NMR of CeCoIn5

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8/15/2016

In Co-NMR measurements on CeCoIn5, we see an increasing peak width below 50K. We interpret this as the growth of antiferromagnetic regions surrounding Cadmium dopants in an otherwise uniformly paramagnetic lattice. The Cadmium acts like a local negative pressure and drives the material toward a quantum critical point. These areas of negative pressure expand at low temperatures. NMR reveals the local nature of the Cadmium–induced magnetic order.

Introduction:

CeCoIn5 is a very interesting material with unique properties. It is a superconductor, a heavy fermion, and a quantum critical metal, so it has been the subject of significant research. A heavy fermion is a metal in which its 4f or 5f electrons can act like they are much heavier than they are and behave like a heavy fluid. A quantum critical metal is a material that at zero temperature, its ground state, undergoes a magnetic phase transition. Because this is not due to a changing temperature, this is due to a quantum change. CeCoIn5's ground states are paramagnetic and antiferromagnetic, and its magnetic phase transition can be induced through Cadmium doping.

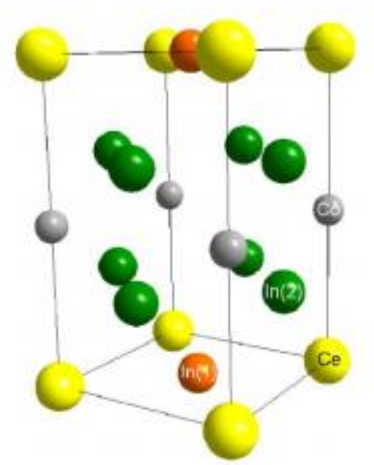


Figure 1: Unit cell of CeCoIn5: When doped with Cd, the Cd atoms typically replace the In atoms in the unit cell. These atoms apply less chemical pressure on the surrounding lattice than the In atoms, causing the surrounding lattice to feel less pressure.

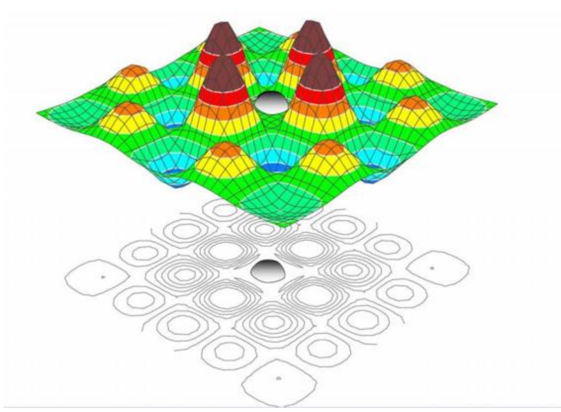


Figure 2: A histogram with distance represented on the horizontal plane and magnetization represented by the plot's depth. If a cadmium atom (the grey sphere in the middle) is placed into an otherwise uniformly magnetized CeCoIn5, a pattern of antiferromagnetic magnetization forms around it. As temperature gets lowered these areas of magnetization surrounding the cadmium atoms will expand.

Cadmium doping in CeCoIn5 reduces the chemical pressure. The cadmium atoms take the place of Indium atoms in the unit cell, and the chemical pressure they apply to the lattice structure is less than that of the indium atoms (Fig. 1). Therefore, where there is a Cadmium atom, the material "feels like" it is under less pressure. Because this material is already quantum critical, these areas of low pressure cause an area of antiferromagnetic magnetization to form (Fig. 2). Because of the material's magnetic phase properties (Fig. 3), these areas of magnetization expand as temperature is lowered because smaller changes in pressure are needed to nudge the material toward antiferromagnetism.

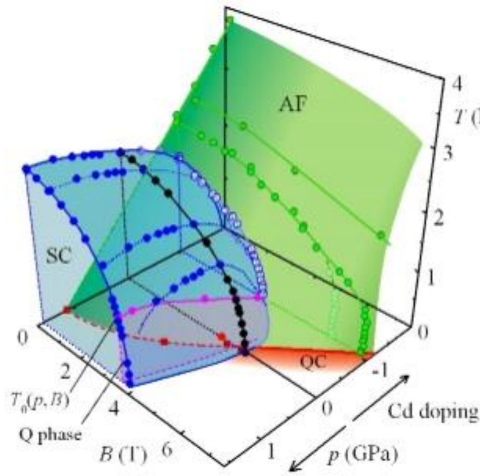


Figure 3. Phase diagram of CeCoIn5: At ambient pressure, material is quantum critical. At negative pressure, especially as temperature decreases, material trends from paramagnetic toward antiferromagnetic.

