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Report:

We have measured infinite-layer nickelates thin films (around 10 nm in thickness), with three different Sr doping levels, *i.e.* 0 %, 5 % and 20 %. From the transport point of view, the sample with 20% Sr doping is the only to exhibit a transition toward the superconducting state at around 10 K, while the 5% is a bad metal and the undoped sample shows an insulating behaviour within the entire measured range (20-300 K). This last result, while corroborates the bulk finding [Hayward, M. A. & Rosseinsky, M. J. *Solid State Sci.* **5**, 839–850 (2003)], it is at odd with the only (up to now) report for thin films for which a bad-metal behaviour is displayed also for the undoped case. Our RIXS and XAS (TEY) measurements were performed at 20 K for a series of infinite-layer nickelate thin films prepared without capping layer. In literature [Hepting *et al*. Nat. Mat. (2020) and Rossi *et al.* ArXiv (2020)], the most recent RIXS studies on infinite-layer nickelate thin films are using samples with a $SrTiO_3$ capping layer, and independently from the Sr-doping level, the O K-edge XAS spectra do not show any pre-peak structure. It is nevertheless speculated that such a finding might be somehow strictly related to the presence of capping layer. In order to get useful insight on this point we decided to perform the topotactic reduction without capping layer. Theoretical results give a larger charge transfer value if compared with cuprates (4 eV against 2 eV) in undoped infinite-layer nickelates. This larger charge transfer value is necessarily weakening the Ni 3d-O 2p hybridization degree, but a close inspection of the DFT+U calculations [Karp et al. PRX (2020)] help to delineate a rather different reality with the O2p bands which are smoothly bleeding over E_F and, hence, denoting a non-zero overlaps with the Ni 3d bands. Interestingly, in our series of samples we could observe such a pre-peak structure in the O K-edge spectra. Figure 1 shows the pre-peak structure of the O K-edge spectra after an opportune baseline subtraction in order to compare the relative intensity of the measured features as s function of the Sr-doping. Our preliminary result/analysis point at a more cuprate-like behaviour [CT Chen at al. PRL (1991)] of the doping effect, that bring the hole to reside not simply in the Ni-3d sector but, most likely, in the Ni 3d-O 2p charge transfer states.

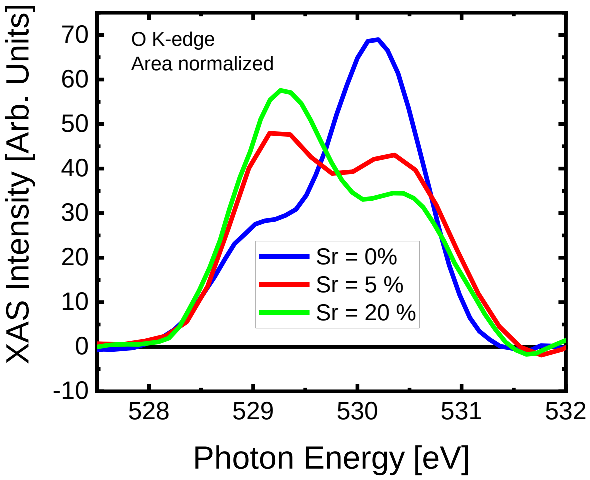

Figure 1 O K-edge pre-peak structure for three different Sr doping content of infinite-layer nickelate thin films after a base line subtraction.

XAS data acquired at the Ni L_3-edge (TEY mode), confirmed the expected low Ni valence state value ($Ni^{(1+x)+}$), and the square-planar geometry, although not so robust, for the infinite-layer phase. A close inspection of such spectra rendered also important information about the spatial anisotropy of the Ni 3d derived states. A pronounced shoulder around 852.6 eV is present only for light polarized in the out-of-plane direction (with respect to the sample surface), and we can associate this low energy shoulder to an anisotropic hybridization degree between Nd $5dz^2$ and Ni $3dz^2$ states especially for the undoped and 5% Sr-doped samples that are no superconducting. Figure 2 shows these results.

