Interface Trap-Free, 100% Yield, Wafer-Scale, Non-Volatile Optically-Guided Memory Array from Cumulatively-Stacked Small Molecules/Fluoropolymer/Copper-Oxide Nanoparticles Heterostructure

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Optically-guided memory devices, with its photo-response, allow for photodetector and memory functions to be combined in a single device. As a result, the issue of unnecessary signal delay can be alleviated by reducing the metal wire between the photodetector and the memory device, and the function of two different devices can be performed simultaneously, which enables an image recognition system to be miniaturized. With such advantages, optically-guided memory is therefore considered to be highly promising as a potential key component for next-generation applications where image detection and processing capabilities are paramount. Here, a wafer-scale $12 \times 12$ dinaphtho[2,3-b:2′,3′-f]thieno[3,2-b]thiophene (DNTT)-based optically-guided memory transistors (OMT) array with a cumulatively stacked small-molecules/fluoropolymer/copper-oxide nanoparticles structure is demonstrated. The proposed OMT is formed in 4 different states depending on the light intensity. Furthermore, read current ($I_{\text{read}}$) and threshold voltage ($V_{\text{th}}$) in the programming state (P-state) of OMT is maintained stably even after 20 days. Based on the optimized DNTT thickness, a wafer-scale $12 \times 12$ OMT array is fabricated consisting of 144 devices for text image detection with 100% yield. This study also demonstrates a text image detection with non-volatile memory characteristics depending on the presence or absence of light irradiation.

1. Introduction

In this era of the 4th industrial revolution, many technologies are developing at tremendous speeds, and artificial intelligence,[1–3] autonomous vehicles,[4–6] and telemedicine[7–9] are emerging applications that are gaining considerable attention. In each of these applications, the development of image sensor-based camera technology is crucial. Specifically, in each domain, the subsequent execution procedure of the system is determined based on the image information that has just been obtained. Further, since these applications can directly affect daily life, more accurate image detection and high-speed detection are essential. Therefore, there is an urgent need for the development of a high-performance image sensor for the stable commercialization of innovative technologies based on image data detection and processing.

Meanwhile, an optically-guided memory transistor (OMT) is a single-structure memory device in which a photodetector generates photocurrent in response to light exposure. This device combines that photodetector function with a memory function that stores light signal information through charge trapping. As a result, the memory device itself can adjust the memory state depending on the incident light properties such as wavelength and intensity.[10–13] In the conventional image information detection process, there is a need to transmit optical information detected through a photodetector to a memory cell, which causes an unavoidable propagation delay in the information transfer process.[14] However, the use of the above-mentioned OMT reduces the difficulties caused by that delay due to its structure wherein optical information is stored in the memory at the same time as the photo-response. Further, the overall system size is reduced because functions that are typically performed by different devices—such as photodetection and information storage—are integrated into one device. Due to these advantages, OMTs have recently received considerable attention.

However, OMTs still have some limitations. i) First, the results of most prior studies only showed a single sweep of the transfer curve characteristics of the OMTs.[15–18] For reliable memory characteristics, the same current characteristics must be secured when the same voltage is applied regardless of the voltage sweep direction. Therefore, it is necessary to demonstrate
stable operation characteristics by showing the dual sweep operation in the transfer curve of the OMT according to the memory state. i) Second, previously reported studies showing memory characteristics through light only showed the programmed state and the erased state.\[^{19-21}\]\n
In a conventional memory device, this is only sufficient if specific logical values such as 0 and 1 can be distinguished. However, since the core of OMT is to store information about light, multi-state memory characteristics according to wavelength or light intensity—which are involved in light information—should be required. ii) Third, the materials that have primarily been used for a floating-gate have been noble metals such as Au and Ag or high-cost organic materials, thus requiring a high-cost process for mass-production that is limited by their high scarcity.\[^{22-24}\]\n
Therefore, there is a need for research on alternative materials that are relatively low-cost and stable even when exposed to ambient air. iv) Fourth, most emphasized, the cumulative deposition of heterogeneous layers with a trap-free interface is needed. As the floating-gate structure is made through at least four multilayered depositions (i.e., blocking dielectric, floating-gate, tunneling dielectric, and semiconductor channel layer)\[^{25-28}\]\n
each layer must have a surface without interfacial traps or defects to achieve operational stability and reduced leakage current during programming and erasing. v) Finally, although many studies have been reported to implement OMT characteristics, most studies have demonstrated only the unit device level. It is essential to obtain a sufficient yield on a large-area substrate and develop an array level beyond unit devices are essential for applications with more practical functions such as image detection.

