Graphene-based sensors for detecting special nuclear materials

Purdue University
Yong P. Chen (PI), Igor Jovanovic (Co-PI)

Academic Research Initiative (ARI) Grantees Conference, Washington DC April 8, 2009
What is G.U.A.R.D.

Graphene-based Ultrasensitive Advanced Radiation Detectors
(for gamma rays and neutrons – SNM detection)

Relevance and Goals
- Ultimate vision: Array/stack of graphene-based radiation sensors with ultrahigh spatial and energy resolution to detect special nuclear materials
- Potential Advantages over state-of-the-art (high purity Ge or superconductor TES): high speed (not based on charge collection); high energy/spatial resolution; room temperature operation; compact/portable size
- Potentially transformational impact: new paradigm of nuclear radiation detection based on field effect

Technical Approach
- Primary approach: use sharp field effect in graphene to detect electric perturbations by ionizing radiation
- Secondary approach: use graphene composite to sense physico-chemical changes due to radiation
- Technical challenges:
  - new approach(es), new material [2004-], new science
  - speed/sensitivity/energy resolution unknown [promising]
  - Exploratory, higher-risk [high-pay off]
  - (Scalable production of materials/devices) [rapid progress!]

(How) Can graphene work (well)?

“graphene-based”=
Graphene + absorber + …
Outline

• Overview
  – Team
  – Program Vision and Milestones
• Graphene for Radiation Sensor
  – Radiation detection
  – Graphene properties
• Research Progresses and Accomplishments
  – Modeling of ionizing radiation in materials
  – Modeling of graphene response
  – Graphene Field Effect transistors
  – Proof of concept experiments
  – Reliability and effect of charged irradiation on graphene
• Education and Outreach
• Future Work & Conclusions
ARI-GUARD Team @ Purdue

Faculty
Prof. Yong P. Chen (Physics & ECE)
Prof. Igor Jovanovic (Nuclear Engineering)

Postdoc
Dr. Romaneh Jalilian

A diverse and interdisciplinary team

Graduate Students (see their posters!)

Isaac Childres (Physics)
Mike Foxe (Nuclear E)
Gabriel Lopez (ECE)

Undergraduate students (Spring 2009)
Sarah Alexander (CheE), Justin Gregory (NE), Elaine Li (ME), Stephen Schiffli (ECE)

Graduate Students (see their posters!)

Nanomaterials and nanodevices
Nanofabrication and characterization of graphene devices
Graphene composites & radiation response
Local-gated graphene field effect transistors
High mobility GFET

Nanofabrication
Graphene devices and GFET
Effects of charged particle irradiation and reliability

Modeling of radiation-absorber interaction
Laser and gamma ray measurements

Graphene material fabrication
COMSOL modeling of GFET
GFET characterization and high speed GFET
GFET on small bandgap semiconductors for radiation

Part-time staff help (nano device fabrications): Dr. Yi Xuan (Fall 2008), Dr. Jifa Tian (Spring 2009)

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Overall Program Vision

• Phase I (09/2008-08/2011):
  basic material science and device physics to
  find the optimal scheme to use graphene to
  sense ionizing radiation from SNM → science

• Phase II (09/2011-08/2013):
  – Detector & integrated architecture
    development and performance optimization
    → engineering/product development
Fist Year Program Milestones

Program/Personnel
• 09/2008: Funding arrived, GUARD program kicks off
• 09/2008: 2 Graduate RAs hired: Isaac Childres and Mike Foxe
• 01/2009: Postdoc (Romaneh Jalilian) joins
• 01/2009: 3rd Graduate RA Gabriel Lopez hired --- core team completed!

Technical
• 12/2008: Graphene devices fabricated and field effect observed
• 12/2008: Optical and Gamma Radiation Measurement Setup
• 02/2009: MCNP/CASINO modeling of radiation-absorber interaction
• 03/2009: Local field effect on graphene demonstrated
• 03/2009: COMSOL modeling of response of GFET on local ionized charge (by radiation)
• 03/2009: photo irradiation sensor based on GFET demonstrated
Research Facilities @ Purdue

Birck Nanotechnology Center (BNC)

Applied Physics Laboratory

Nuclear Engineering

Physics

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Yong P. Chen (yongchen@purdue.edu)
Part II: Why **Graphene** for Radiation Detection

SNM Radiation Sensing --- some state of art: High Resolution is Desirable

- HPGE (High Purity Ge)
  
