Increase in Generation of Poly-Crystalline Silicon by Atmospheric Pressure Plasma-Assisted Excimer Laser Annealing

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Abstract: In this work, a novel atmospheric pressure plasma-assisted excimer laser annealing method for increasing the generation efficiency of poly-crystalline silicon from amorphous silicon layers is presented. Here, both the plasma and the laser propagate coaxially in order to generate energetic synergies. The influence of different process gases and plasma discharge modes as well as the working distance were investigated. Depending on the particularly applied plasma, the crystalline area was increased by a factor of approx. 1.1 to 1.9, where the highest efficiency was observed when introducing an argon plasma beam to the annealing process.

Key words: Excimer laser annealing, laser-plasma processes, amorphous silicon, polycrystalline silicon, display technology.

1. Introduction

For a sensitive representation of images, modern flat screen displays are based on thin film transistors (TFT), where each pixel is supplied with variable voltages. The cost-efficient realisation of the required integrated circuits is achieved by the so-called system on glass (SOG) technique. Here, amorphous silicon (α-Si), which is gained from gaseous silane (SiH₄), is deposited onto a glass substrate. This α-Si layer is then converted into polycrystalline silicon (p-Si) in order to fabricate semiconductor channels [1].

Due to the comparatively low softening point of the used glass substrates, resulting in a thermal limitation of the process of $T_{\text{max}} \approx 500-650$ °C, low temperature poly silicon technology (LTPS) is applied [2]. Here, the application of pulsed excimer laser irradiation affects a short-time melting of the silicon layer without any significant heating of the glass substrate. In this context, several processes for large-scale crystallisation based on specifically shaped laser beams such as excimer-laser-annealing (ELA) allow the generation of large crystallite spots by single- or multi-pulse annealing [3].

In the case of ELA, the applied laser fluence $\Phi$ corresponds to the fluence which is required for melting the entire α-Si layer. Here, the silicon layer is molten except of minor polycrystalline zones at the silicon-glass interface. Starting at these zones, the lateral and columnar crystallisation, i.e. the generation of p-Si, takes place. As a result, the crystallites exceed the layer thickness of the initial silicon layer. This effect is known as super lateral growth (SLG) [2, 4], resulting in a high electron mobility within the p-Si layer and thus enabling faster TFT switching times [5]. ELA is applied for the production of active matrix liquid crystal display (AM-LCD)- or active matrix...
organic light-emitting diode (AMOLED)-TFT and is performed by the use of a xenon chloride (XeCl) excimer laser with a laser wavelength $\lambda$ of 308 nm and an average power $P$ of 315 W. By beam shaping optics, the raw beam is homogenised and converted to a line beam. The fluence within the line focal area amounts to approx. 350 mJ/cm² [1, 6].

The processing of p-Si TFT by the use of ELA at $\lambda = 308$ nm is a critical step in the fabrication of flat-screen displays. The demands on displays of larger size such as Gen 8 (2,200 × 2,500 mm) as well as rapidly growing markets are a motivation for display manufacturers to apply new, more efficient technologies. In this field of action, production costs are a direct function of throughput. The crystalline quality, variation in grain size and uniformity are the influencing variables of high quality p-Si TFT. Further, the laser power represents the physical and economical limitation. In this work, we present an atmospheric pressure plasma-assisted excimer laser annealing (APP-ELA) method for increasing the efficiency of generation of poly-crystalline silicone [7].

For this purpose, a laser-plasma common-path setup was assembled in order to allow a simultaneous treatment of $\alpha$-Si-coated substrates with both excimer laser irradiation and different atmospheric pressure plasmas. In previous work, it was already shown that such a hybrid setup for simultaneous laser-plasma ablation allows significant enhancements in comparison to a pure laser ablation process [8-10].

2. Materials and Methods

In order to investigate the influence of assisting atmospheric pressure plasmas (APP) on the efficiency of ELA processes, a hybrid experimental setup as represented in Fig. 1 was realised. Here, a XeCl excimer laser LEAP 130C from Coherent GmbH with a wavelength of 308 nm was used as laser source.

From the raw beam, a nearly flat-top sub-aperture beam was generated by introducing a diaphragm with an inner diameter of 2 mm to the laser outlet. This sub-aperture beam was then focused by a convex lens with a focal length of 100 mm, resulting in a focus fluence $\Phi$ of $75.95 \pm 2.55$ mJ/cm².

