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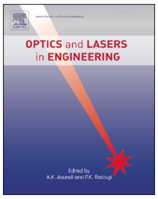

Femtosecond laser processing of indium-tin-oxide thin films

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ABSTRACT

Micro- and nano-scale crystalline indium-tin-oxide (c-ITO) patterns fabricated from amorphous ITO (a-ITO) thin films on a glass substrate using a (low NA 0.26) femtosecond laser pulse that is not tightly focused are demonstrated. Different types of c-ITO patterns are obtained by controlling the laser pulse energies and pulse repetition rate of a femtosecond laser beam at a wavelength of 1064 nm: periodic micro c-ITO dots with diameters of ~1.4 μm, two parallel c-ITO patterns with/without periodic-like glass nanostructures at a laser scanning path and nano-scale c-ITO line patterns with a line width ~900 nm, i.e. ~1/8 of the focused beam's diameter (7 μm at $1/e^2$).

1. Introduction

Indium tin oxide (ITO) is widely used as a transparent conducting oxide material with high transparency in the visible region of the spectrum. Since the crystalline structure usually has more physical properties than an amorphous structure does, the as-deposited amorphous-ITO (a-ITO) thin film usually undergoes furnace annealing at a temperature higher than 200 °C [1] and changes to crystalline-ITO (c-ITO) film. To achieve local and rapid thermal heating, a laser annealing process with a nanosecond pulsed laser [2,3] or ultra-short pulsed laser [4] of ITO is presented. The annealed c-ITO film is then usually patterned by a photolithography process, which is a complicated and time-consuming process, e.g. involves photoresist coating and baking, photoresist exposure and developing, etching and photoresist stripping.

Laser processing is a mature technology intended for use in patterning ITO film, i.e. selectively removing the ITO and leaving the desired ITO pattern. Some researchers have investigated the ablation process of ITO thin films with a long pulsed laser, i.e. nanosecond [5–8]. Recently, femtosecond laser pulses for processing ITO thin film have been investigated with positive results [9–12], e.g. selectively removing the ITO and leaving the desired ITO pattern as transparent electrodes for devices.

Since an a-ITO thin film has a more rapid etching rate than a c-ITO structure [13], some researchers utilized the long pulsed laser annealing process followed by wet etching to fabricate a c-ITO pattern from a-ITO thin films [3,14–16]. In our previous studies, a process to fabricate a crackless c-ITO pattern by the high repetition rate (80 MHz) femtosecond laser-induced crystallization process with a heat accumulation effect was reported [17]. However, due to the extended width caused by the heat accumulation effect, the line width of the fabricated c-ITO pattern was usually greater than that of the focused beam diameter. Recently, researchers [18] have been using a high repetition rate femtosecond laser resonator (85 MHz) with a high numerical aperture (NA 1.3) to anneal the polycrystalline ITO film into a different phase which is more resistive in hydrochloric acid and researchers follow up with wet etching to fabricate sub-wavelength nanostructuring. However, the process requires the use of a high NA lens; accordingly, it is difficult to precisely control the focal point.

In this study, micro- and nano-scale crystalline indium-tin-oxide (c-ITO) patterns are obtained from a-ITO thin film by a not tightly focused (NA 0.26) femtosecond laser (wavelength 1064 nm, variable pulse repetition rate 0.01–1 MHz) irradiation, followed by chemical etching. The results show that by careful control of the laser pulse energy and pulse repetition rate, different types of c-ITO patterns, e.g. periodic micro c-ITO dots, two parallel c-ITO patterns with/without periodic-like glass nanostructures and nano-scale c-ITO line patterns can be obtained.

2. Experiments

A femtosecond fiber laser (FemtoPower 1060-3uJ-s, Fianium Inc.) with a central wavelength of 1064 nm, a pulse width <500 fs, a variable repetition rate 0.01–1 MHz and a maximum pulse energy of

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~3 μJ was used for processing the a-ITO thin film in air. The laser beam entered an objective lens (numerical aperture 0.26, M Plan Apo NIR, Mitutoyo) and was subsequently incident in the normal direction on the surface of an a-ITO coated specimen mounted on an X–Y positioning stage. The focal spot diameter on the specimen surface was approximately 7 μm (at $1/e^2$). In the present experiments, a-ITO thin films with a thickness of ~100 nm were deposited on borosilicate glass substrates (NEG OA10) using a DC magnetron sputtering system.

The structures on the a-ITO film were then fabricated by translating the sample stage under the control of a PC-based micro-positioning system with a precision of better than 1 μm. After laser processing, the samples were immersed in a 0.05 mol/L oxalic acid etchant at 50 °C for 2 min. Due to the more rapid etching rate of the a-ITO thin film compared to the c-ITO structure [13], the etching process resulted in the complete removal of the non-irradiated a-ITO film, leaving only a crystalline c-ITO structure. After etching, the processed area of the surface was observed using an optical microscope and scanning electron microscope (SEM, FE-SEM 7001).

