth. Although the *effective* thickness as measured using time-resolved conductivity measurements [2] appears to be roughly the same for both processes in comparable cases (that is, similar a-Si layer thickness and laser energy density) [2,73], this does not mean that the dwell time of crystallites in the melt is the same: in the case of random nucleation and growth the liquid contains a larger fraction of crystalline material and the total thickness should be larger to yield the same conductivity as in epitaxial growth. Therefore crystallites cannot grow to the same size in epitaxial EC as in EC yielding fgp-Si. Very small nuclei may be incorporated in the single crystal by alignment at high temperatures immediately after solidification. The liquid layer thickness has also been suggested be different in the different modes of lateral EC [33,38]: a large thickness in combination with a low velocity in the regime where large grains are formed, a small thickness and a high velocity when small grains are formed.

### 3.4. Discussion

As is evident from the previous sections, the overall features of in-depth EC in ion-implanted a-Si layers are reasonably well understood. This is in contrast with the lack of detailed understanding of the processes on an atomic scale leading to triggering and propagation of EC. As has been mentioned before, the typical time (after surface melting) needed for triggering of EC by random nucleation and growth is 10–15 ns. A comparison to the dwell time in the  $\ell$ -Si layer during propagation of EC ( $\sim 3$  ns) suggests that the nucleation mechanism responsible for propagation of EC is different from that leading to triggering. Although we have given suggestions to explain the various

observations, definite conclusions cannot yet be drawn.

#### 4. Conclusions

We have discussed and compared different modes of EC in a-Si and a-Ge. There is conclusive experimental evidence for EC mediated by melting of the amorphous phase in both a-Si and a-Ge. It has been shown that in a-Si this process can proceed both in an epitaxial way and by random nucleation and growth. There are indications that the situation for a-Ge is similar, but no specific attempts have been made so far to verify this statement. Solid-phase EC of a-Si through random nucleation and growth has been claimed to occur, but we feel that the experimental evidence is not yet complete. To our knowledge this mode has not been observed in a-Ge, which is attributed to the difference in nucleation rates between a-Si and a-Ge. Solid-phase epitaxial EC occurs in neither material, which can be understood from the fact that the maximum epitaxial regrowth rates are too small by several orders of magnitude.

The major difficulty in making a detailed description of EC is the lack of understanding of crystal nucleation in the amorphous and liquid phases. Nevertheless the overall features of EC are now reasonably well understood.

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