Atomic vibrations in glasses:
50 years ago,
today,
and in future

A. Chumakov, A. Monaco, G. Monaco, I. Sergueev, W. Chrichton, O. Leupold, R. Rüffer
ESRF
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Outlook

Aus dem Institut für Physik im Max-Planck-Institut für medizinische Forschung, Heidelberg

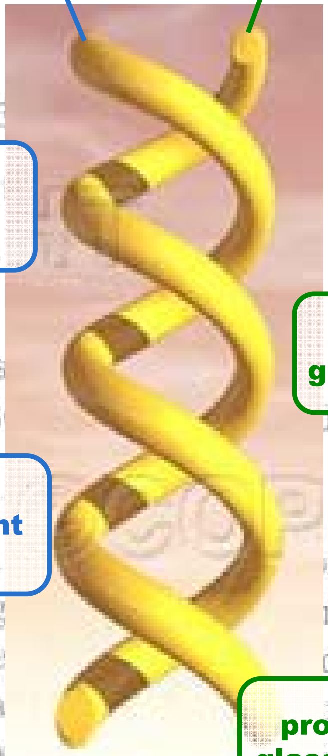
Kernresonanzfluoreszenz von Gammastrahlung in Ir^{191}

Von Rudolf L. Mössbauer

In future

Today

50 years ago



Mössbauer effect

Glass dynamics

new view of nuclear resonant scattering

new view of glass dynamics

help from nuclear resonant scattering

problems in glass dynamics

Introduction

Aus dem Institut für Physik im Max-Planck-Institut für medizinische Forschung, Heidelberg

Mössbauer effect

Glass dynamics

Kernresonanzfluoreszenz von Gammastrahlung in Ir^{191}

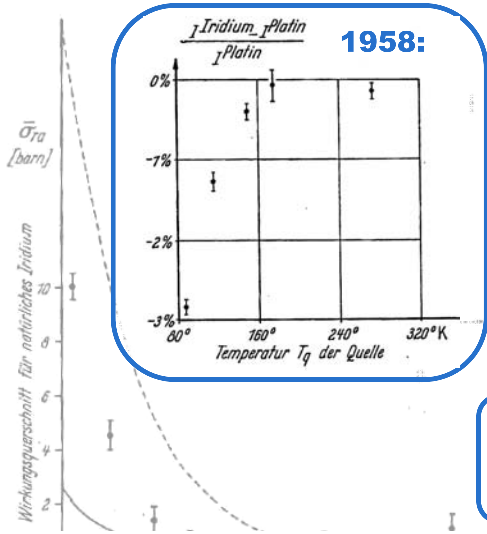
Von

RUDOLF L. MÖSSBAUER

> 50 years ago ...

RUDOLF L. MÖSSBAUER:

jeder der vier Intensitäten $I_t^{Ir}(T_1)$, $I_t^{Pt}(T_1)$, $I_t^{Ir}(T_2)$, $I_t^{Pt}(T_2)$. Bei der *einzelnen* Intensitätsmessung betrug der statistische Fehler 0,04 bis 0,05 % und die Meßzeit 12 bis 20 min. Täglich wurde eine Meßreihe aufgenommen. Um systematische Fehler auszuschließen, wurde die Geometrie bei jeder Meßreihe etwas variiert, durch Drehung der Absorber

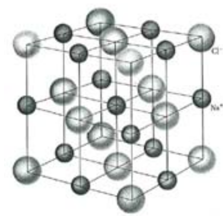


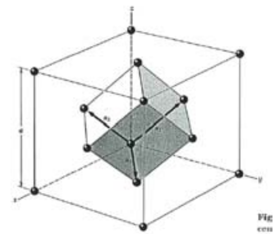
before 1958: nuclear absorption increases with T° ?

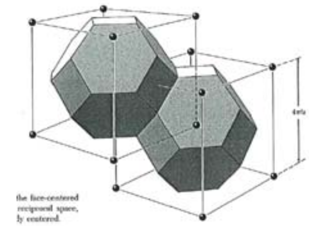
before 1971: glass dynamics: sound waves ..?

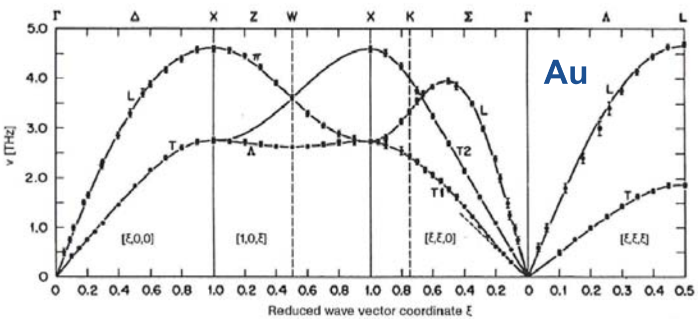
before 1971:

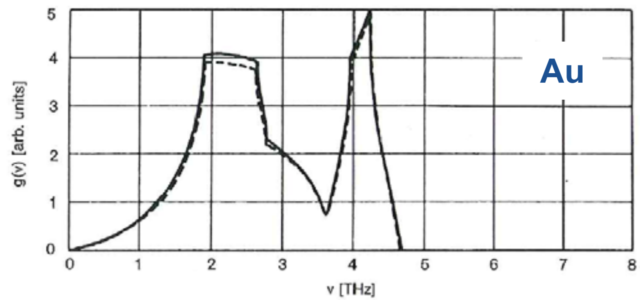
Crystals: translational periodicity



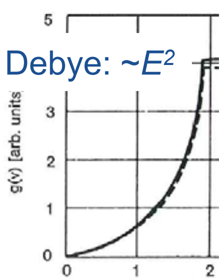








Glasses: no periodicity, no zone boundaries: ideal elastic medium



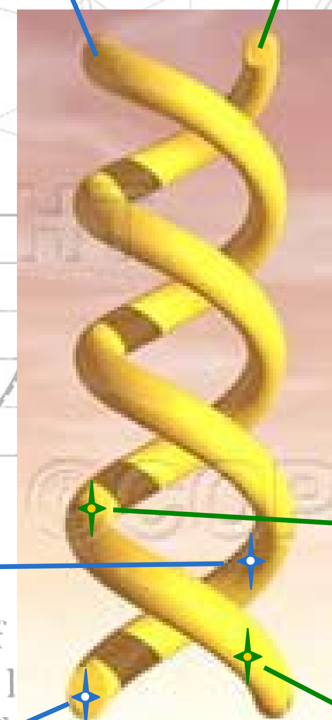
The low temperature heat capacity of pure dielectric crystalline solids is known (Chapter 5) to follow the Debye T^3 law, precisely as expected from the excitation of long wavelength phonons. The same behavior was expected in glasses and other amorphous solids—the point was so obvious that it did not encourage experimental investigation.

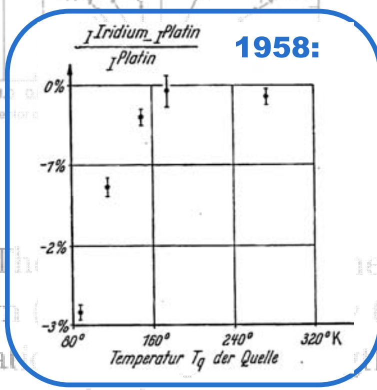
C. Kittel

1971:
crash of blissful ignorance:

Mössbauer effect

Glass dynamics





1971: thermodynamic anomalies !!!

before 1958: nuclear absorption increases with T° ?

before 1971: glass dynamics: sound waves ..?

Introduction

specific heat: additional vibrational states?

