

Theoretical studies of 2D systems

Nithaya Chetty

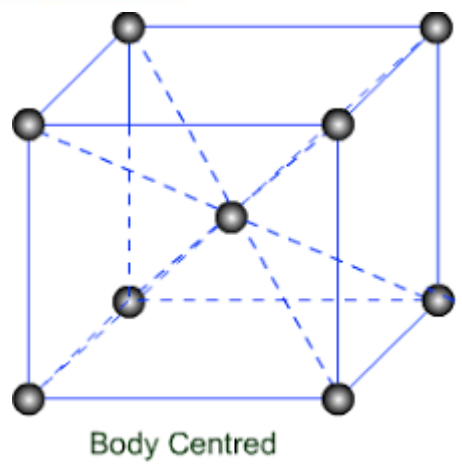
University of Pretoria, South Africa

To the students

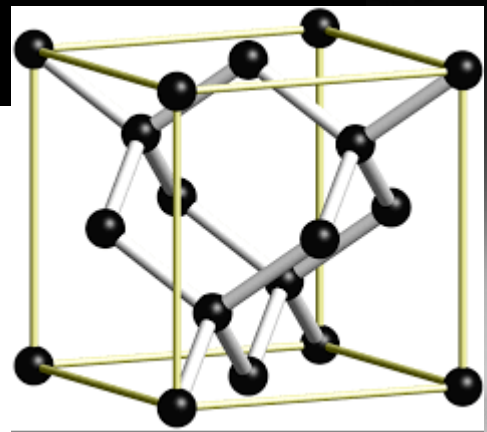
- Material science is arguably the most important discipline that needs to be developed in Africa
 - Many initiatives are already underway
- Science is foremost an empirical endeavor
 - Experimental equipment intensive
 - Governments/institutions need to invest more in equipment
 - Need more access to international laboratories
 - Need more scientific and technical training
- Theoretical and Computational studies are accessible and cheap
 - All you need are smart students (of which there are aplenty in Africa)
 - Access to moderate to good computing facilities



iron ore



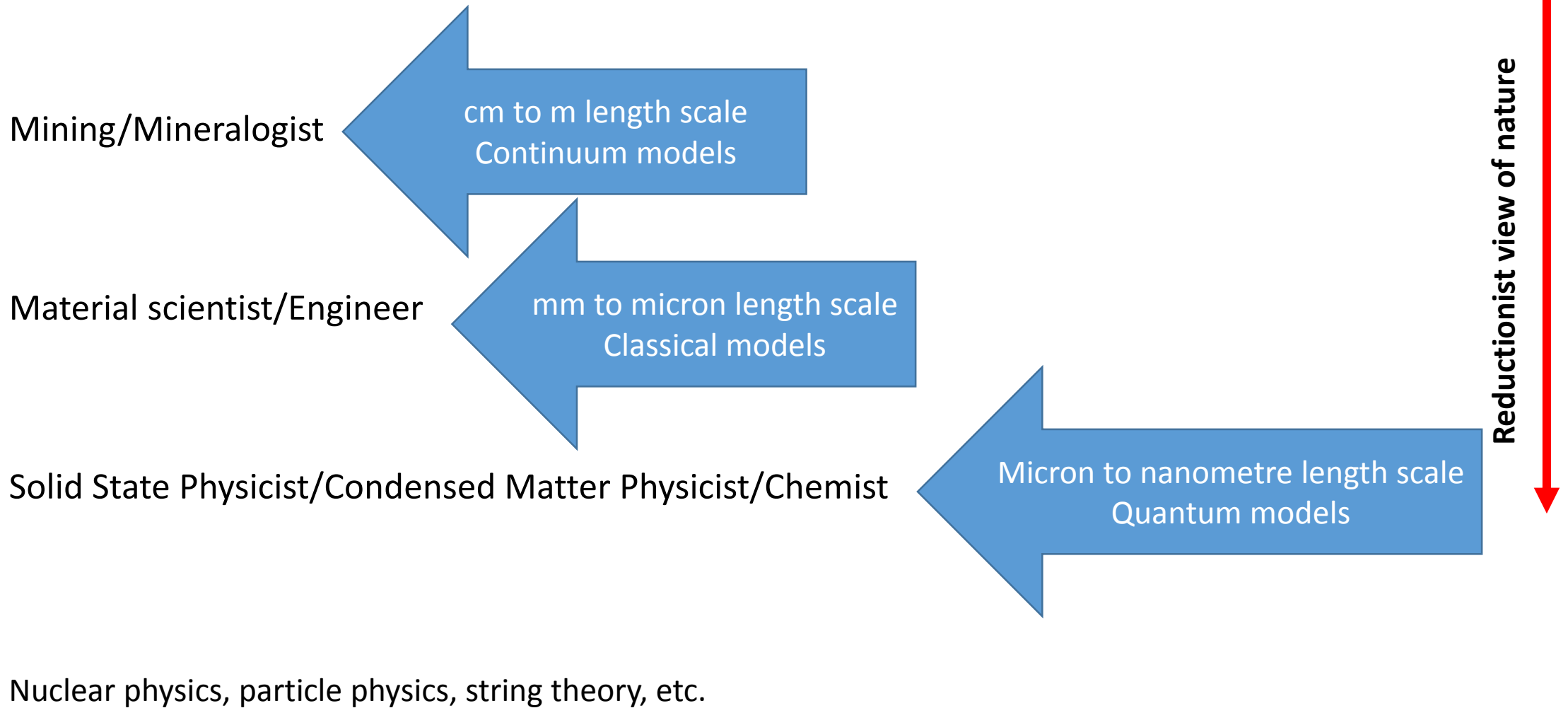
rough diamond



cut and polished diamond

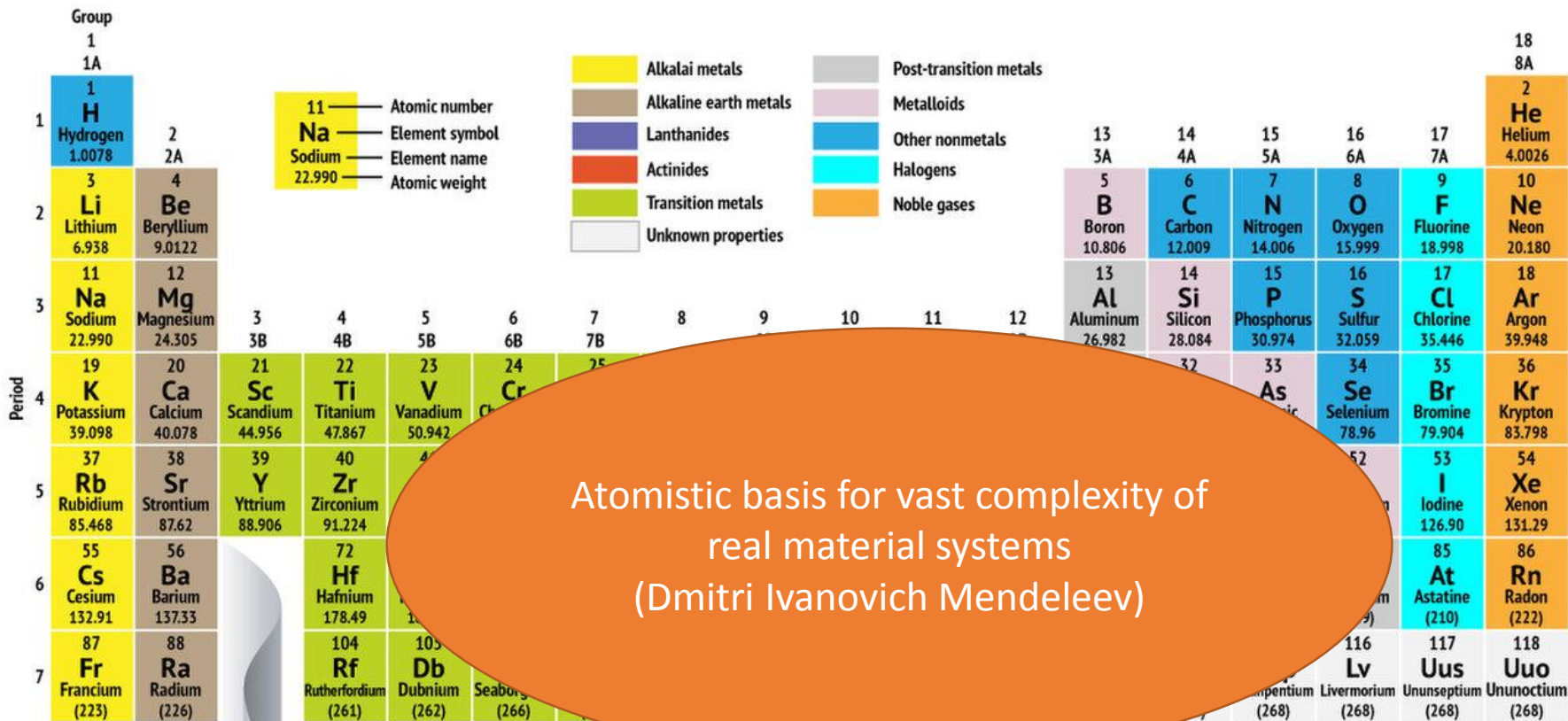


Hierarchy of disciplines



Why only 110 elements?

