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The creation and Retention at ambient of the metastable superconducting state with a 151 K T_c

Paul C. W. Chu

*Department of Physics & TCSUH
University of Houston*

- Collaborators:
TCSUH – **L. Z. Deng**, T. Hasamahoro, A. Safezoddeh, T. Bonke, T. R. Fujikawa, D. Schulze, S. Kazibwe, D. Rolston, Z. Wu and M. Gooch/ B. Karki, P. R. Hoser;
Rice U– B. Gao, T. Chen and P. C. Dai; **IV**- M. Julian and R. P. Prasakumar;
ANL- H. Zhou and J. Smith; **Jilin U** - Y.M. Ma et al.; and **CAS** – C. Q.
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Superconductivity is one of the oldest but continues to be a full of vigor field of study ever since its discovery more than a century ago for its scientific lure and technological potential.

**To raise the T_c above the prevailing records
has been the driving force for the century
old active effort on superconductivity**

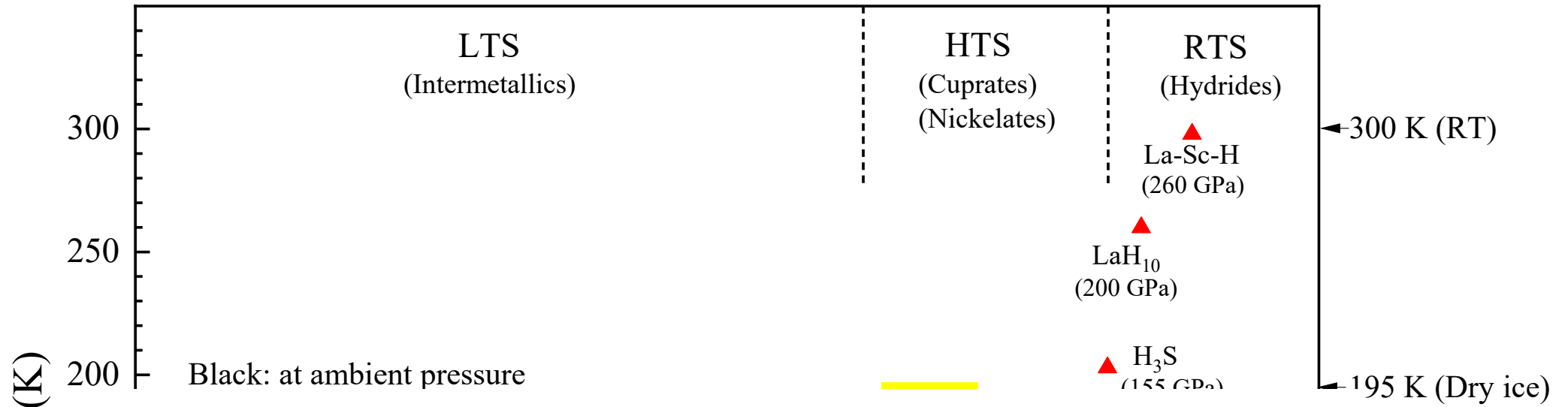
Study

(to discover, to understand and to apply)

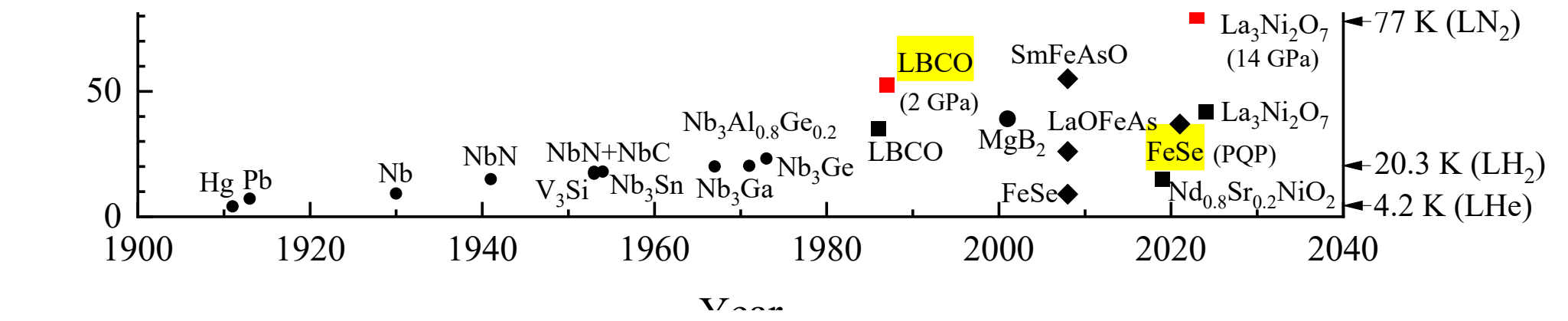
- *New materials? from elements to compounds; from simplicity to complexity*
 - *Universal rules?*
 - *New mechanisms?*
- *Chemical and physical?*
 - *Ultrahigh pressures
(via a metastable state)?*

Superconductivity – Science and Technology

- Science – Edisonian vs Einsteinian: T_c , mechanism
 - BCS: $T_c = 1.14\theta_D \exp(-1/NV)$ – electron pairing, attractive interaction, macroscopic coherent state
 - McMillan: $T_c = [\langle\omega\rangle/1.20] \exp\{-[1.04(1+\lambda)]/[\lambda-\mu^*(1+0.62\lambda)]\}$
 - Ginzburgh; $T_c \sim \exp(1/g)$
 - Matthias: $T_c \sim e/a$ 4.75, 6.4 (LTS); Presland: $T_c/T_{c,max+} \sim 1 \sim 82.6 (n-n_o)^2$ (HTS)
 - characteristic temperature $\Theta \sim 1/M^{-\alpha}$
 - e-ph interaction, mass and instabilities, **LTS**
 - e-e interaction, magnetic fluctuations, e-e correlations, excitons, anyons, magnons, plasmons, demons, electronic instabilities,... **HTS**
 - e-ph interaction, **RTS** (hydrides)



All record high T_c s are under extreme pressures and the current record ~ 300 K



Record High T_c -Superconductors (LTS, HTS and RTS):

they are all relative, a moving target– T_c , operating temp, materials, pressure....

- Edge of the universe – 0.3 K (*The cosmo background radiation, Penzias and Wilson, 1964*)
- **LTS** Liquid He - 4.2 K [*Hg – 4.2 K*] (*Kamerlingh Onnes, 1911*)
- Liquid H₂ – 20.3 K [*Nb₃(Ge,Al) – 21 K, 23.2 K*] (*Matthias et al. 1967; Testardi et al. 1973*)
- **HTS** Liquid N₂ – 77 K [*RBCO - 90's K*] (*Chu et al. 1987*) – ***the holy grail then***
- Space Shuttle - 100 K [*BSCCO - 110 K*] (*Maeda et al. 1988*)
- Liquid Natural Gas - 120 K [*TBCCO - 120 K*] (*Hermann et al. 1988; [HBCCO- 133 K]*) (*Schilling et al. 1993*)
- CF₄ - 148 K [*HBCCO -164 K, 31 GPa*] (*Chu et al. 1993*) – [but high pressure required and ozone layer depletion]
- **RTS** - ≥ 203 K [*Hydrides under very high pressures*]– ***the Holy grail now***
 - H₃S – ~ 203 K at ~ 155 GPa (Eremets et al. 2015)*
 - LaH₁₀ - ~ 260 K at ~ 180-200 GPa (Hemley et al. 2019)*
 - C-S-H - ~ 287 K at – 267 GPa (Dias et al. 2000)***
 - Lu-N-H - ~ 294 K at 1GPa (Dias et al. 2023) – the holy grail then?***
 - Pb-Cu-P-O - ~403 K at 1atm (Lee et al. 2023)***
 - La-Sc-H – 296 K at 260 Gpa (Y. M. Ma ET AL. 2025)*

#5

- We now enter the RTS era of hydrides by replacing the difficult temperature barrier unfortunately by an even more formidable pressure barrier
 - A possible solution-
controlled Pressure Quench Protocols (PQP)
and a new record T_c at ambient

To search for new record Tc's at ambient: e.g.

1. to modify an existing compound system, through optimal doping, **Nb₃Ge- 23.2 K (1973)**
2. to find a new compound system, e.g. the first quaternary compound **YBCO- 93 K (1987)**
3. to modify the existing compound through high pressure, e.g. **HBCCO- 133 K (1993)**
[and **164 K at 31 GPa (1993)**]
4. to retain the metastable phase with minimum efforts by PQP - a non-chemical route – **HBCCO 151 K (2026)**

Pressure-Quench-Process (PQP)

