Neutron spin-echo

on triple-axis spectrometers

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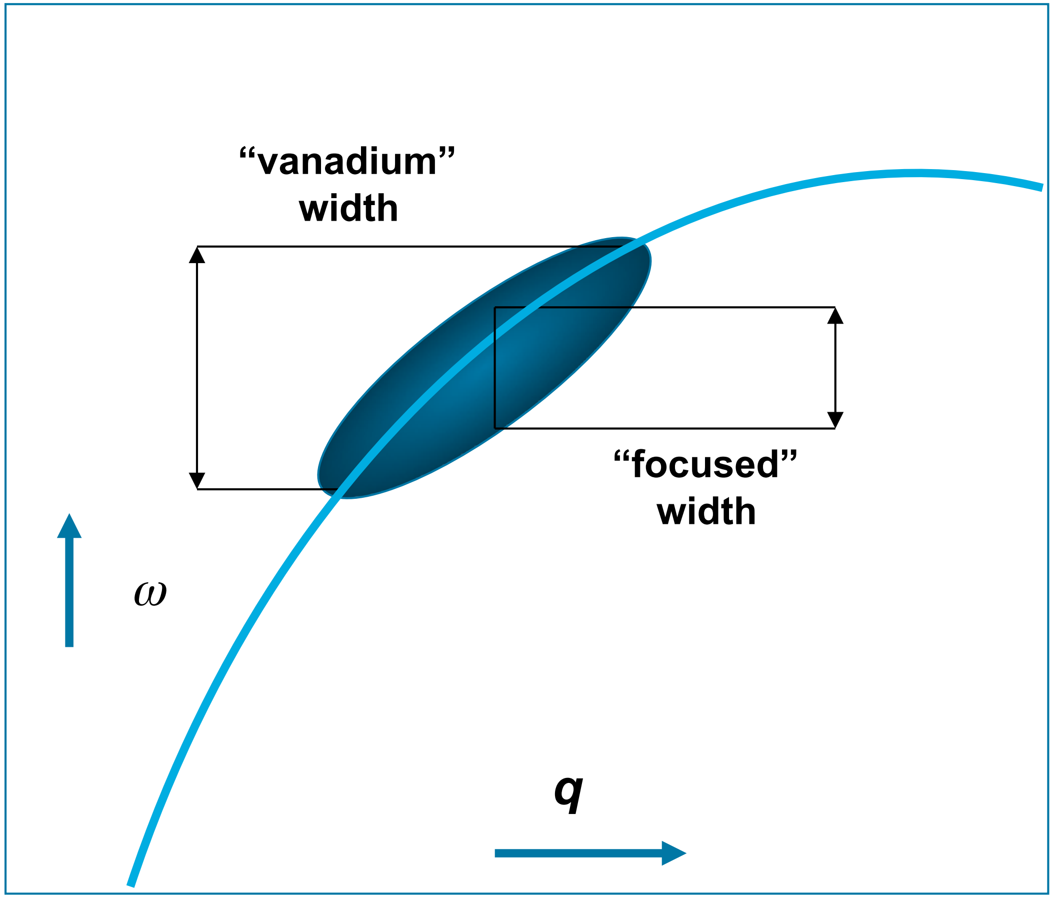
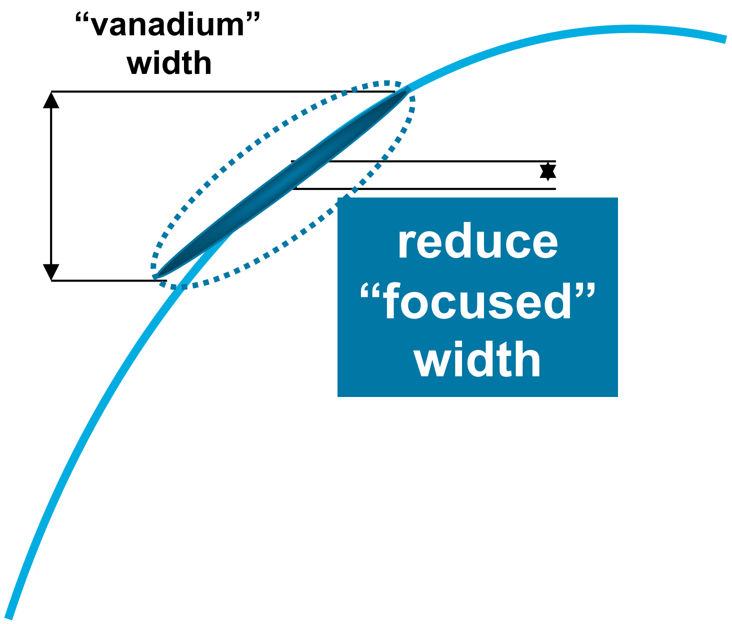
Neutron TAS

Neutron **T**hree-**A**xis **S**pectrometers:

- access to large Q,ω range
- energy resolution $\Delta E/E \approx$ 5-10%
- efficient for $\omega(q)$
- lacking resolution for $\Gamma(q)$

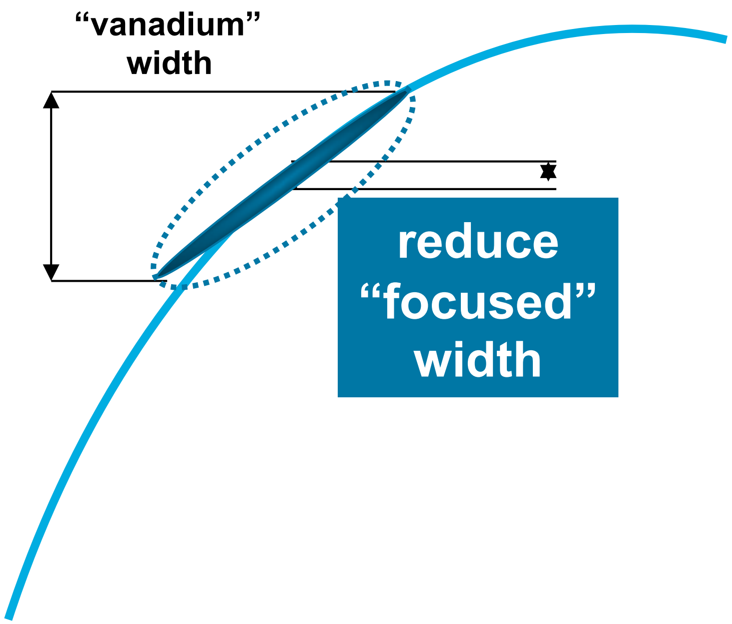


TAS resolution



normal TAS setup with perfect monochromator & analyzer crystals (Si, Ge)

TAS resolution



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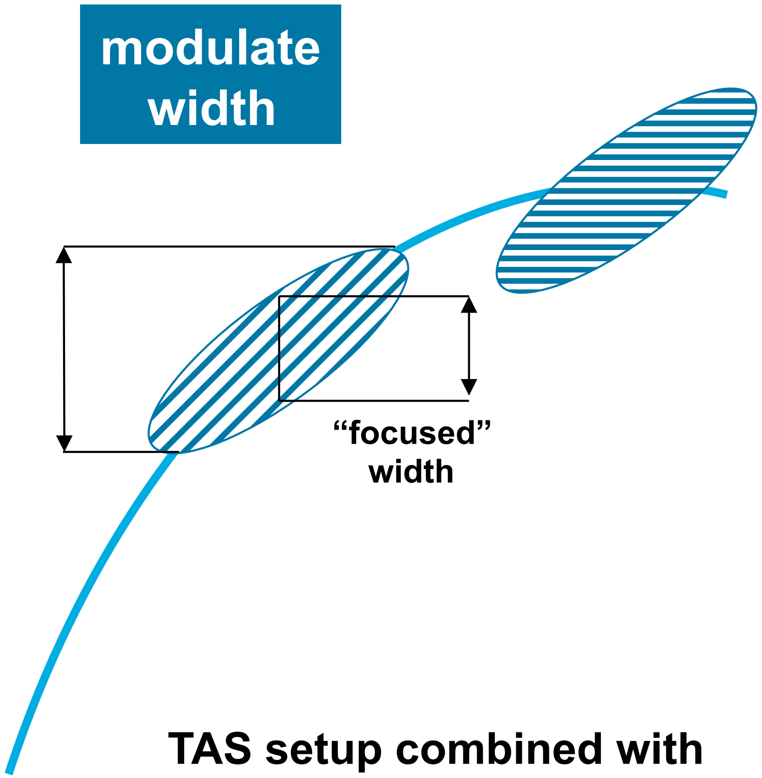


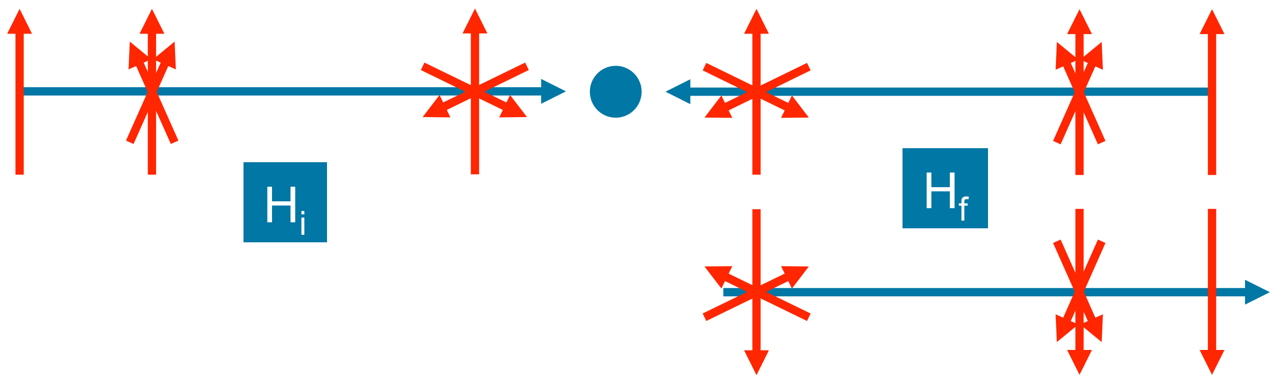
TAS setup combined with
spin-echo
(TOF Fourier technique)

Spin-echo principle

Larmor precesssion:

$$\phi = \gamma_L \frac{H\,l}{v_n}$$

γ_L = 18324 rad $Oe^{-1}s^{-1}$



Spin-echo condition:

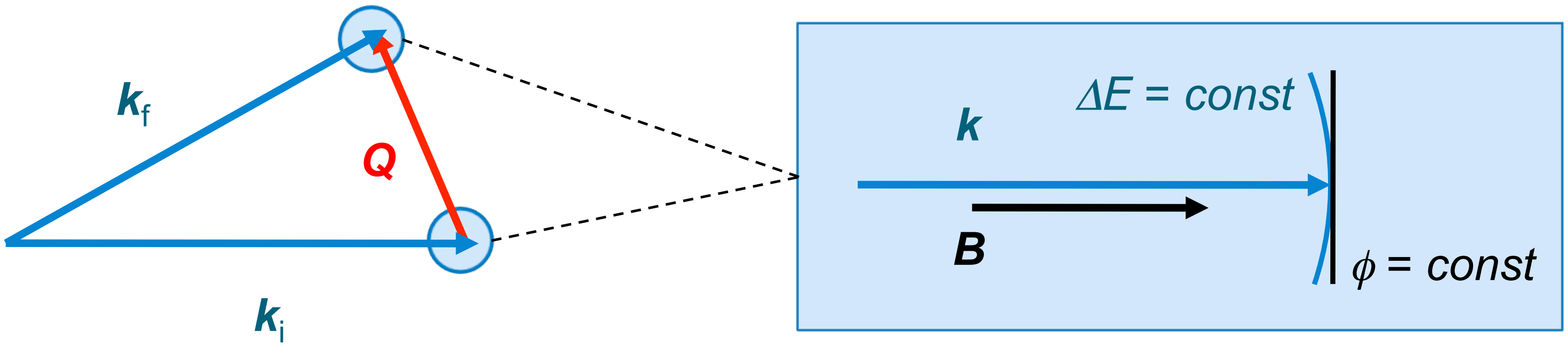
$$\Delta\phi = \phi_f - \phi_i = \gamma_L \frac{H_f l_f - H_i l_i}{v_n} = 0$$

stationary phase

ϕ = *const* surfaces perpendicular to $\boldsymbol{k}_i$, $\boldsymbol{k}_f$

TAS + spin-echo

The simple case – combining the traditional QENS spin-echo & TAS



overall spin-echo phase is only stationary for $\Delta E = const$, i.e. $\omega(\boldsymbol{q}) = const$

