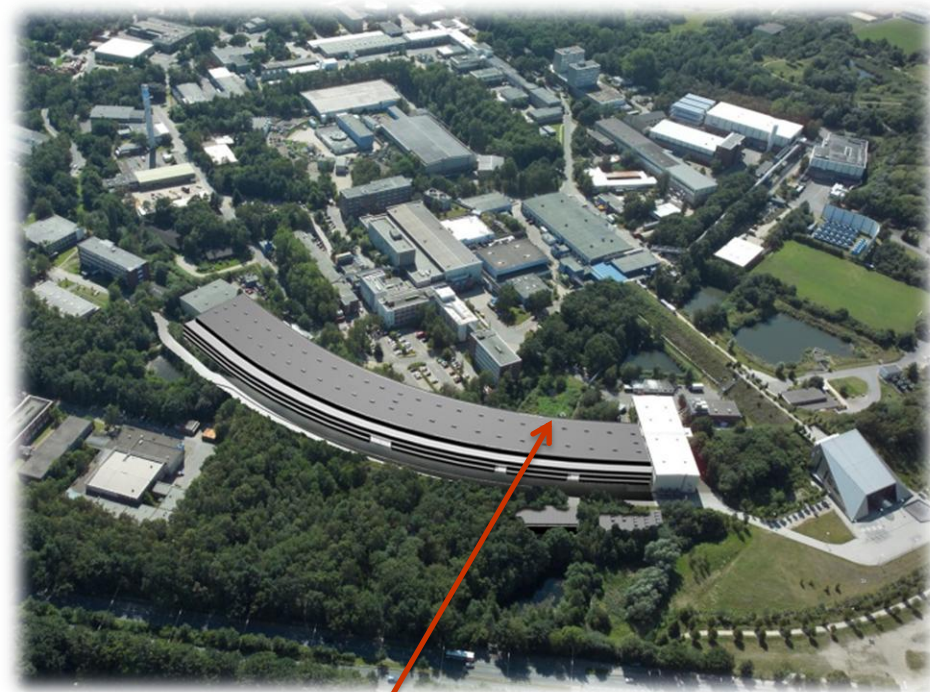


Ядерное резонансное рассеяние СИ при энергиях выше 30 кэВ. Развитие и применение метода.

Илья Сергеев, PETRAIII, Hamburg, Germany



P01 @ PETRA III

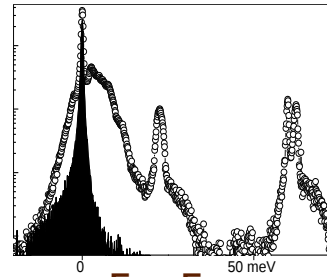
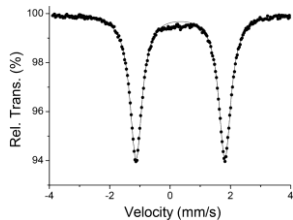


ID18 @ ESRF

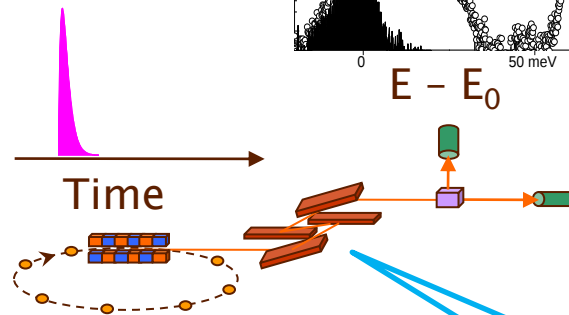
Линии ядерного резонансного рассеяния:

Nuclear Resonance Scattering

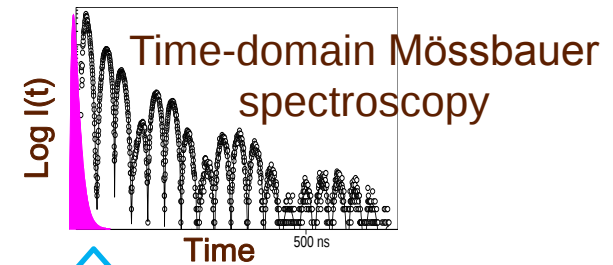
Mössbauer spectroscopy



Nuclear Inelastic Scattering
gives phonon density of states
nuclear resonance works as analyzer



Fourier transformation



Combines properties of

Mössbauer spectroscopy

- element (isotope) selectivity,
- sensitivity to electronic, magnetic and lattice properties

Synchrotron Radiation

- small beam size
- beam collimation

Key optical element –
high resolution monochromator
 $\Delta E \sim 1 - 5 \text{ meV}$

Mössbauer isotopes

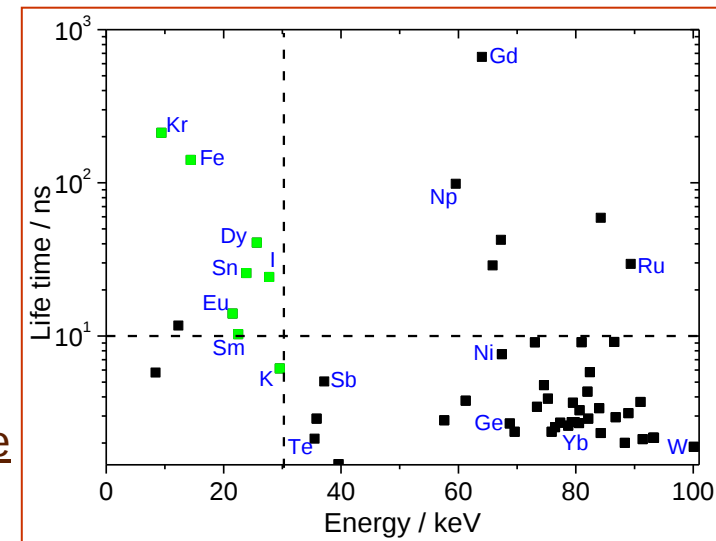
H	W - Mössbauer isotopes																He	
Li	Be											B	C	N	O	F	Ne	
Na	Mg	Fe - Nuclear Resonance Scattering										Al	Si	P	S	Cl	Ar	
K	Ca	Sc		Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y		Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**																

Lanthanides*	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Actinides**	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Available Mössbauer isotopes:
time MS:

 $^{57}\text{Fe}, ^{119}\text{Sn}, ^{151}\text{Eu}, ^{149}\text{Sm}, ^{161}\text{Dy}, ^{121}\text{Sb}, ^{125}\text{Te}, ^{61}\text{Ni}, ^{73}\text{Ge}$

NIS:

 $^{57}\text{Fe}, ^{119}\text{Sn}, ^{151}\text{Eu}, ^{149}\text{Sm}, ^{161}\text{Dy}, ^{121}\text{Sb}, ^{125}\text{Te}$ 

Applications of time Mössbauer spectroscopy

Collimation of the beam



High brilliance of the source



- measurements under extreme conditions
high / low T
high pressure
magnetic field
- surfaces and nanoscale materials

PRL **109**, 026403 (2012)

PHYSICAL REVIEW LETTERS

week ending
13 JULY 2012

Reentrant Valence Transition in EuO at High Pressures: Beyond the Bond-Valence Model

N. M. Souza-Neto,^{1,2,*} J. Zhao,¹ E. E. Alp,¹ G. Shen,³ S. V. Sinogeikin,³ G. Lapertot,⁴ and D. Haskel^{1,†}

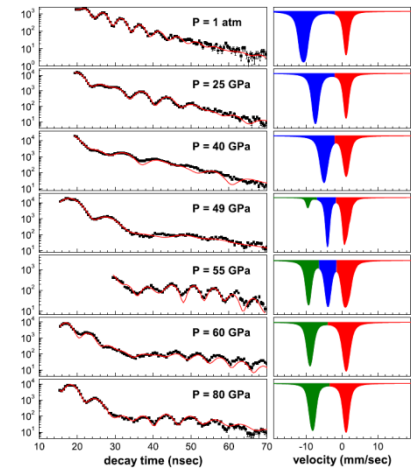
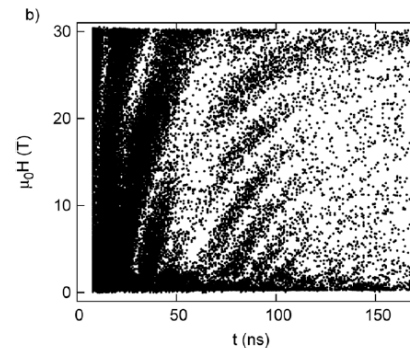
PRL **104**, 087601 (2010)

PHYSICAL REVIEW LETTERS

week ending
26 FEBRUARY 2010

Nuclear Forward Scattering of Synchrotron Radiation in Pulsed High Magnetic Fields

C. Strohm,^{*} P. Van der Linden, and R. Rüffer



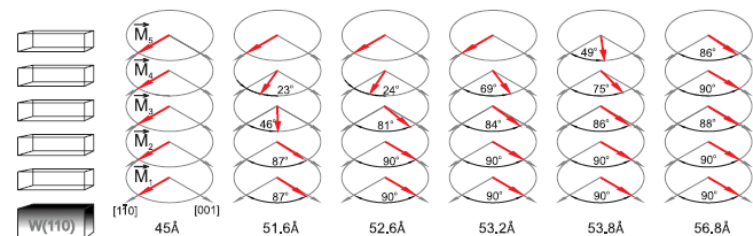
PRL **105**, 027206 (2010)

PHYSICAL REVIEW LETTERS

week ending
9 JULY 2010

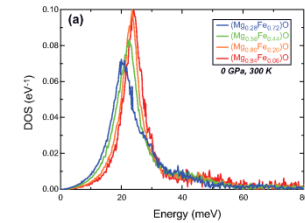
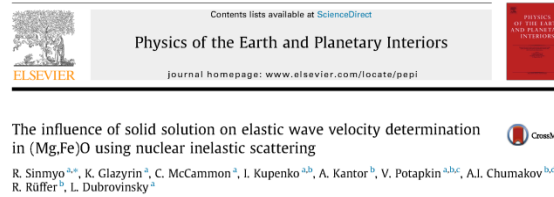
Noncollinear Magnetization Structure at the Thickness-Driven Spin-Reorientation Transition in Epitaxial Fe Films on W(110)

T. Ślęzak,¹ M. Ślęzak,¹ M. Zajac,^{1,2} K. Freindl,^{1,3} A. Kozioł-Rachwał,¹ K. Matlak,¹ N. Spiridis,³ D. Wilgocka-Ślęzak,³ E. Partyka-Jankowska,⁴ M. Rennhofer,^{1,*} A. I. Chumakov,² S. Stankov,^{2,†} R. Rüffer,² and J. Korecki^{1,3}



Applications of NIS

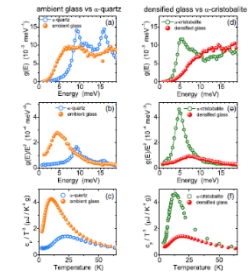
- Geoscience
- glass physics
- nanoscale materials
- thermoelectrics



PRL 112, 025502 (2014) PHYSICAL REVIEW LETTERS week ending 17 JANUARY 2014

Role of Disorder in the Thermodynamics and Atomic Dynamics of Glasses

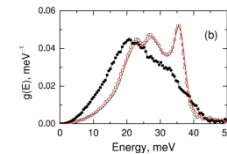
A. I. Chumakov,^{1,*} G. Monaco,^{2,1} A. Fontana,^{2,3} A. Bosak,¹ R. P. Hermann,^{4,5} D. Bessas,^{4,5,1} B. Wehinger,¹ W. A. Crichton,^{1,4} M. Krisch,¹ R. Rüffer,¹ G. Baldi,⁶ G. Carini Jr.,⁷ G. Carini,⁸ G. D'Angelo,⁸ E. Gilotti,⁶ G. Tripodo,⁸ M. Zanatta,^{9,3} B. Winkler,¹⁰ V. Milman,¹¹ K. Refson,¹² M. T. Dove,¹³ N. Dubrovinskaia,¹⁴ L. Dubrovinsky,¹⁵ R. Keding,¹⁶ and Y. Z. Yue^{17,8}



PRL 100, 235503 (2008) PHYSICAL REVIEW LETTERS week ending 13 JUNE 2008

Vibrational Properties of Nanograins and Interfaces in Nanocrystalline Materials

S. Stankov,¹ Y. Z. Yue,^{2,3} M. Miglierini,⁴ B. Sepiol,⁵ I. Sergueev,¹ A. I. Chumakov,¹ L. Hu,^{2,3} P. Svec,⁶ and R. Rüffer¹



Nanoscale

RSC Publishing

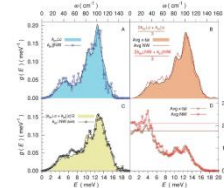
PAPER

View Article Online
View Journal | View Issue

Phonon spectroscopy in a Bi₂Te₃ nanowire array

Cite this: *Nanoscale*, 2013, 5, 10629

Dimitrios Bessas,^{†ab} William Tollner,^c Zainul Aabdin,^d Nicola Peranio,^d Ilya Sergueev,^{†e} Hans-Christian Wille,^f Oliver Eibl,^g Kornelius Nielsch^c and Raphaël P. Hermann^{*ab}



