Rapid and selective green laser activation of InGaZnO thin-film transistors through metal absorption

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ABSTRACT
We used a pulsed green laser to activate indium gallium zinc oxide thin-film transistors (IGZO TFTs). The IGZO films with large bandgaps (> 3 eV) were easily activated by heat delivered by a pulsed green laser to the gate, source, and drain metal electrodes. The IGZO TFTs were quickly and selectively activated in the absence of conventional annealing. Compared to the IGZO TFTs that were annealed at 300°C, the IGZO TFTs that were activated via pulsed green laser irradiation exhibited superior electrical characteristics: a field effect mobility of 11.98 ± 0.64 cm² V⁻¹ s⁻¹, a subthreshold swing of 0.33 ± 0.02 V dec⁻¹, and an on/off ratio of 8.28 × 10⁹ ± 7.42 × 10⁹, which were attributable to increases in the number of metal–oxide (M-O) bonds and oxygen vacancies, and reduced levels of OH-related species. The pulsed green laser broke weak chemical M-O bonds in the IGZO films through dihydroxylation of the OH-related species, and then strengthened the residual M-O bonds via heat transfer from the metal electrodes. This new activation process could replace conventional annealing and is expected to expand the applications of flexible and transparent devices.

1. Introduction
Several attempts have been made to fabricate oxide thin-film transistors (TFTs) on flexible substrates to meet the increasing demand for flexible displays [1–5]. Sputter-processed indium gallium zinc oxide (IGZO) TFTs have already been commercialized and used in backplanes due to their high optical transparency, low off-current, and high mobility compared to amorphous silicon (a-Si) TFTs [6–8]. However, IGZO TFTs are difficult to fabricate on flexible substrates, such as polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) that have low glass transition temperatures, since IGZO TFTs require heating to over 300°C. Thus, various activation processes, including conventional heat treatment and processes that use other energy sources, have been developed to activate IGZO TFTs at low temperatures [9–14].

In this study, green laser irradiation was used to activate IGZO TFTs. The concentrated green laser energy rapidly and selectively activated the IGZO TFTs. The green laser has a lower photon energy (~ 2.3 eV) than a UV laser (~ 6.7 eV), thus reducing damage to thin films and flexible substrates during irradiation. Also, the green laser is advantageous in terms of process time because it makes the activation time shorter than in other activation methods [15]. Despite these advantages, IGZO TFT activation via visible light, including green light, has rarely been used because the bandgap of the IGZO film is generally greater than 3 eV and the IGZO film hardly absorbs visible laser energy [16]. In this study, the gate, source, and drain metal electrodes were all involved in the activation process [Figure 1(a)]. The green laser was used to irradiate parts of the source/drain electrodes and the back channel, where the thermal energy generated in the metal electrodes is transferred to the TFT channels. A pulsed rather than continuous wave laser was used to minimize degradation of the IGZO film and enable efficient heat absorption and transfer. In our previous study, we could not activate IGZO TFTs selectively and sufficiently due to the p⁺Si wafer substrate and the continuous wave laser, respectively [13].

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2. Experiments

2.1. Fabrication process of a-IGZO TFTs with green laser activation

Mo (the gate electrode) and SiO₂ (the gate insulator) were prepared on glass substrates, and channeled IGZO films (~40 nm thick) were deposited onto the SiO₂ (120 nm thick) via radiofrequency (RF) sputtering with a power of 150 W, a working pressure of 5 mTorr, and a deposition time of 5 min. As the oxygen partial pressure was 0 Torr, the as-deposited IGZO films were conductive, with a resistance of about 10 kΩ. Next, Al source and drain electrodes were deposited on the IGZO films/SiO₂ via RF sputtering. Green laser radiation was delivered to part of the back channel and to the source and drain electrodes. We used a diode-pumped solid-state (DPSS) laser (LRS-0532; Laserglow Technologies Inc.). The laser was controlled using transistor-transistor logic (TTL), and damage to the IGZO TFTs during the irradiation was minimized by changing the wave mode from continuous to pulsed or vice versa. As shown in Figure S1(a)–(c), the relationship between the laser power density and the distance was Gaussian, and the maximum energy density was 300 mJ cm⁻² near the laser center. The effective beam diameter was 650 μm, which is wider than the diameter of the channel (150 μm). The gate, source, and drain electrodes were adequately irradiated. We used a joystick to move the laser freely along the x, y, and z axes (maximum...
speed, 100 mm/s) for selective activation of the IGZO TFT regions. To optimize the laser activation, the focal length, frequency, irradiation time, and energy density were adjusted as appropriate.

