ANNUAL SCIENTIFIC PROGRESS REPORT

National Nuclear Security Administration Stewardship Stockpile Academic Alliance Research Grant #DE-FG52-06NA26205

The focus of this grant, entitled "Experimental investigations of magnetic, superconducting, and other phase transitions in novel f-electron materials at ultra-high pressures using designer diamond anvils," is to explore the novel properties of f-electron compounds under pressure, with a particular emphasis on the physics of superconductivity, magnetism, and their interactions. This report is a synopsis of the research that was undertaken from 6/2006 – 6/2007.

PRESSURE MEASUREMENTS

a. URu_2Si_2

The moderately heavy fermion compound URu_2Si_2 was discovered over 20 years ago, but still remains a perplexing and interesting compound owing mostly to its transition into an ordered state near T_0 = 17.5 K. The order parameter of this state has yet to be identified, even after 20 years of research, and, as such, this ordered state has come to be known as the "hidden order" (HO) state. In addition to this HO state, URu_2Si_2 exhibits another phase transition into a superconducting state at T_c = 1.5 K. It has been inferred previously from neutron diffraction data that these two ordered phases at ambient pressure coexist, prompting concerns over the nature of the HO state and its relation to superconductivity. To that end, we have prepared high-quality, single crystal specimens of URu_2Si_2 and prepared them for high-pressure, electrical resistivity measurements, utilizing a hydrostatic piston-cylinder clamp device in our facility, to explore the HO and superconducting states up to approximately 25 kbar.

The HO transition manifests itself in the resistivity as a trough-and-peak structure reminiscent of the spin density wave (SDW) transition of elemental chromium. We found that the qualitative shape of this transition persists up to the highest pressures measured. The HO transition temperature, T_0, was found to exhibit a distinct kink in its pressure dependence at P_c = 15 kbar: below P_c, $T_0(P)$ is linear with a slope near 0.1 K/kbar; and, above P_c, $T_0(P)$ is linear with a slope near 0.23 K/kbar. This kink at P_c corresponds to a dramatic change in the magnitude of the ordered moment as determined through neutron diffraction, where the magnetic moment above P_c is consistent with bulk antiferromagnetism (AFM). With increasing pressure, the superconducting critical temperature, T_c, was smoothly and monotonically suppressed towards zero temperature near 15 kbar, or P_c. The coincidence of the disappearance of superconductivity and the kink in T_0, possibly indicative of a crossover from a HO state to an AFM state, suggested that the HO and superconducting states were in competition. Previous specific heat measurements suggested the HO state partially gapped a portion of the Fermi surface (FS). From this, we analyzed the transition temperatures of the ordered states of URu_2Si_2 in the context of a competition for FS fraction. Through this analysis, we found that the increase in T_0 corresponds to an increase in the portion of FS gapped by the HO transition. The increase in this gapped portion of the FS leaves fewer electrons to undergo pairing into the superconducting state. Our analysis agrees extremely well with previous specific heat studies under pressure, and indicates that the HO transition fully gaps its portion of the FS near P_c.

By fitting the electrical resistivity to a form including scattering from gapped spin excitations, the magnitude of a gap in the spin-excitation spectrum could be quantified. We found that the magnitude of this gap changes near P_c. Furthermore, the height of the resistive anomaly associated with the HO state decreases with increasing pressure up to P_c, after which it remains roughly constant. This behavior is consistent with the gapping of the FS, indicated by our previous analysis. As the HO state gaps more of the FS, there are fewer states into which quasiparticles can scatter, thus reducing the magnitude of the resistivity. Above P_c, when the HO state has fully gapped its portion of the FS, the number of states into which quasiparticles can scatter is pressure-independent, yielding a pressure-independent value for the magnitude of the resistive anomaly.

The presence of a gap in the spin-excitation spectrum and a gap at the FS strongly suggests that a SDW-like instability occurs at the HO transition temperature T_0. The onset of bulk AFM above P_c is intriguing and could be explained in a SDW scenario where a SDW instability induces local ordering.

b. $URu_{2-x}Re_xSi_2$

The Re-substituted URu_2Si_2 system, $URu_{2-x}Re_xSi_2$, provides a unique opportunity to examine the pressure dependence of a HO state whose ambient pressure transition temperature and associated correlations are suppressed with increasing x. Using high-quality single crystals, we measured several compositions of $URu_{2-x}Re_xSi_2$ under pressure. The qualitative trough-and-peak structure of the HO transition in the pure compound persisted with increasing Re-content, although the absolute value of the resistivity changed due to impurity effects. With applied pressure, the HO transition temperature T_0 exhibited a kink in its pressure dependence at P_c = 15 kbar, identical to the pure compound, for all values of x. This persistence of the value of P_c with Re-content suggests that Re-substitution does little to affect the crossover from the HO state to the bulk AFM state.