In this paper, we present a wafer-scale 12× 12 array of 144 dinaphtho[2,3-b:2′,3′-f]thieno[3,2-b]thiophene (DNTT)-based OMTs with a cumulatively stacked small-molecules/fluoropolymer/copper-oxide nanoparticles (CuO NPs) structure. The proposed OMTs satisfy the above-described five requirements i)–v). To understand the optically-guided memory operation, we analyze the optically-programming and erasing characteristics according to the thickness of the DNTT. The proposed OMT is not programmed under the dark while under light irradiation, the devices provide four distinct memory states as a function of the light intensity. Furthermore, read current ($I_{\text{Read}}$) and threshold voltage ($V_{\text{th}}$) in the programming state (P-state) of OMT are maintained stably even after 20 days non-volatile. Based on the explored OMT device, we fabricate a 12× 12 OMT array consisting of 144 devices on a 4-inch wafer. It is emphasized that the fabricated 144 OMTs exhibit 100% yield of the optically-guided memory operation with no hysteresis behavior, and it is confirmed that the devices have a low variation of $I_{\text{Read}}$ in the P-state and erased state (E-state), presenting their uniformity. Finally, we demonstrate a test image detection for alphabet “G” with non-volatile memory properties with or without light irradiation while programming through $I_{\text{Read}}$ and $V_{\text{th}}$ mapping.

The proposed OMT was composed of boron-doped silicon with $1.46 \times 10^{16}$–$4.44 \times 10^{18}$ atoms·cm$^{-2}$ and SiO$_2$ as the back gate and blocking dielectric, respectively. CuO NPs and CYTOP were formed by spin coating on SiO$_2$ as floating-gate and tunneling dielectrics, respectively. Then, 56 nm thick DNTT and 100 nm thick Au were deposited as channel and source/drain electrodes using a thermal evaporator. The channel length and width of the fabricated OMT are 100 and 1000 μm, respectively. The fabrication process of OMT is depicted in detail in Figure S1, Supporting Information. Figure 1a shows a structural illustration of the proposed OMT. The cross-section scanning electron microscope (SEM) image of the fabricated OMT showing each layer is cumulatively stacked without a pin-hole formation (Figure 1b). Figure 1c shows the optical microscopy image of the fabricated OMT. Figure 1d shows the absorbance of DNTT used as a channel in the proposed OMT, which indicates that DNTT absorbs visible light between 450 and 500 nm. Figure 1e shows an atomic force microscopy (AFM) image of the surface of a Si/SiO$_2$ substrate coated with CuO NPs to be applied as a floating-gate. The measured AFM image of the CuO NPs exhibited island-shaped particles periodically with a height of several tens of nanometers, thus indicating that the CuO NPs are uniformly distributed without an aggregation. The implication of the island-shaped particles as a floating-gate will be discussed on a prevention of interference effect in adjacent devices in an array. Regarding the roughness of the CuO NPs surface, it has previously been reported that when DNTT used as a channel layer is deposited on a rough surface, it leads to significantly degraded electrical characteristics—including effective mobility—due to the formation of many grain boundaries.\[^{29}\]\n
Therefore, we analyzed the surface roughness through AFM images of each layer while depositing CuO NPs, CYTOP, and DNTT, respectively (Figure 1e–g). The root-mean-square surface roughness ($R_s$) was 1.829 nm immediately after CuO NPs coating, and it improved to 0.483 nm after CYTOP coating. Therefore, the DNTT channel layer could be formed without degradation. On the other hand, after the DNTT was deposited, the surface roughness increased to 7.091 nm, but rather the increased DNTT surface roughness enables to increase the contact area with the Au electrode to facilitate charge injection.\[^{30}\]\n
Figure 1h shows a plot for comparing the surface roughness of each layer.

