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In this project, we designed and engineered hetero-interfaces of 2-dimensional (2D) van der Waals (vdW) materials for the realization of novel quantum electronic states. We employed molecular beam epitaxy (MBE) combined with nanofabrication techniques to form functional nanostructures that can be utilized for new electronic device applications. We have demonstrated growth of topological insulator, Bi2Te3 on the surface of hBN single crystals by MBE method. We also use transmission electron microscopy (TEM) analysis for the structural of the atomically sharp interface between hBN and Bi2Te3. Finally, we have developed unprecedentedly clean graphene supercoductor junctions that allowed to study transport across the van der Waals interface between the conductor and superconductor. Our observation of gate tunable transitions between retro intraband and specular interband Andreev reflections opens a new route for future experiments that could employ the gate control of the Cooper pair injection process.

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NBIT Final Report for AOARD Grant 134122

"Nano Electronics on Atomically Controlled van der Waals Quantum Heterostructures"

3/30/2015

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Abstract: In this project, we designed and engineered hetero-interfaces of 2-dimensional (2D) van der Waals (vdW) materials for the realization of novel quantum electronic states. We employed molecular beam epitaxy (MBE) combined with nanofabrication techniques to form functional nanostructures that can be utilized for new electronic device applications. We have demonstrated growth of topological insulator, Bi2Te3 on the surface of hBN single crystals by MBE method. We also use transmission electron microscopy (TEM) analysis for the structural of the atomically sharp interface between hBN and Bi2Te3. Finally, we have developed unprecedentedly clean graphene supercoductor junctions that allowed to study transport across the van der Waals interface between the conductor and superconductor. Our observation of gate tunable transitions between retro intraband and specular interband Andreev reflections opens a new route for future experiments that could employ the gate control of the Cooper pair injection process.

Introduction: The biggest challenge of nanoelectronic device development is reducing the power consumption in current electronic devices. Necessity of novel quantum electronic device applications has been called up for low-power, high-speed, high-density electronic application and for a new route for novel quantum computing, where quantum device operation may play a significant roles. For developing novel electronics, one also require a new material platforms. One of the most exciting recent scientific discoveries has been the emergence of a new class of low dimensional materials. The ultimate 2-dimensional (2D) limit of a layered materials systems has attracted intense research focus. In these systems, weak van der Waals (vdW) forces hold the layers together. This new class of materials, termed van der Waals materials, includes graphene, hexagonal boron nitride (h-BN), and transition metal chalcogenides. Together these materials represent various electronic systems, including semiconductors, metals, superconductors, and topological insulators. Our research efforts have been largely built upon the possibility of creating these new materials platform.

Designing and building metastable structures with interfacial heterojunctions between vdW atomic layers allowed us to build novel nanoscale engineered devices. In order to acquire precise atomic controllability of the vdW interface, we use controlled engineered materials growth approach using MBE on vdW substrate such as hBN. We also have been focusing on the electron transport across the hetero-interface between two dissimilar materials. Electron transport in two particular vertical functional structures has been explored: (i) atomically sharp interfaces of planar superconductor/VdW conductor such as NbSe2/graphene interface and (ii) topological insulator grown on hBN.

Experiment:

<u>vdW epitaxial growth of TI on hBN.</u> We use molecular beam epitaxial growth to obtain high-quality Bi_2Se_3 thin films on h-BN layers and their structural and electrical characteristics. Both large-area chemical vapor-deposited (CVD) single-crystalline h-BN layers and h-BN microflakes mechanically cleaved from a bulk single crystal were used as substrates. The Bi_2Se_3 thin films with atomically smooth terraces over a large area were obtained by employing two-step growth method. Further, we investigated epitaxial relationship and structural properties of the Bi_2Se_3 films on h-BN layers using high-resolution transmission electron microscopy. In this study, we report on the molecular beam epitaxial growth of Bi_2Se_3 thin films on h-BN and their structural quality examined by high-resolution transmission electron microscopy (HR-TEM). Bi_2Se_3 thin films were grown on h-BN layers using a custom-built MBE system dedicated to $(Bi_xSb_{1-x})_2(Se_yTe_{1-y})_3$ materials. The base pressure of the growth chamber was in the range of 10^{-10} Torr. High purity Bi (99.999%) and Se (99.9999%) fluxes were provided by Knudsen cells and measured using a quartz crystal microbalance. The growth was carried out under Se-rich conditions with a typical Se/Bi flux ratio of ~15. The growth rate was maintained at ~0.3 quintuple layer (QL)/min, solely determined by the Bi flux. The typical film thickness was 6-10 QLs. For substrate preparation, h-BN layers were transferred by the mechanical exfoliation technique either onto SiO_2/Si substrates or TEM compatible chips. Prior to Bi_2Se_3 growth, thermal cleaning was carried out at 400°C for 30 min in an ultrahigh vacuum.

