Electronic Materials and Photonics Division
Room 14 - Session EM+MI+TF-MoM

Growth, Electronic, and Magnetic Properties of Heusler Compounds

Moderators: Rehan Kapadia, University of Southern California, Seth King, University of Wisconsin - La Crosse

8:20am **EM+MI+TF-MoM-1 Semiconducting Half-Heusler Heterostructures Grown by Molecular Beam Epitaxy**, ***Anthony Rice***, *S Harrington, D Pennachio, M Pendharkar, C Palmstrøm,* University of California at Santa Barbara

Half-Heusler (hH) compounds are an attractive family of materials for a number of applications due to their wide range of properties, including half-metallic ferromagnetism and topologically non-trivial surface states. Additionally, those containing 18 valence electrons per formula unit are predicted to show a semiconducting band gap [1]. This suggests the possibility of a single multifunctional material composed of compounds with the same crystal structure throughout which makes use of the diverse hH properties not accessible by traditional III-V technology as well as more traditional band gap engineering.

In this presentation, the heterointerface formed between the 18 valence electron semiconducting hHs, CoTiSb and NiTiSn, is investigated. Layered structures with both NiTiSn and CoTiSb, have been successfully grown on MgO(001) substrates using molecular beam epitaxy. Transmission electron microscopy and X-ray diffraction (XRD) data suggest separate layers with sharp interfaces. X-ray photoelectron spectroscopy (XPS) data shows no evidence of intermixing, with component peaks attenuating as expected. XPS is used to measure the valence band offset, which suggests a type-I heterojunction.

Through the use of CoTiSb buffer layers, the integration of NiTSn with III-V substrates is demonstrated. Previous attempts at direct growth of NiTiSn on III-Vs has proven unsuccessful due to the high reactivity of nickel with III–Vs. Reflection high-energy electron diffraction intensity oscillations during growth are observed for these structures, consistent with layer-by-layer growth. XRD interference fringes suggest abrupt interfaces. Higher quality NiTiSn is ultimately achieved, with lower carrier concentrations and higher mobility. Interface transport, both laterally and vertically, is also explored.

This work was supported in part by the Vannevar Bush Faculty Fellowship (ONR-N0014-15-1-2845) and NSF-MRSEC (DMR-1121053). The UCSB MRL Shared Experimental Facilities are supported by the MRSEC Program of the NSF under Award No. DMR 1121053; a member of the NSF-funded Materials Research Facilities Network. A part of this work was performed in the UCSB Nanofabrication Facility which is a part of the NSF funded National Nanotechnology Infrastructure Network.

[1] T. Graf, C. Felsar, and S. Parkin. Progress in Solid State Chemistry 39 (2011) 1-50

8:40am **EM+MI+TF-MoM-2 Towards Topotronics: Combining Chemical Potential Tuning and Strain Engineering to Realize Surface Dominated Transport in Topological Heusler Thin Films**, ***Shouvik Chatterjee***, *J Logan, N Wilson, M Pendharkar, C Palmstrøm,* University of California at Santa Barbara

Heusler compounds have emerged as an exciting material system where realization of functional and tunable novel topological phases might be possible[1-4]. PtLuSb is one such compound that has been shown to host topologically non-trivial surface states[5]. However, being a semi-metal without a bulk band gap, exotic transport and thermodynamic properties expected from topological surface states are obscured by contributions from trivial bulk carriers that limits possible device applications[6]. Furthermore, natural defects in the compound leads to unintentional p-type doping resulting in the surface Dirac point lying above the chemical potential[5,6,7].

In this talk, I will present our efforts to address both these issues by a combination of carrier doping and substrate induced bi-axial strain to shift the chemical potential and attempt to open up a bulk gap, respectively. I will show experimental evidence of chemical potential tuning in Au alloyed $Pt_{1-x}Au_xLuSb$ thin films where the surface Dirac point can be pushed below the Fermi level. In addition, it is possible to open a bulk-band gap by application of compressive bi-axial strain on thin films synthesized on lattice mismatched substrates. Realization of surface dominated transport in topological Heusler thin films will open up avenues for realization of many exotic phenomena such as quantum anomalous Hall effect[8], axion insulators[9], topological superconductivity[10] and their potential device applications.

