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2021 2023

Honeycomb nitrides layers as playground for Xenes formation

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SiC(0001)

Sn Sb Pb Bi   (Xene)   2D

Xene

The data obtained in during this research period has proven the concept of using nitride layer on SiC as a base for development of group IV and V Xene 2D materials. Further developments of the topic are expected in the future which may prove useful in the field of spintronics.

A single silicon nitride layer on SiC(0001) with honeycomb dangling bonds arrangement has been successfully grown and characterized by variety of surface science techniques. This nitride layer is an interesting material itself promising a lot of future applied and scientific potential. We have also carried out extensive computational study of possibility of honeycomb lattice 2D layers consisting of Sn, Sb, Pb, and Bi atoms (Xenes). Calculations confirms high probability to synthesize such layers on nitride buffer, confirming suggested significant prospects of such approach. The Xene layers made of heavy elements exhibit unique spin-polarized electronic structure. Experiments will be done in future continuing the progress achieved during this project period.

Surface physics

2D materials semiconductors spin-polarization Dirac materials Xenes Silicon carbide

１．研究開始当初の背景

Graphene opened a new chapter in physics of 2D-materials. The fascinating properties of graphene, such as massless Dirac fermion nature of valence electrons, quantum Hall effect and others, in reality are attributed to the very simple fact of honeycomb structural symmetry of graphene's atomic lattice. Seemingly simple tight-binding prediction model when includes spin-orbit interaction (SOI) and strong on-site interaction results in fascinating physical outcomes such as Mott insulator transition, topological insulator phase transition, spin liquid state and spin-polarized edge currents as shown in Kane-Mele-Hubbard model. Most of these interesting and potentially practically useful physics is, however, is not realized in graphene owing to very small SOI of carbon atoms. Honeycombs made of heavier elements (called Xenes) are of particular interest. Experiments show, however, that these materials are unstable in free form, and it is hard to find suitable substrate for synthesis of such materials. There is, however, a candidate to resolve this issue, namely, $Si2N_3$ monolayer on SiC(0001) surface, which offers a unique opportunity for the growth of heavy element Xenes as has been suggested based on preliminary calculations. The purpose of our research was to investigate fully the properties of this unique overlayer, as it represents a very interesting system itself, including Mott insulating state. Then use it to synthesize heavy element Xenes and their nanostructures and investigate their properties.

２．研究の目的

The purpose of our research was to find a way to grow epitaxial silicon nitride (Si_2N_3) buffer layer on SiC(0001) and explore it's potential for synthesis of various elements Xenes possessing interesting physical properties. The following goals has been postulated:

- Prepare laboratory environment for carrying out the research on the topic;
- Optimize Si_2N_3 growth on SiC for quality;
- Characterize properties of Si_2N_3 layer by experimental studies and by calculations;
- Attempt to grow Xenes layers composed of Pb, Sb, Bi, Sn, and Tl elements;
- Characterize the Xenes layers experimentally and by calculations;
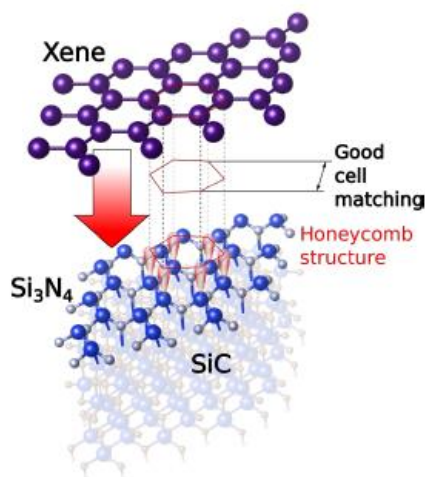- Experiment with various substrate surface morphologies and nanostructured materials.

Fig. 1. Epitaxial Xenes growth on silicon nitride on SiC.

３．研究の方法

To reach the goals both we were intended to use both experimental and computational approaches. To investigate if the suggested structures could be grown in principle, the extensive *ab initio* calculations (DFT method) have been performed investigating the stability of honeycomb structures of metal layers on Si_2N_3 vs rectangular and other arrangements. The electronic structure calculations have also been performed to make sure that interesting electronic properties are indeed expected and where to look in the experimental data to see them.

The experimental part had to be carried out using a broad set of surface science characterization techniques such as low-energy electron diffraction (LEED) for structure determination, atomic force microscopy (AFM) and scanning tunneling microscopy (STM) for surface morphology and atomic level topology investigations, as well as chemical analysis using X-ray photoemission spectroscopy and electronic structure observations using angle-resolved photoemission spectroscopy (ARPES). Large variety of experimental analysis implied extensive collaboration with other laboratories.

