A Flexible High-Performance Photoimaging Device Based on Bioinspired Hierarchical Multiple-Patterned Plasmonic Nanostructures

Yoon Ho Lee, Tae Kyung Lee, Hongki Kim, Inho Song, Jiwon Lee, Saewon Kang, Hyunhyub Ko, Sang Kyu Kwak,* and Joon Hak Oh*  

1. Introduction

Iconic sensory memory is a process by which the eye retains the sensations of interactions following cessation of external stimuli. Thus, it helps organisms recognize their environment and manipulate objects during performance of daily activities. The eye detects external photonic stimuli and conveys this sensory information to the brain through afferent neurons to form iconic memory, which allows organisms to remember the impressions of optical stimuli. Natural hierarchical structures of photonic systems have been identified in insects such as moths and butterflies. The insect eye is composed of units called ommatidia, which comprise a complex sensory system containing hierarchically structured components, i.e., cornea, sensory receptors, and optic nerves. The micro- and nanopatterned structures in insect eyes provide several advantages, such as light focusing by hemispherical lenses, antireflection effects by unique nanoscale surface architectures, dewetting nanostructures for waterproof characteristics, greater depth of visual field, a wide angle field of view, sensitivity to motion, and negligible aberration.

For those reasons, the ommatidia with hierarchical structured cornea play a critical role in amplifying and transferring visual signals to brain through optic nerves, enabling the spatiotemporal perception of various visual signals (Figure 1a).

Here, inspired by the sophisticated sensory system of insect eyes, we have developed flexible high-performance photoimaging device (PID), which can simultaneously detect and record incoming photonic signals at a low driving voltage (threshold voltage, $V_T$). This has been realized by combining organic photodiodes (OPDs) with hierarchical multiple-patterned plasmonic electrodes and organic nonvolatile resistive switching memory devices (ORRs) in a tandem structure (Figure 1b). Compared to devices incorporating nonpatterned and flat electrodes, the OPD arrays employing multiple-patterned plasmonic electrodes as single-layered back reflectors showed highly effective light trapping results including scattering and plasmonic properties. Furthermore, the $V_T$ of this multiple-patterned PID was less than half of that of comparable conventional devices. The resulting flexible PID showed excellent mechanical stability and well-resolved spatiotemporal mapping of incident optical signals under ambient conditions. In addition, the light information stored in the device could be easily erased and restored so that multiplex usage was possible. To date, most of the plasmonic nanostructures have been used only in a single-type organic optoelectronic device. The integration of light sensing and recording components into a single platform on a flexible substrate is of great importance for developing versatile optoelectronic and photonic systems.
importance to realize advanced light-sensing systems such as flexible photoimaging systems or electronic eyes (E-eyes). To the best of our knowledge, our results first demonstrate flexible organic optoelectronic arrays functionalized with highly effective plasmonic nanostructures.

Theoretical calculations and scanning near-field optical microscopy (SNOM) analyses were also performed to substantiate the strong plasmonic effects of the multiple-patterned electrodes. Our results suggest that the introduction of hierarchical, multiple-patterned electrodes into OPDs and PIDs is a promising means of improving the performance of optoelectronic devices by significantly enhancing light absorption. The OPDs and PIDs described herein could pave the way for flexible photonic devices due to their low cost, light weight, and mechanical flexibility. Moreover, these properties also make our PID suitable for use in portable flexible scanners, environmental monitoring systems, and E-eyes for human–machine interfaces.\[12–16\]

2. Results and Discussion

To clarify the benefits of multiple-patterned electrodes, we also prepared flat-, nanopost-, and grating-patterned electrodes for comparison. The light trapping effects of our multiple-patterned electrodes are illustrated in Figure 2a. Reflected incident light passes through the active layer multiple times by scattering (upper side of the electrode), leading to an enhancement in light absorption. The OPDs and PIDs described herein could pave the way for flexible photonic devices due to their low cost, light weight, and mechanical flexibility. Moreover, these properties also make our PID suitable for use in portable flexible scanners, environmental monitoring systems, and E-eyes for human–machine interfaces.\[12–16\]

