Size-dependent Conduction near the Super-conductor/Insulator Transition in thin TiN-Films

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Outline:

- global conductance vs. conductivity
  as a probe of long-range interactions

- collective charging energy in arrays of tunnel junctions
  resulting from 2-dim Coulomb interaction

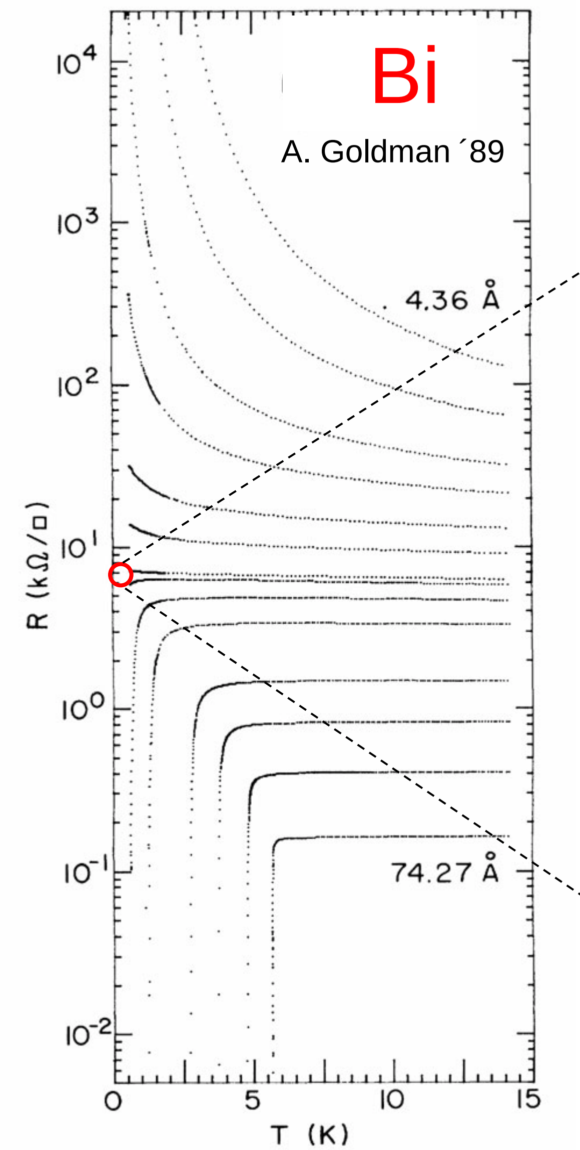
- size dependence of R(T), R(B) and I(V)

- comparison to expectations from
  Berezinskii-Kosterlitz-Thouless transition

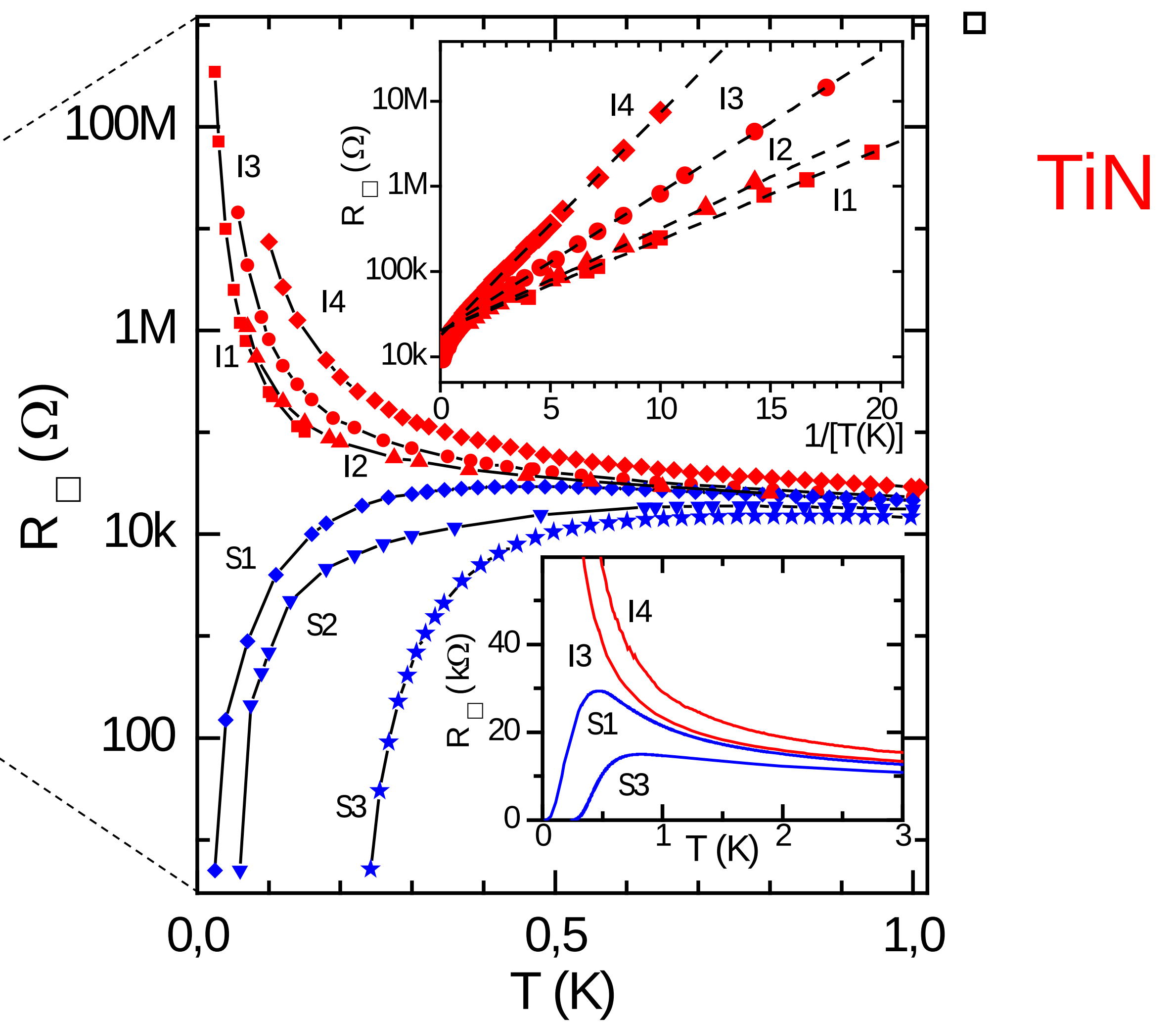
In this talk, 'insulators' are characterized by
an exponential R(T) dependence.

direct superconductor/insulator transition

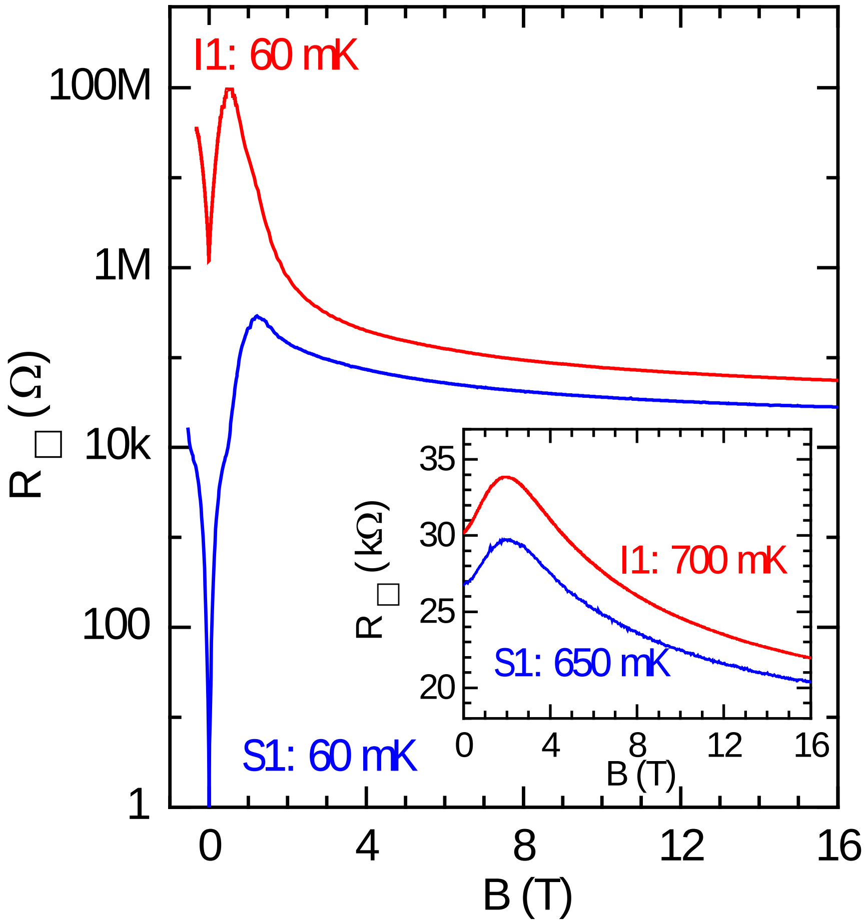
with Arrhenius-like conductance on the insulating side



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



initially *positive* magnetoresistance in both: metallic and *insulating* state



Qualitatively similar behavoir for insulating and superconducting films

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

Superconductivity important for insulating behavior ?!

quasi-metallic phase at high B

low B:
‚Cooper-pair' - insulator !

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

How to model the Cooper-pair insulator:

- thermally activated conductance

  expt.: Palaanen & Hebard, Ovadyahu, and many others
  theory: Fistul, Baturina, Vinokur, PRL 100 (2008)
  Feigel'man, Ioffe, Kravtsov, Cuevas,
  Ann. Physics 325, 1368 (2010);
  Feigelman, Ioffe, Mezard, PRB 82, 184534 (2010)

- strongly non-monotonic magnetoresistance

  expt.:Gantmakher et al., Shahar et al., Baturina et al. and others
  Theory: Dubi, Meir, Avishai, Nature 449 876 (2007)

- non-linearities in IV-characteristics

  expt. Baturina et al.; Ovadia et al.,
  theory: Vinokur et al., Altshuler et al.

emergent inhomogeneity in superconducting condensate

→ exploit knowledge about Josephson junction array (JJAs)

Josephson arrays in the charging regime

"Unbinding of Charge-Anti-Charge Pairs in Two-Dimensional arrays of small Tunnel Junctions"

J. Mooij, B.J. van Wees, L.J. Geerlings, M. Peters, R. Fazio and G. Schön,
Phys. Rev. Lett. **65**, 645 (1990)

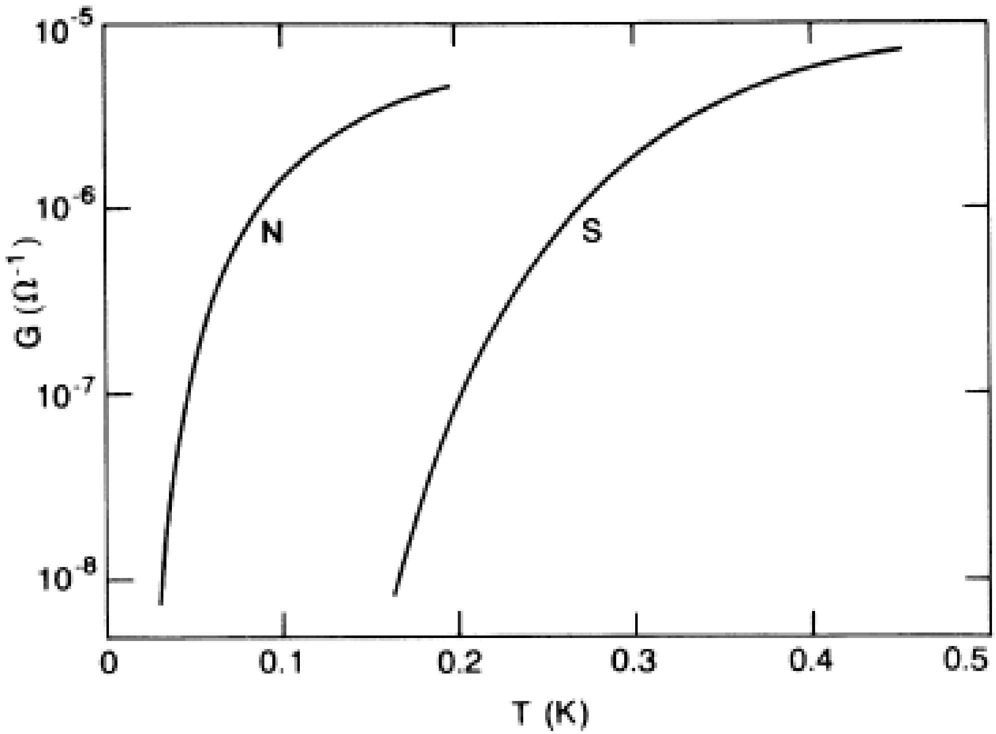


FIG. 3. Measured conductance of an array of (100 nm)^2 aluminum tunnel junctions, 190 cells long and 60 cells wide. *N* is in the normal state (magnetic field of 3T applied); *S* in the superconducting state.

consider JJ-arrays in the limit $E_J < E_C$

superconducting array turns insulating at much higher temperatures !

