Collective and quasiparticle properties of nanocrystals and their arrays

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The fundamental competition between order and disorder lies at the heart of materials science and technology. Interactions between atoms or electrons with sub nanometer spacings can lead to collective organization into states with long-range order. Order in electronic states gives rise to physical consequences such as magnetism, ferroelectricity and superconductivity. In bulk materials, the collective electronic properties associated with ferromagnetic and superconducting order yield effects that are exceptionally robust and useful. Collective organization of quasiparticles gives rise to a number of characteristic length scales. Examples include the coherence length and the minimum grain size in superconductors, and in ferromagnets the domain wall width, the superparamagnetic limit, and the maximum single-domain radius. These length scales generally exceed or are comparable to geometrical parameters of nanostructured materials and are, on the other hand, not far removed from the microscopic length scales such as inter-electron spacing. For these reasons nanostructured materials represent an important new frontier for the study of collective electronic behavior. This interdisciplinary research team will study the physical consequences of nanometer-size dimensions on collective and independent electron properties in individual nanocrystals and in controlled nanocrystal arrays.

This program brings together expertise in the epitaxial growth of self-assembled semiconductor quantum dots, expertise in colloidal synthesis of both metal and semiconductor nanoparticles, expertise in nanoscale optical spectroscopy, expertise in scanning tunneling and magnetic force microscopy, SQUID magnetometry and cantilever micro-magnetometry, and expertise in mesoscopic and many-body condensed matter theory. We intend to work closely together, with materials synthesis efforts informed by theoretical ideas on interesting effects that might be realized in novel nanoparticle systems, and the modeling and theory effort informed by materials ideas on nanoparticle synthesis. Both efforts will ultimately be guided by optical, magnetic, tunneling, and transport measurements of nanoparticle physical properties.

The materials systems we will focus on will be the controlled two- and three-dimensional arrays of metal and semiconductor nanocrystals realized by colloidal synthesis, and molecular beam epitaxy. We intend to study ferromagnetic, superconducting, and normal metal nanocrystals, and ferromagnetic, semimagnetic, and normal semiconductor quantum dots. Coulomb blockade and independent electron properties of both metallic and semiconductor nanoparticles have already been extensively studied. In ferromagnetic and superconducting systems the emphasis to date has been on the collective properties that underlie, for example, the use of ferromagnets for information storage and the potential use of small superconductors as quantum-bits. As these particles become smaller, the physics of the magnetic anisotropy barriers essential for information storage will be altered, and superconductivity will be destroyed, respectively. This regime is a frontier for fundamental physics and for materials physics and chemistry.

Below we show two examples that highlight some of our accomplishments: (a) Novel growth and magnetic properties of Co quantum platelets on Si(111) surface; and (b) Coherent optical manipulations of excitonic quantum bits in semiconductor quantum dots.

(a) Novel growth and magnetic properties of Co quantum platelets on Si(111) surface

Nanometer-sized magnetic particles exhibit a wide range of fascinating magnetic phenomena, and have attracted continuous attention both scientifically and technologically. In order to incorporate magnetic innovations into existing Si-based semiconductor technology, many studies have attempted to grow magnetic nanostructures on semiconductor surfaces to utilize charge carrier's spin in conventional electronic devices [1,2]. However, achieving adequate particle size control and silicidation suppression are still very challenging and a subject of extensive investigation. Here we show a novel growth process which leads to the formation of magnetic nanostuctures with well-controlled size and shape on Si without observable silicide formation.

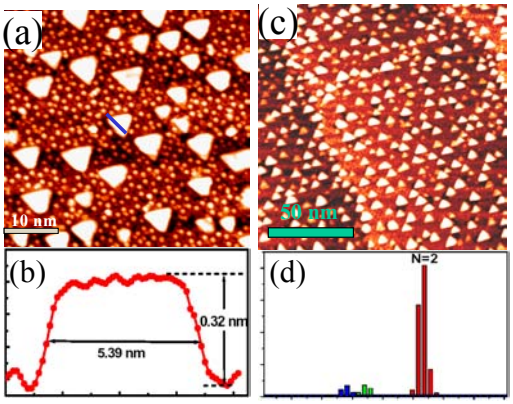

Fig. 1 (a) A STM image of Co platelets grown on Si(111) 7x7 decorated with order array of Al clusters. (b) Line profile across one of the platelets. (c) Larger scale STM image. (d) histogram of size distribution of the Co platelets

Under proper growth condition, it was discovered that Al atoms can form ordered cluster array on Si(111) 7x7 surface where each half-unit cell (HUC) contains exactly 6 Al atoms [3,4]. Using such a surface as a template, we succeeded in growing Co nano-platelets with well-controlled size and shape, as illustrated in Fig.1. These nano-platelets are triangular and are exactly two monolayer thick. Furthermore, the lateral length of the platelets are quantized in unit of the HUC for Si(111) 7x7. With fine tuning of the growth parameters, we can also achieve a uniform size distribution of the nanoplatelets whose lateral size is mostly 2 units of HUC (N=2) (see Fig. 1 (c) and (d))

