The electrical and photo-electrical properties of exfoliated MoS\(_2\) were investigated in the dark and in the presence of deep ultraviolet (DUV) light under various environmental conditions (vacuum, N\(_2\) gas, air, and O\(_2\) gas). We examined the effects of environmental gases on MoS\(_2\) flakes in the dark and after DUV illumination through Raman spectroscopy and found that DUV light induced red and blue shifts of peaks (E\(_{2g}^1\) and A\(_{1g}\)) position in the presence of N\(_2\) and O\(_2\) gases, respectively. In the dark, the threshold voltage in the transfer characteristics of few-layer (FL) MoS\(_2\) field-effect transistors (FETs) remained almost the same in vacuum and N\(_2\) gas but shifted toward positive gate voltages in air or O\(_2\) gas because of the adsorption of oxygen atoms/molecules on the MoS\(_2\) surface. We analyzed light detection parameters such as responsivity, detectivity, external quantum efficiency, linear dynamic range, and relaxation time to characterize the photoresponse behavior of FL-MoS\(_2\) FETs under various environmental conditions. All parameters were improved in their performances in N\(_2\) gas, but deteriorated in O\(_2\) gas environment. The photocurrent decayed with a large time constant in N\(_2\) gas, but decayed with a small time constant in O\(_2\) gas. We also investigated the characteristics of the devices after passivating by Al\(_2\)O\(_3\) film on the MoS\(_2\) surface. The devices became almost hysteresis-free in the transfer characteristics and stable with improved mobility. Given its outstanding performance under DUV light, the passivated device may be potentially used for applications in MoS\(_2\)-based integrated optoelectronic circuits, light sensing devices, and solar cells.
application because of their high surface-to-volume ratio. MoS₂ sheets are sensitive detectors of NO, NO₂, NH₃, N₂, and triethylamine gases.[11–15] However, the large variation in the electrical transport properties of MoS₂ is generally attributed to extrinsic or environmental factors. These effects may significantly limit the exploration of the intrinsic properties of MoS₂. Investigating the environmental factors that influence the reliability and stability of MoS₂ FETs is important. Detailed and comparative investigations on environmental gases effects are needed to understand the overall performance of MoS₂ FETs and undesirable effects should be minimized or removed to enhance device reliability and stability.

In this study, we fabricated few-layer (FL) MoS₂ FETs and investigated electrical transport properties under various environmental conditions. We analyzed the change in on-state current under different environmental conditions. We observed the hysteresis behavior in transfer characteristics, where oxygen on the MoS₂ surface plays a vital role in hysteresis. Gas environment significantly affects the characteristics of MoS₂ FETs under deep ultraviolet (DUV) light. Thus, we investigated the time-dependent photosresponse of MoS₂ FETs in the presence of various environmental gases. We also studied the maximum photocurrent saturation, responsivity, detectivity, external quantum efficiency (EQE), linear dynamic range (LDR), and relaxation time after switching light ON and OFF under various environmental conditions. Finally, we investigated the characteristics of devices after passivating by Al₂O₃ film on the MoS₂ surface. The devices became almost hysteresis-free and stable FETs with improved mobility. The effect of Al₂O₃ on MoS₂ flakes was discussed to explore the basic and important parameters regarding light detection.

2. Experimental section

We fabricated FL-MoS₂ FETs using a conventional approach. FL-MoS₂ flakes were transferred to clean 300-nm-thick SiO₂ layer on p++ Si substrate with Scotch tape via mechanical cleavage method.[16] FL flakes were initially identified under an optical microscope (Figure 1(a)) and further confirmed through Raman spectroscopy. The thickness of the flakes was estimated as ~3.6 nm by atomic force microscopy, which indicated the presence of five layers (Supporting Information Figure S1). Large patterns formed through photo-lithography for each device, and fine electrodes for source and drain were completed after electron beam-lithography. The source–drain contacts of Cr/Au (10/80 nm) were deposited via thermal evaporation. Electrical transport measurements were performed using a Keithley 2400 source meter (Beaverton, OR, USA) and a Keithley 6485 picoammeter (Beaverton, OR, USA). The final optical device image is illustrated in Figure 1(b). All of the measurements were carried out at room temperature. Raman spectra were recorded with a Renishaw microspectrometer equipped with a 514 nm laser at room temperature. DUV light (λ = 220 nm and average intensity of 11 mW cm⁻²) was used for illumination.

