Under high pressure, simple molecular solids transform into non-molecular (extended) solids as compression energies approach the energies of strong covalent bonds in constituent chemical species. As a result, it is common to observe the transformation of molecular solids into more compact extended structures with more itinerant electrons, which softens repulsive interatomic interactions at high density. This allows exotic properties to be tuned, such as mechanical strength, nonlinear second harmonic optical properties, electric and optical conductivities, and magnetic properties of condensed-matter systems. Carbon dioxide, for example, exhibits a richness of high-pressure polymorphs with a great diversity in intermolecular interaction, chemical bonding, and crystal structures. Thus, group IV sulfides, in comparison with their chemical analog CO$_2$, provide opportunities to exploit the relationship between the structural phase transition, electronic metallization, and superconductivity. We present integrated spectral, structural, resistance, and theoretical evidence for several systems of simple molecular group IV sulfides that undergo pressure-induced electronic phase transitions to novel metallic, magnetically ordered, and/or superconducting states.

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