Thermal and Quantum Fluctuations in Superconductor Nanograins

Correlations and coherence in quantum systems
Évora, Portugal, 8-12 October 2012

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MAX-PLANCK-GESELLSCHAFT

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Phys. Rev. B 84, 104525 (2011)
I. Brihuega, A. M. García-García, PR,
M. M. Ugeda, C. H. Michaelis, S. Bose, K. Kern

Phys. Rev. Lett. 108, 097004 (2012)
PR, A. M. García-García

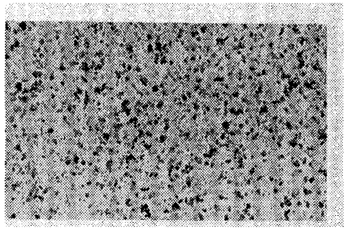
Motivation

Technological developments permit to synthesize and characterize high quality nano-structures

Properties of single superconducting nano-grains can be studied by STM

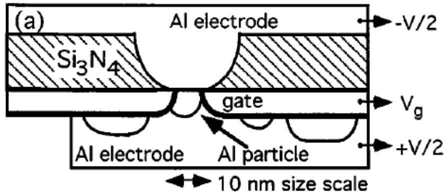
It is now possible to address the role of thermodynamic fluctuations on the stability of the superconducting state

Granular material



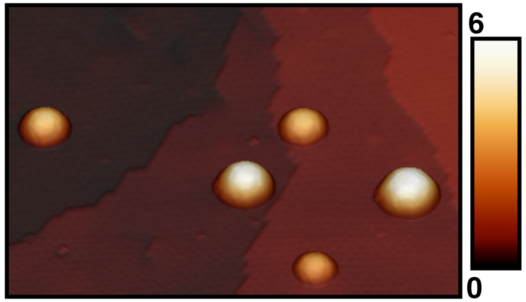
B. Abeles, Roger W. Cohen, and G. W. Cullen
Phys. Rev. Lett. 17, 632–634 (1966)

Single grains



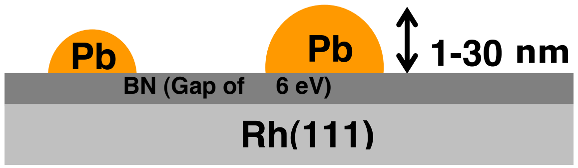
D. C. Ralph, C.T. Black, and M. Tinkham
PRL 74, 3241 (1995).

Isolated single grains

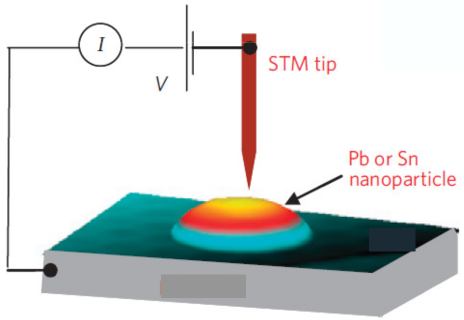


STM data
isolated Pb nanoparticles of different sizes

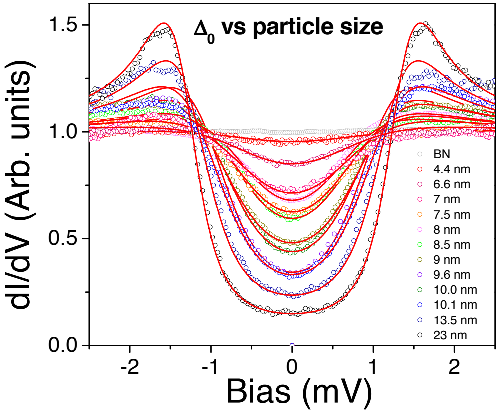
S. Bose et al.
Nature Materials 9, 550–554 (2010)

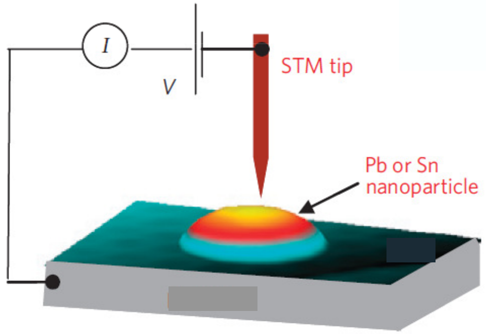


Isolated Pb nanoparticles grown on top of an insulating layer



Experimental output: Tunneling conductance





$$G(V) = \frac{dI}{dV} = G_{nn} \frac{d}{dV} \int_{-\infty}^{\infty} N_s(\omega)[f(\omega) - f(\omega - eV)]d\omega$$

Density of states of the SC nanoparticle

Topics of this talk:

Description of a clean, finite-size conventional superconductor.

Test the applicability of this description to realistic grains.