TAS + spin-echo

General SE condition:

$$\left.\frac{\partial \Delta\phi}{\partial k_{i,f}}\right|_{\Delta E = const.} = 0 \quad \longrightarrow \quad \frac{H_f l_f}{H_i l_i} = \left(\frac{k_f}{k_i}\right)^3$$

optimum field ratio

stationary phase

ϕ = *const* surfaces perpendicular to $\boldsymbol{k}_i$, $\boldsymbol{k}_f$

$$\Delta\phi = \tau_F \frac{\Delta E}{\hbar}$$

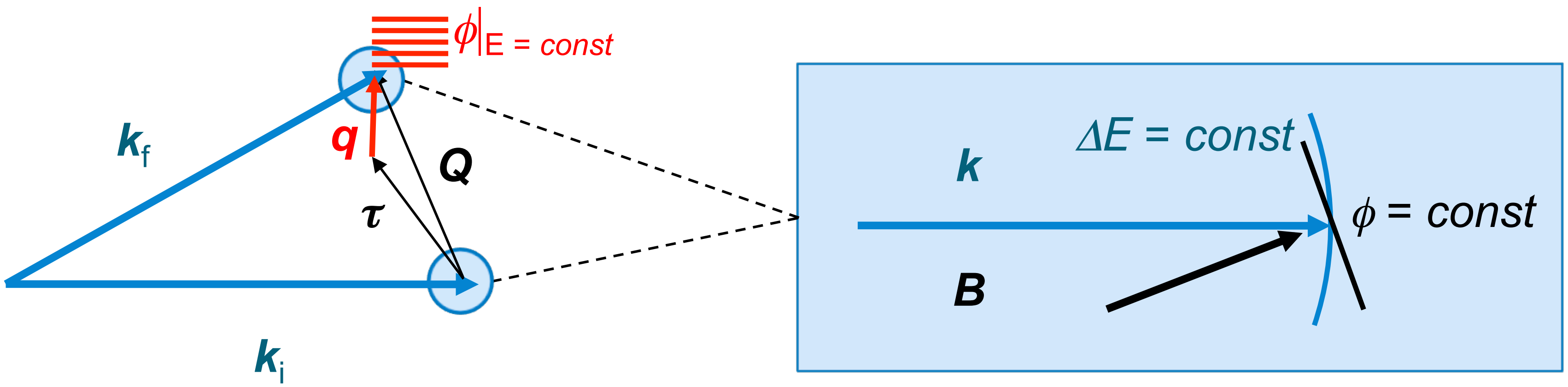
phase shift

Fourier time τ_f

$$\boxed{\tau_F = \gamma_L \frac{2m_n^2}{\hbar^2} \frac{H_f l_f}{k_f^3}}$$

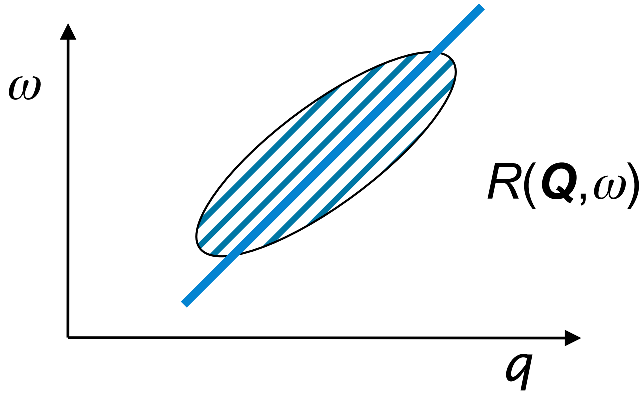
TAS + spin-echo

The sophisticated case – matching the slope of a dispersion $\omega(\boldsymbol{q})$



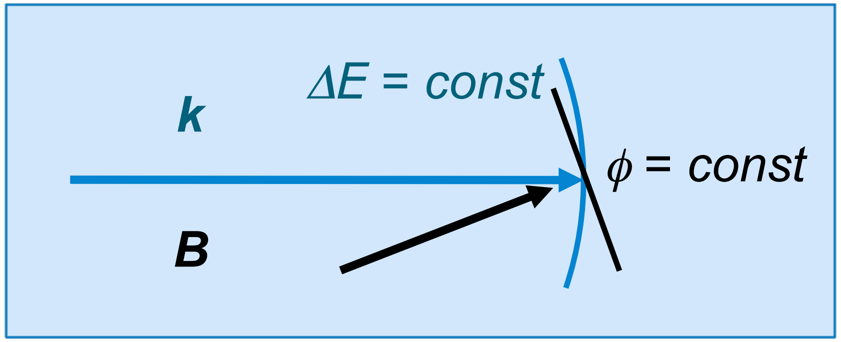
- each point of the resolution ellipsoid corresponds to a combination $\boldsymbol{k}_i$, $\boldsymbol{k}_f$
- we need to manipulate the phase fields around $\boldsymbol{k}_i$, $\boldsymbol{k}_f$ and project them onto $R(\boldsymbol{Q},\omega)$

→ rotate $\boldsymbol{B}$ with respect to $\boldsymbol{k}_i$, $\boldsymbol{k}_f$



TAS + spin-echo

The sophisticated case – matching the slope of a dispersion $\omega(\boldsymbol{q})$



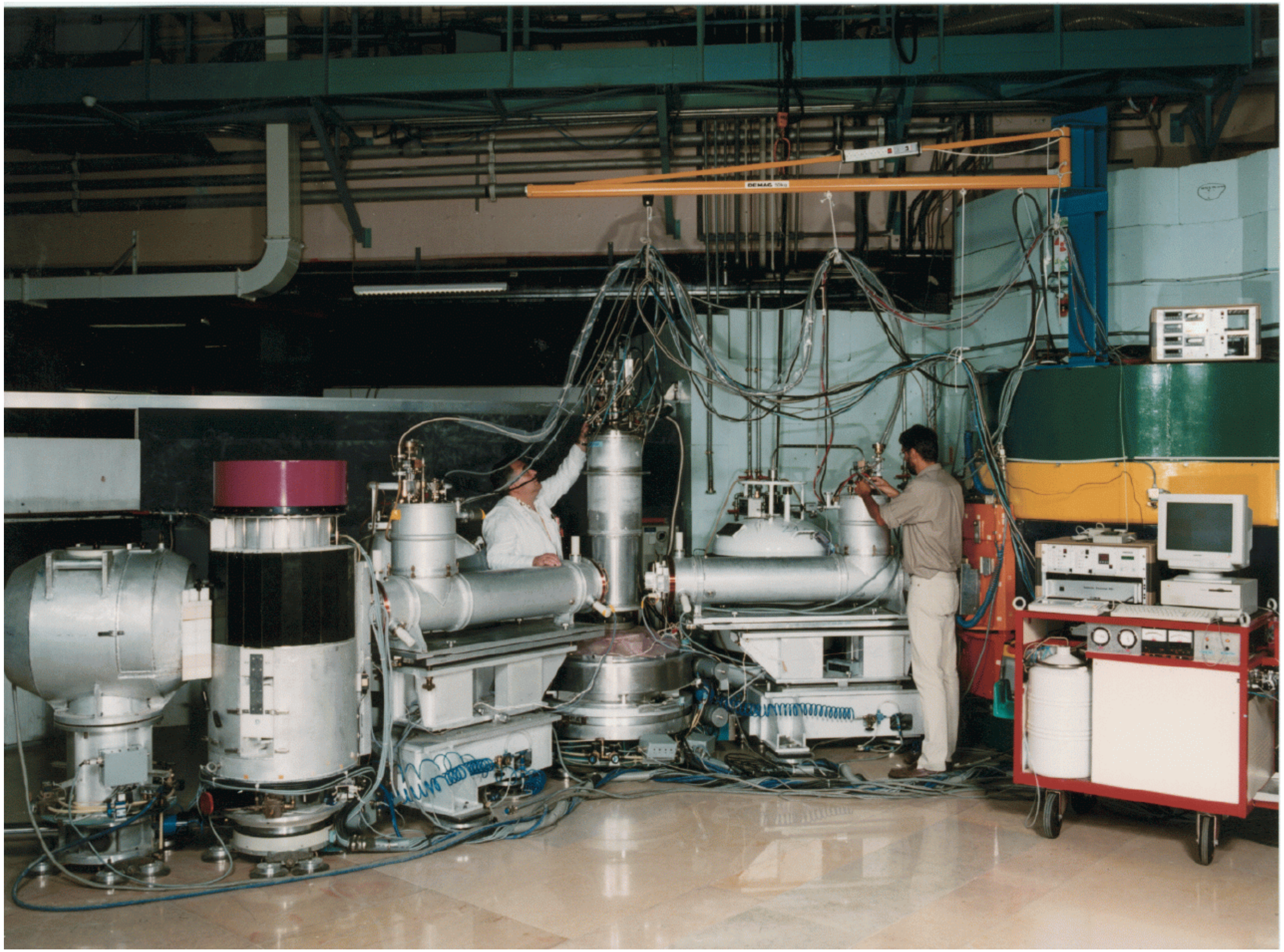
we need to manipulate the phase fields around $\boldsymbol{k}_i$, $\boldsymbol{k}_f$ and project them onto $R(\boldsymbol{Q},\omega)$

- rotate $\boldsymbol{B}$ with respect to $\boldsymbol{k}_i$, $\boldsymbol{k}_f$
- incline the field boundary with respect to $\boldsymbol{k}_i$, $\boldsymbol{k}_f$

difficult with solenoids ...

... look for a more flexible technique!

IN20 – TASSE (1998 – 2015)



IN20 TASSE setup

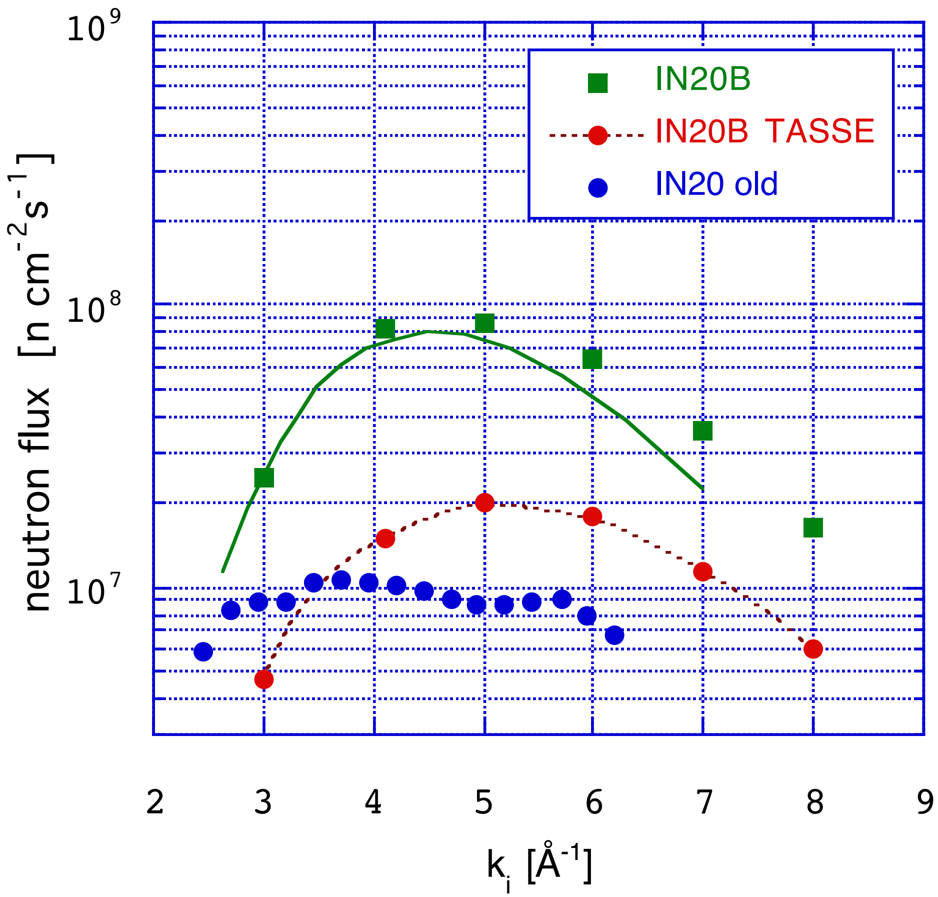
- **Q-range: 1 - 7 Å-1**
- **ΔE range (TAS): 0 - 40 meV**

- **OSF superconducting solenoids**
- **max. field integral 1 Tm**
- **Fourier times:**
  - **k_f = 2.662 Å-1 0.015 - 1.5 ns**
  - **k_f = 4.1 Å^{-1} 0.002 - 0.4 ns**

