Large area patterning using interference and nanoimprint lithography

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ABSTRACT

Interference lithography (IL) is the best suited technology for the origination of large area master structures with high resolution. In prior works, we seamlessly pattern areas of up to 1.2 x 1.2 m² with periodic features, i.e. a diffraction grating with a period in the micron range. For this process we use an argon ion laser emitting at 363.8 nm. Thus, feasible periods are in the range of 100 µm to 200 nm. Edge-defined techniques or also called (self-aligned) double patterning processes can be used to double the spatial frequency of such structures. This way, we aim to reduce achievable periods further down to 100 nm. In order to replicate master structures, we make use of nanoimprint lithography (NIL) processes. In this work, we present results using IL as mastering and NIL as replication technology in the fields of photovoltaics as well as display and lighting applications. In photovoltaics different concepts like the micron-scale patterning of the front side as well as the realization of rear side diffraction gratings are presented. The benefit for each is shown on final device level. In the context of display and lighting applications, we realized various structures ranging from designed, symmetric or asymmetric, diffusers, antireflective and/or antiglare structures, polarization optical elements (wire grid polarizers), light guidance and light outcoupling structures.

Keywords: interference lithography, micro replication, nanoimprint lithography, solar cells, wire grid polarizer

1. INTRODUCTION

The patterning of surfaces on the micro- and nanoscale is of importance for markets such as the manufacturing of solar cells or displays. Optical effects related to such surfaces are the manipulation of light propagation, reflection or absorption. In solar cells, a patterning of surfaces is applied to maximize light in-coupling, internal light path lengths and hence the total absorption within the semiconductor material [1]. In industrial fabrication, textures applied to silicon solar cells are typically the size of several micrometers and are realized masklessly and are thus stochastically arranged. In research, there are various concepts for realizing highly efficient textures based on lithographic techniques ranging from the microscale [2–6] down to the sub-micron regime [7–10]. The successful transfer of such concepts requires up-scalable high-resolution patterning techniques. We are working on nanoimprint lithography (NIL) processes in order to open up an industrially feasible pattern replication technology [11].

Especially in display applications, very large areas have to be patterned homogeneously and seamlessly as stitching processes cannot meet the given requirements. Examples for optically active films in displays are e.g. brightness enhancement films (BEF) [12] as well as antiglare, antireflection and diffuser structures [13]. Another essential optical functionality required in displays is provided by polarizers. There, excellent performance can be achieved by wire grid polarizers; however, especially when to be applied in the visible range, the pitch of metallic lamellae has to be very small (< 100 nm) [14]. Thus, for the fabrication of such structures solely processes like electron beam lithography or laser interference lithography and often additionally so-called edge defining techniques or double patterning approaches are applied to reduce the pitch of the linear gratings [15,16]. Electron beam lithography is typically not applicable for the patterning of very large areas. Even though interference lithography (IL) is in principle capable of patterning very large areas [17], largest reported sample sizes for the combination of IL and double patterning was only up to 5.5 x 5.5 cm² [14]. Most approaches making use of such elaborate mastering processes rely on nanoimprint lithography processes as replication technology in order to compensate high initial costs [18].

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In this work, we present interference lithography and nanoimprint lithography as versatile and powerful technologies, which allow the manufacture of sophisticated surface patterns. The basics of these techniques as well as some technological achievements are described in the section micro- and nanopatterning techniques. In the subsequent section, some applications in the context of photovoltaics are shown, which lead to an improvement compared to the state of the art. Finally, we present first results of our efforts to combine interference lithography and double patterning with the aim of originating master structures with periods around 100 nm on very large areas.

