Insulating and Superinsulating States in thin TiN Films

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GK 638

Question:

What happens, when the attractive and repulsive components of the Coulomb interaction compete:

- Does first superconductivity and then the metallic state break down -
- or vice versa?

Is it possible to realize a correlated many-body state from *localized* Cooper pairs, which is an insulating analog of the BCS-ground state ?

In 2d superconducting films the zero-resistance state breaks down via the unbinding of thermally excited vortex/anti-vortex pairs:

vortex Berezhinskii-Kosterlitz-Thouless transition

The zero resistance state is characterized by a well defined *phase* and uncertain *number* of Cooper pairs.

Is it possible to realize the inverse case of well defined *number* and uncertain *phase* ?

Yes – in granular films / Josephson networks.

If $E_J < E_C$, numbers are fixed by Coulomb blockade

In such systems a charge Berezhinskii-Kosterlitz-Thouless transition is predicted to exist !

BKT physics in a Josephson junction array

combine $E_p = 2\pi E_J \ln\left(\frac{L}{d}\right)$ and $\Delta S_p = k_B \ln\left(\frac{L^2}{d^2}\right)$

binding energy of a vortex-antivortex pair

entropy of a vortex-antivortex pair

$$\longrightarrow \quad \Delta F = E_p - T\Delta S = (2\pi E_J - 2k_B T)\ln\frac{L}{d}$$

$$\longrightarrow \quad T_{BKT} = \frac{\pi E_J}{k_B}$$

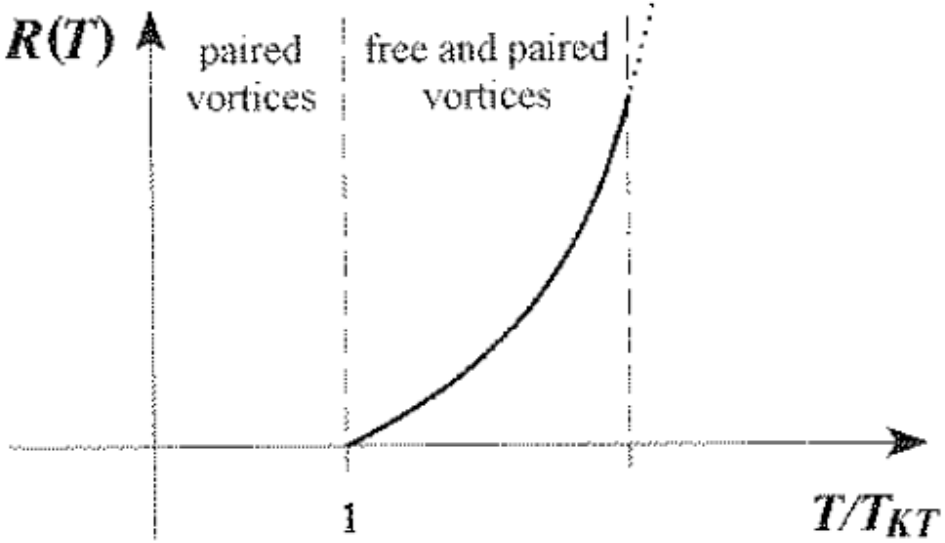
$$\langle V \rangle = R_0 \left(n_f(I) d^2\right) \frac{L}{d} I$$

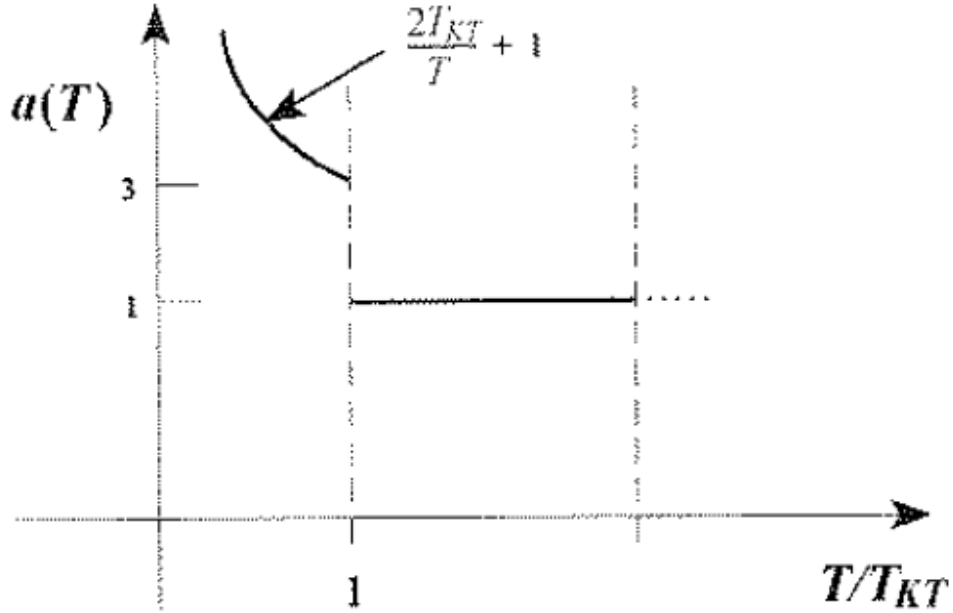
$$n_f = \begin{cases} =0 & T < T_{KT} \\ \geqslant 0 & T \geqslant T_{KT} \end{cases}$$

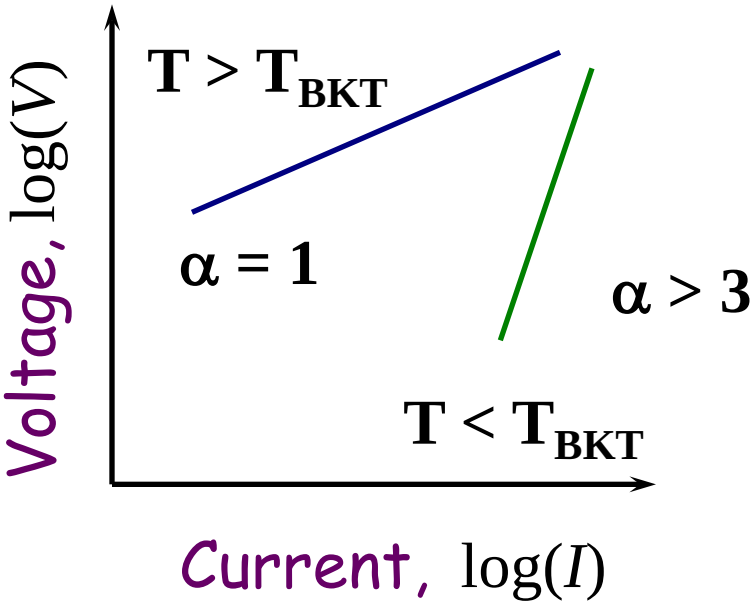
$n_f(I)$:

number of free vortices, depends on current

BKT physics: *I-V* characteristics (vortices)



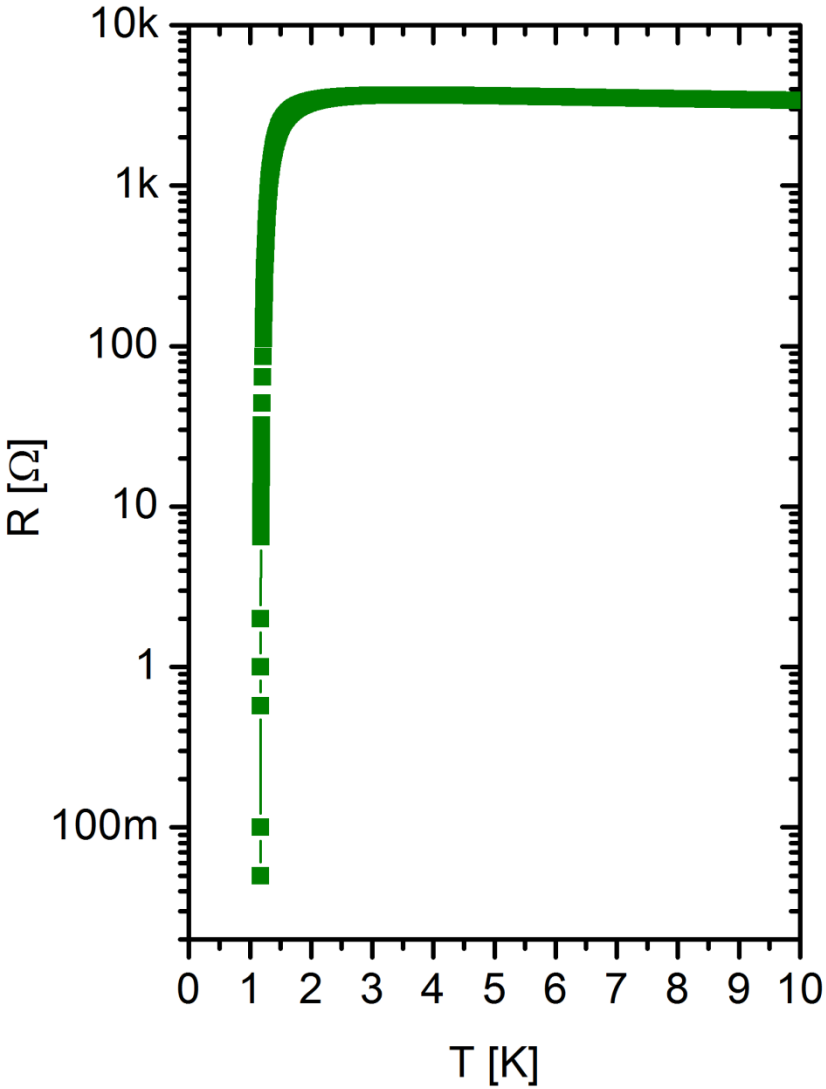




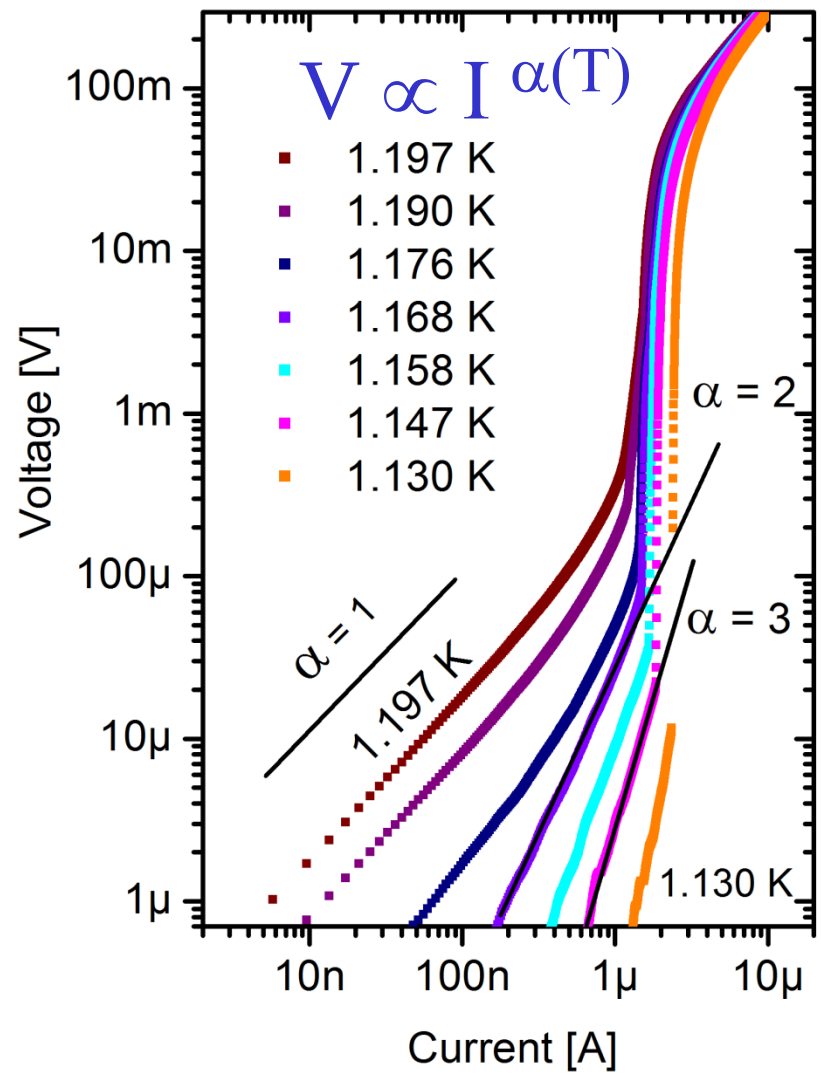
$$V \propto I^{\alpha(T)}$$

Vortex BKT transition in TiN thin films

linear response regime



current - voltage characteristics



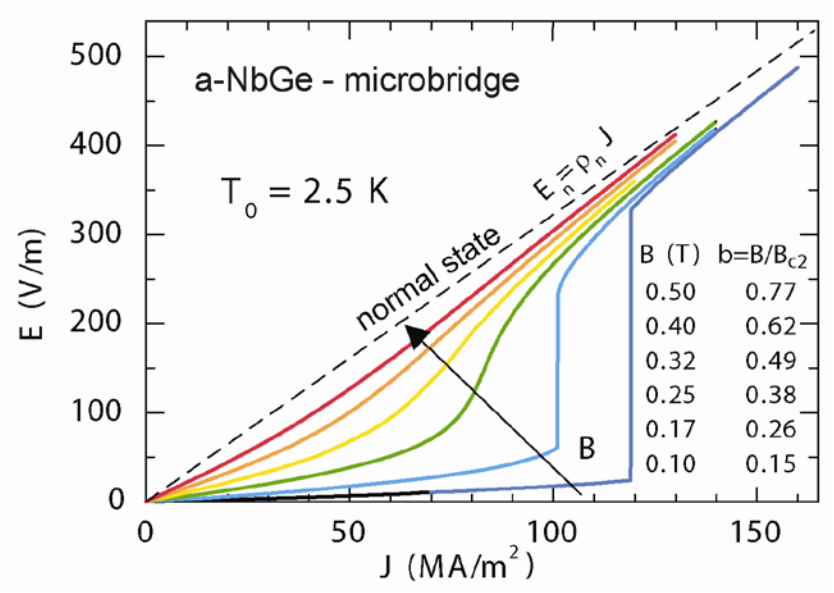
experimental data by A. Mironov and T. Baturina, Novosibirsk

Caution: there may be different types of jumps

Example: Larkin-Ovchinnikov instability
in strongly driven vortex flows

quasiparticles boil off from strongly driven vortex cores

- shrinking of the vortex core
- decrease of vortex viscosity
- incease of vortex velocity and dissipation



(much thicker films
20 nm > ξ)

D. Babic, C.S. et al.
PRB **65**, 645 (2004)

completely different mechanism for jumps in V(I) than BKT

In this talk, 'insulators' are characterized by
an Arrhenius-like R(T) dependence

