



ARL-TR-9711 • JUNE 2023



Multiscale Mathematical Modeling and Design for the Realization of Novel 2-D Functional Materials (Summary Technical Report, May 2014–Mar 2022)

by Joseph Myers and Pani Varanasi

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Joseph Myers and Pani Varanasi
DEVCOM Army Research Laboratory

REPORT DOCUMENTATION PAGE

1. REPORT DATE		2. REPORT TYPE		3. DATES COVERED	
June 2023		Summary Technical Report		START DATE 5/01/2014	END DATE 5/31/2022
4. TITLE AND SUBTITLE Multiscale Mathematical Modeling and Design for the Realization of Novel 2-D Functional Materials (Summary Technical Report, May 2014–Mar 2022)					
5a. CONTRACT NUMBER		5b. GRANT NUMBER		5c. PROGRAM ELEMENT NUMBER	
5d. PROJECT NUMBER W911NF-14-1-0247		5e. TASK NUMBER		5f. WORK UNIT NUMBER	
6. AUTHOR(S) Joseph Myers and Pani Varanasi					
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) DEVCOM Army Research Laboratory ATTN: FCDD-RLR-EW Research Triangle Park, NC 27709				8. PERFORMING ORGANIZATION REPORT NUMBER ARL-TR-9711	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) US Army Research Office 800 Park Offices Drive Research Triangle, NC 27709-2211		10. SPONSOR/MONITOR'S ACRONYM(S)		11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION/AVAILABILITY STATEMENT DISTRIBUTION STATEMENT A. Approved for public release: distribution unlimited.					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT Summary Technical Report for the FY16 Multi-University Research Initiative effort titled “Multiscale Mathematical Modeling and Design Realization of Novel 2-D Functional Materials” led by Professor Mitch Luskin at the University of Minnesota, which was active from 28 May 2014 through 15 March 2022. The objective of the effort has been to further our fundamental understanding of van der Waals bonded, layered 2-D heterostructures, using novel strongly linked multiscale computational methods and guided by concurrent experiments to both inspire the theory and benefit from its predictions in order to design flexible electronics and more sensitive detection capability.					
15. SUBJECT TERMS 2-D materials, weakly interacting multilayers, superconductivity in 2-D systems, incommensurate heterostructures, Military Information Sciences, Photonics, Electronics, and Quantum Sciences, Sciences of Extreme Materials, STR					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	18. NUMBER OF PAGES 16	
a. REPORT UNCLASSIFIED	b. ABSTRACT UNCLASSIFIED	c. THIS PAGE UNCLASSIFIED			
19a. NAME OF RESPONSIBLE PERSON Joseph Myers				19b. PHONE NUMBER (Include area code) (919) 549-4245	

STANDARD FORM 298 (REV. 5/2020)

Prescribed by ANSI Std. Z39.18

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1. Executive Summary

The FY14 Multidisciplinary University Research Initiative (MURI) topic titled “Multiscale Mathematical Modeling and Design Realization of Novel 2-D Functional Materials” led by Professor Mitch Luskin at University of Minnesota was active from 28 May 2014 through 15 March 2022. The objective of the effort has been to further our fundamental understanding of the geometric deformations and resulting electronic phenomena that occur in layered sheets of flexible 2-D electronic materials when twisted in relation to adjacent sheets. Their key results included new multiscale models for electronic phenomena in rotated 2-D sheets and predicted the existence of superconductivity in some rotated sheets, which was soon after confirmed by experiments. Future studies will use these new tools to search for new phenomena that can enable new capabilities in areas such as single photon detection (SPD) for ultra-sensitive detection devices.

This effort invented the field of Twistronics, a term and an area that did not exist before this effort was initiated by the MURI team. It brought together mathematicians, material scientists, computational modelers, and atomic physicists to investigate the mathematics, geometry, flexible connectivity, and solid-state physics of these newly engineered 2-D structures as well as the models to predict and engineer new structures for new phenomena. The MURI focused on developing mathematical tools and related computational methods for modeling the properties of 2-D sheets, which distorted each other through electronic interactions when brought together. These models were developed in close collaboration with experimental challenges and needs. These systems became the subject of intense research efforts a few years ago, starting with the discovery of graphene, followed by the realization of a much broader class of 2-D layered structures with unique semiconducting or insulating properties, depending on the material. More recently, the discovery of superconductivity and other correlated phases in twisted moiré systems with flat bands sparked our efforts to investigate new moiré superlattice systems of interest and to derive and simulate models that accurately describe their electronic properties.

The novel aspect of this class of materials is the true 2-D character of each layer, all of which interact via weak chemical forces between adjacent layers. The specific goal of this MURI has been to provide the mathematical and computational underpinnings for accurately modeling these materials to guide experimental efforts toward device realization.

