How far can we get with in-beam UCN production at the ESS?

Oliver Zimmer

Nordita workshop on Particle Physics with Neutrons at the ESS, Stockholm, 10-14 December

“In-beam” (in contra-distinction from “in-pile”) means:

UCN source is installed in a cold neutron beam far from a moderator

Advantages of “in-beam” w.r.t. “in-pile”:

- Lower radiation level
  - $\rightarrow$ lower cooling power required
  - $\rightarrow$ low temperature (down to 0.5 K) attainable
  - $\rightarrow$ access for UCN reflectors for maximum UCN density
- Experiment can be close to source or even “in-situ”
  - $\rightarrow$ low UCN transport losses (see proposed nEDM searches)
- Low backgrounds
- Easy access to the source for troubleshooting

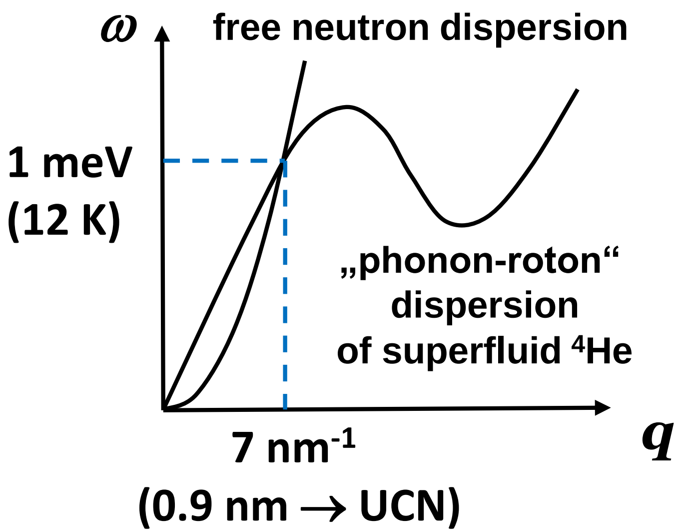
Disadvantages:

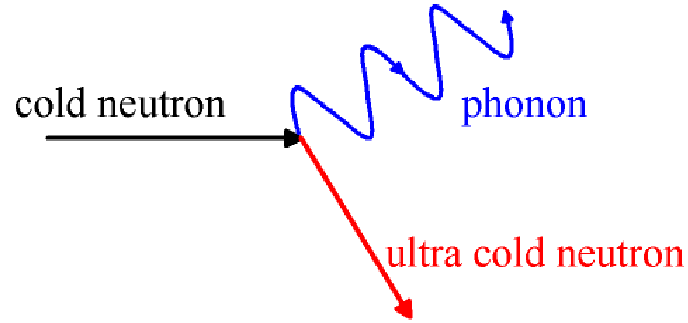
Cold neutrons from limited solid angle used for conversion to UCNs

- $\rightarrow$ lower UCN production rate
- $\rightarrow$ lower UCN fluxes
- $\rightarrow$ lower total UCN numbers in big vessels

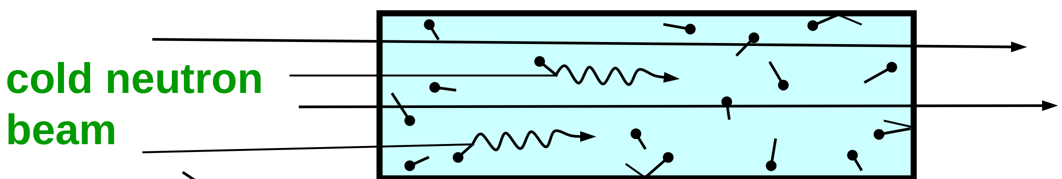
UCN production in superfluid He

R. Golub, J.M. Pendlebury, PL **53A** (1975) 133





T [K]	τ_{max} [s]
1	100
0.8	310
0.7	510
0.5	820
0	880



converter $\sigma_{capture}$ (**^{4}He) = 0**

$$\rho_{UCN} = P\tau$$

$$\tau^{-1} = \tau^{-1}_{decay} + \tau^{-1}_{upscattering} + \tau^{-1}_{capture} + \tau^{-1}_{wall\ losses}$$

→ need T < 0.5 - 0.6 K
and low-loss walls
(e.g. magnetic reflector)

Facts about the planned ESS moderator (Ken Andersen):

Moderator brilliance $\frac{d\Phi}{d\lambda d\Omega}$ at 9 Å:

$1.3 \times 10^{13}\ \mathrm{s^{-1}cm^{-2}sr^{-1}Å^{-1}}$ (peak at 5 MW)

$5.2 \times 10^{11}\ \mathrm{s^{-1}cm^{-2}sr^{-1}Å^{-1}}$ (average at 5 MW)

Usable moderator surface:

$3(\mathrm{vertical}) \times 8(\mathrm{horizontal})\ \mathrm{cm}^2$

Beam extraction with mirrors:

Mirror made of natural nickel ($m = 1$):

$$1.73\ \mathrm{mrad/Å} \leftrightarrow 15.6\ \mathrm{mrad/9Å} \leftrightarrow \Omega = 2.4 \times 10^{-4}\ \mathrm{sr}$$

UCN production rate density at ESS moderator surface:

$$\dot{\rho} \approx 6.2\ \mathrm{s^{-1}cm^{-3}} \times m^2 \qquad \dot{\rho} \approx 5 \times 10^{-8}\ \mathrm{Å cm^{-1}} \left.\frac{d\Phi}{d\lambda}\right|_{9Å}$$

For comparison, at ILL:

H172B monochromatic beam (SUN-2):

$\approx 5\ \mathrm{s}^{-1}\mathrm{cm}^{-3},\ 8 \times 8\ \mathrm{cm}^2$

H523 (SuperSUN):

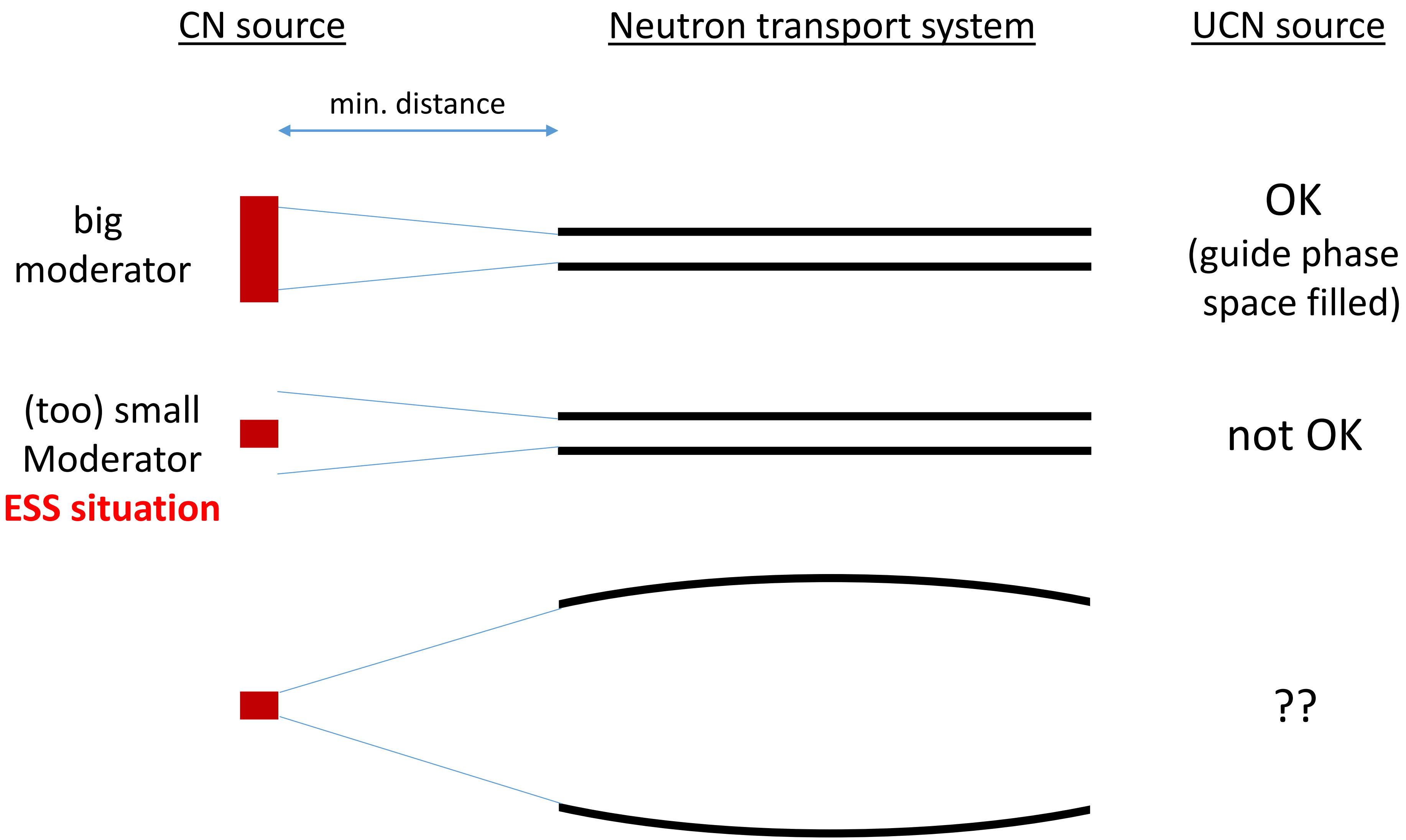
$\approx 15\ \mathrm{s}^{-1}\mathrm{cm}^{-3},\ \varnothing\, 7\ \mathrm{cm}$

Challenges for ESS in-beam UCN source:

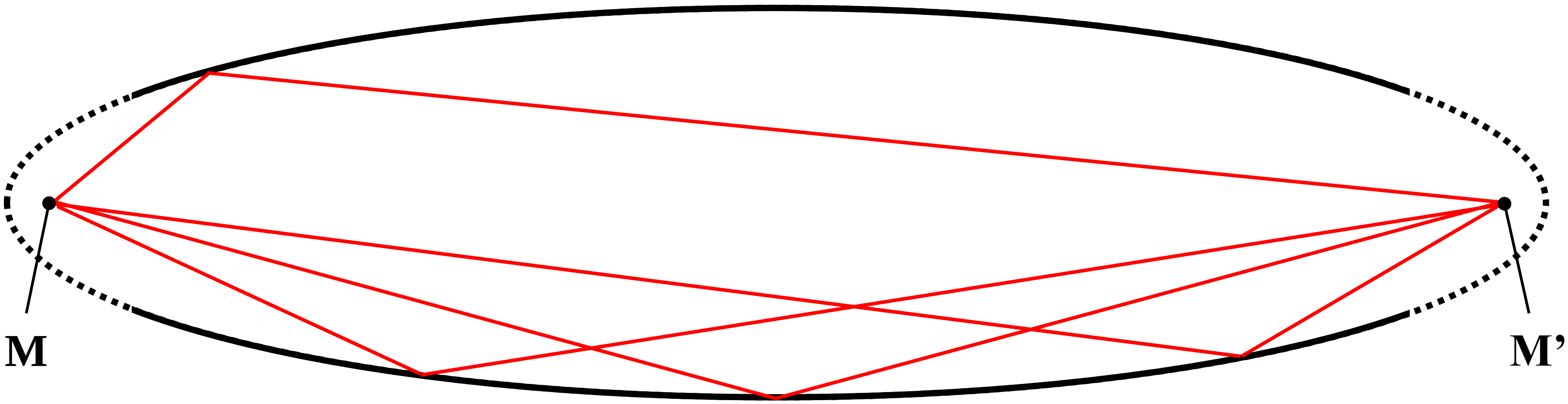
(a) Need high brilliance transfer from moderator to UCN source for $\dot{\rho}$ to come close to $6.2\ \mathrm{s}^{-1}\mathrm{cm}^{-3} \times m^2$

(b) A larger and colder moderator would increase the total number of UCNs after accumulation (which is ∝ source volume); the moderated spectrum would best peak at $9\ \mathrm{Å}$.

(a) How to deliver the neutrons to the UCN source?



Is an elliptic guide an imaging device?



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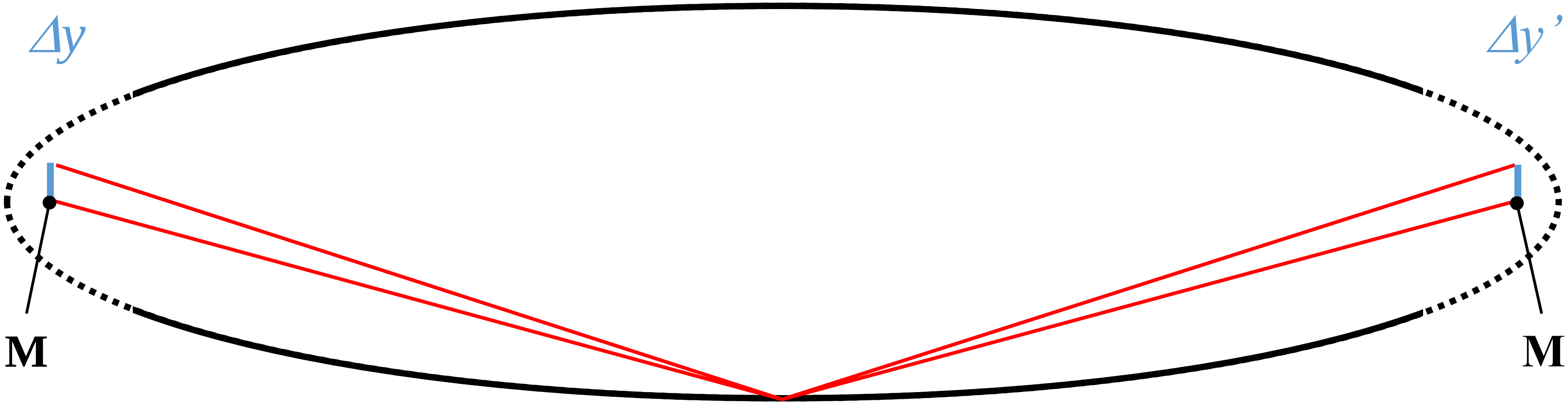

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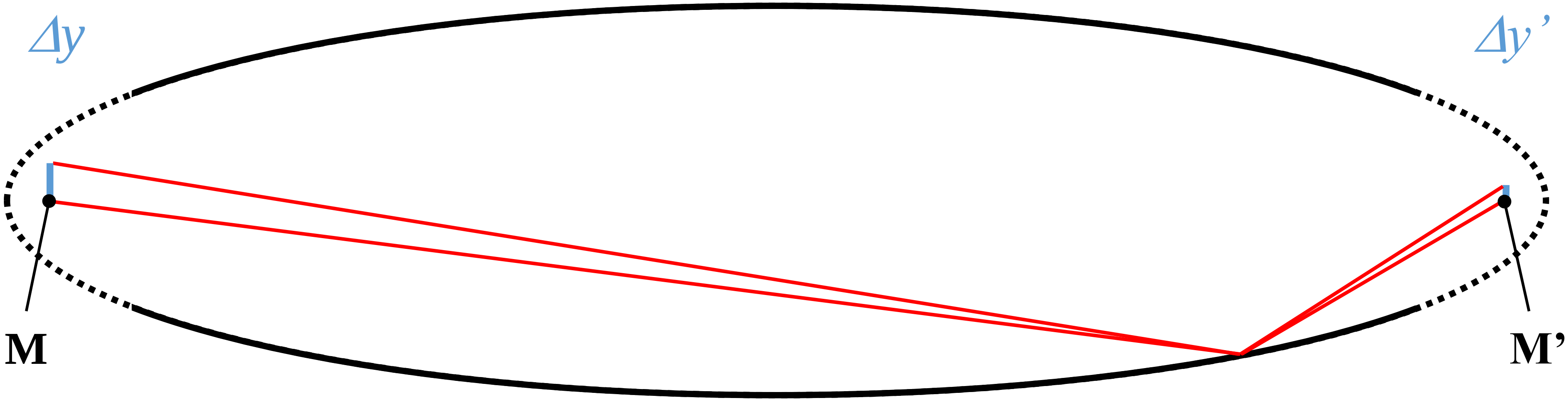

