TECHNICAL PROGRESS REPORT

MAJOR RESEARCH ACCOMPLISHMENTS

Development of the relaxation method of specific heat for measurements at miliKelvin temperatures and high magnetic fields

During this period of time we have made a strong strife to further develop the specific heat method for very low temperatures and the highest accessible fields. The specific heat is the most important probe that provides the most basic information on low temperature sates of solids. It is of particular importance to strongly correlated electron systems and heavy fermions, whose main hallmark is strongly enhanced low temperature specific heat. The discovery of non-Fermi liquid behavior in f-electron systems, manifesting itself as divergent C/T (specific heat divided by temperature) at T=0, necessitated accurate measurement of the specific heat to the lowest accessible temperatures. Finally, it has been shown that external magnetic field is the most appropriate and readily available parameter that can tune a physical system to a quantum critical point. Moreover, the magnetic field is extremely convenient and indispensable tool to study all heavy fermion systems due to their proximity to magnetism and since their relevant energies are of order $\mu_B H$. However, despite this paramount importance of such studies, neither of the traditional techniques of specific heat is appropriate for low temperatures and high fields, particularly for small samples. Among the three group of methods: adiabatic, ac, and relaxation, this last one seem to be most promising. But even in this case we have showed that large systematic errors obscure the results due to a weak coupling between electrons and nuclear degrees of freedom that show up at sufficiently large fields and small temperatures. These large errors are most often incorrectly associated with a conventional t_2 effect (imperfect thermal contact between the sample and addenda). The t_2 correction procedure is inappropriate for these hyperfine effects. In particular, we have showed that in an extreme case of pure Cu at 1 K and fields of order 10 T, the measured time constants relate more closely to a nuclear spin-lattice relaxation time T_1 than to electronic heat capacity. Thus this method has a potential to simultaneously measure the specific heat and nuclear spin-lattice relaxation time T_1. This measurement of T_1 has a large potential for applications in the field regime inaccessible to a traditional NMR technique.

In order to further advance the relaxation technique of the heat capacity in high fields and low temperatures, the P.I. has teamed up with Yasu Takano, one of the founders of Microkelvin laboratory in Gainesville and expert on very low temperatures and NMR techniques, and hired a postdoctoral associate (Hiroyuki Tsujii) having previous experience in measurements at low temperatures and high fields. We have constructed a calorimeter for a top loading dilution refrigerator for the Tallahassee 20 T superconducting magnet and performed successful measurements on $Ce_{0.01}La_{0.99}Al_3$ and S=1 quasi 1D-antiferromagnet to temperatures as low as 60 mK. In addition, we have built two calorimeters (for each of the P.I.'s) for resistive 33 T magnets. We have successfully tested them to 350 mK and 30 T.

The group has spent four weeks in Tallahassee tackling various technical problems and performing measurements, in addition to time in Gainesville devoted to the design and construction of apparatus. In the process we have discovered that a number of problems related to heat capacity measurements were not treated correctly in the past. We have solved some of them, such as distributive heat capacity (poor sample conductance) or contribution due to weak link. Short discussion of these problems and their solution has been submitted for publications. More thorough treatment will be presented in papers in preparation to the Review of Scientific Instruments.

Role of chemical disorder and sample annealing on non-Fermi liquid behavior of UCu_4Pd and related $UCu_{5-x}Pd_x$ alloys

Two studies have been undertaken to study the role of chemical disorder on Cu/Pd sites on the ground states in UCu_4Pd, a canonical U-based non-Fermi liquid, and related alloys. The first study performed in a collaboration with D.E. MacLaughlin by low temperature muon spin relaxation technique found inhomogeneously distributed f-electron spin fluctuations rates and time-field scaling of relaxation rates in both UCu_4Pd and in $UCu_{3.5}Pd_{1.5}$, indicative of long-lived spin fluctuations. In UCu_4Pd the scaling exponent is small and temperature independent but varies with temperature in a similar manner to that observed for some canonical spin-glasses. This last observation corroborates our initial hypothesis that the ground state in $UCu_{3.5}Pd_{1.5}$ corresponds to a spin-glass with $T_f=0$ (quantum spin glass).

