Understanding vibrational contributions to phase stability and transport in actinides

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While inelastic neutron scattering drove early studies on the lattice dynamics of actinides, issues with neutron absorbing isotopes, the absence of large crystals, and safety issues associated with handling large quantities of radioactive materials left a large gap in our understanding. Plutonium metal, for example, exhibits a rich phase diagram that includes six different allotropes spanning density variations of more than 30% and sits at the crossover from itinerate to localized 5f electrons in the periodic table – and yet very basic questions about its vibrational and electronic degrees of freedom remained unresolved for decades. For the actinide oxides, on the other hand, phonon thermal transport under irradiation is of critical importance to advanced nuclear reactor designs, but fully understanding phonon transport requires knowledge of the phonon dispersion curves including the line shapes. The measurement situation for lattice dynamics in the more challenging actinides improved dramatically with the advent of high energy resolution synchrotron based inelastic X-ray scattering (IXS) spectrometers, which have no isotope requirements and crystal sizes can be as small as a few microns – allowing measurements of crystal grains within a polycrystal sample. The first measured phonon dispersion curves for plutonium metal and the first phonon density of states of plutonium dioxide were both obtained using IXS rather than neutron scattering. IXS has also opened measurements of phonons at high pressures and in thin films using grazing incidence. The high atomic numbers of the actinides make them particularly well suited to IXS measurements in small geometries since the X-ray scattering per atom increases as the square of the atomic number. In this talk, I will highlight progress made in our understanding phase stability and thermal transport in actinides, with a particular emphasis on key insights obtained using IXS.