Why study mesoscopic physics?

There are a lot of ways in which nm–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 crossover between classical and quantum physics
  - ~ 1 μm scale – phase coherence, weak localization, Aharanov-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 for 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)

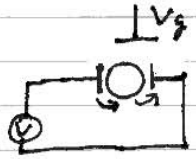
1. Basic introduction to Coulomb blockade in quantum dots
   Simple spin physics, Zeeman spin splitting
   Spin-orbit effects on Zeeman splitting, including random-matrix-theory ideas

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

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

for aluminum oxide junctions $(100\times100)\,nm^2$ $C \sim 0.5\,fF$
$\Rightarrow E_c \sim 0.2\,meV$ or $(2K)\,k_B$

for $(5\times5)\,nm^2$ junctions, $C \sim 1\,aF$ $E_c = 80\,meV \sim (800\,K)\,k_B$

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" $\ll \frac{e^2}{2C}$

$\Rightarrow \frac{\hbar}{\Delta t} \ll \frac{e^2}{2C}$ ← lifetime of charge on capacitor

$\Rightarrow \frac{\hbar}{RC} \ll \frac{e^2}{2C}$ → $R \gg 2\frac{\hbar}{e^2}$ $\frac{h}{e^2} = 25.8$ "resistance quantum"

When $R \gg \frac{h}{e^2}$ and eV is comparable to E_c, at most one excess electron at a time can occupy the island ⇒ single electron tunneling.

With a tunable gate voltage V_g, can make a single-electron transistor. V_g tunes the energy cost for adding an electron.

Energy of island for $V=0$ $= E(n \text{ electrons}) = \frac{(ne)^2}{2C} - neV_g\frac{C_g}{C}$

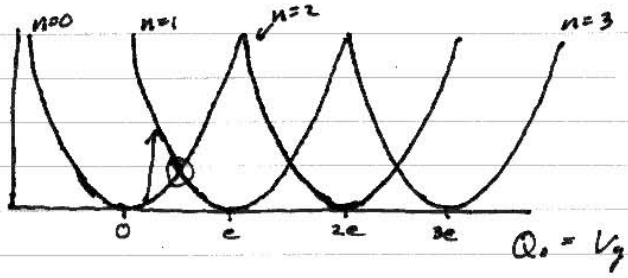
$$= \frac{1}{2C}[ne - V_g C_g]^2 - \frac{(V_g C_g)^2}{2C}$$

Now what matters is $E(n\pm1) - E(n)$ = energy required to change the number of electrons by one. This means we can ignore the last term because it is independent of n.

$\Rightarrow E(n) = \frac{1}{2C}[V_g C_g - ne]^2 \equiv \frac{1}{2C}[Q_0 - ne]^2$ where $Q_0 \equiv V_g C_g$ is called the "offset charge", but really it is due to the effects of an external potential.

Graphically:

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

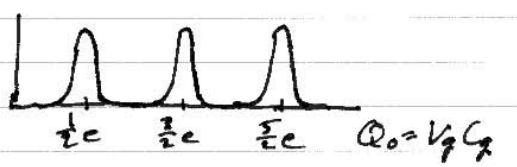


The different charge states have different energies. With a gate voltage, you can change how many electrons are in the ground state of the island

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

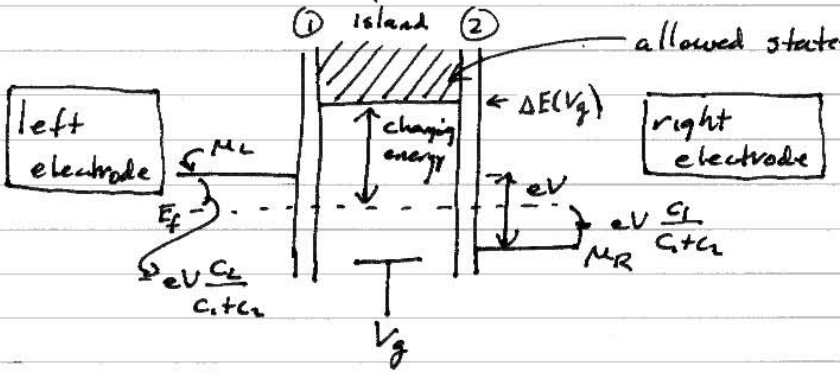
At $Q_0/e = \frac{1}{2}$ + integer values, 2 different charge states are degenerate ground states. At these values of gate voltage, charges can flow with zero Coulomb energy cost. The gate voltage has tuned away the Coulomb blockade.

Conductance $\frac{dI}{dV}$



Another picture for the same physics:

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



(I also assume that C_g is small compared to C_1 and C_2)

The threshold for tunneling for this sign of voltage is when

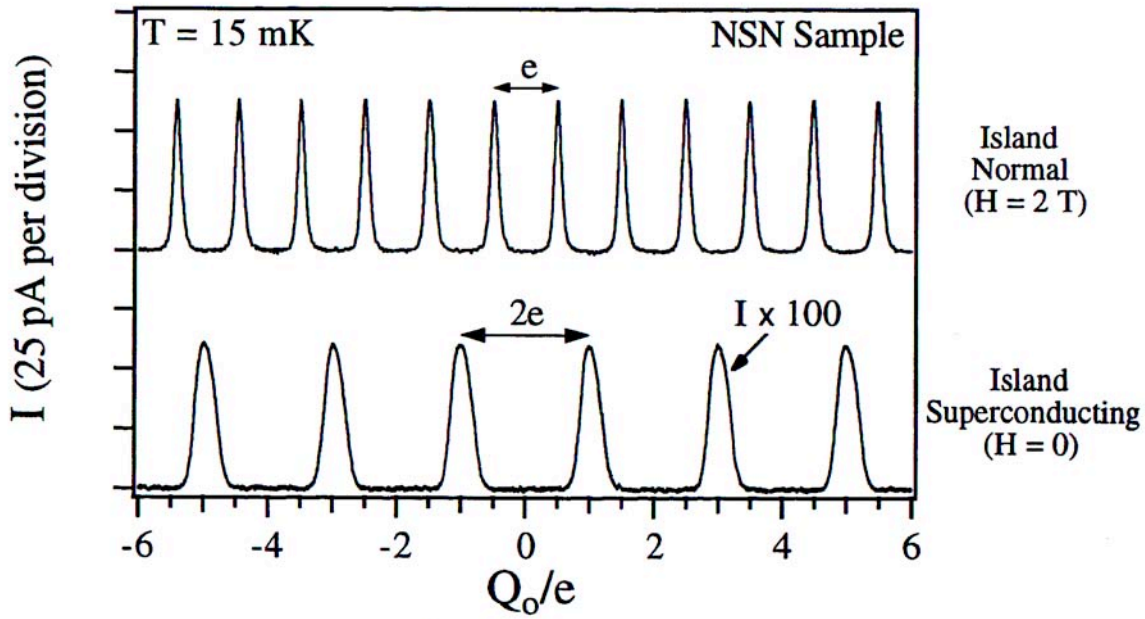
$$\mu_L = E_0(V_g) \quad \text{or} \quad eV\frac{C_1}{C_1+C_2} = \Delta E(V_g)$$

For the opposite sign of bias $eV\frac{C_2}{C_1+C_2} = \Delta E(V_g)$

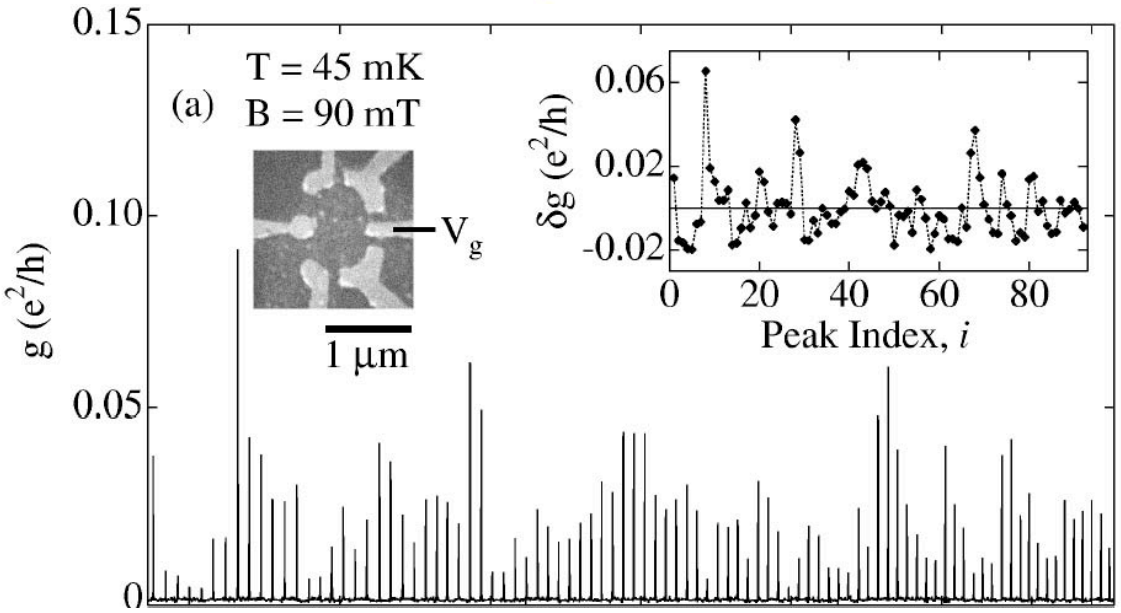
V_g shifts the energy levels on the island up or down relative to the Fermi level in the electrodes. When the levels are shifted down to E_f, there is no charging energy.

