Defects and Tunneling Systems: Disordered Quantum Solids

Introduction tunneling systems

Tunneling systems in disordered crystals thermal properties dielectric properties interacting TS incoherent tunneling

Tunneling systems in amorphous solids

thermal properties dielectric and elastic properties spectral diffusion experiments on single tunneling systems

Tunneling



crystal with point defects

→ amorphous solids

possible structural configurations with atomic tunneling systems:



typical values: $\Delta / k_{\rm B} < 10$ K $d \sim 1$ Å $\hbar \Omega / k_{\rm B} \sim 300$ K V $/ k_{\rm B} < 1000$ K

double-well potential



Atomic Tunneling Systems: two level approximation

total wave function $\psi = a \psi_{
m l} + b \psi_{
m r}$

$$H_{ll} = \int \psi_l^* H \psi_l \, \mathrm{d}^3 x$$
$$H_{\mathrm{rr}} = \int \psi_r^* H \psi_r \, \mathrm{d}^3 x$$
$$H_{\mathrm{lr}} = \int \psi_l^* H \psi_r \, \mathrm{d}^3 x$$
$$S = \int \psi_l^* \psi_r \, \mathrm{d}^3 x$$

minimizing *E*: $\partial E / \partial a = 0$ $\partial E / \partial b = 0$

$$a (H_{\rm ll} - E) + b(H_{\rm lr} - ES) = 0$$

 $a (H_{\rm lr} - ES) + b(H_{\rm rr} - E) = 0$

 $\longrightarrow (H_{\rm ll} - E)(H_{\rm rr} - E) - (H_{\rm lr} - ES)^2 = 0 \quad E_{\pm} = \frac{1}{2} \left(\hbar\Omega \pm \sqrt{\Delta^2 + 4H_{\rm lr}^2} \right)$ energy zero point $H_{\rm ll,rr} = (\hbar\Omega \pm \Delta)/2$

in addition: $S \approx 0$, overlap is small, V is large

$$E = E_{+} - E_{-} = \sqrt{\Delta^{2} + 4H_{\rm lr}^{2}} = \sqrt{\Delta^{2} + \Delta_{0}^{2}}$$

Atomic Tunneling Systems



Tunneling systems in crystals

 $\label{eq:control} \int d^{\text{often more than two minima}} \Delta \approx 0$

example: KCI:Li

- Li⁺ substitutes K⁺
- \blacktriangleright ionic radius: $r_{
 m Li^+} < r_{
 m K^+}$
 - 8 off-center positions in <111> direction



potential minima at $r = \frac{d}{2}(\alpha, \beta, \gamma)$ with $\alpha, \beta, \gamma = \pm 1$

quantum states



 A_{2u} : (1,1,1),

- $T_{2g} : (1,1,0); (1,0,1); (0,1,1) \,,$
- T_{1u} : (1,0,0); (0,1,0); (0,0,1),
- A_{1g} : (0,0,0).

tunneling states with cubic symmetry



KCI:Li, KBr:C

KCI:OH, LiF:OH

NaBr:F

example: $\langle 111 \rangle$ tunneling states

a) partition function

$$Z = \sum_{s} e^{-E_{s}/k_{\rm B}T} = 1 + 3e^{-\Delta_{0}/k_{\rm B}T} + 3e^{-2\Delta_{0}/k_{\rm B}T} + e^{-3\Delta_{0}/k_{\rm B}T}$$
$$= \left(1 + e^{-\Delta_{0}/k_{\rm B}T}\right)^{3}$$

b) internal energy

$$U = \frac{N}{Z} \sum_{s} E_{s} e^{-E_{s}/k_{\rm B}T} = 3N\Delta_{0} \frac{1}{1 + e^{\Delta_{0}/k_{\rm B}T}}$$
$$U = \frac{3}{2} N\Delta_{0} \left[1 - \tanh\left(\frac{\Delta_{0}}{2k_{\rm B}T}\right) \right]$$

c) specific heat

$$C_{\rm TS} = \frac{3nk_{\rm B}}{\varrho} \left(\frac{\Delta_0}{2k_{\rm B}T}\right)^2 \operatorname{sech}^2\left(\frac{\Delta_0}{2k_{\rm B}T}\right)$$



Schottky peak



example: KCI: Li

- ► above 1K T³ dependence is observed
- below 1 K additional contribution: Schottky peak
- ▶ just 20 ppm Li dominates specific heat

tunneling system contribution to specific heat

isotope effect observed:proof of tunneling effect

$$\int_{6}^{7} \Delta_0 / k_{\rm B} = 1.1 \, {\rm K}$$

 $\int_{6}^{6} \Delta_0 / k_{\rm B} = 1.65 \, {\rm K}$



J.P. Harrison, Phys. Rev. 171, 1037 (1968)

