NAMBE

Room Cummings Ballroom - Session NAMBE2-TuA

Oxides II

Moderator: Zach Cresswell, Idaho National Laboratory

3:45pm **NAMBE2-TuA-8 Signatures of Bosonic Coupling in Superconducting $LiTi_2O_4$ Thin Films***, **Zubia Hasan**, G. Pan,* Harvard University*; M. Barone,* Cornell University*; C. Brooks,* Harvard University*; A. Kaczmarek,* Cornell University*; S. Sung,* Harvard University*; E. Mercer,* Northeastern University*; S. Sharma,* Arizona State University*; I. El Baggari,* Harvard University*; K. Nowack,* Cornell University*; A. Botana,* Arizona State University*; B. Faeth,* Cornell University*; A. De La Torre Duran,* Northeastern University*; J. Mundy,* Harvard University

The mechanisms behind unconventional superconductivity have been intensely studied over the past few decades. Leading this thrust has been the high T_c cuprates, whose pairing 'glue' has been widely debated. $LiTi_2O_4$, a spinel oxide material, is an unconventional superconductor that preceded the cuprates [1]. However, despite having one of the highest T_c (~13.7 K) for a non-cuprate oxide, little is known about its' superconducting mechanism, with reports of both unconventional pairing [2] and traditional phonon-mediated BCS-like behavior [3]. There have also been signs of orbital and spin fluctuations persisting up to ~100 K, based on angle-dependent transport data [4]. Nevertheless, it remains unclear which mechanisms—spin fluctuations, electron-phonon coupling or mixed valency—are essential for superconductivity in $LiTi_2O_4$. Despite interest in this compound, the direct visualization of its' band structure has been inhibited due to the difficulty of cleaving the highly three dimensional single crystals of $LiTi_2O_4$. Here, we utilize a novel approach to incorporate Li in a Molecular Beam Epitaxy (MBE) system and synthesize phase pure, highly crystalline $LiTi_2O_4$ thin films for the very first time via MBE. The atomically smooth surfaces that are enabled by MBE growth allow us to interrogate the electronic band structure via Angle Resolved Photoemission Spectroscopy (ARPES) experiments. Our work reveals the presence of strong bosonic coupling in the compound. The bands intriguingly show a kink resembling cuprate-like band renormalizations. Our data indicates strong correlations: the band centered at Γ shows a 'kink' at around E_B~40 meV and a quasi-particle peak and incoherent tail suggestive of coupling to a bosonic mode. We see that this mode is present at all values of k_F and k_Z and persists above T_c. We discuss the origin of the kinks in $LiTi_2O_4$, providing broader insight into the pairing symmetry present in this superconducting system.

[1] D. C. Johnston et al, Mater. Res. Bull. 8, 777–784 (1973).
[2] H. Xue et al, ACS Nano 16 (11), 19464 (2022).
[3] C. P. Sun et al, Phys. Rev. B 70, 054519 (2004).
[4] K. Jin. et al, Nat. Commun. 6, 7183 (2015)

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4:00pm **NAMBE2-TuA-9 Defect Engineering in Thin Films of the Pyrochlore Frustrated Magnet $Tb_2Ti_2O_7$***, **Margaret Anderson**, I. El Baggari, C. Brooks, T. Powell,* Harvard University*; C. Lygouras,* Johns Hopkins University*; A. N'diaye,* Lawrence Berkeley National Laboratory*; S. Koohpayeh,* Johns Hopkins University*; J. Nordlander,* Paul Drude Institute, Germany*; J. Mundy,* Harvard University

