Final Report for ONR grant award N00014-17-1-2785

Controlling Atomic-Scale Quantum Magnetic Structures

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Overview

This grant supports research in quantum magnetism of single-atom spins, magnetic sensing, and pulsed spin resonance using a scanning tunneling microscope. IBM has demonstrated major progress in all three sets of goals during the 2017–2020 funding period. This progress is documented in twelve publications in high-impact journals, and summarized below.

This effort is basic exploratory research in the electronic and magnetic properties of atoms, molecules and nanostructures that are positioned or assembled on surfaces with atomic-scale precision. This work employs low-temperature scanning tunneling microscopy (STM), together with the technique of single-atom electron spin resonance (ESR) using STM. The experiments were performed using atoms deposited on two atomic layers of MgO, which provides controlled isolation from the substrate and a well-ordered array of binding sites.

The proposed work falls in three main categories: (1) Exploring single-atom spins, in which our spin resonance method is improved in capability, extended to apply to new atoms species, and exploited for probing the magnetic character of atoms on surfaces. (2) Magnetic and sensing and imaging at the atomic scale, to determine the positions and orientations of spins in nanostructures and molecules. (3) Developing control over the quantum state of atomic spins by using pulsed spin resonance techniques, in order to use them as qubits in quantum information and sensing applications.

Following IBM Almaden's demonstration of ESR in STM in 2015 [Bau15][Pau16], we have gone on to achieve the following under ONR support:

- Measurement of magnetic dipole interaction between atoms positioned on a surface [Cho17].
- Sensing the magnetic field of bistable magnets formed from single Ho atoms [Nat17].
- ESR of spin-1/2 Ti atoms to observe their dipole and exchange interactions [Yan17].

- Modeling and controlling the energy relaxation [Pau17] and coherence times [Wil18a].
- Sensing the hyperfine interaction [Wil18b] and its use to hyperpolarize the nuclear spin in Cu atoms [Yan18].
- Controlling [Yan19a] and imaging [Wil19a] interaction between adatoms and a magnetic tip.
- Pulsed spin resonance for direct observation of Rabi oscillation and spin echoes [Yan19b].
- Use of strongly-coupled spin-1/2 atoms to observe long-coherence clock transitions [Bae18] and highly entangled states in spin arrays [Yan21].

Results and publications for the 2017–2020 funding period

The following sections summarize our main results and publications supported by this grant.

1. Magnetic dipole sensing [Cho17]

Each spin resonant atom shows an ESR frequency that is proportional to the magnetic field at its location, making it a sensitive detector of the local magnetic environment. We calibrated the moment of Fe atoms to high precision ($5.44 \pm 0.03\ \mu_B$) by measuring the ESR splitting of Fe pairs assembled at precise spacings. The $1/r^3$ dependence indicates magnetic dipole coupling, with rigid out-of-plane orientation due to the large anisotropy when bound to oxygen in MgO. Having calibrated the Fe atom, it was then used as a sensor to measure moments of other spins, such as Co atoms on MgO ($5.88\ \mu_B$). These moments are close to the free-atom value of $6\ \mu_B$ for both Fe and Co, which indicates that Fe and Co have largely unquenched orbital moments on this surface despite the ligand field from bonding to an O atom of the MgO surface. This out-of-plane easy axis anisotropy makes Fe sensitive to the out-of-plane component of the magnetic field.

We showed that precise Fe structures can be assembled on MgO, and that ESR spectra of Fe sensors can be used to precisely locate an unknown moment, by using "nano GPS", in which positions are calculation in a manner analogous to the Global Positioning System. In a proof-of-concept demonstration, we determined the moment and location of a "target" Fe spin, treated as an unknown, by analyzing the ESR spectra of "sensor" Fe atoms positioned a few nm away. This method should allow determination the magnetic structure of unknows such as magnetic molecules. Our initial experiments in probing a molecule having two spin centers, $Tb_2ScN@C_{80}$, were hampered by pinning of the molecule at step edges; ESR spectra using a single Fe sensor showed magnetic two-state switching in the molecule, which is an encouraging step.