Figure 1. a) Schematic image of insect eye. b) Schematic image of PID. The functionalities of insect eye are mimicked by an organic photosensor (ommatidium for light detection), an organic memory device (occipital lobe for visual information storage), and a hierarchical multiple-patterned plasmonic electrode (hierarchical patterned cornea for light signal amplification).

polydimethylsiloxane (PDMS) stamp, which contains various nanopost and grating patterns, was prepared by combining block-copolymer and nanoimprinting lithographies\[21\] (see more details in Methods in the Experimental Section and Figure S1, Supporting Information). Multiple patterns were combined with a 273 nm pitch grating pattern and 64 nm sized (average diameter) nanoposts to maximize the trapping of green light (530–560 nm).\[21–23\] This specified wavelength range is centrally located in the visible spectrum perceived by human eyes and is widely used in medical applications.\[24,25\] The multiple-patterned poly(3-hexylthiophene) (P3HT):[6,6]-phenyl-C₆ yên butyric acid methyl ester (PCBM) active layer of the OPD was fabricated by nanoimprinting lithography. Atomic force microscopy (AFM) revealed the formation of multiple-patterned active layers (Figure S2, Supporting Information). A nanopatterned back reflector was formed by subsequent deposition of a 100 nm thick Al electrode onto the nanostructured active layer (Figure 2b). Figure 2c shows UV–vis absorption spectra of the P3HT:PCBM active layer fabricated with different patterns without the Al electrode layer. Similar absorption behaviors were observed over the range of visible light. However, the absorption spectra of the active layers with 20 nm thick semitransparent Al layer clearly showed the enhancement in light absorption, as shown in Figure 2d. The semitransparent Al layer thinner than actual electrode thickness was used for UV–vis spectroscopic measurements. The grading-patterned and nanopost-patterned active layers exhibited enhanced absorption compared with flat active layer owing to the light-trapping effects engendered by Al layer. On the other hand, multiple patterns showed greatly enhanced absorption over the whole range of visible light.

To clarify the effects of patterns on the UV–vis absorption spectra in more details, extinction spectra were calculated for each patterned system with a 20 nm thick Al layer using the
discrete dipole approximation (DDA) method (see Calculation Method in Experimental Section, Figures S3 and S4, Supporting Information). Figure 2e shows that the extinction spectra of four types of patterned systems showed similar trends, including a range of maximum intensity (505–545 nm) that agreed with the experimental results (554 nm) shown in Figure 2d. The multiple-patterned system yielded the most intense extinction, with an enhancement of $\approx 5.33\%$ (averaged over the wavelength region of our interest) over that of a flat-patterned device. The greatest contribution to this enhancement came from the inclusion of the grating pattern, which produced a $\approx 4.12\%$ enhancement, due mainly to scattering. The effect was less evident around 635 nm, where absorption was predominant (Figure S5, Supporting Information). Nano-posts patterned with a 1 nm depth from the flat pattern produced a $\approx 0.52\%$ enhancement, while those with a 4 nm depth from the grating pattern produced a $\approx 1.21\%$ increment. Thus, the structural aspects of both the grating pattern and the depth of the nanoposts were important for the enhancement of light absorption.

The performances of our OPDs were evaluated with 532 nm light, which is the available wavelength in our system closest to the maximum absorption wavelengths of pure active layer (519 nm) and active layer with semitransparent Al layer (554 nm). A schematic image and typical current density–voltage ($J–V$) curves of these OPDs (2000 $\mu W$ cm$^{-2}$) are shown in Figure 3a and Figure S6 (Supporting Information), respectively. The enlarged $J–V$ curves clearly revealed that the enhancement in photocurrent for the multiple-patterned OPDs was greater than that of singular grating- or nanopost-patterned OPDs (Figure 3b). Figure 3c shows the on–off photoswitching characteristics of the OPDs under green light with a bias of 1 V and a 30 s duration. All of the devices maintained stable output signals under repeated on/off cycles.

