Shoji Tanaka

1927 - 2011

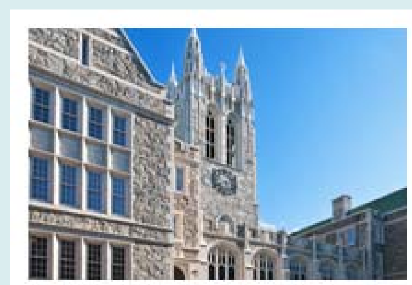
Eliashberg-McMillan Parameters of Polysulfur Nitride, $(SN)_x$

Session W21: Focus Session:
Search for New Superconductors- Electron-Phonon Coupling
W21.00014, Room 254A, Thursday, March 1, 2012, 1:51 PM

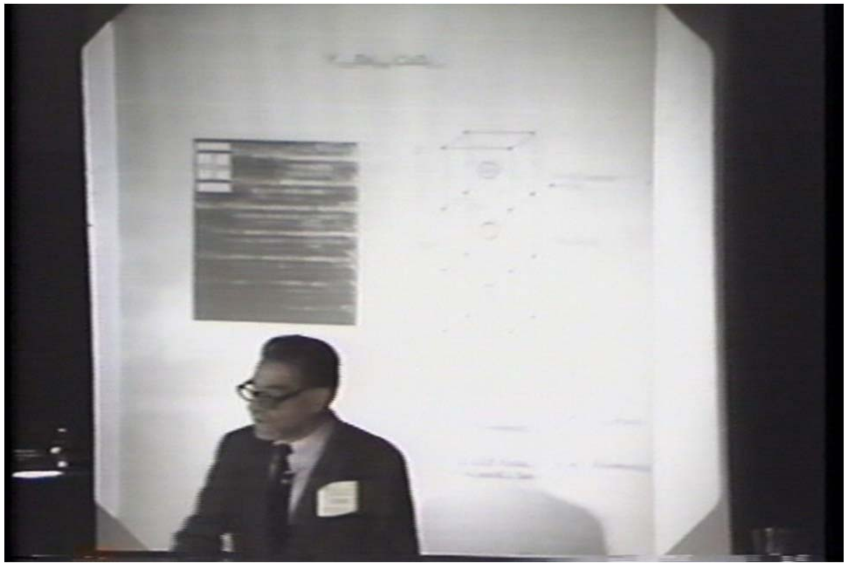
Paul Michael Grant

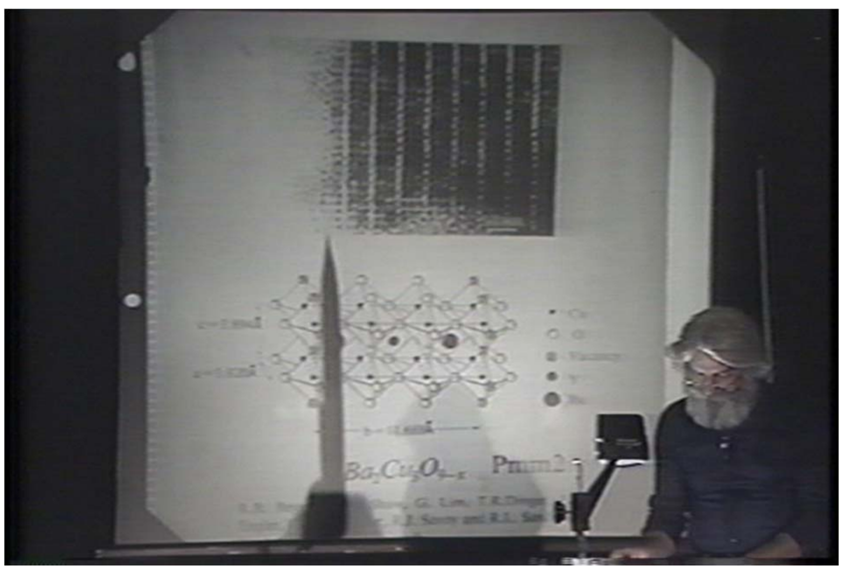
Aging IBM Pensioner

(research supported under the IBM retirement fund)



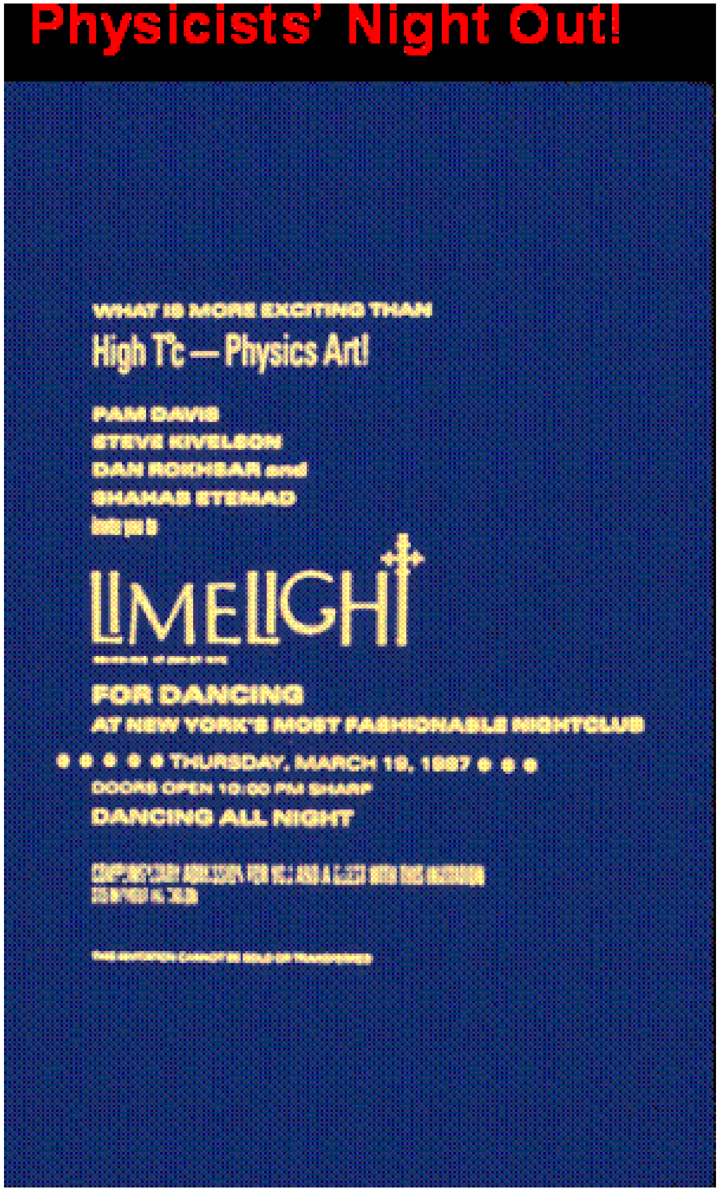
25 Years Since Woodstock!





Ballroom, NY Hilton – 1987
3000+ Nerds

Watch Physicists Behaving Badly!

