Arbitrarily shaped Si nanostructures by glancing angle ion beam sputter deposition

Christian Patzig¹, André Miessler¹, Tansel Karabacak², and Bernd Rauschenbach*,¹,³

¹Leibniz-Institut für Oberflächenmodifizierung e.V. (IOM), Permoserstraße 15, 04318 Leipzig, Germany
²University of Arkansas at Little Rock, 2801 South University Avenue, Little Rock, AR 72204, USA
³Universität Leipzig, Institut für Experimentalphysik II, Linnéstraße 5, 04103 Leipzig, Germany

Received 11 November 2009, revised 22 February 2010, accepted 26 February 2010
Published online 6 April 2010

PACS 68.55.A–, 68.55.J–, 81.07.–b, 81.15.Cd, 81.16.–c

*Corresponding author: e-mail bernd.rauschenbach@iom-leipzig.de, Phone: +49 341 2352308, Fax: +49 341 2352313

Using glancing angle deposition by ion beam sputtering, sculptured thin films (STFs) consisting of various Si nanostructures of manifold shapes, such as inclined and vertical columns, screws, and spirals, were deposited on Si substrates. It will be shown that morphology, shape, and diameter of the structures are influenced and can thus be controlled by adjusting various deposition parameters, including substrate temperature and ratio of substrate rotational speed to film deposition rate. Especially the temperature-controlled surface diffusion is found to play an important role in the growth of STFs.

Cross-sectional scanning electron microscopy micrograph of helical Si nanostructures, deposited with ion beam sputter glancing angle deposition.

© 2010 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction

Among the many variables that influence the growth of a thin film by physical vapor deposition in terms of, e.g., density, crystal structure and morphology, the deposition angle \( \theta \) between the direction of the incoming particle flux, and the substrate normal plays a key role when the films morphology is to be influenced [1]. In contrast to the compact thin film deposition as depicted in the upper part of Fig. 1, where the flux of particles that contribute to the growth of the film reaches the substrate parallel to the substrate normal, the concept of oblique angle deposition (OAD) is such that the particle flux will strike the substrate under a highly oblique angle \( \theta \) to the substrate normal (usually, \( \theta \geq 80^\circ \)). If the nucleation process is governed by Volmer-Weber or Stranski-Krastanov like growth and the surface diffusion is kept low, nuclei on the substrate surface with distinct heights \( h_S \) will exist [2]. After nucleation, high \( \theta \) values ensure that the nuclei shadow the substrate region opposite to the direction of the incoming particle flux on a shadowing length \( l \) that is a function of seed height and deposition angle: \( l = h_S \tan(\theta) \).

The inset of Fig. 1 shows a sketch of the geometrical relations between incoming particle flux and islands on the substrate surface. As a result of this atomic-scale self-shadowing, the deposition of further particles, as soon as first nuclei have formed on the substrate, is restricted to the tops of these nuclei [1, 3]. Hence, instead of a compact thin film, sculptured thin films (STFs) comprising columnar, needle-like structures, slanted toward the direction of the incoming particle flux, evolve, as sketched in Fig. 1.

With the introduction of a suitable azimuthal substrate rotation around the substrate normal during deposition, the OAD concept is expanded to the concept of glancing angle deposition (GLAD) [4], allowing to additionally alter the morphology of the growing structures. This ability to tailor the microstructure of thin films with the recipe “OAD plus azimuthal substrate rotation” was first discovered six decades ago [5], but a widespread use of GLAD did not start until the mid-1990s with the first experiments by Robbie et al. [6–8]. Depending on various deposition parameters, such as deposition angle \( \theta \) and ratio \( \rho = r/\omega \) of deposition rate \( r \) to substrate rotational speed \( \omega \), STFs comprising nanostructures with multiplex morphologies, including columnar and helical structures, can easily be deposited with GLAD [1, 9]. As an example, Fig. 1 depicts the
deposition of vertical columnar structures with GLAD. The simplicity of this deposition technique with its ability to "tailor" the morphology of nanostructured thin films of various materials makes this deposition approach an ideal candidate for the fabrication of functional thin films whose functionality is based on the morphological dependence of various physical properties. Applications of glancing angle deposited thin films include square spiral photonic crystals [10–12], polarization filters [13], polarized light emitters [14], broadband antireflection coatings [15, 16], narrow bandpass optical filters [17], high sensitivity surface enhanced Raman spectroscopy [18], high sensitivity surface enhanced fluorescence [22] substrates, and biomimetization [23], to name only a few.

In this paper, the growth of Si nanostructures with ion beam sputter GLAD on bare planar substrates will be examined. First, some general aspects of GLAD at room temperature (RT), in particular the dependence of the structure morphology on both, deposition time tdep and ratio \(\rho = r/\omega\) of deposition rate \(r\) to substrate rotational speed \(\omega\) will be shown and discussed. After those more general aspects, the influence of the substrate temperature \(T_S\) in the temperature range \(RT \leq T_S \leq 360^\circ\) on the growth of glancing angle deposited structures at different \(\rho\) values will be examined and discussed. Most GLAD experiments described in literature so far were done at \(T_S = RT\), arguing that high \(T_S\) values would increase the surface diffusion length of the incoming particles, which is undesired for GLAD as it counteracts the self-shadowing process (as a rule of thumb, the ratio of substrate temperature \(T_S\) to melting point \(T_M\) of the material to be deposited should be \(T_S/T_M < 0.3\) for negligible surface diffusion [2, 24]). Only few reports concerning the influence of elevated \(T_S\) on glancing angle deposited nanostructures exist so far [25–29], and none of them discusses the influence of \(T_S\) on the growth of screw- and spiral-like structures. In the present study, however, the influence of the surface diffusion on the growth of helical structures grown at low and intermediate substrate rotational speed \(\omega\) (with respect to the deposition rate) is evaluated, and it will be shown that \(T_S\) influences the growth of the structures in terms of merging behavior, film density, and diameter of the structures. Furthermore, it will be illustrated that in a certain range of \(T_S\), it is possible to deposit separated helical nanostructures with diameters of approximately 30 nm. Those diameters stay roughly constant, which is in contrast to the commonly described broadening of the structure diameter \(w\) of columnar GLAD-grown structures with increasing film height \(h\) [30]. A simple growth model to describe this behavior will be given, and the results will be compared with \((2+1)\)-dimensional Monte Carlo (MC) simulations of the GLAD of helical STFs at different surface diffusivities. Additionally, some first results on the behavior of spiral-like Si STFs under mechanical load will be presented.

