

Nanoscience, Nanotechnology, Nanomaterials

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Acoustically Active Surfaces Based on Piezoelectric Microstructures

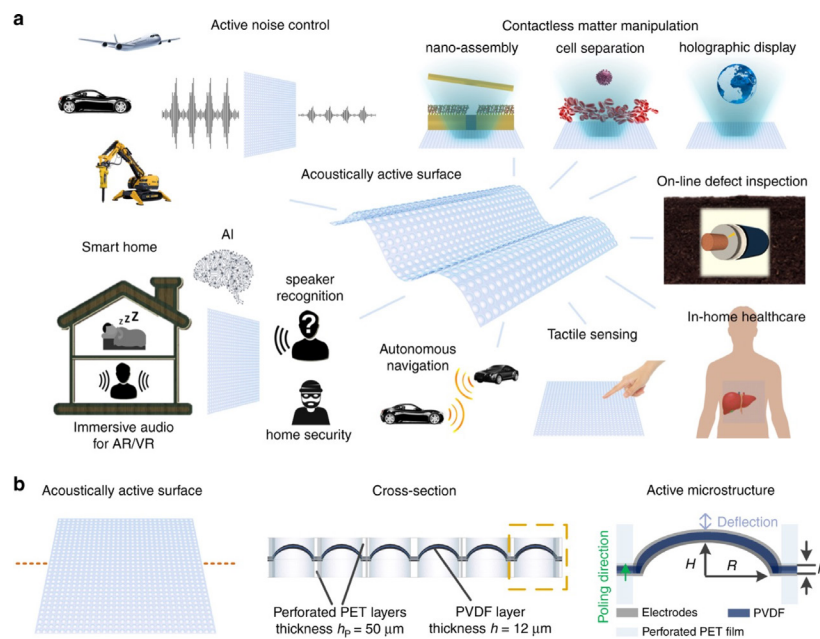
T. Dang, J. Han, J. Lang, V. Bulović
Sponsorship: Ford Motor Company, Lendlease Group

Acoustic transducers have attracted significant attention due to their ubiquitous application in modern-day technologies, such as active noise control, human-machine interfaces, ultrasonic imaging, and tactile sensing (Figure 1a). The industrial and consumer demand for using sound as a sensing and actuation medium has encouraged the development of cost-effective, scalable, and high-performance loudspeakers. Commercial speakers typically generate sound based on electrostatic or piezoelectric effects; among these, piezoelectric loudspeakers have stood out due to their simple structures and low power consumption.

In this project, we develop a flexible thin-film acoustic transducer based on an ensemble of free-standing microstructures using a piezoelectric polyvinylidene fluoride (PVDF) sheet. The

microstructures follow the shape of a dome fabricated through a vacuum-induced self-aligned micro-embossing process (Figure 1b). These PVDF dome arrays are sandwiched between two layers of perforated polyethylene terephthalate (PET) sheets, which serve as both the embossing mold and a protection layer to ensure the free vibration of the piezoelectric microstructures.

The performance of the speakers is subsequently measured, and they demonstrate excellent sound generation and sensing capabilities as well as high sensitivity and large bandwidths. Our ongoing work focuses on scaling up the current speakers and integrating them into various sound-generating applications.



▲ Figure 1: Concept and structure of acoustically active surfaces. (a) Application prospects of acoustic surfaces. (b) Schematic showing the cross section of an acoustic surface and a magnified picture of an active microstructure within the array.

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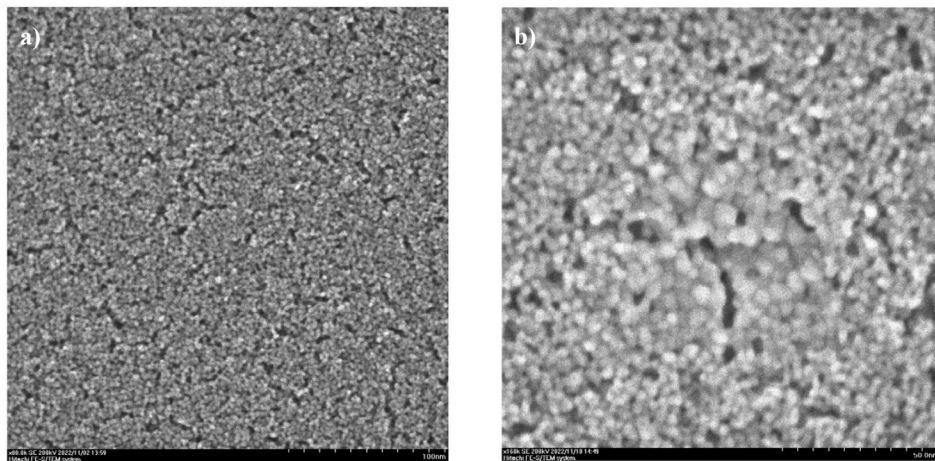
Morphological Change of Hydrogen Gas Doped Metal Oxide Nanoparticles under Charge Flow

J. Geng, M. Saravanapavanantham, A. Penn, V. Bulović
Sponsorship: Samsung

Quantum dot light-emitting diodes (QD-LEDs) emit color-pure, easily tunable, bright light and are thus a promising new technology for lighting and display applications. However, before market viability, they must be made more efficient and durable. Zinc magnesium oxide (ZnMgO) nanoparticles serve as an excellent electron transport layer in indium phosphide QD-LEDs. It has been observed that operating QD-LEDs under hydrogen gas flow causes performance improvement over a short timescale.

To investigate this effect, we simultaneously flowed hydrogen gas over a drop-cast layer of ZnMgO nanoparticles and imaged the layer in an in-situ transmission electron microscope (TEM). Figure 1 shows images of the nanoparticles before and during

hydrogen gas flow. We observed that the nanoparticles coarsen over a few minutes once doped with hydrogen and irradiated with the electron beam, which simulates the passage of charge through the device. The resulting doped and irradiated spots are comprised of larger, fewer dots and significant cracks. This effect only occurs in areas of dense nanoparticle population, where particles may merge. These observations allow us to hypothesize that the larger dots allow for better charge transport through the layer due to fewer necessary grain boundary crossings. These results provide a physical explanation for improved QD-LED performance under hydrogen and may inform further development of the InP QD-LED.



