Polarization-Sensitive Photodetector Using Patterned Perovskite Single-Crystalline Thin Films

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Hybrid organic–inorganic perovskite single crystals, compared with their polycrystalline counterparts in thin films, are free from grain boundaries and have fewer defects, and therefore are promising in high-performance optoelectronic devices. Recently, the crystal-structure anisotropy in perovskites has been utilized to achieve polarization-sensitive photodetectors. Here, it is shown that under 532 nm linearly polarized illumination, although MAPbBr$_3$ (MA$=$CH$_3$NH$_3$)$^+$ single crystals possess weak in-plane anisotropy, nanogratings made on the crystal surface lead to high polarization photodetection sensitivity of $\approx$2.2 comparable with that of the isotropic perovskite crystals and nanowires, while enhancing both the photodetection responsivity and external quantum efficiency. The surface nanopattern induced polarization sensitivity may find interesting applications in future optoelectronic devices.

1. Introduction

Photodetectors are important optoelectronic devices, and are widely used in communication, imaging, sensing, and spectroscopy.[1] According to the orientation of electromagnetic wave oscillation, light can be divided into different polarization categories.[1a] The non-polarized photodetectors are usually only sensitive to the intensity and wavelength of the incident light, while polarized photodetectors are explored to directly detect polarized light in addition to the intensity and wavelength.[2] Polarized-light detection is important in applications spanning optical communications, optical switching, polarization sensors, and optical radar.[3] Generally, the polarization-sensitive properties in a photodetector arise from either inherent crystal-structure anisotropy (e.g., 2D anisotropic materials[13,4]) or extrinsic geometric effects (e.g., 1D nanowires[5]). For crystal-structure anisotropy, the atoms’ alternative arrangements vary in a 3D space, therefore, leading to the changes of anisotropic material physical and chemical properties according to the observation direction.[6] The mainstream of polarization-sensitive photodetectors using highly anisotropic materials is dominated by inorganic 2D systems (e.g., black phosphorus,[3e,7] metallic graphene,[4b] GeSe$_2$,[8] and MoS$_2$[9]) and heterojunctions.[10,11] However, the synthesis process of these anisotropic materials is complex and potentially expensive.[13] In addition, nanowire based polarization-sensitive photodetectors were also reported, including InP,[12] core/shell GaAs/AlGaAs,[13] Si,[13,14] and GeO$_2$–Ge,[14] however requiring complex high-cost fabrication processes.

Halide perovskites, most notably MAPbX$_3$ ($X =$ Cl$^-$, Br$^-$, or I$^-$), have recently become promising optoelectronic candidates, due to their fascinating physical properties of high absorption coefficient ($\approx 10^4$ cm$^{-1}$), low trap density ($10^3$–$10^{20}$ cm$^{-3}$) and long carrier diffusion length ($\approx 100$ nm–100 $\mu$m).[15] Profiting from the simple synthesis process and their outstanding physical properties, perovskite-based photodetectors have been widely studied.[16] The chemical vapor deposition deposited polycrystalline perovskite photodetectors show good stability and can be self-powered.[17] By using spin-coating method, the facet-dependent photovoltaic[18] and charge transport[19] anisotropies in perovskite polycrystalline thin films were explored. Furthermore, the solution-processed single crystal[13,14,16,20] and nano/microwire[18,21] perovskite polarization-sensitive photodetectors were also studied. For example, Zuo et al.[22] and Ding et al.[20a] reported different optoelectronic properties of MAPbBr$_3$ and MAPbI$_3$ from different perovskite crystal facets, respectively. Afterward, facet-dependent optoelectronic anisotropy properties of MAPbCl$_3$ single crystal were studied by Cheng et al.[22] Ding and co-workers revealed the in-plane polarization-sensitive phenomena of MAPbBr$_3$ and MAPbCl$_3$ crystals.[23a] Beyond the optoelectronic anisotropies in bulk single crystals, Gao et al. designed MAPbI$_3$ nanowire arrays as highly polarization-sensitive photodetectors.[21a] Apart from these organic–inorganic perovskite nano/microwire
photodetectors, all-inorganic\textsuperscript{[1a]} and 2D\textsuperscript{[21c]} perovskite nano/microwire array polarized photodetectors were also reported.

