Tuning the Spin-Orbit Coupled Ground State of Iridates with Pressure

DANIEL HASKEI, Argonne National Laboratory

The electronic ground state of the novel magnetic insulators BaIrO$_3$ [1] and Sr$_2$IrO$_4$ [2] is probed at ambient and high-pressure conditions using x-ray absorption and magnetic circular dichroism measurements. A spin-only description of the magnetic ground state is ruled out, spin-orbit entanglement in 5d states resulting in comparable orbital ($L_z$) and spin ($S_z$) contributions to the localized magnetic moments despite the presence of strong crystal fields and band effects in Ir 5d states. Pressures of $\sim 5$ GPa and 20 GPa quench the “weak” ferromagnetic ordering in BaIrO$_3$ and Sr$_2$IrO$_4$, respectively, despite robust local moments and insulating behavior remaining at these pressures, confirming the Mott character of the insulating gap. The expectation value of the angular part of the S-O interaction, $\langle L \bullet S \rangle$, extrapolates to zero at 80–90 GPa in Sr$_2$IrO$_4$ where an increased bandwidth strongly mixes $J_{\text{eff}} = 1/2, 3/2$ states and S-O interactions no longer dominate the electronic ground state. The likely appearance of a single, metallic band at a pressure of $\sim 1$ Mbar (100 GPa) provides an exciting backdrop for searches of superconductivity at high pressures [3]. Work at Argonne is supported by the U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC-02-06CH11357.