1 DOE Award: DE-FG02-08ER46512
Recipient Institution: Boston University

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PI: Antonio H. Castro Neto

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Year 3 of the project was until 09/01/2014, and since then the project has been at no-cost extension, with essentially no funds to spend.

The present report covers scientific progress (completed or started) during Year 3 (May 1, 2013 until 09/01/2014), and all publications that year as well as subsequent years where work was published on the basis of ideas and progress made that year, and therefore acknowledges the DOE grant in publications.

4 People working on the project and costs

Dr. Anand Sharma has been been supported by the grant as a research associate in years 2 and 3 (since 09/01/2012, until 07/01/2014), at 100% effort. Dr. Sharma's appointment was at the University of Vermont, on a sub-contract from Boston University. Dr. Sharma concluded his appointment on 07/01/2014, at which point the funds had run out. Prof. Valeri N. Kotov, at the University of Vermont, was Co-Investigator and PI on the sub-contract from BU. His effort was 0.5 summer salary in year 3 (2014). A detailed cost breakdown is available via the BU sponsored office: essentially in year 3 (9/1/13 – 8/31/14) the funds for that year were used for research associate salary at the sub-award (Univ. of Vermont) as well as summer salary for the PI and fringe expenses, pretty much exhausting the funds in the grant.

5 Accomplishments

The research in this program involves theoretical investigations of electronic, optical and mechanical properties of graphene and its derivatives, such as bi-layer graphene, graphene heterostructures, strained graphene, as well as graphene on various surfaces. One line of research has been development of theoretical models that support graphene's large array of possible technological applications. For example one of our goals has been the understanding of surface plasmons and spin relaxation mechanisms in graphene, related to novel optoelectronics and spintronics applications. Our current research focus is on understanding the role of correlations in graphene under mechanical deformations, such as strain. The main goal is to describe the mutual interplay

between strain and electron-electron interactions which could lead to the formation of novel electronic phases with strongly modified electronic, magnetic and optical properties. This direction of research contributes to deeper understanding of interactions in graphene - a subject at the forefront of research on graphene and its derivatives.

5.1 van der Waals Interactions, Atom-Graphene Membrane Interactions, Electron Correlations and Strain in Graphene

Long-range electron-electron interactions near the Dirac points can lead to profound effects, such as reshaping of the Dirac cones in suspended graphene as well as interaction-driven formation of "plasmarons" (i.e. quasiparticles bound to plasmons) in doped samples. The interplay of interactions and fermion anisotropy (e.g. due to strain) can lead to novel electronic phases with unconventional properties. It can also enhance significantly the van der Waals (VDW) interaction between two graphene layers (or one strained graphene layer and another material). This effect can be of great importance for graphene-based VDW heterostructures. In addition, interactions of adatoms with graphene, both elastic and inelastic, can depend significantly on strain, which could affect a variety of physical characteristics, such as properties of quantum gases and liquids on graphene (e.g. Helium) as well as quantum reflection properties and sticking rates of atoms to the graphene surface (important for sensors).

- We have discovered that uniaxial strain can enhance significantly the VDW interaction between graphene layers, which could be potentially explored in VDW heterostructure building. The role of electron-electron interactions and their renormalization also affects the VDW force by creating logarithmic corrections to the usual VDW force dependence - an affect we calculated first and which depends on the mutual interplay between interactions and strain. We find that the many-body renormalization contributions to the correlation energy are non-negligible and the vdW interaction energy decreases as a function of increasing distance between the layers due to renormalization of the Fermi velocity, the anisotropy, and the effective interaction. Our analysis can be useful in designing graphene-based VDW heterostructures which, in recent times, has seen an upsurge in research activity.

