Non-Fermi Liquid Behavior, Lattice Disorder, and Clustering in f-electron Intermetallics

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Introduction

The electronic and magnetic properties of many *f*-electron intermetallics are well described as a collection of noninteracting quasiparticles, that is, with Fermi liquid theory (FLT) [1]. Recently, there has been mounting interest in so-called "non-Fermi liquid" (NFL) systems, where atypical electronic and magnetic properties have been measured [2]. Most theories of NFL behavior in *f*-electron intermetallics are at least partially based on the idea of a quantum critical point (QCP) [3], that is, a critical point between Ruderman-Kittel-Kasuya-Yosida (RKKY) and Kondo ground states at zero temperature. As such, many researchers believe that NFL behavior can be generated in a well-ordered crystalline material. This notion poses a problem because many of the materials exhibiting NFL behavior are known to be disordered. Some researchers have therefore been exploring models where disorder is a necessary component. These theories include Kondo disorder [4], where a distribution of *f*-electron/conduction-electron interaction strengths creates NFL behavior with essentially no new physics and can be described by QCP models with the addition of lattice disorder, which creates a larger region in phase space for the critical point (Griffiths' phase) [5]. Therefore, the presence of a disordered lattice is a central dividing line between the various theories.

Our research into this subject has so far focused on the $UCu_{5-x}Pd_x$ system and on UCu_4Pd in particular. This system was thought to be an example of a chemically ordered NFL system until x-ray absorption fine structure (XAFS) measurements at the U L_{III}, Cu K, and Pd K edges [6-8] revealed that a large fraction (~25%) of the Pd atoms were sitting on the 16*e* site of the C15*b* lattice, rather than the 4*c* site as had been previously believed. Interestingly, the measured disorder is not considered to be appropriate for explaining the NFL properties [7, 8].

To further understand the effects of disorder on NFL systems, we undertook a number of high-energy x-ray powder diffraction experiments, to better understand both the effects of disorder in some NFL systems and intermediate-length scales, which are not accessible to XAFS or conventional diffraction experiments. In addition, although the evidence from XAFS for the Pd/Cu site interchange is strong, no independent experiment has yet corroborated this result. For instance, a room-temperature neutron powder diffraction study found little difference between a fit that included some site interchange and one that did not [9]. One way to discern between these kinds of fits is to perform a diffraction study to obtain the displacement parameters (u^2) 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 promise for better isolating the local distortions in the future.

Methods and Materials

Four materials were chosen for these experiments. In this report, we focus on a UCu_4Pd sample from the same (unannealed) batch described in Refs. 6 and 7. A sample of $YbAgCu_4$ (very well ordered with the same crystal structure as UCu_4Pd) was also measured. These results are already reported on in Ref. 10. We also measured the disordered NFL $CeRhRuSi_2$ [11] and the NFL heavy-fermion superconductor $CeCoIn_5$ [12]. Samples were ground to a fine powder, passed through a 20-μm sieve, loaded into quartz tubes, and placed into a He Displex. X-ray scattering measurements were collected in a Laue configuration at BESSRC beamline station 11-ID-C at the APS. 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 [12] as the data were analyzed. Data were fit by using the PDFFIT codes [13]. After other corrections, such as background removal, multiple-scattering removal, and absorption corrections, were applied, the structure factor was isolated.

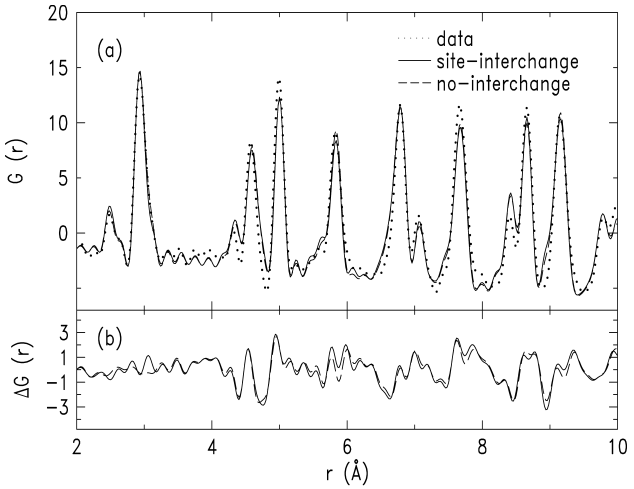

FIG. 1. Panel (a) shows the PDF data (dotted) at 20K and best fits with a site-interchange model (dashed) and a normal model (solid). Panel (b) shows the difference between the fit and the data.

