PCSI
Room Keahou I - Session PCSI-WeM3

Photoemission Spectroscopy
Moderator: Gregory Fuchs, Cornell University

11:40am **PCSI-WeM3-39 Surface and Interface Effects on the Electronic and Magnetic Properties of $NiCo_2O_4$ Thin Films**, ***Arjun Subedi***, *B. Giri, D. Yang,* University of Nebraska–Lincoln*; A. N'Diaye,* Advanced Light Source, Lawrence Berkeley National Laboratory*; T. Komesu, X. Xu, P. Dowben,* University of Nebraska–Lincoln

The surfaces of epitaxial complex oxide thin films could differ chemically from its bulk. The distinctly different surface can lead to different surface electronic and magnetic properties compared to the bulk properties of the thin films. Using angle-resolved x-ray photoelectron spectroscopy (ARXPS) of the $NiCo_2O_4$ (NCO) thin films, NCO thin film surfaces are found to be Ni-rich and there exist surface-to-bulk core level shifts in binding energies of the Ni $2p_{3/2}$ and Co $2p_{3/2}$ core levels [1, 2]. While the Ni-rich conducting NCO thin film surface undergoes irreversible metallic to non-metallic (dielectric) phase transition, the highly reduced surface of the NCO thin film undergoes reversible phase transition from highly dielectric to much diminished dielectric (or highly enhanced metallic) character with temperature [2]. We have proposed a modified Arrhenius-type model for the core level binding energy change with temperature in x-ray photoelectron spectroscopy (XPS) of dielectric oxide thin films, and activation energies for thermal stimulations of the carriers are thus estimated [2]. Spin-polarized inverse photoemission spectroscopy (SPIPES) and angle-resolved x-ray magnetic circular dichroism (XMCD) of Ni-rich NCO thin film show that the surface spin moments of the film are canted, although the same film in the bulk is known to possess perpendicular magnetic anisotropy (PMA). When a thin platinum (Pt) layer was deposited on the NCO film, the spin moment canting at the interface of Pt/NCO increased dramatically, leading to significantly different spin ordering at the interface compared to the bulk spin ordering (PMA) of the NCO thin film. The reversibility in electronic phase transition can ultimately lead to two controlled, programmable and non-volatile electronic phase states, whereas spin-canting surfaces and interfaces make NCO thin films interesting for spintronics.

[1] **A. Subedi,** D. Yang, W. K. Chin, B. Tamang *et al.,* J. Phys.: Condens. Matter **36**, 285001 (2024).

[2] **A. Subedi**, D. Yang, X. Xu, and P. A. Dowben, J. Phys. D: Appl. Phys., in press, (2024). [https://doi.org/10.1088/1361-6463/ad5aa8]

11:45am **PCSI-WeM3-40 Spectroscopic Calculations for Trivalent Lanthanide Ions**, ***Tharnier O. Puel***, University of Iowa*; J. Lizarazo-Ferro, R. Zia,* Brown University*; M. E. Flatté,* University of Iowa

It is known that spin of optically addressable solid-state defects can be coherently manipulated using electric fields, enabling precise control over the photoluminescence spectra of spin defects when integrated into electrical junctions. Additionally, spin defects are also sensitive to electric and magnetic field noise, for example, electric field noise from surface charge fluctuations can be a significant source of spin decoherence. In this study, we examine trivalent lanthanide ion defects in crystals using a semi-empirical Hamiltonian, where parameters are empirically fitted to experimental data. By diagonalizing the Hamiltonian, we are able to predict key spectroscopic properties of these defects, including optical transition rates, oscillator strengths, magnetic dipole transitions, parity-forbidden electric dipole transitions, AC Stark shifts, and g-tensors for low-energy excitations. We have compared our results against the well-established calculations from Carnall et al. The Journal of Chemical Physics 90, 3443 (1989), for the lanthanide ions in LaF_3 and make available the *qlanth* software for easy reproducibility of the results. These findings are essential for advancing the control of rare-earth-based quantum devices and for understanding the mechanisms underlying their decoherence.

