Neutron Research at NIST

Mike and Jack symposium

Hydrogen in metals -

as a postdoc and beyond

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how it all started

I met Jack & Mik … nference

From May 1979 a … nths, co-financed
by NBS,
extended by 9 m

From October 19 … ty of Maryland

post doc time is a good time

A lot of freedom

There was much time to use and to learn about instruments

There was much advice on how to use instruments

Science:

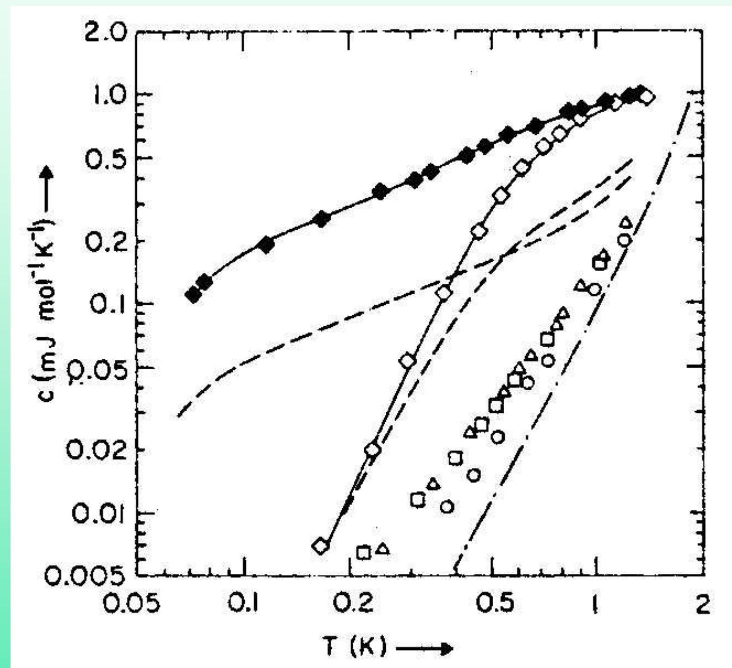
- **diffuse scattering from O defects in Nb**
- **phonons**
- **graphite intercalation compounds**
- **hydrogen tunneling**

As PhD student I did phonons, and so as post doc I should look for H tunneling in $Nb(OH)_x$ by neutron spectroscopy

H tunneling - what is the problem?

C. Morkel, H. Wipf and K. Neumaier, PRL. 40, 947 (1978)

- ○ **pure Nb**
- □ **$NbN_{0.003}$**
- △ **$NbH_{0.002}$**
- ◇ **$NbN_{0.006}H_{0.002}$**
- ◆ **$NbN_{0.003}D_{0.002}$**

