4. TITLE AND SUBTITLE
Classical and Quantum Properties of Magnetic Nanostructures

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13. ABSTRACT (Maximum 200 words)
The reversal mechanisms in arrays of STM-fabricated nanometer-scale iron particles were studied by low temperature integrated ZDEG Hall magnetometry and room temperature magnetic force microscopy. Initially, the magnetic properties of the STM particles were studied in ensembles at low temperature.

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FINAL TECHNICAL REPORT

"Classical and Quantum Properties of Magnetic Nanostructures"
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1. Summary of technical accomplishments

- Imaging and Magnetometry of Magnetic Nanostructures
  a) Arrays of STM-fabricated magnets

The reversal mechanisms in arrays of STM-fabricated nanometer-scale (< 40 nm diameter) iron particles were studied by low temperature integrated 2DEG Hall magnetometry and room temperature magnetic force microscopy. Initially, the magnetic properties of the STM particles were studied in ensembles at low temperature. By complementary low temperature Hall magnetometer and room temperature magnetic force microscope (MFM) measurements, the average magnetic properties of an array of particles are now compared with the properties of individual particles. The magnetic properties of the arrays of iron particles are studied at low temperatures (< 100 K) with a Hall magnetometer fabricated from a Be modulation doped GaAs/Al$_x$Ga$_{1-x}$As heterostructure grown by molecular beam epitaxy. An array is grown 100 nm above the (2.5 μm)$^2$ square active area of a Hall cross. The sensitivity of the Hall magnetometer depends on the placement of the magnets with respect to the active area of the Hall cross. Tilting of the magnetic moments can lead to negative flux coupling from dipoles at an edge of the active area and hysteresis loops which are difficult to interpret. Therefore, all measurements are performed with the external field applied perpendicular to the active area of the Hall magnetometer (henceforth known as the vertical direction). The observed noise level is equivalent to the signal produced by a dipole of $-10^{-19}$ emu located at the center of the active area and oriented with its moment perpendicular to the plane of the active area.

Rotation of the net array magnetization at low temperatures (20 K) has been observed to occur by both reversible and irreversible modes, the latter revealed by Barkhausen jumps. Spatially-resolved measurements at room temperature show the particles to be single domain at this length scale with remanence and coercivity indicating they are not superparamagnetic. Individual particles are observed to switch irreversibly over a small field range (< 10 Oe) between preferred magnetic directions parallel to the growth direction of the particles. Insight about the magnetization reversal process can be gained by a measurement of the switching field, the field at which the rotation of the moments becomes irreversible. Magnetization reversal here tends to occur by Barkhausen jumps rather than by continuous rotation of moments. Repeated sweeps show more variation in the irreversible part of the loop than in the reversible part. The irreproducibility of the discrete jumps could either be due to domain wall motion over a variable energy landscape or random switching of single domain particles. Magnetic force imaging described below supports the latter interpretation.

While a Hall magnetometer measures the average properties of the arrays, a MFM resolves the magnetic state of individual particles and simultaneously provides topographic information. The iron particles show remanent magnetization at room temperature, demonstrating that they are not superparamagnetic at these temperatures. Furthermore, the magnetic moments of the particles appear to have a preferred orientation either parallel or antiparallel to the growth direction. Unlike the hysteresis loops obtained with the Hall magnetometer, however, the magnetic force images do not show a reversible rotation of the moments. The higher temperature of the magnetic force measurements as compared to the Hall bar measurement (300 K vs. 20 K) provides more thermal energy for the moments to overcome anisotropy energy barriers and thus may not allow for a stable arrangement of the moments at an angle from the easy axis. Within the spatial resolution of the MFM, it is not possible to discern whether the magnetization reversal is occurring by a coherent (uniform reversal of the magnetic moments) or an incoherent mode (e.g., nucleation and rapid motion of a domain wall). While the particles appear structurally similar in the topographic images, the
magnetic force image shows that the coercive fields of the particles are not the same. The true shapes of the particles may be more variable than revealed through the carbon coating and/or shape may not be the only source of magnetic anisotropy. Statistics on the magnetization reversal process are also acquired by imaging in constant magnetic fields of increasing strength following a large saturating field in the opposite direction. Although the same average number of particles are observed to switch at each field on different field sweeps, it is not the same particles that switch at each field.