Mössbauer Spectroscopy with ^{61}Ni and ^{73}Ge

^{61}Ni :

$E = 67.4 \text{ keV}$

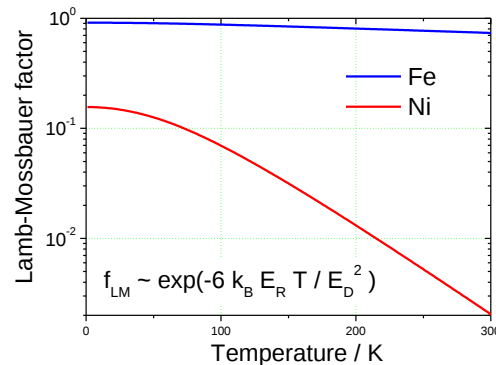
$\tau_0 = 7.60 \text{ ns}$

transition: $3/2^- \rightarrow 5/2^-$

Sources:

- ^{61}Co (β^- , 99 min)
- ^{61}Cu (EC, 3.41 h)

High energy $\Rightarrow$
small Lamb-Mössbauer factor



^{73}Ge :

$E = 68.75 \text{ keV}$

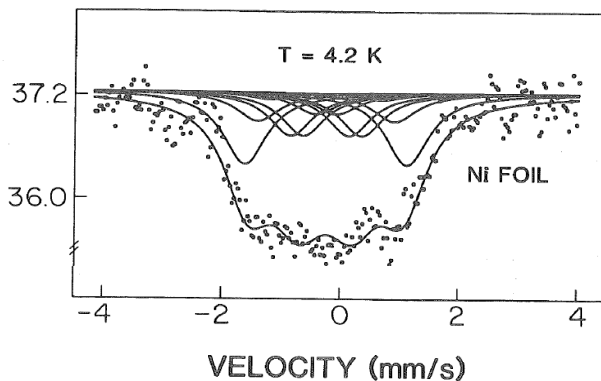
$\tau_0 = 2.51 \text{ ns}$

transition: $9/2^+ \rightarrow 7/2^+$

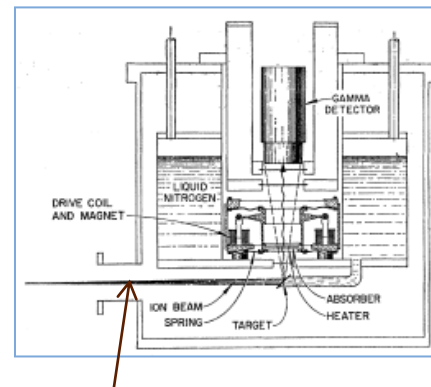
Source:

Coulomb excitation

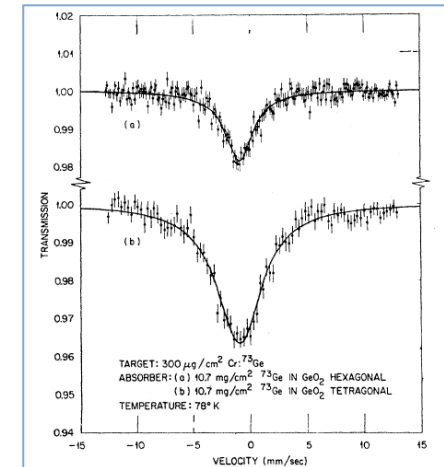
McCammon et al, *Hyp. Int.* **28** (1986) 483



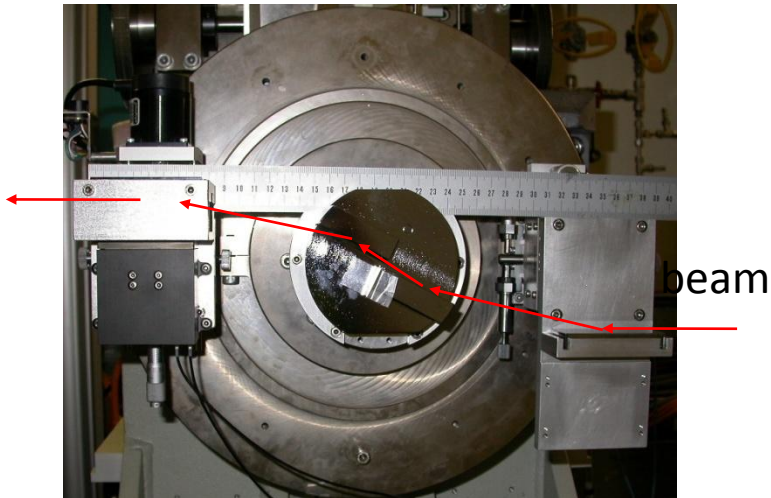
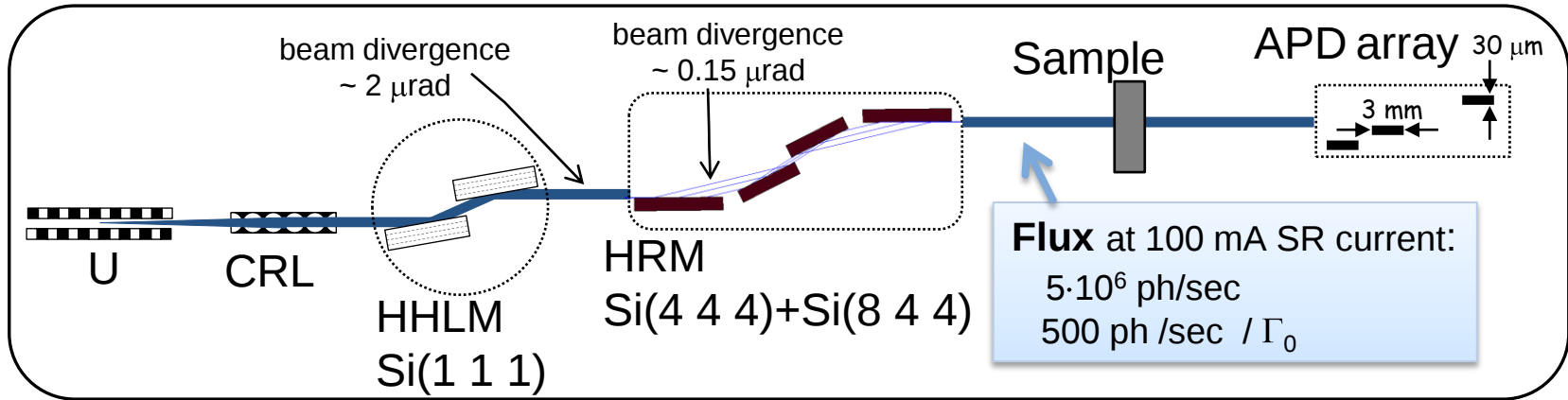
Czjzek et al, *Physical Review* **174** (1968) 331



Ion beam



Experimental setup



Detector:
Array of 16 Si APDs
Efficiency
@ 67keV ~ 20%

Time MS with ^{61}Ni and ^{73}Ge

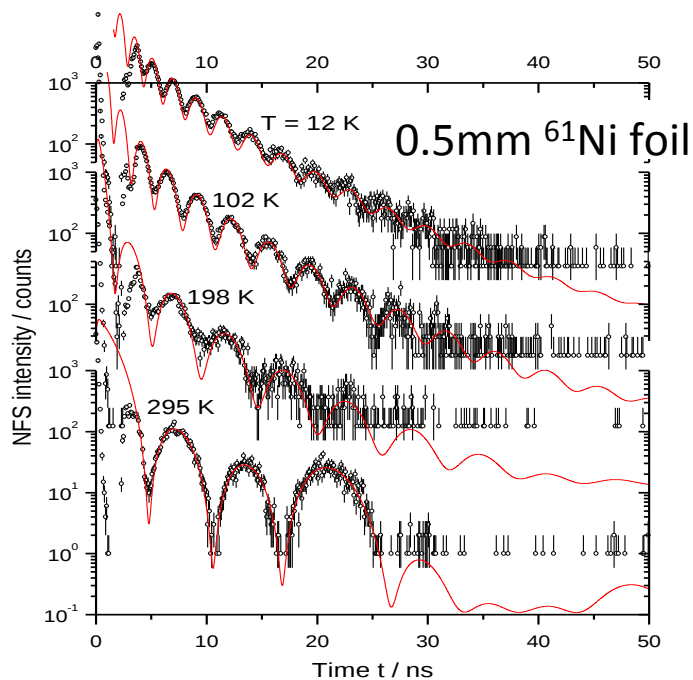
^{61}Ni , $E = 67.413 \text{ keV}$

rotate x-tals by $0.1 - 0.3^\circ$

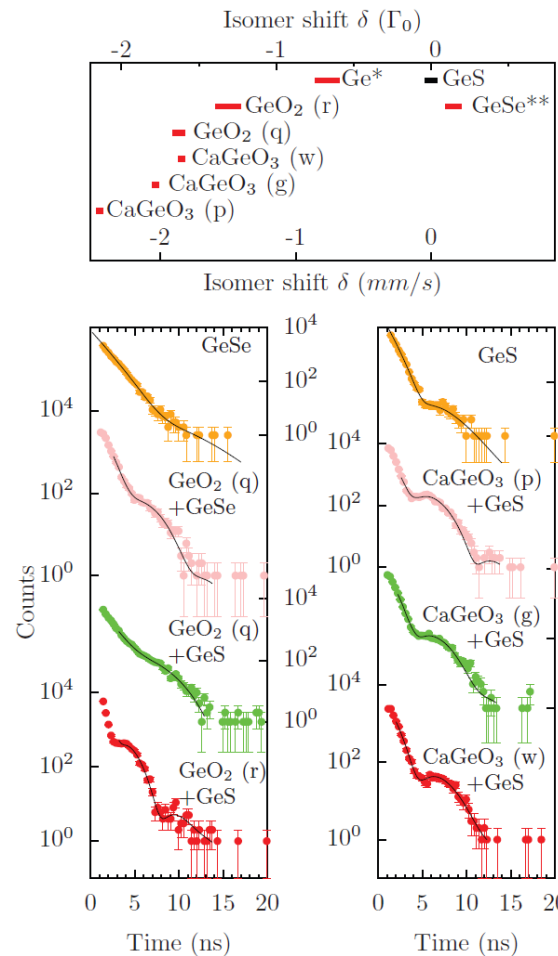
^{73}Ge , $E = 68.752 \text{ keV}$

Range: $64 \text{ keV} < E < 77 \text{ keV}$

Magnetic and elastic properties
of elemental nickel



I. Sergueev et al., PRL 99(2007)097601.



Isomer shift
(coordination number)
of CaGeO_3 :

wollastonite
garnet
perovskite

R. Simon et al., EPL 104(2013) 17006



Highest pressure for magnetism

X-ray diffraction: fcc phase up to 150 / 200 GPa,

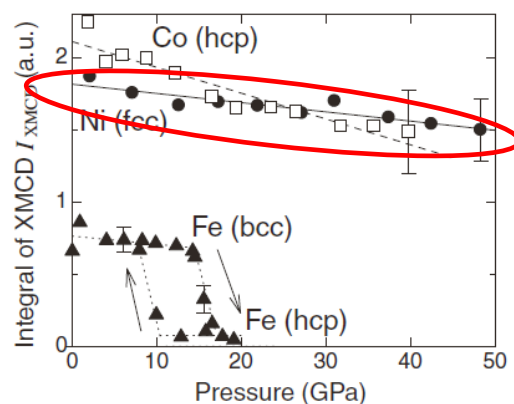
Theory: predicts continuous decrease of magnetic moment up to 300 – 34000 GPa

X-ray magnetic dichroism

Spring-8

Up to 50 GPa

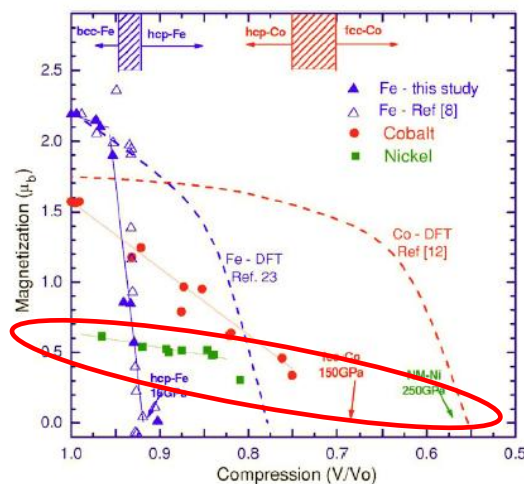
Ishimatsu et al., *J. Phys.Soc.Jpn.*,
76(2007)064703



