Andreev Reflection in Heavy-Fermion Superconductors: Order Parameter Symmetry and Emergence of the Heavy-Electron Liquid in CeCoIn5

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Dozens of undergraduates, NSF, and DoE.

**in memory*

Outline

☐ **Introduction:**

- The heavy-fermion superconductor (HFS) $CeCoIn_5$
- Point Contact Andreev Reflection Tunneling Spectroscopy (PCARTS)
- Blonder-Tinkham-Klapwijk (BTK) theory and it's extension to d-wave
- Definition of the issues (AR at HFSs and spectroscopy of HFs)

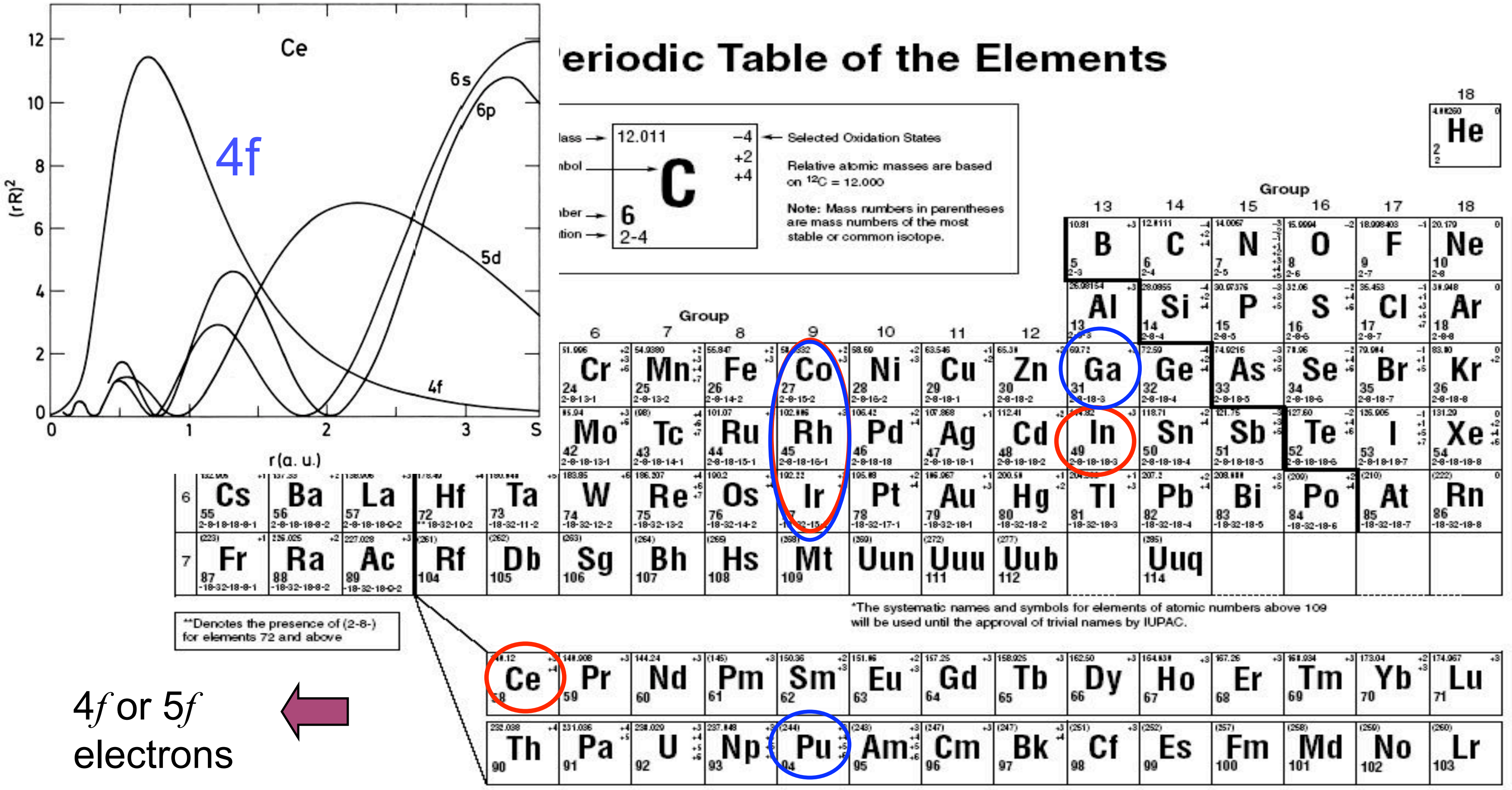
☐ **Results and Discussions:**

- $CeCoIn_5$ along three crystallographic orientations **=>** d_{x2-y2} symmetry (shows spectroscopic ability of PCARTS)
- Modification of the BTK model for heavy fermions: (*two-fluid model, and energy-dependent DoS)*

☐ **Conclusions:**

- PCARTS is a much more powerful technique than we ever expected: OP symmetry, DoS, and AF/S competing orders
- Model (2-fluid + DoS peak) explains data; and now with some understanding.

1-1-5 Heavy-Fermion Compounds



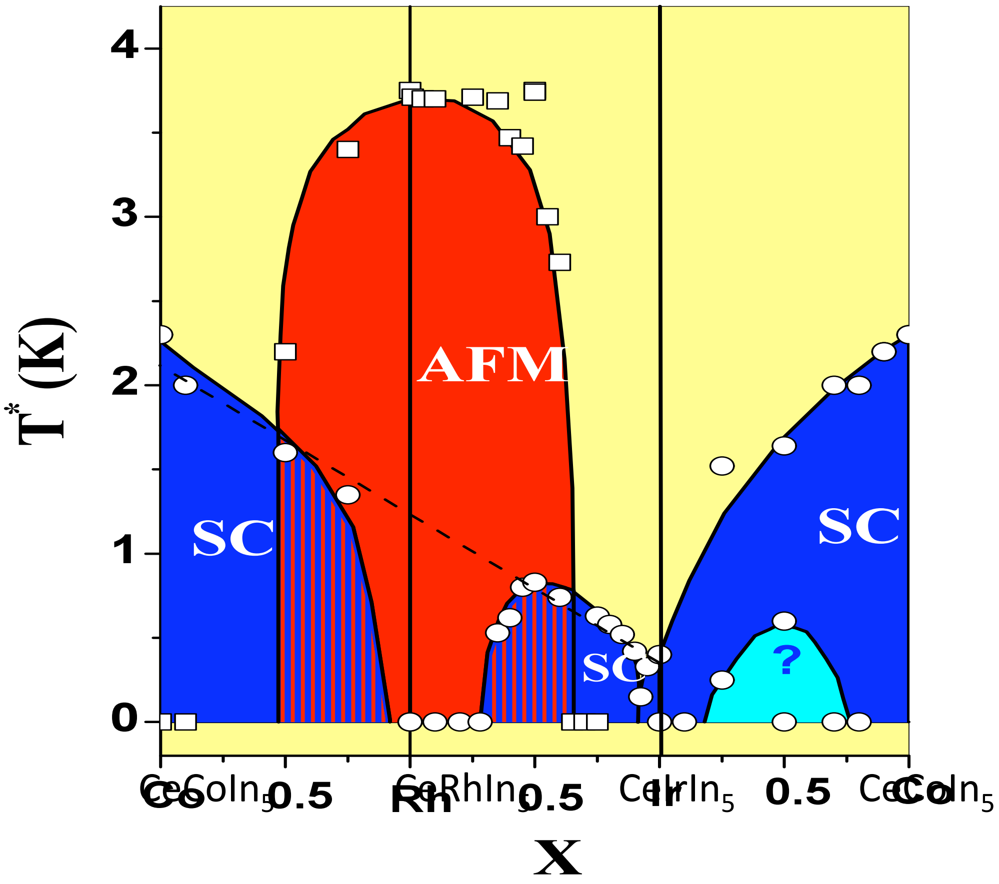
4*f* or 5*f* electrons ⇦

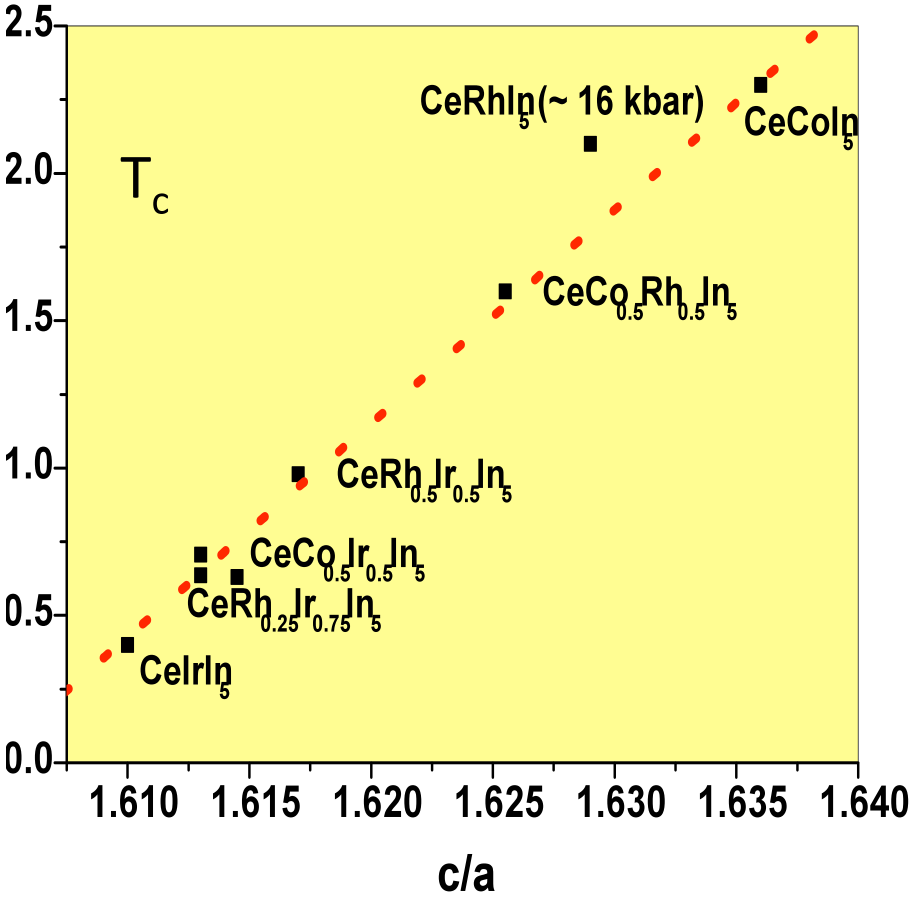
$CeMIn_5$ — $CeCoIn_5$ (T_c=2.3 K, g_{el}=290 mJmol^{-1}K^{-2})

$PuMGa_5$ — $PuCoGa_5$ (T_c=18.5 K, g_{el}=77 mJmol^{-1}K^{-2})

$CeCoIn_5$: "Rosetta Stone for the Kondo Lattice"

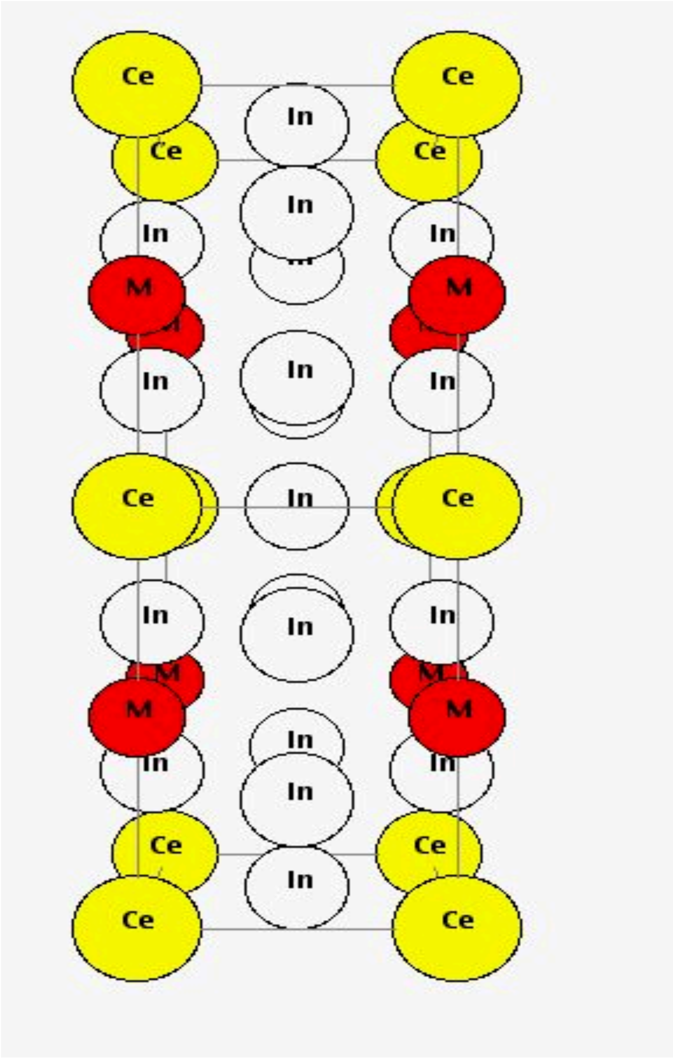
Ce M In_5 (M = Co, Rh, Ir):
Generalized Doping-Temperature Phase Diagram

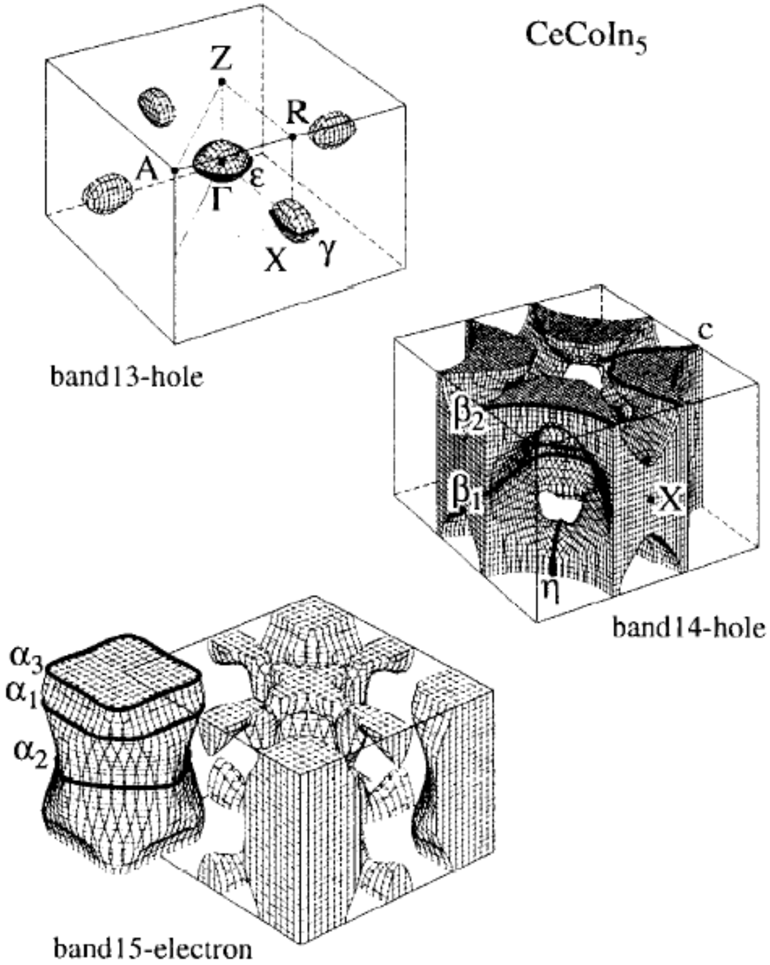




Pagliuso et al., Phys. Rev. B 64 (2001) 100503(R)

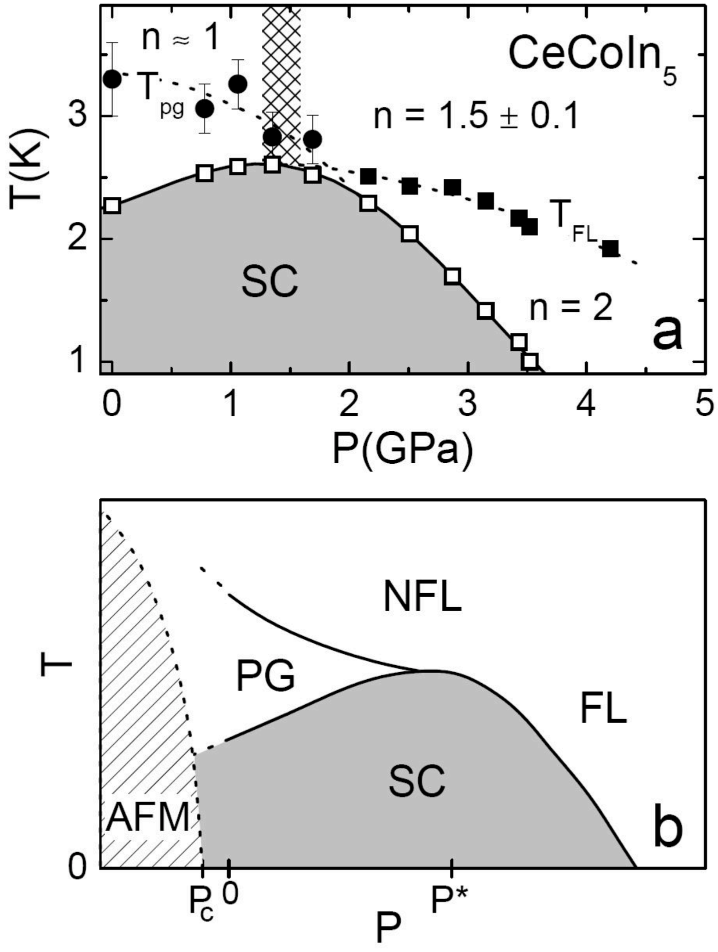
Ce M In$_5$ (M = Co, Rh, Ir): Quasi 2-D Crystal Structure and Fermi Surface:





R. Settai et al., JPCM **13**, L627 (2001)

CeCoIn$_5$: Phase Transitions



*V. A. Sidorov et al., PRL **89**, 157004 (2002)*

- **Quantum Phase Transition**
  with chemical substitution*,
  hydrostatic pressure,
  magnetic field ,
  (similar to cuprates)
- **FFLO Phase Transition**