The OMTs were prepared by sequentially depositing the CuO NPs, CYTOP, DNTT, and Au contact electrodes. A more detailed description of the fabrication process is given in the experiment section below. Figure 2a depicts a schematic symbol of the proposed OMT. The scheme for measuring the optically-guided memory characteristics of the OMT is shown in Figure 2b. For the optically-guided memory operation evaluation, each of the fabricated OMTs was irradiated by visible-region multiple-wavelength LED light, and the wavelength characteristics are provided in Figure S2, Supporting Information. Figure 2c shows the respective transfer curves of the proposed OMT in the pristine state and the E-state. It can be emphasized that the hysteresis-free electrical characteristics were observed. The hysteresis-free operation of the proposed OMT derived from the interfacial trap-free fluoropolymer was maintained even after erasing/programming operation. After erasing operation ($V_{\text{C}} = −100 \text{ V}$) under dark, the $V_{\text{th}}$ of OMT was shifted from $−4.14$ to $−20 \text{ V}$. The shift of the $V_{\text{th}}$ toward the negative gate voltage direction indicates that hole carriers are trapped in the floating-gate at a large negative gate voltage ($V_{\text{C}} = −100 \text{ V}$) in the erasing operation is applied. While the erasing operation was performed under dark, the shift of transfer curve was not observed after the programming operation ($V_{\text{C}} = 100 \text{ V}$) under
dark. Thus, the same memory state as the E-state was maintained (Figure 2d). In contrast, a significant positive shift of $V_{th}$ ($\Delta V_{th} = 21.5$ V) in the transfer curve occurred when programming operation ($V_G = 100$ V) was performed under light illumination at $L_{int} = 5500$ lx ($P_{inc} = 0.81 \text{ mW cm}^{-2}$), resulting in a clearly distinct memory state (Figure 2e). For a quantitative comparison of the programming behavior with or without light illumination, Figure 2f,g shows $I_{\text{Read}}$ and the $V_{th}$ for E-state and two P-state (dark, w/light). As a result of comparing the $I_{\text{Read}}$ for each state, the P-state formed under dark was almost identical to the $I_{\text{Read}}$ of a few pA at $V_D = -1$ V and $V_G = 0$ V in the E-state, resulting that the memory state was not distinguishable. On the other hand, the P-state formed under light of $L_{int} = 5500$ lx led to significantly increased $I_{\text{Read}} = 2 \times 10^{-7}$ A. Therefore, the $I_{\text{Read}}$ ratio of P-state and E-state ($I_{P-state}/I_{E-state}$) could reach up to $2.34 \times 10^3$, which is a 10-fold improvement compared to $I_{P-state}/I_{E-state} = 2.36$ in P-state formed under dark. The increased $I_{P-state}/I_{E-state}$ enabled a clear distinction between the P-state and E-state. The $V_{th}$ shift according to the presence or absence of light irradiation during programming operation also showed the same trend as the change in $I_{\text{Read}}$. The $V_{th}$ of P-state (under dark) was measured to $-19.55$ V shifted by only 0.42 V (1.05% change at the operating voltage of $-40$ V) compared to that of E-state, and there was negligible difference with that of E-state.

On the other hand, the $V_{th}$ of the P-state (w/light) formed under light irradiation ($L_{int} = 5500$ lx) was located at $1.53$ V, shifted by $21.5$ V (53.75% change at the operating voltage of $-40$ V). The memory states of OMT modulated by light irradiation showed the light-dependent memory characteristics of the proposed OMT. The proposed OMT properly switched its state depending on the erasing and programming operations as a function of time (Figure S3, Supporting Information). To further investigate the optically-guided memory properties, we evaluated the retention characteristics of the OMT (Figure 2h). The P-state and E-state of the proposed OMT were stably maintained with the programming/erasing current ratio of about $10^6$ or more even after 2000 s. This high $I_{P-state}/I_{E-state}$ ratio enables the formation of several multi-memory states corresponding to intermediate values between the $I_{P-state}$ and $I_{E-state}$ States, which will be presented in the next paragraphs. Furthermore, we further evaluated the retention characteristics of the proposed OMT, and the measured retention test exhibited that $I_{\text{Read}}$ and $V_{th}$ were maintained unchanged even after 20 days (Figure S4, Supporting Information).

