Thermal neutron scattering law of liquid FLiBe derived from machine learning force field

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Abstract

The thermal scattering law (TSL) data for liquid FLiBe are evaluated in the present work. The self-scattering law component of the total TSL is calculated using a quantum correction method, wherein the self-scattering law is obtained as the Fourier transform of the quantum self-intermediate scattering function, and the width function of the quantum self-intermediate scattering function is related to its classical counterpart through the characteristic function. The Sköld approximation is employed to incorporate the distinct scattering law into the total TSL. To obtain the accurate parameters required for the TSL calculation, a machine learning force field (MLFF) is trained for liquid FLiBe. The results demonstrate the accuracy of the force field and reveal that distinct scattering exerts a significant influence on the accuracy of TSL data for liquid FLiBe.

Full Text

Preamble

Thermal Neutron Scattering Law of Liquid FLiBe Derived from Machine Learning Force Field

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In this work, we evaluate the thermal scattering law (TSL) data of liquid FLiBe. The self-scattering law component of the total TSL is calculated using a quantum correction method, where the self-scattering law is obtained as the Fourier transform of the quantum self-intermediate scattering function. The width function of the quantum self-intermediate scattering function is related to its classical counterpart through a characteristic function. The Sköld approximation is

employed to incorporate the distinct scattering law into the total TSL. To obtain the accurate parameters required for TSL calculations, a machine learning force field (MLFF) is trained for liquid FLiBe. The results demonstrate the high accuracy of the force field and reveal that distinct scattering has a significant influence on the precision of TSL data for liquid FLiBe.

Keywords: Molten salts; Thermal scattering law; Machine learning force field; Molecular dynamics

Introduction

The molten salt FLiBe (66.6% LiF –33.3% BeF_2) has been proposed as an ideal coolant and fuel salt for fluoride-based molten salt reactors (MSRs) due to its excellent properties, including high heat capacity, low neutron absorption cross section, and high boiling temperature [1,2]. With MSRs selected as one of the Generation IV reactor candidates, FLiBe has attracted considerable attention [3,4]. Although FLiBe is primarily used as a coolant, it still possesses a relatively high moderating ratio of 63 [5]. Consequently, thermal neutron scattering in FLiBe significantly affects neutronics simulations.

In the thermal neutron energy region (typically below 5 eV), scattering reactions between thermal neutrons and materials are intimately related to the structural and dynamic characteristics of the material, arising from thermal motion of target nuclei, chemical binding effects, and interference of scattered waves. The thermal scattering law (TSL) for various materials is provided in evaluated nuclear data files (ENDF) [6] to describe energy and momentum transfer during thermal neutron scattering. The thermal scattering cross-section data used in neutron transport calculations are generated by nuclear data processing codes based on TSL data. In recent years, atomic simulation techniques such as lattice dynamics and molecular dynamics simulations have advanced rapidly. Material simulations can provide key parameters required for TSL data evaluation, thereby facilitating assessment of thermal scattering laws for new materials. Several studies worldwide have employed atomic simulation techniques to evaluate TSL data for FLiBe.

The TSL data for solid BeF_2 and LiF were previously evaluated [7,8]. In the work of Mei et al. [7], TSL data for BeF_2 and LiF crystals were calculated using the LEAPR module in the NJOY nuclear data processing code [9]. Wang et al. [8] calculated TSL data using the SIRIUS code. Both LEAPR and SIRIUS calculate TSL for crystalline materials using the phonon expansion model, which requires the phonon density of states as a fundamental parameter. Mei et al. [7] used the density functional theory (DFT) code CASTEP [10] to model BeF_2 and LiF and calculate the phonon density of states, while Wang et al. [8] employed the Vienna Ab initio Simulation Package (VASP) [11] and the PHONON code [12].

In subsequent work, Mei et al. [13] adopted the LEAPR module to evaluate TSL data for liquid FLiBe. In LEAPR, the self-scattering law for liquid materials is

separated into several dynamical modes, with each partial TSL represented by a simple frequency spectrum. The self-scattering law is obtained by convolving all partial TSLs. Mei et al. [13] calculated the self-scattering law of liquid FLiBe using bound vibrational and diffusive modes, where the spectrum for the bound vibrational mode was evaluated using DFT based on solid crystalline FLiBe, and the spectrum for the diffusive mode was represented by the Egelstaff-Schofield diffusion model [14].

