Why study mesoscopic physics?

There are a lot of ways in which nan-μm samples differ from both atomic systems and larger devices. I won’t try to lay out a systematic list — will just give examples:

- **Single-electron effects** — Coulomb blockade can control electrons one by one

- **Near crossovers between classical and quantum physics**
  - 1 μm scale: phase coherence, weak localization, Aharonov-Bohm
  - 100 nm individual quantum states in semiconductors at low T
  - 10 nm individual quantum states in metals

- Can now manipulate single atoms and molecules and control their coupling to the outside world.

Lots of important scales in the 1 nm - 1 μm range

- elastic and inelastic mean free paths of electrons
- spin-flip diffusion length (important in spin-polarized transport)
- domain-wall widths in magnets
- superparamagnetic crossover in magnets
- in superconductors, superconducting coherence and magnetic penetration lengths
- many others...

New discoveries are being made, both experimentally and theoretically. Knowledge being developed is leading to some practical applications. A time of productive ferment, if not always the most organized field.
Plan for Dan Ralph’s Lectures (Tentative)

1. Basic introduction to Coulomb blockade in quantum dots
   Simple spin physics, Zeeman spin splitting
   Spin-orbit effects on Zeeman splitting, including random-matrix-theory ideas

2. Interacting Electrons within Quantum Dots -- the Universal Hamiltonian
   (a) Weak exchange interactions -- non-zero spin states prior to the Stoner instability
   (b) Superconducting pairing
   (c) Ferromagnetic quantum dots

3. Review of Kondo Experiments
   Single-Molecule Devices

4. Nano-magnetics (in metals)
   (a) basics of “giant magnetoresistance” (GMR)
   (b) spin-transfer torques and magnetic dynamics
Coulomb Blockade and Tunneling Effects in Nanostructures

Imagine the circuit contains an object that provides a pool for electrons to enter - e.g., metal grain, semiconductor dot, redox state in a molecule, localized charge state.

If the grain is small, it has a small capacitance $C$ to outside world.

$\Rightarrow$ Can't ignore the classical charging energy $E_c = \frac{e^2}{2C}$ even for one electron.

For aluminum oxide junctions, (100x100)nm$^2$ $C \approx 0.5$ F

$\Rightarrow E_c \approx 0.2$ meV or (2.6) k$\Omega$

For (5x5)nm$^2$ junctions, $C \approx 1$ aF $E_c \approx 80$ meV or (0.08) k$\Omega$

**Conditions for Coulomb blockade**

1. $kT, eV < E_c$ - electrons in electrodes can have insufficient energy to hop onto the island and off the other side.
2. To avoid quantum fluctuations, need "energy uncertainty" $\ll \frac{e^2}{2C}$

$\Rightarrow \frac{kT}{e} \ll \frac{e^2}{2C}$

$\Rightarrow \frac{kT}{eC} \ll \frac{e^2}{2C}$

$\Rightarrow R \gg \frac{2kT}{eC}$

$\Rightarrow \frac{1}{R} = 25.8 \text{ "resistance quantum"}$

When $R \gg \frac{1}{C}$ and $eV$ is comparable to $E_c$, at most one excess electron at a time can occupy the island $\Rightarrow$ simple electron tunneling.
With a tunable gate voltage $V_g$, one can make a single-electron transistor. $V_g$ tunes the energy cost for adding an electron.

$$E(n) = E(n_e) = \frac{Q_0 e^2}{2C} - neV_g \frac{Q_0}{2C}$$

for $V=0$

$$= \frac{1}{2C}[Q_0 - V_g Q_0]^2 - \frac{V_g Q_0}{2C}$$

Now what matters is $E(n+1) - E(n) = \text{energy required to change the number of electrons by one}$. This means we can ignore the last term because it is independent of $n$.

$$E(n) = \frac{1}{2C}[V_g Q_0 - ne]^2 \equiv \frac{1}{2C}[Q_0 - ne]^2$$

where $Q_0 = V_g Q_0$ is called the "offset charge", but really it is due to the effects of an external potential.

Graphically:

The different charge states have different energies. With a gate voltage, you can change how many electrons are in the ground state of the island.

For a fixed value of $Q_0$, there is a minimum energy cost to change charge number (vertical arrows).

At $Q_0 = \pm \text{integer values}$, there are degenerate ground states. At these values of gate voltage, charges can flow with zero Coulomb energy cost. The gate voltage has tuned away the Coulomb blockade.

