Our goals for the second year of this grant were to perform experiments on f-electron materials at low temperatures, high magnetic fields, and high to ultrahigh pressures and to continue setting up our ultrahigh pressure diamond anvil cell facility. In order to accomplish this goal, it was necessary to design and implement the required techniques and equipment essential to investigating materials under extreme conditions. In the previous year, we have studied several interesting materials at low temperatures, high magnetic fields, and high pressures as well as completing the fabrication and installation of our dilution refrigerator system. The following sections of this report serve to highlight the progress we have made towards developing an ultrahigh pressure research facility at UCSD, investigating f-electron and novel materials under extreme conditions, as well as future directions we plan to pursue.

ULTRAHIGH PRESSURE DIAMOND ANVIL CELL FACILITY

a. Dilution Refrigerator

As many f-electron materials exhibit structural, magnetic, and superconducting phase transitions at low temperature, a dilution refrigerator proves to be an extremely useful experimental tool for the thorough investigation of these materials. We purchased our Oxford Kelvinox MX-100 dilution refrigerator during the first year of this grant and it arrived at our laboratory near the end of the first year of this grant. Before installing and operating the refrigerator it was necessary to construct a structure to support the weight of the refrigerator and the associated liquid helium dewar as well as to vibrationally isolate the refrigerator system from the floor as well as the pumps that would be used to operate the system. Air springs were chosen as vibrational dampers for this structure and have proven more than capable of supporting the weight of the system.
while virtually eliminating heating due to external vibrations. An additional hoisting structure was fabricated for the purpose of raising and lowering the dilution refrigerator insert, which is equipped with a sliding seal to prevent excessive liquid helium boil-off, into the dewar. When used with the sliding seal, this hoisting structure provides a quick cooling time for the insert, which takes approximately six hours to cool from 300 K down to liquid helium temperatures, while keeping liquid helium boil-off below 20 liters. With the ancillary structures completed, the dilution refrigerator was tested and certified by an Oxford technician who successfully operated the refrigerator down to ~ 13 mK. An experimental tail was fabricated and attached to the mixing chamber of the dilution refrigerator, the source of the cooling power of the system, on which a variety of high-pressure cells could be attached. The experimental tail provides a thermal link to the mixing chamber while placing the pressure cells and their associated samples in the center of the magnetic field produced by the 9 T superconducting magnet contained in the liquid helium dewar. Electrical resistivity measurements on URu$_2$Si$_2$ under pressure were successfully performed below 50 mK using the dilution refrigerator system.

b. In situ Pressure Calibration

We have previously constructed a ruby fluorescence microscope for calibrating the pressure inside the sample chamber of the diamond anvil cell at room temperature. In addition to room temperature measurements of the pressure, we would also like to be able to calibrate the pressure at low temperatures to account for pressure losses due to differential thermal contraction with changing temperature. To that end, we have designed a custom fiber-optic patch cord for use at the low temperatures and moderate vacuum associated with our measurement apparatus. This patch cord will gather laser light from a 573 nm solid state laser and direct it along the multimode fiber into the cryostat directly at the ruby within the diamonds. The same fiber will collect the fluorescence spectrum and direct it out of the cryostat to the spectrometer. This system should allow pressure calibration as a function of temperature down to 1 K. With this information, we will attempt to minimize the pressure losses due to differential thermal contraction by using additional materials or components with different coefficients of thermal expansion such as Delrin or Belleville springs.
PRESSURE MEASUREMENTS