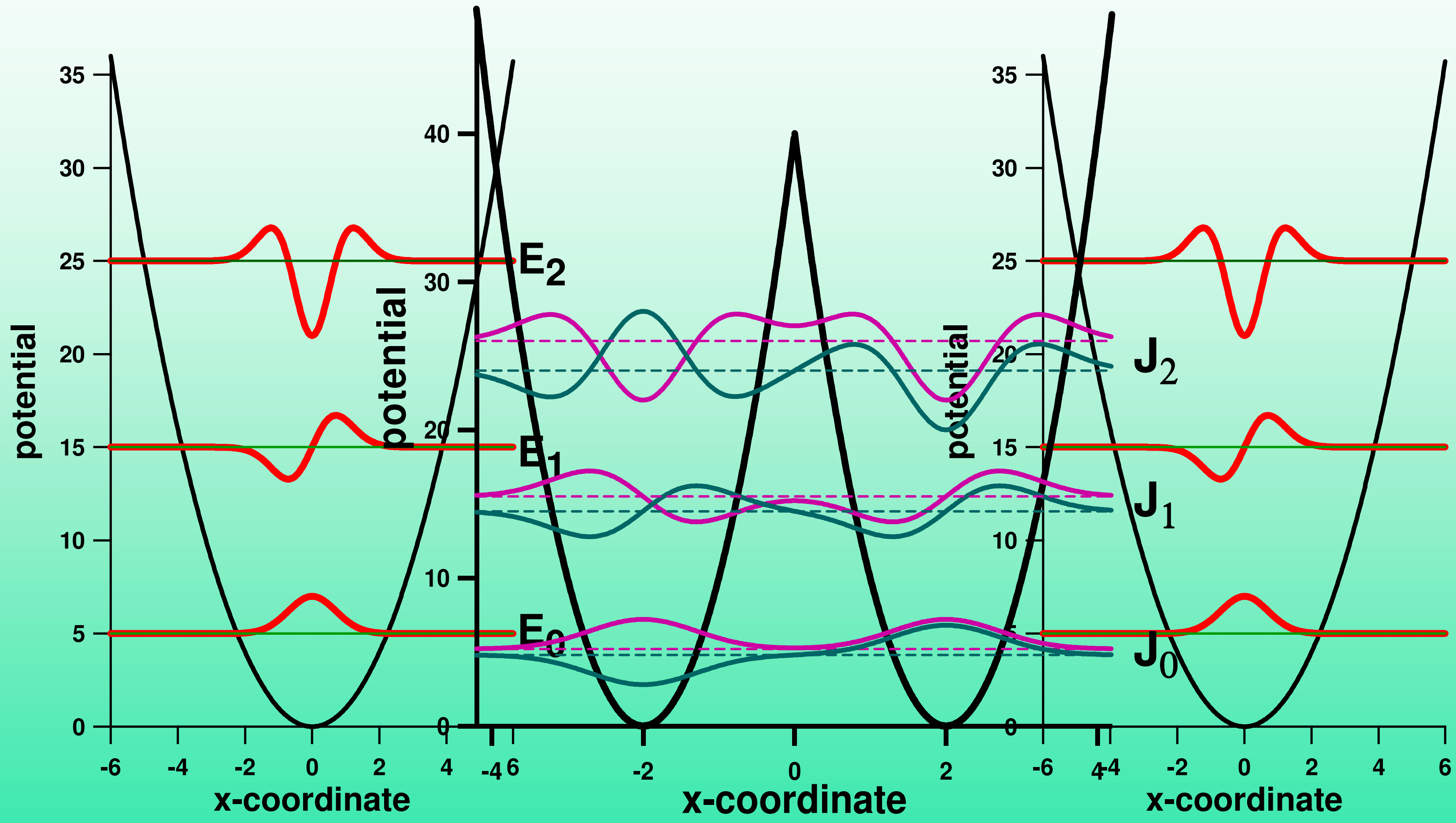


Specific heat anomaly needs the presence of both N and H / D

The anomaly is very sensitive on the hydrogen isotope

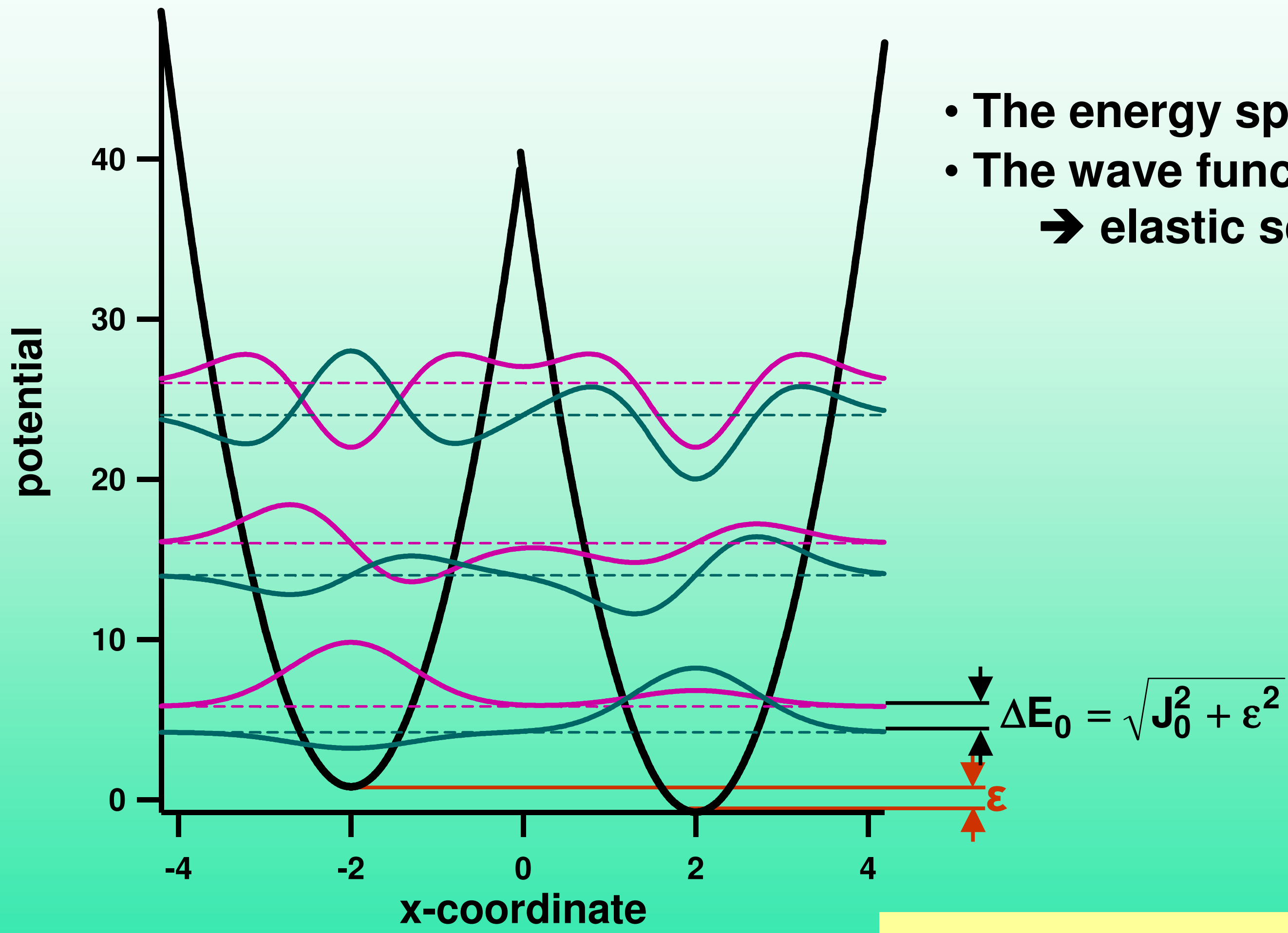
⇨ hydrogen tunneling ⇦

the H-tunneling



In a double minimum potential all oscillator levels become split

an asymmetric potential

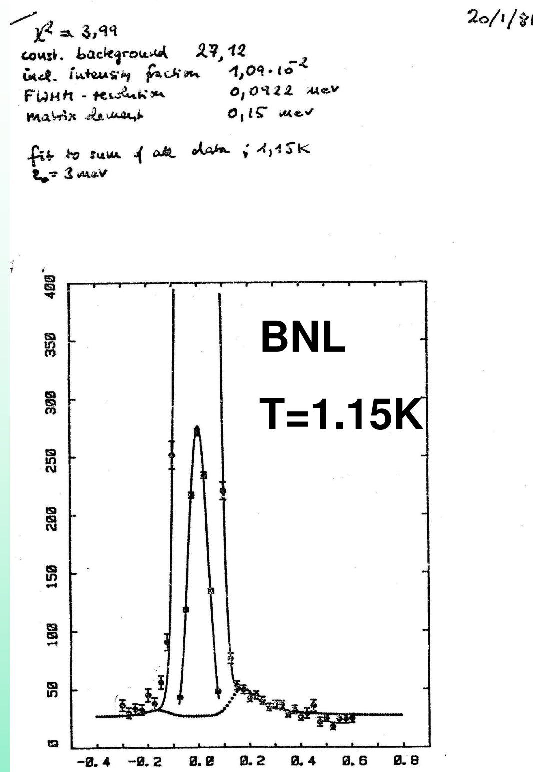


- **The energy splitting increases**
- **The wave functions localize rapidly**
  ➔ elastic scattering

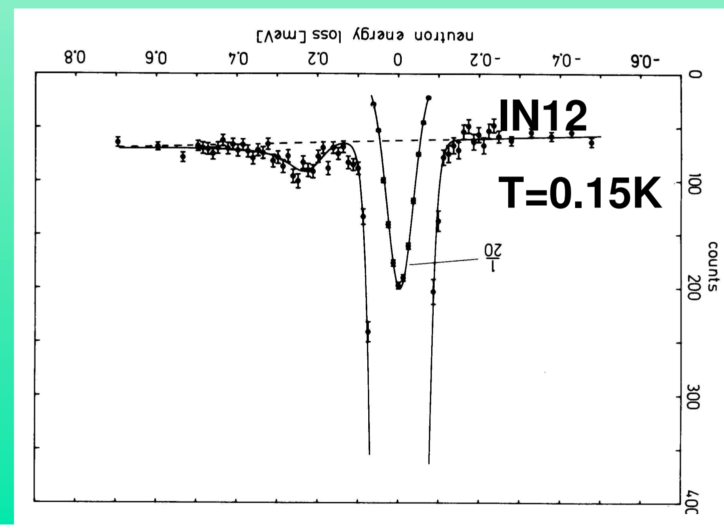
Strain leads towards a glassy system with an energy onset at J_0

the ground state

Brookhaven:
triple axis, resolution 100 μeV,
sample 90 gr of $NbO_{0.013}H_{0.016}$,
counting time ~ 2 days per spectrum



NIST:
triple axis, resolution 92 μeV,
sample 200 gr of $NbO_{0.011}H_{0.010}$,
counting time ~ 4 days per spectrum