2 Experiments

As described in previous studies [31–33], all experiments were performed in a high vacuum load-locked deposition chamber, with a deposition setup as sketched in Fig. 2. The base pressure \(p_0\) of the system is better than \(3 \times 10^{-8}\) mbar. The generation of the Ar ion beam that is used for the sputtering process of the Si target takes place in a high-frequency (HF) ion beam source [34]. During deposition, the Ar gas flow \(f_{Ar}\) was set to \((3.5 < f_{Ar} < 6.2)\) sccm, thus resulting in working pressures \((7.0 \times 10^{-5} < p_{dep} < 1.0 \times 10^{-4})\) mbar. The ion beam source–target distance measures approximately 15 cm and...
the distance between target and substrate is approximately 12 cm. The deposition chamber is equipped with a substrate manipulator that can be moved along all three spatial coordinates. It also allows a stepless adjustment of the deposition angle \( \theta \) between target normal and substrate normal. Additionally, the substrate holder can be rotated azimuthally around the substrate normal with rotational speeds \( 0.01 < \omega < 0.2 \) rev/min. The substrate rotation is computer controlled and can be either continuous or stepwise. A tantalum wire resistance heater and a type K thermocouple are both situated under the substrate holder and can be used to adjust and control the substrate temperature \( T_S \) in the range \( RT \leq T_S \leq 400^\circ C \). The angle \( \alpha_{S-T} \) between the direction of the ion beam and the normal of the sputter target (a sintered disk of amorphous Si with a diameter of 15 cm) is freely adjustable. As the sputtering yield reaches a maximum for \( 60^\circ \leq \alpha_{S-T} \leq 80^\circ \) [35], the angle between ion beam source and target was set to \( \alpha_{S-T} \approx (70 \pm 5)^\circ \), where experimentally a maximum of the deposition rate for otherwise constant deposition parameters was found. The ion energy \( E_{ion} \) was set to be approximately \( 1100 \, eV \leq E_{ion} \leq 1250 \, eV \), and the deposition angle was set to \( \theta \approx 85^\circ \). To reduce the angular spread of the sputtered particles, a slit aperture between target and substrate was used, preventing particles with deposition angles less than \( \theta_{min} \approx (76 \pm 3)^\circ \) from reaching the substrate. At \( T_S = RT \), those deposition conditions resulted in deposition rates \( r \approx (3.5 \pm 0.3) \, \text{nm/min} \).

After growth, the samples were cleaved and examined with scanning electron microscopy (SEM) at 2.5 kV acceleration voltage. The micrograph analysis was done using the commercially available software Scanning Probe Image Processor version 3.2.6.0 (SPIP), with the grain detection and fast Fourier transformation (FFT) modules [36].

To determine the mechanical behavior of spiral-like Si STFs, indentation experiments were performed by means of load–displacement measurements with a nanoindentation measuring setup (UNAT, ASMEC GmbH). The flat punch indenter used for the experiments has the shape of a truncated cone with an opening angle of 29.7°, and a spherical surface with a diameter of 45.65 μm (and therefore a contact area \( A_c \approx 1637 \, \mu m^2 \)). This indenter geometry with its large contact area ensures that a number of spiral-like nanostructures is compressed by the indenter. The computer-controlled indentation experiments were performed in dynamic mode, meaning that the displacement \( h_{dis} \) was recorded during the loading and unloading cycle. The contact stiffness \( S \), which is defined as the slope of the unloading curve at the point of maximum displacement [37]

\[
S = \frac{dF_i}{dh_{dis}} \bigg|_{h_{dis}^{\text{max}}},
\]

where \( F_i \) is the loading force, can be used to extract the averaged spring constant of the nanospirals that were compressed with the indenter. The spring constant \( k \) of a single nanostructure can be estimated as \( k = S/(A_c n) \), with \( n \) as number of spirals per \( \mu m^2 \) and \( A_c \) as contact area (in \( \mu m^2 \)) of indenter and STF.

### 3 Results and discussion

**3.1 GLAD at \( T_S = RT \): General aspects of growth**

The ratio \( \rho = r/\omega \) of deposition rate \( r \) to substrate rotational speed \( \omega \) defines the morphology of the growing structure at \( T_S = RT \). Figure 3 illustrates the three different structure morphologies that are obtainable at different \( \rho \) values when the substrate is continuously rotated. The growth of the structures, regardless of the \( \rho \)-dependent morphology, will always start on seeds that nucleate in the beginning of the growth process with diameters within the 20–30 nm range [31]. Keeping \( r \) constant, \( \omega \) was adjusted by changing \( \rho \). For the ion beam sputter glancing angle deposited Si structures studied here, it was found that a column-like morphology is obtained with \( \rho \leq 20 \, \text{nm/rev} \). Likewise, with \( \rho \geq 120 \, \text{nm/rev} \), spiral-like structures (helical with open core) are obtained. For intermediate \( \rho \) cases, screw-like structures (helical with dense core) that quickly evolve out of separated, single spiral-like fibers are deposited. This \( \rho \)-dependent morphological change is linked to the growth front of the structures “following” the particle flux under glancing angle conditions and low adatom mobility. As most of the incoming particles stick to where they land, i.e., the tops of the growing structures, a continuous change of the particle flux direction thus leads to the growth of helical structures like spirals and screws [1, 38].