H. v.d.Zant, R. Fazio
Phys. Repts. **355**, 235 (2001)

question: what energy scales are relevant in (disordered) Josephson junction arrays?

- thermally activated conductance at $T > E_C/k_B$

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

- collective Coulomb energy:

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

N : number of islands

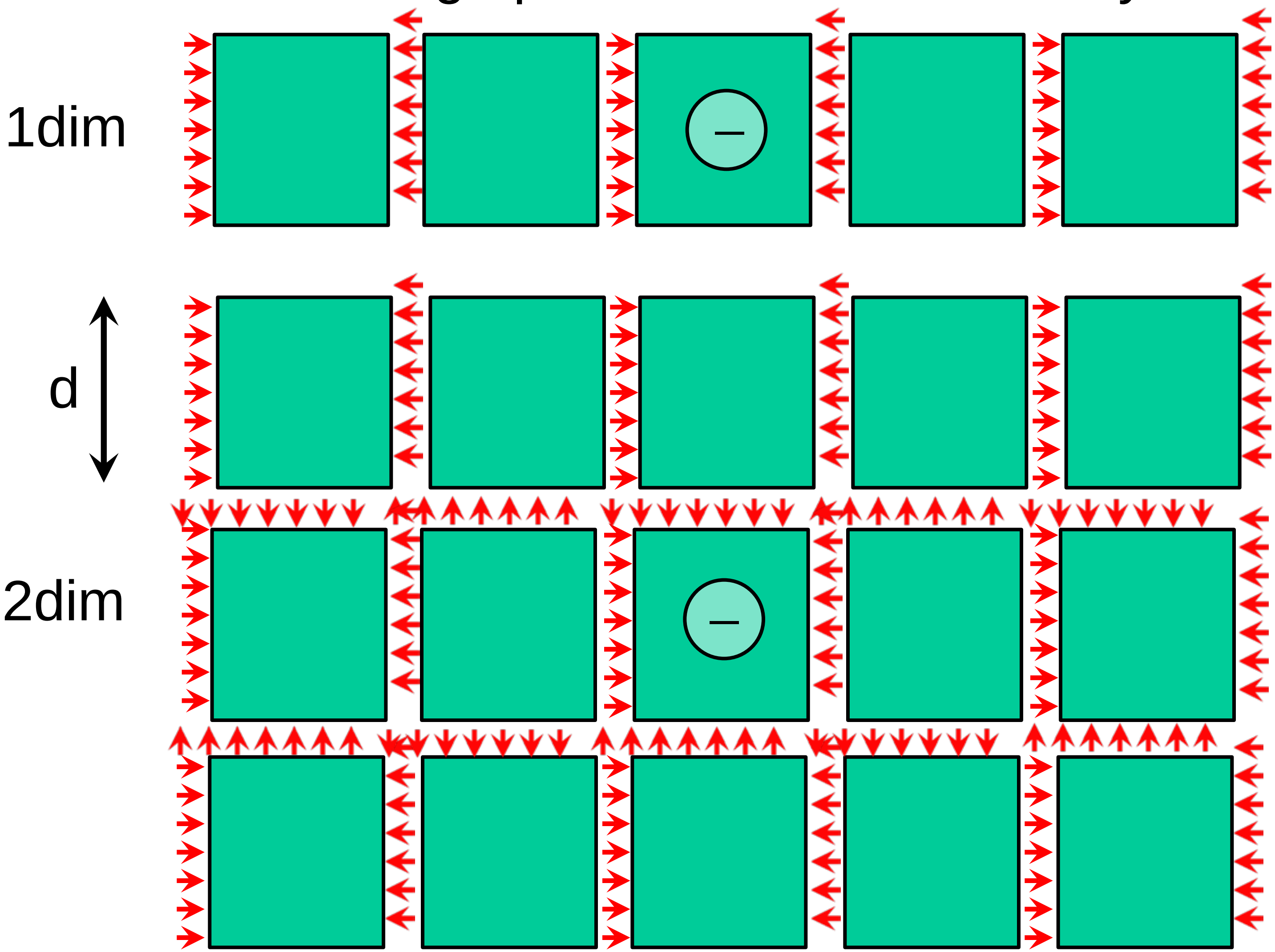
$$N = \min\left\{\frac{L}{d}, \frac{\lambda}{d}\right\}$$

λ is the electrostatic screening length

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

origin of collective Coulomb energy:

1 excess charge polarizes the whole array, if $\lambda > L$

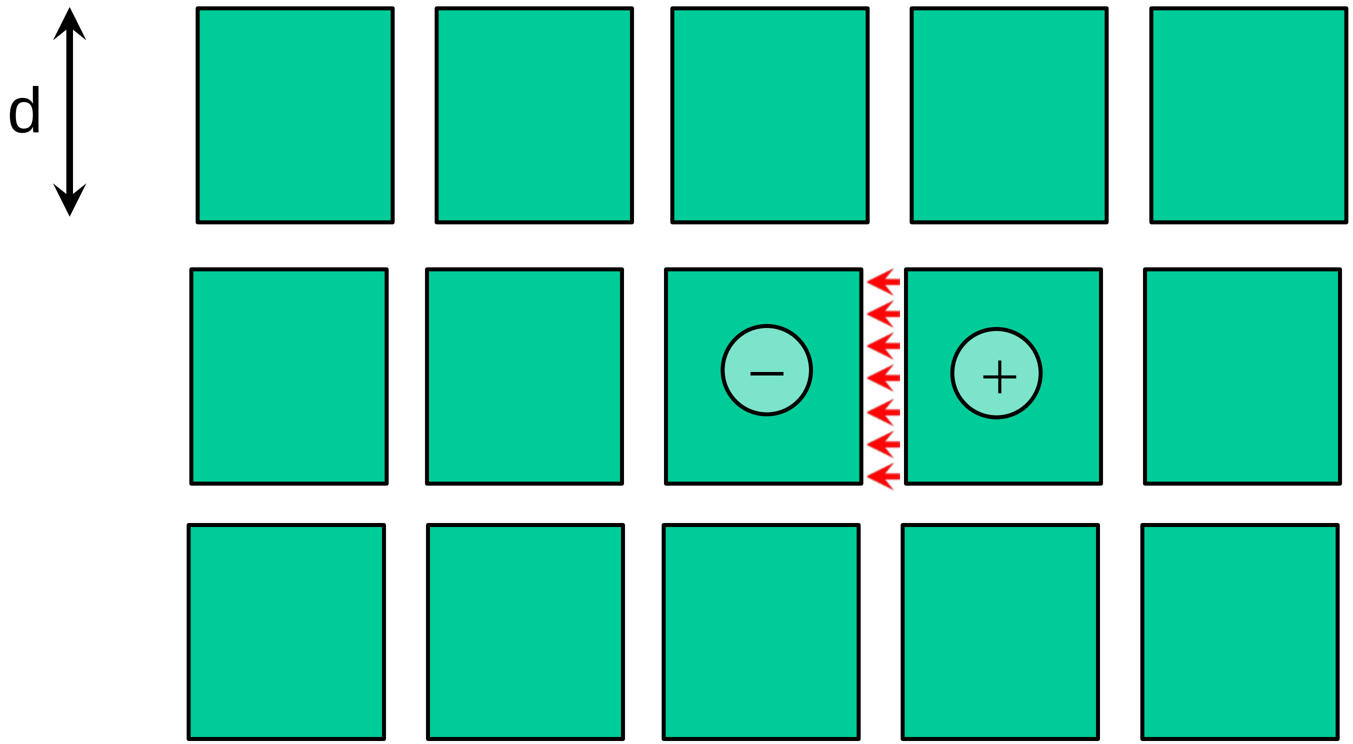


C: capacitance between islands

C_0: capacitance to ground

polarization cloud cut off by screening length $\lambda = d(C/C_0)^{1/2}$

charge/anti-charge pairs bound on adjacent islands cost much less energy:



C: capacitance between islands

C_0: capacitance to ground

adjacent charges cost charging energy E_C

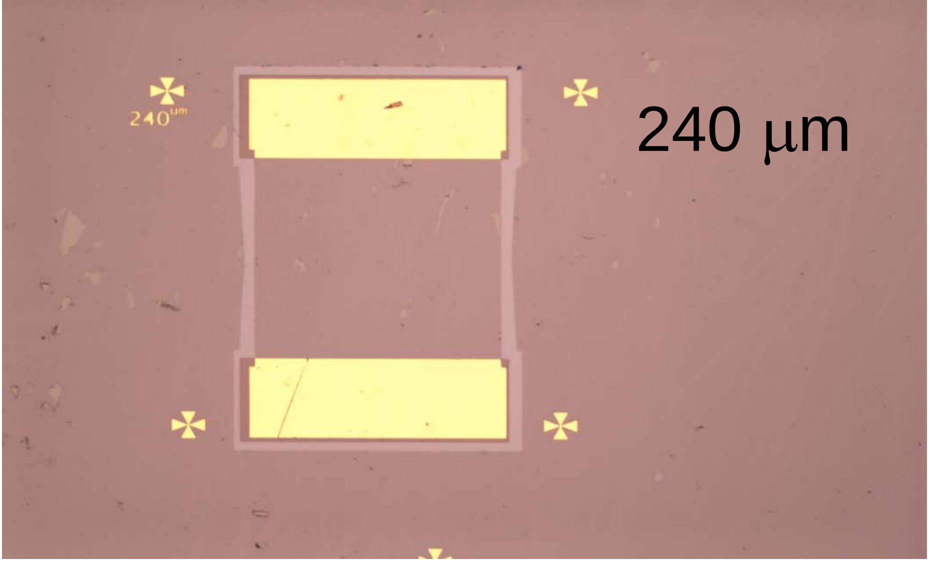
seperating charges costs *collective charging energy* $\Delta_C = E_C/2 \ln(L/d)$

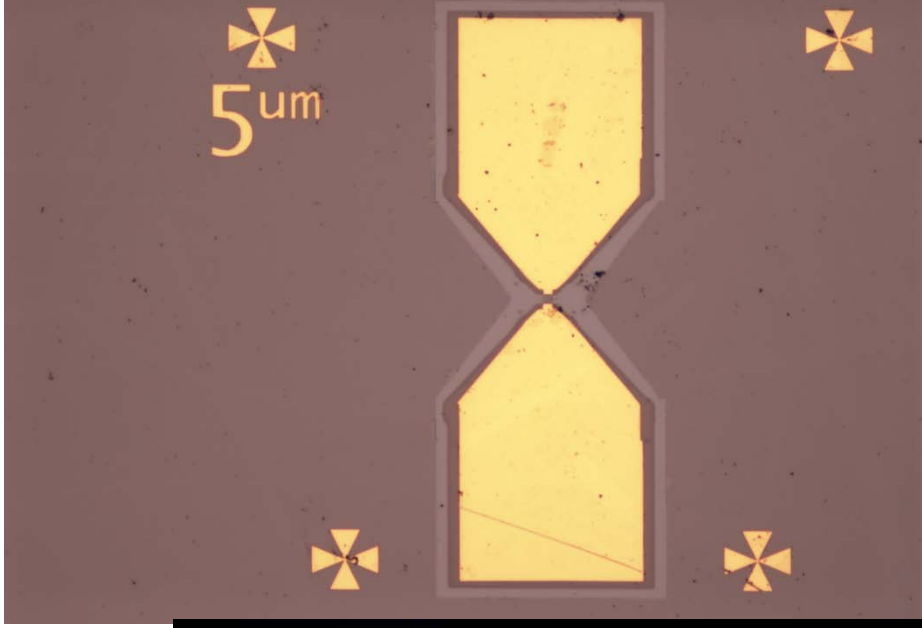
→ logarithmic interaction between charges in array

How to check the relevance of JJ-array models and/or Coulomb charging effects experimentally?