  [charge collection]

- Superconductor TES (Transition Edge Sensor)
  
  [NOT charge collection; use a *sharp feature*]

Can we have a *sharp feature* @ ~ room T that couples to radiation(effect)
What is Graphene

1000+ papers in 2008

- (electrically isolated) “discovered” in 2004
- Building block of many carbon (nano)materials
- New “wonder” semiconductor/semimetal
- Amazing Electrical Properties – (“post Si” electronics/’Moore’)
- Amazing Mechanical Properties --- highest strength (~CNT)
- Amazing Thermal properties – highest thermal conductivity
- Easy to make and work with (2D planar fabrication)

Electrons in graphene

\[ E = p v_F = \hbar k v_F \]
\[ v_F \sim 1 \times 10^6 \text{ m/s} \]

- Dirac equation
- Chiral massless fermions
- [QED/QCD in graphene]

Usual solid          graphene

- High conductivity/mobility (>10X Si @ room T)
- Low (electronic) noise
- Tunable (electr.) properties
- Exposed to environment
  --- excellent sensor mat.

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**Sharp Electric Field Effect in Graphene GFET**

- **Graphene**
- **Semiconductor**
- **Insulator**
- **$V_{\text{gate}}$**

- **Finite R (quantum R)**
- **Low noise**
- **High mobility** (can ballistic)
- **High speed [THz]**
- **High sensitivity** [$dE/E<10^{-3}$]
- **Bandgap eng possible**
- ... all these even at 300K

**“the sharp feature”**

---

**Detection of individual gas molecules adsorbed on graphene**


1. Manchester Centre for Mesoscopic and Nanotechnology, University of Manchester, Manchester, M13 9PL, UK
2. Institute for Microelectronics Technology, 142413 Chernogolovka, Russia
3. Institute for Molecules and Materials, University of Nijmegen, 6525 ED Nijmegen, Netherlands

*Published online: 20 July 2007; doi:10.1038/nnano.2007.196*

The ultimate aim of any detection method is to achieve such a level of sensitivity that individual quanta of a measured entity can be resolved. In the case of chemical sensors, the quantum is one atom or molecule. Such resolution has so far been beyond the reach of any detection technique, including solid-state gas sensors that require them for their exceptional sensitivity. The fundamental reason limiting the resolution of such sensors is fluctuations due to thermal motion of charges and defects, which lead to intrinsic noise exceeding the sought-after signal from individual molecules, usually by many orders of magnitude. Here, we show that micrometre-size sensors made from graphene are capable of detecting individual events when a gas molecule attaches to or detaches from graphene’s surface. The adsorbed molecules change the local carrier concentration in graphene on by one electron, which leads to step-like changes in resistance. The achieved sensitivity is due to the fact that graphene is an exceptionally low-noise material electronically, which makes it a promising candidate not only for chemical detectors but also for other applications where low noise is desired, such as charge, magnetic field or mechanical strain are required.

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**F. Schedin et al’2007**

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GFET for radiation sensing

- Graphene a highly sensitive to detect local Efield-change [single molecule sensitivity]
- $V_{\text{gate}}$ tunable $\rightarrow$ sensitivity and resolution
- NOT relying on collecting/drifting ionized charges; appearance of ionized charges changes electric field
- sensing Efield intrinsically faster than sensing drifted/collection charges
- can work with variety of absorber substrates for gamma/neutron interaction; thin insulator layer

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Yong P. Chen (yongchen@purdue.edu)
Possible alternative schemes: Physico/Chemical Change due to radiation

Graphene membrane ➔ Nanomechanical resonator

→ f and Q could change upon “bulging”

[more suitable as dosimeter]
Graphene Composite+ for Radiation Sensing

Graphene composite:
Graphene sheets dispersed in polymer matrix “bottom up” approach

(www.composite+)
○: semiconductor/absorber nanoparticles

Stankovich et al’06

How would radiation interacts with such material

How would composite respond to radiation in a detectable way?
Part III: Technical Accomplishments (09/2008-)

Focus: GFET based radiation sensor
(sharp change of resistivity due to change of electrical field caused by ionizing radiation)


Poster 2. G. Lopez: Characterization of graphene field effect transistors for use in a radiation sensor

Poster 3. I. Childres: Fabrication of graphene devices and studies of effects of charged-particles irradiation
Modeling: radiation-substrate (absorber) interaction

- MCNP and MCNP-Polimi modeling of Interaction of ionizing radiation (gamma, neutrons) with absorber materials: various semiconductors (Si, Ge, InSb), polystyrene etc.

