Lectures: Condensed Matter II

1 – Quantum dots

2 – Kondo effect: Intro/theory.

3 – Kondo effect in nanostructures

Luis Dias – UT/ORNL

Lecture 1: Outline

- Introduction: From atoms to “artificial atoms”.
- What are Quantum Dots?
- Confinement regimes.
- Transport in QDs: General aspects.
- Transport in QDs: Coulomb blockade regime.
- Transport in QDs: Peak Spacing.

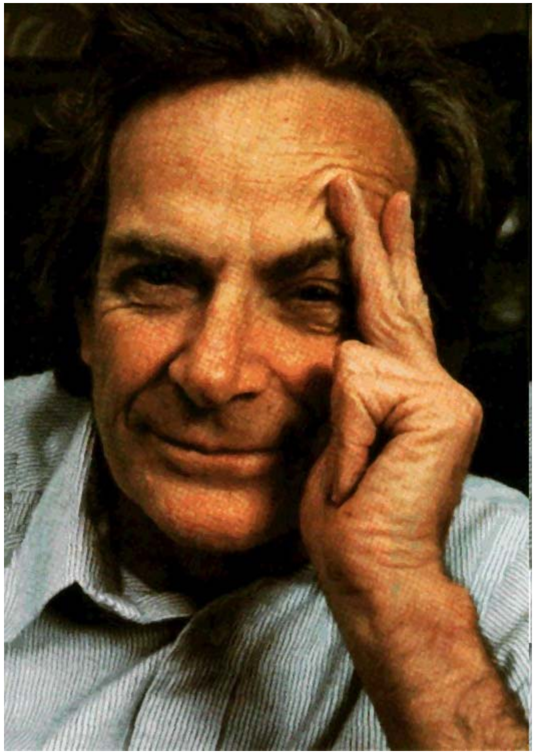
“More is Different”

“ The behavior of large and complex aggregates of elementary particles, it turns out, is not to be understood in terms of simple extrapolation of the properties of a few particles.

Instead, at each level of complexity entirely new properties appear and the understanding of the new behaviors requires research which I think is as fundamental in its nature as any other.“

Phillip W. Anderson, “More is Different”, *Science* **177** 393 (1972)

“More is Different?”



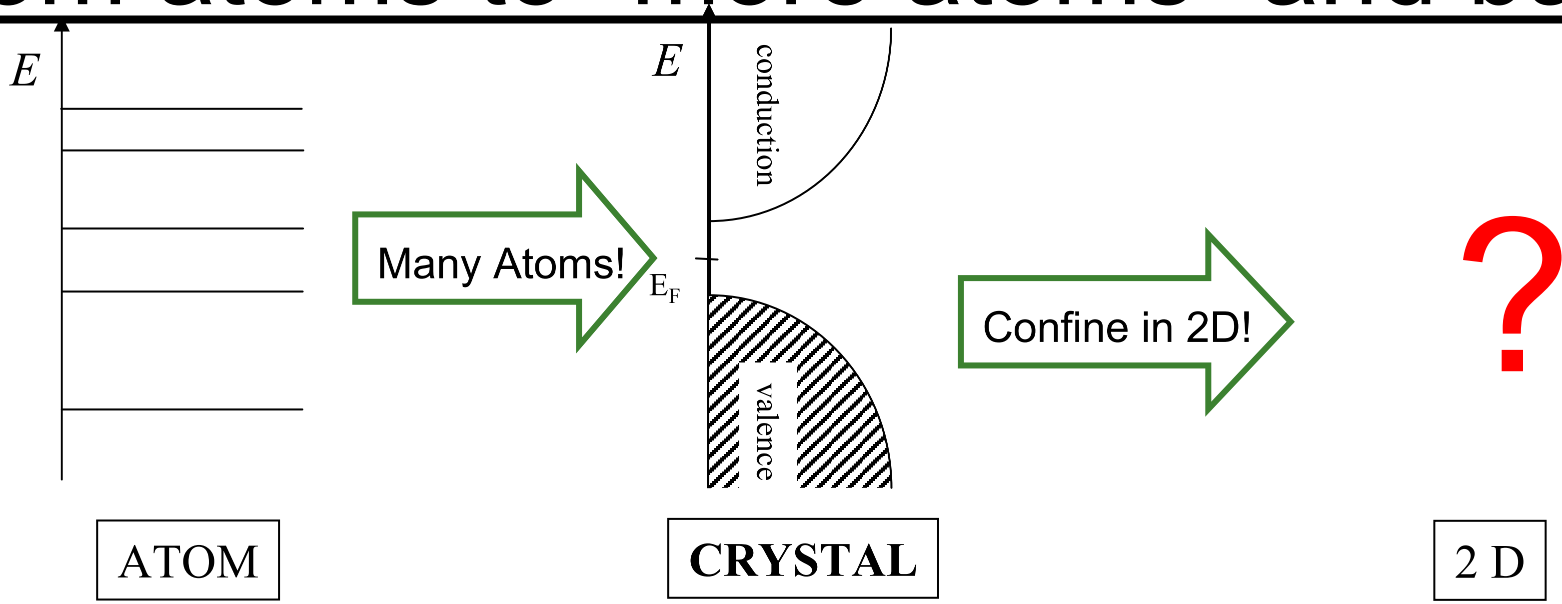
“If, in some cataclysm, all of scientific knowledge were to be destroyed, and only one sentence passed on to the next generation of creatures, what statement would contain the most information in the fewest words?

I believe it is the *atomic hypothesis* that ***All things are made of atoms-little particles that that move around in perpetual motion, attracting each other when they are a little distance apart, but repelling upon being squeezed into one another.***“

In that one sentence, there is an enormous amount of information about the world, if just a little imagination and thinking are applied.

R. P. Feynman – *The Feynman Lectures*

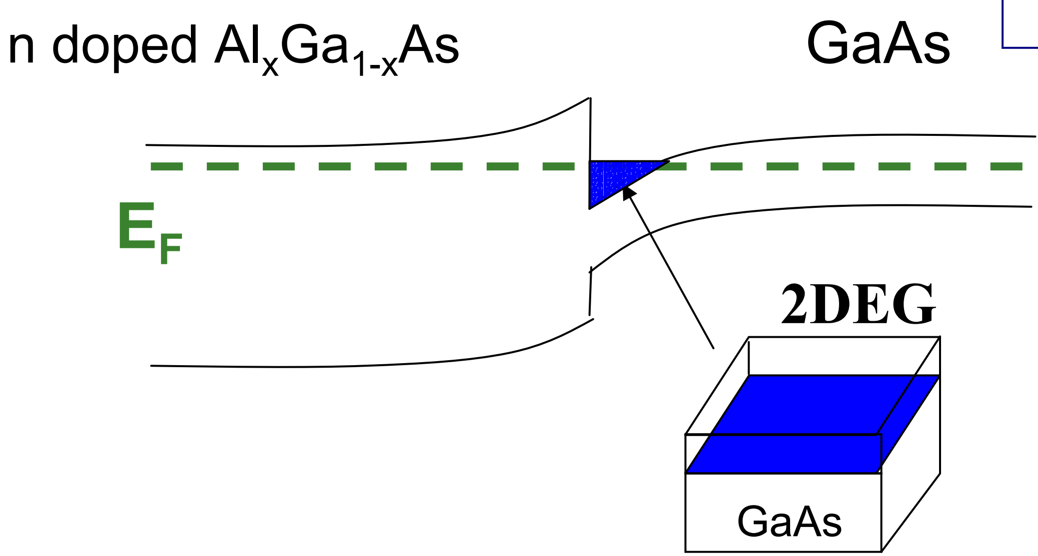
From atoms to “more atoms” and back

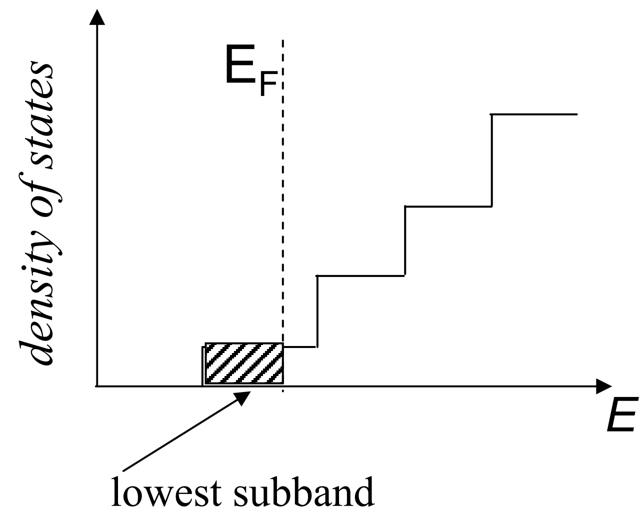


Confined in 1 direction: 2D system

If a thin enough 2D plane of material (containing free electrons) is formed the electrons can be confined to be two dimensional in nature. Experimentally this is usually done in semiconductors.

$$\rho_{2D}(E) = \frac{m}{\pi\hbar^2}\sum_i \Theta(E - E_i)$$

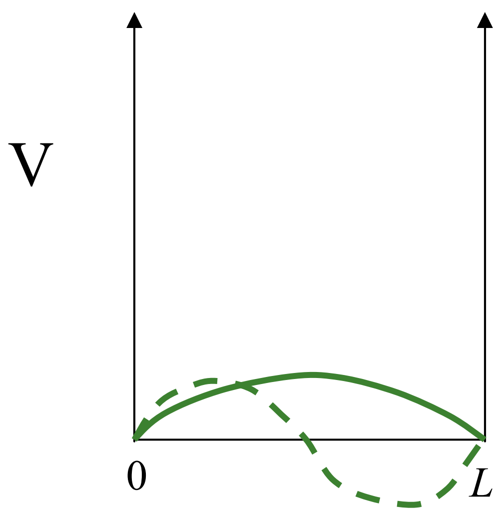




e.g. by growing a large band gap material with a smaller band gap material you can confine a region of electrons to the interface - TWO DIMENSIONAL ELECTRON GAS (2DEG).

More details: Ando, Fowler, Stern *Rev. Mod. Phys.* **54** 437 (1982)

Confinement: Particle in a box



1-d box: wavefunction constrained so that

L = N λ / 2 or
k = 2 π / λ = N π / L

Energy of states given by Schrodinger Equation:

$$\hat{H}\Psi = E\Psi$$

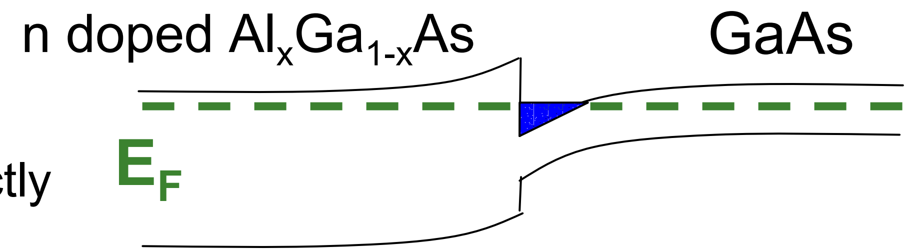
$$\boxed{E_N = \frac{\hbar^2 k_N^2}{2m} = \frac{\hbar^2 \pi^2 N^2}{2mL^2}}$$

Typical semiconductor dots:
L in nm, E in meV range

As the length scale decreases the energy level spacing increases.

$$\boxed{\Delta E = E_{N+1} - E_N \propto \frac{1}{L^2}}$$

Confined in 1 direction: 2D system



Provided the electrons are confined to the lowest subband the electrons behave exactly as if they are two-dimensional i.e. obey 2D Schrodinger equation etc.

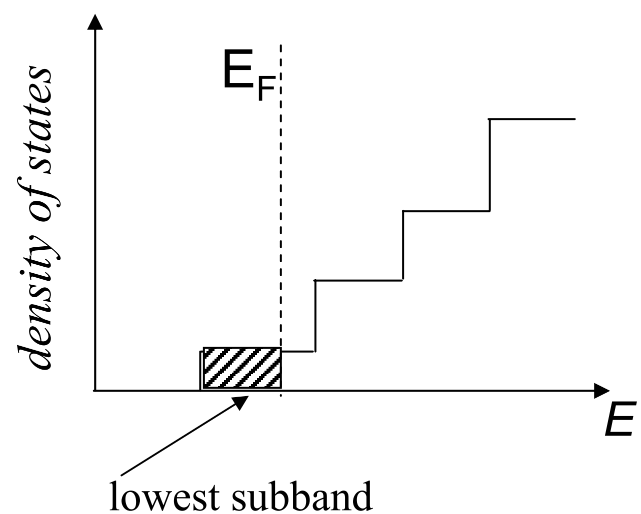
2DEG: Rich source of Physics.