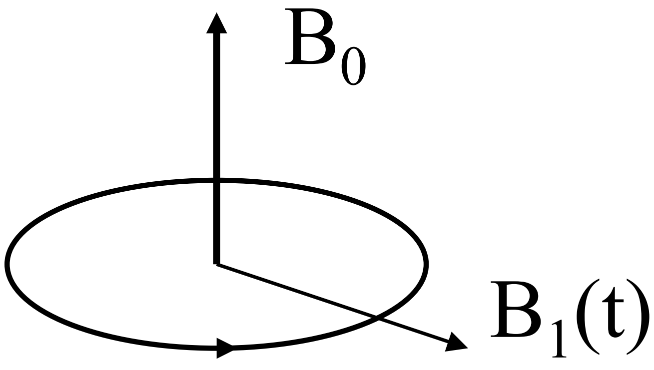
- **horizontally focussing Heusler monochromator & analyzer**

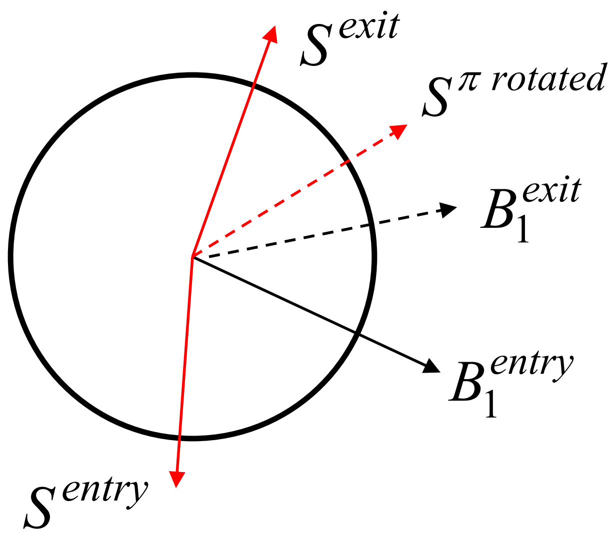
- **k_f = 4.1 Å^{-1}, PG filter**

- **^{74}Ge (96.8%) single crystal; volume 7 cm^3**



Neutron resonant spin echo (NRSE)



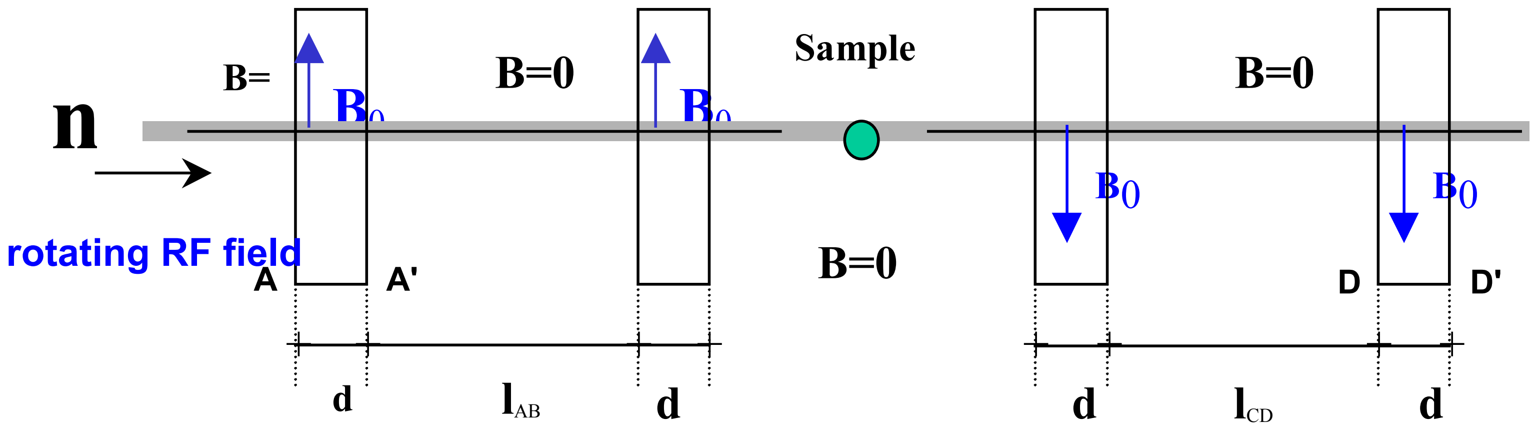


- fixed guide field $B_0 \approx 100$ G
- rotating RF field $B_1 \approx 1$ G with $\omega = \omega_L$
- setup tuned for a π-flip

$$\begin{aligned}\phi_{neutron}^{exit} &= \phi_{RF}^{exit} + (\phi_{RF}^{entry} - \phi_{neutron}^{entry}) \\ &= 2\phi_{RF}^{entry} - \phi_{neutron}^{entry} + \omega_0 d / v\end{aligned}$$

courtesy of R. Pynn, Indiana University

Neutron resonant spin echo (NRSE)



	Time t	Phase field B_r	neutron Spin phase S
A	t_A	ωt_A	0
A’	$t_{A'} = t_A + \frac{d}{v}$	$\omega t_{A'}$	$2\omega t_A + \omega \frac{d}{v}$
B	$t_B = t_A + \frac{l_{AB}+d}{v}$	ωt_B	$2\omega t_A + \omega \frac{d}{v}$
B’	$t_{B'} = t_A + \frac{l_{AB}+2d}{v}$	$\omega t_{B'}$	$2\omega \frac{l_{AB}+d}{v}$
C	t_C	$-\omega t_C$	$2\omega \frac{l_{AB}+d}{v}$
C’	$t_{C'} = t_C + \frac{d}{v}$	$-\omega t_{C'}$	$-\omega \frac{d}{v'} - 2\omega t_C - 2\omega \frac{l_{AB}+d}{v}$
D	$t_D = t_C + \frac{l_{CD}+d}{v'}$	$-\omega t_D$	$-\omega \frac{d}{v'} - 2\omega t_C - 2\omega \frac{l_{AB}+d}{v}$
D’	$t_{D'} = t_C + \frac{l_{CD}+2d}{v'}$	$-\omega t_{D'}$	$2\omega(\frac{l_{AB}+d}{v} - \frac{l_{CD}+d}{v'})$

R. Gähler, R. Golub, T. Keller, Physica B 180&181 (1992) 899

spin-echo condition

$(l_{AB} + d)/v_n = (l_{CD} + d)/v'_n$

courtesy of R. Pynn, Indiana University

Neutron resonant spin echo (NRSE)

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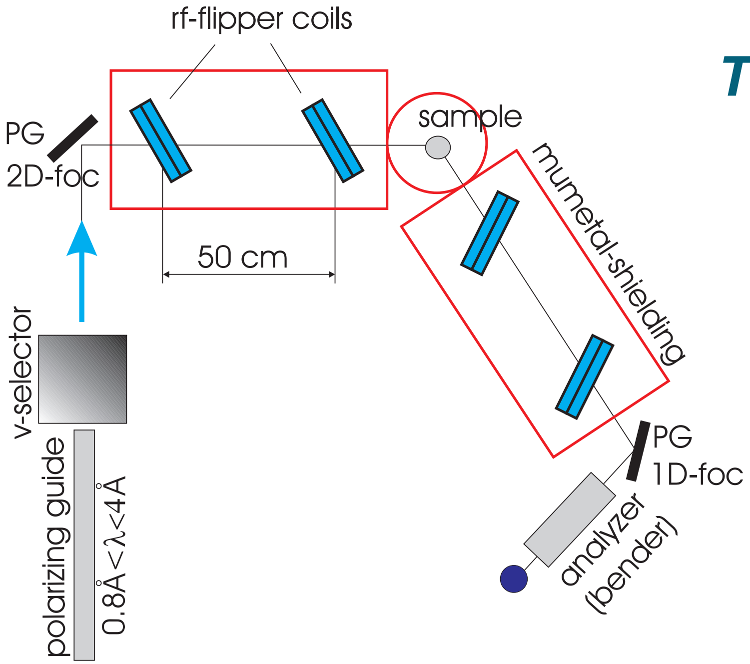
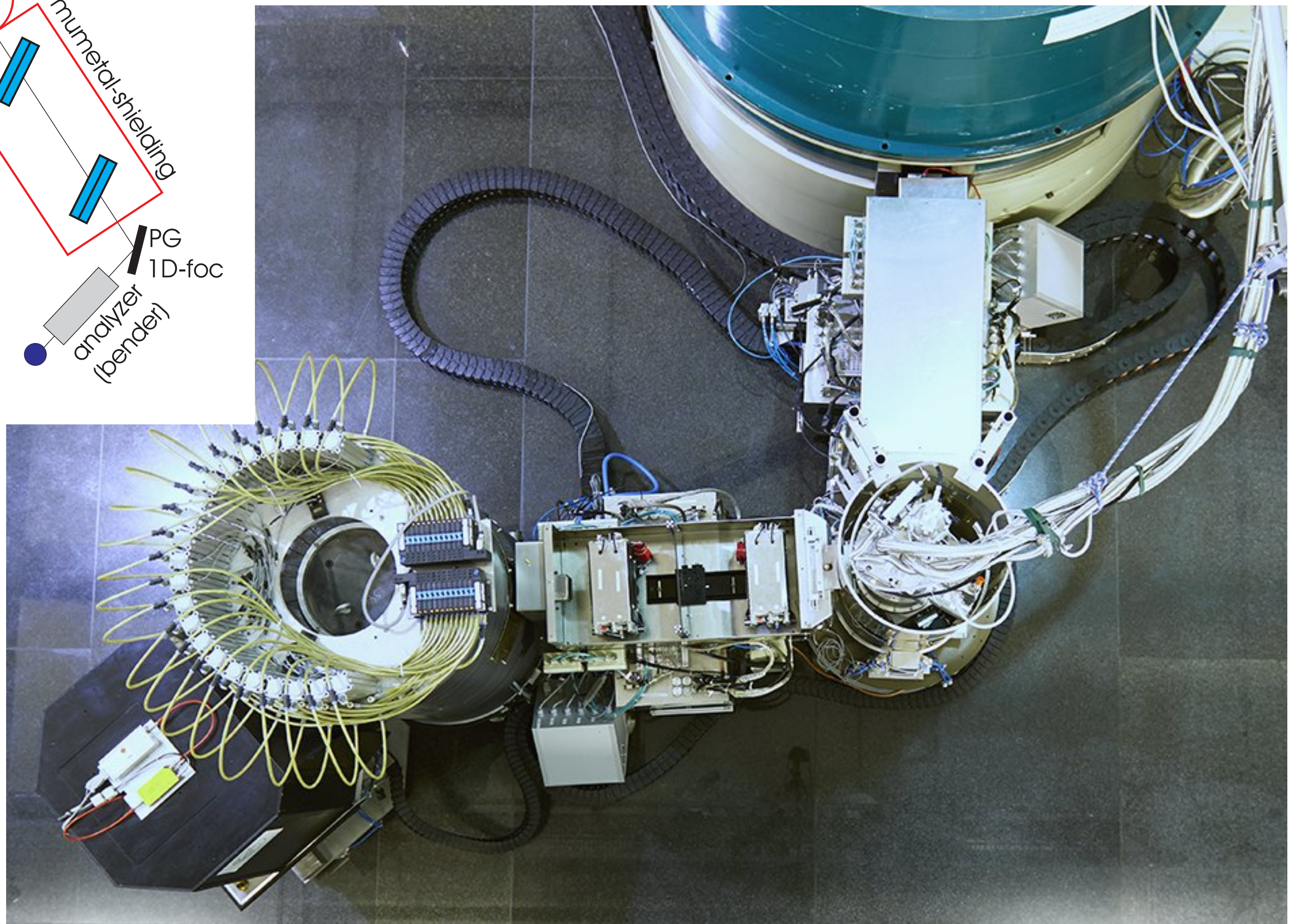
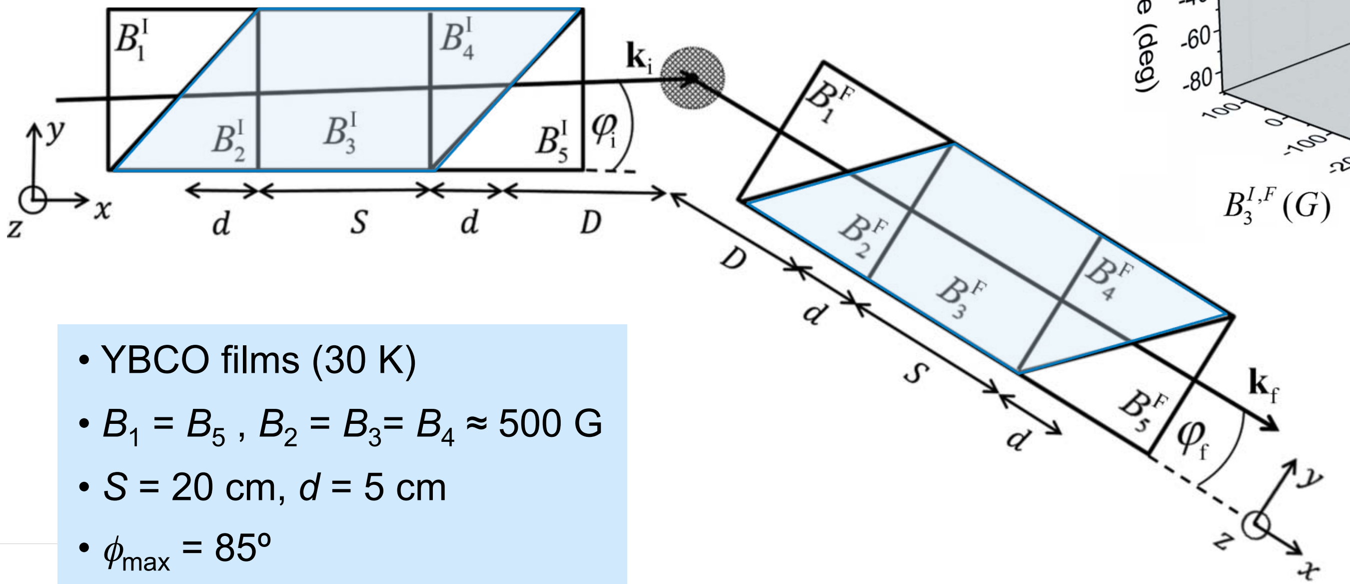


