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6. AUTHOR(S)
Prof. Peter Schiffer

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
University of Notre Dame
Notre Dame, IN 46556

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13. ABSTRACT (Maximum 200 words)

The specific aim of this research program has been to understand the cooperative magnetic properties of geometrically frustrated antiferromagnets which are model frustrated systems. We have performed extensive thermal studies of one such material, gadolinium gallium garnet, which has highly unusual ground state properties. Because of its purity and the energy scale of the spin-spin interactions, this system offers a unique model to study the consequences of strong geometrical frustration. We have characterized a novel spin state in this material which appears to be a three dimensional "spin liquid", and we have shown that it is both distinct from a spin glass and has a highly unusual phase boundary with a long-range-ordered antiferromagnetic state. These results enhance our understanding of geometrically frustrated systems in that they greatly broaden the range of cooperative behavior which has been observed in such systems. In particular we have made the first study of the thermal excitations in a three-dimensional cooperative spin-liquid state. We also have begun studies of two other types of geometrically frustrated systems: a pyrochlore lattice of corner sharing tetrahedra and a kagome lattice of corner sharing triangles. In both of these systems we are examining the evolution of frustration as a function of dilution of the frustrated lattice with non-magnetic impurities.

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Final Report: Geometrically Frustrated Magnets as Model Systems
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PI: Peter Schiffer
University of Notre Dame

This document reports the progress of a research program which has been supported by a five-year PECASE award. Since the principal investigator has moved to a new institution, the remainder of the award will be made through a new grant, and this document is the final report on the initial grant covering approximately one half of the award's funding. The focus of the research program has been the low temperature magnetic phases of a model geometrically frustrated magnet, gadolinium gallium garnet (GGG). The magnetic sublattice of this system consists of two interpenetrating three-dimensional lattices formed of corner-sharing triangles. The high degree of frustration of the antiferromagnetic Gd-Gd exchange interactions leads to a variety of novel behavior in this material. This material is readily available in extremely high quality single crystals which makes it particularly well suited to serve as a model system for the study of geometrical frustration.

The magnetic ground state of GGG changes nature with applied field. Previous studies had demonstrated that the ground state in zero field was a spin glass and that the spins enter a long-range-ordered antiferromagnetic state in fields between 0.7 and 1.4 T. The spin state in intermediate fields, however, remained a mystery since the indications of spin-glass order disappeared and the system showed no evidence of other long range magnetic order. This intermediate-field (IF) state is a three-dimensional spin liquid -- a unique type of magnetic ground state associated with geometrical frustration in which the spins can fluctuate even though the available thermal energy is much smaller than the strength of the spin-spin interactions.

We have completed detailed studies of the thermal conductivity, the heat capacity, the magnetization, and the magnetocaloric properties of GGG in the IF regime. We find no long range order features in the IF regime down to temperatures below $T = 0.06$ K, which is consistent with the theorized properties of a spin-liquid state. We find a variety of interesting properties of the IF state, including decreasing thermal conductivity with increasing magnetic field (anomalous since thermal conduction is through phonons scattered by magnetic excitations which should be suppressed in a magnetic field).

The most interesting data, however, come at the phase boundaries of the IF state. We find definite evidence in the magnetocaloric data that the IF state has a sharp thermodynamic boundary separating it from the low-field spin glass state. This demonstrates that the IF state is a novel cooperative spin state, distinct from a spin glass. The phase boundary of the IF state with the long range ordered state was demonstrated to have a minimum in the field-temperature phase diagram. This is equivalent to reentrance of a disordered state from an ordered state upon cooling, something which happens only rarely in nature since it implies that the disordered state has less entropy than the ordered state. This phase boundary in GGG is perhaps most analogous to the phase boundary between solid and superfluid ^{4}He, one of the most important model systems in condensed matter physics, which also shows such reentrance due to the relative number of phonon modes in the solid and superfluid states. By analogy, the phase boundary in GGG can be viewed as being between a spin liquid and a spin solid. These results enhance our understanding of geometrically frustrated systems in that they greatly broaden the range of cooperative behavior which has been observed in such systems.

A significant portion of the research program has been the implementation of a new measurement tool, a magnetometer for the study of the low temperature magnetization of geometrically frustrated magnets. The magnetization of a magnetic material is the most important parameter in the understanding of its cooperative spin-behavior, since it measures the total vector sum of the spin orientation. How the magnetization varies as a function of temperature and magnetic field at low temperatures gives a direct window into the behavior of the spin ground state and the excitations among the spins. Unfortunately, while commercial instruments exist to measure the magnetization of materials at high temperatures, there is no comparable technology to measure magnetization in the temperature range of interest for the ground state of geometrically frustrated magnets.

This problem is particularly difficult since we need to measure the magnetization as a function of magnetic field in order to understand the behavior, something for which many measurement techniques (such as SQUID magnetometry) are not ideally suited.