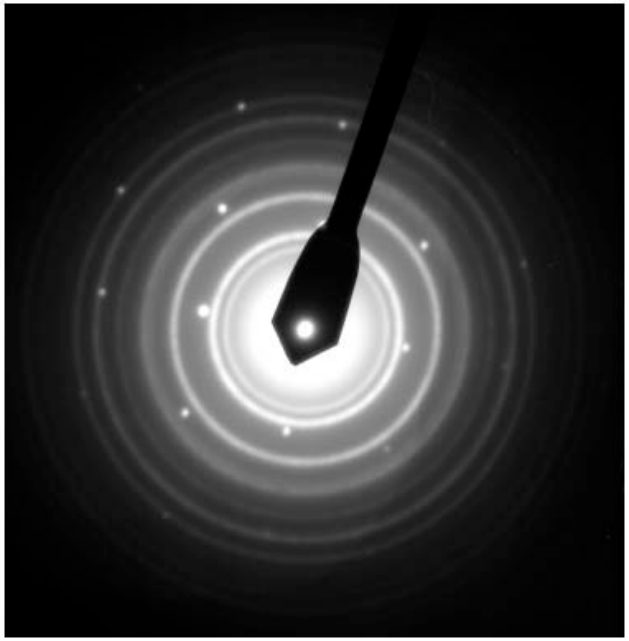
Outline:

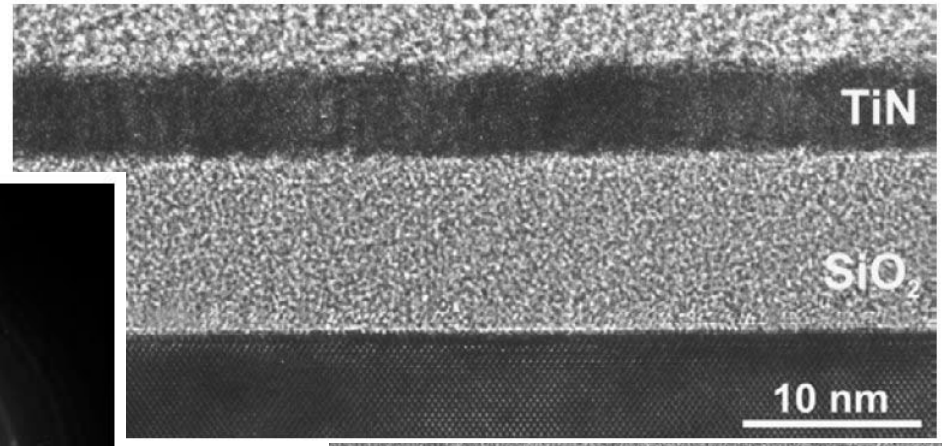
- direct superconductor/insulator transition in thin TiN films
- unusual ‚superinsulating‘ state at very low temperature

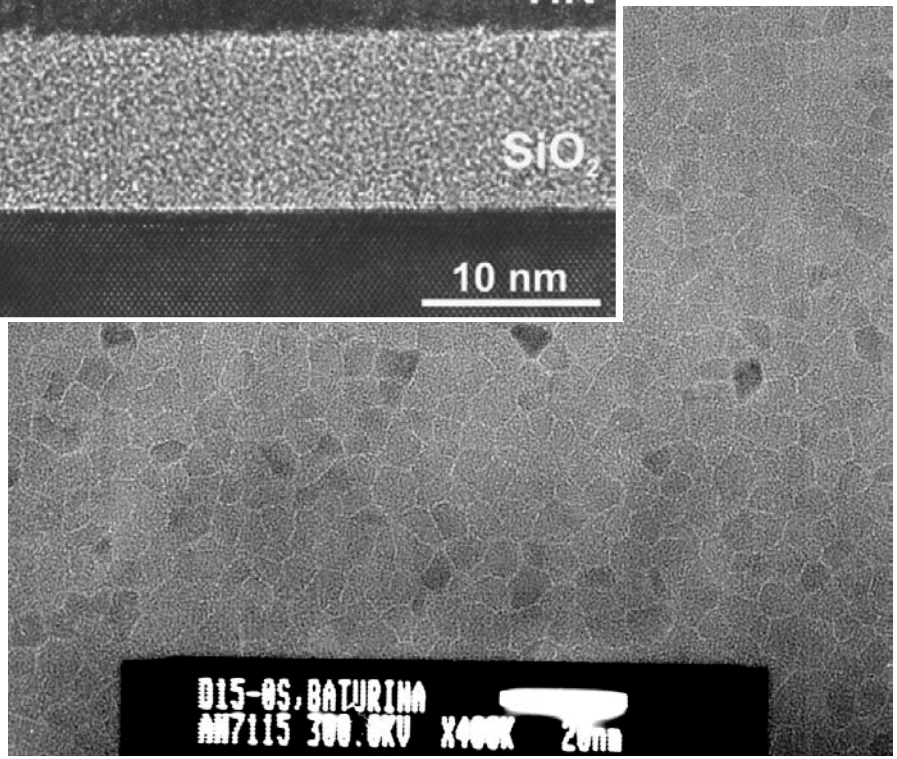
exploring the low-T regime:

- charge Berezhinskii-Kosterlitz-Thouless transition ?
- transition between insulating and superinsulating state:
  dielectric breakdown vs. electron overheating
- size dependence of the resistance per square

Experiments on TiN films







✓ TiN films were formed by atomic layer chemical vapor deposition onto a Si/SiO_2 substrate at 350 ^0C.

✓ Composition:

Ti	N	Cl
1	0.94	0.035

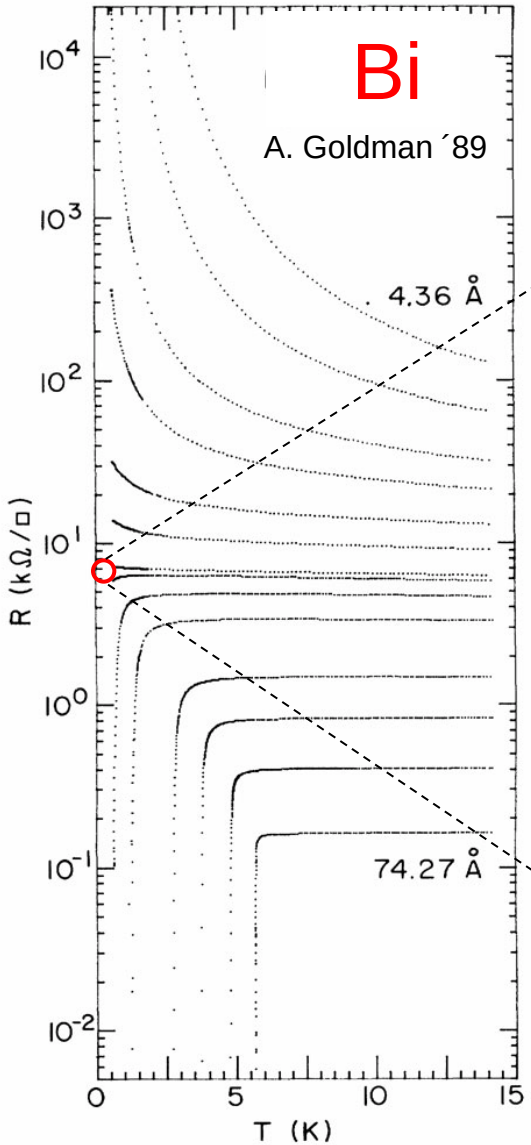
thickness d = 5 nm

carrier density n = 2-4 10^{22} cm^{-3} depending on stochiometry

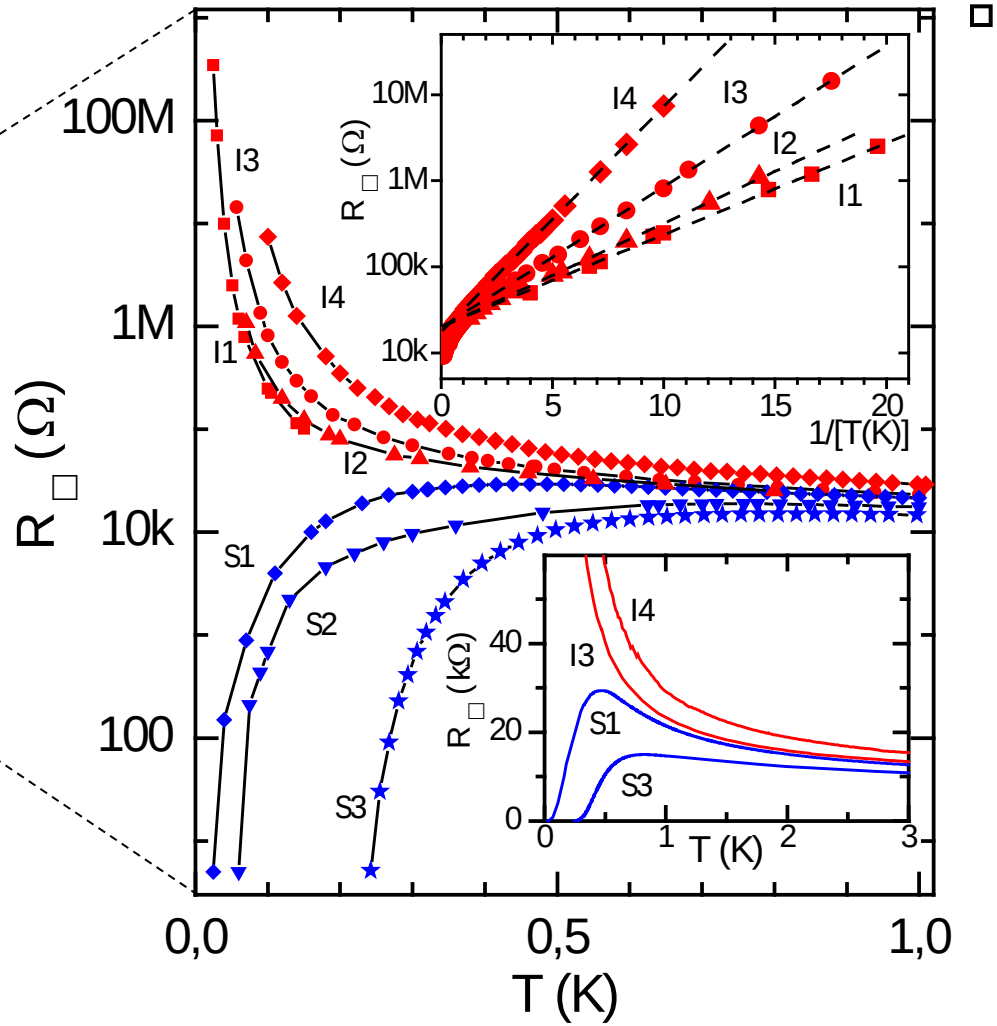
ξ_d ~ 9 nm superconducting coherence length

direct superconductor/insulator transition

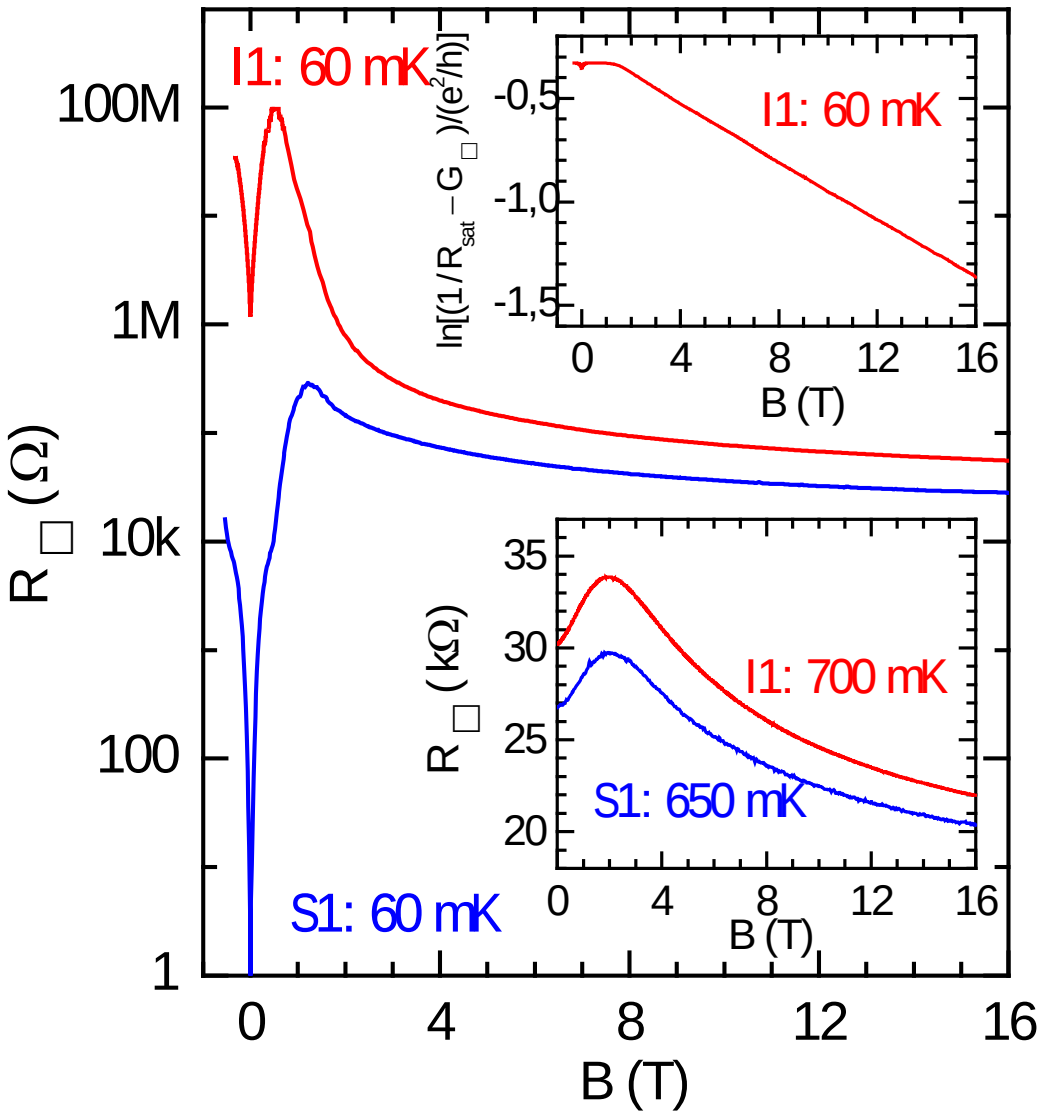
with thermally activated conductance (Arrhenius) on the insulating side



homogeneous TiN-films of 4-18 nm thickness
show a disorder-induced SIT



magnetic field induced resistance enhancement in both: insulating and metallic state



Qualitatively similar behavoir for insulating and superconducting films already in relatively small magnetic fields

Exponential decay of magneto-resistance at high fields:

→ gradual suppression of superconducting OP

Very similar behavior of insulating and superconducting films for T > 600 mK

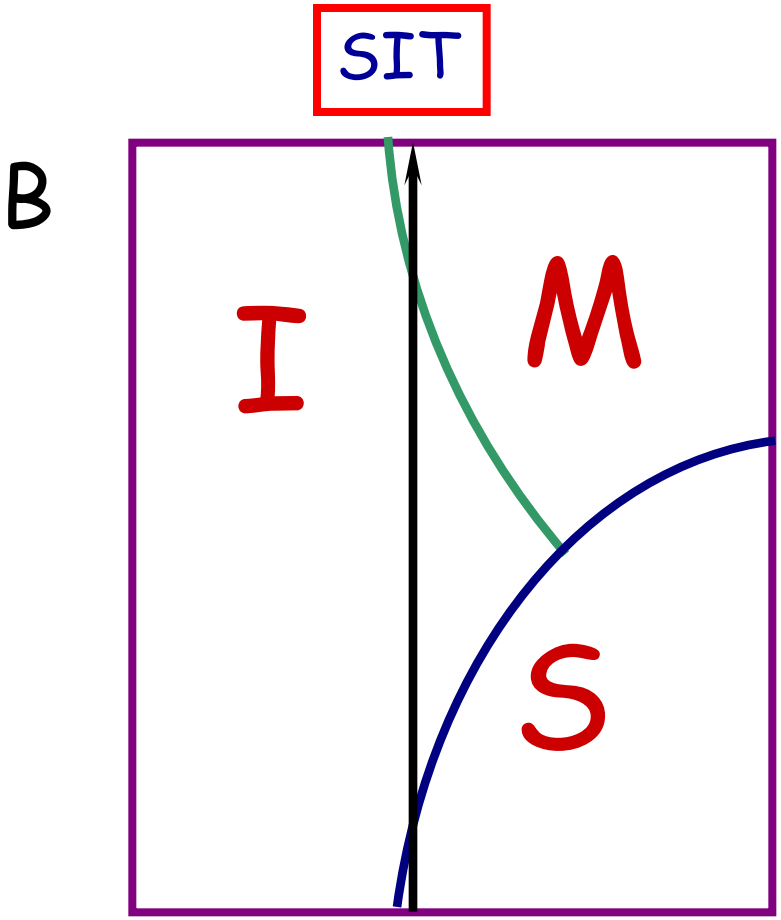
quasi-metallic phase at high B

low B: ‚Cooper-pair' - insulator !

