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LOW DIMENSIONAL CARBON MATERIALS FOR NANOOPTICS AND NANOPLASMONICS

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Final Report**

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This final report describes the development and application of the new synthesis, fabrication, characterization techniques our group has developed for studying fundamental optical and optoelectronic properties in two-dimensional (2D) materials, including graphene, hexagonal boron nitride (h-BN) and molybdenum disulfide, as funded by the AFOSR grant (FA9550-10-1-0410). We have investigated the fundamental properties such as electron cooling and the effect of grain boundary in single layer graphene (SLG). We demonstrated the "patterned regrowth" technique to build spatially-precise 2D circuit out of graphene and hBN. We discovered and studied previously-unseen structures such as the strain soliton in bilayer graphene using dark- field transmission electron microscopy (DF-TEM). Being able to identify 2D multilayer materials with complicated stacking structures enables us to study their unique optical properties, such as excitonic effects in the interlayer excitation in tBLG. Finally, the technique we have developed can be directly applied to study other 2D materials such as molybdenum disulfide and 2D glasses. Novel properties in these materials open up new avenues for studying old and new physics including glass phase transition and valley Hall effect.

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Executive Summary

Controlling the propagation of light, and localizing the energy it carries, is one of the most important scientific challenges of the 21st century. While nanoscale materials, 2D materials in particular, provide exciting new approaches for this purpose, much key information regarding their fundamental optical properties is currently unknown. In addition, there are significant materials issues that need to be addressed in order to produce device geometries that are contamination-free and fully controlled. This final report describes the development and application of the new synthesis, fabrication, characterization techniques our group has developed for studying fundamental optical and optoelectronic properties in two-dimensional (2D) materials, including graphene, hexagonal boron nitride (*h*-BN) and molybdenum disulfide, as funded by the AFOSR grant (FA9550-10-1-0410). We have investigated the fundamental properties such as electron cooling and the effect of grain boundary in single layer graphene (SLG). We demonstrated the “patterned regrowth” technique to build spatially-precise 2D circuit out of graphene and hBN. We discovered and studied previously-unseen structures such as the strain soliton in bilayer graphene using dark- field transmission electron microscopy (DF-TEM). Being able to identify 2D multilayer materials with complicated stacking structures enables us to study their unique optical properties, such as excitonic effects in the interlayer excitation in tBLG. Finally, the technique we have developed can be directly applied to study other 2D materials such as molybdenum disulfide and 2D glasses. Novel properties in these materials open up new avenues for studying old and new physics including glass phase transition and valley Hall effect.

TECHNICAL DESCRIPTION OF WORK

A. Introduction

Over the past decade, two-dimensional (2D) materials, such as graphene, hexagonal boron nitride (*h*-BN) and transition metal dichalcogenides (TMDs), have emerged as promising candidates for novel optoelectronic devices and applications as well as next-generation atomically thin technology. However, the promise of 2D materials-based technology heavily relies on our ability to produce and characterize large-scale 2D films with desired structure and properties. In order to fully utilize the power of 2D materials with various structures, one must first understand the effects of their structural properties – grain boundary, crystal structure and orientation - on their electronic and optical properties. Under this AFOSR grant, our group has developed new synthesis, fabrication, and optical and transmission electron microscopy (TEM) characterization methods to achieve this understanding. Here we describe several methods that are newly developed by our group to efficiently characterize 2D materials, discover new physics for future application and demonstrate fabrication for atomically thin circuit and novel optoelectronics.

In order to achieve high performance devices, we started with studying the electronic properties in monolayer graphene. Our transient photocurrent¹ and absorption microscopy² reveals the mechanism for electron cooling that has long been controversial. Furthermore, we developed direct device fabrication on TEM window and demonstrated the first direct electrical measurements of individual grain boundaries in chemical vapor deposition (CVD) grown graphene with full knowledge of their locations³. A prototype of 2D circuit is also demonstrated through our “patterned regrowth” method enabling next-generation 2D electronics⁴.