In order to perform this measurement, we have developed a highly sensitive field-gradient capacitance magnetometer which can be used to measure the magnetization as a function of both temperature and magnetic field. We have used this magnetometer to complete studies of the magnetization of gadolinium gallium garnet, a three-dimensional frustrated magnet which we have examined previously by other measurement techniques. This material is particularly interesting in that it has a novel "spin-liquid" ground state at low temperatures for a wide range of fields, and one of the goals of our research program is to understand this spin-liquid state. These studies have lead to three important results. We were able to directly confirm that the spin liquid state has no dependence on its magnetic field history, demonstrating that it is indeed distinct from a spin glass state through comparing the field-cooled and zero-field-cooled magnetizations. We were also able to directly prove the existence of a re-entrance of the spin-liquid phase at high magnetic fields where the phase becomes unstable compared to a long-range-ordered antiferromagnetic ground state, something which had only been inferred indirectly based on purely thermal studies. Perhaps most importantly, however, we were able to map out the temperature and field dependence of the magnetization. We find that, for temperatures well below the energy scale of the spin-spin interactions, there is very little temperature dependence to the magnetization. This implies that the spins are closely coupled to their near neighbors and are relatively un-perturbed by thermal fluctuation despite the dynamic nature of the spin liquid state and the absence of long range order.

The combination of our magnetization and specific heat data allow us to analyze the nature of the low temperature excitations in the IF spin-liquid state. The temperature dependence of the specific heat changes with field, suggesting a crossover in the nature of spin excitations between the spin liquid phase and the high-temperature paramagnetic phase. Moreover, both the specific heat and magnetization at low temperatures can be consistently and quantitatively described by a spin-wave-like model of excitations with a gapped quadratic dispersion relation. The gap grows linearly with magnetic field, but appears to have a finite value even in the extrapolation to zero field. This is the first such detailed study of the low temperature thermal excitations in a three dimensional spin liquid, as well as the first evidence for a spin-gap in such a system.

Publications:

"A Study of the Low Temperature Thermal Properties of the Geometrically Frustrated Magnet: Gadolinium Gallium Garnet," Y. K. Tsui, N. Kalechofsky, C. A. Burns, and P. Schiffer, Journal of Applied Physics **85** 4512 (1999).

"Magnetic Field-Induced Transitions From Spin Glass to Liquid to Long Range Order in a 3D Geometrically Frustrated Magnet," Y. K. Tsui, C. A. Burns, J. Snyder, and P. Schiffer, Physical Review Letters **82** 3532 (1999).

"Thermal Studies of the Spin Liquid State and Analog to the Helium-4 Melting Curve in a Geometrically Frustrated Magnet," Y. K. Tsui, C. A. Burns, J. Snyder, and P. Schiffer, Physica B **280**, 296 (2000).

"Analog to the ^{4}He Melting Curve in a Model Geometrically Frustrated Magnet," Y. K. Tsui, J. Snyder, and P. Schiffer, Canadian Journal of Physics (in press).

"Thermodynamic Study of Excitations in a 3D Spin Liquid," Y. K. Tsui, J. Snyder, and P. Schiffer, Physical Review Letters (submitted)

Invited Talks and Seminars

"Geometrical Frustrated Antiferromagnets: Common Behavior and Unique Ground States," **Condensed Matter Seminar, Purdue University**, West Lafayette, Indiana, May 1, 1998.

"Geometrical Frustrated Antiferromagnets: Common Behavior and Unique Ground States," **Seminar, Bell Laboratories**, Murray Hill, New Jersey, June 5, 1998.

"Geometrical Frustrated Antiferromagnets: Common Behavior and Unique Ground States," **Condensed Matter Seminar, University of Toronto**, Toronto, Canada, October 23, 1998.

"Field-Induced Spin Glass to Liquid to Solid Transition in a Three-Dimensional Geometrically Frustrated Magnet," **Condensed Matter Seminar, Princeton University**, Princeton, NJ, December 4, 1998.

"Geometrical Frustration in Magnets: Common Behavior and Unique Ground States" **Condensed Matter Seminar, University of Missouri,** Columbia, MO April 21, 1999.

"Geometrical Frustration in Magnets: Common Behavior and Unique Ground States" **Condensed Matter Seminar, University of Cincinnati,** Cincinnati, OH June 2, 1999.

"Spin Liquid and Analog to the Helium-4 Melting Curve in a Geometrically Frustrated Magnet," **22^{nd} International Conference on Low Temperature Physics**, Helsinki, Finland, August 7, 1999.

"Geometrical Frustration in Magnets: Common Behavior and Unique Ground States" **Condensed Matter Seminar, Pennsylvania State University,** University Park, PA November 8, 1999.

"Spin Liquid Thermodynamics in Gadolinium Gallium Garnet (GGG)" **Highly Frustrated Magnetism 2000** Waterloo, CA , June 13, 2000.

Scientific Personnel Supported

Prof. Peter Schiffer (PI)
Yee-Kin Tsui (graduated Ph.D. Student, Ph.D. awarded in 2000)
Joseph Snyder (current Ph.D. Student, M.S. awarded in 2000)
Steve Potashnik (current Ph.D. Student, partial support)