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Cite as: Appl. Phys. Lett. **89**, 151907 (2006); https://doi.org/10.1063/1.2358922
Submitted: 17 April 2006 • Accepted: 16 August 2006 • Published Online: 11 October 2006

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Arbitrary surface structuring of amorphous silicon films based on femtosecond-laser-induced crystallization

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(Received 17 April 2006; accepted 16 August 2006; published online 11 October 2006)

The arbitrary surface structuring of amorphous silicon (*a*-Si) films was performed by applying the Fourier-transform (FT) method to the femtosecond-laser-induced crystallization. In order to realize the arbitrary structuring, the logo q-Psi was produced in the *a*-Si film by the FT of a computer-generated hologram. The crystallization of *a*-Si was performed using the near-infrared femtosecond-laser pulses. By micro-Raman spectroscopy, scanning-electron microscopy, and transmission-electron microscopy, it was found that the femtosecond-laser pulses induced a localized phase transformation from the amorphous to the crystalline phase, and the spatially selected crystallization of the *a*-Si was responsible for the formation of the two-dimensional pattern. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2358922]

Substantial attention has been focused on amorphous silicon (*a*-Si) and its crystallization for use in active-matrix-based flat-panel displays such as active-matrix liquid-crystal displays and active-matrix organic-light-emitting diodes.[1] Since crystalline silicon (*c*-Si) has larger carrier mobility, faster switching, and higher stability than *a*-Si, the spatially selected crystallization of *a*-Si could lead to a localized control of electrical properties.[2] The crystallization of *a*-Si is typically obtained by laser annealing[3] or solid-phase crystallization.[4] If multiple-beam interference or multi-axis laser-beam scanning is used to create a spatial variation of the laser intensity, laser annealing could result in a multidimensional patterning of the materials. Femtosecond-laser pulses can be used to modify the selected area of amorphous or crystalline silicon. Femtosecond-laser pulses provide an extremely high heating rate in a particular region of the material. This rapid energy deposition makes a laser-induced phase transformation of the amorphous or crystalline structure of the material possible.[5–8] Femtosecond-laser patterning, commonly used to fabricate particular patterns inside a bulk material or on a film surface, has attracted increasing interest due to various device applications such as gratings, waveguides, splitters, directional couplers, optical memory and photonic crystals.[9–12] Although several studies have been conducted on the excimer laser-based crystallization of *a*-Si,[13,14] there are very few reports on the femtosecond-laser-induced crystallization (FLIC).[15,16] Therefore, it is worthwhile to examine the FLIC of an *a*-Si film and its potential applications in micropatterning.

In this letter, we studied the arbitrary surface structuring of *a*-Si films based on the FLIC. In order to realize the arbitrary surface structuring of an *a*-Si film, the logo q-Psi of the Quantum Photonic Science Research Center was produced in the *a*-Si film by means of the Fourier transform (FT) of a computer-generated hologram (CGH). The crystallization of *a*-Si was performed using the near-infrared (NIR) femtosecond-laser pulses. The femtosecond-laser system used in this experiment was a regeneratively amplified Ti:sapphire laser (Spectra Physics, Hurricane) with an output wavelength of 800 nm, a pulse duration of 130 fs, a maximum pulse energy of 1.0 mJ, and a repetition rate of 1 kHz. Hydrogenated *a*-Si films were deposited onto a glass substrate (Corning 1737) by plasma-enhanced chemical-vapor deposition (PECVD) in a gas mixture of silane and hydrogen at a substrate temperature of 270 °C. The resulting film was 100 nm thick. The physical properties of the *a*-Si films created by the PECVD method are described elsewhere.[17,18] The surface morphology of the laser-modified region was investigated by optical microscopy and atomic-force microscopy (PSIA, XE-100). Scanning-electron microscopy (SEM; JEOL, JSM-6630F) was also employed to evaluate both the surface morphology and the crystallization properties of the laser-modified region. The structural properties of the laser-modified region were investigated by micro-Raman spectroscopy. The Raman spectrum for each sample was measured at room temperature using a 514.5 nm line from a stabilized argon-ion laser (Coherent, Innova 307) as the excitation source. In order to obtain the micro-Raman spectra, the laser beam was focused to a spot of 1–2 μm on the sample using a microscope objective (60×). Transmission-electron microscopy (TEM; JEOL, JEM-2010) was used to elucidate the microstructure of the laser-crystallized silicon. The TEM specimens were made by mechanical grinding and ion milling.