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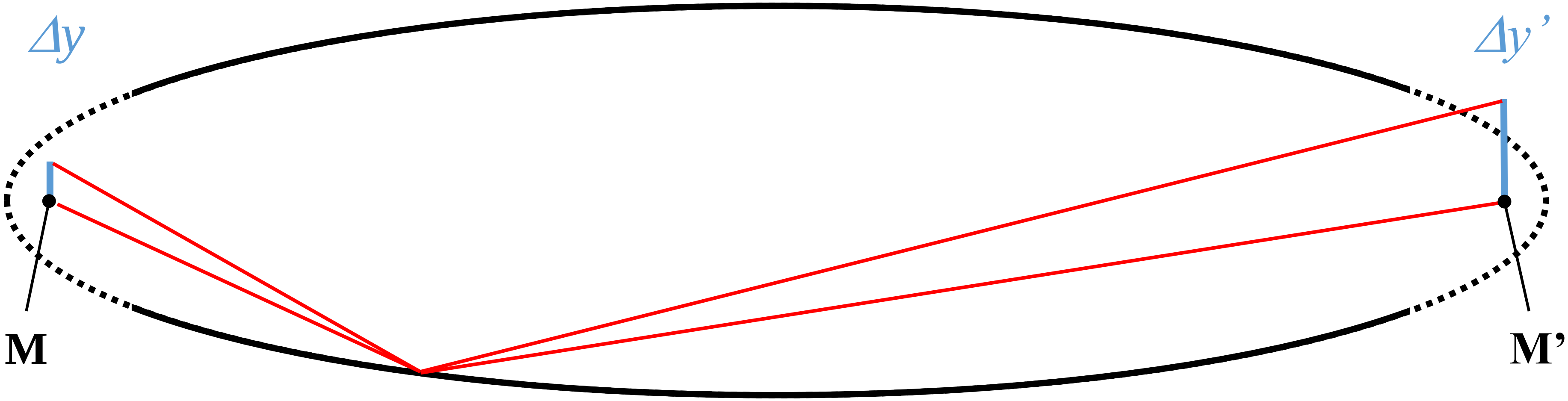

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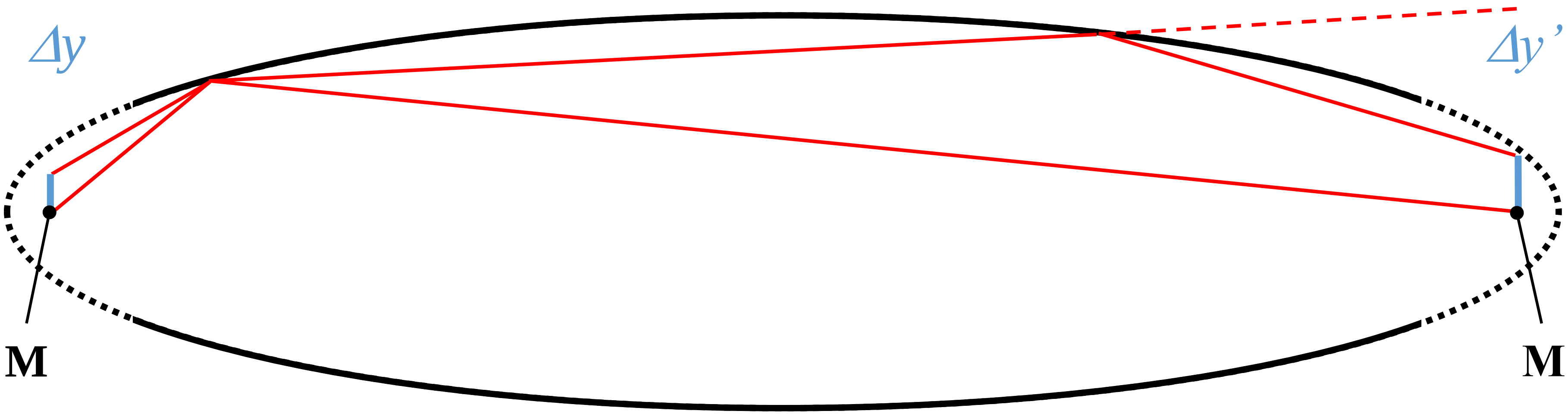

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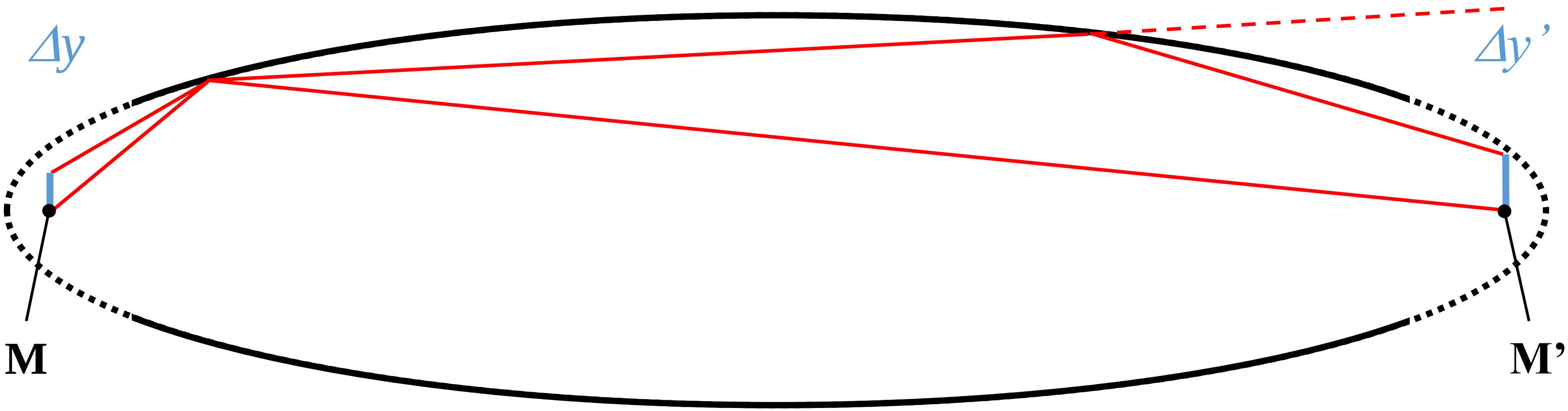

Cussen, Nekrassov, Zendler and Lieutenant:

Multiple reflections in elliptic neutron guides, NIM A 705 (2013) 121

"Transport of neutrons by realistic elliptic guides usually involves many reflections, contrary to the usual expectations."

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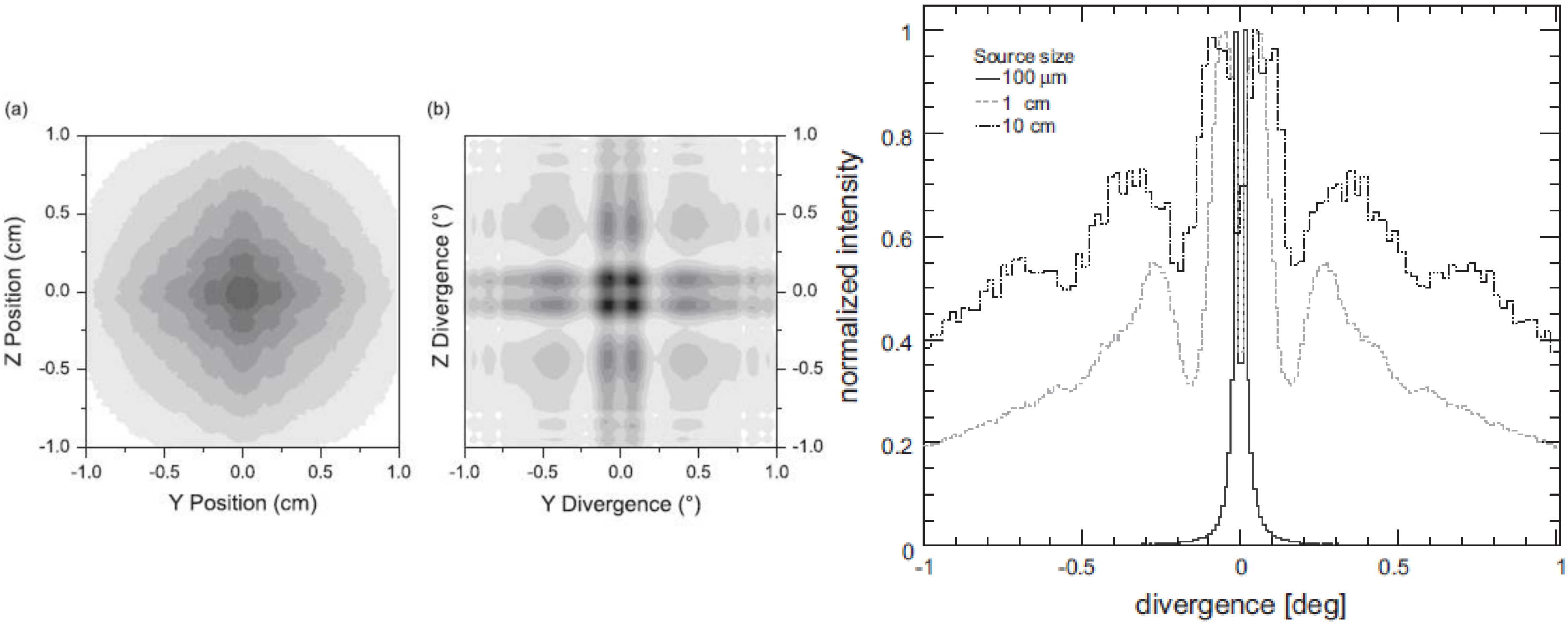

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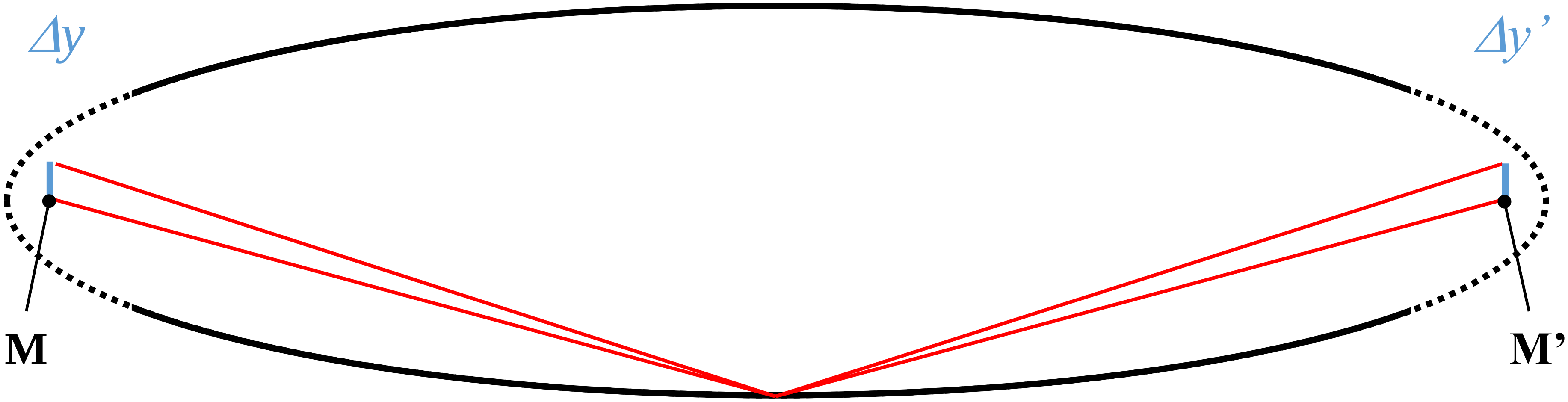
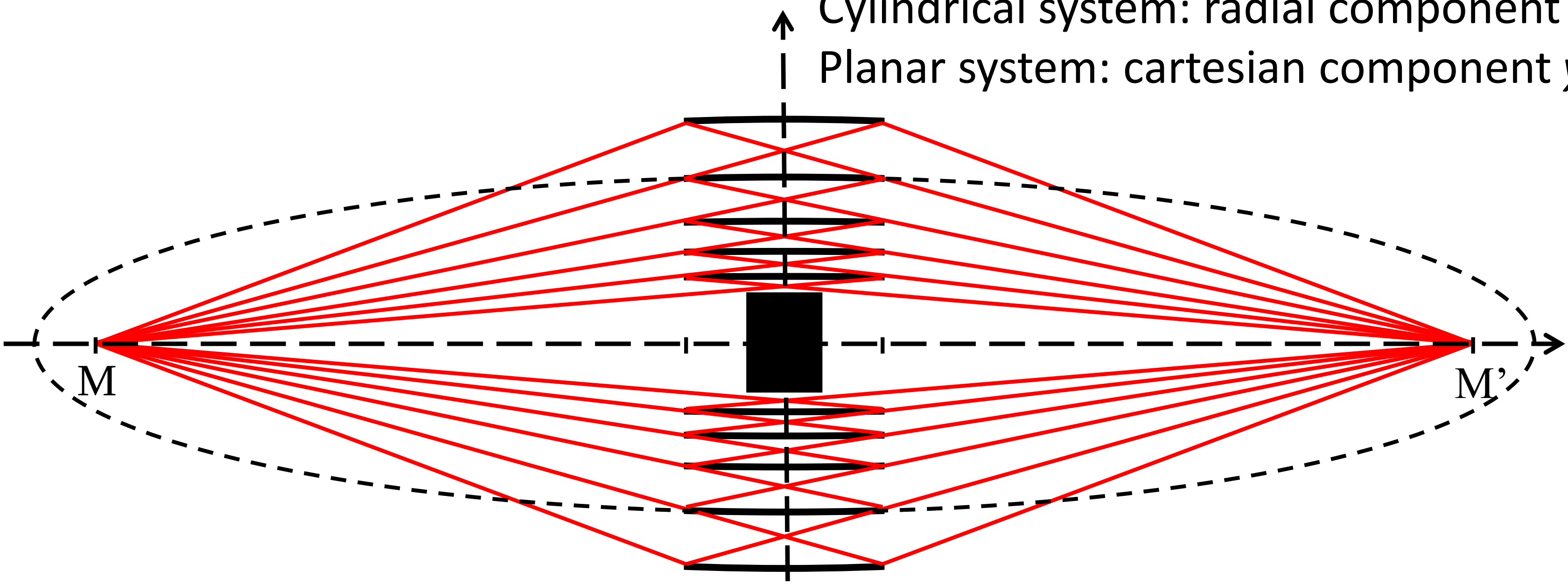


In general NO,

but YES, if we limit the reflection area on the mirror arXiv:1611.07353

Multi-mirror imaging optics for low-loss transport of divergent neutron beams and tailored wavelength spectra