Part I — **Thermal effects**

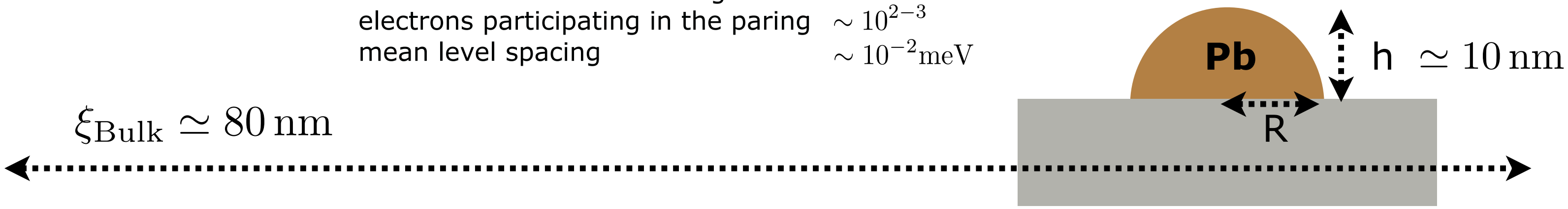
Theoretical analysis that combines thermal and quantum fluctuations in a zero dimension superconductor

Part II — **Thermal+ Quantum**

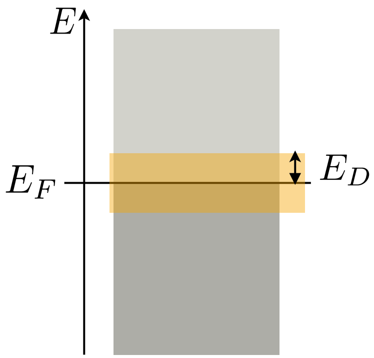
0d samples

typical values for a 10nm grain:

- conduction electrons in the grain $\sim 10^5$
- electrons participating in the paring $\sim 10^{2-3}$
- mean level spacing $\sim 10^{-2}\mathrm{meV}$



BCS Hamiltonian



$$H = \sum_\alpha \varepsilon_\alpha c_\alpha^\dagger c_\alpha - \delta \sum_{\alpha,\alpha'>0} g_{\alpha,\alpha'} c_\alpha^\dagger c_{-\alpha}^\dagger c_{-\alpha'} c_{\alpha'}$$

$$|E_\alpha - \mu| = |\varepsilon_\alpha| < E_D$$

$$g_{\alpha,\alpha'} = g$$

$$\alpha = (k,\uparrow), (-k,\downarrow)$$

$$-\alpha = T(\alpha)$$

$$(k,\uparrow) > 0$$

$$\delta \propto V^{-1} \left(\frac{m}{\hbar}\right)^{-3/2} E_F^{-1/2}$$

Corrections within MF-BCS:

Single particle DOS

$$\varrho(\varepsilon) = \sum_\alpha \delta(\varepsilon - \varepsilon_\alpha)$$

for equally spaced levels: $\varrho(\varepsilon) = \delta^{-1}$ $\quad |\varepsilon| < E_D$

for a nanograin: finite size corrections to ϱ and $g_{\alpha,\alpha'}$

R. Parmenter
Phys. Rev. 166, 392 (1968).

J. M. Blatt and C. J. Thompson,
Phys. Rev. Lett. 10,332 (1963)

integrable and chaotic grains

H. Olofsso *et al.*
PRL 100, 037005 (2008)

A. M. García-García *et al.*
PRL 100, 187001 (2008)

Corrections due to thermal fluctuations

Hubbard-Stratonovich transformation

$$Z = \int \mathcal{D}\Delta^\dagger \mathcal{D}\Delta \mathcal{D}c^\dagger \mathcal{D}c \; e^{-\int_0^\beta d\tau \left\{\sum_\alpha c_\alpha^\dagger [\partial_\tau + \varepsilon_\alpha] c_\alpha + \sum_\alpha \left(c_\alpha^\dagger c_{-\alpha}^\dagger \Delta(\tau) + \Delta^\dagger(\tau) c_{-\alpha} c_\alpha\right) + (g\delta)^{-1} \Delta^\dagger(\tau)\Delta(\tau)\right\}}$$

Static Path Approximation (SPA)

B. Mühlschlegel, D. J. Scalapino, R. Denton
PRB 6,1767 (1972)

$$Z/Z_0 = \int_0^\infty d\,|\Delta|^2 \;\; e^{-\beta \mathcal{A}_0(|\Delta|)}$$

$\Delta(\tau) = \Delta$ classical variable
valid for finite T

$$\mathcal{A}_0\left(|\Delta|\right) = (g\delta)^{-1}\,|\Delta|^2 - \frac{1}{\beta}\int_D d\varepsilon\,\varrho\left(\varepsilon\right)\ln\frac{\left[1+e^{-\beta\xi_0}\right]\left[1+e^{\beta\xi_0}\right]}{\left[1+e^{-\beta|\varepsilon|}\right]\left[1+e^{\beta|\varepsilon|}\right]}$$

$$\xi_0 = \sqrt{\varepsilon^2 + |\Delta|^2}$$

$$\int_D = \int_{-E_D}^{E_D}$$

BCS MF result is recovered for bulk systems $\mathcal{A}_0 \propto (\text{Volume})$

$$\partial_{|\Delta|}\mathcal{A}_0\left(|\Delta|\right) = 0 \qquad\qquad (g\delta)^{-1} = \int_D d\varepsilon\,\varrho\left(\varepsilon\right)\frac{\tanh\left(\beta\xi_0/2\right)}{2\xi_0}$$