One can draw a similar diagram for the case when the Coulomb blockade is overcome by subtracting an electron

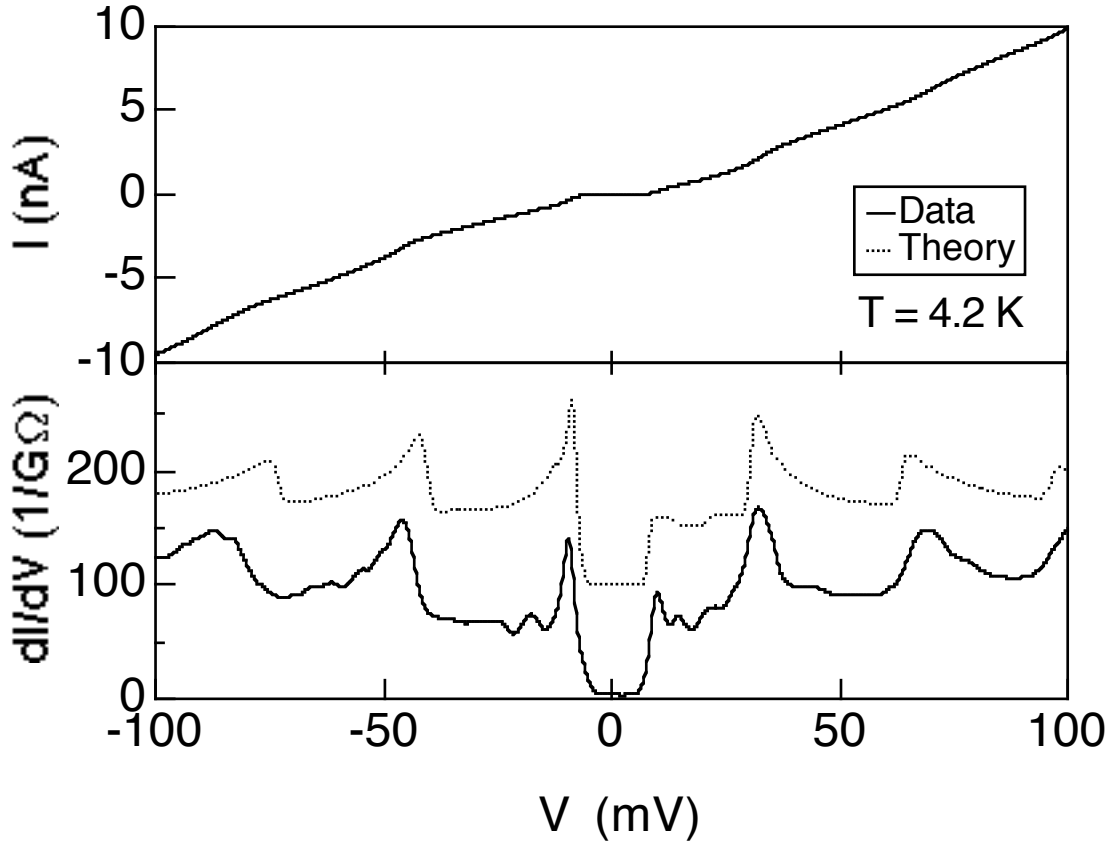
Micron-scale
Aluminum SET
(continuous DOS)
Hergenrother et al.



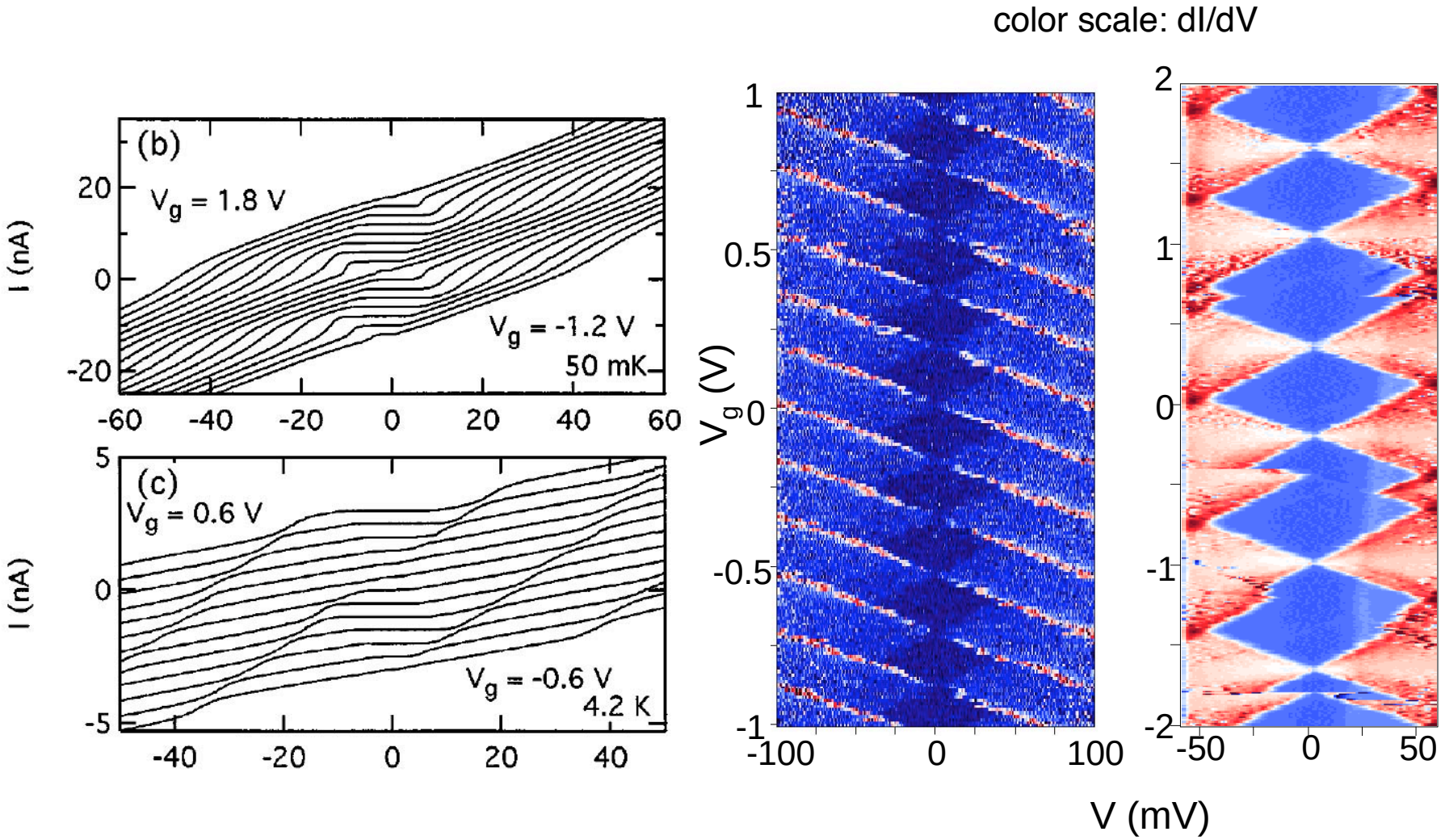
GaAs SET
(discrete DOS)
Patel et al.



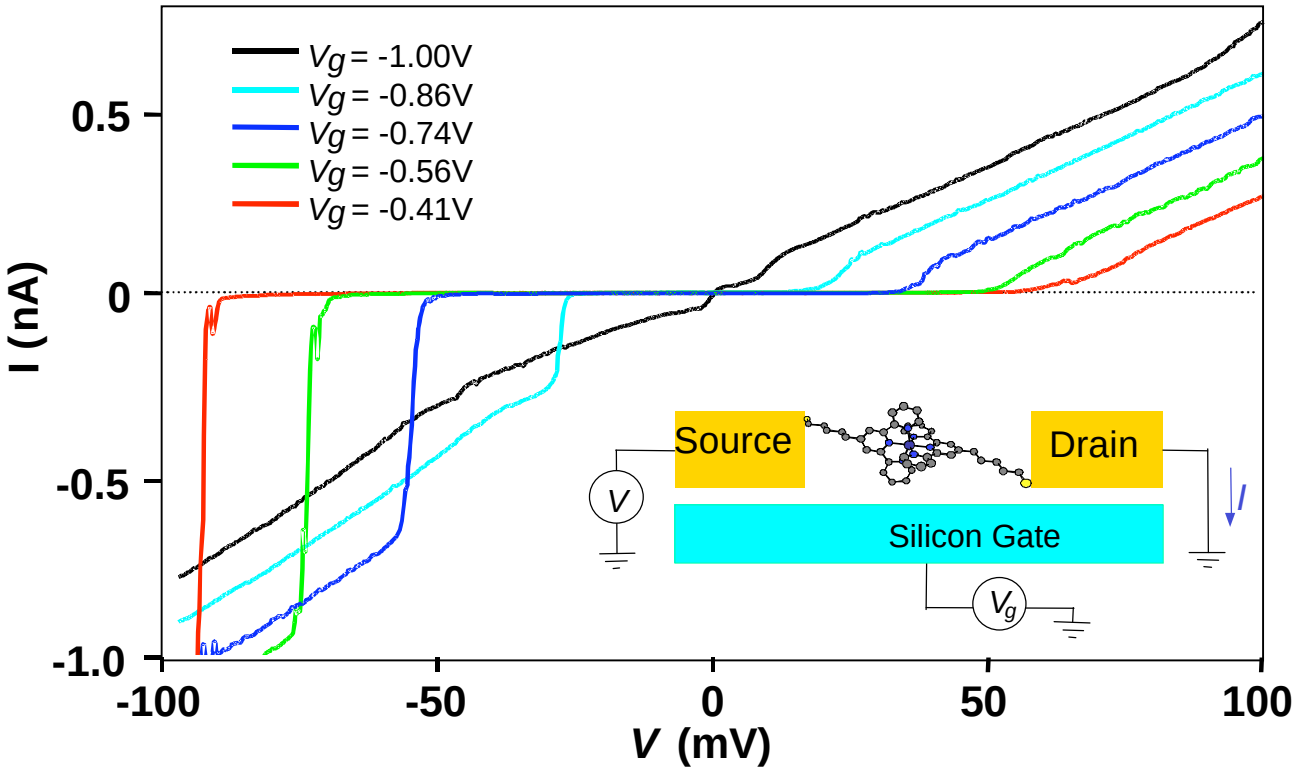
Coulomb-Staircase Curves

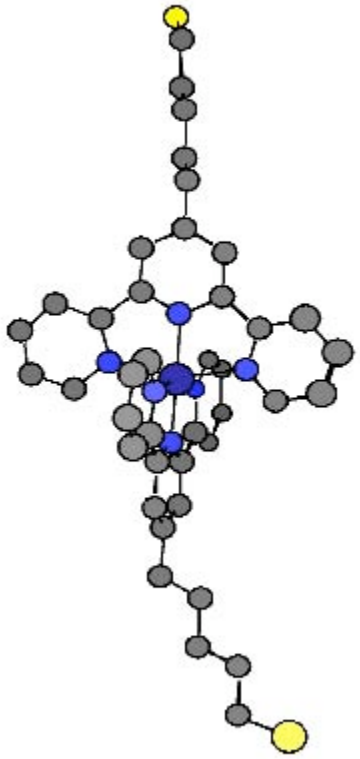


Effects of Gate Voltage on Coulomb Blockade



Coulomb-Blockade Effects in One Molecule

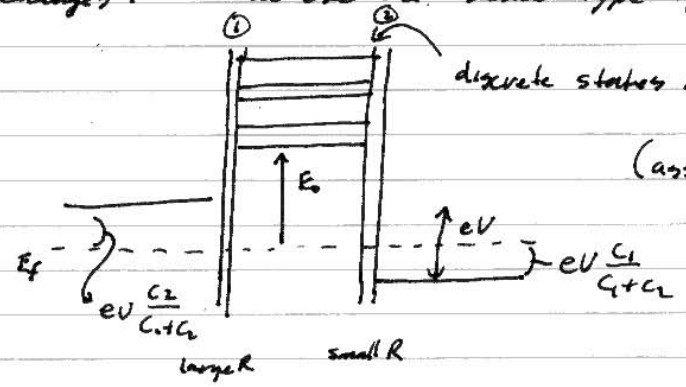




- High resistance (> megaOhms) - single electron charging.
- Coulomb blockade > 150 meV (unstable beyond this).

