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Photonic Crystallization of Two-dimensional MoS$_2$ for Stretchable Photodetectors

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Abstract

Low temperature synthesis of high quality two-dimensional (2D) materials directly on flexible substrates remains a fundamental limitation towards scalable realization of robust flexible electronics possessing the unique physical properties of atomically thin structures. Herein, we describe room temperature sputtering of uniform, stoichiometric amorphous MoS$_2$ and subsequent large area (>6.25 cm$^2$) photonic crystallization of 5 nm 2H-MoS$_2$ films in air to enable direct, scalable fabrication of ultrathin 2D photodetectors on stretchable polydimethylsiloxane (PDMS) substrates. The lateral photodetector devices demonstrate an average responsivity of 2.52 $\mu$W/A and a minimum response time of 120 ms under 515.6 nm
illumination. Additionally, the surface wrinkled, or buckled, PDMS substrate with conformal MoS$_2$ retained the photoconductive behavior at tensile strains as high as 5.72% and over 1000 stretching cycles. The results indicate that the photonic crystallization method provides a significant advancement in incorporating high quality semiconducting 2D materials applied directly on polymer substrates for wearable and flexible electronic systems.

**Introduction**

Flexible and stretchable photodetector devices based on two-dimensional (2D) materials are known to exhibit a rare combination of high optoelectronic performance with the ability to accommodate large amounts of strain. This unique coupling is enabled by the broad optical absorption in graphene and other 2D material systems, quantum confinement of energy carriers in the 2D plane resulting in ultrafast transport dynamics, the van der Waals bonding between the layers, and the enhanced electromechanical properties that arise due to the extreme thinness of the material. Resultant 2D material photodetector devices exhibit rapid photoresponse, semitransparency, and even strainability as high as 200% in crumpled systems. Practical realization of flexible 2D materials in these applications is primarily limited by the lack of large area, transfer-free processing schemes that enable incorporation of the layered materials onto soft, organic substrates and allow for bottom-up device fabrication. Large scale photonic crystallization of amorphous precursors to layered 2D materials directly on soft substrates, as demonstrated in this study, can facilitate the future 2D flexible electronic and optoelectronics that are easily processed at low temperatures for devices including ultra-thin photodetectors.

Synthesis of 2D materials with high crystal quality typically require high temperatures in the range of 500-1000°C coupled with an epitaxial template to facilitate the thermodynamic
reactions stoichiometric materials with low defect densities. To overcome the processing limitations for flexible electronics where the substrate typically cannot accommodate for either of these conditions, the 2D layers must either be grown as polycrystalline films at a much lower substrate temperature, transferred from a rigid substrate to the flexible substrate of interest, or processed by non-thermal annealing techniques in order to achieve similar device performance. Recently, techniques including additive manufacturing have emerged that allow for the transfer of liquid-based 2D inks onto flexible substrates for device manufacturing. These 3D printing approaches enable patternability and low temperature solution processing, however issues with repeatability and film discontinuity require advances before practical devices can be achieved.

Herein, we present a technique that utilizes a large area pulsed light source in order to initiate controllable transformation of amorphous ultrathin MoS$_2$ films to layered 2H-MoS$_2$ on flexible polydimethylsiloxane (PDMS). The phase transformation in this case is performed in open air environment and occurs through absorption of the incident light resulting in selective, local heating of the film to temperatures predicted as high as 390°C without decomposition of the PDMS substrate. Photonic crystallization has been utilized to process materials including graphene inks, metallic nanoparticles, and other nanomaterials. This work highlights photonic crystallization as a process to induce phase transformations in semiconductor 2D materials directly on soft stretchable substrates for flexible photo-devices which can operate under repeated stretching cycles.
Results and Discussion