Recognizing that atomic vibrations and diffusion in solid crystalline FLiBe differ from those in liquid FLiBe, Zhu and Hawari [15] updated the bound vibrational and diffusion spectra by directly simulating liquid FLiBe with the classical molecular dynamics (CMD) code LAMMPS [16], employing the Born-Mayer-Huggins potential [17,18]. The TSL for liquid FLiBe was then calculated using the LEAPR module with these updated spectra.

In the aforementioned molecular dynamics (MD) simulation [15], the generalized frequency spectrum—including both bound vibrational and diffusion modes—was obtained from the Fourier transform of the velocity autocorrelation function (VACF) calculated from ion trajectories generated by LAMMPS. This spectrum was subsequently separated into diffusion and bound vibrational mode components. However, the frequency spectrum calculated this way serves as an approximate substitute for the rigorous quantum mechanical version because MD simulations are based on classical mechanics. Therefore, Zhu [19] proposed a quantum correction (QC) method to incorporate quantum effects. In this approach, the quantum self-scattering law was formulated through the Fourier transform of the quantum self-intermediate scattering function. The width function of the quantum self-intermediate scattering function was related to the classical one via a characteristic function, with the classical width function calculated using the mean squared displacement (MSD) obtained from CMD simulations.

The QC method described above has been implemented in the nuclear data processing code NECP-Atlas [20] to calculate TSL data for H_2O and D_2O, demonstrating good accuracy for liquid materials [21]. However, the CMD simulations in Zhu's works [15,19] employed the Born-Mayer-Huggins potential, whose coefficients were initially parameterized for solid Li_2BeF_4 crystal [22]. This potential was found to have difficulties predicting properties of liquid FLiBe [23], with a softer potential behavior necessary for better prediction of physical properties at high temperatures [15].

Another classical potential is the Polarizable Ionic Interaction Potential (PIM), which includes polarization effects. A previously developed PIM [24] showed high accuracy compared to experimental data. The PIM was constructed from ab initio calculations [25], with parameters fitted by minimizing differences between ab initio data (energies, forces, etc.) and PIM model predictions for given structures. However, the accuracy and flexibility of PIM are constrained by its functional form.

Ab initio molecular dynamics (AIMD) based on quantum mechanics is considered highly accurate. Wang et al. [26] used AIMD to obtain VACF for calculating the generalized frequency spectrum of liquid FLiBe. However, the computational cost of AIMD makes it difficult to predict complex properties requiring large system sizes and long time scales, including the generalized spectrum and diffusion coefficient (calculated from MSD slope) needed for TSL evaluation. MSD depends on dynamic behavior over long time scales, and the generalized frequency spectrum also requires large-scale, long-time simulations to converge to desired accuracy.

Machine learning force field (MLFF), also called machine learning potential, offers a breakthrough that balances accuracy and efficiency by learning potential functions from high-precision ab initio data through machine learning models [27–33]. As demonstrated by previous deep learning potential MD (DPMD) [33] and moment tensor potential (MTP) MD [27] studies on FLiBe, MLFF can reproduce thermophysical properties within experimentally acceptable accuracy. Recent studies [28,29,31,34] showed that MLFF achieves comparable accuracy to ab initio simulations in predicting forces and energies while enabling long-scale MD simulations for large unit cells. This is critical for capturing long-range dynamics in liquid FLiBe and enables ab initio-level precision for TSL evaluation.

The present work focuses on evaluating TSL data for liquid FLiBe by combining MLFF with the QC-based TSL calculation method. The MLFF method implemented in VASP was used to generate the MLFF for FLiBe. MD simulations based on MLFF (MLFF-MD) were performed to obtain parameters required for TSL calculation, and the QC method implemented in NECP-Atlas was used to calculate the TSL.

This paper is organized as follows: Section II describes the methodologies for TSL evaluation and MLFF generation. Section III presents the physical properties of FLiBe predicted by MLFF and the thermal neutron scattering cross sections obtained from the evaluated TSL. Conclusions are given in Section IV.