Conductance $\frac{dI}{dV}$

$\frac{1}{2C} \frac{1}{eC} \frac{1}{2eC} \frac{Q_0 = V_g Q_0}$
Another picture for the same physics:

(assume that the Coulomb blockade is overcome by adding an electron)

The threshold for tunneling for this sign of voltage is when
\[ eV \frac{C_l}{C_l + C_r} = qE(V_g) \]

For the opposite sign of bias
\[ eV \frac{C_l}{C_l + C_r} = \Delta E(V_g) \]

\( V_g \) shifts the energy levels on the island up or down relative to the Fermi level in the electrodes. When the levels are shifted down to \( E_F \), there is no charging energy.

One can draw a similar diagram for the case when the Coulomb blockade is overcome by subtracting an electron.
Micron-scale Aluminum SET (continuous DOS) 
Hergenrother et al.

GaAs SET (discrete DOS) 
Patel et al.
Coulomb-Staircase Curves

![Coulomb-Staircase Curves Graph](image-url)
Effects of Gate Voltage on Coulomb Blockade

color scale: $\frac{dI}{dV}$
Coulomb-Blockade Effects in One Molecule

- High resistance (> megaOhms) - single electron charging.
- Coulomb blockade > 150 meV (unstable beyond this).
Quantum dots - Discrete Electronic States instead of Continuum

What changes? Can use the same type of picture:

(assume lifetime broadening of states is $< kT$)

Coulomb blockade physics is unchanged. But now the turn-on of current is changed. Instead of a continuous turn-on, the current will begin with a series of small steps, as the discrete states provide alternative channels for tunneling.

Other differences from the case of continuous states:
- Spacings between Coulomb blockade peaks will not all be equal, because differences in level spacing can occur.
- The current associated with tunneling through different discrete states can vary, due to differences in individual tunneling matrix elements.

Note: In InAs quantum dots, the tunnel barrier charge with $V$. Allows it hard to resolve excited states in $I-V$ curves.
(a) Coulomb blockade ($\sim 15$ mV)
(b) Level spacing ($\sim 0.5$ mV)
$T = 320 \text{ mK}$

$H = 0.1 \text{ Tesla}$
Tunneling through Individual Quantum States

$\theta << kT$

(a) Current (pA) vs. Bias Voltage (mV)

(b) $dI/dV(\mu S)$ vs. Voltage (mV)

Cobalt lead
1 Tesla
Part of Coulomb Diamond for Aluminum Particle

Color scale denotes $dl/dV$.

Red lines - excited states of $n+1$ electrons
Blue lines - excited states of $n$ electrons
Black lines - ground states
Why study these spectra of discrete states?

- Experiments show that the spectra are not well described by simple models of non-interacting electrons. Instead, they reflect all the different forces and interactions acting on the electrons, with different types of interactions having distinguishable effects.

These spectra are a good way to learn about the consequences of electron interactions, at a very fundamental level.

(Many analogies to atomic and nuclear physics.)

There are different types of forces and interactions acting on electrons in semiconductors, normal metals, magnets, superconductors, carbon nanotubes, other molecules, etc.

In this lecture and the next, I will discuss the consequences of different kinds of forces.

1) Spin-Orbit interactions (for Al, Cu, Ag, Au)

2) Weak exchange interactions and non-zero spin states (compare GaAs and Au)

3) Superconductor pairing (Al)

4) Strong exchange and magnetic anisotropy forces (ferromagnetic Co)
Basic of Spin in Quantum Dots (non-magnetic)

It is possible to tell if a nonmagnetic dot has an even or odd number of electrons, even if the number of electrons is in the 1000's.

One procedure: Look at the lowest-energy tunneling resonance. 
If it undergoes Zeeman spin splitting, then the transition corresponds to even→odd. 
If only one transition appears, moving to higher energy as a function of B, you have odd→even

Single-electron picture:

for electron addition:

\[ \begin{align*} 
B & \uparrow \downarrow \quad \text{even→odd} \\
E & \quad \text{both states empty and available for tunneling} 
\end{align*} \]

\[ \begin{align*} 
B & \uparrow \downarrow \quad \text{odd→even, lower E} \\
E & \quad \text{state is blocked by the odd electron, only one state available for tunneling} 
\end{align*} \]

for electron subtraction:

\[ \begin{align*} 
B & \uparrow \downarrow \quad \text{even→odd} \\
E & \quad \text{both states available} 
\end{align*} \]

\[ \begin{align*} 
B & \uparrow \downarrow \quad \text{odd→even, only filled state is available} \\
E & \quad \text{Kramers Doubt} \text{ only lower state occupied, only one transition visible} 
\end{align*} \]

Alternative Many-body picture: (holds as long as the ground state is spin 0 for even electrons, spin 1 for odd)

\[ \begin{align*} 
\text{even→odd} & \quad s = \frac{1}{2} \text{ in B field} \\
\text{odd→even} & \quad s = 0 
\end{align*} \]
Magnetic-Field Dependence of Aluminum Levels

\[ g = 2.0 \pm 0.1 \] for Al.
\[ \frac{dI}{dV} (\text{M}\Omega)^{-1} \]

For various values of \( V \) (mV):

- 4.0 to 4.4
- 3.8 to 4.0
- 3.6 to 3.8

The graph shows peaks at different voltages, indicating the behavior of the system under different magnetic field strengths. The label \( N \text{ is even} \) suggests that the number of channels or states is even, which is a characteristic of certain quantum systems.

