

# Magnetic Structure of $Ce(Mn_{1-x}Cu_x)_2Si_2$

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## Abstract

In recent temperature dependent magnetization measurements on CeCu<sub>2</sub>Si<sub>2</sub>-based compounds, we found that the magnetization as a function of temperature does not follow the inverse law which usually obeyed by cerium (Ce) ions. In this project, we build a matrix model to calculate the multiple energy levels of Cerium atoms. Then we apply the Boltzmann function to calculate the magnetization as a function of temperature. Mathematical approximation will be applied to this calculation for the limit cases of low temperature and high temperature ends. Finally, the theoretically calculated magnetization curve will be compared with the experimental curves.

## 1 Purpose

The purpose of this paper is to explore what parametric function of our data made our experimental graph curve instead of going in a straight line. This could help make a model that could to calculate any substances magnetic susceptibility more accurately when theoretically done. This will also help us to understand more about the substance that we are currently working on.

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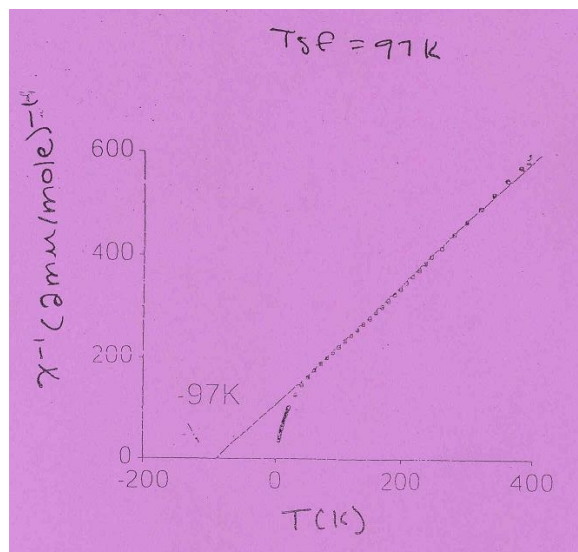


Figure 1: Experimental curve vs Theoretical curve

## 2 Background and Introductory Material

In order to understand what the paper will be talking about, first we need to have some background. A superconductor is an element, inter-metallic alloy, or compound that will conduct electricity without resistance below a certain temperature. Resistance is undesirable because it produces losses in the energy flowing through a material, a microscopic quantum phenomenon. We want a substance that will allow electricity to take the least path of resistance and decrease the heat that is produced. The next question may be; where can we use superconductors? One use is Magnetic-levitation. Transport vehicles such as trains can be made to float on strong superconducting magnets. Another area is in the field is biomagnetism. By shooting a strong superconductor-derived magnetic field into the body, hydrogen atoms that exist in the body's water and fat molecules are forced to accept energy from the magnetic field. They then release this energy at a frequency that can be detected and displayed graphically by a computer, an MRI. Also electric generators made with superconducting wires are more efficient than conventional generators.

### 3 The Experiment

(insert slide 3) These samples were prepared by melting the constituent elements in an atmosphere of argon gas, to reduce oxidation of the metals, by standard arc furnace. The samples were then grounded to a small powder and then examined by the use of x-ray diffraction (XRD). A measurement was then made for the temperature dependent magnetization on the samples for  $x = 0$  using Faraday magnetometer in a magnetic field. For  $x$  not equal to zero, the temperature dependent magnetization was measured by using the physical properties measurement system for quantum design. This brought up a question. Why does the Magnetic Susceptibility,  $\chi(T)$ , for  $CeCu_{(2)}Si_{(2)}$  as a function of temperature have the following behavior:

- For  $T > 50K$ , the inverse of  $\chi(T)$ ,  $\chi^{-1}$  varies almost linearly with temperature
- For  $T < 50K$ ,  $\chi^{-1}$  drops with the decrease of temperature much quicker than the linear behavior

### 4 Road Map

Our goal is to be able to parameterize our data for the magnetic susceptibility as a function of temperature. We first have to solve the eigen equation. This will give us the magnetic moment of an atom and the magnetic energy of the atom when a magnetic field is applied. We then have to solve for the Boltzman distribution to give us the probability that an atom will be in a certain state. After that we can solve for the Brillouin function. This has to be done when one tries to calculate the magnetization of compounds with magnetic moments. This will let us to calculate the susceptibility in respect to temperature. In order to do this we have to also calculate the curie law which will give us the curie constant. We then need to know that state that our Ce atom can exist. We need to know the fractional occupation of the state and the excitation energy. Then we find the inverse chi. Inverse chi is the magnetic susceptibility. Then we fit the inverse chi in respect to high temperature.

