Superconductor-insulator transition in MoC ultrathin films
Transport and STM studies

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SUPPRESSION OF T_c IN THIN FILMS

first noticed in 1938, Nature (London) by Shalnikov in Pb and Sn films ~ hundreds of nm thin

~ LOCALISATION IN METALS & SUPERCONDUCTORS

Ioffe

Ioffe-Regel criterion, *Prog. Semicond*. 1960

When $\boldsymbol{l_{min}}$ ~ ***a*** **, electrons localize**
(***l*** - electron’s mean free path & ***a*** - lattice constant)

$$k_F l_{\min} \approx 1 \quad \left(k_F \cong 1/a\right) \quad (n = k_F^2/2\pi)$$

Regel

Minimum metallic conductivity
(maximum sheet resistance)

$$\sigma_{2D} = \frac{ne^2\tau}{m} = \frac{k_F^2/2\pi\, e^2 l}{m v_F} = \frac{k_F^2/2\pi\, e^2 l}{\hbar k_F} = \frac{e^2}{h} = 1/25.82\mathrm{k}\Omega$$

localization of Cooper pairs *q=2e*

$$\sigma_{2D} = \frac{4e^2}{h} = 1/6.45\mathrm{k}\Omega$$

SUPERCONDUCTOR - INSULATOR TRANSITION (SIT)

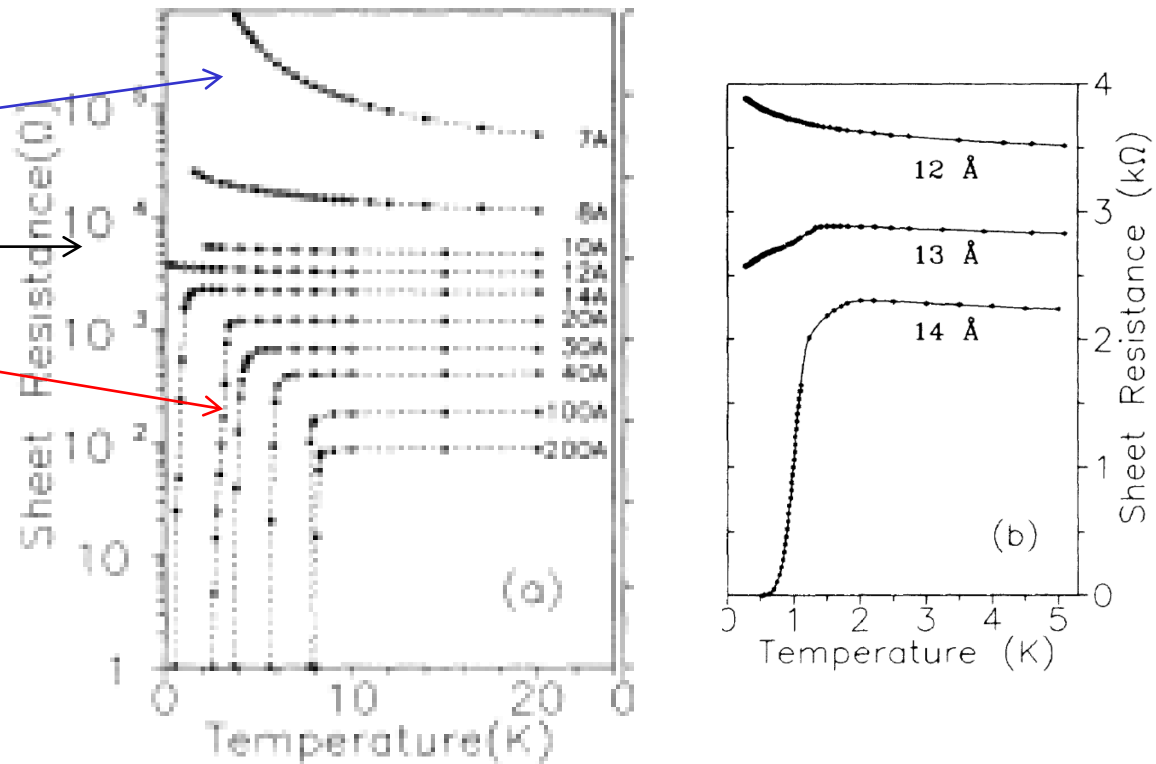
VOLUME 64, NUMBER 25 PHYSICAL REVIEW LETTERS 18 JUNE 1990

Critical Sheet Resistance for the Suppression of Superconductivity in Thin Mo-C Films

S. J. Lee and J. B. Ketterson

insulating films

separatrix @ $R_Q \sim 3$ kΩ
(<6.45 kΩ)
<superconducting films



What is the physics of
1. **suppression of T_c on sc side**
2. **SIT transition**
3. **insulating state**

WHAT CONTROLS T_c ?

PHYSICAL REVIEW B VOLUME 1, NUMBER 3 1 FEBRUARY 1970

Destruction of Superconductivity in Disordered Near-Monolayer Films*

MYRON STRONGIN, R. S. THOMPSON, O. F. KAMMERER AND J. E. CROW
Brookhaven National Laboratory, Upton, New York 11903

T_c versus resistivity ρ

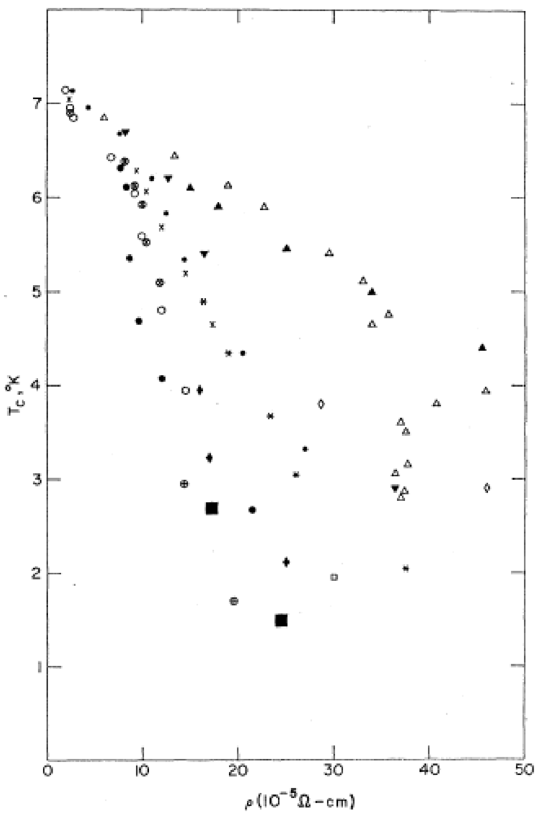


FIG. 7. T_c versus resistivity ρ (see Fig. 5 for symbol notation).

T_c versus inversed thickness

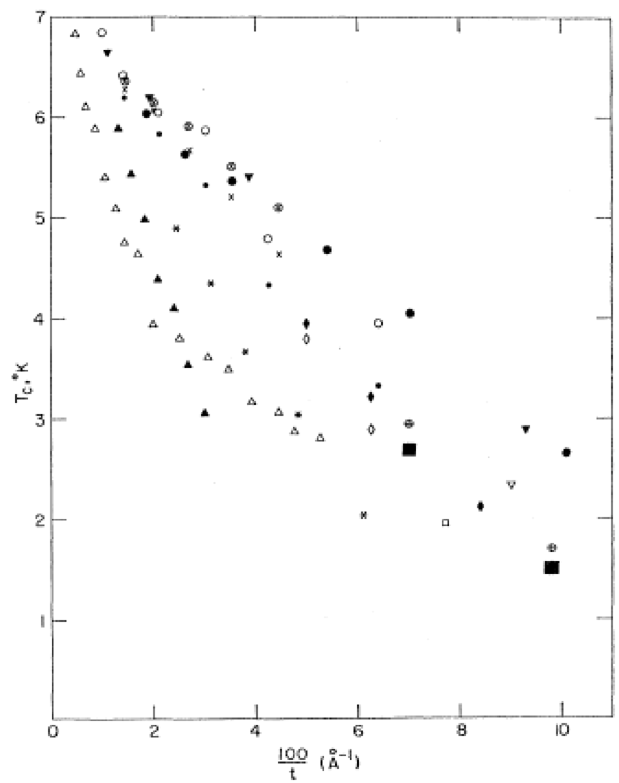


FIG. 6. T_c versus inverse thickness (see Fig. 5 for symbol notation).

T_c versus sheet resistance

seems as best correlation

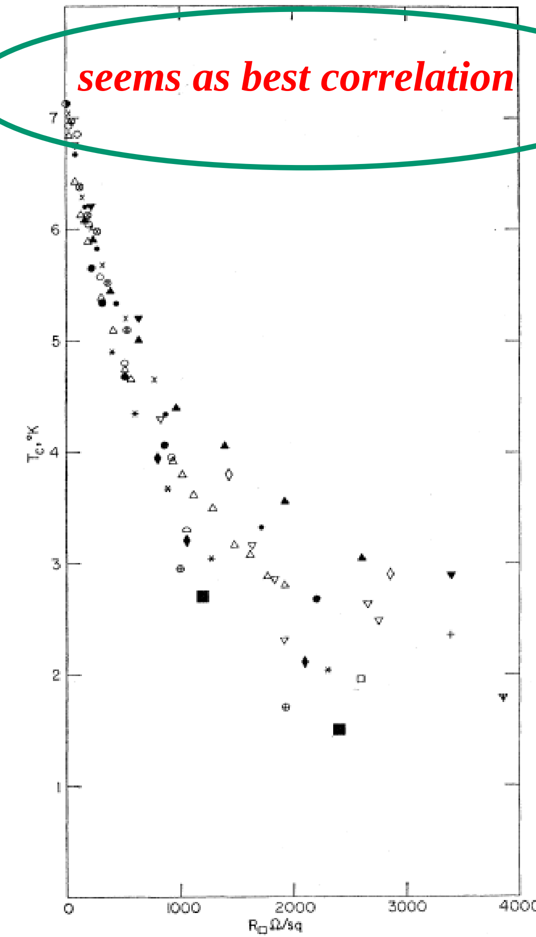


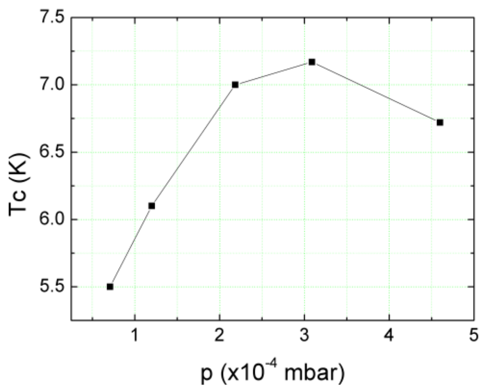
FIG. 5. T_c versus $R_\square$ where $R_\square$ is the resistance/sq area: ⊕, ■—Pb on Ge; +—Pb on Al_2O_3; ♦, ◊—Pb on Ge (deposited at room temperature); ∗—Bi on SiO; ∇, ⊗, ○, •, ●, △, ▲, ▼—Pb on SiO.