II. Methodologies

A. Thermal Scattering Law

By applying the Born approximation and Fermi's potential to solve Schrödinger's equation [35], the double differential scattering cross section can be expressed as:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{\sigma_{coh}}{4\pi}\sqrt{\frac{E'}{E}}S(\alpha,\beta) + \frac{\sigma_{inc}}{4\pi}\sqrt{\frac{E'}{E}}S_s(\alpha,\beta)$$

where E is the incident energy, E' is the outgoing energy, k_B is the Boltzmann constant, T is the temperature, μ is the cosine of the scattering angle in the

laboratory frame, σ_{coh} is the coherent bound atom scattering cross section, σ_{inc} is the incoherent bound atom cross section, and α and β are the dimensionless momentum and energy transfers defined as:

$$\alpha = \frac{(\hbar\kappa)^2}{2Ak_{BT}}$$

$$\beta = \frac{E' - E}{k_{BT}}$$

where A is the mass ratio of scattering target nuclei to neutron, $\hbar$ is the reduced Planck constant, κ is the wave vector of momentum transfer, and ω is defined by the energy transfer $\hbar\omega = E - E'$. In Eq. (1), $S_s(\alpha, \beta)$ is the self-scattering law quantifying contributions from individual atom dynamics, and $S(\alpha, \beta)$ is the coherent scattering law expressed as:

$$S(\alpha, \beta) = S_s(\alpha, \beta) + S_d(\alpha, \beta)$$

where $S_d(\alpha, \beta)$ is the distinct scattering law quantifying interference contributions between different atoms. In Eq. (1), the cross-section term containing $S(\alpha, \beta)$ is known as the coherent scattering cross section, while the term containing $S_s(\alpha, \beta)$ is called the incoherent scattering cross section.

In this work, $S_s(\alpha, \beta)$ is calculated using the QC method. According to Van Hove theory [35], $S_s(\alpha, \beta)$ can be defined as the Fourier transform of the self-intermediate scattering function $I_s(\alpha, t')$:

$$S_s(\alpha, \beta) = \int_{-\infty}^{\infty} I_s(\alpha, t') \exp(-i\beta t') dt'$$

where $\hbar$ is the reduced Planck constant and $t' = k_{BTt}/\hbar$.

Based on the Gaussian approximation, $I_s(\alpha, t')$ can be represented as a Gaussian function [36]:

$$I_s(\alpha, t') = \exp(-\alpha\gamma(t'))$$

where $\gamma(t')$ is the quantum width function expressed as [19,37]:

$$\gamma(t') = \int_{-\infty}^{\infty} \frac{2M}{k_{BT}} f_c(\beta) \left\{ \frac{\exp(\beta/2)}{2\sinh(\beta/2)} [1 - \cos(\beta t')] - i \sin(\beta t') \right\} d\beta$$

where M is the particle mass, and $f_c(\beta)$ is the classical characteristic function that can be evaluated from MSD [19]:

$$f_c(\beta) = \frac{\beta^2}{Mk_{BT}} \int_{-\infty}^{\infty} \exp(i\beta t')\mathrm{MSD}(t')dt'$$

For calculating $S_d(\alpha, \beta)$, the incoherent approximation—where $S_d(\alpha, \beta)$ in Eq. (1) is directly ignored—is conventionally adopted, reducing Eq. (1) to:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{\sigma_{coh} + \sigma_{inc}}{4\pi} \sqrt{\frac{E'}{E}} S_s(\alpha, \beta)$$

However, $\sigma_{coh}/(\sigma_{coh} + \sigma_{inc})$ is large for each nuclide in FLiBe, indicating that $S_d(\alpha, \beta)$ cannot be ignored. The ratios are shown in Table 1 for the bound scattering cross sections.

