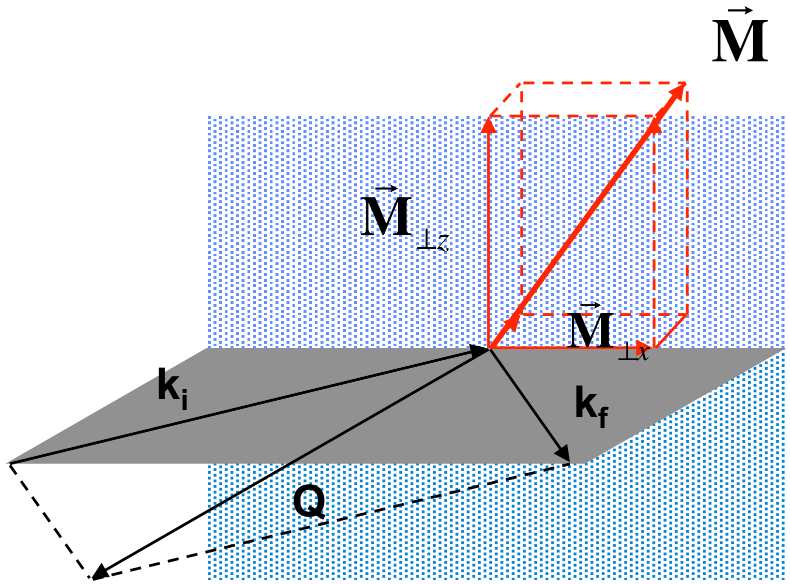
Polarized neutron studies of crystal field effects (in Ce alloys)

Jiří Kulda

Institut Laue-Langevin
Grenoble, France
and
Charles University
Prague, Czech Republic

Magnetic scattering

$$\vec{M}(\vec{Q}) = \sum_j \vec{M}_j(Q)\exp(i\vec{Q}\vec{r}_j)\exp(-W_j)$$



- only projection of **M⊥Q** contributes

$$\vec{M}_\perp(\vec{Q}) = \vec{e}_Q \times \vec{M}(\vec{Q}) \times \vec{e}_Q$$

“Moon’s golden rule”:

$\vec{M}_\perp \parallel \vec{\sigma}_n$	non spin-flip (NSF)	U^{++}, U^{--}
$\vec{M}_\perp \perp \vec{\sigma}_n$	spin-flip (SF)	U^{+-}, U^{-+}

Diagonal polarization analysis

- **Partial intensities (polarized beam):**

$$I_x^{NSF} \approx N^2 + \frac{1}{3} I_{SI} \qquad I_x^{SF} \approx M_{\perp y}^2 + M_{\perp z}^2 + \frac{2}{3} I_{SI}$$

$$I_y^{NSF} \approx \left(N + M_{\perp y}\right)^2 + \frac{1}{3} I_{SI} \qquad I_y^{SF} \approx M_{\perp z}^2 + \frac{2}{3} I_{SI}$$

$$I_z^{NSF} \approx \left(N + M_{\perp z}\right)^2 + \frac{1}{3} I_{SI} \qquad I_z^{SF} \approx M_{\perp y}^2 + \frac{2}{3} I_{SI}$$

- **Use difference signal to extract information:**

$$\chi_y'' \approx M_{\perp y}^2 \approx I_x^{SF} - I_y^{SF}$$

$$\chi_z'' \approx M_{\perp z}^2 \approx I_x^{SF} - I_z^{SF}$$

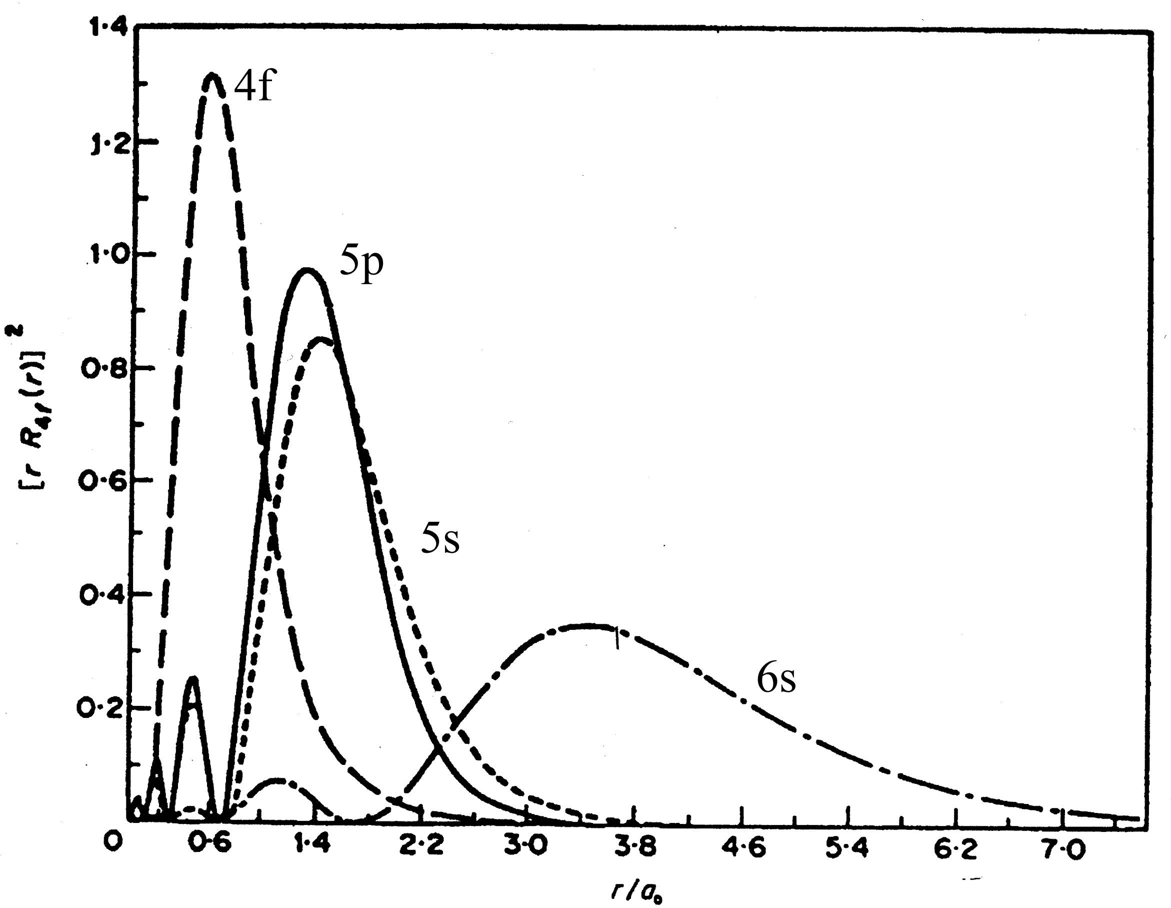
Crystal (electric) field

- interaction of electrons with electrons (exchange) and electric fields of neighbouring atoms
- perturbation of the orientational degeneracy of a free ion – splitting of $\left| M_J \right\rangle$ levels
- magnetism effects depend on the electronic configuration
  - strong for extended 3d shells (> SO)
  - weaker for 4f shells burried behind extended $5p^6$ and $5s^2$ (< SO)
- in rare earths competition with spin-orbit coupling