APS

Up to 70 GPa

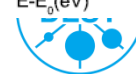
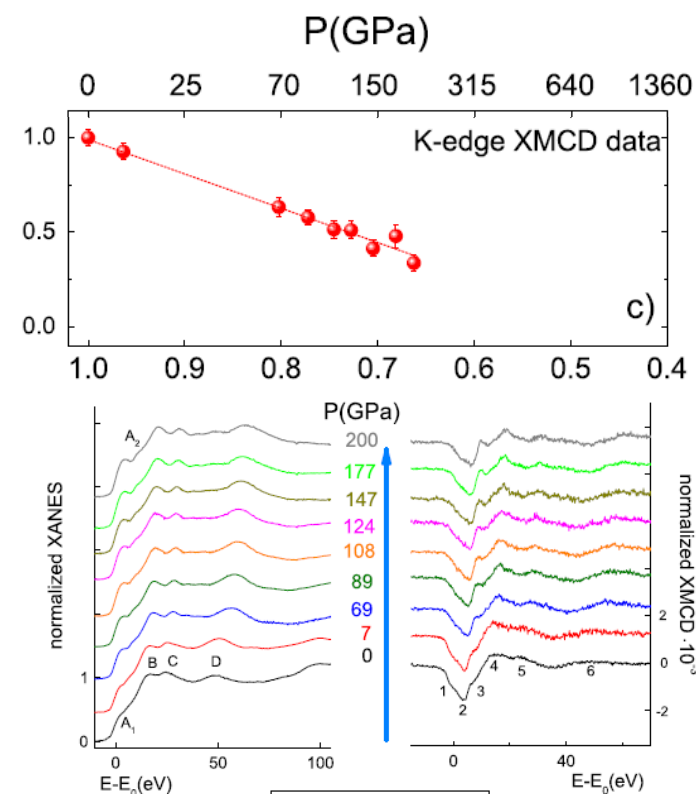
Iota et al., *Appl. Phys. Lett.*
90(2007)042505



ESRF

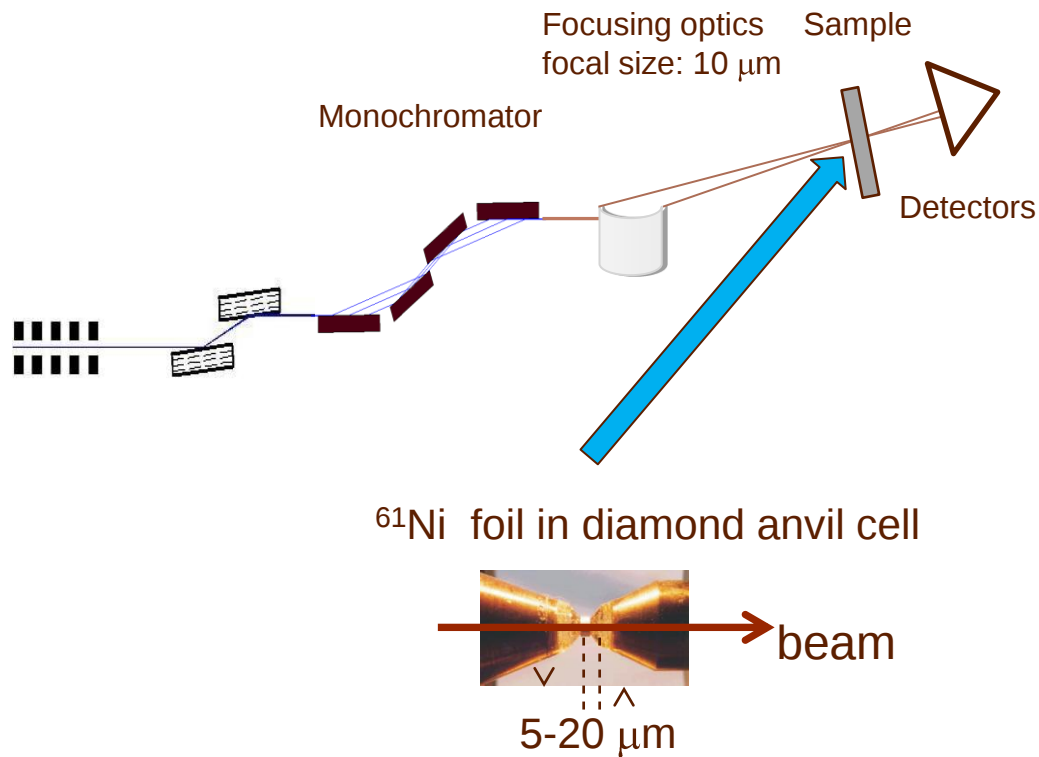
Up to 200 GPa

Torchio et al., *PRL* 107(2011)237202

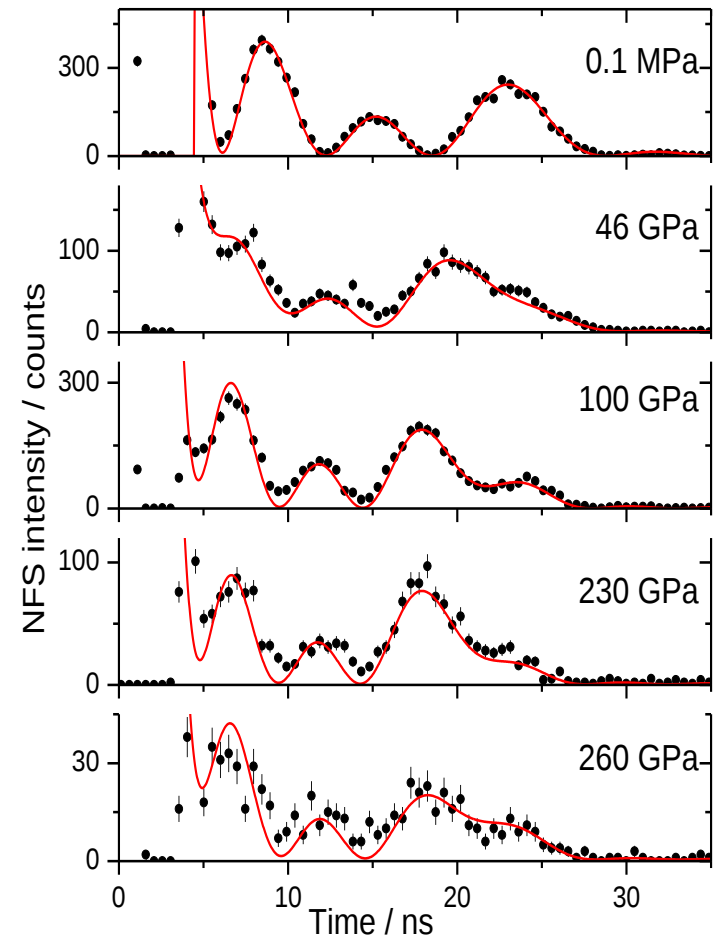


Highest pressure for magnetism

Experimental setup



Measurements



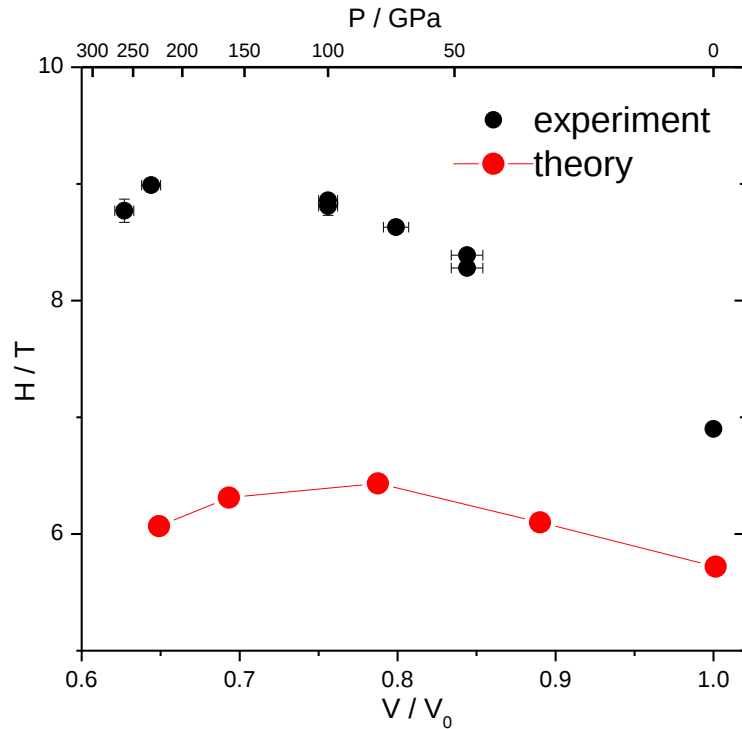
I. Sergueev et al., PRL 111(2013)157601.

H – hyperfine magnetic field



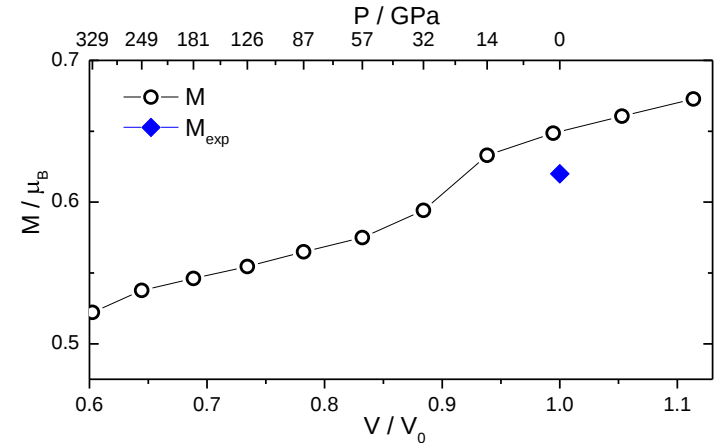
Highest pressure for magnetism

Experiment and theory

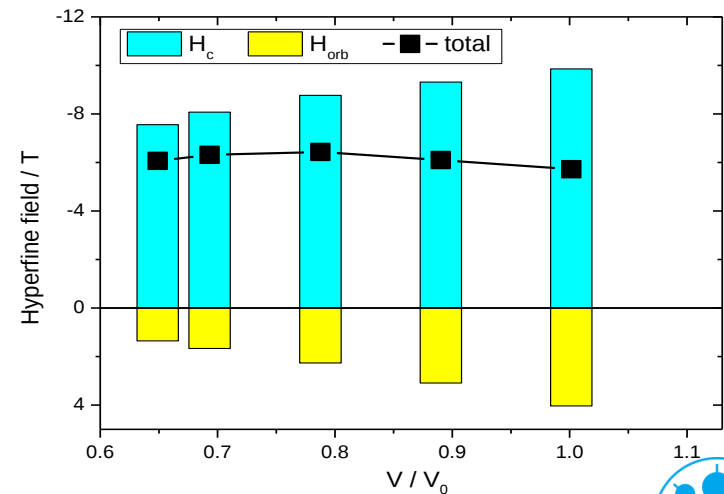


- Ni is ferromagnetic at room temperature up to 260 GPa.
- Measurements at higher pressure are required in order to find critical pressure.