•Nobel Prize in Physics in 1985 to von Klitzing for the Quantum Hall Effect (QHE),

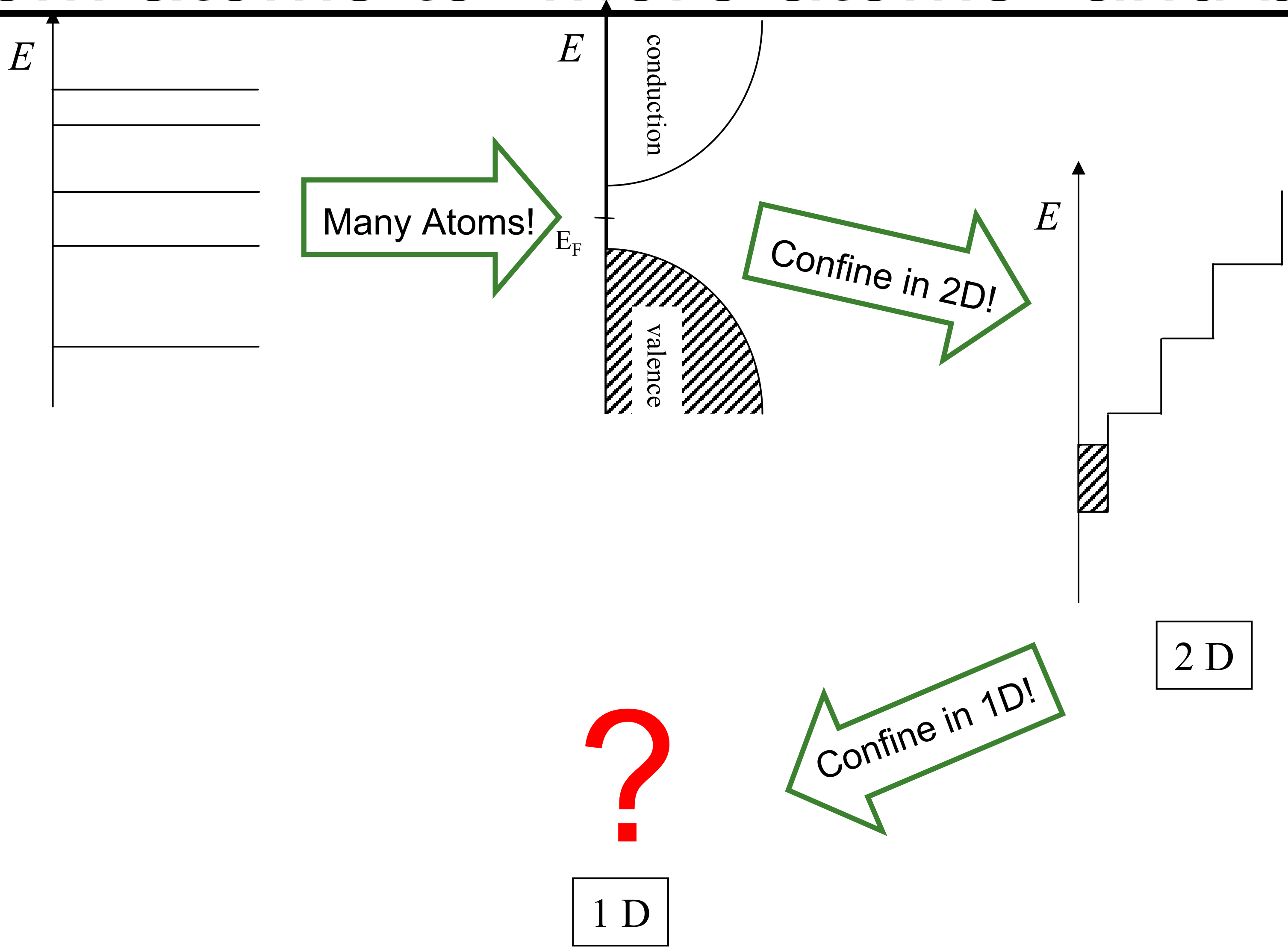
•1998 to Tsui, Stormer and Laughlin for the Fractional QHE

•Semiconductor heterostructures, lithography

•Applications (lasers, QHE, etc.)

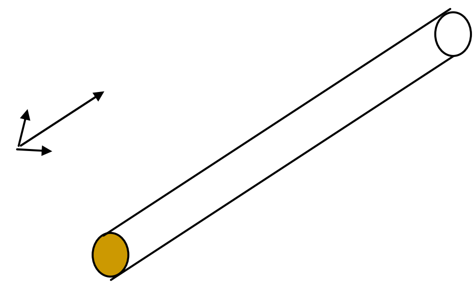


From atoms to "more atoms" and back

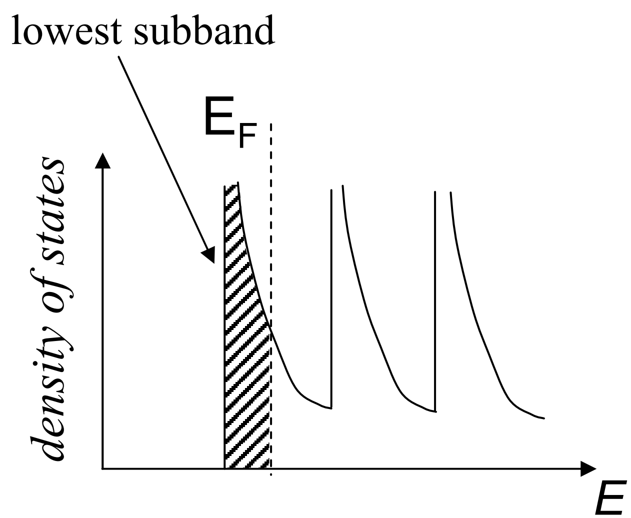


Confined in 2 directions: 1D system

Condition for confinement is that the confinement length of the order of the Fermi wavelength **($L \sim \lambda_F$ or $E_1 <\sim E_F$) .** Then electrons confined in one quantum mechanical state in two directions, but free to move in the third => 1D.



Semiconductors are good for that (hard to see confinement effect in metals)

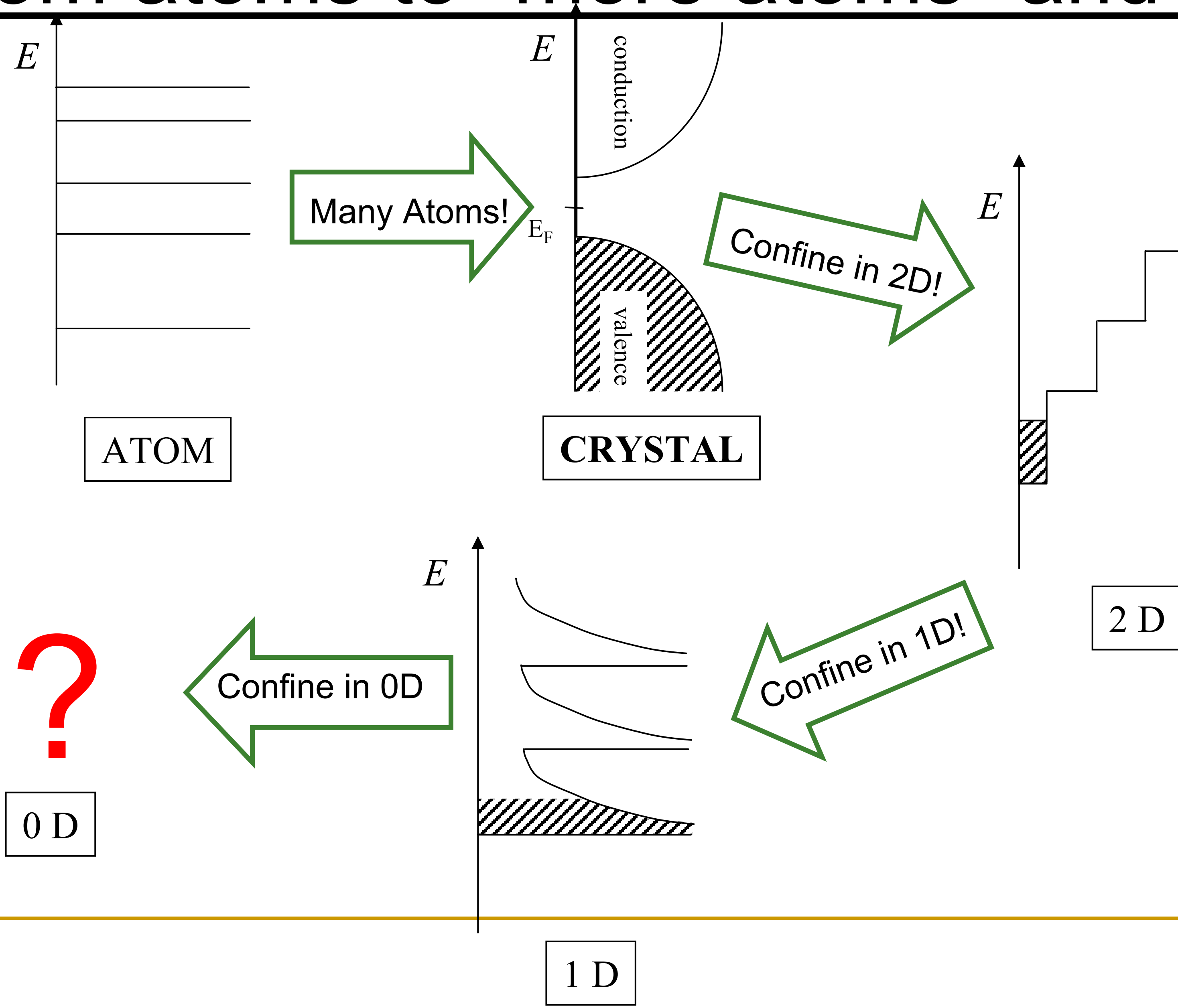


Interestingly, electrons interact differently in 1D compared to 2D and 3D. As an analogy think of cars (electrons) moving along a single track lane. They interact differently compared to cars on dual carriageways or motorways.

Examples include carbon nanotubes, nanowires, lithographically defined regions of 2DEGs etc.

$$\rho_{1D}(E) = \left(\frac{2m}{\pi^2\hbar^2}\right)^{1/2} \sum_i \frac{n_i \Theta(E - E_i)}{(E - E_i)^{1/2}}$$

From atoms to “more atoms” and back



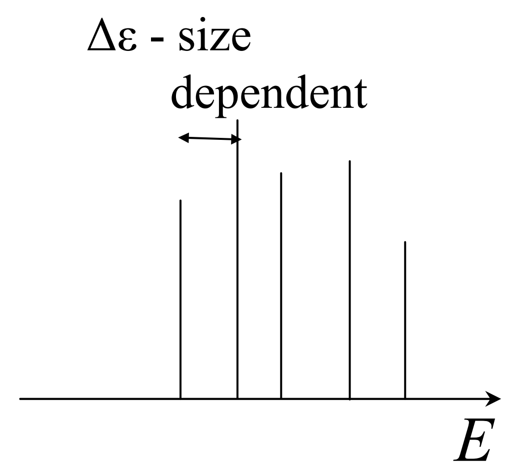
Confined in *all* directions: 0D systems or Quantum Dots

Electron systems confined in all three directions. ‘0D’

man-made droplets of charge

e.g. nanocrystals (nanoparticles)
molecules

degree of confinement does not have to be the same in all directions
=> 2 D quantum dots and 1D quantum dots



fabrication either “bottom up”:

2DEG
small metal islands

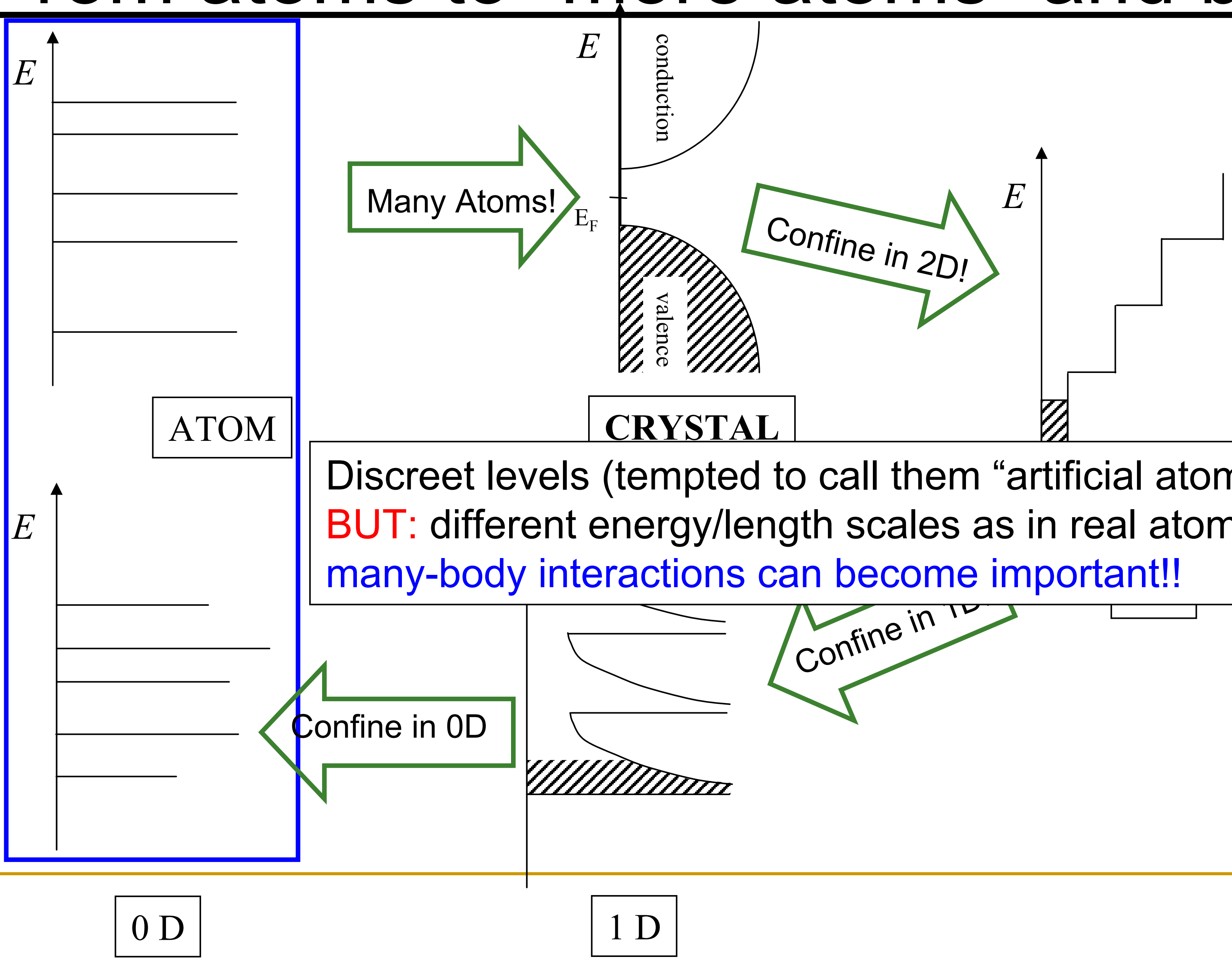
or “top down” fabrication:

e.g. nanotubes
nanowires

or combination of both.

$$\boxed{\rho_{0D}(E) = \sum_i \delta(E - E_i)}$$

From atoms to “more atoms” and back

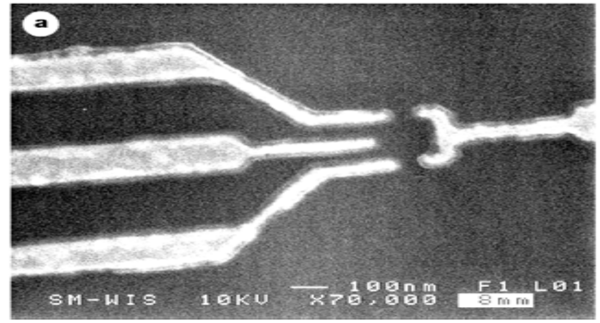


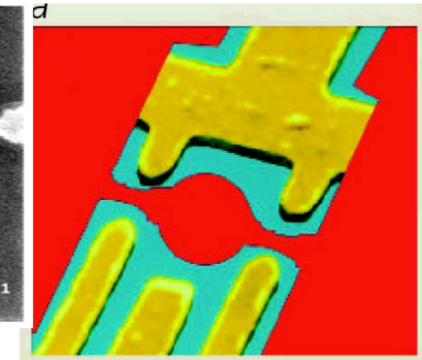
Discreet levels (tempted to call them “artificial atoms”?)
BUT: different energy/length scales as in real atoms.
many-body interactions can become important!!