3. Results and discussion

Fig. 1(a) shows the microscopic image of the five laser-written lines produced on an a-ITO thin film surface using laser pulse energy of 84 nJ, a constant scanning speed of 25 mm/s and a pulse repetition rate in the range of 0.01–1 MHz. Note that the specimen is in an unetched condition. It can be seen that the film within the laser irradiated area is different from that within the unirradiated region. The irradiated sample was then etched, as shown in Fig. 1(b), leaving clearly visible patterns on the glass substrate. At a high pulse repetition rate, i.e. 0.5 and 1 MHz as seen in Fig. 1(b), the center area was ablated and the ablated channels were bordered on either side by two parallel ITO patterns. At a medium pulse repetition rate 0.05 and 0.1 MHz as seen in Fig. 1(b), a continuous c-ITO pattern was formed. At a lower repetition rate, i.e. 0.01 MHz as seen in Fig. 1(b), a discrete c-ITO dot pattern was presented.

For a single scanning path, where the laser beam is at normal incidence and focused on the surface of the sample, the equivalent number (N) of pulses applied to an assumed single laser spot on the ITO film is estimated by $N = D_e \times f/v_s$,where D_e is the focal spot diameter on the specimen surface, f is the pulse repetition rate and v_s is the scanning speed. For example, when $D_e = 7\ \mu\text{m}$ and the scanning speed is $v_s = 25$ mm/s, N is 28, 140 and 280 for a pulse repetition rate of 0.1, 0.5 and 1 MHz, respectively. The net energy density of the laser beam applied to the sample surface can be calculated by $(1-R) \times N \times F$, where R is the surface reflectivity and F is the laser energy density (fluence).

Fig. 2 shows the SEM images of Fig. 1(b). Fig. 2(a) and (b) reveals two regions with different structures: in the center of the laser written pattern, the a-ITO thin film was clearly ablated (width ~2.5 μm) while the glass substrate surface was not damaged. In the edges of the laser written pattern, two parallel c-ITO patterns (width ~1.3 μm for each side) adjacent to the ablated regions were formed. Due to the Gaussian property of the irradiated beam profile, the middle of the laser written area was more irradiated by higher laser energy than the edges. The net energy density of the laser beam was higher than the ablation threshold of the a-ITO thin film, and thus, the center area was ablated. However, when the outer edges were subjected to lower irradiation intensity, the net energy density was still higher than the crystallization threshold of a-ITO thin film, and thus, the two parallel c-ITO patterns adjacent to the ablated regions were formed. It was found that the overall line width (ablated + two parallel c-ITO) decreased with a decreasing repetition rate.

In Fig. 2(c) and (d), it can be seen that after chemical etching, the continuous c-ITO line pattern with ~2.5 μm line width was retained on the glass substrate. This means that in the middle of the laser written area, the irradiated net laser energy was enough to crystallize the a-ITO thin film and the irradiated a-ITO was completely transformed into c-ITO. It was found that the line width decreased with a

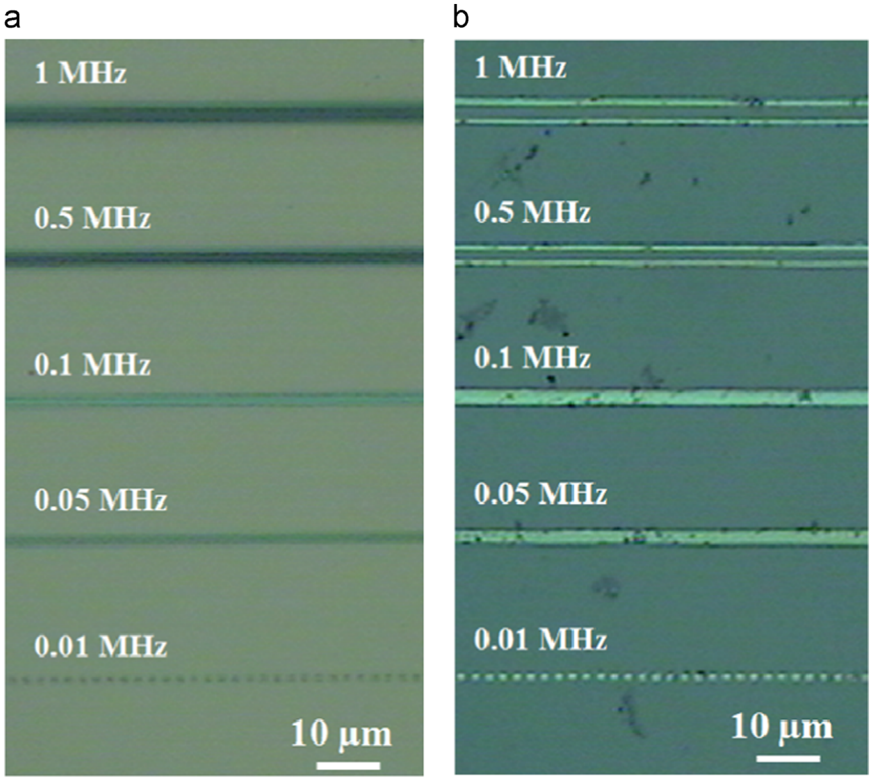

Fig. 1. Microscopic images of laser irradiated ITO thin film surface with different pulse repetition rates (a) before and (b) after etching.