2. Spin relaxation in Fe atoms [Pau17]

Bistability in nanomagnets has long been a goal of the surface science and molecular magnet communities. The large anisotropy of transition metal atoms on insulating films such as Cu_2N and MgO yields long relaxation times T_1, sometimes exceeding a microsecond. We explored the limits of Fe adatom lifetimes by using electrical pump-probe measurements to observe T_1 as a function of tunnel current, voltage, and magnetic field [Pau17]. The ~14 meV anisotropy barrier of Fe on this surface gives it an energy relaxation time of 20 ms for the best conditions obtained. This T_1 is long enough to observe in real time as two-state switching in the tunnel current, which is possible because ~10000 electrons are required to relax the spin through the barrier. This long T_1 occurs for large out-of-plane magnetic fields of several tesla. (Note that for ESR studies, smaller fields are used to obtain ESR frequencies in the range 5–35 GHz that is accessible to us. The smaller field allows the ground states to mix, shortening T_1 to ~100 μs and enabling ESR transitions.) We found that each atomic layer of MgO reduces tunneling conductance by a factor of ~10, and spin scattering by a factor of $\sim 10^2 = 100$ since electrons from the substrate must pass twice through the layer in order to relax the spin. At currents exceeding about 10 pA the tunnel current is the dominant cause of relaxation.

We expect that coherence times T_2 will follow a pattern similar to energy relaxation times T_1, where each tunneling electron or conduction electron impinging on the atomic spin has a cross section for causing energy relaxation, and a cross section for decoherence. Thus we expect that increased coherence is possible by using thicker MgO and low tunnel current. Preliminary experiments (unpublished) show that ESR is possible for Fe and Ti on thicker (3 monolayer) MgO. Further experiments will be needed to determine when other relaxation mechanisms, such as coupling to nuclear spins and phonons, limit coherence times.

3. Ho atoms as bistable magnetic bits [Nat17]

It has long been a goal to determine how small a magnetic structure can show bistability, and previous results showed magnetic bistability in as few as eight Fe atoms at 4 K. We found that single Ho atoms on MgO show magnetic bistability at 4 K due to their large magnetic anisotropy [Nat17]. We showed the bistability of individual Ho by sensing the dipole field using ESR in nearby Fe atoms. The Ho moment was 10.0 μ_B, which matches the moment of a free Ho^{+3} ion in $4f^{10}$ configuration. The Ho could also be read using a small (3%) magnetoresistance signal, and written by using current pulses from the STM tip. The Ho atoms remain stable even when placed as close as 1 nm apart, showing ultra-dense magnetic memory bits are physically possible at low temperature.

4. ESR of titanium atoms [Yan17]

In [Yan17] we showed that individual Ti atoms on MgO are versatile spin resonant centers. In contrast to the large spin of Fe, the Ti has spin $S = 1/2$, with a moment ~0.9 μ_B. These Ti atoms are hydrogenated, so they may be described as TiH molecules, but we refer to them as Ti for simplicity. We found that bare (unhydrogenated) Ti atoms have spin $S = 1$ and a ~2 meV magnetic anisotropy energy, which prevented us from using this bare atom for ESR.

Ti adsorbs on both the oxygen-top sites (Ti_O) and on bridge sites between two oxygen atoms (Ti_B), both being spin resonant with ~0.9 μ_B. This provides us with a fine grid of binding sites for building coupled-spin structures. The unquenched orbital moment slightly reduces the moment from 1 μ_B and introduces g-factor anisotropy, but viewing these as free spin-1/2 centers provides a good starting point for understanding their quantum behavior.

Two Ti atoms couple by exchange interaction when the spacing is less than ~1 nm, and magnetic dipole coupling dominates for larger distances. When the Zeeman energy of two Ti atoms is matched closely enough, we found that the spin-1/2 character allows the $|01\rangle$ and $|10\rangle$ states mix to form singlet ($|01\rangle - |10\rangle$) and triplet ($|01\rangle + |10\rangle$) states. Here 0 represents spin up and 1 represents spin down. This mixing allows more transitions to be detected, and entanglement of the spins to be explored. This subject is investigated more deeply in [Bae18] where increased T_2 is found for singlet triplet "clock" transitions, and in [Yan21] where arrays are explored.

5. Singlet-triplet clock transitions for increased coherence [Bae18]

In [Bae18] we demonstrate the increase in T_2 that is obtained by using singlet-triplet transitions in a pair of strongly-coupled spin-1/2 Ti atoms. For large enough antiferromagnetic exchange coupling J between two Ti spins, the singlet state becomes the ground state even at the ~1 T field we used to give the desired Zeeman splitting. The singlet-triplet transition can be driven in the STM and detected by means of homodyne detection, which appears simply as an asymmetric line shape instead of a peak in the spectrum. That transition is forbidden in traditional ESR, where a uniform RF field is applied to both atoms. We apply the driving field to just one atom of the pair, making new transitions available. Since the singlet-triplet transition is very insensitive to uniform applied fields, it provides a "clock" transition that gives much increased coherence time T_2 compared to a single-spin qubit. This energetic benefit to forming a singlet state is the basis for more complex entangled states in spin arrays, explored in [Yan21] and discussed in section 12.