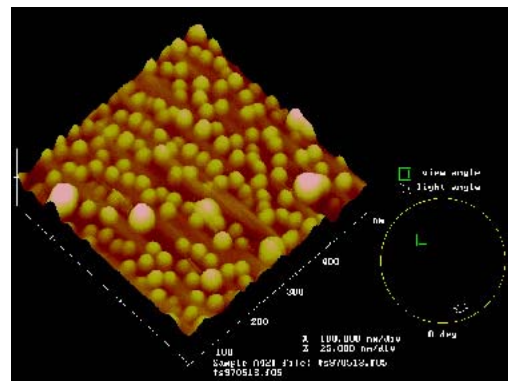
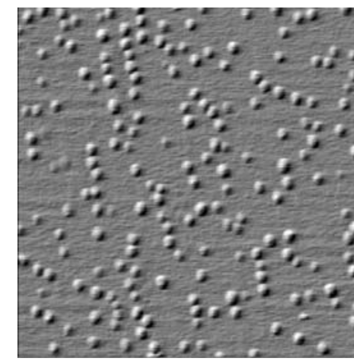
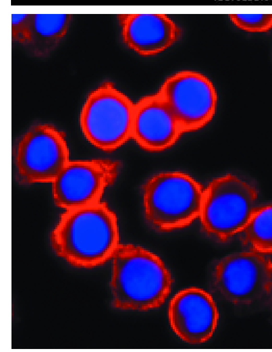
What are *Quantum Dots*?

Semiconductor Quantum Dots:

- Devices in which electrons are **confined** in nanometer size volumes.
- Sometimes referred to as "artificial atoms".
- "Quantum dot" is a generic label: lithographic QDs, self-assembled QDs, colloidal QDs have different properties.

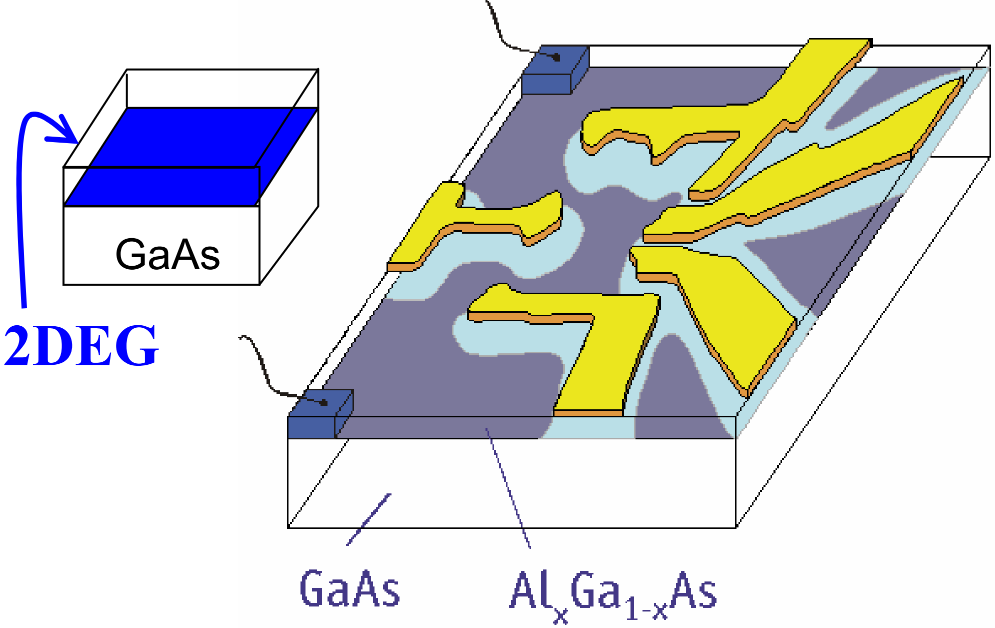


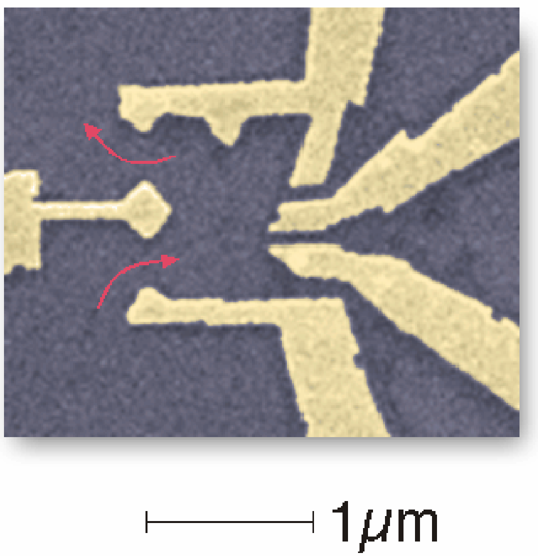



Lithographic Quantum Dots

How to do it in practice? (a question for the experimentalists…)



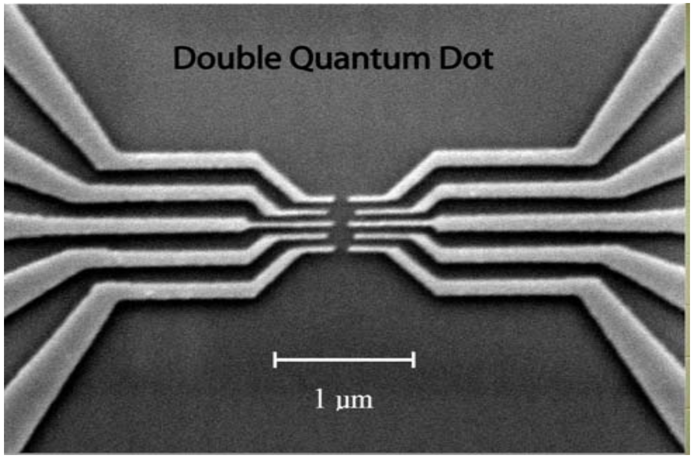


from Charlie Marcus' Lab website (marcuslab.harvard.edu)

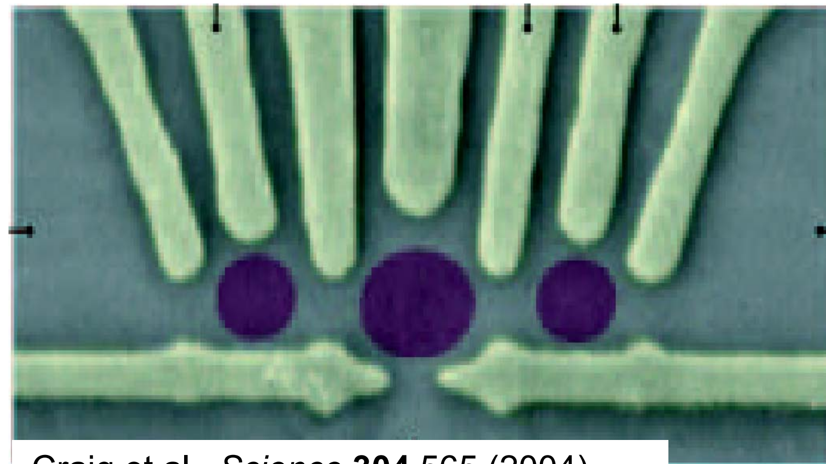
Ingredients:

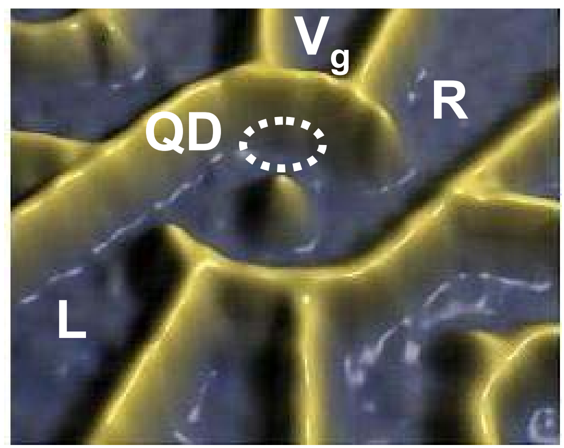
- Growth of heterostructures to obtain the 2DEG
  - (good quality, large mean free-paths)
- Metallic electrodes electrostatically deplete charge: confinement
- Sets of electrodes to apply bias etc.
- LOW TEMPERATURE! (~100 mK)

Lithographic Quantum Dots



Jeong, Chang, Melloch *Science* **293** 2222 (2001)


Craig et al., *Science* **304** 565 (2004)



From:
K. Ensslin's group website

Lithography evolved quite a bit in the last decade or so. Allow different patterns: double dots, rings, etc.

Quantum Dots: transport

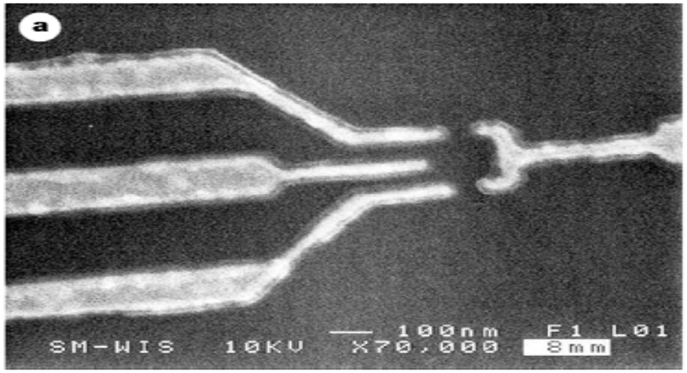
Lithographic Quantum Dots:

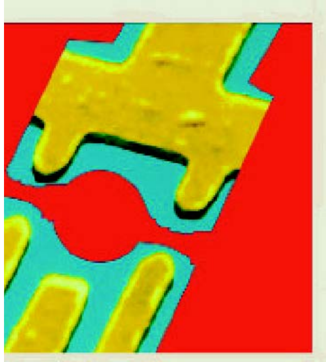
- Behave like small capacitors:

$$\boxed{E_c = \frac{e^2}{C}}$$

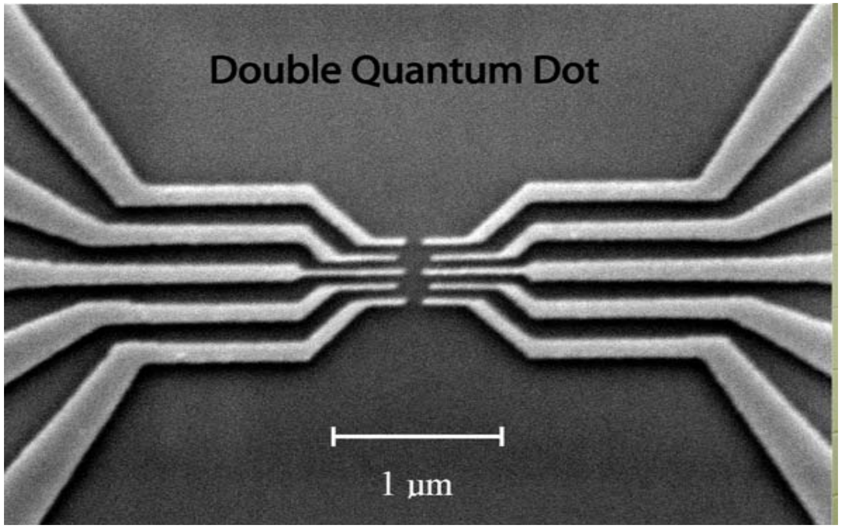
- Weakly connected to metallic leads.
- Energy scales: level spacing ΔE; level-broadening Γ.
- E_C is usually largest energy scale:

$$\boxed{E_c \gg \Delta E, \Gamma}$$



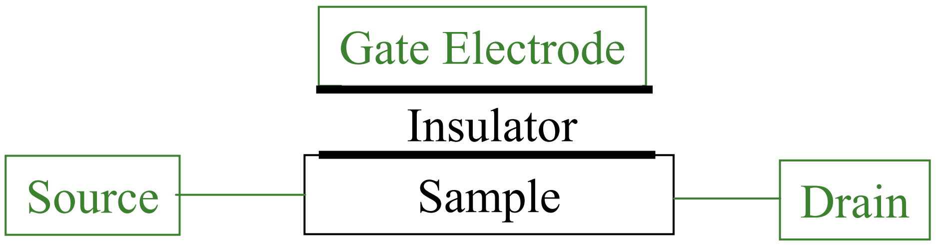


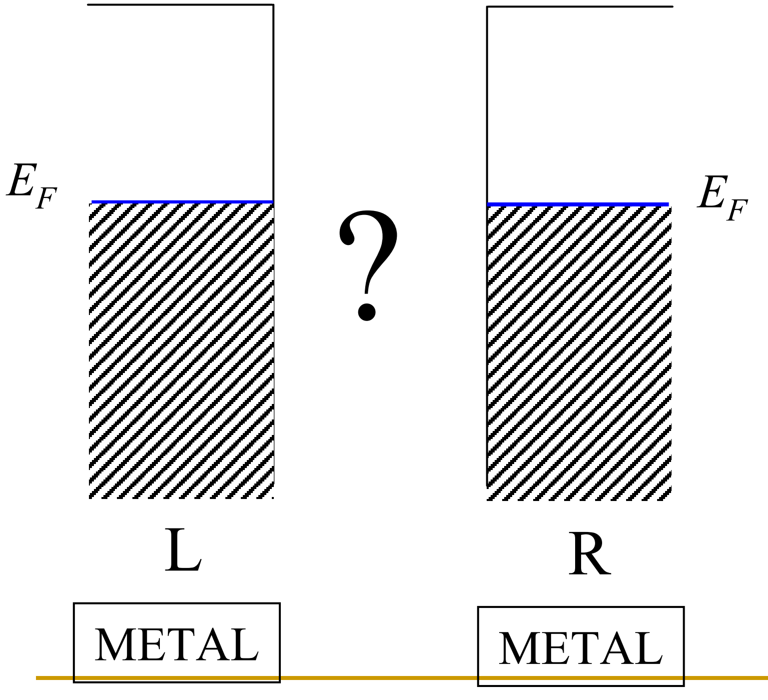
Goldhaber-Gordon *et al.* *Nature* **391** 156 (1998)