Among the pyrochlore oxides, the frustrated magnet $Tb_2Ti_2O_7$ has drawn intense interest as a spin liquid candidate. Its unusual magnetic properties rely on a careful balance of interactions on the frustrated pyrochlore lattice and are exquisitely sensitive to composition. Using reactive-oxide molecular beam epitaxy, we synthesize $Tb_2Ti_2O_7$ thin films on (111)-oriented YSZ substrates and probe their structural and magnetic properties. Single-phase pyrochlore thin films are realized within a large growth window at up to 25% off-composition. Using both scanning transmission electron microscopy and electron energy loss spectroscopy, we investigate defect formation with varying film stoichiometry. Titanium-rich films incorporate titanium excess via antistuffing of both Ti^{3+} and Ti^{4+} locally along slanted $\langle 11\bar{2}\rangle$ antiphase boundaries. In contrast, excess terbium is directly incorporated into the film through homogeneous stuffing and terbium-rich films exhibit mostly $\langle 111\rangle$ antiphase boundaries. DC magnetic susceptibility measurements suggest that defects reduce the frustration index of $Tb_2Ti_2O_7$, with titanium and terbium excess both leading to an enhanced saturated magnetic moment and less negative Curie−Weiss temperature compared to the stoichiometric film, while all films remain paramagnetic down to 1.8K.

4:15pm **NAMBE2-TuA-10 Soft Chemical Manipulation of MBE-Synthesized Ruddlesden-Popper Nickelates***, **Abigail Jiang**, A. Turkiewicz, G. Pan, D. Ferenc Segedin, C. Brooks, J. Mason, J. Mundy,* Harvard University

Recent observations of nickelate superconductivity have established the Ruddlesden-Popper (RP) $Ln_{n+1}Ni_nO_{(3n+1)}$ and square-planar $Ln_{n+1}Ni_nO_{(2n+2)}$ series as important comparisons to the high-T_c cuprates. Both theory and experiment suggest that lower dimensional phases may lead to higher critical temperatures. However, existing soft-chemical methods to achieve a cuprate-like d^9 configuration (i.e. deintercalation of apical oxygens to achieve the square-planar phase) do not apply to the most dimensionally confined n=1 RP due to stoichiometric limitations, prompting alternative chemical modifications to tune electronic structure of RP nickelates. Here, we synthesize low dimensional RP nickelate thin films via atomically precise, reactive-oxide MBE with chemical doping and tunable strain states on a variety of substrates. We also demonstrate new soft-chemical, topotactic methods to modify the anion sublattice with commensurate and promising changes in electronic structure.

4:30pm **NAMBE2-TuA-11 $BaTiO_3$ Films for Integrated Electro-Optics***, **Larissa Little**, B. Fazlioglu-Yalcin, A. Cavanagh, N. Sinclair, T. Zulu, K. Powell, C. Brooks, R. Westervelt, M. Loncar,* Harvard University*; D. Barton,* Northwestern University*; J. Mundy,* Harvard University

Barium titanate has been identified as a promising electro-optic material for wider-scale adoption of integrated electro-optic modulators and other integrated photonic devices. Currently, electro-optic modulators are often bulk crystals of lithium niobate with indiffused waveguides, limiting their size, speed, and efficiency. Although thin film lithium niobate offers improved scalability and higher bandwidth over its bulk counterpart, lithium oxides are not CMOS compatible and there is a growing need for higher bandwidth modulators with lower voltage requirements. Barium titanate is an alternative material platform which exhibits an extremely high electro-optic coefficient (r_{42} ~ 900 pm/V), a reasonably high band gap (> 3eV), a relatively high refractive index (n = 2.4), and is compatible with traditional CMOS processing. Scalable methods for creating high quality, single ferroelectric domain films of barium titanate are therefore extremely attractive for highly efficient modulators and integrated optical devices. Here we use molecular beam epitaxy to grow barium titanate films and characterize the defects and effects that stoichiometry and strain changes have on their properties.