The vertical stacking and lateral structure produced using 2D materials introduce additional structural variations in these crystalline films, which can significantly affect their electronic and mechanical properties, producing entirely new physical phenomena. We developed TEM, Raman, and optical techniques that allow direct structural identification of stacking structures and interlayer interactions⁵. We discovered and characterized new strain soliton states in bilayer graphene that are formed at the boundaries between twin domains⁶. We also studied the optical properties of twisted bilayer graphene (tBLG) and bilayer hBN^{7,8}. Our data confirms the presence of a new kind of dark excitons in tBLG^{9,10} that cannot be seen by normal 1-photon microscopy but can be probed experimentally with 2-photon measurements¹¹.

All the techniques we have developed for graphene and hBN also pave the way to study other

2D materials such as transition metal dichalcogenides (TMDs) and 2D glasses. We discovered the valley Hall effect (VHE) in MoS₂ transistors, where electrons from each energy valley in its band structure spontaneously exhibit a finite Hall effect in the absence of a magnetic field¹². We also observed, using atomic resolution TEM, the structure of 2D glass and its transformation¹³. These results open up the gateway to a deeper understanding of 2D materials and to the basis for future technology such as atomically thin integrated circuitry and valleytronics.

B. Supercollision cooling in graphene electronic relaxation^{1,2}

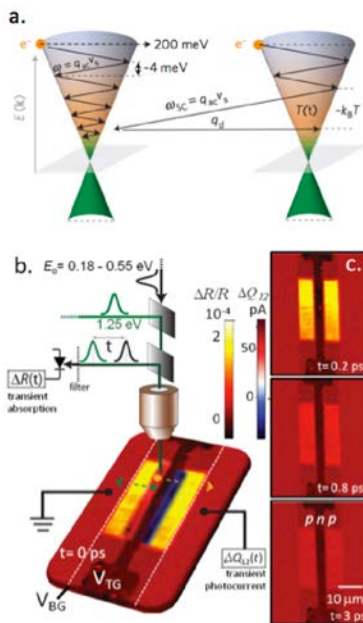


Figure 1. (a) Two proposed cooling mechanisms. Left: hot optical phonon cooling; Right: defects assisted super-collision cooling.

(b) Transient photo-current and absorption (TPC/TA) measurement set-up. (c) Ultra fast movie for electron-relaxation

The cooling of hot electrons in graphene is the critical process underlying the operation of exciting new graphene-based devices, but the nature of this cooling is controversial (fig. 1(a)). We studied hot-electron cooling near the Fermi level by using graphene as a novel photothermal thermometer (fig. 1(b)) that measures the electron temperature as it cools dynamically. These

results are in excellent quantitative agreement with disorder-enhanced supercollision (SC) cooling mechanism over a wide range of electronic (15 to ~3,000K) and lattice (10–295 K) temperatures. We further studied the SC cooling mechanism through transient absorption (TA) microscopy as the optical counterpart (fig. 1(b) and (c)). It shows that disorder-assisted acoustic-phonon SC best describes the rate-limiting relaxation step in graphene electron and hole relaxation as well. Moreover, it is shown that the electron-cooling rate in substrate-supported graphene is twice faster than its suspended counterpart, as one would expect based on the SC model. The confirmation of the cooling mechanism provides a reliable model to determine the electronic temperature in graphene, which is of central importance in designing graphene terahertz plasmonic devices, photodetectors and bolometers.

