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Temperature-dependent x-ray diffraction study of Pd/Cu site interchange in non-Fermi liquid UCu₄Pd *

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A pair distribution function (PDF) analysis of temperature-dependent x-ray diffraction measurements from UCu₄Pd is presented. Fits to the displacement parameters (u^2 's) with a Debye model show better agreement with a model that includes 25% of the Pd atoms on 16e (Cu) sites. In addition, significant non-thermal disorder is observed in the Cu environment, in contrast to previous measurements of local order in the U-Cu pairs.

PACS numbers: 72.15.Qm, 61.10.Ht, 71.23.-k, 71.27.+a

UCu₄Pd displays typical non-Fermi liquid (NFL) characteristics, such as a logarithmically diverging electronic part of the heat capacity, $\Delta C_{el}/T$ and the magnetic susceptibility, $\chi(T)$, as well as linear behavior in the low temperature electrical resistivity, $\rho(T)$ [1]. X-ray absorption fine-structure (XAFS) measurements [2] have shown that between 20-30% of the Pd atoms in this nominally *C15b* structure sit on the 16e (nominally Cu) sites. Although this amount of disorder is fairly substantial, subsequent XAFS measurements [3] of the U-Cu pairs did not display the kind of disorder necessary for the Kondo disorder model (KDM) [4] to generate the measured divergences without invoking some other mechanism besides simple lattice disorder combined with a tight-binding model for the *f*-electron/conduction

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electron hybridization energy, V_{fd} . At this time, arguments exist both in favor and against the three leading theoretical candidates for explaining the NFL behavior in this system, namely, the KDM, various Griffiths' phase models [6, 5] and proximity to a true antiferromagnetic quantum critical point [7]. Understanding the details of the lattice disorder remains an important component for verifying and utilizing these theories.

Although the evidence from XAFS for the Pd/Cu site interchange is strong, there is still no confirming independent experiment corroborating the XAFS result. In particular, a room-temperature neutron powder diffraction study found little difference between a fit that included some site interchange and one that did not [8]. One way to discern between these kinds of fits is to perform a diffraction study to obtain the displacement parameters (u^2 's) as a function of temperature. We have performed such measurements, choosing to analyze the data by fitting to the pair-distribution function (PDF) obtained from the Fourier transform of the reduced structure factor, $F(Q) = (S(Q) - 1)Q$. For this study, the results should be equivalent to the usual Rietveld refinement of the structural parameters, but the technique holds some future promise for better isolating the local distortions.

Details of the sample-growth procedure are described elsewhere [8]. X-ray scattering measurements were performed at 11D-C beam line of BESSRC-CAT, Advanced Photon Source, Argonne National Laboratory. A fixed energy of 98.29 keV was selected with a Si(220) single crystal monochromator. Elastic and inelastic (Compton) scattering data were collected with a solid state detector. The inelastic part was removed with a theoretical model [9] as the data were analyzed. Data were fit using the PDFFIT codes [10]. After other corrections, such as, background removal, multiple-scattering removal and absorption correction, were applied, the structure factor was isolated. A PDF obtained through the Fourier transform of $F(Q)$ with Q between 0.04 and 32 \AA^{-1} is shown in Fig. 1 (a). The PDF data between 1.5 - 10 \AA were fit with two different models, normal (dashed-line) and site-interchange (solid-line). The site-interchange model assumed 25% of the Pd atoms were on the 16e site and 25% of the 4c site was occupied by Cu. The final fit is nearly independent of the choice of model, as indicated in Fig. 1 (b). The lattice constant and the displacement parameter (u^2) of U are independent of the choice of model. However, the u^2 's of Pd and Cu are model dependent. Since the x-ray scattering factor of Pd is larger than Cu, for the site-interchange model the fit reduces the displacement parameter of Pd while increasing Cu's, because a larger displacement parameter diminishes the peak intensity. This result is consistent with Chau *et al.* [8].