The MURI project spent significant effort on developing theory and numerical methods to explore the electronic, mechanical, and diffraction properties of layered electronic sheets as the induced small structural distortions in each other. These

methods resulted in models that are much more accurate than methods based on periodicity (e.g., supercells, which are known to be a poor modeling assumption) and to instead enable exploring the physics of deformed electronic sheets that deform each other through electronic repulsions/attractions. The major experimental goals of this collaborative research included the characterization of 2-D van der Waals heterostructures by measuring structural, electrical, thermal, and mechanical properties to provide experimental inputs and confirmation for theoretical and mathematical modeling of the team.

The more recent discovery of superconductivity and other correlated phases in systems containing isolated regions of no net current drove the investigation of new twisted systems of interest and the derivation and simulation of models that could accurately describe their electronic properties. The larger goal in the applied physics and materials communities, and in our follow-on efforts in a newly started Small Business Technology Transfer Program (STTR) described in the following, is to compute useful ways we can combine these layers in interesting ways that will lead to devices with operational features much improved over traditional materials for optical and electronic applications.

2. Overview of Results

The team developed most of the mathematical tools and related computational methods for modeling the properties of 2-D weakly interacting multilayers in close collaboration with experimental challenges and needs.

Collaborative development focused on the interplay between theory, computation, and experiment for 2-D flexible sheets of electronic materials. The team developed methods for taking advantage of the relative sparsity of connections in the 2-D systems (which still involve twist and curvature embedded in a 2-D way in the 3-D space) to develop useful, relatively fast approximations of solutions to the governing equations. More specifically, a major focus has been calculating the relaxation paths and final positions of charged and neutral molecules due to the several different forces involved. These general methods were made possible by the team's development of methods and codes for constructing a library of mathematical operators for graphene-based materials and others that for the first time includes strain terms and accurate interlayer interactions. The team has used these new mathematical operators within a physics-based approach to compute electronic structure and transport in layers of 2-D electronic structures that do not (and cannot) stack well together.

This impossibility of relaxed stacking and the incommensurate nature of 2-D heterostructures makes it impossible to apply the standard tools of solid-state

physics, which are based on perfect crystalline order. Even for stackings of layers of the same material, the resulting structure can be incommensurate. The team's new approach parametrizes the degrees of freedom by the compact local configuration space rather than real space, thus bypassing the need for the standard supercell approximation and making possible the investigation of true incommensurate structures. The team has developed even more efficient methods by reformulating the mathematical tools to work in a more abstract but efficient space, rather than in physical space. This enables the modeling and computational methods to more accurately account for strain, buckling, electromagnetic fields, vacancies, and twisting in nonmatching 2-D curved sheets of sizes up to millions of atoms. The team has developed a number of methods for computing the states, conductivity, and selective optical activity. Depending on the system size and periodicity, they are now able to choose the best tool for the job at will. The computational methods developed in this MURI have already played a major role in the highly celebrated discovery of superconductivity at the "magic" rotation angle of 1.1° in bilayer twisted graphene. The team's new theory, fast numerical methods, and codes for the electronic structure, transport, mechanical relaxation, and diffraction of ill-matching 2-D sheets of electronics materials are now capable of exploring previously inaccessible physical regimes (viz. small twist angle, low temperature, etc.). These theoretical and computational methods have facilitated experimental investigations of controlled doping by lithium. This work is now helping enable progress toward the larger goal in the applied physics and materials communities of combining these layers in interesting ways that will lead to devices with operational features much improved over traditional materials for optical and electronic applications.

Success Summary: This work has developed new models and understanding of 2-D materials, which have enabled new understanding of the physics of 2-D multilayers and in turn has enabled applications in SPD. The project has created the theoretical foundations for understanding and creating miniaturized, flexible electronics that operate at extremely low power.

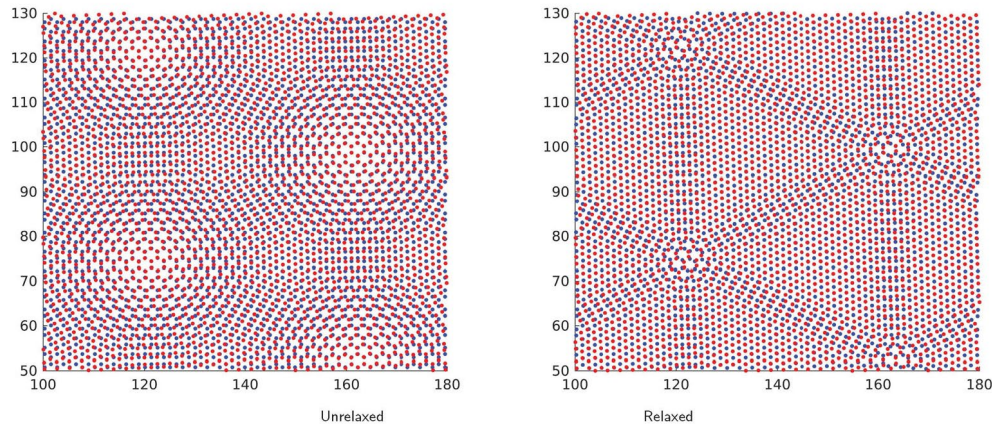
These findings have recently been published in a review article in *Nature–Materials* and are now being commercialized in an STTR sponsored by the Office of the Under Secretary of Defense for Research and Engineering [O/USD(R&E)]. The novel concepts developed in this MURI related to twistronics have also inspired new follow-on MURI topics to be developed within DOD, such as the FY21 Army Research Office MURI program titled "Same is Different" (Program Managers Dr Marc Ulrich and Dr Pani Varanasi).