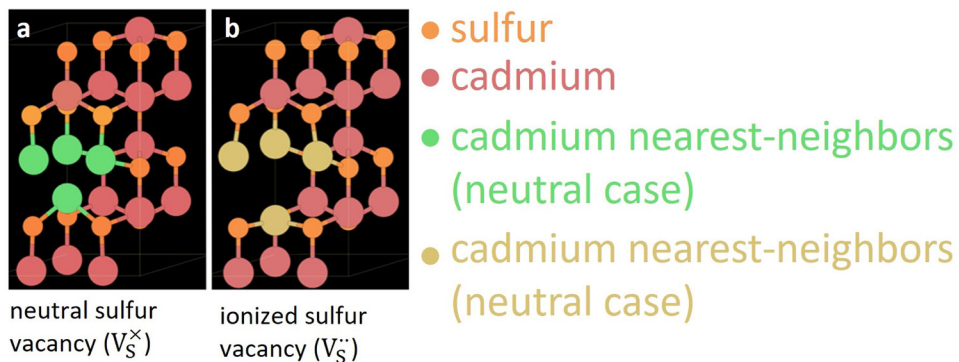
▲ Figure 1: TEM images of ZnMgO nanoparticle layer a) before and b) during hydrogen gas flow.

Using Point Defects to Control Giant Opto-mechanical Effects in Wide-band-gap Semiconductors

J. Dong, Y. Li, Y. Zhou, A. Schwartzman, H. Xu, B. Azhar, J. Bennett, J. Li, R. Jaramillo
Sponsorship: Office of Naval Research (Grant No. N00014-17-1-2661)

Semiconductor point defects that create deep charge traps are often to be avoided because they speed up bad things (e.g., non-radiative recombination) and slow down good things (e.g., photodetector response). Nevertheless, deep traps often feature large charge-lattice coupling phenomena that are fundamentally interesting and could be put to good use. We found experimentally that deep traps in II-VI semiconductors CdS, ZnS, and ZnO are correlated with giant opto-mechanical effects, such as changes in hardness and elastic modulus greater than 50% under relatively mild illumination

(e.g., blue light at 1.5 mW/cm²) during nanoindentation, and that these giant effects can be tuned by materials processing. Giant photoplasticity has been reported and understood conceptually for decades, but the giant photoelastic effect that we observe has not been understood. Using density functional theory, we find that giant photoelasticity can arise from changes in lattice configuration resulting from defect ionization (i.e., DX center phenomena). This places photoelasticity on a firm theoretical footing and paves the way for parametric design of devices using the effect.



▲ Figure 1: Illustrations of changed atomic positions around sulfur vacancy in CdS, as vacancy changes from neutral (a) to doubly ionized state (b). Illustrations project atomic positions computed using density functional theory. Colors highlight changed positions of four Cd atoms nearest vacancy.

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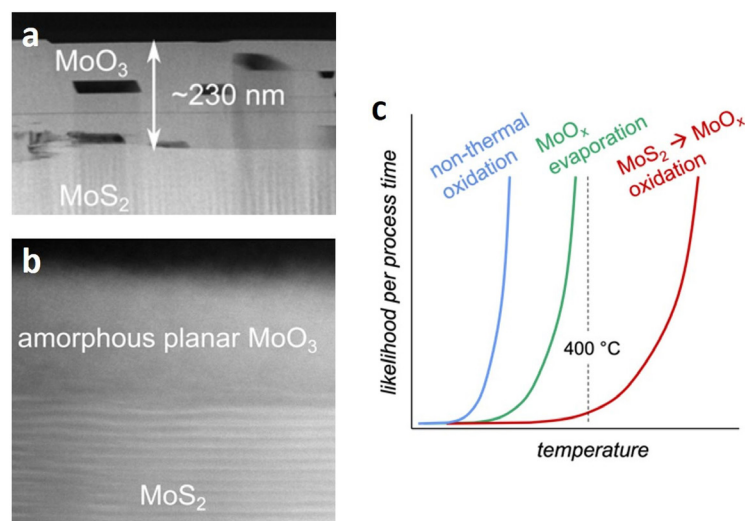
Atomic-scale Mechanisms of MoS₂ Oxidation for Kinetic Control of MoS₂/MoO₃ Interfaces

K. Reidy, W. Mortelmans, S. S. Jo, A. N. Penn, B. Wang, A. C. Foucher, F. M. Ross, R. Jaramillo
Sponsorship: Semiconductor Research Corporation, Office of Naval Research (Grant No. N00014-17-1-2661)

Like most semiconductors, the surfaces of transition metal dichalcogenides (TMDs) are prone to oxidation. TMDs are now on development roadmaps for continued transistor scaling, which increases the importance of controlling TMD oxidation and understanding the electrical properties of interfaces between TMDs and their oxides. A lesson learned from Si microelectronics is that, if at all possible, we should make use of semiconductor native oxides. Particularly for semiconducting TMDs for which the native oxides may be useful dielectrics (or even ferroelectrics), there is an opportunity to better understand the processing-property relationships that control dielectric response, leakage, and interface quality. For TMDs for which the native oxides have easily varied conductivity, there is an opportunity to develop resistive switching functionality, or to develop native oxide electrodes.

Here, we investigate the atomic scale oxidation

mechanisms of the most widely studied TMD, MoS₂. We find that thermal oxidation results in α -phase crystalline MoO₃ with sharp interfaces, voids, and a textured alignment with the underlying MoS₂. Experiments with remote substrates prove that thermal oxidation proceeds via vapor-phase mass transport and redeposition, due to rapid evaporation of the oxide—a challenge to forming thin, conformal planar oxide films. We accelerate the kinetics of oxidation relative to the kinetics of mass transport using a non-thermal oxygen plasma to form a smooth and conformal oxide. The resulting amorphous MoO₃ films can be grown several nanometers thick, and we calibrate the oxidation rate for varying processing conditions. Our results provide quantitative guidance for managing both the atomic scale structure and thin film morphology of oxides in the design and processing of TMD devices.



▲ Figure 1: Kinetics of thermal vs. non-thermal oxidation. (a) Transmission electron micrograph (TEM) of a crystalline, non-uniform MoO₃ layer formed by thermal oxidation of MoS₂. (b) TEM of an amorphous, uniform MoO₃ layer formed by plasma oxidation of MoS₂. (c) Schematic of rates of relevant processes: thermal oxidation, evaporation, and non-thermal oxidation.