- **Anisotropic type-II SC**
- **Heavy-fermion**

liquid $m_{\text{eff}} = 83m_0$

$T^* \sim 45$ K

- **Non-Fermi liquid**

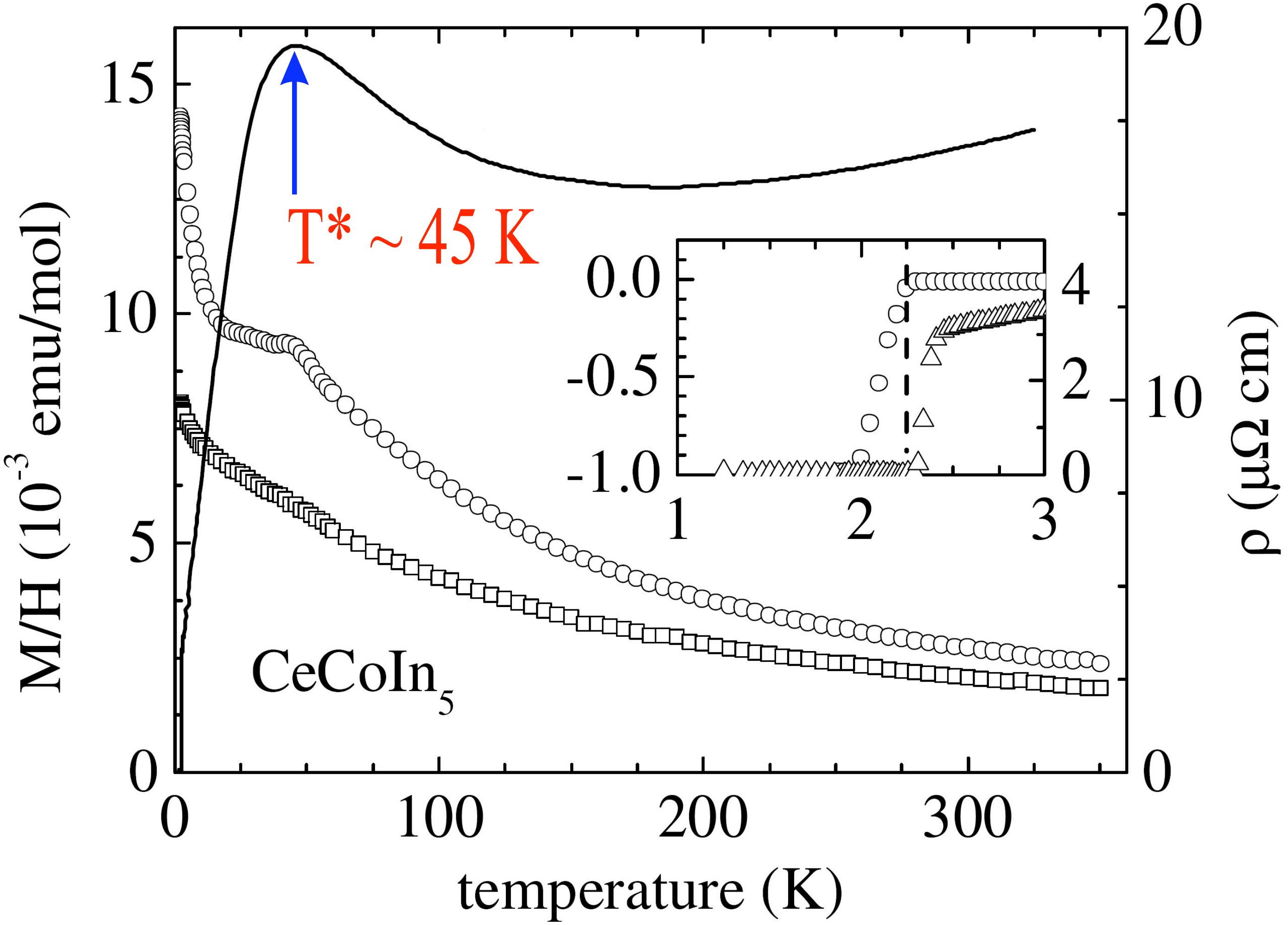
  $\rho \sim T^{1.0 \pm 0.1}$,
  $C_{en} / T \sim -\ln T$,
  $1 / T_1 T \sim T^{-3/4}$

*Cd, later

$CeCoIn_5$: The Ideal HFS for PCARTS

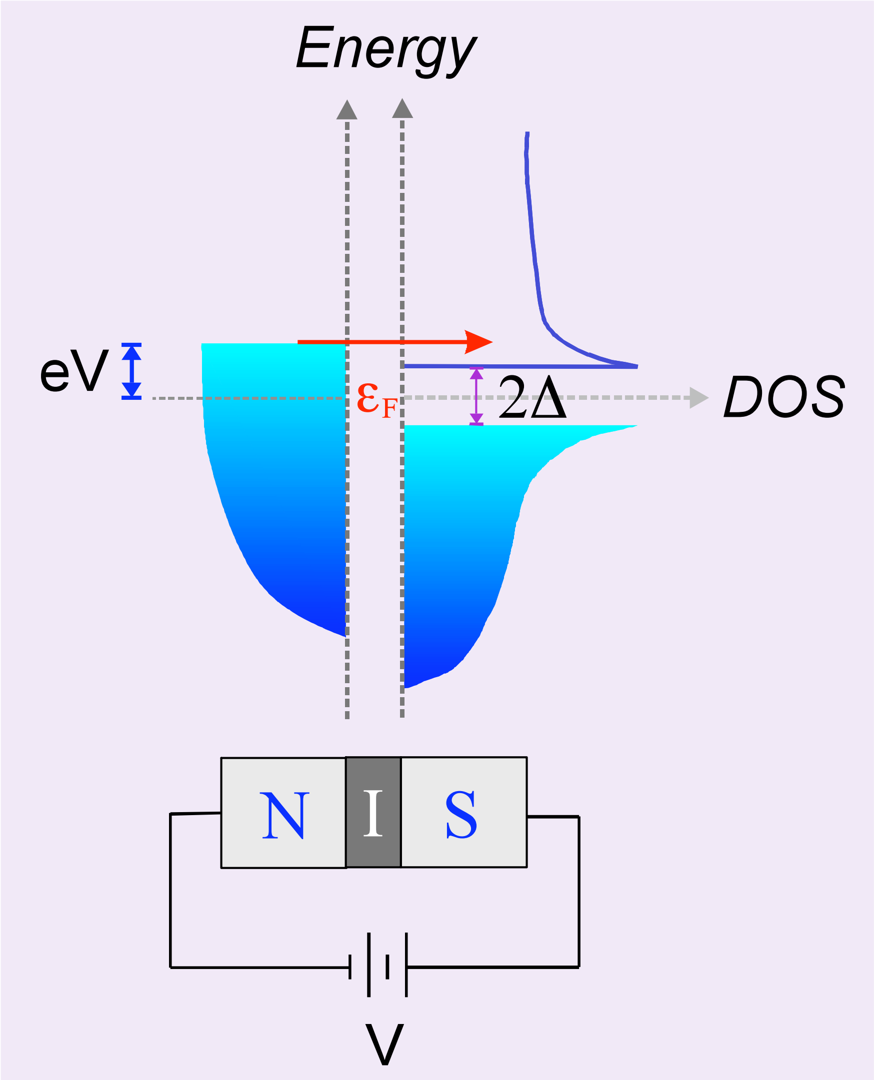
(Point Contact Andreev Reflection Tunneling Spectroscopy)

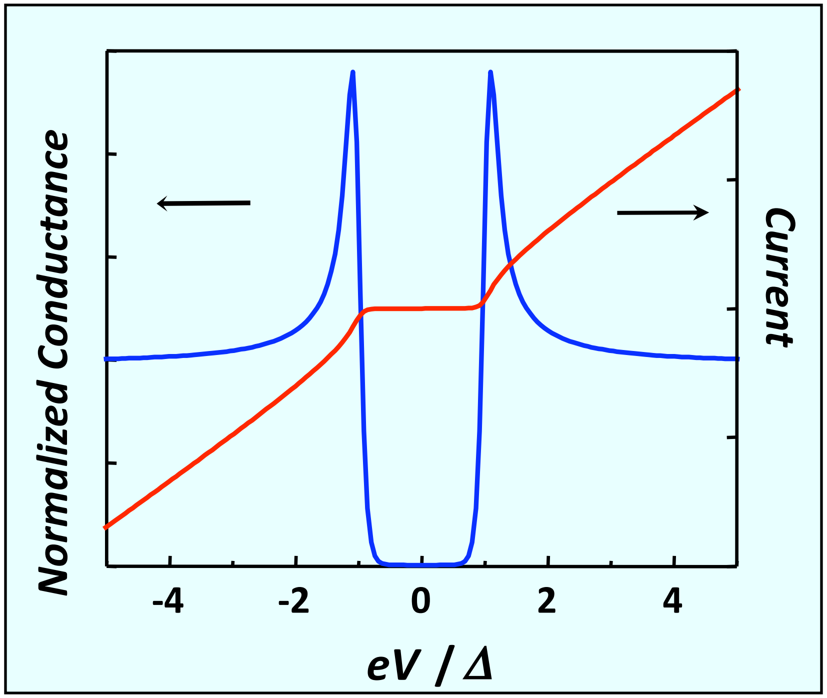
- T_c = 2.3K and T* = 45K; (high for HFS)
- Clean limit (*mfp* ~ 810 Å at T_c, increasing to ~3 µ at 400mK)



C. Petrovic et al., J. Phys.: Condens. Matter **13**, L337 (2001)

Quasiparticle Tunneling: The NIS Junction



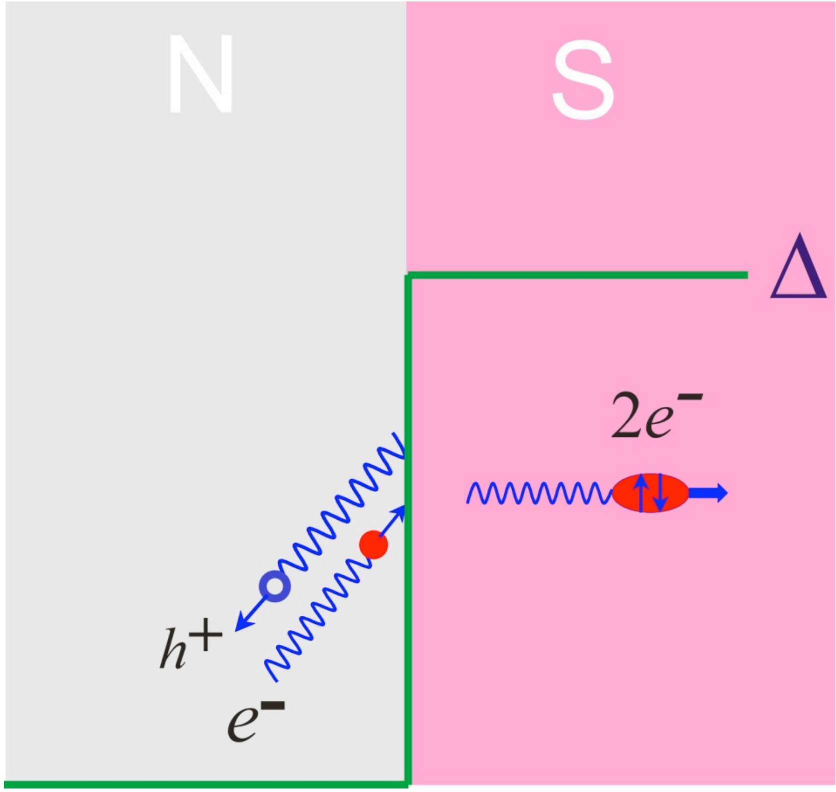


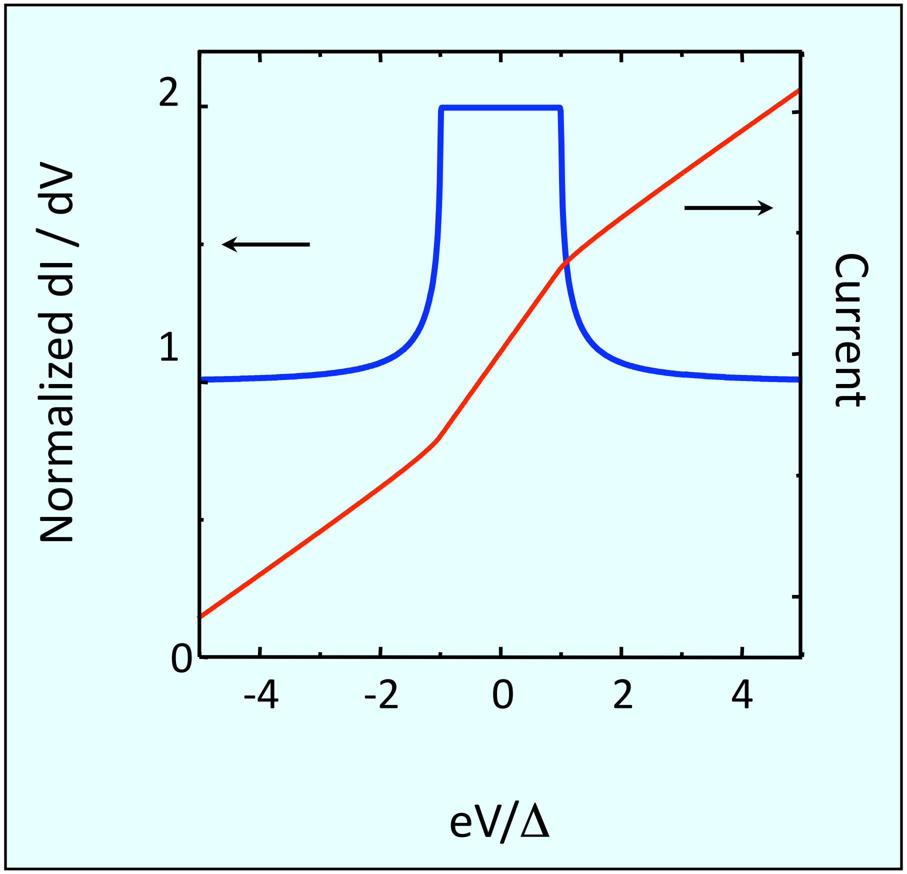
Near Δ, QP-DoS directly observed

Tunneling spectroscopy gives superconducting DOS.

What if no tunnel barrier?

Andreev Reflection: The NS junction



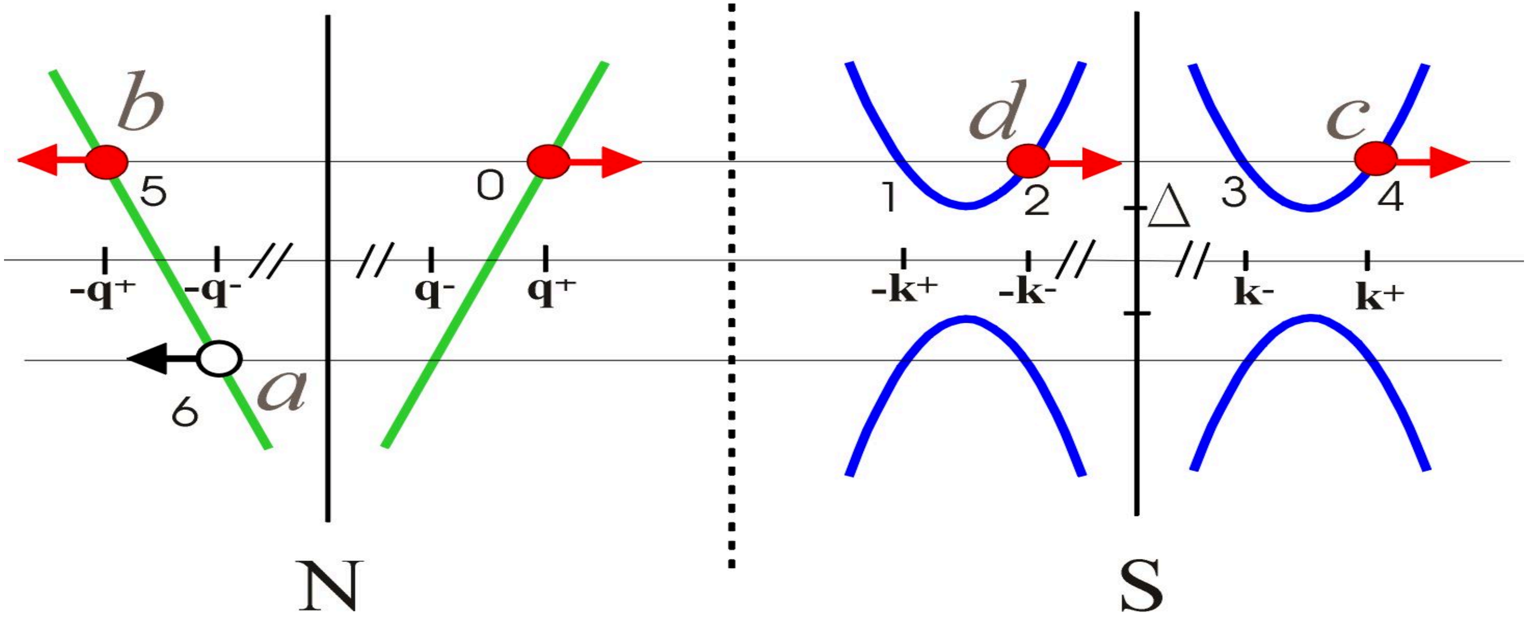


- Incident electron is retro-reflected as a hole, $\boldsymbol{v_h = -v_e}$.
- Conductance is doubled below the gap energy.
- DoS effects can be observed (discussed later) …

Blonder-Tinkham-Klapwijk (BTK) Model

Charge transport across N/S interface - *PRB **25**, 4515 (1982)*

Assumes abrupt 1-D interface and **Ballistic Transport**



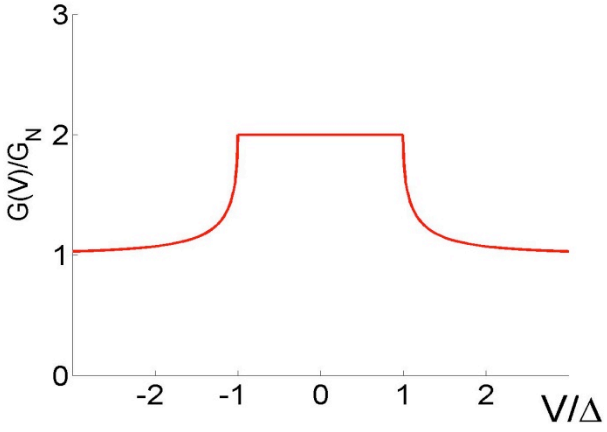
a : Andreev reflection
b : Normal reflection
c : Transmission w/o branch-crossing (electron-like)
d : Transmission with branch- crossing (hole-like)

$$A(E)+B(E)+C(E)+D(E)=1$$

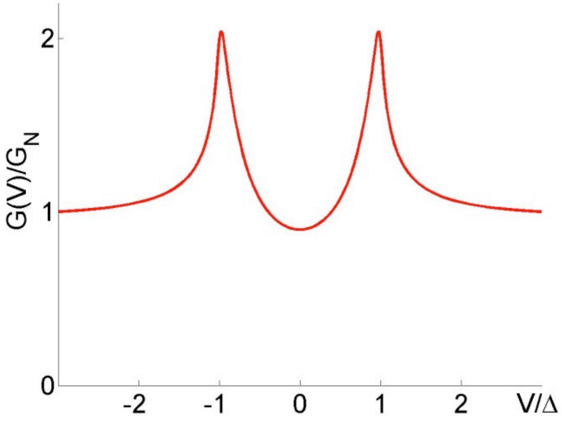
BTK model: Three fitting parameters

Δ = superconducting gap
Γ = Dynes broadening factor (qp scattering rate)
Z_{eff} = barrier strength at the N/S interface

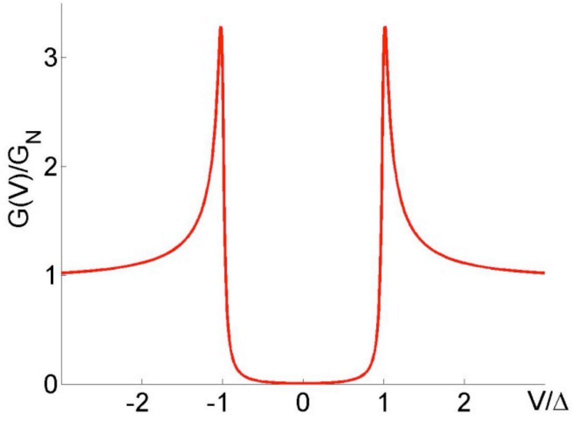
Effect of increasing Z (or Z_{eff}):



Z = 0
Andreev Reflection



Z = 0.5

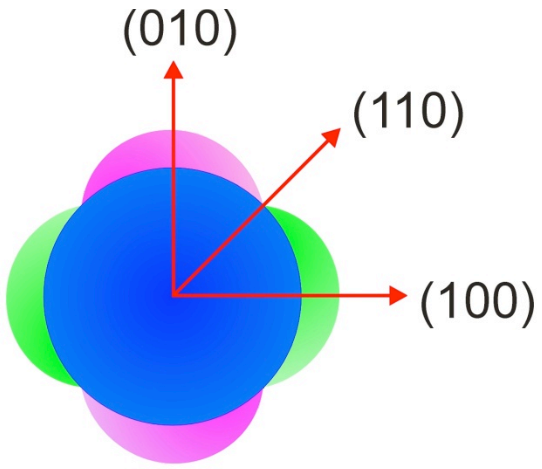


Z = 5.0
Tunneling

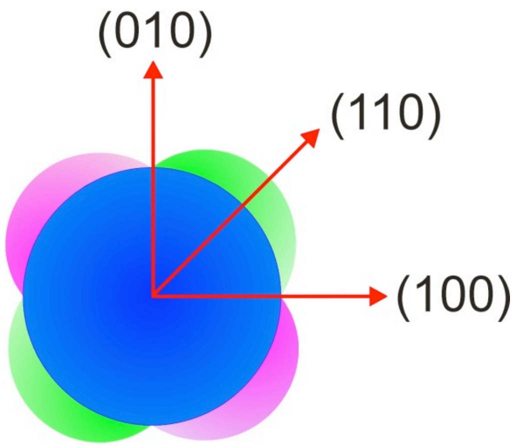
Assuming $\Gamma = 0$ and $\Delta = 1$

OP Symmetry of CeCoIn$_5$: Previous work

- Evidence for the existence of line nodes:
  Power law dep: $C_{en} / T \sim T$, $\kappa \sim T^{3.37}$, $1/T_1 \sim T^{3+\varepsilon}$, $\lambda \sim T^{1.5}$
- Four-fold symmetry of field-angle dep in thermal cond.:
  small angle neutron scattering $\Rightarrow d_x2\text{-}{}_y2$
  specific heat $\Rightarrow d_{xy}$
- Spectroscopic evidence was lacking to determine the locations of line nodes: (110) or (100) i.e. d_{xy} or $d_x2\text{-}{}_y2$?

