Atomically Thin, Optically Isotropic Films with 3D Nanotopography

Myungjae Lee,^# Jong-Hoon Kang,# Fauzia Mujid, Joonki Suh, Ariana Ray, Chibeom Park, David. A. Muller, and Jiwoong Park,*

ABSTRACT: Flat optics aims for the on-chip miniaturization of optical systems for high-speed and low-power operation, with integration of thin and lightweight components. Here, we present atomically thin yet optically isotropic films realized by using three-dimensional (3D) topographic reconstruction of anisotropic two-dimensional (2D) films to balance the out-of-plane and in-plane optical responses on the subwavelength scale. We achieve this by conformal growth of monolayer transition metal dichalcogenide (TMD) films on nanodome-structured substrates. The resulting films show an order-of-magnitude increase in the out-of-plane susceptibility for enhanced angular performance, displaying polarization isotropy in the off-axis absorption, as well as improved photoluminescence emission profiles, compared to their flat-film counterparts. We further show that such 3D geometric programming of optical properties is applicable to different TMD materials, offering spectral generalization over for the entire visible range. Our approach presents a powerful platform for advancing the development of atomically thin flat optics with custom-designed light–matter interactions.

KEYWORDS: atomically thin materials, TMDs, conformal growth, 3D topography, optical isotropy

Compact and tunable optical components are essential building blocks in flat optics for a wide range of applications that require small optical form factors such as lensless imaging,^1–4 beam steering devices,^5–7 and optical computation with stackable diffractive plates.8–10 Recently, atomically thin materials have emerged as a next-generation platform,^11–14 due to their ultimate dimensionality,^15 architectural flexibility,^16,17 diversified library,^18–20 and exotic optical properties.21,22 However, 2D material-based flat optics is at an early stage of development, limited to specific optical configurations such as normal incidence with small numerical apertures.11–13 A primary bottleneck is the appalling angular performance, originating from a fundamental limitation on 2D materials; as a consequence of their atomic thinness, these materials interact only with in-plane polarized light, resulting in negligible out-of-plane responses (i.e., they exhibit strong anisotropy).23–25 Being able to generate out-of-plane optical responses is a key challenge for engineering light–matter interactions in the angular domain.

Here, we present 3D nanostructuring of anisotropic atomically thin materials to form optically isotropic films by creating out-of-plane optical responses (schematically depicted in Figure 1a). The main idea is to tilt and rotate the 2D crystalline domains using a 3D surface, for which the characteristic feature size is smaller than the wavelength of light. The subwavelength-scale reconstruction of 2D materials allows us to engineer the orientations of the anisotropic crystals and manipulate the light fields on the nanoscale topography. This enables the generation of optical isotropy in a predictable and systematic way: by balancing the out-of-plane and in-plane optical responses. To demonstrate this, we use transition metal dichalcogenides (TMDs),26–28 chosen due to their strong light–matter interactions, their ability to be grown directly onto diverse substrates as wafer-scale monolayer films,27 and their ability to be conformally grown in complex 3D geometries while preserving their intrinsic properties (e.g., chemical composition, crystal structure). These qualities enable the 3D topographic design of 2D materials to enhance the wide-angle performance while preserving or enhancing their unique excitonic,26 valley,29 and nonlinear properties.30

Using the aforementioned approach, we generate a 3D nanostructured TMD films, referred to here as dome-TMD. For this, an array of nanoscale dome structures is patterned with an average center-to-center distance (d ~ 150 nm) and height (h ~ 40 nm) that are smaller than visible wavelengths. This is followed by conformal growth of a monolayer TMD onto the prefabricated substrates. Thus, the TMD film is
distributed over the out-of-plane distance \( h \), and the crystalline domains in the film are arranged such that there exist out-of-plane orientations on the dome structures (Figure 1a). While only in-plane orientations occur at the peaks (A), out-of-plane orientations prevail at the bases of the domes (C), and tilted orientations, which are associated with both in-plane and out-of-plane optical responses, are found in the areas between (B).

Figure 1b shows morphology and atomic arrangement of monolayer dome-MoS\(_2\) imaged by scanning electron microscopy (SEM) and high-angle annular dark field (HAADF) scanning transmission electron microscopy (STEM; Methods).

