

PCSI

Room Ballroom South - Session PCSI-SuA

Topological Materials I & Perovskites

Moderators: Scott Crooker, Los Alamos National Laboratory, Peter Krogstrup, Niels Bohr Institute

3:00pm PCSI-SuA-1 Topological Superconductivity and Majorana Zero Modes on β -Bi₂Pd Thin Films, *Can-Li Song*, Tsinghua University, China **INVITED**

The search for Majorana fermions in solid state system is one of paramount research targets in physics today. Majorana fermions are an exotic class of fermions (which are their own antiparticles) and predicted to exist in topological superconductors. By growing β -Bi₂Pd epitaxial films on SrTiO₃ substrates with state-of-the-art molecular beam epitaxy, we here demonstrate a topologically nontrivial superconductivity on β -Bi₂Pd thin films. Majorana zero modes, supported by such a superconducting state, are identified by directly probing quasiparticle density of states within the vortex cores under magnetic field with a cryogenic scanning tunneling microscope. The superconductivity and Majorana zero modes (MZMs) exhibit resistance to intrinsic point and linear defects, characteristic of a time-reversal-invariant topological superconductor. Our discovery of the topological superconductivity and MZMs in single material of β -Bi₂Pd thin films represents one of the major advances in topological quantum physics.

3:30pm PCSI-SuA-7 Origin of the Helicity Dependent Photocurrent in Electrically Gated (Bi_{1-x}Sb_x)₂Te₃ Thin Films, *Yu Pan*, *N Samarth*, *C Liu*, *Q Wang*, Penn State University; *A Yeats*, *D Awschalom*, University of Chicago

Experimental studies of three-dimensional topological insulators have shown that circularly polarized photons can robustly generate a directional helicity-dependent photocurrent at room temperature [1]. Surprisingly, the phenomenon is readily observed at photon energies that excite electrons to states far from those within the spin-momentum locked Dirac cone. The underlying mechanism for the helicity-dependent photocurrent is thus far from obvious and still a mystery. We resolve the puzzle by carrying out a comprehensive study of the helicity dependent photocurrent in (Bi_{1-x}Sb_x)₂Te₃ thin films (Figure 1) as a function of the incidence angle of the optical excitation, its wavelength and the gate-tuned chemical potential (Figure 2). Our observations allow us to unambiguously identify the circular polarized photo-galvanic effect as the dominant mechanism for the helicity-dependent photocurrent. Additionally, we relate the directional nature of the photocurrent to asymmetric optical transitions between the topological surface states and bulk bands. This is rigorously inferred from a first principles calculation that reproduces the distinctive experimental dependence of the helicity-dependent photocurrent on gate voltage and excitation wavelength. The insights provided by our study are important for engineering opto-spintronic devices whose functionality relies on optical steering of spin and charge currents.

[1] McIver, J. W., Hsieh, D., Steinberg, H., Jarillo-Herrero, P. & Gedik, N., *Nature Nanotech.* **7**, 96 (2012).

3:45pm PCSI-SuA-10 High-efficiency Hybrid Perovskite Based Optoelectronic Devices with Technologically Relevant Stability, *Aditya Mohite*, Los Alamos National Laboratory **INVITED**

Hybrid (inorganic-organic) perovskites have demonstrated an extraordinary potential for clean sustainable energy technologies and low-cost optoelectronic devices such as solar cells; light emitting diodes, detectors, sensors, ionic conductors etc. In spite of the unprecedented progress in the past six years, one of the key challenges that exist in the field today is the large degree of processing dependent variability in the structural and physical properties. This has limited the access to the intrinsic properties of hybrid perovskites and led to multiple interpretations of experimental data. In addition to this, the stability and reliability of devices has also been strongly affected and remains an open question, which might determine the fate of this remarkable material despite excellent properties. In this talk, I will describe our recently discovered approach for thin-film crystal growth as a general strategy for growing highly crystalline, bulk-like thin-films of both three-dimensional (3D) and layered two-dimensional (2D) hybrid perovskites that overcomes the above issues by allowing access to the intrinsic charge and energy transport processes within the perovskite thin-films and results in reproducible and stable high performance optoelectronic devices.