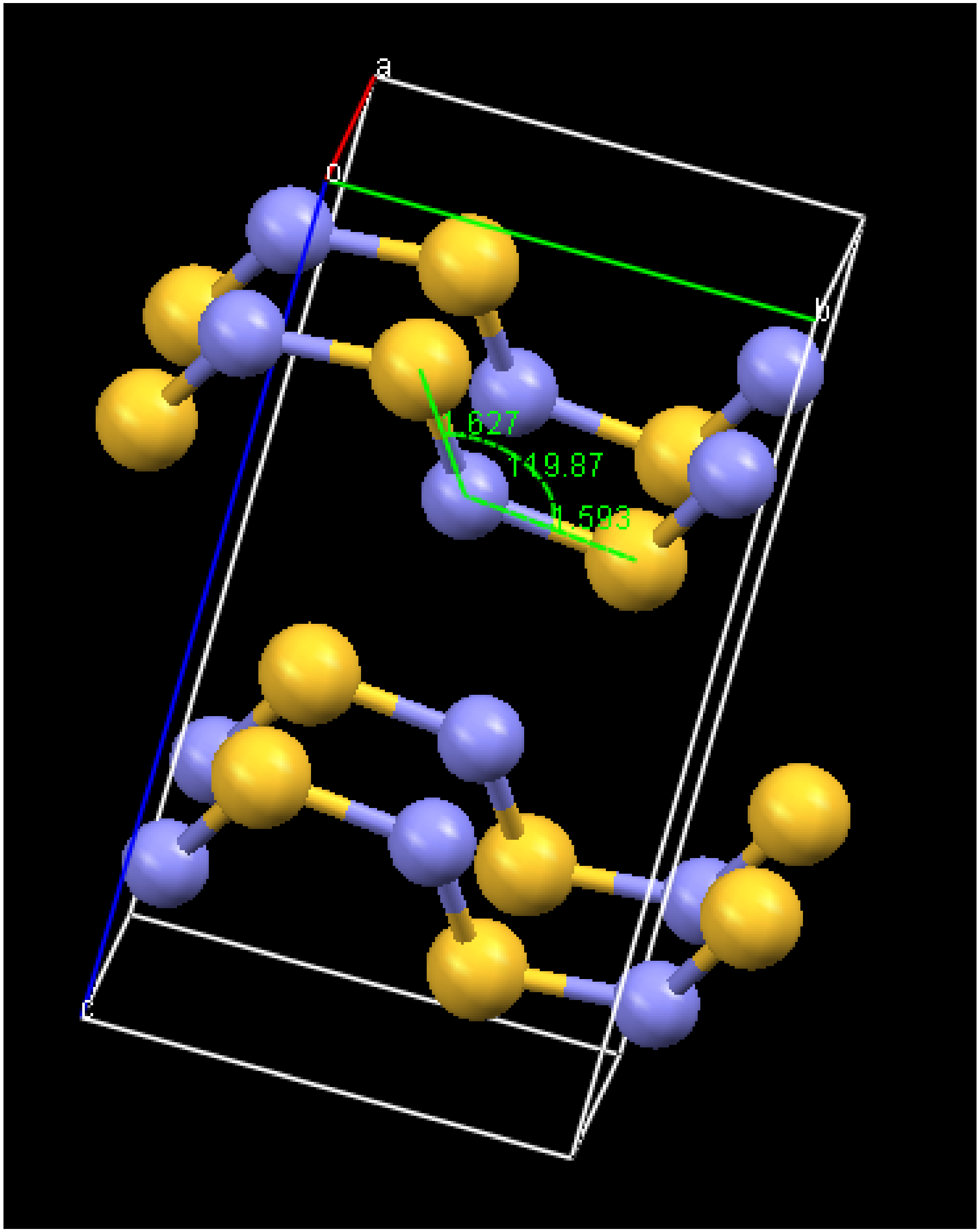


Videos Available on www.w2agz.com
Consult speaker for details
(Cuidao! Los archivos estan muy grandes!)

- Race for the Superconductor (NOVA, 1988)
- The Race for the Prize (BBC, 1988)
- Superconductivity Challenge for America (Reagan, 1987)
- APS Woodstock of Physics (March, NYC 1987)
- MRS Altamont of Materials (April, Anaheim 1987)
- Berkeley People's Park (June, Berkeley 1987)
- IBM 1987-88 Press & Corporate Newsreels

Grazie - Thanks

- Quantum-Espresso Democritos Consortium
  - Paolo Gianozzi & Eyvaz Isaev
  - ...and many more in the PW_Forum community
- JPL/CalTech
  - Paul von Allmen & Trinh Vo
  - ...for assistance using the “Antpile Cluster”



Fabrication of $(SN)_x$

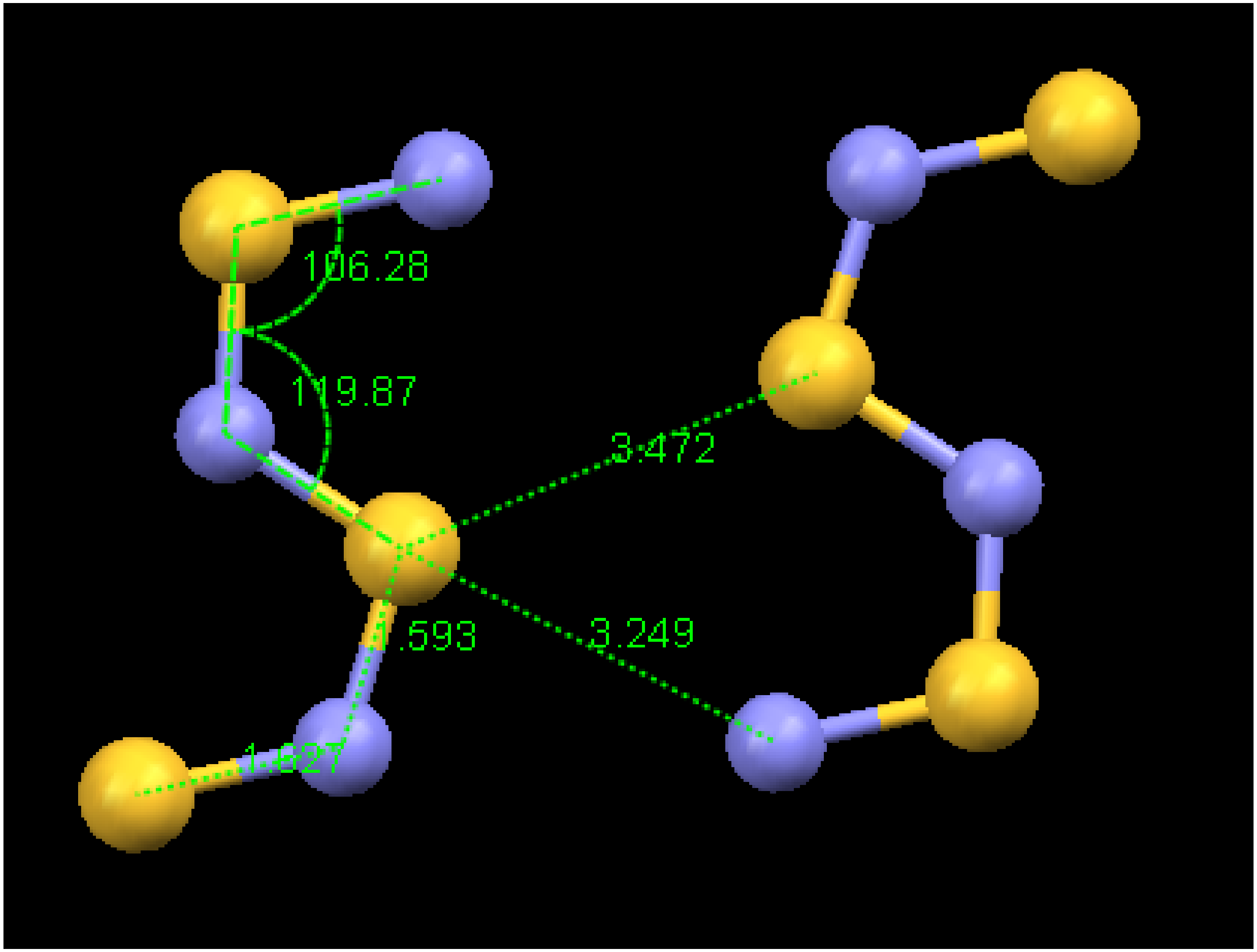
Frank Playfair Burt (1910)

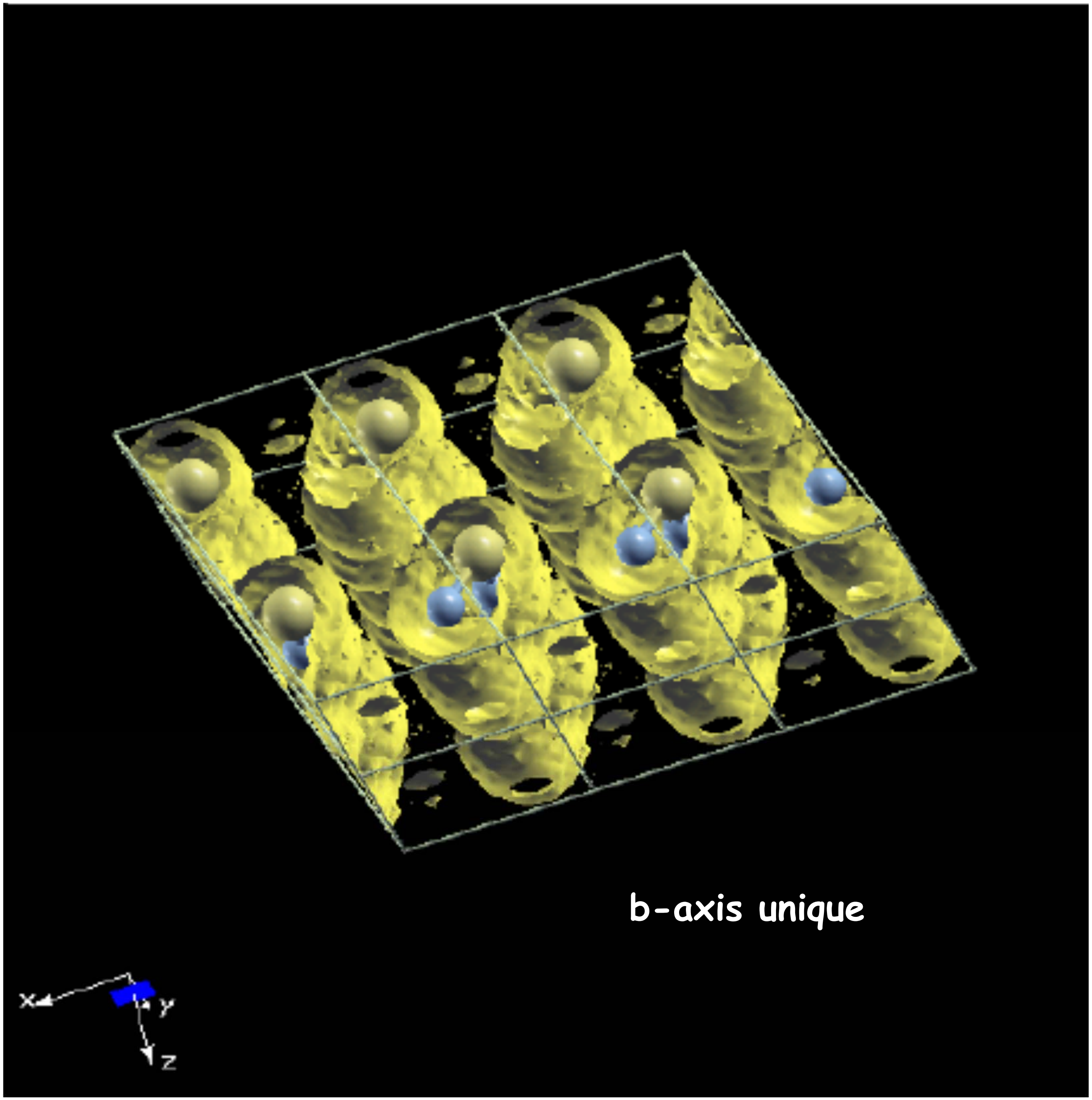
- Thermally split S_4S_4 into S_2N_2
- Ag catalyze to $(SN)_x$
- It's a metal! (Cuidado!)

