Response Letter

Article title: First-principles calculations of zone center phonons and related thermal properties of $MgSiN_2$

Author: Chaiyawat Kaewmeechai, Yongyut Laosiritaworn and Atchara Punya Jaroenjittichai*

Corresponding author: Atchara Punya Jaroenjittichai

Corresponding author's email address: atcharapunya@gmail.com

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Dear Editor and Reviewer

Thank you very much for your comments and suggestions. We already removed typos and improved the writing style more carefully. The reviewer's comment and our responses are listed below. The changes are highlighted in the revised manuscripts.

Comment :

The authors employed first-principles calculations to investigate phonons and thermal properties of $MgSiN_2$. The quality and results of this work is suitable for publication. However, the writing style is encouraged to be improved as well as the typos should be removed. For example "The irreducible symmetry classification gives 11 modes for LO and TO branches of B_1, B_2 and A_1 corresponding to directions x, y and z while only 12 modes for A_2." "At low frequency, the phonon branches are very difficult to see the LO-TO splitting." "However, we found that their C_p are greater than our C_v within 2.2% in range of 80 -800 K [17, 18] and go increasingly greater with increase temperature [18- 19]" "Note that, the assumption of $C_p \approx C_v$ and be use only.." These sentences are difficult to understand.

Responses :

We have rewritten some sentences in the manuscripts and give more explanations here.

1) " The irreducible symmetry classification gives 11 modes for LO and TO branches of B_1, B_2 and A_1 corresponding to directions x, y and z while only 12 modes for A_2."

Rewritten as :

"The 78 vibrational modes consist of 11 longitudinal optical (LO) modes and 11 translational optical (TO) modes for each B_1, B_2 and A_1 and 12 optical modes for A_2. The LO-TO splitting disappears in case of A_2 because it cannot be represented by a component of a vector *x*, *y* or *z* like B_1, B_2 and A_1."

Explanation :

There are 12 modes for each symmetry representation (48 modes in total). The lowest modes of B_1, B_2, and A_1 are all zero (acoustic modes). Thus, from considering the C_{2v}-character table with the changes in phonon frequencies due to the external electric filed along x,y, and z directions, we will obtain 11 modes for A_1, B_2, B_1 modes and 12 modes for A_2. When we include LO-TO splitting, it turns to $11(A_{1L}) + 11(A_{1T}) + 11(B_{1L}) + 11(B_{1T}) + 11(B_{2L}) + 11(B_{2T}) + 12\ (A_2) = 78$ modes.

2) "At low frequency, the phonon branches are very difficult to see the LO-TO splitting."

Rewritten as :

"At Γ point LO-TO spitting can be observed from discontinuity of phonon branches. At low frequency, we indeed observe small LO-TO splitting due to small change in energy."

3) " However, we found that their C_p are greater than our C_v within 2.2% in range of 80 - 800 K [17, 18] and go increasingly greater with increase temperature [18- 19]"

4) " Note that, the assumption of $C_p \sim C_v$ and be use only at low temperature because the thermal expansion of $MgSiN_2$ is not continuing linear [18, 20] at high temperature."

Rewritten as :

"At temperature below 300 K, the resulting C_v are comparable to C_p from [17] and [18] within 0.1 J/(mol K), while at temperature above 300 K, C_p from [17], [18] and [19] are larger. This can be explained by considering the equation of $C_p - C_v = 9V_m T\alpha^2 / \beta_T$ where α, β_T, and V_m are linear thermal expansion coefficient, isothermal compressibility and molar volume respectively. At temperature below 300 K, the term $9V_m T\alpha^2 / \beta_T$ can be neglected [18, 20]."

Yours sincerely,

Chaiyawat Kaewmeechai, Yongyut Laosiritaworn and Atchara Punya Jaroenjittichai