• for an extensive coverage

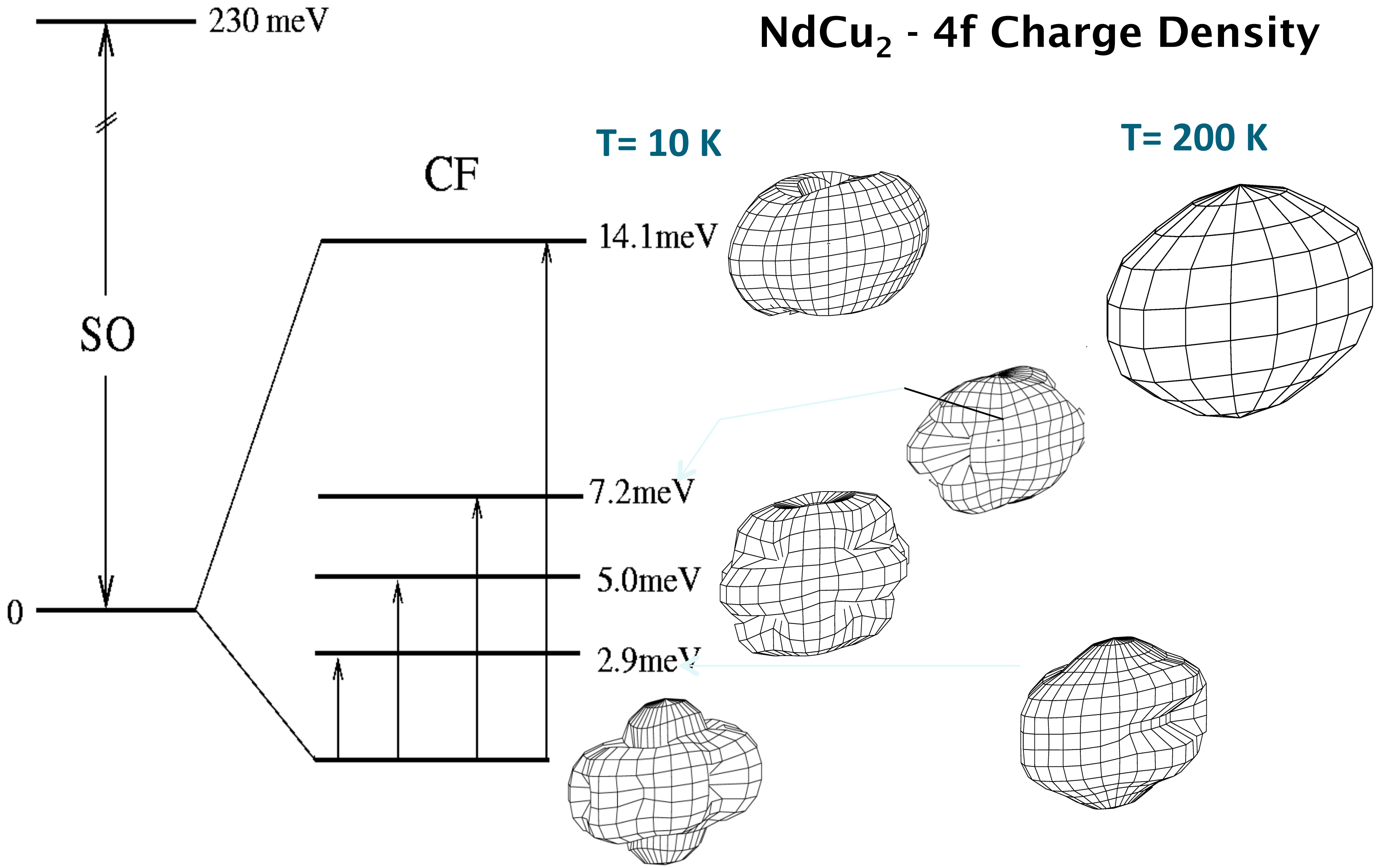
P. Fulde and M. Loewenhaupt, Adv. in Phys. 34, 589 (1985)

Crystal (electric) field



Crystal (electric) field

$NdCu_2$ - 4f Charge Density



courtessy P. Alexeev

CEF Hamiltonian

- for weak CF the handling can be constrained to the lowest multiplet

$$\hat{H}_{CF} = \sum_{l,m} B_l^m O_l^m$$

CEF parameters

Stevens operators (polynomials in J_α)

- further constraints: *l* even; *l* ≤ 6

- for more an extensive coverage

P. Fulde and M. Loewenhaupt, Adv. in Phys. 34, 589 (1985)

CePtSn: neutrons w/o PA

Pnma
TiNiSi-type

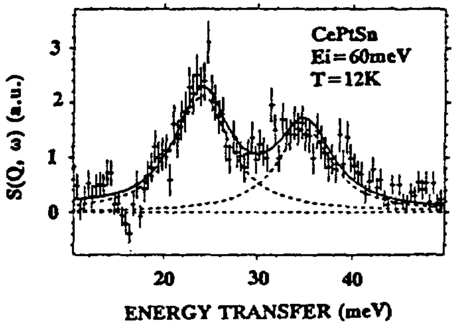


Fig. 2 Inelastic neutron spectrum from CePtSn at 12K.

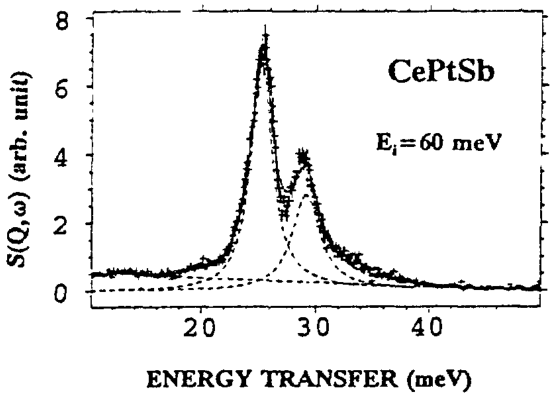


Fig. 2 Inelastic neutron spectrum of CePtSb at 13K.

P6$_3$m
GaGeLi-type

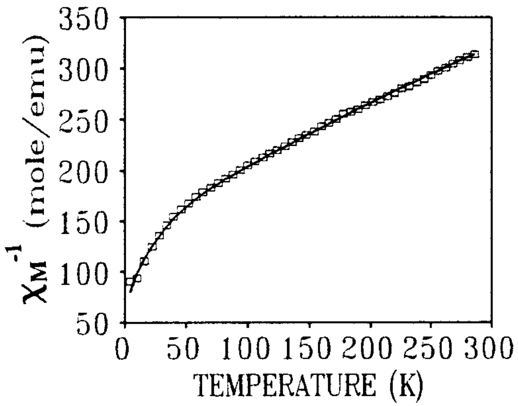


Fig. 3 χ^{-1} versus temperature of CePtSn.

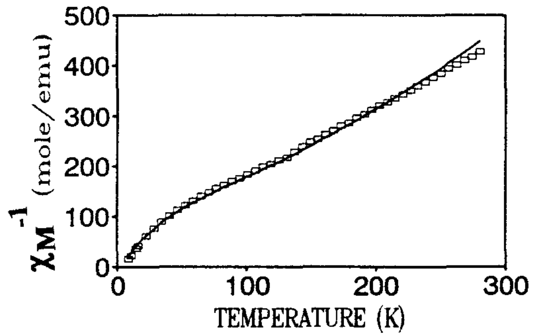
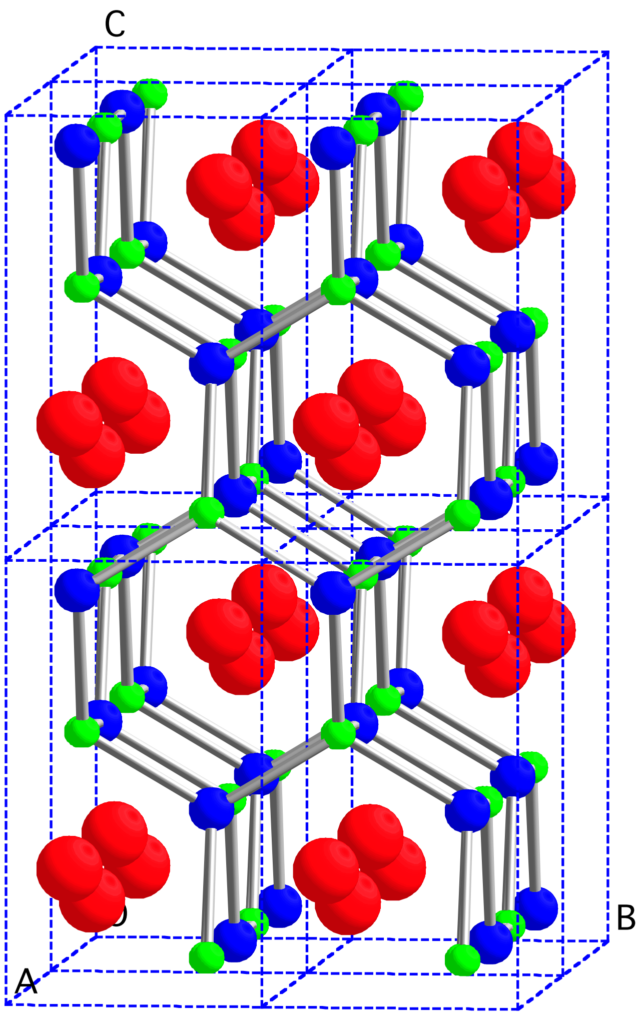


Fig. 3 Reciprocal magnetic susceptibility *versus* temperature for CePtSb; the solid line is the CF fit.

$$I \approx \frac{k'}{k} f^2(Q) \exp(-2W) \sum_{mm'} n_m \left| \langle m' | J^{\perp} | m \rangle \right|^2 \delta\left(\omega_{m'} - \omega_m - \omega\right)$$