The growth and photonic crystallization process that enabled the 2D MoS$_2$ to be directly synthesized on a flexible polymer substrate are depicted schematically in Figure 1a. Initially, the flexible and stretchable PDMS substrates (2.5 cm x 2.5 cm and 0.25 cm thickness) were loaded into an ultra-high vacuum plasma reactor with a residual base pressure of 10$^{-9}$ Torr. Room temperature magnetron sputtering from a polycrystalline MoS$_2$ target yielded continuous large area coverage of the entire PDMS wafer with stoichiometric, amorphous MoS$_2$ films (X-Ray Photoelectron Spectroscopy data in Supporting Figure S2). These films served as precursors for subsequent large area crystallization which was performed with a broadband pulsed flash lamp (Pulse Forge 1300 system) to a resulting crystalline film thickness of 5 nm. This process uses high power (1000-2000 W/cm$^2$), short duration (100-300 µs) pulses of broadband light (200-1500 nm) to illuminate a large area on the order of 1 m$^2$ for thermally processing of thin films. With this specifically tuned photonic curing process, the amorphous MoS$_2$ films can be thermally crystallized to the 2H semiconducting phase of MoS$_2$ on the surface of a PDMS substrate without evidence of substrate decomposition. Previous studies of laser annealing indicate that the transformation is mostly thermal, as the crystallization is dependent upon irradiance (i.e. power and time) rather than strictly fluence.$^{27}$ Crystallization over the entire surface of the 6.25 cm$^2$ samples was uniform as evidence by spatially-resolved micro-Raman spectroscopy, with only slight changes in morphology at the extreme edges (less than 50 µm) likely due to enhanced cooling at the sample edges (Supporting Figure S3).
Figure 1. Photonic crystallization and characterization of MoS$_2$ films. a) Schematic diagram depicting the fabrication of the crystalline MoS$_2$ films on the PDMS substrate with photonic curing process, b) Raman scans of PDMS substrate, amorphous MoS$_2$ and crystalline MoS$_2$ with the inset spectra depicting polarized scans in the parallel (VV) and cross-polarized (VH) direction c) Optical image of samples; bare PDMS, amorphous MoS$_2$ films on the PDMS, and crystalline MoS$_2$ films on the PDMS (from left), and d) Comparison of the absorption behavior between amorphous MoS$_2$ films and crystalline MoS$_2$ films on the PDMS substrate. Logo courtesy of AFRL.

The Raman spectra (514.5 nm excitation) of PDMS, amorphous MoS$_2$, and crystalline MoS$_2$ in Figure 1b reveal the presence of the peak associated with in-plane stretching of 2H MoS$_2$ mode at 383 cm$^{-1}$ ($E_{12g}^1$) and the peak for out-of-plane stretching around 406 cm$^{-1}$ ($A_{1g}$), which are only visible after the photonic crystallization. Polarized Raman spectra in the cross-
polarized configuration (see Experimental Methods for more detailed explanation) provide clues to the orientation of the MoS$_2$ layers. In a backscattering configuration, the out-of-plane A$_{1g}$ mode is expected to disappear for the cross-polarized configuration, which can observed for transition metal dichalcogenide (TMD) layers parallel to the substrate$^{28}$ but not when they are aligned at other angles.$^{29}$ The polarized Raman spectra from the photonically crystallized films shows a significant decrease in intensity of the A$_{1g}$ mode relative to the E$_{12g}^1$ mode, therefore strongly suggesting the presence of MoS$_2$ layers crystallized parallel to the substrate.

As shown in Figure 1c both the amorphous and crystalline MoS$_2$ on the PDMS substrate exhibit similar semitransparency to visible light. Absorbance spectra reveal that the initial amorphous film is featureless with no observable exciton peaks characteristic of crystalline 2D MoS$_2$. The spectra of photonically crystallized MoS$_2$ resolves electronic transitions at 602 nm and 398 nm, consistent with B and C excitonic transitions.$^{30}$ The broadness of the direct excitonic features may be due to defect states present in the polycrystalline MoS$_2$. As these excitonic transitions are highly localized,$^{31, 32}$ the strength of these electronic transitions can be attenuated due to defect trap-state recombination sites.

