Novel Materials

Room Swan BC - Session NM-WeM2

Topological Insulators

Moderator: Kunal Mukherjee, Stanford University

10:30am **NM-WeM2-11 Structure-Property Relationship of the Magnetic Properties of Molecular Beam Epitaxy Grown $(Sb_2Te_3)_{1-x}(Mnsb_2Te_4)_x$- Magnetic Topological Insulators**, ***Ido Levy***, City College of New York, City University of New York; *C. Forrester,* Graduate Center of CUNY and CIty College of New York and Lehman College; *X. Ding, K. Wickramasinghe,* City College of New York; *C. Testelin,* Sorbonne Université, CNRS; *D. Smith,* Arizona State University; *L. Krusin-Elbaum,* City College of New York, City University of New York; *M. Tamargo,* City College of New York

Magnetic topological insulators such as $MnBi_2Te_4$ and $MnSb_2Te_4$ are of great interest due to their promise for realizing exotic physical phenomena and for potential applications in quantum science and quantum computing. However, the control and understanding of the materials properties are still far from adequate. Molecular beam epitaxy (MBE) promises higher control and improved materials properties due to its far from equilibrium conditions and its layer-by-layer growth. Addition of Mn during growth of Sb_2Te_3 results in the formation $MnSb_2Te_4$ septuple layers (SL). Although the individual SLs are ferromagnetic (FM), when SLs stack to form a crystal, their spins align opposite each other, resulting in net antiferromagnetic properties, not conducive to the desired exotic phenomena. It has also been shown that depending on the amount of Mn added, the structure self-assembles into layered QL:SL stacks of the form $(Sb_2Te_3)_{1-x}(MnSb_2Te_4)_x$.

Our group is investigating the MBE growth of the $MnSb_2Te_4$ system. We recently reported the growth of layered QL:SL structures ranging in composition from 100% QL to 100% SLs**.**We showed that modification of the growth conditions by incorporation of a preannealing step and increased growth temperatures leads to samples with varying amounts of SL for the same Mn flux ratios. This suggests that Mn incorporates both as a structural component in SLs and as antisite defects, affecting the magnetic properties of the resulting structures.

Here we present the detailed magnetic properties of the materials using Hall Effect and remanent magnetization measurements. We show that under our MBE growth conditions, all layers with at least a few SLs are FM. Furthermore, their Curie temperatures (T_C) vary as a function of the %SLs in the structure. Results show that: 1) Samples with low %SLs (< 70%) exhibit a single T_C value of 15-20K. 2) Samples with nearly 100% SLs (>90%) also exhibit a single Tc, with a higher value of 35-40K. 3) Samples with intermediate values of 70-85% SLs exhibit unusual T_C behavior, with evidence for two T_C components in the structure (T_{C1} and T_{C2}). For these, the T_{C1} value is ~40K, similar to the value for samples with >90% SLs, while the T_{C2} values are as high as 80K, higher than values reported to date for these materials. We interpret these results on the basis of the distribution of SL and QL layers within the structure, and the excess Mn contained in the layers. We propose that the structures with two T_C values consist of two distinct regions: regions of mostly SLs, that behave as materials with >90%SLs, and regions of SLs surrounding isolated QLs which lead to the higher T_{C2} values.

10:45am **NM-WeM2-12 High Curie Temperature $(Mnsb_2Te_4)_x(Sb_2Te_3)_{1-x}$ Magnetic Topological Insulator Structures Growth by Molecular Beam Epitaxy**, ***Candice Forrester,*** The Graduate Center (CUNY), The City College of New York, Lehman College; *I. Levy,* The Graduate Center (CUNY), The City College of New York; *G. Lopez-Morales,* The Graduate Center (CUNY), Lehman College; *X. Ding, K. Wickramasinghe,* The City College of New York; *C. Testelin,* Sorbonne Université, CNRS, Institut des NanoSciences de Paris; *D. Smith,* Arizona State University; *G. Lopez,* The Graduate Center (CUNY), Lehman College; *M. Tamargo,* The Graduate Center (CUNY), The City College of New York

The interaction between magnetic impurities and topological electronic states of 3D topological insulators (TIs) has attracted many studies of predicted exotic phenomena, which may lead to the observance of quantum anomalous hall effect (QAHE) and realizations of quantum computing, among others.[1]

Mixing of Mn, Sb and Te during crystal growth by bulk growth or epitaxial techniques forms a new crystal phase, $MnSb_2Te_4$, with septuple layer (SL) units rather than typical quintuple layer (QL) units of the undoped TIs. These magnetic TIs display antiferromagnetic (AFM) behavior in bulk, which is not conducive for QAHE, and typically have high bulk conductivity, which limits the ability to detect the surface states and their possible applications. Our group has shown that controlling the Mn incorporation into crystal led to the design of QL:SL structures.[2] These structures exhibit ferromagnetic (FM) behavior and a reduced bulk conductivity depending on the %SLs in the structure. Our group has also recently observed that, for some QL:SL layer ratios, samples exhibit mixed magnetic behaviors, with two distinct Curie temperature (Tc) components (Tc1 and Tc2), and Tc2 values as high as 75-80K.

