3<sup>rd</sup> Lecture NSE

Michihiro Nagao NSF NSE Workshop October 28<sup>th</sup>, 2021



### Learning Goal

- ✓ Understand the length/time range covered by NSE.
- ✓ Understand the principle of NSE operation.
- Understand the types of soft matter problems that can be solved by NSE.
- ✓ Understand the new opportunities for the upgraded NSE at NIST.
- ✓ Understand how to plan a successful NSE experiment.



# Outline

- Dynamic range
- Conventional QENS machine
- Principle of NSE
  - Neutron polarization
  - Larmor precession of neutron spins in a magnetic field
  - Fouier time
  - Measurement of S(Q,t) and S(Q,0)
- NSE data reduction
- Example science on NSE
- Upgrade of the CHRNS-NSE
- Planning a NSE experiment



# **Dynamic Range**



- NSE is one of QENS techniques
- NSE covers smallest Q (biggest size) and longest t (smallest energies or slowest motions) among them



Triple axis spectrometer Monochromatization by Bragg reflection of crystals



#### Time of Flight spectrometer

#### Monochromatization by choppers







Energy resolution 10 - 100  $\mu$ eV







Time of flight instrument (DCS@NCNR)

Energy resolution 10 - 100  $\mu$ eV





To have better energy resolution, i.e., to measure smaller energy exchanged, use smaller incident energy



Energy resolution 10 - 100  $\mu$ eV



To have better energy resolution, i.e., to measure smaller energy exchanged, use smaller incident energy narrower energy distribution





Energy resolution 10 - 100  $\mu$ eV

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To have better energy resolution, i.e., to measure smaller energy exchanged, use smaller incident energy narrower energy distribution

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Neutron wavelength (Å)

15

λ=18Å -> E<sub>n</sub>⊭0.25meV

20

ˈλ=5.7Å -≻ E<sub>n</sub>≈2.5meV

5

Veutron flux (a.u.)

The better the resolution, the smaller the resolution volume and the lower the count rate

# The Idea of NSE

- Traditional define both incident & scattered wavevectors in order to define *E* and *Q* accurately
- Traditional use collimators, monochromators, choppers etc to define both  $k_i$  and  $k_f$
- NSE measure as a function of the difference between appropriate components of k<sub>i</sub> and k<sub>f</sub> (original use: measure k<sub>i</sub>-k<sub>f</sub> i.e. energy change)
- NSE use the neutron's spin polarization to encode the difference between components of k<sub>i</sub> and k<sub>f</sub>
- NSE can use poor monochromatization to increase signal intensity, while maintaining very good resolution



# Neutron Precession in a magnetic field

#### **Neutron Properties**

charge: q = 0mass:  $m_n = 1.675 \times 10^{-27}$  kg life time:  $t_1 = 886.7s$ spin: S = 1/2 [in unit of  $h/(2\pi)$ ] Magnetic moment:  $\mu_n = -9.66 \times 10^{-27}$  J/T



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![](_page_13_Figure_3.jpeg)

The neutron experiences a torque, **N**, from a magnetic field **B** perpendicular to its spin direction

$$\frac{dS}{dt} = N = S \times B$$

Precession with Larmor frequency

$$\omega_L = \gamma B$$

Gyromagnetic ratio:  $\gamma = 1.832 \times 10^8 / s / T$ 

![](_page_13_Figure_9.jpeg)

#### Larmor precession

![](_page_14_Figure_0.jpeg)

![](_page_14_Picture_1.jpeg)

![](_page_15_Figure_0.jpeg)

At NCNR, 0.0001 < J < 0.5 T m, how many turns do you expect when  $\lambda$ =8Å

de Broglie relation: 
$$\lambda = \frac{h}{mv}$$

$$\sim 6 < N(\lambda) = \frac{\varphi}{2\pi} = \frac{\gamma m \lambda}{2\pi h} J = 7370 \times J [T \cdot m] \times \lambda [Å]$$

