Mott *versus* Slater: Nature of antiferromagnetism in the $J_{\rm eff}$ = 1/2 insulator Sr_2IrO_4 and its implications

1. Theoretical study of insulating mechanism in multi-orbital Hubbard models with a large spin-orbit coupling: Is Sr_2IrO_4 a Slater insulator or a Mott insulator? H. Watanabe, T. Shirakawa, and S. Yunoki, arXiv:1402.0935.
2. A. Yamasaki, H. Fujiwara, A. Higashiya, A. Irizawa, O. Kirilmaz, F. Pfaff, P. Scheiderer, J. Gabel, M. Sing, T. Muro, M. Yabashi, K. Tamasaku, H. Sato, H. Namatame, M. Taniguchi, A. Hloskovskyy, H. Yoshida, H. Okabe, M. Isobe, J. Akimitsu, W. Drube, R. Claessen, T. Ishikawa, S. Imada, A. Sekiyama, and S. Suga, Bulk nature of layered perovskite iridates beyond the Mott scenario: An approach from bulk sensitive photoemission study, arXiv:1310.7160.

Recommended with a Commentary by Atsushi Fujimori, University of Tokyo

Mott transition, which occurs as a result of competing electron kinetic energy and electron-electron repulsive Coulomb energy, is observed in many 3*d* transition-metal oxides because the electron band width *W* and the on-site Coulomb energy *U* are of comparable magnitudes. In 4*d* transition-metal oxides, because of the larger spatial extent of the 4*d* orbitals and hence reduced *U/W*, metallic behaviors are much more common. Therefore, the insulating behaviors of some 5*d* oxides in spite of even more extended 5*d* orbitals, in particular of the two-dimensional perovskite Sr_2IrO_4 [with Ir^{4+} ($5d^5$) ions] had been a mystery [1] until B. J. Kim and his co-workers pointed out that the large spin-orbit splitting of the 5*d* levels played an essential role [2]: The Ir 5*d* t_{2g} band is split into $J_{\rm eff}$ = 1/2 and 3/2 bands, and then the relatively narrow half-filled $J_{\rm eff}$ = 1/2 band is further split into the upper and lower Hubbard bands, resulting in the novel $J_{\rm eff}$ = 1/2 Mott insulator.

While the $J_{\rm eff}$ = 1/2 Mott insulator picture well explains the transport properties [1] and is supported by spectroscopic studies [2,3], it was challenged in the following two aspects:

(1) The $J_{\rm eff}$ = 1/2 and $J_{\rm eff}$ = 3/2 states may not be clearly separated and may be hybridized with each other (through low-symmetry crystal field and exchange interaction) [4],

(2) The insulating phase below the Néel temperature $T_{\rm N}$ = 240 K may not be a Mott insulator but a Slater insulator, in which long-range magnetic order opens an insulating gap [5].

While issue (1) does not affect the low-energy physical properties of Sr_2IrO_4 so seriously, issue (2) does, particularly at finite temperatures. If it is a Mott insulator, the gap is due to Coulomb repulsion and the system remains insulating above $T_{\rm N}$. If it is a Slater insulator, on the contrary, the system should be metallic above $T_{\rm N}$. Therefore, experimental and theoretical studies on metallic *versus* insulating behaviors above $T_{\rm N}$ are expected to give a clue. However, local-density-approximation plus dynamical-mean-field-theory (LDA+DMFT) calculations have given conflicting results so far:

metallic [5,6] or insulating above T_N [7]. In order to resolve the issue, one has to go beyond the standard (i.e., single-site) DMFT and to take into account short-range antiferromagnetic (AFM) correlations which may persist above T_N.

In the recommended article, Watanabe, Shirakawa, and Yunoki go beyond single-site theory by applying the variational Monte-Carlo (VMC) method to large two-dimensional clusters. Their results suggest that, for a certain range of parameter U/t ($\sim 3 < U/t < \sim 7$, where t is a hopping integral between neighboring Ir 5d t_{2g} orbitals), the AFM insulating ground state turns into a paramagnetic (PM) metal above T_N, that is, the ground state is a Slater insulator. For a larger $U/t > \sim 7$, the AFM insulating ground state becomes a PM insulator above T_N, that is, the system is a Mott insulator. The AFM ground state itself also changes its character across $U/t \sim 7$: Below $U/t \sim 7$, the PM-to-AFM transition is driven by exchange energy gain while above $U/t \sim 7$ it is driven by kinetic energy gain. They proposed that Sr_2IrO_4 is located near the boundary $U/t \sim 7$. The conflicting DMFT results mentioned above [5-7] can now be attributed to the subtle difference in the choice of parameter U/t.

In the other recommended article, Yamasaki *et al*. have measured the temperature dependence of the gap for Sr_2IrO_4 by photoemission spectroscopy. They found that the AFM gap decreases with temperature and closes above T_N, consistent with the Slater scenario. The same conclusion has been drawn by a recent temperature-dependent scanning-electron-spectroscopy (STS) study [8].

However, in spite of the apparently consistent theoretical and experimental results within the Slater-transition scenario, electrical resistivity does not show an appreciable anomaly at T_N and remains insulating above T_N [1], in favor of the Mott scenario. If we closely look at the photoemission and STS spectra, the gap closure above T_N is indeed not so evident as the authors claim: Spectral weight at the Fermi level (E_F) above T_N is indeed larger that below T_N but still remains very low and a large pseudogap of order ~0.5 eV persists. This is radically different from spectra predicted by the DMFT calculations, in which above T_N a sharp, intense quasi-particle peak grows at E_F. Such a discrepancy between theory and experiment has indeed been observed for many transition-metal oxides showing metal-insulator transitions and has often been attributed to surface effects. But Yamasaki *et al*.'s data were taken under truly bulk-sensitive conductions (using hard x-rays or very low energy photons). Most likely, the AFM correlations and hence the global antiferromagnetic gap structure persist above T_N as the Slater insulator undergoes a transition into a PM metal. If the strong AFM correlation persists above T_N, electrical resistivity may be largely determined by the large scale gap at least at temperatures as high as T_N (~ 240 K) and may not exhibit a pronounced anomaly at T_N irrespective of whether the insulating gap of tiny spectral weight is open or closed.

The physics of short-range AFM correlation discussed above cannot be incorporated in the single-site DMFT but is captured by the VMC study of large clusters. Unfortunately, it is difficult to calculate spectral functions based on the VMC result, but in the future technical development might

enable us to calculate spectral functions and to make direct comparison between theory and photoemission data. If such comparison becomes possible, it will bring about a major advance in our understanding of the physics of correlated systems. In relation to issue (1) above, the entangled spin and orbital components in the $J_{\mathrm{eff}} = 1/2$ state in carrier-doped Sr_2IrO_4 have been predicted to lead to a superconducting state containing both singlet and triplet as well as inter-orbital pairing [9] and, therefore, search for superconductivity in Ir oxides is highly anticipated.

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