Magnetic Field Induced Super-Insulating State and Enhancement of Superconductivity in Ultrathin Pb Films

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Peng Xiong

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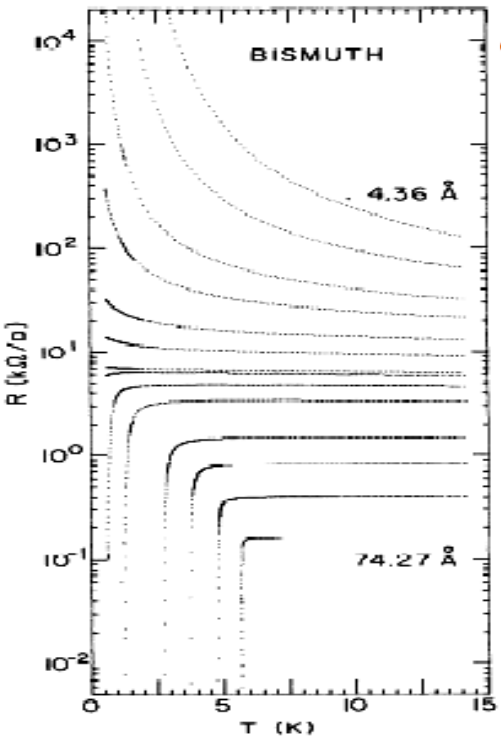
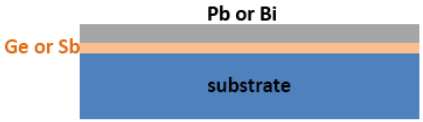
Outline

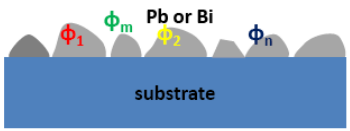
- **Introduction**
- **Experimental Setup and Procedure:**
  - ***in situ* film growth and measurement**
  - **superconductor-insulator transitions tuned by disorder, magnetic impurity, and magnetic field**
- **Perpendicular Field:**
  - **magnetic field-induced super-insulating state**
- **Parallel Field:**
  - **magnetic field enhancement of superconductivity**

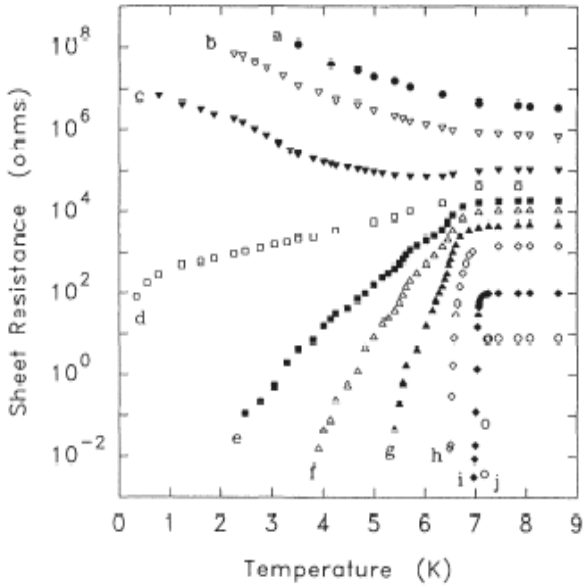
Superconductor-insulator transitions

$$\Psi = \sqrt{\Delta} e^{i\phi}$$









Haviland *et al*, PRL 1989

Barber *et al*, PRB 1993

Signatures of a bosonic SIT

d-tuned SIT in granular films as a model system for Bosonic transition:

Direct:

- **Well-defined order parameter amplitude in insulating state**

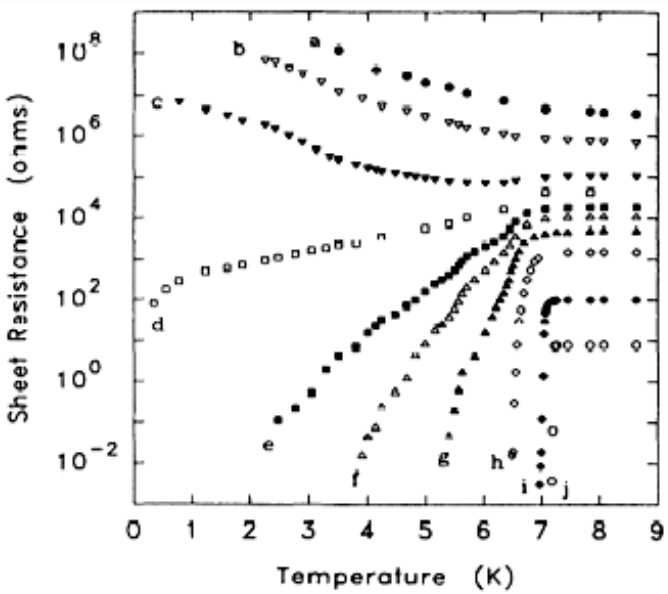
Indirect:

- **Broad resistive transitions**
- **Reentrance behavior in critical region**
- **Activated/hopping transport immediately on insulating side**
- **Large negative MR in insulating state**

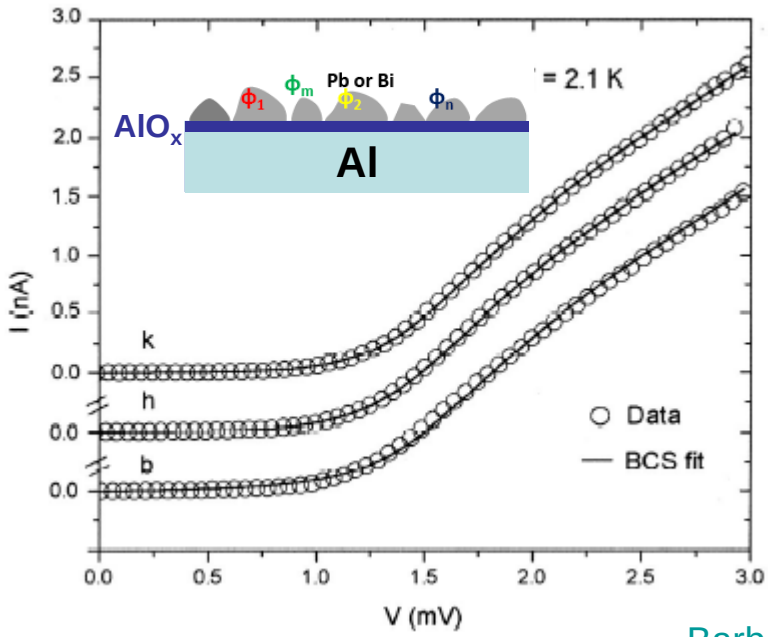
Direct evidence of pairs in insulating state

granular Pb films

d-tuned SIT



NIS tunneling



Barber *et al*, PRB 1994

Merchant *et al*, PRB 2001

$$I_{sn}(T) = \frac{G_n}{e}\int_{-\infty}^{+\infty}\frac{E}{\sqrt{E^2-\Delta^2}}[f(E-eV)-f(E)]dE$$

$$G_n \propto D_N(0)|T|^2$$

Fully developed Δ in the insulating state

Signatures of a bosonic SIT

d-tuned SIT in granular films as a model system for Bosonic transition:

Direct:

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Indirect:

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- **Activated/hopping transport immediately on insulating side**
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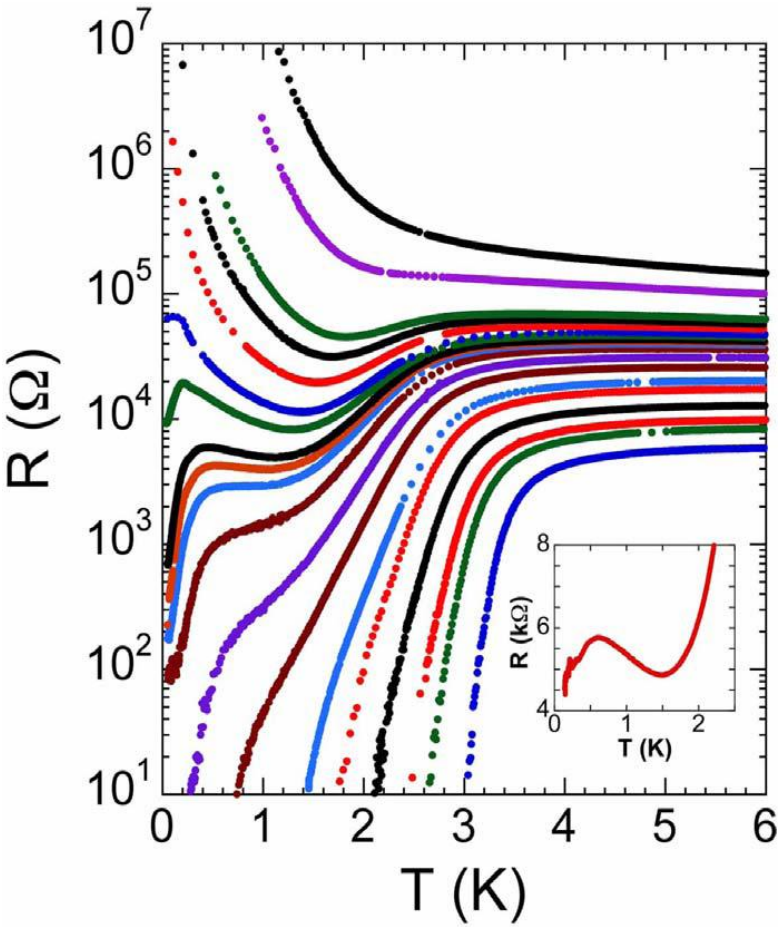
Indirect evidence of pairs in insulating state

granular Bi films

***d*-tuned SIT**



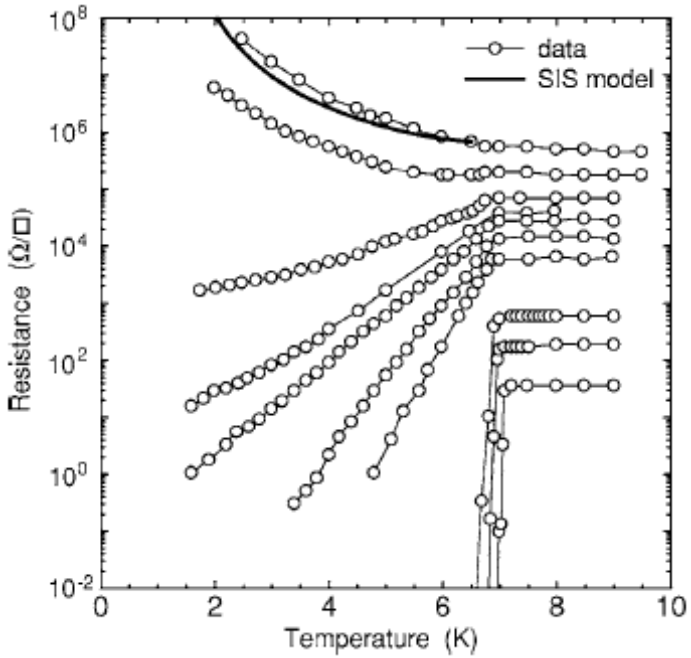
Reentrance and double-reentrance near SIT

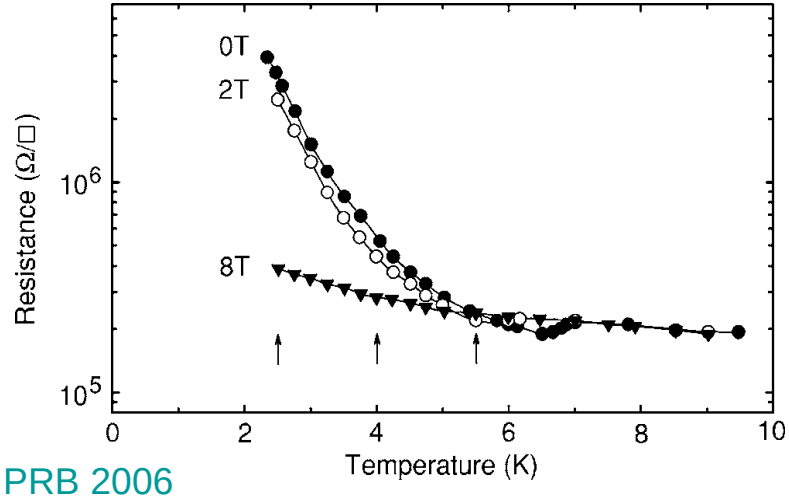
Parendo *et al*, PRB 2007

Indirect evidence of pairs in insulating state

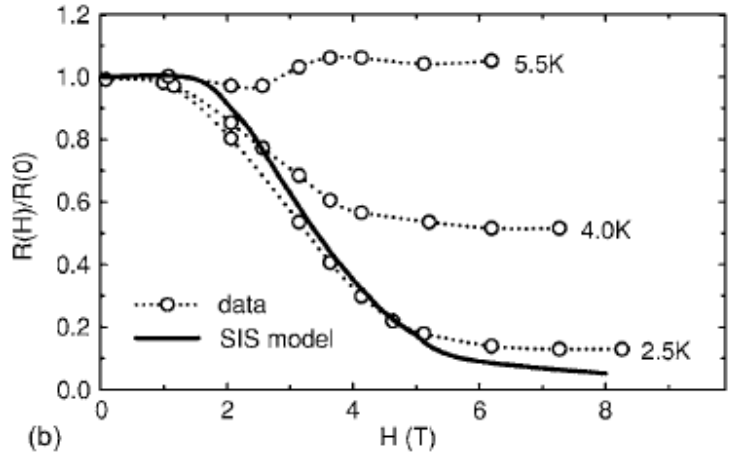
granular Pb films

d-tuned SIT





Barber *et al*, PRB 2006



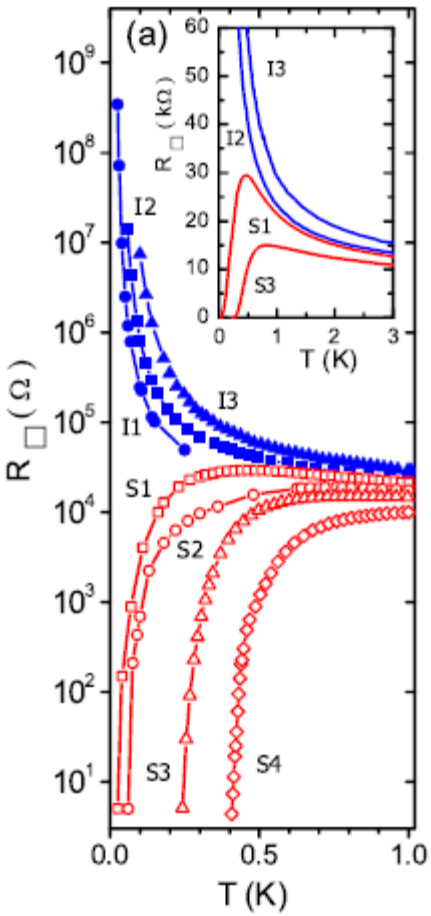
$$I_{ss'}(T) = \frac{1}{eR_n}\int_{-\infty}^{+\infty} \frac{|E-eV|}{\sqrt{(E-eV)^2-\Delta^2}} \frac{|E|}{\sqrt{E^2-\Delta^2}} [f(E-eV)-f(E)]dE$$

Activated transport

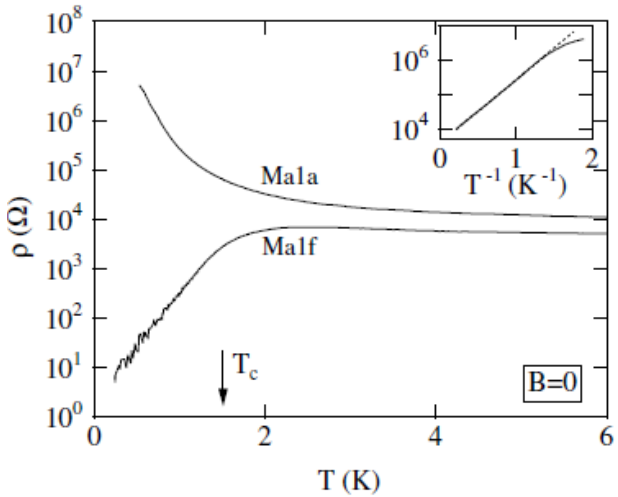
Large negative MR

SITs in 2D homogeneous films?

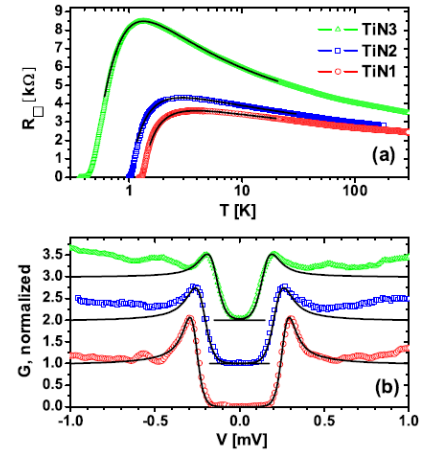
The case of binary compound superconductors: disorder tuning

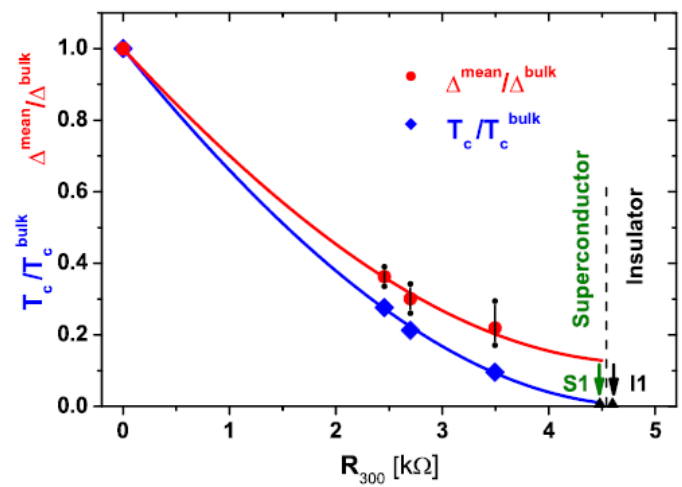


Baturina *et al*, PRL 2007



Sambandamurthy *et al*, PRL 2004

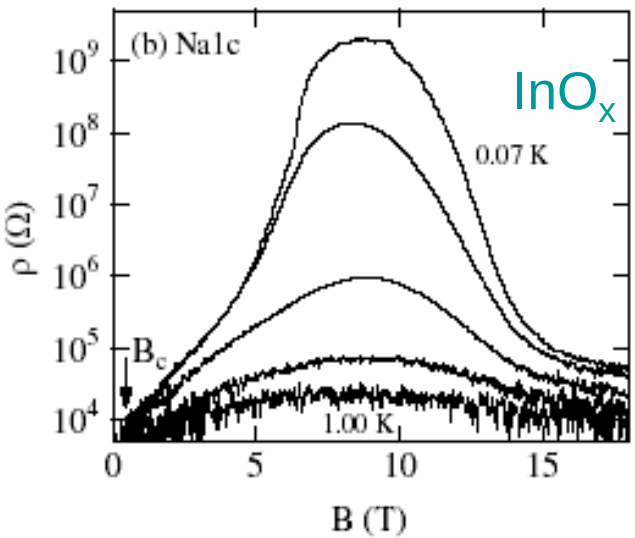




Sacepe *et al*, PRL 2008
Sacepe *et al*, N Phys 2011
Mondal *et al*, PRL 2011
Sherman *et al*, PRL 2012

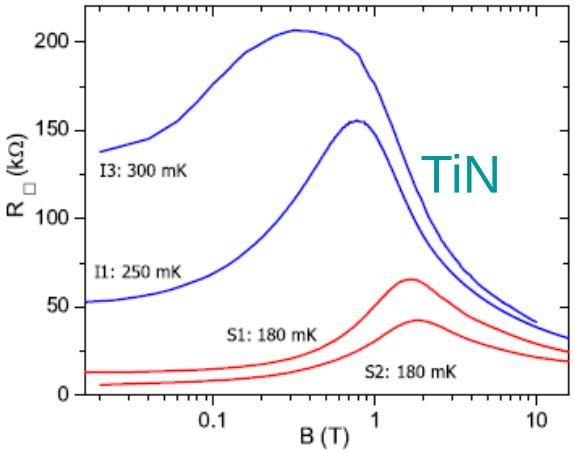
SITs in 2D homogeneous films?

The case of binary compound superconductors: magnetic field tuning

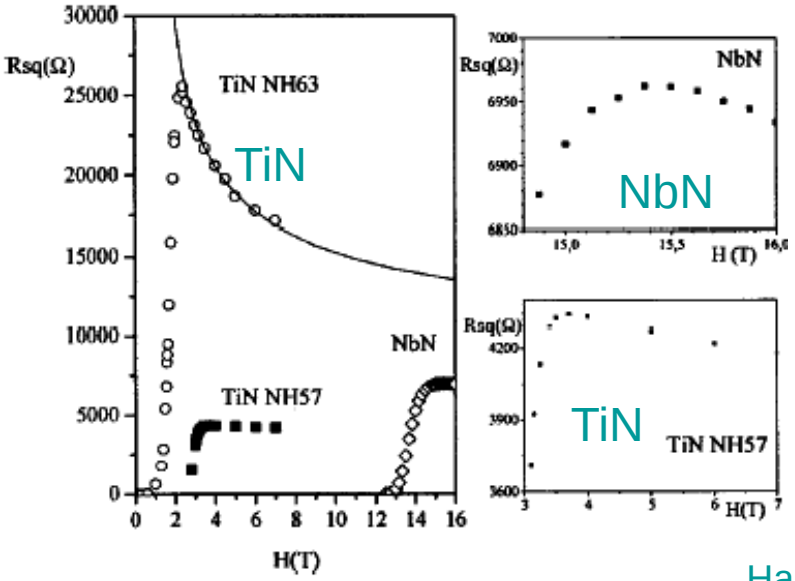


Paalanen *et al*, PRL 1991
Gantmakher *et al*, JETP Lett 2000
Sambandamurthy *et al*, PRL 2004
Steiner *et al*, PRL 2005

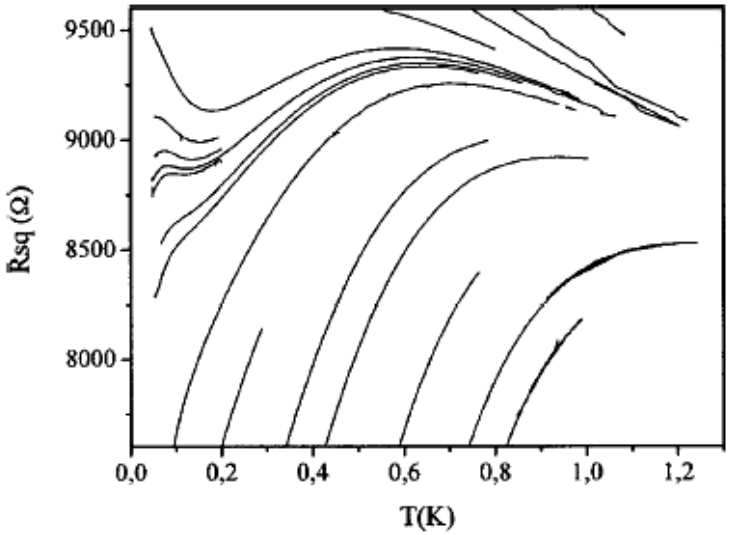
MR peak ➔
Large negative MR
at high field



Baturina *et al*, PRL 2007

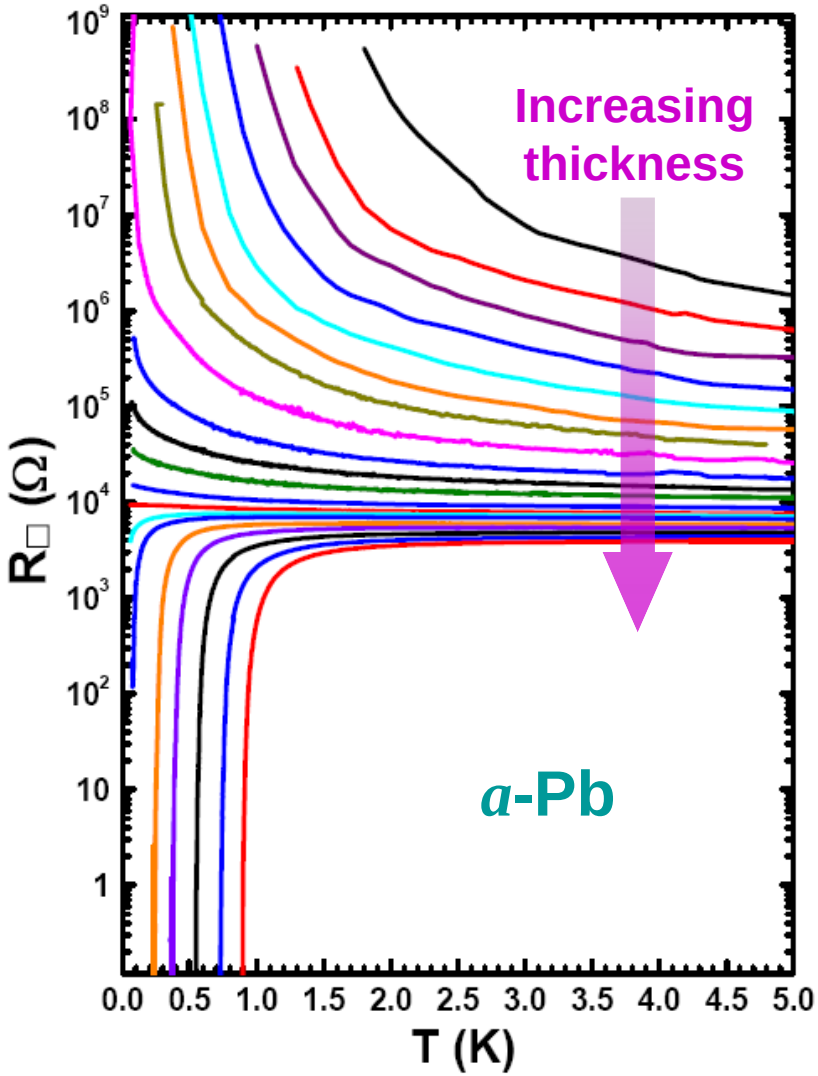


Hadacek *et al*, PRB 2004



Reentrance near B_c

SITs in 2D homogeneous films?



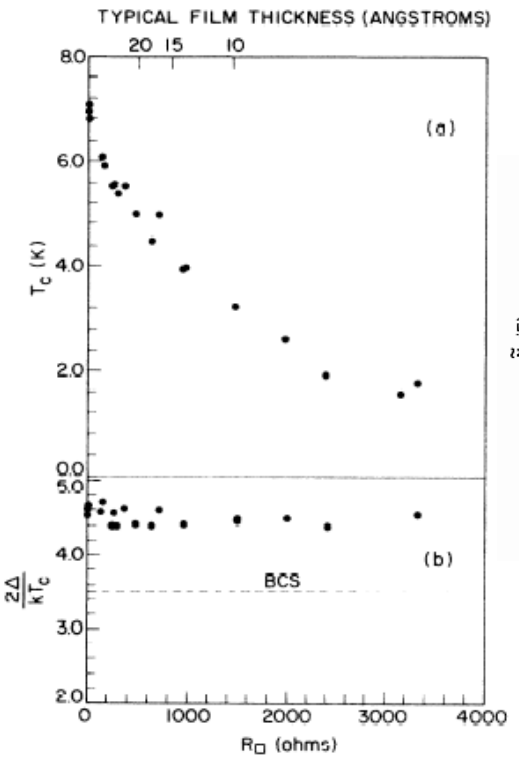
The case of elemental metal superconductors: disorder (thickness) tuning

- **Sharp resistive transitions**
- **Clear phase boundary (no reentrance)**
- **Diffusive transport immediately on insulating side**
- **No significant negative MR in insulating state (small positive low-field MR)**

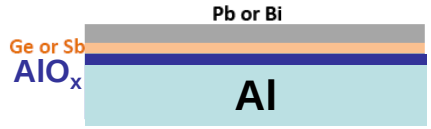
SITs in 2D homogeneous films?

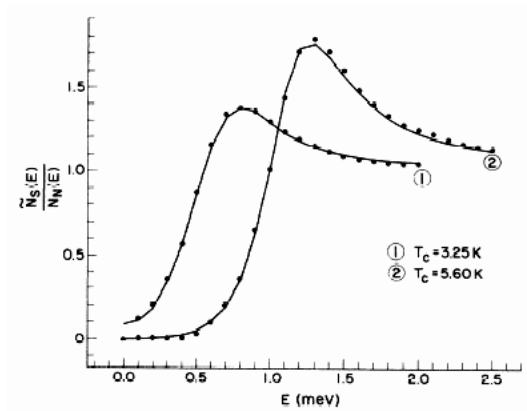
The case of elemental metal superconductors: disorder tuning

Dynes *et al*, PRL 1986



NIS tunneling

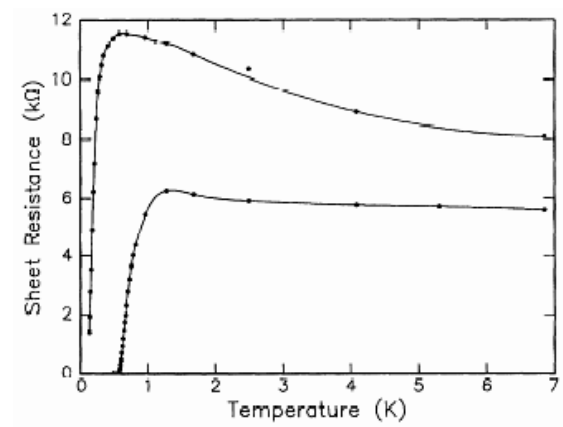


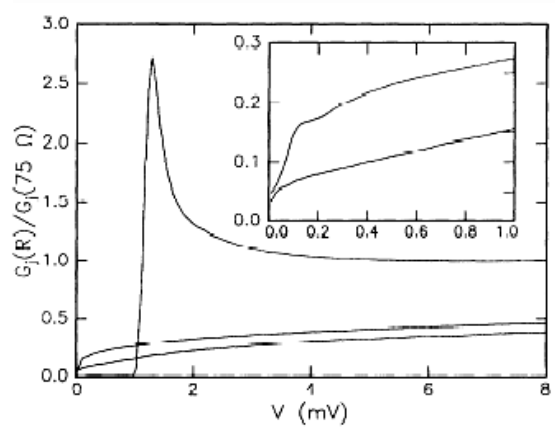


$$\frac{2\Delta}{kT_C}$$ **remains constant**

$$\left(\frac{dI_{sn}}{dV}\right)_T = G_n \int_{-\infty}^{+\infty} \frac{E}{\sqrt{E^2-\Delta^2}} \frac{df(E-eV)}{dV} dE$$

Valles *et al*, PRL 1992



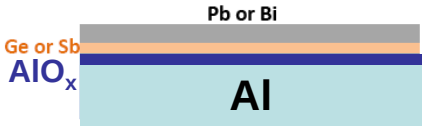


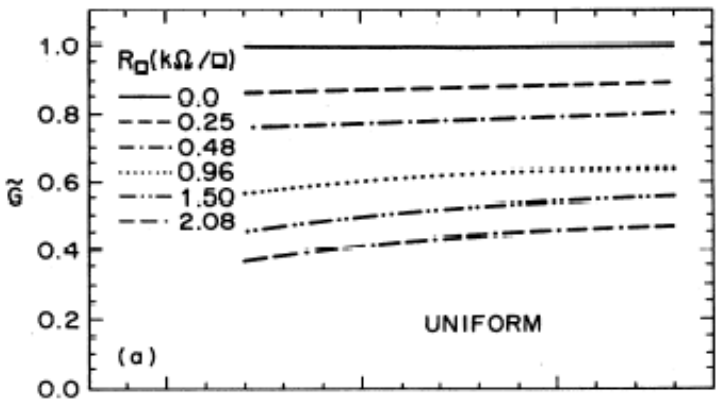
Δ vanishes at SIT

SITs in 2D homogeneous films?