Periodic Table of the Elements



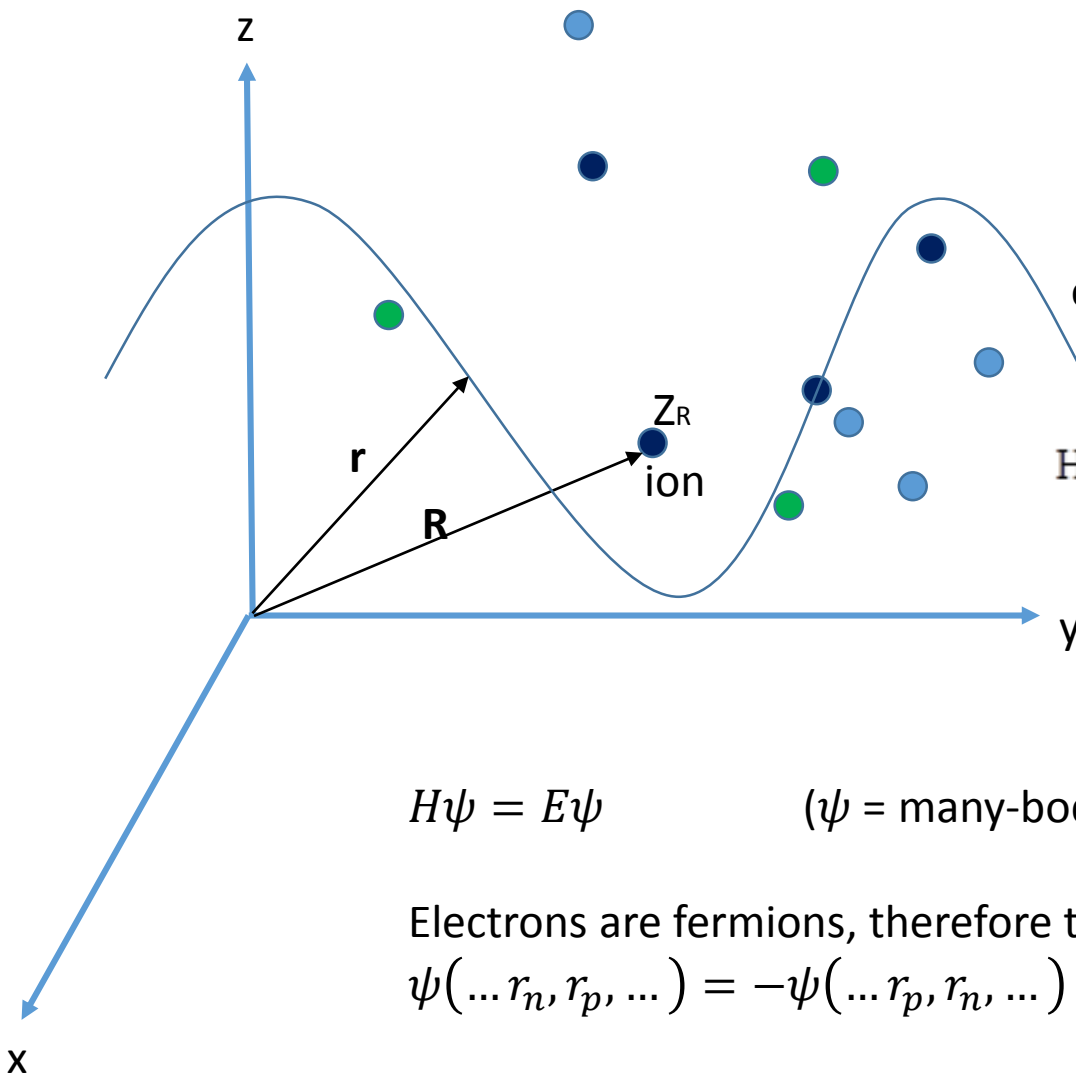
Where have the elements come from?

Lanthanides	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Actinides	89	90	91	92	93	94	95	96	97	98	99	100	101	102	103
	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

- Electronic or magnetic properties, e.g. metals, semiconductors, insulators, semimetals, superconductors (conventional, high temperature), heavy fermions, magnetic materials (ferrimagnetism, ferromagnetism, antiferromagnetism, Curie paramagnetism, Paul paramagnetism, diamagnetism)
- Crystal structures, e.g. BCC, FCC, diamond, graphite, silicon, silicon dioxide, quartz, calcite, mica, graphite, diamond, silicon, silicon dioxide, quartz, calcite, mica
- Groups, e.g. alkali metals, transition metals, noble gases, noble metals
- All of these are complex and interconnected
- Defects (substitutional impurities, interstitial impurities, defect complexes), line defects (stacking faults, dislocations), surface defects (adsorption, desorption, catalysis, semiconductor devices)
- Amorphous, polycrystalline and certain crystalline materials are not periodic, have useful properties and applications, e.g. lighter and harder materials, polycrystalline Si for photovoltaic cells
- Surfaces, e.g. (111) surface, which enables one to consider catalysis and surface reactivity
- Interfaces, e.g. in quantum well structures, transistors, solid state lasers
- Thin films and multi-layers e.g., photovoltaic cells, giant magneto-resistance
- Nanostructures e.g. quantum wells, quantum dots, quantum wires exhibit quantum effects
- Nanosystems often straddle classical and quantum mechanics, and this is why both theoretical and experimental methodologies and instrumentation are applicable
- Variety of properties, e.g. transport properties (electrical and thermal conductivity), crystal phase structures and transitions especially under conditions of temperature and pressure, hardness (brittle, ductile, malleable), elastic properties (bulk modulus, shear modulus, Young's modulus, Poisson ratio), lustre, optical properties (refractive index, absorption coefficient, reflectivity, transmission coefficient, dielectric function)
- Various probes, e.g. light (UV, IR), x-rays, electric fields and magnetic fields, temperature and pressure, neutrons, electrons, positrons to create a variety of excitations and collective excitations, such as spinons (spin waves), phonons (lattice waves), magnons (magnetic waves), polarons (electron-hole excitations), etc.
- Various experimental techniques, e.g. x-ray diffraction, nuclear magnetic imaging, photoemission spectroscopy, scanning electronic microscopy, mass spectroscopy, etc., etc. Techniques applicable to other disciplines and to mainstream society, e.g. magnetic resonance imaging

Much complexity in Solid State Physics

Unifying theory is Quantum Mechanics



electronic wavefunction

$$H = \sum_i \frac{-\hbar^2}{2m} \nabla_i^2 + \sum_R \frac{-\hbar^2}{2M_R} \nabla_R^2 + \frac{1}{2} \sum_{i=j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{1}{2} \sum_{R=R'} \frac{Z_R Z_{R'} e^2}{|\mathbf{R} - \mathbf{R}'|} - \sum_{iR} \frac{Z_R e^2}{|\mathbf{r}_i - \mathbf{R}|}$$

$$H\psi = E\psi \quad (\psi = \text{many-body wave function})$$

Electrons are fermions, therefore the Pauli Exclusion Principle applies

$$\psi(\dots r_n, r_p, \dots) = -\psi(\dots r_p, r_n, \dots)$$

ie : wavefunction is anti-symmetric in the exchange of electron coordinates

The energy is central

- Why is graphite the more stable form of solid carbon?

- $p = -\frac{\partial E}{\partial V}$ basis for pressure induced phase transitions

- $F = -\frac{\partial E}{\partial R}$ basis for calculation of equilibrium structures, molecular dynamics

Elastic constants, phonon frequencies, electrical conductivity, etc, etc etc

Calculate charge density from wavefunction, basis for understanding bonding

Single particle energy spectrum basis for understanding electronic properties of solids (e.g. metals, semiconductors, magnetism, etc., etc.)