Figures 1(a) and 1(b) show the generic mask (CGH) for a two-dimensional (2D) pattern and the optical micrograph

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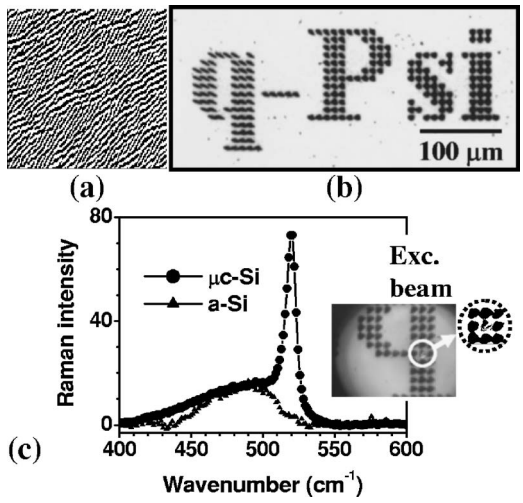

FIG. 1. (a) Generic mask (CGH) for a 2D pattern (the logo q-Psi) and (b) the optical micrograph of the 2D pattern produced in an *a*-Si film by the FT of a CGH. (c) The micro-Raman spectra for the laser-modified and the uxexposed regions in the 2D pattern. Here, q-Psi is the logo of the Quantum Photonic Science Research Center. In the inset image of (c), the *P* is inverted due to the optical layout of the confocal system; the small white circle represents the region in which the micro-Raman measurements were performed. Among the nine dots in the inset, the excitation laser is focused on the center dot. Pulse energy=16 μJ; laser shot number=5000.

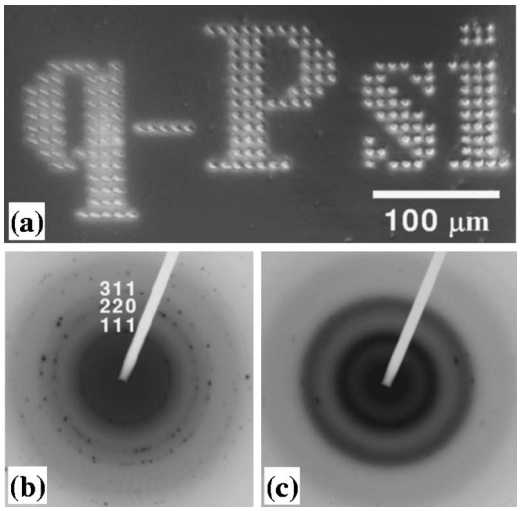

FIG. 2. (a) Plan-view SEM image for the logo (q-Psi) produced in the *a*-Si film by the FT of the CGH, (b) the indexed polycrystalline ED pattern from the bright region, and (c) an amorphous ED pattern from the dark region.

of the 2D pattern (the logo q-Psi) produced in an *a*-Si film by the FT of a CGH, respectively. The 2D pattern was produced in the *a*-Si film using the Fourier-optics method as follows: First, the generic pattern, that is the FT of the object, was designed and then converted to the real CGH element [Fig. 1(a)] using a photolithographic technique. Experimentally, the CGH element was produced by the reactive-ion etching of a quartz substrate with a chrome mask patterned by holographic lithography. Next, the laser structuring of the *a*-Si film was performed by positioning the CGH element just before a FT lens and locating the *a*-Si film near the focus of the lens. The image on the focal plane corresponds to the spatial FT of the CGH pattern. Therefore, the reconstructed pattern becomes a replica of the object. Experimentally, the logo q-Psi of Fig. 1(b) was produced in the *a*-Si film by the FT of the CGH in Fig. 1(a). The focal length of the FT lens is 8.0 mm. Imperfect imaging may be due to lens aberrations.

In order to find the origin of the femtosecond-laser structuring in an *a*-Si film, we measured the micro-Raman spectra for the laser-treated samples. Figure 1(c) shows the micro-Raman spectra for the femtosecond-laser-modified region as well as the uxexposed area in the 2D pattern. Based on previous studies in the literature,[19,20] it is believed that the 520 cm^{-1} band is assignable to the transverse-optic-phonon mode of crystallites with different sizes, whereas a broad and weak band centered at approximately 480 cm^{-1} is associated with the amorphous phase. When the Raman spectrum of the femtosecond-laser-modified region was compared with that of the uxexposed area, the intensity of the crystalline Raman peak was increased through the irradiation of the femtosecond laser. These results indicated that (i) the femtosecond-laser pulses induced a localized phase transformation from the amorphous to the crystalline phase, and (ii) the formation of the 2D pattern was ascribed to the spatially selected crystallization of the *a*-Si.