c. Thorium

Elemental thorium is a conventional superconductor with an ambient pressure critical temperature T_c = 1.4 K. Previous, pressure-dependent measurements revealed a dramatic decrease in T_c with applied pressure; however, above approximately 100 kbar, the value of T_c remained roughly constant, showing a flat pressure dependence up to nearly 160 kbar. Using high-quality, single crystals, we have investigated the superconducting state of thorium up to high pressures. This work was performed using a beryllium-copper diamond anvil cell (DAC) from our facility. We used a designer diamond anvil equipped with microprobes for electrical resistivity measurements, obtained through our collaborations with Dr. Weir at Lawrence Livermore National Laboratory and Dr. Vohra at University of Alabama, Birmingham. The thorium sample was mounted within a beryllium-copper gasket along with a ruby manometer. Ultra-high pressure measurements at very low temperature were performed within the Kelvinox MX-100 ^{3}He-^{4}He dilution refrigerator within our facility.

The critical temperature was tracked up to pressures near 400 kbar, approaching the limit of the beryllium-copper gasket. The pressure-dependent evolution of the superconducting state was similar to previously reported results: T_c decreased with increasing pressure—although not as steeply as the previous results, possibly due to pressure gradients arising from a lack of a pressure-transmitting media; above approximately 100 kbar, $T_c(P)$ flattened and exhibited little pressure dependence out to nearly 400 kbar. In addition, measurements of the critical field curves were attempted and showed little change at high pressures, consistent with the weak pressure dependence of T_c. The critical fields obtained from this experiment were larger than expected, likely due to an inaccurate determination of the penetration of the field into the sample space.

PROJECT PARTICIPANTS

Faculty

Name: M. Brian Maple

Percent Contribution: 3%

Contribution to Project: Research group leader and Principal Investigator.

Assistant Project Scientist

Name: Pei-Chun Ho

Percent Contribution: 58%

Contribution to Project: Designs and performs laboratory experiments in collaboration with PI.

Post-doctoral Research Associate

Name: Benjamin J. Taylor

Percent Contribution: 7%

Contribution to Project: Designs and performs laboratory experiments in collaboration with PI and Assistant Project Scientist. Prepares thin film high temperature superconducting materials.

Graduate Students

Name: Jason R. Jeffries

Percent Contribution: 100%

Contribution to Project: Prepares intermetallic samples and performs high-pressure measurements, responsible for development and implementation of ultrahigh pressure facility.

Name: Nicholas P. Butch

Percent Contribution: 100%

Contribution to Project: Prepares intermetallic samples and performs measurements of magnetic and transport properties of f-electron materials including high pressure measurements.

Name: Todd A. Sayles

Percent Contribution: 34%

Contribution to Project: Prepares intermetallic samples and performs magnetic, transport, and specific heat measurements of f-electron materials.

Name: Diego Zocco

Percent Contribution: 28%

Contribution to Project: Prepares intermetallic samples and performs high pressure electrical resistivity and susceptibility measurements.

Name: Ryan E. Baumbach

Percent Contribution: 18%

Contribution to Project: Prepares thin film and polycrystalline samples of intermetallic f-electron and high temperature superconducting compounds and performs magnetic and electrical transport measurements.

Name: Daniel J. Scanderbeg

Percent Contribution: 14%

Contribution to Project: Prepares thin film and polycrystalline samples of cuprate-based high temperature superconducting compounds and performs magnetic and electrical transport measurements.

Undergraduate Lab Assistants

Name: Colin McElroy

Percent Contribution: 95%

Contribution to Project: Prepares intermetallic f-electron samples and assists in electrical resistivity, magnetization, and specific heat measurements to characterize materials with potentially interesting high pressure properties.

Name: Benjamin T. Yukich

Percent Contribution: 3%

Contribution to Project: Prepares single crystal specimens of URu_2Si_2 and its various substituted systems for high pressure electrical resistivity measurements.

PUBLICATIONS

- D. D. Jackson, J. R. Jeffries, Wei Qiu, Joel D. Griffith, S. McCall, C. Aracne, M. Fluss, M. B. Maple, S. T. Weir, and Y. K. Vohra, "Structure-dependent ferromagnetism in Au_4V studied under high pressure," *Phys. Rev. B* **74** 174404 (2006).

- N. P. Butch, J. R. Jeffries, B. T. Yukich, and M. B. Maple, "Tuning of Hidden Order and Superconductivity in URu_2Si_2 by Applied Pressure and Re Substitution," in *Actinides 2006* **—** *Basic Science, Applications and Technology*, edited by K. J. M. Blobaum, E. Chandler, L. Havela, M. B. Maple, and M. Neu (Mater. Res. Soc. Symp. Proc. **986**, Warrendale, PA, 2007), 0986-OO02-03.