Pb and Bi films on dif. substrates

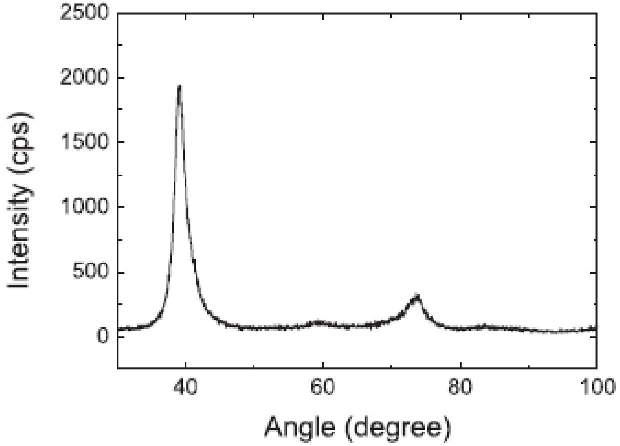
Preparation of our MoC thin films

Reactive magnetron sputtering, target Mo 99.95%
in mixture of Ar and acetylene gas on singleXtalline saphire @ 200 C

optimization of acetylene pressure

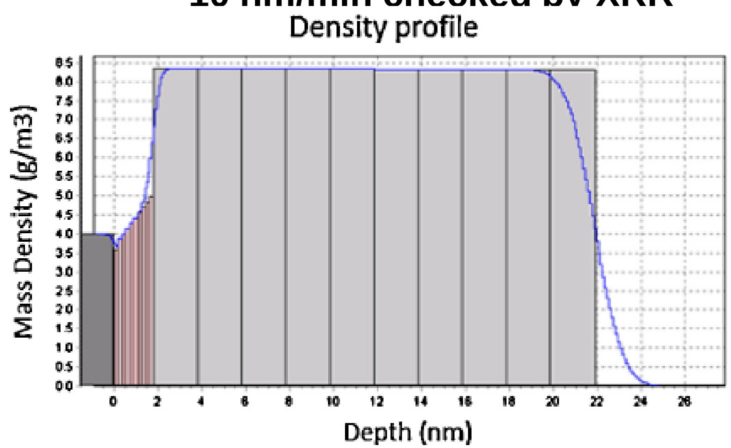


RTG analysis shows MoC peak



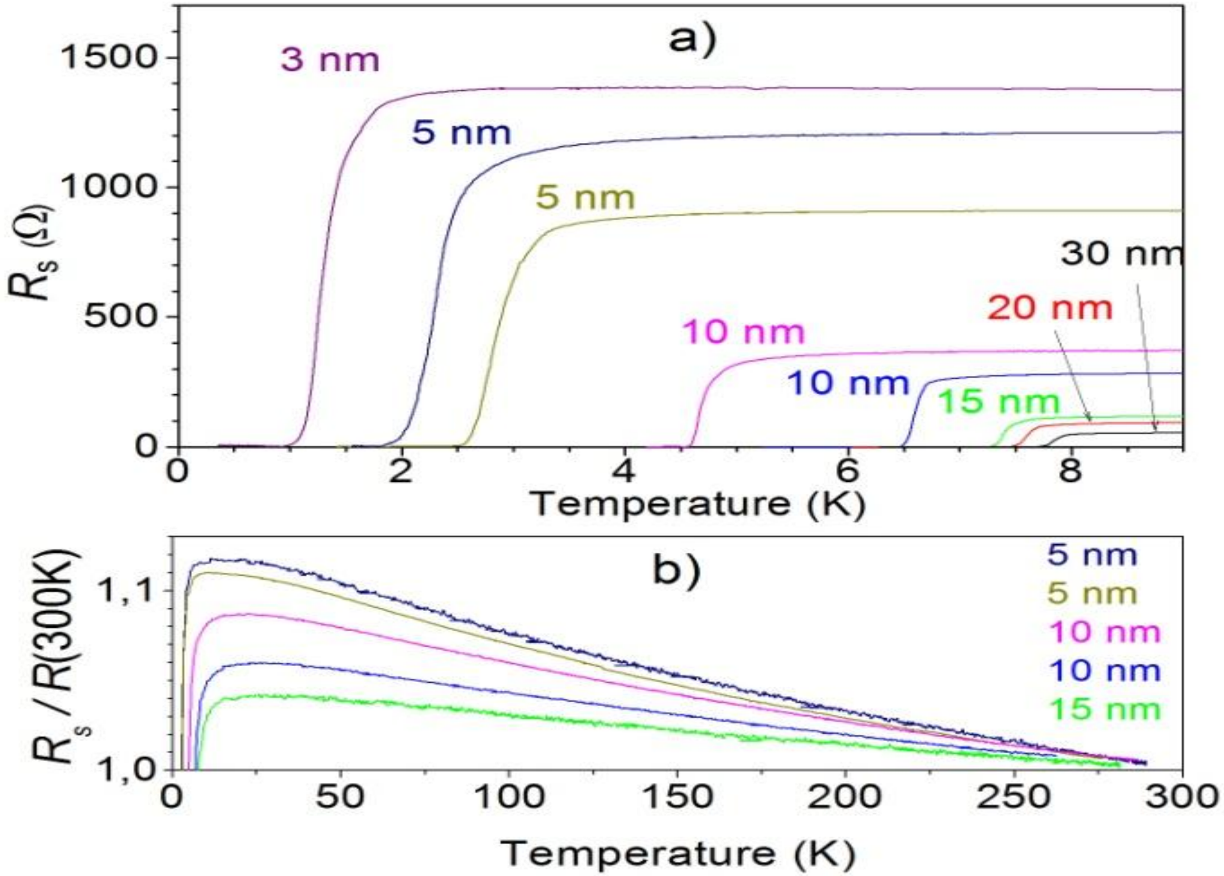
AFM on 1 x 1 mm²
Corrugation 0.5 nm

Thickness controled by sputtering time
~10 nm/min checked bv XRR



Trgala et al., Appl. Surf. Sci. 2014

Transport in MoC thin films

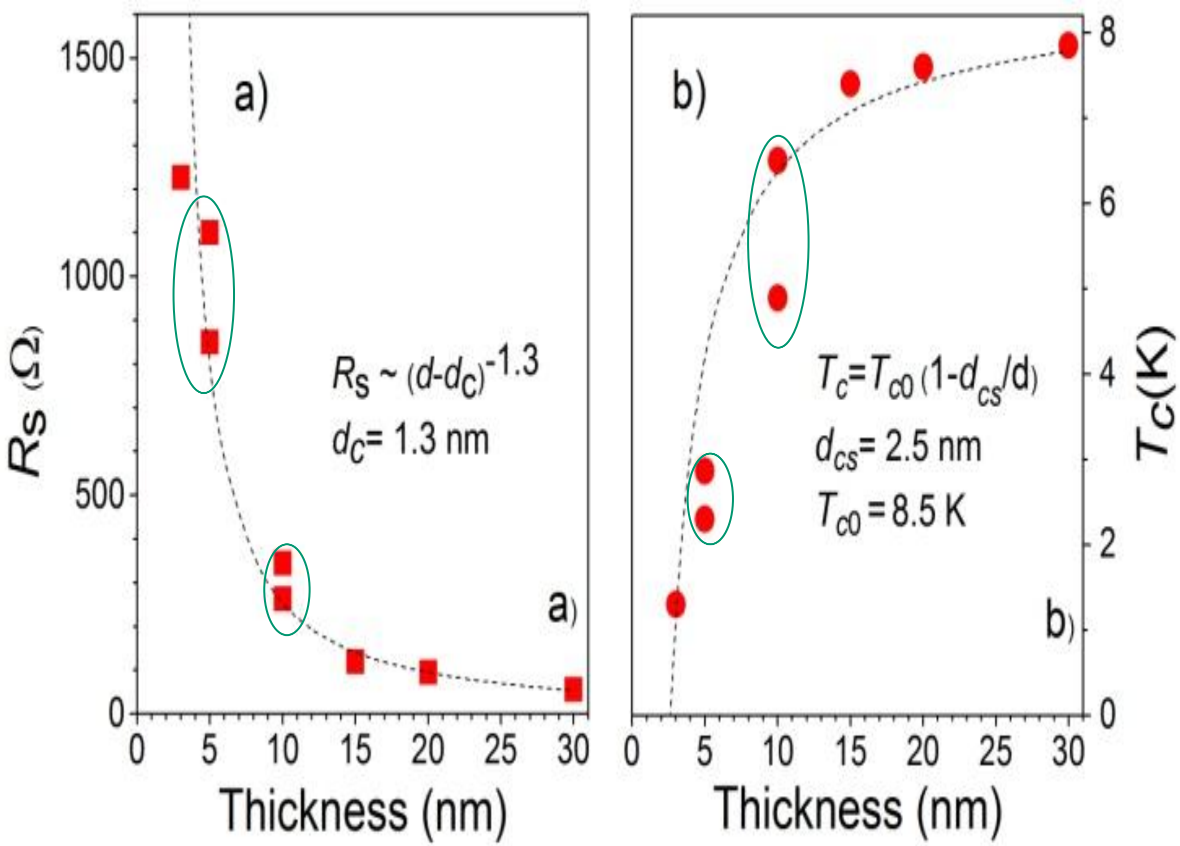


✓ sharp transitions @ T_c for different *d*
✓sheet resistance R_S increase from 50 to 1400 Ω
✓ small quantum corrections due WL and EEI
✓T_c shift from 8 K to 1K