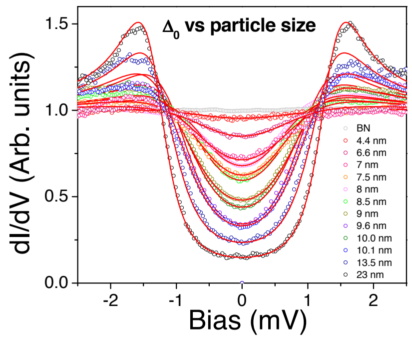
Typical tunneling conductance experiment

$$G(V) = \frac{dI}{dV} = G_{nn}\frac{d}{dV}\int_{-\infty}^{\infty} N_s(\omega)[f(\omega) - f(\omega - eV)]d\omega$$

↓

$$N_D(\omega) = \mathrm{Im}\left(\frac{\omega + i\Gamma}{\sqrt{\Delta^2 - (\omega + i\Gamma)^2}}\right)$$

Dynes expression



Fit to obtain Δ and Γ

No microscopic parameters are needed

Interpretation: Δ SC gap

Γ (Quasiparticle lifetime)$^{-1}$ QP scattering, recombination, experimental apparatus, ...

What about thermal fluctuations due to finite size effects?

Not so simple for finite systems ....
... have the Δ and Γ the same interpretation than for bulk?

.... back to the SPA formula

DOS in the SPA approximation

$$N_{\mathrm{SPA}}(\omega) = \frac{Z_0}{Z}\int_0^{\infty} d\,|\Delta|^2 \; e^{-\beta \mathcal{A}_0(|\Delta|)} N_D(\omega)$$

$$N_D(\omega) \;=\; \mathrm{Im}\left(\frac{\omega + i\Gamma}{\sqrt{\Delta^2 - (\omega + i\Gamma)^2}}\right)$$

agrees with the MF expressions for $\delta \to 0$

Define the order parameter

$$\bar{\Delta}^2 = (g\delta)^2\left[\sum_{\alpha,\alpha'>0}\left\langle c^\dagger_{\alpha'} c^\dagger_{-\alpha'} c_{-\alpha} c_\alpha\right\rangle - \sum_{\alpha>0}\left\langle c^\dagger_\alpha c_\alpha\right\rangle\left\langle c^\dagger_{-\alpha} c_{-\alpha}\right\rangle\right]$$

$\bar{\Delta}$ coincides with the QP gap for $\delta \to 0$

Fit the SPA DOS to experimental measurements to get Γ (Quasiparticle lifetime)

Compute $\bar{\Delta}$ by the SPA formula

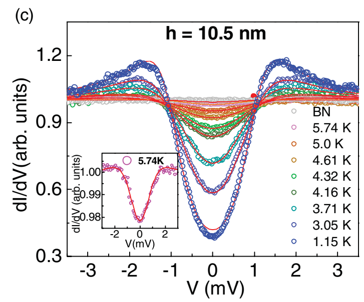
Microscopic theory is needed:

- $\varrho(\varepsilon)$ OK!
- E_D Fixed to the bulk value
- g Coulomb interactions, phonon spectrum...
  ideal world: Eliashberg + ...
  real world: too difficult to get something reasonably simple.
  solution: fit it also!

Why all this?

Dynes expression fits perfectly the experimental data!

There is no direct experimental measure of Δ!



Theoretician's experiment....