Scaling of the arrays offers the possibility of room temperature magnetic storage at this demonstrated level of 45 Gbit/in.², nearly fifty times greater than current technology. The practical realization of a storage technology at this level would obviously require higher rates of deposition for scaling the arrays over larger areas and higher data rates for reading than are currently available with scanning probes. Parallel arrays of scanning probes may overcome current scanning probe bandwidth limits. Improvement is also necessary in the control of the magnitude of the anisotropy of particles. Although the STM forms particles that are ostensibly similar in structure with a preferred anisotropy direction, there is found to be a distribution of local coercive fields. Magnetic properties may thus serve as a more sensitive characterization of nanometer-scale particles. The Hall magnetometer provides a measure of the average magnetic properties of nanometer-scale particles, but requires care in interpretation since particles are not weighted equally. Imaging of individual particles with an MFM shows the effect of the scanning tip on small particles can be significant. Another outstanding challenge is the development of local magnetic probes which are less invasive.

b) *Submicron Room-temperature Ferromagnets in Semiconductor Heterostructures*

Submicron room-temperature ferromagnets have been successfully formed in GaAs semiconductors through a simple process of Mn⁺ ion implantation and subsequent heat treatment. A combination of transmission electron, atomic force, and magnetic force microscopies have been used in conjunction with SQUID magnetization measurements to directly examine the structural and magnetic properties of this new system. After Mn⁺-implantation at various doses, rapid thermal annealing crystallizes in situ submicron GaMn ferromagnetic particles (~200 nm) at the GaAs surface. These GaMn particles are crystalline, some with quasicyristalline-like order. Bulk magnetization measurements show that the GaMn particles are room temperature ferromagnets with a Curie temperature far exceeding room temperature. High resolution magnetic force microscopy (MFM) images on single GaMn ferromagnets reveal that unmagnetized samples contain both magnetic single- and multi-domain particles, but after initial magnetization, the single-domain state predominates, with magnetic moments aligned preferentially along the [001] directions of the GaAs substrate. In particular, magnetic force imaging has been performed in a changing magnetic field (up to 8 kOe) to directly image magnetization reversal of individual single-domain particles. MFM images and magnetic anisotropy of GaMn particles are studied in parallel to dipolar field simulations. Furthermore, submicron GaMn ferromagnets have recently been assembled and self-organized in lithographically patterned GaAs structures (~5 nm) for magnetoelectronic studies. In order to understand the images of mesoscopic magnetic structures taken under external fields, we have characterized the MFM probes by imaging microfabricated current-carrying strips in applied magnetic fields. Patterned micrometer scale lines containing submicron magnetic structures on GaAs are fabricated using lithography in conjunction with broad beam ion implantation.
Micromachined cantilevers for single particle magnetometry: mechanical detection of magnetization