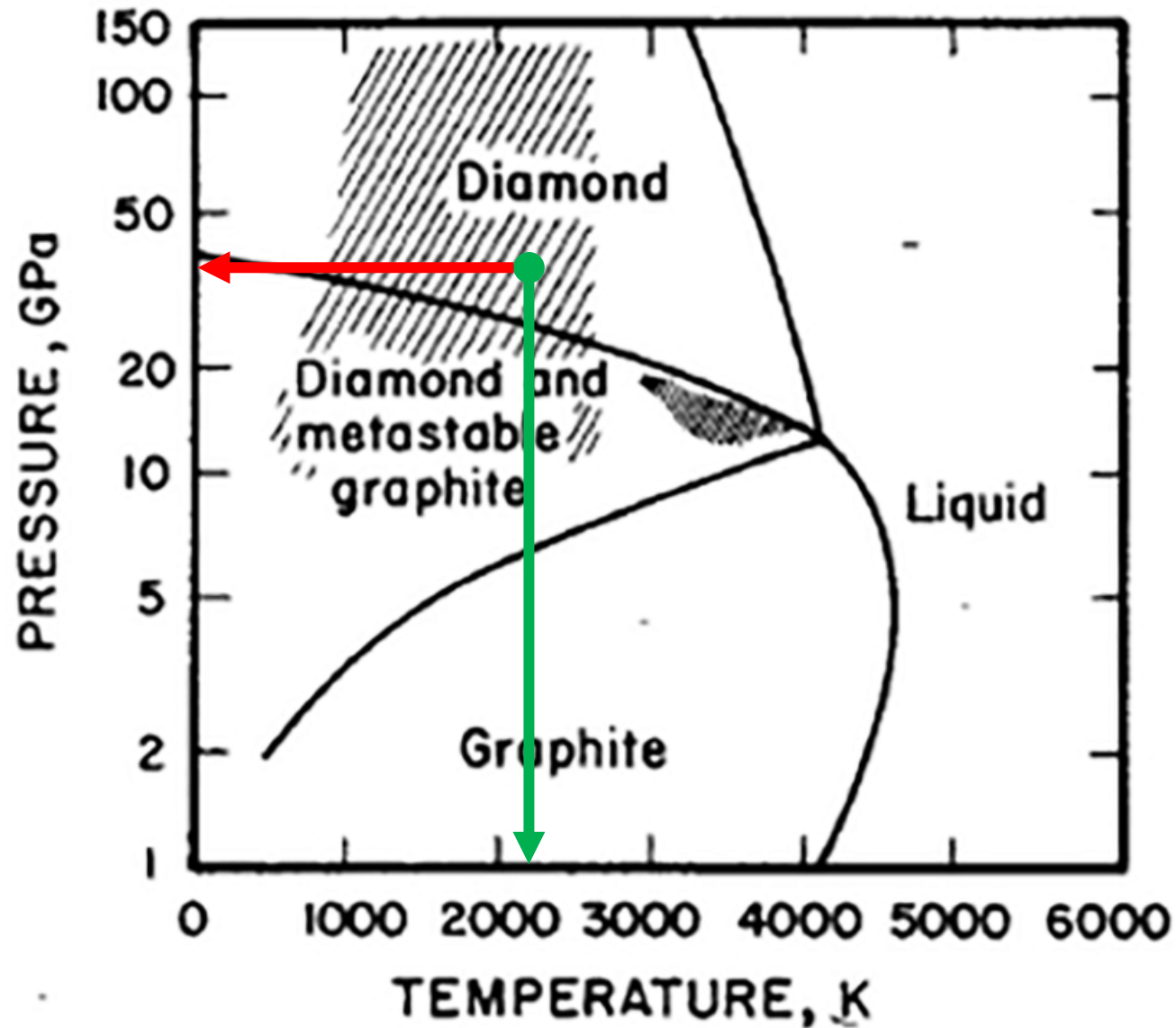
the concept is not new but its deployment for for superconductivity is

- “Most of the alloys used in industrial applications are actually metastable at atmospheric pressure and room temperature, and these metastable phases possess desired and/or enhanced properties that their stable counterpart lack.”
 - Pol Duwez (metallic glass), Francis Kennedy (artificial diamond)
- The high-pressure-induced phases in HTS and RTS can be considered metastable, i.e. kinetically stable but thermodynamically not, protected only by energy barriers. We have taken advantage of these energy barriers to stabilize the metastable or “supercooled” states via a pressure-quench protocol (PQP) at a specific quench-pressure (P_Q) and a chosen quench-temperature (T_Q)
- Simple element -> binary FeSe -> cuprate HBCCO -> hydrides

Gib's free energy consideration– $G(P, T, \eta, \dots)$ *(latent heat, energy barriers)*

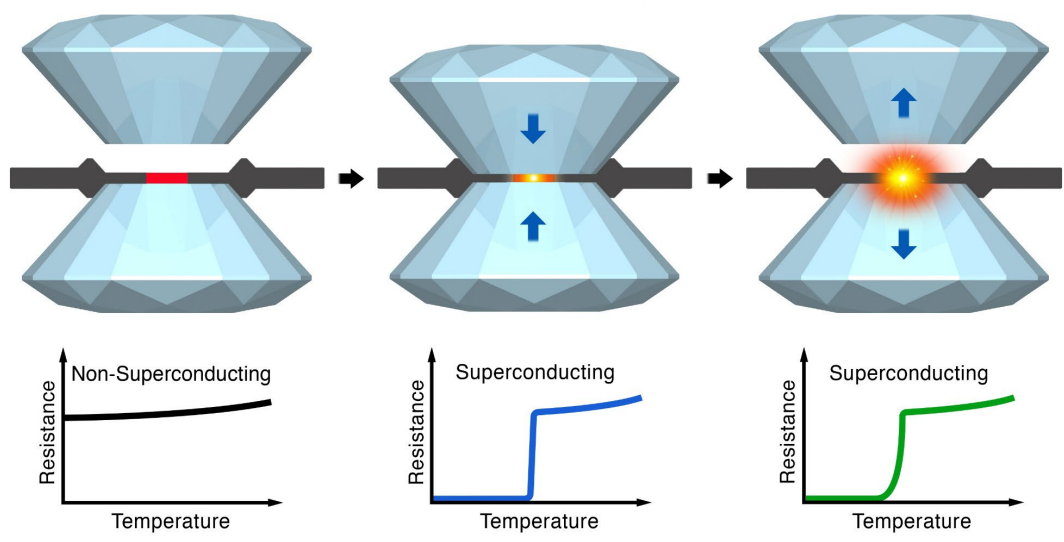
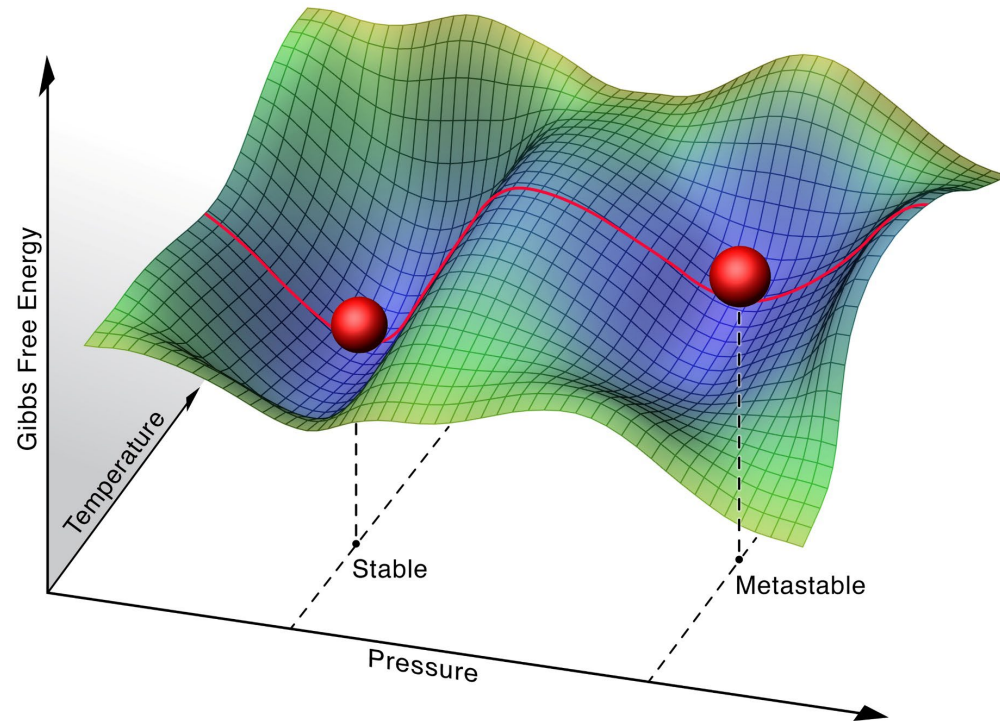
- 1st order -lattice, structure, symmetry
Clapeyron-Clausius equation $dP/dT = \Delta S/\Delta V = l/T \Delta V$
- 2nd order – electronic, often
- 2 1/2 order- Fermi surface topology change

$$\mathcal{N}(\mathcal{E}) = \frac{1}{4\pi^3 N \hbar} \int \frac{dS}{|\mathbf{v}_{\mathbf{k}}|}, \quad (4.6)$$



Francis Bundy
1954

Diamond can be retained by either **pressure** or **temperature**-quench, chemically or physically



Indeed , we found that anomalies of different electron and/or phonon in nature can provide the energy barriers to trap the metastable phases. For instance, we have successfully demonstrated to trap the pressure induced superconducting state in material from Sb, Bi, FeSe, Cu-doped FeSe, $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ in progressing complexity.

The PQP steps to obtain, test and characterize the pressure-induced phase without pressure.

