

LETTERS

Evidence from surface phonons for the (2×1) reconstruction of the $(10\bar{1}4)$ surface of calcite from computer simulation

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ABSTRACT

A new force field for modeling calcium carbonate has been derived that corrects deficiencies of previous models. The model correctly reproduces the structure of the gas phase species, as predicted from ab initio calculations, as well as the bulk structure and properties of calcite and aragonite. With this new model, a (2×1) reconstruction is predicted to occur for the dominant $(10\bar{1}4)$ surface of calcite, involving rotation of half of the surface carbonate anions. This reconstruction matches the results of low energy electron diffraction measured in vacuo and provides the first independent verification of this observation, as well as yielding the atomic detail of the nature of the reconstruction. While there is only a small exothermic energy associated with the formation of the supercell, the presence of an imaginary phonon mode at $(\frac{1}{2}, 0)$ in the two-dimensional Brillouin zone for a single surface cell verifies the existence of the reconstruction.