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Grant Information

Grant Number	HR0011-07-1-0031
Title of Research	Advanced Materials for Quantum Computing
Principal Investigator	Dr. Charles J. O'Connor
Organization	University of New Orleans, New Orleans, LA 70148

FINAL TECHNICAL REPORT – April 28, 2010

Objectives:

The objective of this research project is the development of new solid state qubits for quantum computing using two main strategies: rare earths ions and ruthenates. The study of various sized spin based qubits in different condensed matter systems will allow us to understand and optimize the tradeoff between coherence and controllability. The effort is focused in synthesizing the qubit systems at AMRI-UNO, experimentally probing their suitability for quantum computation, and quantum computing device prototyping.

(1) Solid State Qubits – Dr. Leonard Spinu

Research Progress:

- Sr₂RuO₄ thin films were routinely deposited by Pulsed laser Deposition (PLD) in the last quarter. Several series of samples were prepared using different deposition conditions as substrate temperature, laser fluence and reactive gas pressure. Structural characterization by X-ray diffraction and electron microscopy confirms the ruthenate deposition.
- Low temperature resistivity measurements down to 15 mK using dilution refrigerator do not show superconductive behavior of the films.

Publications:

1. H. Pham, D. Cimpoesu, A. Stancu, and L. Spinu, "Switching behavior of a Stoner-Wohlfarth particle subjected to spin-torque effect," *Journal of Applied Physics*, vol. 103, p. 3, Apr 2008.
2. C. Radu, D. Cimpoesu, A. Stancu, and L. Spinu, "Measurement of the critical curve of a synthetic antiferromagnet," *Applied Physics Letters*, vol. 93, p. 3, Jul 2008.
3. D. Cimpoesu, H. Pham, A. Stancu, and L. Spinu, "Dynamic and temperature effects in spin-transfer switching," *Journal of Applied Physics*, vol. 104, p. 7, Dec 2008.
4. Z. Qu, J. Peng, T. J. Liu, D. Fobes, L. Spinu, and Z. Q. Mao, "Complex electronic states in double-layered ruthenates (Sr_{1-x}Cax)₃Ru₂O₇," *Physical Review B*, vol. 80, pp. 115130-1-115130-7, Sep 2009.

5. B. Qian, Z. Qu, J. Peng, T. J. Liu, X. S. Wu, L. Spinu, and Z. Q. Mao, "Structural, magnetic, and electronic transport properties of $(\text{Sr}_{0.9}\text{Ca}_{0.1})_3\text{Ru}_2\text{O}_7$ single crystal," *Journal of Applied Physics*, vol. 105, p. 3, Apr 2009.
6. W. Bao, Y. Qiu, Q. Huang, M. A. Green, P. Zajdel, M. R. Fitzsimmons, M. Zhernenkov, S. Chang, M. H. Fang, B. Qian, E. K. Vehstedt, J. H. Yang, H. M. Pham, L. Spinu, and Z. Q. Mao, "Tunable $(\delta\pi, \delta\pi)$ -Type Antiferromagnetic Order in $\alpha\text{-Fe}(\text{Te}, \text{Se})$ Superconductors," *Physical Review Letters*, vol. 102, p. 4, Jun 2009.

Presentations:

Switching Behavior of a Stoner-Wohlfarth Particle Subjected to Spin-Torque Effect
 MMM Conference 2007
 November 5-9, 2007, Tampa, FL

Controlled Fabrication of Novel Materials for Nanosensors and Quantum Computing
 Next Generation Materials for Defense 2007
 December 10-12, Arlington, VA

Dynamic and temperature effects in spin-transfer switching
 D. Cimpoesu, H. Pham, A. Stancu, and L. Spinu
 APS March 2008 Meeting, New Orleans, LA, March, 10-14, 2008

Switching behavior of a Stoner-Wohlfarth particle subjected to spin-torque effect
 H. Pham, D. Cimpoesu, A. Stancu, and L. Spinu
 APS March 2008 Meeting, New Orleans, LA, March, 10-14, 2008

Study of magnetization switching in synthetic antiferromagnets
 C. Radu, D. Cimpoesu, L. Spinu, and A. Stancu
 APS March 2008 Meeting, New Orleans, LA, March, 10-14, 2008

Relation between structure and magnetic properties of $\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$
 J. Peng, Z. Qu, L. Spinu, T.J. Liu, D. Fobes, and Z.Q. Mao
 APS March 2008 Meeting, New Orleans, LA, March, 10-14, 2008

Magnetic phase diagram of $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ ($0 < x < 1$)
 Z.Q. Mao, Z. Qu, L. Spinu, J. Peng, T.J. Liu, D. Fobes, V. Dobrosavljevic, H.Q. Yuan, W. Bao, J.W. Lynn
 APS March 2008 Meeting, New Orleans, LA, March, 10-14, 2008

Study of Dipolar Interaction in array of MRAM Cells
 Dorin Cimpoesu, L. Spinu and Al. Stancu
 Poster presentation at the 53rd MMM Conference, Austin, TX, November 10-14 2008

(2) Quantum Computing

co-PI: John Wiley w/ Postdoc Dr. Jonglak Choi

Project Name: Quantum Computing

1. Brief Narrative: Our group is working to grow single crystals of materials for quantum computing project. We are using a four-mirror floating zone furnace for the growth. Our focused targets have been Scheelites and its spin-doped crystals, such as LaNbO_4 that is providing a matrix with no spin, and $\text{La}_{1-x}\text{Ln}_x\text{NbO}_4$ ($0.001 \leq x$, $\text{Ln} = \text{Nd, Eu, and etc.}$).

