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Development of High Magnetic Fields for Energy Research

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Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The primary purpose of work has been to develop the scientific basis for DOE support of a program that would build a novel, nondestructive 100-tesla magnet that would be available as a user facility for cutting-edge, energy-related research and technology at very high magnetic fields.

Background and Research Objectives

High magnetic fields are an extremely powerful tool for understanding the physical properties of materials and for revealing fundamentally new science of condensed matter. Materials research in which very high magnetic fields prove invaluable include studies of high-temperature superconductors, correlated electron phenomena, fermiology, magnetic anisotropy and exchange in rare-earth and transition-metal compounds, field-induced magnetic and electronic phase transitions, reduced dimension/quantum-confined systems, magneto-optical effects, and new routes to synthesis of novel materials. This list, by no means exhaustive, illustrates the key role that magnetic fields play in materials research programs important to the DOE/OBES. In spite of this role, the United States is well behind other developed countries in exploiting the potential of high magnetic fields for energy-related materials research. A positive step, however, has been taken by the National Science Foundation (NSF) to regain U.S. competitiveness in high field research through the establishment of a new National High Magnetic Field Laboratory (NHMFL), an important part of which is at Los Alamos. These new capabilities, available to users within and outside the United States, represent the beginning of an exciting new direction

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for materials research. However, the NSF has set aside no funding at Los Alamos to develop experimental programs that would use these facilities.

Aside from the new NHMFL, there are no high field facilities (above 50 T) in the United States accessible to users in the next decade. Most magnetic field research is conducted at universities, where magnetic fields rarely exceed 10 T. An exception is Princeton University, which has plans for a pulsed magnet capable of reaching 60 T, but this is not a national user facility nor is the 60-T pulsed magnet at Bell Labs. Although excellent science is being performed with existing magnets, these facilities are limited by accessibility, ultimate field, or breadth of experimental techniques and, in most cases, by all three. Opportunities for discovering and understanding new properties of solids and for exploring frontiers opened by the availability of very high magnetic fields and state-of-the-art experimental techniques are substantial. Historically, though, there has not been a community in the United States pushing the limits of materials phenomena at very high fields, an area of research specifically identified as a high priority by National Academy of Science and NSF panels and one that could substantially benefit programs responding to DOE missions in energy research and technology.

To seize these opportunities and respond to a national need, we proposed to establish the foundation for a DOE-NSF partnership that would assure joint leadership for materials research at very high magnetic fields in the United States. This foundation would be built upon the design of a totally new pulsed, high-field magnet that will reach 100 T nondestructively, on implementation of experimental techniques to measure the magnetic properties of materials at these fields, and on initiating forefront high-field research using existing magnets at the NHMFL-Los Alamos. We would expect to attract DOE funding to construct the magnet and to provide support for a principal research team (PRT) composed of DOE and university researchers and led by Los Alamos, whose primary mission would be to perform forefront research on materials at very high fields. This magnet and experimental capabilities, though primarily for PRT use, would be made available to other users of the Los Alamos NHMFL on a basis similar to beamlines at DOE synchrotron and neutron sources.

Importance to LANL's Science and Technology Base and National R&D Needs

This LDRD project represents a new direction in materials research for both Los Alamos and DOE/OBES. Discussions with experimentalists and theorists from DOE laboratories and universities, especially those from University of California (UC) campuses, have indicated enthusiasm for research at fields greater than 50 T and for the

concept of a PRT. The availability of a 100-T nondestructive magnet, reserved primarily for PRT use, will open entirely new opportunities for program development in areas represented by these scientists and important to the DOE mission. Within Los Alamos, the ability to characterize high-temperature superconductors, correlated electron materials, and low-dimensional polymers will be extended and a foundation will be laid for developing new programs in high-density magnetic recording and information-storage devices, high-strength permanent magnet technology, artificially structured materials, and novel routes to materials synthesis, among others. The capability to characterize magnetic properties of electrically conductive materials in 100-T fields and at extremes of very low temperatures will be unique in the world, and by necessity any industrial interest in these areas will come to Los Alamos.