The fabrication of microscopic mechanical cantilevers combined with sensitive displacement detection schemes has resulted in the development of several powerful experimental techniques for micromagnetometry, including a new class of torque magnetometers. We have made a significant advance in this instrumentation by exploiting the fact that the force sensitivity of these techniques can be improved by lowering the spring constant \( k \) of the cantilever (thereby increasing the displacement per unit force) and increasing the resonant frequency \( f \) (decreasing the necessary averaging time). Since most semiconductors and metals have mass densities and elastic moduli within an order of magnitude of each other, the design parameters that afford the greatest opportunities for improvements are the physical dimensions of the cantilever. Specifically, for a rectangular cantilever, one can achieve small \( k \) and large \( f \) by simultaneously decreasing all the dimensions to the submicron scale. We have successfully fabricated cantilevers from III-V based GaAs materials, thereby offering the advantages of integration with optical devices, magnetic systems, and strain sensing elements that utilize the piezoelectric properties of the GaAs to detect the cantilever displacement. We have developed a new process for making sub-micron thickness micro-mechanical cantilevers out of single GaAs epilayers grown by molecular beam epitaxy to operate from room temperature to 30 mK in fields up to 19 T. We have fabricated cantilevers <100 nm thick, \~1 micron wide, and ranging from 1 to 500 microns in length (2000:1 aspect ratio). These dimensions are comparable to those of the most sensitive cantilevers made from silicon (which is a much more mature process), and give a spring constant as small as \( 10^{10} \) N/m. The process we have developed could easily be adapted to work with any of the III-V or even II-VI semiconductor families, allowing the possibility of integrating a cantilever with a magnetic quantum well, for instance. The process has a number of advantages, including a very high yield rate and a design which allows easy access to both sides of the cantilever. In addition, we have built a fiber optic interferometer with a displacement sensitivity of \( 6 \times 10^{10} \) Angstroms per root Hertz, allowing very sensitive detection of the cantilever motion. Initial results using 100-nm thick structures have yielded magnetic sensitivities of \( < 10^{-1} \) emu, and have produced magnetization measurements and magnetic noise spectroscopy. Moreover, such measurements have taken place over a wide variety of temperatures (1K - 300K) and applied magnetic fields (0-8T), as well as in ambient conditions (atmospheric conditions and room temperature). While this process was originally designed for the fabrication of torque magnetometers, fabricating cantilevers out of the magnetic III-V materials or Mn-implanted GaAs could also be advantageous for making extremely high resolution Magnetic Force Microscopy probes.

New biological magnets: bacterial magnetic strings (bionites) & proteins

We are studying the magnetic properties of biologically grown samples using a SQUID magnetometer, in particular looking at thread-like structures called bionites grown by Professor S. Mann at the University of Bath, Great Britain. Bionites are long assemblies of single celled bacteria that fail to completely separate when reproducing and so naturally form long chains of cells. These chains get tangled up and when drawn out of solution form a thread which looks under magnification like a rope made up of many strands of cells. The bionites by themselves are virtually non-magnetic, but can be made magnetic by adding microscopic (10 - 20 nm diameter) \( \text{Fe}_3\text{O}_4 \) particles to the solution from which a bionite is drawn. The magnetic particles become trapped between the strands of cells. The resulting magnetic bionite is a superparamagnet at room temperature. The individual magnetic particles are ferromagnetic with the spins of all the constituent Fe atoms aligned along some axis. However the magnetic moment of each particle is free to move giving paramagnetic properties. (a magnetic moment proportional to and in the same direction as an applied field) for the whole sample. The motion of a magnetic moment is thermally activated and at lower temperatures (below 175 K) the motion becomes blocked and the sample displays hysteresis in the plot of moment versus applied field. There is a clear difference between the magnetic properties measured with an applied field either along the length or across a bionite. There are two simple explanations: Either the \( \text{Fe}_3\text{O}_4 \) particles have some shape, giving a preferred direction of the magnetic moment, and the action of drawing the bionite
thread causes the particles to align with the thread. Or, more likely, the \( \text{Fe}_3\text{O}_4 \) particles are isotropic but the action of drawing the bionite thread causes there to be strain which would set up a preferred magnetization direction.

In an effort to form self-assembled biological magnets, macroscopic magnetic bacterial threads have been formed in which small (10 - 20 nm) \( \text{Fe}_3\text{O}_4 \) particles are intercalated between cell walls. These structures employ a mutant strain of bacteria whose cells are unable to separate from each other on dividing. Using a drawing technique from a suitable surface culture, these “bionites” form a solid thread typically 100 microns in diameter and several centimeters long, resulting in a thread which has a greater tensile strength than steel. Cross-sectional AFM and SEM images of the bionites reveal a single thread containing ~ 10^4 strands of cells close packed and aligned along the length of the thread. The bionites are magnetically doped by incorporating a ferrofluid containing \( \text{Fe}_3\text{O}_4 \) particles to the culture solution, and appear capable of holding a wide variety of magnetic particle sizes. SEM images reveal the presence of these embedded magnetic particles between the cell walls and provide an estimate of the volume fraction of magnetic material. Temperature-dependent SQUID magnetometry measurements show the bionite to be superparamagnetic with a blocking temperature \( T_B \approx 175 \) K. Below the blocking temperature, the bionite magnetization displays a field-dependent hysteresis indicating anisotropic behavior. Angle-dependent magnetization studies in fixed applied fields provides a quantitative measure of the anisotropy and the energy barrier.