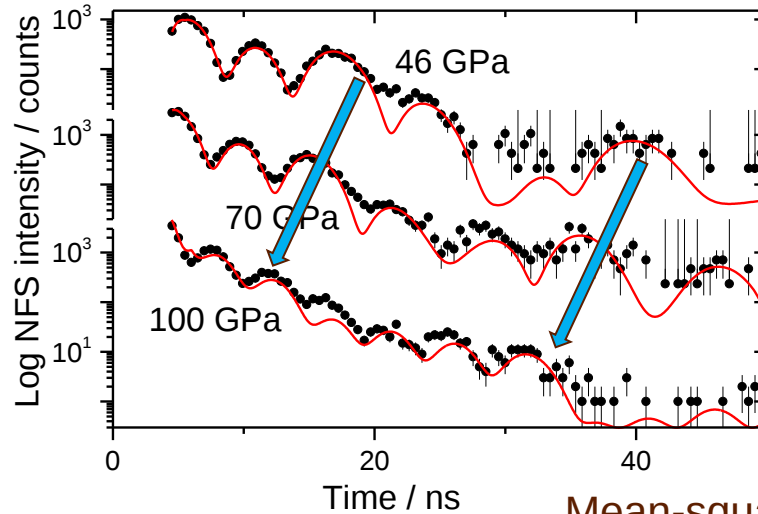
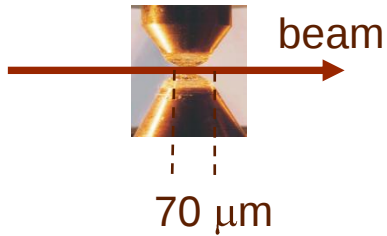
Magnetic moment vs volume (theory)



Hyp. magn. field vs volume (theory)



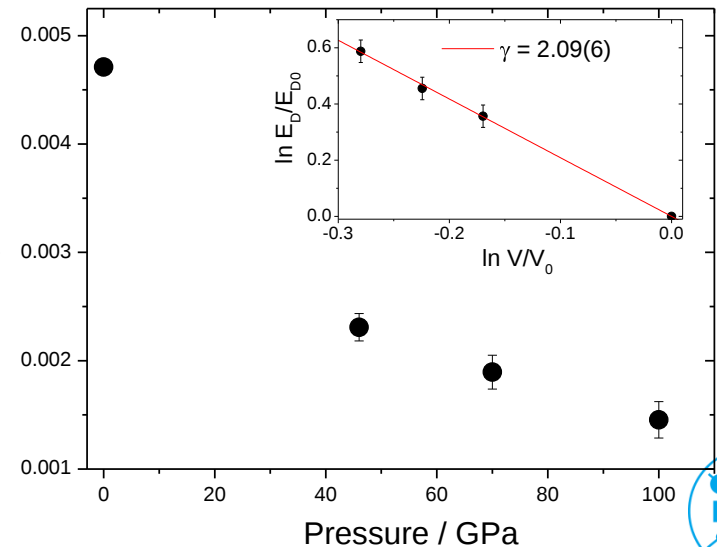
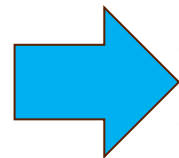
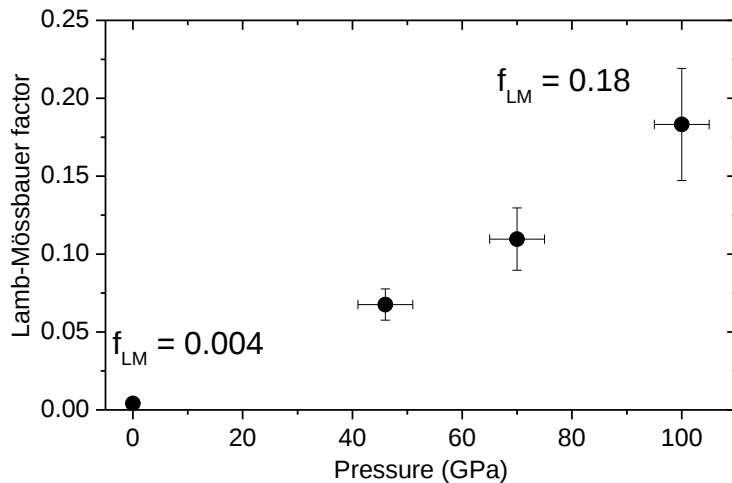
Time MS of Ni at HP. Elastic properties



Changes of the spectra are due to increase of the Lamb-Mössbauer factor

Mean-square vibrations, Debye energy, Grüneisen parameter

Lamb-Mössbauer factor at RT

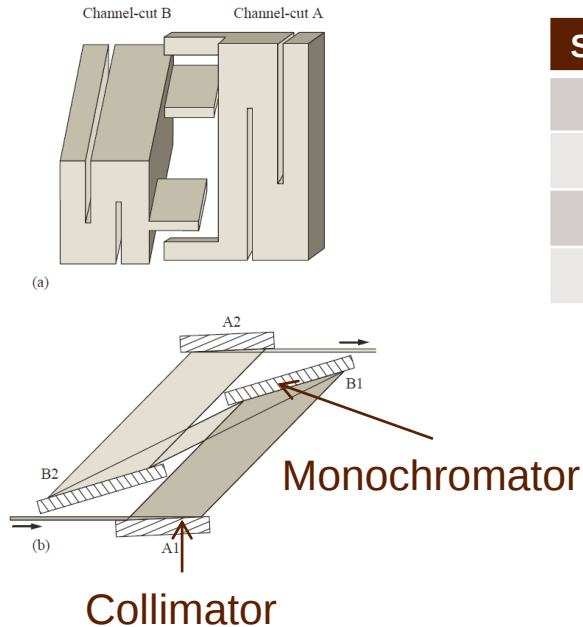


High Resolution Monochromators. Multiple-crystal approach

Si reflections with Bragg angle = 84°

Si reflection	7 7 7	12 12 12	17 17 17
E, keV	13.9	23.9	33.8
ΔE , meV	5	0.7	0.06
$\Delta\theta$, urad	3.4	0.3	0.02
R, %	80	73	22

SR beam divergence:
10-20 urad



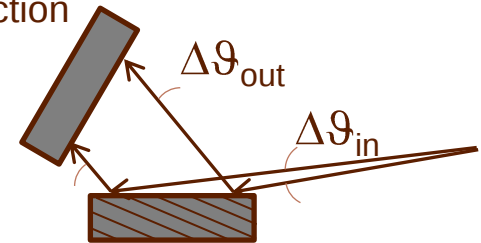
2 steps of collimation:

1. Compound refractive lenses



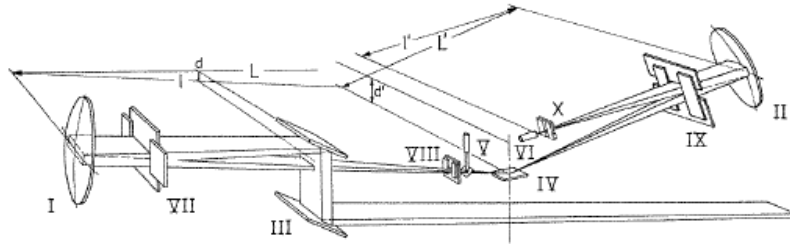
2. Asymmetric reflection

$$b = \frac{\Delta\vartheta_{in}}{\Delta\vartheta_{out}} \approx 10 \div 20$$



Backscattering Monochromators

Burkel, Rep. Prog. Phys. 63(2000)171



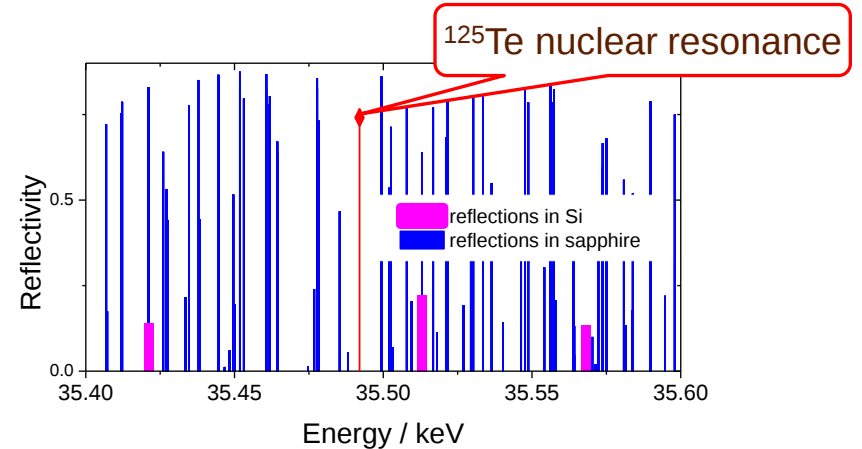
Si reflections with Bragg angle = 90°

Si reflection	7 7 7	12 12 12	17 17 17
E, keV	13.8	23.7	33.6
ΔE , meV	5	0.7	0.06
$\Delta\theta$, urad	970	320	83
R, %	80	73	22

Energy is defined by Si crystal lattice

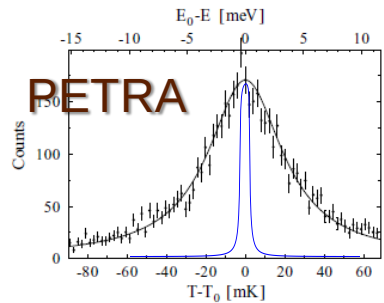
Y.V. Shvyd'ko and E. Gerdau, Hyp. Interact. 123 (1999)

Crystal with low symmetry (sapphire, quartz)



At least 2 reflections in 150-300K
for any $E > 20\text{keV}$

Sapphire backscattering monochromator



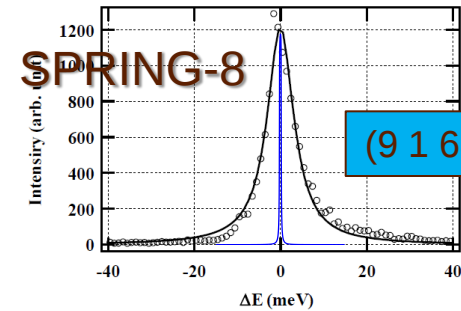
Shvyd'ko et al., EPL 56(2007)309

(3 2 52) @ 25.51 keV

$$\Delta E_{th} = 0.8 \text{ meV}$$

$$\Delta E_{exp} = 8 \text{ meV}$$

I. Sergueev, H.-C. Wille, R. P. Hermann, D. Bessas, Yu. V. Shvyd'ko, M. Zajac and R. R  ffer, J. Synchrotron Rad. 18(2011)

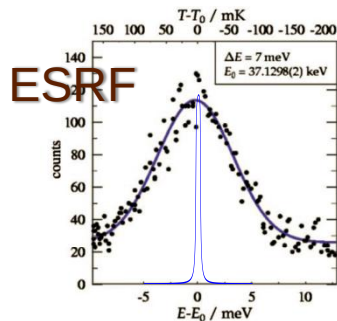


Imai et al., Proc. SPIE, 2007

(9 1 68) @ 35.49 keV

$$\Delta E_{th} = 0.4 \text{ meV}$$

$$\Delta E_{exp} = 8 \text{ meV}$$



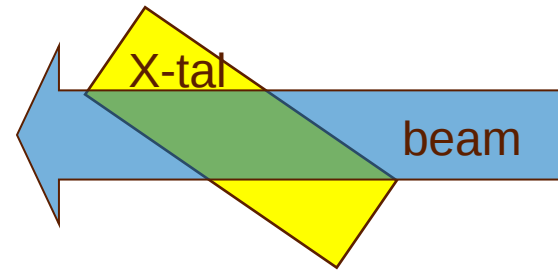
Wille et al., EPL 74(2006)170

(15 13 14) @ 37.13 keV

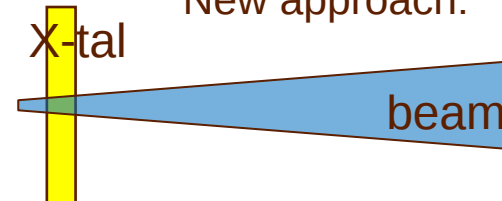
$$\Delta E_{th} = 0.4 \text{ meV}$$

$$\Delta E_{exp} = 7 \text{ meV}$$

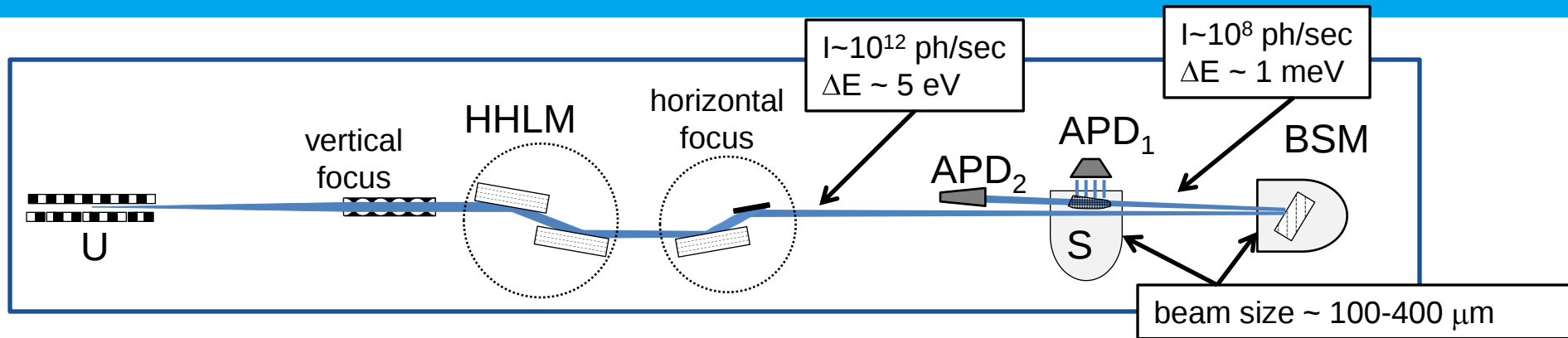
Traditional approach:



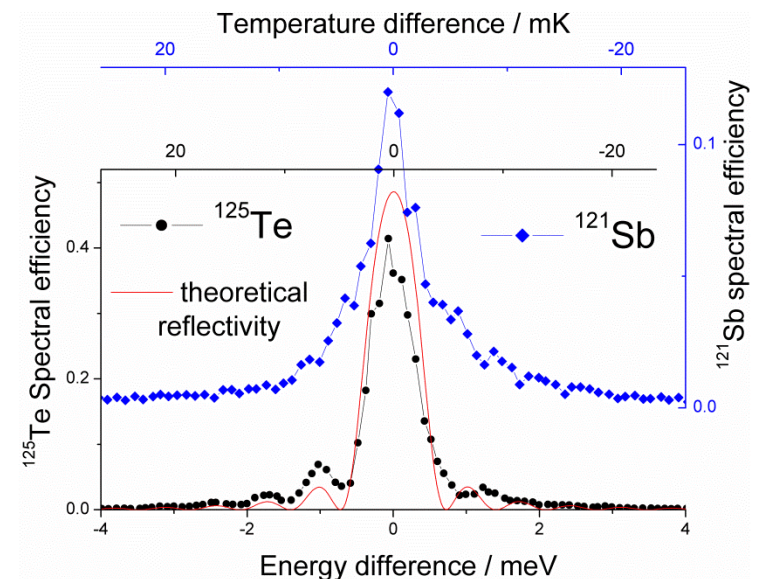
New approach:



Experimental Setup and Instrumental Functions



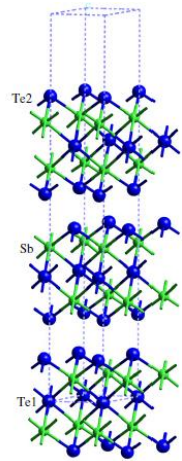
	¹²¹ Sb	¹²⁵ Te	¹¹⁹ Sn	¹²⁹ Xe
E/ keV	37.13	35.49	23.88	39.58
Reflection	(8 16 40)	(9 1 68)	(4 4 45)	(5 17 54)
T / K	237	220	193	291
Angle / degree	59	24	26	49
ΔE_{th} / meV	0.4	0.7	1.0	0.5
ΔE_{exp} / meV	0.7	0.7	1.0	0.9



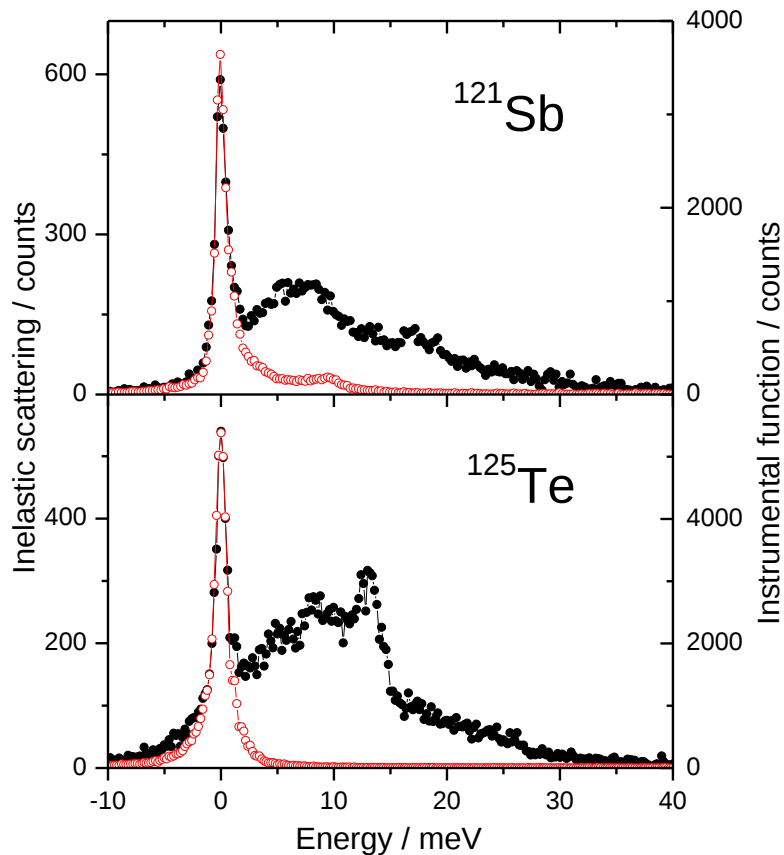
Energy band width ~ 0.7 meV,

$$\Delta E / E = 2 \cdot 10^{-8}$$

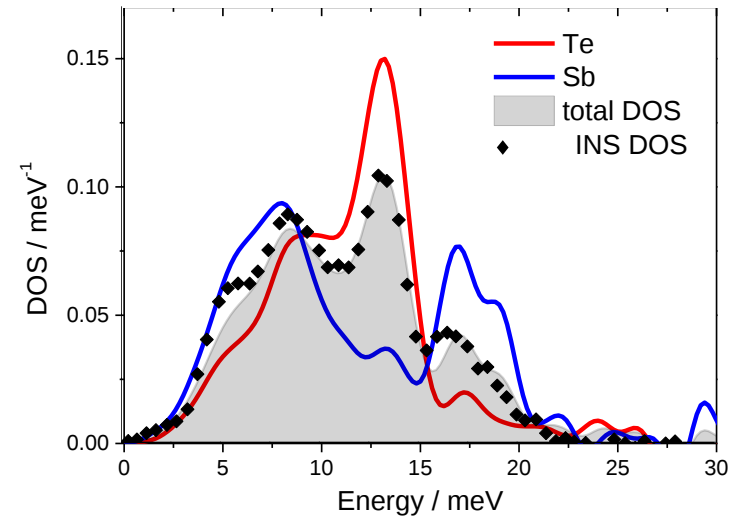
Application. Phonons in Sb_2Te_3



Inelastic spectra



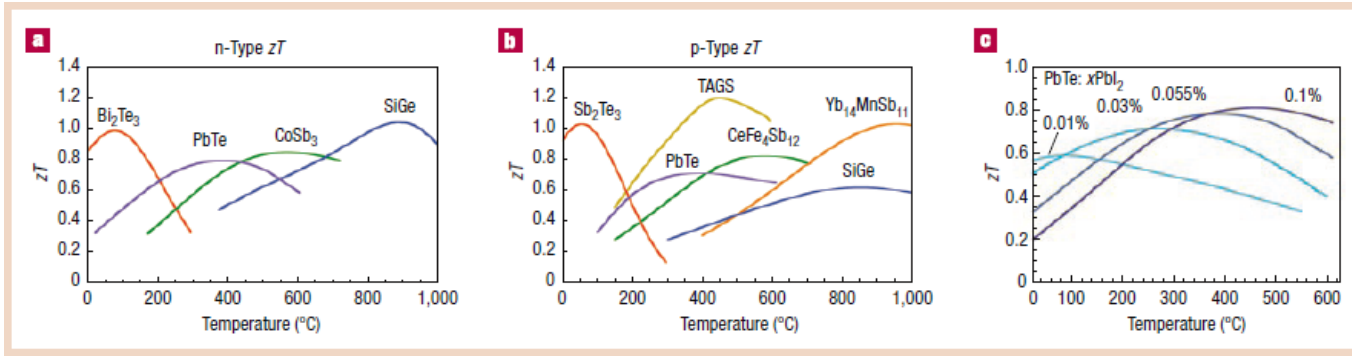
Phonon DOS



Inelastic neutron scattering
on $\text{Sb}(\text{Bi})_2\text{Te}(\text{Se})_3$
Rauh et al., J. Phys. C 14(1981)2705

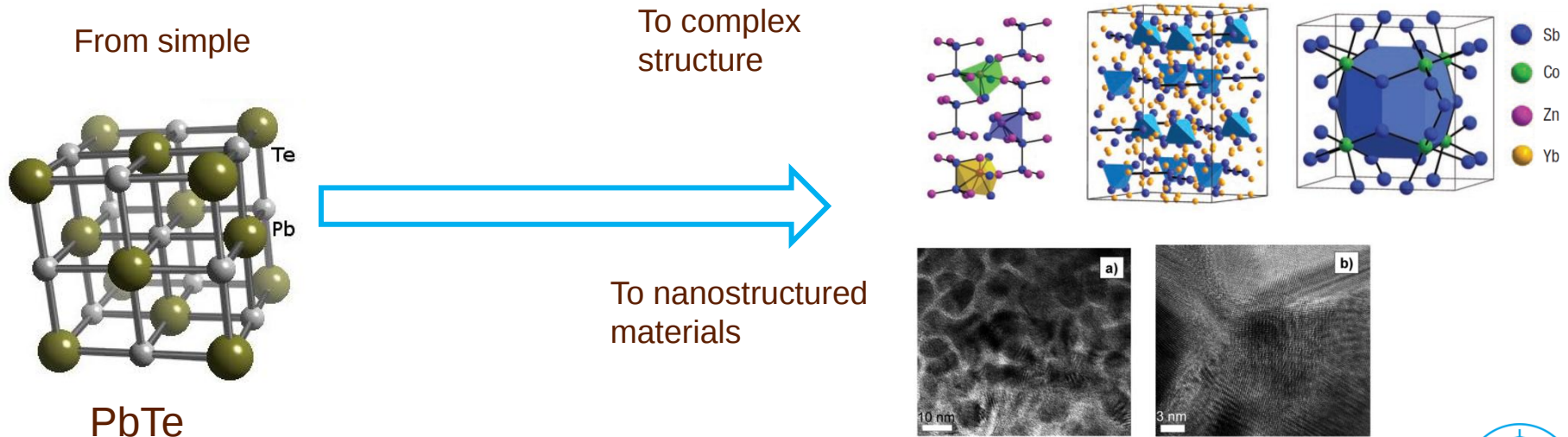
Thermoelectrics. Current trends

Commercial materials



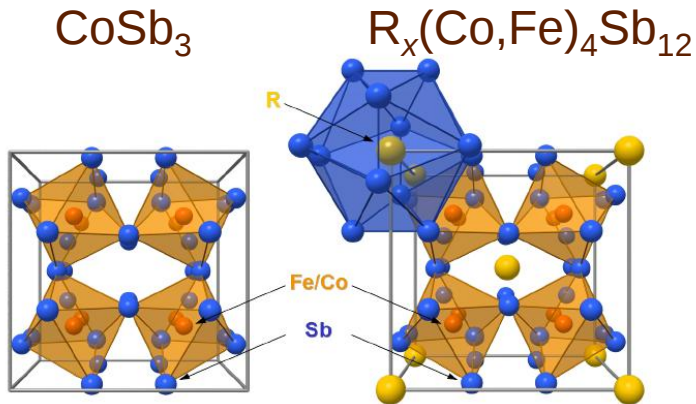
Snyder and Toberer,
Nature Mat. 7(2008)

Search for “phonon glass, electron crystal”



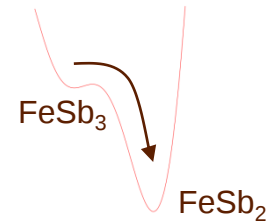
TEM images of a $\text{Si}_{80}\text{Ge}_{20}$ nanocomposite
Wang et al., *Appl. Phys. Lett.*, 93 (2008).

Thermoelectrics. Skutterudites



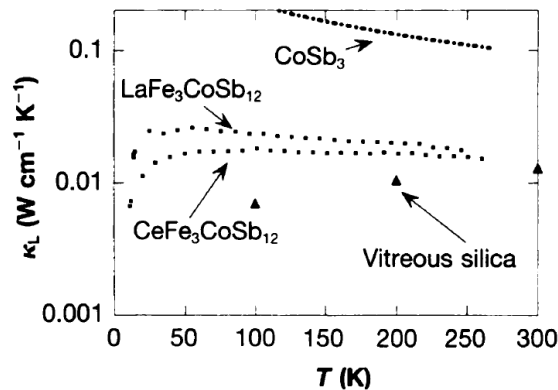
Is CoSb_3 a good reference for filled skutterudite lattice dynamics?

FeSb_3 is thermodynamically unstable.