A 10-nm-thick Al₂O₃ passivation layer was deposited on the FL-MoS₂ at 200°C through atomic layer deposition (ALD; Lucida D100 ALD by NCD Co., Ltd, Daejeon, Korea). During the growth process, 100 cycles with a growth rate of ~1 Å per cycle were completed to reach the desired thickness. Trimethylaluminum and deionized water were used as precursors. Ultra-pure N₂ (99.999%) was used as a carrier and purging gas. The pressure of the growth chamber was maintained at ~5.5 × 10⁻³ torr during deposition.

3. Results and discussion

Raman spectroscopy is a fast and reliable tool that provides information about structural and local perturbation of 2D materials. The Raman spectrum of MoS₂ is dominated by two vibrational modes: E₁₂g, which is attributed to in-plane vibrations of two S atoms with respect to the Mo atom, and A₁g, which corresponds to the out-of-plane vibrations of S atoms in opposite directions. The E₁₂g and A₁g modes are sensitive to the number of layers forming the structure of MoS₂.[17, 18] The Raman spectra of FL-MoS₂ layers were recorded at room temperature with a laser excitation of ~514 nm and a small laser power of ~1.0 mW to avoid the heating effect. Figure 1(c) shows the Raman spectra of pristine and gas-treated (in the dark and under DUV light) FL-MoS₂ (five layers; see Figure S1). The FL-MoS₂ exhibited strong bands at ~383.2 and ~407.7 cm⁻¹, which is ascribed to in-plane vibrational (E₁₂g) and out-of-plane vibrational (A₁g) modes, respectively. The vibrational modes of FL-MoS₂ did not significantly change after treating the samples with O₂ and N₂ gases in the dark. However, we observed a shift toward a lower wave number after DUV treatment of FL-MoS₂ in the presence of N₂ gas. Subsequently we observed a shift toward a higher wave number in the presence of O₂ gas under DUV light (Figure 1(c)). After the DUV treatment in N₂ gas, the red shift of A₁g and E₁g was ~2.2 and ~1.6 cm⁻¹, respectively. The subsequent DUV treatment in O₂ gas environment made the blue shift of A₁g and E₁g by ~2.1 and ~1.5 cm⁻¹, respectively. The red and blue shifts in peak (E₁₂g and A₁g) position are due to doping of electrons and holes on FL-MoS₂ by DUV treatment in N₂ and O₂ gas, respectively. The mechanism of Raman shifts by adsorption and desorption of these gas molecules is attributed to electron-phonon coupling of modes. The A₁g mode couples more strongly with electrons than the E₁g mode. The electron doping in MoS₂ causes softening specifically of its Raman-active A₁g phonon, accompanied by increase in line-width of its Raman peaks.[19] In contrast, the other Raman mode with E₁₂g symmetry is less sensitive to electron doping. The Raman spectrum
for each case was obtained from five different points on the MoS\(_2\) surface, and the results were consistent.

The electrical transport measurements of the FL-MoS\(_2\) FETs (sample-1; \(L = 8.1\) μm and \(W = 7.6\) μm) were carried out under various environmental conditions. Transfer characteristics (drain–current \(I_D\) as a function of back-gate voltage \(V_g\)) were measured at room temperature with fixed source–drain voltage (\(V_{ds}\) = 1 V).

Figure 2(a) presents the transfer characteristics of the FL-MoS\(_2\) FETs in the dark under various environmental conditions. The measurements were performed in vacuum, followed by \(N_2\), air, and \(O_2\) gas environment. The current level of the device was almost similar in the \(N_2\) and vacuum environments but was substantially reduced in the air and \(O_2\) environments. The \(I_{on}/I_{off}\) of our device was \(\sim 10^5\) and remained almost same in vacuum, \(N_2\), air, and \(O_2\) gas flow (Supporting Information Figure S2a).