Density Functional Theory

- Energy is a unique functional of the density
- Recasts the many electron problem into an effective non-interacting problem



$$\langle \Psi | H | \Psi \rangle = E = T_s + E_C + E_{e-ion} + E_{ion-ion} + E_{xc}$$

10 most cited papers in Physical Review over 110 years

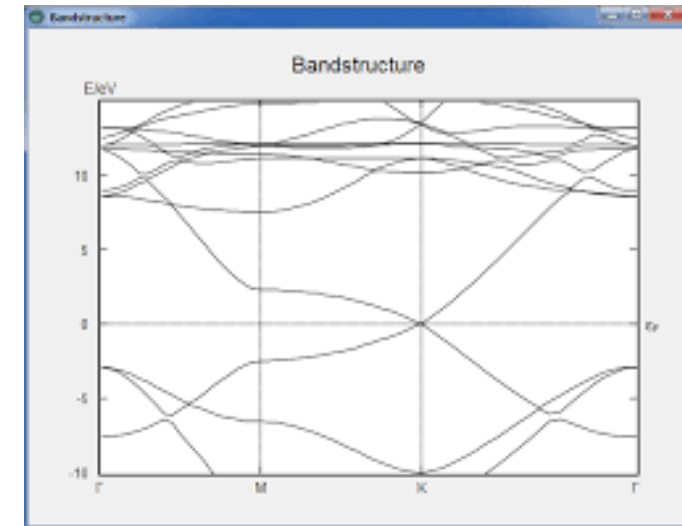
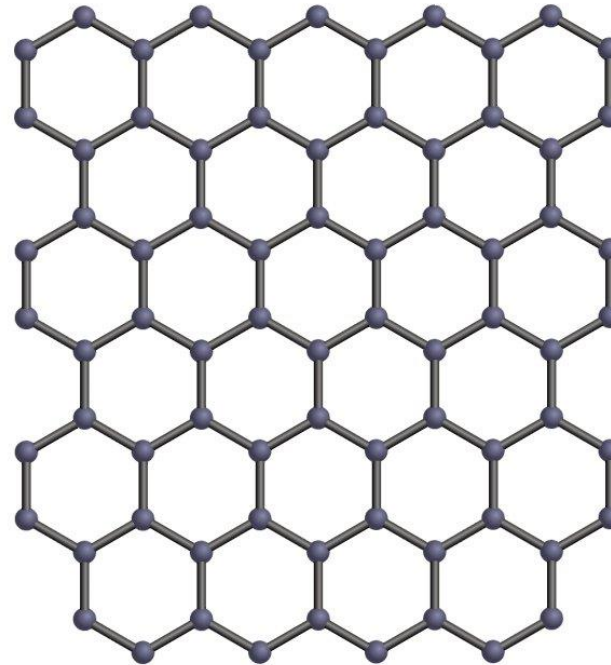
#	Title	Author(s)
1.	<i>Self-Consistent Equations Including Exchange and Correlation Effects (1965)</i>	W. Kohn, L. J. Sham
2.	<i>Inhomogeneous Electron Gas (1964)</i>	P. Hohenberg, W. Kohn
3.	<i>Self-Interaction Correction to Density-Functional Approximations for Many-Electron Systems (1981)</i>	J.P. Perdew, Alex Zunger
4.	<i>Ground State of the Electron Gas by a Stochastic Method (1980)</i>	D. M. Ceperley, B.J. Alder
5.	<i>Theory of Superconductivity (1957)</i>	J. Bardeen, L.N. Cooper, J.R. Schrieffer
6.	<i>Model of Leptons (1967)</i>	S. Weinberg
7.	<i>Linear Methods in Band Theory (1975)</i>	O. K. Andersen
8.	<i>Effects of Configuration Interaction on Intensities and Phase Shifts (1961)</i>	U. Fano
8.	<i>Disordered Electronic Systems (1985)</i>	P.A. Lee, T.V. Ramakrishnan
9.	<i>The Electronic Properties of Two-Dimensional Systems (1982)</i>	T. Ando, A.B. Fowler, F. Stern
10.	<i>Special Points for Brillouin-Zone Integrations (1976)</i>	H.J. Monkhorst, James D. Pack

Theoretical and Computational Solid State Physics Research Group @ Univ. Pretoria

- Density functional methods
- VASP, Quantum Espresso
- University cluster, CHPC

- Currently focussed on 2D materials
- Developmental work on local energy density within DFT
- New work at Centre for Functional Nanomaterials at Brookhaven

Li states on C-H pair vacancy in Graphane

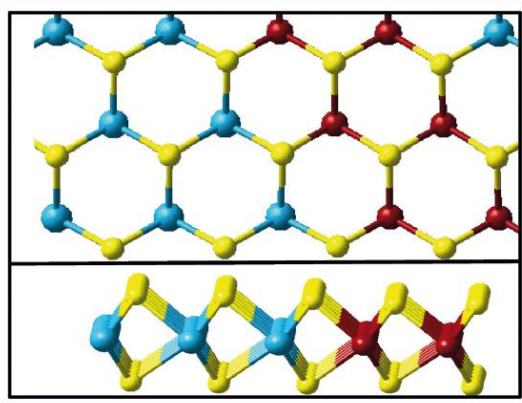


Edwin Mapasha, University of Pretoria
Mahlaga Molepo, University of South Africa

Volume 29 Number 32 16 August 2017

Topical review**Non-contact lateral force microscopy**

A J Weymouth



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Condensed Matter

Volume 29 Number 32 16 August 2017

TOPICAL REVIEW

323001 **Non-contact lateral force microscopy**
A J Weymouth

PAPERS**STRUCTURE, DYNAMICS AND PHASE TRANSITIONS**

325401 **High-pressure structures of yttrium hydrides**
Lu-Lu Liu, Hui-Juan Sun, C Z Wang and Wen-Cai Lu

ELECTRONIC STRUCTURE

325501 **Evolution of the topologically protected surface states in superconductor β -Bi₂Pd from the three-dimensional to the two-dimensional limit**
B T Wang and Elena R Margine

325502 **Static and time-resolved mid-infrared spectroscopy of Hg_{0.95}Cd_{0.05}Cr₂Se₄ spinel**
S Barsaume, A V Telegin, Yu P Sukhorukov, N Stavrias, V A Fedorov, T K Menshchikova and A V Kimel

325503 **Quantum well electronic states in a tilted magnetic field**
C Trallero-Giner, J X Padilha, V Lopez-Richard, G E Marques and L K Castelano

325504 **First-principles studies of chromium line-ordered alloys in a molybdenum disulfide monolayer**
N F Andriambelaza, R E Mapasha and N Chetty

325505 **Hydrogen bond symmetrization and elastic constants under pressure of δ -AlOOH**
Pietro Cortona

325506 **Comparison of nitrogen-vacancy complexes in diamond and cubic SiC: dose dependencies and spin-Hamiltonian parameters**
T L Petrenko and V P Bryksa

CORRELATED ELECTRON SYSTEMS

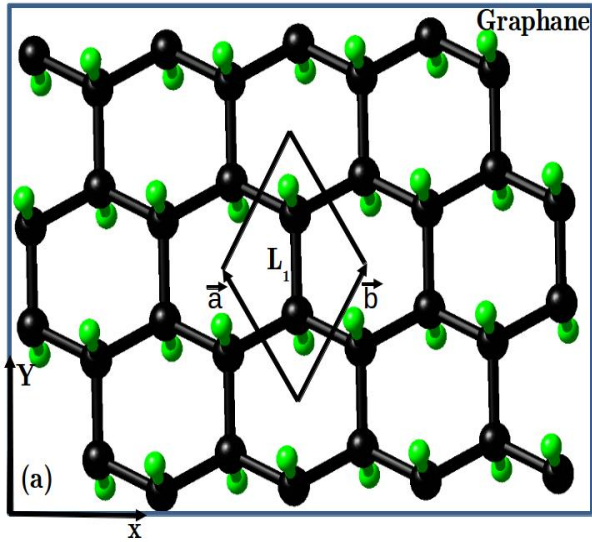
325601 **Magnetic behavior of metallic kagome lattices, Tb₃Ru₄Al₁₂ and Er₃Ru₄Al₁₂**
Sanjay Kumar Upadhyay, Kartik K Iyer and E V Sampathkumaran

PHYSICS OF MATERIALS

325701 **Thermopower and thermal conductivity in the Weyl semimetal NbP**
U Stockert, R D dos Reis, M O Ajeesh, S J Watzman, M Schmidt, C Shekhar, J P Heremans, C Felser, M Baenitz and M Nicklas