Semiconductor surface patterning has been widely applied in integrated optoelectronic devices and optical/optoelectronic circuits.\textsuperscript{[22]} Many surface patterning attempts have been made to improve the properties of Si MSM photodetectors or polarization-sensitive photodiodes with metallic nanostructure arrays,\textsuperscript{[23]} such as Si photovoltaic cells and photodetectors,\textsuperscript{[24]} and GaAs MSM photodetectors.\textsuperscript{[25]} These processes are normally accompanied by reactive ion etching, electron–beam lithography, or chemical wetting etching technologies. These surface patterning technologies are hard to be utilized for halide perovskite monocrystalline thin film surface due to its sensitivity to high temperature\textsuperscript{[26]} and polar solvents.\textsuperscript{[27]} Herein, we study the surface nanopatterned single-crystalline perovskite thin-film photodetector performance and the polarization sensitivity at a certain wavelength. We use a non-destructive epitaxial growth method we reported elsewhere\textsuperscript{[28]} to grow surface nanopatterned MAPbBr$_3$ single-crystalline thin films as polarization-sensitive photodetectors. According to photodetectors with Au–MAPbBr$_3$–Au type, photocurrents, responsivity ($R$), and external quantum efficiency (EQE) are performed to reveal the pattern-induced anisotropic optoelectronic properties.

2. Results and Discussion

2.1. Nanopatterned Monocrystalline Thin Film

As a proof-of-concept experiment, using the method described in ref. [28], we fabricated nanopatterns on single-crystal perovskite thin films for polarization-sensitive photodetection. The pattern was copied from a DVD surface. The nano-gratings on DVD surfaces were first transferred to polydimethylsiloxane (PDMS) and then the patterned PDMS was used as the template (see Experimental Section for details). Surface-nanopatterned single-crystalline MAPbBr$_3$ thin films were characterized using scanning electron microscopy (SEM) and atomic force microscopy (AFM). Figure 1a shows the ultra-smooth surface of the nanopatterned thin film. Several micrometers thick patterned perovskite thin film can be easily realized. All these perovskite single-crystalline thin films exhibit sharp edges (e.g., Figure 1b). More SEM images, for example, the DVD master and a thinner film ($\approx 2$ $\mu$m), are presented in Figure S1, Supporting Information. To characterize the depth information of the surface patterned perovskite single-crystalline thin films, AFM was performed on randomly selected 10 $\mu$m $\times$ 10 $\mu$m area. As indicated in Figure 1c,d, a fine grating structure on the smooth patterned thin film surface and an $\approx 80$ nm depth is clearly revealed.

![Figure 1](image-url)

**Figure 1.** Morphology characterizations of surface-nanopatterned single-crystalline MAPbBr$_3$ thin films. a) Top-view SEM images of surface nanopatterned MAPbBr$_3$ single-crystalline thin films. b) Clear and sharp edges of nanopatterned sample. c) AFM image of nanopatterned sample and d) the corresponding cross-section profile.
Figure 2a illustrates the X-ray diffraction (XRD) patterns of powder MAPbBr₃ and the (100) plane of surface nanopatterned MAPbBr₃ single-crystalline plane. The XRD pattern of MAPbBr₃ powder is indexed as cubic symmetry (Pm-3m) and the MAPbBr₃ lattice parameters are calculated to be 5.92 Å. The sharp peaks of {100} crystal-plane suggest a high quality of nanopatterned MAPbBr₃ single-crystal thin film. The surface patterned sample is also characterized via transmission electron microscopy (TEM), as seen in Figure 2b, and Figure 2c is the corresponding selective area electron diffraction (SAED) image from Figure 2b, which exhibits only reflections related to the cubic structure with viewing direction along the [001] zone axis and high crystalline quality with a measured d spacing of 2.50 Å to the plane (210). To further confirm the single-crystal nature of the perovskite thin film, the surface patterned perovskite thin film was investigated by electron backscatter diffraction (EBSD). In the EBSD measurements, to reduce the damage from electron beam irradiation on nanopatterned sample, we selected a larger area of ≈50 µm × 50 µm containing many striped nanostructures. To prove the whole thin-film is the same coherent single-crystal structure, translation-independent regional mappings of Kikuchi patterns were conducted. As illustrated in Figure 2d, EBSD images were taken on a surface nanopatterned single-crystalline thin film (≈3 mm) from different surface locations, where the single crystal identical crystallographic orientations were seen clearly.

