NORDITA
The Nordic Institute for Theoretical Physics

Topology and Geometry
Beyond Perfect Crystals

Quest for amorphous superconductors of Bi-Sb alloys by irradiation with swift heavy ions

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UAM | FACULTAD DE CIENCIAS

CMAM

Research team

Quest for amorphous superconductors of Bi-Sb …

Qmat-MeV

Quantum materials and sensors by MeV ion implantation

Ref.: PID2021-127498NBI00

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Outline

Quest for amorphous superconductors of Bi-Sb …

Spoiler Alert:

We have not yet succeeded in making amorphous and superconducting Bi-Sb material

- ♣ Introduction and Background
- ♣ Experimental techniques
- ♣ Experimental results (up to now)
- ♣ Ongoing experiments
- ♣ Conclusion and Outlook

Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

Why Bi-Sb materials?

- Bi and Bi-Sb alloys: interesting materials *per se*
  → pure bismuth is the strongest diamagnetic material, it is semimetallic, and the liquid state is denser than the solid (crystal)

- Promising thermoelectric materials
  → promising thermoelectric figure of merit:

$$zT = S^2 \frac{\sigma}{\kappa} T$$

- **Potential amorphous topological superconductors**
  → In amorphous state Bi and Bi-Sb alloys are superconductors with $T_c > 6$ K !

Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

Amorphous

Topological

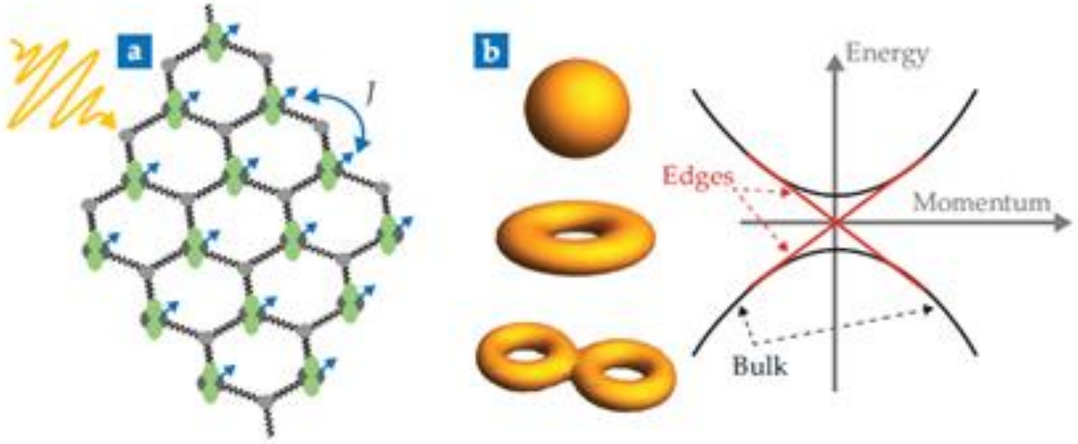
Superconductors

Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

Topological Insulators

QUANTUM MATERIALS



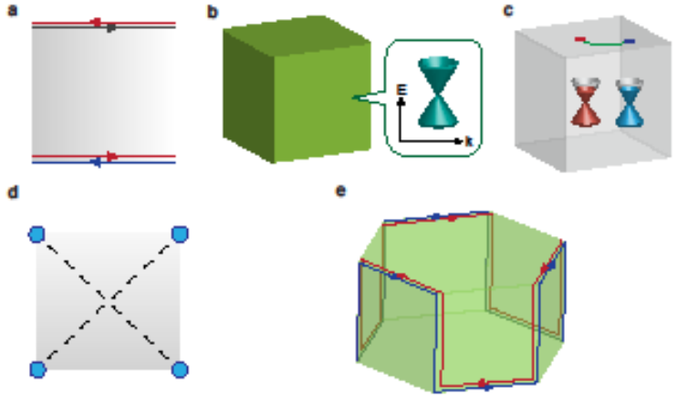
[Rodríguez-Vega, Vergniory and Fiete, Physics Today 2022]

Introduction and Background

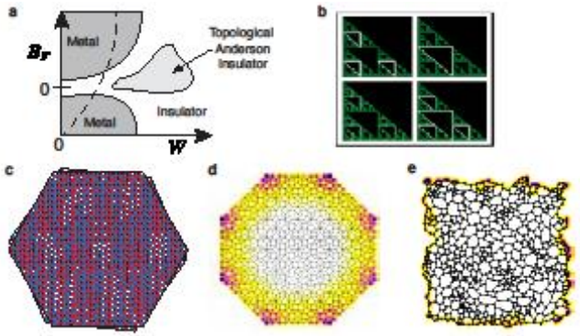
Quest for amorphous superconductors of Bi-Sb …

Amorphous Topological Matter

crystal



amorphous



Chapter 11

Topological Phases of Amorphous Matter

Adolfo G. Grushin

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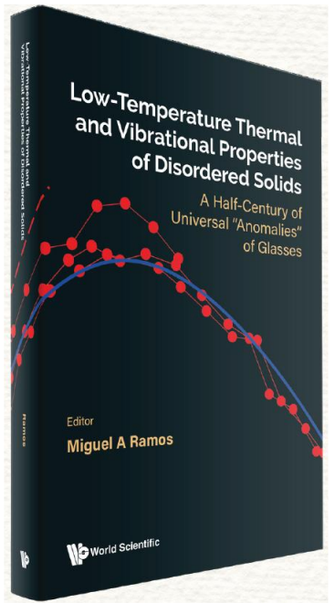
[September 2022]



epl A Letters Journal Exploring the Frontiers of Physics

April 2023

EPL, **142** (2023) 16001

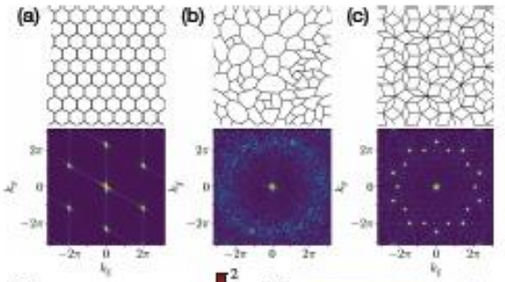
www.epljournal.org

doi: 10.1209/0295-5075/acc2e2

Perspective

Amorphous topological matter: Theory and experiment

Paul Corbae[1,2(a)], Julia D. Hannukainen[3(a)], Quentin Marsal[4(a)], Daniel Muñoz-Segovia[5(a)] and Adolfo G. Grushin[4(a)(b)]



Element \ Supercond. T_c	Crystal	Amorphous
Bi	5 mK	6 K
Be	26 mK	10 K
Al	1.18 K	6 K

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Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

REVIEWS OF MODERN PHYSICS, VOLUME 83, OCTOBER–DECEMBER 2011

Topological insulators and superconductors

Xiao-Liang Qi

Microsoft Research, Station Q, Elings Hall, University of California, Santa Barbara, California 93106, USA and Department of Physics, Stanford University, Stanford, California 94305, USA

Shou-Cheng Zhang

Department of Physics, Stanford University, Stanford, California 94305, USA

(Received 2 August 2010; published 14 October 2011)

IOP Publishing Reports on Progress in Physics

Rep. Prog. Phys. **80** (2017) 076501 (42pp) https://doi.org/10.1088/1361-6633/aa6ac7

Review

Topological superconductors: a review

Masatoshi Sato[1] **and Yoichi Ando**[2]

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[2] Physics Institute II, University of Cologne, Zülpicher Str. 77, 50937 Cologne, Germany

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Accepted for publication 3 April 2017
Published 5 May 2017

Corresponding Editor Professor Sean Washburn

CrossMark

Topological ***Superconductors***

Experimental pathways to search for topological superconductors:

Discovering **intrinsic** topological superconductors (very rare!)

Inducing Superconductivity in materials with nontrivial band topology (topological insulators, semimetals…):

- ♣ Pressure
- ♣ Doping
- ♣ Proximity effect
- ♣ …

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Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

Topological *Superconductors*

Majorana qubits
for topological
quantum computing

Ramón Aguado and
Leo P. Kouwenhoven

[Physics Today, **73**, 6, 44 (2020)]

Researchers are trying to store robust quantum information in Majorana particles and are generating quantum gates by exploiting the bizarre non-abelian statistics of Majorana zero modes bound to topological defects.

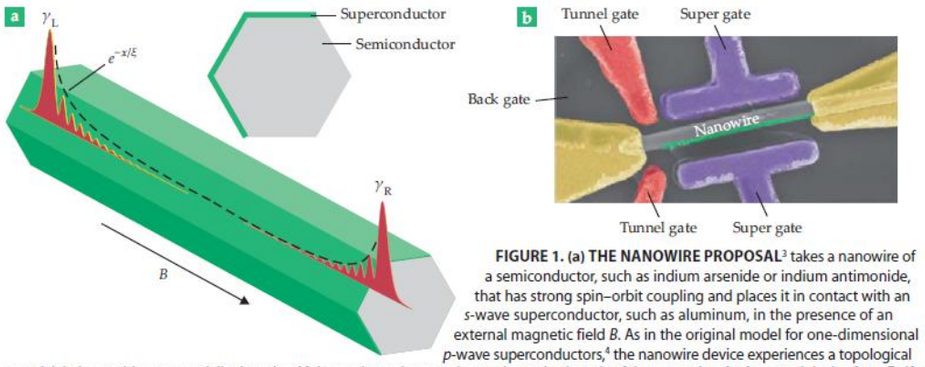
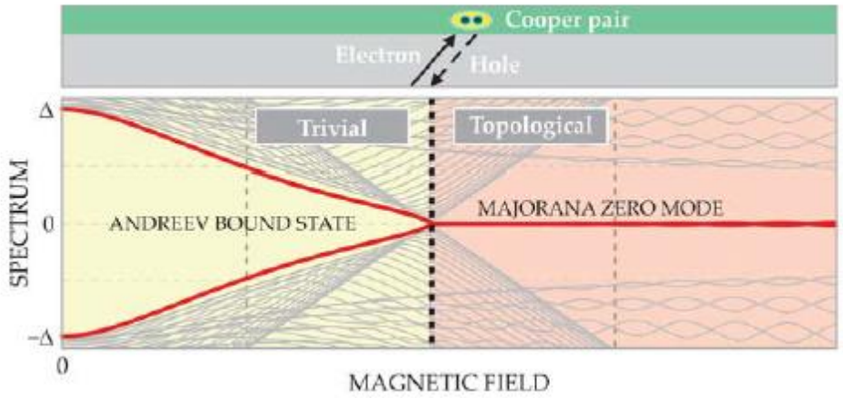
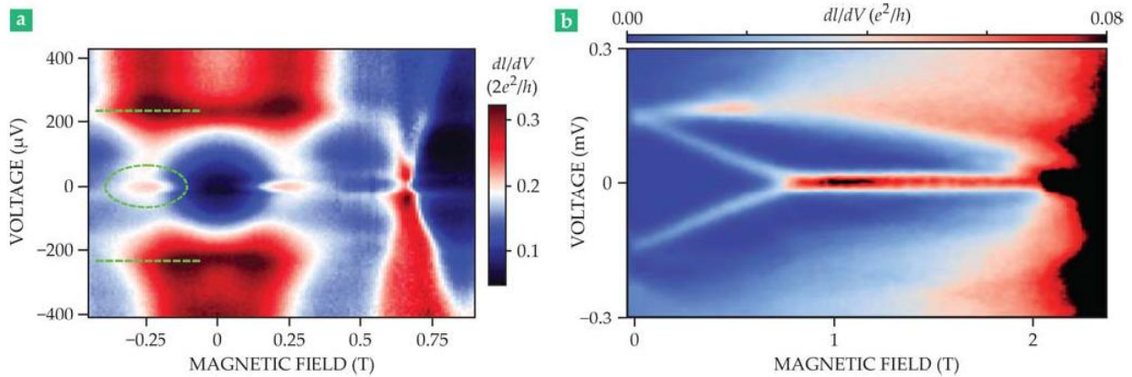


FIGURE 1. (a) THE NANOWIRE PROPOSAL[3] takes a nanowire of a semiconductor, such as indium arsenide or indium antimonide, that has strong spin–orbit coupling and places it in contact with an *s*-wave superconductor, such as aluminum, in the presence of an external magnetic field *B*. As in the original model for one-dimensional *p*-wave superconductors,[4] the nanowire device experiences a topological nontrivial phase with exponentially decaying Majorana bound states, denoted γ_L, at both ends of the nanowire. **(b)** An actual device from Delft University of Technology includes various metallic gates for tuning it to the topological phase by adjusting the nanowire's chemical potential. (Panel a adapted from ref. 3, R. M. Lutchyn, J. D. Sau, S. Das Sarma; panel b adapted from H. Zhang et al., *Nature* **556** 74, 2018.)





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Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

♣ Amorphization of $Bi_{100-x}Sb_x$ films: *Intrinsic* Topological superconductors?

TOPOLOGICAL INSULATORS: Topological materials are insulating in the bulk but conduct an electrical current on the surface or edges.

* One of the first examples of experimentally observed Topological Insulators:

$Bi_{100-x}Sb_x$ ($7 < x < 22$)

TOPOLOGICAL SUPERCONDUCTORS: They have a superconducting energy gap instead of an insulating gap. They are expected to show "Majorana zero modes", very promising for QUANTUM COMPUTING:

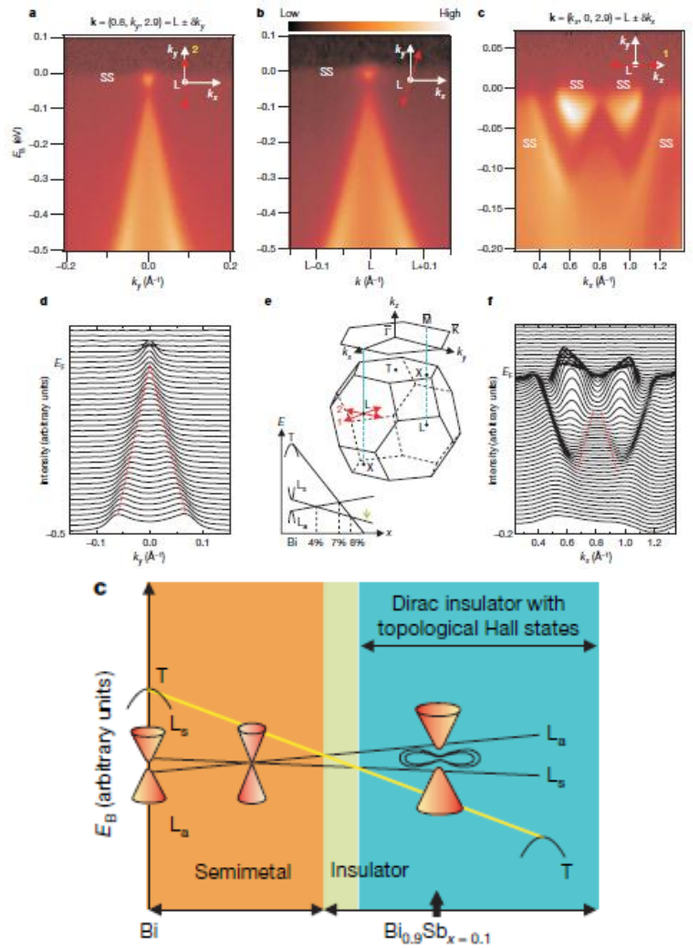
* **$Bi_{100-x}Sb_x$** alloys are **superconducting** only **in amorphous state**!

⇒ **CHALLENGE: To amorphize $Bi_{100-x}Sb_x$ crystals or films by ion irradiation**

A topological Dirac insulator in a quantum spin Hall phase

D. Hsieh[1], D. Qian[1], L. Wray[1], Y. Xia[1], Y. S. Hor[2], R. J. Cava[2] & M. Z. Hasan[1,3]

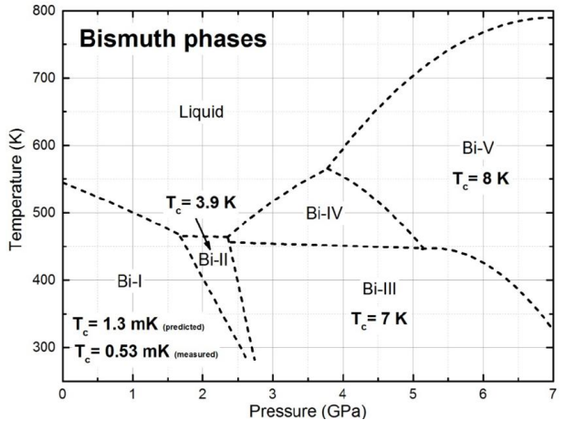
Vol 452|24 April 2008|doi:10.1038/nature06843



Introduction and Background

Amorphization of $Bi_{100-x}Sb_x$

Quest for amorphous superconductors of Bi-Sb …



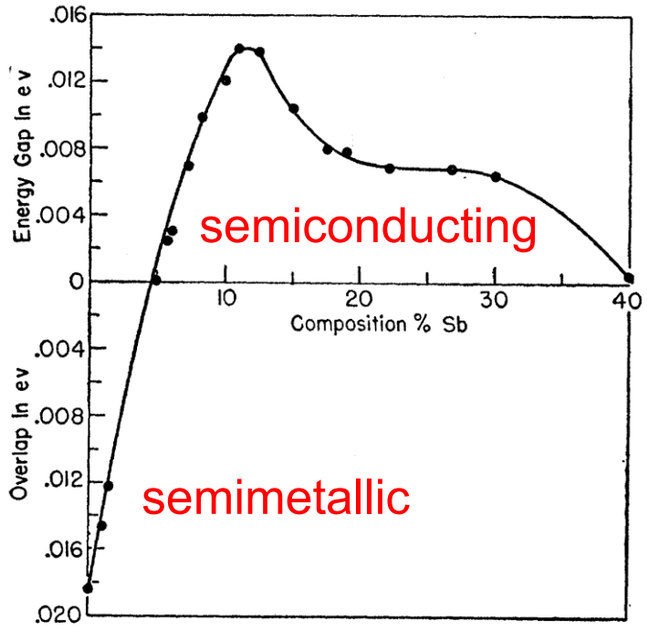


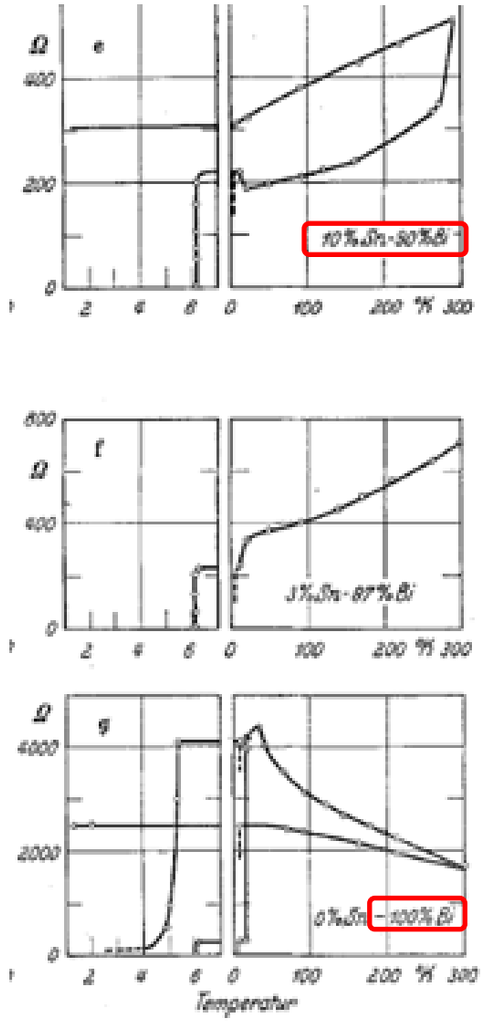
FIG. 8. Activation energy Eg *vs* concentration of Sb.