Quantum dots - Discrete Electronic States instead of Continuum

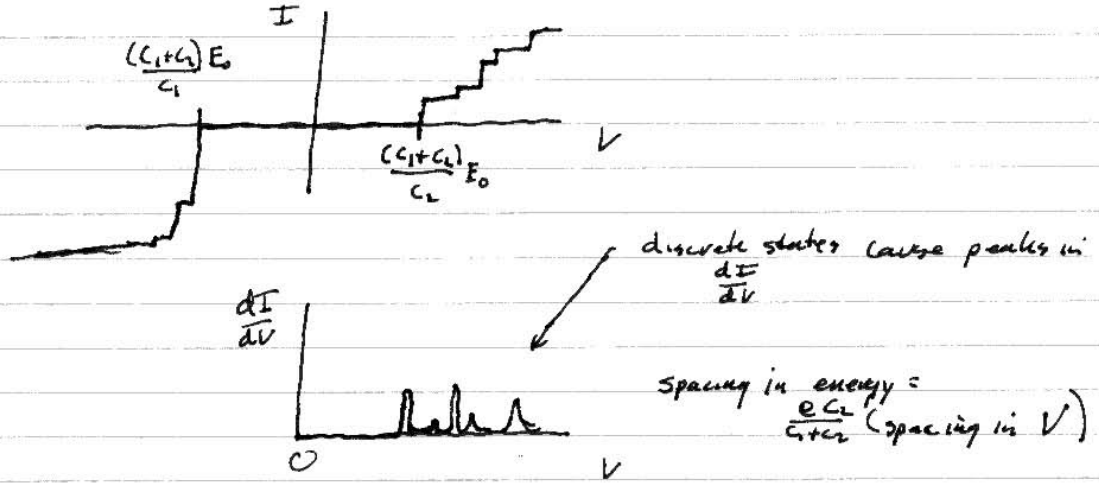
What changes? Can use the same type of picture:



discrete states now, not continuum

(assume lifetime broadening of states is $< k_B T$)

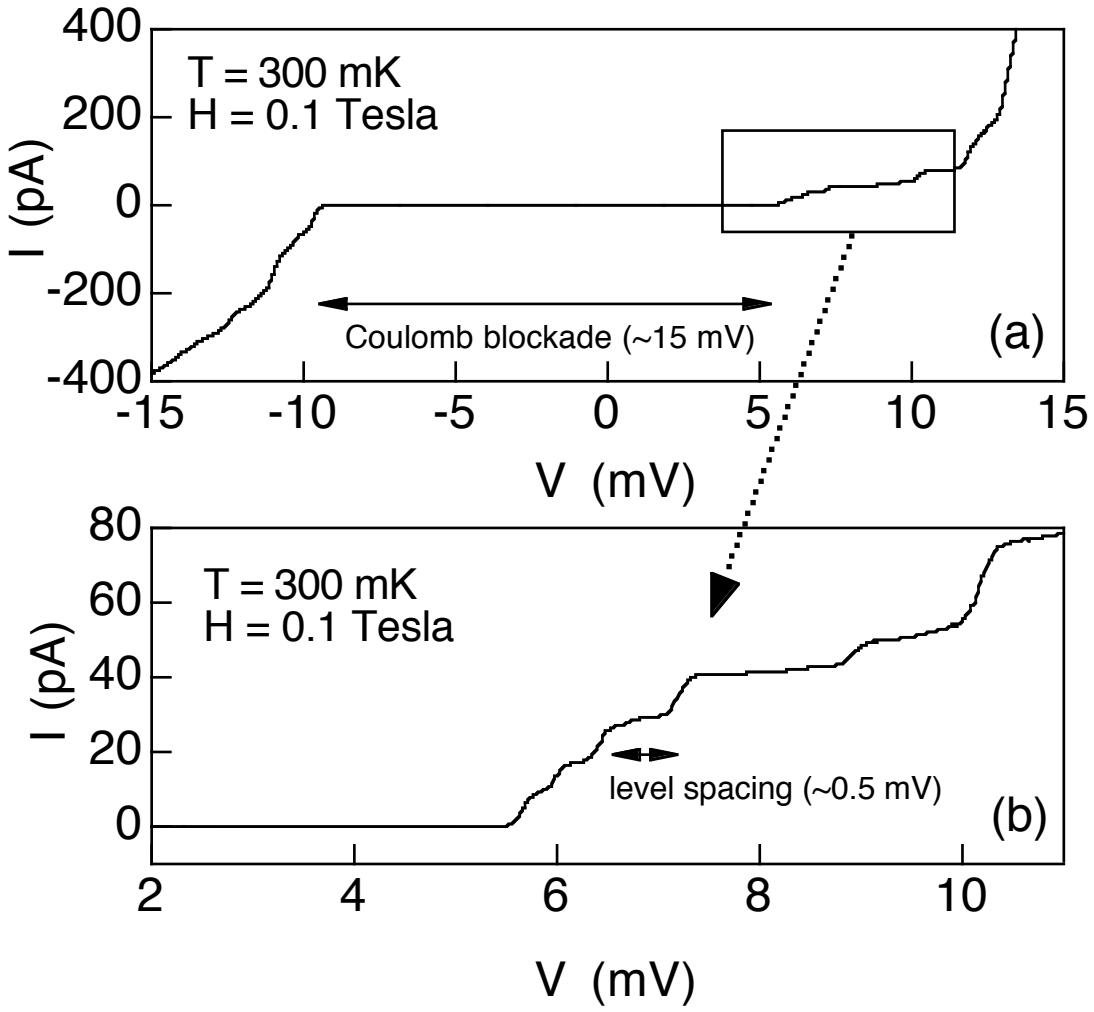
Coulomb-blockade physics is unchanged. But now the turn-on of current is changed. Instead of a continuous turn-on, the current will begin with a series of small steps, as the discrete states provide alternative channels for tunneling

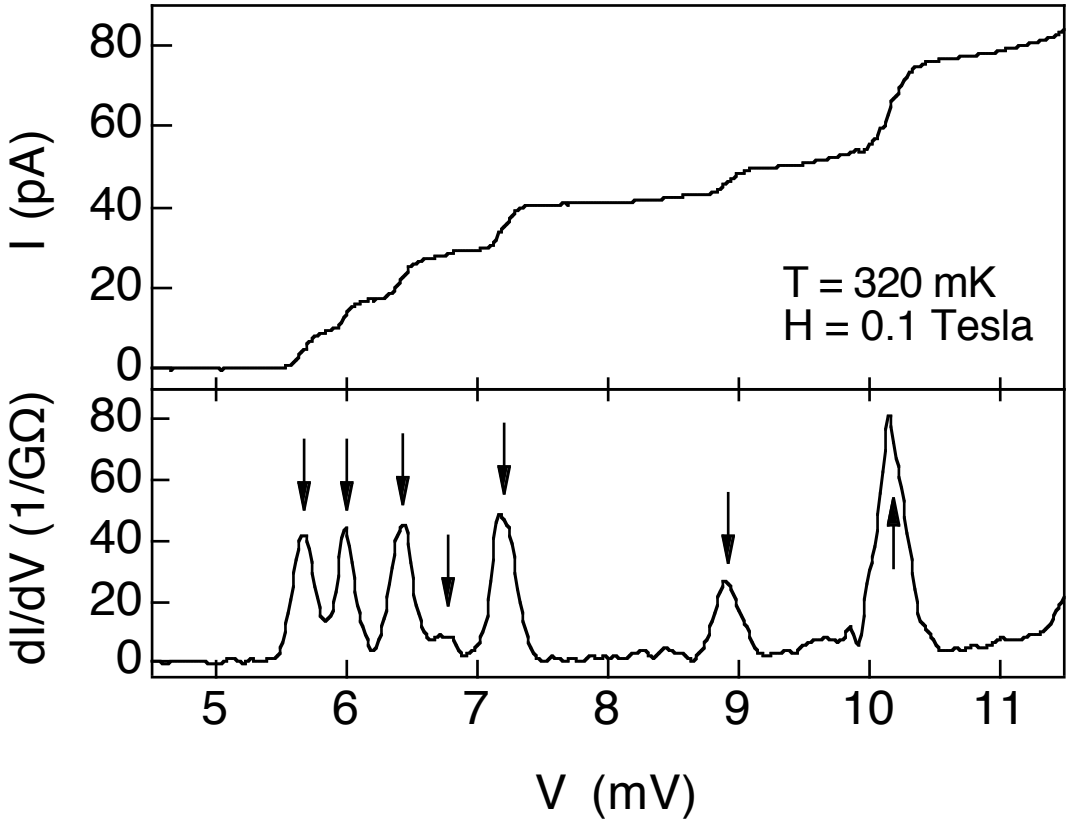


Other differences from the case of continuous states:

- spacings between Coulomb blockade peaks will not all be equal, because differences in level spacing can enter in
- the current associated with tunneling through different discrete states can vary, due to differences in individual tunneling matrix elements.