The chemical states of different elements in the surface nanopatterned thin film were investigated using X-ray photoelectron spectroscopy (XPS). As presented in Figure S2, Supporting Information, XPS diagrams of the patterned MAPbBr₃ thin film show strong signals of Pb, Br, C, and N elements, which prove the presence of these corresponding elements in the sample. The diagrams of Pb 4f, C 1s, and Br 3d show the stable chemical states of these elements.

2.2. Photodetector Performance

The photodetectors were fabricated on both planar and nanopatterned (the same as DVD structures) single crystal thin films. Figure 3a shows the photocurrent measurement schematic diagram of a photodetector based on MAPbBr₃ single crystal under linearly polarized light. To study the effect of linearly polarized light on the photoelectric response of the planar device, those two Au electrodes are designed to be as parallel as possible to the two edges of the monocrystalline thin film. Therefore, the interaction between polarized light and the crystal surface is defined. As a control experiment to show that rotating the polarizer has no influence on the output light power, the relationship between power density of 532 nm light (50 mW cm⁻², filtered from solar simulator) and polarization angles is shown in Figure 3b. It is obvious that the power density is stable under different polarization angles. Figure 3c,d are the photocurrents of MAPbBr₃ single-crystalline thin-film photodetectors without and with surface patterns, respectively, under the linearly polarized light illumination. Dark currents of ≈10⁻⁹ A at an applied voltage of 10 V were seen for both non-patterned and patterned MAPbBr₃ photodetectors due to the high-quality single crystals.[29] Following the linearly polarized light illumination, the photocurrent increases with the rising of the applied voltage for MAPbBr₃ single-crystalline thin-film photodetectors, owing to the high carrier concentration by...
photoexcitation. Besides, different photocurrent features with various polarization angles were observed between planar and nanopatterned photodetectors. Under 532 nm illumination, compared to the non-patterned perovskite single-crystal thin film, the patterned one reveals significant variations in photocurrents at different polarization angles, suggesting that surface gratings enhance anisotropic optoelectronic effects. The maximum photocurrent of the photodetector appears when the light polarization is parallel to the grating lines axial direction and it gradually decreases to its minimum value until the polarization angle reaches 90° (the 0° polarization is defined as the direction parallel to the grating lines or perpendicular to the electrodes where a maximal photocurrent is received for the same bias voltage, as shown in Figure 3a). A further increase in the polarization angle improves the device photocurrent and it totally recovers at 180° polarization, which clearly proves that the photodetector optoelectronic performances exhibit a 180° periodicity. The periodicity is consistent with the polarization angle of the incident light and the direction of the grating lines.

We evaluated both the R and the EQE for the polarization-sensitive photodetectors. The responsivity is given by $R = (I_{\text{light}} - I_{\text{dark}})/P \times S$, where $I_{\text{light}}$ and $I_{\text{dark}}$ are the photodetector current under 532 nm light illumination and dark condition, respectively, $P$ is the illuminant power intensity, and $S$ is the effective area. The EQE is calculated by $\text{EQE} = (R\hbar c)/(e\lambda)$, where $R$ is the responsivity calculated above, $\hbar$ is the Planck constant, $c$ stands for the light velocity, $e$ is an electron charge, and $\lambda$ is the light wavelength. Based on the above formulae, we analyzed the photocurrent conversion capability of the planar and nanopatterned photodetectors fabricated on MAPbBr$_3$ single crystal (100) plane.

The photodetectors’ R and EQE values under polarized lights illumination are presented in Figure 4. Under different polarization-angle irradiation and a power of 50 mW cm$^{-2}$, the planar photodetector’s $R$ and EQE vary in the range 11.4–15.4 mA W$^{-1}$ and 26.6–35.9%, respectively at a 10 V bias. Although the $R$ and EQE are changing accordingly to the polarization angles, such differences are relatively small compared with those of the patterned single-crystal perovskite thin films under 532 nm polarized irradiation. For example, under 10 V bias, the nanopatterned photodetector’s $R$ and EQE can be 35.0 mA W$^{-1}$ and 81.6%, respectively, which are much higher than the planar one. To avoid individual difference, $I$–$V$ curves of three nanopatterned photodetectors and three planar photodetectors at the polarization angle 0° were measured, respectively, and the $R$ and EQE were calculated accordingly, as presented in Figure S3, Supporting Information. These phenomena remarkably demonstrate that the surface nanopatterned (100) plane is much more sensitive to linearly polarized illumination and is more efficient in light collection and optoelectronic performances. Besides, for the nanopatterned photodetector (Figure 4b), the prominent differences of $R$ and EQE between 0°/180° and 90° polarizations are observed. For example, these values at 0°/180° are more than two times compared to those