Figure 2 shows the temperature dependence of the displacement parameters of each atomic species. Using a Debye model, the displacement parameters were fit for the site-interchange and normal models. Results of the

Table 1. Debye temperatures and offsets (u_{static}^2) relative to a Debye model of the measured displacement parameters.

Model	U		Pd		Cu	
	Θ_D (K)	u_{static}^2 (\AA^2)	Θ_D (K)	u_{static}^2 (\AA^2)	Θ_D (K)	u_{static}^2 (\AA^2)
site-interchange	201(20)	0.0008(5)	263(13)	0.0009(4)	552(30)	0.0047(2)
no-interchange	213(16)	0.0009(3)	282(11)	0.0019(2)	428(16)	0.0035(2)

fits are summarized in Table 1. In this analysis, temperature-independent offsets between the models are considered to be due to distortions or disorder relative to the fitting model, and are therefore evidence against a given model. The U data shows a small offset, consistent with zero, for both models. The Pd u^2 only shows a significant offset for the no-interchange model, which is evidence that the site-interchange model is a better explanation of these data. Interestingly, the Cu atoms show significant disorder in both models. This long range disorder (that is, observed on the ~ 10 \AA scale) is in contrast to the observation of local order in the U-Cu pairs observed in the XAFS experiments [3].

In conclusion, we studied site-interchange in UCu_4Pd with PDF analysis and found that we could only discern between the normal and the site-interchange models through an analysis of the temperature-dependent displacement parameters. These diffraction results are more consistent with a site-interchange model, however, significant additional disorder is observed in the Cu environment, likely induced by distortions due to the Pd atoms on the $16e$ sites. Such distortions are likely perpendicular to the U-Cu pairs, since no such disorder is observed in those pairs from XAFS experiments.

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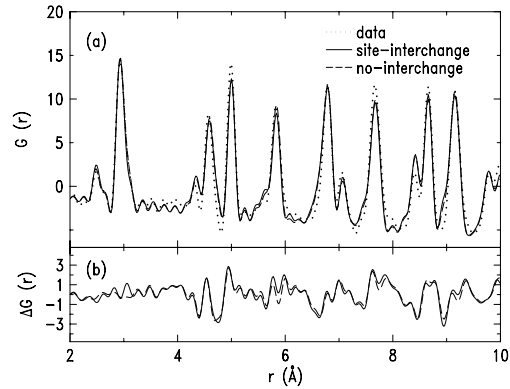


Fig. 1. (a) PDF data (dotted line) at 20 K and best fits with site-interchange (dashed line) and normal (solid line) models. (b) shows the difference between fit and data.

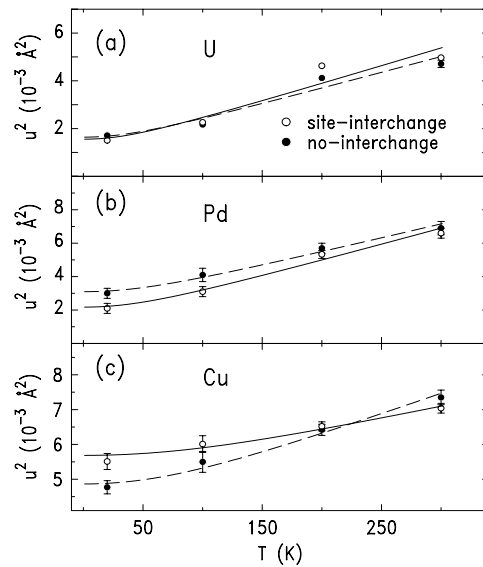


Fig. 2. Thermal factors of (a) U, (b) Pd, and (c) Cu, are obtained from the PDF analysis with two different models, site-interchange (open-circle) and normal (solid dot). The lines are a best fit (Table 1) with a Debye model for site-interchange (solid line) and normal (dashed line).

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