2. MICRO- AND NANOPATTERNING TECHNOLOGIES

2.1 Interference lithography

In interference lithography, coherent laser beams are superimposed on a sample. The resulting interference pattern is applied to expose a photoresist and thereby induce selectively a photochemical modification. After a wet chemical development process, a surface relief is originated, which is a function of the interference pattern. However, it is not necessarily linearly proportional to the intensity distribution. As a result of the interference of waves, the intensity distribution by definition shows a sinusoidal shape. Still, as a result of strongly varying photochemical behavior of resists and by making use of substrate interfaces as structural element a huge variety of profile shapes can be originated [17,19,20].

We use an argon ion laser emitting at a wavelength of 351.1 nm or 363.8 nm. Hence, minimum achievable periods are around 180 nm in theory and around 200 nm in practical terms [17]. One of the main advantages of interference lithography is the broad range of structures and dimensions that can be originated. The principle setup for interference lithography including possible modifications is sketched in Figure 1. Examples of achievable structures in photoresist are shown in Figure 2.

![Figure 1: Schematic view of the basic set-up for two beam interference lithography. The light blue arrows indicate possible set-up modifications leading to different pattern periods (angle between beams), a modulation in different spatial directions (sample rotation between exposures) or the origination of aperiodic features (multi-wave interference by adding a diffuser into a beam).](image-url)
Figure 2: Examples of photoresist structures originated by interference lithography:
(a) linear grating with a period of 230 nm and a depth of 380 nm,
(b) hexagonal “honeycomb” structure with a period of 8 \( \mu \text{m} \), originated by three wave interference,
(c) aperiodic binary structure with defined spatial frequency distribution,
(d) ultra-hydrophobic structure composed of needles (period 2 \( \mu \text{m} \)) and an underlying wavy structure (period 30 \( \mu \text{m} \)).

Another key strength of interference lithography is that very large areas can be patterned seamlessly; however, this requires the phases of the beams to be kept stable relative to each other over potentially very long exposure durations. The largest sample area we patterned with a diffraction grating had a size of 1.2 x 1.2 m\(^2\). Given that exposure times for such sample areas are in the range of several hours, this can only be possible using a closed-loop control of the relative phases (fringe locking) to keep the interference pattern stable in the sample plane [17].
2.2 Nanoimprint lithography

The term nanoimprint lithography (NIL) was introduced by Chou et al. in 1995 [21]. Especially NIL processes relying on UV-curing resist materials are promising in terms of process velocity [22]. We are using various setups for UV-NIL processes ranging from a rudimentary setup, where pressure is applied to a stamp via air pressure [23], over a commercially available Smart-NIL™ tooling from EVG integrated into a mask aligner [11], to an own development of a roller-NIL tooling being very well suited for imprinting on very thin silicon wafers [5].

We use soft stamp materials for the NIL, to be able to pattern large areas with homogeneous residual layers [24]. Depending on feature sizes either soft stamps made of standard polydimethylsiloxane (PDMS) or composite stamps made of hard PDMS, carrying the surface structure, and a soft PDMS bulk can be used [25]. For each tooling described above, stamps are differently mounted (bonded on glass substrates or foils or directly wrapped around a roller).

When it comes to the integration of NIL processes into large scale industrial process chains, a critical issue will be the degradation of the stamps. We assume the molar weight of pre-polymers in the resist as an important factor for the stamp lifetime. An imprint resist with very short polymer chains led to failures after approximately 10 imprints. Additionally, we tested two other resist materials with different molar weights. Confirming our assumption, the one with a slightly higher molar weight led to around 70 successful imprints and for the one with the highest molar weight, we could not detect any degradation after 100 imprints. Currently we are working on a characterisation method to quantify the effect of diffusion into the stamp and thus strengthen the assumption stated above. In the photovoltaics industry the throughput of toolings is supposed to exceed 3000 wafers per hour [26], therefore the number of imprints per stamp has to be raised considerably or the manufacture and changing of stamps has to be cheap, easy and very fast.