To quantitatively investigate the optoelectronic properties of the fabricated OPDs, photoresponsivity ($R$, ratio of the photocurrent to the incident light power), the photocurrent/dark-current ratio ($P$), external quantum efficiency (EQE, ratio of the number of photogenerated carriers enhancing the current to that of the photons irradiated onto the channel area), and the specific detectivity ($D^*$, the smallest detectable signal) were calculated (see more details in Note S1, Supporting Information). The key parameters of OPDs with different patterns are summarized in Table 1 and Table S1 (Supporting Information). The $R$ and $D^*$ values of OPDs, as a function of light intensity, are also presented in Figure S7 (Supporting Information). Enhancements in $R$ and $D^*$ significantly increased as the light intensity decreased, which is a commonly observed phenomenon.\textsuperscript{[26,27]}

Under a light intensity of 20 $\mu W$ cm$^{-2}$, $R$ was improved from 1.48 A W$^{-1}$ for flat OPDs to 7.95 A W$^{-1}$ for multiple-patterned OPDs.
OPDs, while those of grating-only and nanopost-only OPDs were 3.80 and 2.03 A W\(^{-1}\), respectively. It indicated that the enhancement in the photocurrent of the multiple-patterned electrode was greater than those of the grating and nanopost electrodes. Moreover, \(D^*\) was enhanced from 1.6 \(\times\) 10\(^{12}\) Jones for flat OPDs to 11.4 \(\times\) 10\(^{12}\) Jones for multiple-patterned OPDs, while those of grating-only and nanopost-only OPDs were 4.2 \(\times\) 10\(^{12}\) and 3.9 \(\times\) 10\(^{12}\) Jones, respectively.

Furthermore, to explore their potential applicability as flexible sensor platforms, OPD-based sensors were fabricated with indium tin oxide (ITO)-coated polyethylene naphthalate (PEN) substrate (Figure 3d). Electrical characterization was conducted

<table>
<thead>
<tr>
<th>OPD</th>
<th>EQE [%]</th>
<th>(R) [A W(^{-1})]</th>
<th>(P)</th>
<th>(D^*) [Jones]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat</td>
<td>344.1 (±137.8)(^c)</td>
<td>1.48 (±0.59)</td>
<td>11.6 (±4.6)</td>
<td>1.6 (\times) 10(^{12}) (±0.6 (\times) 10(^{12}))</td>
</tr>
<tr>
<td>Grating</td>
<td>884.8 (±128.8)</td>
<td>3.80 (±0.55)</td>
<td>29.6 (±4.3)</td>
<td>4.2 (\times) 10(^{12}) (±0.6 (\times) 10(^{12}))</td>
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<tr>
<td>Nanopost</td>
<td>473.5 (±142.6)</td>
<td>2.03 (±0.61)</td>
<td>47.1 (±14.2)</td>
<td>3.9 (\times) 10(^{12}) (±1.2 (\times) 10(^{12}))</td>
</tr>
<tr>
<td>Multiple</td>
<td>1854.3 (±245.0)</td>
<td>7.95 (±1.05)</td>
<td>106.1 (±14.0)</td>
<td>11.4 (\times) 10(^{12}) (±1.5 (\times) 10(^{12}))</td>
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<thead>
<tr>
<th>PID</th>
<th>(V_T) (V)</th>
<th>(I_{off}) (A, at 1 V bias)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat</td>
<td>5.0 (±0.4)</td>
<td>2.4 (\times) 10(^{-8}) (±0.7 (\times) 10(^{-8}))</td>
</tr>
<tr>
<td>Grating</td>
<td>3.2 (±0.1)</td>
<td>178.2 (\times) 10(^{-8}) (±43.2 (\times) 10(^{-8}))</td>
</tr>
<tr>
<td>Nanopost</td>
<td>3.7 (±0.1)</td>
<td>13.6 (\times) 10(^{-8}) (±3.5 (\times) 10(^{-8}))</td>
</tr>
<tr>
<td>Multiple</td>
<td>2.2 (±0.6)</td>
<td>1497.8 (\times) 10(^{-8}) (±481.2 (\times) 10(^{-8}))</td>
</tr>
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\(^c\)The standard deviation. These parameters were averaged from at least five devices.
at various bending radii ($R_b$s) to assess the mechanical flexibility and operational stability of the OPDs. Bending tests were performed at four $R_b$s: 9, 5, 2, and 1.5 cm. Figure 3e shows the average $I_{\text{light}}$ and $I_{\text{dark}}$ of the devices measured at various $R_b$s, even down to 1.5 cm. Average $I_{\text{light}}$ and $I_{\text{dark}}$ were monitored for up to 1000 cycles, with an interval of 100 cycles at an $R_b$ of 2 cm (Figure 3f). Both factors showed stable retentions throughout repetitive bending tests, indicating that the developed OPDs were mechanically and operationally stable. Furthermore, a $7 \times 7$ multiple-patterned photosensor matrix was fabricated using the same device configuration and fabrication process of the unit device. Figure 3g,h shows schematic and photographic images of the fabricated OPD matrix, respectively. The center of the sensing matrix was illuminated with a green laser source and the photonic signal was illustrated by a 2D histogram as a function of the $P$ value, as shown in Figure 3i. $P$ values over 4000 were observed only from the unit device around (4, 4), which corresponded to the illumination spot of incident laser. This result indicates that these multiple-patterned devices, and their associated fabrication processes, can potentially be used for making low-cost, flexible, and high-performance optoelectronic devices.