## 5 Finding the Magnetic Moment of Ce Ion

We need to solve the eigen equation to obtain the eigen function. This will give us the magnetic moment of the orbital angular momentum of the Ce ions. We first start need to start with the Hamiltonian equation.

$$\hat{H} = T + \mu$$

The Hamiltonian is a function that describes the state of a mechanical system in terms of position and momentum.

$$\hat{H} = \frac{p^2}{2m} - k^* \left(\frac{e}{r}\right)^2$$

We then operate each term on the wave function to normalize the function.

$$\psi = \psi(x, y, z, t)$$

$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}$$

We then need to solve the Schrödinger equation to obtain the eigen function.

$$\hat{H}\Psi = -\frac{\hbar^2 2m^2}{v} \Psi - \frac{ke^2 r}{\Psi}$$

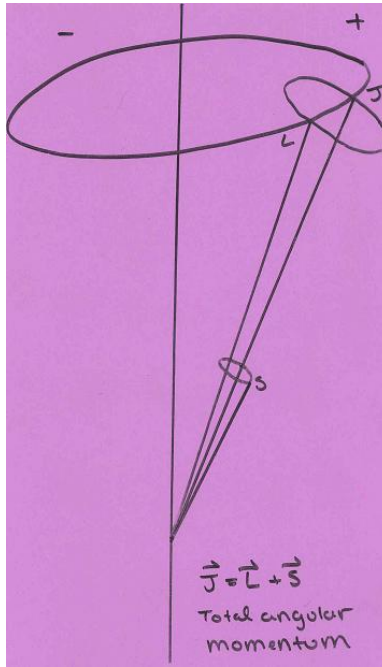
Putting it into spherical coordinates we get:

$$\begin{aligned} -\frac{\hbar^2}{2m} \frac{1}{r^2} \frac{\partial^2}{\partial r^2} \left( r \frac{\partial^2}{\partial z^2} + \frac{1}{r^2} \sin(\theta) \frac{\partial^2}{\partial \theta^2} \left( \sin \theta \frac{\partial^2}{\partial \theta^2} + \frac{1}{r^2} \sin^2(\theta) \frac{\partial^2}{\partial \theta^2} \left( \sin \theta \frac{\partial^2}{\partial \phi^2} \right) \right) \right) \Psi - \frac{ke^2}{r} \Psi \\ = E\Psi \end{aligned}$$

$$\Psi(r, \theta, \phi) = R(r)\Theta(\theta)\Phi(\phi)$$

$$\frac{\partial^2}{\partial \phi^2} + m^2 \phi = 0$$

We are only interested in the  $\phi$  portion,  $\Phi(\phi)^2 e^{im\phi}$ .



Then we need to use spin orbital coupling to find the total angular momentum and magnetic momentum. One has to take in account for the fact that an atom has two angular momentums, orbital and angular spin. To help understand, the earth spins on it's axis, angular spin, and it rotates around the sun, orbital spin. If an atom is in free space so that no external torque acts on it, its total angular momentum is :  $J = L + S$ .

$$J = L - S, L - S + 1, \dots, (L + S - 1), (L + S)$$

For each projection of  $J$  we get certain  $M$  values.

$$M = -J, -J + 1, \dots, (J - 1), J$$

$J$  is the fixed magnitude of thez component, the total angular momentum.  $L$  is the orbital momentum and  $S$  is the angular momentum. The magnetic moment of an atom is in a specific state. This is characterized by a pair of  $J$  and  $M$ 's.