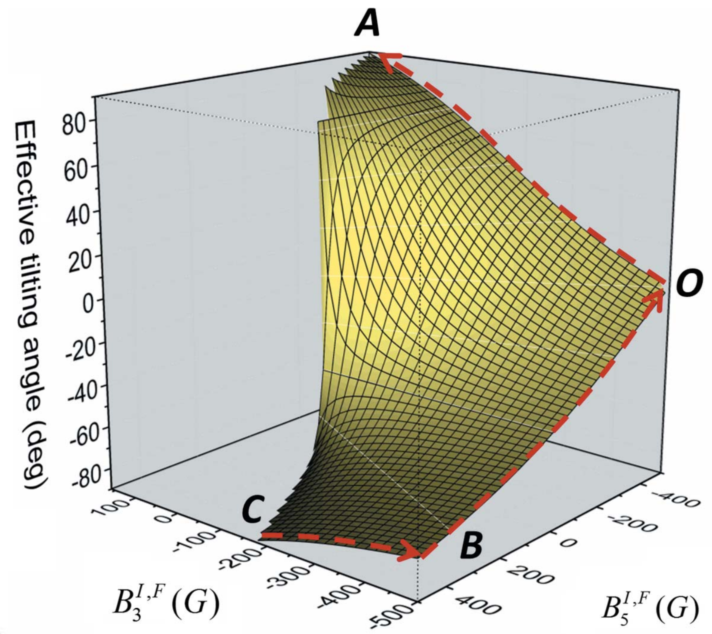
TRISP at FRM-II Munich



Tilted fields

Wollaston prisms

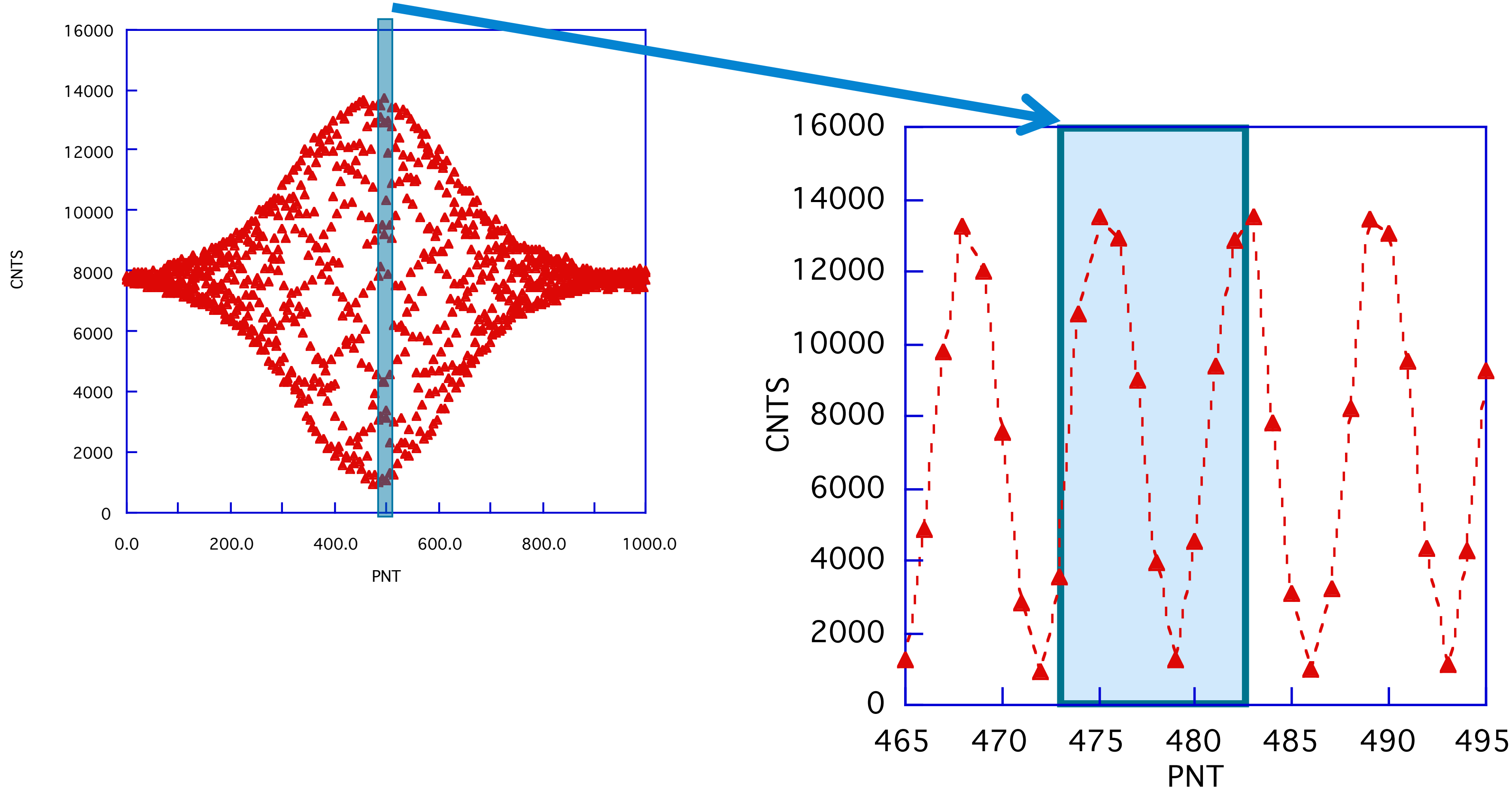




- YBCO films (30 K)
- $B_1 = B_5$, $B_2 = B_3 = B_4 \approx 500$ G
- S = 20 cm, d = 5 cm
- ϕ_{max} = 85º
- $\tau_{max} \approx 200$ ps

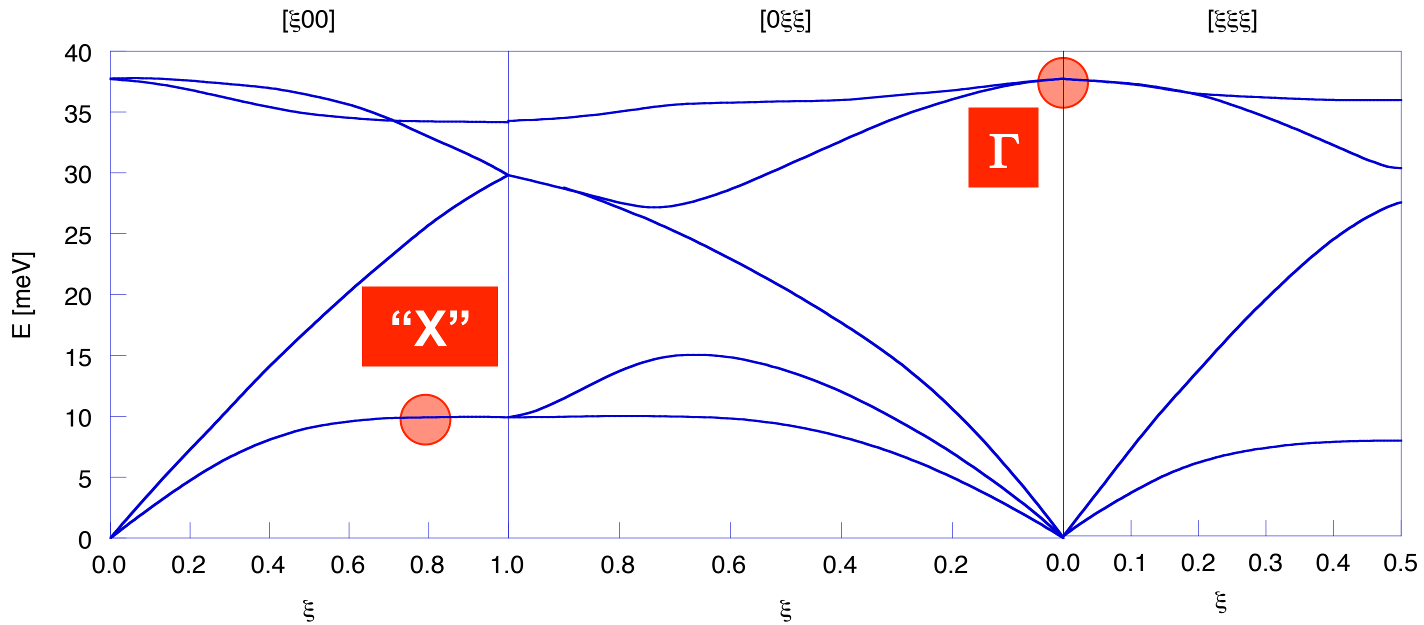
Li F., Pynn R., J.Appl.Cryst. 47 (2014) 1849

Spin-echo pattern



Ge lattice dynamics

Phonon dispersion



- group IV semiconductor
- diamond structure (2 atoms/unit cell)
- nontrivial lattice dynamics
- negative volume expansion

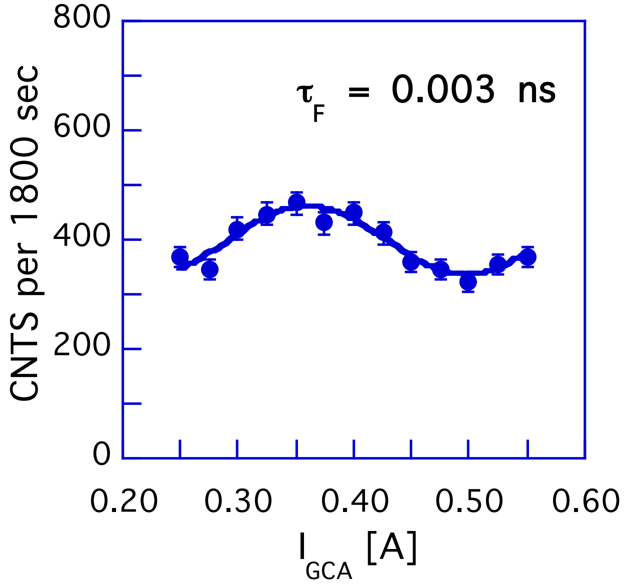
•disorder effects:

✓Ge isotopes (M. Cardona)

✓Si-Ge alloys (E. Courtens)