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Chemical Vapor Deposition Synthesis of Multilayer Hexagonal Boron Nitride

H. Liu, Z. Luo, J. Kong

Sponsorship: EECS Faculty Research Innovation Fellowship

Hexagonal boron nitride (hBN) has attracted tremendous research interest due to its wide band gap of up to ~6 eV, remarkable chemical and physical stability, atomic surface flatness, etc. As such, hBN serves as a promising candidate as quantum emitters, insulators, or support for the next generation of electronic and photonic devices. Recent reports on the wafer-scale synthesis of hBN are limited to monolayer or few layers, which is due to the self-limited growth regime for the growth substrates with low nitrogen solubility. However, monolayer or few-layer hBN is limited in its performance, some of the problems include unstable optical emission, non-satisfied mechanical strength, inability to screen out substrate effects, etc. In this work, we propose to utilize single crystalline metal alloys as

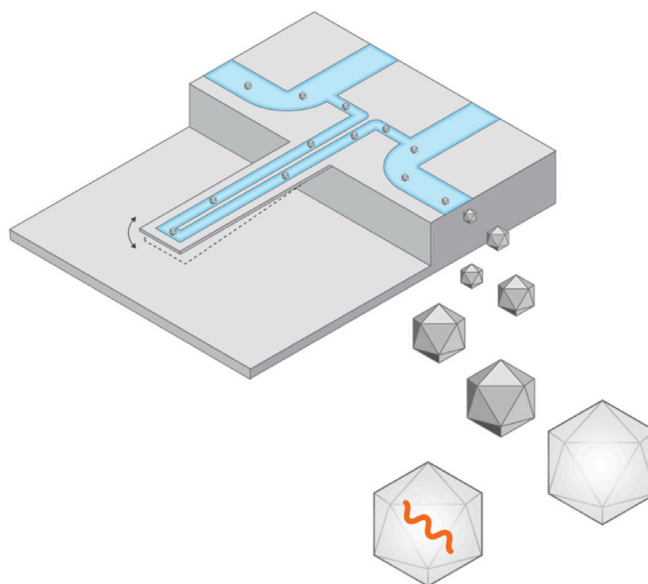
the growth substrate to synthesize multilayer hBN by chemical vapor deposition. The uniformity of both substrates and the as-synthesized hBN films will be characterized by optical contrast and Raman spectroscopy mapping. The crystallinity of the hBN films will be characterized by XRD and the full width at half maximum of Raman peaks corresponding to hBN. Atomic force microscopy will be used to measure the hBN thicknesses and electron microscopy will be conducted to reveal the microstructure of hBN films. The success of this work will help to improve the next generation of 2D materials-based electronic devices, finally leading to a world with a higher information transport rate with less energy consumption compared to the current Si-based devices.

Weighing the DNA Content of Adeno-associated Virus Vectors with Zeptogram Precision using Nanomechanical Resonators

G. Katsikis, I. E. Hwang, W. Wang, V. S. Bhat, N. L. McIntosh, O. A. Karim, B. J. Blus, S. Sha, V. Agache, J. M. Wolfrum, S. L. Springs, A. J. Sinskey, P. W. Barone, R. D. Braatz, S. R. Manalis
Sponsorship: U.S. Food and Drug Administration

Quantifying the composition of viral vectors used in vaccine development and gene therapy is critical for assessing their functionality. Adeno-associated virus (AAV) vectors, which are the most widely used viral vectors for in vivo gene therapy, are typically characterized using polymerase chain reaction (PCR), enzyme-linked immunosorbent assay (ELISA), and analytical ultracentrifugation, which require laborious protocols or hours of turnaround time. Emerging methods such as charge-detection mass spectroscopy, static light scattering, and mass photometry offer turnaround times

of minutes for measuring AAV mass using optical or charge properties of AAV. Here, we demonstrate an orthogonal method where suspended nanomechanical resonators (SNR) are used to directly measure both AAV mass and aggregation from a few microliters of sample within minutes. We achieve a precision near 10 zeptograms, which corresponds to 1% of the genome holding capacity of the AAV capsid. Our results show the potential of our method for providing real-time quality control of viral vectors during biomanufacturing.



▲ Figure 1: Concept of measuring mass of AAVs in solution showing a schematic of a hollow cantilever of length 17.5 μm . Inside the cantilever, we flowed solutions of AAVs with different DNA content or genetic constructs (denoted by orange).

FURTHER READING:

- G. Katsikis, I. E. Hwang, W. Wang, V. S. Bhat, N. L. McIntosh, O. A. Karim, B. J. Blus, S. Sha, V. Agache, J. M. Wolfrum, S. L. Springs, A. J. Sinskey, P. W. Barone, R. D. Braatz, and S. R. Manalis, "Weighing the DNA Content of Adeno-Associated Virus Vectors with Zeptogram Precision Using Nanomechanical Resonators," *Nano Letters*, vol. 22, no. 4, pp. 1511–1517, Feb. 2022.

Modelling Interfacial Competition on Drug Crystal Surfaces Using Molecular Dynamics Simulations

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Sponsorship: MIT UROP Office

Oral administration is often preferred among all drug administration routes due to its simplicity, low cost, and convenience. However, nearly 90% of drug candidates in the pharmaceutical development pipeline have high hydrophobicity, which significantly hinders their dissolvability in the gastrointestinal (GI) tract and limits bioavailability. Our lab has developed a suite of 'bottom-up' methods to template drug nanocrystals by embedding APIs in polymeric matrices as an approach to reduce drug aggregate sizes for improved bioavailability. However, recent experimental results have motivated the need for a mechanistic understanding of the influence of excipient-drug interactions during processing.

In this work, we present the use of molecular dynamics (MD) simulations to advance understanding of how molecular interactions between excipients and drug control the self-assembled nanostructure on drug crystal surfaces. MD is used to explore the compositional effects of surfactant and polymer excipients interacting on the surface of a model hydrophobic API (fenofibrate). The role of surfactant in screening polymer-drug interactions is elucidated as an important mechanism for designing 'bottom-up' small molecule formulation processing schemes. Based on the results of this work, various combinations of drugs, surfactants and polymer matrices can be tested computationally and experimentally, leading to the next generation of oral drug delivery forms.

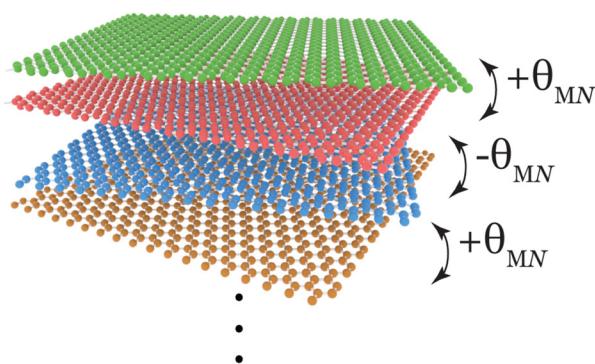
Discovery of a Family of Magic-angle Twisted Graphene Superconductors

J. M. Park, Y. Cao, L. Xia, S. Sun, K. Watanabe, T. Taniguchi, P. Jarillo-Herrero
Sponsorship: Department of Energy, NSF

When materials are thinned down to an atomic limit, properties vastly different from their three-dimensional counterparts appear. Further, these two-dimensional (2D) materials can be stacked to create a new type of platform called van der Waals heterostructures, which exhibit novel phases not present in the constituent layers. More recently, another degree of freedom that controls the relative twist angle between the adjacent layers has been explored, giving rise to the field of moiré systems. In particular, when two layers of graphene were stacked with a relative twist angle of 1.1 degrees, unexpected insulator and superconductor were observed. After the discovery of such magic-angle twisted bilayer graphene (MATBG) system, various kinds of moiré systems were investigated, and novel interaction-driven phenomena including correlated insulators, quantum anomalous Hall effect, ferromagnetism, and generalized Wigner crystal were found.