=> electrically continuous/homogeneously disordered films

✓ two 10 & two 5 nm films with different T_c due small change in preparation conditions

Thickness effect



a) Thickness dependence of $R_S \sim (d - d_c)^{-1.3}$ in classical **percolation theory** => d_c = 1.3 nm, minimum thickness for electrical continuity (MIT)

b) Thickness dependence of $T_c(d) = T_{c0}(1-d_{cs}/d)$ within **GL calculations** (Simonin, 1986) with a **surface term** (decreased DOS) => $d_{cs} \sim 2.5$ nm (good fit for optimally prepared films)

$d_{cs} > d_c$ => first SMT and then MIT

Transport in MoC thin films

4-probe measurements in Corbino geometry

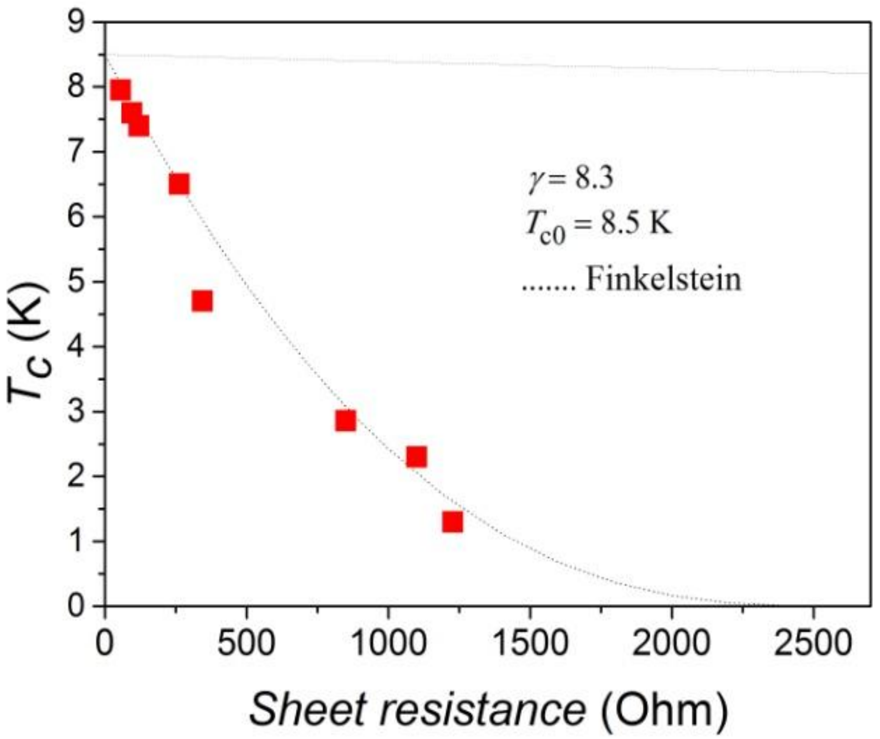
Maekawa & Fukuyama '84 / Finkel'stein '87
Increased diffusivity of electrons in 2D:
decrease of dynamical screening of Coulomb repulsion which compensates SC attraction
$\Rightarrow$ decrease of T_c , eventually $T_c \to 0$

$$Tc = Tc0 * \exp(\gamma)\left[\frac{1/\gamma + t/4 - \sqrt{t/2}}{1/\gamma + t/4 + \sqrt{t/2}}\right]^{(1/\sqrt{2t})}$$

$$\gamma = \ln\left[\frac{\hbar}{k_B T_{c0} \tau}\right] \qquad r = R/(R_Q \pi)$$

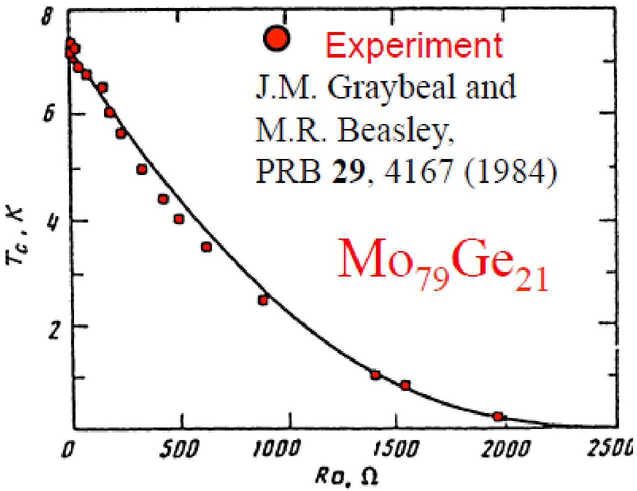
$R_Q = h/e^2 = 25\ k\Omega$

✓Good agreement !!

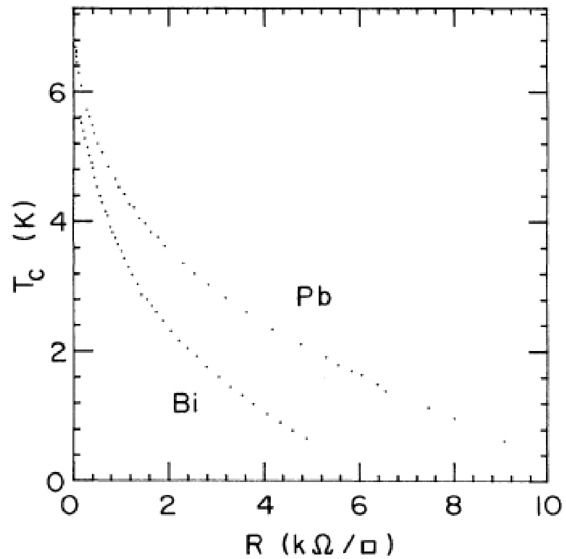


- Suppression of T_c is due to suppression of the amplitude of the sc order parameter
- Balance between sc attraction and Coulomb repulsion does not lead to full localization of electrons
- Two transitions: **superconductor => (bad) metal & metal => (fermionic) insulator**

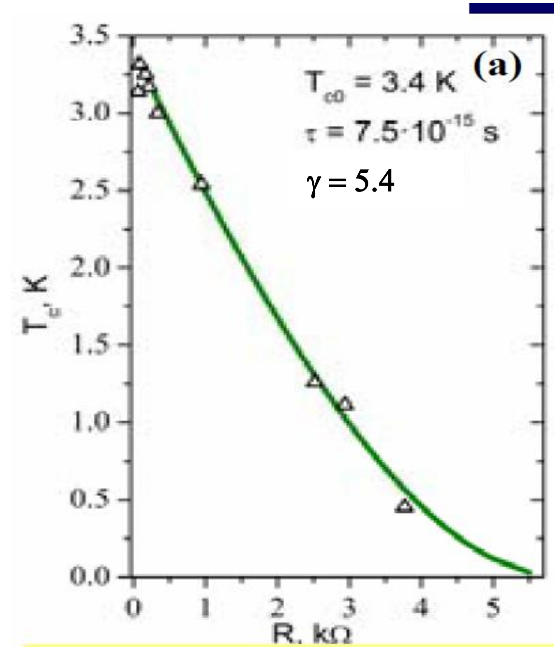
Agreement between Finkel'shtein model and experiment

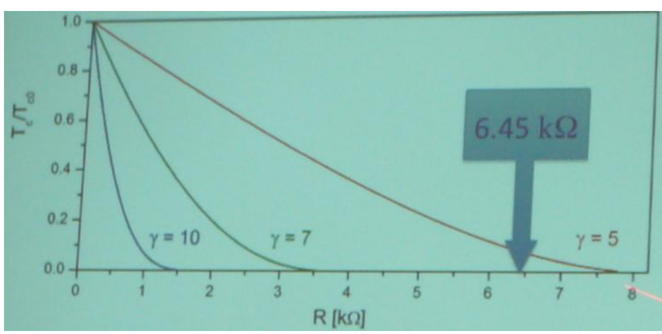


Haviland, PRL 1989



Baturina on TiN





T_{c0} [K]	γ	τ [10^{-16} sec]
1	5	514.6
	7	69.6
	10	3.5
5	5	102.9
	7	13.9
	10	0.7

Fitting parameter $\gamma \sim 5 - 8$

Finkel'stein model

Valid for 2D superconductors: $k_B T_{c0} \ll \hbar/\tau \ll \hbar D/d^2$

τ – relaxation time of qp momentum in normal state

$\hbar D/d^2$ – Thouless energy related to time t_D for qp diffussion through film with thickness *d.*
In 2D films => $l >> d$

If $l << d$ but still $k_B T_{c0} \ll \hbar D/d^2$

In Finkel'stein formula the scattering term $\hbar/t$ must be replaced by Thouless energy $\hbar D/d^2 = (\hbar/\tau)(l/d)^2$

$$Tc = Tc0 * \exp(\gamma) \left[\frac{1/\gamma + t/4 - \sqrt{t/2}}{1/\gamma + t/4 + \sqrt{t/2}} \right]^{(1/\sqrt{2r})}$$

$$\gamma = \ln \left[\frac{\hbar}{k_B T_{c0} \tau} \right] \qquad r = R/(R_Q \pi)$$

→

$$\gamma' = \ln \left[\frac{\hbar D}{d^2 k_B T_{c0}} \right] = \ln \left[\frac{\hbar}{\tau k_B T_{c0}} \left(\frac{l}{d} \right)^2 \right]$$

but this make much smaller effect on T_c ...