A future goal is to drive ESR of two spins that are only weakly coupled, so they may be considered separate qubits: their resonant frequencies perturb each other, but the states remain Zeeman product states. We have learned that this will require a new mechanism for driving the spin that is not in the tunnel junction. We did preliminary (unpublished) tests to show that ESR can be driven by the tip's electric field together with the magnetic field gradient of a neighboring atom on the surface. We are hopeful that this "lateral" driving mechanism will enable multiple resonance in the near future.

6. Probing and controlling coherence [Wil18a]

The coherence properties of Fe atoms were explored in depth in [Wil18a] to determine the limitations and the available improvements in T_2. We found that nearly every tunneling electron decoheres the Fe atom's spin, even though the anisotropy barrier requires ~100 electrons for energy relaxation. For low enough current (< 10 pA) the coherence time $T_2 \approx 50$ ns is set by scattering from substrate conduction electrons that tunnel through the 2 ML MgO film. At higher currents the tunneling electrons are the main cause of decoherence and relaxation of the Fe spin. This knowledge provides a means to choose between maximizing T_2, and consequently giving the highest ESR energy resolution, or sacrificing T_2 to obtain higher current for larger ESR signals and rapid spectra. This study also showed that one can use spin transfer torque to polarize the Fe spin, so we do not need to rely on spontaneous thermalization of the Fe to drive the spin toward the ground state. This spin torque arises from the spin-polarized tunnel current from the tip, and allows us to work at much larger currents, and consequently obtain better ESR signal-to-noise ratios, at the expense of some energy resolution. It also allows us to obtain ESR spectra at up to 4 K (higher than our usual ~1 K working temperature) so other groups that do not have access to lower temperature can also make use of ESR in STM.

7. Hyperfine coupling in Fe and Ti [Wil18b]

Nuclear spins couple to the electronic spins through hyperfine coupling, and appear as a splitting of the ESR spectra into a separate peak for each nuclear spin state. The most abundant Fe and Ti isotopes have zero nuclear spin, but a few percent have larger spins. We observed two peaks for ^{57}Fe (nuclear spin $I = 1/2$) that are equal height and split by 230 MHz, determined by the hyperfine interaction energy in this chemical environment. This equal occupation is a consequence of negligible Zeeman energy, leading to nearly equal Boltzmann occupation at our 0.6 K operating temperature.

For Ti atoms, the isotope ^{47}Ti has nuclear spin $I = 5/2$, so the ESR spectra have 6 peaks, and ^{49}Ti has $I = 7/2$, giving 8 peaks. Since the Ti is hydrogenated, we expect the hydrogen atom to give a splitting of ~1–10 MHz, which is close to the threshold of our sensitivity, but we have not yet seen the effect of this proton unambiguously.

We used the STM tip and voltage pulses to move the Ti spins between oxygen and bridge binding sites. The hyperfine interaction varied strongly depended on the binding site. Ti_B showed a splitting of ~47 MHz. In contrast, for Ti_O the splitting was only ~10 MHz, a result of partial cancellation of the Fermi contact and dipole contributions to the hyperfine interaction. The spectra provide information about the electronic ground state, the atomic state mixing due to neighboring atoms, and properties of the nuclear spin such as the nuclear quadrupole moment.

8. Hyperfine coupling in Copper [Yan18]

We found that copper atoms on MgO have net spin $S = 1/2$ due to their $4s^2 3d^9$ configuration, which leaves one unpaired spin. This provides us with the third element shown to be accessible to ESR in STM. Copper has extraordinarily large hyperfine coupling because its 4s orbital has strong Fermi overlap with the nucleus, and these 4s electrons are significantly spin polarized by interaction with the 3d electrons. We found that spin pumping the electronic spin of Cu resulted in strong nuclear spin polarization, known as hyperpolarization. The mechanism is the conservation of angular momentum: an electron spin flips and a nuclear spin flops. The direction and magnitude of the nuclear polarization is controlled by the direction and amplitude of the current.

The strong nuclear polarization permitted us to detect NMR transitions in individual Cu atoms, the first demonstration of NMR in and STM. These NMR transitions were used to sense the local magnetic environment of the Cu electron spin such as the proximity of the STM tip.

