Understanding Radiation-Induced Polarization Dynamics in Polarized Targets

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Tensor Collaboration Meeting

Jefferson Lab

3-5 June 2026

U.S. DEPARTMENT OF ENERGY | Office of Science

UNIVERSITY of VIRGINIA

Jefferson Lab

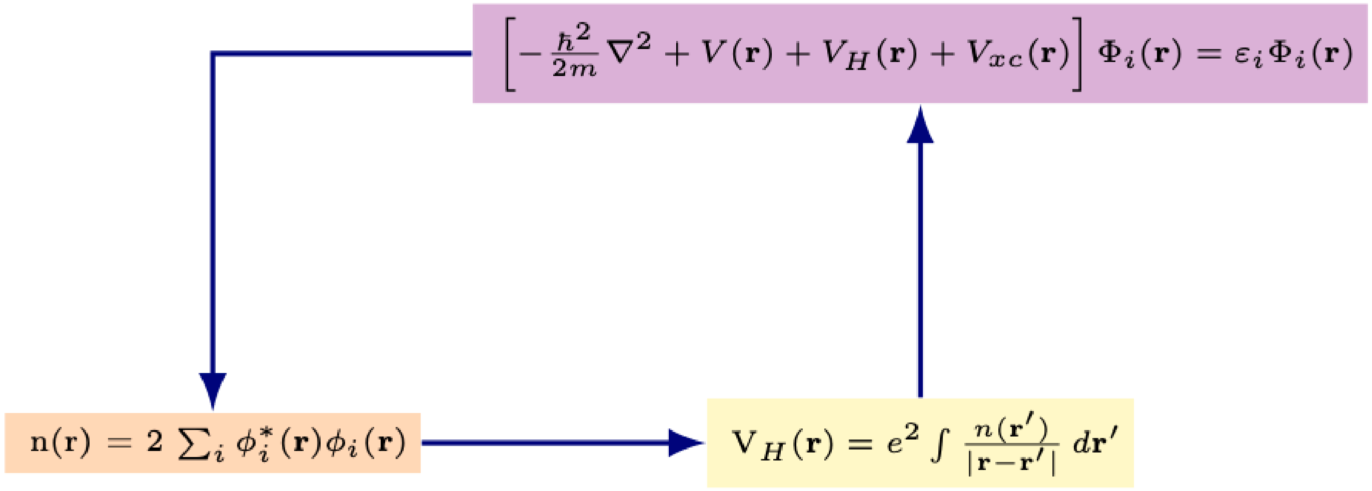
Motivation

- NH_3 and ND_3 have been used as polarized targets for decades.
- Yet we still lack a microscopic explanation of Polarization and relaxation mechanisms.
- Cold irradiation dose behavior of ND_3.
- We aim to connect the radiation-induced radical population and lattice dynamics to the observed polarization behavior.

Recap from the last meeting

- Have been using DFT to calculate fundamental solid-state properties of target systems.

Self Consistency of Kohn-Sham Equation



ESR Parameters

- Expectation values for different electronic, nuclear and cross operators.

The g-Tensor:

Determines the position of the signal in the ESR spectra. There are four contributions to it.

$$\mathbf{g} = g_e \mathbf{1} + \mathbf{g}^{\mathrm{RMC}} + \mathbf{g}^{\mathrm{DSO}} + \mathbf{g}^{\mathrm{PSO}} \quad (1)$$

Spin Zeeman / Isotropic part: free electron g value is 2.002319

$$g_{\mu\nu}^{\mathrm{SZ}} = \delta_{\mu\nu} g_e . \quad (2)$$

Relativistic Mass Correction

$$g^{\mathrm{RMC}} = -\frac{\alpha^2 g_e}{2S} \sum_{k,l} P_{kl}^{\alpha-\beta} \langle \phi_k | \hat{T} | \phi_l \rangle, \quad (3)$$

Diamagnetic Spin-Orbit

$$g_{\mu\nu}^{\mathrm{DSO}} = \frac{\alpha^2 g_e}{4S} \sum_{k,l} P_{kl}^{\alpha-\beta} \langle \phi_k | \xi(r_A) [\mathbf{r}_A r_O - r_{A,\mu} r_{O,\nu}] | \phi_l \rangle \quad (4)$$

Orbital Zeeman: usually the main source of deviation from free electron g-value.

$$g_{\mu\nu}^{\mathrm{PSO}} = -\frac{g_e}{2S} \sum_{k,l} \frac{\partial P_{kl}^{\alpha-\beta}}{\partial B_\nu} \langle \phi_k | \hat{h}_\nu^{SOC} | \phi_l \rangle \quad (5)$$

(Source: ORCA manual, Release 6.1.0)

Hyperfine Coupling (A) Tensor:

It characterizes the interaction between electron and nuclear spin that leads to splitting of the signal.

$$\mathbf{A}_N = A_{\text{iso}}\mathbf{1} + \mathbf{A}^{\text{dip}} + \mathbf{A}^{\text{orb}} + \mathbf{A}^{\text{GC}} \tag{6}$$

Fermi Contact/ Isotropic Part

$$A_{\text{iso}}(N) = \frac{4\pi}{3}\langle S_z\rangle^{-1} g_e g_N \beta_e \beta_N\, \rho(\mathbf{R}_N), \tag{7}$$

