Analyzing LDOS and energy gap fluctuations: quantifying the interplay between electron-electron interactions and disorder in 2D superconductors



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# Effects produced by the interplay between disorder and electron-electron interactions in 2D superconductors

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2D means  $d < (<) \xi$ 

Structure matters !!



Thin film top view

# Effects produced by the interplay between disorder and electron-electron interactions in 2D superconductors

 $\Delta(\vec{r})e^{i\varphi(\vec{r})}$ 

#### Disordered superconducting ultrathin films

#### Granular thin films



Jaeger et al. PRB 40, 182 (1989)

Reviews: Goldman & Markovic Phys. Today 51, 39–44 (1998)

#### Homogeneous thin films



Haviland et al. PRL 62, 2180 (1989)

Gantmakher & Dolgopolov Phys. Usp. 53, 1–49 (2010)

#### Disordered superconducting ultrathin films

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Homogeneous thin films

 $\Delta(ec{r})e^{iarphi(ec{r})}$ 

Fermionic scenario

$$k_F l \rightarrow 1$$

#### Bosonic scenario

Finkelstein mechanism

increased Coulomb repulsion: poor electronic screening reduces  $\lambda$ 



Finkelstein Pis'ma Zh. Eskp. Teor. Fis. 1987

Anderson localization local phase fluctuations



Review: Sacépé et al Nat. Phys. 2020

#### Structure of NbN on Al<sub>2</sub>O<sub>3</sub> (ex situ growth)



STM and in-situ 4 probe R(T)



Moderate disorder  $k_F \ell_e \approx 2 - 3$   $T_c \approx 0.3 T_{c-bulk}$  $R_{\Box} \approx 7 \text{ k}\Omega$ 

In situ R(T)



STM head



Home-made apparatus

# STM/STS

UHV :  $P < 5x10^{-11}$  mbar

In situ growth @  $P < 1x10^{-10}$  mbar

Base T° 300 mK (He<sub>3</sub> single shot) T<sub>electrons</sub>~380 mK (55 hours)

Magnetic Field: 0 – 10 T



Preparation chamber e-beam evaporators Recap: tunneling spectroscopy of superconductors

$$dI / dV(\mathbf{r}) = \int_{-\infty}^{\infty} N_s(E, \mathbf{r}) \left[ \frac{-\partial f(E + eV)}{\partial (eV)} \right] dE$$

#### BCS DOS

Ν

S

$$N_{S}(E) = N_{N}(E) \frac{E}{\sqrt{E^{2} - \Delta^{2}}}$$





Serrier-Garcia et al. PRL 110, 157003 (2013)

#### Structure of NbN on Al<sub>2</sub>O<sub>3</sub> (ex situ growth)



 $\int d_{NbN} = 2.33 \text{ nm}$   $l_{grain} = 2 - 8 \text{ nm}$ 



STM and in-situ 4 probe R(T)



In situ R(T)

Moderate disorder

 $k_F \ell_e \approx 2-3$ 

 $T_c \approx 0.3 \ T_{c-bulk}$  $R_{\Box} \approx 7 \ k\Omega$ 

Emerging electronic and superconducting inhomogeneities

#### STM topography

#### Gap map



See also: Sacepe et al PRL 2008, Chand et al PRLB 2012, Noat et al PRB 2013...

#### Emerging electronic and superconducting inhomogeneities





No local cross-correlation between the grain structure and gap inhomogeneities

Emerging electronic and superconducting inhomogeneities

#### STM topography

#### gap map

auto correlation



Superconducting puddles much larger than NbN nanocrystals

Emerging electronic and superconducting inhomogeneities

#### STM topography

#### gap map

#### auto correlation



 $\implies l_{|\Delta|} \approx 35 \text{ nm} \gg \xi \approx 5 \text{nm} \approx l_{grain}$ 

# Content

Revealing quantitatively the interplay between disorder and electronelectron interactions in 2D superconductors from LDOS analysis

#### 1. Case of moderate disorder in NbN thin films

PHYSICAL REVIEW B 102, 024504 (2020)