2.2. Measurements and analysis method
A parameter analyzer (4156C; Hewlett-Packard) was used to define the transfer characteristics of the IGZO TFTs. The gate voltage was varied from −30 to 30 V but the drain voltage was fixed at 10.1 V. A positive bias stress (PBS) test was performed at a gate voltage of 20 V and a drain voltage of 10.1 V for 3,600 s, and a negative bias illumination stress (NBIS) test was conducted at a gate voltage of −20 V and a drain voltage of 10.1 V for 3,600 s under a white light-emitting diode illumination of 5,700 lux. Spectroscopic ellipsometry (SE) data were derived over the entire energy range (1.24–5 eV; step, 0.02 eV) at an incident angle of 65°. X-ray photoelectron spectroscopy (XPS) was performed in the constant analyzer energy (CAE) mode at 50 eV using an Al K alpha source.

3. Results and discussion
Figure 1(a) shows the green laser activation of the IGZO TFTs. The laser irradiated evenly spaced IGZO TFTs via x- or y-axis beam movement. A green laser with an effective beam diameter of about 650 μm selectively activated the IGZO TFTs. Figure 1(b) shows the detailed activation process (heat absorption and transfer). The laser energy was absorbed almost exclusively by the source, drain, and gate metal electrodes, and not by the IGZO film, the SiO₂ film, or the glass substrate, all of which had bandgaps of at least 3 eV. The absorbed laser energy was converted into heat and transferred to the IGZO channel layer, the property of which instantaneously changed from metallic to semiconducting. Figure 1(c) shows how we maximized the laser energy delivered to the metal electrodes using a beam expander and a focusing lens, rather than the more commonly used homogenizer. The objective lens increased the laser beam diameter, and the collimator provided a constant beam direction. Finally, the focusing lens concentrated the beam on a point. We refer to the objective lens/collimator combination as a beam expander. Figure S1(d) shows a schematic in which the laser beam is focused on a selected point. The final beam diameter (2 W₀) is inversely proportional to the diameter (D₀) of the incident laser. Because the beam diameter increased as it passed through the beam expander, the focusing lens could have reduced the final beam diameter to 650 μm after optimizing the optical characteristics and the distance between the laser head and the devices. The beam diameter was minimized using the beam expander and the focusing lens, and the metal electrodes effectively absorbed the laser energy.

To optimize the electrical characteristics of the activated IGZO TFTs, the laser focal length, frequency, irradiation time, and energy density were adjusted as shown in Figure 2(b)–(e). As indicated in Figure S1(d), the distance between the focusing lens and the IGZO TFTs is the focal length, which critically affected the beam diameter and the laser energy transfer to the metal electrodes. Figure 2(a) and (b) show schematic diagrams of the green laser irradiation and the effect of the focal length on the electrical performance of the IGZO TFTs. When the focal length was 10 cm, the spot size was the smallest, so the energy of the green laser was efficiently transmitted to the IGZO film. When the focal length was 5 cm, the green laser was close to the IGZO TFT, so the spot size (10 cm) was relatively larger than the focal length. When the focal length was 15 cm, the laser was far from the IGZO TFT, so the spot size (10 cm) was relatively larger than the focal length. Thus, the IGZO TFTs had no satisfactory switching characteristic at focal lengths of 5 and 15 cm. The off-current of the IGZO TFTs decreased significantly at a focal length of 10 cm, and then increased once more. At a focal length of 10 cm, the optimally activated IGZO TFTs had a field-effect mobility of 11.98 ± 0.64 cm² V⁻¹ s⁻¹, a subthreshold swing (SS) of 0.33 ± 0.02, and an on/off ratio of 8.28 × 10⁹ ± 7.42 × 10⁹.

Figure 2(c) shows the transfer characteristics of the IGZO TFTs according to the laser frequency. The IGZO TFTs that were irradiated at low frequencies (0, 10, and 50 Hz) exhibited high off- and on-currents and poor switching characteristics. However, the irradiation at 100 Hz did not damage the IGZO TFTs. At higher frequencies (400, 700, and 1,000 Hz), the IGZO TFTs were not adequately activated and exhibited poor switching characteristics (a high off-current and SS) because from 100 to 1,000 Hz, the electrodes reached their maximum temperature at different times. As both the 100 and 1,000 Hz lasers had pulse durations that were half as long as the periods, their average power densities were half their peak power densities. However, the increased temperature rate at which the electrodes absorbed the laser (the heating state) was greater than the decreased temperature rate at which the electrodes did not absorb the laser (the cooling state). Thus, the IGZO TFTs that were activated with a 100 Hz laser reach a higher temperature because the pulse duration of such laser is longer than that of a 1,000 Hz laser even if the laser irradiation times are identical [17].

