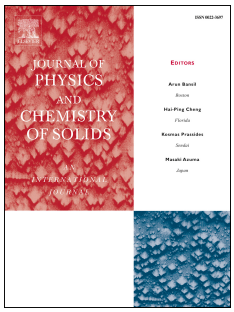
Accepted Manuscript



Lattice dynamical investigation of the Raman and infrared wave numbers and heat capacity properties of the pyrochlores $R_2Zr_2O_7$ (R = La, Nd, Sm, Eu)

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PII: S0022-3697(17)32234-5

DOI: 10.1016/j.jpcs.2018.01.001

Reference: PCS 8365

To appear in: *Journal of Physics and Chemistry of Solids*

Received Date: 22 November 2017

Revised Date: 29 December 2017

Accepted Date: 1 January 2018

Please cite this article as: S. Nandi, Y.M. Jana, H.C. Gupta, Lattice dynamical investigation of the Raman and infrared wave numbers and heat capacity properties of the pyrochlores $R_2Zr_2O_7$ (R = La, Nd, Sm, Eu), *Journal of Physics and Chemistry of Solids* (2018), doi: 10.1016/j.jpcs.2018.01.001.

Lattice dynamical investigation of the Raman and infrared wave numbers and heat capacity properties of the pyrochlores $R_2Zr_2O_7$ (R = La, Nd, Sm, Eu)

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ABSTRACT

A short-range electrostatic forcefield model has been applied for the first time to investigate the Raman and infrared wave numbers in pyrochlore zirconates $R_2Zr_2O_7$ (R^{3+} = La, Nd, Sm, Eu). The calculations of phonons involve five stretching and four bending force constants in the Wilson GF matrix method. The calculated phonon wave numbers are in reasonable agreement with the observed spectra in infrared and Raman excitation zones for all of these isomorphous compounds. The contributions of force constants to each mode show a similar trend of variation for all of these compounds. Furthermore, to validate the established forcefield model, we calculated the standard thermodynamic functions, e.g., molar heat capacity, entropy and enthalpy, and compared the results with the previous experimental data for each compound. Using the derived wave numbers for the acoustic and optical modes, the total phonon contribution to the heat capacity was calculated for all these zirconate compounds. The Schottky heat capacity contributions were also calculated for the magnetic compounds, $Nd_2Zr_2O_7$, $Sm_2Zr_2O_7$ and $Eu_2Zr_2O_7$, taking account of crystal-field level schemes of the lanthanide ions. The derived total heat capacity and the integrated values of molar entropy and molar enthalpy showed satisfactory correlations at low temperatures with the experimental results available in the literature for these compounds. At higher temperatures, the discrepancies may be caused by the anharmonic effects of vibrations, phonon dispersion, distribution of phonon density of states, etc.

Keywords: Pyrochlores; Raman and infrared wave numbers; Lattice dynamics; Heat capacity

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