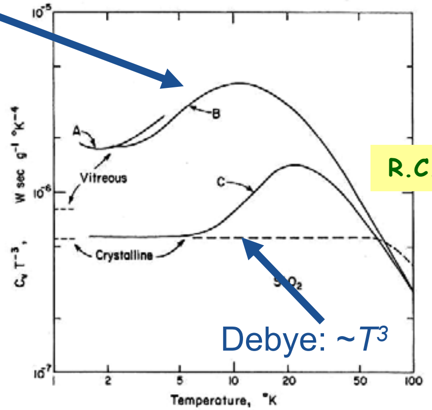


FIG. 4. Specific heat of vitreous SiO_2 and crystal quartz, plotted as $C_v T^{-3}$ vs T. *A*: I. R. Vitreosil (Ref. 29); *B*: vitreous silica (after Refs. 30–32); *C*: α-quartz

thermal conductivity: additional source of scattering ?

R.C.Zeller et al., PRB 4,2029,1971

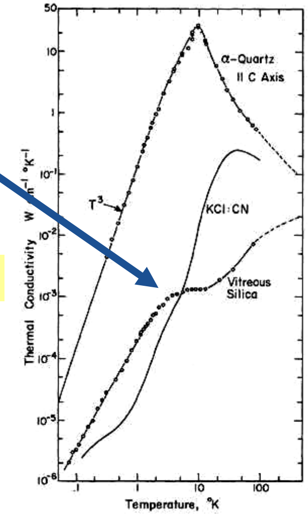
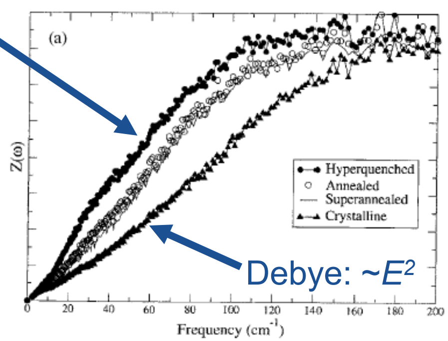


FIG. 1. Thermal conductivity of crystalline and of vitreous SiO_2 and of crystalline KCl : CN ($n_{CN^-} = 4.9 \times 10^{19}$

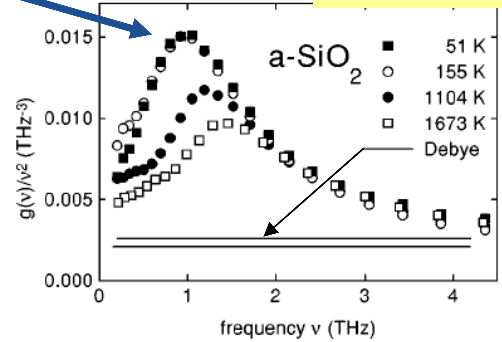
DOS *g(E)*: additional vibrational states !



C.A.Angel et al., J.Phys.:Cond.Matt. 15,S1051,2003

Reduced DOS *g(E)/E²*: the boson peak !

A.Wischnewski et al., PRB 57,2663,1998



Introduction

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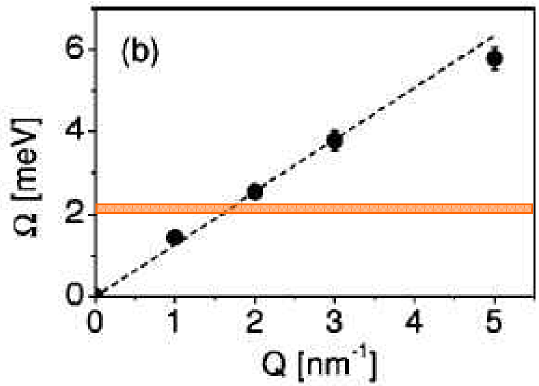
Vibrations and relaxations in a soft sphere **glass**: **boson peak** and structure factors. Authors: , Schober, H. R.. Affiliation: ...
adsabs.harvard.edu/abs/2005cond.mat..2611S - Similar pages

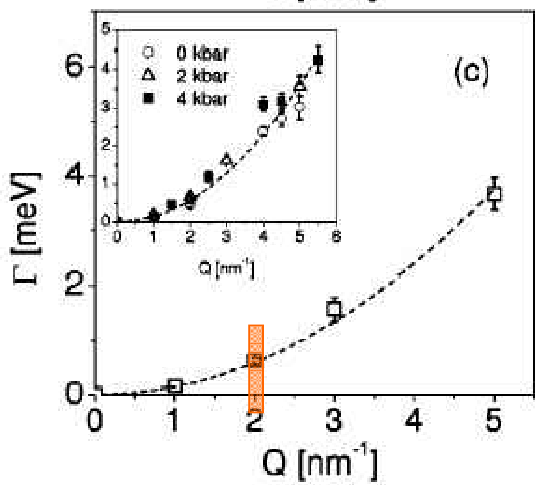
Origin of the **boson peak** in a network **glass** B2O3
Origin of the **boson peak** in a network **glass** B2O3. Authors: , Engberg, D.; Wischnewski, A.; Buchenau, U.; Börjesson, L.; Dianoux, A. J.; Sokolov, A. P.;

Introduction

Introduction

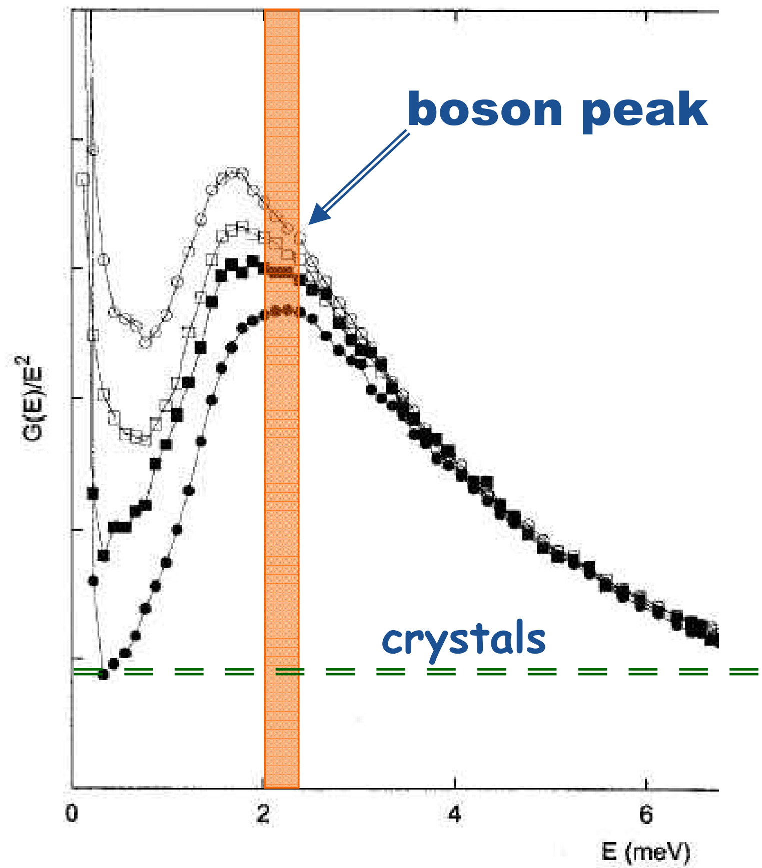
dispersion relations





A.Mermet *et al.*, PRE 66, 31510 (2002)