The focused laser beam was guided coaxially to the particularly applied plasma which was made possible by the use of a cone-shaped plasma source with an inner hollow-core high-voltage (HV) electrode. Depending on the circuitry of both the HV electrode and the ground electrode, this plasma source can be operated in two different modes [11]: First, a dielectric barrier discharge (DBD)-based atmospheric pressure plasma jet can be generated by grounding the internal ground electrode (Fig. 1); second, introducing an external ground electrode to the setup, where the sample itself acts as dielectric for realising a DBD, results in the formation of a fine plasma filament, the plasma beam, with a diameter of approx. 200 µm which is stabilised by the process gas flow (Fig. 1). Experiments were performed using both argon (Ar) and forming gas 90/10 (90% nitrogen (N₂) and 10% hydrogen (H₂)) as process gas. In the case of Ar, both operation modes of the plasma source were investigated whereas for forming gas 90/10, no stable plasma beam was obtained due to the high required ignition voltage in comparison to Ar. The power dissipated into the plasma amounts to approx. 1.9 W in both Ar-operation modes, whereas in the case of forming gas, 0.69 W were implemented.

Besides the mode of operation and the process gas, the working distance $d$ of the $\alpha$-Si-coated samples...
from the plasma source outlet was varied. Experiments were performed in order to identify the most suited set of parameters for a high improvement in efficiency of the plasma-assisted ELA-process. For this purpose, the resulting crystalline areas $A_{\text{crys}}$ of p-Si were compared as the significant key quality parameter. This consideration was carried out by the use of a light microscope Axioskop 2 MAT from Zeiss.

3. Results and Discussion

3.1 Forming Gas Jet APP-ELA

When introducing a forming gas APP jet to the ELA process, where $\Phi$ was $78.5 \pm 1.1 \text{ mJ/cm}^2$, the resulting crystalline area was measurably enlarged in comparison to pure ELA as shown in Fig. 2.

Here, the averaged quotient of the crystalline areas that were achieved by APP-ELA and pure ELA $A_{\text{crys}}(\text{APP-ELA})/A_{\text{crys}}(\text{ELA})$ was 1.09.

3.2 Argon Jet APP-ELA

As shown in Fig. 3, the crystalline area was further increased when applying APP-ELA using an assisting argon jet APP. Here, $\Phi$ amounted to $75.3 \pm 2.2 \text{ mJ/cm}^2$.

By this configuration, an averaged factor $A_{\text{crys}}(\text{APP-ELA})/A_{\text{crys}}(\text{ELA})$ of 1.57 was obtained. However, the plasma-induced increase in crystalline area shows a strong dependency on the working distance $d$ which can approximately be described by a linear interrelationship.

3.3 Argon Beam APP-ELA

By the combination of both ELA, where $\Phi$ was $73.4 \pm 0.9 \text{ mJ/cm}^2$, and an assisting argon beam APP, a significant increase in crystalline area was realised as shown in Fig. 4.

For this combined APP-ELA process, a considerable factor $A_{\text{crys}}(\text{APP-ELA})/A_{\text{crys}}(\text{ELA})$ of 1.90 was achieved. As a result of the applied discharge mode, nearly no dependency of the plasma-induced
increase in crystalline area on the working distance was observed. The plasma-induced effect is visualised by the comparison of the measured crystalline areas for pure ELA and argon beam APP-ELA in Fig. 5.

This comparison further shows that the pure argon plasma beam does not affect the sample surface in terms of crystallisation processes.

3.4 Discussion

The above-presented results clarify the strong influence of the assisting plasma on the efficiency of the ELA process. As shown by the comparison of the particular percentaged plasma-induced increases in $A_{crys}$ in Fig. 6, the applied discharge mode and the choice of the process gas are the crucial factors for achieving an increased generation of poly-crystalline silicon by APP-ELA. The lowest efficiency was resulting when applying a forming gas jet APP, where an average increase in crystalline area $\Delta A_{crys}$ of 9.28% was achieved. It can thus be assumed that in this case, a marginal amount of excited species was generated from the compounds of the used process gas, i.e., nitrogen and hydrogen. This is explained by the fact that, according to Paschen’s law, the required ignition voltage $u_0$ of hydrogen and nitrogen is approx. 12.7-18 times higher with respect to argon.