Determine $T_c(P_A)$ phase relation



Select a target phase at P_A for PQ



Measure $R(T)$ under P_A to confirm the phase selected



Pressure-quench at $P_Q (=P_A)$ and T_Q

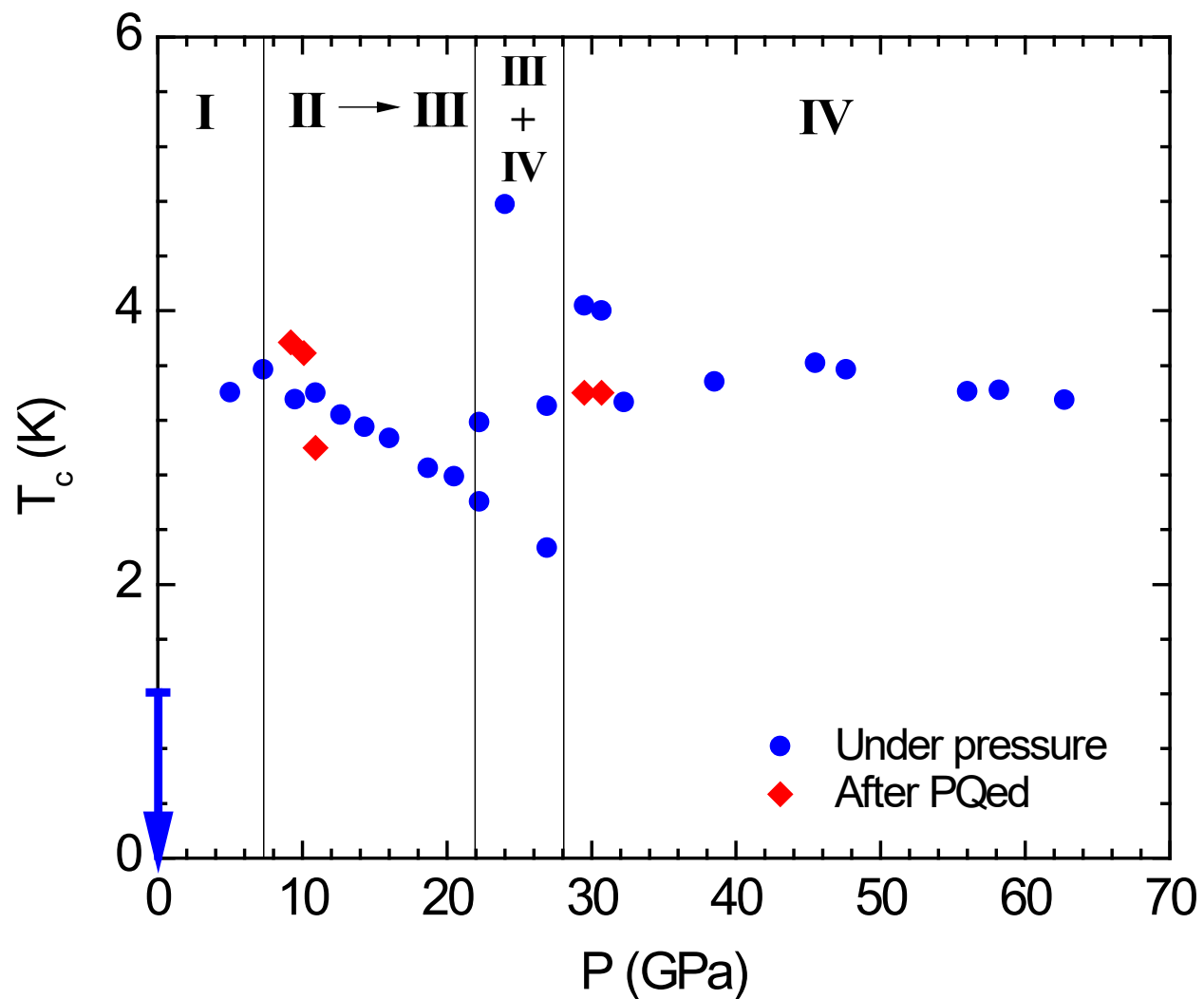


Measure T_c of the retained phase at ambient



Check stabilities of the PQed phases as functions of time and temperature

Sb Single Crystals



Sb single crystal: (blue) T_c at P_A ; (red) T_c at ambient after PQ at P_Q and 77 K

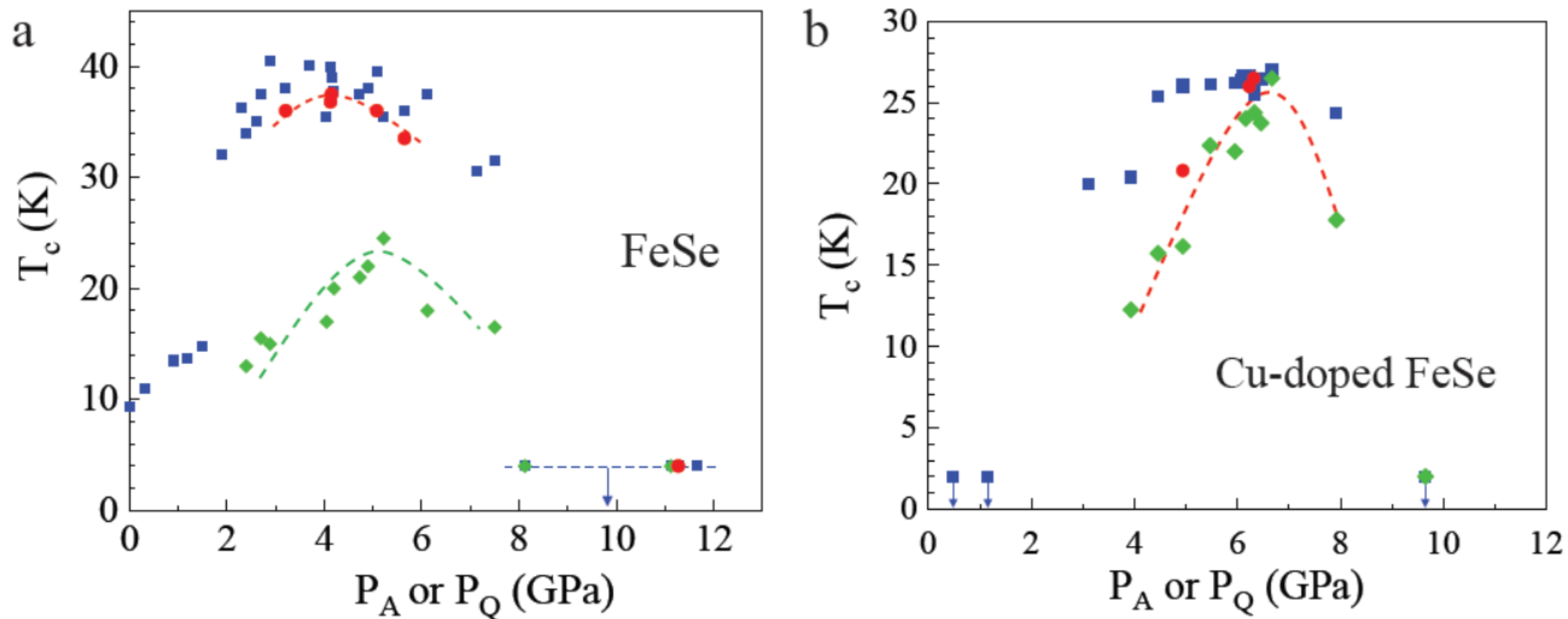
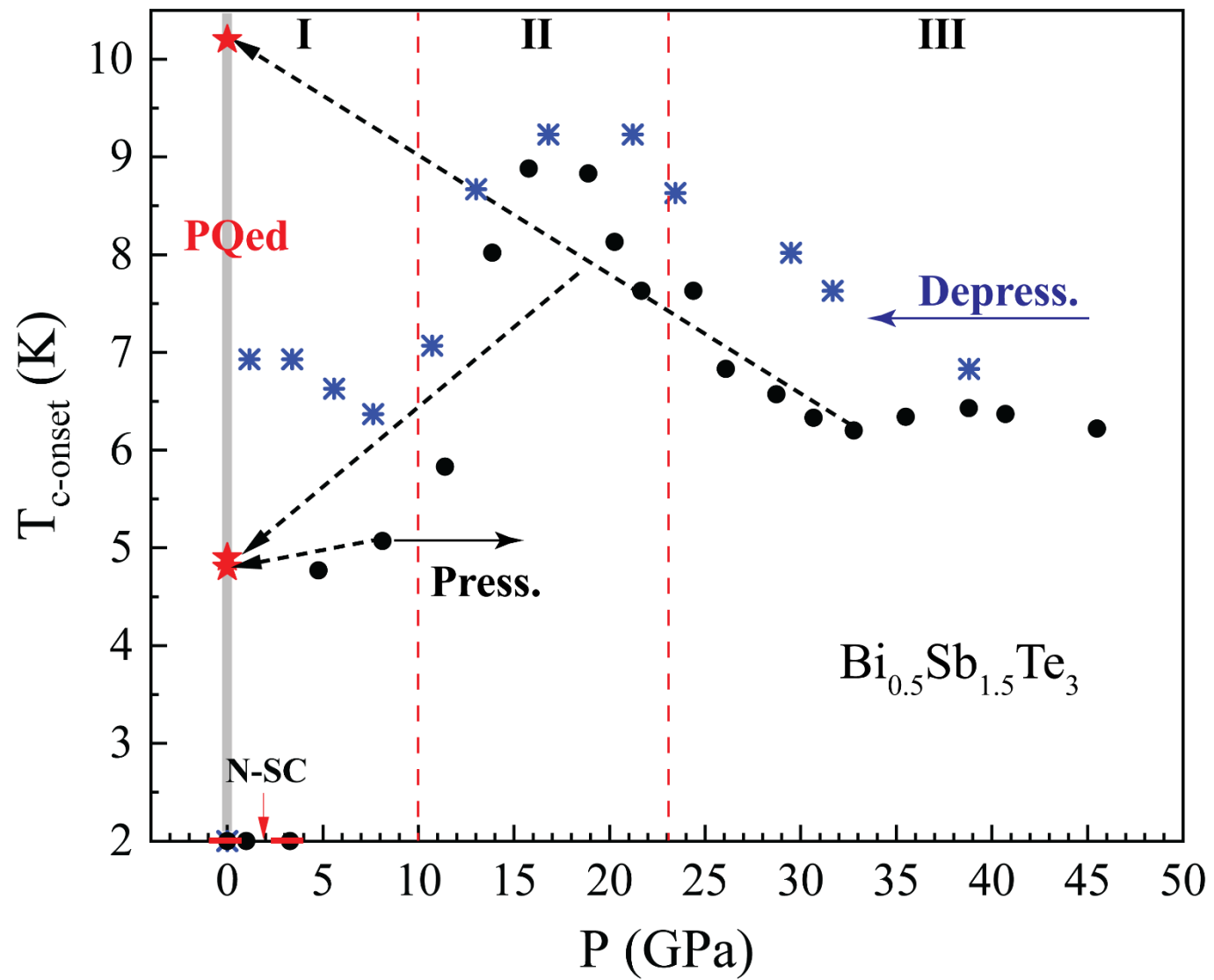
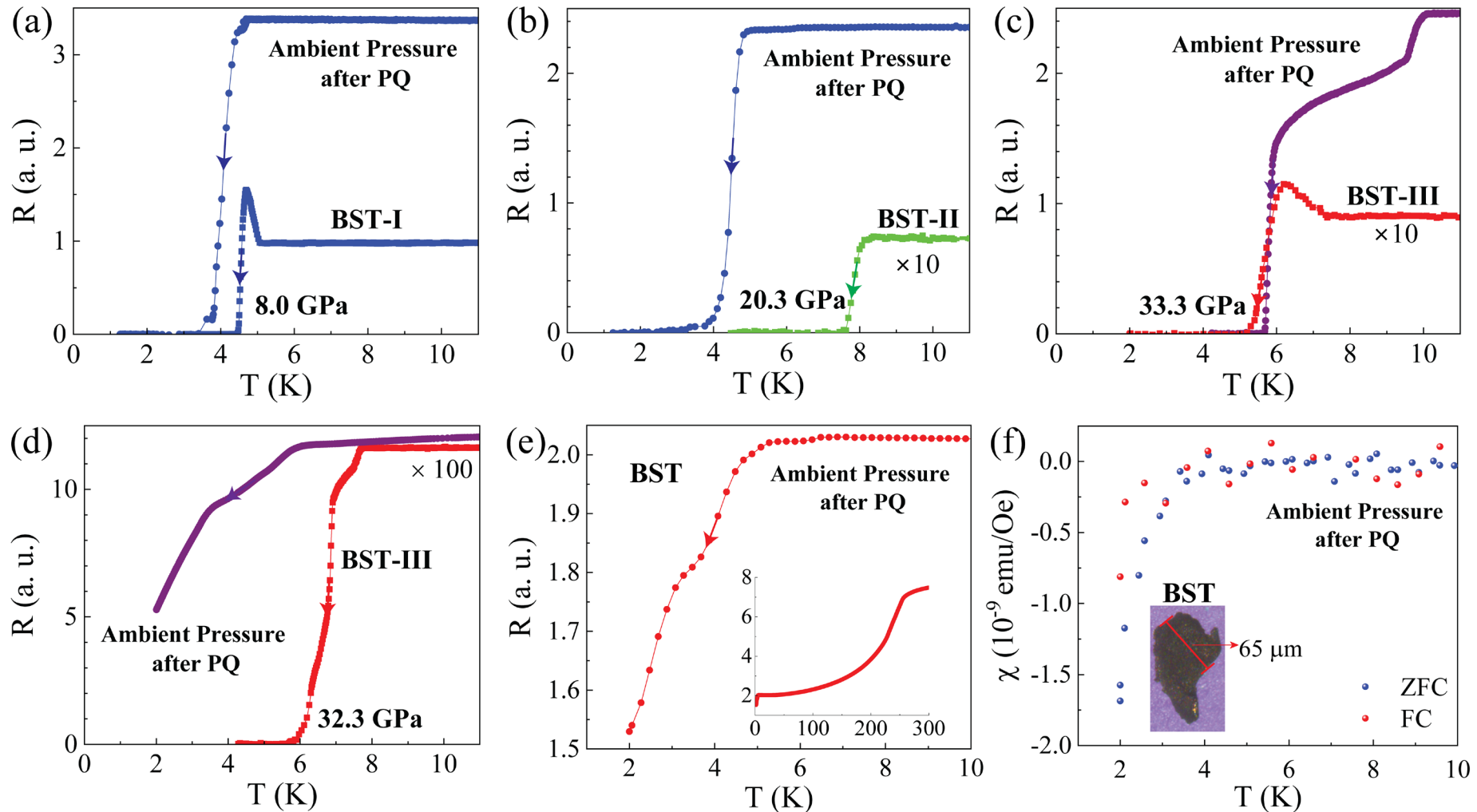


