

Ryan Klein

2019 COMPRES Annual Meeting

Poster Abstract:

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.

Contributing Authors:

Ryan A. Klein<sup>1</sup>, James P. S. Walsh<sup>1</sup>, Samantha M. Clarke<sup>2</sup>, Yinsheng Guo<sup>1</sup>, Danilo Puggioni<sup>3</sup>, Wenli Bi<sup>4,5</sup>, Gilberto Fabbris<sup>4</sup>, Yue Meng<sup>4,6</sup>, Daniel Haskel<sup>4</sup>, E. Ercan Alp<sup>4</sup>, Zhenxian Liu<sup>7</sup>, James M. Rondinelli<sup>3</sup>, Richard P. Van Duyne<sup>1</sup>, Steven D. Jacobsen<sup>8</sup>, Danna E. Freedman<sup>1\*</sup>

Affiliations:

<sup>1</sup> Department of Chemistry, Northwestern University, Evanston, Illinois 60208, United States

<sup>2</sup> Lawrence Livermore National Laboratory, Physical and Life Sciences Directorate, P.O. Box 808, L-350, Livermore, California 94550, United States

<sup>3</sup> Department of Materials Science and Engineering, Northwestern University, Evanston, Illinois 60208, United States

<sup>4</sup> Advanced Photon Source, Argonne National Laboratory, 9700 South Cass Avenue, Lemont, Illinois 60439, United States

<sup>5</sup> Department of Geology, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States

<sup>6</sup> HPCAT, Geophysical Laboratory, Carnegie Institute of Washington, Argonne, Illinois 60439, United States

<sup>7</sup> Department of Civil and Environmental Engineering, Institute of Materials Science, The George Washington University, Washington DC 20052, USA

<sup>8</sup> Department of Earth and Planetary Sciences, Northwestern University, Evanston, Illinois 60208, United States

Presenter: Ryan A. Klein