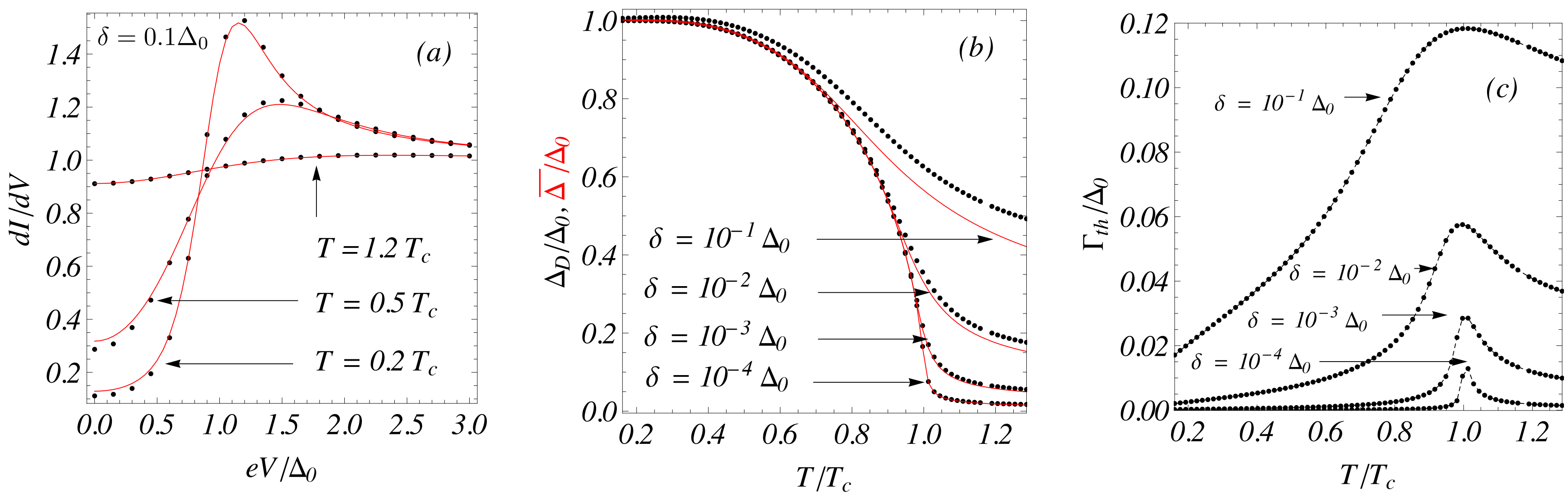
Set $\Gamma = \tau^{-1} = 0.1\Delta_0$

Fit Dynes expression to SPA generated "data": Δ_D $\quad \Gamma_D = \tau^{-1} + \Gamma_{\text{th}}$

has two contributions!

$g = 0.34$

$E_D = 9.0\,\Delta_0$



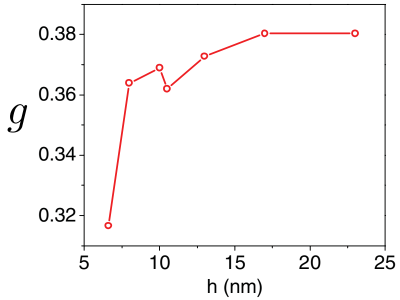
Good fit even for large δ

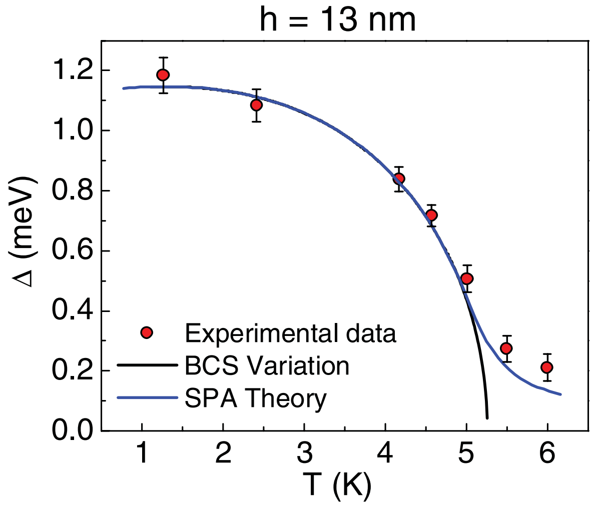
$\bar{\Delta} \simeq \Delta_D$
in the experimental accessible region
$\delta/\Delta_0 < 10^{-2}$

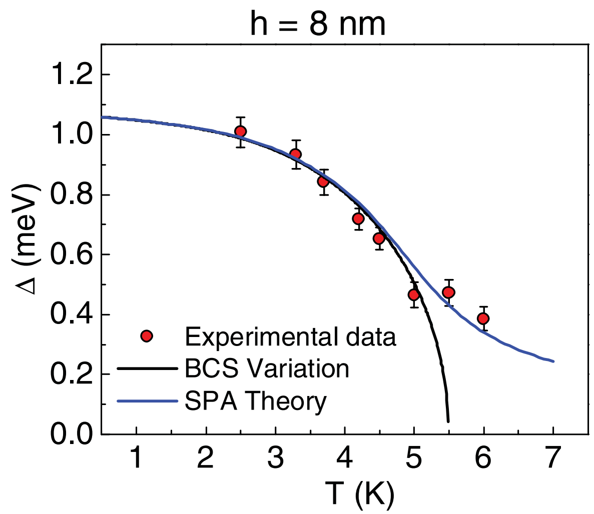
Quantifies thermal fluctuation

Finite size effects:

BCS: corrected DOS
Non BCS: $g(\delta)$
Thermal: SPA







Conclusion of Part I

It is possible to address single SC grains in STM experiments

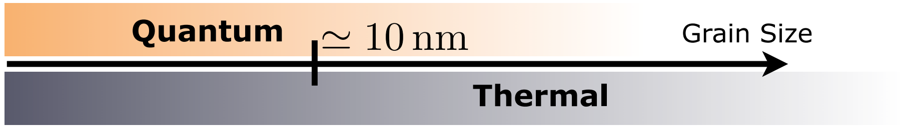
The experimental data is well described by a theoretical model that includes TFs + mean-field finite-size effects

TFs give rise to a finite-energy gap in the “fluctuation dominated regime” around Tc.

