Observation of a novel orbital selective Mott transition in $\mathbf{Ca_{1.8}Sr_{0.2}RuO_4}$

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Mott transition, that is, metal-insulator transition (MIT) driven by electron correlation, has been the subject of intensive research for many decades and continues to provide theoretical and experimental challenges [1]. In the case of "bandwidth-controlled" Mott transition, the ratio U/W between the on-site electron-electron Coulomb repulsion energy U and the one-electron bandwidth W controls the MIT: Above a critical U value, $U_{\mathrm{cr}} \sim W$, all electrons are localized and the system becomes an insulator. Theoretically, in most cases this problem has been studied based on the single-band Hubbard model. Experimentally, for electronic states near the Fermi level, the d-orbital degeneracy is lifted in the cuprate superconductors by a crystal field and the p-orbital degeneracy in organic conductors by the dimerization of molecules. A question then naturally arises how the Mott transition takes place when the orbital degeneracy is not lifted. If all the degenerate orbitals are equivalent, the critical values becomes $U_{\mathrm{cr}} \sim \sqrt{N}W$, where N is the orbital degeneracy [2]. If some of the degenerate orbitals are not equivalent, the situation becomes far from trivial and two alternative scenarios may be considered: (i) MIT's for the different orbitals occur separately [3,4], or (ii) all the orbitals undergo an MIT simultaneously due to finite interaction between the orbitals [5].

This rather conceptual and theoretical problem has become a reality and has been debated for many years since a bandwidth-controlled MIT in the layered oxides $\mathrm{Ca}_{2-x}\mathrm{Sr}_x\mathrm{RuO}_4$ was discovered by Nakatsuji and Maeno [6]. The $x = 2$ end member $\mathrm{Sr_2RuO_4}$ is a famous triplet p-wave superconductor while the $x = 0$ end member $\mathrm{Ca_2RuO_4}$ is a Mott insulator with an ordered moment of $S = 1$ as expected for the localized electronic configuration of the Ru^{4+} ion: $d_{xy\uparrow}d_{xy\downarrow}d_{yz\uparrow}d_{zx\uparrow}$. The MIT occurs at $x \simeq 0.2$ between the antiferromagnetic insulator and an antiferromagnetic metal, and the antiferromagnetic order persists up to $x \simeq 0.5$. The most striking phenomenon is the Curie-Weiss magnetic susceptibility of $S = 1/2$ in the vicinity of the critical concentration $x \simeq 0.5$. The observed $S = 1/2$ local moment means that some of the Ru d electrons are localized and the others are itinerant, that is, an "orbital-selective Mott transition" (OSMT) should take place in this system.

Then the next question arises. Among the three d orbitals, d_{xy}, d_{yz}, and d_{zx}, in the crystal-field-split t_{2g} manifold forming the Fermi surfaces of $\mathrm{Sr_2RuO_4}$, whether the d_{xy} orbital (forming a 2D Fermi surface) or the d_{yz}-d_{zx} orbitals (forming two 1D Fermi surfaces) first undergo an MIT and become localized. One would naively expect that the d_{yz}-d_{zx} bands should first open a Mott gap because the 1D bands should have narrower bandwidths than the 2D d_{xy} band [3]. However, the rotation of the $\mathrm{RuO_6}$ octahedra caused by the substitution of small Ca ions reduces the d_{xy} bandwidth [7,8]. Also, the Ca substitution compresses the $\mathrm{RuO_6}$ octahedra along the z-axis and lowers the d_{xy} level relative to d_{yz}-d_{zx}, enhancing the occupation of d_{xy} [7,8]. In $\mathrm{Sr_2RuO_4}$, where the

RuO_6 octahedron is almost regular, the three d orbitals are rather uniformly occupied: $n_{xy} \simeq n_{yz} \simeq n_{zx} \simeq 4/3 = 1.33$. Optical measurements of the change of Drude weight across the MIT favors the Mott transition in the d_{xy} band rather than in the d_{yz}-d_{zx} bands [9]. If this is the case, the Ca substitution increases the occupation from $n_{xy} \simeq 1.33$ towards ~ 2. But $n_{xy} = 2$ is unlikely because the fully occupied d_{xy} orbital would not allow a Mott transition to occur. Then is $n_{xy} \sim 1.5$ most likely? Can such a non-integer filling allow a Mott transition?

Neupane and co-workers' ARPES studies on $Ca_{2-x}Sr_xRuO_4$ provided deeper insight into the question of how the OSMT took place in such complicated circumstances. The 1D Fermi surfaces of the d_{yz}-d_{zx} bands remained while the 2D Fermi surface of the d_{xy} band disappeared in going from $x \sim 0.5$ to $x \sim 0.2$, i.e., towards the metal-insulator boundary. This is consistent with the optical study mentioned above [9] and supports the view that the OSMT takes place first in the 2D d_{xy} band and then the 1D d_{yz}-d_{zx} bands. The area of the missing d_{xy}-derived Fermi surface was 1.5 electrons per Ru atom. In order to explain the opening of a Mott gap in the d_{xy} band with non-integer filling, they proposed that the $\sqrt{2} \times \sqrt{2}$ superstructure resulting from the rotation of the RuO_6 octahedra was responsible.

Thus the situation in $Ca_{2-x}Sr_xRuO_4$ turned out to be far more complicated than initially debated. Nevertheless, the concept of OSMT should be applied to a wide class of strongly correlated systems. For some f-electron compounds, the coexistence of localized and itinerant electrons have often been proposed [10], which necessarily results from OSMT. The "heavy Fermion" behavior of the spinel-type LiV_2O_4 has been attributed to the coexistence of localized and itinerant d orbitals [11], which in turn necessitates OSMT, too. Considering the delicate balance of different interactions which makes possible the OSMT in $Ca_{2-x}Sr_xRuO_4$, careful examination of various interactions has to be made to establish an OSMT in any system.

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