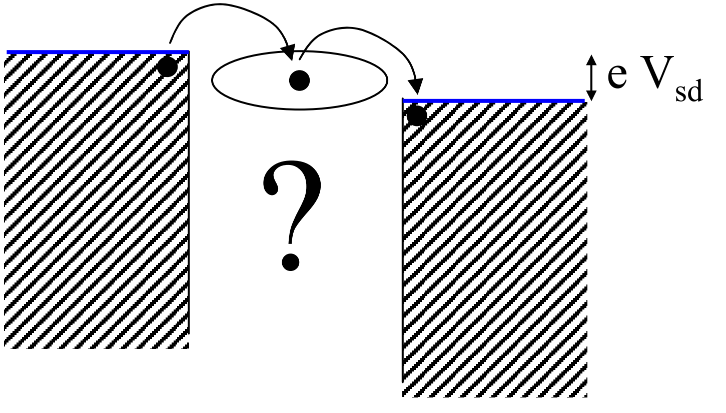


Jeong, Chang, Melloch *Science* **293** 2222 (2001)

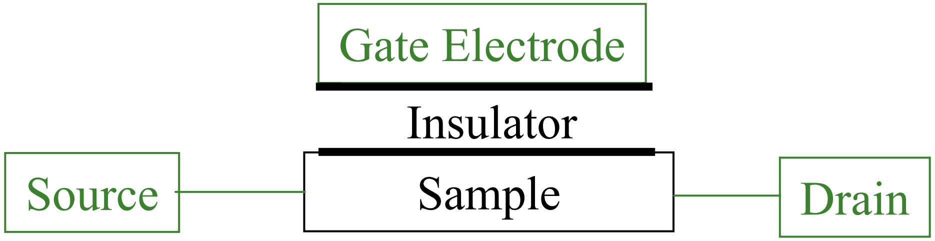
Electrical Transport

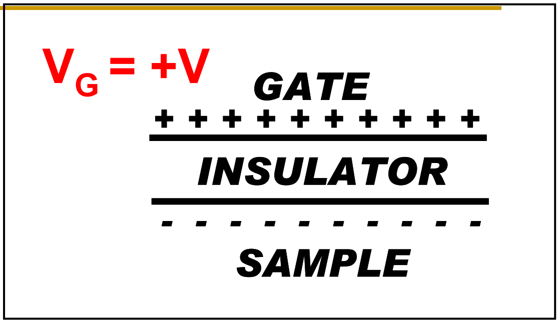


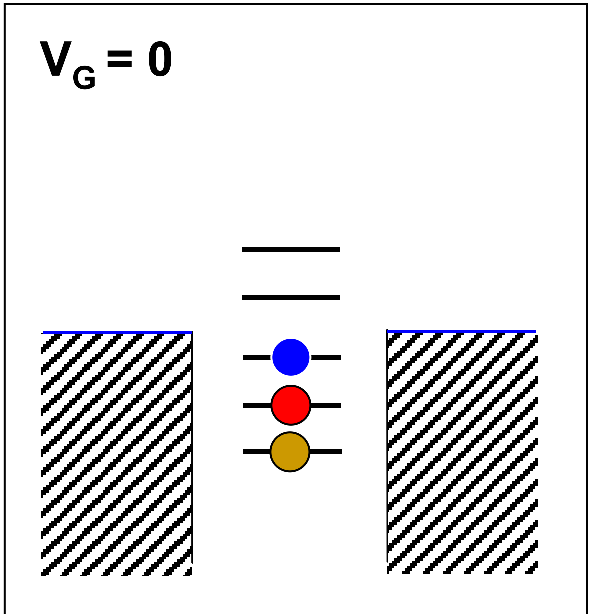


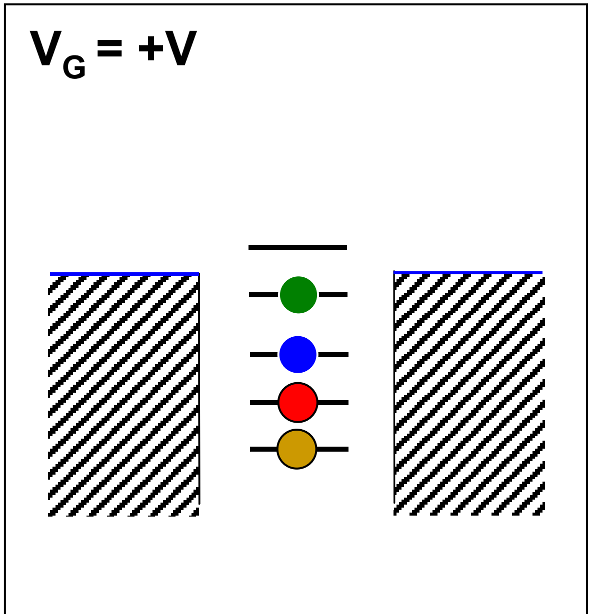


Role of the Gate Electrode

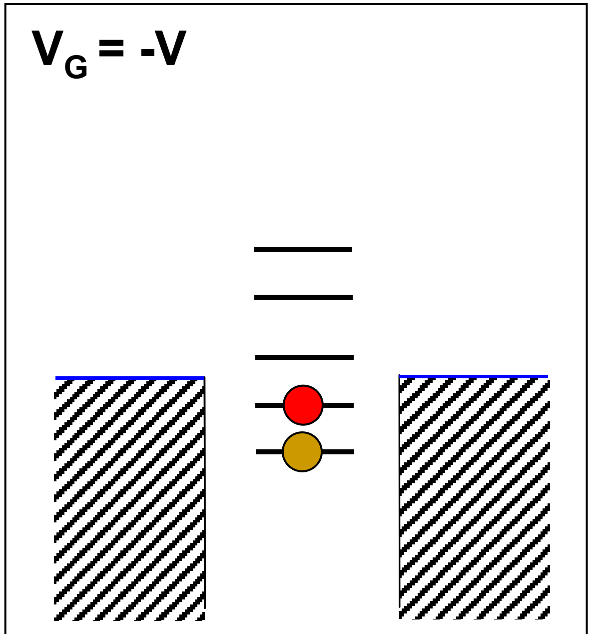








Raise Fermi level – adds electrons

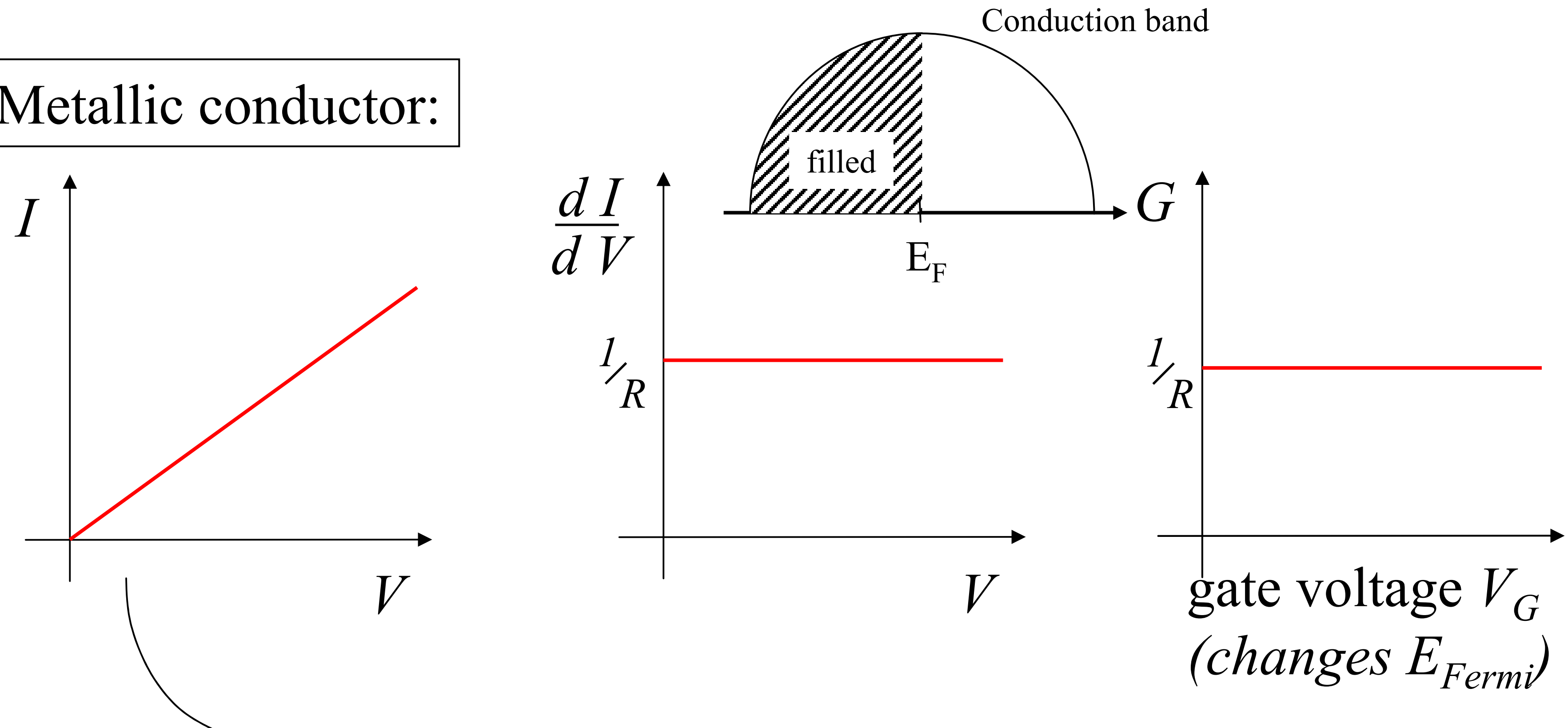


Lower Fermi level – remove electrons

Electrical Transport: Ohm's Law

Ohm's Law holds for metallic conductors => $V = I R$
We can also define a conductance which can be bias dependent $\frac{dI}{dV}$
The **zero bias conductance**, *G*, is conventionally quoted.

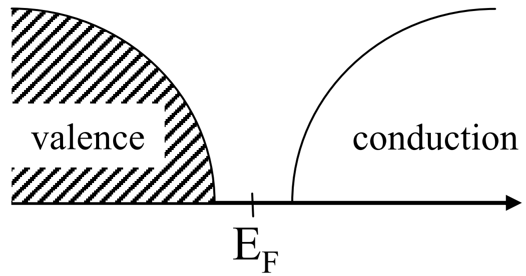
Metallic conductor:

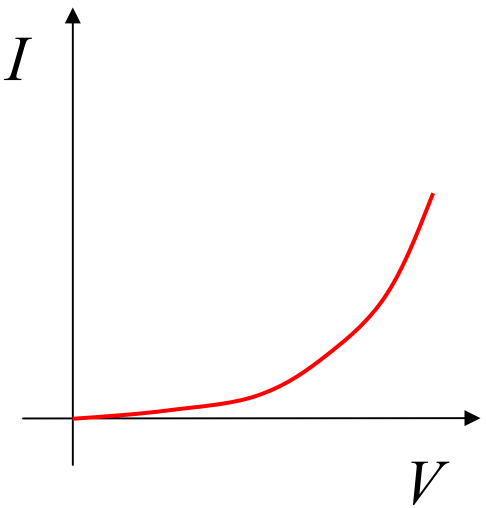


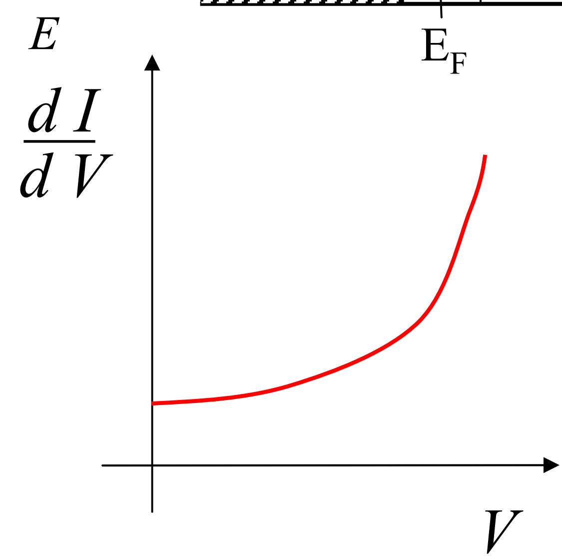
Resistance due to scattering off impurities, mfp ~ 10 nm

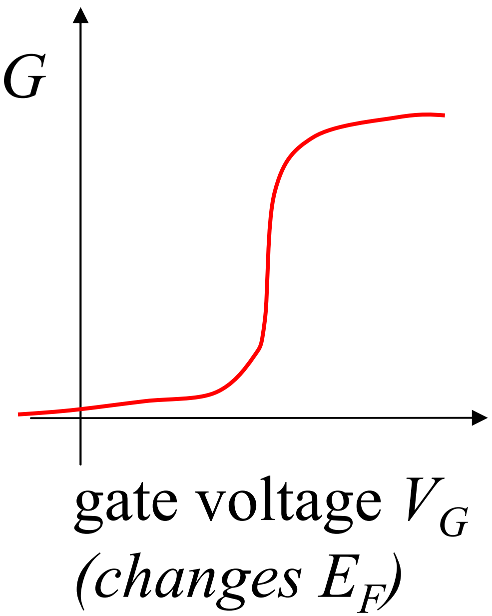
Electrical Transport: semiconductors

Semiconductor:









Semiconductor - nonlinear I - V response

tunneling through Schottky barrier
or out of band gap.