Additionally, there are graphs illustrating the current \( I \) (pA) vs. voltage \( V \) (mV) for different magnetic field strengths:

- 0.03 Tesla
- 3 Tesla

These graphs display step-like increases in current with respect to voltage, typical of systems experiencing quantum tunneling effects or other quantum phenomena under magnetic influence.
odd number of electrons
occupied  unoccupied

even number of electrons
occupied  unoccupied

$g = 1.30$ to $1.82$
$\langle g \rangle = 1.58$
In general, copper is a little more complicated than aluminum.
Note for Al:

- $g = 2$ for all states to within measurement uncertainties
- Linear dependence of energy on $B$
- Simple crossings between up and down spin states

For Cu:

- $g < 2$ for all states
- $g$ differs from state to state
- Avoided crossings of up and down spin states

What causes the difference? The shifts in spin energy vary depending on how close in energy are other orbital states - we should suspect spin-orbit coupling.

Cu ($Z = 29$) is a heavier element than Al ($Z = 13$)

So scattering is stronger.

References for spin-orbit scattering in metals:

R.J. Elliot, Phys Rev. B6, 266 (1954)


Electrons scatter off of impurities, interfaces, grain boundaries, or phonons. This rotates the spin, mixes spin-up and spin-down components in the wave function.

But time-reversal symmetry still holds – Kramer’s degeneracy is still valid.
With spin-orbit interactions, the true energy eigenstates are not pure spin up or spin down (the total electron spin does not commute with $H$). Instead, they are linear superpositions of $\uparrow$ and $\downarrow$:

$$1n, \uparrow\rangle = \alpha_n \uparrow\rangle + \beta_n \downarrow\rangle \quad \text{(related by time-reversal)}$$

$$1n, \downarrow\rangle = -\beta^*_n \uparrow\rangle + \alpha^*_n \downarrow\rangle \quad \text{(symmetry)}$$

1. $g$-factor for spin Zeeman splitting:

$$g_n = 2 \left< n, \uparrow\vert \mathbf{g}_e \vert n, \uparrow\rangle = 2 \left( 1 - \beta_n^2 \right) = 2 \left( 1 - 2 \beta_n^2 \right) \leq 2.$$  

(For weak spin-orbit interactions, real particles have negligible orbital contribution to the $g$-factor, because orbital angular momentum is quenched, unlike atoms. The orbital contribution will be considered below in the regime of strong SO.)

Simple perturbation theory can explain the qualitative features I have described in the Cu dots. (J. Sono, J. Phys. Soc. Jpn. 42, 1457 (1977))

2. Variations in $g$-factors:

$$g_n = 2 \left[ 1 - 2 \sum_{m\neq n} \frac{\left< n, \uparrow\vert \mathbf{H}_{SO} \vert m, \uparrow\rangle \right>^2}{(E_n - E_m)^2} \right]$$

Matrix elements will vary between the unperturbed single-electron states $\Psi_n \equiv \Psi_n$. The energy denominators will also vary.

$\Rightarrow$ for different orbital states $n$, $g_n$ can be different.

3. Avoided crossings:

In crossing region, $2\times2$ Hamiltonian

$$\hat{H}_J = \begin{pmatrix} E_1 + \frac{1}{2}g_\mu_B B & H_{SO} \\ H_{SO}^\dagger & E_1 - \frac{1}{2}g_\mu_B B \end{pmatrix}$$
More sophisticated theory of Spin-Orbit consequences - Random Matrix Theory


\[ \mathcal{H}(\lambda) = S \otimes \mathbf{1} + \frac{i \lambda}{\sqrt{N}} \sum_j A_j \otimes \sigma_j \]

- \( S = N \times N \) symmetric random matrix
- \( A_j = N \times N \) antisymmetric random matrix
- \( \lambda \approx (\lambda_0 \lambda)^{-\frac{1}{2}} \), strength of spin-orbit interaction

Theory can predict:

1. Full probability distribution of q factors for a given value of \( \lambda \) - not just in small-\( \lambda \) perturbative regime

2. The q factor for each energy level should be a tensor, should vary depending on the direction of magnetic field

\[ \text{Zeeman splitting} = \frac{N \lambda^2}{4} \left( g_1 B_1^2 + g_2 B_2^2 + g_3 B_3^2 \right) \]

where \( B_1, B_2, \) and \( B_3 \) are along the principle axes

- Ellipsoid (most complicated form allowed by symmetry for \( S = \mathbf{1} \))

Principle-axis q-factors predicted to vary by factors of 4 or 5 in regime of moderate \( S = (\lambda - 1) \) - not a small effect!

