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Friday 16 January JST 16:55-17:40 (35+10 min)

Dynamic magnetic pair-density function (DymPDF) analysis

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Collaborators

Neutron: Yasuhiro Inamura (JAEA), Kazuki Iida(CROSS), Katsuaki Kodama, Mitsutaka Nakamura (JAEA) Lieh-Jeng Chang (NCKU), Daichi Ueta (KEK), Hiroki Yamauchi (JAEA), Jae-Ho Chung (Korea Univ.)

μSR: Takashi U. Ito, Wataru Higemoto (JAEA)), Yasutomo Uemura (Columbia Univ.)

X-ray: Satoshi Tsutsui, Hiroshi Fukui, Taiushun Manjo (JASRI), Alfred Q.R. Baron (RIKEN)

Supercomputer FUGAKU: Kanako Yoshizawa, Yasuhiro Sunaga, Daisuke Matsuoka, Satoru Asami (RIST)

Samples: Yukio Yasui (Meiji Univ.), Jose Antonio Alonso (CSIC, ICMM)

Acknowledgments

Grants-in-Aid for Scientific Research (C) (**25K08263, 22K04678**, 21K03478, and 19K12648) from the Japan Society for the Promotion of Science. Proposals 2024C0001(BL01), 2024A0053(S1), 2021C0001(BL01), 2020A0052 (BL01) (J-PARC), 2024A1156 (60524, BL35XU@SPring-8). The samples were characterized using a magnetic properties measurement system at the CROSS user laboratory II.

Outline

1. Introduction

    Fourier transform analysis of neutron scattering data

    Pair Distribution Function (PDF)

    Magnetic Pair Distribution Function (mPDF)

    Dynamic Pair Density Function (DyPDF)

2. Dynamic magnetic Pair Density Function (DymPDF)

3. How to get DymPDF by ‘Utsusemi’

4. How to simulate DymPDF by ‘SpinW‘ and ‘Utsusemi’

5. Summary

1. Introduction

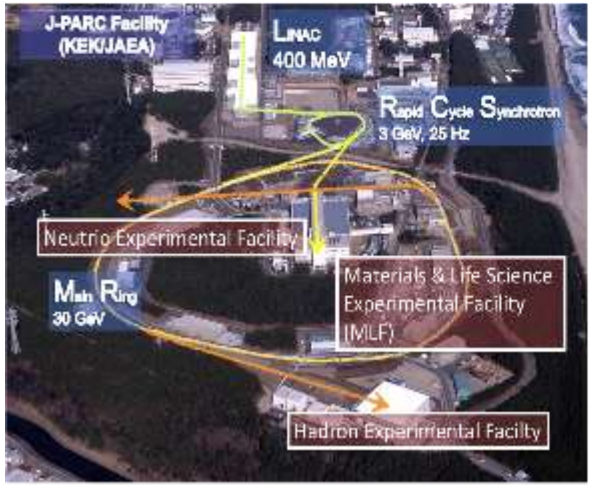
Recent history of the Fourier transform analysis on neutron scattering

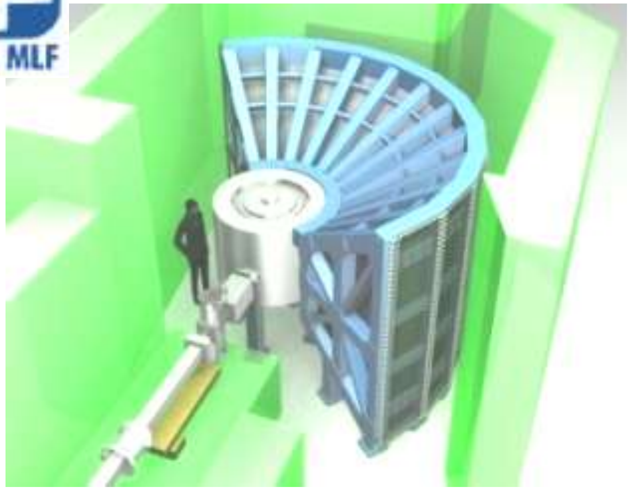
1990 Crystalline PDF	1992 DyPDF	2014 mPDF	2022 DymPDF
T. Egami, *Mater. Trans. JIM* **31**, 163–176 (1990).	A. C. Hannon, M. Arai, *et al*., *J. Non-Cryst. Solids* **150**, 239 (1992).	B. A. Frandsen, X. Yang, S. J. L. Billinge, *Acta Crystallogr. Sect.* A **70**, 3 (2014).	K. Iida, K. Kodama, Y. Inamura, M. Nakamura, L.-J. Chang, S. Shamoto *Scientific Reports* **12**: 20663 (2022).
IPNS, ANL USA	**ISIS, RAL UK**	**ILL France**	**J-PARC MLF Japan**

PDF : Pair Distribution (Density) Function

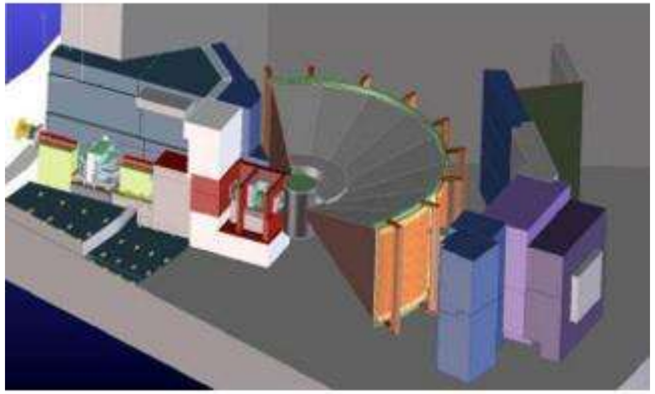
J-PARC : Japan Proton Accelerator Research Complex

- Materials & Life Science Experimental Facility (MLF)



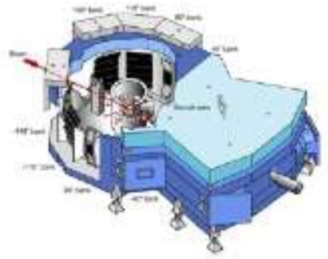


4SEASONS(BL01):
S/N=43.8



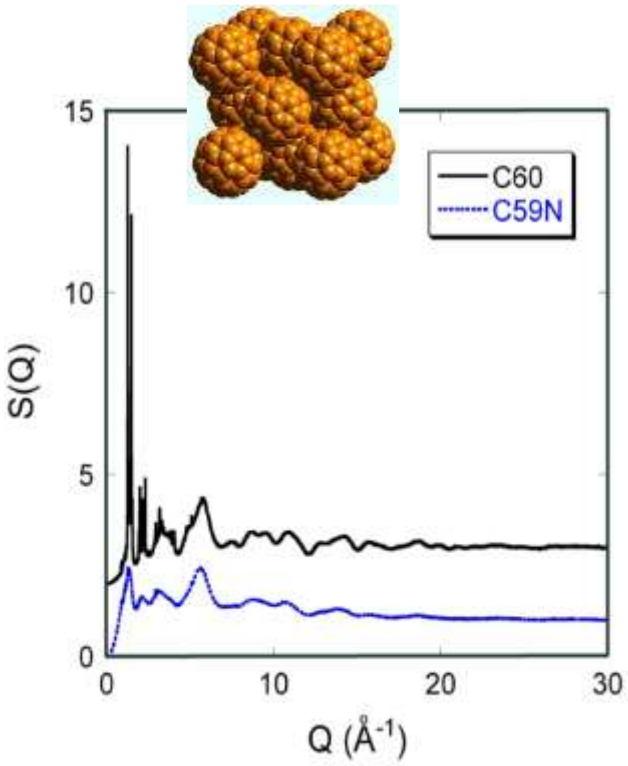
HRC(BL12):

Pair Distribution Function (PDF)



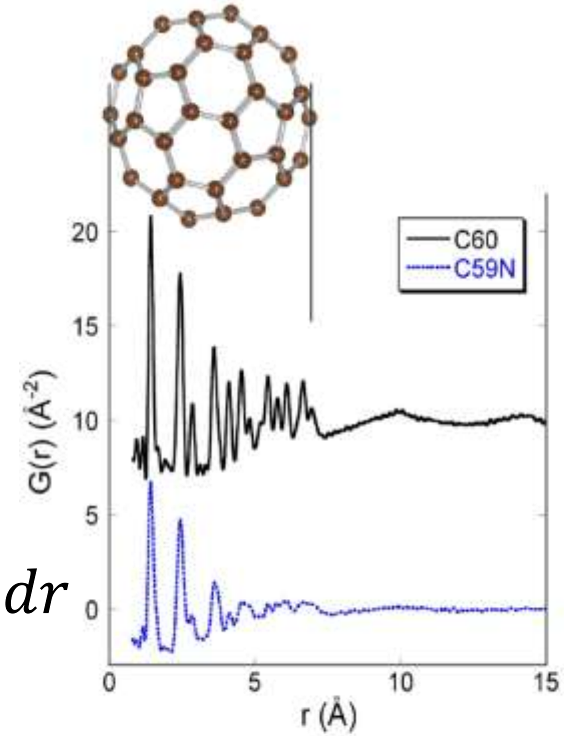
NPDF@Los Alamos

C60 Fullerene,
C59N Azafullerene



Fourier transform

$$G(r) = \frac{2}{\pi}\int_0^\infty Q[S(Q)-1]\sin(Qr)dr$$



Scattering function

Atomic pair distribution function (PDF)

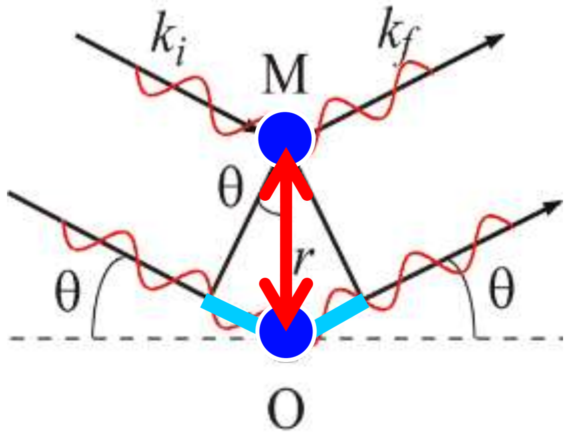
S. Shamoto, J. Phys. Soc. Jpn. **88,** 081008 (2019).