As conceptually illustrated in Figure 1c, a MoS\(_2\) film on a flat surface, referred to here as flat-MoS\(_2\), interacts with light in a narrow angular distribution (since \( \chi_{\text{out}}/\chi_{\text{in}} = 0 \); ratio between out-of-plane and in-plane susceptibility), whereas dome-MoS\(_2\) exhibits a wide angular distribution due to the generation of an out-of-plane response (\( \chi_{\text{out}}/\chi_{\text{in}} > 0 \)). In dome-MoS\(_2\), the light field plays a crucial role in balancing the in-plane and out-of-plane film responses: the light field redistributes on the nanodome topography and is significantly amplified, particularly near the cusps between the domes, where the crystalline MoS\(_2\) domains are predominantly oriented out-of-plane. This effect is shown in our finite-difference time-domain simulations (Figure 1d and Methods). This enhancement of the out-of-plane response results in more isotropic films that have reduced polarization and incidence angle dependence. Such correlation between collective response and near-field interaction is one of the most crucial aspects in our study.

Figure 2a shows false-color optical images of three 2 in. dome-TMD films of monolayer MoS\(_2\), WS\(_2\), and WSe\(_2\) (Methods) that are homogeneous over the wafer scale. These films are produced in two steps (Methods). First, the curved nanostructures are generated by assembling a monolayer of silica nanospheres (diameter \( \sim d \)) on a fused silica substrate, followed by an etching process that transfers the nanosphere pattern into the substrate. After that, monolayer TMD films are conformally grown on these substrates using metal–organic chemical vapor deposition.\(^{16,17,27}\) The nucleation density, growth rate, and growth time are carefully controlled to ensure full monolayer coverage over the entire substrate. This can be seen in the SEM images of dome-MoS\(_2\) (darker regions) in Figure 2b where the film coverage reaches approximately 50%, 75%, (insets), and 100% (main panel) of the fused silica substrate. We further observe a nearly linear increase in the coverage with growth time (Supporting Information S1). The SEM images also show that the MoS\(_2\) covers the surface of the domes uniformly over the peaks and cusps. TEM characterizations confirm that dome-TMD is a polycrystalline film that conformally covers the dome surface by patching many grains whose average size (<100 nm) is smaller than the size of the domes (details will be published elsewhere).

Raman and photoluminescence measurements exhibit almost identical spectra for flat- and dome-MoS\(_2\) (including peak positions). This indicates that the dome-MoS\(_2\) is monolayer, with an averaged strain state similar to that of the flat-MoS\(_2\) (Supporting Information S2). In each dome-TMD film, optical transmission (\( \lambda = 532 \) nm) is measured to map the whole wafer, which shows only a small (<1%) variation along the radial direction of the wafer (Supporting Information S3). We also observe specular reflection with negligible light scattering, similar to a macroscopic partial mirror; i.e., the film is indeed optically flat (Supporting Information S4). These data confirm that our process produces nanostructured optical films with large-scale homogeneity.

We measure the optical properties using either a visible spectrometer or a collimated laser beam under ambient conditions while varying the light wavelength (\( \lambda \)), power, polarization, and incidence angle (\( \Theta \)) (Methods). To determine the absorption (A), the transmittance (T) and reflectance (R) are measured, and then A is calculated using A
plots the dome-MoS2 displays the polarization isotropy, which further diverge (both s- and p-polarization at normal incidence). However, the dependence of the absorption of dome- and flat-MoS2 is independent of the angular dependence (Figure 2d). The absorption in dome-MoS2 is enhanced over that of flat-MoS2 (dotted gray curve). Comparison of these curves shows that there is a similar angular dependence (more dramatic at higher angles. Additional experiments show that the optical response is largely a microscopic light–matter interactions in the near-field regime, leading to the absorption enhancement.