Figure 2(b) illustrates the hysteresis in the transfer characteristics of FL-MoS\(_2\) in the dark under different environmental conditions. We swept the \(V_g\) from –40 to 80 V repeatedly and observed a hysteresis in vacuum. Later on, the same devices were exposed in \(N_2\) gas and found a similar hysteresis trend to that in vacuum. However, further exposure of devices to air and oxygen significantly changed the hysteresis because of adsorption of oxygen atoms/molecules on top of MoS\(_2\) surface.[20] This kind of oxygen adsorption resulted in formation of charge-trapping sites and made the hysteresis prominent in FL-MoS\(_2\) FETs. In transfer characteristics in Figure S2a, we found that the threshold voltages \((V_{th})\) of FL-MoS\(_2\) FETs was near –35, –37, –34, and –28 V in vacuum, \(N_2\), air, and \(O_2\), respectively. Furthermore, we observed that the \(V_{th}\) remained similar in vacuum and \(N_2\) gas but shifted toward positive gate voltages when the devices were exposed to air and \(O_2\) gas. The shift of \(V_{th}\) to positive back gate voltages indicates an electron deficiency in FL-MoS\(_2\) due to exposure to air and oxygen. Such behavior was previously reported for MoS\(_2\)-related FET devices in \(O_2\) environments, which was also attributed to the absorption of oxygen molecules into sulfur or defect states on the MoS\(_2\) surface, which traps the charge carriers.[21, 22]

Figure 2(c) shows the transfer characteristics of the FL-MoS\(_2\) FETs under various environmental conditions in the presence of DUV light. DUV light substantially
improved the drain current of the FL-MoS2 FETs in N2 and in vacuum, but significantly decreased it in air and in O2 gas environments. DUV light reduced the oxygen atom/molecules from the MoS2 nanosheet surface in N2 and vacuum environments. The adsorption of oxygen atom/molecules on the MoS2 nanosheet reduced the number of charge carriers (electrons), causing a decline of drain current, which is consistent with previous reports.[23, 24] Figure 2(d) shows the field-effect mobility (μ) of the FL-MoS2 device under various environmental conditions in the dark and in the presence of DUV light. The mobility (μ) of the FL-MoS2 device was obtained using the following relation:

\[ \mu = \frac{L}{WC_g} \frac{dI_{ds}}{dV_g} \]  

where \( dI_{ds}/dV_g \) is slope of the transfer curve in the linear region, \( C_g \) is the gate capacitance (≈115 aF μm\(^{-2}\)) of Si/ SiO\(_2\) substrates (300 nm), \( V_{ds} \) is the source–drain current for each device, \( L \) is the channel length, and \( W \) is the channel width. In the dark, we estimated the \( \mu_{\text{vacuum}} \), \( \mu_{\text{N2}} \), \( \mu_{\text{air}} \), and \( \mu_{\text{O2}} \) of the FL-MoS2 FETs to be ~15.6, 16.5, 9.1, and 8.1 cm\(^2\) V\(^{-1}\)s\(^{-1}\), respectively. However, under DUV illumination, the \( \mu_{\text{vacuum}} \), \( \mu_{\text{N2}} \), \( \mu_{\text{air}} \), and \( \mu_{\text{O2}} \) of the FL-MoS2 FETs were ~24.8, 50.6, 11.9, and 0.8 cm\(^2\) V\(^{-1}\)s\(^{-1}\), respectively. The FL-MoS2 FETs exhibited a high \( I_D \) and large mobility in N2 but showed a small \( I_D \) in air and O2 environment under DUV illumination.[25] Figure 3 presents the output characteristics of the FL-MoS2 FETs in vacuum, N2, air, and O2 environments in the dark. The output characteristics (\( I_D \) versus \( V_{ds} \)) were measured at a fixed \( V_g \) ranging from −20 V to +60 V with a step of 20 V. The nonlinear I-V characteristics supports that the Cr/Au metal electrodes make Schottky barrier due to the work function difference between metal and MoS2.

