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Photoelectric Effect in Uranium

C. P. Opeil[1], R. C. Albers[1], K. B. Blagoev[1], M. Gulácsi[2], J. C. Lashley[1], P. B. Littlewood[3], M. E. Manley[1], B. Mihaila[1], P. S. Riseborough[4], R. K. Schulze[1], D. J. Thoma[1], H. M. Volz[1] and J. L. Smith[1]*

[1]*Los Alamos National Laboratory, Los Alamos, NM 87545*
[2]*Department of Theoretical Physics, Institute for Advanced Studies, The Australian National University, Canberra, ACT 0200, Australia*
[3]*Cavendish Laboratory, Madingley Road Cambridge CB3 0HE United Kingdom; and*
[4]*Department of Physics Temple University, Philadelphia, Pennsylvania 19122-6099 USA*

We report the work function Φ for single and polycrytalline α-uranium over the temperature range 173≤T≤873K. Photoemission spectroscopy (PES) measurements were made at energies 21 eV and 40 eV. Surfaces were prepared *in situ* by sputter/annealing cycles under UHV conditions. Like most metals the polycrystalline specimens showed little temperature dependence, the change in the work function $\Delta\Phi$ = 1 %. Conversely the single crystal changed by 5 % over the same temperature range. A sudden temperature arrest in $\Delta\Phi$ centered at 500 K was detected in the single crystal. This arrest coincides with the ductile-to-brittle (dilation change) transformation temperature.

KEYWORDS: α-U, Photoemission

1. Introduction

Although uranium is the fundamental element to nuclear physics, many of its condensed matter properties challenge modern band structure theory. The properties are thought to originate from an unusual temperature dependence of strong electron-phonon coupling. The sensitivity of some low-temperature properties such as superconductivity and the charge density wave state vary with sample purity and crystallinity, confirming this assumption.[1)] Surprisingly these effects persist to 500 K resulting in a ductile-to-brittle transformation - an unusual plateau in the constitutive properties[2)] as shown in Fig. 1. The ductile-to-brittle transformation, shown as open squares corresponding to the right-hand y-axis, is a manifestation of temperature activated dislocation and twinning mechanisms.[3)] Although fundamental information on this transformation is sparse, it has long been assumed that it is partly electronic in nature. Therefore, it was anticipated that PES measurements would be a sensitive probe of the ductile-to-brittle transformation. Motivated by 1) large harmonic softening of the phonons and a sharpening of the phonon spectra of high purity uranium single crystals,[4)] and by 2) large changes in the vibrational density of states with moderate impurity additions[5)] we investigate the high temperature PES spectra of α-uranium.

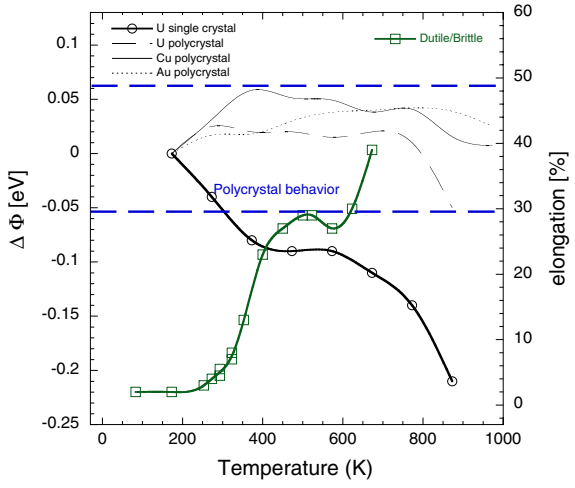

Fig. 1. The open square symbols show the temperature dependence of the macroscopic elongation under load as reported from.[2)] The open circles show our work function results for the single crystal. $\Delta\Phi$ is expressed as the absolute difference from the lowest temperature value of Φ. The squares, triangles, and inverted triangles show our results for polycrystalline samples of uranium, gold, and copper, respectively.

2. Experiment

PES measurements were made using a Perkin-Elmer Physical Electronics Model 5600 ESCA System. Excitation radiation was generated using a SPECS UVS 300 lamp producing from spectral lines from He I and He II with energies 21.21 eV and 40.81 eV, respectively. These crystals grow as very thin, faceted parallelepipeds with typical dimensions of 6 x 6 x 0.2 mm^3. The single crystal was oriented with the c-axis perpendicular to the instrument analyzer. The crystal orientation was confirmed by low-energy electron diffraction and back-reflection Laue. Samples of polycrystalline copper, gold, and arc-melted single-crystal uranium were also measured as standards.