The normal-state DOS: $D(E_F)$

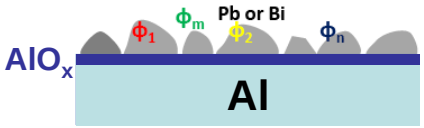
uniform Pb films

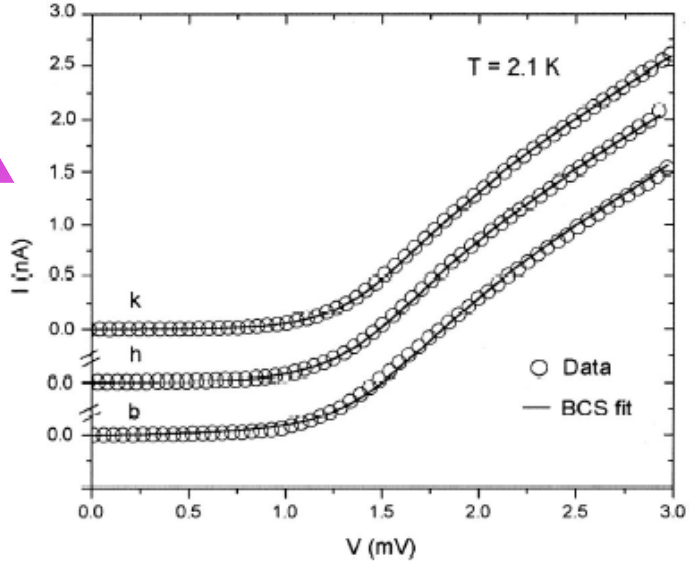




Valles *et al*, PRB 1989

granular Pb films





decreasing thickness

Barber *et al*, PRB 1994

Merchant *et al*, PRB 2001

$$\left(\frac{dI_{sn}}{dV}\right)_T = CD_N(0)|T|^2 \int_{-\infty}^{+\infty} D_S(E)\frac{df(E-eV)}{dV}dE$$

***$D(E_F)$* decreases concomitantly with T_C and Δ**

***$D(E_F)$* remains constant across SIT**

Motivation

Directly compare SITs tuned by

disorder (*d*), magnetic field (*B*), and magnetic impurities (*MI*),

on the <u>same</u> ultrathin <u>homogeneous</u> a-Pb films

Making homogeneous films

All these predictions required experimental verification, and my friend and University mate, Nikolay Zavaritskii, started to measure the critical field of thin films. Theory and experiment fitted perfectly, including the change of the nature of the transition: first order at larger thicknesses and second order at smaller ones. Everything seemed OK but Alexander Shalnikov, the boss of Zavaritskii was not satisfied. He said that the films used by Zavaritskii were bad, since they were prepared at room temperature. The atoms of the metal, evaporated on a glass substrate, could agglomerate, and therefore the film, actually consisted of small droplets. In order to avoid that, Shalnikov recommended to maintain the glass substrate at helium temperature during evaporation and until the measurements were finished. Then every metal atom hitting the surface would stick to its place, and the film would be homogeneous.

A. Abrikosov, *Nobel Lecture*, 2003

Low temperature quench-condensation

Making ultrathin homogeneous films

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
(Received 22 July 1968; revised manuscript received 1 July 1969)

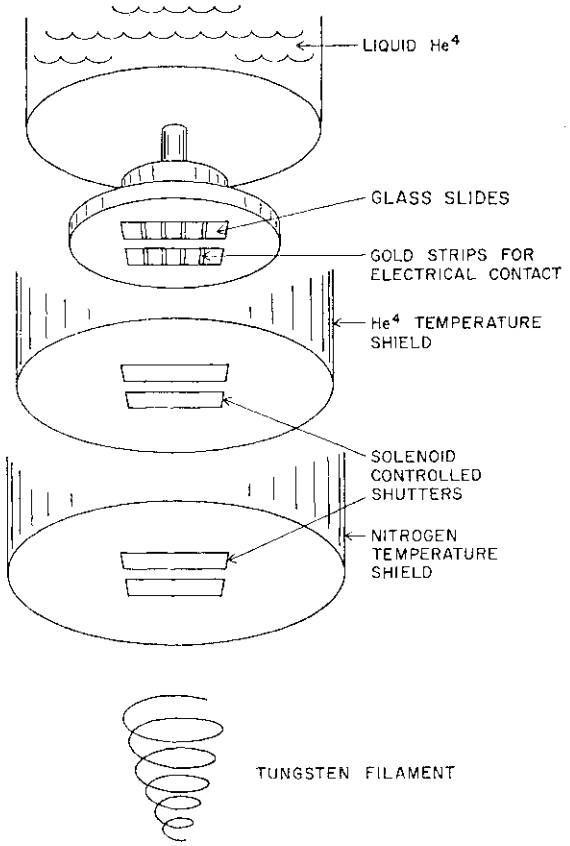
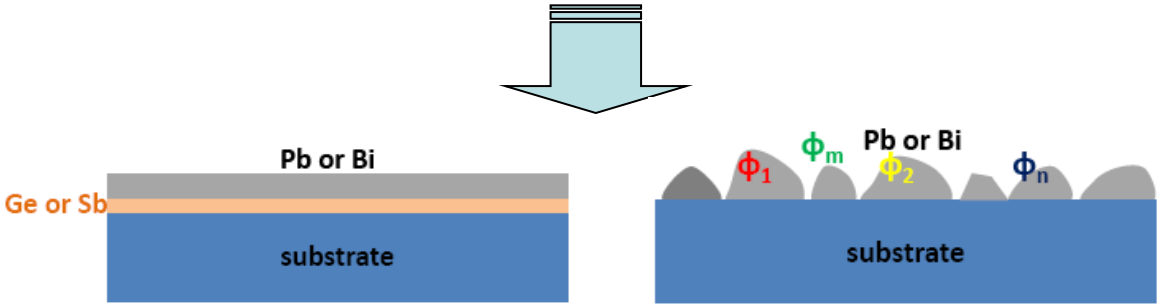


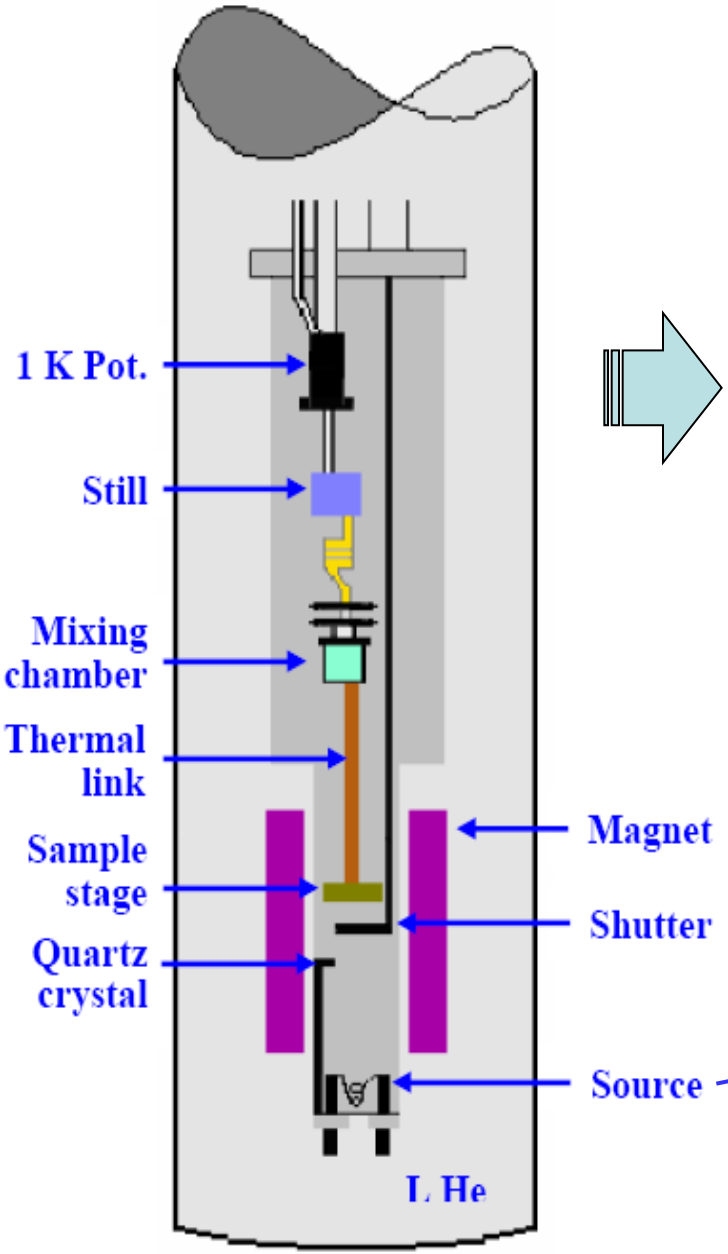
FIG. 3. General experimental setup for evaporating samples.

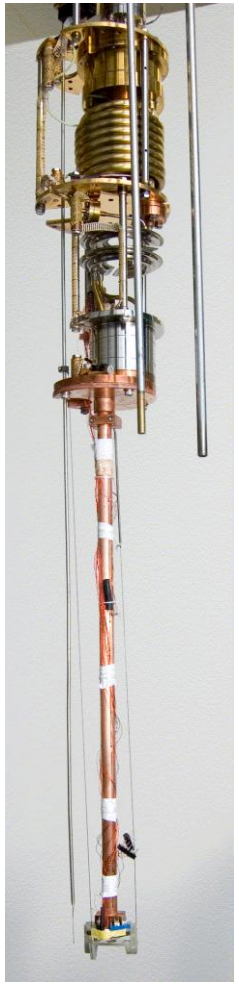
discussed. It is clear that films deposited on previously deposited SiO, Ge, and Al_2O_3 can be made much thinner than films deposited on LiF or glass, immediately showing the important effect of the substrate. In Fig. 5,

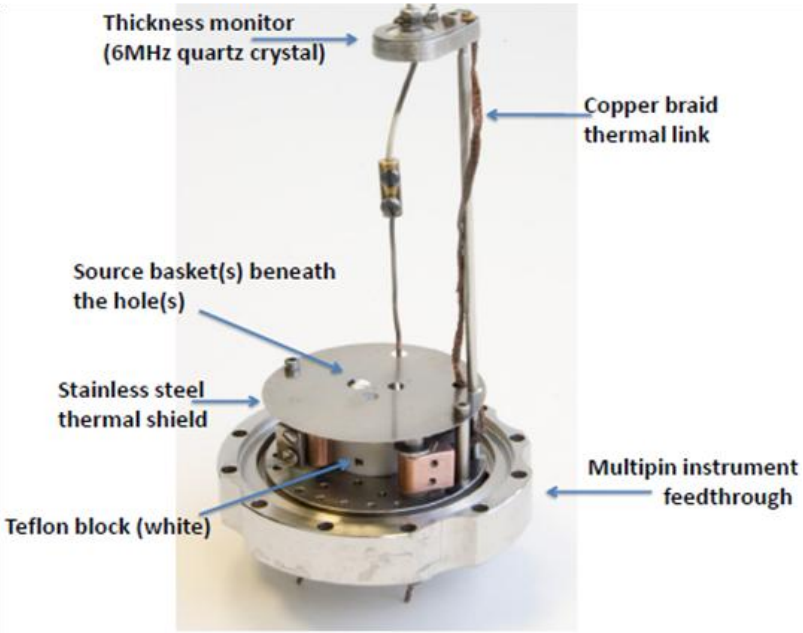


Low temperature quench-condensation with a buffer layer

Experimental Setup



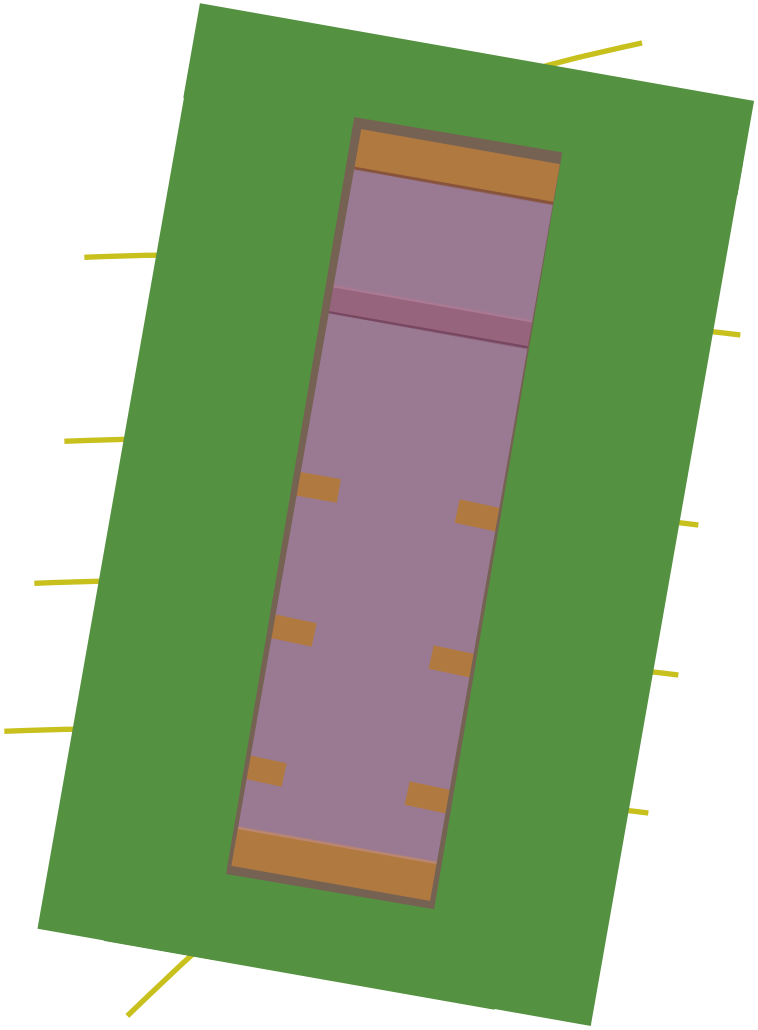




in situ **ultrathin film growth and measurement**

Experimental Procedure

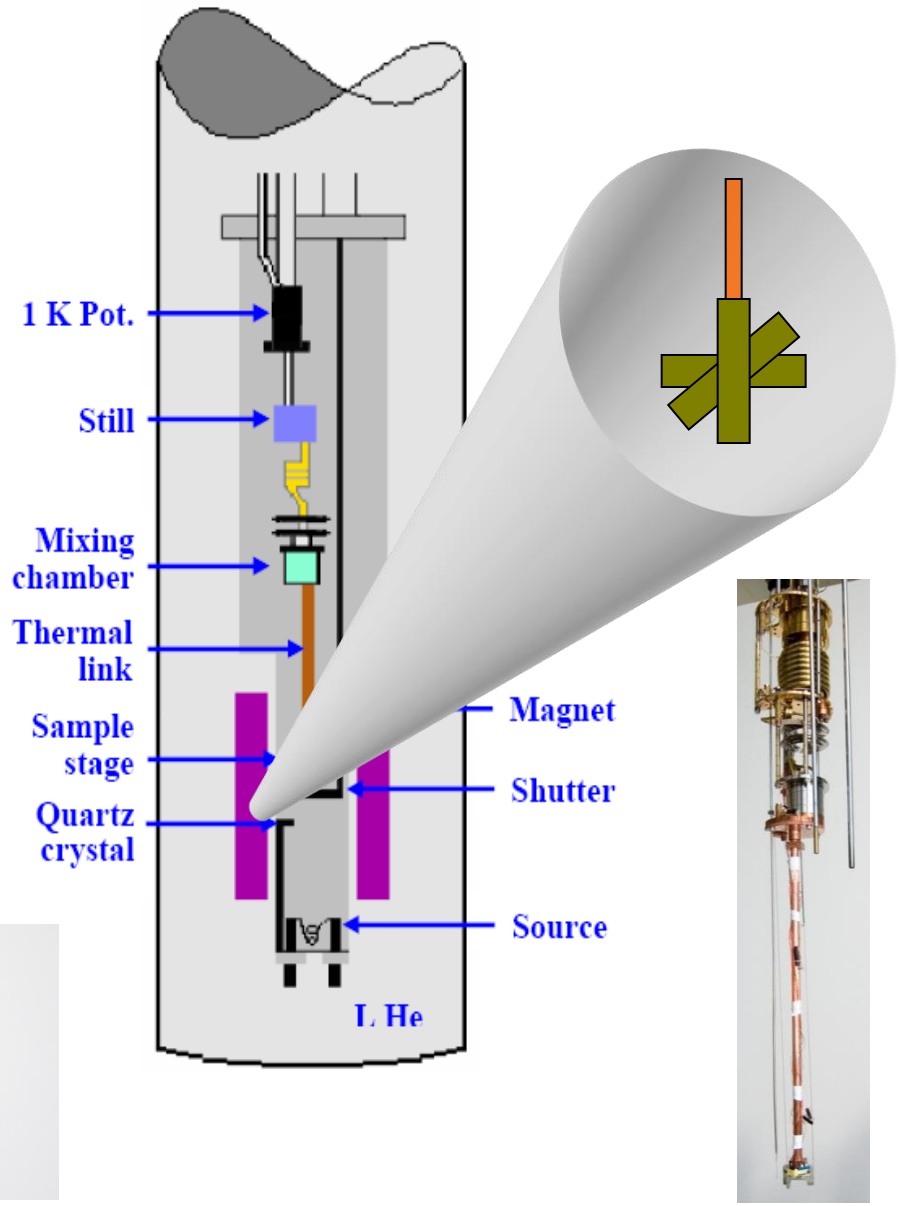
- **Mount a Si wafer on to the Cu sample stage**
- **Deposit ~80 Å Cr/Au contacts through a shadow mask (*ex situ*)**
- **In solder Au wires to contact pads**
- **Deposit 60 Å Al cross stripe**
- **Replace cross stripe mask with Pb shadow mask**
- **Load the dilution unit into cryostat and cool the system**
- **Below 5 K, deposit Sb and Pb**

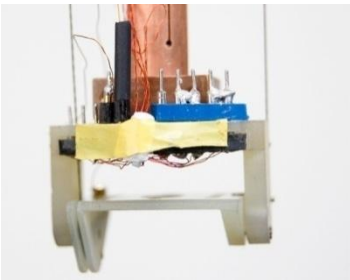


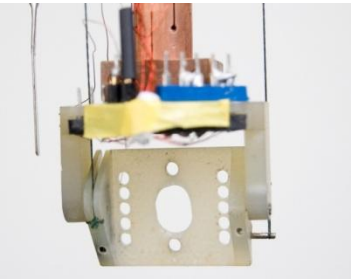
Experimental Setup

Several *in situ* capabilities are crucial:

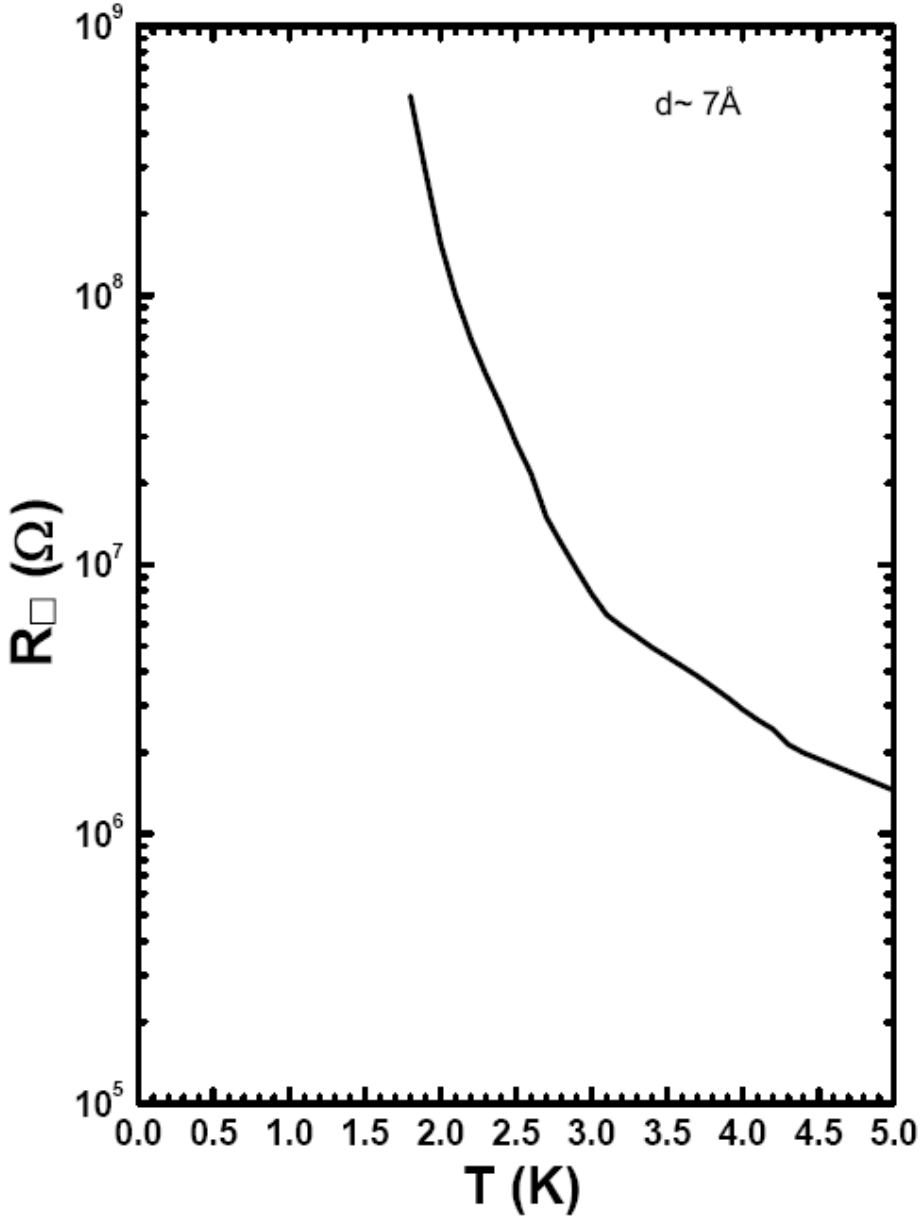
- ***In situ* film growth ➔**
  - Studies of ultrathin films
  - Examination of evolution with film thickness
- ***In situ* sample rotation ➔**
  - Access to both field orientations
  - Fine adjustment of field alignment
- ***In situ* magnetic impurity deposition ➔**
  - Unambiguous determination of effect of paramagnetic impurities



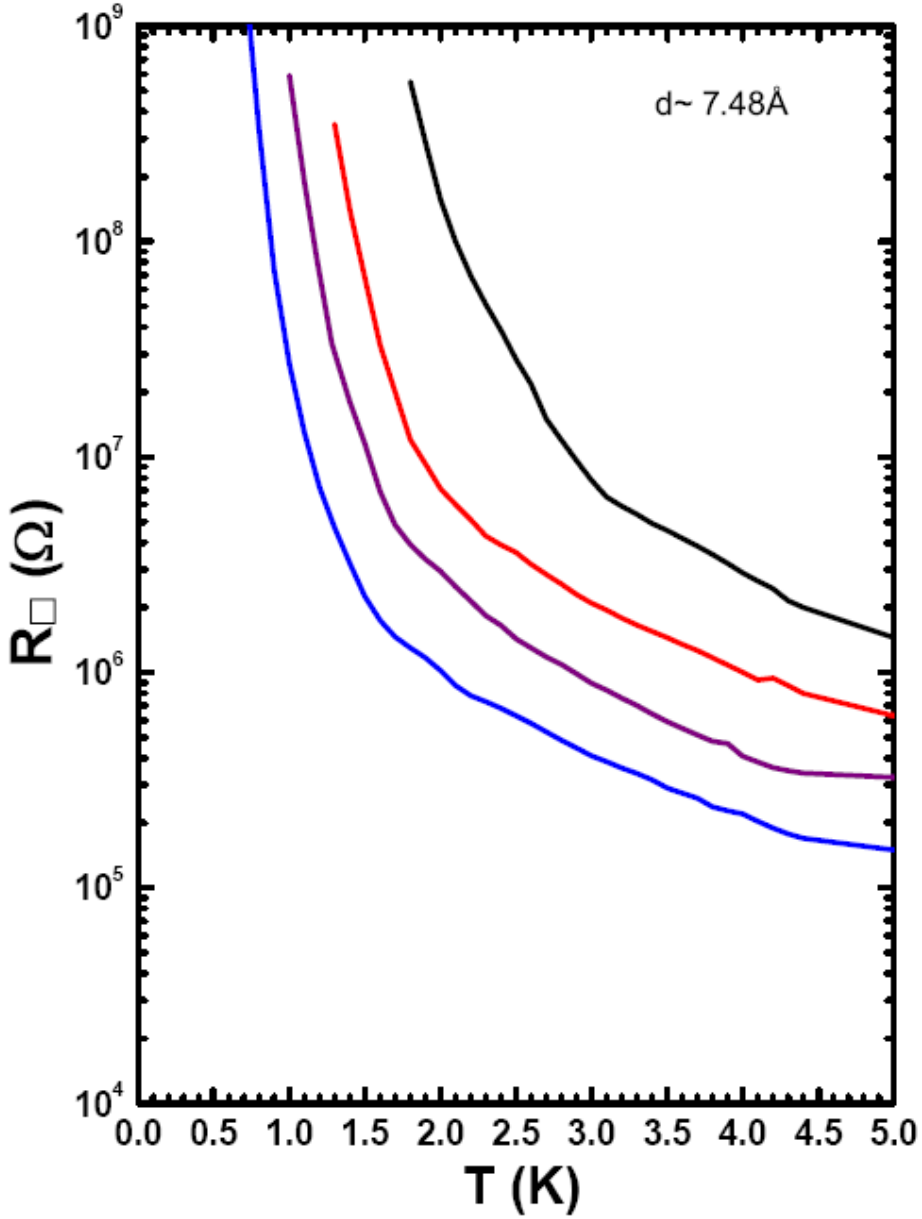




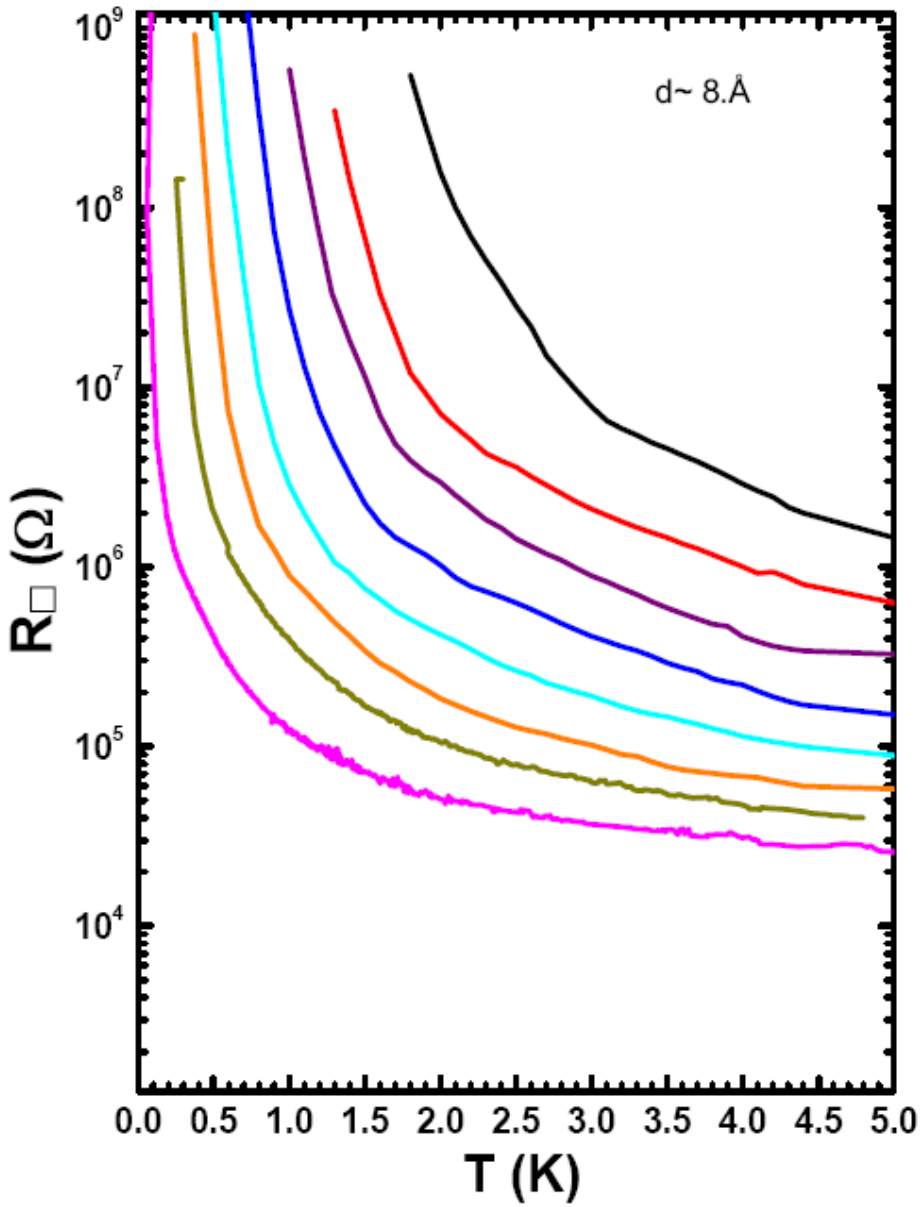
d-tuned SIT


d~ 7Å
10^9
10^8
10^7
10^6
10^5
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