The observations described above (polarization isotropy and absorption enhancement) share two important features. They are each characterized by a broadband (or r-insensitive) absorption enhancement such as $A_{dome}/A_{flat} \approx 1$ (Figure 2c) and $A_{dome}/A_{flat} \approx 2.4$ (Figure 2d). This leads to the main advantage of our 3D nanostructuring: it can be used to enhance the optical performance of TMDs while maintaining their intrinsic optical spectra. In addition, as the performance values are largely determined by the geometry, they are similar for different TMD monolayers. Therefore, we can identify a design principle, which, once established in one TMD material, may be applied to diverse TMD films for angular isotropy (to be discussed in Figure 3). We will further present a general relation between the macroscopic optical properties and microscopic light–matter interactions in the near-field regime (to be considered in Figure 4).

We investigate the off-axis interaction of the nanostructured films to analyze absorption anisotropy on light polarizations (Figure 3a). For this, we consider $\chi_{in}$ ($\chi_{out}$), the in-plane (out-of-plane) sheet susceptibility,9,24 (unit: nm), of the monolayer film. $\chi_{in}$ ($\chi_{out}$) represents the polarizability in response to the electric field, which is correlated with absorption; for example, a freestanding (i.e., without substrate) monolayer film that has $\text{Im} \{ \chi_{in} \} / \text{Im} \{ \chi_{out} \} = 1$ exhibits polarization isotropy of absorption ($A_s = A_p$). Once the film is supported on a dielectric substrate, however, the isotropy is no longer conserved. This is because the macroscopic optical response is determined by the collective, subwavelength-scale interactions along the oscillating direction of electric field. Hence, the dielectric environment (e.g., air, substrate) needs to be considered, particularly for atomically thin films, for which the optical response is largely affected by the interface between the film and its surrounding dielectric.9,24 Because the interaction geometry is distinct for the p- and s-polarizations (Figure 3a), p-polarized light induces a larger polarization density $P = e_0 (E - 1)E$ onto the higher-index substrate.

Figure 2. Wide-angle, isotropic, and broadband absorption enhancement in wafer-scale monolayer dome-TMDs. a) False-color optical images of 2 in. dome-TMDs with wafer-scale uniformity. b) SEM image of continuous and conformal MoS2 monolayer films grown on close-packed nanodome topography. The inset shows the MoS2 films at approximately 50% and 75% surface coverages. c) Angular absorption with s- and p-polarized light in dome- and flat-MoS2. d) Absorption spectra of dome- and flat-MoS2. The flat-MoS2 spectrum is also shown multiplied by 2.4 for comparison.
Im{χ_{out}} and the roughly canceled out, mainly re
dependence. Thus, we may conclude that, in general, dome-
textures provide a more isotropic angular pros
effects of the TMD are
The consequence of this is that the out-of-plane
electric field caused by the fused silica and
our dome-structured MoS2 displays a large value varying
im{χ_{out}} increases, the
angle (Figure 3f).
Now, we consider the microscopic interactions in the near-
field regime to quantitatively explain the macroscopic film
responses. In nanostructured films, the local field of incident
light is redistributed at the subwavelength scale (Figure 4a),
and it determines χ_{in} and χ_{out} of effective optical films. For
quantitative analysis, we first measure the precise shape and
dimensions of the domes using cross-sectional SEM (Figure
4b). Then, we calculate the spatial map of the electric field vector E(r) = E_o + E’(r) using finite-difference time-domain simulations (λ = 532 nm), where E_o is the unperturbed electric field of light and E’(r) is the field caused by the fused silica and the MoS2 monolayer (Methods). The resulting maps of |E|² (Figure 1d) show strongly enhanced fields, as large as |E|² ∼ 5, near the cusps between domes. A map of P_{eff} the

| eps_{substrate} ~ 2.13), while s-polarized light induces a relatively
smaller P onto the lower effective-index medium of the air and
substrate (eps_{eff} ~ 1.61; calculated by a volume-averaging method31). The consequence of this is that the out-of-plane
electric field is more enhanced (due to the larger P) than the
in-plane electric field. Therefore, to obtain A_s ≈ A_p we need to
design smaller χ_{out} (or Im{χ_{in}}/Im{χ_{out}} > 1).

