

# Kondo effect in mesoscopic systems (experimental)

(<http://dasan.sejong.ac.kr/~eom/>)

# Kondo effect in mesoscopic systems (experimental)

## I. Kondo effect in mesoscopic metallic alloys

- (1) Finite size effect in Kondo alloys
- (2) Thermopower of mesoscopic spin glasses
- (3) Superconducting proximity effect in Kondo alloys

## II. Kondo effect in low-dimensional systems

- (1) Kondo effect in a single electron transistor
- (2) Kondo effect in carbon nanotubes
- (3) Kondo effect in a single molecule transistor

# Kondo model

Dilute Magnetic Alloy

Single localized spin impurity embedded in s-like electron sea  
; AuFe, AuMn, CuFe, CuMn, CuCr .....

$$H = \sum_{k,\sigma} \epsilon_k c_{k\sigma}^+ c_{k\sigma} + \epsilon_L (n_{L\uparrow} + n_{L\downarrow}) + H_{sd}$$

Conduction electrons

Localized d-electron

s-d exchange interaction

; formulated by Zener(1951), and Kondo showed(1964) the spin-flip scattering could cause unusual low-temperature behavior by going beyond first Born approximation.

$$H_{sd} = -J_{s-d} \vec{S} \cdot \vec{s}$$

impurity spin

conduction spin

$$= -\frac{1}{2N} \sum_{k,q} J_{kq} [S_L^{(z)} (c_{k+q\uparrow}^+ c_{k\uparrow} - c_{k+q\downarrow}^+ c_{k\downarrow}) + S_L^{(+)} c_{k+q\downarrow}^+ c_{k\uparrow} + S_L^{(-)} c_{k+q\uparrow}^+ c_{k\downarrow}]$$

Non spin-flip process :  $H_{sd}^z$

spin flip process :  $H_{sd}^\pm$

# LogT dependence in R(T)

Scattering amplitude for a channel of  $k \uparrow \Rightarrow k' \uparrow$

$$t_{k\uparrow,k'\uparrow} = t_{k\uparrow,k'\uparrow}^{(1)} + t_{k\uparrow,k'\uparrow}^{(2)} + \dots$$

$$t_{k\uparrow,k'\uparrow}^{(1)} = -\frac{J}{2N} S_z$$

$$t_{k\uparrow,k'\uparrow}^{(2)} = t_{k\uparrow,k'\uparrow}^{(2)} \Big|_z + t_{k\uparrow,k'\uparrow}^{(2)} \Big|_{\pm} = 0 - 2 \left( \frac{J}{2N} \right)^2 S_z N(0) \ln(kT / D)$$

where  $N$  : number of d-electrons,  $N(0)$  : density of state at  $E_F$

$D$  : width of conduction electron distribution around  $E_F$

$$\begin{aligned} J_{k,q} &= J && \text{where } -D < \epsilon_k, \epsilon_q < D \\ &= 0 && \text{otherwise} \end{aligned}$$

$$R_{Kondo}(T) \propto c \frac{2\pi}{\hbar} \left| t_{k\uparrow,k'\uparrow} \right|^2 = c R_m \left[ 1 + \frac{2J}{N} N(0) \ln(kT / D) + \dots \right]$$

This  $\ln T$  dependence combined with the phonon contribution ( $T^5$  dependence) makes a resistance minimum in  $R(T)$ .

# Anderson model

$$H = \sum_{k,\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \epsilon_L (n_{L\uparrow} + n_{L\downarrow}) + \sum_{k\sigma} M_k (c_{k\sigma}^\dagger c_{L\sigma} + c_{L\sigma}^\dagger c_{k\sigma}) + U n_{L\uparrow} n_{L\downarrow}$$

Fano-Anderson model

Mixing term

Coulomb interaction

Mixing term : process where electron hops off of the impurity and becomes a continuum state and vice versa.

Coulomb interaction causes interesting magnetic phenomena.

Anderson model and Kondo model both describe the interaction of a continuum electron with localized electron. The two models are not totally different. There is a canonical transformation which when applied to the Anderson model will transform it into a form similar to the Kondo model. This transformation on the Anderson model produces quite a few terms, of which the Kondo model is a subset. Thus the transformation does not produce exactly the Kondo model. Thus the transformation does not produce exactly the Kondo model, and the two models are not identical.

- Gerald D. Mahan, Many-Particle Physics (Plenum, 1993)

# LogT dependence of Kondo resistivity

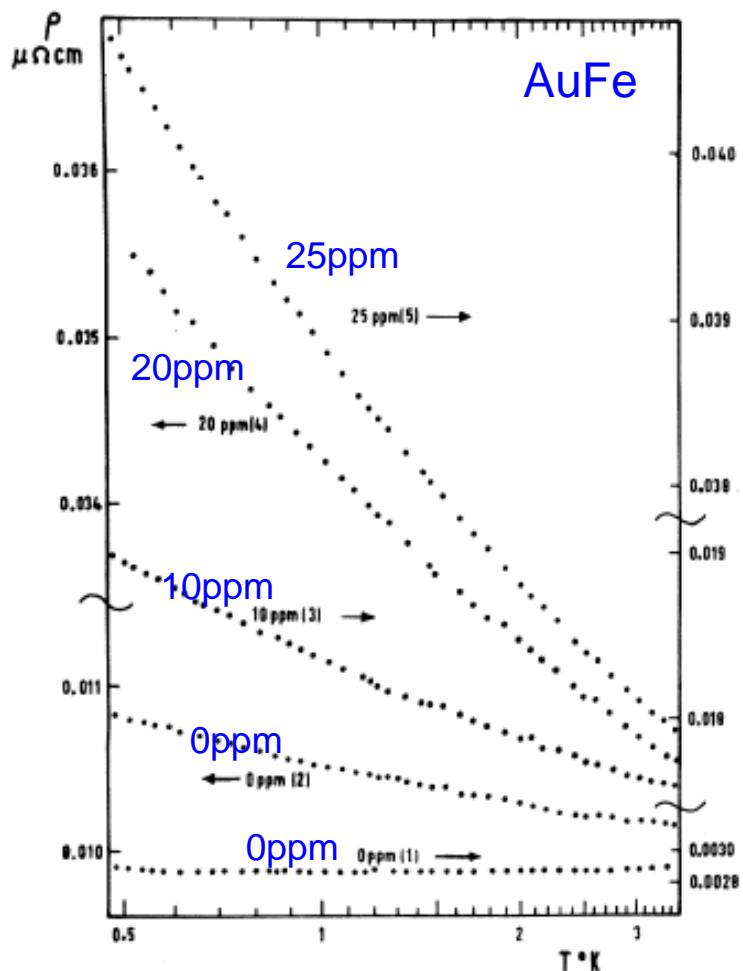


FIG. 2.  $\rho \mu\Omega\text{cm}$  against  $\log_{10} T$  for very dilute AuFe alloys. Nominal Fe concentration in each alloy is shown in ppm Fe and the specimen number in this and subsequent figures is shown in parentheses.

Loram et al., PRB 2, 857 (1970)

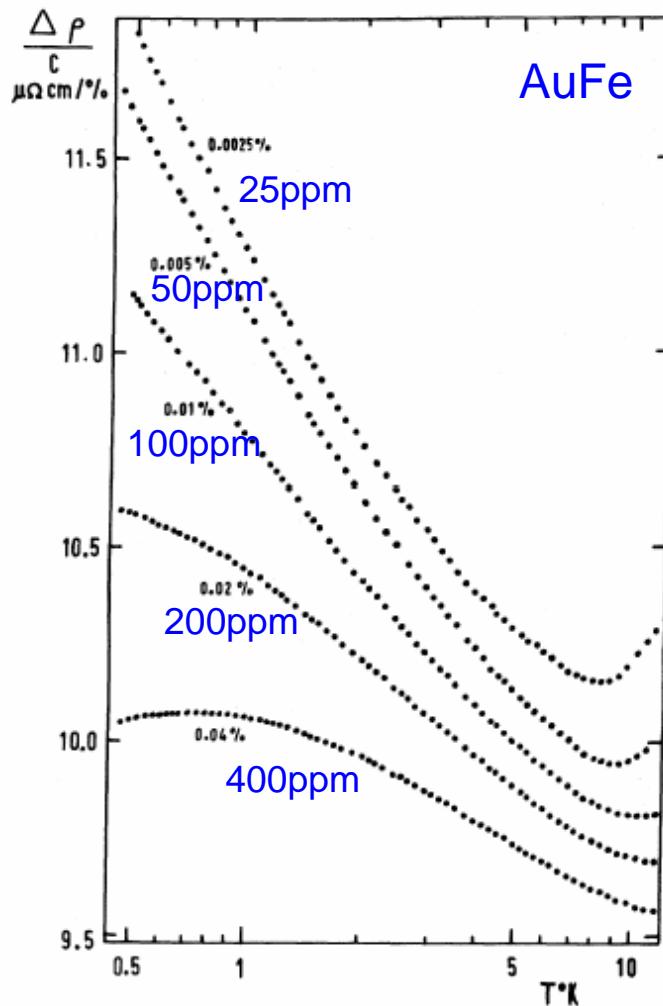


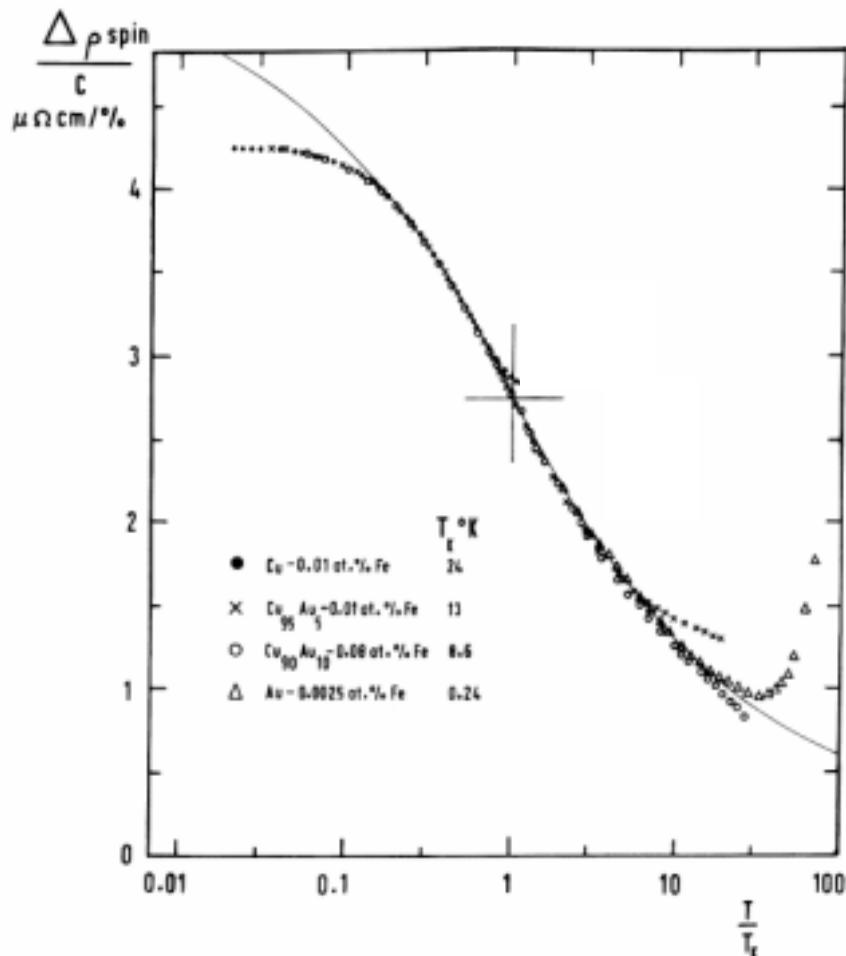
FIG. 3.  $(\Delta\rho/c) \mu\Omega\text{cm}/\text{at. \%}$  against  $\log_{10} T$  for more concentrated AuFe alloys. Nominal Fe concentration is indicated for each alloy.

# Temperature dependence around $T_K$

Kondo effect in resistivity,  $\rho(T)$

When  $T \ll T_K$ ,  $\rho \sim (\rho_0 - cT^2)$  : Unitary limit

When  $T_K \ll T$ ,  $\rho \sim \log(T/T_K)$



When  $T \sim T_K$

Hamann expression for Kondo resistivity

Hamann, Phys. Rev. 158, 570 (1967)

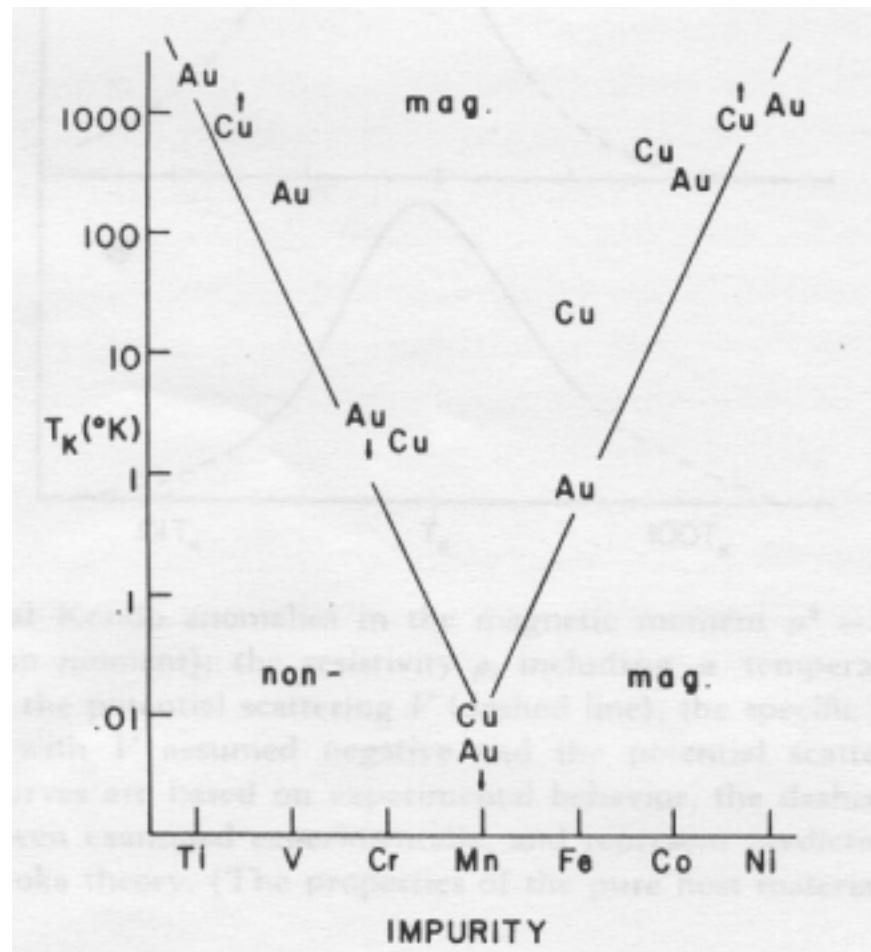
$$\Delta\rho(T) \propto \frac{1}{2} \rho_0 (1 \pm \left[ 1 + \frac{S(S+1)\pi^2}{[\ln(T/T_K)]^2} \right]^{-\frac{1}{2}})$$

$$\begin{array}{lll} T_K > T & , & (-) \\ T_K < T & , & (+) \\ , \rho_0 & & . \end{array}$$

From fitting,  $T_K$  is obtained

$$T_K \sim T_F \exp[-1/\{J N(0)\}]$$

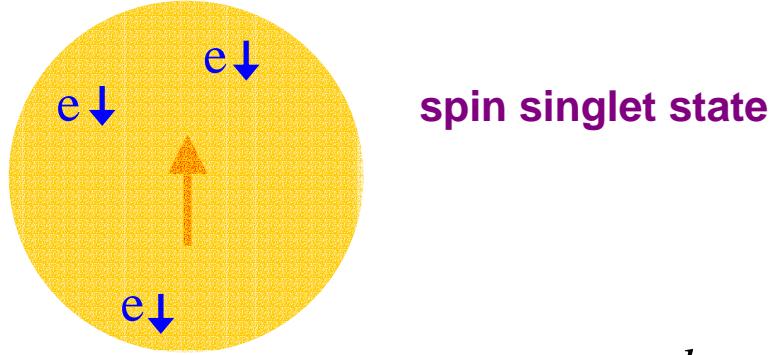
J : s-d exchange coupling, N(0) : conduction electron density of states



# Size of the Kondo screening cloud: the Kondo length

Simple picture

Kondo effect arises from the screening of moment of a magnetic impurity by the conduction electrons in the host metal.



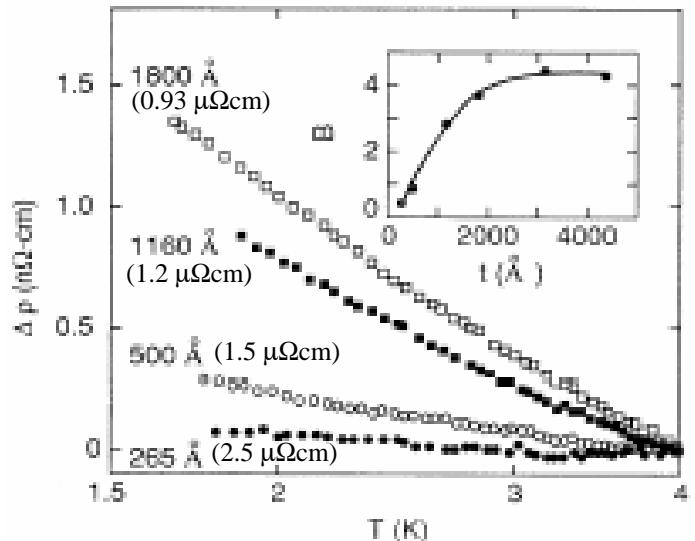
$$\text{Size of the Kondo screening cloud : } \xi_K \approx \frac{\hbar v_F}{kT_K}$$

For AuFe with  $T_K \sim 1$  K,  $\xi_K \sim 10 \mu\text{m}$

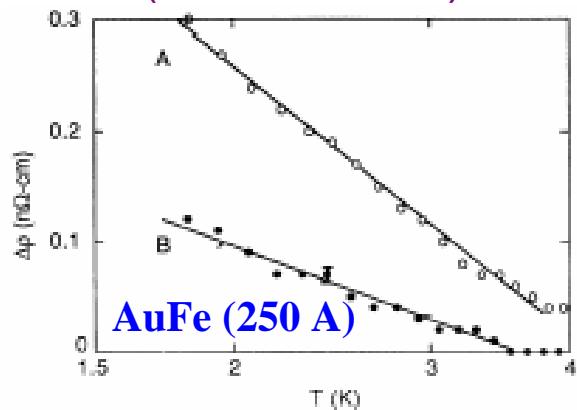
If the Kondo screening cloud is distorted by reducing the sample dimension, the Kondo effect may be suppressed.

# The first observation of size dependence of Kondo effect

AuFe(30ppm) 2D film



Au/AuFe (110 Å / 250 Å)



Chen and Giordano, PRL 66, 209 (1991)

$$\Delta \rho = -B \ln T$$

The coefficient B depends on thickness,  $t$ .

$$\frac{1}{t}$$

kondo slope

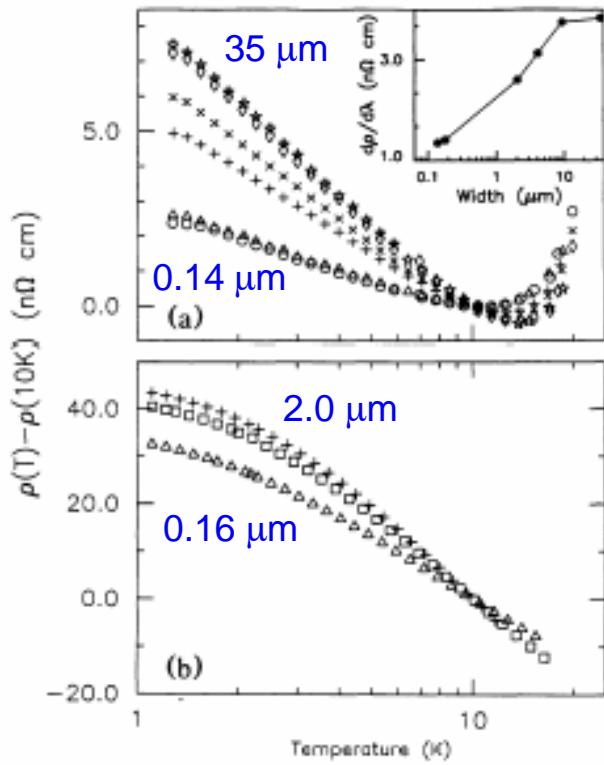
$$\xi_K \approx \frac{\hbar v_F}{kT_K} = 10 \text{ } \mu m \quad \text{where } T_K \sim 1\text{K}$$

Kondo screening cloud can expand to the adjacent Au layer

# Finite size effect in Kondo alloys

CuCr(1000ppm) wire

DiTusa et al., PRL 68, 678 (1992)



(a) 21.2 nm thick film

$$\leftarrow \frac{d\rho}{d(\ln T / T_K)} \text{ vs. width}$$

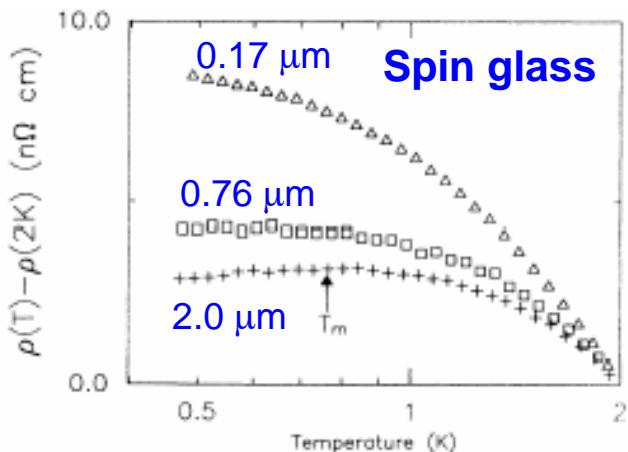
w = 0.14, 0.16, 2, 4, 9.5, 35  $\mu$ m

(b) 98 nm thick film

w = 0.16, 0.76, 2  $\mu$ m

kondo slope

(c) 20 nm thick CuCr(2000ppm) film

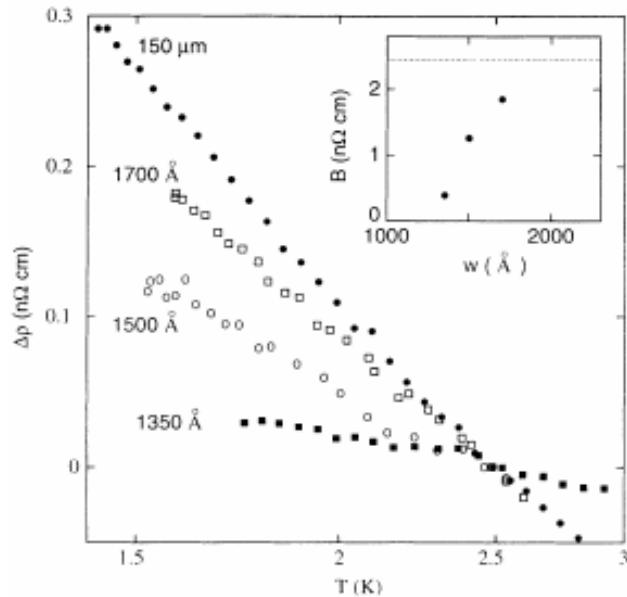


inter-spin interaction

# Finite size effect in Kondo alloys

AuFe(70ppm) wire

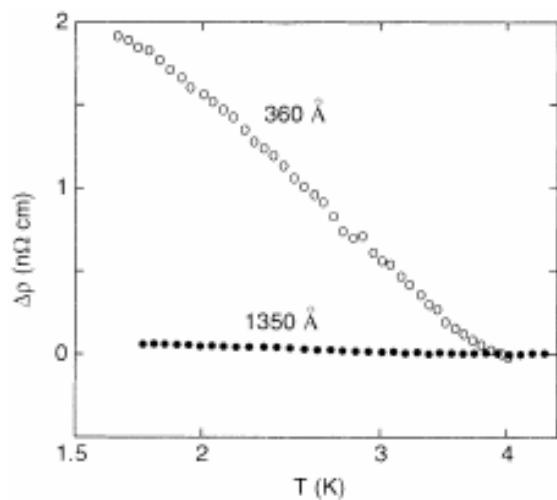
Blachly and Giordano et al., PRB 46, 2951 (1992)