In a parallel continuing effort, using multiple modes of the Atomic Force Microscope, synthetic ferritin (a biomaterialized magnetic protein) has now been manipulated into small patterns, for example a grid of micron by micron magnetic boxes, limited in size only by the accuracy of available mechanical translation stages. Synthetic protein lines are made by literally dragging a contact tip, in contact mode of the AFM, across ferritin on a mica or semiconductor substrate thereby forcing the ferritin out of the line of the tip. Patterns are made by adjusting the sample in accordance with the pattern desired after each line is made. The fundamental limits of the AFM should allow for any submicron pattern fabricated by a series of specifically placed rectangles. Upon project completion the small patterns can potentially be used as optical grids and as small magnets on top of semiconductor devices for spin-dependent patterning of electrons.

- **Spatiotemporal Near-field Spin Spectroscopy in Digital Magnetic Heterostructures and Quantum Dots**

During this grant period we have successfully developed a variety of novel II-VI magnetic semiconductor quantum structures (magnetic quantum dots and doped magnetic semiconductor heterostructures) which have been explored using femtosecond-resolved magneto-optical techniques, including Faraday rotation and near-field spectroscopy. These studies uncovered unexpected new coherent electron spin dynamics in the solid state which persist to room temperature, and offer the promise of new ultrafast magnetoelectronic devices. Spatially-resolved optical microscopy has revealed the existence of “spin quantum dots” with one electron/structure. Parallel studies including the development of low temperature near-field magneto-optical spectroscopy and micromagnetometry have produced new high resolution measurement methods for nanometer-scale magnetic structures, demonstrating spatial resolution of <100 nm and magnetic sensitivities <10^{-6} emu.

A femtosecond-resolved low-temperature near-field scanning optical microscope (NSOM) is used to monitor the spatiotemporal evolution of excitonic spins in digital magnetic semiconductor quantum structures which are laterally patterned with a focused beam of Ga⁺ ions. Polarization-resolved photoluminescence (PL) images at T=5K reveal a spin-dependent energy landscape due to locally depressed Zeeman splittings in the implanted regions. Marked differences between carrier and spin behavior are observed by sharp contrasts in intensity and polarization profiles, showing that excitonic diffusion has a minimal effect on the local magnetic interactions which contribute to Zeeman-split states. Time-resolved measurements suggest that exciton diffusion is driven by a spatially varying energy profile, and acquires a
spin-dependent component in the presence of a magnetic field. The data also demonstrate fundamental limitations on the measurement of polarized PL from semiconductors in the near-field regime.

The heterostructures consist of single 120Å ZnSe/ZnCdSe MBE-grown semiconductor quantum wells (QW) containing a systematic planar distribution of magnetic ions (Mn²⁺). In magnetic fields, traditional magneto-optical data show narrow PL linewidths and large Zeeman splittings, making them ideal systems in which to study local spin-dependent interactions. A low-dosage (10³ μm²) 140 keV 100 nm-diameter focused beam of Ga⁺ ions is used to implant specific patterns in the etched structures. Field-dependent PL experiments are performed with one of two NSOMs at T=4K-300K, where carriers are optically excited and polarization-resolved PL from the n+1 heavy-hole excitonic peak collected in the near field.

NSOM images of the PL intensity and polarization are obtained by scanning an aperture over the sample surface and collecting luminescence at fixed detection energies. The intensity is suppressed in the implanted regions, and recovers slowly in the intrinsic areas. In contrast, the measured polarization is roughly constant in the intrinsic areas, and drops abruptly to zero as one moves into the implanted regions. The slow modulation of the intensity is attributed to exciton diffusion from intrinsic into nearby implanted regions. Spectrally-resolved spatial scans provide a quantitative measure of the energy landscape for the two excitonic spin states.