2. Objectives:

We have focused on the growth of quality single crystal samples, such as superconducting Sr_2RuO_4 , $\text{Sr}_3\text{Ru}_2\text{O}_7$, Scheelite phases $\text{A}_{1-x}\text{Ln}_x\text{WO}_4$, and $\text{La}_{1-x}\text{Ln}_x\text{NbO}_4$ ($0 \leq x \leq 0.001$), by floating zone methods. Additionally, we provided polycrystalline targets for Spinu's group for growing thin film using PLD.

3. Research Progress:

Initially in this study, we have focused on growing single crystals of Scheelites including $\text{A}_{1-x}\text{Ln}_x\text{WO}_4$ ($0.001 \leq x$, $\text{A} = \text{Ca, Sr, Ln} = \text{spin containing f-block elements}$). These tungstates were problematic and turned out to be unsuitable for floating zone methods due problems with fabrication of rods for the growth and uneven melting within the molten zone during growth. Some crystals were grown, but of poor quality. We did have much better success with the growth of niobate-based Scheelite compounds instead and have produced crystals, $\text{La}_{1-x}\text{Ln}_x\text{NbO}_4$ ($0.001 \leq x$, $\text{Ln} = \text{Nd, Eu}$). In order to prepare the pure polycrystalline rods of the Scheelites the preheated La_2O_3 and Nb_2O_5 were used for the reactions, and the optimized temperature for the reactions and annealing were $\sim 1300^\circ\text{C}$ and $\sim 1350^\circ\text{C}$, respectively. All of the dense polycrystalline samples showed monoclinic LaNbO_4 structure confirmed in XRD.

The synthesized polycrystalline rods were loaded into floating zone furnace, and attempted to be grown as single crystals under various environment, such as Ar gas or 10 % O_2 gas flow etc., in atmospheric pressure, $\sim 65\%$ as the power, and up to 4 mm/hr for the growth speed environment as slowest one. Slow cooling was adapted to all of the systems to get the crystals without any defect which could be made during the phase change from tetragonal to monoclinic around 520°C . The grown crystal ingots were easily and sharply cleaved in a slightly tilted angle direction to the growth direction by the slight stress with a laser blade.

XRD scans were performed on the surfaces of the cleaved faces of the crystals, which is the direction of the growth, and showed high crystallinity and preferred orientation to (040) directions that is the long axis of the LaNbO_4 monoclinic system. It means that the growth direction of the crystals is long axis in all of the samples. The Laue pattern and single crystallography measurements on all of the cleaved sample surfaces, however, showed not a single crystal evidences; for example, $\text{La}_{1-x}\text{Eu}_x\text{NbO}_4$ (4 mm/hr, growth speed) has one monoclinic phase, space group = $I_{12/c1}$, $a=5.5315$, $b=11.4933$, $c=5.2168$, $\alpha=90.000$, $\beta=94.159$, $\gamma=90.000$ with several different orientation of crystals in the grown crystal ingots.

XANES measurements on all of the crystals including LaNbO_4 showed the evidence of La^{3+} and Nb^{5+} , and the existence of doped elements, Nd^{3+} and Eu^{3+} as detected in fluorescence measurements.

EPR measurements on all of the recently grown crystals including LaNbO_4 have been showing unexpected spin signatures in the spectra. Neither the niobium nor the lanthanum have unpaired electrons, so the resonances must be arising from a different source. One possibility is that oxygen defects have been produced during the growth process and reduced niobium ions are present. Current efforts are concentrating on lowering the growth rate to $< 4\text{mm/hr}$ in an effort to improve crystal quality. Further an atmosphere with oxygen environment, $\geq 10\%$ should also serve to minimize oxygen defects.

4. Publications: none

5. Presentations: none

(3) Quantum Computing with Magnons

co-PI: Leszek Malkinski w/ Postdoc Dr. Seong-Gi Min

Project Name: Quantum Computing with Magnons

1. Brief Narrative:

Quanta of spinwaves called magnons can be used to exchange quantum information between solid state qubits. The project was driven by the concept of spiwave bus proposed by A. Khitun in 2001 which is presented in Fig.1.