15 nm thick film

Slope decreases as width( $w$ ) is decreased.

kondo slope



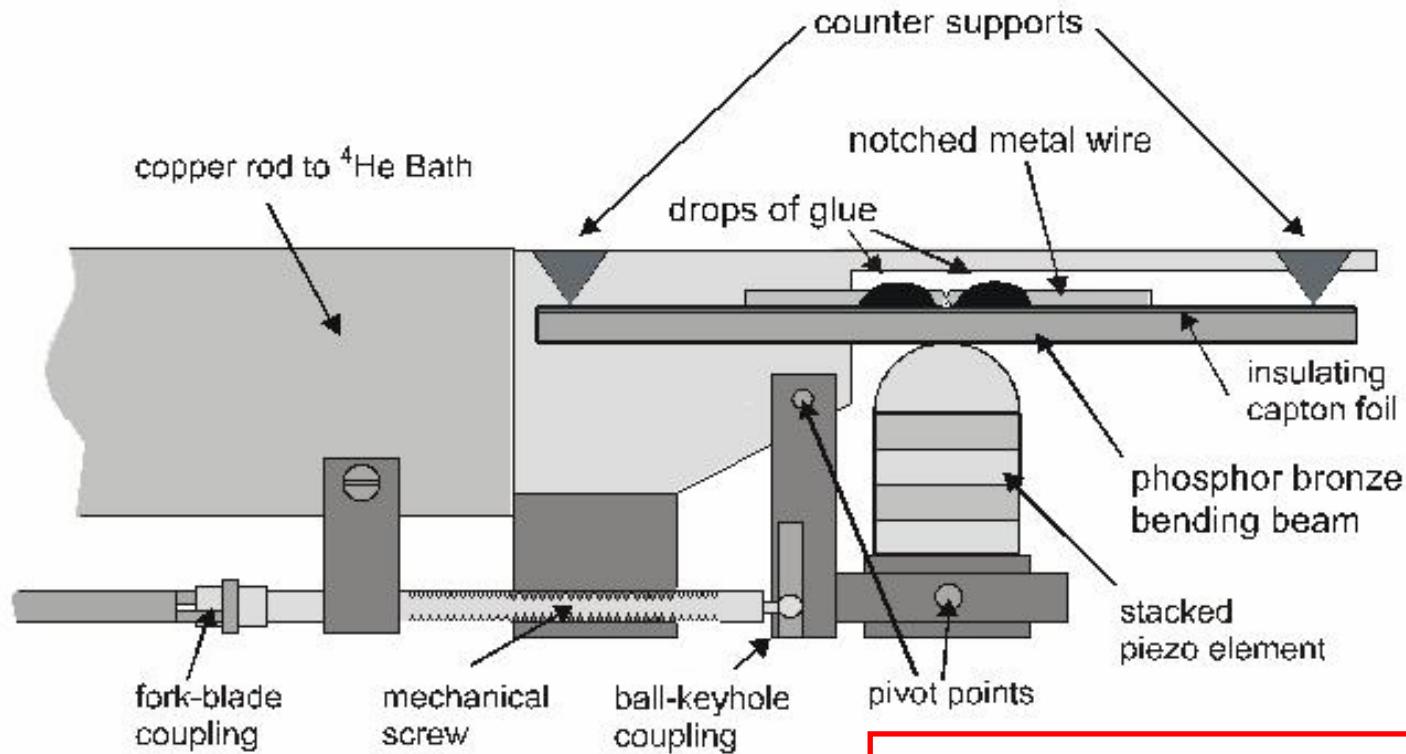
$\Delta\rho$  increases as  $w$  is decreased below 1350  $\text{\AA}$ .

The  $\delta\rho$  by e-e interaction becomes important in 1-D.

$$\delta\rho_{ee}^{(1)} = A_1 \frac{R_S^2 t}{(\pi\hbar/e^2)w} L_T = A_1 \frac{R_S^2 t}{(\pi\hbar/e^2)w} \sqrt{\frac{\hbar D}{kT}}$$

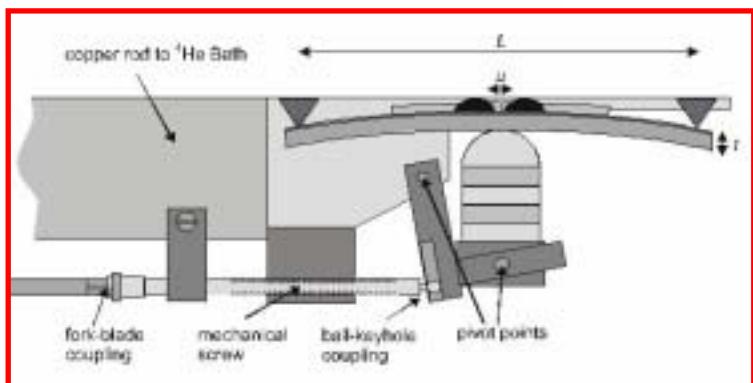
where  $A_1 \sim 1.56$

# Mechanically controllable break junction: A useful tool for size effect study



The diameter of the contact is given by  
Sharvin relation for ballistic contacts  
(Yanson, Sov. J. Low Temp. Phys. 9, 343 (1983))

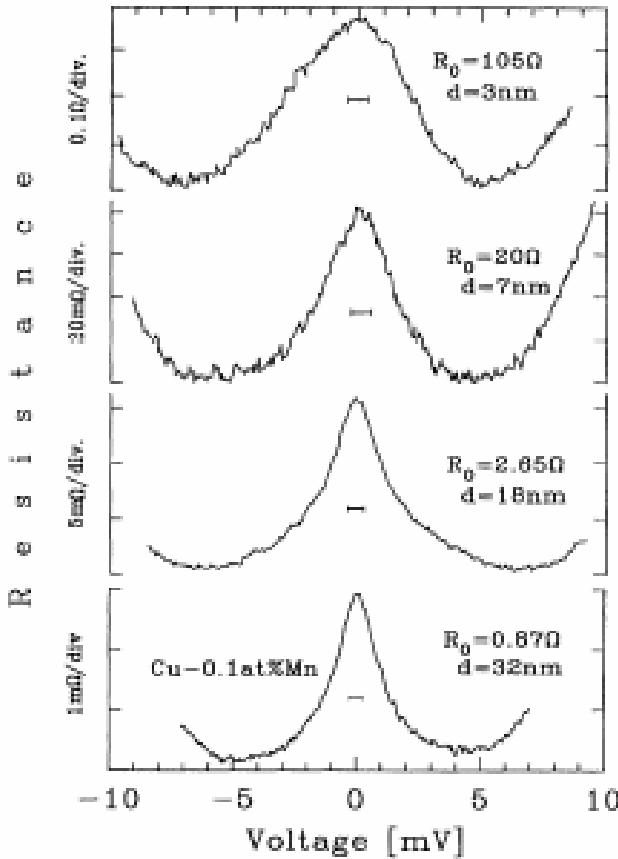
$$k_F d = \sqrt{\frac{8h}{e^2 R}} \quad \left( \text{for } Cu, \quad d[nm] = \frac{30}{\sqrt{R[\Omega]}} \right)$$



# Mechanically controllable break junction

CuMn (1000ppm)

Point contact spectroscopy



$$\frac{\delta R_K}{R_0} \approx 0.003 \text{ for all contact size}$$

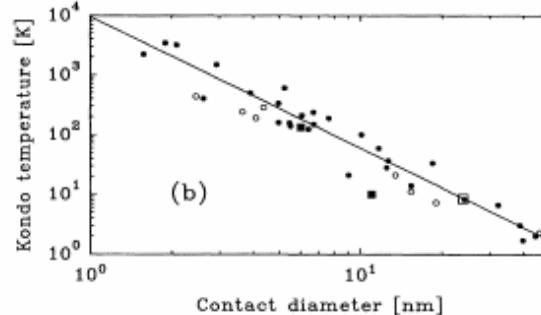
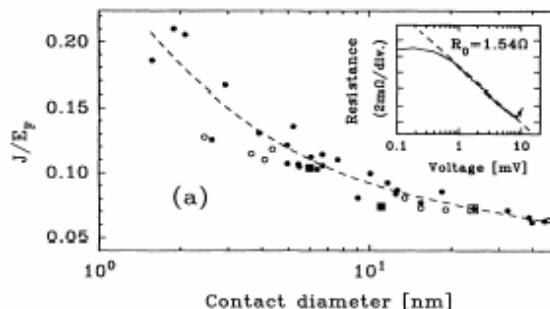
Yanson et al., PRL 74, 302 (1995)

In second-order Born approximation the magnetic impurity contribution to the point contact spectroscopy

Omelyanchuk, Sov. J. Low. Temp. Phys. 11, 211 (1985).

$$-\frac{d^2 I}{dV^2} \approx Q \frac{1}{V} \quad (\text{when } eV \gg kT)$$

$$\text{where } Q = \frac{105}{8} c \frac{k_F d}{R_0} \left( \frac{J}{E_F} \right)^3 \quad \text{for } S(\text{Mn}) = 5/2$$



$$\frac{J}{E_F} = -0.044 \left( \frac{1}{c \sqrt{R_0}} \frac{dR}{d \log_{10} V} \right)^{1/3}$$

c : impurity concentration

$$kT_K = E_F \exp \left( - \frac{2}{3J/E_F} \right)$$

Size 가

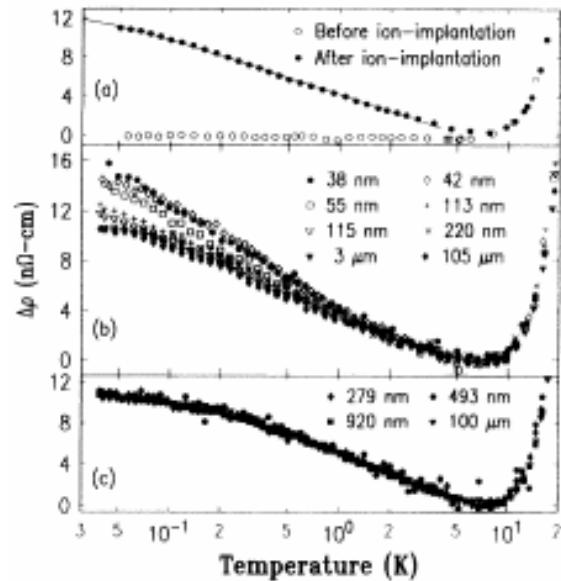
kondo

가 .

# Absence of size dependence of the Kondo resistivity

AuFe (50ppm)

Chandrasekhar et al., PRL 72, 2053 (1994)

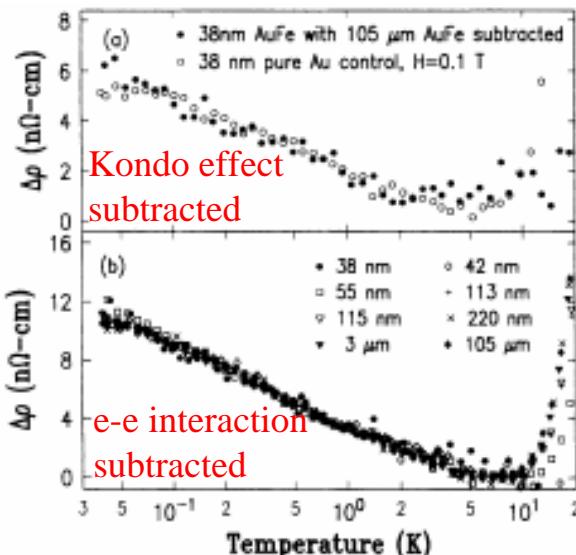


← Slope increases as the width decreases.

← Slope increase is not appreciable when  $w > 300$  nm.

## Contributions to the low temp resistivity

1. Kondo effect
2. e-e interaction (width dependence in 1-D wires)
3. weak localization (negligible in magnetic sample)



For 1-D system,

$$\Delta\rho_{ee}(T) = \alpha \frac{R_s^2 t}{(\pi\hbar / e^2)w} L_t$$

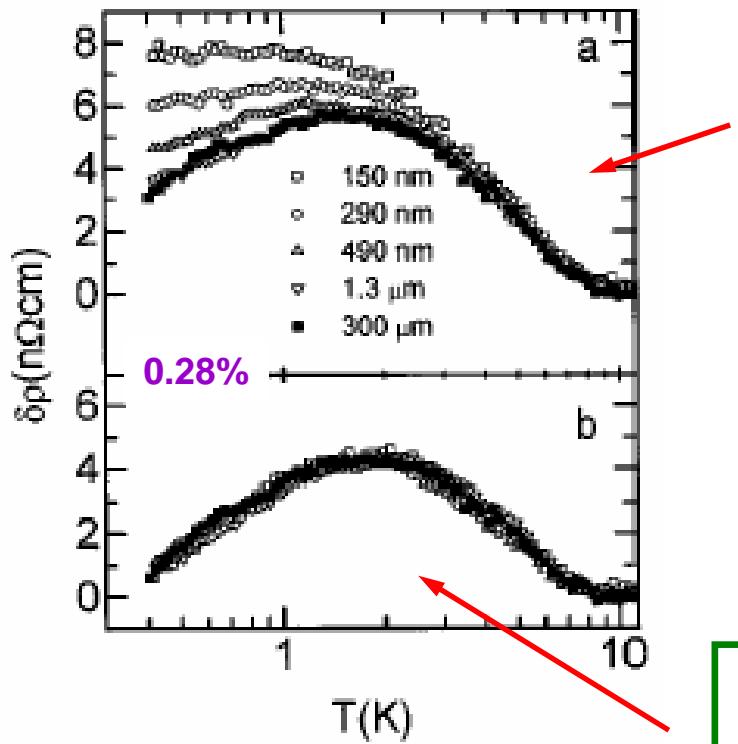
; Altshuler and Aronov, *Electron-electron interactions in disordered systems* (1985)

No size dependence of Kondo resistivity

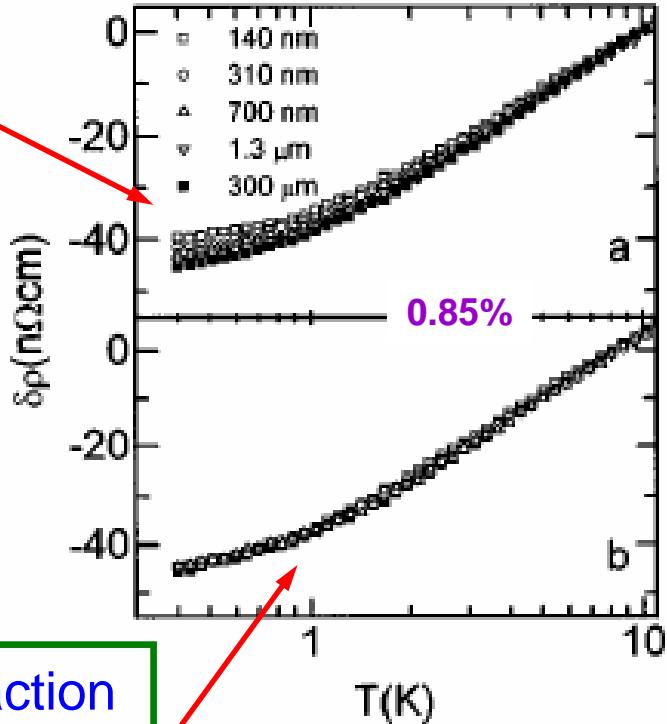
# Size dependence in the spin-glass resistivity of AuFe wires

AuFe (0.28%, 0.85%)

Neuttiens et al., Europhy. Lett. 34, 617 (1996)



Raw data



After e-e interaction  
subtracted

No size dependence of spin-glass resistivity

# Theoretical models for the finite size dependence in the Kondo effect

Spin-orbit interaction gives rise to size dependent magnetic anisotropy.

; Ujsaghy et al., PRL 76, 2378 (1996)

The volume to surface ratio of samples is important factor.

In a disordered samples, weak localization can lead to a finite size dependence in the Kondo resistivity.

; Martin et al., PRL 78, 114 (1997)

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It is obviously interesting to look at other transport properties which are affected by the spin scattering.



thermopower (thermoelectric power)

Mott's formula directly reflects the pronounced energy dependence of the scattering time induced by the magnetic impurity.

; Barnard, *Thermoelectricity in Metals and Alloys* (Taylor & Francis, London, 1972)

# Kondo, Spin glass, Ferromagnet

Noble metal (Au, Ag, Cu, ...) + Magnetic impurity (Fe, Cr, Mn, ...)



: Au

Kondo sample : Au + Fe ( 0.04% )  $T_K = 0.3 \sim 0.8$  K

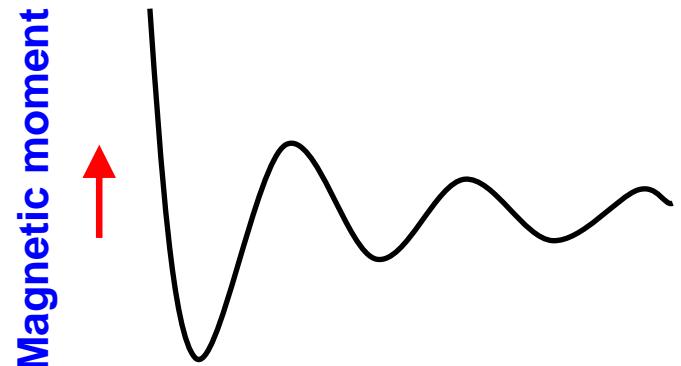
Spin glass : Au + Fe (0.04% ~ 10%)  $T_g = 1 \sim 10$  K

Inhomogeneous Ferromagnet : Au + Fe (10% )  $T_0 > 30$  K

# Metallic Spin Glass

Magnetization of a free electron gas  
by RKKY interaction

$$J_{RKKY}(r) \propto \frac{1}{r^2} \left[ \frac{\cos k_F r}{2k_F r} - \frac{\sin k_F r}{(2k_F r)^2} \right]$$

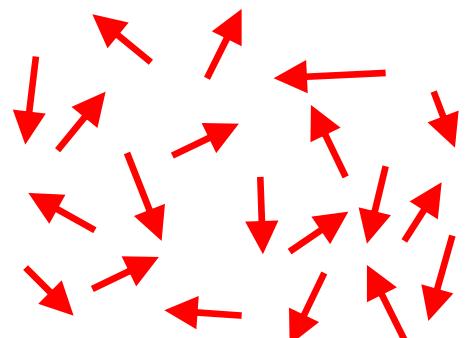


In disordered metal ( $r > l_e$ ) (Jagannathan et al., PRB 37, 436 (1998).)

$$J(r) \propto J \exp(-r / l_e) + 3 \left( \frac{mk_F}{8\pi^3} \right)^2 \frac{1}{r^6}$$

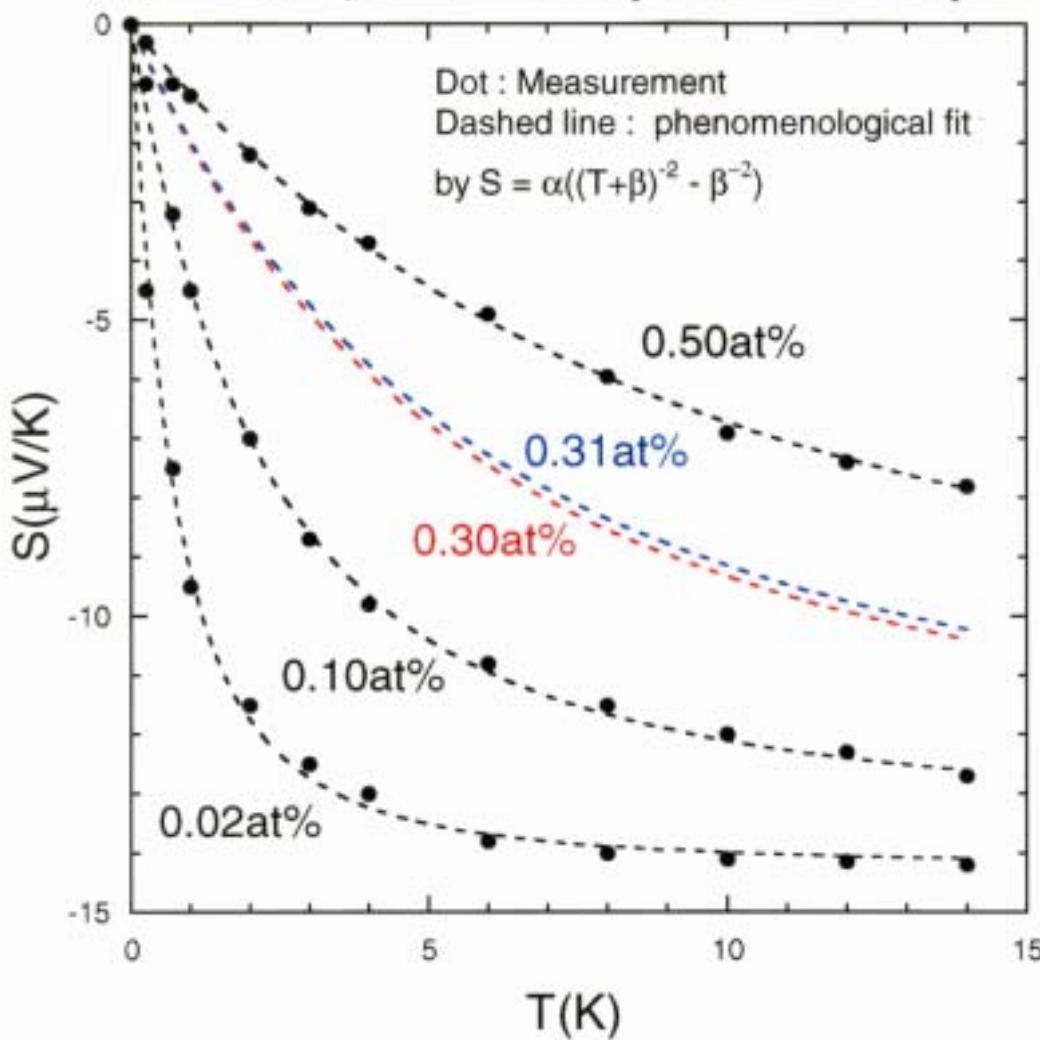
Spins freeze in random directions below  $T_g$ .

- Broad peak in  $R(T)$ ,  $c(T)$
- Sharp peak in  $c(T)$
- Hysteresis and Remanent Magnetization
- No long-range order



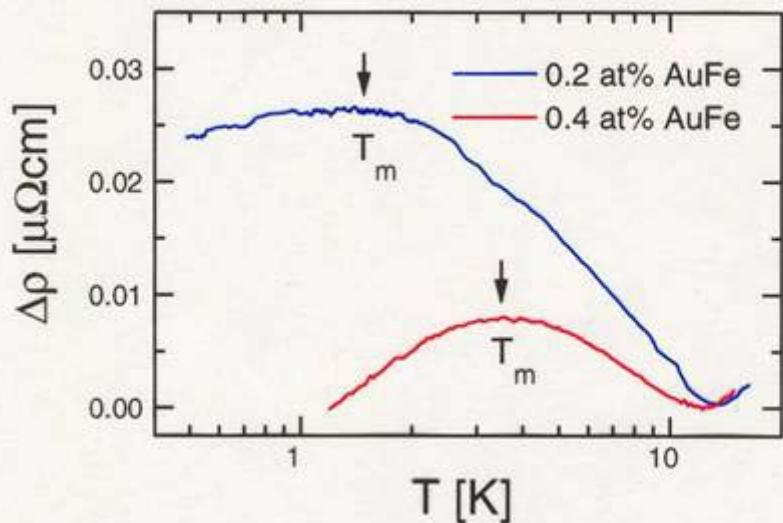
# Thermopower of AuFe

D.K.C.McDonald et al., Proc.Roy.Soc. A266 (1962),166.  
R. D. Barnard, Thermoelectricity in Metals and Alloys.