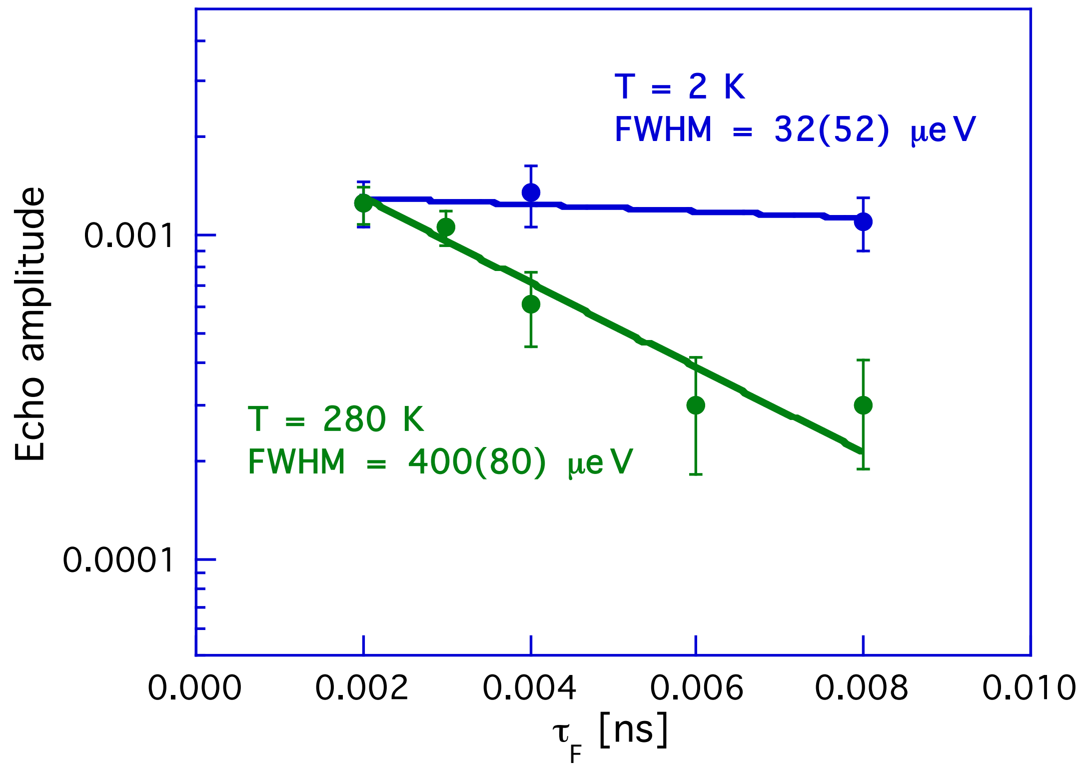
- large perfect crystals available

^{74}Ge: Γ-point phonon width

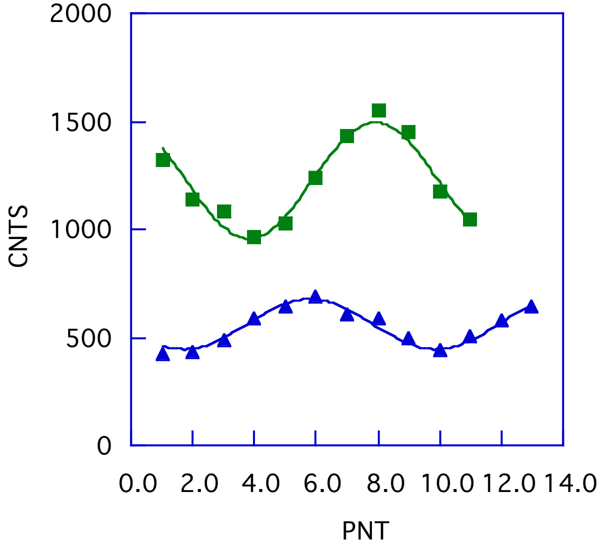


$\boldsymbol{Q}$ = [0 0 6]
ΔE = 37.3 meV

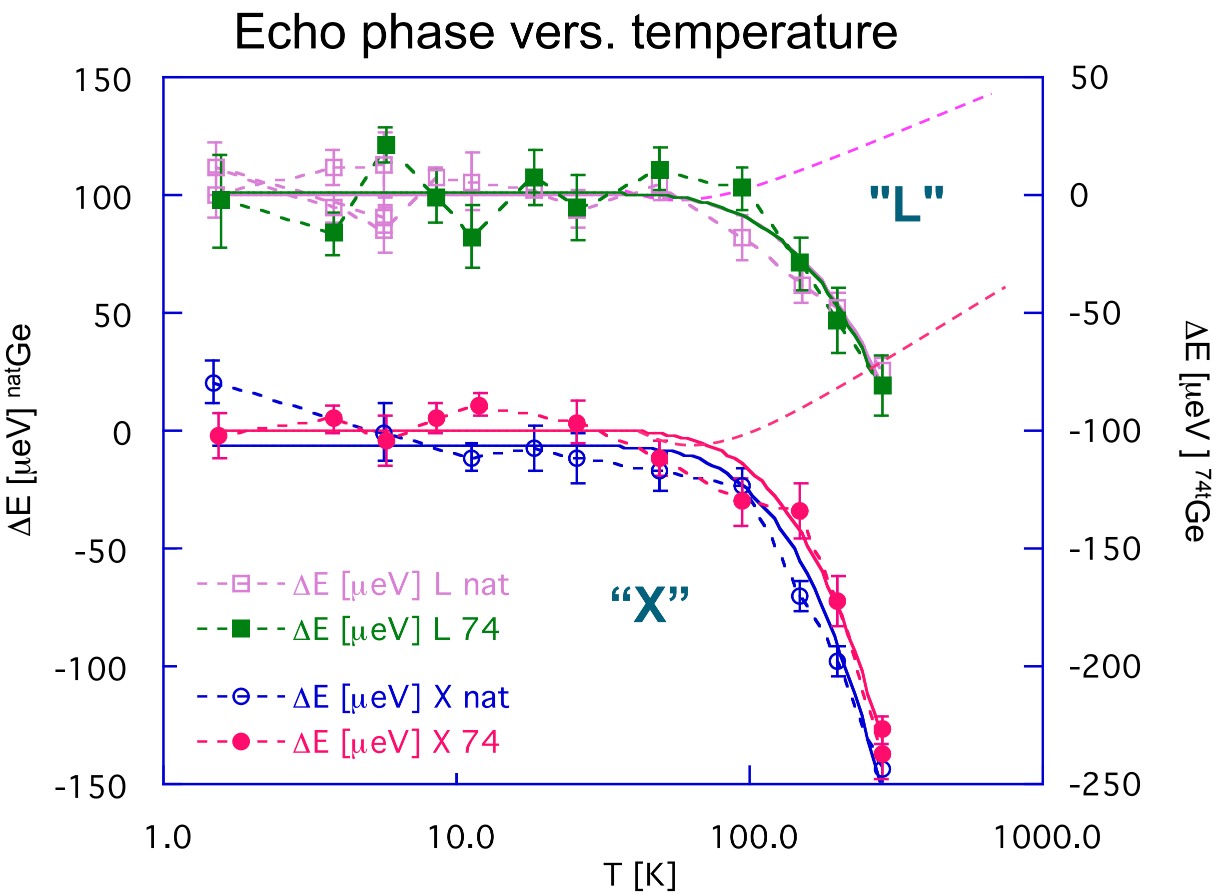
Raman width:
T = 280 K
FWHM = 260 μeV



TASSE - Ge shifts



Isobaric expansion
T = 2 - 300 K, p ≈ 0.1 MPa

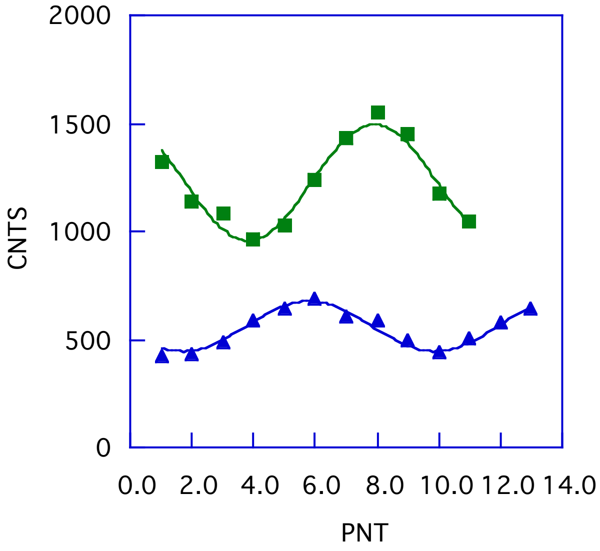


Barron's model:

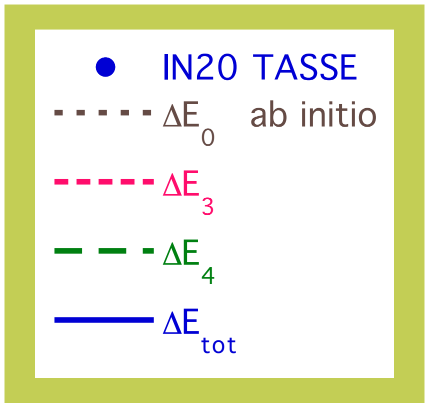
$$\nu(T) = \nu_h \left[1 - \frac{h\nu_g}{k\Theta} \left(n_g + \frac{1}{2} \right) \right]$$

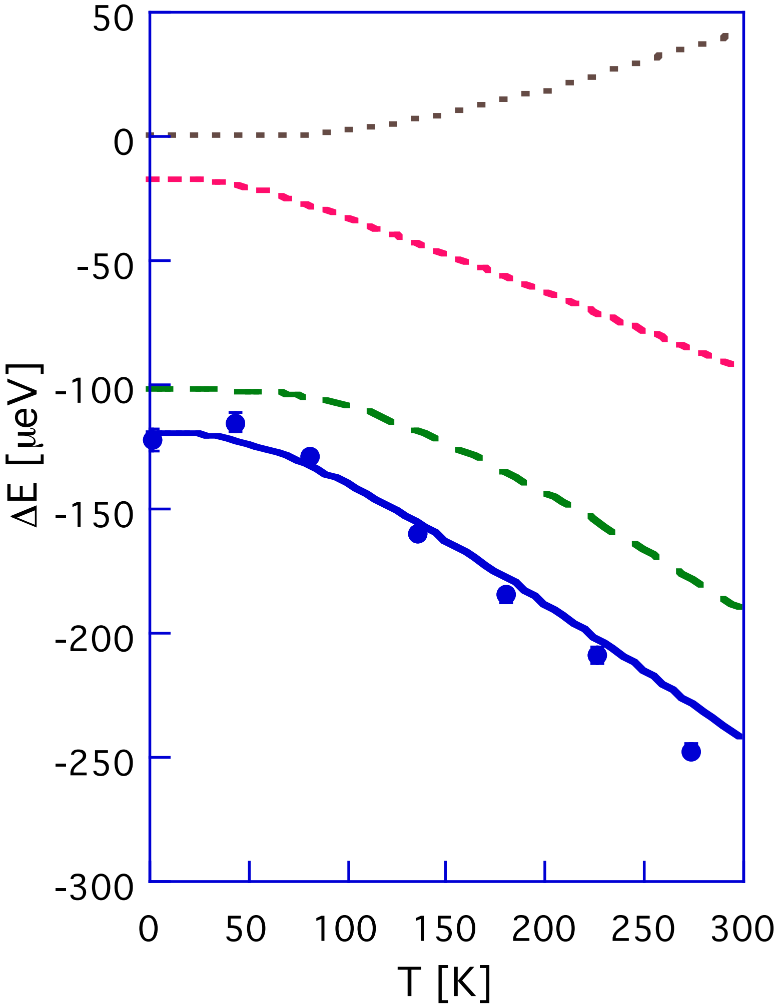
G. Nelin and G. Nilsson,
Phys. Rev. B10 (1974) 612-620.