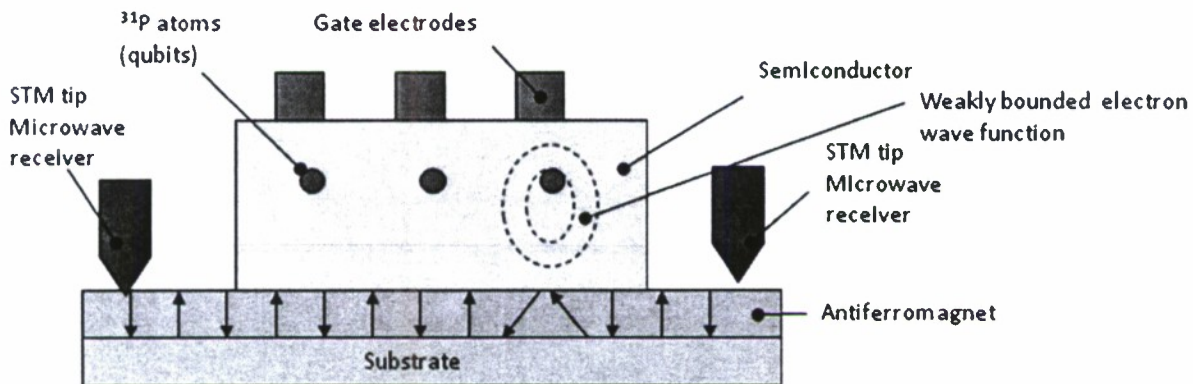


Fig.1. A schematic of Kithun's original concept of spinwave bus for quantum computing.

In his original form the model assumed that weakly bound electrons of implanted phosphorus atoms in silicone can generate spinwaves in 2 dimensional antiferromagnetic film when pushed toward the film by a pulse of negative potential applied to electrodes above the P atoms. The detection of the spiwaves would be achieved using STM tips. Although this concept was clever the practical realization with existing technology is extremely difficult or even impossible.

Therefore, we proposed to investigate the facility of using the spinwaves for quantum communications in a modified form. The major efforts were:

- to find appropriate medium for propagation of spinwaves
- design microwave circuit capable of generating and detecting spinwaves.

2. Objectives:

The main objectives of the research were to fabricate device suitable of locally generating and receiving spinwaves in nanoscale lines. This required two tasks to be accomplished:

- a) Fabricate new materials exhibiting very low attenuation of spinwaves which ensure long distances for propagation of the spinwaves.
- b) Design microwave circuit for generation and detection of the spinwaves.

3. Research Progress:

Our first choice of the material for the propagation of the spiwaves was Yttrium-Iron Garnet which is known for low attenuation of spinwaves. However, we found that very thin submicron YIG films contained defects and were found improper for spin-bus application, unlike thick films where different spinwave modes can propagate for distances of millimeters or even centimeters. Because of large magnetization, transition metal alloys have been considered good candidates for quantum computing applications. The challenge, however, was to sputter deposit these materials in the form of single-crystalline films with substantially lower number of defects compared to polycrystalline ones. We have been using Reflection High Energy Diffraction system purchased through this grant to monitor growth of single crystalline Permalloy films. The morphology of the samples studied by the scanning atomic force microscopy and Fig.2