References:

1. S. Chadov *et al*, Nature Mater, 9, 541 (2010)
2. H. Lin *et al*, Nature Mater, 9, 546 (2010)
3. J. Ruan *et al,* Nature Commun, 10, 11136 (2016)
4. C. J. Palmstrøm, Prog. Cryst. Growth Charact. Mater,62, 371-397 (2016)
5. J. Logan *et al,* Nature Commun, 7, 11993 (2016)
6. S. J. Patel *et al*, Appl. Phys. Lett., 104, 201603 (2014)
7. Y. G. Yu, X. Zhang and A. Zunger, Phys. Rev. B, 95, 085201 (2017)
8. C-Z. Chang *et al,* Science, 340, 167 (2013)
9. L. Wu *et al,* Science, 354, 1124 (2016)
10. L. Fu and C. Kane, Phys. Rev. Lett,100, 096407 (2008)

9:00am **EM+MI+TF-MoM-3 Topology, Magnetism, and Superconductivity in Ternary Half-Heusler Semimetals**, ***Johnpierre Paglione***, University of Maryland, College Park **INVITED**

In materials with non-centrosymmetric crystallographic structures, it has long been known that the lack of inversion symmetry can give rise to odd-parity spin-triplet pairing states. We report superconductivity and magnetism in a new family of topological semimetals, the ternary half Heusler compounds RPtBi and RPdBi (R : rare earth). In this series, tuning of the rare earth f-electron component allows for simultaneous control of both lattice density via lanthanide contraction, as well as the strength of magnetic interaction via de Gennes scaling, allowing for a unique tuning of both the normal state band inversion strength, superconducting pairing and magnetically ordered ground states. Antiferromagnetism with ordering vector (0.5,0.5,0.5) occurs below a Néel temperature that scales with de Gennes factor, while a superconducting transition is simultaneously linearly suppressed. With superconductivity appearing in a system with non-centrosymmetric crystallographic symmetry as well as p-orbital derived spin-3/2 quasiparticles, the possibility of high-spin Cooper pairing (i.e. beyond triplet) with non-trivial topology analogous to that predicted for the normal state electronic structure provides a unique and rich opportunity to realize both predicted and new exotic excitations in topological materials.

9:40am **EM+MI+TF-MoM-5 Electron Counting, Surface Reconstructions, and Electronic Structure of 18 Electron Half Heuslers**, ***Jason Kawasaki***, University of Wisconsin - Madison*; A Janotti,* University of Delaware*; C Palmstrøm,* University of California at Santa Barbara

Half Heusler compounds (composition *ABC*) show great promise for the development of earth abundant thermoelectrics, half metallic ferromagnets for spin injection, and topological heterostructures. In these applications, the electronic structure of surfaces and interfaces are critical to materials performance. However, little is known about how and why the surfaces of these materials reconstruct or their direct effect on electronic properties. Using a combination of molecular beam epitaxy, angle resolved and core level photoemission, scanning tunneling microscopy, and density functional theory (DFT), we investigate the stability, reconstructions, and electronic surface states on the (001) surfaces of CoTiSb, NiTiSn, and FeVSb. These compounds are representative of a large class of 18 valence electron Half Heuslers that are expected to be semiconducting. We find that reconstructions in these compounds are characterized by *C* site (group IV or V) dimerization, as in III-V semiconductors, and this dimerization coincides with *B* site vacancies at the surface. We explain these trends using a simple electron counting model, and predictions from the model are in good agreement with both the experimental data and with DFT calculations. Our combined theoretical and experimental studies provide a rationale for understanding and controlling reconstructions and resultant electronic surface states in Heuslers.

10:00am **EM+MI+TF-MoM-6 Computational Investigation of Heusler Compoundss for Spintronic Applications**, ***Jianhua Ma***, University of Virginia*; W Butler,* University of Alabama

We present first-principles density functional calculations of the electronic structure, magnetism, and structural stability of 378 *XYZ* half-Heusler, 405 X_2YZ inverse-Heusler, 576 X_2YZ full-Heusler compounds. We find that a "Slater-Pauling gap" in the density of states in at least one spin channel is a common feature in Heusler compounds. We find that the presence of such