We quantitatively analyze the absorption anisotropy ratio ρ = (A_s − A_p)/(A_s + A_p) calculated as a function of Im{χ_{in}}/ Im{χ_{out}} and Θ (Supporting Information S7), which is visualized in Figure 3b. As Im{χ_{in}}/Im{χ_{out}} increases, the relative weight of the absorption is shifted from the p-
polarization dominant regime (blue; A_s < A_p) to the s-
polarization dominant one (red; A_s > A_p). It further predicts ρ ~ 0 (polarization isotropy) when Im{χ_{in}}/Im{χ_{out}} ∼ 2.4, as indicated by the black line in Figure 3b, which is insensitive to change in incident angle. Thus, angular isotropy can be achieved in the wide range of Θ by optimizing Im{χ_{in}}/ Im{χ_{out}}. In general, χ_{in} (or χ_{out}) is sensitive to the λ-
dependent properties of TMD. However, the ratio between the two (Im{χ_{in}}/Im{χ_{out}}), where the effects of the TMD are
roughly canceled out, mainly reflects the nanoscale geometry
and the field distribution in the 3D-textured substrate. This makes Im{χ_{in}}/Im{χ_{out}} a general design parameter applicable
for different TMDs, which can be tuned primarily through the
aspect ratio (height/radius) of the dome texture.

We perform absorption measurements to extract Im{χ_{in}}/ Im{χ_{out}} of our nanodrome-structured films (Supporting Information S8). Figure 3c plots Im{χ_{in}}/Im{χ_{out}} vs λ, measured from three different dome-TMDs (MoS2, WS2, WSe2), all of which share the same 3D nanostructured geometry. The graph also includes the values measured from a flat-MoS2 film for comparison. All three dome-TMDs display the same λ-independent value of Im{χ_{in}}/Im{χ_{out}} close to 2. In contrast, the flat-MoS2 displays a large value varying between 7 and 12 (i.e., strong anisotropy) with significant λ
dependence. Thus, we may conclude that, in general, dome-
TMDs show polarization isotropy (i.e., ρ ~ 0). This is what we
observe in the experiments shown in Figure 3d. The values of ρ measured from our dome-TMDs are observed to be close to zero, regardless of the choice of TMD material (MoS2, WS2, WSe2), the incidence angle (Θ between 0° to 60°), or the wavelength (λ between 400 nm and 650 nm).

Our approach for isotropy is not limited to light absorption
but is also applicable to light emission. While light emission
from flat surfaces is directional (from the surface normal),
dome-MoS2 provides a more isotropic angular profile. We use
Fourier-plane imaging (Methods) to demonstrate isotropic
photoluminescence (PL). In the flat film, the angular profile
(Ω(θ,ϕ) = I(θ,ϕ))/Imax) corresponds to the dipole radiation
pattern, where the angular profile changes as a function of cos2 Θ (Supporting Information S9). On the contrary, the emission profile of dome-MoS2 is more uniform than that of flat-MoS2 from center to the edge. The difference between two profiles, displayed in Figure 3e, (Ω_{dome} − Ω_{flat}) shows that the PL uniformity is significantly improved at higher angles (bright yellow ring). The angular plot of the differential change (Ω_{dome} − Ω_{im})/Ω_{im} monotonically increases, compensating for the deficiency of light emission from the flat surface at grazing
angle (Figure 3f).
Figure 4. Enhanced optical responses by nanotopography-driven field redistribution. a) Schematic of light field components projected to tangential planes on a dome structure with or without field redistribution. b) Cross-sectional SEM image of dome-MoS$_2$. c) Light energy flow visualized by Poynting vector. The color bar indicates flux direction (blue, $-\hat{x}$ direction; red, $+\hat{x}$ direction). d) Analytic relationship between absorption enhancement and field enhancement factor ($\alpha$). Blue and red dotted lines indicate the measured and the simulated absorption enhancements, respectively. e) Out-of-plane susceptibility measured in dome- and flat-MoS$_2$. f) Absorption in dome- and flat-MoS$_2$ as a function of light intensity ($\lambda = 532$ nm).

Poynting vector (Figure 4c), visualizes the energy flux of electromagnetic wave, where the flux directions are inverted across the peak of the dome, indicating that the energy is flowing toward the cusps. Similar results are also seen for different wavelengths and light polarizations (Supporting Information S10).