Therefore, to include distinct scattering, $S(\alpha, \beta)$ in this work is calculated using the Sköld approximation [38]:

$$S(\alpha, \beta) = S_s(\alpha/S_i(\kappa), \beta) \cdot S_i(\kappa)$$

where κ is the wave vector transfer variable, and $S_i(\kappa)$ is the Sköld correction factor for the i-th atom, obtained as [39]:

$$S_i(\kappa) - 1 = \frac{\sum_j c_j b_j^{\mathrm{coh}} [S_{ij}(\kappa) - 1]}{\sum_j c_j b_j^{\mathrm{coh}}}$$

where $S_{ij}(\kappa)$ is calculated as [40]:

$$S_{ij}(\kappa) = 1 + 4\pi\rho \int_0^{\infty} dr[g_{ij}(r) - 1] \frac{r^2 \sin(\kappa r)}{\kappa r}$$

where c_j is the mole fraction of the j-th atom, b_i^{coh} is the coherent bound scattering length of the i-th atom, r is the distance between i-th and j-th atoms, and $g_{ij}(r)$ is the radial distribution function (RDF) between i-th and j-th atoms.

In the above calculation model, MSD and RDF serve as input parameters for calculating $S_s(\alpha, \beta)$ and $S(\alpha, \beta)$, respectively. In this work, these variables are obtained from MD simulations based on MLFF.

B. MLFF Generation

Generating MLFF requires a training dataset from ab initio calculations, containing Bravais matrices, atom positions, energies, forces, and stress tensors. MLFF accuracy depends on training dataset quality, making appropriate dataset size selection essential. However, larger training sets demand greater

computational cost. To address this, we adopted the on-the-fly MLFF algorithm [29,30,41] implemented in VASP, which uses active learning during MD simulations. In this algorithm, MD simulations are performed during training, and force field accuracy is estimated via Bayesian error estimation (BEE) of forces at each MD step. If the estimated Bayesian force error for any atom exceeds a set threshold, DFT calculations are performed; otherwise, MLFF is used for that MD step. This algorithm selectively incorporates structures poorly predicted by MLFF into the training set, avoiding excessive DFT calculations and enhancing computational efficiency.

Since this work aims to evaluate TSL for FLiBe within the temperature range of 800–1600 K, MLFF training was performed within this range. MLFF can be trained in a smaller unit cell and then applied to larger systems. Therefore, MD simulations for training were conducted with a 98-atom supercell initially generated by Packmol [42] based on the experimental density of 2.01 g/cm^3 at 823 K. As the structure was randomly generated by Packmol, we first performed an MD simulation using the Langevin thermostat with a 1 fs time step under constant-pressure and constant-temperature (NPT) ensemble at 800 K for 10 ps to accelerate structure equilibration.

Subsequently, the MLFF for FLiBe was trained using the on-the-fly algorithm during three MD simulations. In these simulations, the Langevin thermostat with a 1 fs time step was used; cutoff radii for radial and angular descriptors in MLFF training were set to 8 Å and 5 Å, respectively. The three MD simulations for training were: (i) an MD simulation from 800 K to 1600 K for 40 ps; (ii) an NPT MD simulation at 1600 K for 10 ps; and (iii) a cooling simulation from 1600 K to 800 K for 20 ps, using the equilibrium structure from the 1600 K NPT simulation as the initial configuration.

After training, we obtained an MLFF based on a training set containing 2388 structures. For each atom type, the number of local reference configurations is 4500, selected from atoms in the training structures.

During training, DFT calculations were performed to generate the training dataset. Computational settings were as follows: the Generalized Gradient Approximation (GGA) with Perdew-Burke-Ernzerhof (PBE) functional was used for exchange-correlation; a plane-wave cutoff energy of 600 eV was set; Gaussian smearing with 0.05 eV width determined partial occupancies; electron self-consistency convergence was set to 10^{-6} eV; and a single gamma point was used for Brillouin zone sampling. Projector-augmented-wave (PAW) potentials were used in all calculations. Proper description of van der Waals (vdW) interactions is important for predicting FLiBe density [27,33], so the DFT-D3 method with Becke-Johnson damping was used for dispersion correction.

In the training process, BEE of forces served as a monitoring quantity. Bayesian error estimates the average training set error for new random configurations, reflecting the real error between predicted and DFT values for current configurations. Significant jumps in the BEE curve indicate that the current con-

figuration differs substantially from previously learned structures, suggesting MLFF-predicted forces will deviate from reference AIMD results. Since force data in the training dataset far exceeds energy data [34], force was chosen as the criterion for performing DFT calculations. Fig. 1 [Figure 1: see original paper] shows BEE, root mean square errors (RMSE), and force threshold during training. In the final cooling simulation, the maximum BEE was 0.042 eV/Å; BEE remained below threshold at nearly every MD step, and force RMSE stabilized around 0.043 eV/Å with minimal force field updating. Therefore, the force field meets requirements for property prediction.