Fig. 2 For (a) FeSe and (b) Cu-doped FeSe, creation of the superconducting phases: T_c (blue squares) at different P_A s; and PQ of the selected superconducting phases: T_c (red circles) at ambient after being PQed at 4.2 K and different P_Q s and T_c (green diamonds) at ambient after being PQed at 77 K and different P_Q s.

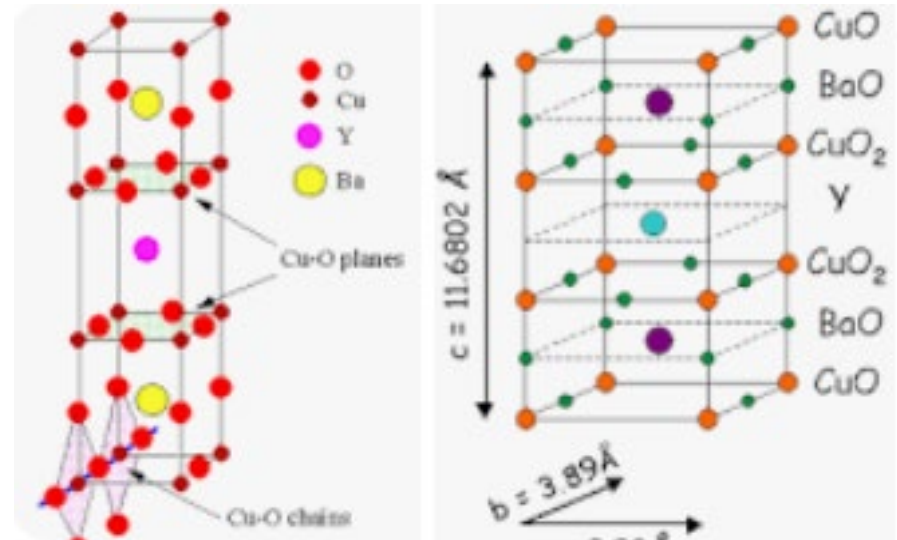


Phase transition may have provided the energy barrier to retain the PQed phase; the PQed Phase is bulk



**A Complex Superconductor needs a
Skeleton to host substructures
Active Component + Charge Reservoir**

1. Laves Phase $RERu_2$
2. A15 $NbSn_3$
3. Chevrel Phase $REMo_6S_8$
4. Rhodium Borides $RERh_4B_4$
5. Cuprates 123 $REBa_2Cu_3O_7$
6. Rare-Earth Hydrides REH_x
RE – skeleton, H – active part & charge
reservoir? – need more data!



***RE affects (1-4) or does not (5, 6?) affect SC.
[cwc]***

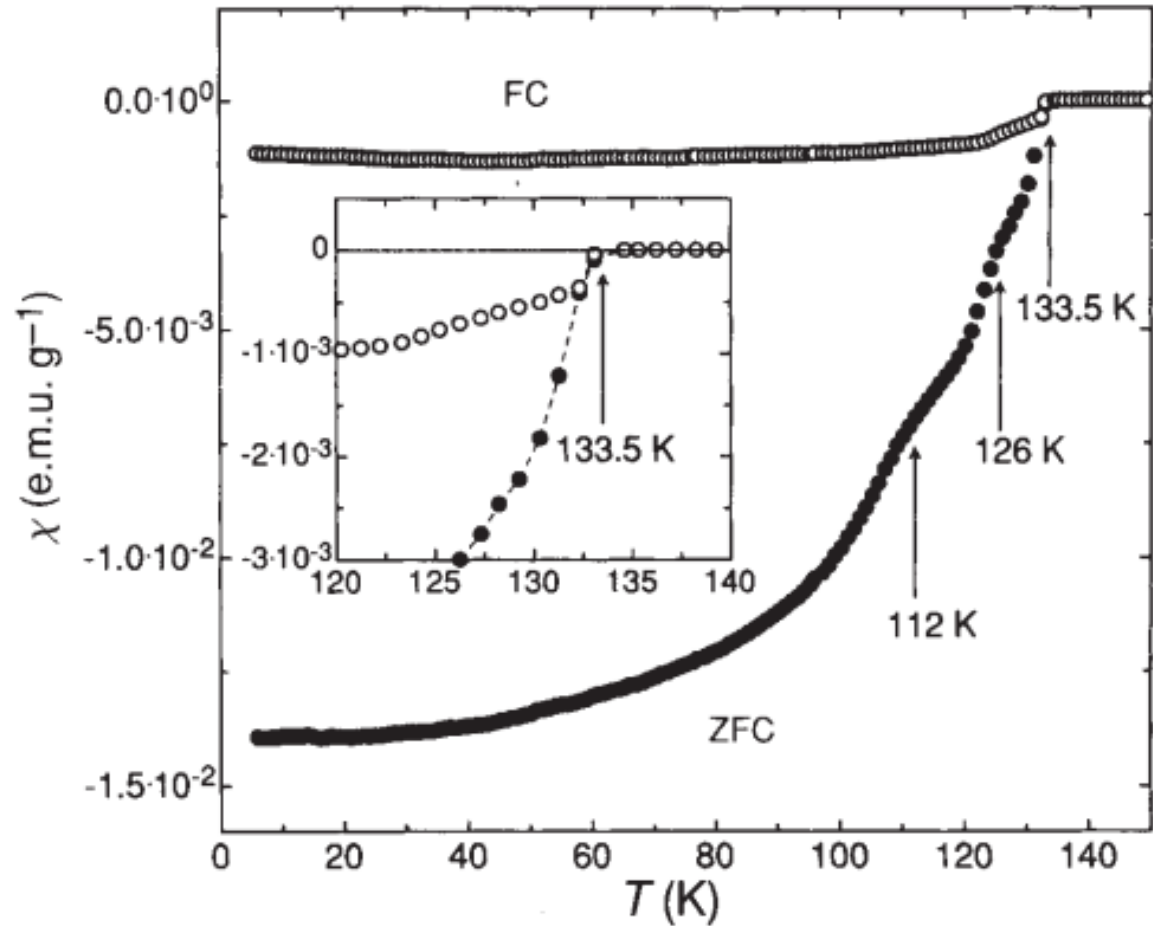
Superconductivity above 130 K in the Hg-Ba-Ca-Cu-O system