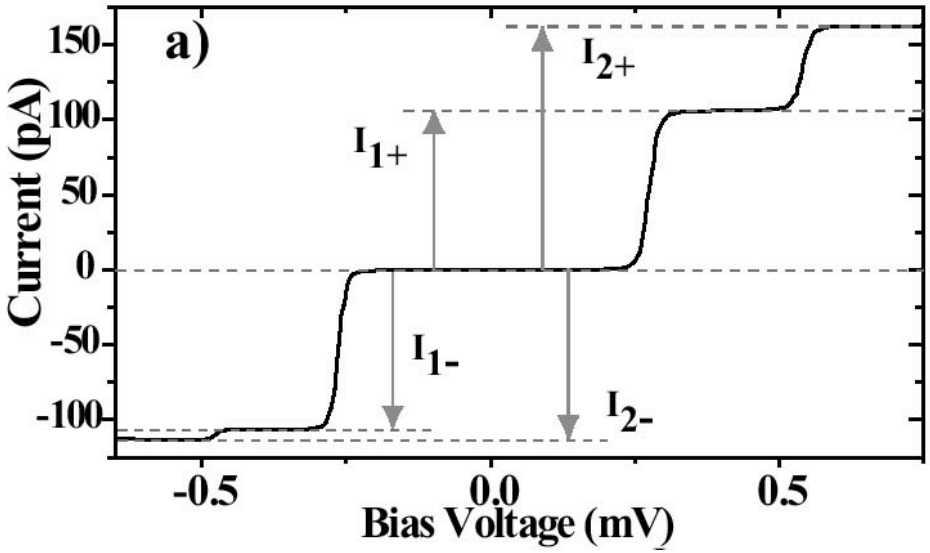
Note: In lateral semiconductor quantum dots, the tunnel barriers change with V. Makes it hard to resolve excited states in I-V curves (data)

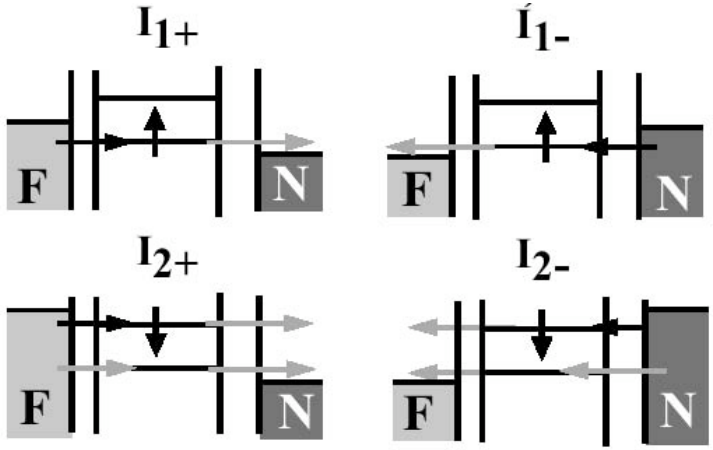

400
200
0
-200
-400
I (pA)
T = 300 mK
H = 0.1 Tesla
Coulomb blockade (~15 mV)
(a)
-15
-10
-5
0
5
10
15
V (mV)
80
60
40
20
0
I (pA)
T = 300 mK
H = 0.1 Tesla
level spacing (~0.5 mV)
(b)
2
4
6
8
10
V (mV)

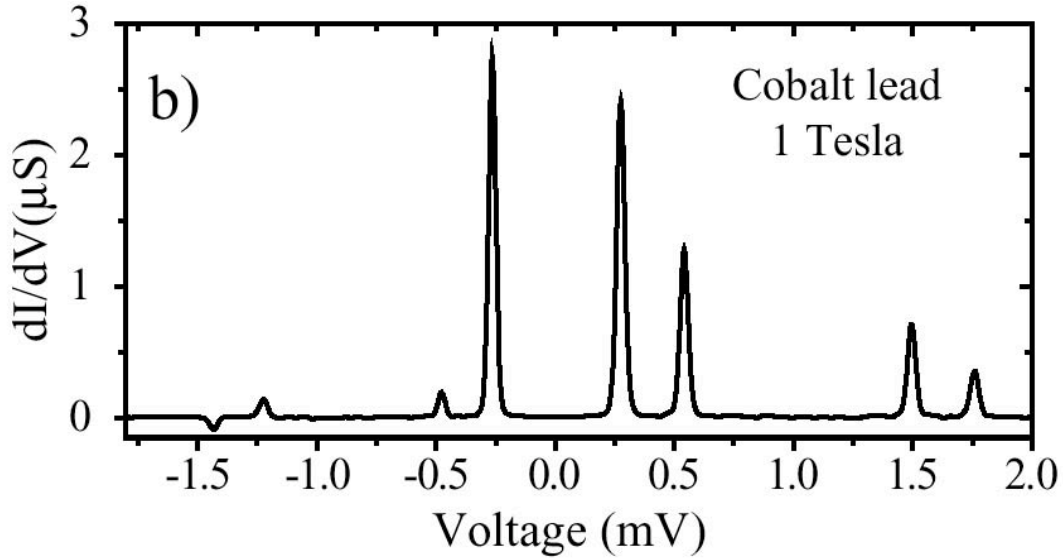

I (pA)
80
60
40
20
0
T = 320 mK
H = 0.1 Tesla
dI/dV (1/GΩ)
80
60
40
20
0
5
6
7
8
9
10
11
V (mV)

Tunneling through Individual Quantum States

$\Gamma \ll kT$

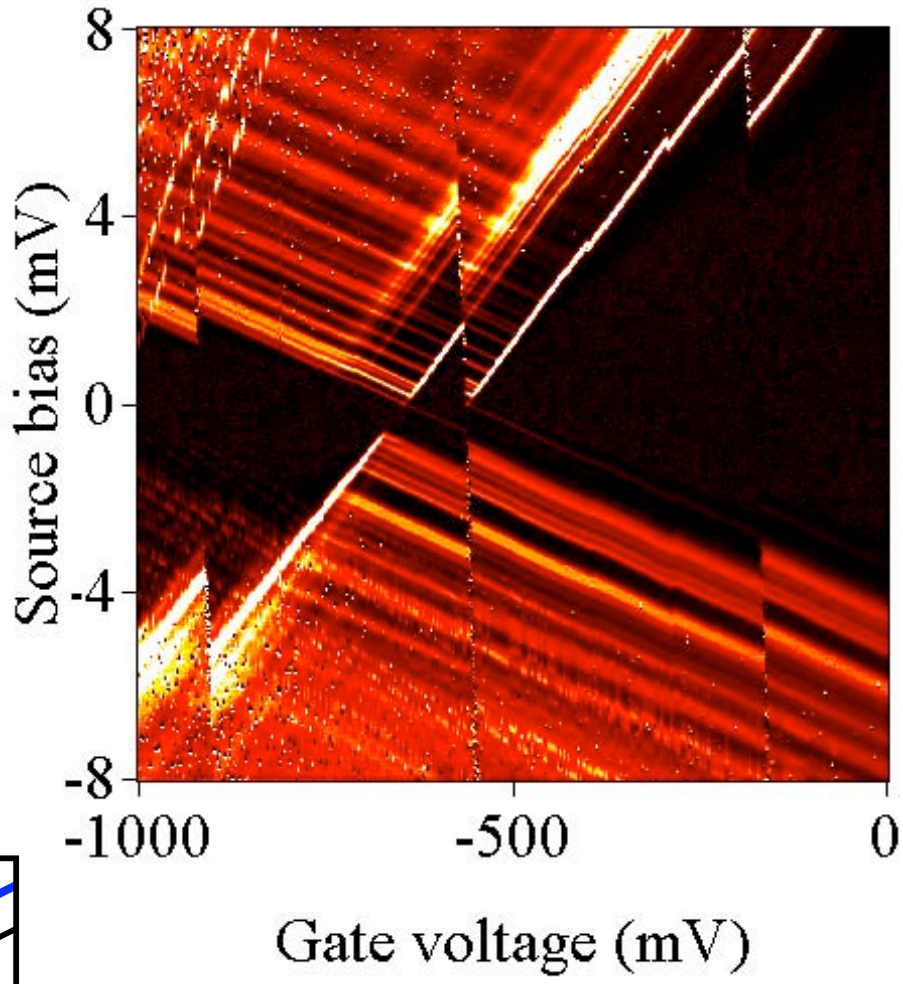


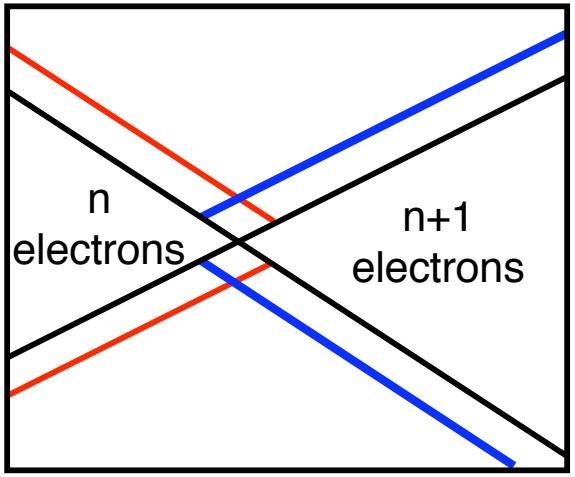




Part of Coulomb Diamond for Aluminum Particle

Color scale denotes dI/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) Superconducting pairing (Al)

4) Strong exchange and magnetic anisotropy forces (ferromagnetic Co)

Basics of Spin in Quantum Dots (nonmagnetic)

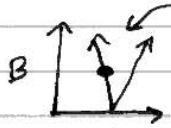
It is possible to tell if a nonmagnetic dot has an even or odd number of electrons, even if the number of electrons is in the 1000's.

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:

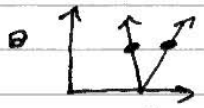
for electron addition

even → odd
both states empty and available for tunneling

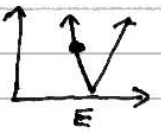


odd → even. lower E state is blocked by the odd electron. Only one state available for tunneling

for electron subtraction:



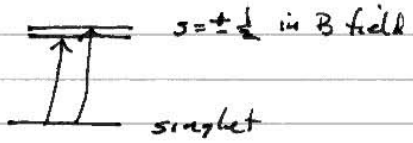
even → odd
both states available



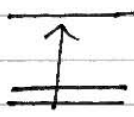
odd-even. only filled state is available.