To assess predictive accuracy, we built a test set of 5000 structures from ab initio configurations randomly selected from AIMD simulations (800–1600 K), independent of the training set. Energy and force predictions for training and test sets are shown in Fig. 2 [Figure 2: see original paper], with RMSE values given in Table 2 . Results demonstrate good agreement between MLFF-MD and AIMD calculations. The excellent accuracy of MLFF-predicted energies and forces provides a solid basis for large-system, long-time-scale MLFF-MD simulations for FLiBe property prediction from 800–1600 K.

C. Calculation Flowchart of TSL Data for Liquid FLiBe

This section summarizes the TSL evaluation framework for liquid FLiBe, shown in Fig. 3 [Figure 3: see original paper]. The workflow is: (i) train MLFF for liquid FLiBe using the on-the-fly algorithm; (ii) test MLFF-predicted forces and energies on a DFT-based test set; (iii) validate trained MLFF against theoretical results and experimental data for liquid FLiBe properties (density, self-diffusion coefficient, RDF, VACF), including our AIMD results and other available theoretical data; (iv) calculate TSL for liquid FLiBe using the QC method based on MSD and RDF from MLFF-MD.

III. Results and Discussion

A. Properties of Liquid FLiBe

This section presents predictions of liquid FLiBe properties—including density, RDF, VACF, diffusion coefficient, and electrical conductivity—to demonstrate trained MLFF accuracy. MLFF-MD simulations were performed with a larger 7000-atom FLiBe system. Before production MD simulations for property calculations, we first performed 10 ps NPT simulations at several temperatures to ensure initial systems reached equilibrium. Production MD simulations were then conducted at NPT ensemble using MLFF at each temperature to obtain properties. All simulations used a time step of 1 fs and pressure of 0 bar.

1. Density Fig. 4 [Figure 4: see original paper] shows density versus temperature from MLFF-MD, compared with other theoretical [27,33,43] and experimental results [44–46]. The MLFF-MD density-temperature curve closely matches MTP-MD [27] and AIMD [43] results. DPMD results underestimate

experimental values, primarily due to absence of dispersion corrections in their training data, which has been shown to overestimate FLiBe volume [43]. From our 800–1600 K results, we fitted the following density-temperature equation:

$$\rho(T) = 2.3808 - 4.1 \times 10^{-4} T$$

where ρ is density in g/cm^3 and T is temperature in Kelvin.

2. Radial Distribution Function The RDF is calculated as:

$$g_{ij}(r) = \frac{n_{ij}(r)/V_{\text{shell}}(r)}{N_i(r)/V(r)}$$

where $n_{ij}(r)$ is the number of j-type atoms in a spherical shell of thickness dr at distance r from an i-type atom, $V_{\text{shell}}(r)$ is the shell volume, $N_i(r)$ is the number of i-type atoms in a sphere of radius r, and $V(r)$ is the sphere volume.

Fig. 5 [Figure 5: see original paper] compares RDFs for F^--F^-, Li^+-F^-, and Be^{2+}-F^- in FLiBe at 973 K from MLFF-MD and AIMD. MLFF-MD RDF curves reproduce AIMD results well. The first RDF peak for Be-F is sharp and high, with the post-peak minimum approaching zero, while the Li-F first peak is broader with a relatively high minimum. This indicates distinct local structures for Li^+ and Be^{2+}, with stronger Be^{2+}-F^- bonding.

From RDFs, average near-neighbor distances and first-shell coordination numbers can be calculated. Table 3 lists average near-neighbor distances. MLFF-MD predicts 1.545 Å for Be^{2+}-F^-, 2.589 Å for F^--F^-, and 1.853 Å for Li^+-F^-, showing good agreement with our AIMD results, experimental data [47], and other theoretical results including AIMD [43], MTP-MD [27], DPMD [33], and PIM-MD [24]. First-shell coordination numbers from various methods are shown in Table 4 . Predicted results are consistent with our AIMD, AIMD [43], MTP-MD [27], and PIM-MD [24]. However, theoretical results differ from experimental data, particularly for F^--F^- coordination numbers. This discrepancy was attributed to difficulty in determining the minimum after the first RDF peak over a wide flat region [27].