3. Results and Discussion

We show the temperature dependence of $\Delta\Phi$ for both polycrystal and single crystal uranium in Fig. 1. In this figure $\Delta\Phi$, taken as the absolute difference between the lowest temperature value, pertains to the left y-axis. One sees that the polycrystal shows little temperature dependence except at approximately 800 K, where there is a decrease in both the single and polycrystalline samples approaching the α-to-β structural phase transition

*E-mail: jlsmith@lanl.gov

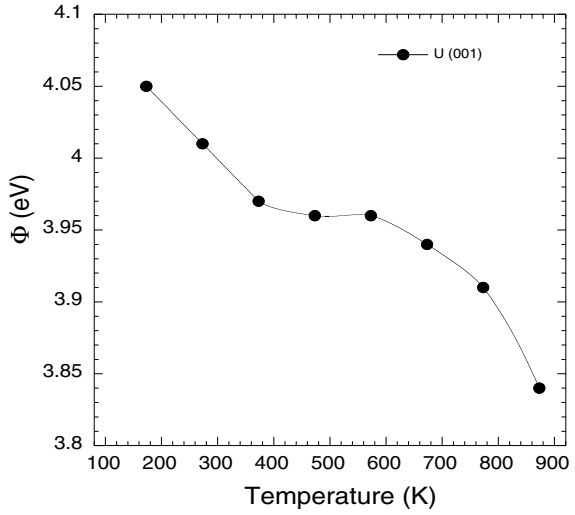

Fig. 2. Temperature dependence of Φ for single crystal along (001) for α-uranium. Presumably the decrease in Φ approaching 900 K signals the impending α-to-β structural phase transition at 935 K.

(T_{tr}=935K). To compare polycrystalline uranium with free electron metals, measurements were made on copper gold, also shown on Figure 1. One sees that the temperature dependence mirrors that of polycrystalline uranium. So much so that the blue dashed limits marks a region in $\Delta\Phi$ where polycrystal results typically fall. Similar results have been reported for polycrystalline uranium.[7] Polycrystalline results depend of crystallographic orientation. The relaxation of the of the distance between surface layers from the bulk-spacing distance can be positive or negative, and they vary with different faces. So it is better to measure a single crystal than to have an average over orientations in a polycrystal.

To the best of our knowledge, Fig. 2 is the first measurement of Φ in single crystal uranium. It shows a rather large variation with temperature, nearly 5 times as large as the polycrystalline samples. At present phenomenological models that rely on the temperature dependence of the volume thermal expansivity[7] are the only explanation for the temperature dependence of Φ in the light actinides. In light of this it seems reasonable to speculate about the similarity between the temperature dependence of the work function and the constitiutive properties, shown on the right hand y-axis. The change in macroscopic elongation with temperature mirrors the temperature dependence of the work function. Although this is simply a comparison between a bulk and a surface property, we suspect that that there may be a common cause for these two anomalies.

From a historical perspective it is interesting to note that 100 years ago Albert Einstein published three papers, one explaining why dim light of higher energy could cause the ejection of electrons from a metal surface while intense light of lower energy could not, giving rise to the concept of the work function.[8] This work had such an impact on science that this year has been called the World Year of Physics 2005.

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1) G. H. Lander, E. S. Fisher, and S. D. Bader, Adv. Phys. **43** (1994) 1.
2) D. N. R. Taplin and J. W. Martin: J. Nucl. Mater. **10** (1963) 134.
3) D. J. Thoma, A. M. Kelly and W. L. Hults, *unpublished data* (2000).
4) M. E. Manley, B. Fultz, R. J. McQueeney, C. M. Brown, W. L. Hults, J. L. Smith, D. J. Thoma, R. Osborn, and J. L. Robertson: Phys. Rev. Lett. **86** (2001) 3076.
5) M. E. Manley, W. L. Hults, J. C. Cooley, R. E. Hackenberg, D. J. Thoma, M. W. Koby, J. L. Smith and K. Littell: Phys. Rev. B **72** (2005) 184302.
6) M. E. Manley, G. H. Lander, H. Sinn, A. Alatas, W. L. Hults, R. J. McQueeney, J. L. Smith and J. Willit: Phys. Rev. B **67** (2003) 052302.
7) T. Durakiewicz, A. J. Arko, J. J. Joyce, D. P. Moore and S. Halas: Surface Science **478** (2001) 72.
8) A. Einstein: Ann. Phys. (Leipzig) **17** (1905) 132.