T. Baturina, C.S. et al., PRL **99**, 257003 (2007)

schematic phase diagram

InO_x, Be, and TiN films



Quantum metallicity at a high-field side of SIT

Magnetic-field-tuned superconductor–insulator transition

Disorder-driven superconductor–insulator transition

$g_c = h/4e^2$

Compare

nonlinear transport (IV-characteristics)

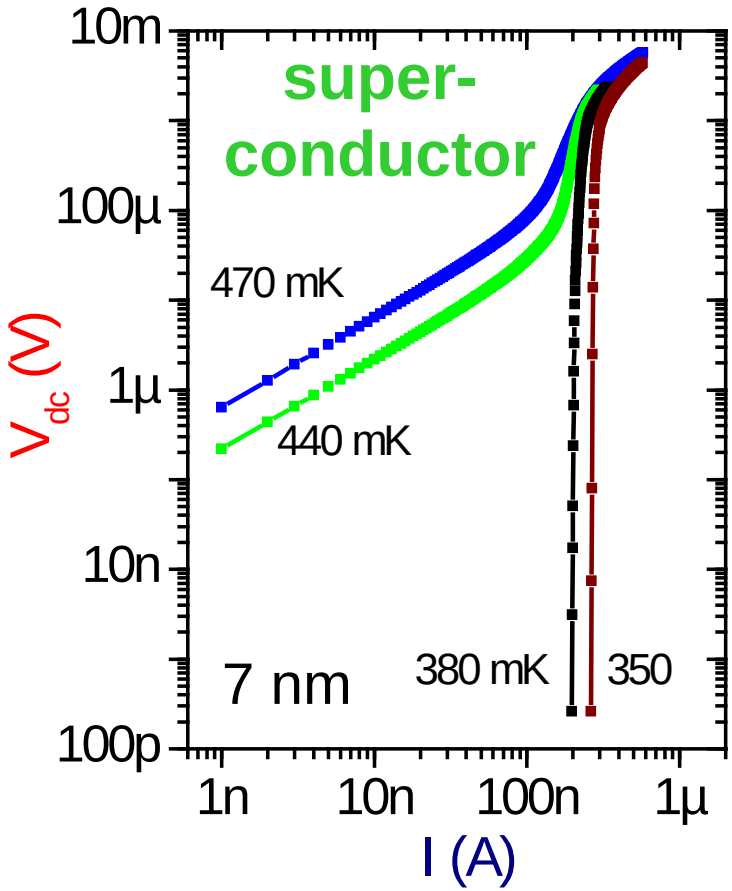
on the

superconducting and insulating

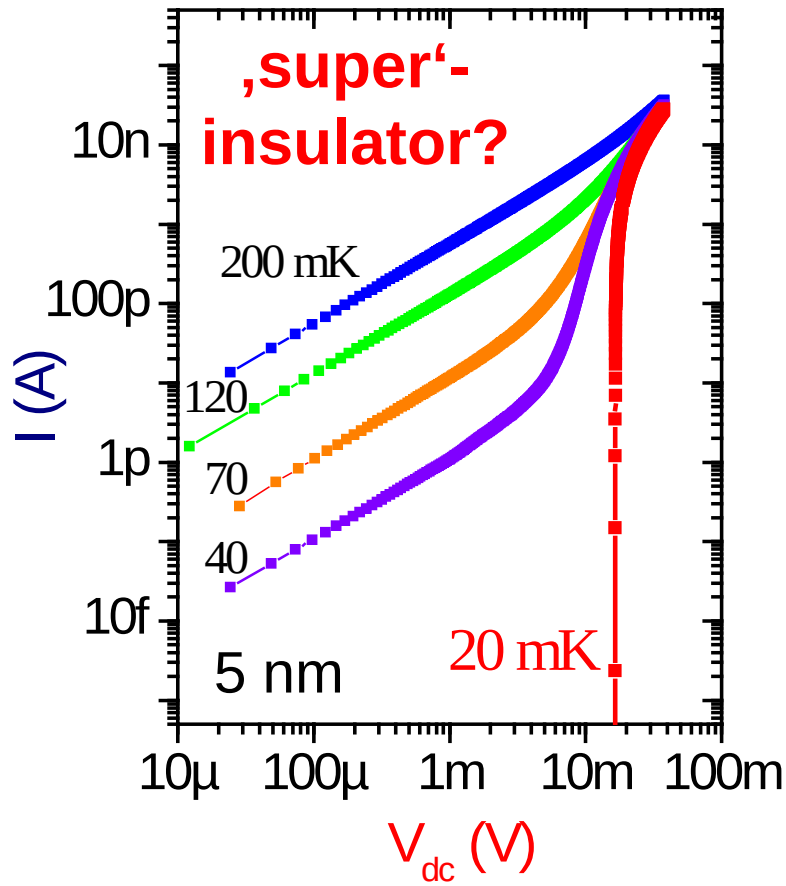
sides of the SIT:

Berezhinskii-Kosterlitz-Thouless transition ?

thermally activated
vortex-antivortex pairs



thermally activated
charge-anticharge pairs ??



Vinokur, Baturina, C.S. et al., Nature **552** (2008)

A similar jump in the IV's has been seen before in amorphous InO_x films in magnetic field :

Sambandamurthy et al. PRL 2004

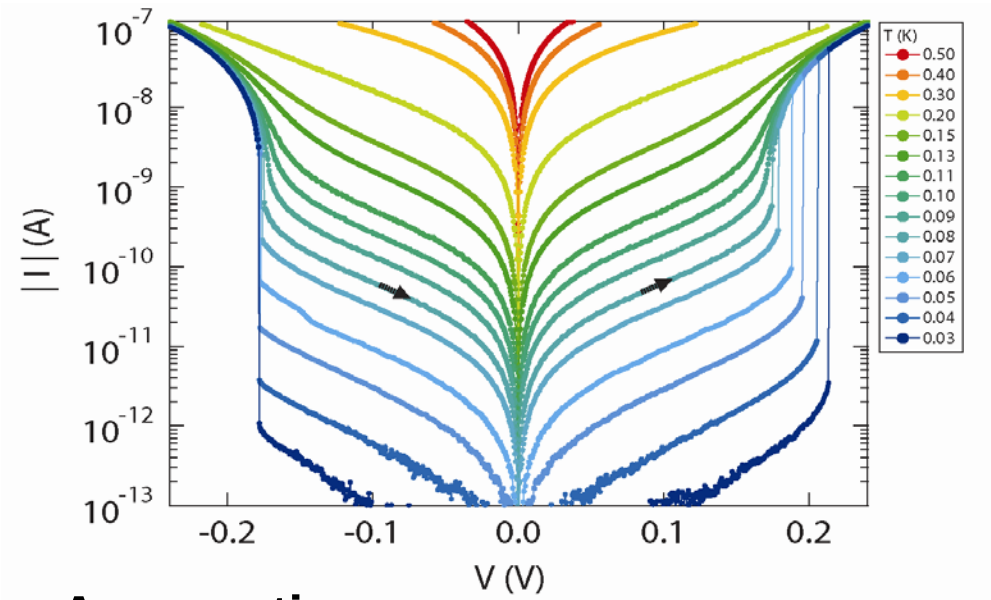
PRL 2005

at Weizmann institute

no clear interpretation at that time

later understood as heating instability (Ovadia et al. PRL 2009)

An important complication : electron heating effects



Volkov & Kogan
Sov. Phys. Usp. **11**, 881 (1969)

Gurevich & Mints,
Rev. Mod. Phys. **59,** 941 (1987)

Ovadia, Sacepe & Shahar
PRL **102**, 176802 (2009)

Altshuler, Kravtsov, Lerner & Aleiner,
PRL **102**, 176803 (2009)

similar to YSi-films (Mott-insulator):

Ladieu, Sanquer & Bouchaud,
PRB **53**, 973 (1996)

Assumptions:

- intrinsic I(V) is linear – no internal energy scales induce non-linearity
- strongly varying R(T) produces heating instability
- decoupling between electrons and phonons (power law IV ~ T^6)

Questions:

- power level 3 orders of magnitude higher than in our TiN-films
- is there really only one energy scale involved (linearity of intrinsic IV‘s ?)
- origin of the highly resistive state?

Phenomenology of Josephson networks

conductance of an artificial Josephson junction array

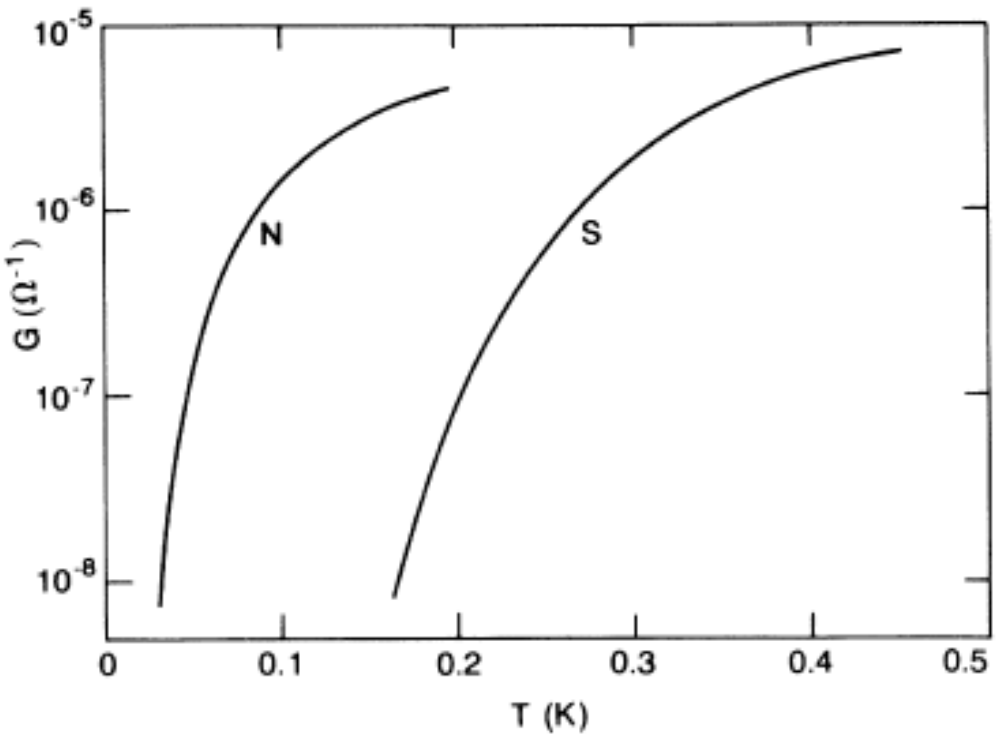


FIG. 3. Measured conductance of an array of $(100\ \mathrm{nm})^2$ aluminum tunnel junctions, 190 cells long and 60 cells wide. *N* is in the normal state (magnetic field of 3T applied); *S* in the superconducting state.

The superconducting array turns insulating at higher temperatures !

J. Mooij, B.J. van Wees, L.J. Geerlings, M. Peters, R. Fazio and G. Schön

Phys. Rev. Lett. **65**, 645 (1990)

H. v.d.Zant, R. Fazio

E. Chow, P. Delsing, D.B. Haviland, PRL 81, 204 (1998).

One-dimensional arrays of small-capacitance Josephson junctions (255, 127, and 63 junctions)

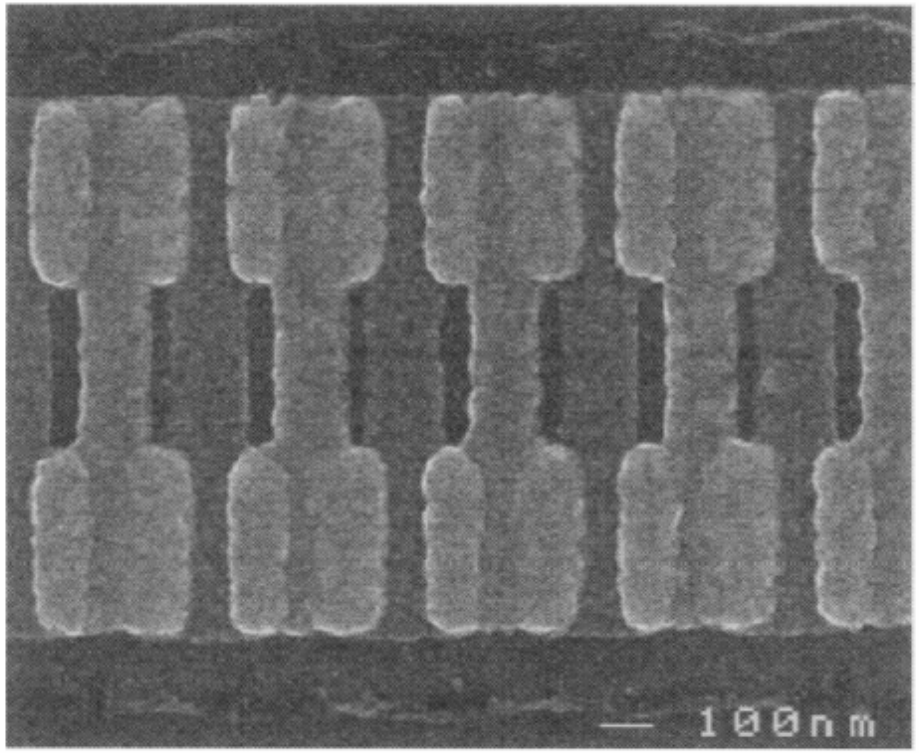


FIG. 1. A scanning electron micrograph of a section of the Josephson junction array. Tunnel junctions are formed at the overlap between the base electrode (darker gray) and the top electrode (lighter gray). The hole between neighboring electrodes forms the SQUID geometry.

