Tuning dimensionality, magnetism and conduction in van-der-Waals Mott insulators

M.J. Coak*[,1;2], D.M. Jarvis[2], H. Hamidov[2;3;4], A.R. Wildes[5], J.A.M. Paddison[6;2], C. Liu[2], C.R.S. Haines[7;2], N. T. Dang[8;9], S.E. Kichanov[10], B.N. Savenko[10], S. Lee[11;12], M. Kratochvilova,[11;12;13], S. Klotz[14], T. Hansen[5], D.P. Kozlenko[10], J.-G. Park[11;12], and S.S. Saxena[2;4]

[1]*Department of Physics, University of Warwick, Gibbet Hill Road, Coventry CV4 7AL, UK*
[2]*Cavendish Laboratory, Cambridge University, J.J. Thomson Ave, Cambridge CB3 0HE, UK*
[3]*Navoi State Mining Institute, 27 Galaba Avenue, Navoi, Uzbekistan*
[4]*National University of Science and Technology "MISiS", Leninsky Prospekt 4, Moscow 119049, Russia*
[5]*Institut Laue-Langevin, CS 20156, 38042 Grenoble Cedex 9, France*
[6]*Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*
[7]*Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge CB2 3EQ, UK*
[8]*Institute of Research and Development, Duy Tan University, 550000 Da Nang, Viet Nam*
[9]*Faculty of Natural Sciences, Duy Tan University, 550000 Da Nang, Vietnam*
[10]*Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, 141980 Dubna, Russia*
[11]*Center for Correlated Electron Systems, Institute for Basic Science, Seoul 08826, Republic of Korea*
[12]*Department of Physics and Astronomy, Seoul National University, Seoul 08826, Republic of Korea*
[13]*Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Charles University, Prague, Czech Republic*
[14]*Sorbonne Universite, IMPMC, CNRS, UMR 7590, 4 Place Jussieu, 75252 Paris, France*

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*e-mail: matthew.coak@warwick.ac.uk

The van-der-Waals antiferromagnets *TM*PS_3, where *TM* = Transition Metal, form an ideal playground for tuning both low-dimensional magnetic and electronic properties [1-4]. These are layered honeycomb antiferromagnetic Mott insulators, long studied as near-ideal 2D magnetic systems with a rich landscape of competing interactions and a variety of magnetic properties across the family.

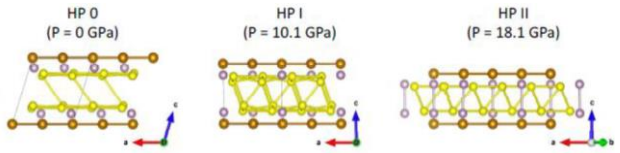

Figure 1. Evolution of the layered crystal structure of $FePS_3$ with increasing pressure [3].

I will give an overview of our work using high pressure as a continuous tuning parameter to control the dimensionality of these materials. Due to the weak physical inter-planar forces in such van-der-Waals materials, pressure gives us clean and selective control over the inter-planar spacing and hence interactions.

I will present magnetic, structural and electrical transport results and compare the behaviour of Fe-, V-, Mn- and $NiPS_3$ as we tune them towards 3D structures – and Mott transitions from insulator to metal. I will focus in particular upon recent results on ultra-high-pressure neutron scattering, which has unveiled an enigmatic form of short-range magnetic order in metallic $FePS_3$. This phase is particularly important as it most likely forms a precursor to superconductivity.

These neutron diffraction results made use of new cutting-edge experimental techniques [5] which have allowed measurements of magnetic structure through powder diffraction up to 20 GPa at the ILL - smashing the previous 10 GPa record.

Piecing together our high-pressure neutron, transport and x-ray results has allowed us to map out the full phase diagram - a first in this crucial family of materials, and most likely to become the archetypal example. We observe multiple transitions and new states, and an overall increase in dimensionality and associated changes in behaviour.

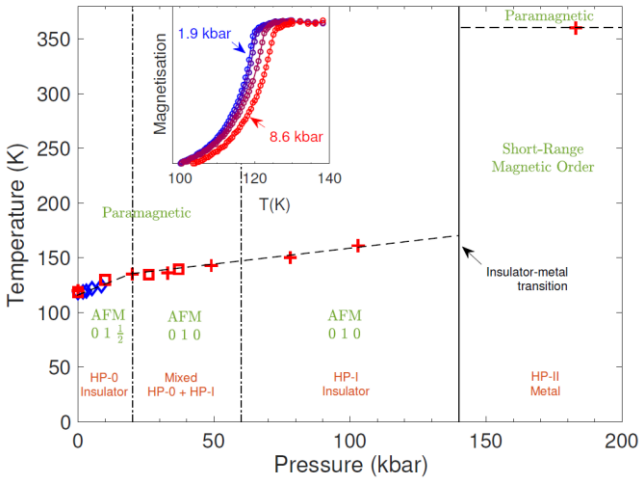

Figure 2. Newly uncovered pressure-temperature magnetic phase diagram of FePS3 - the first in the *TMPX*$_3$ family. Within the strange metallic state at high pressure, a novel short-range-ordered magnetic state is discovered.

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