![Figure 3](image-url) STFs comprising Si nanostructures deposited with GLAD by ion beam sputter deposition: examples for obtainable structure types according to \( \rho = r/\omega \).
However, it was found that this rather broad classification can only be seen as rule of thumb, and is only valid for structure heights \( h \lesssim 800 \text{ nm} \), as the competitive nature of the growth of adjacent structures fosters both structure broadening and structure merging effects, that gradually lead to a change of the structures morphology with increasing structure height \( h \). For columnar structures, deposited with sufficiently fast substrate rotation, the structure diameter \( \omega \) of the surviving structures increases according to a scaling relationship \( \omega \propto h^p \), where \( p \) is a scaling exponent [30]. MC simulations have shown that this broadening effect is correlated with surface diffusion. Low adatom-mobility conditions, that usually exist if \( T_S/T_M < 0.3 \), where \( T_M \) is the melting point of the deposited element [2, 3], tend to exacerbate the increase of \( \omega \) with increasing \( h \) [28]. For the case of Si, at \( T_S = RT \), \( T_S/T_M \approx 0.18 \). Therefore, the surface diffusion length can be considered to be insufficient to allow considerable mass transport away from the impact point of the particle if the experiments are done without extra substrate heating [39]. The incoming particles stick close to the tops of the structures, which in turn increase their diameters as the deposition continues, until neighbored fibers touch each other and merge to form broader structures with increased diameters. Especially for the case of non-columnar structures (slow and intermediate \( \rho \) values), where the growth front of a growing structure overlaps with already existing parts of adjacent structures, this diameter increase can lead to a touching of neighbored, originally separated structures, and a subsequent structure merging. Here, it was found that this merging takes place after the first approximately 200 nm of film growth, as can be seen in Fig. 4a.

The merging effect exacerbates the overall structure broadening. Further structure extinction due to the competitive growth mode in GLAD even at late growth stages can occur as well, since slight differences in structure heights can amplify the self-shadowing process, thereby favoring the “survival” of some of the structures over its neighbors [40]. The extinction of some of the structures is compensated by an increase of \( \omega \) of the surviving structures in order to keep the lateral density constant. Thus, as the structure diameter \( \omega \) increases, the ratio of the pitch \( c \) (height of the structure during one full rotation) to \( \omega \) decreases, leading to a shift of the structures morphology from spiral-like to screw-like, as can be seen in Fig. 4a. A further structure diameter increase (due to both, merging and structure extinction) leads to a further decrease of the pitch-to-diameter ratio, until finally the screw-like character of the structure diminishes and a column-like structure type evolves (Fig. 4b).

Therefore, it is important to notice that the morphology of the growing structures on flat substrates at \( T_S = RT \) is not to be considered constant (except for the case of a fast substrate rotation with \( \rho \lesssim 20 \text{ nm/rev} \) that leads to column-like structures right from the beginning of growth), but rather always changes from spiral-like over screw-like to column-like.

3.2 OAD and GLAD at elevated temperatures

As will be shown in the following subsections, increasing \( T_S \) does significantly alter the structure morphology, as the influence of diffusion-driven processes will increase when the ratio \( T_S/T_M \) of the growing film increases [3, 24].

3.2.1 OAD: Influence of \( T_S \) on the column inclination angle

When STFs are grown using OAD without substrate rotation, the resulting array of nanostructures consists of columns that are slanted into the direction of the particle flux. For OAD of Si with ion beam sputtering at a deposition angle \( \theta \approx 85^\circ \), it is found that the column inclination angle \( \beta \) (i.e., the angle between the column long axis and the substrate normal, as depicted in Fig. 5c) at \( T_S = RT \) is \( \beta = (62 \pm 3)^\circ \). This is in good accordance to the prediction of the *cosine rule*, which was suggested by Tait et al. [41] in order to link the column inclination angle \( \beta \) with the deposition angle \( \theta \) for large \( \theta \) values: \( \beta = \theta - \arcsin \left( 1 - \cos(\theta) / 2 \right) \). This model is based on geometrical assumptions and disregards surface diffusion. Therefore, its validity at increased substrate temperatures is questionable. Hara et al. [42] treated the influence of surface diffusion on the column inclination angle of OAD deposited STFs in a qualitative way, assuming that an enhanced thermally activated and thus non-directional surface diffusion should lead to a deviation of the columnar growth direction away from the substrate normal. Thus, with increased substrate temperature, the column inclination angle should increase as well.

Here, the influence of \( T_S \) on the growth of columnar Si structures with ion beam sputter OAD was investigated in the temperature range \( RT \leq T_S \leq 360^\circ C \). Figure 5a and b shows the difference between columns grown at \( T_S = RT \) and \( T_S = 360^\circ C \). The total film height \( h \) decreases, whereas the column inclination angle \( \beta \) increases with elevated \( T_S \). As increasing \( T_S \) fosters the surface diffusion, this is in accordance to the model of Hara that predicts an increase of the column inclination angle \( \beta \) with an increase of undirected adatom diffusion.