Spin Dipole: Through-space dipole-dipole interaction of nucleus with electron magnetic moment

$$A^{\text{dip}}_{\mu\nu}(N) = P_N \sum_{k,l} \rho_{kl} \left\langle \phi_k \middle| r_N^{-5}(3r_{N\mu}r_{N\nu}\right.$$

$$- \delta_{\mu\nu} r_N^2)\phi_l \tag{8}$$

Spin-orbit contribution: comes from cross terms between spin orbit and nucleus orbit coupling operators

$$A^{\text{orb}}_{\mu\nu}(N) = -\frac{1}{2S} P_N \sum_{k,l} \frac{\partial \rho_{kl}}{\partial I_\nu} \left\langle \phi_k \middle| \hat{h}^{\text{SOC}}_{\mu} \middle| \phi_l \right\rangle \tag{9}$$

$$\hat{h}^{(N)}_{\text{NOC}} = \sum_i \mathbf{r}_{iA}^{-3} \mathbf{l}^{(N)}_{i,\nu} \tag{10}$$

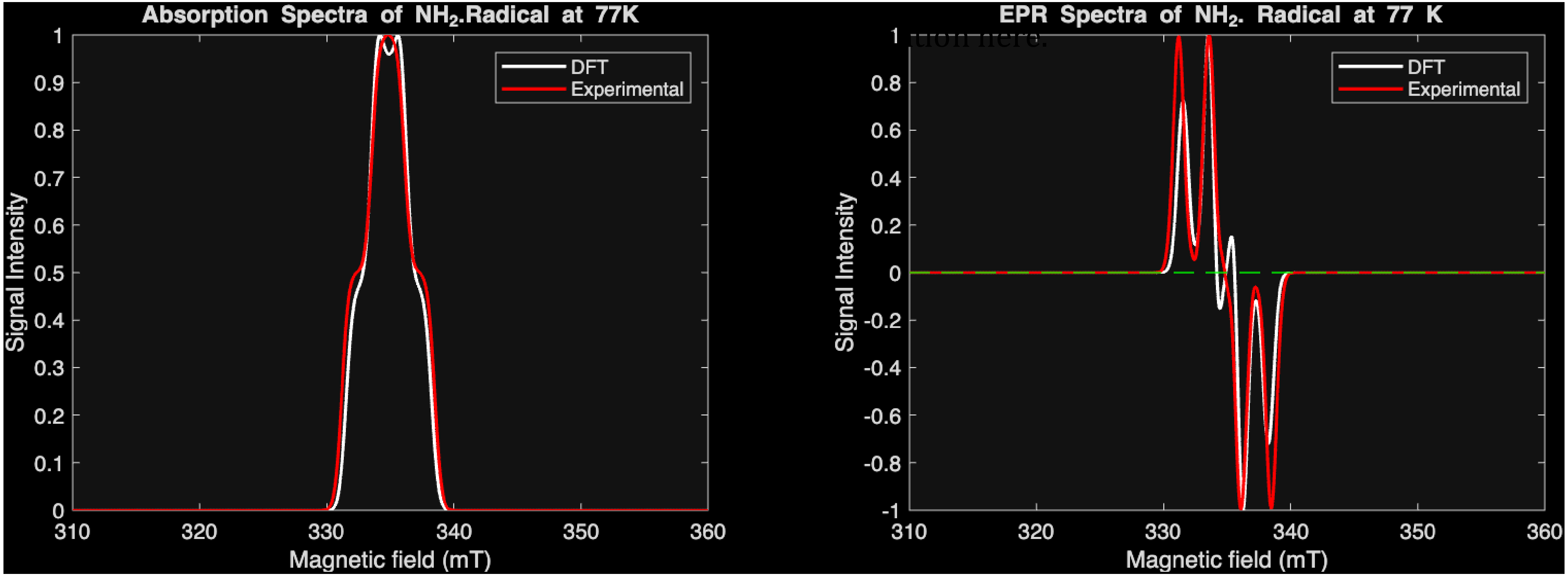
(Source: ORCA manual, Release 6.1.0)