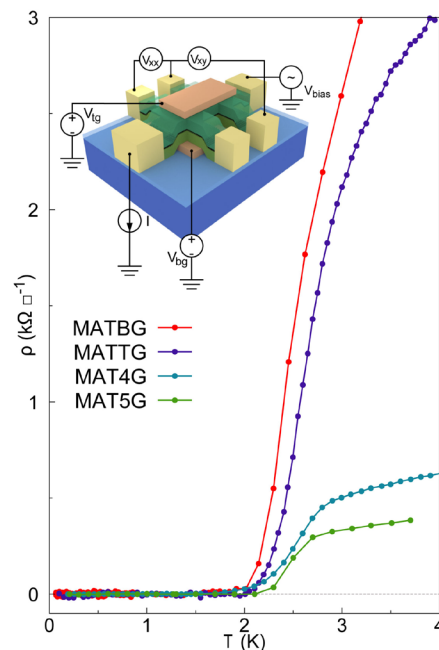
Despite the numerous correlated phases observed

in moiré systems, signatures of robust and reproducible superconductivity have been rare and only found in MATBG and more recently in MAT trilayer graphene. In this study, we report the experimental realization of superconducting MAT four-layer and five-layer graphene, therefore establishing alternating twist magic-angle multilayer graphene as a robust family of moiré superconductors. This finding suggests that the flat bands present in all members play a crucial role in forming the superconductivity. Our measurements in parallel magnetic fields, particularly the investigation of Pauli limit violation and spontaneous rotational symmetry breaking, reveal a clear distinction between the $N=2$ and $N>2$ -layer structures, consistent with the difference between their orbital responses to magnetic fields. Our results expand the emergent family of moiré superconductors, providing novel insight into the design of unconventional superconducting materials platforms.



▲ Figure 1: Magic-angle twisted multilayer graphene, where the adjacent layers are twisted in an alternating direction.

► Figure 2: Zero resistivity shows robust superconductivity in the magic-angle twisted multilayer graphene family.



FURTHER READING

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Predicting Coherent Nanocrystal Orientation in Nanocrystal Superlattices by Minimizing Ligand Packing Frustration

E. K. Price, W. A. Tisdale

Sponsorship: DoE Office of Science, Basic Energy Sciences, NSF Graduate Research Fellowships Program (GRFP)

Semiconducting, colloidal nanocrystals (NCs) have size-tunable optical and electronic properties and show promise for next-generation sensing, photovoltaic, and computing devices. For integration into optoelectronic devices, NCs are assembled into ordered superlattices (SLs). Since the SL structure can influence charge and thermal transport in these devices, robust engineering control over SL formation is needed. A number of factors influence the self-assembly of colloidal NCs, including the well-studied impacts of nanocrystal size and shape. In recent years, the importance of both bound and unbound organic ligands on self-assembly outcomes has been highlighted; the ligand length, ligand coverage, bound and unbound ligand fractions, and ligand interactions can all influence the resulting NC SL structure. In this work, we consider how the classic influence of nanocrystal shape can impact ligand packing frustration and influence the coherent orientation of non-spherical NCs in NC

SLs. Through the application of the freely jointed chain model to estimate conformational entropy, we find that minimizing the packing frustration of the ligand layer may explain experimental observations of NC alignment in NC SLs. We validate our model by comparing to published X-ray scattering data showing the kinetics of lead sulfide (PbS) superlattice formation. These data show that as solvent is evaporated, the SL exhibits a Bain-like distortion, contracting from an fcc to bcc configuration, and the coherent NC tilt relative to the substrate changes from 9.7° to 0° . The thermodynamic understanding of coherent NC orientation in NC SLs explored in this work may inform the synthesis of high-quality films with minimal disorder for integration into optoelectronic devices and enable the experimental realization of predicted theoretical properties such as band-like transport in NC SLs.

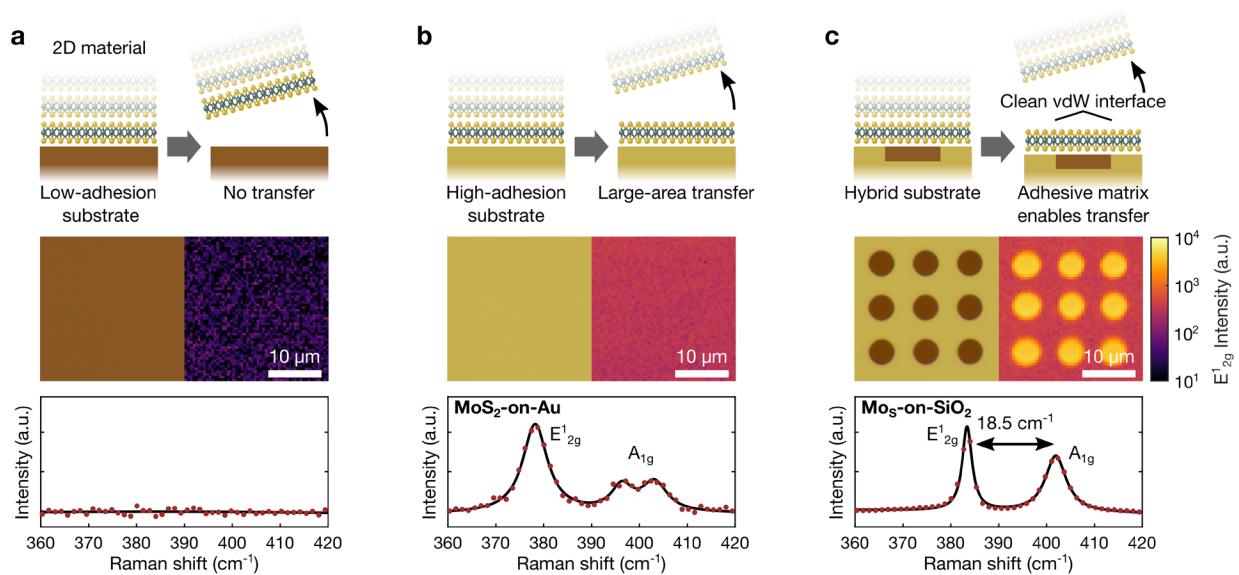
Van Der Waals Integration Beyond the Limits of Van Der Waals Forces