Nuclear spins inside the material inform of these patterns of magnetization. In this experiment, nuclear magnetic resonance (NMR) was used to observe the changes of nuclear spins in Cadmium-doped (=7.5%) CeCoIn5 as the magnetization was manipulated by the temperature. The sample was placed such that its c-axis was parallel with an external magnetic field of 11.7 T.

NMR:

The magnetic moments of nuclei generally align with an external magnetic field. The frequency at which the magnetic moment of a nucleus precesses around the field is called its Larmor frequency, and this frequency reflects both a nucleus's intrinsic qualities and its electronic environment. Nuclear magnetic resonance (NMR) can measure this frequency of precession of multiple different nuclei within one material, and these readings shed light on multiple types of electronic environments.

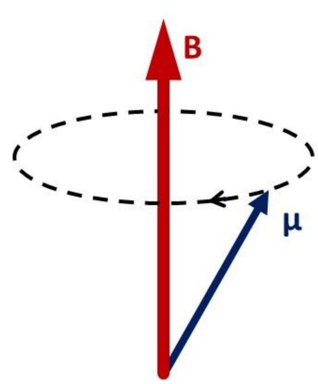


Figure 4: The magnetic moments of nuclei, when knocked off axis of an external field, will precess around the field at the nucleus's Larmor frequency.

To measure NMR, the material being studied is placed inside of a radiofrequency (RF) coil. This coil is the inductor in an LC circuit with an adjustable characteristic frequency ω:

$$\omega = \frac{1}{\sqrt{LC}}$$

Current is pulsed through the circuit at different frequencies (one can also hold frequency constant and sweep the field). These current pulses create a time-dependent magnetic field within the coil perpendicular to the much larger external field. When the frequency of the current matches the Larmor frequency of the nucleus, resonance occurs and the magnetic moments of the nuclei will much more fully get knocked over by the perpendicular field pulse. When this occurs, the RF coil can detect a much larger signal through induction than when the frequencies are not equal. However, the signal-to-noise ratio is still relatively low, so many of these signals are averaged at each frequency.

Current is pulsed through the circuit in the Hahn echo sequence (Fig. 5). There is first a current pulse that knocks the nuclear moments in the material perpendicular to the original field. Instead of immediately testing for resonance, the spins are briefly allowed to diphase. Dephasing occurs because the magnetic field is not perfectly uniform, so the Larmor frequencies of the nuclei differ ever so slightly. Therefore, as time passes, the nuclei get more and more out of sync. After the dephasing, another field pulse is applied that knocks the spins π radians. There, because the dephased nuclear moments were flipped 180 degrees, they converge for the same reason they dephased. Only then is the signal measured. There are two reasons for applying the second current pulse: first, it is difficult for the electronics and the inductor to, in a matter of microseconds, switch from applying a large voltage to detecting a small one, and second, the signal can be measured as the spins approach and descend from being perfectly in phase, instead of just measuring the peak and subsequent dephasing.