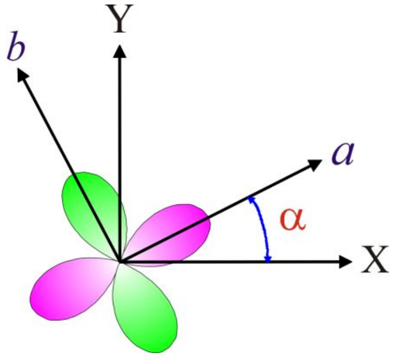


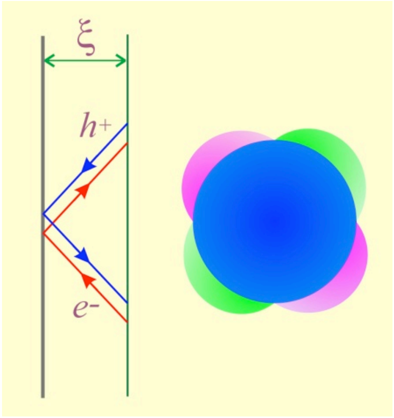
$d_{x^2-y^2}$



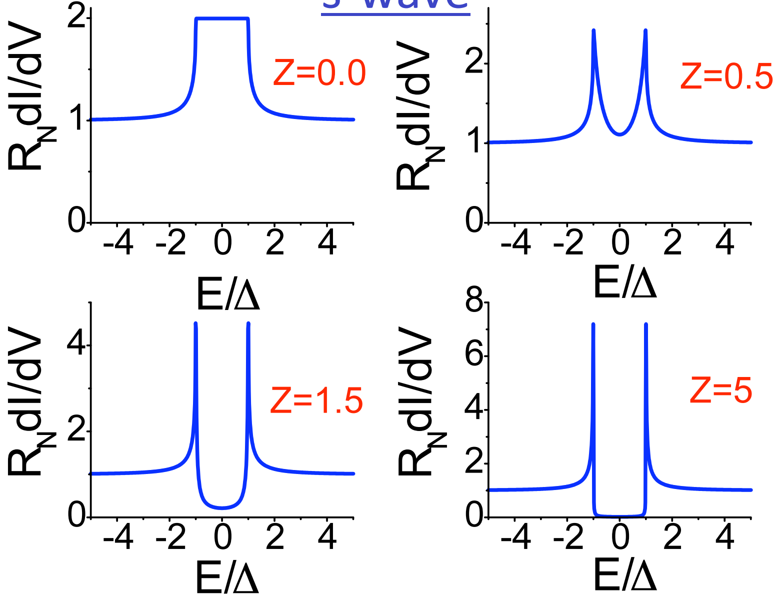
d_{xy}

BTK Model for *s-wave* *and extended to d-wave*

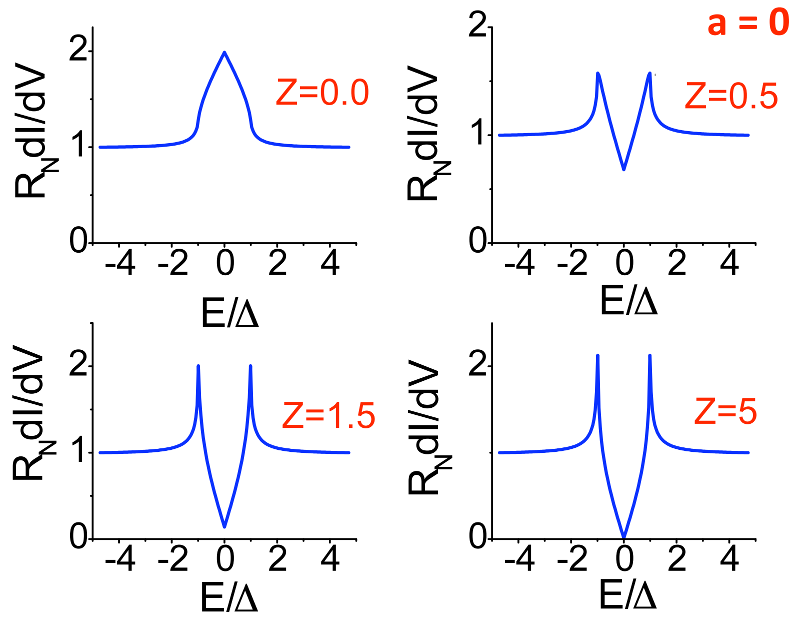




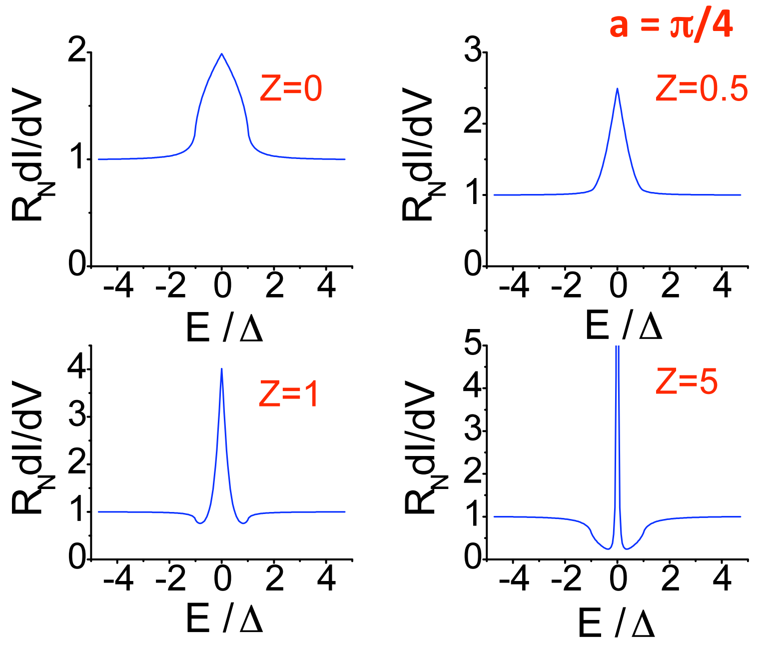
s-wave



d-wave: c-axis or lobe direction



d-wave: nodal direction

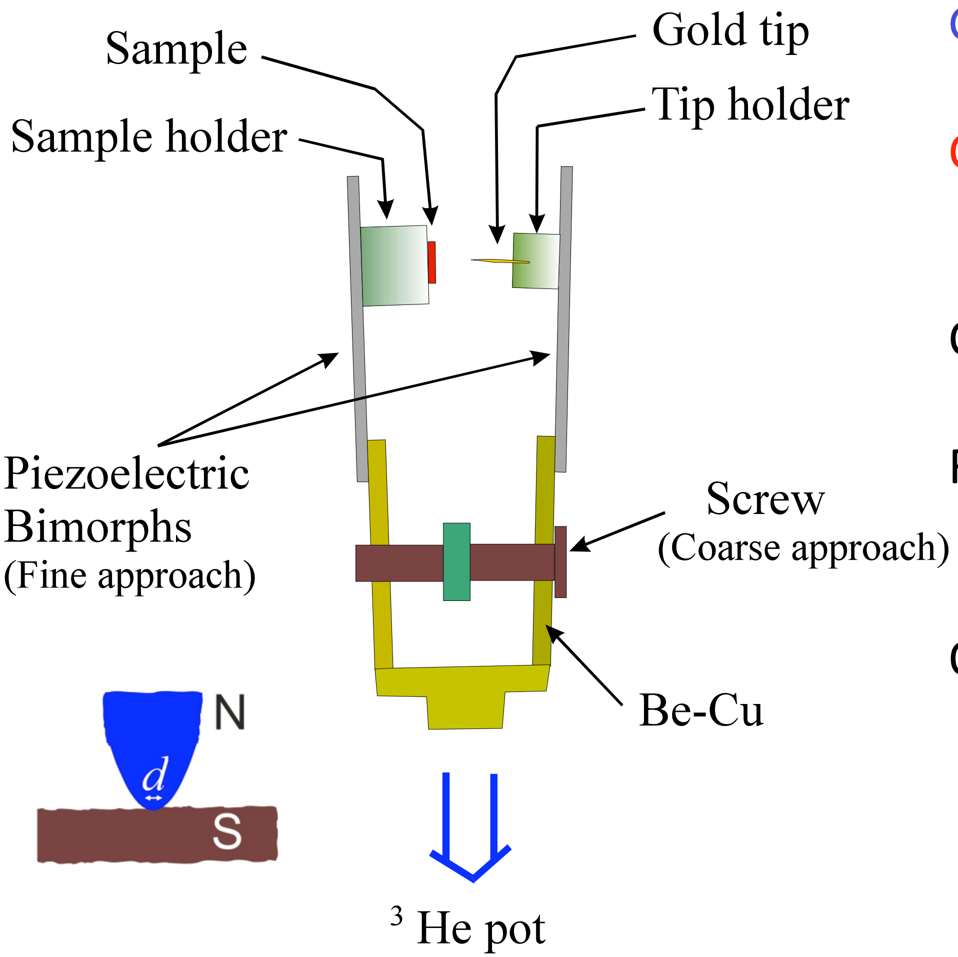


Our Experiments:

Point Contact Andreev Reflection Spectroscopy **(PCARS)**

1) Cantilever-Andreev-Tunneling (**CAT**) Rig

W.K. Park, LHG, RSI (06).



Gold tip

- electrochemically etched

$CeCoIn_5$ single crystal

- *(001), (110) and (100)* oriented
- etch-cleaned using H_3PO_4

Coarse approach

- done before inserting probe

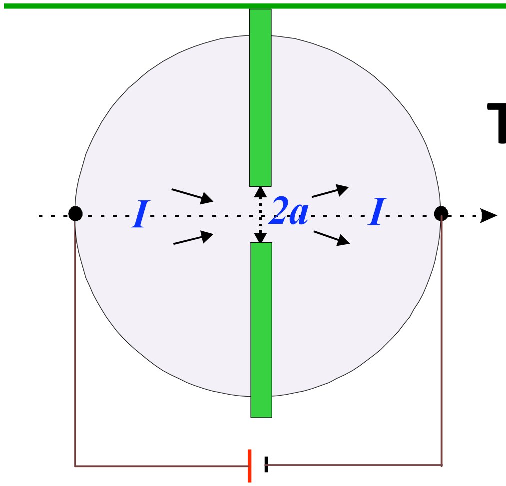
Fine approach

- done during cool down
- piezo driven by computer control

Operation range

- Temperature : down to 300mK
- Magnetic Field : up to 12T

Basics of PCARTS: Contact Regimes



To do spectroscopy, the contact <u>must</u> be in the

Ballistic or Sharvin limit ➔

Contact diameter < mfp in either material

Wexler's Formula (Proc. Phys. Soc. 89, 927 (1966)

For our experiment (R_N = 1-4 Ω) and

* Upper limit of 2a = 46 nm

* l_{el} at T_c= is 81 nm (from thermal conductivity), and up to 4-5 μm at 400mK.

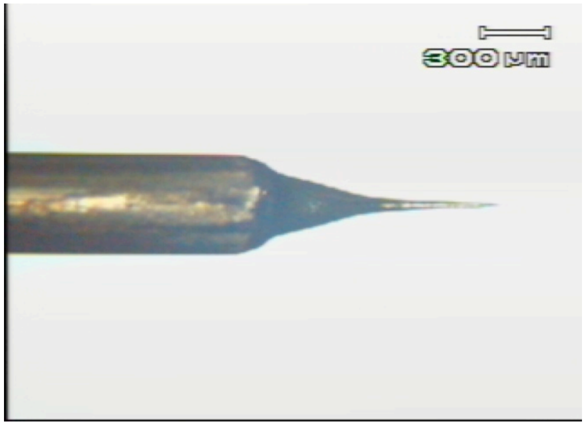
Resistance within Sharvin limit

But this is NOT enough: Also MUST show (we have):

1. **Junction resistance is T-independent**
2. **REPRODUCIBILITY**

Basics of PCS: Tip production

The sharp gold tip is electrochemically etched in hydrochloric acid



For our experiment (R_N = 1-4 Ω) and **not T-dep**:

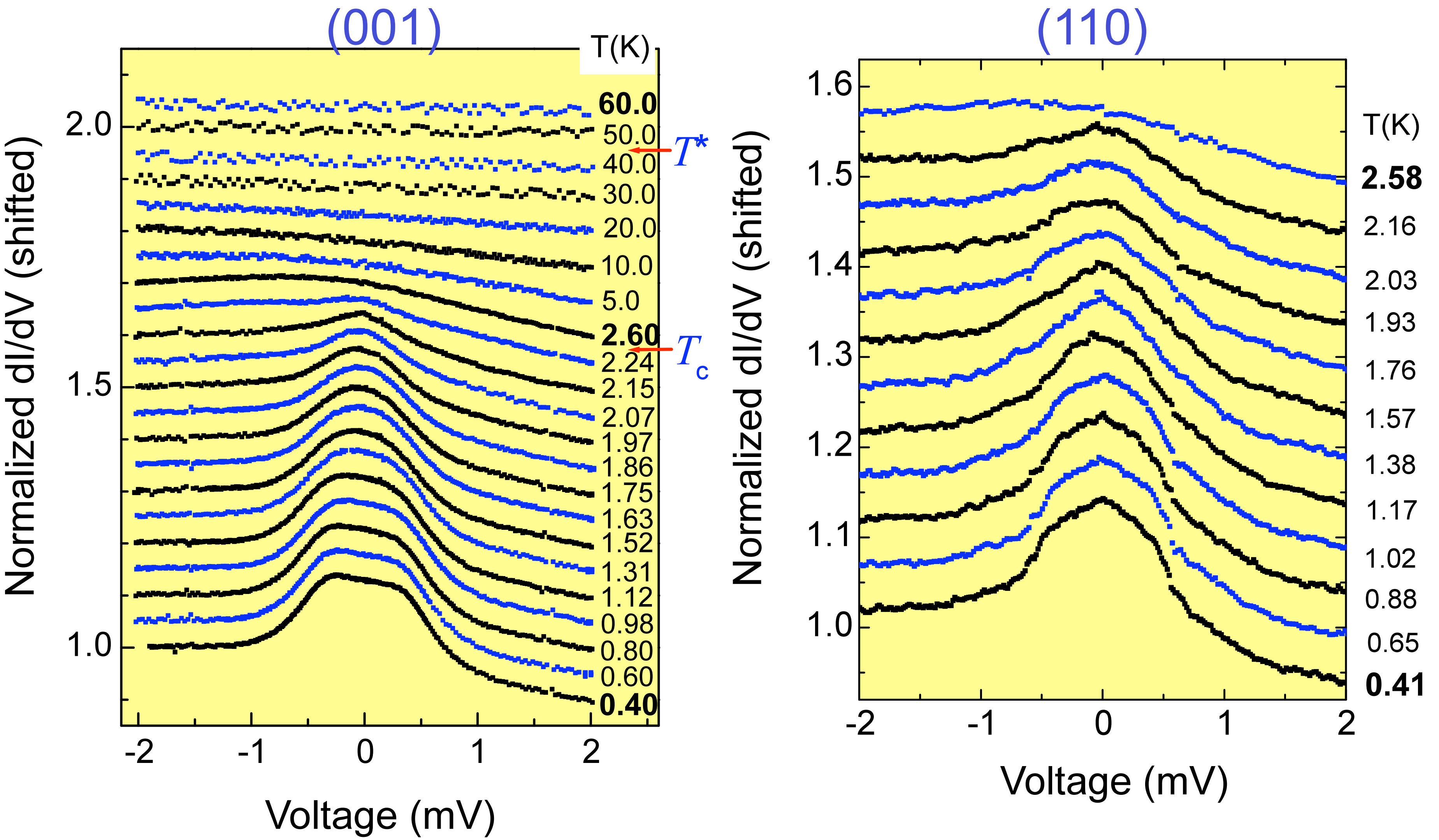
* Upper limit of 2a = 46 nm

* l_{el} at T_c= is 81 nm (from thermal conductivity),

and increases with decreasing T, to 4-5 μm at 400mK.

Our experiments are in the Sharvin Limit,
and are reproducible.

