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

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Abstract

The thermal scattering law (TSL) data of liquid FLiBe is evaluated in the present work. The self-scattering law component of the total TSL is calculated using a quantum correction method, where the self-scattering law is calculated as the Fourier transform of quantum self-intermediate scattering function, and the width function of the quantum self-intermediate scattering function is associated with the classical one by the characteristic function. The Sköld approximation is used to include the distinct scattering law into the total TSL. To obtain the accurate parameters requires for the TSL calculation, a machine learning force field (MLFF) is trained for liquid FLiBe. The results highlight the accuracy of force field, and shows that distinct scattering has obvious influence on the accuracy of TSL data of liquid FLiBe.

Full Text

Preamble

The thermal scattering law (TSL) data of liquid FLiBe is evaluated in the present work. The self-scattering law component of the total TSL is calculated using a quantum correction method, where the self-scattering law is calculated as the Fourier transform of the quantum self-intermediate scattering function, and the width function of the quantum self-intermediate scattering function is associated with the classical one by the characteristic function. The Sköld approximation is used to include the distinct scattering law in 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 highlight the accuracy of the force field and show that distinct scattering has an obvious influence on the accuracy of the TSL data of liquid FLiBe.

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

Introduction

The molten salt liquid FLiBe (66.6% LiF -33.3% BeF_2) has been proposed as an ideal coolant and fuel salt in fluoride-based molten salt reactors (MSR) [?, ?] for its excellent features, such as high heat capacity, low neutron absorption cross section, and high boiling temperature [?, ?]. With the selection of MSR as one of the candidate generation IV reactors, FLiBe has garnered significant attention [?]. Although FLiBe is used primarily as a coolant, it still has a relatively high moderating ratio of 63 [?]. Therefore, thermal neutron scattering of FLiBe has significant effect on the neutronics simulation of the reactor.

In the thermal neutron energy region (normally less than 5 eV), the scattering reactions between thermal neutrons and materials are closely related to the material structure and dynamic characteristics due to the thermal motion of the target nuclei, the binding effect of chemical bonds, and the interference effect of scattered waves. The so-called thermal scattering law (TSL) for different materials is provided in the evaluated nuclear data files (ENDF) [?] to describe the energy and momentum transfer when thermal neutrons scatter with various materials. 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 simulation and molecular dynamics simulation of materials have developed rapidly. Simulations of materials can provide the relevant key parameters required for the evaluation of TSL data, thereby promoting the evaluation of thermal scattering laws for some new materials. Several works based on atomic simulation techniques have been performed worldwide to evaluate the TSL data of FLiBe.

The TSL data of solid BeF_2 and LiF were evaluated previously [?, ?]. In the work of Mei et al. [?], the TSL data of BeF_2 and LiF crystals were calculated using the LEAPR module in nuclear data processing code NJOY [?]. Wang et al. [?] calculated the TSL data using the SIRIUS code. In both LEAPR and SIRIUS, the TSL data for crystalline materials is calculated with the phonon expansion model, and the phonon density of states is the basic parameter needed for the TSL calculation. The density functional theory (DFT) based code CASTEP [?] was used to model BeF_2 and LiF to calculate the phonon density of states in the work of Mei et al. [?], and the Vienna Ab initio Simulation Package (VASP) [?] and the PHONON code [?] were used in the work of Wang et al. [?].

In the later work of Mei et al. [?], the LEAPR module was adopted to evaluate the TSL data of liquid FLiBe. In LEAPR, the self-scattering law for liquid materials is separated into several dynamical modes, and for each mode, the corresponding partial TSL is represented by a simple frequency spectrum. The self-scattering law is finally obtained by convolution of all the partial TSLs. Mei et al. [?] calculated the self-scattering law of liquid FLiBe represented by bound vibrational mode and diffusive mode, and the spectrum for bound vibrational mode was evaluated using DFT based on solid state crystalline FLiBe; the spectrum for diffusive mode was represented with the Egelstaff-Schofield diffusion model [?].

Due to the fact that the bound vibration and diffusion of atoms in solid state crystalline FLiBe are different from those in liquid FLiBe, Zhu and Hawari [?] updated the bound vibrational and diffusion spectra by directly simulating the liquid FLiBe with the classical molecular dynamics (CMD) code LAMMPS [?]. The Born-Mayer-Huggins potential [?, ?] was used in the LAMMPS simulations. The TSL for liquid FLiBe was also calculated using the LEAPR module with the updated spectra.

