Spin stripe order in a square planar trilayer nickelate

J. Zhang,[1,2] D.M. Pajerowski,[3] A.S. Botana,[1,4] H. Zheng,[1] L. Harriger,[5] J. Rodriguez-Rivera,[5,6] J.P.C. Ruff,[7] N.J. Schreiber,[8] B. Wang,[1] Y.-S. Chen,[9] M.R. Norman,[1] S. Rosenkranz,[1] J.F. Mitchell,[1] and D. Phelan[1]

Like medieval knights chasing the *Holy Grail*, condensed matter physicists have long been allured by the glitter of high temperature superconductivity. The most famous advance in this effort was the Nobel-prize winning discovery in 1986 by Bednorz and Müller of superconductivity in layered copper oxides. The ensuing years have revealed that these materials have certain common features that seem to be part of the 'recipe' for copper oxide superconductivity: they are layered materials containing Cu-O planes; they consistently possess the same number of electrons in the copper 3d orbitals, which are strongly polarized with holes in the $d_{x^2-y^2}$ orbitals; and they are proximal to states where the electron spins order antiferromagnetically. Finding similar superconductivity in other transition metal oxides has thus far proven fruitless, but are copper oxides really the only ones? Nickel, which sits one column to the left of copper on the periodic table, has long been considered a tantalizing alternative for achieving superconductivity in a copper-free transition metal oxide. Recently, trilayer nickelates with the chemical formula $R_4Ni_3O_8$ (where R is a trivalent rare earth), which possess low valence nickel cations in square planar coordination, are particularly interesting candidate materials because they possess many of the same characteristics identified above as common to copper oxide superconductors [1]. Given the importance of magnetic interactions in the copper oxide superconductors, as well as in the iron pnictide superconductors, unraveling the magnetism in these trilayer nickelates is essential for understanding their behavior as well as their potential as superconductors. However, relatively few detailed investigations have been performed because single crystals have been unavailable, leading to a critical gap in our understanding of the magnetic behavior of these compounds.

Recently, single crystals of $La_4Ni_3O_8$ (La-438) have become available as a result of the advent of high-pressure floating zone technology. La-438 crystals exhibit a semiconductor-insulator transition on cooling that is concomitant with the formation of charge stripes of higher and lower oxidation state on the nickel sites [2]. One of the major questions remaining is whether or not there is magnetic ordering in the ground-state, and if there is, how it relates to the charge ordering that is observed. Unfortunately, previous neutron powder diffraction experiments proved inconclusive. No magnetic Bragg reflections were observed from polycrystalline materials, either because the peaks were too weak to be detected by powder diffraction, or because the system did not develop long-range order. In order to resolve this quandary, single crystal neutron diffraction measurements were performed on MACS [3], which was chosen because of its large cold neutron flux, which is beneficial for investigating small single crystals with weak magnetic moments, and because of its spin-polarization capabilities.

Weak satellite reflections were observed in the two-dimensional plot of neutron intensity in the ground-state (Fig. 2A). Spin-polarized diffraction data, collected on MACS using the ^{3}He polarizers and analyzers, conclusively evidenced a magnetic origin to the scattering – thus proving that $La_4Ni_3O_8$ does have a magnetically-ordered ground-state (Fig. 2B, C). Using the charge-stripe state as a starting point, an antiferromagnetic spin-stripe-ordering model was developed in which ordered spins are present on Ni^{1+} sites (S = 1/2) and absent on Ni^{2+} sites (S = 0), as shown in Fig. 3. The nearest neighboring Ni^{1+} sites have antiferromagnetic interactions within the basal plane, as well as antiferromagnetic interactions out of the basal plane. The data are consistent with a quasi-two-dimensional ordering that occurs within Ni-O trilayers that are uncoupled along the c-axis.