Andreev Reflection Conductance of Au/CeCoIn$_5$

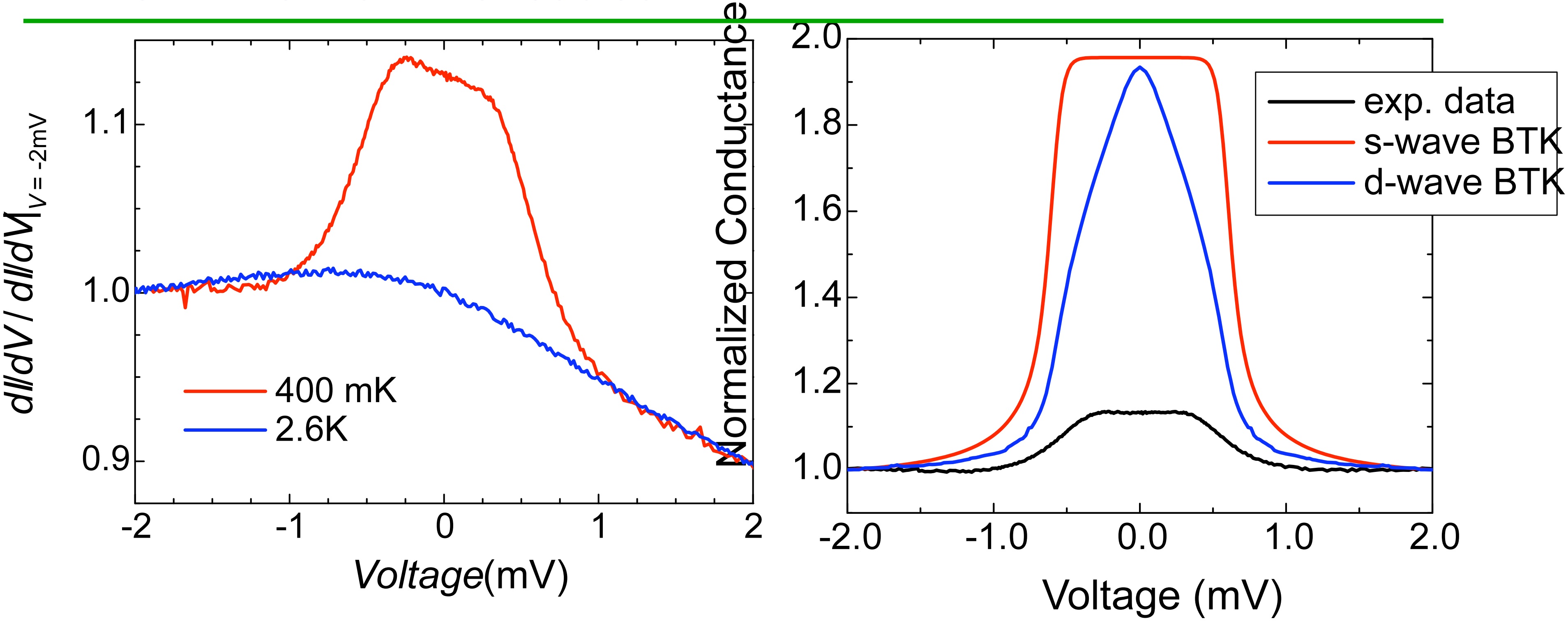


(001)

(110)

Conductance asymmetry begins at T^* and saturates below T_c

Definition of the issues



1. **Understanding charge transport across HF interface**

   Existing models cannot account for

   Andreev reflection at the HFS/N interface

2. **Spectroscopic studies of CeCoIn$_5$**

   OP symmetry, mechanism, DoS …

3. **Competing AF and S phases in Cd:CeCoIn$_5$?**

Effective Barrier Strength, Z_{eff}:

$$Z_{eff}=\sqrt{Z^2+\frac{(1-r)^2}{4r}}, \qquad r = v_{FN}/v_{FS}$$

- Takes into account the mismatch of the Fermi velocities
- Has worked well for a <u>wide</u> range of materials, but not for HFS

For the HFS/N interface $Z_{eff} \geq 1$ (tunneling limit)
=> Andreev reflection should never occur!

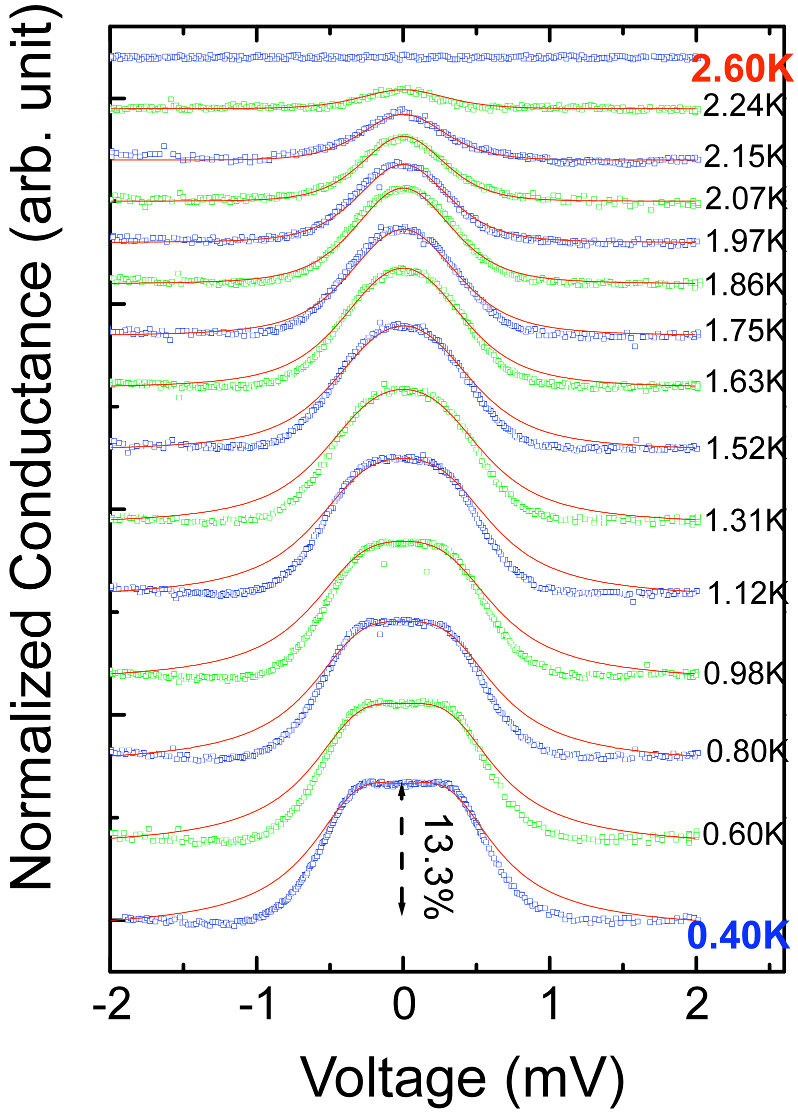
Suppressed AR is routinely observed cannot be explained by existing theories.

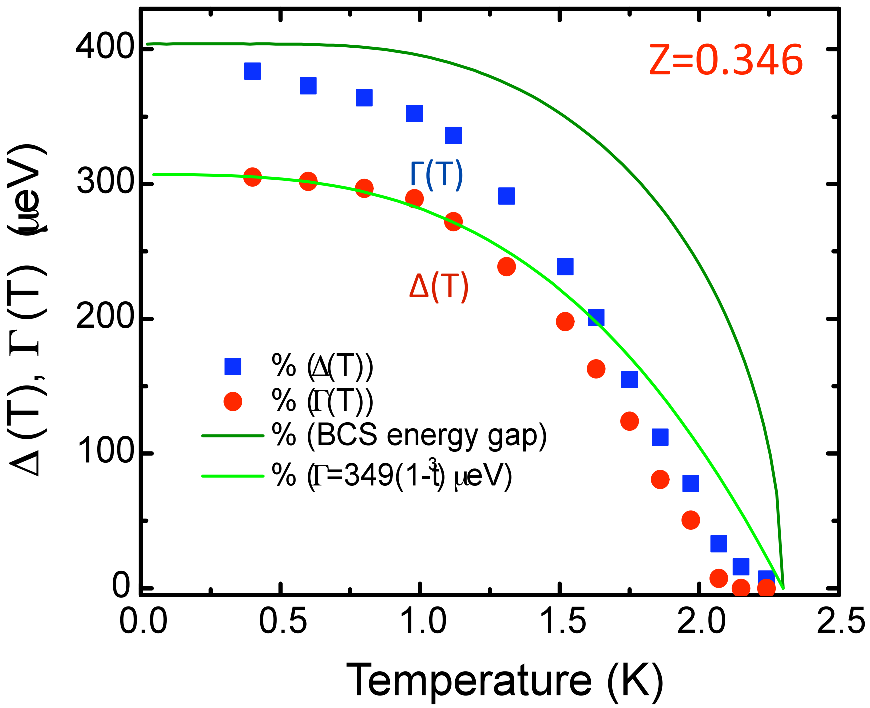
Will can account for this by:

1. Relaxing mismatch constraint (Deutscher, Nozières ‘94)
2. Two-fluid model (Nakatsuji, Pines & Fisk ‘04)
3. Energy-dependent DoS

Earlier (unsuccessful) Fits of the T-dep Conductance

(also, s-wave & d-wave fits are indistinguishable)

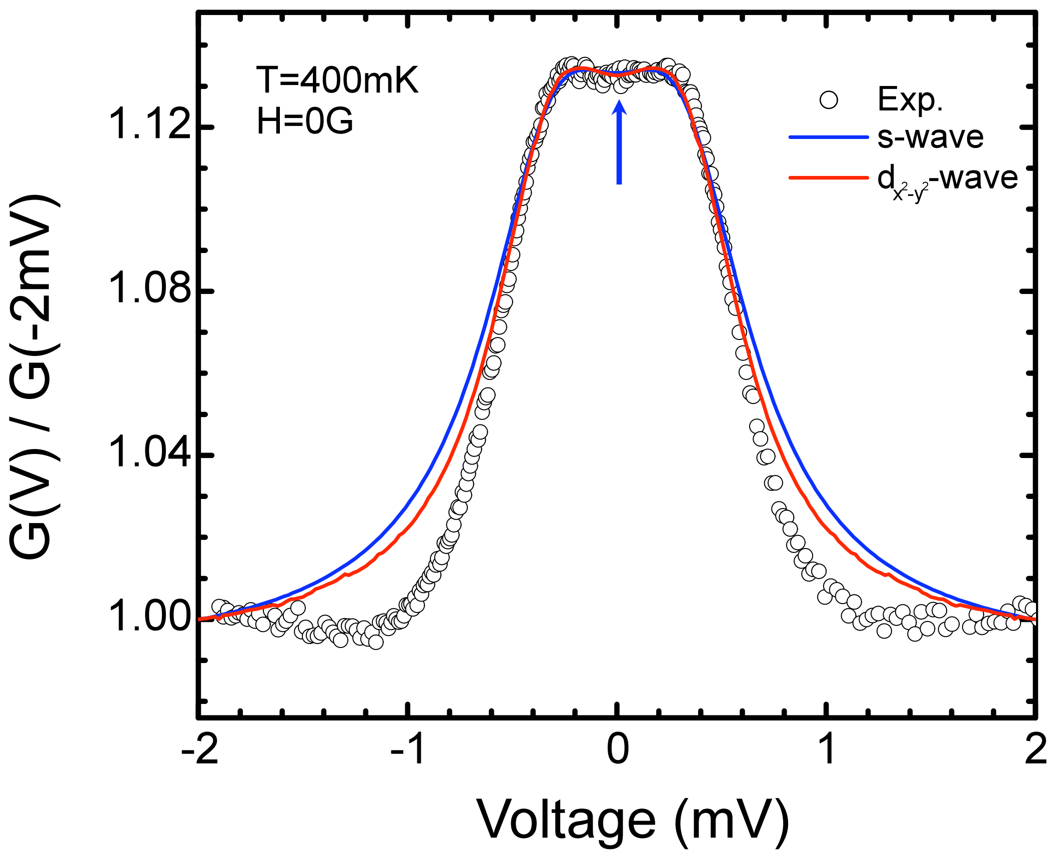




This decreasing Γ with decreasing T is not physically meaningful: (i.e., breakdown of BTK).

Conductance Fit at Lowest Temperature

d-wave vs. s-wave fitting



Fitting Parameters

	s-wave	d-wave
Z	0.346	0.215
Δ(μeV)	384	460
Γ(μeV)	305	220

(can’t differentiate)

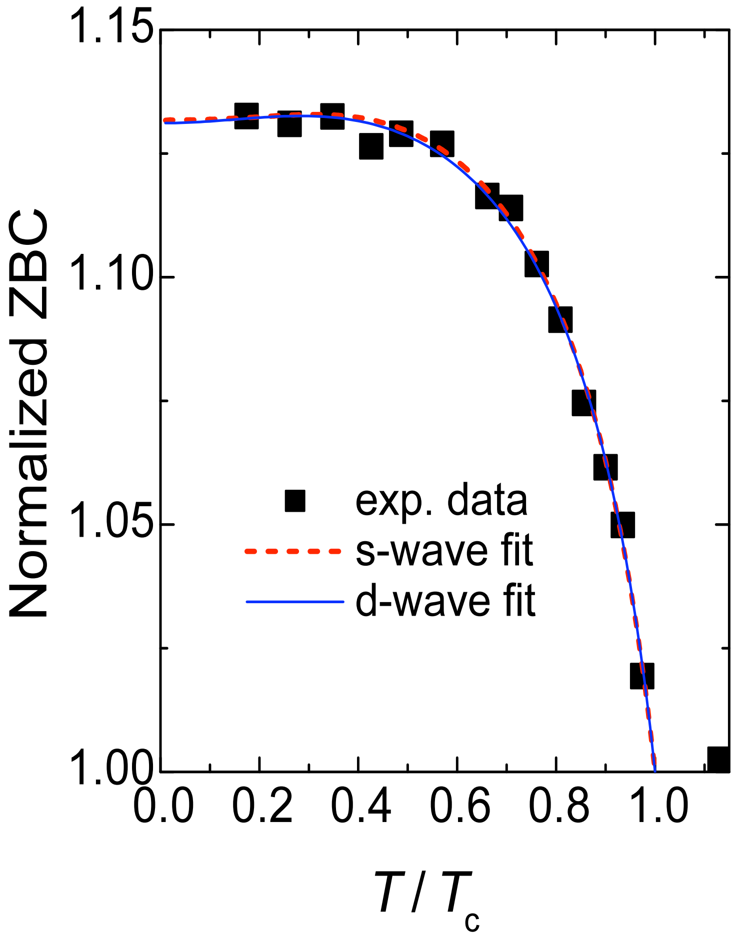
BUT:

$$\frac{2\infty}{kT_{Bc}} = 4.64$$

⇒ strong coupling (as expected)

FIRST reasonable result using BTK theory

Zero-bias Conductance (ZBC) vs. Temp. Fit



Fitting Parameters

	s-wave	**d-wave**
Z	0.346	0.365
Δ	Δ(0) = 349 meV,	$\Delta(T, f) = \Delta(0)\cos(2\theta)$, $\Delta(T) = 2.35 k_B T_c \times \tanh(2.06 (T_c/T-1)^{1/2})$
Γ	$\Gamma(t) = 0.86\,\Delta(0) \times (1-t^3/3)$	**218 meV**

$$\geq = \frac{\hbar}{\Pi}$$

d-wave fit: T-indep and “reasonable” Γ

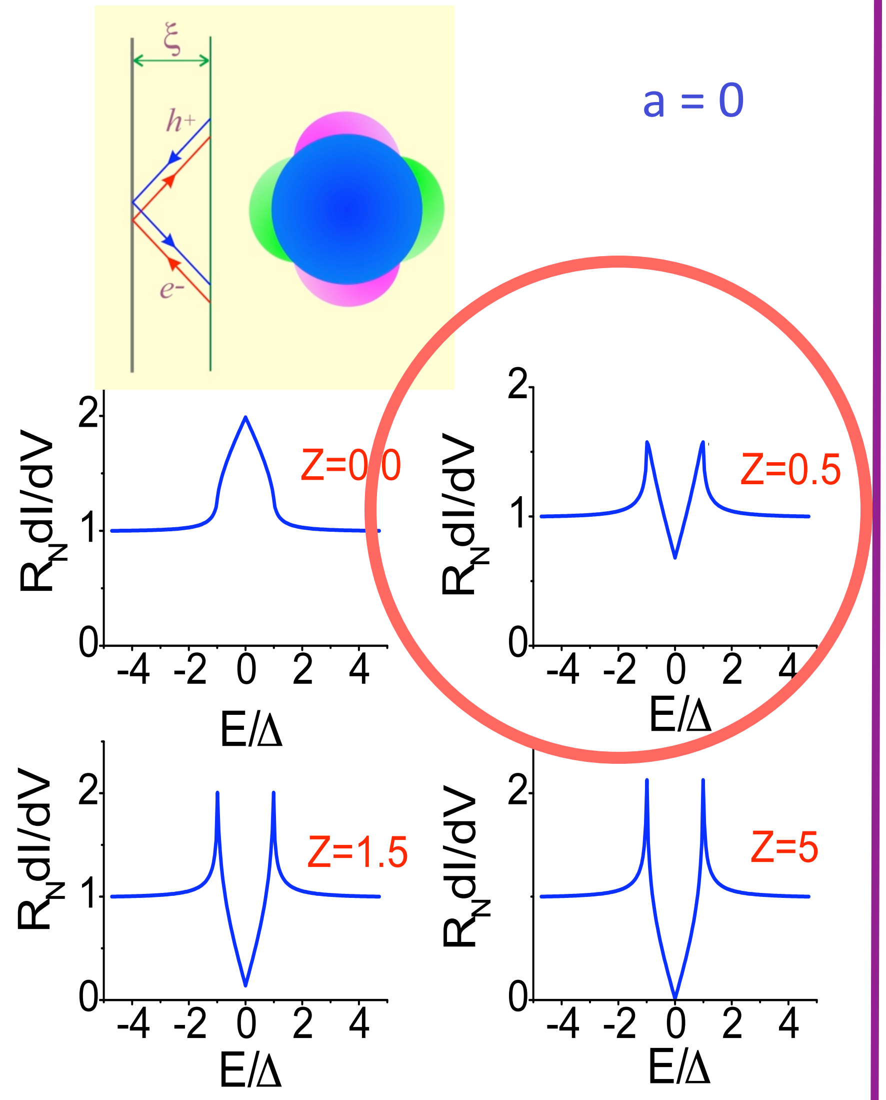
⇒ d-wave (as expected)