In the above mentioned molecular dynamics (MD) simulation [?], the generalized frequency spectrum including the bound vibrational and diffusion mode was obtained from the Fourier transform of the velocity autocorrelation function (VACF) which was calculated from the ion trajectories generated by LAMMPS. The generalized frequency spectrum was then separated into the spectra for diffusion mode and bound vibrational mode. The frequency spectrum calculated this way is an approximate substitute for the one in rigorous quantum mechanics, because the MD simulation is based on classical mechanics. Therefore, Zhu [?] proposed a quantum correction (QC) method to incorporate the quantum effects. In the proposed method, the quantum self-scattering law was formulated by the Fourier transform of the quantum self-intermediate scattering function. The width function of the quantum self-intermediate scattering function was associated with the classical one by the characteristic function. The classical width function was calculated using the mean squared displacement (MSD) obtained from the CMD simulation.

The QC method described above has been applied in nuclear data processing code NECP-Atlas [?] to calculate the TSL data of H_2O and D_2O, showing good accuracy for liquid materials [?]. However, the CMD simulation was performed using the Born-Mayer-Huggins potential in Zhu' s works [?, ?]. The Born-Mayer-Huggins potential coefficients were initially parameterized for solid Li_2BeF_4 crystal [?]. It was found that this potential has difficulties in predicting the properties of liquid FLiBe [?], and a softer behavior of the liquid FLiBe potential is necessary to better predict physical properties at high temperature [?].

Another classical potential is the Polarizable Ionic Interaction Potential (PIM) which involves polarization effects. The PIM developed previously [?] showed high accuracy compared to experiment over the properties. The PIM was constructed from ab initio calculations [?]. Specifically, the parameters of PIM were fitted by minimizing the difference between the ab initio data (energies, forces, etc.) and those predicted by PIM model for a given structure. However, the accuracy and flexibility are constrained by the given function form for the PIM model.

The ab initio molecular dynamics (AIMD) based on quantum mechanics is regarded as a highly accurate method. Wang et al. [**?**] used AIMD method to obtain the VACF for calculating the generalized frequency spectrum of liquid FLiBe. However, the computational costs of AIMD makes it difficult to predict complex properties where large system size and long time scale are needed, including the generalized spectrum and diffusion coefficient (calculated by the slope of MSD) which are required for the TSL evaluation. The MSD is dependent on the dynamics behavior over a long time scale, and the generalized frequency spectrum also requires large scale and long time scale to converge to the desired accuracy.

Machine learning force field (MLFF), also called machine learning potential, offers a breakthrough that balances the accuracy and efficiency by learning potential functions from high-precision ab initio data through machine learning models \cite{28-34}. As demonstrated by previous deep learning potential MD (DPMD) [**?**] and moment tensor potential (MTP) MD [**?**] focusing on FLiBe, MLFF can reproduce thermophysical properties of FLiBe within experimentally acceptable accuracy. Recent studies [**?**, **?**, **?**, **?**] showed that MLFF has comparable accuracy with ab initio simulation in predicted forces and energies while achieving 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 the evaluation of the TSL data of liquid FLiBe by combining the MLFF with the QC based TSL calculation method. The MLFF method implemented in VASP was used to generate the MLFF for FLiBe. The MD simulations based on MLFF (MLFF-MD) were performed to obtain parameters required for TSL calculation. The QC method implemented in NECP-Atlas was used to calculate the TSL.

The structure of this paper is organized as follows. Section II describes the methodologies for TSL evaluation and MLFF generation. Section III shows the results of physical properties of FLiBe predicted by MLFF, and the thermal neutron scattering cross sections obtained from the evaluated TSL in the present work. Some 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 [**?**], the double differential scattering cross section is reduced and can be calculated as:

$$\frac{d\mu}{dE'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}\left(\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)\right)$$

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 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, α and β are the dimensionless momentum transfer and dimensionless energy transfer, which are defined as:

$$\alpha = \frac{E' + E - 2\mu\sqrt{EE'}}{Ak_{BT}} = \frac{(\hbar\kappa)^2}{2Mk_{BT}}$$