In most cases, the area close to the laser spot is damaged by the rapid temperature rise during laser irradiation, and the material may melt or evaporate. Similarly,
when a laser irradiates thin films, the films are distorted or transformed in other ways. The laser frequency is critical and laser pulsing is essential to prevent damage to thin films. Figure 3(a) and (b) show schematic diagrams and micrographs of the IGZO TFTs after low (0 and 10 Hz) and high (100 and 1,000 Hz) green laser irradiation, respectively. As shown in Figure 3(c) and (d), low-frequency irradiation (< 10 Hz), including continuous irradiation (0 Hz), significantly damaged the films, which would adversely affect their electrical performance unlike high frequency irradiation (100 and 1000 Hz).

In general, heat treatment of IGZO TFTs is associated with changes in the activation energy triggered by the high temperature and the long duration thereof. At a high temperature (> 300°C), the activation may be very rapid, while at a low temperature (< 150°C), no activation is evident. This must be considered when activation is performed using a laser. The peak power density, which is related to heat, is considered more important than the irradiation time. However, lasers that directly irradiate thin films damage the films. Both an appropriate irradiation time and high laser power density are required for good activation with minimal thin film damage.

Figure 2(d) shows the transfer characteristics of the IGZO TFTs according to their laser irradiation time. The characteristics were optimal at 60 s. When the irradiation time was longer, the electrical performance was degraded because the IGZO films and the metal electrodes were damaged. As shown in Figure 2(e), the IGZO TFTs were not activated at all when the laser power density was low, and were sufficiently activated for high electrical performance only at a high energy density of 300 mJ cm⁻².

Thus, we confirmed that IGZO films, which generally do not absorb visible light, can only be activated using a pulsed green laser (i.e. in the absence of conventional heat treatment) if the laser parameters are optimized (focal length, 10 cm; frequency, 100 Hz; irradiation time, 60 s; and energy density, 300 mJ cm⁻²).

To prove that laser absorption occurred at the gate (Mo), source (Al), and drain (Al) electrodes, the transfer characteristics of the IGZO TFTs with different electrodes were compared as shown in Figure 4. Mo, Al, and transparent ITO electrodes were used. When ITO was used to fabricate the gate, source, and drain electrodes, the IGZO TFTs were not activated, as shown in Figure 4(a). When Mo served as the gate electrode and ITO, as the source and drain electrodes, and when ITO served as the gate electrode and Al, as the source and drain electrodes, the transfer characteristics were appropriate, as shown in Figure 4(b) and (c). Thus, the activation did not involve the ITO electrodes, which have high bandgaps, but only the Mo and Al electrodes. The activation was successful when Al served as the source and drain electrodes and when Mo served as the gate.
Figure 3. (a) Schematic diagrams and (b) microscope images of the IGZO TFTs activated with green laser irradiation. Microscope images of the IGZO TFTs (c) after low-frequency (0 and 10 Hz) and (d) high-frequency (100 and 1,000 Hz) green laser irradiation.

electrode, as in the optimized IGZO TFTs described above.

Figure 5(a) summarizes the transfer characteristics of the IGZO TFTs that were not activated and of those that were activated using a continuous wave (CW) laser, a pulsed laser, and thermal-only (300°C) conditions. The inactivated and CW-activated TFTs exhibited metallic properties, and the other TFTs were semiconductive. Pulsed laser activation was associated with better electrical performance than thermal-only activation [Figure 5(a) and Table S1].

The results of the PBS and NBIS tests shown in Figure 5(b) indicate that the laser-activated IGZO TFTs were more stable than those subjected to thermal-only activation. The PBS test revealed positive $V_{th}$ shifts of 1.6 V (for the thermal-only activation) and 1.4 V (for the pulsed green laser activation). In the NBIS test, there were negative $V_{th}$ shifts of $-12.2$ V (for the thermal-only activation) and $-6.8$ V (for the pulsed green laser activation). Thus, the pulsed green laser activation more significantly reduced the levels of the uncoordinated oxygen species in the IGZO thin films than did the thermal-only activation.

Figure 5(c) summarizes the field-effect mobility, SS, and on/off ratio data of 10 different IGZO TFTs that were subjected to thermal-only or pulsed green laser activation. The IGZO TFTs that were subjected to pulsed green laser activation exhibited better electrical characteristics and greater uniformity.