This field enhancement explains our main observation of absorption enhancement ($A_{\text{dome}}/A_{\text{flat}} \approx 2.4$). Since the optical absorption in MoS$_2$ is highly anisotropic, we consider the tangential component of the electric field, $\mathbf{E}_t(\theta, \phi) = \mathbf{E} \times \hat{n}$, where $\theta$ and $\phi$ are the angular coordinates and $\hat{n}$ is the surface normal vector (see Figure 4a). The absorption enhancement factor $A_{\text{dome}}/A_{\text{flat}}$ is then calculated from the ratio between the integrated value of $|\mathbf{E}_t|^2$ over the surface of a dome-MoS$_2$ film and the integrated value of $|\mathbf{E}_t|^2$ over a flat-MoS$_2$ film. Figure 4d plots the values of $A_{\text{dome}}/A_{\text{flat}}$ calculated for different total fields $E_o + \alpha E'$. Here, we introduce the unitless number $\alpha$ to represent conditions with no field enhancement ($\alpha = 0$), enhancement with the full strength predicted by our simulation ($\alpha = 1$), as well as other conditions. The graph shows that $A_{\text{dome}}/A_{\text{flat}} \sim 1.1$ when $\alpha = 0$, confirming that the enlarged surface area alone provides only $\sim$10% of the absorption increase, as slanted MoS$_2$ crystals absorb less. In contrast, we find that $\alpha = 1$ leads to $A_{\text{dome}}/A_{\text{flat}} \sim 2.3$ (red dotted line), which is close to the measured value ($A_{\text{dome}}/A_{\text{flat}} \sim 2.4$, blue dotted line). This agreement confirms that field enhancement quantitatively explains the increased absorption in dome-MoS$_2$.

Absorption enhancement in dome-MoS$_2$ indicates Im\(\chi_{\text{out}}\) is enhanced by approximately a factor of 2 due to the field enhancement (Supporting Information S8). We also observe a nearly 1 order of magnitude increase in Im\(\chi_{\text{out}}\) shown in Figure 4e. The local fields are concentrated at the cusp regions between the domes, which are predominantly occupied by the out-of-plane crystal orientations, which thus results in an even greater increase in Im\(\chi_{\text{out}}\) compared to Im\(\chi_{\text{in}}\), bringing balance between $\chi_{\text{in}}$ and $\chi_{\text{out}}$ for isotropic films (i.e., Im\(\chi_{\text{in}}\)/Im\(\chi_{\text{out}}\) ~ 2, shown in Figure 3c). In addition to linear susceptibility, an enhanced local field is also expected to strengthen high-order susceptibility:30 associated with nonlinear optical properties such as the onset of absorption saturation.32 To test this, we measure the absorption of dome- and flat-MoS$_2$ films (Figure 4f) as a function of the intensity of a continuous-wave laser beam ($\lambda = 532$ nm, $\Theta \sim 0^\circ$) (Methods). The values measured from the flat-MoS$_2$ film are constant over the entire intensity range, confirming that the response remains in the linear regime. In contrast, absorption measured from the dome-MoS$_2$ film deviates from the lower-intensity values above $\sim$10 kW/cm$^2$, a behavior that is reversible upon changing the light intensity. A similar power-dependent absorption is observed in Z-scan measurements (Supporting Information S11). This confirms that our dome-MoS$_2$ behaves as a saturable absorber at a relatively low light power.33 This observation is consistent with the field enhancement in dome-MoS$_2$.

Our work demonstrates that 3D nanostructuring of anisotropic 2D materials is a powerful and versatile approach to produce optically isotropic atomically thin films. Geometry-controlled crystal orientation and topography-driven field redistribution enable the generation and enhancement of out-of-plane optical responses to manipulate light–matter interactions in angular domains, in a given anisotropic atomically thin material. Since the concept of our geometric approach is material-independent, it can be applied to a variety of 2D materials beyond TMDs, such as graphene or hexagonal boron nitride, that absorb photons with lower (THz, infrared) or higher (ultraviolet) energies. In addition, the diverse library of 2D materials, which are available as metals, semiconductors, and insulators, can provide flexibility in the choice of dielectric...
constant. Our thin films compatible with wafer-scale, uniform, and optically flat substrates advance the development of compact and robust optical systems with atomically thin materials.