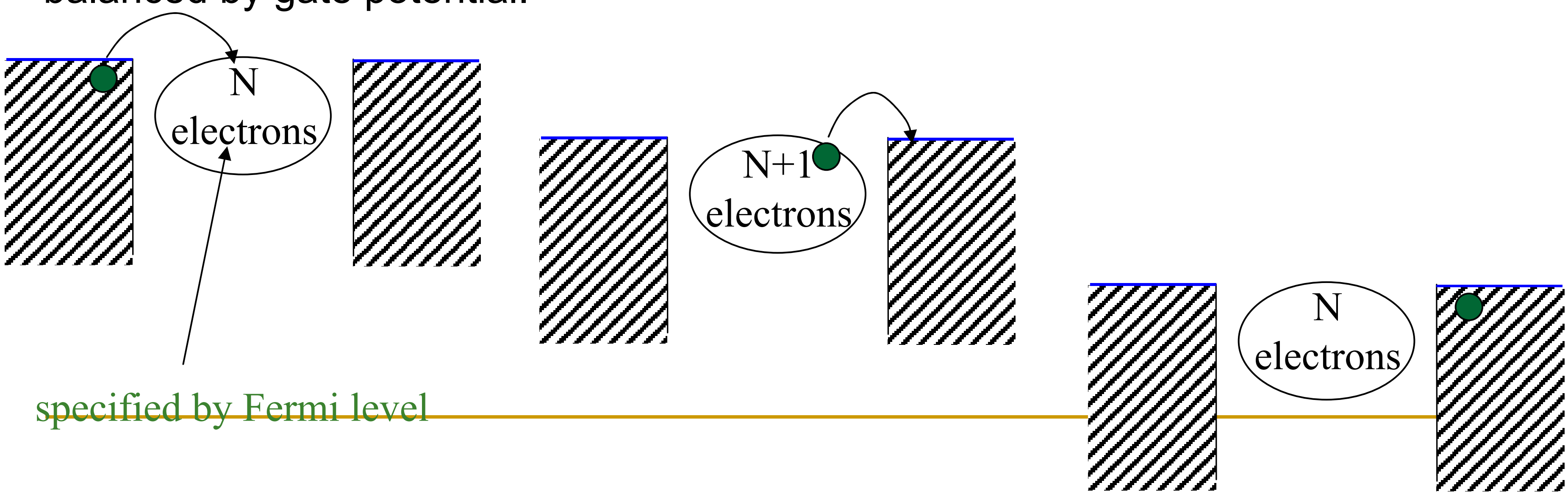
Conductance through quantum dot

Quantum dots contain an integer number of electrons.

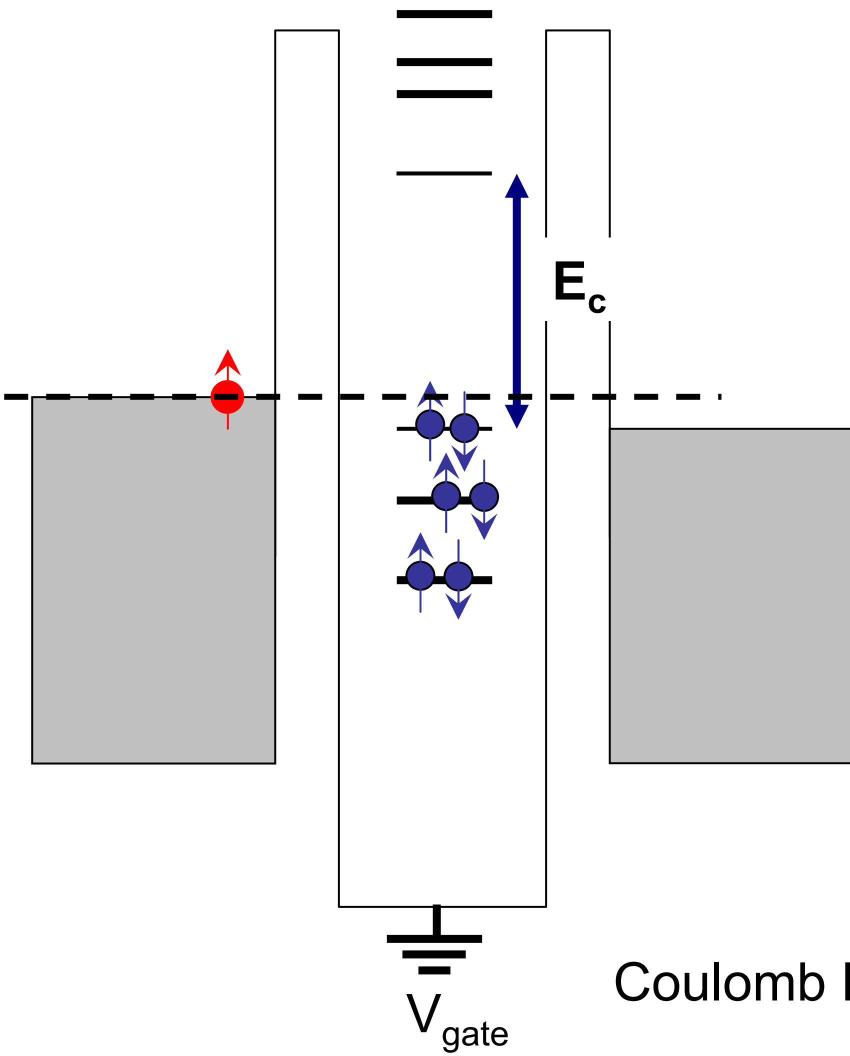
Adding an electron to the QD changes its energy => electrostatic charging energy $\frac{Q^2}{2C}$

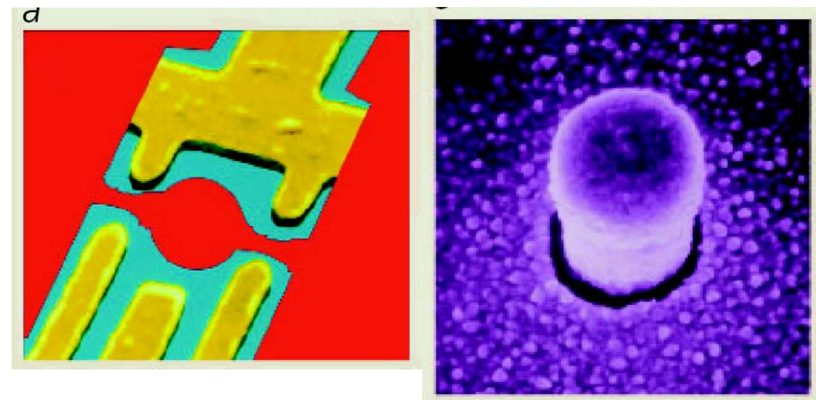
In order for a current to pass an electron must tunnel onto the dot, and an electron must tunnel off the dot.

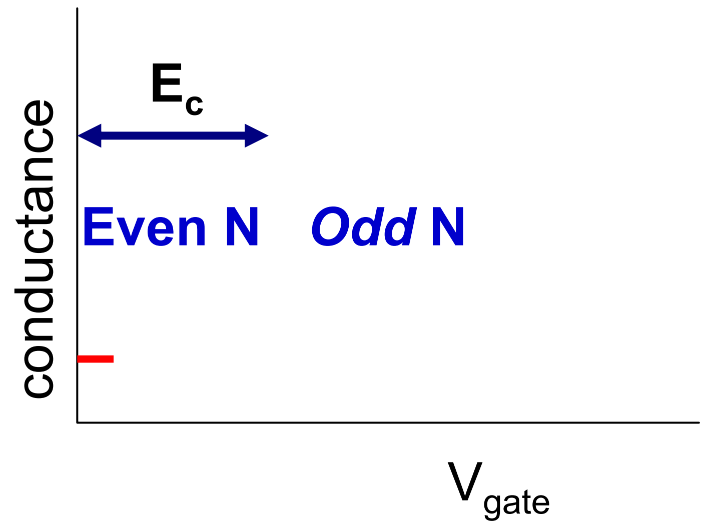
For **conduction at zero bias** this requires the energy of the dot with *N* electrons must equal the energy with *N+1* electrons. i.e. charging energy balanced by gate potential.