To measure the photodetector response, the MoS2 nanosheet device was irradiated with DUV light (sample-2; \( L = 1.3 \mu m \) and \( W = 3.15 \mu m \)). In Figure 4(a) we show the transient current response to a cycle of
Two key figures of merit were measured to quantify detector performance. Current responsivity \( (R_{\lambda}) \) and detectivity \( (D^*) \) were defined as follows [26]:

\[
R_{\lambda} = \frac{I_{\text{ph}}}{P A}
\]

\[
D^* = \frac{R_{\lambda} A^2}{\sqrt{2 e I_{\text{dark}}}}
\]

where \( I_{\text{ph}} \) is the photo-excited current, \( P \) is the light power intensity, and \( A \) is the effective area of the photodetector. Responsivity is defined as the photocurrent generated per unit power intensity of the incident light on the effective area of a photoconductor. The power intensity of our DUV incident light and the area of device were 11 mW cm\(^{-2}\) and 4.09 \( \mu \)m\(^2\), respectively. In Equation (3), \( e \) is the absolute value of the electron charge and \( I_{\text{dark}} \) is the current density in dark. Figure 4(d) shows the responsivity and detectivity of our devices, where \( R_{\lambda} \) was estimated to be ~65.9, 27.9, 14.7, and 154
An important factor which is the denominator in the formula (EQE = hνR_{λ}/e_{λ}). We enhanced our EQE to 8600% using smaller wavelength together with N2 gas environments. The N2 gas provides a supportive environment for photocurrent generation. The LRD (= 20 log (I_{ph}/I_{dark}), where I_{ph} is the photocurrent measured at a light intensity of 11 mW cm^{-2}) was measured under DUV illumination. The measured LDR values of the MoS2 nanosheet device were ~29.5, 23.16, 20, and 34.4 dB in vacuum, air, O2, and N2 environments, respectively. The results demonstrate that MoS2 nanosheet devices in N2 gas can be potential photodetectors in the future.

Relaxation response time or decaying behavior is another key factor in photodetector performance. Decaying behavior is observed after photocurrent saturation under various environmental conditions. Relaxation data were extracted from Figure 4(b), when light was turned off after photocurrent saturation. The response time obtained from 1D nanostructures or graphene oxide-based photodetectors ranges from seconds to several tens of minutes.[33–35] The wide range of response time is attributed to the difference in the materials or device structures. The dynamic response to kAW^{-1} in vacuum, air, O2, and N2 gas environments, respectively. D', which is measured in Jones units (1 Jones = 1 cm Hz^{1/2} W^{-1}), ranged from ~10^{13}–10^{14}. These data indicated the best response in N2 gas, which is more than ~10^{4} times higher than that of the previously reported 2D layered material devices on Si/SiO2 substrates.[26–30]

To investigate photodetector performance further, two other critical parameters, namely EQE and LDR (typically presented in the unit of dB), were measured as shown in Figure 5(a) and (b), respectively. The EQE (= hνR_{λ}/e_{λ}, where h is a plank constant, c is the speed of light, R_{λ} is the responsivity at a DUV wavelength of 220 nm, and e is the electron charge) is defined as the number of electron–hole pairs excited by one absorbed photon. The EQE was estimated to be ~3700, 1500, 830, and 8600% in vacuum, air, O2, and N2 environments, respectively. Compared with previously reported devices, our MoS2 devices on the Si/SiO2 substrate exhibited higher EQE in N2 gas.[24,28,31,32] There are three factors which improve external quantum efficiency (EQE) of our devices; higher responsivity, light of lower wavelength and N2 gas environment. The wavelength, λ, is an important factor which is the denominator in the formula (EQE = hνR_{λ}/e_{λ}). We enhanced our EQE to 8600% using smaller wavelength together with N2 gas environments. The N2 gas provides supportive environment for photocurrent generation. The LRD (= 20 log (I_{ph}/I_{dark}), where I_{ph} is the photocurrent measured at a light intensity of 11 mW cm^{-2}) was measured under DUV illumination. The measured LDR values of the MoS2 nanosheet device were ~29.5, 23.16, 20, and 34.4 dB in vacuum, air, O2, and N2 environments, respectively. The results demonstrate that MoS2 nanosheet devices in N2 gas can be potential photodetectors in the future.

Figure 4. Photocurrent response in various environments. (a) Time-dependent photocurrent response of the device in different environments at V_{ds} = 1 V and V_{g} = 0 V. DUV light was first turned on for 60 s and then turned off for 60 s. (b) Photocurrent saturation of the FL-MoS2 FET in vacuum and N2 gas (left axis and bottom axis), and in air and O2 gas (right axis and top axis) at (V_{ds} = 1 V and V_{g} = 0 V). (c) Saturated photocurrent values in vacuum, N2 gas, air, and O2 gas. (d) Modulation of photocurrent response parameters; maximum responsivity (left axis) and detectivity (right axis) in different environments.
Figure 5. Light detection response. Crucial factors to be calculated in vacuum, \(N_2\), air, and \(O_2\) for (a) external quantum efficiency and (b) linear dynamic range. The data are obtained from Figure 4(b), (c), and (d).