Alternative Many-body picture: (holds as long as the ground state is spin 0 for even electrons, spin ½ for odd)

even → odd



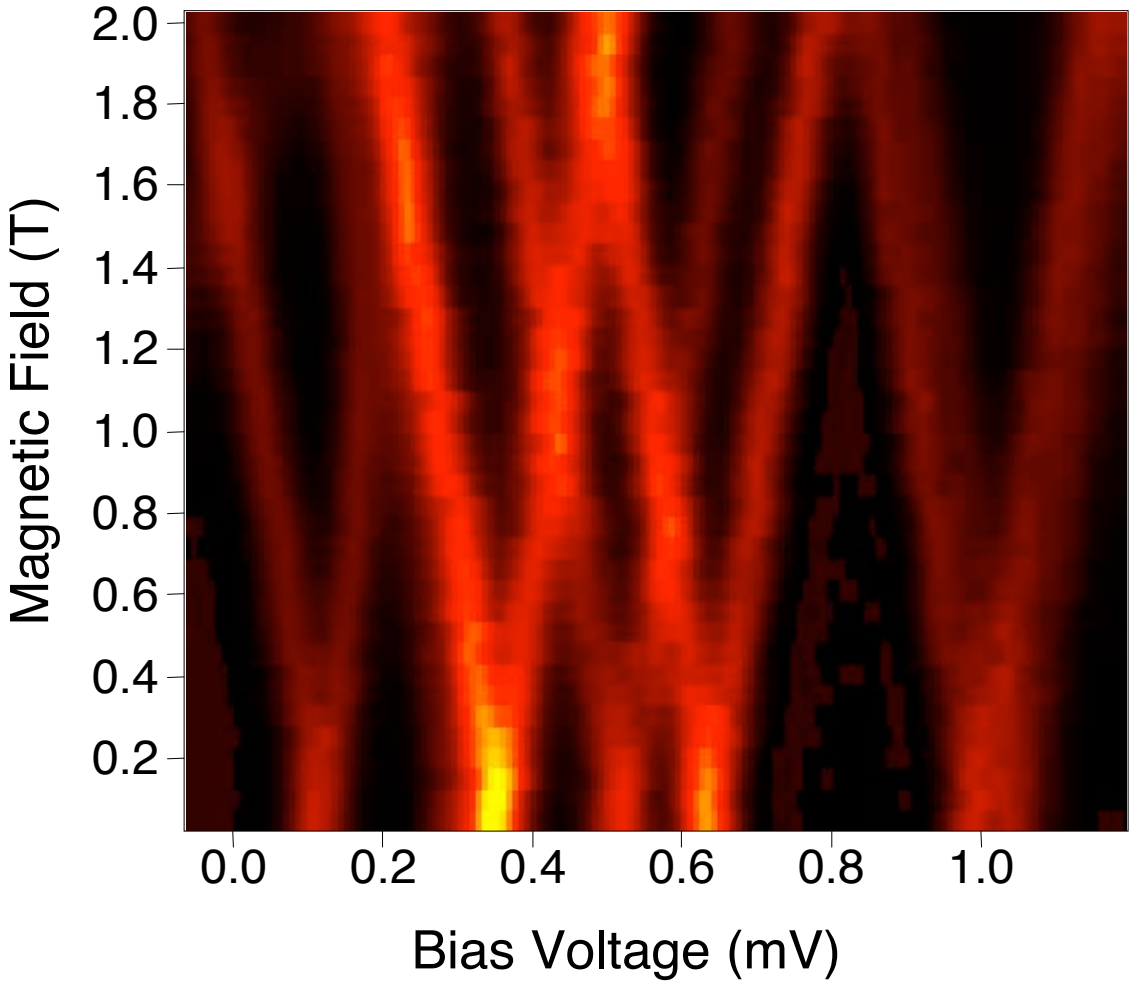
odd → even



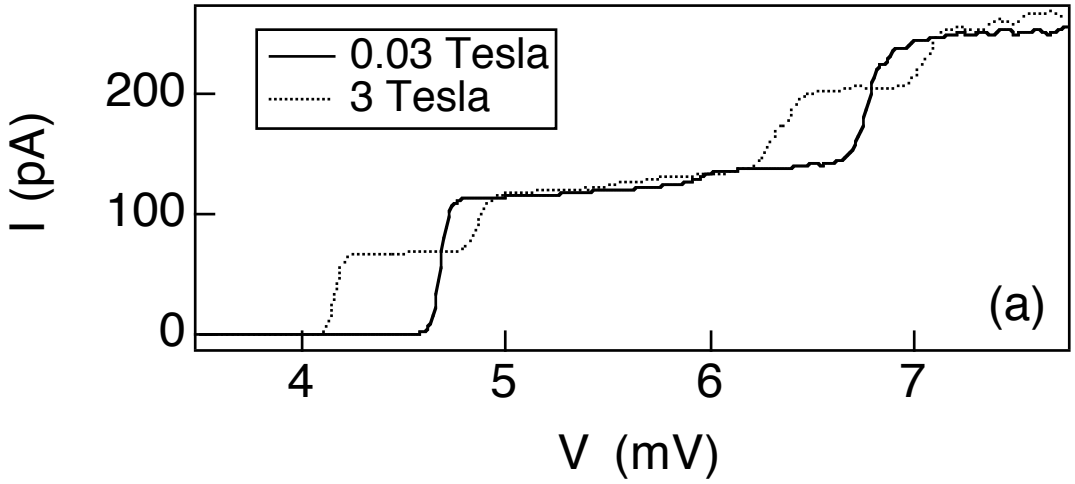
only lower state occupied.
Only one transition is visible

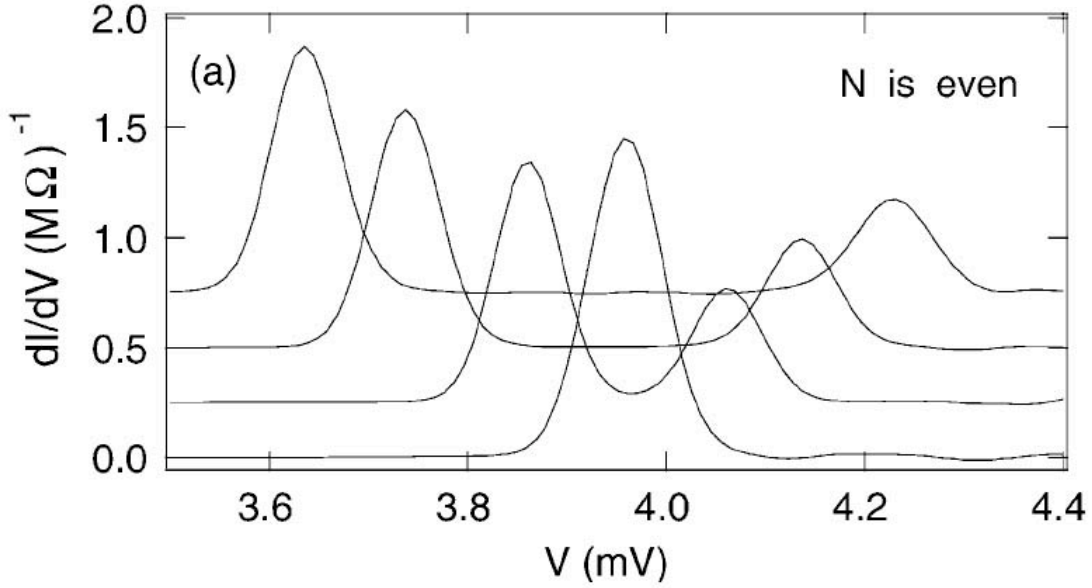
(<Show data for Al + Cu>)

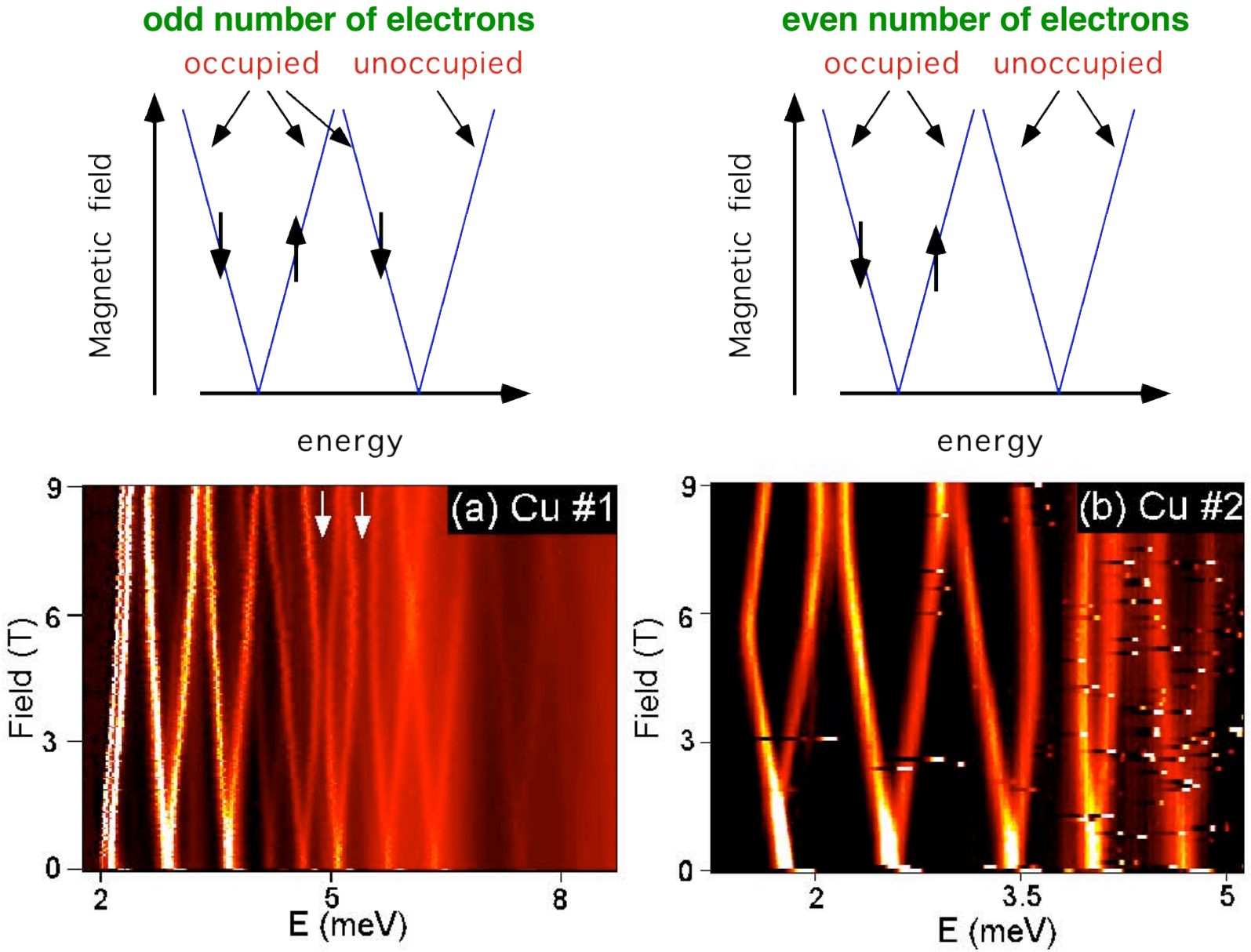
Magnetic-Field Dependence of Aluminum Levels



g = 2.0 ± 0.1 for Al.