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

where A is the mass ratio of scattering target nuclei to neutron, M is the mass of scattering target nuclei, $\hbar$ is the Planck constant divided by 2π, 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 which quantifies the contribution to the system dynamics from the atom itself, and $S(\alpha, \beta)$ is the coherent scattering law which is expressed as:

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

where $S_d(\alpha, \beta)$ is the distinct scattering law, which quantifies the contribution to the system dynamics from the interference 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 the present work, $S_s(\alpha, \beta)$ is calculated using the QC method. According to the Van Hove theory [**?**], $S_s(\alpha, \beta)$ can be defined as the Fourier transform of the self-intermediate scattering function $I_s(\alpha, t')$:

$$S_s(\alpha, \beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} I_s(\alpha, t') \exp(-i\beta t') dt'$$

where $\hbar$ is the Planck constant divided by 2π, $t' = k_{BTt}/\hbar$.

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

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

where $\gamma(t')$ is the quantum width function represented as [**?**, **?**]:

$$\gamma(t') = \int_0^\infty \frac{2M}{k_B\beta^2} f_c(\beta) \left\{ \frac{\exp(\beta/2)}{2\sinh(\beta/2)}[1-\cos(\beta t')] - i\frac{\sin(\beta t')}{2} \right\} d\beta$$

where M is the mass of the particle, $f_c(\beta)$ is the classical characteristic function, which can be evaluated from MSD [**?**]:

$$f_c(\beta) = \frac{\beta^2}{Mk_{BT}} \int_0^\infty \exp(i\beta t') MSD(t') dt'$$

As for the calculation of $S_d(\alpha,\beta)$, the incoherent approximation where $S_d(\alpha,\beta)$ in Eq. (1) is directly ignored is adopted conventionally, consequently, Eq. (1) becomes:

$$\frac{d\mu}{dE'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}(\sigma_{coh}+\sigma_{inc})S_s(\alpha,\beta)$$

However, as shown in Table 1 , the ratio $\sigma_{coh}/(\sigma_{coh}+\sigma_{inc})$ is non-negligible for each nuclide in FLiBe compared to that of H, indicating that neglecting the distinct scattering term $\sigma_{coh}S(\alpha,\beta)$ may therefore affect the total scattering law of FLiBe.

Therefore, to include the distinct scattering, $S(\alpha,\beta)$ in the present work is calculated with the Sköld approximation [**?**]:

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

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

$$S_i(\kappa) - 1 = \frac{\sum_j c_j b_j^{coh}[S_{ij}(\kappa)-1]}{\left(\sum_j c_j b_j^{coh}\right)^2}$$

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

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

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 the i-th and j-th atoms, and $g_{ij}(r)$ is the radial distribution function (RDF) between the i-th and j-th atoms.

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

B. MLFF Generation

The generation of MLFF requires a training dataset generated by ab initio calculations, containing the collection of Bravais matrices, atom positions, energies, forces, and stress tensors. The accuracy of MLFF depends on the quality of the training dataset used for machine learning. Thus, selecting an appropriate size of training dataset is essential for the accuracy of MLFF. However, larger training size requires greater computing cost. To solve this problem, the so-called on-the-fly MLFF algorithm [**?**, **?**, **?**] implemented in VASP that used active learning strategy during the MD simulations was adopted in the present work. In the algorithm, MD simulations are performed during training, and the accuracy of the force field is estimated based on the Bayesian error estimation (BEE) of forces calculated at each MD step. For each MD step, if the estimated Bayesian force error for all atoms exceeded a set threshold, then DFT calculation is performed; otherwise, MLFF is used in this MD step. This algorithm is designed to selectively incorporate structures which are predicted poorly by MLFF into the training set, avoiding excessive DFT calculations to enhance computational efficiency.

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

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

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

During the above training process, DFT calculations were carried out to generate the training dataset. The computational settings for these calculations are as follows. The Generalized Gradient Approximation (GGA) of Perdew-Burke-Ernzerhof (PBE) functional was used for the exchange-correlation functional. An energy cutoff of 600 eV for the plane-wave basis set was set and Gaussian smearing with a width of 0.05 eV was used to determine the partial occupancies for each orbital in the system. The energy convergence of 10^{-6} eV was set for the electron self-consistent loop, and a single gamma point was used for the Brillouin zone sampling.

