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Inelastic Neutron Scattering Measurements of Phonon Dispersion Relations in Andalusite and Sillimanite, Al_2SiO_5

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Abstract

This paper reports inelastic neutron scattering (INS) measurements of the phonon dispersion relations of the aluminium silicate minerals andalusite and sillimanite, Al_2SiO_5 . The single crystal INS measurements were undertaken using the Triple-axis-spectrometer at the Dhruva reactor, Trombay for andalusite and at the Oak Ridge National Laboratory, U.S.A for sillimanite. The phonon dispersion relations (upto 50 meV) along various high symmetry directions have been measured and have been analyzed on the basis of lattice dynamics shell model calculations. The calculated structure factors based on the model calculations were used as guides for planning these single crystal measurements and were used to identify regions in reciprocal space with large cross-sections. The calculated structure factors have been very useful in the planning, execution and analysis of the experimental data. The calculated phonon dispersion relations are found to be in good agreement with the measured data.

Introduction

Andalusite, sillimanite and kyanite are the three important polymorphs of the geophysically important aluminium silicate minerals Al_2SiO_5 . Earlier, we had undertaken detailed lattice dynamical studies involving experimental inelastic neutron scattering measurements of the phonon density of states and theoretical shell model calculations of the vibrational and thermodynamic properties of these minerals [1,2]. In this paper, we report inelastic neutron scattering measurements of the phonon dispersion relations of andalusite and sillimanite and their interpretations based on these shell model results. Measurement of the phonon dispersion relation using the inelastic neutron scattering technique and theoretical calculations using interatomic potentials form an ideal combination for a microscopic study of the vibrational properties and thermodynamic properties of minerals [1-3].

The importance of the three aluminium silicate polymorphs andalusite, sillimanite and kyanite is well known in mineralogy [4]. They share a common feature in their crystal structure, i.e. chains of edge-shared AlO_6 octahedra running parallel to the *b*-axis. The transformations amongst the three Al_2SiO_5 polymorphs involve a change in primary coordination of one of the Al atoms which is in tetrahedral coordination in sillimanite, five-coordination in andalusite and in octahedral coordination in kyanite. Both andalusite and sillimanite have orthorhombic structures with space groups *Pnmm* and *Pnma*, respectively and *Z*=4. There are 96 phonon branches at every wavevector corresponding to the 32 atoms/unit cell.

The shell model developed earlier [1,2] was used to compute the one-phonon neutron cross-section for scattering from a single crystal for each phonon mode to identify the regions in reciprocal space with large cross-sections for andalusite and sillimanite. These were used as guides to plan the experiment while from the actual INS measurements, the mode frequencies were ascertained. The calculated phonon dispersion relations are found to be in good agreement with the measured data.

Experimental details

The measurements of the phonon dispersion relations of andalusite were carried out on the Triple-Axis-Spectrometer (TAS) at ambient pressure and room temperature at the Dhruva reactor, Trombay. This TAS uses a Cu (111) monochromator and pyrolytic graphite (PG) (0002) analyser. A PG filter was used to remove the second order contributions. A natural single crystal (with a volume of about 10 cm³) of andalusite was used for the experiments. The measurements were undertaken using the constant Q mode with E_f , the scattered neutron energy fixed at 14.8 meV. The phonon measurements were made in the (100) direction and phonons upto 40 meV energy were measured using incident neutron energies varying from 15 to 55 meV. The measurements of the phonon dispersion relations of sillimanite were undertaken along three high symmetry directions using the triple-axis spectrometer at the Oak Ridge National Laboratory.

Results

The typical neutron groups associated with a few phonons in andalusite obtained from inelastic neutron scattering measurements at the Dhruva reactor, are shown in Fig. 1. The group theoretical representations to which the phonon belong are as indicated by the Greek letters. The numbers in parentheses indicate the coordinates of the wave vector transferred in the neutron scattering process in units of reciprocal lattice vectors. The ordinate is neutron energy in arbitrary units as the neutrons are counted for varying periods of time.

The calculated and measured phonon dispersion relations in andalusite and sillimanite are as shown in Figs. 2 and 3. The agreement between the experimental data and theoretical predictions is quite satisfactory. A comparison of the neutron group intensities associated with each phonon mode with the calculated one-phonon cross-section indicates that the agreement between the two is quite satisfactory. The calculated cross-sections have played a vital role in the planning, execution and analysis of the experimental results and emphasize the role of theoretical models in the analysis of the complex experimental data.