ESR Lines

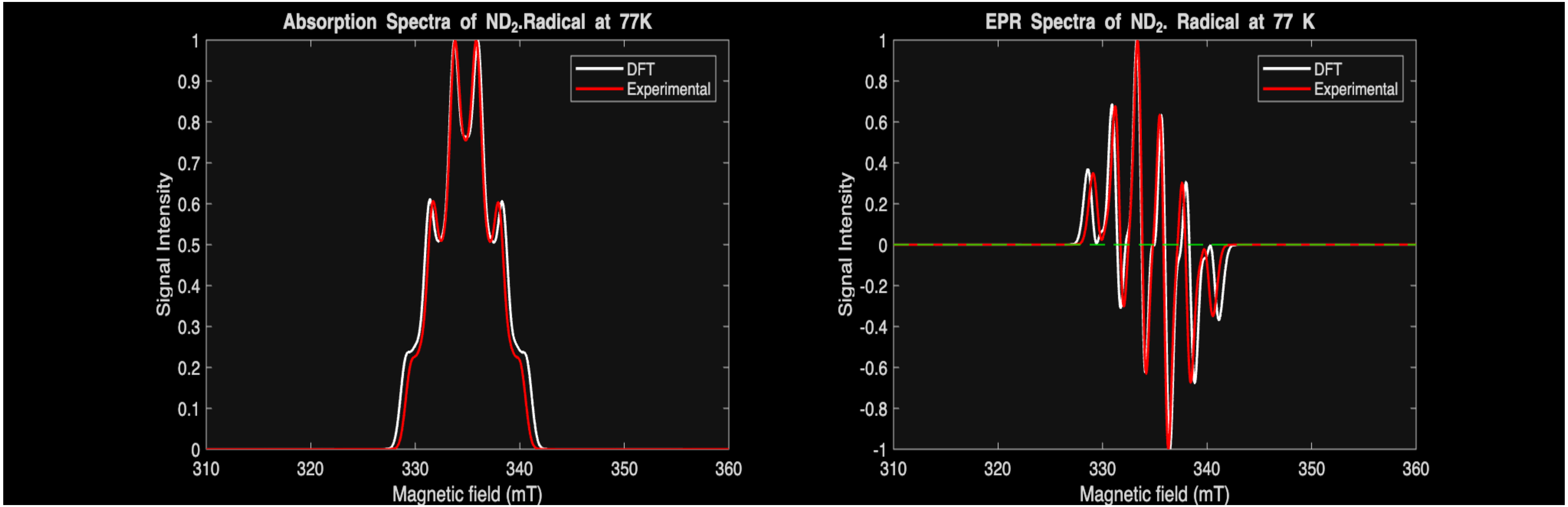
- Calculated **A** and **g**-tensors for both **$\dot{N}H_2$** and **$\dot{N}D_2$**
- $\hat{H}_{ESR}=\mu_B \mathbf{B_0} \cdot g_e \cdot \hat{\mathbf{S}} + \sum \hat{\mathbf{S}} \cdot A \cdot \hat{\mathbf{I}} - g_n \mu_N \sum \mathbf{B_0} \cdot \hat{\mathbf{I}}$
- Constructed ESR line with EasySpin.

Experimental g & A values
[1] Köksal *et al.*, 1985.
[2] Peyerimhoff *et al.*, 1990.
[3] DeMarco *et al.*, 1998.

$\dot{N}H_2$ Radical



$\dot{N}D_2$ Radical

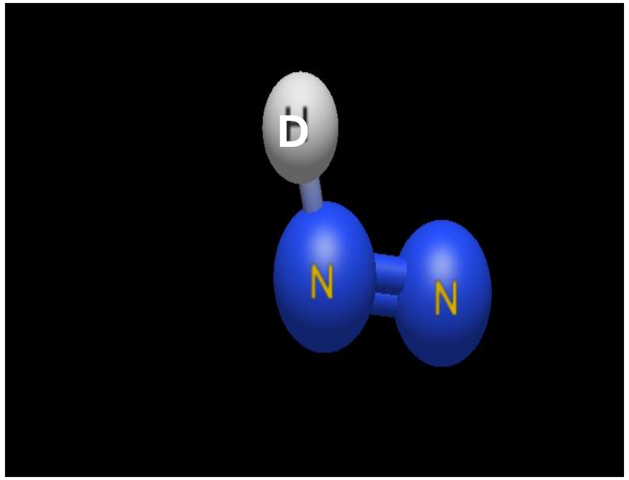


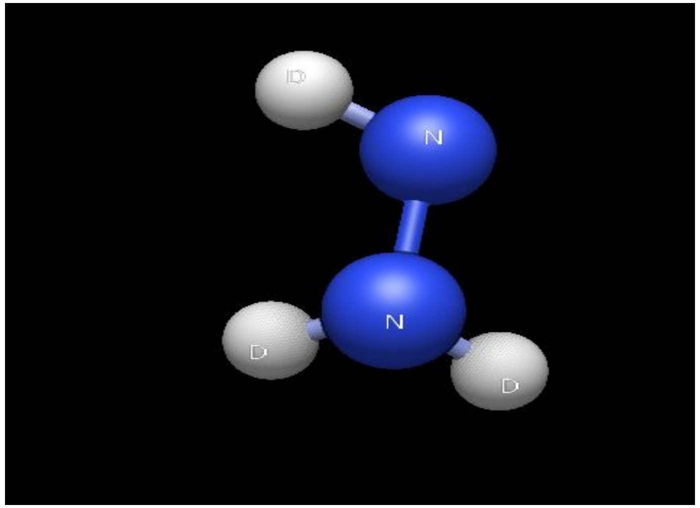
- ESR frequency ≅ 9.5GHz at 334 mT field. Linewidth for $\dot{N}H_2$ is **4.2 mT** and for $\dot{N}D_2$ is **3.8 mT** as measured by D.G. Crabb *et al. J. Chem. Phys. 108(1998) 1423.*

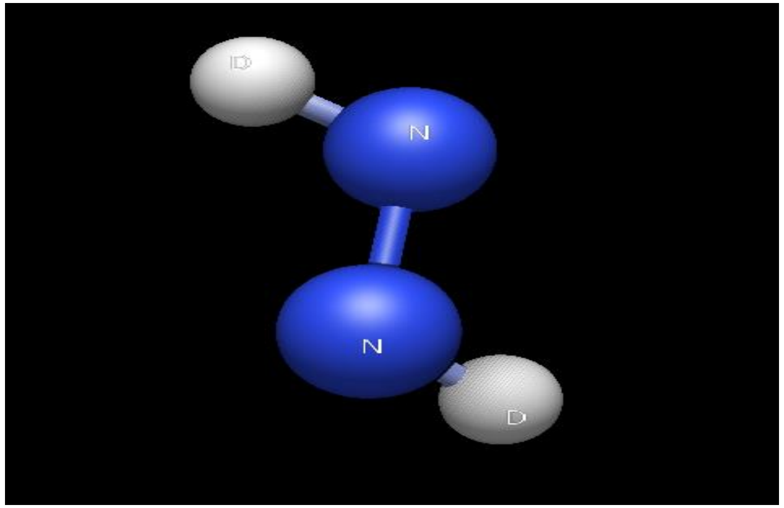
- Spin-Spin/ Dephasing relaxation time T_{2e} for this linewidth would be in nanoseconds.