The decrease of the film height \( h \) with increasing \( T_S \) is a consequence of the increase of \( \beta \). As shown in Fig. 5c, the
critical height, a merging of originally separated fibers sets in, leading to spiral-like structures with increased spiral arm diameters in the case of \( \rho = 175 \text{ nm/rev} \) or to screw-like structures in the case of \( \rho = 100 \text{ nm/rev} \). As discussed in the previous subsection, increasing \( T_S \) leads to enhanced surface diffusion and influences the growth and morphology of oblique angle deposited columnar Si structures. In the following, the influence of \( T_S \) on the growth of glancing angle deposited Si nanostructures with helical and vertical column-like morphology will be presented and discussed.

Figure 6 shows the morphological evolution of Si nanostructures with increasing \( T_S \) and the same deposition time \( t_{\text{dep}} = 155 \text{ min} \), for the cases of slow (\( \rho = 175 \text{ nm/rev} \)) and intermediate substrate rotational speed (\( \rho = 100 \text{ nm/rev} \)). As can be seen in the figure, the morphology of the structures changes distinctly with increasing \( T_S \). In the following, the term \( h_{\text{crit}} \) will be used to indicate the critical height at which single, non-merged spiral fibers that start growing in the early deposition phase are still observable in the STFs. The trends for both, critical height \( h_{\text{crit}} \) and total STF height \( h \) with increasing substrate temperature \( T_S \) are shown in Fig. 7. Clearly, two \( T_S \) regions have to be distinguished.

In the \( T_S \) range from RT to 300 °C, an increase of the critical height of merging \( h_{\text{crit}} \) is evident for both \( \rho \) values. At \( \rho = 175 \text{ nm/rev} \), \( h_{\text{crit}} \) increases from \( h_{\text{crit}}(\text{RT}) \approx 150 \text{ nm} \) to \( h_{\text{crit}}(300^\circ\text{C}) \approx 300 \text{ nm} \), and at \( \rho = 100 \text{ nm/rev} \), \( h_{\text{crit}} \) increases from approximately 150 to 350 nm if \( T_S \) is altered in the same way. The helical fibers with diameters within the range of \( w \approx (20 - 30) \text{ nm} \) that emerge in the beginning of growth stay separated over a larger thickness range with increasing \( T_S \). Likewise, with increasing \( T_S \) the film height \( h \)

### 3.2.2 GLAD with slow and intermediately fast continuous substrate rotation

A slow continuous substrate rotation was gained with adjusting the ratio \( \rho = r/\omega \) to \( \rho = 175 \text{ nm/rev} \) (with \( \omega = 0.02 \text{ rev/min} \) at \( r = 3.5 \text{ nm/min} \) at \( T_S = \text{RT} \)). With this \( \rho \) value, the glancing angle deposited Si structures at \( T_S = \text{RT} \) have a spiral-like morphology. Likewise, an “intermediate” \( \rho \) value of \( \rho = 100 \text{ nm/rev} \) was chosen to represent deposition conditions leading to screw-like structures at RT. In both cases, at \( T_S = \text{RT} \) the growth starts with the evolution of spiral fibers, having diameters of approximately 20 nm. At a certain

---

**Figure 5** (online color at: www.pss-b.com) Cross-sectional SEM micrographs of columnar Si structures deposited at \( T_S = \text{RT} \) (a), and at \( T_S = 360^\circ\text{C} \) (b). Inclination angle \( \beta \), film height \( h \), and column length \( h_{\text{col}} \) as function of \( T_S \) (c).

Column length \( h_{\text{col}} = h/\cos(\beta) \approx (1300 \pm 100) \text{ nm} \) stays nearly constant for all examined substrate temperatures. Therefore, the effect of re-evaporation in the examined temperature range \( \text{RT} \leq T_S \leq 360^\circ\text{C} \) can be considered negligible. With the same deposition time \( t_{\text{dep}} \), the same amount of material is deposited in the columnar thin films.

---

**Figure 6** Cross-sectional SEM micrographs of Si nanostructures grown at \( \rho = 175 \text{ nm/rev} \) (left column) and at \( \rho = 100 \text{ nm/rev} \) (right column). (a) \( T_S = \text{RT} \), (b) \( T_S = 300^\circ\text{C} \), and (c) \( T_S = 360^\circ\text{C} \). The dashed lines indicate the critical height \( h_{\text{crit}} \) for each substrate temperature \( T_S \). The scale is valid for all micrographs.
Figure 7 (online color at: www.pss-b.com) Influence of $T_S$ on the critical height of merging $h_{\text{crit}}$ and the total film height $h$ for (a) slow substrate rotation ($\rho = 175 \text{ nm/rev}$) and (b) intermediate substrate rotational speed ($\rho = 100 \text{ nm/rev}$).

decreases. Since the amount of deposited material is the same for all examined substrate temperatures, and re-evaporation should be negligible even at $T_S = 300 \degree C$ as discussed for the case of OAD in the precedent subsection, this film thickness decrease indicates an increase of the films overall density. Thus, the morphology changes from broad, merged structures (spiral-like for $\rho = 175 \text{ nm/rev}$ and screw-like for $\rho = 100 \text{ nm/rev}$) with large interstructure distances at $T_S = RT$ to non-merged, densely packed spirals at $T_S = 300 \degree C$ for thicknesses $h < h_{\text{crit}}$.

