

Electron Dephasing Times in Disordered Metals

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Quantum Phenomena at Low Temperatures ULTI III Users Meeting Lammi Biological Station, January 2004 **Experimental Method**

Weak-localization magnetoresistances

Electron dephasing times as function of temperature



Outline

- Experimental electron dephasing times from weaklocalization studies: low-field magnetoresistance
- Electron-phonon scattering in disordered metals weakened or enhanced electron-phonon interaction static or vibrating defects (impurities)
- Very short dephasing lengths in some cases
- Saturation in electron dephasing time as T→ 0 magnetic-impurity scattering ? two-level systems ? electron-electron interactions ?

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The Problem

• To study the electron-phonon interaction in disordered metals



Short Mean Free Path and the Dirty Limit

 $\operatorname{Ti}_{100-x}\operatorname{Al}_x$



• Ti₇₃Al₂₇ has a very high resistivity

 \Rightarrow Suitable for 3D weak localization studies

- Sn-doped Ti₇₃Al₂₇ alloy possesses strong spin-orbit scattering
- $\Rightarrow \tau_{o}$ is the only fitting parameter

Mooij, Phys. Stat. Sol. (a) 17 (1973) 521



ho(10 K) \approx 225 $\mu\Omega$ cm $l \approx$ 2 Å (mean free path) $ql \approx$ 0.006 T << 1

- In these alloys, the electron-phonon interaction is well within the dirty limit
- The exponent of temperature for τ_{ep}^{-1} is $p \approx 2$

Electron-Phonon Time in Superconducting Ti₈₈Sn₂₂ Alloys



Jian, Lin (1996)

Electron-Phonon Interaction in Disordered Metals

• For many years, the electron-phonon interaction in impure conductors has been thought to be well understood

Pippard (1955); Schmid (1973, 1985, 1986); Sergeev, Reizer (1986); Belitz (1987)

Conventional wisdom:

• Impurity atoms (defects) move in phase with vibrating lattice atoms, resulting in long-wavelength phonons being unable to scatter electrons (the `Pippard ineffectiveness condition')

$$\tau_{ep}^{-1} \propto T^4 l \qquad (ql < 1)$$

Cf. weakened e-ph interaction, compared with the pure case: $\tau_{ep}^{-1} \propto T^3$ (ql > 1)

Pippard Ineffectiveness Condition

• Electrons having very short mean free path are not effective in scattering long wavelength phonons

$$l < \lambda_{ph} = \frac{2\pi}{q}$$
 or $ql < 1$

• In disordered metals, the electron wavenumber is subject to an uncertainty of

$$\Delta k \sim \frac{1}{l}$$

 \Rightarrow Effective scattering requires:

$$q > \Delta k \sim \frac{1}{l}$$
 or $ql > 1$

Cf. Kittel: Quantum Theory of Solids, Ziman: Electrons and Phonons

Experimental situation:

• A $\tau_{ep}^{-1} \propto T^2$ has been observed in metal films and narrow wires from time to time

 \Rightarrow But, basically, no systematic studies on both the temperature and mean free path dependences of τ_{ep}

Theoretical situation (up to 1999):

• "We have thus no evidence now indicating the existence of a new mechanism of phase relaxation which has heretofore not been analyzed theoretically and which could account for a function of the type $\tau_{\omega} \sim T^{-2}$."

> [Altshuler, Aronov, Gershenson, Sharvin, Sov. Sci. Rev., Sect. A 9 (1987) 223]

Theories seem to suggest that a T^2 dependence is not possible

Why 3D Superconducting Ti_{1-x}Al_x Alloys

- Arc-melted, single-phased alloys for $x \le 0.13$
- Resistivity (disorder) increases linearly with increasing x \Rightarrow to study disorder dependence of τ_{ep}
- Diffusion constant can be determined from H_{c2} measurement
- Fe, Co, Cr, and Ni do not form localized moments in a Ti host
- 3D mesoscopic samples: $(L_{\phi} < L_x, L_y, L_z)$

$$\tau_{\varphi}^{-1} = \tau_{ep}^{-1} + \tau_{ee}^{-1} + \tau_{s}^{-1} \approx \tau_{ep}^{-1}$$

(at not too low temperatures)



The resistivity increases linearly with increasing x

$Ti_{1-x}AI_x$ alloys with a wide range of disorder: Compositional Disorder

- The temperature dependence of τ_{ep} is difficult to measure
- The disorder dependence of τ_{ep} is even much more difficult to measure



Lin & Wu, Europhys. Lett. 29, 141 (1995)

Why non-superconducting Au₅₀Pd₅₀ thick films

- A prototypical disordered metal, by DC or RF sputtering
- Resistivity was "tuned" by adjusting the deposition rate
 ⇒ a wide range of electron mean free path
- Strong spin-orbit scattering $\Rightarrow \tau_{\varphi}$ is the sole fitting parameter
- Absence of superconductivity