C. Optimizing Electrical Properties of Polycrystalline Graphene³

When graphene is grown by chemical vapor deposition (CVD), it produces many grain boundaries (GBs) in contrast to exfoliated graphene that is single crystalline. These grain boundaries (GBs) could potentially serve as a scattering center for charge carriers and therefore degrade the electrical and optoelectronic performance (including THz). To determine the effects of GBs, we have directly measured the electrical properties of GBs with simultaneous knowledge of their locations and structures for the first time (fig 2). After the

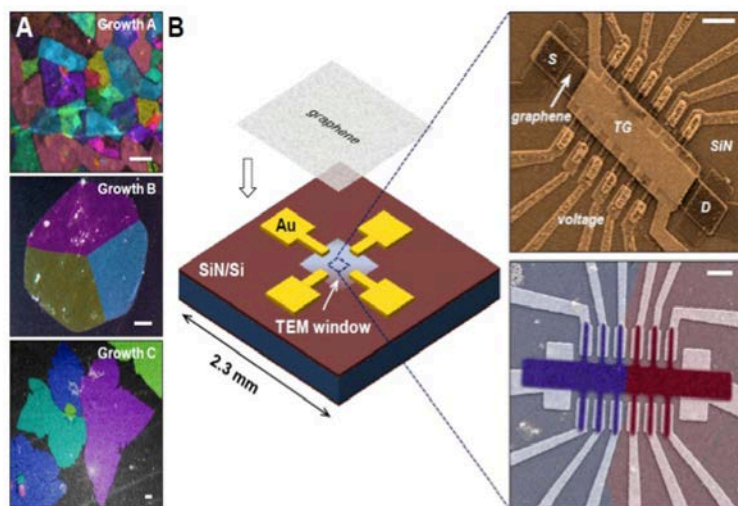


Figure 2. a, DF-TEM images of CVD graphene grown under different conditions. b, Schematic of TEM chip compatible with e-beam lithography and electrical measurements. Top: SEM image of graphene bar device. Bottom: overlaid SEM and DF-TEM image of a device crossing a single GB. All scale bars - 1 μm .

grain structure is characterized via DF-TEM (fig. 2(a)), we use e-beam lithography to fabricate a device consisting of an individual grain boundary (fig. 2(b)) directly onto the TEM window. Unexpectedly, we found out that the electrical conductance can be improved by one order of magnitude when GBs has better inter-domain connectivity. Our study suggests that polycrystalline graphene with good stitching may allow for

uniformly high electrical performance rivaling that of exfoliated samples, and we demonstrated this using optimized growth conditions and device geometry.

D. Graphene and Boron Nitride Lateral Heterostructures⁴

Precise spatial control over the electrical properties of thin films in large scale is the key capability enabling the production of modern integrated circuitry. Large scale, high performance atomic membranes can now be made via CVD, but controlled fabrication of lateral heterostructures has not been reported before. We reported the first spatially controlled synthesis of lateral junctions between electrically conductive graphene and insulating h-BN using a versatile and scalable process, “patterned regrowth”. (fig. 3) We demonstrated that our resulting films form mechanically continuous sheets. Conductance measurements confirmed laterally insulating behavior for h-BN regions, while the electrical behavior of graphene sheets maintains excellent properties, with low sheet resistances and high carrier mobilities. In addition, we fabricated vertical heterostructures of these patterned films using