Figure 6. Photocurrent decaying behavior in different environments. Transient property of FL-MoS\(_2\) FETs under different conditions and data fitting (red line) with a single exponential equation. (a) Decay of drain current in vacuum. (b) Decay of drain current in \(N_2\). The device exhibits a slow decay with a large time constant. (c) Decay of drain current in air and \(O_2\). The drain current decays with a small time constant. (d) Relaxation time constants of the device in vacuum, \(N_2\), air, and \(O_2\).
films.[37,38] In the present study, electron-capturing impurity states were reduced under DUV light in N$_2$ gas environment but were enhanced under DUV light in O$_2$ gas environment. Thus, the decay time was significantly longer in N$_2$ environment and substantially shorter in O$_2$ environment. Evidently, decaying behavior was robustly dependent on the surrounding environment.

We measured the electrical transport of the FL-MoS$_2$ FETs (sample-3; L = 1.5 μm and W = 1.2 μm) in vacuum at a bias voltage $V_{ds}$ = 1 V to study the passivation effect by high-k dielectric materials to circumvent the environmental effects. The transfer characteristic of the pristine device at room temperature in the dark is shown in Figure 7(a), where a pronounced hysteresis can be observed. A 10-nm-thick passivation layer of Al$_2$O$_3$ was deposited on the same device through atomic layer deposition (ALD). After depositing the passivation layer, the transfer characteristics of the device were examined in the dark under various environmental conditions (Figure 7(b)). The MoS$_2$ device was almost hysteresis-free with increased drain current and exhibited n-type. The Al$_2$O$_3$ layer on the MoS$_2$ surface served as a DUV illumination for relaxation time can be expressed as [28, 36]:

$$I_{ph}(t) = I_{dark} + A \exp \left( -\frac{t}{\tau_{decay}} \right)$$

(4)

where $A$ is a scaling constant, $\tau_{decay}$ is a time constant for decaying, and $t$ is the time after DUV light is switched on or off. The time constant ($t$) can be calculated by fitting the experimental data. Figure 6(a–c) describes the decaying photocurrent behavior in N$_2$, vacuum, air, and O$_2$ environments, respectively, and the red lines indicate the fitting data. The relaxation time was calculated to be ~215.9, 79.4, 11.1, and 3.7 s in N$_2$, vacuum, air, and O$_2$, respectively (Figure 6(d)). These results demonstrated that the decay process was very slow in N$_2$ and significantly fast in O$_2$ gas. Thus, the adsorbates and defect states originating from ambient air, water, and oxygen atoms/molecules play an important role in photocurrent relaxation and electron-hole pair recombination dynamics. In general, the surface-adsorbed oxygen significantly affects the photoresponse of MoS$_2$ and ZnO

Figure 7. Electrical transport measurements of passivated FL-MoS$_2$ FET. (a) Transfer characteristics of the FL-MoS$_2$ FET device in the dark. Transfer characteristics are measured in vacuum. The linear scale is on the right axis, and the logarithm scale is on the left axis. (b) Hysteresis in transfer characteristics after passivation. The measurements were performed in the dark under different environmental conditions with $V_g$ from −60 V to +80 V. (c) Transfer characteristics under DUV illumination. The measurements were carried out in vacuum at $V_{ds}$ = 1 V. The power intensity and wavelength of light were 11 mW cm$^{-2}$ and 220 nm, respectively. (d) Field–effect mobility of the passivated FL-MoS$_2$ FET device in the dark and under illumination. The measurement was conducted in vacuum.
protecting or capping layer. Hence, the effect of external environments was effectively disregarded.