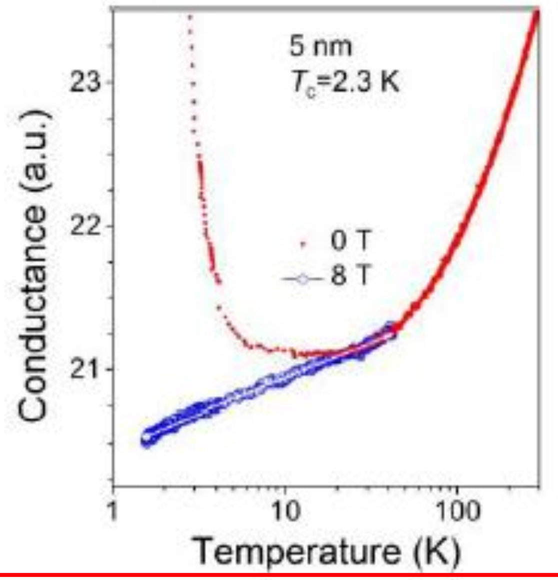
In the following we **determine the Thouless energy** in MoC films
and compare with $k_B T_{c0}$ and $\hbar/\tau$

l_T =

Thouless energy: 2D *vs* 3D character of MoC films

Disordered metal: Bloch-Gruneisen (d*R*/dT>0) + quantum corrections to sheet resistance (d*R*/dT<0)

Temperature dependence sheet conductance of MoC



Experiment:
3D to 2D transition at low temperatures

$L_T = \sqrt{2\pi\hbar D/(k_B T)}$ ~ *d* @ 30 – 40 K

i.e. $2\pi\hbar D/d^2$ = 30 -40 K

=> $k_B T_{c0} < (hD/d^2 \sim 10\, k_B T_c) \ll \hbar/\tau$

=>> $l \ll d$

We have only quasi 2D electrons in MoC

Quantum corrections = weak localization + Altshuler-Aronov can be used to test 2D or 3D character of the electron transport

In quasi-2D : $l < d < \xi, L_T$

l: mean free path, *d*: film thickness, ξ: coherence length

L_T: thermal coherence length
D: diffusion coefficient

$$L_T = \sqrt{2\pi\hbar D/(k_B T)}$$

WL + AA in conductivity =>

$$\frac{\Delta G^{WL}(T) + \Delta G^{ID}(T)}{G_{00}} = A \cdot \ln\left[\frac{k_B T \tau}{\hbar}\right];$$

In 3D: $l, L_T < d < \xi$

WL + AA in conductivity =>

$$\Delta G^{AA} + \Delta G^{WL} = A\sqrt{T} + BT^{p/2}$$

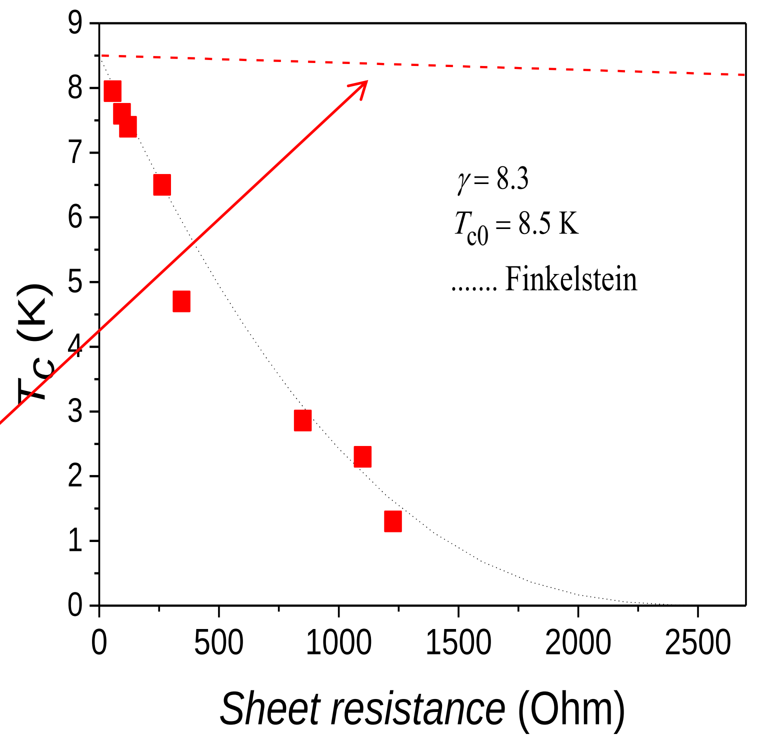
Determination of Thouless energy from B_{c2}

- We measured $B_{c2}(T)$ from $R_S(T,B)$
- We determined $B_{c2}(0) = 0.69(dH_{c2}/dT)T_c$; also $\xi(0) = \sqrt{(\Phi_0/2\pi B_{c2}(0))}$
- Diffusion constant $D = \frac{0.407\pi k_B}{e}\left(\frac{dH_{c2}}{dT}\right)^{-1}$
- transverse Thouless energy $\pi\hbar D/d^2$ determined

=>> **Thouless energy is below 100 K for all MoC films (see Table)**

$k_B T_{c0} < (hD/d^2 \sim 10\, k_B T_c) << \hbar/\tau$

but then the renormalized Finkelstein makes small effect on Tc



(Same arguments hold for MoGe, Graybeal & Beasley, TiN, Baturina, ……)

Effectively Finkelstein mechanism does not work

What is the real mechanism of T_c suppression ?

$k_F l$ from Hall & resistivity measurements

- We measured te Hall coefficient from Hall voltage deduced from field sweeps +- 8T @ 200 K

- Charge-carrier density determined $n \sim 10^{23}$ cm^{-3} not changing upon decrease of thickness (!!)

- Ioffer-Regel product $k_F l$ determined; drops from 4.1 (15nm) to 1.3 (3 nm)

$$k_F l = \frac{\hbar(3\pi^2)^{2/3}}{e^{5/3}}\left[\frac{R_H^{1/3}}{R_\square d}\right]$$

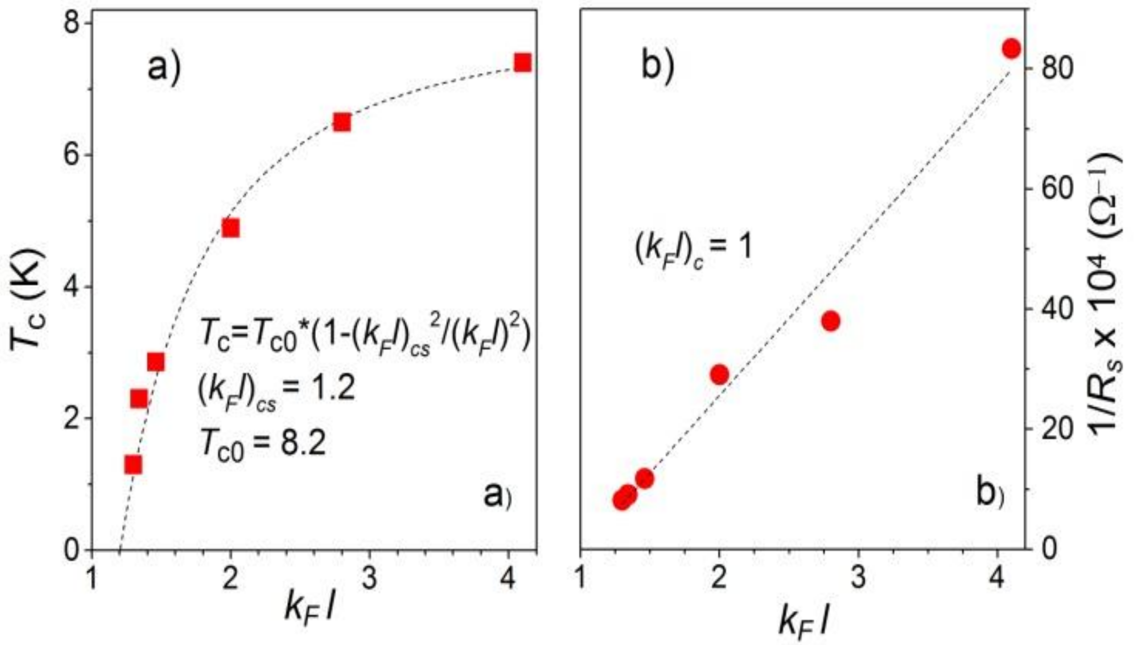
d [nm]	R_s[Ω] @ 288 K	T_c K)	R_H x10^{11} [ΩmT^{-1}]	N*10^{23}cm^{-3}	$k_F.l$	H_{c2}[T]	ξ [nm]	D[cm^2/s]	$\pi^2\hbar D/d^2$ [K]
30	56	7.95	-	-	-	-	-	-	-
20	95	7.6	-	-	-	-	-	-	-
15	120	7.4	3.75	1.7	4.1	10.7	5.48	0.52	17.4
10	263	6.5	3.75	1.7	2.8	9.4	5.78	0.53	40
10	344	4.9	3.13	1.9	2	9.5	5.8	0.39	29.4
5	850	2.86	3.8	1.7	1.46	5.3	7.8	0.39	118
5	1100	2.3	3.8	1.7	1.34	5.3	7.8	0.33	100
3	1227	1.3	3.9	1.7	1.3	-	-	-	-

T_c versus $k_F l$

- We found that T_c follows the best $k_F l$, better than Finkelstein T_c (R_s), even films with the same thickness but different T_c and R_s are well fitted.

- The data can be fit to the Anderson localization model $T_c = T_{c0}(1-[(k_F l)_c{}^2/(k_F l)]^2)$ with $(k_F l)_{cs} \approx 1.2$ and $T_{c0} = 8.2$ K.

