



First-principles study of the high-pressure Raman spectrum of FeS₂ pyrite

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Pyrite (FeS₂) is the most abundant of all metal sulphide minerals and is ubiquitous. Its physical and chemical properties at high-pressure and high-temperature have been extensively studied due to the possible role of the Fe-S system in the formation, evolution and composition of the Earth's core and more generally of the terrestrial planets' core (e.g. Venus, Mars). We present the results of plane-wave density functional calculations of the Raman vibrational frequencies of pyrite as a function of pressure. These calculations, performed within the local density approximation (LDA) and employing norm-conserving pseudopotentials, give a very good description of the structural and electronic properties at 0 GPa as well as 50 GPa. The Γ -point phonon frequencies have been calculated using the linear response method as implemented in the CASTEP code (Segall et al., 2002). These are found to be in good agreement with the recent experimental data acquired up to 50 GPa by Kleppe and Jephcoat (2004). Our study enables us to discuss the structural origin of the evolution of Raman modes with pressure and to predict the behaviour of this semiconductor at higher pressures where a metallization may occur by closure of the band gap (Cervantes et al., 2002). According to shock compression data, pyrite is stable up to the pressure of at least 320 GPa (Ahrens and Jeanloz, 1987). The study of the phonon modes provides another way to investigate a possible structural phase transition.

References

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