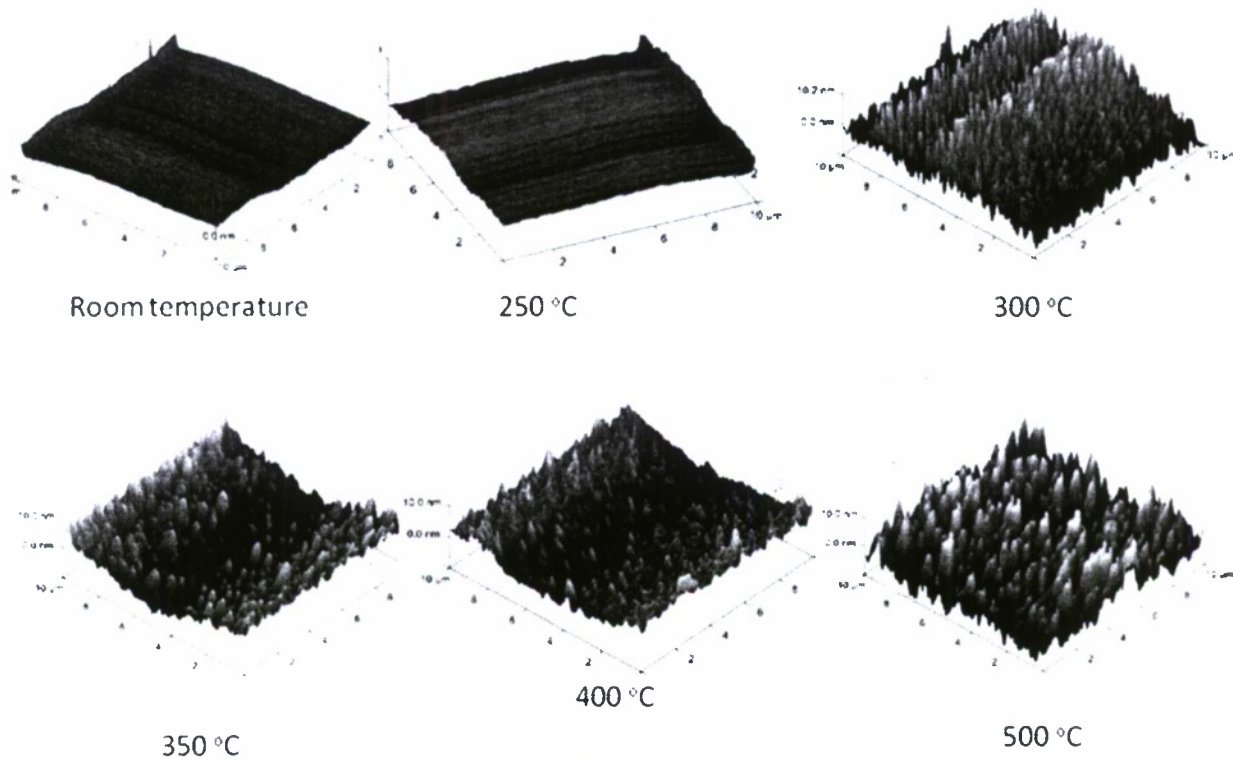


Fig.2. Effect of the substrate temperature on the morphology of thin Py films.

The examples of the ferromagnetic resonance measurements of the films are presented in Fig. 3. And numerical values of the linewidth are summarized in table 1. The best conditions for sputtering were found for the sputtering on a substrate at 250°C where about 31 Oe linewidth was achieved at 9.8 GHz independent on the orientation of the field with respect of the film.

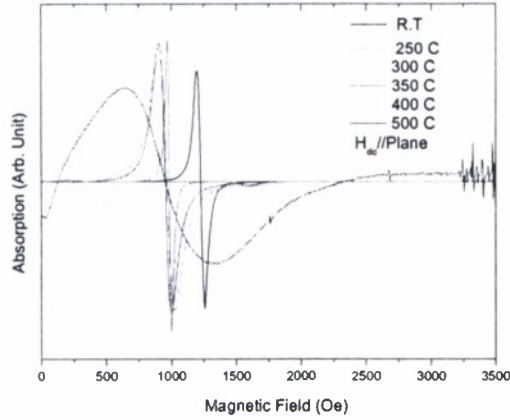


Fig. 3. The ferromagnetic resonance curves of the Py films grown in different conditions

Temp.	ΔH_{p-y} (Oe) Y-axis	ΔH_{p-x} (Oe) x-axis
R.T	58.65	50.83
250	31.28	31.28
300	113.39	35.19
350	93.84	23.46
400	117.3	31.28
500	660.8	93.84

Tab.1. Summary of the effect of the substrate temperature on the FMR linewidth measured with the field in- and out- of film plane

Another issue discovered in terms of resonance linewidth was the effect of film thickness and capping layer on the linewidth.

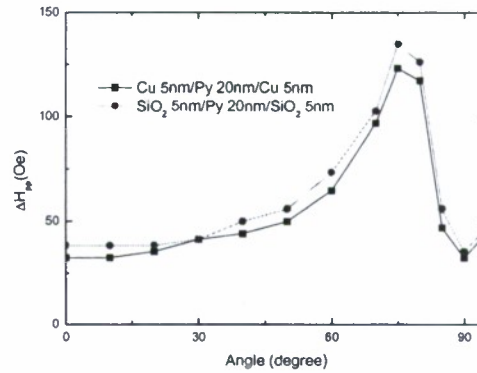
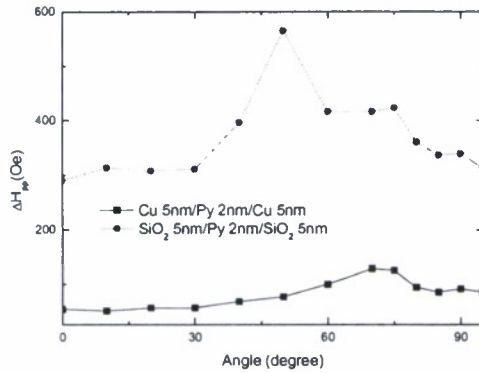


Fig. 4. Angular dependencies of the FMR linewidth for the samples with thickness of 2 nm (a) and 20 nm (b) which were capped with Cu (black curves) or SiO₂ (red curves).

It was found that the linewidth increases with decreasing thickness of the Py film and that the presence of SiO₂ capping layer dramatically increases the damping for the thinnest films with thickness below 10 nm. Therefore, films with the thickness ranging from 20 to 50 nm were selected for the fabrication of the microwave circuits.

Different types of microwave circuits have been tested the most promising design is presented in Fig. 5. The stripline of thin Py film has been patterned using optical lithography, whereas the spinwave emitting and receiving antennas on the top of the Py strip were fabricated by the means of electron lithography.