４．研究成果

During our study silicon nitride layer on SiC(0001) has been successfully grown and characterized. Atomic model with honeycomb arrangement of dangling bonds which was suggested to favor growth of Xenes has been confirmed both by LEED analysis and STM measurements (Fig. 2(a,b)). Spin-resolved DFT calculations have shown, surprisingly that nitride layer is an insulator with ferromagnetic dangling bond electron spin arrangement (Fig. 2(d)). Such a unique for 2D material spin behavior might be very interesting when applied to Xenes materials on-top of it. Unfortunately, more detailed analysis of spin dependent electronic properties proved to be hard to investigate in calculation due to the lack of computation power of existing equipment.

Moreover, experiments show that another ordered Si cluster-like surface can be grown by further increasing Si concentration on the surface (Fig. 2(c)). The details of this new surface phase will be further investigated in following studies.
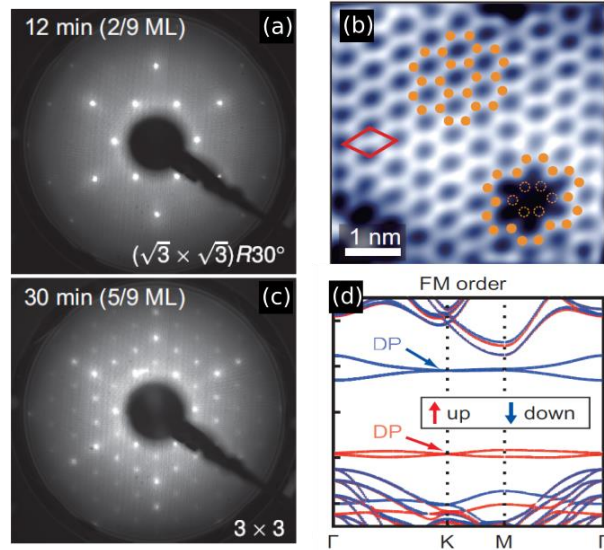

Fig. 2. LEED pattern (a) and STM image (b) of honeycomb nitride layer; (c) new Si cluster-on-nitride (3x3) structure LEED pattern; (d) band structure of honeycomb nitride with ferromagnetic spin dangling bonds.
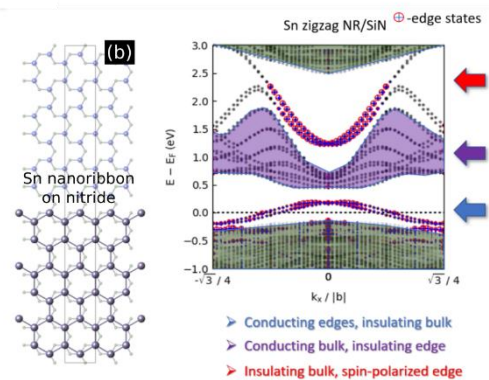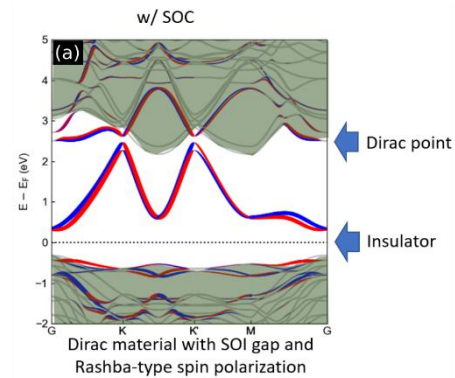
The extensive DFT modelling, and calculations show that Xenes consisting of Sn, Sb, Pb, Bi are stable in honeycomb configuration on Si_2N_3, unlike on bare SiC surface. The investigation of electronic structure show, that such Xenes, although not freestanding, exhibit very unique electronic structure with Dirac cones affected by rather noticeable Rashba-type spin polarization on inclusion of SOI into calculation. These makes them interesting candidates for spintronic devices. Calculations of Xenes nanoribbons show the possible topological edge states appearing (Fig. 3).

Unfortunately, owing to COVID restrictions in the initial stages of the project and some serious complications with equipment working conditions, experimental part of the Xenes growth on the nitride layer has not been successfully performed during research project period. However, we did apply a significant amount of resources and workhours to meaningfully improve our research environment to continue study on this very interesting topic even after supporting project period is finished. Our initial experiments show a lot of promise, and we will be continuing research to fulfill our initial goals.

Fig. 3. DFT study on stanene (Sn Xene) on honeycomb silicon nitride layer. (a) spin-resolved band strucutre, (b) calculation of stanene nanoribbon electronic states.

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Currently two manuscripts are being prepared for publications on the research results. One is in the structure determination and characterization of novel 2D honeycomb silicon nitride layer on SiC(0001). The other one is the result of computational work on Xenes/Si2N3/SiC structure stability and electronic bands properties.

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