**ASSOCIATED CONTENT**

1. Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.1c02478.

Methods; nanoscale surface coverage and macroscopic optical transmission; Raman and photoluminescence of monolayer MoS2 films; dome-TMD films with 2 in. scale homogeneity; specular reflection in nanoscale textured films; wide-angle absorption enhancement over visible wavelengths; surface area increase in dome-TMD; polarization anisotropy in anisotropic 2D films; thin-film susceptibility of anisotropic 2D films; dipole radiation and emission profile in flat-MoS2; polarization dependence of field distribution in dome-MoS2; Z-scan measurement on saturable absorption in dome-MoS2 (PDF)

**AUTHOR INFORMATION**

**Corresponding Author**

Jiwoong Park — James Franck Institute, University of Chicago, Chicago, Illinois 60637, United States; Department of Chemistry and Pritzker School of Molecular Engineering, University of Chicago, Chicago, Illinois 60637, United States; Email: jwpark@uchicago.edu

Authors

Myungjae Lee — James Franck Institute, University of Chicago, Chicago, Illinois 60637, United States

Jong-Hoon Kang — Department of Chemistry, University of Chicago, Chicago, Illinois 60637, United States; orcid.org/0000-0002-5109-8402

Fauzia Mujid — Department of Chemistry, University of Chicago, Chicago, Illinois 60637, United States

Joonki Suh — Department of Chemistry, University of Chicago, Chicago, Illinois 60637, United States

Ariana Ray — Department of Physics, Cornell University, Ithaca, New York 14853, United States

Chiboom Park — James Franck Institute, University of Chicago, Chicago, Illinois 60637, United States; Department of Chemistry, University of Chicago, Chicago, Illinois 60637, United States; orcid.org/0000-0003-603-292X

David A. Muller — School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853, United States

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.nanolett.1c02478

**Author Contributions**

#M.L. and J.-H.K. had equal contribution.

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

We thank Jaehyung Yu, Jae-Ung Lee, Yu Zhong, Shi En Kim, Alexander A. High, and Sarah B. King for their helpful discussions. Primary funding for this work comes from Air Force Office of Scientific Research (FA9550-16-1-0347) and MURI projects (FA9550-18-1-0480 and FA9550-16-1-0031). Materials growths done by J.-H.K. and C.P. were supported by Samsung Advanced Institute of Technology. F.M. acknowledges support by the NSF Graduate Research Fellowship Program under Grant No. DGE-1746045. A.R. and electron microscopy at the Cornell Center for Materials Research are supported by the NSF-MRSEC grant DMR-1719875. The Titan microscope was acquired with the NSF MRI grant DMR-1429155. This work makes use of the characterization facilities of the University of Chicago MRSEC (NSF DMR-2011854) and the fabrication facilities of the Pritzker Nanofabrication Facility at the University of Chicago, which receives support from SHyNE Resource (NSF ECCS-4201542205), a node of NSF’s NNIC network.