Time-resolved measurements are performed using a frequency-doubled mode-locked Ti:Sapphire laser producing 130fs pulses. Both luminescence intensity autocorrelation (LIA) and pump-probe absorption techniques are exploited to obtain time- and spatially-resolved information about the lifetime of radiative excitonic states. The lifetime is reduced for both spin states in implanted regions, but the changes are much more pronounced for the spin-down state. These differences are attributed to an enhanced diffusion of spin-down excitons from the implanted to the nearby intrinsic regions, driven by the spin-dependent potentials.

We have recently succeeded in the fabrication and direct observation of 0D exciton confinement in wide-gap II-VI quantum dots. These nanostructures are formed during the strained layer epitaxy of (cubic) CdSe (E_g = 1.75 eV) on ZnSe (E_g = 2.8 eV) with a lattice mismatch ~ 7%. The 0D nature of the confined electronic states in these quantum dots is directly revealed through PL spectroscopy with high spatial resolution (~100 nm) carried out using a low-temperature near-field scanning optical microscope. The smooth, inhomogeneously broadened lineshape of typical far-field PL spectra is seen to evolve into a spectrum characterized by sharp, resolution-limited (0.8 meV) reproducible spectral features in the near-field, arising from a convolution of the delta-function density-of-states of each individual quantum dot region and an envelope determined by the statistical distribution of quantum dots. Measurements in II-VI CdSe quantum dots electronically coupled to adjacent MnSe magnetic layers have revealed the existence of magnetic quantum dots, exhibiting field-tunable Zeeman splittings in individual dots. These near-field optical spectra at low temperatures demonstrate the ability to fabricate and measure genuine “spin quantum dots” in applied magnetic fields, and explore spin-dependent phenomena in zero-dimensional structures. The observation of 0D states in II-VI nanostructures opens up exciting possibilities for studying static and dynamic spin dependent phenomena in “quantum spin dots” by the incorporation of magnetic ions into II-VI nanostructures.

- **Room-Temperature Spin Memory in Two-Dimensional Electron Gases**

Recent discoveries in gases that spin ensembles may be used collectively as single quantum elements have renewed optimism that coherent electronics may eventually be realized as a basis for computation. The advantages for computation are unprecedented speed (exponentially faster than present technology for many calculations) and potentially high levels of integration. Semiconductors offer the advantage that spin orientation of carriers (electrons and holes) induces strong optical non-linearities that may be used to establish and to probe electronic coherences. This discovery demonstrates, for the first time, an optically active solid state system in which electron spin coherence persists for nanoseconds at room temperature.
Time-resolved Kerr reflectivity of two-dimensional electron gases in II-VI semiconductor heterostructures provides a direct measure of electron spin precession and relaxation over a temperature range from 4 to 300 kelvin. The introduction of n-type dopants into these systems increases the electronic spin lifetimes over three orders of magnitude relative to insulating counterparts, a trend that is also observed in doped bulk semiconductors. As the electronic spin polarization in these systems survives for nanoseconds - far longer than the electron-hole recombination lifetime - this technique reveals thousands of 15 gigahertz per tesla spin precession cycles within an electron gas. Remarkably, in contrast to theoretical expectations, these spin beats are only weakly temperature dependent and persist to room temperature. The discovery suggests an opportunity for practical room-temperature ultrafast coherent magneto-electronics in semiconductors, and the first demonstration of the potential for constructing quantum computing devices using semiconductor technology. In addition, the measurement technique has been shown to enable real-time spin resonance measurements of microscopic conductors and insulators.

2. Students Funded Under this Award and Present Placement

Postdoctoral Student: Dr. Paul Crowell
(performing research under award)
Assistant Professor, Department of Physics
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Dr. Jing Shi
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Phoenix, AZ

Funded Graduate Students:
Vladimir Nikitin
IBM Storage Systems Division
San Jose, CA

Sivas Gider
IBM Storage Systems Division
San Jose, CA

3. Publications from Previous Grant Support

Peer-reviewed journal articles


**Conference Papers**


4. **Invited Talks During Past Grant Period**


5. Contributed Talks During Past Grant Period


