Electron Dephasing and Energy Exchange in Diffusive Metal Wires:

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Prologue

Resistivity of metals

High T, phonons

Low T, impurities & disorder

dR/dT > 0 always

De Haas & de Boer, 1934
But $\frac{dR}{dT} < 0$ in some samples!

De Haas, de Boer, & van den Berg, 1934
Suspect magnetic impurities

Fe in Cu:

J.P. Franck, Manchester, Martin (1961)

But how do they work?

FIGURE 3. The electrical resistance of dilute copper-iron alloys. The bars indicate the point of minimum resistance. The points shown □ were taken after re-annealing the 0-1% alloy.
The solution

The s-d exchange model: \( H = \sum_i J \vec{s}_i \cdot \vec{S} \)

Kondo’s result: \( \delta\rho \propto -B \log\left( \frac{T}{T_K} \right) \Rightarrow \frac{d\rho}{dT} < 0 \) !!

Kondo temperature: \( k_B T_K \approx E_F \left( e^{-\frac{1}{\nu J}} \right) \)

\( \nu \) = density of states at \( E_F \)
Moral of the story: magnetic impurities dominate the low-temperature resistivity of metals, even at concentrations as low as 0.01%.
Jump ahead 20 years … 1980’s

Matthiessen’s rule:

\[
\frac{1}{\tau} = \frac{1}{\tau_{\text{disorder}}} + \frac{1}{\tau_{\text{el-ph}}} + \ldots
\]

Wrong!

\[
\rho = \frac{m}{ne^2} \frac{1}{\tau}
\]

Elastics preserve quantum phase coherence
destructs quantum phase coherence

Electrons maintain quantum phase coherence over distance \( L_0 \gg l_e \)

Low T:

\[
\frac{1}{\tau_{\text{inelastic}}} \ll \frac{1}{\tau_{\text{elastic}}}
\]
Electron transport in diffusive regime

1. Elastic scattering (film boundaries, impurities) \( l_e = v_F \tau_e \)
   - diffusive states \( D = \frac{1}{3} v_F l_e \)

2. Inelastic scattering (phonons, other electrons, spins)
   - loss of phase coherence \( L_\phi = \sqrt{D \tau_\phi} \)
   - energy exchange between electrons
Why Is the Phase Coherence Time Important?

• $\tau_\phi$ limits quantum transport phenomena:
  – normal metals: weak localization, UCF, Aharonov-Bohm
  – superconductors: proximity & Josephson effects

• Localization theory assumes $L_\phi > \xi$
  – no M-I transition if $\tau_\phi$ saturates at low temperature

• example of quantum system coupled to environment
Predictions for $\tau_\phi$ at low $T$

(Altshuler & Aronov, 1979)

At low $T$, $\tau_\phi$ limited by e-e interactions

Screening depends on dimensionality

(At energy $E$, compare $\sqrt{\hbar D/E}$ with transverse dimensions)

$\tau_\phi$ depends on dimensionality

$\ll$ wires $\gg$ (1d regime): $L_\phi = \sqrt{D\tau_\phi} >$ transverse dimensions

($E \sim \hbar / \tau_\phi$ rule the game)
$\tau_{\phi}(T)$ in wires

(Altshuler, Aronov, Khmelnitskii, 1982)

$\tau_{\phi} = \left( A T^{2/3} + B T^3 \right)^{-1}$

$A = \frac{1}{\hbar} \left( \frac{\pi k_B^2}{4v_F L w t} \frac{R}{R_K} \right)^{1/3}$

Screened Coulomb interaction at d=1
Temperature dependence of $\tau_\phi$
confirmation of AAK theory in quasi-1D

Echternach, Gershenson, Bozler, Bogdanov & Nilsson, PRB 48, 11516 (1993)
The Experimental Controversy
Mohanty, Jariwala and Webb, PRL 78, 3366 (1997)

Saturation of $\tau_\phi$:
Artifact of measurement? If not, is it intrinsic?
Measuring $\tau_\phi(T)$

Interference of time-reversed paths $\Rightarrow$ “weak-localization” correction to $R$

$B$ reduces weak-loc. correction

$B \sim 0.25 \text{ mm}$

$\tau_\phi = (D\tau_\phi)^{1/2}$
Measuring $\tau_\phi(T)$ : raw data

$\Delta R/R$

$\Delta R$ grows as $T$ decreases

$\Delta R$ saturates below ~ 100mK

$5N = 99.999 \%$ source purity

$6N = 99.9999 \%$ source purity

1 ppm of impurities:

100 atoms ~ 25 nm

Ag(6N) & Au(6N):

Ag(5N) & Cu(6N):
\( \tau_\phi(T) \) in Ag, Au & Cu wires

Saturation of \( \tau_\phi \) is sample dependent

- Ag 6N, Au 6N → agreement with AAK theory
- Ag 5N, Cu 6N → saturation of \( \tau_\phi(T) \)

Low T behavior vs. Purity:

- 5N = 99.999 % source material purity
- 6N = 99.9999 %

Saturation of \( \tau_\phi \) is sample dependent
Quantitative comparison with AAK theory for clean samples

\[ \tau_\phi = \left( A T^{2/3} + B T^3 \right)^{-1} \]

<table>
<thead>
<tr>
<th>Sample</th>
<th>( A_{thy} ) (ns(^{-1}) K(^{-2/3}))</th>
<th>( A ) (ns(^{-1}) K(^{-2/3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag(6N)a</td>
<td>0.55</td>
<td>0.73</td>
</tr>
<tr>
<td>Ag(6N)b</td>
<td>0.51</td>
<td>0.59</td>
</tr>
<tr>
<td>Ag(6N)c</td>
<td>0.31</td>
<td>0.37</td>
</tr>
<tr>
<td>Ag(6N)d</td>
<td>0.47</td>
<td>0.56</td>
</tr>
<tr>
<td>Au(6N)</td>
<td>0.40</td>
<td>0.67</td>
</tr>
</tbody>
</table>

F. Pierre et al.,
PRB 68, 0854213 (2003)

\[ A_{thy} = \frac{1}{\hbar} \left( \frac{\pi k_B^2}{4v_F Lw \sqrt{R}} \right)^{1/3} \]
Investigation of inelastic processes

1st method: \( \tau_\phi \)

Another process dominates in not-so-pure samples?

2nd method: measure energy exchange rates

Distribution \( f(E) \) reflects the exchange rates
Background: Shot noise in diffusive metal wires

Steinbach, Martinis and Devoret, PRL 76, 3806 (1996)

Theory:

Nagaev 1992, 1995
Kozub & Rudin, 1995

\[ S_I = \frac{4}{RL} \int \int dx dE \ f(x, E)(1 - f(x, E)) \]

What does \( f(x, E) \) look like?
Distribution function -- textbook case
(no shot noise)

\[ \tau_D = \frac{L^2}{D} \]

Assumes complete thermalization -- \( t_D \gg t_{\text{electron-phonon}} \)

Never true in mesoscopic metal samples at low T!
Distribution function for $\tau_D \ll \tau_{\text{electron-phonon}}$

$f(x,E)$ shaped by energy exchange

$\tau_D = \frac{L^2}{D}$

$\tau_D \ll \tau_{\text{interaction}}$

$\tau_D \gg \tau_{\text{interaction}}$

free electrons

“hot” electrons
Aside 1: Current through a tunnel junction

$$I = e \left( \Gamma_→ - \Gamma_← \right)$$

$$\Gamma_→ = \frac{2\pi v_F^2}{\hbar} \int dE \left| \langle M \rangle \right|^2 n_L(E) n_R(E+eV) f_L(E) (1 - f_R(E+eV))$$

$$\Gamma_← = (1 - f_L(E)) f_R(E+eV)$$

$$I = \frac{1}{eR_T} \int dE n_L(E) n_R(E+eV) \times (f_L(E) - f_R(E+eV))$$

NN junction:
$$n(E) = 1 \quad f(E) = \overline{T}$$

$$\Rightarrow I = \frac{V}{R_T}$$
Conductance of an N-X junction at $T=0$

\[
l = \frac{1}{eR_T} \int dE \ n_L(E) n_R(E + eV) \\
\quad \times \left( f_L(E) - f_R(E + eV) \right) \\
= \frac{1}{eR_T} \int_{-eV}^{0} dE \ n_X(E + eV) \\
= \frac{1}{eR_T} \int_{0}^{eV} dE \ n_X(E)
\]

\[
\frac{dl}{dV} = \frac{1}{R_T} n_X(eV)
\]

Spectroscopy of $n_X$
How to measure $f(E)$:
tunnel spectroscopy using an N-S junction

$E, \Delta$
N out of equilibrium: spectroscopy of \( f(E) \)

\[
I = \frac{1}{eR_T} \int dE \ n_S(E) \left[ f_N(E-eV) - f_S(E) \right]
\]

\[
\frac{dI}{dV} = \frac{-1}{R_T} \int dE \ n_S(E) f'_N(E-eV)
\]
Experimental setup

L = 5 to 40 µm

Diffusion time: \( \tau_D = \frac{L^2}{D} = 1 \) to 60 ns

\[
\frac{dI}{dV}(V) \xrightarrow{\text{numerical deconvolution}} f(E)
\]
Summarize how to measure $f(E)$:

- **U=0 mV**
  - The plots show $f(E)$ and $dI/dV$ vs. $E$ (meV) for $U=0$ mV.