Briefly, photovoltaic devices fabricated using 3D hybrid perovskites show hysteresis-free response, with high degree of reproducibility, with an average efficiency of planar devices approaching ~18%. Photo-physical,

electrical characterization and theoretical modeling attribute the improved performance to reduced defects (bulk and interface) and improved charge-carrier mobility in large-grain devices. Finally, I will also describe some new results on Ruddlesden-Popper phase perovskites based devices. In our first few attempts, we fabricated solar cells with efficiency approaching 13% as compared to the previous best of 4.5%. This phenomenal increase in efficiency is attributed to the near single-crystalline quality thin-films with a strongly preferential out-of-plane alignment of the inorganic perovskite component that facilitates efficient charge transport. Photovoltaic devices exhibit no hysteresis or degradation in performance under continuous operation and withstand an illumination intensity up to 4-Suns. Importantly, these devices with layered perovskites exhibit extraordinary, technologically relevant stability with no loss in performance with for ~2000 hours under humidity and 1-SUN full spectrum illumination.

4:15pm PCSI-SuA-16 The Origin of High Photovoltaic Efficiencies in Large-grain Organic-Inorganic Perovskites, *Jean-Christophe Blancon*, *W Nie*, *A Neukirch*, *S Tretiak*, Los Alamos National Laboratory; *L Cognet*, Institut d'optique & CNRS; *A Mohite*, *J Crochet*, Los Alamos National Laboratory

Organometallic perovskites have attracted considerable attention after promising developments in energy harvesting and other optoelectronic applications. Notably, the recent synthesis of high-crystallinity thin films with grain sizes of hundreds of microns has offered a new opportunity for the development of efficient solar cells with high reproducibility and photostability [1][2]. However, it is crucial to obtain a deeper understanding of the intrinsic photo-physics and transport properties of perovskites with relevant structural characteristics. Here, we will present an investigation of the dynamics of photogenerated charge carriers in large-area grain organometallic perovskite thin films via confocal time-resolved photoluminescence spectroscopy. Our findings show that the bimolecular recombination of free charges is the dominant decay mechanism at excitation densities relevant for photovoltaic applications [3]. Bimolecular coefficients were found to be on the order of 10⁻⁹ cm³/s, comparable to typical direct-gap semiconductors, yet significantly smaller than what is theoretically expected. We also will provide evidence that there is no degradation in carrier transport in these thin films due to electronic defects through modeling of the photoluminescence kinetics and open circuit voltage characteristics of a photovoltaic cell. We conclude that suppressed electron-hole recombination and transport that is not limited by defects provide a microscopic model for the superior performance of large-area grain hybrid perovskites for photovoltaic applications.

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[1] W. Nie, et al., *Science* **347**, 522 (2015).

[2] W. Nie, et al., *Nat. Commun.* **7**, 11574 (2016).

[3] J.-C. Blancon, et al., *Adv. Func. Mater.* **26**, 4283 (2016).

4:20pm PCSI-SuA-17 Origin of Photo-absorption and Photo-emission in Two-dimensional Ruddlesden-Popper Perovskites, *Jean-Christophe Blancon*, *H Tsai*, *W Nie*, *A Stier*, Los Alamos National Laboratory; *L Pedesseau*, INSA de Rennes; *C Stoumpos*, *M Kanatzidis*, Northwestern University; *J Even*, INSA de Rennes; *S Crooker*, *J Crochet*, *A Mohite*, Los Alamos National Laboratory