Look for *size dependence* of transport properties:

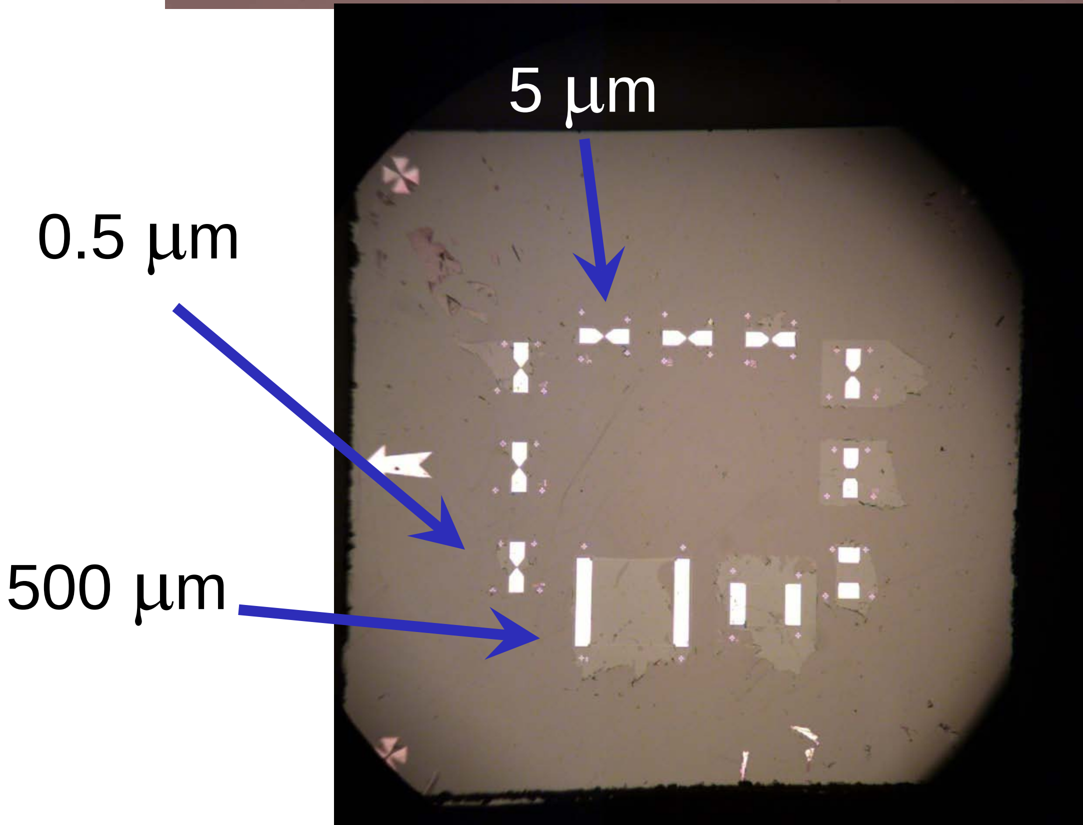
- size dependent activation energy ?
- size dependent magnetoresistance ?
- I(V) – characteristics ?



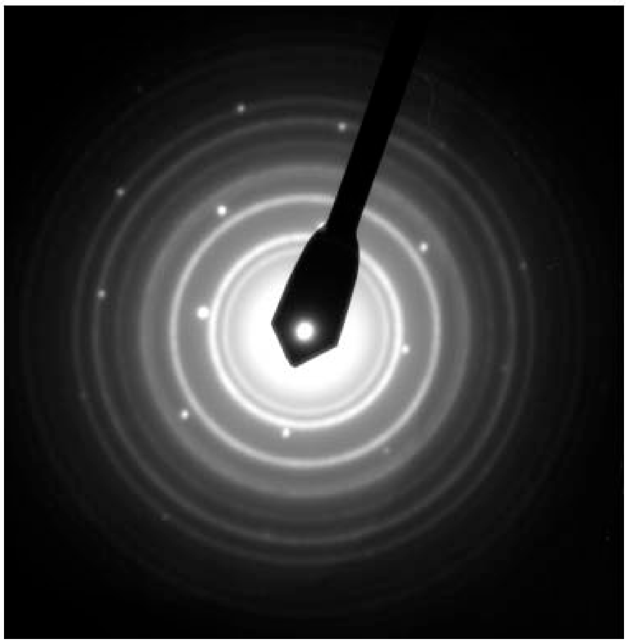


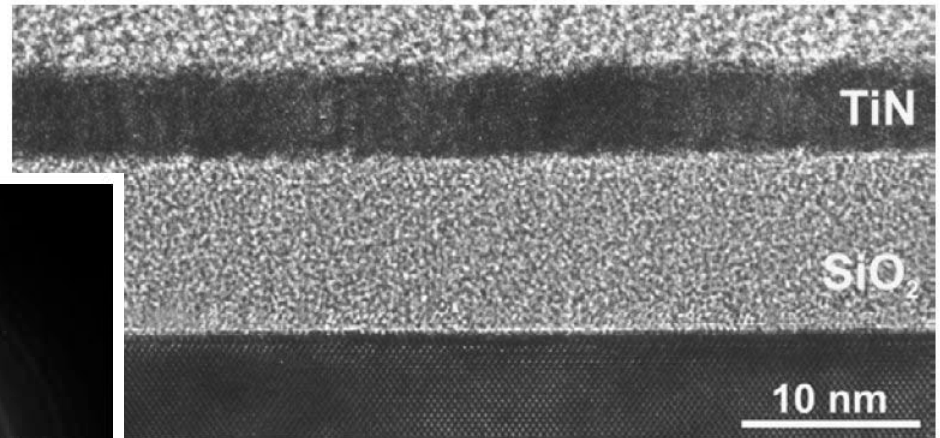
dependence of $R_{\square}$ on sample size:

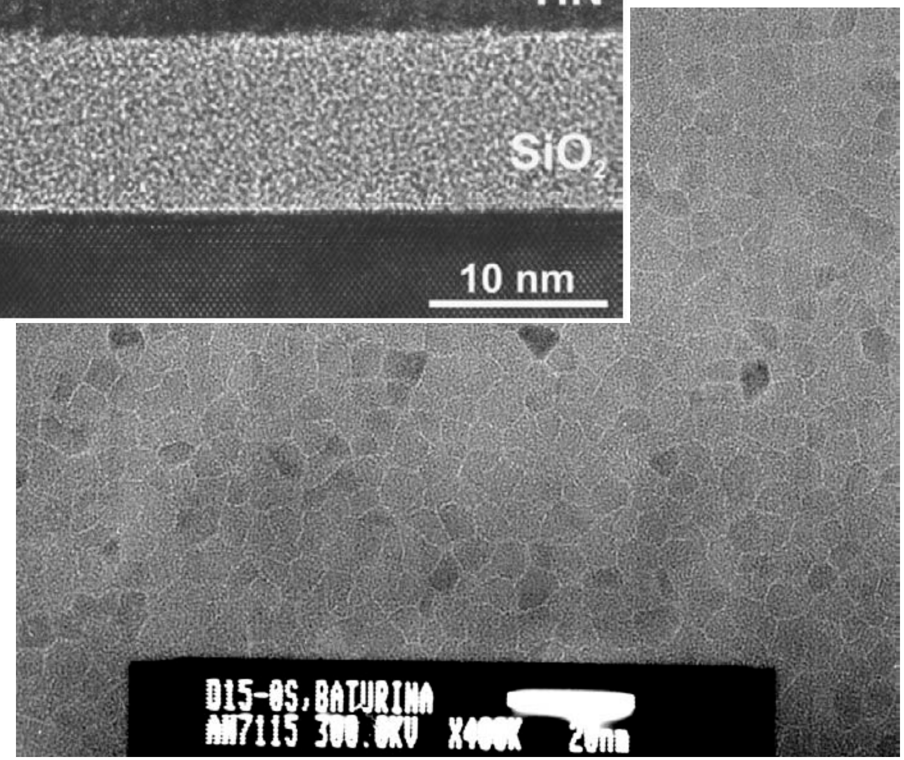
prepare squares between 0.5 and 500 µm side length



characterization of the TiN films







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

✓ Composition:

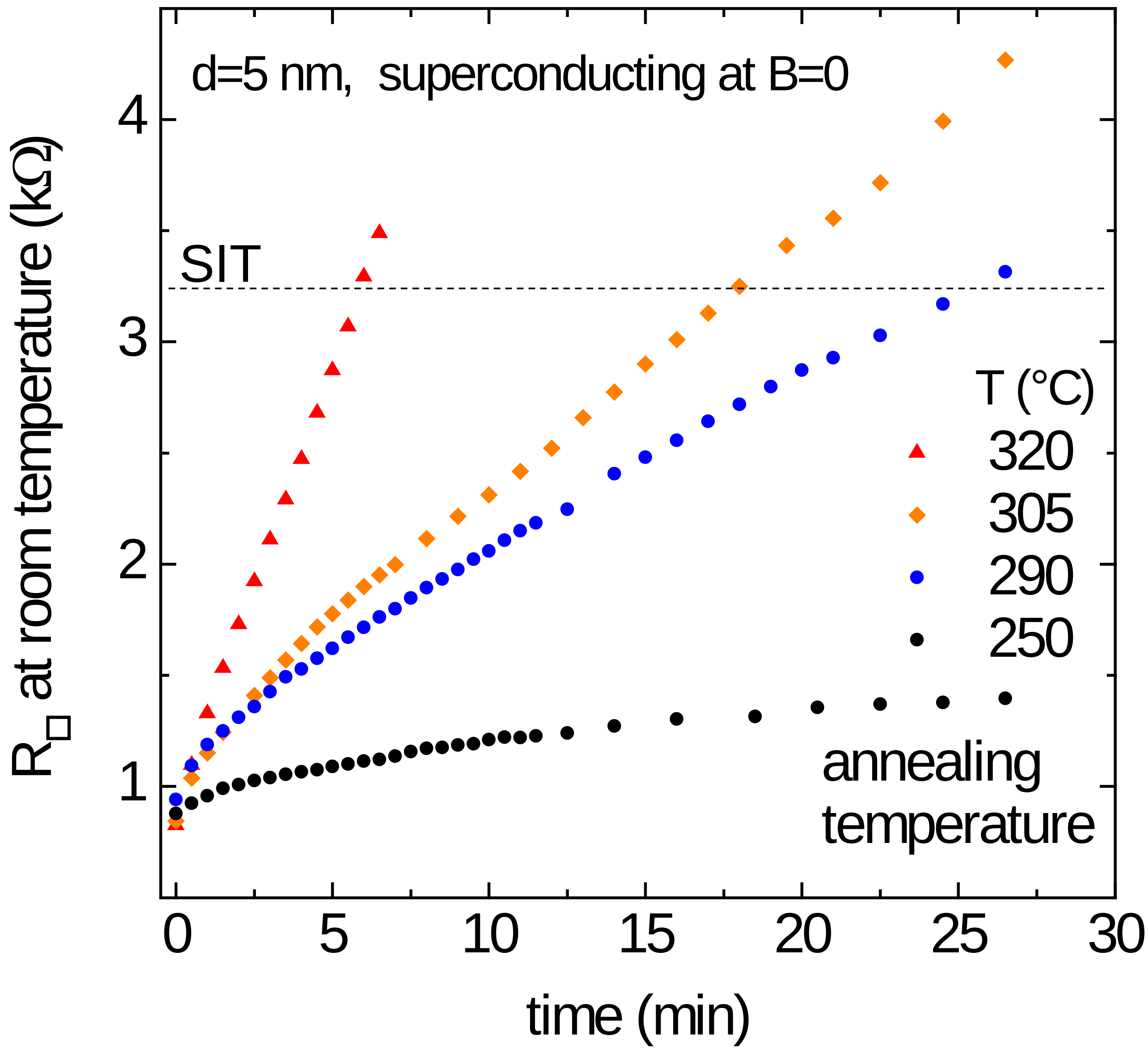
Ti	N	Cl
1	0.94	0.035

thickness d = 5 nm

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

ξ_d ~ 9 nm superconducting coherence length

controlled oxidation of TiN films in air



Is there granularity in TiN?