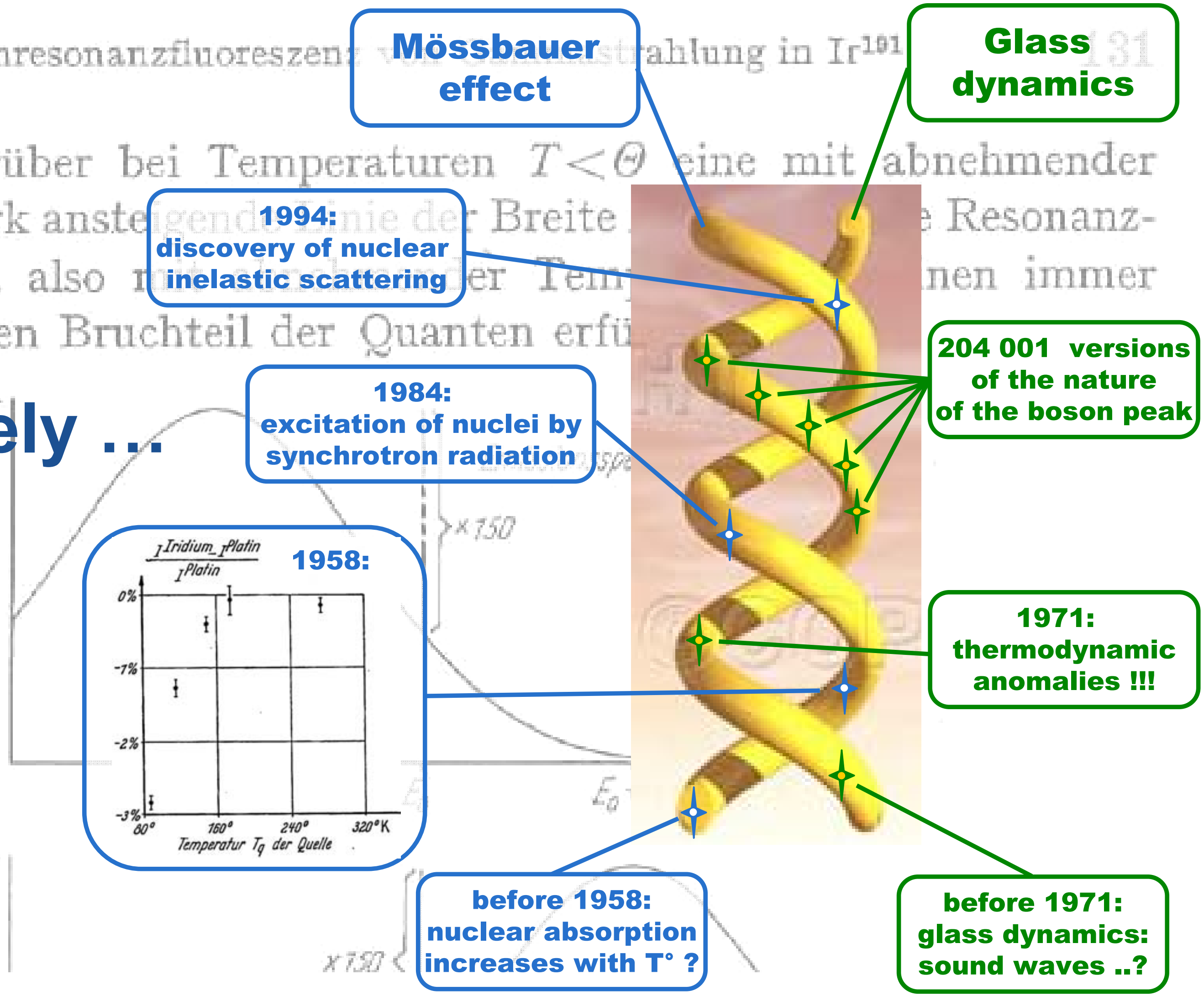
density of states



L.Saviot *et al.*, Phil.Mag.B 82, 533 (2002)

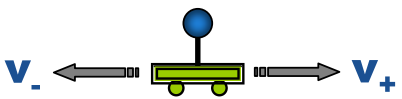
Introduction

Fortunately …



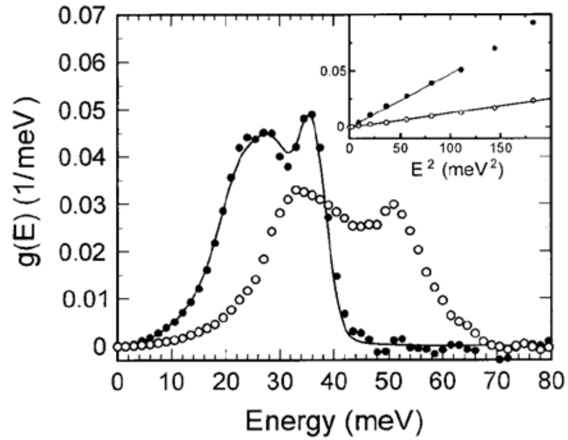
Experimental

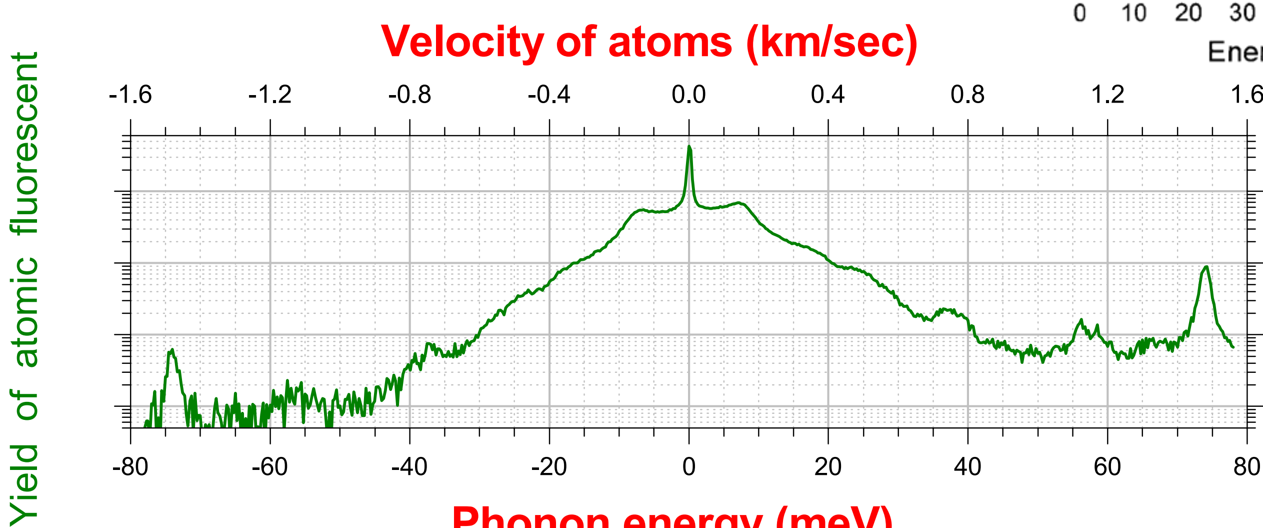
Nuclear Inelastic Scattering: resonant absorption of x rays by moving nuclei:



^{57}Fe nuclei

DOS in absolute and correct scale:





Experimental

Raman and neutron scattering

you can do almost everything,
but not always convenient
and not necessarily precise

Nuclear inelastic scattering

you can only cut,
but precisely, sharp, deep,
and exactly where you need it

132 RUDOLF L. MÖSSBAUER:

4. Versuchsanordnung

Fig. 2 zeigt die Versuchsanordnung, Fig. 3 den
Kryostaten. Die Absorber, zwei je etwa 0,4 mm d
bzw. Platinbleche von 35 mm Durchmesser waren
der Abkühlung eine ungehinderte Kontraktion d
Untersucht wurde die Absorption der beim I
ausgesandten 129 keV Gammastrahlung in Iridi
Zerfallschema [7] und das beobachtete Spektrum,
der 95 d-Aktivität von Os^{185}
enthält [8]. Die harten, beim K-Einfang von Os^{185} ausgesandten Linien
von Re^{185} bei 640 keV und bei 875 keV durchsetzten die Absorber