- Compton electron transport through substrate (CASINO)  
  M. Foxe et al  
  (see poster 1)

Gamma ray in silicon  
→ conduction region
COMSOL Modeling: GFET response to radiation ionized charges

Gabe Lopez et al., poster 2
Energy and Direction Resolution

GFET

absorber

radiation

graphene 1

graphene 2

(b)
Material/Device Fabrication of Graphene and GFET

“exfoliation” (scotch tape)

Graphene on doped Si with 300 nm oxide

E-beam lithography

Develop PMMA

Cr/Au evaporation

Lift off

Lithography → Nanodevices

Doped Si

SiO₂

PMMA

Cr/Au

Photo by Sambandamurthy
How to identify graphene

Optical microscopy -- seeing is believing

Raman Spectroscopy
(also sensitive to defects in graphene)

Many layers

Quantum Hall Effect
(magnetoresistance)

I.Childres et al (see poster 3)
GFET Characterization

Semiconductor=Si
Insulator=300nm SiO2

See G Lopez poster 2
Local Field Effect: by Side Gate

Graphene is sensitive to local electric field

Isd (A)

Vsd=2mV

Vsd=-2 mV

Vsg (V)
Scanning Local Field Effect GFET –Coated Nanowire/AFM tip

M.M. Yazdanpanah, NaugaNeedles LLC
[Ga/Ag NW coated with parylene] + AFM tip

Tip/base as absorber?

J. Romaneh et al., work in progress
Proof of Concept: Photo-actuated GFET

Also tried photo-resistor
similar to photodiode
Also tried MOSFET
See G. Lopez et al. poster 2

Photodiode
- Vds of GFET at 20HZ @Vg=6V
- Vds of GFET at 100HZ Ids=10uA
- Vds of GFET at 500HZ

Radiation [this case, laser]
Charged-particles irradiation: e-beam

Motivation: effect of energetic charged particles (e.g., electrons) (long term) reliability of GFET radiation sensors

30keV electron beam
I= 0.15nA; Time=5mins; Expose area: 50um x 50um
Estimated dose: 112.5 e-/nm²

e- beam adds to graphene negative (n-) charges

(I. Childres poster 3)
Charged-particles irradiation: O\(^+\) ions

O\(^+\) ions generated in a “microwave” plasma chamber

- O\(^+\) ions add to graphene positive (\(p^+\)) charges

Defect creation studied also by:
- Raman spectroscopy
- AFM (atomic force microscopy)
- time-dependent behavior

I. Childres et al., in preparation
See poster 3
Other Work on Graphene-based Materials

• Fabricating Graphene on Ge & other substrates

(w/t 30nm germanium oxide)

• Synthesizing Graphene Oxide & Graphene Composite
Recent Breakthrough: Large-scale CVD-grown Graphene Film

Collaboration with Univ. of Houston Center for Advanced Materials

- Grown by CVD/segregation on Ni then transferred to any substrate
- >~cm in size; scalable low cost production
- Mobility ~5000 cm²/Vs
- Transparent, bendable and stretchable


See also:
- & MIT (2009)

Scalable production of graphene is possible
Publications/Presentations

• 10th International Conference on Applications of Nuclear Techniques (Crete, June 2009) (I. Childres et al. on irradiation actuated GFET) [accepted]

• In preparation:
  – IEEE NSS/MIC (Orlando 2009) on graphene radiation detector & modeling
  – Manuscript on charged-particle irradiation effect
  – Manuscript on local gate/charge FET

Other publications/presentations:
• arXiv 0901.1136 (2009) [large scale graphene/FET]
(Near) Future Work

- Gamma rays / neutron response
- GFET on undoped substrates
- Speed/sensitivity/energy resolution
- GFET on other substrates
  - Ge; InSb; CZT, TIBr ....
    (higher-Z & low/medium band-gap substrates)
  - B/Li containing substrates
- Explore graphene composites (“bottom up”)
- ... integrated detector/sensor & architecture
- ......
Part VI: Education & Outreach

Interdisciplinary training of US scientists/engineers for S&T challenges in nuclear security

- Postdoc mentoring --- Dr. Romaneh Jalilian: develop dual expertise in nanomaterials/devices and radiation detection