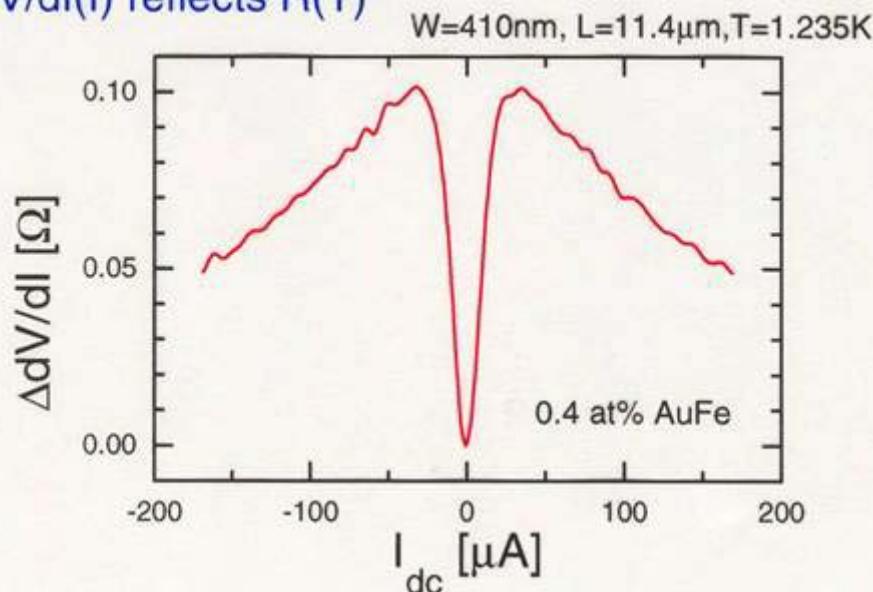


# Differential resistance of AuFe spin glass

Broad resistance maximum in  $R(T)$



$dV/dI(I)$  reflects  $R(T)$



Eom et al., PRL (1996)

Broad resistance maximum.

$T_m$  increases as  $c$  increases.

Differential resistance,  $dV/dI(I)$

$I$                        $I_{ac}$

$I_{ac}$     frequency                      response

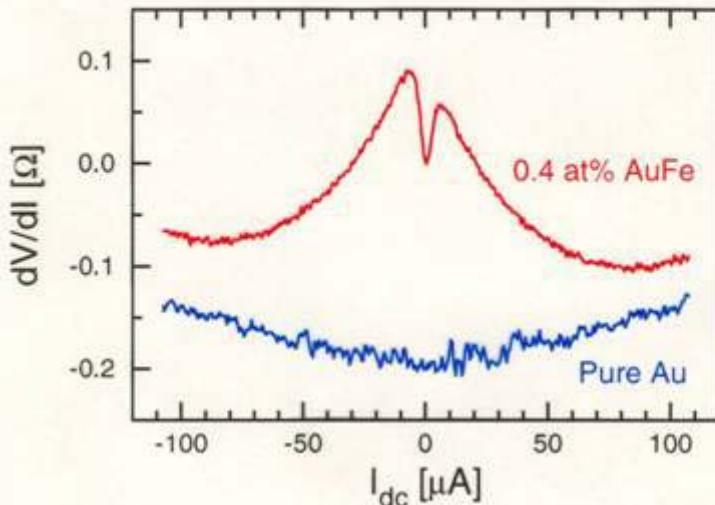
. (Lock amplifier      )

←  $I$

symmetric

# Asymmetric $dV/dI$ of AuFe wire

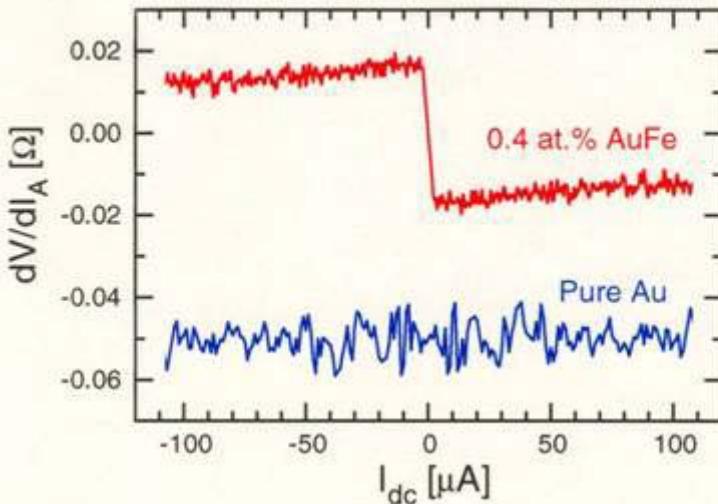
$dV/dI$  of AuFe vs.  $dV/dI$  of Au



$w = 85 \text{ nm}$   
 $T = 1.705 \text{ K}$

$w = 117 \text{ nm}$   
 $T = 1.355 \text{ K}$

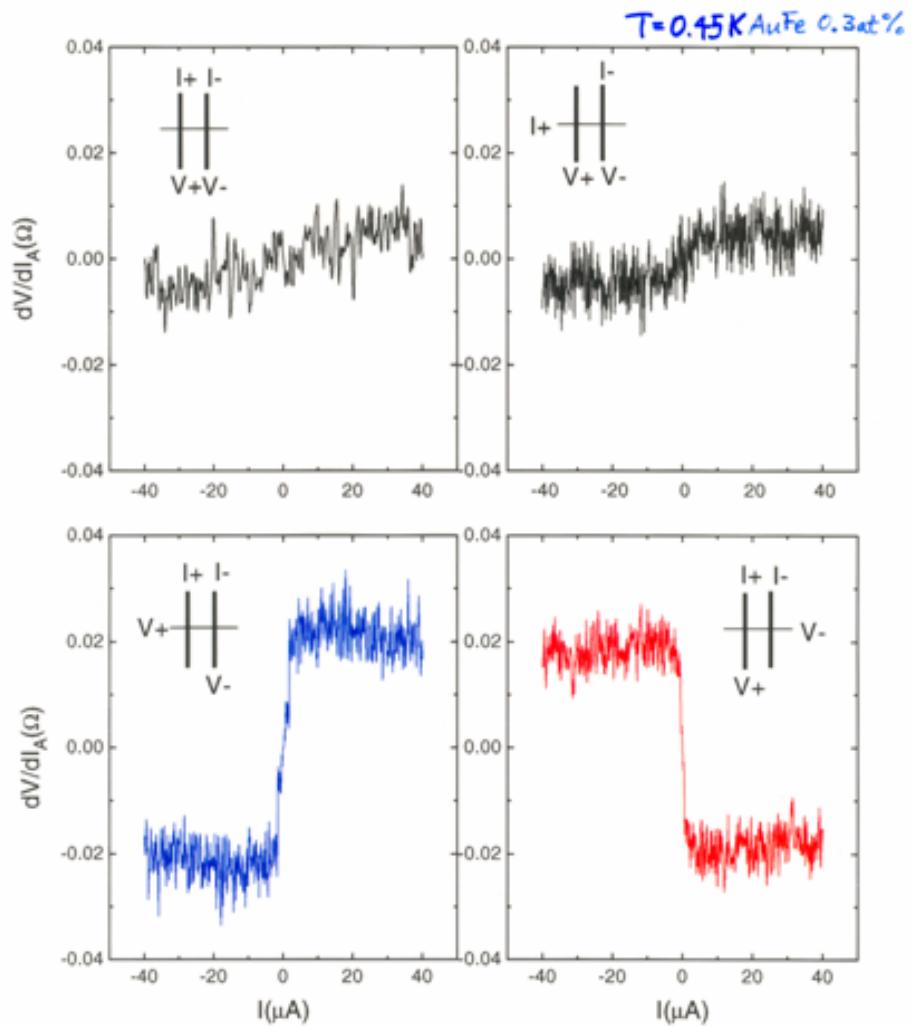
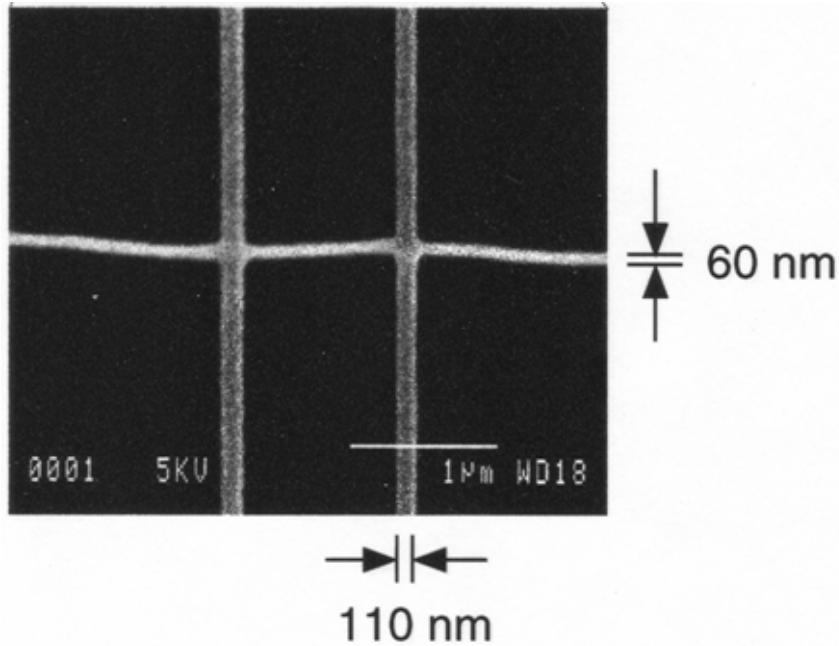
Antisymmetric component,  $dV/dI_A$



Voltage probe width 가  
 $dV/dI$  asymmetric .

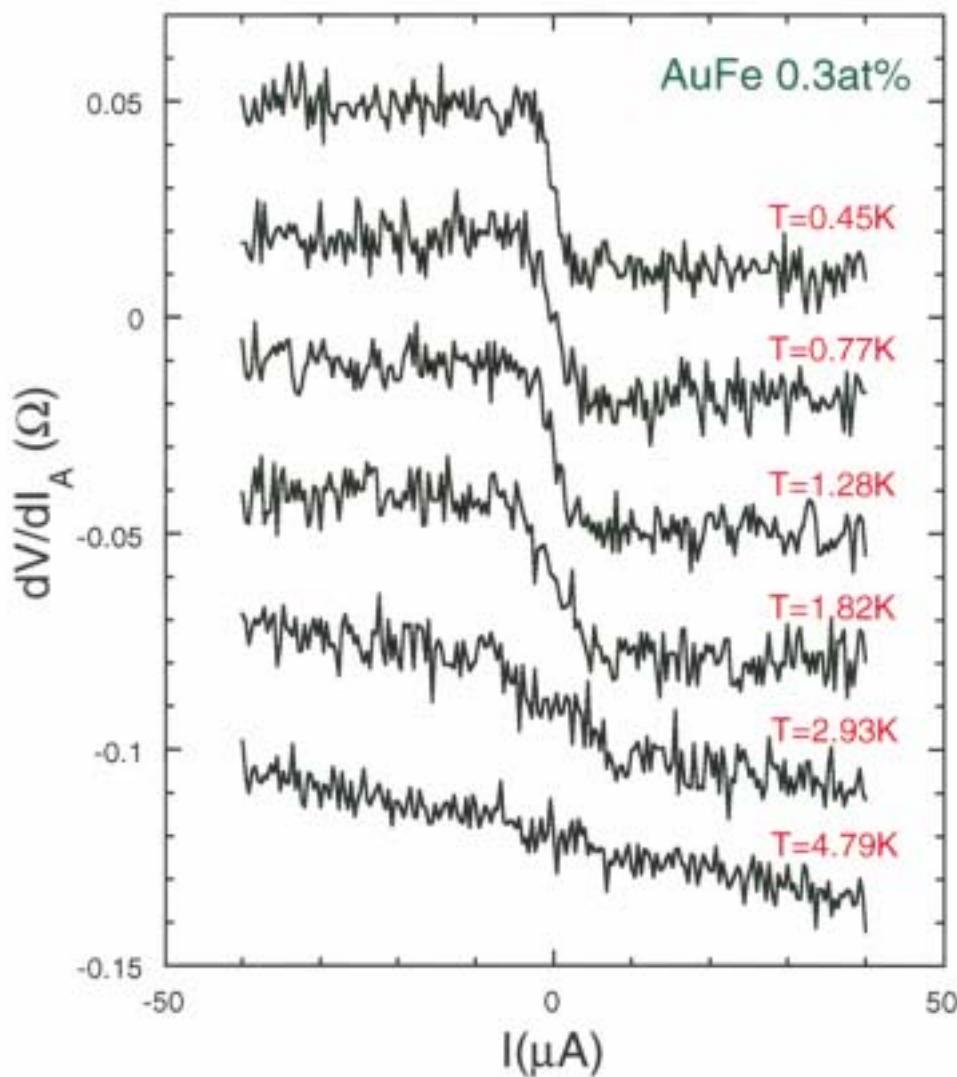
# Antisymmetric part of $dV/dI$

AuFe(0.3%) film thickness = 30 nm



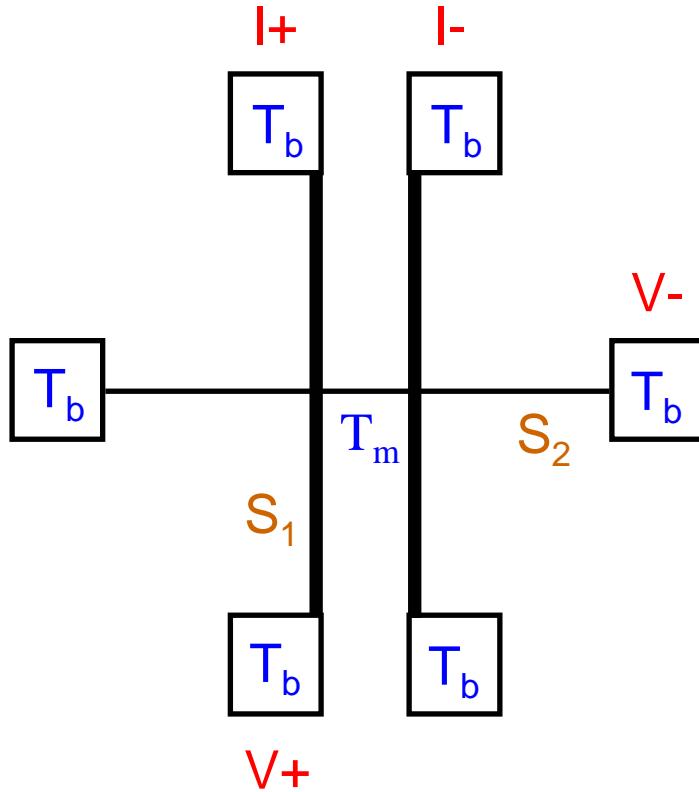
Voltage probe width 가  
 $dV/dI$  asymmetric .

# Temperature dependence of antisymmetric part of $dV/dI$



# Thermoelectric effect

Measurement configuration



Measured voltage

$$V = I R + \int_{T_b}^{T_m(I)} S_1 dT + \int_{T_m(I)}^{T_b} S_2 dT$$

$$= I R + \int_{T_b}^{T_m(I)} (S_1 - S_2) dT$$

where  $S_1$  : thermopower of wide wire  
 $S_2$  : thermopower of narrow wire

$$\frac{dV}{dI} = R + [S_1 - S_2]_{T=T_m} \times \frac{dT_m}{dI}$$

Resistance  
(symmetric in  $I$ )

thermoelectric part  
(antisymmetric in  $I$ )

$T_m(I)$

$dV/dI$

wire width

$\Delta S$

.

# Electron heating by dc current

K.E. Nagaev, PRB 52, 4740 (1995)

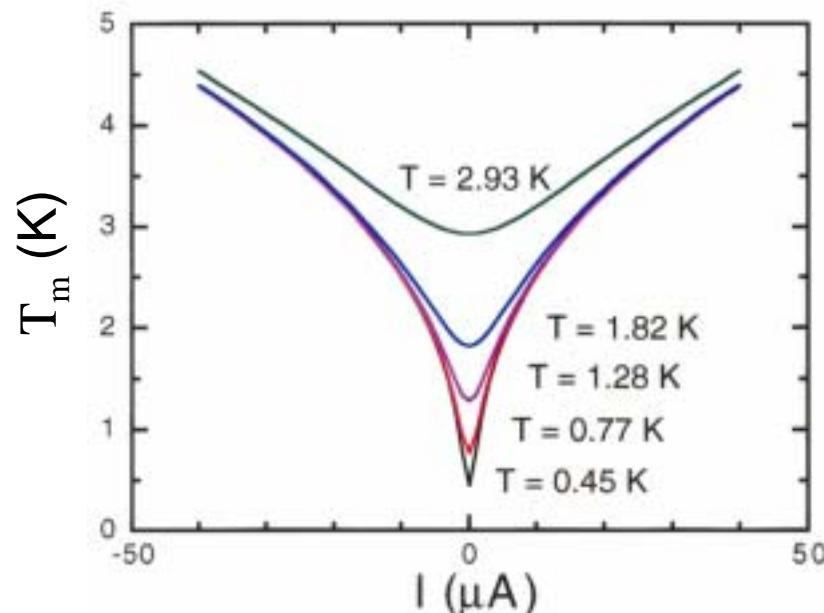
$$\frac{\pi^2}{3} k_B^2 L^2 \frac{\partial}{\partial x} \left( T_e \frac{\partial T_e}{\partial x} \right) = -(eIR)^2 + \frac{24\zeta(5)\alpha_{ph}L^2k_B^5}{(k_B\Theta_D)^2\hbar D} (T_e^5 - T_b^5)$$

Electronic heat conduction  
through wire itself

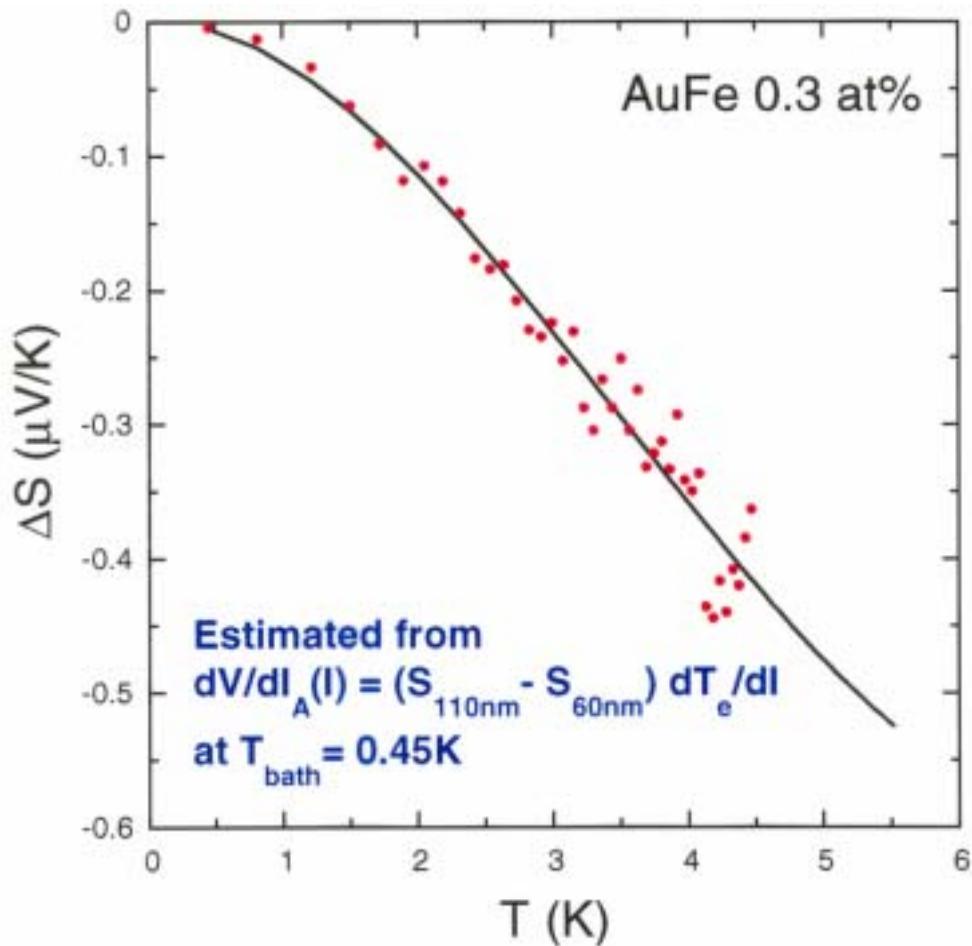
Joule heating

electron-phonon coupling

$T_m(I)$  can be obtained for each bath temperature  $T_b$



$$\Delta S (= S_{110\text{nm}} - S_{60\text{nm}})$$

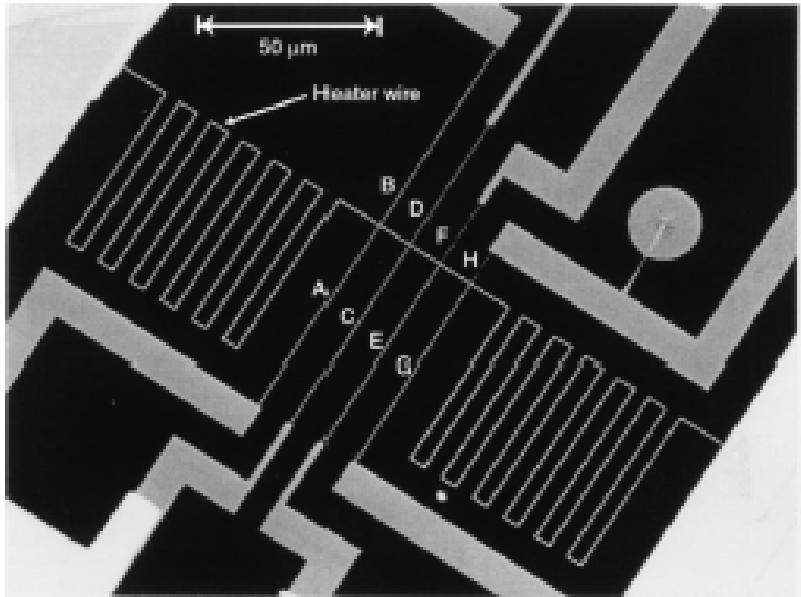


AuFe spinglass      thermopower      size effect 가 .