$Al/Al_2O_3/Al$ tunnel junctions

$A_{loop} = 0.12\ \mu m^2$ is the effective area of the SQUID loop.

$A \simeq 0.039\ \mu m^2$ is junction area

$R_T = 4.9\ k\Omega \pm 6\%$

$C \simeq 3.5$ fF

electrostatic screening length

$\Lambda \equiv (C/C_0)^{1/2} \simeq 10$

$E_{J0}/E_C \simeq 142\ \mu eV/23\ \mu eV = 6.1$

$$E_J = E_{J0} |\cos \pi B A_{loop}/\Phi_0|$$

1-d JJ – arrays show similar phenomenology !

super-conducting state at low B-fields

insulating state at intermediate B-fields

cusp-like B-dependence of threshold voltage in insulating regime

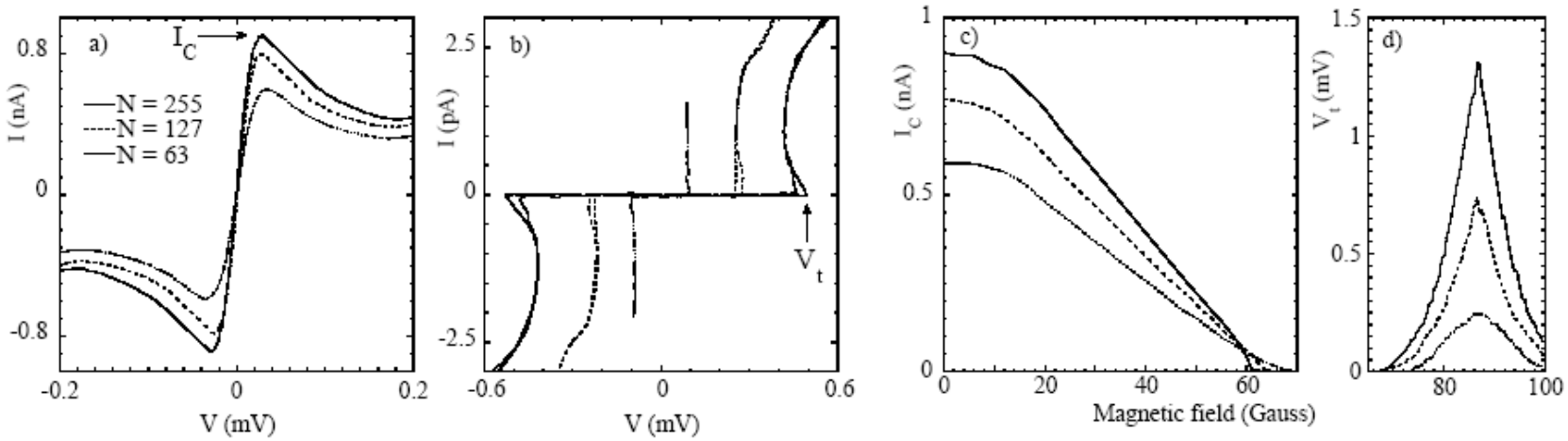
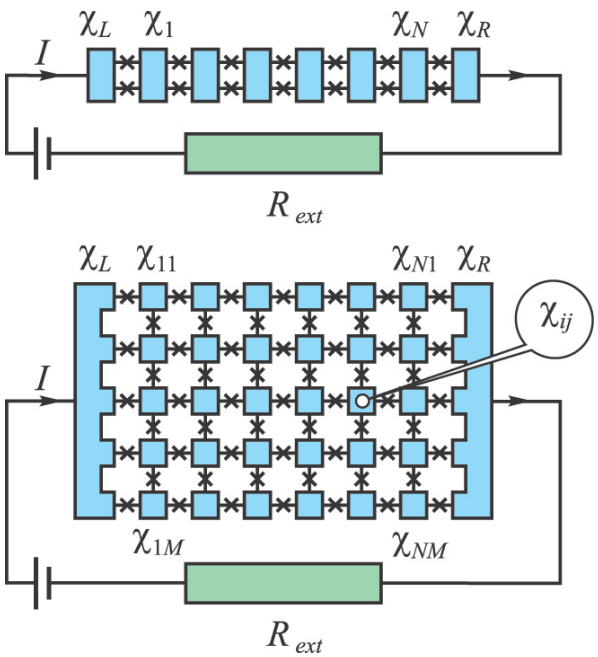
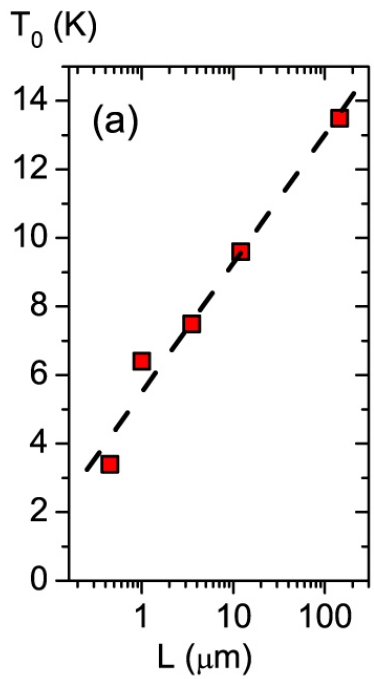


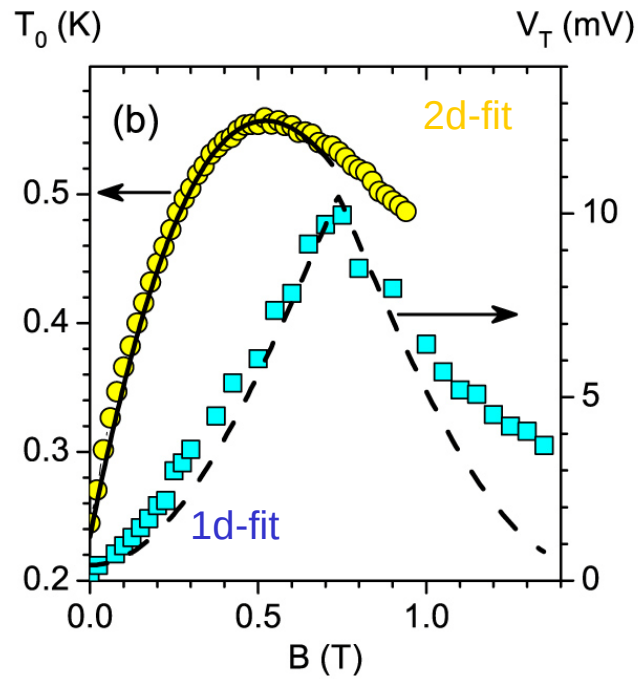
FIG. 2. Dependence of the I-V curves on array length, N, $T = 50$ mK. (a) The I-V curves at $B = 0$ G showing Josephson-like behavior and the critical current I_C. (b) The I-V curves at $B = 71$ G showing the Coulomb blockade of Cooper-pair tunneling and the threshold voltage V_t. (c) The magnetic field dependence of I_C. (d) The magnetic field dependence of V_t.

formation of a collective charging energy

model films as network of Josephson junctions







Kowal & Ovadyahu
Sol.Stat.Comm. '94

M. Fistul,
V. Vinokur, T. Baturina,
PRL **100**, 086805 (2008)

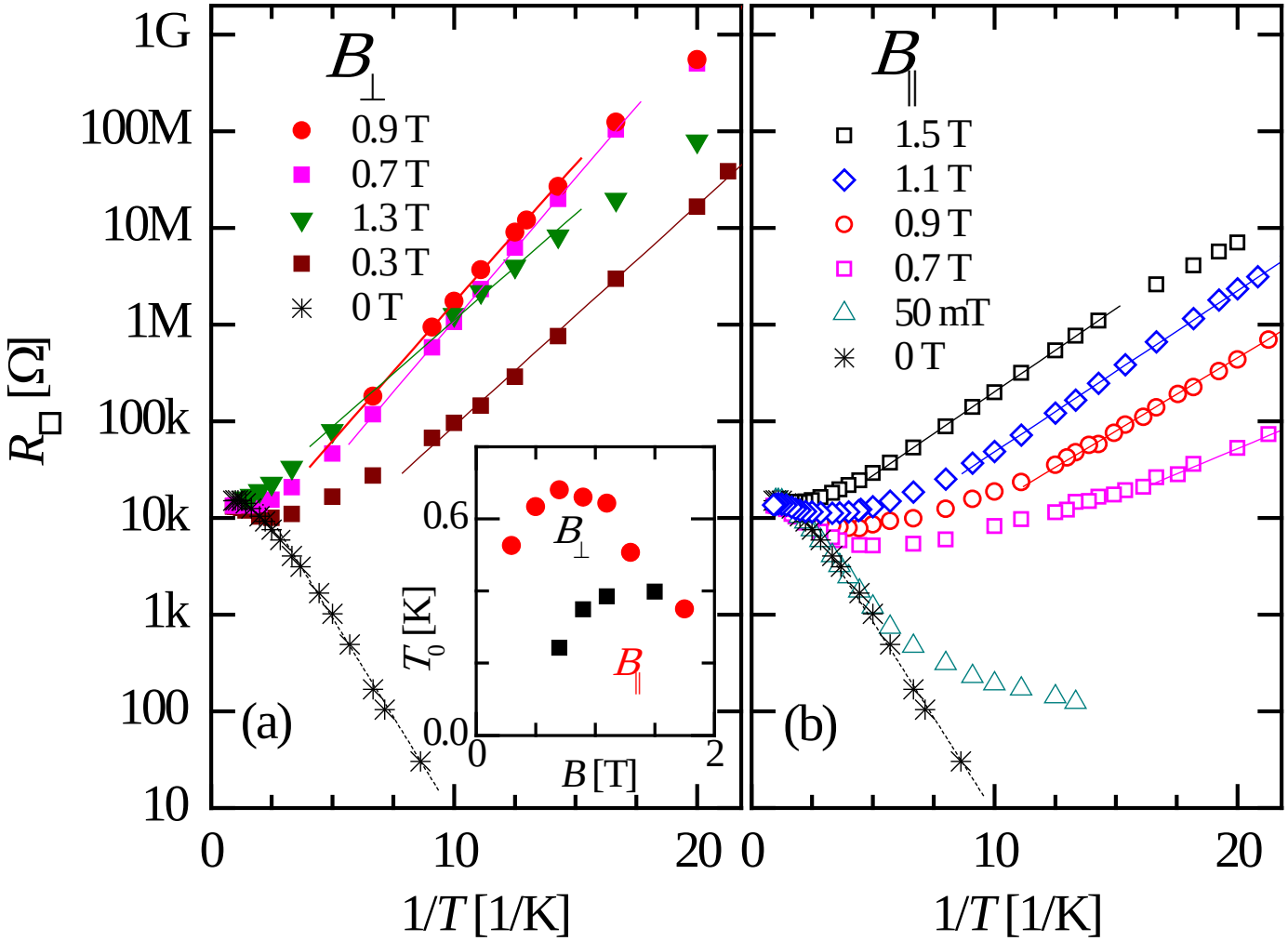
$$\Delta_c(B) = \Delta_c[1 - \alpha E_J(B)/E_c]$$

Assume existence of SC islands in structurally homogeneous films

and formation of a collective Coulomb gap depending on sample size :

nonmonotonic B-dependence of activation energy T_0 and threshold voltage V_T resulting from frustration of Josephson coupling between SC islands

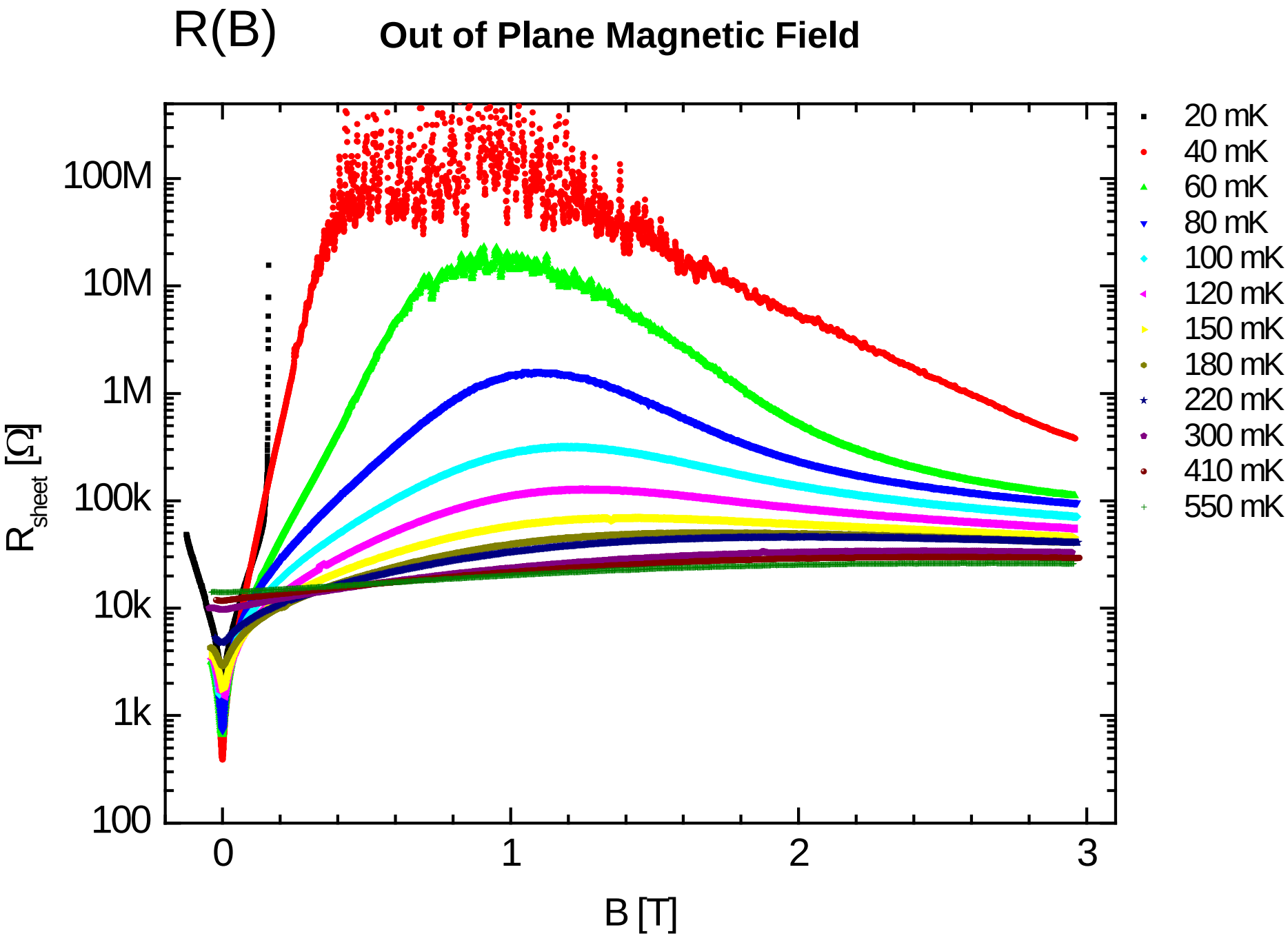
New data on a sample superconducting at B=0