Nuclear and advanced materials are a noted competency of the Laboratory. The addition of a 100-T magnet and techniques to characterize the magnetic properties of these materials at such high fields will represent a significant advancement in this competency, comparable to the introduction of the Los Alamos Neutron Science Center (LANSCE). The potential importance of 100-T nondestructive magnetic fields is recognized explicitly in the Laboratory's Institutional Plan.

Scientific Approach and Accomplishments

Very high magnetic fields, approaching 100 T, are generated in two ways. Most common in Japan and Europe is the discharge of a very large capacitor bank into a single-turn-coil. Much less common is an approach that relies on explosive compression of magnetic flux within a single-turn coil that is magnetically charged initially from a relatively small capacitor bank, as is done at Los Alamos or in Russia. In either approach, the magnet and/or sample under study is destroyed. This is expensive, labor intensive, and not conducive to systematic studies of materials. Further disadvantages include relatively poor field homogeneity, extremely short pulse times (typically a few microseconds), low repetition rates, and restricted access for both personnel and samples. These numerous disadvantages severely limit types of experiments that can be performed and materials that can be studied.

We have realized that the same design techniques being used to develop a NHMFL 60-T long-pulse (100 msec) magnet, powered by a unique 1.4-GW motor-generator set at Los Alamos, could be combined with our existing technology of capacitor-driven multi-turn, relatively long-pulse (10 msec) 50-T magnets to produce a hybrid magnet that reaches 100 T with a pulse duration of 10 msec or more. This would be unprecedented. It would push the combination of pulse duration, volume, homogeneity, and pulsed-rise time several

orders of magnitude beyond any existing 100-T facility and would be nondestructive. The pulse duration alone would be 1000 times longer than is currently available (in Japan). The relatively slow rise time would permit, for the first time, 100-T studies on electrically conducting materials and experiments at very low temperatures.

To achieve all objectives of this project we have

- designed controls of the capacitor bank that would drive the 50-T insert of the 100-T magnet, controls of the 50-T outsert and overall 100-T system controls;
- developed new and improved existing experimental techniques that could be applied to study materials in 100-T fields;
- developed the community of pulsed-field users; and
- demonstrated that interesting new science can be done in very high pulsed fields.

We have been successful in each of these areas, and as a result the DOE/OBES has funded a new Los Alamos program to build a DOE 100-T "beamline" at the NHMFL. This new program, which began in FY 1996, is funded at approximately \$800K/year and is expected to continue at this level for the foreseeable future. Once the 100-T magnet is built, we anticipate that comparable level of support from DOE will allow development of a principal research team, composed of DOE and university researchers and led by Los Alamos, that would perform forefront research at very high magnetic fields. In addition to this \$800K/year, we also have attracted an NSF-funded program (\$125K over two years) to develop the capability to perform specific heat measurements in pulsed fields and at low temperatures. From the point of view of program development objectives, this LDRD project has been successful.

The following points highlight additional accomplishments of this project:

1. New Magnetometer. We have developed a new type of magnetometer capable of measuring the magnetic moment of tiny single crystals in pulsed fields. This device relies on detecting the force produced from the interaction of a magnetic field with the magnetic moment of a sample mounted on the end of an ultra-miniature cantilever. We have fabricated cantilevers from both silicon and GaAs using molecular beam epitaxial growth techniques and have produced 100-nm thick cantilever beams that can measure forces as small as 10^{-13} Newtons. This magnetometer has been tested in static fields to 30 T and at temperatures from 0.03 to 300 K. It is at least as sensitive as any other magnetometer developed and can be used in pulsed fields where others cannot be used.

2. New Dilatometer. We have designed and built a new device that measures field-induced changes in the length (magnetostriction) of a sample. This device, called a dilatometer, is fabricated from single-crystal Si and can resolve relative length changes of

10^{-6} on samples that are only a few millimeters long. Its major advantage is that Si is electrically nonconductive and, therefore, not susceptible to induced currents and associated resistive heating in pulsed magnetic fields, unlike dilatometers constructed from copper or silver.

3. Data Acquisition Systems. We have developed new data acquisition systems that improve the signal/noise ratio by over a factor of two for virtually any measurement (other than optical) that would be made in a pulsed magnet. Along these same lines, we have designed and applied a new approach to magnetoresistivity measurements, probably the most common of all to be made in pulsed fields, that permits these measurements to be made with an accuracy of 0.1% at pulsed fields to 60 T. All of these improvements (1 through 3) are directly applicable to experiments in the 100-T magnet.