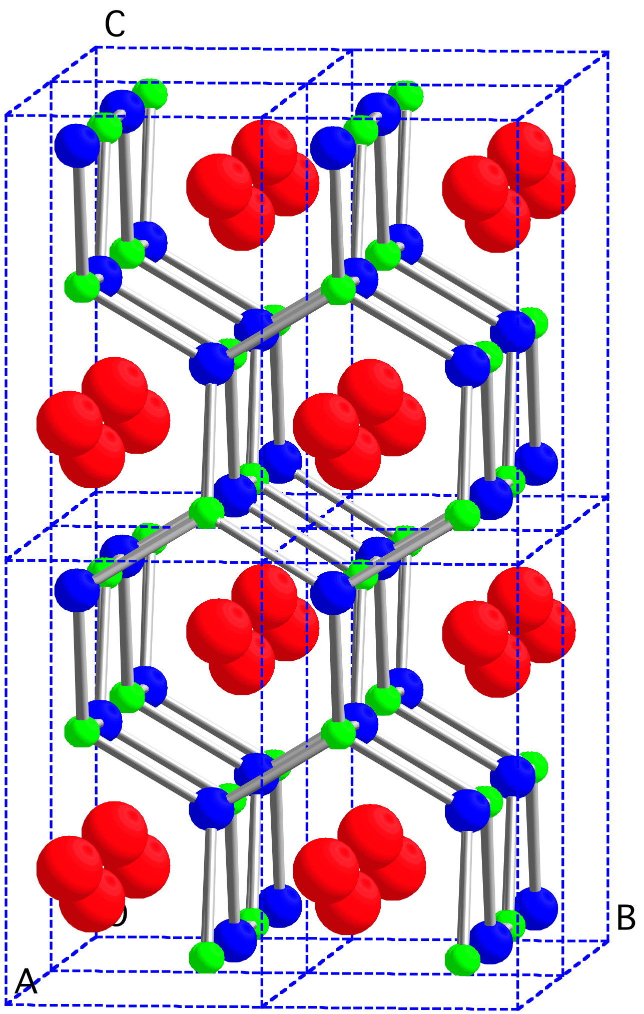
D. T. Adroja and B. Rainford, Phys. B 194-196, 363 (1994)

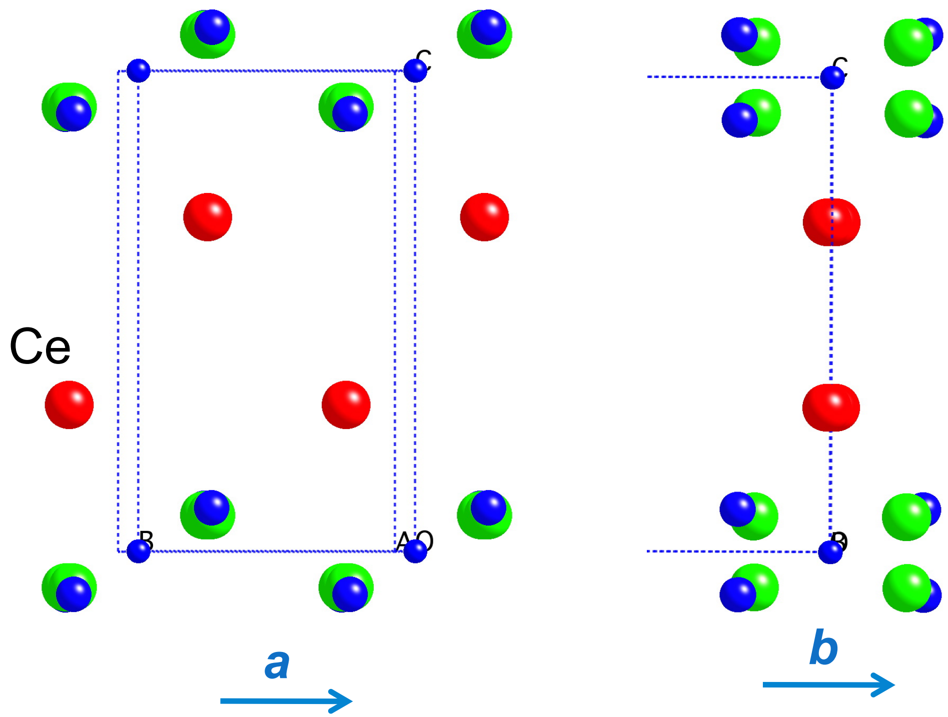
CePtSn



- structure TiNiSi-type (*Pnma*)
- strong Kondo effect (T_K = 10 K)
- antiferromagnet (T_N = 7.5 K)
- RKKY exchange interaction
- propagation ***q*** = (0 0.5 0) + spin slips
- first order (?) transition T_M = 5 K
- magnetization anisotropy (***a*** easy)

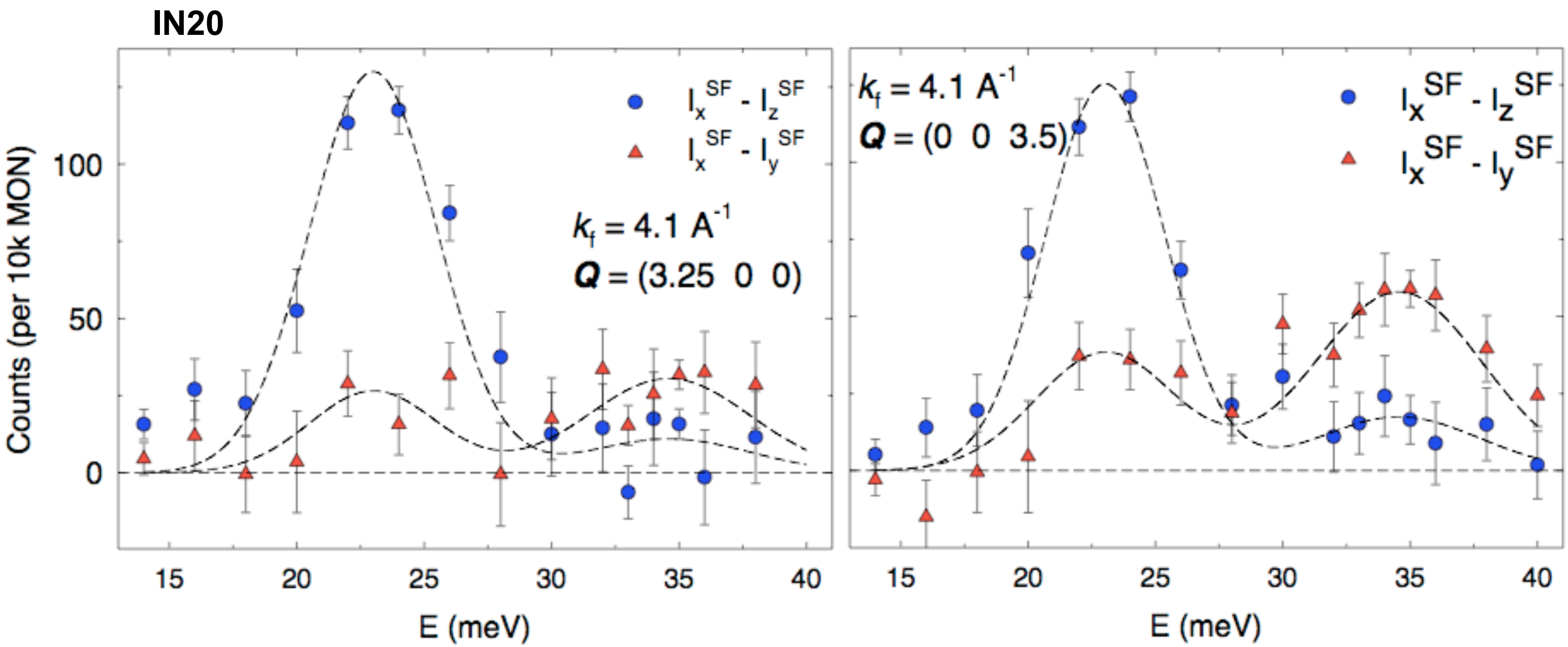
CePtSn





- structure TiNiSi-type
- orthorombic (*Pnma*)
- Ce atoms lower site symmetry (monoclinic)
- Ce groundstate $^2F_{5/2}$ split into 3 doublets (Kramers ion)

CePtSn: neutrons with PA



$$I_x^{SF} - I_{y,z}^{SF} \approx \frac{k'}{k} f^2(Q)\exp(-2W)\sum_{mm'} n_m \left|\langle m'|J_{y,z}|m\rangle\right|^2 \delta\left(\omega_{m'} - \omega_m - \omega\right)$$

B. Janousova et al., PRB 69, 220412(R) (2004)

Crystal field in CePtSn

- Ce $^2F_{5/2}$ Hamiltonian (3 doublets)