3. APPLICATIONS

3.1 Solar cell applications

Looking at the state-of-the-art in industrial fabrication of silicon solar cells, especially multicrystalline silicon (mc-Si) solar cells suffer from a non-optimal surface texturing. There, one cannot make use of a unique crystal orientation and related anisotropic etching in alkaline solutions leading to pyramidal textures in <100> monocrystalline silicon wafers. Therefore, typically isotropic etching in acidic solutions is applied to generate a so-called isotexture [27]. This isotexture allows a homogeneous appearance; however, achieved optical properties are rather moderate. A quantity describing the
performance of a front side texture in solar cells is the weighted reflectance. The weighting in this case is applied considering the photon flux of the AM1.5g solar spectrum [5]. Values of the weighted reflectance for the isotexture are around 25% or even above without and around 6% with a silicon nitride antireflection coating. As comparison, the state-of-the-art in monocrystalline silicon is around 11% and between 1 and 2%, respectively. Former world record efficiencies for mc-Si made use of the so-called honeycomb texture [2,28]. Therefore, there has been some activity in various research groups to find adequate processes to originate a honeycomb texture on multicrystalline silicon using e.g. laser ablation [4], laser ablation of etching masks [3], inkjet masking [6] or NIL [5] as alternative processes to photolithography.

The basic process chain for the realization of the honeycomb texture with the processes of interference lithography, NIL and plasma etching is described thoroughly in [5]. In the following, the process chain is described briefly and recent results published in [29] are summarized. We used three beam interference lithography to originate large area (25x25 cm²) master structures having a hexagonal symmetry and a pitch of 8 µm. From these master structures first so-called nickel shims, fabricated by electroplating from external suppliers, and then from these shims working stamps made of PDMS were replicated. Using such PDMS stamps, a UV-curable resist was patterned on large area mc-Si substrates (156 x 156 mm²), which was subsequently applied as etching mask in a plasma etching process. Finally, the surface was smoothened using wet chemical etching. Figure 4 shows a scanning electron microscopy (SEM) micrograph of the resulting texture. Also shown in Figure 4 is the result of a hybrid process chain consisting of interference lithography, photolithography as well as replication technologies in order to do a re-mastering [11]. In the re-mastered structure both the periodic features realised by interference lithography and written features (e.g. a contact grid geometry) are defined by photolithography. After a stamp replication both feature types are transferred within on NIL process.

![Figure 4: Left: SEM micrograph showing a honeycomb textured surface of a multicrystalline silicon substrate. Right: Here, additionally a spacing for a contact finger was already introduced into the master structure, so that both features, the honeycomb texture as well as the geometry for the contact grid, are defined in one NIL step. In both cases the primary mastering was done using interference lithography. [11]](image)

Optically this honeycomb texture reached excellent values for the weighed reflectance of 6% without and around 1% with antireflection coating. These values are well below the ones for the isotexture, which is the benchmark on mc-Si, and even superior to pyramidal textures on monocrystalline silicon as they are described above. Mc-Si solar cells were fabricated using screen printing processes and an aluminium back-surface-field (Al-BSF) cell structure as described in [29]. The optical gain was also demonstrated on device level, where the better optical performance is transferred to a gain in short circuit current density (Jsc) of 1.3 mA/cm². Unchanged electrical properties are reflected in constant values for the open circuit voltage. Thus the optical gain directly leads to a gain in solar cell efficiency of 0.5 percent absolute to 17.8%.