In all the calculations, the projector-augmented-wave (PAW) potentials were used. It has been pointed out that the proper description of van der Waals (vdW) interactions is important for the prediction of density for FLiBe [**?**, **?**]. Thus, the DFT-D3 method with Becke-Johnson damping function was used to perform the dispersion correction.

In the training process, BEE of forces was used as a training monitoring quantity. The Bayesian error is an estimate for the average error of the training set when considering a new random configuration, and it reflects the real error between predicted values and DFT values for the current configuration. When a significant jump occurs in the BEE curve, it indicates that the current configuration is quite different from the previously learned structures, and the forces predicted by MLFF are thus expected to deviate from the reference AIMD results. Since the amount of forces in the training dataset is much larger than that of energy [**?**], force was chosen as the criterion for whether to carry out DFT calculation.

The threshold for the BEE of forces depends on the system and temperature. The typical attainable values for threshold provided on the VASP [**?**] website are 0.02 eV/Å around 300-500 K, and 0.06 eV/Å around 1000-2000 K. As the FLiBe system in this paper is a liquid system at high temperatures, which usually has a higher error than high symmetry solids at low temperature, the initial thresholds for the three parts of the force field training were set as 0.03, 0.02, and 0.02 eV/Å respectively. Furthermore, the threshold used in this paper is a variable controlled by the on-the-fly algorithm of VASP, which is updated during the MLFF training process.

Fig. 1 [Figure 1: see original paper] shows the BEE, root mean square errors (RMSE), and threshold criterion of force during the training process. As shown in Fig. 1, in the final cooling simulation, the maximum value of BEE is 0.042 eV/Å; the BEE remains below the threshold at nearly every MD step, and the RMSE of force remains stable at around 0.043 eV/Å with the force field barely updating. Earlier studies [**?**] have pointed out that force errors smaller than 0.1 eV/Å are generally sufficient for predicting properties such as density, RDF, diffusion coefficient, and so on. Therefore, it is considered that the force field meets the requirements for predicting the properties.

To assess the predictive accuracy of the trained force field, we built a testing set of 5000 structures from ab initio structures randomly selected from AIMD simulation based on DFT from 800 K to 1600 K, and the testing set is unrelated to the training set. The energy and force predictions for training set and testing set are shown in Fig. 2 [Figure 2: see original paper], and the RMSE of energy and force are given in Table 2 . The results demonstrate good agreement between MLFF-MD and AIMD calculations. The excellent accuracy of energy and force predicted by MLFF provides a good basis for the MLFF-MD simulations in large systems with long time scales, which can be used for property prediction of FLiBe system from 800 K to 1600 K.

C. The Calculation Flowchart of TSL Data of Liquid FLiBe

In this section, we provide a summary of the evaluation framework for the TSL data of liquid FLiBe adopted in the present work, as shown in Fig. 3 [Figure 3: see original paper]. The workflow is as follows: (i) Train the MLFF for liquid FLiBe based on on-the-fly MLFF algorithm. (ii) Test the predicted forces and energies by MLFF on a testing set containing DFT data. (iii) Validate the trained MLFF against the theoretical results and experimental data of liquid FLiBe properties, including density, diffusion coefficient, RDF, and VACF. The theoretical results used for validation include our AIMD results and available theoretical results from other works. (iv) Calculate the TSL of liquid FLiBe using the QC method based on the MSD and RDF from MLFF-MD.

III. Results and Discussion

A. Properties of Liquid FLiBe

In this section, the properties of liquid FLiBe, including density, RDF, VACF, diffusion coefficient, and electrical conductivity, are predicted to show the accuracy of the trained MLFF. The MLFF-MD simulations were performed with a larger FLiBe system of 7000 atoms. Before the production MD simulations used for calculating the properties, we first performed NPT simulations for 10 ps at several temperatures to make sure that the initial systems for the production MD simulations reach the equilibrium state. Then, several production MD simulations were performed at NPT ensemble using the MLFF at each temperature to obtain the properties. The time step and pressure of all the simulations were set as 1 fs and 0 bar, respectively.

