Thermal neutron scattering data for $^7\text{LiF}$ and $\text{BeF}_2$

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Abstract. Based on the coherent elastic, incoherent elastic, coherent inelastic and incoherent inelastic scattering processes, a code named SIRIUS is developed to produce thermal neutron scattering data for crystals in ENDF-6 format. The phonon band structures and projected phonon densities of states of $^7\text{LiF}$ and $\text{BeF}_2$ crystals were calculated by Hellman-Feynman Theorem combined with a lattice dynamics direct method. Finally, the thermal neutron scattering data for $^7\text{LiF}$ and $\text{BeF}_2$ crystals are given.

1. Introduction

Thermal neutron scattering data are widely used in nuclear engineering applications, such as reactor design, radiation shielding, long-lived nuclear waste transmutation, Boron Neutron Capture Therapy. The study of thermal neutron scattering started from 1950s, which is due to the needs of the reactor design. Subsequently, GASKET code was developed at General Atomic [1,2], and was used to generate early thermal neutron scattering data. Now, LEAPR module in NJOY [3,4] code is widely used to calculate ENDF-6 format thermal neutron scattering law (TSL) data.

At present, some evaluated nuclear data libraries such as ENDF/B library [5], JENDL library [6], JEFF library [7] and IAEA nuclear data library contains the sublibrary of TSL data for about 23 moderators. However, new requirements for the development of current nuclear engineering, especially for the fourth generation reactor design need more TSL data. For example, in molten salt reactor, TSL data for Beryllium fluoride and Lithium fluoride are needed. Mei and Cai applied the CASTEP code and the modified NJOY code to generate the thermal neutron scattering data for LiF and BeF$_2$ crystals [8] and investigated the thermal neutron scattering data for molten salt Flibe based on the dynamic performance for Flibe [9]. To date, no thermal neutron scattering data in the ENDF format are available for Beryllium fluoride and Lithium fluoride.

Recently, a code named SIRIUS [10] is developed at IAPCM to generate the thermal neutron scattering data for solid in ENDF-6 format. In this paper, SIRIUS code is utilized to calculate the thermal neutron scattering data of $^7\text{LiF}$ and $\text{BeF}_2$ crystals.

2. Theoretical models

The theory of scattering of thermal neutron by crystals is exposed in details in many textbook [11,12] and is briedly considered here. There are four types of scattering, coherent elastic scattering, incoherent elastic scattering, coherent inelastic scattering and incoherent inelastic scattering.

The expression of the coherent elastic scattering cross section for polycrystalline material is:

$$\sigma_{el,coh}(T) = \frac{\sigma_{coh} \pi^2 h^2}{2N\nu_0 m E} \sum_{\tau} \frac{|F(N)|^2}{\tau} \exp(-4W(T) E_i) \quad (1)$$

The expression of the differential incoherent inelastic scattering cross section is:

$$\frac{d\sigma_{inc}(T)}{d\Omega} = \frac{\sigma_{inc}}{4\pi} \exp(-2W(T) E(1-\mu)) \quad (2)$$

Where $W(T) = \frac{1}{2\pi^2} \int_0^{\beta_{max}} \frac{1}{\beta^2} \coth(\frac{\beta}{2}) \rho(\beta) \beta d\beta$ is the Debye-Waller integral divided by the atomic mass, $T$ is the temperature of the scattering medium, $N$ and $\nu_0$ are the number of basis atoms and the volume of the primitive unit cell, respectively. $m$ is the mass of the neutron, $|F(N)|$ is the crystallographic structure form factors, $\tau$ is the length of reciprocal vectors, $E_i$ are the “Bragg edges”, $E$ is the incident neutron energies in the laboratory system, $\mu$ is the cosine of the scattering angle, the subscript coh means coherent and inc means incoherent. $\sigma_{coh}$ and $\sigma_{inc}$ are the incoherent and coherent bound scattering cross section for the material, respectively. For mixed moderator, the coherent elastic scattering cross section is more complicated and can be written as:

$$\sigma_{el,coh}(T) = \frac{\pi^2 h^2}{2N\nu_0 m E} \sum_{\tau} \left[ \sum_{j=1}^{N} \sqrt{\sigma_{coh,j} \exp(-2W_j(T) E_i) \exp(i2\pi \tau \cdot \vec{d}_j)} \right]^2 \quad (3)$$