Here we report the further enhancement of the Tc2 values by modification of the MBE growth conditions. In particular, growth rates and the Mn beam equivalent pressure (BEP) ratios were varied in this study. Samples with a lower growth rate of about 0.5-0.6 nm/min, compared to ~0.9-1.0 nm/min used previously, and with Mn BEP ratios between 0.07 and 0.09, resulted in structures having 79-89% SLs that also exhibit two distinct Tc values, one Tc value (Tc1) at ~40-50K and a high Tc value (Tc2) of 100K and above. (Fig. 1) These high Tc values are significantly higher than any values reported to date for these materials. Reducing the Mn BEP ratio to levels of 0.04-0.06 led to the observation of a single Tc value of ~30K and evidence for the coexistence of FM and AFM phases, suggesting the approach to stoichiometric $MnSb_2Te_4$ growth, with little excess Mn. (Fig. 2) We will present these results as they relate to the $(MnSb_2Te_4)_x(Sb_2Te_3)_{1-x}$ structural properties and the details of the growth mechanism. We will perform first-principle calculations using the Ising model to describe and understand the possible magnetic interactions that lead to samples with such high Curie temperatures.

1. R. Yu, et al., *Science* 2010, 329, 61−64.
2. I. Levy et al., *Crystal Growth & Design* 2022 https://doi.org/10.1021/acs.cgd.1c01453 (Published Online)

11:00am **NM-WeM2-13 Structural and Magnetotransport Properties of $MnBi_2Te_4$-based Heterostructure Grown by Molecular Beam Epitaxy**, ***Seul-Ki Bac***, *K. Koller, J. Wang, L. Riney, M. Zhukovskyi, T. Orlova, X. Liu, B. Assaf,* University of Notre Dame

The intrinsic magnetic topological insulators get enormous attention due to the interplay of the topological nontrivial electronic states and the magnetic order, which produces quantum phenomena, such as the quantum anomalous Hall effect and the axion insulator state. Although most research has been accomplished in exfoliated $MnBi_2Te_4$ thin flakes, it remains a big challenge to prepare high-quality thin films with well-controlled compositions and thicknesses. Here, we grow three different types of $MnBi_2Te_4$-based films by molecular beam epitaxy (MBE) and analyze structural and magnetotransport properties. Three samples are spontaneously obtained during the growth depending on growth conditions: a single $MnBi_2Te_4$ (1-phase), a composite structure with $MnBi_2Te_4$ and Bi_2Te_3 (2-phase), and a composite structure with $MnBi_2Te_4$, Bi_2Te_3, and MnTe (3-phase). We distinguish each phase using structural analysis, Raman spectroscopy and x-ray diffraction, and magnetotransport measurement. Our studies disclose insights on optimizing the MBE growth conditions and differentiating the types of $MnBi_2Te_4$-based films.

11:15am **NM-WeM2-14 MBE Growth and Thermo-/Magneto-Transport Properties of Ternary $(Bi,Sb)_2(Te,Se)_3$ Films with High Mobility**, ***Patrick Taylor***, US Army Research Laboratory; *H. Chi,* Massachusetts Institute of Technology; *B. Wooten, J. Heremans,* Ohio State University; *H. Hier, O. Vail,* US Army Research Laboratory; *J. Moodera,* Massachusetts Institute of Technology

We report the results from the exploration of the MBE growth and characterization of ternary *p*-type $(Bi,Sb)_2Te_3$ and *n*-type $Bi_2(Te,Se)_3$. Comprehensive temperature, field and angular dependent magnetotransport measurements of these ternary $(Bi,Sb)_2(Te,Se)_3$films show band-insulator behavior and display low carrier density on the order of 10^{18} cm^{-3} and a record high mobility exceeding 10^4 cm^2 V^{-1} s^{-1} at 2 K. The remarkable manifestation of strong Shubnikov–de Haas (SdH) quantum oscillation under 9 T at liquid helium temperatures, as well as the analyses therein, has allowed direct experimental investigation of the epitaxial layer electronic structure. Thermal phenomena including the Seebeck and Nernst coefficients are found to be consistent with the understanding obtained from magneto-transport measurements.The primary significance of these results is a path forward for topological spintronics with low bulk carriers and unprecedented magnetoelectric functionalities.

11:30am **NM-WeM2-15 MBE Growth of High Mobility Topological Crystalline Insulators in Proximity with a Magnetic Insulator***, J. Wang,* University of Notre Dame*; M. Ozerov,* National High Magnetic Fields Lab*; T. Wang, M. Zhukovskyi, T. Orlova,* University of Notre Dame*; D. Smirnov,* National High Magnetic Fields Lab*; V. Lauter,* Oak Ridge National Laboratory*; X. Liu,* ***Badih Assaf****,* University of Notre Dame

Topological insulators (TIs) are promising materials for spintronic and quantum devices due to the fact that they host Dirac fermions with spin-momentum locking. Spintronic devices based on TIs generally consist in a TI proximitized with a magnetic insulator. But the Dirac fermions in the TI can be gapped by magnetism. The magnetic proximity induced gap has never been measured but is important to quantify to properly evaluate the impact of the magnetic layer on the TI. Here, report an MBE synthesis scheme of a topological crystalline insulator (TCI) in proximity with a magnetic insulator that yields very high mobilities needed to evaluate this gap. The mobility is high enough (~1m^2/Vs at 4.5K) to allow us to extract the gap using magnetoinfrared spectroscopy. Our measurements allow us to conclude that the magnetic proximity gap cannot be larger than 20meV. Considering the size of the Fermi surface in TIs and TCIs, such a small gap likely preserves the helicity of topological states, making TIs ideal for spintronic devices, despite their fragility to magnetic exchange interactions.

11:45am **NM-WeM2-16 Closing Remarks and Thank Yous***,*

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