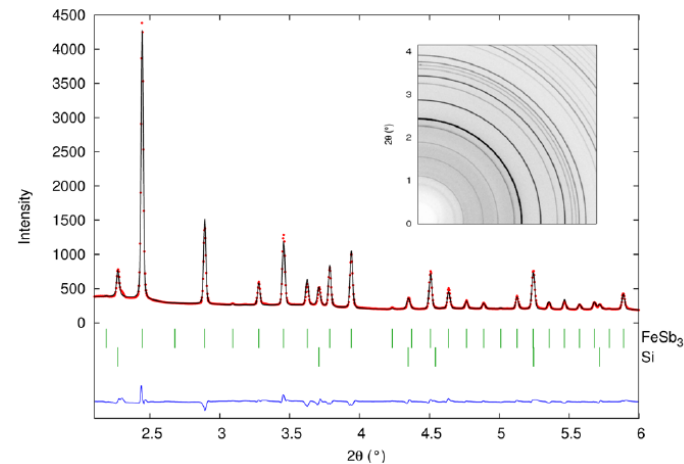
Synthesis of Metastable FeSb_3 using multilayer precursors
(Hornbostel et al., J. Am. Chem. Soc. 1997, 119, 2665)

Lattice thermal conductivity



Sales et al., Science, 272(1996)

→ 1-1.5 μm films of **FeSb_3**

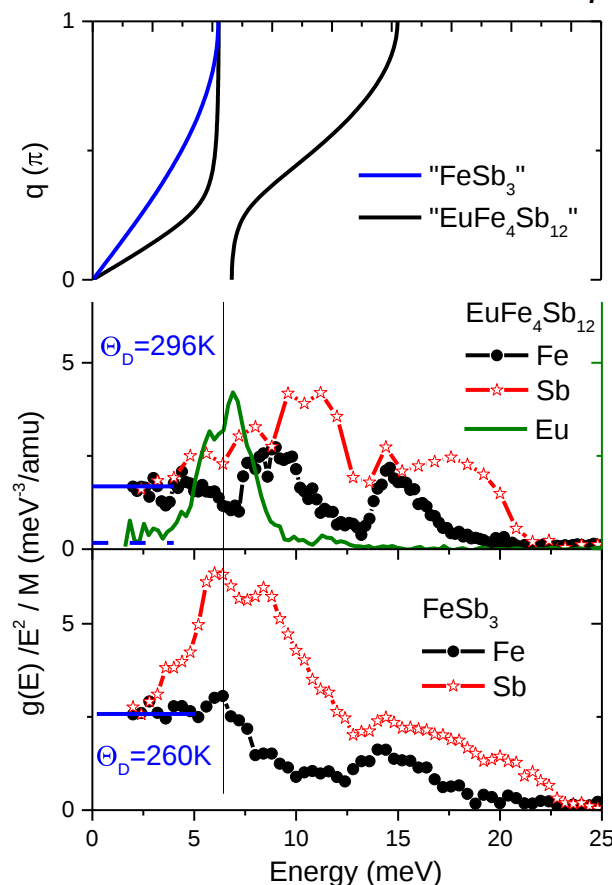
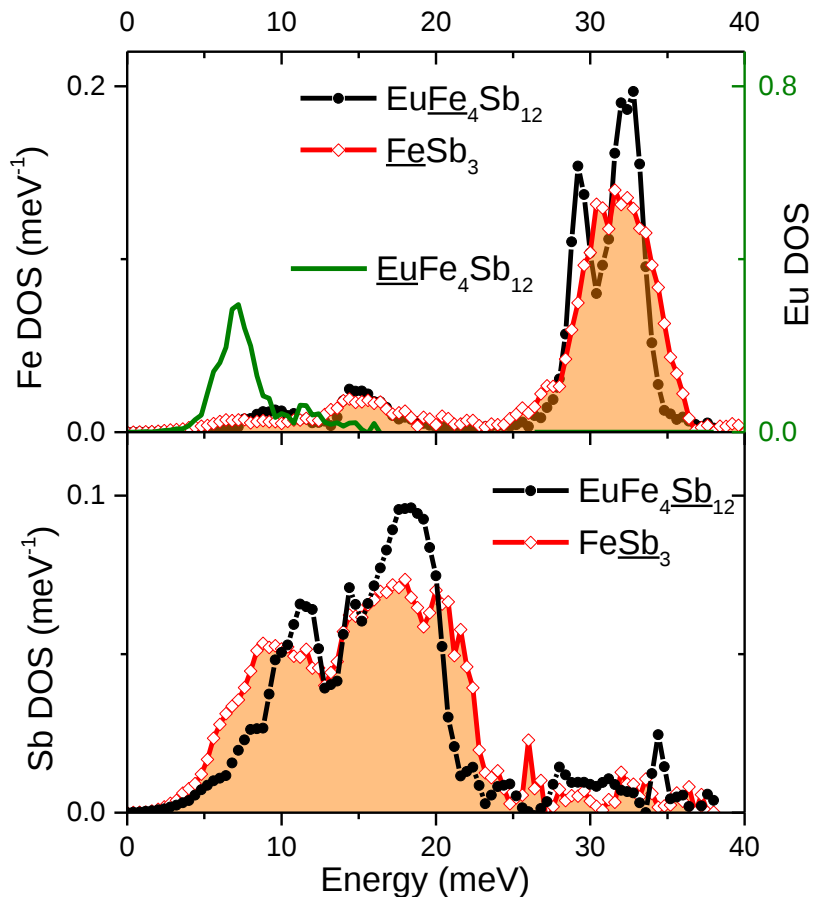
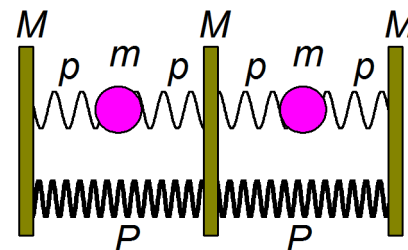


Möchel et al, Phys. Rev. B, 84(2011)064302



Phonons in skutterudites. $\text{EuFe}_4\text{Sb}_{12}$ vs FeSb_3

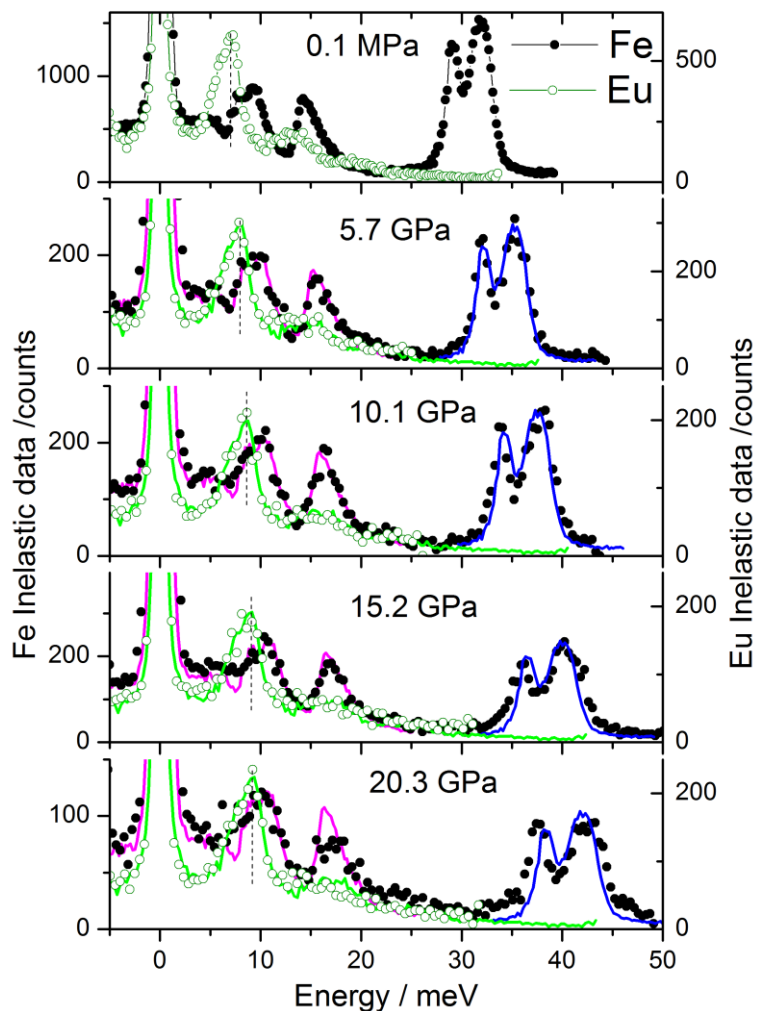
Nuclear Inelastic Scattering at
 ^{151}Eu , ^{57}Fe , ^{121}Sb nuclear resonances



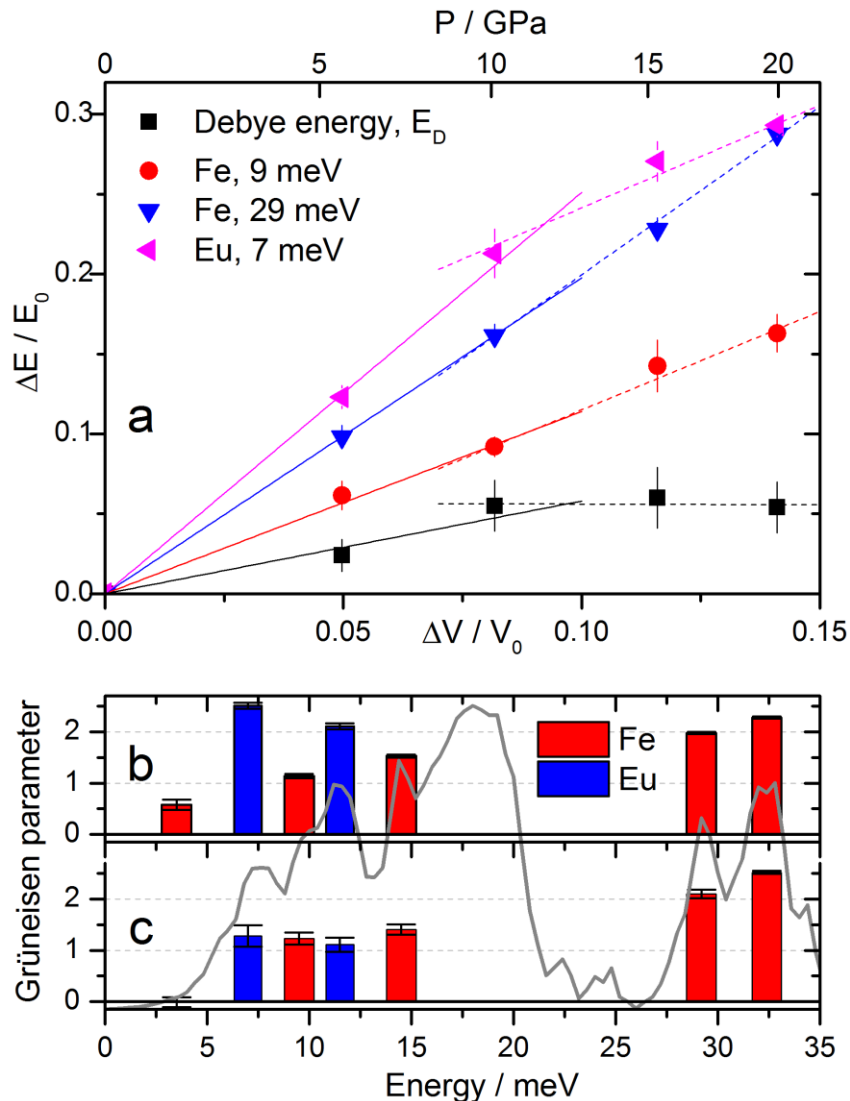
Möchel et al, Phys. Rev. B, 84(2011)064302
 I. Sergueev, in prep.

Phonons in skutterudites. High pressure study

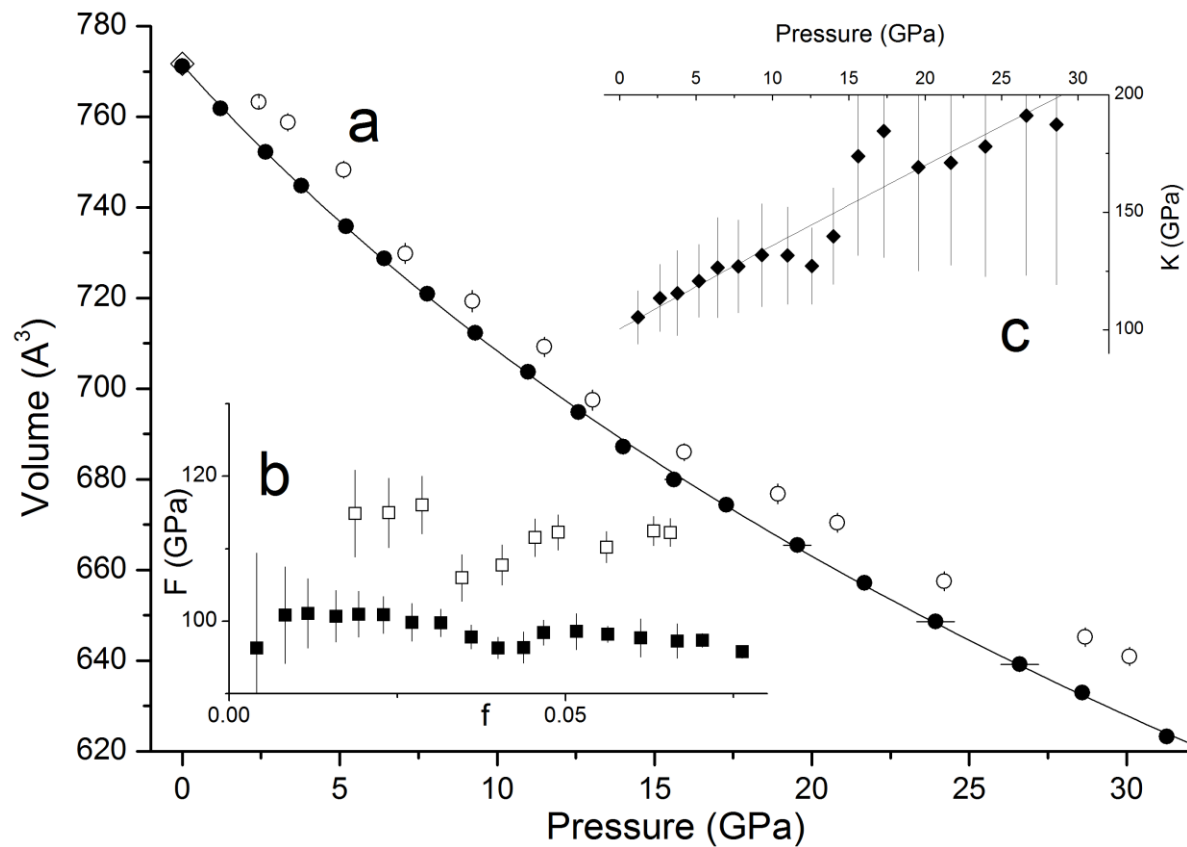
Experiment.



