Fast simulation of disordered crystals using Gaussian copulas

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Substitutional disorder is ubiquitous in crystalline materials, yet generating physically realistic disordered structures remains computationally challenging. We present an algorithm that employs Gaussian copulas to rapidly generate disordered crystal structures consistent with observed diffuse scattering patterns. The method works by first generating correlated Gaussian random variables with a carefully chosen covariance matrix, then applying thresholding to transform these into discrete site occupancies. For binary disorder with equal probabilities, the relationship between Gaussian and binary correlations follows the analytical arcsin law, enabling straightforward implementation. The technique naturally extends to non-equal probability distributions and multi-state disorder through modified thresholding schemes. The algorithm requires only the generation of Gaussian random variables and a single FFT operation for periodic systems, offering significant computational advantages over conventional supercell Monte Carlo methods. This enables modeling of disordered crystals with efficiency similar to that of Reverse Monte Carlo (RMC); however, in contrast to RMC, the method also provides a direct analytical relationship between input parameters and the observed pair distribution function. We demonstrate the method on experimental diffuse scattering from the model system of Prussian Blue analogues. By combining tools from signal processing and statistical modeling with crystallographic applications, this work provides crystallographers with a practical, efficient tool for modeling disordered materials.