- sheet conductance $1/R_S$ as a function of $k_F l$ is almost linear with $(k_F l)_c \rightarrow 1$



$(k_F l)_{cs}$ **>** $(k_F l)_c$ => first SMT and then MIT

II. Local DOS by STM/S

STM NEAR SIT TRANSITION

Superconductivity:
macroscopic wave function
with amplitude Δ and phase φ

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

Fermionic mechanism

Disorder-enhanced Coulomb int.
destroys Copper pairs => **SMT**
At higher disorder bad metal goes
to Fermi insulator via **MIT**
Amplitude fluctuations

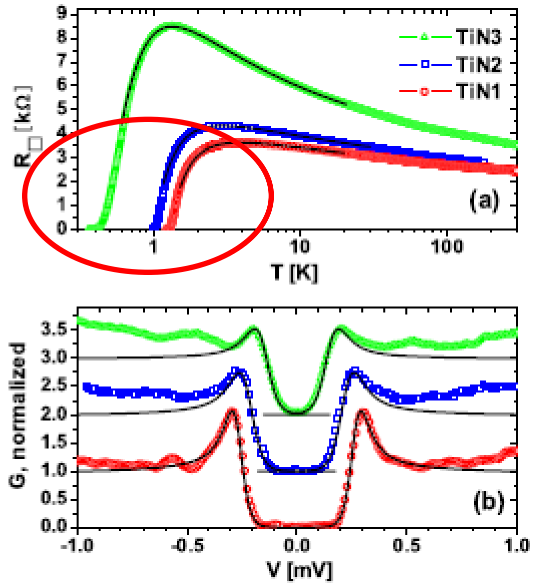
Bosonic mechanism

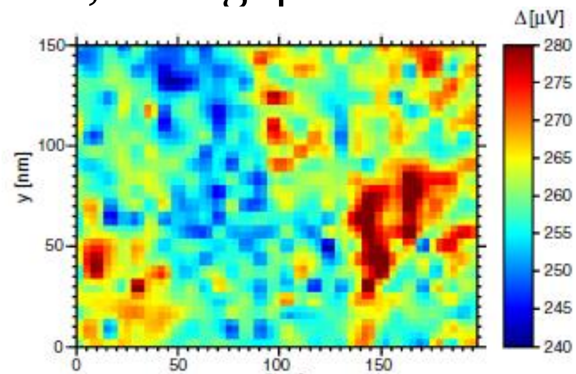
Superconductor => directly to insulator **SIT**
Cooper pairs survive with finite Δ
without long range phase coherence
Phase fluctuations

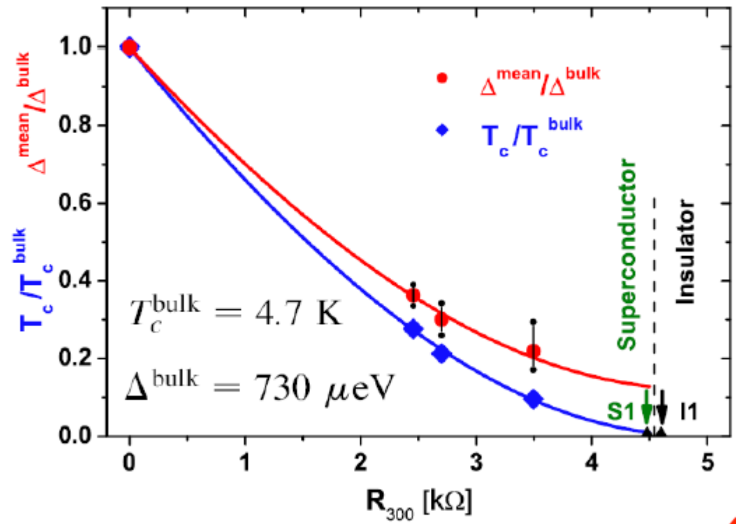
Local studies of superconductivity (Δ) by
Scanning Tunneling Spectroscopy are chalenging

Scanning tunneling spectroscopy in ultrathin TiN films

Film thickness 3-5 nm, coherence length ξ=10 nm, strong quantum corrections







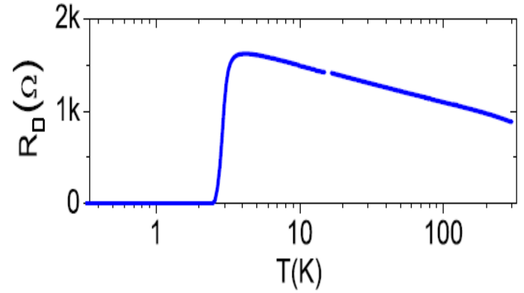
	R_{300} kΩ	T_c K	$\bar{\Delta}$ μeV	σ μeV	$\sigma/\bar{\Delta}$	$\bar{\Delta}/k_B T_c$
TiN1	2.45	1.3	265	11	0.04	2.37
TiN2	2.7	1.0	220	13	0.06	2.55
TiN3	3.5	0.45	160	⋯	⋯	4.13

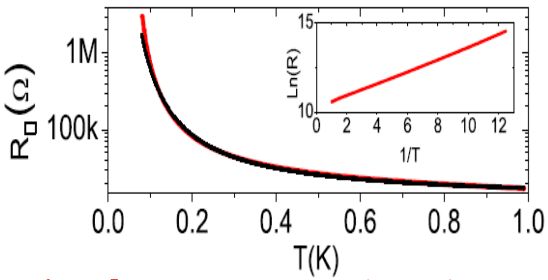
Disorder driven:
decrease of T_c, inhomogeneity of gap Δ (20%)
increased Δ/ kT_c : T_c =>0 *while* Δ remains finite

Sacepé et al., PRL2008

Planar tunnel junctions on real insulating film compared with superconducting film