Using argon as process gas, significantly higher averaged $\Delta A_{crys}$ of 53.67% (jet mode) and 89.71% (beam mode), respectively, were obtained. This increase in crystalline area when applying argon APP-ELA can partially be explained by plasma heating of the substrate surface and an accompanying reduction of the band gap. In order to verify such a heating, the temperature of the sample surface during pure argon plasma jet and beam treatment was determined by the use of an infrared camera Vario Cam from InfraTec/JENOPTIK. In jet mode, a maximum substrate surface temperature $T_{\text{max}}$ of approx. 38 °C was measured whereas $T_{\text{max}}$ amounted to approx. 88 °C when applying a plasma beam to the substrate. This heating corresponds to a reduction of the band gap $E_g(T)$ according to the Varschni equation:

$$E_g(T) = E_g(T = 0K) - \frac{\alpha T_{\text{max}}}{T_{\text{max}} + \beta}$$  \hspace{1cm} (1)

where $\alpha$, $\beta$ and $E_g(T = 0K)$ are material constants, of approx. 0.36% (jet mode) and 1.5% (beam mode), respectively [12].
However, since no crystallisation effect was observed for pure argon plasma beam treatment (Fig. 5), the actual increase in crystallisation turns out to be due to synergetic effects of both the particular APP and the incoming laser irradiation. Such behaviour was also observed in previous work where APP-assisted ablation of optical glasses [8], aluminium [9] and ceramics [10] was investigated. In this context, inelastic collision processes can be referred to as the dominating underlying effect. Within the plasma volume, excited argon atoms and metastable argon species, the latter with an energy $E$ of 11.5 eV, are generated. Following, these species are lost by radial diffusion and de-excitation at the substrate surface [13]. The involved energy transfer into the amorphous silicon layer is notably facilitated by the incoming laser irradiation and the accompanying heating and partial melting of the near-surface region. The observed strong dependency of the crystallisation efficiency on the working distance in the case of argon jet APP-ELA can thus be explained by a decreasing presence of argon metastables in direction of propagation of the argon plasma jet. This is also confirmed by the comparatively constant results for argon beam APP-ELA, where the argon metastable density can be assumed to be approximately constant over the whole length of the plasma beam. The marginal effect that was observed for forming gas jet APP-ELA further suggests the dominant influence of argon metastables on the presented method.

4. Conclusions and Outlook

The presented atmospheric pressure plasma-assisted excimer laser annealing method allows a significant increase in generation of poly-crystalline silicon. It was shown that in comparison to pure laser annealing, the crystalline area is nearly doubled when applying an argon beam APP. This effect is of high interest for a number of applications such as display technology or the production of photovoltaic cells since atmospheric pressure plasmas stand out due to a high cost- and energy-efficiency and thus contribute to sustainable industrial processes. As an example, the presented method could be used to reduce the required laser energy for laser annealing. In addition, due to the characteristics of dielectric barrier discharges, thermal influences on the substrate are minimised or even avoided.

In order to characterise the APP-ELA process in more detail, further experiments such as an investigation of the available plasma rotation temperatures and its influence on lattice vibrations in silicon and hence interband transitions of electrons during photon absorption will be carried out in future work. The influence of a temporal synchronisation of both the plasma and laser pulse will be investigated, too. Further, first approaches for an improvement of the presented method by up-scaling the plasma discharge area and adapting it to a large-scale laser line focal area are currently performed.

In terms of energetic considerations, APP-ELA offers a higher efficiency than pure ELA. For instance, in order to crystallise an area of 0.166 mm², a laser fluence of 118 mJ/cm² was required for ELA whereas in the case of APP-ELA, the same effect was achieved when applying a laser fluence of 74 mJ/cm². In commercial industrial scale, ELA is performed by using laser pulses with a pulse energy of 1,050 mJ at a repetition rate of 300 Hz and a crystallisation speed of 28 cm²/s [1]. This corresponds to a laser power of 315 W. For the presented APP-ELA method, crystallisation can already be achieved at a laser power of 196 W, corresponding to a reduction factor of 1.6.

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