The excitons that you might not expect

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Excitons are a key to explore two-dimensional (2D) layers and nanosystems: Since the beginning of bottom-up nanoribbon fabrication, the predicted excitonic signatures are opening the way to understanding the structure [1], the growth process [2], the edge and distortion design [3], as well as many other phenomena of central importance for applications [4]. First-principles theory and computation beyond density functional theory is the main tool enabling such advances.

Excitons of 2D systems are now the leading actor also in a new movie, maybe less expected within the Chem2Dmat community: the Excitonic Insulator (EI). The name refers to the long-sought state of matter that may result when the electron-hole binding energy exceeds the band gap: in this case, a new excitonic ground state would from spontaneously, in analogy with the condensation of Cooper pairs in a superconductor. The search of the EI has long been hindered by the lack of materials with sufficient exciton binding, which in most systems scales with the gap size due to screening. This is why dimensionality and environment of 2D systems are central.

I will discuss how the EI phase is indeed realized in two-dimensional systems where screening of long-range Coulomb interaction is strongly reduced, and exciton binding is greatly enhanced. Using many-body perturbation theory, we single out the best candidate materials and discriminate between spurious insulating behaviour and genuine EI fingerprints, which are solely due to electron-electron interactions. We show that transition metal dichalcogenides are excellent candidates for the EI realization. In the case of MoS_2, we analyze the EI formation both in the monolayer [5] and in the bulk phase at high pressure [6]. The latter is found to be prone to the condensation of genuine excitons of finite momentum above 30 GPa, whereas the phonon dispersion remains regular; the EI sustains an out-of-plane permanent electric dipole moment with an antiferroelectric texture in the layer plane. We identify a unique Raman fingerprint for the EI formation and recognize that it was previously observed experimentally, thus providing direct spectroscopic confirmation of an ideal excitonic insulator phase in MoS_2.

Work in collaboration with D. Prezzi, M. Rontani, D. Varsano. Thanks to the whole CNR-Nano - UniMoRe team for many discussions and contributions. Work supported in part by EXC-INS (PRIN Project n. 2017BZPKSZ) and MaX-Materials design at exascale (European Centre of Excellence for HPC applications, EU grant n. 824143).

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