Structure of $(SN)_x$

Cohen, et al., (1976)

- Monoclinic $P2_1/c$
- a=4.485, b=3.767, c=8.452, β=106.43
- 4 N, 4 S, 2 chains
- S=32.06, N=14.007
- S[Ne]$3s^2 3p^4$, (3d ?) N[He]$2s^2 2p^3$
- 44 electrons
- 22+ bands
- Many phonon IRs (24)


106.28
119.87
3.472
1.593
3.249
1.627



b-axis unique
x
y
z

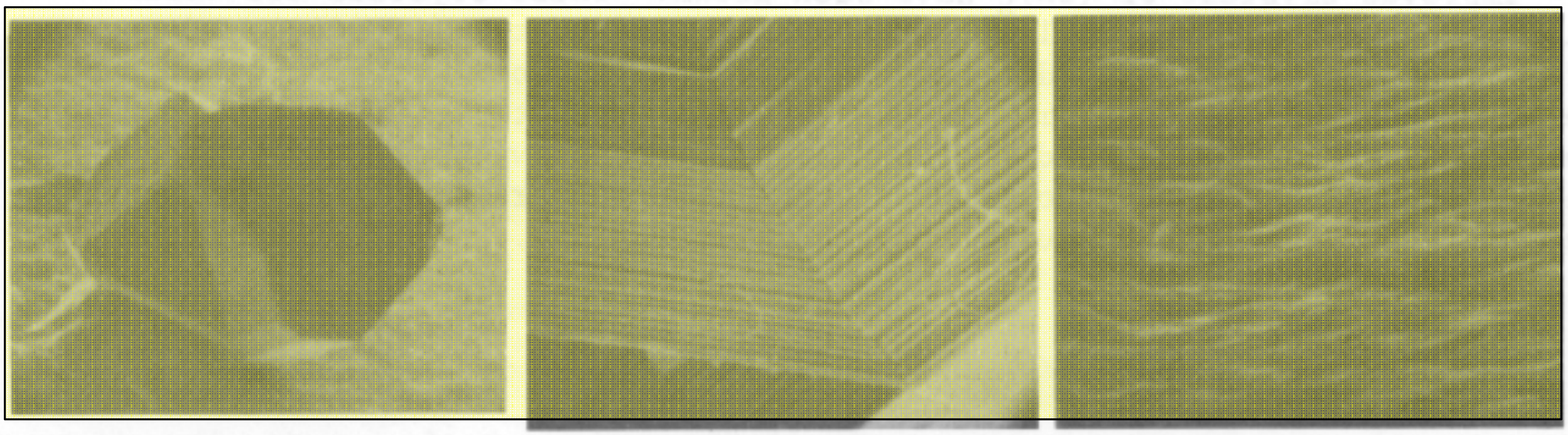
$(SN)_x$: "It's a Yellow Bunch of Straws"

The preparation and characterization of crystals of the superconducting polymer, $(SN)_x$

G.B. Street, H. Arnal, W.D. Gill, P.M. Grant, R.L. Greene

IBM Research Laboratory, San Jose, CA 95193 USA

Received 2 July 1975. Available online 9 May 2003. Communicated by R. A. Huggins.



a b c

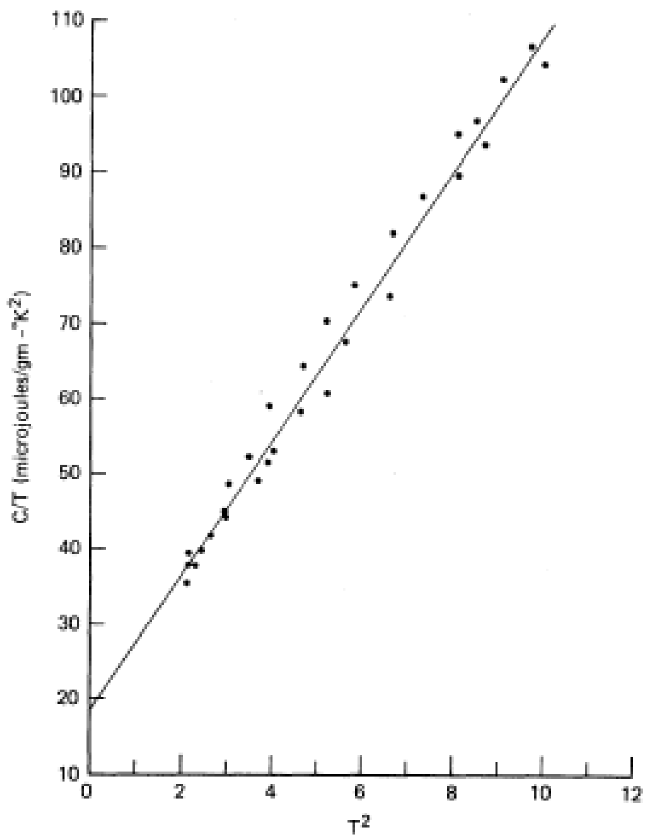
FIG. 2

(a) Twinned crystal of $(SN)_x$ magnified 20 times.
(b) Fibres parallel to b axis of each crystal magnified 250 times.
(c) Fibres terminating on faces at angles to b axis magnified 2,400 times.

First IBM Experiment (1974)

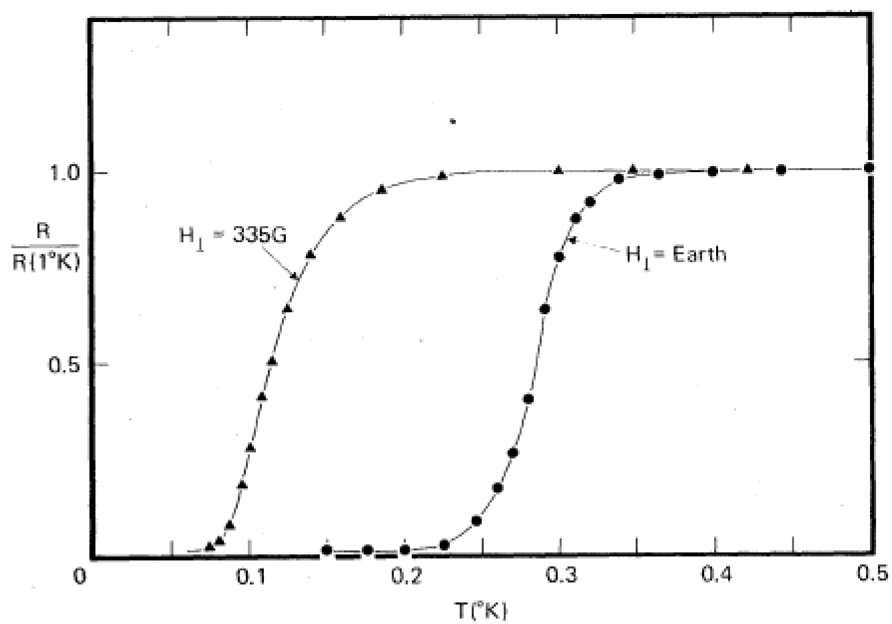
Low-Temperature Specific Heat of Polysulfur Nitride, $(SN)_x$

R. L. Greene, P. M. Grant, and G. B. Street
IBM Research Laboratory, San Jose, California 95193
(Received 25 September 1974)



1975: Superconductivity Discovered at 330 degrees in Polysulfurnitride, (SN)x

...but the units are in millikelvin !