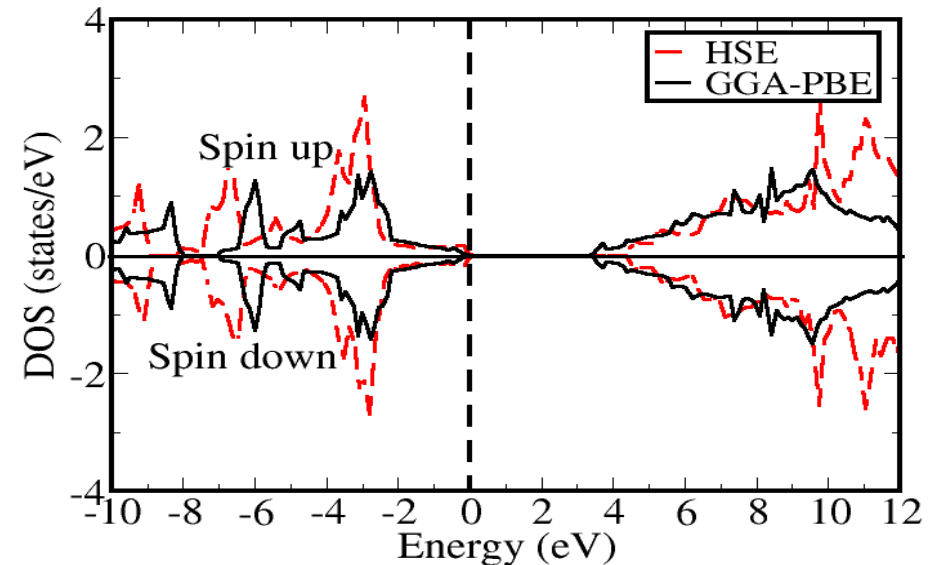
325702 **Oscillatory deviations from Matthiessen's rule due to interacting dislocations**
Chu-Liang Fu and Mingda Li

Overview of graphane



	Methods	a_0	d_{C-C}	Δ_{hC}	d_{C-H}	E_{gap}
This work	HSE06	2.52	1.53	0.44	1.10	4.51
	GGA-PBE	2.54	1.54	0.46	1.11	3.38
Other work	HSE06	2.52 ^a	1.53 ^a		1.10 ^a	4.60 ^a
	HSE06	2.52 ^b	1.53 ^b			4.38 ^b
	GGA-PBE	2.54 ^c	1.54 ^c		1.12 ^c	3.50 ^c
	GGA-PBE	2.52 ^d	1.52 ^d		1.10 ^d	3.50 ^d
	GGA-PBE	2.54 ^e		0.46 ^e		

- High volumetric hydrogen density
- Wide energy band gap
- Graphane is thermodynamically stable at room temperature
- Excellent candidate for energy storage devices
- Suitable for room temperature nano-technological devices



V_H defects in graphane

- Hydrogen vacancies (V_H) simplest defects
- DFT studies V_H introduces magnetism in graphane
 - localized magnetic moment $1 \mu_B$ per V_H
- Experiment reveals the magnetic features in a nearly full hydrogenated graphene
- We have investigated the effect of charge doping on the electronic properties of V_H
 - Charge doping fine tunes the electronic structure
 - Alters the localised magnetic moments
 - V_H defect can be used to design spintronic devices (spin dependent electron transport)

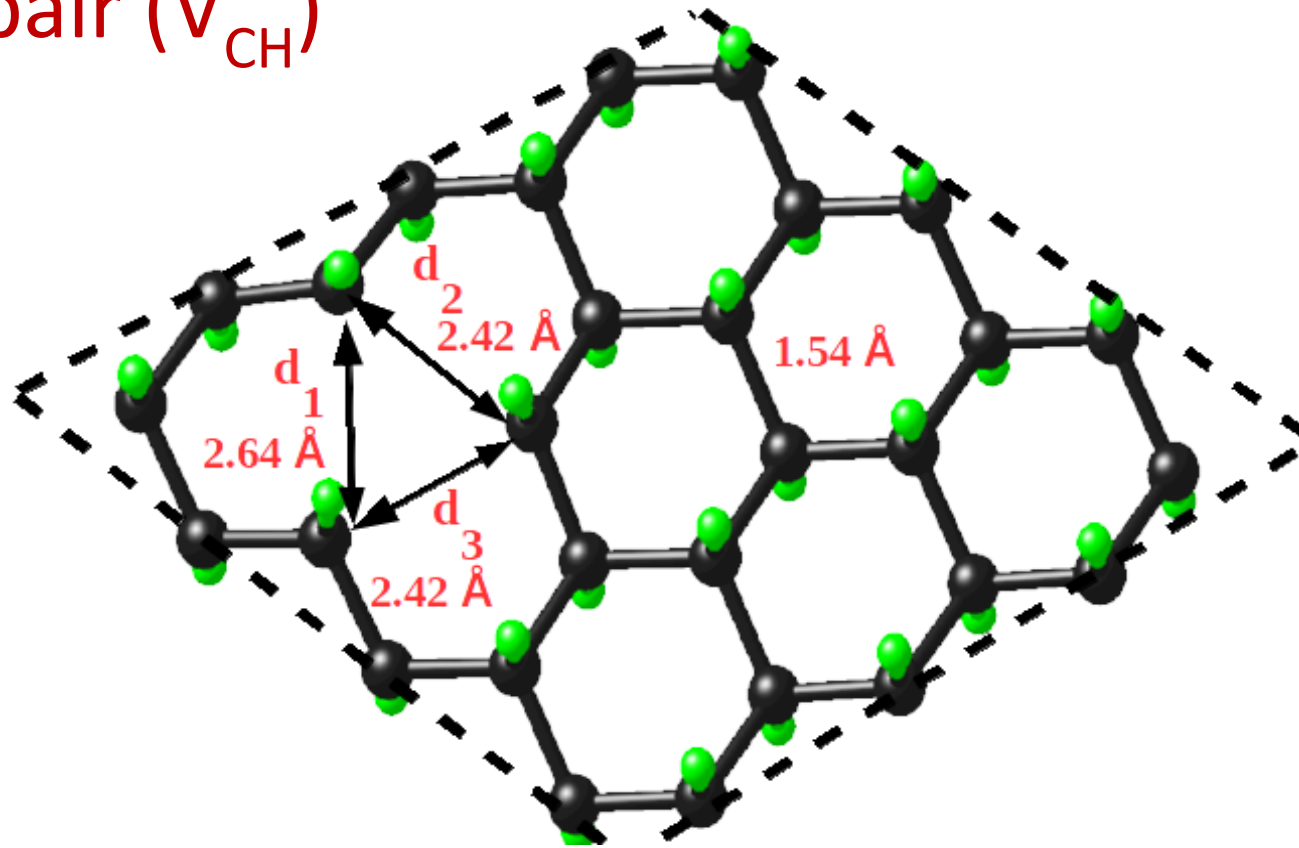
V_{CH} defects in graphane

- Experimentally reported carbon-hydrogen pair V_{CH} vacancies
 - achieved using high-energy ion beam pathways
- DFT studies V_{CH} defect induces the mid-gap states with a magnetic moment of $1 \mu\text{B}$ in a graphane monolayer.
 - magnetic moment arises from the dangling bonds on the three C atoms surrounding the V_{CH} vacancy
 - mainly contributed by C 2p orbitals
- Controlling the induced magnetism in a V_{CH} defected graphane could be essential for technological devices

V_{CH} defects in graphane

- We study interaction of a Li atom with a V_{CH} vacancy also considering the effect of charge states using HSE06 functional
- Vienna *ab initio* simulation packages (VASP) code
- The Heyd, Scuseria, and Ernzerhof functional (HSE06 functional)
- Projector augmented wave (PAW) method
- The energy cut-off was set to 500 eV
- The 10×10×1 k-mesh generated using Monkhorst-Pack scheme was used
- For large supercell systems, a 4×4×1 k-mesh was used
- The interlayer spacing along the z-axis was set to 15 Å to reduce the periodic image interactions

C-H vacancy pair (V_{CH})



- The C atoms surrounding the V_{CH} vacancy undergo structural distortions
- The graphane monolayer threefold symmetry breaks due to Jahn-Teller effect
- The three C atoms surrounding V_{CH} defect form an asymmetric triangular arrangement

