Upon attempting to improve energy conversion efficiency, attention has been paid to intermetallic materials, especially metal disilicides, for next-generation gas turbine engine due to their remarkable mechanical properties, oxidation resistance, and stability at elevated temperature. Unlike metals or alloys, it exhibits variety of crystal structures and interatomic bonding deviates from the average between metallic bonding and covalent bonding [1]. Their unique bonding, on the other hand, bring about difficulties to obtain systematic understanding of mechanism behind the properties.

Coefficient of thermal expansion (CTE) of crystal lattice is one of the most important properties since difference in CTEs between a silicide blade and a neighboring layer causes fracture or spallation of the upper layer. According to the equation that defines Grüneisen parameter, $\gamma$, CTE, $\alpha$, is given by $\alpha = \gamma C_v\rho K_T$ where $C_v$, $\rho$, and $K_T$ are heat capacity at constant volume, density, and bulk modulus, respectively [2]. While origins of other three properties are clear, the way to control Grüneisen parameter is as difficult as to control CTE itself according to the equation and thus dependence of CTE on materials is often attributed only to Grüneisen parameter with factors that determine CTE remain unclear. Besides, although impurity is often added to adjust lattice matching between phases to control microstructure, its impact on CTE remains unclear, disabling to predict the lattice matching at elevated temperature. In order to enable data-driven materials design with better CTE and to optimize CTE without deteriorating other properties for practical application at elevated temperature, candidates of factors that have influence on CTE needs to be determined.

In this study, systematic calculations of CTEs of metal silicides including disilicides have been carried out by \textit{ab initio} lattice dynamics within quasi-harmonic approximation using VASP and phonopy codes [3], [4] to obtain accurate forces between atoms and phonon states, respectively. Sufficiently good numerical accuracy was imposed in the calculations to obtain accurate free energy as functions of temperature and volume. Through determining equilibrium lattice constants as a function of temperature, CTE was derived as a function of temperature for a given crystal structure and chemical composition.

Figure 1 shows crystal structures of MSi$_2$ and M$_2$Si where M denotes metallic
Both of them have cubic fluorite crystal structures with M and Si occupying the opposite sites. The ratio of M to Si are different between the two, resulting in different numbers of M-M and Si-Si bondings. It brings about the difference in coordination numbers of M and Si, with 8 Si around M in MSi₂ and 4 Si around M in M₂Si. Figure 1 compares CTEs of MSi₂ and M₂Si for selected M. These results show that there is negative correlation between bulk modulus and the CTEs, and this trend follows the aforementioned equation. It also shows that CTEs of MSi₂ is greater than that of M₂Si in general. These suggest that the CTEs in silicides decreases with increasing ratio of M-M bonding to Si-Si bonding. However, additional calculations of CTEs for pure metals and Si show that the CTEs of Si is smaller than that of metals, indicating that the CTEs of silicides are not determined simply by the balance between bondings species in a unitcell. In addition, it is revealed that the magnitude of influence of net charge on the CTEs is different between the two classes of silicides, indicating that interatomic bonding specific to silicides needs to be further analyzed for optimizing CTEs of disilicides for practical use.

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