Physical picture:

The orbital wavefunctions in a neutral particle are highly oscillatory, essentially random functions. When the orbits are coupled to spin, the spin properties also take on random properties. There will be anisotropic in space, so the q-factors will depend on the direction of the magnetic field

- The principle axes for different states in one particle should be random, not associated with shape of crystal axes.

\( g_1, g_2, g_3 \) emerge from a matrix diagonalization. The spread in their values is related to eigenvalue repulsion.
Random Matrix Theory Predictions: Distributions of the principal g-factors for different strengths of spin-orbit interaction.
• With one fitting parameter per sample, both the average g-factor and the standard deviation are described well.
• Significant differences in spin-orbit strength even for particles made of the same metal.
Anisotropy of g-factors
Variations from Quantum State to Quantum State

Red: Cu#1
Blue: Cu#2
Principal-axis directions are randomly oriented in space.
Check agreement between experiment and RMT predictions

**Single parameter fit:** \( \Delta \) (spin-orbit scattering strength) is determined by matching the experimental and theoretical values of \( \langle g_1^2 \rangle + \langle g_2^2 \rangle + \langle g_3^2 \rangle \).

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<th>Exp.</th>
<th>RMT</th>
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<tbody>
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<td>( g_1 )</td>
<td>1.3</td>
<td>1.25</td>
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<tr>
<td>( g_2 )</td>
<td>0.8</td>
<td>0.76</td>
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<tr>
<td>( g_3 )</td>
<td>0.4</td>
<td>0.52</td>
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<td>1.59</td>
</tr>
<tr>
<td>( g_2 )</td>
<td>1.2</td>
<td>1.12</td>
</tr>
<tr>
<td>( g_3 )</td>
<td>0.9</td>
<td>0.96</td>
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Theory and experiment are in excellent agreement.
One Mystery: The g factors in gold nanoparticles are smaller than expected

Strong spin-orbit scattering limit (Matveev et al.)

\[ \langle g^2 \rangle = \frac{3}{\hbar^2} \Delta_{so} + \frac{l}{L}, \text{ where } \Delta \sim 1 \]

For a ballistic nanoparticle, \( l \sim L \)
\[ 2 \geq \langle g \rangle \geq 1 \]

For a diffusive nanoparticle, \( l < L \)
\[ \langle g \rangle \leq 2 \]

We see \( \langle g^2 \rangle \) as small as 1/50.

If we assume the orbital part does not contribute, the spin contribution to the average g factor gives an estimate for \( \Delta_{so} \) consistent with weak localization measurements.

Are the nanoparticles much more disordered than we expect so that they can quench the orbital contribution, or is there some shortcoming in the theory?
Summary

- Coulomb blockade, single-electron tunneling are ubiquitous when thinking about the physics of small-scale objects weakly coupled to electrodes.

- We developed pictures to understand the I-V characteristics when: level width $\Delta < kT <$ level spacing $<$ charging energy. This corresponds to relatively high resistance tunnel junctions with $R > \hbar/e^2$ (for lower-resistance junctions, cotunneling and Kondo physics come into play.)

- The study of spin Zeeman splitting is a good test to learn about spin-orbit coupling.

- Mesoscopic effects bring some surprises — e.g. very large fluctuations in g-factors between different orbital levels in the same metal quantum dot. Random matrix theory can be applied in explaining many results, but some mysteries remain.

Next time: Using measurements of electron-in-a-box energy levels to explore the consequences of different types of electron-electron interactions, and applications of a "universal Hamiltonian".
References for Ralph Lecture 1

General discussions of Coulomb blockade:
C. W. J. Beenakker, PRB 44, 1646 (1991)

Review of energy-level spectroscopy in metal nanoparticles

Spin-Orbit in nanoparticles
Matveev et al., PRL 85, 2789 (2000)
Gorokhov and Brower, PRB 49, 155417 (2004)
experiment: J. R. Petta and D. C. Ralph, PRL 87, 226801 (2001)
Petta and Ralph, PRL 89, 156802 (2002)