- We have also explored in detail the related problem of atom-graphene interactions and the effect of strain on those for a variety of atoms; this lead us to the concept of "adsorption by design". In this part of our research we aim to understand how the van der Waals force between neutral adatoms and a graphene layer is modified by uniaxial strain and electron correlation effects. A detailed analysis is presented for three atoms (He, H, and Na) and graphene strain ranging from weak to moderately strong. We show that the van der Waals potential can be significantly enhanced by strain, and present applications of our results to the problem of elastic scattering of atoms from graphene. In particular, we find that quantum reflection can be significantly suppressed by strain, meaning that dissipative inelastic effects near the surface become of increased importance. Furthermore, we introduce a method to independently estimate the Lennard-Jones parameters used in an effective model of He interacting with graphene, and determine how they depend on strain. At short distances, we find that strain tends to reduce the interaction strength by pushing the location of the adsorption potential minima to higher distances above the deformed

graphene sheet. This opens up the exciting possibility of mechanically engineering an adsorption potential, with implications for the formation and observation of anisotropic low-dimensional superfluid phases.

- Inelastic properties of adatoms approaching the surface of graphene are governed by interactions with flexural phonons at finite temperature ("hot graphene membrane"). Here we have explored in great depth a variety of issues, starting from the infrared dynamics of low-energy atoms interacting with a sample of suspended graphene at finite temperature. The dynamics exhibits severe infrared divergences order by order in perturbation theory as a result of the singular nature of low-energy flexural phonon emission. Our model can be viewed as a two-channel generalization of the independent boson model with asymmetric atom-phonon coupling. This allows us to take advantage of the exact non-perturbative solution of the independent boson model in the stronger channel while treating the weaker one perturbatively. In the low-energy limit, the exact solution can be viewed as a resummation (exponentiation) of the most divergent diagrams in the perturbative expansion. As a result of this procedure, we obtain the atoms Green function which we use to calculate the atom damping rate, a quantity equal to the quantum sticking rate. A characteristic feature of our results is that the Greens function retains a weak, infrared cutoff dependence that reflects the reduced dimensionality of the problem. As a consequence, we predict a measurable dependence of the sticking rate on graphene sample size. We provide detailed predictions for the sticking rate of atomic hydrogen as a function of temperature and sample size.

- We have studied the influence of uniaxial strain on the ferromagnetic exchange instability in graphene and have found that itinerant magnetism can occur at much smaller (strain-dependent) value of the electron-electron interactions, relative to the unstrained isotropic graphene. This can lead to magnetism, with rather large magnetization, in strongly anisotropic graphene-based systems.

- We have also published a major review article on electron-electron interactions in graphene in Reviews of Modern Physics. It reviews research on all aspects of electron correlation physics in single and bi-layer graphene.

5.2 Magnetic Moments, Orbital Currents and designer Spin/Orbital Liquids in Graphene-based Systems and Artificial Honeycomb Mott Lattices

This part of our research program deals with the conditions for formation of localized as well as itinerant magnetic moments under different geometries and applied strain as well as spin relaxation mechanisms relevant to spintronics applications. All these are effects induced by electron correlations. We explore a variety of scenarios for creation of localized magnetic and valley moments, and lattices of those, including the possibility of macroscopic magnetic and valley (loop current) order, for example in graphene on hexagonal boron nitride (h-BN) substrate, which exhibits a triangular superlattice of nearly circular mass defects. This is an unconventional state of matter similar to anomalous Hall materials. We have also proposed a novel way to create

artificial, graphene-based Mott insulators, by arranging lattices of Coulomb impurities on top of graphene on SiC substrate (which induces a gap in graphene). Such arrangements can lead to a promising route for observation of a spin-orbital (i.e. spin-valley) quantum liquid – a truly new state of matter.

- We have investigated the phase diagram of the Anderson impurity model describing localized magnetic states, due to electron correlation, on adatoms in strained graphene. Detailed predictions were made for the values of the Hubbard U and strain necessary to generate magnetic moments.

- We have examined in detail the role of Coulomb interactions in the emergence of macroscopically ordered states in graphene supported on hexagonal boron nitride (h-BN) substrates. Due to incommensuration effects with the substrate and interactions, graphene can develop gapped low-energy modes that spatially conform into a triangular superlattice of quantum rings. In the presence of these modes, we show that Coulomb interactions lead to spontaneous formation of chiral loop currents in bulk and to macroscopic spin-valley order at zero temperature. We show that this exotic state breaks time-reversal symmetry and can be detected with interferometry and polar Kerr measurements.