The objective of the second project initiated is to optimize the preparation conditions of UCu_4Pd and related alloys and to study how the improvements in the synthesis of $UCu_{5-x}Pd_x$ affect the low temperature properties. One of our observations is that the high temperature annealing affects much more strongly the low temperature state of pure UCu_5 and alloys corresponding to small x than alloys with x close to 1. This is obviously inconsistent with the general belief that the Cu/Pd interchange is the relevant disorder. Our other conclusion is that the room temperature resistivity ratio (RRR=R(300 K)/R(0)), previously argued (and generally believed) to be a measure of sample quality can not be used as the quality criterion for these alloys. In particular, the annealing increases RRR for x close to 1 but decreases RRR for x close to 0. Our best UCu_5 alloys show very large increase of the resistivity below 1 K suggesting a proximity to a metal-insulator transition. One of our working hypotheses, supported by at least tenfold decrease of the electronic specific heat coefficient below 1 K, is that a temperature driven metal-insulator transition can be observed in this compound provided that the sample is free of Cu-inclusions.

Ground state of $CeAl_3$

Our discovery of impurity driven instabilities in $CeAl_3$, the first discovered heavy fermion material, which for decades has been considered as a canonical example of non-magnetic, Fermi-liquid, strongly m*-enhanced system, has prompted renewed interest in this material. Two competing explanations have been proposed: disordered antiferromagnetism (our hypothesis) and anisotropic Kondo model (AKM). Our continuing work on this material was to distinguish between these two scenarios. Comprehensive alloying and magnetic field studies performed by us clearly disprove the applicability of AKM. Our most recent investigation on strongly dilute alloys (such as $Ce_{0.01}La_{0.99}Al_3$) indicate a single-impurity S=1/2 Kondo ground state with unusually small value of T_K (smaller than 1 K).

Publications acknowledging the support of the Department of Energy

Specific heat of S=1 quasi 1D-antiferromagnet NDMAP in magnetic fields, H. Tsujii, Z. Honda, B. Andraka, K. Katsumata, and Y. Takano, submitted to the 23rd International Conference on Low Temperature Physics, Hiroshima, Japan, August 20-27, 2002

Distributed t_2 effect in relaxation calorimetry, H. Tsujii, B. Andraka, K. Muttalib, and Y. Takano, submitted to the 23rd International Conference on Low Temperature Physics, Hiroshima, Japan, August 20-27, 2002

The alloying study of $CeAl_3$, H. Tsujii, Y. Takano, and B. Andraka, Physica B 312-313, 200 (2002).

Slow spin dynamics in non-Fermi-liquid $UCu_{5-x}Pd_x$, x=1.0 and 1.5, D.E. MacLaughlin, R.H. Heffner, O.O. Bernal, G.J. Nieuwenhuys, J. E. Sonier, M.S. Rose, M.B. Maple, and B. Andraka, Physica B 312-313, 200 (2002).

Calorimeter for a top loading dilution refrigerator in high magnetic fields, H. Tsujii, B. Andraka, and Y. Takano, in preparation for Review of Scientific Instruments (short version submitted to the 23th International Conference on Low Temperature Physics, Hiroshima, Japan, August 20-27, 2002.

Annealing and non-Fermi liquid behavior properties of $UCu_{5-x}Pd_x$, C. Rotundu and B. Andraka, abstract submitted to the conference on Magnetism and Magnetic Materials, November 11-15, 2002; to be published in J. Appl. Phys.

