Imaging valence and excited states of fullerenes in momentum space

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One of the key milestones in advancing the performance of molecular electronic and photonic devices is to gain a comprehensive understanding of the electronic properties and rich excited state dynamics of this class of materials. In this context, momentum-resolved photoemission in combination with photoemission orbital tomography (POT) has been established as a powerful tool to study the band structure of molecular films and to reveal the degree of localization of molecular valence orbitals by their characteristic emission pattern in momentum space.

In this contribution, we exploit these capabilities of POT to study the valence and excited states of fullerenes grown on noble metal surfaces. For the most prototypical fullerene, the buckyball C_{60}, we will show that the valence states show signatures of an atomic crystal-like band structure with delocalized π- and localized σ-orbitals [1]. The situation is significantly different for thin films of larger fullerenes such as C_{84} or of the endohedral fullerene $Sc_3N@C_{80}$, where the valence states are strongly localized on the carbon cage of the molecules.

In addition, we provide a first insight into the momentum space signatures of the excited state dynamics of C_{60} and other fullerenes obtained by time-resolved two-photon momentum microscopy [2]. For an optical excitation with 3.1eV photons, we are able to identify characteristic emission patterns even in the small momentum space range accessible our experiment. These signatures are discussed in the context of the recently proposed charge transfer and Frenkel exciton character of these states [3,4].

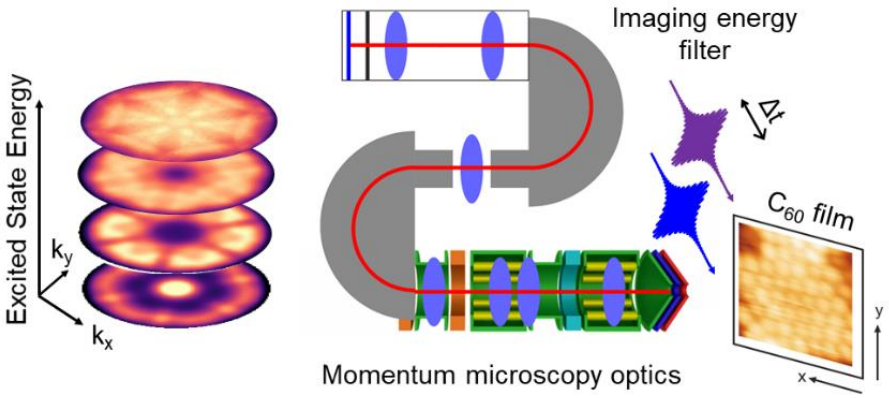

Fig. 1. Schematic sketch of the tr-2-photon momentum microscopy experiment for excited state spectroscopy.

[1] N. Haag et al., Phys. Rev. B 101, 165422 (2020)
[2] F. Haag et al., Rev. Sci. Instrum. 90, 103104 (2019)
[3] B. Stadtmüller et al., Nat. Commun. 10, 1470 (2019)
[4] S. Emmerich et al., J. Electron. Spectros. Relat. Phenomena 252, 147110 (2021)