Fills large solid angle from small source

Single reflections with well-defined reflection angles

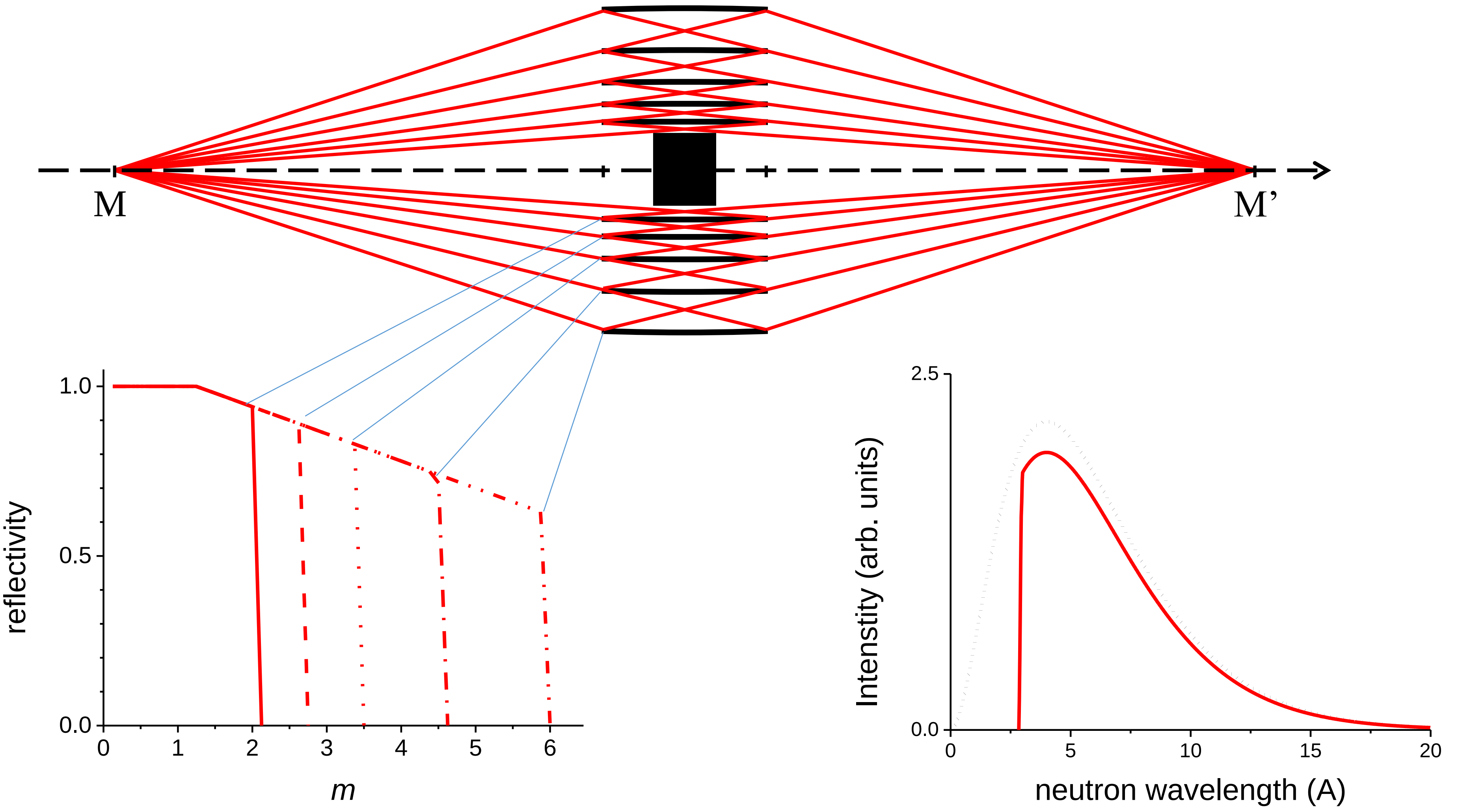
$\rightarrow$ no garland reflections

$\rightarrow$ beam divergence (q-resolution) adjustable with scrapers

$\rightarrow$ pure spectra adaptable to need of instrument

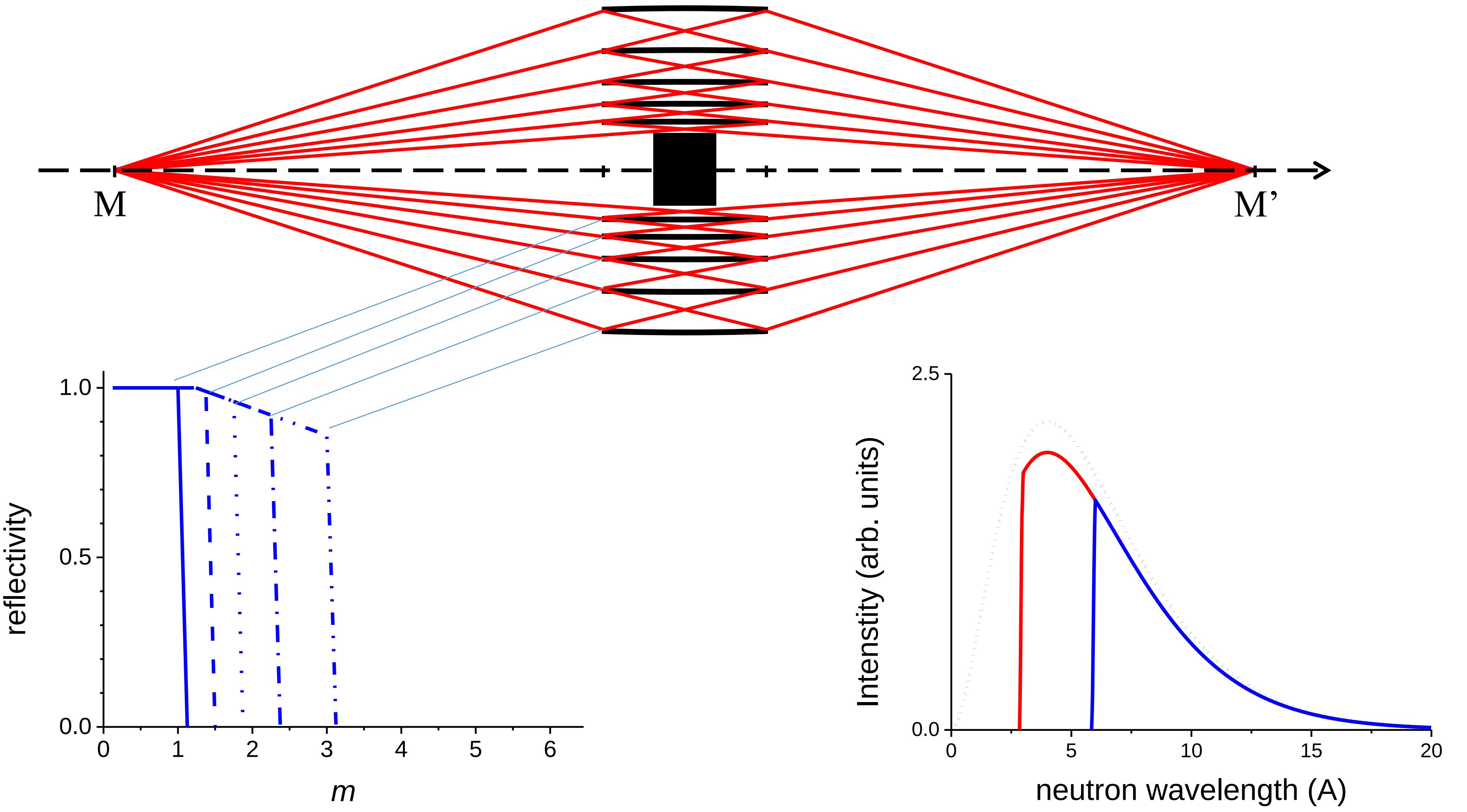
Broadband supermirrors (polarising or non-polarising)

- *m*-value tuning to a common short-wavelength cutoff:



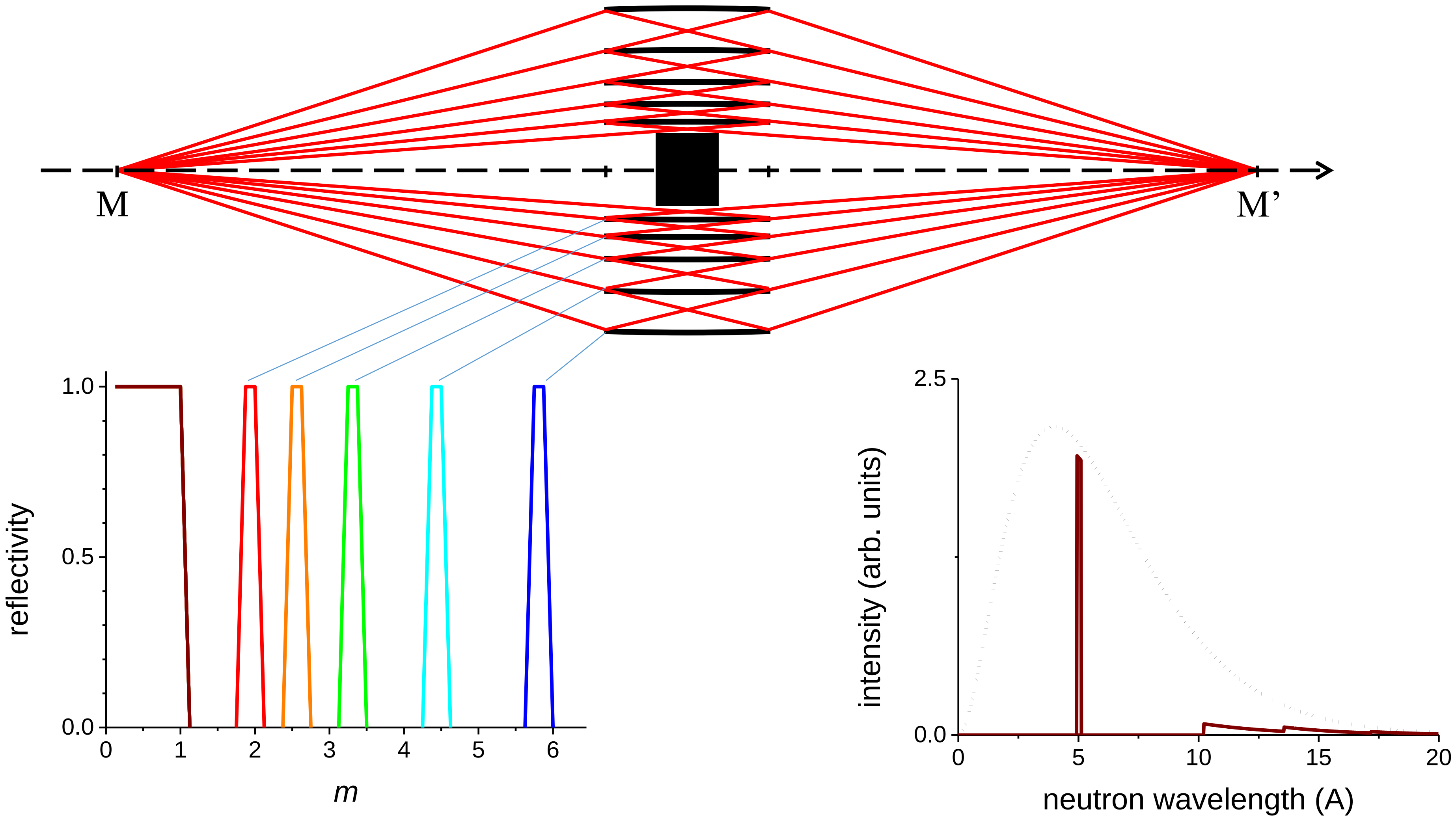
Broadband supermirrors (polarising or non-polarising)

- *m*-value tuning to a <u>common short-wavelength cutoff</u>:

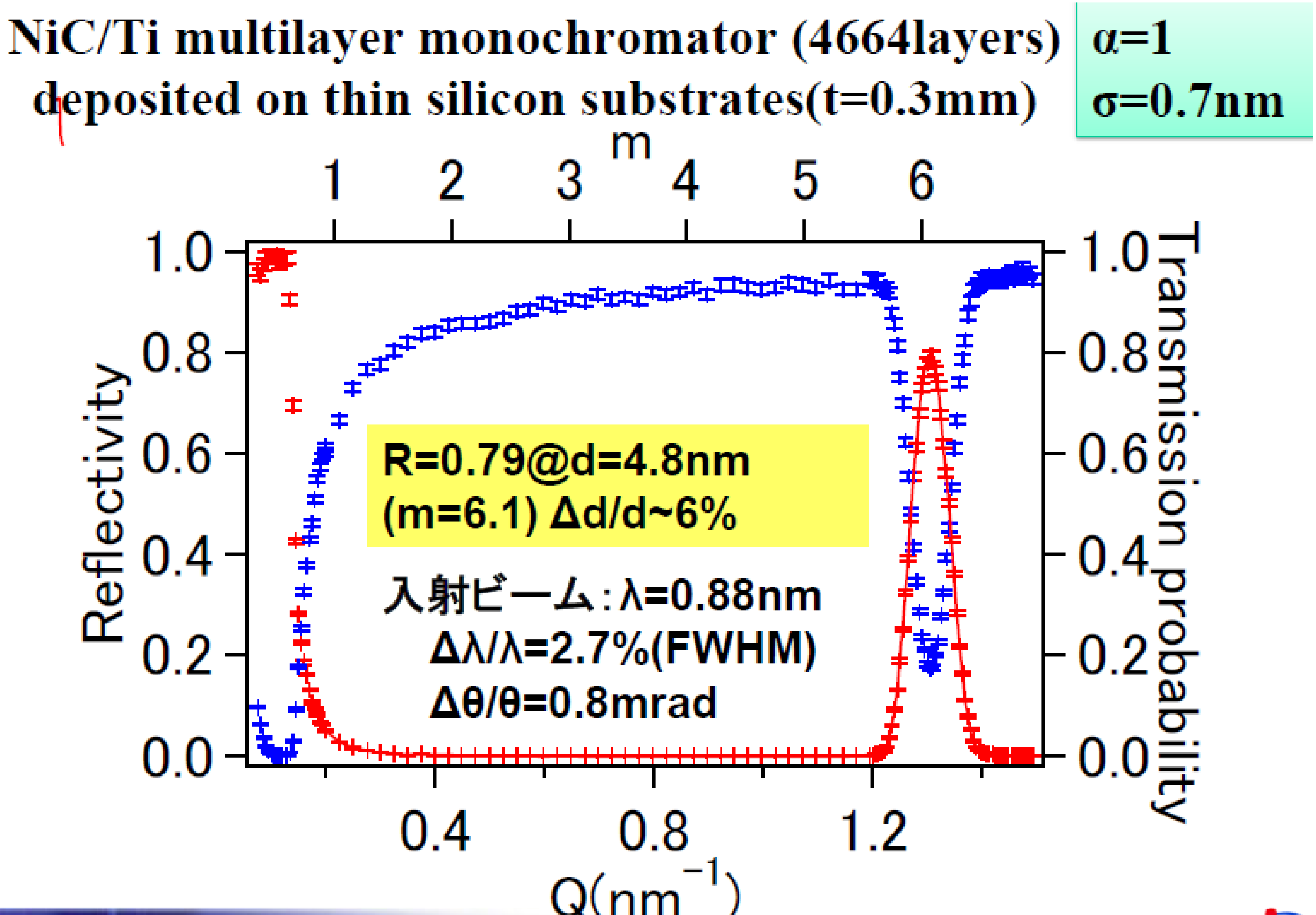


Bandpass supermirrors (polarising or non-polarising)

- monochromation to a common wavelength band:

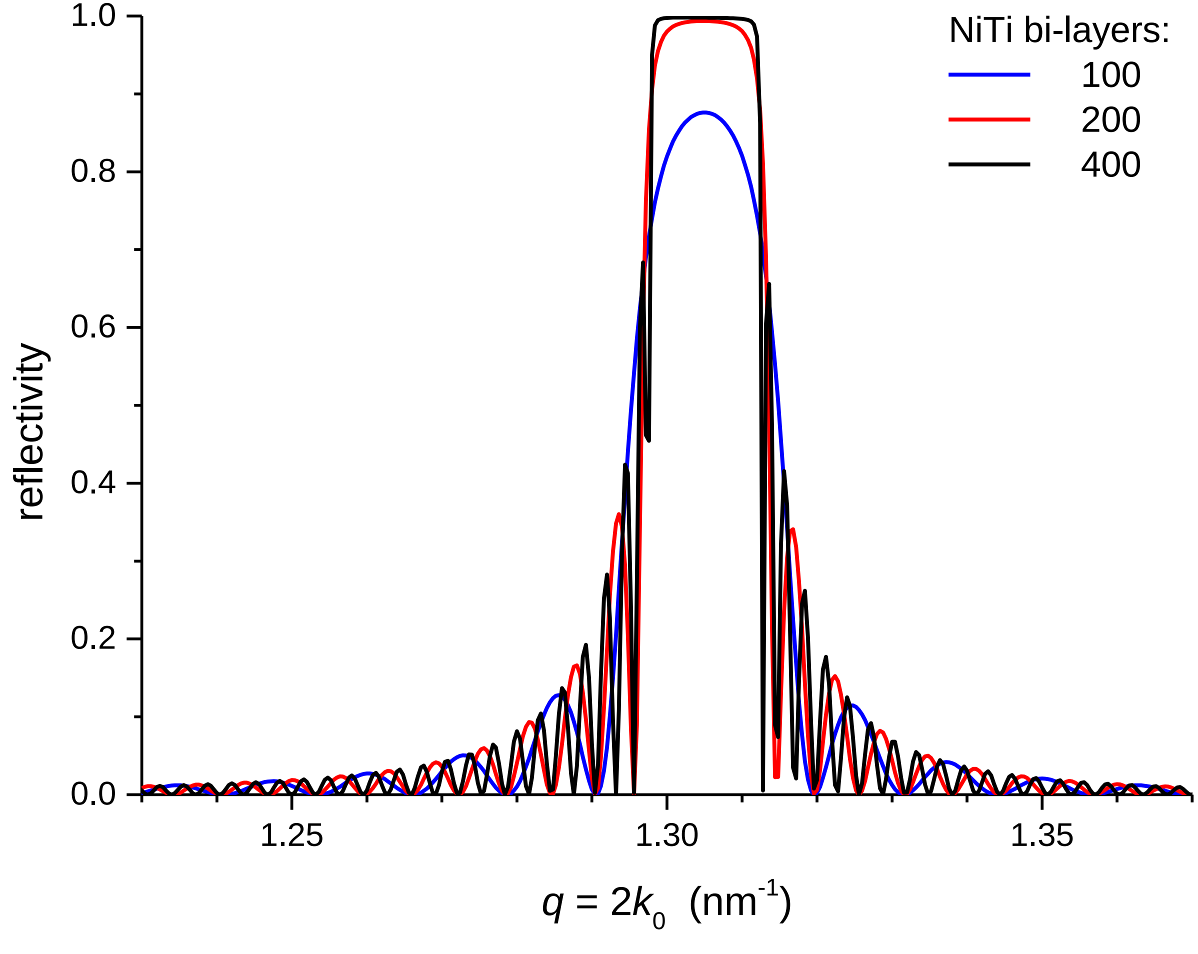


Masahiro Hino:



Sergei Masalovich, NIM A 705 (2013) 121:

Analysis and design of multilayer structures for neutron monochromators and supermirrors



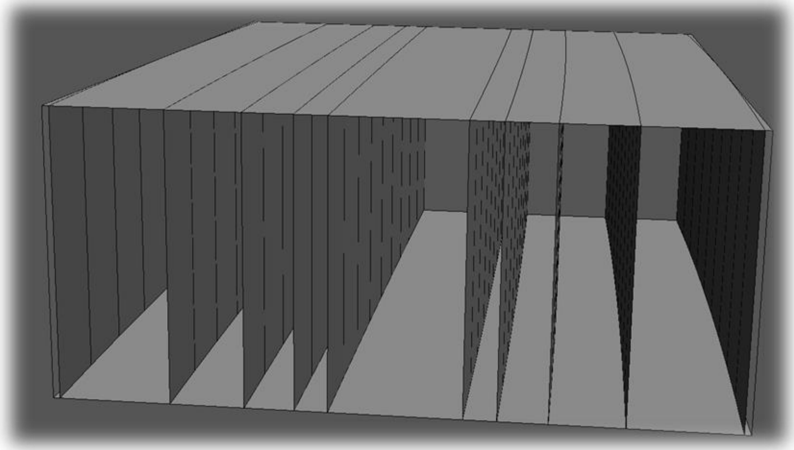
$m = 6$

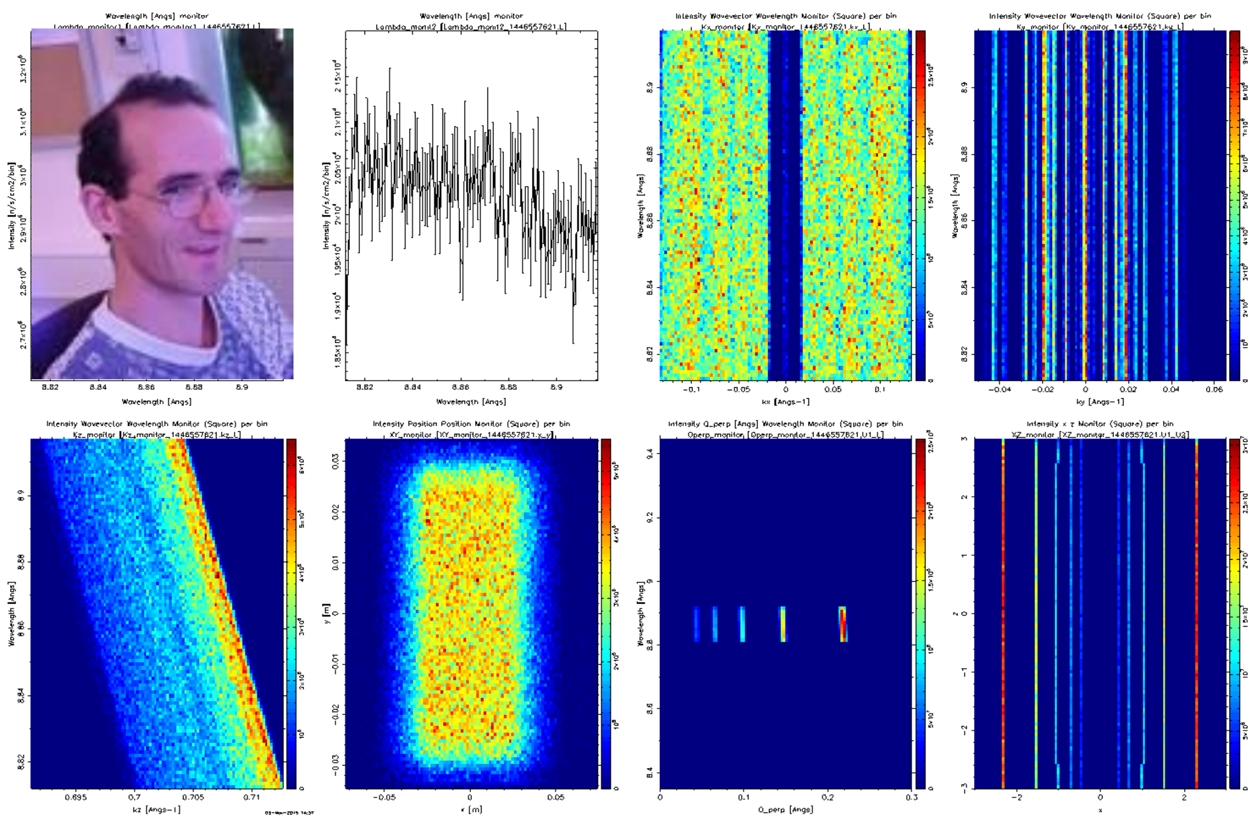
Quarter-wave layers can be adapted to select the width of the plateau reflectivity

Imaging works!

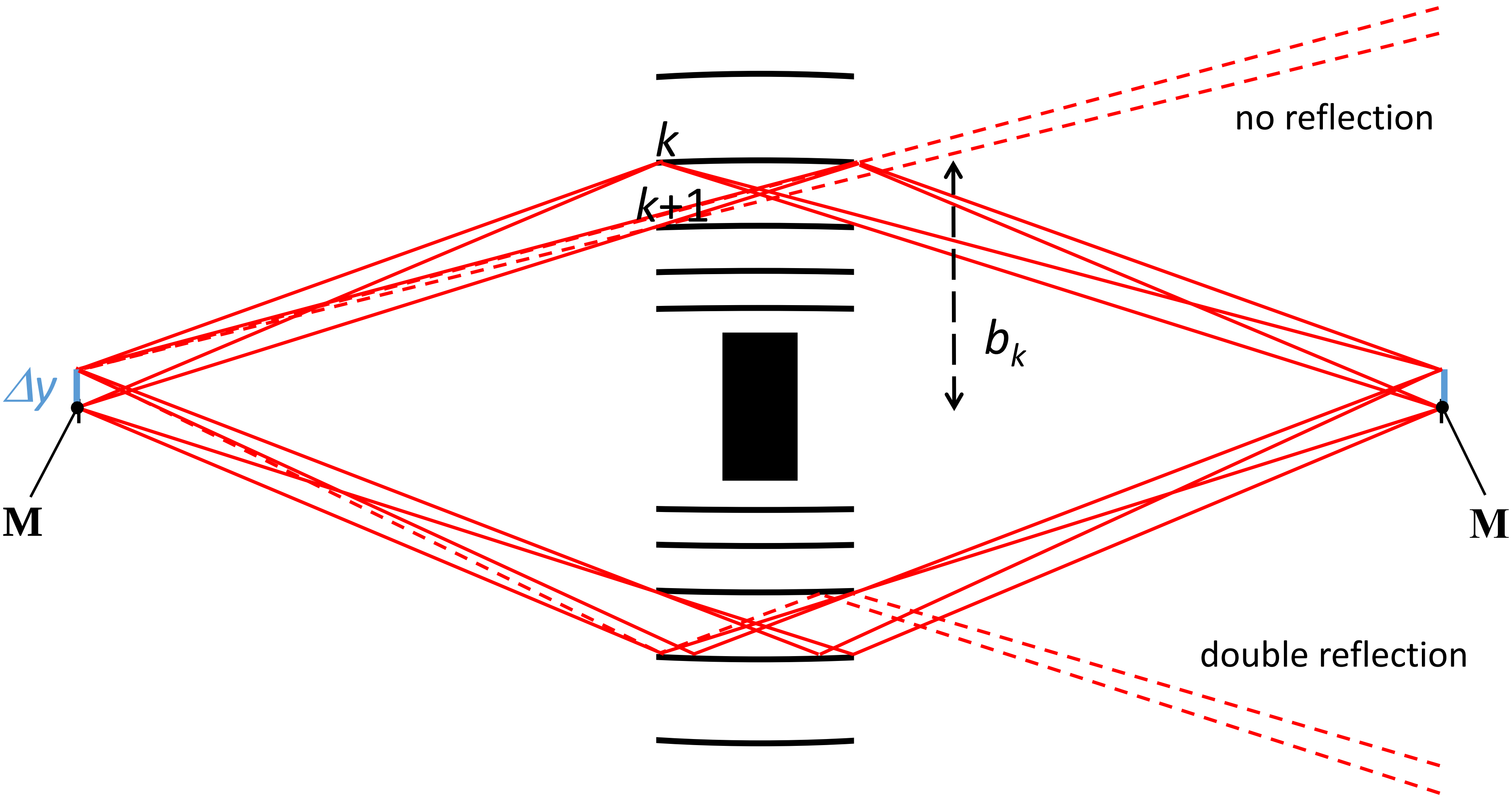
A planar elliptic multi-mirror already available as a McStas component

thanks to Emmanuel Farhi





Geometrical neutron losses due to finite source size



These losses are ~ $\Delta y/b_k$ and hence largest for the innermost mirrors

Examplary system for 9 Å for a He-II UCN source
(MM' = 30 m, mirror length = 2 m):

k	$y_k\,(\pm l)$ [m]	b_k [m]	a_k [m]	$\xi_k\,(-l)$ [^{0}]	$\theta_k\,(\pm l)$ [^{0}]	$\theta_k\,(0)$ [^{0}]	m_k
−2	2.3073	2.3123	15.1772	9.359	8.782	8.763	10
−1	2.0189	2.0233	15.1358	8.206	7.699	7.682	8.80
0	1.7665	1.7704	15.1041	7.192	6.746	6.731	7.70
1	1.5457	1.5491	15.0798	6.300	5.909	5.896	6.74
2	1.3525	1.3555	15.0611	5.518	5.175	5.163	5.90
3	1.1834	1.1860	15.0468	4.832	4.531	4.521	5.16
4	1.0355	1.0378	15.0359	4.230	3.967	3.958	4.51
5	0.9061	0.9081	15.0275	3.703	3.472	3.464	3.95
6	0.7928	0.7946	15.021	3.241	3.039	3.032	3.46
7	0.6937	0.6952	15.0161	2.837	2.660	2.654	3.02
8	0.6070	0.6083	15.0123	2.483	2.328	2.322	2.65
9	0.5311	0.5323	15.0094	2.173	2.037	2.032	2.32
10	0.4647	0.4658	15.0072	1.901	1.782	1.779	2.03
11	0.4066	0.4075	15.0055	1.664	1.560	1.556	1.78
12	0.3558	0.3566	15.0042	1.456	1.365	1.362	1.55
13	0.3113	0.3120	15.0032	1.274	1.194	1.192	1.36
14	0.2724	0.2730	15.0025	1.115	1.045	1.043	1.19
15	0.2384	0.2389	15.0019	0.975	0.914	0.912	1.04
16	0.2086	0.2090	15.0015	0.854	0.800	0.798	0.91
17	0.1825	0.1829	15.0011	0.747	0.700	0.699	0.80
18	0.1597	0.1600	15.0009	0.653	0.613	0.611	0.70

ESS moderator (Δy = 1.5 cm): losses < 10 % even for m < 1 mirrors

Advantages of this type of optics (in fact of more general interest than only for a UCN source):

- Efficient brilliance transfer from small moderator
- Small-wavelength cut-off
- Low backgrounds of unwanted neutrons at instrument
- Monochromation of primary beam possible
- *q*-resolution (divergence) adaptable by scrapers
- Mirrors far away from source $\rightarrow$ small radiation damage
- Practical: easier exchange of beam tubes
- Options: stack several planar systems with different properties

(b) Can we produce more very cold neutrons?

They would indeed be useful for everyone:

Neutron scattering community:

For pulsed VCN sources, the following gains can be assumed for resolution and intensity:

	resolution at fixed geometry	Intensity at fixed resolution
SANS	λ^{-1}	λ^{0}
Reflectometry	λ^{-1}	λ^{2}
TOF-INS	λ^{-3}	λ^{2}
NSE	λ^{-3}	λ^{2}-λ^{4}

From proceedings of workshop on application of a VCN source at Argonne, 2005

Particle physics community:

Counting statistics improvements e.g. for

- neutron-antineutron oscillation experiment
- beam neutron EDM experiment
- in-beam UCN source

Namiot's proposal (1974):

"phononless cooling of neutrons to extremely low temperatures"

Namiot, Sov. Phys. Dokl. 18, 481 (1974)

Cascaded neutron-deuteron spinflip scattering in a fully polarised medium in 30 T magnetic field

Energy transfer per nd spin flip collision: $\sim 0.1\ \mu\text{eV/T}$

Zeeman energy of unpaired electron: $116\ \mu\text{eV/T}$

Usable for cooling?

A suitable system should be paramagnetic because:

no dispersion $\rightarrow$ no kinematic restrictions $\rightarrow$ **scattering cascadable!**

…look for weakly absorbing paramagnetic species:

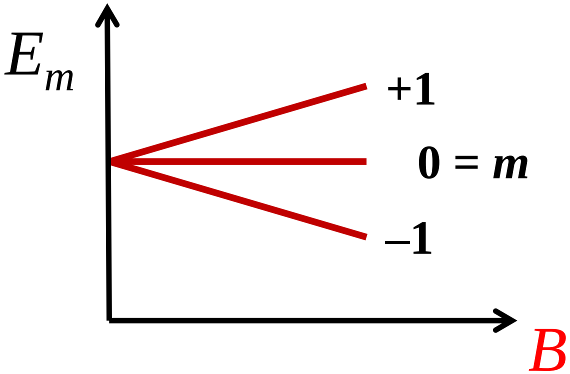
Species	S	g_- $(T \to 0)$	σ_a (mbarn)
electron	$1/2$	$1/3$	0
^2H	$1/2$	$1/3$	0.519 (7)
^1H	$1/2$	$1/3$	332.6 (7)
^{15}N	$3/2$	1	0.024 (8)
^{14}N	$3/2$	1	1910 (30)
$^{16}O_2$	1	$4/3$	2×0.10 (2)
natural O_2	1	$4/3$	2×0.19 (2)

0.12 meV/T

paramagnetic molecule !