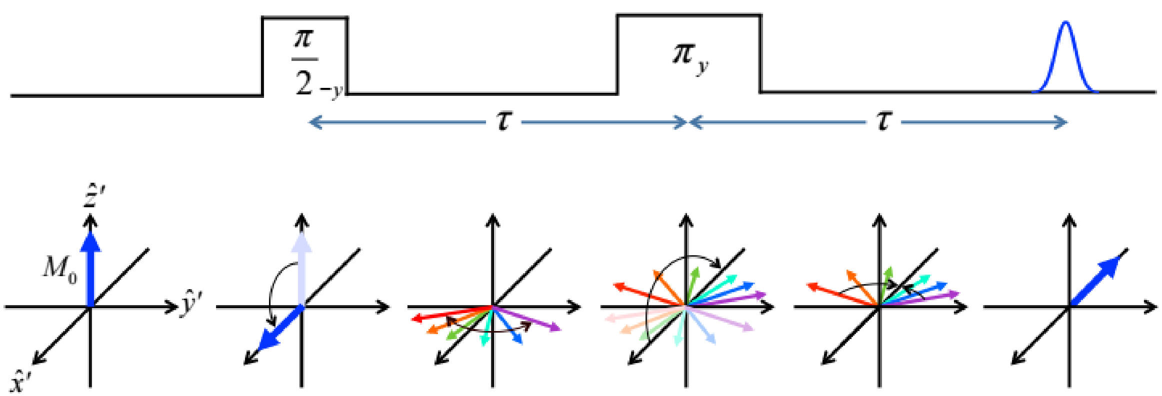


Figure 5: The Hahn echo sequence: First, the spins are aligned with the external magnetic field. Second, current is pulsed through the RF coil long enough to flip the spins □/2 radians. Third, the spins relax around the external field. Fourth, current is pulsed through the RF coil long enough to flip the spins □ radians. Fifth, the spins, which are still relaxing at the same speed, converge. Sixth, when the spins finally converge, the signal is measured. The frequencies at which significant signal is detected tell us about the electronic environment the nucleus is in.

The nuclear spin of a specific nucleus in a material can be characterized by its Hamiltonian:

$$H = H_z + H_q + H_{dip} + H_{hyp}$$

where $H_z$is the Zeeman interaction, H_q is the quadrupolar interaction, H_{dip} is the dipole interaction, and H_{hyp} is the hyperfine interaction.

The hyperfine interaction is the effect the spins of neighboring electrons have on the spin of the nucleus. This interaction is very important to understand for CeCoIn5, because the material undergoes a magnetic phase transition, and its electron spins will change. The dipole interaction is how the spins of neighboring nuclei affect the spin of the measured nucleus, and the quadrupolar interaction is how the charges of the surrounding electrons interact with the spin of a nucleus with spin greater than ½. The quadrupolar interaction is interesting for us because both Cobalt and Indium have spins greater than ½.

These three terms are important for interpreting data, but the Zeeman interaction is by far the largest contributor to the total nuclear spin Hamiltonian. In fact, in a liquid, the other three terms can effectively be ignored because nuclei in liquids interact much less with their neighbors than they do in solids. The Zeeman interaction is the effect an external magnetic field has on the nuclear spin:

$$H_z = \gamma \hbar H_0 \cdot I$$

where H_z is the applied external magnetic field, I is the spin of the isotope, and γ is the ratio of the magnetic dipole moment to the angular momentum, or the gyromagnetic ratio. The gyromagnetic ratio is unique to each isotope and gives the isotope's Larmor frequency. This frequency gives the frequency at which the magnetic moment of a nucleus will precess around a magnetic field:

$$\omega_L = \gamma H$$

where H is the external magnetic field. A Larmor frequency can be predicted based solely on the applied external magnetic field, and when the observed Larmor frequency differs from this prediction, it is due to the influence of the other three interactions. The difference, in percentage, between the predicted and measured Larmor frequencies is called the Knight Shift (K).

Results and Discussion:

We first performed frequency sweeps of the Cobalt site in Cadmium doped (= 7.5%) CeCoIn5 at varying temperatures with a constant field of 11.7 T. The sample was oriented so that the c-axis was parallel to the 11.7 field. This orientation was constant throughout the experiment as of the time this paper was written.

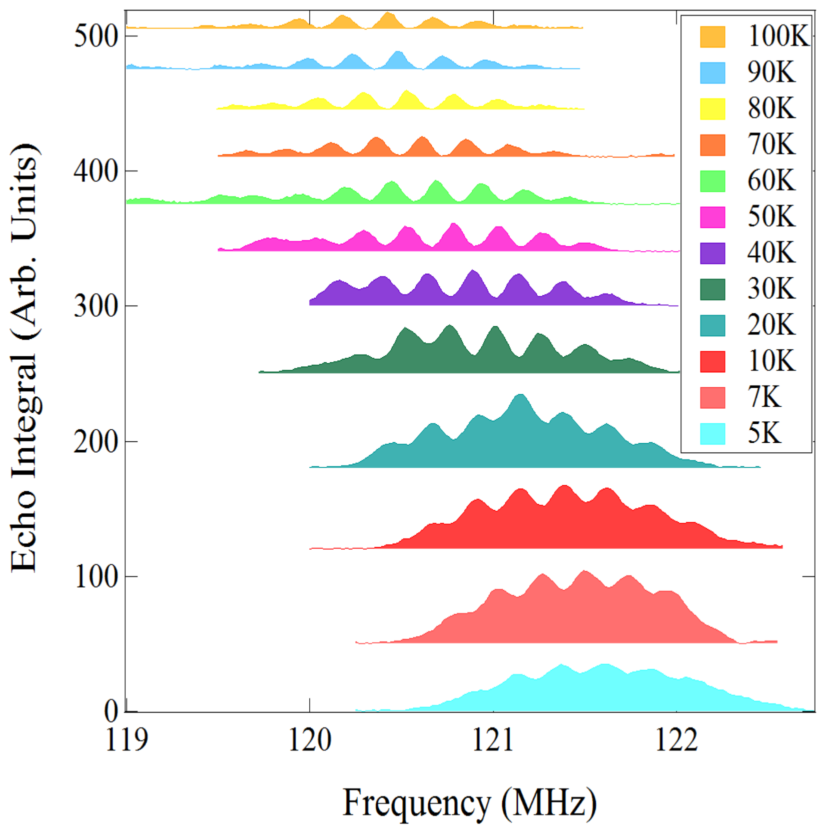
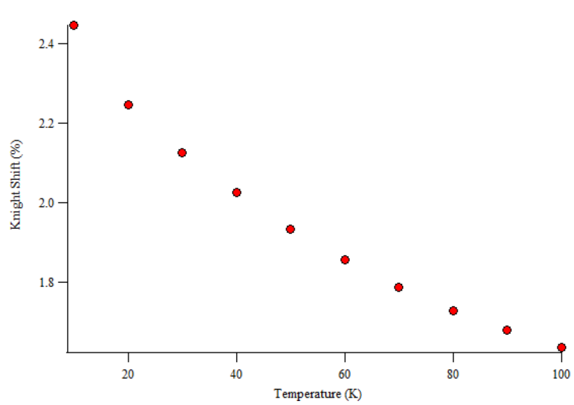