Possible paramagnetic complex (cold irradiation)

- NH_3 polarization degrades under beamtime due to radiation damage but in ND_3, it improves.







Radical: **Ṅ=ND**

g_{iso}= 2.0002716, Δg= -0.0020477

A_{iso}(MHz): D= 75.3, N1= 76.29, N2= 15.62

Radical: **ṄD-ND_2** (Hydrazines)

g_{iso}= 2.0037656, Δg= 0.0014463

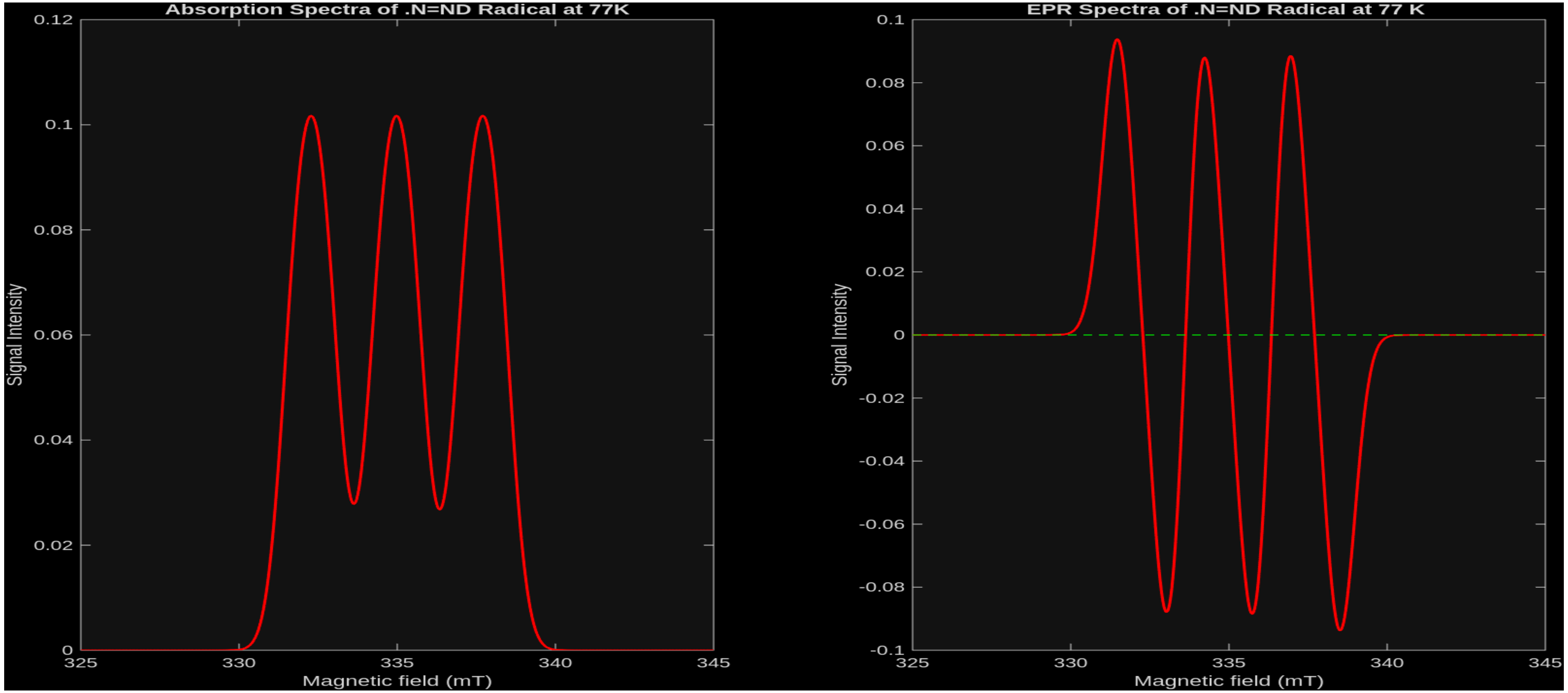
A_{iso}(MHz): D1=-6.8905,D2=-1.4110,D3= 0.6610 ,N1= 28.7719, N2= 24.8832