SECOND reasonable result using BTK theory

Calculated BTK conductance using a d-wave OP

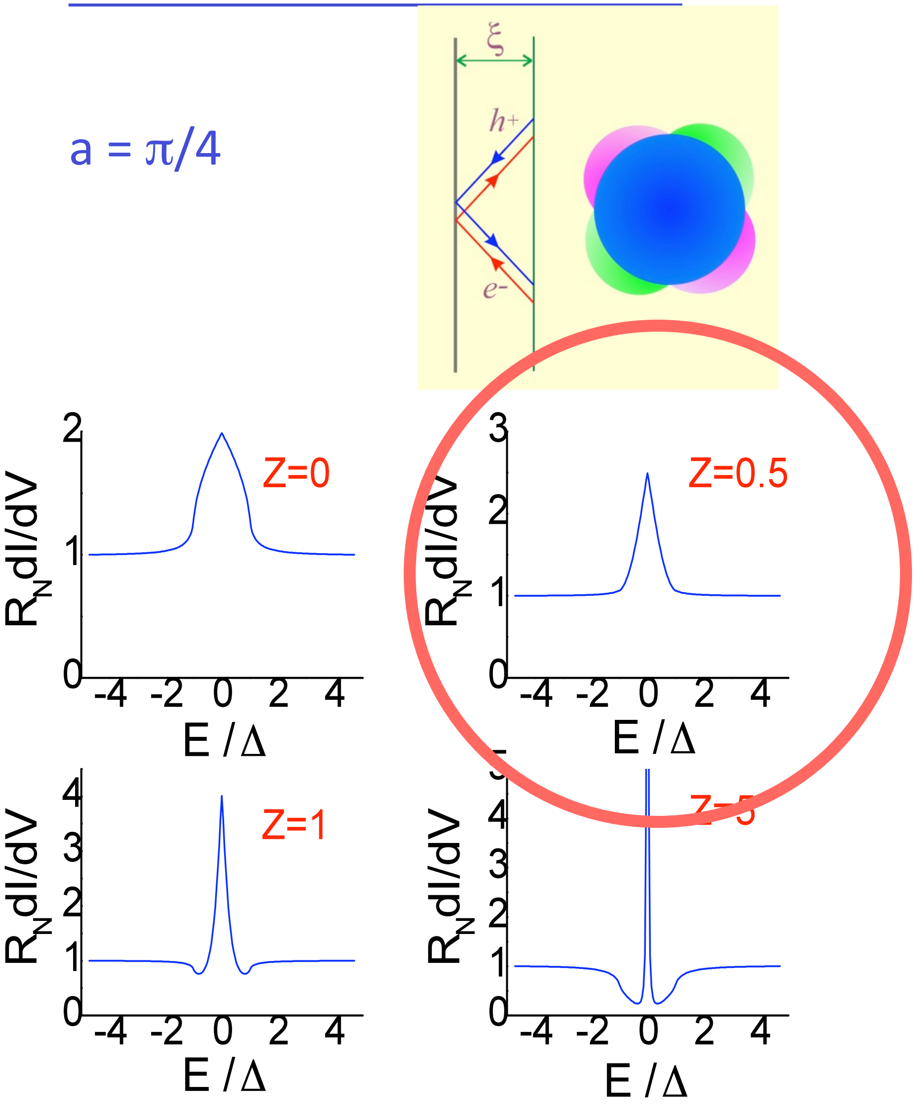
d-wave: c-axis or lobe direction

a = 0



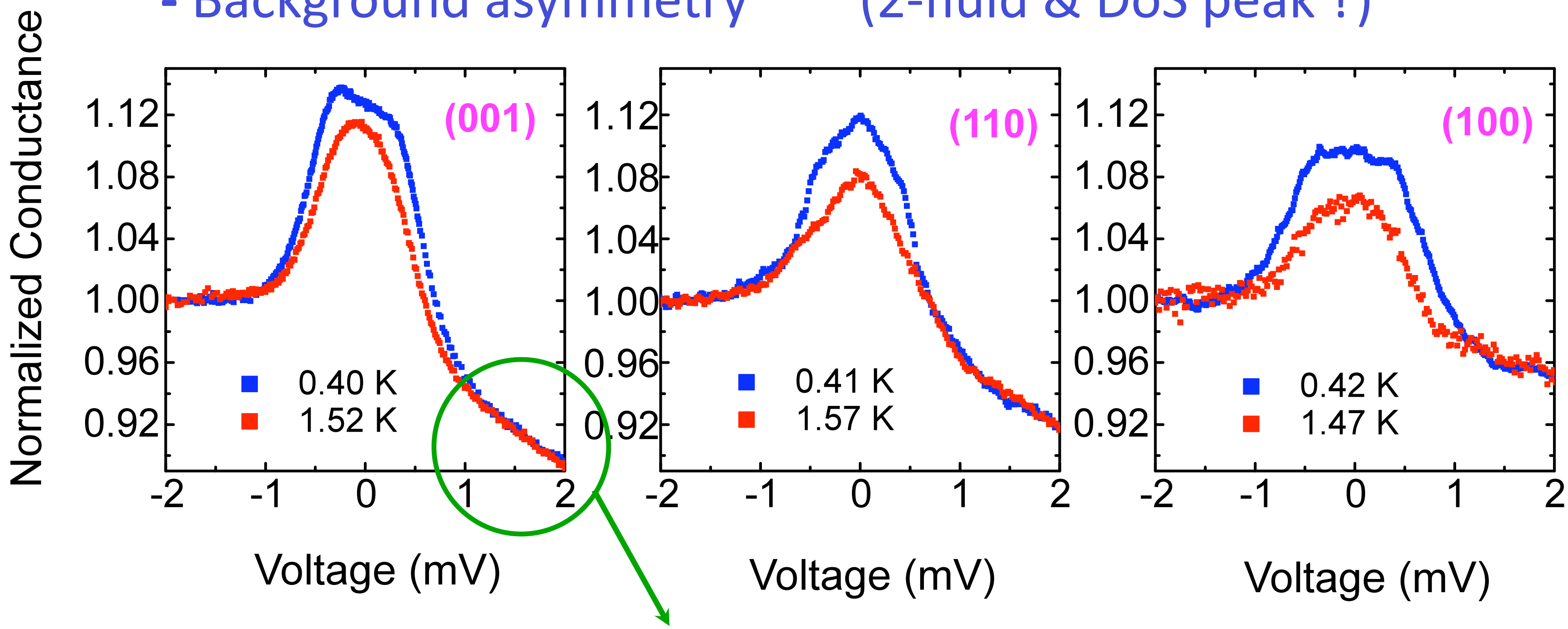
d-wave: nodal direction

a = π/4



Consistency Along Three Orientations

- Conductance magnitude (AR)
- Conductance width (Δ)
- Background asymmetry (2-fluid & DoS peak ?)

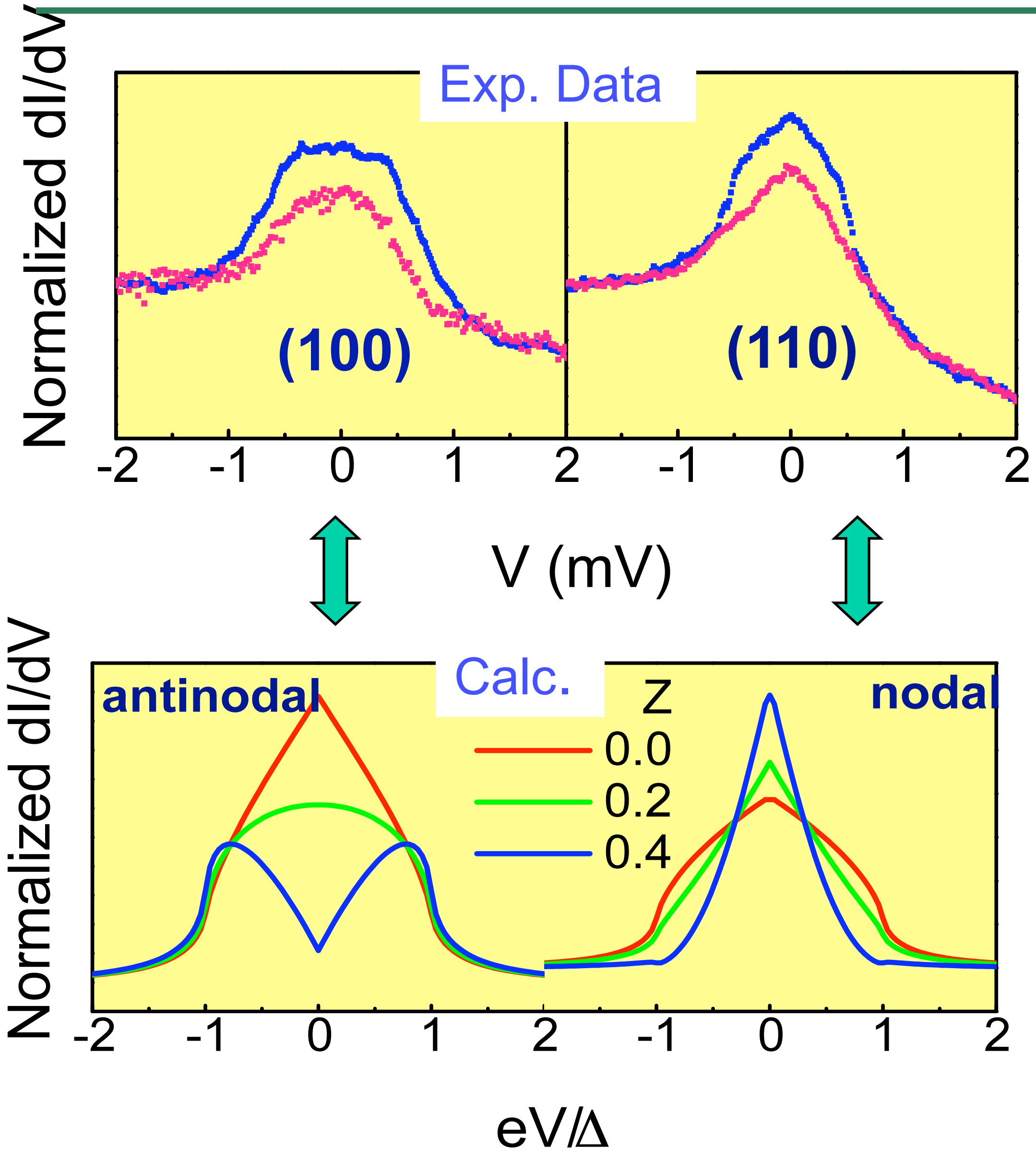


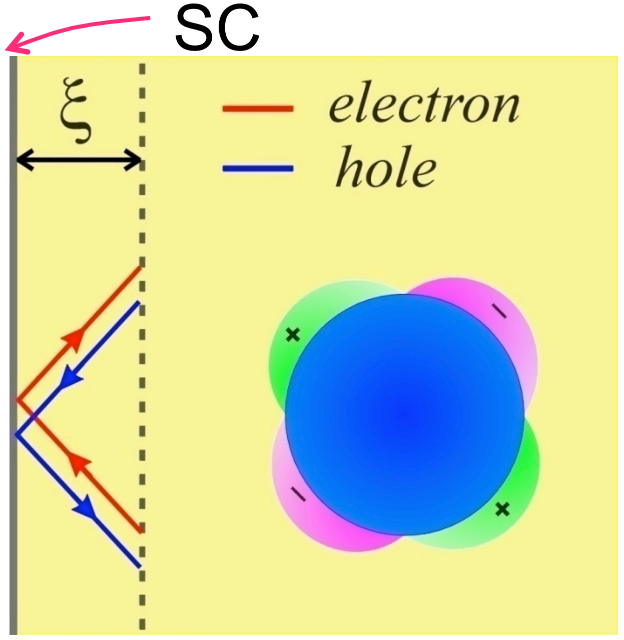
(+) $CeCoIn_5$; (−) Au

=> Adding electrons to $CeCoIn_5$ above the Fermi energy is more difficult than removing them

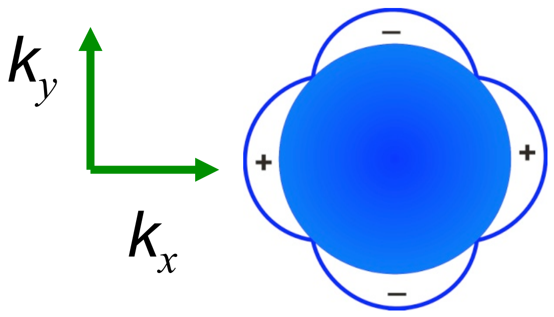
Note also the shapes of the conductance curves

Spectroscopic Evidence for $d_{x^2\text{-}y^2}$ Symmetry



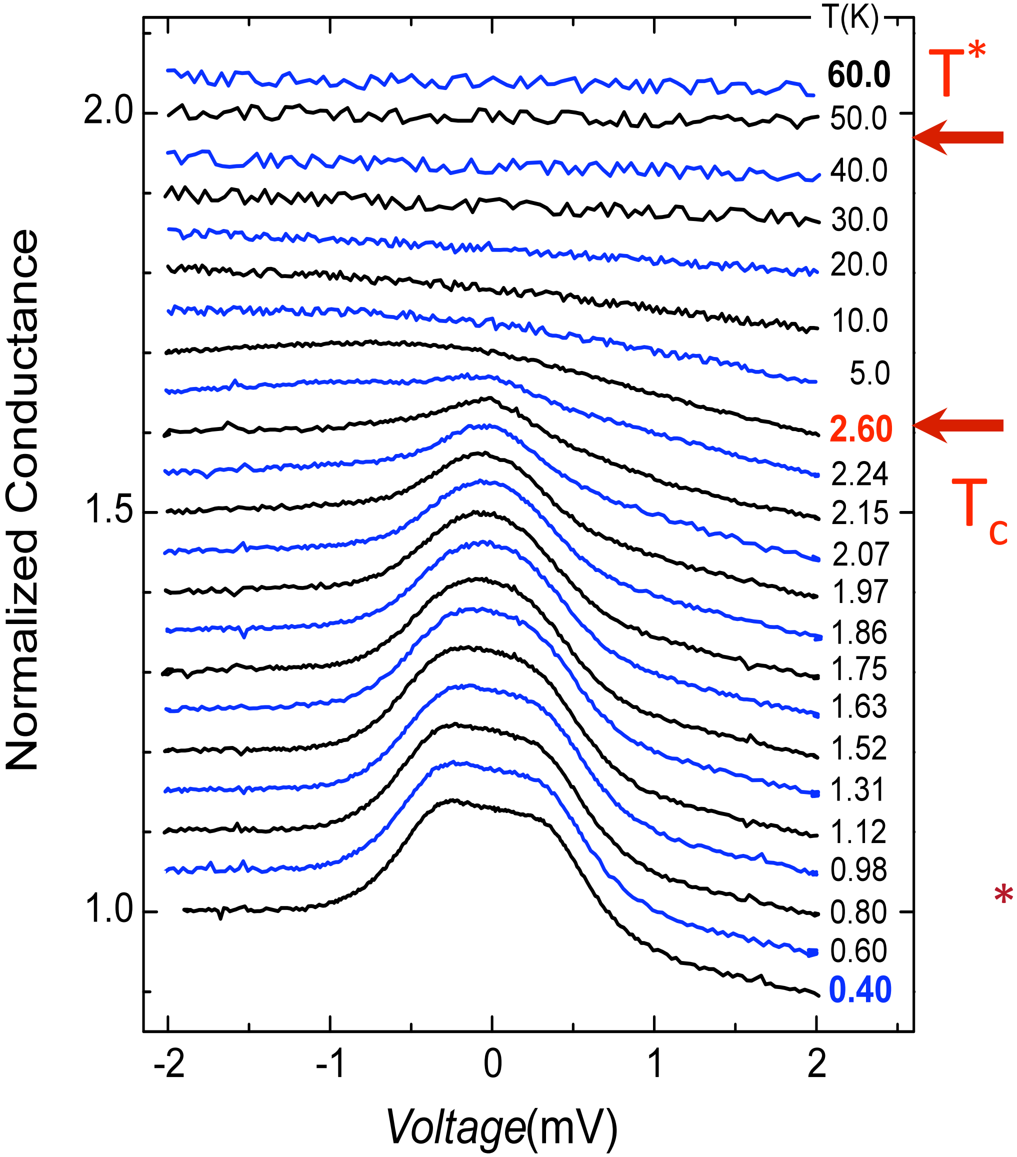


Andreev Bound States (ABS)



WKP et al., PRL **100**, 177001 (2008)

Background Conductance Asymmetry of Au/$CeCoIn_5$

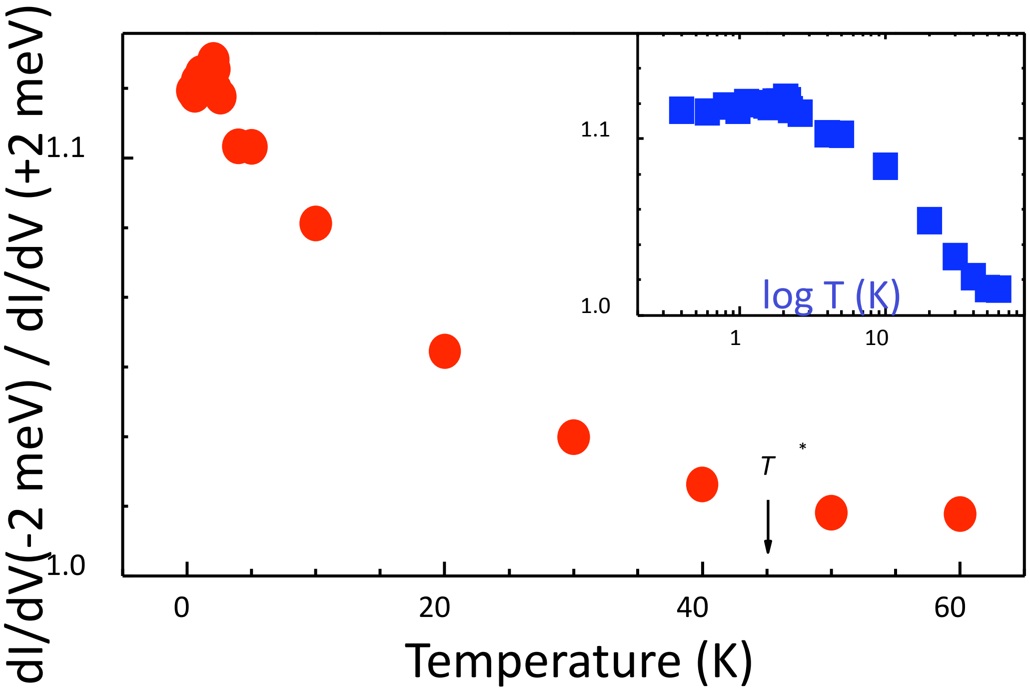


Background develops an asymmetry* at the heavy-fermion liquid coherence temperature,
T* ~ 45 K.

This asymmetry gradually increases with decreasing temperature until the onset of superconducting coherence,
T_c =2.3 K.

