X Workshop on Novel Methods for Electronic Structure Calculations
04th – 06th December 2023
La Plata – Argentina

Magnetic order, magnetic excitations and magnetoelastic interactions in a two-dimensional van-der Waals system

D. Vaclavkova [a], C. Faugeras [a], M. Potemski [a], A. R. Wildes [b], B. Grémaud [c], Andrés Saúl [d],

[a] *Laboratoire National des Champs Magnétiques Intenses, CNRS-UGA-UPS-INSA-EMFL, Grenoble, France.*

[b] *Institut Laue-Langevin, Grenoble, France.*

[c] *Aix Marseille Université, Université de Toulon, CNRS, CPT, Marseille, France. MajuLab, CNRS-UCA-SU-NUS-NTU International Joint Research Unit, 117542 Singapore Centre for Quantum Technologies, National University of Singapore, 2 Science Drive 3, 117542 Singapore.*

[d] *Aix-Marseille Université, Centre Interdisciplinaire de Nanoscience de Marseille-CNRS (UMR 7325), Marseille, France.*

email:

The discovery of graphene, followed by the development of exfoliation techniques of other van der Waals (vdW) systems, has motivated a great interest to study the unique properties of quasi two-dimensional (2D) compounds. Interestingly, many vdW compounds are magnetic and antiferromagnetic (AFM) order is very often observed in semiconductors with relatively large effective interactions resulting on intermediate transition temperatures (75 - 210 K) and AFM resonances in the THz range close to the optical phonon's frequencies. The van der Waals antiferromagnetic systems thus represent an extremely rich playground for studying non-trivial interactions between electronic, magnetic, and vibrational properties with potential applications.

In this work, we are interested in $MnPS_3$, a member of the metal phosphorus tri-chalcogenides family MPX_3 (M = Fe, Co, Ni, M and X = S, Se). The magnetic centers in $MnPS_3$ are the Mn^{2+} ions. They form a distorted honeycomb lattice showing AFM Néel order below 78 K with magnetic moments perpendicular to the planes.

In the first part of the talk, I will discuss the magnetic order and the physical origin of the magnetic anisotropy taking into account that one expects a negligible spin-orbit coupling associated to the 3d5 electronic configuration of the Mn^{2+} ions. I will compare single ion and magnetic dipolar contributions to available magnon band and magnetic spin resonance measurements.

In the second part, I will show Raman scattering measurements as a function of temperature where some of the modes show a blue shift when the temperature is raised and cross the AFM-paramagnetic transition. I will present density functional theory (DFT) calculations of the effective exchange interactions and Raman active phonon modes which show that the peculiar behavior with temperature of the low energy phonon modes can be explained by the symmetry of their corresponding normal coordinates which involve the virtual modification of the super-exchange angles associated with the leading antiferromagnetic (AFM) interactions.