To understand the operating mechanism of the proposed OMT, we performed ultraviolet photoelectron spectroscopy (UPS) and UV–vis spectroscopy to investigate the energy band structure information of the DNTT. Figure 3a,b shows...
secondary cut-off region and valence band edge region. The Fermi level ($E_F$) and highest occupied molecular orbital (HOMO) level of the DNTT measured by UPS were equal to $-4.42$ and $-4.97$ eV, respectively. In addition, the energy bandgap ($E_g$) of the DNTT measured from UV–vis spectroscopy was 2.61 eV (Figure 3c), and we additionally extracted that the lowest unoccupied molecular orbital (LUMO) level of the DNTT was $-2.36$ eV based on the obtained UPS and UV–vis-based HOMO level and energy band gap values. Considering that the work function ($W_F$) of Au was equal to $-4.7$ eV according to previously reported study,[31] with the energy band information of the DNTT and Au mentioned above, we represented the energy band diagrams as shown in Figure 3d. The difference between the HOMO level of DNTT ($-4.97$ eV) and the $W_F$ of Au ($-4.7$ eV) corresponding to the hole injection barrier ($\Phi_{b,hole}$) is 0.27 eV. On the other hand, the electron injection barrier ($\Phi_{b,electron}$), the difference between the LUMO level of DNTT and the $W_F$ of Au ($-4.7$ eV) is 2.34 eV, which is 8.6 times larger than that of the hole injection barrier. In the structure of the DNTT channel and Au contact electrode, the formation of a low hole injection barrier ($\Phi_{b,hole} = 0.27$ eV) and electron injection barrier ($\Phi_{b,electron} = 2.34$ eV) implies that hole injection is preferable while electron injection behavior is limited, which is directly related to the programming mechanism of the proposed OMT. The energy band diagram of the proposed OMT to explain the operating mechanism is provided in Figure 3e–g. In the erasing operation of $V_G = -100$ V, due to the low hole carriers injection barrier ($\Phi_{b,hole} = 0.27$ eV), the hole carriers injected from the Au to the DNTT penetrate the tunneling dielectric by the high gate voltage and are trapped in the CuO NPs (Figure 3e). As a result, a negative shift of the transfer curve appeared (Figure 2c). After that, when programmed under the
dark \((V_G = 100~V)\), it showed almost the same transfer curve characteristics as in E-state despite the high positive gate voltage (Figure 2d). As mentioned above, electron injection into the DNTT cannot be achieved due to the significantly high electron injection barrier, \(\Phi_{b,\text{electron}}\), as large as 2.34 eV, which resulted in no trappable electrons in the floating-gate CuO NPs (Figure 2n). The difficulty of electron injection from the Au to the DNTT still exists when programmed under light irradiation. However, when light is irradiated, even if electrons are not injected from the Au, an excitation of electron carriers occurs in the DNTT that has absorbed photon energy. At this regime, the generated electrons can be trapped in the CuO NPs by a high positive gate voltage (\(V_G = 100~V\)) (Figure 3g). The positive shift in the transfer curve appears as a result of the trapping of electrons in the floating-gate (Figure 2e). As the thickness of the DNTT increased, the absorbance increased as shown in Figure S5, Supporting Information, and the increase in absorbance means an increase in the number of excited electrons, and a positive shift of the transfer curve was available as shown in Figure S6, Supporting Information. The observed DNTT thickness-dependent memory characteristics supports that the optically-induced programming behavior is due to electron carriers generated from the light absorption of the DNTT.