$$\vec{\mu} = -g_J \mu_b m$$

$g_J$  is call the 'g' factor. It is a conversion factor and there is a certain 'g' factor for every state. For example, and electron has a 'g' factor of  $\frac{1}{2}$ .  $\mu_b$  is the Bohr magnetization. This is another scaling factor for an atomic magnetic moment.

$$\vec{\mu}(z) = -g_J \mu_b m$$

$\vec{\mu}(z)$  is the  $z$  component of the magnetic moment.

$$g_J = \frac{1 + J(J + 1) + S(S + 1) - L(L + 1)}{2J(J + 1)}$$

This is a degenerate state ion. This means that for the same  $J$  values we get different values for  $M$ . In this compound we see 6 specific states by  $J$  and  $M$  because we have 3  $L$ 's values and for each  $L$  we have 2  $S$ 's. If a magnetic field is applied to a metal, a preferred orientation splits the state and isotropy is ruined. For our lowest energy level:

$$L = 3, S = \frac{1}{2} \quad J = L - S = \frac{5}{2}$$

$$M = -\frac{5}{2}, -\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}, \frac{3}{2}, \frac{5}{2}$$

With out a magnetic field applied to  $\hat{H}$  the E and atomic states have the same energy. The different energies are given by:

$$E_m = -\mu_0 \mu \hat{H} = -\mu \beta$$

,  $E_m$  is the potential energy.

$$J\hat{H} = J\hat{H}(z) = \hat{H}J(z) = \hat{H}m$$

The higher the field, the larger the spread will be. This will make us worried about overlapping of the states.

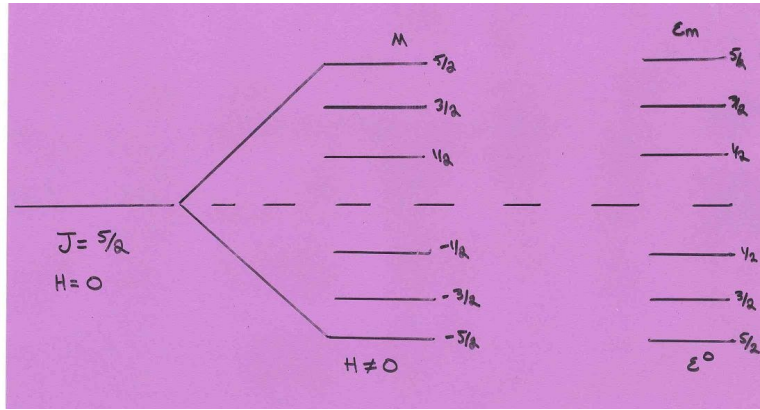
$$E(m) = mg_J \mu_0 \mu_b \hat{H}$$

$$\text{With } m = -J, -J + 1, \dots, (J - 1), J$$

Or

$$E(m) = mE^0$$

$$E^0 = g_J \mu_0 \mu_b \hat{H} = \text{constant}$$



$E^0$  is the first order correction. We then assume that there are  $N$  Ce atoms per unit volume in the material. Then by the Boltzmann distribution according to the energy, the probability,  $P_z$ , of finding an atom in a specific state is given by:

$$P(m) = \frac{e^{\frac{E_m}{k_\beta T}}}{z} = \frac{e^{\frac{E_m}{k_\beta T}}}{\sum_{m=-J}^J e^{\frac{E_m}{k_\beta T}}}$$

$$F = \frac{E_m}{k_\beta T}$$

## 6 Magnetization of Material

Then we need to calculate the magnetization of the material.

$$M = N \cdot \frac{\sum_{m=-J}^J -g_J \cdot m \cdot \mu_b \cdot e^{\frac{-g_J \mu_0 m \mu_b \hat{H}}{k_\beta T}}}{\sum_{m=-J}^J e^{\frac{-g_J \mu_0 m \mu_b \hat{H}}{k_\beta T}}}$$

$$\text{Set } x = \frac{g_J \mu_b \mu_0 \hat{H}}{k_\beta T}$$

$$d \ln(x) = \chi^{-1} dx$$

$$de^{mx} = me^{mx} dx$$

$$\text{Then } M = Ng_J \mu_b \frac{d}{dx} \left( \ln \sum_{m=-J}^J e^{-mx} \right)$$