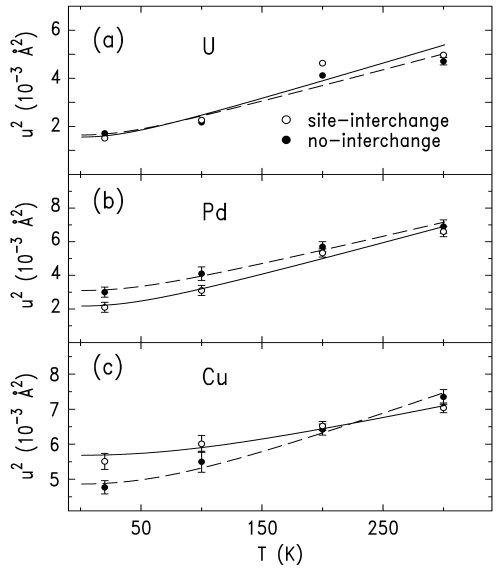

FIG. 2. Thermal factors are obtained from the PDF analysis with two different models, site-interchange (open-circle) and normal (solid dot), for (a) U, (b) Pd, and (c) Cu. Lines are the best fit with a Debye model for the site-interchange (solid) and normal (dashed) fitting models.

Results

A PDF $G(r)$, from UCu_4Pd data, obtained through the Fourier transform of $F(Q)$ with Q between 0.04 and 32 $Å^{-1}$, is shown in Fig. 1(a). The PDF data between 1.5 and 10 Å were fit with two different models, normal (dashed line) and site-interchange (solid line). The site-interchange model assumed that 25% of the Pd atoms were on the 16*e* site and 25% of the 4*c* sites were 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 also 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, the fit reduces the displacement parameter of Pd while increasing that of Cu for the site-interchange model, because a larger displacement parameter diminishes the peak intensity. This result is consistent with Ref. 9. Figure 2 shows the temperature dependence of the displacement parameters of each atomic species. We used a Debye model to fit the displacement parameters for the site-interchange and normal models. Further results of the fits are summarized in Ref. 10.

High-quality data and fits were also obtained from $CeRhRuSi_2$ and $CeCoIn_5$. These data agree with the nominal structures well, with no indications of lattice disorder, apart from the random Rh/Ru site substitution within the *I4/mmm* space group for $CeRhRuSi_2$.

In this analysis, temperature-independent offsets in the Debye models are considered to be due to distortions or disorder relative to the fitting (normal or site-interchange) model and are therefore evidence against a given model. The U data show a small offset, consistent with zero, for both models. The Pd u^2 only shows a significant offset for the no-interchange model (0.0019 $Å^2$ versus 0.0009 $Å^2$ for the site-interchange model), which is evidence that the site-interchange model provides 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 Å scale) is in contrast to the observation of local order in the U-Cu pairs observed in the XAFS experiments [7, 8]. However, a similar effect was observed in the fits to the $YbAgCu_4$ data and thus may be an experimental artifact.

A similar analysis of the data from both $CeRhRuSi_2$ and $CeCoIn_5$ indicates very little disorder, especially for $CeCoIn_5$. Although these results will be the topic of a future paper, they continue to indicate that very little bond length disorder exists in the NFL materials we have studied so far. Therefore, the concepts of chemical pressure are likely to be applicable.

All of this analysis is aimed at getting a fairly general picture of any lattice disorder present in these systems. However, in the Griffiths' phase theories [5], some level of clustering is expected, especially in the theories that emphasize antiferromagnetic interactions. Therefore, two important future experiments remain to be performed on, at least, UCu_4Pd. First, if very high-Q data can be obtained (beyond about 40 $Å^{-1}$), it should be possible to differentiate between Cu-Cu pairs (nominally at 2.49 Å) and short Pd-Cu pairs due to Pd atoms on the nominally Cu sites (2.56 Å according to the XAFS measurements

[6, 7]. In addition, this may allow a rough determination of any Pd-Pd pairs where both Pd atoms are on the nominally Cu sites and thereby allow for an estimate of any possible clustering. Another possible route for obtaining such clustering information would be to perform a difference-PDF experiment around the Pd K edge to isolate only those scattering paths that involve Pd atoms. We have attempted such experiments, at both beamline 7-2 at the Stanford Synchrotron Radiation Laboratory and beamline 12-BM at the APS, with little success.

Discussion

We have reported site-interchange in UCu_4Pd with PDF analysis of high-energy x-ray powder diffraction data and found that we could discern between the normal model and the site-interchange model only through an analysis of the temperature-dependent displacement parameters. No other disorder is unambiguously observed. A similar conclusion is reached from fits to $CeRhRuSi_2$ and $CeCoIn_5$ data. This study perpetuates the possibility that chemical substitution in many NFL materials does not introduce further lattice disorder and can, in fact, be thought of as applying chemical pressure.

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