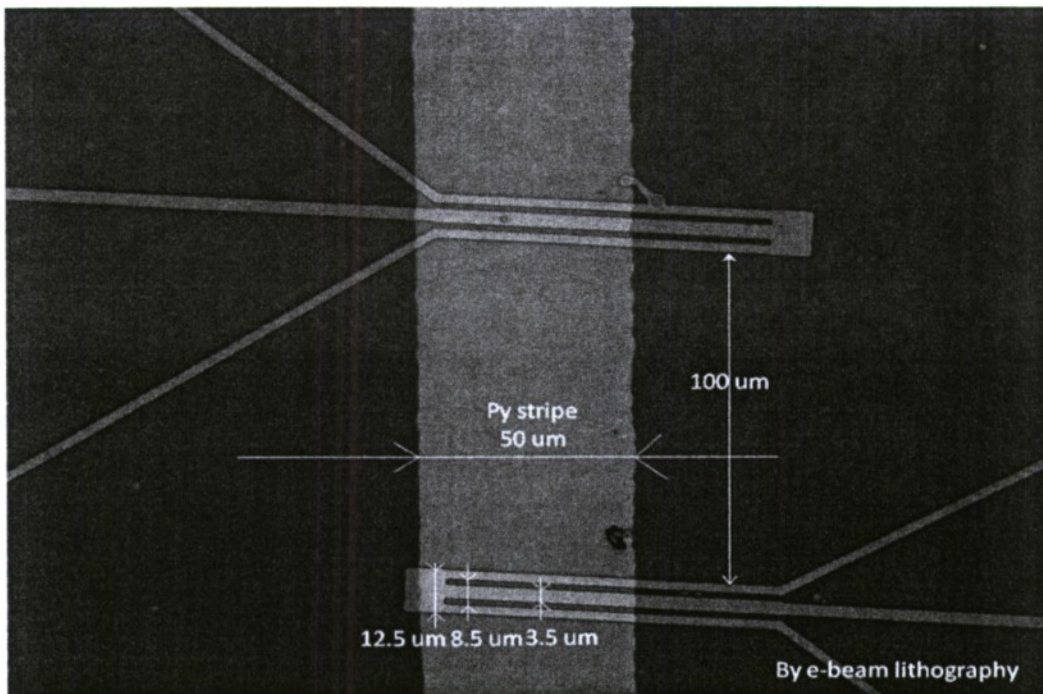


Fig. 5 Scanning field emission electron microscope image of the permalloy stripe and the microwave antennas to generate and detect spinwaves propagating in Py.

Because the model of the Network Analyzer at AMRI is not suitable for calibration procedures, the device presented in Fig.5 is currently under testing at the University of Colorado at Colorado Springs, where this kind of calibration can be performed to reduce stray noise.

4. Publications: None

5. Presentations:

L. Malkinski, "Microwave properties of magnetic nanostructures" Seminar at Tulane University, Department of Physics, October 20, 2009, New Orleans

"Fabrication of and magnetic particles of 2-dimensional arrays of elongated magnetic particles "

Poster presentation at the Conference on Magnetism and Magnetic Materials (Nov 2008) Austin, Texas

"Ferromagnetic resonance of arrays of arrays of nanosized permalloy strips"
Presentation at International Magnetism Conference Sacramento, CA May 2009

"Microwave magnetic properties of NiFe nanostrips"
Presentation at International Magnetism Conference Sacramento, CA May 2009

(4) co-PI: Irinel Chiorescu, Florida State University

Project Name: *Advanced Materials for Quantum Computing*

1. Brief Narrative:

It is proposed to use the NHMFL experimental capabilities to analyze samples that will fit with the research activities of the DARPA grant on Advanced Materials for Quantum Computing. NHMFL provides world-class resources for performing studies in high magnetic fields and high frequencies. We used in the first phase the Bruker pulsed EPR setup working in the X-band, followed by fabrication and measurement techniques developed in my group. We have demonstrated coherent spin manipulation of a Cr ions ($S=1/2$) as well as of the more complex system with Mn ions, $S=5/2$. For the later, precise determination and manipulation of spin Hamiltonian was required. It is important to note that we have developed a 20 GHz spectroscopy cavity for millikelvin studies which allowed the observation, for the first time, of the field-dependent non-linear Meissner effect in a superconductor.

2. Objectives:

The main goal of this subcontract is to probe the quantum spin dynamics in novel magnetic materials, as discussed in the proposal of the DARPA grant on Advanced Materials for Quantum Computing. This will be achieved by using a pulsed EPR technique. Below the main research directions are given as in the original proposal, and in Section 3 their fulfillment (as well as other progresses made) will be discussed:

- 1) Characterization of the intra-molecular interactions creating the anisotropy terms and the final form of the spin Hamiltonian.
- 2) Gain a better understanding of decoherence mechanisms by analyzing absorption line shapes.
- 3) Perform coherent spin manipulation by applying nanosecond pulses in the mm-wave domain.

