FINAL TECHNICAL REPORT: DOE DEFG02-99ER45789
12/1/2012-11/30/2015

Project Title: Quantum Coherence and Random Fields at Mesoscopic Scales
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March 1, 2016

Accomplishments: We seek to explore and exploit model, disordered and geometrically frustrated magnets where coherent spin clusters stably detach themselves from their surroundings, leading to extreme sensitivity to finite frequency excitations and the ability to encode information. Global changes in either the spin concentration or the quantum tunneling probability via the application of an external magnetic field can tune the relative weights of quantum entanglement and random field effects on the mesoscopic scale. These same parameters can be harnessed to manipulate domain wall dynamics in the ferromagnetic state, with technological possibilities for magnetic information storage. Finally, extensions from quantum ferromagnets to antiferromagnets promise new insights into the physics of quantum fluctuations and effective dimensional reduction.

A combination of ac susceptometry, dc magnetometry, noise measurements, hole burning, non-linear Fano experiments, and neutron diffraction as functions of temperature, magnetic field, frequency, excitation amplitude, dipole concentration, and disorder address issues of stability, overlap, coherence, and control. We have been especially interested in probing the evolution of the local order in the progression from spin liquid to spin glass to long-range-ordered magnet.

(A) Quantum Annealing and Optimization: We have performed a proof-of-concept experiment [1] on a crystal containing 10^{20}, rather than hundreds, of quantum spins, which replicates some of the features of the current generation of much smaller specialized computers [2].

One of the key aspects of optimization is the process by which the special purpose computer settles into a solution to questions akin to the traveling salesman problem. The solutions to the problem exist in a landscape where the heights and depths of features are the total distance traveled – the best solution corresponds to the deepest valley. To find the deepest valley, the optimizer hops between valleys either by climbing to intermediate saddle points and then descending again, or via quantum tunneling between valleys. The first represents thermal annealing, while the second corresponds to quantum annealing. The relative weights of the thermal and quantum processes determine the nature of the final valley found [3-7].

The experiment looked at the valleys found after annealing with different rations of weights in the crystalline quantum magnet, $LiHo_xY_{1-x}F_4$. In this material, at temperatures near absolute zero, the speed and strength of thermal annealing can be controlled by rods of sapphire attached to a helium dilution refrigerator with more or less contact with the crystal. At the same time, the rate of quantum annealing can be controlled by means of a (transverse) magnetic field that acts to set the rate of quantum tunneling in the magnetic sample.

We find that when the system reaches its final valley via thermal annealing alone, it was dramatically different from the state reached when the thermal annealing was weakened and quantum annealing was turned on. We show in Fig. 1 a schematic of the state found after quantum annealing, where certain regions of the crystal have spins in quantum superposition and others have definite classical characteristics, marked by fixed up or down arrows; classical annealing of the same system leaves behind regions exclusively of the latter variety. Applied to practical and programmable quantum optimization computers [8], the results imply that quantum optimizers could obtain very different types of solutions to problems such as the traveling salesman problem when compared with conventional annealing techniques, a finding that will affect both the design and use of quantum optimization systems.