9. Scanning ESR for magnetic imaging [Wil19a]

It is a long-standing goal to have a spin-resonant sensor to measure the 3-dimensional pattern of the magnetic field near a surface. As a step toward this goal, we explored the interaction of a known spin-resonant atom on the surface interacting with the unknown moments on the disordered STM tip. To do this we scanned the tip in a 3D raster pattern and mapped out the resonant energy of the surface atom for both Fe and Ti. We found that coupling is dominated by dipole coupling for some tips, and exchange coupling for other tips. Thus, the surface adatom serves as a scanning ESR sensor to image the tip field with sub-Angstrom resolution. Simple models of the tip moments qualitatively capture the spin pattern. A lesson here is that the disordered tip is complex and may have multiple spins, induced spins, and buried spins that are revealed by the ESR atom. This provides a proof-of-principle that scanning ESR in an STM can be a potent tool. In the future we would like to reverse the roles of tip and surface: placing a spin resonant sensor on the tip would allow reproducible surface structures to be imaged.

10. Tip-adatom coupling [Yan19a]

The exchange coupling between the magnetic tip and a Ti spin on the surface was explored for a wide range of coupling energies [Yan19a]. We varied the position of a magnetic tip above a Ti spin on the surface to demonstrate the precise control of the exchange bias experienced by the Ti spin. This measurement covered an energy range of 4 orders of magnitude by adjusting the height of the STM tip. We combined ESR with inelastic electron tunneling spectroscopy (dI/dV) to seamlessly map out the different energy scales, which ranged from an effective tip field of 1 mT to 10 T. Coupling was exponential over the range covered, except at greatest tip height, where the crossover to dipole coupling begins. This control of exchange bias over a wide span provides a means to control quantum state properties in arrays, and gives tunable exchange bias for local spin doping in arrays. This study gives quantitative field gradient measurements, which sheds light on the ESR driving mechanism in STM. We confirmed that the driving results from the electric-field driven motion of the atomic spin in the strong exchange field gradient of the tip.

11. Toward qubits: Pulsed spin resonance [Yan19b]

Pulsed spin resonance treats the spins as qubits by using a sequence of radio-frequency (RF) pulses to Rabi rotate a spin to a desired quantum state [Yan19b]. This gives more control and information than the continuous-wave (CW) ESR used thus far, which yields the resonant frequencies and line widths. We demonstrated standard pulsed sequences to observe Rabi oscillation, Ramsey fringes, and spin echoes in individual Ti atoms, and in coupled spin systems made from pairs of Ti atoms.

The decay time of the Rabi oscillation gives a direct measure of T_2*, which is the effective coherence time when subject to inhomogeneous broadening, which is a consequence of measuring the average over an ensemble of spins. Even though only one spin is being probed in our expriments, ensemble broadening still takes place because the spectra give an average over many sequential measurements, repeated rapidly, in order to obtain sufficient signal. Each measurement is slightly different due to fluctuating magnetic fields from nearby spins, and from unintended movement of the magnetic tip. We found $T_2^* \approx 50$ ns for Ti atoms in the limit of low tunnel current. A spin echo measurement removes most the effect of inhomogeneous broadening by cancelling slowly fluctuating fields. We performed spin echo measurements and found an increased coherence time $T_2 \approx 190$ ns.

We further demonstrate coherent operations on engineered atomic dimers to show that pulsed spin resonance is accessible even in coupled multiple-spin structures.

12. Entangled magnetic states in assembled arrays [Yan21]

We constructed artificial quantum magnets by assembling 3–4 Ti atoms into arrangements such as linear and square patterns. These coupled spins feature strong quantum fluctuations due to the antiferromagnetic exchange interactions between neighboring atoms. The low spin of Ti permits the fluctuations because the exchange interaction can swap the spins. In contrast, large spins such as Fe tend to have Ising type interaction due to the magnetic anisotropy that prefers one axis. We performed ESR on individual spins within each quantum magnet in order to characterize the collective states and transition energies. This gives atomic-scale access to properties of a resonating valence bond (RVB) state, a particular kind of spin liquid. The tunable atomic-scale magnetic field from the STM tip allows us to further characterize and engineer the quantum states. These results present a new method explore quantum magnets at the atomic scale for use as a tunable quantum simulator and as a testbed for quantum magnetic materials.

Publications supported by this ONR Grant

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Status Report for proposed Goals, Milestones, and Tasks

Quantum magnetism of single-atom spins

Goal 1: Measure magnetic properties of new atom species
Milestone: Demonstrate increased spin lifetimes for individual magnetic atoms on a surface.
Achieved: Ho atoms show long-term magnetic stability as seen by an ESR sensor atom [Nat17].
Technical Task: Deposit atoms of candidate elements on atomically clean thin insulating surfaces and characterize their binding sites and magnetic properties.
Achieved: Ti characterized on oxygen and bridge sites with and without attached H [Yan17].