Coulomb Blockade in Quantum Dots

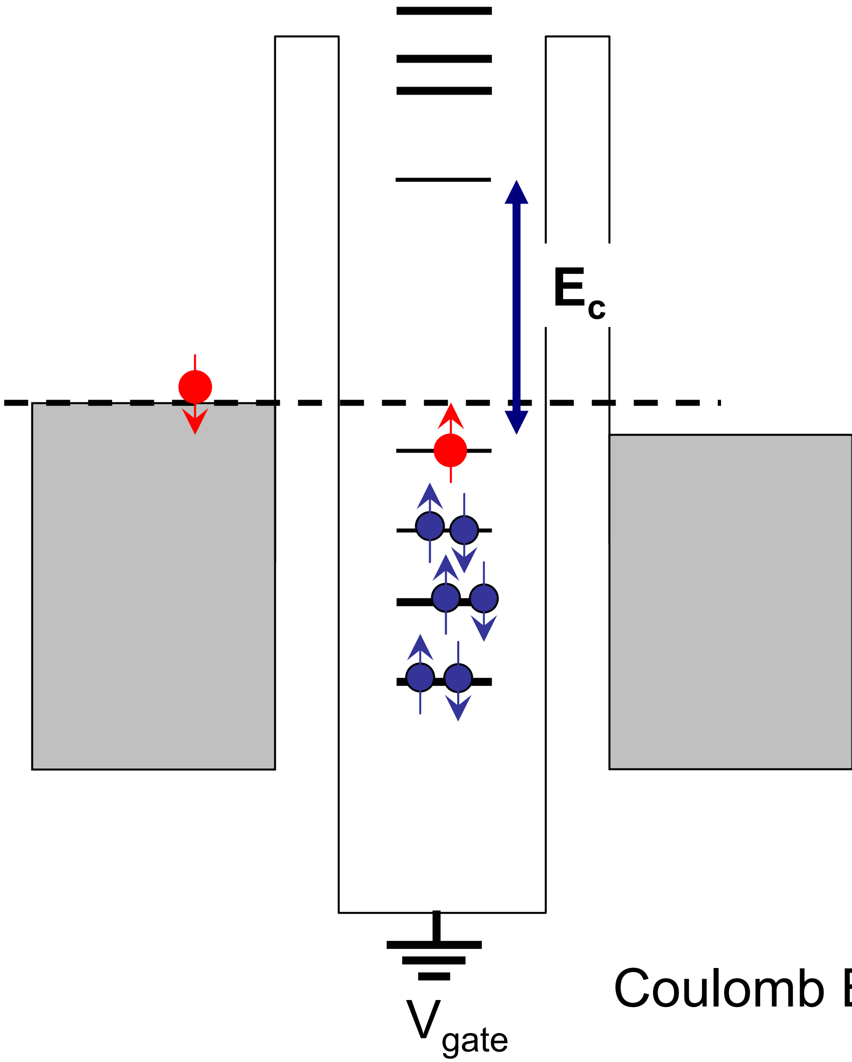


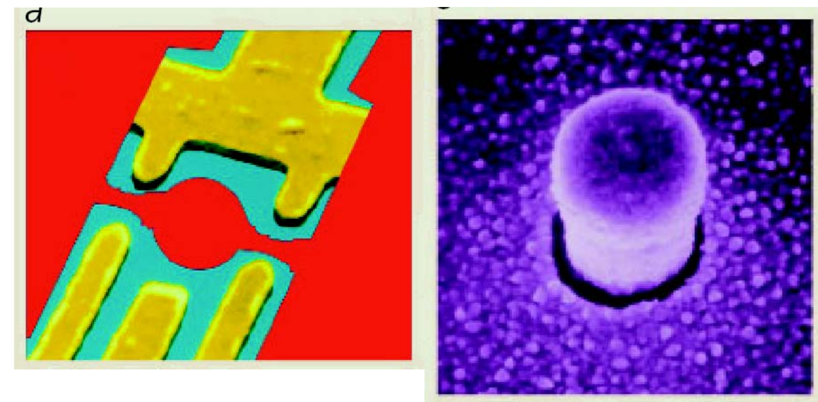


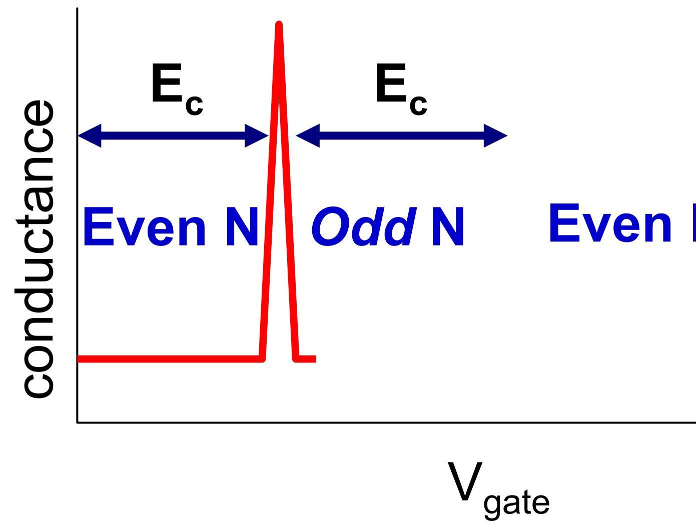


Coulomb Blockade in Quantum Dots

Coulomb Blockade in Quantum Dots

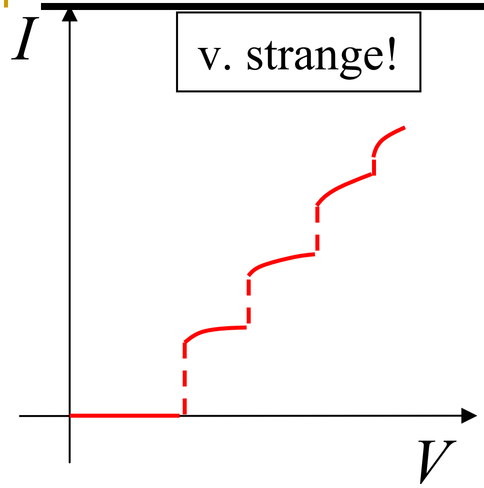


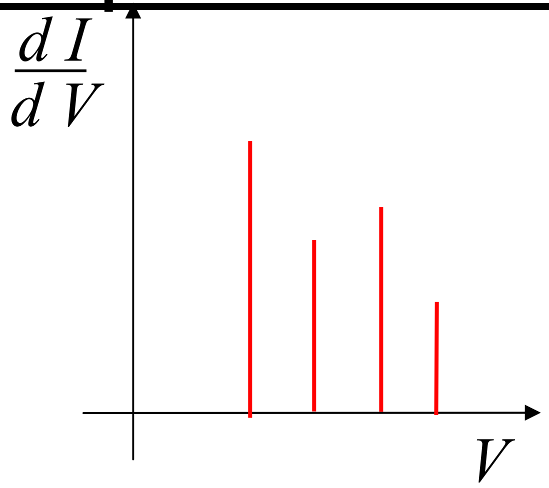


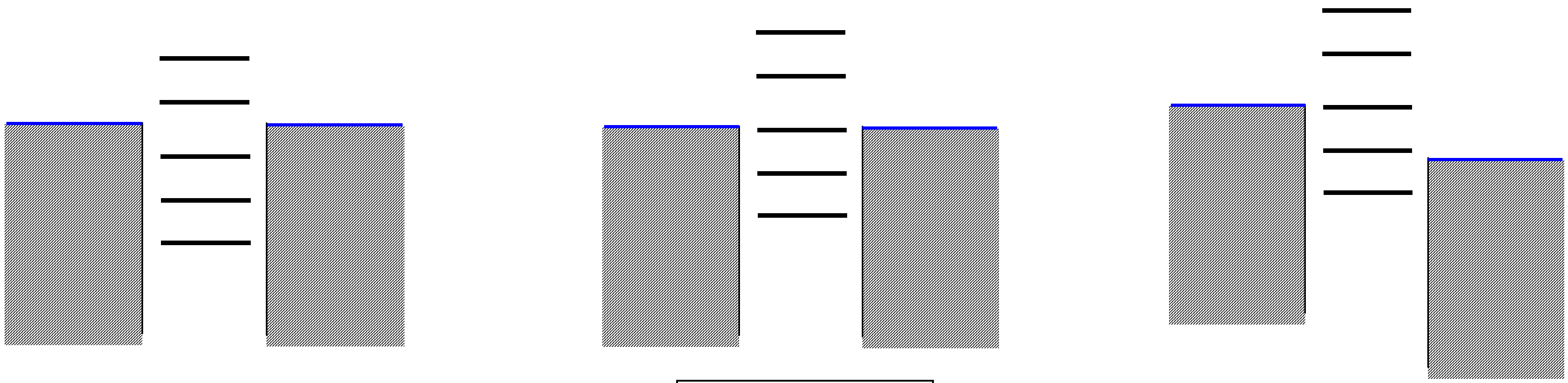


Coulomb Blockade in Quantum Dots

Electrical Transport: Coulomb staircase







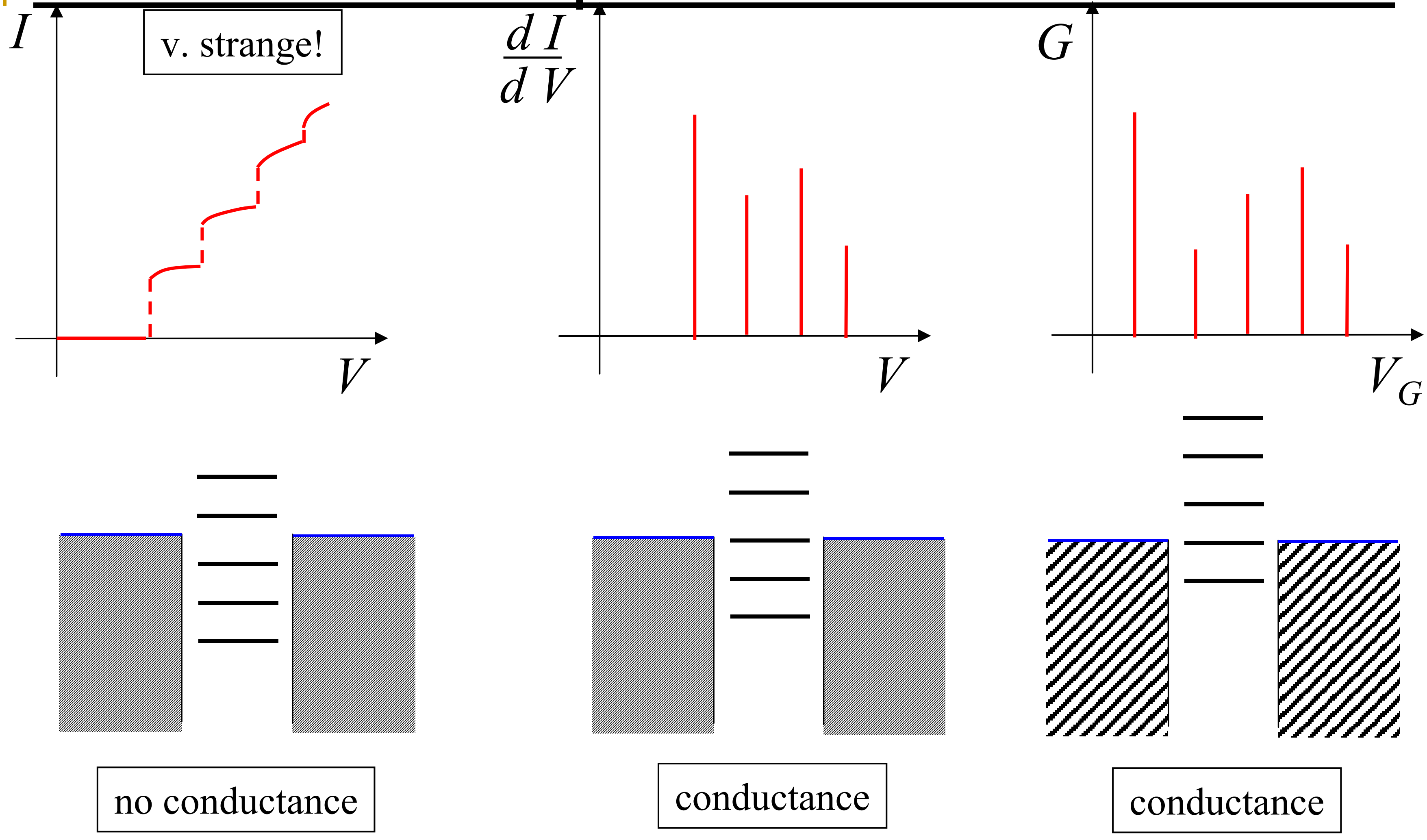
no conductance

conductance

increased conductance

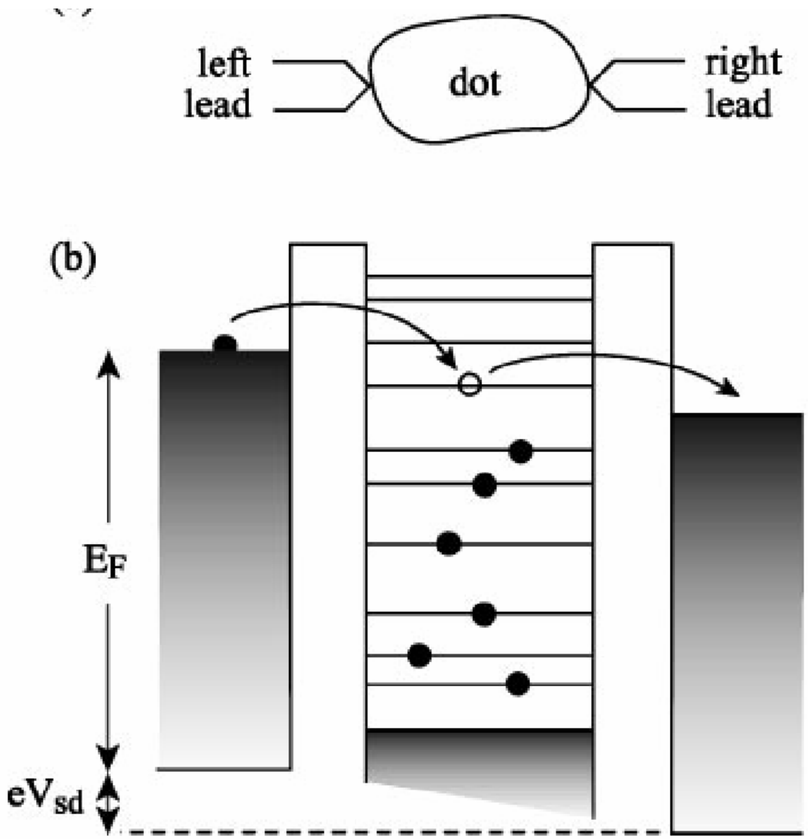
Probing the energy levels in the 'artificial atom'

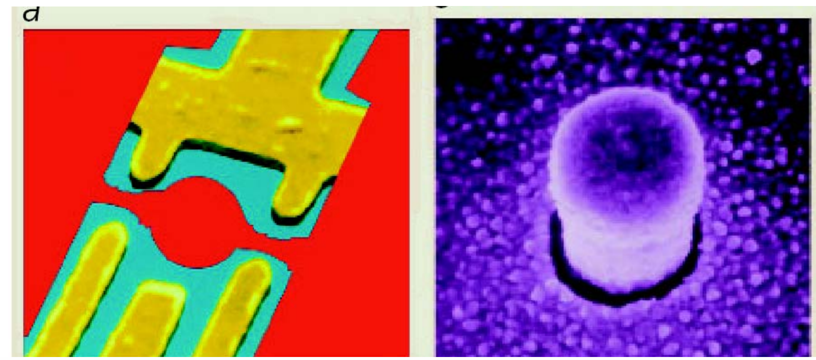
Electrical Transport: Coulomb staircase

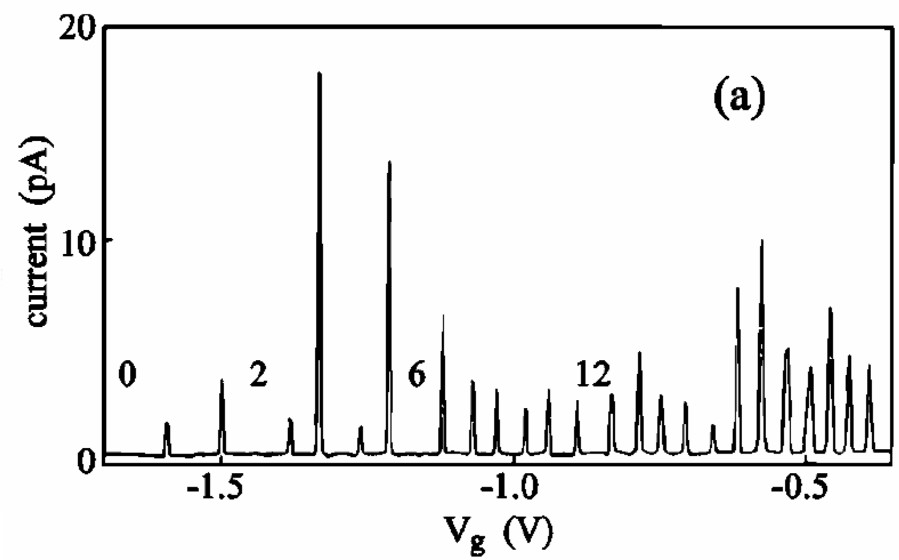


Probing the energy levels in the 'artificial atom'

