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14. ABSTRACT
We report a nitrogen delta-doping technique to engineer monolayer planes of spin in single crystal diamond films. Isotopically pure 15N2 gas is briefly introduced to form a thin N-doped layer (1-2 nm thick) during chemical vapor deposition of a diamond film. Post growth electron irradiation creates vacancies and subsequent annealing forms nitrogen-vacancy centers (NV)s while mitigating the crystal damage. We identified doped NVs by the characteristic hyperfine signature of the rare 15N isotope in electron spin resonance measurements. We confirm the doped NV depth dispersion is less than 4 nm by doping NVs in the 12C layer of an isotopically engineered 13C/12C/13C layer structure and probing the coupling between the doped NVs and the 13C nuclear spins. Furthermore, using a second Carbon ion-implantation technique, spins have been formed a few nanometers from the surface while preserving the coherence times in excess of 750 microseconds at room temperature. These spin-engineered quantum states have been used both to probe the local crystal strain, as well as serve as sensors for local electric, magnetic, and thermal fields using both electronic and optical control schemes.

15. SUBJECT TERMS
Spintronics, quantum information processing, diamond, chemical vapor deposition, spin engineering, quantum electronics

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SYSTHESIS AND ENGINEERING OF DIAMOND FOR NANOELECTRONICS, PHOTONICS AND QUANTUM INFORMATION PROCESSING

RESEARCH ACCOMPLISHMENTS

AFOSR #FA9550-09-1-0540

- *Engineering spins in diamond with nanometer-scale precision*

The excellent spin properties of the diamond nitrogen-vacancy (NV) center motivate many applications from sensing to quantum information processing. Still, external electron and nuclear spin sensing are limited by weak magnetic dipole interactions, requiring NVs be within a few nanometers of the surface and retain long spin coherence times (T_2). We report a nitrogen delta-doping technique to create artificial NVs meeting both these requirements. Isotopically pure $^{15}N_2$ gas is briefly introduced to form a thin N-doped layer (1-2 nm thick) during chemical vapor deposition of a diamond film. Post growth electron irradiation creates vacancies and subsequent annealing forms NVs while mitigating the crystal damage. We identified doped NVs by the characteristic hyperfine signature of the rare ^{15}N isotope in electron spin resonance measurements. We confirm the doped NV depth dispersion is less than 4 nm by doping NVs in the ^{12}C layer of an isotopically engineered $^{13}C/^{12}C/^{13}C$ layer structure and probing the coupling between the doped NVs and the ^{13}C nuclear spins. Furthermore, using a second Carbon ion-implantation technique, spins have been formed a few nanometers from the surface while preserving the coherence times in excess of 750 μs at room temperature.

- *Epitaxial growth of ferromagnetic EuO on diamond*

We report the epitaxial integration of phase-pure EuO on both single-crystal diamond and on epitaxial diamond films grown on silicon utilizing reactive molecular-beam epitaxy. The epitaxial orientation relationship is (001) EuO ∥ (001) diamond and [110] EuO ∥ [100] diamond. The EuO layer is nominally unstrained and ferromagnetic with a transition temperature of 68 ± 2 K and a saturation magnetization of 5.5 ± 0.1 Bohr magnetons per europium ion on the single-crystal diamond, and a transition temperature of 67 ± 2 K and a saturation magnetization of 2.1 ± 0.1 Bohr magnetons per europium ion on the epitaxial diamond film.

- *Fluorescence thermometry using the quantum coherence of single spins in diamond*

We demonstrate fluorescence thermometry techniques with sensitivities approaching 10 mK·Hz−1/2 based on the spin-dependent photoluminescence of nitrogen vacancy (NV) centers in diamond. These techniques use dynamical decoupling protocols to convert thermally induced shifts in the NV center's spin resonance frequencies into large changes in its fluorescence. By mitigating interactions with nearby nuclear spins and facilitating selective thermal measurements, these protocols enhance the spin coherence times accessible for thermometry by 45-fold, corresponding to a 7-fold improvement in the NV center’s temperature sensitivity. Moreover, we demonstrate these techniques can be applied over a broad temperature range and in both finite and near-zero magnetic field environments. This versatility suggests that the quantum coherence of single spins could be practically leveraged for sensitive thermometry in a wide variety of biological and microscale systems.