 $U_{\text{ELAWARE}} \times 10^4$  © Center for Neutron Science

A combination of magnetic fields

$$\varphi = \gamma \frac{\int Bdl}{v} = \gamma \frac{J}{v}$$
$$\Delta \varphi = \gamma \left(\frac{J}{v} - \frac{J}{v}\right) = 0$$

![](_page_16_Picture_2.jpeg)

![](_page_17_Figure_0.jpeg)

![](_page_17_Picture_1.jpeg)

![](_page_18_Figure_0.jpeg)

![](_page_18_Picture_1.jpeg)

![](_page_19_Figure_0.jpeg)

VERSITY OF ELAWARE © Center for Neutron Science

#### Scattering event: neutron beam

![](_page_20_Figure_1.jpeg)

• elastic scattering

$$\overline{\varphi} = \left( \gamma \left( \frac{J}{v} - \frac{J}{v} \right) \right)_{f(\lambda)} = 0$$
INVERSITY OF ELAWARE.

 $P_x \approx 1$ 

#### Scattering event: neutron beam

![](_page_21_Figure_1.jpeg)

• quasielastic scattering

$$\bar{\varphi} = \left\langle \gamma \frac{J}{v} - \gamma \frac{J}{v + \delta v} \right\rangle_{f(\lambda)} \neq 0$$

![](_page_21_Picture_4.jpeg)

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 $P_{\chi} \neq 1$ 

Relation of 
$$\langle \varphi \rangle = \omega t = \frac{\Delta E}{\hbar} t$$
  
 $\langle \varphi \rangle = \langle -\frac{\gamma J}{v} + \frac{\gamma (J + \delta J)}{v + \delta v} \rangle = \frac{\gamma m}{h} \langle -\lambda J + (\lambda + \delta \lambda)(J + \delta J) \rangle \approx \frac{\gamma m}{h} (J \delta \lambda + \lambda \delta J)$ 

On the other hand, we know energy exchange can be written

$$\Delta E = \hbar \omega = \frac{h^2}{2m} \left[ \frac{1}{\lambda^2} - \frac{1}{(\lambda + \delta \lambda)^2} \right] \approx \frac{h^2}{m} \frac{\delta \lambda}{\lambda^3} \longrightarrow \delta \lambda = \frac{m\lambda^3}{2\pi h} \omega = \frac{m\lambda^3}{h^2} \Delta E$$
Then,
$$\langle \varphi \rangle \approx \left[ \frac{\gamma m^2 J \lambda^3}{2\pi h^3} \Delta E + \frac{\gamma m \lambda}{h} \delta J \right]$$
Fourier time
When  $\delta J = 0$ 
(symmetric condition)
$$\langle \varphi \rangle \approx \frac{\gamma m^2 J \lambda^3}{2\pi h^3} \Delta E = \frac{\gamma m^2 J \lambda^3}{2\pi h^2} \omega \longrightarrow t = \frac{\gamma m^2 J \lambda^3}{2\pi h^2} = \frac{m\lambda^2}{h} N(\lambda)$$

![](_page_22_Picture_3.jpeg)

![](_page_23_Figure_0.jpeg)

![](_page_23_Picture_1.jpeg)

![](_page_24_Figure_0.jpeg)

![](_page_24_Picture_1.jpeg)

![](_page_25_Figure_0.jpeg)

 $\tilde{E}_{c}$  © Center for Neutron Science

# In reality, wavelength distribution $f(\lambda)$

$$I_{NSE} \propto \int \int_{-\infty}^{\infty} f(\lambda) S(Q, \frac{\Delta E}{\hbar}) \frac{1 + \cos(\varphi)}{2} d\frac{\Delta E}{\hbar} d\lambda = \frac{1}{2\hbar} \left[ \int \int_{-\infty}^{\infty} f(\lambda) S(Q, \frac{\Delta E}{\hbar}) d\Delta E d\lambda + \int \int_{-\infty}^{\infty} f(\lambda) S(Q, \frac{\Delta E}{\hbar}) \cos(\varphi) d\Delta E d\lambda \right]$$
  