= 0 because of symmetry

$$\hat{H}_{CF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_2^2 O_2^2 + B_4^2 O_4^2 + C_2^2 \Omega_2^2 + C_4^2 \Omega_4^2 + C_4^4 \Omega_4^4$$

orthorombic monoclinic

Stevens operators

$$O_n^m = \left(O_n^m + O_n^{-m}\right)/2$$

$$\Omega_n^m = -i\left(\Omega_n^m - \Omega_n^{-m}\right)/2$$

Crystal field in CePtSn

Diagonal matrix elements

$$\left|\left\langle n\middle|J_i\middle|n\right\rangle\right|^2$$

supplied by bulk measurements

	Experiment	Theoretical calculations	
		Monoclinic H_{CF}	Orthorhombic H_{CF}
	(this work)	(this work)	(Diviš *et al.*[a])
Δ_1(meV)	23.0(4)	24.0(8)	23.3
Δ_2(meV)	34.6(7)	34.6(–)	35.2
I_1/I_2	1.45(9)	1.34(25)	0.758
$\|\langle 0\|J_a\|1\rangle\|^2$	262(71)	270(15)	324
$\|\langle 0\|J_b\|1\rangle\|^2$	777(54)	715(12)	385
$\|\langle 0\|J_c\|1\rangle\|^2$	166(40)	167(12)	168
$\|\langle 0\|J_a\|2\rangle\|^2$	474(69)	518(20)	20
$\|\langle 0\|J_b\|2\rangle\|^2$	113(56)	101(12)	405
$\|\langle 0\|J_c\|2\rangle\|^2$	245(32)	259(16)	730

Eigenfunctions of the CF Hamiltonian

$\|\Psi_0\rangle$	$(0.62+0.42i)\|+\frac{1}{2}\rangle$	$+(0.15+0.43i)\|-\frac{3}{2}\rangle$	$-0.48\|+\frac{5}{2}\rangle$
	$0.75i\|-\frac{1}{2}\rangle$	$+(0.27+0.37i)\|+\frac{3}{2}\rangle$	$+(0.27-0.40i)\|-\frac{5}{2}\rangle$
$\|\Psi_1\rangle$	$(-0.56+0.13i)\|+\frac{1}{2}\rangle$	$+(0.75+0.06i)\|-\frac{3}{2}\rangle$	$-0.32\|+\frac{5}{2}\rangle$
	$0.58i\|-\frac{1}{2}\rangle$	$-(0.22+0.72i)\|+\frac{3}{2}\rangle$	$+(0.07+0.31i)\|-\frac{5}{2}\rangle$
$\|\Psi_2\rangle$	$(-0.30+0.15i)\|+\frac{1}{2}\rangle$	$-(0.28-0.39i)\|-\frac{3}{2}\rangle$	$+0.82\|+\frac{5}{2}\rangle$
	$0.33i\|-\frac{1}{2}\rangle$	$-(0.23-0.42i)\|+\frac{3}{2}\rangle$	$-(0.73-0.36i)\|-\frac{5}{2}\rangle$

CePtSn: magnetisation curves

Pnma
TiNiSi-type

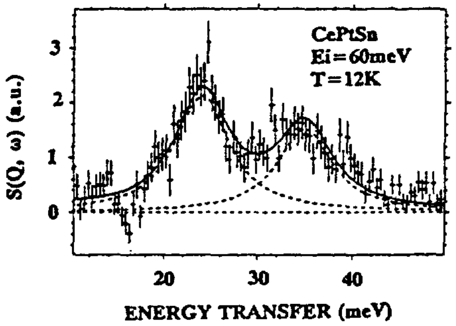


Fig. 2 Inelastic neutron spectrum from CePtSn at 12K.

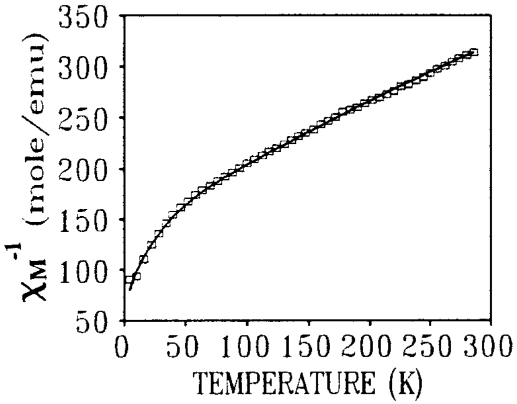
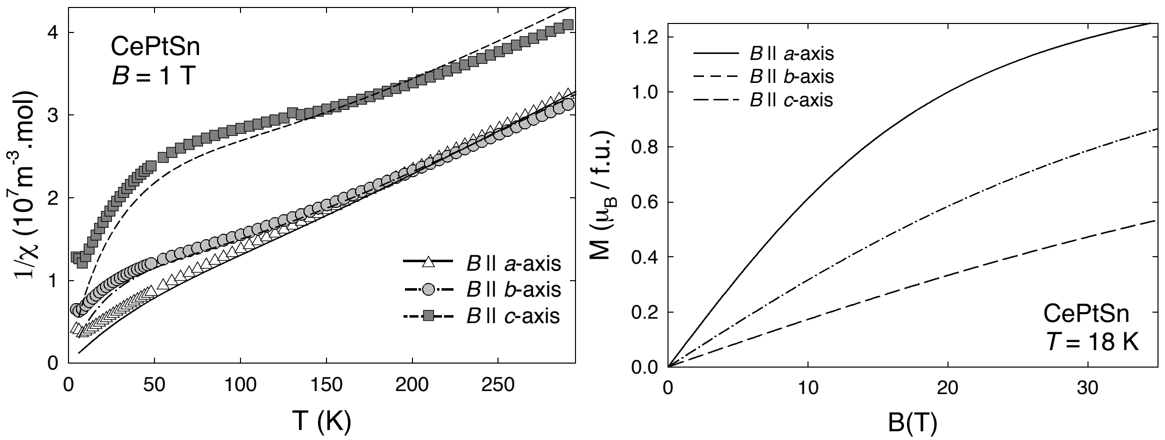


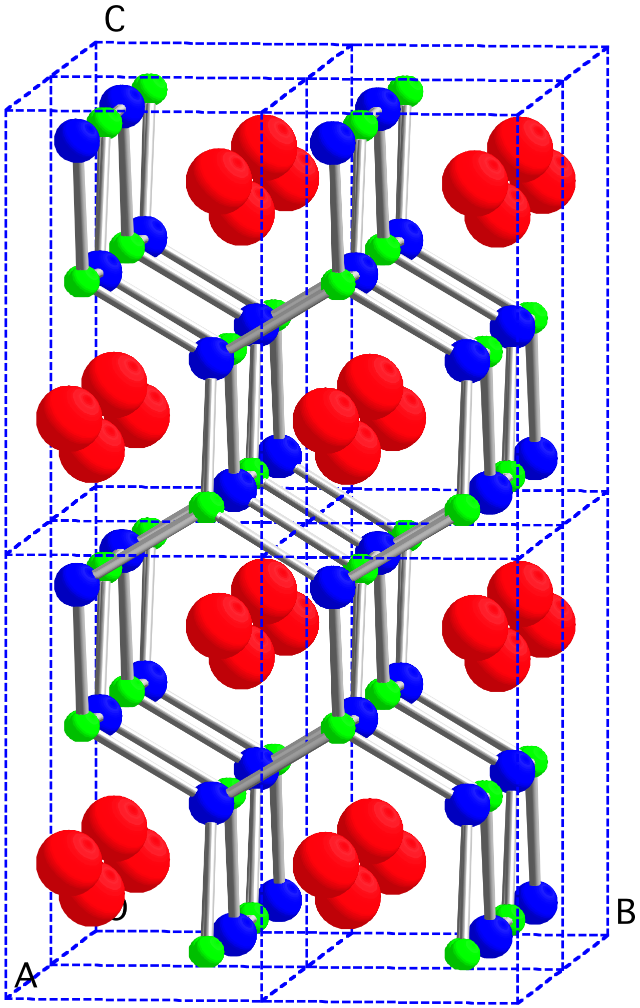
Fig. 3 χ^{-1} versus temperature of CePtSn.

