Laboratory Directed Research and Development – Los Alamos National Laboratory

Probing Correlated Electron Behavior in Uranium-235

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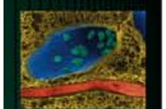


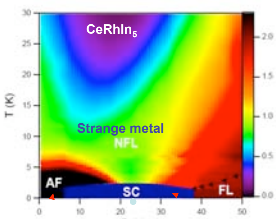
Innovation for Our Nation

Why is this research exciting?

If we can control the properties of materials, we may be able to solve many of the problems in the world today, like the looming energy crisis. Materials of the 21st century required to accomplish this task will be ones that we can "tune" to do a certain thing (such as carry electricity) easily and cheaply.

A class of materials containing rare-earth or actinide elements, called strongly correlated electron materials, shows promise for this necessary tunability and control. In these compounds, one set of mobile "conduction" electrons (the ones that carry electricity in a metal) talks, or interacts, with another set of electrons from the rare earth or actinide atom that are magnetic and located close to the nucleus.

The interaction of these two sets of electrons in the material leads to interesting and complex behavior such as superconductivity, magnetism, or the coexistence of the two behaviors at the same time, which may lead to future energy technologies. This complex behavior is easily tuned by such conditions as temperature, pressure, or a magnetic field. Just as we now really understand simple materials such as silicon, which has lead to incredible advances in electronics and computers, we need to understand these more complicated materials at the same deep level to be able to control them.



Temperature-pressure (T-P) phase diagram of the antiferromagnet $CeRhIn_5$ showing the coexistence of magnetism and superconductivity and, anunusual "strange metallic" state under pressure

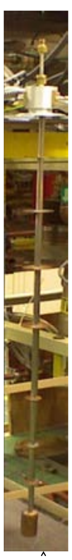
Magnet

Superconductor

T. Park et al. Nature (2008)

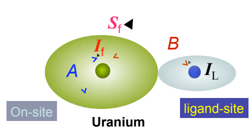
One way to understand the behavior of these complex materials is to look at them from the "inside out", at the microscopic level. We can use nuclear magnetic resonance (NMR) to "see" the nuclei themselves and how they interact with their environment. Since we are most interested in the magnetic atoms, we will use NMR to measure these nuclei, in particular an isotope of uranium, U-235

What is the approach?



Direct NMR of an actinide or lanthanide nucleus is generally quite difficult due to the strong "on-site" coupling (*A*) between the U nucleus and the surrounding magnetic (*5f*) electrons, leading to nuclear relaxation rates that are typically too fast (10^{-9} to 10^{-6} sec) for NMR detection (> 10^{-6} sec). In an ordered state (superconducting or magnetic) the processes (usually the tiny precession or "jiggling" of the *5f* electron spins) that relax the nuclei are suppressed, and direct detection may be possible.

"On-site" interaction of uranium nuclear spin (I_f) with magnetic 5f electrons with spin (S_f) located close to the nucleus. The uranium atom may also be influenced by a neighboring atom (ligand).

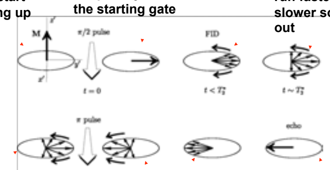


A bit of quantum mechanics happens in order to find the U-235 "signal"—a runner's analogy

1. Spins (runners) start out pointing up

2. Apply torque to put them in the plane just like runners at the starting gate

3. Some runners will run faster, some slower so they spread out



4. Now "flip" them and make them run in the opposite direction

5. Slower runners will have less distance to travel at a slower pace, while faster runners will cover a longer distance more quickly

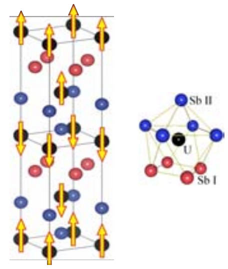
6. Both slow and fast runners arrive back at the starting line at the same time (to produce the "spin echo" signal

Study "cool" materials with a measurement probe (left) and a cryostat to get to a temperature of -270 C

What have we learned so far?

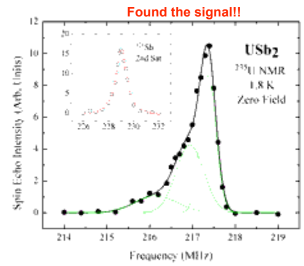
Find the right system: USb_2

Orders magnetically close to room temperature (T_N = 203 K to be precise) so that at very low temperature (2 K) the U-235 nuclei will have slowed down enough (hopefully!) to see a signal

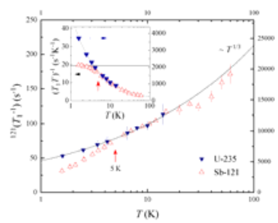


Crystal structure of USb_2. Black: U-235 atoms, blue and red: two Sb atoms in different positions in the structure. The arrows represent the spins of the U atoms in an up-down-up-down (antiferromagnetic) arrangement

Found the signal!!



U-235 signal vs frequency in USb_2 at T=1.8 K. The frequency of 217 MHz corresponds to an "internal" magnetic field produced by the antiferromagnetic arrangement of U spins of 277 Tesla—larger than any magnetic field that can be generated in the laboratory!



Spin-lattice relaxation rate $1/T_1$ in USb_2 of both the Sb-121 and U-235 nuclei

Nuclear spin-lattice relaxation rate: $1/T_1$ ("time" for "runners" to relax back to their un-torqued position)

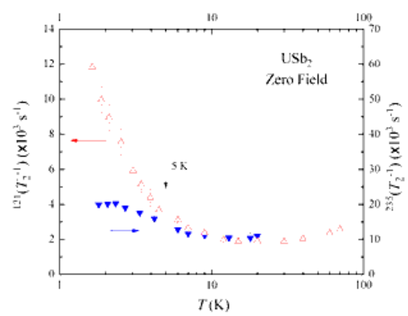
- Similar T-dependence for both nuclei above 5 K.

$1/T_1 \sim T^{1/3}$

- For ^{121}Sb, abrupt change occurs below ~ 5K, which coincides with other measurements that reveal **an energy gap in the magnon spectrum (like a hurdle the "runners" have to jump over)**.

Why different temperature dependences of the two nuclei?

On-site ^{235}U nuclei are relaxed by magnons (fluctuations of magnetic structure) but for ^{121}Sb (ligand nuclei) the scattering by conduction electrons dominates below temperature of 5 K.



Spin-spin relaxation rate $1/T_2$ in USb_2 of both the Sb-121 and U-235 nuclei showing the "time" taken to return to the starting line, due to relaxation by "jiggling" or precession of the uranium spins, or magnons.

Nuclear spin-spin relaxation rate: $1/T_2$ ("time" taken to reach starting line again after "runners" reverse direction)

- Similar T-dependence for both nuclei above 5 K.

- Below 5 K, the two nuclei reveal different temperature dependence probably due to how the different nuclei are affected by the magnons, or jiggling of the uranium spins.

We successfully measured nuclear relaxation rates $1/T_1$ and $1/T_2$ of U-235, for the first time, in a strongly correlated electron uranium compound

Why is this important for our nation?

From these direct U-235 nuclear magnetic resonance measurements on uranium compounds, we are just beginning to understand the complex behavior of strongly correlated electron materials at the microscopic level. We hope that future work on these interesting materials leads to the fundamental understanding of why they show such unusual and complicated behavior, which may lead to future energy technologies and applications.

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