Coulomb Blockade in Quantum Dots



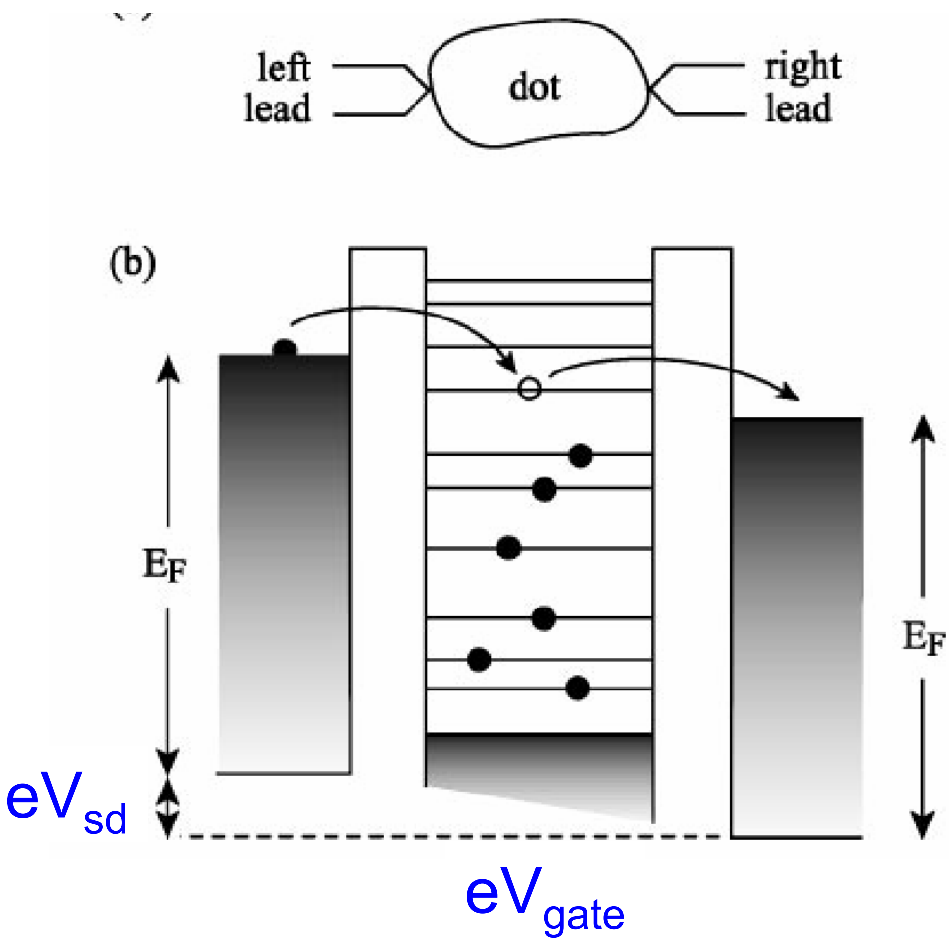




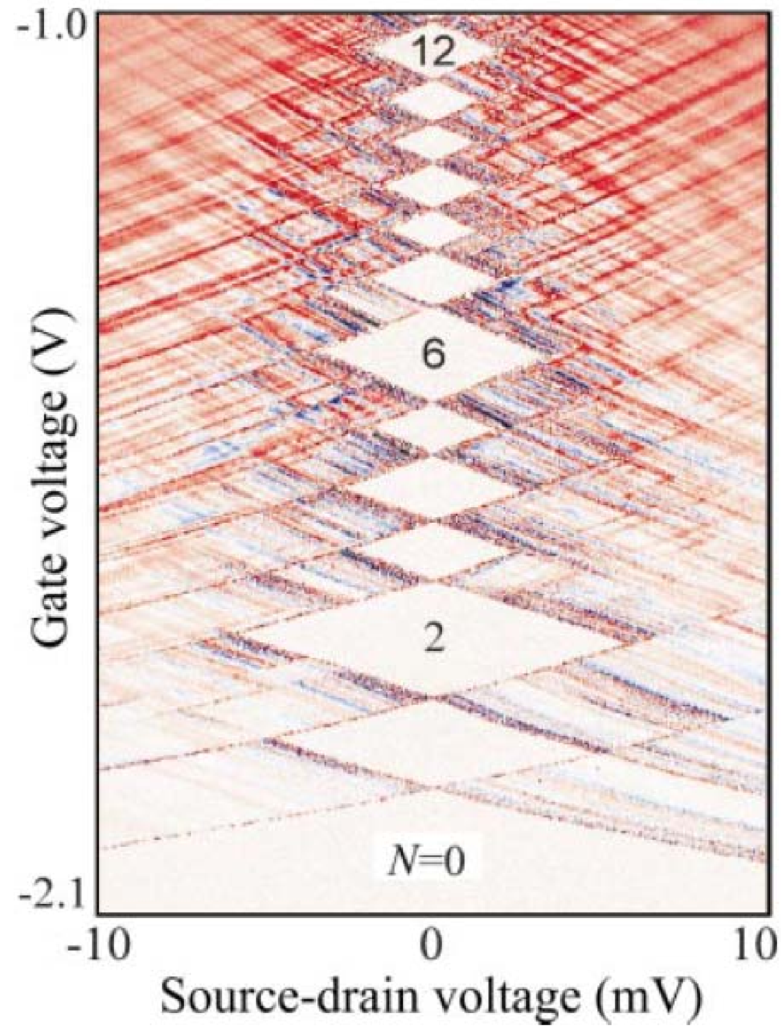
Coulomb Blockade in Quantum Dots: “dot spectroscopy”

Y. Alhassid *Rev. Mod. Phys.* **72** 895 (2000).

“Coulomb Diamonds” (Stability Diagram)

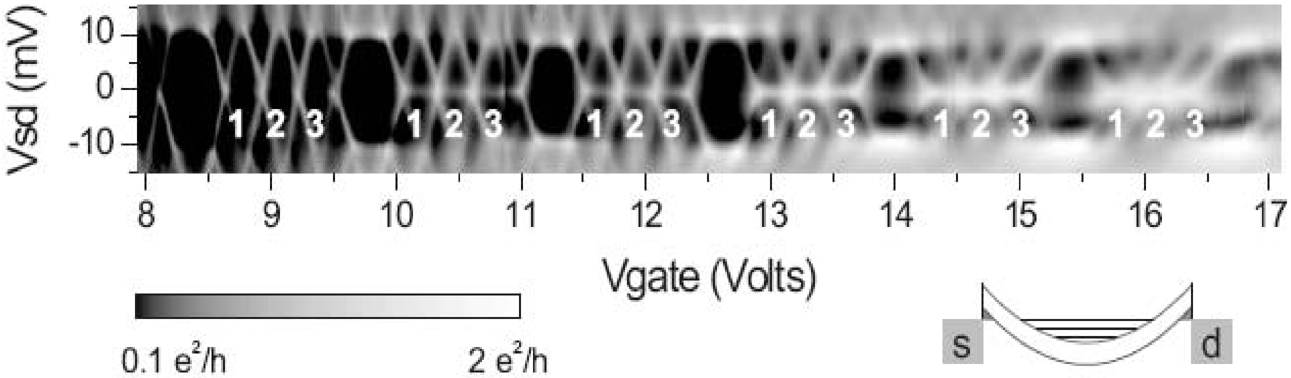


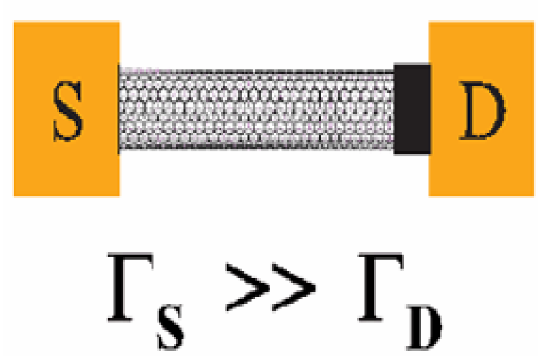
Coulomb Blockade in Quantum Dots



L. P. Kouwenhoven et al. *Science* **278** 1788 (1996).

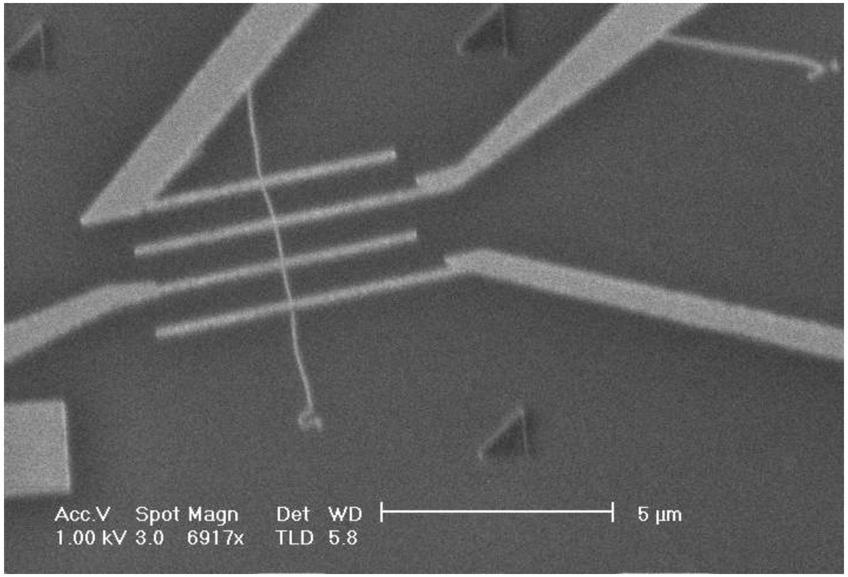
"Carbon nanotube Quantum dots".





Makarovski, Zhukov, Liu, Filkenstein *PRB* **75** 241407R (2007).

- Carbon nanotubes depsited on top of mettalic electrodes.
- Quantum dots defined *within* the carbon nanotubes.
- More structure than in quantum dots: "shell structure" due to *orbital* degeneracy.



Gleb Filkenstein's webpage: http://www.phy.duke.edu/~gleb/

Charging Energy Model

Energy of N particles on QD can be split into the energy levels and the charging energy

$$E_N = \varepsilon_1 + \varepsilon_2 + \cdots + \varepsilon_N + \frac{(Ne)^2}{2C}$$

An extra contribution is given by the gate electrode,

$$\alpha V_G N$$

α is the capacitive coupling of the dot to the gate.

The number of electrons on the QD is determined by the Fermi energy, $\rightarrow$ if

$$E_N + \alpha V_G N < E_F < E_{N+1} + \alpha V_G (N+1)$$

then there will be N electrons on the QD.

The no. of electrons on the QD is adjusted by the gate potential.

Conductance peak spacing

The energy of $N+1$ electrons is

$$E_{N+1} = \varepsilon_1 + \varepsilon_2 + \cdots + \varepsilon_{N+1} + \frac{(N+1)^2 e^2}{2C}$$

The difference in energy between N and $N+1$ electrons is

$$\Delta E_{N\to N+1} = E_{N+1} - E_N = \varepsilon_{N+1} + (2N+1)\frac{e^2}{2C}$$

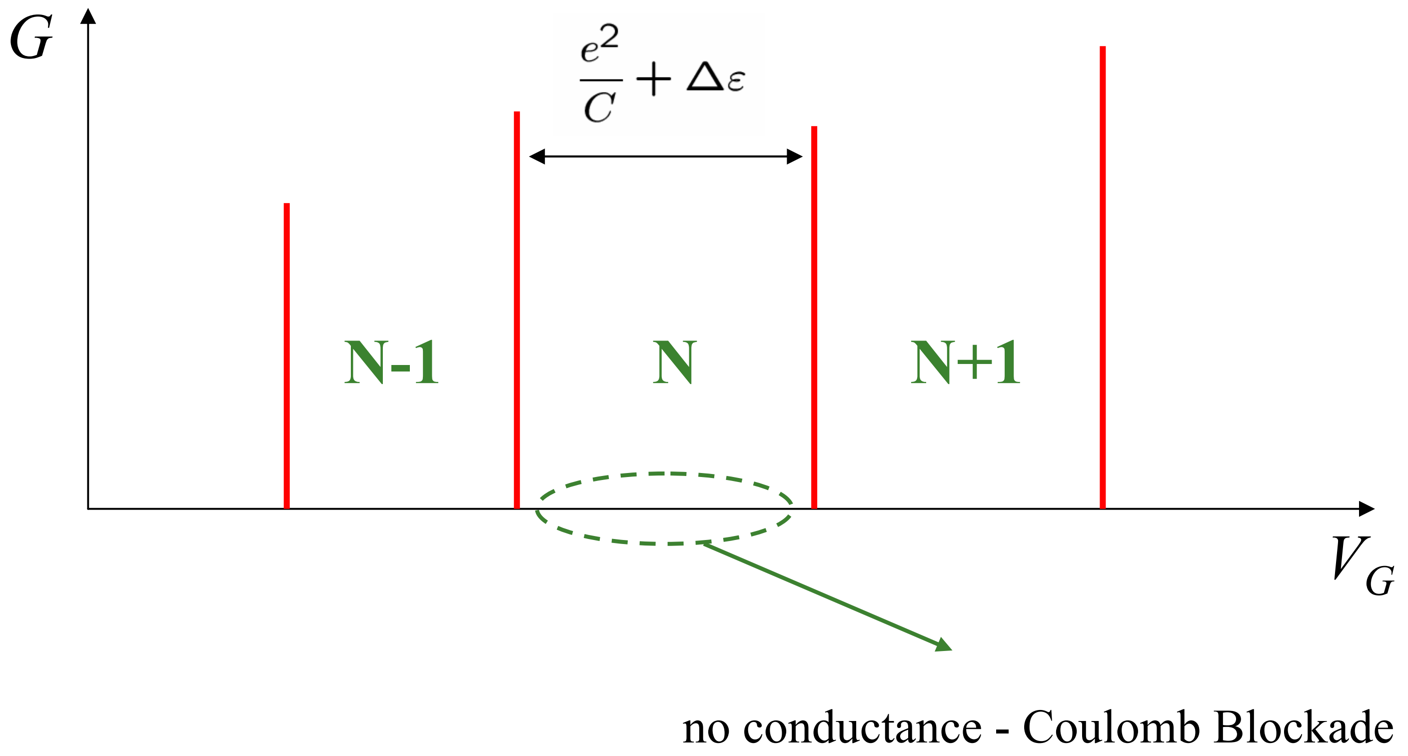
The separation between conductance peaks in this model is given by

$$\alpha \Delta V_G = \Delta E_{N\to N+1} - \Delta E_{N-1\to N} = \varepsilon_{N+1} - \varepsilon_N + \frac{e^2}{C}$$