1. Density In Fig. 4 [Figure 4: see original paper], the density against temperature obtained by MLFF-MD is shown and compared with other theoretical [**?**, **?**, **?**] and experimental results \cite{46-48}. It can be seen that the temperature fitting curve of density obtained by MLFF-MD is close to the results of MD simulations based on the machine-learning potential MTP [**?**] noted as MTP-MD and AIMD [**?**]. The results of DPMD [**?**] underestimate the experimental values, and this is mainly due to the absence of dispersion corrections in their training data, which has been proved to overestimate the volume of FLiBe [?]. From the results obtained by this work at the temperature range from 800 K to 1600 K, we fitted a density-temperature equation as shown below:

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

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

2. Radial Distribution Function The RDF is calculated as follows:

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

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

The RDF of F^--F^-, Li^+-F^-, and Be^{2+}-F^- in FLiBe at 973 K obtained by MLFF-MD and AIMD calculations are compared and shown in Fig. 5 [Figure 5: see original paper]. The RDF curves of MLFF-MD simulations well reproduce those by AIMD simulations performed in the present work. The first peak of RDF for Be-F pair is sharp and high, and the minimum after the peak in the RDF curve approaches zero, while the first peak of RDF for Li-F pair is broader with a relatively high minimum. This indicates that the local structures of Li^+ and Be^{2+} are quite different, and the bonding of Be^{2+} and F^- is stronger.

Based on the RDF, the average near-neighbor distances and the first shell coordination numbers can be calculated. Table 3 lists the average near-neighbor distances. The average near-neighbor distances for Be^{2+}-F^-, F^--F^-, and Li^+-F^- predicted by MLFF-MD are 1.545 Å, 2.589 Å, and 1.853 Å, respectively, showing good agreement with the AIMD results performed in the present work, experimental results [?], and other theoretical results including the AIMD simulation results [?], the machine-learning potential MTP-MD [?] and DPMD [?], and the CMD results of PIM-MD [?]. The first shell coordination numbers obtained with different methods are shown in Table 4 . The predicted results are also consistent with those based on AIMD simulations performed in the present work, AIMD [?], MTP-MD [?], and PIM-MD [?]. However, it is found that the theoretical results differ from the experimental results, especially the coordination number of F^--F^-. The difference was documented to be due to the fact that the minimum after the first peak of RDF, which directly affects the calculated coordination number, could not be well determined over the wide flat region [?].

3. Velocity-Auto Correlation Function Based on the atomic velocities obtained by MLFF-MD and AIMD, the VACF were calculated as:

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

where $v_i(0)$ and $v_i(t)$ denote the velocity of the i-th atom in the ensemble at time 0 and time t, respectively. As shown in Fig. 6 [Figure 6: see original paper], the VACF predicted by MLFF-MD matches well with AIMD simulations. The VACF curves decay first, and oscillate until converging to zero. The Be^{2+} ions exhibit the fastest VACF decay rate, followed by Li^{+}, with F^{-} showing the lowest decay rate. Compared to F^{-} and Li^{+}, Be^{2+} seems to show a longer oscillation time up to about 150 fs. This was attributed to the formation of the fluoro-beryllate anion ${BeF_4}^{2-}$ [**?**].

4. Diffusion Coefficient The diffusion coefficient can be obtained from the slope of MSD:

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

where $r_i(t)$ and $r_i(0)$ are the position of the i-th atom in the system at time t and 0, and N is the number of atoms.

The MSD of Be, F, and Li in FLiBe from 800 K to 1600 K were calculated from MLFF-MD simulations. Fig. 7 [Figure 7: see original paper] shows the MSD of Be, F, and Li in FLiBe from 800 K to 1600 K calculated from the MLFF-MD simulations for 10 ps with a timestep of 1 fs. With the increase of time, the ions in FLiBe continue to move and the MSD increases, showing the linear relationship between MSD and time in liquid FLiBe.

In ENDF libraries, the TSL of inelastic scattering is stored in the form of a two-dimensional interpolation table of α and β, thereby requiring a set of input α and β values. In this work, MSD is the main input parameter for the self-scattering law calculation. For TSL in a specific range of α, the input is the MSD with specific time step, which means the time step of MSD for TSL calculation should be determined by the magnitude of α. The MSD with larger time step and longer total time was used for lower α, and the MSD with smaller time step and shorter total time was used for larger α. Table 5 shows the different time step and total time of MSD files in this work for different α ranges.

Fig. 8 [Figure 8: see original paper] shows the diffusion coefficients of Be, F, and Li ions in FLiBe calculated by MLFF-MD in this work, comparing with those calculated by machine-learning potential based MD including MTP-MD [**?**] and DPMD [**?**], the CMD of PIM-MD [**?**], and experimental results [**?**, **?**]. Compared with other works, our results are close to the results of PIM potential.