Pair Distribution Function (PDF)

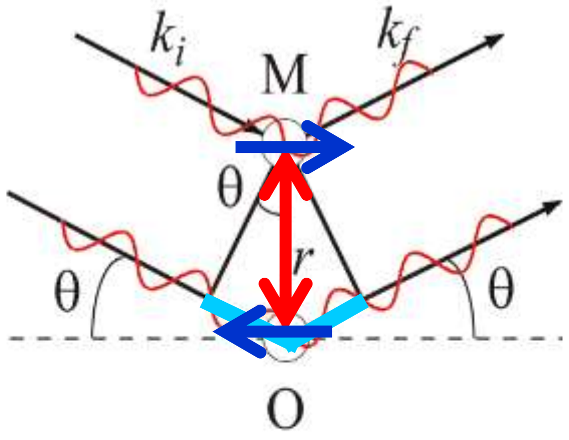
Elastic scattering by two atoms (O and M)

Sum of phase difference : $(k_f - k_i) \cdot r$

Bragg condition $(k_f - k_i) \cdot r = 2\pi n$



Atomic pair correlation



Spin pair correlation

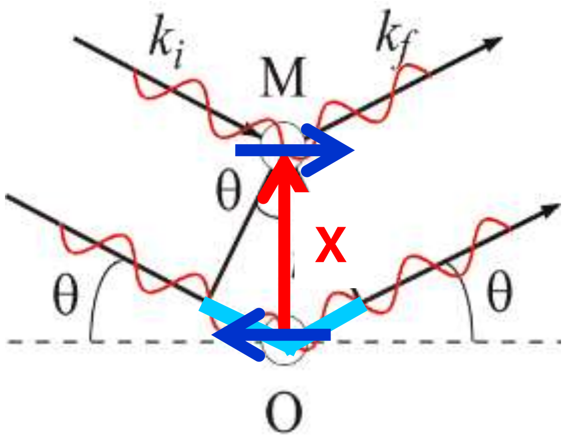
mPDF analysis

$$f(r) = \frac{2}{\pi} \int_0^{\infty} Q \left[\frac{I_m}{\frac{2}{3} N_s S(S+1)(\gamma r_0)^2 f_m^2(Q)} - 1 \right] \sin Qr \, dQ \quad (1)$$

$$= \frac{1}{N_s} \frac{3}{2S(S+1)} \sum_{i \neq j} \left[\frac{A_{ij}}{r} \delta(r - r_{ij}) + B_{ij} \frac{r}{r_{ij}^3} \Theta(r_{ij} - r) \right], \quad (2)$$

$$A_{ij} = \langle S_i^y S_j^y \rangle, \; B_{ij} = 2 \langle S_i^x S_j^x \rangle - \langle S_i^y S_j^y \rangle$$

$$\hat{\mathbf{x}} = \frac{\mathbf{r}_j - \mathbf{r}_i}{|\mathbf{r}_j - \mathbf{r}_i|} \qquad \hat{\mathbf{y}} = \frac{\mathbf{S}_i - \hat{\mathbf{x}}(\mathbf{S}_i \cdot \hat{\mathbf{x}})}{|\mathbf{S}_i - \hat{\mathbf{x}}(\mathbf{S}_i \cdot \hat{\mathbf{x}})|}$$

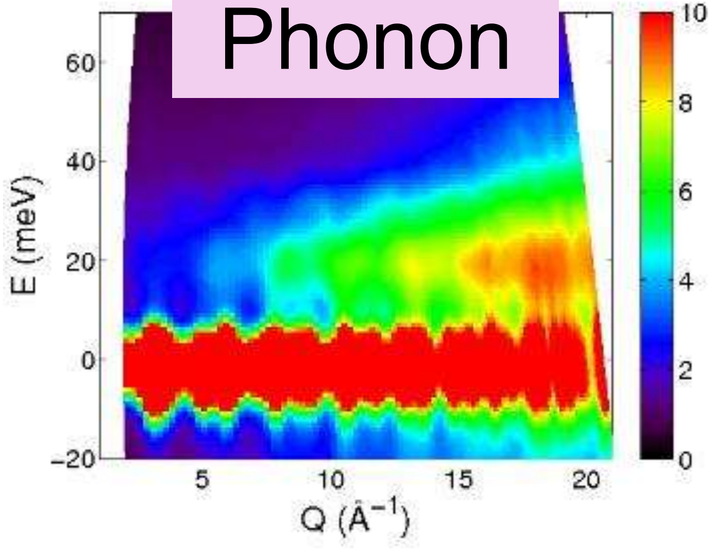


B. A. Frandsen, X. Yang, S. J. L. Billinge, *Acta Crystallogr. Sect.* A **70**, 3 (2014).

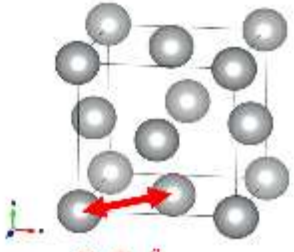
Dynamic Pair Density Function (DyPDF)

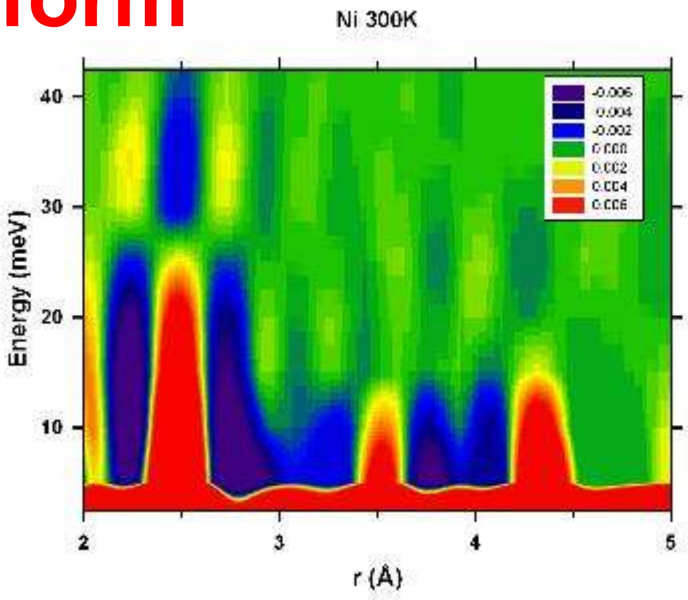
Fourier transform

Phonon



Polycrystalline Ni





DyPDF of Ni

Alex C. Hannon, Masatoshi Arai, Roger N. Sinclair, Adrian C. Wright, “A dynamic correlation function for amorphous solids”, Journal of Non-Crystalline Solids,150, 239 (1992)

Wojtek Dmowski et al. “Local lattice dynamics and the origin of the relaxor ferroelectric material.” Phys. Rev. Lett. 100, 137602 (2021).

Lattice component of the dynamic structure factor

$$\frac{S_L(Q,E)}{\langle 1+n(E)\rangle}-1=\frac{\left(k_i/k_f\right)I(Q,E)-S_S(Q,E)-A_0}{S_S(Q,E)}$$

$$S_S(Q,E)=BQ^2\exp(-Q^2\langle u^2\rangle/3)\frac{\langle 1+n(E)\rangle}{E}$$

Alex C. Hannon, M. Arai et al., J. Non-Cryst. Solids 150, 239 (1992).

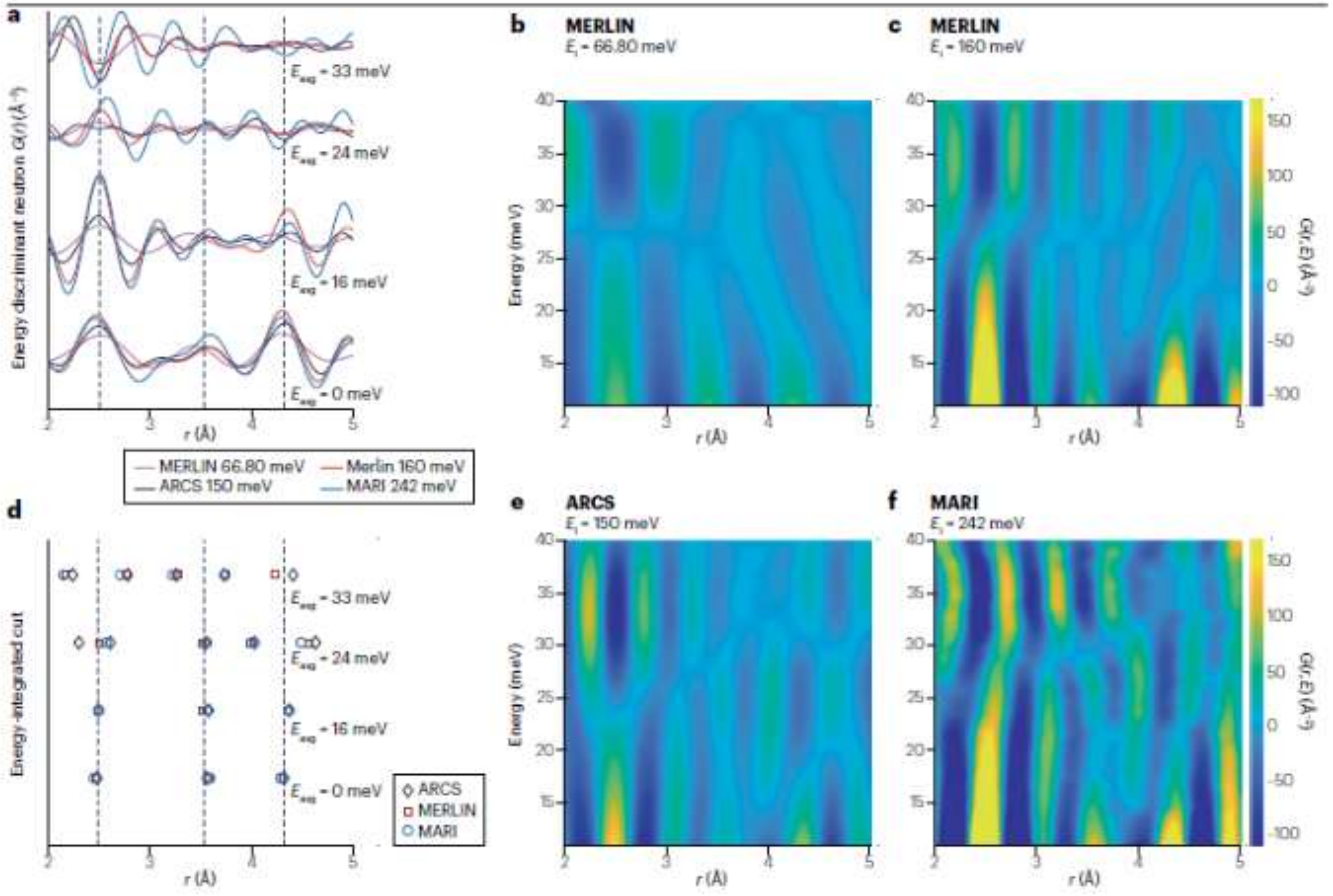
Lattice component of the dynamic pair-density function

$$D_L(r,E)=\frac{2}{\pi}\int_{Q_{min}}^{Q_{max}}Q\left[\frac{S_L(Q,E)}{\langle 1+n(E)\rangle}-1\right]w(Q)\sin(Qr)dQ$$

Window function $w(Q)=\frac{Q_{max}}{\pi Q}\sin\left(\frac{\pi Q}{Q_{max}}\right)$

Nature Reviews Physics (2023). (Technical review)

DyPDF, D(r,E)



Optimizing the dynamic pair distribution function method for inelastic neutron spectrometry

Kody A. Acosta, Helen C. Walker & Allyson M. Fry-Petit

Nature Reviews Physics **5,** 236–249 (2023). (Technical review)
https://doi.org/10.1038/s42254-023-00564-5

2. Dynamic magnetic Pair Density Function (DymPDF)

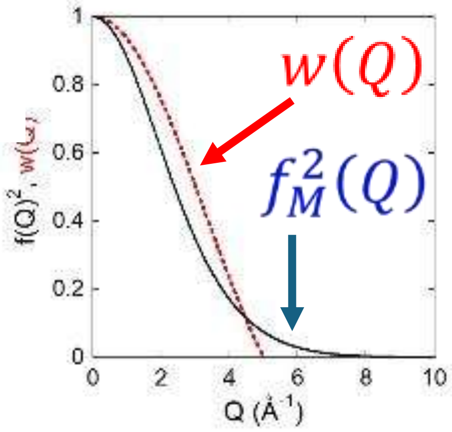
Magnetic component of the dynamic structure factor

$$\frac{S_M(Q,E)}{\langle 1+n(E)\rangle}-1=\frac{(k_i/k_f)I(Q,E)-A(Q-Q_{max})(Q+Q_{max})-A_0}{\frac{2}{3}N_m g^2 S(S+1)(\gamma r_0)^2\langle 1+n(E)\rangle f_M^2(Q)}$$