IN12 at ILL:
triple axis, resolution 76 μeV,
sample 200 gr, $NbO_{0.002}H_{0.002}$,
counting time ~ 3 days per spectrum

the H-tunneling 1: defect concentration

<u>**IN6 at ILL:**</u>

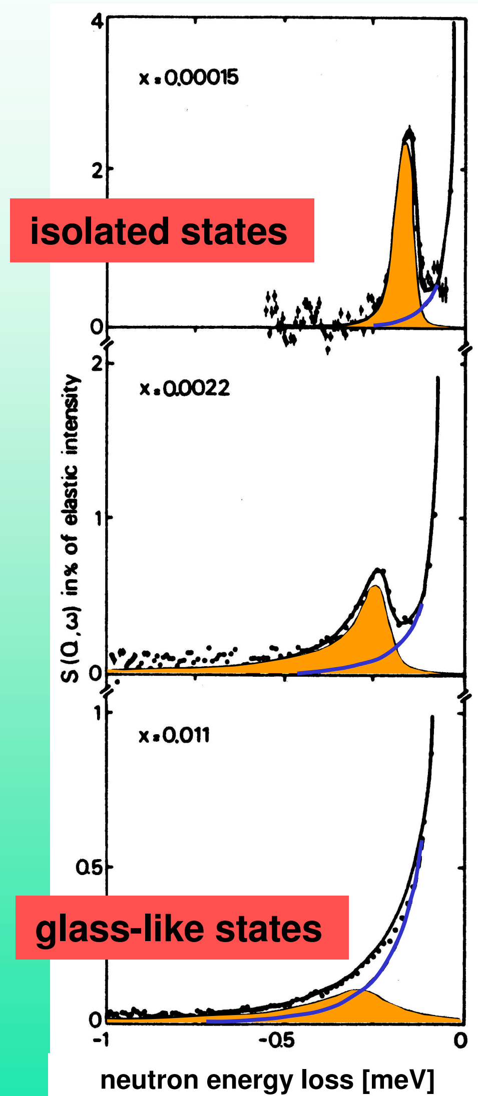
focusing time of flight, resolution 60 μeV

150 gr $NbO_{0.002}H_{0.002}$ (like IN12),

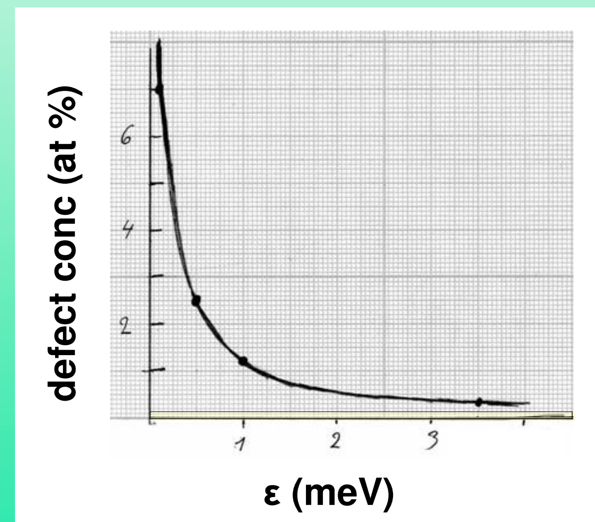
counting time ~ 6 hours

T = 0.05 K

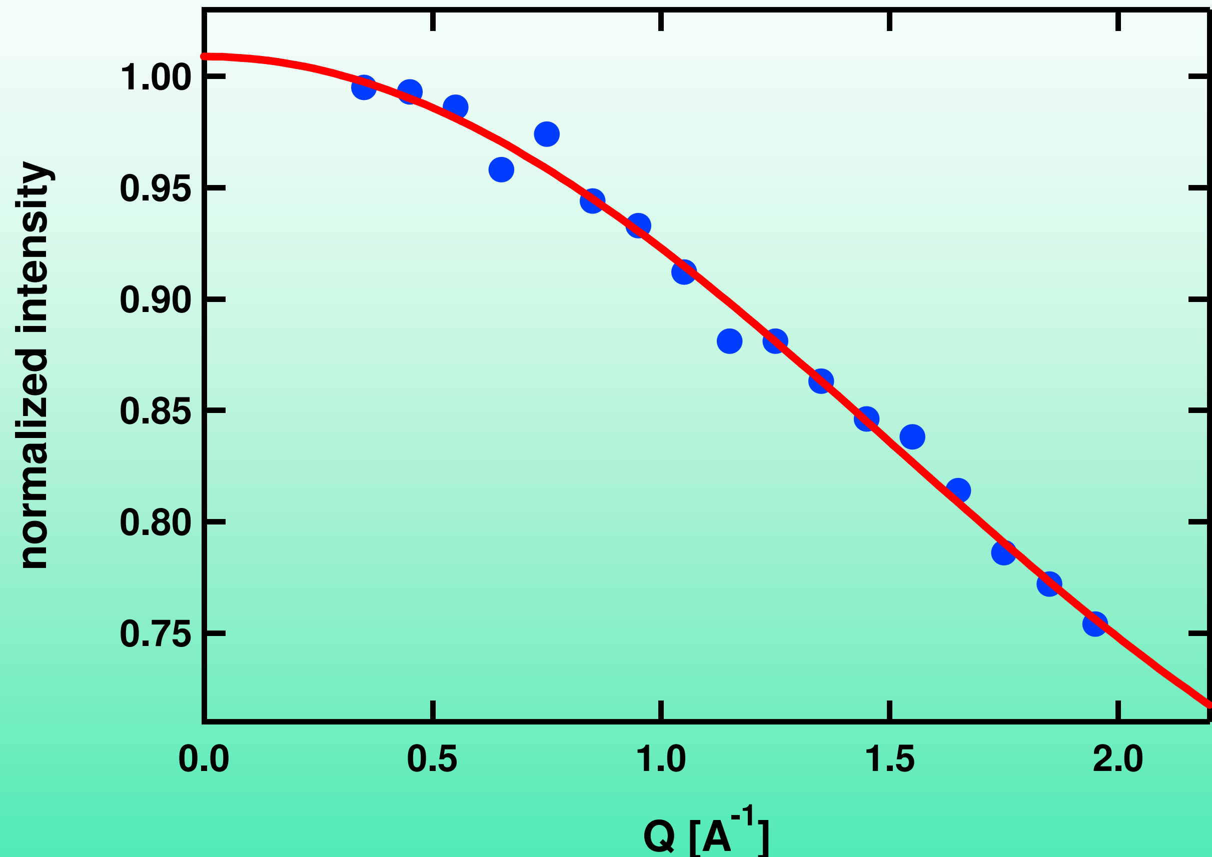
J_0 = 0.22 meV



Mutual interactions between defects cause the distortions ε, and tunneling occurs only for concentrations $x<10^{-3}$