A. Schilling, M. Cantoni, J. D. Guo & H. R. Ott

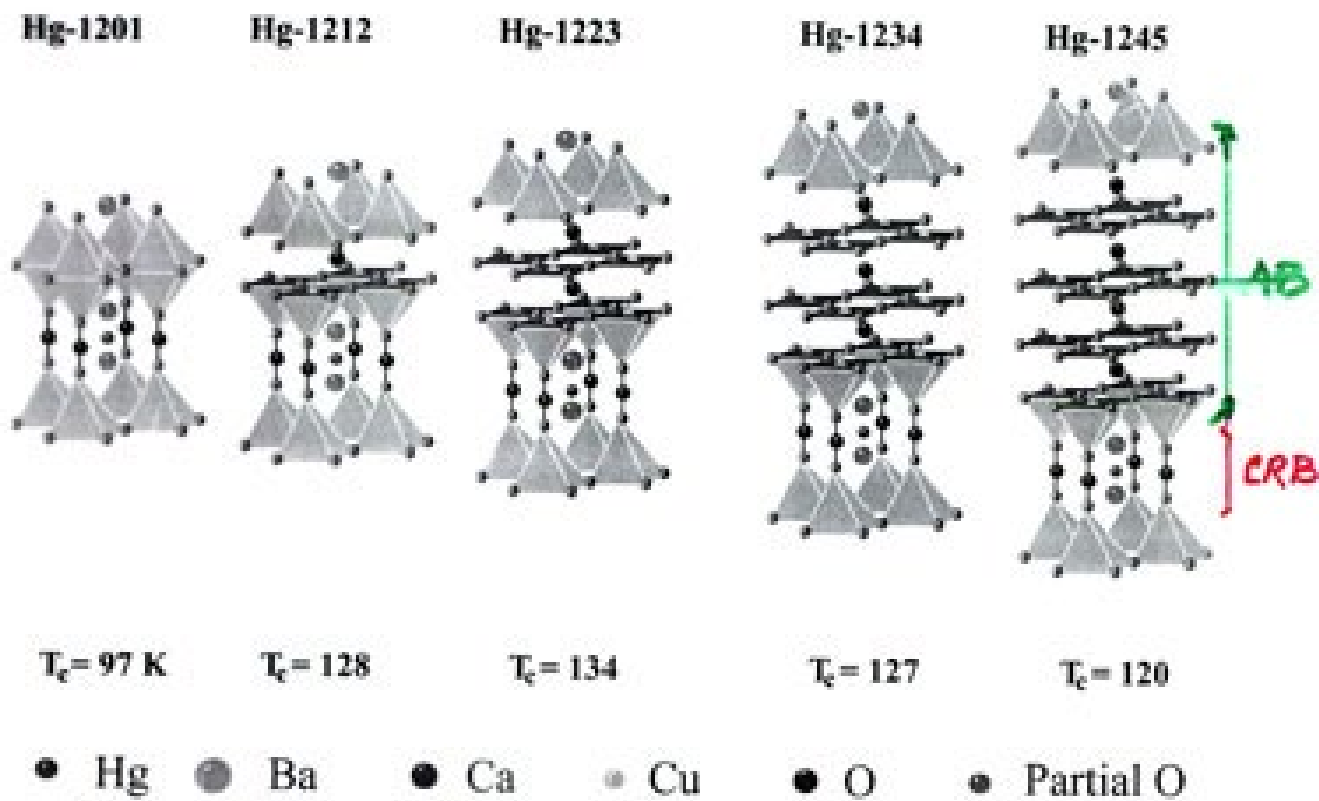
Laboratorium für Festkörperphysik, ETH Höggerberg,
8093 Zürich, Switzerland

THE recent discovery¹ of superconductivity below a transition temperature (T_c) of 94 K in $\text{HgBa}_2\text{CuO}_{4+\delta}$ has extended the repertoire of high- T_c superconductors containing copper oxide planes embedded in suitably structured (layered) materials. Previous experience with similar compounds containing bismuth and thallium instead of mercury suggested that even higher transition temperatures might be achieved in mercury-based compounds with more than one CuO_2 layer per unit cell. Here we provide support for this conjecture, with the discovery of superconductivity above 130 K in a material containing $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{1+x}$ (with three CuO_2 layers per unit cell), $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+x}$ (with two CuO_2 layers) and an ordered superstructure comprising a defined sequence of the unit cells of these phases. Both magnetic and resistivity measurements confirm a maximum transition temperature of ~ 133 K, distinctly higher than the previous established record value of 125–127 K observed in $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ (refs 2, 3).

The structural similarity of $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Hg-120; ref. 1) to a member of the thallium-containing family of copper oxides, $\text{TlBa}_2\text{CuO}_5$ (Tl-1201), suggests the existence of compounds with the general composition $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta}$. The transition temperatures of the thallium-containing analogues, $\text{TlBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+3}$, range from <10 K ($n=1$, ref. 4) to ~ 110 K ($n=3$, ref. 5). In this sense, transition temperatures exceeding 100 K may be expected also in the Hg-Ba-Ca-Cu-O



Schematic Structures of the Hg-Compounds



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HBCCO 1223: $T_c = 134 \text{ K}$ at ambient; 164 K at 31 GPa

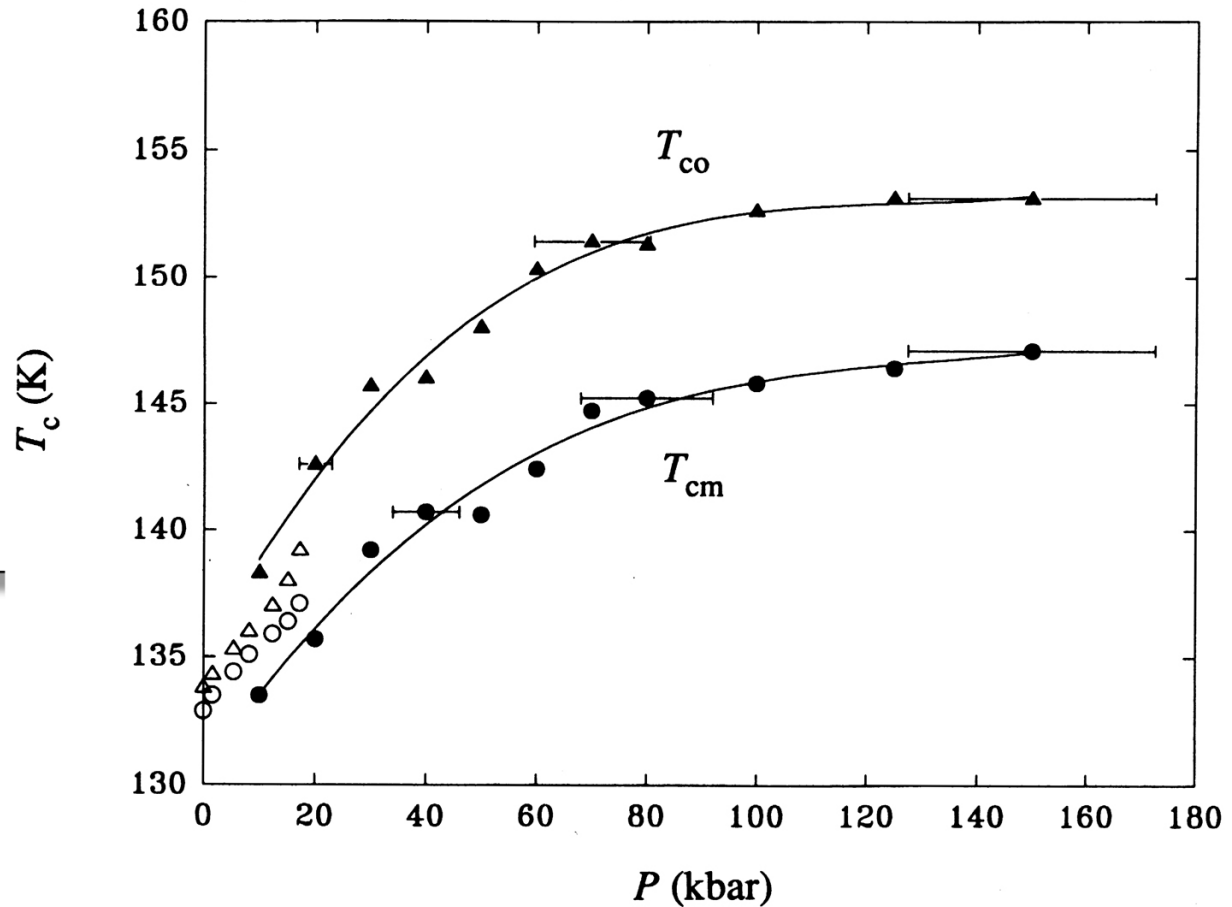
Superconductivity above 150 K in $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ at high pressures

C. W. Chu, L. Gao, F. Chen, Z. J. Huang,
R. L. Meng & Y. Y. Xue

Department of Physics and Texas Center for Superconductivity,
University of Houston, Houston, Texas 77204-5932, USA

RECENTLY, superconductivity at >130 K was reported^{1,2} in multi-phase samples of the compound system $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta}$ ($\text{Hg-12}(n-1)n$) with $n = 1, 2, 3, \dots$. Single-phase Hg-1223 was subsequently synthesized and found to be superconducting at a record temperature of 135 K (ref. 3). It remains to be seen if a much higher transition temperature (T_c) than 135 K can be achieved in this compound system. The application of pressure generally increases the T_c of underdoped layered cuprates, with a pressure derivative of T_c that decreases with increasing doping^{4,5}. Preliminary studies on Hg-1223 ⁶ showed a pressure-induced increase in T_c without any sign of saturation up to 17 kbar. Here we report the observation of superconductivity at up to 153 K in Hg-1223 at 150 kbar, with a main transition at 147 K. This observation provides an indication that superconductivity at >150 K may be possible in this system at ambient pressure, if suitable forms of chemical substitution can be found.

(Nature 1993)



Superconductivity up to 164 K in $\text{HgBa}_2\text{Ca}_{m-1}\text{Cu}_m\text{O}_{2m+2+\delta}$ ($m=1, 2,$ and 3) under quasihydrostatic pressures

L. Gao, Y. Y. Xue, F. Chen, Q. Xiong, R. L. Meng, D. Ramirez, and C. W. Chu

Department of Physics and Texas Center for Superconductivity, University of Houston, Houston, Texas 77204-5932