Magnetic component of the dynamic pair-density function

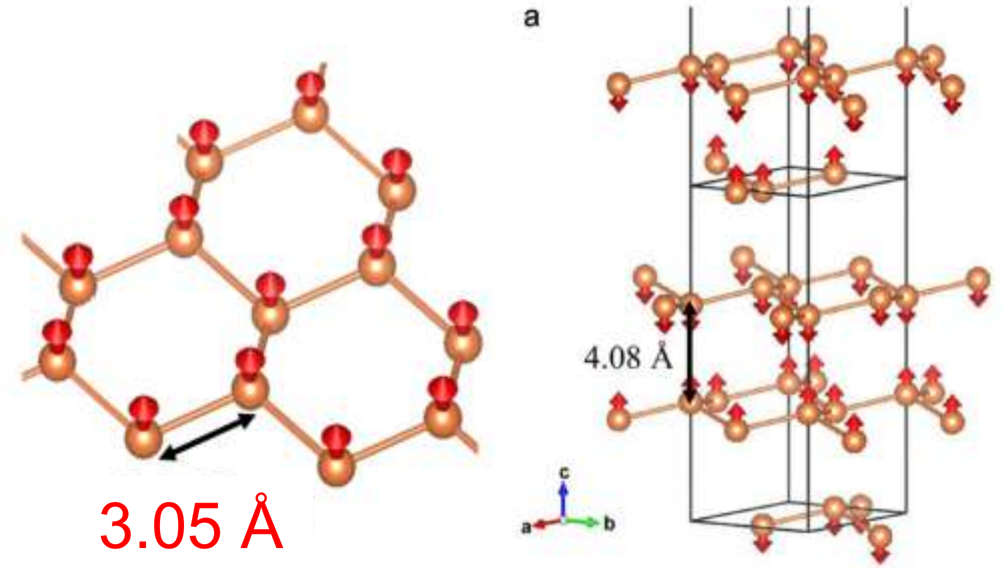
$$D_M(r,E)=\frac{2}{\pi}\int_{Q_{min}}^{Q_{max}} Q\left[\frac{S_M(Q,E)}{\langle 1+n(E)\rangle}-1\right]w(Q)\sin(Qr)\,dQ$$

Window function $$w(Q)=\frac{Q_{max}}{\pi Q}\sin\left(\frac{\pi Q}{Q_{max}}\right)$$



K. Iida, K. Kodama, Y. Inamura, M. Nakamura, L.-J. Chang, S. Shamoto, *Sci. Rep.* 12: 20663 (2022).

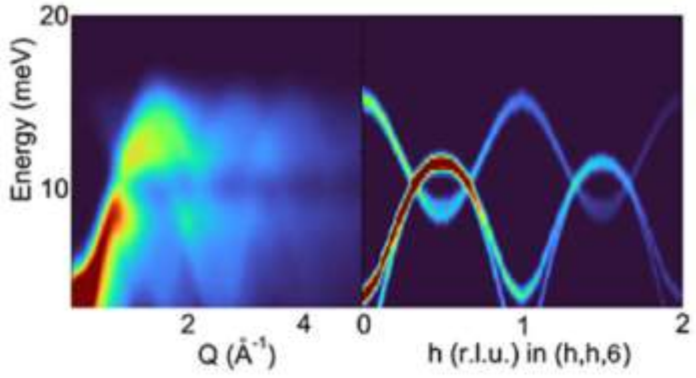
$Fe^{2+}TiO_3$



Crystal structure

T_N = 59 K

Spin-wave dispersion (SpinW)



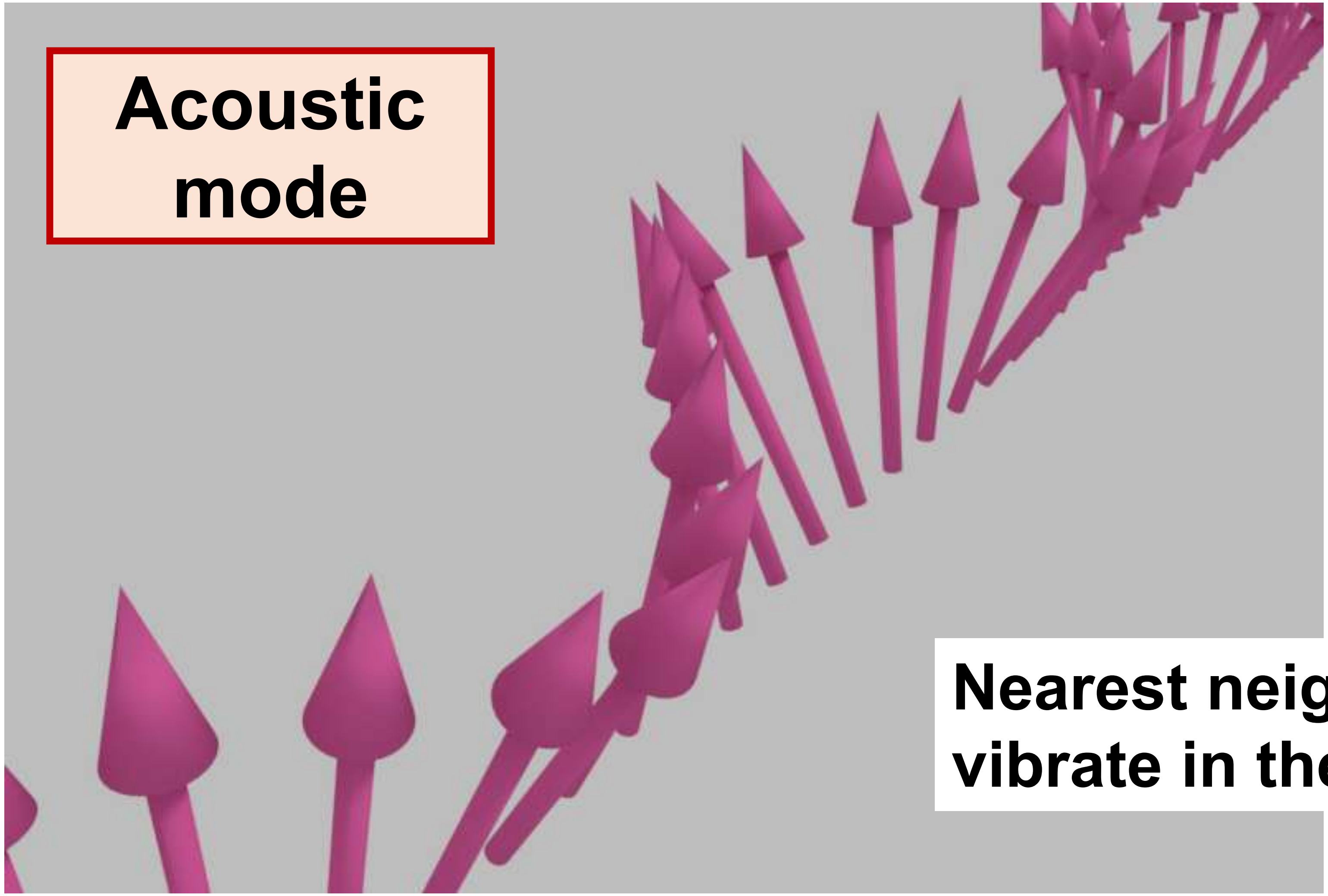
Powder average

Spin-wave dispersion

SpinW

H. Kato, Y. Yamauchi et al., *J. Phys. C: Solid State Phys.* **19**, 6993 (1986).
S. Toth, and B. Lake, *J. Phys.: Condens. Matter* **27**, 166002 (2015).

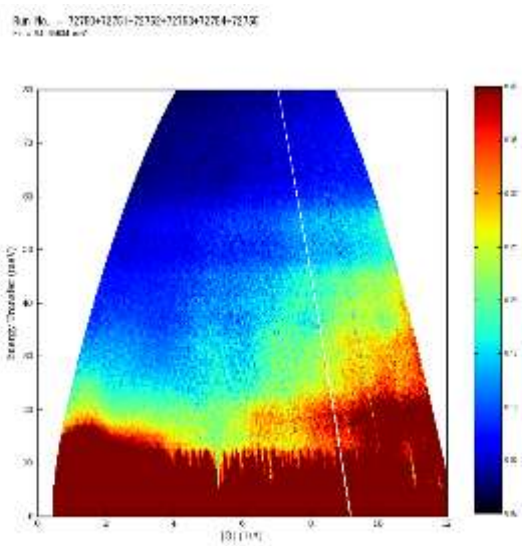
Classical limit of spin dynamics



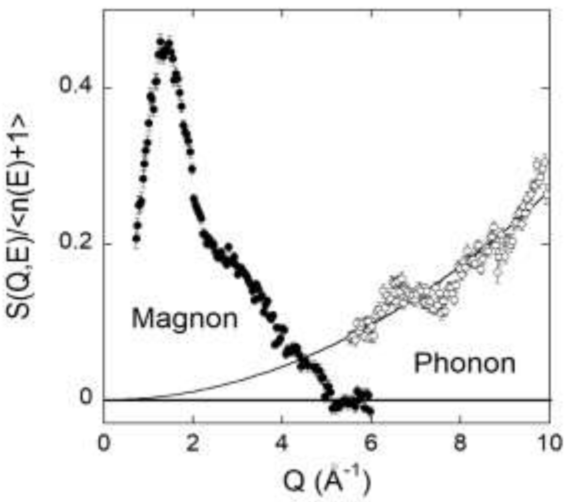
Nearest neighbor spins vibrate in the same direction!

https://staff.aist.go.jp/v.zayets/spin3_68_heat.html

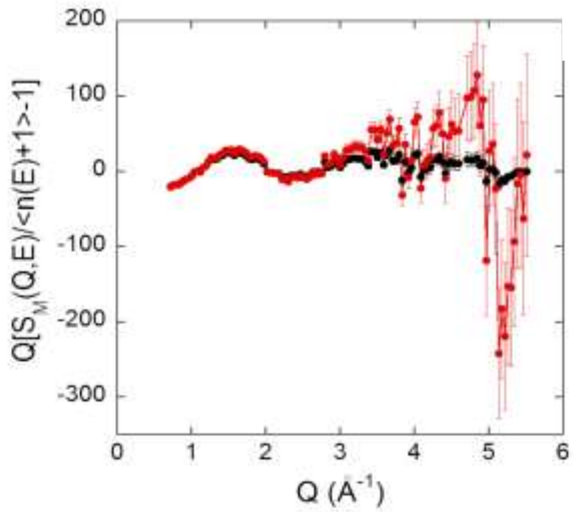
Separation of magnon from phonon



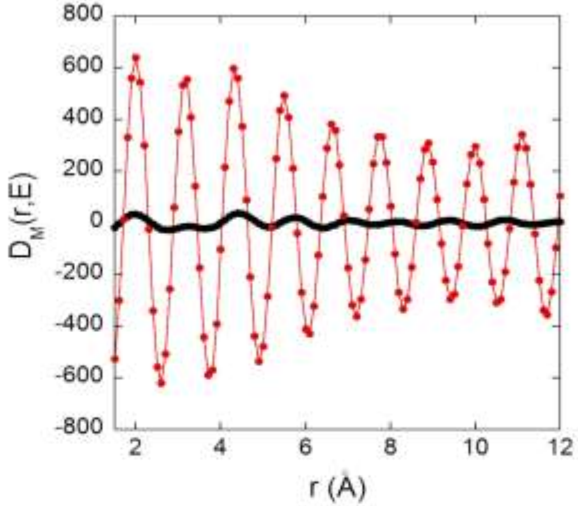
E_i = 95 meV



E = 15 meV



DymPDF



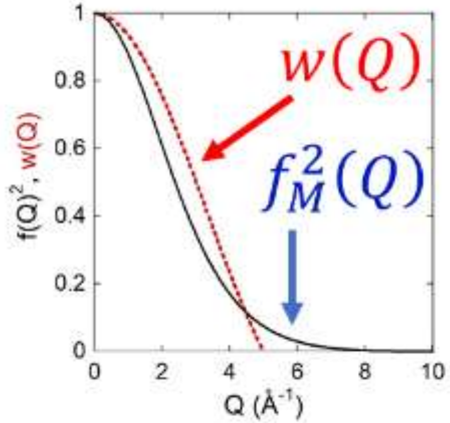
Hints:

Fe^{2+} gS = 4 μ_B

Fe b_c = 9.45 barn

Ti b_c = -3.37 barn

3O b_c = 5.805 barn



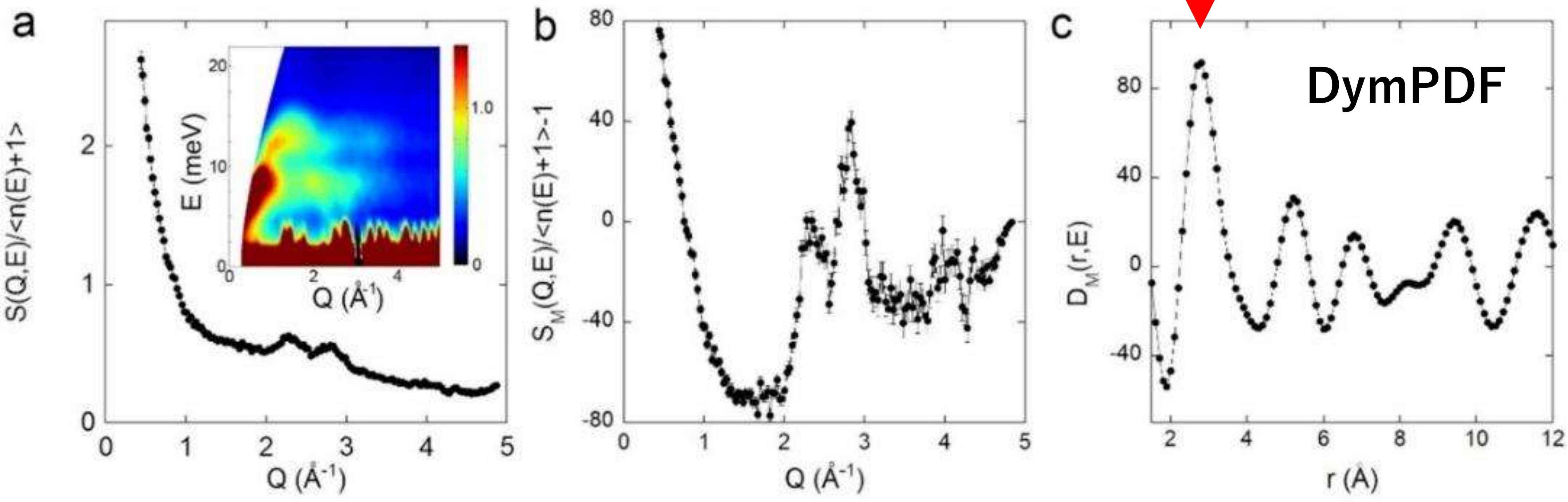
Window Function

Data Calculation Process of DymPDF

sliced at E= 6 meV (T = 8 K)

E_i= 46 meV

3.05 Å



x Q → Fourier transform

K. Iida, K. Kodama, Y. Inamura, M. Nakamura, L.-J. Chang, S. Shamoto, *Sci. Rep.* 12: 20663 (2022).

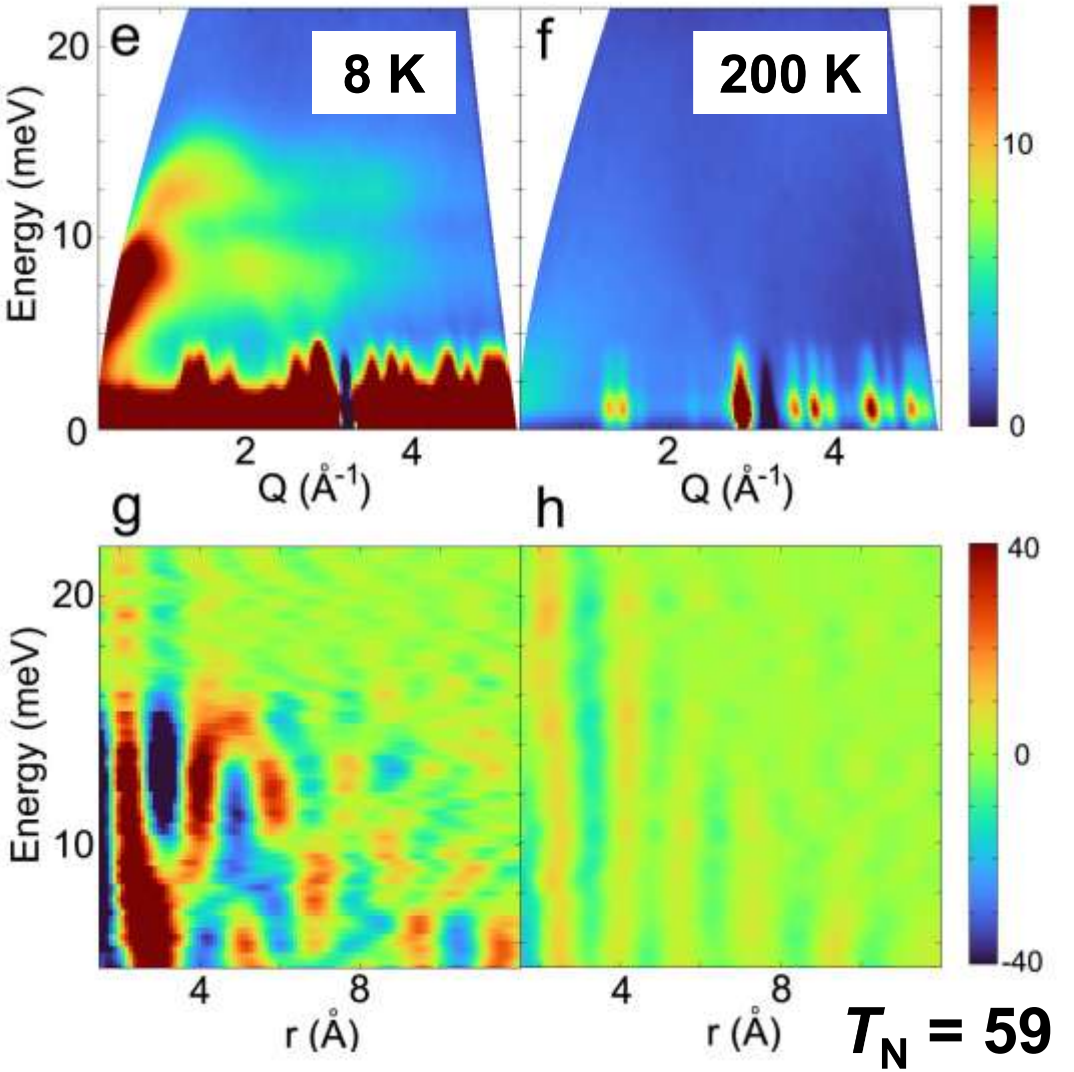
Dynamic magnetic Structure Factor to DymPDF

Dynamic magnetic Structure Factor

Fourier transform

Dynamic magnetic Pair-Density Function (DymPDF)

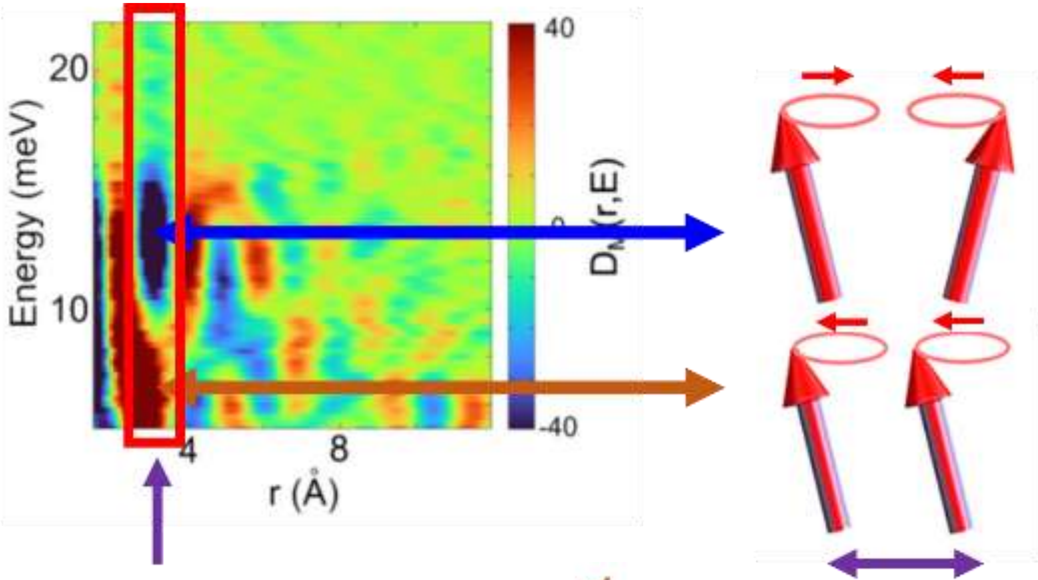
$FeTiO_3 : D_M(r,E)$



T_N = 59 K

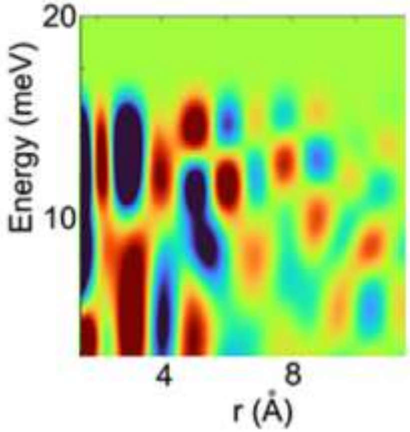
Observation of two spin fluctuation modes

BL01
4SEASONS

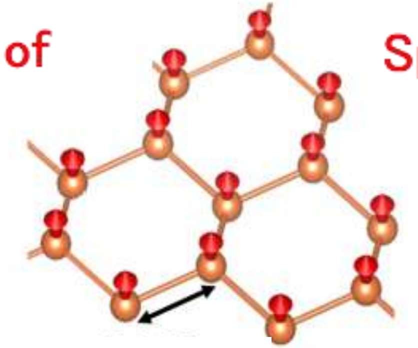


Optical mode

Acoustic mode



Spin distance of
3.05 Å



3.05 Å

Spin distance of
3.05 Å

Simulation
(SpinW+Utsusemi)