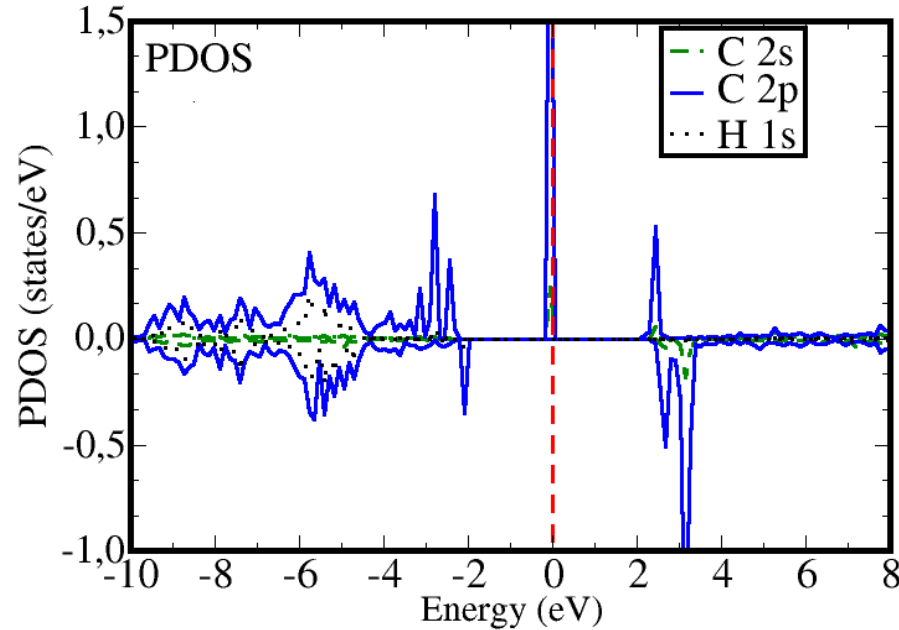
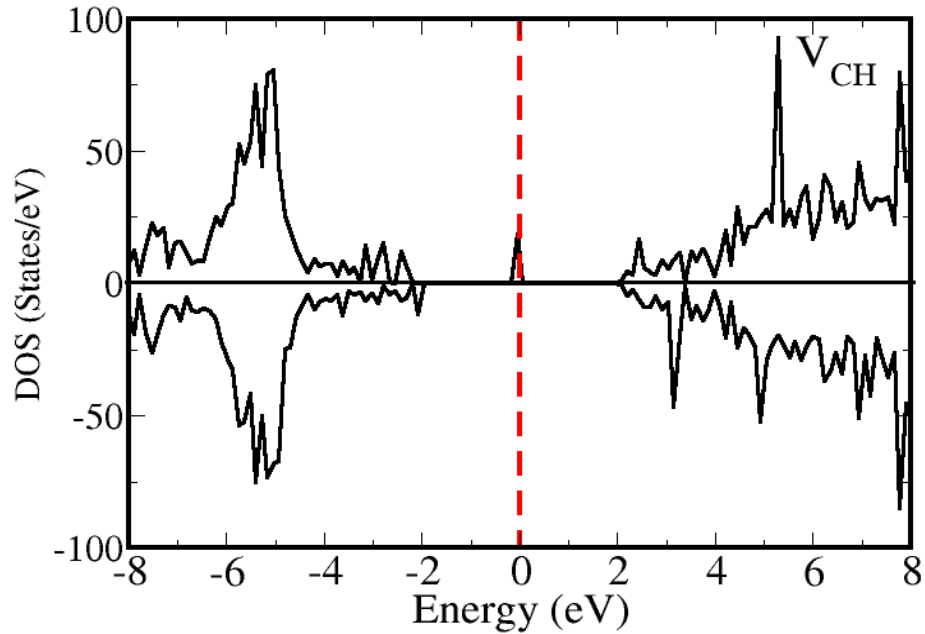
Thermodynamic stability of V_{CH}

- Formation energy 6.05 eV (HSE06) for V_{CH}
- Agrees well with the GGA value [1]
- The high formation is needed to first break the C-C bonds
- V_{CH} is realizable experimentally, can be stable at room temperature [2]

[1] Pujari *et al* J. Phys. Chem. C, **113**, 21063-21067 (2009).

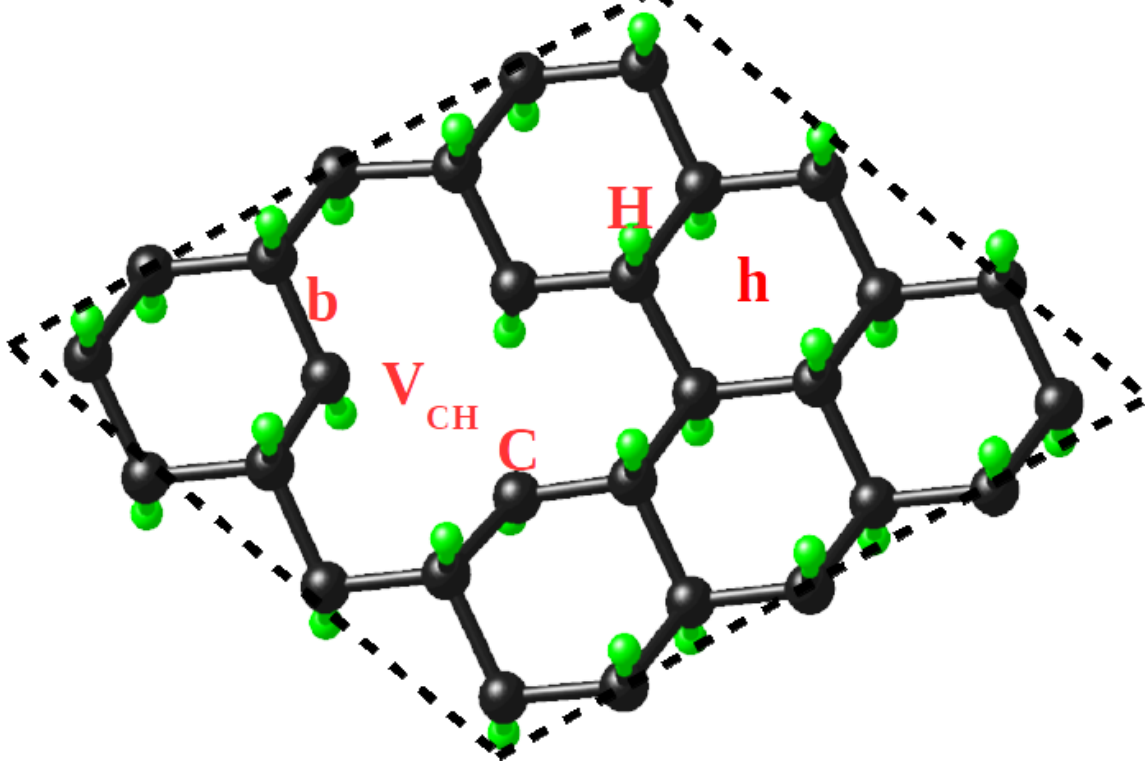
[2] Elias *et al*. Science. **323**, 610 (2009).

Electronic properties of V_{CH}



- V_{CH} defect induces spin up mid-gap states within the graphene band gap (half metallic behaviour)
- This is mainly contributed by the C 2p states as shown by PDOS
- As a consequence, the system becomes magnetic with the magnetic moment of $1 \mu_B$ [1]

Li above V_{CH} graphane



Different Li adsorption sites on a V_{CH} graphane.

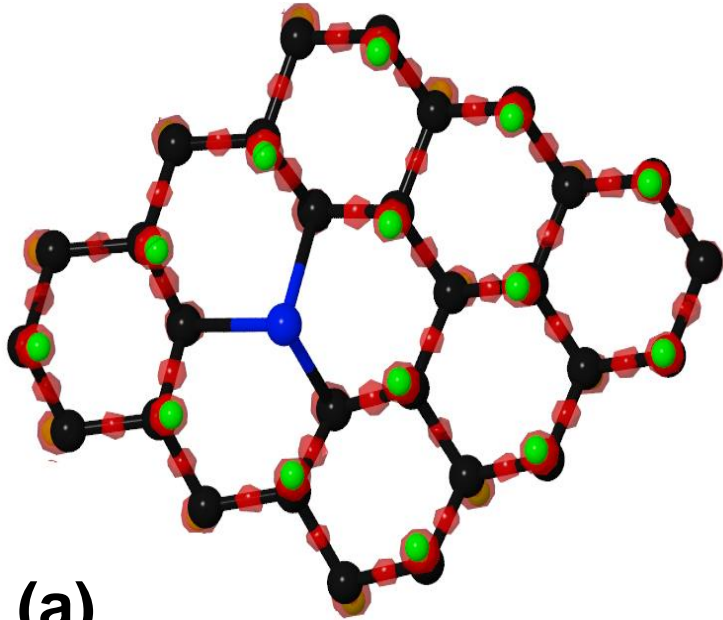
The binding energy E_B (eV), charge transfer q (e) and Li height d (Å) for various Li configurations.