3. Velocity Autocorrelation Function Using atomic velocities from MLFF-MD and AIMD, VACF was calculated as:

$$\text{VACF}(t) = \frac{1}{N} \sum_{i=1}^{N} \langle v_i(0) \cdot v_i(t) \rangle$$

where $v_i(0)$ and $v_i(t)$ are velocities of the i-th atom at times 0 and t, respectively. As shown in Fig. 6 [Figure 6: see original paper], MLFF-MD VACF matches AIMD well. VACF curves initially decay, then oscillate before converging to

zero. Be^{2+} ions show the fastest VACF decay, followed by Li^+, with F^- showing the slowest decay. Be^{2+} exhibits longer oscillation times up to ~150 fs compared to F^- and Li^+, attributed to formation of fluoro-beryllate anion $BeF_4{}^{2-}$ [15].

4. Diffusion Coefficient The diffusion coefficient is obtained from the MSD slope:

$$\text{MSD} = \langle \sum_{i=1}^{N} [r_i(t) - r_i(0)]^2 \rangle$$

where $r_i(t)$ and $r_i(0)$ are positions of the i-th atom at times t and 0, and N is the number of atoms.

MSD for Be, F, and Li in FLiBe from 800–1600 K was calculated from MLFF-MD simulations. Fig. 7 [Figure 7: see original paper] shows MSD for 10 ps with 1 fs timestep. With increasing time, ions continue moving and MSD increases, showing linear MSD-time relationship in liquid FLiBe.

In ENDF libraries, inelastic scattering TSL is stored as a two-dimensional interpolation table of α and β, requiring input α and β values. In this work, MSD is the main input parameter for self-scattering law calculation. For TSL in specific α ranges, MSD with specific timesteps is required, meaning MSD timestep for TSL calculation should be determined by α magnitude. Larger timestep and longer total time MSD were used for lower α, while smaller timestep and shorter total time MSD were used for higher α. Table 5 shows different timesteps and total times for various α ranges.

Fig. 8 [Figure 8: see original paper] compares diffusion coefficients of Be, F, and Li ions from MLFF-MD with machine-learning potential MD (MTP-MD [27], DPMD [33]), PIM-MD CMD [24], and experimental results [48,49]. Our results are close to PIM potential predictions. Theoretical diffusion coefficients for Li and Be are all lower than experimental data, which previous works [19,24,43] suggested may contain errors due to relatively large values and high activation energies.

5. Electrical Conductivity Based on diffusion coefficients, electrical conductivity is given by the Nernst-Einstein equation [50]:

$$\sigma = \frac{e^2}{V k_{BT}} \sum_i N_i z_i^2 D_i$$

where e is elementary charge, V and T are system volume and temperature, k_B is Boltzmann constant, and N_i, z_i, and D_i are number, charge, and diffusion coefficient of the i-th ion.

Fig. 9 [Figure 9: see original paper] shows electrical conductivity calculated from Section III.A.4 diffusion coefficients, compared with experimental data [24]. MLFF-MD-derived conductivities are close to experimental values.

B. Thermal Neutron Scattering Cross Sections of FLiBe

Based on MSD and RDF from MLFF-MD simulations, TSL and thermal neutron scattering cross sections for liquid FLiBe were calculated using the QC method in NECP-Atlas. To better compare force field effects on TSL, FLiBe cross sections were also calculated using the QC method based on CMD simulation with the same Born-Mayer-Huggins potential as previous works [15,19,51].

The newly released ENDF/B-VIII.1 library [52] includes TSL data for liquid FLiBe evaluated by Zhu et al. [51], where TSL was separated into two partial TSLs for bound vibrational and diffusive modes under the incoherent approximation. We compare our results with ENDF/B-VIII.1 evaluation.

