Geophysical Research Abstracts, Vol. 9, 05764, 2007
SRef-ID: 1607-7962/gra/EGU2007-A-05764
© European Geosciences Union 2007

Prediction of stable isotopes fractionation by first-principles methods.

M. Méheut (1), M. Lazzeri (1), E. Balan (1,2), F. Mauri (1)

(1)Institut de Minéralogie et de Physique de la Matière Condensée , (2)Institut de Recherche et Développement - UMR CEREGE (meheut@impmc.jussieu.fr , Fax: +33-1-44273785)

Quantitative modelling of isotope fractionation is a key-problem in geochemistry. Equilibrium isotopic fractionation is a thermodynamic property which is determined by the vibrations (phonons) of the system and, so far, calculations have been mainly based on semi empirical models. In our work, we use ab-initio methods, based on density functional theory, to determine the vibrational properties and, thus, the isotopic fractionation. Ab initio methods are a computational method used to solve the equation of quantum mechanics and to predict, with precision, the properties of a large class of real systems. Standard ab initio total-energy calculations can be used to obtain the phonons at the center of the Brillouin zone of a crystal. More sophisticated techniques, such as the density functional perturbation theory approach (DFPT, Baroni et al. 2001) are required to obtain the complete phonon dispersion. This information is then used to calculate the free energy of a material and hence, its thermodynamic properties. We will show that ab initio DFPT can be used to predict fractionation factors within an accuracy of typically 5% at low temperature on several systems (Meheut et al.). This method is particularly usefull for poorly known systems, or systems for which fractionation factors are hardly experimentally accessible. Selected examples of stable isotope fractionation in the system quartz, kaolinite, water will be presented.

Bibliography

Baroni S. , S. de Gironcoli and A. Dal Corso (2001) Phonons and related crystal properties from density-functional theory Rev. Mod. Phys. 73, pp 515-562

Meheut M., Lazzeri L., Balan E. and Mauri F. (2007) Equilibrium isotopic fractionation in the kaolinite, quartz, water system: predictions form first-principles density-functional theory. Submitted to Geochim. Cosmochim, Acta