From Black Silicon to Photovoltaic Cells, Using Short Pulse Lasers

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Abstract. Laser created Black Silicon has been developed since 1998 at Harvard University. The unique optical and semiconducting properties of the black silicon first lead to interesting applications for sensors (photodetectors, thermal imaging cameras…). Other applications like Photovoltaic solar cells have been rapidly identified, but it took more than ten years of research and development before demonstrating a real improvement of the photovoltaic efficiency on an industrial multi-crystalline solar cell. This paper is a brief review of the use of black silicon for photovoltaic cells.

Keywords: Black Silicon, Laser, Photovoltaics, Solar Cell, micro and nanostructuration….
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The Origin of Black Silicon

The idea of trapping light in a semiconductor device by total internal reflection has been proposed as early as 1968, and the increase of optical absorption in textured semiconductor films has been analyzed by Yablonovitch et al. in 1983 [1]. In the 1980s black silicon was discovered using reactive ion etching (RIE). Researchers noticed that a strong roughening of silicon could occur at the bottom of silicon cavities or trenches when ion etching with $SF_6/O_2/CHF_3$ plasmas [2-3]. This strong anisotropic etching created the first "spike" structures with high aspect ratios and the typical black color. The forming of "grass" on the silicon surface was then a major problem when etching silicon vertically and was attributed to all kinds of micromasks deposited or grown on the silicon. However the particular optical properties of this black silicon was identified for silicon solar cells [4-19; 33-34; 36, 48-49] along with other potential applications like field emission devices [20], MEMS, modifications of hydrophobic properties (wetability) [21], and creation of AFM tips. The RIE technique worked also with other silicon-etch gases, e.g. CF_4, NF_3, SiF_4, CF_3Br. or Cl_2.

At the end of the 1990's, similar black silicon structures have been obtained at Harvard [4-5] by irradiating silicon with femtosecond laser pulses, in the presence of a gas containing sulfur hexafluoride. The surface of silicon develops a self-organized microscopic structure of micrometer-sized cones. The resulting material has many remarkable properties, such as an enhanced absorption that extends to the infrared

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below the band gap of silicon [6-11; 19; 29-35; 37], including the wavelengths for which unmodified silicon is transparent. The sub-gap absorption is caused by deep donor states (or bands, depending on doping level) of sulfur (or other chalcogenides). Specific mechanism is promotion of electrons from defect state/band to conduction band, and is related to optical cross-section of the deep. This particular enhancement of the silicon absorption in the visible and mid-IR range has already found applications for light sensing devices commonly used in security, defense, consumer, industrial, medical, and automotive applications [19]. Recent results [12-17; 19; 48-50] also show that a laser process can produce black silicon with or without SF_6 (Figure 1), with some potential for photovoltaic cells; these results are discussed in the next paragraph.

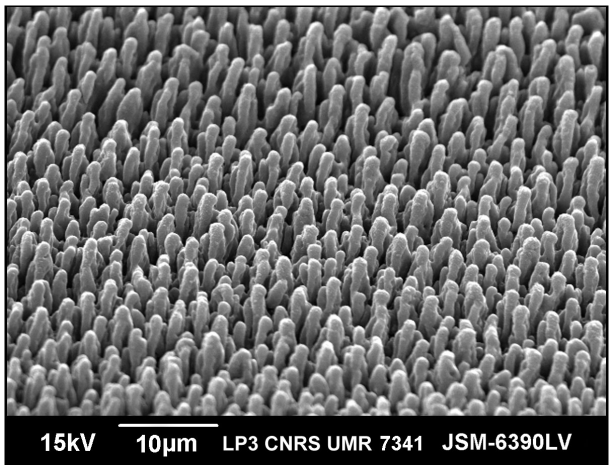

FIGURE 1. SEM photograph of a black silicon structure (conical shape or "penguin-like") created by femtosecond laser irradiation under vacuum.

Formation mechanisms

Laser-induced small periodic structures are created on the surface after the first laser shots (Fig. 2a). These small structures, known as "Laser Induced Periodic Surface Structures" (LIPSS or "ripples"), are similar to capillary waves with a sub-micrometer periodicity (close to the laser wavelength) [38-47]. They are formed at low energy density and/or number of pulses. They are spaced by a distance close to the laser wavelength. These structures are generally attributed to the incident pulse interference with light scattered from defects at the surface, setting up an inhomogeneous melt depth and the formation of capillary waves, which freeze in place [sipe]. However, it has also been shown that for fs laser interaction and depending on the laser parameters (fluence and number of shots), the laser excitation of surface plasmons can also produce such ripples [22-24, 42-44; 46-47].

