**Electronic and structural response to pressure in the hyperkagome-lattice Na$_3$Ir$_3$O$_8$**

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The hyper-kagome lattice iridate Na$_3$Ir$_3$O$_8$, closely related to spin liquid candidate Na$_4$Ir$_3$O$_8$, is unusual in that spin-orbit interactions acting on the 1/3-filled Jeff = 1/2 state lead to a semimetallic ground state, in contrast to the conventional insulating Mott state stabilized by S-O interactions in the 1/2-filled Jeff = 1/2 state of other iridates including Na$_4$Ir$_3$O$_8$. We have studied the evolution of crystal structure, electronic structure, and transport in Na$_3$Ir$_3$O$_8$ under high pressure using x-ray diffraction, x-ray absorption near edge structure, and electrical resistance measurements in a diamond anvil cell. The study was augmented by the use of *ab initio* calculations, which provided insight into pressure-induced changes in crystal and electronic structure. We found that Na$_3$Ir$_3$O$_8$ transforms from a semimetal to an insulator under pressure, with an estimated energy gap that increases to about 130 meV at P ~ 9 GPa. At approximately 10 GPa, a cubic-to-monoclinic structural transition takes place between two insulating phases. This structural transition features a sizable volume collapse and a high-pressure phase characterized by apparent dimerization of Ir-Ir distances, wide dispersion of Ir-O-Ir bond angles, and an increase in the occupation of 5d states. Although the energy gap is reduced in the high-pressure phase, insulating behavior remains to the highest pressures ~1 Mbar. The strongly pressure-dependent insulating gap shows a positive correlation with the expectation value of the angular part of the S-O interaction, $\langle \mathbf{L} \cdot \mathbf{S} \rangle$, which may indicate an active role of S-O interactions in stabilizing the insulating state that emerges in the compressed Na$_3$Ir$_3$O$_8$ structure.

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