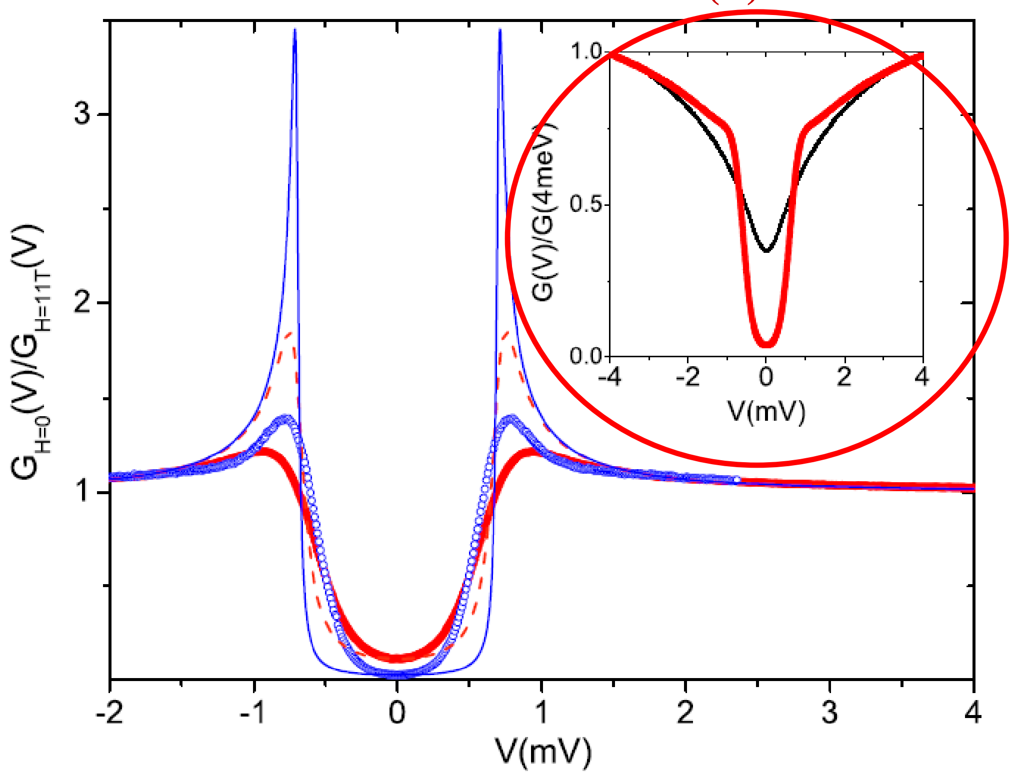
superconductor, $T_c \sim 3$ K





insulator, $R = R_{s0} \exp(T_0/T)$, $R_{s0} = 11.5$ kΩ

Raw dI/dV(V) not normalized



Gap remains about same in insulating phase, but coherence peaks are missing

amorphous InO_x films

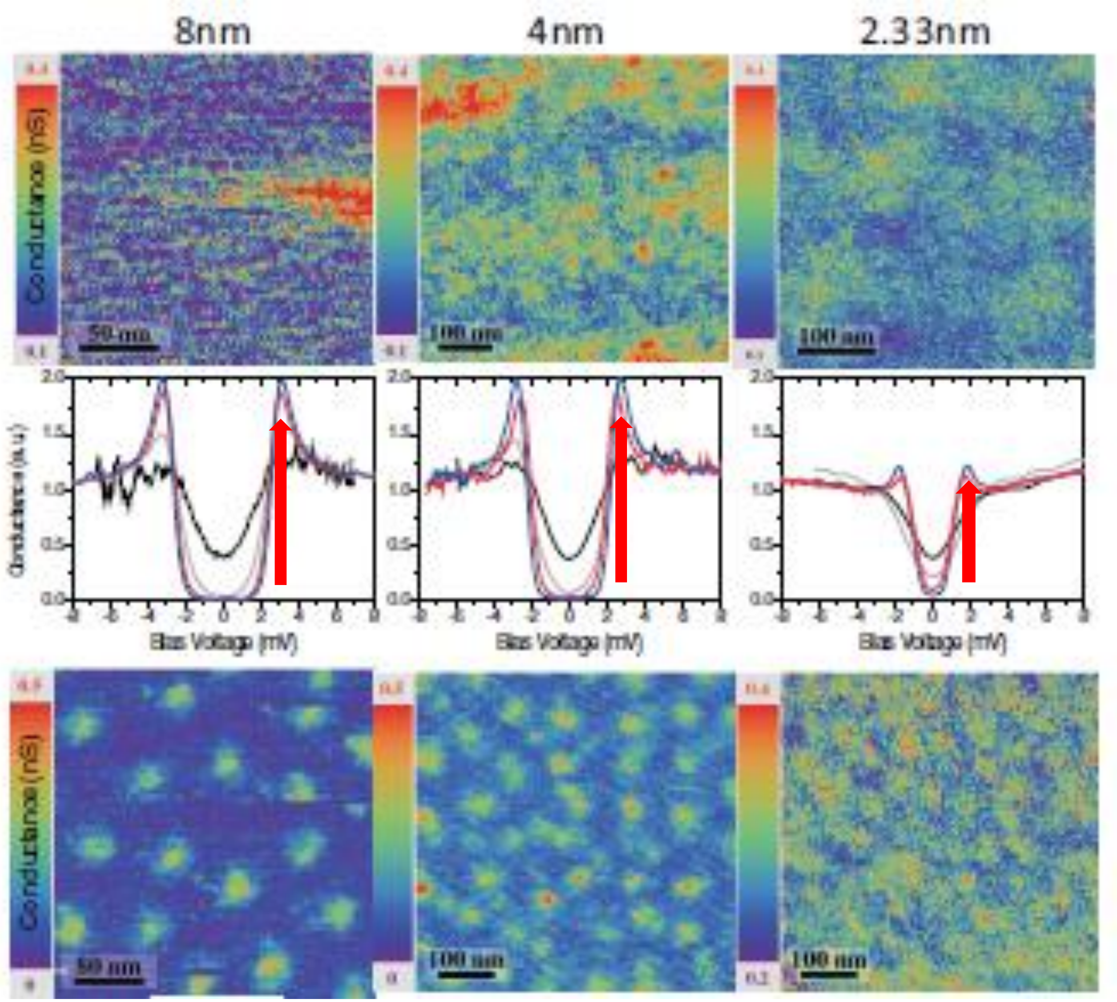
Sherman, PRL 2012

NbN films studied by Roditchev

Tunnel Cond. @ gap

Inhomogeneities

Vortex images



Fading out of vortex image
Lost phase coherence

STS phenomenology

On approach to SIT

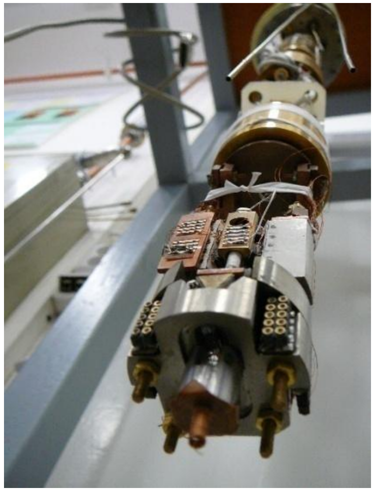
- ✓ Δ decrease more slowly than T_c, $2\Delta/k_BT_c$ increase
- ✓ Δ inhomogeneity on scale of ξ
- ✓ pseudogap appearance
- ✓ coherence peaks in SC DOS suppression
- ✓ Fading out of vortex image

All support bosonic scenario

- **Is bosonic scenario universal ?**
- **Or scenario depends on material parameters ?**
- **If yes, how ?**

STM - experimental setup in Košice

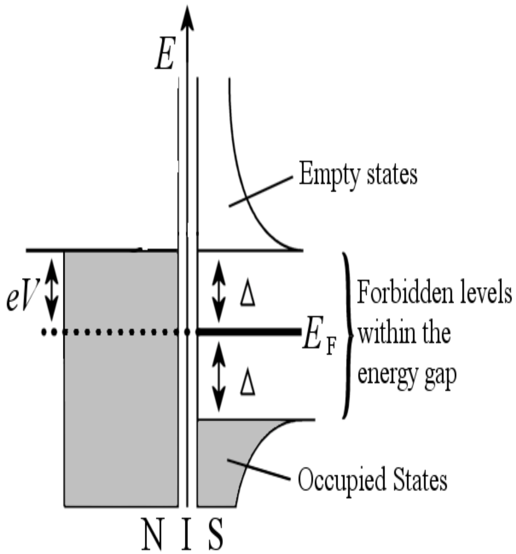
- **home made STM head**
- **Dulcinea SPM controller by Nanotec**
- **280mK: Janis SSV 3He refrigerator**
- **8T Janis cryomagnetic system**



Scanning Tunneling Spectroscopy (STS)

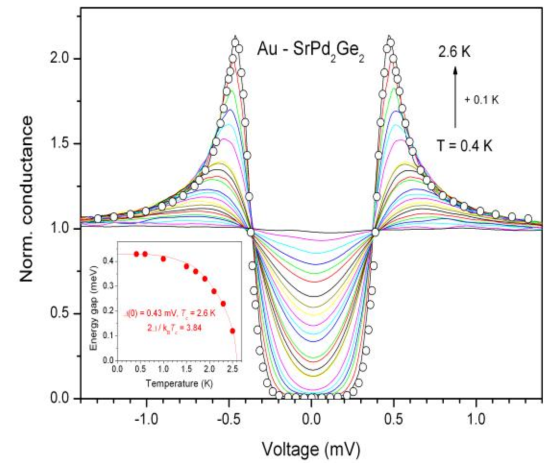
S-I-N junction with Au tip

$$dI(V)/dV \propto A \int N_S(E)\,[\partial f(E-eV)/\partial f(eV)]\,dE$$



$$N_S^{BCS} = \mathrm{Re}\left\{\frac{E}{\sqrt{E^2-\Delta^2}}\right\}$$

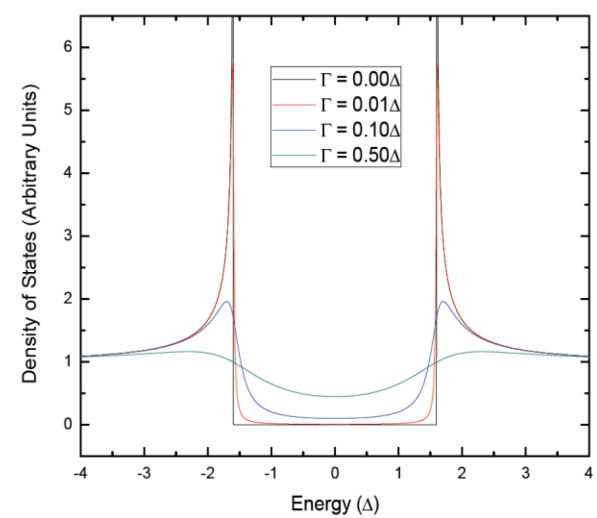
At low *T dI/dV* is proportional to the LDOS of SC



Low temperatures are important to resolve from

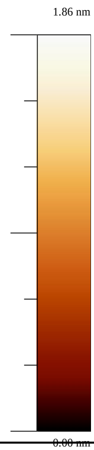
Finite lifetime effect (Dynes formula):

$$N_S^{BCS} = \mathrm{Re}\left\{\frac{E-i\Gamma}{\sqrt{(E-i\Gamma)^2-\Delta^2}}\right\}$$

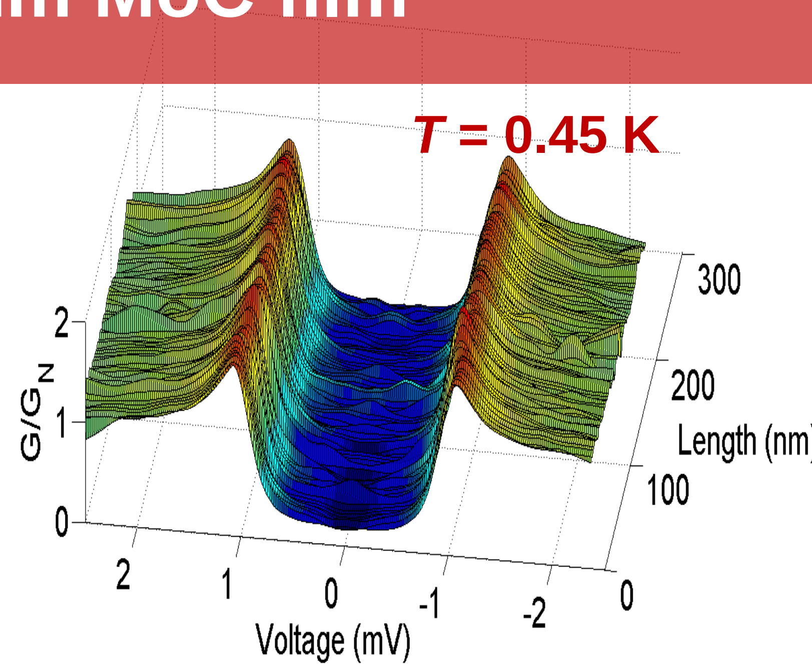


STM on 10 nm MoC film

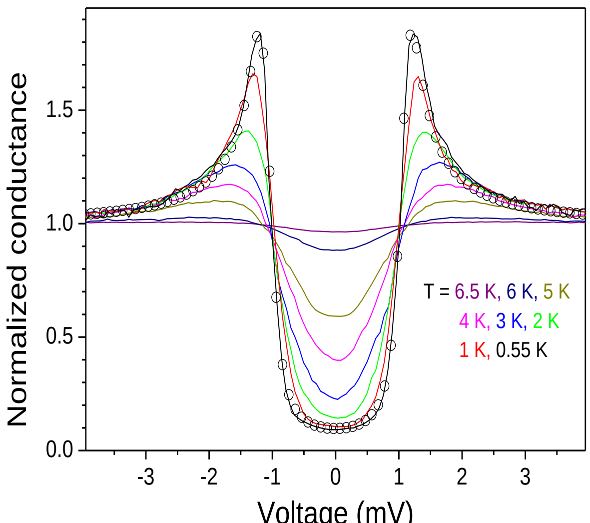
***T* = 0.45 K**



Polycrystalline films, oblate Xstal ~ 20 nm

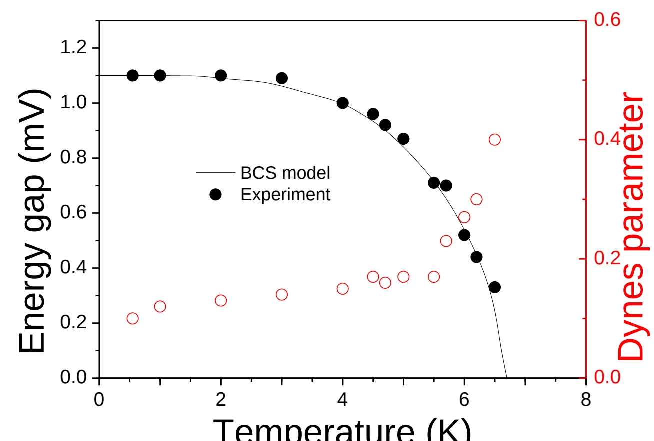


100 spectra along 200 nm line
very homogenous, Δ variations less than 5%



pronounced gap peaks
In-gap states ~ 10%
$2\Delta/k_B T_c = 3.8$

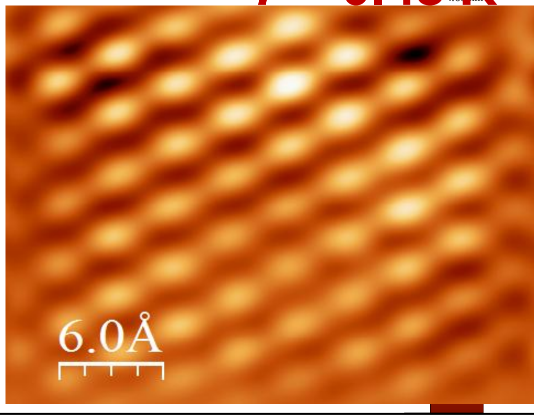
$$N_S^{Dynes} = \mathrm{Re}\left\{ \frac{E - i\Gamma}{\sqrt{(E - i\Gamma)^2 - \Delta^2}} \right\}$$