In order to further study the FLIC of *a*-Si, we investigated the structural properties of the laser-crystallized silicon by SEM and TEM. Figure 2 shows a plan-view SEM image and the selected-area-electron-diffraction (SAED) patterns from the 2D pattern obtained by TEM. Here, the SAED patterns of Figs. 2(b) and 2(c) correspond to the bright and dark regions of Fig. 2(a), respectively. The plan-view SEM micrograph clearly reveals that the spatially selected crystallization of the *a*-Si is responsible for the formation of the 2D pattern. The SAED pattern from the bright region [Fig. 2(b)] confirms that polycrystalline silicon was indeed produced by the FLIC and the crystals had a diamond cubic structure, as can be seen from the indexed ED pattern. Meanwhile, the SAED pattern from the dark region [Fig. 2(c)] remained nearly amorphous as evidenced by diffuse diffraction rings. A TEM analysis of the bright field image of the crystallized region indicated that an average grain size of ~100 nm was obtained through laser crystallization using 5000 laser shots from the 16 μJ beam.

Here we explain the advantage of the proposed method and the physics behind it. If an amorphous film undergoes the FLIC, the proposed method is beneficial because the arbitrary surface structuring of the amorphous film is quickly achieved by means of the FT of the CGH pattern. In this method, the CGH element generates the corresponding spot-array pattern on the focal plane, so that a single collimated beam is split into spot arrays in an arbitrary intensity distribution.[21] If femtosecond-laser pulses are incident to the CGH element, the generated spot-array pattern produces the target pattern of the CGH through spatially selected crystallization. If a CGH element for the arbitrary pattern is prepared, the arbitrary structuring of amorphous material can be achieved by the FLIC method. In fact, another type of a 2D pattern of 20×20 dots was created using the same method as producing the logo. As expected, the micro-Raman spectra showed that the formation of the 2D array was attributable to the spatially selected crystallization of *a*-Si. In our approach, the arbitrary surface structuring of *a*-Si films was carried out by applying the spatial FT method to the FLIC. In principle, the NIR femtosecond-laser pulses can induce multiphoton-absorption (MPA)-induced crystallization, because the *a*-Si

film exhibits a negligible amount of single-photon absorption (SPA), despite an appreciable amount of MPA in the incident wavelength region. Theoretically, laser structuring by MPA can generate the finer structures than that by SPA.[6,22,23] Next, we consider the grain size of the laser-crystallized silicon produced by femtosecond-laser annealing. In this study, the average grain size of ~100 nm was observed in the laser-crystallized region. The average grain size of ~100 nm is small for device application. However, the grain size can be increased by thermal annealing, where the laser-crystallized regions act as seeds for nucleation. Without the seed, crystallization is developed via homogeneous nucleation, which tends to produce a larger number of nucleation sites and a finer grain size compared to heterogeneous nucleation. If seeds are supplied via the spatially selective-laser crystallization, the preexisting seeds provide the preferential nucleation sites for crystallization, so that the final grain size is likely to be increased owing to the heterogeneous nucleation. Further growth of the grain size might be pursued by properly selecting the film thickness, the substrate, the underlayer, and the capping layer.

In conclusion, the arbitrary surface structuring of *a*-Si films was performed by applying the spatial FT method to the FLIC. The *a*-Si films were prepared on a glass substrate using PECVD. In order to realize the arbitrary surface structuring of an *a*-Si film, the logo q-Psi was produced in the *a*-Si film by means of the FT of the CGH. The crystallization of *a*-Si was performed using the NIR femtosecond-laser pulses. By micro-Raman spectroscopy, SEM, and TEM, it was found that (i) the femtosecond-laser pulses induced a localized phase transformation from the amorphous to the crystalline phase, and (ii) the spatially selected crystallization of the *a*-Si was responsible for the formation of the 2D patterns. If an amorphous film undergoes the FLIC, the proposed method is beneficial because the arbitrary surface structuring of the amorphous film is quickly achieved using the FT of the CGH pattern.

This work was supported by the Korea Science and Engineering Foundation (KOSEF) through Quantum Photonic Science Research Center.

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