The magnetic properties of such Co quantum platelets array were investigated using SQUID magnetometer after capping the surface with 20 nm of Au. Shown in Fig. 2(a) is the magnetization-field (M-H) hysteresis loop measured at 4.2K when the magnetic field is applied perpendicular to the surface. The in-plane loop (not shown) was also measured and exhibited no hysterisis, implying that the easy axis perpendicular to the sample surface. Most interestingly, there are distinct step structures observed in the hysteresis loop. Theoretical investigations attributed such structures to a novel pinning mechanism of "spin" pairs (each platelets is treated like a giant spin either pointing up or down). Monte Carlo simulations based on an Ising-like Hamiltonian reproduced the step structure very well (shown in Fig. 2(b)).

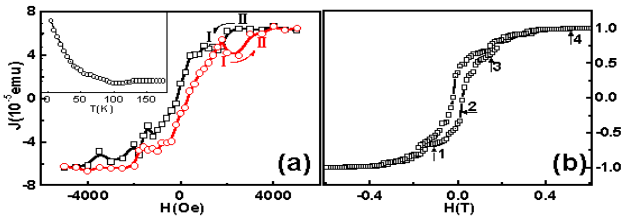

Fig. 2 (a) The M-H hysteresis loop measured at 4.2K when the magnetic field is applied perpendicular to the surface. The inset shows that the Blocking temperature of the Co quantum platelets is roughtly 50 K. (b) Theoretical calculation of the M-H hysteresis loop using Monte Carlo simulation based on an Ising-like Hamiltonian

This example demonstrates how materials synthesis, characterization, and theory work hands-in-hands together to control novel magnetic properties through novel nanostructure engineering [5].

(b) Coherent manipulations of excitonic quantum bits in semiconductor quantum dots

Quantum computing relies on the capability of setting the state of a two-level system to an arbitrary linear combination of its two eigenstates. The system, referred to as a quantum bit or qubit, can be "rotated" entirely from the lower state to the upper state and vice-versa. Due to their atomic-like energy structure, semiconductor quantum dots (QDs) are ideal candidates for qubits and have been envisioned as the fundamental building blocks of quantum information processing. By using the discrete excitonic states in QDs as qubits, we demonstrated high quality factor qubit rotations in InGaAs quantum dots.

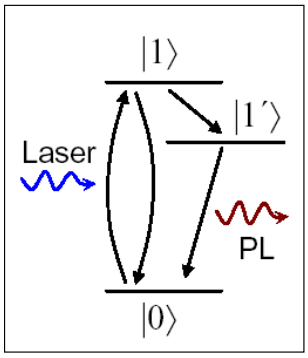

Fig. 3 QD energy structure. The QD is resonantly excited to the first excited excitonic state |1⟩. It then relaxes non-radiatively to the excitonic ground state |1′⟩ and finally radiatively to |0⟩ (no exciton).

Figure 3 shows how the qubit, formed by states |0⟩ and |1⟩ can be addressed resonantly with a picosecond laser pulse. Here |0> represents exciton vacuum (no electron hole pair in the QD). State |1> is the exciton excited state. State |1′⟩ is the exciton ground state which is also a spectator state used to monitor the population of state |1⟩. Under resonant excitation with a strong electromagnetic field, the population of |1⟩ undergoes periodic oscillations as a function of the input pulse area θ, which is proportional to the square root of the average laser intensity. These qubit rotations, also called Rabi oscillations, are shown in figure 4 where the ground state PL, which is proportional to the population of state |1⟩, was recorded as the intensity was varied at fixed pulse width.

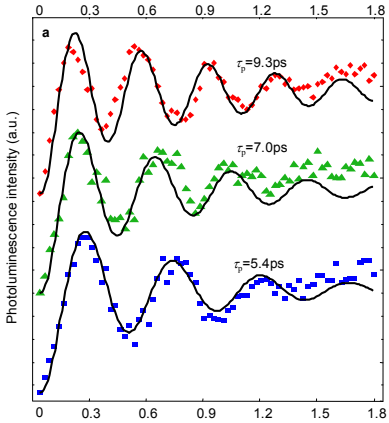

Fig. 4 Behavior of Rabi oscillations (RO) for a particular excitonic qubit driven by an external laser at different laser pulse width τ_p. The PL from the |1′⟩ to |0⟩ transition was recorded while the average intensity was varied. Each oscillation corresponds to a complete rotation of qubit driven by the external laser pulse

These are the longest oscillations reported so far in quantum dots and make it possible to carefully study damping characteristics. As shown in Fig. 4 oscillation amplitudes are damped exponentially, and the damping rate increases sub-linearly with the intensity. This leads to a quality factor $Q=\Omega/\Gamma=15$, where Ω is the Rabi frequency (the frequency at which the qubit is driven between |0⟩ and |1⟩) and Γ the damping rate. Understanding the source of the damping, plays the most critical role toward practical implementation of quantum computation [6]

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