Adsorption site	E_B (eV)	q (e)	d (Å)
Li- V_{CH}	1.72	0.87	1.54
Li-C	0.60	0.82	1.66
Li-h	0.58	0.96	1.70
Li-H	-1.19	0.19	4.19
Li-b	0.96	0.80	1.59

- Li- V_{CH} configuration gives the most thermodynamic stable structure
- Li- V_{CH} has the shortest value of d (Å)
- Li- V_{CH} bond strongly with the graphane substrate although ionically

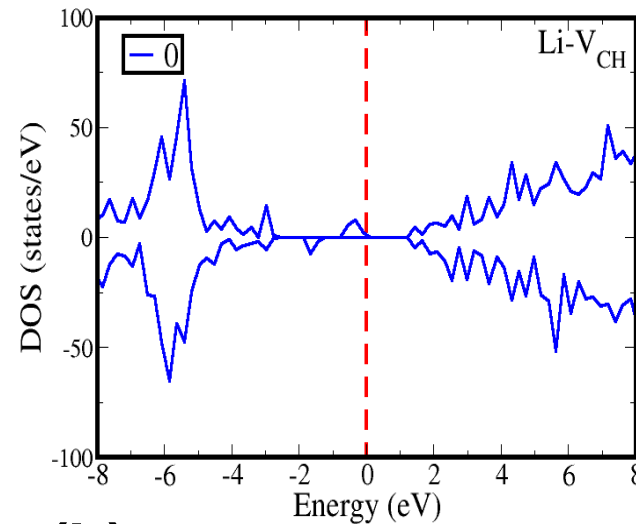
Electronic properties of Li-V_{CH}

Charge density difference ($\Delta\rho$)

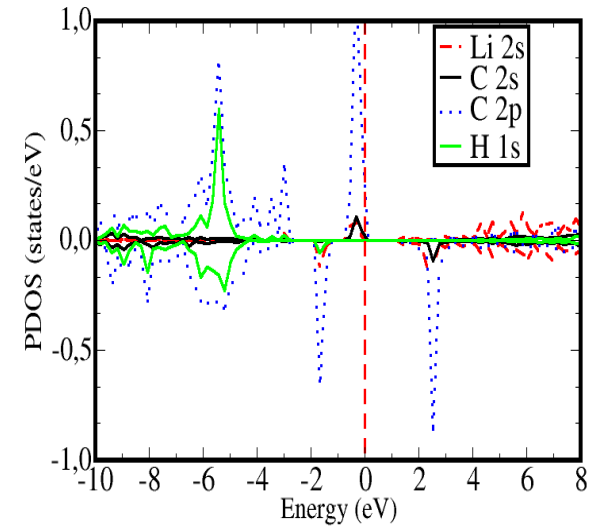


(a)

Density of states



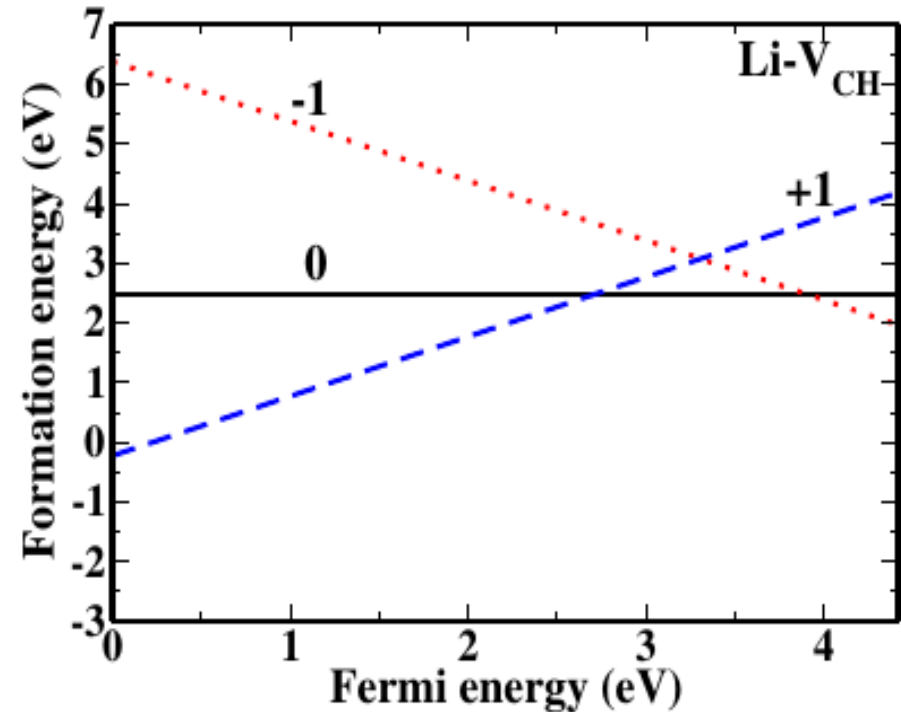
(b)



(c)

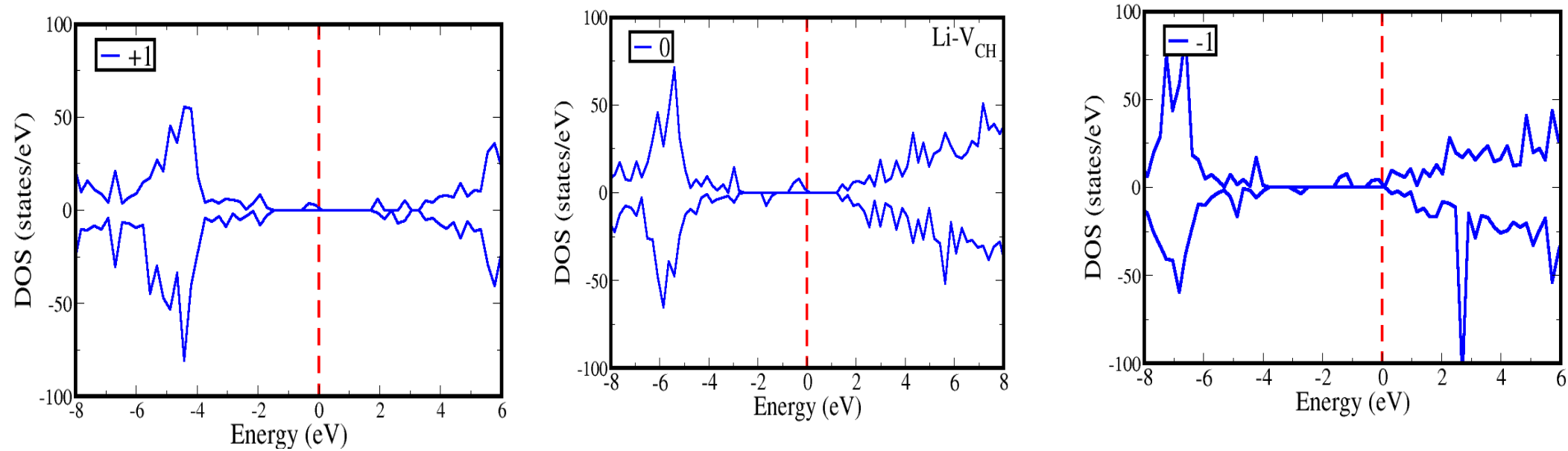
- The electronic charge densities are depleted from the C atoms, accumulated in C-C bonds
- Li atom at global minimum region
- Li mid-gap states are noted in Fig (b)
- Hybridization mainly contributed by Li 2s and C 2p

Thermodynamic stability of Li-V_{CH}



- At 0 charges state, the formation energy of Li-V_{CH} is 2.46 eV
- This is lower than that of V_{CH} defect of 6.05 eV
- The donor transition level $\epsilon(0/+1)$ is at 2.70 eV relative to VBM in the band gap
- Li-V_{CH} configuration may act as deep donor

Dependant of DOS for Li-V_{CH} on Charge states



- The Li ion enhances the V_{CH} induced magnetism
- The -1 charge doping shift the Fermi level toward the CBM further increasing magnetism.
- The +1 charge doping shifts the Fermi level toward the VBM reducing magnetism.
- Charge doping fine tunes the electronic and magnetic properties of Li-V_{CH}

Modification of the band offset in boronitrene

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¹Physics Department, University of Pretoria, Pretoria 0002, South Africa,
and ²National Institute for Theoretical Physics, Johannesburg, 2000, South Africa

(Dated: August 12, 2011)

Using density functional methods within the generalized gradient approximation implemented in the Quantum Espresso codes, we modify the band offset in a single layer of boronitrene by substituting a double line of carbon atoms. This effectively introduces a line of dipoles at the interface. We considered various junctions of this system within the zigzag and armchair orientations. Our results show that the "zigzag-short" structure is energetically most stable with a formation energy of 0.502 eV, and with a band offset of 1.51 eV. The "zigzag-long" structure has a band offset of 1.99 eV. The armchair structures are non-polar while the zigzag-single structures show a charge accumulation for the C-substituted B and charge depletion for the C-substituted N at the junction. Consequently there is no shifting of the bands.