- Graduate students
  - Mike Foxe (Nucl E): MCNP/CASINO; nuclear detectors; recipient of ANS Student Conference best presentation award.
  - Isaac Childres (Physics): particle physics; nanofabrication; nanodevices
  - Gabe Lopez (ECE): Semiconductors, radiation sensors; COMSOL, High speed GFET and nanoelectronics

### ARI/GUARD PhD Curriculum

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<thead>
<tr>
<th>Radiation Detection &amp; Nuclear Engineering</th>
<th>General Experimental Physics/Measurement Techniques</th>
</tr>
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<tbody>
<tr>
<td>Glenn F Knoll, Radiation detection and measurement 3rd ed, 2000</td>
<td>WR Leo, Techniques for nuclear and particle physics experiments, a how-to-approach, 1994</td>
</tr>
<tr>
<td>G. Lutz, Semiconductor radiation detectors, 2007</td>
<td>Nanoscience and nanotechnology</td>
</tr>
<tr>
<td>FH Attix, Introduction to Radiological physics and radiation dosimetry, 1986</td>
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ARI Education (cont.)

Graduating/Training new generation of US scientists/engineers trained with highly-interdisciplinary background/approaches to address S&T challenges for nuclear security

Undergraduate Students/Interns --- leveraged by:

New courses developed at Purdue:
- Spring 2009: PHYS 570X “Carbon nanophysics and devices” (Y. Chen)
- “Detection for nuclear security” (I. Jovanovic, in preparation)
Outreach Activities

Educating public on importance of S&T research for nuclear security

• Purdue Homeland Security Institute (PHSI)

• Lecture to High School Teachers attending Workshop at National Center for Learning and Teaching of Nanoscale Science and Technology (NCLT) (K-)9-12

Engaging with national lab/industry for collaboration/R&D

• Visit to national labs: ORNL (Jovanovic), ANL (Chen)

• Industrial Collaboration/Connections:
  – Canberra Industries
  – Naugle needles
  – Semiconductor Research Corporation (SRC)-Nanoelectronics Research Initiative (NRI)
  – SEMatech
  – IBM
ARI/GUARD Website and Wiki

GUARD Website (public)
https://www.physics.purdue.edu/quantum/GUARD

ARI/GUARD Group Wiki (internal)
http://aripu.wikispaces.com/

Graphene-based Ultrasensitive Advanced Radiation Detector (GUARD)

Put logo here centered?

Recent News:
Website Updated!

The Project:

Develop radiation sensors and detector architectures based on graphene, a novel electronic material with many exceptional properties of potential use for special nuclear material (SNM) detection. This technology has the potential to significantly outperform current detectors, such as the gamma-ray detectors based on high-purity germanium (HPGe), in terms of the energy and spatial resolution, speed and functionality.

Relevance for SNM detection: The graphene-based detector we aim to develop could be used to detect both SNM-produced photons and neutrons when used in conjunction with various high-attenuation or hydrogenous materials. Unlike the established detector technologies based on electron collection in semiconductors, the graphene-based detector would instead utilize a sharp change in electrical conductivity of graphene in the presence of transient electrostatic potential resulting from the charge produced by ionizing radiation. The use of electrostatic potential instead of charge collection eliminates the requirement for high carrier mobility and lifetime that characterizes conventional charge collection-based detectors. Three key advantages could result from the use of this novel detection method. In the absence of leakage current, it is possible to (1) use inexpensive absorber materials of low purity which may be available in large volumes, (2) use semiconductors such as Ge at room temperatures, and (3) deploy the next generation of ultra-narrow band semiconductors such as InSb as absorbers, without the requirement for active cooling. Therefore, a potential exists for unprecedented energy resolution at room temperatures, in addition to low cost, large volume, and environmental tolerance. The proposed research is thus synergistic with several key program thrust areas and would have a transformational impact on SNM detection.

Intellectual Merits: One of the most important enabling factors for future SNM detection technologies will be the development of radiation detectors based on new materials, concepts, and designs that maximize the efficiency of the system while producing the best possible energy resolution. Our approach is unique and innovative in comparison to the SNM radiation detectors proposed and developed to date in several key aspects. First, the project utilizes a novel material, graphene, which exhibits unique properties with high potential impact on the next-generation radiation sensors. Second, our approach utilizes a previously unexplored material property to detect charge produced by ionizing radiation. Third, this novel detection method is not only compatible with the existing well-developed absorber materials such as Ge, but it also offers a unique opportunity to deploy other narrow-bandgap materials such as InSb without temperature, purity, carrier mobility, or lifetime constraints. Finally, our approach is scalable by stacking or through the use of novel graphene composite structures. When combined, these key features of our approach indicate a potential for a paradigm shift in nuclear detection, with significant impact beyond SNM detection and extending into medical applications, monitoring of nuclear fuel cycle, and high-sensitivity detectors for particle and nuclear physics.

