Unusual magnetic phenomena at the surfaces of lanthanide materials: Insights from ARPES and XAS experiments

Denis Vyalikh

Donostia International Physics Center, 20080 Donostia/San Sebastián, Basque Country, Spain

IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain

www.ikerbasque.net/en/denis-vyalikh

denis.vyalikh@dipc.org

For a long time, lanthanide (Ln) materials have attracted considerable interest due to their rich and exotic properties. These include complex magnetic phases, unconventional superconductivity, valence fluctuations, heavy-fermion and Kondo behavior, and non-Fermi-liquid properties. A key aspect of the involved physics is the interplay between itinerant electrons and the lattice of localized 4f moments. It is important to note that the surfaces of such materials often receive less attention than their bulk. However, it is reasonable to anticipate that the 4f-driven physics at the surface can be even richer and more compelling than in the bulk. The lack of inversion symmetry, spin-orbit coupling (SOC), the appearance of surface states and resonances, relaxation and reconstruction, as well as strong changes in the crystal-electric field near and at the surface, are driving forces for novel 4f-driven phenomena, phases, and temperature scales that differ significantly from those in the bulk.

We will focus on a class of LnT_2Si_2 materials, where T represents transition metal atoms. In addition to their unique bulk properties, these materials exhibit rather unusual phenomena at the surface and can be considered as models for studying the peculiarities of 4f physics within the non-centrosymmetric Si-T-Si-Ln surface-silicide blocks. In these systems, the strength of spin-orbit coupling (SOC) can be tuned by choosing appropriate transition metal (T) atoms. The SOC gradually increases by substituting Co (3d) with Rh (4d), and further with Ir (5d). As a competing effect, exchange magnetic interactions can be introduced by incorporating elementary 4f magnets like Gd as the Ln component. Since the orbital moment of the Gd 4f shell vanishes (L = 0), the pure and large spin moment of Gd provides a strong and robust source of magnetic phenomena. A rotation of the 4f moments relative to the surface normal can be achieved by coupling with the crystal electric field (CEF). To exploit notable CEF effects, a non-vanishing orbital moment is required, as in the case of Ho or Tb. This allows for the implementation of an exchange magnetic field with varying strength and orientation at the surface, competing with the Rashba SOC field and offering additional opportunities to manipulate the properties of the 2D electrons in the Si-T-Si-Ln system. Another element to consider is the Kondo effect, which can be introduced by inserting elements with an unstable 4f shell, such as Yb or Ce. This presents the opportunity to explore the interplay between 2D electrons and 4f moments within a 2D Kondo lattice in the presence of spin-orbit coupling and a non-centrosymmetric environment.

By performing systematic ARPES and XAS experiments based on the aforementioned chain of thought, we have realized most of these scenarios and demonstrated that the Si-T-Si-Ln surfaces of the LnT_2Si_2 materials serve as a versatile platform for studying the fundamental properties linked to *f-d* interactions at reduced dimensionality. These surfaces act as a "construction kit" comprising Rashba SOC, Kondo interactions, crystal-electric fields, and magnetic exchange with varying strengths. Their mutual combination allows for the design of systems for different scenarios and the study of 2D electron states in the presence of these competing interactions.

In this talk, I will present the most interesting results that reveal novel 4f-driven properties and related temperature scales at the surfaces of the discussed LnT_2Si_2, as well as $CeIrIn_5$ and $LnCo_2P_2$ materials. We will also demonstrate how an essential property of 4f moments, as their orientation in the individual Ln layers, can be reliably derived from the line shapes of classical momentum-resolved 4f photoemission spectra. This opens up significant opportunities to control the 4f-derived magnetic properties in Ln-based heterostructures.

The results carry strong implications for how novel functional and quantum materials can be developed by using thin layers of 4f-materials as building blocks. In these systems, different combinations of fundamental interactions can be realized, and their combination offers the potential to predict and create novel materials with new functionalities.

Own related references:

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