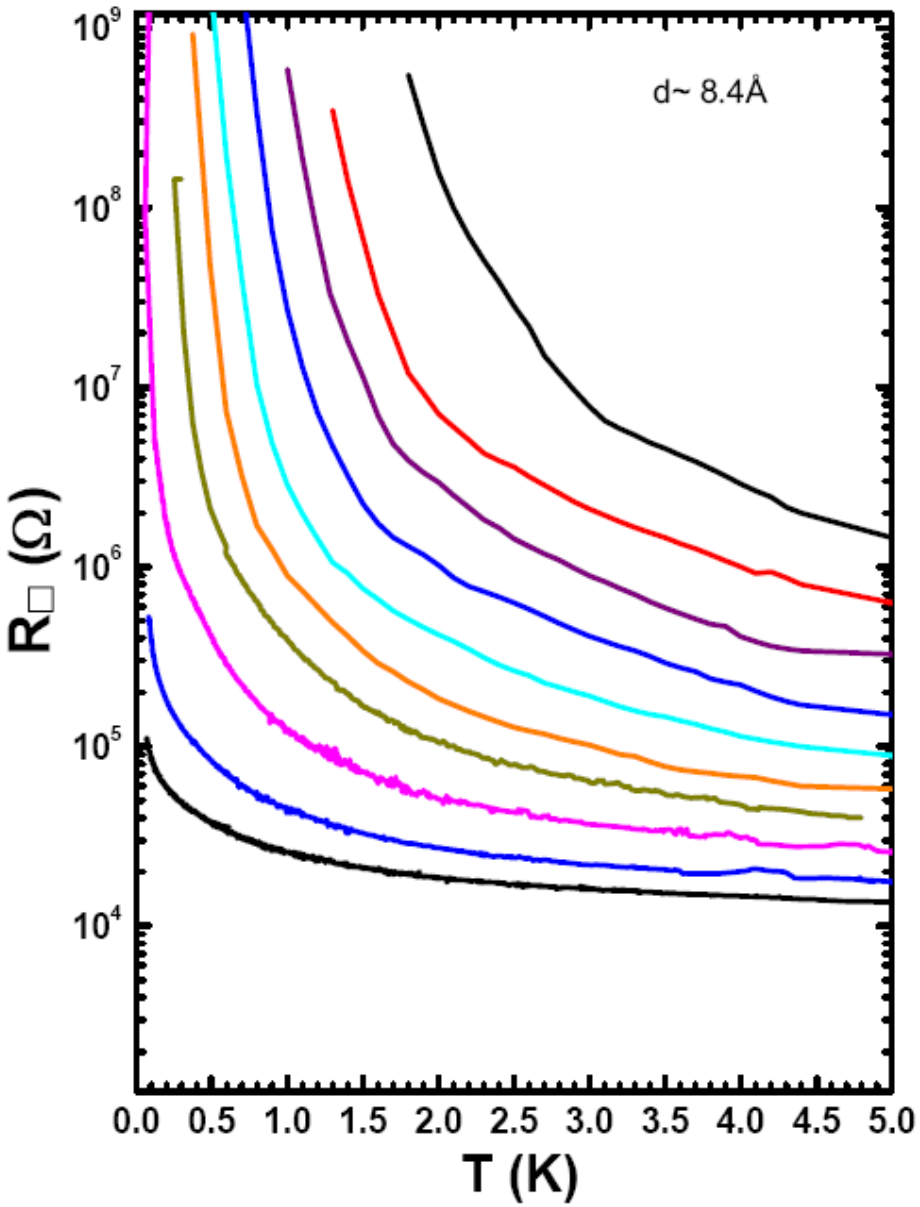
d-tuned SIT


d~ 7.48Å
10^9
10^8
10^7
10^6
10^5
10^4
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

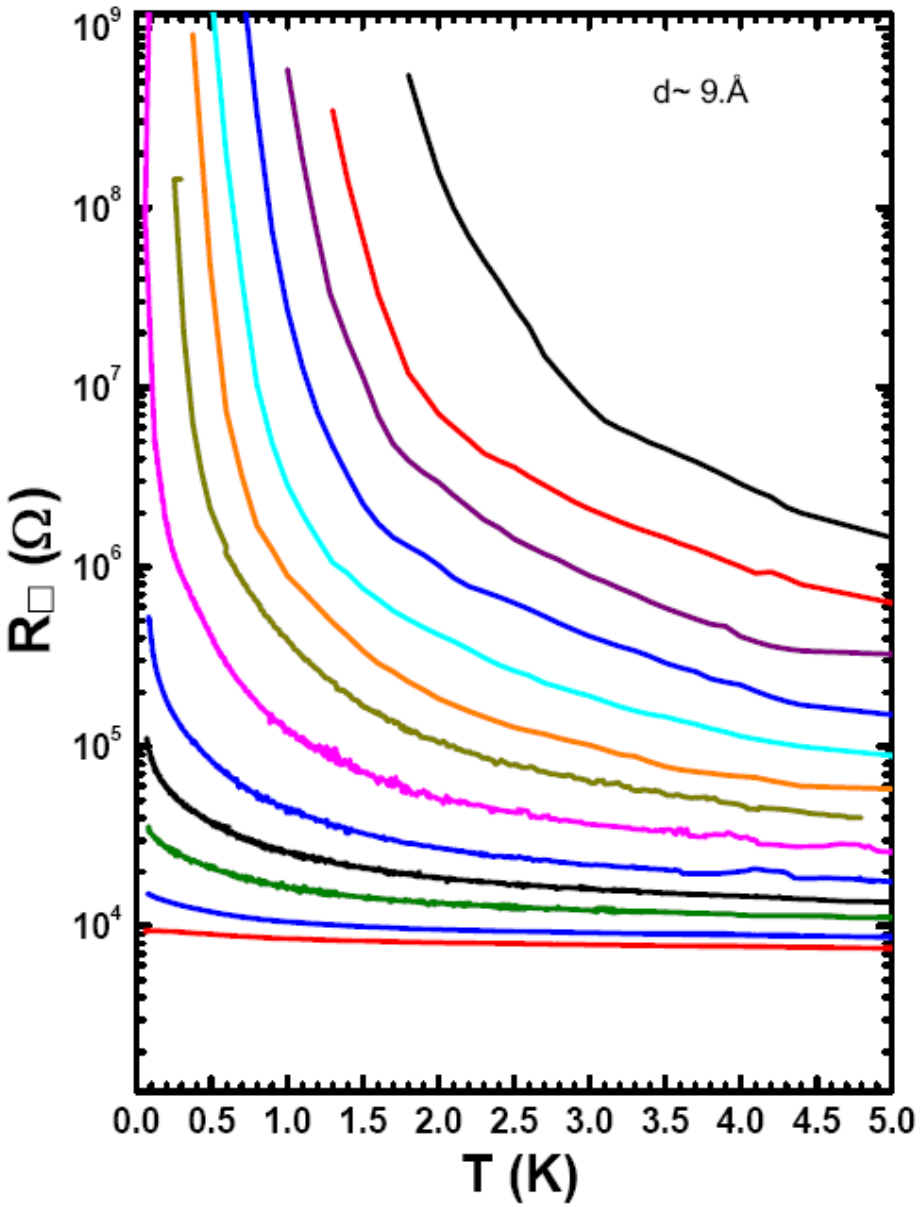
d-tuned SIT


d~ 8.Å
10^9
10^8
10^7
10^6
10^5
10^4
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

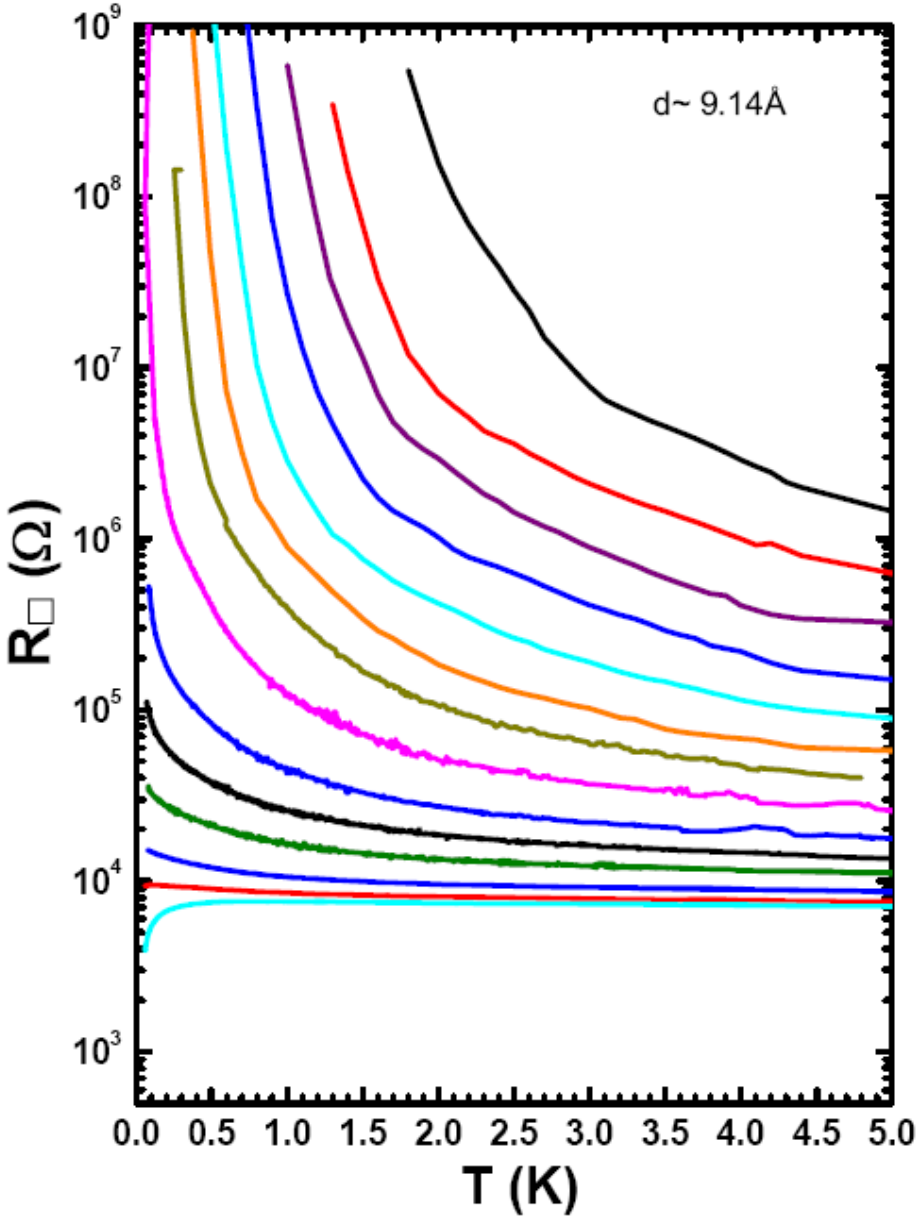
d-tuned SIT


d~ 8.4Å
10^9
10^8
10^7
10^6
10^5
10^4
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

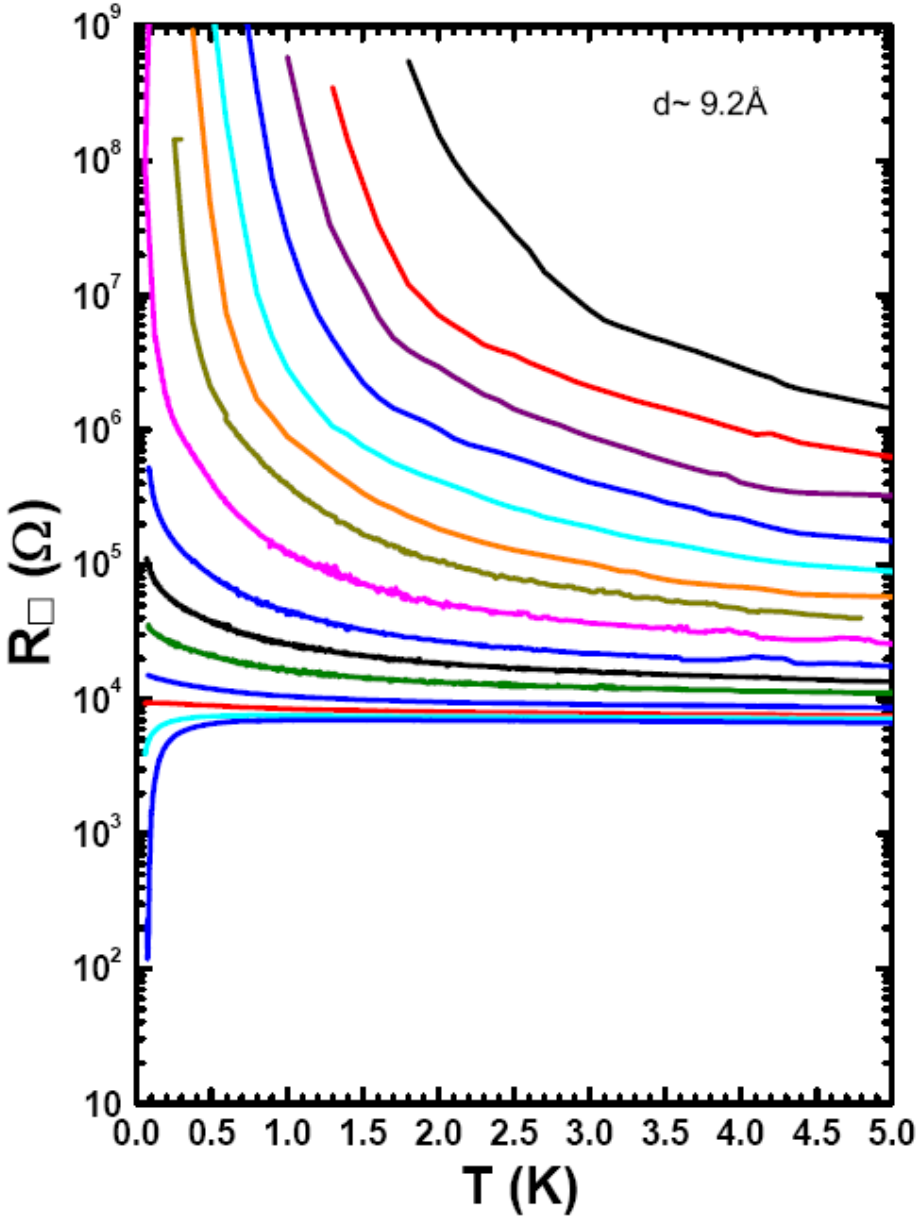
d-tuned SIT


d~ 9.Å
10^9
10^8
10^7
10^6
10^5
10^4
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

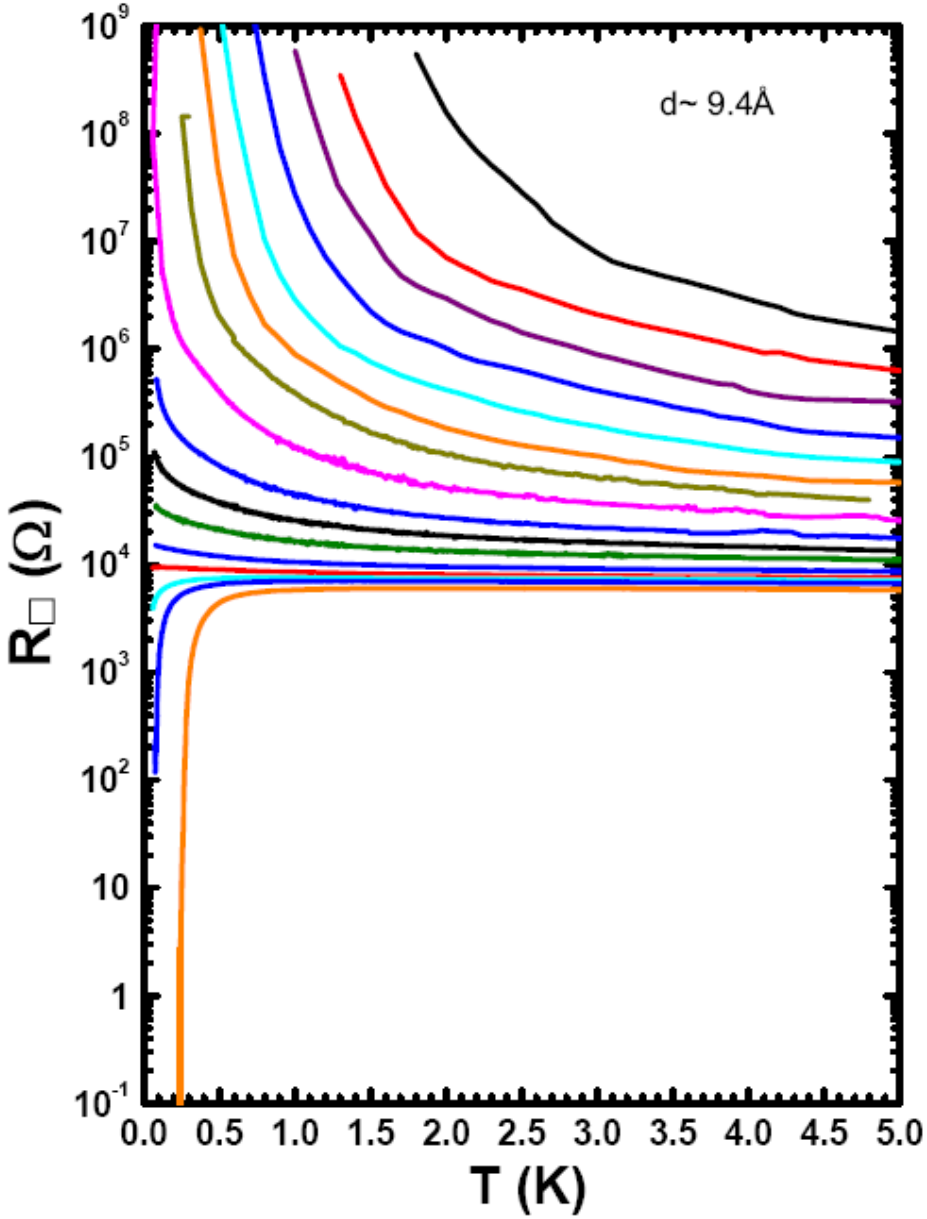
d-tuned SIT


d~ 9.14Å
10^9
10^8
10^7
10^6
10^5
10^4
10^3
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

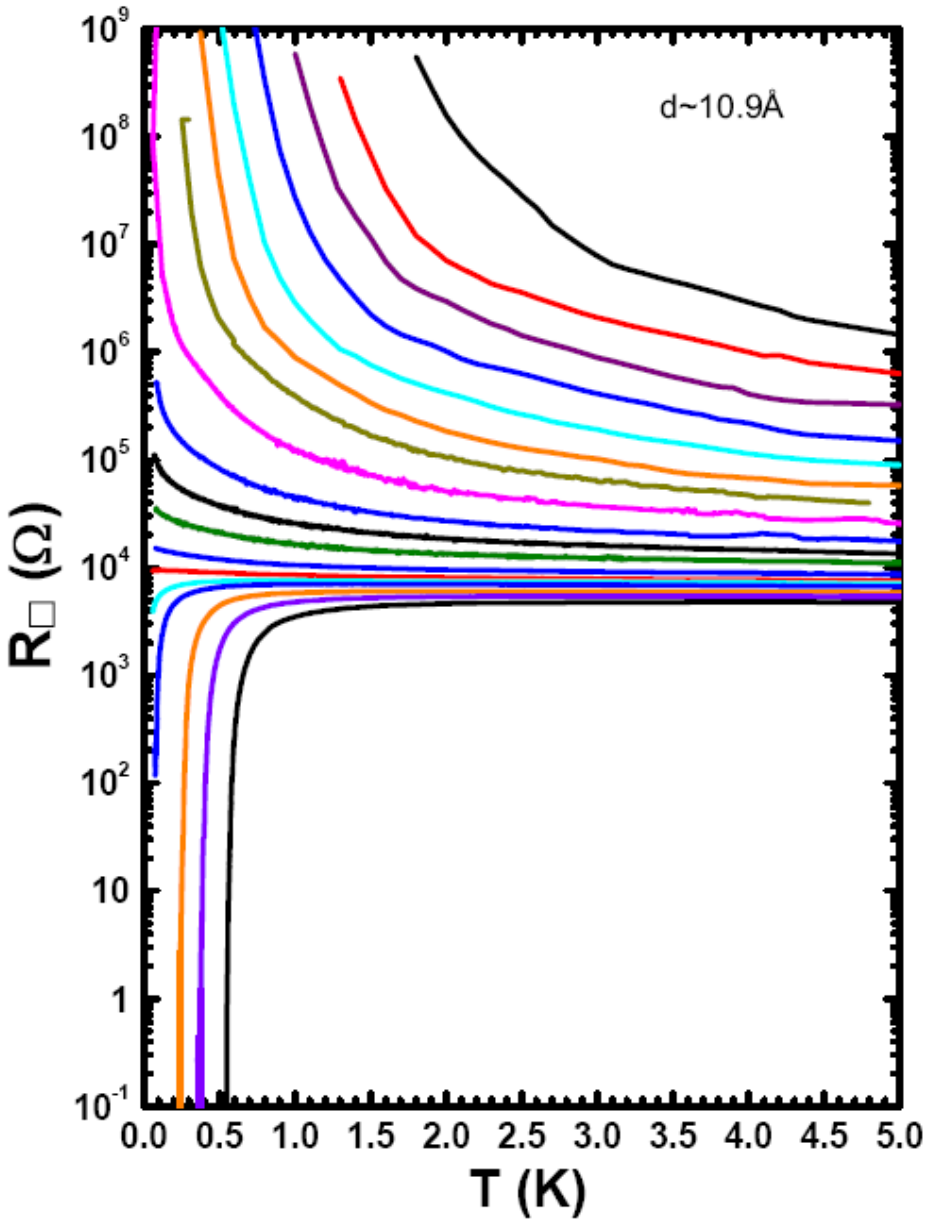
d-tuned SIT


d~ 9.2Å
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

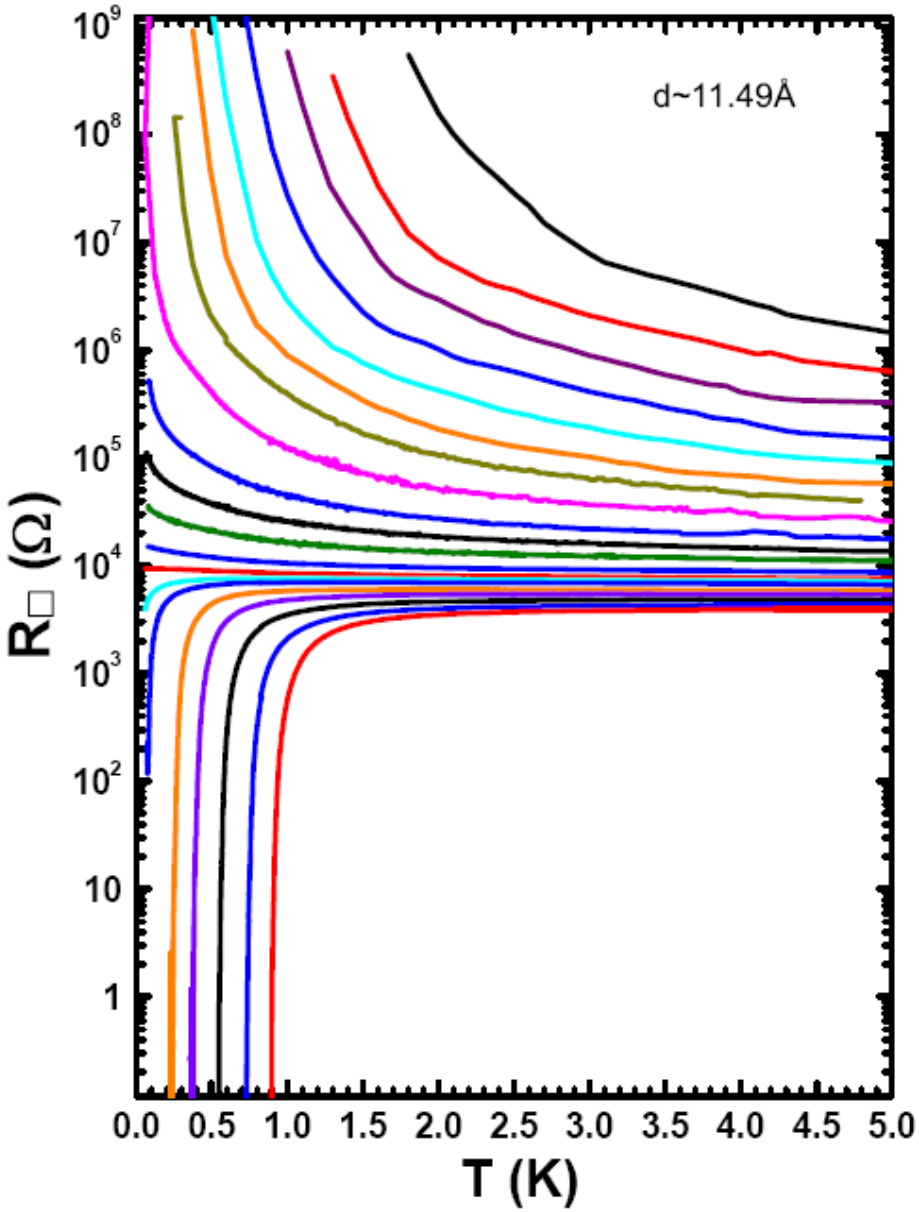
d-tuned SIT


d~ 9.4Å
$R_\square$ (Ω)
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10
1
10^{-1}
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

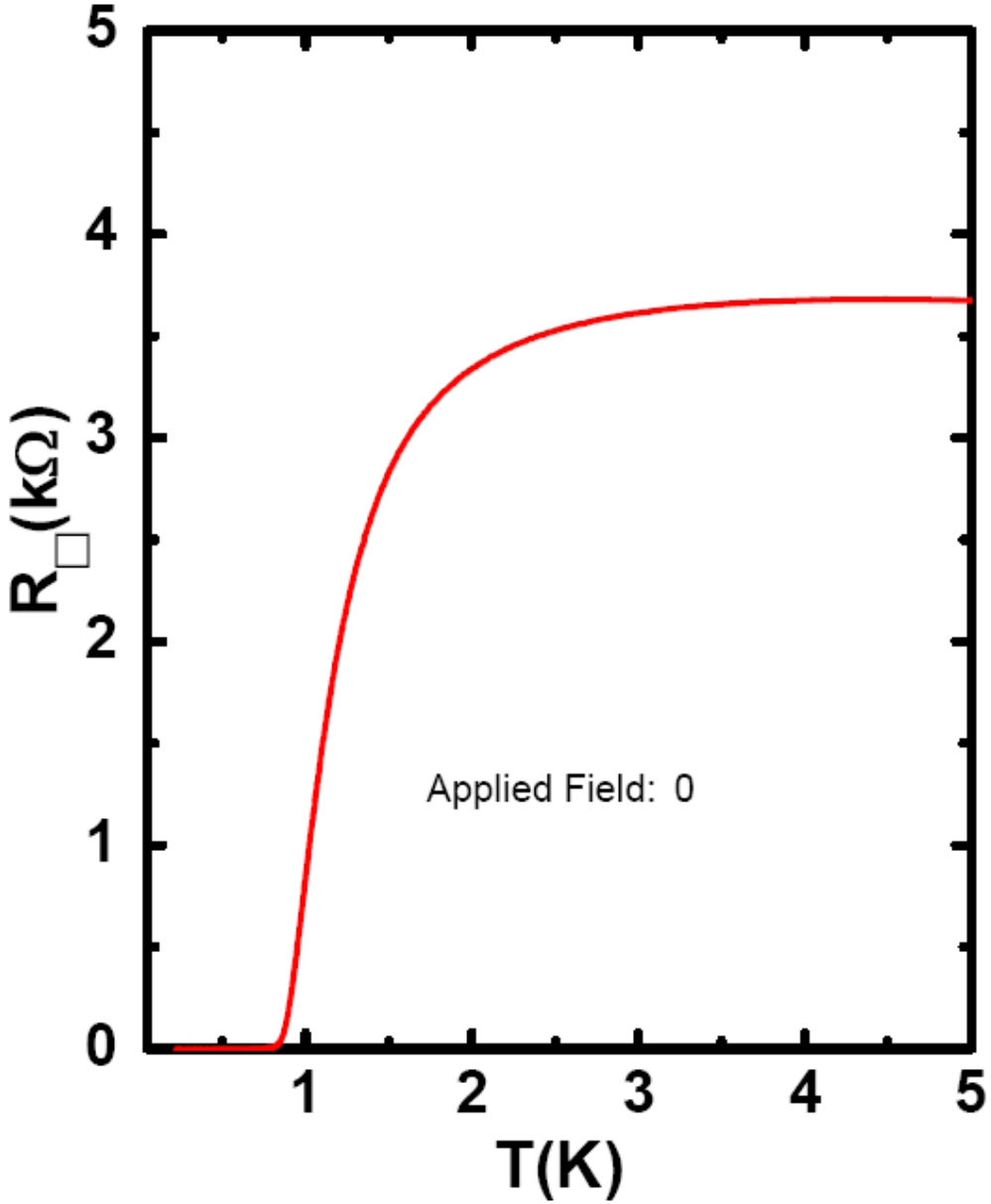
d-tuned SIT


d~10.9Å
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10
1
10^{-1}
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