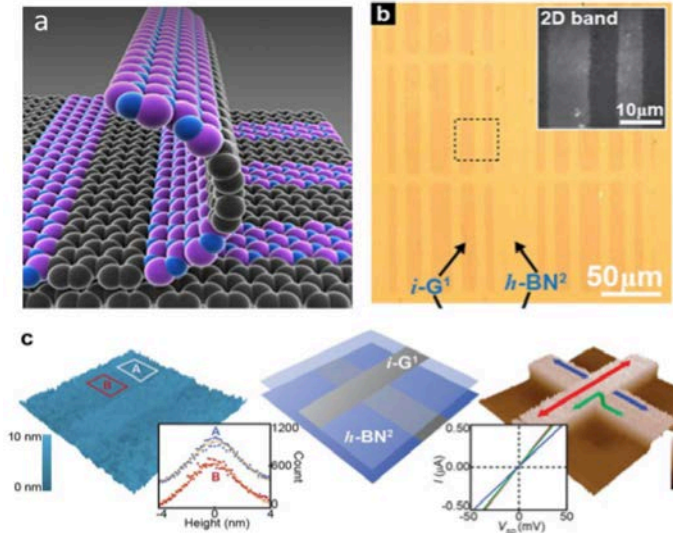


Figure 3. **a**, Illustration of graphene/*h*BN heterostructure. **b**, Optical image of transferred graphene(dark)/*h*BN heterostructure on SiO₂/Si substrate. Inset: Raman 2D band image. **c**, Stacked heterostructure films show a flat surface (left-AFM topography) and well defined conductive regions (right- EFM).

subsequent transfers, which showed that they have flat surfaces even without further mechanical or chemical polishing and that graphene strips in different layers can make good electrical interlayer contacts. (fig. 3(c)) Our results represent an important step towards developing atomically thin integrated circuitry, enabling the fabrication of electrically isolated active and passive elements embedded in continuous, one atom thick sheets.

E. Stacking order and formation of strain soliton in multilayer graphene^{5,6}

Multilayer atomic membranes with complicated stacking orders have often been observed in CVD synthesis along with single layer. They exhibit different properties from monolayer and

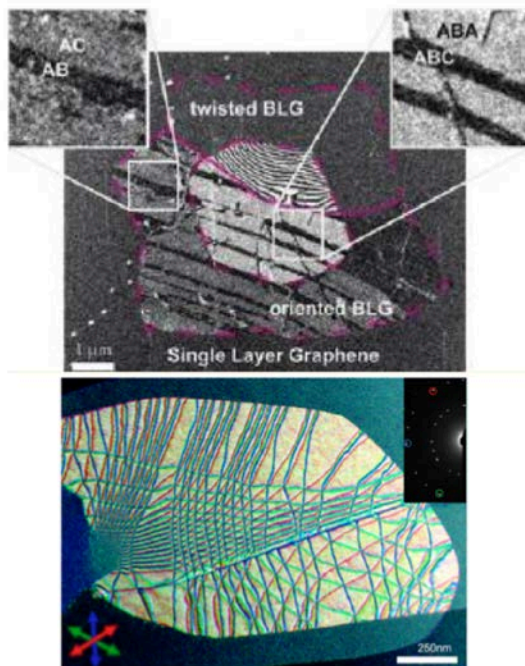


Figure 4. (top) dark field TEM image shows stacking structure and strains. (bottom) Color-coded darkfield TEM images of strain solitons in BLG. Each color (red, blue, green) corresponds to the displacement vector of the soliton.

can be useful in novel devices. However, developing controlled growth of these structures requires efficient large-scale characterization. We used dark-field transmission electron microscopy with different tilting angle for rapid and accurate determination of key structural parameters (twist angle, stacking order, and interlayer spacing), as shown in fig. 4(top). With this technique, we find that Bernal-stacked bilayer is the preferred structure for bilayer graphene (BLG) in our CVD growth, accounting for 70% of the BLG region. Moreover, we investigated the stacking order in Bernal stacked BLG. In Bernal stacked BLG, more than one structural ground states (AB or BA) exist and they are connected by a topologically protected defect lines. Using high resolution and DF-TEM, we showed that these boundaries are

strain solitons, whose properties are determined by interlayer interaction and strain, minimizing the total energy. Significantly, these soliton lines exhibit characteristic atomic configurations each associated with an interlayer displacement vector which can be read out using DF-TEM imaging (fig. 4(b)). We observed similar strain soliton lines in *h*-BN bilayers as well and their presence in BLG introduces novel ways to modify the electrical, optical, and mechanical properties of these bilayer systems.

F. Structure dependent optical properties of bilayer graphene and *h*-BN^{7,8}

Multilayer 2D materials possess various complicated stacking orders and interlayer structures not found in their monolayer counterparts, which bring in many new properties. As a basis for such complicated multilayer structures, we start with the structure and optical properties of bilayer atomic membranes of graphene and *h*-BN.