 $\Rightarrow \tau_{\omega}(T \rightarrow 0)$ can be directly measured

$$\tau_{\varphi}^{-1} = \tau_{ep}^{-1} + \tau_{ee}^{-1} + (\tau_{\varphi}^{0})^{-1} \approx \tau_{ep}^{-1} + C$$
experimentally
measured



• The 3D weak-localization theoretical predictions (solid curves) can well describe the experimental data

Au₅₀Pd₅₀ Thick Films: Structural Disorder

- The temperature dependence of $\tau_{\rm ep}$ is difficult to measure
- The disorder dependence of τ_{ep} is even much more difficult to measure



 $\tau_{\varphi}^{-1} = \mathbf{C} + \tau_{ep}^{-1} = \mathbf{C} + \mathbf{AT}^{p} \implies \tau_{ep}^{-1} \propto \mathbf{T}^{2}l$

Zhong & Lin, Phys. Rev. Lett. 80, 588 (1998)







0.8

 $D (\text{cm}^2/\text{s})$

0.4

Zhong et al., Phys. Rev. B 66, 132202 (2002)

1.2

1.6



pp. 6041-6047

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Electron-phonon interaction in disordered conductors: Static and vibrating scattering potentials

A. Sergeev and V. Mitin Department of ECE, Wayne State University, Detroit, Michigan 48202 (Received 2 July 1999; revised manuscript received 23 September 1999)

See, also, Europhys. Lett. 51 (2000) 641

• Defects such as heavy (light) impurities and tough boundaries may not move in phase with deformed lattice atoms

⇒ static impurities result in *enhanced* e-ph interaction:

$$au_{ep}^{-1} \propto T^2 l^{-1}$$

Cf. vibrating impurities \Rightarrow standard result: $\tau_{ep}^{-1} \propto T^4 l$

Predictions of the Sergeev-Mitin theory

k = 1, if impurities move in phase with deformed lattice atoms *k* = 0, if impurities remain *completely static*



- The *T* dependence of τ_{ep}^{-1} can change from T^4 to T^2 The disorder dependence of τ_{ep}^{-1} can change from l to l^{-1}

Comparison with Previous Works

- 3D Granular films are not microscopically homogeneous
 ⇒ no disorder dependence was observed
- 3D amorphous metals are already in the limit of strong randomness ⇒ level of disorder cannot be "tuned"





microscopically homogeneous: Al atoms randomly substitute for Ti lattice sites $\frac{\text{microscopically inhomogeneous:}}{\text{metal or insulator grains of tens}} or hundreds \text{\AA}$

Comparison with Previous Works (continued)

In reduced dimensions:

many fitting parameters: $\tau_{in}^{-1} = \tau_{ep}^{-1} + \tau_{ee}^{-1} = AT^{p} + BT^{q}$ even worse: $\tau_{ep}^{-1} < \tau_{ee}^{-1}$

• Phonon dimensionality is not well defined, depending on the film thickness, phonon wavelength, acoustic transparency of the film-substrate interface, etc.



• We have shown that a *T*² dependence is often observed in real metals

• But, other power law is not impossible

⇒ Apart from the total level of disorder, the temperature and disorder dependences of τ_{ep} is very sensitive to the microscopic quality of the defects

Question:

Can one observe the T^4 dependence

Wu *et al.*, Phys. Rev. B 57, 11232 (1998)

Hafnium and Titanium thin films on sapphire substrates



• From thermal conductance measurement, a T^{-4} dependence was observed between 40-700 mK

• But, a weaker temperature dependence above 0.7 K, where $ql \approx 0.04 \ll 1$

Gershenson *et al.*, Appl. Phys. Lett. 79, 2049 (2001)



Gershenson *et al*., Appl. Phys. Lett. 79, 2049 (2001) Electrons and phonons at sub-Kelvin temperatures: validation of the disorder-mediated scattering theory

I. J. Maasilta, J. T. Karvonen, J. M. Kivioja^{*}, and L. J. Taskinen NanoScience Center, Department of Physics, P.O. Box 35, FIN-40014 University of Jyväskylä, Finland (Dated: October 4, 2003)

From Joule heating the electron gas and measuring both the electron and the lattice temperatures simultaneously, τ_{ep} is determined



- First observation of disordermediated e-ph scattering in Cu thin films: $\tau_{ep}^{-1} \sim T^4$
- Measuring temperatures:
 60–135 mK (900 Å)
 60–195 mK (450 Å)

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Electron-phonon heat transport and electronic thermal conductivity in heavily doped silicon-on-insulator film

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* Heavily doped Si is in the dirty limit at mK and the phonon system has a complete phonon drag

- The heat flow between electron and phonon systems has a T^6 dependence $\Rightarrow \tau_{ep}^{-1} \propto T^4$ for the e-ph interaction relaxation time
- Measuring temperature: 100-500 mK



