Disorder-Phonon Coupling in the Correlated Disorder System γ-($U_{1-x}Mo_x$)

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Understanding the role of disorder, and the correlations that exist within it, is one of the defining challenges in contemporary materials science. However, there are few material systems, devoid of other complex interactions, that can be used to systematically study the effects of correlated disorder arising from crystallographic conflict. The pseudo-*bcc* uranium molybdenum system is however an exemplar case study and as such we fabricated thin (~ 300 nm) epitaxial films of γ-($U_{1-x}Mo_x$) alloys in the range $0.17 < x < 0.31$ to be studied that the ID28 beamline (ESRF, France).

We established, via extensive diffuse x-ray scattering studies [1], that the intrinsic symmetry conflict; where uranium, which prefers a locally anisotropic environment, is forced into an isotropic *bcc* global symmetry, produces a new form of intrinsically tuneable correlated disorder where every atom is displaced to form a short-range superstructure with correlations existing over nanometre sized regions. This serves to modulate the local crystallographic periodicity, thus giving rise to the possibility of a form of disorder-phonon coupling. To investigate this possibility one alloy composition was measured with grazing incidence inelastic x-ray scattering (GI-IXS) and compared with extensive *ab-initio* modelling [2].

We discovered strong disorder-phonon coupling that relaxes degeneracy conditions at the **P** position, hardens the LA-2/3⟨111⟩$_p$ mode ubiquitous to monotonic *bcc* crystals and produces significant phonon linewidth broadening at almost all positions in the 1st Brillion zone [1]. This broadening is almost entirely due to the presence of correlations, and results in a strong reduction of the phononic contribution to thermal conductivity. Similar effects are expected in any system able to host comparable levels of crystallographic conflict and may be exploited when designing future functional materials. As the first IXS study, to our knowledge, on a thin film heterostructure composed of multiple single crystal layers this work also highlights the power of GI-IXS and may serve as a blueprint for future studies of thin film structures or devices with similar or greater complexity.

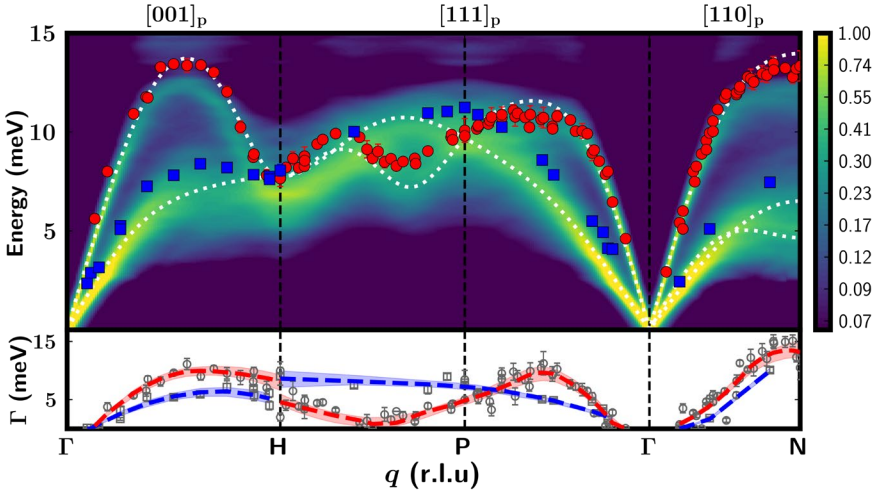

Figure 1: (Top Panel) Experimental (x = 0.23) and theoretical (x = 0.25) phonon dispersion curves. Transverse (longitudinal) acoustic modes are shown as blue squares (red circles) and theoretical results for two separate theoretical approaches are shown as dashed white lines and as a colour map, respectively. All directions are within the bcc BZ. (Bottom Panel) Raw linewidths are shown as grey squares (TA) and circles (LA) with deconvoluted linewidths shown by dashed blue (TA) and red (LA) trendlines with errors shown as confidence bands.

References

[1] D. Chaney et al., Phys. Rev. Materials **5**, 035004 (2021)

[2] A. Castellano et al., Phys. Rev. B **101**, 184111 (2020)