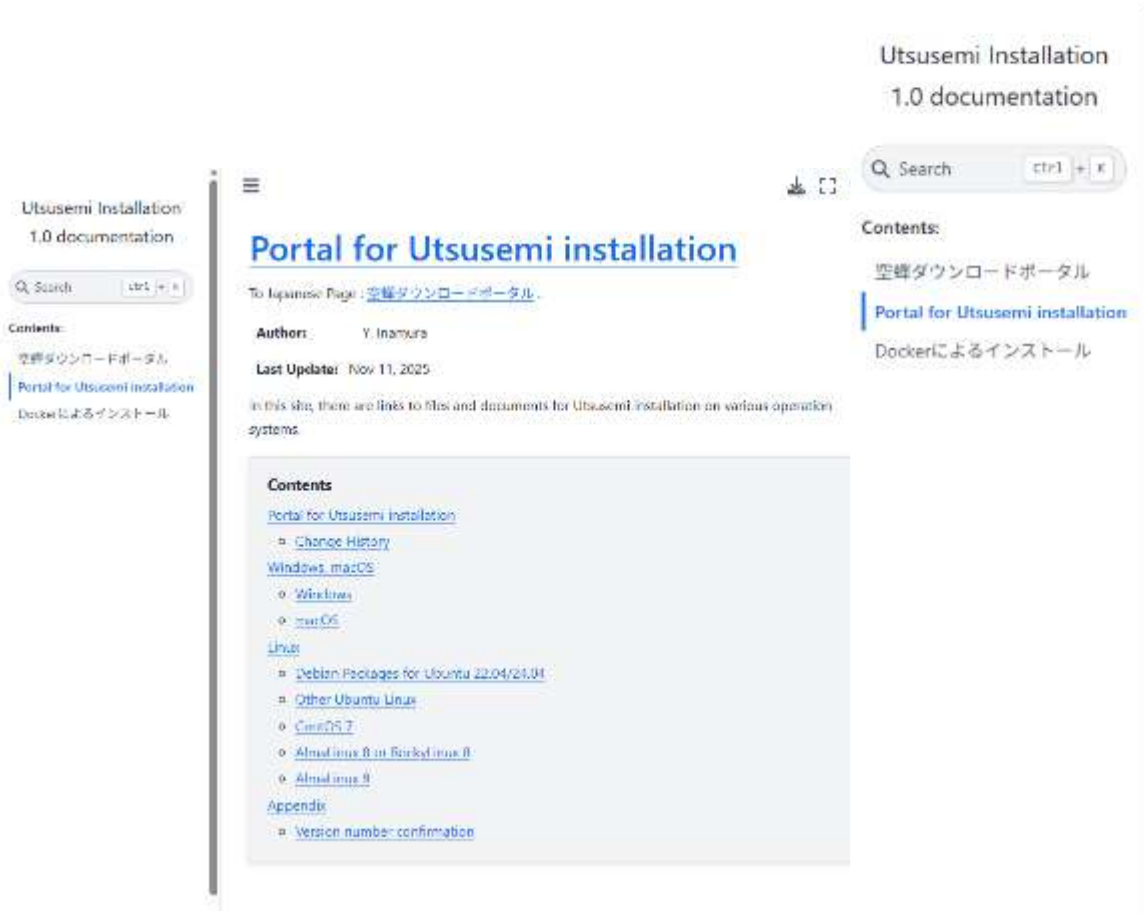
空蟬 UTSUSEMI FOR NEUTRON SCATTERING

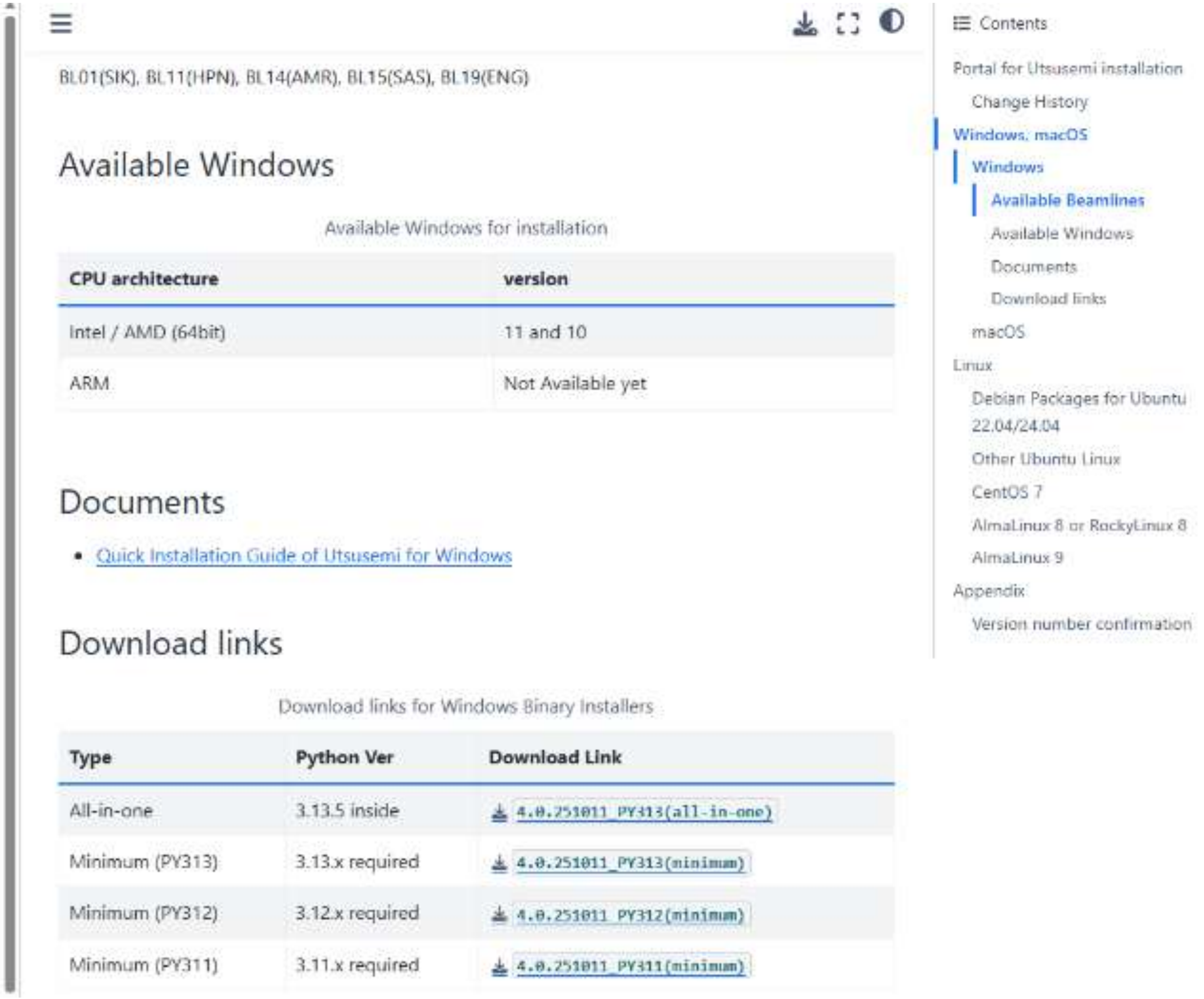
K. Iida, K. Kodama, Y. Inamura, M. Nakamura, L.-J. Chang, S. Shamoto, *Sci. Rep.* 12: 20663 (2022).

3. How to get DymPDF by ‘Utsusemi’

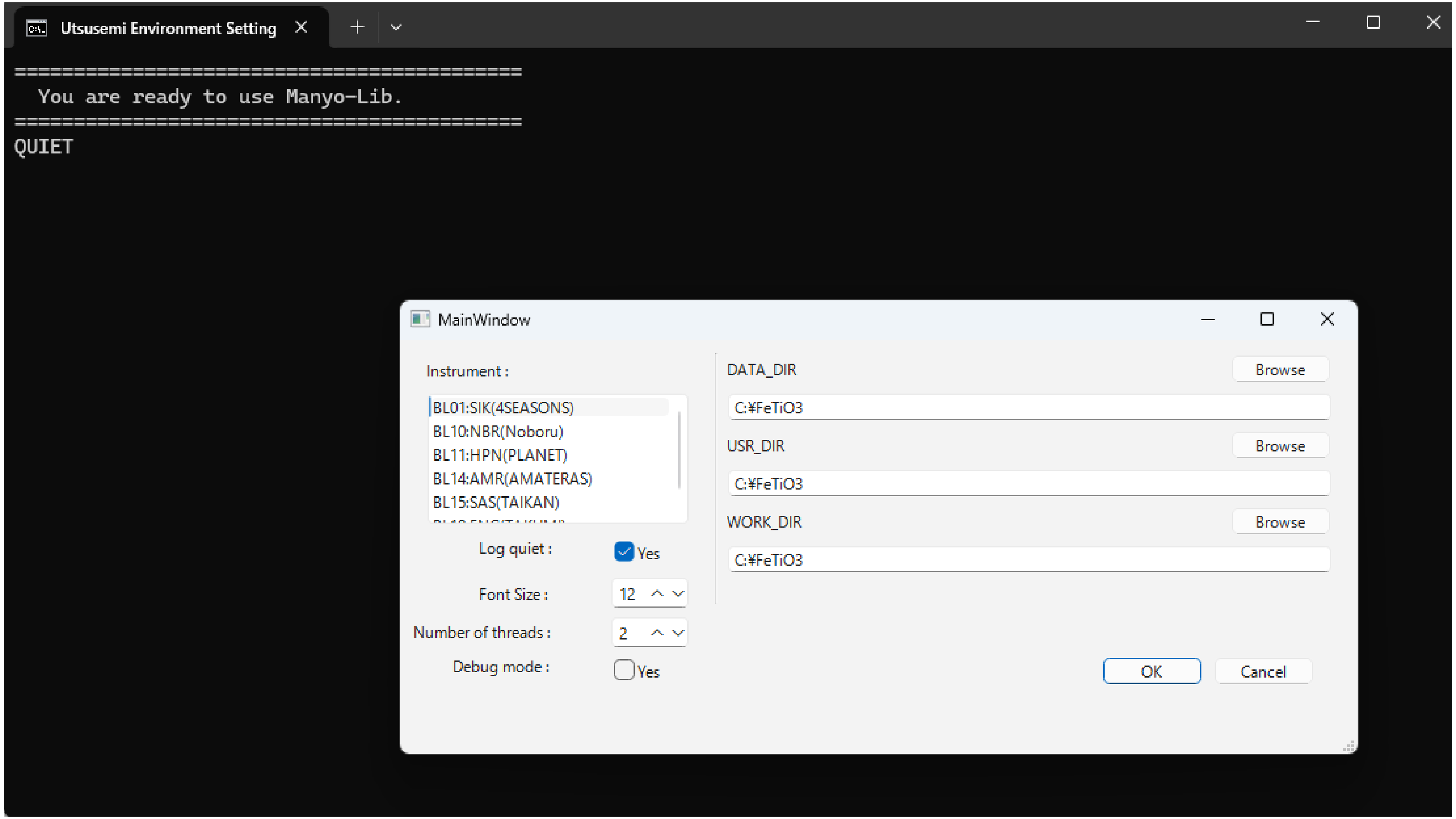
Utsusemi
https://mlfinfo.jp/groups/comp/en/utsusemi.html

Utsusemi Installation
Windows 11, MacOS, Unix (Ubuntu)



