Insight into zeolite nanogrowth through the analysis of vibrational spectra in terms of internal coordinates

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I. Introduction

Zeolites are microporous inorganic materials, mostly with aluminosilicate components, which exhibit crystal structures containing pores and cages large enough to permit the diffusion of small molecules. Zeolites are indispensable in many industrial applications, e.g. in heterogeneous catalysists, absorption and molecular separation, and ion-exchange. These applications and the extension of their application field in many domains are a motivation for further investigation aiming at an even deeper insight into the behaviour of a zeolite.

Understanding how zeolites nucleate and grow is of fundamental scientific and technological importance. Insight into the molecular mechanisms of structuring of silica can lead to the development of new hierarchical materials promising high potential for optimization of processes in catalysis and molecular separation.

II. Theoretical and computational method

An efficient protocol is presented to identify signals in vibrational spectra of silica oligomers based on theoretical molecular dynamics simulations. The method is based on the projection of the atomic velocity vectors on the tangential directions of the trajectories belonging to a predefined set of internal coordinates. In this way only contributions of atomic motions along these internal coordinates are taken into consideration. The new methodology is applied to the spectra of oligomers and rings, which play an important role in zeolite synthesis. A suitable selection of the relevant internal coordinates makes the protocol very efficient but relies on intuition and theoretical insight. The simulation data necessary to compute vibrational spectra of relevant silica species are obtained through molecular dynamics (MD) using proper force fields. The new methodology - the so-called velocity projection method - makes a detailed analysis of vibrational spectra possible by establishing a one-to-one correspondence between a spectral signal and a proper internal coordinate. It offers valuable perspectives in understanding the elementary steps in silica organization during zeolite nanogrowth. The so-called velocity projection method is generally applicable on data obtained from all types of MD and is a highly valuable alternative to normal mode analysis (NMA) which has its limitations due to the presence of many local minima on the potential energy surface.

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