11:50am **PCSI-WeM3-41 Brillouin-Zone-Selection Effects in Angle-Resolved Photoemission Spectroscopy of Silicon**, ***Niels van Venrooij***, University of Iowa, Netherlands*; P. Constantinou, T. Stock,* University College London, UK*; V. Strocov,* Paul Scherrer Institut, Switzerland*; G. Aeppli,* ETH Zurich, Switzerland*; N. Curson, S. Schofield,* University College London, UK*; M. Flatté,* University of Iowa

The advancement of semiconductor-based atomic-scale quantum electronics hinges on a deep understanding of the electronic properties of subsurface δ-layers[1]. In this rapidly evolving field, soft X-ray angle-resolved photoemission spectroscopy (SX-ARPES) has emerged as a pivotal, non-destructive probing tool[2]. The large energy range of SX-ARPES allows for measurements across a broad momentum space, covering multiple Brillouin zones. During these measurements, interference effects from photoemission across different atoms in a unit cell can lead to pronounced intensity fluctuations between adjacent Brillouin zones, even causing some bands to vanish. In this study, we present the first detailed observations of such photoemission structure factor effects from a cubic semiconductor, revealing periodic fluctuations in both the valence and conduction band states of δ-doped silicon. By applying a simple tight binding calculation to Fermi's golden rule we are able to calculate the structure factor of Silicon and reproduce our experimental findings with a minimal number of approximations. Our findings pave the way for investigations at higher photon energies in the hard X-ray domain, crucial for exploring deeper δ-layers (~10 nm) typical in silicon quantum electronic devices.

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[1] J. G. Keizer, S. Koelling, P. M. Koenraad, and M. Y. Simmons, "Suppressing Segregation in Highly Phosphorus Doped Silicon Monolayers," ACS Nano, vol. 9, no. 12, pp. 12537–12541, 2015, doi: 10.1021/acsnano.5b06299.

[2] V. N. Strocov et al., "Soft-X-ray ARPES at the Swiss Light Source: From 3D Materials to Buried Interfaces and Impurities," Synchrotron Radiat News, vol. 27, no. 2, pp. 31–40, 2014, doi: 10.1080/08940886.2014.889550.

11:55am **PCSI-WeM3-42 A Topological Superconductor Tuned by Electronic Correlations**, ***Haoran Lin***, University of Chicago*; C. Jacobs,* West Virginia University*; C. Yan,* University of Chicago*; G. Nolan,* University of Illinois at Urbana-Champaign*; P. Singleton, Y. Bai, Q. Gao, G. Berruto, D. Nguyen,* University of Chicago*; X. Wu,* Chinese Academy of Sciences, China*; C. Liu,* Penn State University*; N. Guisinger,* Argonne National Laboratory*; P. Huang,* University of Illinois at Urbana-Champaign*; S. Mandal,* West Virginia University*; S. Yang,* University of Chicago

A topological superconductor, characterized by either a chiral order parameter [1] or a chiral topological surface state in proximity to bulk superconductivity [2], is foundational to topological quantum computing. Similar to other topological phases of matter, it can be profoundly tuned by electronic correlations through the modification of low-energy Fermiology, but not elucidated so far.

We present the study of a unique topological superconducting phase emerging in 10-unit-cell-thick $FeTe_xSe_{1-x}$ films grown on $SrTiO_3$ substrates [3]. By combining molecular beam epitaxy (MBE) growth with *in-situ* angle-resolved photoemission spectroscopy (ARPES) (Fig. 1), we investigate the electronic band structures of these thin films. When the Te content x exceeds 0.7, we observe a rapid increase of the effective mass for the Fe d_{xy} band, with the emergence of a topological surface state and superconductivity; however, near the FeTe limit, the system enters an incoherent regime where the topological surface state becomes unidentifiable, and superconductivity is suppressed. Theory [4] suggests that the electron-electron interactions in the odd-parity xy^- band with a strong d_{xy} character lead to an orbital-selective correlated phase. Our work establishes $FeTe_xSe_{1-x}$ thin films as a unique platform where electronic correlations sensitively modulate topological superconductivity, suggesting opportunities to use tunable electron-electron interactions to engineer new topological phases in a broad class of materials.

[1] A. Kitaev, AIP Conf. Proc. 1134, 22 (2009).

[2] L. Fu and C. L. Kane, Phys. Rev. Lett. 100, 096407 (2008).

[3] H. Lin et al. In Review (2024).

[4] M. Kim, S. Choi, W. H. Brito, and G. Kotliar, Phys. Rev. Lett. 132, 136504 (2024).

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