The morphological and microstructural characteristics of Bi_2Se_3 films on h-BN were investigated using field-emission scanning electron microscopy (FE-SEM), atomic force microscopy (AFM), and HR-TEM. For plan-view TEM observations, the h-BN transferred onto a holey carbon TEM grid or a SiN_x/Si membrane chip was employed as a substrate.[13] Samples for cross-sectional TEM imaging were prepared by a focused ion beam machine. A 200 kV field-emission TEM (JEOL JEM-2100F) was used for selected-area electron diffraction (SAED), bright field (BF) imaging, and high-resolution imaging.

<u>Specular Andreev Reflection across the vdW interfaces.</u> Electrons incident from a normal metal onto a superconductor are reflected back as holes- a process called Andreev reflection. In a typical metal with larger Fermi energy than superconducting gap, the reflected hole is retraces the path taken by the incident electron. In graphene with low disorder, the Fermi energy can be tuned to be smaller than the superconducting gap. In this unusual limit, the holes are expected to be reflected specularly at the superconductor-graphene interface due to the onset of interband Andreev processes where the effective mass of reflected holes change sign. In this experiment we present measurements of gate modulated Andreev reflections across the low disorder van der Waals interface formed between graphene and the superconducting NbSe2. We find that the conductance across the graphene-superconductor interface exhibits a suppression when the Fermi energy is tuned to values smaller than the superconducting gap, a hallmark for the transition between intraband retro- and interband specular- Andreev reflections.

In this project, we employ a novel non-invasive approach to fabricate normal metal (N) and superconductor (SC) interfaces with an unprecedent energy resolution close to the the neutrality of graphene. For this purpose we electronically couple a high mobility hBN/ bilayer graphene (BLG) device and a 20-100 nm thin NbSe2 flake. NbSe2 is a van der Waals SC with a critical temperature Tc~7K and a large energy gap. We use the dry-vdW transfer technique and a current annealing method to achieve ultra-clean, atomically sharp, and highly transparent Ohmic interfaces between graphene and NbSe2. The devices were fabricated on heavily degenerated Si substrates topped with 300 nm SiO2, where a back gate voltage Vbg is applied to tune the Fermi energy of the BLG channel.

Results and Discussion:

vdW epitaxial growth of TI on hBN. The crystalline quality of the optimized film was examined by TEM. The SAED patterns obtained from the area highlighted in the inset of Fig. 1(a) with a selected area aperture size of ~1.3 μm is shown in Fig. 1(a). It revealed that the formation of undesirable defects such as low-angle grain boundaries were significantly suppressed. The statistical distribution of the in-plane alignment of Bi2Se3 thin films was estimated as a standard deviation width narrower than ±1° in the SAED intensity profile of Bi2Se3. The interface of the Bi2Se3/h-BN heterostructure was investigated by cross-sectional HR-TEM. The TEM image of Fig. 1(b) shows the clear layered structures and atomically abrupt interface in Bi2Se3/h-BN. From the image, the lattice spacings between adjacent planes were measured to be 0.96 and 0.34 nm in the Bi2Se3 thin film and h-BN layer, corresponding to the d-spacings of Bi2Se3 (0003) and h-BN (0002), respectively. Importantly, even though the two-step growth method was employed, the film had uniform crystalline quality down to the first quintuple layer without any amorphous layer or other interfacial layers, forming atomically sharp interface with the underlying h-BN layers. Moreover, no extended crystal defects, such as threading dislocations and stacking faults, could be observed at the interface through high-resolution analyses, presumably due to stress relaxation through the noncovalent vdW heterointerface. All the microstructural features showed the high structural quality of Bi2Se3 thin films grown on h-BN layers.

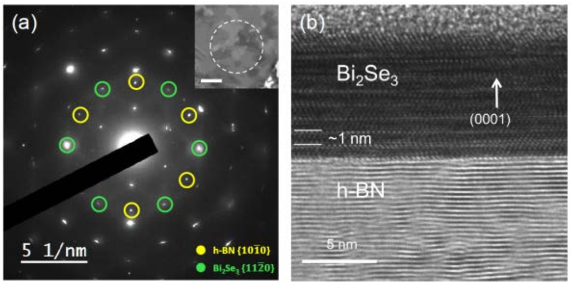

FIG. 1. Structural properties of two-step grown Bi2Se3 thin films on h-BN layers with the optimal conditions. (a) SAED patterns obtained from the area shown in the inset (scale bar, 500 nm). (b) Cross-sectional HR-TEM image of the Bi2Se3 thin films.