D. T. Adroja and B. Rainford, Phys. B 194-196, 363 (1994)



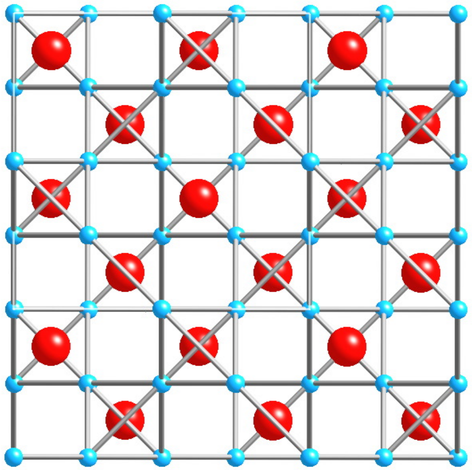
B. Janousova, PhD thesis (2004)

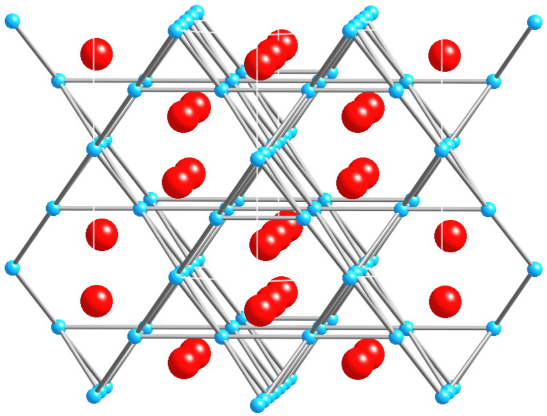
CePtSn

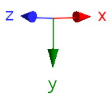


- neutron scattering by transitions between CEF split levels is a single-site process
- access to local site-symmetry for general lattice positions
- PA reveals directly the angular momentum projections
- bulk techniques & powder scattering provide access only to the symmetrized part

CeAl$_2$







- cubic structure (MgCu2) Fd-3m
- Ce sits in a general position

Ce [0.125 0 0]   Al [0.5 0.5 0.5]

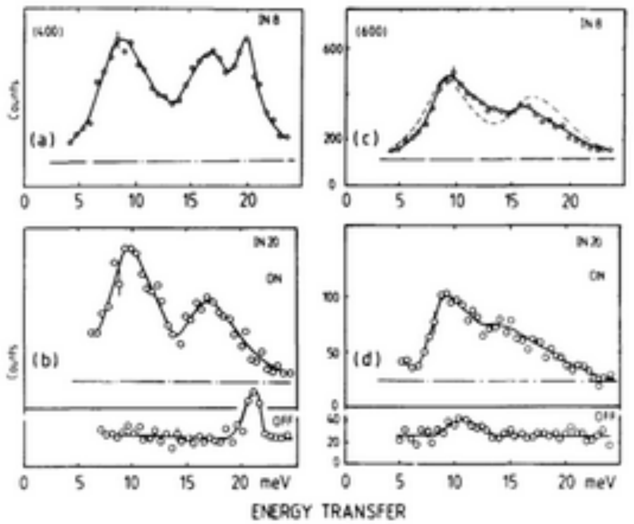
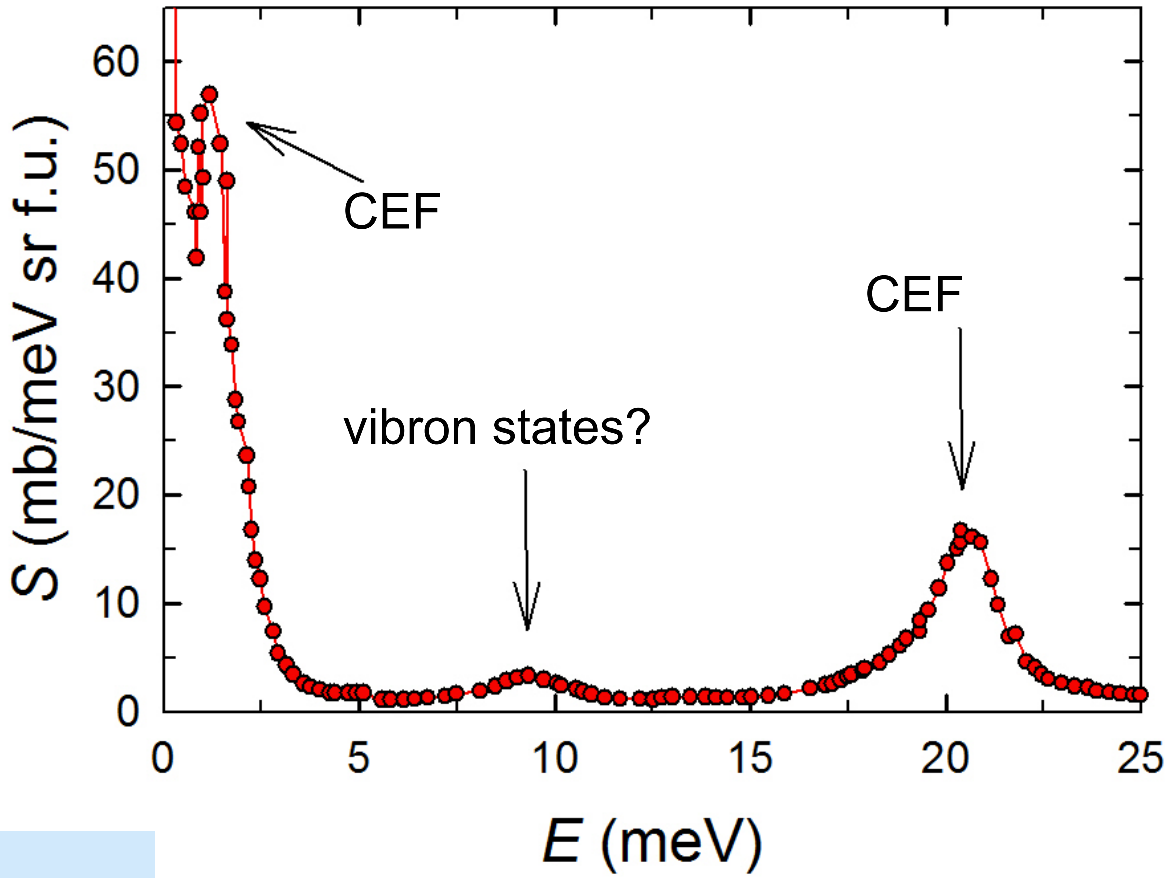


Fig. 2. Inelastic neutron spectra of CeAl$_2$ at (004) and (006) which are nonequivalent points for the diamond Ce sublattice. Data taken at $T = 6$ K on IN8 and IN20, typical measuring times per point are 10 and 14 min, respectively. For other experimental conditions and explanations see text.

"unexpected" signature of a triplet splitting in a cubic structure

Loewenhaupt et al., JMMM 93-94 (1987) 73

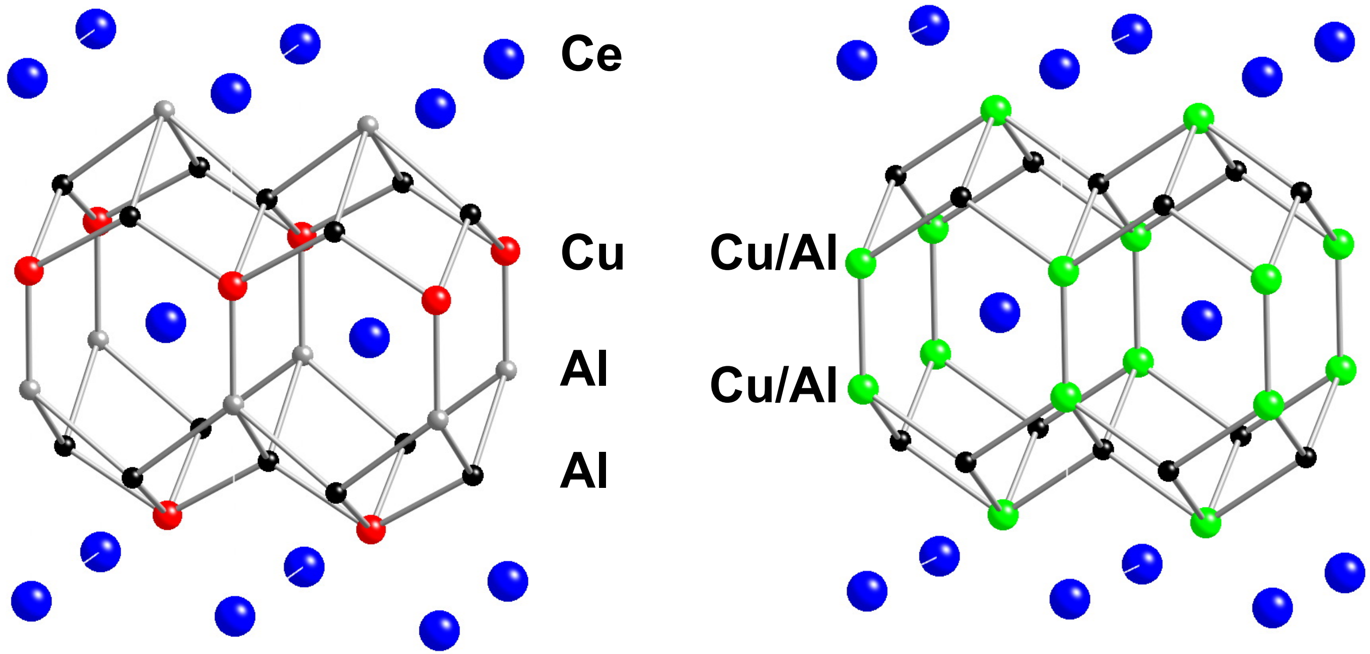
$CeCuAl_3$



ISIS
unpolarized TOF

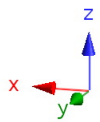
Adroja et al., PRL 108, 216402 (2012)