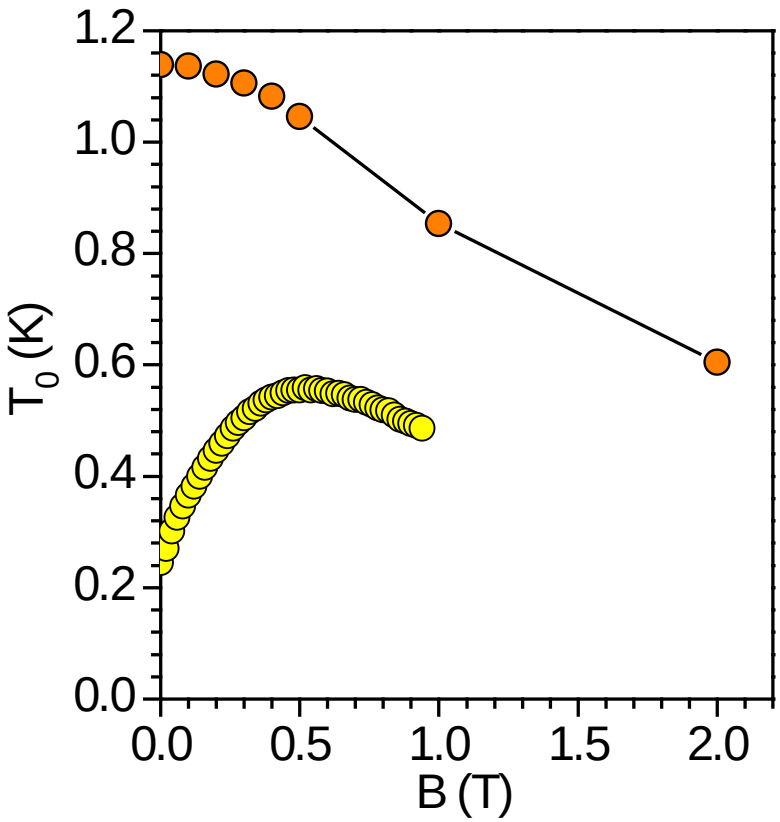
determine R from dc-IV-characteristics:

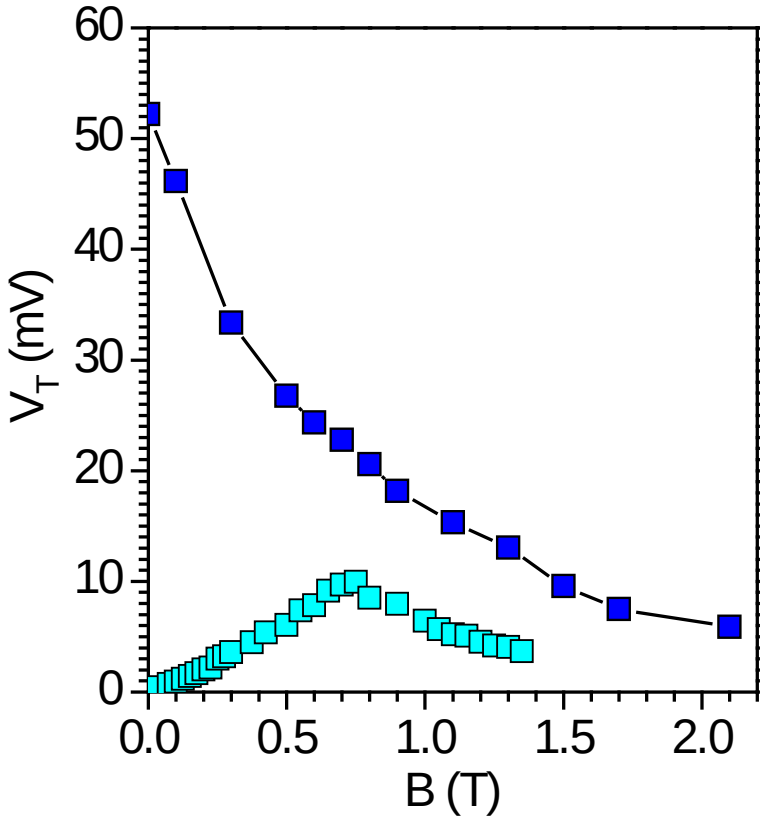
very high resistances accessible!

resistance in perpendicular field much higher than in parallel field


R(B)
Out of Plane Magnetic Field
100M
10M
1M
100k
10k
1k
100
R_{sheet} [Ω]
0
1
2
3
B [T]
20 mK
40 mK
60 mK
80 mK
100 mK
120 mK
150 mK
180 mK
220 mK
300 mK
410 mK
550 mK

Compare samples deeper in the insulating regime

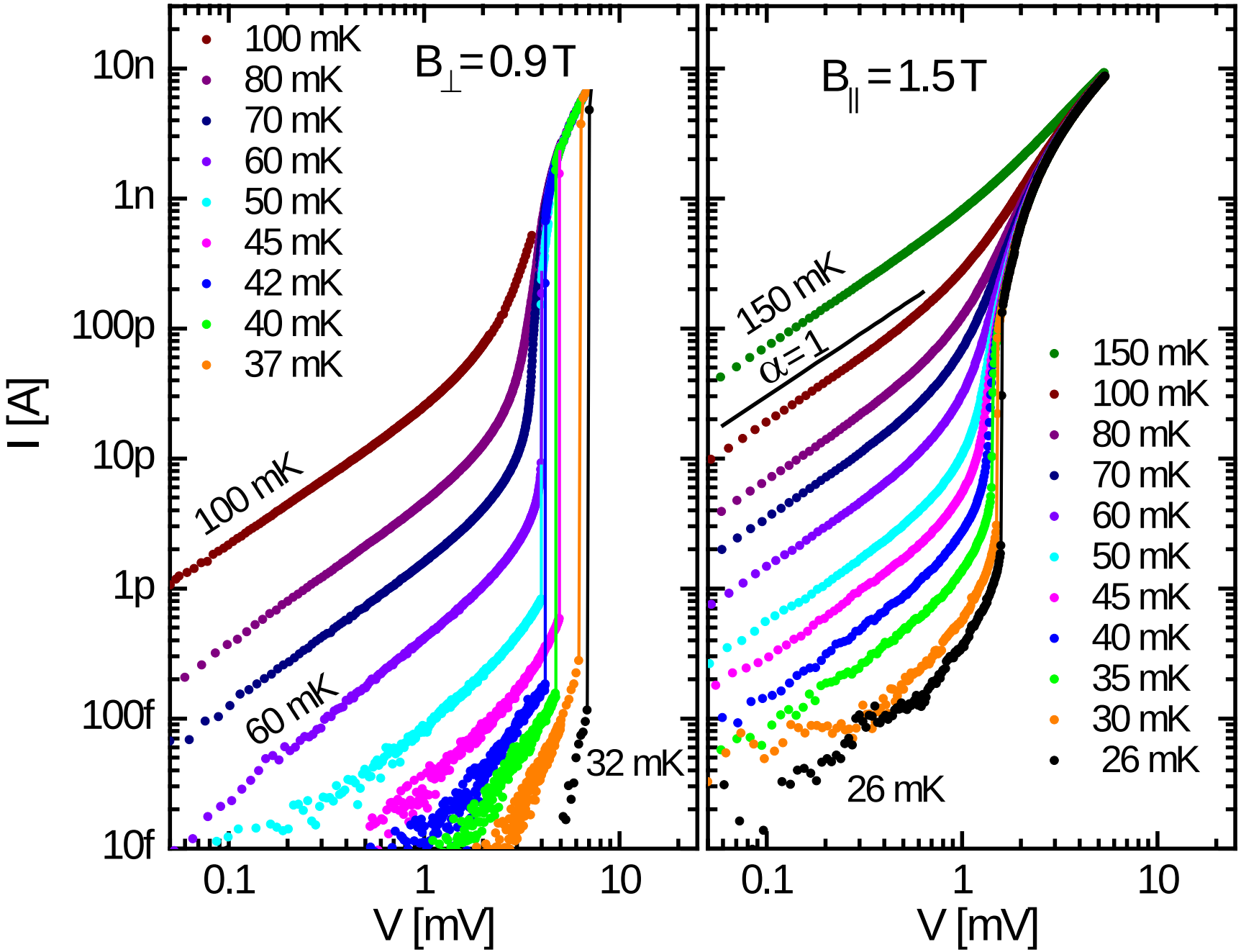




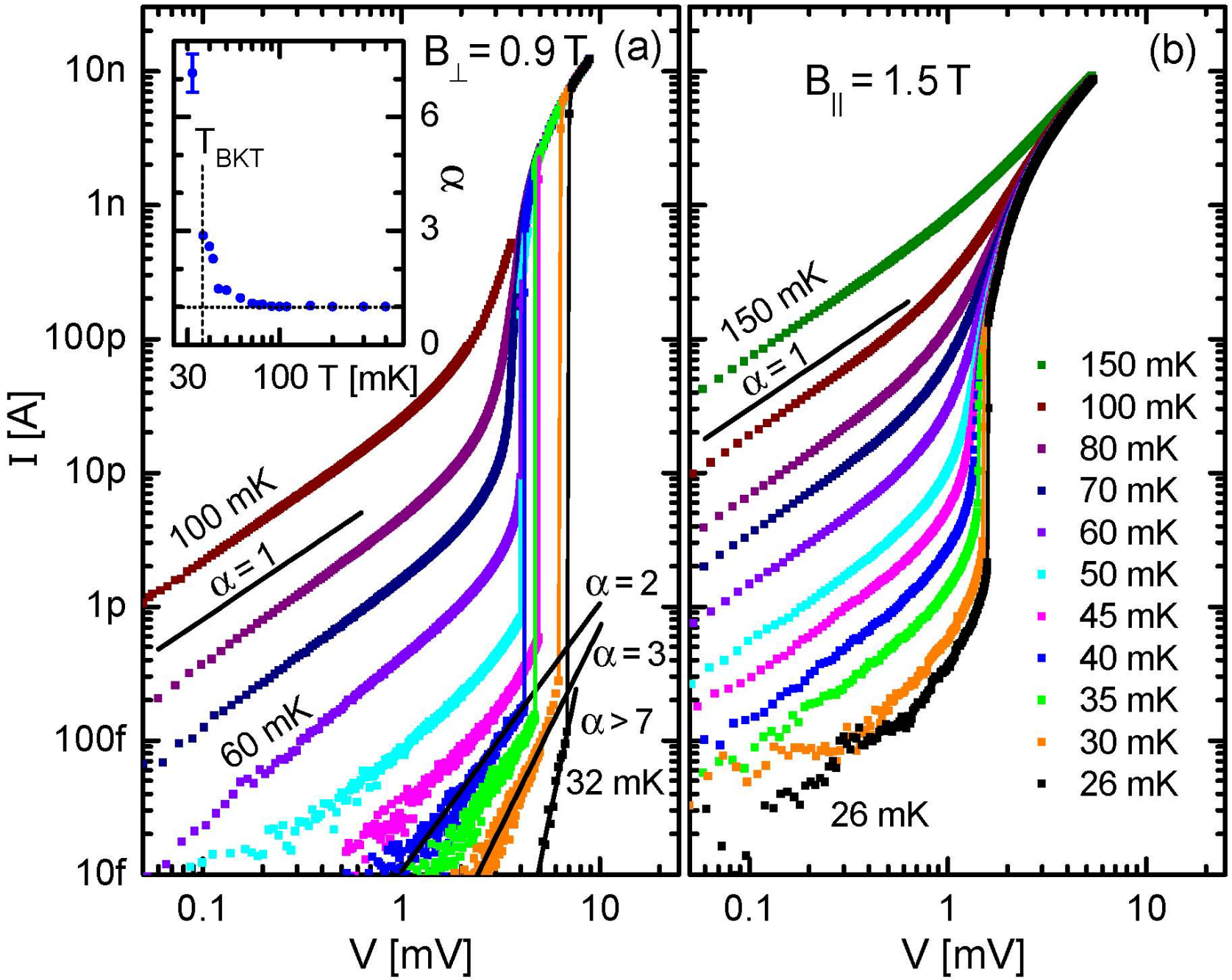
both the positive magnetoresistance and initial rise of T_0 and V_T disappear

$T_0(B)$: horizontal slope; $V_T(B)$: finite slope

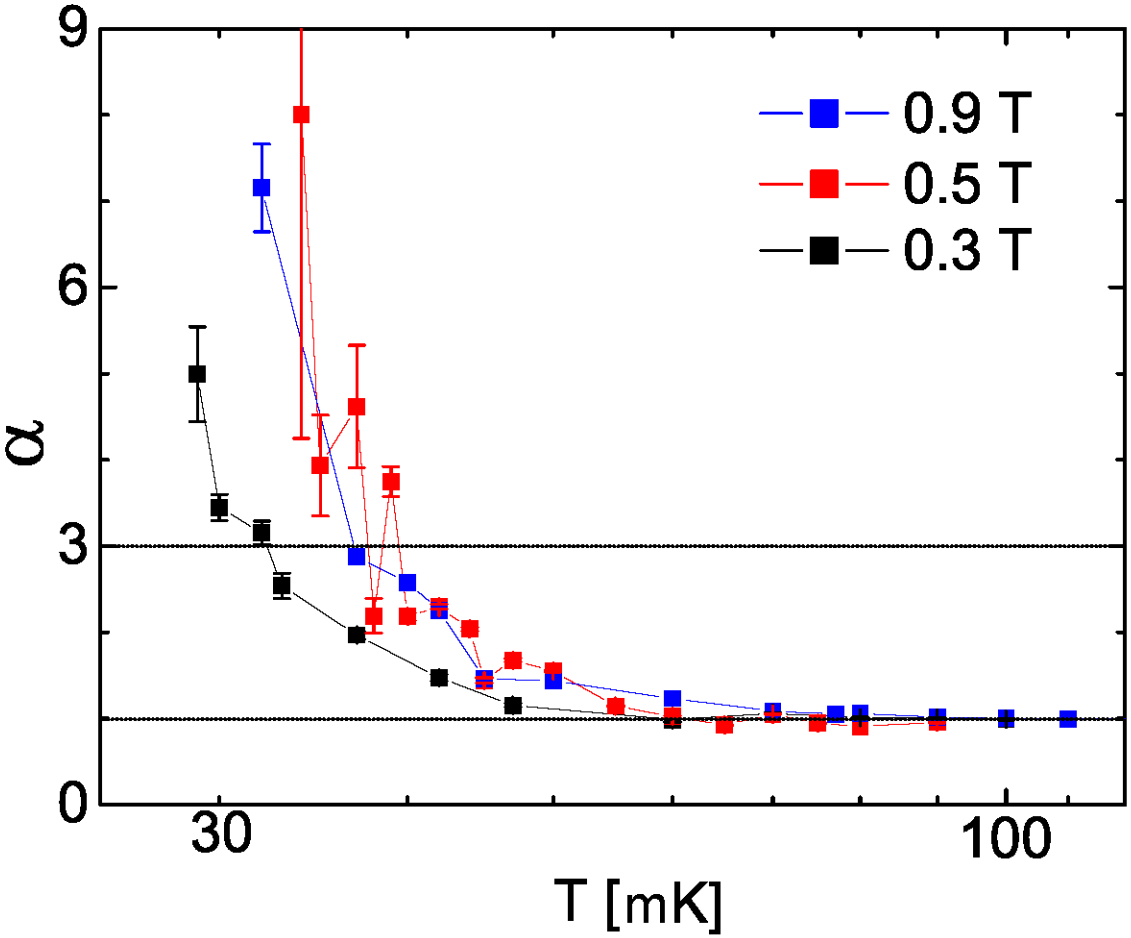
Again jumps in IV, but also current below jump measurable



IV's display power-law behavior at the lowest temperatures !