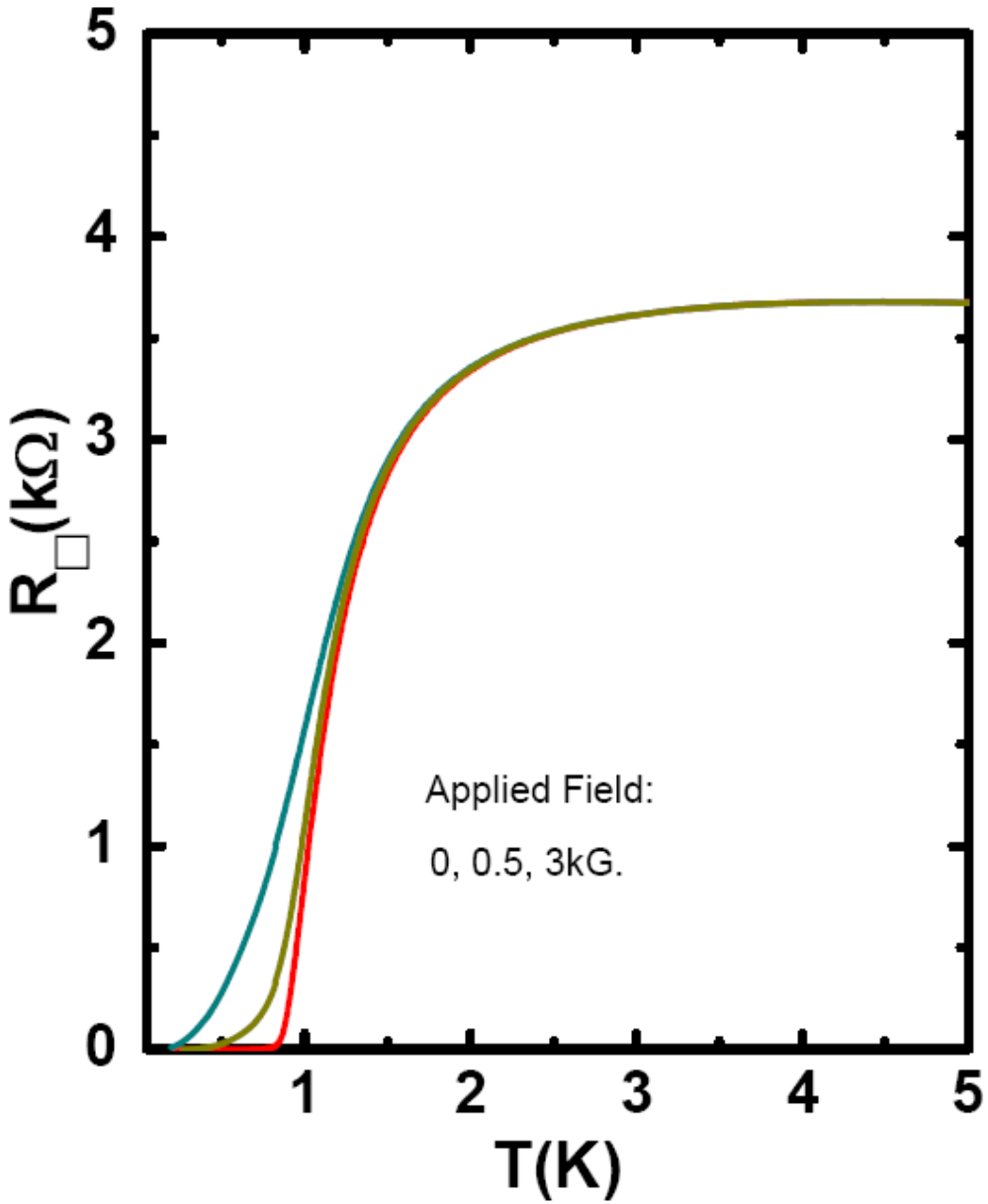
d-tuned SIT


d~11.49Å
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10
1
$R_\square$ (Ω)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
T (K)

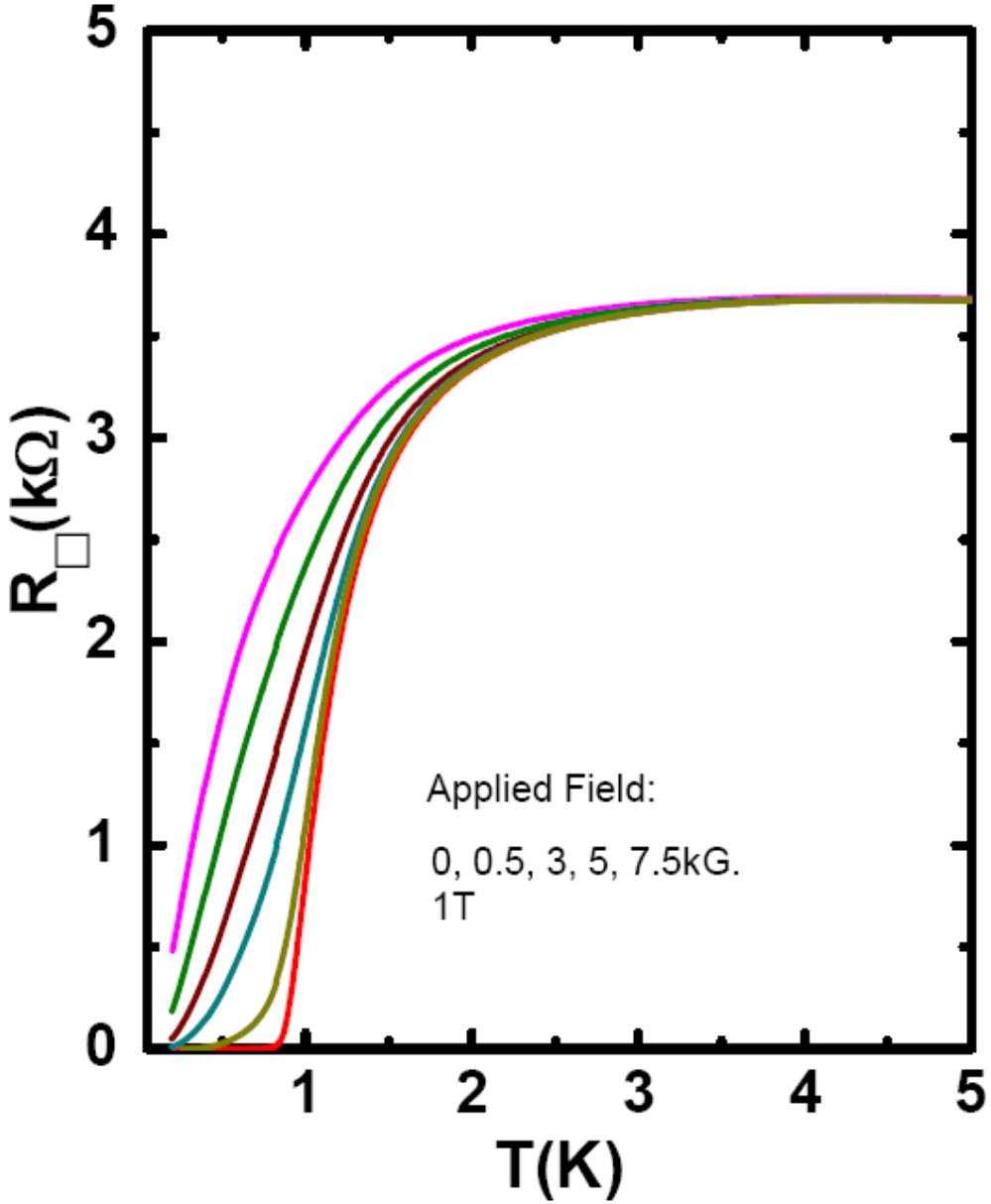
B-tuned SIT



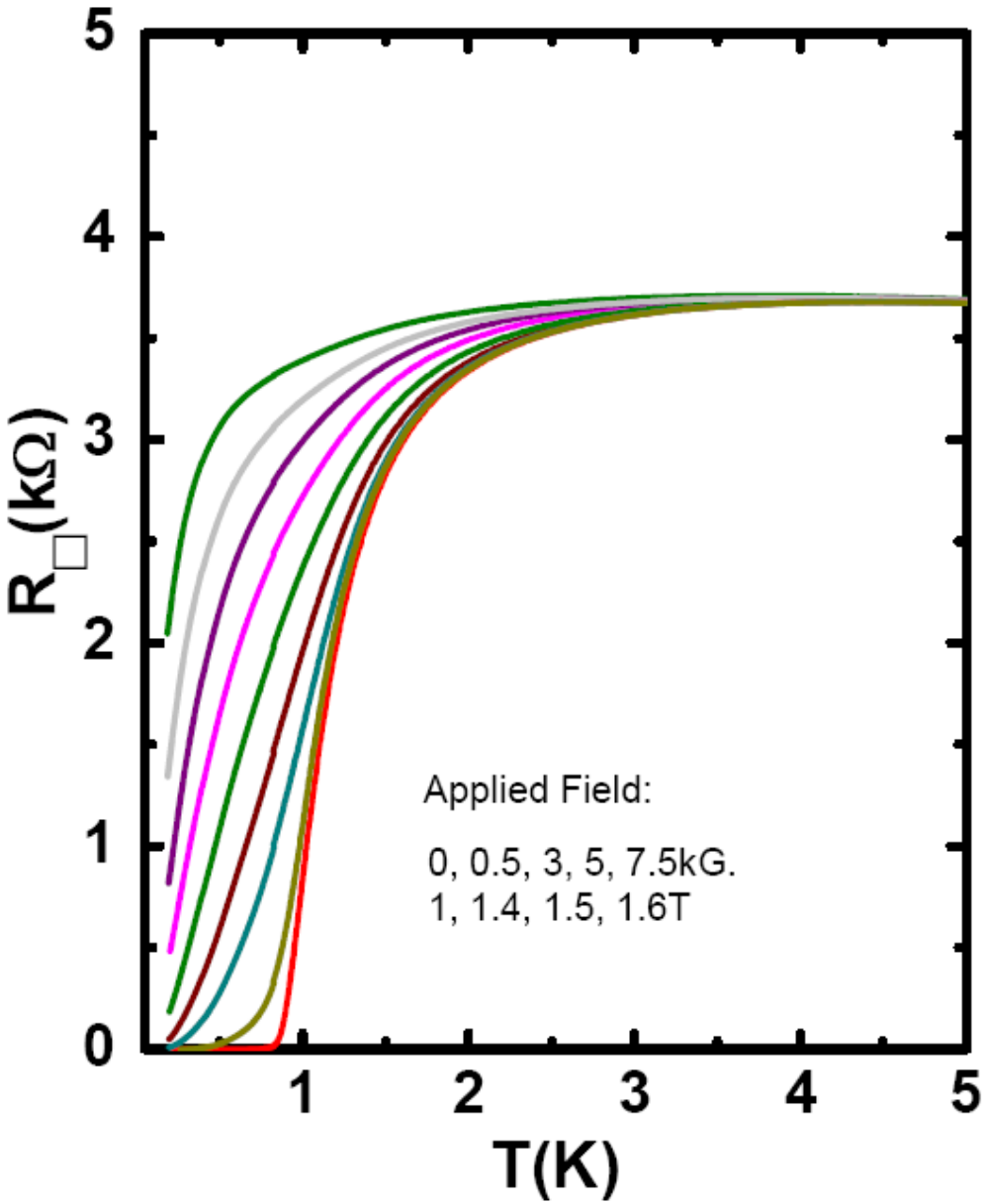
B-tuned SIT



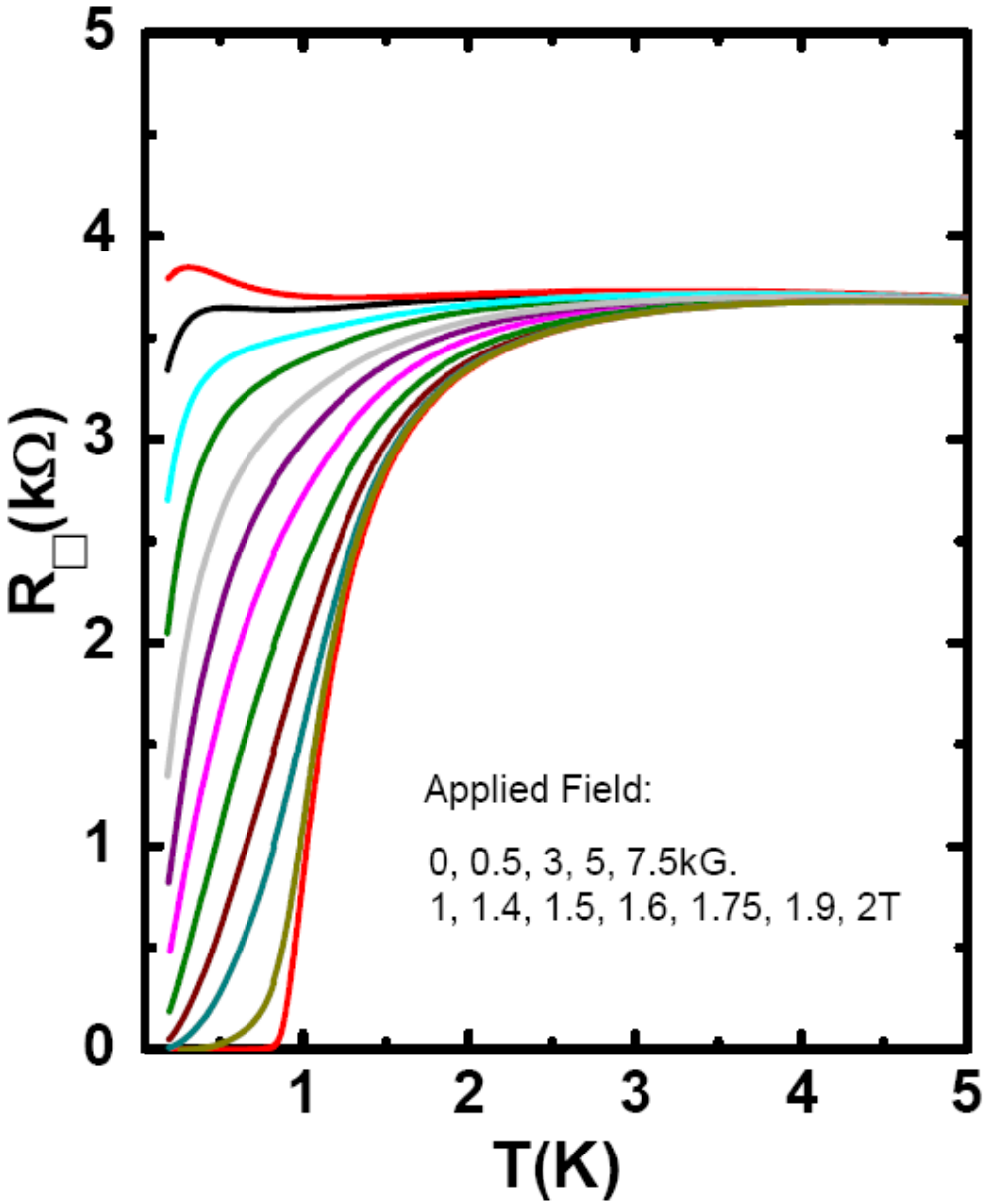
B-tuned SIT



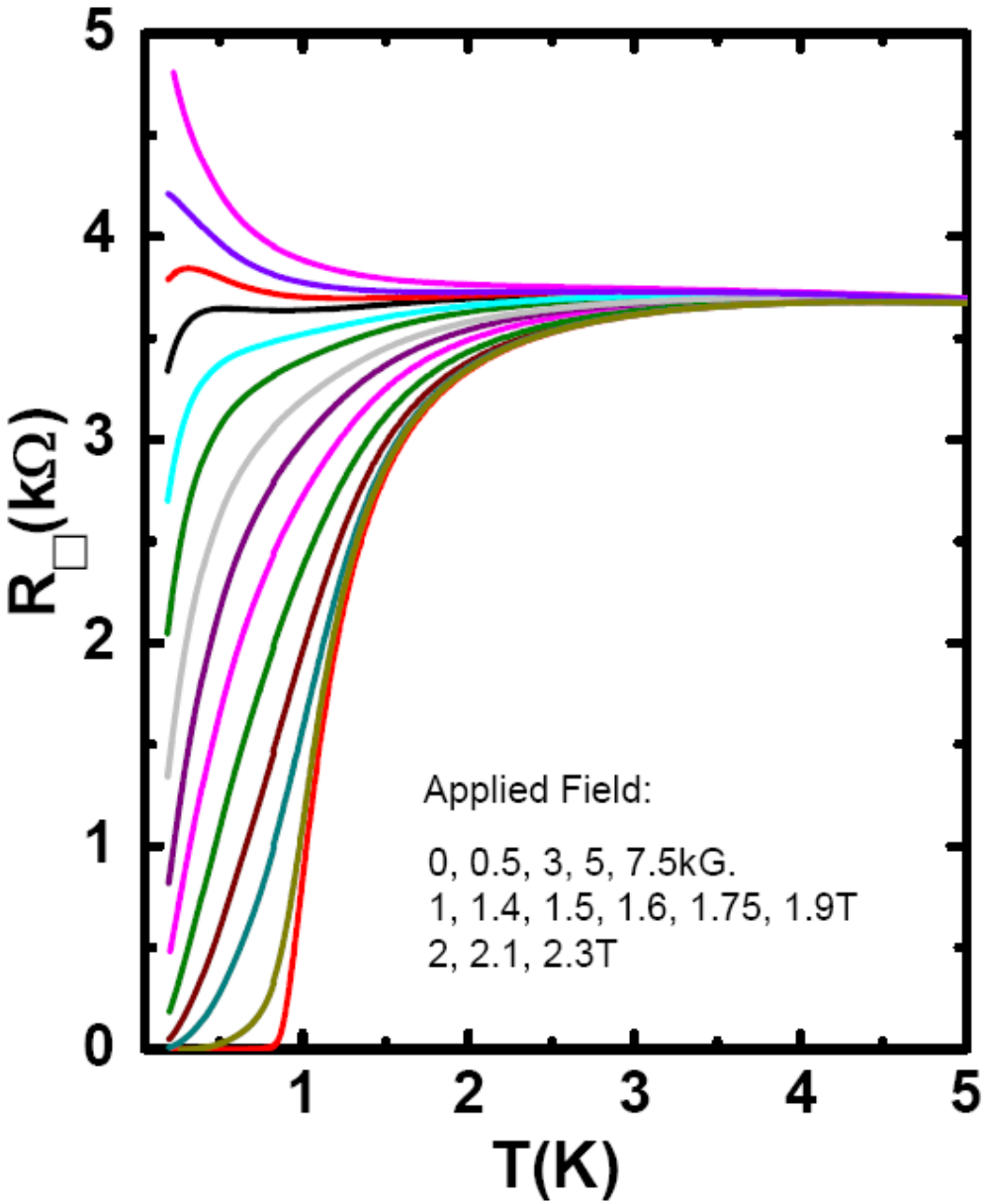
B-tuned SIT


5
4
3
2
1
0
$R_{\square}(k\Omega)$
1
2
3
4
5
T(K)
Applied Field:
0, 0.5, 3, 5, 7.5kG.
1, 1.4, 1.5, 1.6T

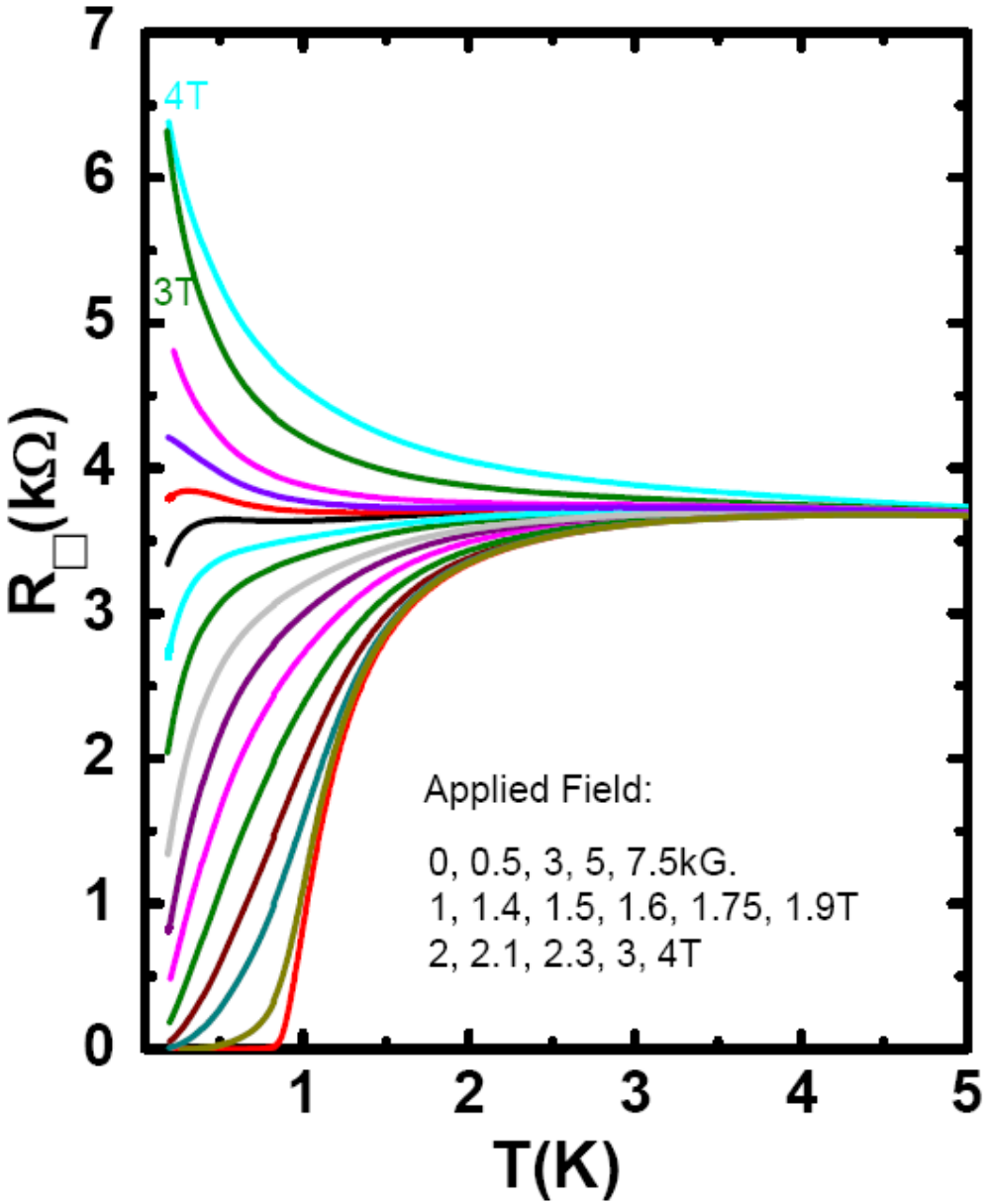
B-tuned SIT


5
4
3
2
1
0
$R_{\square}$(kΩ)
1
2
3
4
5
T(K)
Applied Field:
0, 0.5, 3, 5, 7.5kG.
1, 1.4, 1.5, 1.6, 1.75, 1.9, 2T

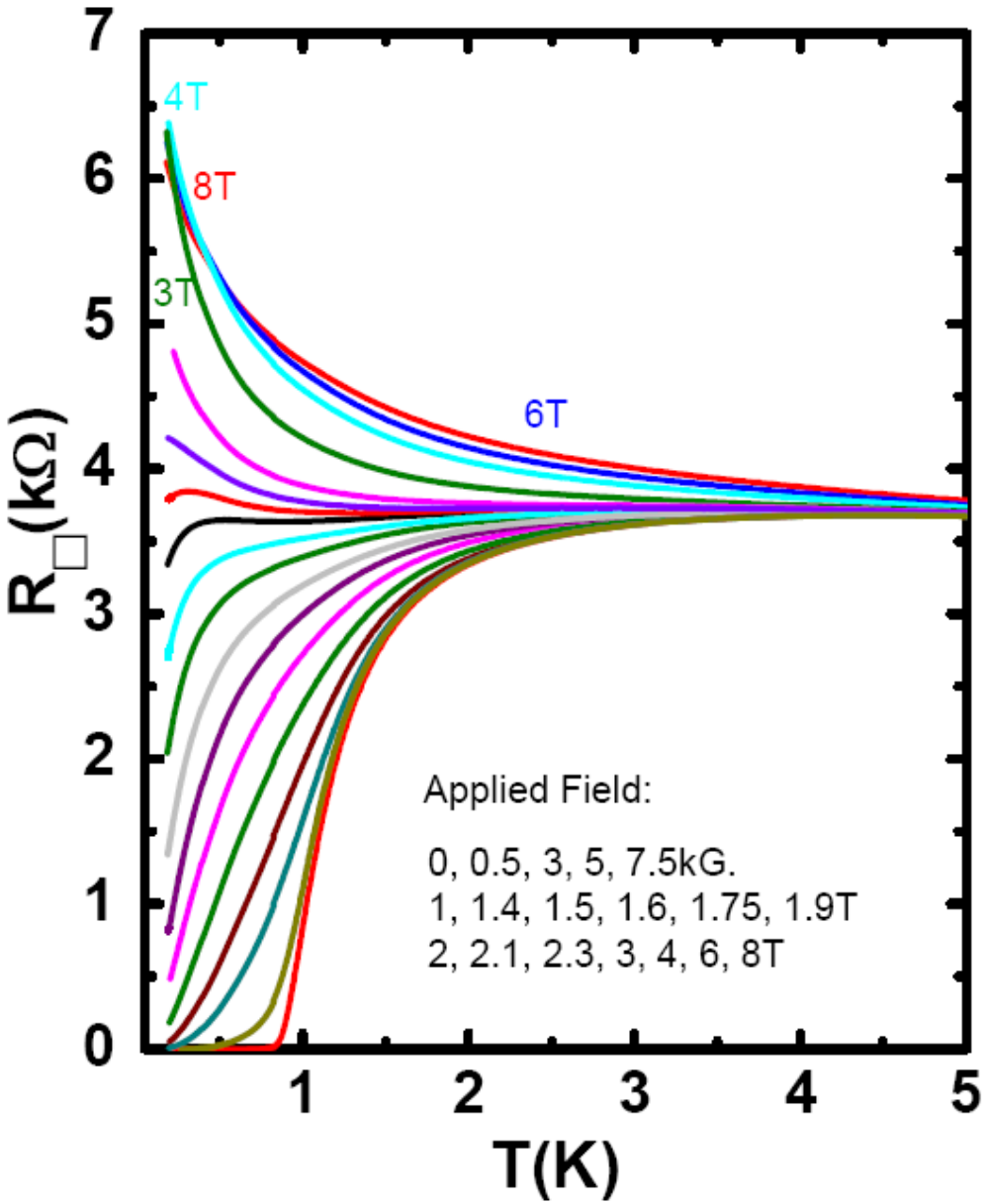
B-tuned SIT


5
4
3
2
1
0
$R_{\square}$(kΩ)
1
2
3
4
5
T(K)
Applied Field:
0, 0.5, 3, 5, 7.5kG.
1, 1.4, 1.5, 1.6, 1.75, 1.9T
2, 2.1, 2.3T

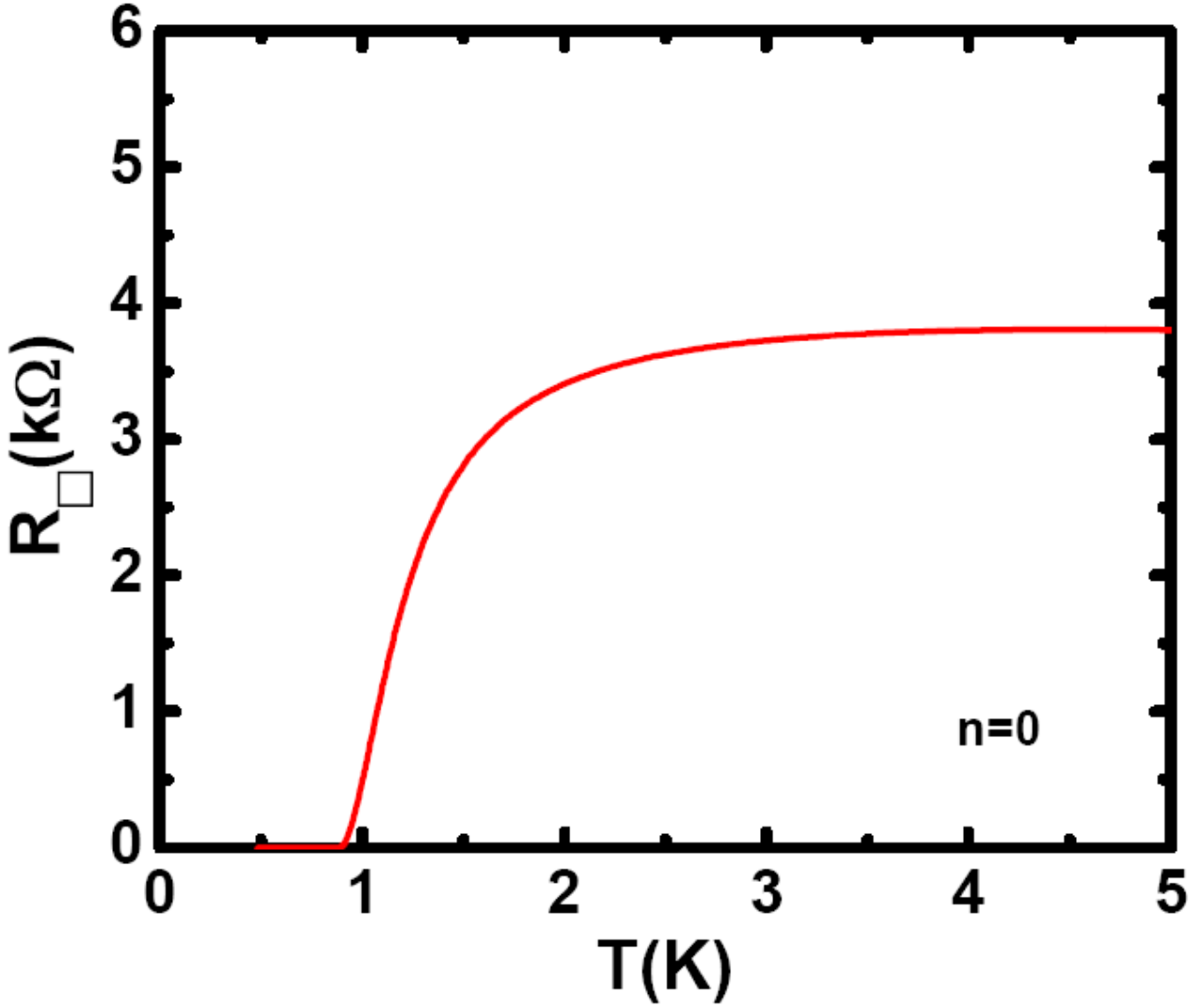
B-tuned SIT


4T
3T
Applied Field:
0, 0.5, 3, 5, 7.5kG.
1, 1.4, 1.5, 1.6, 1.75, 1.9T
2, 2.1, 2.3, 3, 4T
R□(kΩ)
T(K)
0
1
2
3
4
5
6
7
1
2
3
4
5