Twisted bilayer graphene (tBLG), where two stacked graphene layers are rotated

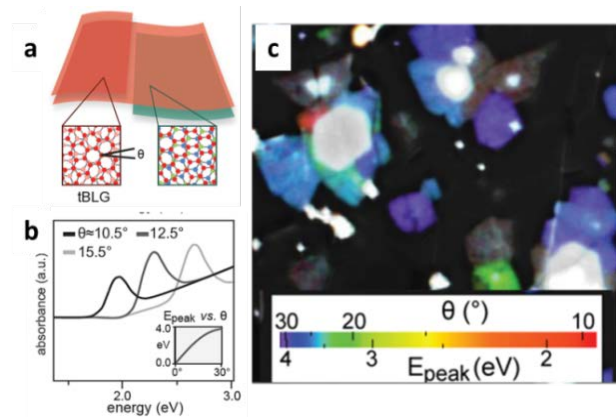


Figure 5. a, Schematic of tBLG. b, Plot of theoretical tBLG peak energy (E_{peak}) vs. θ . c, False color image of θ in tBLG domains, determined optically for a $\sim 1500 \mu\text{m}^2$ area.

by a relative angle θ (fig. 5(a)), exhibits unique interlayer optical behaviors dependent on θ . We performed quantitative optical characterization of tBLG with known θ up to 30° by combining TEM and broadband DUV-Vis-NIR hyperspectral imaging.⁸ We observed, as functions of θ , enhanced optical absorption at the energies where the single layer graphene bands hybridize. This establishes a structure (θ)-property relationship in tBLG, enabling purely optical measurements of this angle on arbitrary substrates for the first time (fig. 5(c)).

Due to its lower symmetry, more variations in stacked structures exist in *h*-BN bilayers. Our optical second harmonic generation (SHG) measurements combined with DF-TEM confirmed the correlation between stacking orders and their optical properties. As SHG is generated in a non-centrosymmetric material (system without inversion symmetry), a strong SHG signal was observed only in regions corresponding to non-centrosymmetric *h*-BN (AB stacking). Significantly, the efficiency of SHG observed in AB *h*-BN bilayers is much stronger than that of naturally abundant AA' stacked *h*-BN. Our study suggests that the optical properties of stacked atomic membranes can be engineered by modulating their stacking orders.

G. Excitonic effects in the optical response of tBLG and its hidden dark excitons^{9,10,11}

Strong Coulomb interaction has been shown important in single layer two-dimensional materials, but the role it plays in multilayer atomic membranes remains unexplored. Our DUV-Vis-NIR hyperspectral imaging provides us a quantitative probe into the full optical

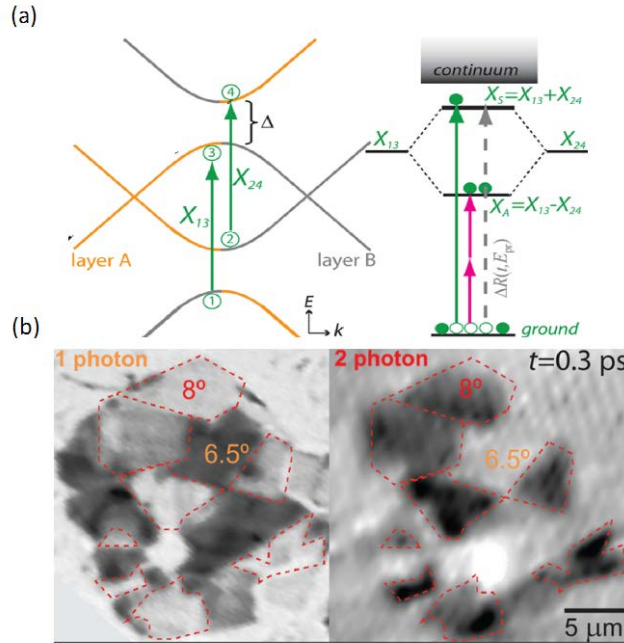


Figure 6. (a) Left: tBLG band structure shows interlayer excitonic transition, X_{13} and X_{24} . Others are forbidden by selection rule. Right: The degenerate X_{13} and X_{24} rehybridize to form 1-photon allowed X_S state and 2-photon allowed X_A state. (b) Left: 1-photon excite X_S in 6.5° domain. Right: 2-photon excite X_A in 8° domain.

spectra of twisted bilayer graphene.