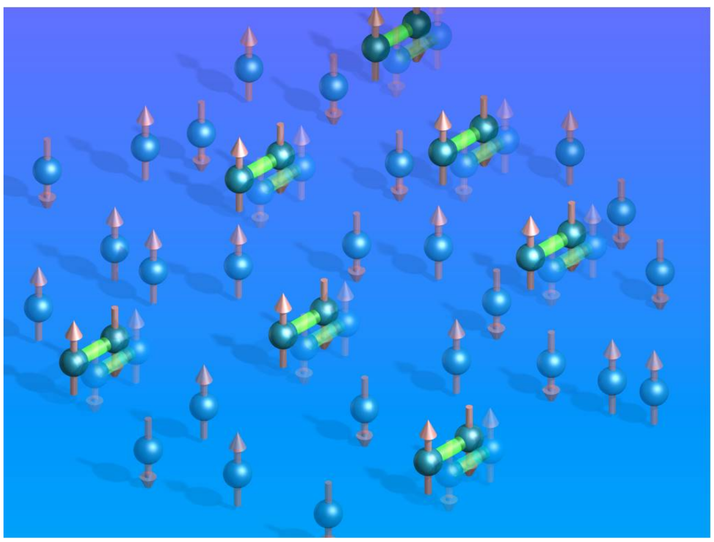

Fig. 1: The spheres represent the atoms of a quantum magnet, the arrows denote their spin orientation. The green bars indicate coupling between atoms, and the adjacent ghosted images illustrate quantum tunneling between different spin directions. This particular spin configuration displays a quantum solution to the computational problem of determining the ground state in a spin liquid [From Ref. 7].

The key difference involves the collective nature of the relaxation process. The small values of transverse field are orders of magnitude smaller than the field scales required to produce single-ion effects. Describing this system using the Hamiltonian for the Ising Model in transverse field,

$$H = -\sum_{i,j}^{N} J_{ij}\sigma_i^z\sigma_j^z - \Gamma\sum_i^N \sigma_i^x \qquad (1)$$

where the σ's are Pauli spin matrices and the J_{ij}'s are longitudinal couplings, permits a quantitative evaluation of the pertinent energies. Experiments and mean-field theory calculations find the mixing term Γ to be approximately quadratic in H_t for $H_t < 20$ kOe, with Γ(1 kOe) = 30 mK [9-12]. Clearly, in the single ion picture only transverse fields on the order of 1 kOe or more will be relevant at temperatures of order 1 K; however, we have observed unmistakable effects on the evolution of the magnetic system for transverse cooling fields as small as 10 Oe. We argue that in order for the interaction energy between a magnetic moment and a transverse field this small to be relevant at T ~ 0.1 K, the moment must be comprised of order 100 spins adding coherently.

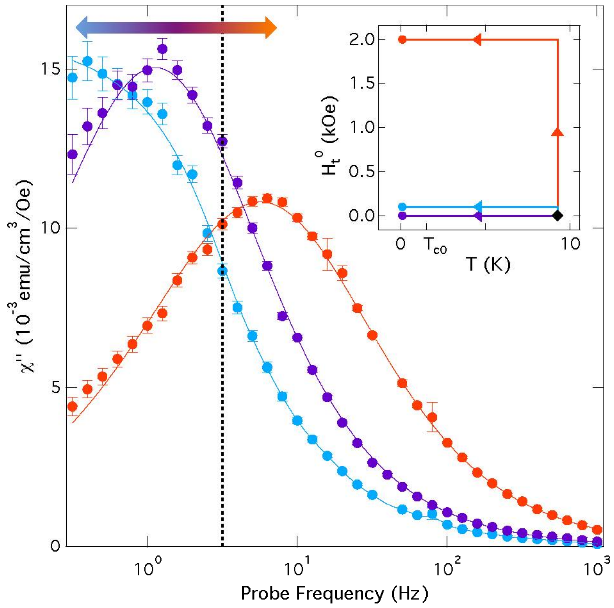

Fig. 2: Contrasting response of the imaginary part of the susceptibility to small and large transverse fields (inset) after cooling in a constant energy configuration. The shift down in frequency is unexpected, indicating collective behavior of hundreds of spins transiting the free energy surface and changing the system's ground state. At large transverse fields, tunneling probabilities are enhanced and the spectrum moves to higher frequency [Following Ref. 1].

As also deduced from the hole-burning results, the natural interpretation of this many-body effect is that clusters of spins are the relevant degrees of freedom. Indeed, the interaction energy of a field of 100 Oe with a single spin is ~3 mK, negligible compared to thermal fluctuations at T = 90 mK. These data therefore indicate that the field-cooled relaxation is a many-body effect in which clusters of spins act coherently, reacting to an applied field *en masse* in order to find a deeper local minima in the free-energy landscape as the sample cools. It also suggests a strategy to similarly enhance quantum relaxation in crystals of Li(Ho,Y)F_4 at higher dipole (Ho) concentration, where long-range magnetic order emerges.

