1. Abstract

The Booster grant our research group received from ISEN enabled us to initiate a theory-inspired approach to the synthesis of new heavy fermion superconductors through harnessing electron delocalization to create a new class of superconductors. Careful consideration of a new theoretical foundation for heavy fermion superconductors suggests that enabling electron delocalization in intermetallic compounds will lead to the discovery of new heavy fermion superconductors. Towards those ends, during the funding period our laboratory identified a list of candidates for synthesis and initiated the synthesis and doping of the selected candidates. Our results arising from ISEN funding were used as the preliminary results section of an application to a federal funding agency.

2. Energy Relevance

The successful fusion of theory and experiment to develop new materials for superconductivity has the potential for a transformative impact in energy transmission. With the increasing consumption of electricity for transportation and the advent of plug-in hybrid and electric cars, superconductivity is rapidly becoming a vital part of the renewable energy picture. While significant research efforts are devoted to portable energy, in the form of batteries and energy generation from renewable resources, inadequate chemistry research is devoted to power transmission. This oversight is dangerous because the DOE projects our established power grid technology cannot sustain the current density required to accommodate new electric technologies.1

3.1 Hypothesis and theory inspired methodology for selecting synthetic targets

The booster grant proposed generating electron delocalization for the rational synthesis of new superconductors relying heavily on the nascent theoretical framework.2-4 Our approach to synthesizing new superconductors is inclusion of first-row transition metals that demonstrate itinerant magnetism in their bulk form. This approach presupposes that itinerant magnetism in cerium, neptunium, and uranium is the direct cause of superconductivity in heavy fermion superconductors. Thus, we assume that another element that behaves similarly in the bulk form could create a superconducting compound.

In the early 1980s, Kemtko delineated a clear dichotomy between metals with bonding electrons, which are delocalized in a conductor, and localized magnetism mediating electrons.5 Very few metals reside on the border of this trend and are able to both conduct and couple magnetically. The new theoretical framework for heavy fermion superconductivity leans heavily on this observation, making note that one f-element metal on the border of itinerancy and magnetism resides in every heavy fermion superconductor, those elements being Ce, U, Np and Pu.6-8 In known heavy fermion superconductor compounds $T_c$ increases as the radial extent of the orbitals increases. In the context of a double Kondo effect this trend is attributed to the increased overlap between the magnetism mediating electron and the conduction band, creating a “stronger” double Kondo effect (as depicted in Figure 1). The greater radial extent of the 3d orbitals relative to 4f and 5f orbitals suggests that synthesizing heavy fermion superconductors with transition metals will increase the transition temperature. Thus far, the highest transition temperature for a heavy fermion superconductor is 18 K with no theorized upper limit.9 To test this theory, we incorporated iron, manganese, and cobalt into...
structure types known to produce heavy fermion superconductivity while carefully considering how the different metallic radii will affect orbital overlap.

2.2. Synthetic targets and results

Rational synthetic design of new heavy fermion systems requires an approach to incorporating Mn, Fe, or Co into a lattice capable of mediating heavy fermion behavior. The compounds best suited for studying heavy fermion superconductivity are layered compounds in which planes of atoms, which mediate superconductivity, reside. Such planes are indicated in Figure 2. Most known heavy fermion superconductors crystallize in tetragonal space groups with planes of heavy atoms interspersed with main group elements. Based on this observation we employed two design principles for synthetic target selection: tetragonal cells with a plane of transition metal atoms.

Our initial doping studies focused on substitution of different transition metals to develop a continuum of properties. We synthesized the solid solutions of Y4Fe5Co3-xGa16, Y4Co5Mn3-xGa16, Y4Ni5Cr3-xGa16 and Y4Co5Cr3-xGa16 pictured in Figure 2. These compounds were synthesized using standard solid-state synthesis techniques, beginning by arc melting binary precursors. Binary precursors were then combined under an inert atmosphere and ground together. The powder was then combined with excess of a molten flux and transferred to an alumina crucible, capped with quartz wool and placed into a quartz tube. The quartz tubes were sealed under vacuum and placed into a furnace. Samples were heated above the melting point of the flux and removed from the furnace while the flux was molten. The tubes were then turned over and centrifuged to perform a high temperature filtration. The remaining flux was removed by chemical etching prior to physical measurements. Compounds were characterized by single crystal x-ray diffraction, scanning electron microscopy, and powder x-ray diffraction. The aggregate of these results confirmed the incorporation of other transition metals onto the lattice sites. Magnetic data were obtained for these compounds, demonstrating a change from the parent compounds. Additional magnetic characterization will be necessary to fully extract the new magnetic properties produced through doping.

Invited Presentations Resulting from this ISEN Funding

(1) Missouri University of Science and Technology - Rolla, MO, October 2013
(2) Argonne National Laboratory - Lemont, IL, July 2013
(3) National High Magnetic Field Laboratory - Tallahassee, FL, August 2013
(4) ACS National Meeting “Exxon Award Symposium” – San Francisco, CA, August 2014

3. References