Expect for Zeeman system:

0.12 meV/T



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$0.12\ \text{meV/T}$

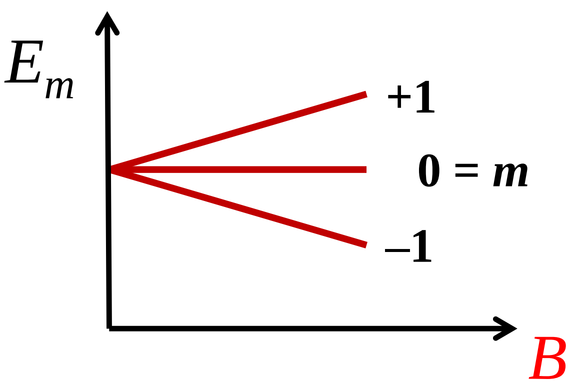
paramagnetic molecule !

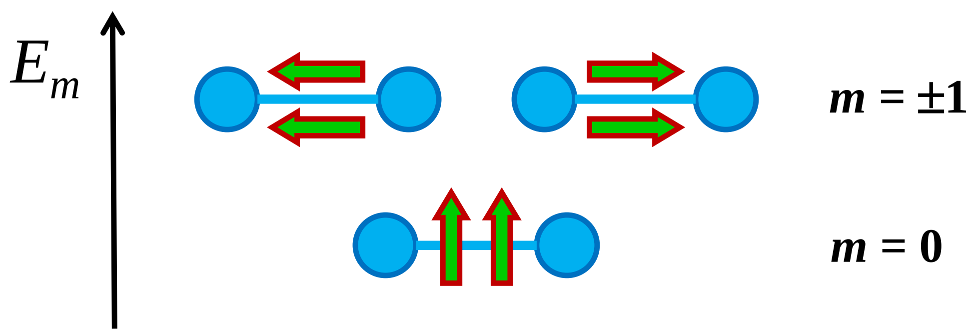
Expect for Zeeman system:

$$0.12\ \text{meV/T}$$

but O_2 has triplet zero-field splitting:

$$\approx 0.4\ \text{meV} = k_B \times \mathbf{4.6\ K}\,!$$





…and it's there **without *B*-field !**

Solid oxygen is antiferromagnetic at low T (and dangerous)

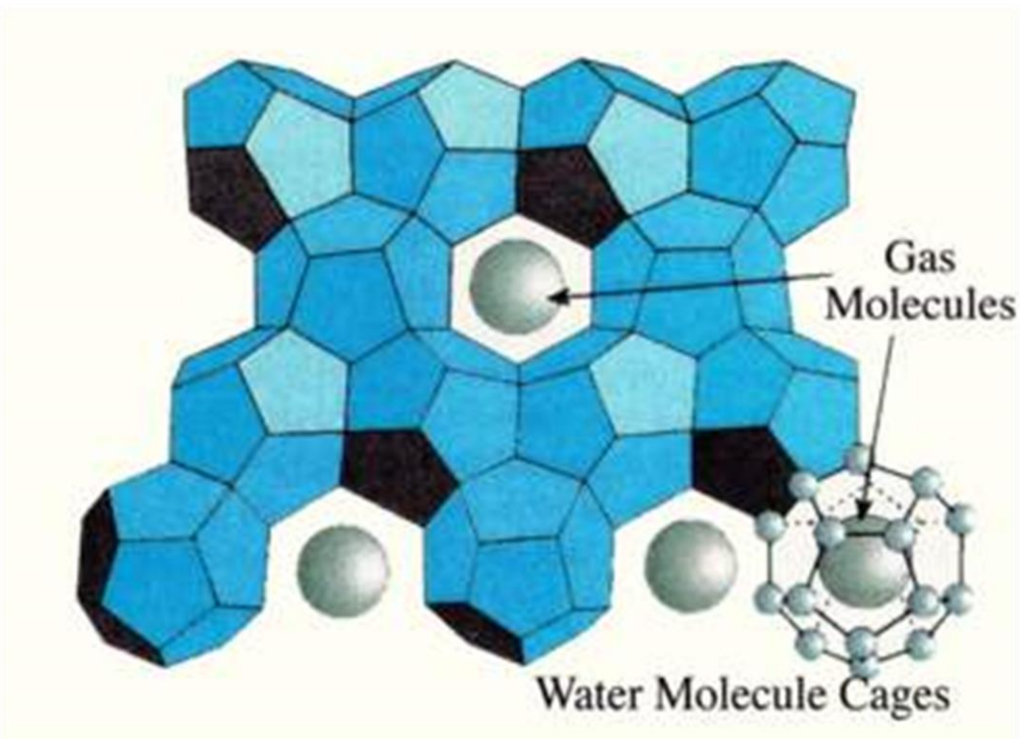
O_2 hydrate clathrate:

O_2 density $\approx 4.2 \times 10^{21}/\mathrm{ccm}$ (90 % cage filling)

$\rightarrow$ stays paramagnetic at liq.He temperatures

$\rightarrow$ metastable (not explosive)

$\rightarrow$ neutron survival $> 0.1\ s$ if fully deuterated



Methane hydrate clathrate

Inelastic scattering cross section for paramagnets?

Start from first-order time-dependent perturbation theory:

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\eta\to\eta'} = \frac{k'}{k}\left(\frac{m_{\mathrm{n}}}{2\pi\hbar^2}\right)^2 \sum_{\lambda\lambda'} p_\lambda \left|\langle \mathbf{k}'\eta'\lambda'\right| \mathcal{H}_{\mathrm{m}}(\mathbf{r}) \left|\mathbf{k}\eta\lambda\rangle\right|^2 \delta\left(E_{\lambda'} - E_\lambda + E' - E\right)$$

• • •

to arrive at (for molecular oxygen with zero-field splitting):

$$\boxed{\Sigma^{\pm}\left(E \to E'\right) = n_{\mathrm{pc}}\sigma_{\mathrm{m}}\frac{k'}{k}g_{\pm}(T)\, f_{\pm}(E)\,\delta\left(E \pm E^{*} - E'\right)}$$

$$\sigma_{\mathrm{m}} = 4\pi b_{\mathrm{m}}^2 = 3.66\ \mathrm{barn} \qquad g_{+}(T \to 0) \to 0$$

$$g_{-}(T \to 0) \to 4/3$$

5 Appendix: Neutron scattering cross sections

In this appendix we derive the inelastic neutron scattering cross sections needed for the analysis of neutron conversion and cascade cooling by paramagnetic centers. The first part covers simple Zeeman systems of atomic or ionic paramagnetic centers without zero-field splittings. The second part deals with the triplet state of molecular oxygen without external magnetic field. The analysis follows standard procedures presented in textbooks on neutron scattering theory [28, 29] up to the point, where we evaluate the thermal averages of time-dependent spin operators without neglecting energy transfers to or from the neutron. While this can in fact be easily accomplished for paramagnetic systems, expressions for such inelastic cross sections seem not to appear in the literature, probably because the usually small energy transfer in the diffuse scattering associated with an electron spin flip is only of limited interest for structural studies. As argued in the main text, the inelastic neutron scattering due to the zero-field splitting in oxygen seems to have already been observed in two experimental studies [44, 45], where it was however temptatively interpreted as a crystal field effect. Also for this reason a comprehensive presentation of the corresponding cross sections seems useful.

5.1 Spin dependent neutron scattering cross sections for a Zeeman system without zero-field splittings

We analyze neutron scattering by atomic or ionic paramagnetic centers polarized in a static external magnetic field and derive partial cross sections for electron spin flip and electron non-spin flip processes, with and without neutron spin flip. We start from the double differential cross section for magnetic neutron scattering in first order time dependent perturbation theory, which is given by

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\eta\to\eta'} = \frac{k'}{k}\left(\frac{m_{\rm n}}{2\pi\hbar^2}\right)^2 \sum_{\lambda\lambda'} p_\lambda \left|\langle \mathbf{k}'\eta'\lambda'|\,\mathcal{H}_{\rm m}(\mathbf{r})\,|\mathbf{k}\eta\lambda\rangle\right|^2 \delta\left(E_{\lambda'}-E_\lambda+E'-E\right). \tag{46}$$

Here a neutron with mass $m_{\rm n}$, wavevector $\mathbf{k}$, kinetic energy E and quantum number η for the projection of the neutron spin onto the z axis defined by the external, static magnetic field $\mathbf{B}_0 = (0,0,B_0)$, is scattered into a final state with $\mathbf{k}'$, E' and η'. The probed system undergoes a transition from an initial state $|\lambda\rangle$ characterized by a set of quantum numbers λ and energy E_λ to a final state characterized by λ' and energy $E_{\lambda'}$. The cross section in Eq. (46) includes a sum over final states λ' and thermal averaging over the initial states by means of statistical weight factors p_λ.

The Hamiltonian $\mathcal{H}_{\rm m}(\mathbf{r}) = -\boldsymbol{\mu}_{\rm n}\cdot\mathbf{B}(\mathbf{r})$ of the interaction of the neutron magnetic moment $\boldsymbol{\mu}_{\rm n} = g_{\rm n}\mu_{\rm N}\boldsymbol{\sigma}/2$ with the local magnetic field $\mathbf{B}(\mathbf{r})$ in the paramagnetic system has matrix elements between plane wave states $\mathbf{k}$ and $\mathbf{k}'$ that can be expressed as

$$\mathcal{H}_{\rm m}(\boldsymbol{\kappa}) = \langle \mathbf{k}'\,|\mathcal{H}_{\rm m}(\mathbf{r})|\,\mathbf{k}\rangle = \frac{1}{2}\mu_0 g_{\rm n}\mu_{\rm N} g_{\rm e}\mu_{\rm B}\boldsymbol{\sigma}\cdot\mathbf{Q}_\perp(\boldsymbol{\kappa}), \tag{47}$$

where μ_0 is the magnetic vacuum permeability, $g_{\rm n}\approx -3.826$ is the g-factor of the neutron, $\mu_{\rm N}$ is the nuclear magneton, $g_{\rm e}\approx -2.002$ is the g-factor of the electron, $\mu_{\rm B}$ is the Bohr magneton, $\boldsymbol{\sigma}/2$ is the neutron spin in units of $\hbar$ expressed by the vector of Pauli matrices $\boldsymbol{\sigma} = (\sigma_x,\sigma_y,\sigma_z)$, and the scattering vector

$$\boldsymbol{\kappa} = \mathbf{k}-\mathbf{k}' \tag{48}$$

is the momentum transfer to the scattering system in units of $\hbar$. The vector

$$\mathbf{Q}_\perp = \widehat{\kappa}\times(\mathbf{Q}\times\widehat{\kappa}) = \mathbf{Q}-(\mathbf{Q}\cdot\widehat{\kappa})\,\widehat{\kappa} \tag{49}$$

is the component of a vector $\mathbf{Q}$ perpendicular to $\boldsymbol{\kappa}$ ($\widehat{\kappa}$ is the unit vector of $\boldsymbol{\kappa}$), which can be shown to be in general proportional to the Fourier transform of the atomic magnetization $\mathbf{M}(\mathbf{r})$ due to both, spin and orbital angular momentum of the unpaired electrons. We can limit our attention to the case where unpaired electrons are located close to equilibrium positions of paramagnetic centers, and where individual electron spins of the center j couple to a total spin $\mathbf{S}_j$ with quantum number S. For the weakly absorbing species quoted in Table 1 the total orbital angular momentum $\mathbf{L}_j$ vanishes. For low-energy neutron scattering S is a conserved quantum number while its z component, characterized by a quantum number m, may change by one unit. Under these circumstances the vector $\mathbf{Q}$ can be shown to take the form

$$\mathbf{Q} = \sum_j \mathbf{Q}_j = \sum_j F_j(\kappa)\exp(i\boldsymbol{\kappa}\cdot\mathbf{R}_j)\,\mathbf{S}_j, \tag{50}$$

wherein $\mathbf{R}_j$ denotes the position of the jth paramagnetic center and

$$F_j(\kappa) = \int \widetilde{s}_j(\mathbf{r})\exp(i\boldsymbol{\kappa}\cdot\mathbf{r})\,d^3r \tag{51}$$

is the magnetic form factor with $\widetilde{s}_j$ denoting the density of unpaired electrons of the jth ion, divided by their number, so that $F_j(0) = 1$.

The cross section is given in Eq. (46) for specific transitions between neutron spin states $|+\rangle$ and $|-\rangle$ with respect to the external magnetic field. From the standard representation of the Pauli matrices the corresponding matrix elements follow as

$$\langle +|\,\boldsymbol{\sigma}\cdot\mathbf{Q}_\perp\,|+\rangle = Q_{\perp z}, \qquad \langle -|\,\boldsymbol{\sigma}\cdot\mathbf{Q}_\perp\,|-\rangle = -Q_{\perp z}, \tag{52}$$

and

$$\langle -|\,\boldsymbol{\sigma}\cdot\mathbf{Q}_\perp\,|+\rangle = Q_{\perp x}+iQ_{\perp y}, \qquad \langle +|\,\boldsymbol{\sigma}\cdot\mathbf{Q}_\perp\,|-\rangle = Q_{\perp x}-iQ_{\perp y}, \tag{53}$$

with the first (second) pair describing transitions without (with) neutron spin flip. Considering first the cross sections for magnetic neutron spin flip scattering, we use Eqs. (47) and (53) in Eq. (46) and write

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\pm\to\mp} = b_{\rm m}^2\frac{k'}{k}\sum_{\lambda\lambda'} p_\lambda \langle\lambda|\,Q_{\perp x}^\dagger \mp iQ_{\perp y}^\dagger\,|\lambda'\rangle\langle\lambda'|\,Q_{\perp x}\pm iQ_{\perp y}\,|\lambda\rangle\,\delta\left(E_{\lambda'}-E_\lambda+E'-E\right), \tag{54}$$

where

$$b_{\rm m} = \frac{1}{2}\mu_0 g_{\rm n}\mu_{\rm N} g_{\rm e}\mu_{\rm B}\frac{m_{\rm n}}{2\pi\hbar^2} = 5.404\ \text{fm} \tag{55}$$

is the magnetic scattering length. Continuing to follow the standard procedure to evaluate the cross section, the δ function is expressed as

$$\delta\left(E_{\lambda'}-E_\lambda+E'-E\right) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}\exp\left(i\left(E_{\lambda'}-E_\lambda\right)t/\hbar\right)\exp\left(i\left(E'-E\right)t/\hbar\right)dt. \tag{56}$$

Since $|\lambda\rangle$ are eigenstates of the Hamiltonian $\mathcal{H}_0$ of the system,

$$\exp(i\mathcal{H}_0 t/\hbar)\,|\lambda\rangle = \exp(iE_\lambda t/\hbar)\,|\lambda\rangle. \tag{57}$$

One can define time dependent operators as

$$Q_{\perp\alpha}(t) = \exp(i\mathcal{H}_0 t/\hbar)\,Q_{\perp\alpha}\exp(-i\mathcal{H}_0 t/\hbar), \tag{58}$$

where $\alpha = x, y, z$ are cartesian coordinates with the z axis pointing along the external magnetic field. Using Eq. (50) with this definition, one can write

$$Q_{\perp\alpha}(t) = \sum_j F_j(\kappa) \exp(i\kappa \cdot \mathbf{R}_j(t)) S_{\perp j\alpha}(t), \tag{59}$$

where

$$\mathbf{S}_{\perp j}(t) = \mathbf{S}_j(t) - (\mathbf{S}_j(t) \cdot \widehat{\kappa}) \widehat{\kappa}, \tag{60}$$

in analogy to Eq. (49). Under the usual assumption that the orientations of the electron spins do not affect positions and motion of the nuclei, the thermal averages can be factorized for the nuclear coordinates and electron spins. Using also the closure relation $\sum |\lambda'\rangle \langle\lambda'| = 1$ and denoting the thermal average $\sum p_\lambda \langle\lambda|...|\lambda\rangle$ by brackets $\langle...\rangle$, the cross section becomes

$$\begin{aligned}\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\pm\to\mp} &= \frac{b_{\mathrm{m}}^2}{2\pi\hbar}\frac{k'}{k}\int_{-\infty}^{\infty}\sum_{jj'}\langle\exp(-i\kappa\cdot\mathbf{R}_{j'})\exp(i\kappa\cdot\mathbf{R}_j(t))\rangle F_{j'}^*(\kappa) F_j(\kappa) \\ &\times \langle (S_{\perp j'x} \mp iS_{\perp j'y})(S_{\perp jx}(t) \pm iS_{\perp jy}(t))\rangle \exp(i(E'-E)t/\hbar)\, dt.\end{aligned} \tag{61}$$