* el-h asymmetry described by Nakatsuji, Pines & Fisk, PRL **92**, 016401 (2004)

Background Conductance Asymmetry

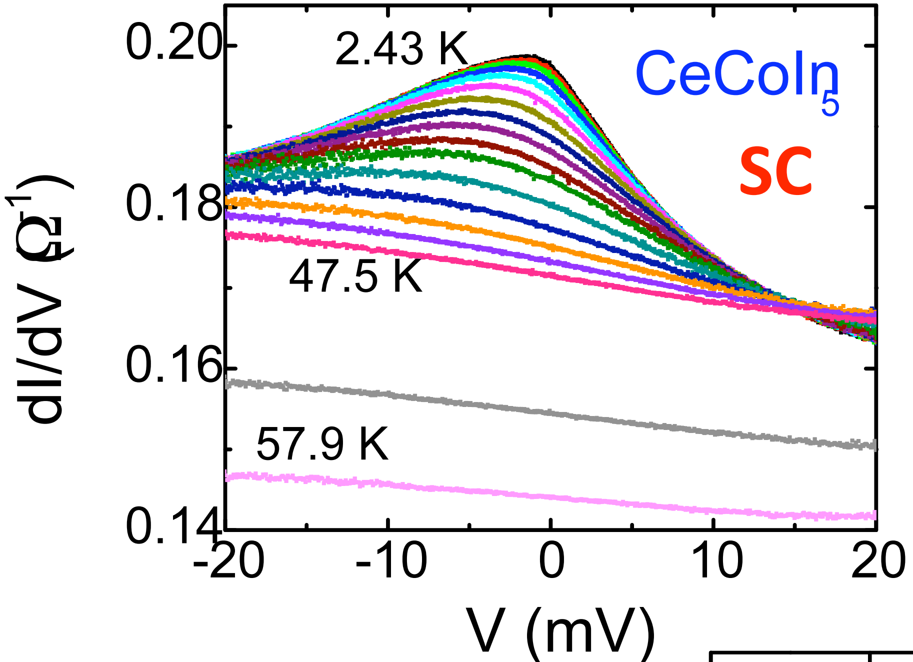


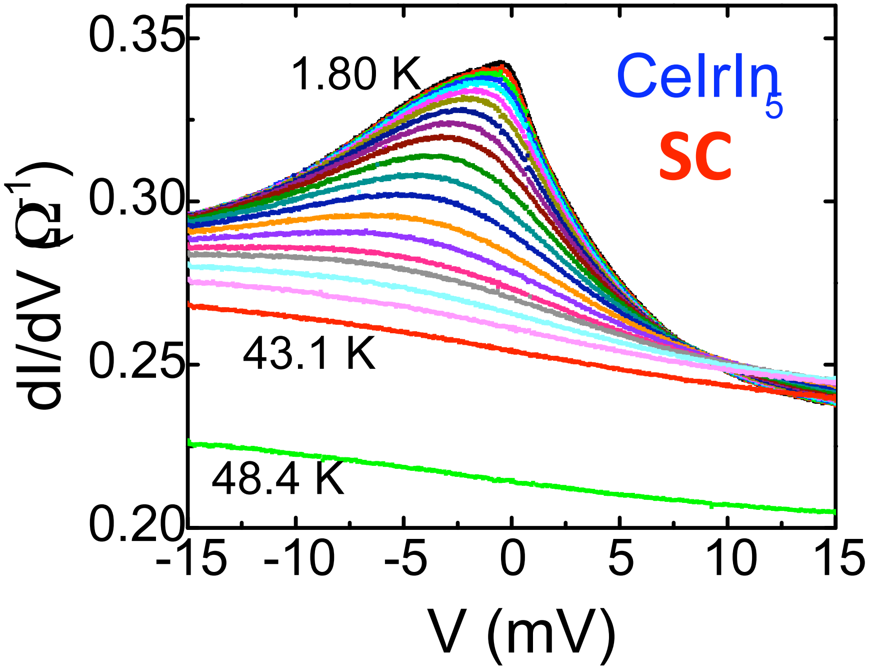
T-dep of background conductance asymmetry follows that of:

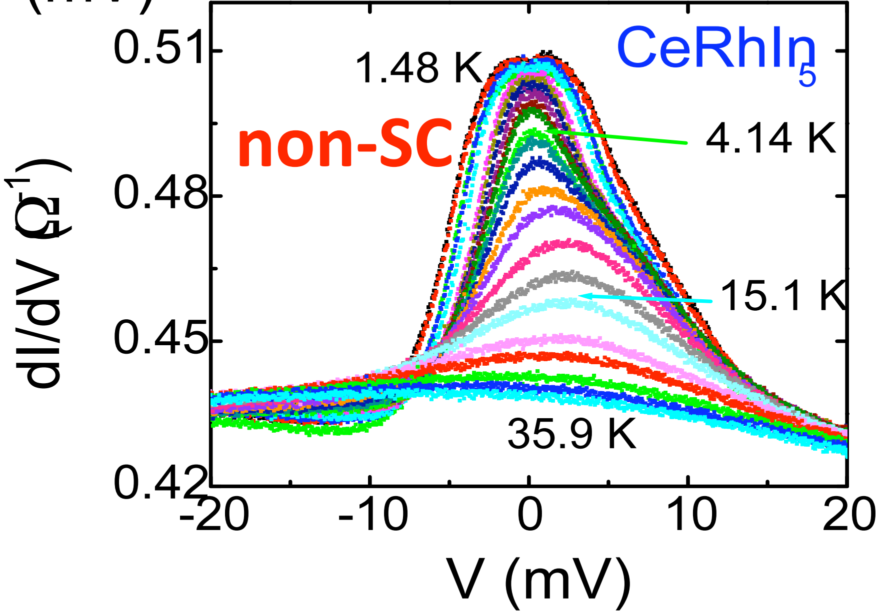
- Spectral weight (specific heat): Nakatsuji, Pines, Fisk, PRL ‘04
- NMR Knight shift (spin susceptibility): Curro et al., PRB ’04

Described by Y.-F. Yang and D. Pines,
“Universal Behavior in Heavy Electron Materials” (cond-mat)

Background Conductance Asymmetry of Au/CeMIn$_5$





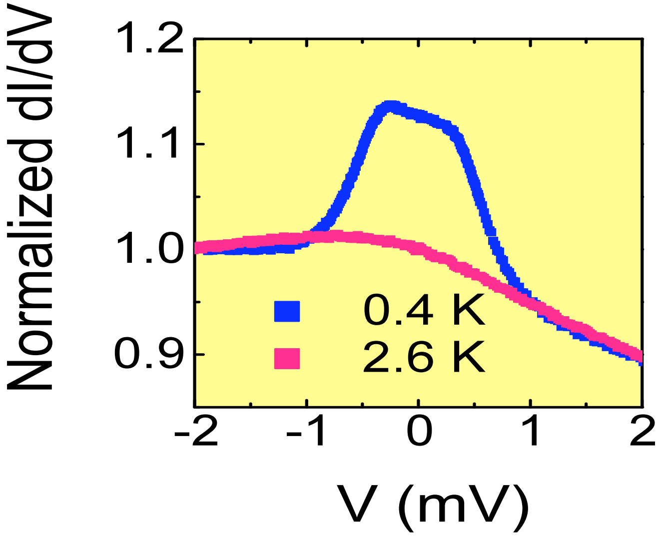


- **Co- & Ir-115: qualitatively similar**

- Rh-115: additional structure due to AFM (also, in Cd-doped Co-115)

Why is the conductance asymmetric?

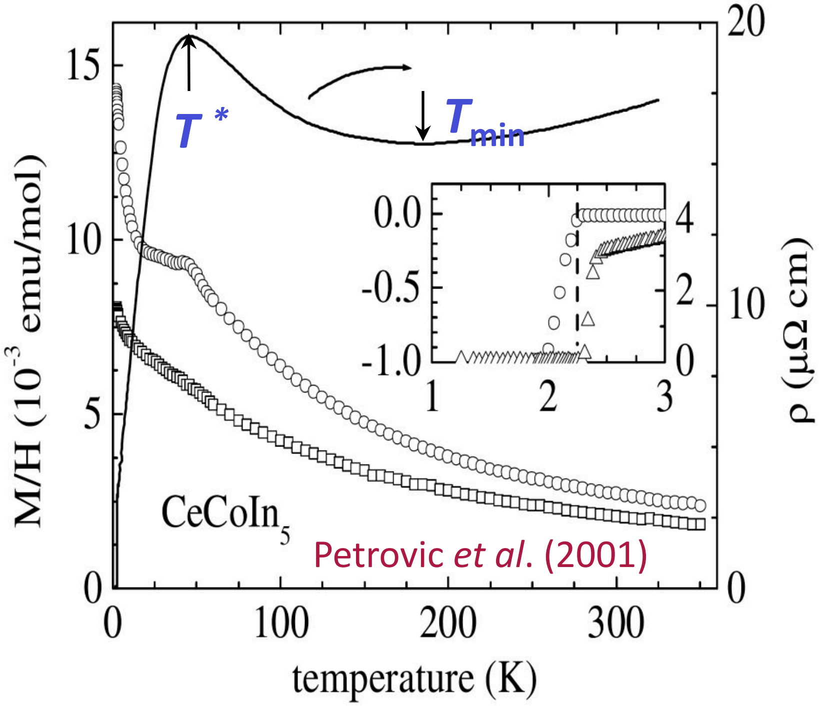
- Asymmetry is reproducible; conductance is always smaller when HFs are biased positively for the two SC 115s.



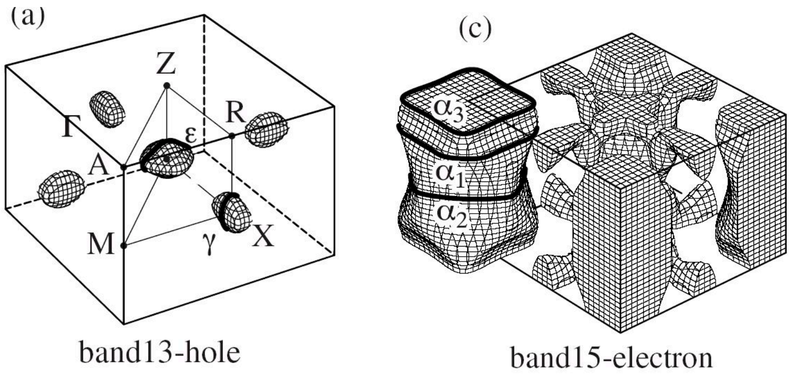
Relevance of Proposed Models

- Competing order (Hu & Seo, PRB 2006)
  - Does not explain STS data on UD-Bi2212, nor our $\mathrm{CeIrIn_5}$ data.
- Non-Fermi liquid behavior (Shaginyan, Phys. Lett. A 2005)
  - Asymmetry is still seen in field-induced Fermi liquid regime.
- Large Seebeck effect in HF + thermal regime (Itskovich-Kulik-Shekhter, Sov. JLTP 1985): asymmetry persists in SC states.
- Energy-dependent QP scattering (Anders & Gloos, Physica B 1997)
  - Explains both reduced signal & asymmetry, but unclear origins.
- Strongly energy-dependent DOS (Nowack & Klug, LT Phys. 1992)

Two-fluid picture of heavy fermions

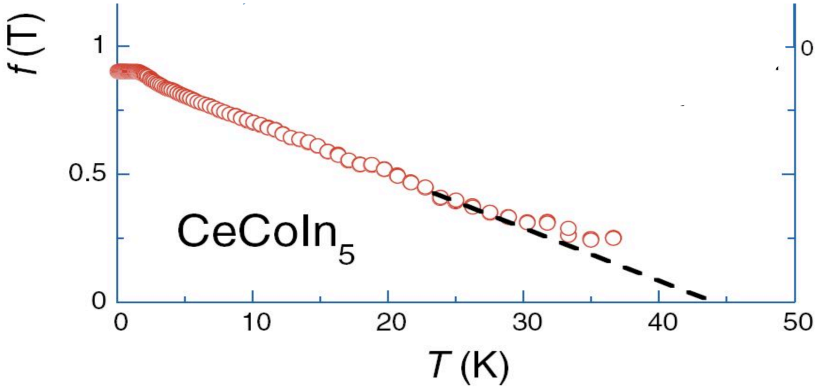


Petrovic *et al*. (2001)



Shishido *et al*. (2002)

- Emerging heavy fermions in Kondo lattice systems below a coherence temperature, *T* * (~ 45 K in CeCoIn$_5$).
- *f* (T) : relative weight of heavy-fermion liquid, increases with decreasing *T* and saturated below 2 K. Nakatsuji, Pines, Fisk, PRL **92**, 016401 (2004).

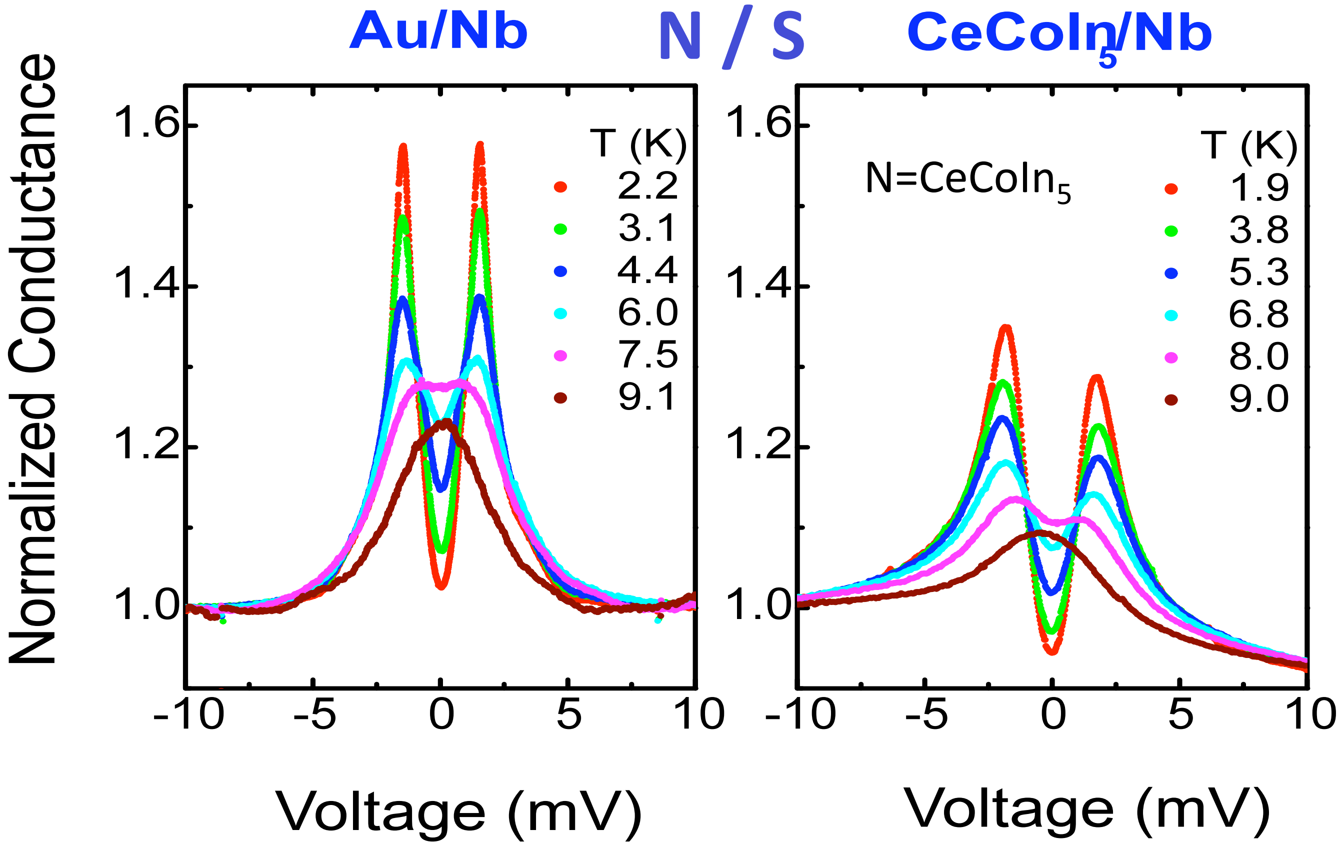


- This two-fluid picture appears valid in other heavy-fermion systems. Curro *et al*., PRB **70**, 235117 (2004).
- “Heavy electrons superconduct but light electrons don’t.” Tanatar *et el*., PRL **95**, 067002 (2005).

More support for 2-fluid model in $CeCoIn_5$

PCARTS for both N/S junctions of Au/Nb & $CeCoIn_5$/Nb are comparable, where there is no 2-fluid model for S Nb so all the Cooper pairs participate in the AR.

Recall for N/S Au/$CeCoIn_5$ is greatly reduced and we argue that "one of the 2 fluids does not participate in the AR"

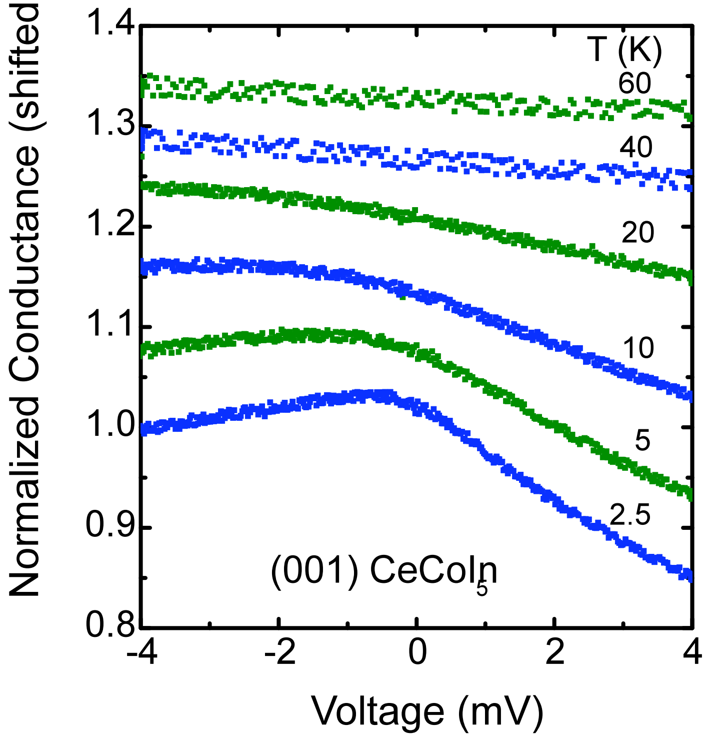


Asymmetry in Background Conductance modeled:

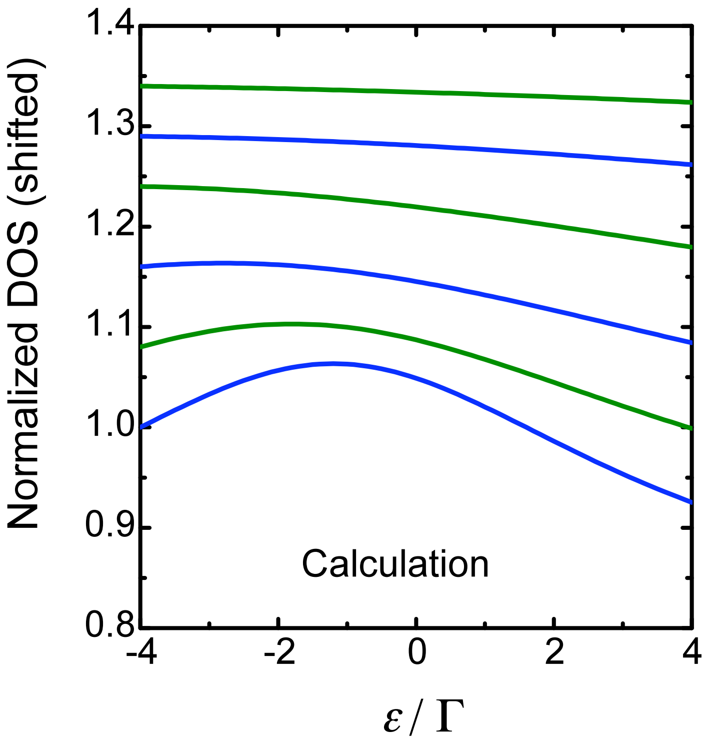
The temperature dependence of the Background Asymmetry follows that of the 2-fluid model.