The sample needs to have:

• *ql* < 1

- Contain little non-vibrating disorder
- Have 3D phonons coupled to electrons by the deformation potential

The Heat flow from electrons to phonons:

The change of electron temperature is determined by:

 $dP = C_e dT_e / \tau_{ep}$ (*C_e*: electronic heat capacity)

By substituting $\tau_{ep}^{-1} = \alpha T^{p} \Rightarrow P = \Sigma \Omega (T_e^{p+2} - T_{ph}^{p+2})$

 Σ : material-dependent electron-phonon coupling constant Ω : the volume of the sample

Yuan-Liang Zhong Chiidong Chen Juhn-Jong Lin



Supported and free-standing films and wires for studies of τ_{ep}

Samples were prepared by *e*-beam lithography technique

 \Rightarrow e.g., phonon confinement effect







Very Short Electron Dephasing Lengths

 The dephasing length in dirty multi-wall carbon nanotubes, determined from weak-localization studies, is only ~ 10 nm



Low Temperature Laboratory/HUT

Very Short Dephasing Length: Cu-SiO₂ Nano-Granular Films

• Cu_x -(SiO₂)_{1-x} films were prepared by co-sputtering on glass substrates, 1 µm thick



as-sputtered: ~ 1-3 nm

annealed: ~ 10 nm

Cu_x-(SiO2)_{1-x} nano-granular films

The dephasing length is only ~ tens nm near the quantum percolation threshold



Zhang *et al.*, Phys. Rev. Lett. 86, 5562 (2001)

 Cu_x -(SiO2)_{1-x} nano-granular films

- At a few degrees Kelvin, the thermoelectric power is very small; it is linear in $T \Rightarrow$ typical metal behavior
- There is no indication of a huge, broad Kondo bump (or dip)
- ⇒ Seemingly no sign of (appreciable) magnetic impurities



Chen *et al*., Appl. Phys. Lett. 81, 523 (2002)

Importance of Three-Dimensional Structures



* There is an increased contrast between the saturation and the strong dependence of $\tau_{in}(T)$ with increasing sample dimensionality

Minimized magnetic contamination:

• 3D samples are insensitive to surface effects (substrates, interfaces, paramagnetic oxidation)

• 3D samples do not require sophisticated lithographic processing

Lin *et al.*, J. Phys. Soc. Jpn. 72, 7 (2003), Suppl. A

Effect of Thermal Annealing on the Dephasing Time

 \Rightarrow testing the role of magnetic scattering and dynamical defects



Moderately-disordered films: ρ_0 (as-prepared) ~ 100 μΩ cm

• Thermal annealing results in a decrease in disorder

• τ_{φ} (**T** \rightarrow **0**) increases with decreasing disorder

 One might think that a decrease in disorder could be accompanied by a decrease in TLS

Lin *et al.*, Europhys. Lett. 57. 872 (2002)

Difficulties in comparison to TLS theories:

- Number concentration of two-level tunneling modes is unknown
- Coupling between conduction electrons and a TLS is poorly understood
- Dynamical properties of real defects are unclear
- *Cf.* Zawadowski et al. (1999, 2003) Imry et al. (1999) Galperin (2003)

.....

Magnetic scattering and Kondo effect



"The Kondo effect is very sensitive to disorder; decreasing disorder enhances the Kondo effect."

⇒ Stronger magnetic scattering with increasing annealing

Effect of Thermal Annealing on the Dephasing Time

AuPd thick film



Strongly-disordered films:

 ρ_{0} (as-prepared) ~ 500 $\mu\Omega$ cm

• Thermal annealing results in a decrease in resistivity by a factor ~ 6

• τ_{φ} ($T \rightarrow 0$) remains basically unchanged

• A picture based on TLS cannot apply

Lin *et al.*, Europhys. Lett. 57, 872 (2002)

τ_{φ} (*T* \rightarrow **0**) in Strongly Disordered Metals

• The measured τ_{φ}^{0} cannot be due to random magnetic contamination, e.g. $\tau_{\varphi}^{0} \propto n_{m}^{-1}$



Conclusion

- Apart from the total level of disorder, e-ph interaction is very sensitive to the microscopic quality of the defects
- "Electron-phonon engineering" may be promising
 weakened (T⁴) or enhanced (T²) e-ph interaction is possible
 - ⇒ How to observe the T^4 dependence over a wider T range? How to observe the disorder dependence: T^4l ?
- The appearance of very short dephasing lengths of ~ 10 nm (e.g., in carbon nanotubes and Cu-SiO₂ nano-granular films) is not understood
 - \Rightarrow Do we really have a large amount of magnetic impurities?

Conclusion (continued)

- Saturation in $\tau_{\varphi}(T \rightarrow 0)$ cannot be readily explained in terms of TLS models
- Magnetic-scattering induced dephasing cannot explain the saturation of τ_{φ} found in strongly disordered metals