Wavelength resolution smeared  $S(Q)$ 
$$\int d\lambda \int_{-\infty}^{\infty} d\Delta E f(\lambda) S(Q, \frac{\Delta E}{\hbar}) \cos\left[ \frac{\gamma m^2 J \lambda^3}{2\pi \hbar^3} \Delta E + \frac{\gamma m \lambda}{\hbar} \delta J \right] = \int d\lambda \int_{-\infty}^{\infty} d\Delta E f(\lambda) S(Q, \frac{\Delta E}{\hbar}) \cos\left( \frac{\gamma m \lambda}{\hbar} \delta J \right)$$
$$= \int f(\lambda) \cos\left( \frac{\gamma m \lambda}{\hbar} \delta J \right) d\lambda \times \int_{-\infty}^{\infty} S(Q, \frac{\Delta E}{\hbar}) \cos\left( \frac{\Delta E}{\hbar} t \right) d\Delta E$$

![](_page_26_Figure_2.jpeg)

$$I_{NSE} \propto S(Q) + S(Q,t) \exp\left(-\Lambda^2 \gamma^2 \frac{m^2}{h^2} \delta J^2\right) \cos\left(\gamma \frac{m}{h} \lambda \delta J\right)$$

 $\Lambda:$  relating to the wavelength spread  $\Delta\lambda$ 

Period of oscillation  $\propto \lambda$ 

Decay of oscillation relates to the wavelength distribution

![](_page_26_Picture_7.jpeg)

# **NSE principles - summary**

- If a spin rotates anticlockwise & then clockwise by the same amount it comes back to the same orientation
  - Need to reverse the direction of the applied field
  - Independent of neutron speed provided
- The same effect can be obtained by reversing the precession angle at the mid-point and continuing the precession in the same sense
   Use a π rotation (π flipper)
- If the neutron's velocity is changed by the sample, its spin will not come back to the same orientation
  - The difference will be a measure of the change in the neutron's speed or energy.

![](_page_28_Figure_0.jpeg)

![](_page_29_Figure_0.jpeg)

![](_page_30_Figure_0.jpeg)

# NSE: coherent vs incoherent

$$\frac{S(Q,t)}{S(Q,0)} = \frac{S_{coh}(Q,t) - \frac{1}{3}S_{inc}(Q,t)}{S_{coh}(Q,0) - \frac{1}{3}S_{inc}(Q,0)}$$

NSE is known for the investigation of the coherent dynamics.

Incoherent scattering intensity is reduced to -1/3 in NSE. The best achievable flipping ratio is 0.5.

![](_page_31_Figure_4.jpeg)

![](_page_31_Picture_5.jpeg)

# Spin-flip/Non-spin-flip Scattering

Sample scattering events sometimes involve with spin-flip scattering

Coherent scattering: Non-Spin-Flip Scattering Isotope incoherent scattering: Non-Spin-Flip Scattering Spin incoherent scattering: Spin-Flip Scattering -- 2/3 Spin-Flip probability  $I_{NSF} = I_{coh} + I_{i-inc} + \frac{1}{3}I_{s-inc}$   $I_{cotal} = I_{coh} + I_{i-inc} + I_{s-inc} = I_{NSF} + I_{SF}$  $I_{SF} = \frac{2}{3}I_{s-inc}$ 

Separation of coherent + isotope-incoherent from spin-incoherent scattering

$$I_{coh} + I_{i-inc} = I_{NSF} - \frac{1}{2}I_{SF}$$
  $I_{s-inc} = \frac{3}{2}I_{SF}$ 

![](_page_32_Picture_5.jpeg)

### NSE: coherent vs incoherent

$$\frac{S(Q,t)}{S(Q,0)} = \frac{S_{coh}(Q,t) - \frac{1}{3}S_{inc}(Q,t)}{S_{coh}(Q,0) - \frac{1}{3}S_{inc}(Q,0)}$$

NSE is known for the investigation of the coherent dynamics.