- **U=0.2 mV**
  - The plots show $f(E)$ and $dI/dV$ vs. $E$ (meV) for $U=0.2$ mV.
Effect of the diffusion time $\tau_D$ on $f(E)$

longer interaction time $\Rightarrow$ more rounding

H. Pothier et al., PRL 79, 3490 (1997)
Compare strength of interactions

- Ag 6N (99.9999%): $\tau_D = 1.8$ ns

- Cu 5N (99.999%): $\tau_D = 2.8$ ns

- Au 4N (99.99%): $\tau_D = 1.8$ ns

Effect of material? Effect of purity?
Compare Dependence on U

U=0.1, 0.2, 0.3 & 0.4 mV

Observe scaling law in **Au 4N** & **Cu 5N** but **not** in **Ag 6N**
Calculation of $f(x,E)$


$$\frac{\partial f(E)}{\partial t} + D \frac{\partial^2 f(E)}{\partial x^2} = l_{in}(x,E,f) - l_{out}(x,E,f)$$

Boundary conditions:

$$f_{x=0}(E) = f_{x=L}(E) = \text{Fermi function}$$
Calculation of $f(x,E)$


$$D \frac{\partial^2 f(E)}{\partial x^2} = l_{\text{in}}(x,E,\{f\}) - l_{\text{out}}(x,E,\{f\})$$

e-e interactions:

$$l_{\text{out}}(x,E,\{f\}) = \int dE' d\varepsilon K(\varepsilon) f(E) \left[1 - f(E - \varepsilon)\right] f(E') \left[1 - f(E' + \varepsilon)\right]$$

(Altshuler, Aronov, Khmelnitskii, 1982)
Theory of screened Coulomb interaction in the diffusive regime

(Altshuler & Aronov, 1979)

Prediction for 1D wire:

\[ K(\varepsilon) = \frac{K}{\varepsilon^{3/2}} \left( \propto \int \frac{dq}{D^2 q^4 + \omega^2} \right) \]

\[ \kappa = \left( \sqrt{2D} \pi \hbar^{3/2} \nu_F S_e \right)^{-1} \]
Experiment vs. Theory

Ag 6N:
experiment agrees with theory

Cu 5N, Au 4N, Ag 5N:
• energy exchange stronger than predicted
• \( K(\varepsilon) = \kappa \varepsilon^{-2} \) fits data
Comparison of the results of the two methods

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<tr>
<th></th>
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<th>$f(E)$</th>
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<td>$\text{Ag}_6\text{N}$</td>
<td>$\propto T^{-2/3}$</td>
<td></td>
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<tr>
<td>$\text{Ag}<em>{5\text{N}}\text{Cu}</em>{6\text{N},5\text{N},4\text{N}}\text{Au}_{4\text{N}}$</td>
<td>saturation</td>
<td></td>
</tr>
<tr>
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<td></td>
<td>$K(\varepsilon) = \frac{\kappa}{\varepsilon^{3/2}}$</td>
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$K(\varepsilon) \propto \frac{1}{\varepsilon^{3/2}}$
Comparison of the results of the two methods

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<td>Coulomb interactions</td>
</tr>
<tr>
<td>$\text{Ag}_5\text{N}$, $\text{Cu}_6\text{N,5N,4N}$, $\text{Au}_4\text{N}$</td>
<td>saturation</td>
<td>Other mechanism</td>
</tr>
<tr>
<td></td>
<td>$\propto$</td>
<td>fast relaxation rates</td>
</tr>
<tr>
<td></td>
<td>$K(\varepsilon) = \frac{\kappa}{\varepsilon^{3/2}}$</td>
<td>$K(\varepsilon) \propto \frac{1}{\varepsilon^{3/2}}$</td>
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</tbody>
</table>