Charge-Berezhinski-Kosterlitz-Thouless-like transition smeared by disorder



IV characteristics become non-linear below 60 mK

power law

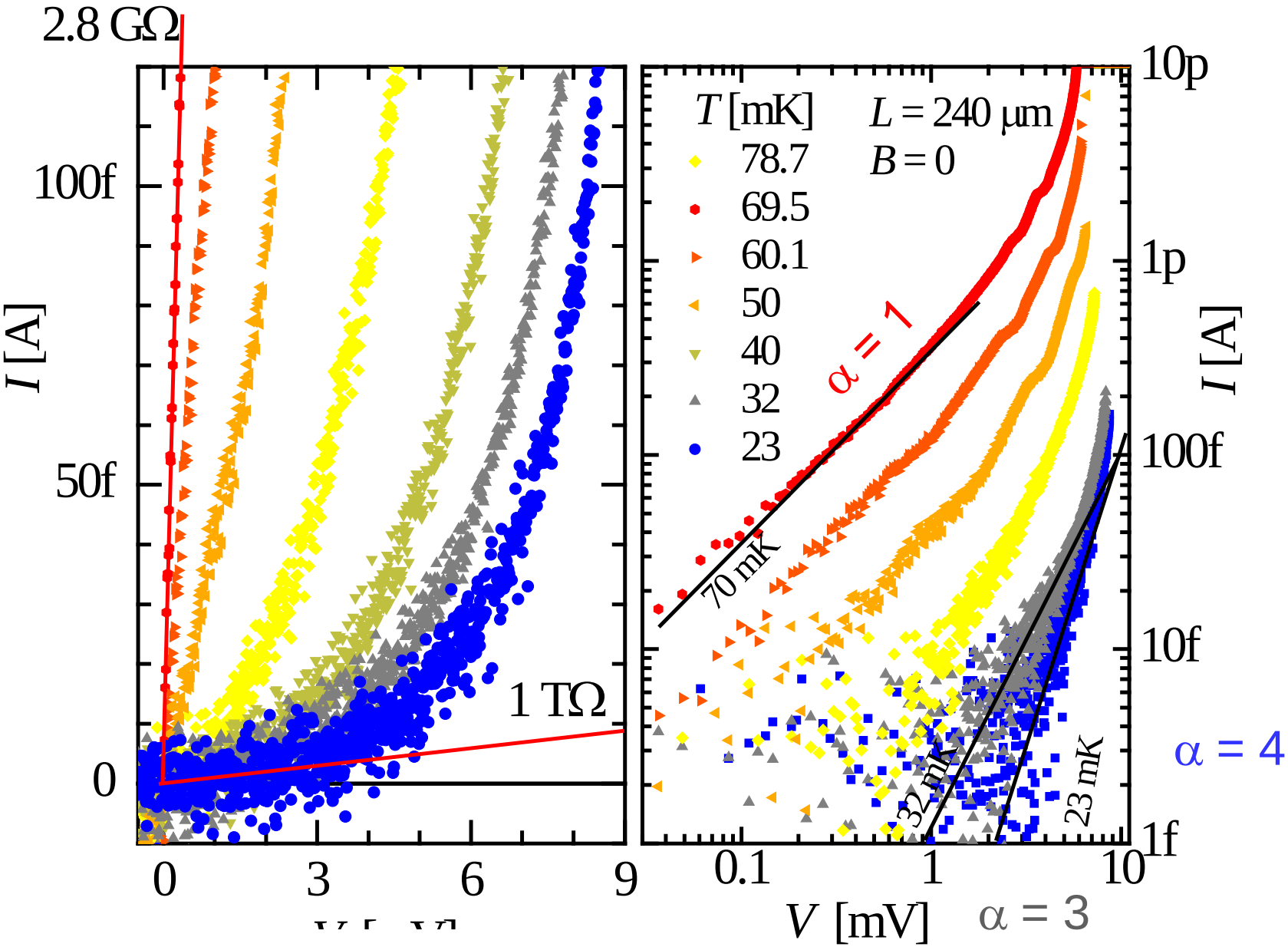
$$I(V) \propto V^{\alpha(T)}$$

with strongly T- dependent exponent α(T)

BKT transition usually assigned to $\alpha = 3$

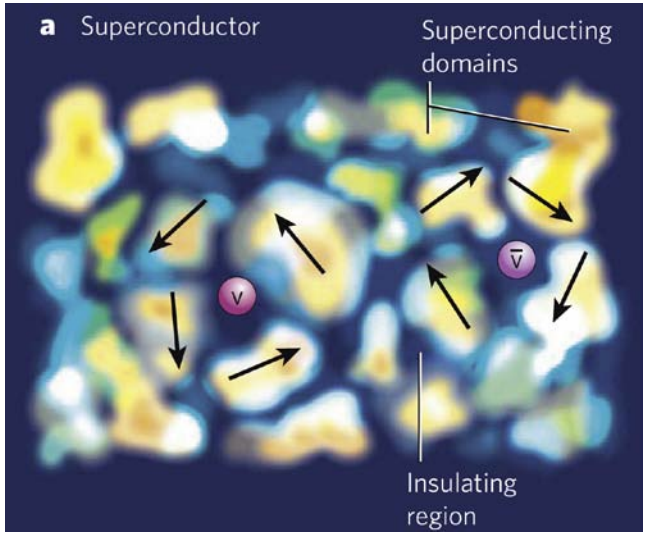
D. Kalok, C.S. et al., arXiv:1004.5153

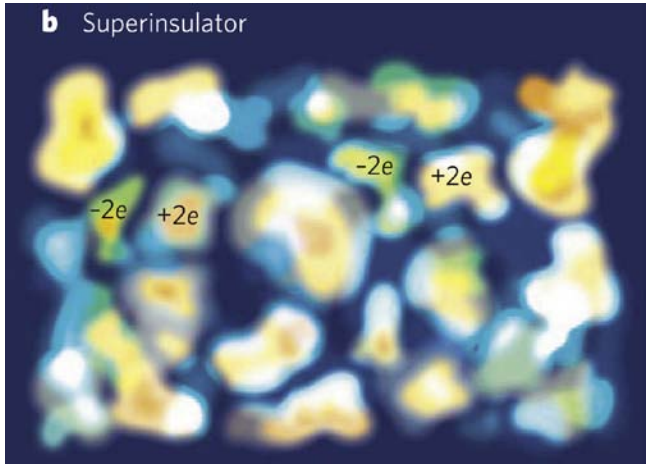
Another sample – insulating at B = 0

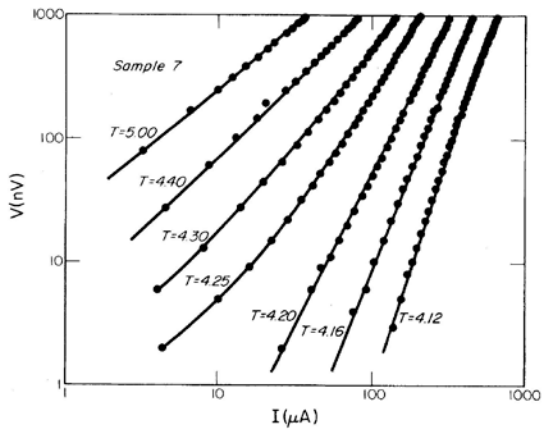


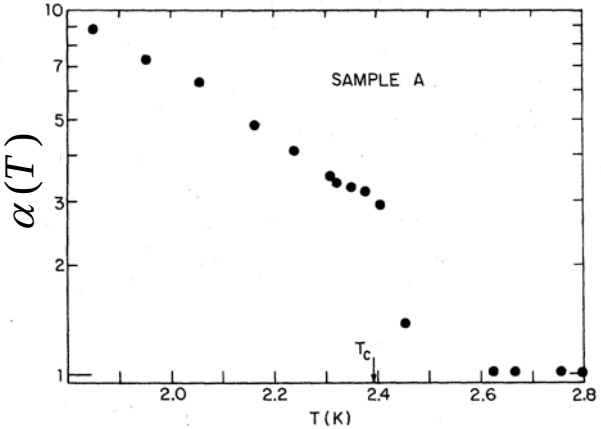
Very close analogy to vortex BKT-transition:

proximity-coupled
JJ-arrays (Pb/Sn)









Resnick et al., PRL '81, Abraham at al., PRB '82

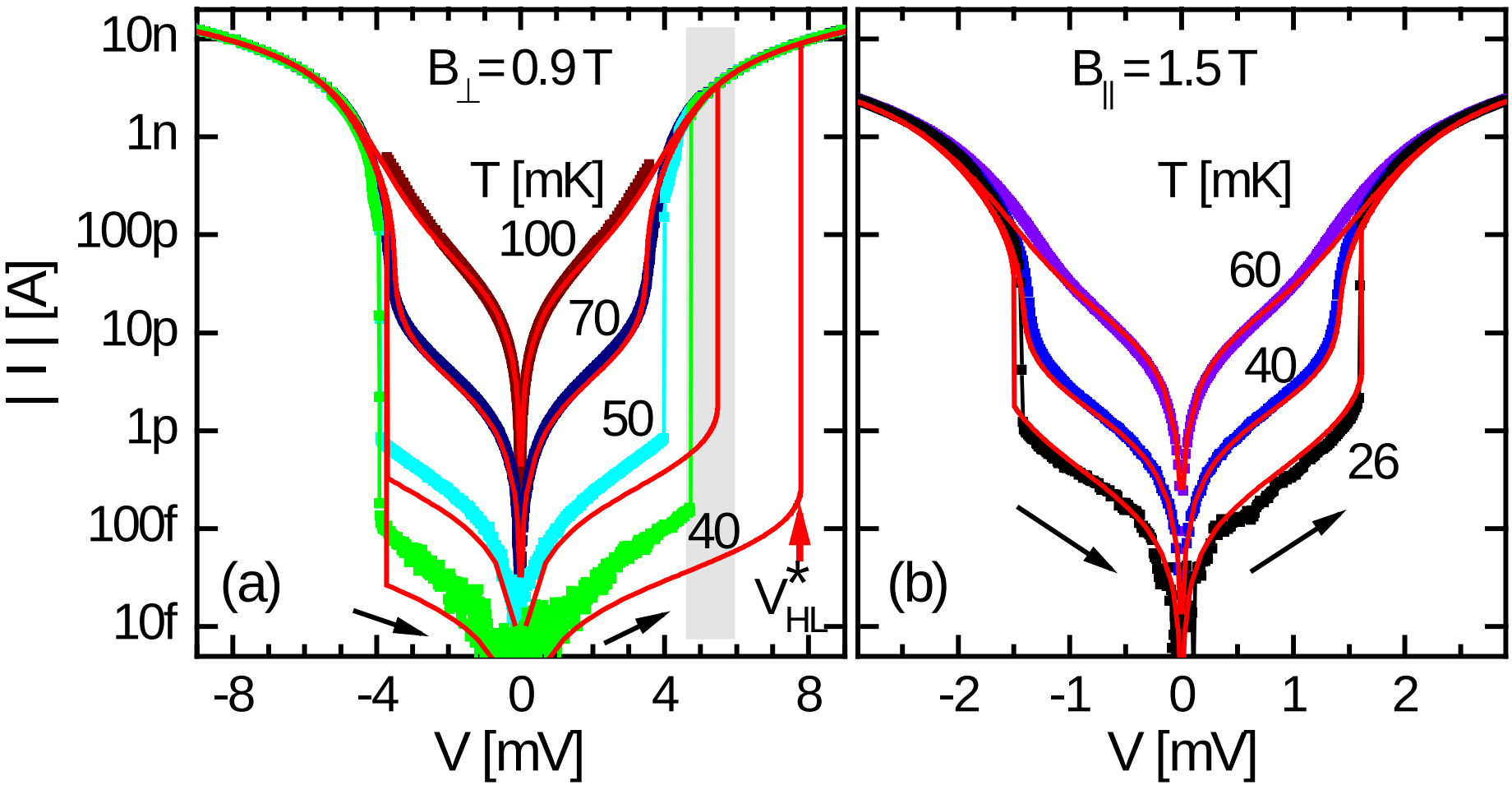
charge-BKT - transition:

logarithmic interaction between charge/anti-charge pairs required

very high effective dielectric constant needed to force electric field lines in plane!