a. PrOs$_4$As$_{12}$

Single crystals of the filled skutterudite compound PrOs$_4$As$_{12}$ were obtained from Z. Henkie (Polish Academy of Sciences) and were characterized via measurements of bulk magnetization, electrical resistivity, and specific heat. In collaboration with S. McCall and M. W. McElfresh (Lawrence Livermore National Laboratory), specific heat measurements were extended to high field. In this heavy fermion compound, two bulk phase transitions are observed at ~2.2 K, below which the material shows behavior consistent with antiferromagnetic order. Specifically, magnetization measurements in field demonstrate a lack of magnetic hysteresis and provide evidence of a Pr$^{3+}$ crystal field-split magnetic ground state, while the low-temperature electrical resistivity shows behavior consistent with the opening of a magnon energy gap. In applied magnetic field, the antiferromagnetic phase is suppressed by ~1.7 T, makes a transition into another ordered phase, currently speculated to be quadrupolar in nature, that is suppressed by ~3.2 T. In the paramagnetic state of PrOs$_4$As$_{12}$, both resistivity and specific heat measurements show clear signs of single-ion Kondo behavior. Under applied pressure, the electrical resistivity was measured at several pressures up to 23 kbar, between 1 K and 300 K, and no significant pressure dependence was found. However, it is expected that at higher pressures, the low-temperature ordered phases of PrOs$_4$As$_{12}$ will be suppressed, and we plan to measure the electrical resistivity at pressures exceeding 30 kbar.

b. Au$_4$V

We have measured the electrical resistivity of the ferromagnetic compound Au$_4$V under hydrostatic pressure up to 25 kbar. The electrical resistivity of Au$_4$V as a function of temperature displays a characteristic kink corresponding to ferromagnetic ordering in the system at approximately $T_C = 45$ K. With applied pressure, the Curie temperature of a single-crystal sample increases to a value of $T_C = 52$ K. A crystalline sample of Au$_4$V was filed into a fine powder and, with our collaborator Sam Weir, loaded into a DAC at LLNL. High-pressure electrical resistivity measurements were performed on the Au$_4$V powder at LLNL up to a pressure of almost 200 kbar. Initial results are in excellent
agreement with the lower pressure hydrostatic cell data taken at UCSD. The high-pressure diamond anvil studies indicate that the Curie temperature of Au₄V continues to increase to a value of Tᵥ ≈ 90 K at a pressure of approximately 200 kbar. Above 200 kbar, no resistive transition due to the onset of ferromagnetism could be resolved. The dramatic increase in the Curie temperature suggests an increase in the magnetic exchange interaction.

c. Dilute Au₁₋ₓVₓ Alloys

In an effort to understand the dramatic increase in the Curie temperature—and likely the magnetic exchange interaction—of Au₄V, we have synthesized low V concentration Au₁₋ₓVₓ alloys with x = 0.1, 0.05, 0.02, 0.01, 0.005, and 0.0025. Electrical resistivity measurements of the alloyed samples exhibit characteristic signatures of the presence of local moments in the Au host with the concentrations below 1% (x = 0.01) displaying an upturn in the electrical resistivity at low temperature, a classic hallmark of the Kondo effect. A sample with 0.5% vanadium was mounted in a hydrostatic pressure cell and electrical resistivity measurements were performed from 300 – 1 K. The characteristic minimum is seen to increase slightly with increasing pressure. Analysis of the data reveals an increase in the Kondo temperature Tᵥ of the sample from ~180 K at ambient pressure to ~200 K at almost 30 kbar. Like the Curie temperature in Au₄V, the increase in the Kondo temperature of Au₀.₉₉₅V₀.₀₀₅ suggests an increase in the magnetic exchange interaction. With our collaborators at Lawrence Livermore National Laboratory, another sample of Au₀.₉₉₅V₀.₀₀₅ has been mounted in a designer diamond anvil cell for electrical resistivity measurements to ultrahigh pressures. The data from these ultrahigh pressure electrical resistivity measurements will be compared to the data from Au₄V at ultrahigh pressures in an effort to determine the interesting magnetic behavior in Au₄V.