level spacing
Δε

charging energy

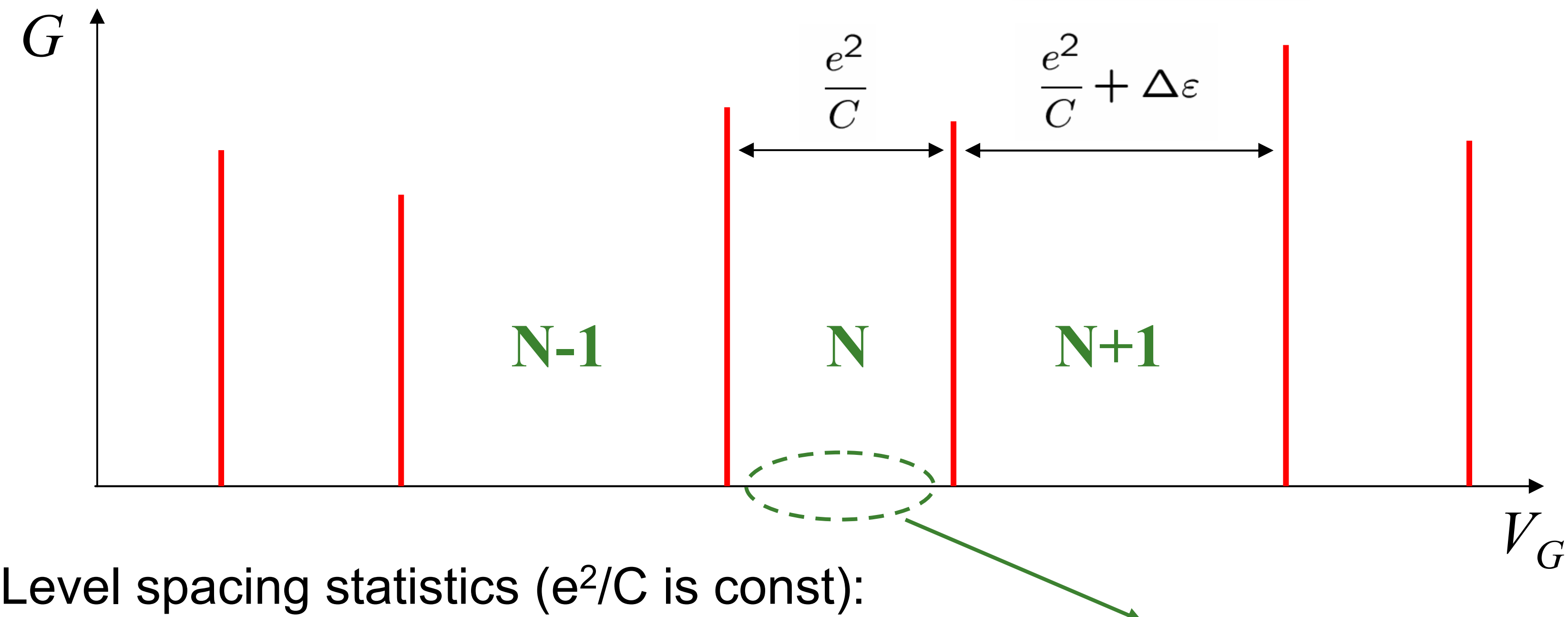
Conductance peak spacing II



Conductance peak spacing III: spin degeneracy

In the event of degeneragy, e.g. spin degeneracy

$$\varepsilon_N = \varepsilon_{N+1}$$

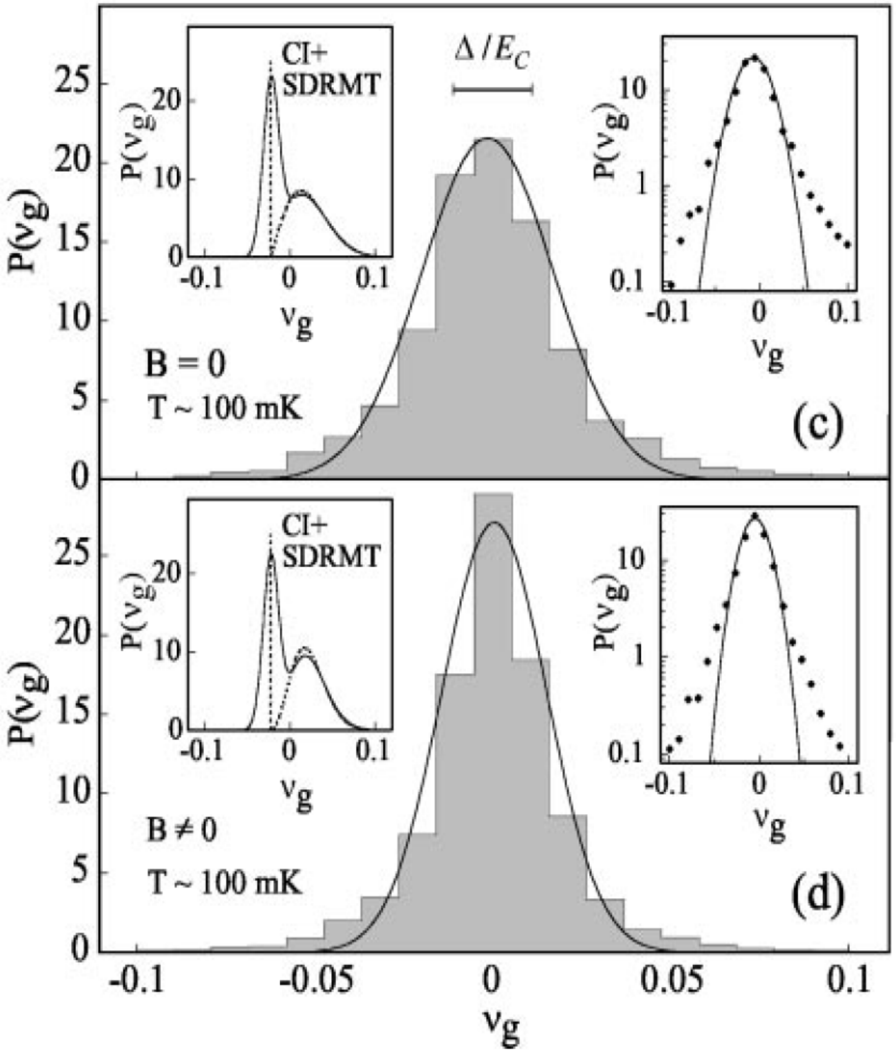


Level spacing statistics (e^2/C is const):

$$\boxed{\begin{array}{l} P(E_{N+1} - E_N) = P(\Delta E) \text{ for even } N \\ P(E_{N+1} - E_N) = \delta(E) \text{ for odd } N \end{array}}$$

no conductance - Coulomb Blockade

Peak spacing statistics: e-e interactions



Sivan et al. *PRL* **77** 1123 (1996).

$$P(E_{N+1} - E_N) = P(\Delta E) \text{ for even } N$$
$$P(E_{N+1} - E_N) = \delta(E) \text{ for odd } N$$

Level spacing distribution P(ΔE):
Theory: CI+RMT prediction
Superposition of two distributions:
• even N: large, chaotic dots:
P(ΔE) obeys a well known
RMT distribution (*not* Gaussian)
• odd N: Dirac delta function.

Experiment:
P(ΔE) gives a Gaussian.

Possible explanations:
Spin exchange, residual e-e interaction effects*.

* Jiang, Baranger, Yang *PRL* **90** 026806 (2003).

Applications of a ‘SET’

• These devices are often call **single electron transistors** - conductance modulated by gate voltage between on and off states and the mechanism is single electron transport.

•Most obvious proposed application is for replacing conventional FET.

•By making stable energy states can have defined spin on the dot => spintronics

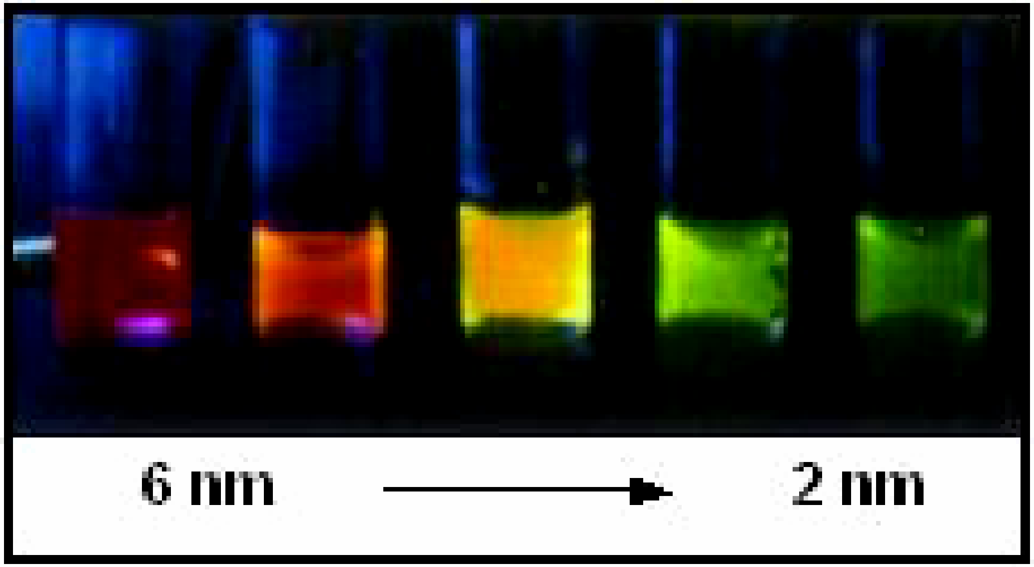
•Quantum computing through ‘mixing’ spins etc.

•Fundamental science

Colloidal quantum dots (nanocrystals)

Same electronic quantisation => enhanced **optical properties**
greater specificity of colour
greater intensity of emission

In addition reducing size increases band gap => tunable color



(d) Colloidal CdSe nanocrystals dissolved in toluene. Each vial contains CdSe nanocrystals of a different size, ranging from about 2 to 6 nm. All solutions were excited with a hand-held UV lamp and a photograph of the fluorescence was recorded. The small (2 nm) nanocrystals emit green, and the large (6 nm) ones emit red light.

Nanotechnology 14 (2003) R15–R27

Already in the market!
•$750 (10 mg kit, 6 colors) at www.nn-labs.com
Applications:
• Most used: biological “tagging”
• Other ideas: prevent counterfeiting money, IR emmitters (“quantum dust”)

Applications

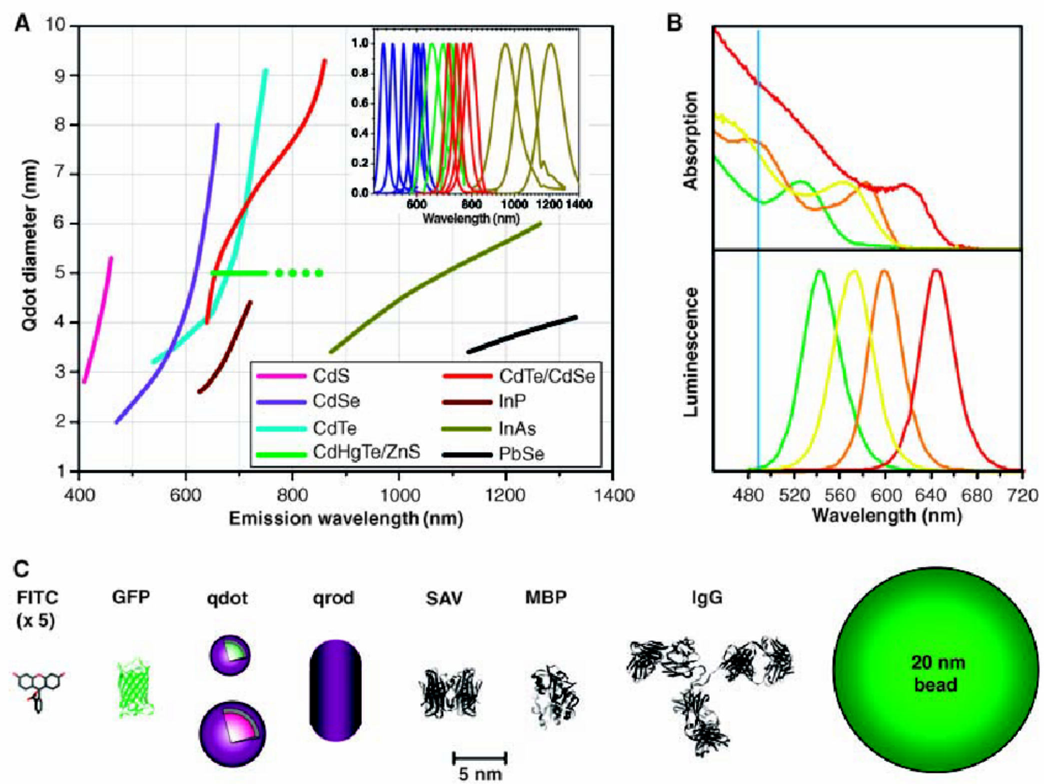


Fig. 1. (**A**) Emission maxima and sizes of quantum dots of different composition. Quantum dots can be synthesized from various types of semiconductor materials (II-VI: CdS, CdSe, CdTe…; III-V: InP, InAs…; IV-VI: PbSe…) characterized by different bulk band gap energies. The curves represent experimental data from the literature on the dependence of peak emission wavelength on qdot diameter. The range of emission wavelength is 400 to 1350 nm, with size varying from 2 to 9.5 nm (organic passivation/solubilization layer not included). All spectra are typically around 30 to 50 nm (full width at half maximum). Inset: Representative emission spectra for some materials. Data are from (*12*, *18*, *27*, *76–82*). Data for CdHgTe/ZnS have been extrapolated to the maximum emission wavelength obtained in our group. (**B**) Absorption (upper curves) and emission (lower curves) spectra of four CdSe/ZnS qdot samples. The blue vertical line indicates the 488-nm line of an argon-ion laser, which can be used to efficiently excite all four types of qdots simultaneously. [Adapted from (*28*)] (**C**) Size comparison of

Quantum Dots for live cells, in vivo imaging, and diagnostics'
X. Michalet et al.; Science, (2005), **307**, p.538-544