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Supplementary information

Enabling Silicon-on-silicon photonics with pedestaled Mie resonators.

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1 About the crystallinity of the structures

The structures presented in this work are made of monocrystalline silicon with substrate, pedestal, and spheroid-like particle, all sharing the same crystalline orientation. This fact is a direct consequence of the fabrication process. Surface diffusion in crystalline silicon is activated at high-temperatures (T > 900ºC) in deoxidizing ambient, usually in Ar and/or H_2 ambient with very low oxygen partial pressure. These temperatures are well bellow the melting temperature of silicon and mass transport occurs mainly through surface diffusion of Si atoms. Surface atoms migrate from high surface energy regions toward lower energy ones, stacking there following the underlying crystal ordering. Stacking faults can potentially occur in the volumes where material accumulates, but the structure essentially remains monocrystalline. All transformation process occurs in solid phase. The crystalline nature of resonant silicon structures can be assessed through Raman measurements [1]. To avoid the signal of the crystalline Silicon substrate, some a mount of spheroids were scraped from the sample surface and transferred to an aluminium substrate. Measurements were taken with an InVia Raman Microscope from Renishaw using a 532nm laser for excitation with a focus spot of 700nm, well below the size of the measured particles (2µm). Measurements were taken with a 100x objective, and an exposure time of 0.5 s at 1% of the laser power. Figure S1 shows the typical Raman spectrum of a single spheroid, where a sharp crystalline silicon peak is observed at 519 cm^{-1} with a full width at half maximum of exactly 4.5 cm^{-1}. This values are in good accordance to crystalline silicon values, respectively 520 cm^{-1} and 4.5 cm^{-1}, and the slight peak shift can be probably attributed to residual stresses on the structures introduced during the cooling down or due to a slight oxidation of the surface. Other examples of the crystalline quality of the material after transformation by surface diffusion can be found in the literature. For instance, in the Silicon-on-Nothing technique [2] an array of pores etched on silicon are transformed into a free-standing crystalline layer through a high-temperature annealing. In this particular example, researchers have reported [2,3] that the quality of the layer produced by surface diffusion is comparable to bulk silicon, without any observable stress or defects, even though a rather large amount of Si migrates in the process.

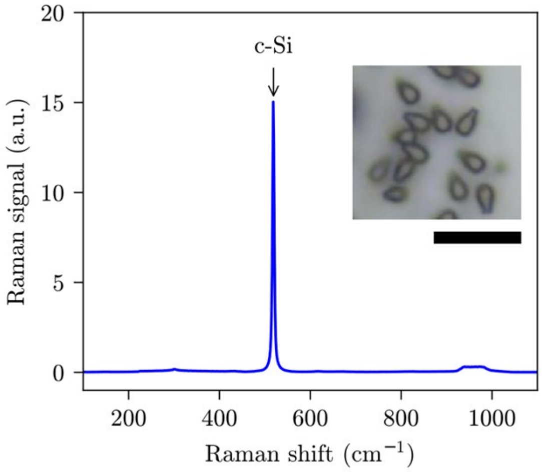

Fig. S1. Raman spectrum of a single pedestalled spheroid. A sharp peak is obtained at 519 cm^{-1}. The inset shows a microscope image of the measured particles over the aluminium substrate. The scale-bar represents 10 µm.

The transformation process can be modelled through the Mullin's equation for surface diffusion, that assumes that the process is isotropic. According to this model, changes in the surface energy arise due to changes in the surface curvature. However, due to the anisotropic nature of crystalline silicon, facets also develop in the surface of the annealed structures since the surface energy of high symmetry planes tend to be lower. The development of facets can also be considered a proof that the structures are monocrystalline. Not only facets develop in the surfaces, but facets in all pedestalled spheroids are equal and oriented in the same direction, as it can be appreciated in figure 1(b) of the main article. In order to verify that the facets correspond to high symmetry planes we have physically measured the angle of some of them with respect to the (100) plane. For this purpose we have carved out half of an spheroid with a focused ion beam facility, leaving a cross section parallel to the (110) plane as defined by the main flat of the wafer. Figure S2(a) shows an SEM image of the carved pedestalled spheroid. Prior the ion beam milling we deposited *in situ* a Pt layer by e-beam assisted gas deposition in order to protect the surface and reduce curtain and beam-tail effects [4]. The image was stretched vertically to correct for the viewing angle, and the angle of the most prominent facets was verified to match the angle of high symmetry planes. In particular, in the figure the planes *b* and *d* matches the (111) plane, the plane *a* matches the (311) crystal plane and the plane c matches the (110) plane. Using this planes as a reference and applying symmetry principles, several other planes were identified over the structures, as shown in figures S2(b) and S2(c). Notice that the contrast of the planes in image S2(c) is rather feeble since the structures are freezed before reaching a stable, minimum surface energy, shape and not all facets are strongly developed. In fact, the most clear facets are those of the {111} family, which is known to present the lowest surface energy [5– 7]. Stable formation of Si facets of the families {111}, {100}, {110} and {311} has also been reported in the annealing of closed voids in Si [8].