Figure 3. a) Schematic diagram of the photodetector using a MAPbBr$_3$ nanopatterned (the same as DVD structures) single crystal (Au electrodes with a channel of 1 mm in width and 30 µm in length) devices under linearly polarized light. b) Power density versus polarization angles. c) Photocurrent versus voltage plots ($I$–$V$ curves) of the non-patterned device in the dark and under 50 mW cm$^{-2}$ 532 nm illumination. d) Photocurrent versus voltage plots ($I$–$V$ curves) of the nanopatterned photodetector in the dark and under 50 mW cm$^{-2}$ 532 nm illumination.
at 90°. This suggests that 0°/180° linearly polarized light under 532 nm illumination absorbs more light. Normalized photore-
sponse of the nanopatterned and planar photodetectors were
employed to obtain the rise time and fall time. The rise time
and the fall time of the nanopatterned photodetector were ≈0.10
and 0.13 s, respectively, while the rise time and the fall time of
the planar photodetector were ≈0.28 and 0.13 s, respectively, as
shown in Figure S4, Supporting Information. Limited by our
apparatus during the transient photocurrent measurements,
the rise time and the fall time should not be regarded as our
photodetectors’ inherent response speed. The response time is
expected to be shorter and on par with the carrier lifetime.\[17a]\nMeanwhile, we find that the maximal polarization sensitivity
(over 2.2) can be obtained at a bias voltage of 1.11 V, which is
defined as the 0°/180° polarized photocurrent divided by the
90° polarized photocurrent. Figure 4d plots the sensitivity-
related curves under the illumination of 532 nm polarized light
at different bias voltage. The polarization-dependent normalized
photocurrent is given in Figure 4e, exhibiting a periodicity
of 180° and the normalized photocurrent of >2.2 at both polarization
angles of 0° and 180°. To know the stability of our device,
the photocurrent of a surface nanopatterned single-crystal
perovskite thin film stored in the air under ambient condition
for 7 days without any encapsulation was characterized for
comparison. As illustrated in Figure S5, Supporting Information,
the average stability of the photodetector can be over 85% after being stored in air for 7 days.

Polarization-sensitive photodetection based on nanopat-
terned single-crystal perovskite thin films is reported in our
work. Previously, the photoelectric anisotropy was shown as
a result of the anisotropy of the crystal surface. It is generally
believed that, for non-patterned single crystals, charge-density
differences and linearly polarized light absorption are the two
factors contributing to the optoelectronic anisotropy under
linearly polarized light. To understand the influence of the
MAPbBr$_3$ (100) facet crystal structure on the optoelectronic an-
isotropy properties, the band structure (Figure S6, Supporting
Information) and the 2D (Figure 5a) and 3D (Figure 5b)
charge–density maps were calculated. The calculated MAPbBr$_3$
narrower band gap (2.087 eV) is attributed to the GGA corre-
lation potential and exchange. Although the MAPbBr$_3$ pos-
sesses a cubic crystal structure, a weak anisotropic feature of
charge densities for (100) plane is observed. It can be seen that
the electron densities are different along the defined polariza-
tion angles of 0° and 90°, which may be due to the CH$_3$NH$_3^+$
groups’ random distributions in the crystal lattice.\[34\] In addi-
tion, the imaginary part of the dielectric function can lead to
the linearly polarized light absorption, which may result in anis-
otropic optoelectronic properties.\[35\] As shown in Figure 5c,
it can be seen that the light absorption below 450 nm has dif-
ferent features. However, at ≈532 nm which is also near the
absorption threshold for MAPbBr$_3$ single crystal, the imaginary
parts of the dielectric function show weak difference along var-
dious polarization angles. Here, we believe that the small differ-
ences in charge density diversities in (100) planes can lead to
differences in light absorption, electron–hole pair generation,
charge transfer, and recombination after photoexcitation.\[36\]
The above explanations are consistent with the observed exper-
imental result in non-patterned photodetector.