KCI: CN (same symmetry than KCI:Li) example:

T³ dependence subtracted

solid line: Schottky peak



P.P. Peressini, J.P. Harrison, R.O. Pohl, Phys. Rev. 182, 939 (1969)

- broadening at higher concentrations contributions of pairs
- double maximum structure at highest concentration

Thermal conductivity:

phonon transport, but resonant absorption via TS

hole in differential thermal conductivity





T.F. McNelly, Ph.D. Thesis (Cornell University 1974)

strong impact at maximum

reduction of thermal conductivity
 factor of 500 for 50 ppm OH⁻

- isotope effect observed
- confirms that TS are responsible for heat resistance

scattering rate / mean free path



P.P. Peressini, J.P. Harrison, R.O. Pohl, Phys. Rev. 180, 926 (1969)





- high temperature: classical 1/T dependence
- low temperature: quantum mechanical plateau $\propto 1/\Delta_0$
- isotope effect clearly observed
- ► solid line → theoretical description assuming isolated TS

C. Enss, M. Gaukler, S. Hunklinger, M. Tornow, R. Weis, A. Würger, Phys. Rev. B **53**, 12094 (1996)



- high temperature: classical 1/T dependence
- Iow temperature: quantum mechanical plateau $\propto 1/\Delta_0$
- maximum in between: levels contribute that couple linear to strain field

G. Weiss, M. Hübner, C. Enss, Physica B **263-264**, 388 (1999)

Concentration dependence: example dielectric susceptibility of KCI with TS









 $\checkmark \Delta \neq 0$ widely distributed

 \blacktriangleright Δ_0 also widely distributed

distribution function \longrightarrow standard tunneling model

 $P(\Delta, \lambda) \,\mathrm{d}\Delta \,\mathrm{d}\lambda = P_0 \,\mathrm{d}\Delta \,\mathrm{d}\lambda$

$$\begin{split} P(\Delta,\lambda) &\longrightarrow P(E,\Delta_0) \quad \text{with} \quad E^2 = \Delta^2 + \Delta_0^2 \\ \Delta_0 &= \hbar \Omega \mathrm{e}^{-\lambda} \end{split}$$



$$P(E, \Delta_0) d\Delta_0 dE = P(\Delta, \lambda) \left| \frac{\partial \lambda}{\partial \Delta_0} \right| \left| \frac{\partial \Delta}{\partial E} \right| d\Delta_0 dE$$
$$= P_0 \frac{E}{\Delta_0 \sqrt{E^2 - \Delta_0^2}} d\Delta_0 dE$$

density of states

$$D(E) = \int_{\Delta_0^{\min}}^{E} P(\Delta_0, E) \, \mathrm{d}\Delta_0 = P_0 \, \ln \, \frac{2E}{\Delta_0^{\min}}$$



W.A. Phillips, J. Low. Temp. Phys. **7**, 351 (1972) P.W. Anderson et al., Philos. Mag. **25**, 1 (1972)



$$C_V = \left(\frac{\partial u}{\partial T}\right)_V = \frac{\pi^2}{6} D_0 k_{\rm B}^2 T \propto T$$

total specific heat

 $C_V = AT + BT^3 + C_{\text{Debye}}$

- ► additional T^3 term \longrightarrow quasi-harmonic modes
- linear term $\sim T^{1.3}$ instead of $\sim T$
 - good agreement but glass is non-equilibrium system not all TS can contribute in measuring time



R.C. Zeller, R.O. Pohl, Phys. Rev. **B 4**, 2029 (1971) J.C. Lasjaunias et al., Sol. State Commun. **17**, 1045 (1975)

Heat release

effective density of states



$$\tau_{1} = A \left(\frac{E}{\Delta_{0}}\right)^{2} \frac{1}{E^{3}} \tanh\left(\frac{E}{2k_{B}T}\right)$$

$$P_{0} \frac{E}{\Delta_{0}\sqrt{E^{2} - \Delta_{0}^{2}}} d\Delta_{0} dE$$

heat release of amorphous solids

$$\dot{Q} = \frac{\pi^2 k_{\rm B}^2}{24} P_0 (T_1^2 - T_0^2) \frac{1}{t}$$



M. Schwark, M. Kubota, R.M. Mueller, F. Pobell, J. Low Temp. Phys. **58**, 171 (1985)

Thermal conductivity

much lower than in crystals

low temperatures: resonant scattering by TLS

$$\ell^{-1} \propto \omega \tanh\left(\frac{\hbar\omega}{2k_{\rm B}T}\right)$$

with dominant phonon approximation:

$$\hbar \overline{\omega} \approx k_{\rm B} T \quad \text{and} \ \ell^{-1} \propto \overline{\omega} \propto T$$

$$\Lambda \propto C_{\rm Debye} \ell$$

$$\swarrow T^{-1}$$

$$T^{3} \qquad T^{-1}$$



R.C. Zeller, R.O. Pohl, Phys. Rev. B 4, 2029 (1971)



