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

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The electronic ground state of the novel magnetic insulators BaIrO₃ [1] and Sr₂IrO₄ [2] is probed at ambient and highpressure 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 5*d*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 5*d* states. Pressures of ~ 5 GPa and 20 GPa quench the "weak" ferromagnetic ordering in BaIrO₃ and Sr₂IrO₄, 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, <**L**•S>, extrapolates to zero at 80–90 GPa in Sr₂IrO₄ where an increased bandwidth strongly mixes $J_{eff} = 1/2$, 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 ~ 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.

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[2] D. Haskel et al., Phys. Rev. Lett. 109, 027204 (2012).

[3] F. Wang and T. Senthil, Phys. Rev. Lett. 106, 136402 (2011).