Collaborations on ARPES study of quantum materials

Here I summarize my activities during my visit to ICC-IMR as a visiting associate professor hosted by Prof. Masaki Fujita. In particular, I briefly describe two projects in ongoing collaboration with the Fujita group on ARPES study of quantum materials, i.e., 3D negative electronic compressibility in electron-doped cuprate superconductors, and doping dependence ARPES study of complex oxides based on vacuum annealing.

During my visit to ICC-IMR as a visiting associate professor，I had the opportunity to do the following things: to work closely with Prof. Masaki Fujita's group; to visit Prof. Y. Koike's group, Prof. T. Sato's lab and Prof. H. Kumigashira's group; to visit the synchrotron lightsource UVSOR at the Institute for Molecular Science in Myodaiji, Okazaki; to attend the 12th International Conference on Spectroscopies in Novel Superconductors (SNS2019) at University of Tokyo. In particular, through numerous face-to-face discussions with various members of the Fujita group, exchanges of ideas were very effective and a few projects for collaborations based mainly on ARPES measurements were identified. I will briefly summarize two of them in the following.

Project 1: 3D Negative electronic compressibility (NEC) in electron-doped cuprate superconductors.

My previous group at Boston College found with ARPES in an iridate material Sr3Ir2O7 first evidence for 3D NEC [1], which is a new emergent phenomenon driven by strong electron correlations. This finding points to a distinct pathway towards an uncharted territory of NEC featuring bulk correlated metals with unique potential for applications in low-power nanoelectronics and novel metamaterials beyond reach of their 2D cousins [2].

Preliminary evidence obtained by my current group suggests the occurrence of NEC in electron-doped cuprates Nd2-xSrxCuO4 (NCCO) at 0.04≤x≤0.10 and its crossover onto a positive electronic compressibility (PEC) regime at higher doping levels (Fig. 1a-b). Consistent with this preliminary experimental result, single-band Hubbard model calculation performed by Prof. A. Bansil's group at Northeastern Univ. obtains a NEC phase which crosses over into PEC beyond a boundary dependent of the tight-binding band structure parameters and doping level (Fig. 1c). According to the calculation, NEC in this system is driven by the antiferromagnetic correlations (tunable by an effective on-site Coulomb repulsion in the calculation) which are strongly weakened with increasing electron doping, reminiscent of the case of Sr3Ir2O7 [3]; the NEC-PEC crossover occurs as these correlations become too weak and the valence band crosses the Fermi level forming hole pockets that tend to drive the chemical potential up.

If confirmed, these results would constitute the first experimental case of NEC in cuprates that had eluded early observation. It would also constitute the first case of NEC-PEC crossover in a bulk metal, which occurs between two metallic phases. We plan to perform an integrative study to scrutinize these results, in terms of the deep valence band and core level shifts and low-energy band structure based on both bulk Ce and surface K doping. Combining both doping approaches allows to assess the role of disorders in connection with the recent controversy about the effect of annealing condition on the electronic properties of electron-doped cuprates.

Related to this project, the Fujita group has provided samples of NCCO as well as PrLa0.6CexCuO4-d for a comparative study.

Project 2: Doping dependence ARPES study of complex oxides based on vacuum annealing.

We are in the process of developing a new experimental technique in order to obtain entire doping evolution of materials electronic structure on a given cleaved surface without the need of changing samples as in most traditional doping dependence studies.

The idea of this capability is derived from Ref. [4], which showed that sample surface can survive after repeated vacuum annealings, and ARPES data quality remained good after one month's repeated treatments and measurements. The authors did their sample annealing in a MBE chamber with a poor vacuum, and the

treatment process involved multiple sample transfers; these two factors tend to reduce the sample surface lifetime. Besides, the quenching mechanism was largely absent, except when the sample is transferred from the MBE chamber after fresh annealing back onto the liquid-helium cryostat--a "slow" process that would take ~10 mins.

We intend to perform vacuum annealing & quenching of samples in the same chamber where ARPES measurements are carried out. The procedure is as follows: First, laser heating a given cleaved surface of single-crystal sample up to 1300 C for a few seconds/mins, then remove the laser and the sample will be quenched instantly as the sample has been sitting on the liquid helium cryostat during the entire treatment. Each of such a treatment cycle can alter the oxygen content of the sample surface which is then subject to subsequent ARPES measurement.

In light of the potentially substantial improvements in our design, our treatment process should offer a much cleaner, efficient and effective way to tune (decrease) the sample surface oxygen content in-situ. The dream experiment in pursuit with this new technique is to study quantum criticality in ARPES, which requires finetuning of doping level and measurement with high precision and reproducibility --challenges that have prevented the use of advanced experimental techniques other than transport measurements for the quantum criticality research, although those transport studies were already quite impactful.

Related to this project, the Fujita group has provided hole-doped cuprate superconductor Bi2Sr2CuO6+x samples [5] for the ongoing testing required for our new experiment technique.

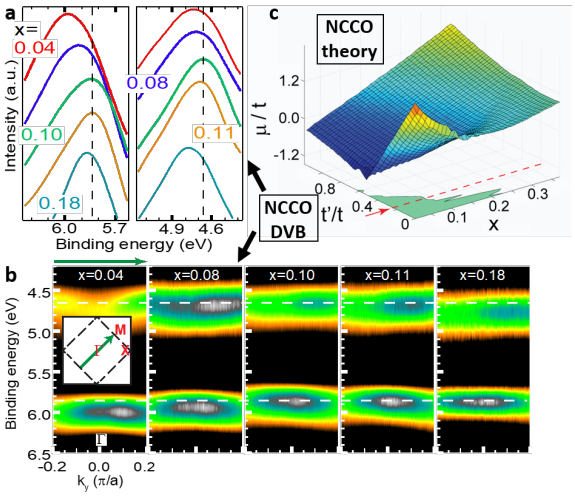

Fig.1 Doping dependence of the (**a**) spectra (second derivative) at Γ and (**b**) dispersion along a momentum cut through Γ of two deep valence bands on Nd2-xCexCuO4 at 70 K. (**c**) Hubbard model calculation of the chemical potential as functions of x and the ratio between next- and nearest-neighbor hopping parameters.

References

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