J. H. Eggert and H. K. Mao

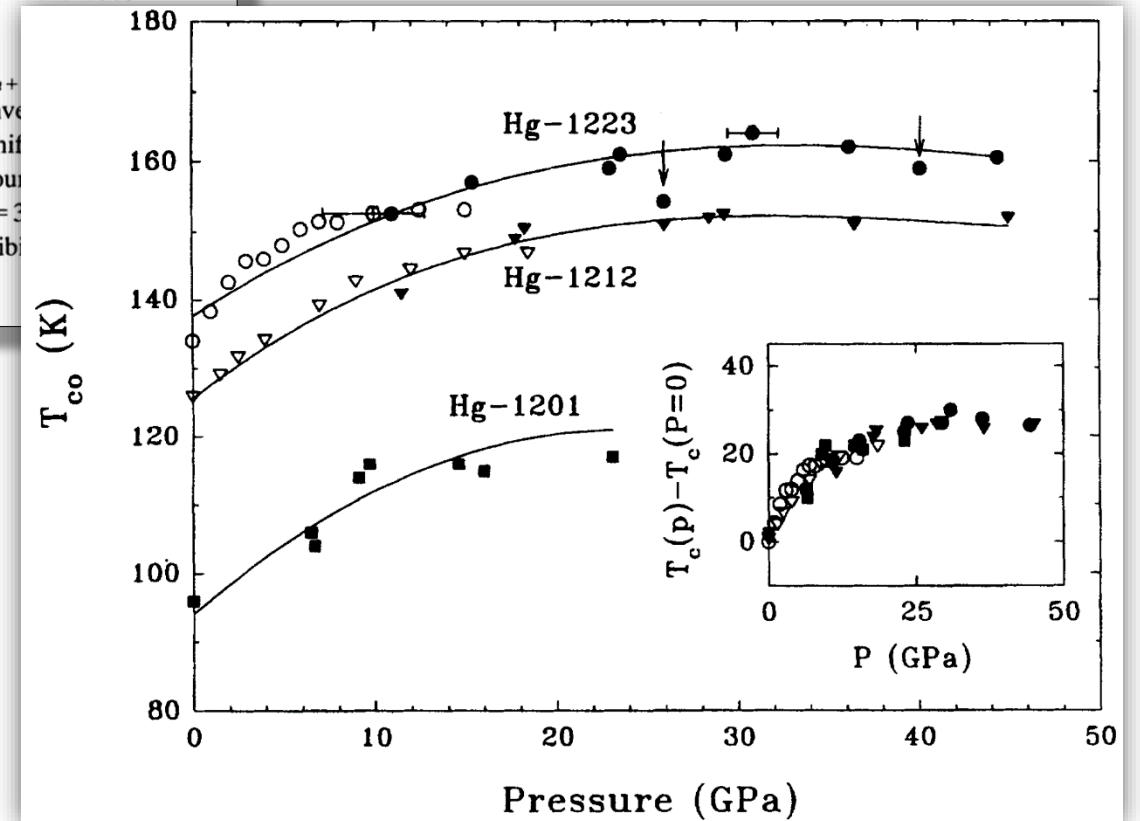
Geophysical Laboratory, Carnegie Institution of Washington, 5251 Broad Branch Road NW, Washington, D.C. 20015-1305

(Received 24 May 1994)

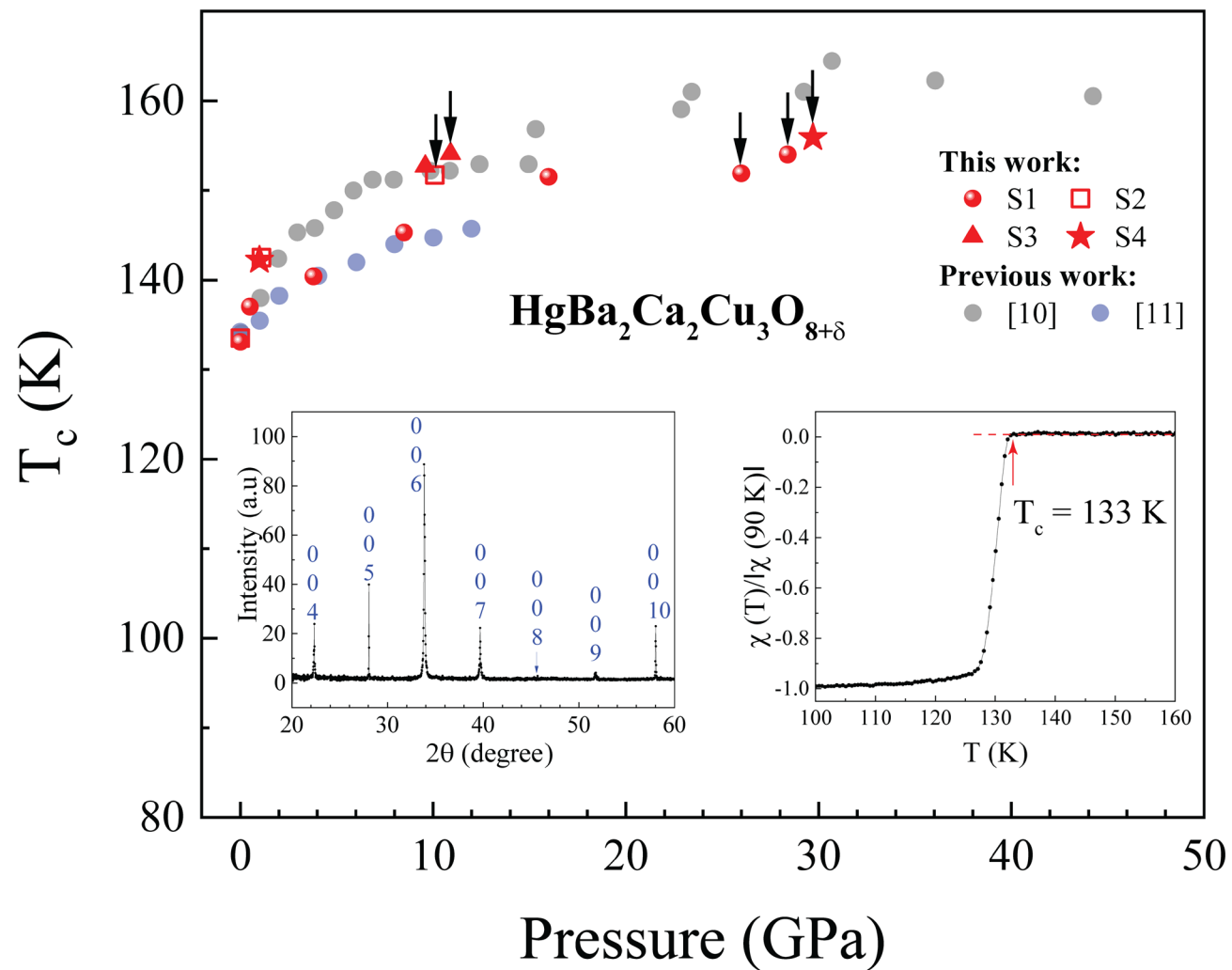
The superconducting transition temperatures (T_c 's) of optimally doped $\text{HgBa}_2\text{Ca}_{m-1}\text{Cu}_m\text{O}_{2m+2+\delta}$ (Hg 1:2: $m-1$: m) with $m=1, 2,$ and 3 and $\text{Hg}_{1-x}\text{Pb}_x\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ [Hg(x Pb) 1:2:2:3] have been investigated resistively under quasihydrostatic pressures up to 45 GPa. There seems to be a universal upward shift of T_c under pressure, regardless of m , for all Hg 1:2: $m-1$: m , implying a common origin for all compounds. Record high T_c 's of 164, 154, and 118 K were reached for the optimally doped Hg 1:2: $m-1$: m with $m=3, 2,$ and $1,$ respectively. However, the T_c enhancement is suppressed by Pb substitution, suggesting the possibility that Hg plays an important role in these compounds.

Large and universal T_c enhancement in Hg-12($n-1$)

PRB 50, 4260(R) (1994)

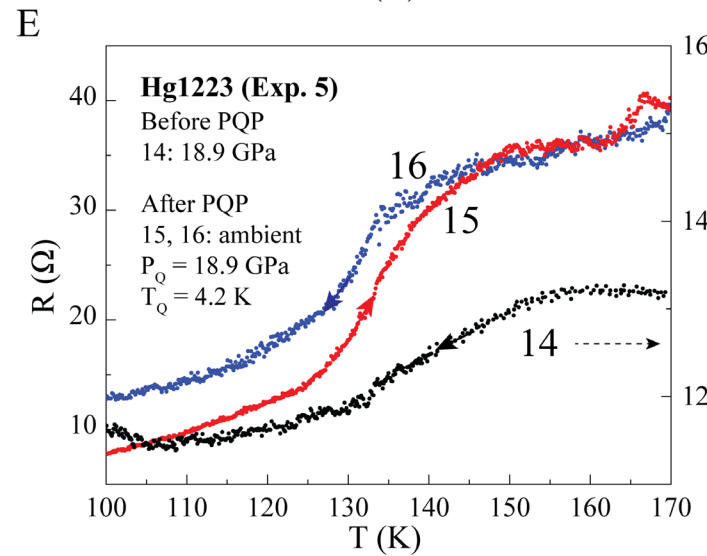
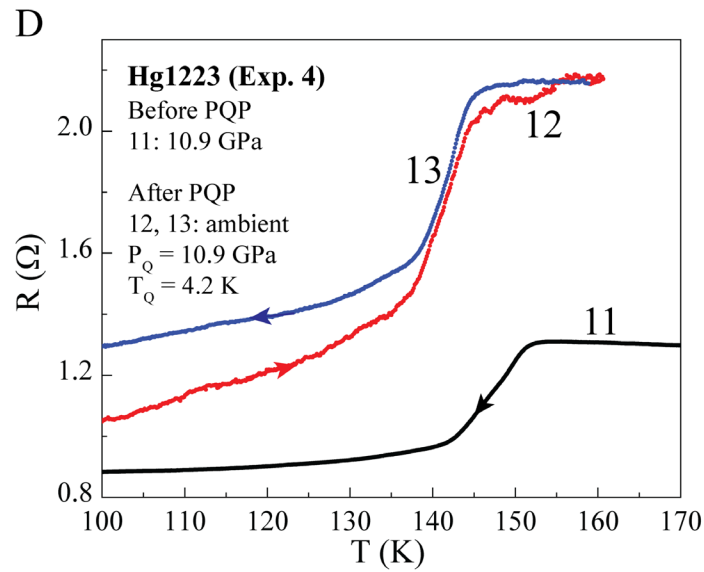
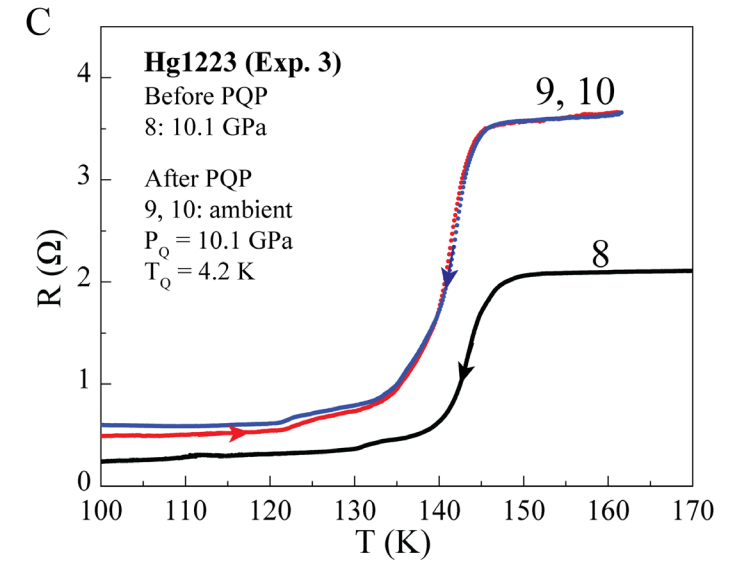
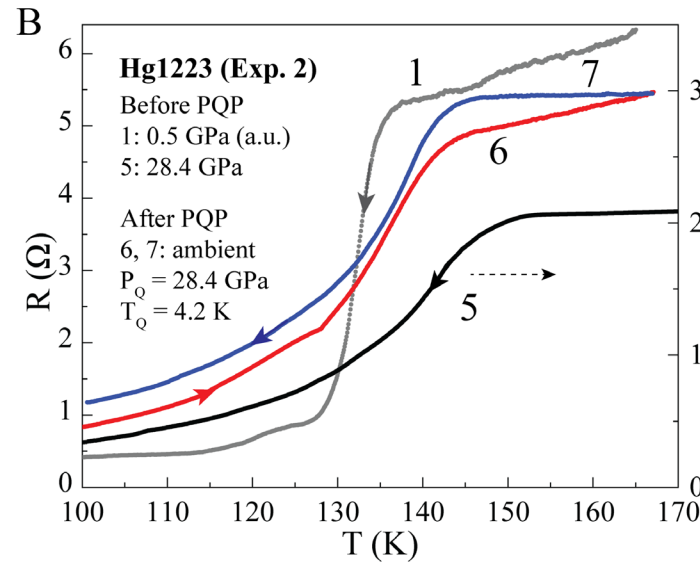
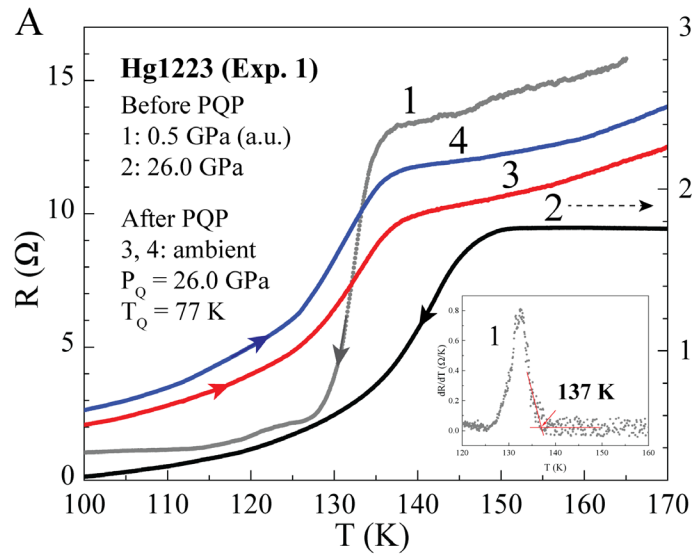