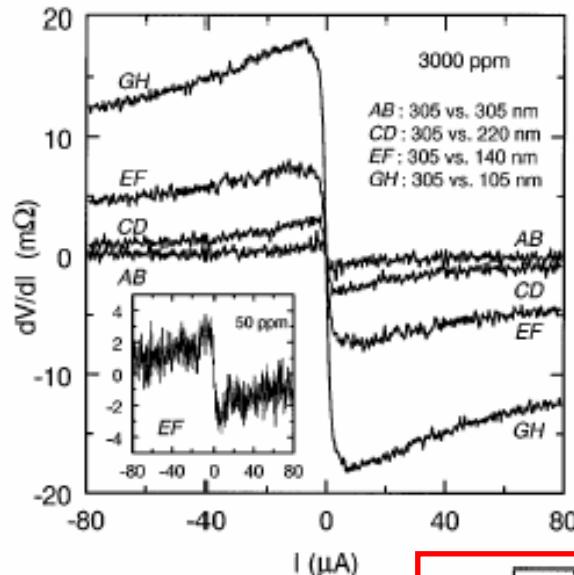
# Size dependent thermopower in Mesoscopic AuFe wires

AuFe(0.3%) film thickness = 30 nm

Strunk et al., PRL 81, 2982 (1998)



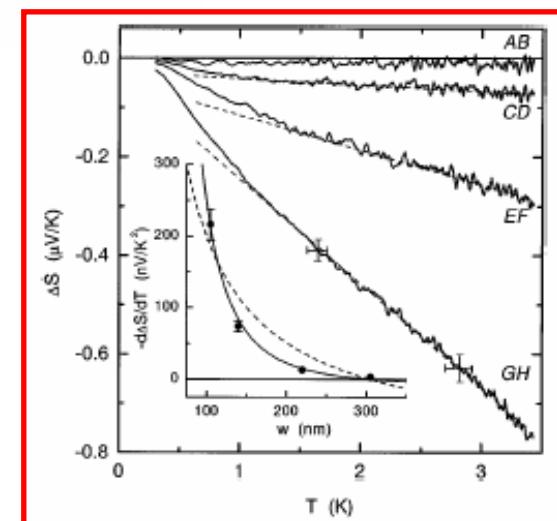
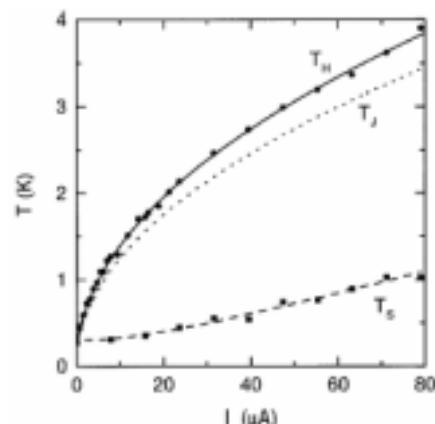
Antisymmetric part of  $dV/dI$



Measuring  $T_m$  by noise spectrum

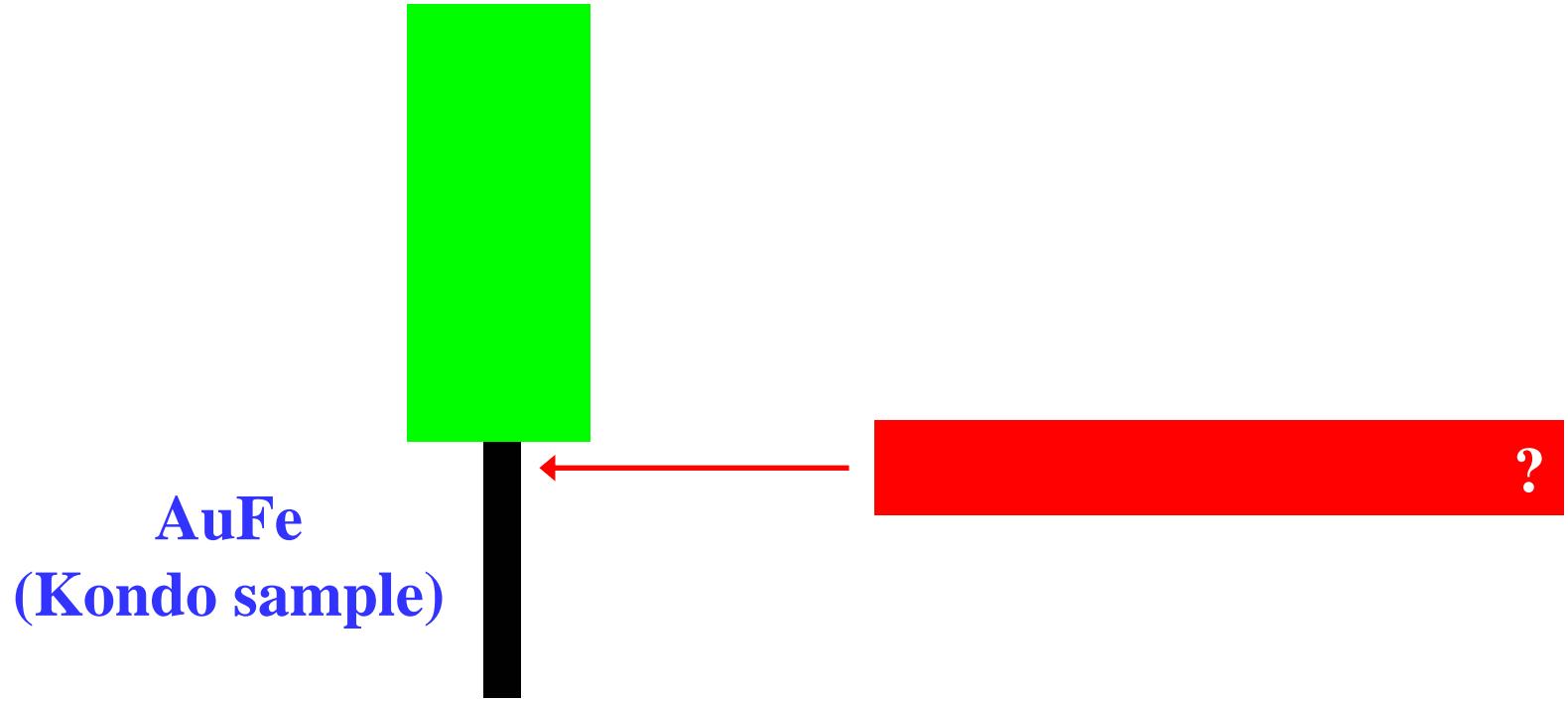
; Henny et al., APL 71, 773 (1997)

$$S_V(I) = 4kT_m(I)R$$



# Superconducting proximity effect in Kondo alloys

Eom, JKPS 42, L313 (2003)



Magnetic impurity

phase coherence length ( $L_\phi$ )가

,

(

)가

?

# Motivation

?

$$\text{exchange field} \Rightarrow \xi_{ex} = \sqrt{\frac{hD}{2\pi k_B T_{Curie}}} \approx 1\text{-}3 \text{ nm}$$

: transport

(Kawaguchi et al., PRB 1992 : Fe/Nb multilayer)

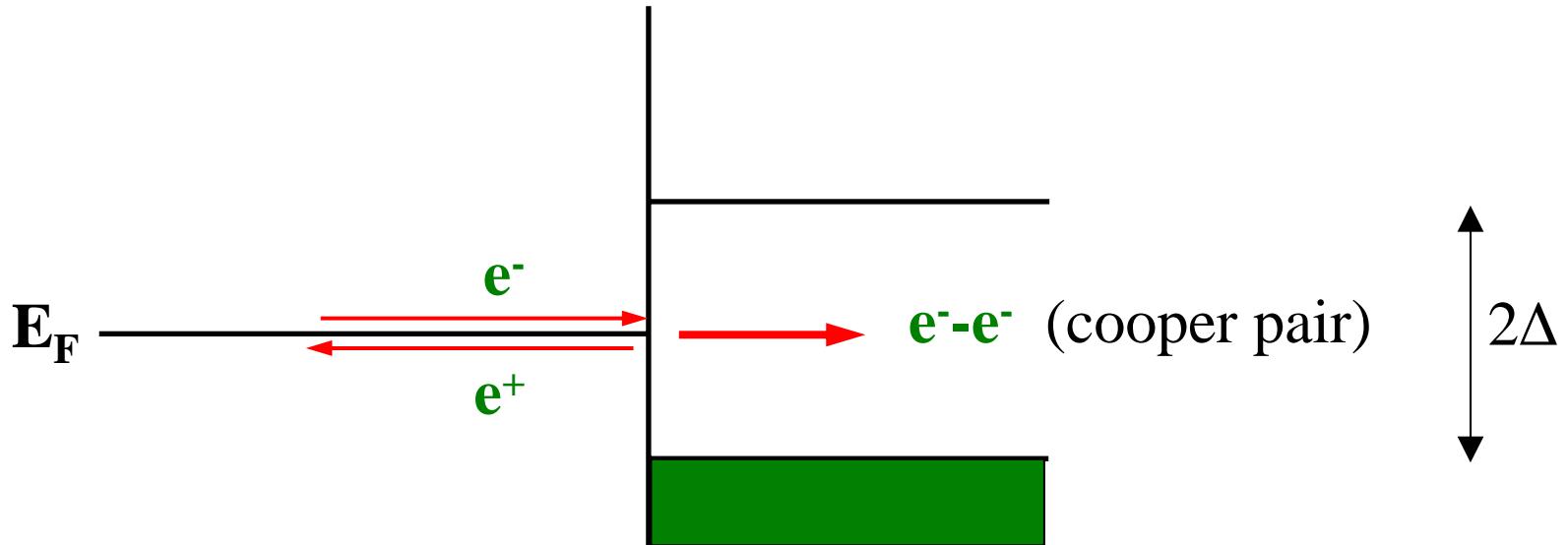
## mesoscopic F/S structures

1. Superconducting proximity effect in a mesoscopic ferromagnetic wire ; Giroud et al., PRB (1998).
2. Proximity effects in superconductor-ferromagnet junctions ; Lawrence and Giordano, J. Phys. (1999).
3. Giant mutual proximity effects in F/S nanostructures ; Petrashov et al., PRL (1999).

=>  $\xi_{ex}$

# Superconducting proximity effect

: Andreev



1.  
2.

가  
cooper pair

가 .

(a)  
(b)

:

(Phase-memory effect).  
(Retro-reflection).

/ (N/S)

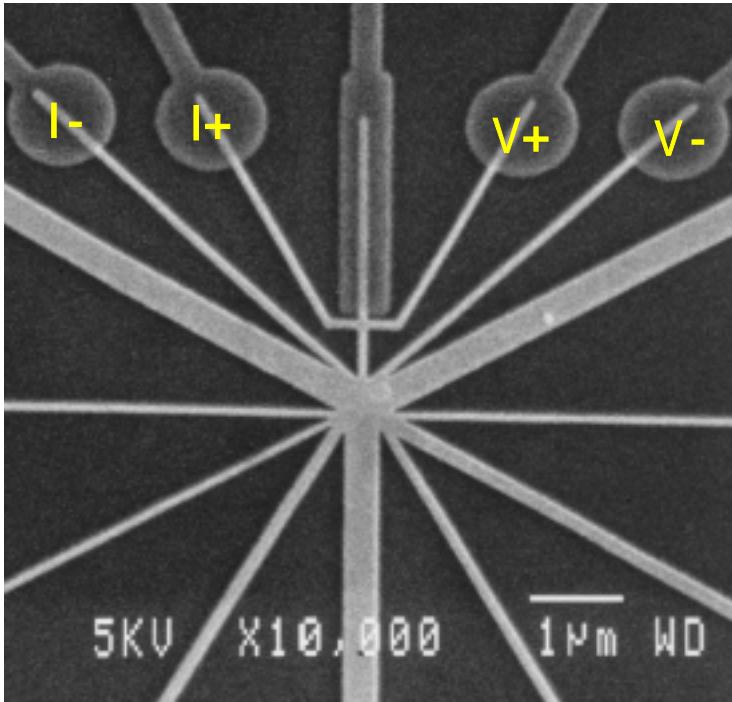
- pair correlation

$$L_T \quad (= (hD/2\pi kT)^{0.5})$$

- Au                     $L_T$                     1 K                     $\sim 0.3 \mu\text{m}$

: Hekking, Schoen, Averin, *Mesoscopic Superconductivity*  
Physica (Amsterdam) 203B (1994).

AuFe /Al



## Kondo sample

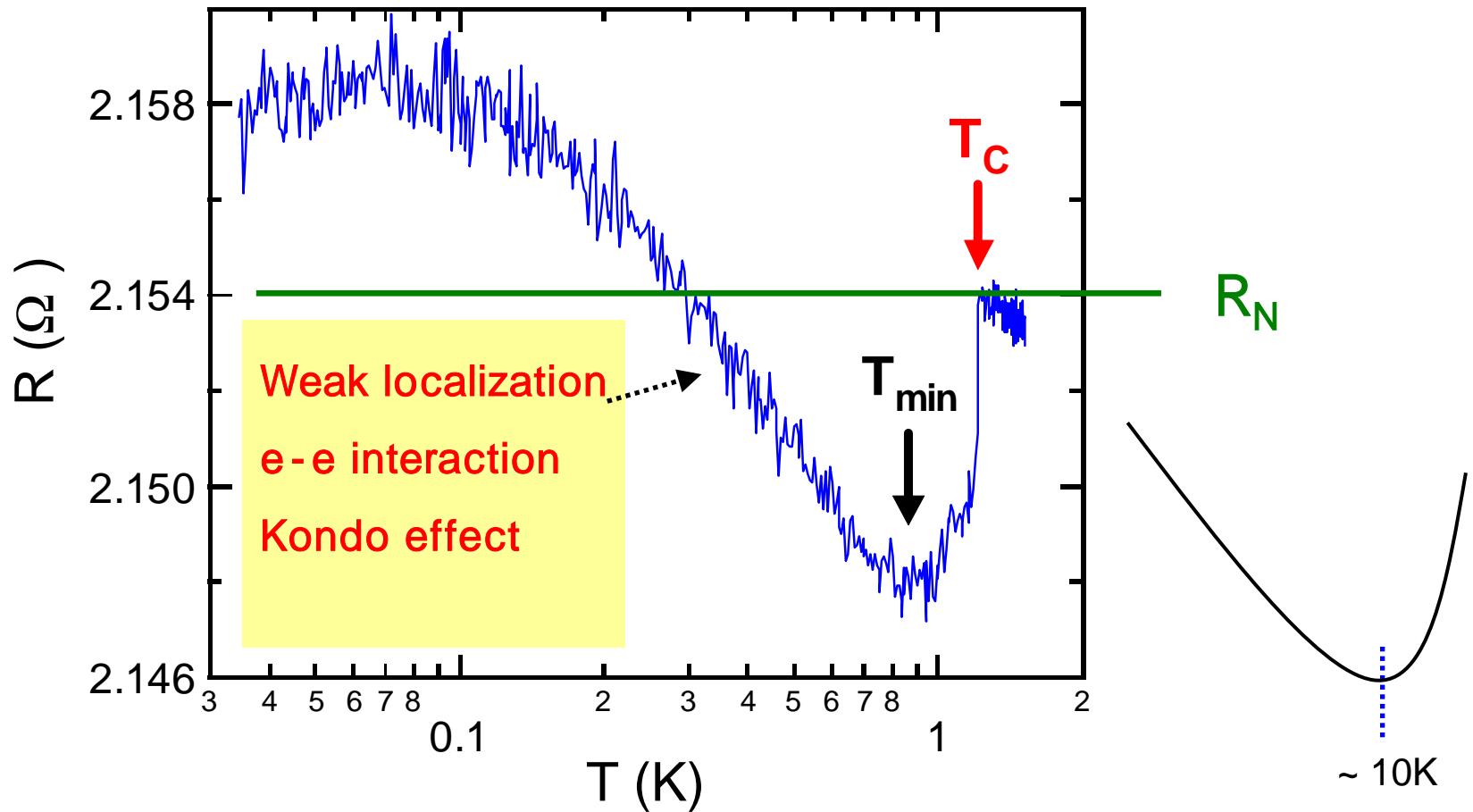
Fe : 100 ppm (0.01%)

Film

AuFe = 51 nm

Al = 75 nm ( $T_c = 1.15$  K)

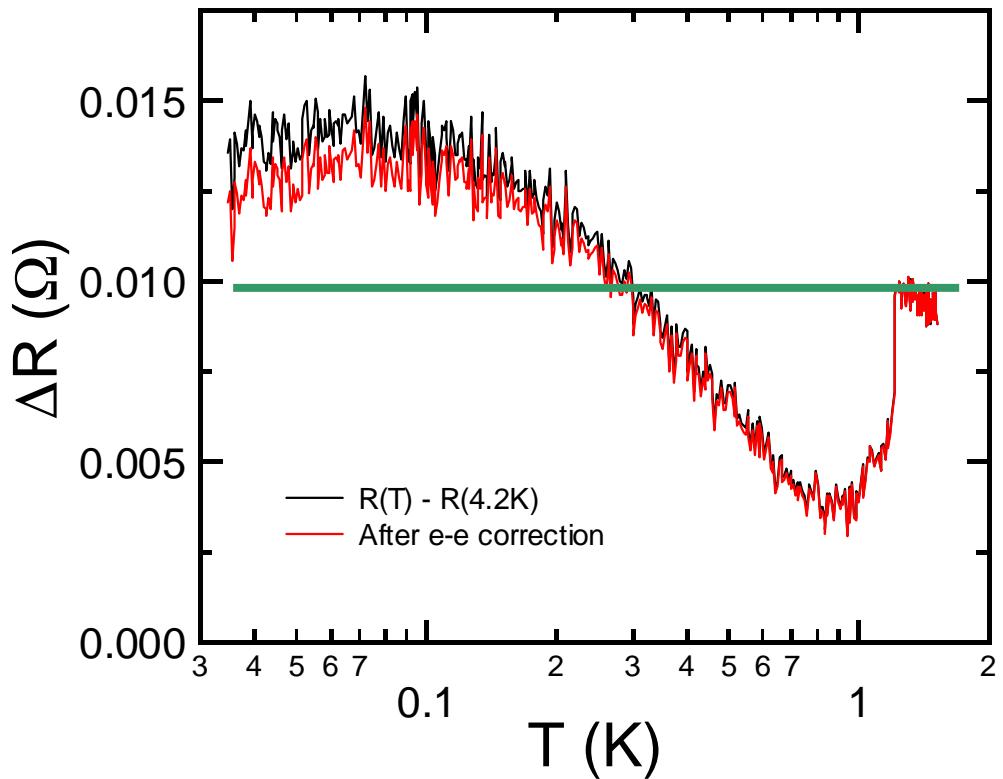
AuFe/



# Repulsive e-e interaction correction

Altshuler and Aronov (1985)

$$\delta\rho_{ee}(T) - \delta\rho_{ee}(T_0) = \alpha \frac{R_{\square}^2 t}{(\pi\hbar / e^2)W} L_T$$



e - e interaction

# Temperature dependence of Kondo resistivity

When  $T \ll T_K$ ,  $\rho \sim \rho_0 - cT^2$

$T_K \ll T$  ,  $\Delta\rho(T) \propto \ln(T / T_K)$

$T \sim T_K$  , Hamann expression (Phys. Rev. 1967)

$$\Delta\rho(T) \propto \frac{1}{2} \rho_0 (1 \pm \left[ 1 + \frac{S(S+1)\pi^2}{[\ln(T / T_K)]^2} \right]^{-\frac{1}{2}})$$

$T_K > T$  , (-) ,

$T_K < T$  , (+) .

,  $\rho_0$  .

$\Delta\rho$

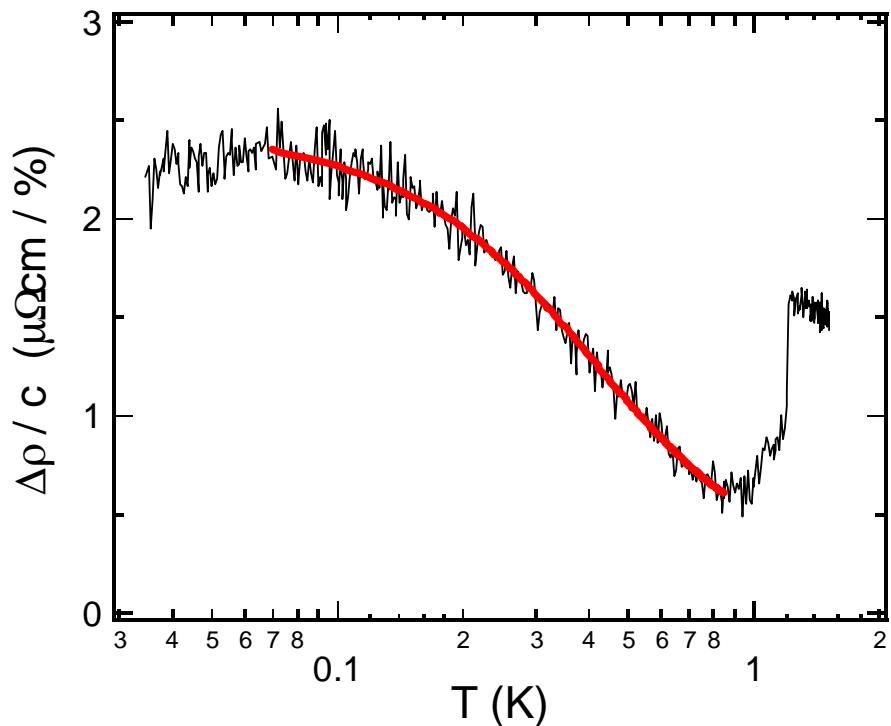
fitting

$T_K$

$S$

.

# Fitting by Hamann expression



Fit parameter :  $T_K = 0.41$  K,  $S = 0.12$

$T_K$

AuFe  $T_K$

$S$   $(0.5 \sim 1.0)$

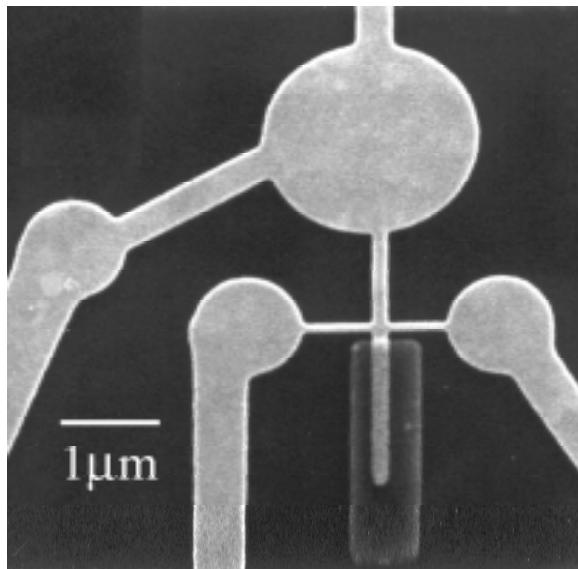
가

=>

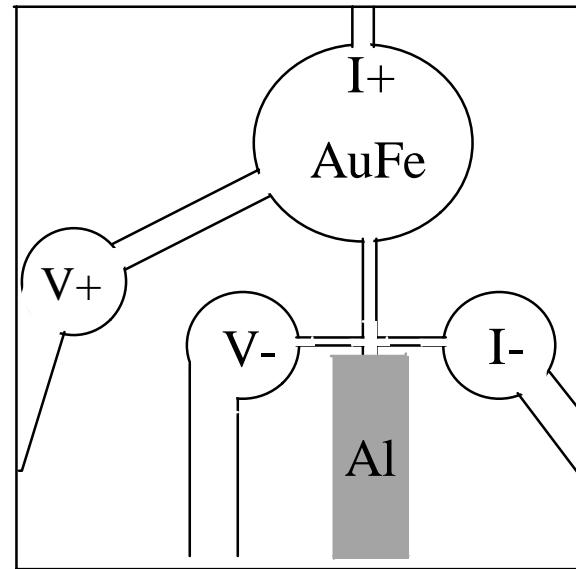
Kondo

AuFe /Al

Fe : 100 ppm (0.01%)



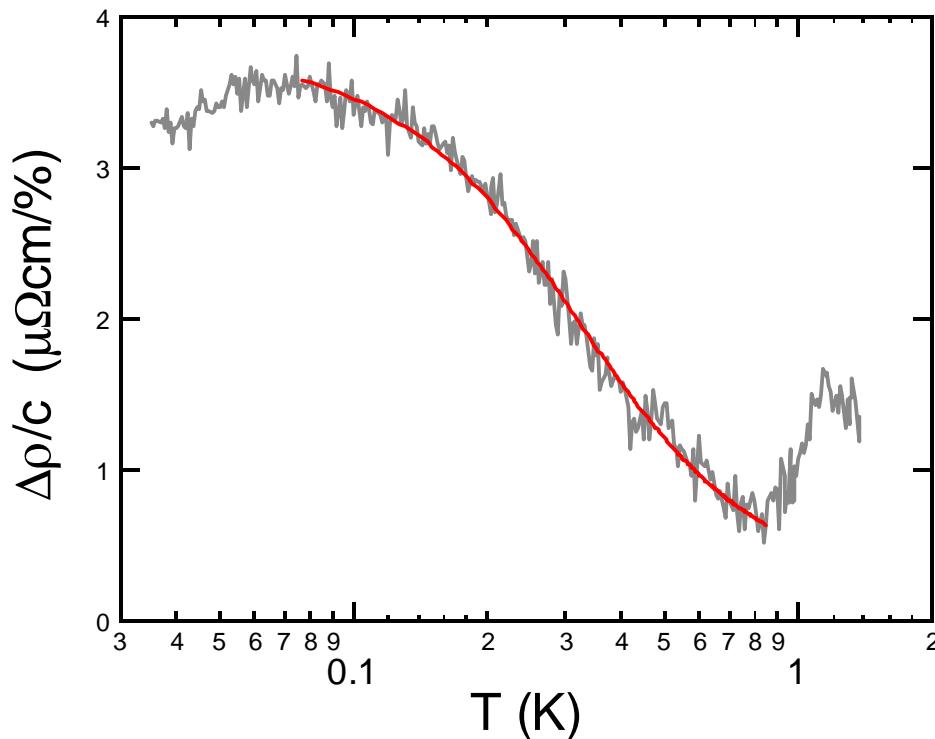
Al = 75 nm  
AuFe = 51 nm



AuFe/

.