Invited talks

Magnetic field dependence of $Ce_{1-x}La_xAl_3$, March 2002 Meeting of the American Physical Society, Indianapolis, IN

Non-Fermi liquid behavior in UCu4Pd and related alloys, The European Conference Physics of Magnetism'02, Poznan, Poland, July 1-5, 2002; to be published in Physica Status Solidi; plenary talk

Exotic ground states in U-based heavy fermions, International Symposium: 50th Anniversary of the First 5f-Electron Ferromagnet Discovery and 50th Anniversary of Polish Academy of Sciences Foundation, July 6-8, 2002, Wroclaw

WORK PLAN FOR THE NEXT REPORTING PERIOD

High magnetic field specific heat study of canonical heavy fermion systems

We intend to capitalize on our unique ability, developed during the current reporting period, to study materials at extreme temperature/field conditions. The initial measurements to 14 T will be performed in Gainesville and subsequent studies to 33 or 45 T in Tallahassee. The P.I. has applied for two weeks measuring time in the National High Magnetic Field Laboratory in Tallahassee. One week will be for the superconducting 18 T magnet with a dilution refrigerator, the second week for the resistive 33 T magnet. The P.I. intends to apply for the 45 T hybrid magnet (with a dilution refrigerator) during the spring of 2003. There is also a possibility to have access to 45 T field this fall through our collaborator, Y. Takano, who has applied for this system for the fall. Such high field/low temperature studies, in addition to testing theoretical models, carry high potential for important new discoveries.

UCd_{11}- Magnetic phase diagram and "hidden order"

We propose high field study of the specific heat in UCd_{11}. UCd_{11} is one of the better known heavy fermion systems. It orders antiferromagnetically below 5 K (T_N). Its behavior in the paramagnetic state is unique among heavy fermion metals and has been a subject of controversies that have not been resolved yet. The specific heat divided by temperature (C/T) above T_N is linear in T^2, similarly to ordinary metals, albeit with a huge value of the linear coefficient ? (700 mJ/K^2mol). Assuming that this linear specific heat is due to heavy electrons results in a very large value of the entropy at temperatures of order 15 K. In our previous investigation of this metal we have not detected a significant decrease in the specific heat (and entropy) in fields to 16 T. Moreover, we have noticed a puzzling behavior of the antiferromagnetic anomaly and specific heat below T_N. Magnetic field reduces T_N extending the temperature region of C/T proportional to T^2 such that no characteristic increase in C/T, typical of a Kondo effect, has been observed in the highest field used. The objective of the proposed investigation is to get insight into this unique system by reducing its T_N to zero. Based on our results in fields to 16 T, we expect the antiferromagnetism to be completely suppressed by fields of order 30 T. Below are listed some of the specific questions we intend to answer. What is the nature of the state corresponding to T_N=0, does it correspond to a quantum phase transition? Does the "normal-like" temperature dependence of the specific heat extends to the lowest temperatures in fields greater than the critical field? Another specific goal of this study is related to the possibility of observing two different phase transitions in sufficiently high magnetic fields. This possibility has been raised by the zero-field investigation, the study at different combinations of magnetic–field pressures, and the 16 T specific heat study. The zero-field anomaly has a rather unusual shape that has been suggested to be due the superposition of two transitions occurring at nearly identical temperatures. These hypothetical two transitions can be split by fields greater than 16 T.

Normal state of UBe_{13}

UBe_{13} is one of the most fascinating heavy fermion compounds that has been recently classified as a non-Fermi liquid. Were the 45 T hybrid system available to us, UBe_{13} would be the primary choice. Previous magnetic field studies of UBe_{13} have indicated a very week response of the specific heat to magnetic fields in the normal state. Thus the main effect of the field is to suppress the superconductivity to 0 allowing one to study the normal state to low temperatures.