Where the subscript $j$ means the $j$th atom in the unit cell. $\tau$ is the reciprocal lattice vector, $\vec{d}$ is atomic position of the
The calculation procedure for coherent elastic scattering of the mixed material goes as follows. Firstly, the Eq. (3) is used to calculate the whole coherent elastic scattering cross section is incoherent approximation is used [13]. The incoherent approximation is considered so small that can be neglected, and the thermal neutron scattering law is given as

\[ S_{\text{coh}}(\alpha, \beta, T) = \frac{3\hbar^2}{2mk_B T} \exp \left( \frac{\beta}{2} \right) \frac{1}{\Delta \beta \beta} \left( \sum_{j} \rho(\beta_j) \sum_{\alpha, \beta, \Delta \alpha, \Delta \beta} F_{\alpha, \beta}^{2}(\vec{\alpha}, \vec{\beta}) \Delta \beta \beta \right) \]

(8)

where \( F_{\alpha, \beta}^{2}(\vec{\alpha}, \vec{\beta}) \) is the scattering vector, \( \vec{e}_{\alpha, \beta} \) is the polarisation vector of the scattering model in position \( d \), \( s \) stands for the double index \( q \) and \( j, j \) is the polarisation index (\( j = 1, 2, 3 \)). \( q \) is the wavevector. The conditions \( q = \vec{q} - q_j \) and \( \beta = \beta_j \) must be satisfied in the calculation. The calculated scattering law is an average value in \( [\alpha, \alpha + \Delta \alpha] \) and \( [\beta, \beta + \Delta \beta] \).

Finally, based on the theory, a code program SIRIUS is constructed to generate thermal neutrons scattering data in ENDF-6 format. In order to calculate coherent inelastic scattering, both the PDOSs and the phonon dispersion relation for materials are needed, otherwise, PDOSs for materials are enough.

3. Computational details

3.1. The phonon density of states

Beryllium fluoride (BeF\(_2\)) has a \( \alpha \)-quartz-type structure and belongs to space group P3121 [18] with three beryllium atoms and six fluoride atoms in the unit cell. Be: (0.4782, 0, 1/3), (0, 0.4782, 2/3), (0.5218, 0.5218, 0) and belongs to space group FM3M [19] with four lithium fluoride atoms and four fluoride atoms in the unit cell. F: (0, 0, 0), (0, 1/2, 1/2), (1/2, 0, 1/2), (1/2, 1/2, 0). The phonon band structures and projected phonon densities of states for BeF\(_2\) crystal and the structure for 7LiF crystal and the structure for 7LiF crystal is given as

\[ S_{\text{coh,1}p}(\alpha, \beta, T) = \frac{3\hbar^2}{2mk_B T} \exp \left( \frac{\beta}{2} \right) \frac{1}{\Delta \beta \beta} \left( \sum_{j} \rho(\beta_j) \sum_{\alpha, \beta, \Delta \alpha, \Delta \beta} F_{\alpha, \beta}^{2}(\vec{\alpha}, \vec{\beta}) \Delta \beta \beta \right) \]

(8)

The magnitude of reciprocal space vector of BeF\(_2\) is

\[ |\tau| = 2\pi \sqrt{4/3a^2(h^2 + k^2 + l^2)} + 1/c^2l^2 \]

(9)

Lithium fluoride (7LiF) has a face centred cubic structure and belongs to space group Fm3m [19] with four lithium atoms and four fluoride atoms in the unit cell. F: (0, 0, 0), (0, 1/2, 1/2), (1/2, 0, 1/2), (1/2, 1/2, 0) Li: (1/2, 1/2, 1/2), (1/2, 0, 0), (0, 1/2, 0), (0, 0, 1/2). The magnitude of reciprocal space vector of 7LiF is

\[ |\tau| = 2\pi \sqrt{(h^2 + k^2 + l^2)/a} \]

(10)

The computed lattice parameters of BeF\(_2\) and 7LiF are in well agreement with experimentally reported [20–22]. The lattice constants values for BeF\(_2\) are \( a = 4.85 \) Å, and \( c = 5.335 \) Å. The lattice constants value for 7LiF is \( a = 4.026 \) Å. The structure for BeF\(_2\) crystal and the structure for 7LiF are shown in Fig. 1.

The phonon band structures and projected phonon densities of states for 7LiF and BeF\(_2\) crystals were calculated by Hellman-Feynman Theorem combined with a lattice dynamics direct method using the Vienna Ab initio Simulation Package (VASP) and the PHONON code [23,24]. For 7LiF crystal, the model used a 3 × 3 × 3 supercell composed of 216 atoms and for BeF\(_2\)
Figure 1. Structure of BeF$_2$ crystal (left)/LiF crystal (right).

Figure 2. The calculated partial PDOSs of $^7$Li and F atoms.

