

Summary of thesis:

Variation of the electronic states of Ca_2RuO_4 and Sr_2RuO_4 under uniaxial pressures

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Competition and cooperation among spin, orbital, and lattice degrees of freedom are key concepts to understand intriguing phenomena in condensed matter systems. As one of such fascinating systems, the layered perovskite ruthenates $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$ have been attracting wide interest for their variety of electronic states originating from multiple degrees of freedom: for example, Ca_2RuO_4 is an antiferromagnetic (AFM) Mott insulator^[1], whereas Sr_2RuO_4 is a leading candidate for a spin-triplet superconductor^[2].

For studying a system like $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$ with orbital degree of freedom, uniaxial pressure (UAP) is expected to be effective because it will couple well with the orbital which spatially spreads along a certain direction. UAP can realize different crystal structures depending on pressure direction and selectively control the symmetry of the crystal differently from hydrostatic pressure.

Therefore, we have studied the electronic states of Ca_2RuO_4 and Sr_2RuO_4 under UAPs with three different pressure directions: two in-plane directions, $[100]_{\text{T}}$ and $[110]_{\text{T}}$, and the out-of-plane direction, $[001]_{\text{T}}$, using the tetragonal notation. $[100]_{\text{T}}$ and $[110]_{\text{T}}$ direction are parallel and diagonal to the in-plane Ru-O bond of the RuO_6 octahedra, respectively. Single crystals of Ca_2RuO_4 were provided by Prof. F. Nakamura in Kurume Inst. of Tech., and Sr_2RuO_4 were synthesized in our Lab lead by Prof. Y. Maeno.

For revealing the in-plane UAP effect on Ca_2RuO_4 , we performed magnetization and resistivity measurements^[3]. We succeeded in inducing the ferromagnetic metallic (FM-M) phase. The mechanism of this Mott transition is considered to be the same as that under hydrostatic pressure: the out-of-plane flattening distortion of RuO_6 octahedra is released, and the xy and $\{yz, zx\}$ bands of Ru $4d$ electrons approach energetically and overlap. The critical pressures of the FM-M phase for the UAPs (0.4 GPa for $P//[100]_{\text{T}}$ and 0.2 GPa for $P//[110]_{\text{T}}$) are substantially lower than that for the hydrostatic pressure (0.5 GPa^[4]). Interestingly, the critical pressure of the FM-M phase and the pressure dependence of FM component of magnetization were found to be highly anisotropic. These peculiar

anisotropic results can be naturally understood as a consequence of the orthorhombic crystal distortions in Ca_2RuO_4 and existence of orthorhombic crystalline twin domains in the sample.

Surprisingly, the out-of-plane UAP effect on Ca_2RuO_4 was opposite to a simple prediction^[5]; from resistivity measurements, we have clarified that the insulating gap is suppressed from 3000 K to 700 K. This result suggests that Ca_2RuO_4 approaches a metallic state under the out-of-plane UAP with a mechanism, which is different from that of the Mott transition under in-plane UAP. Since the crystalline field splitting between the xy and $\{yz, zx\}$ bands should become larger, this gap suppression is attributed to the enhancement of the band width as a result of the release of rotation or/and tilting distortion of RuO_6 octahedra.

In the study of the out-of-plane UAP effect on the superconductivity (SC) of Sr_2RuO_4 , we used Sr_2RuO_4 -Ru eutectic crystals in order to compare the UAP-originated 3-K SC with the interfacial 3-K SC in the same sample^[6]. We previously revealed that the onset of superconducting transition temperature T_c of Sr_2RuO_4 without Ru inclusions is enhanced from 1.5 K to 3.2 K, which is the same as the onset T_c of the SC realized near interfaces between Sr_2RuO_4 and Ru in the eutectic crystal^[7]. In this thesis, we newly proposed the out-of-plane UAP dependence of the spatial distribution of 3-K SC in Sr_2RuO_4 -Ru from AC susceptibility and resistivity measurements.

The in-plane UAP effect on Sr_2RuO_4 was also revealed to be significantly anisotropic from AC susceptibility measurements; we clarified that $P//[100]_T$ rather than $P//[110]_T$ is favorable for inducing 3-K SC. This result can be understood consistently with previous studies^[8].

Our results strongly demonstrate the effectiveness of UAP to control the electronic state of systems which have multiple degrees of freedom.

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