$\Delta(T)$ BCS-like, broadening Γ increases with T

5 nm MoC film

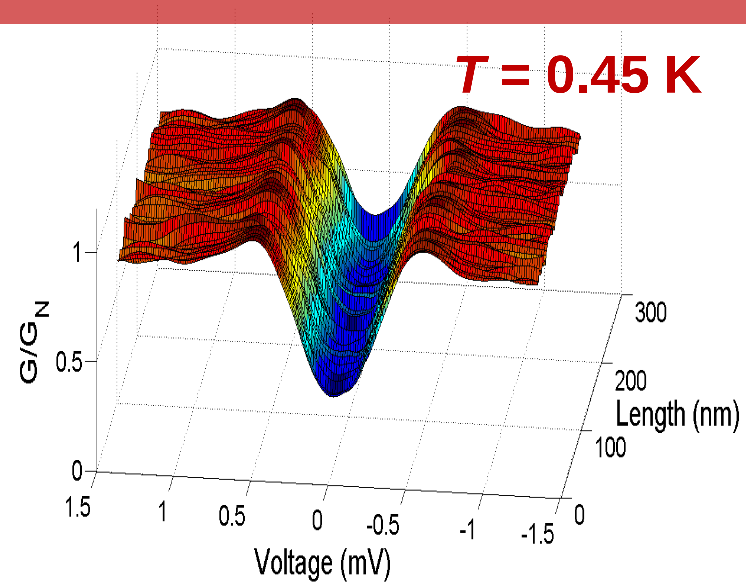
T = 0.45 K



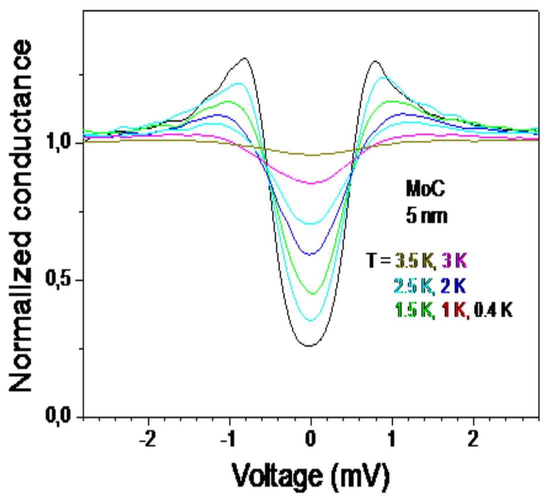
atomic lattice
Γ not from some dead layer

Polycrystalline films, oblate Xstal ~ 10 nm
corrugation ~ 0.7 nm

T = 0.45 K



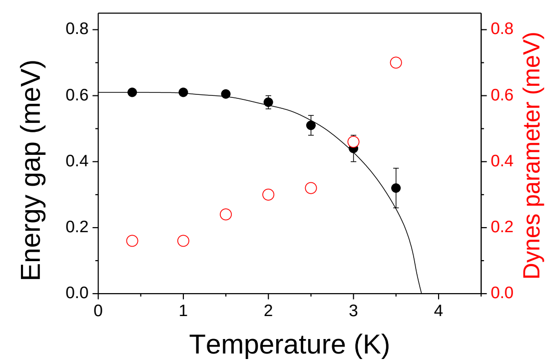
Up to 100 spectra along 200 nm line
Variation of the peak distance ~ 10% (see further)



gap peaks
In-gap states ~ 30% σ_n
$2\Delta/k_BT_c$ = 3.8

$$N_S^{Dynes} = \mathrm{Re}\left\{\frac{E - i\Gamma}{\sqrt{(E - i\Gamma)^2 - \Delta^2}}\right\}$$

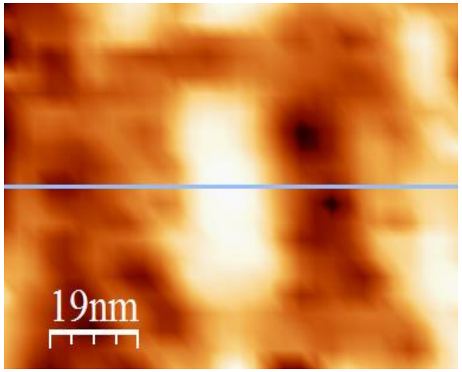
$$\Gamma = 0.3\Delta$$



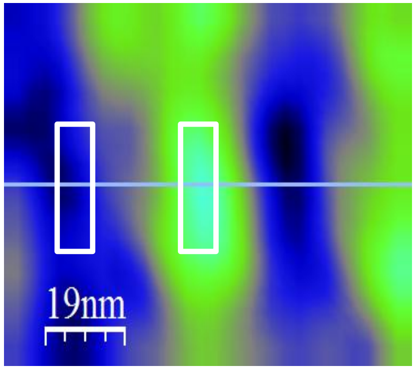
$\Delta(T)$ BCS-like, broadening Γ increases with T

Origin of variation of the gap value in 5 nm MoC

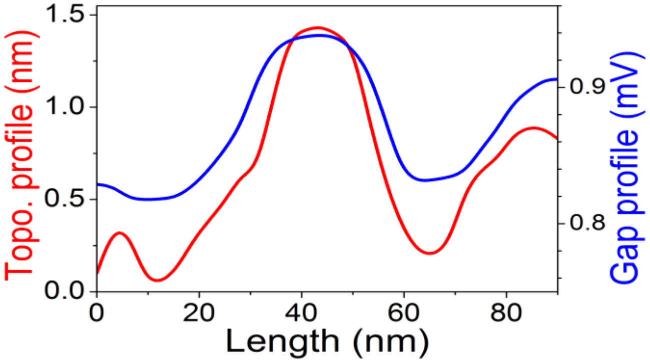
95 x 65 nm^2 topography



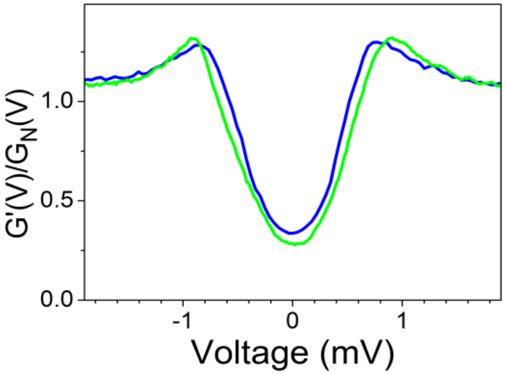
gap map of same area



Corrugation of 1.5 nm
and gap profile (10% variation)
along gray lines



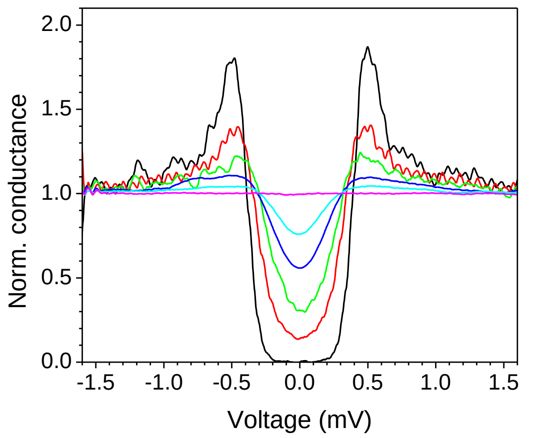
Spectra averaged over
green & blue rectangles

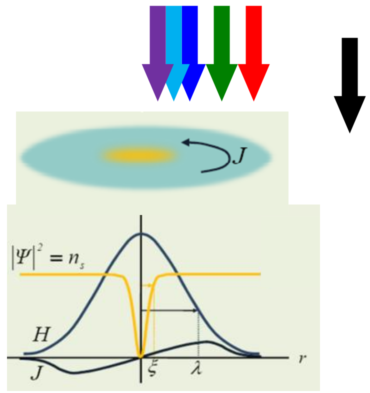


Variation of gap-peak position follows surface corrugation !

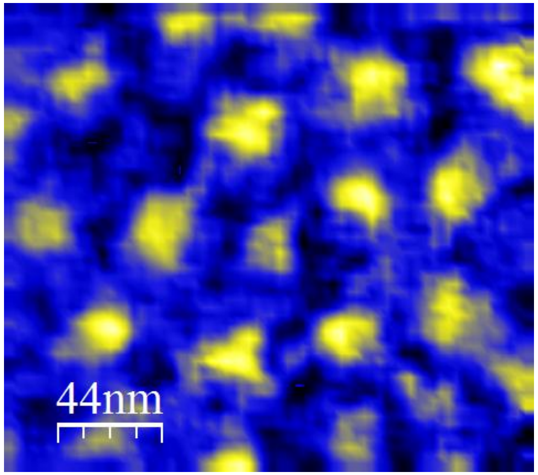
Vortex imaging on 5 nm MoC film

Spectrum along vortex in dirty SC





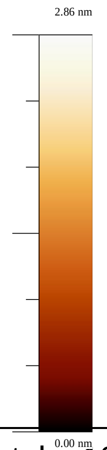
surface area 220 x 190 nm²
***T* = 450 mK, *B* = 1 T, Abrikosov lattice *a* = 50 nm**
distorted vortex lattice,