PACS numbers: 61.48.Gh, 68.35.bg, 73.22.Pr

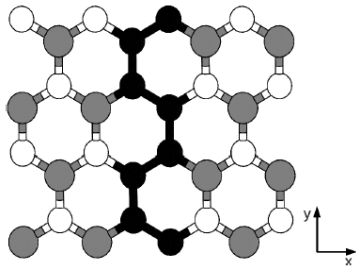


FIG. 1. The BN/C armchair-double structure showing a double line of C atoms. B atoms (●), N atoms (○) and C atoms (●). Because of the nonpolar lines comprising both B and N atoms parallel to the junction, no dipole is created at the junction by the double line of C atoms

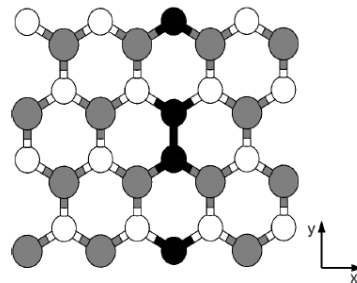


FIG. 2. The BN/C armchair-single structure showing a single line of C atoms. B atoms (●), N atoms (○) and C atoms (●). Because of the nonpolar lines comprising both B and N atoms parallel to the junction, no dipole is created at the junction by the single line of C atoms

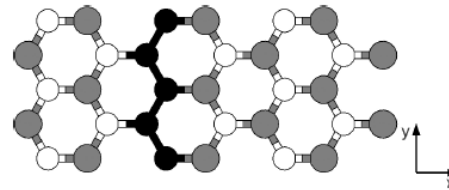


FIG. 3. The BN/C zigzag-short structure showing a double line of C atoms. B atoms (●), N atoms (○) and C atoms (●). Because of the alternating polar lines of B and N atoms parallel to the junction, a line of dipoles is created at the junction by the double line of C atoms

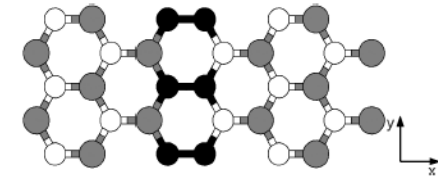


FIG. 4. The BN/C zigzag-long structure showing a double line of C atoms. B atoms (●), N atoms (○) and C atoms (●). Because of the alternating polar lines of B and N atoms parallel to the junction, a line of dipoles is created at the junction by the double line of C atoms

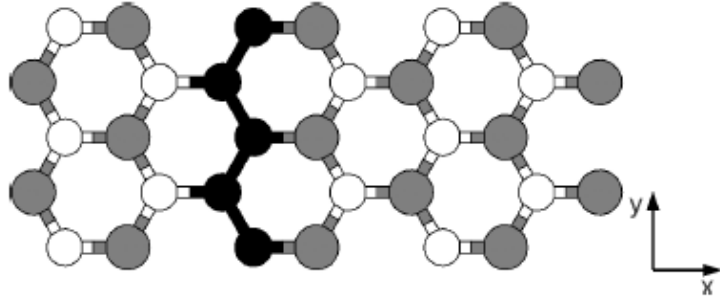


FIG. 3. The BN/C zigzag-short structure showing a double line of C atoms. B atoms (●), N atoms (○) and C atoms (●). Because of the alternating polar lines of B and N atoms parallel to the junction, a line of dipoles is created at the junction by the double line of C atoms

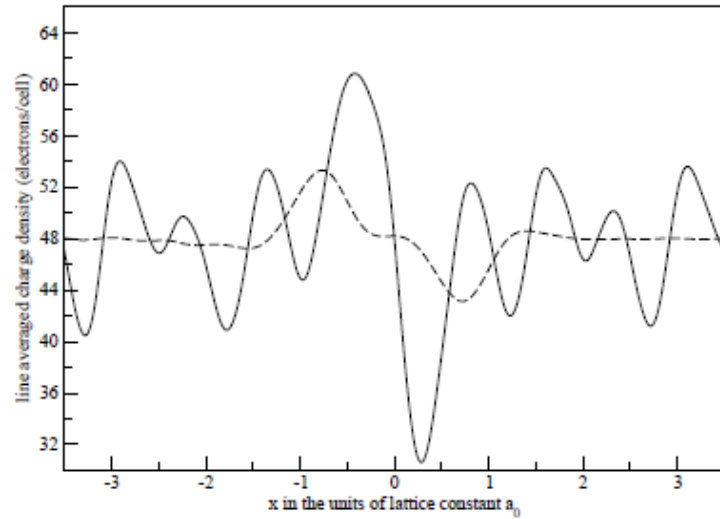


FIG. 9. The BN/C zigzag-short structure showing the dipole across the junction. The line-averaged charge density $\bar{n}(x)$ as a solid line and the macroscopic average $\tilde{n}(x)$ as a dashed line.

TABLE II. The values for the dipole (eV), electric field (10^9 V m^{-1}) and heat of formation ΔH_f (eV).

structure	ΔV_{gap}	E_{field}	ΔH_f
armchair-double	-	-	1.042
armchair-single	-	-	1.174
zigzag-long	1.99	5.568	1.764
zigzag-short	1.51	6.081	0.502

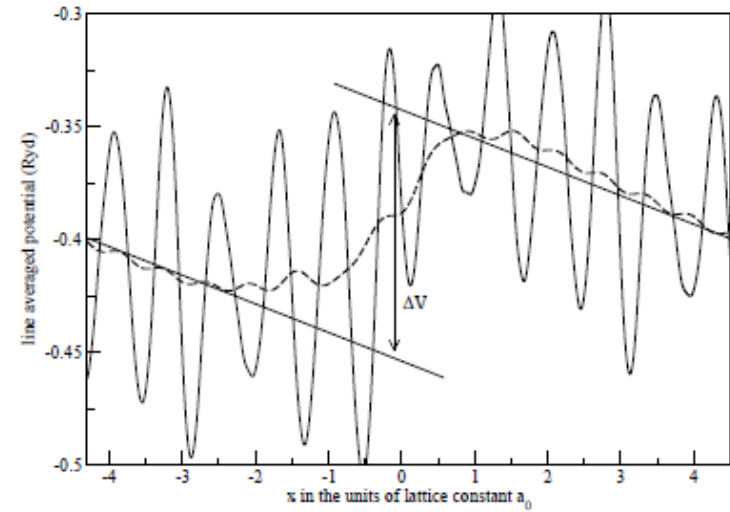


FIG. 10. The BN/C zigzag-short structure showing the band-offset. The line-averaged potential $\bar{V}(x)$ as a solid line and the macroscopic average $\tilde{V}(x)$ as a dashed line.

Mechanical properties of graphene and boronitrene

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We present an equation of state (EOS) that describes how the hydrostatic change in surface area is related to two-dimensional in-plane pressure (\mathcal{F}) and yields the measure of a material's resilience to isotropic stretching (the layer modulus γ) as one of its fit parameters. We give results for the monolayer systems of graphene and boronitrene, and we also include results for Si, Ge, GeC, and SiC in the isostructural honeycomb structure for comparison. Our results show that, of the honeycomb structures, graphene is the most resilient to stretching with a value of $\gamma_{\text{C}} = 206.6 \text{ N m}^{-1}$, second is boronitrene with $\gamma_{\text{BN}} = 177.0 \text{ N m}^{-1}$, followed by $\gamma_{\text{SiC}} = 116.5 \text{ N m}^{-1}$, $\gamma_{\text{GeC}} = 101.0 \text{ N m}^{-1}$, $\gamma_{\text{Si}} = 44.5 \text{ N m}^{-1}$, and $\gamma_{\text{Ge}} = 29.6 \text{ N m}^{-1}$. We calculate the Young's and shear moduli from the elastic constants and find that, in general, they rank according to the layer modulus. We also find that the calculated layer modulus matches the one obtained from the EOS. We use the EOS to predict the isotropic intrinsic strength of the various systems and find that, in general, the intrinsic stresses also rank according to the layer modulus. Graphene and boronitrene have comparable strengths with intrinsic stresses of 29.4 and 26.0 N m^{-1} , respectively. We considered four graphene allotropes including pentaheptite and graphdiyne and find that pentaheptite has a value for γ comparable to graphene. We find a phase transition from graphene to graphdiyne at $\mathcal{F} = -7.0 \text{ N m}^{-1}$. We also consider bilayer, trilayer, and four-layered graphene and find that the addition of extra layers results in a linear dependence of γ with \mathcal{F} .