d. URu₂Si₂

The f-electron compound URu₂Si₂ is a very interesting material that displays many novel properties including: moderately heavy fermion behavior with m* ~ 25 mₑ; a “hidden” order transition at Tᵥ = 17.5 K, which coincides with a small-moment antiferromagnetic transition at the same temperature; and a superconducting transition at Tᵥ ≈ 1.3 K. Previous experiments from our laboratory suggest that the “hidden” order
and the superconductivity both occupy a fraction of the Fermi surface. Upon the application of pressure, it has been shown that the “hidden” order gives way to bulk antiferromagnetism at $P_c \approx 15$ kbar a pressure near where the superconductivity extrapolates to zero temperature. We have performed electrical resistivity measurements as a function of temperature, pressure, and applied magnetic field. Preliminary results of the evolution of the upper critical field $H_{c2}$ for the superconductivity reveal a change in curvature at low fields. The results of these measurements indicate that the superconductivity and $H_{c2}(0)$, the upper critical field at zero temperature, are indeed suppressed at $P_c = 15$ kbar. Further studies of the “hidden” order transition as a function of pressure are underway to determine the nature of this material in the vicinity of 15 kbar.

**FUTURE DIRECTIONS**

We will continue our efforts to establish an ultrahigh pressure facility at UCSD by completing the integration of fluorescence spectroscopy into our cryostats and fabricating diamond anvil cells for use at dilution refrigerator temperatures. We will continue our efforts at the investigation of quantum criticality and heavy fermion behavior in f-electron materials. We will study the effects of pressure on the moderately heavy fermion compounds URu$_2$Si$_2$ and URu$_{2-x}$Re$_x$Si$_2$ in order to investigate the possible correlation between superconductivity, “hidden” order, and antiferromagnetism as well as the possible reemergence of a second superconducting phase at ultrahigh pressures where antiferromagnetism is suppressed. We will investigate the alloyed compound CeRh$_{1-x}$Co$_x$In$_5$ as a function of pressure and magnetic field for $x > x_c$, where $x_c = 0.75$, in an effort to explore the possible interplay of the field-tuned and pressure- or chemical substitution-tuned quantum critical points in this system. We plan to investigate other compounds in the 1-1-5 family of compounds—those compounds with chemical formula LnMX$_5$, where Ln represents an element from the lanthanide series; M represents a transition metal element Co, Rh, or Ir; and X represents a group IIIIB element Ga or In—in order to better understand the quantum criticality present in CeCoIn$_5$. Doping of rare earth elements on the Ce site could provide valuable insight into the role of valence fluctuations on the non-Fermi liquid behavior seen in these systems. Additional ultrahigh
pressure measurements on members of the skutterudite family of compounds, such as those previously measured like PrFe$_4$Sb$_{12}$ or PrOs$_4$As$_{12}$, could elucidate interesting as of yet unrevealed behavior at high pressures where the lattice constants are more greatly affected.
PROJECT PARTICIPANTS

Faculty
Name: M. Brian Maple
Percent Contribution: 10%
Contribution to Project: Research group leader and Principal Investigator.

Administrative Assistant
Name: Carolyn Rosado
Percent Contribution: 25%
Contribution to Project: Performs tasks benefiting the project including preparing or assisting in the development of manuscripts, illustrations, publication artwork, technical presentations; arranging travel of PI, graduate students, or collaborating faculty; and maintaining our research database.

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

Name: Nicholas P. Butch
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**Contribution to Project:** Prepares intermetallic compounds with an emphasis on the synthesis of 1-1-5 alloys.


**PUBLICATIONS**


**ABSTRACTS**


**INVITED TALKS OF P. I., M. B. MAPLE**


Strongly correlated electron phenomena in Pr-based filled skutterudite compounds, 2nd US-Japan Workshop on Synchrotron Radiation and Nanoscience, San Diego, California, April 4-6, 2005.


Pr-doped YBCO: a model system for studies of high temperature superconductivity and vortex physics, UC/Los Alamos Workshop, University of California, Santa Barbara, May 13-14, 2005.


Non-Fermi liquid behavior near magnetic quantum critical points in uranium based compounds, International Conference on Strongly Correlated Electron Systems (SCES05), Vienna, Austria, July 26-30, 2005.


**POSTER SESSIONS**