Two possiblities:

structural granularity

Stranski-Krastanov (island) growth mode: film closure at d = 4nm

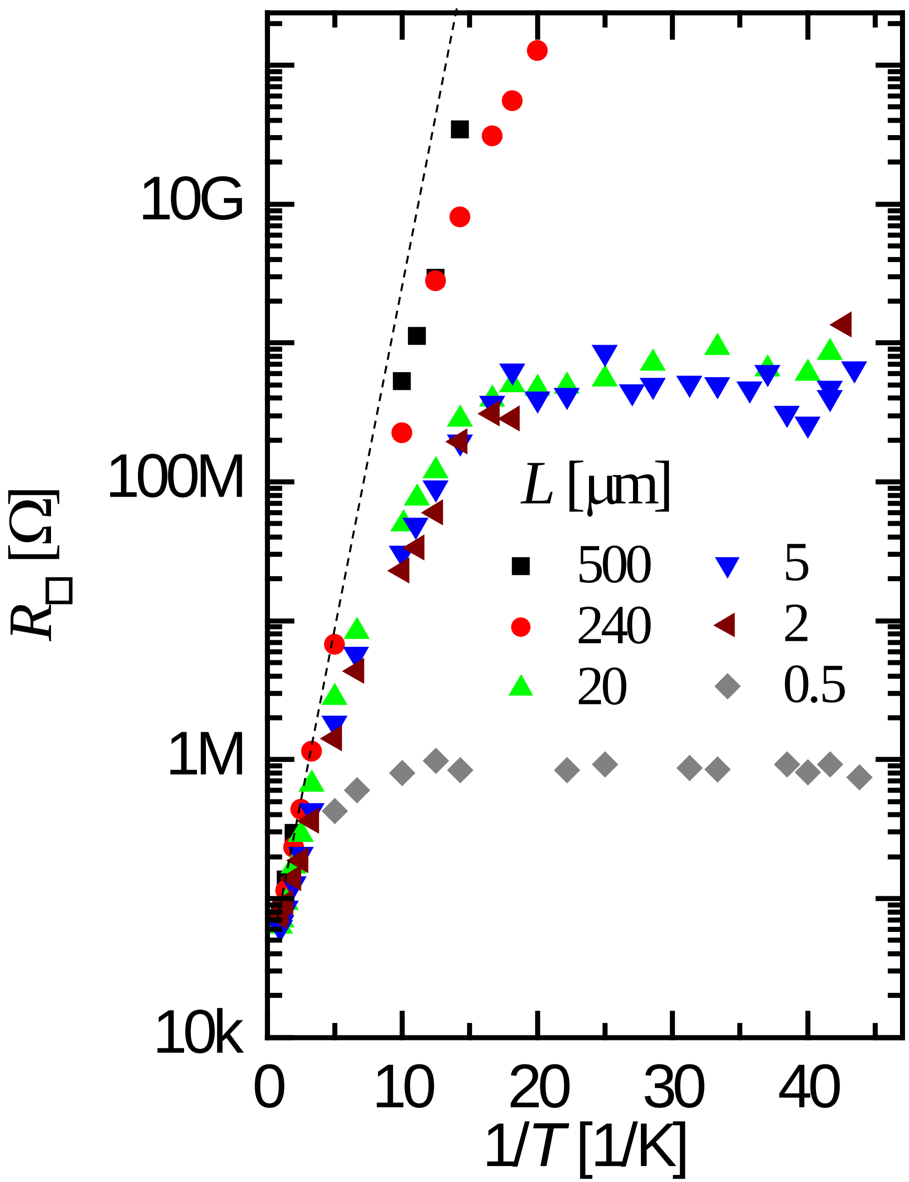
preferred oxidation at grains boundaries?

'self-induced' granularity

disorder potential produces Josephson coupled SC 'droplets'

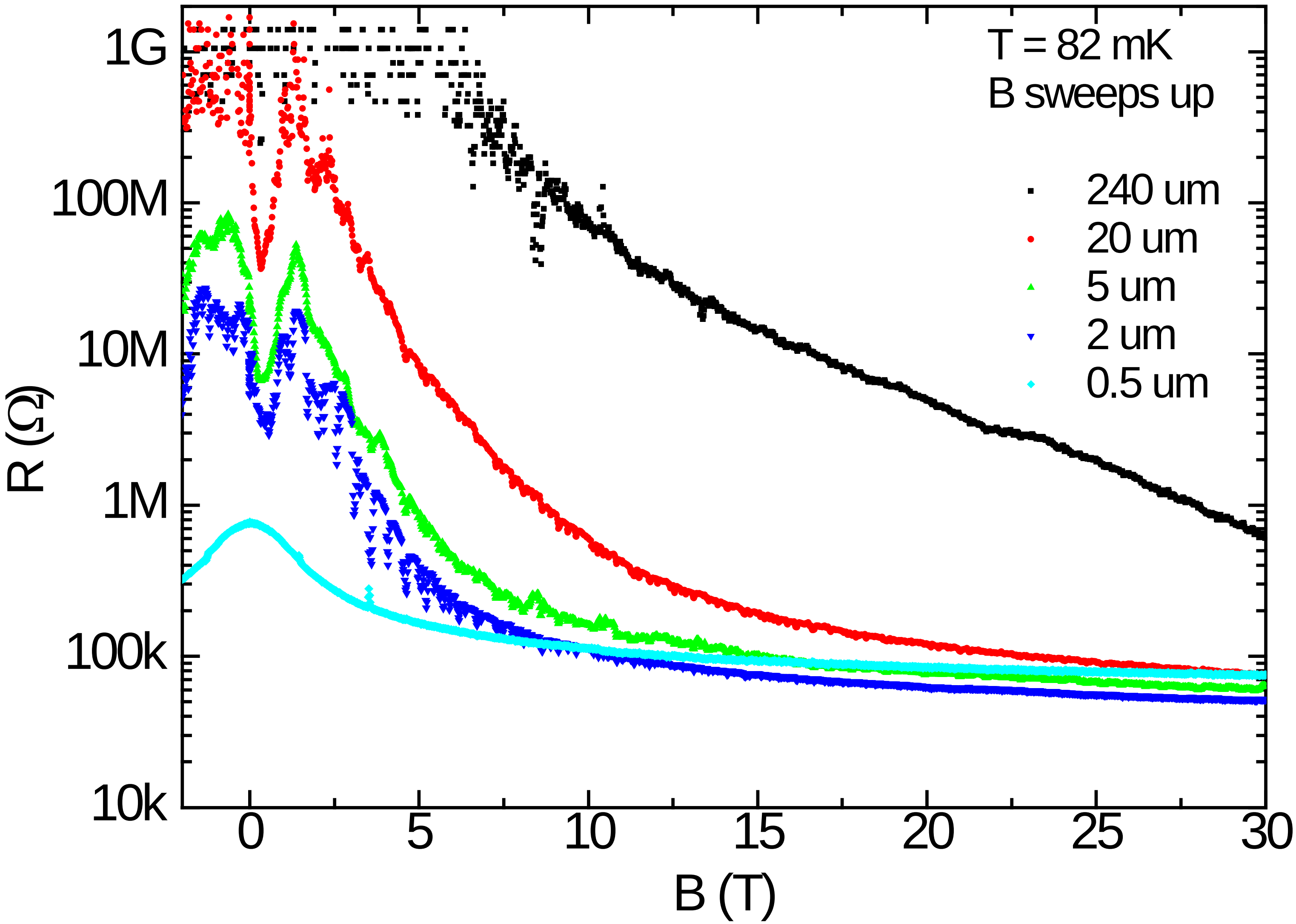
Kowal & Ovadyahu, Sol.Stat. Comm '94
Trivedi et al. PRL '98
Sacepe et al. PRL 2008, Nat.Phys. 2011
Bouadim, et al. Nat.Phys. 2011

$R_\square(T)$ is indeed *size dependent*:

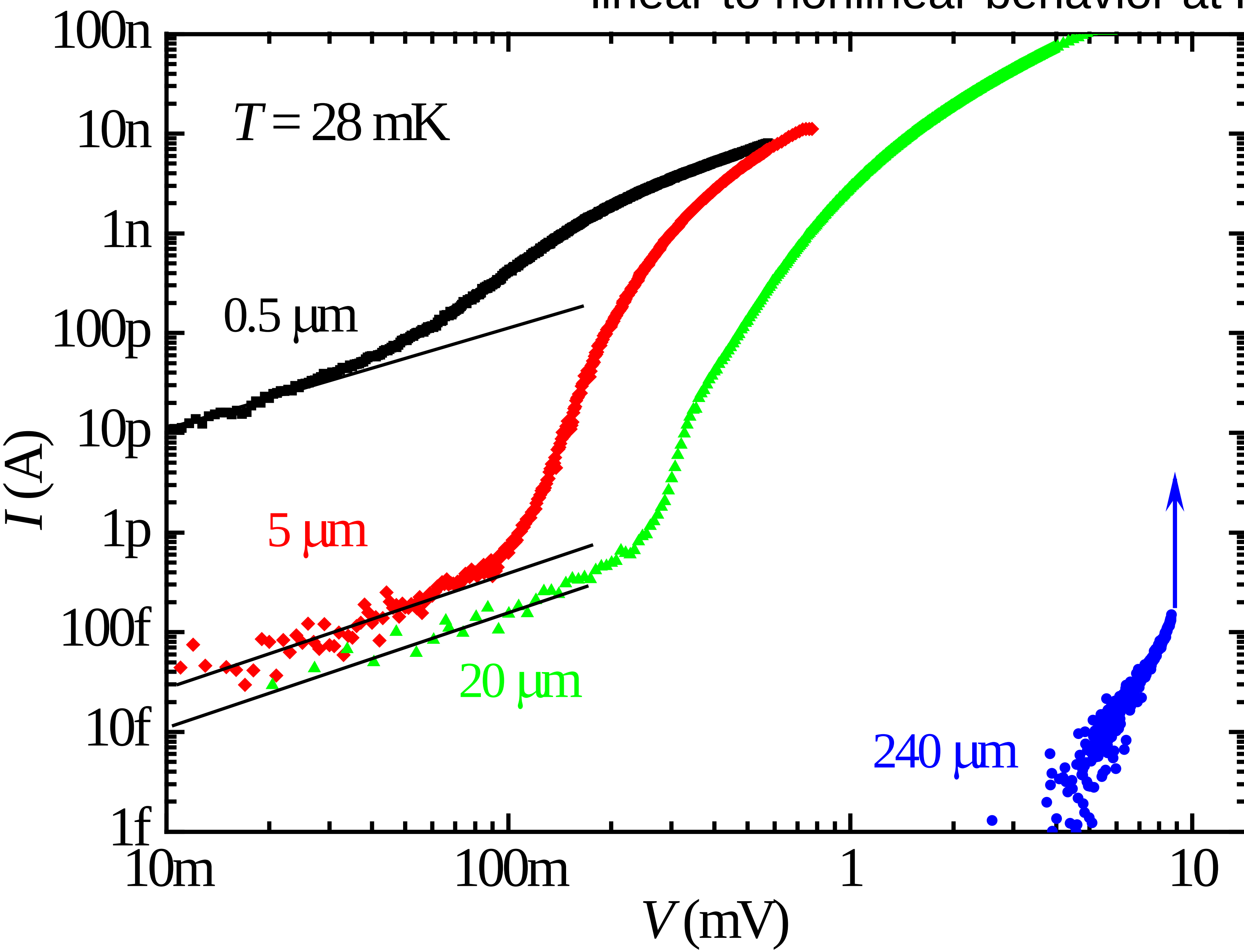


- sample insulating at B = 0
- each point extracted from full IV-characteristic
- larger samples more insulating
- saturation of R at low T, except for the largest samples
- Arrhenius behavior at higher temperature

