Sensitivity of thermal diffuse scattering to correlated disorder: A study of the disordered rocksalts $KBr_{1-x}Cl_x$

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The analysis of diffuse scattering in compounds with substitutional disorder mainly focuses on the static description of local chemical environments, while the changes to the dynamics of the system due to correlated disorder are widely ignored. However, it is clear from supercell lattice dynamical calculations [1], that correlated disorder affects the phonon dispersion, as different degrees of band splitting and/or broadening are observed depending on the degree and nature of local order.

We analyse the experimentally observed diffuse scattering in the solid solution series $KBr_{1-x}Cl_x$ ($0 \leq x \leq 1$), Figure 1a. The width of the the projected diffuse scattering line cuts (Figure 1b) serves as our measure for the sensitivity of the diffuse scattering to the change in composition (Figure 1c).

Using our model system, we establish the sensitivity of thermal diffuse scattering to the degree and nature of local order. We model different degrees of static correlated disorder and by utilising supercell lattice dynamical calculations we demonstrate the effect of correlated short range order on the first order thermal diffuse scattering. We show that the correlated disorder significantly affects the dynamics in a system and cannot be modelled by the commonly used virtual crystal approximation.

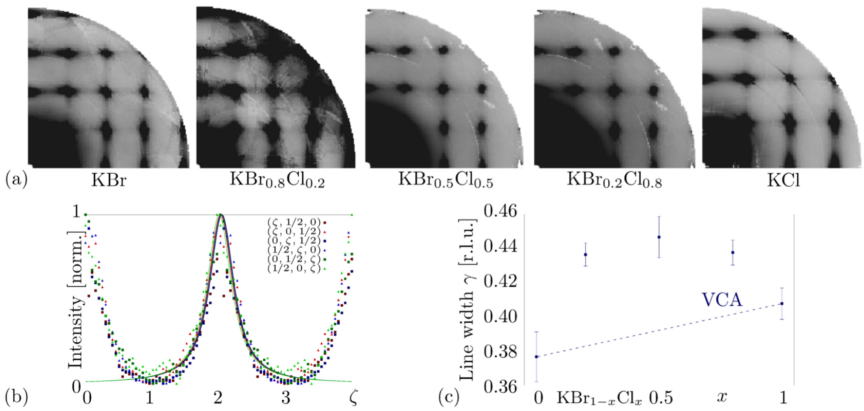

Figure 1: (a) Symmetry averaged *hk*0-sections of experimentally observed single crystal diffuse scattering for $KBr_{1-x}Cl_x$ with x = 0.0, 0.2, 0.5, 0.8, 1.0. (b) Cuts through the diffuse scattering of $KBr_{0.2}Cl_{0.8}$ along ⟨ζ 1/2 0⟩. The intensity is projected into one Brillouin zone ($0 \leq \zeta < 2$) and normalized to the intensity range 0 to 1. The intensity is repeated for $2 \leq \zeta \leq 4$ to show the peak shape that is fitted with a Lorentzian for the range $1 \leq \zeta \leq 3$. Fits are shown as solid lines. For more details see text. (c) Width γ of the observed diffuse scattering in relative lattice units (r. l. u.) as a function of composition. The dashed line indicates the expected trend from a virtual crystal approximation (VCA).

[1] Overy, A. R., Cairns, A. B., Cliffe, M. J., Simonov, A., Tucker, M. G. & Goodwin, A. L. (2016). *Nature comm.* 7(1), 1–8.