It is observable that the critical height at $T_S = 300 \degree C$ is different for both $\rho$ values. With the slow substrate rotation ($\rho = 175 \text{ nm/rev}$) only 85% of the maximum $h_{\text{crit}}$ value at $\rho = 100 \text{ nm/rev}$ are obtained. The difference in $h_{\text{crit}}$ for both $\rho$ values is believed to be a consequence of the different amount of adjacent structures a single fiber is directly able to interfere with for both, slow and intermediate substrate rotational speeds. They can be explained with a simple geometrical consideration: in the case of a slow substrate rotation, the growing spiral fiber, if not merging to broader structures, circumscribes a cylinder with a diameter of (150 ± 15) nm, as shown in Fig. 8a. In the case of intermediate substrate rotational speed, the imaginary cylinder has a smaller diameter of (85 ± 10) nm (Fig. 8b). If the seeds starting to grow off are considered to be hemispherical with a diameter of approximately 30 nm, it follows that one spiral arm in the case of the slow substrate rotation interferes in the growth of approximately 25 adjacent structures, whereas one spiral arm in the case of the intermediate substrate rotation directly intercepts the growth direction of only approximately 8 neighbored structures. Thus, it seems possible that there is greater interstructure competition in the case of the slow substrate rotation ($\rho = 175 \text{ nm/rev}$), which can be an explanation for the earlier structure merging as compared to the intermediate substrate rotation ($\rho = 100 \text{ nm/rev}$) case.

For both $\rho$ cases it can be concluded that in the $T_S$ range between RT and approximately 300 °C, the merging of primarily separated, fibrous nanospirals to broader structures is delayed and the critical height of merging $h_{\text{crit}}$ is increased, thus leading to the growth of STFs that consist of separated, densely packed, spiral-like fibers.

A further increase of $T_S$ from 300 to 360 °C results again in a significant change of the structure morphology, accompanied by a drastic drop of $h_{\text{crit}}$: as Figs. 6 and 7 show, at $T_S = 360 \degree C$, broad, screw-like structures with values of $h_{\text{crit}}$(360 °C) ≈ 115 nm and diameters $w$ between 250 and 500 nm do exist for both $\rho$ cases.

Obviously it is possible to grow separated, helical nanostructures of Si with diameters in 20–40 nm range using GLAD at elevated temperatures, as long as the total film thickness $h$ is kept below $h_{\text{crit}}$ for the cases of slow and intermediately fast substrate rotations.

To evaluate the changes during film growth at constant $T_S$, a set of films was grown at $T_S = 300 \degree C$ for different deposition times $t_{\text{dep}}$ to examine the film morphology before and after reaching $h_{\text{crit}}$. As in principle the effects are the same for both $\rho$ values, this was done with $\rho = 100 \text{ nm/rev}$ only. The results are shown in Fig. 9. As can be seen, if $h < h_{\text{crit}}$ ($t_{\text{dep}} = 85 \text{ min}$, $h \approx 220 \text{ nm}$), truly separated spiral fibers without structure broadening and merging are deposited. The onset of merging is reached for $h \approx h_{\text{crit}}$ ($t_{\text{dep}} = 155 \text{ min}$, $h \approx 400 \text{ nm}$) and is characterized by the existence of merged clusters that are embedded in-between some still separated spiral fibers. Finally, when $h > h_{\text{crit}}$ ($t_{\text{dep}} = 175 \text{ min}$, $h \approx 450 \text{ nm}$), the formerly separated spiral fibers are merged into screw-like structures with increased diameters.

3.2.3 GLAD with fast continuous substrate rotation As already discussed in a previous publication [38], if the effect of enhanced $T_S$ on the morphological
The temperature-driven morphological changes for slow and intermediate substrate rotation as described here can be understood considering the effects of surface diffusion, as also explained in a previous work [38].

In the beginning of growth, the film consists of separated, spiral-like fibers. The top of one growing spiral-like structure locally resembles a slanted rod. At $T_S = RT$, as discussed above, the adatom mobility will be insufficient to foster a significant motion of the incoming particles before they are deposited with sufficiently fast substrate rotation) is evaluated, merging and densification are fostered with increasing $T_S$ and therefore increasing the adatom-diffusion lengths. Similar experiments as described in the previous subsection, except for $r$ which was set to $r = 17$ nm/rev led to the growth of vertical Si columns.

Briefly, the following effects are observable with increasing $T_S$ from RT to approximately 360 °C, as is also illustrated in Fig. 10: a decrease of the STF thickness $h$ takes place, from $h(\text{RT}) = 490$ nm to 73% of this value: $h(360 \, ^\circ C) = 360$ nm. Additionally, the structure number density $n$ ($\mu$m$^2$) decreases with increasing $T_S$. Obviously, at $\rho \approx 17$ nm/rev an increase of $T_S$ from RT to 360 °C changes the structure morphology from separated columns to densely packed columns with larger average column diameters $\langle w \rangle$. A $\langle w \rangle$ value calculation by averaging over approximately 50 columns using SPIP [36] on top-view SEM micrographs of the STFs deposited at different $T_S$ indicates a change of the average column diameters from $\langle w \rangle \approx 115$ nm at $T_S = \text{RT}$ to $\langle w \rangle \approx 140$ nm at $T_S = 360°$.

From the top-view SEM micrographs of the columnar structures deposited at different $T_S$, the power spectral density (PSD) function curves were calculated as well. When the PSD data is drawn versus the spatial frequency $f$, the column–column separation $\lambda$ of the STF that comprises vertical, columnar structures can be deduced from the maximum frequency $f_{\text{max}}$ [43]. The results are shown in Fig. 10f, showing that $f_{\text{max}}$ shifts to larger values with increasing $T_S$. This means that the spatial wavelength, i.e., the column–column separation $\lambda$, decreases with increasing temperature, from $\lambda(\text{RT}) \approx 179$ nm to $\lambda(360 \, ^\circ C) \approx 156$ nm. This result is in accordance with MC simulations of the growth of columnar nanostructures deposited with $\theta = 85°$ and with different numbers of diffusion steps (DS), which indicate an increase of $f_{\text{max}}$ with increased surface diffusion as well [44, 45].