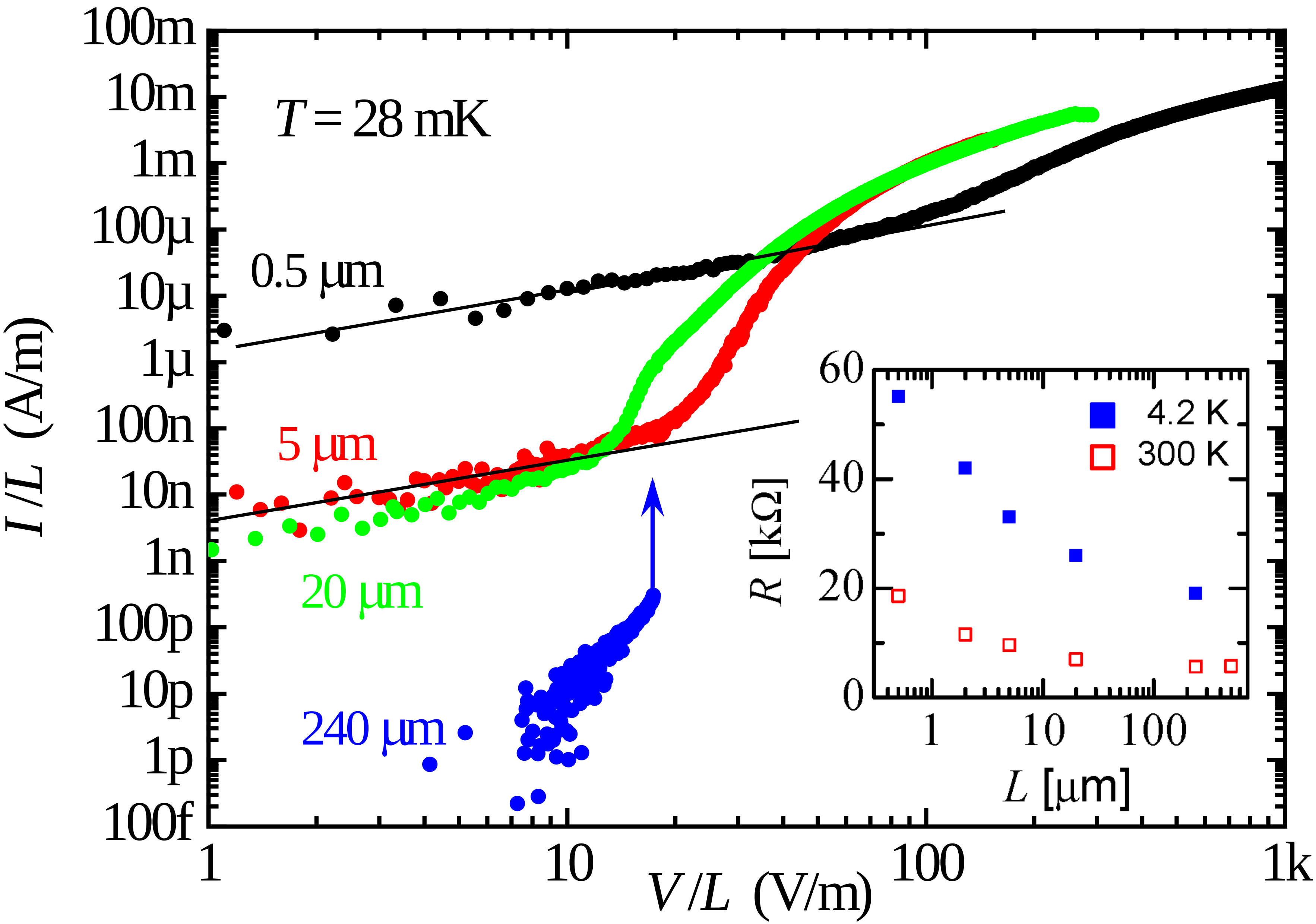
magnetoresistance strongly depending on sample size



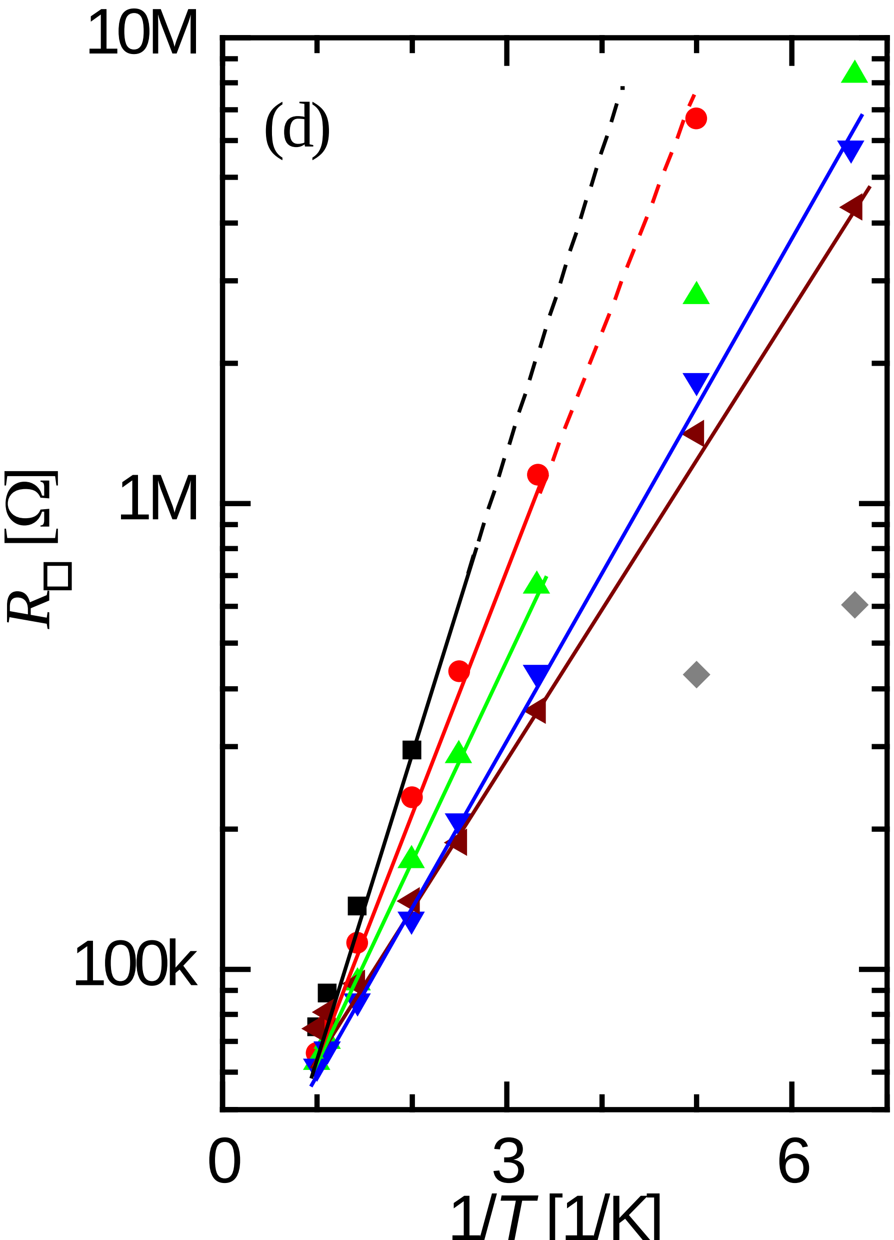
Measurement at High Magnetic Field Laboratory Nijmegen *size dependent* IVs display cross-over from linear to nonlinear behavior at low T



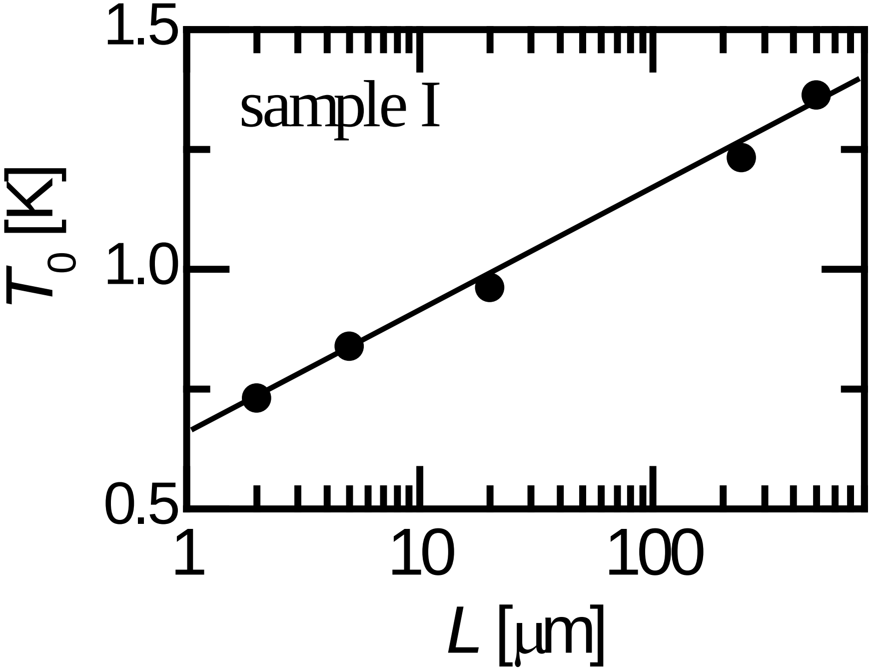
size dependent IVs display cross-over from linear to nonlinear behavior at low T



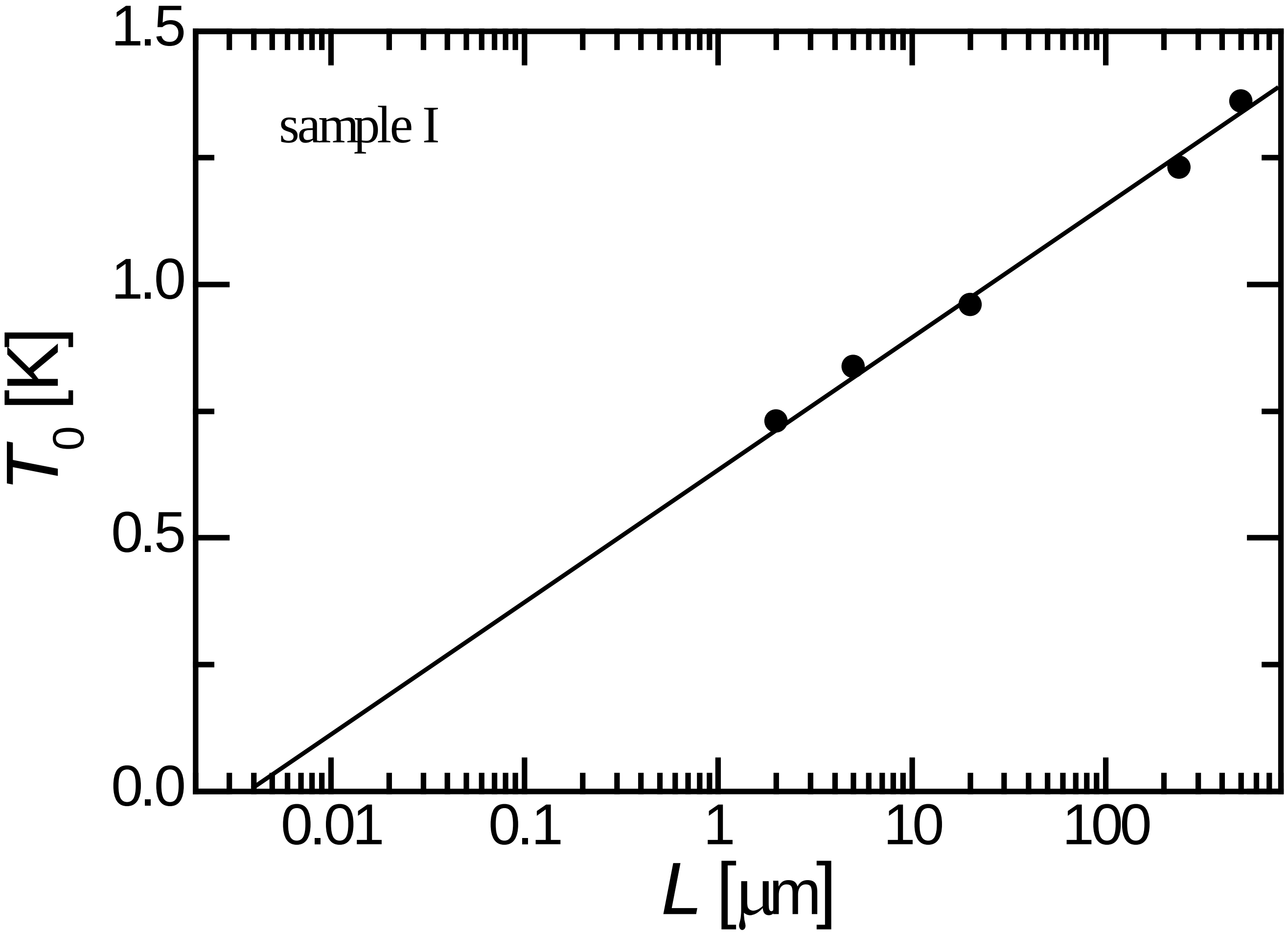
extract *size dependent* activation energy:



logarithmic increase of T_0 with size L

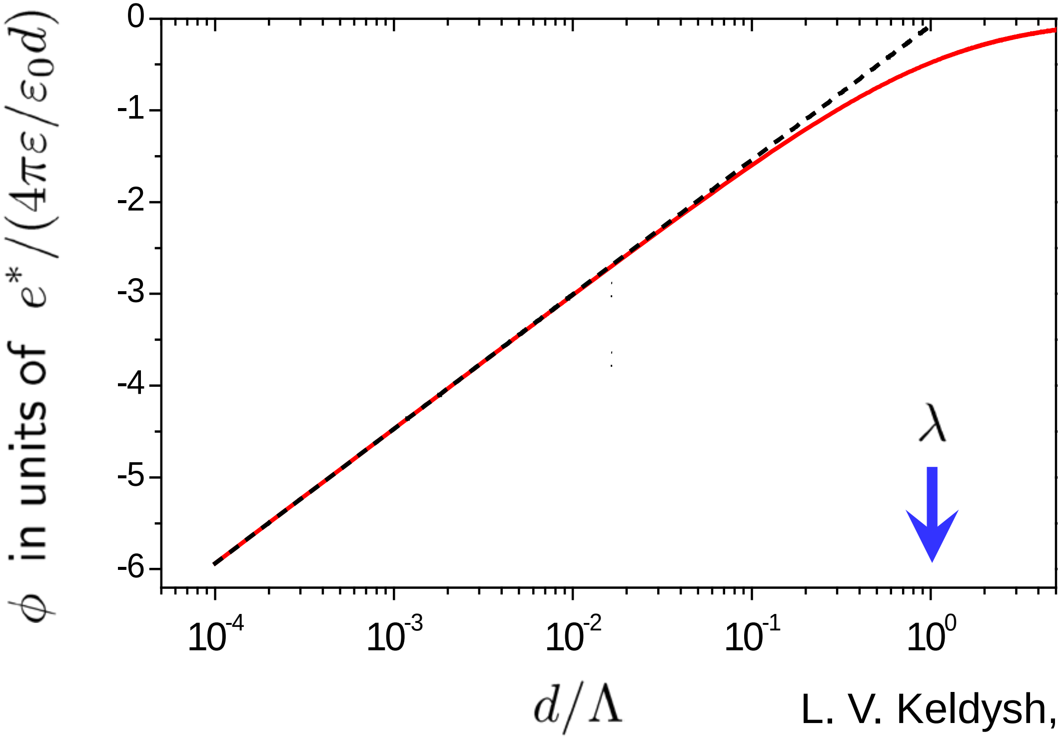


extract network parameters governing activation energy:



$$k_B T_0 = \Delta_C/2 = \frac{E_C}{4} \ln\left(\frac{L}{d}\right) \longrightarrow \begin{cases} E_C/k_B = 0.04\,\text{K} \\ d = 4\,\text{nm} \end{cases}$$

employ continuum model to extract dielectric constant:



electrostatic penetration depth

$$\lambda = \frac{\varepsilon d}{\varepsilon_1 + \varepsilon_2} \simeq 240\,\mu\mathrm{m}$$

compatible with measured sizes

L. V. Keldysh, JETP Lett. **29**, 658 (1979)

interaction energy between charge/anti-charge pair:

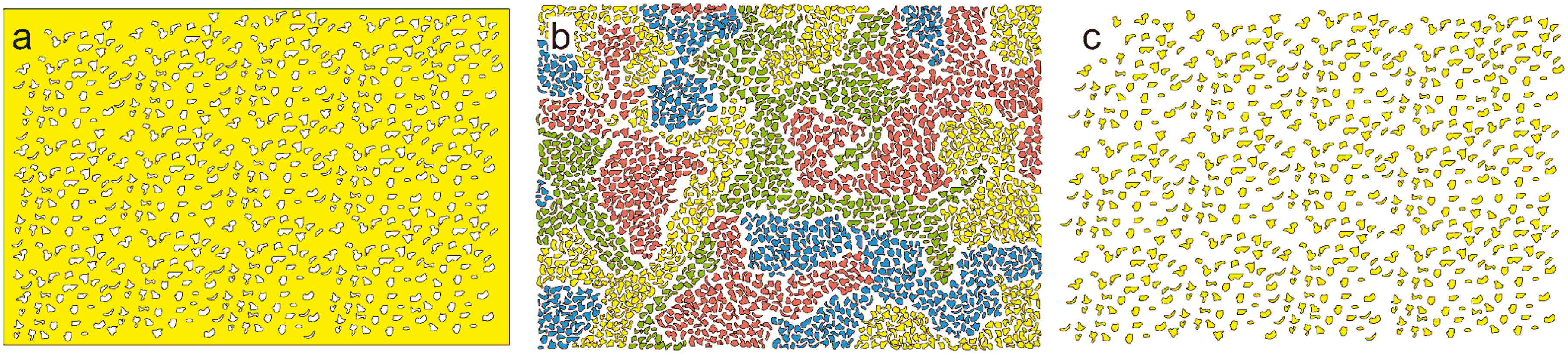
$$\phi(r) = \frac{(e^*)^2}{4\pi\varepsilon\varepsilon_0 d} \ln\left(\frac{r}{\lambda}\right)$$

for $d << r << \lambda$

$$E_C \simeq \frac{(e^*)^2}{(4\pi\varepsilon\varepsilon_0 d)} \longrightarrow \varepsilon \simeq \frac{(e^*)^2}{E_C 4\pi\varepsilon_0 d} \simeq 4 \cdot 10^5$$

large value!

explanation for large values of ε?



a) localized Cooper-pairs for droplets in superconducting matrix:

⟶ **superconducting films** (see B. Sacepe, Nat. Phys 2011)

b) insulating droplets form percolating network wirh diverging ε at the percolation threshold

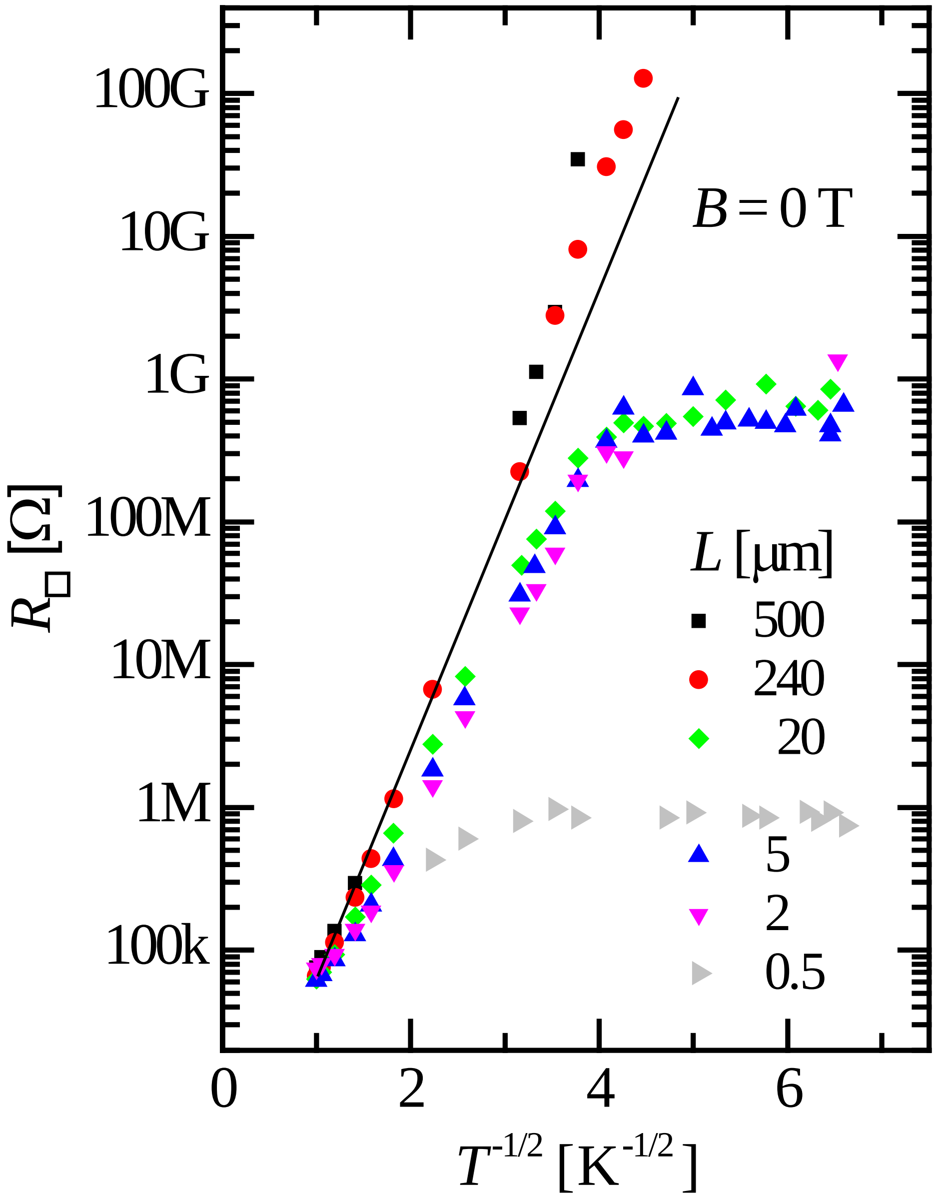
⟶ **superconductor/insulator transition**

c) superconducting droplets in insulating matrix

⟶ **Cooper-pair insulator**

to be verified experimentally!

behavior not far from Efros-Shklovskii,



but deviations at

- high temperature:

  $\sim \exp(T_0/T)$

- low temperature:

  – upturn in large samples

  – saturation in small samples

see also tin/graphene granular films:
A. Allain, Z. Han, and V. Bouchiat,
Nature Materials 11, 590 (2012).