Radical: **ṄD-ṄD**

g_{iso}= 2.0028297, Δg= 0.0005104

A_{iso}(MHz):D1=18.1879, D2=18.1901,N1=49.9620, N2= 49.9592

ESR Line widened

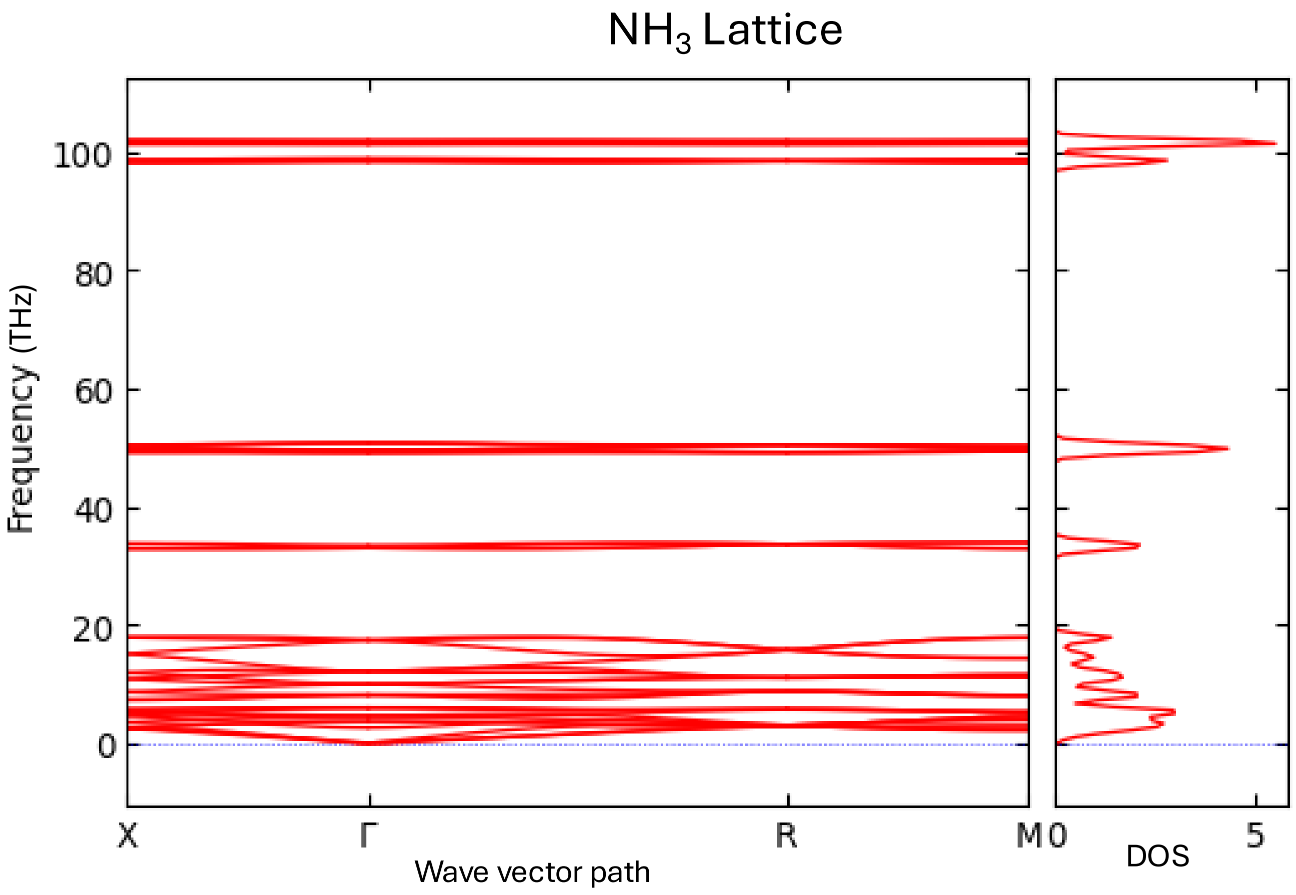


- Line flattened due to structural , dipolar , hyperfine complexity.
- While narrow line is better for microwave frequency tuning during DNP, wider line doesn’t rule out these radicals being formed.
- Need to extend it to spectral density at microwave frequency, it’s overlap with deuteron larmor frequency, spin-lattice relaxation time etc.