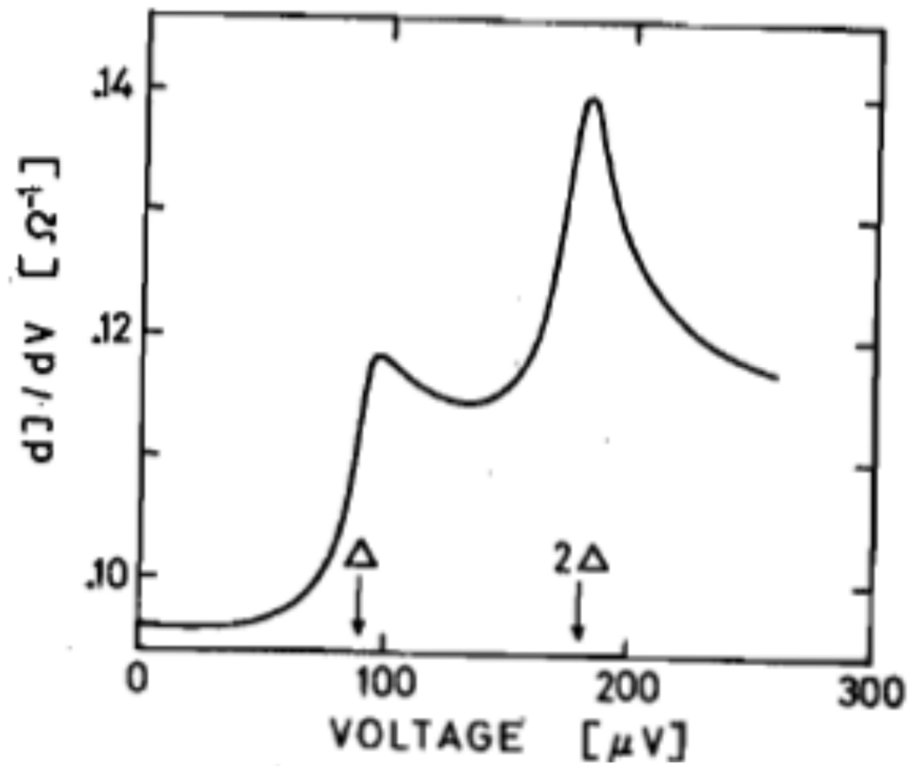
Superconductivity in Polysulfur Nitride $(SN)_x$

R. L. Greene and G. B. Street
IBM Research Laboratory, San Jose, California 95193

and

L. J. Suter*†
Department of Physics, Stanford University, Stanford, California 94305
(Received 27 January 1975)

Precursor to a Nobel Prize



Tunneling Investigation of Superconducting $(SN)_x$

G. Binnig and H.E. Hoenig
Physikalisches Institut der Universität Frankfurt, Germany

Z. Physik B 32, 23–26 (1978)

Lattice Specific Heat of $(SN)_x$

Specific heat of polysulfur nitride, $(SN)_x$

J. M. E. Harper*
Department of Applied Physics, Stanford University, Stanford, California 94305

R. L. Greene, P. M. Grant, and G. B. Street
IBM Research Laboratory, San Jose, California 95193
(Received 22 July 1976)

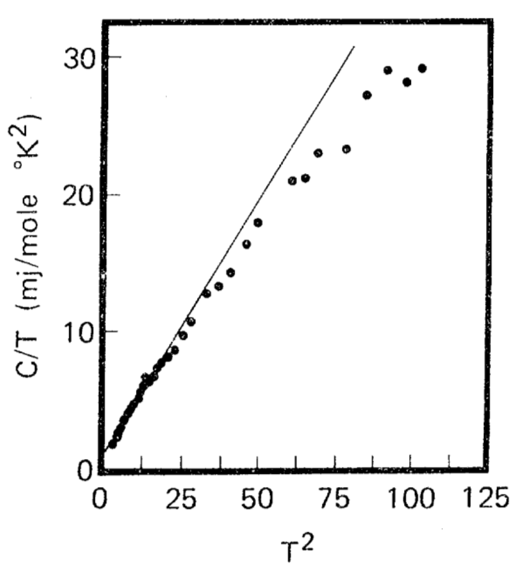


FIG. 2. Specific heat of $(SN)_x$ from 1.5 to 10 °K. Solid line shows least-squares fit to data from 1.5 to 3.2 °K given by $C=\gamma T+\beta T^3$ with $\gamma=0.83\pm0.09$ mJ/mole °K^2 and $\beta=0.41\pm0.02$ mJ/mole °K^4.

From Experiment:

- $\Theta_D \sim 148$
- $\lambda \sim 0.31$
- T_C = 330 mK
- Can We Compute λ ?

Coming Up!

DFT Computational Toolbox

- Software (PWscf – Quantum Espresso)
  - Self Consistent Field (SCF) Parameters
    - Becke-Lee-Yang-Parr (BLYP) Pseudopotential
    - 12 x 12 x 12 Monkhorst-Pack Grid
    - 32 Ry G-lattice cutoff, 10^{-9} Ry convergence
  - non-SCF Inputs
    - Electron & Phonon Bands: 221 k-points navigating BZ
    - Electron-Phonon λ: 2x2x2 MP Grid; 10^{-11} Ry converge
- Hardware
  - JPL Antpile Cluster: 8 Supermicro server boards
  - Two days cpu time