(B) Nonlinear Dynamics of Coherent Spin Clusters: As discussed above, the study of extended quantum-coherent states serves both as a laboratory for fundamental quantum mechanics and as a benchmark for the practical requirements of quantum computation. In order to understand the dynamics and energetics of these coherent states, it is necessary to study how they interact with each other and with the outside world. Minimizing the interactions between the coherent state and a continuum of states in the broader environment is an important goal for realizing an effective quantum computer [13-16]. However, these environmental couplings are often weak compared to the transitions inside the coherent structure, making it difficult to study them directly. Recently, it has been posited that many weak couplings of this sort can be probed by pumping the system into a nonlinear response regime [17], saturating the discrete transition associated with the coherent state, and using the emergence of a Fano resonance to characterize the interactions with the continuum states [18]. In this work [19], we use a magnetic adaptation of the Fano resonance technique [20] to examine the coupling between coherent spin clusters quantum-mechanically isolated from their neighbors in the quantum spin liquid, $LiHo_{0.045}Y_{0.955}F_4$, and the bath of dilute spins in which they are embedded. Significantly, we find that varying the microscopic Hamiltonian via application of a transverse magnetic field can be used to tune these interactions without destroying the coherence of the cluster states (Fig. 3).

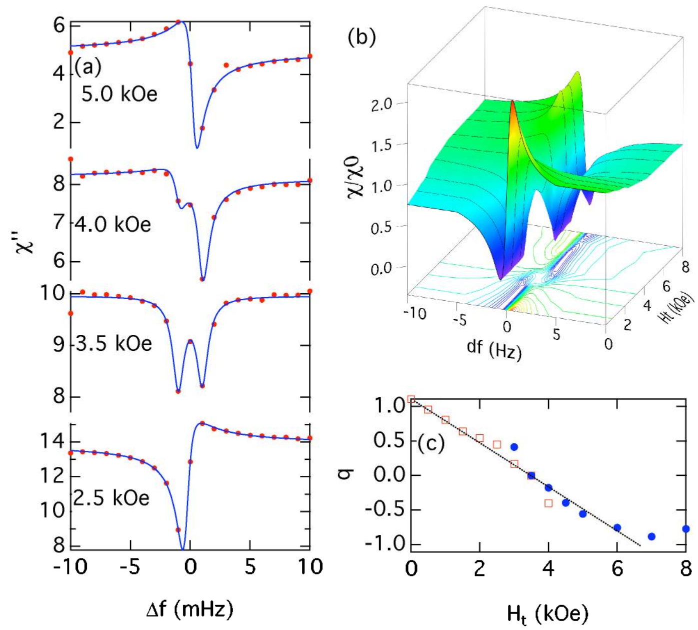

Fig. 3: Nonlinear absorption in the presence of a 202 Hz, 0.3 Oe pump field as a function of transverse field at T = 0.11 K. (a) Measured absorption (points), and fits to Fano resonance forms. Tuning energy levels with a transverse field avoids the decoherence effects associated with heating. (b) Normalized absorption as a function of frequency and transverse field. (c) Fano coupling parameter *q* vs. transverse field. *q* describes the relative transition pathways with weak connection to a spin bath [Following Ref. 19].

(C) Geometric Frustration: We have extended our studies of coherent clusters of spins [21-23] effectively decoupled from the surrounding magnetic environment (so-called "quantum protectorates" [24,25]) in spin liquids to geometrically-frustrated systems. In the case of GGG ($Gd_3Ga_5O_{12}$), spin freezing is suppressed classically by geometrical frustration, without the need for dilution. Frustration refers to the inability of complex systems to simultaneously satisfy all constraints, epitomized by the triangular antiferromagnet. If a spin on a particular triangle has two antiparallel neighbors, the latter two must be parallel and therefore do not satisfy their own pairwise preference for antiparallelism. The outcome for a large network of triangles is that the number of lowest energy spin configurations is not finite, as for a square lattice antiferromagnet, but grows linearly with the number of spins, resulting in a finite T = 0 entropy [26-28].