3. Research Progress:

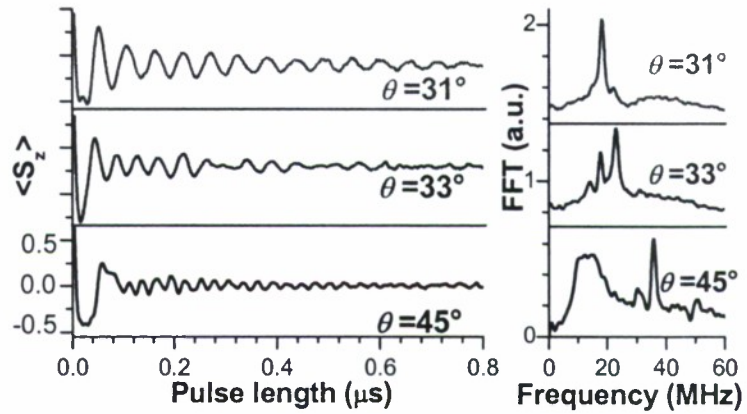
In brief, we (i) performed research aiming to an in-depth understanding of the anisotropic spin Hamiltonian in the case of Mn ions with $S=5/2$ (objective #1); (ii) analyzed the decoherence processes in Cr $S=1/2$ and Mn $S=5/2$ ions and a general study on phonon-bottleneck (objective #2) and (iii) performed spin-photon coherent operations in these two spin systems (objective #3). In addition, this work resulted in the development of a 20 GHz spectroscopy setup and sample deposition methodology for low temperatures, which allowed us to demonstrate for the first time the field-dependent non-linear Meissner effect in a superconductor.

Coherent multi-photon spin manipulation in $S=5/2$ system

We focused a great deal of our research on manipulating the spin Hamiltonian of Mn ion with spins $S=5/2$ diluted in MgO. This choice was motivated by the very low anisotropy due to the symmetry and by the higher value of Mn spin ($S=5/2$). The multi-level system

is almost an harmonic one. A magnetic field applied under a variable angle with the crystal axes acts as an external parameter tuning “in-situ” the harmonicity of the levels. We have performed measurements and developed a model allowing the understanding of this effect. Experimentally, it turns out the Rabi frequency can be effectively tuned by the off-diagonal terms of the spin diagonal, here tuned in-situ by the external field.

Figure 1. From Ref. 1. The Rabi frequency of the Mn spins is recorded for three rotation angles θ of the applied field within the horizontal plane (001). The Fast Fourier Transform of the oscillations is given in the second panel. It is found that for a particular angle (here 31 deg), the system becomes truly harmonic and behaves like a two-level system.



In the Fig. 1, the Rabi frequency is given for three angles between the applied field in the (001) plane and the [100] axis, together with the corresponding Fourier transform (FFT). It is found that at a particular angle (1st graph) the multi-level system shows one Rabi frequency. For an angle just slightly different (2nd curve), the FFT indicates two close Rabi frequencies. Away from this angle (3rd angle), the FFT of the Rabi oscillations indicates a broad peak, probably due to short multi-photon processes. This technique of spin dynamics led to a very sensitive method to observe the role of spin anisotropy on its energy levels and dynamics.

The figure 2 shows Rabi oscillations performed at 30K for three different powers (a) and a contour plot (b) of experimental FFT peaks (red dots) compared with our numerical simulations. The experimental points fit very well with the theory. Panel (c) shows an experimental FFT trace with

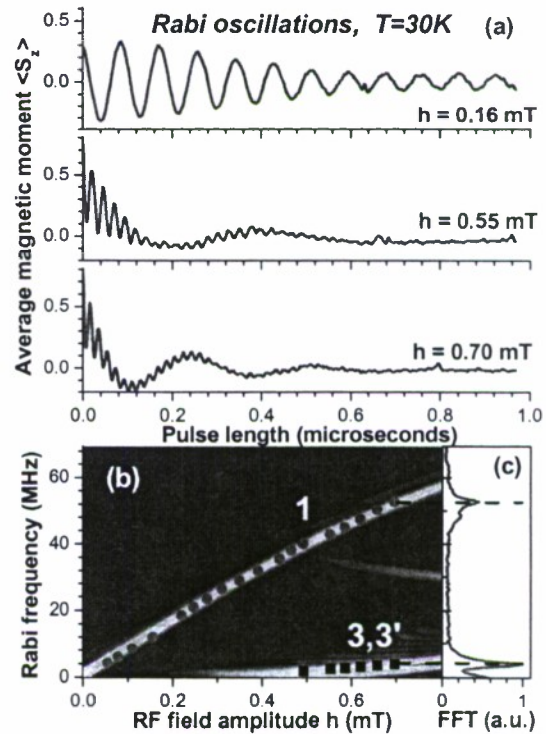


Figure 2. Rabi oscillations at 3 powers (a) and simulated contour plot the FFT of the oscillations (b) compared with the experimental points. An actual FFT trace is shown in (c) indicating single and triple photon Rabi oscillations.

two main peaks corresponding to single and triple-photon driven Rabi oscillations.

Such systems can be a viable alternative to exquisite materials as diamond containing N vacancies.

Controlling decoherence by eliminating nuclear spins, the case of Cr^{5+} $S=1/2$ ions.