3.2.4 Growth model The temperature-driven morphological changes for slow and intermediate substrate rotation as described here can be understood considering the
buried under newly arriving ones. Thus, the particles that reach the tip of the growing spirals will not travel long distances along the sides of the already existing fibers, but will rather stick in close vicinity to their impact points, thus gradually increasing the spirals’ diameter. Finally, neighbored structures are broad enough to touch and merge together, as sketched in Fig. 11a. Increasing $T_S$ to 300 °C enhances the adatom mobility, giving the incoming particles the possibility to travel along the sides of the spirals for a certain distance and preventing a broadening and early merging of the spiral fibers, as shown in Fig. 11b.

Figure 11c underlines this point: whereas for the case of $T_S = 300$ °C, increasing $t_{dep}$ from 8 to 30 min at $\rho = 100$ nm/rev leads to no significant merging of the single spiral fibers that are observable in the beginning of the growth process, at $T_S = RT$ after $t_{dep} = 30$ min merged structures with diameters in the range of $w \lesssim 100$ nm have developed out of single spirals that exist in the beginning of the growth. According to this, the average interstructure separation $\lambda$ stays about constant at $T_S = 300$ °C with increasing $t_{dep}$ from 8 to 30 min, whereas $\lambda$ increases for the same variation of $t_{dep}$ at $T_S = RT$. In Fig. 11d, the PSD function graphs of the top-view SEM micrographs of Fig. 11c indicate that at elevated temperatures, with increasing $t_{dep}$ from 8 to 30 min the peak maxima of the spatial frequency $f_{max}$ show only a slight decrease according to a change in $\lambda$ from $\lambda(8 \text{ min}) \approx 34$ nm to $\lambda(30 \text{ min}) \approx 42$ nm. Different from that, for $T_S = RT$, $\lambda$ increases from $\lambda(8 \text{ min}) \approx 34$ nm to $\lambda(30 \text{ min}) \approx 114$ nm (although a clear PSD peak at $T_S = RT$ and $t_{dep} = 30$ min is difficult to observe, the shift of the peak to lower frequencies with increasing deposition time is obvious).

The densification of the films with increasing $T_S$, visible through the decrease of the overall film thickness $h$, can also be interpreted as a consequence of the enhanced surface diffusion: at $T_S = RT$, shadowing effects are dominant, compared to diffusion-driven mass transport. In this case, slight height differences of neighbored spirals and merged screw-like structures exacerbate the shadowing effect. As a consequence, smaller structures terminate their growth and are overgrown by adjacent ones. The surviving structures capture all the incoming particle flux and the resulting, underdense STF consists of structures with large interstructure distances. With increasing $T_S$, however, the adatom diffusion along the sides of the spiral fibers favors the growth of densely packed spirals with approximately equal diameters. The delay of structure merging and extinction in this case results in the STF remaining dense, thus having an overall film height $h$ being less than in the RT case.

If $T_S$ is increased to values at which the adatom mobility becomes sufficiently large, enabling the diffusing particles to overcome the distances between single fibers, early merging results in broad, screw-like structures. Unlike as in the case of $T_S = RT$, where the shadowing effect is dominant, at elevated temperatures ($T_S = 360$ °C in this case) mass transport by surface diffusion plays a significant role. Screw-like structures deposited at $T_S = 360$ °C are therefore broader and denser than screws (at $\rho = 100$ nm/rev) and merged spirals (at $\rho = 175$ nm/rev) grown at $T_S = RT$.

3.2.5 Comparison with MC simulations

As shown in the previous sections, the surface diffusion, determined by the substrate temperature $T_S$, influences the growth of ion beam sputter glancing angle deposited Si structures in terms of structure morphology, diameter, and film density. It is therefore interesting to compare those experimental results with GLAD simulations that incorporate the effect of variable surface diffusion.

Concerning the influence of $T_S$ on GLAD of Si nanostructures with continuous substrate rotation and different substrate rotational velocities, the most outstanding morphological changes occur at low and intermediate $\rho$ values, such as the transition from films consisting of merged, broad structures to films of densely packed, separated, fibrous spirals with delayed merging at increased $T_S$. 

---

**Figure 11** (online color at: www.pss-b.com) Illustration of the surface diffusion along the sides of the single fibers and the evolution of the fiber diameter with time at (a) low and (b) high $T_S$. (c) Top-view micrographs of experimental results at $\rho = 100$ nm/rev. Highlighted in white are already merged structures at $T_S = RT$ after $t_{dep} = 30$ min. In (d), the PSD function curves calculated from the SEM micrographs in (c) are shown. Whereas at $T_S = 300$ °C, the maximum peak position remains roughly constant with $t_{dep}$ (dashed lines), at $T_S = RT$ there is a clear shift toward lower frequencies with increasing $t_{dep}$. 

---
Simulating the influence of surface diffusion on the GLAD of STFs at slow substrate rotation \( \omega \) with the MC simulation code of Karabacak et al. [30] partially reflects the experimentally gained results in a qualitative way. Briefly, the three-dimensional (3D) MC simulations include an oblique deposition angle \( \theta \), substrate rotation, and surface diffusion. The substrate rotation is simulated with a change of the azimuthal angle between successive particles sent one after another. Furthermore, the model allows sidewall sticking to simulate truly 3D structures. Surface diffusion is simulated by randomly choosing a surface or bulk atom within the vicinity of the impact point of the deposited particle and letting this atom diffuse to another nearest neighbor location by “hopping” from lattice point to lattice point. When the diffusing atom comes to rest, this DS is repeated with a new atom until an assigned number of DS is made. An alteration of the ratio \( \rho = r/\omega \) of vertical deposition rate \( r \) to substrate rotational speed \( \omega \) can be incorporated in the simulations by changing the number of particles per rotation (NPR). Additionally, different film heights (and therefore deposition times) can be simulated by changing the number of deposited particles (NOP). Details of the MC simulation approach can be found in previously published studies [30, 46, 47].

