Giant orbital polarization of Ni\textsuperscript{2+} in square planar environment

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Understanding the electronic behavior of Ni\textsuperscript{2+} in a square planar environment of oxygen is the key to unravel the origin of the recently discovered superconductivity in the hole doped nickelate \(\text{Nd}_0.8\text{Sr}_0.2\text{NiO}_2\). To identify the major similarities/dissimilarities between nickelate and cuprate superconductivity, the study of the electronic structure of Ni\textsuperscript{2+} and Cu\textsuperscript{2+} in an identical square planar environment is essential. In order to address these questions, we investigate the electronic structure of \(\text{Sr}_2\text{CuO}_3\) and Ni doped \(\text{Sr}_2\text{CuO}_3\) single crystals containing \((\text{Cu/Ni})\text{O}_3\) square planar units. Our polarization dependent X-ray absorption spectroscopy experiments for Ni\textsuperscript{2+} reveal very large orbital polarization, which is a characteristic feature of high \(T_c\) cuprate. This arises due to the low spin \(S=0\) configuration with two holes in Ni \(3d_{x^2−y^2}\) orbitals - in contrast to the expected high spin \(S=1\) state from Hund’s first rule. The presence of such \(S=0\) Ni\textsuperscript{2+} in hole doped nickelate would be analogous to the Zhang Rice singlet. However, the Mott Hubbard insulating nature of the Ni\text{O}_2 unit would point towards a different electronic phase space of nickelates, compared to high \(T_c\) cuprates.

INTRODUCTION

Ever since the discovery, understanding the mechanism of high temperature superconductivity in cuprates remains an open problem in condensed matter physics [1]. Over the years, the importance of several factors, such as the antiferromagnetic, insulating phase of the parent compound, large orbital polarization, strong hybridization between Cu \(d\) and O \(p\) states, and Zhang Rice (ZR) physics have been recognized to play a role in the hole doped superconducting phase [2–4]. The recent finding of superconductivity in Sr doped \(\text{NdNiO}_2\) having Ni in a square planar oxygen environment is a monumental development [15], and strongly implies that the ground state of \(\text{CuO}_2\) and dominant \(d^{10}\text{L}_0\) configurations (\(\text{L}\) denotes a hole in O \(2p\) state) - contrary to the pure \(d^9\) state expected from an ionic picture [43].

In this work, we have investigated the electronic and magnetic structure of \(\text{Sr}_2\text{CuO}_3\) (SCO) and \(\text{Sr}_2\text{Cu}_{0.9}\text{Ni}_{0.1}\text{O}_3\) (SCNO) single crystals by magnetic measurement, synchrotron based X-ray linear dichroism (XLD) measurements, density functional theory (DFT) and cluster calculations. We observed extremely large orbital polarization of Ni in \(\text{Sr}_2\text{Cu}_{0.9}\text{Ni}_{0.1}\text{O}_3\) single crystal, which arises due to the low spin \(S=0\) configuration with completely unoccupied \(3d_{x^2−y^2}\) orbitals of Ni\textsuperscript{2+}. Ni doping of SCO also enhances the contribution of the \(d^9\) configuration in the ground state of Cu\textsuperscript{2+}.
Our comprehensive study has also affirmed Mott Hubbard insulating nature of the NiO4 unit and larger charge transfer energy compared to high Tc cuprate.

RESULTS

Single crystals of SCO and SCNO, grown by the traveling solvent floating zone technique [44] have been investigated in this study. We first discuss the results of polarization dependent XAS (X-ray absorption spectroscopy) experiment on Cu L3, L2 edges, which is known to be a very powerful technique to probe orbital symmetry of cuprates [45–48]. In Fig. 1(c), we compare linear polarization dependent XAS of Cu L3 and L2 edges for SCO and SCNO. XAS (I_c) of SCO recorded with in-plane (H) polarized light consists of sharp peaks around 930.5 eV and 950.5 eV, which are associated with the transition from the spin-orbit split Cu 2p3/2 and Cu 2p1/2 core levels, respectively, and the final state configuration is 2p3d10 (2p denotes a core hole in Cu 2p states). The much lower intensity of these peaks in XAS (I_c) with the out-of-plane (V) polarization signifies that the upper Hubbard band (UHB) is predominantly contributed by Cu d_{x^2-y^2} orbitals, which is expected for a d⁰ electronic configuration in D_{4h} symmetry. The features around 934 eV and 954.5 eV, which are much stronger for the out-of-plane polarization, are related to the transitions into Cu 3d_{3z^2-r^2} derived states, which become partially unoccupied due to the hybridization with Cu 4s states [47]. Upon Ni doping, the intensity of the white line increases significantly for both polarizations though the line shape remains unchanged (Fig. 1(c)). The origin of the increase of the d⁰ configuration upon Ni doping will be discussed later in this paper.