This enables us to study the interlayer exciton optically through linear absorption spectroscopy. Our result suggested that the excitonic effects can largely alter the lineshape of the spectrum in tBLG. Furthermore, our theoretical calculation predicts the existence of an

previously-unseen, one-photon forbidden dark exciton state X_A (fig. 6(a), right), besides the bright exciton X_S . We utilized

two-photon excitation to break the selection rule and successfully found the dark state at $\sim 0.37\text{eV}$ (right, fig. 6(b), for 8° tBLG). Moreover, this dark state is

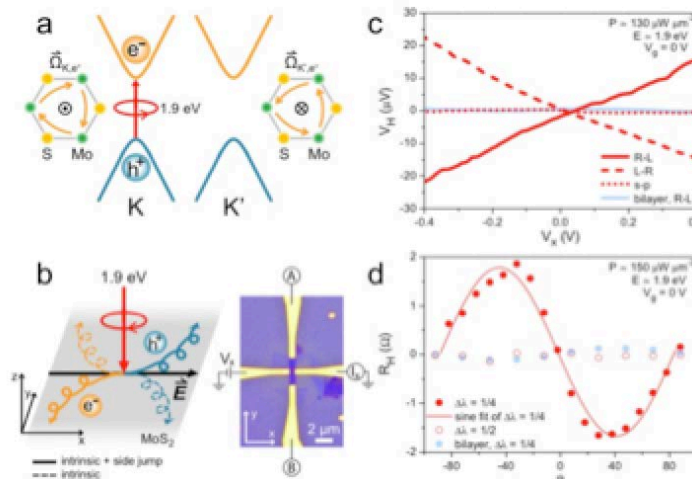
disconnected from the continuum states in graphene. Therefore, it does not have

the Fano resonance line shape that is present in single layer graphene excitons producing a

lifetime as long as 66 ± 4 ps, which is surprisingly long for a metallic system.

H. Valley Hall effect in monolayer MoS2 transistors¹²

Transitional metal dichalcogenides (TMDs) appears as another low dimensional system



that offers a playground for new

Figure 7. (a) The valley-dependent optical selection rules and the electrons at the K and K' valleys that experience opposite effective magnetic fields. (b) Schematic of a photo induced Hall effect driven by a net valley polarization, and an image of the Hall bar device. (c) The source-drain bias (V_x) dependence of the Hall voltage (V_H) for right-handed (red, solid), left-handed (red, dashed) and linearly polarized (red, dotted) excitations. (d) Dependence of the anomalous Hall resistance on the excitation ellipticity.

nanooptics. For example, electrons in monolayer (ML) MoS₂ display a novel electrical behavior associated with the extra valley degree of freedom in addition to charge and spin. Due to the unique crystalline structure of ML MoS₂, electrons from each valley spontaneously exhibit a finite Hall effect in the absence of a magnetic field, a phenomenon called the valley Hall effect (VHE) (fig. 7(a) and (b)). To measure the VHE, we shone circularly polarized light onto a MoS₂ Hall bar device to create a population imbalance between the two valleys to create Hall effect as shown in fig. 7(b). This VHE is active only under circularly polarized light (fig. 7(c)) with its sign and magnitude sensitive to the light polarization (fig. 7(d)). This new DOF has the potential to be used as an information carrier in next-generation electronics and valleytronics.