- *All-optical control of a single solid-state spin using coherent dark states*

The study of individual quantum systems in solids, for use as quantum bits (qubits) and probes of decoherence, requires protocols for their initialization, unitary manipulation, and readout. In many solid-state quantum systems, these operations rely on disparate techniques that can vary widely depending on the particular qubit structure. One such qubit, the nitrogen-vacancy (NV) center spin in diamond, can be initialized and read out through its special spin-selective intersystem crossing, while microwave electron spin resonance techniques provide unitary spin rotations. Instead, we demonstrate an alternative, fully optical approach to these control protocols in an NV center that does not rely on its intersystem crossing. By tuning an NV center to an excited-state spin anticrossing at cryogenic temperatures, we use coherent population trapping and stimulated Raman techniques to realize initialization, readout, and unitary manipulation of a single spin. Each of these techniques can be performed directly along any arbitrarily chosen quantum basis, removing the need for extra control steps to map the spin to and from a preferred basis. Combining these protocols, we perform measurements of the NV center's spin coherence, a demonstration of this full optical control. Consisting solely of optical pulses, these techniques enable control within a smaller footprint and within photonic networks. Likewise, this unified approach obviates the need for both electron spin resonance manipulation and spin addressability through the intersystem crossing. This method could therefore be applied to a wide range of potential solid-state qubits, including those which currently lack a means to be addressed.

HONORS AND AWARDS DURING CURRENT PERIOD

- Bethe Lecturer, Cornell University (2014)
- Marker Lecturer, Pennsylvania State University (2014)
- Nobel Week Dialogue Panelist, Sweden (2013)
- Gothenburg Mesoscopic Lecturer, Chalmers, Nobel Institutes of Physics, Sweden (2013)
- Slichter Lecturer, University of Illinois (2013)
- Elected, European Academy of Sciences (2013)

Publications

1. "Epitaxial growth of europium monoxide on diamond," A. Melville, T. Mairoser, A. Schmehl, M. Fischer, S. Gsell, M. Schreck, D. D. Awschalom, T. Heeg, B. Holländer, J. Schubert, J. Mannhart, and D. G. Schlom. *Appl. Phys. Lett.* **103**, 222402 (2013)

2. "Quantum computing with defects," L. Gordon, J. R. Weber, J. B. Varley, A. Janotti, D. D. Awschalom, and C. G. Van de Walle, *MRS Bulletin* **38**, 802 (2013)

3. "Fluorescence thermometry enhanced by the quantum coherence of single spins in diamond," D. M. Toyli, C. F. de las Casas, D. J. Christle, V. V. Dobrovitski, and D. D. Awschalom, *Proc. Natl. Acad. Sci. USA* **110**, 8417 (2013)

4. "All-optical control of a solid-state spin using coherent dark states," C. G. Yale, B. B. Buckley, D. J. Christle, G. Burkard, F. J. Heremans, L. C. Bassett, and D. D. Awschalom, *Proc. Natl. Acad. Sci. USA* **110**, 7595 (2013)

5. "Quantum Spintronics: Engineering and Manipulating Atom-Like Spins in Semiconductors," D.D. Awschalom, L.C. Bassett, A.S. Dzurak, E.L. Hu and J.R. Petta, Invited Article, *Science* **339**, 1174 (2013)

6. "Quantum Control over Single Spins in Diamond," V.V. Dobrovitski, G.D. Fuchs, A.L. Falk, C. Santori and D.D. Awschalom, Invited Article, *Annu. Rev. Condens. Matter Phys*. **4**, 23 (2013)

7. "Engineering and quantum control of single spins in semiconductors," D.M. Toyli, L.C. Bassett, B.B. Buckley, G. Calusine and D.D. Awschalom, *MRS Bulletin* **38**, 139 (2013)

8. "Nanoscale Nuclear Magnetic Resonance with a Nitrogen-Vacancy Spin Sensor," H. J. Mamin, M. Kim, M. H. Sherwood, C. T. Rettner, K. Ohno, D. D. Awschalom, and D. Rugar, *Science* **339**, 557 (2013)

Invited Talks

1. D.D. Awschalom, Hans Bethe Lectures, Cornell University, Ithaca, NY, April 7-11, 2014. "Beyond Electronics: Abandoning Perfection for Quantum Technologies", Physics Colloquium, April 7, 2014; "Ultrafast quantum control of single electron orbital and spin dynamics in diamond", Physics Seminar, April 8, 2014; "Engaging Diamonds in the Quantum Age", Public Lecture, April 9, 2014.

