Unusual Magnetic Phenomena and Novel Temperature Scales at the Surfaces of 4f Materials: Insights from ARPES

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For a long time, Rare-Earth (RE) intermetallic materials have attracted considerable interest because of their rich and exotic properties. They include the complex magnetic phases, unconventional superconductivity, valence fluctuations, heavy-fermion and Kondo behavior and non-Fermi-liquid properties. A key point of the involved physics is the delicate interplay between itinerant electrons and the lattice of localized 4*f* states. Note here that the surfaces of such materials often do not receive as much attention as their bulk. However, it is reasonable to anticipate that the *f*- driven physics within the surface area can be even much richer and more compelling than that in the bulk. Lack of inversion symmetry and spin-orbit coupling (SOC), appearance of surface-electron states and resonances, relaxation and reconstruction, as well as strong changes of the crystal-electric field near and at the surface are the driving forces for novel *f*- driven phenomena, phases and temperature scales that are in remarkable difference to those in the bulk.

We will focus on a class of RET_2Si_2 compounds, where T is transition metal atoms, which possess the $ThCr_2Si_2$ type structure. Besides their unique bulk properties, as we will see these materials reveal a rather unusual phenomena at the surface and can be considered as models for studying the peculiarities of 4*f* physics within the non-centrosymmetric Si-T-Si-RE surface-silicide blocks. There, the strength of spin-orbit coupling (SOC) can be tuned by choice of suitable transition metal (T) atoms. It gradually increases by exchanging Co (3*d*) for Rh (4*d*) and further for Ir (5*d*). As a competing ingredient, exchange magnetic interaction may be exploited by inserting elementary 4*f* magnets like Gd as the RE component. Because the orbital moment of the Gd 4*f* shell vanishes (***L*** = 0), the pure and large spin moment of Gd will be a strong and robust source of magnetic phenomena. A rotation of the 4*f* moments to a certain angle relative to the surface normal may be achieved by coupling to a crystal electric field (CEF). To make use of notable CEF effects, a non-vanishing orbital moment ***L*** is needed, like for instance in Ho or Dy. Then, this option allows to implement an exchange magnetic field with different strength and orientation at the surface, which competes with the Rashba SOC field and creates additional possibilities to manipulate the properties of the 2D electrons within the considered Si-T-Si-RE system. As the next ingredient, the Kondo effect can be introduced by inserting elements with unstable 4*f* shell as Yb or Ce. This gives the opportunity to explore the interplay of the 2D electrons with 4*f* moments within a 2D Kondo lattice in the presence of spin-orbit coupling and a non-centrosymmetric environment.

Performing systematic photoemission experiments based on aforementioned chain of thoughts, we have realized the most of such scenarios and demonstrated quite nicely that the Si-T-Si-RE surfaces of the RET_2Si_2 materials serves as a versatile playground for studying the fundamental properties linked with *f-d* interactions at reduced dimensionality. It represents a kind of construction kit with Rashba spin-orbit coupling, Kondo interaction, crystal-electric fields and magnetic exchange with different strengths as building blocks. Their mutual combination gives the opportunity to design systems for different scenarios and to study the physics of 2D electron states in the presence of these competing interactions.

In this talk, the most interesting results, which unveil the novel *f*-driven properties and related temperature scales at the surfaces of the discussed RET_2Si_2 as well as of $CeTIn_5$ and $RECo_2P_2$ materials will be presented. It will be also shown that so essential property of 4*f* moments as their orientation in the individual RE layers can be reliably and readily derived from the line shapes of the classical momentum-resolved 4*f* photoemission spectra. This opens great opportunities to control the 4*f*-derived magnetic properties in RE-based heterostructures.

The obtained results make a call for detailed studies of the properties emerging at the surfaces and sub-surface areas of many strongly correlated Ce-, Eu-, and Yb-based materials with layered, quasi-2D structure. They also bear strong implications for how novel functional and quantum materials can be devised using thin layers of *f*-materials as building blocks. In such systems, different combinations of fundamental interactions can be realized. Combining them with one another gives the opportunity to predict and create novel materials with new functionalities.

References:

[1]. G. Poelchen et al., **Interlayer Coupling of a Two-Dimensional Kondo Lattice with a Ferromagnetic Surface in the Antiferromagnet $CeCo_2P_2$**, ACS Nano **16** 3573 (2022).

[2]. D. Usachov et al., **Estimating the Orientation of 4*f* Magnetic Moments by Classical Photoemission**, J. Phys. Chem. Lett. **13** 7861 (2022).

[3]. M. Mende, et al., **Strong Rashba Effect and Different *f-d* Hybridization Phenomena at the Surface of the Heavy-Fermion Superconductor $CeIrIn_5$**, Advanced Electronic Materials **8** 2100768 (2022).

[4]. S. Schulz, et al., **Classical and cubic Rashba effect in the presence of in-plane 4*f* magnetism at the iridium silicide surface of the antiferromagnet $GdIr_2Si_2$**, Physical Review B **103** 035123 (2021).

[5]. D. Yu. Usachov et al., **Cubic Rashba Effect in the Surface Spin Structure of Rare-Earth Ternary Materials**, Physical Review Letters **124** 237202 (2020).

[6]. G. Poelchen, et al., **Unexpected differences between surface and bulk spectroscopic and implied Kondo properties of heavy fermion $CeRh_2Si_2$**, npj Quantum Materials **5** 70 (2020).

[7]. S. Schulz et al., **Emerging 2D-ferromagnetism and strong spin-orbit coupling at the surface of valence-fluctuating $EuIr_2Si_2$**, npj Quantum Materials **4** 26 (2019).

[8]. A. Generalov et al., **Strong spin-orbit coupling in the noncentrosymmetric Kondo lattice**, Physical Review B **98** 115157 (2018) (Editors' Suggestion).

[9]. A. Generalov et al., **Spin orientation of two-dimensional electrons driven by temperature-tunable competition of spin-orbit and exchange magnetic interactions**, Nano Letters **17** 811 (2017).

[10]. S. Patil et al., **ARPES view on surface and bulk hybridization phenomena in the antiferromagnetic Kondo lattice $CeRh_2Si_2$**, Nature Communications **7** 11029 (2016).

[11]. A. Chikina et. al., **Strong ferromagnetism at the surface of an antiferromagnet caused by buried magnetic moments**, Nature Communications **5** 3171 (2014).