**Editors' Suggestion** 

Spectroscopic evidence for strong correlations between local superconducting gap and local Altshuler-Aronov density of states suppression in ultrathin NbN films

C. Carbillet,<sup>1</sup> V. Cherkez,<sup>1</sup> M. A. Skvortsov,<sup>2,3,\*</sup> M. V. Feigel'man,<sup>3,2</sup> F. Debontridder,<sup>1</sup> L. B. Ioffe,<sup>4,3</sup> V. S. Stolyarov,<sup>1,5,6</sup> K. Ilin,<sup>7</sup> M. Siegel,<sup>7</sup> C. Noûs,<sup>8</sup> D. Roditchev,<sup>1,9</sup> T. Cren,<sup>1</sup> and C. Brun<sup>1,†</sup>

#### 2. Case of weak disorder in Pb single atomic layer

PHYSICAL REVIEW B 107, 174508 (2023)

**Editors' Suggestion** 

# Local density of states fluctuations in a two-dimensional superconductor as a probe of quantum diffusion

Mathieu Lizée,<sup>1,2,\*</sup> Matthias Stosiek,<sup>3</sup> Igor Burmistrov,<sup>4,5,†</sup> Tristan Cren,<sup>2</sup> and Christophe Brun<sup>2,‡</sup>

NbN film moderate disorder

$$k_F \ell_e \approx 2 - 3$$
  $T_c \approx 0.25 T_{c-bulk}$ 

#### STM topography

Gap map



 $l_{|\Delta|} \approx 27 \text{ nm} \gg \xi \approx 5 \text{nm} \approx l_{grain}$ Carbillet et al PRB 102, 024504 (2020)

NbN film moderate disorder

$$k_F \ell_e \approx 2 - 3$$
  $T_c \approx 0.25 T_{c-bulk}$ 

#### Local dl/dV spectra

Gap map



$$\begin{split} |\Delta|(\vec{r}) \\ l_{|\Delta|} \approx 27 \text{ nm} \gg \xi \approx 5 \text{nm} \approx l_{grain} \\ \end{split}$$
 Carbillet et al PRB 102, 024504 (2020)

NbN film moderate disorder  $k_F \ell_e \approx 2 - 3$   $T_c \approx 0.25$   $T_{c-bulk}$ 



 $|\Delta|(\vec{r})$ 

Strong locally varying Altshuler-Aronov background

NbN film moderate disorder  $k_F \ell_e \approx 2 - 3$   $T_c \approx 0.25$   $T_{c-bulk}$ 

local dl/dV spectra at location  $\vec{r}$ 



Power law fitting of the Altshuler-Aronov background

Local dl/dV spectra



#### cross correlation



#### cross correlation



#### suppression of the tunneling DOS induced by Coulomb effect

$$\nu(E) = \nu_0 e^{-S(E)} \qquad S(E) = \frac{2}{R_Q} \int_E^{1/\tau} \frac{d\omega}{\omega} R(\omega)$$

with  $R_Q = h/e^2$ 

 $R(\hbar\omega)$  spreading resistance between the diffusive scale  $r_{in}(\hbar\omega)$  and field propagation scale  $r_{out}(\hbar\omega)$ 

Altshuler et al PRL 1980, Levitov & Shytov JETP Lett. 1997, Andreev & Kamenev PRB 1999 Finkelstein JETP 1983

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 $r_{in}(\hbar\omega) = \sqrt{\frac{\hbar D}{\hbar\omega}} \qquad R(\hbar\omega) = \frac{R_{\Box}}{2\pi} ln \frac{r_{in}(\hbar\omega)}{r_{out}(\hbar\omega)} \\ R(\hbar\omega) = \sqrt{\frac{\hbar D\hbar\omega_0}{\hbar^2\omega^2}} \qquad R(\hbar\omega) = \frac{R_{\Box}}{2\pi} ln \frac{\omega_0}{\omega}$ 

Altshuler et al PRL 1980, Levitov & Shytov JETP Lett. 1997, Andreev & Kamenev PRB 1999 Finkelstein JETP 1983