ROLE OF RESIDUAL IMPURITIES?
The two puzzles

\( \tau_\phi(T) \) measurements

\[ \begin{array}{cccc}
\text{Ag 6N} & \text{Au 6N} & \text{Ag 5N} & \text{Cu 6N} \\
\tau_\phi \text{ (ns)} & 0.3mV & 0.2mV & 0.1mV \\
\end{array} \]

\( f(E) \) measurements

Anomalous interactions in the less pure samples
The Kondo effect again

Collective effect:

Formation of a singlet spin state

\[ k_B T_K \approx E_F e^{-1/\nu J} \]

Spin-flip scattering

\[ J \sigma \cdot \bar{S} \]

\( \Rightarrow \) increased resistivity
\( \Rightarrow \) reduction of \( \tau_\phi \)

\[ R \] (total scattering rate)

\[ \gamma_{sf} \]

\[ T_K \]

Kondo phonons

\[ \gamma_{sf} \]

spin-flip
Nagaoka-Suhl expression of the spin-flip scattering rate near $T_K$

$$\tau_{sf}^{-1} = \frac{1}{\tau_{sf}} = \frac{c_{\text{mag}}}{c_{\text{imp}}} \frac{\pi^2 S(S+1)}{\pi \hbar v_F \left( \pi^2 S(S+1) + \ln^2 \left( \frac{T}{T_K} \right) \right)}$$

Weak temperature dependance near $T_K$!!

$\text{Link to } \tau_{\phi}(T) \text{ saturation?}$
From $\tau_{sf}$ to $\tau_{\phi}$

Another important timescale: $\tau_K$

Lifetime of the spin state of a magn. imp.

( V.I. Fal'ko, JETP Lett. 53, 340 (1991) )

If $\tau_K < \tau_{sf}$

Other electrons matter

Randomising effect

$$\frac{1}{\tau_{\phi}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{e-ph}} + \frac{1}{\tau_{sf}}$$

If $\tau_K > \tau_{sf}$

The spin states of the mag. imp. seen by time-reversed electrons are correlated

$$\frac{1}{\tau_{\phi}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{e-ph}} + \frac{2}{\tau_{sf}}$$
Comparison of $\tau_{sf}$ and $\tau_K$

$k_B T$ $E_F$ $\nu_F k_B T$ concentration of electrons that can spin-flip

$C_{imp}$ concentration of magnetic impurities

If $\nu_F k_B T > c_{imp}$ then $\tau_K < \tau_{sf}$

$\frac{1}{\tau_\phi} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{e-ph}} + \frac{1}{\tau_{sf}}$

Numerically, for Au, Ag, Cu, ...

$T > 40 \text{ mK} \times c_{imp} \text{ (ppm)}$
Effect of magnetic impurities on $\tau_\phi$

$$\tau_\phi = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{e-ph}} + \frac{1}{\tau_{sf}}$$

Spin-flip rate peaks at $T_K$:

$$\tau_\phi(T_K) = \frac{0.6 \text{ ns}}{c_{\text{imp}} \text{ (ppm)}}$$
Effect of magnetic impurities on $\tau_\phi$

F. Pierre et al., PRB 68, 0854213 (2003)

Fit parameters:
Ag(5N) bare: 0.13 ppm
+ 0.3 ppm : 0.40 ppm
+ 1 ppm : 0.96 ppm

Above $T_K$ : partial compensation of e-e and s-f

apparent saturation
Why can’t we just detect magnetic impurities with $R(T)$ (the original Kondo effect)?

1 ppm of Mn is invisible in $R(T)$ (hidden by e-e interactions)
Source material purity vs. sample purity: Cu samples

- In all Cu samples $\tau_\phi(T)$ saturates at low $T$.
- $\tau_\phi(T)$ is strongly reduced but shows no dip.

Magnetic impurities are invisible in R(T).
Measure $\tau_\phi(B)$ from Aharonov-Bohm oscillations

$\delta G (e^2/h)$ vs $B$ (T)

$T=100$ mK

Cu ring
AB oscillations increase with B
⇒ presence of magnetic “impurities”!
What about energy exchange?
Energy exchange mediated by magnetic impurities
Kaminski and Glazman, PRL 86, 2400 (2001)