- Search for Record High Ambient Pressure T_c in HBCCO via PQP



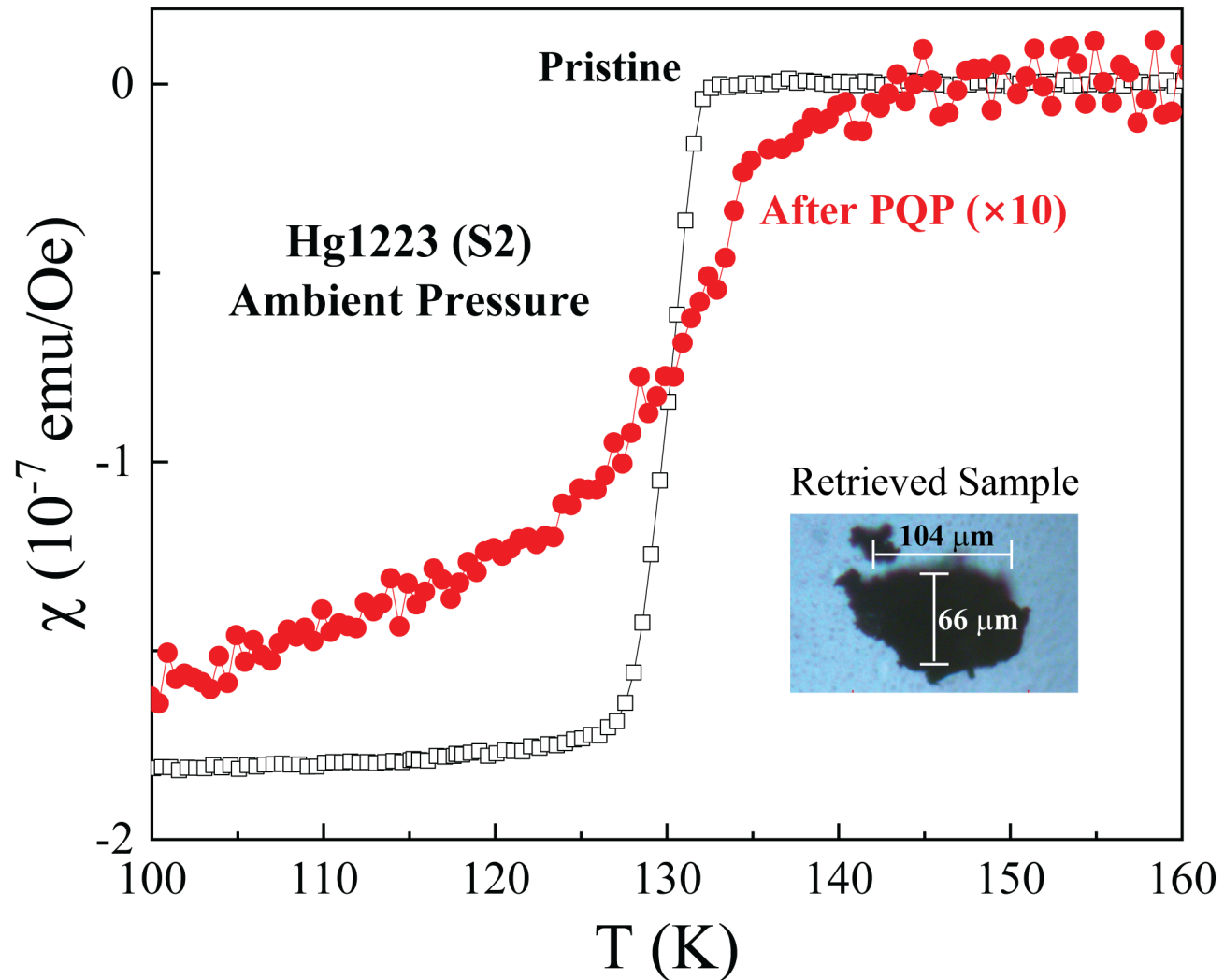


Hg1223 – RTs of PQP (Exp. 1-5)



F

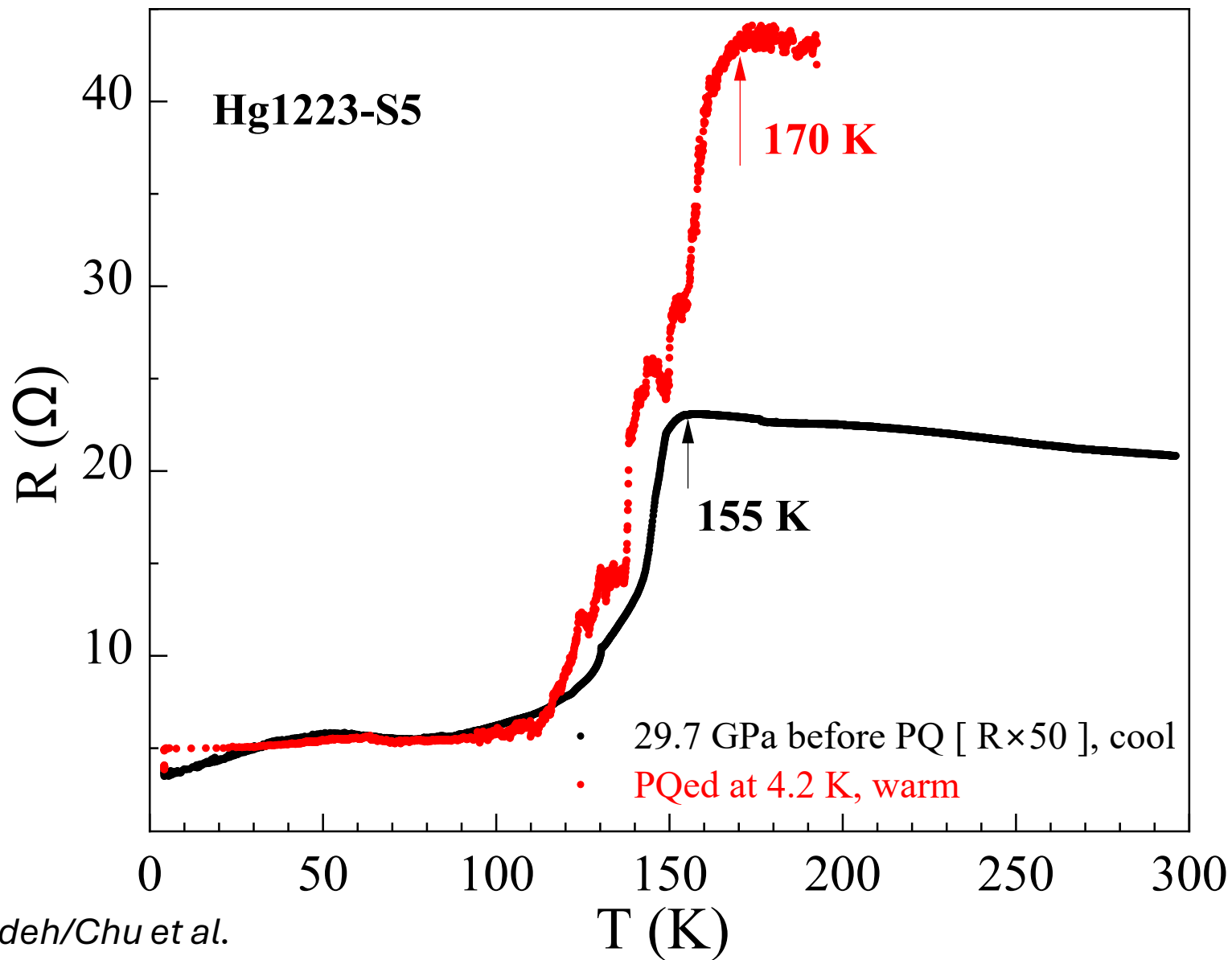
Exp.	1	2	3	4	5
Sample	S1	S1	S2	S3	S4
T_Q (K)	77	4.2	4.2	4.2	4.2
P_Q (GPa)	26.0	28.4	10.1	10.9	18.9
Target T_c (K) at P_Q	152	154	152	154	158
Retained T_c (K) at ambient P	139	147	147	149	151



DC magnetic results for pristine Hg1223 crystal #1 (source of S2) before PQP (black squares) and following retrieval from the DAC after PQP (red circles). Inset: part of Hg1223 (S2) retrieved from the DAC after PQP.