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with  $R_Q = h/e^2$ Homogeneous case:  $\nu(E) \propto E^{\alpha(E)}$  $\alpha(E) = \frac{R_{\Box}}{2\pi R_O} ln \frac{\hbar \omega_0}{E}$ 

#### suppression of the tunneling DOS induced by Coulomb effect

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with  $R_Q = h/e^2$   $\Rightarrow$  Homogeneous case:  $\nu(E) \propto E^{\alpha(E)}$   $\alpha(E) = \frac{R_{\Box}}{2\pi R_Q} ln \frac{\hbar \omega_0}{E}$  $/\tau \approx 0.3 \text{ eV}$ 

 $\frac{\hbar}{\tau} \approx 0.3 \text{ eV}$   $\frac{\hbar\omega_0}{\kappa} \approx 10 \text{ eV}$   $R_{\Box} \approx 7 \text{ k}\Omega$  E = 5 - 30 meV

 $\alpha_{th}(E) \approx 0.29$ 

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 $\hbar\omega_0$  and  $\hbar/\tau$  from Semenov et al. PRB 2009



Inhomogeneous case:

Assumption that fluctuations of  $\alpha(\vec{r})$  and  $|\Delta|(\vec{r})$  originate from fluctuations in 2D resistivity:

 $\rho(\vec{r}) = R_{\Box} + \delta \rho(\vec{r})$ 

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The spreading resistance possess a local term for  $|\vec{r} - \vec{r}_0| < l_{\alpha}$ 



Inhomogeneous case:

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 $\rho(\vec{r}) = R_{\Pi} + \delta \rho(\vec{r})$ 



$$\delta\alpha(\vec{r}, E) = \frac{\delta\rho(\vec{r})}{2\pi R_Q} ln \frac{E}{\hbar D/l_{\alpha}^2}$$

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 $\rho(\vec{r}) = R_{\Box} + \delta \rho(\vec{r})$ 

 $\alpha(\vec{r}) = \langle \alpha \rangle + \delta \alpha(\vec{r})$ 

 $l_{\alpha} \approx 18 \text{ nm}$   $\sigma_{\alpha} \approx 0.03 \implies \sigma_{\rho}$ ?  $D \approx 0.5 \text{ cm}^2/\text{s}$ 





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 $l_{\alpha} \approx 18 \text{ nm}$   $\sigma_{\alpha} \approx 0.03 \implies \sigma_{\rho} \approx 1.1 \text{ k}\Omega$   $D \approx 0.5 \text{ cm}^2/\text{s} \qquad \frac{\sigma_{\rho}}{R_{\Box}^{max}} \approx 16\%$ Carbillet et al PRB 102, 024504 (2020)



Inhomogeneous case:

local Finkelstein picture to explain of  $|\Delta|(\vec{r})$  fluctuations

 $\rho(\vec{r}) = R_{\Box} + \delta\rho(\vec{r}) \qquad \delta\Delta(\vec{r}) = -\frac{\delta\rho(\vec{r})}{6\pi R_Q} \ln^3 \frac{\hbar\omega_D}{\Delta_{bare}}$  $\Delta(\vec{r}) = \langle\Delta\rangle + \delta\Delta(\vec{r})$ 

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 $\sigma_{\rho} \approx 1.1 \text{ k}\Omega$   $\langle \Delta \rangle \approx 1.22 \text{ meV}$   $\hbar \omega_D \approx 300 \text{ K} \implies \sigma_\Delta \approx 0.09 \text{ meV}$   $\Delta_{bare} \approx 2.85 \text{ meV} \qquad \sigma_\Delta / \langle \Delta \rangle \approx 7.4\%$ Carbillet et al PRB 102, 024504 (2020)

# $\Delta(\vec{r})$ experimental distribution

1.2

Gap width (meV)

1.3

1.4

1.1

1.0

Conclusion of part 1

Moderate disorder in ultrathin NbN films  $\begin{cases} k_F \ell_e \approx 2 - 3 \\ T_c \approx 0.25 T_{c-bulk} \end{cases}$ 

Emergent electronic inhomogeneities explained by a local Finkelstein picture: meV scale and tens meV scale linked