We also theoretically investigated the distributions and average intensities of the induced electric field (i.e., $|E(\omega)|^2/E_0^2$) in the OPD systems (Figure 4a) constructed with a 100 nm thick Al layer (Figure S3, Supporting Information). In all of the patterned systems, the electric field was concentrated at the interface between the Al and P3HT:PCBM layers. The multiple patterns showed higher electric field intensities in both the Al and active layers. Interestingly, the electric field in the middle of the active layer in the grating-patterned system was locally enhanced in a periodic fashion along the top of the Al grating, where surface plasmons were greatly generated. The enhancement was more increased by incorporating nanopost patterns, which induced large generation of excitons in the active layer side, as confirmed by backward scattering data (Figure S8, Supporting Information). To further investigate the effects of multiple patterns, we described the difference plot of $|E(\omega)|^2/E_0^2$ intensities of multiple- and grating-patterned systems (Figure 4b). It showed that the $\Delta|E(\omega)|^2/E_0^2$ intensity was increased at the interface between Al and active layers ($\Delta|E(\omega)|^2/E_0^2_{\text{max}} = 16.7$) and at the middle of active layer ($\Delta|E(\omega)|^2/E_0^2_{\text{max}} = 0.15$). Consequently, multiple patterns induced the largest electric field enhancements. This was likely due to increased excitons in the active layer via scattering by surface plasmons of Al layer and consequently the OPD efficiency was much improved. By investigating the effect of electric field direction, we further found that the grating pattern exhibited dominant response to the light due to $y$-direction polarization (Figure S9, Supporting Information).

For quantitative understanding of the effects of multiple-patterned electrodes, we also investigated the local electric field distributions on patterned electrodes using SNOM. AFM topographs and the near-field optical images for Al electrodes are presented in Figure 5 and Figure S10 (Supporting Information). The flat and nanopost-patterned electrodes showed similar SNOM images (and optical properties), presumably due to the AFM resolution limits (probe aperture diameter = 60 nm). However, the SNOM images of the grating patterns exhibited a high scattering intensity at the trough (green rectangles), while those of multiple-patterned electrodes exhibited the high scattering intensity at the crest (blue rectangles; 532 nm wavelength). These results indicated that, while the intensity of electric field of the grating pattern was focused on the trough, the intensity of electric field of the multiple-patterned electrode was higher at the crest due to the larger sized nanopatterns. Overall, the theoretical calculation and SNOM analysis agreeingly support that utilizing the multiple patterns is highly promising for