$(SN)_x$

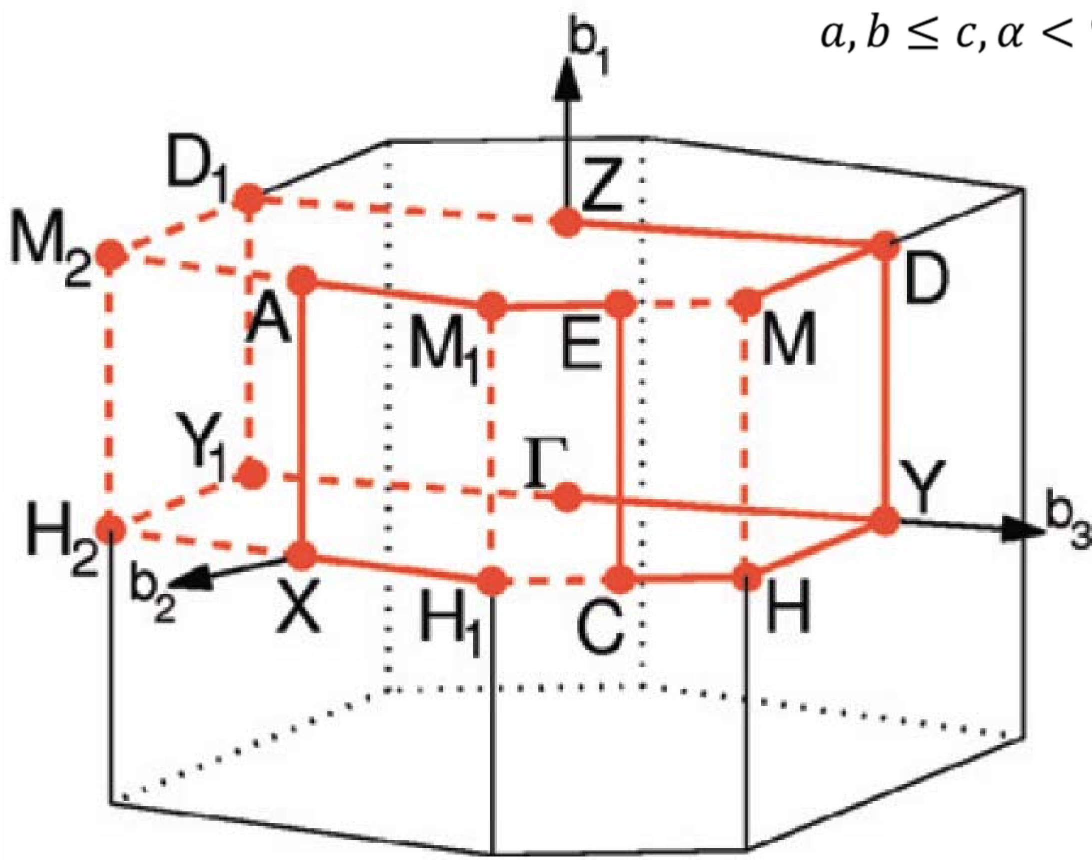
Reciprocal Space

Brillouin zone of MCL lattice

Setyawan, Curtarolo

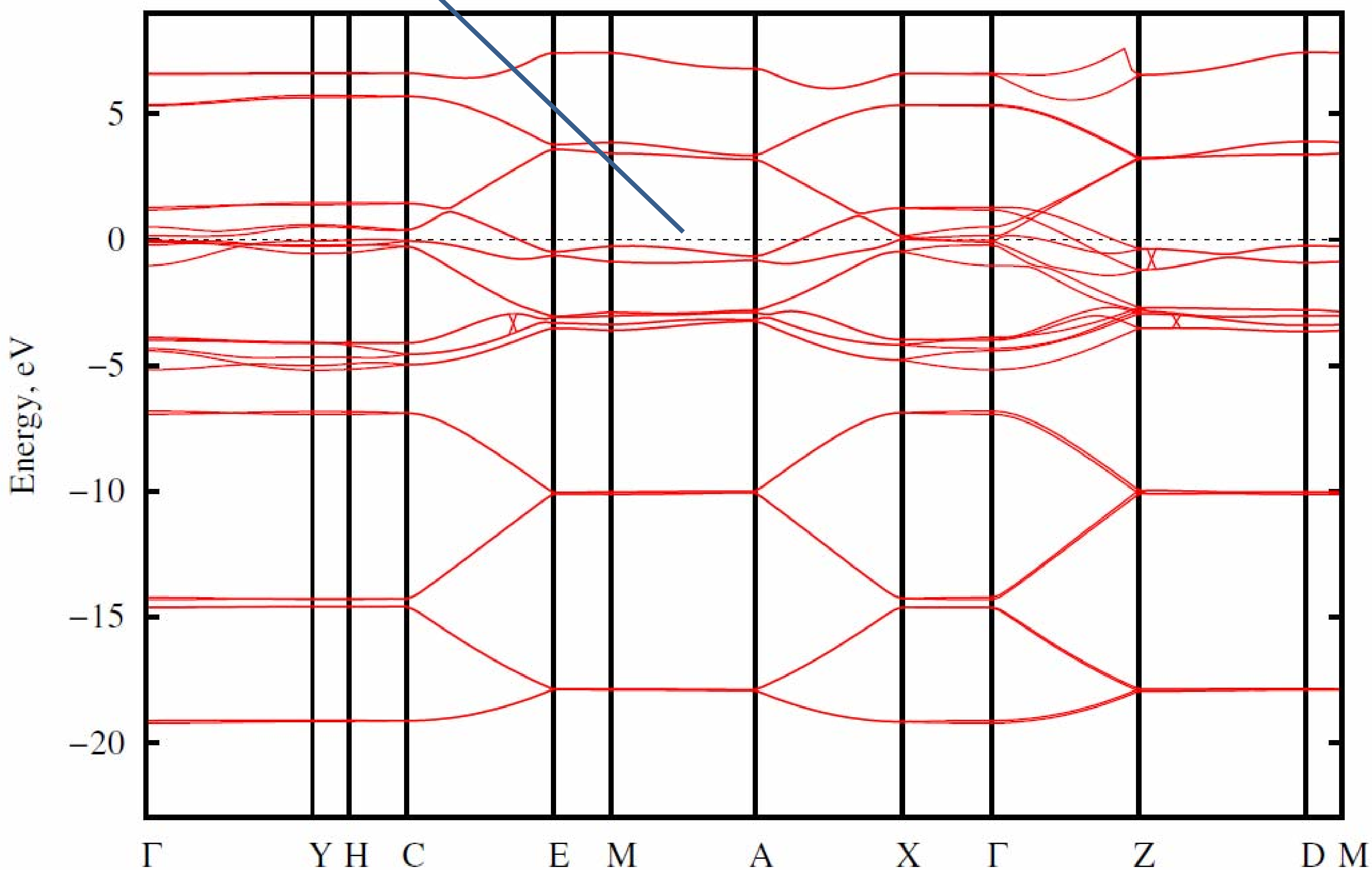
Computational Materials Science 49 (2010) 299–312

$a, b \leq c, \alpha < 90°, \beta = \gamma = 90°$



$(SN)_x$
Electron Band Structure

Note:
Multiple FS Crossings!

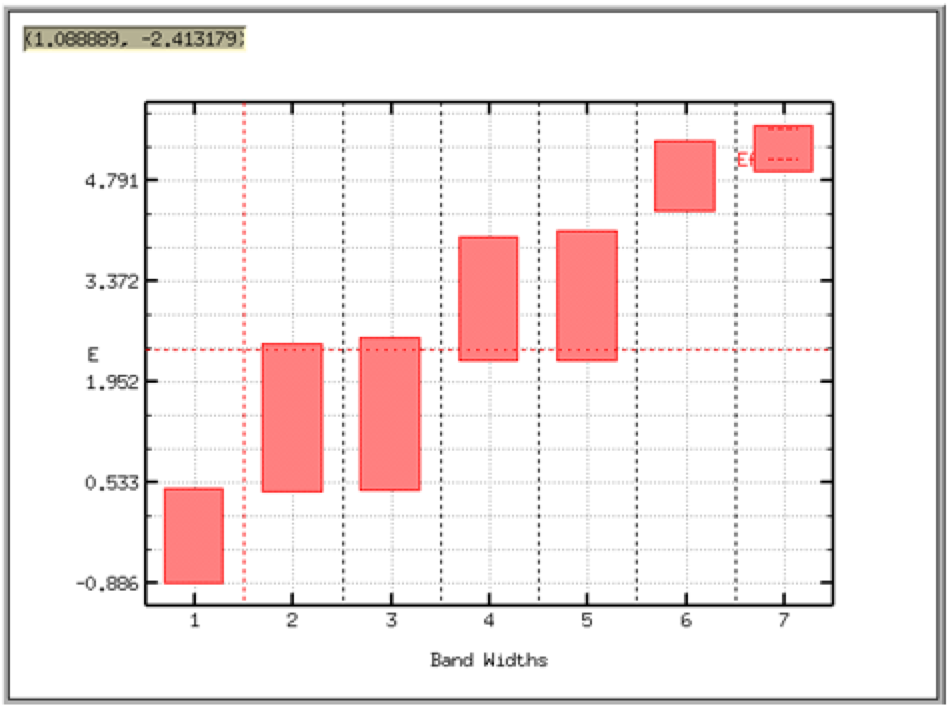


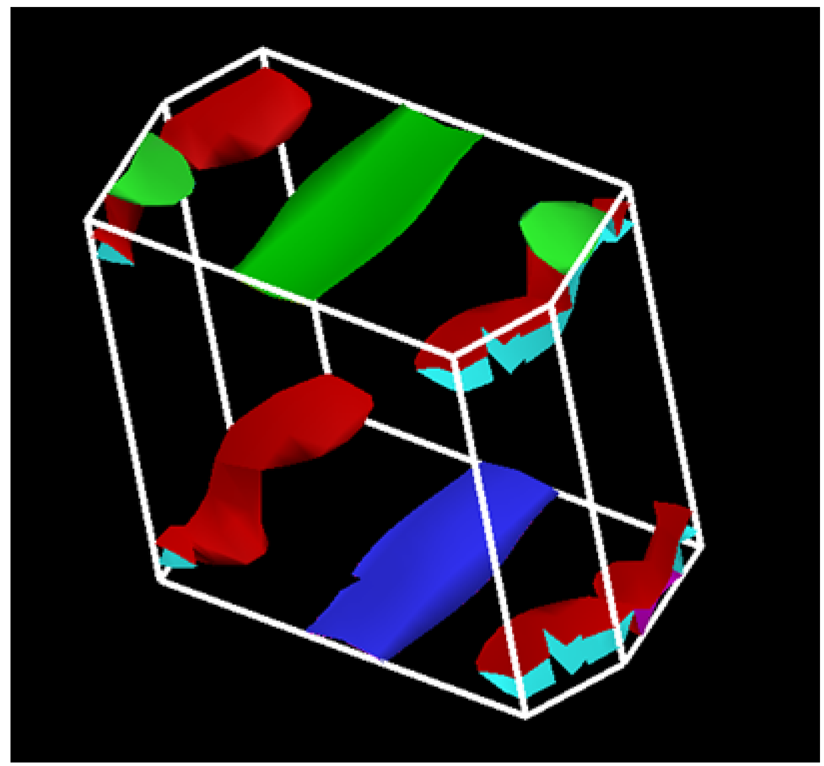
$(SN)_x$

Fermiology

Note!:

- Two band superconductor (a la MgB_2)
- Significant nesting (a la MgB_2)