the inelastic structure factor



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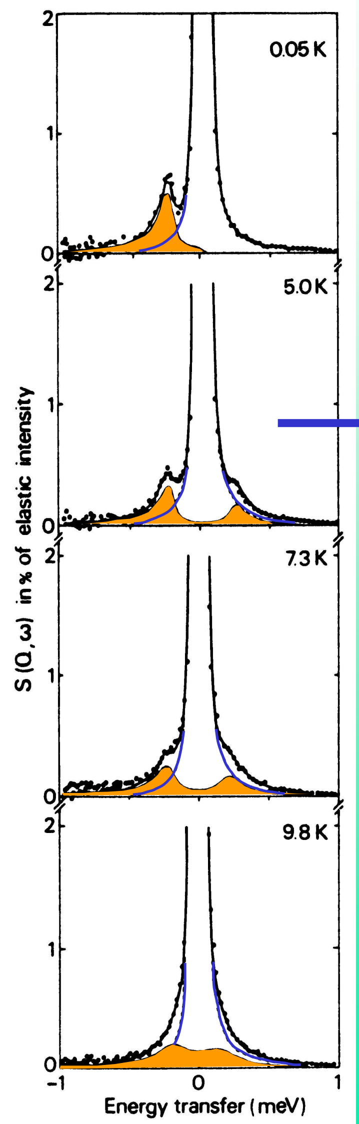
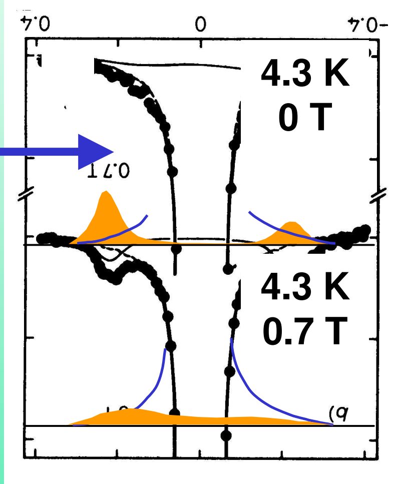
Elastic scattering 65%

Tunneling distance 1.05 Å < a < 1.35 Å (ideal nn tetrahedral distance = 1.17 Å)

the H-tunneling 2: temperature



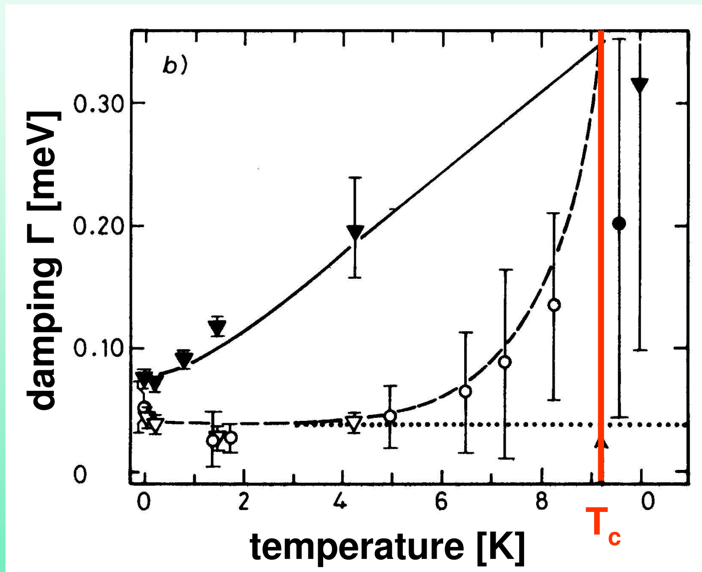
Temperature dependence
150 gr, $Nb(OH)_{0.002}$, counting time 6 hours



Magnetic field dependence
50 gr $Nb(OH)_{0.0002}$, counting time 12 hours

The coupling to the conduction electrons destroyes the coherent tunneling state

electron defect coupling



for metallic state

J. L. Black 1981

$$\Gamma = \frac{\pi}{4}\lambda^2 E \coth\left(\frac{E}{k_B T}\right)$$

Γ = 0.043 meV for T=0 K

for BCS superconductor

$$\Gamma = \pi \lambda^2 \frac{k_B T}{\exp\left(\Delta^{gap}(T)/k_B T + 1\right)}$$

Γ = 0 for T=0 K

$\Delta^{gap}(T)$ = gap energy with $\Delta(T)$ = 1.53 meV for Nb

λ = interaction strength $\rho(\varepsilon_F)V$

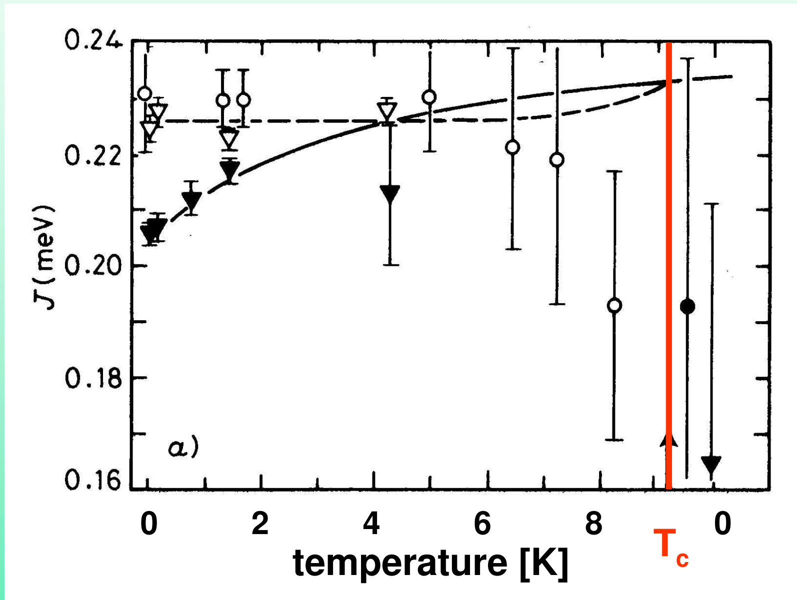
fit: λ = 0.45

metallic Nb: λ = 0.55

Coherent tunneling regime below ~ 10K

renormalisation of the splitting

Kondo, Yamada, Black, Grabert, Teichler



Open symbols sc state

Filled symbols nc state

$$\ln\left(\Delta E^{n} \big/ \Delta E^{s}\right) = \frac{\lambda^2}{4}\left(\ln\frac{\Delta E^{n}}{\Delta E^{s} + 2\Delta^{gap}} + I\left(\frac{\Delta E^{s}}{2\Delta^{gap}}\right)\right)$$

