LA-UR- 01-2512

Title:

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Submitted to:

4th Pacific Rim International Conference
Honolulu, Hawaii
December 11 -15, 2001

Los Alamos
NATIONAL LABORATORY

Magnetism and Superconductivity of Uranium and Intermetallic Compounds

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Heat capacity, resistivity, and phonon density of states have been measured on uranium and reported already. Many of the results are on single crystals of purity that has been unavailable before. Some intermetallic compounds have been measured that are in the class of so-called heavy-fermion materials. We present here the latest results along with a discussion of the occurrence of superconductivity or magnetism in these materials.

Keywords: *uranium superconductivity, uranium magnetism*

1. Introduction

Before 1970, uranium compounds seemed to exhibit superconductivity or magnetism in an unpredictable manner. Then Hill demonstrated that if the uranium atoms were closer together than about 3.51 Å, the compound could be superconducting, and if they were further apart, the compound could be magnetic.(1) This occurs, we now know, because the 5f electrons of the light actinide elements sit of the edge between localized and itinerant behavior, that is, magnetism or superconductivity, respectively.(2) Thus if the 5f wavefunctions overlap, they hybridize, and if they do not overlap, they localize. Thus Hill showed that in compounds, as long as the nearest-neighbor atom did not hybridize with the uranium 5f electrons, the spacing of the uranium atoms determined superconductivity or magnetism. This was not a sufficient condition, merely a necessary one. Later, a few uranium compounds were discovered with a large uranium spacing that were nonetheless superconducting.(3) These, so-called, heavy-fermion superconductors have an electron effective mass for the superconducting electrons that can be a thousand times heavier than that of a bare electron. This comes about by the formation of quasiparticles that show their localized-electron parentage, where the mass would be infinite, and there is not yet a well accepted theoretical explanation for this behavior.

The most interesting problem to understand is that it is the same electrons in uranium that compete for different ground states, unlike superconductors with localized 4f electrons that order magnetically while other electrons become superconducting. Recently, the ferromagnetic compound UGe_2 was seen to be superconducting under pressure.(4) Such a possibility had been predicted theoretically two years earlier,(5) but it had not been well received because of the long-held view that the only form of magnetism that could not co-exist with superconductivity was ferromagnetism because it had a finite average magnetization. The most exciting question right now is to determine the pairing state of the superconducting electrons. Here higher purity samples would be a great help.

2. Pure Uranium

There is a need for better samples for all of these materials. Usually it is the uranium that needs to be purer. We have single crystals of uranium now that are made by electrodeposition in a salt bath at a temperature such that the room-temperature α phase is stable, and they are of very high purity.(6) Single crystals cannot be grown from the melt because there are two higher temperature phases. Earlier work on single crystals and a fine summary of the properties of uranium metal is in Lander *et al.*(7) As these authors discuss, there are many open questions. Recently, some of us found a remarkable softening of the phonons as polycrystalline uranium powder was heated to almost its melting point.(8) The phonon density of states softened far more than expected from its lattice expansion, and the phonons remained harmonic. Clearly the bonding of the uranium must change as a function of temperature. This is unique among elements that have been measured.

Recent heat-capacity measurement on the single crystals showed that the three known charge-density-wave transformations below liquid-nitrogen temperature were first order, showed an improved value for entropy, and found a low-temperature Debye temperature that agreed with ultrasound measurements for the first time ever.(9) These measurements also showed a bulk superconducting transition. The superconductivity of uranium is a very old and very troublesome issue as described by Lander *et al.*(7) Very simply stated, if the charge density waves are inhibited by pressure (≈ 1.1 GPa), uranium is a bulk superconductor. Polycrystalline samples, where the orthorhombic crystal structure leads to strains at low temperature, have usually been thought only to be superconducting at grain boundaries, that is, filamentary superconductivity. This question has been controversial for fifty years.

3. Experimental Procedure and Results

We report here on resistivity measurements performed on the single crystals grown in the salt bath. The crystals tend to contain planar inclusion of salt, and we selected samples where we thought we could see the fewest inclusions. The resistivity was measured at the National High Magnetic Field Laboratory at Los Alamos and Tallahassee. A four-lead technique was used with the leads attached with conducting epoxy. An 18 T superconducting magnet was used to apply a magnetic field. Temperatures below 1.4 K were obtained with a dilution refrigerator.

Figure 1 shows the derivative of the resistivity of a crystal that was cooled to pumped liquid-helium temperature and then warmed. The three charge-density-wave transitions are marked with their usual α_i notation. Figure 2 shows the magnetoresistance of a crystal with the magnetic field applied parallel to the basal plane. We did not determine the orientation within the plane. The inset of fig. 3 shows the superconducting transition by measuring the resistance of the crystal. The curve of the superconducting critical field uses temperatures taken as the onset of the transition, and thus ignores the "foot" at low temperatures.