For the nanopatterned single-crystal perovskite thin films,
the polarization-dependent optical absorption is enabled by the
surface nanogratings. As illustrated in Figure 5d, the 0°
and 180° polarized light show a higher optical absorption which
responds to the polarization direction along the grating line’s
axial direction. With the gradual rotation of the polarized light
from 0°, the absorption intensity decreases to its minimum

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**Figure 4.** a,b) R and EQEs under polarized illumination for planar and grating MAPbBr$_3$ photodetectors, respectively. c) Sensitivity versus bias voltage for the polarization angle of 0°. d) Polarization dependence of normalized photocurrent at 1.11 V.
value at 90°. These simulated results are in good agreement with the observed phenomena in our experiments mentioned above, and suggest that the polarized photodetection is achieved by structure-induced polarization-dependent optical absorption. Such an interesting anisotropic absorption phenomena can be attributed to the different dielectric constants in different directions of the grating lines and the surrounding air due to the high aspect ratio of the grating lines.\[5a,21b\] The performances of our nanopatterned polarized-sensitive single-crystal perovskite thin film, including sensitivity, responsivity, detectivity, and response capacity, are summarized in Table 1.

3. Conclusion

In summary, we have developed and demonstrated polarization sensitive photodetection based on surface nanopatterned single-crystal MAPbBr\(_3\) perovskite thin films. Using planar and nanopatterned single-crystal perovskite thin films, we investigate the crystal orientation within (100) plane induced optoelectronic anisotropy. Unlike the weak polarization sensitivity of the planar photodetector at 532 nm, the nanopatterned single-crystal perovskite thin films show polarization sensitive photocurrents, responsivity, and EQE. The experimental and simulation results in our work reveal that surface-grating structures enhance light absorption according to the polarization direction of the light. The observed strong polarization sensitivity as a result of the surface grating orientations may find useful optoelectronic applications.

4. Experimental Section

**Chemicals and Reagents:** MABr (99%), PbBr\(_2\) (≥98%), lead iodide (99%), and DMF (anhydrous, 99.8%), were purchased from Sigma-Aldrich. Dow Corning Sylgard 184 kit was purchased from Ellsworth Adhesives Europe Ltd. All chemicals were used as received without any further purification.

**Nanopatterned Single-Crystalline Thin Film Preparation:** The film was prepared using the authors' previous method.\[28\] In detail, the perovskite solution was made by dissolving a 1:1 molar ratio of PbBr\(_2\) and MABr in DMF solvent mixture in a N\(_2\) glovebox. The resulting concentration was 0.7 g mL\(^{-1}\). The solution was stirred for 12 h and filtered using PTFE filter with 0.2 µm pore size. The solution was then filled into the gap between two substrates and heated at 80 °C for 2–4 h, then the temperature was decreased to 60 °C slowly, the upper substrate was changed into a patterned PDMS master, and the capillary action process repeated; the solution was preheated at 60 °C. After another 2–4 h, the patterned single-crystalline thin films with several millimeters in width were fabricated.
Table 1. Comparison of the nanopatterned photodetector polarization sensitivity with some other materials.

