LETTER

REPLY TO JANOSCHEK ET AL.: The excited δ-phase of plutonium

Albert Migliori[a,1], Per Söderlind[b], Alexander Landa[b], Franz J. Freibert[a], Boris Maiorov[a], B. J. Ramshaw[a], and Jon B. Betts[a]

In a recent PNAS paper (1) we explain the anomalous temperature dependence of the elasticity in δ-plutonium (δ-Pu) in terms of a first-principles model that includes multiple energy configurations attributed to spin fluctuations.

Our model (1) captures the highly unusual behavior of decreasing elastic moduli with increasing temperature at constant volume. This is a falsifiable test that any model must pass but none, other than the one we present (1), has. This test is a difficult one, as usually the smaller volume state required to keep volume constant is elastically stiffer, contrary to measurements. Our theory yields a modest decrease by about 15%, whereas experiments show a 40% decrease in the 100–600 K range in δ-Pu. The thermal effect is thus larger in our experiment, but the anomalous behavior of negative temperature dependence is clearly captured by our parameter-free model. Certainly, one cannot expect our simple model to agree perfectly with the experiment because it ignores lattice vibrations.

We state (1) that the model is consistent with a weakly temperature-dependent magnetic susceptibility because the spin and orbital magnetic contributions counter each other. This result does not depend on a numerically exact cancellation between these components, but rather only that they are antiparallel and similar in magnitude, something that we now appear to agree upon (2).

The model by Niklasson et al. (3) predicts a spin moment of 4.76 μ_B in plutonium, and their speculation that Kondo physics may play a role was thus not supported by their own calculations but presumably motivated by the lack of experimental observation of magnetic moments in δ-Pu [this has of course changed (4)].

We also point out that our theory does not address a nonmagnetic "ground state" of δ-Pu but targets excitations and spin fluctuations through constrained calculations (5) that include restrictions on crystal structure, magnetic structure, and magnetic-moment magnitude.

We believe it is obvious that conclusions from the earlier experimental report (6) are inconsistent with the latest neutron-scattering experiments (4), as well as comments made in Janoschek et al. (2). Ref. 6 is entitled "Absence of magnetic moments in plutonium" and reports: "We maintain that there is no evidence whatsoever that magnetism either ordered or disordered exists in δ-Pu." This statement is not reconcilable with the two statements by Janoschek et al. in ref. 4: "The combination of our neutron spectroscopy and DMFT [dynamical mean-field theory] results unambiguously establishes that the magnetism in δ-Pu is not 'missing,' but dynamic," and in ref. 2: "the DLM [disordered local moment] approach of Migliori et al. (1) is not in accord with the experimentally determined magnetism in δ-Pu."

Acknowledgments

This work was supported as part of the Materials Science of Actinides, an Energy Frontier Research Center funded by the US Department of Energy, Office of Science, Basic Energy Sciences under Award DE-SC0001089; and under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

1 Migliori A, et al. (2016) Origin of the multiple configurations that drive the response of δ-plutonium's elastic moduli to temperature. *Proc Natl Acad Sci USA* 113(40):11158–11161.
2 Janoschek M, et al. (2017) Relevance of Kondo physics for the temperature dependence of the bulk modulus in plutonium. *Proc Natl Acad Sci USA* 114:E268.
3 Niklasson AMN, et al. (2003) Modeling the actinides with disordered local moments. *Phys Rev B* 67:235105.
4 Janoschek M, et al. (2015) The valence-fluctuating ground state of plutonium. *Sci Adv* 1(6):e1500188.
5 Dederichs PH, Blügel S, Zeller R, Akai H (1984) Ground states of constrained systems: Application to cerium impurities. *Phys Rev Lett* 53:2512–2515.
6 Lashley JC, Lawson A, McQueeney R, Lander GH (2005) Absence of magnetic moments in plutonium. *Phys Rev B* 72:054416.

[a]Los Alamos National Laboratory, Los Alamos, NM 87545; and [b]Lawrence Livermore National Laboratory, Livermore, CA 94550

Author contributions: A.M., P.S., A.L., F.J.F., B.M., B.J.R., and J.B.B. wrote the paper.

The authors declare no conflict of interest.

[1]To whom correspondence should be addressed. Email: migliori@lanl.gov.