 $l_{grain} = 2-8 \text{ nm}$  $\sigma_{\rho}/R_{\Box}^{max} \approx 16\%$  $\rho(\vec{r}) = R_{\Pi} + \delta \rho(\vec{r})$  $\xi \approx 5 \text{ nm}$  $\alpha(\vec{r}) = \langle \alpha \rangle + \delta \alpha(\vec{r})$  $\sigma_{\alpha}/\alpha \approx 10\%$  $l_{\alpha} \approx 18 \text{ nm}$  $\Delta(\vec{r}) = \langle \Delta \rangle + \delta \Delta(\vec{r})$  $\sigma_{\Lambda}/\langle\Delta\rangle\approx7\%$  $l_{|\Delta|} \approx 27 \text{ nm}$ 

 $\blacksquare$  Low  $q < q_D$  matter !! Interface resistance of nanocrystals mostly contribute to  $\delta \rho(\vec{r})$ thus linked to the grain structure



ANR

DED BY TH



# Thanks

#### Theory







Clémentine Carbillet PhD





Misha Skvortsov





Misha Feigel'man Lev Ioffe



Dimitri Roditchev



Tristan Cren



François Debontridder



Christophe r Brun

#### Growth of NbN films



Kostia Ilin, KIT Germany

# Content

Revealing quantitatively the interplay between disorder and electronelectron interactions in 2D superconductors from LDOS analysis

1. Case of moderate disorder in NbN thin films Carbillet et al PRB 102, 024504 (2020)

2. Case of weak disorder in Pb atomic monolayer Lizée et al. PRB 107, 174508 (2023)

PRL 108, 017002 (2012)

Th

Enhancement of the Critical Temperature of Superconductors by Anderson Localization

I. S. Burmistrov,<sup>1</sup> I. V. Gornyi,<sup>2,3,4</sup> and A. D. Mirlin<sup>2,4,5,6</sup>

2D systems, electron-electron interaction in particle-hole and Cooper channels,  $\sigma$ -model renormalization group framework Short-range spatial fluctuations of  $\lambda(r)$  over  $l_{\lambda} \ll \xi$ this physically corresponds to screened Coulomb interactions

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#### Extension of the works of Feigel'man et al.:

PRL 98, 027001 (2007)	PHYSICAL	REVIEW	LETTERS	week ending 12 JANUARY 2007

Th Eigenfunction Fractality and Pseudogap State near the Superconductor-Insulator Transition

M. V. Feigel'man,<sup>1</sup> L. B. Ioffe,<sup>2,1</sup> V. E. Kravtsov,<sup>3,1</sup> and E. A. Yuzbashyan<sup>2</sup>

and also Feigel'man et al. Annals of physics 325, 1390 (2010)

3D systems, close to the mobility edge

PRL 108, 017002 (2012)

PHYSICAL REVIEW LETTERS

week ending 6 JANUARY 2012

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PRL 108, 017002 (2012)

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Exp

PHYSICAL REVIEW LETTERS

week ending 6 JANUARY 2012

Enhancement of the Critical Temperature of Superconductors by Anderson Localization

I. S. Burmistrov,<sup>1</sup> I. V. Gornyi,<sup>2,3,4</sup> and A. D. Mirlin<sup>2,4,5,6</sup>

Disorder induced multifractal superconductivity in

monolayer niobium dichalcogenides

Zhao et al. Nature Phys. 15, 904 (2019)

Exp Visualization of Multifractal Superconductivity in a Two-Dimensional Transition Metal Dichalcogenide in the Weak-Disorder Regime

Rubio-Verdu et al. Nano Lett. 20, 5111 (2020)

Puzzling: in both experiments the level disorder  $k_F \ell_e > 10$  is too low for theory to explain  $T_C$  enhancement

PRL 108, 017002 (2012)

Th

PHYSICAL REVIEW LETTERS

week ending 6 JANUARY 2012

Enhancement of the Critical Temperature of Superconductors by Anderson Localization

I. S. Burmistrov,<sup>1</sup> I. V. Gornyi,<sup>2,3,4</sup> and A. D. Mirlin<sup>2,4,5,6</sup>

Choose an appropriate 2D superconductor to probe first the weak-disorder limit !

