Lack of Out-of-Plane Dispersion of the Magnetic Excitations of Charge-Stripe Ordered $La_{2-x}Sr_xNiO_{4+\delta}$.

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The dependence of the magnetic excitations in the out-of-NiO-plane direction of charge stripe ordered $La_{2-x}Sr_xNiO_{4+\delta}$ $x = 0.3$ and $\delta = 0.11 \pm 0.01$ were investigated by inelastic neutron scattering. No dispersion was observed for the magnetic excitations in the out-of-plane $(0, 0, l)$ direction of reciprocal space of $La_{2-x}Sr_xNiO_{4+\delta}$, indicating an upper limit of the out-of-plane spin interaction of $J_\perp < 0.25$ meV.

Charge order, dynamic charge stripes, spin stripes and high temperature cuprate superconductivity remain a highly active field of research decades after the discovery of charge-stripe order in a La-based cuprate.[1)] Extensive research has been undertaken to understand insulating charge ordered materials to aid this research.[2,3)] With the vast amount of research conducted into both charge ordered materials and cuprates it is important to keep a clear understanding of what points have been proven, and what are reasonable assumptions that are made about these materials.

Recent studies of $Pr_{3/2}Sr_{1/2}NiO_4$ have highlighted the potential importance of inter-layer spin interactions in layered charge order materials.[4)] Magnetic excitations in the equivalent material $La_{2-x}Sr_xNiO_{4+\delta}$ (LSNO) have been reported (without data shown) to show no dispersion of the magnetic spin excitations in the out-of-Ni-O plane direction,[5,6)] consistent with limited data on La_2NiO_4.[7)] In this paper we report inelastic neutron scattering scans of the magnetic excitations of two doping levels of charge-stripe ordered LSNO, consistent with a lack of a significant inter-layer spin interaction in LSNO.

We chose to study an oxygen doped, and a Sr doped sample due small differences in their ordered structure.[2,8)] Large single crystals of $La_2NiO_{4.11\pm0.01}$ ($\delta = 0.11$) and $La_{1.7}Sr_{0.3}NiO_4$ ($x = 0.3$) were grown using the floating-zone technique,[9)] with oxygen content of as grown crystals determined by thermogravimetric analysis.[10)] Both crystals were approximately 15 g rods of 45 mm length and 8 mm diameter, with the $\delta = 0.11$ crystal having been previously studied.[11)] Inelastic neutron scattering on the $\delta = 0.11$ were taken on IN3 at the Insitiut Laue-Langevin, and on the $x = 0.3$ on TASP, at the SINQ neutron source of the Paul Scherrer Institut. The monochromator was vertically focused and the analyzer horizontally focused on both instruments. The sample was orientated on both instruments so that (h, h, l) positions in reciprocal space could be accessed, referenced with the tetragonal unit cell for the $x = 0.3$ $a = b = 3.84(1)$ Å, $c = 12.7(1)$ Å and for the $\delta = 0.11$ $a = b = 3.87(1)$ Å, $c = 12.6(1)$ Å. In both the $x = 0.3$ and $\delta = 0.11$ the magnetic excitations disperse up to at least 80 meV.[10,12)]

The $x = 0.3$ was characterised by neutron diffraction during the TASP experiment with; a spin ordering temperature $T_{SO} = 162.5 \pm 2.5$ K, a spin reorientation temperature of $T_{SR} = 18 \pm 1.5$ K, and the magnetic order at base temperature has an incommensurability of $\varepsilon = 0.317 \pm 0.001$. This is consistent with a stoichiometric oxygen content.[13)]

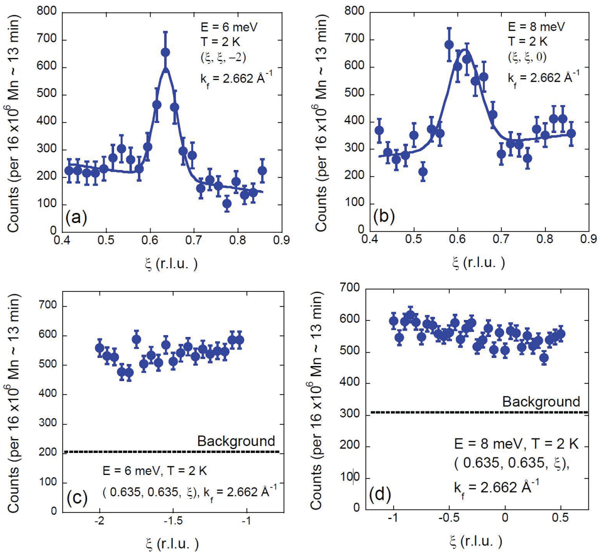

Fig. 1. (Color online) (a, b) constant energy scans of the magnetic excitations of $La_2NiO_{4.11\pm0.01}$ scanned parallel to $(h, h, 0)$ at measured at $l = -2$ and $E = 6$ meV, and $l = 0$ and $E = 8$ meV respectively. (c, d) constant energy scans at the centres of the magnetic peaks in (a) and (b) scanned in the $(0, 0, l)$ showing no variation with l.