Figure 6: Frequency sweeps of Co site in CeCoIn5. Scans are offset for clarity. Temperatures increase vertically.

As the temperature decreased, the Cobalt site's Knight shift increased. Also, at low temperatures, the peaks broadened. This trend is due to the increasing hyperfine interaction, which indicates an increasing coupling between the Co nuclei spins and surrounding electron spins. The quadrupolar interaction can be seen in the seven frequency peaks at each site. These frequencies are correlated with the seven transition energies of a spin 7/2 particle.



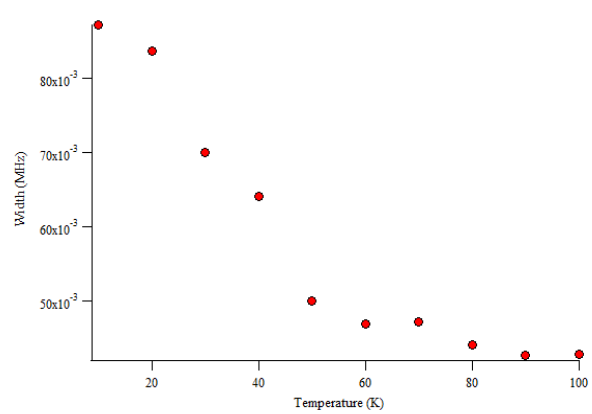


Figure 7: Knight shift of Co site as a function of temperature. The Knight shift increased as the temperature decreased.

Figure 8: Average width of Co frequency peaks as a function of temperature. Below about 50K, the peak broadens as the temperature decreases.

The increase of the width of the resonant peaks is particularly interesting. This is likely the direct result of the areas of antiferromagnetic magnetization surrounding the Cadmium atoms expanding. As the temperature decreased and the material neared its quantum critical point, the areas of low pressure surrounding the cadmium atoms induced larger areas of antiferromagnetic magnetization (Fig. 2). As more nuclei are affected by these changing magnetic environments, more of their magnetic resonant frequencies will change.

So, at high temperatures, when the material is almost uniformly paramagnetic with only small areas of antiferromagnetic magnetization surrounding the cadmium atoms, one expects the resonant frequency peaks to be sharp and precise. As the temperature decreases and the areas of magnetization expand, there will be a greater number of nuclei that will have different Larmor frequencies, so one would expect that the peaks will broaden and be less clear. This change is visible in Figure 8.

Second, the Indium (1) site was observed.

