Magnetization Measurements of Antiferromagnetic $SrCu_2(BO_3)_2$ using a VSM

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(July 28, 2004)

Abstract

$SrCu_2(BO_3)_2$ is a low-dimensional antiferromagnetic compound that has undergone many different magnetization studies; however, to date none have addressed the temperature dependence of the magnetization. Measuring this sample's magnetism versus magnetic field at varying temperatures will provide insight into its quantum mechanical nature and help strengthen or refute the current theory. This experiment utilizes a vibrating sample magnetometer and DC magnet at the National High Magnetic Field Laboratory and covers a magnetic field range up to 30.1 T and a temperature range from 1.4 to 12.3 K. Our data show that the magnetization of the sample does in fact depend the temperature of the sample.

I. Introduction

Physicists began studying magnetic properties of various materials many decades ago and continue today. The quantum mechanical nature of magnetism was identified around 1930, but the full understanding of the phenomenon has yet to be achieved, even today. The motivation to study low dimensional materials arises from the suspected connection between high temperature superconductors and antiferromagnetism. Some oxides, a copper oxide for instance, naively appears to be an insulator. However, depending on the amount of oxidation, the material can become either an insulating antiferromagnet or a superconductor when cooled to a sufficiently low temperature. Moreover, theorists believe that the antiferromagnetism may play some underlying role in the superconductivity, rather than the two being disjunctive phenomena.

$SrCu_2(BO_3)_2$ is a prime candidate for magnetometry measurements because it is either one of two, or *the only* example of a two-dimensional spin-gap system. Unlike other, classical antiferromagnetic materials, at zero temperature this material is not magnetic. Furthermore, it does not become magnetic until it is subjected to a certain amount of magnetic field. Measuring this sample's magnetism versus magnetic field will provide further insight into its quantum mechanical nature, as well as provide data for comparison to a theory not fully completed yet. The vibrating sample magnetometer (VSM) will be used to characterize the magnetism of $SrCu_2(BO_3)_2$ in varying magnetic fields at the National High Magnetic Field Laboratory in Tallahassee, Florida. Although earlier magnetization measurements have been performed on this sample, these were carried out using a pulse magnet and the temperature could not be controlled. The VSM technique is advantageous because it allows the measuring of magnetization at a constant temperature. The setup includes a powerful resistive magnet, a vibrating mechanism, detection coils, and a lock-in amplifier.

The scope of this experiment is limited in that it is designed to only measure the magnetization as a function of magnetic fields at different temperatures. However, the findings may be used to guide the understanding of other two-dimensional spin-gap systems found in the future; likewise, the findings may also influence the quantum mechanical theory for these types of materials, which has not been fully completed to date.

Magnetism

In Physics, materials can be classified by one of three types of magnetizations depending on their magnetic susceptibility, paramagnetism, diamagnetism, or ferromagnetism [1]. Paramagnetism, diamagnetism, and ferromagnetism are both very well understood in Physics and their engineering capabilities have been widely explored.

Antiferromagnetism, however, still provides some mysteries. The subject of low-dimensional antiferromagnets that strongly exhibit quantum mechanical behavior has only been around ten years and there is much competition among physicists to be the first to experimentally discover the behavior of these materials and either strengthen or refute the current theories.

The magnetization of a material can provide crucial information about the intrinsic properties of the materials as a whole, as well as the nature of the interactions of the constituent ions. In the case of quantum mechanical antiferromagnets, magnetization measurements are used to clarify the nature of spin-singlets that occur at the ground state [2]. Classically, at zero field, the magnetic dipole moments of an antiferromagnetic material line up antiparallel to each of its nearest neighbors. This arrangement is the lowest energy state; thus, this state is preferred. For special antiferromagnets, however, this is not the quantum mechanical ground state. $SrCu_2(BO_3)_2$ is one of these special antiferromagnetic materials. The structure of this compound is a two-dimensional system, made up of separate layers with weak coupling between each of the sheets and strong couplings between the copper ions within the same sheet (see Fig. 1 [3]). An interesting ground state occurs when the magnetization of the material is zero, but the applied magnetic field is not. Previous studies found that under very high magnetic fields, the magnetization curves have plateaus at one-fourth and one-eighth of the full magnetic moment of Cu^{2+} ions.