a gap at the Fermi energy in one or both spin channels contributes significantly to the stability of a Heusler compound. We calculate the formation energy of each compound and systematically investigate its stability against all other phases in the open quantum materials database (OQMD). We represent the thermodynamic phase stability of each compound as its distance from the convex hull of stable phases in the respective chemical space and show that the hull distance of a compound is a good measure of the likelihood of its experimental synthesis. We find low formation energies and mostly correspondingly low hull distances for compounds with X = Co, Rh, or Ni, Y = Ti or V, and Z = P, As, Sb, or Si in half-Heulser compounds. In the half-Heusler family, we identify 26 18-electron semiconductors, 45 half-metals, and 34 near half-metals with negative formation energy that follow the Slater-Pauling rule of three electrons per atom. In the inverse-Heusler family, we identify 14 asymmetric 18-electron semiconductors, 50 half-metals, and 42 near half-metals with negative formation energy. In the full-Heusler family, we identify 8 24-electron semiconductors and 23 half-metals with negative formation energy. Our calculations predict several new, as-yet unknown, thermodynamically stable phases, which merit further experimental exploration—RuVAs, CoVGe, FeVAs in the half-Heusler structure, and NiScAs, RuVP, RhTiP in the orthorhombic MgSrSi-type structure. Further, two interesting zero-moment half-metals, CrMnAs and MnCrAs, are calculated to have negative formation energy. In addition, our calculations predict a number of hitherto unreported semiconducting (e.g., CoVSn and RhVGe), half-metallic (e.g., RhVSb), and near half-metallic (e.g., CoFeSb and CoVP) half-Heusler compounds to lie close to the respective convex hull of stable phases, and thus may be experimentally realized under suitable synthesis conditions, resulting in potential candidates for various semiconducting and spintronics applications.

11:00am **EM+MI+TF-MoM-9 Growth, Electronic, and Magnetic Properties of Half-Heusler $CoTi_{1-x}Fe_xSb$**, ***Sean Harrington***, *A Rice, T Brown-Heft, A McFadden, M Pendharkar,* University of California at Santa Barbara*; O Mercan, L Çolakerol Arslan,* Gebze Technical University, Turkey*; C Palmstrøm,* University of California at Santa Barbara

Recent predictions suggest the semiconducting half-Heusler compound, CoTiSb, exhibits half-metallicity when substitutionally alloyed with Fe. However, to date, few studies have examined the growth of high-quality single crystal thin films of Fe-alloyed CoTiSb. Here, we report the epitaxial growth of the substitutionally alloyed half-Heusler series $CoTi_{1-x}Fe_xSb$ by molecular beam epitaxy and the influence of Fe on the structural, electronic, and magnetic properties. $CoTi_{1-x}Fe_xSb$ epitaxial films are grown on InAlAs grown on InP (001) substrates for concentrations $0 \le x \le 1$. The films are epitaxial and single crystalline, as measured by reflection high-energy electron diffraction and X–ray diffraction. For films with higher Fe content, a lower growth temperature is necessary to minimize interfacial reactions. Using *in-situ* X-ray photoemission spectroscopy, only small changes in the valence band spectra from pure CoTiSb are detected. For films with $x \ge 0.05$, ferromagnetism is observed in SQUID magnetometry with a Curie temperature >400K. The saturation magnetization of the series increases linearly with Fe content as 3.4 μ_B/Fe atom. In comparison, there is a much smaller magnetic moment when the Fe is substituted on the Co site ($Co_{1-x}Fe_x$-Ti-Sb) indicating a strong dependence of the magnetic moment with site occupancy. A cross over from both in-plane and out-of-plane magnetic moments to only in-plane occurs for higher concentrations of Fe. Ferromagnetic resonance indicates a transition from weak to strong interaction as Fe content is increased. Temperature dependent transport shows a gradual semiconductor to metal transition with thermally activated behavior for $x \le 0.3$. Anomalous Hall effect and magneto resistance are investigated for the x=0.3 and x=0.5 films revealing large differences in the electronic scattering mechanisms and transport behavior depending on Fe content.

11:20am **EM+MI+TF-MoM-10 High Spin-Polarization and Perpendicular Magnetic Anisotropy in Single-Crystal Full-Heusler Co_2MnAl/Fe_2MnAl Superlattice**, ***Tobias Brown-Heft***, *A McFadden, J Logan,* University of California at Santa Barbara*; C Guillemard,* University of Lorraine, France*; P Le Fevre, F Bertran,* Synchrotron SOLEIL, France*; S Andrieu,* University of Lorraine, France*; C Palmstrøm,* University of California at Santa Barbara