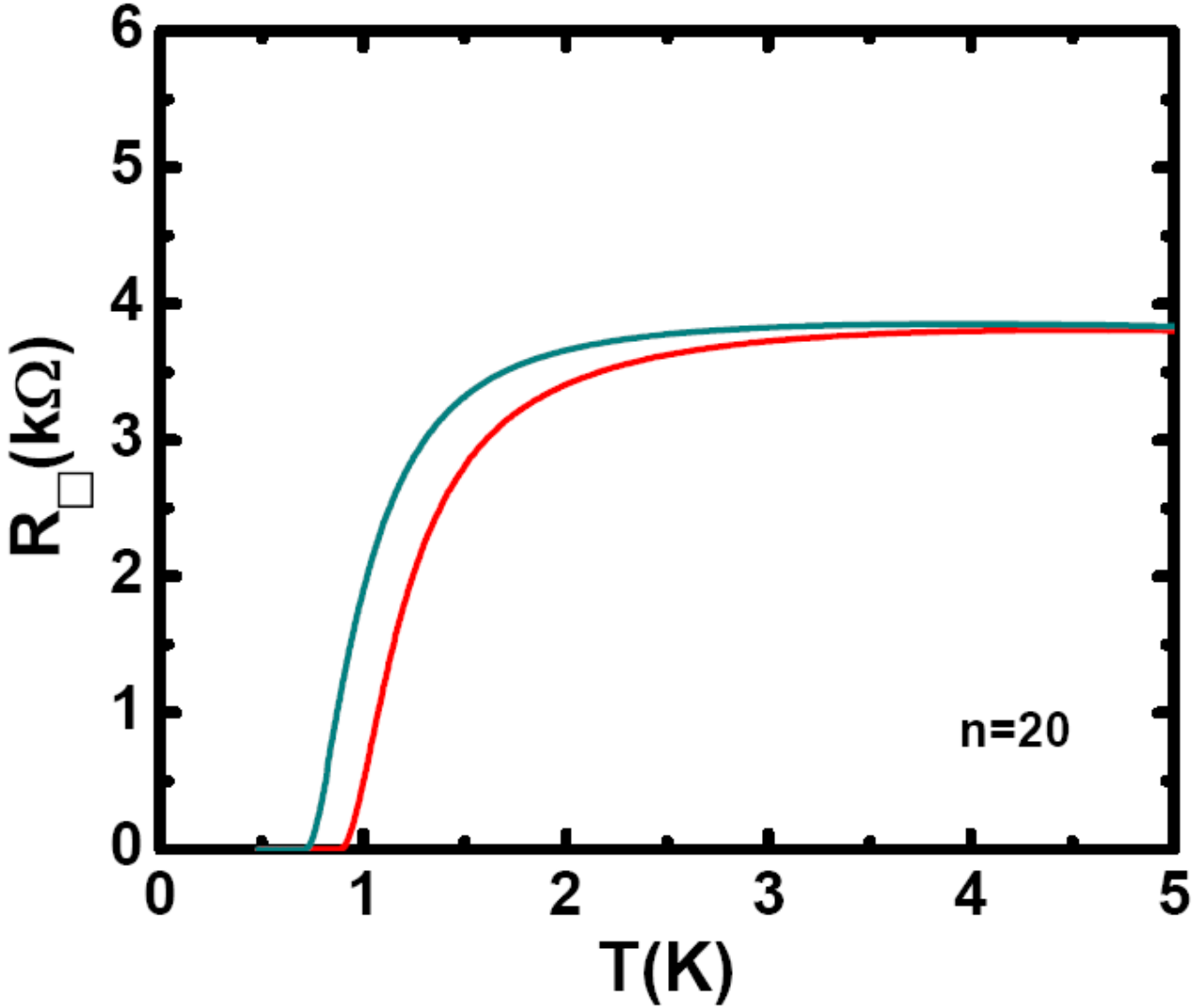
B-tuned SIT


7
6
5
4
3
2
1
0
1
2
3
4
5
T(K)
R□(kΩ)
4T
8T
3T
6T
Applied Field:
0, 0.5, 3, 5, 7.5kG.
1, 1.4, 1.5, 1.6, 1.75, 1.9T
2, 2.1, 2.3, 3, 4, 6, 8T

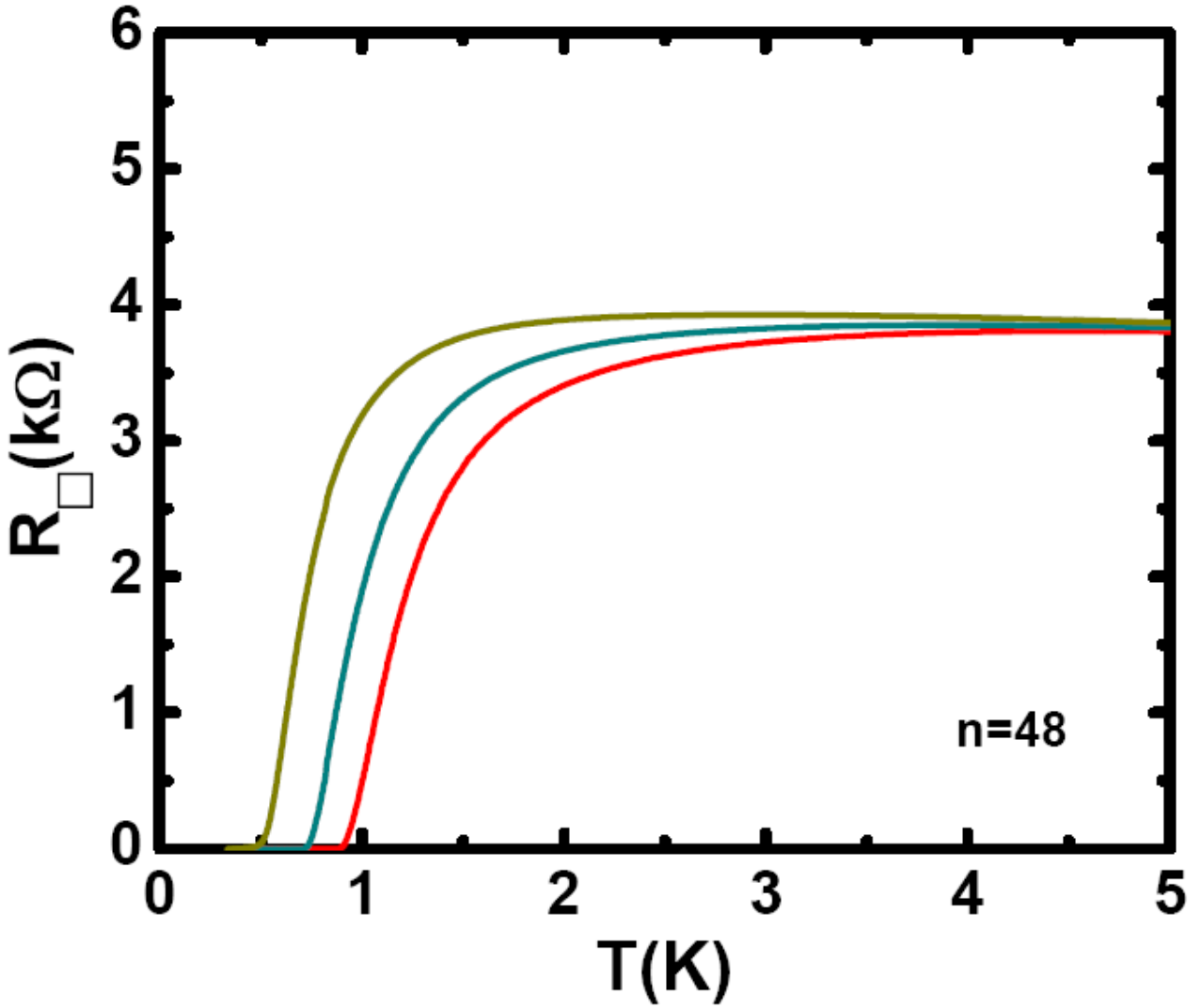
MI-tuned transition



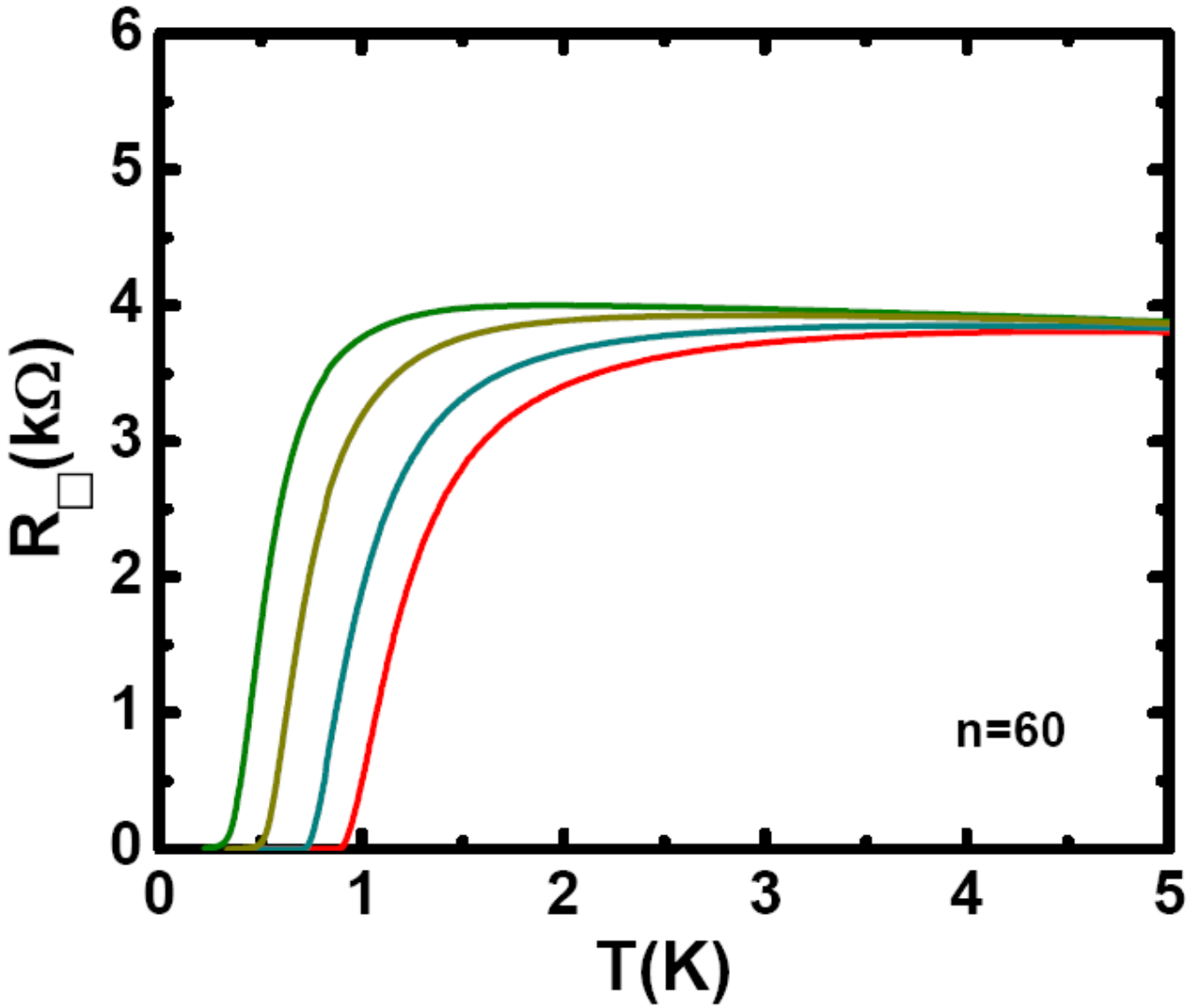
MI-tuned transition



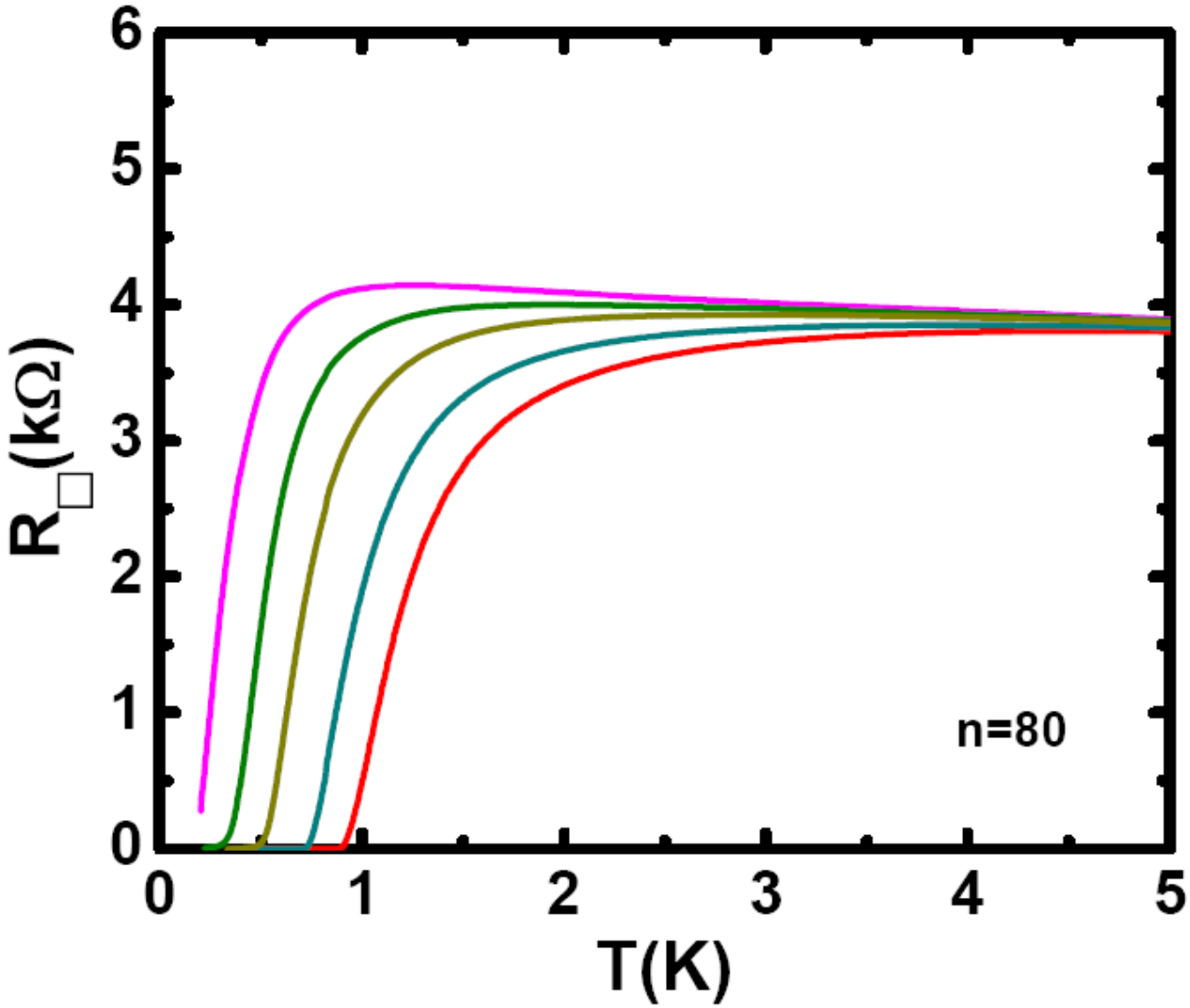
MI-tuned transition



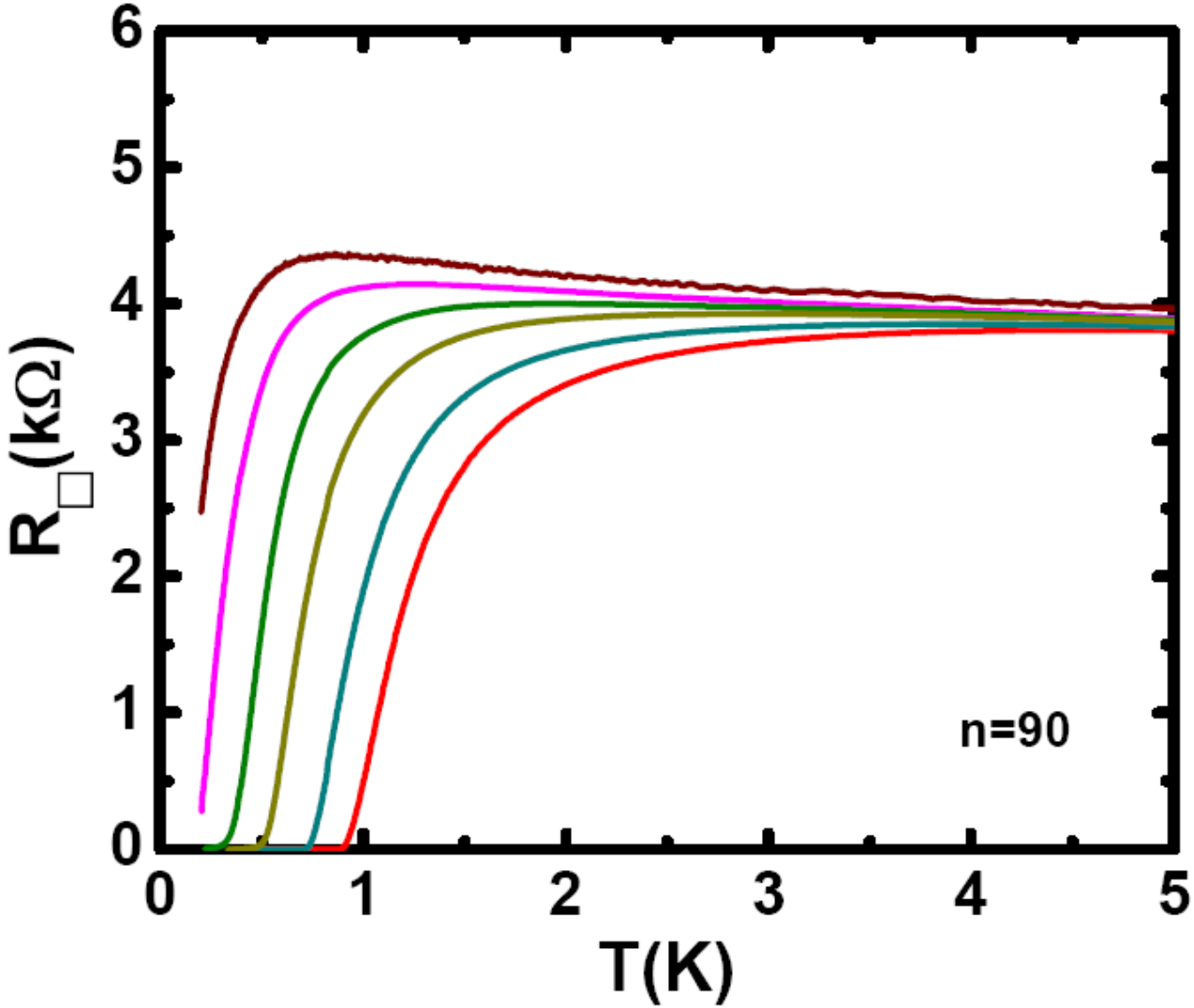
MI-tuned transition



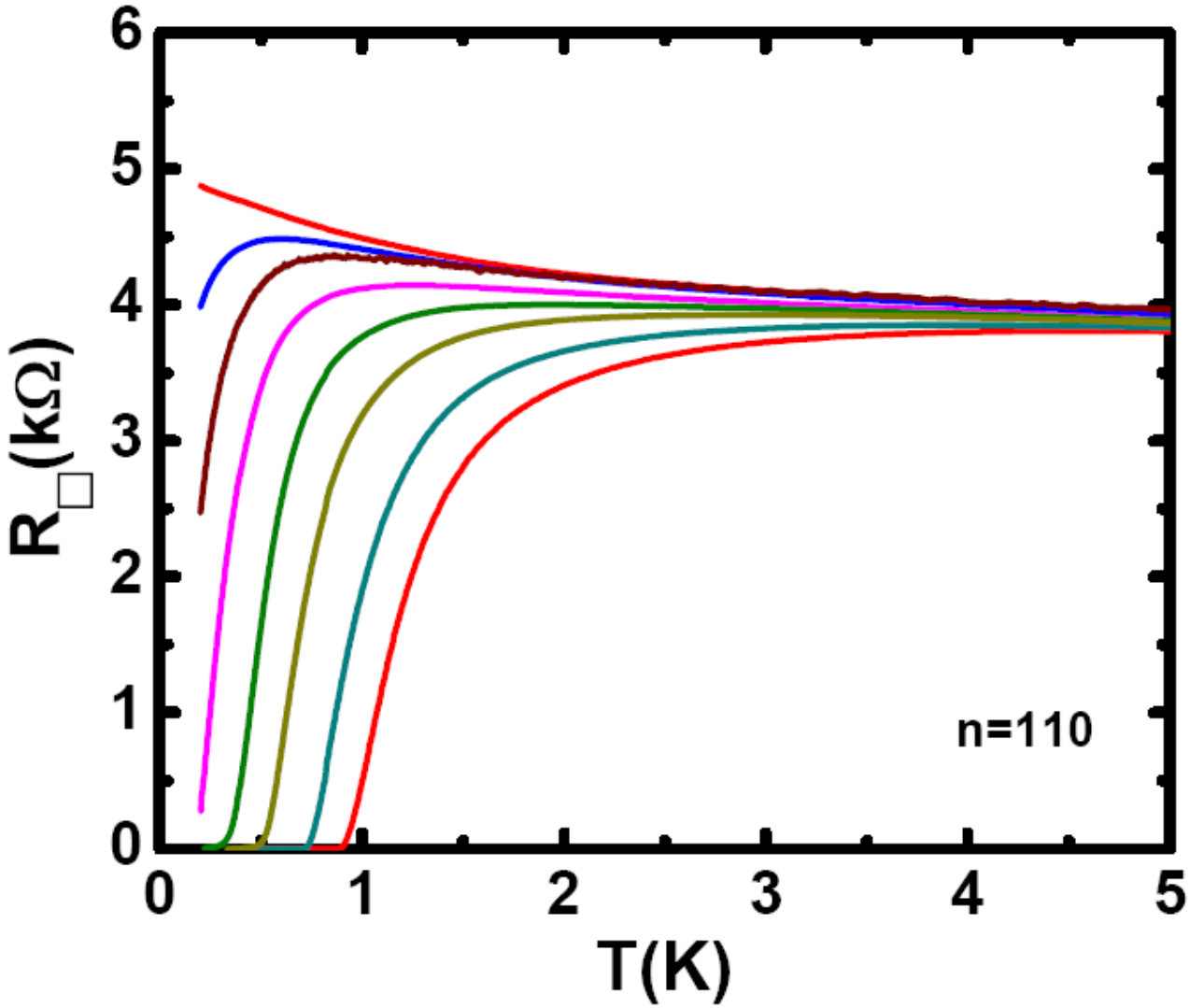
MI-tuned transition



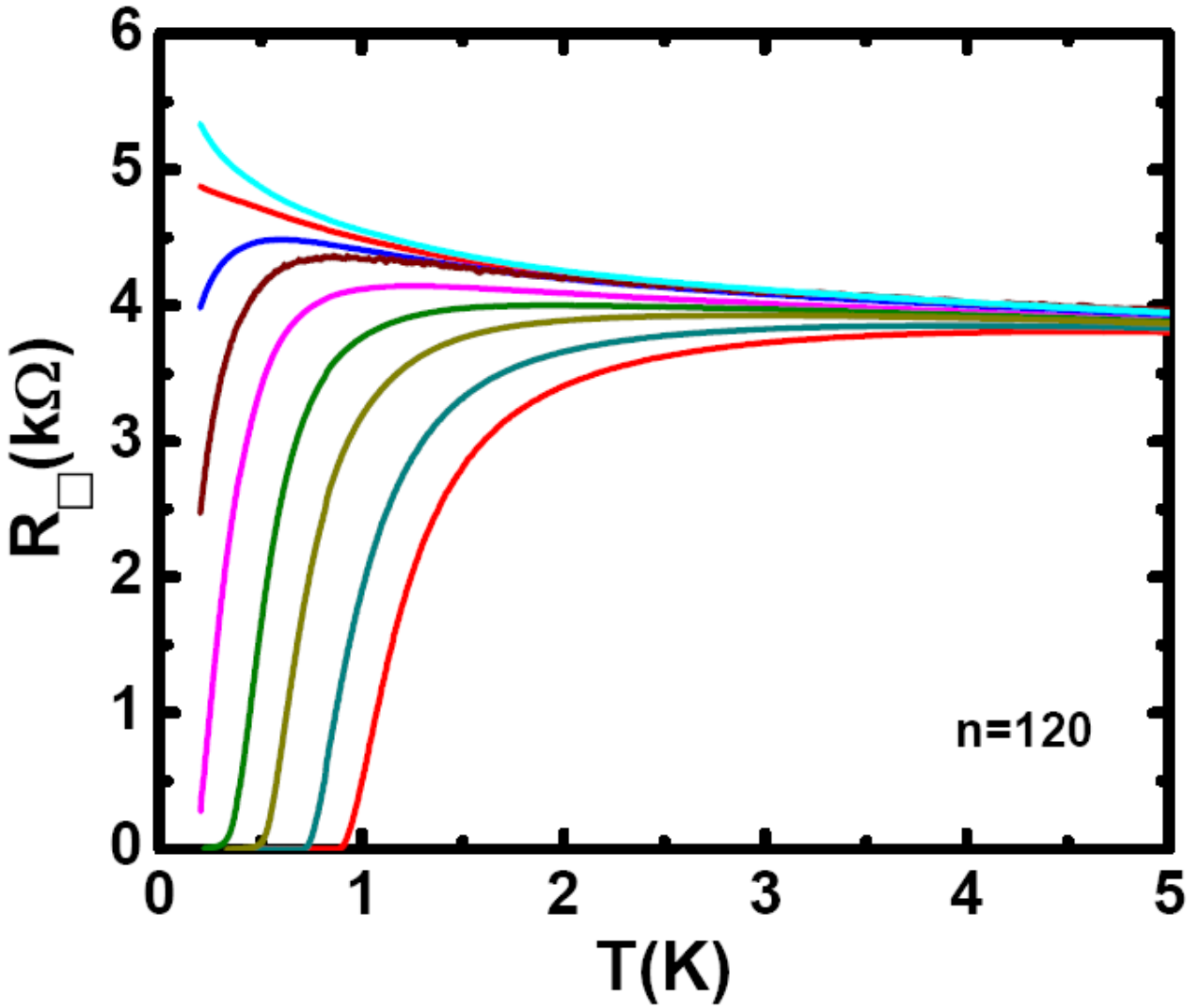
MI-tuned transition



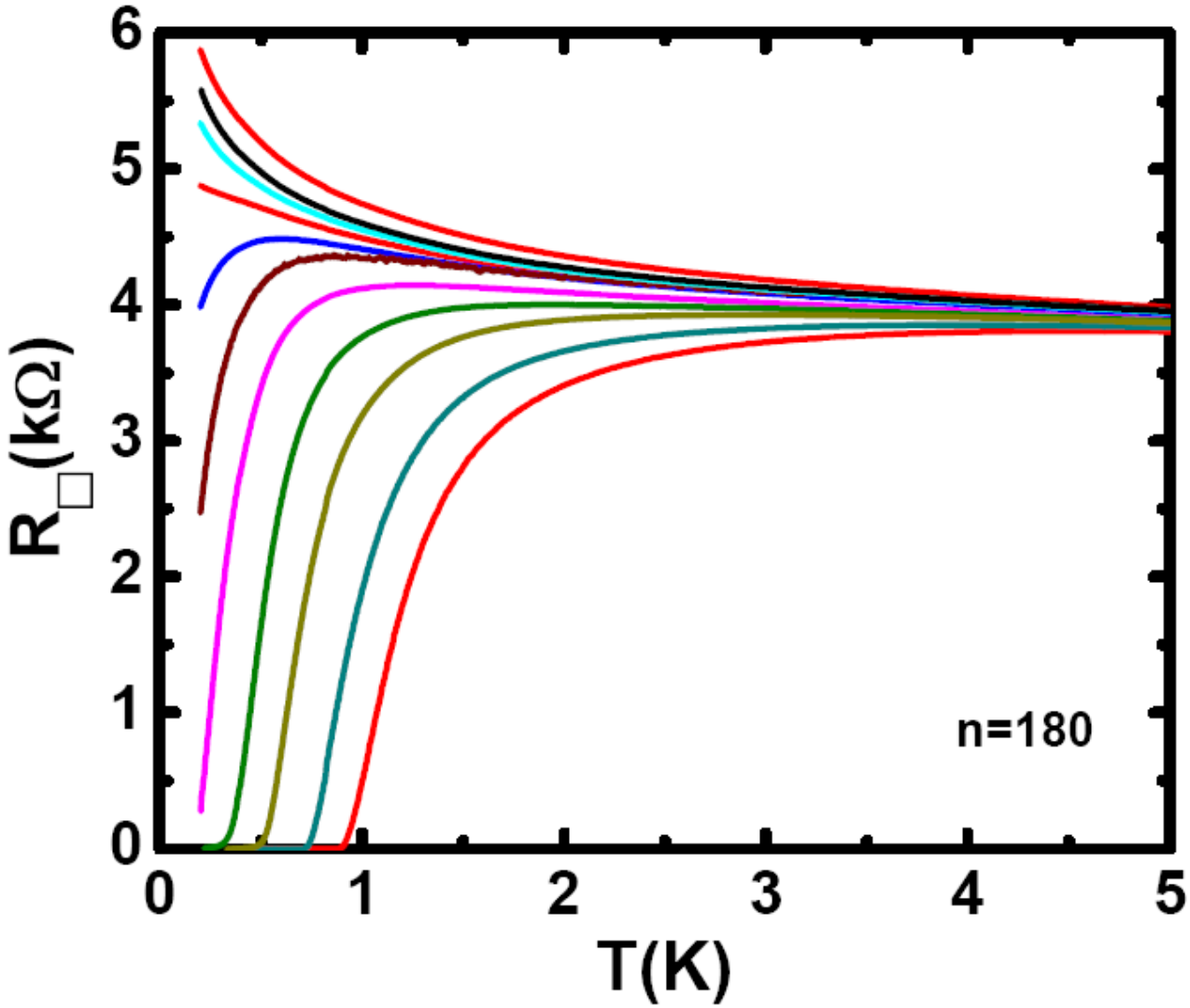
MI-tuned transition



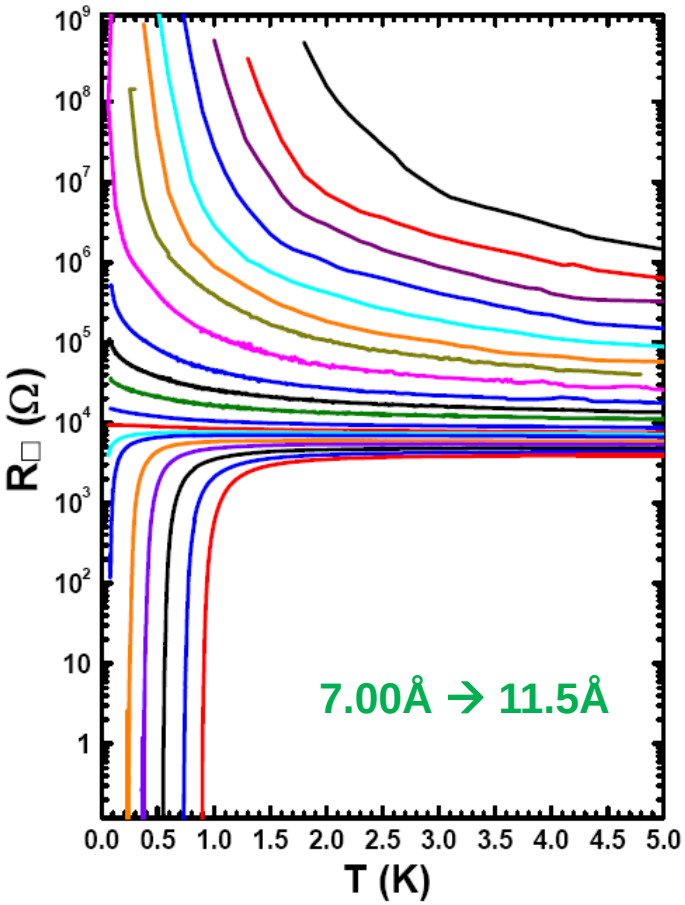
MI-tuned transition



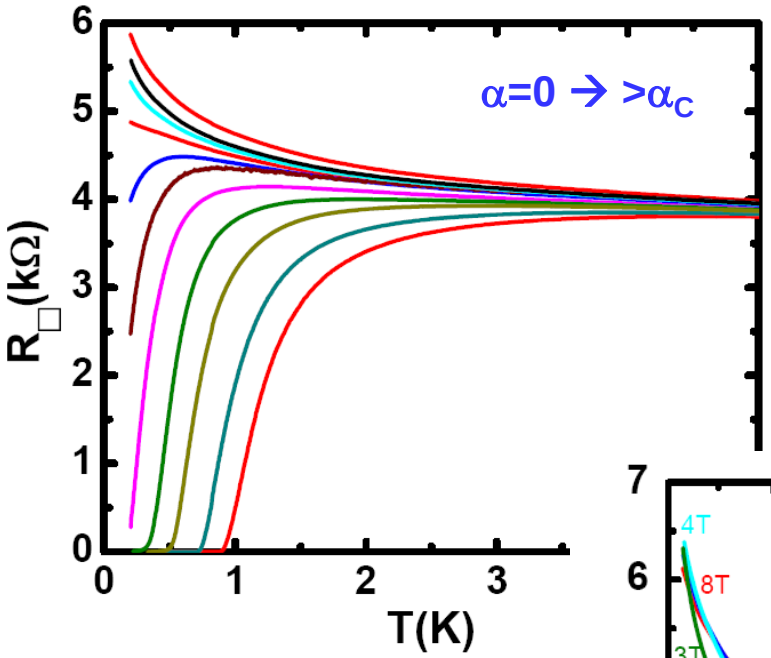
MI-tuned transition



Direct Comparison

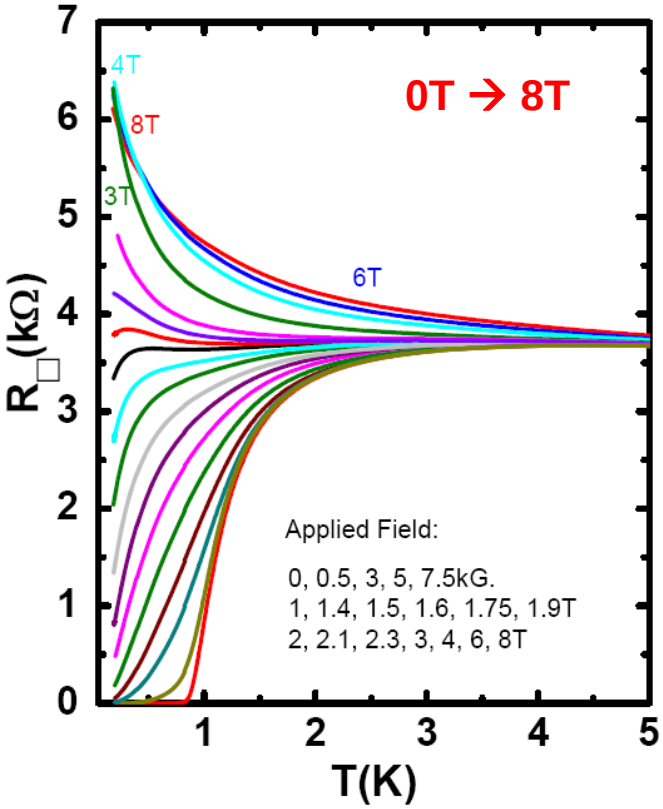


disorder (thickness)