Incoherent scattering intensity is reduced to -1/3 in NSE. The best achievable flipping ratio is 0.5. However, the main limitation to Othe study of incoherent of scattering by NSE is the Q coverage of instrument.

Recent advancements in NSE instrumentation aim to overcome this limitation (WASP at ILL).

![](_page_33_Figure_5.jpeg)

Most important is to avoid *Q* areas where coherent and incoherent intensity cancel each other.

![](_page_33_Picture_7.jpeg)

![](_page_34_Figure_0.jpeg)

#### Data reduction: intensity vs phase (echo signal) and t

![](_page_35_Figure_1.jpeg)

#### Data reduction: intensity vs phase (echo signal) and t

![](_page_36_Figure_1.jpeg)

#### Polarized intensity vs phase (echo signal)

![](_page_37_Figure_1.jpeg)

Fitting the echo  
Fitting the echo  

$$I_{NSE} \propto S(Q) + S(Q, t) \exp\left(-\Lambda^2 \gamma^2 \frac{m^2}{h^2} \delta J^2\right) \cos\left(\gamma \frac{m}{h} \lambda \delta J^2\right)$$

$$I_{NSE} \propto S(Q) + S(Q, t) \exp\left(-\Lambda^2 \gamma^2 \frac{m^2}{h^2} \delta J^2\right) \cos\left(\gamma \frac{m}{h} \lambda \delta J^2\right)$$

$$I_{0} \text{ Average Intensity}$$

$$I_{0} \text{ Echo point}$$

$$I_{0} \text{ Echo point}$$

$$I_{0} \text{ Echo point}$$

$$I_{0} \text{ Echo width, function}$$

$$I_{0} \text{ Echo width, function of } (\lambda)$$

$$I_{0} \text{ Echo width distribution}$$

$$I_{0} \text{ Echo width distribution}$$

$$I_{0} \text{ Echo width distribution}$$

$$I_{0} \text{ Echo point}$$

$$I_{0} \text$$

# The physical information is all in the amplitude

$$\frac{I(Q,t)}{I(Q)} \propto \frac{2A}{Up - Dwn}$$

# Incidentally, in this way, both polarization and detector efficiency effects are taken care off.

![](_page_39_Picture_3.jpeg)

# A small portion of the echo will do

![](_page_40_Figure_1.jpeg)

"/var/nse/m3514" 10% DS/D20 1mm Q=0p06 Q=5.984e+08 m-1 t=5.03804e-10 s

![](_page_40_Picture_3.jpeg)

#### Polarized intensity vs t

![](_page_41_Figure_1.jpeg)

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# **Resolution normalization**

![](_page_42_Figure_1.jpeg)

Even for an elastic scatterer the echo signal will decrease with the increase of the Fourier time. Inhomogeneities in the • magnetic field will depolarize the beam. In Neutron Spin-Echo the resolution can be simply divided out from the data.

![](_page_42_Picture_3.jpeg)

![](_page_43_Figure_0.jpeg)

![](_page_43_Picture_1.jpeg)

### In reality, 2D detector

![](_page_44_Figure_1.jpeg)

![](_page_44_Figure_2.jpeg)

Each pixel has an echo

at multiple Fourier time points

+ multiple scattering angles

Data reduction software is available Data Analysis and Visualization Environment DAVE @ NCNR

#### Question: What do the blue lines define?

#### 2D detector: 32 x 32 pixels with 1 cm<sup>2</sup> resolution

![](_page_44_Picture_9.jpeg)

#### Phase map

![](_page_45_Figure_1.jpeg)

Phase (=echo point) varies with detector pixel. Why?

#### Phase map

![](_page_46_Figure_1.jpeg)

Phase (=echo point) varies with detector pixel.

