PCSI

Room Ballroom South - Session PCSI-SuA

New Developments in Oxide Materials & Growth

Moderator: Alex Demkov, The University of Texas

4:00pm **PCSI-SuA-1 Crystal-Chemical Origins of the Ultrahigh Conductivity of Metallic Delafossites*, Chris Leighton,*** University of Minnesota **INVITED**

The delafossites are a class of complex oxides with general formula ABO_2 that have been available synthetically since 1971 [1]. Some of these delafossites are metallic, where conductive triangular sheets of A^{1+} ions are interspersed with insulating $B^{3+}O_6$ edge-sharing octahedral layers, generating a remarkably simple electronic structure at the Fermi level [1,2]. Only in the last 10-20 years, however, was it understood that despite their highly anisotropic complex-oxidic nature,metallic delafossites (particularly $PdCoO_2$ and $PtCoO_2$) are the *most conductive oxides known*, for reasons that remain poorly understood [1]. In particular, their room-temperature resistivity is lower than Au and their low-temperature resistivity falls as low as 8 nW cm, implying mean-free-paths of ~20 mm [1]. These extraordinary values have led to a slew of recent advances [1]. To reach such low-temperature values, it is widely accepted that these materials must be ultrapure and ultraperfect, although the methods for their bulk growth (which produce only small crystals) are not typically capable of such [1].

In this presentation, we first report a new approach to $PdCoO_2$ crystal growth, using a novel chemical vapor transport method to achieve order-of-magnitude gains in size and mass, the highest structural qualities yet reported, and record residual resistivity ratios ($440 < RRR < 760$) [3]. Nevertheless, the first detailed mass spectrometry measurements on these materials reveals that they are *not* ultrapure, typically harboring 100s-of-part-per-million impurity levels [3]. Through detailed crystal-chemical analyses, we resolve this apparent dichotomy, showing that the vast majority of impurities are forced to reside in the insulating Co-O octahedral layers, leaving the conductive Pd sheets highly pure (~1 ppm impurity concentrations). These purities are shown to be in quantitative agreement with measured residual resistivites [3]. We thus conclude that a previously unconsidered "sublattice purification" mechanism is essential to the ultrahigh low-temperature conductivity and mean-free-path of metallic delafossites [3], opening up many exciting device possibilities.

Work supported primarily by the US Department of Energy through the University of Minnesota Center for Quantum Materials under DE-SC0016371.

[1] Mackenzie, *Rep. Prog. Phys*. **80**, 032501 (2017).

[2] Zhang, Saha, Tutt, Chaturvedi, Voigt, Moore, Garcia-Barriocanal, Birol and Leighton, *Phys. Rev. Mater*. **6**, 115004 (2022).

[3] Zhang, Tutt, Evans, Sharma, Haugstad, Kaiser, Ramberger, Bayliff, Tao, Manno, Garcia-Barriocanal, Chaturvedi, Fernandes, Birol, Seyfried Jr., Leighton, arXiv:2308.14257

4:40pm **PCSI-SuA-9 Superconductivity and Magnetism in Infinite-Layer Nickelate Heterostructures*, Jennifer Fowlie,*** SLAC National Lab **INVITED**

Nickel and copper are nominally very similar in chemistry so the search for superconductivity in nickelates is a story as old as the quest to understand the high temperature superconductivity of the cuprates.

In this talk, I will introduce the recent discovery of superconductivity in infinite-layer nickelates [1] and the ever-growing family of nickelate superconductors. I will touch on some of the materials challenges involved before summarizing the key physics we have learned so far including results from x-ray scattering [2] that identify a Mott-Hubbard-like character to the infinite-layer nickelate electronic structure as well as a significant rare earth 5d influence at the Fermi level. In particular I will focus on muon spin rotation [3] that reveals local magnetism in these materials that 1) onsets at rather high temperature, 2) is independent of the rare earth 4f electrons, 3) appears to be robust to doping 4) is antiferromagnetic and possibly short-range-ordered in nature and 5) coexists with superconductivity at low temperatures.

Finally, I will come back to the comparison between nickelates and cuprates and discuss how the disparities in the magnetic properties may be understood.

[1] D. Li et al, Nature **572**, 624 (2019).

[2] H. Lu et al, Science **373**, 213 (2021).

[3] J. Fowlie et al, Nat. Phys. **18**, 1043 (2022).

5:20pm **PCSI-SuA-17 The Redox Chemistry of Oxide Molecular Beam Epitaxy*, Oliver Bierwagen,*** Paul-Drude-Institut für Festkörperelektronik Leibniz-Institut im Forschungsverbund Berlin, Germany

The materials class of crystalline oxides provides a plethora of functional (dielectric, semiconducting, superconducting, ferroelectric, or ferromagnetic) properties. For harnessing this potential, molecular beam epitaxy (MBE) has proven an established method that realizes high quality oxide thin films. In its mostly used variety, the corresponding cation is evaporated from an effusion cell in vacuum onto the heated substrate where it gets oxidized by molecular oxygen, ozone, or an oxygen plasma.