As shown in Fig. 8, the theoretical results of the diffusion coefficients of Li and Be are all lower than experimental data. It was pointed out in previous works [?, ?, ?] that the experimental values may contain errors for the relatively large values and high activation energy.

5. Electrical Conductivity Based on diffusion coefficient, the electrical conductivity can be given by the Nernst-Einstein equation [?]:

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

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

Based on Eq. (17), the electrical conductivity was calculated from the diffusion coefficients obtained in Section III A 4, and compared with the experimental electrical conductivity [?] in Fig. 9 [Figure 9: see original paper]. The electrical conductivities calculated from the diffusion coefficients obtained by MLFF-MD are close to the experimental curve.

B. Thermal Neutron Scattering Cross Sections of FLiBe

Based on MSD and RDF calculated from MLFF-MD simulations, the TSL and thermal neutron scattering cross sections of liquid FLiBe were calculated using the QC method implemented in nuclear data processing code NECP-Atlas. In order to better compare the influence of force field on the TSL, the cross sections of FLiBe were also calculated using the QC method based on CMD simulation with Born-Mayer-Huggins potential the same as previous works [?, ?, ?].

The newly released evaluated nuclear data library ENDF/B-VIII.1 [?] has included the TSL data of liquid FLiBe evaluated by Zhu et al. [?] where the TSL was separated into two partial TSLs for bound vibrational mode and diffusive mode under the incoherent approximation. Here we also show the comparison between the results by the present work and ENDF/B-VIII.1 evaluation.

1. Double Differential Scattering Cross Section In Fig. 10 [Figure 10: see original paper] to Fig. 12 [Figure 12: see original paper], the double differential scattering cross sections (DDSCS) of Be, F, and Li at the incident energy $E = 0.075$ eV were calculated under the incoherent approximation and compared with those obtained from ENDF/B-VIII.1. As can be seen, the peak values of the DDSCS calculated based on MLFF-MD are relatively higher, while the DDSCS based on CMD simulations are closer to those of ENDF/B-VIII.1. This is mainly due to the difference of the MSD calculated using different potentials. As shown in Table 6 , the diffusion coefficients predicted by MLFF-MD are much lower than the values predicted by CMD simulations and those given in Zhu' s work [?], where the same potential as the ENDF/B-VIII.1 FLiBe evaluation [?] was used.

As shown in Fig. 10 to Fig. 12, the up-scattering ($E' > E$) is more dominant than down-scattering ($E' < E$). When the scattering angle increases, the DDSCSs broaden to a wider range due to the higher momentum transfer. Meanwhile, the inelastic scattering becomes dominant, along with the decrease of the magnitude of the quasi-elastic scattering peak around the energy transfer $\hbar\omega = E - E' = 0$. Because of the lower diffusion coefficients predicted by MLFF-MD than CMD, the translational diffusion of FLiBe is more suppressive, which results in a narrower quasi-elastic scattering peak. Therefore, the scattering intensity is concentrated over a narrower energy transfer range, resulting in a higher peak of the DDSCS.

2. Integrated Scattering Cross Section Fig. 13 [Figure 13: see original paper] shows the integrated thermal scattering cross sections of Be, F, Li, and FLiBe at 873 K, which were calculated using the incoherent approximation. It can be seen that the cross sections of Be and Li calculated based on MLFF-MD are slightly lower than those of ENDF/B-VIII.1 in the energy range from 10^{-5} eV to 10^{-2} eV, and the cross section of F based on MLFF-MD is significantly lower than ENDF/B-VIII.1 in the same energy range. The deviation between the thermal scattering cross sections of MLFF-MD and those of ENDF/B-VIII.1 arises from differences in force fields and TSL calculation method. Considering the influence of cross section of each nuclide, the total scattering cross section of FLiBe is lower than ENDF/B-VIII.1 in the cold neutron energy region less than 5×10^{-3} eV. Moreover, obvious differences of scattering cross sections can be seen when comparing our MLFF-MD results with CMD results due to the difference of force fields.

The results also show that our CMD results with Born-Mayer-Huggins potential are different from ENDF/B-VIII.1 on each nuclide, and close to ENDF/B-VIII.1 on the total cross section, suggesting the important influence of TSL evaluation methodology.

