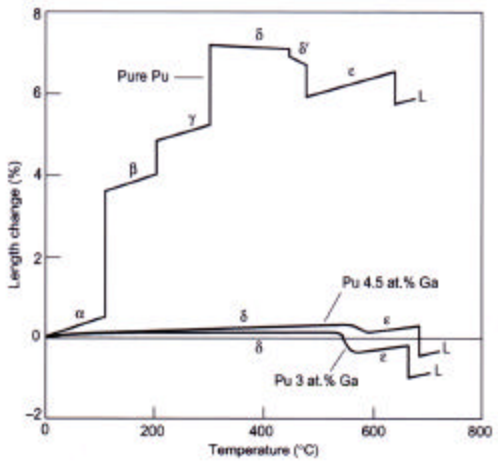
Report:

Phonon dispersion curves (PDCs) in plutonium (Pu) and its alloys have defied measurement for the past few decades since the discovery of this element in 1941. This is due to a combination of the high thermal-neutron absorption cross section of plutonium and the inability to grow the large single crystals (with dimensions of a few millimeters) necessary for inelastic neutron scattering. These limitations have recently been overcome by using a tightly focused undulator x-ray micro-beam scattered from single-grain domains in polycrystalline specimens. This experimental approach has been applied successfully to map the complete PDCs of an fcc δ-Pu-Ga alloy using the high resolution inelastic x-ray scattering (HRIXS) capability on ID28.

Pu is well known to have complex and unique physico-chemical properties [1]. Notably, the pure metal exhibits six solid-state phase transformations with large volume expansions and contractions along the way to the liquid state: $\alpha \rightarrow \beta \rightarrow \gamma \rightarrow \delta \rightarrow \delta' \rightarrow \varepsilon \rightarrow$ liquid. Unalloyed Pu melts at a relatively low temperature ~640ºC to yield a higher density liquid than that of the solid from which it melts, **(Figure 1)**. PDCs are key experimental data to the understanding of the basic properties of Pu materials such as: force constants, sound velocities, elastic constants, thermodynamics, phase stability, electron-phonon coupling, structural relaxation, etc.



Pu Crystal Structure		Density (g/cc)
α	Simple Monoclinic	19.86
β	Body-Centered Monoclinic	17.70
γ	Face-Centered Orthorhombic	17.14
δ	Face-Centred Cubic	15.92
δ'	Body-Centered Tetragonal	16.00
ε	Body-Centered cubic	16.51
L	Liquid	16.65

Figure 1 Transformations in Pu to different crystal structures are accompanied by very large volume changes. Alloying with Ga or Al avoids the transformation to γ, β and α and stabilizes the δ phase all the way to room temperature and below [1].

The complete PDCs for an fcc Pu-0.6 wt% Ga alloy are plotted in **Figure 2**, and represent the first full set of phonon dispersions ever determined for any Pu-bearing materials [2]. The solid (red) curves are calculated using a standard Born-von Kármán (B-vK) force constant model. An adequate fit to the experimental data is obtained if interactions up to the fourth-nearest neighbours are included. The dashed (blue) curves are recent dynamical mean field theory (DMFT) results by Dai et al. [3].

The elastic moduli calculated from the slopes of the experimental phonon dispersion curves near the Γ point are: C_{11} = 35.3 ±1.4 GPa, C_{12} = 25.5 ±1.5 GPa and C_{44} = 30.53 ±1.1 GPa. These values are in excellent agreement with those of the only other measurement on a similar alloy (1 wt % Ga) using ultrasonic techniques as well as with those recently calculated from a combined DMFT and linear response theory for pure δ-Pu [3]

Several unusual features, including a large elastic anisotropy, a small shear elastic modulus C', a Kohn-like anomaly in the T_1[011] branch, and a pronounced softening of the [111] transverse modes are found. These features can be related to the phase transitions of plutonium and to strong coupling between the lattice structure and the 5f valence instabilities. The HRIXS results also provide a critical test for theoretical treatments of highly correlated 5*f* electron systems as exemplified by recent dynamical mean field theory (DMFT) calculations for δ-plutonium.

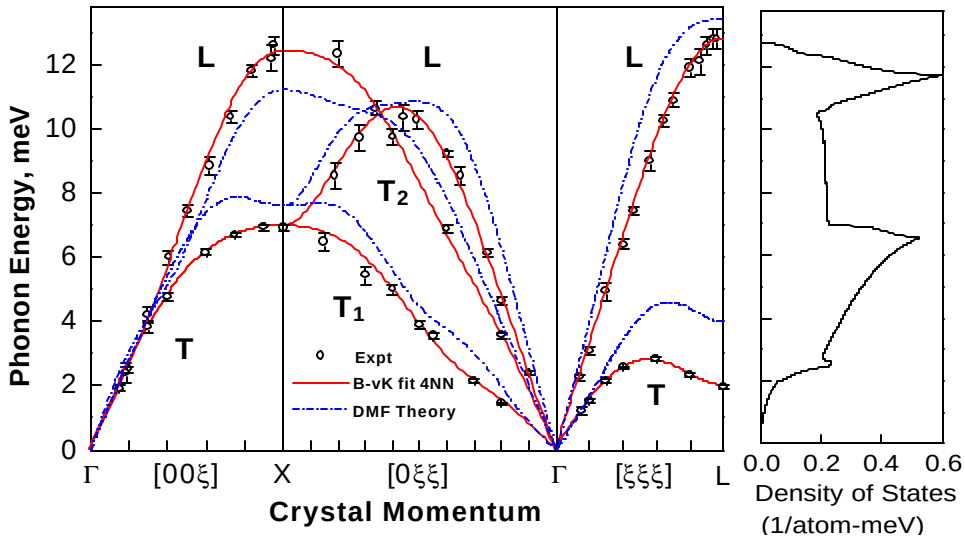


Figure. 2. Phonon dispersions along high symmetry directions in δ-Pu-0.6 wt% Ga alloy. The longitudinal and transverse modes are denoted L and T respectively. The experimental IXS data are shown as circles. Along the [0ξξ] direction, there are two transverse branches [011]<01-1> (T_1) and [011]<100> (T_2). Note the softening of the TA[ξξξ] branch towards the L point. The lattice parameter of our samples is a = 0.4621 nm. The red solid curves are the fourth-nearest neighbour Born-von Kármán model fit. The derived phonon density of states, normalized to 3 states per atom, is plotted in the right panel. The blue dashed curves are calculated dispersions for pure δ-Pu based on DMFT [3].

The experimental-theoretical agreements shown in Figure 2 in terms of a low shear elastic modulus C', a Kohn-like anomaly in the T1[011] branch, and a large softening of the T[111] modes give credence to the DMFT approach for the theoretical treatment of 5*f* electron systems of which δ-Pu is a classic example. However, quantitative differences remain. These are the position of the Kohn anomaly along the T_1[011] branch, the energy maximum of the T[111] mode s and the softening of the calculated T[100] branch near the X point, which is not observed experimentally. These differences are significant and thus provide a framework for refined theoretical treatments. Systematic HRIXS experiments as a function of temperature in the fcc Pu-Ga alloy are underway.

References

[1] S.S. Hecker, *Challenges in Plutonium Science*, Los Alamos Sci, **26**, 290 (2000).
[2] Joe Wong et al., *Science*, **301**, 1078 (2003)
[3] X. Dai et al., *Science* **300**, 953 (2003)