Figure 12 shows the simulated evolution of helical glancing angle deposited nanostructures with a deposition angle \( \theta = 85^\circ \) and different numbers of DS. It is observable that similar morphological changes as seen in the experiment take place in the simulations. Low surface diffusion (DS = 10) leads to the growth of bundled structures without clear boundaries, whereas high surface diffusion (DS = 300 and 1000) fosters the growth of STFs that consist of separated spirals with smaller diameters as compared to the low surface diffusion case.

Just as in the experiment, the overall density of the film decreases with increasing DS, which is reflected in a decrease of the film height by approximately 28% from DS = 10 to DS = 300 for otherwise constant deposition parameters (in the experiments with \( \rho = 100 \text{ nm/rev} \), the film height decreases by approximately 30% from \( T_S = RT \) to \( T_S = 300 \text{ °C} \)). Therefore, the simulations support the assumptions of the simple growth model developed in the previous section at least partly. Both, simulations and experiments indicate a surface-diffusion driven morphological change of the growing nanostructures from broad, merged structures to an STF of densely packed, smooth spirals with equal diameters. However, the comparison between experiment and simulation provides similar results only for a certain range of \( T_S \) or, respectively, DS. The experiments show that for both, slow (\( \rho = 175 \text{ nm/rev} \)) and intermediate (\( \rho = 100 \text{ nm/rev} \)) substrate rotational speeds, an increase of \( T_S \) to temperatures above approximately 300 °C results in the formation of merged, broad screw-like structures with low \( h_{\text{crit}} \) values being in the range of \( h_{\text{crit}} \approx 115 \text{ nm} \), as can be seen in Fig. 6. This early broadening effect, being attributed to the surface diffusion length becoming sufficiently large to overcome the distances between single spiral fibers right in the beginning of the growth process, cannot be seen in the simulations. Figure 12 shows that if DS is increased from 300 to 1000 DS, the film thickness and therefore film density remains constant, whereas the film itself remains consisting of separated, fibrous spirals. Obviously, other effects not taken into account in the simulation but existent in the experiments, such as the incident kinetic energy of the sputtered particles, influence the growth of the structures as well.

### 3.3 Spring constants of Si nanospiral arrays

When the ratio of vertical deposition rate to substrate rotational speed \( \rho = r/\omega \) is adjusted to be larger than approximately 120 nm/rev, STFs comprising spiral-like Si structures (helices with open core) are deposited on the substrate, as already described in previous sections of this report. Such spiral-like STFs, if flexible enough to withstand defined loads without plastic deformation, could be promising candidates for a variety of applications in the sensing domain, for example, as electromechanical actuators [48]. This motivates the examination of their mechanical behavior, especially in comparison to macroscopic spirals, as will be presented here in terms of the spirals spring constant \( k \).

![Figure 12: Change of structure morphology for GLAD simulations with total number of deposited particles NOP = 5 \times 10^7 and number of particles per rotation NPR = 1 \times 10^7 and different number of DS. Top row: top view, bottom row: cross-section.](image)

![Figure 13: Cross-sectional SEM micrographs of a 4-turned Si spiral-like STF (sample A) (a) and a 13-turned Si spiral-like STF (sample B) (b). The micrograph in the center shows one spiral of sample A in enlargement to underline the parameters needed for the evaluation of the theoretical values of the spring constant \( k_{\text{theo}} \): spiral arm diameter, \( W \): lateral structure expansion (i.e., outer spiral diameter), \( \beta \): inclination angle.](image)
Table 1 Dimensions of two helical Si STF samples used for the determination of their spring constants.

<table>
<thead>
<tr>
<th>sample</th>
<th>h (nm)</th>
<th>w (nm)</th>
<th>W/2 (nm)</th>
<th>β (°)</th>
<th>γ</th>
<th>n (μm²)</th>
<th>k_{exp} (N/m)</th>
<th>k_{theo} (N/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>2850 ± 50</td>
<td>160 ± 50</td>
<td>260 ± 25</td>
<td>58 ± 3</td>
<td>4</td>
<td>16 ± 4</td>
<td>64 ± 12</td>
<td>5.9</td>
</tr>
<tr>
<td>B</td>
<td>8400 ± 50</td>
<td>250 ± 70</td>
<td>300 ± 50</td>
<td>58 ± 3</td>
<td>13</td>
<td>10 ± 4</td>
<td>107 ± 35</td>
<td>7.1</td>
</tr>
</tbody>
</table>

h, spiral height; w, spiral arm diameter; W, lateral structure expansion (i.e., outer spiral diameter); β, inclination angle; γ, number of turns; and n, areal structure density (number of spirals per μm²). The resulting spring constants in experiment and theory are labeled k_{exp} and k_{theo}.

To analyze the elastic deformation of spring-like Si STFs, two samples comprising spirals with different structure parameters have been deposited with GLAD: one 4-turned spiral-like sample of total height \( h = 2850 \pm 50 \) nm (sample A) and one 13-turned spiral-like sample of total height \( h = 8400 \pm 50 \) nm (sample B). Figure 13 shows both samples in cross-section.