The two-dimensional equivalent of bulk pressure is force per unit length (denoted \mathcal{F}) where an in-plane hydrostatic force causes a uniform change in area of the two-dimensional lattice. Force per unit length is expressed as the first derivative of the energy with respect to surface area:

$$\mathcal{F} = -\frac{\partial E}{\partial A} \quad (1)$$

and has units N m^{-1} . Positive \mathcal{F} represents a hydrostatic 2D compression while negative \mathcal{F} represents a uniform stretching. The two-dimensional equivalent of the bulk modulus, which we refer to as the layer modulus, is then defined as

$$\gamma = -A \frac{\partial \mathcal{F}}{\partial A}. \quad (2)$$

The bulk modulus represents the resistance of a bulk material to compression, whereas the layer modulus represents the resistance of a 2D material to stretching.

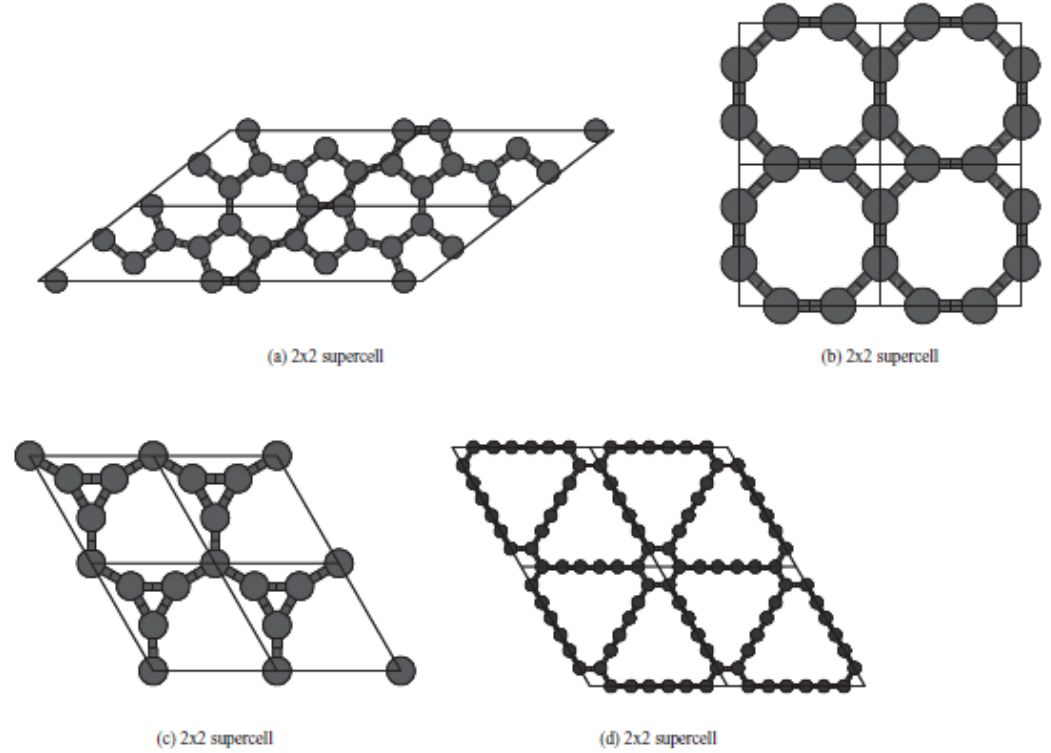


FIG. 1. The four graphene allotropes: (a) C1 (pentaheptite) consisting of pentagons and heptagons, (b) C2 consisting of squares and octagons, (c) C3 consisting of triangles and enneagons, and (d) C4 (graphdiyne) consisting of two acetylenic linkages between hexagons.

TABLE I. EOS fit parameters for honeycomb structures, graphene allotropes, and layered graphene (equilibrium area per atom pair A_0 in \AA^2 , lattice constants a and b , relaxed interlayer distance t and buckling parameter Δ in \AA , layer modulus γ_0 in N m^{-1} , γ'_0 dimensionless, γ''_0 in m N^{-1} , and cohesive energy per atom pair E_{coh} in eV).

	A_0	a	b	γ_0	γ'_0	γ''_0	E_{coh}
C	5.277	2.47		206.6	4.33	-0.0306	15.2
BN	5.468	2.51		177.0	4.37	-0.0454	13.8
SiC	8.303	3.10		116.5	4.79	-0.0688	11.2
GeC	9.068	3.24		101.0	5.33	-0.0722	9.3
Si	12.959	3.87 ^a		44.5	1.79	-0.6826	7.2
Ge	14.171	4.04 ^b		29.6	4.23	-1.5710	5.8
C1	5.480	7.48	4.75	192.3	4.35	-0.0338	14.7
C2	5.944	3.45		174.7	4.33	-0.0361	14.2
C3	6.399	3.84		153.2	4.55	-0.0511	13.8
C4	8.629	9.47		110.2	4.51	-0.0742	13.7
Four-graphene (LDA)	5.187	2.45 ^c		863.4	4.29	-0.0072	17.0
Three-graphene (LDA)	5.187	2.45 ^c		647.6	4.29	-0.0096	17.0
Two-graphene (LDA)	5.187	2.45 ^c		431.8	4.28	-0.0146	17.0
Graphene (LDA)	5.186	2.45		215.9	4.28	-0.0286	17.0

^a $\Delta = 0.45$.

^b $\Delta = 0.68$.

^cAtom relaxation for 2D EOS gave $t = 3.33$.

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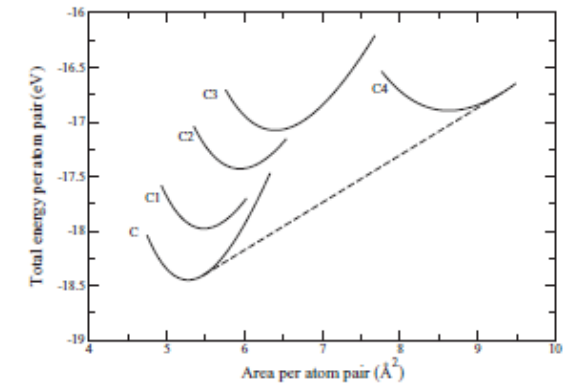


FIG. 5. EOS for graphene and four allotropes showing a phase transition from graphene to C4 at $\mathcal{F} = -7.0 \text{ N m}^{-1}$.

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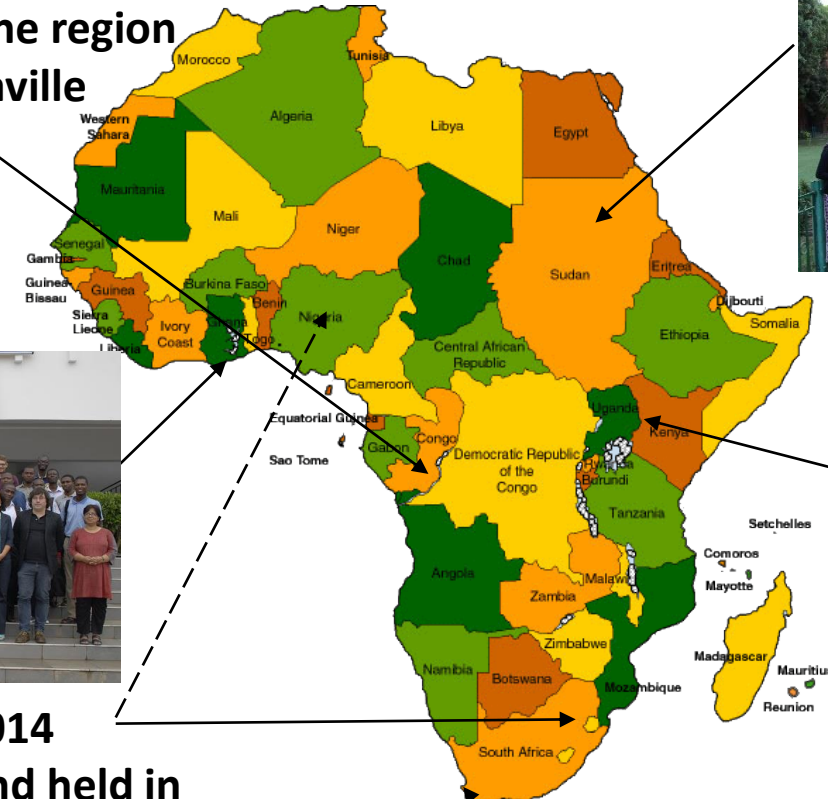
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