# Fitting by Hamann expression



Fit parameter :  $T_K = 0.32$  K,  $S = 0.10$

$T_K$	AuFe	$T_K$	.
$S$	$0.5 \sim 1.0$	가	.

=>

Kondo

# Mesoscopic dilute magnetic systems

- Kondo regime

**Kondo length for a typical AuFe films:**

$$\xi_K = \frac{hv_F}{k_B T_K} \approx 20\mu m \quad \text{in ballistic limit}$$

$$\xi_K = \sqrt{\frac{hD}{k_B T_K}} \approx 0.5\mu m \quad \text{in diffusive limit}$$

lithography ,  
가 .

## **II. Kondo effect in low- dimensional systems**

**Kondo effect in a single electron transistor**

**Kondo effect in carbon nanotubes**

**Kondo effect in a single molecule transistor**

# Kondo effect in a single electron transistor

## MIT and Weizman Institute of Science

- Kondo effect in a SET; Nature 39, 156 (1998).
- From the Kondo regime to the mixed-valence regime in a SET ; PRL 81, 5225 (1998)

## Delft University, Netherlands

- A tunable Kondo effect in quantum dots; Science 281, 540 (1998)
- The Kondo effect in the unitary limit; Science 289, 2105 (2000)
- Kondo effect in an integer-spin quantum dot; Nature 405, 764 (2000)

## Ludwig-Maximilians-Universitat, Germany

- Anomalous Kondo effect in a QD at nonzero bias; PRL 83, 804 (1999)

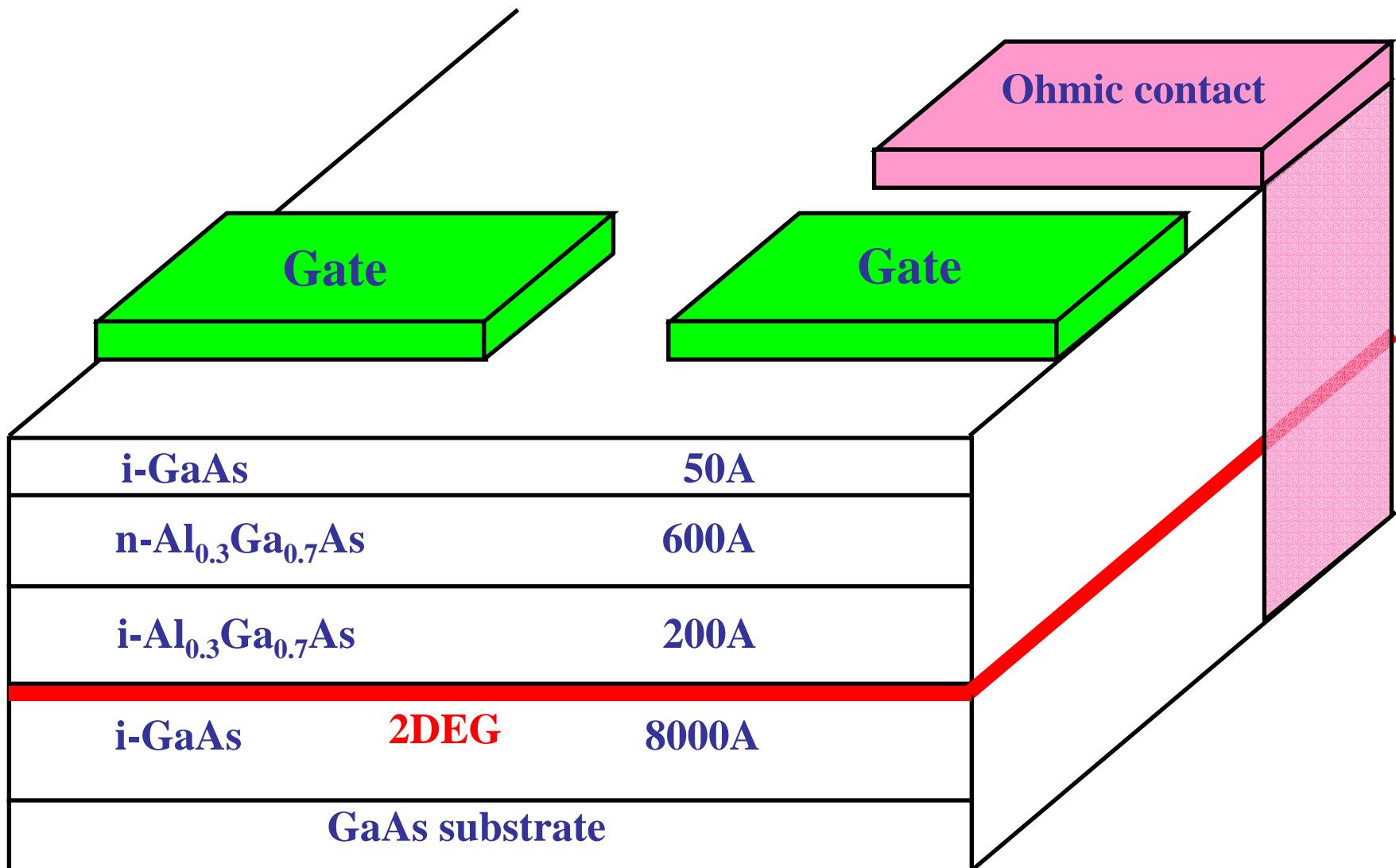
## Max-Planck-Institute, Germany

- Quantum dot in high magnetic fields; PRB 64, 033302 (2001)

## Cavendish Laboratory, UK

- Kondo effect in a quantum antidot; PRL 89, 226803 (2002)

# Example of sample structures of 2DEG



# Coulomb blockade oscillation

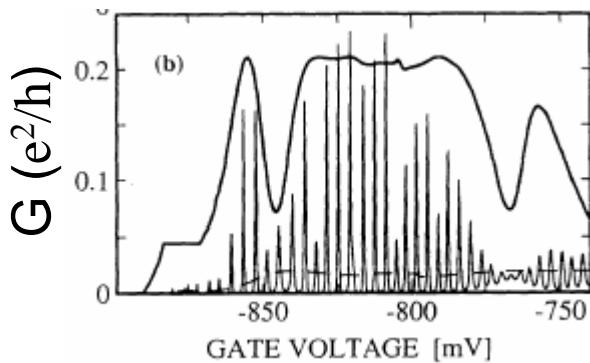
When  $kT < e^2/2C$

Kastner, Phys. Today 24 (Jan 1993)

Electrostatic energy of a charge  $Q$  on the dot

$$E = QV_g + \frac{Q^2}{2C} = \frac{1}{2C} (Q + CV_g)^2 - CV_g^2$$

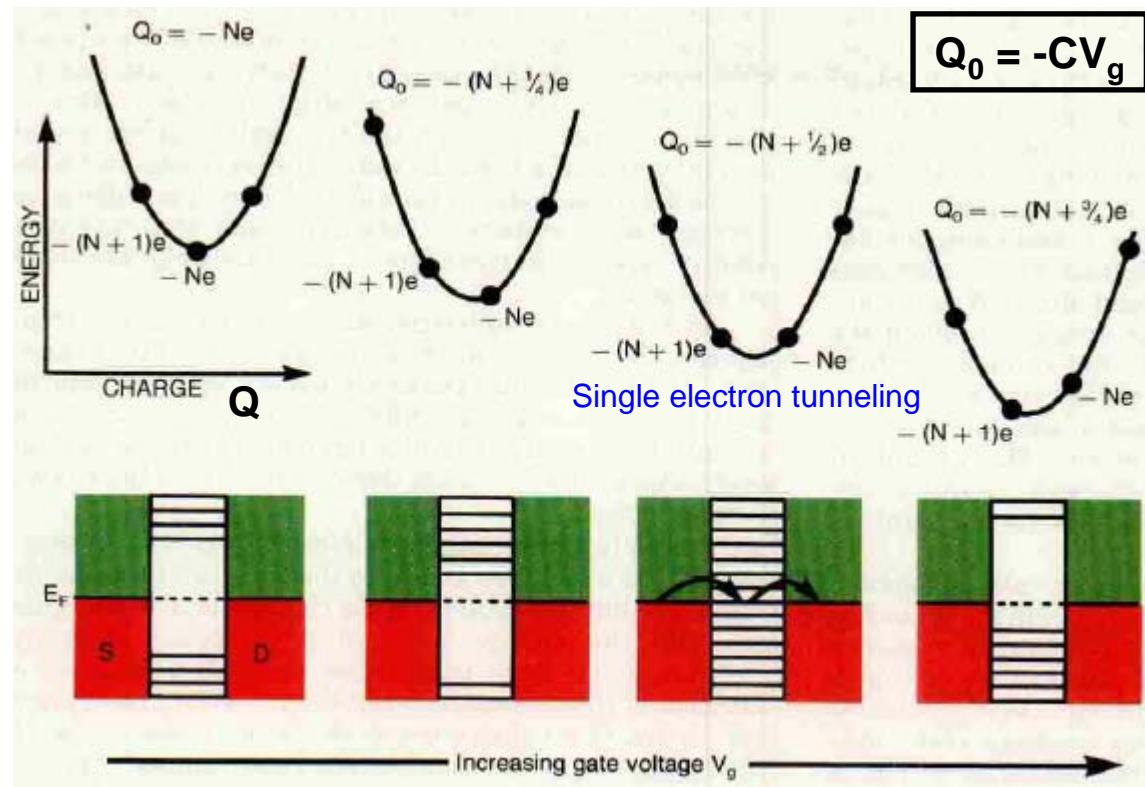
$$Q = \dots, -(N+1)e, -Ne, -(N-1)e, \dots$$



Johnson et al., PRL 69, 1592 (1992)

Evenly spaced oscillation

$$\Delta V_g = e/C$$



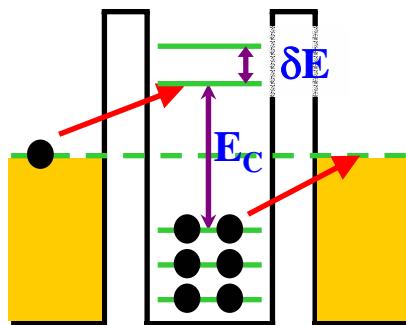
# Co-tunneling (2<sup>nd</sup> order process)

Glazman and Pustilnik, cond-mat/0302159

At low temperatures,  $T \ll E_C$ , conduction through the dot in the Coulomb blockade valleys is exponentially suppressed.

Going beyond the lowest-order perturbation, the following “co-tunneling” process contributes to the activationless transport.

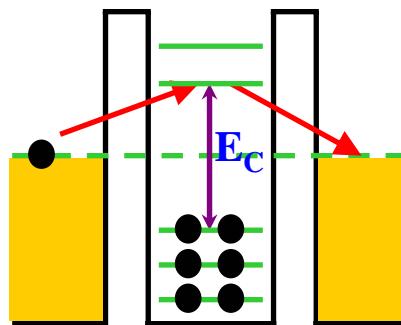
Inelastic co-tunneling



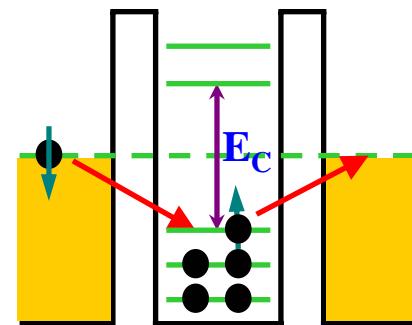
$$G_{in} \propto \frac{e^2}{h} G_L G_R \left( \frac{T}{E_C} \right)^2$$

e-h pair

Elastic co-tunneling



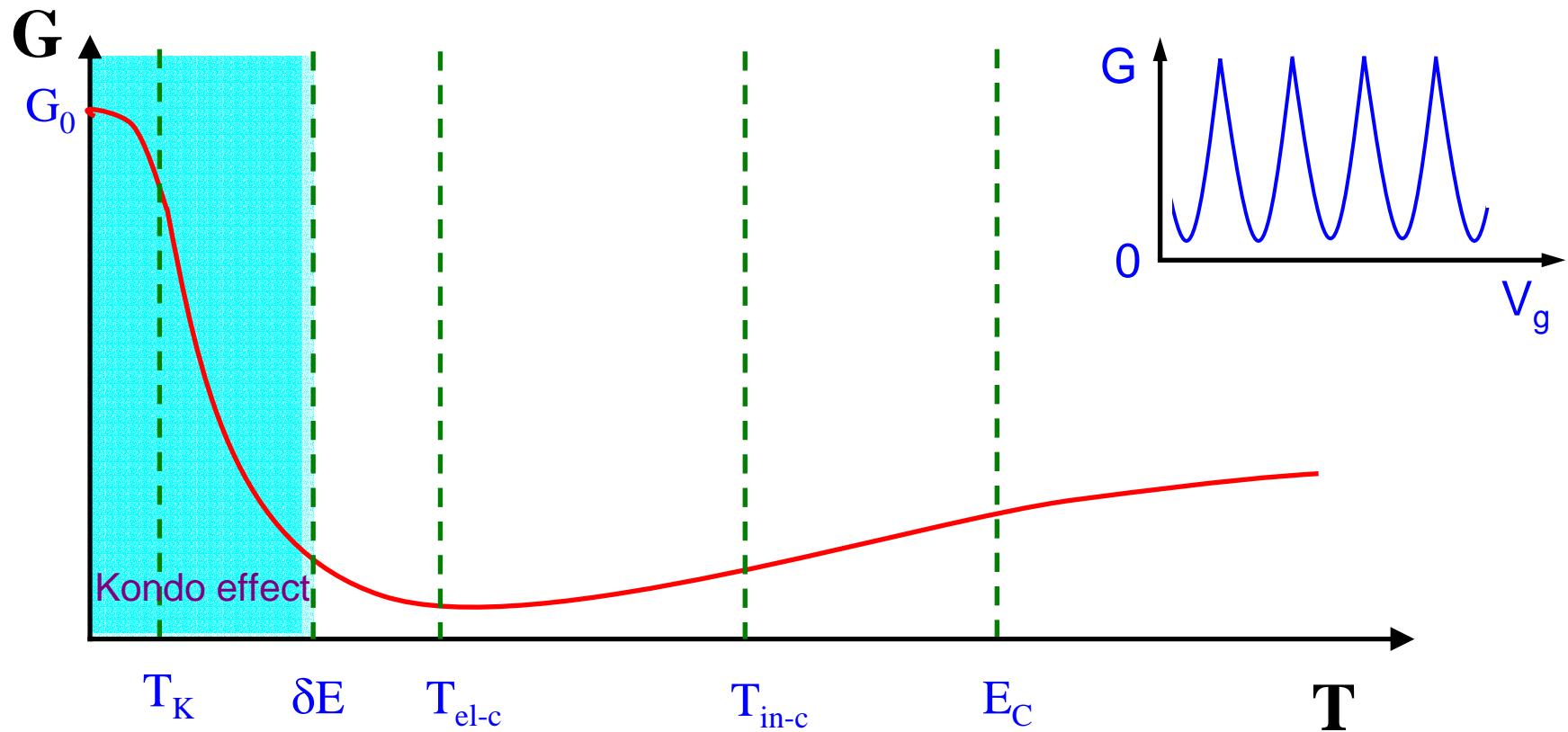
Non spin-flip



Spin-flip

Transmission amplitude diverges logarithmically when the energy of an incoming electron approaches 0.

# T dependence of the conductance in the Coulomb blockade valley with $N_d = \text{odd}$



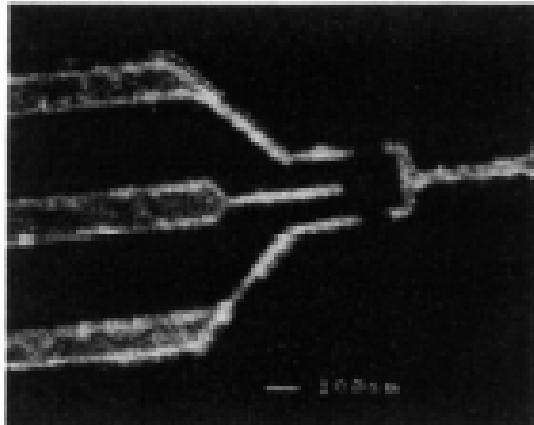
$$G = G_0 [1 - (\pi T / T)^2] \quad \text{where} \quad T \ll T_K$$

$$G = G_0 \frac{3\pi^2 / 16}{[\ln(T / T_K)]^2} \quad \text{where} \quad T_K \ll T \ll \delta E$$

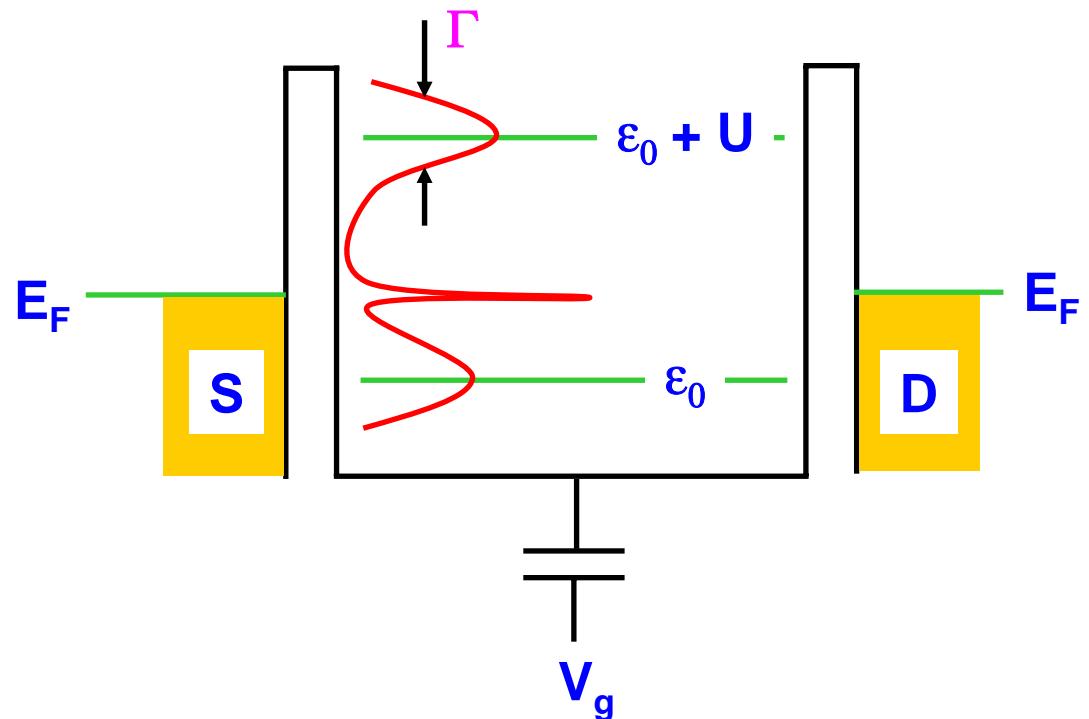
# Single electron transistor (SET)

SET is approximated as a single localized state (Anderson model).  
Electron tunnels into the leads with rate  $\Gamma/h$ .

Kondo temperature :  $k T_K \approx 4 - 250 \mu eV$  ( $T_K \approx 0.05 - 3 K$ )



Goldhaber-Gordon et al., PRL 81,  
5225 (1998)



where

$\epsilon_0$  : bare-level energy

$U$  : Coulomb interaction energy

# Advantages of quantum dot in 2DEG

1. Tunneling rate of leads ( $\Gamma$ ) is controlled by the point-contact Voltage.

$\varepsilon_0 / \Gamma \ll -0.5$  : Kondo regime

$-0.5 < \varepsilon_0 / \Gamma < 0$  : mixed-valence regime

$\varepsilon_0 / \Gamma > 0$  : empty orbital regime

2.  $\varepsilon_0$  of quantum dot is controlled by  $V_g$

$$\varepsilon_0 = \alpha e V_g + \text{const.}$$

$\alpha$  : coupling constant to relate  $V_g$  and  $\varepsilon_0$ .

3.  $\Gamma \propto \alpha$

.

## Constant interaction model

Charging energy  $U$  is assumed to be constant, independent of the number of electrons in quantum dot.

Conductance of resonant tunneling at  $T = 0$

$$G_{T=0} = 2 \frac{e^2}{h} \frac{\Gamma_L \Gamma_R}{(E - E_0)^2 + \Gamma^2} \quad \text{where } \Gamma = (\Gamma_L + \Gamma_R) / 2$$

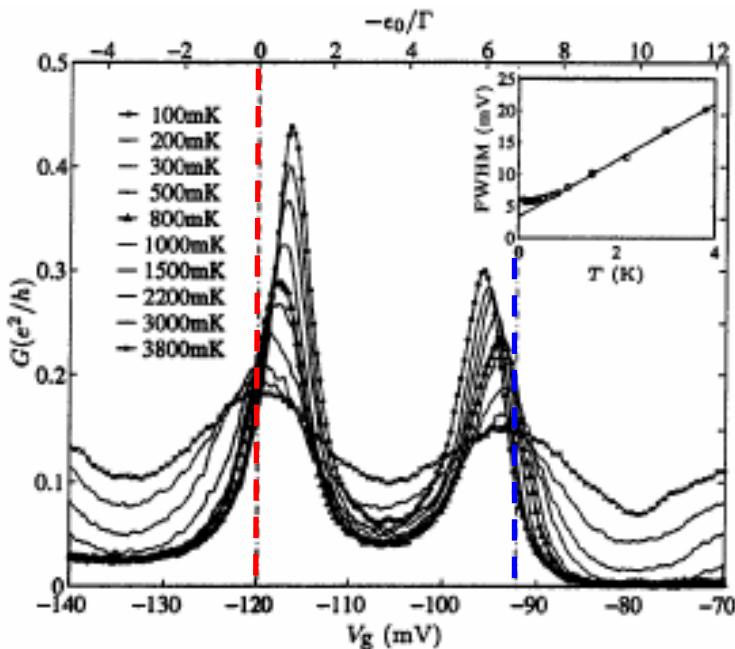
at finite  $T$

$$\begin{aligned} G &= 2 \frac{e^2}{h} \frac{\Gamma_L \Gamma_R}{4\Gamma^2} \int dE \left\{ \frac{\Gamma_L \Gamma_R}{(E - E_0)^2 + \Gamma^2} \right\} \left( -\frac{df}{dE} \right) \\ &= 2 \frac{e^2}{h} \frac{\Gamma_L \Gamma_R}{\Gamma^2} \int \frac{dE}{kT} \left\{ \frac{\Gamma_L \Gamma_R}{(E - E_0)^2 + \Gamma^2} \right\} \operatorname{sech}^2 \left( \frac{E - \alpha e V_g}{2kT} \right) \end{aligned}$$

FWHM of  $G$  is given by  $\Delta = \frac{0.78\Gamma + 3.52kT}{\alpha e}$

# Obtaining $\alpha$ and $\Gamma$ from experiment

Goldhaber-Gordon et al., PRL 81,  
5225 (1998)



$$n_d = 0 \quad 1 \quad 2$$

## FWHM of the left peak

$$\Delta = \frac{0.78\Gamma + 3.52kT}{\alpha e}$$

Fitting       $\alpha$        $\Gamma$

$$\alpha = 0.069 ; \Gamma = 295 \mu\text{eV}$$

Coulomb interaction energy

$$U = \alpha e \Delta V_g = 1.9 \text{ meV}$$

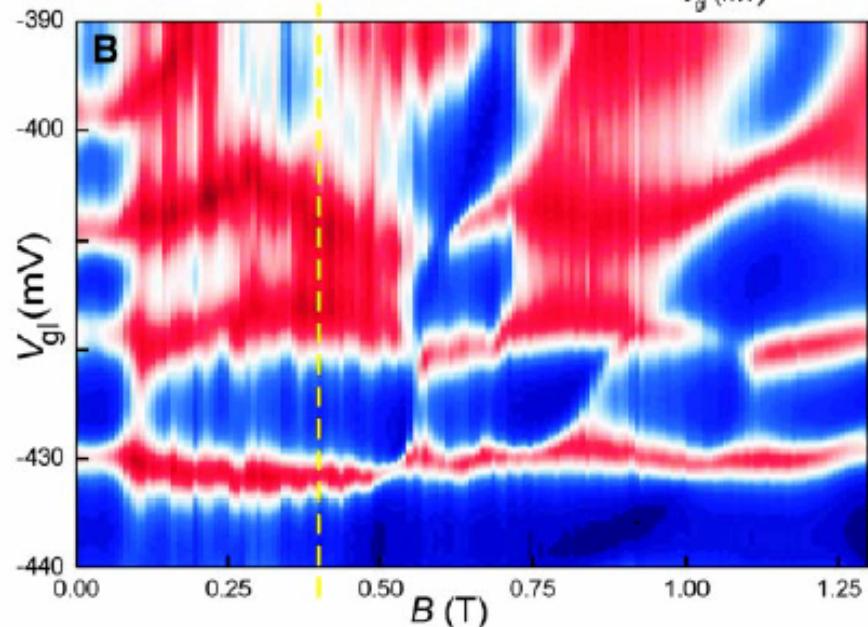
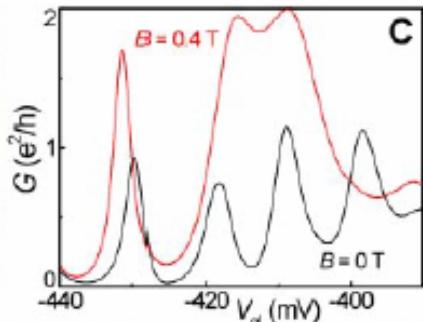
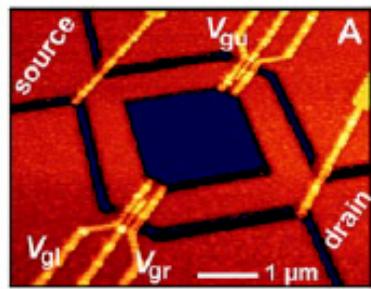
Recall

$$\epsilon_0 = \alpha e V_g + \text{const.}$$

# Kondo effect in a single electron transistor

van der Wiel et al., Science 289, 2105 (2000)

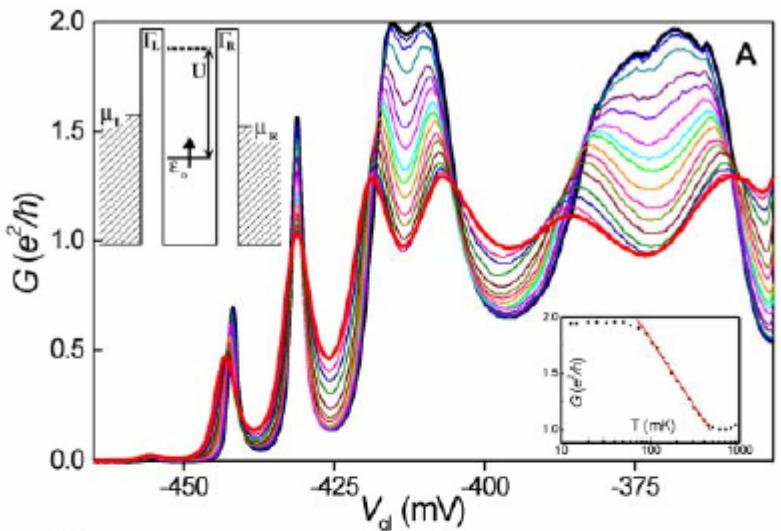
T=15 mK



- Upper gate ( $V_{gu}$ ) pinches off the upper arm.
- Regular Coulomb blockade oscillations at  $B=0$ .
- Small  $B$  brings about a different transport regime (stronger Kondo effect).