Check consistency between experiment and theory for LDOS and gap energy spatial fluctuations

#### Model 2D system: Pb/Si(111) single atomic layer

ultrahigh vacuum growth: P~10<sup>-11</sup> mbar range

Si(111)-7x7

#### monolayer Pb/Si(111)



C. Brun et al. Nature Physics 10, 444 (2014) C. Brun et al. Supercond. Sci. Technol. 30, 013003 (2017)

#### Model 2D system: Pb/Si(111) single atomic layer

large scale

small scale



Polycrystalline structure with grain size  $l_{grain} = 2-10 \text{ nm}$ ARPES: single 2D band with strong Rashba spin-orbit coupling

#### Model 2D system: Pb/Si(111) single atomic layer

#### average superconducting properties at T = 0.3 K



 $\langle \Delta \rangle \approx 350 \ \mu eV$   $\Gamma \approx 10 \ \mu eV$   $T_c \approx 1.8 \ K$ estimated  $k_F \ell_e \approx 30 - 100$   $\xi \approx 50 \ nm \gg l_{grain}$ 

consistent with Zhang et al. Nature Phys. 2010, Yamada et al. PRL 2013, Cherkez et al PRX 2014

#### very small local energy gap fluctuations at T = 0.3 K



Exp:  $\sigma_{\Delta}/\langle\Delta\rangle \approx 1.1 \%$ Th:  $\sigma_{\Delta}/\langle\Delta\rangle \approx 0.5 - 1 \%$   $\sigma_{E_{max}} \approx \sqrt{c/g} (\Gamma/\langle\Delta\rangle)^{2/3}$ with  $c \approx 0.3$ 

 $\langle \Delta \rangle \approx 350 \ \mu eV \quad \sigma_{\Delta} \approx 4 \ \mu eV$ 

 $\Gamma \approx 10 \ \mu eV$  estimated  $g = k_F \ell_e \approx 30 - 100$ 

#### large local density of states fluctuations at T = 0.3 K



reminder:  $\xi(0) \approx 50$  nm !!

reveal the multifractality of the underlying wavefunctions

quantifying local density of states fluctuations at T = 0.3 K

normalized variance of  $\eta(E) \propto dI/dV(E)$ 



#### Conclusion of part 2

weak disorder in a Pb/Si(111) 2D single layer  $k_F \ell_e \approx 30 - 100$  $l_{grain} = 2-10$  nm

screening of e-e interaction: spatial fluctuations of  $\lambda(r)$  over  $l_{\lambda} \ll \xi$ 

relative LDOS fluctuations  $\approx 10 \%$   $\sigma_{\Delta}/\langle \Delta \rangle \approx 1 \%$ 

quantitative agreement with multifractal 2D superconductivity

Lizée et al. PRB 107, 174508 (2023)



# Thanks

#### Experiments, data analysis





Mathieu Lizée student

**Tristan Cren** 



#### Christophe Brun

# Theory



Igor Burmistrov Landau Institute Russia



Matthias Stosiek Post-doc Japan







$$\begin{split} \langle \delta \rho(E_1, \mathbf{r}_1) \delta \rho(E_2, \mathbf{r}_2) \rangle &= \frac{\rho_0^2}{2\pi g} \operatorname{Re} \left\{ [1 + X_{E_1} X_{E_2}^*] K_0 \left( R \sqrt{\frac{2\gamma_{\Phi} - iE_1 / X_{E_1} + iE_2 / X_{E_2}^*}{\hbar D}} \right) \\ &- [1 - X_{E_1} X_{E_2}] K_0 \left( R \sqrt{\frac{2\gamma_{\Phi} - iE_1 / X_{E_1} - iE_2 / X_{E_2}}{\hbar D}} \right) \right\}, \end{split}$$



# Experimental dependence of auto-correlation



# Reducing the thickness of crystalline thin films

50

40

30

20

10

0

28

26

24



Pb films grown on Si(111) in ultrahigh vacuum

> Not exhaustive ! *Strongin et al. PRL 1973 Pfennigstorf et al. PRB 2002 Ozer et al. Nat. Phys. 2006 Eom et al. PRL 2006 Brun et al. PRL 2009 Qin et al. science 2009*