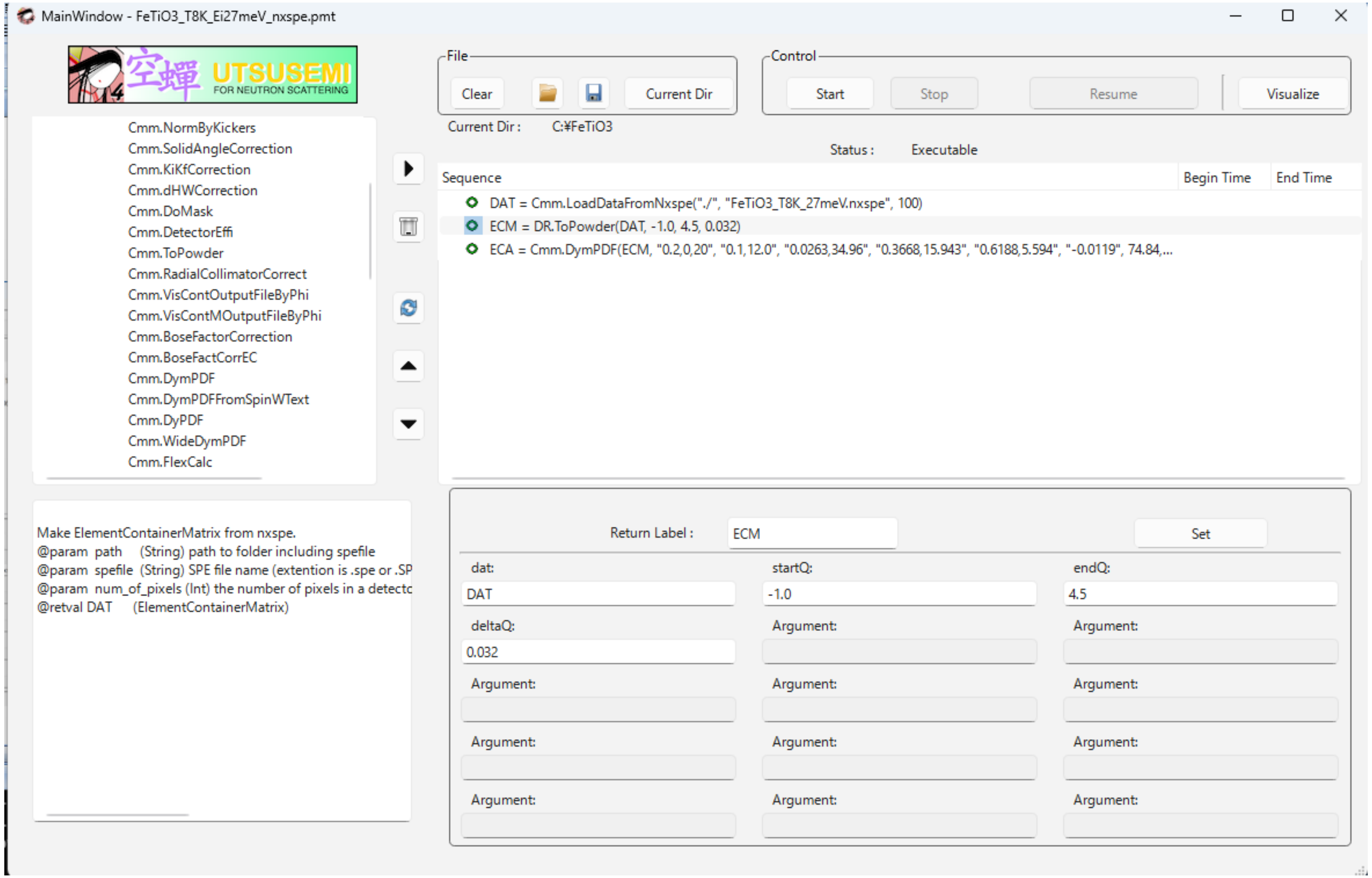


After installation of
‘Utsusemi.4.0.251011.PY313.x86_64.msi’
‘Utsusemi Environment Settings’



Nxspe file

'Sequence Editor', Functions, SIK,
DAT=Cmm.LoadDataFromNxspe
ECM=DR.ToPowder
ECA=Cmm.DymPDF



Available data formats:
SPE, Nxspe, Dump,
Srlz, ManyoBinary,
NeXus

‘Sequence Editor’

