Probing the Effect of Electronic Substitution on the Correlated Electron Physics in $CeNi_2Ge_2$

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Introduction

The topic of strong electronic correlations in crystalline materials has attracted intense interest for several decades. Such interactions produce unusual behaviors including heavy charge carrier quasiparticles, unconventional superconductivity, exotic magnetism, and even the breakdown of Fermi-liquid behavior.[1] While these phenomena are intriguing, the ability to describe them theoretically and to control them in real materials remains a long-standing challenge. Experimentally, one method that can be used to achieve such control is chemical substitution, since a change in the composition of a material changes its properties through variation of parameters such as the unit cell volume and charge carrier density. Here we investigate Ge → As substitution in the strongly correlated electron material, $CeNi_2Ge_2$.[2] Such tuning is expected to independently control the chemical potential, and will provide insight into how the strong electronic correlations are thus modified.

Motivation

A central purpose of this study is to probe a newly proposed global phase diagram (Figure 1) that describes the nearly independent effects of electron/hole doping and unit cell volume on the magnetic and non-magnetic phases that occur in Ce compounds with the $ThCr_2Si_2$[3] structure.

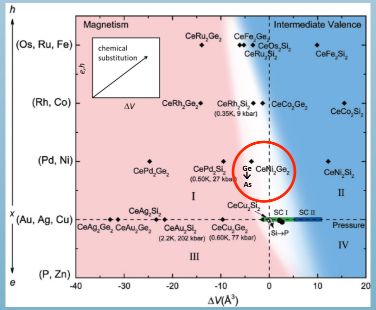


Figure 1. *Proposed Global Phase Diagram of Ce Family Compounds with the $ThCr_2Si_2$ Structure.*[4]

Methodology

Single Crystals of $CeNi_2(Ge_{1-x}As_x)_2$ were grown using the self-flux growth method in alumina crucibles sealed under vacuum in quartz ampoules.[4] Starting elements Ce:Ni:Ge:As had a purity of at least 99.9%, and were massed according to the stoichiometric ratios 1 : 9.5 : 9.5-x : x respectively. The ampoule was heated in resistive furnace according to the heating curve seen in Figure 2. At the final temperature, 975°C, the ampoule was centrifuged in order to separate the crystals from the flux.

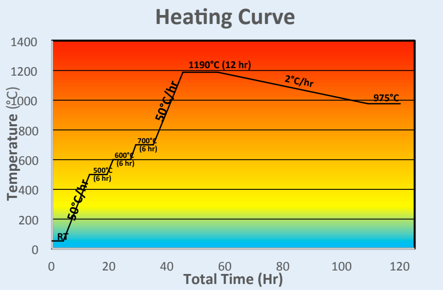
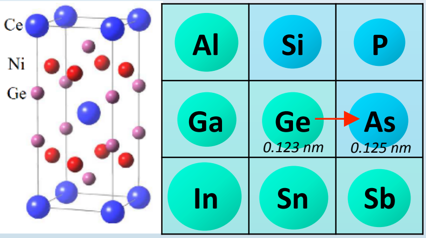


Figure 2. *Heating Curve for the $CeNi_2(Ge_{1-x}As_x)_2$ Growth.*

Figure 3. *Sample images of as-grown $CeNi_2(Ge_{1-x}As_x)_2$ single crystals. Imperfections on surface are residual flux.*

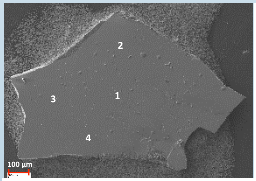
Above: Single Crystal Shaped like the State of Florida (by chance).

Results



Al	Si	P
Ge	Ge → As	As
In	Sn	Sb

Figure 4. *(Left) Crystal Structure of $CeNi_2Ge_2$. (Right) Section of Periodic Table of Elements Surrounding Ge.*



X_{nom}	X_{meas}
0.0125	0.08
0.0375	0.145

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Element    Wt %     At %   K-Ratio     Z        A        F
  CeL     36.43    21.20   0.3425   0.9116   1.0149   1.0161
  NiK     28.25    39.23   0.3030   1.1038   0.9476   1.0257
  GeK     32.13    36.10   0.3145   1.0136   0.9656   1.0000
  AsK      3.19     3.47   0.0314   1.0107   0.9731   1.0000
 Total   100.00   100.00
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Figure 5. *(Top Left) Scanning Electron Microscope Image of a $CeNi_2(Ge_{1-x}As_x)_2$ Crystal with Four Scanned Locations Indicated. (Top Right) Nominal Concentrations (x_{nom}) and Actual Measured Concentrations (x_{meas}) of for two crystals from separate batches. (Bottom) Example Results from actual EDS Measurement.*

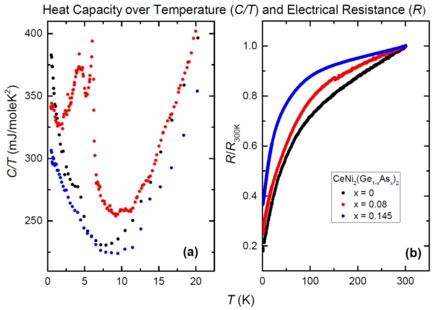

Figure 6. *(a) Heat Capacity Divided by Temperature (C/T) vs. T and (b) Room T Normalized Resistivity, R/R_{300K}, $CeNi_2(Ge_{1-x}As_x)_2$.*

Results (Cont'd.)

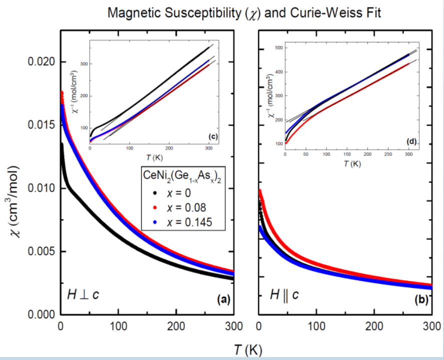

Figure 7. *(a) and (b), Magnetic Susceptibility $\chi = M/H$ in Magnetic Field H = 5000 Oe Applied both Parallel (//) and Perpendicular (⊥) to the c-axis for $CeNi_2(Ge_{1-x}As_x)_2$. (c) and (d) Curie-Weiss Fits.*

Conclusion and Further Research

Ge → As substitution in $CeNi_2Ge_2$ is accomplished using the flux growth technique, where the parent compound behavior is noticeably impacted. Study will be furthered through growth and testing of crystals with higher arsenic concentrations, as well as conducting further tests such as neutron scattering and NMR.

Acknowledgements

Single Crystal Diffraction measurements were performed by Nate Falb using equipment in the Siegrist Lab at the NHMFL. Electron dispersive spectroscopy measurements were performed by Yu-Che Chiu and Fumitake Kametani at the ASC-NHMFL.

I would like to thank the National High Magnetic Field Laboratory, the Research Experiences for Undergraduates program, Dr. Ryan Baumbach and the Baumbach Group for the opportunity to perform research in a wonderful environment this summer.

This work was performed at the NHMFL, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490, the State of Florida and the DOE. A portion of this work was supported by the NHMFL User Collaboration Grant Program (UCGP).

Works Cited: [1] G. R. Stewart, Reviews of Modern Physics 73, 797 (2001). [2] P. Gegenwart, *et al.* Phys. Rev. Lett. 82, 1293 (1999). [3] Y. Lai, *et al.* Separating the effects of volume and carrier concentration on the heavy fermion state. Manuscript in Preparation. [4] P.C. Canfield, *et. al.* Cryst. Growth 225 (2001) 155.