with $\Delta E^{n} = 0.224$ meV and $\Delta E^{s} = 0.206$ meV

local modes

The local modes energies in the free and trapped states are the same

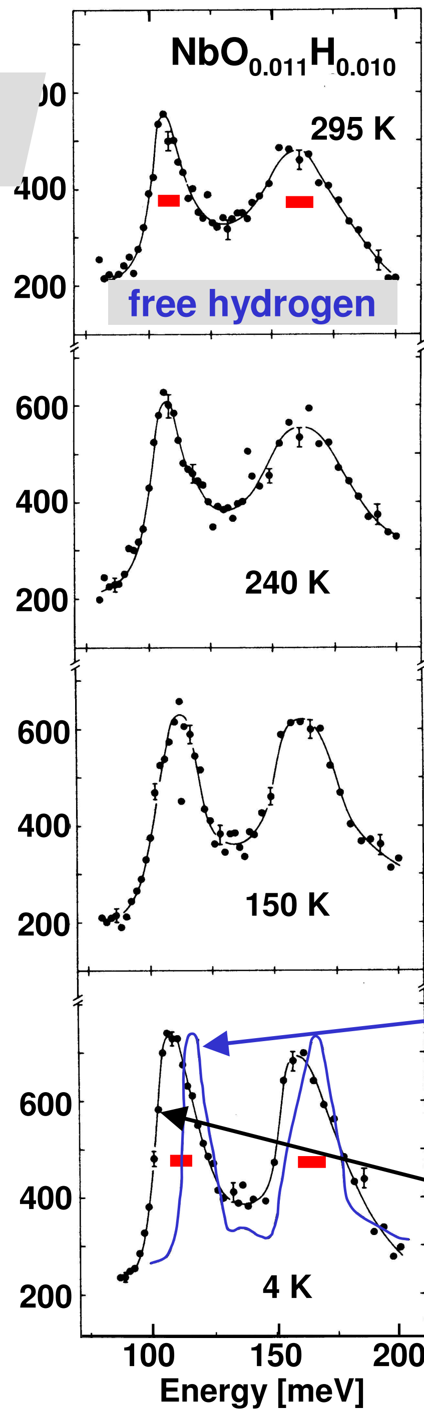
H always occupies tetrahedral sites

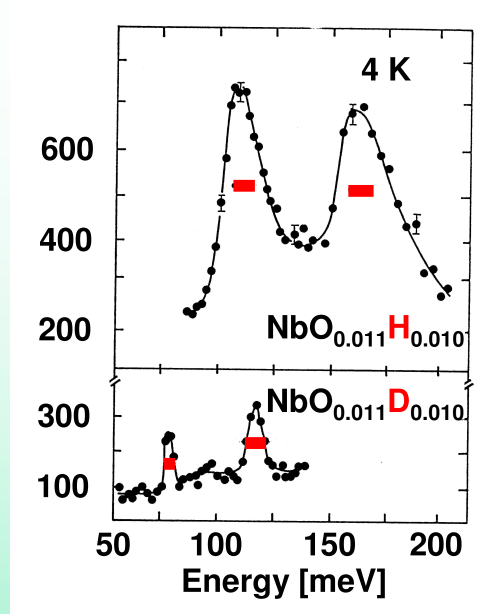
The line profiles in α-phase NbH_x and in $Nb(OH)_x$ have large widths

The line widths in the ordered hydride phase are smaller than in the disordered phase

The line widths depend strongly on the H-isotope

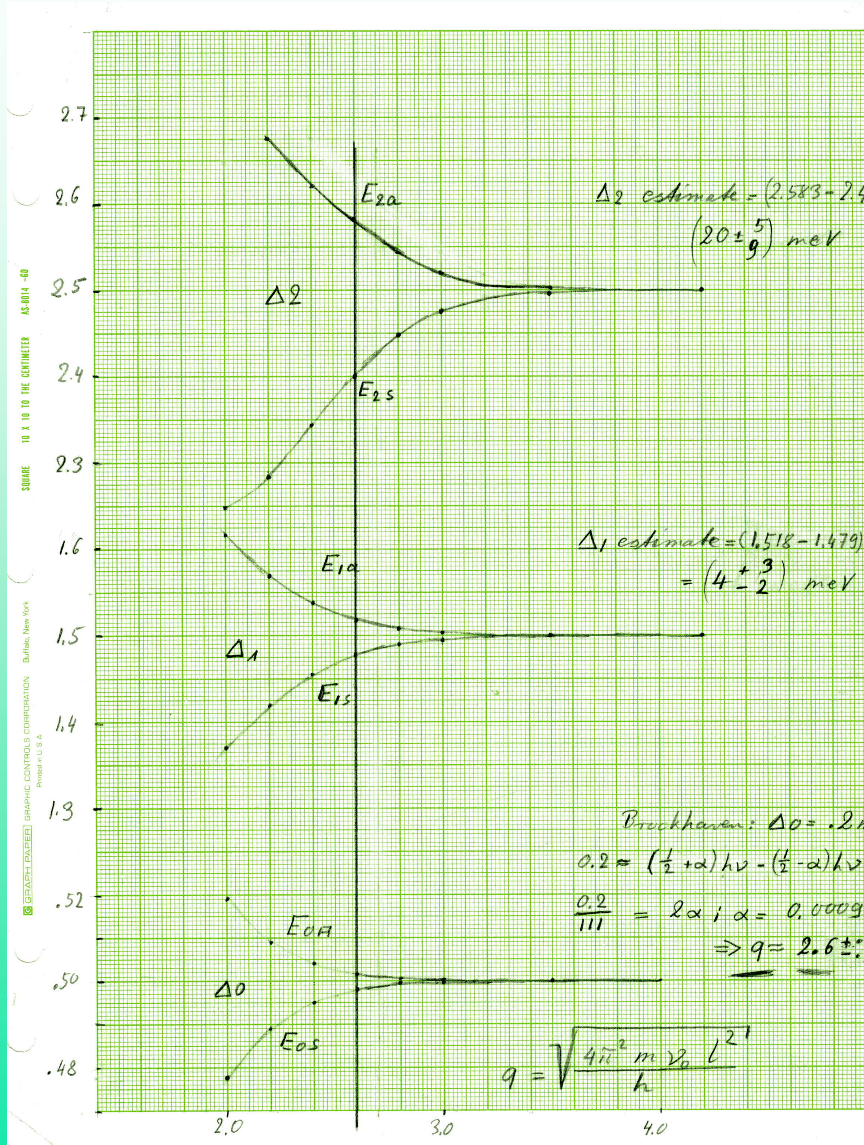
Is this due to the tunnel splitting J_1?

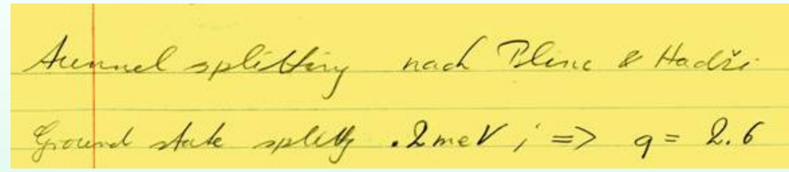




a strong isotope effect

an excited state splitting?