Based on these results, we analyzed whether the proposed OMT distinguish and store light information depending on the light intensity. Figure 4a shows an illustration of the device structure of OMT, and Figure 4b,c illustrates the state in which holes and electrons are trapped in CuO NPs in the E-state and P-state, respectively. We initialized the OMT through the erasing operation (\(V_G = -100~V, 3~s\)) before performing the programming operation for storing light information. We used three light intensities of 880 lx \((P_{\text{inc}} = 0.13~\text{mW cm}^{-2})\), 3800 lx \((P_{\text{inc}} = 0.56~\text{mW cm}^{-2})\), and 5500 lx \((P_{\text{inc}} = 0.81~\text{mW cm}^{-2})\) to compare the memory states formed according to the light intensity. Then, we observed that the amount of positive shift in the transfer curve increased as a function of the light intensity (Figure 4d–f). The P-state of OMT was determined depending on the light intensity irradiated during programming. It is noted that we denoted the programming states as P-state i (880 lx), P-state ii (3800 lx), and P-state iii (5500 lx), respectively. We emphasize that hysteresis-free electrical characteristics were observed in all memory states regardless of the memory state. The hysteresis-free electrical properties of OMT indicated that charge trapping in CuO NPs occurs only by erasing/programming operations. As another observation, the hysteresis-free operation indicates that charge trapping did not occur at the interface between the DNTT channel and the SiO2 tunneling dielectric surface. This behavior resulted from, as mentioned previously, the interfacial trap-free fluoropolymer of CYTOP as the tunneling dielectric. Figure 4g,h shows the comparison of \(I_{\text{Read}}\) and \(V_{th}\) of memory states formed as light intensity increases (from P-state i to P-state iii), respectively. \(I_{\text{Read}}\), which was 1.24 pA in E-state, increased to 1.12 and 13 nA in P-state i and P-state ii as the light intensity gradually increased.
increased to 880 and 3800 lx, respectively. When the high intensity light (5500 lx) was irradiated, $I_{\text{Read}}$ was measured to 1.44 µA in P-state iii, about $10^6$ more than E-state, which was measured to 1.24 pA. Similarly, $V_{\text{th}}$, which was measured to −6.26 V in E-state, was positively shifted to −1.59 and −0.24 V in P-state i and P-state ii, respectively. Also, $V_{\text{th}}$ shifted positively by as large as 21 V from −6.26 (E-state) to 14.73 V (P-state iii) when irradiated with the light intensity of 5500 lx. As the increased $I_{\text{Read}}$ and $V_{\text{th}}$ shift of OMT changes incrementally as the light intensity increases, the light intensity-dependent multi-level memory operation can be available in the proposed OMTs. We measured the retention of four-distinct memory states, such as the E-state and three P-states (P-state i, P-state ii, P-state iii), respectively (Figure 4i). The four memory states were maintained unchanged for 2000 s, and in particular, the current difference between adjacent states also maintained distinguishable as large as about 100 times. This multi-level operation was obtained by the considerably high current ratio between P-state and E-state as large as $\approx 10^7$.

Next, we fabricated a 12 × 12 OMT array with a total of 144 devices on a 4-inch wafer substrate using the proposed OMT. The fabricated OMT array is shown in Figure 5a. The transfer curves of all OMTs constituting the array in the pristine state are shown in Figure 5b. Through the transfer curve of the pristine OMTs, 144 devices exhibited uniform characteristics with average threshold voltage of $−2.94 \pm 0.64$ V. It is worthy note that the optically-guided memory characteristics operated with a yield of 100% in the evaluated 144 devices, as shown in Figure 5c. All 144 fabricated devices exhibited transfer curve shift behavior depending on the light-induced electron trapping in the floating-gate. The linear-scale transfer curve, which approximates the $V_{\text{th}}$ in the E-state and P-state of all OMTs constituting the 12 × 12 array, is shown in Figure S7, Supporting Information. We also checked an interference issue between adjacent devices during the programming or erasing. The programming operation of a single OMT did not interfere with other adjacent OMTs memory states around the programmed device (Figure S8, Supporting Information). This is because...
CuO NPs, a floating-gate, were dispersed in the island form without additional patterning process, thereby separating the floating-gates between adjacent devices. Figure 5d, e shows mapping images of the $I_	ext{Read}$ when a gate voltage of 0 V is applied in the E-state and P-state, respectively. The measured $I_	ext{Read}$ mapping images confirmed that the P-state and E-state of all OMTs...
clearly distinguishes $I_{\text{read}}$ depending on the optically-guided memory behavior with a high $I_{\text{P-state}}/I_{\text{E-state}}$ of 105 (Figure S9, Supporting Information). In addition, the distribution of $V_{\text{th}}$ of E-state and P-state for 144 OMTs are given in Figure 5f,g. The $V_{\text{th}}$ distribution was equal to ~6.73 ± 0.8 V for the E-state and was equal to 30.7 ± 8.2 V for the P-state. Furthermore, we demonstrated a selectively-image detection test through the sequentially-proceeded device-to-device programming in the fabricated 12 × 12 array as shown in Figure 5h. We determined the illuminated and non-illuminated device addresses, representing 12 array as shown in Figure 5h. We determined the tially-proceeded device-to-device programming in the fabri-

The distinct alphabet “G” text image was obtained in the extracted $V_{\text{th}}$ values in a selectively programmed 12 × 12 OMT array (Figure 5i).