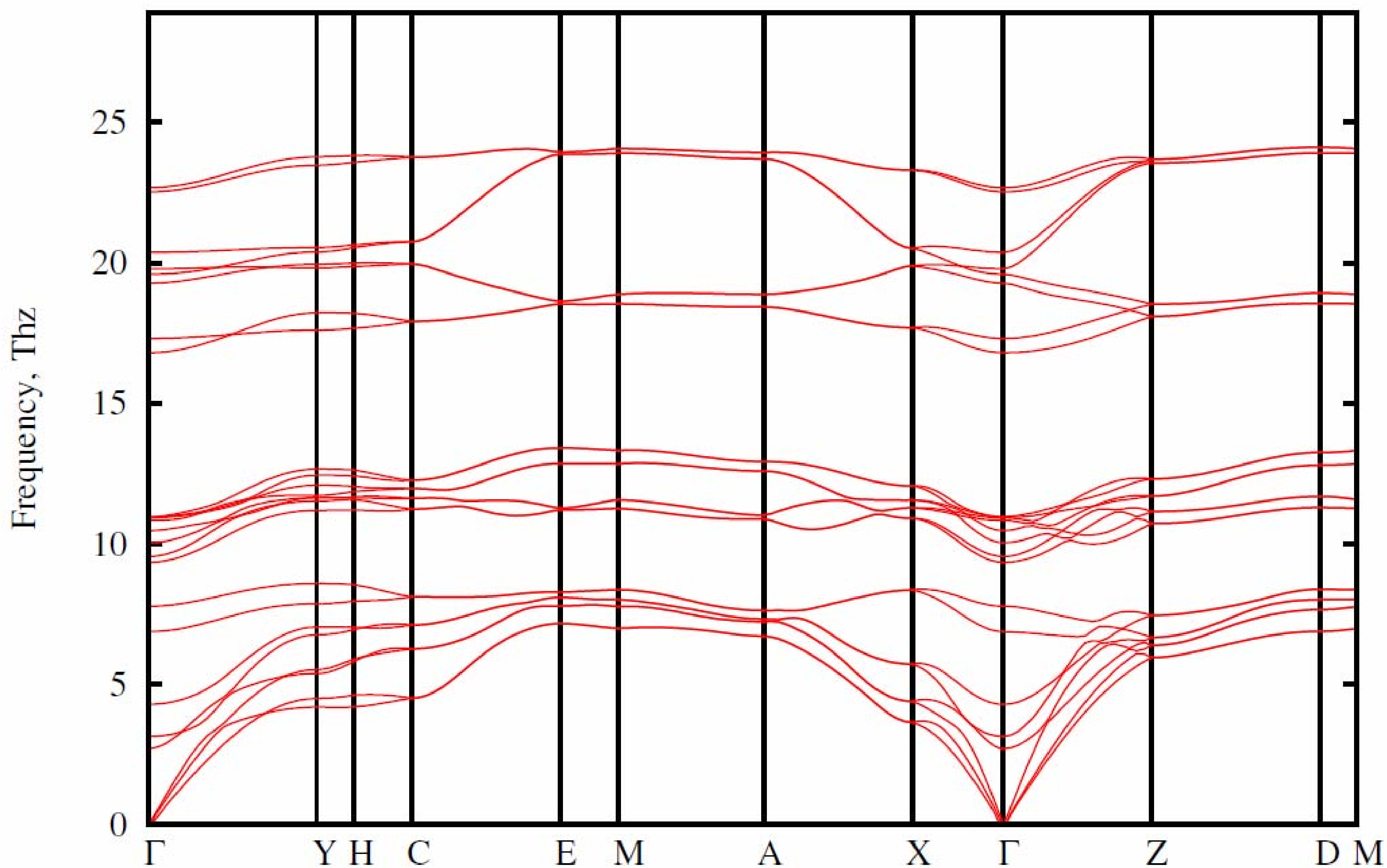




$(SN)_x$

Phonon Dispersion

24 IR's
WOW!



Electron-Phonon Coupling

Eliashberg-McMillan

$$H_{el-ph} = \sum_{\mathbf{k},\mathbf{q},\nu} g_{\mathbf{k+q},\mathbf{k}}^{\mathbf{q}_\nu,mn} c_{\mathbf{k+q}}^{\dagger m} c_{\mathbf{k}}^{n} (b_{-\mathbf{q},\nu}^{\dagger} + b_{\mathbf{q},\nu})$$

$$\lambda_{\mathbf{q},\nu} = \frac{2}{N(\varepsilon_F)\omega_{\mathbf{q},\nu}} \sum_{mn} \sum_{\mathbf{k}} \left| g_{\mathbf{k+q},\mathbf{k}}^{\mathbf{q}_\nu,mn} \right|^2 \delta(\varepsilon_{\mathbf{k+q},m} - \varepsilon_F)\delta(\varepsilon_{\mathbf{k},n} - \varepsilon_F)$$

$$\alpha^2 F(\omega) = \frac{1}{N(\varepsilon_F)} \sum_{mn} \sum_{\mathbf{q},\nu} \delta(\omega - \omega_{\mathbf{q},\nu}) \sum_{\mathbf{k}} \left| g_{\mathbf{k+q},\mathbf{k}}^{\mathbf{q}_\nu,mn} \right|^2 \delta(\varepsilon_{\mathbf{k+q},m} - \varepsilon_F)\delta(\varepsilon_{\mathbf{k},n} - \varepsilon_F)$$

$$\lambda = 2\int_0^\infty \frac{\alpha^2 F(\omega)}{\omega} d\omega = \sum_{\mathbf{q},\nu} \lambda_{\mathbf{q},\nu}$$

Need to compute $g_{\mathbf{k+q},\mathbf{k}}^{\mathbf{q}_\nu,mn}$!

e-p Interaction in the DFT/LDA Formalism

$$g_{\mathbf{k}+\mathbf{q},\mathbf{k}}^{\mathbf{q}_{\nu},mn} = \sqrt{\hbar / 2\omega_{\mathbf{q},\nu}} \left\langle \psi_{\mathbf{k}+\mathbf{q},m} \left| \Delta V_{KS}^{\mathbf{q},\nu} \right| \psi_{\mathbf{k},n} \right\rangle$$

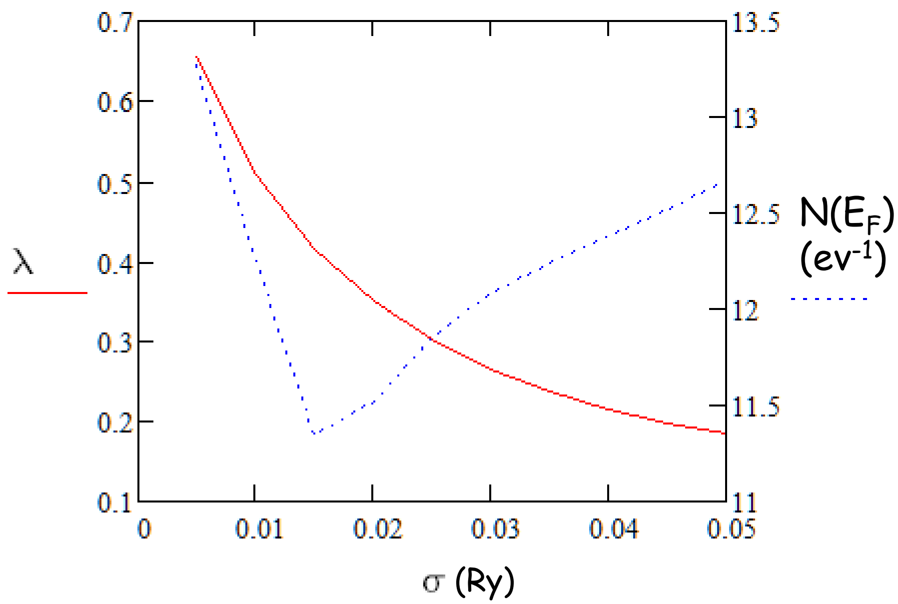
$$\Delta V_{KS}^{\mathbf{q},\nu} = \sum_{\mathbf{R}} \sum_{s} \frac{\partial V_{KS}}{\partial \vec{u}_{s,\mathbf{R}}} \cdot \vec{u}_{s}^{\mathbf{q},\nu} \frac{e^{i\mathbf{q}\cdot\mathbf{R}}}{\sqrt{N}}$$

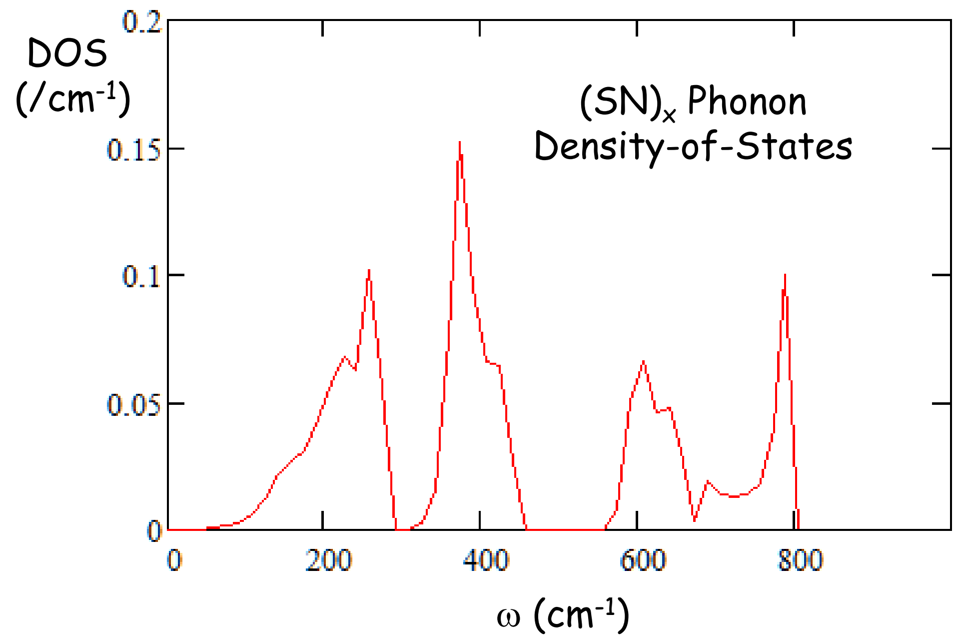
2x2x2 MP Grid; 10-11 Ry converge

$$T_C = \frac{\Theta_D}{1.45} \exp\left(-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)} \right)$$

