Jarosite, a mineral with a kagomé lattice, displays magnetic frustration yet orders magnetically below 65 K. As magnetic frustration can engender exotic physical properties, understanding the complex magnetism of jarosite comprises a multi-decade interdisciplinary challenge. To address this challenge, we use applied pressure to smoothly vary jarosite’s structure without manipulating the chemical composition, enabling a chemically invariant structure–function magnetocorrelation study. Using single-crystal and powder X-ray diffraction, we identify two pressure-induced phase transitions. By harnessing a suite of magnetic techniques under pressure, including SQUID-based magnetometry, time-resolved synchrotron Mössbauer spectroscopy, and X-ray magnetic circular dichroism, we construct the magnetic phase diagram for jarosite up to 120 GPa. Notably, we demonstrate that the magnetic ordering temperature increases dramatically to 240 K at 40 GPa, and then vanishes above a critical pressure of 45 GPa. Additionally, we conduct X-ray emission spectroscopy, Mössbauer spectroscopy, UV-visible absorption spectroscopy, Raman spectroscopy, and Fourier-transform infrared spectroscopy experiments, along with density functional theory calculations, to comprehensively map the magnetic and electronic structures of jarosite at high pressure. We use these maps to construct chemically-pure magnetostructural correlations which fully explain the nature and role of the magnetism in jarosite at extreme conditions.

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