In summary, we demonstrated the optically-guided memory array in the 4 inch wafer-scale, consisting of 144 devices with 100% yield. Due to the cumulatively stacked small-molecules/fluoropolymer/CuO NPs structure, all 144 devices had hysteresis-free switching behaviors as well as the statistical test provided uniform $V_{\text{th}}$ distribution with 2% and 20.2% variation and of P-state and E-state, respectively. Compared with previously reported OMTs in the last 3 years (Table S1, Supporting Information),[10,13,19,32–34] our results showed the pos-

The data that support the findings of this study are available from the author.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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2. Experimental Section

Synthetic Procedure of Copper-Oxide Nanoparticles and Preparation of Coating Solution: CuO NPs were synthesized via a simple solvothermal method. Copper (II) acetate monohydrate (≥98%, Sigma–Aldrich) and tetrathiafulvalene were used as a copper precursor and a hydroxide source, respectively. First, 1.5 mmol copper precursor was dissolved in 30 mL of anhydrous ethanol (94–96%, Alfa Aesar) under vigorous stirring. At this time, 3.25 mL of TMAH solution was added to 6.75 mL of anhydrous ethanol in a separate container. After the copper precursor was completely dissolved, the precursor solution was transferred to an oil bath preheated to 75 °C. The reaction was initiated by adding the TMAH solution to the copper precursor solution dropwise at regular intervals for 10 min. The continuous condensation and hydrolysis reaction was maintained for 2 h under constant stirring at a speed of 500 rpm. As the reaction progressed, the color of the solution changed to black. The as-synthesized CuO NPs were purified and collected through centrifugation in n-hexane (extra pure, DUKSAN) at 4000 rpm for 10 min. To prepare the coating solution, CuO NPs were re-dispersed in chloroform (anhydrous, ≥99%, Sigma–Aldrich) and absolute ethanol co-solvent (chloroform, 75%, v/v) at concentrations of 5 mg mL$^{-1}$. The average size of CuO NPs in coating solution was measured to 16.8 ± 4.02 nm (Figure S10, Supporting Information).

Fabrication Process of Optically-Guided Memory Transistor: 300 nm thick SiO$_2$/Si substrate was prepared. The substrate was cleaned by sonication in ethanol and IPA for 10 min each. For the floating-gate layer, 0.35 mL of CuO NPs solution was dropped on a Si/SiO$_2$ substrate, spin-coated at 3000 rpm for 30 s, and dried at 120 °C for 30 min. Tunneling dielectric was formed by mixing CYTOP and solvent 1:10, dropping 0.2 mL, and spin coating at 1000 rpm for 30 s. In addition, annealing was performed at 120 °C for 1 h to dry the CYTOP solvent. Then, 56 nm thick DNTT and 100 nm thick Au as channel and source/drain electrodes were deposited using a thermal evaporator and patterned with a shadow mask.

Characterization: The size distribution of CuO NPs in coating solution was measured by dynamic light scattering spectrophotometer (ELS-8000, Otsuka Electronics Co. Ltd.). The memory characteristics of OMT were obtained using a Keithley 4200 semiconductor parameter analyzer in ambient conditions. The memory operation was measured according to light irradiation using multi-wavelength white LED (Figure S2, Supporting Information). The intensities of multi-wavelength white LED were classified into three levels through a manual lever. 880, 3800, and 5500 lx lights was used corresponding to $P_{\text{inc}} = 0.13, 0.56$, and 0.81 mW cm$^{-2}$ to measure the programming states (P-state i, ii, iii) according to the light intensity, respectively. White LED was only applied during the programming operation, and the erasing operation was performed under dark.