^{74}Ge: X-point frequency shifts

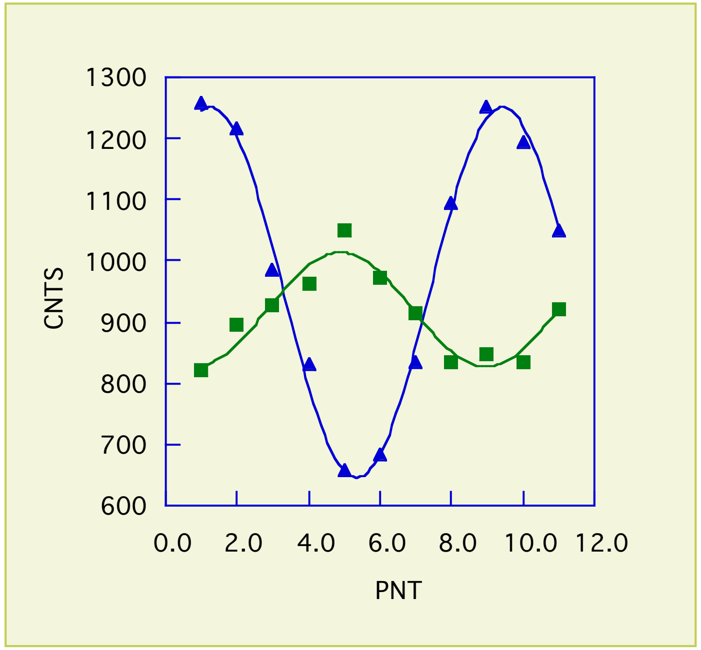


Q = [3 3 1.8]
ΔE = 9.8 meV

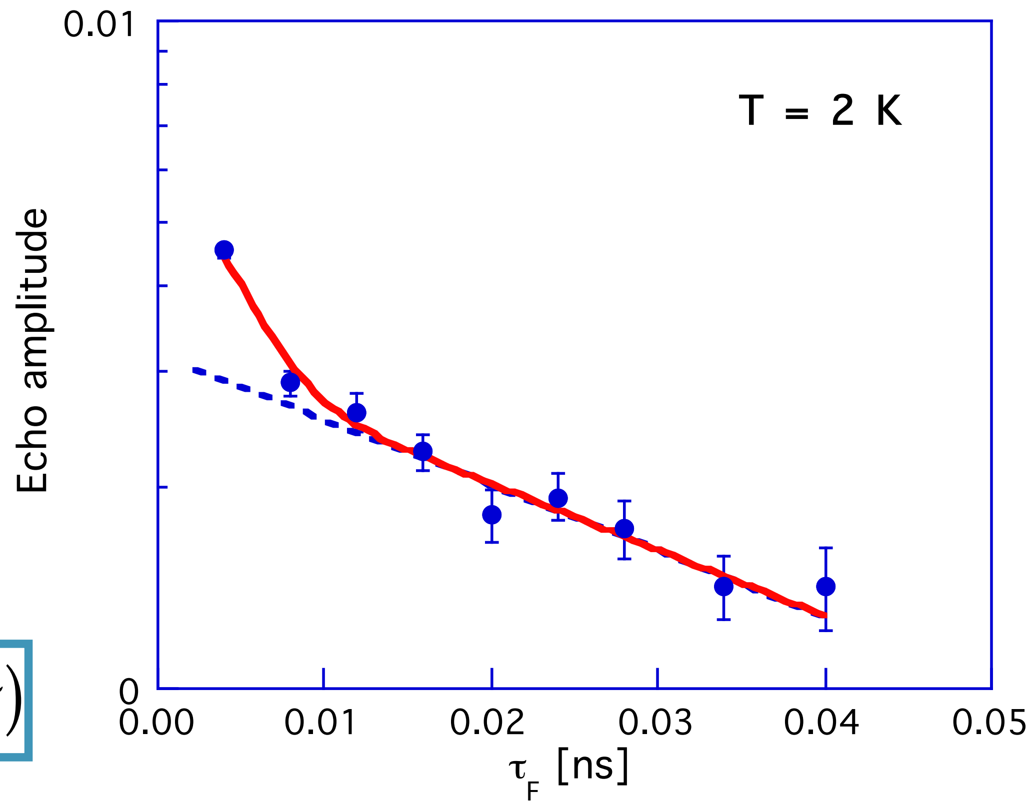




^{74}Ge: X-point phonon width



Echo amplitude vers. Fourier time



Exponential decay:

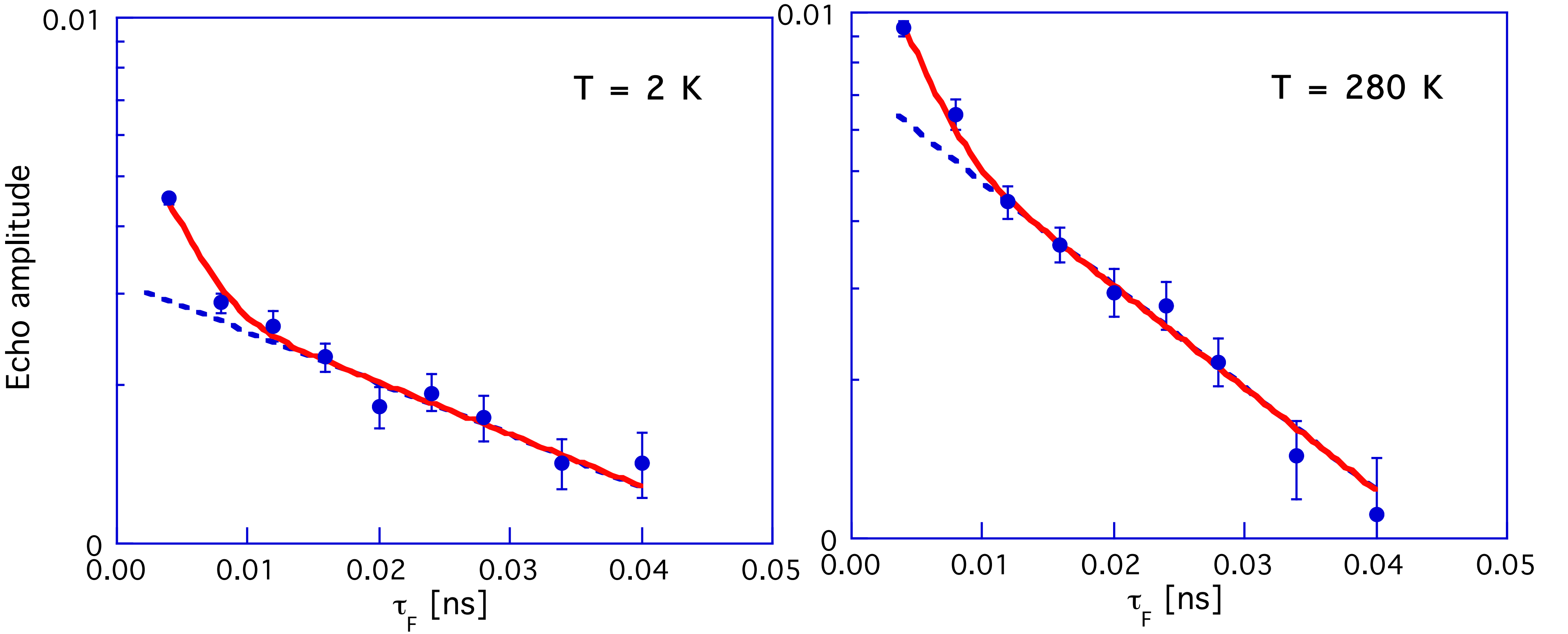
$$\boxed{I(\tau) = I_0\left[1 + c\exp\left(-g\tau^2\right)\right]\exp\left(-\Gamma\tau\right)}$$

TAS resolution
Gaussian

phonon life-time
Lorentzian

^{74}Ge: X-point phonon width

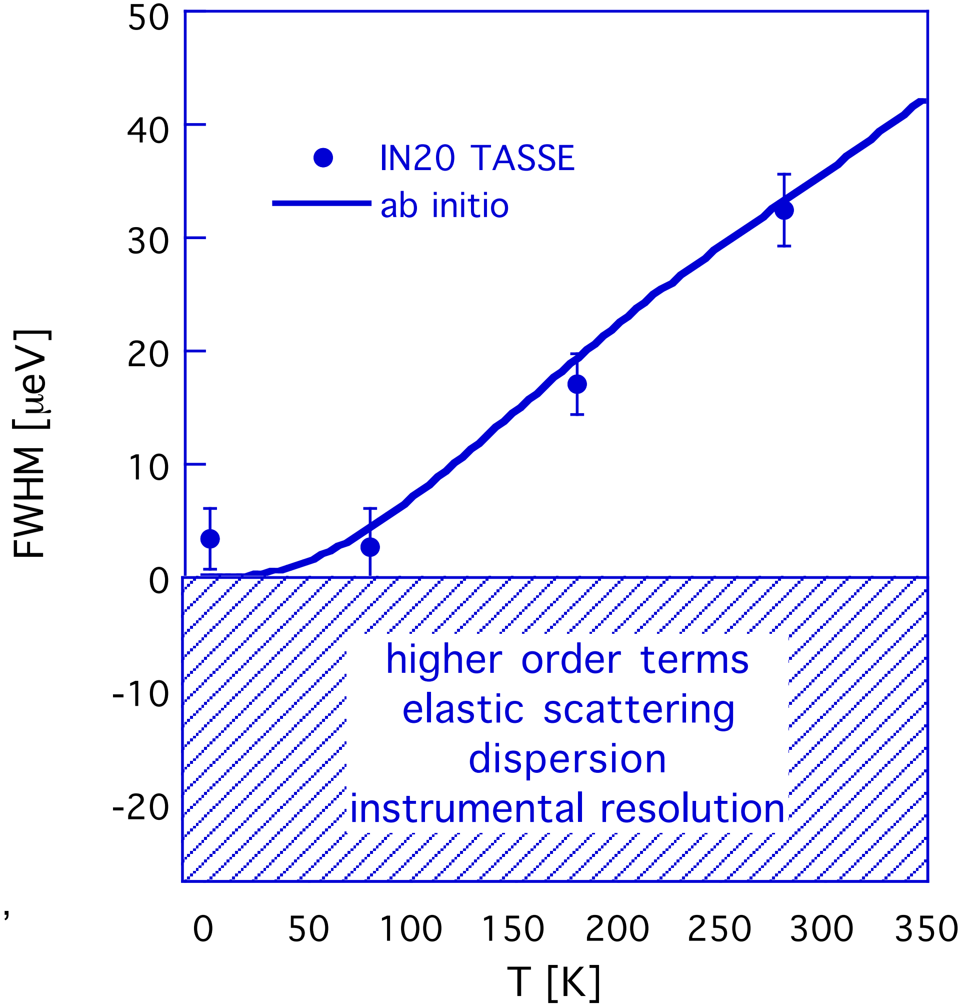
Echo amplitude vers. Fourier time



^{74}Ge: X-point phonon width

Ab initio calculations:

- *lowest (3rd) order in amplitude*
- *sum processes negligible*
- *difference processes T > 50 K*



J. Kulda, A. Debernardi, M. Cardona et al., Phys. Rev. B **69** (2004) 045209

^{74}Ge: X-point phonon width

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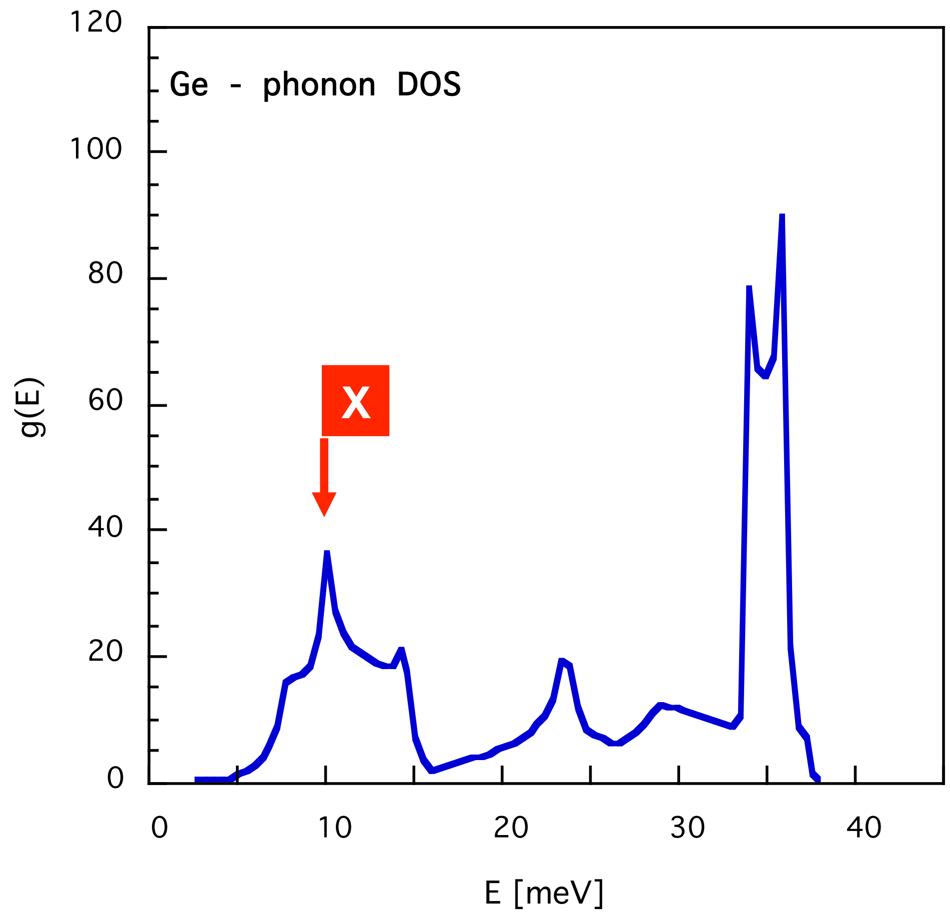
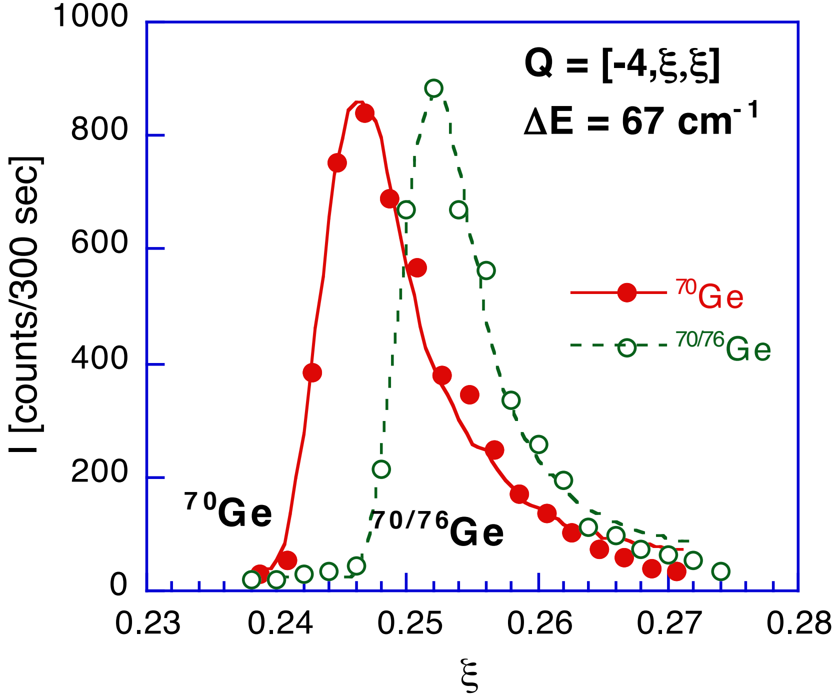