The magnetron sputtering and photonic crystallization process results in a uniform nanocrystalline MoS$_2$ coating over the polymer substrate. Cross-sectional TEM imaging from reveals a continuous layered structure with a film thickness of 5 nm (Figure S1). The morphology of the film and substrate is dominated by buckles on the surface of the PDMS upon exposure to both the film deposition processes and the photonic crystallization.$^{33}$ Two-dimensional graphene and TMD materials have been known to “crumple” or “wrinkle” under an applied strain load, which has been shown to improve both the mechanical properties and photodetector properties.$^{13, 14}$ The evolution of the buckled topology of the MoS$_2$/PDMS system
is shown in Figure 2a. Initially prior to any MoS$_2$ processing or deposition, the PDMS is nearly atomically smooth with an RMS roughness of 0.04 nm. The coupled processes of exposure to vacuum and energetic plasma species during amorphous MoS$_2$ deposition increases the RMS roughness to 4.32 nm and periodic surface wrinkles of around 200 nm in diameter are prevalent over the entire surface. This surface wrinkling and buckling has been known to occur during ultra-high vacuum deposition by a combination of exposure to vacuum condition as well as plasma bombardment during the growth process, allowing for bulging at the MoS$_2$/PDMS interface.\textsuperscript{33, 34} Upon crystallization, the local heating within the MoS$_2$ and the coefficient of thermal expansion mismatch between the two interfaces results in surface wrinkling with larger diameters approaches 1 µm as well as an increase in the RMS roughness to 9.16 nm. It is notable that previous studies observed a change in the morphology of MoS$_2$ after laser annealing to aligned crystallites, where the photonic curing in this case resulted in random and isotropic morphology.\textsuperscript{27} The use of photonic crystallization rather than laser-based approaches allows for large area industrially scalable processing with relatively low equipment cost relative to a larger laser setup. Additionally, the uniform thermal heating across the wafer provides for uniform buckling across the wafer except at the extreme edges (shown in Supporting Figure S3). The buckled nature of PDMS and uniform coating of MoS$_2$ from the deposition process are presumed to be key enablers to stretchability in the photodetector demonstrations described later.\textsuperscript{35} This controlled buckling phenomena in PDMS results from several mechanisms including vacuum exposure, plasma interaction, and thermal mismatch, and can potentially be controlled and tailored for future stretchability enhancement.\textsuperscript{36, 37}
Figure 2. MoS$_2$/PDMS surface morphology. a) AFM surface scans of bare PDMS, amorphous MoS$_2$ on PDMS, and crystalline MoS$_2$ on PDMS with scale bars of 5 µm, and b) RMS roughness of the films at the same conditions.

The threshold for photonic crystallization in the amorphous films was identified by systematic investigation of the applied photonic energy fluence and broadband light pulse duration. Raman intensity of the A$_{1g}$ peak post-annealing at different pulse energies and times is shown in Figure 3a. For the case of 5 nm MoS$_2$ on PDMS, the optimal range for crystallization is within a photonic pulse duration of 125 µs to 300 µs and fluence of 0.85 to 1.20 J/cm$^2$ and with a step size between sample conditions of ~0.1 J/cm$^2$ and 50 microseconds. The condition in which the A$_{1g}$ peak was most intense for the photonically crystallized MoS$_2$ films occurred under an illumination energy of 1.14 J/cm$^2$ and 250 µs pulse time. At these conditions, the linewidth of the A$_{1g}$ peak was 10.1 cm$^{-1}$, indicating that the grain size is in the tens of nanometers.$^{38}$ Above a particular threshold of incident energy, and the heat generated from incident light source in the MoS$_2$ layer coupled with the large coefficient of thermal expansion mismatch caused film cracking and delamination of the MoS$_2$ film from the PDMS substrate (Supporting Figure S5). After this point, the films were not electrically continuous and could not be evaluated in large area optical device tests. Raman studies indicate that the film within
these cracked areas are in fact crystalline and controlling the crack formation/delamination process is critical for high quality films. The MoS$_2$ thickness of 5 nm was utilized in this study due to the strong absorption at that material thickness, and future experiments on thinner films (especially approaching monolayer) will require larger lamp sources with higher power outputs to achieve similar local temperatures.

Figure 3. Optimization of the photonic crystallization process. a) Raman intensity of the $A_{1g}$ peak post-annealing at different pulse energies and times, b) The estimated transient temperature of the MoS$_2$ during crystallization for selected illumination energy densities and inset depicts the temporal thermal profile of the MoS$_2$ at the optimal annealing conditions (1.14 J/cm$^2$ and 250 µs pulse length).
The peak temperature in both the amorphous MoS$_2$ film and the top layer of PDMS during the rapid annealing process were estimated using SimPulse software thermal modeling (NovaCentrix) for a 5 nm MoS$_2$ on 1 mm PDMS substrate. The modeling software considers first principles approaches coupled with the magnitude and time characteristics of the photonic pulse of light, the material absorption properties at the broadband wavelengths (from Figure 1d), and relevant material properties including thermal conductivity and specific heat. Figure 3b depicts the estimated transient temperature of the MoS$_2$ during crystallization for selected illumination energy fluences. The process conditions described above for the highest degree of crystallization resulted in a peak estimated temperature of 390°C (denoted by the star in Figure 3b). This is a critical observation in that the onset of decomposition of PDMS occurs around 250 °C,\(^{39}\) which highlights the benefits of selective and transient heating from the photonic curing technique as we do not observe decomposition within the polymer at the appropriate conditions. A modeled temporal thermal profile of the MoS$_2$ is shown in the inset in Figure 3b depicts the film cooling from as peak temperature of 390°C to room temperature within 25 ms. In previous works, growth and crystallization of MoS$_2$ occurred at temperatures above 350°C on SiO$_2$ substrates, in the same range as the temperature predicted by the Simpulse model software.\(^{38, 40}\) Additionally, The MoS$_2$ films reveal partial oxidation observed in the XPS analysis after illumination (Supporting Figure S2), indicating that the temperature of the film during photonically induced crystallization exceeded 330 °C, which was the critical temperature reported for MoS$_2$ decomposition in air.\(^{41, 42}\) From these observations, future photonic crystallization experiments conducted in inert ambient environments (i.e, argon gas) are necessary to entirely reduce or eliminate oxidation. The low thermal conductivity of the amorphous film material and the polymer substrate assist in isolating the heat in the film, as do the significant interfacial elastic
modulus mismatch resulting in low interfacial thermal conductance. The film/substrate interface
does not reach these high temperatures due to transient heating and poor interfacial thermal
transport, indicating that the photonic annealing is considerably more advantageous than
conventional heating.