Next, we have performed similar polarization dependent XAS experiments at Ni L3, L2 edges of SCNO at 300 K. As shown in Fig. 2(a), the two intense peaks around 853 eV & 870 eV in the spectra (I_{ab}) recorded with the in-plane polarization, correspond to the transitions from Ni 2p3/2 and Ni 2p1/2 core levels respectively. The spectrum (I_c) recorded with the out-plane polarization shows significantly low intensity indicating that the holes predominantly occupy the d_{x^2-y^2} bands. The peaks around 856 eV and 874 eV, show higher intensity in I_c than I_{ab}. Similar to the case of Cu, these features are related to the hybridization of Ni 3d_{3z^2-r^2} and Ni 4s states. Since the experimental realization of large orbital polarization (OP) in Ni based oxides, with important implications for superconductivity, has been a topic of paramount interest [11, 14, 51, 52], we have further evaluated OP of Ni in the present case. According to the sum rule of linear polarization, the ratio of holes in d_{3z^2-r^2} and d_{x^2-y^2} [11] is defined as

$$X = \frac{I_{3z^2-r^2}}{I_{x^2-y^2}} = \frac{\int h_{3z^2-r^2}(E)dE}{\int h_{x^2-y^2}(E)dE}$$

with $I_{3z^2-r^2}/I_{x^2-y^2} = 3I_{h}^{4d} - I_{c}$. By this definition, $X=1$ corresponds to equal hole population in $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals and $X=0$ signifies 100% $d_{x^2-y^2}$ character of holes. We have obtained $X \approx 0.23$ (also see Supplementary Materials) in the present case. This implies 81% of the holes occupy Ni $d_{x^2-y^2}$ orbital, which is the highest among the existing literature of Ni based complex oxides (see Table I).

| System | $X$ | % of holes in Ni $d_{x^2-y^2}$ |
|--------|-----|-----------------------------|
| SrCuO2/LaNiO3 | 0.7 | 59% |
| LaTiO3/LaNiO3/LaAlO3 | 0.55 | 65% |
| La1Ni3O8 | 0.4-0.5 | 67-71% |

The orbital polarization $(P)$ is defined in the literature in
terms of the electronic occupation as [11]

\[ P = \frac{n_{x^2-y^2} - n_{3z^2-r^2}}{n_{x^2-y^2} + n_{3z^2-r^2}} = \left| \frac{4}{n'} - 1 \right| \frac{(X-1)}{(X+1)} \]  

(2)

where \( n_{x^2-y^2} = 2 - h_{x^2-y^2} \) and \( n_{3z^2-r^2} = 2 - h_{3z^2-r^2} \) are the number of electrons in \( d_{x^2-y^2} \) and \( d_{3z^2-r^2} \) orbitals, respectively. It is important to note that the value of \( n' = n_{x^2-y^2} + n_{3z^2-r^2} \) cannot be determined unambiguously for many compounds due to the presence of charge transfer from O 2p to Ni 3d orbitals [11, 53]. In the present case, \( n' = 2 \) as the ground state wavefunction is almost entirely contributed by the \( d^0 \) configuration (discussed in the later part of the paper). We have obtained \( P \approx 63\% \) for the SCNO, which is also larger compared to all existing literatures on Ni based complex oxides [8, 11, 14, 51, 52, 54].