- Quantum spin-orbital liquids are elusive states of matter in which quantum fluctuations of orbitals and spins conspire to create strongly correlated states which do not break any symmetries. A promising development in the observation of those states would be the creation of artificial Mott insulators where antiferromagnetic correlations between spin and orbital degrees of freedom can be designed. Here we propose to exploit the fact that massive Dirac fermions form bound states with spin and valley degrees of freedom in the vicinity of a Coulomb impurity. In the presence of electron-electron interactions, the interaction of the spin and valley polarized electrons bound to different Coulomb impurities naturally maps into a superexchange interaction with SU(4) symmetry. We propose that quantum spin-orbital liquids can be engineered in artificial Coulomb impurity lattices on the surface of gapped honeycomb substrates, such as graphene on SiC. This could open up a new research direction, in a solid-state setting, for creation and detection of artificial quantum liquids which are truly new quantum states of matter.

6 Publications

(excerpts of acknowledgement sections also included)

"Designing Quantum Spin-Orbital Liquids in Coulomb Impurity Lattices on Gapped Honeycomb Substrates," by Xu Dou, Valeri N. Kotov, and Bruno Uchoa, arXiv:1602.01477, submitted and currently under consideration in Nature Scientific Reports.

We are grateful to Kevin Beach, Frederic Mila and K. Mullen for numerous stimulating discussions. X. Dou and B. Uchoa acknowledge the University of Oklahoma for partial support. B. Uchoa was supported by NSF CAREER grant NMR-1352604. V. N. Kotov acknowledges support by the U.S. Department of Energy (DOE) grant DE-FG02-08ER46512. B. Uchoa thanks the Aspen Center of Physics where this work was partially completed.

"Infrared Dynamics of Cold Atoms on Hot Graphene Membranes," by Sanghita Sengupta, Valeri N. Kotov, and Dennis P. Clougherty, arXiv:1603.03476, accepted in Physical Review B (2016), to appear (Editor Code: BR12706).

The research of V. N. Kotov was supported by the U.S. Department of Energy (DOE) grant DE-FG02-08ER46512.

"Adsorption by design: Tuning atom-graphene van der Waals interactions via mechanical strain," by Nathan S. Nichols, Adrian Del Maestro, Carlos Wexler, and Valeri N. Kotov, Physical Review B **93**, 205412 (2016).

We are grateful to D. Clougherty for numerous stimulating discussions related to the subject of this work and we acknowledge M. Cole for his insights into the physics of adsorption. The research of V. N. Kotov was supported by the U. S. Department of Energy (DOE) Grant No. DE-FG02-08ER46512.

"Valley order and loop currents in graphene on hexagonal boron nitride," by Bruno Uchoa, Valeri N. Kotov, and M. Kindermann, Physical Review B **91**, 121412(R) (2015).

V.N.K. was supported by US DOE Grant No. DE-FG02-08ER46512, and M.K. by NSF Grant No. DMR-1055799.

"van der Waals forces and electron-electron interactions in two strained graphene layers," by Anand Sharma, Peter Harnish, Alexander Sylvester, Valeri N. Kotov, and A. H. Castro Neto, Physical Review B **89**, 235425 (2014).

We are grateful to D. Clougherty and A. Del Maestro for numerous stimulating discussions on the subject of dispersion forces. The research of A. Sharma, V. N. Kotov, and A. H. Castro Neto was supported by the U.S. Department of Energy (DOE) Grant No. DE-FG02-08ER46512. A. Sylvester acknowledges financial assistance from the Research Experiences for Undergraduates (REU) Program of the National Science Foundation (No. DMR-1062966).

"Effect of uniaxial strain on ferromagnetic instability and formation of localized magnetic states on adatoms in graphene," by Anand Sharma, Valeri N. Kotov, and Antonio H. Castro Neto, Physical Review B **87**, 155431 (2013).

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