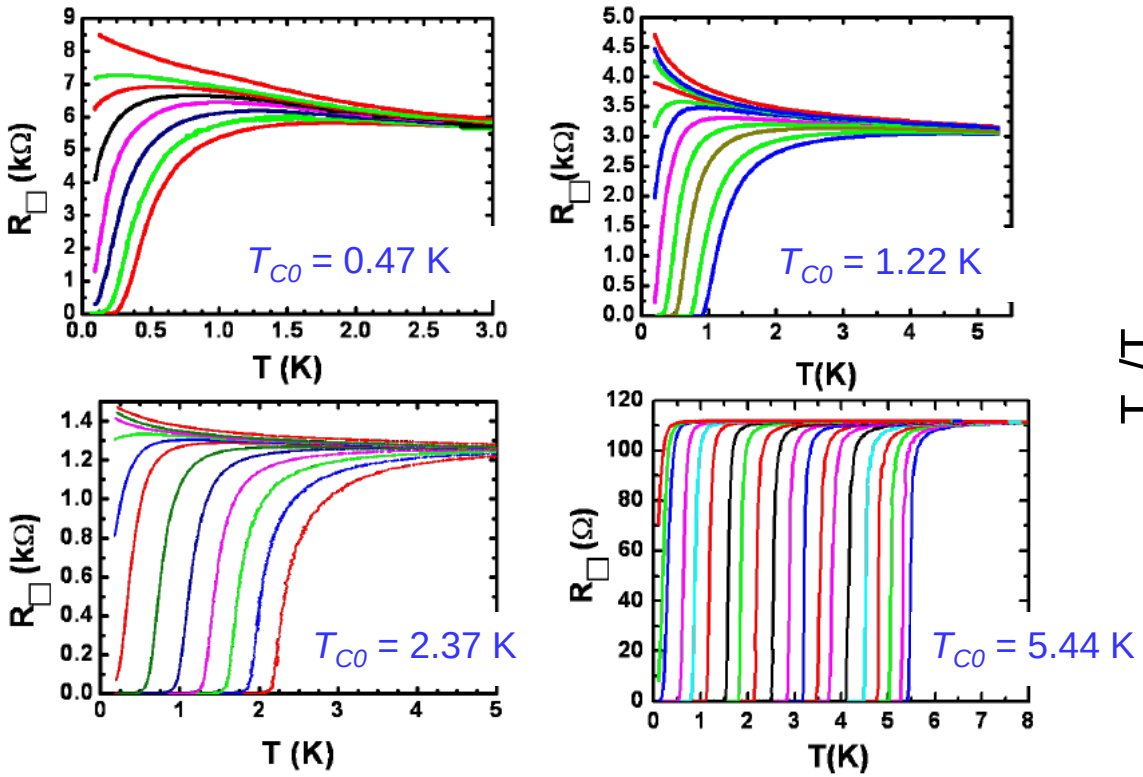
magnetic impurity

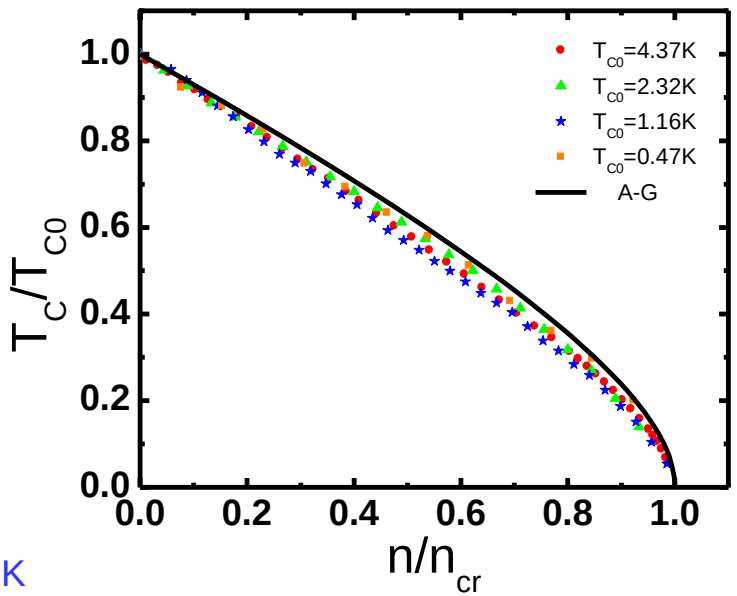
perpendicular field



Parker *et al*, Europhys. Lett. 2006

MI-tuned transition

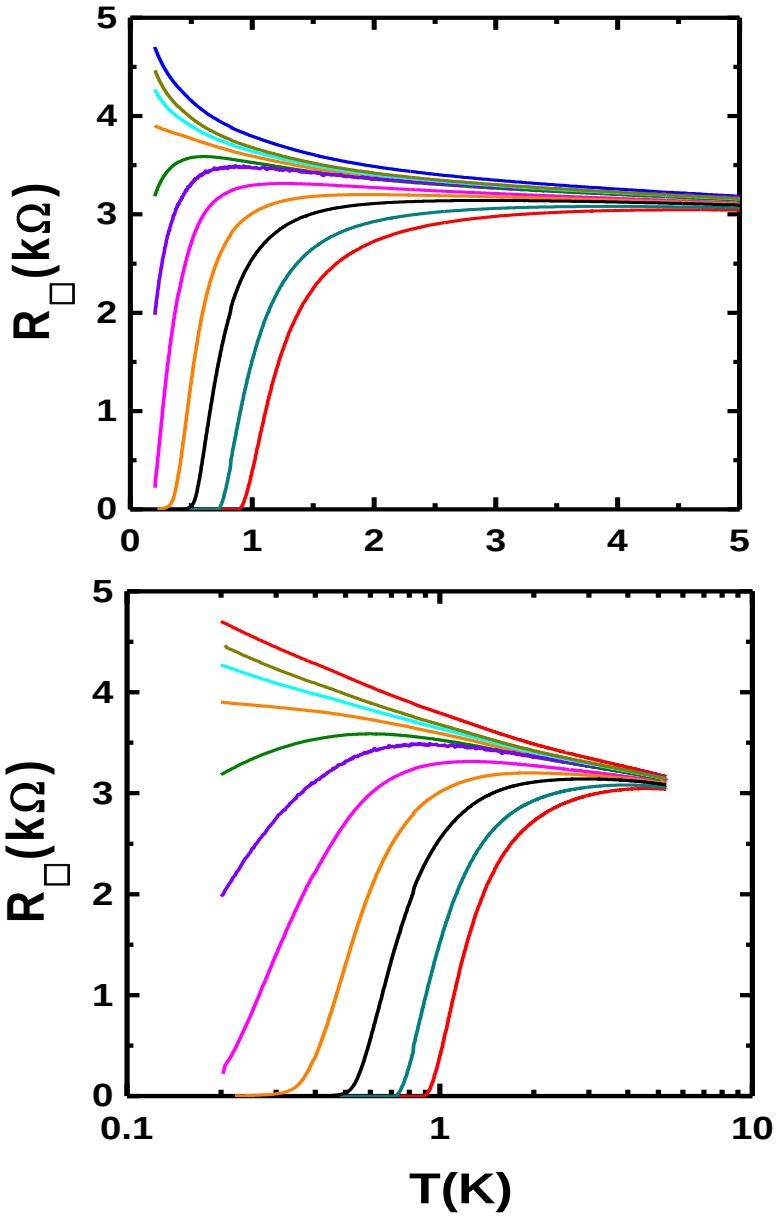




- **Destruction of superconductivity well described by A-G**
- **No superconducting pairing in insulating state**

Woolf and Reif, PRL 1962 Skalski *et al*, PR 1964

MI-tuned transition



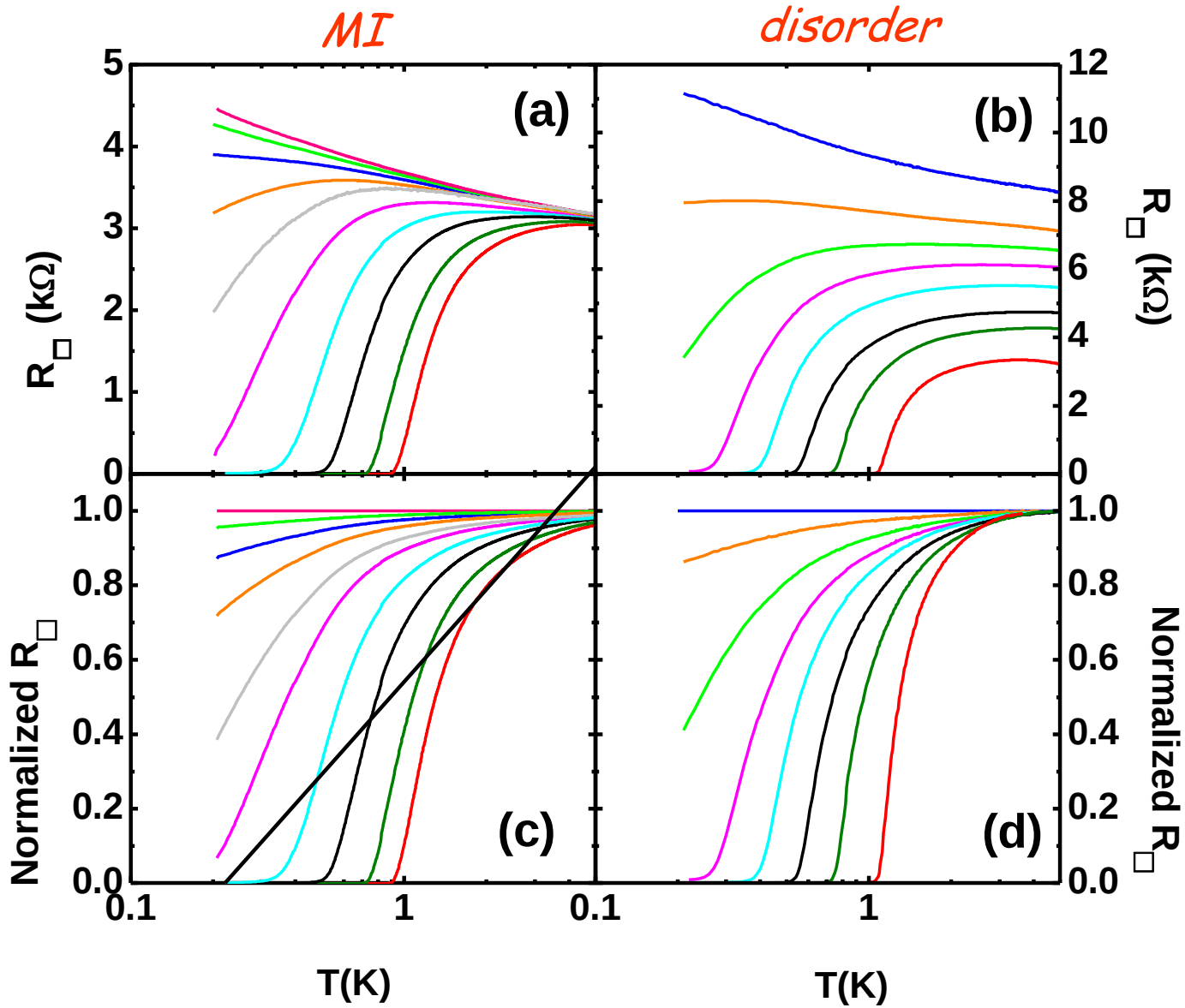
MI-tuned SIT as a model system for Fermionic transition:

- **Well-defined resistive transitions**
- **Clear phase boundary (no reentrance)**
- **Diffusive (weakly localized) transport on 'insulating' side**
- **No significant negative MR in insulating state (small positive low-field MR)**

d-tuned vs MI-tuned transitions

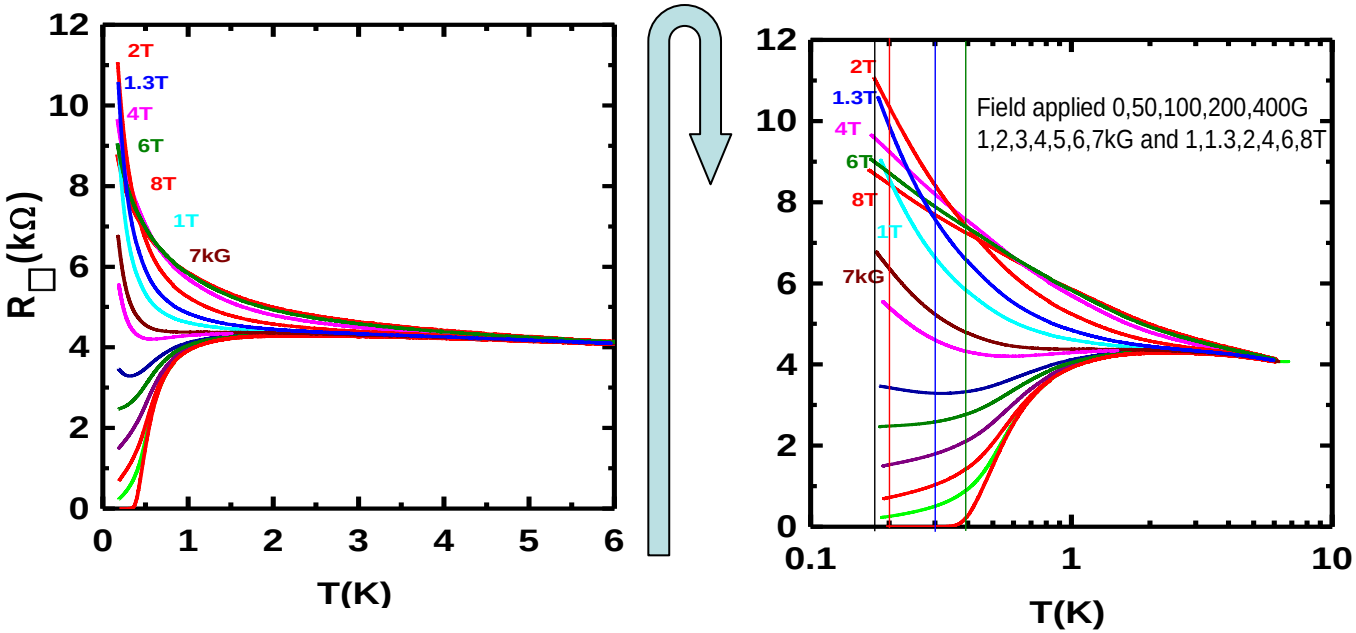
Raw data

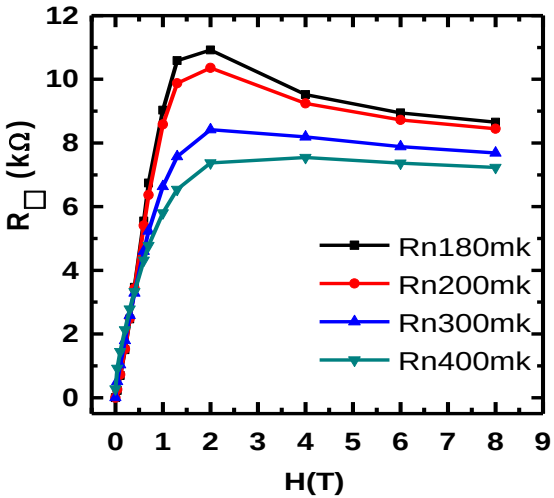
Quantum corrections normalized out (after Chervenak and Valles, PRB 1999)



Parker *et al*, Europhys. Lett. 2006

B-tuned SIT





Shahar *et al*, InOx
Baturina *et al*, TiN

Increasing magnetic field:
superconducting → activated-like → diffusive

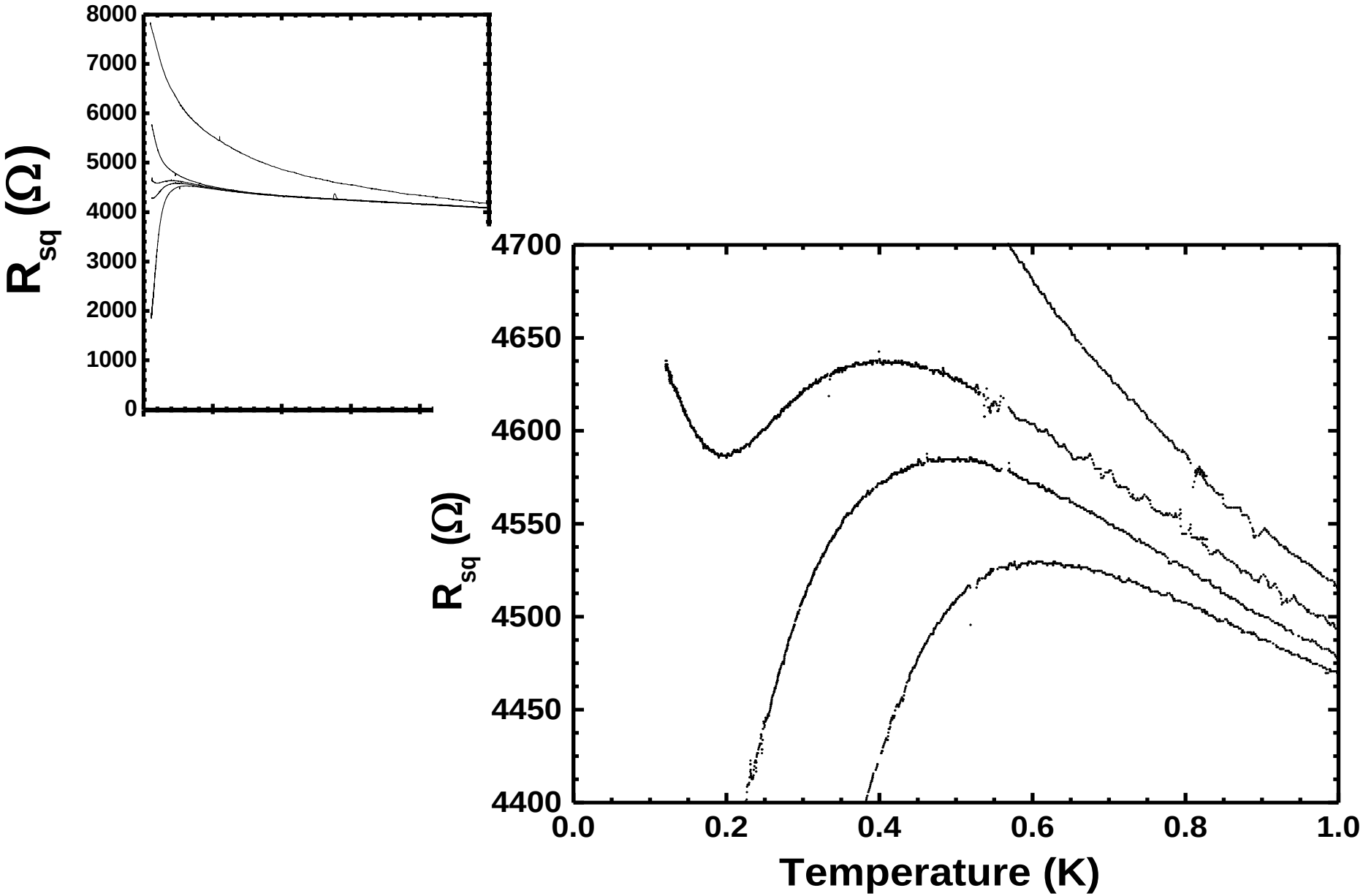
In the insulating state,

i) just above H_C, strong activated-like *T*-dependence;

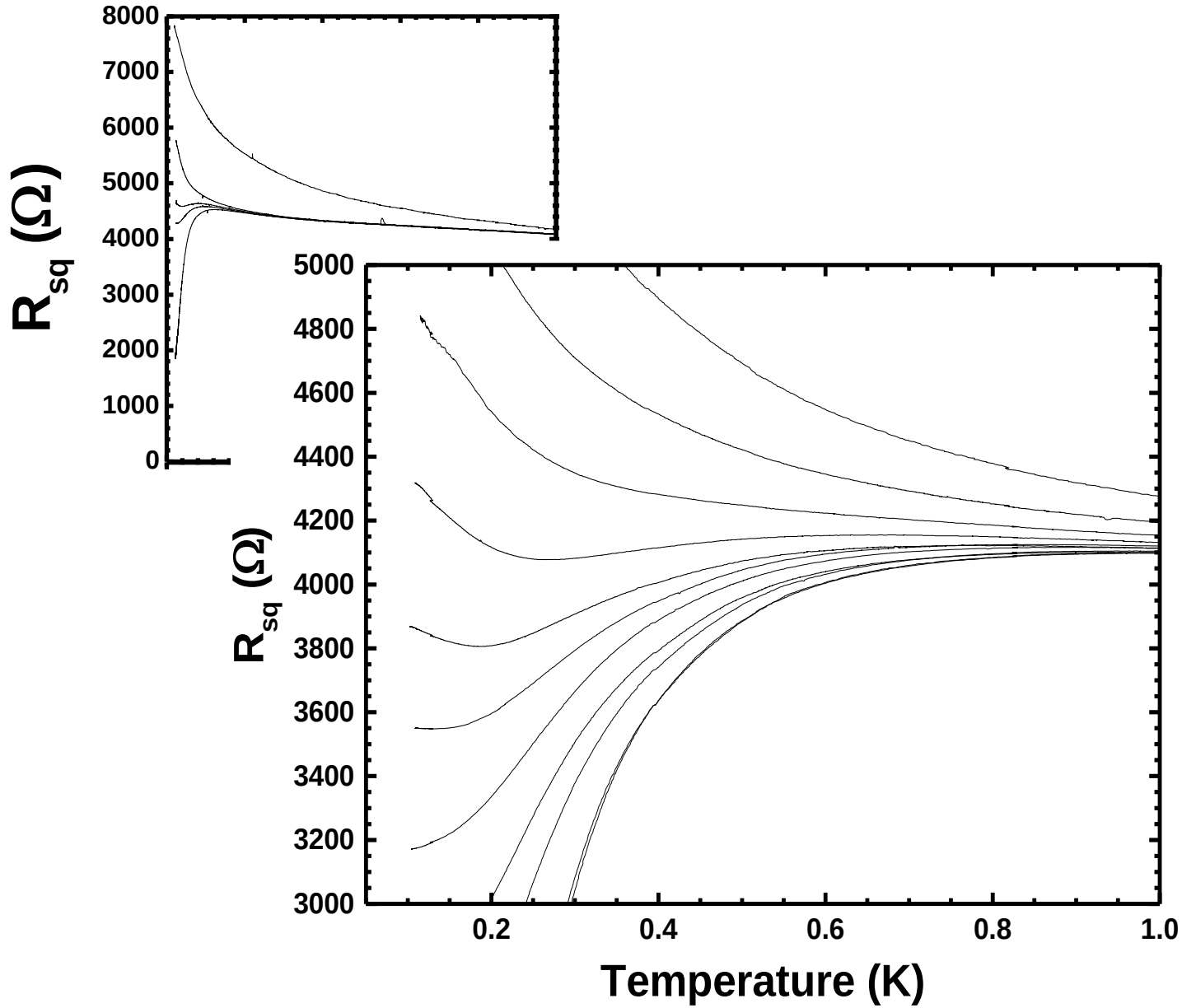
ii) weaker logarithmic dependence at much higher fields

- Non-monotonic MR (MR peak) at low temperatures
- Reentrant behavior near H_C; no clear phase boundary for the "SIT"

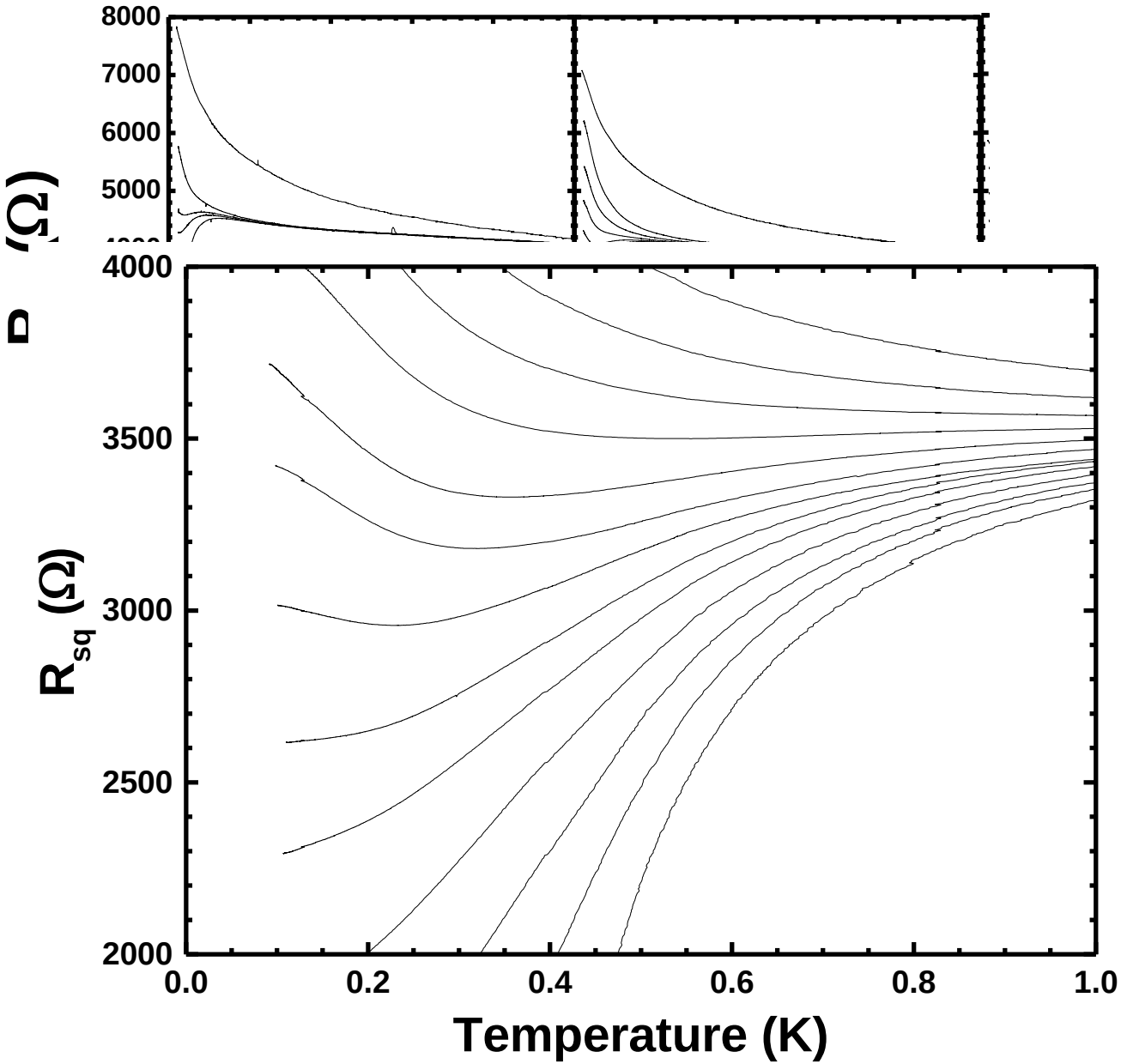
B-tuned SIT: evolution of the reentrant behavior