To confirm the band alignments of the activated IGZO films, we measured the valence band offset ($E_{FV}$) between the Fermi level ($E_F$) and the valence band maximum
Figure 4. Schematic images and transfer characteristics of the IGZO TFTs with different electrode materials. Source/drain electrodes: ITO for (a) and (b), and Al for (c) and (d). Gate electrodes: ITO for (a) and (c), and Mo for (b) and (d).

Figure 5. (a) Transfer characteristics of the IGZO TFTs activated with various methods. (b) PBS and NBIS tests of the IGZO TFTs activated with 300°C thermal-only activation and with a pulsed green laser. (c) Statistical data on the field-effect mobility, S.S., and on/off ratio of several IGZO TFTs activated with 300°C thermal-only activation and with a pulsed green laser (10 devices each).

(EV), and the optical bandgap. Figure 6(a) shows the EV data that were derived using XPS in the region of the valence band energy. The IGZO films that were not activated, and those that were subjected to thermal-only and pulsed green laser activation, exhibited EV values of 3.48, 3.17, and 3.27 eV, respectively. Figure 6(b) shows that the optical bandgaps (the imaginary parts of the absorption coefficients derived from the SE spectra) were 3.46, 3.51, and 3.52 eV, respectively. Figure 6(c) shows the band alignments that were derived using the measured EV and optical bandgaps. The as-deposited IGZO films (that were created under an oxygen partial pressure of 0%) exhibited very high carrier concentrations (~9.1 x 10⁵¹ cm⁻³) [9] because the EV was at the top of the conduction band minimum and degenerated the semiconductive characteristics, similar to the untreated IGZO films shown in Figure 6(c). However, for the activated (semiconductive) IGZO films, the EV was located in
the bandgap between the conduction and valence bands, as in typical semiconductors.

In the semiconductor theory, the $E_{CF}$ is inversely proportional to the carrier concentration of a thin film [11]. In this study, the $E_{CF}$ of the IGZO films that were activated by the pulsed green laser was lower than that of the films that were subjected to thermal-only activation [Figure 6(c)]. Thus, the IGZO films that were activated by the pulsed green laser exhibited relatively higher carrier concentrations. The percolation model of oxide semiconductors indicates that a high oxide carrier concentration is associated with a high Hall mobility of oxide thin films and a high field effect mobility of oxide TFTs [6]. The transfer characteristics shown in Figure 2 and Table S1 demonstrate that the laser-activated IGZO TFTs had higher field effect mobilities than the IGZO TFTs that were subjected to thermal-only activation.

Figure 7(a)-(c) show the XPS O 1s spectral surface data. We explored the physical properties of the IGZO films before their activation and after their thermal-only and pulsed green laser activation. The spectra reflect changes in the oxygen composition, and thus, the M-O bonding level, oxygen vacancies ($V_o$ values), and hydroxyl group (-OH) peak values. The peak values were derived by summing the raw data via deconvolution based on the Gaussian distribution. For both activation methods, the M-O bond, $V_o$, and -OH peaks were located...
Figure 7. XPS O 1s spectra of the IGZO films activated with (a) no treatment, (b) 300°C thermal-only activation, and (c) a pulsed green laser.

at 530.3 ± 0.1, 531.1 ± 0.1, and 532.1 ± 0.2 eV, respectively. In most of the IGZO films, the M-O bonding was greater than the V<sub>O</sub> value, but the V<sub>O</sub>:M-O bonding ratio differed slightly according to the activation method used. The non-activated films exhibited lower M-O bonding and higher V<sub>O</sub> concentrations than the activated films.

Figure 8. Atomic force microscopy (AFM) images and line profiles of the IGZO films activated with (a) no treatment, (b) 300°C thermal-only activation, and (c) a pulsed green laser.

Figure 9. Schematic images of the mechanism of the IGZO TFTs activated with a pulsed green laser (a) before irradiation, (b) shortly after irradiation, and (c) 60 seconds after irradiation.
because the IGZO films were deposited by sputtering under a partial oxygen pressure of 0 Torr. The activated films exhibited high M-O bonding strength and low $V_o$ values because annealing strengthens the bonds between metal and oxygen atoms. Remarkably, the pulsed green laser (in the absence of heat treatment) produced an activation effect similar to that of conventional heat treatment at 300°C. Moreover, compared to conventional thermal activation, pulsed green laser activation reduced the levels of the OH-related species in the IGZO films and increased the $V_o$ values and the M-O bonding strength.