P. F. Satterthwaite, W. Zhu, P. Jastrzebska-Perfect, M. Tang, H. Gao, H. Kitadai, A.-Y. Lu, Q. Tan, J. Kong, X. Ling, F. Niroui
Sponsorship: NSF Center for Energy Efficient Electronics Science (E3S) (ECCS-093951), NSF EAGER (2135846), NSF Graduate Research Fellowship Program (1745302)

Fabrication of pristine van der Waals (vdW) interfaces between two-dimensional (2D) and other materials is essential for emerging optical and electronic devices. The weak vdW forces at the interface of interest cannot, however, be readily tuned to enable direct integration of arbitrary materials. Conventionally, this is addressed by transferring the 2D material using sacrificial layers, solvents and high-temperatures, introducing damage and contaminants. Conventional device integration further requires post-transfer fabrication steps which can lead to device performance being constrained by the processing artifacts rather than the intrinsic materials properties.

Here, we introduce a novel fabrication platform, adhesive matrix transfer, which enables direct 2D material-to-device integration in a single, dry step.

This is achieved by decoupling the forces enabling transfer from the forces at the interface of interest. For example, 2D semiconductors such as MoS_2 cannot be directly transferred to dielectrics such as SiO_2 , low adhesion but important substrates for device integration (Fig. 1a). In contrast, direct transfer of monolayer MoS_2 to gold can be achieved (Fig. 1b), but MoS_2 -on-gold is of limited technological value. By using gold as an adhesive matrix surrounding SiO_2 , MoS_2 -on- SiO_2 can be directly fabricated with no exposure to solvents or polymers (Fig. 1c). Using this transfer, we further demonstrate clean, direct fabrication of MoS_2 transistors demonstrating the ability of our platform to enable direct 2D material-to-device integration.



▲ Figure 1: Adhesive matrix transfer. (a) Direct transfer of MoS_2 to SiO_2 is not possible due to weak adhesive interactions. (b) MoS_2 can, however, be directly transferred to gold. (c) Using gold as an adhesive matrix, MoS_2 -on- SiO_2 can be directly fabricated. Top row shows schematic illustration of experiments. Middle row shows optical micrographs, and Raman intensity mapping. Bottom row shows characteristic Raman spectra.

Nonplanar Nanofabrication via Interface Engineering

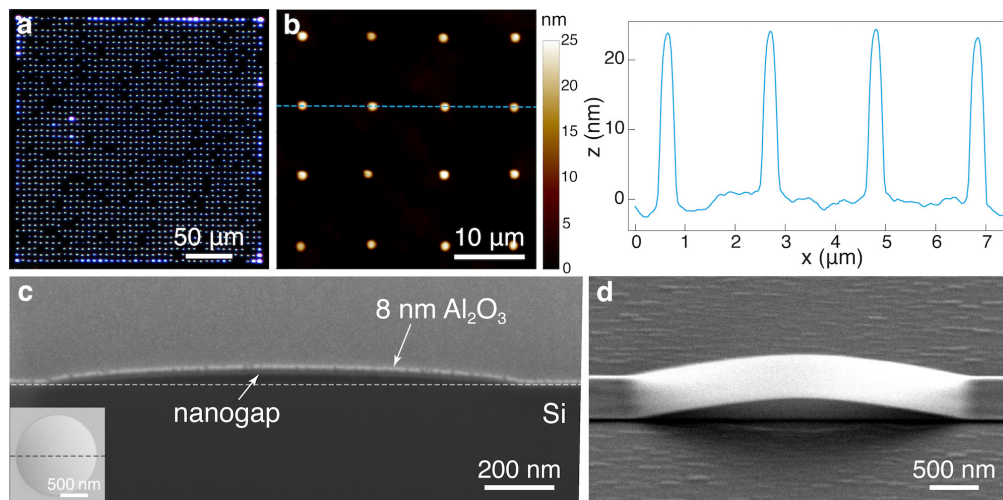
S. O. Spector, P. F. Satterthwaite, F. Niroui

Sponsorship: NSF Graduate Research Fellowship, Samsung Electronics Fellowship

Nonplanar nanostructures, composed of suspended ultrathin films and nanogaps, are foundational for next-generation miniaturized nanoelectromechanical devices, photonic elements, and metamaterials. However, resolution constraints and instabilities due to nanoscale forces limit their fabrication through conventional top-down techniques.

This work reports a new approach for scalable fabrication of suspended, ultrathin nanostructures with controlled nanoscale gaps, without the use

of a sacrificial layer. We engineer surface adhesive forces through a patterned molecular monolayer to enable controlled delamination of an oxide thin film in predetermined locations. By extending standard, wafer-scale, and conventionally compatible planar fabrication techniques, we form nonplanar structures with thicknesses < 10 nm and nanogaps reaching < 10 nm. Using this approach, we demonstrate ultrathin mechanical resonators in the megahertz range with applications in ultrasensitive sensors.



▲ Figure 1: (a) Darkfield image of an array of dome-shaped Al₂O₃ nanostructures. (b) Atomic force microscopy (AFM) and line scan of a nanostructure array. (c) Cross-sectional scanning electron microscopy (SEM) of a fabricated 8 nm-thin suspended membrane. (d) SEM of a doubly clamped, 8 nm-thin mechanical resonator.

Role of Local Gyrotropic Force in Distortion-limited High-speed Dynamics of Antiferromagnetic Skyrmion

E. A. Tremsina, G. S. D. Beach

Sponsorship: NSF GRFP, DARPA TEE Program, SMART, one of seven centers of nCORE, a Semiconductor Research Corporation program, sponsored by NIST

Magnetic solitons, quasiparticles formed by a local twist in the magnetization, are great candidates for use in novel spintronic devices. The key question for their use in practical applications, however, is the maximum speed of propagation through magnetic media. For one-dimensional solitons (domain walls), the high-speed dynamics are well understood and are limited in antiferromagnetic materials by relativistic-like effects, namely velocity saturation towards a speed akin to the speed of light, and Lorentz-like width contraction. Two-dimensional solitons (skyrmions) instead exhibit elliptical distortions followed by a full breakdown at a critical limiting velocity, and the exact nature of this

process is yet to be understood. Here, the unique capabilities of a fully atomistic model are used to perform an extensive and systematic study of soliton dynamics. As a result, a physical explanation for skyrmion deformations, which are attributed to the local imbalance of the gyrotropic forces, is derived using numerical simulation data. It is shown also that the inherent skyrmion structure impedes their ability to even reach the velocity regime where relativistic effects could begin to occur. These results expand the understanding of the fundamental properties of magnetic skyrmions, in particular, their dynamical stability at high speeds, as well as their potential use for spintronic applications.