We have investigated the effects of up to 1% Neodymium substitution for Gallium on the ac magnetic response at temperatures below 1 K in both the linear and nonlinear regimes [29]. Substitutional disorder actually drives the system towards a more perfectly frustrated state, depressing any signatures of ordering such as a plateau in the low frequency dissipative response (Fig. 4). The magnetic Nd substitution apparently compensates for the effects of imperfect Gadolinium/Gallium stoichiometry and, at the same time, more closely demarcates the boundaries of isolated, coherent clusters composed of hundreds of spins. Optical measurements of the local Nd environment substantiate the picture of an increased frustration index with doping. Correlating the linear ac magnetic susceptibility with the non-linear response at both 15 Hz and 5 kHz, where features in the linear spectrum indicate that holes may be burned, should provide insights into the continuum of frustrated states as well as the nature of the quantum-mechanically isolated degrees of freedom.

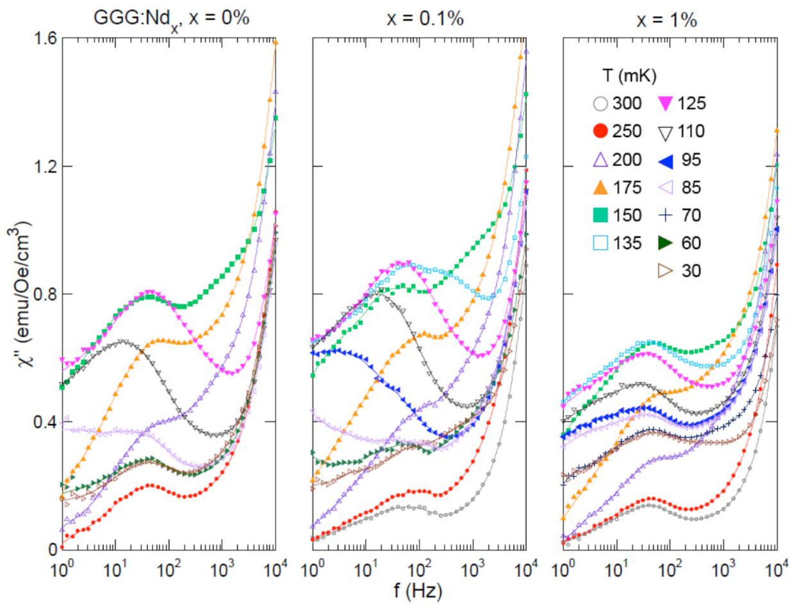

Fig. 4: The imaginary part of the ac magnetic susceptibility over 4 decades of frequency at milliKelvin temperatures for crystals of GGG with 0, 0.1% and 1% Nd doping. The disorder shifts and modulates the low frequency signatures of local magnetic order and spin cluster physics [From Ref. 29].

(D) Tunable Domain Pinning in Ferromagnets: It is no accident that techniques to mechanically and magnetically harden and soften materials coincide. The same impurities and grain boundaries that prevent dislocations from flowing prevent magnetic domains from switching. Strategies to pin magnetic domains typically are implemented during materials preparation by varying the composition, structure, and morphology, locking in a set of properties that cannot be modified subsequently. Of particular technological interest is ultra-high density magnetic storage, where bits are warmed close to or above their ferromagnetic transition temperature for writing and then cooled for long-term retention and reading [30]. A more powerful approach would be to adapt material properties *in situ* as circumstances demand, continuously tuning the energetics of domain reversal without changing temperature.

We adopt a new tactic to regulate isothermal magnetization reversal in real materials by exploiting the random-field Ising model (RFIM), where a site-random magnetic field acts to orient magnetization locally in competition with the underlying exchange couplings that favor homogeneous magnetism [31,32]. Random fields act on ferromagnets via their pinning of domain walls, creating barriers to motion that increase with random field amplitude [33,34]. The rising barrier height decreases the probability of reversal at a given applied field, and if the amplitude of the random field contribution can be controlled continuously, then the pinning potential can be increased or decreased on demand.