ARI Grantees Conference
Washington DC, 04/08/2009
ARI-GUARD events

- Weekly ARI/GUARD Meeting
- reports/progresses uploaded on Wiki

- Seminars

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<th>Fall 2008</th>
<th>Topic</th>
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<tr>
<td>Peter Yu (UC Berkeley)</td>
<td>Semiconductors for Radiation Detection</td>
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<td>George Milley (UIUC)</td>
<td>Nuclear Energy and Hydrogen Economy</td>
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<th>Spring 2009</th>
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<td>Jean Paul Allain (Purdue Nucl Eng)</td>
<td>Simulated Experiments of Particle and Plasma-Surface Interactions at the Nanoscale</td>
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<tr>
<td>Jie Lian (RPI)</td>
<td>Materials Behavior under Extreme Environments: Nuclear Engineering Application and Nano-scale Materials Design</td>
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Acknowledgments

• Current and Past Team Members

• Colleagues/Collaborators:
  Prof. David Koltick (Purdue Physics/Applied Physics Lab)
  Prof. Leonid Rokhinson (Purdue Physics)
  Materials providers:
  Prof. Peide Ye (Purdue ECE)
  Prof. Hao Li (U Missouri MAE)
  Prof. Dima Dykin (Northwestern U)
  Prof. Qingkai Yu (U Houston ECE)

• NSF

• DHS-DNDO
Conclusions

Graphene sensors for SNM radiation (γ/n) detection
• compatible with a wide variety of absorber materials
• NOT drifting/collecting charges
• based on sharp field effect
• exciting promises:
  high speed, high sensitivity,
  excellent energy resolution, room T
• potentially opening a new approach for radiation detection

Initial accomplishments --- focus on proof-of-concept
• Modeling/design of graphene FET radiation sensor
  MCNP/CASINO modeling of γ/n-substrate interaction
  COMSOL modeling: demonstrate GFET response to radiations
• Fabricating/testing graphene & GFET (graphene field effect transistor)
  high quality graphene material fabricated
  global and local electric field effect demonstrated
  demonstrated laser-irradiation actuated GFET

On-going/future work
• fabricate/test GFET on a variety of absorbers: Si, Ge, InSb, CZT, TlBr, ...
• radiation response experiments
  gamma rays
  neutrons
  alpha/beta etc
• graphene radiation detector:
  study energy resolution, sensitivity, speed etc.
  detector design, architecture and integration
• charged-particle radiation on graphene/GFET effects of O+ ions & e− beams demonstrated
  reliability of GFET radiation sensors

Education/Outreach/Collaboration
• interdisciplinary training of students for nuclear security S&T challenges
• educating public on importance of S&T research for nuclear security
  industry/national lab collaboration
• graphene composite (physico-chemical approach)

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A historic and futuristic remark...

Radiation detection has been very closely coupled with advances in semiconductors.

**How Can graphene do it (well)?**

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“The gravest danger we face—nuclear terrorism…”
--- 07/16/08 at Purdue University (Summit on Confronting New Threats)

“And the biggest threat that we face right now is not a nuclear missile coming over the skies. It’s in a suitcase.”
--- 09/28/2008 during 1st Presidential debate
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  - gamma rays
  - neutrons
  - alpha/beta etc
- graphene radiation detector:
  - study energy resolution, sensitivity, speed etc
  - detector design, architecture and integration
- graphene composite (physical/chemical approach)

Education/Outreach/Collaboration
- interdisciplinary training for nuclear security S&T challenges
- educating public on importance of S&T research for nuclear security
- industry/national lab collaboration

ARI Grantees Conference
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**Faculty**
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- Prof. Igor Jovanovic (Nuclear Engineering)

**Postdoc**
- Dr. Romaneh Jalilian

**A diverse and interdisciplinary team**
- Nanomaterials and nanodevices
- Nanofabrication and characterization of graphene devices
- Graphene composites & radiation response
- Local-gated graphene field effect transistors
- High mobility GFET

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