References

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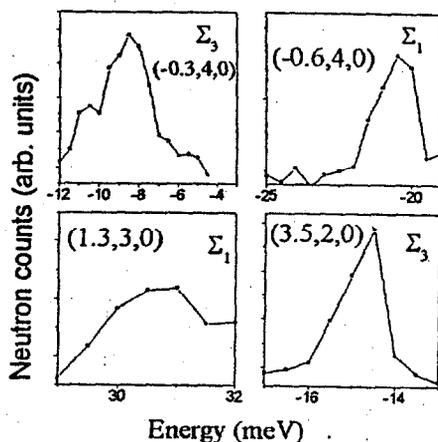


Fig1: Typical phonon groups in andalusite measured using the TAS at Dhruva.

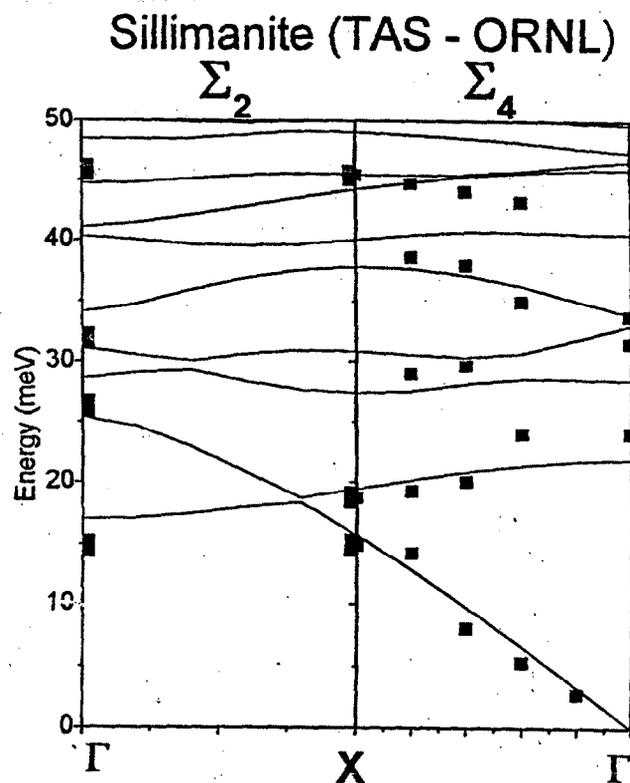
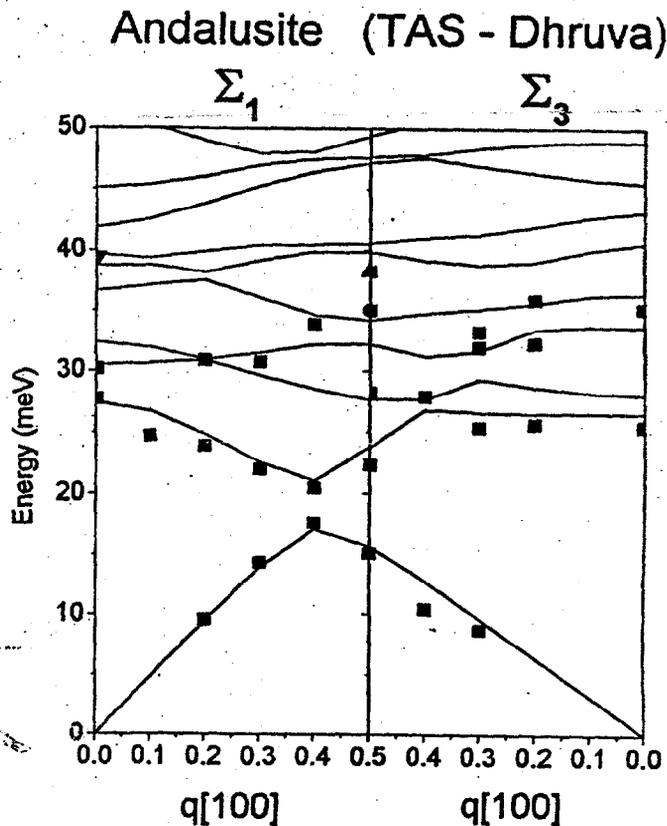


Fig 2: Calculated (solid lines) and measured phonon dispersion relations in andalusite and sillimanite.