Phonon Bands

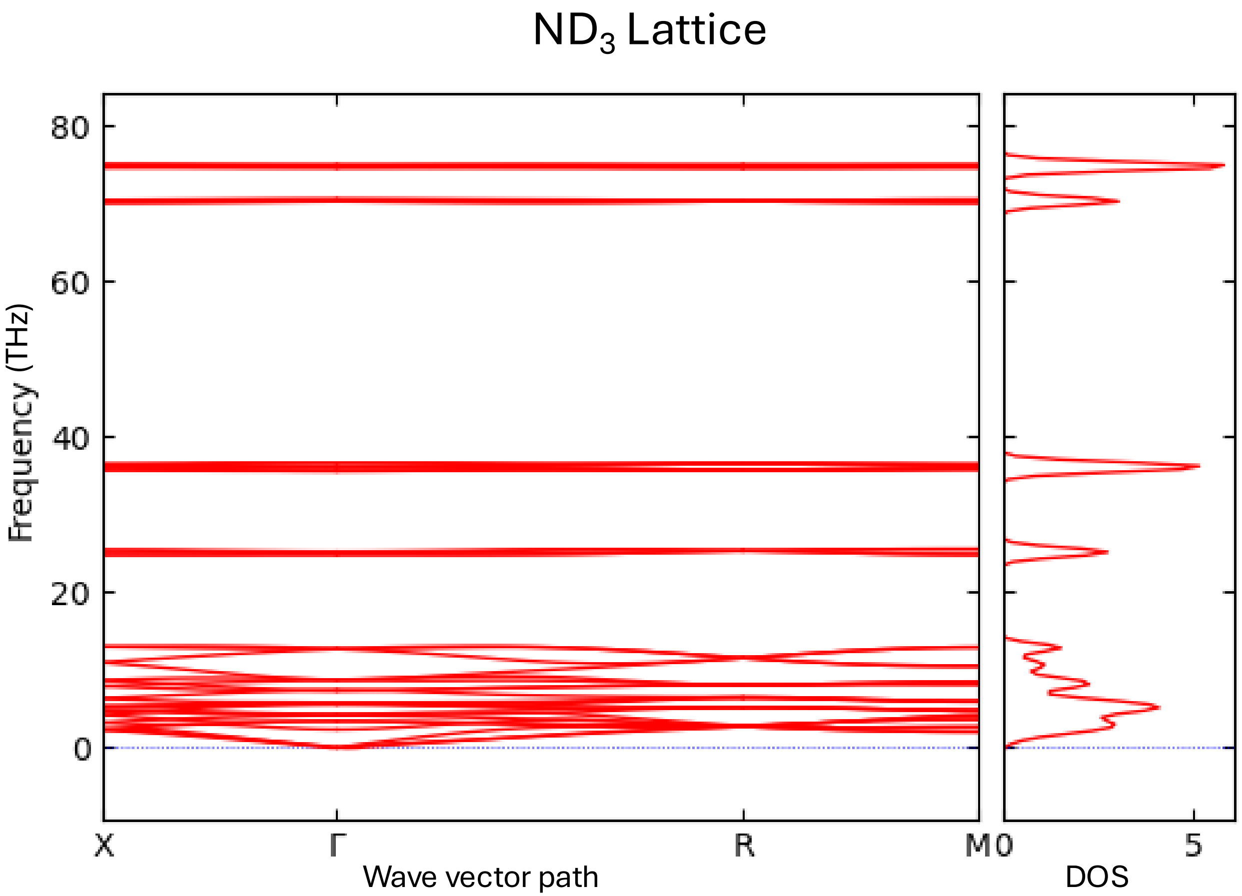
- Gives natural/fundamental vibration of lattice at quantum temperatures.

  - Conversion 100 THz = 0.41 eV.
  - Consistent with band measured by B. Monserrat *et al*. Physical Review B, 2015 .



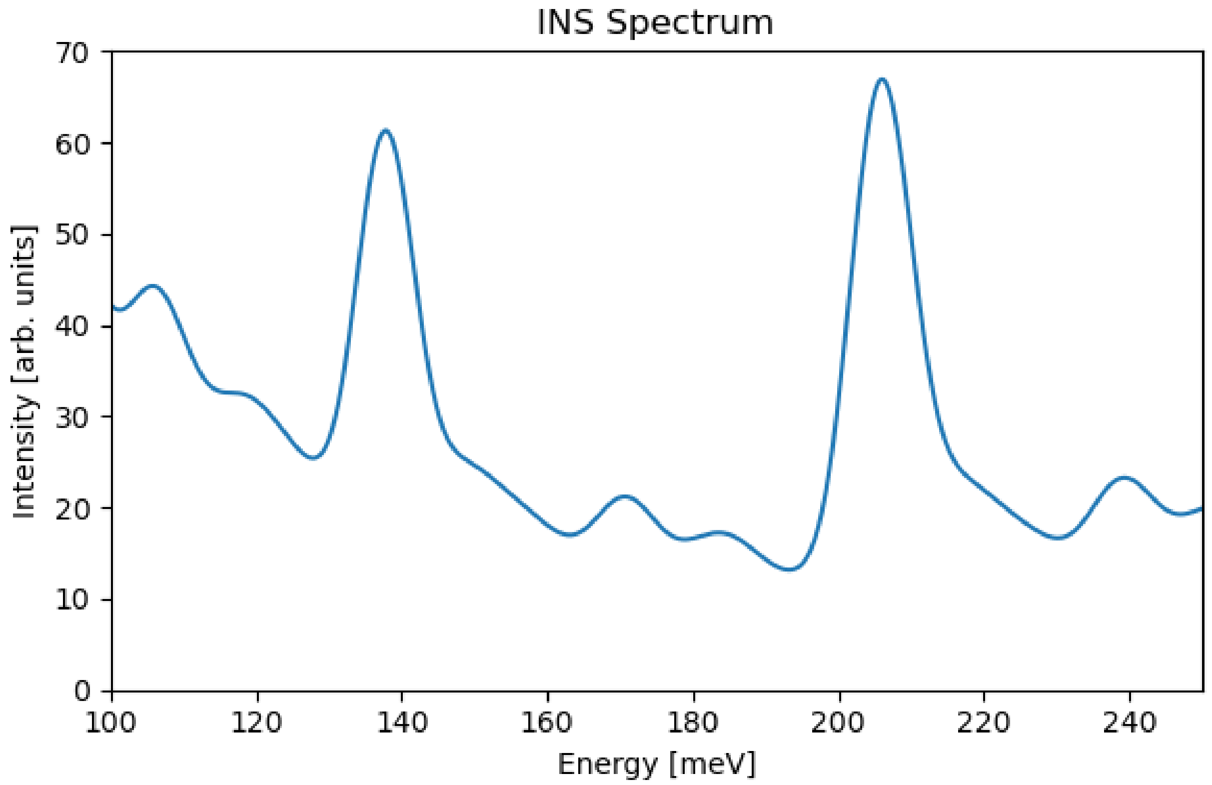
ND_3

- Follows $f_{new} = \frac{f}{\sqrt{M}}$ as predicted by theory.



