## Low dimensional systems

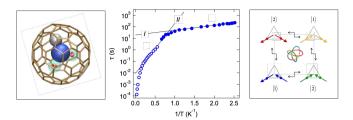
## Prof. Thomas Greber



We study objects like zero-dimensional endofullerene molecules and two-dimensional (2D) boron nitride layers in view of their functionality as nano-materials. Single-molecule magnetism is the focus of the fullerene research, where we apply x-ray absorption and a sub-Kelvin superconducting quantum interference device. In the activity of the 2D materials, we grow the highest quality boron nitride on substrates up to the four-inch wafer scale with chemical vapor deposition, subsequent exfoliation, and implementation in devices. At UZH Irchel, we use a dedicated clean room, optical microscopy, inkjet printing, and surface science tools such as low-energy electron diffraction, photoemission, and scanning tunneling microscopy for these purposes. At the Swiss Light Source, we performed photoemission and x-ray absorption spectroscopy experiments. https://www.physik.uzh.ch/groups/greber/

## Lifting the zero field degeneracy of 4f ion pairs

The single molecule magnetism in endohedral fullerenes with dilanthanides like Tb<sub>2</sub> or Dy<sub>2</sub> displays a low energy excitation that can not be understood with a pseudospin model without hybridisation. The pseudospins depict the total angular momentum states  $\pm J_z$  that impose for the dilanthanides four states  $|\uparrow\uparrow\rangle=|1\rangle$ ,  $|\downarrow\downarrow\uparrow\rangle=|2\rangle$ ,  $|\downarrow\downarrow\rangle=|\bar{1}\rangle$  and  $|\uparrow\downarrow\rangle=|\bar{2}\rangle$ . In zero field the ferromagnetic pairs  $|1\rangle$  and  $|\bar{1}\rangle$  are degenerate, as well as the two antiferromagnetic pairs  $|2\rangle$  and  $|\bar{2}\rangle$ . With an ansatz that considers quantum tunneling of the magnetization between the four pseudospin states a tunneling rate can be determined from the Arrhenius barrier in the magnetization lifetimes at sub-Kelvin temperatures. The empirical fact that the approach to thermal equilibrium contains ground state information implies that thermal fluctuations exceeding a given energy-splitting of states accelerate the equilibration.



Endohedral  $Tb_2ScN@C_{80}$ . The left panel shows the molecule with the two magnetically interacting Tb 4f 8 electron systems (red tori). Center experimental zero field magnetisation lifetimes (A. Kostanyan et al., Phys. Rev. B (2020)). Two Arrhenius processes are indicated. While II with a barrier of 10.5 K corresponds to the energy difference in a ferromagnetic or an antiferromagnetic ground state, the low energy excitation I has a barrier of 1 K and is proposed to be due to the lifting of the degeneracy by hybridisation of the four pseudospin states shown on the right.  $|1\rangle$  (yellow) and  $|\bar{1}\rangle$  (blue) are degenerate. Though, they hybridise with  $|\bar{2}\rangle$  (red) and  $|\bar{2}\rangle$  (green), which yields a non-degenerate, non-magnetic ground state. The panel shows the four states and their connections via single pseudospin flips in Hilbert space. The step arrows indicate the quantum tunneling of the magnetisation, and the different energies of the pure states. The central logo is borrowed from the 2025 unesco International Year of Quantum Science and Technology and stands here for the mixing of the four pseudospin states.

Importantly, the lifting of the zero-field degeneracy of the ground state should enable pseudospin control and manipulation.

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## **Highlighted Publications:**

- Monolayer calibration of endofullerenes with x-ray absorption from implanted keV ion doses
  W.C. Lee, L. Yu, Y. Zhang, A. A. Popov, C. Coletti, B. Delley, J. Oscarsson, M. W. Ochapski, Z. M. Gebeyehu, L. Martini, M. Muntwiler, D. Primetzhofer, and T. Greber, J. Vac. Sci. Technol. A 42, 023406 (2024)
- Quantum Tunneling of the Magnetization in Systems with Anisotropic 4f Ion Pairs: Rates from Low-Temperature Zero-Field Relaxation
  T. Greber, ACS Omega, 9, 37183 (2024)
- Benchmarking the integration of hexagonal boron nitride crystals and thin films into graphene-based van der Waals heterostructures
  T. Ouaj et al., 2D Mater. 12, 015017 (2025)