**REFERENCES**

(1) Khorasaninejad, M.; Capasso, F. Metalenses: versatile multifunctional photonic components. Science 2017, 358, No. eaam8100.
(2) Khorasaninejad, M.; Chen, W. T.; Devlin, R. C.; Oh, J.; Zhu, A. Y.; Capasso, F. Metalenses at visible wavelengths: Diffraction-limited focusing and subwavelength resolution imaging. Science 2016, 352, 1190–1194.
(3) Adams, J. K.; Boomminathan, V.; Avants, B. W.; Vercosa, D. G.; Ye, F.; Baraniuk, R. G.; Robinson, J. T.; Veeraraghavan, A. Single-frame 3D fluorescence microscopy with ultraminature lensless FlatScope. Sci. Adv. 2017, 3, No. e1701548.
(4) Greenbaum, A.; Luo, W.; Su, T. W.; Göröcs, Z.; Xue, L.; Iskiman, S. O.; Coskun, A. F.; Mudanyali, O.; Ozcan, A. Imaging without lenses: achievements and remaining challenges of wide-field on-chip microscopy. Nat. Methods 2012, 9, 889–895.
(5) Arbabi, A.; Horie, Y.; Bagheri, M.; Faraon, A. Dielectric metasurfaces for complete control of phase and polarization with subwavelength spatial resolution and high transmission. Nat. Nanotechnol. 2015, 10, 937–943.
(6) Yang, M.; Horie, Y.; Shibukawa, A.; Brake, J.; Liu, Y.; Kamali, S. M.; Arbabi, A.; Ruan, H.; Faraon, A.; Yang, C. Wavefront shaping with disorder-engineered metasurfaces. Nat. Photonics 2018, 12, 84–90.
(7) Shaltout, A. M.; Lagoudakis, K. G.; van de Groep, J.; Kim, S. J.; Vučković, J.; Shalaev, V. M.; Brongersma, M. L. Spatiotemporal light control with frequency-gradient metasurfaces. Science 2019, 365, 374–377.
(8) Silva, A.; Monticone, F.; Castaldi, G.; Galdi, V.; Alù, A.; Engheta, N. Performing mathematical operations with metamaterials. Science 2014, 343, 160–163.
(9) Zhou, Y.; Zheng, H.; Kravchenko, I. I.; Valentine, J. Flat optics for image differentiation. Nat. Photonics 2020, 14, 316–323.
(10) Lin, X.; Rivenson, Y.; Yaridmci, N. T.; Velì, M.; Luo, Y.; Jarrahi, M.; Ozcan, A. All-optical machine learning using diffractive deep neural networks. Science 2018, 361, 1004–1008.
(11) van de Groeep, J.; Song, J. H.; Celano, U.; Li, Q.; Kik, P. G.; Brongersma, M. L. Exciton resonance tuning of an atomically thin lens. Nat. Photonics 2020, 14, 426–430.
(12) Back, P.; Zeytinoglu, S.; Ijaz, A.; Kroner, M.; Imamoğlu, A. Realization of an electrically tunable narrow-bandwidth atomically thin mirror using monolayer MoSe2. Phys. Rev. Lett. 2018, 120, 037401.
(13) Hu, G.; Hong, X.; Wang, K.; Wu, J.; Xu, H.-X.; Zhao, W.; Liu, W.; Zhang, S.; Garcia-Vidal, F.; Wang, B.; Lu, P.; Qiu, C.-W. Coherent steering of nonlinear chiral valley photons with a synthetic Au–WS2 metamaterial. Nat. Photonics 2019, 13, 467–472.
(14) Brongersma, M. L. The road to atomically thin metasurface optics. Nanophotonics 2020, 10, 643–654.
(15) Manzeli, S.; Ovchinnikov, D.; Pasquier, D.; Yazyev, O. V.; Kis, A. 2D transition metal dichalcogenides. Nat. Rev. Mater. 2017, 2, 17033.
(16) Kang, K.; Lee, K.-H.; Han, Y.; Gao, H.; Xie, S.; Muller, D. A.; Park, J. Layer-by-layer assembly of two-dimensional materials into wafer-scale heterostructures. Nature 2017, 550, 229−233.

(17) Xie, S.; Tu, L.; Han, Y.; Huang, L.; Kang, K.; Lao, K. U.; Poddar, P.; Park, C.; Muller, D. A.; DiStasio, R. A.; J.; Park, J. Coherent, atomically thin transition-metal dichalcogenide superlattices with engineered strain. Science 2018, 359, 1131−1136.

(18) Caldwell, J. D.; Aharonovich, I.; Cassabois, G.; Edgar, J. H.; Gil, B.; Basov, D. N. Photonics with hexagonal boron nitride. Nat. Rev. Mater. 2019, 4, 552−567.

(19) Amani, M.; Lien, D.-H.; Kiriya, D.; Xiao, J.; Azcatl, A.; Noh, J.; Madhavapathy, S. R.; Addou, R.; KC, S.; Dubey, M.; Cho, K.; Wallace, R. M.; Lee, S.-C.; He, J.-H.; Ager, J. W., III; Zhang, X.; Yablonovitch, E.; Javey, A. Near-unity photoluminescence quantum yield in MoS2. Science 2015, 350, 1065−1068.