(i) cooling rate

and

(ii) density

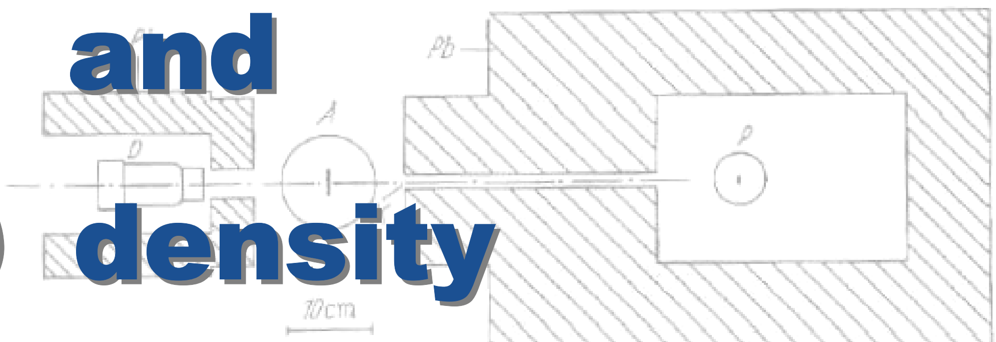


Fig. 2. Versuchsgeometrie. A Absorber-Kryostat; P Kryostat mit Quelle; D Detektor: NaJ(Tl)-Kristall (22 mm hoch, 40 mm Durchmesser) und Photomultiplier; K Kollimator (Bohrung 18 mm); A und P werden von den Armen eines schweren Stativs getragen

cooling rate:

Energy Landscape Model

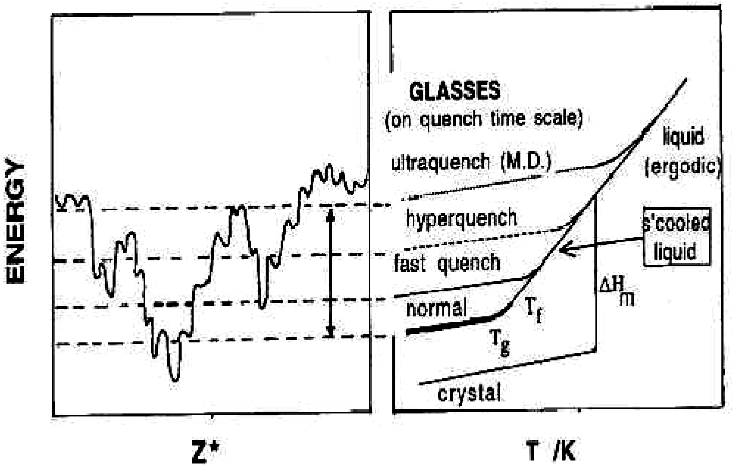


FIGURE 1. Depiction of the relation between the energy of glasses and the rate of quenching. LHS shows the trapped system energy in relation to its energy landscape, represented in the common (but highly over-simplified) two dimensional form appropriate to constant volume systems.

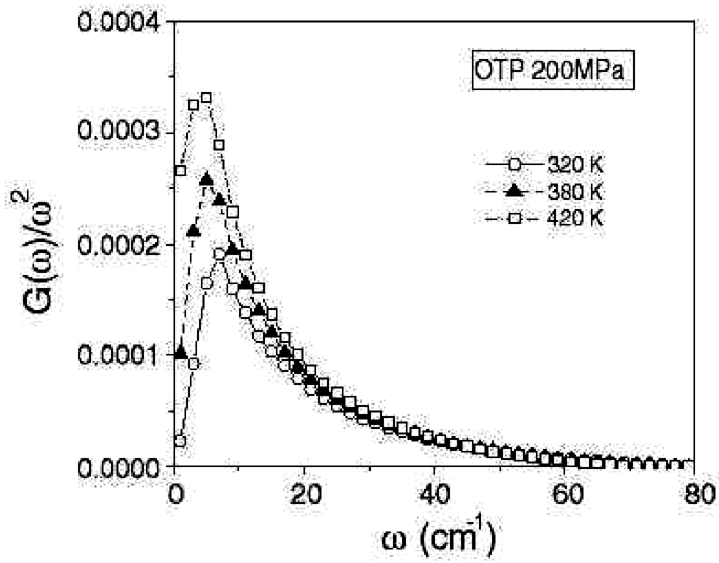


Figure 4. The second moment of the VDOS for a model molec inherent structures characterized by the structural temperatures peak intensity increases with increasing structural temperature.

C.A.Angell *et al.*, ~JPCM 15, S1051 (2003)

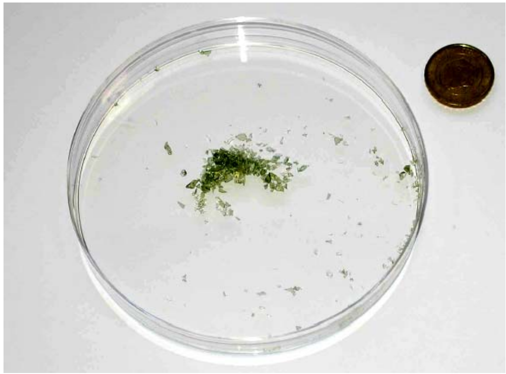
Dependence of DOS on cooling rate:

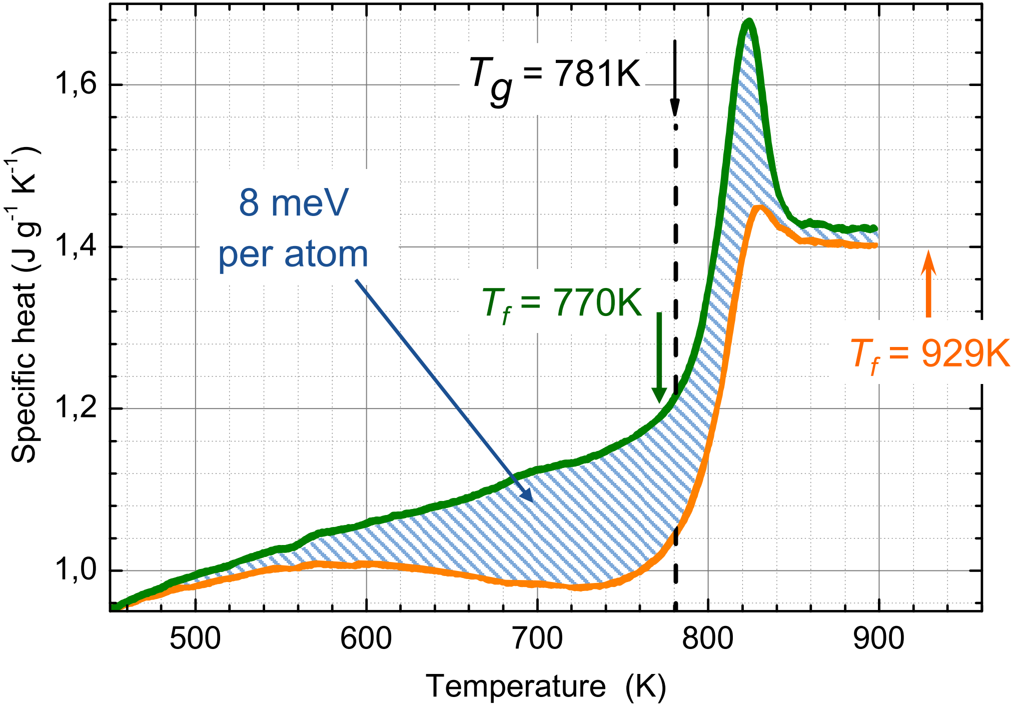
- quenched from melt to water with a cooling rate of 1500 K/s,
- annealed for 30 min at *Tg* + 4K, cooled down with a rate of 2 K/s

Sodium silicate glass:

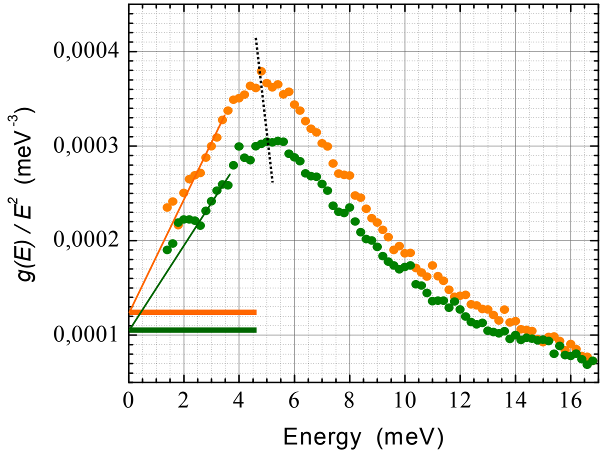
$(SiO_2)_{74}$ $(Na_2O)_{16}$ $(FeO)_6$ $(CaO)_4$

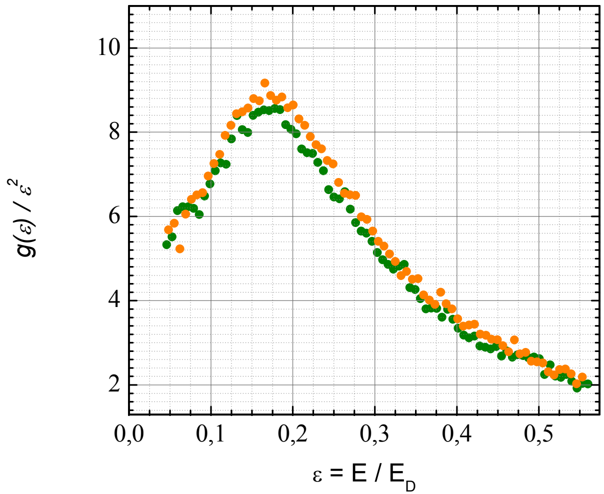
Tg = 781 K





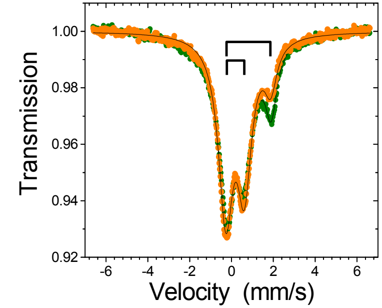
A.Monaco *et al*, PRL **96** 205502 (2006)



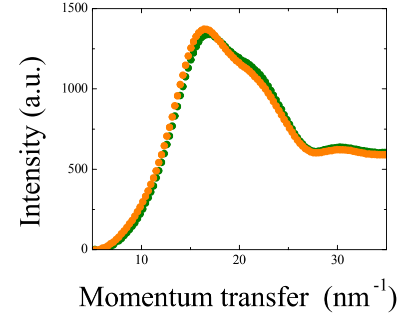


- quenched
- annealed

microscopic properties: Mössbauer spectroscopy



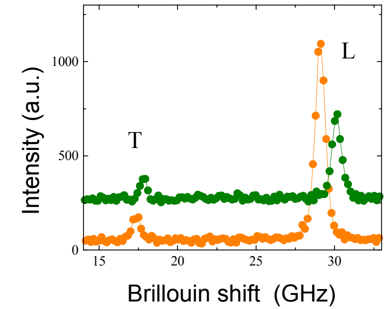
intermediate-range properties: X-Ray Scattering



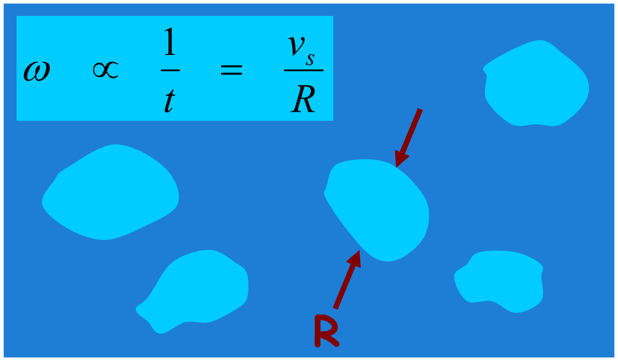
macroscopic properties: ρ & Brillouin Light Scatt.

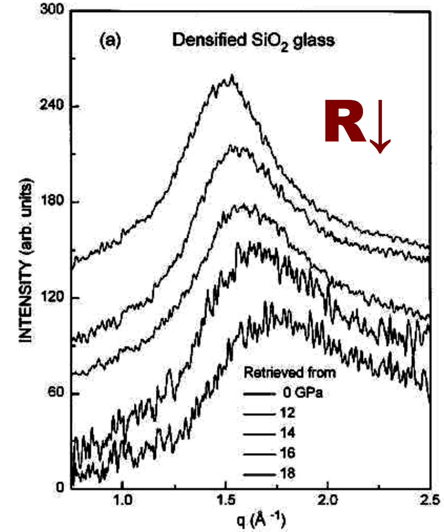
ρ = 2.404(1) g/cc

ρ = 2.439(5) g/cc



density:
cluster model





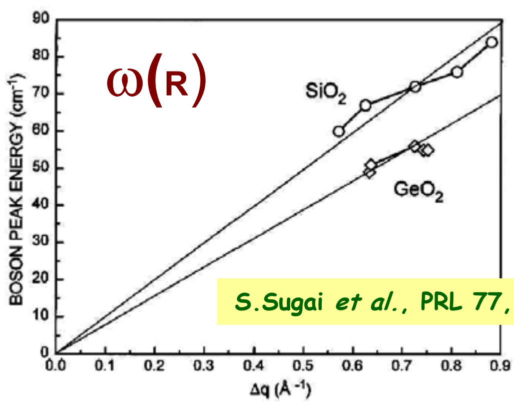
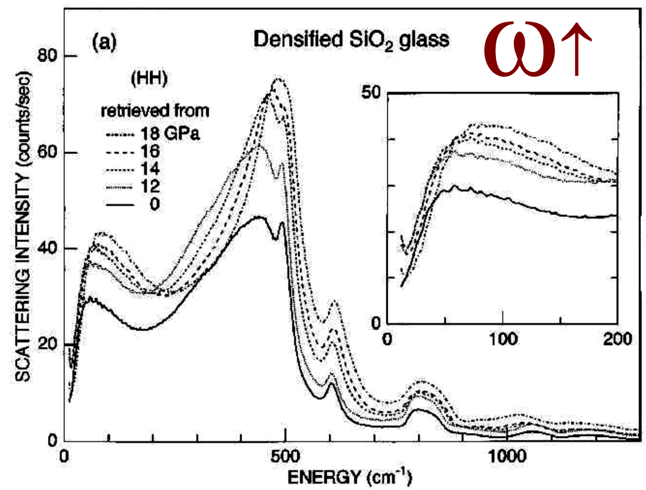


FIG. 5. Boson peak energy as a function of Δq.