**Figure 4.** a) $|E(\omega)|^2/E_0^2$ distributions and $|E(\omega)|^2_{\text{avg}}/E_0^2$ intensity profiles according to height of each patterned system with 100 nm thick Al layer. $k$ (black arrow) and $E$ (green arrow) indicate the directions of incident light and electric field, respectively. b) Difference plot of $|E(\omega)|^2/E_0^2$ intensities between multiple- and grating-patterned systems. Note that all results are calculated at 532 nm wavelength.
enhancing the plasmonic effect and eventually the performance of optoelectronic devices. Grazing incidence X-ray diffraction analysis of patterned P3HT:PCBM films also indicated that the change of molecular packing during the nanoimprinting process was not a major factor in determining the performance of OPDs (see more details in Note S2, Figures S11, S12 and Table S2, Supporting Information).

We also fabricated a flexible multiple-patterned PID by integrating multiple-patterned OPDs with ORSs. A schematic diagram and photograph of flexible PID consisting of a PEN/ITO/PEDOT:PSS/P3HT:PCBM/Al/tris(8-hydroxyquinolinato) aluminum (Alq₃)/Au structure are presented in Figure 6a, b, respectively. The I–V characteristics of separate OPD and ORS devices are shown in Figure S13a, b (Supporting Information), respectively. Voltage applied to the PID (V_{APPLIED}) led to a voltage drop across the OPD and ORS, depending on the specific resistance. Under light illumination, significant changes of resistance in the OPD, which occurred upon light illumination, resulted in changes in V_{APPLIED} across the ORS. The ORS was changed from a high resistance state to a low resistance state (LRS) and the incoming photonic signal was retained by the ORS, while V_{APPLIED} to the ORS was not sufficient to retain light information under dark conditions (Figure 6c).[4] Therefore, the enhancement in photocurrent supplied by the OPD resulted in a reduced V_T of PID, allowing for a lower driving voltage (see more details in Note S3, Figure S13c, Supporting Information).

Figure 6d shows the I–V characteristics of the PID based on different patterns under a reverse bias. In the multiple-patterned PID, V_T was significantly reduced from 5.0 V (flat) to 2.2 V due to the enhanced photocurrent. In contrast, V_T values of Al-grating- and Al-nanopost-only PIDs were 3.2 and 3.7 V, respectively (Table 1). These results are in line with the trends observed in the OPDs and the theoretical calculations. Furthermore, the PIDs showed stable switching endurances for multicycle usage (Figure S14, Supporting Information). To investigate the capability of simultaneous detection and storage of incoming photonic signals, a multiple-patterned PID composed of an array of 7 × 7 pixels was used to record ‘P’ patterned laser light through a mask (140 mW cm⁻²). The I–V characteristics of the elements in the array are given in Figure S15 (Supporting Information). As illustrated in Figure 6e, devices only within the illuminated P-patterned area could be programmed to LRS by a reverse voltage sweep over 2.2 V (V_T), providing a well-resolved spatiotemporal photomap. In this device, the illuminated light could not only be detected, but also recorded by the memory array. Promisingly, the flexible device was able to retain the current mapping of the incoming photonic signals after 1000 bending cycles (Figure 6e, R_b = 2 cm) and for 2 days (Figure 6f). Furthermore, such light information stored in the device could be erased easily and reprogrammed to allow multicycle usage.