Goal 2: Develop theoretical model for quantum magnetism of the atoms
Task: Develop and extend models of quantum magnetism of atoms on surfaces to apply to new atoms.
Achieved: Modeling of Fe [Pau17], hydrogenated Ti [Yan17] and Cu spins [Yan18] describes dI/dV and ESR spectra, coupling, and field dependence. Nuclear spin including nuclear quadrupole moment and binding site dependence also modeled quantitatively.

Goal 3: Determine the mechanisms of electric-field-driven spin resonance in STM
Task: Evaluate new candidate atom species for spin-resonance performance.
Achieved: Driving by piezoelectric motion in tip's exchange field characterized [Yan19a].

Goal 4: Seek improved spin resonance by exploring different atom species
Milestone: Demonstrate improved spin resonance by using different atom/surface combinations.
Achieved: Understanding T_2 of Fe: dependence on I, V, T, and spin torque [Wil18a].
Longer T_2: obtained by using echo techniques with Ti atoms [Yan19].

Goal 5: Measure spin resonance of atoms on thicker insulating layers
Some progress: ESR spectra of Fe (both sites) on 3 ML of MgO obtained [unpublished].

Magnetic sensing and imaging

Goal 6: Remotely sense the magnetic properties of molecules and nanostructures
Milestone: Construct atomically precise arrangements of sensor atoms for probing molecule's magnetism.
Technical Task: Implement spectrum deconvolution method for analyzing spectra of multi-spin targets.
Some progress: Proof of principle [Cho16] and single Fe sensing of $Tb_2ScN@C_{80}$ [unpublished].

Goal 7: Precise atom manipulation of new atom species
Task: Extend atom manipulation capabilities to apply to new atom/surface combinations.
Achieved: Manipulation of Ti at two binding sites (oxygen top and bridge) to form precise structures [Yan17][Bae18][Yan19b][Yan21][Nat17]. Manipulation of Ho atoms [Nat17].

Goal 8: Determine the position of the spin centers by remote sensing of their magnetic fields

Milestone: Image the magnetic structure within individual magnetic molecules by remotely sensing their magnetic fields.

Some Progress: Ho atoms probed by single Fe sensors show $1/r^3$ distance dependence [Nat17].

Goal 9: Magnetic imaging of multiple spin centers

Milestone: Image the magnetic structure of a molecule or nanostructure having more than one spin center.

Some Progress: for atoms: Imaging spin resonant atoms using non-resonant tip [Wil19a].

Minimal progress: for molecules: Multiple spin centers within a molecule not yet resolved due to difficulty of controlling $Tb_2ScN@C_{80}$ position near Fe sensors [unpublished].

Single-atom quantum bits

Goal 10: Use radio-frequency pulses to control the quantum state of an atom

Task: Setup instrumentation for high-frequency pulsed experiments to control qubit states.

Milestone: Demonstrate one-qubit gates by using pulsed spin resonance of single atoms.

Achieved: Pulsed spin resonance of Fe and Ti_O to show Rabi oscillation [Yan19].

Goal 11: Implement general single-qubit gates

Task: Implement pulsed spin resonance techniques and measure Rabi-oscillation signal

Milestone: Evaluate single-qubit gate fidelity by performing quantum state tomography.

Achieved: Ramsey fringes in Ti with detuning show effect of X and Y gates [Yan21].

Some progress: States inferred from Ramsey [Yan21] but full tomography requires greater T_2.

Goal 12: Implement echo techniques and measure the resulting improvement in coherent properties

Task: Implement spin echo techniques and evaluate improvement in T_2

Achieved: Spin echoes in Ti show T_2 improvement [Yan21].

Goal 13: Control two or more spin resonant atoms together

Task: Setup for double-resonance experiments by implementing two-frequency driving.

Some progress: ESR of 2–4 coupled spins driven at a single frequency [Yan17][Bae18][Yan19].

Some progress: Electron-nuclear double resonance observed in a single Cu atom [Yan18].

Further progress in areas closely related to the ONR proposal

- **Hyperfine coupling**

Achieved: Observe hyperfine coupling in ESR for Fe and Ti atoms; employ it as a sensor for local binding chemistry [Wil81b].

Achieved: Use hyperfine coupling to polarize a nuclear spin; observe NMR transitions [Yan18].

- **Entangled Spin Arrays**

Achieved: Driving and sensing of singlet-triplet clock transitions in tuned dimers [Bae18].

Achieved: Resonating Valence Bond states observed in spin-1/2 arrays [Yan21].

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