Part II

Thermal fluctuations: For a finite size system Δ is not a well defined quantity

important near Tc



Quantum fluctuations: $\Delta(\tau)$ is a dynamic quantity

most important for low temperatures

The role of fluctuation (thermal + quantum) in the behavior of the order parameter in finite size systems

(thermal + quantum) fluctuation is a long standing problem in the theory of superconductivity in nanograins - also relevant in cold atom and nuclear physics.

Phys. Rev. Lett. 108, 097004 (2012)
PR, A. M. García-García

Hamiltonian for a 0d sample

$$H = \sum_{\alpha} \varepsilon_\alpha c^\dagger_\alpha c_\alpha - g\delta \sum_{\alpha,\alpha'>0} c^\dagger_\alpha c^\dagger_{-\alpha} c_{-\alpha'} c_{\alpha'}$$

- exact solution - Richardson equations - useful for T=0
  Von Delft, Sierra, Braun, Dukelsky...
- no results for finite T

Hubbard-Stratonovich field

$$Z = \int \mathcal{D}\Delta^\dagger \mathcal{D}\Delta \mathcal{D}c^\dagger \mathcal{D}c \; e^{-\int_0^\beta d\tau \left\{ \sum_\alpha \begin{pmatrix} c_{\alpha,1}(\tau) \\ c^\dagger_{\alpha,-1}(\tau) \end{pmatrix}^\dagger \begin{pmatrix} \partial_\tau + \varepsilon_\alpha & \Delta(\tau) \\ \Delta^\dagger(\tau) & \partial_\tau - \varepsilon_\alpha \end{pmatrix} \begin{pmatrix} c_{\alpha,1}(\tau) \\ c^\dagger_{\alpha,-1}(\tau) \end{pmatrix} + (g\delta)^{-1}\Delta^\dagger(\tau)\Delta(\tau) \right\}}$$

Integrate out the electrons

$$\frac{Z}{Z_0} = \int \mathcal{D}\Delta^\dagger \mathcal{D}\Delta \; e^{-\beta\mathcal{A}[\Delta]}$$

$$\beta\mathcal{A}[\Delta] = (g\delta)^{-1} \int_0^\beta d\tau \, \Delta^\dagger(\tau)\Delta(\tau) - \mathrm{tr}\left\{\ln\left[-G^{-1}\right] - \ln\left[-G_0^{-1}\right]\right\}$$

$$G^{-1} = -\begin{pmatrix} \partial_\tau + \varepsilon_\alpha & \Delta(\tau) \\ \Delta^\dagger(\tau) & \partial_\tau - \varepsilon_\alpha \end{pmatrix} \qquad G_0^{-1} = -\begin{pmatrix} \partial_\tau + \varepsilon_\alpha & 0 \\ 0 & \partial_\tau - \varepsilon_\alpha \end{pmatrix}$$

Approximations

BCS:

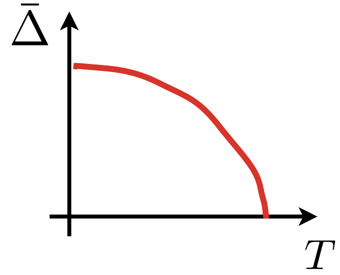
Mean field approximation

$\mathcal{A}[\Delta] \propto N$ (number of electrons participating in the paring)

saddle-point $\partial_\Delta \mathcal{A}[\Delta] = 0$

$$(g\delta)^{-1} = \sum_\alpha \frac{\tanh(\beta\xi_\alpha/2)}{2\xi_\alpha} \qquad \xi_\alpha = \sqrt{\varepsilon_\alpha^2 + |\Delta|^2}$$

Gap equation

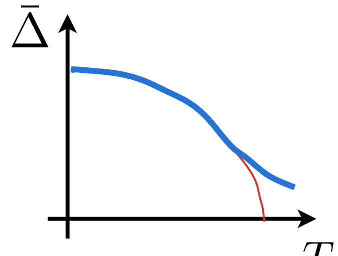


SPA:

Static path approximation

$\Delta(\tau) = \Delta$

Integrate over Δ exactly



rounding of the transition

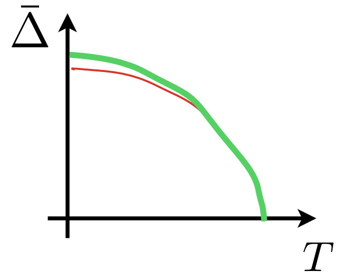
RPA:

Random phase approximation

$\Delta(\tau) = \Delta + \delta\Delta(\tau)$

Δ obtained by the saddle-point condition

$\delta\Delta(\tau)$ Gaussian fluctuations

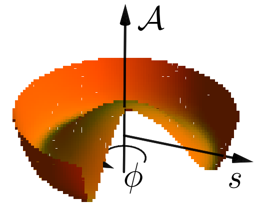


increasing of $\bar{\Delta}$ at low T

To take into account thermal and quantum fluctuations: combine SPA + RPA

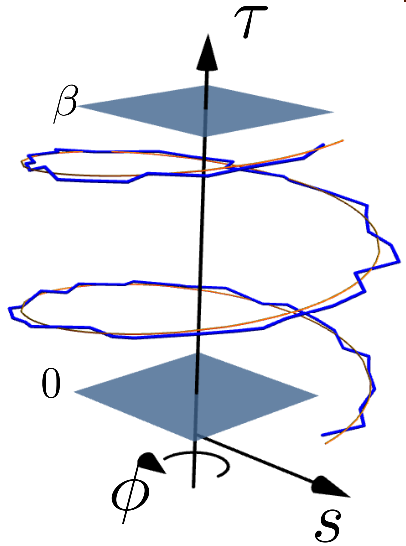
Rossignoli and Canosa
Ann. of Phys. 275, 1-26 (1999)