R. B. Stephens, Phys. Rev. B 8, 2896 (1973)

E

Coupling to Electric and Elastic Fields



relaxational processes

 \longrightarrow modulation of Δ

$$\delta \Delta = 2 \gamma e$$

 $\delta \Delta = 2 \mathbf{p} \cdot \mathbf{F}$





$$\mathcal{H}_{\rm S} = \frac{1}{E} \begin{pmatrix} \Delta & -\Delta_0 \\ -\Delta_0 & -\Delta \end{pmatrix} (\gamma \,\tilde{e} + \boldsymbol{p} \cdot \boldsymbol{F})$$

 $\Psi_+ \qquad \qquad E = \hbar \omega$

Echo experiments:

coherent regime: $t \ll \tau_1, \tau_2 \rightarrow \infty$ two-level approximation:

applied rf field: $F = F_0[\exp(i\omega t) + \exp(-i\omega t)] = 2F_0 \cos(\omega t)$

Schrödinger equation: $i\hbar \frac{\partial \Psi}{\partial t} = [H_0 + H_S] \Psi = \begin{bmatrix} H_0 + p \frac{\Delta_0}{E} F_0 \left(e^{i\omega t} + e^{-i\omega t} \right) \end{bmatrix} \Psi$ Ansatz: $\Psi(t) = a_1(t) \Psi_- e^{-i\omega_1 t} + a_2(t) \Psi_+ e^{-i\omega_2 t} \longrightarrow \begin{cases} a_1(t) = \cos\left(\Omega_R t\right) \\ a_2(t) = -i\sin\left(\Omega_R t\right) \\ & \int \\ \Omega_R = \frac{1}{\hbar} \frac{\Delta_0}{E} \mathbf{p} \cdot \mathbf{F}_0 \\ & \text{Rabi frequency} \end{cases}$



Origin of echo



two-pulse echo decay

- sensitivity five orders of magnitude
- non-exponential decay

what determines the decay: spectral diffusion



and thermally fluctuating TS



Two Level Systems and 1/f noise

Noise generating two level systems are

- Strain dipoles that couple to the strain field (phonons)
- Electric dipoles (if they are charged) that couple to an electric field (photons)

Not all TLS have electric dipole moments but all TLS are strain dipoles

TLS systems may cause fluctuations in carrier number, mobility, flux, critical current, dielectric constant, etc.



Tunneling Systems in Superconducting Quantum Devices

TLS generate noise & dissipation in

- MOSFETs & single-electron transistors
- micro-mechanical resonators
- single-photon detectors, nanowires
- superconducting resonators and qubits



APL **97**, 252501 (2010) – phase qubit PRL **95**, 046805 (2005) – charge qubit PRB **84**, 235102 (2011) – *E*_J fluctuations

TLS are found

- in surface oxides
- in / on the substrate
- at interfaces
- in tunnel junctions

in Josephson junctions:



- hydroxide defects
- dangling bonds
- electrons trapped at interfaces: Kondo- / Andreev Fluctuator

1/f Flux Noise in SQUIDs

1/f^α with 0.58 < α< 0.80 Wellstood *et al.*, APL **50**, 772 (1987)



Hypothetical noise source Noise from SQUID(2) or I_{b1} Noise from $I_{\Phi1}$ Symmetric fluctuations in $I_{01} \& I_{02}, R_1 \& R_2$, or $L_1 \& L_2$ Antisymmetric fluctuations in I_{01} and I_{02} Antisymmetric fluctuations in L_1 and L_2 Antisymmetric fluctuations in R_1 and R_2 Fluctuations in external magnetic field Noise from substrate Noise from SQUID support Liquid helium in cell Heating effects Motion of flux lines trapped in SQUID

Noise would not appear as flux noise Noise would depend on M_i Noise would not appear as flux noise a S_{Φ} would scale as I^2 S_{Φ} would scale as V^2 $S_{\Phi}^{1/2}$ would scale as SQUID area Should depend on material Should depend on material Should change in absence of helium Should depend on power dissipated Should depend on material



"Universal" 1/f flux noise

Independent of: inductance, materials, geometry Not due to fluctuating vortices (seen in wires too thin to have a vortex)

Mechanism not fully known

Paramagnetic Susceptibility

 \mathbf{T}

Paramagnetism: Magnetization M is proportional to the magnetic field H $\chi \mid \chi \mid M = \chi H$

Curie Susceptibility: $\chi \sim \frac{1}{\tau}$

- Consider a toroidal current loop (SQUID) with spins on the surface.
- Current produces B field that polarizes spins.
- Polarized spins contribute to M and flux Φ .
- Flux $\Phi = LI \leftrightarrow$ Magnetization M = χH .

