What can we learn from the nickelate vs. cuprate phase diagrams?

Character of the "normal state" of the nickelate superconductors
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Recommended with a Commentary by Atsushi Fujimori, National Tsunghua University

The recent discovery of superconductivity in the infinite-layer nickelate $Nd_{1-x}Sr_xNiO_2$ [1] has attracted tremendous interest because the superconductivity occurs in the hole-doped square lattice of the Ni^+ ions, which have the same d^9 configuration as the Cu^{2+} ions in the cuprates. Therefore, information from the nickelates, namely, similarities to and differences from the cuprates, will help us to elucidate the mechanisms of the superconductivity and the unusual normal-state properties of the cuprates from new perspectives. For such purposes, experimental data from nickelates with as high quality as those from the cuprates are indispensable. In a recent breakthrough in the $Nd_{1-x}Sr_xNiO_2$ thin-film synthesis using $(LaAlO_3)_{0.3}(Sr_2TaAlO_6)_{0.7}$ (LSAT) substrates, the growth and reduction conditions were optimized and extended defects were eliminated, as reported in the recommended paper. This allowed the authors of the paper to investigate the intrinsic transport properties of $Nd_{1-x}Sr_xNiO_2$.

The recommended paper reports similarities between the nickelates and cuprates, including the *strange metal* behaviors, $\rho(T) \propto T$, near a quantum critical point (QCP) buried behind the superconducting dome (T_c dome). Here, the temperature at which the resistivity upturn occurs decreases with x and goes to zero at the QCP. This similarity between the nickelates and cuprates is remarkable considering the differences between the two systems such as the Mott-Hubbard- *versus* charge-transfer-type electronic structures [2] and the single-band *versus* multi-band structures [3, 4]. In particular, in spite of the fact that the parent compounds of the nickelates are metals due to the presence of electron pockets, the phase diagram is qualitatively similar to that of the cuprates, as shown in Fig. 1.

Furthermore, the differences between the two systems are even more intriguing. Compared to the cuprates, the nickelate show the following features:

(i) The T_c dome is shifted toward higher hole concentrations by $x \sim 0.05$.

(ii) The QCP is shifted toward a lower hole concentration from $x \sim 0.19$-0.20 to 0.16.

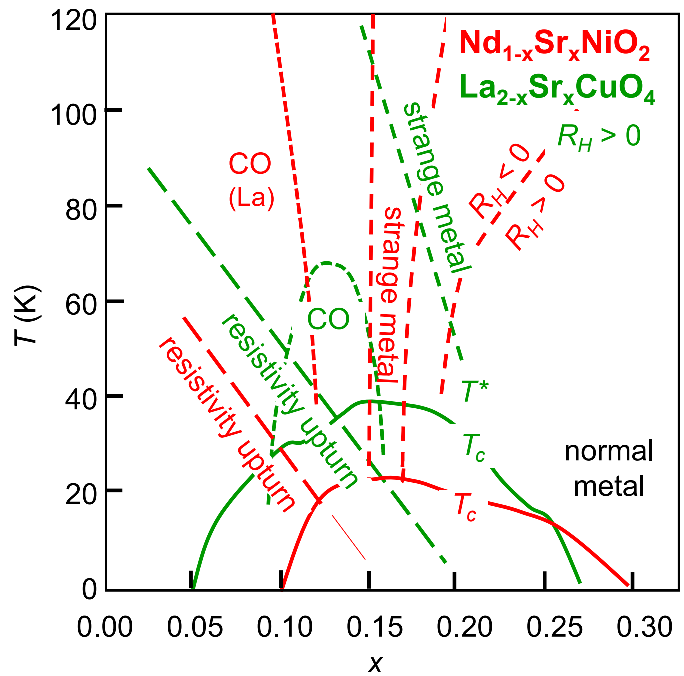

Figure 1: Phase diagram of $Nd_{1-x}Sr_xNiO_2$ (red) compared with that of the cuprate $La_{2-x}Sr_xCuO_4$ (LSCO, green) reported in the recommended paper. The charge-ordered (CO) phase of $La_{1-x}Sr_xNiO_2$ has been adopted from Ref. [5]. As for LSCO, the strange metal phase which appears above the pseudogap temperature T^* is taken from Refs. [6–8], and the CO phase from Ref. [9]. The resistivity upturn below T_c for LSCO has been measured under high magnetic fields, where the superconductivity is suppressed [10]

As for (i), if T_c is determined by the density of holes in the $d_{x^2-y^2}$ band, the presence of the electron pockets would increase the hole density and would shift the T_c dome toward lower x. It appears that, on the contrary, the presence of the electron pockets depletes the holes in the $d_{x^2-y^2}$ band, possibly through the formation of electron-hole pairs or excitons. The sign change of the Hall coefficient $R_{\rm H}$ in the overdoped region (Fig. 1) would reflect the shrinkage of the electron pockets and the recovery of the hole density with increasing x.