2. D.D. Awschalom, Marker Lectures, Pennsylvania State University, University Park, PA, March 26-28, 2014. "Engaging Diamonds in the Quantum Age", Public Lecture, March 26, 2014; "Beyond Electronics: Abandoning Perfection for Quantum Technologies", Physics Colloquium, March 27, 2014; "Ultrafast quantum

control of single electron orbital and spin dynamics in diamond”, Physics Seminar, March 28, 2014.

3. D.D. Awschalom, “Quantum spintronics”, The International Meeting on Spintronics for Integrated Circuit Application and Beyond, Tokyo, Japan, March 13, 2014.

4. D.D. Awschalom, “Mobile electron spin resonance with spins in optically-trapped nanodiamonds”, Symposium on Exploring the Foundations of Magnetism with New Nanoscale Probes, Annual Meeting of the American Association for the Advancement of Science, Chicago, IL, February 13-17, 2014.

5. D.D. Awschalom, “Ultrafast quantum control of single electron orbital and spin dynamics in diamond”,International Symposium on Topological Quantum Technology, University of Tokyo, Tokyo, Japan, January 27-30, 2014.

6. D.D. Awschalom, "Beyond electronics: abandoning perfection for quantum technologies", Physics Colloquium, University of Chicago, Chicago, IL, January 16, 2014.

7. D.D. Awschalom, “Beyond electronics: abandoning perfection for quantum technologies“, Goteborg Mesoscopic Lecture, Royal Swedish Academy of Sciences & Nobel Institutes of Physics, Chalmers University, Goteborg, Sweden, December 6, 2013.

8. D.D. Awschalom, Experimentalist of the Week, “Abandoning perfection: quantum control of defects for sensing and information processing”, KITP Program on Spintronics: Progress in Theory, Materials, and Devices, Kavli Institute for Theoretical Physics, Santa Barbara, CA, November 18-22, 2013.

9. D.D. Awschalom, “Beyond electronics: abandoning perfection for quantum technologies”, Physics Colloquim, Northwestern University, Evanston, IL, November 18, 2013

10. D.D. Awschalom, “Beyond electronics: abandoning perfection for quantum technologies”, Materials Science Special Colloquium, Argonne National Laboratory, Illinois, October 3, 2013

11. D.D. Awschalom, D.D. Awschalom, “Quantum control of single spins in semiconductors for sensing and information processing”, Ultrafast Magnetism Conference, Strasbourg, France, October 28-November 1, 2013.

12. D.D. Awschalom, “Beyond electronics: abandoning perfection for quantum technologies”, Munich Center for NanoScience Workshop on Nanosciences: Great

Adventures on Small Scales, Venice International University, Venice, Italy, September 16-20, 2013.

13. D.D. Awschalom, Keynote Speaker, “Beyond electronics: abandoning perfection for quantum technologies”, Swiss Nanoconvention, Basel, Switzerland, May 23-24, 2013.

14. D.D. Awschalom, Slichter Colloquium, “Beyond electronics: abandoning perfection for quantum technologies Department of Physics, University of Illinois, Urbana, IL, April 17, 2013.

15. D.D. Awschalom, “Mobile quantum sensing with spins in optically trapped nanodiamonds”, March Meeting of the American Physical Society, Baltimore, MD, March 18-22, 2013.

16. D. D. Awschalom, “Quantum Information Processing with Silicon Carbide Spin Qubits”, WPI-AIMR Symposium, Sendai, Japan, February 18-21, 2013.

17. D.D. Awschalom, “Breaking the Wall of Traditional Electronics”, International Conference on Future Breakthroughs in Science and Society, Falling Walls Conference, German Ministry of Education, Berlin, Germany, November 9, 2012.

18. D.D. Awschalom, “Engaging spins in semiconductors for quantum information processing”, Sixth International Conference on Spontaneous Coherence in Excitonic Systems (ICSCE 6), Stanford University, Palo Alto, CA, August 27^{-}31^{s}, 2012.

19. D.D. Awschalom, “Engaging single spins in semiconductors for quantum information processing”, Seventh International Conference on Physics and Applications of Spin-related Phenomena in Semiconductors, Eindhoven University of Technology, The Netherlands, August 5-8, 2012.

20. D.D. Awschalom, “Quantum control of single spins in diamond and SiC”, Low Energy Electrodynamics of Solids conference (LEES-2012), Napa, California July 22-27, 2012.

21. D.D. Awschalom, “Quantum control of single spins in diamond and SiC”, 19th International Conference on Magnetism, Busan, Korea, July 8-13, 2012.