Metal-graphene Interface in Evaporative-deposition of Various Metals on Graphene

Z. Wang, H. Wang, J. Zhu, T. Pieszkow, X. Zheng, S. A. Vitale, Y. Han, J. Kong

Sponsorship: MIT DMSE and EECS, Rice University Materials Science and NanoEngineering, MIT Lincoln Laboratory

Graphene, the first member in the two-dimensional (2D) materials family, has unique electrical, optical, and mechanical properties, which opens vast opportunities for applications. The metal-graphene interface is crucial for many research studies and applications but has not been understood thoroughly due to the difficulty in characterizing it. Here we observed that when 8-15 nanometers of metal is deposited onto chemical vapor

deposition- (CVD) grown graphene, certain types of metal exhibit a change in optical contrast compared to the area without graphene underneath, but for others no such change is noticeable. In this work we carried out various investigations to understand the reasons behind the phenomena and explored their potential applications.

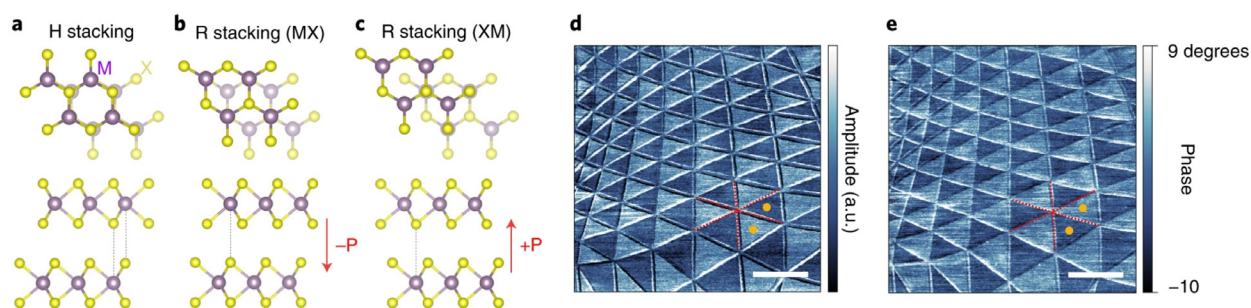
Interfacial Ferroelectricity in Rhombohedral-stacked Bilayer Transition Metal Dichalcogenides

X. Wang, K. Yasuda, Y. Zhang, S. Liu, K. Watanabe, T. Taniguchi, J. Hone, L. Fu, P. Jarillo-Herrero
Sponsorship: DoE, ARO, Gordon and Betty Moore Foundations, NSF

Two-dimensional (2D) ferroelectrics have great potential for dense and low-consumption nonvolatile memory applications. However, there are rare experimental reports due to the demand of layered polar bulk material. Van der Waals (vdW) materials enable the fabrication of heterostructures by stacking one layer of crystal on top of another at a controlled angle. These heterostructures combine characteristics of the individual building blocks but can also exhibit physical properties absent in the parent compounds through interlayer interactions and therefore can greatly expand the design space of 2D ferroelectrics.

Here we report on a new family of nanometer-thick, 2D ferroelectric semiconductors, in which the individual constituents are well-studied non-ferroelectric monolayer transition metal dichalcogenides (TMDs), namely WSe_2 , MoSe_2 , WS_2 , and MoS_2 . Specifically, we cut a monolayer TMD into two pieces and stack one half on top of the other

in parallel. This forms the rhombohedral-stacking (R-stacking) configuration different from the natural 2H phase. We demonstrate that robust out-of-plane ferroelectricity exists in such R-stacked bilayer TMDs, and the polarization can be flipped via in-plane sliding motion between the two monolayers. We visualize the ferroelectric domains as well as electric-field-induced domain wall motion with piezoelectric force microscopy. Furthermore, we probe the polarization switching of the bilayer via an adjacent graphene layer in a ferroelectric field transistor geometry and quantify the ferroelectric built-in interlayer potential, which is in good agreement with first-principles calculations. Our demonstration of ferroelectricity in stacking-engineered TMD bilayers consolidates the feasibility of engineering 2D ferroelectric semiconductors and opens up a broad way of engineering various functional heterostructures out of non-ferroelectrics.



▲ Figure 1: (a) H-stacked bilayer TMD. M, metal atom (W or Mo). X, chalcogen atom (S or Se), (b) MX and (c) XM stacking forms of R-stacked bilayer TMD, (d) amplitude and (e) phase images of vertical piezoelectric force microscopy (PFM) on twisted MoSe_2 . Scale bars, 200 nm.

FURTHER READING

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Electrical Switching of a Bistable Moiré Superconductor

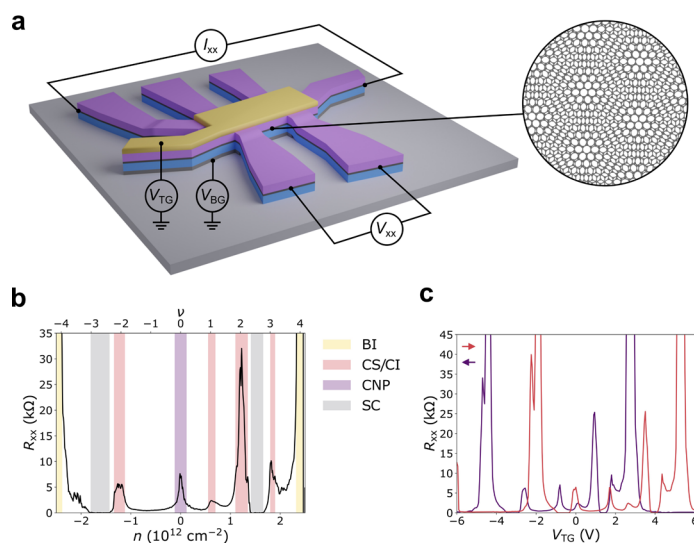
D. R. Klein, L.-Q. Xia, D. MacNeill, K. Watanabe, T. Taniguchi, P. Jarillo-Herrero

Sponsorship: Air Force Office of Scientific Research, Army Research Office, Gordon and Betty Moore Foundation, NSF

Electrical control of superconductivity is critical for nanoscale superconducting circuits including cryogenic memory elements, superconducting field-effect transistors (FETs) and gate-tunable qubits. Superconducting FETs operate through continuous tuning of carrier density, but no bistable superconducting FET, which could serve as a new type of cryogenic memory element, has been reported.