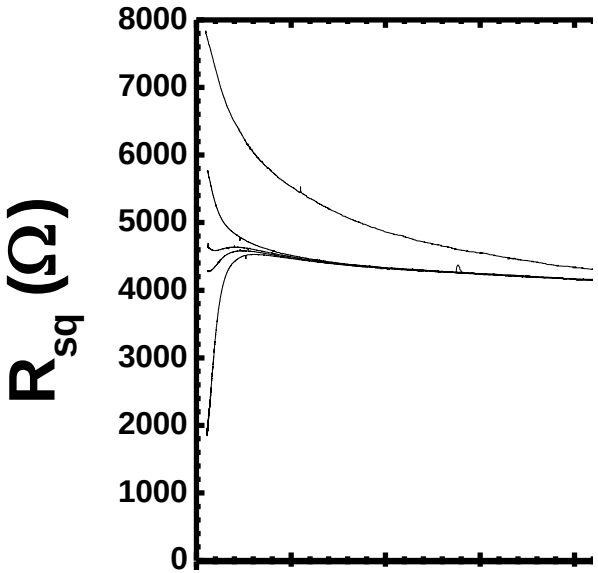
B-tuned SIT: evolution of the reentrant behavior

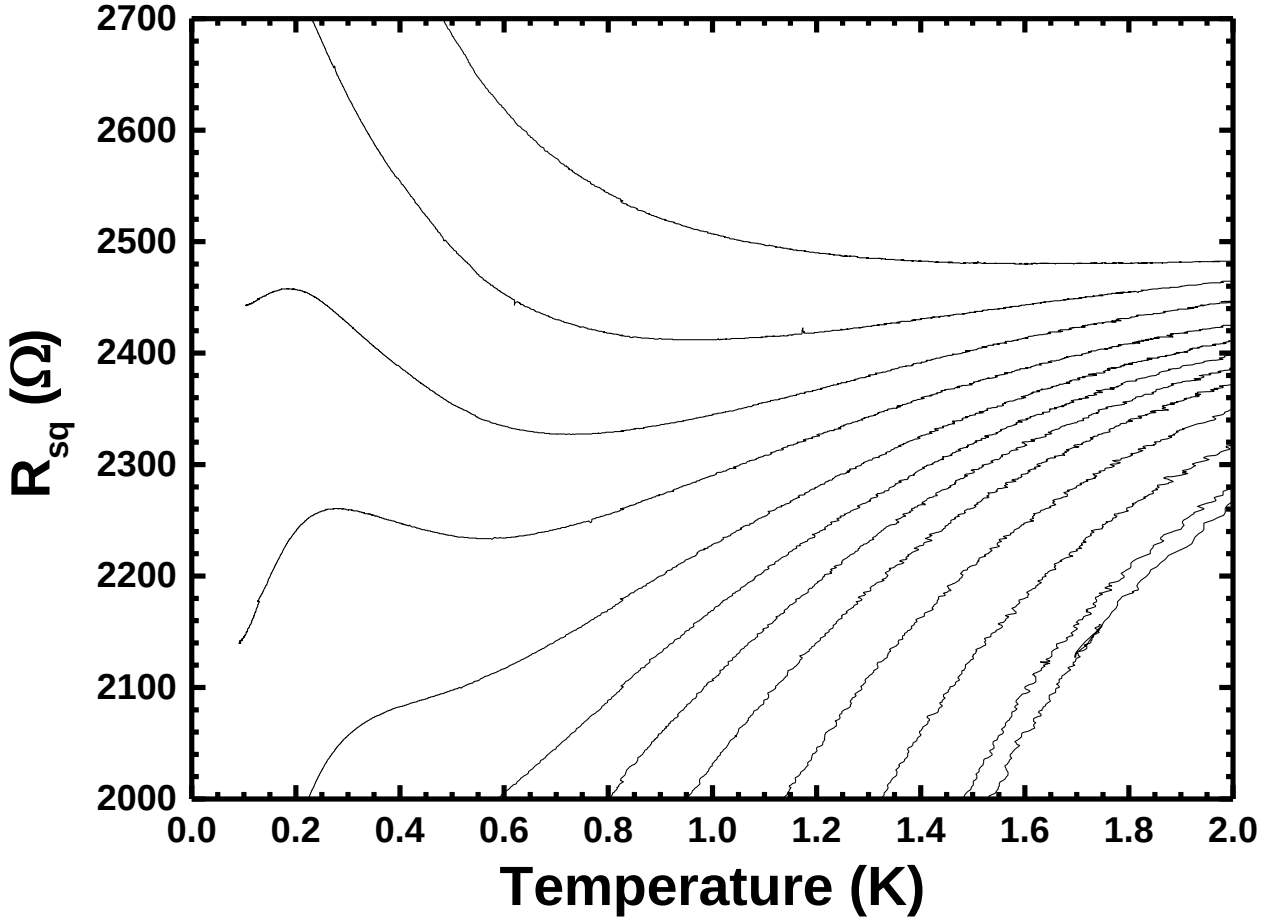


B-tuned SIT: evolution of the reentrant behavior

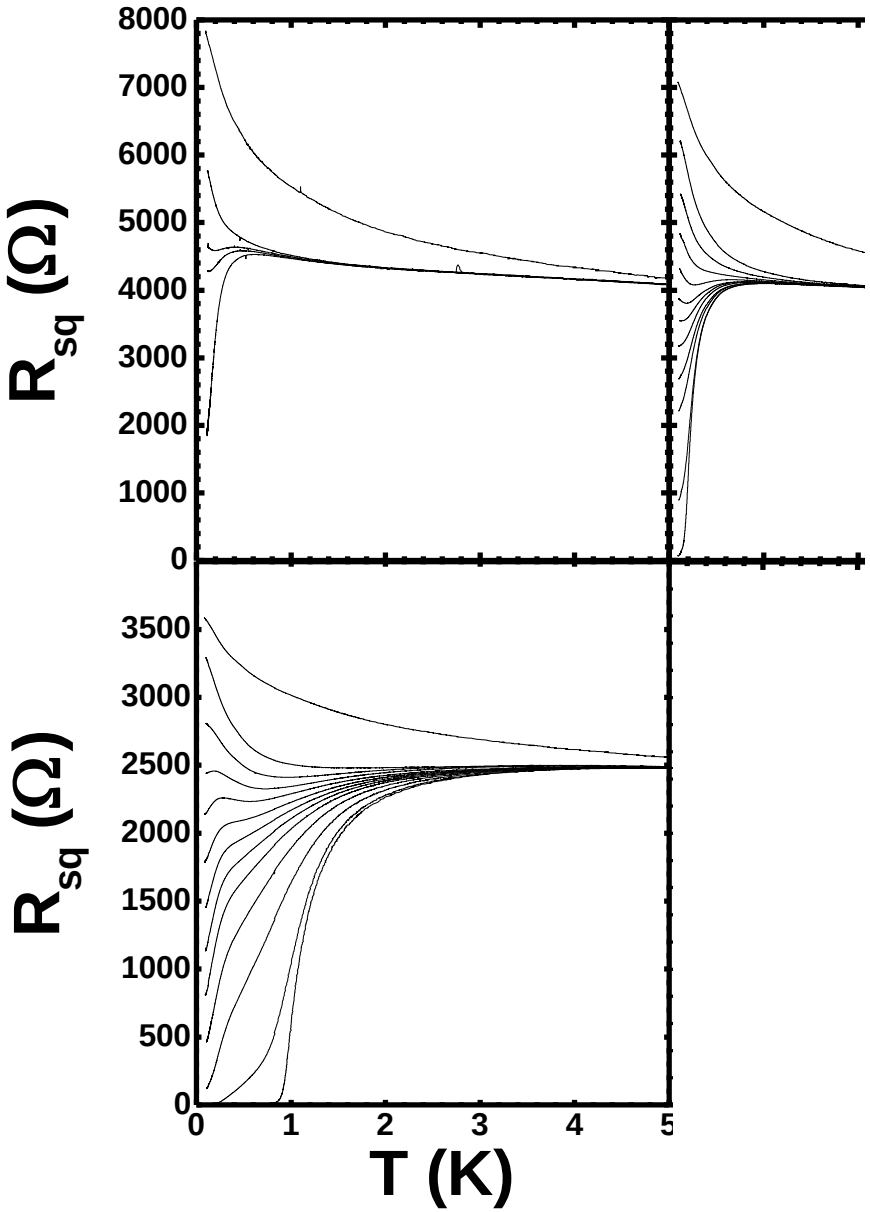


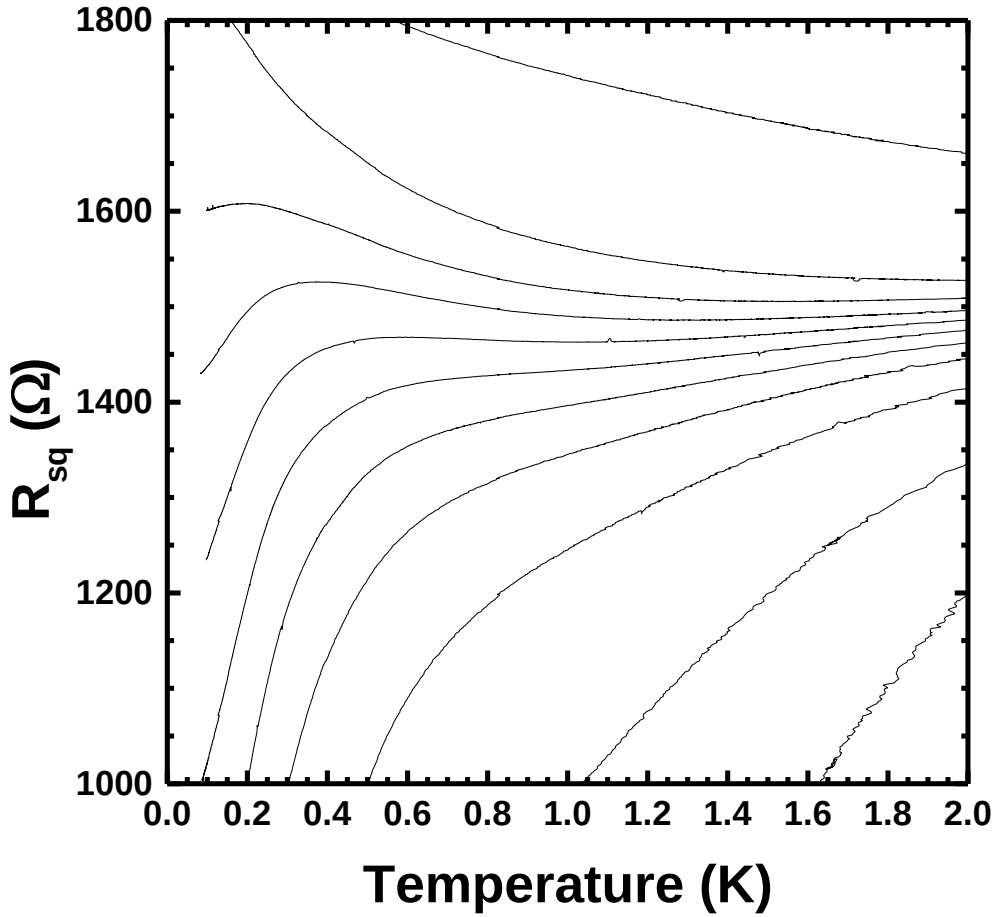
B-tuned SIT: evolution of the reentrant behavior



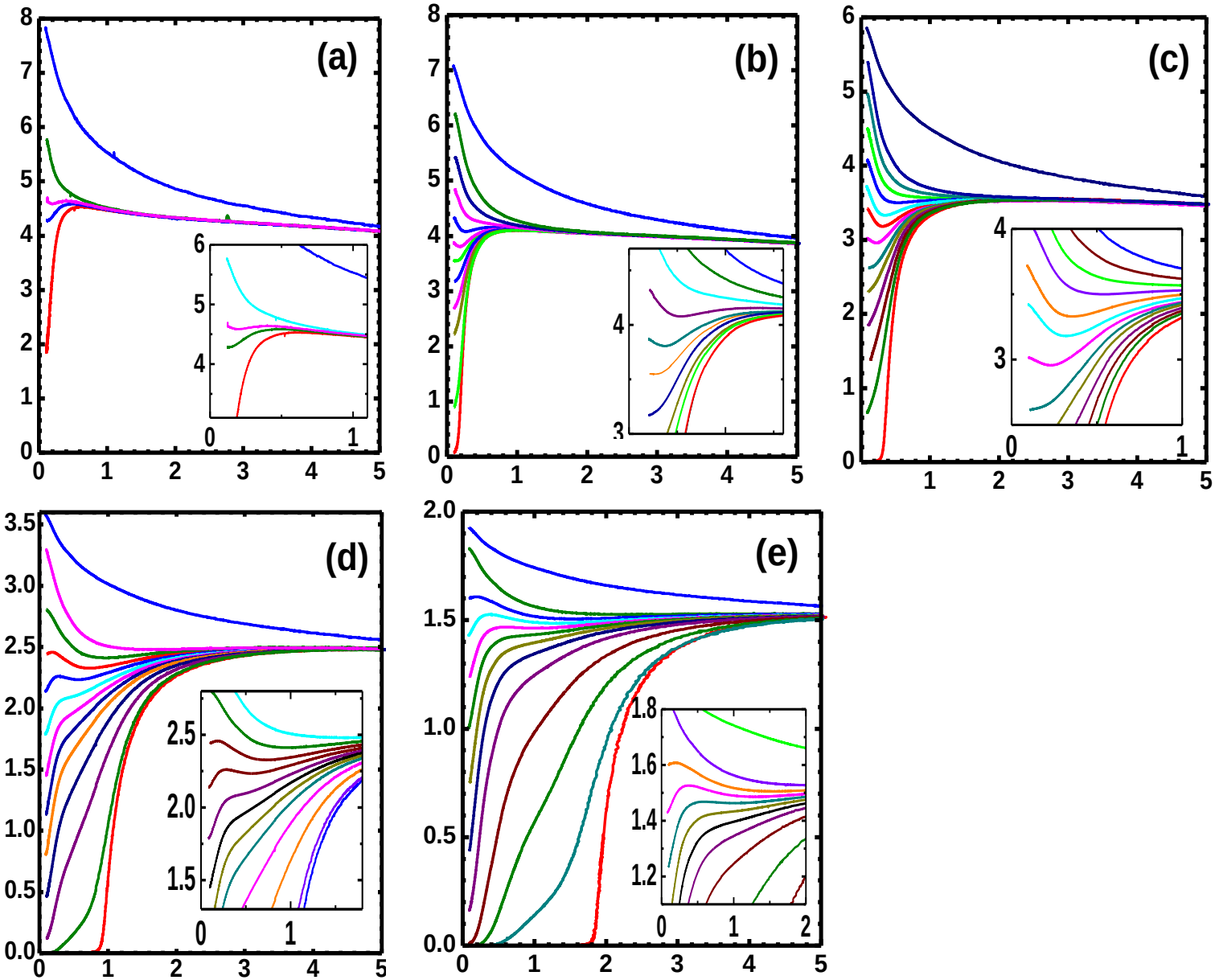


B-tuned SIT: evolution of the reentrant behavior

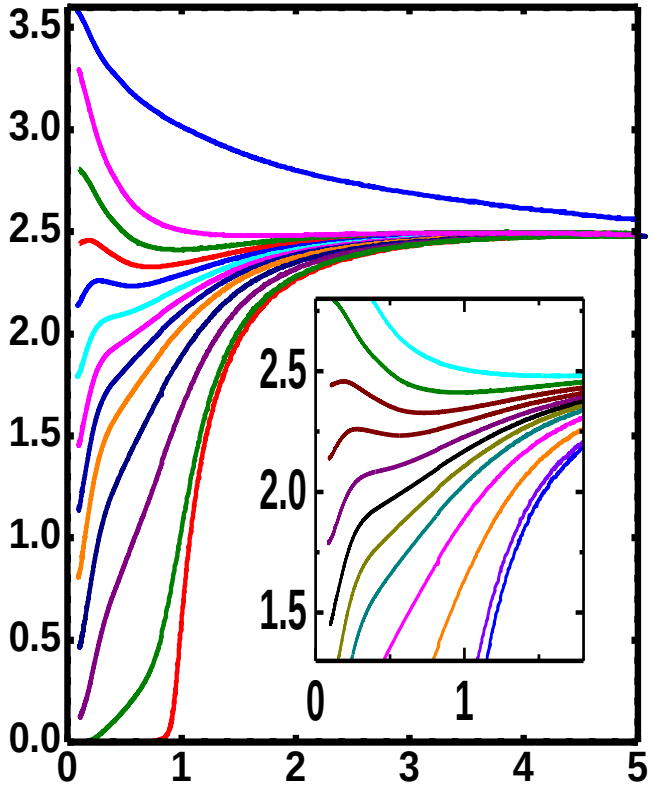




Reentrance in B-SIT

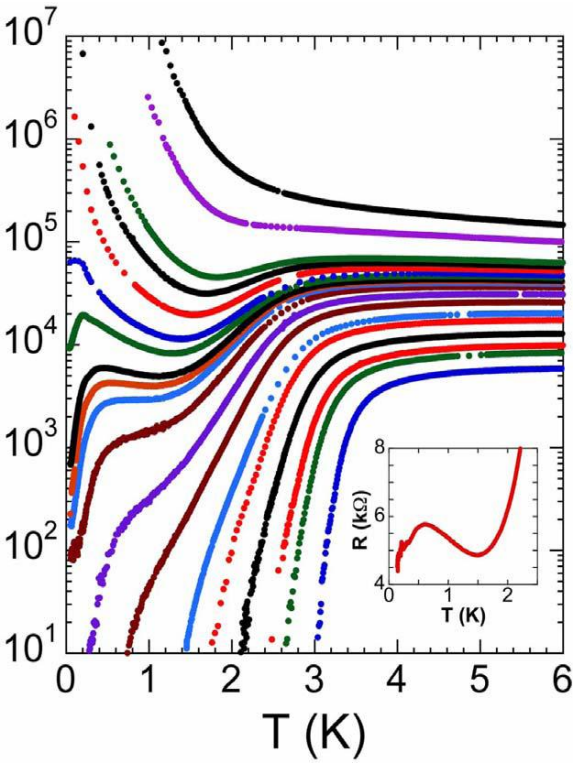


Reentrance: *a*-Pb vs granular Bi vs HTS



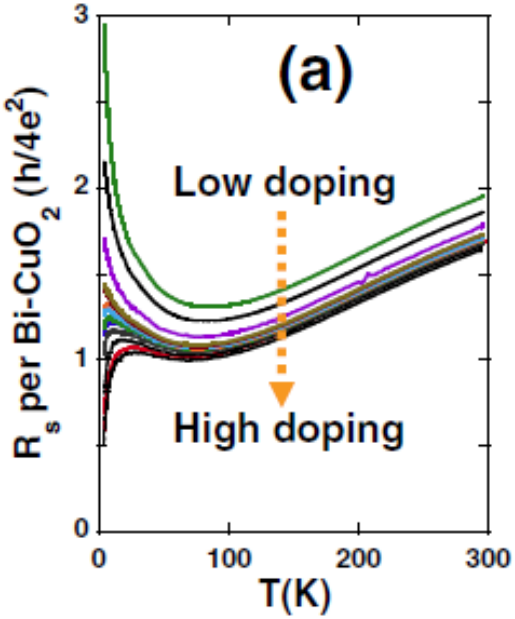
B-tuned SIT, homogeneous Pb

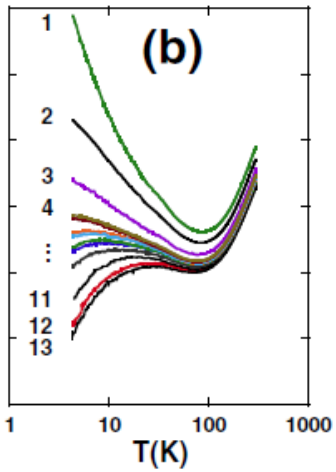
Parker *et al*, Europhys. Lett. 2006



d-tuned SIT, granular Bi

Parendo *et al*, PRB 2007





p-tuned SIT, BSCCO:La

Oh *et al*, PRL 2006

Summary: B-tuned SIT

- **Several key aspects of the *B*-tuned SIT are similar to those of the *d*-tuned SIT in <u>granular</u> films.**
- **Perpendicular magnetic field induces mesoscale phase separation near H_c, resulting in an insulating state with localized superconducting puddles;**

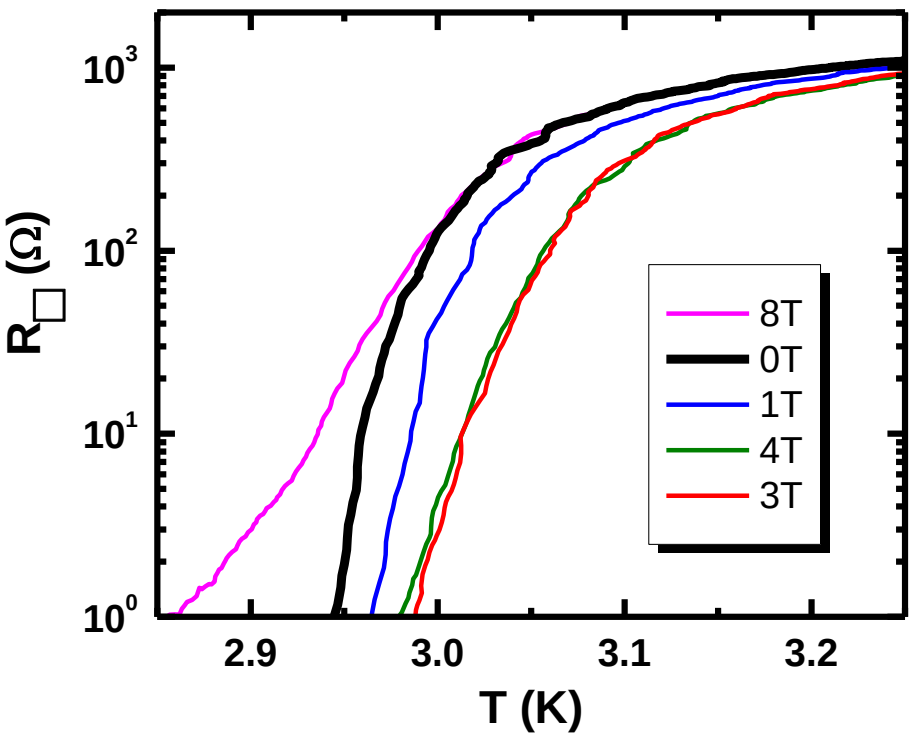
  possibly due to mesoscopic fluctuations and local enhancement of H_{c2}

  Spivak and Zhou, PRL 1995
  Galitski and Larkin, PRL 2001

- **R_N determines the Josephson coupling strength between the superconducting islands, i.e., whether *R* goes to zero or infinity at $T = 0$.**

⇒ The field-tuned SIT in homogeneous *a*-Pb films is <u>bosonic</u> in nature.

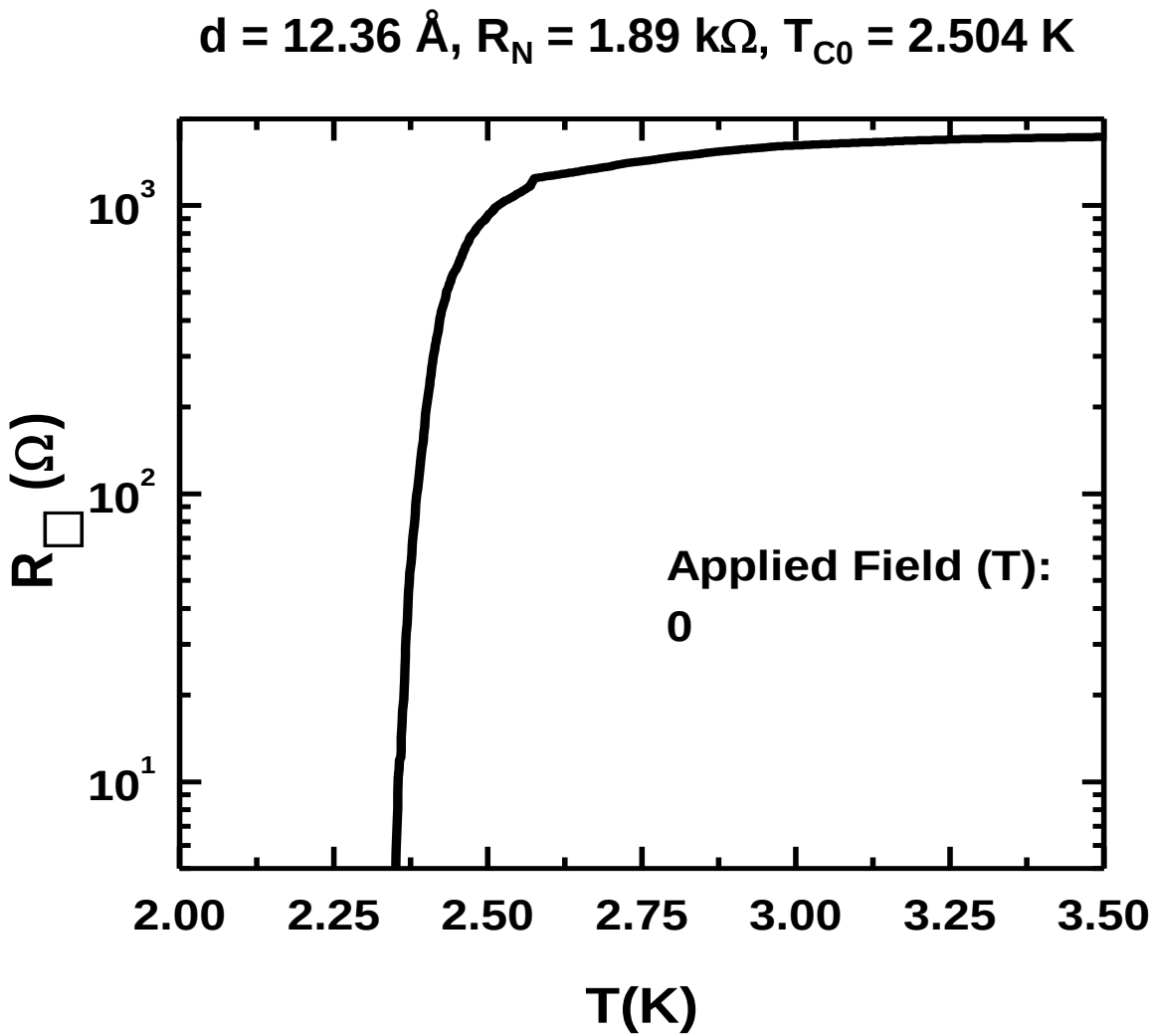
Parallel field



- **Effect of parallel field is <u>very</u> weak.**
- **Strong <u>enhancement</u> of superconductivity by parallel field**