Sign of >170 K SC at ambient pressure?

2024/12/03 eve -04



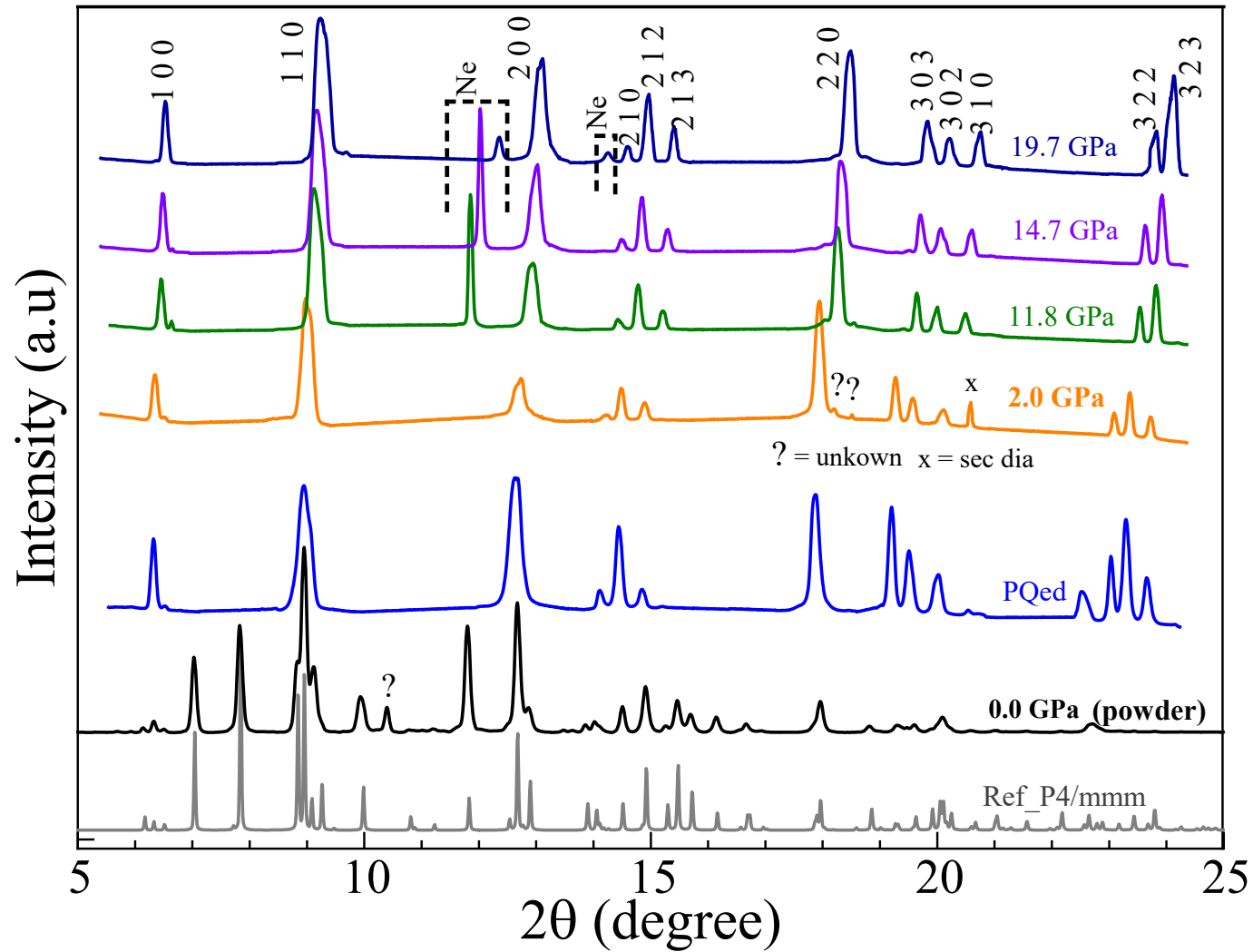
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Chu/Deng Group

Deng, Habamahoro, Safezoddeh/Chu et al.

XRD at 300 K and different P's

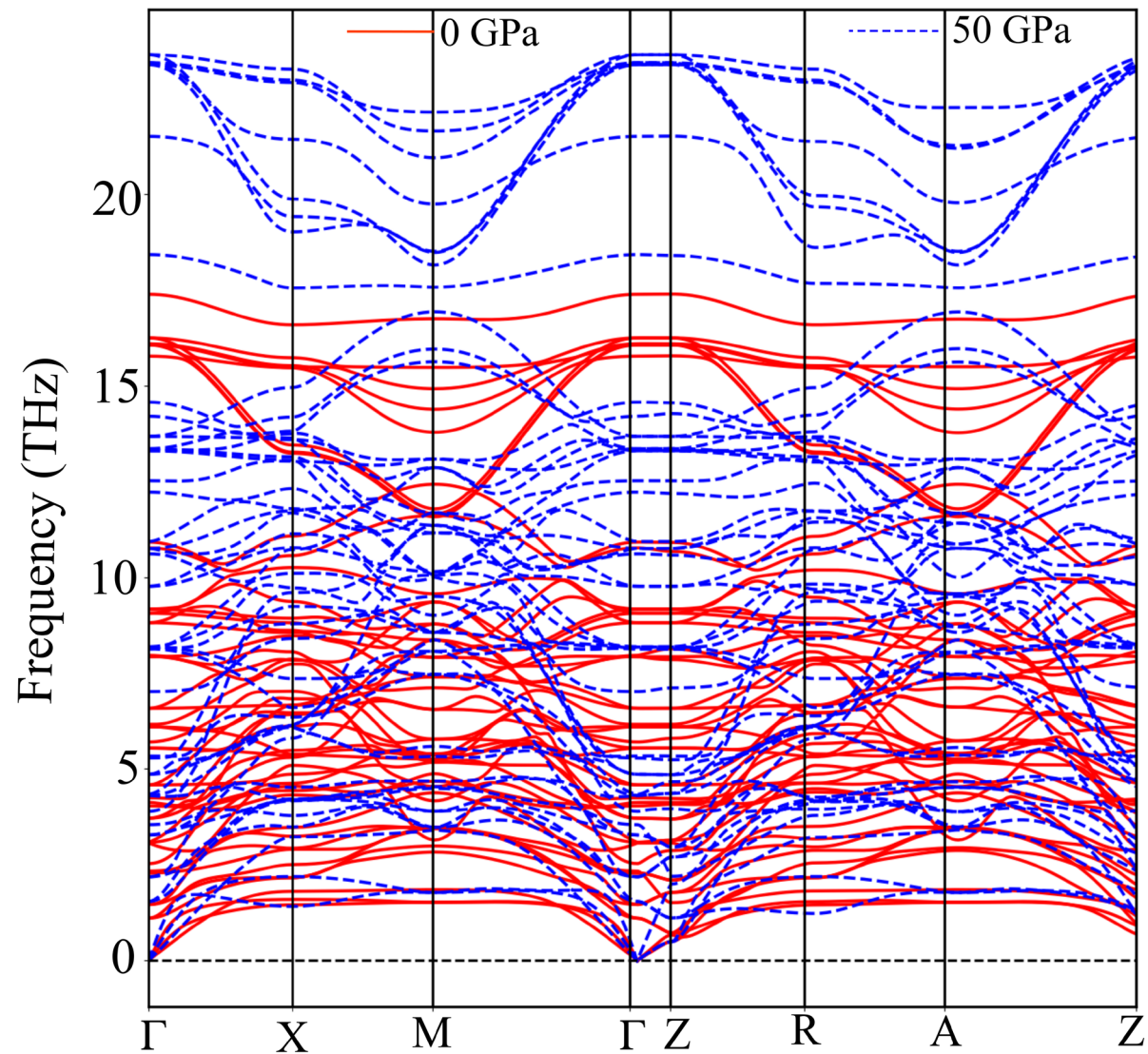
Hg1223- S6 [$P_Q = 19.7$ GPa, $T_Q = 50$ K]



- Structural transition
- ↓
- Lattice parameter
- ↓
- Bond angle/length

No structural transition generated under pressure!

DFT – Phase Stability



Knows, Unknowns & Perspective

- Knowns: $T_c \sim 151$ K , bulk, no PQP induced structural transition, defects, highly reproducible
- Crucial Parameters & their correlations for successful PQP's: T_c , J_c and mechanical properties
- PQP Control and Performance: micro-and/or nano-structures, P_Q and T_Q
- Characterization of the PQed phases inside and outside DAC
- Prerequisites for Materials for optimal PQP: lattice, electronic(FSTC) and/or other metastabilities(FSTC)
- Temporal and thermal stabilities
- $T_c(P_Q)$ before PQP $>$ T_c after PQed at P_Q
- Quantity: from PQP to application
- Challenges: small size, misalignment from PQP to characterization

***This may just be the beginning of an exciting Journey of PQP !!!
(Hope AI will help shorten the journey!!!)***

Deng et al., Ambient-pressure 151-K superconductivity in $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ via pressure quench, Proc. Natl. Acad. Sci. U.S.A. 123 (11) e2536178123, <https://doi.org/10.1073/pnas.2536178123> (2026).

Prasankumar et al., The path to room-temperature superconductivity: A programmatic approach, Proc. Natl. Acad. Sci. U.S.A. 123 (11) e2520324123, <https://doi.org/10.1073/pnas.2520324123> (2026).

Thank you!

Any comments and suggestions are welcome!