To take care of the sum we used the formula for a finite series for an even function.

$$\begin{aligned} \sum_{m=-J}^J e^{-mx} &= \sum_{m=-J}^J e^{+mx} \\ &= e^{Jm} + e^{-J-1} + \dots + e^{(J-1)x} + e^{Jx} \\ &= e^{-Jx} (1 + e^x + e^{2x} + \dots + e^{2Jx}) \\ &= e^{-Jx} \frac{e^{(2J+1)x} - 1}{e^x - 1} \end{aligned}$$

To simplify things, we let:  $S = 1 + e^x + e^{2x} + \dots + e^{2Jx}$

$$Se^x = e^x + e^{2x} + \dots + e^{(2J+1)x}$$

$$Se^x - S = e^{(2J+1)x} - 1$$

$$S(e^x - 1) = e^{(2J+1)x} - 1$$

$$S = \frac{e^{(2J+1)x} - 1}{e^x - 1}$$

Where  $S = \text{JoJimbo parameter}$

To calculate magnetization:

$$\begin{aligned}
M &= Ng_J\mu_b \frac{d}{dx} \left[ \ln \frac{e^{J+1} - e^{Jx}}{e^x - 1} \right] \\
&= Ng_J\mu_b \frac{d}{dx} \left[ \ln \frac{e^{J+1} - e^{-Jx}}{e^{\frac{x}{2}} - e^{-\frac{x}{2}}} \cdot \frac{1}{e^{\frac{x}{2}}} \right] \\
&= Ng_J\mu_b \frac{d}{dx} \left[ \ln \frac{e^{(J+1)x} - e^{(-J+\frac{1}{2})x}}{e^{\frac{1}{2}x} - e^{-\frac{1}{2}x}} \right] \\
\sinh &= \frac{e^x - e^{-x}}{2} \\
M &= Ng_J\mu_b \frac{d}{dx} \left[ \ln \frac{(\sinh(J + \frac{1}{2})x)}{(\sinh \frac{1}{2}x)} \right] \\
M &= ng_J\mu_b J \beta_J(y)
\end{aligned}$$

Where  $\beta_J(y)$  = Brillouin function. The Brillouin function is a special function that arises in the calculation of magnetization of compounds with a magnetic moment.

$$\begin{aligned}
\beta_J(y) &= \frac{2J+1}{2J} \coth \frac{2J+1}{2J} y - \frac{1}{2J} \coth \frac{y}{2J} \\
y &= Jx = Jg_J\mu_b\mu_0 \frac{H}{k_B T}
\end{aligned}$$

## 7 Calculating the Magnetic Susceptibility

The magnetic susceptibility is the degree of magnetization of material in response to the magnetic field.

$$\mu = \chi H$$

Where  $\mu$  = magnetization of material and  $\chi$  = dimensionless volume magnetic susceptibility. We assume:  $\mu_0 = 4\pi \times 10^{-7} \frac{t}{Am^{-1}}$  and  $\beta = \mu_0 H = .1$  Tesla; since  $\mu_b = 9.274 \times 10^{-24} Hm^2$ . At room Temperature (298K) for  $J = \frac{5}{2}$ ,  $L = 3$ ,  $S = \frac{1}{2}$ .