Zeitschrift für Physik, Bd. 146, S. 27—38 (1956)

Supraleitung und elektrischer Widerstand neuartiger Zinn-Wismut-Legierungen

Von

W. BUCKEL und R. HILSCH



PHYSICAL REVIEW VOLUME 147, NUMBER 1 8 JULY 1966

Superconducting Transitions of Amorphous Bismuth Alloys*†

J. S. SHIER‡ AND D. M. GINSBERG§
Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois

(Received 20 December 1965)

Sample	At.% impurity	Thickness (Å)	Resistivity ($\mu\Omega$ cm)	T_c °K	Transition width (mdeg)
a	0	750±40	125±7	6.154	5.8
b	0[a]	690±40	96±6	6.173	4.6
c	0.53 Tl	630±40	101±6	6.164	4.9
d	1.02 Tl	610±40	114±7	6.158	6.0
e	1.02[a] Tl	...	...	6.173	4.5
f	2.05 Tl	550±40	97±7	6.167	5.7
g	3.32 Tl	820±40	134±7	6.181	5.6
h	6.15 Tl	...	...	6.205	6.0
i	9.52 Tl	550±40	114±8	6.220	5.6
j	13.16 Tl	730±40	114±8	6.253	...
k	1.03 Pb	920±40	144±6	6.218	5.2
l	2.02 Pb	740±40	129±7	6.261	6.3
m	3.08 Pb	750±40	130±7	6.292	5.3
n	5.09 Pb	700±40	118±7	6.374	6.2
o	1.08 Sb	890±20	141±3	6.142	5.1
p	1.08 Sb	480±20	141±6	6.092	9.5
q	2.95 Sb	1070±20	144±3	6.125	4.5
r	5.02 Sb	540±20	125±5	6.049	6.5
s	7.96 Sb	1130±20	138±2	6.032	5.0

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Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

Amorphization of $Bi_{100-x}Sb_x$

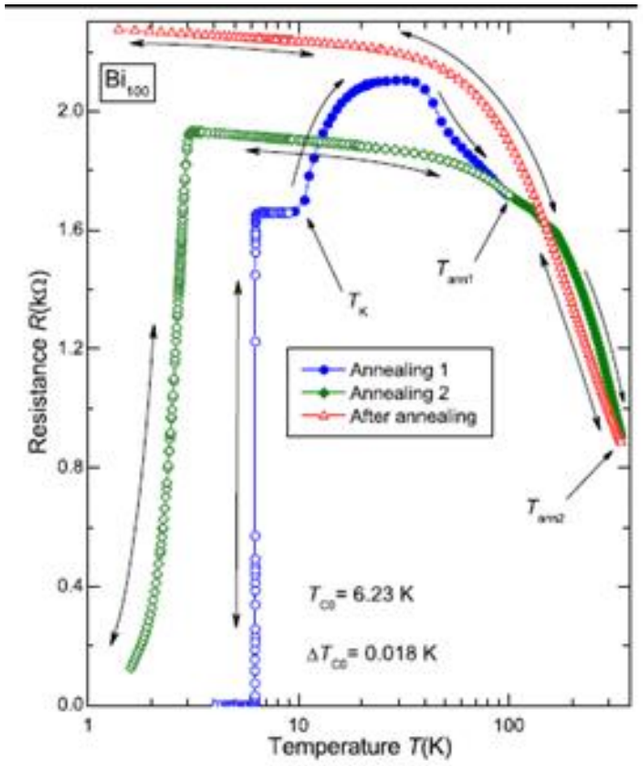
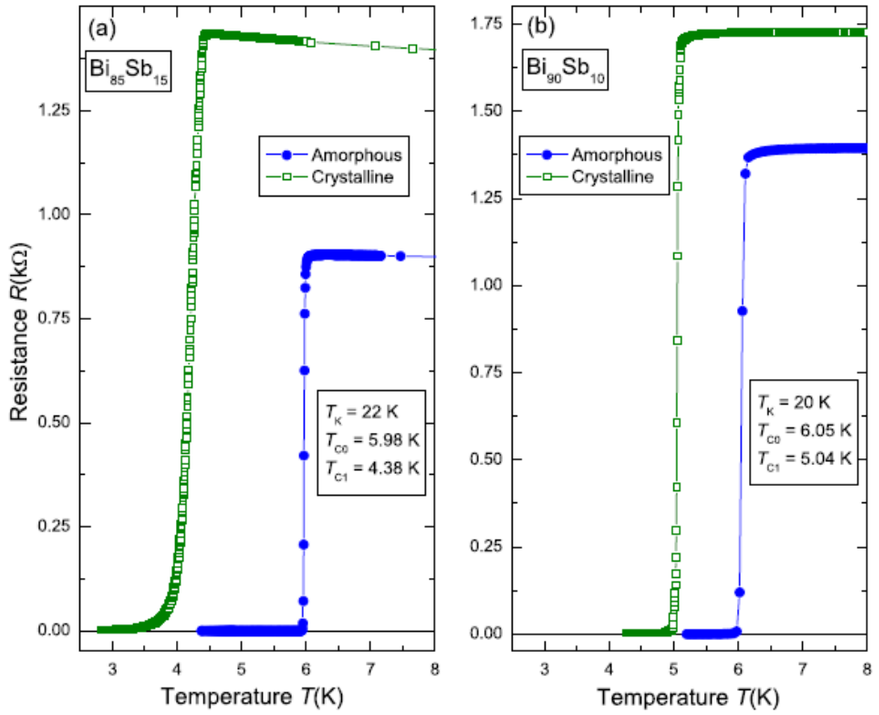


Figure 1. Resistance of the *in-situ* prepared Bi film, measured immediately after preparation and during annealing. Symbols: ⟶ irreversible changes; ⟷ reversible changes of the resistance. The experimental data plotted with open symbols are after annealing and refer to the reversible state.

IOP Publishing — Superconductor Science and Technology
Supercond. Sci. Technol. **30** (2017) 015013 (9pp) — doi:10.1088/0953-2048/30/1/015013

Superconductivity in the amorphous phase of topological insulator Bi_xSb_{100-x} alloys

J Barzola-Quiquia[1,2], C Lauinger[3], M Zoraghi[1], M Stiller[1], S Sharma[4] and P Häussler[2]



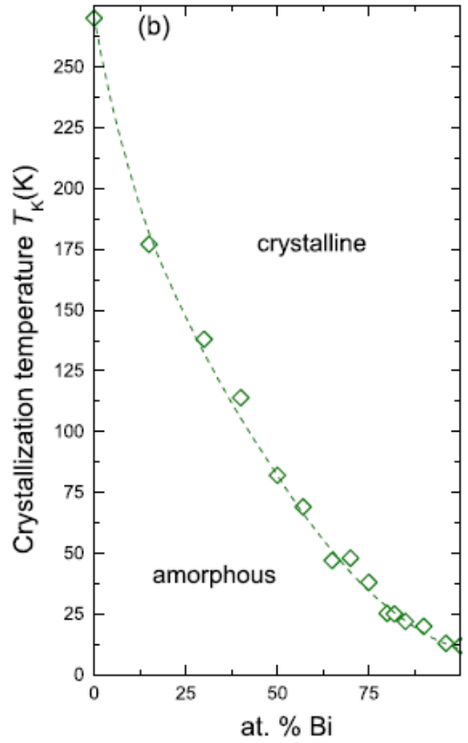


Figure 3. Resistance of (a) $Bi_{85}Sb_{15}$ and (b) $Bi_{90}Sb_{10}$ films. A superconducting transition in the amorphous state and after crystallization is visible. The crystalline phase was obtained after annealing the sample up to $T = 60$ K.

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Introduction and Background

Quest for amorphous superconductors of Bi-Sb …

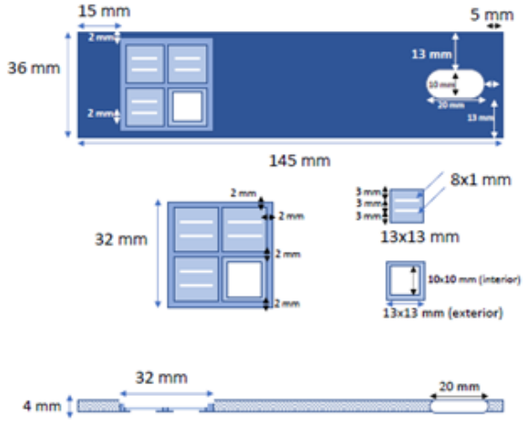
⇒ **<u>CHALLENGE</u>: To amorphize $Bi_{100-x}Sb_x$ polycrystalline samples by ion-beam irradiation**

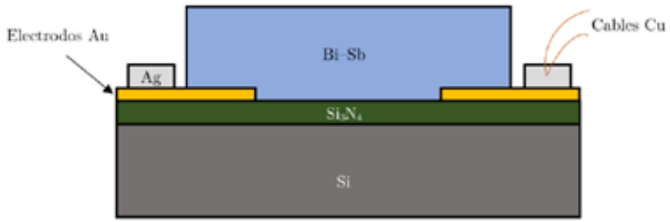
Experimental Techniques

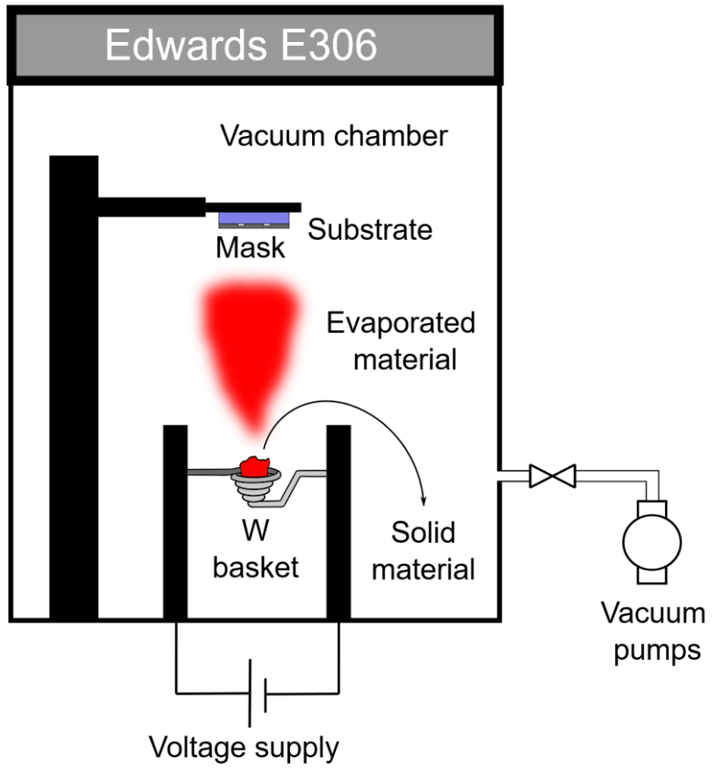
Quest for amorphous superconductors of Bi-Sb …

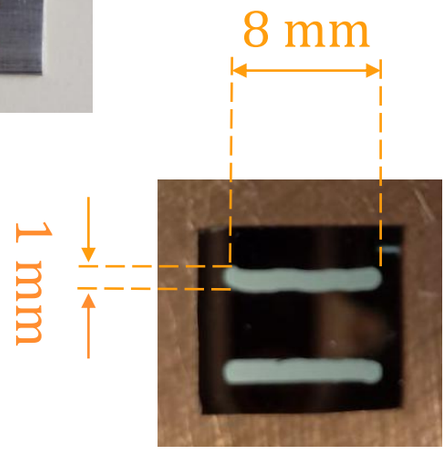
Growth by thermal evaporation

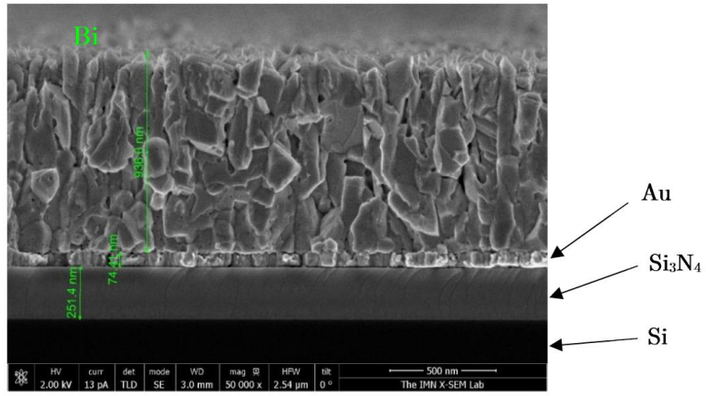
High purity Bi/Sb
(99.995 %)











→ ***Trying different substrates, evaporation procedures, etc.***

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Experimental Techniques

Quest for amorphous superconductors of Bi-Sb …

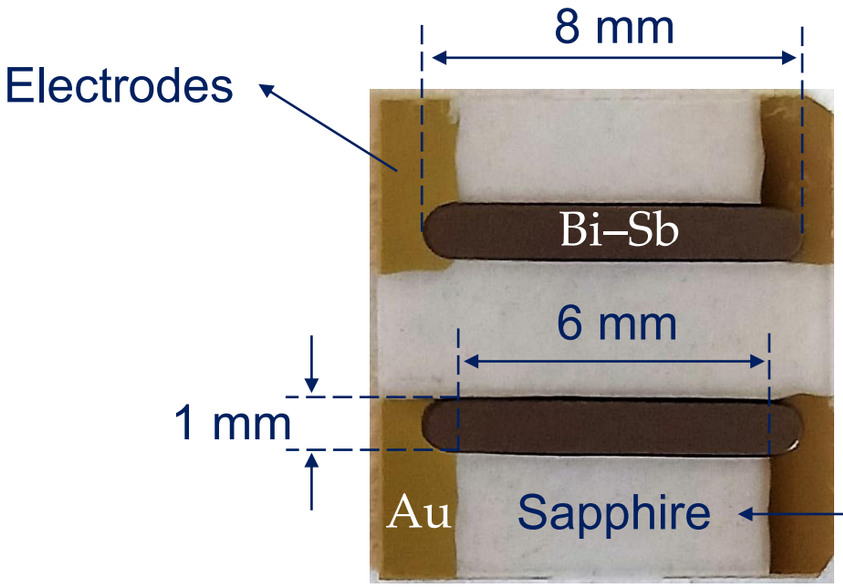
Structural and morphological characterization

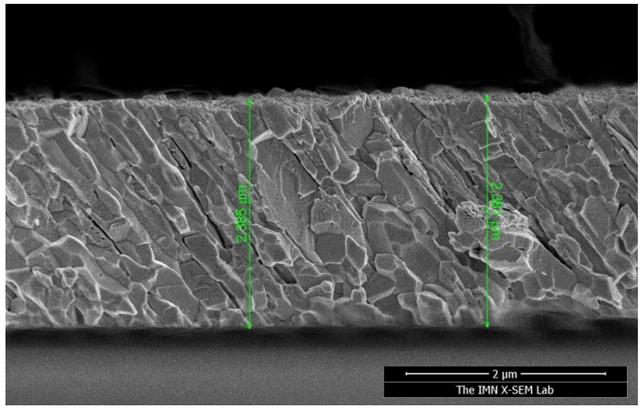
$Bi_{100-x}Sb_x$
~10 μm - thick crystalline films

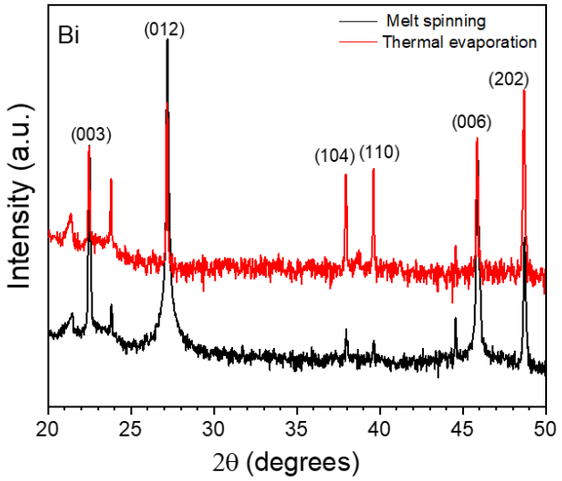
x = 0, 5, 10, 15

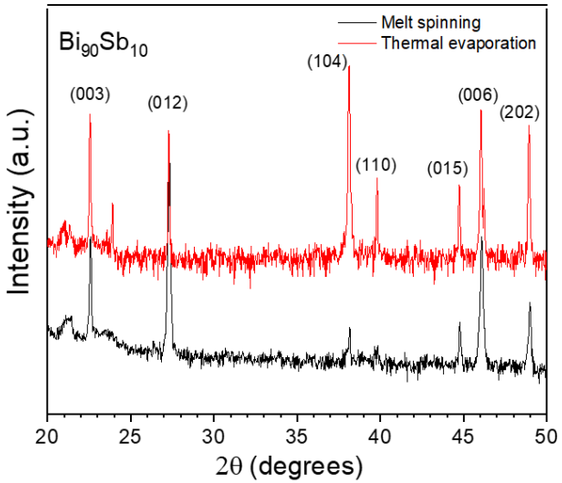
Characterization by:

- Profilometry
- Scanning Electron Microscopy (SEM) + EDX
- X-ray diffraction









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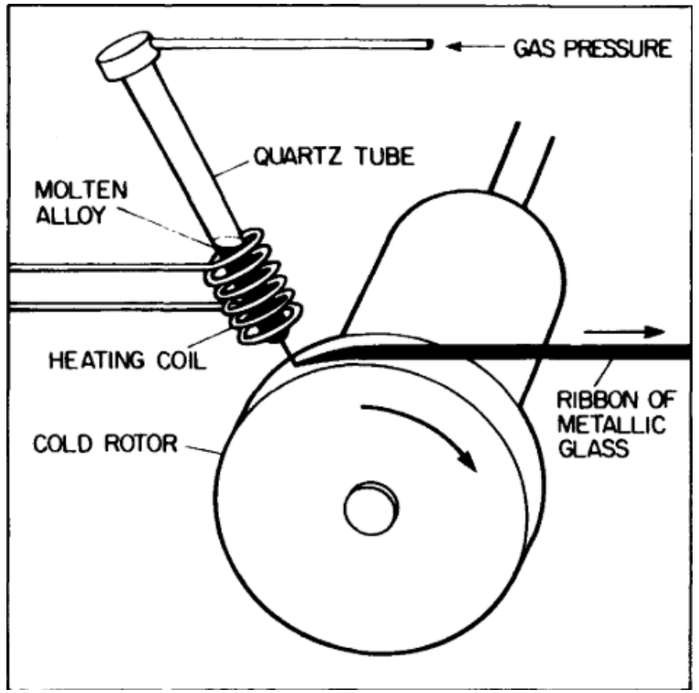
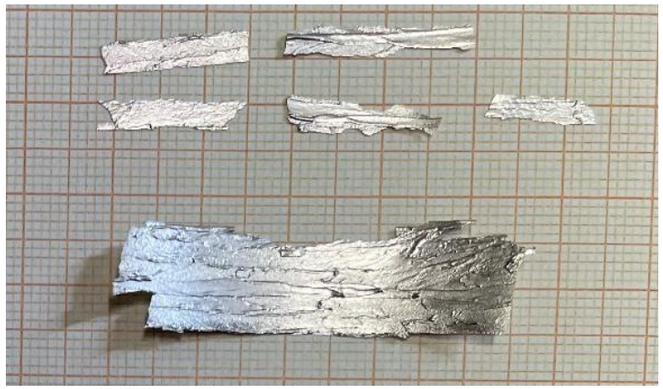
Experimental Techniques

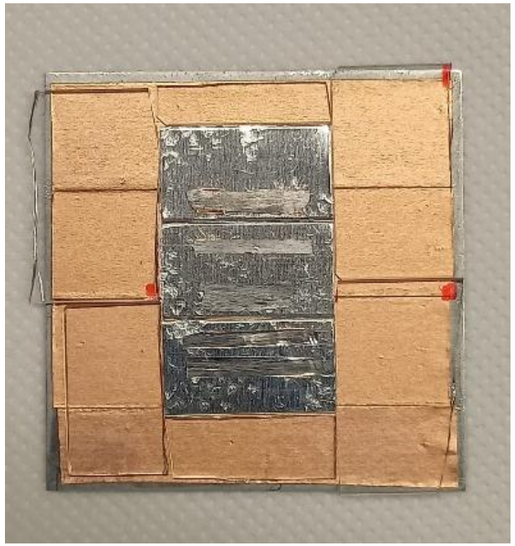
Quest for amorphous superconductors of Bi-Sb …