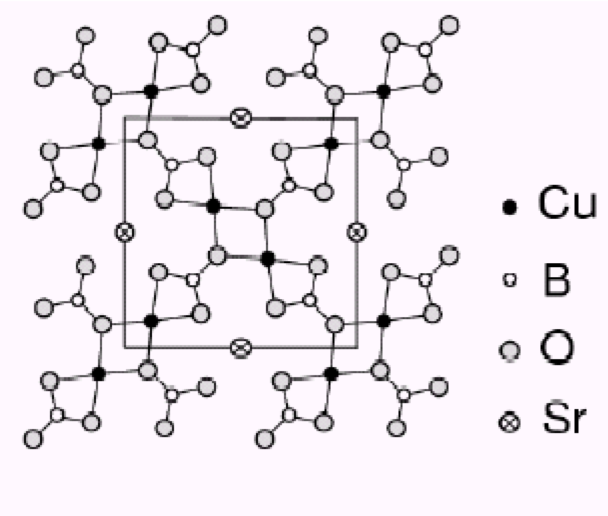

FIGURE 1. Schematic of $SrCu_2(BO_3)_2$.

On a previous run, Professor Takano and his research team found some new interesting features of the magnetization curves of this compound. The focus of my project was to attempt to reproduce these new anomalies and possibly discover something about their origin.

II. Experimentation

Resistive Magnet

The DC magnet utilized in this experiment is one housed at the National High Magnetic Field Laboratory at Florida State University. The facility is powered by a 40 million watt (MW) power supply and is highly regulated. This represents 10 % of Tallahassee's total generating capability. The complex has an extensive water-chilling plant that stores 1 million gallons of chilled water and is capable of producing 8,000 tons of ice per day [4]. The magnet itself is a DC bitter magnet and can reach 33 T.

Vibrating Sample Magnetometer

As before, the vibrating sample magnetometer was the instrument to be used to measure the magnetization of the $SrCu_2(BO_3)_2$, for fields ranging from 0 to 30.1 T and a temperature range of 1.4 to 12.3 K. During the first run, however, a significant amount of noise was present in the signal. Therefore, the first task was to become familiar with the nature of the vibrating sample magnetometer and seek out possible causes and remedies for the noise problem. Many papers were found offering various solutions including techniques to increase the sensitivity, decrease the temperature-dependent noise, and increase the signal to noise ratio (SNR) while decreasing the noise caused by electronics and modulation. The more promising solutions were forwarded to the VSM technician in Tallahassee for further consideration.

Methods

Once we were assured that the noise problem had been addressed, the second run was set to begin. Initially the sample is securely placed in the probe and lowered into a refrigeration system within the core of the resistive magnet. The sample is then cooled to a predetermined temperature using various cryogenics including liquid nitrogen, ^{4}He, and ^{3}He. Once the cooling process is completed the VSM is checked thoroughly to ensure that the pick-up coils are located in the middle of the magnet, and the sample is sitting in the middle of the coils. Next, a calibration is performed using a nickel standard for which the magnetization is well known. Once the centering and calibration is achieved the experiment can begin. The magnet is initialized and the sweeping of the field begins. The sample is vibrated between the detection coils and a voltage is generated due to the change in flux caused by the sample (see Fig. 2 [5]). The voltage is related as a signal and stored for further analysis. Using a computer program the magnetic field is changed from 0 to 30.1 T at a constant rate, and the temperature is recorded for each sweep. During the second run, we were able to collect data for temperatures ranging from 1.4 to 12.3 K for two sample orientations.