To evaluate the feasibility of the photonically crystallized MoS$_2$ for flexible devices,
lateral photodetector devices were fabricated with Ti/Au contacts forming a 10 µm channel
length, as shown in the inset of Figure 4a. Under illumination of 515.6 nm wavelength laser
(spread size 100 µm estimated by the razor blade method) over the entire device channel, laser
power dependent photocurrent was observed with an applied external bias voltage of 1V as
shown in Figure 4b. The increase in photocurrent is a direct result of photo-induced carriers
within the active device region, indicating semiconductor device quality material over many
micrometers. Our device results in Figure 4b depict a relatively constant photoresponsivity
versus laser intensity, implying that photovoltaic mode is dominant in photocurrent generation.$^{43,44}$

This is determined by the relationship between photocurrent and incident laser power in the
expression $I_{ph} \propto P^\gamma$, where a $\gamma$ value of 1 indicates photodetector performance dominated by the
photovoltaic effect rather than photothermal effect (Figure S8). The conductivity of the MoS$_2$
channel in dark mode is estimated at 9.1 $\Omega^{-1}\text{cm}^{-1}$, which is a relatively high for a thin film MoS$_2$
and the conductivity increase of the MoS$_2$ channel with laser illumination of 59.84 W/cm$^2$ was
1.2 $\Omega^{-1}\text{cm}^{-1}$. $^{45}$ Overall, the photoresponsivity of our device without the inclusion of any dark
current contributions averaged 2.52 $\mu$A/W and maintained a constant value while the laser power
increased from 4.71 W/cm$^2$ to 59.8 W/cm$^2$. 

Figure 4. 2D MoS$_2$ photodetector performance. a) A schematic representation of 2D MoS$_2$ photodetector with a laser illumination. Inset: An optical microscopic image of the actual device. b) Power dependent photocurrent measured from fabricated MoS$_2$ photodetector. Inset: A photograph of the photocurrent measurement setup with the laser illumination. c) Time-resolved photocurrent measured from the MoS$_2$ photodetector. d) Rise time and fall time of the photocurrent measurement, e) Normalized current measured from the device under the range of tensile strain, from 0 to 5.72% and inset image of the stretching setup, and f) Mechanical durability of the fabricated device from cyclic testing.

The time-resolving, dynamic on/off behavior of the device is shown in Figure 4c as the incoming light is cycled on and off, with rise and fall times (within 10% of maximum and minimum current) of 240 and 214 ms, respectively, as shown in Figure 4d, at the highest laser power tested. The fastest response time was observed at the lowest laser power, with a rise and fall time of 122 and 120 ms, respectively (a full set of rise time and fall time analysis is shown in Supporting Table S1). The performance of our photodetector device with photonically crystallized MoS$_2$ films is lower in absolute responsivity in comparison to previous works of visible photodetectors made of single- or few-layer TMDs (Supporting Table 2), but the response time is comparable and the ease of processing and direct on polymer synthesis makes the approach very enticing for future device fabrication. As multi-layer MoS$_2$ is an indirect
bandgap semiconductor, the photocurrent amplitude is reduced in comparison to devices fabricated from single 2D layers and is also sensitive to the presence of defects and impurities. It is anticipated that further developments in the photonic annealing process (employing an inert atmosphere, reducing layer thickness, etc.) will improve photoresponse and device performance even further.