Impurity and magnetic field study of UCu_5

UCu_5 belongs to the least understood and most interesting heavy fermion metals waiting for its re-discovery. It exhibits an unusual coexistence of magnetic order and heavy electrons. Our understanding of heavy fermions is based on the competition of magnetism and Kondo effect. Normally, magnetic order prevents the development of heavy electrons. Conversely, heavy electrons exhaust the paramagnetic entropy preventing a full blown magnetic order. UCu_5 violates both these principles. Heavy electrons develop at temperatures below 5 K i.e., from a magnetically ordered state (T_N=15 K). Secondly, there is a second transition at 1 K that strips quasiparticles of their heavy masses. The low temperature state has been investigated by two groups by magnetic fields. Both groups have indicated a strong sensitivity of the low temperature C/T to magnetic fields, with one group reporting a decrease while the other increase of C/T in fields. This observation correlates with our results showing a strong sensitivity of $UCu_{5-x}Pd_x$ to preparation conditions. The nature of the low temperature state in UCu_5 and how this state evolves upon doping with Pd have obvious bearing on the explanation of the non-Fermi liquid behavior of UCu_4Pd.

Collaborative studies (in progress)

We intend to continue our synergetic collaborations in the investigation of f-electron systems. We have either prepared or will prepare samples for collaborative measurements. These collaborative investigations include:

a) µSR investigation of $Ce_{1-x}La_xAl_3$ alloys with D.E. MacLaughlin (x<0.2). The goal is to investigate the evolution of magnetism between x=0 and x=0.2
b) Optical studies of the 1 K transition in UCu5; a new collaboration with Maureen Reedyk from Canada. The first sample has been shipped, new samples will be sent after further optimization of preparation conditions and initial characterization in the P.I.'s laboratory
c) NMR studies of $U_{1-x}Y_xRu_2Si_2$ with O. Bernal

New materials

We constantly are searching for new materials with unusual low temperature states. We are also assisted in this search by our collaborators from the Polish Academy of Sciences in Wroclaw, Poland.

CONTRIBUTION TO THE DEVELOPMENT OF HUMAN RESOURCES

We were fortunate in securing assistance in our investigations from two individuals, Costel Rotundu (graduate student) and Hiroyuki Tsujii (postdoctoral associate). Costel has joined us mid May as the first year graduate student. He is scheduled to take the preliminary exam in August this year. Based on the comments of his last year teachers he was one of the best first-year students and should not have any difficulties with passing this exam. Since he has already satisfied all core corses requirements the first year he will join us full-time at the end of August (he is spending more than 40 hours a week in tha laboratory already). So far, he has been involved in the synthesis of samples and their characterization by X-ray diffraction. In addition he has learnt cryogenic techniqes and performed specific heat or resistivity measurements under my ot Tsujii's supervision. He has also participated in measurements in Tallahasse.

Hiroyuki Tsujii has been trained in measuring specific heat using the relaxation technique. After spending several weeks in the P.I.'s laboratory and measuring specific heat in fields to 14 T he became a leading person in implementing this technique for high magnetic fields and temperatures to 60 mK in the National High Magnetic Field Laboratory in Tallahassee. He has participated in the design of calorimeters and assembled two calorimeters for measurements at Tallahassee and Gainesville. We hope to support him also during the next budget year.

PERSONEL SUPPORTED BY THIS AWARD

Hiroyuki Tsujii, Postdoctoral Research Associate

COLLABORATIONS

Y. Takano, K. Ingersent, G.R. Stewart (Florida), D.E. MacLaughlin (Riverside), R. Troc (Wroclaw, Poland), M. Reedyk (Brock University, Canada), O. Bernal (CalState)

CURRENT AND PENDING SUPPORT

<u>Current</u>

Synthesis and Study of Unconventional f-Electron Metals, Department of Energy, DE-FG02-99ER45748, February 1, 2002 – January 31, 2005, FY02 $90,016

Synthesis and Characterization of Unconventional Heavy Fermion Materials, National Science Foundation, DMR-0104240, November 1, 2001 – October 31, 2004, FY02 $110,440

<u>Pending</u>

None