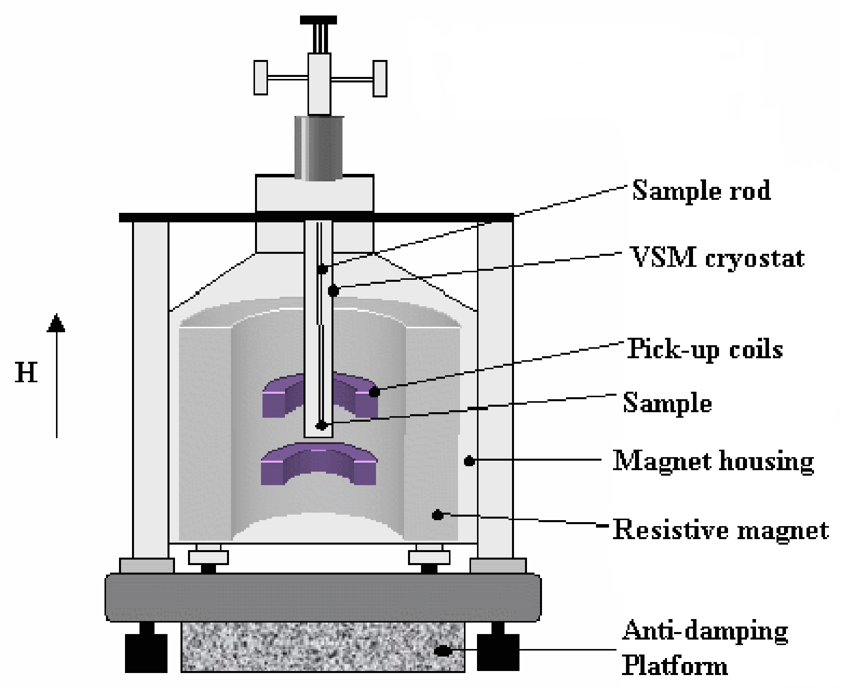

FIGURE 2. VSM schematic.

III. Results & Data Analysis

In 1999, Kageyama and his associates conducted a similar experiment for the temperature range of 1.7 to 400 K and using a pulse magnet, reached fields of 45 T. The rapidly changing magnetic field changes the magnetization isentropically rather than isothermally in an ideal case; that is, the experiment is performed without changing the entropy taking no account of the temperature changes [2]. In practice, however, there is a real possibility of the sample experiencing heating from the metallic components of the magnetometer that can be heated by the induced eddy currents. Despite this probability of heating of the sample, the data show the unique feature of $SrCu_2(BO_3)_2$, the nearly zero magnetization until the critical field, H_c given by Eq. (1), where Δ is the energy gap for the lowest excitations, k_B is the Boltzman constant, g is the g factor, and μ_B is the Bohr magnetron [1].

$$H_C \approx \frac{k_B \Delta}{g \mu_B} \tag{1}$$

At the lowest temperatures, our results agree with that of Kageyama's study, thereby reaffirming the existence of the energy gap of 30 K. However, unlike their experiment our data are designed to also detect any temperature dependence, which has not been previously investigated. Our data, pictured in Figs. 3 and 4 clearly show that with any given field, the magnetization clearly increases with increasing temperature.