Inelastic Neutron Scattering

- Gives 2D picture of energy and momentum transfer during scattering



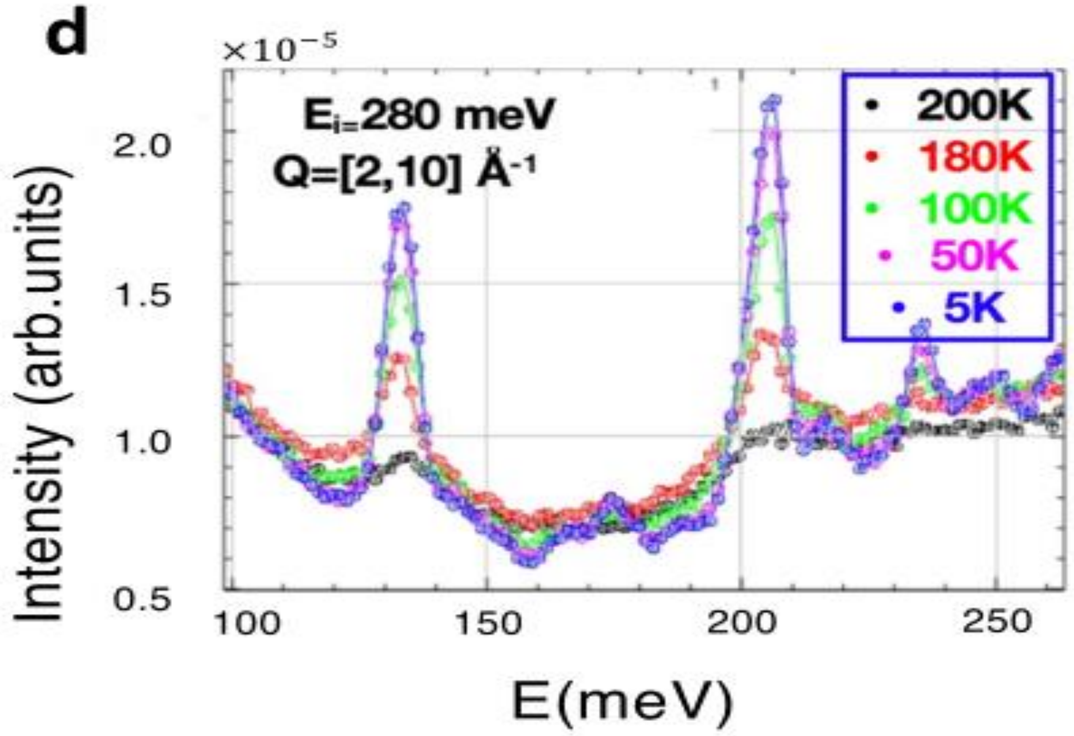
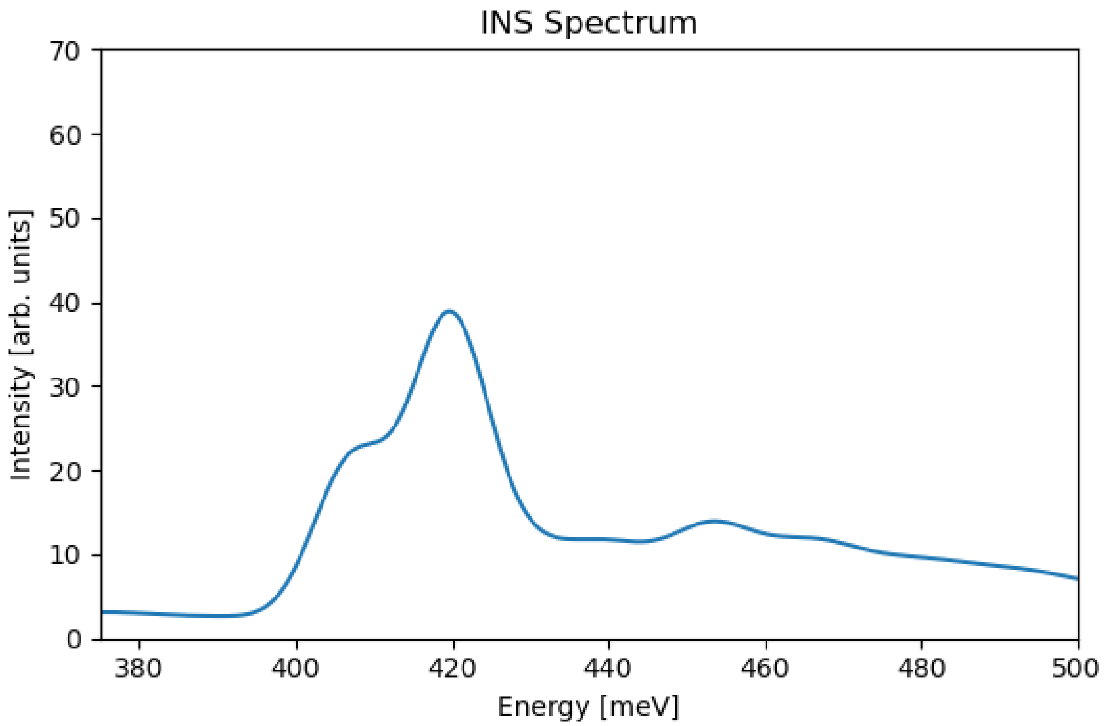