In order to understand the origin of this unprecedentedly large orbital polarization, we have calculated XAS [\( I(ab + L) / 2 \)] and XLD [\( I(ab - L) \)] of Ni \( L_{3/2} \) edges considering a NiO\(_4\) cluster with \( D_{4h} \) symmetry and Ni\(^{2+}\) ionic configuration using CTO4XAS [55] and Quanty [56] programs that incorporate ligand field multiplet theory. First, we have simulated the XAS and XLD spectra (Fig. 2(b)) for the high spin \( S = 1 \) configuration [57, 58] with \( U = 7 \) eV and \( \Delta = 9 \) eV (all other parameters, used for the simulations have been listed in Ref. 59). Clearly, neither the simulated XAS nor the XLD matches with the corresponding experimental spectra. In particular, the experimentally observed \( L_2\)-edge XAS shows only one peak around 870 eV whereas the simulated spectra shows two peaks. The shape of the simulated XLD is completely different and the amount of anisotropy is significantly lower compared to the experimental data. We have also simulated XAS and XLD with the same values of \( U \) and \( \Delta \) but with low spin configuration \( (S = 0) \) by adjusting crystal field parameters [59], which ensures the following expected crystal field splitting \( d_{3z^2-r^2} < d_{xy} < d_{y^2} < d_{x^2-y^2} \) for a square planar environment [49]. The excellent matching (Fig. 2(c)) between the experimentally observed and simulated spectra with low spin configuration establishes Ni\(^{2+}\) is in \( S = 0 \) state in the square planar environment. The ground state of Ni\(^{2+}\) is strongly dominated by the \( |d^8\rangle \) configuration (\( \approx 0.97 \) \( |d^9\rangle + 0.03 \) \( |d^6\rangle \)). Simulated spectra with \( U > \Delta \) also do not match with experimental observation (see Supplementary Materials), implying electronically NiO belongs to the Mott Hubbard insulating region of ZSA phase diagram [40] - strikingly similar to the recent experimental reports of the hole doped NdNiO\(_2\) [16, 32]. Due to the large \( \Delta \), Ni \( d \) is less hybridized with O \( p \), compared to the Cu \( d-O-p \) hybridization. Thus, the replacement of a fraction of Cu by Ni pushes holes from O \( p \) towards Cu \( d \) around the dopant Ni. This enhances the relative contribution of \( d^0 \) configuration (also confirmed by magnetic measurement, discussed later in the paper), resulting in the observed strong increase in the white line intensity in Cu XAS upon Ni doping (Fig. 1(c)).

The increase of the \( d^0 \) component of Cu due to the presence of nonmagnetic Ni\(^{2+}\) is also evident in bulk magnetic measurements. The low-temperature magnetic susceptibility (\( \chi \)) plots of pristine, 1%, and 10% Ni doped SCO samples are shown in Fig. 3. As expected, \( \chi \) for the pristine sample is very small due to the antiferromagnetic coupling between the neighboring Cu spins. Upon doping with Ni at the Cu-site, the -O-Cu-O-Cu-O- chain breaks into finite length segments due to an effective spin 0 on the Ni ion. The number of seg-
configuration with fitted that to CW equation for obtaining the effective moment from low-temperature limit \([62]\). Hence, we used the data below 10 K and all the terms mentioned above, reduces to the CW equation in the temperature range, one should also include not only the chain susceptibility, i.e., the susceptibility of an endless antiferromagnetic chain \([60]\), but also the boundary contribution arising from the staggered moment at the free ends of a chain segment \([39, 61]\). However as shown by Sirker et al., the generalized expression of \(\chi\) including all the terms mentioned above, reduces to the CW equation in the low-temperature limit \([62]\). Hence, we used the data below 10 K and fitted that to CW equation for obtaining the effective moment from which the spin state of Ni can be inferred. The data for 1\% Ni doped SCO sample has been adapted from Ref. 39.