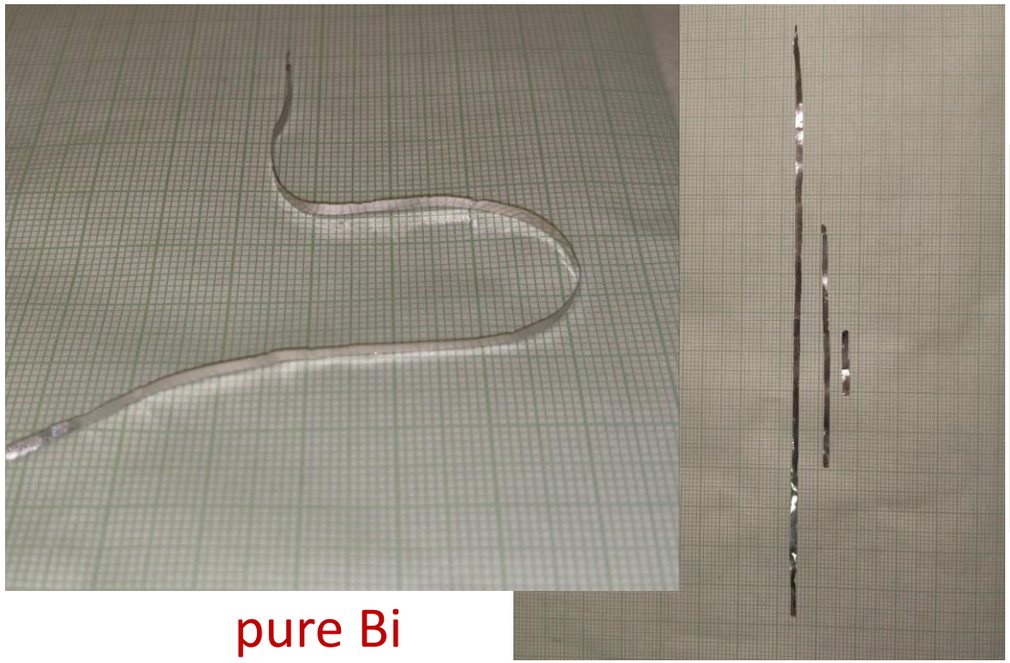
Growth by melt spinning



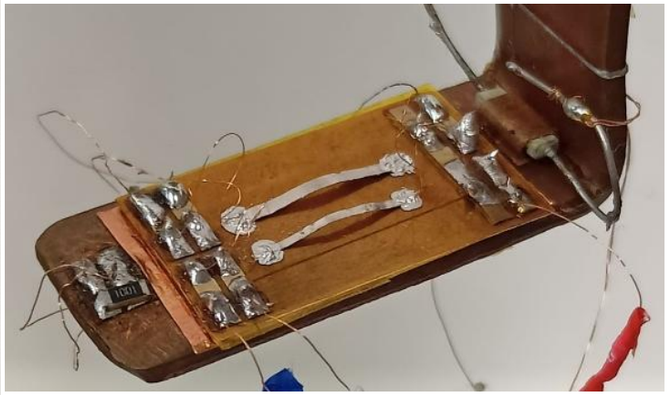
Bi-Sb







pure Bi



Vicente Madurga
Cristina Favieres
José Vergara

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Universidad Pública de Navarra
Nafarroako Unibertsitate Publikoa
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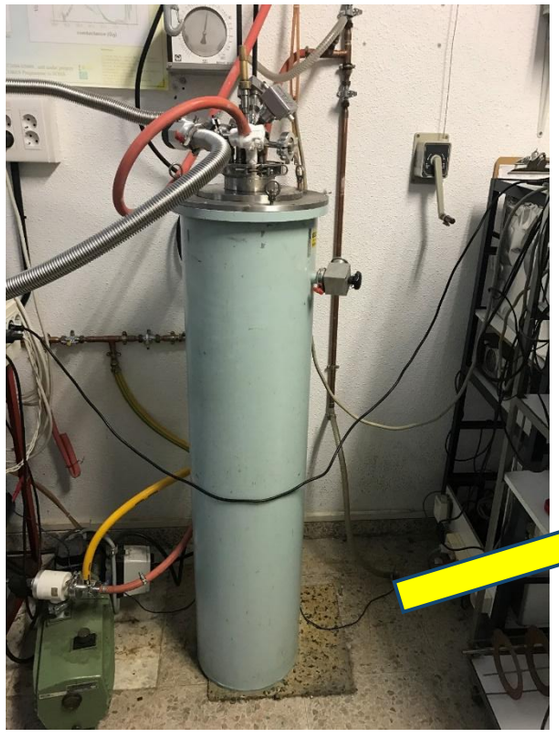
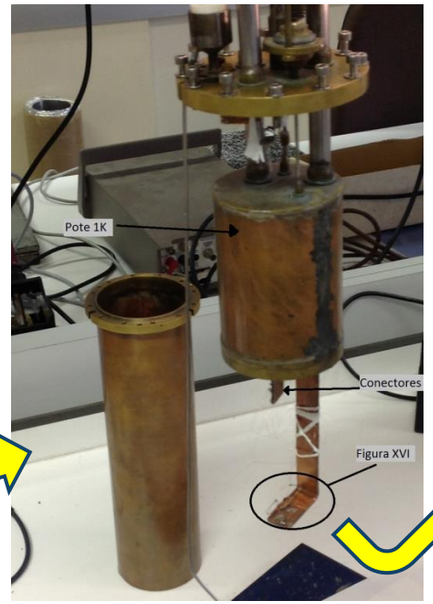
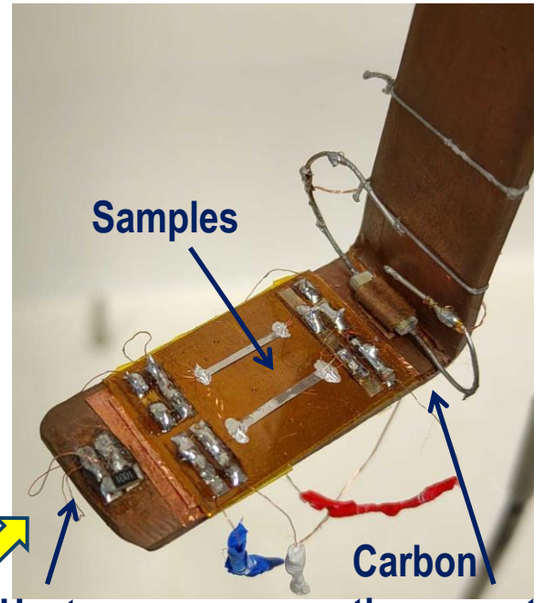
Experimental Techniques

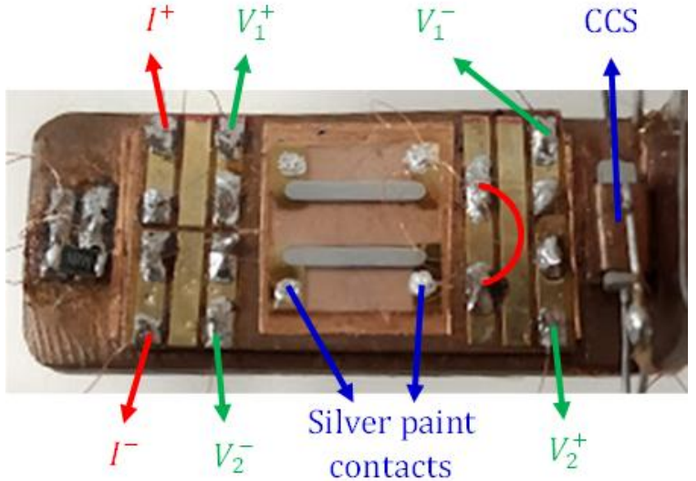
Quest for amorphous superconductors of Bi-Sb …

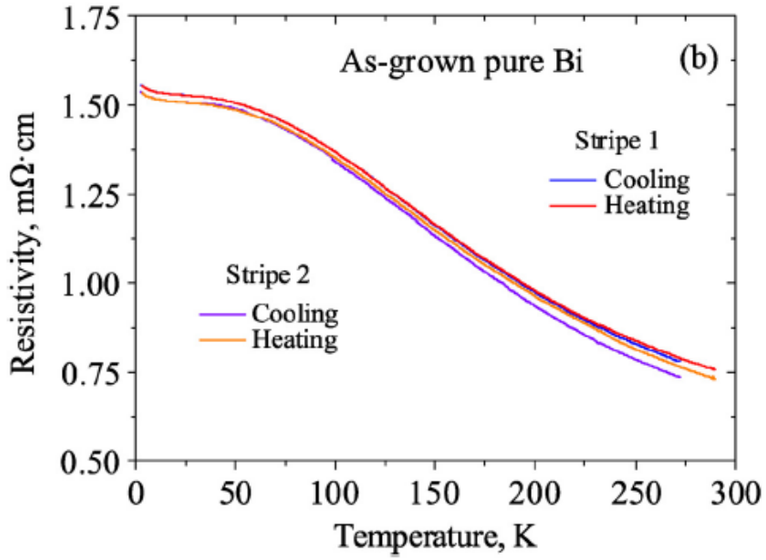
Low-temperature electrical resistivity











[Andrino-Gómez *et al.*, Low Temp. Phys. **50**, 389 (2024)]

- ^{4}He cryostat (with a “1K pot”)
- T ~ 2 K – 300 K
- P <10^{-7} mbar
- 4-point resistance measurements

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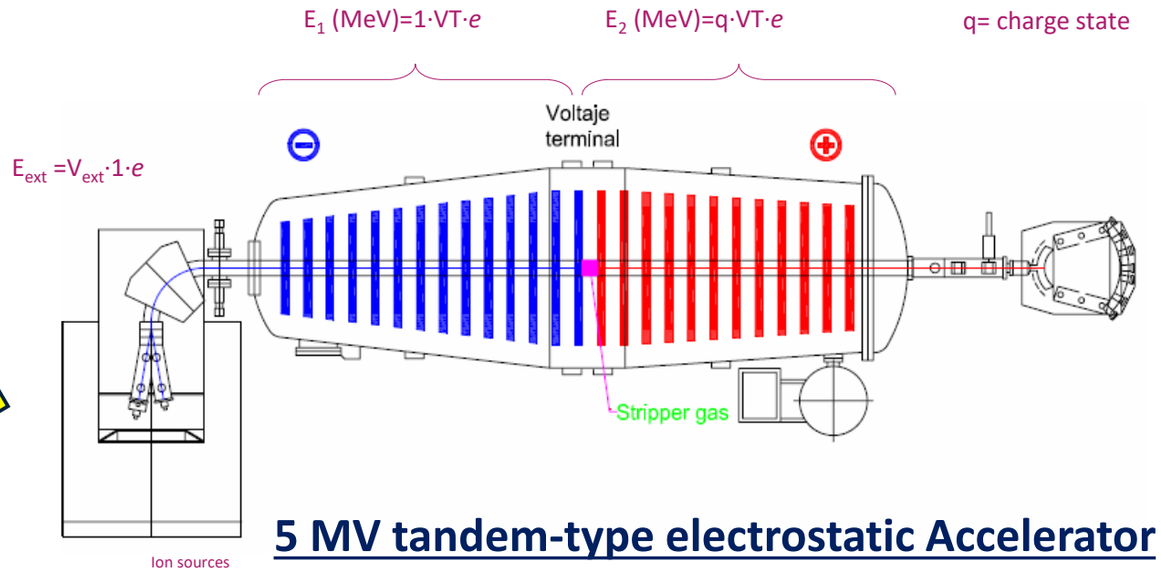
Experimental Techniques

Quest for amorphous superconductors of Bi-Sb …

= **Center for Micro-Analysis of Materials (@ UAM)**

Ion beam accelerator

$E_{max}=E_{ext}+VT\cdot(1+q)\cdot e$ → **Energy: 10´s of MeV**



Two ion sources:

- Duoplasmatron: H, He
- Sputtering: Solid targets, many beam species used & molecular beams

- Unique energy range in Iberian peninsula
- High stability (ripple below 10^{-5})

5 MV tandem-type electrostatic Accelerator

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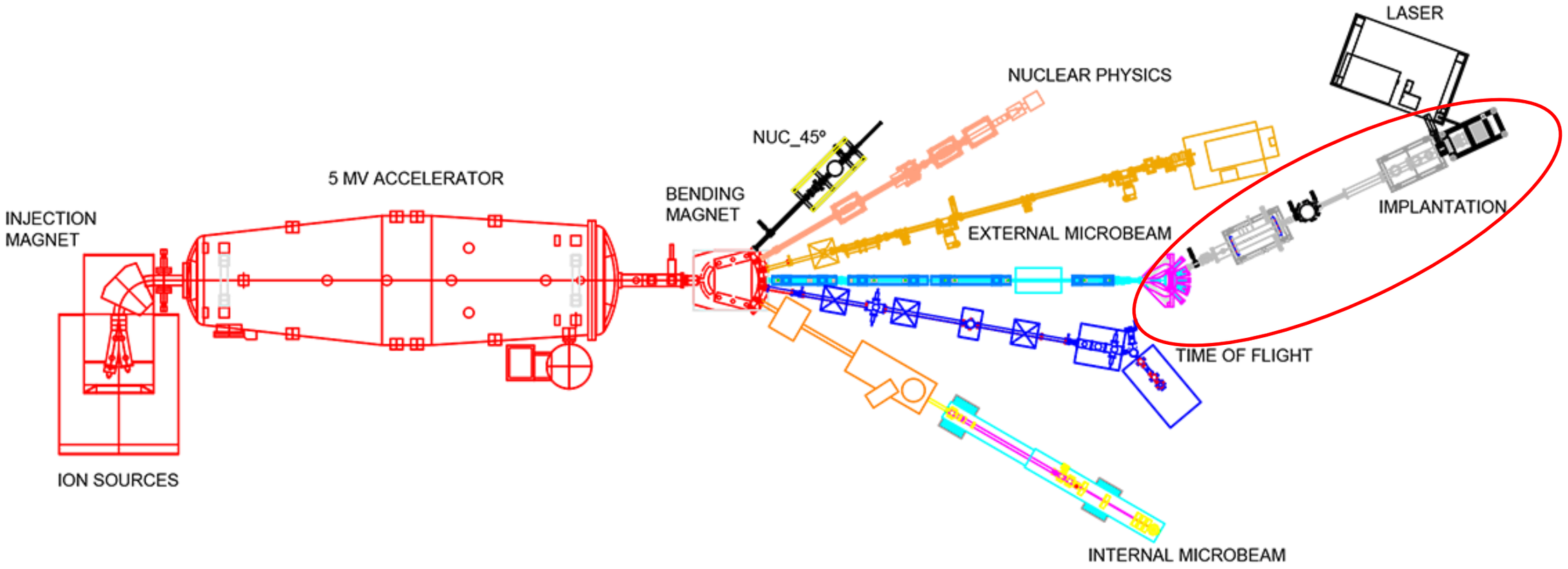
Experimental Techniques

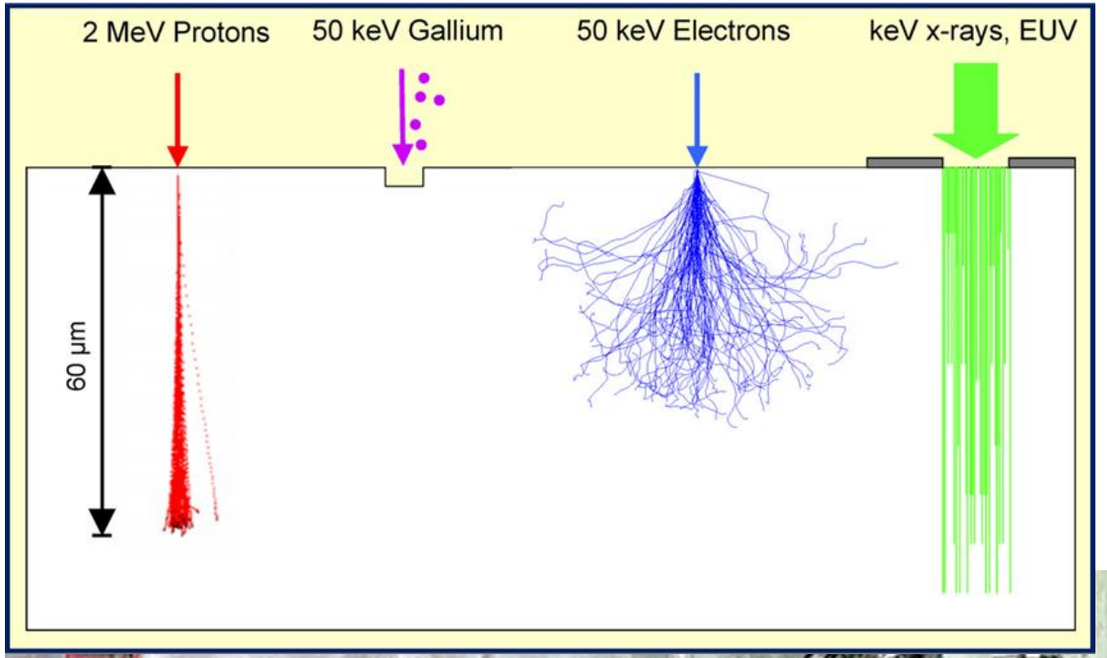
Quest for amorphous superconductors of Bi-Sb …