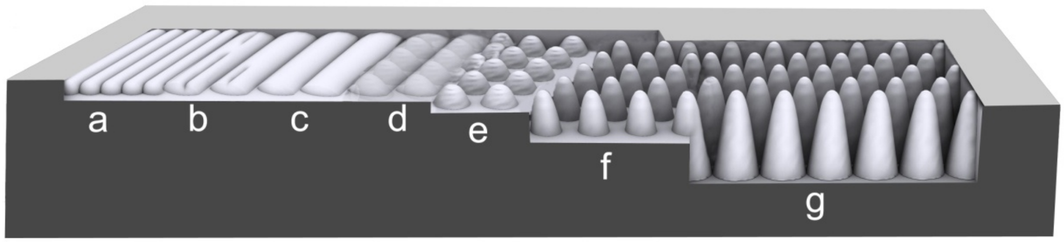

FIGURE 2. Formation of 'black silicon' with increasing laser energy/number of shots: from small parallel structures ('ripples' 700–800 nm periodicity) to the final "spikes" or "cones" (size 5–10 µm).

For higher energy densities and number of pulses these capillary waves tend to collapse (Figure 1 b-d) to form a larger and more hydrodynamically stable structure, like "beads" (Fig. 1, e). The absorption of light on these beads is not uniform: the ablation is maximized in the valley between the beads which tends to amplify the phenomena (Figure 1f, 1g) and creates more erected structures (cones or "penguins", spikes…). The formation of the first ripples and the following transformation of these ripples into spikes as a function of laser parameters, such as energy density, numbers of shots, and laser polarization has been extensively studied in the past few years [22-24; 38-47] and the main differences with structures obtained with longer pulses (ns) are now quite identified.

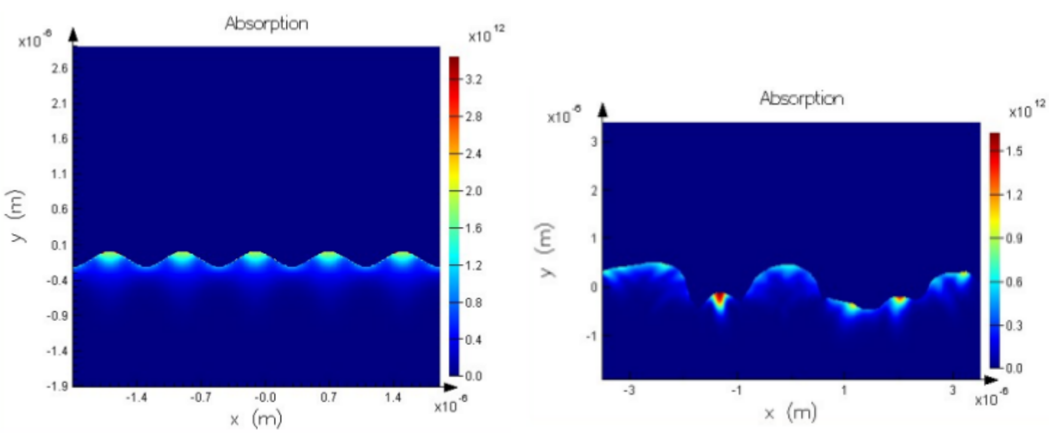

FIGURE 3. FTDT simulation of the absorption of the incident laser radiation on a cross section. Left: ripple structures; Right: ripples and beads (Arbitrary Intensity Units) from [24].

Advanced FDTD simulations have been performed [24] to understand the evolution of the micro structure shape with the fluence, number of pulses and polarization. Since a fs laser irradiation strongly modifies the electron distribution in the first femtoseconds, a new optimized dielectric function has to be taken into account for the excited states of the silicon. Once can noticed on Figure 3 (left) that the absorption of the EM field is higher on top of the ripples. In the presence of ripples and beads (Figure 3, right) the absorption is also maximized on the ripples, not on the beads. This phenomenon probably explains the disappearance of the ripples and the formation of larger structures like beads and eventually cones. Figure 4 shows 3D FDTD simulations that have been performed on excited silicon beads to understand the oval shape of these structures during their formation. Results show that the polarization induces an increased absorption on the sides of the beads, perpendicularly to the polarization, leading to a faster ablation in that direction. These simulations are in good agreement with the SEM top view observations where the lateral dimension of the structure is smaller perpendicular to the polarization. Increasing the number of shots usually decrease this dissymmetry, probably because of a saturation process.