# Unitary limit of the Kondo effect in SET

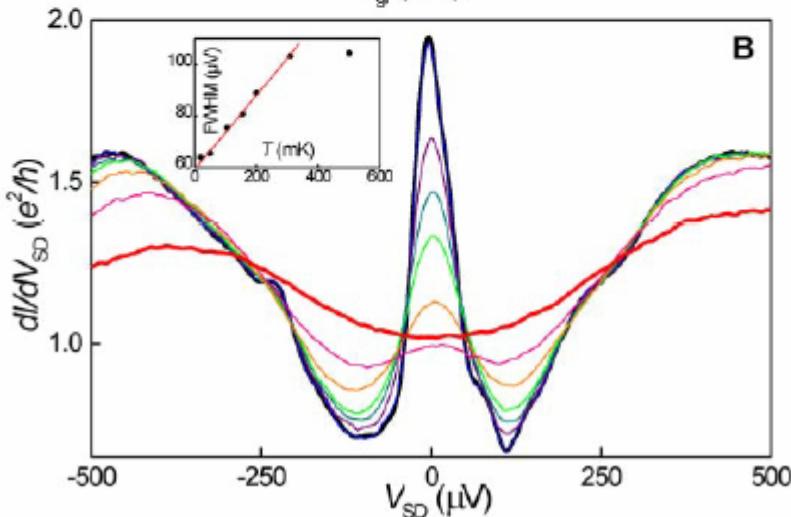
: 15 mK – 800 mK, B= 0.4T



**G(T)**

$G$  at  $V_{gl} = -413$  mV shows logarithmic T dependence (inset), and saturates below 90 mK (unitary limit)

This experiment shows a unitary limit  
 $= 2e^2/h$  ( $\Gamma_R = \Gamma_L$  )



**Kondo resonance peak**

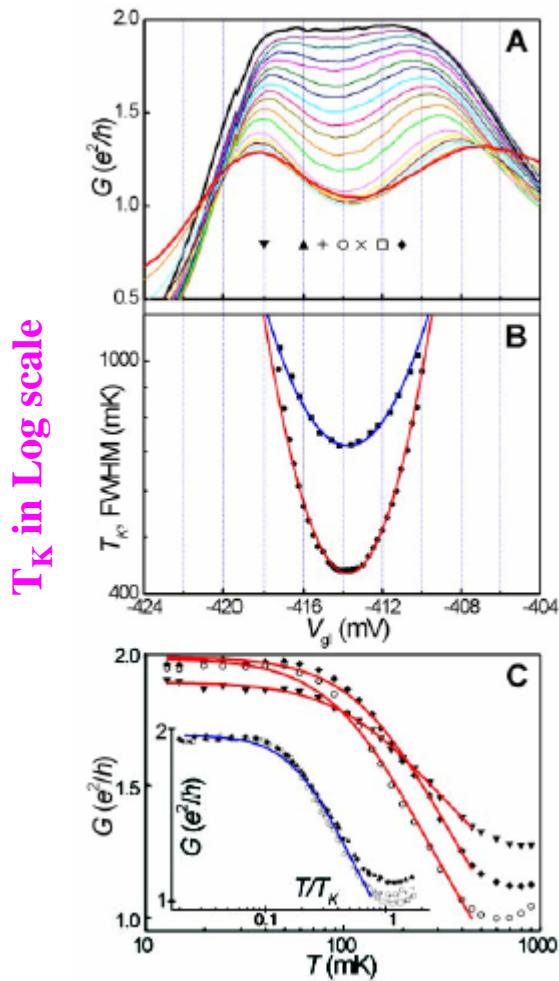
$V_{gl}$  was fixed at -413 mV.  
 $V_{SD}$  was biased between S and D.

# Kondo temperature: $T_K$

In Anderson model,

; Costi et al., J. Phys.: Condense. Matter **6**, 2519 (1994)

$$kT_K = \frac{\sqrt{\Gamma U}}{2} \exp\left[\frac{\pi\varepsilon_0(\varepsilon_0 + U)}{\Gamma U}\right]$$



An empirical function

; Goldhaber-Gordon et al., PRL **81**, 5225 (1998)

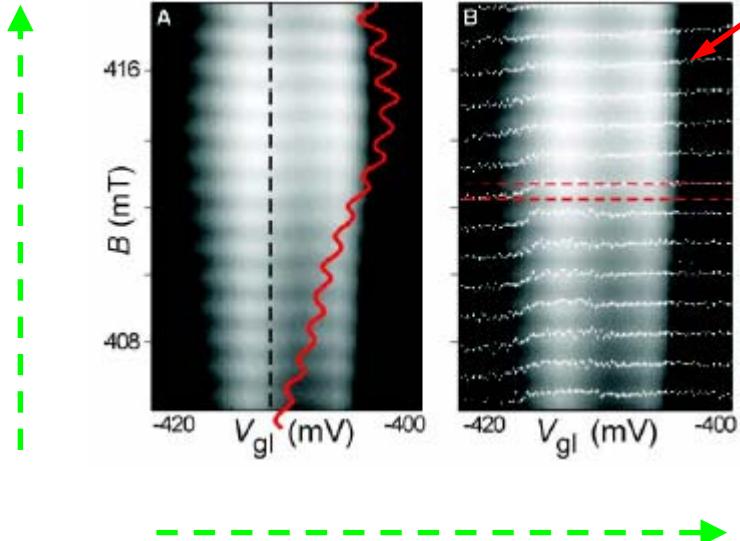
$$G(T) = G_0 \left( \frac{1}{1 + (2^{1/S} - 1) T^2 / T_K^2} \right)^S$$

: universal functional form of  $T/T_K$

- $T_K$  is obtained from fitting.
- $S$  is a fit parameter, but is almost constant ( $\sim 0.2$ ) in the Kondo regime.

# AB conductance oscillation and Coulomb blockade oscillation

Aharonov-Bohm oscillation



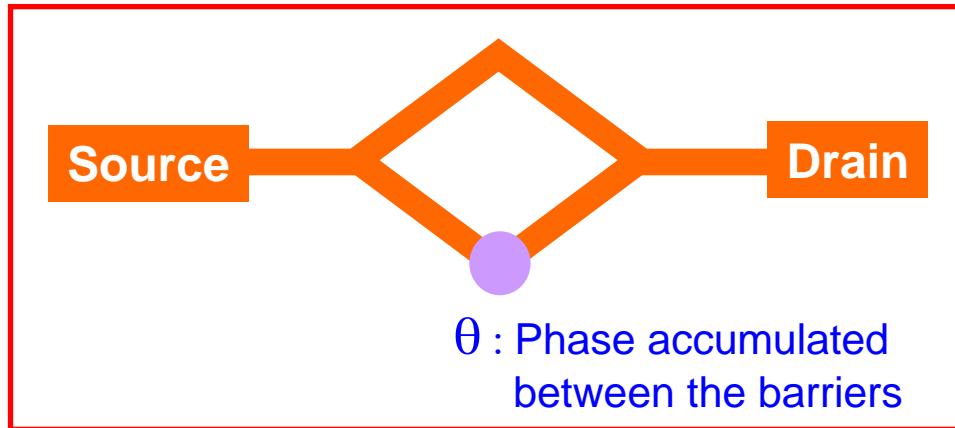
conductance maximum

$\pi$ -phase flip is observed when stepping through the left Coulomb peak

2-terminal geometry causes rapid phase change by  $\pi$ .

; Yacoby et al., PRB 53, 9583 (1996)

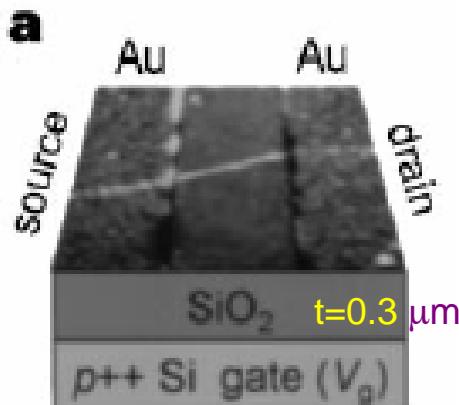
Coulomb blockade oscillation



No phase change is observed in right Coulomb peak.

Why? Yet to be understood.

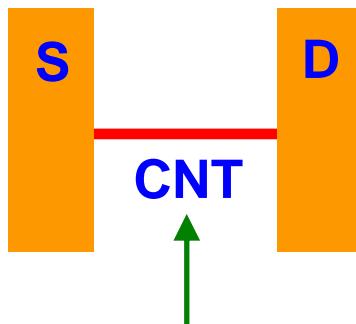
# Kondo effects in carbon nanotubes



Nygard et al., Nature 408, 342 (2000)

## Carbon Nanotube

L=300 nm, 2 nm dia.  
metallic single-walled carbon nanotube



1-D quantum dots  
(Kondo impurity)

Au contact	CNT	transmission probability P
------------	-----	----------------------------

P << 1 : Coulomb blockade

P ~ 0.9 : diffusive 1-D wire

intermediate P : Kondo effect

Kondo resonances for many quantum dots

The number of quantum dots is about number of carbon atoms : ~

# Various structures of carbon nanotube

<http://home.hanyang.ac.kr/~nanotube>

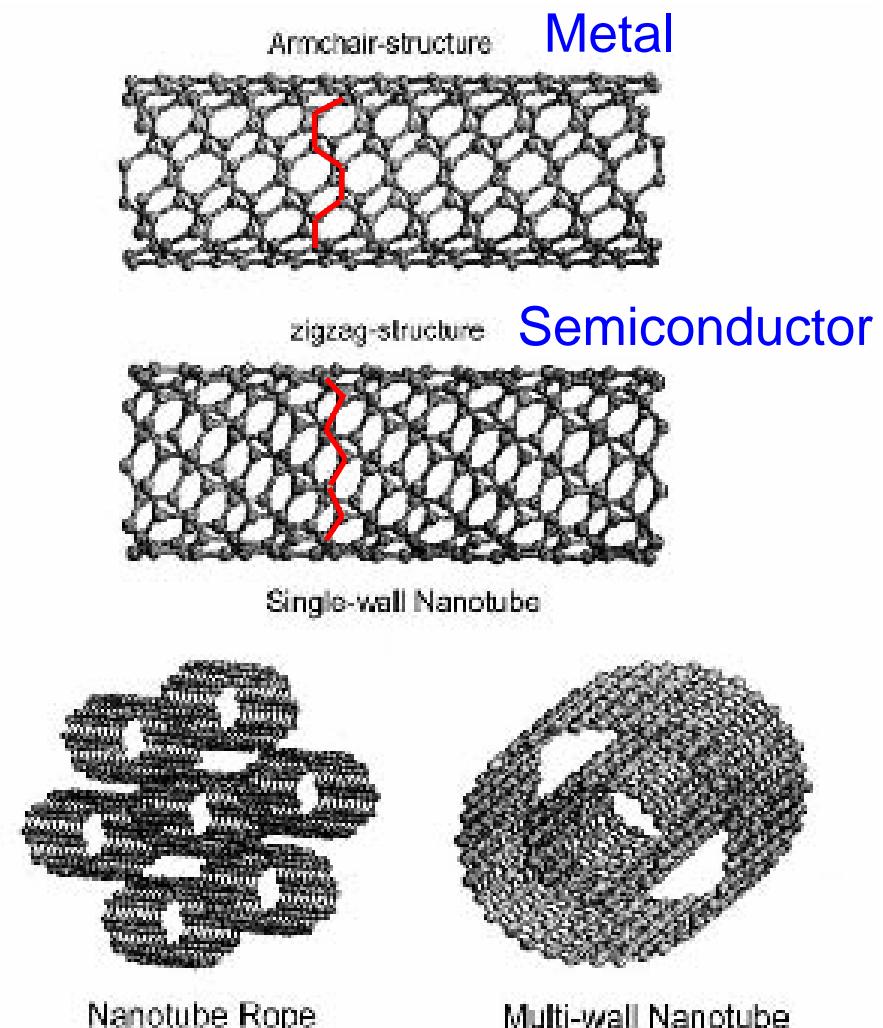
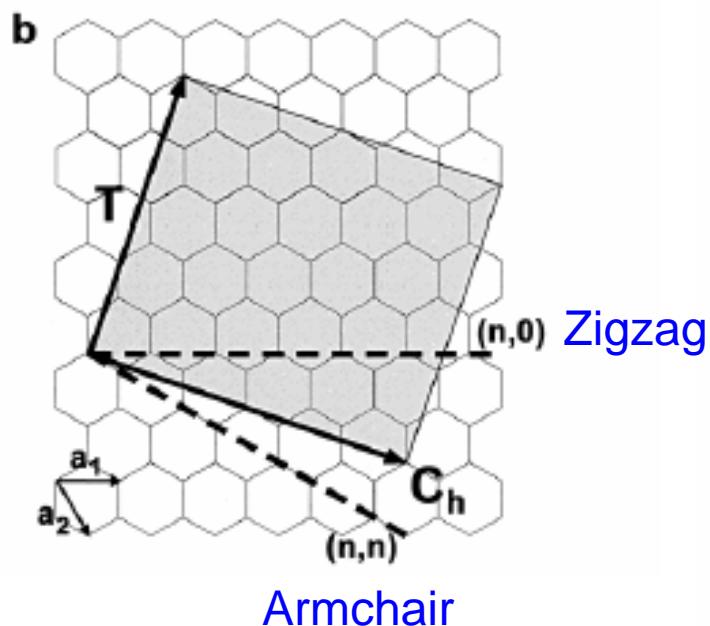
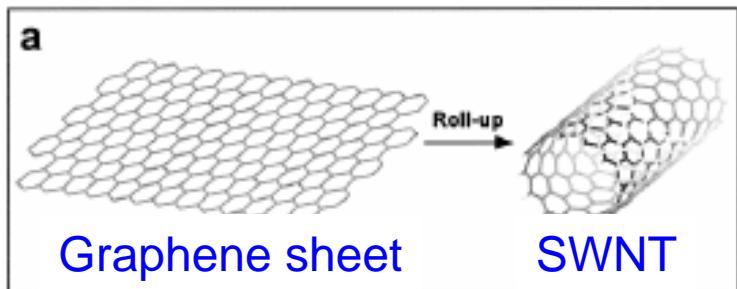
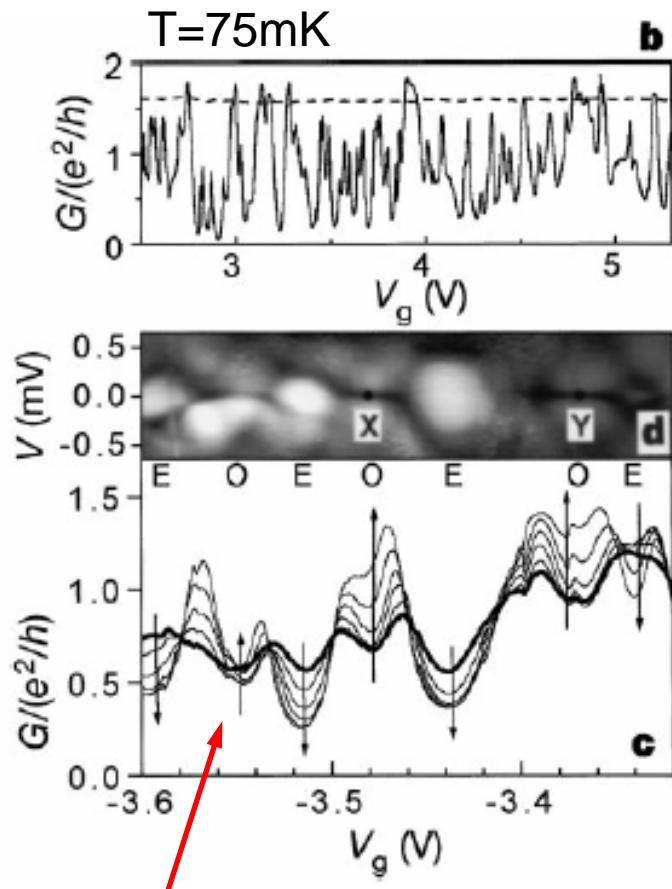


Fig. 1. Various structures of carbon nanotubes.

# Temperature dependence of conductance G

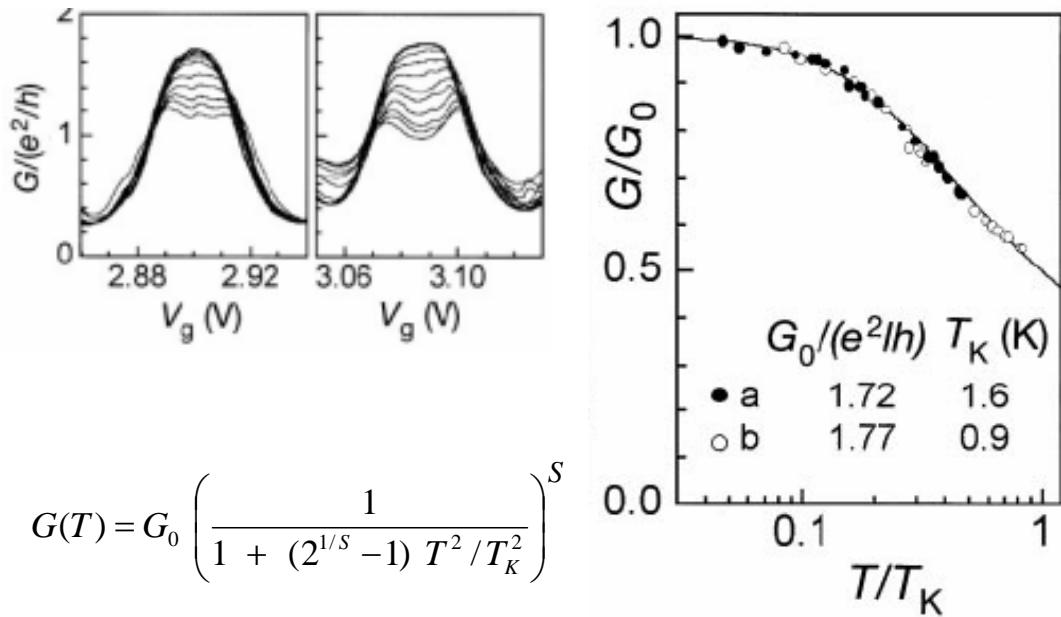


( $T = 75, 125, 180, 245, 320, 490, 560, 780 \text{ mK}$ )

→  $G$  fluctuates as  $V_g$  is swept.

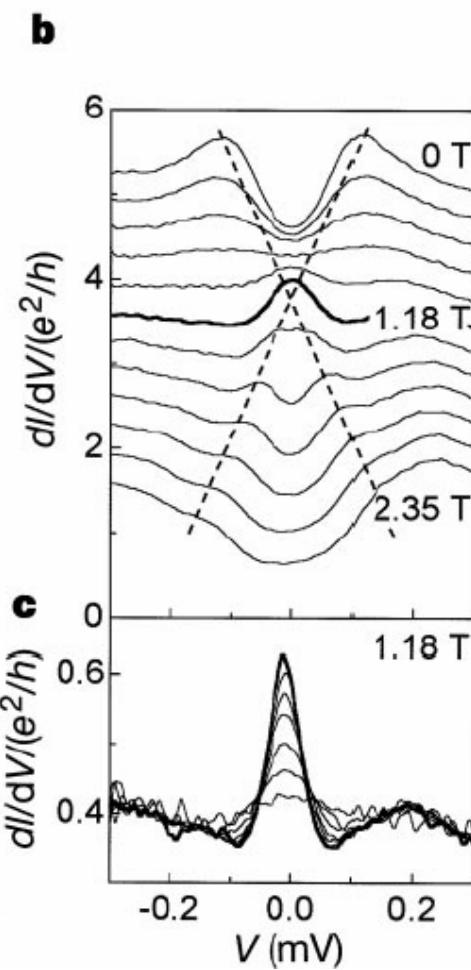
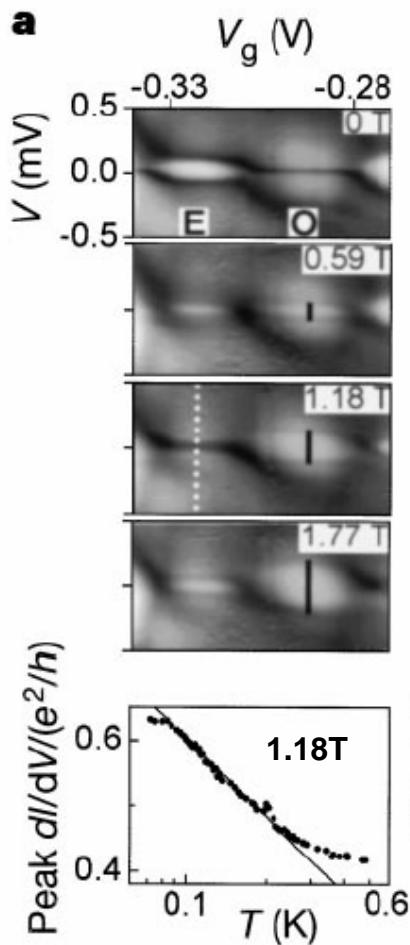
(a darker region represents a higher  $G$ )

LogT dependence of Kondo resonance



$T_K, s$  : fitting parameters

# A generic Kondo effect for even-N quantum dot



Real Kondo resonance (O)

At  $V_g = -0.295V$

=> Peak splits by  $2g\mu_B B/e$ .