As for (ii), one should know first why the resistivity upturn occurs in the two systems. In the recommended paper, it is pointed out that the upturn cannot be explained by an extrinsic effect such as disorder but by an intrinsic electron correlation effect such as charge order (CO). Figure 1 shows that the CO occurs at lower x in the nickelates [5] than in the cuprates [9], which is correlated with the different QCP positions between the two systems. This in turn suggests that CO plays a role in the formation of the QCP. The CO scenario is consistent with the observation that the QCP occurs universally at $x = 0.19-0.20$ in various cuprates in spite of the varying Fermi-surface shapes and topologies [11]. Other kinds of symmetry breaking such as time-reversal symmetry breaking (TRSB) [12] and rotational symmetry breaking (RSB) [13] have also been proposed as the origin of the QCP at $x = 0.19 - 0.20$ in the cuprates. The latter scenarios have been tested by various experimental methods including Kerr rotation for TRSB [14] and elasto-resistivity for RSB [15]. Similar experiments are highly desired for the nickelates.

Now the recommended paper has given much deeper insight into the physics of the nickelates than before, the next step would be to apply low energy spectroscopic tools such as ARPES and STM/STS. As for the cuprates, at least for LSCO, transport and spectroscopy show anomalies at the same "pseudogap" temperature T^* [7,8] (Fig. 1). Studies on nickelates using ARPES and/or STM/STS are challenging but will make the second breakthrough.

From the chemistry point of view, the difficulties in the synthesis of the superconducting nickelates compared to the cuprates would be due to the unusual valence state of Ni^+. In order to deduce electronic structure parameters such as the on-site Coulomb energy U and the p-to-d charge-transfer energy Δ, and hence the Mott-Hubbard *versus* charge-transfer character [2], core-level photoemission spectroscopy combined with model calculations is more accurate than x-ray absorption spectroscopy and inelastic x-ray scattering [16]. The new high-quality thin films will allow one to perform bulk-sensitive hard x-ray core-level photoemission and will give us a more quantitative information about chemical bonding in the nickelates.

References

[1] D. Li, K. Lee, B. Y. Wang, M. Osada, S. Crossley, H. R. Lee, Y. Cui, Y. Hikita, and H. Y. Hwang, Nature **572**, 624 (2019).

[2] M. Jiang, M. Berciu, and G. A. Sawatzky, Phys. Rev. Lett. **124**, 207004 (2020).

[3] H. Sakakibara, H. Usui, K. Suzuki, T. Kotani, H. Aoki, and K. Kuroki, Phys. Rev. Lett. **125**, 077003 (2020).

[4] C.-J. Kang and G. Kotliar, Phys. Rev. Lett. **126**, 127401 (2021).

[5] M. Rossi, M. Osada, J. Choi, S. Agrestini, D. Jost, Y. Lee, H. Lu, B. Y. Wang, K. Lee, A. Nag, Y.-D. Chuang, C.-T. Kuo, S.-J. Lee, B. Moritz, T. P. Devereaux, Z.-X. Shen, J.-S. Lee, K.-J. Zhou, H. Y. Hwang, W.-S. Lee, arXiv:2112.02484.

[6] H. Takagi, T. Ido, S. Ishibashi, M. Uota, S. Uchida, and Y. Tokura, Phys. Rev. B **40**, 2254 (1989).

[7] Y. Ando, S. Komiya, K. Segawa, S. Ono, and Y. Kurita, Phys. Rev. Lett. **93**, 267001 (2004).

[8] M. Hashimoto, T. Yoshida, K. Tanaka, A. Fujimori, M. Okusawa, S. Wakimoto, K. Yamada, T. Kakeshita, H. Eisaki, and S. Uchida, Phys. Rev. B **75**, 140503(R) (2007).

[9] R. Comin and A. Damascelli, Annu. Rev. Condens. Matter Phys. **7**, (2016) 369.

[10] G. S. Boebinger, Y. Ando, A. Passner, T. Kimura, M. Okuya, J. Shimoyama, K. Kishio, K. Tamasaku, N. Ichikawa, and S. Uchida, Phys. Rev. Lett. **77**, 5417 (1996).

[11] B. Keimer, S. A. Kivelson, M. R. Norman, S. Uchida, and J. Zaanen, Nature **518**, 179 (2015).

[12] C. M. Varma, Phys. Rev. Lett. **83**, 3538 (1999).

[13] Y. Huh and S. Sachdev, Phys. Rev. B **78**, 064512 (2008).

[14] R.-H. He, M. Hashimoto, H. Karapetyan, J. D. Koralek, J. P. Hinton, J. P. Testaud, V. Nathan, Y. Yoshida, H. Yao, K. Tanaka, W. Meevasana, R. G. Moore, D. H. Lu, S.-K. Mo, M. Ishikado, H. Eisaki, Z. Hussain, T. P. Devereaux, S. A. Kivelson, J. Orenstein, A. Kapitulnik, and Z.-X. Shen, Science **331**, 1579 (2011).

[15] K. Ishida, S. Hosoi, Y. Teramoto, T. Usui, Y. Mizukami, K. Itaka, Y. Matsuda, T. Watanabe, and T. Shibauchi, J. Phys. Soc. Jpn. **89**, 064707 (2020).

[16] M. Hepting, D. Li, C. J. Jia, H. Lu, E. Paris, Y. Tseng, X. Feng, M. Osada, E. Been, Y. Hikita, Y.-D. Chuang, Z. Hussain, K. J. Zhou, A. Nag, M. Garcia-Fernandez, M. Rossi, H. Y. Huang, D. J. Huang, Z. X. Shen, T. Schmitt, H. Y. Hwang, B. Moritz, J. Zaanen, T. P. Devereaux, and W. S. Lee, Nat. Mater. **19**, 381 (2020).