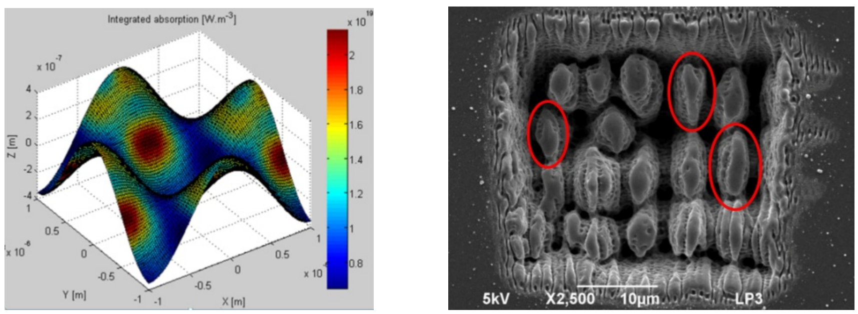

FIGURE 4. Left: 3D FDTD simulations showing the enhanced absorption on the bead sides, perpendicular to the polarization; Right: SEM top view showing the elliptical shape of the silicon structures, perpendicular to the polarization [24].

Crystallographic study of different structures (ripples, beads, spikes) have been observed with an HR-TEM on cross-sectioned samples: these measurements revealed the recrystallisation of the material at different stages (Figure 5). For small features like ripples (Figure 5 left), HR-TEM observations show a nice re-growth of the crystal structure (mainly c-Si).

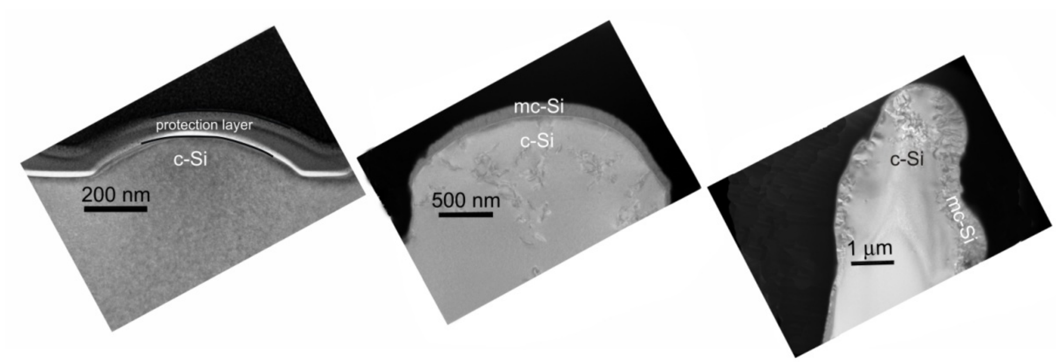

FIGURE 5. HR-TEM views of different structures: ripples, beads and spikes, from left to right, showing the modified Si crystal structures.

However, for larger features (beads and spikes) the crystallographic structure appeared to be more complex. We can observe a transformed multi-crystalline (mc-Si) ‘skin’ layer (which probably corresponds to the rapid melted/solidified layer) on top of the beads (Fig. 5, center) and spikes (Fig. 5, right), with a thickness of a few hundreds of nanometers, while the core of these structures remains mainly monocrystalline. However, the presence of a modified crystalline structure is often observed well beneath this melted layer. When irradiating with of a dopant gas (like SF_6), it has been shown that the core of the spike can be heavily doped on a μm scale, while the supposed melted layer is only in the order of 100 nm [17-18]. This enhanced modification that allows the diffusion of the dopant away from the liquid phase is still under investigation.

Photovoltaic improvements

The black silicon has rapidly proven to be very efficient for photodetectors with a small applied bias, with an extended absorption in the near infrared [19]. However the progress made for photovoltaic cells have been slow and challenging. The first photovoltaic cell made out of black silicon at Harvard had a very poor PV efficiency (1.6 %). They were made with the p-n junction created during the fs laser treatment with SF_6 gas, followed by a thermal annealing at 1075 K for one hour. The annealing step proves critical in improving the efficiency of these samples, confirming that an improvement of the crystal structure has to be performed to increase the lifetime of the carriers. Also the metallization step (evaporation of Cr/Au finger grid contacts onto the microstructured surface) probably induced a large series resistance added from non ideal electrical contact.