Guo et al. Science 2004

20

22

Thickness N (ML)

18

16

# Superconductivity in 1 atomic layer of Pb/Si(111)

50 nm 100 nm 0.30 d ь 0.35 0.25 0.30 0.20 (Vm)∆ ⊲(mV) 0.25 √7×√3 Pb SIC Pb 0.20 0.15 Data o Data 0.15 💻 BCS theory 0.10 BCS theory 0.10 T\_= 1.83 K T\_ = 1.52 K 0.05 0.05 0.3 0.6 0.9 1.2 1.5 1.8 D. 0 0.20.4 0.6 0.8 1.01.2 1.4 1.6 Т (K)  $\pm 100$ 

 $\sqrt{7} \times \sqrt{3}$ 

Coverage:1.20 Pb atom for 1 Si

 $T_{c} = 1.5 \text{ K}$ 

Stripedincommensurate (SIC)

Coverage:1.30 Pb atom for 1 Si

 $T_{c} = 1.8 \text{ K}$ 

Zhang et al. Nat. Phys. 6, 104 (2010)

# DFT : decoupling of surface states from bulk at E<sub>F</sub>



Small changes of the Pb-6px,y states due to the substrate DFT : Jung and Kang Surf. Sci 601, 555 (2011)

# Macroscopic resistivity of 1 atomic layer of Pb/Si(111)

Micro-four point probes – spacing = 20 microns



 $T_{c-transport} = 1.1 \text{K} < T_{c-STM} = 1.8 \text{ K}$ 

*Uchihashi et al. PRL 107, 207001 (2011)* (In monolayer) *Yamada et al. PRL 110, 237001 (2013)* (Pb monolayer)

# Existence of strong Rashba SOC in Pb/Si(111)

- Strong spin-orbit coupling in Pb/Si(111) layer
- 3D inversion always broken in a surface layer

Split-off in (x,y) plane of the 2D electron bands

 $H_{\rm so} = \alpha(\boldsymbol{\sigma} \times \mathbf{p}) \cdot \mathbf{n} \quad \text{Spi}$ 

Spins put in plane by  $H_{SO}$ Spins  $\perp$  to **p** 

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Bychkov, Rashba, JETP Lett. 39, 78 (1984) LaShell et al. PRL77, 3419 (1996)

# DFT+SOC and spin-ARPES of the Pb-SIC monolayer



#### Fermi surface



Helical spin texture

DFT: Ren PRB 94, 07543 (2016) spin-ARPES: Brand PRB 96, 035432 (2017)

# Summary of the electronic properties of the Pb-SIC monolayer

- 1 Rashba split band (Pb  $6p_{x,y}$  orbitals)
- m ~ 1.2 m<sub>0</sub>
    $k_F \sim 1.36 \text{ Å}^{-1}$  

   E<sub>F</sub> ~ 0.8 eV
    $\lambda_F \sim 4.6 \text{ Å}$

True 2D electronic confinement 6p<sub>x,y</sub> states at E<sub>F</sub>

 $\Delta << 2|\alpha|p_F << E_F$ 0.3meV 100meV 800meV

 $\xi \sim 50 \text{ nm}$   $\ell_e \sim 5 \text{ nm}, \text{ k}_F \ell_e > 20 \text{ very far from SIT }$ Mean-level spacing of single-electron states in a coherence volume  $\delta = E_F / (\xi/a)^2 \sim 0.1 \text{ meV} \sim \Delta/3$ 

# Electron-phonon coupling from DFT



Assumptions: perfect  $\sqrt{3}x\sqrt{3}$  Pb/Si(1111) - No SOC - Fixed Si atoms

Noffsinger and Cohen Solid State Commun. 151, 421 (2011)



 $\xi_0 = (\hbar D/\Delta)^{1/2} \approx 47$ nm Nat. Phys. 10, 444 (2014) Very good agreement also with: Cherkez et al PRX 2014





#### B=40mT

Justification: see 3D Usadel calculations by Kupriyanov & Golubov

Nature Commun. 9, 2277, 2018