In addition, field-effect mobility increased after passivation by Al₂O₃ film. The improvement of device performance after deposition of a high-k dielectric is associated with the suppression of Coulomb scattering, modification of phonon dispersion, the difference of Al₂O₃ and SiO₂ dielectric constants (3.9 for SiO₂ and 9.0 for Al₂O₃), and the removal of impurities during ALD growth at 200°C.[39–41] Extensive theoretical work including the calculation of phonon dispersion relations in MoS₂, the calculation of scattering rates on phonons and charge impurities, is necessary to describe these phenomena completely. After the deposition of a 10-nm-thick Al₂O₃ layer, the electrical properties were also measured under DUV illumination in vacuum at Vds = 1 V. A huge increment in drain current was observed (Figure 7(c)). The field-effect mobility of the pristine and passivated devices in the dark and in the presence of DUV illumination was measured in vacuum, and the mobility was ~18, 32, and 104 cm² V⁻¹ s⁻¹, respectively (Figure 7(d)).

We explored the photoresponse of the FL-MoS₂ FETs (sample-4; L = 2 µm and W = 8.2 µm) passivated by the Al₂O₃ layer (10 nm). The transfer characteristics of the device are shown in Figure S2d (Supporting Information). The time-dependent response under DUV light is described in Figure 8(a) with switching light ON and OFF for a 60 s cycle at (Vds = 1 V and Vg = 0 V) in vacuum. As shown in Figure 8(a), the passivated device exhibited a slow response because of the suppression of impurities and defect states by the Al₂O₃ layer. However, the photocurrent of the passivated device enhanced compared with that of the pristine device. In Figure 8(b), the maximum photocurrent saturation values (ΔImax = I_saturation – I_dark) of the pristine and passivated FL-MoS₂ FETs were found to be ~20.6 and 24.2 μA, respectively. Rλ and D’ were also investigated as shown in Figure 8(c). The maximum responsivity and detectivity of the pristine device were ~11 × 10³ AW⁻¹ and ~6.3 × 10¹² Jones, respectively. However, those after deposition of the Al₂O₃ layer were ~14 × 10³ AW⁻¹ and ~8.2 × 10¹² Jones, respectively. To investigate further the effect of passivation, the relaxation time (τ_decay) of the pristine and passivated devices was examined (Figure 8(d)). The data for decaying time were obtained from Figure 8(b) and exponentially fitted by the single exponential Equation (4). The carrier lifetimes for the pristine and passivated devices were ~30.3 and 284.5 s, respectively.
As depicted in Figure 8(d), the relaxation time of the passivated device was significantly longer than that of the pristine device. This result can be attributed to the fact that the intermediate states of defects and oxygen constitutes are reduced after Al₂O₃ deposition and those excited electrons take a long time to relax.

4. Conclusions

In conclusion, we fabricated FL-MoS₂ FETs and studied the effects of environmental gases in the dark and under DUV illumination. We determined that DUV light induced red and blue shifts of Raman peak (E₁₉₁₂ and A₁₉₂₀₆) positions in the presence of N₂ and O₂ gases, respectively. We investigated the electrical transport measurements of FL-MoS₂ FETs under various environmental conditions and found that device performance did not degrade in N₂ gas in the dark. However, O₂ gas significantly reduced the ON-current state. In the dark, the environmental gases did not modify the Ion/Ioff of the device and remained consistent at ~10⁻⁵. The threshold voltage and hysteresis were nearly similar in vacuum and N₂ gas but shifted toward positive gate voltage accompanied with a large hysteresis loop in air and O₂ gas. However, the environmental gases changed the characteristics of the FL-MoS₂ FETs under DUV illumination. The photocurrent response and saturated photocurrent of the FL-MoS₂ FETs under various environmental conditions were examined to investigate light detection parameters, such as responsivity, detectivity, external quantum efficiency, and linear dynamic range. The results demonstrated a strong photoresponse in N₂ gas but a poor photosresponse in O₂ gas. The photocurrent relaxation after turning off DUV light was also examined under various environmental conditions. The lifetime of a carrier was longer in N₂ gas but shorter in O₂ gas, indicating that N₂ gas helped the device recover from defects and impurities, while O₂ gas adversely affected device characteristics. Furthermore, we passivated the device by depositing the Al₂O₃ layer to protect MoS₂ from environmental effects. We observed almost hysteresis-free transfer characteristics with improved electrical and photoelectrical responses and field-effect mobility after deposition of the passivation layer. We also found that r decay increased after Al₂O₃ layer deposition. This phenomenon may be attributed to the screening of impurities and defects from the MoS₂ surface. Thus, we developed improved, stable, reliable, and hysteresis-free FL-MoS₂ FETs for various nanotechnology applications.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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