Ion beam accelerator

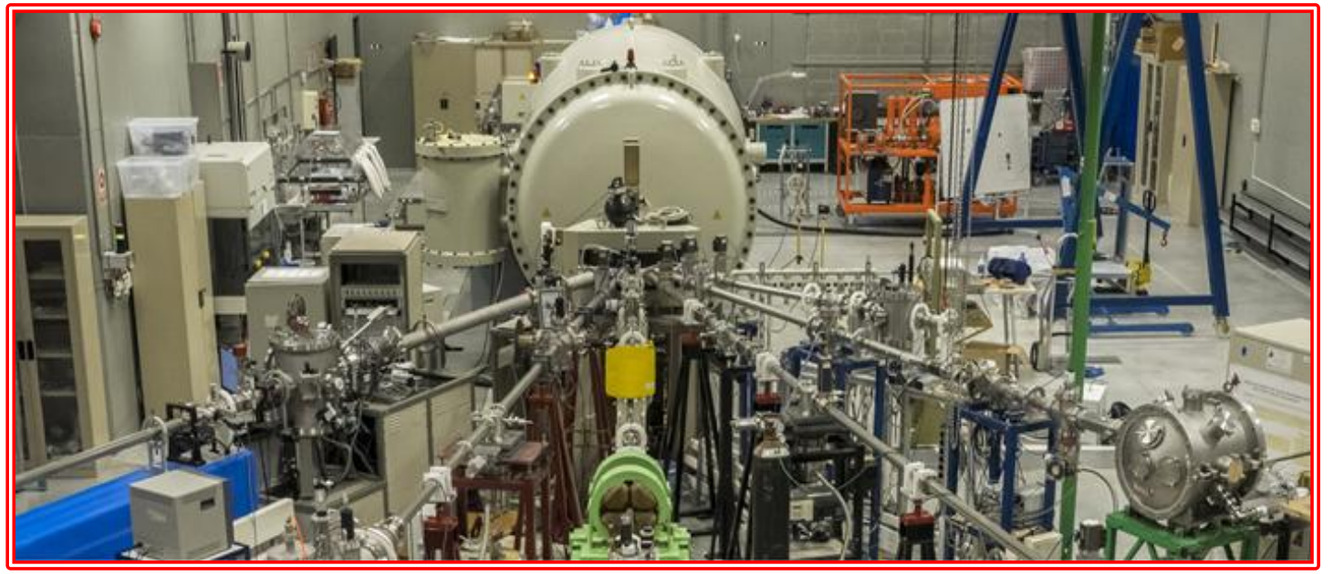


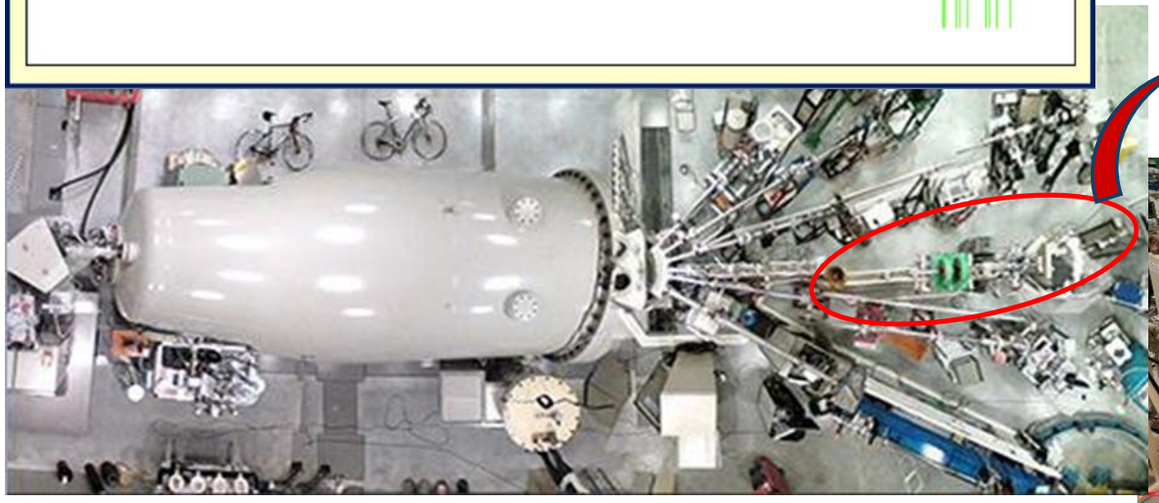
Experimental Techniques

Quest for amorphous superconductors of Bi-Sb …

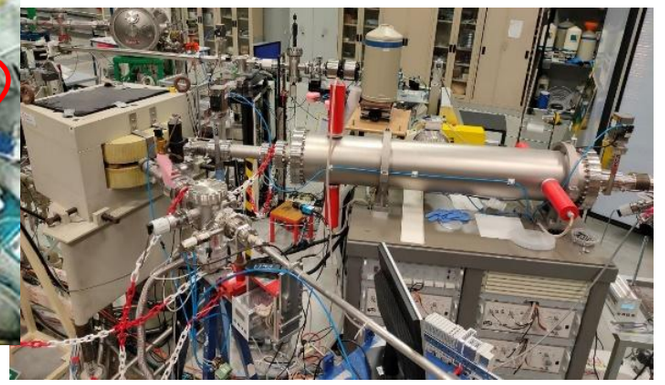


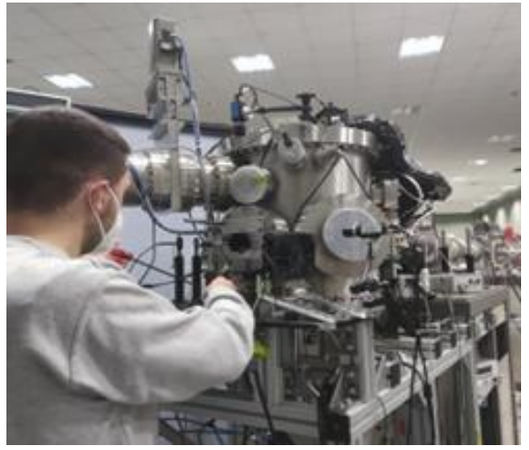
Ion beam accelerator





IMPLANTATION LINE





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Experimental Techniques

Quest for amorphous superconductors of Bi-Sb …

Ion beam accelerator

Ion Beam Analysis (IBA)

Ion Beam Modification of Materials (IBMM)

Experimental Techniques

Quest for amorphous superconductors of Bi-Sb …

Ion Beam Analysis (IBA)

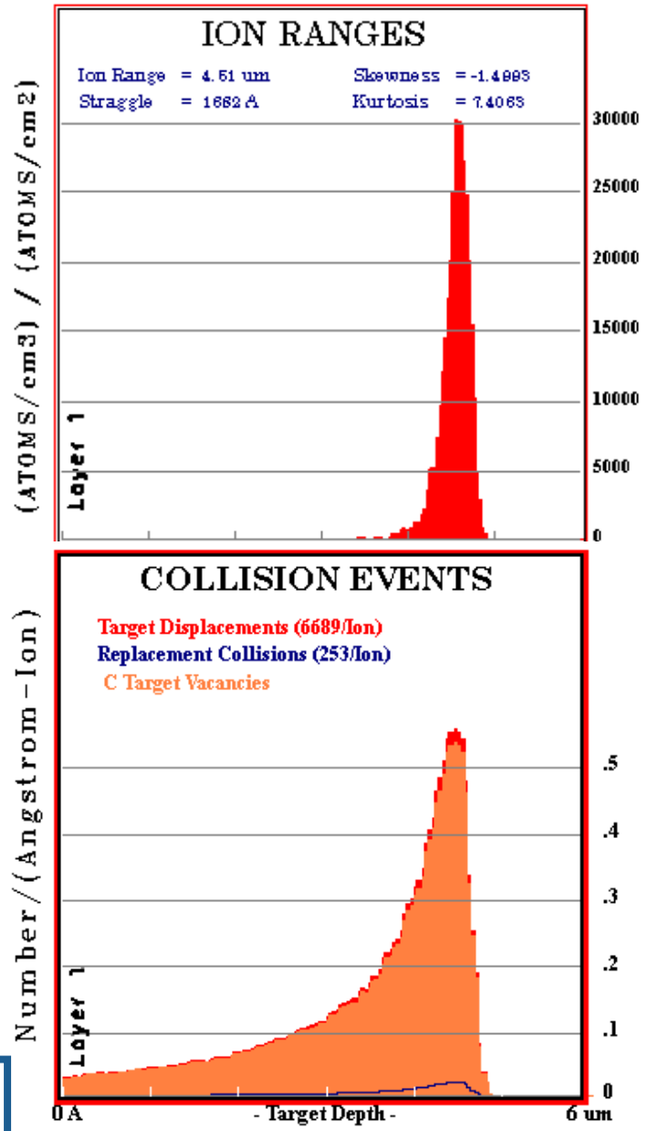
Ion Beam Modification of Materials (IBMM)

- Electronic and nuclear stopping force

  [→ **SRIM simulations**] *
- Non-trivial variation vs. depth (Bragg peak), μm scale
- Area affected by single ion has nm lateral dimensions
- Choose species, energy, fluence, dose rate

* http://www.srim.org/

Ion beam accelerator



35 MeV [Br^+] / diamond

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Experimental Techniques

Quest for amorphous superconductors of Bi-Sb …

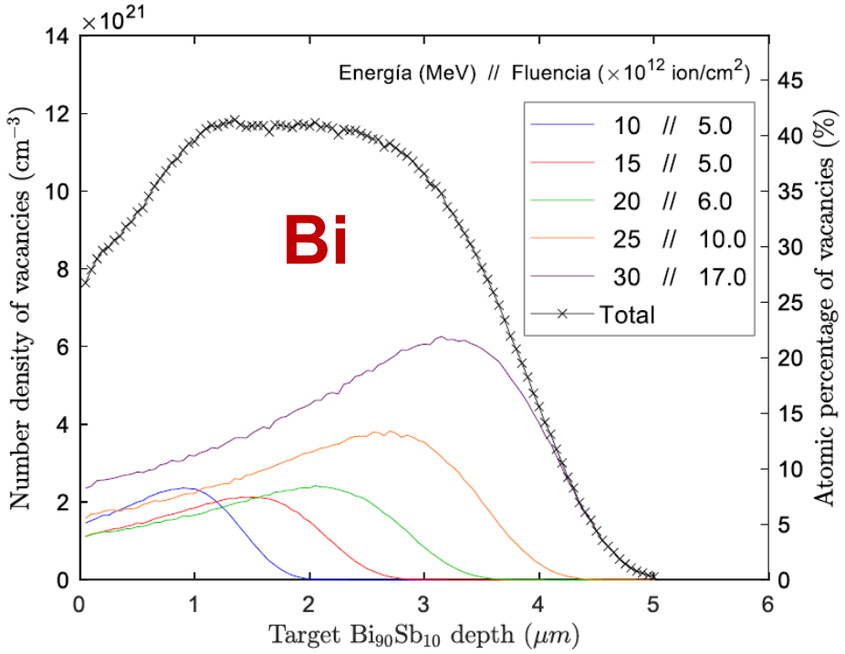
Objectives and working plan

♣ Explore the possibility of obtaining (robust) subsurface **amorphous** (⇒ **superconducting**) regions in Bi and Bi-Sb alloys by irradiation with Swift heavy ions

♣ Prepare (polycrystalline) samples of $\mathbf{Bi_{100-x}Sb_x}$ (x = 0–15) by different methods

♣ Irradiate ~1 cm^2 samples at CMAM (mainly by **10-40 MeV Bi** or **I** ions)

♣ Characterize samples before and after irradiation by:
- SEM, EDX, XRD, profilometry
- **electrical conductivity (2–300 K)**



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Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

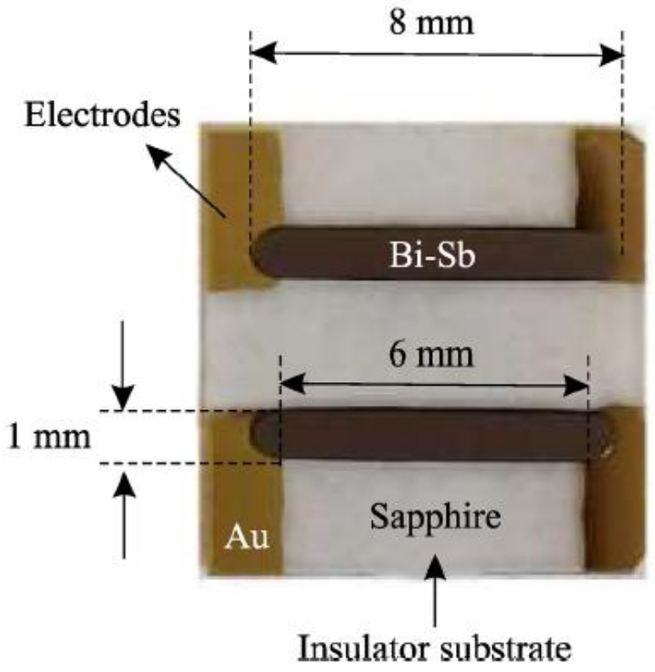
Films by thermal evaporation

Low Temperature Physics ARTICLE pubs.aip.org/aip/ltp

Low-temperature electrical conductivity of ion-beam irradiated Bi-Sb films

Cite as: Fiz. Nizk. Temp. **50**, 427–433 (May 2024); doi: 10.1063/10.0025622
Submitted: 26 March 2024

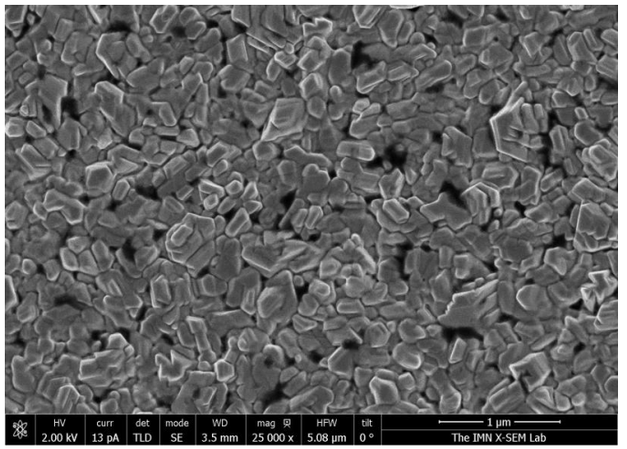
A. Andrino-Gómez,[1,2,3,4] M. Moratalla,[1,3,4] A. Redondo-Cubero,[2,3,4] N. Gordillo,[2,3,4] and M. A. Ramos[1,3,4,a)]



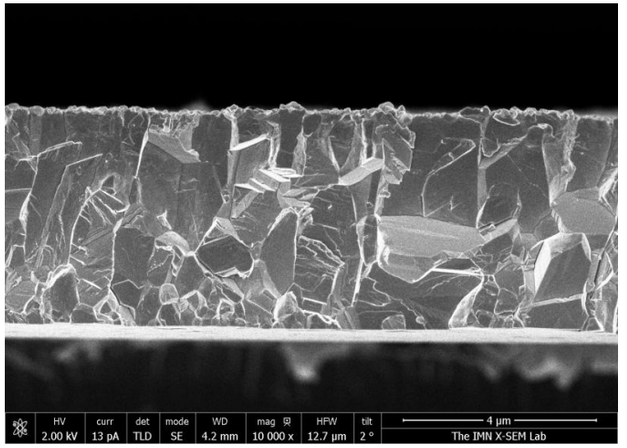
$Bi_{100-x}Sb_x$
~10 μm - thick crystalline films

x = 0, 5, 10, 15

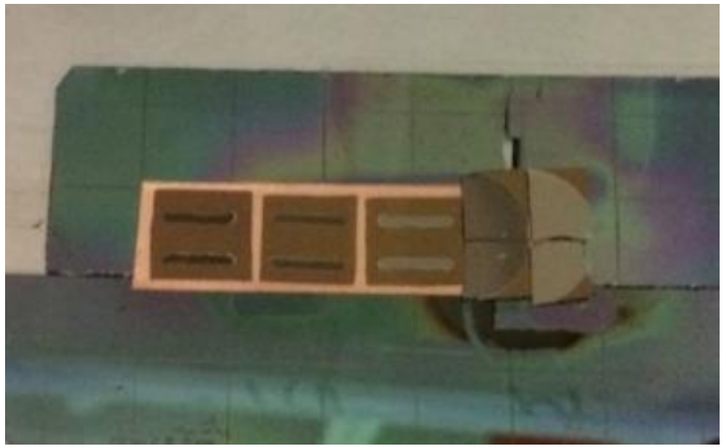
top view



cross section



→ Irradiation with Bi ions



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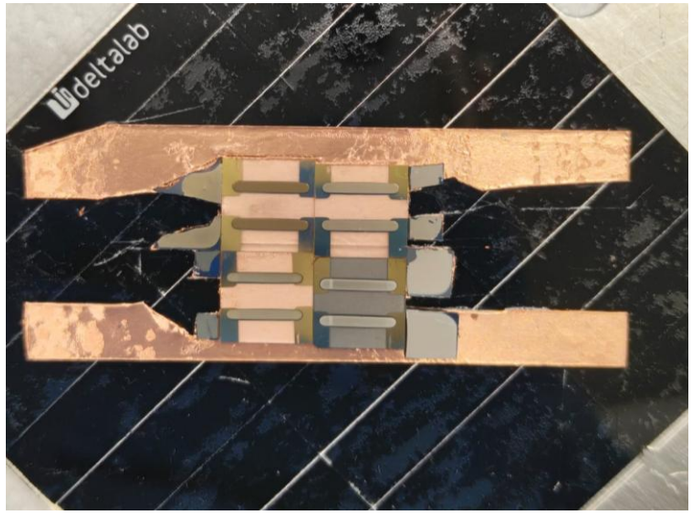
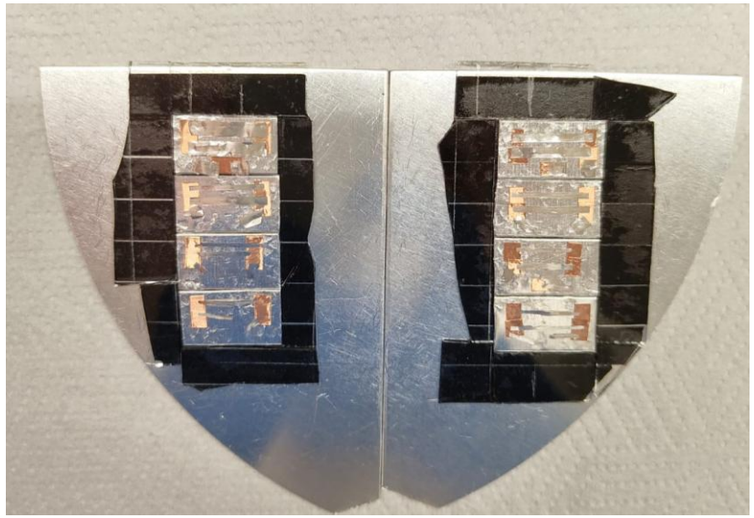
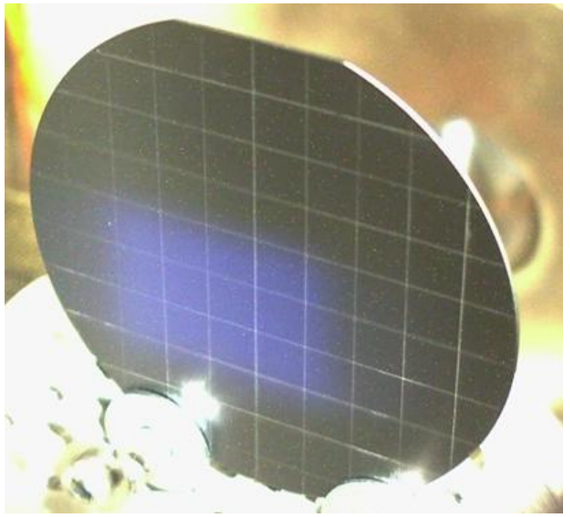
Experimental Results (up to now)

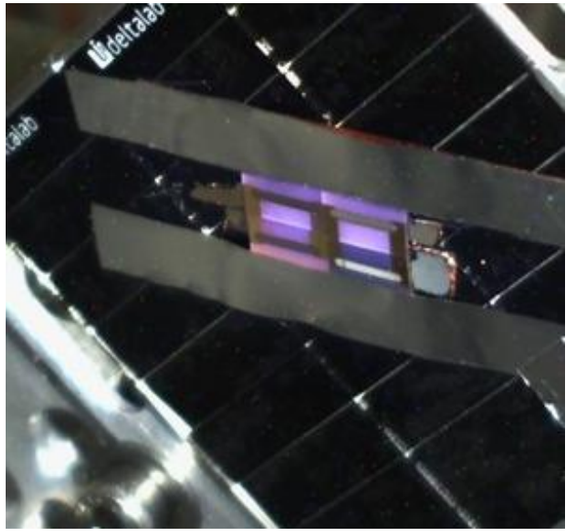
Quest for amorphous superconductors of Bi-Sb …

Films by thermal evaporation









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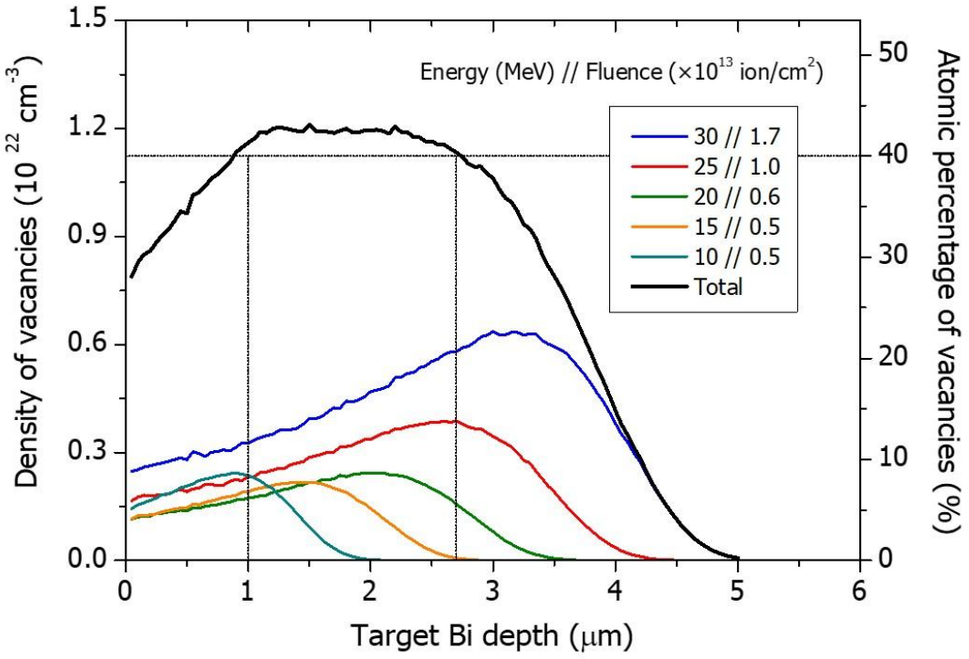
Experimental Results (up to now)