Grüneisen parameter



High pressure diffraction



Sapphire monochromator. Perspectives

Improvement of the sapphire quality

at 35 keV

thickness	ΔE / meV
1 mm	0.7
2 mm	0.3
5 mm	0.1

at 50 keV

thickness	Refl-ty / %
1 mm	2
2 mm	7
5 mm	30

Resolution to measure:

- phonon line broadening
- mean sound velocity

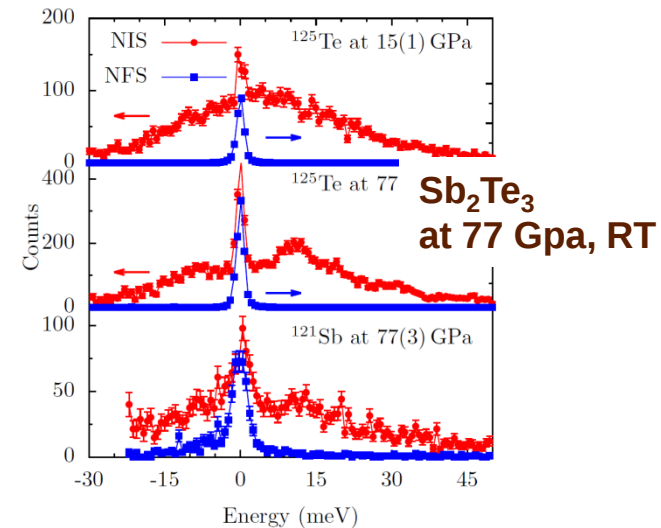
New elements:

^{183}W 46.5 keV
 ^{189}Os 36.2 keV
 ^{238}U 44.9 keV
 ^{240}Pu 42.8 keV

Apply for extreme conditions (high P):

Feasibility high pressure study:

R.Simon et al., Semicond. Sci. Technol. 00 (2014)



Requires setup to perform
low T/ high pressure
measurements

Sapphire ultraoptics for synchrotron radiation

Helmholtz-Russia Joint Research Group HRJRG-402

Partners:

Russia - RAS Shubnikov IC, Moscow

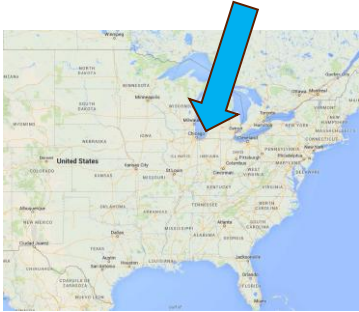
Germany – Forschungszentrum Jülich, DESY, ANKA



Acknowledgment

Argonne National Laboratory

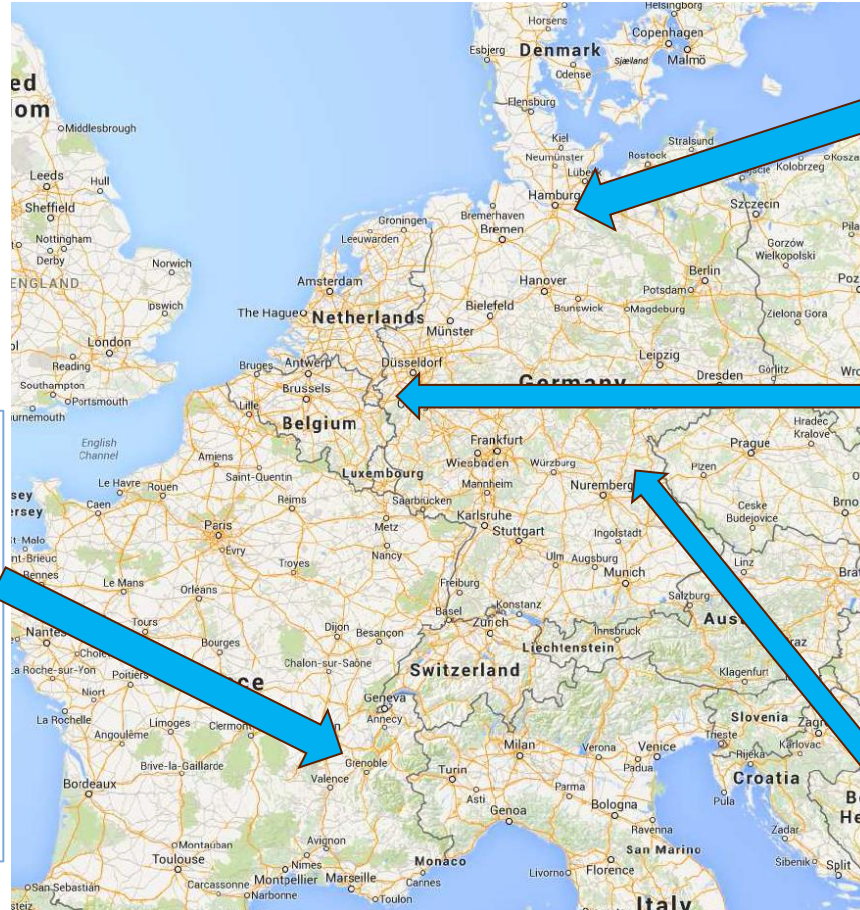
Yuri V. Shvyd'ko



ESRF

Nuclear Resonance Group

Rudolf Rüffer, Alexander Chumakov, Jean-Philippe Celse, Cornelius Strohm, Thanh Hai Deschaux-Beaume, Marcin Zajac, Vasily Potapkin, Dimitrios Bessas



