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## Revealing spin polarization in compensated spin systems on ultrafast timescales

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Fundamental to the advancement of spintronics and quantum technology in solids is the ability to encode, manipulate, and store information about the spin angular momentum of electrons on ever faster timescales. To date, ferromagnets have been the natural driving target for these efforts, leading to optical manipulation schemes that can alter the spin order on the timescale of optical excitation. However, intrinsic limitations of ferromagnets regarding miniaturization and efficiency have prompted the search for compensated spin systems that can still support (ultrafast) spin functionalities.

In this contribution, we will introduce two different classes of compensated spin systems as highly flexible platforms to control the spin polarization and spin functionalities of optically excited charge carriers on ultrafast timescales using fs light pulses.

First, I will focus on heterostructures consisting of molecules and non-magnetic transition metal dichalcogenides (TMDs) with hidden spin polarization [1]. Such hidden spin polarizations arise in centrosymmetric layered TMDs such as WSe<sub>2</sub>, which contain subunits with spin polarization bands that compensate each other overall, see Fig. 1a. Using time-, spin- and momentum-resolved photoemission, I will show how interfacial charge transfer processes between the optically excited molecule and the prototypical TMD WSe<sub>2</sub> can lead to a layer-dependent shift of the WSe<sub>2</sub> valence band structure, ultimately revealing the hidden spin polarization of the system on a fs timescale [2].

Furthermore, I will provide insights into the ultrafast response of altermagnets - a new class of compensated magnets with a momentum-split band structure. For the case of RuO<sub>2</sub> [3], we combine ab initio calculations with time-resolved MOKE experiments (see Fig. 1b) to probe the optically excited spin polarization in this compensated magnet. We find a clear correlation between the sign and magnitude of the optically generated spin polarization and the direction of the light polarization vector of the exciting light pulses with respect to the RuO<sub>2</sub> crystal structure. This provides a clear strategy for the optical generation and control of spin-polarized carriers in altermagnets.

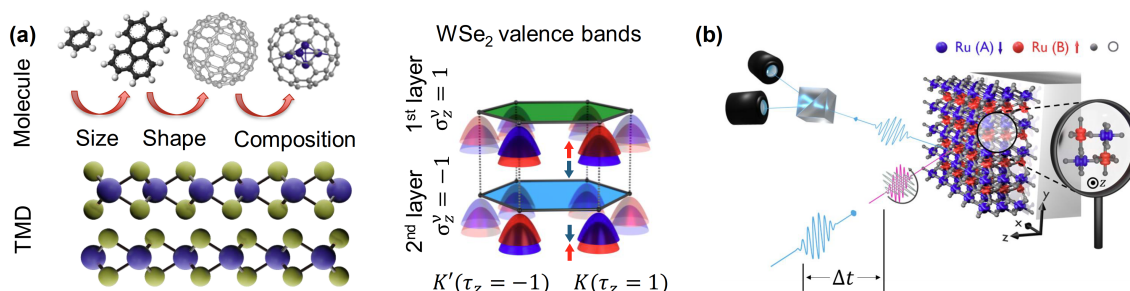


Figure 1: (a) Graphical representation of the tunability of molecular TMD heterostructures and sketch of the layer- and spin-dependent valence band structure of the two noninteracting WSe<sub>2</sub> layers of the bulk unit cell, in which the spin polarization vanishes at each point in the Brillouin zone. (b) Illustration of the pump-probe approach to explore the excited state spin polarization of RuO<sub>2</sub> after fs light pulse excitation.

- [1] J. M. Riley et al Nat. Phys. 10, 835–839 (2014).  
[2] B. Arnoldi et al. Nat. Commun. 15, 3573, (2024).  
[2] O. Fedchenko et al. Sci. Adv. 10, eadj4883 (2024).

Für diese Zeit steht eine Kinderbetreuung nach vorheriger Anmeldung zur Verfügung.

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