Load–displacement curves at four different maximum forces \( F_{\text{max}} \) = 100, 200, 300, and 400 mN were taken from both samples by means of nanoindentation with a flat punch indenter, at 20 different spots for each applied \( F_{\text{max}} \) on both samples. The spring constants \( k_{\text{exp}} \) were deduced from the contact stiffness \( S = dF/dh_{\text{dis}} \) (i.e., the slope of the unloading curve in the point of maximum displacement \( h_{\text{dis}} \) using \( k = S/A_{\text{c}}n \), where \( A_{\text{c}} \) is the contact area of the indenter with respect to the sample surface, and \( n \) is the areal structure number density of the spring-like nanostructures. The results for both samples are shown in Table 1.

Experimentally, the 4-turned Si spirals of sample A exhibit a spring constant of \( k_{\text{exp}} = (64 \pm 12) \) N/m, whereas the 13-turned Si spirals of sample B show a value of \( k_{\text{exp}} = (107 \pm 35) \) N/m.

For macroscopic springs, the following formula containing the material parameters (Young’s modulus \( E \) and Poisson ratio \( ν \)) as well as the morphological sample parameters (arm diameter \( w \), lateral structure expansion (“coil diameter”) \( W \), structure inclination angle \( β \), and areal structure density \( n \)) can be used to calculate the spring constant \( k_{\text{theo}} \): \([49–51]\):

\[
k_{\text{theo}} = \frac{Gw^4}{8W^3n} \left( 1 - \frac{3w^2}{16W^2} + \frac{3 + ν}{2(1 + ν)} \tan^2(90° - β) \right)^{-1}.
\]

In this formula, the shear modulus \( G = E/[2(1 + ν)] \) is used. With the use of \( E = (160 \pm 5) \) GPa (a value experimentally obtained by nanoindentation on a dense, amorphous Si film deposited on a Si substrate under normal particle incidence \( θ = 0° \)), \( ν = 0.278 \) [52], and the morphological sample parameters as listed in Table 1, the theoretical spring constants can be calculated.

Following the above equation, the Si spirals of the 4-turned sample A should exhibit a spring constant of \( k_{\text{theo}} = 5.9 \) N/m, whereas the 13-turned Si spirals of sample B should have a value of \( k_{\text{theo}} = 7.1 \) N/m.

Comparing experiment and theory, it is obvious that although the trends concerning \( k_{\text{exp}} \) and \( k_{\text{theo}} \) are the same when comparing sample A with sample B (the 13-turned spirals of sample B are stiffer than the 4-turned spirals of sample A, exhibiting higher \( k \) values in both theory and experiment), the absolute values of \( k_{\text{exp}} \) and \( k_{\text{theo}} \) are differing in about one order of magnitude for both samples. However, those discrepancies in the spring constant values can be understood as follows: on the one hand, the third and fourth order power terms in the above equation demand a precise knowledge on the spiral dimensions, which cannot be given, as, for example, neighbored spirals are not uniform in diameter on unpatterned substrates, and the diameter of the spiral arm \( w \) for a single spiral is not constant, but increases with increasing structure height. Within the error bars of \( w, W, β, \) and \( n \), the theoretical values thus vary between \( (0.8 ≤ k_{\text{theo}} ≤ 27.1) \) N/m (sample A), or between \( (0.9 ≤ k_{\text{theo}} ≤ 41.6) \) N/m (sample B), respectively. On the other hand, it is likely that the spirals in the STFs do not stay separated, but touch each other under mechanical load. Thus, when experimentally determining \( k \) with nanoindentation, the resulting contact stiffness \( S \) cannot be seen as sum of the \( k_{\text{exp}} \) values of \( n \) independently reacting spirals, but should rather be seen as reaction of a whole “bed” of interconnected springs.

When trying to determine the spring constants of glancing angle deposited STFs consisting of spring-like SiO structures, Seto et al. [49] found \( k_{\text{exp}} \) values that are larger than the theoretically expected values \( k_{\text{theo}} \) in about one order of magnitude as well. Therefore, the observed discrepancies in theoretically expected and experimentally gained spring constant values are not surprising.

4 Conclusions GLAD of Si by ion beam sputter deposition proved to be an auspicious technique for the deposition of complex nanostructures in a straightforward bottom-up deposition process. The ability to tailor the structure morphology by adjusting the ratio \( ρ = r/ω \) of vertical deposition rate \( r \) to substrate rotational speed \( ω \) has been demonstrated. The influence of the substrate temperature \( T_S \) on the growth and morphological evolution of the OAD- and GLAD-grown Si STFs has been evaluated. Increasing \( T_S \) when depositing inclined columnar structures with the OAD concept has been found to lead to an increase in the column inclination angle, thus supporting already existing models that predict this behavior [42]. When increasing \( T_S \) while performing GLAD of Si with constant azimuthal substrate rotation at different substrate rotational speeds \( ω \), it was found that certain \( T_S \) windows exist that enable the deposition of helical STFs comprising fibrous Si
nanospirals with diameters in the 20–30 nm range, that do not show the typical, RT-GLAD inherent effects of structure broadening and merging. A growth model, explaining this behavior in terms of enhanced adatom diffusion with increased $T_S$, is backed by 3D MC simulations of GLAD at slow substrate rotational speed with different numbers of simulated DS. It could also be shown that the prediction of the mechanical behavior of GLAD-grown helical STFs is not possible in an easy way (e.g., applying an equation that is valid for macroscopic, separated spirals, on the nanoscopic spiral-like STFs) and should rather be done experimentally to obtain reliable results.

Acknowledgements The authors would like to acknowledge Dr. J. W. Gerlach for the fruitful discussions, H. Neumann for his help with the ion beam technique, and Prof. Dr. T. Höche and D. Hirsch for their assistance at the SEM. This work is supported by the project P3 within the DFG research group FOR 522 “Architecture of nano- and microdimensional structure elements,” grant number RA 641/21-2.

References