This contribution will review the peculiar redox chemistry of oxide MBE, related to the existence of suboxides with significantly higher vapor pressure than their cation elements, not only taking place at the growth surface but also in effusion cells. I will discuss the implications of this chemistry on growth rate, film composition, and flux emanating from the effusion cells with the example of the plasma-assisted MBE growth of the semiconducting oxides Ga_2O_3, In_2O_3, GeO_2, SnO_2 and its suboxide SnO. Three major reactions describe the suboxide-related chemistry:

(1.) The reaction of the metal with its oxide towards the suboxide, which can be utilizied to etch oxide films in-situ [1,6], but also provides efficient suboxide sources [2].

(2.) The reaction of the cation element with oxygen, resulting in unintentional suboxide fluxes from elemental sources, leading to unexpectedly high cation incorporation in the grown films [3]. On the substrate, the same reaction provides the *p*-type oxide SnO [4] as well as an efficient way of removing a cation layer in-situ. Generally, the suboxide formation on the growth front is the first step during oxide growth, its desorption limits the growth rate.

(3.) The reaction of the suboxide with activated oxygen in the source leads to source passivation [9]. On the growth front, it is the second step that completes oxide growth. A kinetic growth model that involves both these steps, describes the observed, peculiar growth rate dependence on metal-to-oxygen flux ratio and substrate temperature for SnO_2, Ga_2O_3, In_2O_3 [5], and GeO_2 [6]. A simpler, single- and zero-step growth kinetics can be realized when the suboxide instead of an elemental cation flux is provided to the growth front, as demonstrated for SnO_2 [2], Ga_2O_3 [7], and SnO [8] films grown by suboxide MBE (S-MBE).

5:25pm **PCSI-SuA-18 Optical Phonon Modes in $LaInO_3$: Lattice Dynamics and Complete Polarization Analysis of Raman-Active Modes*, Hans Tornatzky,*** Paul-Drude Institute for Solid State Electronics, Germany*; Z. Galazka,* Institut für Kristallzüchtung, Germany*; R. Gillen,* Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Germany*; O. Brand, M. Ramsteiner, M. Wagner,* Paul-Drude Institute for Solid State Electronics, Germany

$LaInO_3$ is part of the family of ABO_3 perovskites, and is considered promising for next generation devices, such as for power electronics, due to its band gap of about 4.5 eV. A detailed knowledge of phonon modes in $LaInO_3$ is important as they determine a number of material properties, such as the mechanical and elastic properties, thermal transport and carrier dynamics, phonon-assisted optical excitations, and many more. However, little is known about the vibrational properties of this material. In this study, we investigate the lattice dynamics by polarization- and angle-resolved Raman spectroscopy and density functional theory (DFT). We experimentally observe all but one of the Raman active modes and compare them to our simulated values from DFT. Furthermore, we present the DFT-derived phonon dispersion relation along the high symmetry directions in reciprocal space and depict the oscillation patterns for selected phonons at the Γ point. Finally, we determine the relative Raman tensor elements of the observed modes from the angular dependence of their corresponding scattering efficiencies (cf. Fig. 2).

5:30pm **PCSI-SuA-19 Non-Trivial Electronic States in the EuO/$KTaO_3$ Interface Revealed by Quantum Oscillations in High Magnetic Fields*, K. Rubi,*** Los Alamos National Laboratory*; M. Dumen, S. Chakraverty,* Institute of Nano Science and Technology, India*; S. Zeng, A. Ariando,* National University of Singapore*; M. Chan, N. Harrison,* Los Alamos National Laboratory

The coexistence of electric-field controlled superconductivity and spin-orbit interaction in two-dimensional electron gas (2DEG) based on complex oxides (e.g., $SrTiO_3$ and $KTaO_3$) hold great promise for advancement in spintronics and quantum computing. However, a comprehensive understanding of the electronic bands that give rise to the multifunctional character of these 2DEGs remains elusive. To address this, we recently

investigated quantum oscillations in the magnetoresistance of the $KTaO_3$-2DEG in high magnetic fields (60 T).

$KTaO_3$ is a 5*d* transition metal oxide, exhibiting a lighter effective mass of electrons and a stronger spin-orbit interaction at its conducting surface/interface than its counterpart $SrTiO_3$ [1-2]. A high-mobility spin-polarized 2DEG with the superconducting feature is discovered at the $EuO/KTaO_3$ interface [3]. In this talk, I will present novel insights into the electronic states of the $EuO/KTaO_3$ interface investigated through Shubnikov-de Haas (SdH) oscillations (Fig.1a). Remarkably, we observed a progressive increase in cyclotron mass and oscillation frequency with the magnetic field (Fig. 1b and c), indicating the presence of non-trivial electronic bands [4]. Besides providing experimental evidence for topological-like electronic states in $KTaO_{-3}$-2DEG, these findings shed light on the recent predictions of

[1] K. Rubi *et al*., npj Quantum Materials **5**, 1 (2020); King, P. D. C. *et al*. Phys. Rev. Lett.**108**, 117602 (2012).

[2] K. Rubi *et al*., Phy. Rev. Research **3**, 033234 (2021).

[3] H. Zhang *et al*., Phys. Rev. Lett. **121**, 116803 (2018); Liu *et al*., Science **371**, 716 (2021).

[4] K. Rubi *et al*., arXiv:2307.04854 (2023).

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