Recently, experimentally realized magic-angle twisted bilayer graphene (MATBG) emerged as a highly-tunable platform for studying strongly correlated phenomena, including superconductivity, correlated insulators, orbital magnetism and anomalous Hall effect, and strange metal behavior. Separately, Bernal-stacked bilayer graphene aligned to its insulating hexagonal boron nitride gate dielectrics has been reported to manifest gate hysteresis and bistability. Here we report the observation of this

same hysteresis in MATBG with aligned boron nitride layers. This bistable behavior coexists alongside the strongly correlated electron system of MATBG without disrupting its correlated insulator or superconducting states (see Figure 1). This all-van der Waals platform enables configurable switching between different electronic states of this rich system. To illustrate this new approach, we demonstrate reproducible bistable switching between the superconducting, metallic, and correlated insulator states of MATBG using gate voltage or electric displacement field. The combination of these results with other configurable MATBG superconducting devices, including Josephson junctions and superconducting quantum interference devices, will enable an additional control knob over the electronic states and pave the way for a new generation of switchable moiré graphene superconducting electronics.



▲ Figure 1: a. Schematic of the MATBG device. b. Measured longitudinal resistance R_{xx} . The band insulators (BI), correlated semimetals/insulators (CS/CI), charge neutrality point (CNP) and superconductivity (SC) regions are highlighted. c. Hysteresis of R_{xx} versus applied top gate voltage V_{TG} depending on gate sweep direction.

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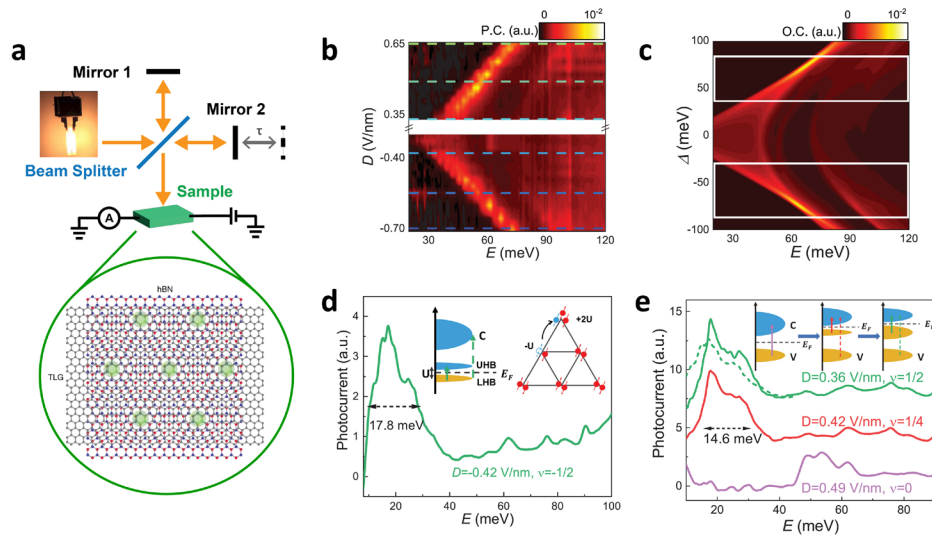
Spectroscopy Signatures of Electron Correlations in a Trilayer Graphene/hBN Moiré Superlattice

J. Yang, G. Chen, T. Han, Q. Zhang, Y.-H. Zhang, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, Z. Shi, T. Senthil, Y. Zhang, F. Wang, L. Ju

Sponsorship: Center for Integrated Quantum Materials, NSF Grant No. DMR1231319

Two-dimensional (2D) materials, since their discovery in 2005, have attracted great interest among scientists. Specifically, the idea of creating a moiré superlattice by stacking 2D materials vertically allows people to combine the properties of various 2D materials and to manipulate the electrons via electrostatic gating, making it a great platform to study exotic physics phenomena. Among them, rhombohedral- (ABC) stacked trilayer graphene/hexagonal boron nitride moiré superlattice (ABC-TLG/hBN) has been found to be a promising system since it has large tunability and is free of twisting angle disorders. People have observed correlated insulator, superconductivity, and topological state in transport studies. In contrast, the spectroscopic study of ABC-TLG/hBN is still absent, mainly due to the size-mismatch between device dimension (typically $\sim 10 \mu\text{m}$) and wavelength (30-100 μm), which makes the signal-to-noise ratio very small. In this work, we report spectroscopy measurements of dual-gated ABC-

TLG/hBN using a special Fourier-transform infrared (FTIR) photocurrent spectroscopy method. We observe a strong optical transition between moiré minibands that narrow continuously as we increase the band-gap via electrostatic gating, indicating a reduction of the single-particle bandwidth. At half-filling of the valence flat band, a broad absorption peak emerges at ~ 18 milli-electron volts, representing the direct optical excitation across an emerging Mott gap. Similar photocurrent spectra are obtained in two other emerging correlated insulating states at quarter- and half-filling of the first conduction band at finite magnetic field. Our work is the very first spectroscopic study of an ABC-TLG/hBN system and provides key parameters of the Hubbard model to understand electron correlation. Further, this technique can be applied to many other graphene-based moiré systems that also lack spectroscopic studies and provides more insights into many exotic physics phenomena.



▲ Figure 1: (a) Illustration of FTIR photocurrent spectroscopy and ABC-TLG/hBN superlattice. (b) Experimental results and (c) calculations of charge-neutral point photocurrent spectra at different displacement fields, where the narrowing of inter-band transition peaks indicates suppressing of bandwidth W . (d) First direct measurement of on-site interaction U via photocurrent spectra at half-filling. (e) Spectra at correlation gaps at quarter- and half-filling at finite magnetic field.