A higher $V_o$ increases the carrier concentration. The $V_o$ sites serve as donors, and if they are close to the acceptor states near the conduction band minimum, the carrier mobility increases via the formation of electron transport paths [18]. As the conduction band minimum is in the s-state in oxide (but not hydroxide) lattices, M-O bonding also plays an important role in carrier transport [19]. In this study, compared to the $V_o$ concentration (32.9%) after the thermal-only activation, the concentration after the laser activation was slightly higher (34.1%), thus enhancing the carrier concentration and mobility. The laser irradiation reduced the levels of the OH-related species in the oxide films, which increased the carrier mobility [20]. As shown in Figure 7(c), after the green laser irradiation, the proportion of the OH-related species decreased from 13.2% to 8.3%. Thus, the pulsed laser activation dehydroxylated the OH-related species and increased the M-O bonding strength and the $V_o$, which in turn improved the electrical properties.

Figure 8(a)–(c) show atomic force microscopy (AFM) images of the IGZO films that were not activated, and of the films that were activated by the thermal-only and laser approaches. We obtained their arithmetical mean height ($S_a$) and the root mean square (RMS) height ($S_q$). After their activation, both the $S_a$ and $S_q$ increased compared to the untreated devices, as reported previously [21,22]. The $S_a$ and $S_q$ of the laser-activated films were slightly greater than those of the films that were subjected to thermal-only activation. The red lines show the locations of the peak thickness. The values were 16 and 8 nm after the laser activation and the thermal-only activation, respectively.

In general, a ripple is generated when a thin film melts because of the interference between the incident laser light and the light scattered by the surface [23]. However, we did not observe a ripple, as shown in the AFM images in Figure 8(c). Unlike with the thermal-only activation, under the laser activation, the IGZO film damage was not completely eliminated. However, the laser did not appear to have a major effect on the characteristics of the IGZO films and the IGZO TFTs, as the $S_a$ and $S_q$ values did not differ significantly.

Figure 9(a)-(c) show schematics of the IGZO TFT characteristics before and after the laser irradiation. A green laser photon (532 nm) has an energy of approximately 2.33 eV [24]. As the binding energies of weak chemical bonds and the In-O, Zn-O, and Ga-O bonds in IGZO films are below 2 eV, they are broken when the laser directly irradiates the films [25]. However, the effects are insignificant compared to those of a UV laser with a short wavelength and high energy. The bonds broken via direct green laser irradiation strengthen M-O bonding when heat is transferred from the metal electrodes. Thus, weak bonds are broken and strong bonds are formed when heat is transferred, which improves the

Figure 10. Flexible IGZO TFT activated with a pulsed green laser on a PET substrate: (a) schematic structure, (b) images, and (c) transfer characteristics.
electrical properties. As the XPS O 1 s results showed, our laser process increased the M-O bonding and the V o level in the IGZO films, and decreased the number of the OH-related species, compared to conventional annealing. The numbers of the OH-related species decreased because the laser broke the weak chemical bonds and the dehydroxylated OH-related species, thereby promoting M-O bonding and V o generation [1,20,26].

We used a thermocouple to measure the temperature during the irradiation. After connecting the thermocouple meter to an IR detector and putting the thermocouple meter on an IGZO film, we measured the maximum temperature of the IGZO film during the green laser irradiation (Figure S2). The maximum IGZO film temperature between the source and drain electrodes was 187.3°C, which was adequate to dehydroxylate the OH-related species. The energy transferred to the channel through this process activates the IGZO TFT, enhancing the M-O bonding.

Figure 10(a)–(c) show the creation of the IGZO TFTs on a flexible PET substrate before the TFTs were activated using laser. A SiO2/ZrO2 thermal diffusion barrier was deposited on the PET to prevent heat transfer from the gate electrode to the PET substrate. The field effect mobility was 8.62 cm² V⁻¹ s⁻¹, the SS was 0.85 V dec⁻¹, and the on/off ratio was 3.09 × 10⁷. Thus, pulsed green laser activation can be used to prepare flexible IGZO TFTs and can be utilized in various flexible electronic parts such as sensors and memories [27].

4. Conclusion

We used green laser irradiation to activate IGZO TFTs instead of conventional heat treatment, which can damage flexible substrates. Only the gate, source, and drain electrodes absorbed the light and generated heat, which activated the TFTs. The electrical performance of the TFTs was better than that of the TFTs that were subjected to conventional heat activation because the numbers of their M-O bonds and oxygen vacancies increased, and the levels of their OH-related species decreased, compared to those of the TFTs subjected to conventional heat activation. This process is expected to broaden the range of substrates for flexible and transparent devices.

Disclosure statement

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