$CeCuAl_3$



I4mm ideal

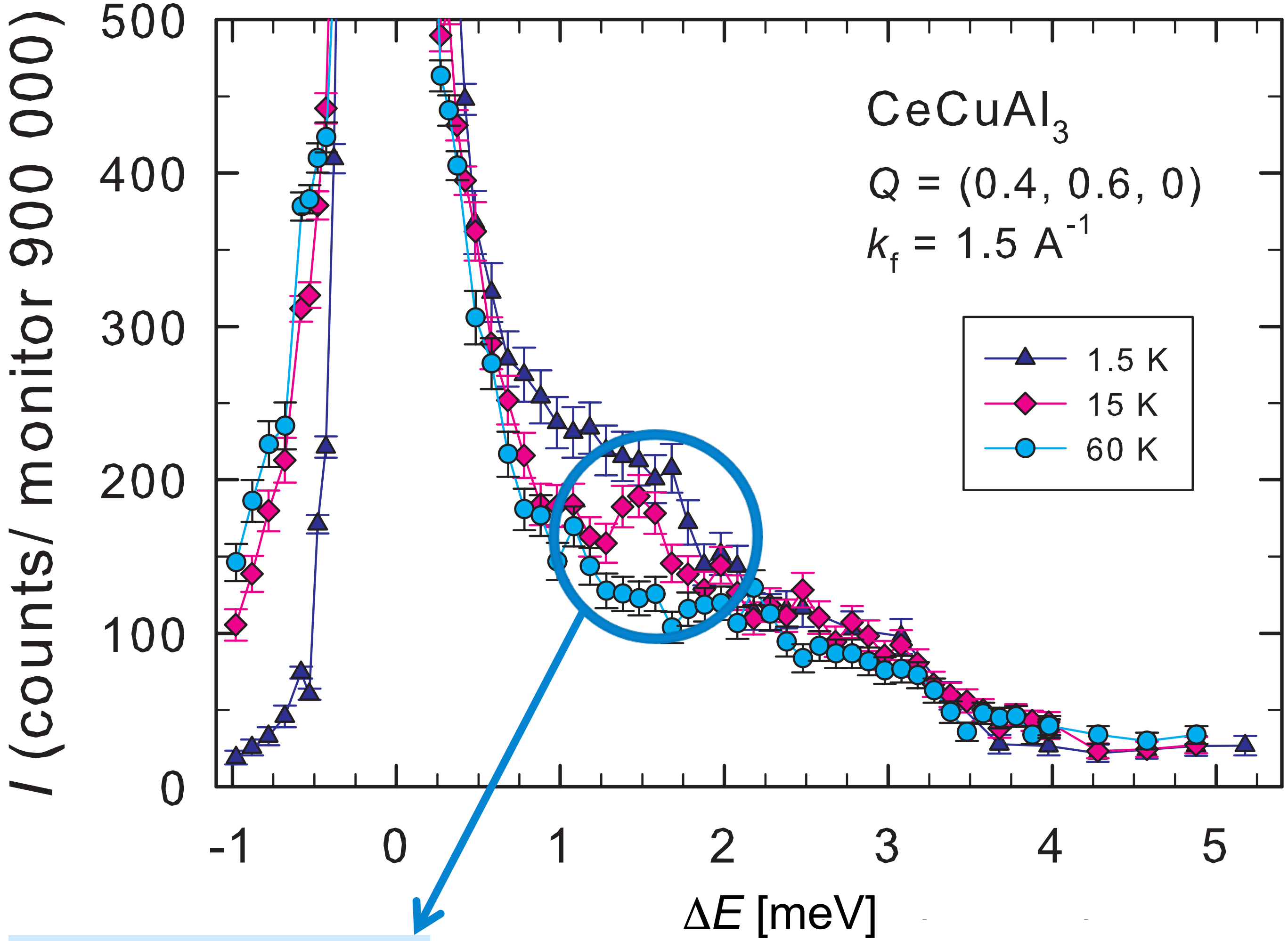
I4/m mm disordered



- tetragonal ($BaNiSn_3$) I4mm
- Ce sits in a special position

BUT Cu/Al are probably disordered

ThALES: $CeCuAl_3$ (unpolarized)



CEF signal?

Polarization analysis needed!

Concluding remarks

- the splitting of the ground state energy depends on the local (single ion) environment
- for atoms in general positions it will not correspond to the full crystal symmetry

details of crystal structure are important

- CEF excitation lines are (in an ideal case) dispersionless
- but their intensities may be strongly anisotropic

only single crystal data provide full information

neutron spectroscopy with PA provides

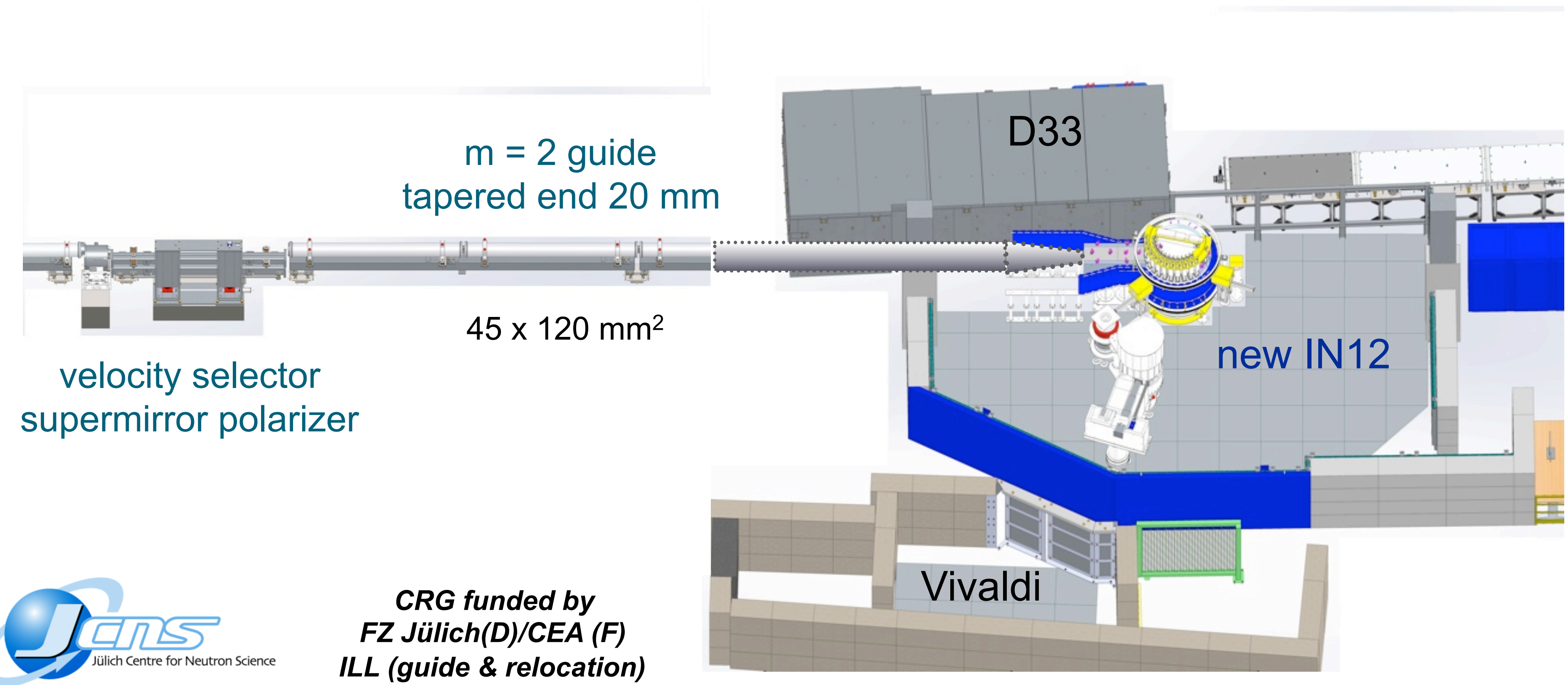
- clean separation of CEF excitations and phonons (even on a polycrystal)
- extensive information on the CEF Hamiltonian (on monocrystals)

use neutron PA & crystals whenever possible!