Part II :: SPA+RPA (i)



Decompose the field Δ in phase and amplitude $\Delta(\tau) = s(\tau) e^{i\phi(\tau)}$

$$\Delta(0) = \Delta(\beta) \quad \Rightarrow \quad \begin{array}{l} s(0) = s(\beta) \\ \phi(0) = \phi(\beta) + 2\pi M \end{array}$$



$M = 2$

Consider the fluctuations

$$s^2(\tau) = s_0^2 + \delta s^2(\tau)$$

$$\phi(\tau) = \phi_0 + 2\pi M \tau/\beta + \delta\phi(\tau)$$

Expand the action

$$\mathcal{A}[s, \phi, M] = \mathcal{A}_0(s_0) \qquad \textbf{SPA}$$

$$+ i\pi \sum_k \left(1 - \frac{\varepsilon_k}{\xi_{0k}}\right) \frac{1}{\beta} M + \left(\sum_k \frac{s_0^2}{2\xi_{0k}^3}\right) \frac{1}{\beta^2} (\pi M)^2 \qquad \textbf{Non-trivial phase configurations}$$

$$+ \frac{1}{2} \sum_{m \neq 0} \begin{pmatrix} \tilde{s}_{-m}^2 \\ \phi_{-m} \end{pmatrix} . \Xi(s_0)_m . \begin{pmatrix} \tilde{s}_m^2 \\ \phi_m \end{pmatrix} \qquad \textbf{RPA}$$

$$s_m^2 = \frac{1}{\beta} \int d\tau \, e^{i\Omega_m \tau} \delta s^2(\tau)$$

$$\phi_m = \frac{1}{\beta} \int d\tau \, e^{i\Omega_m \tau} \delta\phi(\tau)$$

$$\tilde{s}_m^2 = \left[\beta \sum_k \frac{1}{2\xi_{0k}} \tanh\left(\frac{\xi_{0k}}{2}\right)\right] s_m^2$$

all eigenvalues of $\Xi(s_0)$ are positive

...previous works we were mixing these contributions

The role of the non-trivial phase configurations

Paths with winding number $M \neq 0$ cost energy

$$Z = \int_{-\pi}^{\pi} \frac{d\phi_0}{2\pi} \int_0^{\infty} ds_0^2 \int \mathcal{D}'\tilde{s}^2\, \mathcal{D}'\phi\; e^{-\beta[\mathcal{A}_0(s_0)+\ldots]} \left[\sum_M e^{\underbrace{-i\pi \sum_k \left(1-\frac{\varepsilon_k}{\xi_{0k}}\right) M - \left(\sum_k \frac{s_0^2}{2\xi_{0k}^3}\right)}\frac{1}{\beta}(\pi M)^2} \right]$$

$$\simeq \left[\sum_M e^{-i\pi \langle N \rangle_{s_0} M + \pi^2 \frac{T}{\delta} M^2} \right]$$

$T \gg \delta$: paths with $M \neq 0$ can be discarded

$T = 0$: the sum imposes that the GS has an even number of particles
Matveev and Larkin PRL 78, 3749 (1997)

\+ Charging effects (self capacitance of the grain) -> odd,even effects

To understand the role of fluctuations we consider M=0 only

Correction due to $M \neq 0$ configurations can be easily included

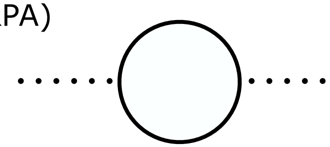
Nevertheless, it is important to identify the modes with non-trivial windings

Collective modes cause the Gaussian fluctuations to diverge in the cartesian case

Fluctuation matrix

$$\Xi_{i,j} = \beta \frac{\delta^2}{\delta X_i \delta X_j} \mathcal{A}[s,\phi] \quad \text{with} \quad X_i = s(\tau), \phi(\tau)$$

Sum all bubble-like diagrams (RPA)



$$\Xi\left(s_0\right)_m = \begin{pmatrix} \Xi_m^{\tilde{s}^2\tilde{s}^2} & \Xi_m^{\tilde{s}^2\phi} \\ \Xi_m^{\phi\tilde{s}^2} & \Xi_m^{\phi\phi} \end{pmatrix}$$

$\Omega_n = 2\pi\beta^{-1}n$ Matsubara frequencies

Cross terms

$$\Xi_m^{\tilde{s}^2\phi} = -\Xi_m^{\phi\tilde{s}^2} = \ldots \sim 0$$

can be neglected for a typical DOS (particle-hole symmetry)