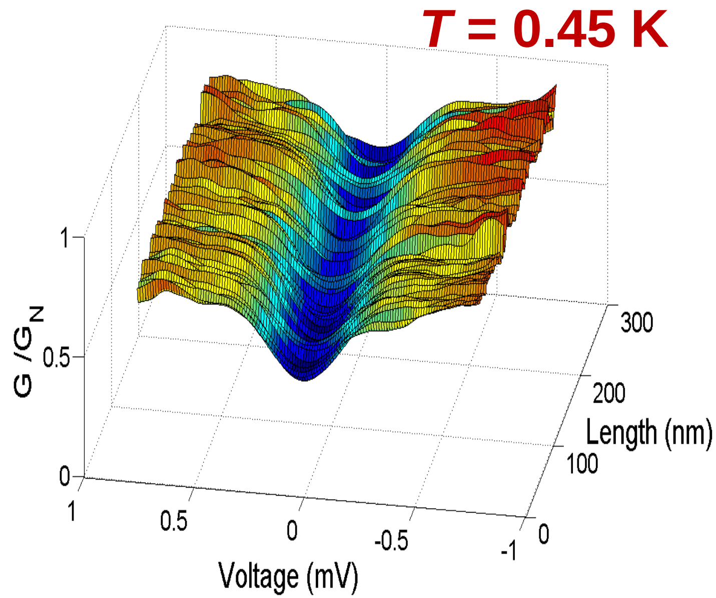
presence of vortices in MoC suggests
long range phase coherence of the superconducting condensate

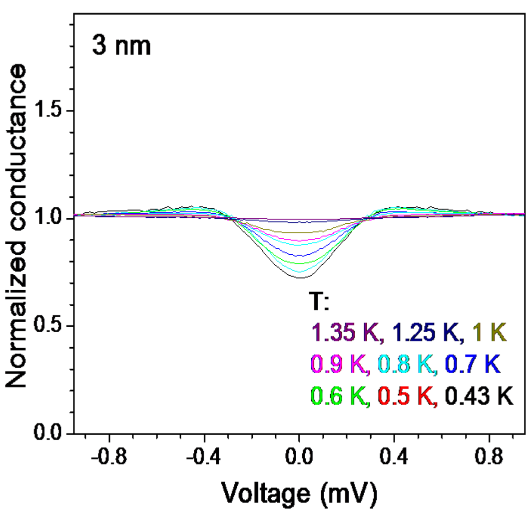
3 nm MoC film

***T* = 0.45 K**



Polycrystalline films, oblate Xstal ~ 10 nm





- 100 spectra along 200 nm line
- variations among spectra ?
- *V*-shape background appears
- suppressed gap peaks but still Dynes works
- in-gap states ~ 80% σ_n

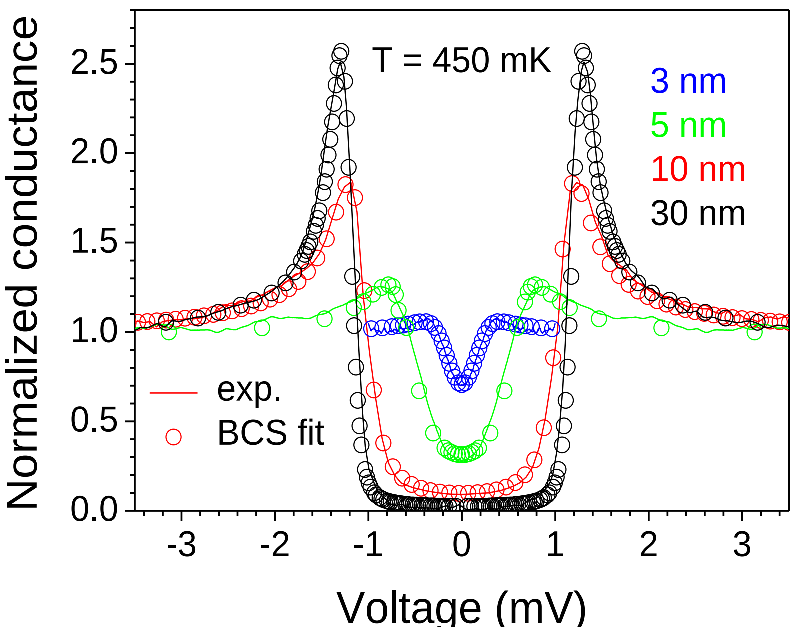
$$N_S^{Dynes} = \mathrm{Re}\left\{ \frac{E - i\Gamma}{\sqrt{(E - i\Gamma)^2 - \Delta^2}} \right\}$$

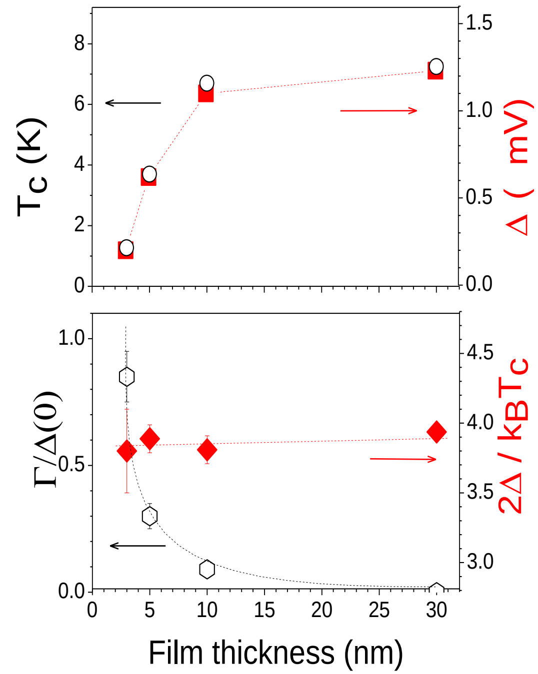
$$\Gamma = 0.8 - 0.9\Delta$$

$2\Delta/k_B T_c$ = 3.5 - 4

Summary of STM/S

We add 30 nm film





STM spectra in MoC upon increased disorder (T_c from 8K down to 1 K & $k_F l$ from 4 to 1)

- Little variation of the gap
- T_c *and* Δ decrease almost same way with $2\Delta/k_B T_c$ ~ 3.8
- No pseudogap
- Presence of vortices suggest global phase coherence

=> *Fermionic scenario !*

- Increasing in-gap states (described by Dynes Γ) =>> what mechanism of pair breaking?

Conclusions

- **Ultrathin films from 30 down to 3 nm thin prepared with T_c decreasing from 8 K down to 1 K, sheet resistances up to 1400 Ohm and $k_F l$ ~1; films are not enough 2D for Finkelstein model**

STM/S

- **sc phase/order parameter is homogeneous in space**
- **T_c *and* Δ decrease in same way with $2\Delta/k_B T_c$ ~ 3.7 – 3.9**
- **Increasing in-gap states (Γ) or gapless SDOS**

(*CWR transmission*

- **Strong pair breaking out of Mattis-Bardeen scenario)**

Questions

- **Mechanism of Tc suppression for films with $l \ll d$?**
- **Mechanism of pair breaking leading to gapless SDOS ?**
- **(Mechanism of strong losses in CWR ?)**


Спасибо

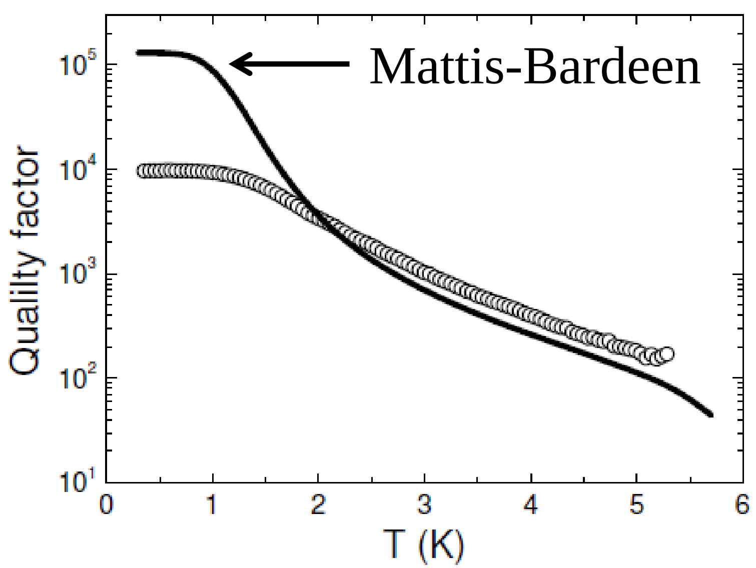
III. Complex conductivity in coplanar wave resonator

Complex conductivity from transmission of CWR

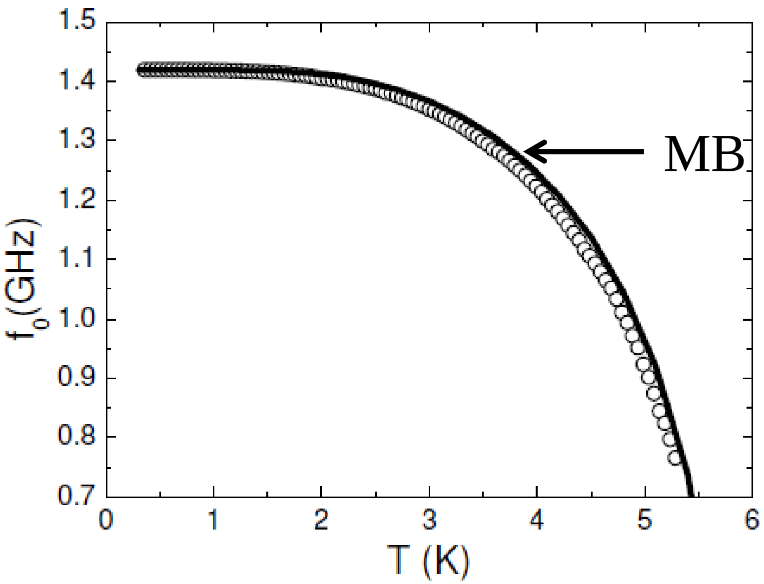
CWR patterned on 10 nm MoC film by optical lithography/etched by ion milling

Transmission measurements => temp. dependence of

- resonance frequency f_0 (~ imaginary part of impedance ~ inductance)
- quality factor Q (~real part of impedance ~ resistive losses)



Losses at low temperatures are much higher than predicted by Mattis-Bardeen
Extra pair breaking present



Resonance frequency falls below Mattis-Bardeen prediction

See also Driessen, Klapwijk PRL 2012
who introduced broadened DOS due mesoscopic fluctuations

M. Žemlička, submitted