| Materials                              | Sensitivity | Responsivity [A W⁻¹] | Detectivity [Jones] | Response time | Stability          | Ref.          |
|----------------------------------------|-------------|-----------------------|---------------------|---------------|--------------------|---------------|
| MAPbBr₃ patterned single crystal        | ≈2.2        | 8 × 10⁻¹ @ 1.11 V     | 1.08 × 10¹⁰         | ≈0.1 s        | 7 days in air >85% | This work     |
| MAPbBr₃ single crystal                 | ≈3          | 1.5 × 10⁻¹ @ 1 V      | 10¹⁰                | –             | –                  |              |
| CsPbBr₃ polycrystalline film           | 0.3 @ 0 V   | 1.15 × 10¹³           | 0.2 s               | 1 week        | ≈60%               | [30]          |
| MAPbI₃ polycrystalline film            | 0.1 @ 10 V  | 1.02 × 10¹²           | 0.3 ms              | 1 week        | <20%               | [31]          |
| MAPbI₃ nanowire                        | 1.3         | 4.95 @ 1 V            | 2 × 10¹³            | <0.1 ms       | One month 28%      | [21a]         |
| (BA)₂(MA)PbBr₃ single crystal          | 2           | –                     | 1.1 × 10⁸           | ≈20 µs        | –                  | [3b]          |
| (FPEA)₂PbI₃ single crystal             | 2.1         | –                     | 10¹¹                | –             | –                  | [20d]         |
| CsPbBr₃ single-crystalline arrays      | 2.6         | 40–1377               | –                   | 21.5 µs       | –                  | [22b]         |
| Black phosphorus                       | 3.5         | 3.5 × 10⁻¹⁴           | –                   | 40 µs         | –                  | [7]           |
| GeSe                                   | 1.09        | –                     | –                   | –             | –                  | [32]          |
| ReS₂                                   | 3.5         | 10¹                  | –                   | –             | –                  | [33]          |
| Core/shell GaAs/AlGaAs nanowire        | 2           | –                     | –                   | –             | –                  | [12]          |
| Si nanowire                            | 5           | –                     | –                   | –             | –                  | [18]          |
| GeO₂–Ge nanowire                       | 1.4         | 10⁴                  | 3.8 × 10¹²          | –             | –                  | [14]          |

**Morphological and Structural Characterization:** The MAPbBr₃ single crystal film SEM images were obtained by JEOL JEM7600F, FEI Nova NanoSEM 650, and operating at an accelerating voltage of 5 kV in low vacuum mode. With the aid of EDX (attached to FEI Tecnai F20), the chemical composition of the film was determined at a voltage of 10 kV. The EBSD analysis was performed at a voltage of 10 kV. All AFM experiments were carried out in tapping mode under ambient conditions (Dimension ICON SPM system, Brooke, USA). Commercial silicon tips with a nominal spring constant of 40 N m⁻¹ and a resonance frequency of 300 kHz were used in all experiments. The Bruker D8 Advance diffractometer with Bragg–Brentano geometry collected X-ray powder diffraction data and operated with Ni filtered CuKα radiation (λ = 1.5418 Å) in the 2θ range from 5° to 70°.

**Device Fabrication and Photodetector Measurements:** For photodetector fabrication, the SiO₂/Si substrate was cleaned with water, ethanol and acetone, then with O₂ plasma. Non-patterned and nanopatterned MAPbBr₃ single crystal thin films were grown onto the SiO₂/Si substrates. Then, 200 nm Au was deposited using thermal evaporation with a shadow mask. The channel width and length were controlled as 1 mm and 60 µm, respectively. To investigate the planar and grating photodetectors’ linearly polarized optoelectronic properties, the photocurrents were collected using Keithley 2400 in the air at room temperature. The influence of crystal surface gratings on photodetectors’ optoelectronic properties were also compared under the same conditions. By adjusting the angle of the collimated polarizer, linearly polarized light with various polarization angles could be obtained.

**Computation Details:** The simulation was performed using the total energy code CASTEP. In the calculation, the Perdew–Wang generalized gradient approximation (GGA) exchange–correlation functional was adopted; the electron wave function of each k point was expanded into a plane wave basis set, which was determined by the energy cut-off point of 600 eV. The Monkhorst–Pack grid parameters for k-point sampling are 5 × 5 × 5. After structural optimization, the optical properties of MAPbBr₃ under different polarization directions were calculated, and the imaginary part of the dielectric function was obtained. According to the electronic properties of the MAPbBr₃ (100) plane, the band structure and charge density map of the crystal were analyzed.

The RCWA computation was performed using RSoft DiffractMOD. For simplification, the perovskite was considered dispersion free. In the authors’ work, the single crystal thin film thickness of ≈5 µm was applied. The absorbance relative to reflectance was calculated because transmittance was negligible for the samples. The absorption (A) spectrum was obtained indirectly from the reflectivity (R): \( A = 1 – R \). A similar approach to the reported Si subwavelength grating absorber on a thick Si substrate.

**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 supports the findings of this study are available in the supplementary material of this article.
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