![FIG. 3. Temperature dependence of magnetic susceptibility (\(\chi\)) and fitting by Curie-Weiss (CW) law. In principle, depending on the temperature range, one should also include not only the chain susceptibility, i.e., the susceptibility of an endless antiferromagnetic chain \([60]\), but also the boundary contribution arising from the staggered moment at the free ends of a chain segment \([39, 61]\). However as shown by Sirker et al., the generalized expression of \(\chi\) including all the terms mentioned above, reduces to the CW equation in the low-temperature limit \([62]\). Hence, we used the data below 10 K and fitted that to CW equation for obtaining the effective moment from which the spin state of Ni can be inferred. The data for 1\% Ni doped SCO sample has been adapted from Ref. 39.](image)

ments thus produced being \(p + 1 \sim p\) for large \(p\), where \(p\) is the concentration of Ni in the sample. Assuming a statistically random distribution of Ni ions over the chain length, it is reasonable to consider that one-half of the chain segments will have an odd number of Cu spins, and even for the remaining. Since Cu-spins are antiferromagnetically paired, the net moment on the even-length segments tends to 0 at sufficiently low-temperatures \([63]\). In contrast, the odd-length segments remain with an uncompensated Cu spin, which results in an enhancement in the magnetic response of Ni doped samples, as seen in our measurement (Fig. 3). A very weak Curie-constant in the pristine sample can be attributed to the presence of intrinsic defects. We fitted the low-temperature susceptibility of our samples using the Curie-Weiss (CW) law: \(\chi = \chi_0 + C/(T - \theta_{CW})\) where \(\chi_0\) is the temperature independent contribution arising due to Van Vleck paramagnetism and core diamagnetism, \(C\) is the Curie-constant, and \(\theta_{CW}\) is the Weiss temperature. The best fit parameters for the three samples are tabulated in Table II.

It is reassuring to note that the value of \(C\) increases by a factor of 10 on increasing the Ni concentration from \(p = 0.01\) to 0.1. \(\chi_0\) for SCNO is positive and about an order of magnitude higher than for SCO. Such an enhancement is likely due to the Van Vleck paramagnetism associated with unoccupied orbitals of Ni\(^{2+}\) ion in the low-spin state \([64]\). From \(C\), the \(\mu_{eff} (=\sqrt{8C})\) for the two Ni doped samples \((p = 0.01\) and 0.1\) turned out to be \(\sim 0.12 \mu_B\) and \(\sim 0.38 \mu_B\) per formula unit respectively. The average effective moment on the odd-length segments \((=\sqrt{8C}/(0.5p))\) turns out to be 1.74 \(\mu_B\) and 1.70 \(\mu_B\), respectively for the two samples. These values are close to an effective moment of \(\sim 1.73 \mu_B\) expected for a \(S = 1/2\) \((d^9\) system\), confirming our conclusion of Cu XAS measurement.

To obtain a microscopic understanding of our experimental observations, we have carried out first-principles density functional theory (DFT) calculations for Sr\(_2\)CuO\(_3\) and Sr\(_2\)Cu\(_{0.95}\)Ni\(_{0.05}\)O\(_3\) [see Methods for details of the calculations]. The calculated orbital projected density of states (DOS) are shown in Fig. 4. In contrast with the transition metal oxides with positive \(\Delta\), the bonding states of SCO are primarily contributed by Cu \(d\) orbitals due to negative \(\Delta\) \([50]\). The Cu \(d_{xy}, d_{xz}, d_{yz}\) and \(d_{3z^2−r^2}\) bonding states are spread around 6 eV below the Fermi level (Fig. 4(b)). The valence band and conduction band edge of undoped SCO comes predominantly from O \(p\) states (inset of upper panel of Fig. 4(a)), as expected for the covalent insulating nature of SCO \([42]\). The \(d_{x^2−y^2}\) antibonding states are partially unoccupied and show a pronounced contribution to the upper Hubbard band, in line with our XAS results. We find that the magnetic moment on Cu is close to 0.19 \(\mu_B\). The slight overestimation of this moment, compared to the experimentally observed moment \((\sim 0.06\mu_B)\) \([34]\) is likely to be related to the presence of the quantum fluctuation effects in SCO, which are not accounted for in a mean field approach like DFT.. The much smaller moment, compared to the expected moment of 1.73 \(\mu_B\) for a pure \(d^9\) \((S=1/2)\) configuration, again signifies that the ground state of Cu\(^{2+}\) in SCO is predominantly \(d^{10}L_2\) configuration with small admixture with \(d^9\). In Ni doped SCO, there is a small increase in Cu magnetic moments, which range from 0.19 to 0.25 \(\mu_B\) and a net moment of \(\sim 0.13 \mu_B\) per formula unit. This also supports our observation about the increase of white line intensity of Cu XAS (Fig. 1(c)) and magnetic measurement i.e. the enhancement of \(d^9\) configuration of Cu upon Ni doping. For Ni doped SCO, the contribution arising from different Ni \(d\) orbitals is shown in Fig. 4(c) (also see lower panel of Fig. 4(a)). We find that the antibonding states of Ni \(d_{x^2−y^2}\) orbital are completely unoccupied, while all other Ni \(3d\) states are completely filled. This low spin \(S=0\) configuration of Ni with holes in the \(d_{x^2−y^2}\) orbital is in excellent agreement with

