Correlated disorder-to-order crossover in the local structure of $K_xFe_{2-y}Se_{2-z}S_z$ superconductor

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A detailed account of the local atomic structure and disorder at 5 K across the phase diagram of the high-temperature superconductor $K_xFe_{2-y}Se_{2-z}S_z$ ($0 \leq z \leq 2$) is obtained from neutron total scattering and associated atomic pair distribution function (PDF) approaches [1]. Various model-independent and model-dependent aspects of the analysis reveal a high level of structural complexity on the nanometer length scale. Evidence is found for considerable disorder in the c-axis stacking of the $FeSe_{1-x}S_x$ slabs without observable signs of turbostratic character of the disorder. In contrast to the related FeCh (Ch = S, Se)-type superconductors, substantial Fe-vacancies are present in $K_xFe_{2-y}Se_{2-z}S_z$, deemed detrimental for superconductivity when ordered. Our study suggests that the distribution of vacancies significantly modifies the iron-chalcogen bond-length distribution, in agreement with observed evolution of the PDF signal. A crossover-like transition is observed at a composition of $z \approx 1$, from a correlated disorder state at the selenium end to a more vacancy-ordered (VO) state closer to the sulfur end of the phase diagram. The S-content-dependent measures of the local structure are found to exhibit distinct behavior on either side of this crossover, correlating well with the evolution of the superconducting state to that of a magnetic semiconductor toward the $z \approx 2$ end. The behavior reinforces the idea of the intimate relationship of correlated Fe-vacancy order in the local structure and the emergent electronic properties.

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