We have extended earlier (DOE-supported) work at milliKelvin temperatures on dipolar ferromagnets [35,36] by considering a rare-earth ferromagnet, $Nd_2Fe_{14}B$, with high crystalline anisotropy and a tendency to form elongated grains. Within a grain, the individual spins are exchange-coupled, yielding a strong interaction energy that magnetizes the entire grain and yields a Curie temperature of order 300° C [37]. Each grain acts like a single effective spin with a moment equivalent to millions of Bohr magnetons because of the crystalline and shape anisotropy, and couples to neighboring grains via the dipolar interaction. This ensemble of grains mimics the collection of individual dipoles in previously studied RFIM materials [35,36,38] and may be expected to exhibit comparable behavior.

We show in Fig. 5 a series of nested subloops for transverse fields of 0 and 6 kOe. The subloops allow us to explore the energy hierarchies that form during nested longitudinal field trajectories [39]. Each measurement begins from the saturated magnetization state. The longitudinal field is then adiabatically ramped through a series of nested subloops (with the maximal h_l decreasing by 1 kOe per subloop) that cover either the low or high longitudinal field ranges. As the subloop series progresses, the sample converges on its equilibrium magnetization, and the series of nested loops forms a hierarchy of energies at each "turning point" [40].

The subloops in the low longitudinal field regime (Fig. 5a,b) narrow as the transverse field is applied. Importantly, application of the transverse field does not change the essential self-similarity of the set of subloops. We contrast this evolution with the behavior of the nested subloops at high longitudinal field (Fig. 5c,d). First, we note the clear broadening in the presence of transverse field, consonant with increased M-H energy storage due to increased pinning. Second, we observe that the subloops taken at H_t = 0 exhibit the same sort of self-similarity exhibited in the low-field regime. With application of a transverse field, however, this self-similarity is lost and there is a monotonic change in slope of each subsequent nested subloop. Computer simulations demonstrate that for small values of randomness below a critical threshold, hysteresis subloops maintain their self-similar nature and collapse onto the same full-scale hysteresis curve, but that in the strong randomness limit, subloops are no longer self-similar and scale inwards in a manner qualitatively similar to our experimental results at high longitudinal and transverse fields [40]. This implies that the effect of the transverse field is to increase the relative effects of disorder and pinning via augmentation of the random fields.

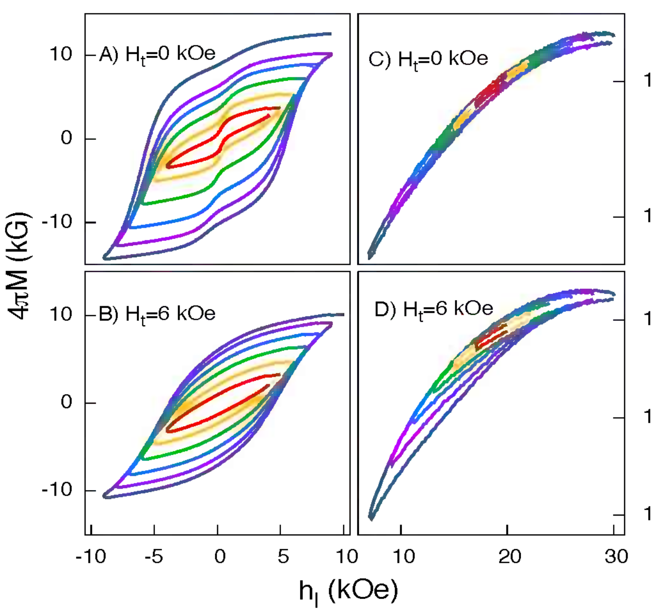

Fig. 5: Effects of transverse field on subloop evolution in the rare-earth ferromagnet $Nd_2Fe_{14}B$.
(a,b) Nested subloops at low longitudinal field at room temperature. The subloops remain self similar in zero and 6 kOe of transverse field.
(c,d) Nested subloops at high longitudinal field. Applying a transverse field *broadens* and rotates the subloops, destroying their self-similar nature [Following Ref. 39].