(20) Ni, G. X.; Wang, L.; Goldflam, M. D.; Wagner, M.; Fei, Z.; McLoud, A. S.; Liu, M. K.; Kellmann, F.; Ozylmaz, B.; Castro Neto, A. H.; Hone, J.; Fogler, M. M.; Basov, D. N. Ultrafast optical switching of infrared plasmon polaritons in high-mobility graphene. Nat. Photonics 2016, 10, 244−247.

(21) Edalati-Boostan, S.; Cocchi, C.; Draxl, C. MoTe2 as a natural hyperbolic material across the visible and the ultraviolet region. Phys. Rev. Mater. 2020, 4, 085202.

(22) Harutyunyan, H.; Beams, R.; Novotny, L. Controllable optical negative refraction and phase conjugation in graphite thin films. Nat. Phys. 2013, 9, 423−425.

(23) Matthews, L.; Pulci, O.; Bechstedt, F. Influence of out-of-plane response on optical properties of two-dimensional materials: First principles approach. Phys. Rev. B: Condens. Matter Mater. Phys. 2016, 94, 205408.

(24) Majerus, B.; Dremetsika, E.; Lobet, M.; Henrard, L.; Kockaert, P. Electrodynamics of two-dimensional materials: Role of anisotropy. Phys. Rev. B: Condens. Matter Mater. Phys. 2018, 99, 125419.

(25) Oliva-Leyva, M.; de la Cruz, G. G. Unveiling optical in-plane anisotropy of 2D materials from oblique incidence of light. J. Phys.: Condens. Matter 2019, 31, 335701.

(26) Ugeda, M. M.; Bradley, A. J.; Shi, S. F.; Felipe, H.; Zhang, Y.; Qi, D. Y.; Ruan, W.; Mo, S. K.; Hussain, Z.; Shan, Z. X.; Wang, F.; Louie, S. G.; Crommie, M. F. Giant bandgap renormalization and excitonic effects in a monolayer transition metal dichalcogenide semiconductor. Nat. Mater. 2014, 13, 1091−1095.

(27) Kang, K.; Xie, S.; Huang, L.; Han, Y.; Huang, P. Y.; Mak, K. F.; Kim, C.-J.; Muller, D. A.; Park, J. High-mobility three-atom-thick semiconducting films with wafer scale homogeneity. Nature 2015, 520, 656−660.

(28) Jin, G.; Lee, C.-S.; Liao, X.; Kim, J.; Wang, Z.; Okello, O. F. N.; Park, B.; Park, J.; Han, C.; Heo, H.; Kim, J.; Oh, S. H.; Choi, S.-Y.; Park, H.; Jo, M.-H. Atomically thin three-dimensional membranes of van der Waals semiconductors by wafer-scale growth. Sci. Adv. 2019, 5, No. eaaw3180.

(29) Gong, S. H.; Alpeggiani, F.; Sciacca, B.; Garnett, E. C.; Kuipers, L. Nanoscale chiral valley-photon interface through optical spin-orbit coupling. Science 2018, 359, 443−447.

(30) Liu, H.; Li, Y.; You, Y. S.; Ghimire, S.; Heinz, T. F.; Reis, D. A. High-harmonic generation from an atomically thin semiconductor. Nat. Phys. 2017, 13, 262−265.

(31) Zhang, C.; Yi, P.; Peng, L.; Ni, J. Optimization and continuous fabrication of moth-eye nanostructure array on flexible polyethylene terephthalate substrate towards broadband antireflection. Appl. Opt. 2017, 56, 2901−2907.

(32) Martinez, A.; Sun, Z. Nanotube and graphene saturable absorbers for fibre lasers. Nat. Photonics 2013, 7, 842−845.

(33) Wang, K.; Wang, J.; Fan, J.; Lotya, M.; O’Neil, A.; Fox, D.; Feng, Y.; Zhang, X.; Jiang, B.; Zhao, Q.; Zhang, H.; Coleman, J. N.; Zhang, L.; Blau, W. J. Ultrafast saturable absorption of two-dimensional MoS2, nanosheets. ACS Nano 2013, 7, 9260−9267.