Report:

We have measured the complete phonon dispersion curves of an fcc δ-Pu-Ga alloy using the high resolution inelastic x-ray scattering capabilities at ID28. The result will be published in Science [1] in a forthcoming issue (Aug. 22) and represents the first full phonon dispersions ever determined for any Pu-bearing materials. .

A novel strain-enhanced re-crystallization technique [2] was used to fabricate a large-grain polycrystalline bulk sample of a Pu-Ga alloy containing ~0.6 wt % of Ga. The Ga stabilizes the otherwise high temperature (329-451 ^{o}C) pure fcc δ-phase Pu to room temperature [3]. The specimen, 2.8 mm in diameter, ~10 micron thick with a grain size ranges from 75 to 100 μm was mounted in a tested leak-proof double-containment cell. A focused beam, ~30 μm x 60 μm with a flux of ~2 x 10^{9} photons/s was first used to survey and map single-crystal domains in the polycrystalline microstructure of the specimen. The selected grain was then oriented appropriately to the desired scattering geometry for each of the longitudinal branches along the three principal directions in the fcc lattice, and phonon energies were recorded as a function of scattering angle (q-vector).

The resulting PDCs along the three principal symmetry directions in the fcc lattice are displayed in Fig. 1, together with a fit using a standard Born-von Kármán (B-vK) force constant model [4]. An adequate fit to the experimental data is obtained if interactions up to the fourth-nearest neighbours are included. The dashed curves are recent DMFT (dynamical mean field theory) results by Dai et al. [5].

The elastic moduli calculated from 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 [6] as well as with those recently calculated from a combined DMFT and linear response theory for pure δ-Pu [5]

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. Our results also provide a critical test for theoretical treatments of highly correlated 5f electron systems as exemplified by recent dynamical mean field theory (DMFT) calculations for δ-plutonium.

As evident in Fig. 1, the present IXS experiment validates the main qualitative predictions of a recent DMFT calculation for δ Pu [5] in terms of (a) a low shear elastic modulus C', (b) a Kohn-like anomaly in the T1[011] branch, and (c) a large softening of the T[111] modes. Such experimental-theoretical agreements give credence to the DMFT approach for the theoretical treatment of 5f electron systems of which Pu is a classic example. However, while there is good qualitative agreement between theory and experiment, quantitative differences indeed exist. These are: 1)

position of the Kohn anomaly along the T_1[011] branch; 2) the energy maximum of the T[111] modes and 3) the softening of the calculated T[100] branch near the X point, which is not observed experimentally. These differences are significant and thus provide the framework for refined theoretical treatments and further experiments in Pu and other 5f systems.

Our future IXS experiments are aimed at elucidating the nature of the T[111] softening as a function of Ga content, temperature and pressure and its association with a martensitic-like transformation of the fcc δ-phase to the monoclinic α' phase at low temperature (~150K).

References and Notes

1. Joe Wong et al., Science, **301** (2003), Aug. 22 issue
2. J.C. Lashley et al., *Scripta Mater.* **44**, 2815 (2001).
3. S.S Hecker, *Los Alamos Sci*, **26**, 290 (2000).
4. M. Born and K. Huang, *Dynamical Theory of Crystal Lattices*, (Clarendon Press) Oxford (1954)
5. X. Dai et al., *Science* **300**, 953 (2003)
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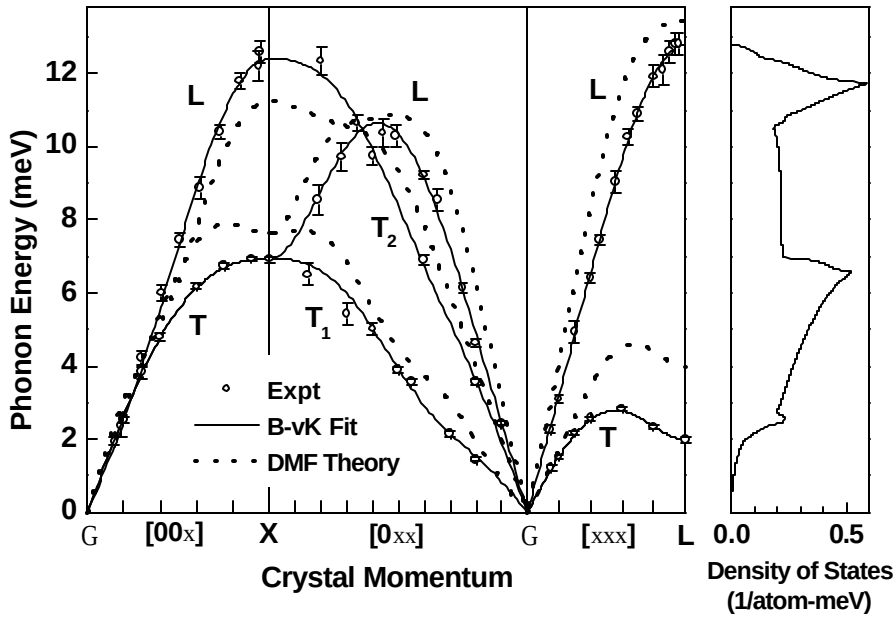

Fig. 1. Phonon dispersions along high symmetry directions in δ-Pu-0.6 wt% Ga alloy. The longitudinal and transverse modes are denoted L and T respectively. The experimental data are shown as circles. Along the [0ξξ] direction, there are two transverse branches [011]<01-1> (T_1) and [011]<100> (T_2). Note the softening of the TA[ξξξ] branch towards the L point. The lattice parameter of our samples is a=0.4621 nm. The 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 dashed curves are calculated dispersions for pure δ-Pu based on DMFT (5).