To demonstrate the utility of direct fabrication and creation of this buckled topology potential of our photonically crystalized 2D photodetectors for stretchable electronics, the devices were measured under the range of tensile strains from 0% to 5.72% as well as 1000 cycles of stretching-releasing tests. The MoS$_2$ photodetector on PDMS was mounted on a stretching stage which allows precise strain control using a micromanipulator, and the photocurrent was measured under different tensile strains. In Figure 4e, the measured photocurrent was normalized with the photocurrent value measured with no externally applied tensile strain. The normalized photocurrent remained constant up to 5.72% tensile strain, suggesting the material system remains stable under the range of applied tensile strain. Beyond this strain, the sample continuously detached from the stretching stage and could have sustained further stretching with a more robust mounting setup. Raman spectra collected under the same applied strain also show the expected changes in peak frequencies and linewidths (Supporting Figure S9) indicating that the MoS$_2$ is undergoing some strain during the stretching process. We attribute the performance stability in the devices under stretching to the uniform coating over the buckled PDMS surface, coupled with the inherent flexibility of the 2D semiconducting MoS$_2$.

In addition to examining the maximum tensile load, mechanical durability of the fabricated device was also demonstrated with the MoS$_2$ photodetector mounted to the stretching stage. The generated photocurrent from the device was measured in the unstrained condition
periodically over 1000 cycles of 4% tensile strain applied. The photocurrent values were
normalized with the photocurrent value measured after one cycle of stretching-releasing, and the
normalized current is displayed in Figure 4f. The resulting current level is consistent over 1000
cycles of stretching-releasing, with a minor decrease of 25% near the conclusion of the
experiment. This result implies that MoS$_2$ can be further fabricated into stretchable devices with
mechanical robustness for repeated cycling during use in applications such as wearable and
flexible electronics.

**Conclusion**

In summary, we demonstrated a large area and low temperature 2D material synthesis
platform through room temperature sputtering and photonic crystallization of 5 nm MoS$_2$ films
directly on a polymer substrate for flexible and stretchable optoelectronics. The phase
transformation of amorphous to polycrystalline 2H MoS$_2$ occurs through absorption of the
incident broadband light resulting in selective, local heating of the film to temperatures
sufficiently high to induce crystallization, but not induce decomposition of the PDMS substrate.
The crystalline MoS$_2$ on the PDMS substrate was readily then employed in a lateral, two-
terminal photodetector exhibiting a linear responsivity with respect to the laser power of 2.52
µA/W and a minimum response time of 120 ms upon illumination with 514.6 nm laser
excitation. Furthermore, the photodetectors were mechanically stretchable up to 5.72% strain
and over 1000 cycling events at a tensile strain of 4%. The results described in this study
indicate that photonic crystallization may be an effective integration strategy for large area
semiconducting 2D materials on stretchable and flexible substrates suitable for a new generation
of electronics.
Experimental Methods

PDMS substrate preparation and deposition of amorphous MoS$_2$

An elastomeric PDMS substrate was fabricated by curing a PDMS precursor (Sylgard 184, Dow Corning Corp) on a smooth silicon wafer in a dish. We used a mixture of PDMS precursor and curing agent (10 : 1 by weight) that had been degassed under vacuum. After the PDMS mixture was cured at 60 °C for 6 h using a vacuum oven, the PDMS was peeled off from the Si wafer. Then the substrates were cut up into smaller pieces (2.5 x 2.5 cm). The surface that was cast onto the flat silicon surface to form an extremely flat PDMS substrate for deposition. The PDMS samples were introduced via a vacuum load-lock and mounted on an electrically grounded fixture. Substrates were rotated at a rate of approximately 100 rpm while positioned 7 cm from the MoS$_2$ target. The synthesis chamber was pumped to a base pressure of < 10$^{-9}$ Torr and was then filled with ultra-high purity argon gas at a constant flow rate of 25 sccm to a pressure of 10 mTorr. All MoS$_2$ films were grown at room temperature via magnetron sputtering using a solid 3.3 cm diameter MoS$_2$ target of 99.95% purity. Pulsed dc power (60 W at 65 kHz) was applied to the MoS$_2$ target to initiate the sputtering process.

Photonic curing of MoS$_2$

The photonic crystallization was performed on a PulseForge 1300 photonic curing system manufactured by NovaCentrix. Processing parameters such as the fluence, pulse length, and the temporal evolution of the pulse shape were controlled to obtain the optimized annealing condition to crystallize amorphous MoS$_2$ films on PDMS substrate.

