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Magnetic structure determination and putative anisotropic exchange coupling in the hybrid organic-inorganic perovskite (CH₅NH₂NH₂NH₃)₂MnCl₄

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The interest in low-dimensional magnetism (LDM) is manifold, but can be summarized in two categories. First, it provides a rich test bed for fundamental research in many-body quantum phenomena to study the influences of restricted exchange pathways, frustration and enhanced quantum fluctuations. Second, LDM has been shown to have links with various phenomena such as high-temperature superconductivity, fractional excitations, spin liquids and multiferroicity, which are at the forefront of global solid-state research.

Metal-organic compounds (MOC) serve as an essentially infinite toolbox for building model systems for LDM research. Owing to their modular structure, the strength, type and direction of magnetic exchange can be tuned by the ligand molecules, which connect the magnetic ions. The perovskite Mn-PEA [fully (C₆H₅CH₂CH₂NH₃)₂MnCl₄] is considered to be one of the ideal layered 2D antiferromagnets, minimizing out-of-plane interactions by separating magnetically active Mn²⁺ ions by long organic chains.

We have successfully performed neutron diffraction (D10+, ILL) and inelastic neutron scattering experiments at the cold multiplexing spectrometer HODACA (JRR-3) and TAS (JRR-3, HANARO) on non-deuterated Mn-PEA single crystals to determine the magnetic structure and its spin wave excitations. The magnetic moments are antiferromagnetically aligned along the *c*-axis, with a slight canting into the *ab*-plane. The spin wave dispersion is completely *l*-independent which is characteristic of negligible exchange interactions across the separation layer of organic ligands. Using spinW, we apply linear spin wave theory including exchange coupling up to the next-nearest neighbors and DMI between nearest neighbors to account for the observation of a second dispersion branch along *h*.

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