Future Capability: This effort is helping enable SPD, which is enabling more sensitive sensing devices, such as for improved night vision, spectral sensing for detecting composition at a distance, and more.

Knowledge Nuggets: Despite visual appearances, 2-D twisted graphene patterns are aperiodic in all directions and lie on a 2-D surface with nonzero curvature almost everywhere. This can be understood through the nature of the mechanical relaxation (Fig. 1).

Mechanical Relaxation

- ▶ Incommensurate. **Moiré patterns are not periodic!**
- ▶ When two layers are closely aligned, they form large-scale moiré patterns.
- ▶ This leads to slowly varying long-range atomistic relaxation.
- ▶ Electronic edge states and localization, valleytronics, topologically protected conduction



M. Lusk (UofM)

Incommensurate 2D Materials

March 27, 2019

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Fig. 1 Large-scale moiré patterns in closely aligned 2-D layers

- New mathematical models for new governing mathematical operators (i.e., Hamiltonians) of 2-D heterostructures can now be relatively quickly constructed, from which electronic structure and transport can now be relatively quickly computed.
- This new ability to compute electronic structure and transport relatively quickly and accurately is now enabling materials scientists and microelectronics engineers to tackle design of ultra-sensitive detection devices.

3. Knowledge Transitions

The Multiscale Mathematical Modeling and Design for the Realization of Novel 2-D Functional Materials MURI has produced over 150 peer-reviewed journal articles and conference presentations, developed novel software, shared ideas and papers with US Army Combat Capabilities Development Command (DEVCOM) Army Research Laboratory (ARL) scientists, trained 16 new PhDs with placement into new jobs including at the University of Minnesota and Harvard University, and gave the now widely used name “Twistronics” to this new field.

3.1 Publications

A complete set of publications is available in the Final Report Appendix (p. 7).

3.2 Engagements with DOD

ARL–Army Research Directorate scientists Madan Dubey and Mahesh Neupane actively assisted in device fabrication and sample design with the MURI team, helped guide the team by sharing what types of applications they were interested in, and received knowledge products from the MURI during the course of the work.

Naval Research Laboratory scientists William Oliver and team collaborated with the MURI team in developing measurement techniques for thermal transport.

Co-Principal Investigator (PI) Philip Kim (Harvard) received a Vannevar Bush Fellowship in 2018 while conducting this DOD-supported research.

O/USD(R&E) scientists have tracked this effort and are now supporting it as one of their own small business commercialization efforts.

3.3 Software

Research-quality software used to conduct the investigation developed under this award is available at:

<https://github.com/stcarr/leo2d>

https://github.com/stcarr/kp_tblg

POC for download/install questions is Dr Stephen Carr,

stephen_carr1@brown.edu

3.4 Commercial Transitions

FY22–25: This work is now being transitioned commercially through the O/USD(R&E)-sponsored STTR Phase 2 80437-OD-ST1 (2022–2025), by Truventic Inc, PI Gonzalez: “Ultra-Sensitive Microwave, THz, and IR Sensors Based on Tunable Josephson Junctions, Realized in Graphene Moiré Superconductors”. This is developing commercial SPD capability to enable more sensitive sensing devices, such as for improved night vision, spectral sensing for detecting constitution at a distance, and more.

Appendix. Research Performance Progress Report (RPPR) Final Report

The appendix appears as a pdf attachment to the report.

List of Symbols, Abbreviations, and Acronyms

2-D	two-dimensional
ARL	Army Research Laboratory
DEVCOM	Combat Capabilities Development Command
DOD	Department of Defense
FY	fiscal year
MURI	Multi-University Research Initiative
PI	Principal Investigator
O/USD(R&E)	Office of the Under Secretary of Defense for Research and Engineering
RPPR	Research Performance Progress Report
SPD	single photon detection
STTR	Small Business Technology Transfer Program

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