Charge Berezinskii-Kosterlitz-Thouless transition

- Combine free motion of particles in 2d with *logarithmic* interaction potential

→ competition between binding energy and entropy

→ binding/un-binding transition of pairs of particles

- applicable to superconducting (vortices) and insulating (charges) side of the transition

$$E_p = 2E_C \ln\left(\frac{L}{d}\right) \qquad \Delta S_p = k_B \ln\left(\frac{L^2}{d^2}\right)$$

$$\Delta F = E_p - T\Delta S_p = (2E_C - 2k_B T)\ln\left(\frac{L}{d}\right)$$

→ $T_{BKT} = E_C/k_B$

E_C : charging energy of single island

Charge-Berezinskii-Kosterlitz-Thouless transition

- high temperature regime: $T >> T_{BKT}$

→ mobile charges with a density ~ $\exp(-\Delta_C/2k_BT)$

→ thermally activated conductance

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

- low temperature regime: $T \sim T_{BKT}$

→ binding into (neutral) charge/anti-charge pairs

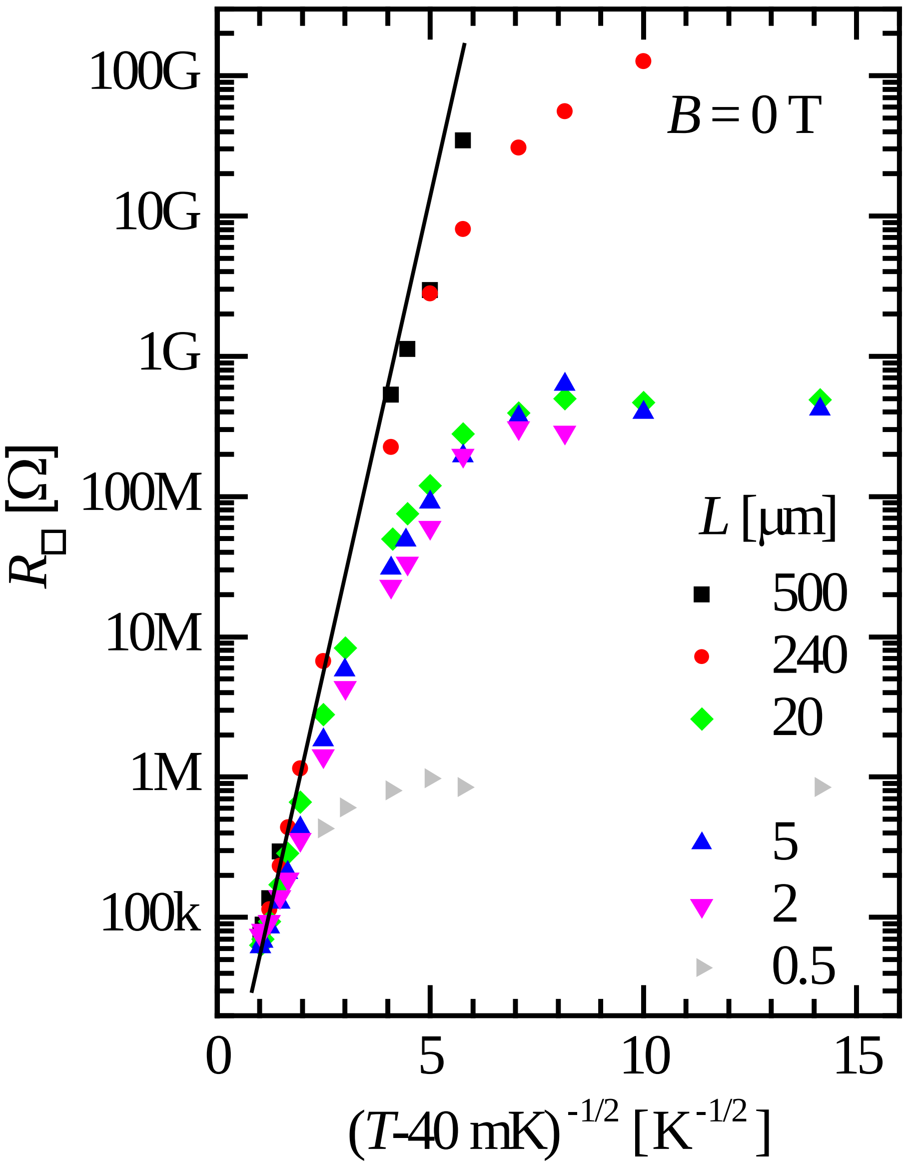
→ very few residual mobile charges

with density $n(T) \;\propto\; \frac{1}{\xi^2(T)}$

- $\xi(T)$ is a correlation length describing the distance between charged excitations

J. E. Mooij *et al*., PRL. **65**, (1990).

best fit: BKT-like 'square-root cusp formula'



$$R_\square(T) = R_T \left(\frac{\xi(T)}{\xi_0}\right)^2 = R_T \frac{A^2}{2\pi} \exp\left(\frac{2b}{\sqrt{|1 - T/T_{\rm BKT}|}}\right)$$

where

$$\xi(T) = \xi_0 \exp\left(\frac{b}{\sqrt{|1-T/T_{\rm BKT}|}}\right)$$

and A and b are constants of order 1

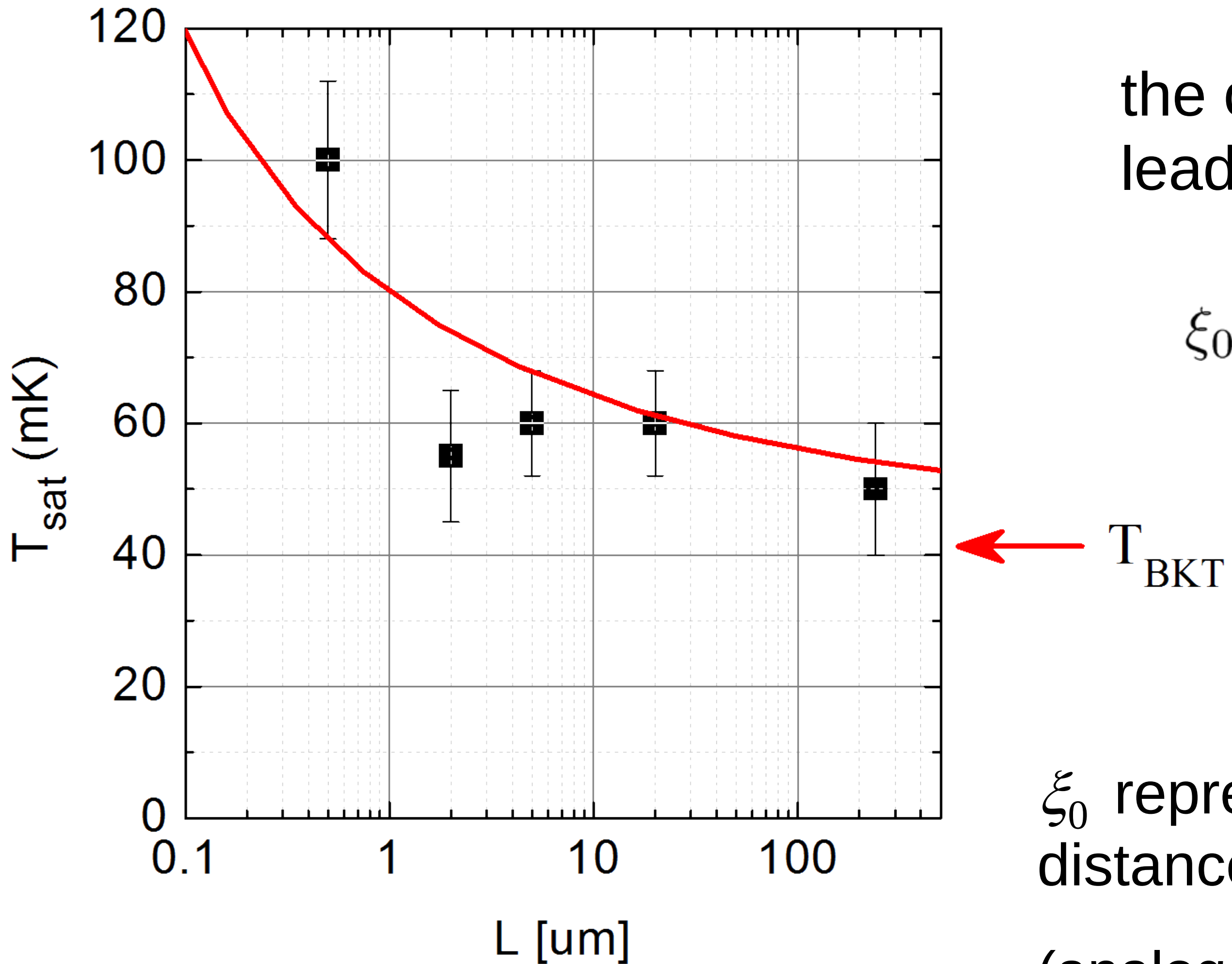
$$T_{\rm BKT} = E_C/k_B \simeq 40\,{\rm mK}$$

is the BKT-transition temperature extracted from the size-dependence $T_0(L)$

Divergence of R(T) is expected to be cut-off at $\xi(T) = L$:

Explanation of saturation of *R*(*T*) !

rough estimate of ξ_0 from saturation temperatures



the condition $\xi(T) = L$ leads to :

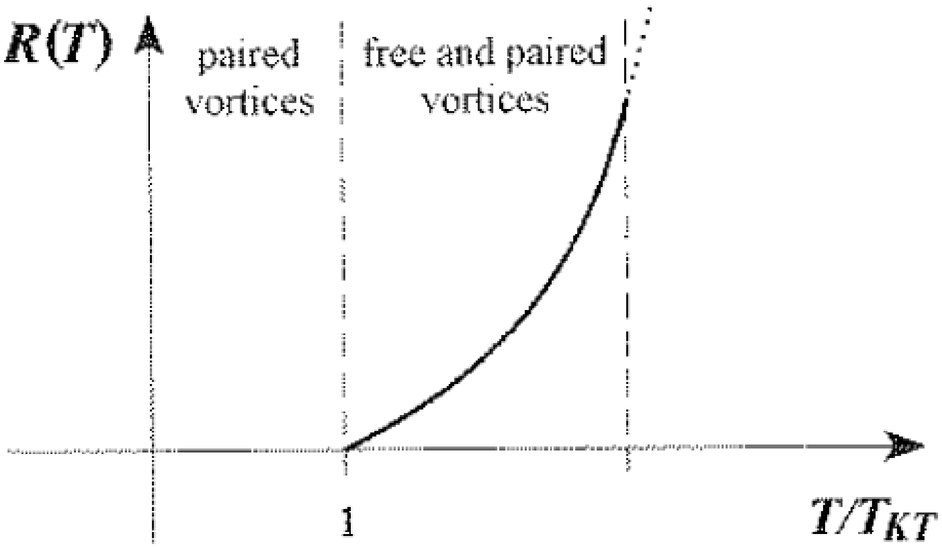
$$\xi_0 \simeq 60 \pm 30 \text{ nm}$$

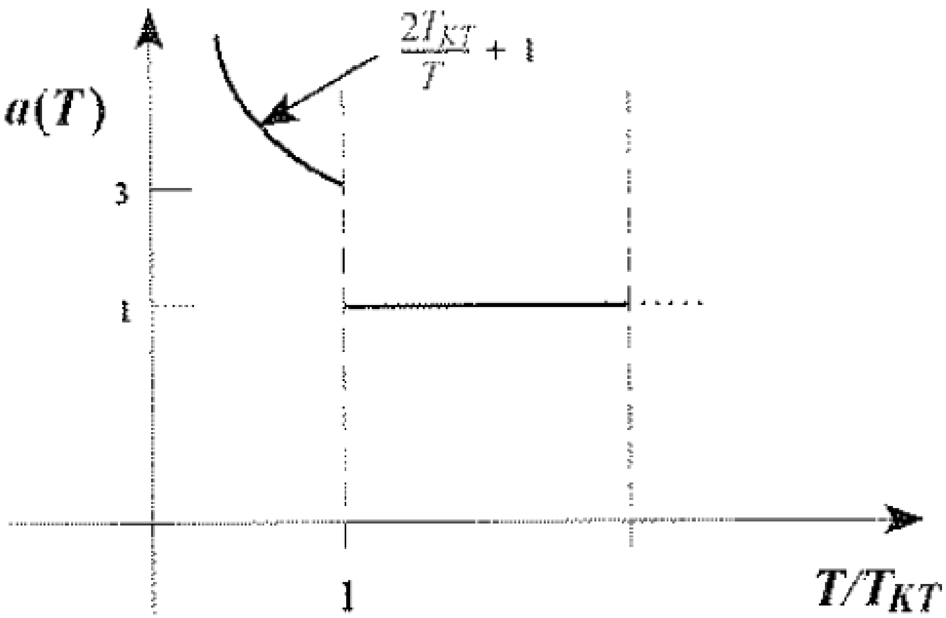
ξ_0 represents a minimal distance of charge solitons