4. User Community. By securing approximately \$200K in University of California Directed Research and Development (UCDRD) funding to support visits and experiments by University of California faculty, postdocs, and students, we have begun to develop and nurture an important community of future 100-T users. UC campuses involved in the UCDRD program include San Diego, Los Angeles, Riverside, Santa Barbara, Berkeley, and Santa Clara. Excitement in the broader community of pulsed-field users also is growing. For example, Lucent Technologies/Bell Labs will have their high-field expert spend quarter-time at the NHMFL-Los Alamos Pulsed Field Facility with the explicit intent of becoming a significant user of the 100-T magnet.

5. Kondo Insulators. Correlated-electron materials called "Kondo insulators" are believed to represent a simple limit of the more general and complex problem described by the Anderson Lattice Hamiltonian (ALH). The ALH remains unsolved but is thought to embody the physics appropriate to all strongly correlated-electron materials. Consequently, it is important to study Kondo-insulators, with the hope that their understanding can be generalized to a broader class of materials.

Pulsed-field magnetoresistance measurements by Bell Labs on a prototypical Kondo insulator $\text{Ce}_3\text{Bi}_4\text{Pt}_3$ suggested that a 60-T field changed this material from an insulator to a metal. Their interpretation of this result is that Zeeman splitting of the valence and conduction bands closed the small (~ 60 K) indirect gap in this material. This interpretation does not require any correlated-electron physics and, consequently, does not distinguish this material from any other "ordinary" small-gap system. From earlier neutron scattering experiments, we have shown that this small gap also manifests itself in the magnetic susceptibility of $\text{Ce}_3\text{Bi}_4\text{Pt}_3$.

To test the Bell Labs interpretation, we performed magnetic susceptibility measurements in pulsed fields to 60 T and found no indication for a field-induced decrease

in the gap (Publication 3). This, we believe, can be understood because the important energy scale is set by electronic correlations, and this scale is on the order of 300 K (or equivalently ≥ 300 T), not 60 K. The gap is just a reflection of the underlying correlations, and until these correlations are sufficiently perturbed by a field, there should be no change in the gap.

Our measurements also suggest that the Bell Labs results can be understood if there were a few impurity states in the gap, which is not unexpected and to which resistivity measurements would be most sensitive. Indeed, we find from magnetic susceptibility measurements that impurity states are present and magnetically frozen out in fields of the order of 40 to 50 T.

6. Magnetic-Magnetic Transition. We have discovered a field-induced magnetic-magnetic transition in the correlated-electron antiferromagnet CeRh_2Si_2 . This transition occurs in a field of 25 T at a temperature of 4 K. Careful analysis of the magnetic phase diagram deduced from a series of measurements to 60 T at $4 < T < 70$ K shows that there is striking resemblance of these states to the unexplained magnetic states found in isostructural CeCu_2Si_2 (Publication 4). In CeCu_2Si_2 it is believed that the very small moment magnetic states are associated intimately with the development of heavy-fermion superconductivity below $T_c = 0.9$ K in zero magnetic field. (Indeed, CeRh_2Si_2 also becomes a superconductor with $T_c = 0.40$ K when antiferromagnetism is suppressed by applied pressure.)

This discovery is important because we know that the magnetism in CeRh_2Si_2 is of a local-moment type and we understand its origin. Even though the ordered moments differ by a factor of eight ($\sim 1.7 \mu_B$ in CeRh_2Si_2 versus $\sim 0.2 \mu_B$ in CeCu_2Si_2), we now logically can deduce that the magnetic states in CeCu_2Si_2 are of the local-moment type, but the moments are reduced due to electronic correlation (Kondo) effects. This resolves a long-standing controversy over the origin of magnetism in CeCu_2Si_2 .

Summary

We have made significant scientific discoveries that would not have been possible without very high magnetic fields, have developed or improved experimental techniques directly applicable to research at 100-T fields, have made progress in developing a community of future 100-T users, and have secured new programmatic funding that will enable the construction and use of a dramatically new tool, a 100-T nondestructive magnet, for energy-related research.

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