The dynamics of the domain walls can be elucidated through Barkhausen noise [41,42], the noise generated by switching domains occurring at audio frequencies under the influence of a changing longitudinal field. We have demonstrated [43] that the Barkhausen noise can be suppressed by the application of a transverse field (Fig. 6), reinforcing the interpretation of isothermal hardening of the magnet. Measurements at a series of longitudinal magnetic fields, transverse magnetic fields, and temperatures have permitted comparisons to scaling theories for the RFIM.

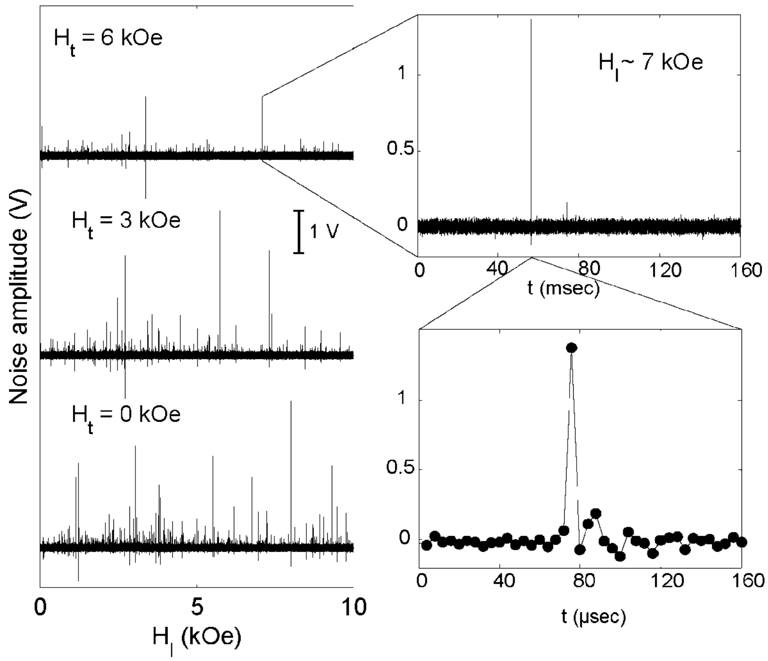

Fig. 6: Barkhausen noise recorded on sweeping the longitudinal magnetic field through a hysteresis loop in the ferromagnet $Nd_2Fe_{14}B$. All data are taken at fixed temperature. The noise decreases with increasing transverse magnetic field as the transverse field induces local random magnetic fields that pin the domains. The ability to alter the pinning potential in this manner may have applications in magnetic storage [From Ref. 43].

Journal Publications

1. "Dipolar Antiferromagnetism and Quantum Criticality in $LiErF_4$," C. Kraemer, N. Nikseresht, J. O. Piatek, N. Tsyrulin, B. Dalla Piazza, K. Kiefer, B. Klemke, T. F. Rosenbaum, G. Aeppli, C. Gannarelli, K. Prokes, A. Podlesnyak, Th. Straessle, L. Keller, O. Zaharko, K. W. Kraemer, and H. M. Rønnow, Science **336**, 1416 (2012). Acknowledgment: "Work at the University of Chicago was supported by the U.S. Department of Energy Basic Energy Sciences."
2. "Sub-Kelvin ac Magnetic Susceptometry," M.A. Schmidt, D.M. Silevitch, N. Woo, and T.F. Rosenbaum, Rev. Sci. Instrum. **84**, 013901 (2013). Acknowledgment: "This work was supported by the U.S. DOE Basic Energy Sciences (Grant No. DE-FG02-99ER45789)."
3. "Using Thermal Boundary Conditions to Engineer the Quantum State of a Bulk Magnet," M.A. Schmidt, D.M. Silevitch, G. Aeppli, and T.F. Rosenbaum, Proc, Nat. Acad. Sci. **111**, 3689 (2014). Acknowledgment: "The work at The University of Chicago was supported by Department of Energy Basic Energy Sciences Grant DE-FG02-99ER-45789."
4. "Reversible Disorder in a Room Temperature Ferromagnet," S.L. Tomarken, D.M. Silevitch, G. Aeppli, B.A.W. Brinkman, J. Xu, K. A. Dahmen, and T.F. Rosenbaum, Advanced Functional Materials **24**, 2986 (2014). Acknowledgment: "The work at The University of Chicago was supported by Department of Energy Basic Energy Sciences Grant No. DE-FG02-99ER-45789."
5. "Quantum Tunneling vs. Thermal Effects in Experiments on Adiabatic Quantum Computing," D.M. Silevitch, T.F. Rosenbaum, and G. Aeppli, Eur. Phys. J. Special Topics **224**, 25 (2015). No acknowledgment permitted.

