A neptunium analog of the LaFeAsO tetragonal layered compound has been synthesized and characterized by a variety of experimental techniques. The occurrence of long-range magnetic order below a critical temperature *T*N = 57 K is suggested by anomalies in the temperature-dependent magnetic susceptibility, electrical resistivity, Hall coefficient, and specific-heat curves. Below *T*N, powder neutron diffraction measurements reveal an antiferromagnetic structure of the Np sublattice, with an ordered magnetic moment of 1.70 ± 0.07μB aligned along the crystallographic *c* axis. No magnetic order has been observed on the Fe sublattice, setting an upper limit of about 0.3μB for the ordered magnetic moment on the iron. High-resolution x-ray diffraction data were measured as a function of temperature (5 < T <300 K) on the powder diffraction beam line ID31, using an incident beam energy of 35 keV. The powdered sample for this experiment was doubly encapsulated because it was put inside a Kapton tube (50 µm) and inserted in a Plexiglas hollow cylinder with a wall thickness of 200 µm. The results exclude the occurrence of lattice transformations down to 5K, in sharp contrast to the observation of a tetragonal-to-orthorhombic distortion in the rare-earth analogs, which has been associated with the stabilization of a spin-density wave on the iron sublattice. Instead, a significant expansion of the NpFeAsO lattice parameters is observed with decreasing temperature below T_N, corresponding to a relative volume change of about 0.2% and to an Invar behavior between 5 and 20 K. First-principles electronic structure calculations based on the local spin density plus Coulomb interaction and the local density plus Hubbard-I approximations provide results in good agreement with the experimental findings.

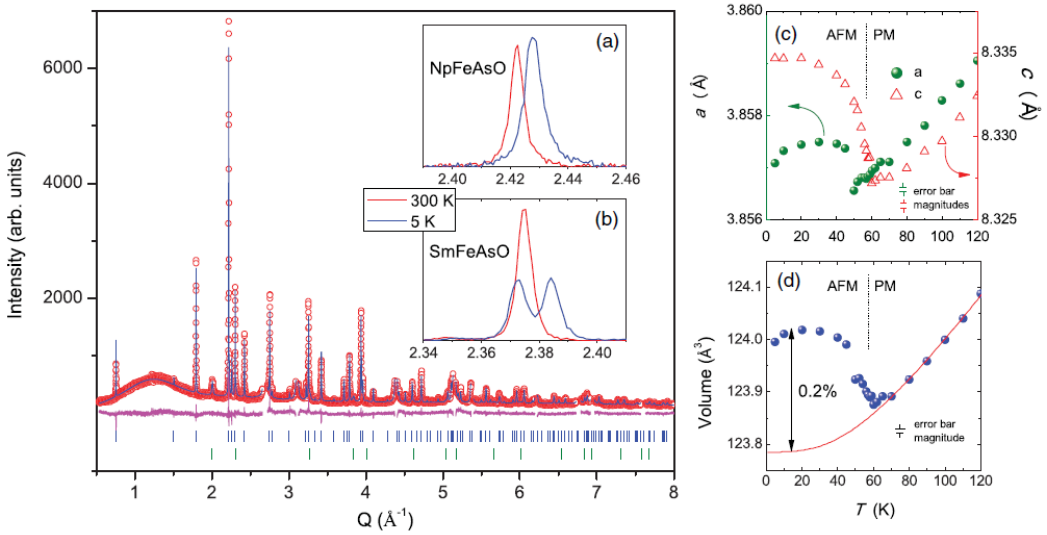

FIG. 1. Fit (solid blue line) of the refined structural model to room-temperature x-ray powder diffraction data (red circles) for NpFeAsO, confirming the tetragonal P4/nmm ZrCuSiAs-type structure. The data were collected on ID31 with a wavelength of λ = 0.35A° . Tick marks are shown for two refined phases: NpFeAsO (top, blue) and an 8% NpO_2 impurity (bottom, green). Insets (a) and (b) make a comparison of data for NpFeAsO and SmFeAsO where only SmFeAsO shows the splitting of the room-temperature (red line) tetragonal (P4/nmm) (111) Bragg reflection into the orthorhombic (Cmma) (201) and (021) reflections at 5 K (blue line). Insets (c) and (d) show the variation in the a and c lattice parameters and of the unit cell volume as a function of temperature, revealing a marked negative thermal expansion below T about 60 K in NpFeAsO. The red line through the volume data is a fit to the second order of the Gr¨uneisen approximation for the zero-pressure equation of state.