R. Fazio Nature **552** (2008) news & views

magnetic field: perpendicular parallel



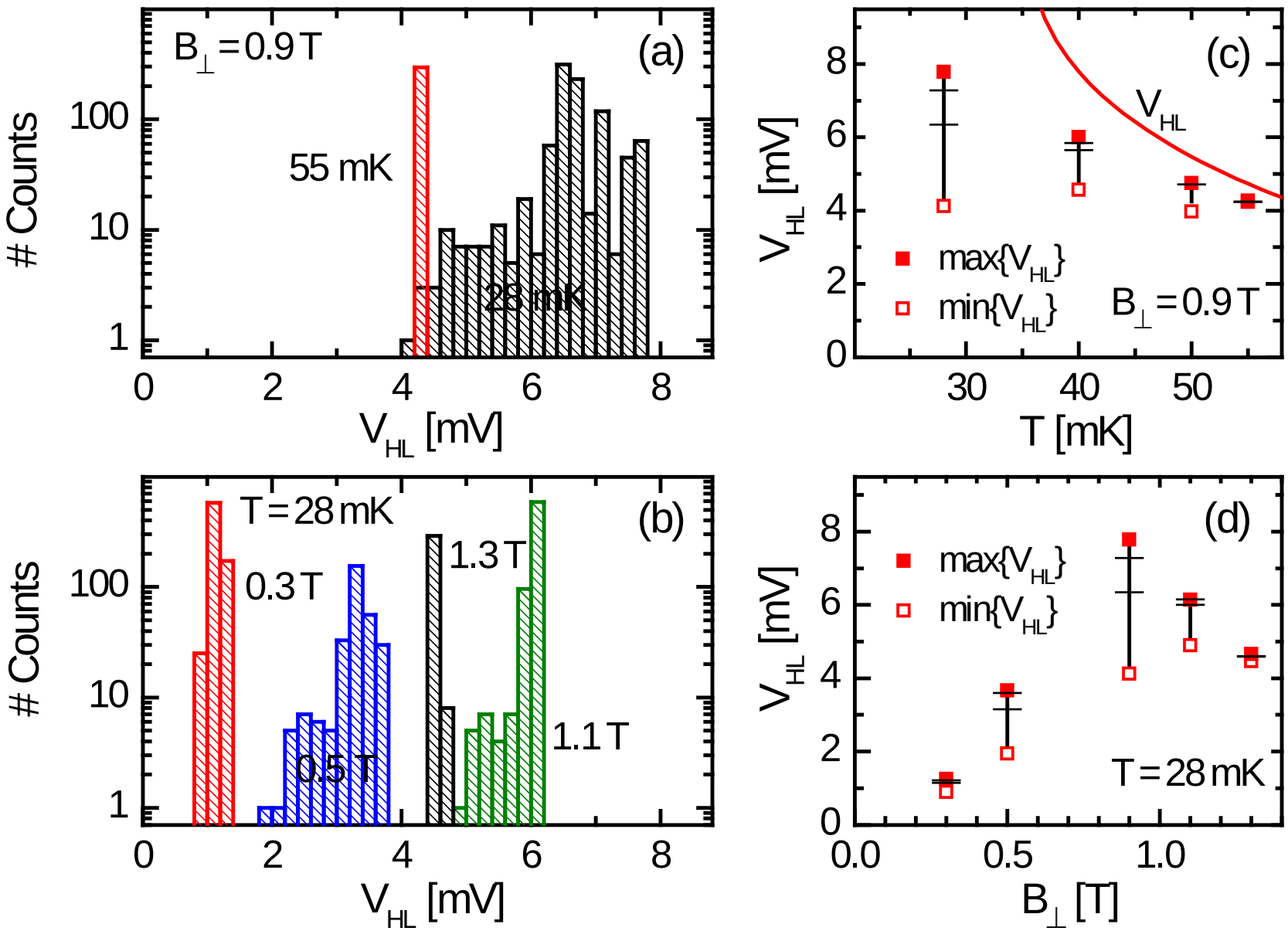
dots: experiment

red: model of electron heating by electron-phonon decoupling

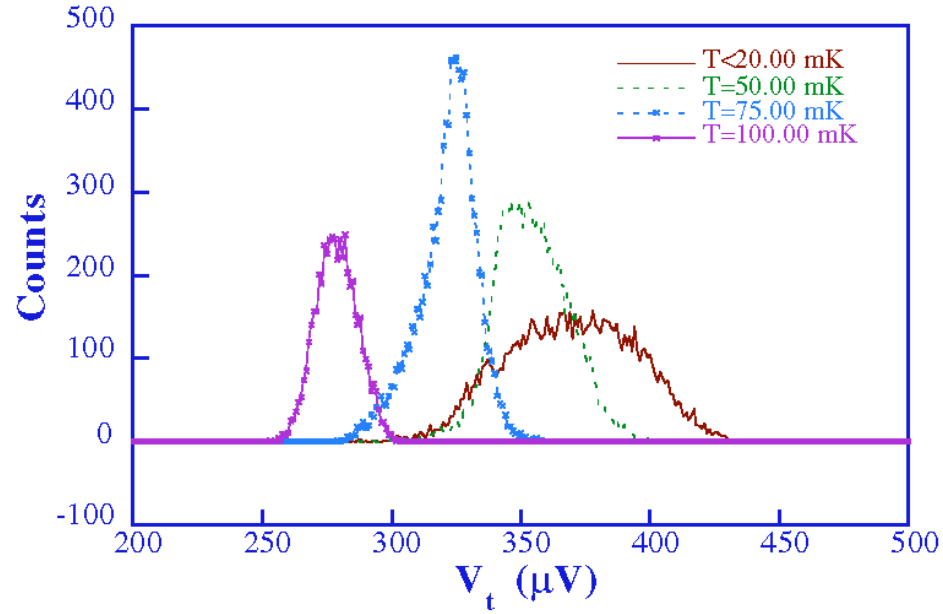
at T < 40 mK the heating scenario systematically overestimates I and V_{Th}

see also Ovadia et al., PRL 102, 176802 (2009)

Histograms display broad range of switching voltage

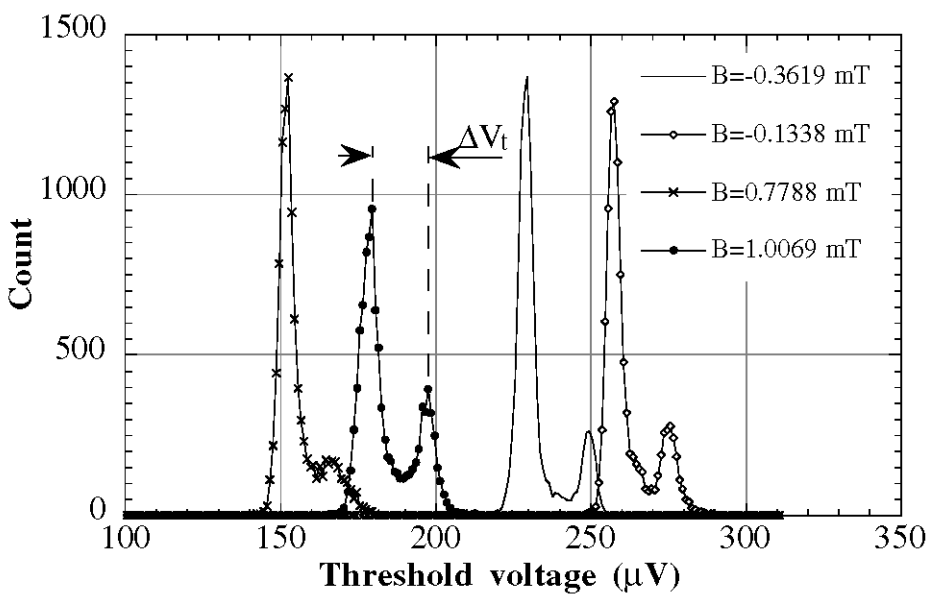


Compare data on an artificial insulating JJ-array:



168 x 168 junctions
R = 27 kΩ per junction

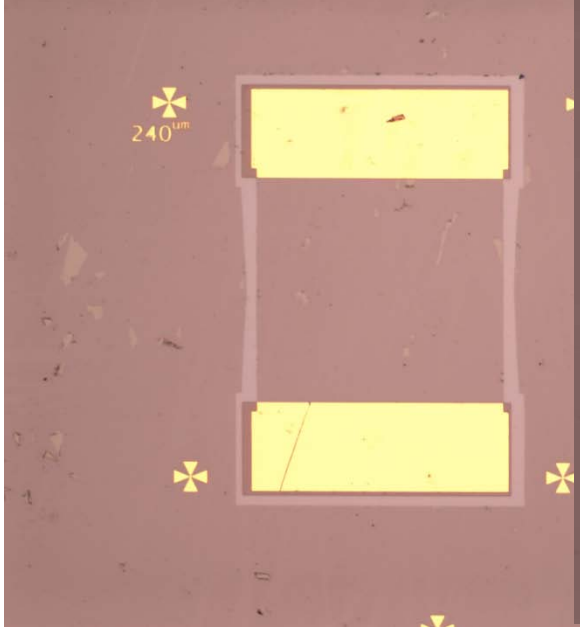
voltage switching with width 30% of $V_{T,max}$

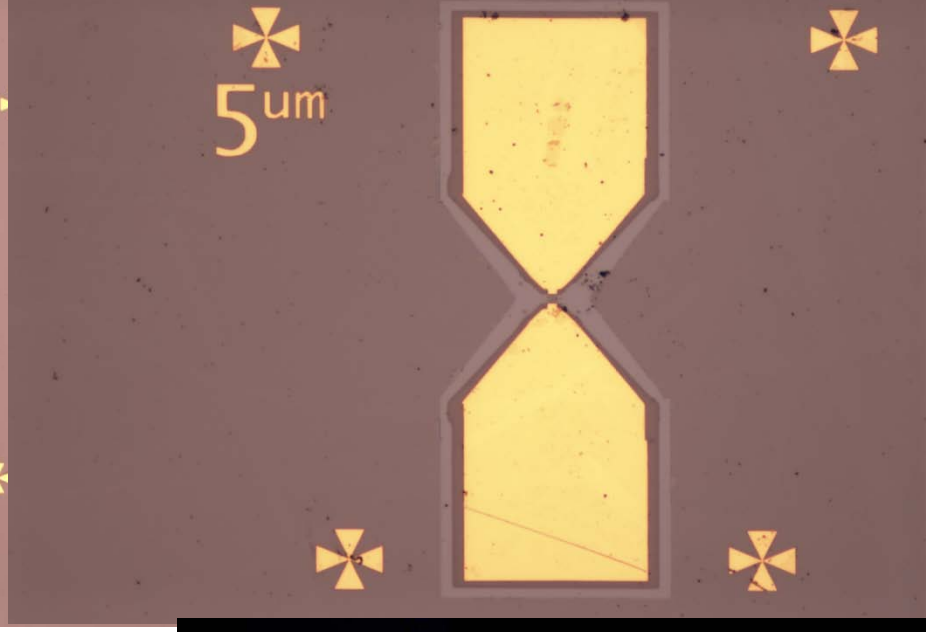


non-monotonic variation with magnetic field

sometimes two peaks observed

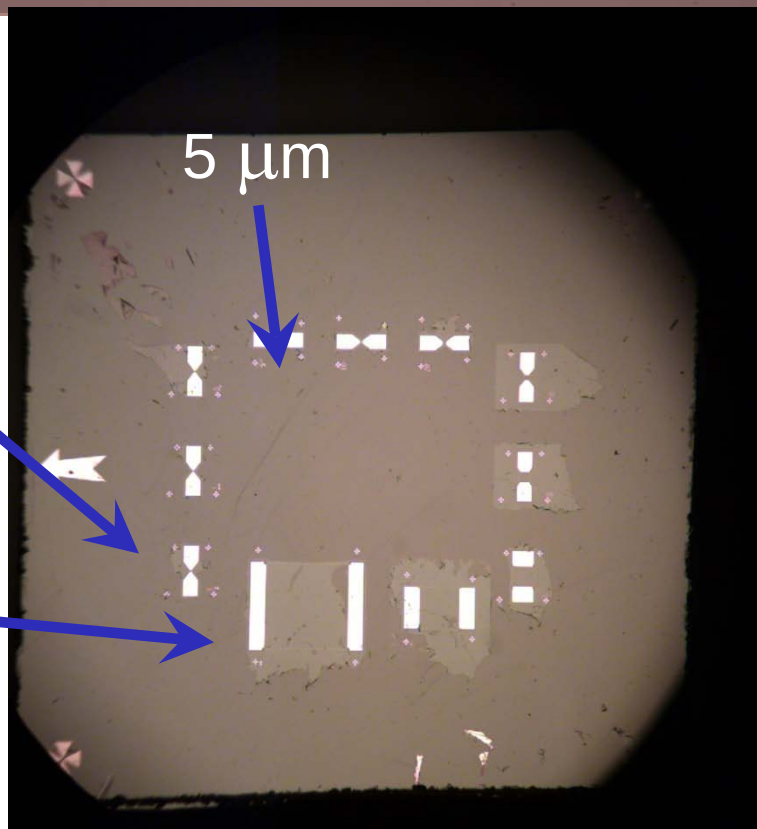
T. Bergsten et al. Chalmers '96



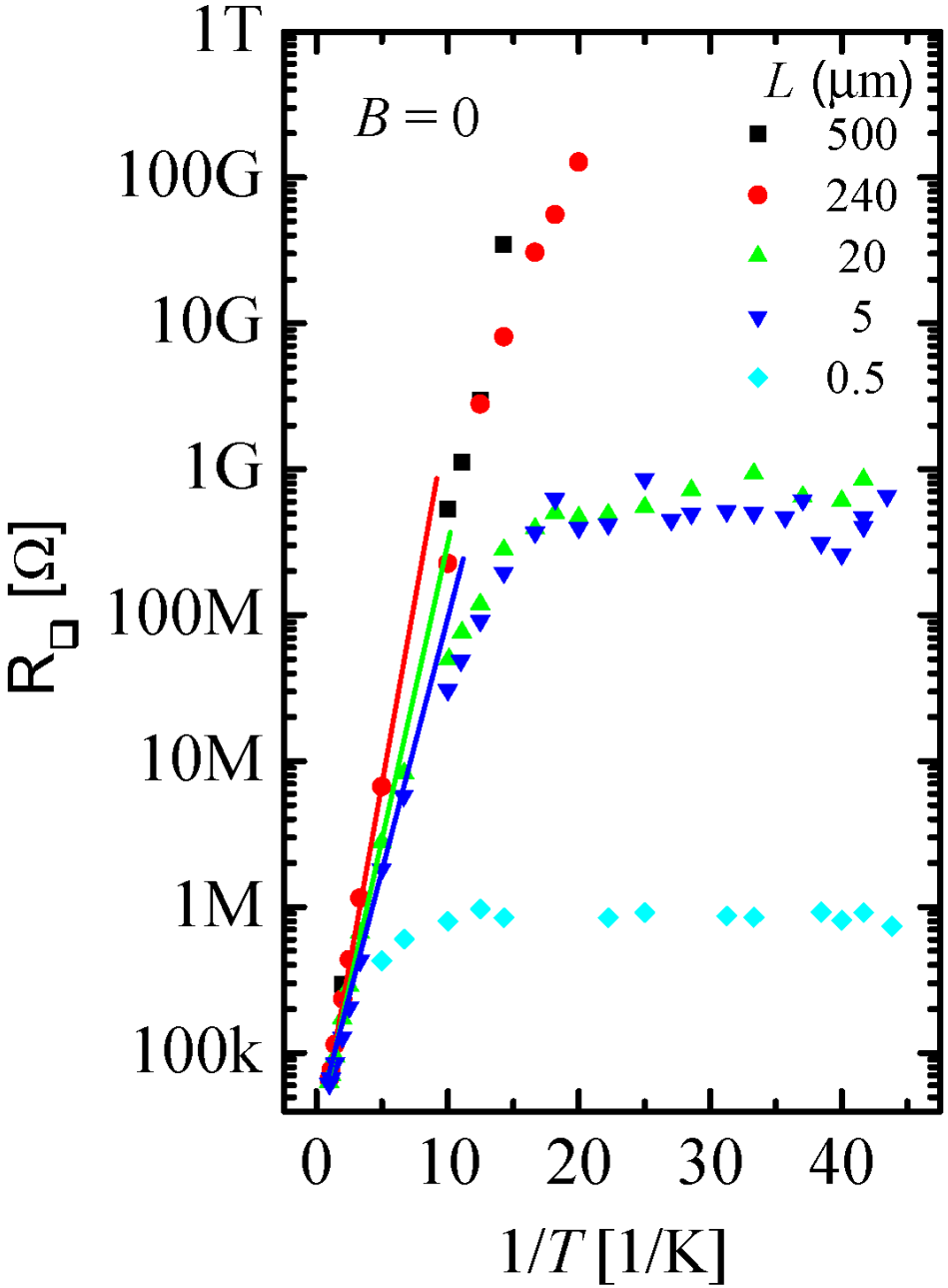


Study dependence of $R_{\square}$ on sample size:

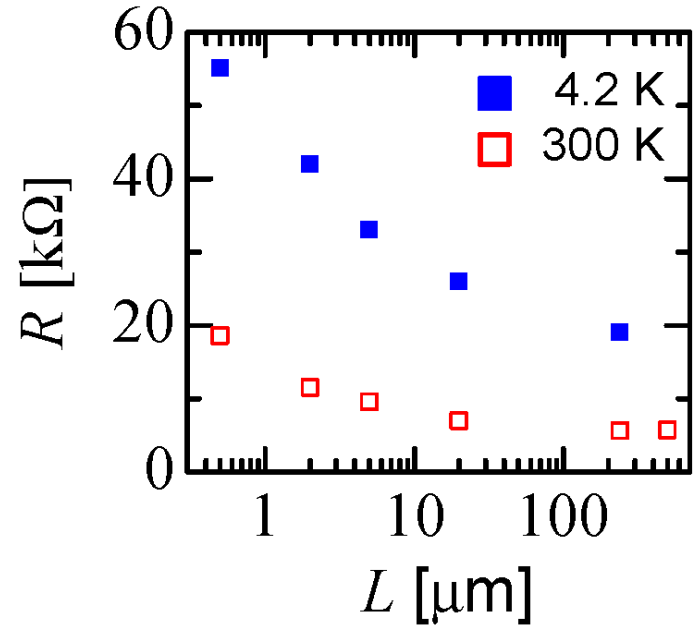
prepare squares between 0.5 and 500 μm side length

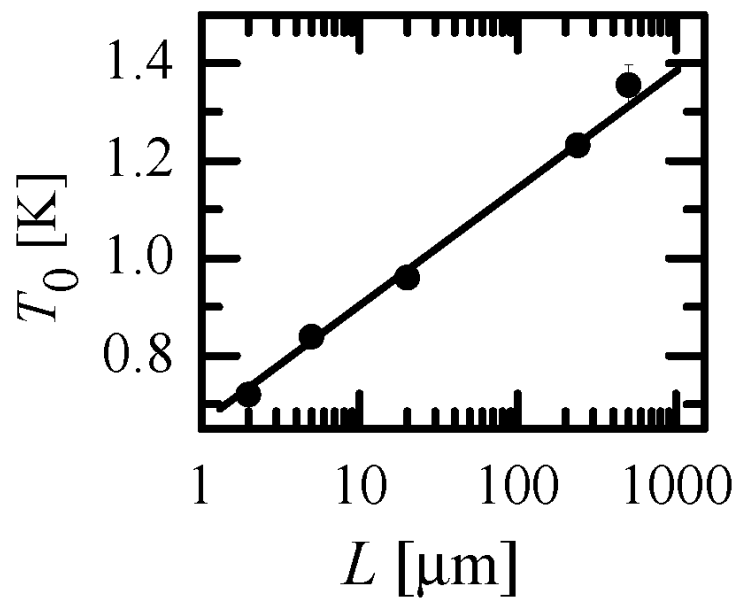


R(T) becomes *size dependent*:

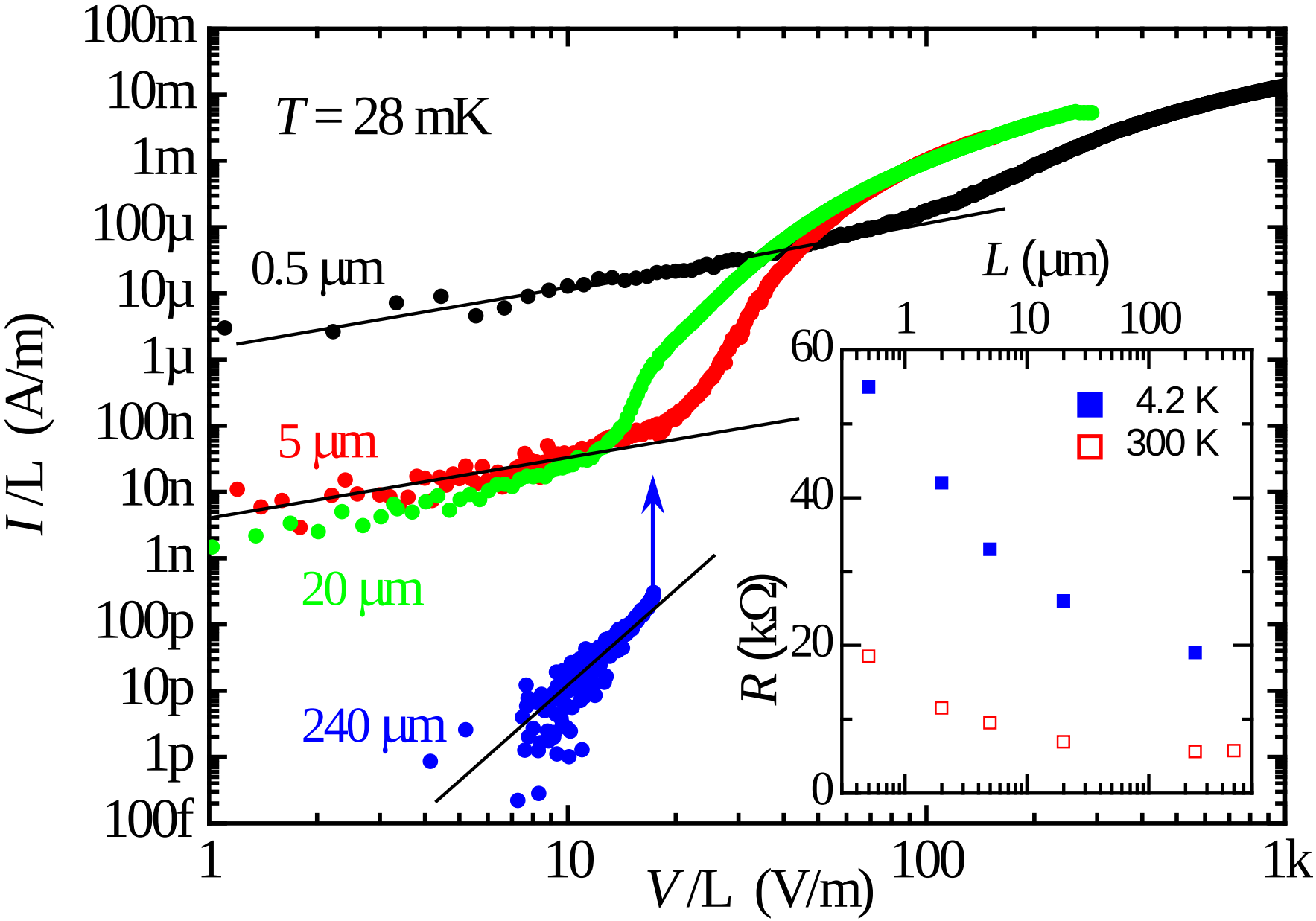


sample B: insulating at B = 0





size dependent IVs display cross-over from linear to nonlinear behavior at low T



characteristics of a granular model

- thermally activated conductance $T > T_{SI} = E_c / k_B$

$$R \propto \exp[\Delta_c/(2k_B T)]$$

- collective Coulomb energy:

$$\Delta_c = \begin{cases} E_c N/2, & 1D \\ E_c \log N, & 2D \end{cases}$$

N : number of islands

$$N = \min\left\{\frac{L}{d}, \frac{\lambda}{d}\right\}$$

where λ is screening length

- Threshold voltage depinning, $eV_T \sim E_c L/d$

Review: *Minnhagen, RMP 59, 1001 (1987)*

Fistul, Vinokur, Baturina, PRL **100**, 086805 (2008)

Is there a granular structure of the SC OP ?

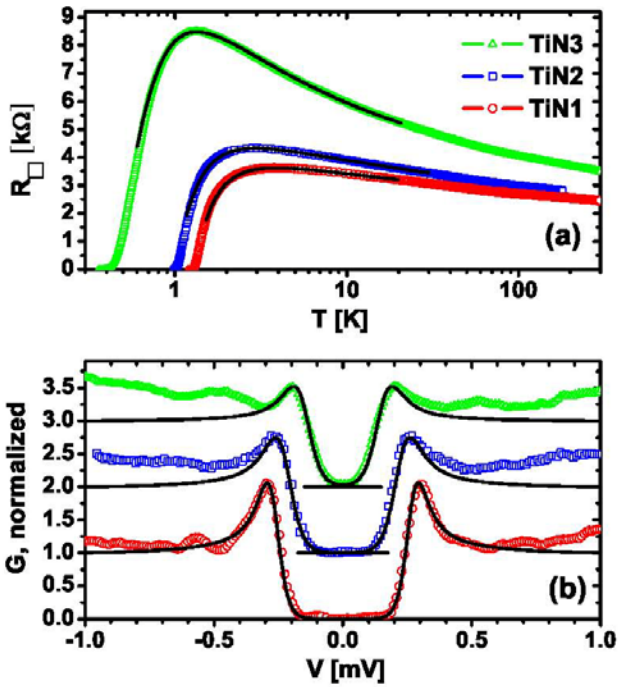


FIG. 1: (a) Sheet resistance $R_\square$ versus temperature for three samples. The solid lines are fits according to localization-interaction and superconducting fluctuations corrections. The legend of panel (a) describes the two panels. (b) Normalized differential tunneling conductance measured at $T = 50\,\mathrm{mK}$ (dots). Spectra are shifted for clarity. The BCS fits (solid lines) were calculated with the following parameters: TiN1 - $\Delta = 260\ \mu\mathrm{eV}$ and an effective temperature $T_{eff} = 0.25\,\mathrm{K}$; TiN2 - $\Delta = 225\ \mu\mathrm{eV}$, $T_{eff} = 0.32\,\mathrm{K}$; TiN3 - $\Delta = 154\ \mu\mathrm{eV}$, $T_{eff} = 0.35\,\mathrm{K}$.

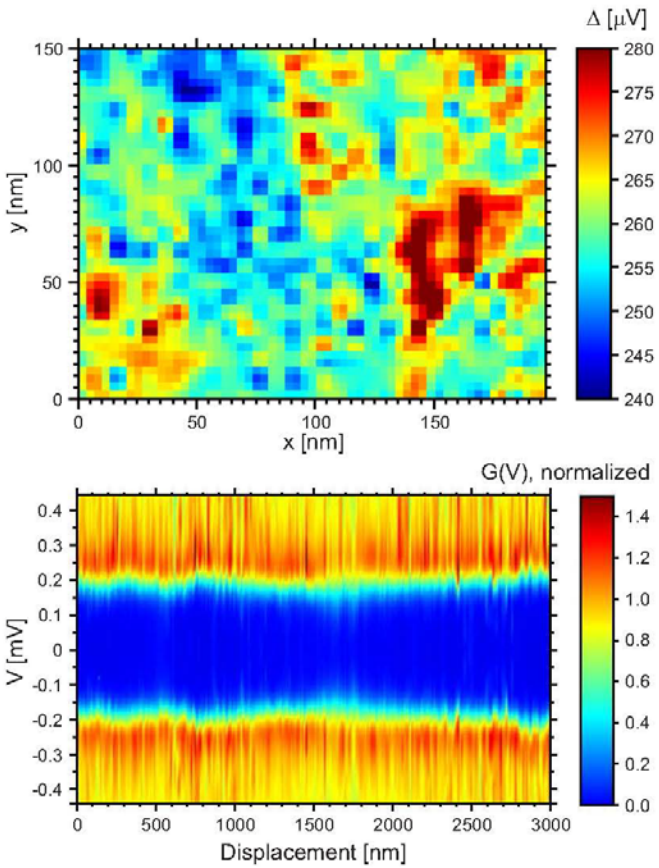


FIG. 2: Top: The colour map of spatial fluctuations of Δ on TiN1. Inhomogeneities of the superconducting properties show up on a scale of a few tens of nanometers. Bottom: Spectra measured along a straight line on TiN2. The BCS-like LDOS fluctuates symmetrically around the Fermi level.

Theory predictions:

Ghosal, Randeria, Trivedi,

PRL ('98), PRB ('01)

Dubi, Meir, Avishai

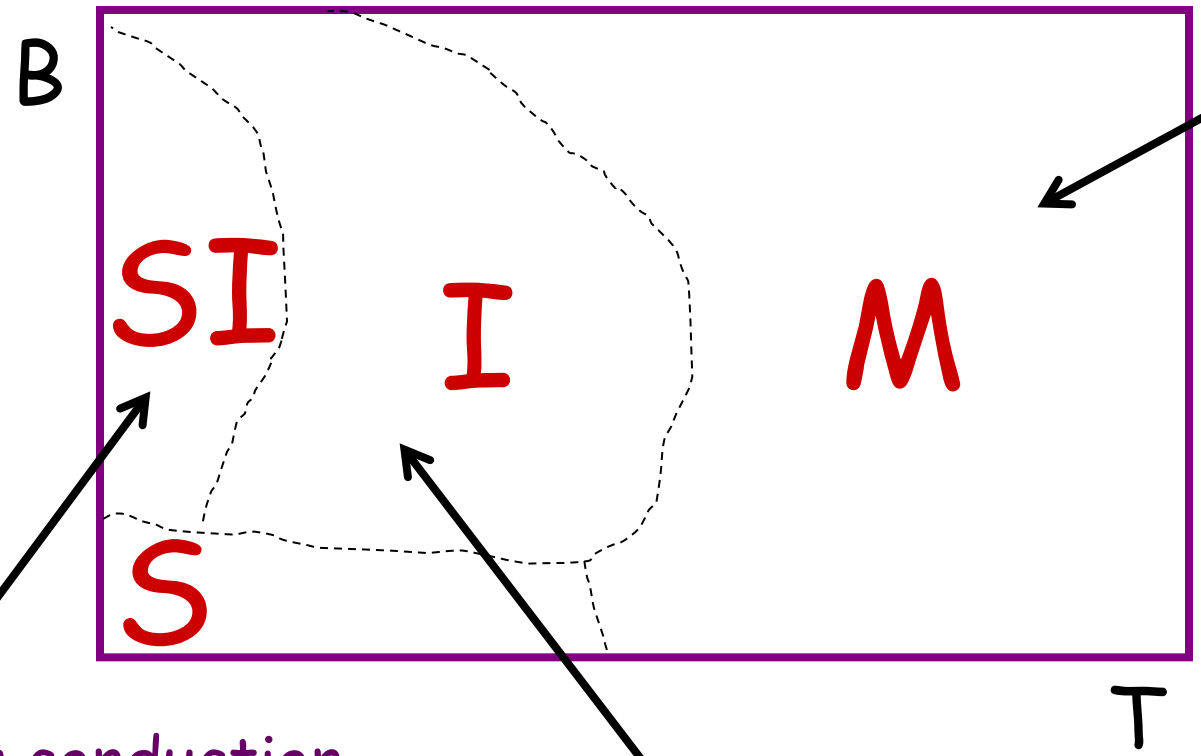
Nature ('07)

Experiment on TiN: PRL 101, 157006 (2008)

B. Sacepe, C. Chapelier, T. I. Baturina, V.M. Vinokur, M.R. Baklanov, and M. Sanquer

schematic phase diagram (fluctation broadened)

(sample superconducting at B=0)



metal with quantum corrections

'Super'-insulator

nonlinear conduction,
extreme T-dependence
dielectric breakdown

'Cooper-pair' insulator

linear conduction,
thermally activated conductance
heating instability of I(V)

Conclusions

- insulating state with thermally activated (Arrhenius) conductance

- evolution from a insulator with activated conductance to a much lower conductance ('superinsulating') state at very low T

- power-law behavior of IV's at the lowest temperatures
- statistical distribution of switching voltages as in dielectric breakdown

→ *charge*-Berezhinskii-Kosterlitz-Thouless-like crossover

- logarithmic size dependence of activation energy on sample size

- R(T) and I(V) show even stronger size dependence at the lowest T

Open Questions

- existence/origin of island formation ?

- dielectric properties:

  logarithmic interaction between charges ?

  anomalies in the dielectric properties ?

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