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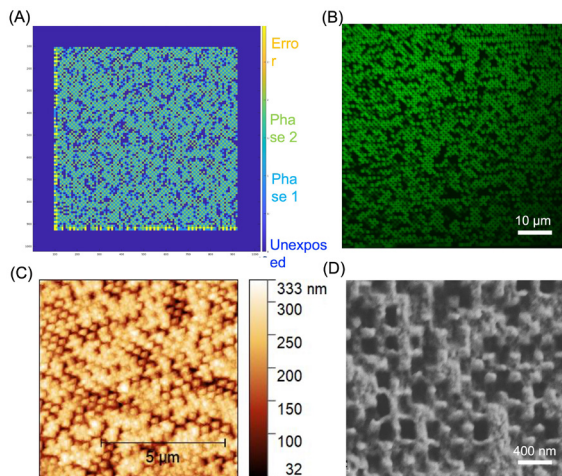
Nanofabrication of Metasurface Structures for Holography, Using Laser Ablation-based Implosion Fabrication

G. Yang, Q. Yang, T. Nambara, Y. Kunai, Y. Salamin, M. Soljačić, P. T. C. So, E. S. Boyden
Sponsorship: Fujikura Corp.

Nanofabrication techniques have advanced the development of precise optical components for various fields, including imaging, sensing, and communication. We present a laser ablation-based implosion fabrication technique for constructing metasurface structures with nanoscale precision for holography and diffractive optical element (DOE) applications. Our process utilizes a two-photon laser to activate photosensitizers, breaking polymer chains to create topological nanostructures and metasurfaces with varying height gradients. We designed a phase mask metasurface transmitting light with varying thicknesses, generating phase changes with three quantization levels. The implosion fabrication method built intricate, free-form three-dimensional (3D) architectures with nanometer precision. We obtained multiple layers of diffractive structures with varying refractive indices (RI) by adjusting the thickness of patterned structures. Comparing the designed mask and fluorescent image of the patterned region (Figure 1a, b) showed high correlation in vox-

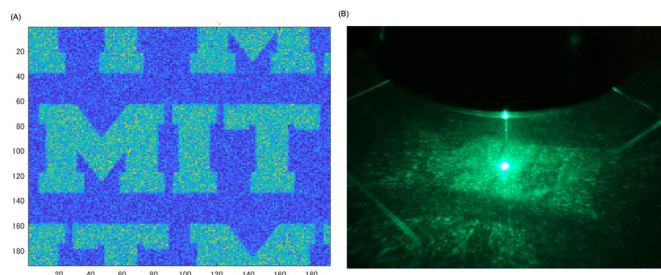
el unit distribution and precision. Scanning electron microscopy and atomic force microscopy (AFM) confirmed successful construction of metasurface structures with varying height gradients (Figure 1c, d), each unit with a feature size of 200-250 nm and height gradient difference of 100 nm.

Using coherent light, the metasurface generated a far-field diffraction pattern reconstructing the MIT logo with high contrast, resolution, and minimal aberrations (Figure 2). The fabricated metasurface structures can produce far-field diffraction hologram effects when illuminated with a monochromatic light source. Our innovative laser ablation-based implosion fabrication technique has the potential to create personalized DOEs with nanoscale 3D structural accuracy and full-color 3D visual effects. This breakthrough holds tremendous promise for revolutionizing the field of holography and creating opportunities in the development of advanced optical devices.



◀ Figure 1: A) The designed mask of different levels; B) Fluorescent image of a plane of the patterned layer; C) AFM image and D) AFM image of the metasurface.

▼ Figure 2: A) The simulated image of reconstructed MIT logo; B) The far-field diffraction effect, displayed as the MIT logo.



FURTHER READING

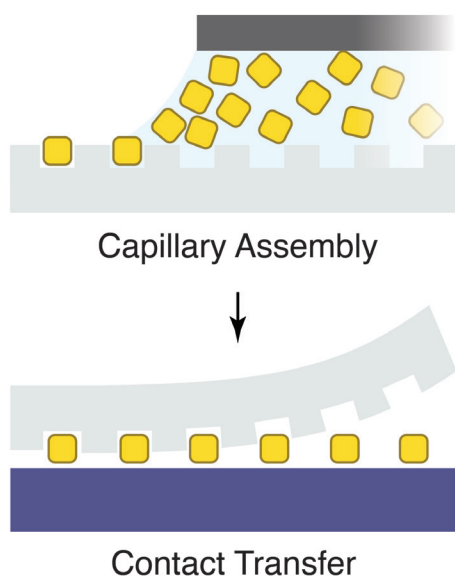
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Contact Printed Nanoparticles as Building Blocks for Active Nanodevices

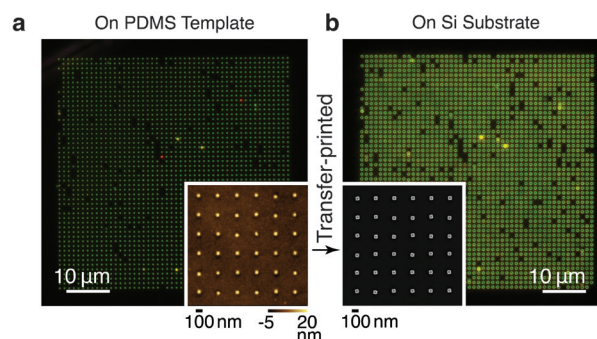
W. Zhu, P. F. Satterthwaite, P. Jastrzebska-Perfect, R. Brenes, F. Niroui
Sponsorship: NSF Award CMMI-2135846

Nanoparticles, efficiently formed through bottom-up synthesis with diverse composition, structure, and functionalities, exhibit unique properties compared to their top-down fabricated counterparts. Leveraging these properties requires deterministic patterning of nanoparticles with the desired spatial order. Here, we present a versatile, scalable, and pristine approach for deterministic integration of nanoparticles into active structures and devices with single-particle resolution. Our approach, named nanoparticle contact printing, first spatially assembles nanoparticles into a topographical template, and then prints them onto diverse surfaces and interfaces. By engineering interfacial interactions, our approach promotes high-yield nanoparticle transfer without requiring solvents,

surface treatments, or sacrificial layers as is conventionally needed. With our approach, surfaces remain pristine and are accessible for integration into functional structures. We demonstrate this through a particle-on-mirror plasmonic cavity model system, where >2000 gold nanocubes are deterministically patterned onto template-stripped gold with sub-50 nm positional accuracy and minimized inter-structural variability. We further highlight the integration opportunities offered by our technique by fabricating arrays of emitter-coupled nanocavities. In addition, we use this platform to demonstrate mechanically active molecular junctions with sub-nm tunability as building blocks of miniaturized nanoelectromechanical sensors and actuators.



▲ Figure 1: Schematic overview of nanoparticle contact printing: nanoparticles are assembled onto topographical template through capillary assembly, followed by contact transfer onto receiving substrate.



▲ Figure 2: (a) Dark-field image of assembled 50 x 50 array of individual gold nanocubes, with AFM image inset highlighting assembled nanocubes. (b) Dark-field image of nanocube array after contact transfer onto Si substrate, with SEM image inset showing transferred nanocubes.

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