Another optical concept, relying on smaller periodically arranged features, is the realization of rear side diffraction gratings to enhance internal light paths and thus the light trapping in solar cells [10]. Again we applied the process chain comprising of NIL and plasma etching to realize such a grating. The period of the grating was chosen to be 1 µm according Ref. [30]. The whole process chain for solar cell fabrication as well as the embedding of the patterning...
processes and results are thoroughly described in [31]. This concept of rear side gratings is especially interesting for very thin solar cells as they suffer from incomplete absorption in the near infrared regime and thus light trapping gains in importance. In [31], this is demonstrated theoretically and experimentally for solar cell thicknesses of 250 µm, 150 µm and 100 µm. The processing of such thin silicon wafers of course is very demanding especially when thinking about a mechanical moulding process as for NIL. We succeeded in patterning etching masks for rear side gratings on wafers as thin as 50 µm using the Smart-NIL as well as our in-house developed roller-NIL tooling. A photograph of such a very thin and thus flexible silicon wafer is shown in Figure 5. Also shown is an SEM of the resulting pattern after plasma etching [11]. The grating induced gain in J_{sc} for such structures is in the range of 1.2 mA/cm² for solar cells with a planar front and a thickness of 250 µm [32]. The corresponding efficiency enhancement for these cells is 1.1 % absolute. For only 100 µm thick cells the grating induced gain in J_{sc} increases to 1.8 mA/cm².

Figure 5: Left: Photograph of an imprinted crossed grating with a pitch of 1 µm on a 50 µm thick 4´´ monocrystalline silicon wafer. The bowing of the wafer highlights its flexibility. Right: SEM micrograph showing the pattern in silicon after a plasma etching process. [11]

3.2 First steps for double patterning using very large area toolings

The origin of double patterning techniques is related to consequent miniaturization in microelectronic applications [33]. The use of such techniques to originate sub-100 nm periodic gratings was demonstrated in several publications before [15,16,34]. One quite obvious application is the origination of wire grid polarizers for display applications. Still, this technology has not been reported on very large area. The very basic idea of double patterning is sketched in Figure 6.

Figure 6: Sketch of the intended process chain for double patterning in order to double the spatial frequency.

We demonstrated the technology of interference lithography being capable of realizing seamless periodic patterns up to the square meter scale. Having a patterned resist layer on this substrate area of course for a successful double patterning an adequate deposition technology is required. In [15] the different deposition techniques evaporation, sputtering and
atomic layer deposition (ALD) are compared concerning their feasibility for double patterning. It is concluded that ALD is best suited because of its conformal deposition, which is especially important on the steep sidewalls. A drawback typically connected with ALD is the very low deposition rate, which might not even be critical here. More critical is the maximum substrate size we are aiming for. To open up an up-scalable process route we intend to use a sputtering tool, as it is applied in industry for the coating of architectural glass. The tooling we use is capable of processing sample areas of up to 1.5 x 4 m². Besides the deposition also large area plasma etching is required. As the large area deposition tool has also the possibility to be used for sputter etching, both processes can be carried out in the same tool.

In very first tests, we used interference lithography to originate linear gratings in AZ-MIR701 photoresist with a period of 300 nm. Then we tested sputter coating processes using different target materials like e.g. Cr, Ti, TiO₂, Pt and Au. In a next step we performed sputter etching tests using argon as well as a mixture of argon and oxygen as etching gases. As it can be seen in Figure 7, up to now we did not fully succeed in setting up a complete double patterning process. This can be rather regarded as first steps of developing this process chain on very large area toolings.

As next steps for this double patterning sequence, we will optimize the homogeneity of the hard layer deposited by sputter coating. However, most importantly we have to optimize the plasma etching process in order to achieve a high directionality of the etching process and investigate potential influences for the pattern stability.

4. CONCLUSION AND OUTLOOK

We have shown interference lithography and nanoimprint lithography as versatile large area patterning techniques. As application examples, we motivated solar cells and very high resolution structures for wire grid polarisers. In solar cells, we already succeeded in introducing defined periodic structures and thereby enhance conversion efficiency. Both for honeycomb textures on multicrystalline silicon as well as rear side diffraction gratings a gain was demonstrated on device level. For the second application, the realization of very high resolution patterning using the double patterning technique, we have only shown the first steps. However, already in this early stage of development we applied toolings being capable of processing very large areas. We will continue to work on these processes in order to originate master structures with periods around 100 nm on large areas. The replication of such structures using NIL processes also will be demanding and will be the scope of future work.

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