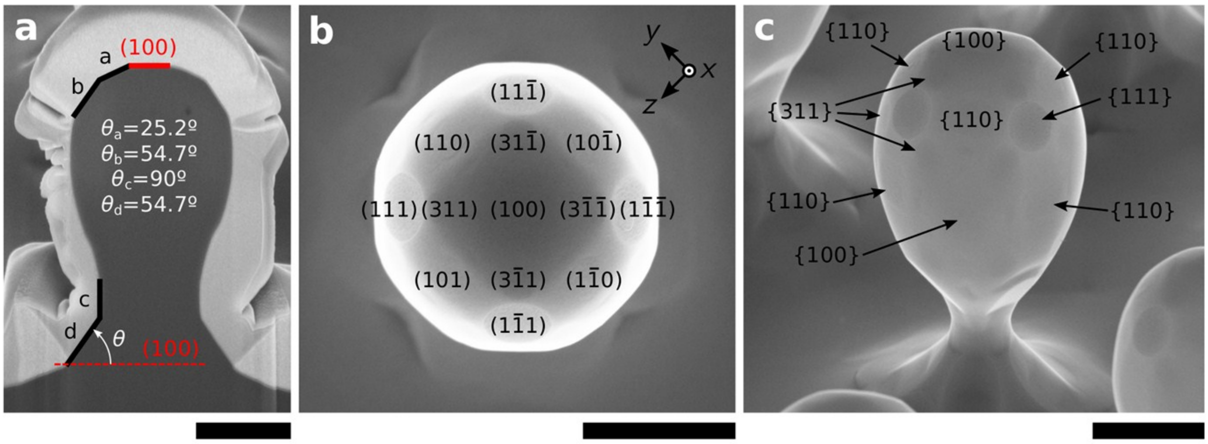

Fig. S2. (a) SEM image of an spheroid cut through the middle with focused ion bean to reveal the angle of the surface facets. (b) Top view SEM image of an spheroid with the main facets labelled. (c) Bird's eye SEM image of an spheroid with the main symmetry facets labelled. The scalebars in all three panels represent 1µm.

2 Process scalability

Transformation by surface diffusion is an scale-independent process and, as a consequence, the structures presented in this work can potentially be scaled down to sub-micrometre dimensions. As an example, in Figure S3 we show the preliminary results of our first attempts to scale the structures from 2µm down to 1µm. A square array of pillars with 1µm diameter and around 3µm in height were fabricated using the exact same process as for the 2µm pedestalled structures. The pillars were then annealed for 1,5h in Ar(95%)+H_2(5%) ambient at 1150ºC. As the figure shows, pedestalled spheroids with a diameter around 1µm were produced, although a few spheroids with a diameter around 900nm can be found. Contact lithography of 1µm features is in the limit of our laboratory capabilities and, as a result, the diameter of the resulting pillars were not consistent, existing noticeable size differences between neighbouring pillars. This explains the dispersion observed in the sizes and also the fact that many spheroids disappeared during the annealing. This would correspond to those with the narrowest diameters, as they would have completely pinched off during the annealing.

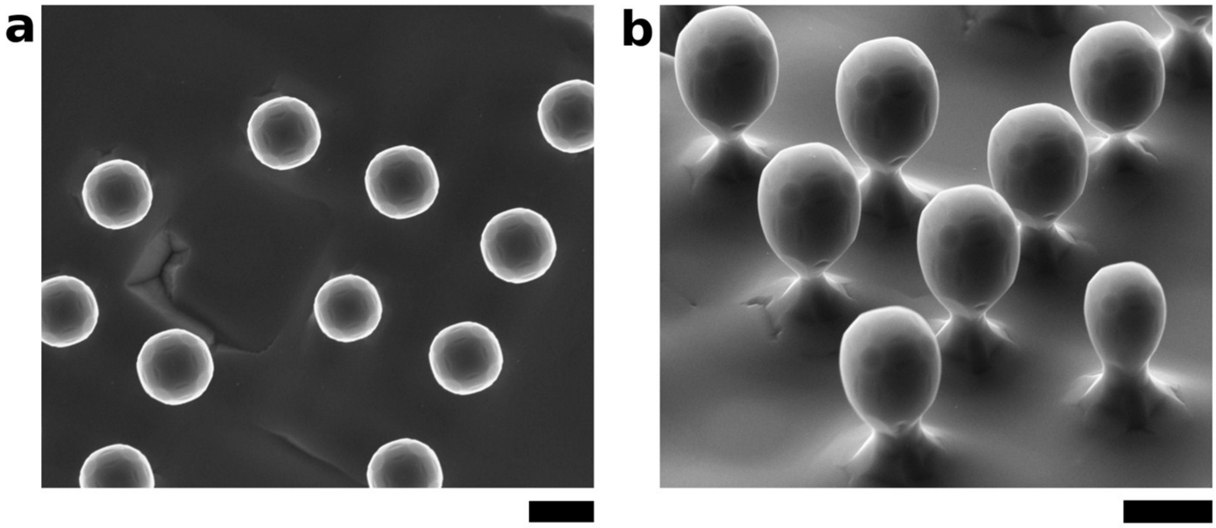

Fig. S3. SEM images of a sample of 1µm pedestalled spheroids: (a) top view, (b) bird's eye view. Scale bars represent 1µm.

After the preliminary results shown in Figure S3, we foresee no fundamental limitation that should prevent scaling the structures further down into the sub-micrometre scale. We believe that spheroids down to 100nm should be realizable by employing better lithography techniques (e.g. electron beam or nanoimprint lithography) and after fine tuning of the process. We believe that the main limitations when scaling down the structures will be related to process inhomogeneities. These might be caused due to lithography issues, such as in fig S3, due to surface roughening and pit formation during the annealing due to slow removal of the native SiO2 [9], and due to both the type and quality of the gases and furnace employed [10]. Finally, contamination of the surface, if present, will also become critical for very small structures, since impurities will interfere with the surface diffusion process introducing inhomogeneities and the formation of pits and defects.

Finally, it is worth mentioning that depending on the application pedestalled spheroids can be produced as both single isolated particles or be very densely packed along the plane. Lateral contact between spheroids during the annealing is possible for very dense initial pillars, although this is not desirable as contacting spheroids will start to sinter and become quickly distorted.

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