0.03 Tesla
3 Tesla
I (pA)
200
100
0
4
5
6
7
V (mV)
(a)

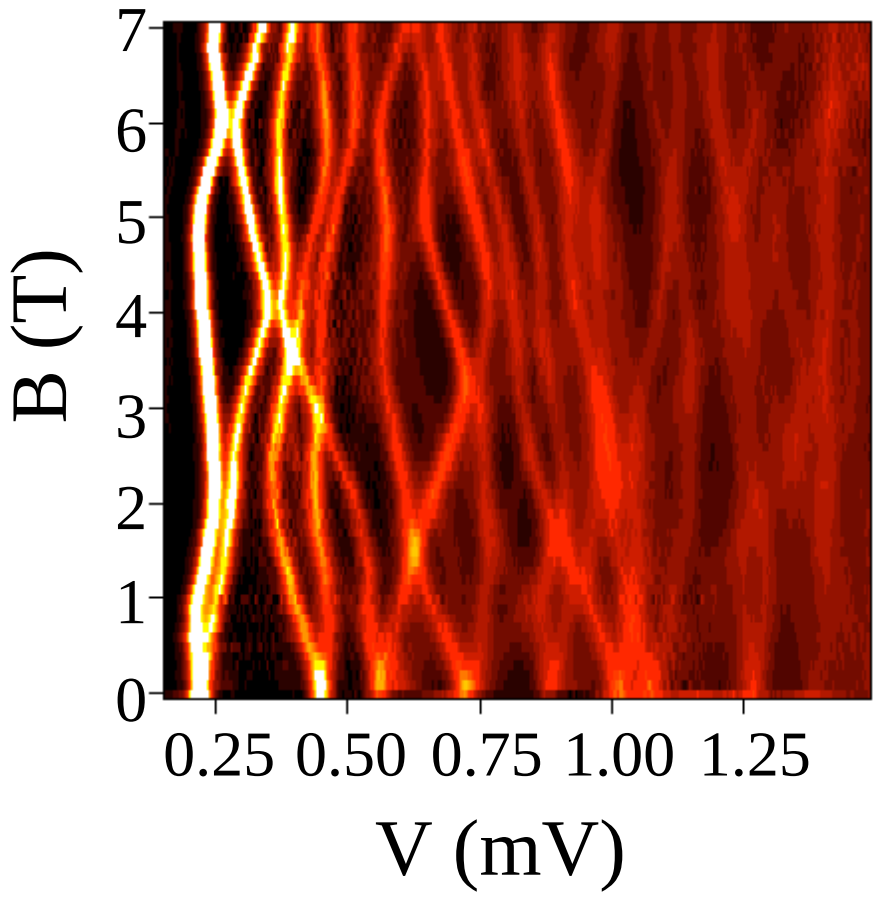

(a)
N is even
dI/dV (MΩ)$^{-1}$
2.0
1.5
1.0
0.5
0.0
3.6
3.8
4.0
4.2
4.4
V (mV)



g = 1.30 to 1.82

<g> = 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:

① $g < 2$ for all states
② g differs from state to state
③ 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 $\sim Z^4$

References for spin-orbit scattering in metals:
R. J. Elliot Phys Rev. 96, 266 (1954)
J Fabian and S. Das Sarma, J. Vac. Sci. Technol. B 17, 1708 (1999)

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 $\downarrow$:

$$|n,\text{"}\uparrow\text{"}\rangle = \alpha_n |\uparrow\rangle + \beta_n |\downarrow\rangle$$
$$|n,\text{"}\downarrow\text{"}\rangle = -\beta_n^* |\uparrow\rangle + \alpha_n^* |\downarrow\rangle$$

→ Related by time-reversal symmetry

① g-factor for spin Zeeman splitting:

$$g_n = 2\langle n,\text{"}\uparrow\text{"}|\sigma_z|n,\text{"}\uparrow\text{"}\rangle = 2(|\alpha_n|^2 - |\beta_n|^2) = 2(1-2|\beta_n|^2) \leq 2.$$

(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. Sone, J. Phys. Soc. Jpn. 42, 1457 (1977))

② <u>fluctuations in g-factors</u>

$$g_n = 2\left[1 - 2\sum_{m\neq n} \frac{|\langle\psi_{m\downarrow}|H_{so}|\psi_{n\uparrow}\rangle|^2}{(E_n - E_m)^2}\right]$$

matrix elements will vary between the unperturbed single-electron states ψ_m & ψ_n

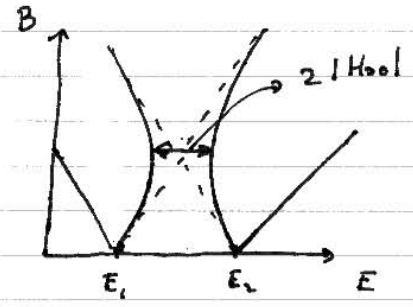
the energy denominators will also vary.

⇒ for different orbital states n, g_n can be different.

③ <u>avoided crossings</u>

in crossing region, 2×2 Hamiltonian

$$\mathcal{H} = \begin{pmatrix} E_1 + \frac{1}{2}g\mu_B B & H_{so} \\ H_{so}^* & E_2 - \frac{1}{2}g\mu_B B \end{pmatrix}$$



More sophisticated theory of Spin-Orbit consequences - Random Matrix Theory

references: Brouwer et al., Phys Rev. Lett 85, 369 (2000)
Matveev et al., Phys Rev. Lett 85, 2789 (2000)
Gorokhov and Brouwer, Phys Rev. B 69, 155417 (2004)

$$\mathcal{H}(\lambda) = S \otimes \mathbf{1} + \frac{i\lambda}{\sqrt{4N}} \sum_j A_j \otimes \sigma_j$$

S = $N \times N$ symmetric random matrix
A_j = $N \times N$ antisymmetric random matrix
$\lambda \sim (\tau_{so}\delta)^{-1/2}$, strength of spin-orbit interaction

Theory can predict:

① Full probability distribution of g factors for a given value of λ - not just in small-λ perturbative regime

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

$$(\text{Zeeman splitting})^2 = \frac{\mu_B^2}{4}\left(g_1^2 B_1^2 + g_2^2 B_2^2 + g_3^2 B_3^2\right)$$

where B_1, B_2, and B_3 are along the principle axes
⇒ ellipsoid (most complicated form allowed by symmetry for $S = \frac{1}{2}$)

principle-axis g-factors predicted to vary by factors of 4 or 5 in regime of moderate SO. ($\lambda \sim 1$) - not a small effect!

Physical picture:

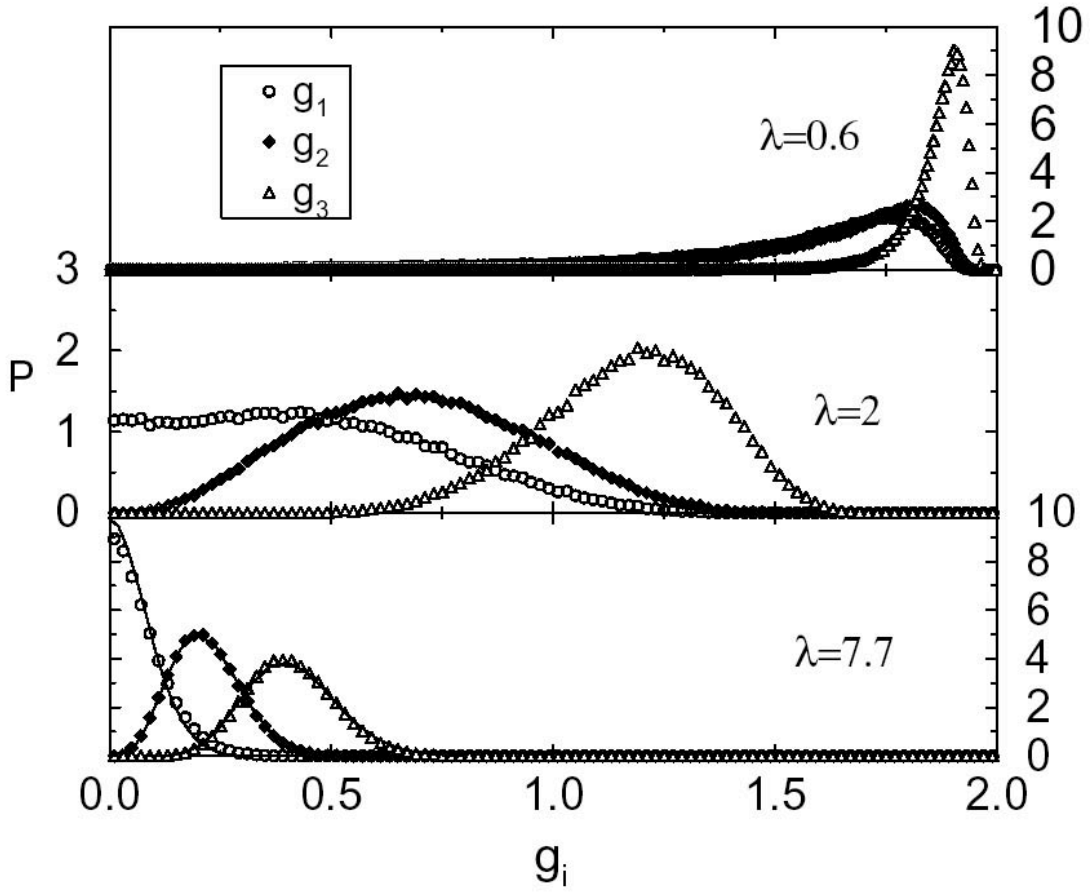
The orbital wavefunctions in a metal particle are highly oscillatory, essentially random functions. When the orbits are coupled to spin, the spin properties also take on random properties. These will be anisotropic in space, so the g-factor will depend on the direction of the magnetic field