6. “Interplay of Disorder and Geometrical Frustration in Doped Gadolinium Gallium Garnet,” N. Woo, D.M. Silevitch, C. Ferri, S. Ghosh, and T.F. Rosenbaum, J. Phys.: Cond. Matt. **27**, 296001 (2015). Acknowledgment: “Work at The University of Chicago was supported by the Department of Energy Office of Basic Energy Sciences, Grant No. DE-FG02-99ER-45789.”

7. “Barkhausen Noise in the Random Field Ising Magnet $Nd_2Fe_{14}B$,” J. Xu, D.M. Silevitch, K.A. Dahmen, and T.F. Rosenbaum, Phys. Rev. B **92**, 024424 (2015). Acknowledgment: “Work at The University of Chicago was supported by the Department of Energy Office of Basic Energy Sciences, Grant No. DE-FG02-99ER-45789.”

8. “Sub-Kelvin Magnetic and Electrical Measurements in a Diamond Anvil Cell with *in situ* Tunability,” A. Palmer, D.M. Silevitch, Y. Feng, Y. Wang, R. Jaramillo, A. Banerjee, Y. Ren, and T.F. Rosenbaum, Rev. Sci. Instrum. **86**, 093901 (2015) [Editor’s Pick]. Acknowledgment: “D.M.S. acknowledges support from the U.S. Department of Energy Office of Basic Energy Sciences (Grant No. DE-FG02-99ER45789).”

9. “Spin Order and Kagome Magnetization of Single Crystalline $Cu_4(OH)_6FBr$,” T.-H. Han, J.A. Schlueter, and J. Singleton, preprint. Acknowledgment: “T.-H.H. thanks the support from Grainger Fellowship provided by the Department of Physics, University of Chicago and the support from the Department of Energy (DOE) (DE-FG02-99ER45789).”

Book

A. Dutta, G. Aeppli, B.K. Chakrabarti, U. Divakaran, T.F. Rosenbaum, and D. Sen, *Quantum Phase Transitions in Transverse Field Models: From Statistical Physics to Quantum Information* (Cambridge University Press, New Delhi, 2015), ISBN 978-1-107-06879-7.

In recent years, there has been an upsurge of studies interconnecting the phenomena of quantum phase transitions, non-equilibrium dynamics, and quantum information and computation. These studies are important from the viewpoint of fundamental physics as well as for developing new quantum technologies. This book is the first attempt to connect these different fields, mentioning both the promises and the problems and incorporating discussions of the most recent technological developments.

Acknowledgment: “Thomas Rosenbaum acknowledges DOE BES Grant No. DE-FG02-99ER45789.”

Patent

System and Method for Manipulating Domain Pinning and Reversal in Ferromagnetic Materials; D.M. Silevitch, T.F. Rosenbaum, and G. Aeppli; US Patent No. 8,558,333; October 15, 2013.

Personnel: Graduate Student: Nayoon Woo (now at Intel Labs, Oregon); Research Scientist (partial support): Daniel Silevitch.

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