Surprisingly, several characteristics were discovered that distinguish $La_4Ni_3O_8$ from copper oxides or single-layer nickelates. First, in these latter compounds, charge is the primary order parameter, and spin is a secondary order parameter that follows the charge. However, for $La_4Ni_3O_8$

1 Argonne National Laboratory, Lemont, IL 60439
2 Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
3 Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
4 Arizona State University, Tempe, AZ 85287
5 NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899
6 University of Maryland, College Park, MD 20742
7 CHESS, Cornell University, Ithaca, NY 14853
8 Cornell University, Ithaca, NY 14853
9 The University of Chicago, Lemont, IL 60439

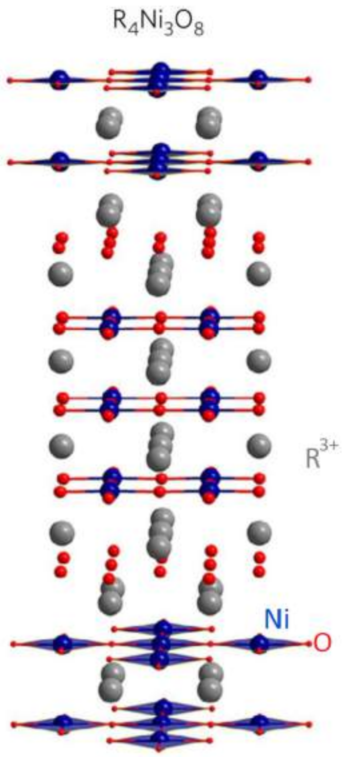

FIGURE 1: Structural motif of $R_4Ni_3O_8$ compounds (R = La, Pr, Nd). After [1].

the strong coupling between the charge and spin leads to a situation in which neither order parameter appears secondary to the other. Second, both the polarized data as well as density functional theory calculations indicate that the ordered spins point parallel and antiparallel to the c-axis. This contrasts with other layered oxides, where the spin prefers to orient in the basal plane.

These neutron scattering experiments have laid a foundation for our understanding of the magnetic correlations in $La_4Ni_3O_8$, which possesses an electron count that matches the over-doped regime of the superconducting cuprates. These experiments have revealed how a complex interplay of the spin, charge, and orbitals results in coupled charge and spin stripes. The next challenge in this work is to modify the electron count so that it overlaps the optimal concentration of the copper oxide superconductors and then to study how the magnetic and charge interactions respond.

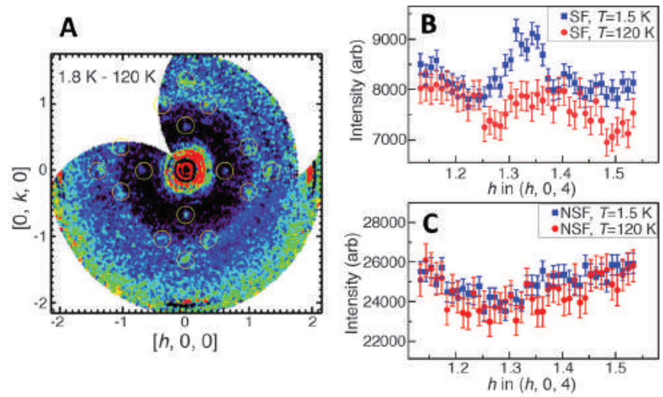

FIGURE 2: (A) Unpolarized neutron diffraction data on a specimen of $La_4Ni_3O_8$ showing weak superlattice peaks in the ground-state. (B, C) Observation of the peak in the Spin-Flip (SF) channel evidences magnetic ordering. After [3].

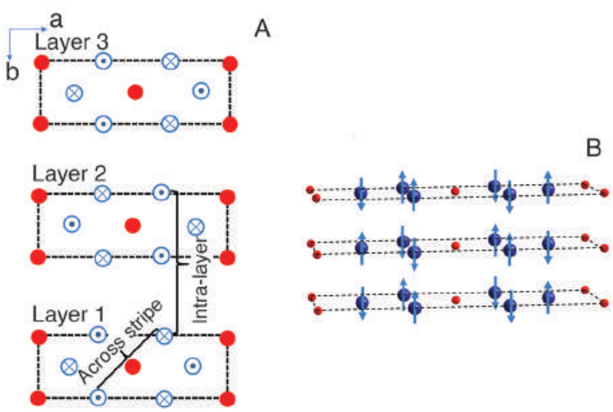

FIGURE 3: Spin-stripe pattern. Blue spheres refer to Ni^{1+} cations, and red spheres to Ni^{2+} cations. After [3].

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