Generic Kondo resonance

At  $V_g = -0.322V$

Kondo-like effect at  $B = 1.18T$

The dashed line shows  $g\mu_B B/e$  slope.

=> Peak splitting follows Zeeman splitting

Log T dependence of the  $dI/dV$  peak

Kondo-like resonance

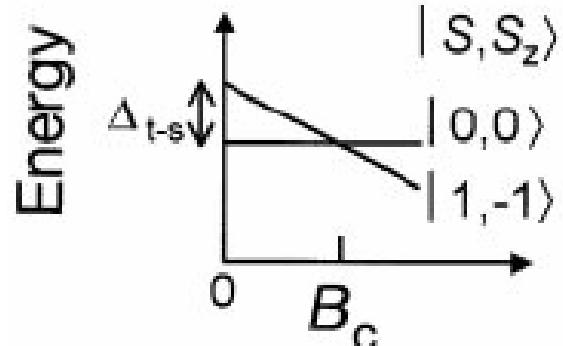
# Spin-flipping higher order transitions in a magnetic field

At  $B=0$ , ground state is a singlet( $S=0$ ).

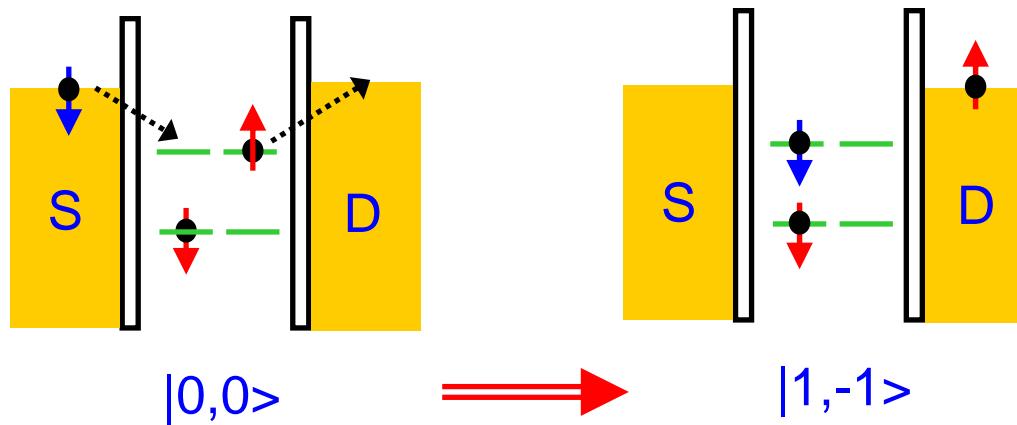
Pustilnik et al., PRL 84, 1756 (2000)

A triplet( $S=1$ ) state is  $\Delta_{t-s}$  above the ground state.

At a certain  $B$ , singlet  $|0,0\rangle$  triplet  $|1,-1\rangle$  are in degeneracy.



## Singlet-triplet alternation



## Magnetic field

Spin-flipping occurs as electrons co-tunnel, leading a generic Kondo resonance.

# Co cluster on a single-walled Carbon Nanotube

Odom et al., Science 290, 1549 (2000)

Co was thermally evaporated onto single walled-CNT.

STM spectroscopic measurements at T= 5K

Apply V between STM tip and single walled-CNT, and measure I or  $dI/dV$ .

=> Spectral density of energy state

Kondo resonance was observed.

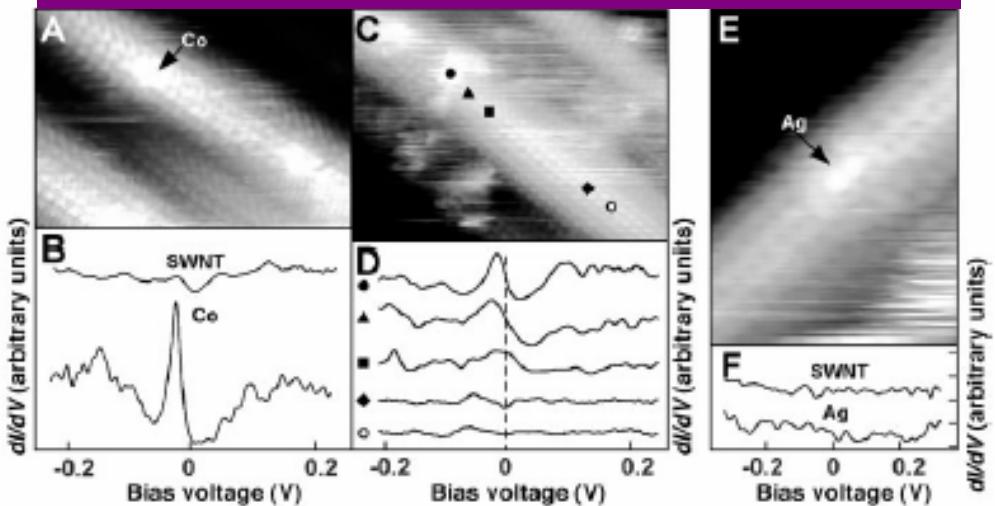
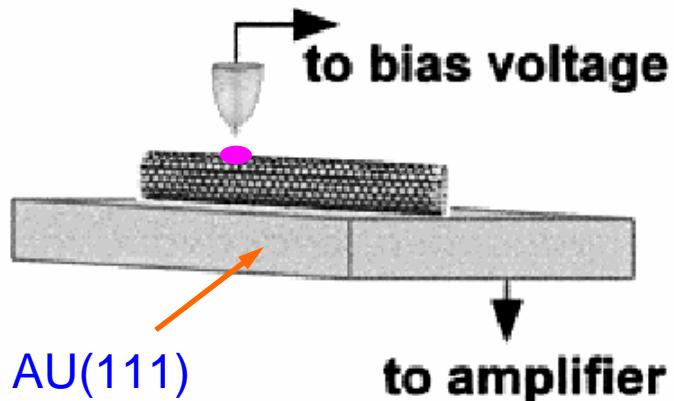


Fig. 1. STM topographic images and spectroscopic measurements on small clusters situated on SWNTs. (A) Atomically resolved image of 0.5-nm Co clusters on an individual nanotube. (B)



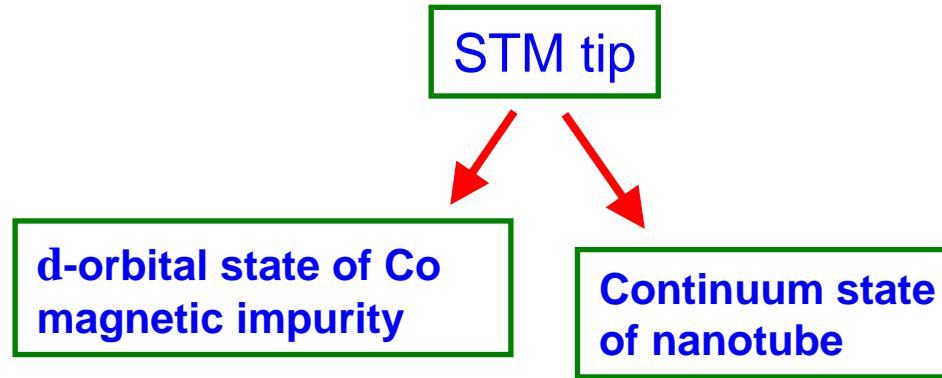
## Fabrication

- Spin coating nanotube suspensions of dichloroethane
- Co was thermally evaporated onto SWNT in situ at low temperature

# Fano model

Fano, Phys. Rev. 124, 1866 (1961)

Two possible channels



Fano's model (the  $U=0$  case of the Anderson model) describes this situation.

# Asymmetric peak shape

## Transition rate in Fano model

$$R(\varepsilon) = R_0(\varepsilon) \frac{(q + \varepsilon')^2}{1 + \varepsilon'^2} \quad \text{where} \quad \varepsilon' = \frac{\varepsilon - \varepsilon_0}{kT_K}$$

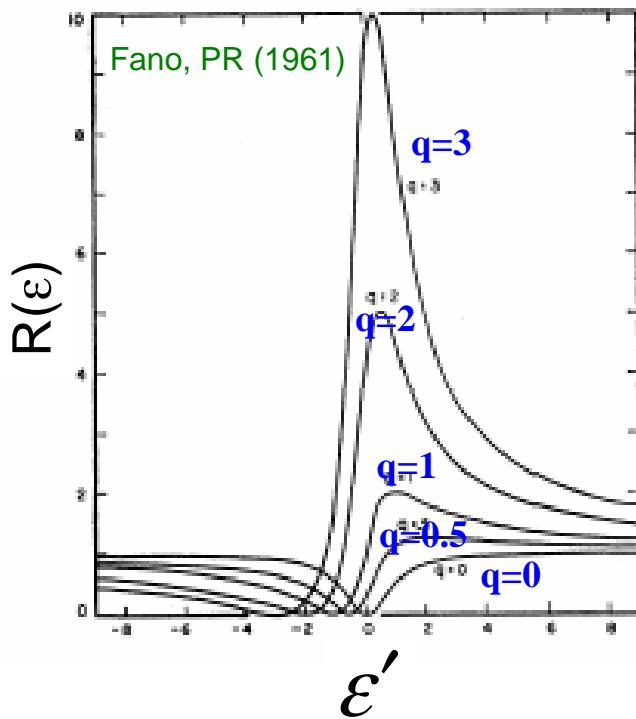
$\varepsilon_0$  resonance energy

discrete state (d-orbital state)

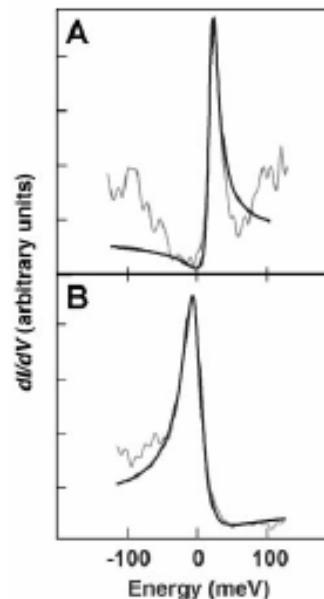
$q$  (“interference parameter”)  $\propto \frac{\text{transition amplitude to } (d\text{-orbital})}{\text{transition amplitude to } (\text{continuum state})}$

,  $R_0(\varepsilon)$  transition rate in the absence of

.



dl/dV spectrum of Co on SWNT



Co size  $\sim 0.5$  nm

$q = 2.7$ ,  $T_K = 93$ K

Co size  $\sim 0.7$  nm

$q = -3.3$ ,  $T_K = 185$ K

In AuCo bulk,  $T_K \sim 300$  to 700 K

# Co cluster on a Au(111) surface

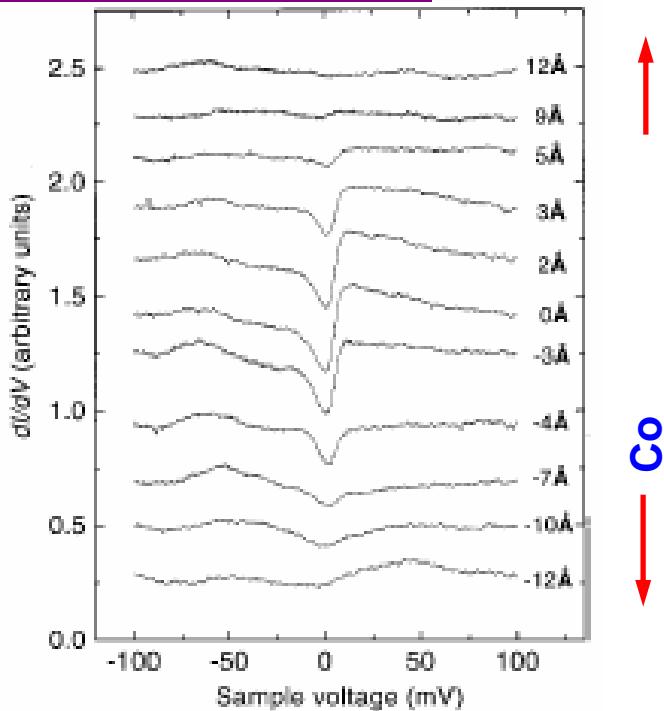
Madhavan et al., Science 280, 567 (1998)

Co was thermally evaporated onto Au(111) surface.

STM spectroscopic measurements at T= 4K.

Kondo resonance was observed, and the shape of the Kondo peak can be explained by Fano's formalism.

## STM spectroscopy



A similar situation as Co on SWNT  
except for the continuum state  
: Au single crystal

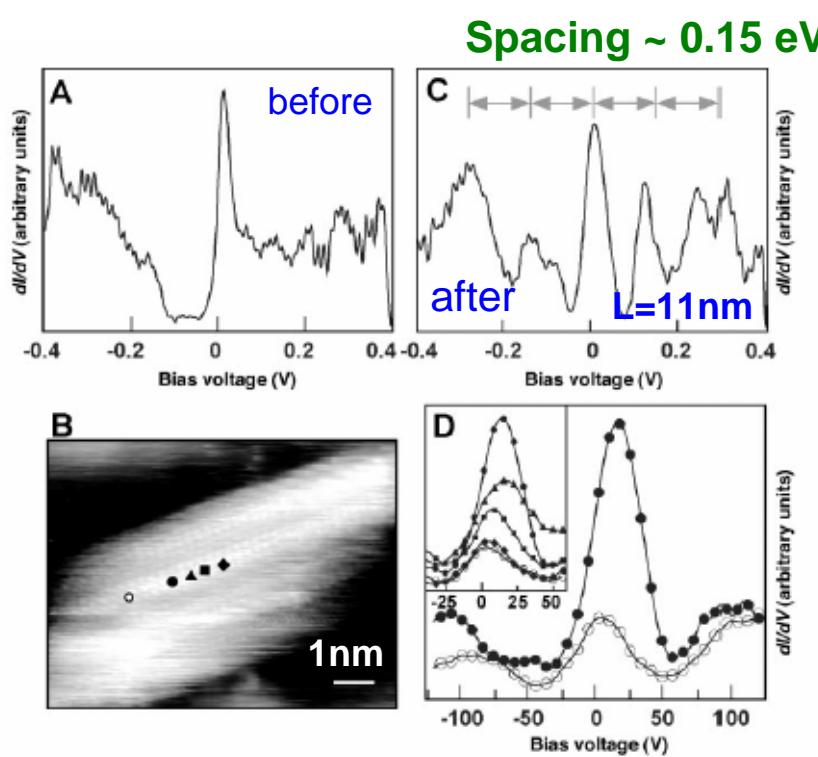
$$q = 0.7, T_K = 64\text{K}$$

In general,  $|q|$  is smaller than that of  
Co on SWNT.

# Co cluster on a SWNT quantum box

Odom et al., Science 290, 1549 (2000)

Using a voltage pulse (7V and 100 $\mu$ s),  
SWNT was cut into a segment.



Energy level spacing

$$\delta E = \frac{\hbar^2 k_F}{m} \Delta k$$

For 1D particle in a box potential,  $\Delta k = \pi/L$

$$\delta E = \frac{\hbar v_F}{2L} \approx \frac{1.67}{L \text{ [nm]}} \text{ eV}$$

, where  $v_F$  is the Fermi velocity of graphene.

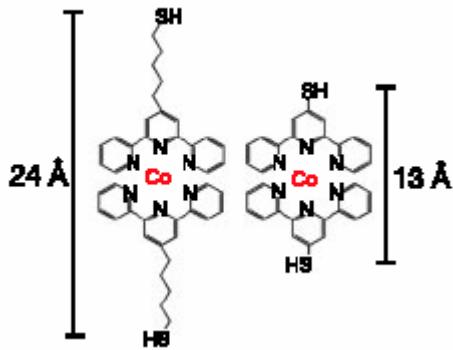
# Kondo effect in a single molecule transistor

Jiwoong Park et al., Nature 417, 722 (2002)

Molecule  $[\text{Co}(\text{tpy}-\text{(CH}_2)_5-\text{SH})_2]^{2+}$  or  $[\text{Co}(\text{tpy-SH})_2]^{2+}$  where tpy: erpyridinyl

=> Co ion + Linker molecule with different lengths

Fabrication

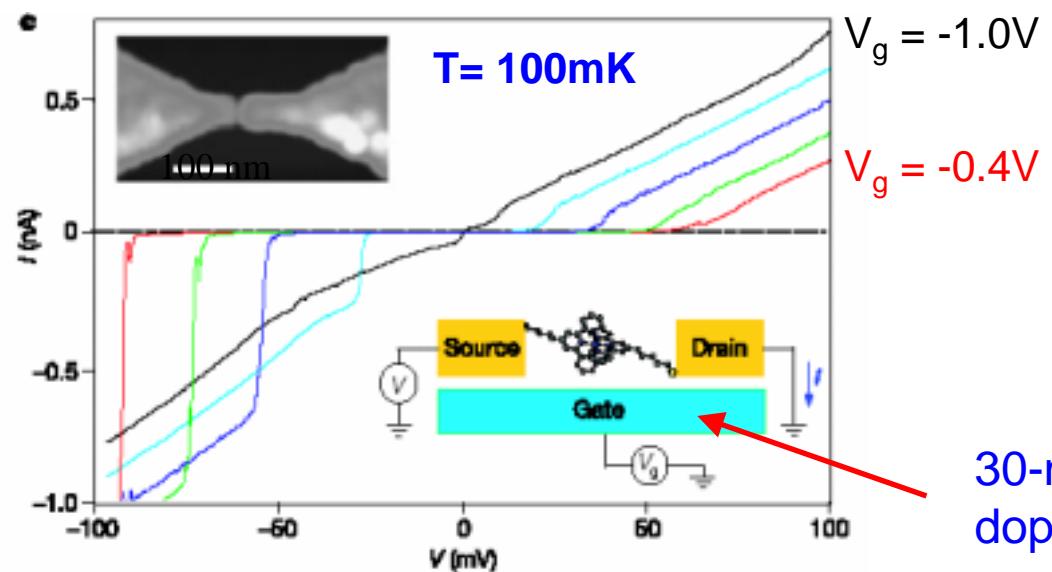


Continuous Au nanowire ( $w= 10-15\text{nm}$ ,  $L= 200-400\text{nm}$ ).

⇒ A self-assembled monolayer on the Au wire.

⇒ Apply dc voltage ( $\sim 0.5\text{V}$ ) to break the Au wire by electromigration.

This produces a gap about 1-2 nm-wide.



I-V reflects the property of a single electron transistor

30-nm  $\text{SiO}_2$  layer on degenerately doped Si Substrate: gate electrode

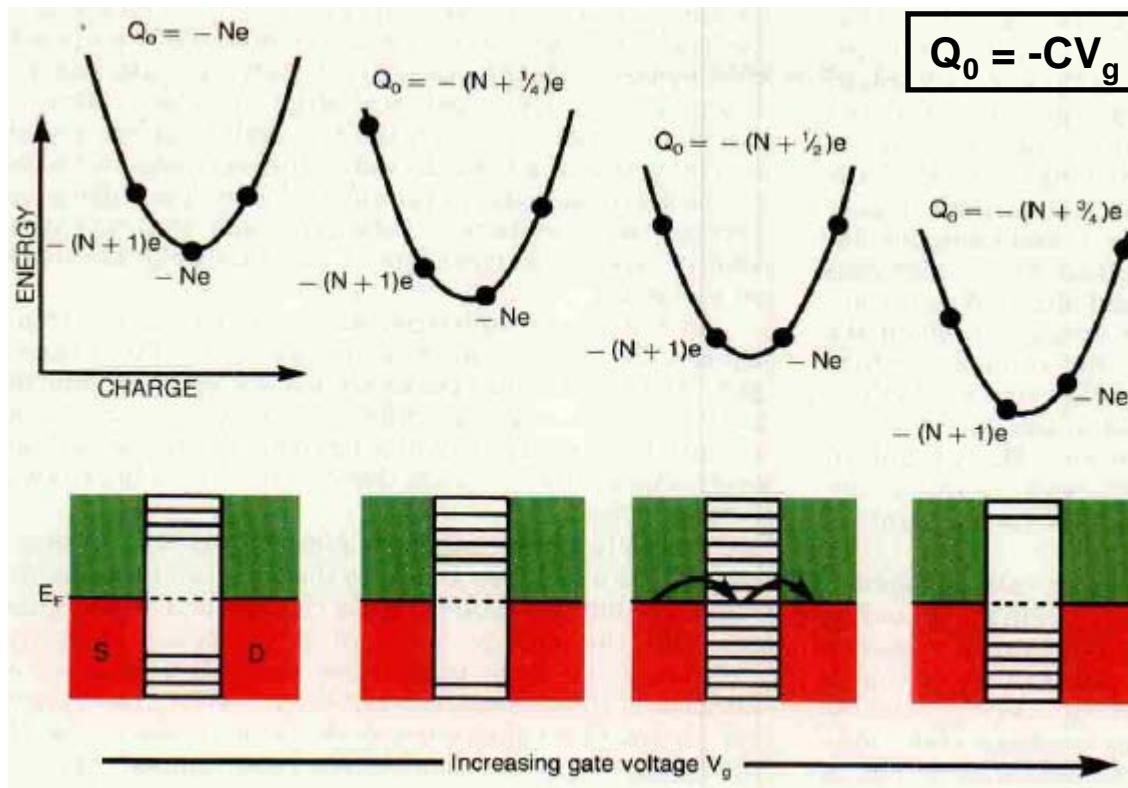
# Recall [ Coulomb blockade oscillation ]

Kastner, Phys. Today 24 (Jan 1993)

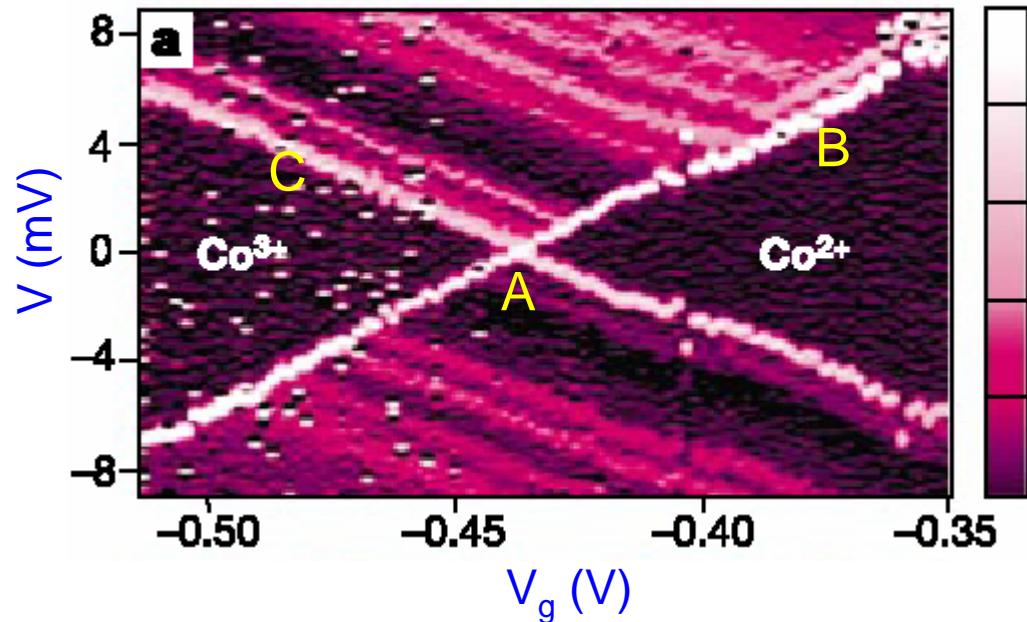
Electrostatic energy of a charge  $Q$  on the dot

$$E = QV_g + \frac{Q^2}{2C} = \frac{1}{2C}(Q + CV_g)^2 - CV_g^2$$

$$Q = \dots, -(N+1)e, -Ne, -(N-1)e, \dots$$



# Differential conductance ( $dI/dV$ ) of single molecule transistor



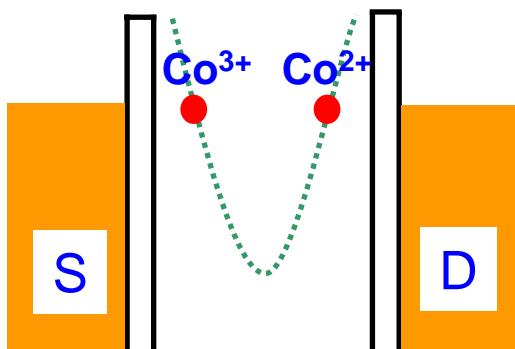
Co ion works as a quantum dot in SET

; No such behavior has been found without Co ion

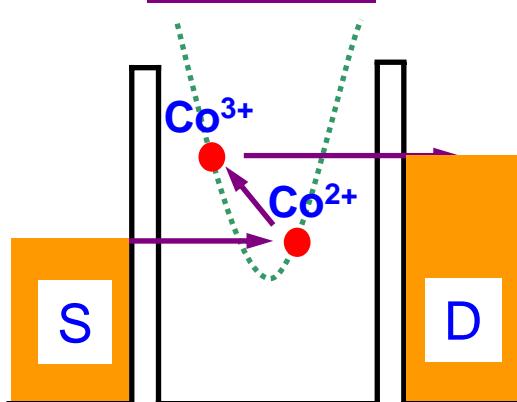
Energy of a quantum dot

$$E = QV_g + \frac{Q^2}{2C} = \frac{1}{2C}(Q + CV_g)^2 - CV_g^2$$

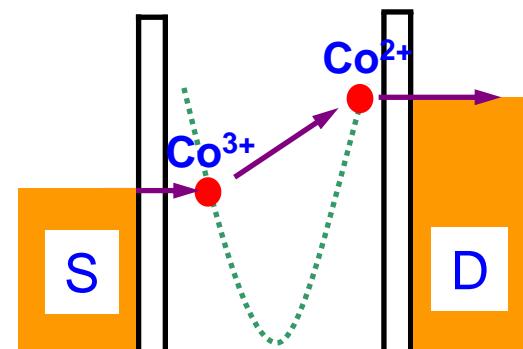
(A)  $V = 0$



(B)  $V > 0$



(C)  $V > 0$



$\text{Co}^{3+}$  and  $\text{Co}^{2+}$  degenerate

$\text{Co}^{2+}$  to  $\text{Co}^{3+}$  transition

$\text{Co}^{3+}$  to  $\text{Co}^{2+}$  transition

# Kondo effect out of equilibrium

Meir et al., PRL 70, 2601 (1993)

