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A semiempirical Hartree-Fock and an analytical tight-binding approach to solid-state calculations for inelastic neutron scattering

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The scattering function, $S(Q,\omega)$, representing the interaction between the incident neutron and the pseudopotential, representing an ensemble of atoms, may be determined in a variety of ways: Via classical force-field approaches, through electronic structure calculations, through analytical approaches and through a combination of one or both of the first two approaches and applying molecular dynamics to study systems under non-equilibrium conditions. Electronic structure calculations quickly become computationally intensive and opaque once more than several hundred atoms are involved in a cluster or unit cell that represents the system under investigation. Another completely different approach is to assume that every solid-state system under study is fundamentally different and solve the specific interaction equation analytically, using for example, a so-called tight-binding methodology.

In the case of neutron spectroscopy many systems investigated are dominated by hydrogen vibrations as the total scattering coefficient, total~82 Barn is approximately an order of magnitude larger than most total scattering coefficients from other elements. Scatter from hydrogen is mainly incoherent and therefore may be considered localised allowing potentially simple molecular models to be employed in the study of hydrogenous materials. Here a well-known Hartree-Fock semi-empirical calculation scheme is presented that is lightning fast for small molecules and adequate for large molecules, clusters, and dynamical studies.
Force constants are obtained by diagonalisation of the mass-weighted Hessian matrix to obtain the vibrational frequency for each mode assuming that each atomic-pair vibrates in a simple-harmonic fashion. The scattering function, $S(Q,\omega)$, is then calculated firstly assuming a one-atom oscillator, then secondly more thoroughly for each atom-pair vibration within the molecule/cluster. A number of benchmark molecules are reported on. Interestingly these rapid calculations give a very good estimation of the scattering function that can be used for initial peak identification and further analysis of the detailed electronic and vibrational structure.

Lastly the first results for an analytical approach are reported here. Rather than calculate an entire system, parts are estimated analytically, and vibrational spectra are very simply calculated.

Topics

Chemistry and Crystallography

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