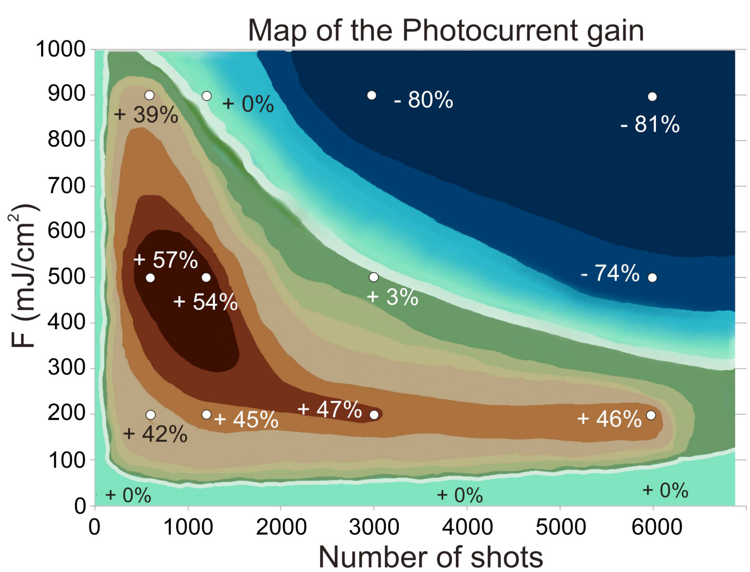

FIGURE 6. Improvement (%) of the photocurrent measured by LBIC as a function of the laser parameters: fluence (200, 500 and 900 mJ/cm2) and number of shots (600, 1200, 3000 and 6000), the false colours are used as an eye-guide, sample: c-Si [17].

Later, we have tried alternative methods at LP3: for instance by separating the doping process from the irradiation process. Mono and multicrystalline silicon were irradiated with ps and fs laser without using SF_6 gas (mainly under vacuum and air). The junction was made afterwards (boron doping) using Plasma Immersion Ion Implantation to create the 3D p+ junction (PULSION IBS system, BF_3, 2–8 kV, 10^{14}–10^{15} at./cm^2) and thermally annealed (900°C, 30 mn, under N_2). The junction depth obtained by this method is measured by Secondary Ion Mass Spectrometry (SIMS). The average junction depth is estimated to be about 150 nm, which is much shallower than the 3D laser structures; therefore, the junction follows the topography of the structures. Depositing Al contacts on the top on the treated area, we were able to obtain the amplification of photocurrent (measured by Light Beam Induced Current (LBIC) by 50% compared with the untreated surface area, using a standard AM1.5 solar illumination [12-17].

Sinton Suns-Voc measurements show that the efficiency of the PV cell increases up to 7%. This efficiency is still low compared to typical commercial crystalline solar cells, but the very promising for the black silicon since many cell parameters still have to be optimized (structure, doping, annealing, metallization, passivation…), in order to fully exploit the potential of the technique. Among the advantages of this laser texturization, we can point out the excellent optical properties: reflectivity less than 5% in the visible for both ps and fs laser treatments without using additional ARC coating. The laser texturization is independent of crystal orientation (good for mc-Si) and has a limited impact on the environment (reduced amount of chemicals, corrosive gases, acid and water). The laser treatment can also replace the SDR (Saw Damage Removal) step.

The black silicon structures still need some improvements to make it suitable for high efficiency photovoltaic cells. For instance, the improvement of the skin layer (topography, crystallographic structure and dopant incorporation) is under investigation in the 2012-2014 B-CELL project (LP3-CNRS/Harvard/IBS collaboration) [51].

To overcome the problem of free carrier recombination, a few approaches have been proposed: a wet etch to remove the defect layer is the most common means, either acidic or alkaline solution etch can be applied [48]. Contrary to the concept of removing the damage layer, laser annealing might be applied to heal the defects. The laser annealing could be an interesting option [26-30] since it is a dry process and it is relatively easy to be integrated with the laser texturization process. Another option is to define the junction away from the damage layer, which can be realized by the combination of minimizing the thickness of laser defect layer and formation of a deep p-n junction.

Recent results

Recently, SiOnyx and German research institute ISC Konstanz have made 17%+ efficient multi-crystalline solar cells using a classic isotexture (HF/HNO_3) followed by a picosecond laser treatment (Coherent AethonTM tool with a TaliskerTM picosecond laser). This improvement represents a 0.3% absolute efficiency boost over the standard isotexture.

The topography of these surfaces is more moderate than some of the historical laser produced spike structures. The resulting surface is more easily passivated and metalized. The process was designed to be fully compatible with the standard industrial PV techniques (standard diffused emitter, surface passivation, front-side silicon nitride ARC, and screen-printed metal contacts).

Standard 156 mm multicrystalline wafers were processed and a comparison was made between cells receiving standard isotexture, isotexture etch combined with ps laser texture, and deep acid polish etch followed by the ps laser texture. An efficiency improvement of 0.3% (absolute) was measured for the cells that received laser texture on top of isotexture, compared to isotexture only. The average efficiency for the laser-textured cells was just over 17%. An important point is that not only is the efficiency increased, but in addition the process variation is reduced. Efficiency binning is improved by a factor of 2X for laser texture compared to isotexture, a benefit we have observed on numerous cell lines. This improvement is due to tighter cell current distributions enabled by the more uniform surface achieved using laser texture compared to isotexture. As a result, the problematic low-end tail of the distribution can be reduced or eliminated, which goes directly to the cell manufacturer's revenue and profit margin.

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