Why?

Neutron trajectories are different. Each neutron trajectory, the magnetic field integral *J* may be different.

Difference in J ( $\delta J$ ) changes the precession angle: potential reason to reduce the instrument resolution

$$\langle \varphi \rangle \approx \left[ \frac{\gamma m^2 J \lambda^3}{2\pi h^3} \Delta E + \frac{\gamma m \lambda}{h} \delta J \right]$$

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# Science on NSE

#### **Coherent dynamics**

Density fluctuations corresponding to some SANS pattern length (Å)

Diffusion Shape fluctuations (Internal dynamics) Polymer dynamics Liquids and Glass systems

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Incoherent dynamics

Self-dynamics (hydrogen atoms)

Magnetic dynamics

Spin glasses

![](_page_47_Figure_8.jpeg)

![](_page_47_Picture_9.jpeg)

# Self-diffusion of a particle - diffusion

Here, we assume self-part of the van Hove correlation function that follows Gaussian shape

$$G_{s}(r,t) = \frac{1}{(2\pi)^{\frac{3}{2}}\sigma^{3}(t)}e^{-\frac{r^{2}}{2\sigma^{2}(t)}}$$

Experiment and theory suggest the Gaussian width follows  $\sigma(t) = \sqrt{2Dt}$ 

The second moment of  $G_s(r,t)$ corresponds to the mean square displacement (MSD),  $\langle r^2 \rangle$ 

A property of Gaussian functions  $\langle e^{iQ[r_i(t)-r_j(0)]} \rangle = e^{-\frac{Q^2}{6} \langle |r_i(t)-r_j(0)|^2 \rangle}$ 

$$\sigma(t)$$

$$|r^2\rangle = \int_{-\infty}^{\infty} r^2 G_s(r,t) dr = 6Dt$$

$$S(Q,t) = e^{-DQ^2t}$$

# Self-diffusion of a particle - diffusion

A particle in interest undergoes many collisions with neighboring particles

Trajectory is a random walk

Probability distribution  $G_s(r, t)$  is the solution of the diffusion equation  $\frac{\partial G_s(r, t)}{\partial t} = D\nabla^2 G_s(r, t) \qquad D \text{ is the diffusion constant}$  $G_s(r, t) = (4\pi Dt)^{-3/2} e^{-\frac{r^2}{4Dt}}$ 

Space Fourier Transform

$$S(Q,t) = e^{-DQ^2t}$$

![](_page_49_Picture_6.jpeg)

# **Diffusing colloidal particles**

![](_page_50_Figure_1.jpeg)

#### In general...

Decay gets faster as Q increases

- Smaller Q -> larger length scale
  - Larger objects move slower
- Larger Q -> smaller length scale
  - Smaller objects move faster

![](_page_50_Figure_8.jpeg)

![](_page_51_Figure_0.jpeg)

### Diffusion of microemulsion droplets

S.-C. Wang, P. Mirarefi, A. Faraone, and C. Ted Lee, Jr., *Biochemistry* 50, 8150 (2011).

### Diffusion and internal dynamics of proteins

#### Lysozyme solution + azoTAB surfactant

![](_page_52_Figure_3.jpeg)

8 0.05

0.1

![](_page_52_Figure_4.jpeg)

![](_page_52_Figure_5.jpeg)

![](_page_52_Picture_6.jpeg)

0.15 Q (Å<sup>-1</sup>) 0.2

0.25

#### When beads are connected – polymer dynamics

![](_page_53_Figure_1.jpeg)

#### MSD for a chain segment

![](_page_54_Figure_1.jpeg)

![](_page_54_Picture_2.jpeg)

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![](_page_55_Figure_0.jpeg)

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E. Senses, S.M. Ansar, C.L. Kitchen, Y. Mao, S. Narayanan, B. Natarajan, and A. Faraone, Phys. Rev. Lett. 118, 147801 (2017).