Fig: INS spectrum with DFT phonons and **OCLIMAX** software (left) and experimental measurements by T. M. Linker *et al.* Nature Communications, 2024 (right)

Molecular Bands



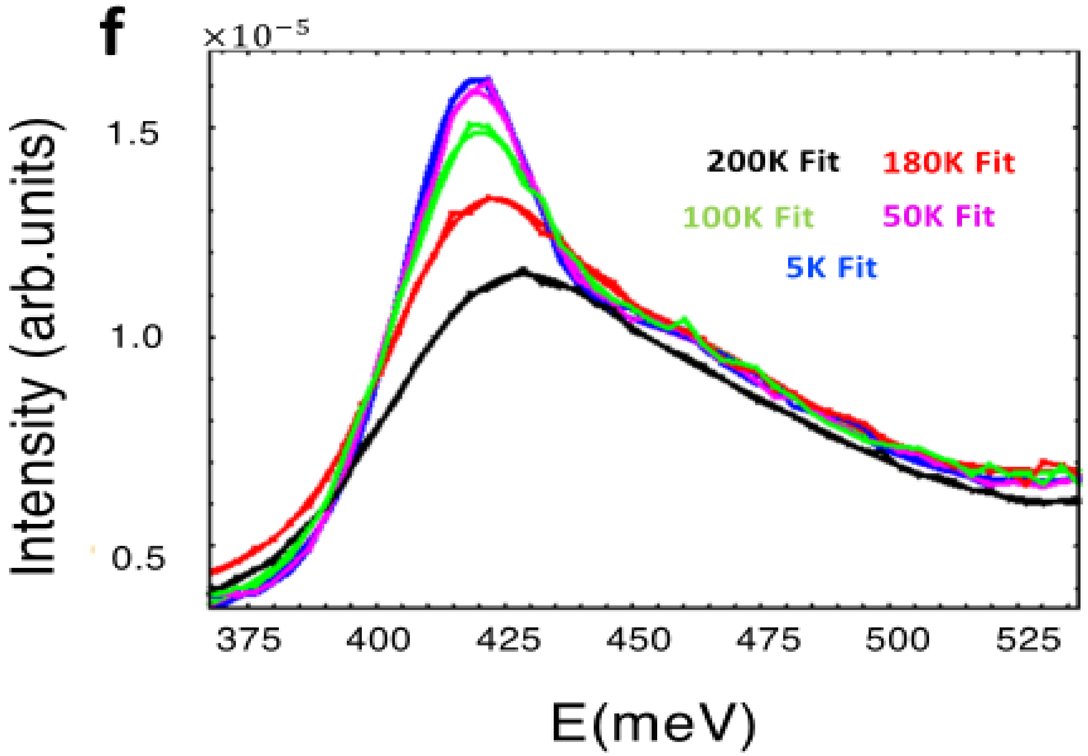
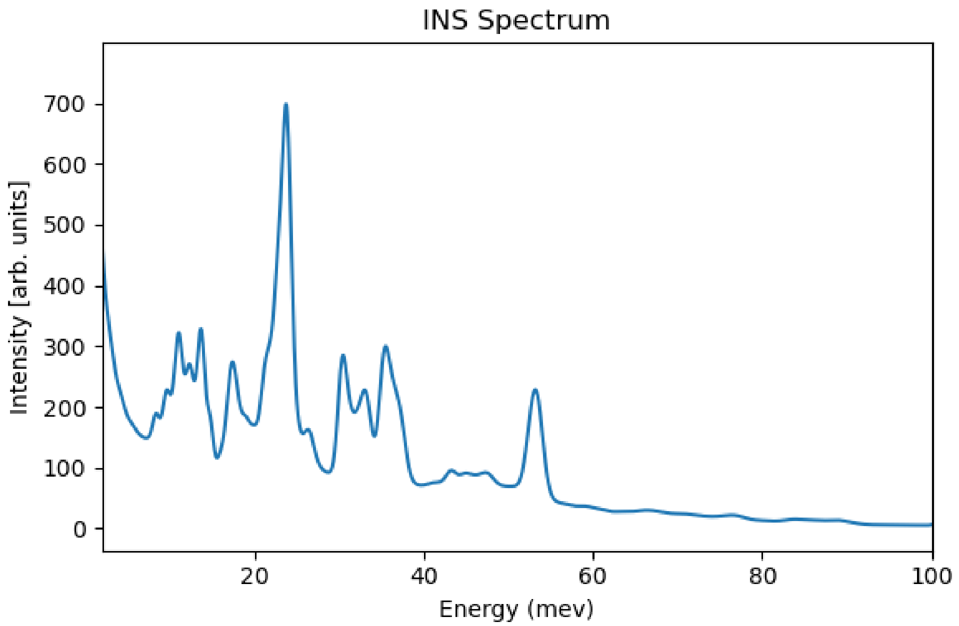


Fig: INS spectrum with DFT phonons and OCLIMAX software (left) and experimental measurement by T. M. Linker *et al.* Nature Communications, 2024 (right)

Lattice Band

- Dependent of the lattice.
- Mainly contributing to spin lattice $\mathbf{T_{1e}}$ and $\mathbf{T_{1n}}$ relaxation.



Next Steps

- Spin-Phonon coupling
- Spin-lattice relaxation time for both NH_3 and ND_3
- Cold dose behavior of ND_3