4:45pm **NAMBE2-TuA-12 Exploration of Erbium-Doped Oxide Thin Films on Silicon for Quantum Memory-Oriented Nanophotonics Development***, **Ignas Masiulionis**,* University of Chicago/Argonne National Laboratory*; G. Grant,* University of Chicago*; R. Chebrolu,* University of Chicago / Argonne National Laboratory*; A. Dibos, J. Zhang, F. Heremans, S. Guha,* Argonne National Lab

Trivalent erbium ions (Er^{3+}) are promising defects for developing photon-enabled quantum communication technology due to their emission at telecom wavelengths (~1.5 µm). Certain classes of materials present appealing conditions for hosting erbium defects for quantum communication, such as oxides having the potential for long spin defect coherence times [1]. Titanium dioxide (TiO_2), in particular, has been investigated as an erbium host due to its ease of fabrication and well-known optical properties; certain complex oxides similarly have been chosen due to their low (<2%) in-plane lattice mismatch with silicon. Here, we discuss the growth and post-growth treatment of these materials for use in nanophotonics and eventual integration with quantum communication technologies, focusing on key metrics such as host surface roughness and Er^{3+} spectral diffusion linewidths. We identify particular optimizations that may be made, for example, that post-growth anneals at moderate temperatures can narrow Er^{3+} spectral diffusion linewidths in TiO_2 without significant roughening of the film. Additional spectroscopy (photoluminescence excitation (PLE) spectroscopy and transient spectral holeburning (TSHB)) plus traditional materials characterization (X-ray diffraction (XRD), atomic force microscopy (AFM)) allow us to ensure the crystalline host quality is suitable for ongoing quantum memory device development.

[1] S. Kanai, et al. PNAS. 119, e2121808119 (2022).

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Science Research Center, with additional support from the U.S. Department of Energy Office of Science Basic Energy Sciences, Materials Sciences and Engineering Division.

5:00pm **NAMBE2-TuA-13 Simultaneous Optical and Microstructural Characterization of Er-Doped CeO_2 on Silicon***, **Gregory Grant,*** University of Chicago*; J. Zhang,* Argonne National Laboratory*; I. Masiulionis,* University of Chicago*; S. Chattaraj, K. Sautter,* Argonne National Laboratory*; S. Sullivan,* memQ*; R. Chebrolu,* University of Chicago*; Y. Liu, J. Martins, J. Niklas, A. Dibos,* Argonne National Laboratory*; S. Kewalramani,* Northwestern University*; J. Freeland, J. Wen, O. Poluektov, F. Heremans,* Argonne National Laboratory*; D. Awschalom,* University of Chicago*; S. Guha,* Argonne National Laboratory

Erbium-doped cerium dioxide ($Er:CeO_2$) is a promising defect-host combination for applications in quantum memories and single photon emitters for wide-area fiber optic-based quantum networks. This is due to the combination of the telecom-compatible (~1.5 µm) 4f-4f transition of Er, the predicted long electron spin coherence time of defects in CeO_2 [1], and the small lattice mismatch between silicon and CeO_2. Here we report on the epitaxial growth of low-doped $Er:CeO_2$ thin films on silicon using molecular beam epitaxy (MBE), with controlled Er concentration in the 1-100 ppm regime. We verify the CeO_2 host structure via thorough microstructural study, and in tandem characterize the spin and optical properties of the embedded Er^{3+} ions as a function of doping density. This allows identification of trends that can enable quantum memory technologies, and simultaneously yields insights into what improvements must be made to the thin films for these applications. Notably for quantum memories, when studying the Er^{3+} Z_1-Y_1 optical transition near 1530 nm at liquid helium temperatures, we find spectral diffusion-limited homogeneous linewidths as narrow as 5 MHz [2], a promising result towards quantum memory technologies. Based on our study, we discuss routes towards improvement of the optical and spin linewidths via growth optimization and post-growth treatment of the $Er:CeO_2$ films.

[1] S. Kanai, et al., PNAS. 119, e2121808119 (2022).

[2] G. Grant, et al., APL Mater. 12, 021121 (2024).