I. Atomic resolution imaging of structure and rearrangements in 2D glass¹³

By combining aberration corrected TEM with atom-by-atom spectroscopy, we reconstructed the full structure of the 2D glass supported by a graphene window and

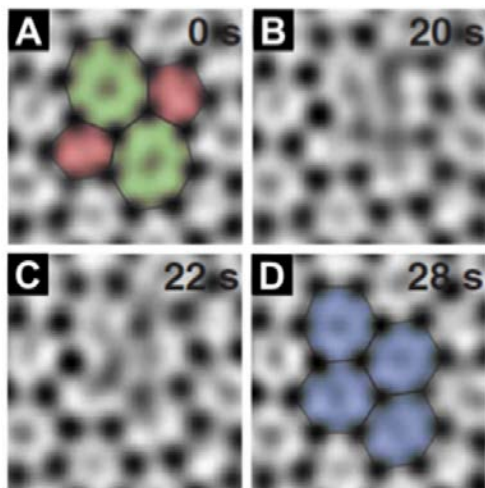


Figure 8. TEM images showing a ring rearrangement that transforms a 5-7-5-7 cluster into a 6-6-6-6 cluster.

identified it as a bi-tetrahedral layer of SiO₂ only 3 atoms thick. Our atomic resolution images, which bear a striking resemblance to Zachariasen's original cartoon models of glasses drawn in 1932, introduce powerful new methods to test long-standing theoretical predictions of glass structure and dynamics against experimental data. For instance, atoms in this disordered 2D solid were imaged in response to local strain, whose motions were then analyzed using ring statistics and pair distribution functions for short-, medium-, and long-range order. We also used the

electron beam to excite atomic rearrangements in 2D glass (fig. 8), producing rich and beautiful videos of glass bending and breaking, as well as the exchange of atoms at a solid/liquid interface. Detailed analysis of these videos reveals a complex dance of elastic and plastic deformations, phase transitions, and their interplay³. These examples illustrate the wide-ranging and fundamental materials physics that can now be studied at atomic-resolution via transmission electron microscopy of 2D glasses.

J. Conclusion

In summary, we developed new synthesis, fabrication and characterization methods for 2D materials. These techniques allow for characterization of structure related electrical, optical and optoelectronic properties in the nano-scale. These new characterization methods assisted in creating specialized forms of 2D films, specifically graphene-*h*BN atomic integrated circuit. New optical characterization methods allowed direct structural identification of stacking structures realizing new properties for novel optoelectronics. This work also paves the way for fabrication and characterization of a host of 2D materials, revealing new physical phenomenon such as valley Hall effect and new experimental platforms for studying glass transformation.

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FA9550-10-1-0410

Principal Investigator Name

The full name of the principal investigator on the grant or contract.

Jiwoong Park

Program Manager

The AFOSR Program Manager currently assigned to the award

Dr. Harold Weinstock

Reporting Period Start Date

08/01/2010

Reporting Period End Date

07/31/2015

Abstract

Controlling the propagation of light, and localizing the energy it carries, is one of the most important scientific challenges of the 21st century. While nanoscale materials, 2D materials in particular, provide exciting new approaches for this purpose, much key information regarding their fundamental optical properties is currently unknown. In addition, there are significant materials issues that need to be addressed in order to produce device geometries that are contamination-free and fully controlled. This final report describes the development and application of the new synthesis, fabrication, characterization techniques our group has developed for studying fundamental optical and optoelectronic properties in two-dimensional (2D) materials, including graphene, hexagonal boron nitride (h-BN) and molybdenum disulfide, as funded by the AFOSR grant (FA9550-10-1-0410). We have investigated the fundamental properties such as electron cooling and the effect of grain boundary in single layer graphene (SLG). We demonstrated the "patterned regrowth" technique to build spatially-precise 2D circuit out of graphene and hBN. We discovered and studied previously-unseen structures such as the strain soliton in bilayer graphene using dark-field transmission electron microscopy (DF-TEM). Being able to identify 2D multilayer materials with complicated stacking structures enables us to study their unique optical properties, such as excitonic effects in the interlayer excitation in tBLG. Finally, the technique we have developed can be directly applied to study other 2D materials such as molybdenum disulfide and 2D glasses. Novel properties in these

materials open up new avenues for studying old and new physics including glass phase transition and valley Hall effect.