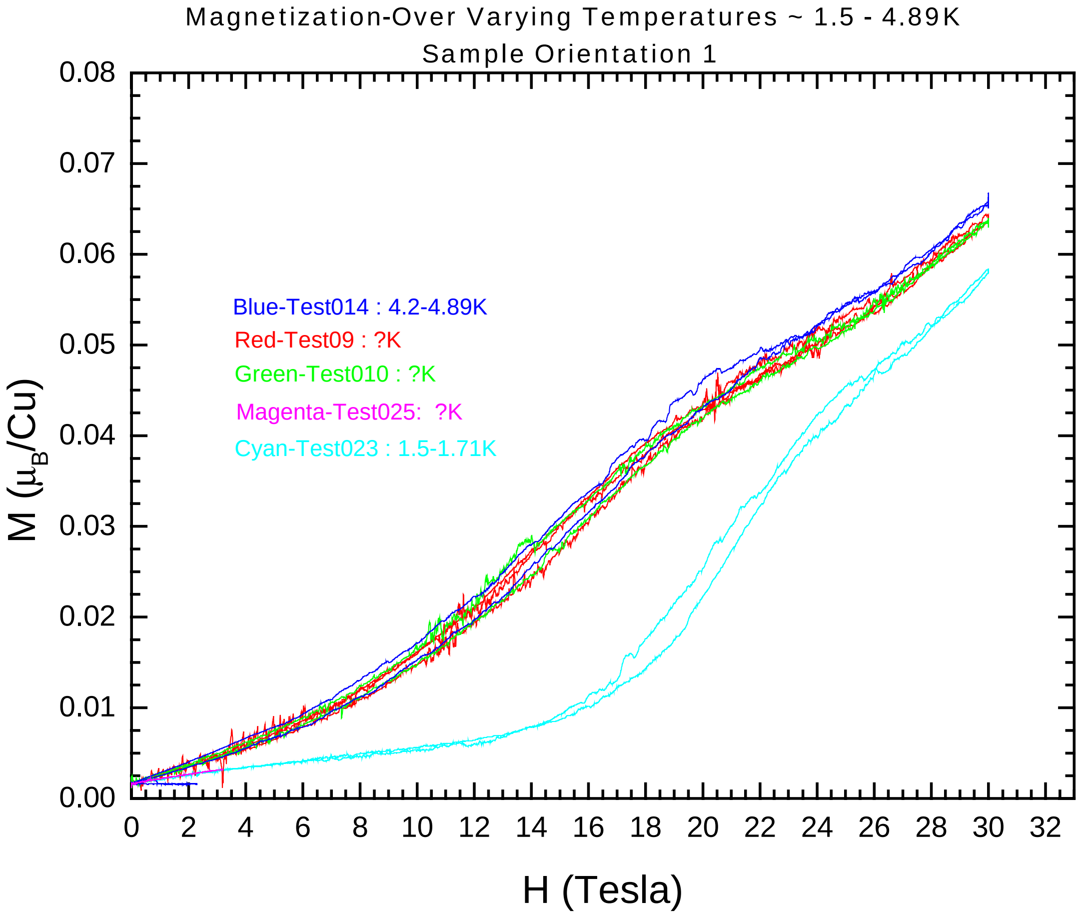

FIGURE 3. Magnetization in Bohr magnetrons per copper ion of $SrCu_2(BO_3)_2$ versus the magnetic field for the first sample orientation, note an off set was used .

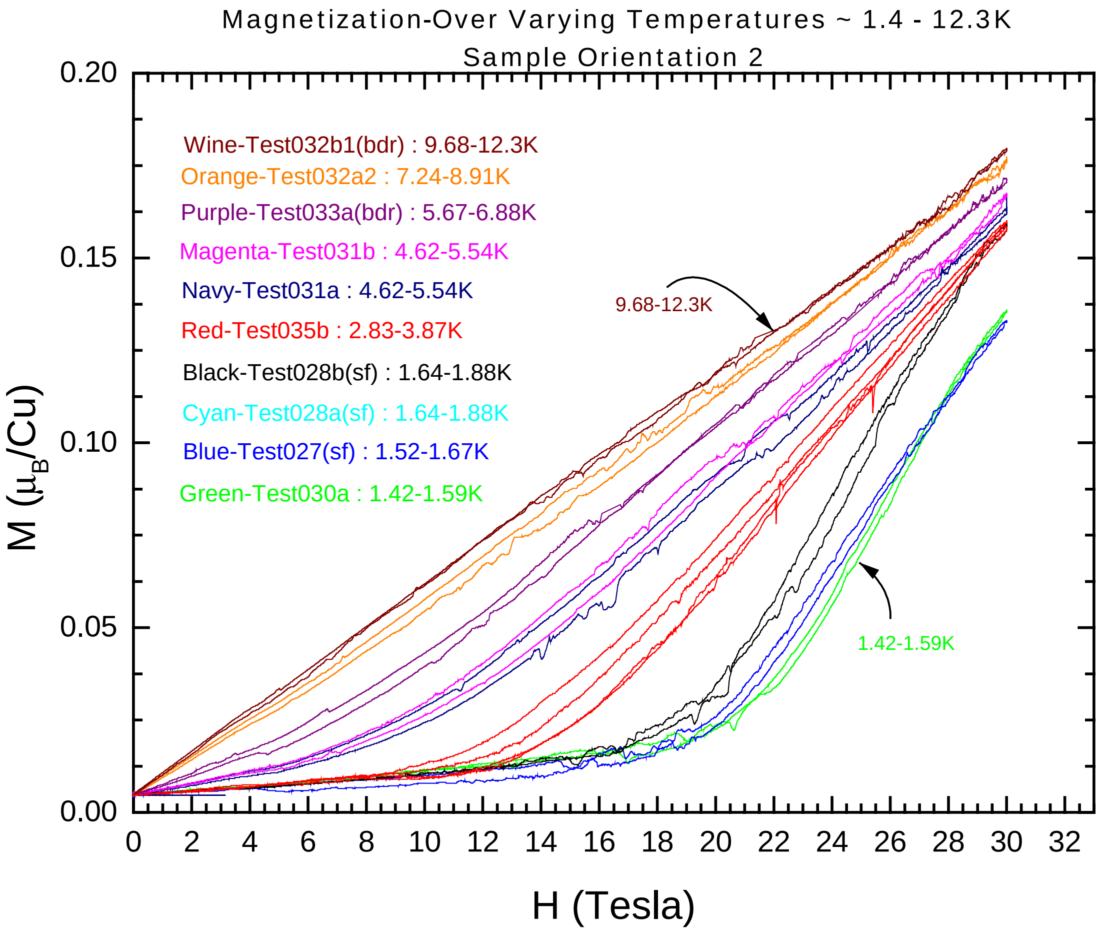

FIGURE 4. Magnetization in Bohr magnetrons per copper ion of $SrCu_2(BO_3)_2$ versus the magnetic field for the second sample orientation.