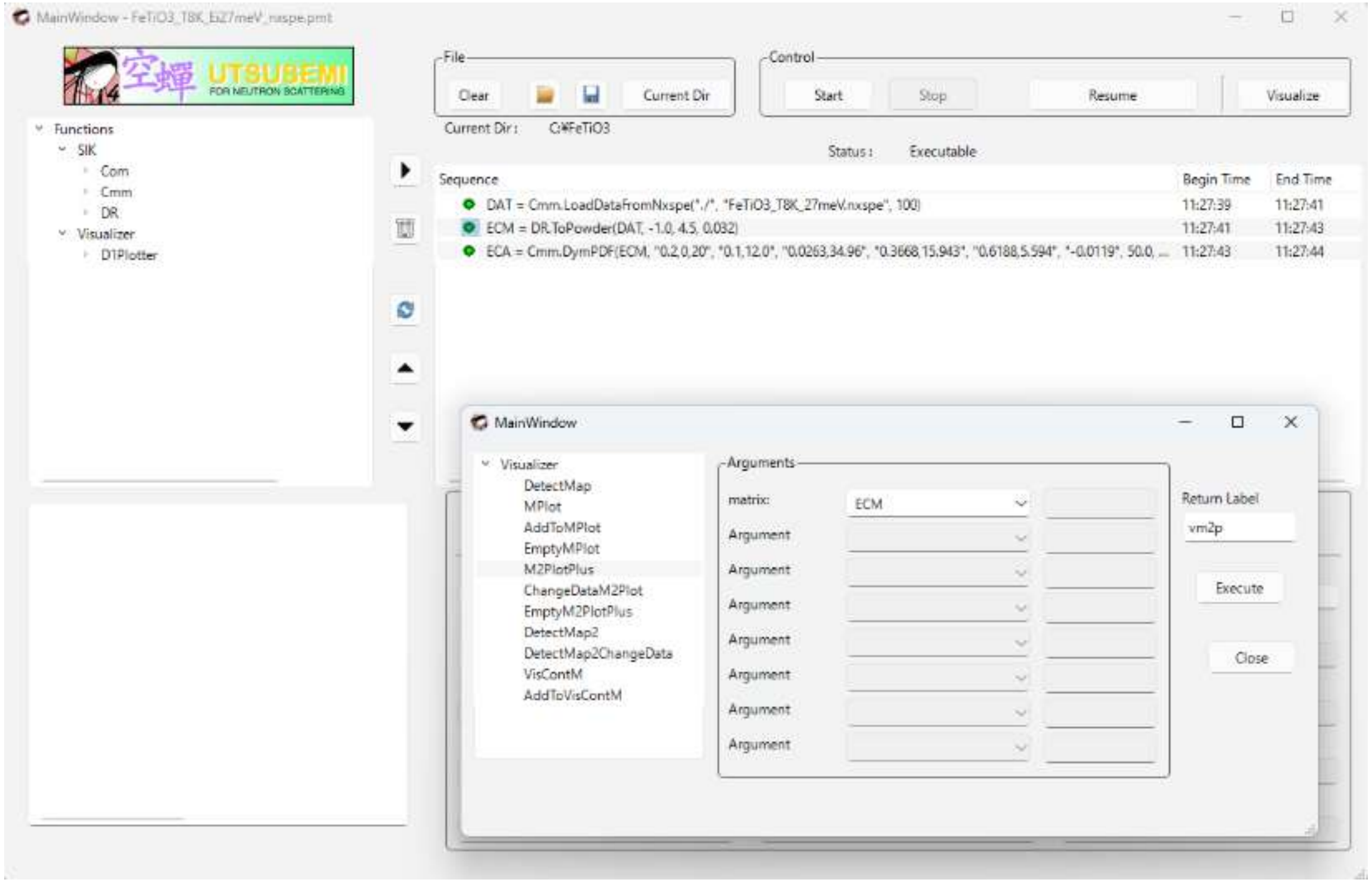
Start

Visualize M2PlotPlus

matrix ECM

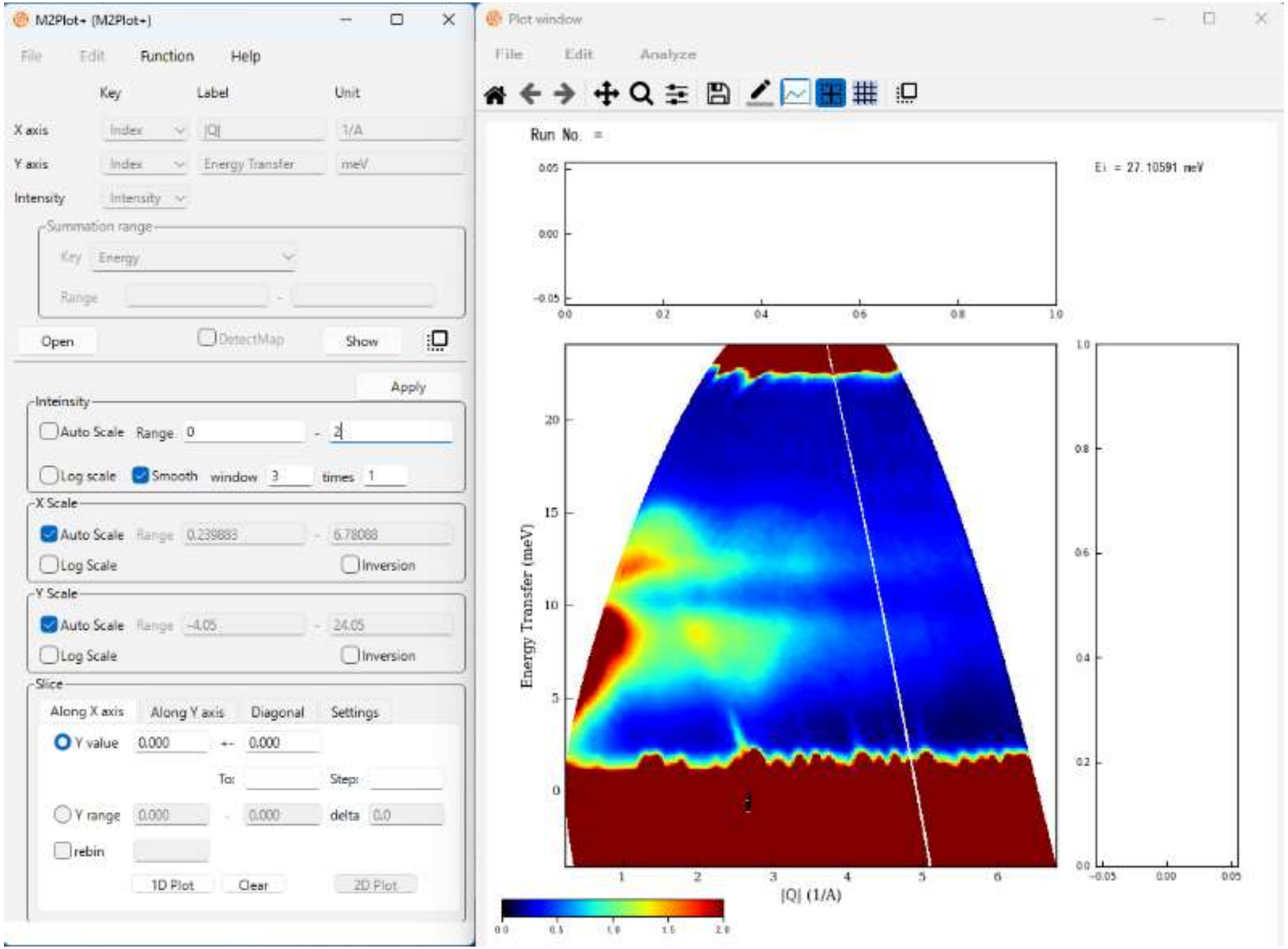
Return Label vm2p

Execute



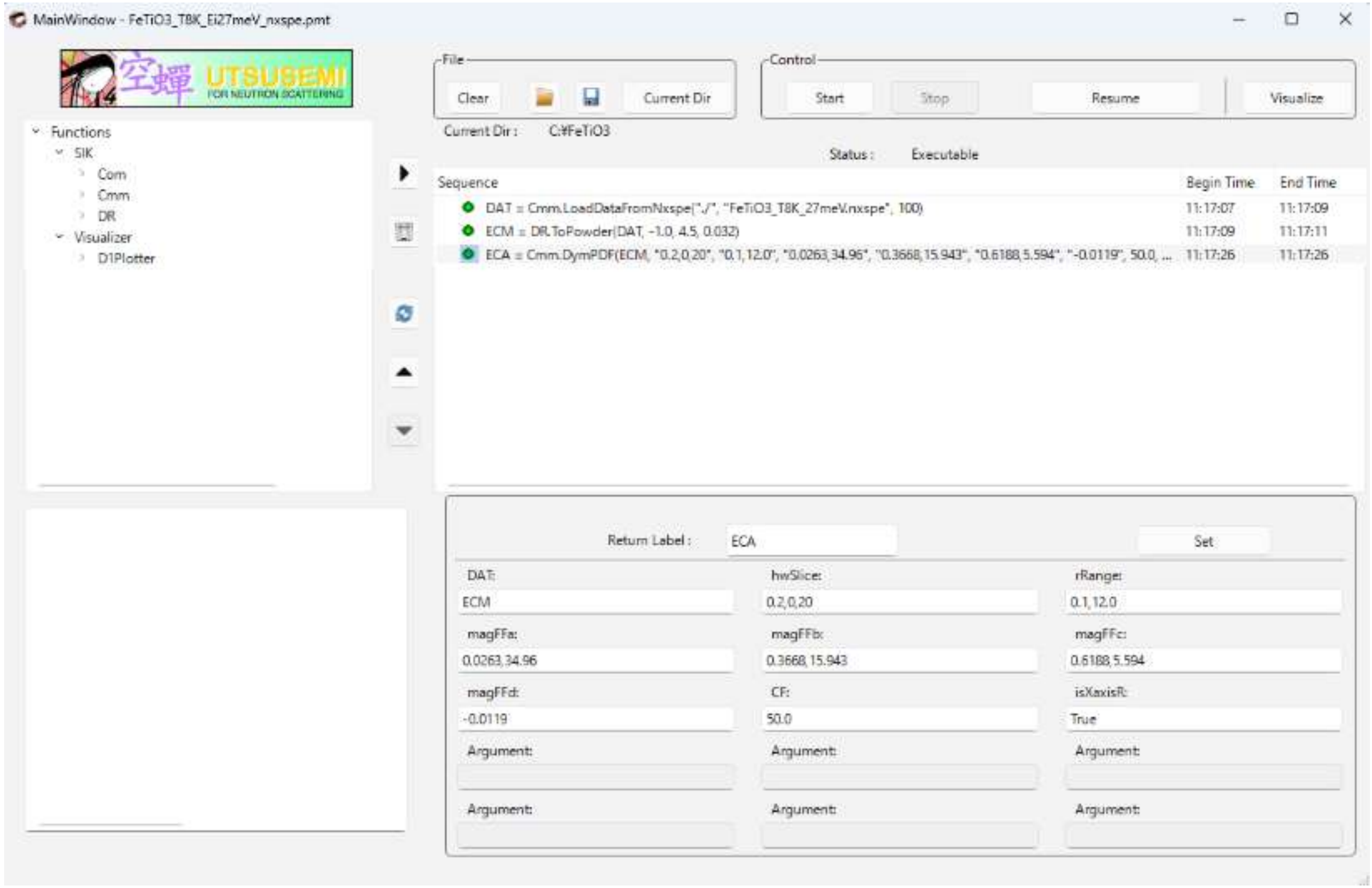
‘Sequence Editor’
M2Plot+

Dynamic structure factor S(Q,E)



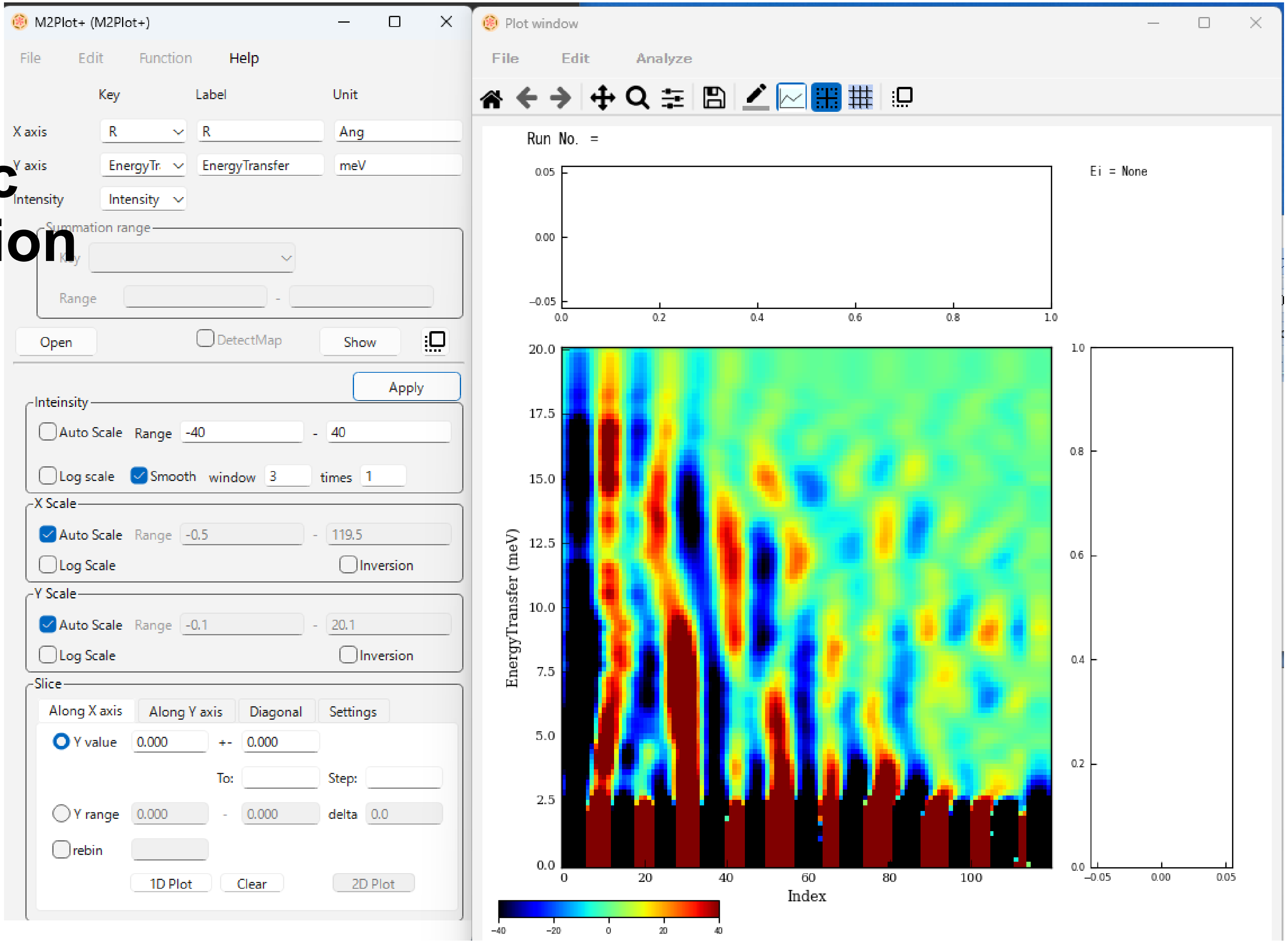
‘Sequence Editor’

Visualize M2PlotPlus
matrix ECA
Return Label vm2p
Execute



‘Sequence Editor’
M2Plot+

Dynamic magnetic pair density function $D_M(r,E)$



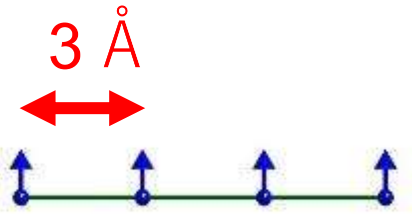
4. How to simulate DymPDF by ‘SpinW’ and ‘Utsusemi’

DymPDF SpinW simulation (MATLAB)

Simple model magnets: S= 1, J_a= -1 meV

1D chain Ferromagnet

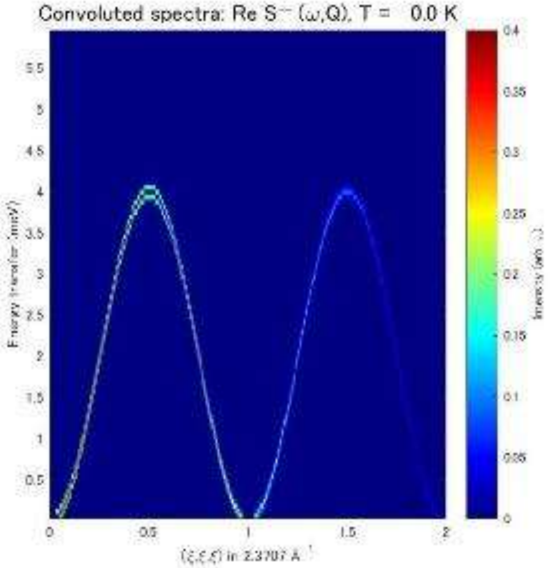
$$H = \sum_{i,j} J_{ij} S_i S_j = J_a (S_i S_{i-1} + S_i S_{i+1})$$

