UC Irvine

UC Irvine Previously Published Works

Title

Inelastic neutron scattering study of U₃Pt₃Sb₄

Permalink

<https://escholarship.org/uc/item/0nv0z5wf>

Journal

Physica B: Physics of Condensed Matter, 206-207(C)

ISSN

0921-4526

Authors

Rainford, BD
Adroja, DT
Severing, A
et al.

Publication Date

1995-02-01

DOI

10.1016/0921-4526(94)00491-D

Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

Inelastic neutron scattering study of $\text{U}_3\text{Pt}_3\text{Sb}_4$

B.D. Rainford^{a,*}, D.T. Adroja^a, A. Severing^b, E.A. Goremychkin^c,
J.D. Thompson^d, Z. Fisk^d

^a*Physics Department, Southampton University, Southampton SO17 1BJ, UK*

^b*Institut Laue Langevin, BP156, F-38042 Grenoble Cedex 9, France*

^c*ISIS Facility, Rutherford Appleton Laboratories, Chilton, OX11 0QX, UK*

^d*Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

Abstract

Inelastic neutron scattering measurements on cubic $\text{U}_3\text{Pt}_3\text{Sb}_4$ reveal well-defined crystal field and spin-orbit excitations, consistent with a $\text{U}^{4+} 5f^2$ configuration of the uranium ion. The magnetic ground state is a crystal field singlet. This may be taken as confirmatory evidence that this compound is not a Kondo insulator, but a band semiconductor.

The magnetism of uranium-based compounds is fascinating since the 5f electrons show behaviour intermediate between a localised and an itinerant nature. This is due to the greater delocalisation of the 5f electrons (compared with, for example, the 4f electrons in the rare earths), as a consequence of their hybridisation with electrons on neighbouring atoms. The localised limit is, in fact, rather rare for uranium: there are only a handful of U intermetallics which display a perfectly defined valence state, as indicated by the existence of well-defined crystal field splittings.

Our interest in the cubic compound $\text{U}_3\text{Pt}_3\text{Sb}_4$ arose from the fascinating behaviour of the isostructural compound $\text{Ce}_3\text{Pt}_3\text{Bi}_4$. This is a Kondo insulator [1], with a transport gap of 5 meV; it also displays a 'spin gap' of 12 meV in its dynamical susceptibility [2], as measured by inelastic neutron scattering. $\text{U}_3\text{Pt}_3\text{Sb}_4$ was another possible candidate for a Kondo insulator compound [3], since it shows a semi-conducting behaviour in its resistivity, has a very small electronic heat capacity coefficient γ , and a temperature independent susceptibility at low temperatures. The

family of Kondo insulators all appear to show moderately large hybridisation between the f electrons and the conduction electrons, i.e. they can be classed as intermediate valence materials. It is relatively easy to distinguish this class of materials by their spin dynamics: inelastic neutron scattering measurements show a rather broad quasi-elastic response. The spin gap then develops at low temperatures as a hole in this response at low frequencies. Our study of $\text{U}_3\text{Pt}_3\text{Sb}_4$ was designed to investigate the character of the dynamical response, and to look for evidence of spin gaps.

Measurements were made on a large polycrystalline sample of $\text{U}_3\text{Pt}_3\text{Sb}_4$ using the HET time of flight spectrometer at the ISIS pulsed neutron facility, Rutherford Appleton Laboratories. Spectra were measured with incident neutron energies of 20, 60, 120, 250 and 600 meV, with the sample held at 14 K in a top loading closed cycle refrigerator. It is clear from the data (Figs. 1, 2, 3) that the compound displays clear crystal field (CF) and intermultiplet excitations. The number of CF excitations is consistent with a $\text{U}^{4+} 5f^2$ configuration. Spectra measured with 20 meV incident neutrons showed no sign of a quasi-elastic response at low temperatures. We deduce that the crystal field ground state is a singlet. The U ion in the

* Corresponding author.