3. Conclusions

In summary, we developed insect eye-inspired flexible PIDs with low operation voltages by vertically stacking OPDs and ORSs, where a multiple-patterned back reflector triggering
Various plasmonic effects were incorporated into the OPD. Compared with flat-electrode OPDs, the multiple-patterned OPDs showed greatly enhanced $R$ (by more than 437%), whereas single-patterned OPDs composed of either grating patterns or nanoposts exhibited 157% and 37% increments in $R$, respectively. We confirmed the plasmonic characteristics of these devices using theoretical calculations and SNOM analyses. A flexible multiple-patterned $7 \times 7$ photosensor matrix showed the capability of a highly sensitive spatiotemporal photomapping with high mechanical durability, demonstrating its high potential for use in flexible image sensors. We also fabricated multiple-patterned plasmonic PIDs, which can simultaneously detect and store light information. The $V_T$ of the multiple-patterned PIDs was reduced by more than 50% compared to the...
flat-electrode system. Furthermore, these flexible PIDs retained light signal mapping capability over 1000 bending cycles and even for 2 days while maintaining the ability to be erased and rewritten. This multiple-patterned plasmonic electrode can be easily applied to a large area and is highly suitable for fabricating high-performance and low-driving-voltage photosensing systems. This system is expected to open up a new avenue for realizing next-generation photonic systems, such as E-eyes for humanoid robots and human–machine interfaces, flexible photosensors, and wearable medical diagnostic systems.

4. Experimental Section

Experimental Method: Fabrication and Characterization of Multiple Plasmonic Patterns. As illustrated in Figure S1 (Supporting Information), a polystyrene (PS)-b-poly(methyl methacrylate) (PMMA) solution (2 wt% in toluene) was spin coated (~70 nm thickness) onto a SiO2/Si wafer. Then, the substrate was annealed for 48 h at 180 °C in vacuum oven. A grating-patterned PDMS stamp (~7 mm thickness) was obtained by pouring a PDMS precursor solution (1:10 ratio of curing agent to silicone elastomer) onto a fluorine treated grating mold (Thorlab, GH13-36U, Periodicity = 278 nm) and curing at 60 °C for 2 h. The grating-patterned PDMS stamp was placed on to PS-b-PMMA layer and imprinted for 10 min at 130 °C. After cooling of substrate at room temperature, the PDMS stamp was lifted off. Then PMMA was selectively etched with a traditional UV/acetone acid treatment to make multiple-patterned PS film. Subsequently, inductively coupled plasma (ICP) etching was performed for 5 min to remove the SiO2 layer using multiple-patterned PS pattern as the mask. (CF3/CHF3/Ar flow rate was 10/30/10 sccm), then the multiple-patterned SiO2 wafer was cleaned and treated with fluorinated self-assembled monolayer. To make a multiple-patterned PDMS stamp, conventional liquid PDMS mixture was poured consecutively on the substrate. AFM images of flat, nanopost, grating, multiple-patterned PDMS stamps, and size distribution analysis of multiple patterns were investigated at our previous paper.[21] After baking at 60 °C for 2 h, the solidified multiple-patterned PDMS stamp could be easily obtained. The scanning electron microscope (SEM) images were obtained using a Hitachi cold SEM microscope. The AFM images and thickness were obtained using a Veeco AFM microscope and Park System NX10 in a tapping mode. ICP etching was carried out using a Sntek Dry Etcher, ICP.

Fabrication and Characterization of OPDs: For OPDs, photodiodes were fabricated with the structure of glass (or PEN)/ITO/PEDOT:PSS/P3HT:PCBM/Al. PEDOT:PSS (Heraeus) was spin coated onto a precleaned ITO at 5000 rpm for 40 s and then dried at 130 °C for 30 min. Subsequently, a dichlorobenzene (ODCB) solution containing P3HT (~50% regioregularity) and PCBM (~95% regioregularity) was spin casted at 2000 rpm for 60 s, followed by annealing at 80 °C for 30 min. For patterning process of photoactive layer, the patterned PDMS mold was placed on the photoactive layer under pressure in the annealing process. After removing the PDMS mold, 100 nm Al intermediate electrode was thermally evaporated on the photoactive layer under a vacuum (~10−6 Torr) without mask. For 7 × 7 array, 300 nm Alq3 (Sigma-Aldrich, without mask) and 80 nm Au (with mask) were thermally evaporated. The area of the Au electrode defined the active area of the device as 0.84 mm2. The current–voltage characteristics of PIDs were measured in ambient condition using a Keithley 4200-SCS semiconductor parametric analyzer. (the compliance current at reverse bias was defined as 1000 μA).