Films by thermal evaporation

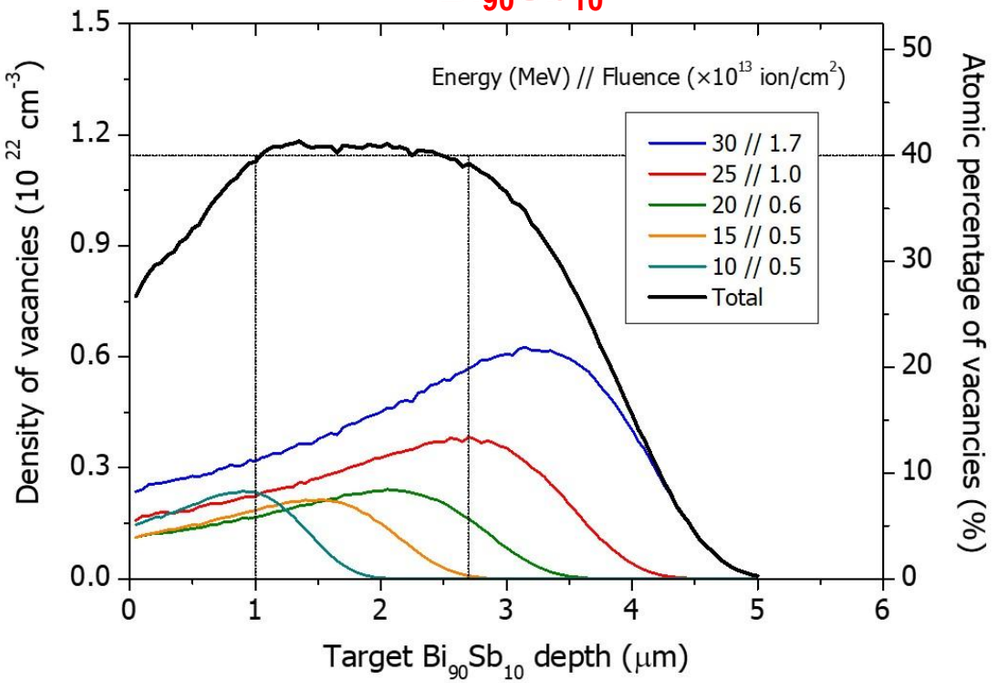
Quest for amorphous superconductors of Bi-Sb …

- Irradiation of evaporated films with **10–30 MeV Bi** self-ions
- Nominal **40% atomic vacancies** in the range **≈1–3 μm**

Bi



$Bi_{90}Sb_{10}$



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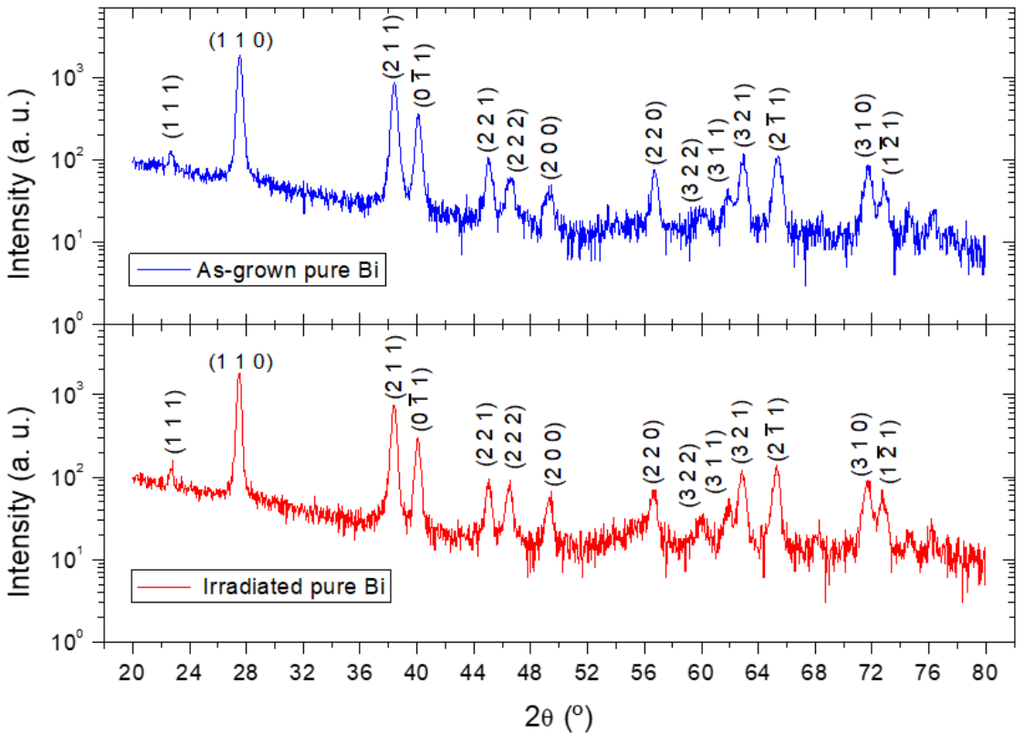
Experimental Results (up to now)

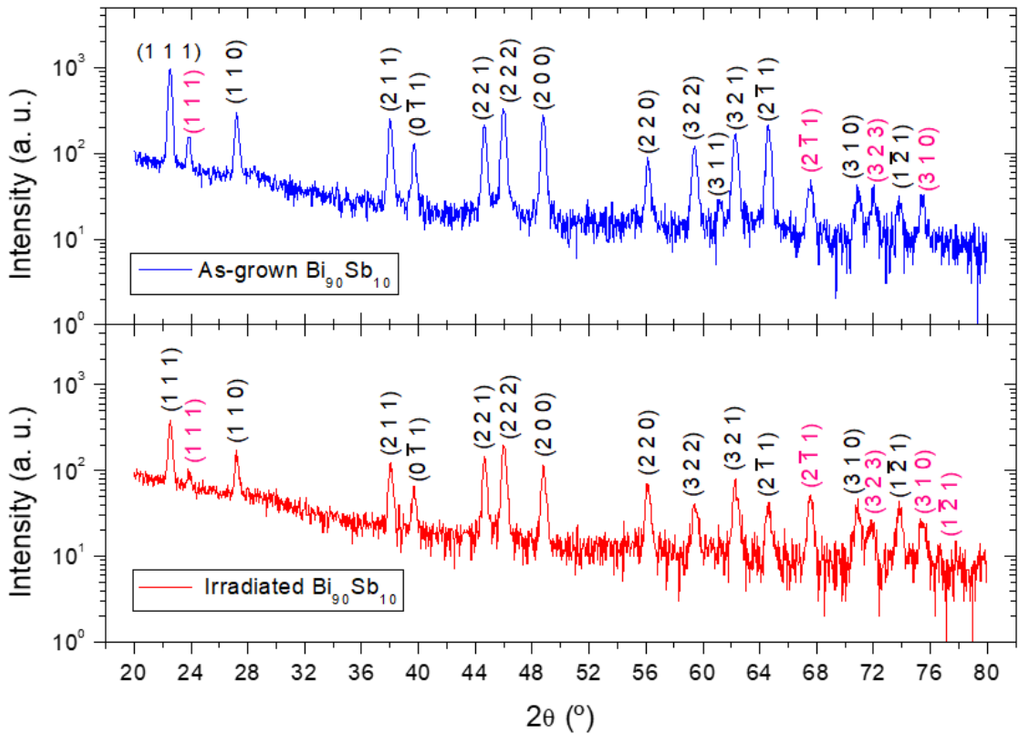
Quest for amorphous superconductors of Bi-Sb …

Films by thermal evaporation

X-ray diffraction



Bi

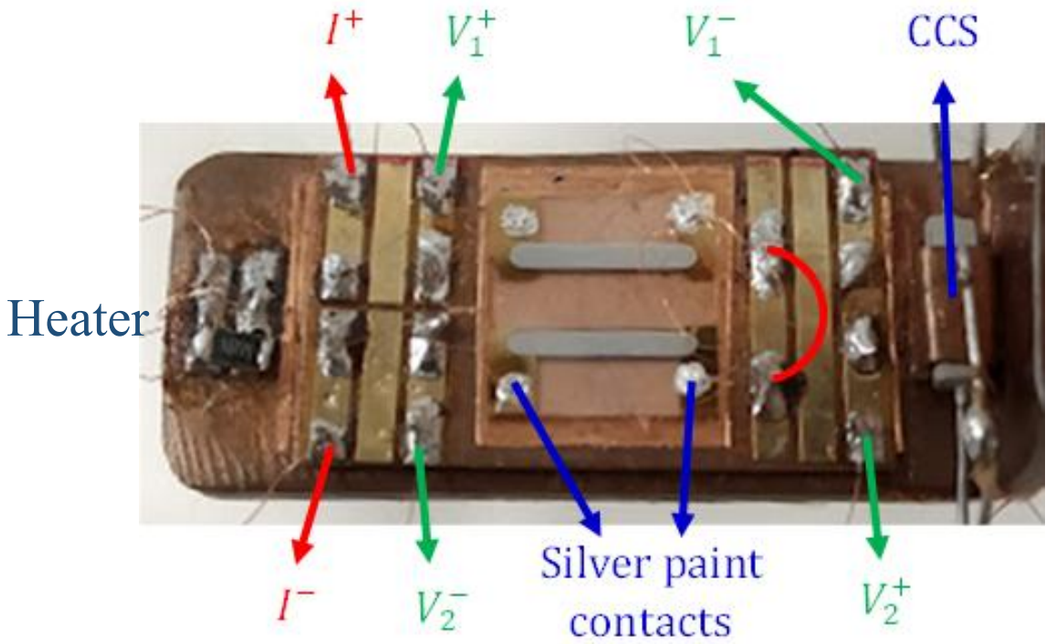


$Bi_{90}Sb_{10}$

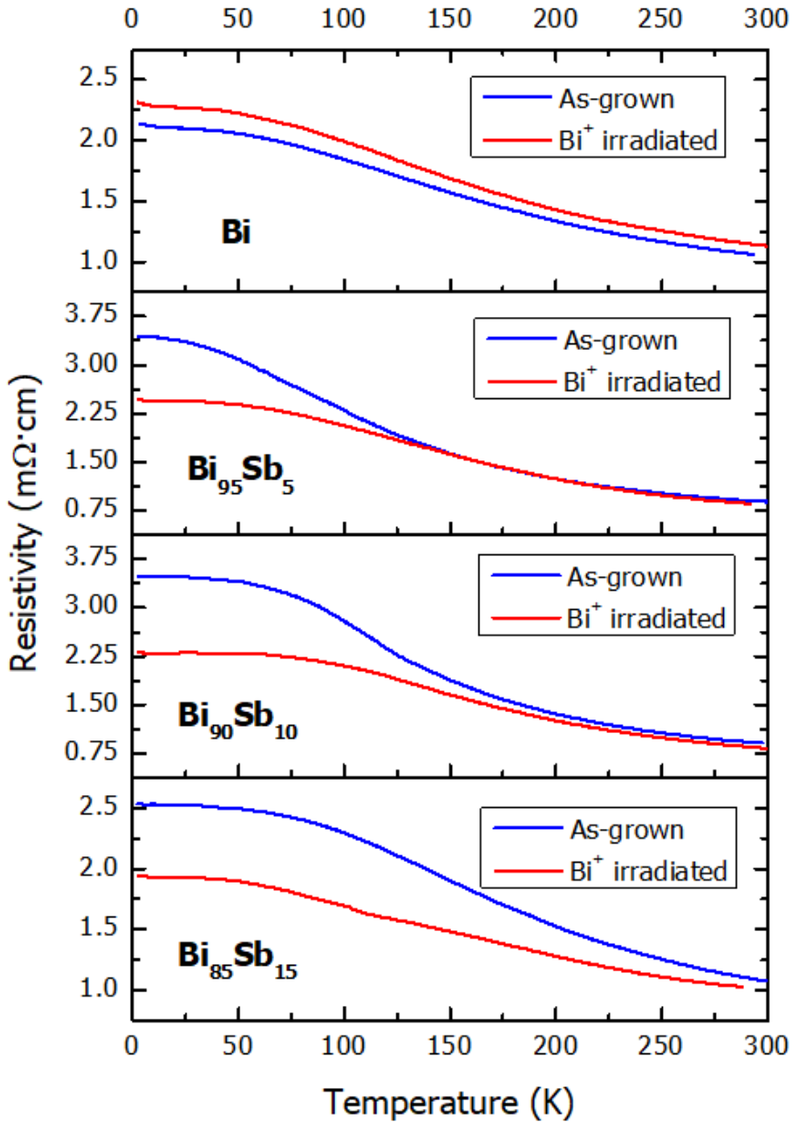
Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Electrical resistivity



Films by thermal evaporation

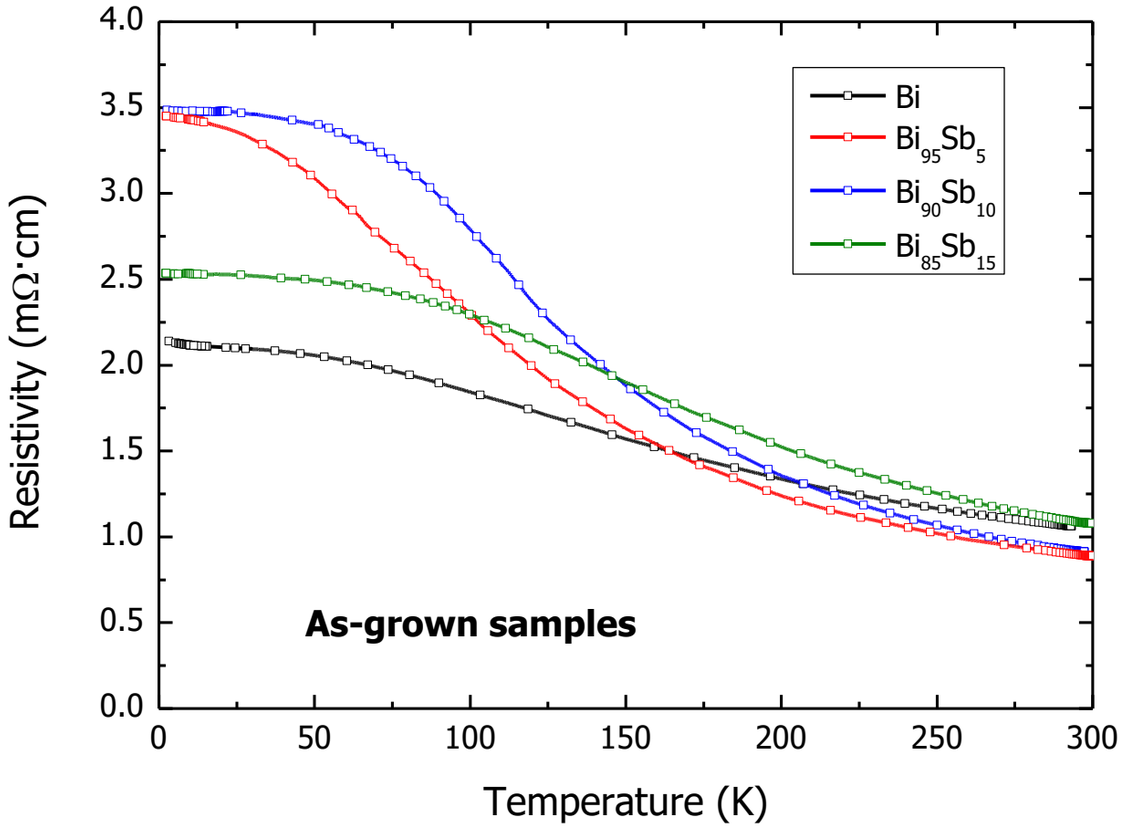


Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Films by thermal evaporation

Electrical resistivity



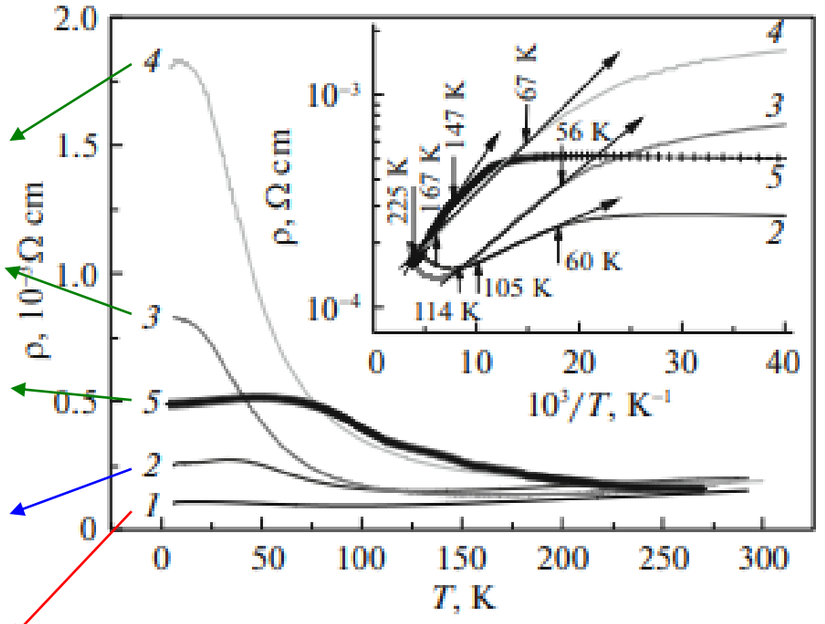
$Bi_{83}Sb_{17}$ (d = 900 nm)

$Bi_{84}Sb_{16}$ (t = 23 μm)

$Bi_{83}Sb_{17}$ (d = 200 nm)

$Bi_{91}Sb_{9}$ (t = 27 μm)

$Bi_{97}Sb_{3}$ (t = 12 μm)



[A. Nikolaeva *et al*. 2019]

1, 2, 3: foils.

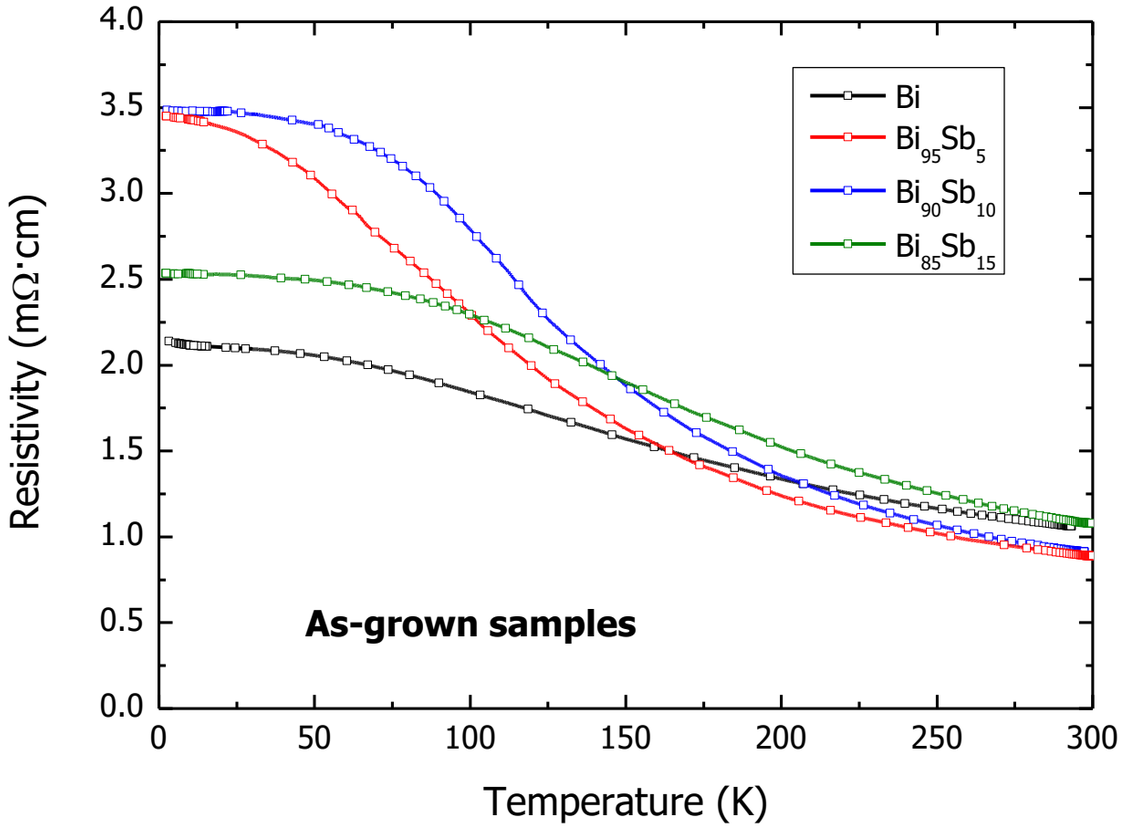
4, 5: wires.