Fig. 1. Spectrum of $\text{U}_3\text{Pt}_3\text{Sb}_4$ measured with 120 meV incident neutrons at 14 K. The histogram is an estimate of the phonon scattering extrapolated from the high angle detector bank.

cubic $\text{Y}_3\text{Au}_3\text{Sb}_4$ structure is at a site with a rather low point symmetry (S_4), so we would expect nine singlet states from the crystal field, and therefore eight excitations from the ground state. The broad peak seen near 25 meV in Fig. 1 (data for 120 meV incident neutrons, 2 m detector bank) is seen to have better resolved structure in Fig. 2 (data for 60 meV incident neutrons, 4 m detector bank). The solid line in Fig. 2 is a fit to five Lorentzian peaks, centred at 12.1, 23.9, 25.7, 30.0 and 36.5 meV. In addition there is the well-defined peak at 69.5 meV (Fig. 1). Therefore six

Fig. 2. Spectrum of $\text{U}_3\text{Pt}_3\text{Sb}_4$ measured in the low angle 4 m bank for 60 meV incident neutrons. The estimated phonon scattering has been subtracted. The solid line is a fit to five Lorentzians (see text).

out of eight possible excitations energies are determined. This rules out the possibility of U^{3+} ($5f^3$, for which we would expect five crystal field doublets and four excitations) or U^{5+} ($5f^1$ —three CF doublets, two excitations). The data are probably the most detailed excitation spectra for a uranium ion measured by neutrons.

The data presented in Fig. 3 were measured with 600 meV incident neutrons, and the points show the spectrum after subtraction of the scattering from the empty sample holder. The peak near 400 meV energy transfer corresponds to the $^3\text{H}_4$ to $^3\text{F}_2$ intermultiplet transition of the U^{4+} ion. A fit to a Lorentzian line shape gives the peak centre as 392 meV. In UO_2 this excitation is found at 448 meV, while in UPd_3 it occurs at 385 meV [4]. The shift to lower energies can be understood as arising from a reduction in the Slater F_2 integral parameter. It is significant that the shift is smaller in semi-conducting $\text{U}_3\text{Pt}_3\text{Sb}_4$, compared with metallic UPd_3 .

The singlet crystal field ground state explains the temperature independent susceptibility of this compound at low temperatures. The relatively narrow line widths of the crystal field excitations indicate that the uranium ion is highly localised. This rules out the possibility that $\text{U}_3\text{Pt}_3\text{Sb}_4$ is a Kondo insulator. The line widths of the excitations do not increase significantly with temperature, up to 100 K, suggesting only weak spin-lattice relaxation. This would be consistent with a low carrier density, suggested by the low value of the Sommerfeld coefficient γ of the heat

Fig. 3. Spectrum of $\text{U}_3\text{Pt}_3\text{Sb}_4$ measured with 600 meV incident neutrons, after subtraction of scattering from empty sample holder. The peak at 392 meV is attributed to the $^3\text{H}_4$ to $^3\text{F}_2$ intermultiplet transition of the U^{4+} ion.

capacity and the activated form of the electronic conductivity. The neutron data is supportive of the conclusion that $\text{U}_3\text{Pt}_3\text{Sb}_4$, like other $\text{U}_3\text{T}_3\text{Sb}_4$ compounds, is a band semi-conductor [5].

For the S4 point symmetry of the U site it can be shown [6] that there is only one second-order crystal field parameter B_2^0 , two independent fourth-order (B_4^0 and B_4^4) and three sixth-order parameters (B_6^0 , B_6^4 and B_6^{-4}). We have verified this by direct calculation with the superposition model approach, using a suitable choice of quantisation axes. The coefficient of the O_4^{-4} term can be made to vanish by rotation of the coordinates about the azimuthal angle. Further work is in progress to determine these parameters by fitting the measured spectral profiles.

References

- [1] M.F. Hundley, P.C. Canfield, J.D. Thompson, Z. Fisk and J.M. Lawrence, *Phys. Rev. B* 42 (1990) 6842.
- [2] A. Severing, J.D. Thompson, P.C. Canfield, Z. Fisk and P. Riseborough, *Phys. Rev. B* 44 (1991) 6832.
- [3] P.C. Canfield, J.D. Thompson, Z. Fisk, M.F. Hundley and A. Lacerda, *J. Magn. Magn. Mater.* 108 (1992) 217.
- [4] R. Osborn, K.A. McEwen, E.A. Goremychkin and A.D. Taylor, *Physica B* 163 (1990) 37.
- [5] T. Takabatake, S. Miyata, H. Fujii, Y. Aoki, T. Suzuki, T. Fujita, J. Sakurai and T. Hiraoka, *J. Phys. Soc. Japan* 59 (1990) 4412.
- [6] U. Walter, *J. Phys. Chem. Solids* 45 (1984) 401.