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Archival Publications (published) during reporting period:

1. M. W. Graham, S. Shi, D. C. Ralph, J. Park and P. L. McEuen, "Photocurrent Measurements of Supercollision Cooling in Graphene", *Nature Physics* 9, 103-108 (2013)
2. M. W. Graham, S. Shi, Z. Wang, D. C. Ralph, J. Park, and P. L. McEuen, "Transient Absorption and Photocurrent Microscopy Show Hot Electron Supercollisions Describe the Rate-Limiting Relaxation Step in Graphene," *Nano Letters* 13, 5497-5502 (2013)
3. A. W. Tsen, L. Brown, M. P. Levendorf, F. Ghahari, P. Y. Huang, C. S. Ruiz-Vargas, R. W. Havener, D. A. Muller, P. Kim, and J. Park, "Tailoring Electrical Transport across Grain Boundaries in Polycrystalline Graphene," *Science* 336, 1143-1146 (2012)
4. M. P. Levendorf*, C. J. Kim*, L. Brown, P. Y. Huang, R. W. Havener, D. A. Muller, and J. Park, "Graphene and Boron Nitride Lateral Heterostructures for Atomically Thin Circuitry," *Nature* 488, 627-632 (2012)
5. L. Brown*, R. Hovden*, P. Huang, M. Wojcik, D. A. Muller, and J. Park, "Twinning and Twisting of Tri- and Bi-layer Graphene," *Nano Letters* 12, 1609-1615 (2012)
6. J. S. Alden, A. W. Tsen, P. Y. Huang, R. Hovden, L. Brown, J. Park, D. A. Muller, and P. L. McEuen, "Strain Solitons and Topological Defects in Bilayer Graphene," *PNAS* 110, 11256-11260 (2013)
7. R. W. Havener, C.-J. Kim, L. Brown, J. W. Kevek, J. D. Sleppy, P. L. McEuen, and J. Park, "Hyperspectral imaging of structure and composition in atomically thin heterostructures," *Nano letters* 13, 3942-3946 (2013)
8. C. J. Kim, L. Brown, M. W. Graham, R. W. Havener, R. Hovden, P. L. McEuen, D. A. Muller, and J. Park, "Stacking Order Dependent Second Harmonic Generation and Topological Defects in h-BN Bilayers," *Nano Letters* 13, 5660-5665 (2013)
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10. Y. Liang, R. Soklaski, S. Huang, M. W. Graham, R. W. Havener, J. Park and L. Yang, "Strongly bound excitons in gapless two-dimensional structures," *Physical Review B* 90, 115418 (2014)
11. H. Patel, R. W. Havener, L. Brown, Y. Liang, L. Yang, J. Park, and M. W. Graham, "Tunable optical excitations in twisted bilayer graphene form strongly bound excitons," *Nano Letters* 15, 5932-5937 (2015)
12. K. F. Mak, K. L. McGill, J. Park and P. L. McEuen, "The valley Hall effect in MoS2 transistors," *Science* 344, 1489-1492 (2014)
13. P. Y. Huang, S. Kurasch, J. S. Alden, A. Shekhawat, A. A. Alemi, P. L. McEuen, J. P. Sethna, U. Kaiser, and D. A. Muller, "Imaging atomic rearrangements in two-dimensional silica glass: watching silica's dance", *Science*, 342, 224-227 (2013)

Changes in research objectives (if any):

Change in AFOSR Program Manager, if any:

Extensions granted or milestones slipped, if any:

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, \$K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

Report Document

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Appendix Documents

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