In this work, we studied a new material, $\text{Cr}:\text{K}_3\text{NbO}_8$ which could have potential impact in the quantum computing arena. We reported coherent spin manipulation on Cr^{5+} ($S=1/2$, $I=0$) doped K_3NbO_8 , which constitutes a dilute two-level model relevant for use as a spin qubit. The crystalline structure is shown in Fig. 3, atop of recorded coherent spin oscillations. Rabi oscillations are observed for the first time in a spin system based on transition metal oxides up to room temperature (Ref. 2). At liquid helium temperature the phase coherence relaxation time T_2 reaches ~ 10 microseconds and, with a Rabi frequency of 20 MHz, yields a single qubit figure of merit QM of about 500.

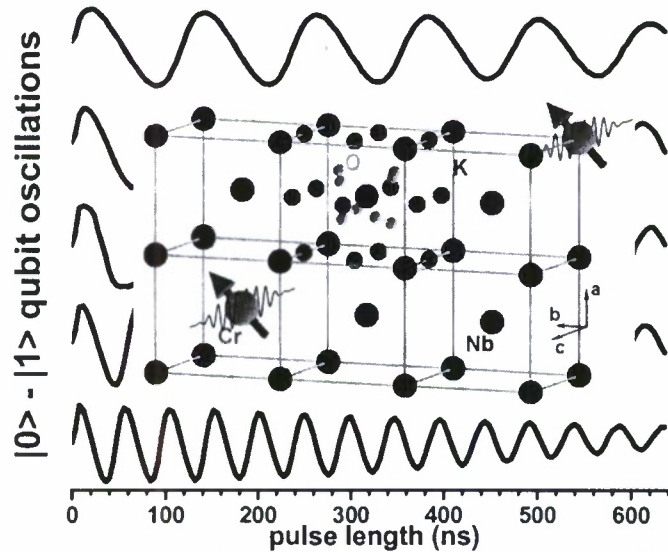


Figure 3. Crystalline structure of K_3NbO_8 containing randomly diluted Cr^{5+} spins ($S=1/2$, $I=0$). The backdrop shows recorded Rabi oscillations of the chromium spins.

This shows that a diluted ensemble of Cr^{5+} is a potential candidate for solid-state quantum information processing. It is important to note that, in contrast to the 2DEG-based quantum dots where the free electron overlaps over the dot's nuclei, the Cr spins interact only with the neighboring non-zero spin nuclei of the crystal, since the Cr does not have nuclear moment (90% natural abundance for $I=0$). This material is almost isotropic and therefore suitable to on-chip applications (doesn't matter how it falls on a chip) and is synthesized through traditional chemistry methods. Further improvements of the material are planned, to further decrease the amount of nuclei carrying magnetic moments. Aside decoherence mechanisms due to nuclear spins, we have studied, on a broader level, how the phonon bottleneck phenomena develop in spin systems with very low energy splitting and the findings have been reported in Ref. 3.

Entrapping micro-crystals for performing on-chip ESR studies

We developed a new method of entrapping micro-crystals containing spins in a well defined location on a substrate's surface. Traditional cavity ESR measurements are than performed on a mesoscopic crystal at 34 GHz. Polycrystalline diluted Cr^{5+} spins were entrapped as well and measured while approaching the lower limit of the ESR sensitivity. The nearby figure 4 (a) shows a photograph of a single crystal of $\text{Cr}:\text{K}_3\text{NbO}_8$ entrapped in the area delimited by the dark contour. The crystal is encased in optical photoresist, as shown by the grey surface of the contour.