To actually get the asymmetry, we model a peak in the DoS below E_F (reasonable for Kondo Lattices)

Data for T>Tc



Model with Lorentzian peak at -2.1 meV, HWHM=5meV



Two-channel Model Based on Lorentzian DOS

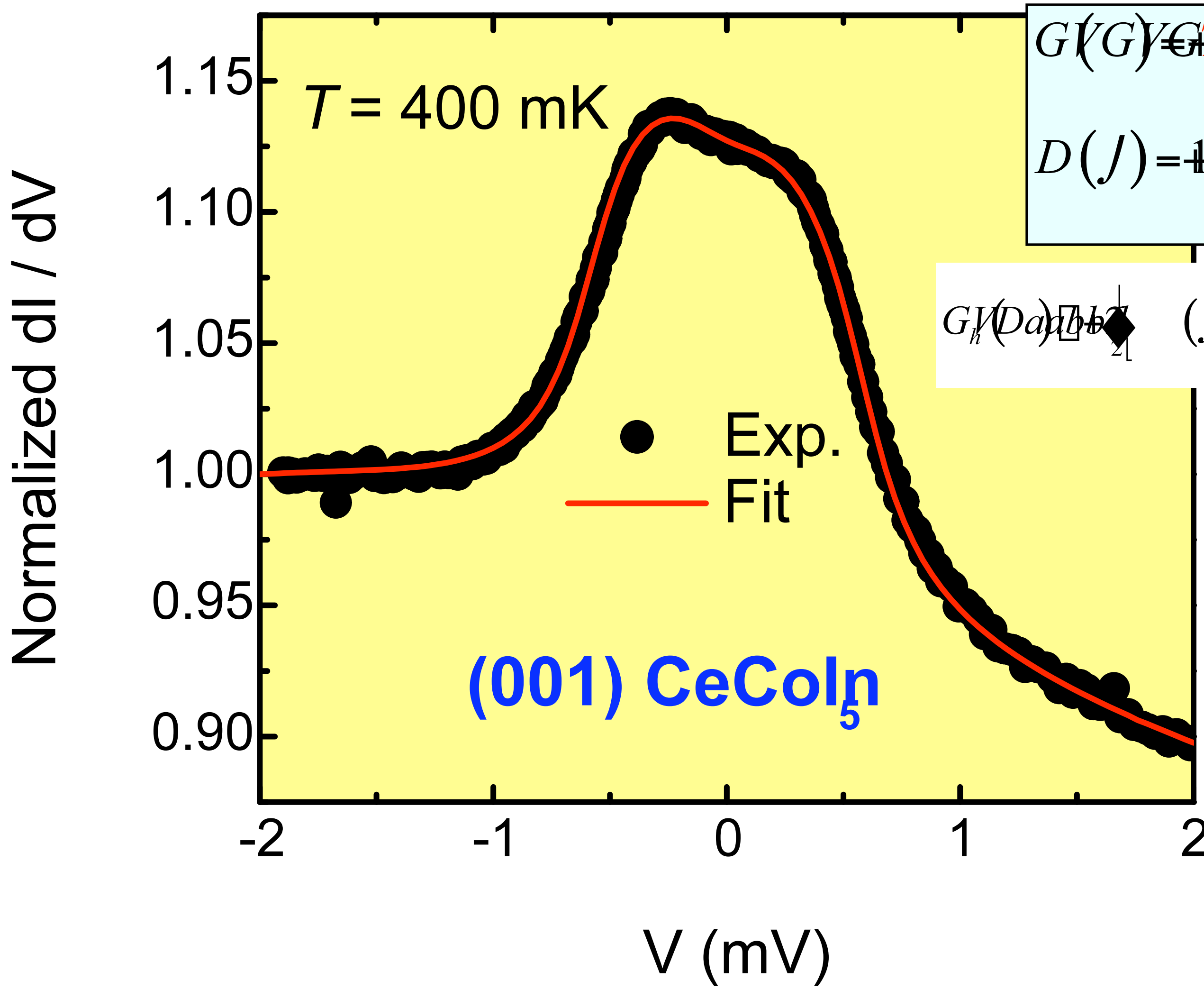
SC heavy el.

Non-SC light el.

$$G(V)=\omega_h G_h(V)+(1-\omega_h)G_l(V)$$

$$D(\varepsilon)=1+\sigma\frac{\Gamma^2}{\Gamma^2+2(\varepsilon-\varepsilon_0)^2}$$

$$G_h(V)\ldots(\varepsilon)\frac{\ldots(\varepsilon\ldots)}{\ldots(eV)}\ldots$$



fitting parameters

ω_h **= 0.51**

$\sigma = 1$, $\Gamma = 5$ meV

$\varepsilon_0 = -2.1$ meV

Z = 0.28

$\Gamma_{\text{Dynes}} = 95$ μeV

$\Delta = 600$ μeV

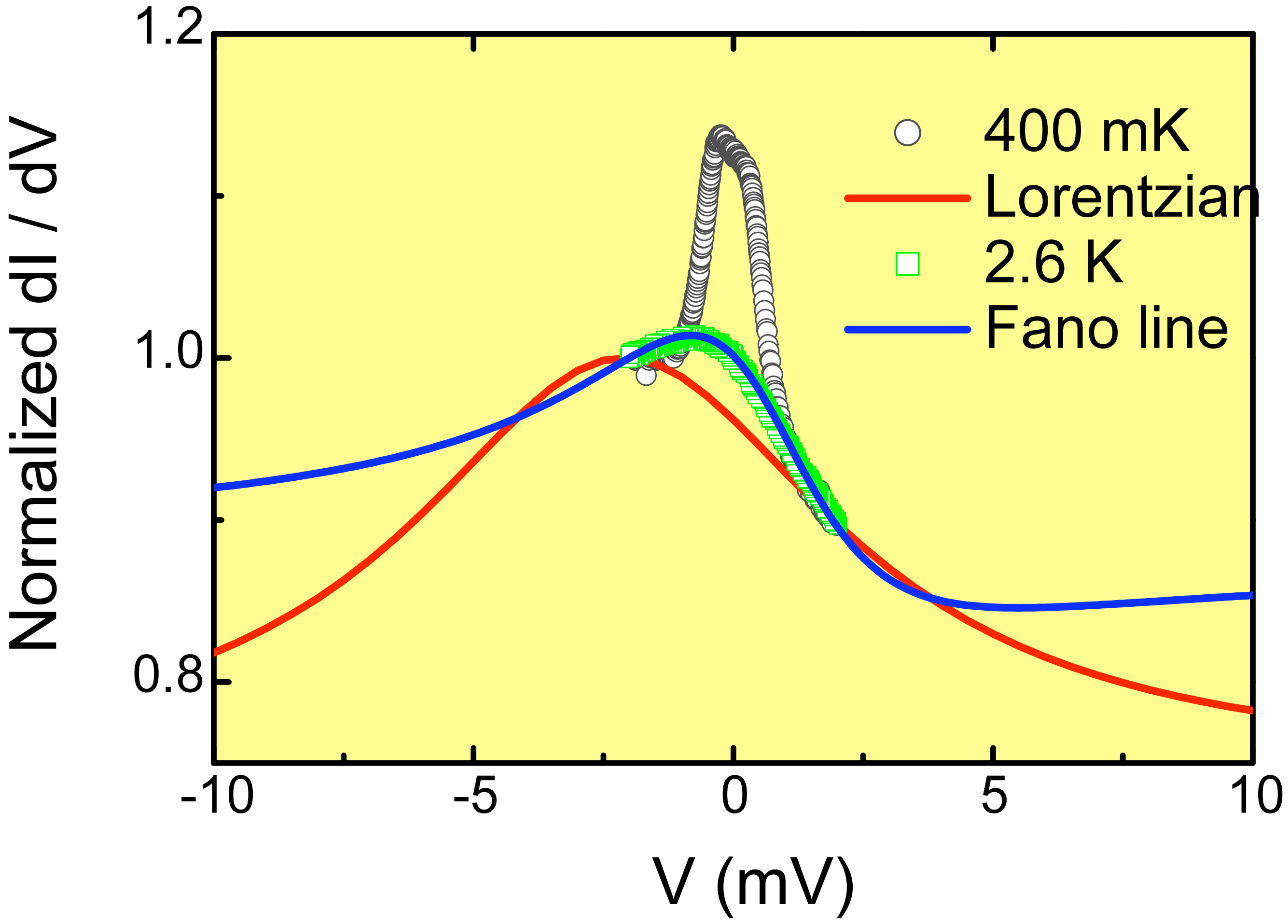
***cf.* 1-channel BTK fit**

$\Gamma_{\text{Dynes}} = 218$ μeV

$\Delta = 460$ μeV

WKP *et al.*, PRL **100**, 177001 (2008)

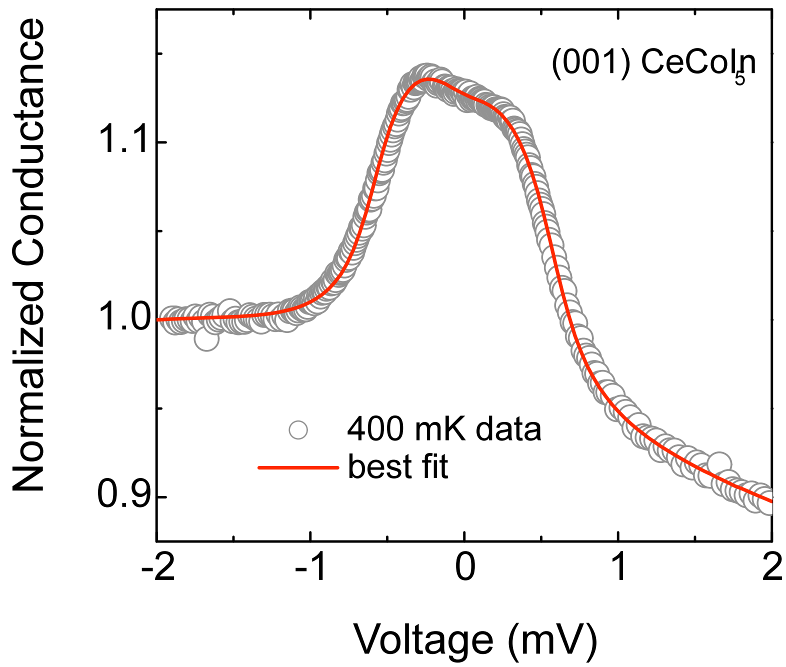
High-Temperature Deviation



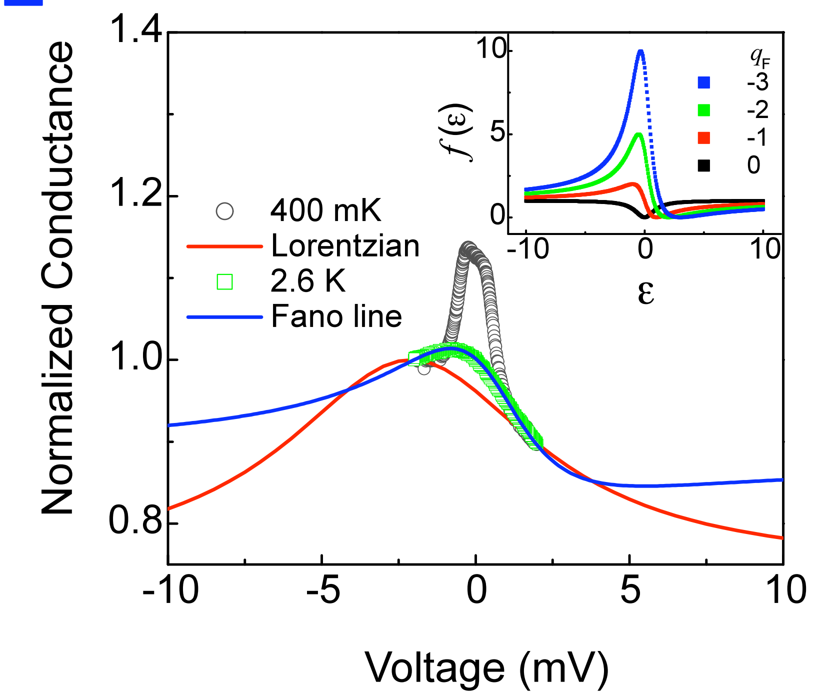
- Do not fit to a Lorentzian but to a Fano line-shape.

Model fits magnitude of AR, asymmetry and T-dep !

Data (circles) and fit
(red line) is excellent

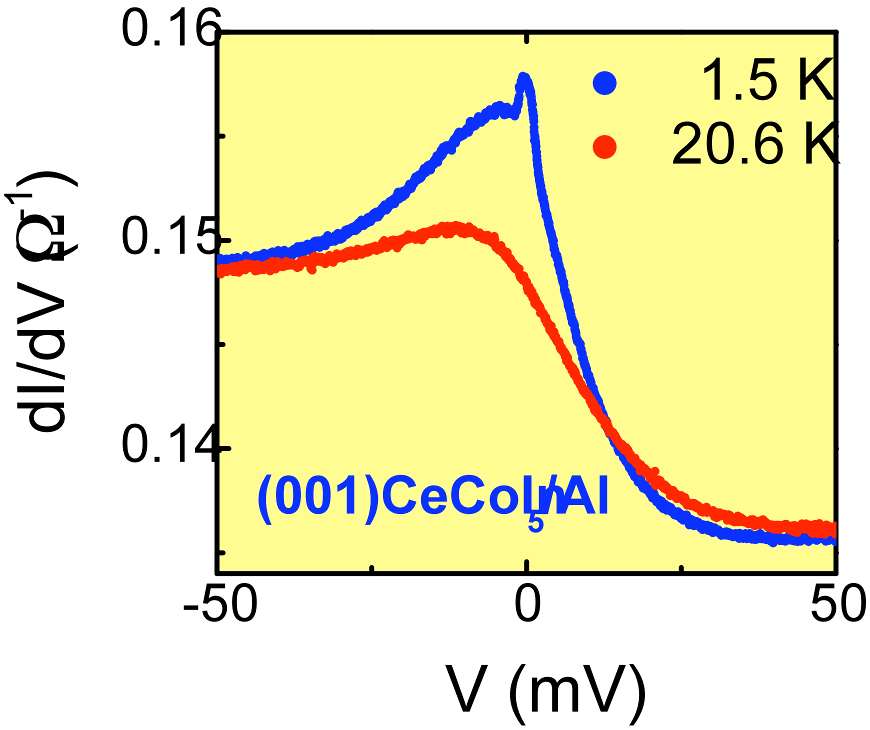


Best fit over wide T-range
with a **Fano lineshape**

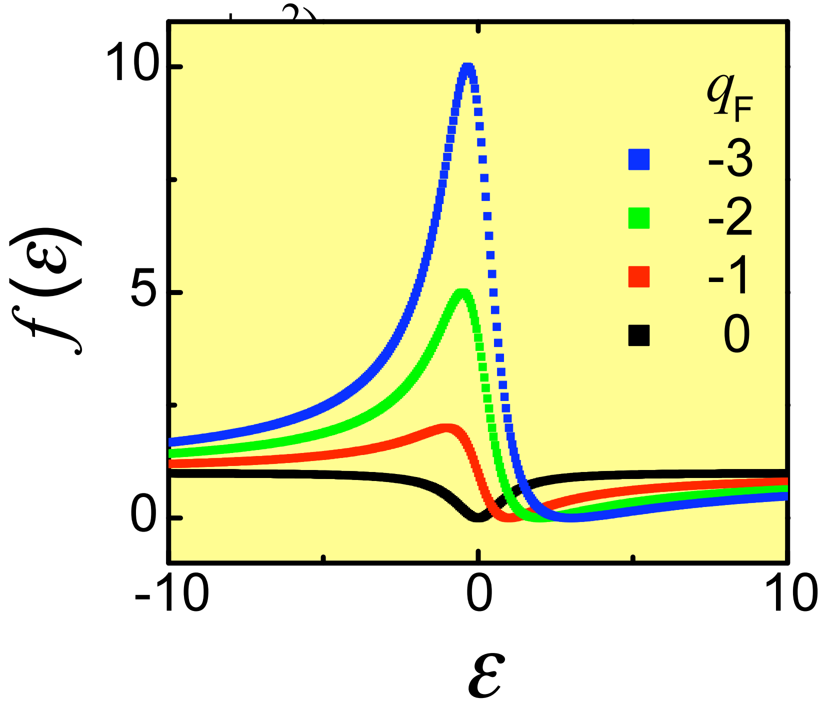


- Fit and consistency with other data: Measure DoS !?
- Fano may be explained by interference between f-electrons and conduction electrons via spin-flip (Kondo) scattering.

Fano Effect in Kondo Lattice?

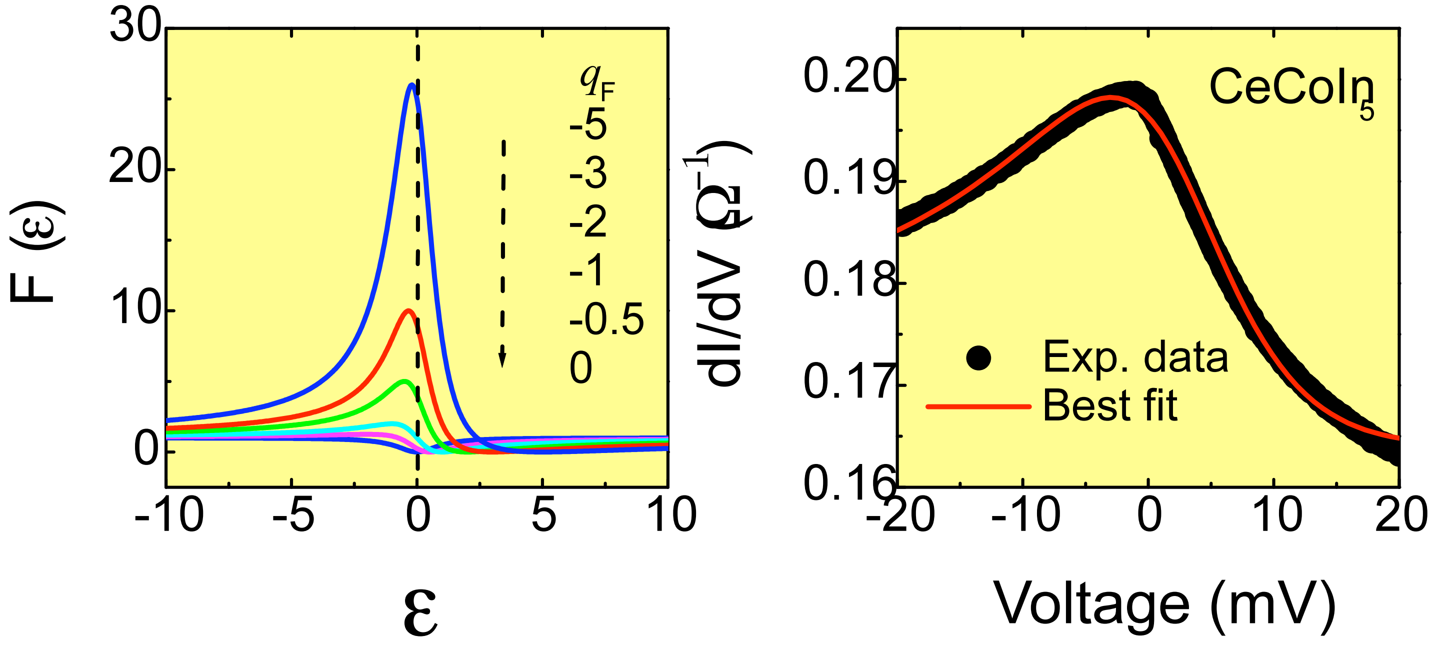


$f(e)=(q_F+e)^2/(1$



- Conjecture: Fano interference effect between two conduction channels: heavy-electron band and conduction electron band.
- Fano factor can have negative value (interference), and peak position below Fermi level can mean the Kondo resonance above Fermi level.
- Underlying microscopic picture is being investigated, which should provide valuable insight into the Kondo lattice physics.