$(SN)_x$

Electron, Phonon, Electron-Phonon Properties



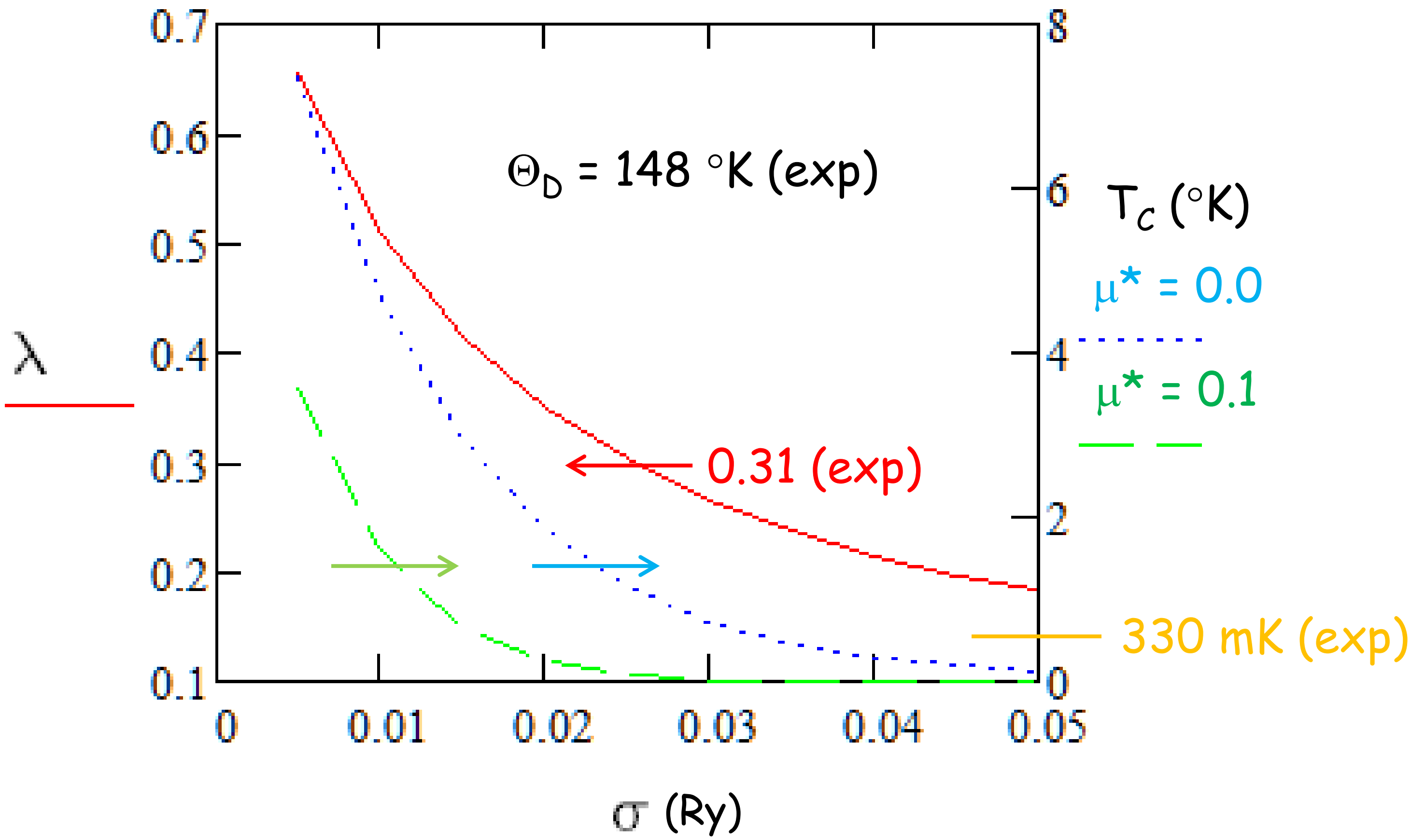


What is σ ?

$(SN)_x$

{according to Eliashberg-McMillan}

$$T_C = \frac{\Theta_D}{1.45}\exp\left(-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right)$$



$(SN)_x$
Observations & Conclusions

Given the rather similar topological nature of the Fermiology of $(SN)_x$ and MgB_2, let's compare their E-M parameters† and computed T_c.

		T_c (°K)
SN_x:	$\Theta_D = 148$, $\mu^* = 0.05$, $\lambda = 0.3$	0.4
MgB_2:	$\Theta_D = 653$, $\mu^* = 0.10$, $\lambda = 1.0$	38

Clearly, it is Θ_D that dominates the difference in T_c between the two compounds, and not values of λ whether driven by exceptionally strong e-p modes or nesting.

Were λ to be 1.0 for $(SN)_x$, T_c ~ 11 °K (...*yawn*...) !
2.5 23 °K (...*hmm*...)

† SN_x: This talk; Harper, et al (1997)
MgB_2: An and Pickett (2001); Susner, et al (2007)

$(SN)_x$
Follow-On

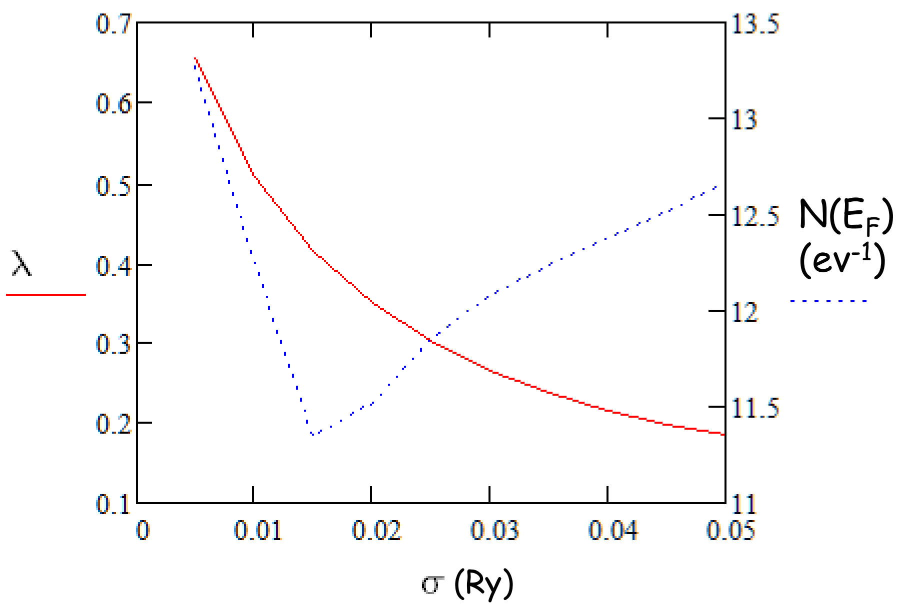
- Explore details of λ dependence on phonon IR
- Compare with MgB_2
- Then, why is T_C for MgB_2 ten times larger than $(SN)_x$?

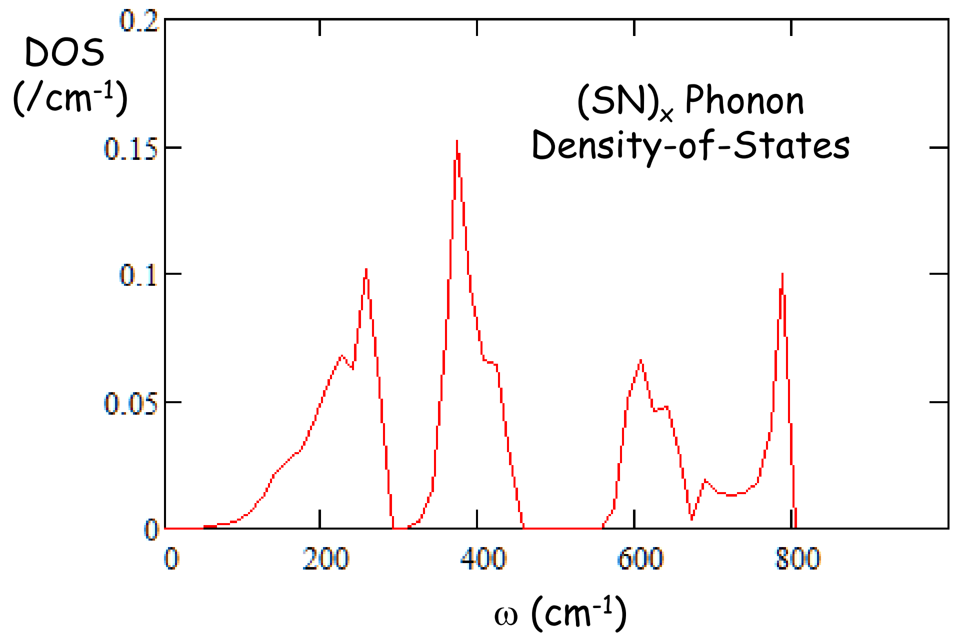
- What really is the upper limit for T_C within e-p mediated pairing?
- Could it be RT or above as speculated by Alex Mueller?