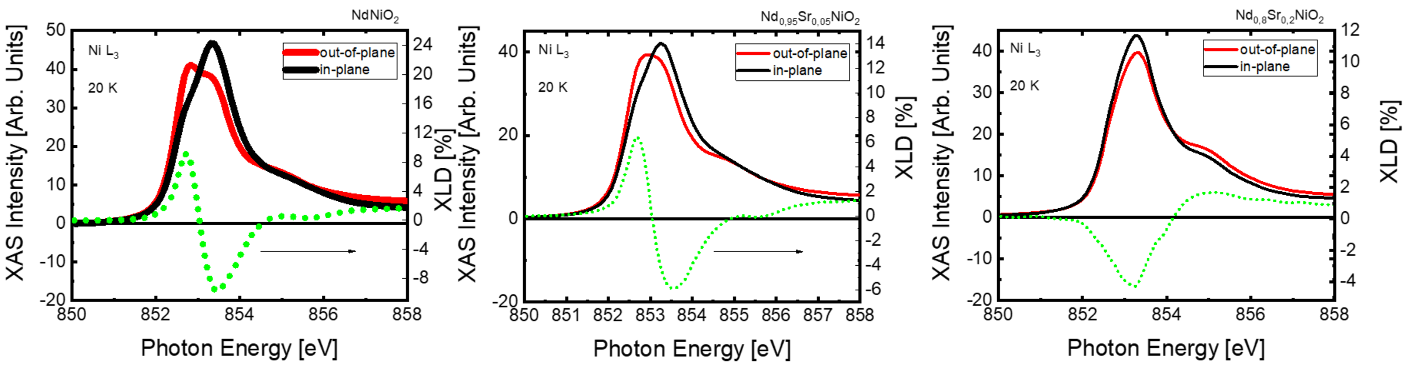

Figure 2 XAS at the Ni L3-edge for three Sr doping content showing a strong in-plane/out-of-plane anisotropy, most likely reflecting the Ni-Nd hybridization spatial anisotropy (see Figure 3).

Finally, our RIXS energy maps show in the surroundings of this incident energy (852.6 eV) and at a 0.6 eV energy loss (Y-axis) a peculiar feature (so far, indeed, explained as a result of the Ni-Nd hybridization), that disappears as the Sr content is increased and, more importantly, also when the light polarization is parallel to the sample surface (in-plane direction). Figure 3 shows the energy maps in a small energy loss range for all three samples acquired with both light polarizations.

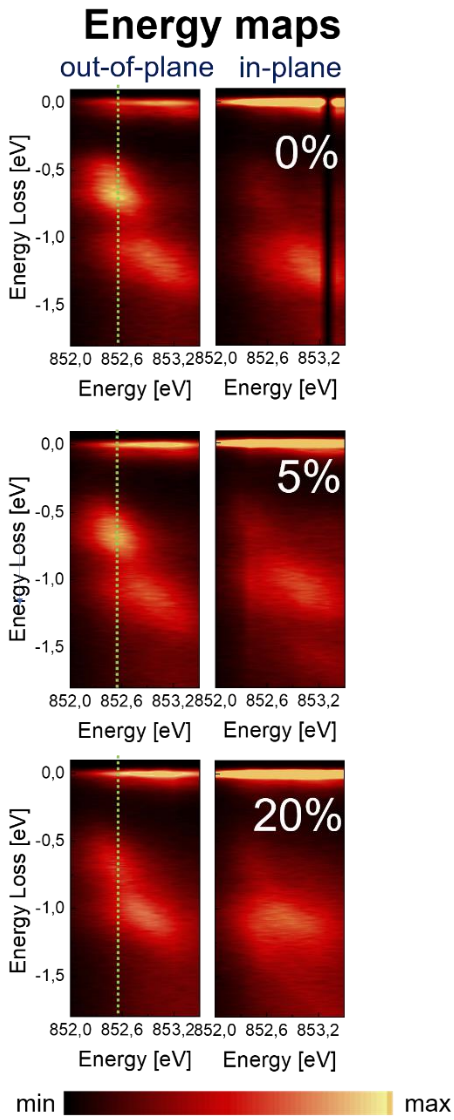

Figure 3 RIXS energy maps acquired in the th 10° tth 90° geometry with both LH (out-of-plane) and LV (in-plane) light polarization for three different Sr-doping level.

So basically, with our set of data we can speak in favour of an anisotropic Nd-Ni hybridization that mostly take places in the out-of-plane direction, and is characterized by a decreasing strength upon hole-doping the system. This led to believe that the Nd-Ni hybridization may play a role in the stabilization of the superconducting state. As already evidenced in several DFT+U studies the Nd 5d bands cross E_F, creating electron pockets at Γ and A points of the Brillouin zone. As a result, the Ni $3dx^2$-$y^2/3dz^2$ bands are self-doped with holes (electrons transfer from Ni to Nd orbitals), but despite this transferring undoped samples are not superconducting perhaps mimicking the underdoped regime of cuprates. By externally adding holes to the system (Sr doping), the Fermi level is pushed down, and the Nd-Ni hybridized levels are now emptied. As a result, the (chemically) hole-doped Ni 3d planar orbital states gain further spectral weight at the Fermi level and, drive the system I nthe superconducting state.

Finally, with our results, we can offer a model in which the superconducting state in infinite-layer nickelates is the result of a competition between an anisotropic Ni-Nd hybridization and Sr-doping which effect is to decrease the spectral weight at Fermi levels of such Nd-Ni hybridized levels (a draft is being prepared). Regarding the pairing mechanism our preliminary results (energy/momentum maps), tell that magnons do not enter in the game, since we do not observe them for the 20% Sr-doped sample. Further analysis and measurements are nevertheless necessary to confirm this point.