Ferromagnetic contacts are used as a source of spin-polarized current in many spintronic devices. Desired properties for ferromagnetic contacts used in magnetic tunnel junctions and other next-generation memory elements are perpendicular magnetic anisotropy and 100% spin polarization at the Fermi level (half-metallicity). Heusler compounds are strong candidates for this purpose as many have been predicted and observed to be half-metals (e.g. Co_2MnSi), while others exhibit perpendicular magnetic anisotropy (e.g. $Co_2FeAl/MgO(001)$). However, until now both properties have not been observed by experiment in a single material. J. Azadani *et al* have predicted that perpendicular anisotropy can be combined with half-metallicity by growing atomic-period superlattices of two different Heusler compounds [1]. We have successfully grown a single-crystal superlattice formed by layers of Co_2MnAl and Fe_2MnAl with periodicity of one to three unit cells using molecular beam epitaxy. X-ray diffraction reciprocal space mapping reveals that the superlattice is compliant to the substrate to at least 20 nm film thickness, sustaining strains from -3.0% (tensile) on MgO(001) to +2.3% (compressive) on GaAs(001). The film strain is accommodated via tetragonal distortion of c/a = 0 .96 to 1.06, respectively. The tetragonal distortion on GaAs(001) contributes to perpendicular magnetic anisotropy, resulting in films exhibiting out-of-plane magnetic easy axes at temperatures below 200K. Films with aluminum content higher than nominal stoichiometry may also help to induce perpendicular magnetization by reducing saturation magnetization, thereby lowering thin-film shape anisotropy. Superlattice structure was verified using electron energy loss spectroscopy in TEM, which shows low interface diffusion of cobalt and iron and high elemental contrast between individual superlattice layers. S pin polarization of >90% near the Fermi level has been measured directly via spin-resolved photoemission spectroscopy. Spin-resolved photoemission spectra suggest that the termination layer near a tunnel barrier interface should be Co_2MnAl-like, and may benefit from further composition tuning. This work was supported in part by C-SPIN, one of the six centers of STARnet, a Semiconductor Research Corporation program, sponsored by MARCO and DARPA. We also acknowledge support from the Vannevar Bush Faculty Fellowship.

[1] J. G. Azadani *et al*. J. Appl. Phys. 119, 043904 (2016).

11:40am **EM+MI+TF-MoM-11 Formation of the Epitaxial MgO/Full-Heusler Co_2MnSi Interface: Oxygen Migration and Elemental Segregation**, ***Anthony McFadden***, *T Brown-Heft, N Wilson, J Logan, C Palmstrøm,* University of California at Santa Barbara

Magnetic tunnel junctions (MTJs) are an increasingly important emerging technology for both magnetic random access memory (MRAM) and spintronics applications. MTJs utilizing CoFeB magnetic electrodes and MgO tunneling barriers have received considerable interest for use in MRAM as desirable properties including perpendicular magnetic anisotropy, high tunneling magnetoresistance ratio, and current induced switching have been demonstrated. While CoFeB/MgO based MTJs have demonstrated remarkable performance, devices utilizing ferromagnetic Heusler compounds have the potential to surpass CoFeB based technologies due to a much higher predicted spin polarization. In addition, many Heusler candidates have even been predicted to be half-metallic (100% spin polarized at the Fermi-level). Of all predicted half-metals, the full-Heusler Co_2MnSi has received considerable attention as it is quite stable (ΔH_F = -0.441 eV/atom), has a high Curie temperature (T_c=985K), and a large minority-spin energy gap (571 meV). While Heusler based MTJs have the potential to surpass current CoFeB based technology, the spin polarization of Heusler compounds has been shown to be sensitive to atomic ordering, adding an additional challenge to materials growth and integration.

In the present work, the formation of the $MgO/Co_2MnSi(001)$ interface has been studied *in-situ* using X-ray photoelectron spectroscopy (XPS). Co_2MnSi layers were grown on Cr-buffered MgO(001) substrates by coevaporation of elemental sources in ultrahigh vacuum while MgO was grown on the Co_2MnSi layers using e-beam evaporation of stoichiometric source material. It was found that partial oxidation of the Co_2MnSi surface was unavoidable during e-beam evaporation of MgO with oxygen bonding preferentially to Mn and Si. Interestingly, oxidation draws Mn and Si to the surface, resulting in an MgO/Co_2MnSi interface with composition significantly different from the unoxidized Co_2-MnSi surface. In addition, Mn and Si oxides at the MgO/Co_2MnSi interface were reduced following annealing in UHV with a corresponding migration of oxygen from the interface into the MgO. The results of XPS studies have been correlated with temperature dependent transport measurements of fully epitaxial $CoFe/MgO/Co_2MnSi$ MTJs which were observed to be highly sensitive to post-growth annealing temperature.

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