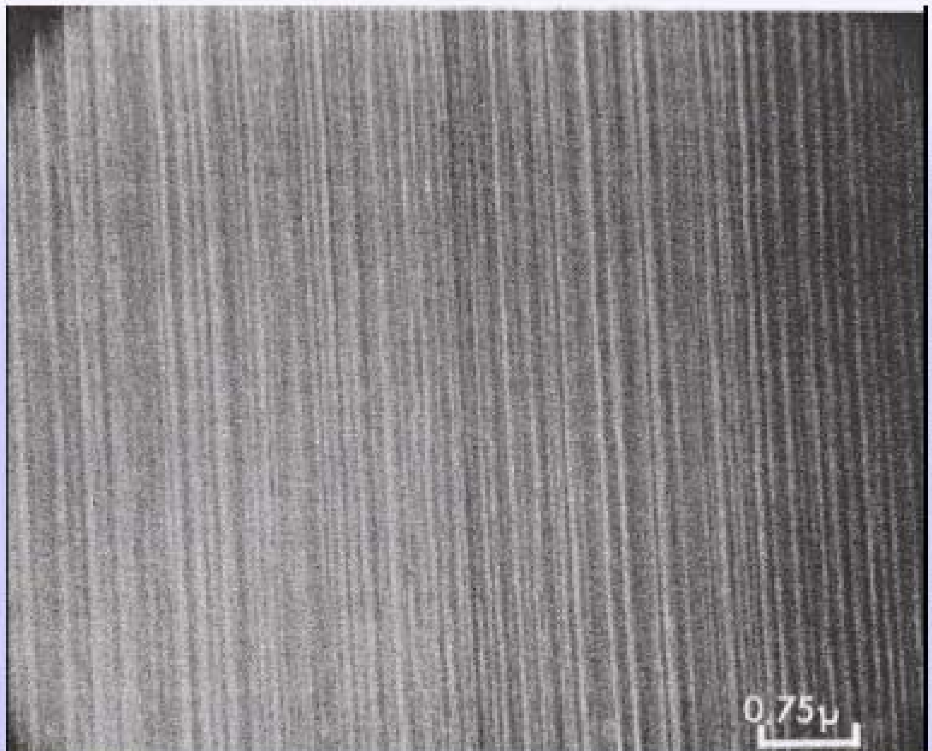
$(SN)_x$

Electron, Phonon, Electron-Phonon Properties

See Previous Slides...



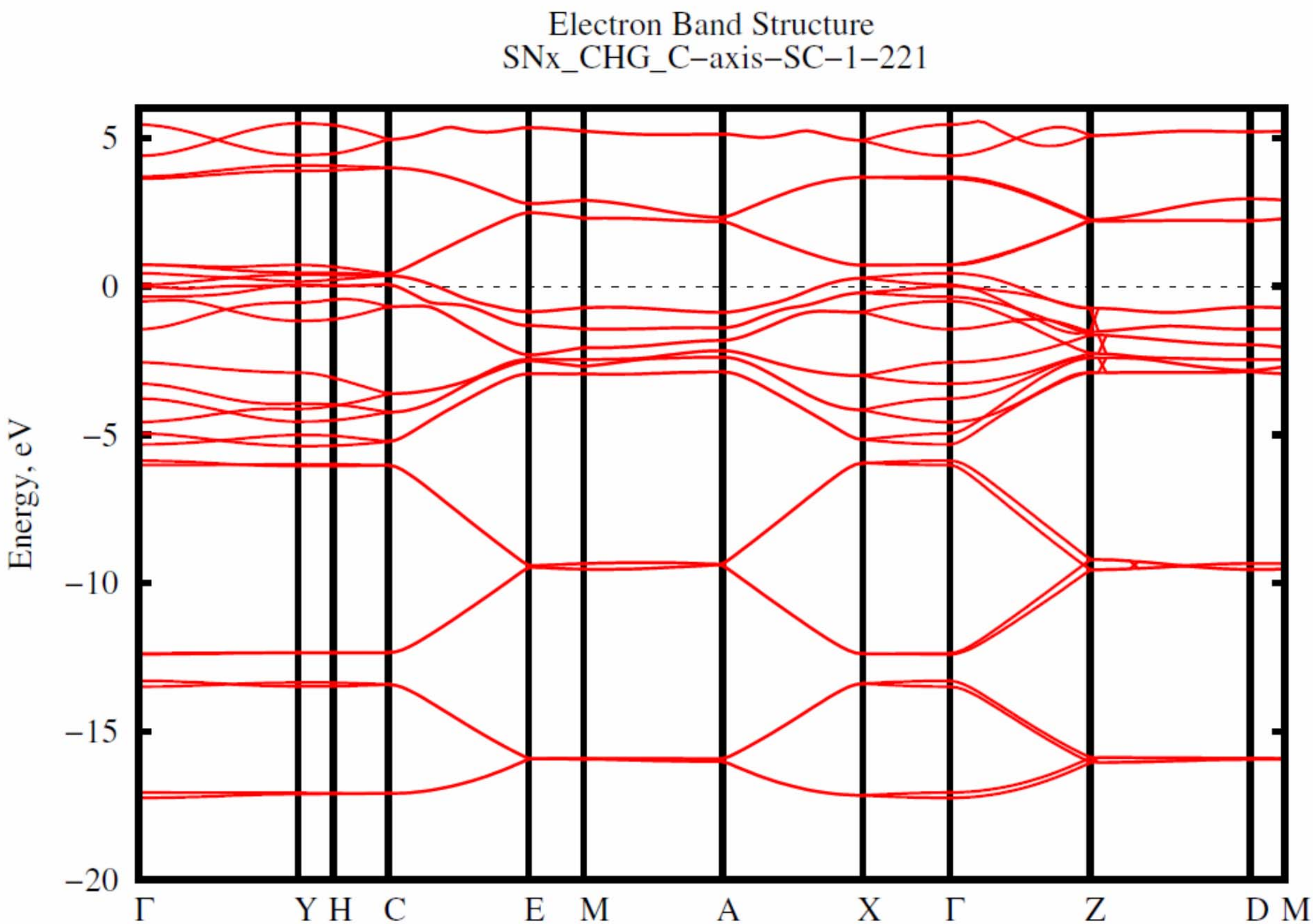




Electron micrograph of a smooth area of an SN_x Crystal

"Experimental Apparatus"

- Linux box running SUSE 10.1
- Dual Xeon (1.2 GHz) Motherboard, 2 GB RAM, 133 MHz FSB, Vintage ca. 2003
- Democritos Quantum Espresso Package
  - pw.x, 4x4x4 MP grid
  - ph.x, tr2_ph = 1e-11
  - 1 q-point => 25 hours wall time
  - Still not a fully converged calculation!


Electron Band Structure
SNx_CHG_C−axis−SC−1−221
Energy, eV
5
0
−5
−10
−15
−20
Γ
Y
H
C
E
M
A
X
Γ
Z
D
M

EMAD, Cont'd

$$T_C = \frac{\Theta_D}{1.45}\exp\left(-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right)$$

E-M

$$\langle\omega\rangle = \frac{2}{\lambda}\int \alpha^2 F(\omega)d\omega$$

A-D

$$T_C = \frac{\langle\omega\rangle}{1.20}\exp\left(-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right)$$

Need to compute $g^{\mathbf{q}\nu,mn}_{\mathbf{k}+\mathbf{q},\mathbf{k}}$!

Lattice Specific Heat of $(SN)_x$

Specific heat of polysulfur nitride, $(SN)_x$

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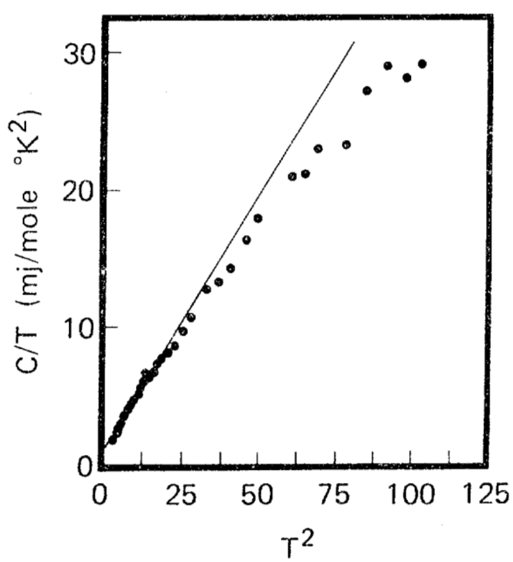

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- Θ_D ~ 103…148…170 °K ?
- λ ~ 0.31 ?
- $2\Delta/kT_C$ ~ 6.5 ?
- T_C = 330 mK
- Well ?

Coming Up!