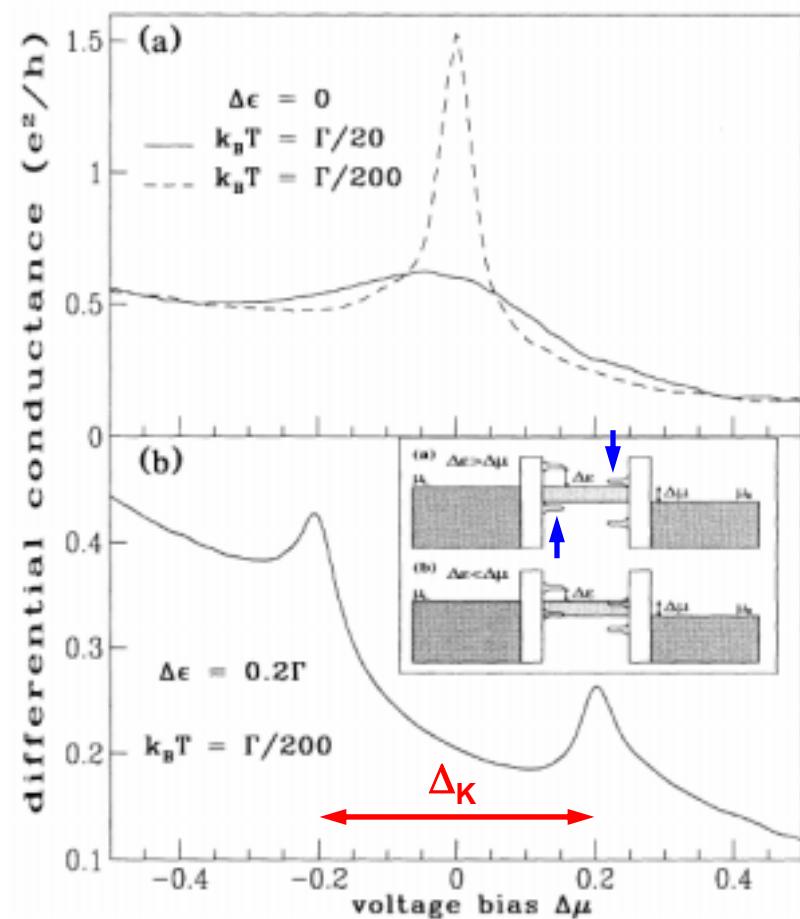
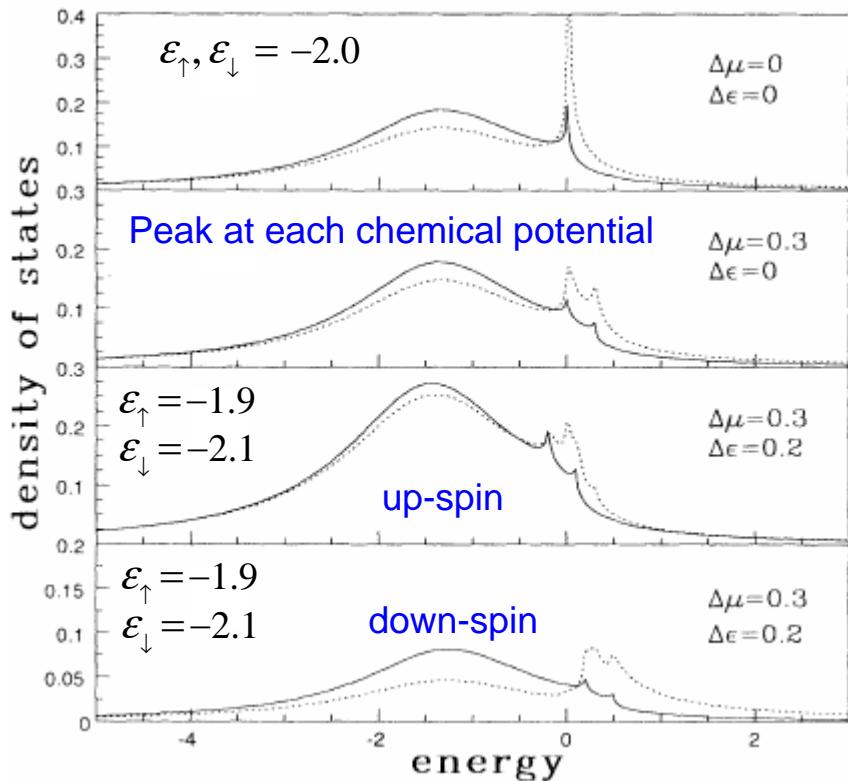
“Kondo effect in a voltage bias” was studied through the Anderson Hamiltonian using perturbation theory, noncrossing approximation, equation of motion, variational wave-function calculation methods.

## Results

1. A voltage bias causes the Kondo peak to split.  
=> A peak in the density of states at the chemical potential of each lead.
2. In a magnetic field, the Kondo peaks shift away from the chemical potentials by Zeeman splitting  $\Delta = g\mu_B B$ .  
=> Kondo peaks shift down for up-spin channel.  
Kondo peaks shift up for down-spin channel.

# Kondo peak splitting in a magnetic field

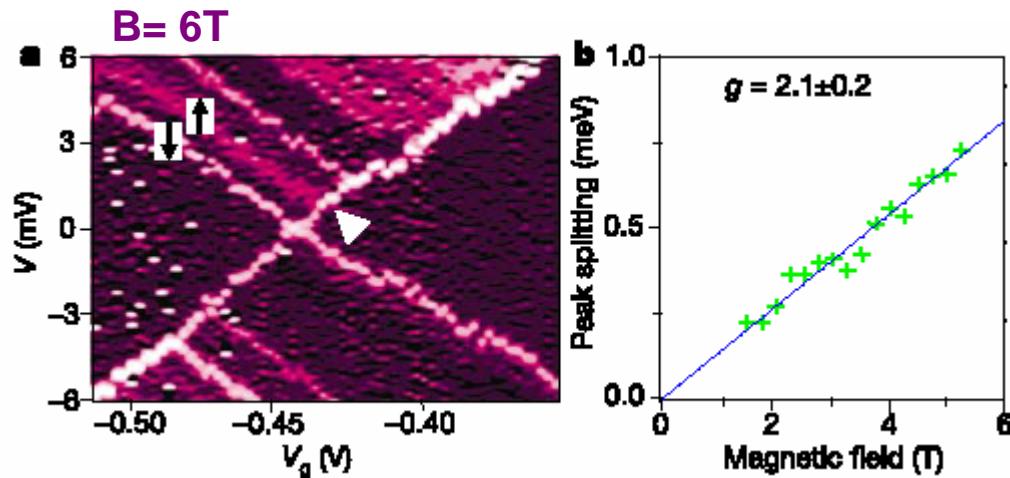
## Density of state of an Anderson impurity



Kondo peak splitting  $\Delta_K$  is twice the ordinary Zeeman splitting.

$$\Delta_K = 2g\mu_B B$$

# Magnetic field dependence of the single molecule transistor



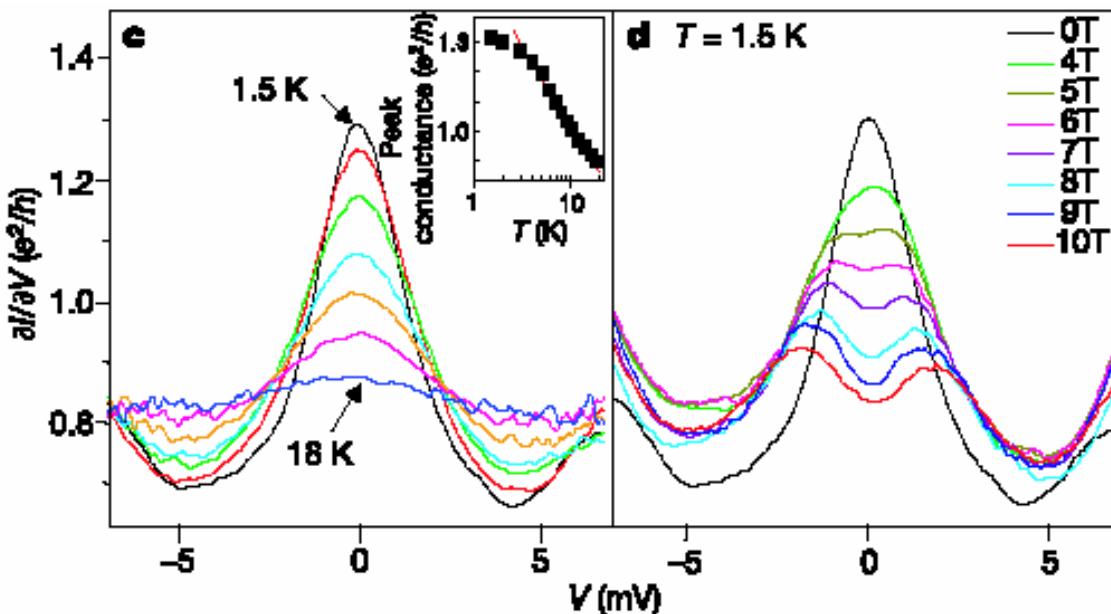
$\text{Co}^{2+}$  ion ( $3d^7$ ) : odd number of e  
 $S = 1/2$

$\text{Co}^{3+}$  ion ( $3d^6$ ) : even number of e  
 $S = 0$

Zeeman splitting

$$\Delta = g \mu_B H$$

Kondo resonance for a SMT of  $[\text{Co}(\text{tpy-SH})_2]^{2+}$



LogT dependence of the Kondo peak

Kondo peak splitting varies linearly with  $H$

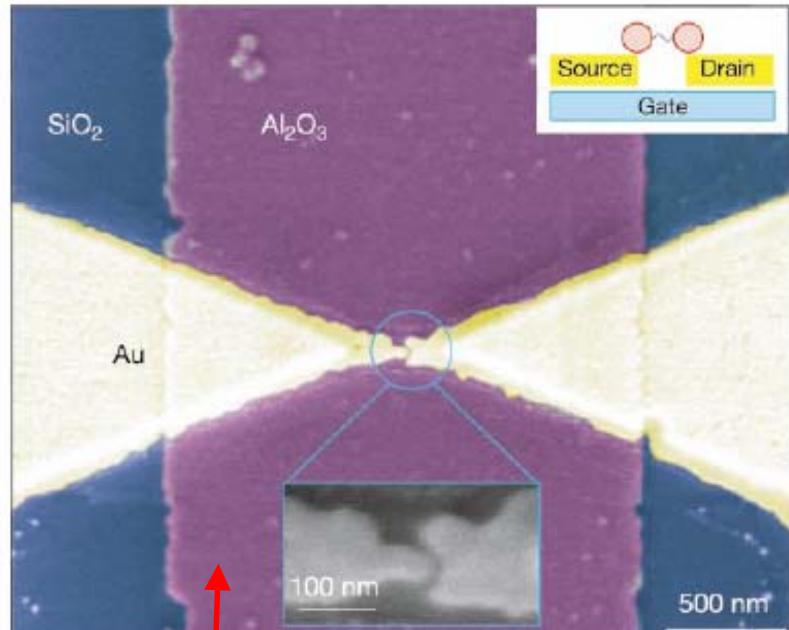
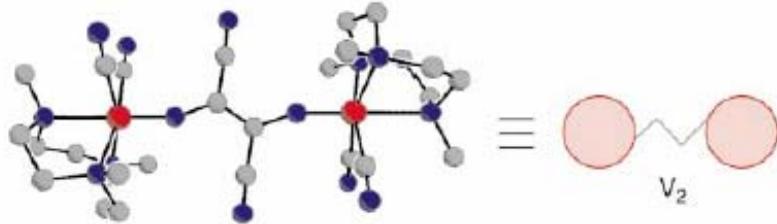
$$\Delta_K = 2g\mu_B H$$

( $g \approx 2$ )

# Single-molecule transistor of an individual V<sub>2</sub> molecule

spin impurity

Liang et al., Nature 417, 725 (2002)

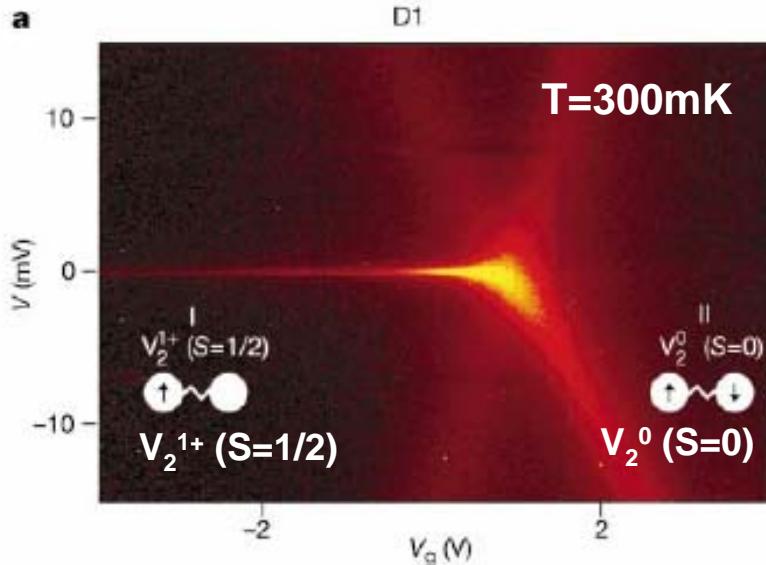


3-nm oxide Al layer: gate electrode

Fabrication —  
Electromigration-induced break-junction technique

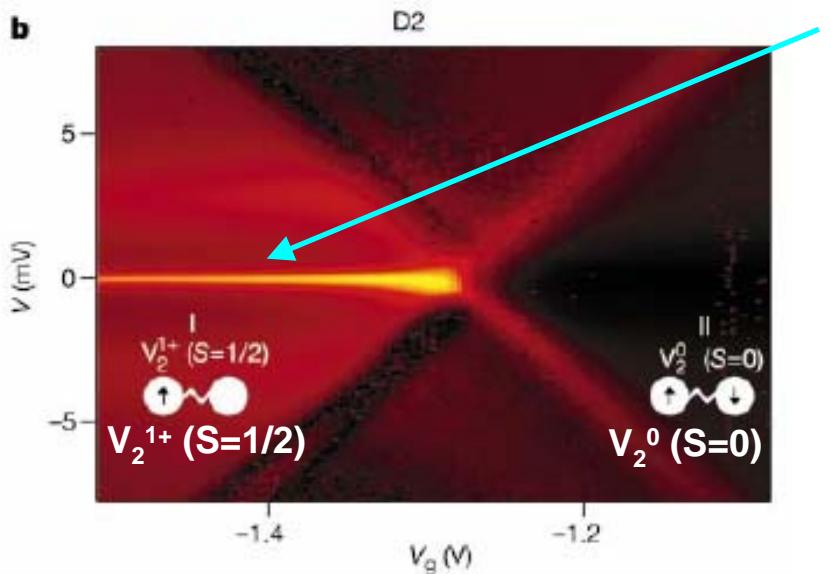
Gap between two electrode is  
~ 1 nm.

# Differential conductance $dI/dV$ of $V_2$ transistor



Scale: Dark red(0) to bright yellow( $1.55\text{e}^2/\text{h}$ )

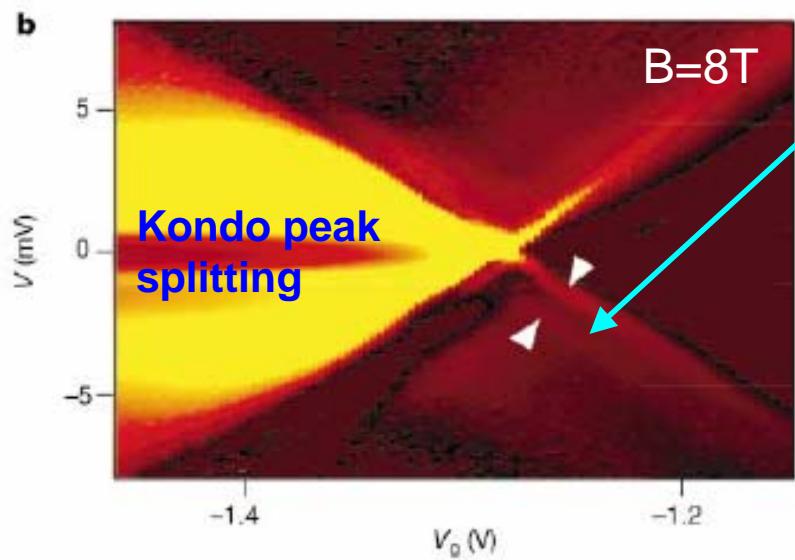
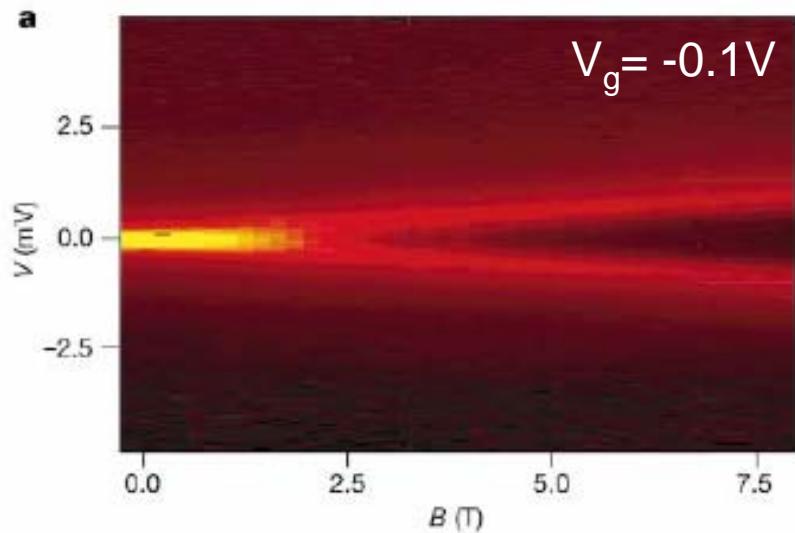
SET characteristic was observed.



Kondo resonance peak at  $V_2^{1+}$  state

Only appears when the molecule has non-zero spin.

# Magnetic field dependence



Splitting of Kondo peak

$$\Delta_K = 2g\mu_B B$$

Twice the ordinary  
Zeeman splitting

$$(\mu_B \approx 58 \text{ } \mu eV / T)$$

From experiment

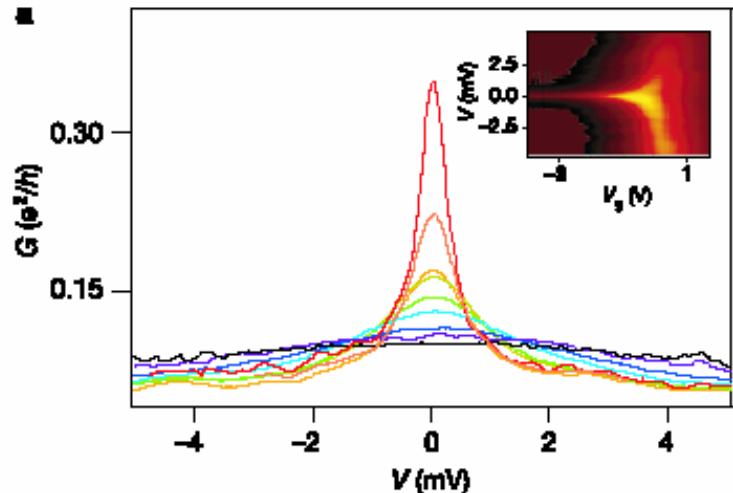
$$\frac{\Delta_K}{eB} = \frac{e}{B} \frac{\delta V_K}{\delta V} \approx 230 \text{ } \mu eV / T$$

Zeeman splitting

From experiment

$$\frac{\Delta}{eB} = \frac{e}{B} \frac{\delta V}{\delta V} \approx 115 \text{ } \mu eV / T$$

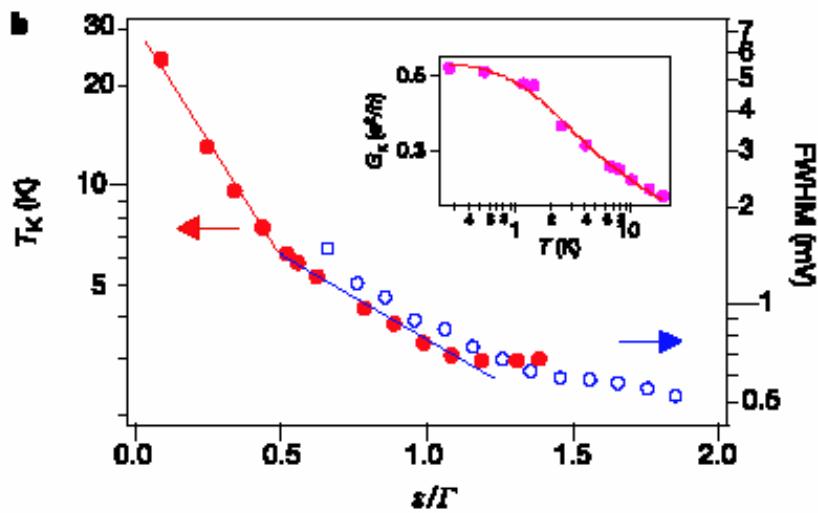
# Temperature dependence of Kondo peak



Kondo peak at  
 $T = 0.3, 1.0, 2.0, 3.1, 4.2, 6.3, 9.0, 14, 20$  K.  
(  $V_g$  was set to -2.25V )

For each  $\epsilon/\Gamma$ , the Kondo peak  $G_K(T)$  were plotted.

Inset :  $G_K(T)$  at  $\epsilon/\Gamma = 0.43$



Fit to the empirical formula

$$G_K(T) = G_0 \left( \frac{1}{1 + (2^{1/s} - 1) T^2 / T_K^2} \right)^s$$

Fitting yields  $T_K = 12.2$  K and  $s = 0.19$

$\epsilon$  : energy of the localized electron ( $\epsilon = \alpha e V_g + \text{const}$ )

$\Gamma$  : level width due to the coupling to the electrodes