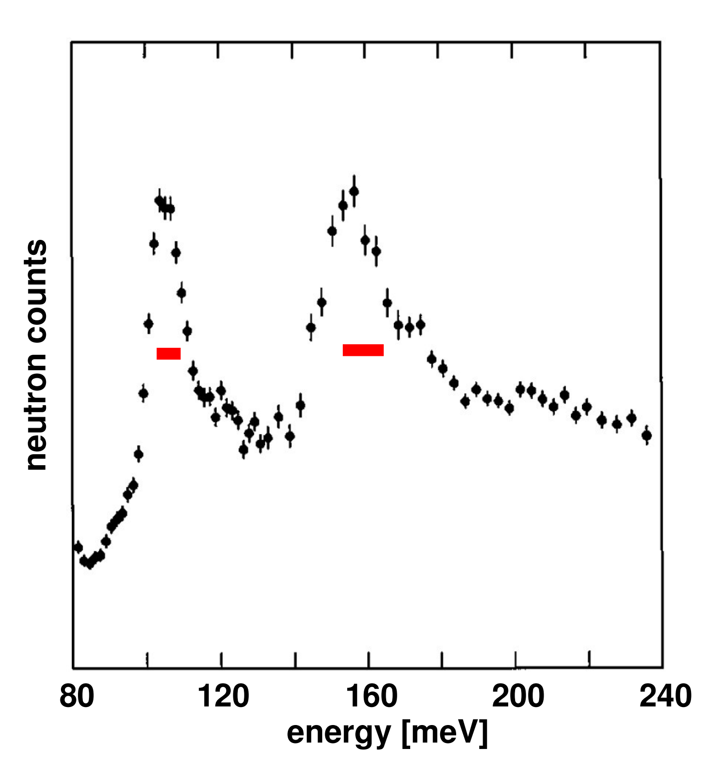
excited state splitting

90 g of $NbN_{0.004}H_{0.003}$ @ 4.2 K

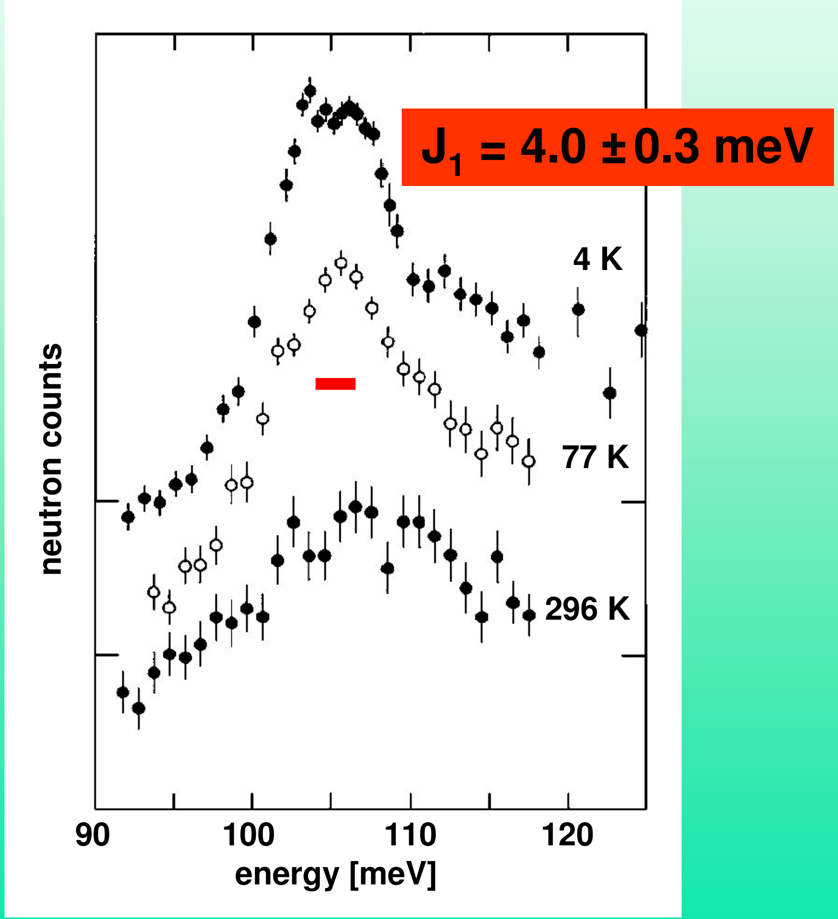
trash

a new high resolution spectrometer

Standard setup:
Cu220 with 40'/40' collimation and Be filter analyser:
6.3 meV FWHM at E=105 meV



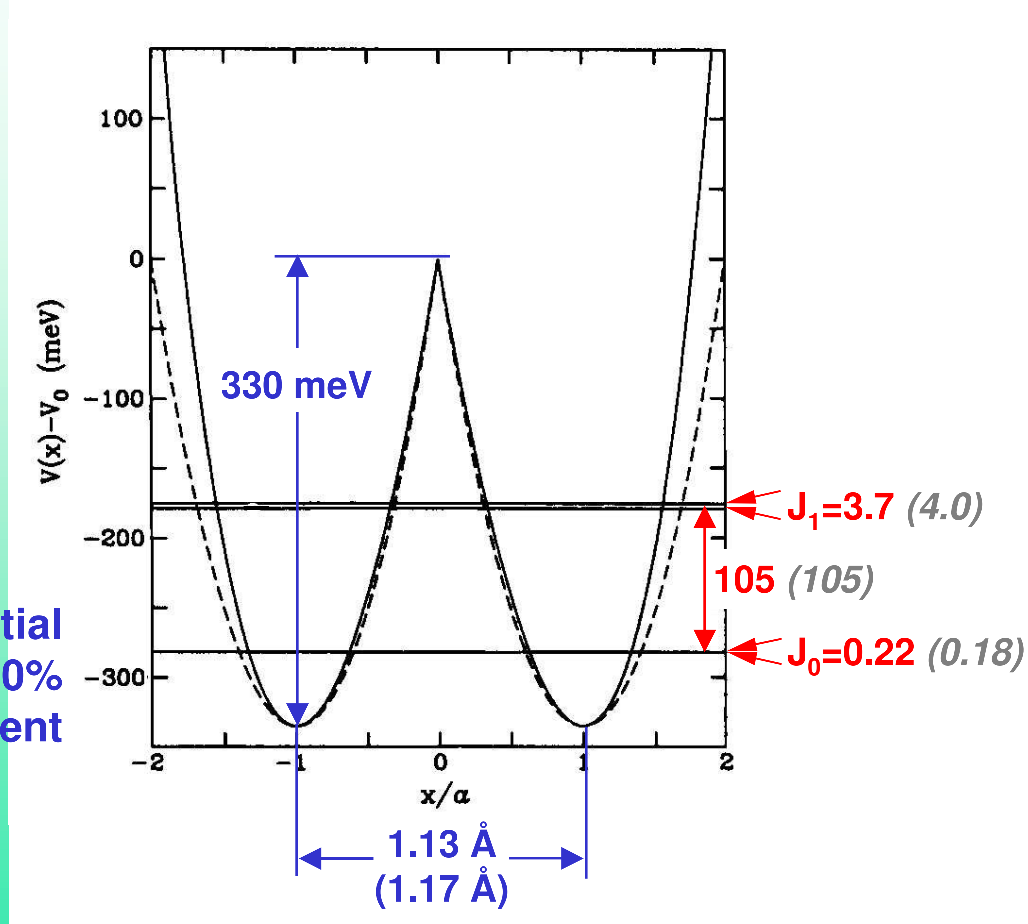
Cu 220 with 20'/20' collimation and a composite Be-graphite-Be filter analyser:
2.6 meV FWHM at E=105 meV



excited state splitting

Model needs to account simultaneously for the vibrational level and for the splittings J_0 and J_1

A harmonic double well potential (dashed line) hardened by a 10% admixture of a quartic component