It will be useful to employ the raising and lowering operators defined by

$$S_j^{\pm} = S_{jx} \pm iS_{jy}, \tag{62}$$

which fulfill the relation

$$S_{j'x}S_{jx}(t) + S_{j'y}S_{jy}(t) = \frac{1}{2}\left(S_{j'}^+ S_j^-(t) + S_{j'}^- S_j^+(t)\right). \tag{63}$$

For further evaluation of the spin operator products in the cross section one notes that for a paramagnetic system in an external magnetic field applied in z direction, the total z component of the electron spin is a constant of motion, and therefore

$$\sum_j [S_{jz}, \mathcal{H}_0] = 0. \tag{64}$$

The operators $S_j^{\pm}$ then change the z component of the total spin of the system by one unit so that

$$\left\langle S_{j'}^+ S_j^+(t)\right\rangle = \left\langle S_{j'}^- S_j^-(t)\right\rangle = 0, \tag{65}$$

and therefore also

$$\left\langle S_{j'x}S_{jy}(t) + S_{j'y}S_{jx}(t)\right\rangle = \frac{1}{2i}\left\langle S_{j'}^+ S_j^+(t) + S_{j'}^- S_j^-(t)\right\rangle = 0. \tag{66}$$

Also,

$$\left\langle S_{j'z}S_{jx}(t)\right\rangle = 0, \qquad \left\langle S_{j'z}S_{jy}(t)\right\rangle = 0, \tag{67}$$

and due to equivalence of the x and y axes,

$$\left\langle S_{j'x}S_{jx}(t)\right\rangle = \left\langle S_{j'y}S_{jy}(t)\right\rangle. \tag{68}$$

Since for a paramagnet there are no correlations between spins of different centers $j \neq j'$,

$$\left\langle S_{j'\alpha}S_{j\alpha}(t)\right\rangle = \langle S_{j'\alpha}\rangle\langle S_{j\alpha}\rangle + \delta_{jj'}\left(\langle S_{j\alpha}S_{j\alpha}(t)\rangle - \langle S_{j'\alpha}\rangle\langle S_{j\alpha}\rangle\right). \tag{69}$$

In presence of a static magnetic field in z direction, $\langle S_{jz}\rangle \neq 0$ but $\langle S_{jx}\rangle = \langle S_{jy}\rangle = 0$. The spin correlation functions entering the cross section are thus given by

$$\left\langle S_{j'x}S_{jx}(t)\right\rangle = \left\langle S_{j'y}S_{jy}(t)\right\rangle = \delta_{jj'}\langle S_xS_x(t)\rangle, \tag{70}$$

and

$$\left\langle S_{j'z}S_{jz}(t)\right\rangle = \langle S_z\rangle^2 + \delta_{jj'}\left(\langle S_zS_z(t)\rangle - \langle S_z\rangle^2\right), \tag{71}$$

where by omission of the index j we focus attention on a medium containing a single paramagnetic species without anisotropy effects due to electrostatic crystal fields. The cross section for neutron spin flip scattering thus becomes

$$\begin{aligned}\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\eta\neq\eta'} &= \frac{b_{\mathrm{m}}^2}{2\pi\hbar}\frac{k'}{k}\int_{-\infty}^{\infty}\sum_{jj'}\langle\exp(-i\kappa\cdot\mathbf{R}_{j'})\exp(i\kappa\cdot\mathbf{R}_j(t))\rangle |F(\kappa)|^2 \\ &\times \left[\delta_{jj'}\left(\frac{1}{4}(1+\widehat{\kappa}_z^4)\langle S^+S^-(t) + S^-S^+(t)\rangle + (\widehat{\kappa}_z^2 - \widehat{\kappa}_z^4)\left(\langle S_zS_z(t)\rangle - \langle S_z\rangle^2\right)\right)\right. \\ &\left. + (\widehat{\kappa}_z^2 - \widehat{\kappa}_z^4)\langle S_z\rangle^2\right] \times \exp(i(E'-E)t/\hbar)\, dt.\end{aligned} \tag{72}$$

where we have written $\eta \neq \eta'$ instead of $\pm \to \mp$, since the cross section is found to be independent on the neutron's spin flipping from up to down or vice versa, in contrast to nuclear scattering by polarized nuclei. The cross section for magnetic neutron non spin flip scattering can be derived accordingly, with the replacement of the matrix element product in Eq. (54) by $\langle\lambda| Q_{\perp z}^{\dagger} |\lambda'\rangle \langle\lambda'| Q_{\perp z} |\lambda\rangle$. This results in

$$\begin{aligned}\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\eta=\eta'} &= \frac{b_{\mathrm{m}}^2}{2\pi\hbar}\frac{k'}{k}\int_{-\infty}^{\infty}\sum_{jj'}\langle\exp(-i\kappa\cdot\mathbf{R}_{j'})\exp(i\kappa\cdot\mathbf{R}_j(t))\rangle |F(\kappa)|^2 \\ &\times \left[\delta_{jj'}\left(\frac{1}{4}(\widehat{\kappa}_z^2 - \widehat{\kappa}_z^4)\langle S^+S^-(t) + S^-S^+(t)\rangle + (1-\widehat{\kappa}_z^2)^2\left(\langle S_zS_z(t)\rangle - \langle S_z\rangle^2\right)\right)\right. \\ &\left. + (1-\widehat{\kappa}_z^2)^2\langle S_z\rangle^2\right] \times \exp(i(E'-E)t/\hbar)\, dt.\end{aligned} \tag{73}$$

The time dependence of the spin observables is governed by the Hamiltonian of a paramagnetic center in the external magnetic field, i.e.

$$\mathcal{H}_0 = -g\mu_{\mathrm{B}}B_0S_z. \tag{74}$$

The energy levels are given by the eigenstates of S_z with quantum number m,

$$\mathcal{H}_0 |m\rangle = E_m |m\rangle, \tag{75}$$

with

$$E_m = -g\mu_{\mathrm{B}}B_0 m. \tag{76}$$

The g-factors of the paramagnetic centers listed in Table 1 are $g \approx -2$. Energy transfers to or from the neutron may occur in units of the Zeeman energy denoted as

$$E^* = |g\mu_{\mathrm{B}}B_0|. \tag{77}$$

The system in thermal equilibrium at temperature T is chacterized by a partition function Z, with the population probabilities of the states $|m\rangle$ given by

$$p_m = \frac{\exp(-\beta E_m)}{Z}, \qquad Z = \sum_m \exp(-\beta E_m), \tag{78}$$

where the sum extends over the values $-S \leq m \leq S$, and

$$\beta = (k_{\mathrm{B}}T)^{-1} \tag{79}$$

with the Boltzmann constant k_{B}.

Evaluating first the matrix elements of operators S_z in Eqs. (72) and (73), we note that

$$\langle S_z \rangle = \sum_m p_m \langle m| S_z |m\rangle = \sum_m p_m m \tag{80}$$

and

$$\langle S_z S_z(t)\rangle = \sum_m p_m \langle m| S_z S_z(t) |m\rangle = \sum_m p_m m^2 = \langle S_z^2\rangle \tag{81}$$

are both time independent and thus describe scattering without electronic spin flip. The partition function of the Zeeman system is given by

$$Z = \sum_m \exp(-mx) = \frac{\sinh\left(\left(S+\frac{1}{2}\right)x\right)}{\sinh\frac{x}{2}}, \qquad x = -\beta g\mu_{\mathrm{B}} B_0, \tag{82}$$

from which, using Eqs. (80) and (81), follow the thermal average values of the spin observables S_z and S_z^2 as

$$\begin{aligned}\langle S_z\rangle &= \frac{1}{Z}\sum_m m\exp(-mx) = -\frac{1}{Z}\frac{dZ}{dx} \\ &= -\frac{1}{2}\left((2S+1)\coth\left(\frac{x}{2}(2S+1)\right) - \coth\frac{x}{2}\right),\end{aligned} \tag{83}$$

and

$$\begin{aligned}\langle S_z^2\rangle &= \frac{1}{Z}\sum_m m^2\exp(-mx) = \frac{1}{Z}\frac{d^2Z}{dx^2} \\ &= S(S+1) + \langle S_z\rangle\coth\frac{x}{2}.\end{aligned} \tag{84}$$

Next we analyze the matrix elements involving the operators $S^{\pm}$ in Eqs. (72) and (73). Application of the time independent operators to a state with quantum number m results in

$$S^{\pm}|m\rangle = \sqrt{S(S+1) - m(m\pm 1)}\,|m\pm 1\rangle. \tag{85}$$

Employing the time dependent operators

$$S^{\pm}(t) = \exp(i\mathcal{H}_0 t/\hbar)\, S^{\pm}\exp(-i\mathcal{H}_0 t/\hbar), \tag{86}$$

and using Eq. (85) and Eq. (86) with Eq. (75), we obtain

$$\begin{aligned}\langle S^{\pm}S^{\mp}(t)\rangle &= \sum_m p_m \langle m| S^{\pm}S^{\mp}(t)|m\rangle \\ &= \sum_m p_m \left(S(S+1) - m(m\mp 1)\right)\exp\left(i\left(E_{m\mp 1} - E_m\right)t/\hbar\right).\end{aligned} \tag{87}$$

These thermal averages thus describe electronic spin flips and associated energy transfer from or to the neutron. Using Eqs. (80) and (81) they can be expressed as

$$\langle S^{\pm}S^{\mp}(t)\rangle = \left(S(S+1) - \langle S_z^2\rangle \pm \langle S_z\rangle\right)\exp\left(\pm i g\mu_{\mathrm{B}} B_0 t/\hbar\right). \tag{88}$$

with the explicit temperature dependences of the thermal averages given in Eqs. (83) and (84).

The cross sections given in Eqs. (72) and (73) can now be evaluated, using Eqs. (63), (81) and (88), with integration over time and collecting the terms that correspond to electronic spin flip and those which do not. We denote the partial cross sections with electronic spin flip leading to a loss (gain) in neutron energy by a superscript $-$ $(+)$, and those without electronic spin flip by a superscript 0, i.e.

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)^{\pm}_{\eta\neq\eta'} = \frac{A}{4}\left(1+\widehat{\kappa}_z^4\right)\left(S(S+1) - \langle S_z^2\rangle \pm \langle S_z\rangle\right)\delta\left(E - E' \pm E^*\right), \tag{89}$$

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)^{\pm}_{\eta=\eta'} = \frac{A}{4}\left(\widehat{\kappa}_z^2 - \widehat{\kappa}_z^4\right)\left(S(S+1) - \langle S_z^2\rangle \pm \langle S_z\rangle\right)\delta\left(E - E' \pm E^*\right), \tag{90}$$

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)^{0}_{\eta\neq\eta'} = A\left(\widehat{\kappa}_z^2 - \widehat{\kappa}_z^4\right)\left(\langle S_z^2\rangle - \langle S_z\rangle^2 + \langle S_z\rangle^2\sum_j \exp\left(i\kappa\cdot\mathbf{R}_j\right)\right)\delta\left(E' - E\right), \tag{91}$$

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)^{0}_{\eta=\eta'} = A\left(1 - \widehat{\kappa}_z^2\right)^2\left(\langle S_z^2\rangle - \langle S_z\rangle^2 + \langle S_z\rangle^2\sum_j \exp\left(i\kappa\cdot\mathbf{R}_j\right)\right)\delta\left(E' - E\right). \tag{92}$$

The common factor

$$A = N b_{\mathrm{m}}^2 \frac{k'}{k}\exp(-2W)\left|F(\kappa)\right|^2, \tag{93}$$

contains the total number N of paramagnetic centers, and the Debye-Waller factor $\exp(-2W)$, where $2W = \kappa^2\langle u_\kappa^2\rangle$, and $\langle u_\kappa^2\rangle$ is the mean square displacement of a paramagnetic center in direction of κ.

The cross sections in Eqs. (89) and (90) involving an electron spin flip with energy transfer $\pm E^*$ are incoherent; they do not contain terms due to interferences of amplitudes from different paramagnetic centers. They vanish if the energy of the incident neutron is too small to compensate for the Zeeman energy needed to flip a single electron spin. In the opposite limit, $E \gg E^*$, and if one is not interested in the energy transfer, neglect of E^* in the δ functions and summing up the partial cross sections for electron spin flip and non-spin flip leads to equations found in the text books.

The electron non spin flip cross sections given in Eqs. (91) and (92) describe elastic scattering (if neglecting the neutron Zeeman energy in case of neutron spin flip scattering, the approximation adopted here). They contain an incoherent diffuse term and a coherent term proportional to $\langle S_z\rangle^2$ due to interferences of amplitudes from different paramagnetic centers, which may show up in Bragg peaks, or lead to small angle scattering contrast for instance for agglomerations of paramagnetic centers immersed in a non-magnetic solvent. Another noteworthy feature is the fact that the coherent cross section with neutron spin flip does not vanish in directions for which $\widehat{\kappa}_z^2 - \widehat{\kappa}_z^4 \neq 0$, i.e. when κ does not point parallel or perpendicular to the applied magnetic field.

For our calculations on neutron conversion and cooling we are primarily interested in the neutron energy changing total cross sections. After integration of $\widehat{\kappa}_z^2$ and $\widehat{\kappa}_z^4$ over solid angle,

$$\int \widehat{\kappa}_z^2 d\Omega = \frac{4}{3}\pi, \qquad \int \widehat{\kappa}_z^4 d\Omega = \frac{4}{5}\pi, \tag{94}$$

we can write them as

$$\left(\frac{d\sigma}{dE'}\right)^{\pm} = \left(\frac{d\sigma}{dE'}\right)^{\pm}_{\eta\neq\eta'} + \left(\frac{d\sigma}{dE'}\right)^{\pm}_{\eta=\eta'} = N\sigma_{\mathrm{m}}\frac{k'}{k}\exp(-2W)\, g_{\pm}(T)\, f_{\pm}(E)\,\delta\left(E \pm E^* - E'\right), \tag{95}$$

where we have defined $\sigma_{\mathrm{m}} = 4\pi b_{\mathrm{m}}^2 \approx 3.66$ barn and

$$g_{\pm}(T) = \frac{1}{3}\left(S(S+1) - \langle S_z^2\rangle \pm \langle S_z\rangle\right), \tag{96}$$

with $\langle S_z\rangle$ and $\langle S_z^2\rangle$ given by Eqs. (83) and (84). The functions $f_{\pm}(E)$ account for the magnetic form factor, which is discussed in the main text.