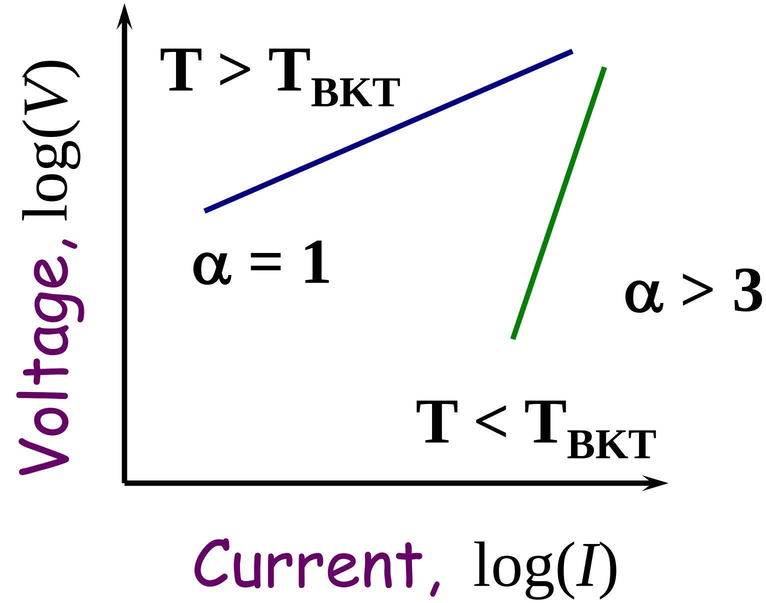
(analog to size of vortex core)

L. Benfatto *et al.*, PRB **80**, 214506 (2009)

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



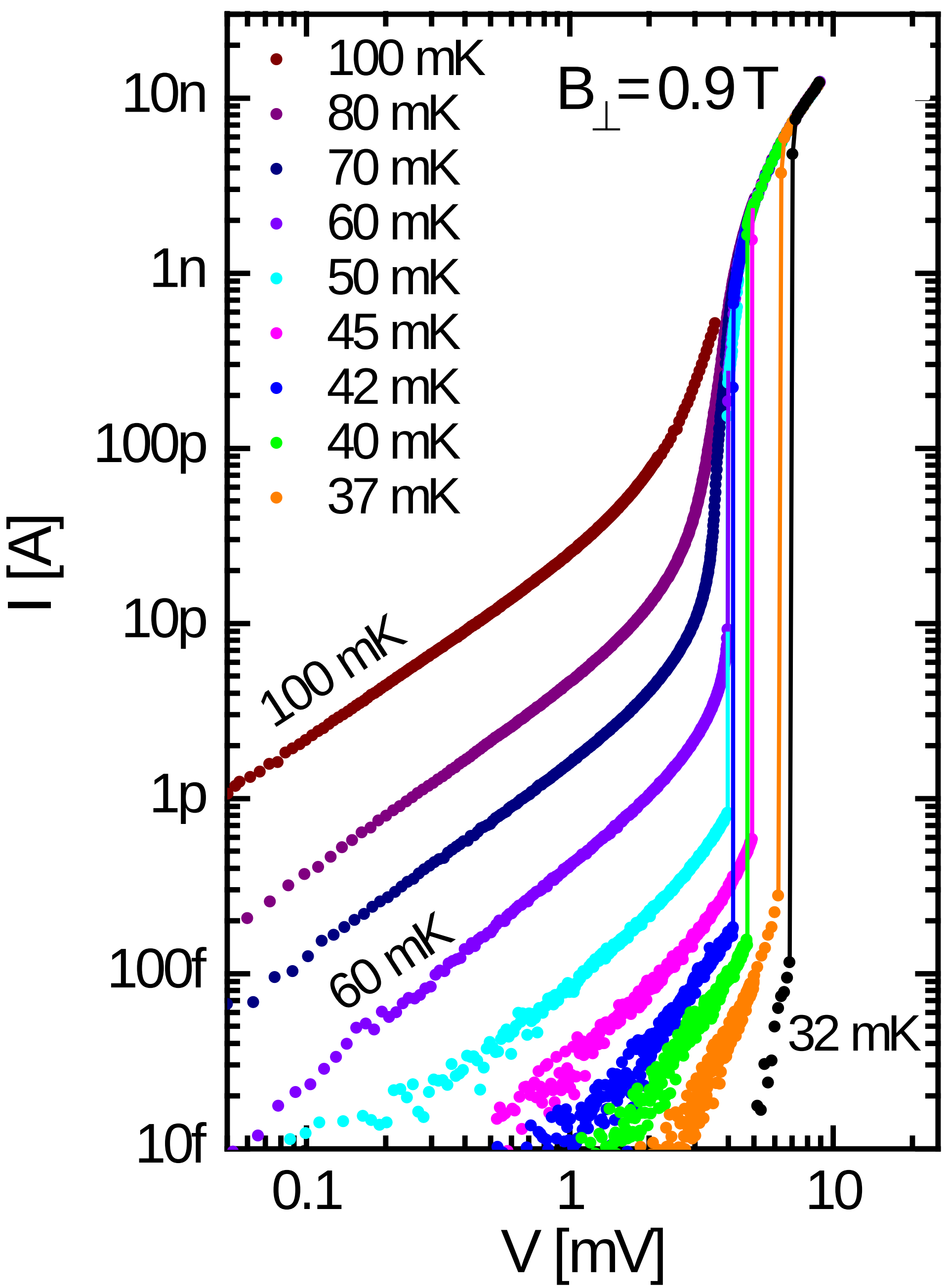




$$V \propto I^{\alpha(T)}$$

A dual (I ⟷ V) scenario is expected to hold in the charge regime $E_C > E_J$

strongly non-linear IV-characteristics at the lowest T:



intermediate temperature:
(50 mK < T < 300 mK):

jump probably induced by electron heating

see data in InO_x (Weizmann)

Sambandamurthy et al. PRL (2005)
Ovadia et al. PRL **102** (2009)
Altshuler et al., PRL **102** (2009)

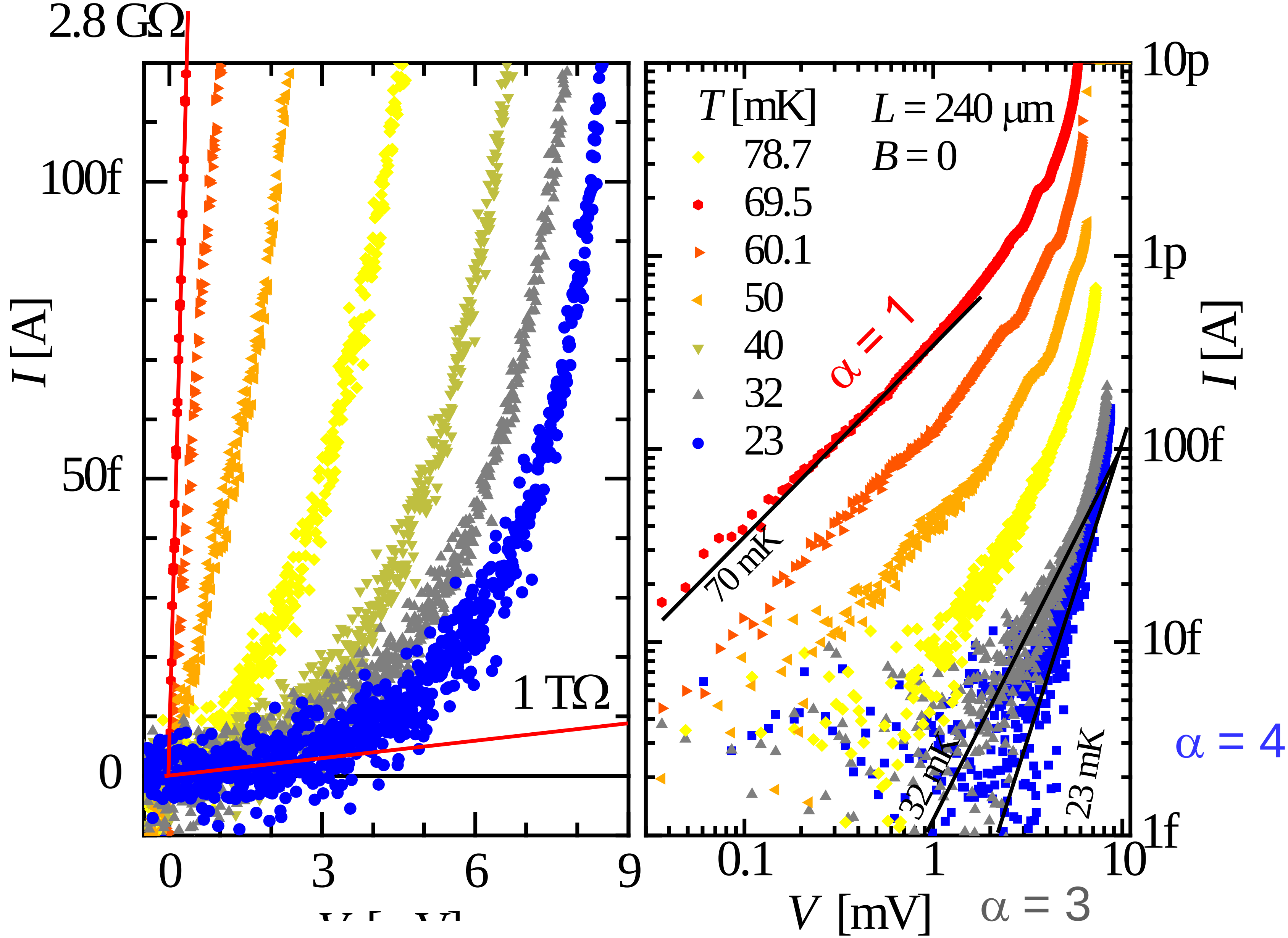
At the lowest T:

I(V) develops power law behavior with exponent α(T)

random scatter of switching voltage

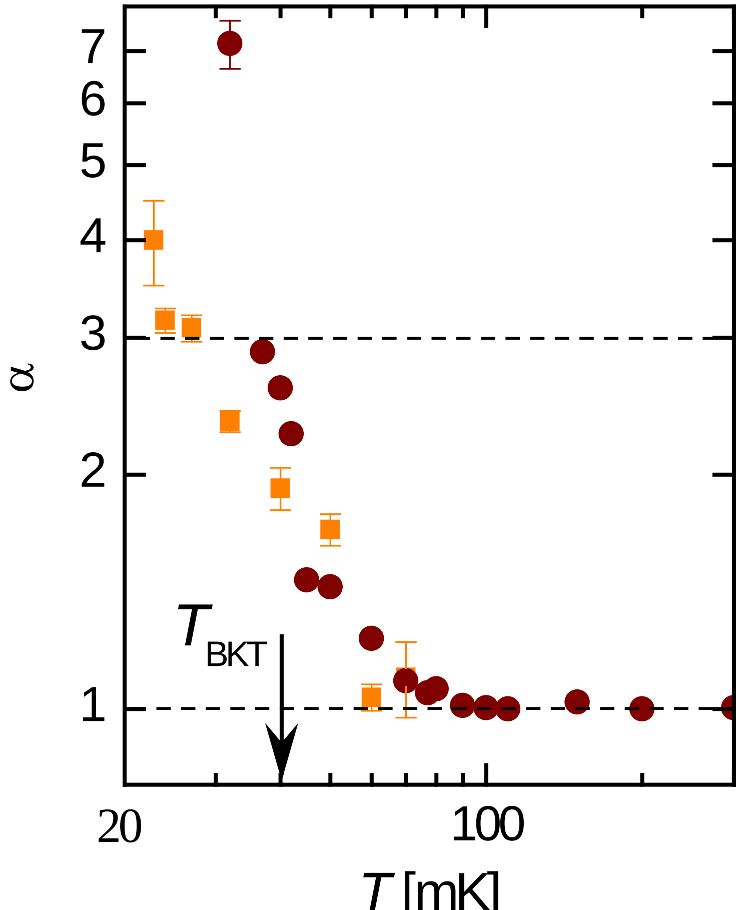
nature of insulating state?

Another sample – insulating at B = 0



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

L. Benfatto *et al.*, PRB **80** (2009)



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 $\alpha = 3$

very good agreement with independent estimate of T_{BKT} from activation energies

Conclusions

- dramatic size dependence of all transport properties near the SIT

- this size dependence can be explained in terms of a ***charge-BKT-transition***, i.e., a large electrostatic penetration depth and a diverging correlation length

- non-linear I(V)-characteristics coincide very well with analysis of size dependence

the conductance vanishing at ***finite*** T_{BKT} supports the notion of a '***superinsulator***' Vinokur et al., Nature **452** (2008)

Open Questions

- direct measurement of dielectric constant

- hyperactivated behavior

- role of quantum phase slips, or analogous processes

-role of electron heating effects