Experimental report

02/10/2016

Proposal: 7-01-428 **Council:** 4/2015

Title: Experimental measurement of the phonon DoS for cementite (Fe3C): applications to isotopic fractionation processes in the mantle

Research area: Other...

This proposal is a new proposal

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Samples: Fe3C

Instrument	Requested days	Allocated days	From	To
IN4	6	4	30/10/2015	03/11/2015

Abstract:

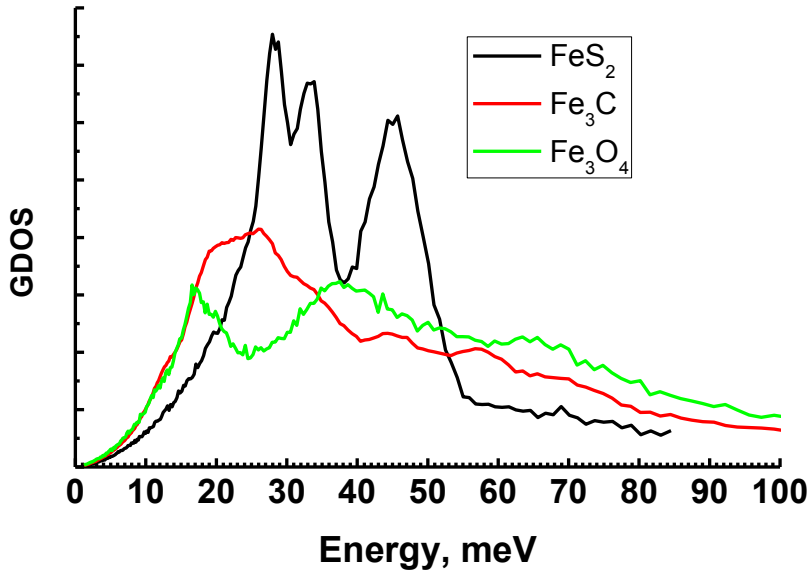
The project is aimed at experimental investigation of phonon density of states (PDOS) in cementite (Fe3C) at different temperatures using Inelastic Neutron Scattering (INS). The data obtained in our work will be used to calculate the carbon equilibrium stable isotope fractionation factor (so-called beta-factor) for Fe3C. The results are crucially important for understanding carbon behaviour and isotopic fractionation in deep Earth. Knowledge of the PDOS allows to calculate the phonon contribution into thermal thermodynamic properties of cementite and to find magnetic contribution into thermodynamic quantities such as heat capacity from comparison with appropriate calorimetry measurements. The PDOS obtained at different temperatures will allow us to estimate the anharmonic correction on thermodynamic properties of cementite (including the isotope factor).

Distribution of stable isotopes between different phases and minerals provides important and often unique information about geochemical processes in the Earth interiors. The reduced isotopic partition function ratio (so-called β-factor) is the main physical quantity in the theory of stable isotope fractionation. In many cases direct experimental measurements of the β-factor are difficult to perform and several theoretical methods to derive its values were suggested. In particular, a novel approach of the β-factor calculations from the PDOS was developed by members of our team. It is based on application of the first-order thermodynamic perturbation theory and allows to express the β-factor in terms of the kinetic energy of the nucleus of interest and appropriate isotopic mass difference. If the partial PDOS of the nucleus is known, the kinetic energy (and the β-factor) can be calculated directly.

The present project was directed to experimental measurements of phonon DOS of several iron compounds important for geosciences: FeS_2, Fe_3C, Fe_3O_4 using Inelastic neutron scattering (INS). The principal novelty of the project is combination of the neutron and synchrotron data. Whereas INS gives phonon DOS of the compound, our previous synchrotron-based measurements performed at ESRF provided partial pDOS for iron. Subtraction of the Fe pDOS allows extraction of light element (C, S, O) contribution and calculation of their isotope fractionation properties.

Experiment.

Phonon DOS of high purity pyrite FeS_2, cementite Fe_3C and magnetite e_3O_4 were measured at IN4 instrument in broad temperature range from 120 to 500 K. Representative curves for 500 K are shown on Fig. 1. Calculation of the isotopic properties from comparison of INS and synchrotron data are currently underway.



Since the studied compounds, except pyrite (FeS_2), are magnetic, it is important to consider possible magnetic contribution to thermodynamic properties (*e.g.*, to heat capacity). At IN4 experimental geometry magnetic excitations can be excluded by omitting data from detector channels corresponding to small angles. Our measurements indicate that lattice contribution dominates over magnetic.