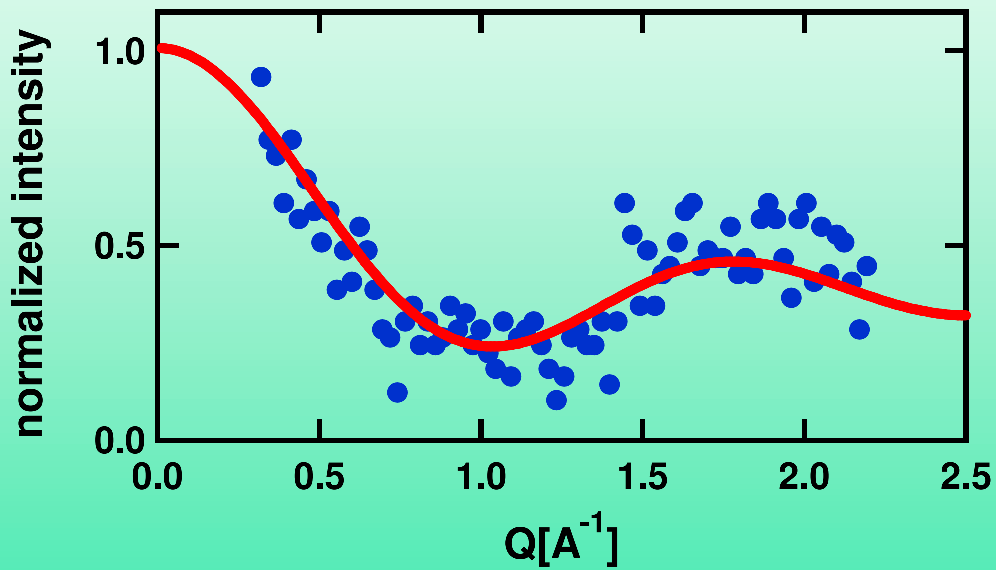


J. J. Rush et al., Euro Phys. Lett, 1999

What about the 25 meV broad excitation at 160 meV, ?FANS?

the O-H geometry

Diffuse scattering in $Nb(OD)_{0.002}$ at 100 K



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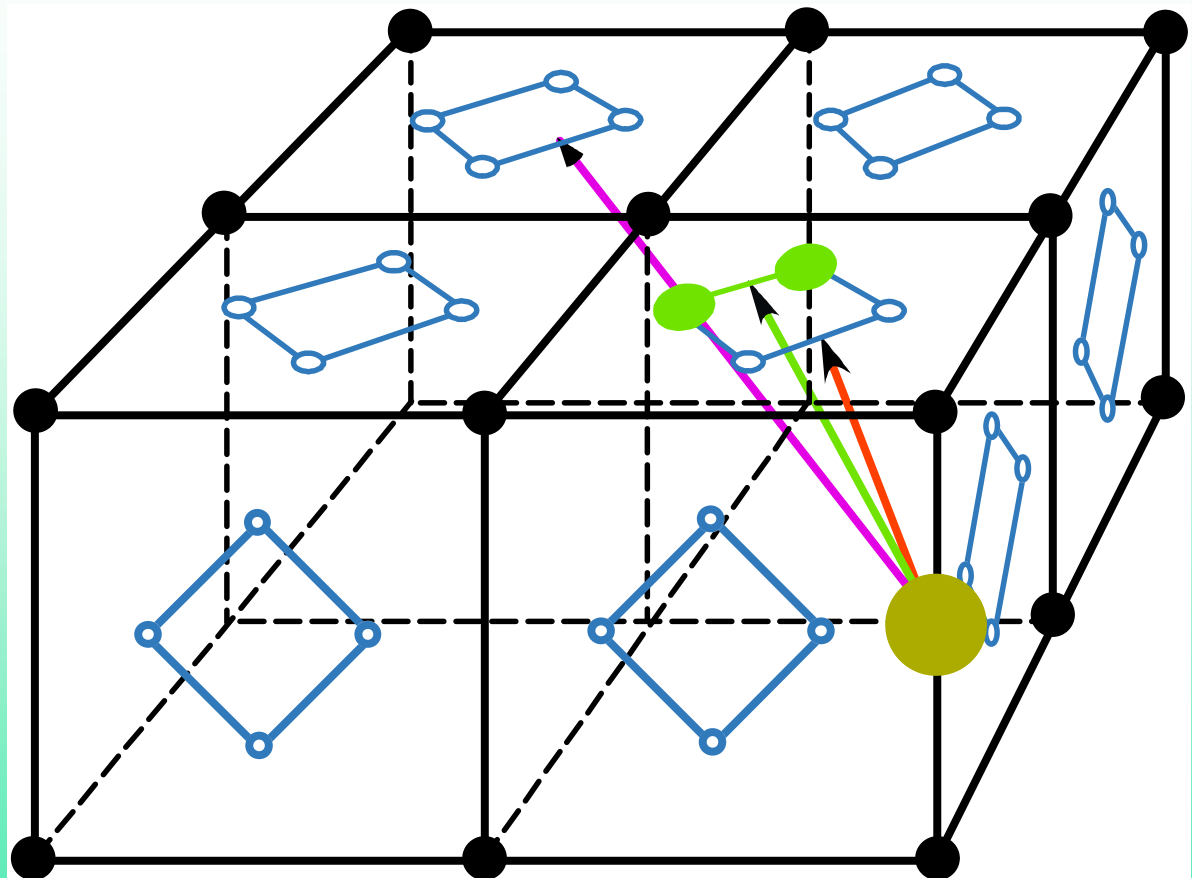
The trapping distance is large ~ 4.0 +/- 0.5 Å

the geometry



the 1st nearest symmetrical double minimimum is at 2.5 Å

the 2nd nearest symmetrical double minimimum is at 3.4 Å

the 3rd nearest symmetrical double minimimum is at 6.1 Å

summary

•A comprehensive understanding of H tunneling in $Nb(OH)_x$

•Ground state and the excited state level splitting

•Potential (1D double minimum) decribing consistently the excitations

•Coherent ground state tunneling below ~ 10K

•Electrons are the dominating damping mechanism

•Temperature dependence of the line width both in nc and sc state, the finite Γ at T=0 in the metallic state, the renormalisation of J_0 are consistently explained with J_0= 0.224 meV and λ=0.45

•The strain introduces a glass-like state

unique features?

substitutional traps $Nb(TiH)_x$ with a more complex trap geometry and thus a more complex level scheme

rare earth-hydrogen systems YH_x, ScH_x where the hexagonal structure itself provides double minima potentials

but the interstitial traps $Nb(OH)_x$ are the most completely studied system of translational atomic tunneling

what remains?

list of publications

7. Neutron diffuse-scattering intensities in niobium, J.M. Rowe and A. Magerl,Phys. Rev. B 21, 1706 (1980)

9. Observations of low-energy excitations in NbD: A simple lattice-dynamical model, A. Magerl, J.M. Rowe and D. Richter, Phys. Rev. B23, 1605 (1981)

12. Tritium vibrations in niobium by neutron spectroscopy, J.J. Rush, A. Magerl, J.M. Rowe, J.M. Harris and J.L. Provo, Phys. Rev. B 24, 4903 (1981)

19. Local hydrogen vibrations in Nb in the presence of interstitial (N,O) and substitutional (V) impurities, A. Magerl, J.J. Rush, J.M. Rowe, D. Richter and H. Wipf, Phys. Rev. B 27, 927 (1983)

20. Phonons in LiC6, H. Zabel, A. Magerl and J.J. Rush, Phys. Rev. B 27, Rapid Comm. 3930 (1983)

21. Diffusive motion of alkali atoms in graphite: a quasi-elastic neutron scattering study, H. Zabel, A. Magerl, J.J. Rush and A.J. Dianoux, Proceedings of the Symposium on Intercalated Graphite, Boston, USA, 1982, Materials Research Symposia Proceedings, Vol. 20, ed. M.S. Dresselhaus, G. Dresselhaus, J.E. Fischer and M.J. Moran, North Holland, New York, Amsterdam, Oxford 1983