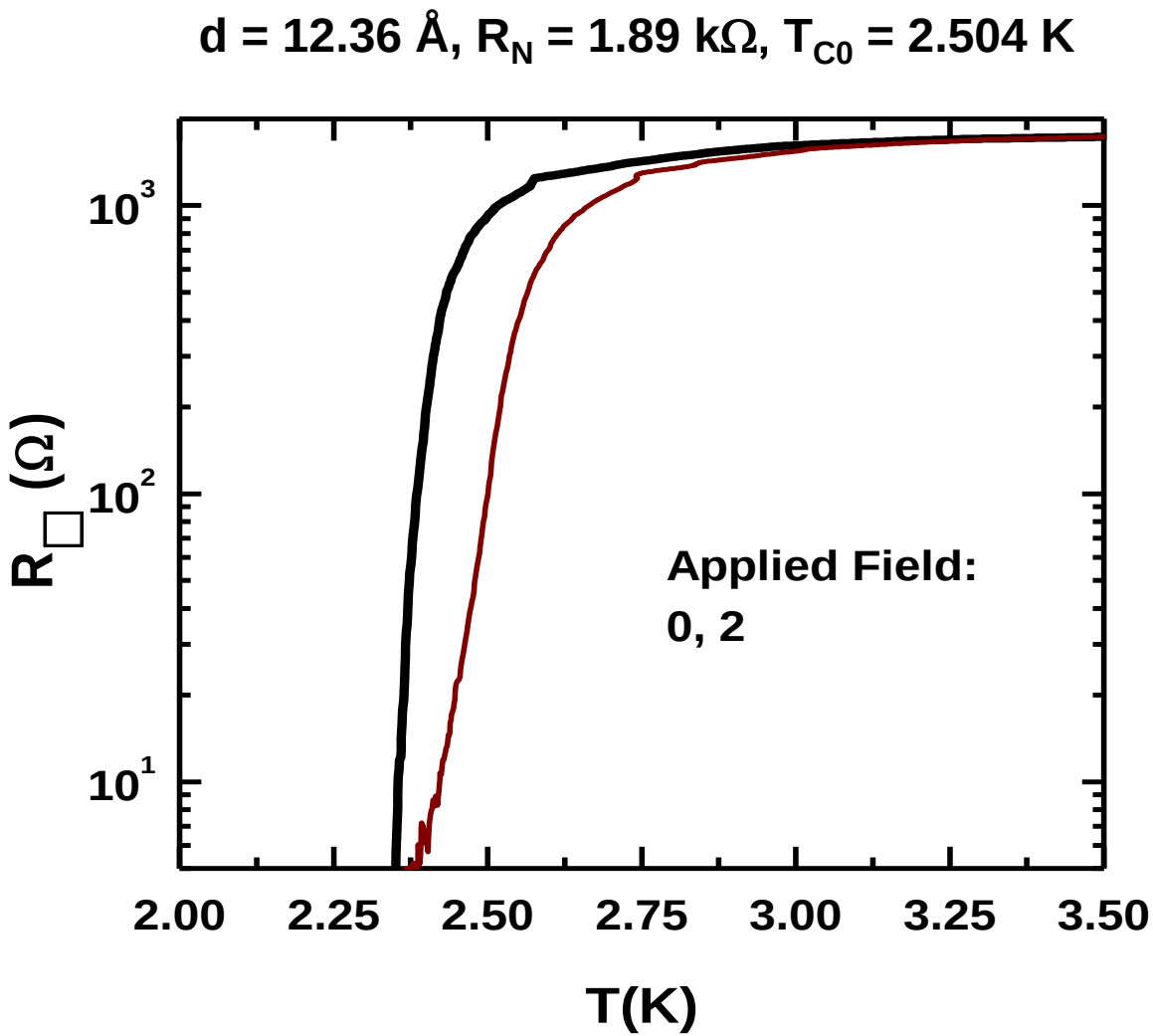
  clear T_C enhancement

Parallel field



T_C (0T) = 2.504 K

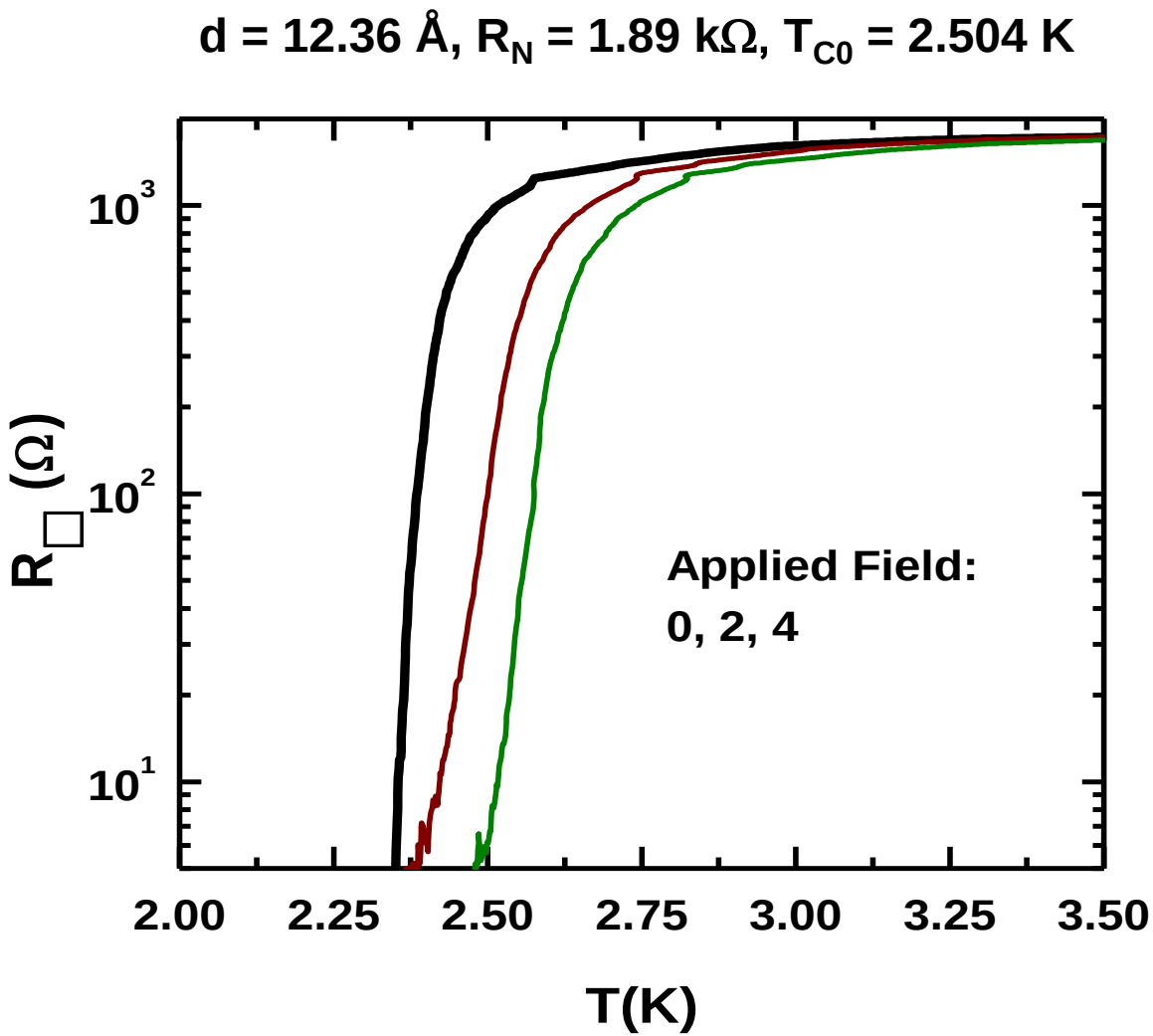
Parallel field



T_C (0T) = 2.504 K

T_C (2T) = 2.645 K

Parallel field

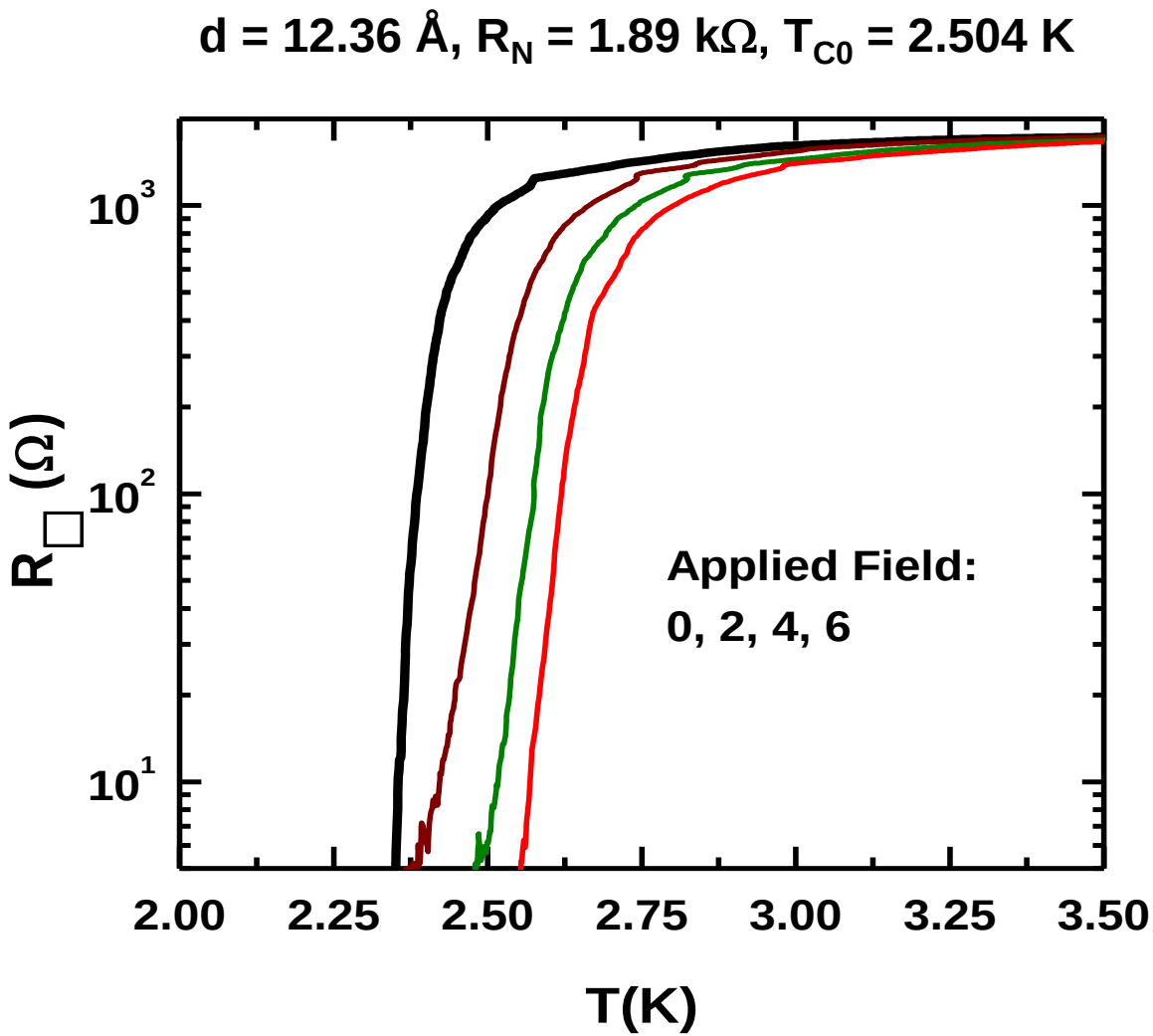


T_C (0T) = 2.504 K

T_C (2T) = 2.645 K

T_C (4T) = 2.721 K

Parallel field



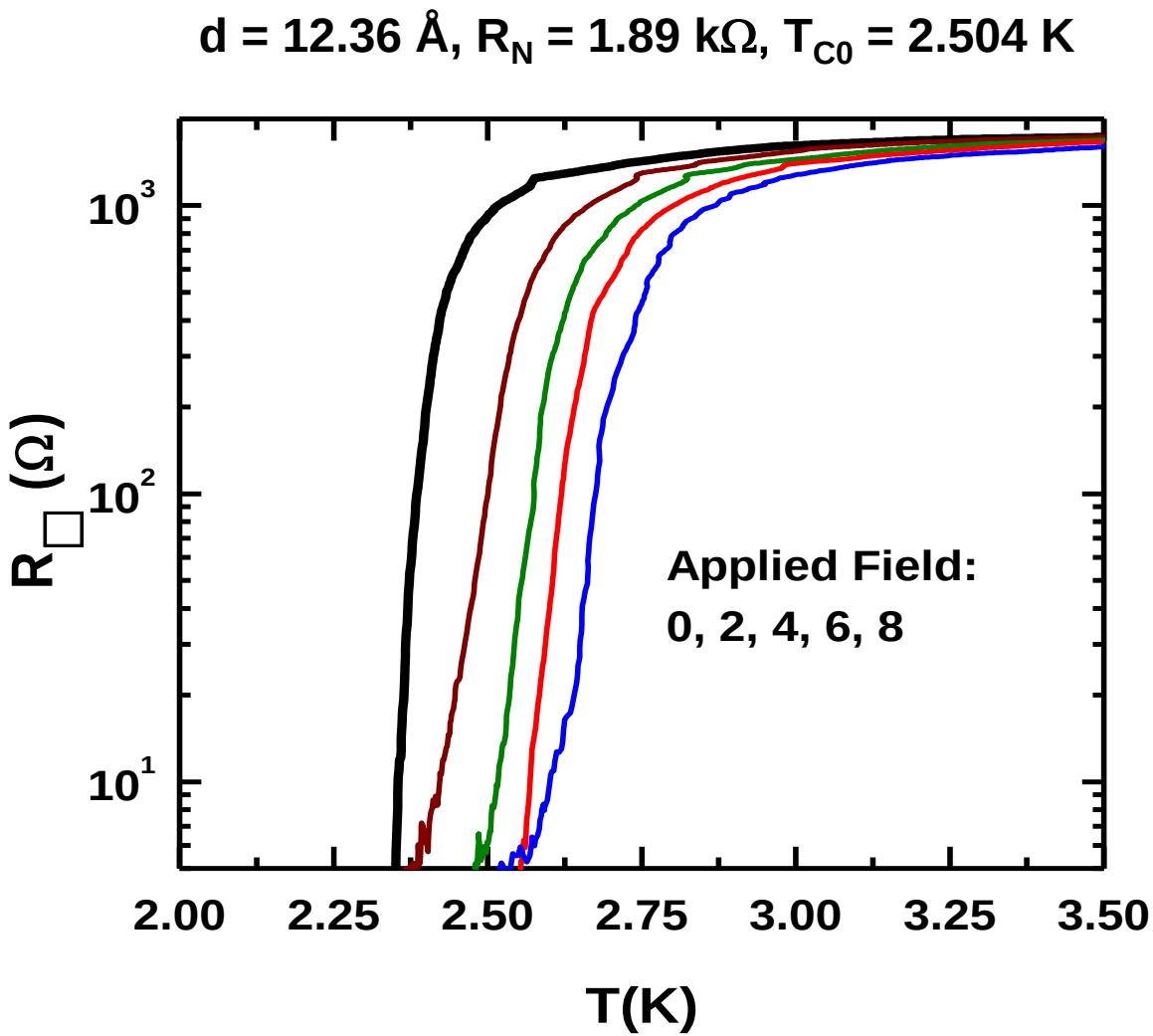
T_C (0T) = 2.504 K

T_C (2T) = 2.645 K

T_C (4T) = 2.721 K

T_C (6T) = 2.777 K

Parallel field



T_C (0T) = 2.504 K

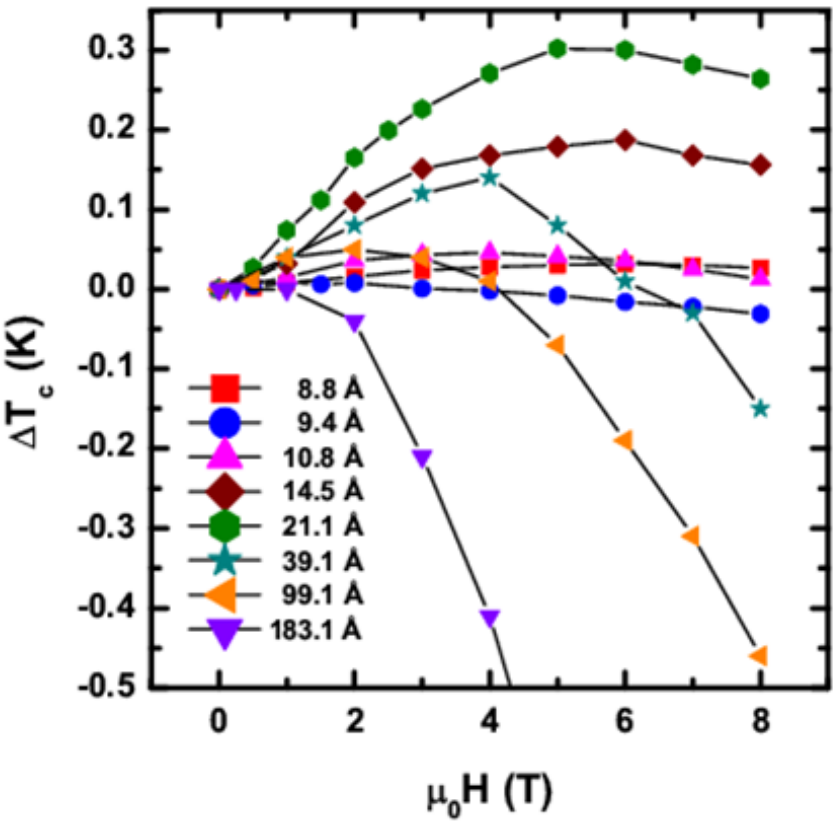
T_C (2T) = 2.645 K

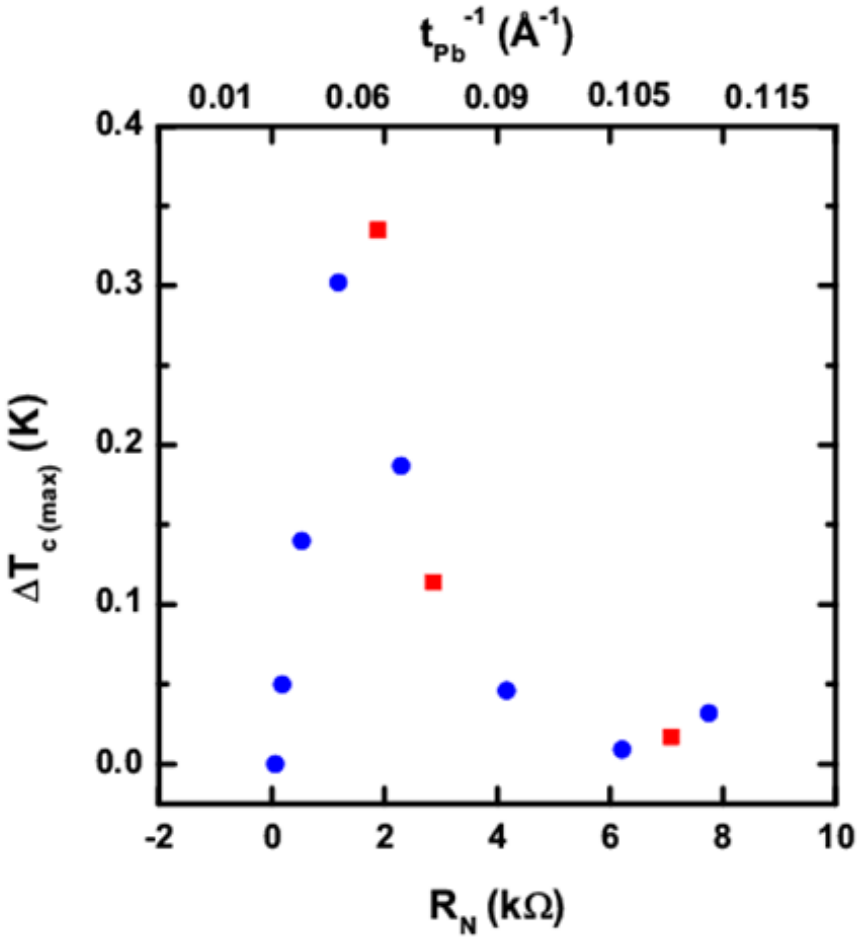
T_C (4T) = 2.721 K

T_C (6T) = 2.777 K

T_C (8T) = 2.839 K

thickness dependence

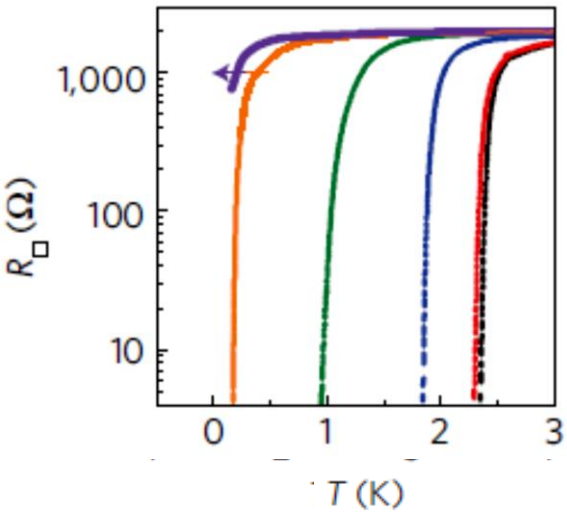




***T_c* enhancement exhibits a non-monotonic dependence on film thickness.**

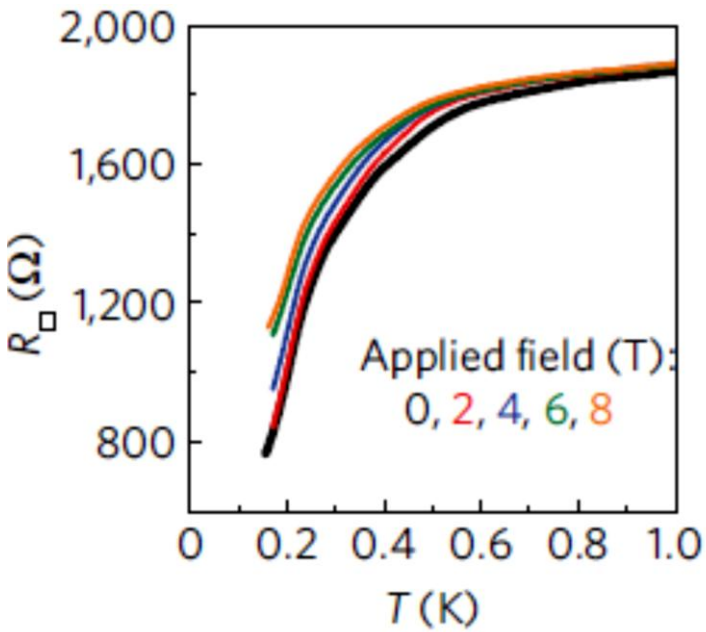
Gardner *et al*, Nature Phys. 2011

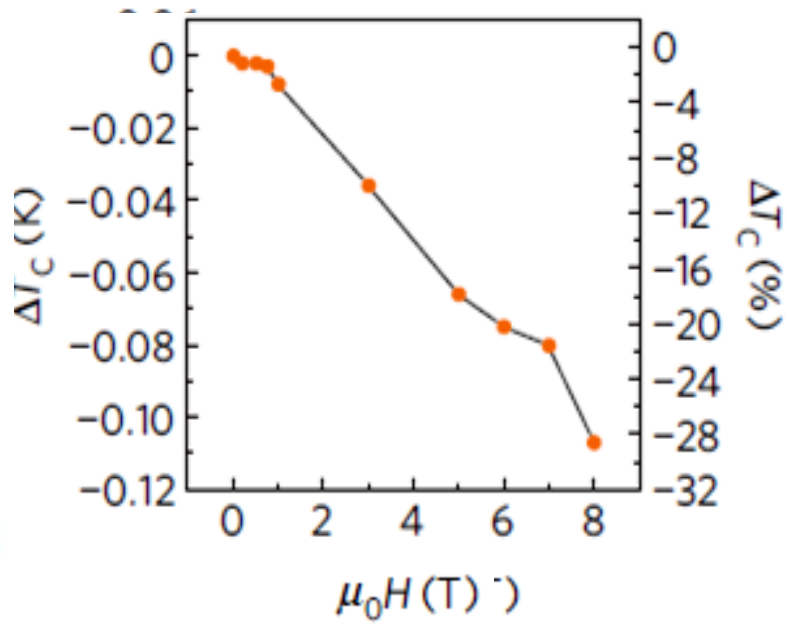
Effect of paramagnetic impurity



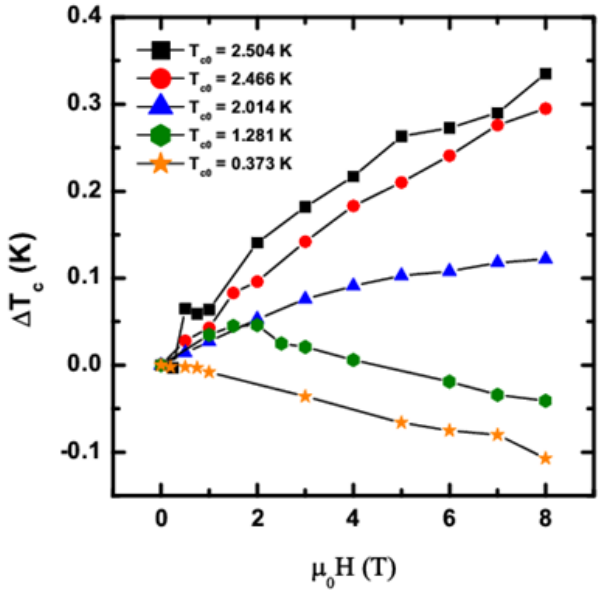
- Magnetic impurities were deposited in well controlled increments
- Field-enhanced superconductivity was systematically suppressed with increased MI concentration

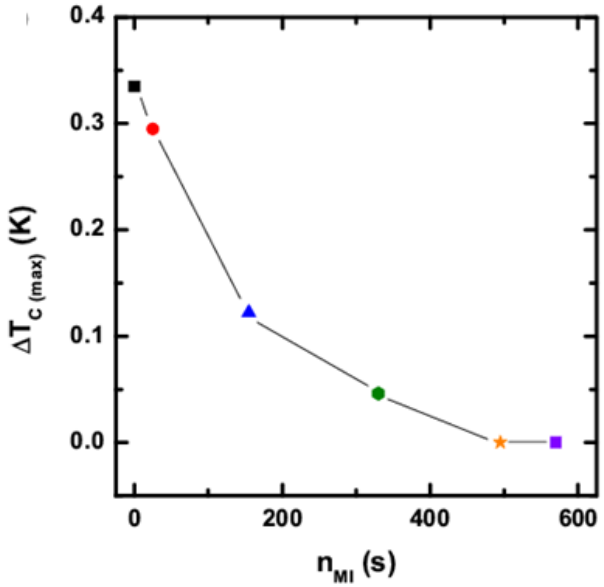
Gardner *et al*, Nature Phys. 2011

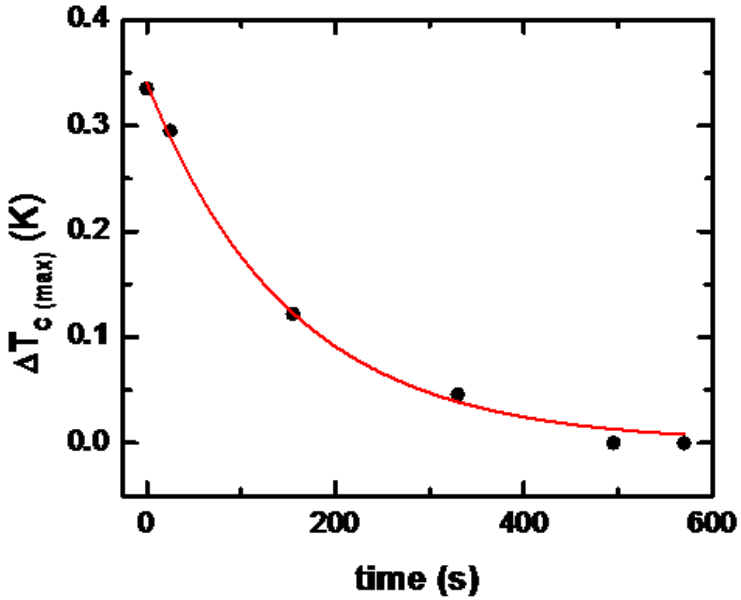




Effect of paramagnetic impurity





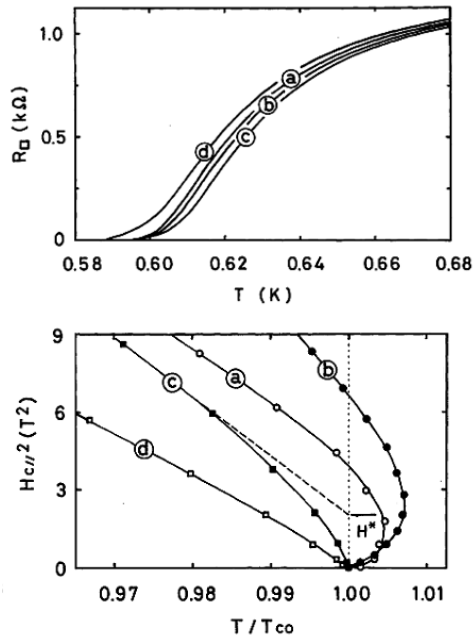


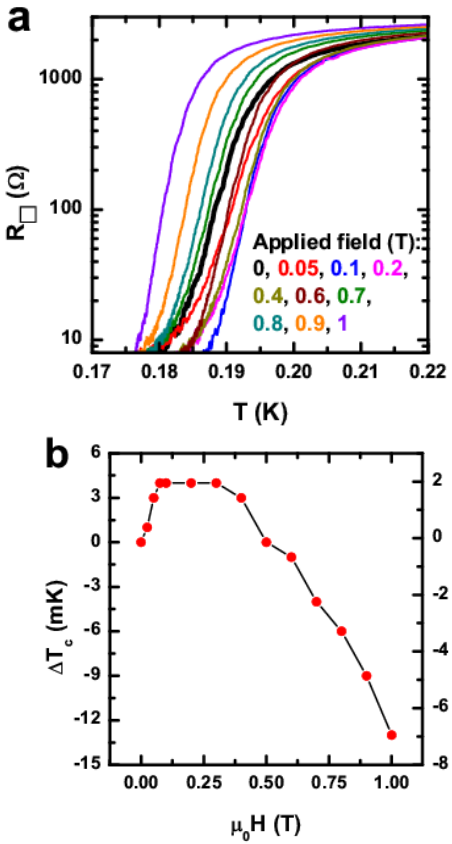
Different materials

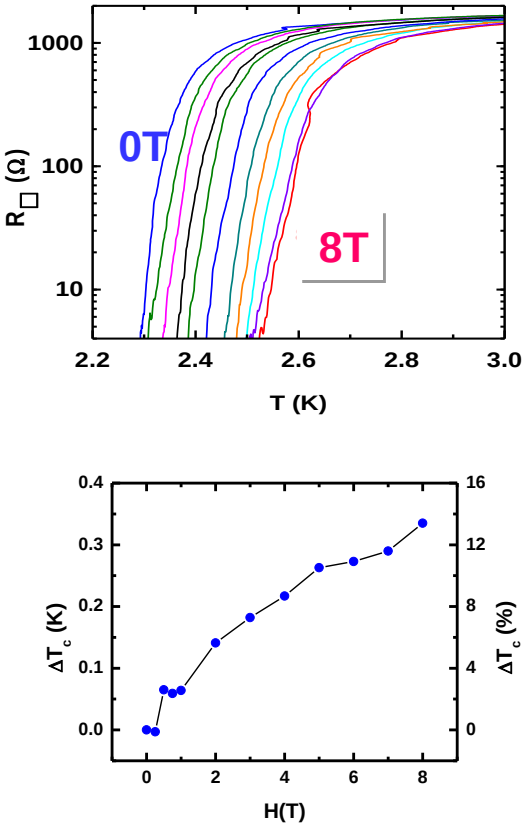
Meservey & Tedrow, *Phys. Rep.* 1994

Seguchi et al, *J. Phys. Soc. Jpn.* 1992, 1993

Gardner *et al*, Nature Phys. 2011





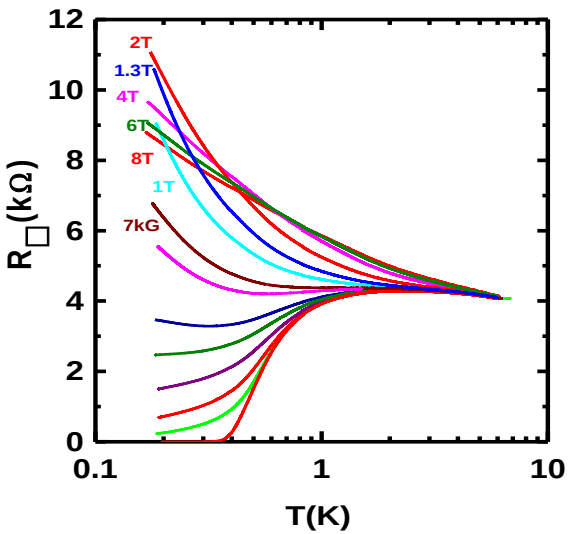


0%	< 1%	~ 2%	> 13%
thin Al	thin AuGe	LAO/STO	thin Pb

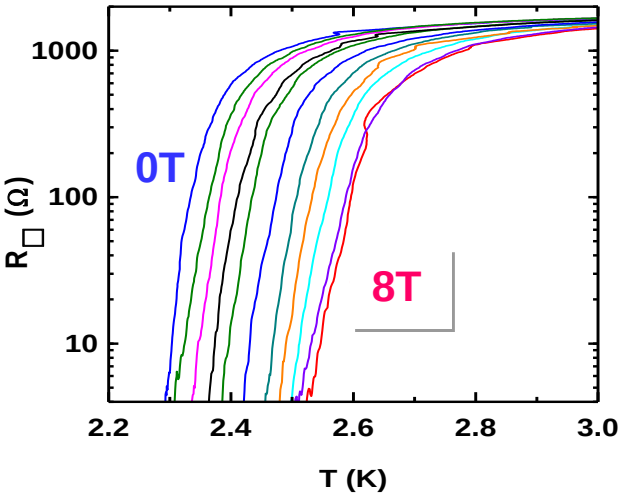
increasing spin-orbit coupling strength

Summary

In ultrathin homogeneous films of *a*-Pb:



Perpendicular magnetic field induces a super-insulating state having localized pairs.



Parallel magnetic field enhances superconductivity:

- **Large magnetic field-induced increase of mean-field T_C**
- **Presence of paramagnetic impurity is detrimental to the field-enhancement effect**
- **Essential role of spin-orbit coupling**