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Author Index

Bold page numbers indicate presenter

— A —
Anderson, M.: NAMBE2-TuA-9, **1**
Awschalom, D.: NAMBE2-TuA-13, 2
— B —
Barone, M.: NAMBE2-TuA-8, 1
Barton, D.: NAMBE2-TuA-11, 1
Botana, A.: NAMBE2-TuA-8, 1
Brooks, C.: NAMBE2-TuA-10, 1; NAMBE2-TuA-11, 1; NAMBE2-TuA-8, 1; NAMBE2-TuA-9, 1
— C —
Cavanagh, A.: NAMBE2-TuA-11, 1
Chattaraj, S.: NAMBE2-TuA-13, 2
Chebrolu, R.: NAMBE2-TuA-12, 1; NAMBE2-TuA-13, 2
— D —
De La Torre Duran, A.: NAMBE2-TuA-8, 1
Dibos, A.: NAMBE2-TuA-12, 1; NAMBE2-TuA-13, 2
— E —
El Baggari, I.: NAMBE2-TuA-8, 1; NAMBE2-TuA-9, 1
— F —
Faeth, B.: NAMBE2-TuA-8, 1
Fazlioglu-Yalcin, B.: NAMBE2-TuA-11, 1
Ferenc Segedin, D.: NAMBE2-TuA-10, 1
Freeland, J.: NAMBE2-TuA-13, 2
— G —
Grant, G.: NAMBE2-TuA-12, 1; NAMBE2-TuA-13, **2**
Guha, S.: NAMBE2-TuA-12, 1; NAMBE2-TuA-13, 2
— H —
Hasan, Z.: NAMBE2-TuA-8, **1**
Heremans, F.: NAMBE2-TuA-12, 1; NAMBE2-TuA-13, 2
— J —
Jiang, A.: NAMBE2-TuA-10, **1**
— K —
Kaczmarek, A.: NAMBE2-TuA-8, 1
Kewalramani, S.: NAMBE2-TuA-13, 2
Koohpayeh, S.: NAMBE2-TuA-9, 1
— L —
Little, L.: NAMBE2-TuA-11, **1**
Liu, Y.: NAMBE2-TuA-13, 2
Loncar, M.: NAMBE2-TuA-11, 1
Lygouras, C.: NAMBE2-TuA-9, 1
— M —
Martins, J.: NAMBE2-TuA-13, 2
Masiulionis, I.: NAMBE2-TuA-12, **1**; NAMBE2-TuA-13, 2
Mason, J.: NAMBE2-TuA-10, 1
Mercer, E.: NAMBE2-TuA-8, 1
Mundy, J.: NAMBE2-TuA-10, 1; NAMBE2-TuA-11, 1; NAMBE2-TuA-8, 1; NAMBE2-TuA-9, 1
— N —
N'diaye, A.: NAMBE2-TuA-9, 1
Niklas, J.: NAMBE2-TuA-13, 2
Nordlander, J.: NAMBE2-TuA-9, 1
Nowack, K.: NAMBE2-TuA-8, 1
— P —
Pan, G.: NAMBE2-TuA-10, 1; NAMBE2-TuA-8, 1
Poluektov, O.: NAMBE2-TuA-13, 2
Powell, K.: NAMBE2-TuA-11, 1
Powell, T.: NAMBE2-TuA-9, 1
— S —
Sautter, K.: NAMBE2-TuA-13, 2
Sharma, S.: NAMBE2-TuA-8, 1
Sinclair, N.: NAMBE2-TuA-11, 1
Sullivan, S.: NAMBE2-TuA-13, 2
Sung, S.: NAMBE2-TuA-8, 1
— T —
Turkiewicz, A.: NAMBE2-TuA-10, 1
— W —
Wen, J.: NAMBE2-TuA-13, 2
Westervelt, R.: NAMBE2-TuA-11, 1
— Z —
Zhang, J.: NAMBE2-TuA-12, 1; NAMBE2-TuA-13, 2
Zulu, T.: NAMBE2-TuA-11, 1