| Doping \((p)\) | \(\chi_0\) (emu \(\text{mol}^{-1}\text{Oe}^{-1}\)) | \(C\) (emu \(\text{mol}^{-1}\text{Oe}^{-1}\)) | \(\theta_{CW}\) (K) | \(\mu_{eff}\) \((\mu_B)\) | \(\mu_{eff\,B}\) \((\mu_B)/1/2\) Ni) |
|---|---|---|---|---|---|
| 0 | \(\sim 5 \times 10^{-5}\) | 0.0005(4) | -0.6 | \(\sim 0.06\) | - |
| 0.01 | \(\sim 3 \times 10^{-5}\) | 0.0018(2) | -1(1) | \(\sim 0.12\) | \(\sim 1.74\) |
| 0.1 | \(\sim 5 \times 10^{-4}\) | 0.018(2) | -2(1) | \(\sim 0.38\) | \(\sim 1.70\) |
our experimental observations.

DISCUSSION

To summarize, our element sensitive X-ray absorption spectroscopy experiments with linear polarized light on $\text{Sr}_2\text{Cu}_{0.9}\text{Ni}_{0.1}\text{O}_3$ have found that 81% holes of Ni$^{2+}$ sites occupy the $d_{x^2−y^2}$ orbital, which is the highest among all Ni containing oxides reported so far. Ni doping in $\text{Sr}_2\text{CuO}_3$ also results in hole transfer from O$^p$ to Cu$d$ orbitals. Cluster calculations have not only confirmed the Mott Hubbard insulating nature of NiO$_4$ units but also revealed that the observed giant orbital polarization arises due to low spin ($S=0$) configuration of Ni$^{2+}$. This nonmagnetic state of Ni$^{2+}$ has been further corroborated by ab initio theory.

Finally, we also note that our investigated system has several parallels with the recently discovered superconducting nickelates. Our system comprises of NiO$_4$ units forming one dimensional chains, while the superconducting nickelates have a two dimensional NiO$_2$ plane. It is expected that the increase in the dimensionality of the system would further enhance $\Delta$ [65], and therefore our conclusions about the Mott-Hubbard nature and the low spin configuration of Ni$^{2+}$ should also hold for nickelate superconductors. The larger charge transfer energy might be responsible for the observed lower $T_c$ in hole doped nickelates [20, 21, 27].

MATERIALS AND METHODS

Single crystals of SCO and SCNO were grown under O$_2$ atmosphere using the traveling solvent floating zone technique as reported elsewhere [44]. All the experiments reported here are carried out on as grown crystals. The magnetic susceptibility was measured using a Physical Property Measurement System, Quantum Design, USA. X-ray absorption spectra (XAS) with vertically (V) and horizontally (H) polarized X-ray were recorded at room temperature in 4-ID-C beamline of Advanced Photon Source, Argonne National Laboratory, USA.

All theoretical calculations have been performed within the framework of density functional theory as implemented in the Quantum Espresso package [66]. The wave functions are expanded in a plane wave basis set with kinetic and charge density cut offs set to 45 Ry and 450 Ry respectively. The exchange-correlation interactions are taken into account by the Perdew-Burke-Ernzeroff form of generalized gradient approximation [67]. The ion-electron interactions are described by ultrasoft pseudopotentials [68]. The on-site Coulomb interaction is modeled using LDA+$U$ approach in the linear-response method of Cococcioni and de Gironcoli [69], with $U = 5$ eV and 9 eV on Ni and Cu, respectively.

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