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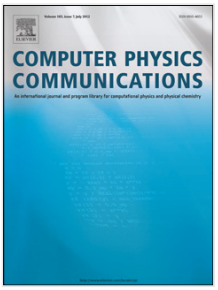

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A systematic fitting procedure for accurate force field models to reproduce ab initio phonon spectra of nanostructures

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Abstract: A fitting procedure is presented to use a valence force field model to generate the phonon spectrum of large nanostructures. This approach uses a relatively large number of parameters (~50) in order to generate the accurate ab initio phonon spectrum. Since the emphasis is in the accuracy rather than the transferability, it can only be used in similar bonding environments. Because of this, a reliable and automatic fitting procedure is essential. We discuss the detailed aspects of the fitting procedure, including the stages of fitting, the type of ab initio values used for the fitting, the weighting factors for different quantities, the number of ab initio data points needed, as well as the uniqueness of the parameters. We found that the parameters cannot be determined uniquely, indicating interdependence of the parameters. Nevertheless, the different parameters resulted from different fits all give accurate phonon spectrum compared to ab initio results. We have used the fitted valence force field model to study the phonon spectra of CdSe nanowires.

Keywords: Classical Force Field Model; Fitting; Phonon Spectrum; Polar Semiconductor; Nanostructure; Surface

Abbreviations: The abbreviation "DCM" is used to refer to "double charge model" in this paper, as discussed in Section 2 and described in Ref. 30.

1 Introduction

Classical force field Hamiltonians, which represent the total energy of a given system in terms of atom-atom pair interactions [1-5], three-body interactions, embedded-charge energies [6,7], and bond-angles energies [8], have been one of the main approaches for material science simulations. This includes the Lennard-Jones pair potential model for ionic systems [2], the Tersoff three-body potential for systems like Si [9], the embedded atom model for metallic systems [10], the valence force field (VFF)