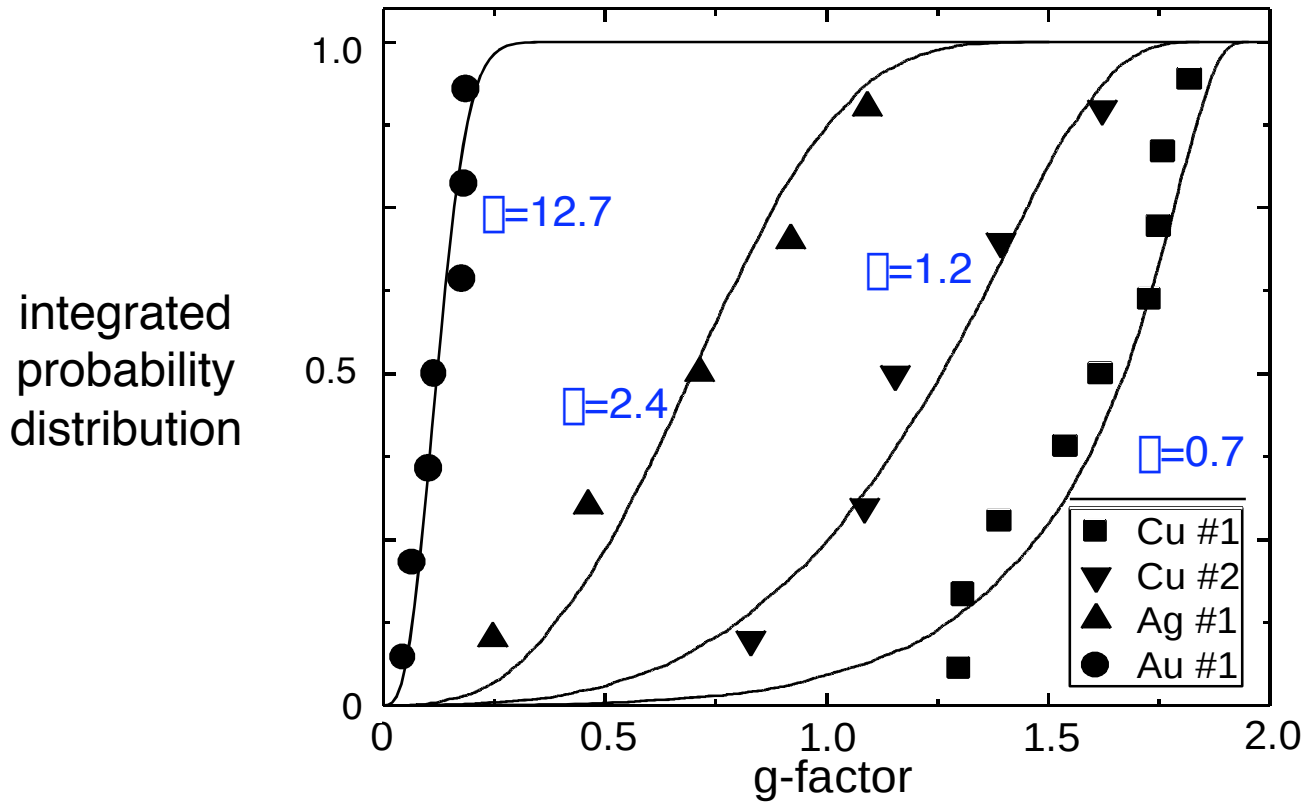
⇒ The principle axes for different states in one particle should be random, not associated with shape or crystal axes.

g_1, g_2, g_3 emerge from a matrix diagonalization. The spread in their values is related to eigenvalue repulsion.

cont'd ↓

Random Matrix Theory Predictions: Distributions of the principal g-factors for different strengths of spin-orbit interaction.

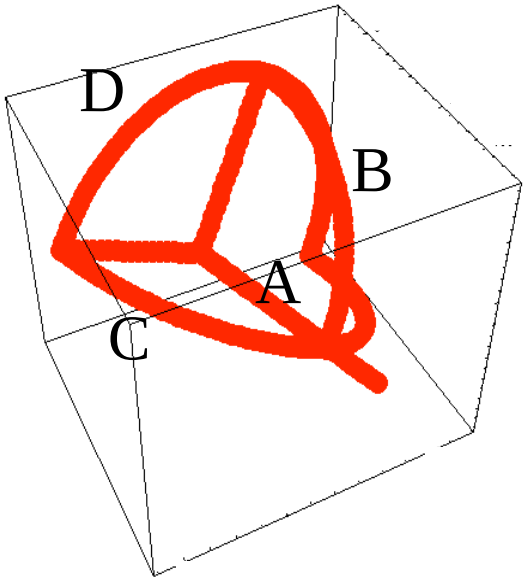


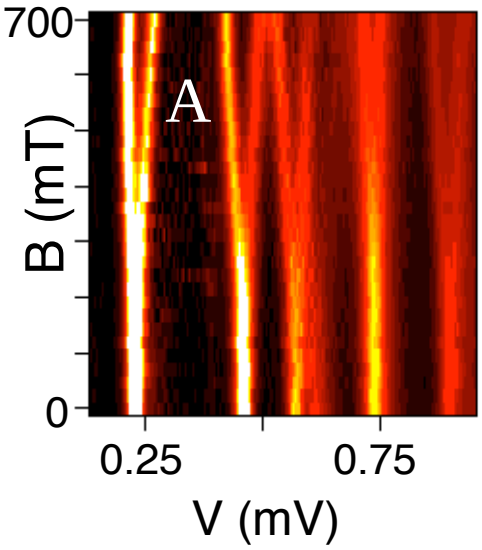


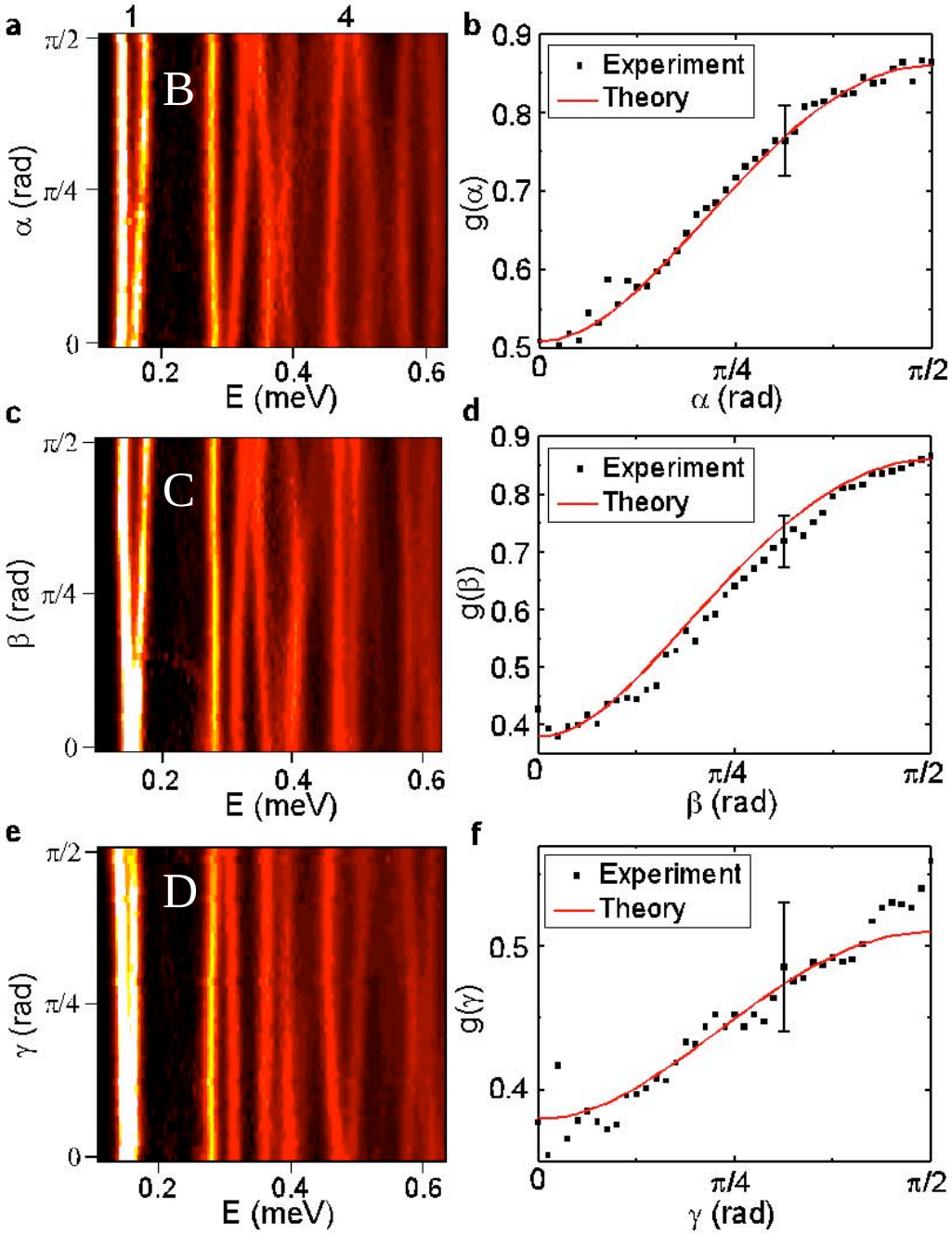
• With one fitting parameter per sample, both the average g-factor and the standard deviation are described well.