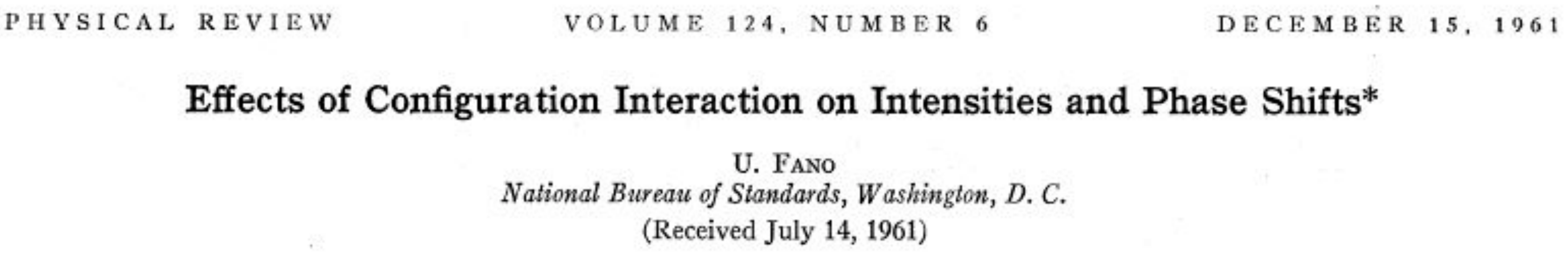
Conductance Model based on Fano Formula



$F(\varepsilon) = (q_F+\varepsilon)^2 / (1+\varepsilon^2)$, $\varepsilon \equiv (E-E_0)/(\Gamma/2)$, $dI/dV = C \cdot F(\varepsilon)+G_0$

- q_F=-2.14, E_0=2.23 meV, $\Gamma/2$=11.13 meV, C=0.0061 Ω^{-1}, G_0=0.164 Ω^{-1}
- negative q_F value - interference; positive E_0 - Kondo resonance above E_F; large G_0 - large portion is not involved in interference.
- Fano interference effect between two conduction channels, into heavy-electron band and conduction electron band.

Fano Resonance

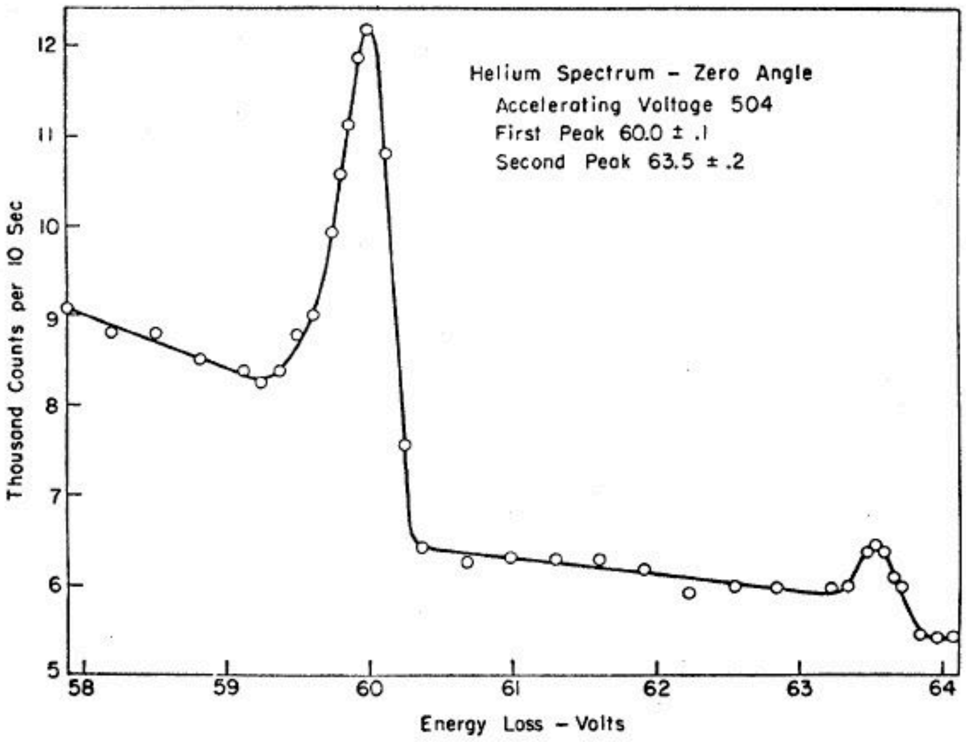


PHYSICAL REVIEW VOLUME 124, NUMBER 6 DECEMBER 15, 1961

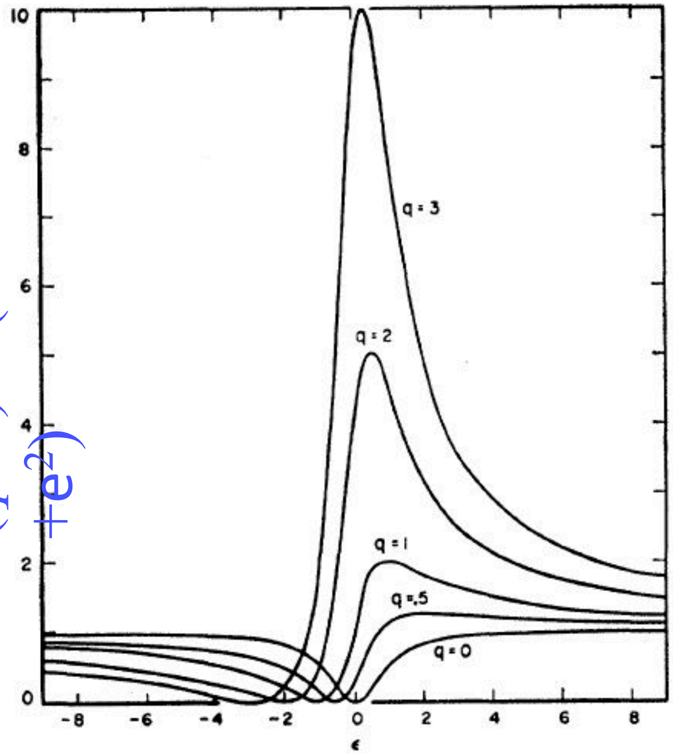
Effects of Configuration Interaction on Intensities and Phase Shifts*

U. FANO
National Bureau of Standards, Washington, D. C.
(Received July 14, 1961)

Electron-Helium inelastic scattering



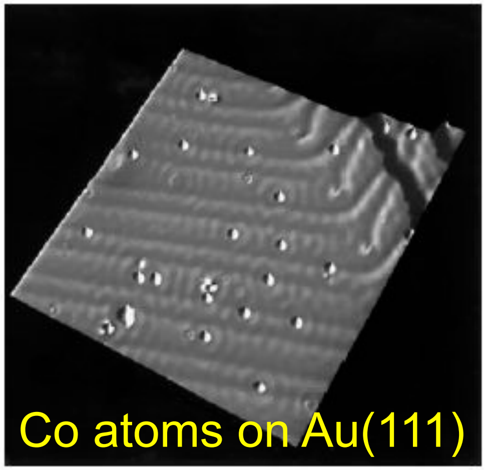
Probability ratio for transition to discrete and continuum



$(q+e)^2 / (1+e^2)$

e, relative to e_{res}

Fano / Kondo Resonance in Single Impurities



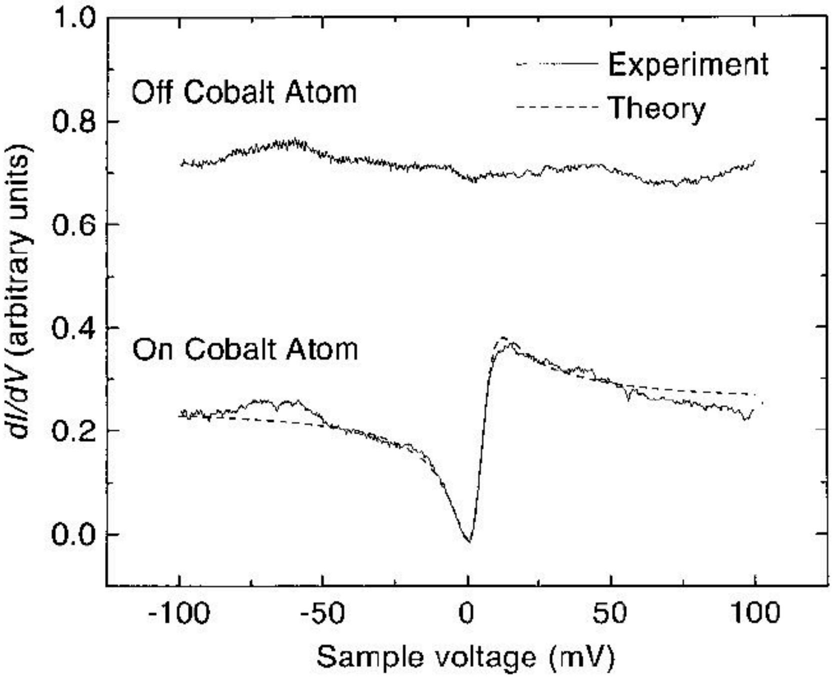
V. Madhavan et al., Science 280, 567 (1998)

$$\frac{dI}{dV}(V)=\frac{4e^2}{\hbar}\rho_{\text{tip}}\left[\pi\sum_k |\hat{M}_{tk}|^2\delta(eV-\varepsilon_k)\right]\frac{(\varepsilon'+q)^2}{1+\varepsilon'^2}+C$$

$$A(\varepsilon)=M_{at}+\sum_k M_{kt}V_{ak}P\left(\frac{1}{\varepsilon-\varepsilon_k}\right)$$

$$qe^{i\theta}=\frac{A}{B}$$

$$B(\varepsilon)=\pi\sum_k M_{kt}V_{ak}\delta(\varepsilon-\varepsilon_k).$$



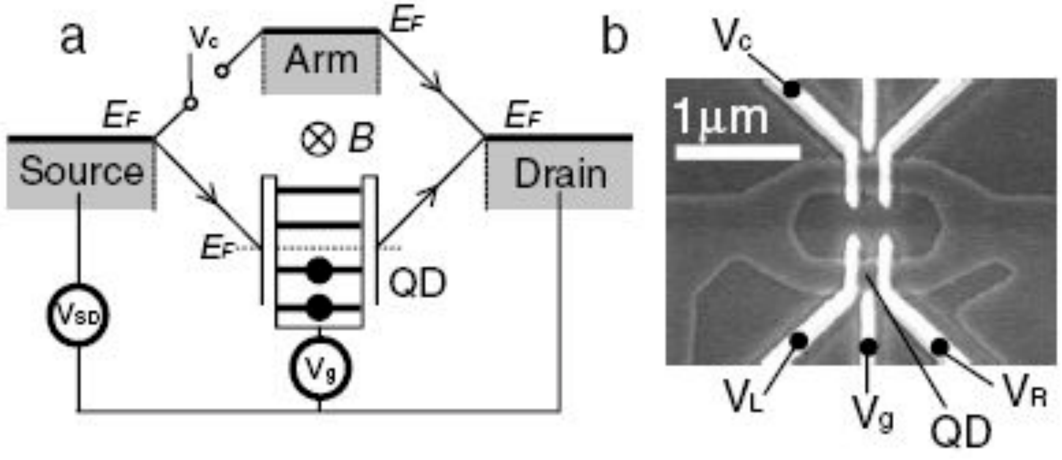
A: coupling to atomic orbital, direct or indirect via virtual transitions involving band electrons

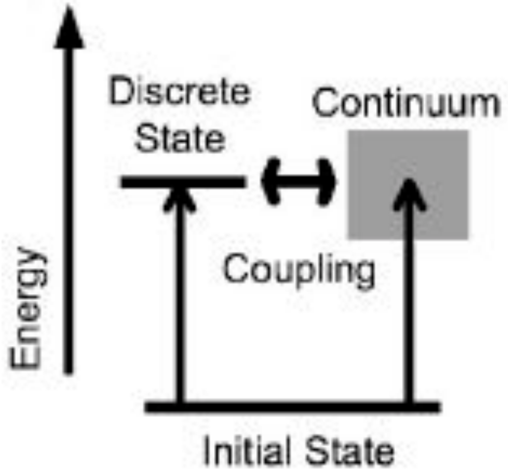
B: coupling to conduction electron continuum

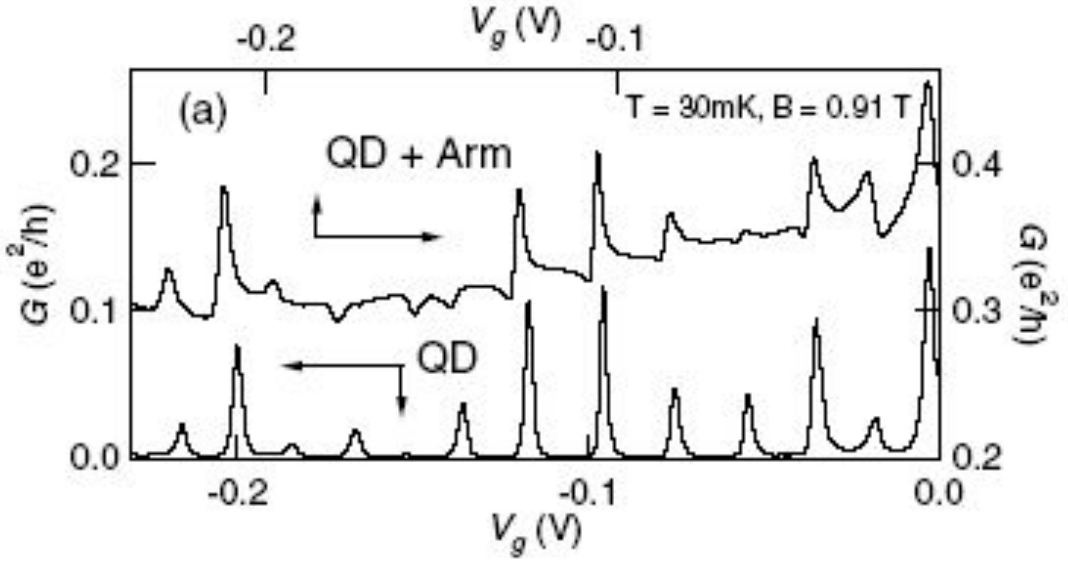
Other groups: Schneider, Eigler, Lieber, Kern, Zhao, Berndt, …

Fano Resonance in Quantum Dots

K. Kobayashi et al., PRL 88, 256806 (2002)



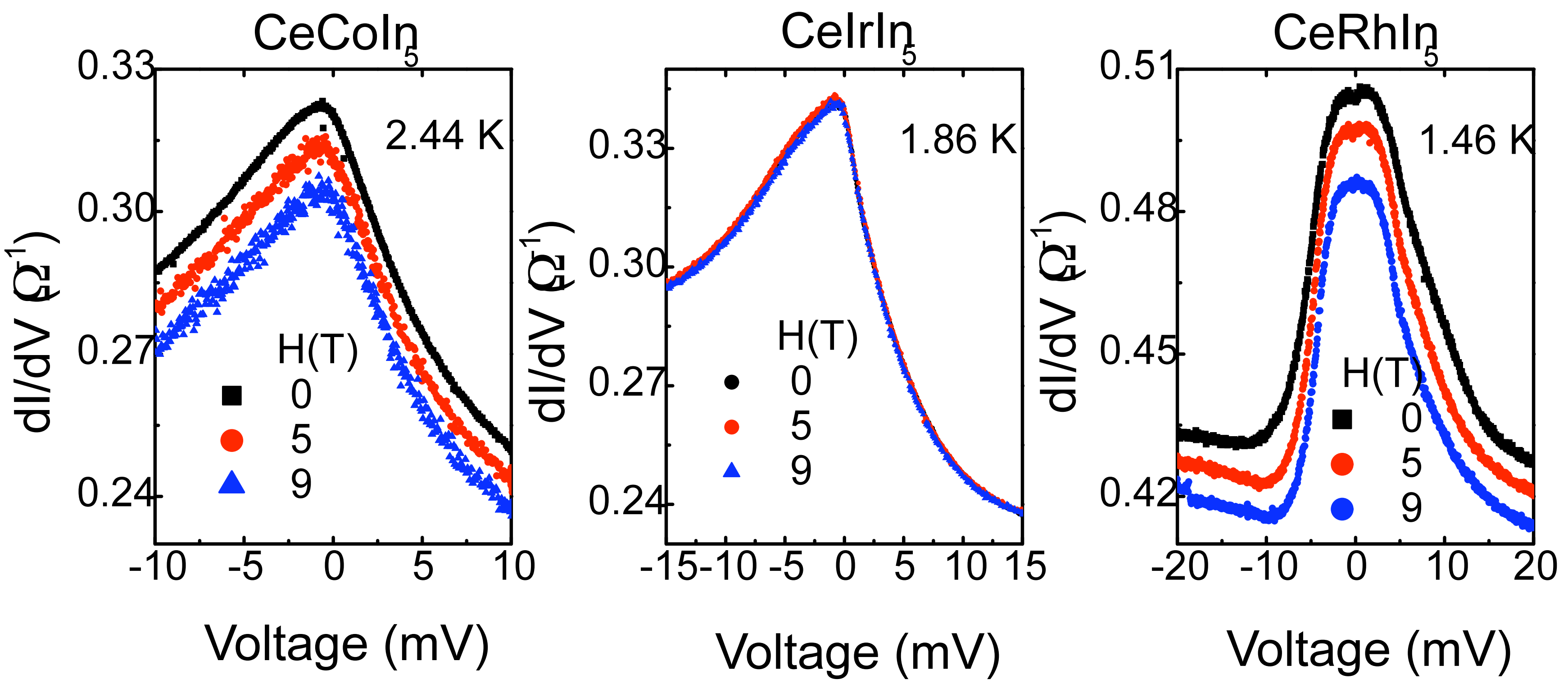




"The Fano effect is essentially a single-impurity problem describing how a localized state embedded in the continuum acquires itinerancy over the system."

Magnetic Field Dependence

H // *ab*-plane



- Conductance asymmetry: nearly independent of magnetic field.
- Only very little magnetoconductance

Comments / Summary

- The 115 family are unusually clean among heavy fermion materials, making it highly feasible to do PCS on them.

- The conductance spectra show a clearest example of Andreev reflection in heavy fermions: directional measurements nail down the order parameter symmetry.

- Interpretation of the conductance asymmetry as a manifestation of Fano resonance in this Kondo lattice systems is a new approach: contains interesting implications for the understanding of how the system evolves as a function of temperature.

- Several relevant issues have been indentified, delineating what is understood and what needs further investigation.

Conclusions

- ☐ **Strength of the PCARS method**
  - First spectroscopic demonstration of d_{x2-y2} symmetry in $CeCoIn_5$
  - Density of states effects measured! (energy-dependent DoS; peak)
- ☐ **Kondo Lattice Properties:**
  - Two-fluid model
  - Energy-dependent DoS given by a Fano resonance possibly due to the interference of the f-electrons with the conduction electrons.