IN12B cold TAS

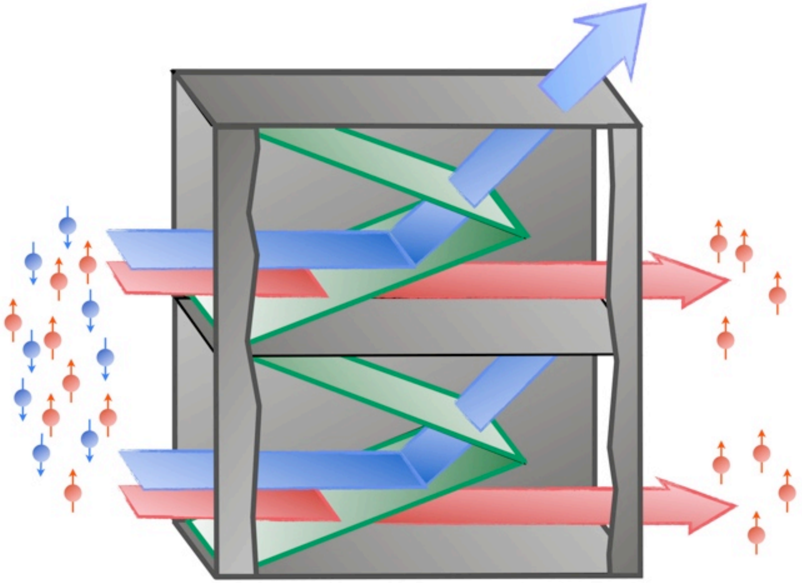
Three Axis Low Energy Spectrometer

- m = 2 cold guide
- extended kinematic range
- full 15T compatibility
- polarised beam option

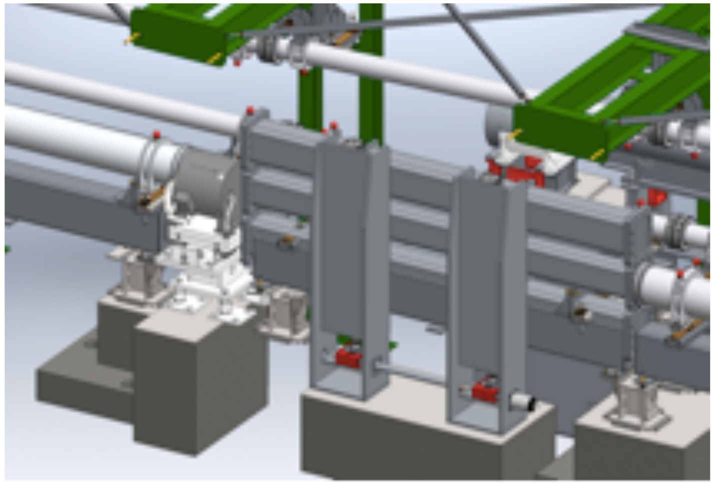


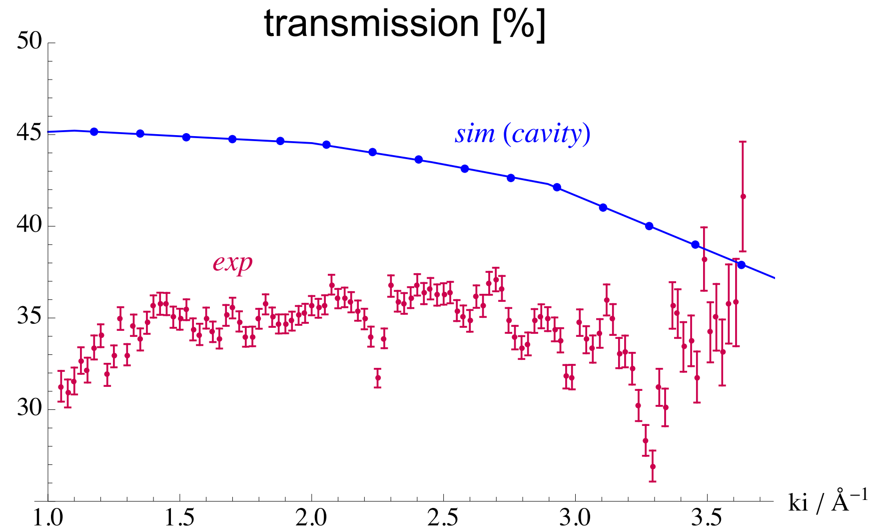
CRG funded by
FZ Jülich(D)/CEA (F)
ILL (guide & relocation)