1. Double Differential Scattering Cross Section Figs. 10–12 [FIGURE:10–12] show double differential scattering cross sections (DDSCS) for Be, F, and Li at incident energy $E = 0.075$ eV, calculated under incoherent approximation and compared with ENDF/B-VIII.1. MLFF-MD-based DDSCS peak values are relatively higher, while CMD-based DDSCS are closer to ENDF/B-VIII.1. This primarily reflects differences in MSD calculated using different potentials. As shown in Table 6 , MLFF-MD diffusion coefficients are much lower than CMD predictions and values from Zhu' s work [19], which used the same potential as the ENDF/B-VIII.1 FLiBe evaluation [51].

Because MLFF-MD predicts lower diffusion coefficients than CMD, translational diffusion in FLiBe is more suppressed, resulting in a narrower quasi-elastic scattering peak. Consequently, scattering intensity concentrates over a narrower energy transfer range, producing higher DDSCS peaks.

2. Integrated Scattering Cross Section Fig. 13 [Figure 13: see original paper] shows integrated thermal scattering cross sections for Be, F, Li, and FLiBe at 873 K, calculated using incoherent approximation. MLFF-MD-based cross sections for Be and Li are slightly lower than ENDF/B-VIII.1 in the 10^{-5}–10^{-2} eV range, while the F cross section is significantly lower. Deviations arise from differences in force fields and TSL calculation methods. Considering each nuclide' s contribution, the total FLiBe scattering cross section is lower than ENDF/B-VIII.1 in the cold neutron region below 5$×10^{-3}$ eV. Obvious differences appear when comparing MLFF-MD with CMD results due to force field variations. CMD results with Born-Mayer-Huggins potential differ from ENDF/B-VIII.1 for individual nuclides but are close for total cross section, suggesting important methodological influences.

Fig. 14 [Figure 14: see original paper] shows thermal scattering cross sections for Be, F, Li, and total FLiBe at six temperatures from 800–1600 K, calculated

using incoherent approximation. Cross sections increase with temperature and converge to free-atom values above 1 eV, consistent with theory.

3. Effect of Distinct Scattering on Thermal Scattering Law The above cross sections were calculated with incoherent approximation, ignoring $S_d(\alpha, \beta)$ in Eq. (4). Distinct scattering in liquid materials can be included via Sköld approximation. Sköld correction factors $S_i(\kappa)$ for F, Li, and Be at 873 K were calculated using Eqs. (11) and (12) and shown in Fig. 15 [Figure 15: see original paper]. Be and Li correction factor curves appear slightly negative at some κ points, violating the physical condition $S_i(\kappa) \geq 0$. To avoid non-physical negative TSL values, these negative points were set to zero.

Fig. 16 [Figure 16: see original paper] compares thermal scattering cross sections for Be, F, and Li at 873 K calculated using Sköld approximation versus incoherent approximation. Sköld approximation results show oscillations due to distinct scattering. Be and F show more obvious deviations than Li compared to incoherent approximation results, explained by σ_{coh} dominating the $\sigma_{coh} + \sigma_{inc}$ sum for Be and F (Table 1).

Due to lack of experimental data, cross-section verification remains challenging. FLiBe cross-section measurements are needed to confirm theoretical validity and guide evaluation method improvements.

IV. Conclusion

In this work, an MLFF for liquid FLiBe was trained using the on-the-fly machine learning algorithm in VASP to evaluate thermal neutron scattering law. Liquid FLiBe properties—including density, RDF, VACF, diffusion coefficient, and electrical conductivity—were predicted using the trained MLFF and compared with our AIMD calculations, other theoretical results, and experiments. MLFF-MD predictions generally show good agreement with experiments and AIMD calculations.

Thermal scattering cross sections for FLiBe from 800–1600 K were calculated based on MLFF-MD and compared with CMD-based results and the latest ENDF/B-VIII.1 library. Both MLFF-MD and CMD results used the same QC method, so their cross-section discrepancies demonstrate significant force field influence on evaluated TSL. Comparative analysis of Sköld versus incoherent approximation reveals that thermal scattering cross sections accounting for distinct scattering differ substantially from those ignoring it. Due to TSL calculation method differences and force field variations, incoherent approximation-based MLFF-MD cross sections deviate from ENDF/B-VIII.1.

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