Two-dimensional Ruddlesden-Popper layered perovskites (RPLPs) have recently emerged as an alternative to the classic bulk organic-inorganic hybrid perovskites, mainly due to significantly improved photo- and chemical-stability in optoelectronic devices [1][2]. They also offer a richer chemical playground as compared to their 3D counterparts, which promises a wider range of functionalities for this layered material. Few recent encouraging developments in optoelectronic applications, notably in energy harvesting and light emitting [2][3], have already been demonstrated in these two-dimensional layered perovskites. However, further development and optimization of devices will require a deeper understanding of the intrinsic photo-physics and transport properties of the phase-pure RPLP materials. Here we investigate the physical properties of photo-generated charge carriers in phase-pure (BA)₂(MA)_{n-1}Pb_nI_{3n+1} layered perovskite family [4], for which the *n*-number can be experimentally tuned thus the thickness of the perovskite layer. It is found that the photo-absorption and photo-emission properties of thin films with *n*>2 are dominated by bound excited states associated with edge-states of perovskite layers, which rule the characteristics of thin-film solar cells and light-emitting diodes. On the other hand, optical and electronic properties in RPLP crystals derive from both quantum and dielectric confinements of carriers in the two-dimensional perovskite layers, which are promising for future applications of color-tunable photon sources. Opportunities to

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control and switch between these states is also relevant for novel technology.

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- [1] I. C. Smith, et al., *Angew. Chem. Int. Ed.* **53**, 11232 (2014).
- [2] H. Tsai, et al., *Nature* **536**, 312 (2016).
- [3] M. Yuan, et al., *Nat. Nanotechnol.* **11**, 872 (2016).
- [4] C. C. Stoumpos, et al., *Chem. Mater.* **28**, 2852 (2016).

4:25pm **PCSI-SuA-18 Tin Oxide Atomic Layer Deposition on Hybrid Lead Halide Perovskites for Tandem Photovoltaics**, *Axel Palmstrøm, K Bush, J Raiford, M McGehee, S Bent*, Stanford University

Hybrid lead halide perovskites are promising candidates for low cost, thin film light absorbers; they have a tunable band gap and have demonstrated efficiencies as high as 22.1%. As such, these materials are of interest for wide-bandgap absorbers in tandem photovoltaics. Hybrid lead halide perovskites have rough surfaces and are sensitive to temperature and oxidative conditions, making many deposition processes incompatible with this material. Typical perovskite solar cells employ spin-deposited organic selective transport layers and evaporated metal contacts on top of the perovskite absorber. These organic selective transport layers have two main drawbacks for tandem solar cells: first, the rough perovskite surface requires thick organic layers for complete coverage, resulting in significant optical losses and second, these organic materials are incompatible with the types of sputter processes used to deposit high quality transparent contacts (such as indium-tin oxide). We apply tin oxide by atomic layer deposition (ALD) as a dual-purpose layer to achieve electron selectivity and sputter protection with high optical transmission.

In this work, we investigate by x-ray diffraction thin film perovskite stability under various ALD reaction conditions on three perovskite materials: $\text{Cs}_{0.25}\text{FA}_{0.75}\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_3$ (1.2 eV), $\text{Cs}_{0.17}\text{FA}_{0.83}\text{Pb}(\text{Br}_{0.17}\text{I}_{0.83})_3$ (1.6 eV) and $\text{Cs}_{0.17}\text{FA}_{0.83}\text{Pb}(\text{Br}_{0.5}\text{I}_{0.5})_3$ (1.8 eV). We then investigate the role of a LiF/C_{60} surface passivation layer on the stability and performance with tetrakis(dimethylamino)tin(IV) and water that degradation is driven by metal organic precursor exposure, rather than by moisture, and that LiF/C_{60} improves surface stability. Finally we demonstrate the efficacy of an optimized tin oxide layer for realizing both high efficiency and stability in perovskite-perovskite and perovskite-silicon tandem devices. We achieve an NREL certified 23.6% efficiency and over 1000 hr stability with a monolithic perovskite-silicon tandem architecture

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