22. Planar diffusive motion of alkali-metal intercalant atoms in graphite, H. Zabel, A. Magerl, A.J. Dianoux and J.J. Rush, Phys. Rev. Lett. 50, 2094 (1983)

23. Influence of substitutional and interstitial impurities on local hydrogen modes in Nb, A. Magerl, J.J. Rush, J.M. Rowe, D. Richter and H. Wipf in the "Proceedings of the International Symposium on the Electronic Structure and Properties of Hydrogen in Metals" edited by P. Jena and C.B. Satterthwaite,Plenum Press New York, 1983

24. Dynamical properties of alkali intercalates in graphite, A. Magerl, H. Zabel, J.J. Rush and A.J. Dianoux, Synthetic Metals 7, 227 (1983)

25. Phonons in LiC6 and in heavy alkali metal-graphite intercalation compounds, A. Magerl, H. Zabel and J.J. Rush, Synthetic Metals 7, 339 (1983)

26. Elastic neutron scattering results on C24Rb(D2)x in the dilute concentration regime, H. Zabel, J.J. Rush and A. Magerl, Synthetic Metals 7, 251 (1983)

38. Local modes in dilute metal hydrogen alloys, A. Magerl, J.J. Rush and J.M. Rowe, Phys. Rev. B 33, 2093 (1986)

63. Diffusion and melting in two dimensions: a quasielastic neutron scattering study of alkali metals in graphite, H. Zabel, A. Magerl, J.J. Rush and M.E. Misenheimer, Physical Review B 40, 7616 (1989)

70. Rapid low-temperature hopping of hydrogen in a pure metal: the ScHx system, I.S. Anderson, N.F. Berk, J.J. Rush, T.J. Udovic, R.G. Barnes, A. Magerl and D. Richter, Phys. Rev. Letters, 65, 1439 (1990)

104. Excited-state vibrational tunnel splitting of hydrogen trapped by nitrogen in niobium, J. J. Rush, T. J. Udovic, N. F. Berk, D. Richter and A. Magerl, Euro. Phys. Letters, 48, 187 (1999)

...and beyond?

Forged my thinking about physics

Forged my thinking about other disciplines

A post doc time is an extremely fruitful period

...and afterwards?

Inauguration of new building

July 19, 2005

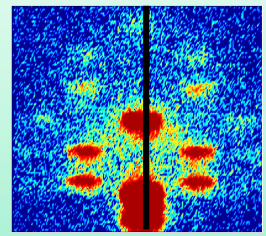
…and science?

<u>Neutron scattering</u>

Oxygen diffusion in perovskite derived structures

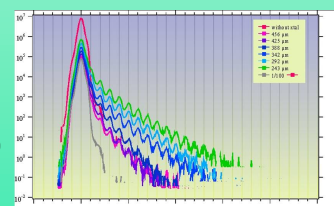
Lubricants under shear

Surface induced ordering in liquids



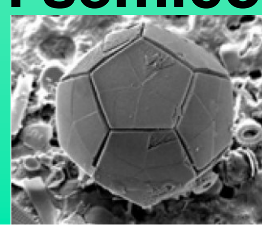
<u>X-ray scattering</u>

Time resolved diffraction (10 ps)



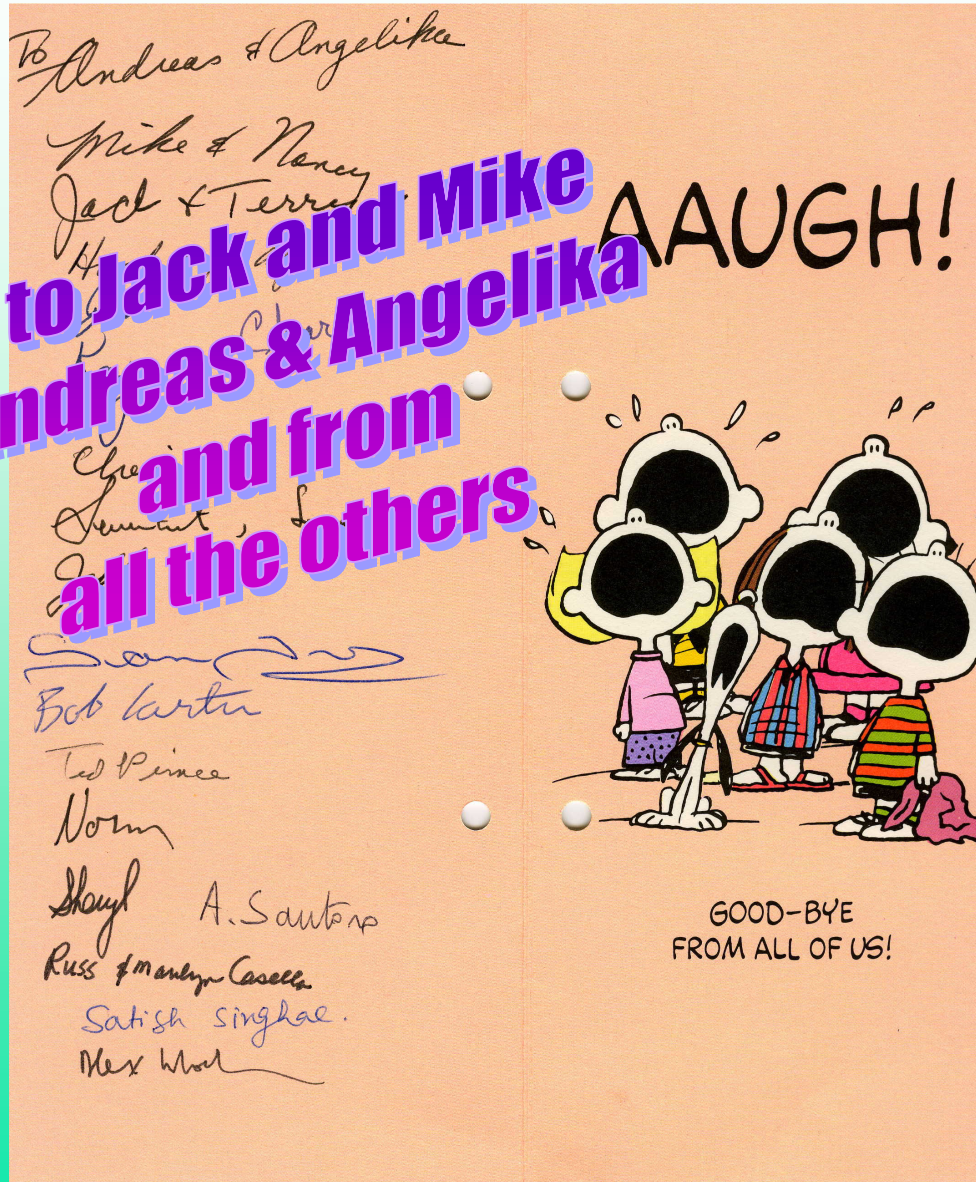
Defect structures in semiconductors (Si, SiC, Si-Ge)

Bio-mineralization



travel back home, Dec. 20, 1980

Sept. 9, 2005



to Jack and Mike
Andreas & Angelika
and from
all the others