In this experiment, the Bi2Se3 thin films were grown on h-BN layers using molecular beam epitaxy in combination with the electron microscopic observations. The surface morphology and structural quality of the films depended strongly on the growth temperature. Furthermore, Bi2Se3 thin films with atomically smooth terraces over a large area have been obtained by employing two-step growth method. Most importantly, by using h-BN as both a substrate for the thin film growth and a supporting layer for TEM measurements, microstructural defects such as low-angle grain boundaries in the films could be readily visualized, providing useful feedbacks for growth. The optimized samples exhibited epitaxial formation of high structural quality Bi2Se3 thin films on h-BN with reduced concentration of structural defects and abrupt and clean heterointerfaces. More generally, we believe that the experimental scheme demonstrated in this report could be a promising method for preparation of high quality Bi2Se3 thin films and readily expaned to many other heterostructures based on 2D layerd materials.

Specular Andreev Reflection across the vdW interfaces.

The ability to tune Fermi energy enables us to investigate the characteristic Andreev reflection (AR) signal at the continuous transition from large to small Fermi energy. Fig. 2 (a) shows the characteristic longitudinal resistance Rxx of the BLG channel vs. Vbg. An upper bound of Fermi energy variation 1 meV was estimated from the full-width-half-maximum (FWHM) peak of Rxx(Vbg). Fig. 2(b) displays simultaneous measurements of normalized differential conductance as a function of both the bias current and Vbg. For large bias, the normalized conductance exhibits a similar characteristic behaviour, showing the characteristic conductance dip around the zero bias condition. However near the charge neutrality point (CNP), the conductance traces exhibit drastic variations: the conductance dip at zero bias turns into a peak and then is suppressed back again as Fermi energy goes through the CNP. While overall the experimental findings are in qualitative agreement with the theoretical estimates, there are certain quantitative mismatches. Notably, the experimental conductance map has an overall higher conductance with broadened features. In addition, we observe a pronounced region of reduced conductance around for a wide range of Fermi energy that is not present in the theoretical map. All these deviations can be attributed to imperfections due to a realistically broadened N/SC interface, inelastic scattering at finite temperatures and the presence of small potential fluctuations, whose quantitative descriptions go beyond the scope of our simple theoretical model.

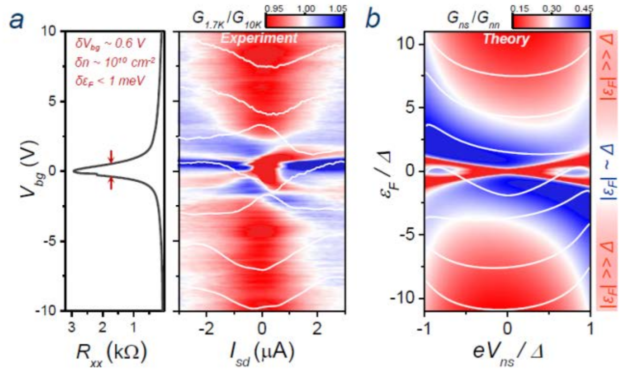

FIG. 2. Gate tunable Andreev reflections. (a) Left : Rxx vs. Vbg showing the characteristic channel resistance peak at the CNP with a full-width of half-maximum. Right : color map of normalized differential conductance as a function of bias current and the gate voltage with line-traces of the conductance for various fixed gate voltages. (b) Color map of the theoretically obtained normalized differential conductance allows to continuously probe the conductance changes.

In this study we have developed unprecedentedly clean BLG/NbSe2 based N/SC junctions that allowed to study ARs at low Fermi energy. Our observation of gate tunable transitions between retro intraband and specular interband ARs opens a new route for future experiments that could employ the gate control of critical angle, which can be continently and independently altered with gate and bias voltage. Most importantly our finding help to draw a general picture of the exact physical processes underlying ARs.

List of Publications and Significant Collaborations that resulted from your AOARD supported project: In standard format showing authors, title, journal, issue, pages, and date, for each category list the following:

a) papers published in peer-reviewed journals,

"Selective excitation of Fabry-Perot or whispering-gallery mode-type lasing in GaN microrods," Hyeonjun Baek, Jerome K. Hyun, Kunook Chung, Hongseok Oh, and Gyu-Chul Yi, Applied Physics Letters 105, 201108 (2014)

b) papers published in peer-reviewed conference proceedings,

None

c) papers published in non-peer-reviewed journals and conference proceedings,

None

d) conference presentations without papers,

- Winter Workshop on Quantum Materials in Condensed Matter Physics, Seoul Korea (2015)
- International Workshop on Field-Effect Transistors and Functional Interfaces, Kashiwa, Japan (2014)
- 7th International Conference on Molecular Electronis, Strasbourg, France (2014)
- Pincare Seminar on Dirac Matter, Paris, France (2014)
- Materials Science Society Meeting, San Francisco (2014)

e) manuscripts submitted but not yet published, and

"Specular Interband Andreev Reflections in Graphene," D. K. Efetov, L. Wang, C. Handchin, K. B. Efetov, J. Shuang, R. Cava, T. Taniguchi, K. Watanabe, J. Hone, C. R. Dean, and P. Kim, submitted to Nature Physics (2015)

Attachments: Publications a), b) and c) listed above if possible.

DD882: As a separate document, please complete and sign the inventions disclosure form.