Amplitude modes

$$\Xi_m^{\tilde{s}^2\tilde{s}^2} = \frac{\sum_k r_k \left[1 - \frac{4(\varepsilon_k)^2}{\Omega_m^2 + (2\xi_{0k})^2}\right]}{2\beta s_0^2 \left[\sum_k r_k\right]^2} \sim \text{gapped}$$

$$\frac{\delta s_0^2}{\bar{s}_0^2} \sim \frac{\sqrt{\delta T}}{\bar{s}_0}$$

divergence near Tc controlled by the mean level spacing

$$\bar{s}_0 \gg T \;\rightarrow\; \frac{\delta s_{m=1}^2}{\delta s_0^2} \sim 1 + \frac{2\pi T}{\bar{s}_0}$$

important near T=0

$$\bar{s}_0 \ll T \;\rightarrow\; \frac{\delta s_{m=1}^2}{\delta s_0^2} \sim \frac{1}{\ln\left(\frac{2\pi T}{\bar{s}_0}\right)}$$

negligible near Tc

$\bar{s}_0$ typical value of s_0

Phase modes

$$\Xi_m^{\phi\phi} = \sum_k r_k \frac{2\beta s_0^2 \Omega_m^2}{\Omega_m^2 + \left(2\xi_{0k}\right)^2} \sim \frac{\beta}{\delta}\Omega_m^2 \longrightarrow \delta^{-1}\int_0^\beta d\tau \left(\partial_\tau \delta\phi\right)^2$$

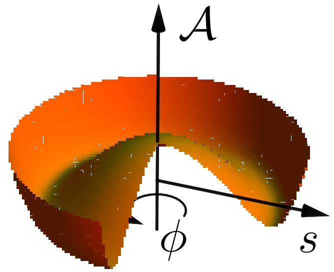
$$r_k = \frac{1}{2\xi_{0k}} \tanh\left(\frac{\beta\xi_{0k}}{2}\right)$$

Usual phase dynamic term in the action

Phase stiffness controlled by the mean level spacing

Part II :: Results

Integrate the Gaussian fluctuations to get the corrections to SPA



$$Z/Z_0 = \int_0^\infty ds_0^2 \; e^{-\beta[\mathcal{A}_0(s_0)+\mathcal{A}_1(s_0)]}$$

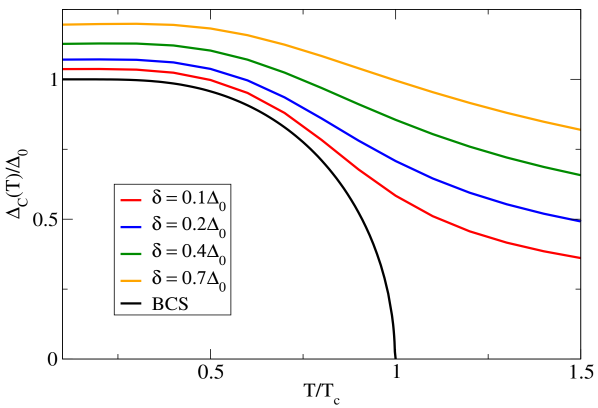
$$\mathcal{A}_1[s_0] = \frac{1}{2}\int d\nu \left[n_b(\nu) - \frac{1}{\beta\nu}\right]\frac{1}{2\pi i}\left\{\ln\left[\widetilde{C}\left(\nu + i0^+\right)\right] - \ln\left[\widetilde{C}\left(\nu - i0^+\right)\right]\right\}$$

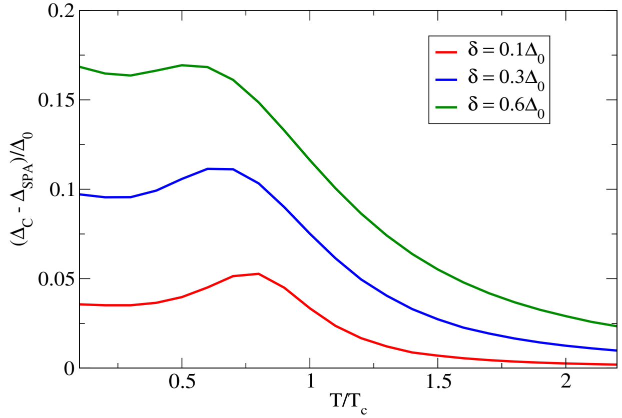
$$\widetilde{C}(z) = \left(-z^2 + 4s_0^2\right)\left(-z^2\right)\left[\int_D d\varepsilon\, \varrho(\varepsilon)\, \frac{r(\xi)}{-z^2 + (2\xi)^2}\right]^2 + \left(-z^2\right)\left[\int_D d\varepsilon\, \varrho(\varepsilon)\, \frac{2\varepsilon\, r(\xi)}{-z^2 + (2\xi)^2}\right]^2$$

$$r(\xi) = \frac{1}{2\xi}\tanh\left(\frac{\beta\xi}{2}\right)$$

Results

(for a constant density of states $\varrho(\varepsilon) = 1/\delta$)





For T=0 RPA result:

$$\bar{\Delta}(T=0) = \Delta_0(1 + \alpha\delta/E_D)$$
$$\alpha \simeq 1 > 0$$

For $T \gg T_C$ SPA result

For intermediate temperatures, differences from SPA results increase as a consequence of the combined effect of thermal and quantum fluctuations.