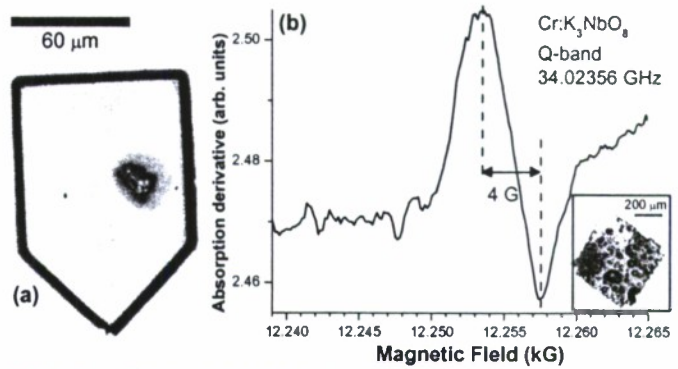


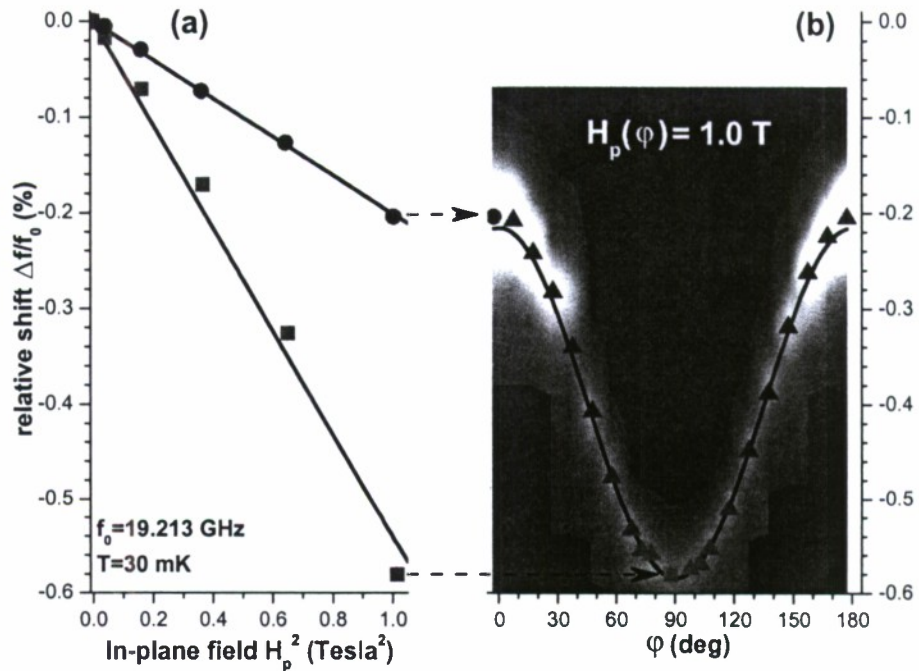
Figure 4. Photograph of a $\text{Cr}:\text{K}_3\text{NbO}_8$ crystal (a) and ESR spectra of an ensemble of crystals (b). From Ref 4.

In panel (b) the room temperature ESR signal of diluted Cr^{5+} spins is shown. The insert shows the actual sample, consisting of few tens of single crystals, in order to achieve a measurable signal by the Q-band spectrometer. The obtained signal is the result of 40 averages, as required by the small size of the signal.

First-time observation of the field dependent Non-Linear Meissner (NLME) effect

We developed a heterodyne-based ESR setup aiming to perform spectroscopy studies in high magnetic fields and low temperatures (in our dilution refrigerator). Using this setup, we reported the first demonstration of the nonlinear Meissner effect (NLME), as observed in a planar superconducting cavity by detection of its resonant frequency dependence of an in-plane magnetic field. The use of low power probing in films thinner than the London penetration depth minimizes vortex penetration and allows NLME manifestation. The coupling between the cavity mode and the Meissner screening currents is modeled classically to describe the findings. Our method probes the macroscopic phase coherence of a condensate and can explore the pairing symmetry in unconventional superconductors.

Figure 5. Relative shift of cavity resonance for in-plane fields along two perpendicular directions (squares and dots) as a function of field. Black lines are through-zero fits. (b) Transmission contour plot as a function of in-plane field rotation (blue: 0 μ W, white: 11 μ W) at constant in-plane field $H_p(\varphi)=1.0$ T. Maxima, shown by symbols, are in excellent agreement with our theoretical curve (black line).



4. Publications:

1. S. Bertaina, L. Chen, N. Groll, J. Van Tol, N.S. Dalal, I. Chiorescu, "Multiphoton coherent manipulation in large-spin qubits", *Physical Review Letters* **102**, 050501 (2009)
2. Nellutla, S.; Choi, K.-Y.; Pati, M.; van Tol, J.; Chiorescu, I. and Dalal, N.S., "Coherent Manipulation of Electron Spins up to Ambient Temperatures in Cr^{5+} ($S=1/2$) doped K_3NbO_8 ", *Phys. Rev. Lett.* **99**, 137601 (2007).
3. L. Chen, I. Chiorescu, "Coherent spin rotation in the presence of a phonon bottleneck effect", submitted
4. N. Groll, S. Bertaina, M. Pati, N.S. Dalal, I. Chiorescu, "Entrapment of magnetic micro-crystals for on-chip ESR studies", accepted for publication in *Journal of Applied Physics*.
5. N. Groll, A. Gurevich, I. Chiorescu, "Observation of the non-linear Meissner effect", to be submitted.

5. Presentations:

- Oral presentation at "The 2nd Workshop on Current trends in Nanoscopic and Mesoscopic Magnetism", September 1-5, 2008 Delphi, Greece.
- Invited talk at the International Conference "Materials for Electrical Engineering", Bucharest - Romania, June 2008
- Invited talk at the Workshop on "Dynamics and manipulation of quantum systems", Tokyo 20-22 October 2008.
- Aspen winter school on Unifying Themes in Condensed Matter (Jan. 2009): Invited presentation and 2 poster presentation of graduate students in our group.
- APS March Meeting in Pittsburgh, 2009: Invited presentation for I. Chiorescu and two oral presentations for graduate students in our group.



ADVANCED MATERIALS RESEARCH INSTITUTE

UNIVERSITY OF NEW ORLEANS

May 3, 2010

Dr. William S. Coblenz
DARPA Program Officer
3701 North Fairfax Drive
Arlington, VA 22203-1714

Re: DARPA Grant No. HR0011-07-1-0031, "Advanced Materials for
Quantum Computing," Final Technical Report, April 28, 2010

Dear Dr. Coblenz:

Enclosed per the requirements of the above referenced Grant is one copy of our Final Technical Report dated April 28, 2010. Should you have any questions, please let me know.

Sincerely yours,

Charles J. O'Connor, Ph.D.
Principal Investigator and
Director of AMRI

CJOC/ird

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