Virtual state

Reinforced by Kondo effect
Energy exchange mediated by magnetic impurities vanishes when $g\mu_B B >> eU$
Aside 2: Inelastic tunneling

initial state:

final state:
P(E) = probability to give energy E to environment
\( \text{P}(E) \) depends on environmental impedance

\[
\begin{align*}
\frac{dl}{dV} &= \frac{1}{R_t} \int_0^{\text{e}_V} \text{P}(E) \, dE \\
\text{P}(E) &= \frac{1}{2\pi\hbar} \int e^{iEt/\hbar + J(t)} \, dt \\
J(t) &= 2\int_0^{+\infty} \frac{d\omega}{\omega} \operatorname{Re}[Z(\omega)] \frac{1}{R_K} (e^{-i\omega t} - 1)
\end{align*}
\]

At \( T=0 \), one obtains:
Resistive environment

For \( eV < \frac{\hbar}{RC} \),

\[
\frac{dl}{dV} \propto \left( V^{\frac{2R}{R_K}} + \text{cst.} \right)
\]

\( C = 0.92 \text{fF} \)

\( T = 40 \text{ mK} \)

\( R/R_K = 0.08 \) (\( R = 2.06 \text{k}\Omega \))
Measure $f(E)$ at $B \neq 0$ using Dynamical Coulomb Blockade

d$I$/d$V$ $\rightarrow$ $f(E)$ $\rightarrow$ electron-electron interactions
A controlled experiment

Effect of 1 ppm Mn on interactions?
Experimental data at weak B

\[ U = 0.1 \text{ mV} \]
\[ B = 0.3 \text{ T} \]
Experimental data at weak and at strong B

U = 0.1 mV
B = 0.3 T
B = 2.1 T

Very weak interaction

implanted

bare
Coherence time measurements on the same 2 samples

Fits:

$C_{\text{bare}} = 0.1$ ppm
$C_{\text{implanted}} = 0.9$ ppm
Full U,B dependence

Bare

C=0.1 ppm

U=0.1 mV

U=0.2 mV

U=0.3 mV

Implanted

C=0.9 ppm

2.1 T

1.8 T

1.5 T

1.2 T

0.9 T

0.6 T

0.3 T
Comparison with theory \( s = \frac{1}{2} \)

Goeppert, Galperin, Altshuler and Grabert, PRB 64, 033301 (2001)

**Implanted**

- C=0.9 ppm

**Bare**

- C=0.1 ppm

<table>
<thead>
<tr>
<th>U (mV)</th>
<th>V (mV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>-0.2</td>
</tr>
<tr>
<td>0.2</td>
<td>0.0</td>
</tr>
<tr>
<td>0.3</td>
<td>0.4</td>
</tr>
<tr>
<td>0.4</td>
<td>0.8</td>
</tr>
<tr>
<td>0.5</td>
<td>1.0</td>
</tr>
<tr>
<td>0.6</td>
<td>1.1</td>
</tr>
<tr>
<td>0.7</td>
<td>1.2</td>
</tr>
</tbody>
</table>

**2.1 T**

- 1.8 T
- 1.5 T
- 1.2 T
- 0.9 T
- 0.6 T
- 0.3 T
Moral of the story: even at concentrations as low as 1 ppm, magnetic impurities have a large influence on low-temperature electronic transport in metals.
Four consequences of electron-electron interactions in quasi-1D diffusive wires (Altshuler & Aronov)

- loss of phase coherence: $\tau_\phi \sim T^{-2/3}$ (AA+Khmelnitskii)
- energy exchange between quasiparticles:

$$\propto \varepsilon^{-3/2}$$

- correction to resistance: $\delta R(T) \sim T^{-1/2}$
- correction to tunneling DOS, or dynamic Coulomb blockade: $dI/dV \sim V^{2R/R_Q}$
References

• Dephasing

• Energy Exchange

• Both
Evidence for extremely dilute magnetic impurities even in purest samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Imp.</th>
<th>$T_K$(K)</th>
<th>c (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag(6N)a</td>
<td>Mn</td>
<td>0.04</td>
<td>0.009</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>&quot; b</td>
<td></td>
<td>0.011</td>
</tr>
<tr>
<td></td>
<td>&quot; c</td>
<td></td>
<td>0.0024</td>
</tr>
<tr>
<td></td>
<td>&quot; d</td>
<td></td>
<td>0.012</td>
</tr>
<tr>
<td>Au(6N)</td>
<td>Cr</td>
<td>0.01</td>
<td>0.02</td>
</tr>
</tbody>
</table>

1 ppm: In the wire, 0.01 ppm = 3 impurities/µm

100 atoms ~ 25 nm
Compare $\tau_\phi$ data with AAK and GZS theories