In figure 1 we show inelastic neutron scans of the magnetic excitations of the $\delta = 0.11$ that pass through the magnetic zone centre at $E = 6$ meV and $E = 8$ meV scanned parallel to (hh0) (a-b) and parallel to (00l) (c-d) respectively, above the out-of-plane anisotropy gap at 2 K. The scans parallel to (hh0) both show a peak in the magnetic excitations, where as the scans parallel to (001) have an intensity that is independent of l.

In figure 2 we show similar scans of the magnetic excitations of the $x = 0.3$ as those shown for $\delta = 0.11$, both above (6 meV) and below (1.2 meV) the out-of-plane

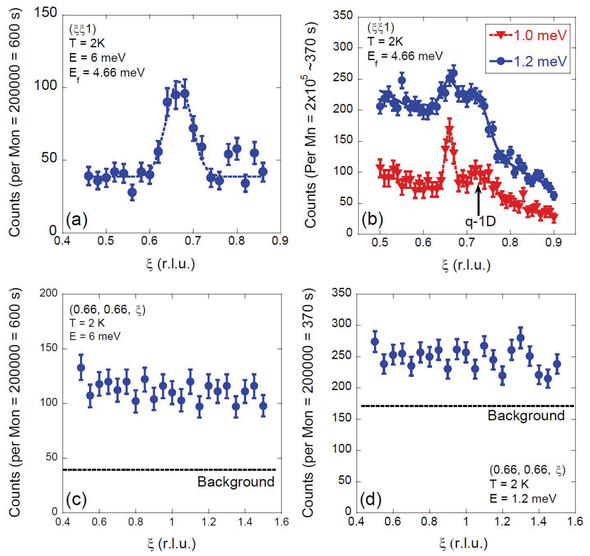

Fig. 2. (Color online) (a, b) constant energy scans of the magnetic excitations of $La_{1.7}Sr_{0.3}NiO_4$ scanned parallel to $(h, h, 0)$ measured at 6 meV for $l = 1$, and 1.0 meV for $l = 3$ & 1.2 meV for $l = 1$ respectively. For 1.0 meV & 1.2 meV an additional q-1D magnetic excitation from the spins of the charge-stripe electrons is also observed.[5,14,15] (c),(d) constant energy scans at the centres of the magnetic excitations from the order spins in (a) and (b), for both 6 meV and 1.2 meV scanned parallel to $(0, 0, l)$ no variation with l is observed.

anisotropy gap. In the scans parallel to $(hh0)$ the magnetic excitations from the ordered spin stripes produce a peak centred at $h = 0.66$. Additionally at 1.2 meV there is an excitation peak centred at $h = 0.73$ consistent with a gaped quasi-1 dimensional antiferromagnetic excitations (q-1D) from the spins of the charge stripe electrons, observed for other doping levels.[5,14,15] An additional scan at 1.0 meV clearly resolves the q-1D from the magnetic excitations from the ordered spins. Consistent with the $\delta = 0.11$ the magnetic excitations from the spin stripes of $x = 0.3$ are peaked in scans parallel to $(hh0)$ but lack an l dependence, both above and below the out-of-plane anisotropy gap for $x = 0.3$.

We note that in the $x = 0.3$ the magnetic excitations from the spin stripe order are centred on the magnetic Bragg peaks, unlike the small offset wave vector in the $\delta = 0.11$.[16] This is suggestive that the offset in the $\delta = 0.11$ is associated with the lattice distortion caused by the excess oxygen supercell structure.[8,17]

Unlike the magnetic excitations of $Pr_{3/2}Sr_{1/2}NiO_4$ which have an l dependence and dispersion, the magnetic excitations of $La_{2-x}Sr_xNiO_{4+\delta}$ are observed here to be featureless in this direction consistent with a lack of dispersion. An upper estimate on the out-of-plane spin interaction can be obtained by assuming the dispersion occurs at lower energy, with 1 meV bandwidth = $4J_\perp$, meaning $J_\perp < 0.25$ meV. We therefore conclude that any out-of-plane spin interaction is no more than a perturbation on the in-plane spin interactions, which were determined in earlier studies.[5,10,12]

We note a similar finding is observed for several doping levels of $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Ba_xCuO_4$ for $x = 0 - 0.11$,[18–20] and in a limited set of data on the charge ordered cobaltate $La_{5/3}Sr_{1/3}CoO_4$.[21] For $Pr_{2-x}Sr_xNiO_4$ the determination of the magnetic state of the Pr may be illuminating on the origin of the out-of-plane spin wave dispersion, considering Pr contributes to the magnetic structure of Pr_2NiO_4.[22,23]

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