Abstract Submitted
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Enthalpy of molecular solids beyond the harmonic approximation: application to hydrogen storage[1] NIKOLAI ZARKEVICH, D.D. JOHNSON, Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, 1304 W. Green St. Urbana, IL 68101. — With low potential energy barriers, the harmonic approximation for phonon modes can be invalid [1]. Molecular solids are composed of strongly-bonded molecules held together by relatively weak intermolecular forces. As intermolecular interactions are usually not harmonic, a new theoretical approach is needed to obtain enthalpies of molecular solids at finite temperature. We develop such a theory for molecular solids and liquids, and apply it to obtain enthalpy differences between various phases from the first principles. We also calculate Gibbs free energy, and show that a phase diagram (e.g., a van't Hoff plot) can be constructed as a graphical solution of the Gibbs equation. To exemplify important applications, we consider materials and reactions for the high-capacity hydrogen storage. [1] Phys.Rev.Lett.97, 119601 (2006).

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