Experimental Results (up to now)

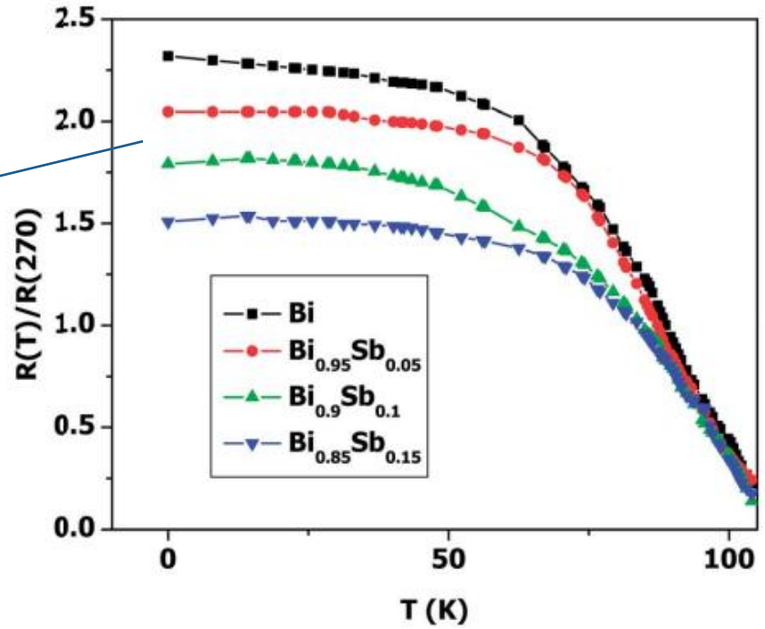
Quest for amorphous superconductors of Bi-Sb …

Films by thermal evaporation

Electrical resistivity



Nanowires:
t = 40 nm



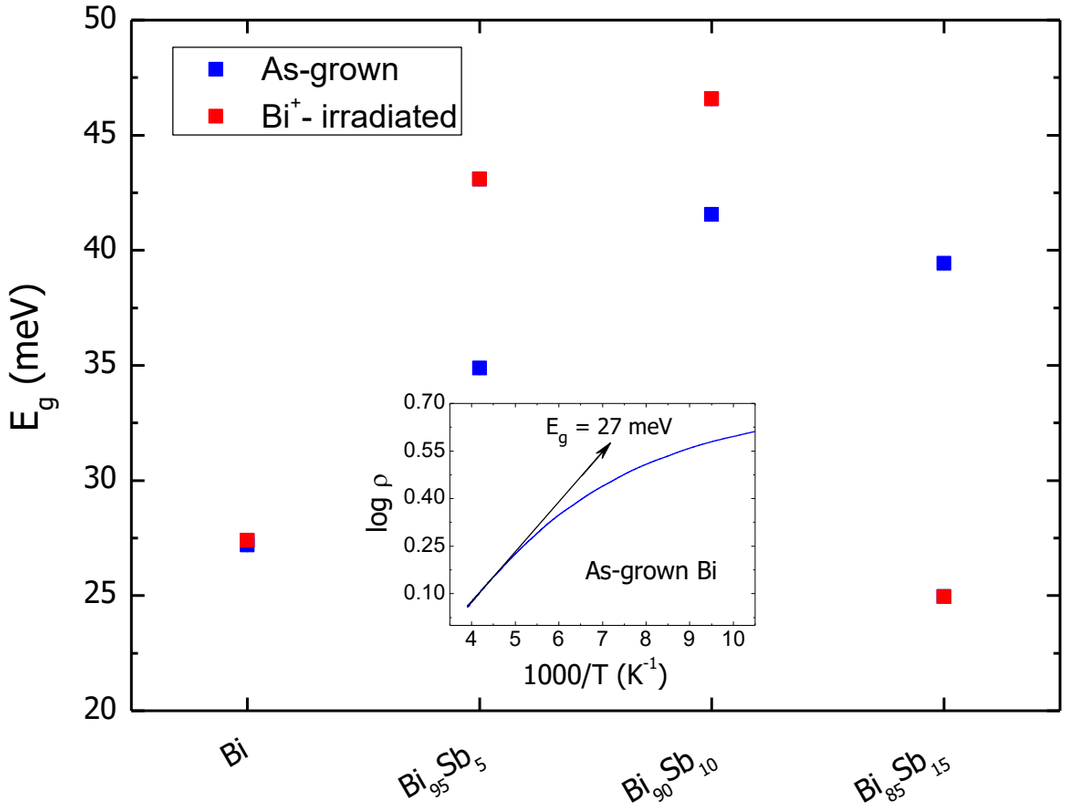
[S. Tang and M. S. Dresselhaus 2014]

Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Films by thermal evaporation

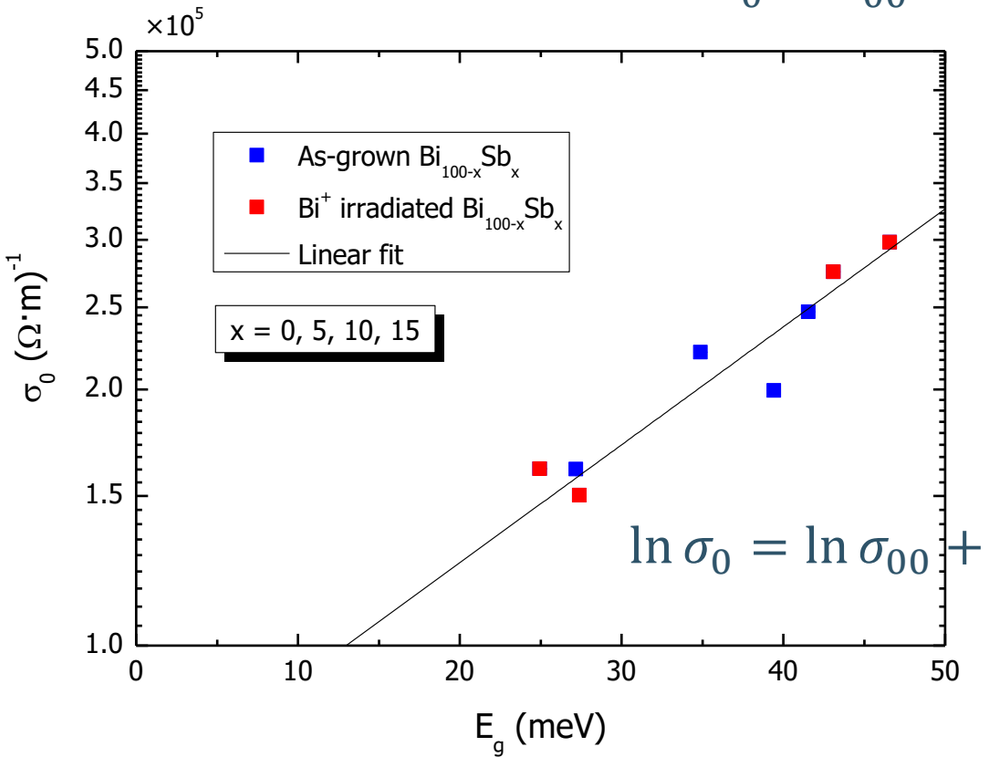
$$\rho(T) = \rho_0 e^{E_g/2k_B T}$$



Meyer-Neldel (1937) Rule: $\sigma_0(E_g)$

$$\frac{1}{\rho} \equiv \sigma = \sigma_0 e^{-E_g/2k_B T} = \sigma_{00} e^{E_g/2k_B T_0} e^{-E_g/2k_B T}$$

$$\sigma_0 = \sigma_{00} e^{E_g/2k_B T_0}$$



$$\ln \sigma_0 = \ln \sigma_{00} + \frac{1}{2k_B T_0} E_g$$

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Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

- Small-gap **semiconducting** behaviour following the **Meyer–Neldel** empirical rule, even for expected **semimetal** pure Bi.
- **Resistivity** **increases** with **irradiation** in pure **Bi** samples but **decreases** in **Bi-Sb** samples
- Sample **method of preparation** highly **affects** the electrical properties.

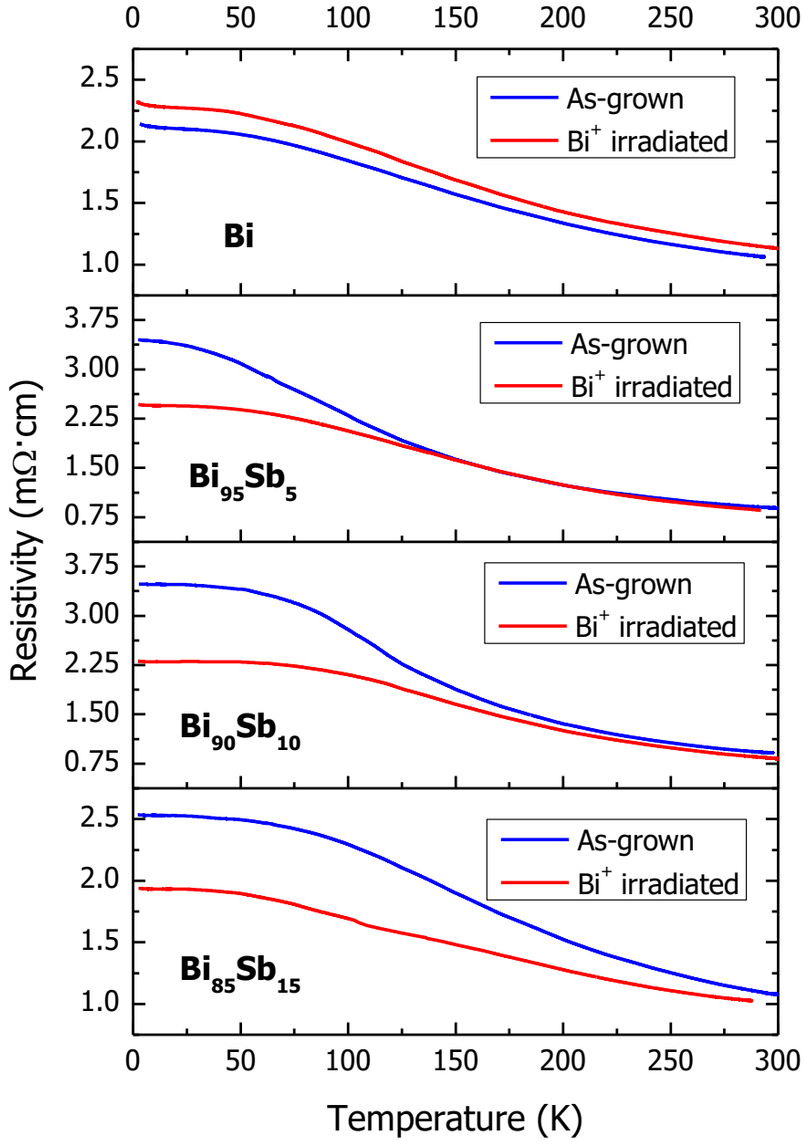
Low Temperature Physics ARTICLE pubs.aip.org/aip/ltp

Low-temperature electrical conductivity of ion-beam irradiated Bi-Sb films

Cite as: Fiz. Nizk. Temp. **50**, 427-433 (May 2024); doi: 10.1063/10.0025622
Submitted: 26 March 2024

A. Andrino-Gómez,[1,2,3,4] M. Moratalla,[1,3,4] A. Redondo-Cubero,[2,3,4] N. Gordillo,[2,3,4] and M. A. Ramos[1,3,4,a)]

Films by thermal evaporation



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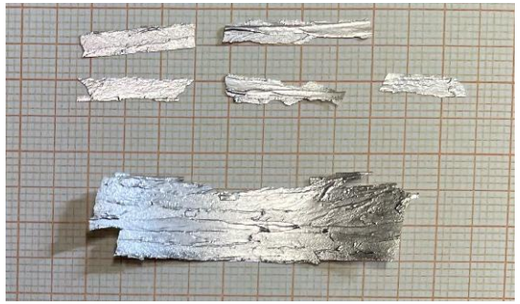
Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Bi-Sb

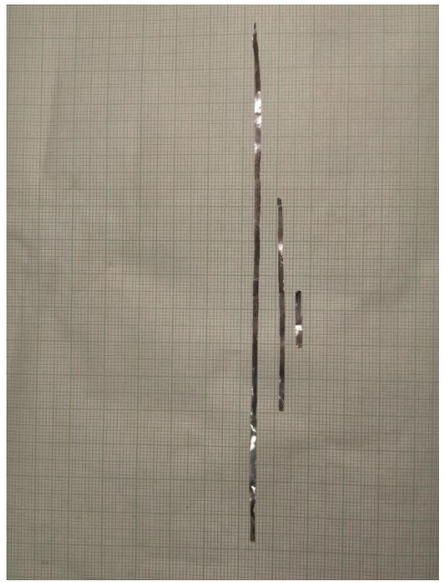


$Bi_{100-x}Sb_x$

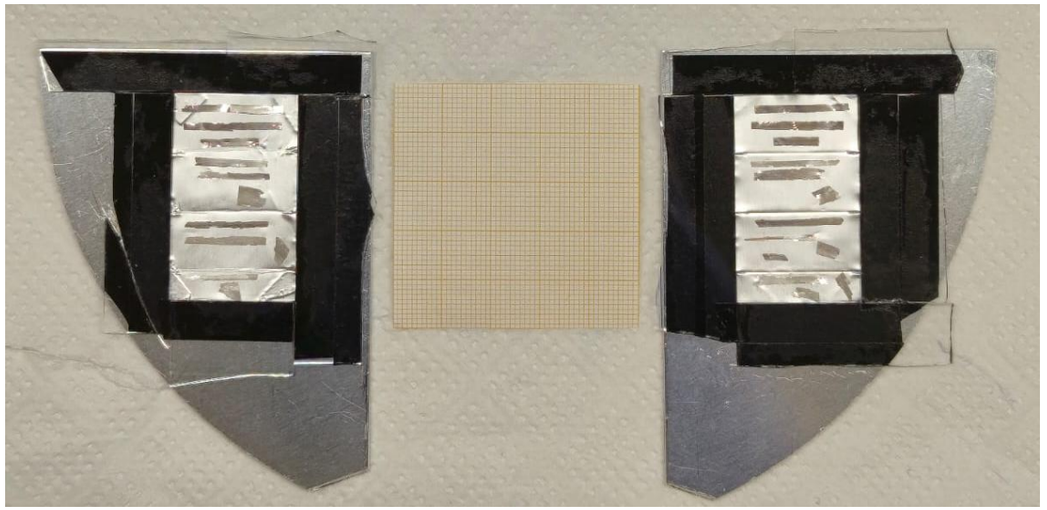
~10-40 μm - thick crystalline ribbons

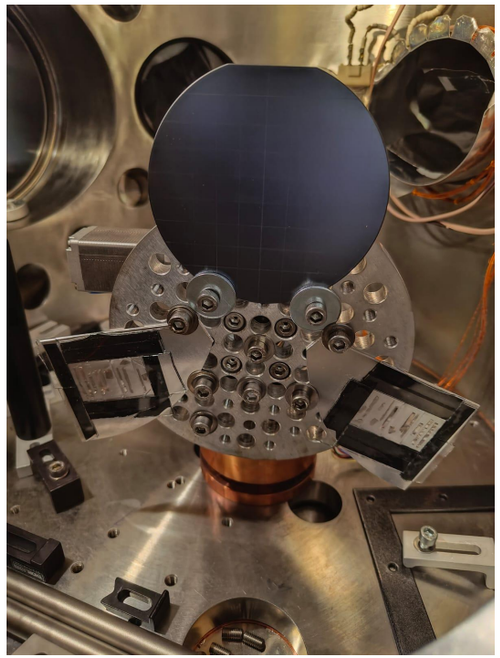
x = 0, 5, 10, 15

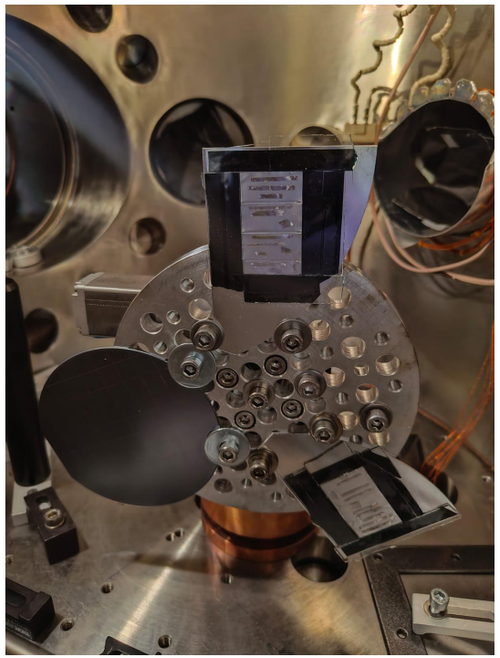
pure Bi



Ribbons by melt spinning







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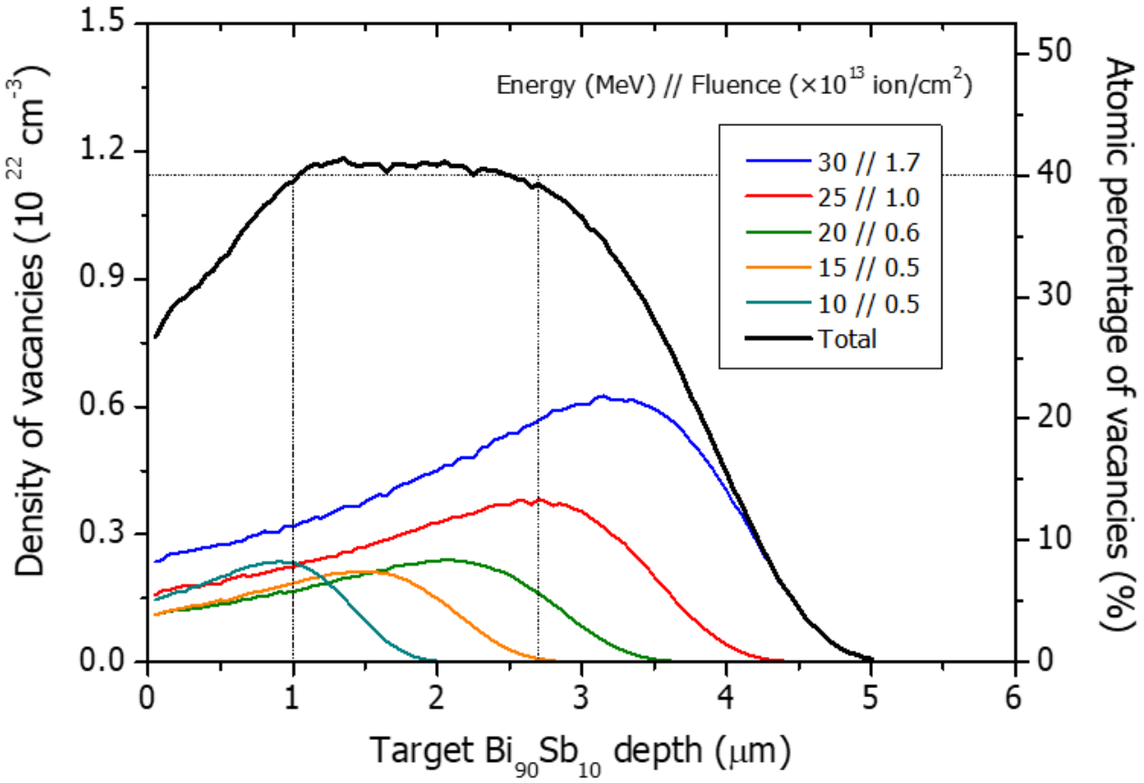
Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Exhaustive study with different methods of sample preparation