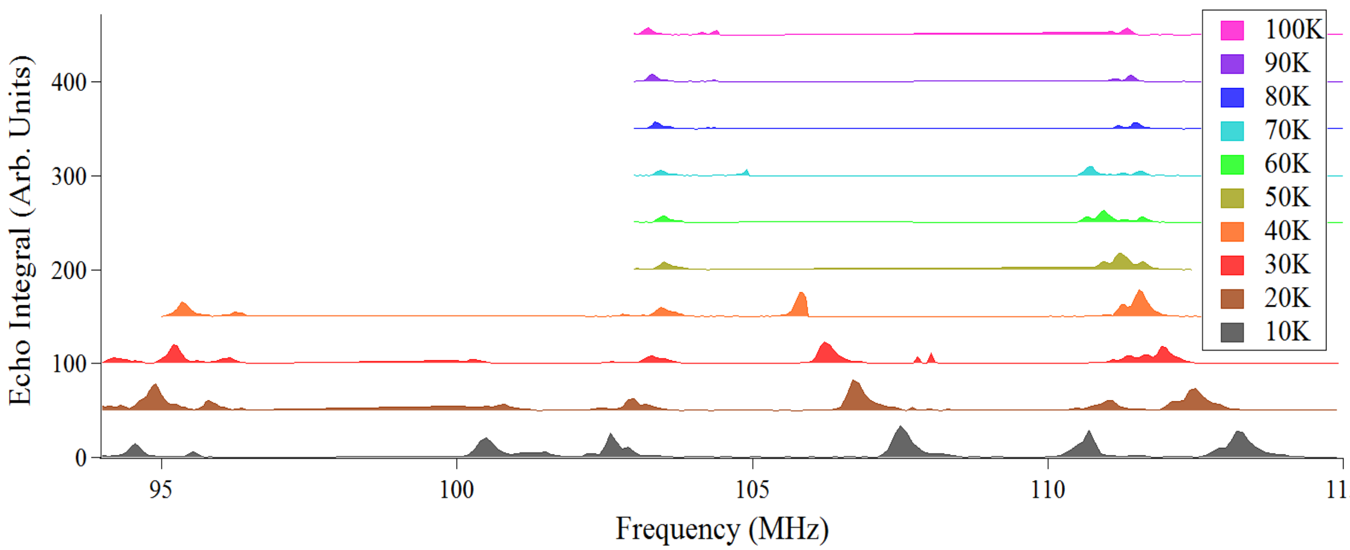


Figure 9: Frequency sweeps of the In(1) sites at varying temperatures. Scans are offset for clarity. At the time of writing, scans were still in progress, so the data are incomplete. Resonant peaks at 10K are at ≈ 94 MHz, 103 MHz, and 11 MHz. As temperature increases, the frequencies of these peaks increase until about 50K, and then start decreasing slightly.

At the time of the writing of this paper, these scans were incomplete. Qualitatively, one can observe the movement of the In(1) peaks (Fig 9). Of particular interest are the low temperature scans showing features of the individual peaks. One can see, for example, in the 103 MHz peak at 10K, that there are side peaks along with the main peak. These peaks could represent location in relation to the Cadmium atoms, and the corresponding breakdown of the lattice surrounding the Cadmium atoms.

Future Work:

More data should be taken from this material, and the analysis of the new data along with the data collected by the time of writing should prove interesting.

The data show signs of the effects the cadmium atoms have on the Cobalt and Indium (1) atoms. There are also data available of CeCoIn5 with different levels of Cd doping (=10%, 15%). If all these data were combined with simulations, a clear and informative picture could be formed of the effects Cadmium doping has in a quantum critical material.

Also, the Indium (2) sites could be analyzed, and angular dependence of the material could be tested.

References

Curro, N.J. (2011). Quadrupolar NMR of Superconductors. *Encyclopedia of Magnetic Resonance.*

Curro, N.J. (2009). Nuclear magnetic resonance in the heavy fermion superconductors. *Rep. Prog. Phys. 72*(026502), p. 24.

Curro, N.J. (2016). Nuclear magnetic resonance in Kondo lattice systems. *Rep. Prog. Phys. 79*(064501), p. 12.

Curro, N.J., Young, B., Urbano, R.R. et al. Hyperfine Fields and Magnetic Structure in the B Phase of CeCoIn5. *J Low Temp Phys* (2010) 158: 635. doi:10.1007/s10909-009-9967-y

Estry, A. (2015). Nuclear magnetic resonance study of three dimensional dirac semimetal Na3Bi. http://london.ucdavis.edu/~reu/REU15/Papers/estry.pdf

S. Zaum, K. Grube, R. Schäfer, E. D. Bauer, J. D. Thompson, & H. v. Löhneysen.(2011). Towards the Identification of a Quantum Critical Line in the (p, B) Phase Diagram of CeCoIn5 with Thermal Expansion Measurements. *Phys. Rev. Lett., 106*(087003).