

Tracking charge, spin and light polarization in space & time in emerging semiconductors

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Solution-processable semiconductors like halide perovskites and certain molecules are promising for next-generation spin-optoelectronic applications [1]. Yet, we don't fully understand what governs spin and light polarization in such materials, and even less how these are affected by chirality [2].

In this talk, I will give an overview of our recent efforts to understand the spin-optoelectronic performance of these materials through time-, space- and polarization-resolved spectroscopy and microscopy.

For investigating halide perovskite films, we initially relied on photoluminescence (PL) microscopy to study charge carrier dynamics in high-performing mixed-halide systems, and, combined with transient absorption and other techniques showed that carrier funneling to low-energy site can have surprising consequences for solar cell and light-emitting diode (LED) performance [3].

We subsequently developed this approach further to arrive at a more complete picture enabling a time,x,y,z,-dependent 4-dimensional carrier tracking scheme to gain insights not just in the horizontal but also vertical carrier diffusivity [4]. We achieve this by measuring the thickness-dependent PL, analyzed upon varying distances from the excitation center, including from the back of the sample, and model the energy- and time-dependent carrier dynamics through a drift-diffusion-recombination approach.

Then, we wondered we what else these wonderful materials could be finding applications for beyond solar cells and LEDs and arrived at the question how electron spins are moving in such disordered semiconductors. Given the strong spin-orbit coupling in these materials, circularly polarized light can be used to optically pump spin populations and read them out, an approach that to date had only been used as a bulk read-out until then. We thus pushed such light-helicity-dependent, broadband circular dichroism (CD) to diffraction-limited spatial and 15 fs time resolution for creating a spin cinematography technique to witness the ultrafast formation of spin domains due to local symmetry breaking and spin-momentum locking in halide perovskite films [5]. While we remain agnostic about the microscopic origin of this locally varying amount of inversion-symmetry breaking, we suspect both strain fields and chemical composition disordered to be largely responsible for the observed patterns, although local halide demixing alone would be insufficient to explain our observations, given the PL energy maps don't correlate fully with the transient CD maps.

Moving beyond using circularly polarized light as only a probe to study spin-dynamics, I will then briefly explain the fundamentals and artefacts involved in measuring circularly polarized luminescence (CPL) emitted from a material reliably and introduce an open-access methodology and code to do so for the community [6].

Finally, in an effort to study dynamic phenomena related to chirality in physics and chemistry at large, we yet again strived to develop the existing spectroscopy toolkit further beyond the state-of-the-art. Thus, I will show our most recent development of a transient sensitive broadband full Stokes-vector spectroscopy technique with unprecedented time- and polarization resolution to track for the first time the emergence of chiral light emission across an excited-state lifetime with highest accuracy and across a large dynamic range [7].

[1] *Nature Reviews Materials* **8**, 365 (2023).

[2] *Nature Reviews Chemistry* **9**, 208 (2025).

[3] *Nature Photon.* **14**, 123 (2020).

[4] *Nature Mater.* **21**, 1388 (2022).

[5] *Nature Materials* **22**, 977 (2023).

[6] *Advanced Materials* **35**, 2302279 (2023).

[7] *Nature* **643**, 675 (2025).

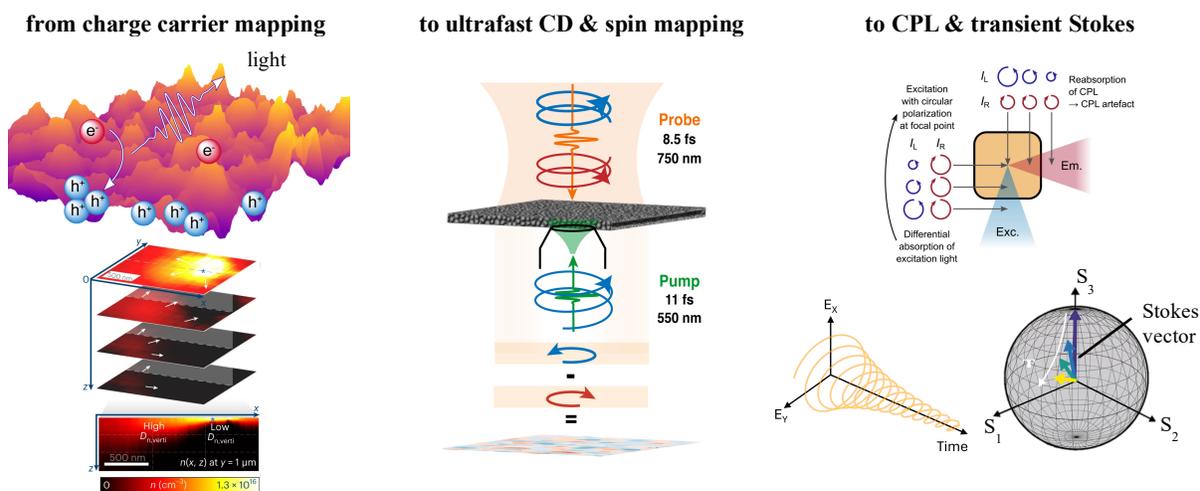


Figure 1: Our group employs and develops time-resolved optical spectroscopy to study and ultimately control charge, spin and light polarization dynamics in emerging semiconductors.

Background Information:

The Feldmann Lab at EPFL develops and employs ultrafast magneto-chiroptical spectroscopy to understand the next generation of soft semiconductors. The overarching goal is to maximize energy efficiency for a sustainable future by unlocking applications ranging from flexible light-weight solar cells & displays all the way to entirely new applications in quantum information processing.

Sascha studied Chemistry at Heidelberg University (Germany) and completed his PhD in Physics at the University of Cambridge (UK) in 2020, where he continued to work as an independent EPSRC Doctoral Prize Fellow. In July 2022, Sascha started his own group at Harvard University through a Rowland Fellowship. Since 2024 he is a tenure-track assistant professor and head of the Laboratory for Energy Materials at EPFL, while also remaining affiliated as SCR member of Winthrop House at Harvard.