This behavior is not like that of ordinary, classical magnets in disordered states. For these magnets, the magnetization is inversely proportional to temperature as shown by Eq. (2) [6].

$$M = \frac{N(g\mu_B)^2 B}{3k_B T} \tag{2}$$

In classical antiferromagnets, the magnetization does increase with increasing temperature as seen in Eq. (3) [6],

$$M = \frac{k_B T x}{\mu_o g \mu_B \lambda} \tag{3}$$

if they are in the antiferromagnetically ordered state at temperatures below T_N, the Néel temperature, given by Eq. (4), where C is the Curie constant, A is the strength of the mean field for spins in site one, and Γ is the strength of the mean field for spins in site two. In the singlet case, in which sites one and two are equivalent cryptographically, then Γ is equal to –A and T_N becomes CA [6].

$$T_N = \frac{C}{2}(A - \Gamma) \tag{4}$$

However, $SrCu_2(BO_3)_2$ is known to be disordered in the field and temperature ranges studied in our experiment, therefore our findings are new and reflect the extremely quantum mechanical nature of this material.

At fields below the critical field of 20 T, the temperature dependence can only be understood in terms of the energy gap. As shown in Kageyama's paper, the ground state in this field region has zero magnetization; moreover, thermally it is not easy to excite the material because the energy gap must be overcome. At fields above H_C, the ground state is magnetic. In this field region no such energy gaps exist for magnetic excitations. Therefore, when compared

to the region below H_C, the magnetization changes more slowly with decreasing temperature.

The increase of magnetization with increasing temperature indicates that the lowest excitations have more magnetization than the ground state.

It should also be noted that in the earlier study, the sample was polycrystalline, unlike our study which utilized a single crystal sample. In 2000, Kageyama and his associates published another study, this time with a single crystal sample of $SrCu_2(BO_3)_2$ [7]. Again, our results are in agreement.

IV. Conclusions

Overall, the experiment was successful and the data we accumulated agreed with previous studies conducted on the sample. Unlike any of the other studies, however, our data clearly show that the magnetization of the sample depends the temperature of the sample. More problems did arise with the amount of noise in regard to the signal of the vibrating sample magnetometer. These problems are currently being addressed, and when remedied, the study can continue further for new orientations of the original sample or new samples all together.

V. Acknowledgements

First and foremost, I would like to thank Professor Yasumasa Takano for allowing me the opportunity to work with him for the second summer in a row. I would also like to thank the people of the Microkelvin Laboratory, Donna Balkcom, and Wayne Bromstead for their continual help and support. Thank you to the University of Florida Physics Department, Professor Kevin Ingersent, and Professor Alan Dorsey for allowing me the opportunity to be a part of this program. Lastly, I would like to express my appreciation for the funding of the REU program by the National Science Foundation.

References

[1] M. Ali Omar, *Elementary Solid State Physics* (Addison-Wesley Publishing Company, Inc.,US, 1993).

[2] H. Kageyama, K. Yoshimura, R. Stern, N.V, Mushnikov, K. Onizuka, M. Kato, K. Kosuge, C. P. Slichter, T. Gogo, and Y. Ueda, Phys. Rev. Lett. **82**, 3168 (1999).

[3] K. Kodama, M. Takigawa, M. Horvatić, C. Berthier, H. Kageyama, Y. Ueda, S. Miyahara, F. Becca, and F. Mila, *Science* **298,** 395 (2002).

[4] National High Magnetic Field Laboratory website, *Magnetic Science and Technology,* http://www.nhmfl.gov/magtech/ .

[5] Schematic diagram of a vibrating sample magnetometer, at URL www.uc.edu/~jph/emma/mag/html .

[6] M. Cardona, P. Fulde, K. von Klitsing, and H. J. Queisser, Eds., *Theory of Magnetism* (Springer-Verlag Berlin Heidelberg, New York, 1998).

[7] K. Onizuka, H. Kageyama, Y. Narumi, K. Kindo, Y. Ueda, and T. Goto, J. Phys. Soc. Japan **69**, 1016 (2000).