Quantification of Correlated Disorder in Alloy Systems Through Complex PDF Modelling

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Rietveld analysis provides a clear picture of the structure for materials that may be described using classical crystallography. Detailed information on atomic scale ordering and nanostructure in order-disorder phase transitions is not easily accessible, however, when using Rietveld analysis, a result of the so-called "nanostructure problem." The depth and inverse nature of this problem requires complex modelling to fit observed data, so that structure – processing – property relationships can be leveraged to tailor material performance.

Here we probe the correlated nature of chemical short-range order and displacement disorder in prototypical order-disorder Au-Cu systems. We achieve this through a novel approach, fitting multiple datasets. Detailed Rietveld fitting of high-resolution powder diffraction data gives quantitative information on microstructure and atomic structure. This also allows us point out the subtle effects in the XRD patterns that may be easily overlooked, as Rietveld analysis will instantly reveal mis-fitting models, highlighting the need for pair distribution function (PDF) measurements. Tuning an evolutionary algorithm for global optimization of large atomic ensembles to fit an observed PDF and quantify details of chemical short-range order. The PDF-derived are further refined using density functional theory (DFT) to elucidate the subtle bond length fluctuations associated with correlated disorder. Applications of this technique to both ambient and high-temperature conditions are discussed, and linked to the superstructure peaks and diffuse scattering found in the XRD patterns.