SNOM Measurement: SNOM images were obtained using a confocal SNOM/AFM microscope (Alpha 300, WITec). Al electrodes were scanned in the SNOM contact reflection mode with a set-point of 1 V. The scanning size and velocity at the SNOM image was 1.5 × 1.5 μm and 3 μm s−1. A 532 nm laser wavelength was coupled to the subwavelength aperture (60 nm in diameter) to generate the optical near field on metal surfaces. The scattered light from the sample was collected by a microscope objective (0.4 NA, 20 x). These SNOM images were simultaneously recorded with the AFM topography images for direct correlations. Each patterned Al electrode was prepared by thermal deposition process of Al on each patterned SiO2/Si wafer.

Measurement of The Complex Refractive Index of Semiconductor: The complex refractive index of semiconductor was measured with ellipsometer (J.A.Woollam). P3HT:PCBM layer was prepared by spin-coating process on the hydrophilic treated SiO2/Si wafer (4 nm thick SiO2/Si).

Calculation Method: Discrete Dipole Approximation. In this study, we used the DDA method[22–30] to calculate optical properties (i.e., extinction, scattering, and absorption) and |E(ω)|2/E2 distributions of the four patterned systems (i.e., flat, grating, nanopost, and multiple patterns). Briefly, in the DDA method, the optical properties of target system are calculated by dipole moments, which are induced by discretizing the system into dipole points under local electric field. An essential criterion should be adopted for reliable calculation; |m|/kd<1, where |m| is a complex refractive index, k is a reciprocal value of incident wavelength, and d is a dipole spacing between adjoining dipoles. d was set to be 1 nm in this study. To perform the DDA calculation, DDSCAT program (ver 7.3.0) developed by Drain and Flatau[31] was used.

Model Systems: Figure S3 (Supporting Information) shows the structural information of model systems, where periodic boundary conditions are applied to x- and y-axis directions. The ambient environment is air. In the Figure S3a,b (Supporting Information), flat-type system used for flat and nanopost patterns and grating-type (i.e., hemiellipsoid shape) system used for gratings and multiple patterns are presented, respectively. For the flat and grating systems, the length in x-axis is 1 nm. The complex refractive indexes of Al and P3HT:PCBM are obtained from Palik’s handbook[31] and our experiment measurement (Figure S16, Supporting Information), respectively. For the calculation of optical properties, the thickness of Al layer (i.e., thickness in Figure S3a,b) is set to be 20 nm. For the calculations of |E(ω)|2/E2 distributions and angular far-field scattering, the thickness of Al layer is set to be 100 nm. Figure S3c,d (Supporting Information) shows the random patterns for nanopost- and multiple-patterned systems, respectively, where the depth of random patterns are 1 and 4 nm, respectively. These patterns are introduced into the interfaces of flat and grating-type systems of Al and P3HT:PCBM layers. Notably, for the random patterned systems, we investigated the influence of random shape of pattern on the extinction spectra and the |E(ω)|2/E2 distributions by considering three types of patterns (Figure S4a, Supporting Information). The shape of pattern induced marginal effects on extinction spectra (Figure S4b,c, Supporting Information) and |E(ω)|2/E2 distributions (Figure S4d,e, Supporting Information) because the depths of pattern were too thin (i.e., 1–4 nm) compared.
to the thickness of Al and P3HT:PCBM layers. For that reason, we used the pattern type I as the representative pattern for the nanopost- and multiple-patterned systems.

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.

Keywords
discrete dipole approximation calculation, organic optoelectronic devices, photoimaging devices, plasmonic effect, scanning near-field optical microscopy

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