- Co-evaporation of pure materials
- Thermal evaporation of Goodfellow stoichiometric alloys
- **Melt spinning** method

- Irradiation of Bi-Sb samples with **10–30 MeV Bi** self-ions

- Nominal **40% atomic vacancies** in the range **≈1–3 μm**

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Experimental Results (up to now)

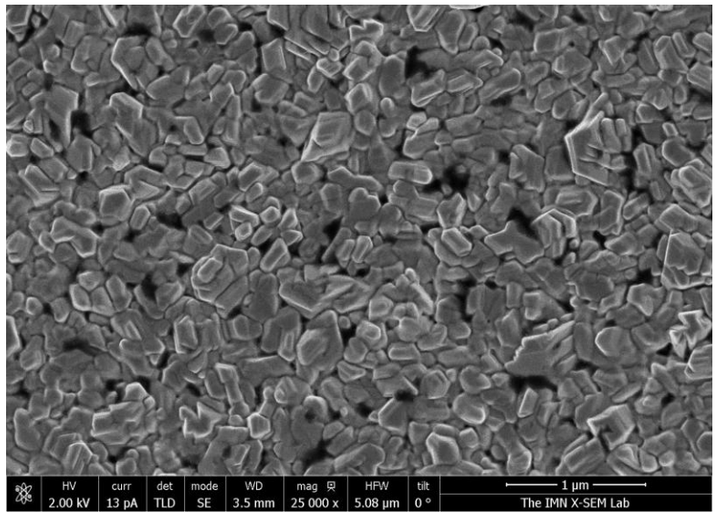
Quest for amorphous superconductors of Bi-Sb …

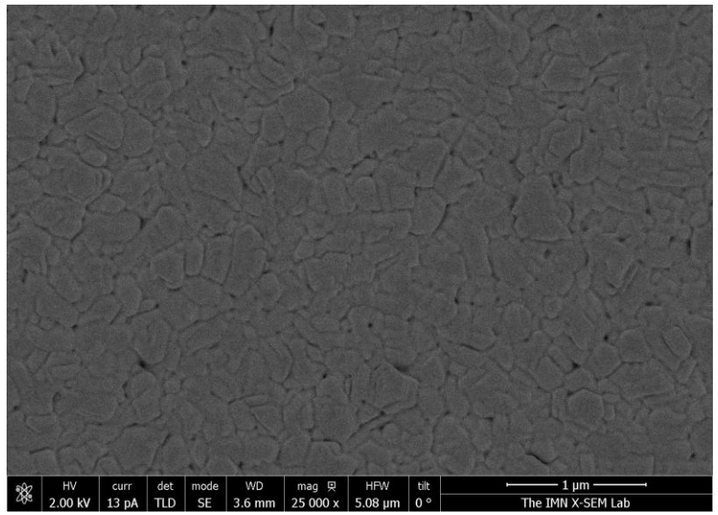
Exhaustive study with different methods of sample preparation

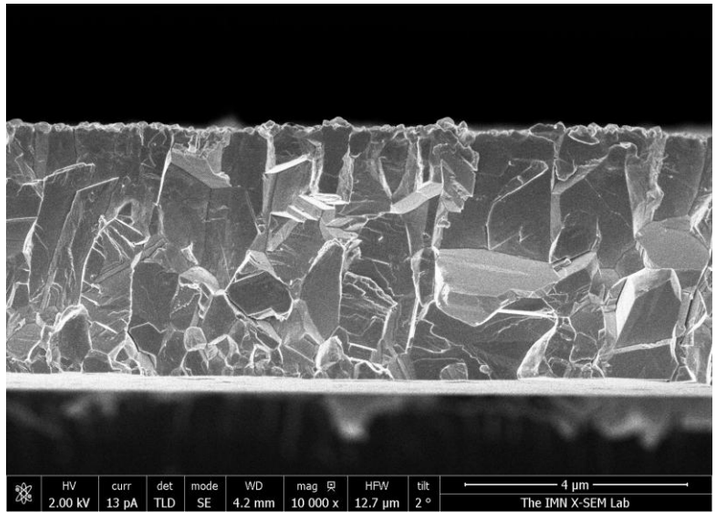
SEM
(Electronic microscopy)

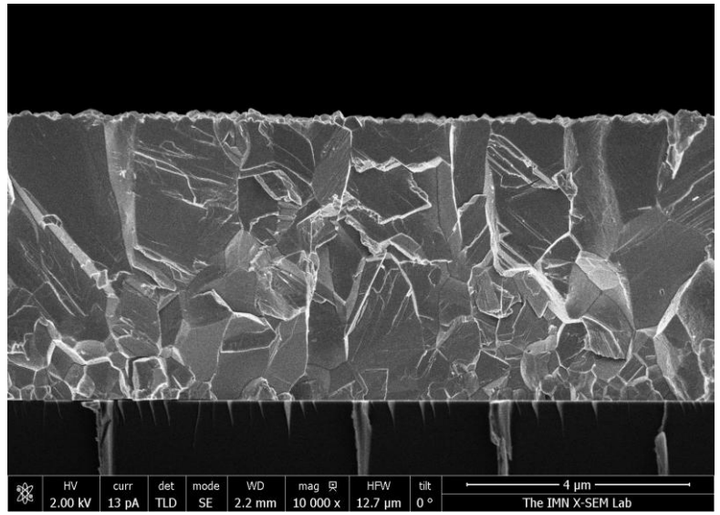
$Bi_{90}Sb_{10}$

Before irradiation









After irradiation

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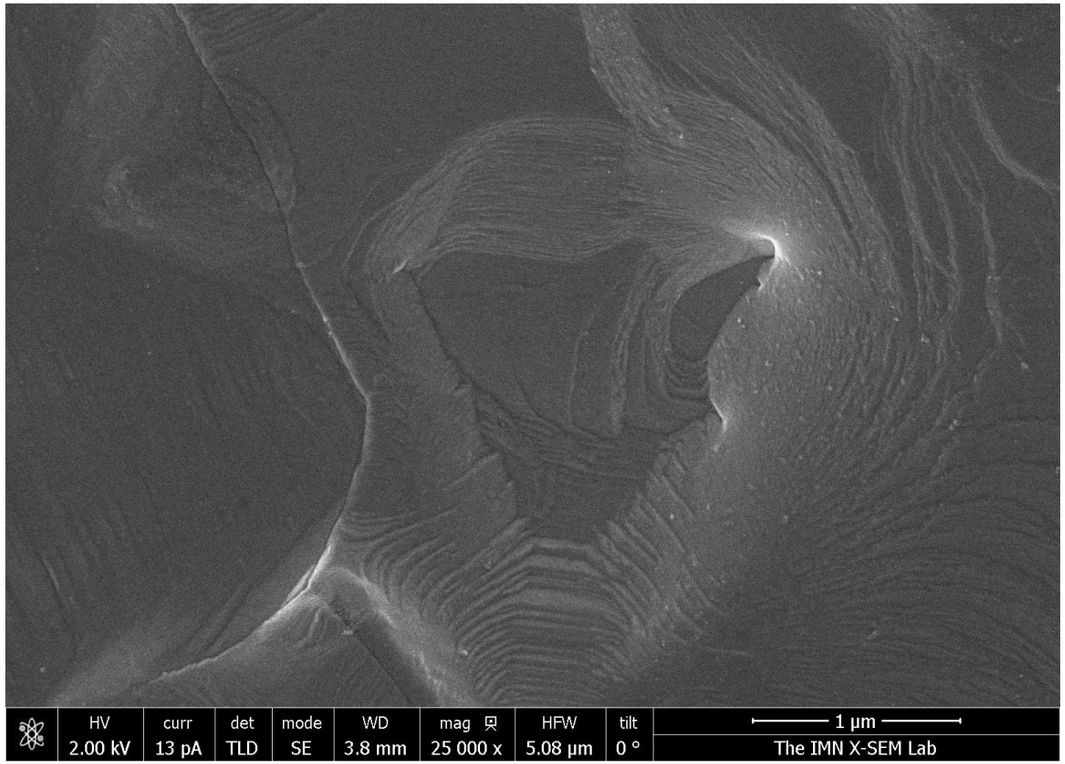
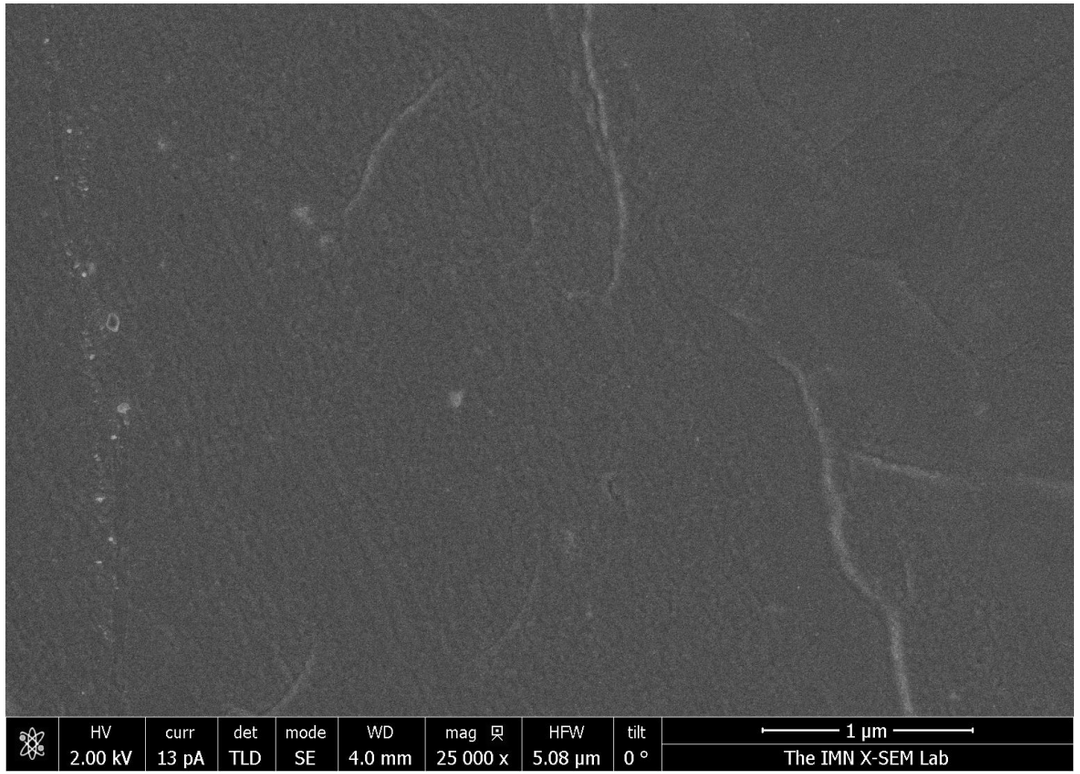
Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Exhaustive study with different methods of sample preparation

$Bi_{90}Sb_{10}$

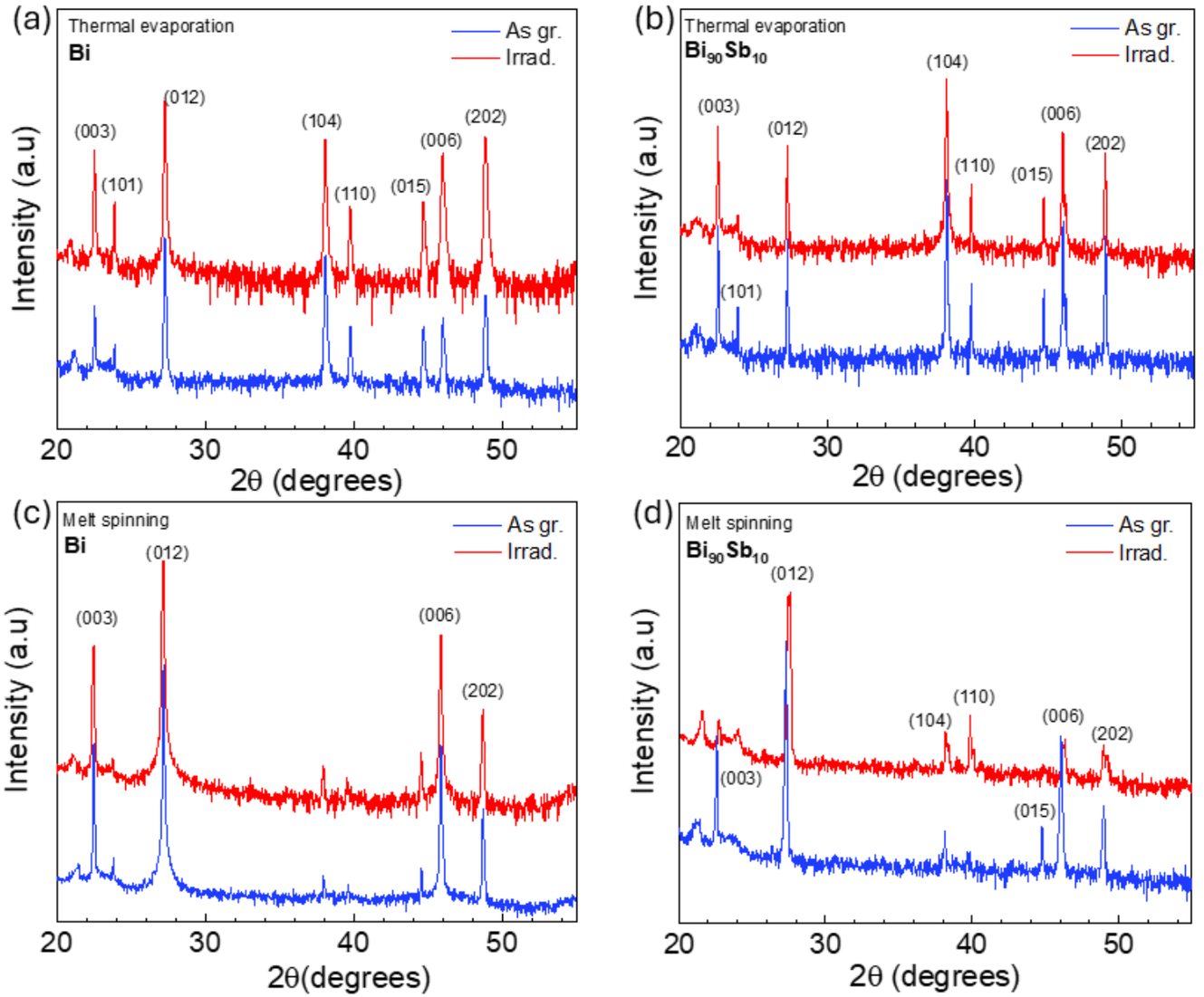




Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …

Exhaustive study with different methods of sample preparation



Bi

→ **Thermal evaporation**

$Bi_{90}Sb_{10}$

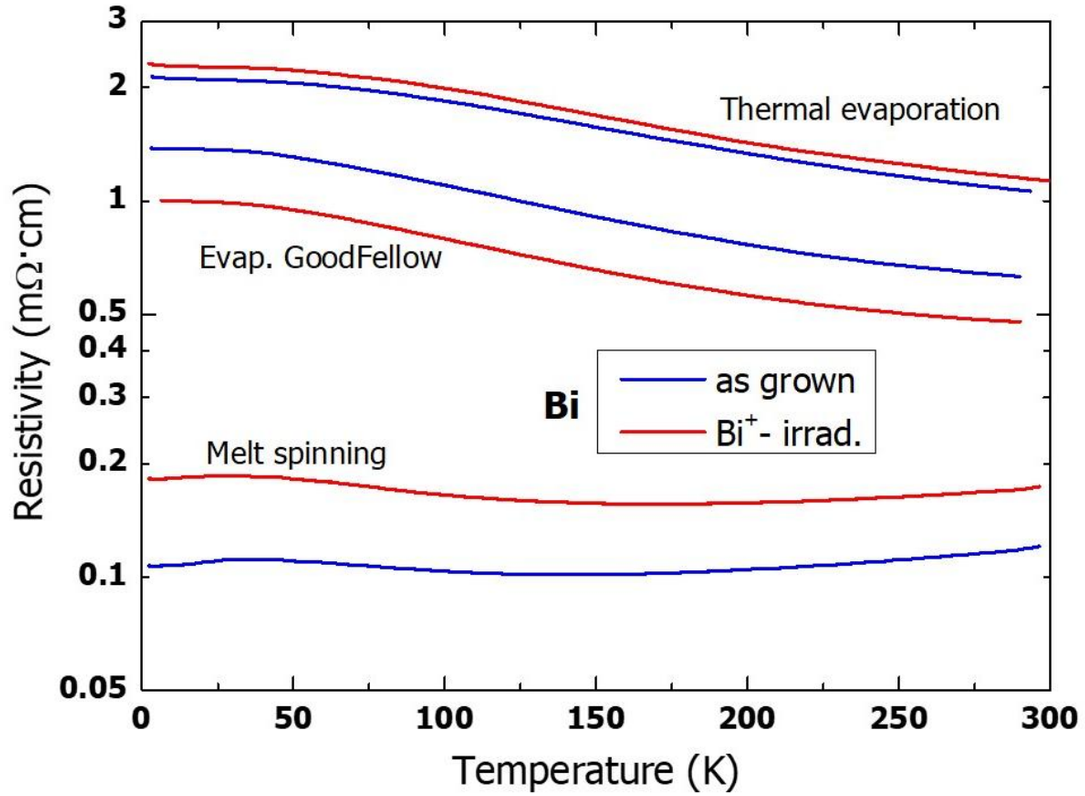
→ **Melt spinning**

Experimental Results (up to now)

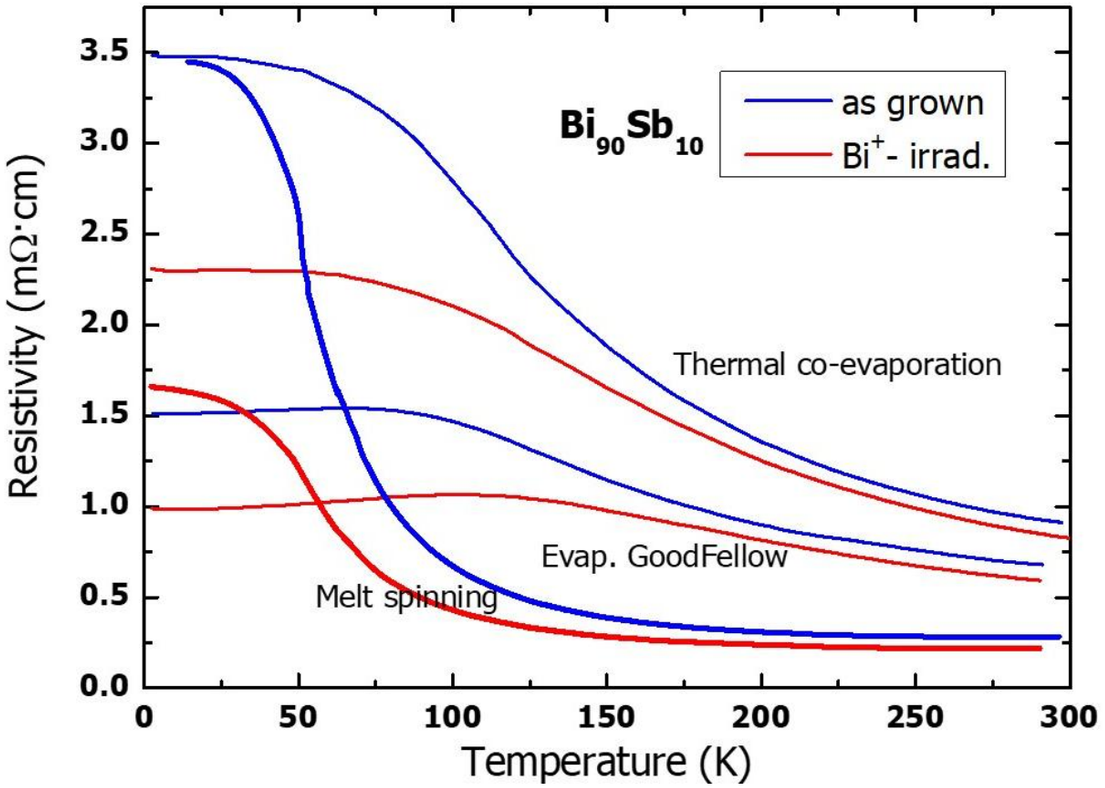
Quest for amorphous superconductors of Bi-Sb …