$$g_J = 1 + \frac{\frac{5}{2}(\frac{5}{2} + 1) + \frac{1}{2}(\frac{1}{2} + 1)}{2(\frac{5}{2})(\frac{5}{2} + 1)} = 3(3 + 1)$$

$$y = \frac{g_J T \mu_b \mu_0 H}{k_\beta T} = .00048$$

With  $.0048 \ll 1$  and  $\beta = 10$  Tesla.  $\beta$  is the magnetic field.  $Y = .048 \ll 1$  when  $t > 300k$  and  $y \ll 1$ . With  $\beta_J(y) = \frac{J+1}{3J}y + y^3$  and  $\coth y = \frac{1}{4} + \frac{1}{3}y + 0y^3$ , we get:

$$M = n g_J \mu_b J \beta_J(y)$$

$$= \frac{n \mu_0 g_J^2 J(J+1) \mu_b^2 H}{3 k_\beta T}$$

$$\chi = \frac{m}{H} = \frac{c}{T}$$

The Curie constant is when the susceptibility is temperature dependent and it's values are determined by relative strength of thermal energy and magnetic interaction energy. Then the Curie constant:

$$c = \frac{n \mu_0 g_J^2 J(J+1) \mu_b^2}{3 k_\beta}$$

The effective moment is  $u_{\text{eff}} = g_J \sqrt{J(J+1)} \mu_b$ .  $c = \frac{n \mu_b u_{\text{eff}}^2}{3 k_\beta}$  and  $\chi = \frac{c}{T}$ .

This curie constant is a relation for paramagnetic materials.  $T =$  ferromagnetic curie temperature For  $J = \frac{5}{2} g_J = \frac{6}{7} u_{\text{eff}} - 2.54 \mu_b$  We know that only 2 states can exist: E2 which is a  $4f^1$  orbital and E1 which is a  $4f^0$  orbital For a compound we know that here is a certain number of electrons that can fill it's orbital. For the  $4f^1$ , all of the shells are filled except the 'f' orbital. The  $4f^0$  orbital, all of the shells are filled but there is one electron in the 'f' orbital.

$$\chi = \frac{n \mu_0}{3 k_\beta} = \frac{u_1^2 v(t) + u_2^2 (1-v(t))}{T + t s f} \quad u_1 = g_{J_1} \sqrt{J_1(J_1 + 1)} \mu_b$$

$$u_2 = g_{J_2} \sqrt{J_2(J_2 + 1)} \mu_b$$

$v(t)$  is fractional occupation of state. Fractional occupation state means that if you have  $N$  occupation sites, the fractional occupation is how many of those sites will be occupied.  $E_{ex} = E_1 - E_2$  which is the excitation energy. The excitation state of a system is any quantum state of the system that has a higher energy than the ground state.

$$v(T) = \frac{6}{6 + e^{-E_{ex}(k_{\beta}(T+tsf))}}$$

$$\chi(t) = \frac{c}{T + tsf} v(T)$$

$$c = \frac{n\mu_0}{3k_{\beta}} (2.54\mu_b)^2$$

$$\chi^{-1}(T) = \frac{T + tsf}{c} v^{-1}(T)$$

If the graph was a straight line the slope would be  $\frac{t}{c}$  but here it is  $\frac{1}{c}$ . So the curie constant will be  $\frac{1}{\text{slope}}$  which is unchangeable.

$$\chi^{-1}(T) = \frac{T+tsf}{.82} \frac{6e^{\frac{-E_{ex}}{k_{\beta}(T+tsf)}}}{6}$$

$T_{sf}$  and  $E_{ex}$  are fitting parameters. We can use the inverse chi as a function of temperature at  $10K$  to get an initial value of  $E_{ex}$  for our fittings.

With our  $tsf = 40K$ ,  $t = 10K$ , and  $\chi^{-1}(T) = 63.3$ ; to solve for our  $E_{ex}$ :

$$\frac{-E_{ex}}{k_{\beta}50} = .229$$

$$E_{ex} = E_2 - E_1 = 74k_{\beta} = 6.4MeV$$

$$\chi^{-1}(T) = \frac{T + 40}{.82} \frac{6e^{\frac{-74}{k_{\beta}(T+40)}}}{6}$$

## 8 Conclusion

The reason for doing a theoretical calculations is to see whether or not a compound will work before starting an experiment. When doing the theoretical calculation we get a straight line but our data shows that our line isn't straight but curved. This tells us that our theoretical equation is not correct. This project is to see if we can get a better theoretical equation that will give us a line that will be closer the experimental.

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