Fig. 14 [Figure 14: see original paper] shows the thermal scattering cross sections of Be, F, Li, and the total FLiBe at six different temperatures from 800 K to 1600 K calculated by incoherent approximation. The thermal scattering cross section obtained in this work becomes larger when the temperature increases, and converges to the free atom cross section at energies above 1 eV, which is consistent with theory.

3. Effect of Distinct Scattering for Thermal Scattering Law The thermal scattering cross sections shown above were calculated with the incoherent approximation where $S_d(\alpha, \beta)$ in Eq. (4) is ignored as described in Section II. The $S_d(\alpha, \beta)$ of liquid materials can be included in $S(\alpha, \beta)$ by using the Sköld approximation. The Sköld correction factors $S(\kappa)$ of F, Li, and Be at 873 K were calculated with Eqs. (11) and (12), and shown in Fig. 15 [Figure 15: see original paper]. The Sköld correction factor curve for Be appears to be slightly negative at several κ points between 2.6 and 2.8 Å^{-1}, which violates the physical condition $S(\kappa) \geq 0$.

In order to avoid non-physical negative values of TSL due to these negative points, we smoothly increased these minor negative $S(\kappa)$ values to small positive values while ensuring that the shape of the original negative peak remained unchanged. Since the magnitudes of negative values in $S(\kappa)$ of Be are small and only present in a narrow region of 0.2 Å^{-1}, its influence on the thermal scattering law is negligible.

In Fig. 16 [Figure 16: see original paper], the thermal scattering cross sections of Be, F, and Li in FLiBe at 873 K calculated by Sköld approximation are compared with those calculated by incoherent approximation. Fig. 17 [Figure 17: see original paper] shows the total scattering cross sections of FLiBe ($2LiF\text{-}BeF_2$) at 873 K calculated by Sköld approximation and incoherent approximation. As can be seen, the results of Sköld approximation show some oscillations due to distinct scattering. In the evaluated nuclear data libraries such as ENDF/B-VIII.1 and JENDL-5, similar oscillations have also been observed in the total cross sections of D_2O after considering the distinct scattering term with the Sköld approximation. In the Sköld approximation calculation, the used correction factors $S(\kappa)$ are derived from the radial distribution function (RDF) calculated by molecular dynamics (MD) simulations. The $S(\kappa)$ contains spatial correlation information among the ions in FLiBe, directly reflecting the static structural characteristics of FLiBe. Based on this structural input, the Sköld approximation predicts the coherent scattering law. Consequently, the oscillations observed in the thermal scattering cross section are fundamentally linked to the microscopic structure of FLiBe. The results of Sköld approximation for Be and F show more obvious deviations than Li when comparing to the results calculated with incoherent approximation, which can be explained by the fact that the σ_{coh} of Be and F as shown in Table 1 dominate the summation of σ_{coh} and σ_{inc}.

Due to the lack of experimental data, verifying the cross section has become a problem. The measurement of cross sections for FLiBe is needed to confirm the validity of the theoretical results and guide the possible improvement of the evaluation method.

IV. Conclusion

In this work, a MLFF for liquid FLiBe was trained with the on-the-fly machine learning algorithm implemented in VASP, to evaluate the thermal neutron scattering law. The properties including density, RDF, VACF, diffusion coefficient, and electrical conductivity of liquid FLiBe were predicted using the trained MLFF, and compared with our AIMD calculations, available theoretical results from other works, and experiments. The results showed that properties predicted by MLFF-MD are generally in good agreement with experiments and AIMD calculations.

The thermal scattering cross sections of FLiBe from 800 K to 1600 K were calculated based on MLFF-MD and compared with those calculated based on CMD simulation, and those calculated based on the latest evaluated nuclear data file ENDF/B-VIII.1. Our MLFF-MD and CMD results both employed the same QC method, thus the discrepancies of their thermal cross sections demonstrate that the force field significantly influences the evaluated TSL. The comparative analysis of thermal scattering cross sections calculated using the Sköld approximation versus the incoherent approximation reveals that the thermal scattering cross sections of liquid FLiBe accounting for distinct scattering are quite different from those which ignore distinct scattering. Due to the differences in TSL calculation method and the force fields used for MD, the thermal scattering cross sections calculated using the incoherent approximation based on MLFF-MD show deviations from those calculated based on ENDF/B-VIII.1.

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