TASSE vers. high resolution TAS



- *same position accuracy*
- *HR TAS faster and easier*
- *TASSE better for FWHM*

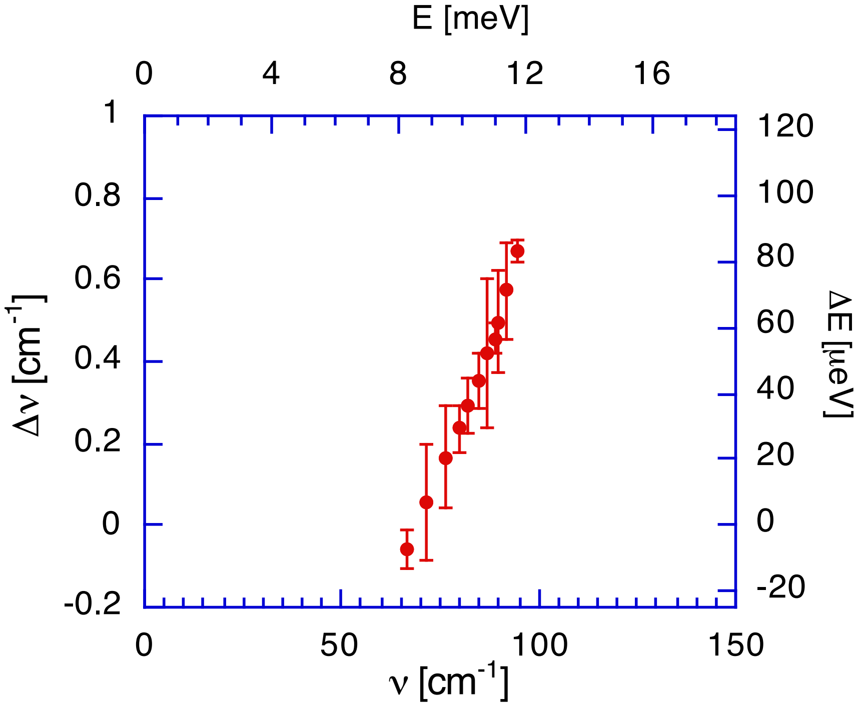
A. Goebel et al., Phys. Rev. B58 (1998) 10510

IN20

Si111/Si111

$R_M = R_A = 50$ m

sample volume < 0.5 cm^3



TASSE vers. high resolution TOF

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PHYSICAL REVIEW B **93**, 134404 (2016)

Anomalous thermal decoherence in a quantum magnet measured with neutron spin echo spectroscopy

F. Groitl,[1,*] T. Keller,[2,3] K. Rolfs,[1,†] D. A. Tennant,[1,4,‡] and K. Habicht[1]

[1]*Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany*

[2]*Max Planck Institute For Solid State Research, 70569 Stuttgart, Germany*

[3]*Max Planck Society Outstation at the FRM II, 85748 Garching, Germany*

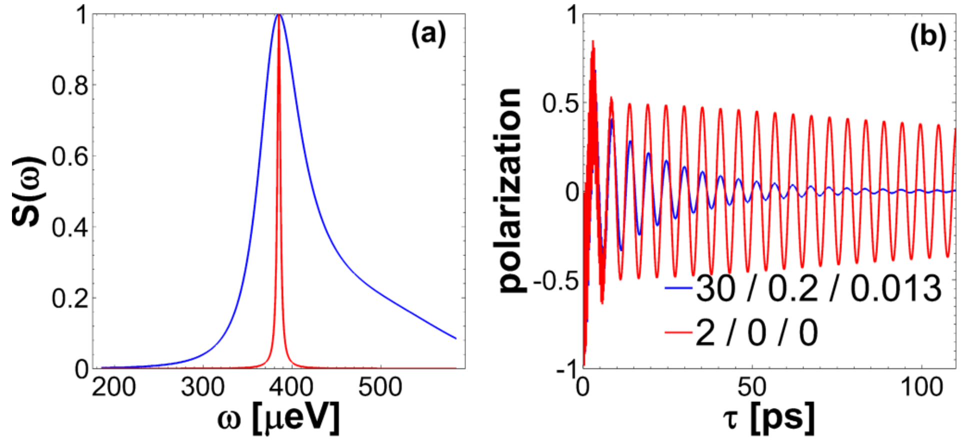
[4]*Technische Universität Berlin, Institut für Festkörperphysik, 10623 Berlin, Germany*

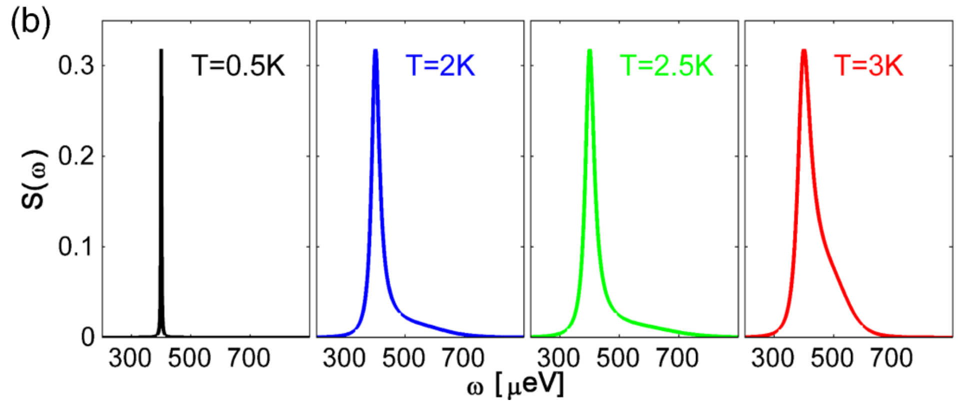
(Received 28 May 2013; revised manuscript received 24 February 2016; published 4 April 2016)

The effect of temperature dependent asymmetric line broadening is investigated in $Cu(NO_3)_2{\cdot}2.5D_2O$, a model material for a one-dimensional bond alternating Heisenberg chain, using the high resolution neutron-resonance spin echo (NRSE) technique. Inelastic neutron scattering experiments on dispersive excitations including phase sensitive measurements demonstrate the potential of NRSE to resolve line shapes, which are non-Lorentzian, opening up a new and hitherto unexplored class of experiments for the NRSE method beyond standard linewidth measurements. The particular advantage of NRSE is its direct access to the correlations in the time domain without convolution with the resolution function of the background spectrometer. This application of NRSE is very promising and establishes a basis for further experiments on different systems, since the results for $Cu(NO_3)_2{\cdot}2.5D_2O$ are applicable to a broad range of quantum systems.

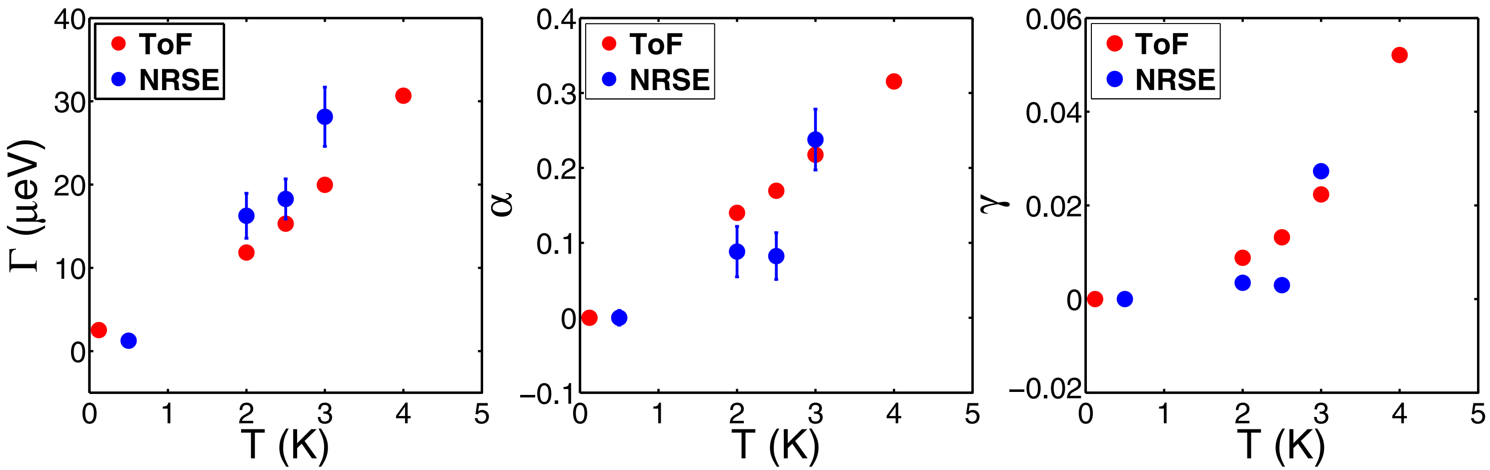
DOI: 10.1103/PhysRevB.93.134404

TASSE vers. high resolution TOF



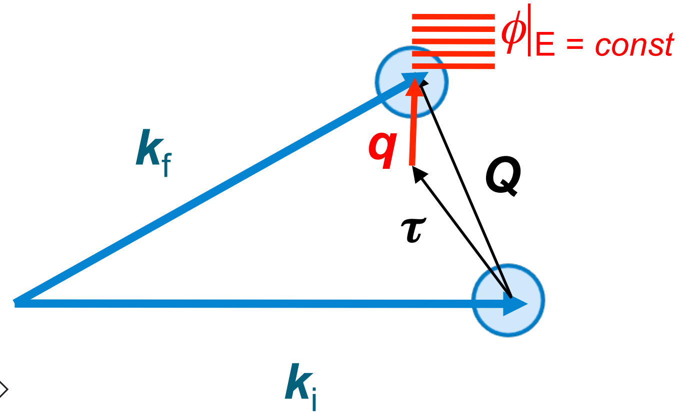


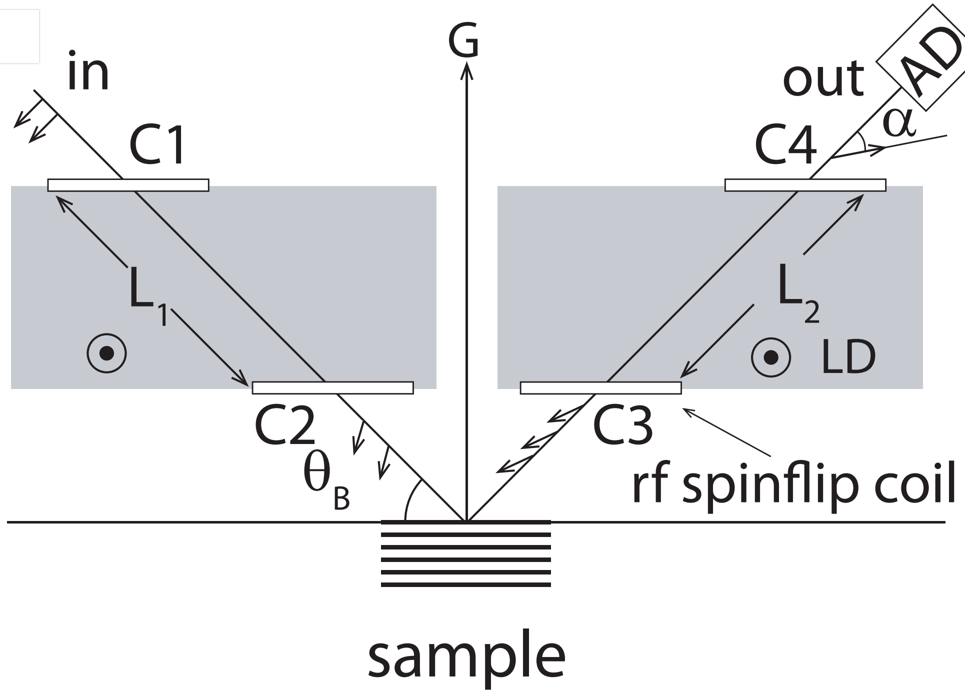
$$S(\omega) = \frac{1}{\pi} \frac{1}{1 + [\omega/\Gamma - \alpha(\omega/\Gamma)^2 + \gamma(\omega/\Gamma)^3]^2}.$$