5.2 Cross sections for the molecular oxygen spin triplet system

Here we consider magnetic neutron scattering by an assembly of unoriented oxygen molecules with motions frozen out. The molecules are assumed to be kept sufficiently far apart from each other to avoid magnetic ordering. This can be achieved using the cage structures discussed in the main text. Our primary interest is the scattering involving transitions between magnetic levels within the triplet state, which is inelastic due to the molecular zero-field splitting. For unoriented molecules and without external magnetic field there is no global quantization axis in the system. It is therefore appropriate to start from the magnetic scattering cross section for unpolarized neutrons (see, e.g., [29]),

$$\frac{d^2\sigma}{d\Omega dE'} = b_{\mathrm{m}}^2 \frac{k'}{k} \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \widehat{\kappa}_\alpha \widehat{\kappa}_\beta\right) \sum_{\lambda\lambda'} p_\lambda \left\langle \lambda \right| Q_\alpha^\dagger \left| \lambda' \right\rangle \left\langle \lambda' \right| Q_\beta \left| \lambda \right\rangle \delta\left(E_{\lambda'} - E_\lambda + E' - E\right), \tag{97}$$

using the same notation of states and transition operators as in the previous section. Each oxygen molecule is characterized by a coordinate $\mathbf{R}_j$ of its center of gravity and relative positions $\mathbf{l}_{j1}$ and $\mathbf{l}_{j2}$ of the two atoms. Projection of the total spin onto the molecular axis, $\mathbf{l}_{j1} - \mathbf{l}_{j2}$, provides a good quantum number. As we do not deal with nuclear scattering, the atomic coordinates do not explicitly occur as variables in the cross section but manifest implicitly as a site dependence of the spin eigenstates. Also the magnetic form factor depends on the molecular orientation, which we can however take as isotropic for our purposes (see section 2). Taking electronic spins and spatial coordinates as independent quantities we write

$$\begin{aligned} \frac{d^2\sigma}{d\Omega dE'} &= \frac{b_{\mathrm{m}}^2}{2\pi\hbar} \frac{k'}{k} \left|F(\kappa)\right|^2 \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \widehat{\kappa}_\alpha \widehat{\kappa}_\beta\right) \times \\ &\times \int_{-\infty}^{\infty} \sum_{jj'} \left\langle \exp\left(-i\kappa \cdot \mathbf{R}_{j'}\right) \exp\left(i\kappa \cdot \mathbf{R}_j(t)\right) \right\rangle \left\langle S_{j'\alpha} S_{j\beta}(t) \right\rangle \exp\left(i\left(E' - E\right) t/\hbar\right) dt. \end{aligned} \tag{98}$$

For uncorrelated oxygen molecules,

$$\left\langle S_{j'\alpha} S_{j\beta}(t) \right\rangle = \left\langle S_{j'\alpha} \right\rangle \left\langle S_{j\beta}(t) \right\rangle = 0 \qquad (j \neq j'). \tag{99}$$

We are thus left with a single sum over an assembly of unoriented and independent triplet spin systems,

$$\begin{aligned} \frac{d^2\sigma}{d\Omega dE'} &= \frac{b_{\mathrm{m}}^2}{2\pi\hbar} \frac{k'}{k} \exp(-2W) \left|F(\kappa)\right|^2 \times \\ &\times \int_{-\infty}^{\infty} \sum_j \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \widehat{\kappa}_\alpha \widehat{\kappa}_\beta\right) \left\langle S_{j\alpha} S_{j\beta}(t) \right\rangle \exp\left(i\left(E' - E\right) t/\hbar\right) dt. \end{aligned} \tag{100}$$

The product $\langle \exp(-i\kappa \cdot \mathbf{R}_j) \exp(i\kappa \cdot \mathbf{R}_j(t)) \rangle$ is the Debye-Waller factor denoted by $\exp(-2W)$. In the sum over j any molecular orientation appears with equal weight with respect to the given direction $\widehat{\kappa}$, of which the differential cross section is obviously independent. We may therefore define for each molecule its own coordinate system and replace

$$\sum_j \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \widehat{\kappa}_\alpha \widehat{\kappa}_\beta\right) \left\langle S_{j\alpha} S_{j\beta}(t) \right\rangle = N \left\langle \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \widehat{\kappa}_\alpha \widehat{\kappa}_\beta\right) S_\alpha S_\beta(t) \right\rangle. \tag{101}$$

On the right side the brackets include angular averaging in addition to the thermal averaging over molecular spin states. Accordingly we have omitted the site index j to the spin operators.

For further evaluation we choose local cartesian coordinates with z axis parallel to the molecular axis and take the x and y axes in directions for which their projections on κ are equal, i.e.

$$\widehat{\kappa}_x = \widehat{\kappa}_y = \frac{1}{\sqrt{2}} \sin\vartheta, \qquad \widehat{\kappa}_z = \cos\vartheta, \tag{102}$$

where ϑ is the angle between κ and the molecular axis. The triplet states of the oxygen molecule are labelled by quantum numbers $m = -1, 0, +1$ characterizing the spin state projection along the symmetry axis of the molecule. The Hamiltonian (without external magnetic field) is given by

$$\mathcal{H}_0 = DS_z^2 - \frac{2}{3}D, \tag{103}$$

which accounts for the energy difference by the zero-field splitting constant D of the states with $m = \pm 1$ and the $m = 0$ state [71]. It commutes with S_z, and since the spin operators obey the same algebra as in the Zeeman case (with different meaning of the states), with the definition of raising and lowering operators in Eq. (62), we use Eqs. (63), (66), (67) and (81), and obtain

$$\left\langle \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \widehat{\kappa}_\alpha \widehat{\kappa}_\beta\right) S_\alpha S_\beta(t) \right\rangle = \frac{1}{3} \left\langle \left(S^+ S^-(t) + S^- S^+(t)\right) \right\rangle + \frac{2}{3} \left\langle S_z^2 \right\rangle. \tag{104}$$

The eigenenergies of $\mathcal{H}_0$ are given by

$$E_m = \left(m^2 - \frac{2}{3}\right) D. \tag{105}$$

The partition function as defined in Eq. (78) follows as

$$Z = \exp(2x) + 2\exp(-x), \qquad x = \frac{\beta D}{3}. \tag{106}$$

Using Eq. (87) with Eq. (105), we obtain

$$\left\langle S^+ S^-(t) + S^- S^+(t) \right\rangle = 4 \frac{\exp(iDt/\hbar) + \exp(-\beta D)\exp(-iDt/\hbar)}{1 + 2\exp(-\beta D)}. \tag{107}$$

The total scattering cross sections with neutron energy loss $(-)$, energy gain $(+)$ thus become

$$\left(\frac{d\sigma}{dE'}\right)^{\pm} = N\sigma_{\mathrm{m}} \frac{k'}{k} \exp(-2W)\, g_\pm(T)\, f_\pm(E)\, \delta\left(E \pm D - E'\right), \tag{108}$$

where

$$g_-(T) = \frac{4}{3} \frac{1}{1 + 2\exp(-\beta D)}, \tag{109}$$

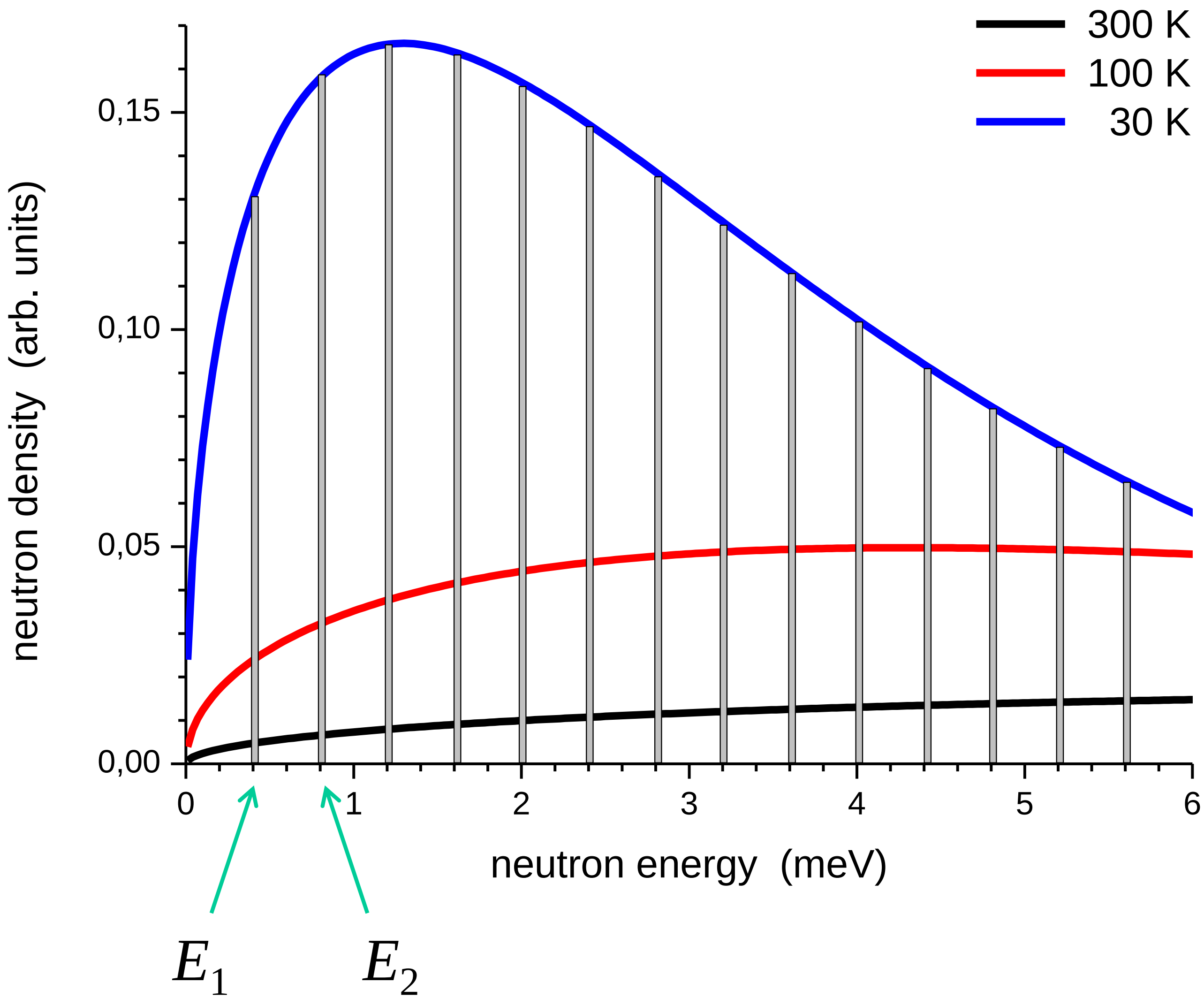
and

$$g_+(T) = \frac{4}{3} \frac{\exp(-\beta D)}{1 + 2\exp(-\beta D)}. \tag{110}$$

The functions $f_\pm(E)$ account for the magnetic form factor as discussed in the main text. The cross sections fulfill the relation of detailed balance, as they have to.

Neutron moderation by the paramagnetic system

Neutron groups ($j = 0, 1, 2, \ldots$): $E_j = jE^* + \Delta$



For paramagnetic <u>cascade cooling</u> of neutrons

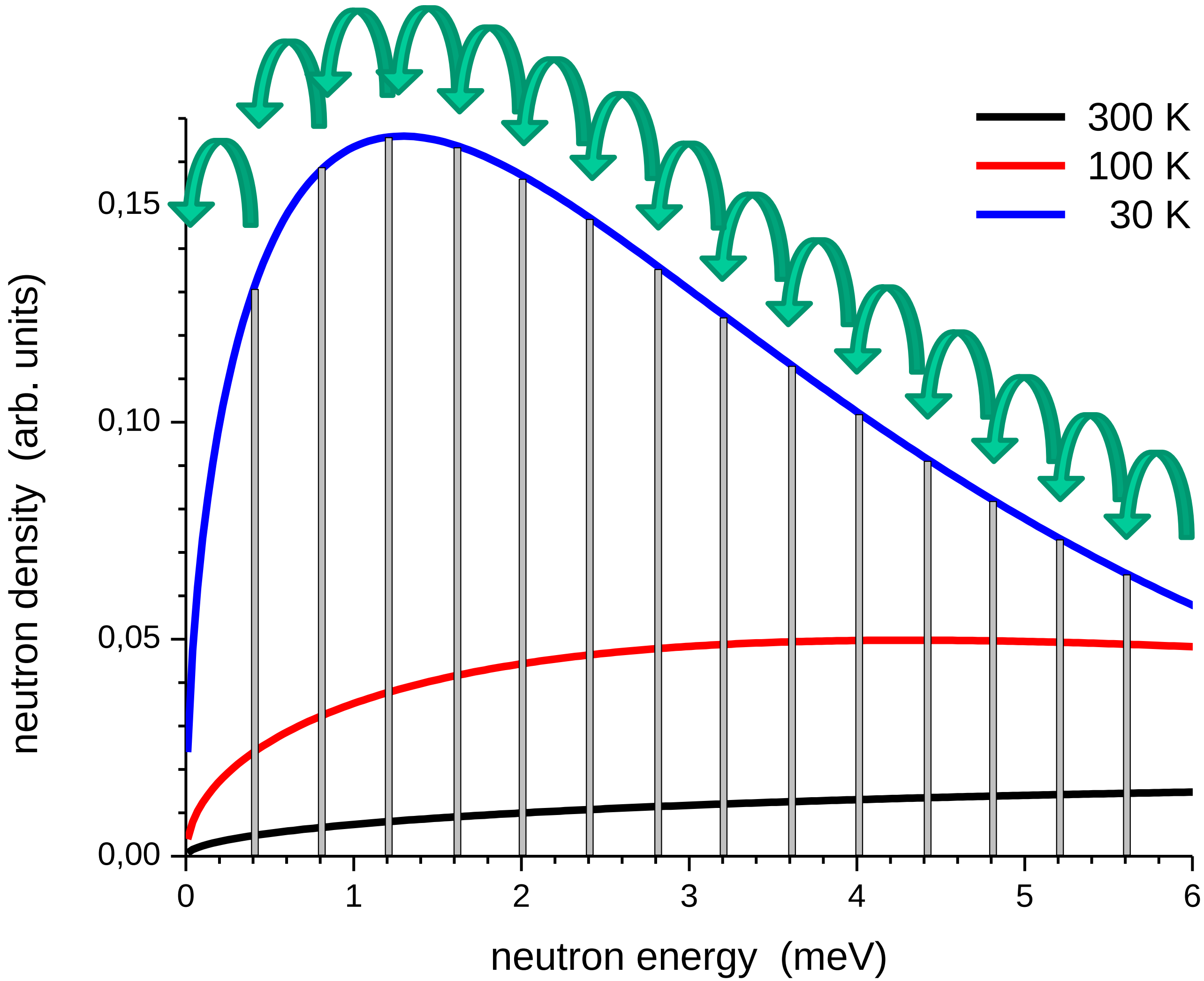
Solve rate equations for infinite medium:

$$\frac{dn_j}{dt} = \overset{\text{source}}{s_j} \overbrace{\boxed{+ n_{j+1}\tau^{-1}_{j+1\to j} + n_{j-1}\tau^{-1}_{j-1\to j}}}^{\text{feeding}} \overbrace{\boxed{- n_j\tau^{-1}_{j\to j-1} - n_j\tau^{-1}_{j\to j+1}}}^{\text{depopulation}} \overset{\text{absorption}}{- n_j\tau^{-1}_{\mathrm{a}}}$$

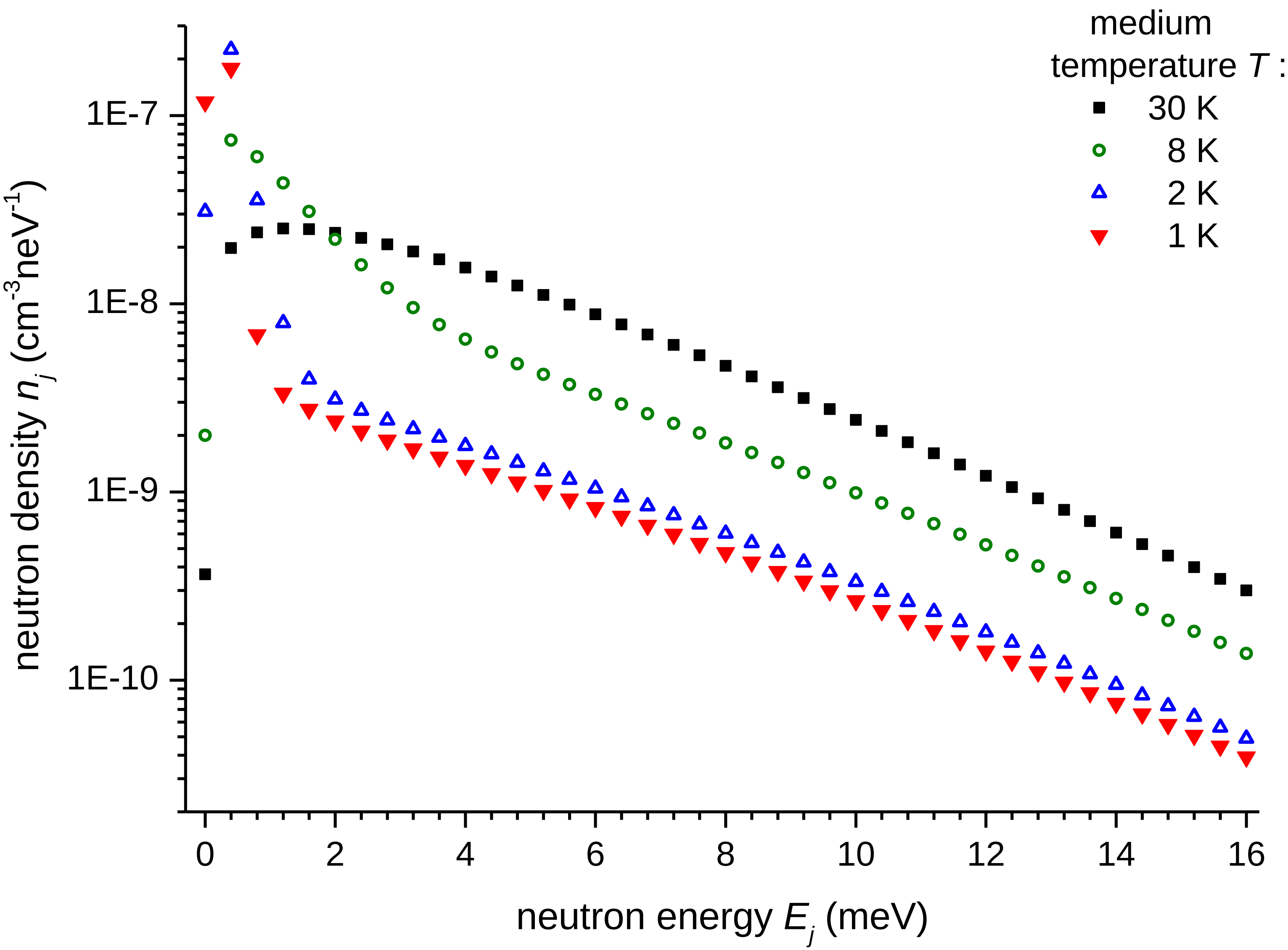
$$\tau^{-1}_{j\to j\pm 1} = n_{\mathrm{pc}}\sigma_{\mathrm{m}} g_{\pm}(T)\, f_{\pm}(E_j)\, v_{j\pm 1}$$

and calculate stationary solutions...