 $\Phi \leftrightarrow M$, $L \leftrightarrow \chi$, $I \leftrightarrow H$

Flux Noise in SQUIDs

- Noise ~ $(1/f)^{\alpha}$ where $0.5 < \alpha < 1$.
- 1/f flux noise in SQUIDs is produced by fluctuating magnetic impurities.
- Paramagnetic impurities produce flux ~ 1/T on AI, Nb, Au, Re, Ag, etc.





Evidence Indicates Spins Reside on Metal Surface



- Flux noise scales with surface area of the metal in the SQUID.
- Magnetic impurities in the bulk superconductor would be screened.
- Weak localization *dephasing time* τ_{φ} *grows as T decreases* (Bluhm *et al.*). If spin impurities in the bulk limited τ_{φ} , τ_{φ} would saturate at low T (Webb).
- Concentration ~ 5 × 10¹⁷/m² implies a *spacing of ~1 nm* between impurities if spin moment is 1 μ_B.
- Lee *et al.* proposed adsorbed neutral OH are the spins but spin reorientation barrier ~ 600 K.

Where do the spins causing flux noise come from?

- Oxygen (O_2) molecules adsorbed on the surface.
- Consistent with flux noise independent of material and scaling with surface area.





Molecular oxygen is paramagnetic. O_2 molecule has 2 unpaired electron spins in the triplet state (S=1) with magnetic moment = $2\mu_B$.

Another Flux Noise Source: Hydrogen Atoms

- Surface treatments do not remove all flux noise sources.
- ESR measurements on sapphire indicate hydrogen atoms
 - 1.42 GHz ESR line splitting matches free H atom
 - Flux noise measurements find *peak at 1.4 GHz*
 - High frequency flux noise can cause qubit relaxation.



Quintana et al. PRL 2017

Summary of Flux Noise Superconducting SQUIDs

- Flux noise in SQUIDs is produced by magnetic impurities such as
 - Paramagnetic O₂ molecules adsorbed on the surface.
 This explains that
 - noise scales with surface area
 - flux noise independent of materials
 - Hydrogen atoms embedded in and adsorbed on metallic oxide layer and substrate
- Surface treatments can help remove impurities.

Candidate dielectrics for quantum circuits



- a-Si:H has ~ factor 10 lower loss than SiNx (defects overconstrained from fourfold bond coordination?)
- Even in qubits with "simple" single-layer fabrication, lossy native oxides will limit T₁ times (2D transmon data compatible with surface loss)
- Crystalline dielectrics should be much better
- Surface chemistry of metals is also important (tantalum)

Tunneling Systems in Superconducting Quantum Devices



Qubit-TLS interaction: via TLS electrical dipole moment \vec{p}

Qubit: $\hat{H} = \frac{\hat{\Phi}^2}{2L} + \frac{\hat{Q}^2}{2C} + E_J$ for $\overline{p} = 2 \text{ D} = 2 \cdot 0.2 \text{ eÅ}$ $\sim -\overline{p} |\vec{E}| \approx h \cdot 10 \text{ N}$ el. Field: $\vec{E} = \frac{\hat{Q}}{t \ C} \approx 1000 \ \text{V/m}$ $\Rightarrow g = \overline{p} \left| \vec{E} \right| \approx h \cdot 10 \text{ MHz}$ qubit-TLS coupling strength

Two-Level-Systems Strain Spectroscopy



G. Grabovskij, J. Lisenfeld, G. Weiss, A.V. Ustinov et al., Science 338, 232 (2012)

1/f Critical Current Noise

- Fluctuating TLS in the tunnel junction with electric dipole moment produce fluctuations in the tunneling matrix element resulting in 1/f noise in the critical current.
- TLS theory: $S_{crit current}(f) \sim TL^5/f$ (Constantin and Yu, 2007)
- Experiment on tunnel resistance: $S_R(f) \sim T/f$



Charge Noise

- Fluctuating electric dipoles in tunnel junction barriers and insulating materials produce image charges in nearby superconductors and, hence, **low frequency** 1/f charge noise.
 - Experiment: $S_Q(f) \sim T^2/f$
 - TLS Theory: $S_Q(f) \sim T/f$





Second Spectrum – "NOISE of the NOISE"



Summary of Squid Circuit Noise

- Flux noise in SQUIDs is produced by magnetic impurities such as
 - paramagnetic O_2 molecules adsorbed on the surface
 - H atoms embedded in and adsorbed on metallic oxide layer
- Fluctuating two level systems with electric dipole moments in the tunnel barrier and insulating materials produce
 - Dielectric loss
 - Energy splittings due to coupling with qubit energy levels
 - Charge noise
 - Critical current noise
- Need better and cleaner materials, fabrication processes, surface treatments, device designs, etc.