Larmor coded diffraction

- *same principle of "focusing"*
- *fast (Bragg peaks)*
- *Xray-like resolution $\Delta d/d \approx 10^{-4} - 10^{-5}$*





Rekveldt T.M., Kraan W., Keller T.,
J. Appl. Cryst. 35 (2002) 28

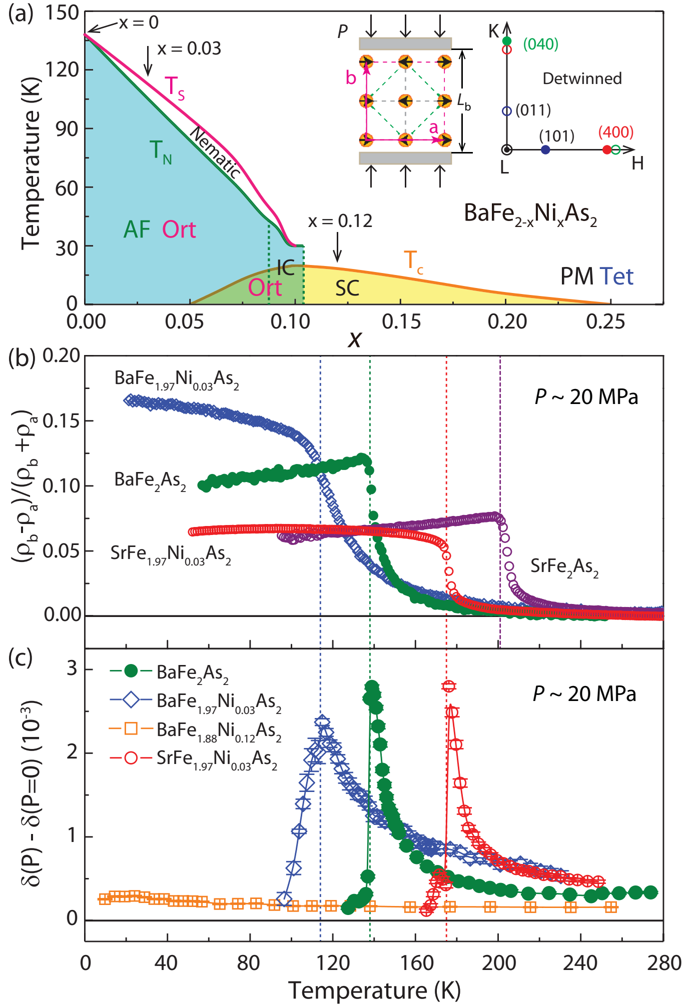
Larmor coded diffraction

$BaFe_{2-x}Ni_xAs_2$

- orthorombic low-T phase detwinned by uniaxial pressure
- investigation of impact on phase transition

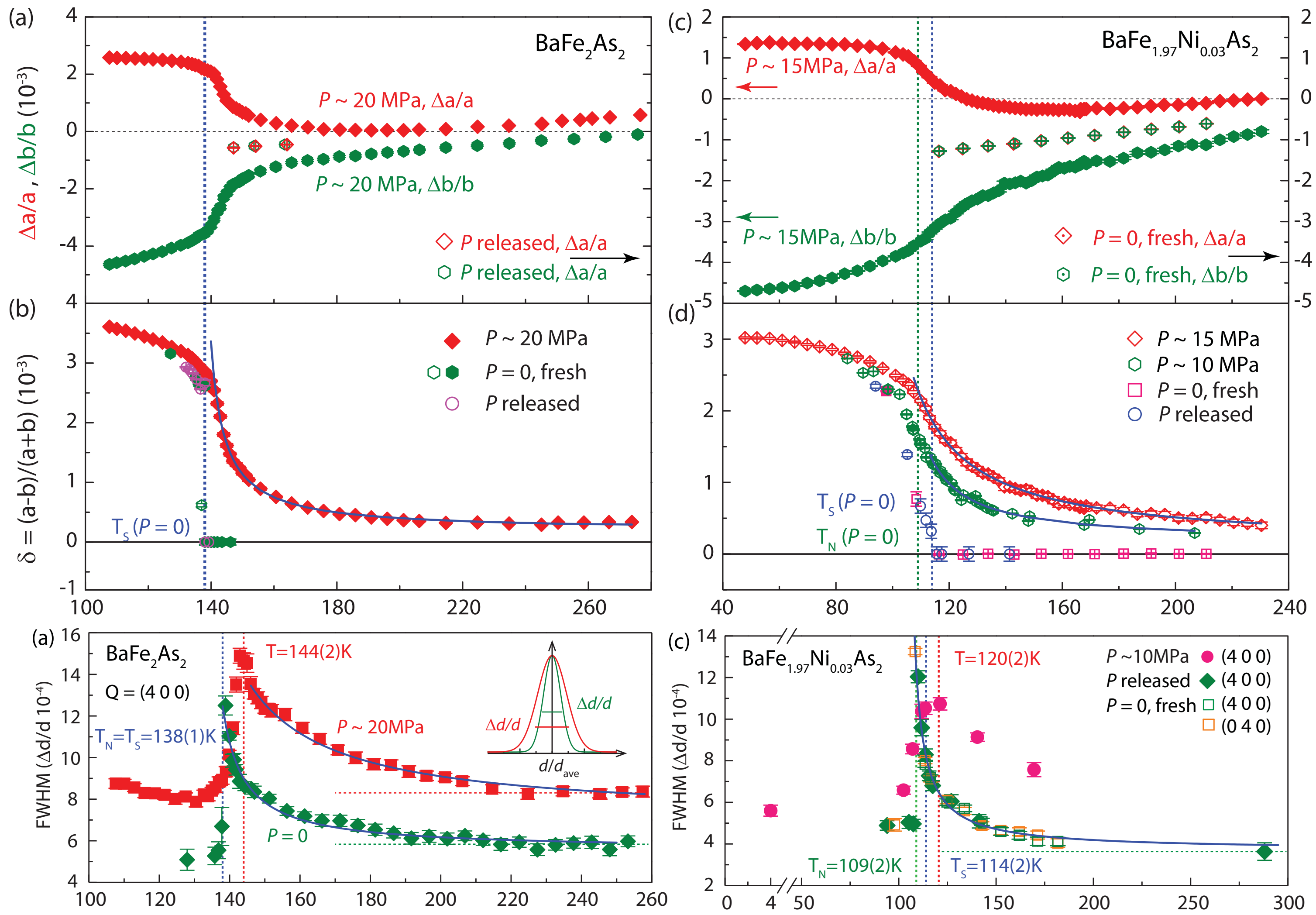
resistivity anisotropy

lattice distortion

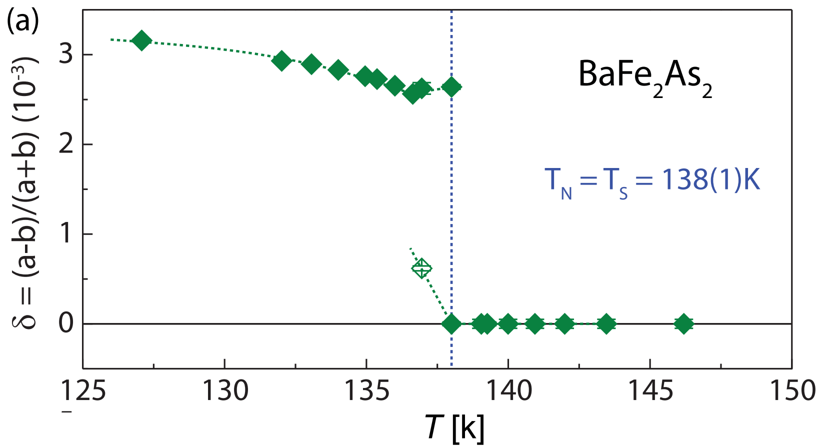


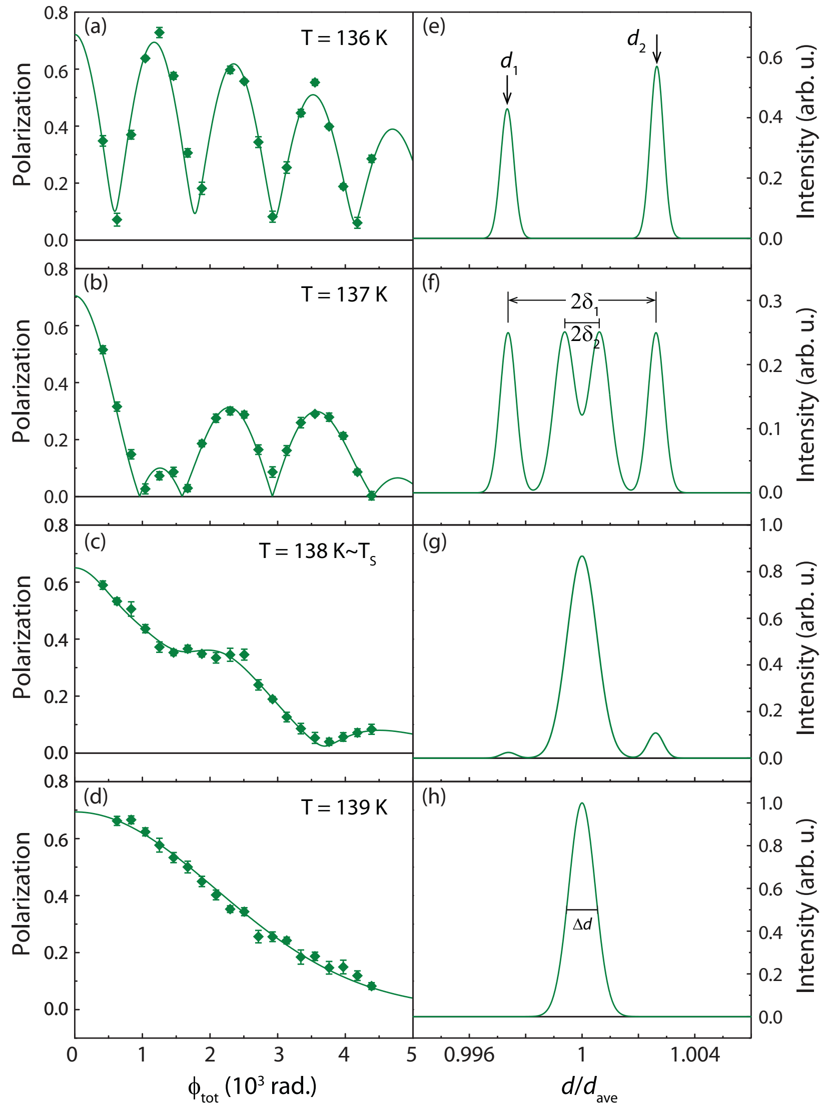
Lu X. et al., Phys. Rev. B93 (2016) 134519

Larmor coded diffraction



Larmor coded diffraction





- *phase coexistence*
- *several d_{hkl} values present*
- *beats in the SE spectrum*

Concluding remarks

IN20 TASSE:

- **range in Q,ω:** *wide ≈ thermal neutron TAS*
- **line positions:** *straightforward & sensitive (≤ 5 μeV)*
- **line widths:** *involved & time-consuming (≥ 5 μeV)*
- **need** *for enhanced luminosity*
  but **limited** *due to dispersion*

^{74}Ge: X-point phonon

- **excellent agreement with *ab initio* calculations**
- *ω(T)* *importance of 4th (and higher) order terms*
- *Γ(T)* *dominated by difference processes*