Exhaustive study with different methods of sample preparation

Bi



$Bi_{90}Sb_{10}$



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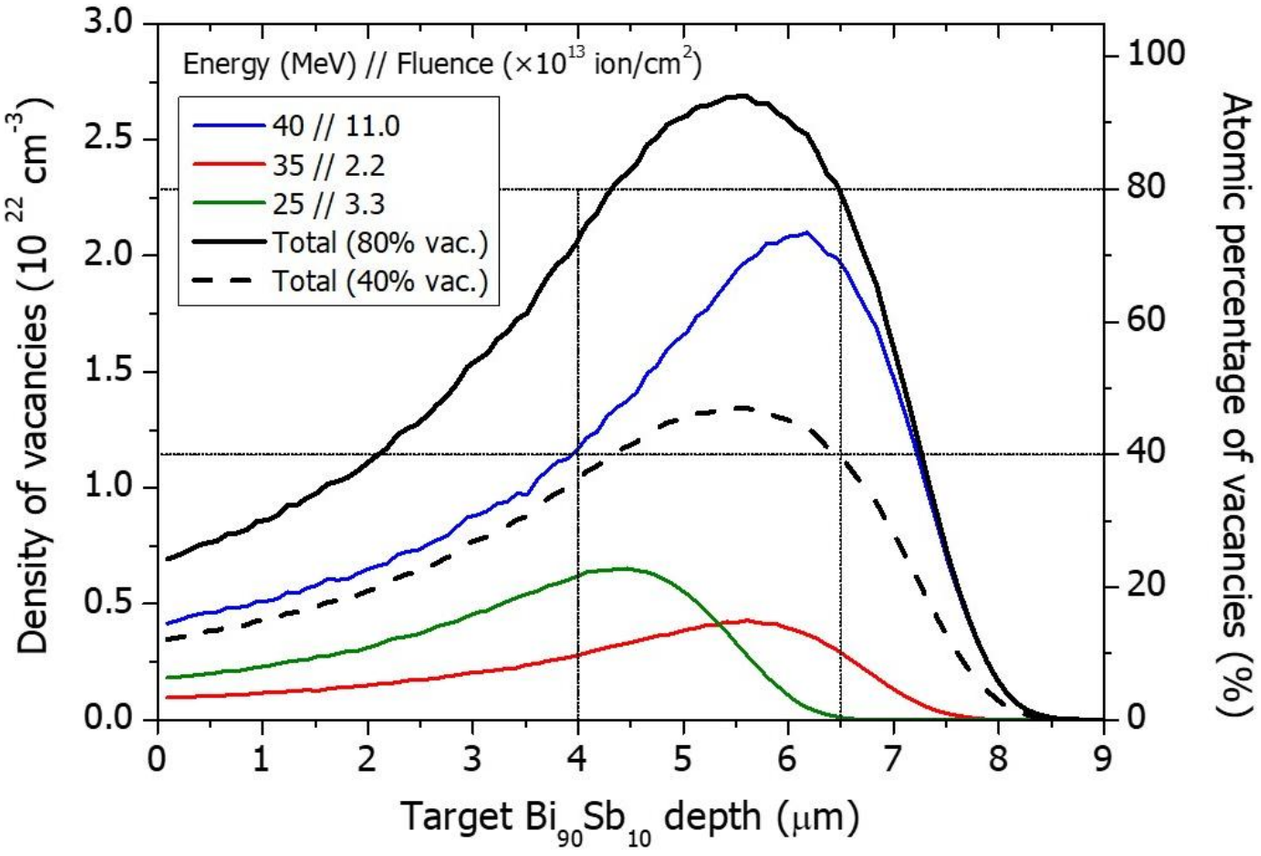
Experimental Results (up to now)

Quest for amorphous superconductors of Bi-Sb …



Another turn of the screw

- **(now focusing on melt-spun ribbons)**
- Irradiation with more efficient **Iodine** ions with **25–40 MeV**
- Nominal **40% atomic vacancies** in a deeper range ≈ **4–7 μm**
- Also DOUBLING the fluences to **80% atomic vacancies**

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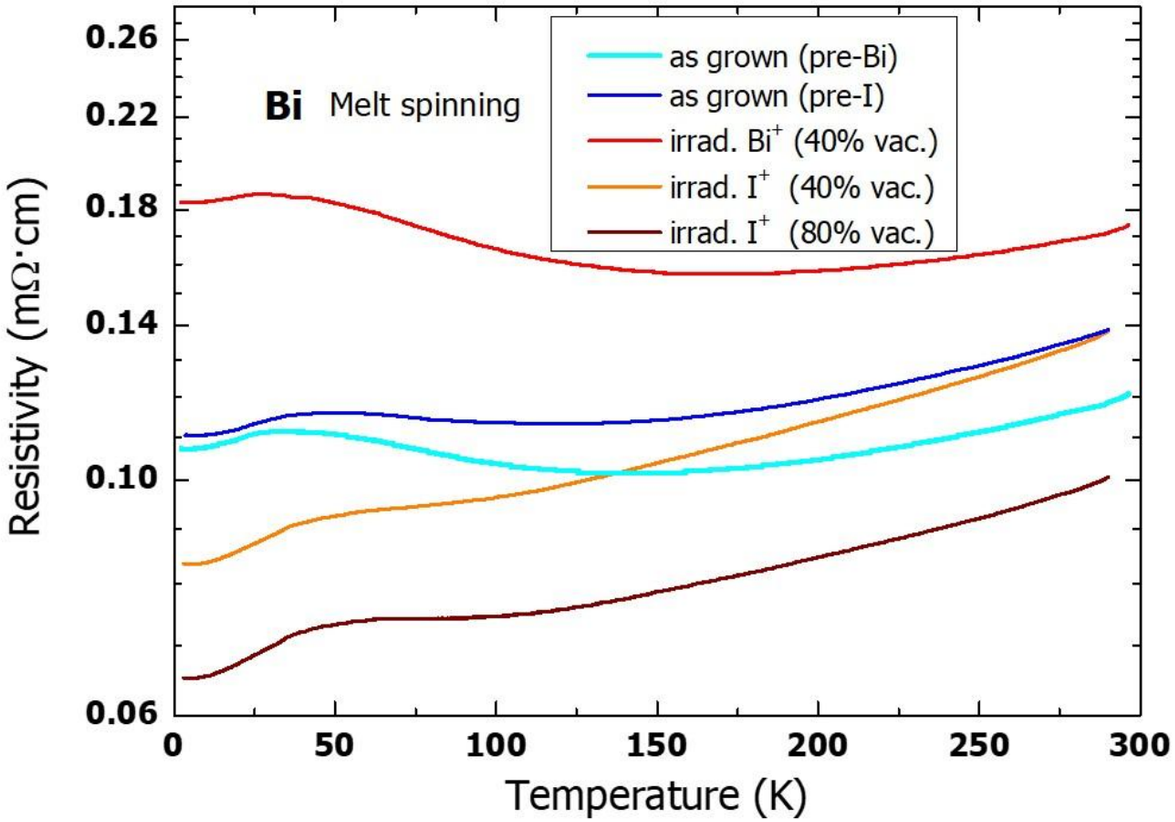
Experimental Results (up to now)

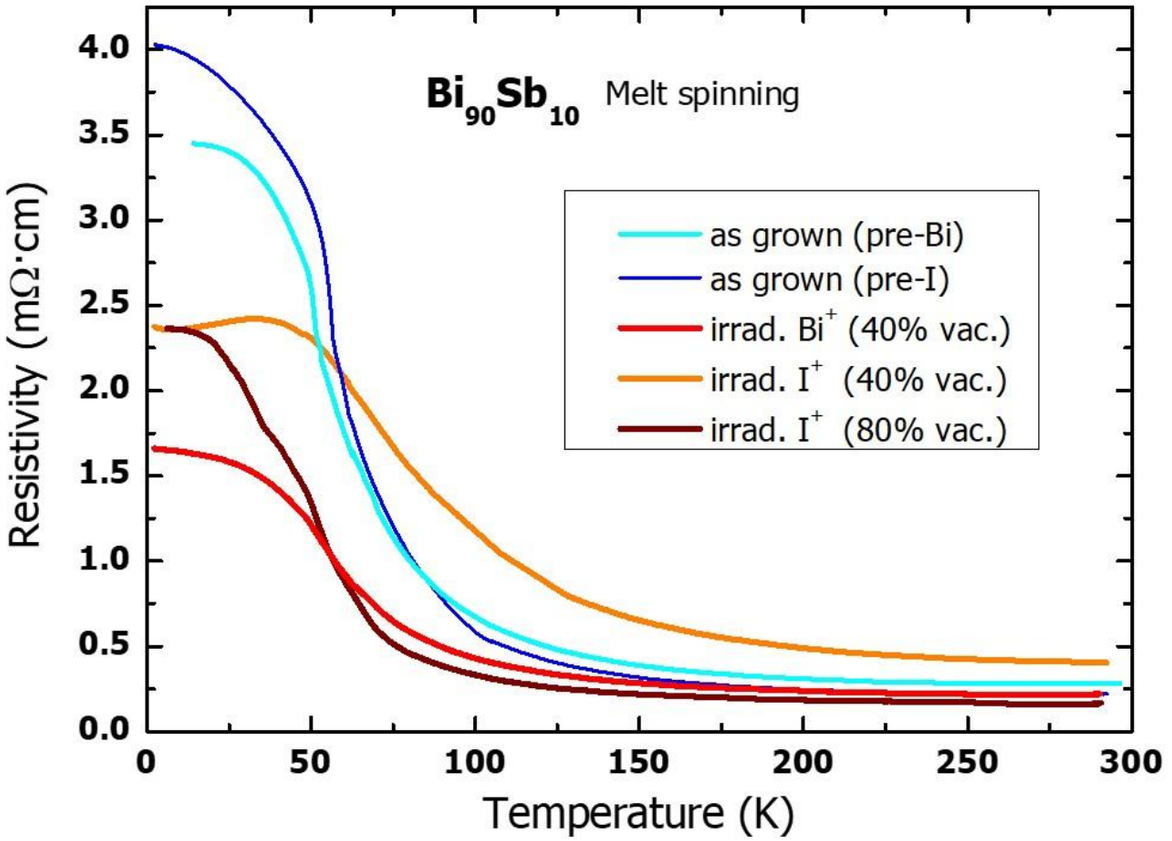
Another turn of the screw

Quest for amorphous superconductors of Bi-Sb …

Bi



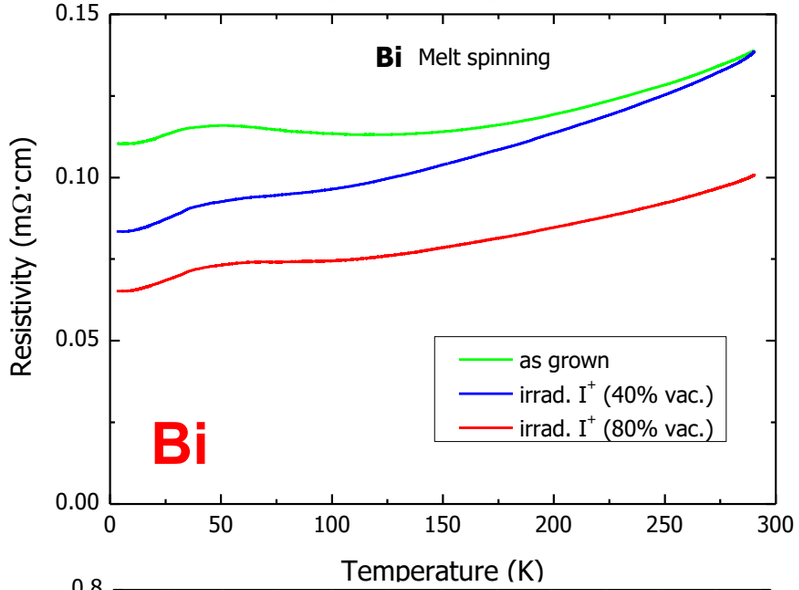
$Bi_{90}Sb_{10}$

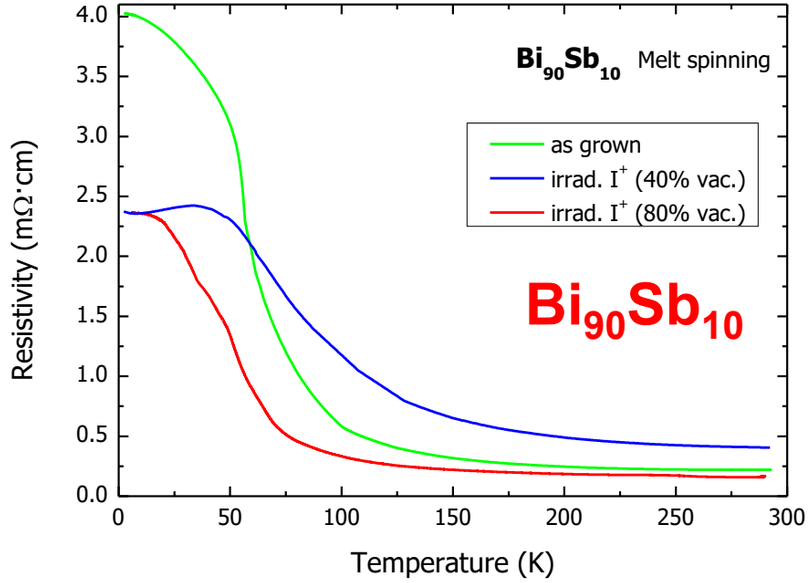


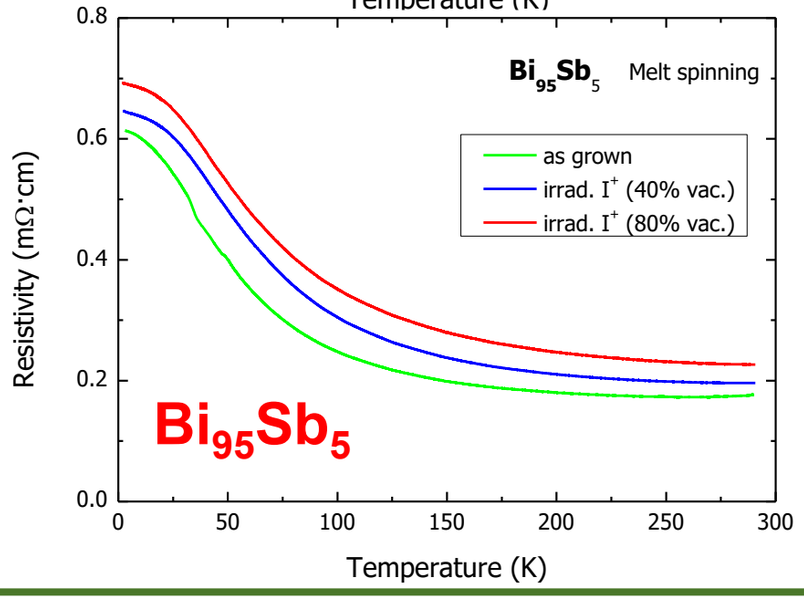
Experimental Results (up to now)

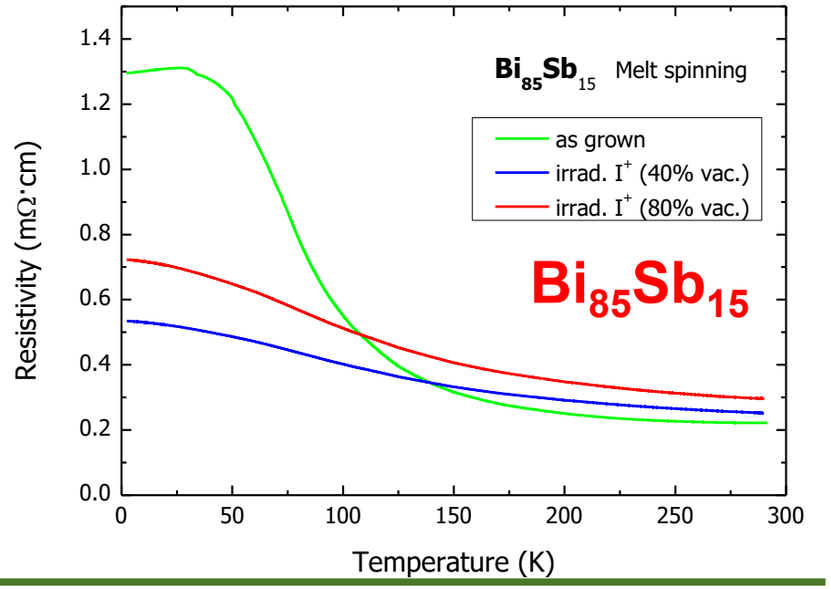
Quest for amorphous superconductors of Bi-Sb …

Irradiation with **Iodine** ions
(40% / 80% vacancies)
for ***x* = 0, 5, 10, 15.**









Bulk single crystals @ R.T.:

ρ = **0.13 mΩ·cm** for pure Bi

$\rho \approx$ **0.2 mΩ·cm** for $Bi_{90}Sb_{10}$

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Ongoing experiments

Quest for amorphous superconductors of Bi-Sb …

- We have irradiated further samples:

  - new, longer and thinner ribbons by melt spinning
  - comercial foils of pure Bi

- … with **25–40 MeV Iodine** with ×**2** and ×**3** the reference fluences (40%)

- We are currently characterizing those samples…

Summary and conclusions

Quest for amorphous superconductors of Bi-Sb …

- We have prepared, irradiated and characterized different types of $Bi_{100-x}Sb_x$ samples (*Materials Science Matters!*)

- We have irradiated **all types of Bi-Sb** samples with 10–30 MeV **Bi ions**, aiming to generate damage ~3 μm in depth (with nominally 40% atomic vacancies)
  → Only melt-spun ribbons exhibited the electrical behaviour of bulk crystals
  → Low-T resistivity curves typically decreased with increasing irradiation

- Focusing only on **melt-spun ribbons**, we also have irradiated with **25–40 MeV I ions**, aiming to generate damage in a deeper range ~3.5–7 μm (with nominally 40% and also 80% atomic vacancies)
  → These irradiations further improved (slightly) the electrical conductivity

- Structural and electrical measurements show that irradiated samples still remain polycrystalline (**not amorphous**) and hence **non-superconducting**, but rather semimetallic (Bi**)** or small-gap semiconducting (Bi-Sb)

Oulook

Quest for amorphous superconductors of Bi-Sb …

- Concluding measurements and characterization on **new samples** with **higher I-ion fluences** for all studied compositions of $Bi_{100-x}Sb_x$ (x = 0–15).

- Planned similar I-ion irradiations but **at lower temperatures (77 K)** @HZDR

- Trying to conduct **irreversible densification** of $Bi_{100-x}Sb_x$ samples (before and after irradiation)

Quest for amorphous superconductors of Bi-Sb alloys by irradiation with swift heavy ions

¡Gracias!

Thank You!

Tack!