- J. R. Jeffries, N. P. Butch, B. T. Yukich, and M. B. Maple, "Competing Ordered Phases in URu_2Si_2: Hydrostatic Pressure and Re-substitution," *Phys. Rev. Lett.* (submitted).

- J. R. Jeffries, N. P. Butch, B. T. Yukich, and M. B. Maple, "The Evolution of the Ordered States of Single Crystal URu_2Si_2 under Pressure," *Phys. Rev. B* (in preparation).

- J. R. Jeffries, N. P. Butch, B. T. Yukich, and M. B. Maple, "The Hidden Order State under Pressure in Single Crystals of $URu_{2-x}Re_xSi_2$," *Phys. Rev. B* (in preparation).

- T. A. Sayles, W. M. Yuhasz, J. Paglione, T. Yanagisawa, J. R. Jeffries, M. B. Maple, Z. Henkie, A. Pietraszko, T. Cichorek, R. Wawryk, Y. Nemoto, and T. Goto, "Thermodynamic and transport studies of the ferromagnetic filled skutterudite compound $PrFe_4As_{12}$," *Phys. Rev. B* (in preparation).

- R. E. Baumbach, P.-C. Ho, T. A. Sayles, M. B. Maple, R. Wawryk, T. Cichorek, A. Pietraszko, and Z. Henkie, “Non-Fermi liquid behavior in a fluctuating valence system, the filled skutterudite compound $CeRu_4As_{12}$,” (in preparation).

ABSTRACTS

- S. Francoual, N. Harrison, M. Jaime, S. Baily, A. Lacerda, N. P. Butch, and M. B. Maple, “Effects of Rhenium Doping on the High Magnetic Field versus Temperature Phase Diagram of URu_2Si_2,” *Bull. Am. Phys. Soc.* **51**, 574 (2007).

- J. R. Jeffries, N. P. Butch, B. T. Yukich, and M. B. Maple, “The Evolution of the Hidden Order Phase in $URu_{2-x}Re_xSi_2$ under Pressure,” *Bull. Am. Phys. Soc.* **51**, 575 (2007).

- N. P Butch, J. R. Jeffries, B. T. Yukich, T. A. Sayles, J. Paglione, P. –C. Ho, and M. B. Maple, “The Search for Quantum Criticality in the $URu_{2-x}Re_xSi_2$ Phase Diagram,” *Bull. Am. Phys. Soc.* **51**, 575 (2007).

INVITED PRESENTATIONS

- M. B. Maple, “Novel types of superconductivity in f-electron materials,” Conferment of the Honorary Professorship of the W. Trzebiatowski Institute for Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw, Poland, September 6, 2006.

- M. B. Maple, “Strongly correlated electron phenomena in filled skutterudite lanthanide osmium antimonides,” 6th International Conference on f-elements, Wroclaw, Poland, September 8, 2006.

- M. B. Maple, “Tuning of hidden order and superconductivity in URu_2Si_2 by applied pressure and Re doping,” Fall MRS’06 Actinides III Symposium, Boston, Massachusetts, November 27, 2006

- M. B. Maple, “Experimental investigation of magnetic, superconducting, and other phase transitions in novel f-electron materials at ultrahigh pressures,” National Nuclear Security Administration Stewardship Science Academic Alliance Symposium, Washington, D.C., February 5-7, 2007.

- J. R. Jeffries, “Competing Ordered Phases in URu_2Si_2,” Arete Associates, Thousand Oaks, CA, March 16, 2007.

- N. P. Butch, “Probing the Unusual Properties of URu_2Si_2 via Applied Pressure and Re Substitution,” Los Alamos National Laboratory, Los Alamos, NM, March 23, 2007.

- J. R. Jeffries, “Competing Ordered Phases in URu_2Si_2: Pressure and Substitution,” Lawrence Livermore National Laboratory, Livermore, CA, March 27, 2007.

- J. R. Jeffries, “Competing Ordered Phases in URu_2Si_2: Pressure and Substitution,” Sandia National Laboratory, Livermore, CA, May 9, 2007.

- J. R. Jeffries, “Competing Ordered Phases in URu_2Si_2: Pressure and Substitution,” Stanford University, Stanford, CA, July 11, 2007.

POSTER SESSIONS

- J. R. Jeffries, N. P. Butch, D. D. Jackson, S. T. Weir, Y. K. Vohra, and M. B. Maple, “Evolution of Ordered States under Pressure in f- and d-electron Systems,” Poster Session: National Nuclear Security Administration Stewardship Science Academic Alliances Symposium, Washington, D. C., February 5-7, 2007.