Characterization
The backscattered Raman spectra were acquired with a Renishaw in Via Raman Microscope using 514.5 nm excitation. The incident laser power was kept below 1 mW to avoid heating of the samples. Parallel- and cross-polarized Raman spectra (denoted as VV and VH, respectively, where V and H are the vertical and horizontal electric field vectors of the incident excitation laser) were also collected in the backscattering configuration from the crystallized MoS$_2$. Compositional changes in the MoS$_2$ films associated with the photonic annealing process were characterized via X-ray Photoelectron Spectroscopy (XPS). Scans were collected with a Surface Science Instruments M-Probe XPS system equipped with a monochromatic Al K$_\alpha$ x-ray source (photon energy of 1486.7 eV) operated at 50W. All peak positions were calibrated to an adventitious carbon C 1s peak value of 284.5 eV. Analysis was based on the position of the peaks, their area, and their full width half maximum with fitted to a Shirley background.

UV-Vis spectra were acquired on a CRAIC UV-Vis optical microscope with a spot size of 10 microns. Blank PDMS were taken as reference spectra and spectra were acquired using 16 ms dwell time and averaged over 16 spectra.

**Atomic force microscopy profiles**

Images were obtained in non-contact mode with an Antimony coated Si probe (Force Constant = 0.01 N/m). Scale bars are 5 micron. Surface roughness measurements were performed after performing a tilt correction to account for sample drift. MoS$_2$ film heights were measured by masking a portion of the PDMS substrate, MoS$_2$ deposited, and removal of the mask. The height difference between unmasked and deposited film were taken as film thicknesses. All images were processed using Bruker Nanoscope Analysis Software.

**SimPulse modeling of transient temperature profiles**
Simpulse modeling software was provided by the PulseForge 1300 photonic curing system manufacturing by NovaCentrix. Input to the simulation was the creation of a simulated layer structure with 5 nm MoS$_2$ and 1 mm thick PDMS substrate. The average absorption properties from MoS$_2$ (12% absorption) gathered from Figure 1d and thermal properties of amorphous MoS$_2$ were used from previous works.$^{38}$ The broadband light source spectrum can be found on the company website and further details with regards to the Simpulse software.

**Photodetector fabrication and photocurrent measurement**

Metal electrodes (Ti/Au: 25nm/50nm) were deposited on the fabricated MoS$_2$ on PDMS by e-beam evaporator (Temescal FC-2000 electron beam evaporator Ferrotec USA, CA) at 2.5x10$^{-6}$ Torr with a shadow mask. The dimension of photodetector is defined by the shadow mask’s window size.

To characterize the photoresponse of the fabricated MoS$_2$ photodetector array photocurrent was measured with a diode laser (516 nm, Thorlab, NJ), a sourcemeter (Keithley 2614B, Tektronix, OR), and microprobe station. Laser power dependent photocurrent was measured under the range of laser power, from 4.8 mW to 0.4 mW. The diameter of laser spot was approximately 100 μm measured by the razor blade method.

**Photodetector fabrication for strain independent photoresponse measurement**

Metal electrodes (Ti/Au: 25nm/50nm) were deposited on the fabricated MoS$_2$ on PDMS by e-beam evaporator (Temescal FC-2000 electron beam evaporator Ferrotec USA, CA) at 2.5x10$^{-6}$ Torr with a shadow mask, under a bending strain (~10% tensile strain). To avoid possible breakage at metal electrodes while the tensile strain was applied, the metal electrode was deposited under a tensile strain higher than the maximum strain employed for photocurrent measurements. After deposition, the bending strain was released, and the photodetector array
sample was mounted on a stretching stage. Using the stretching stage, tensile strain was applied to the sample, and photocurrent was measured in the range of tensile strains, from 0% to 5.72%. For durability test, the sample was repeatedly stretched and released for 1000 cycles, with the maximum tensile strain of 4%. Photocurrent was measured at 1, 2, 5, 10, 20, 50, 100, 200, 500, 1000th cycle to show the consistency.

Supporting information

The accompanied supporting information includes thickness measurement of deposited MoS$_2$ films via TEM, XPS characterization before and after annealing, SEM images of film morphology, photodetector performance, Raman analysis under strain, and a comparison to other published results on MoS$_2$ photodetectors.

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Photonic crystallization of 2D molybdenum disulfide (MoS\(_2\)) on PDMS is demonstrated as an effective direct fabrication tool to enable stretchable photodetectors that allow for up to 5.72 % strain and over 1000 stretching cycles.