• Significant differences in spin-orbit strength even for particles made of the same metal.

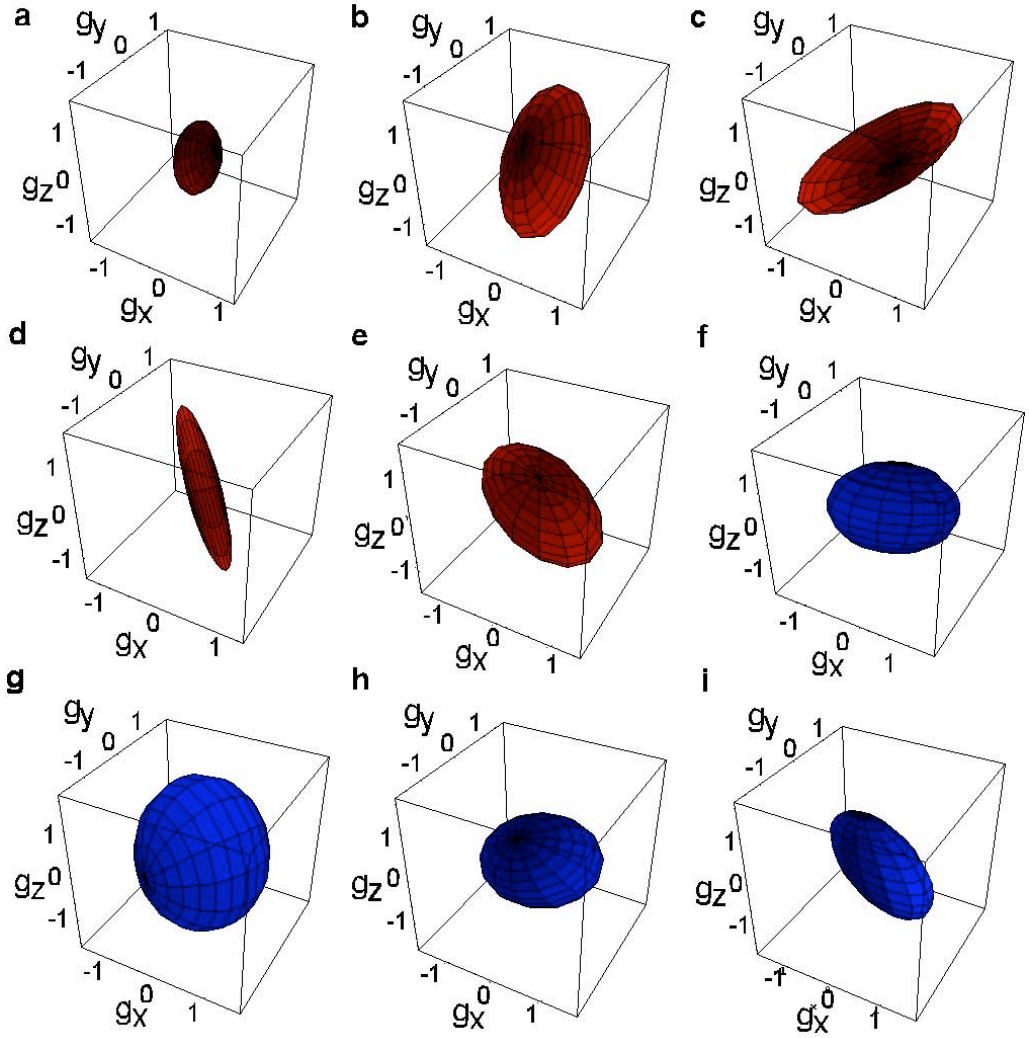
Anisotropy of g-factors







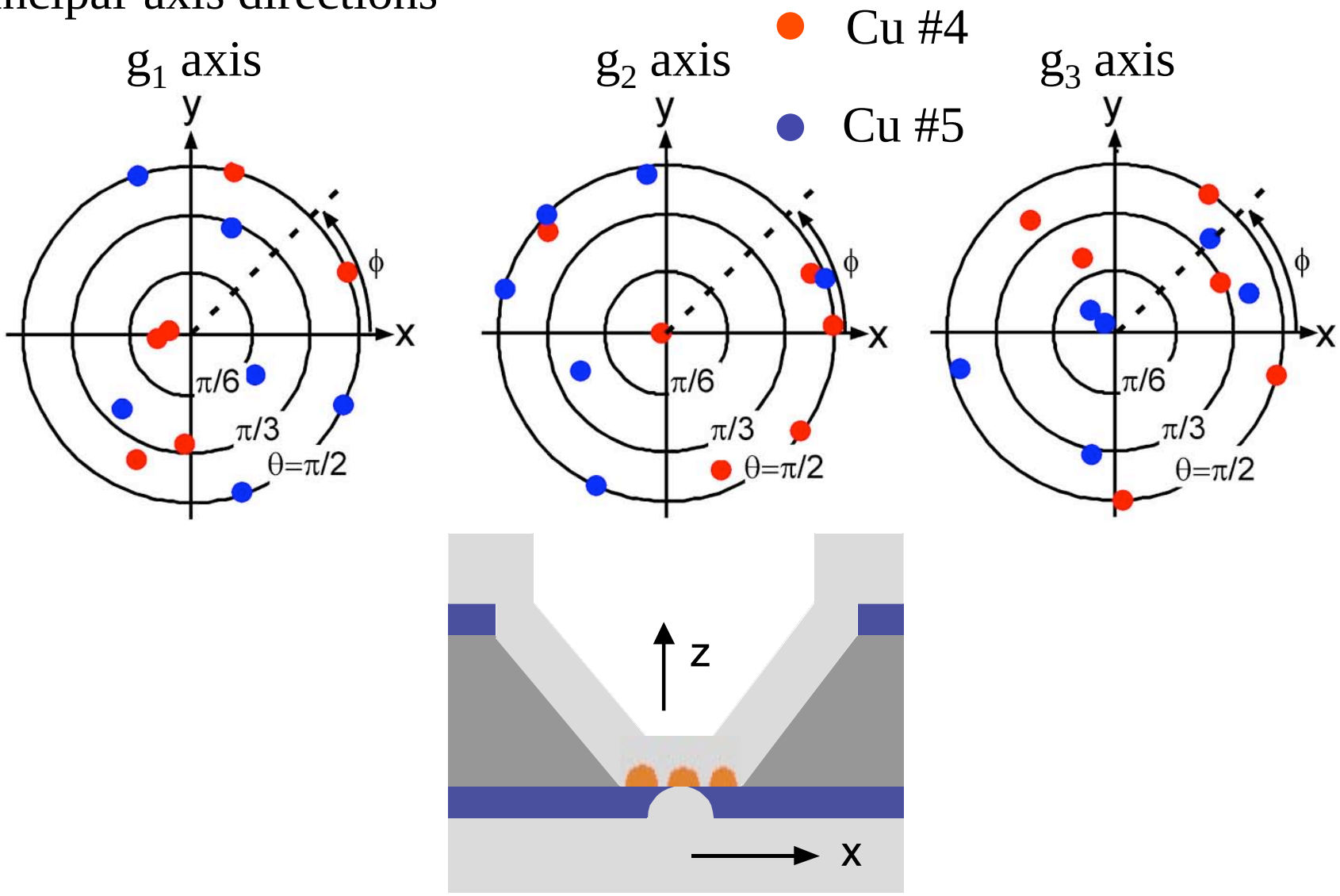
Variations from Quantum State to Quantum State



Red: Cu#1

Blue: Cu#2

Principal-axis directions



Principal-axis directions are randomly oriented in space.

Check agreement between experiment and RMT predictions

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

Cu #4
□=1.8

	Exp.	RMT
$<g_1>$	1.3	1.25
$<g_2>$	0.8	0.76
$<g_3>$	0.4	0.52

Cu #5
□=1.1

	Exp.	RMT
$<g_1>$	1.6	1.59
$<g_2>$	1.2	1.12
$<g_3>$	0.9	0.96

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.*)

$$\langle g^2 \rangle = \frac{3}{\hbar}\tau_{so}\tau + \tau\frac{l}{L}$$, where $\tau \sim 1$

spin contribution

orbital contribution

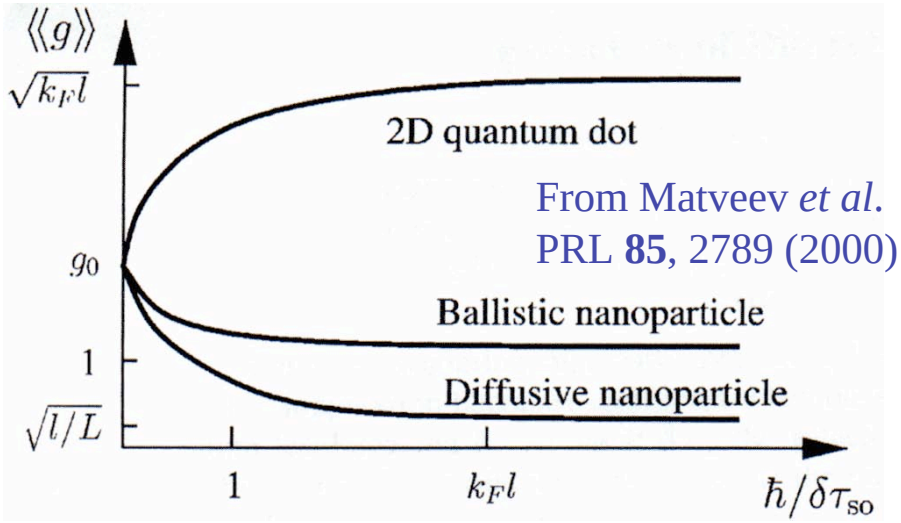
For a ballistic nanoparticle, $l \sim L$

$\longrightarrow$ $2 \geq \langle g \rangle \geq 1$

For a diffusive nanoparticle, $l < L$

$\longrightarrow$ $\langle g \rangle \to 2$

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 τ_{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 $\ll kT <$ level spacing $<$ charging energy. This corresponds to relatively high resistance tunnel junctions with $R \gg h/e^2$
  (For lower-resistance junctions, cotunneling and Kondo physics come into play.)

- The study of spin Zeeman splitting is a good tool 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".

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