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#### 1000 More neutrons! Cold source upgrade at the 100 NCNR (planned in 2023) Smaller Q to access larger length 10 scales! t (ns) Use longer wavelength neutrons λ=17Å 0.1 λ=14Å Larger t to access longer time λ=11Å scales! λ=8Å 0.01 λ=6Å

λ=4.5Å

#### Future direction of the NIST-CHRNS-NSE

- Use longer wavelength
  - neutrons and increase magnetic field integral

 $\underline{m}^2$ 

 $t = \gamma_{\overline{z}}$ 

 $Q(Å^{-1})$ 

<sup>9</sup>0.1

0.001

#### NCNR cold source upgrade

![](_page_57_Figure_1.jpeg)

![](_page_57_Picture_2.jpeg)

20 30

3.0

20

10

10

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### Improve magnetic field strength

![](_page_58_Picture_1.jpeg)

- Coil shape determines J<sub>max</sub>
- Neutrons at different trajectories feel different J, which limits ability of NSE spectroscopy

Asymmetric Coil Shape = Optimized for NSE

![](_page_58_Figure_5.jpeg)

![](_page_58_Figure_6.jpeg)

Design on IN15 at ILL by Bela Farago

Design on J-NSE-Phoenix at JCNS by Michael Monkenbusch

![](_page_58_Picture_9.jpeg)

#### Improve magnetic field inhomogeneity

![](_page_59_Figure_1.jpeg)

$$r^2 = x^2 + y^2$$

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Spiral cut to realize an array of concentric loops with

$$r \propto \sqrt{n}$$

will compensate r<sup>2</sup> inhomogeneity Mezei, *Lect. Notes Phys.* **128**, 178 (1979).

![](_page_59_Figure_7.jpeg)

Version at Juelich

Fresnel coil: technology from the 1970s

Version at ILL

#### Gains

![](_page_60_Figure_1.jpeg)

- Improving data rate a factor 10 to access 100 ns (currently by 11 Å, in future by 8 Å)
  - More gain at longer wavelengths
- Routine operation up to 300 ns
  - Currently 100 ns
- Maximum achievable time to 700 ns
  - Current record 300 ns
- Out reach & education
  - This workshop!

![](_page_61_Figure_0.jpeg)

# Planning a NSE experiment

- Which dynamics do you want to identify?
  - Prepare a few clear targets why you measure dynamics of which system
- Most probably you would like to know the structure before even thinking to study dynamics
  - Dynamics are measured simultaneously with static structures
  - Intensity distributions including coherent/incoherent
- Guestimate timescale of the motion
  - NMR?, dielectric spectroscopy?, DLS?, NBS?, simulation?...
- Gather enough amount of materials
  - Typical sample size 30 mm x 30 mm x 1 4 mm
- Discuss with an expert

![](_page_62_Picture_11.jpeg)

![](_page_62_Figure_12.jpeg)

### Learning Goal

- Understand the length/time range covered by NSE
  - Nanometer and nanosecond scales
- Understand the principle of NSE operation
  - Larmor precession to decouple instrument and probe energy resolution
- Understand the types of soft matter problems that can be solved by NSE
  - Various coherent soft matter dynamics
- Understand the new opportunities for the upgraded NSE at NIST
  - Extended Fourier time range with increased data rate
- Understand how to plan a successful NSE experiment
  - Better to (almost must) know static structures, before dynamics studies

# Presentation 10/29 10:10 am

 Use what you learned from this workshop to present your research that can be benefit from NSE (the one in your application or something else). Send it to Kuo-Chih by 11:59 pm kuo-chih.shih@nist.gov

#### **Proposal Review (Optional)**

• If you like, you can submit a proposal through our IMS proposal system (we will tell you how to do this, of course) and reviewers will review your proposal and give you feedback.

![](_page_64_Picture_4.jpeg)

**Polymer: Main Conference Room** 

Membrane: Key Room

Protein: Tubman Room

![](_page_65_Picture_3.jpeg)