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Magnetism in a family of $S = 1$ square lattice antiferromagnets, NiX_2(pyz)$_2$ (X = Cl, Br, NCS; pyz = pyrazine) J. LIU, University of Oxford, J.L. MANSON, T.J. WOODS, K.E. CARREIRO, H.E. TRAN, Eastern Washington University, S.H. LAPIDUS, K.H. STONE, P.W. STEPHENS, State University of New York, Y. KOHAMA, Los Alamos National Laboratory, J.S. MOELLER, University of Oxford, F.L. PRATT, P.J. BAKER, ISIS, T. LANCASTER, A. ARDAVAN, S.J. BLUNDELL, University of Oxford, J. SINGLETON, Los Alamos National Laboratory, P.A. GODDARD, University of Warwick — The crystal structures of NiX_2(pyz)$_2$ (X = Cl, Br and NCS, henceforth Ni-Cl, Ni-Br and Ni-NCS, respectively), were determined at 298 K from synchrotron powder X-ray diffraction data. All three compounds consist of two-dimensional (2D) $[\mathrm{Ni(pyz)_2}]^{2+}$ square lattices spaced by X ligands, resulting a staggered stacking fashion of 2D layers. Long-range antiferromagnetic order occurs below 1.5 (Ni-Cl) and 1.9 K (Ni-Br and Ni-NCS) as determined by heat capacity and Muon-spin relaxation. The single-ion anisotropy and g factor of Ni-Br and Ni-NCS were measured by electron spin resonance where no zero-field splitting was found. The magnetism of Ni-NCS is interpreted by the 3D simple cubic Heisenberg model with the Ni-pyz-Ni interaction $J_{pyz} = 0.70$ K. A good overall agreement was found between the pulsed field magnetization data, magnetic susceptibility and T_N for Ni-NCS. Ni-Cl and N-Br are characterized as quasi-2D antiferromagnets with the interlayer magnetic coupling significantly suppressed by varying the X ligand.

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