Figure 3. The calculated partial PDOSs of Be and F atoms.

crystal, the model used a $2 \times 2 \times 2$ supercell composed of 72 atoms. The k-point meshes in the full edge of the Brillouin were sampled by $8 \times 8 \times 8$. Figure 2 shows the calculated partial POSs of $^7$Li and F atoms for $^7$LiF. Fig. 3 shows the calculated partial POSs of Be and F atoms for BeF$_2$. There both were compared with the result from Mei and Cai [8].

Obviously, the calculated partial PDOSs of LiF are similar as the result from Mei and Cai [8], but the calculated partial PDOSs of BeF$_2$ are different. BeF$_2$ crystal is low symmetrical crystal, need more supercell in the calculation to get the reasonable result. The same method were used to calculate the PDOSs, however different codes and initial inputs were used. To validate these, more experimental data are needed.

3.2. Thermal neutron scattering data

Based on the PDOS given above, and the bound atom cross section given in Table 1 [25], the thermal neutron scattering law data were calculated using SIRIUS code. Here the incoherent approximation was used in inelastic scattering.

According to the bound atom cross section, the incoherent cross section and coherent cross section are same important for $^7$Li. So the thermal neutron scattering data for $^7$LiF crystals were divided into four parts. The first part was the coherent elastic scattering data for the whole $^7$LiF crystal. The second part was the incoherent inelastic scattering data for F in $^7$LiF. The third part was the incoherent inelastic scattering data for $^7$Li in $^7$LiF. The last part was the incoherent elastic scattering data for $^7$Li in $^7$LiF. In this work, the interference effects in inelastic didn’t considered, and the same in BeF$_2$. Figure 4 shows the thermal neutron scattering cross section for $^7$LiF crystal compared with the result from Mei and Cai [8] at 300 K.

Table 2 shows the result of effective temperatures and Debye-Wall integrals divided by the atomic mass for $^7$LiF crystal.

For BeF$_2$ crystal, the incoherent cross section can be neglected, so the thermal neutron scattering law data for BeF$_2$ crystals were divided into three parts. The first part was the coherent elastic scattering data for the whole BeF$_2$ crystals. The second part and the third part were incoherent inelastic scattering data for Be in BeF$_2$ and F in BeF$_2$, respectively. Figure 5 shows the thermal neutron scattering cross section for BeF$_2$ crystal compared with the result from Mei and Cai [8] at 300 K.

Table 3 shows the result of effective temperatures and Debye-Wall integrals divided by the atomic mass for BeF$_2$ crystal.

As showed in Fig. 4 and Fig. 5, the present scattering cross sections have significant difference with the Ref. [8]. For LiF crystal, the present PDOSs of Li and F are similar to the reference, but the present total inelastic scattering cross section is about twice as much as the reference. Maybe in the reference work, the input value of number
Table 3. Effective Temperatures and Debye–Waller integrals divided by the atomic mass for BeF₂ crystal.

| T (K) | Be in BeF₂ | F in BeF₂ |
|-------|------------|-----------|
|       | W(eV⁻¹)   | T_{eff}(K) | W(eV⁻¹)   | T_{eff}(K) |
| 300   | 6.819620  | 542.1     | 18.38910  | 472.4     |
| 400   | 8.762772  | 596.4     | 24.34908  | 541.1     |
| 500   | 10.74532  | 663.8     | 30.33266  | 618.5     |
| 600   | 12.75204  | 739.9     | 36.32943  | 701.6     |
| 700   | 14.77426  | 821.8     | 42.33428  | 788.8     |

of principal atoms in THERMR module for NJOY code was not set correctly. The difference in the elastic Bragg edges might be caused by the magnitude of reciprocal space vector. In NJOY code, the magnitude of reciprocal space vector for FCC structure is not the same as this work. For BeF₂ crystal, difference of the fractional coordinates of atoms in unit cell and PDOSs are all the reasons, which cause the different scattering cross sections.

4. Conclusions

In summary, the thermal neutron scattering cross section for ⁷LiF and BeF₂ crystals have been calculated in this work using a combination of ab-initio simulations, lattice dynamics, and SIRIUS code. Compared with the Ref. [8], obvious difference can be found in PDOSs and scattering cross sections, the possible cause which may lead these had been analysed in the previous section. However, to validate these calculated thermal neutron scattering data, experimental data are needed.

More work should be done in future. The first is to improve SIRIUS code for liquid materials. The second is to generate more thermal neutron scattering data for other materials such as silicon carbide and molten salt Flibe and so on.

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