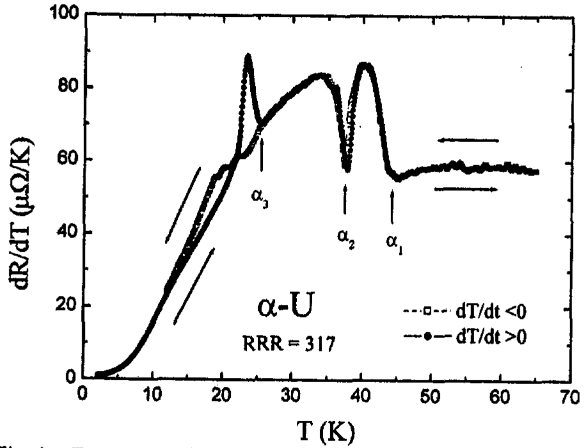

Fig. 1. Temperature derivative of the low-temperature resistivity of single-crystal uranium taken cooling and heating.

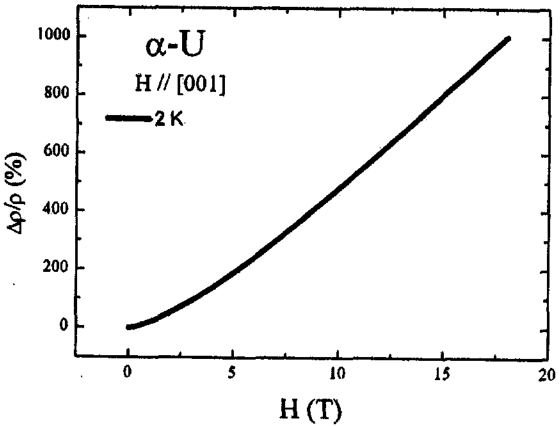

Fig. 2. The magnetoresistance of uranium at a temperature of 2 K.

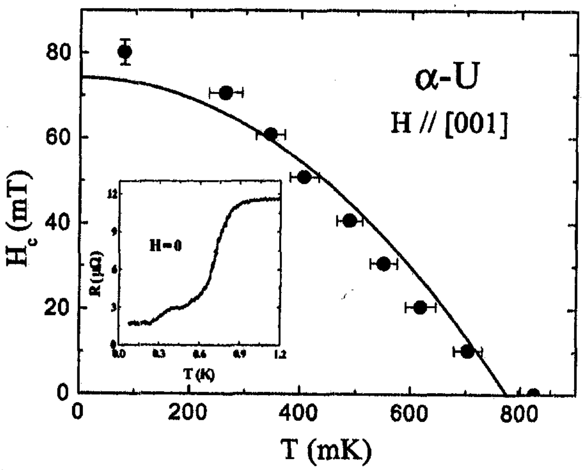

Fig. 3. The superconducting critical field of uranium where the temperature is taken as the onset of the transition. The inset shows that the transition in not uniform, which is assumed to be caused by salt inclusions. The curve is the usual parabolic fit.

4. Discussion and Conclusions

The derivative of the resistivity in Fig. 1 is the most precise measurement seen yet that addresses the hysteresis of the charge-density-wave transitions. The two higher temperature transitions are incommensurate with the lattice, while the lowest temperature one is commensurate. It thus can be viewed as a crystal structure change, and it is the only one that shows hysteresis. This is the first such measurement. As was seen in Lashley *et al.*,[9] these first-order transitions must be very subtle in fact.

The magnetoresistance of uranium has not been measured to high fields before. At low fields, it begins as quadratic, as expected for a simple metal. As it switches over to a more linear behavior it is more interesting, and we have no explanation for this yet.

The inset of fig. 3 shows an inhomogeneity in the sample because it has two transitions. There must be a salt inclusion that stresses a part of the sample. Nonetheless, the resulting critical field looks like a textbook critical-field curve, which has a parabolic temperature dependence. It is plotted using the onset temperature of the transition, but any criterion gives the same shape.

There are many interesting questions, which are discussed in most of the references, that we can address with single crystals of uranium and with higher purity uranium for making compounds. We believe that the preliminary work reported here holds the promise of answers to these questions.

Acknowledgments

Work at Los Alamos National Laboratory and Argonne National Laboratory was performed under the auspices of the U.S. Dept. of Energy. Measurements at the National High Magnetic Field Laboratory were supported by the National Science Foundation.

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