DESY, PETRA III

P01

Hans-Christian Wille,
Pavel Alexeev

Forschungszentrum Jülich

JCNS-2

Raphael Hermann,
Anne Hounen, Benedict Klobes, Ronnie Simon, Atefeh Jafari

Bayerisches Geoinstitut

Leonid Dubrovinsky

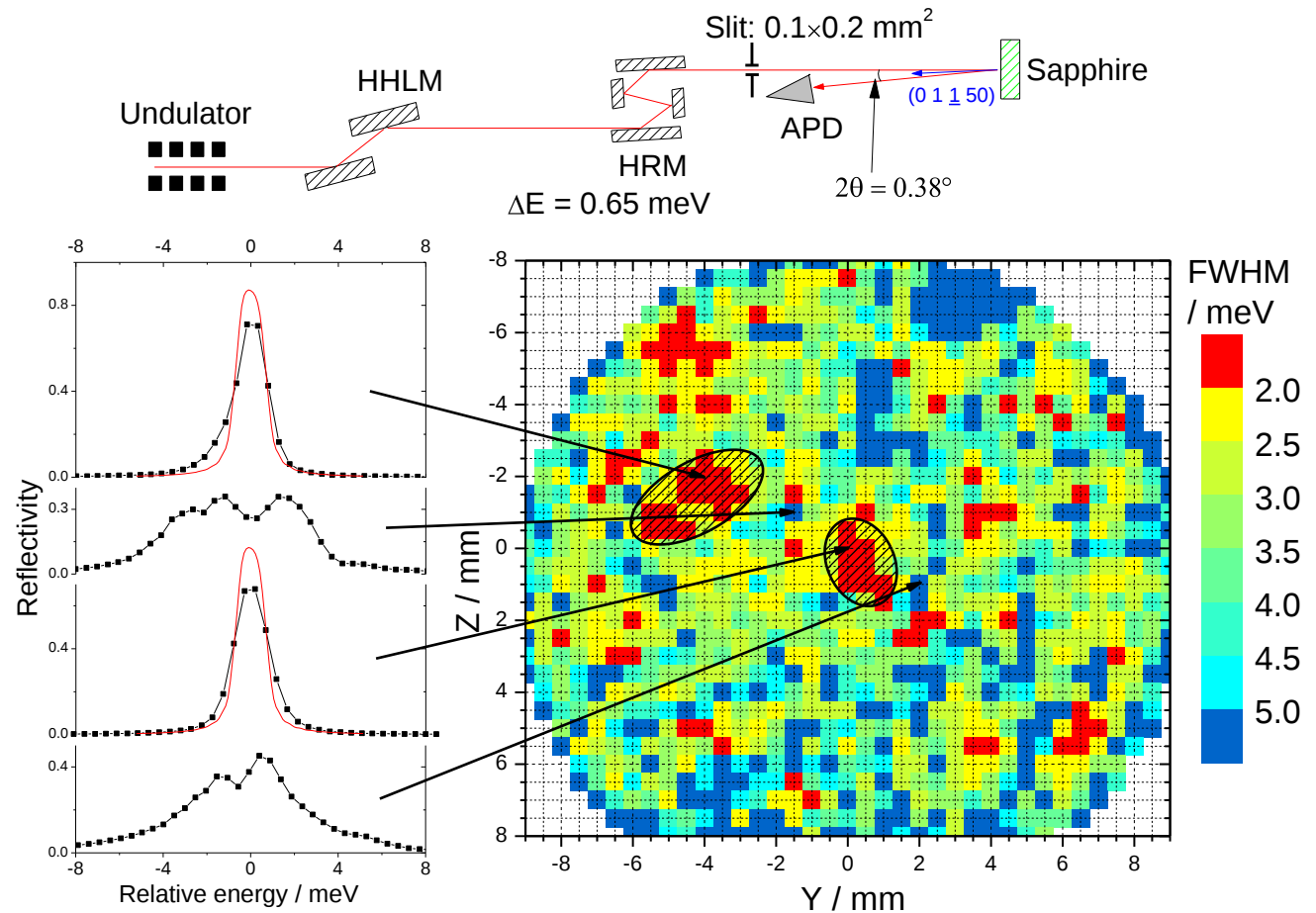


Спасибо
за

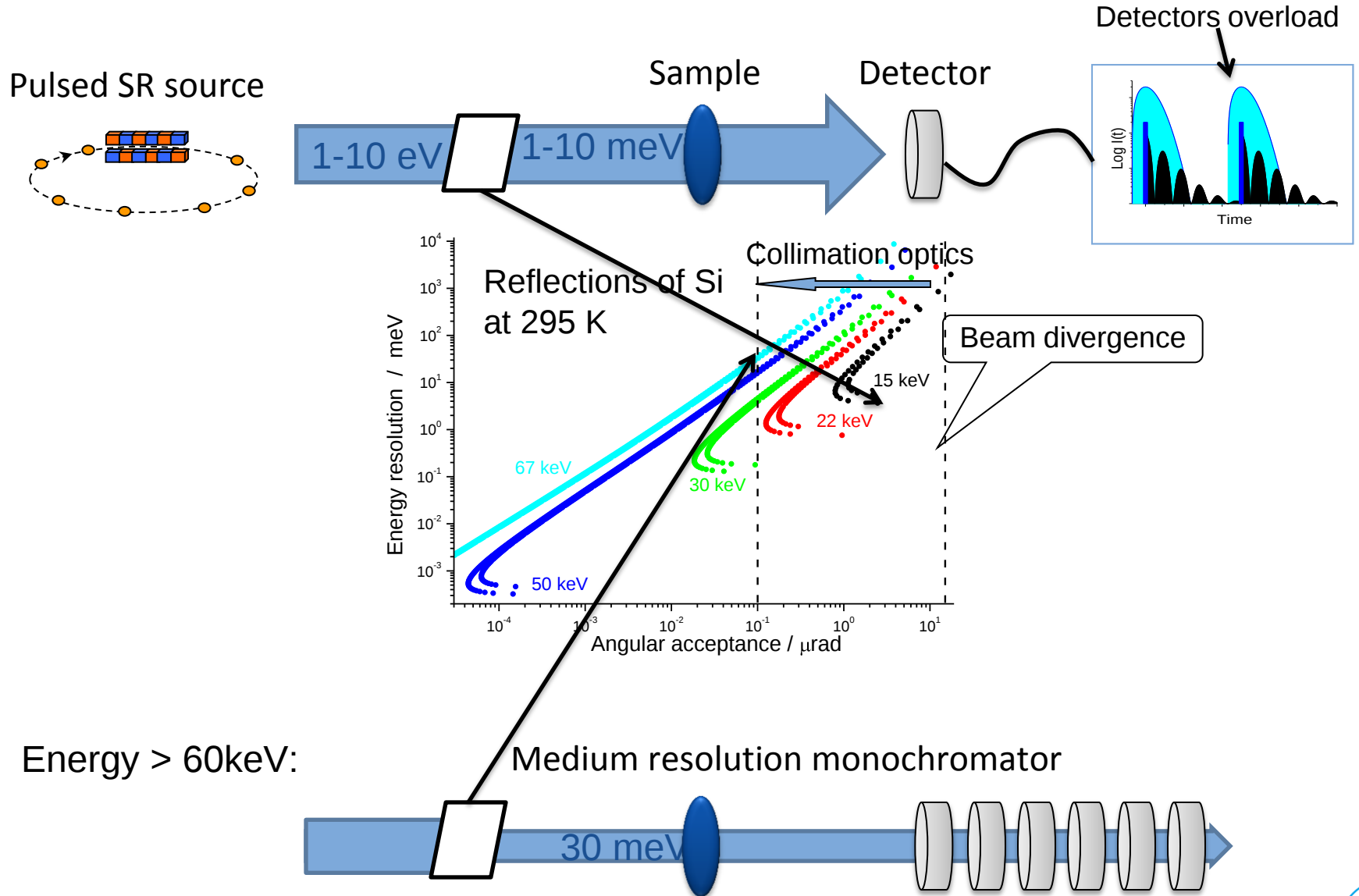
ВНИМАНИЕ



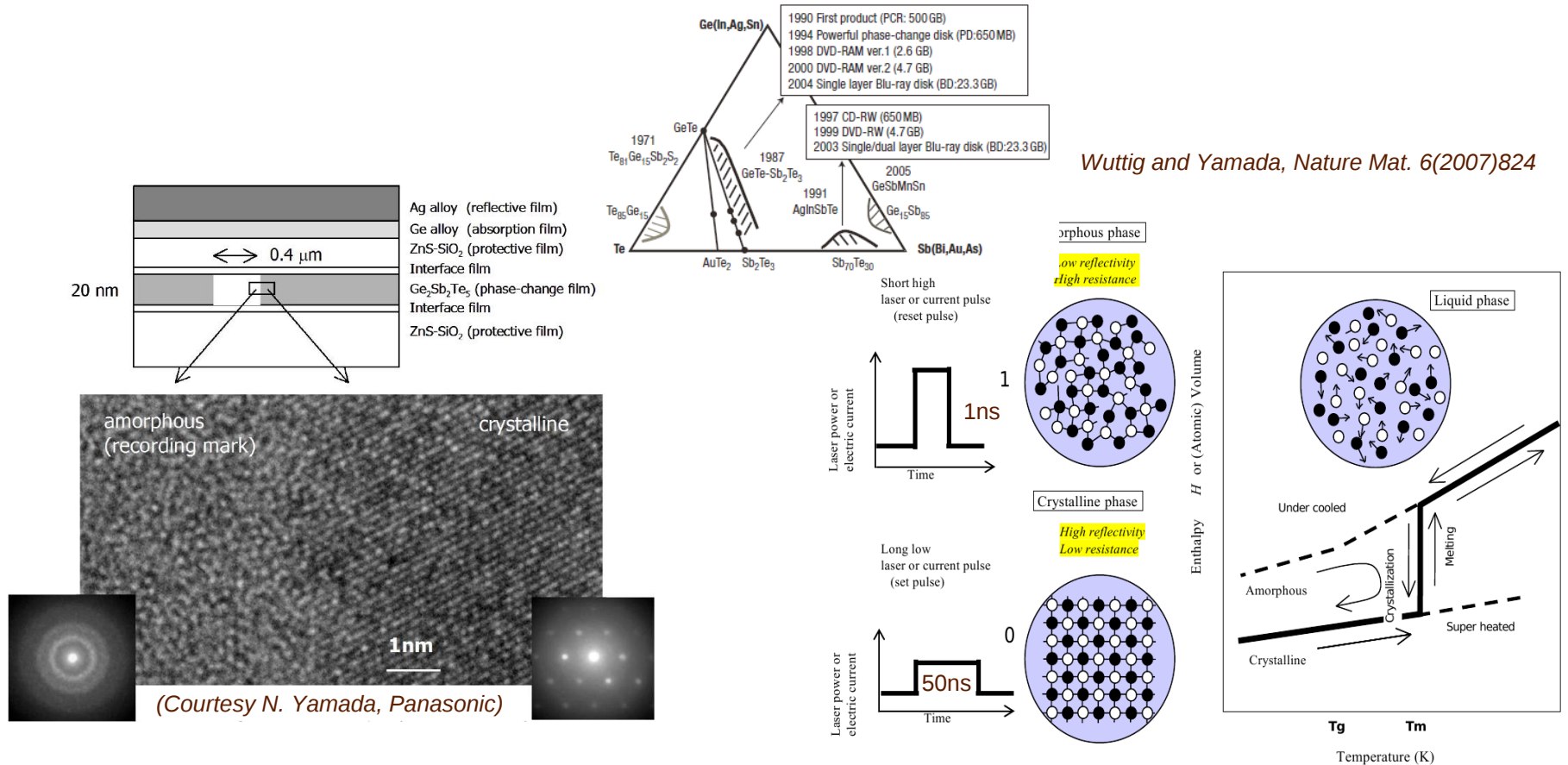
Quality of sapphire crystals



Time Mössbauer spectroscopy above 60 keV



Phase change materials for storage devices



Candidate for memory materials:

- Fast – ~10 ns
- Dense – $\varnothing \ll 50\text{nm}$
- Stable – several years /bit
- Long-lived – 10¹² cycles

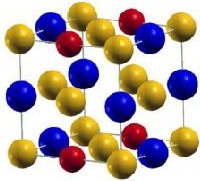
Today: optical contrast: storage media, DVD, Blu-Ray.

Future: resistivity contrast: fast non volatile memory!



Elastic properties of $\text{Ge}_2\text{Sb}_2\text{Te}_4$

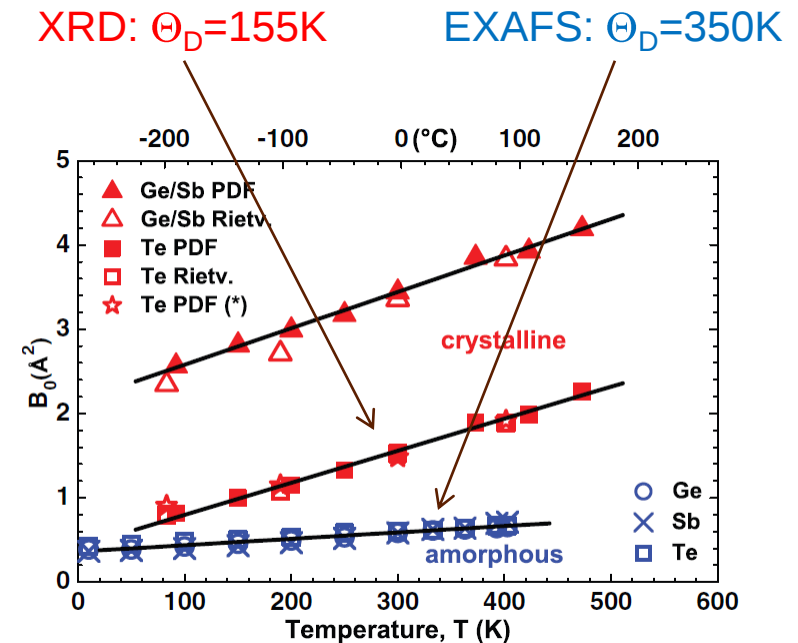
Hardening upon crystallization



	Amorphous	Crystalline
C_{11}	31 GPa	48 GPa
V_L	2280 m/s	2770 m/s
C_{44}	10 GPa	16 GPa
V_T	1300 m/s	1600 m/s
V_{Debye}	1440 m/s	1770 m/s

Blachowicz et al., *J. Appl. Phys.* **102**, 093519 (2007)

Softening upon crystallization



Matsunaga et al., *Adv. Funct. Mater.* **21**(2011)2232

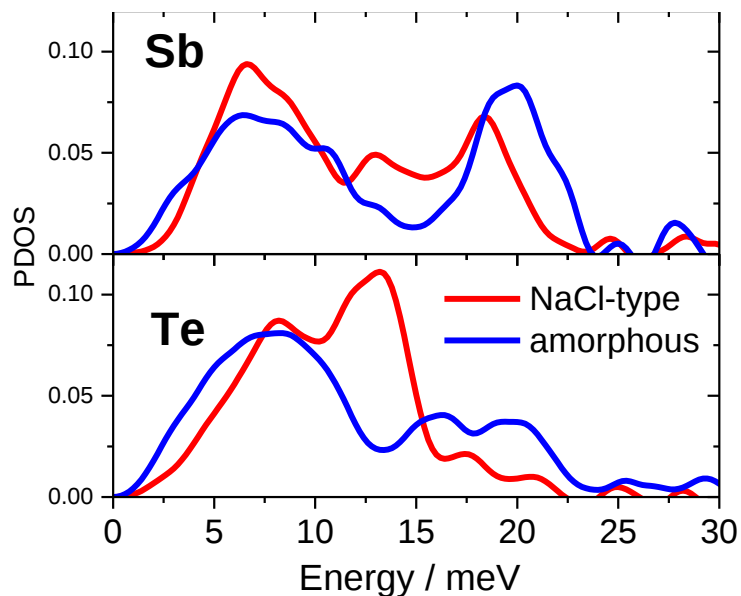
How we can reconcile this?



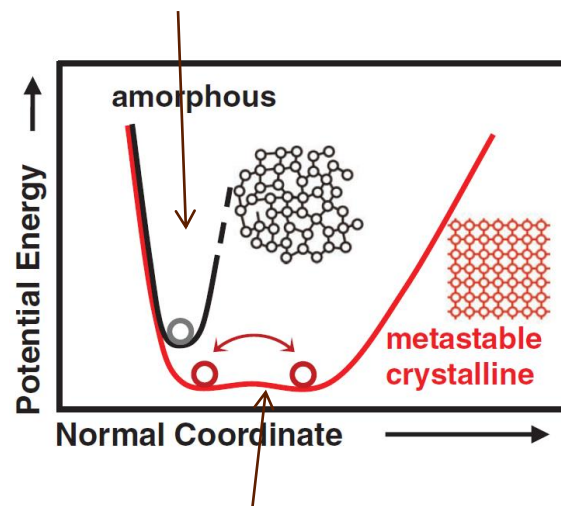
Amorphous and crystalline state of $\text{Ge}_2\text{Sb}_2\text{Te}_4$

Matsunaga et al., Adv. Funct. Mat., 21(2011)2232

Sample: $\sim 1 \mu\text{m}$ film deposited on Al, $T=20 \text{ K}$



Amorphous:
Strong covalent bonding



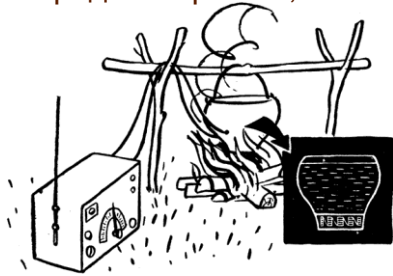
Crystalline:
Weak "resonance" bonding

	Sb		Te	
	B, Å ²	F, N/m	B, Å ²	F, N/m
Amorphous	0.184(2)	97(4)	0.198(2)	84(4)
NaCl	0.166(2)	72(4)	0.160(2)	68(4)

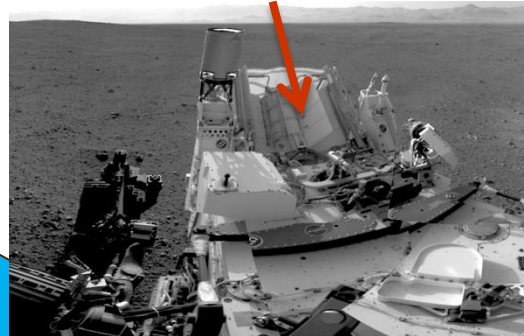
Combination of NIS on ^{125}Te and ^{121}Sb and INS (IXS) $\Rightarrow$ Ge PDOS

Thermoelectrics. Applications

ТГ-1, «партизанский котелок»
Юрий Маслаковец,
Ленинградский физтех, 1943



Radioisotope Thermoelectric Generator,
Mars Science Laboratory
<https://solarsystem.nasa.gov/rps/rtg.cfm>



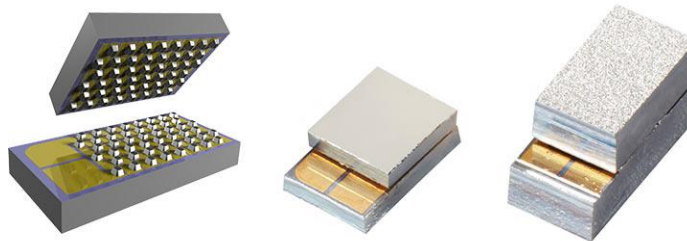
Thermoelectric stations for gas pipe lines
<http://www.telgen.ru/>



Recovery of thermal waste
BMW TEG



Peltier Coolers
<http://www.micropelt.com>



<http://biolitestove.com/products/campstove/>