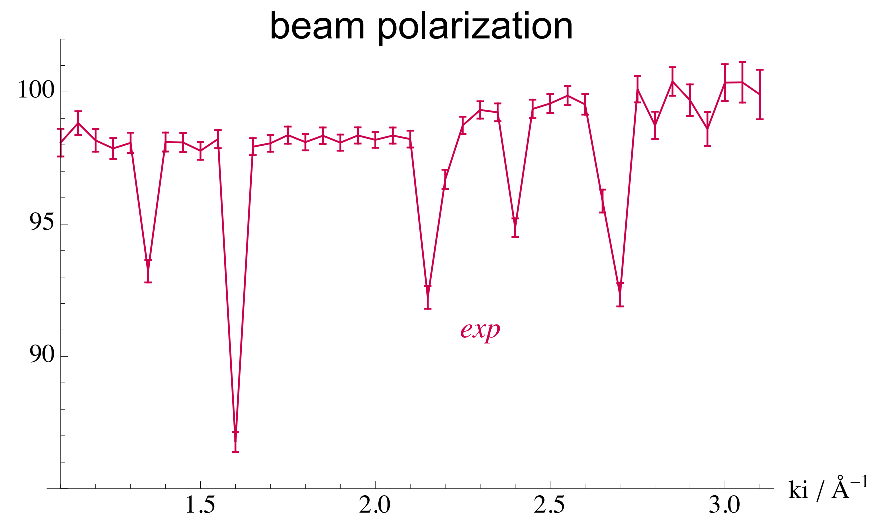
IN12B polarizer



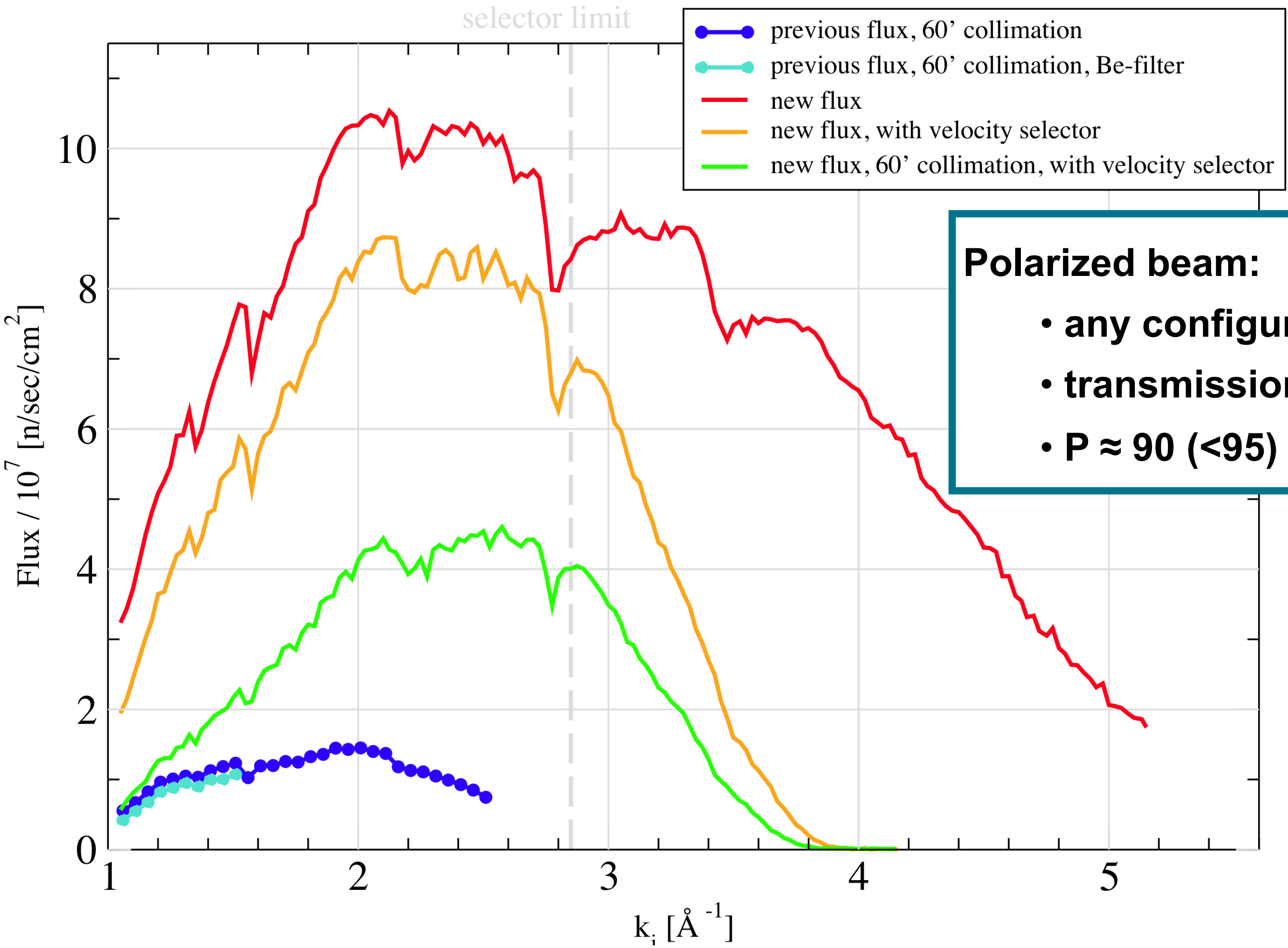
polarizing mirrors with m=0.7 / 3.8







IN12B neutron flux

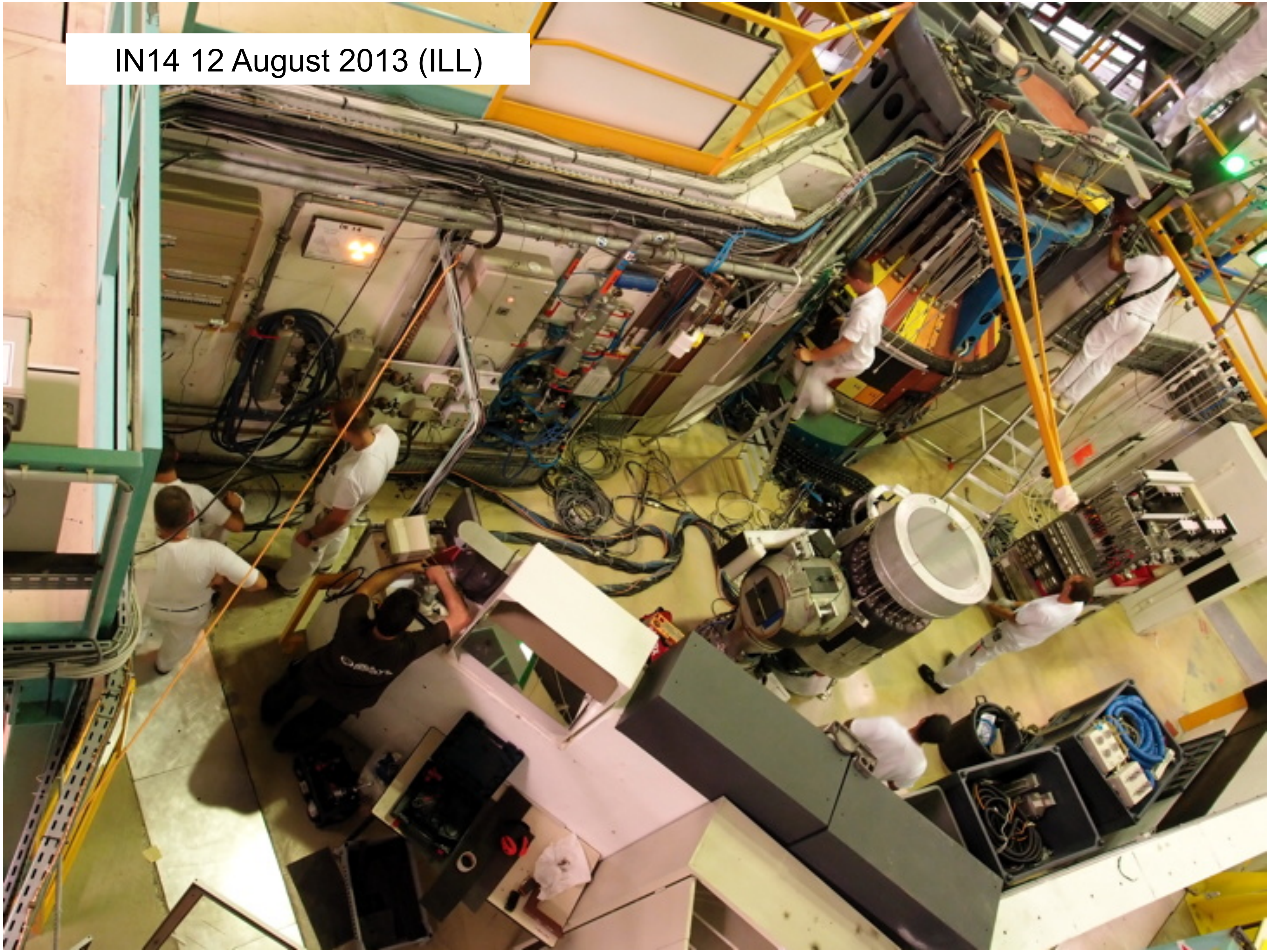


Polarized beam:

- **any configuration**
- **transmission 35%**
- **P ≈ 90 (<95) %**

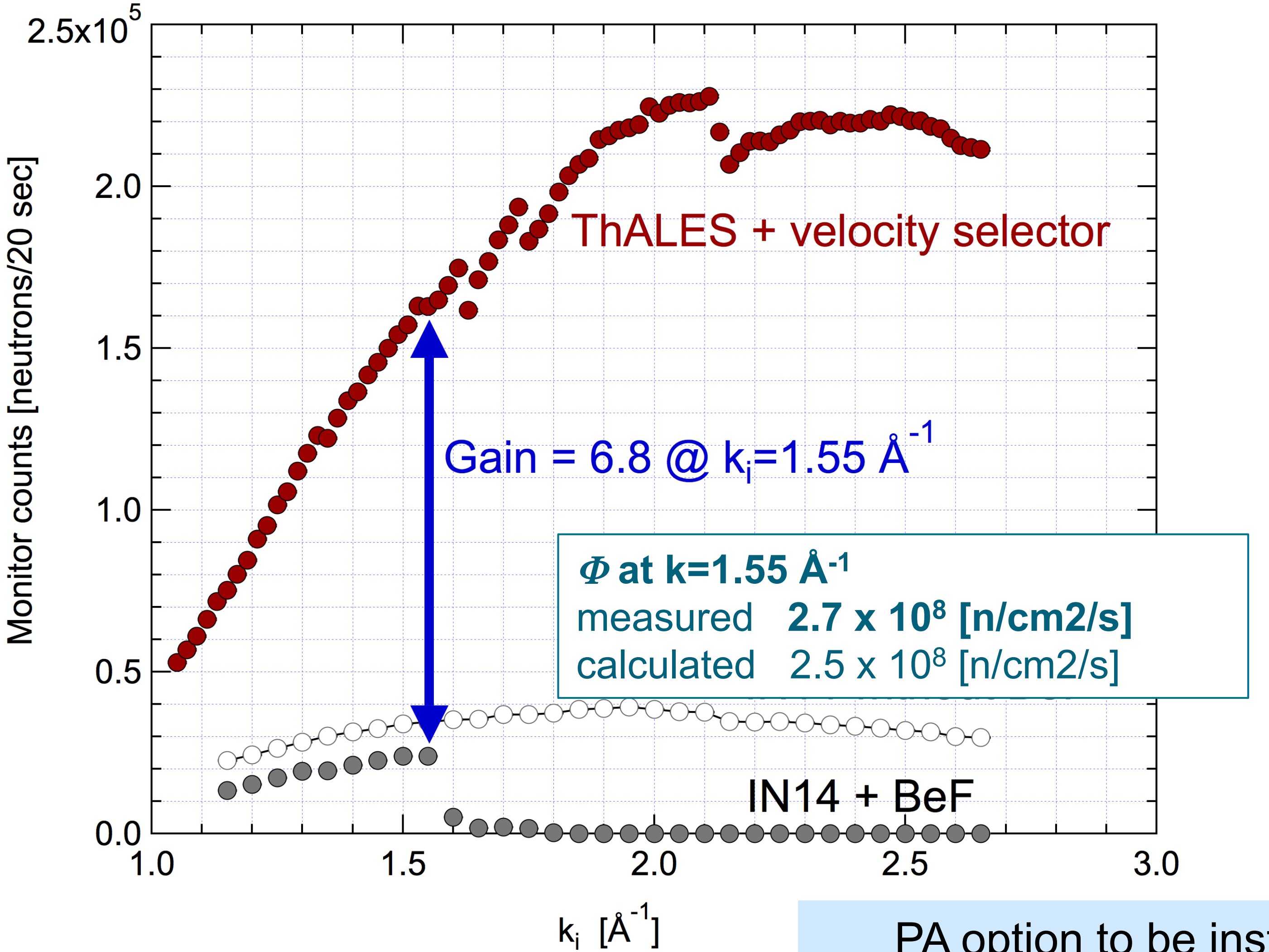
Jülich Centre for Neutron

IN14 12 August 2013 (ILL)



ThALES (ex-IN14)
12 September 2014 (ILL)

ThALES flux



PA option to be installed & commissioned in 2016