Structure Analysis of Metal-Chalcophosphate Layered Systems Using Micro-ED

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Metal chalcophosphates, M2P2Q6 (M = transition metals; Q = chalcogen), are notable among the van der Waals materials family for their potential magnetic ordering that can be tuned with an appropriate choice of the metal or chalcogen. Their hexagonal layered systems are composed of "ethane-like" [P2Q6]4– units, where each P is tetrahedrally bonded to three S and one P atom with a honeycomb extended structure. Each metal cation fills 2/3 of the octahedral sites, while the remaining 1/3 is filled by the P–P dimers. The P–P dimer bond distance is dependent on the metal cations, which can lead to the stretching or shrinking of the entire crystal structure. Amongst Metal chalcophosphates, Fe2P2S6 and Co2P2S6 show unique magnetic properties; magnetic ordering in Fe2P2S6 has been reported in several studies, and transformations at the Néel temperature (TN) have been reported accompanied by a structural distortion. In addition, alloying different metal cations within the metal thiophosphates leads to altered magnetic properties.

In this work we investigate the structural details of an alloyed Fe2P2S6 and Co2P2S6 system using micro electron diffraction (micro-ED). Micro-ED is one of the very few methods that provide both in- and out-of-plane structural information, which is indispensable for layered materials, such as the one studied here. We utilized Gatan's hybrid pixel electron detector (Stela camera) in conjunction with DigitalMicrograph Latitude D software to automatically collect datasets from multiple pre-selected areas of the sample. After data acquisition, diffraction intensities were extracted using MOSFLM followed by structure solution performed using JANA software.

This work provides a fundamental structural framework for mixed metal thiophosphate systems, which can assist in future studies on electronic and magnetic applications of this emerging class of binary cation materials.