S.Sugai et al., PRL 77, 4210 (1996)



Dependence of DOS in glasses on density:

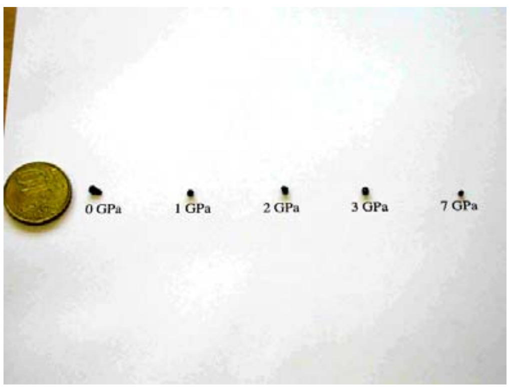
Heated up to *Tg*-70K, densified for 10 min at 1 GPa
at 2 GPa
at 3 GPa

Permanently densified samples:

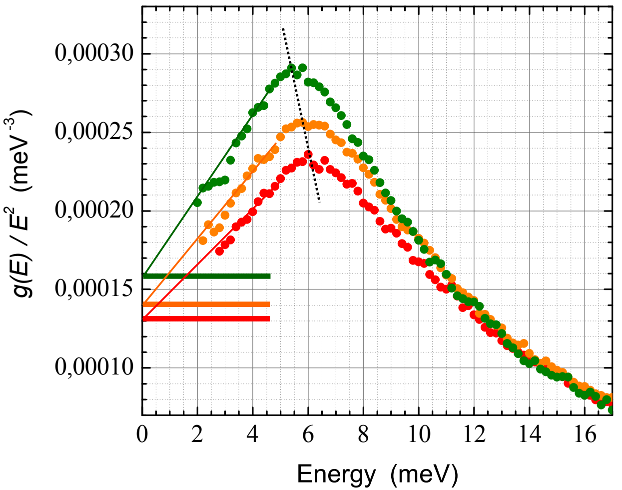
sodium silicate glass

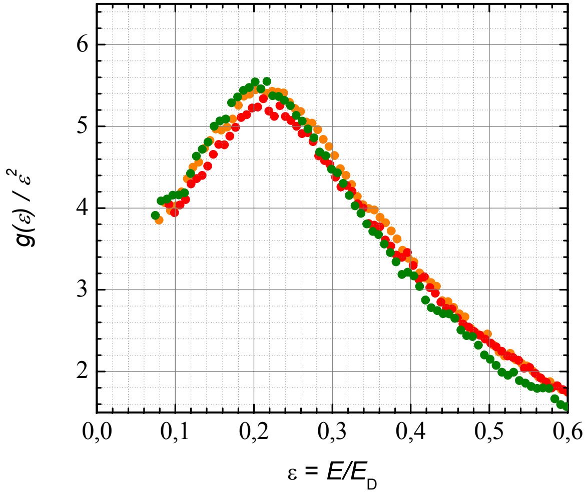
Na_2 Fe Si_3 $O_{8.5}$

Tg = 744 K



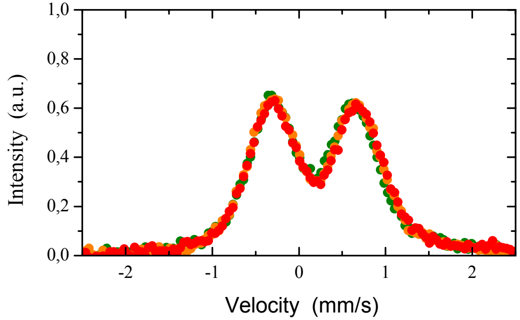
A.Monaco *et al*, PRL **97** 135501 (2006)



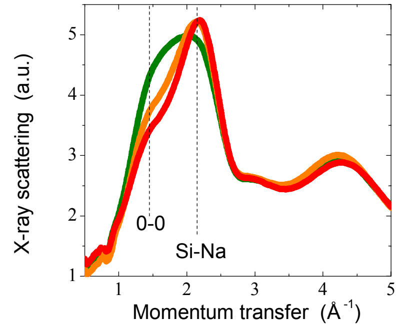


microscopic properties: Mössbauer spectroscopy

- at 1 GPa
- at 2 GPa
- at 3 GPa



intermediate-range properties: X-Ray Scattering

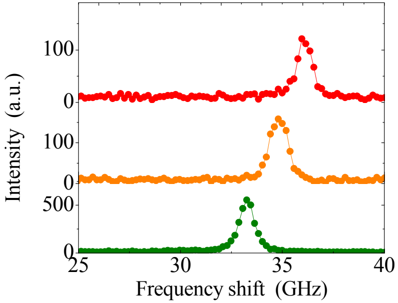


macroscopic properties: ρ & Brillouin Light Scatt.

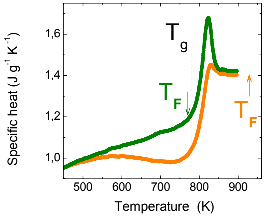
ρ = 2.72(2) g/cc

ρ = 2.87(2) g/cc

ρ = 2.88(4) g/cc

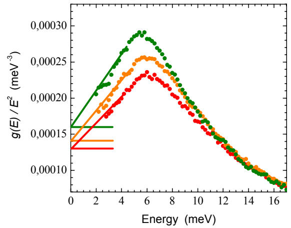


Already summary ?

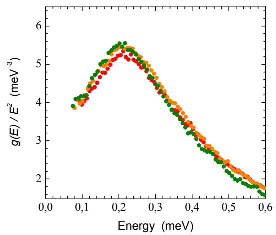


For glasses with various cooling rate (*Tf* = ***Tg*-10K**, ***Tg*+150K**) ;

density ($\Delta\rho/\rho$ **= 6%**) .



Extrapolation of DOS to $E \to 0$ is consistent with the Debye level
as E=0 there is nothing but sound



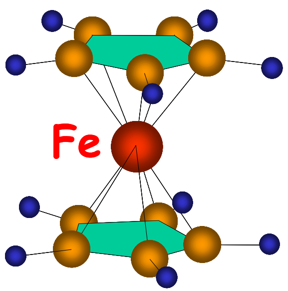
Transformation of DOS is described by changes of elastic medium
as it should be for sound waves

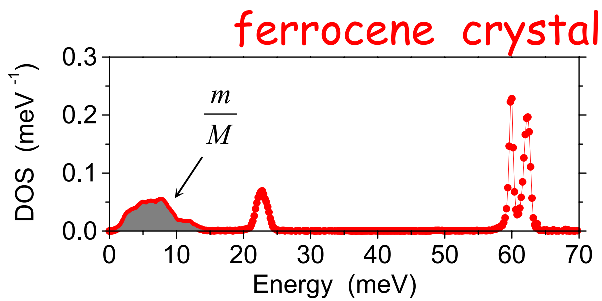
Functional dependence?

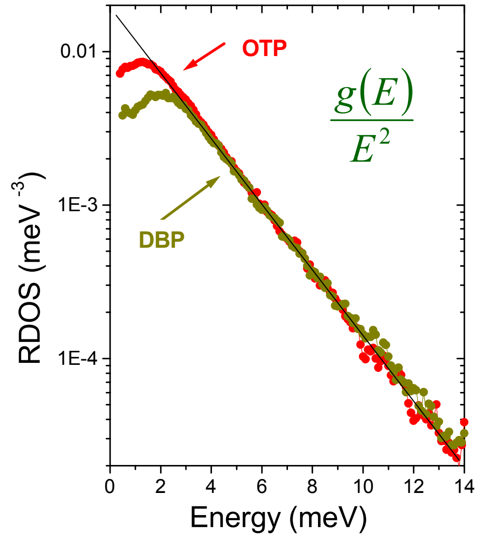
functional dependence:

generic motions of molecules and occasional molecular modes

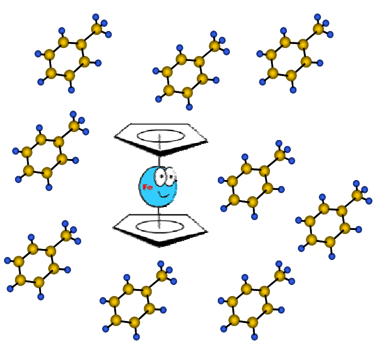
ferrocene

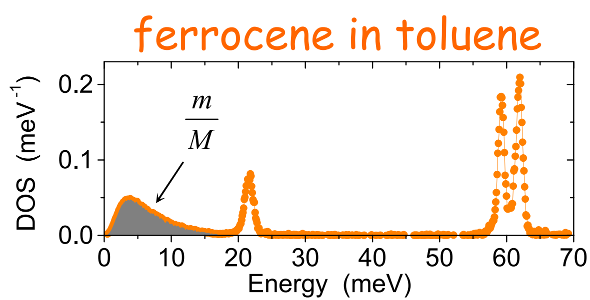






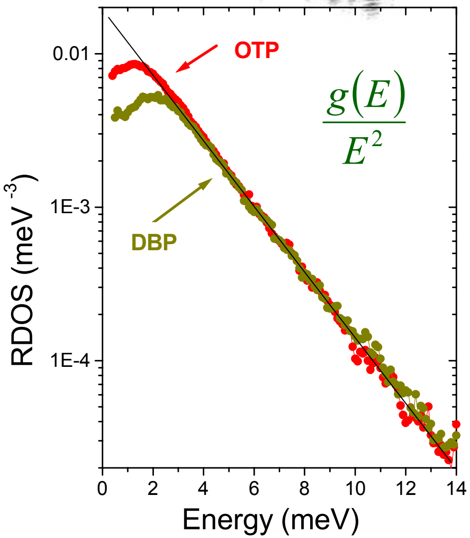
toluene

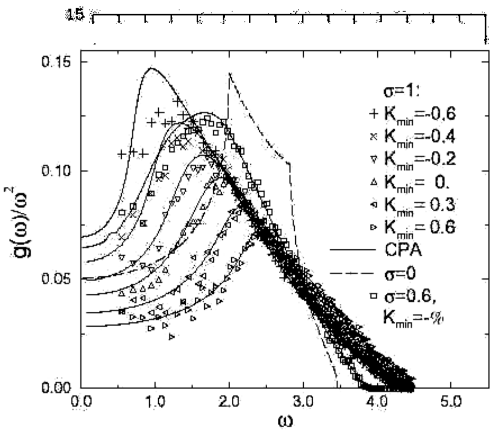




$$g(E) \propto E^2 \exp\left(-E/E_0\right)$$
$$E > E_B$$

A.Chumakov *et al*, PRL **92**, 245508 (2004)





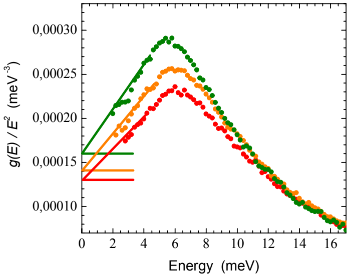
Suspected model:	yes	no
Localized vibrations in a cluster		✓
Same, but with various cluster sizes		✓
Soft potential modes		✓
Mode coupling theory		✓
Sound waves in disordered medium	✓	

positive match!

$E^2 \exp(-E/E_0)$

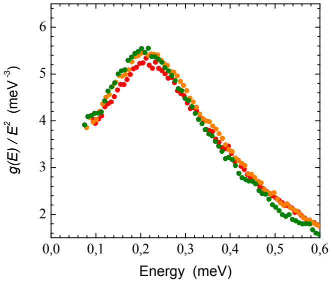
functional behavior is consistent with sound wave model

Summary:



Extrapolation of DOS to $E \rightarrow 0$ is consistent with the Debye level

at $E \rightarrow 0$ there is nothing but sound

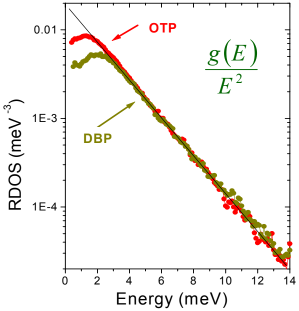


Transformation of DOS is described by changes of elastic medium

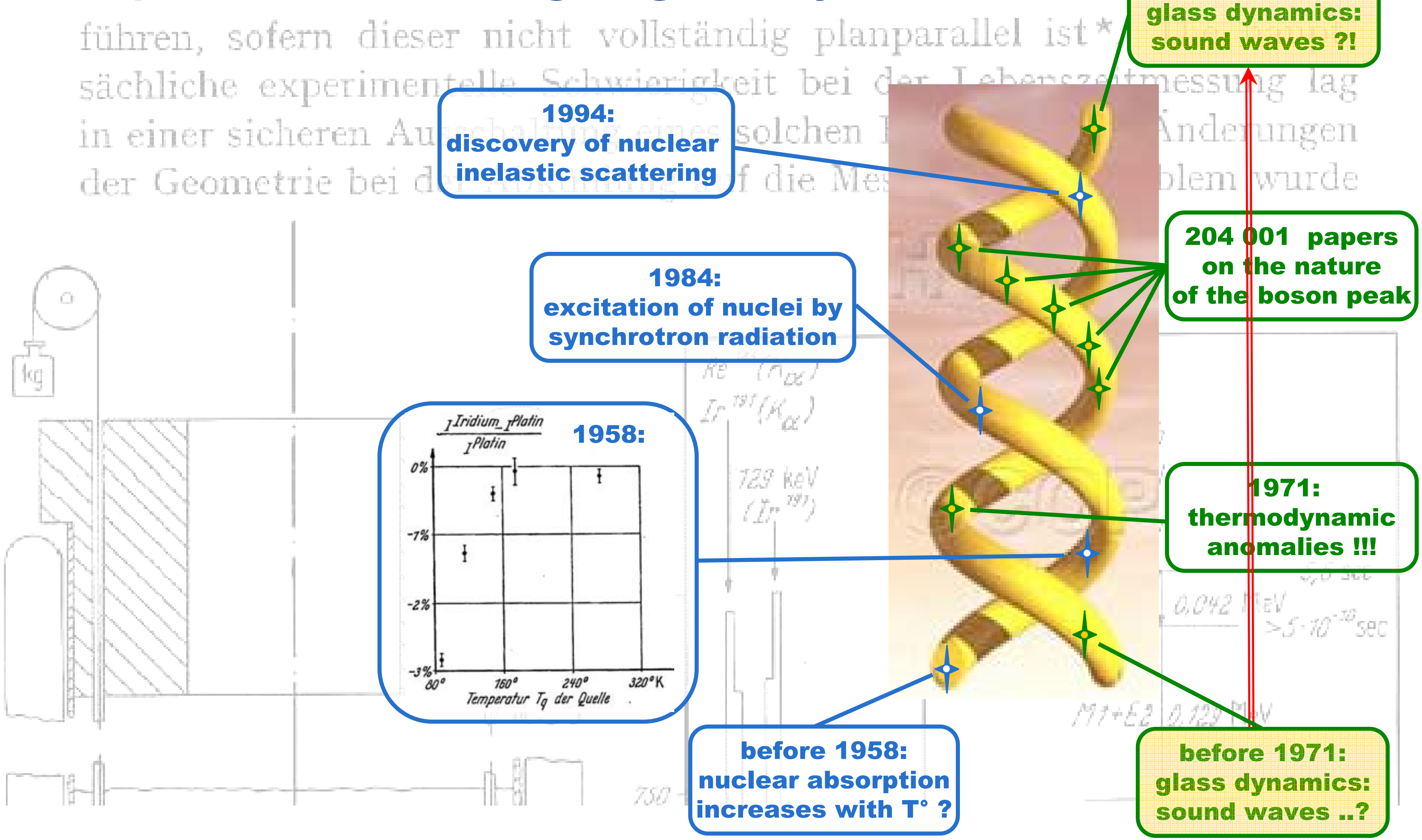
entire DOS transforms like sound

DOS follow functional dependence of sound waves

at E above the boson peak:
nothing but sound

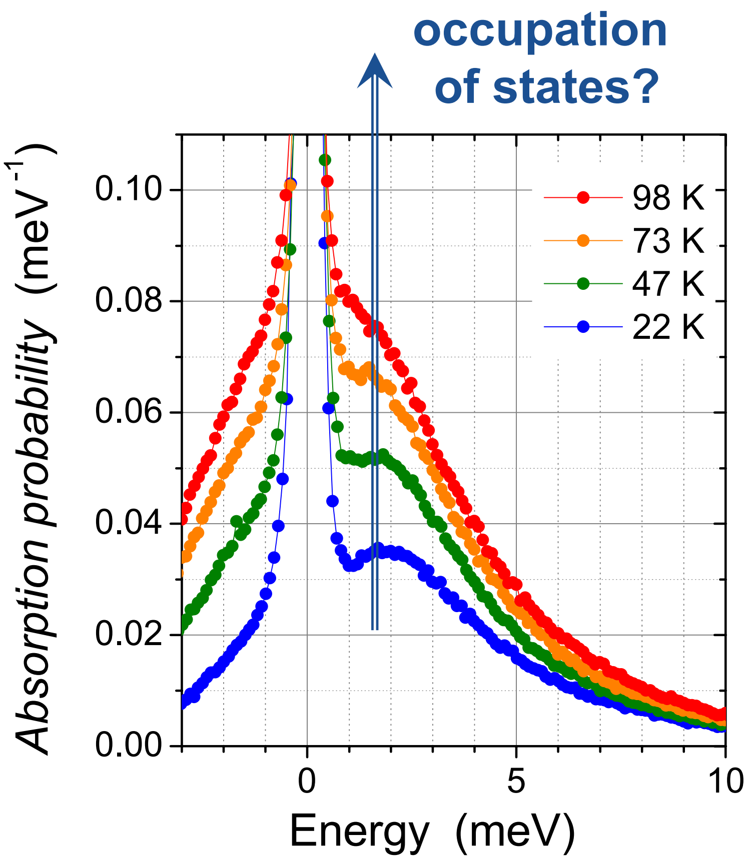


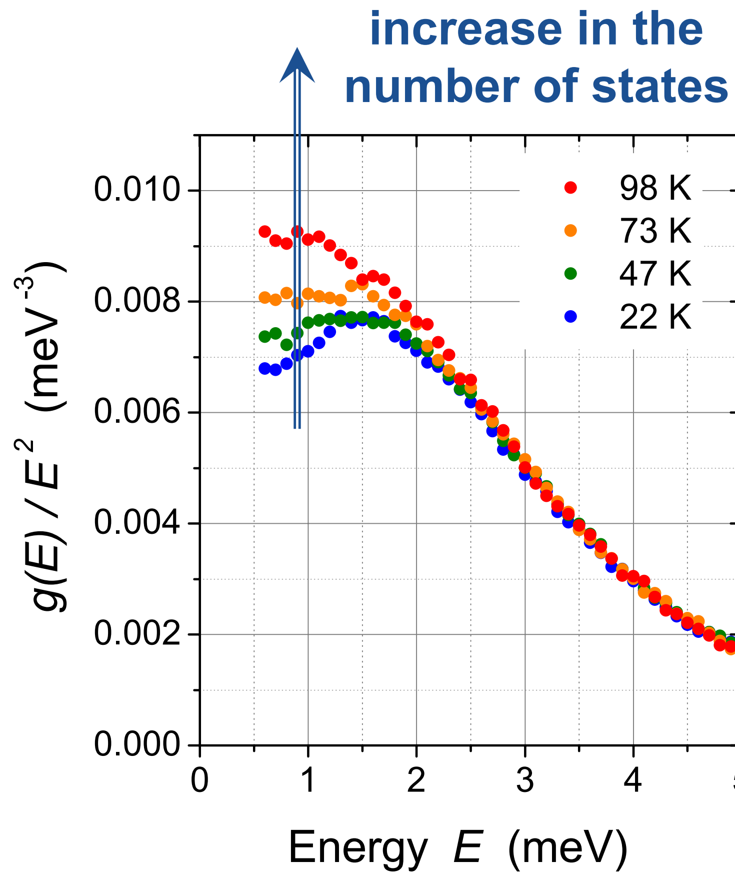
a loop in understanding of glass dynamics:



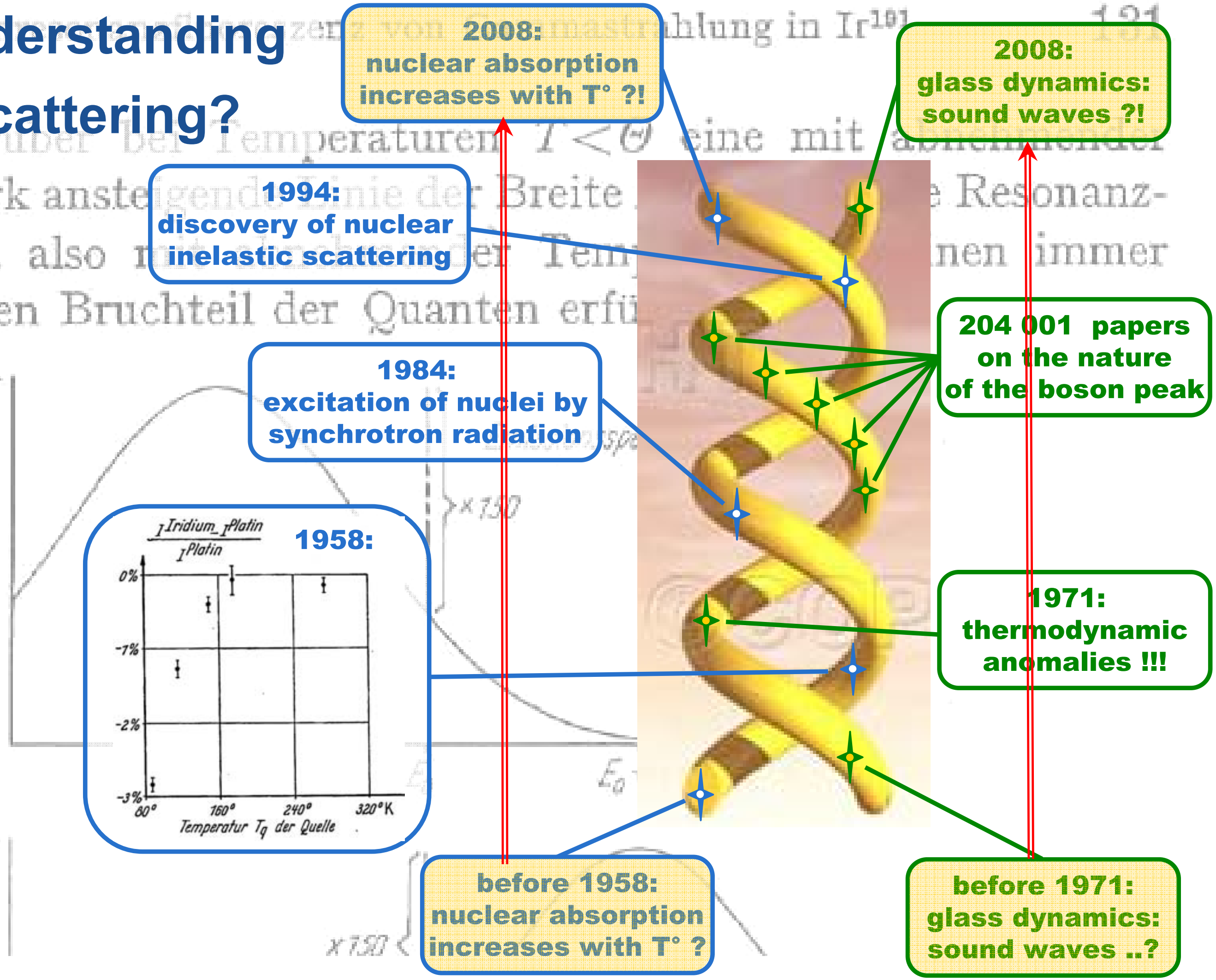
temperature dependence of nuclear absorption:

toluene glass





a loop in understanding of nuclear scattering?



Acknowledgements

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