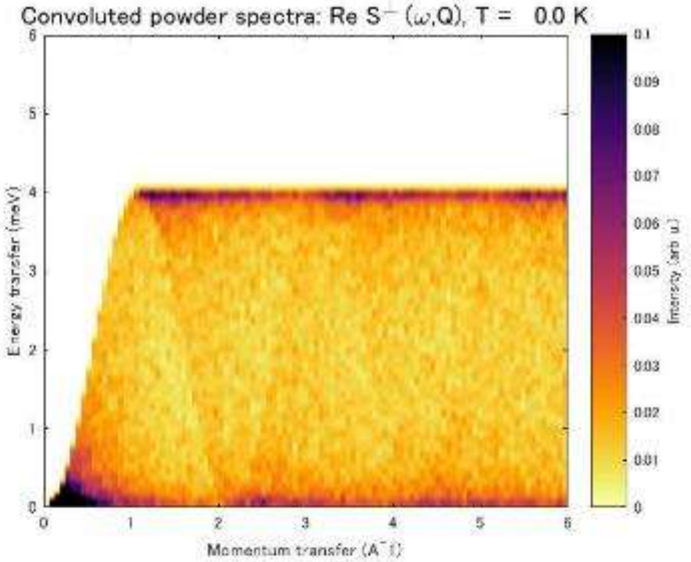


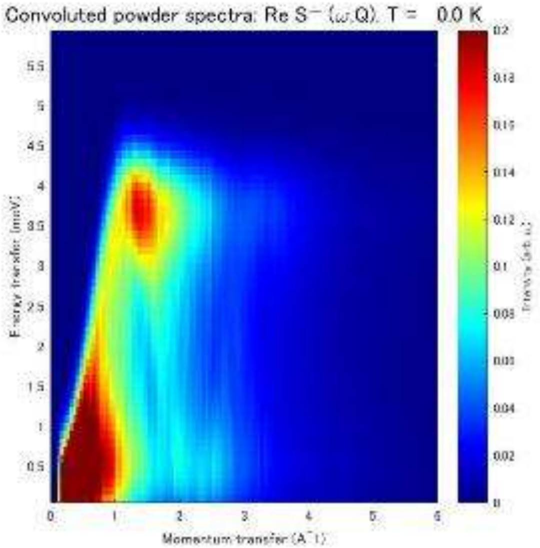
$$E(k) = 2J_a S(1 - \cos ka)$$

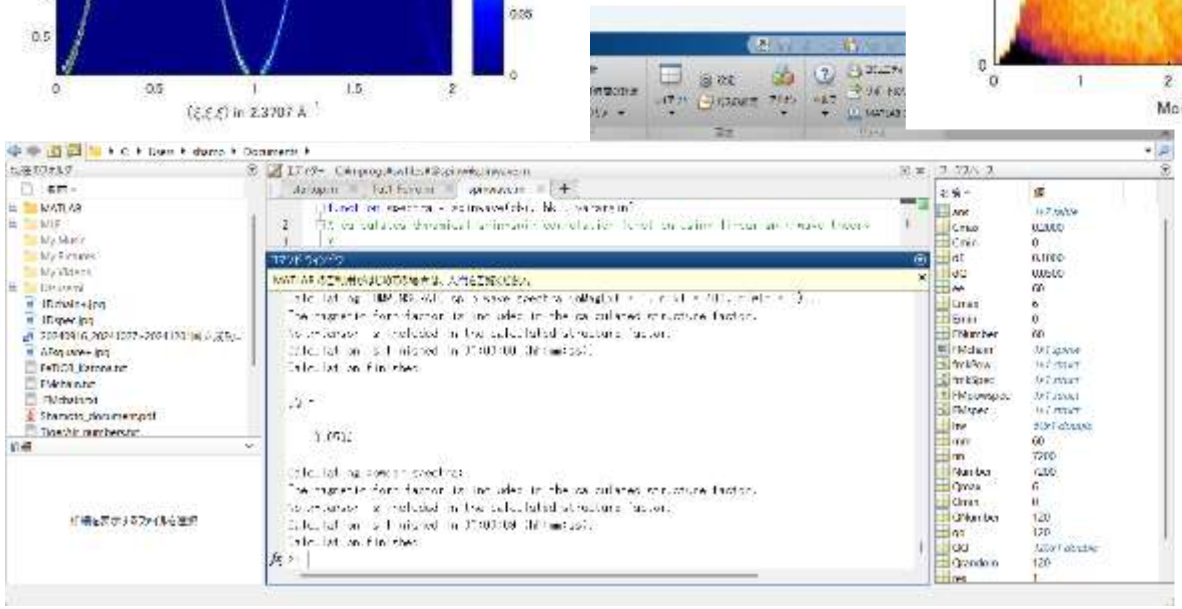
The SpinW software package is based on the linear spin-wave theory with the Holstein–Primakoff approximation.

Tutorial 1 in SpinW homepage “Spin wave spectrum of the Heisenberg ferromagnetic nearest-neighbor spin chain”









https://spinw.org/tutorials/

‘Sequence Editor’, Functions, SIK, ECA=Cmm.DymPDFFromSpinWText

‘Sequence Editor’

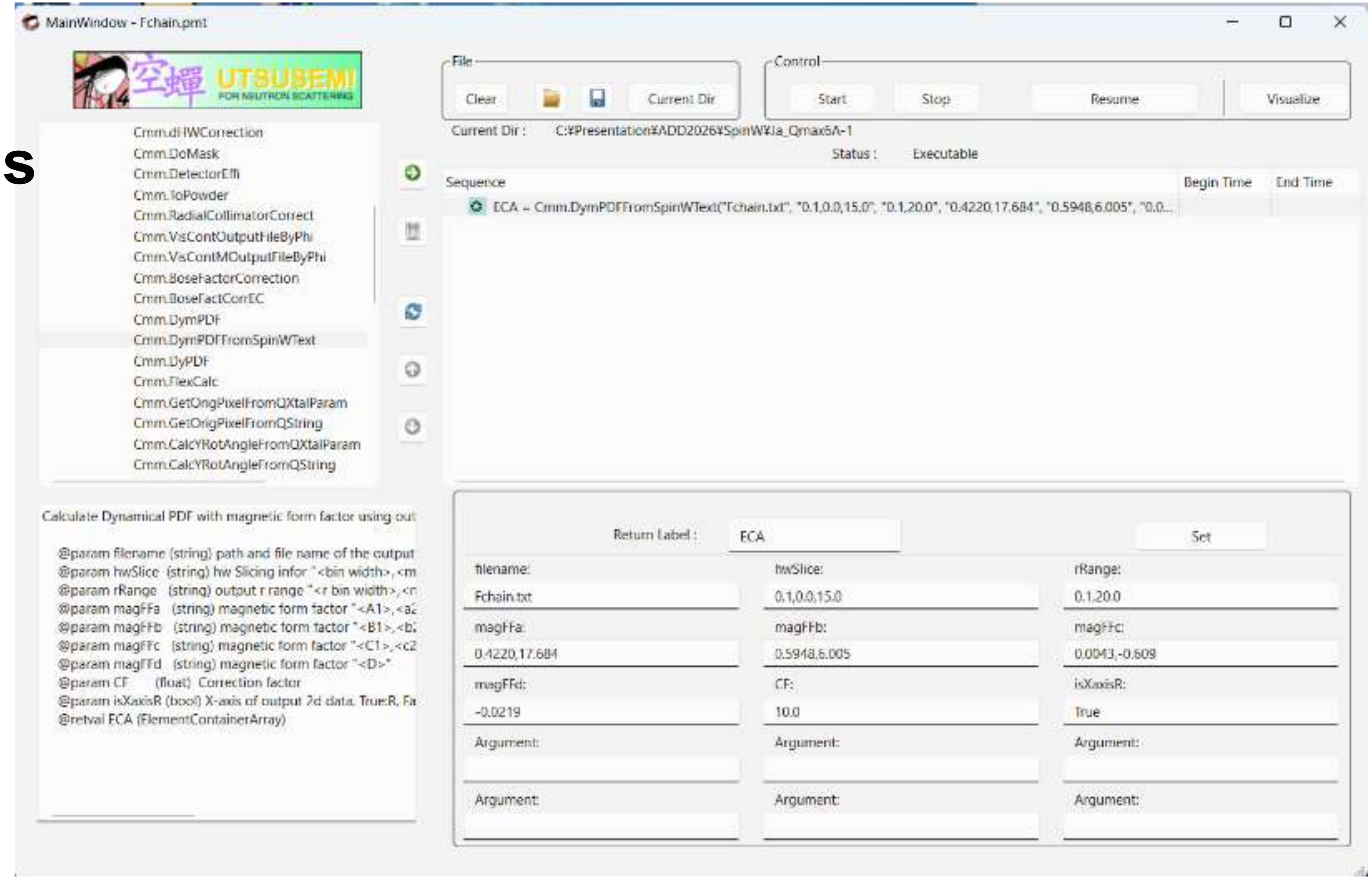
Start

Visualize M2PlotPlus

matrix ECA

Return Label vm2p

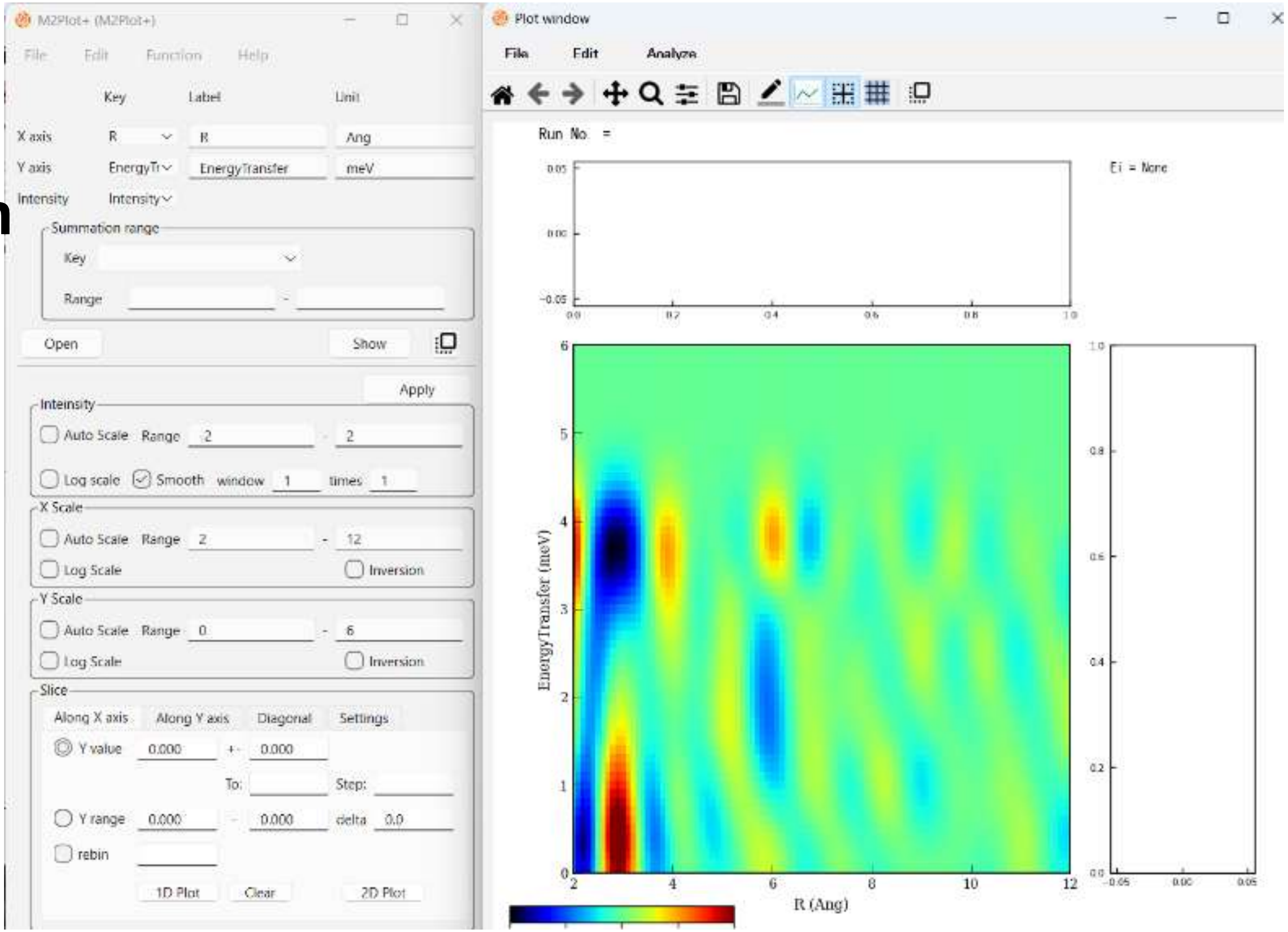
Execute



‘Sequence Editor’

M2Plot+

Dynamic magnetic pair density function $D_M(r,E)$



5. Summary

1990
Crystalline PDF
IPNS, ANL USA

PDFgetN
Billinge, S. J. L.
https://www.diffpy.org/products/pdfgetx.html

PDFFIT
Proffen Th. and Billinge, S. J. L.
https://www.xray.cz/ecm-cd/soft/xray/general/discus/discus/pdffit.html

1992
DyPDF
ISIS, RAL UK

DyPDF (Utsusemi)
https://mlfinfo.jp/groups/comp/en/utsusemi.html

2024 CROSS technical development program

2014
mPDF
ILL France

mPDF
Billinge, S. J. L.
https://mpdf.github.io/

mPDF
Billinge, S. J. L.
https://mpdf.github.io/

2022
DymPDF
J-PARC MLF Japan

DymPDF (Utsusemi)
https://mlfinfo.jp/groups/comp/en/utsusemi.html

2021 CROSS technical development program

SpinW+Utsusemi
https://spinw.org/