Conclusion of Part II

Divergences that plagued previous calculations can be cured identifying zero energy modes.

Thermal and quantum fluctuations can be combined in a single theoretical framework.

Expression for $\bar{\Delta}(T)$ valid for all temperatures to leading order in δ/Δ_0

Further work...

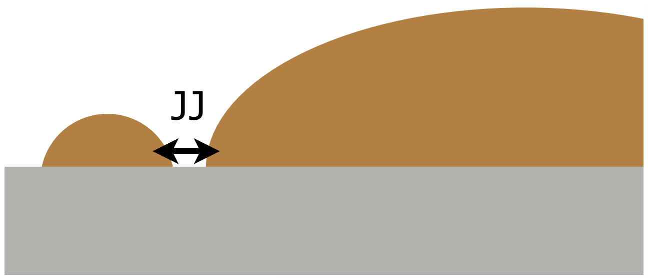
Enhancement of Δ by quantum and thermal fluctuations is not an indication that superconductivity is more robust?

No. Phase coherence is required!

How “superconducting” is a grain?

Possible clue:

The role of fluctuation in Josephson current



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ingredients:

BCS Finite Size Corrections ⟷ Semiclassical single particle DOS
García-García

Thermal Fluctuations ⟷ Path integral - SPA (static phase approximation)
Scalapino

Quantum fluctuations ⟷ Richardson Eq.
Von Delft, Sierra, Braun, Dukelsky...

preparation:

Define SPA DOS - including all the ingredients

Define the gap for a finite system

mise en plat:

Comparison with experimental results

Deviations form MF are known for T=0

$$H = \sum_{\alpha} \varepsilon_\alpha c_\alpha^\dagger c_\alpha - \lambda\delta \sum_{\alpha,\alpha'>0} c_\alpha^\dagger c_{-\alpha}^\dagger c_{-\alpha'} c_{\alpha'}$$

Exact solution -> Richardson equations

$$-\frac{1}{\lambda\delta} + \sum_{j=1;j\neq i}^{m} \frac{1}{E_i - E_j} = \frac{1}{2}\sum_{k=1}^{n} \frac{1}{E_i - \varepsilon_k} \quad i = 1, ..., m$$

GS energy $$E = 2\sum_{i=1}^{m} E_i + \sum_B \epsilon_B$$

Pair breaking excitation $$e^b = \epsilon_a + \epsilon_b + E_{g.s.}(\epsilon_a, \epsilon_b) - E_{g.s.}$$

quite complicated....

Interpretation of a (complicated) δ/Δ_0 expansion is simple...

Richardson, Yuzbashyan, Altshuler

$$\Delta^b = \Delta_0 \left(2 - \frac{\delta}{\Delta_0} + \frac{\delta}{\Delta_0}\frac{\Delta_0}{E_d} \times c^{te} + ...\right)$$

Energy to break a pair

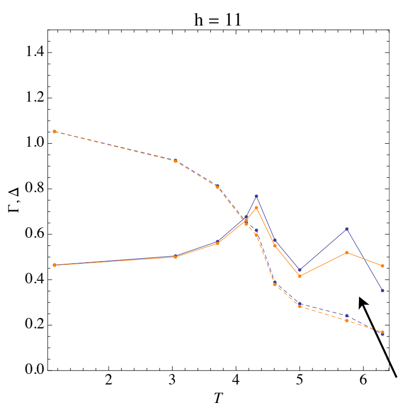
Blocking effect

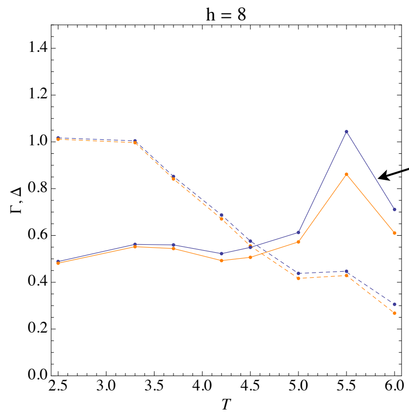
only important for $\delta \simeq \Delta_0 (h < 5\text{nm} \;\; \text{for Pb})$

prescription: remove the two energy levels closest to the E_F

use the single particle DOS : $\varrho_{1\mathrm{P}}(\varepsilon) = \varrho_{1\mathrm{P+S}} - \Theta(|\varepsilon| - \delta/2)$

Real life... and for the less good





low signal-to-noise ratio

$\Gamma_D = \tau^{-1} + \Gamma_{\text{th}}$

τ^{-1} should not see the transition!

Further work

Non conventional SC

Theory for $\lambda(\delta, T)$? sources of decoherence ?

SPA -> SPA + QF include quantum gaussian fluctuations

more accurate
complicated expressions
problems for T=0