For low temperature of the medium, i.e., $k_B T < E^* \cong 4.6$ K:



Stationary neutron group populations in O_2 clathrate



Moderator peculiarities:

Bragg-cutoff length: $\lambda_{\mathrm{B}} \approx 2.0\ \mathrm{nm}$

whereas $E^* = 0.4\ \mathrm{meV}$ corresponds to $1.41\ \mathrm{nm}$

$\rightarrow$ O_2-hydrate is a flux trap for neutrons still to be converted to VCN!

$$\boxed{\Sigma_{\mathrm{a}} \ll \Sigma_{\mathrm{ie}} \ll \Sigma_{\mathrm{e}}}$$

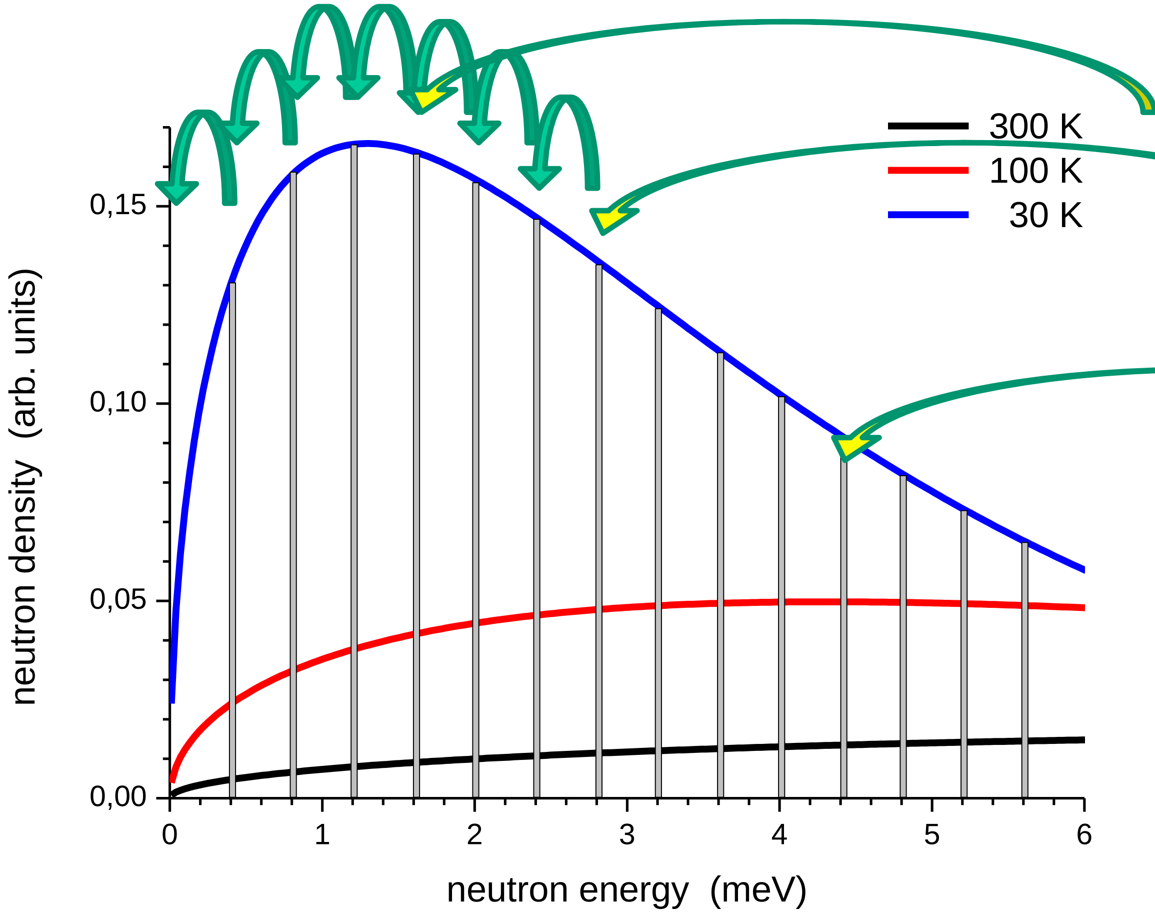
Diffusion length (for inelastic magnetic processes):

$$L_{\mathrm{d}} = 1/\sqrt{3\Sigma_{\mathrm{ie}}\left(\Sigma_{\mathrm{e}} + \Sigma_{\mathrm{ie}}\right)} \qquad L_{\mathrm{d}} \approx 10\ \mathrm{cm}$$

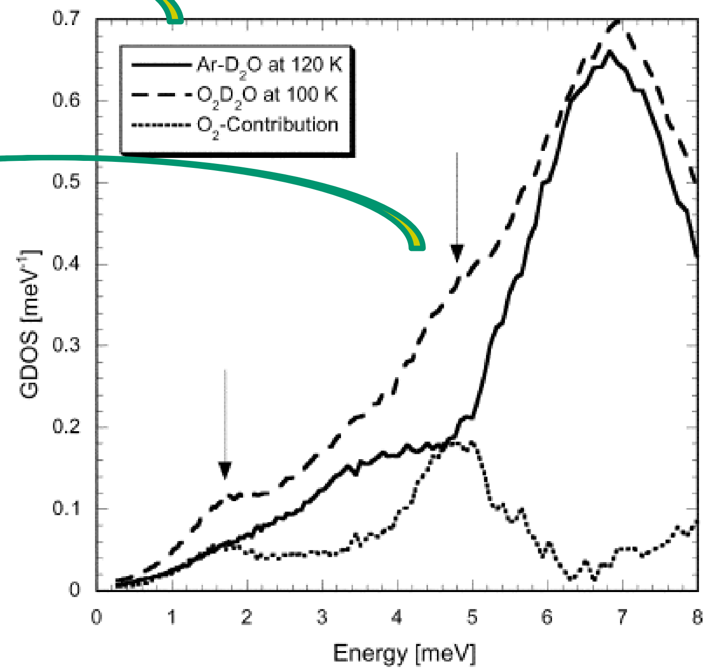
for full moderation if there were only paramagnetic cooling

Short-circuiting of long paramagnetic cascade by Einstein modes at 4.8 meV?

→ moderator could become much smaller



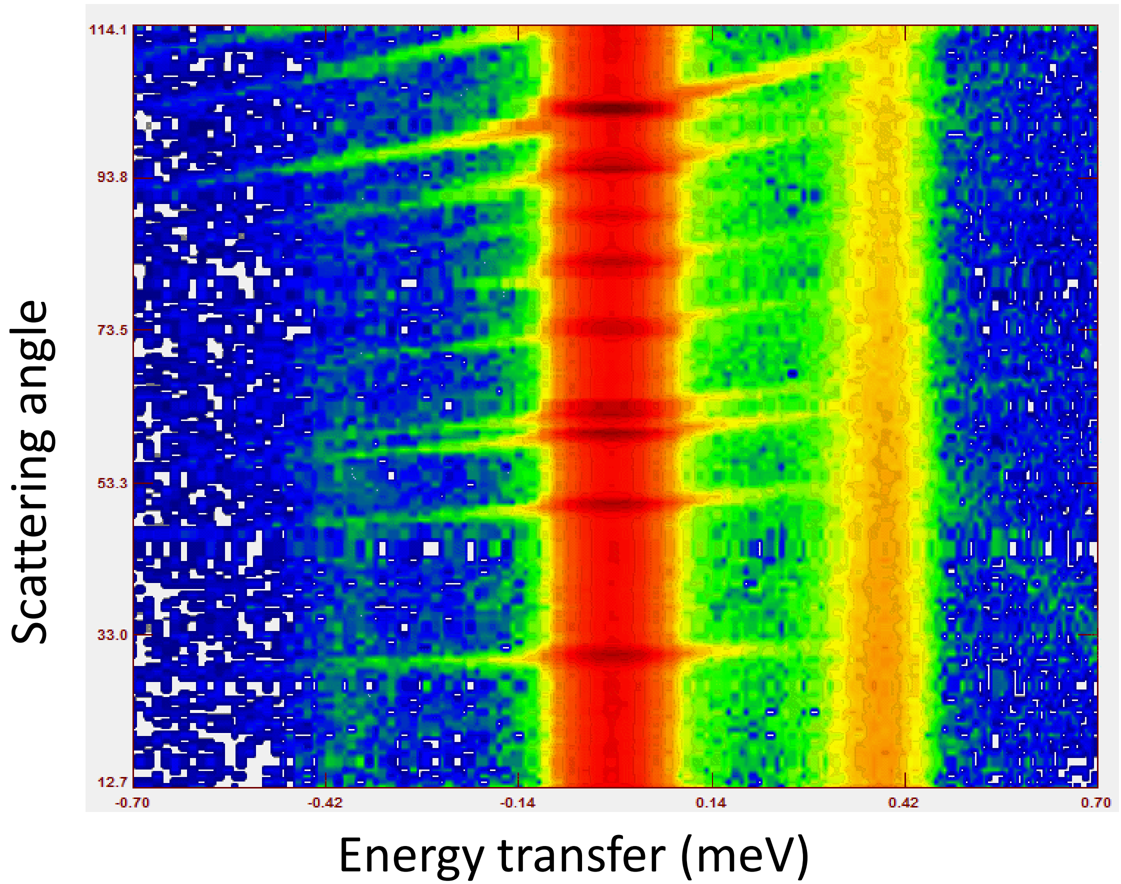
Chazallon, Itoh, Koza, Kuhs, Schober, Chem. Phys. Phys. Chem. 4, 4809 (2002)



First experiments done at D20, IN4, IN6 and D7

Goal: determine absolute cross sections

A. Falenty, T. Hansen, M. Koza, W. Kuhs, O. Zimmer

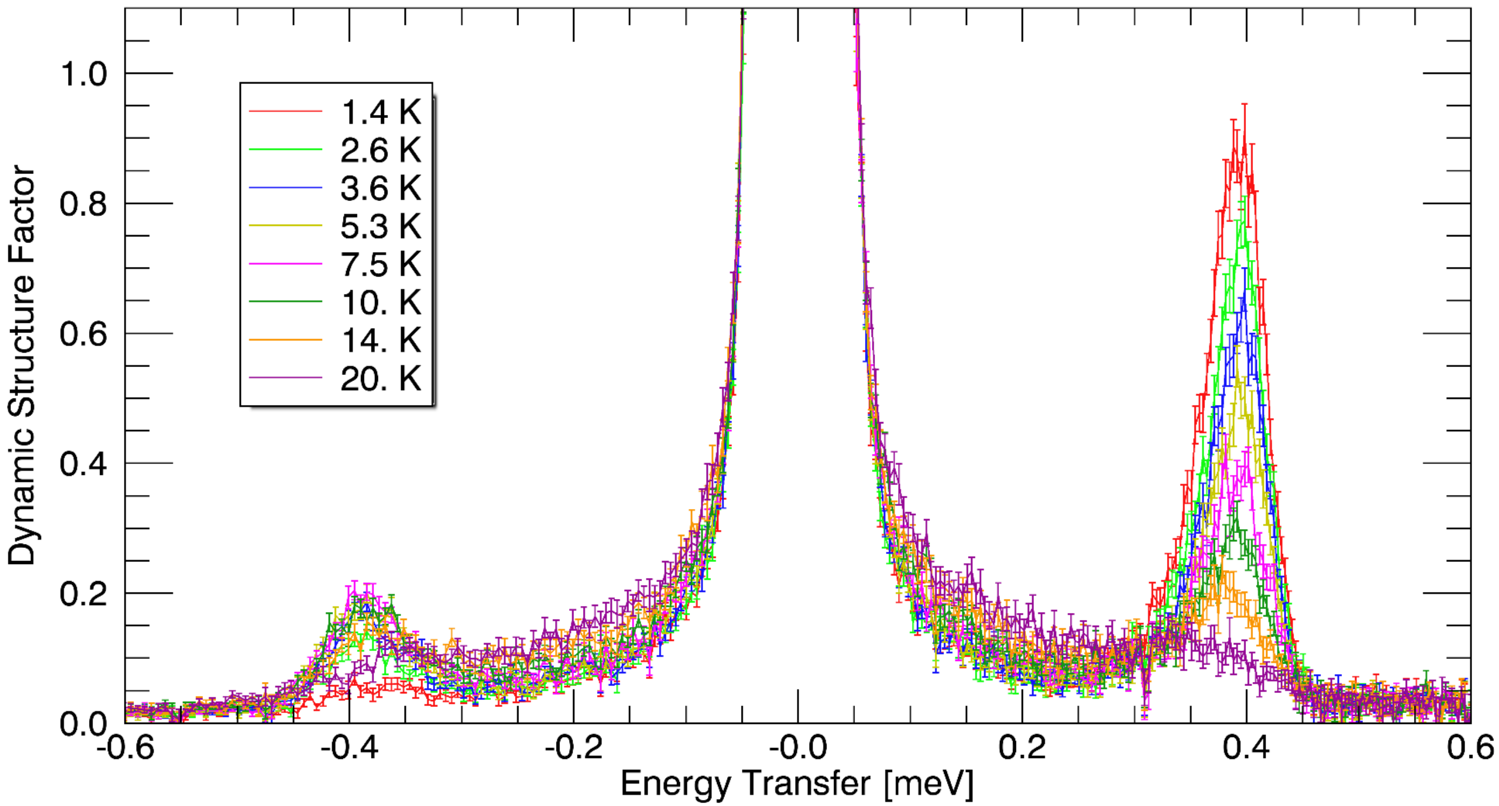


shows dispersion-free excitation at 0.4 meV, magnetic form factor

First experiments done at D20, IN4, IN6 and D7

Goal: determine absolute cross sections

A. Falenty, T. Hansen, M. Koza, W. Kuhs, O. Zimmer



Magnetic intensity seems in agreement with theoretical prediction

For more details, please have a look at my paper:

"Neutron conversion and cascaded cooling in paramagnetic systems for a high-flux source of very cold neutrons"

Phys. Rev. C **93**, 035503 (2016)

Conclusion:

Things need to be done right for significant gains with respect to the current state of the art but might then be worthwhile…