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Lattice dynamics and Raman spectrum of BaZrO3 single crystals

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BaZrO3 is a perovskite that exhibits the ideal cubic structure with space group Pm-3m from ambient conditions down to 2 K [1]. However, theoretical studies by DFT yield a more complex picture: the cubic phase is predicted to be slightly unstable, with an unstable phonon mode at the R point, which should give rise to an antiferrodistortive (AFD) transition to a low-symmetry phase with octahedral tilts [1]. Furthermore, our recent DFT calculations [2] indicate that the three possible tilted phases resulting from this instability (I4/mcm, Imma, R-3c), are nearly degenerate, and hardly lower in energy than the cubic phase. Experimentally, BaZrO3 exhibits a strong Raman signal whose origin is unclear, since first-order Raman processes are forbidden in the cubic phase by symmetry. It has been tentatively explained by structural distortions at the nanoscale [3], or by 2nd-order Raman scattering.
In this work, we took advantage of the recent synthesis of good quality BaZrO3 single crystals [4] to perform Raman measurements in a wide range of temperatures (from 4 K to 1200 K) and under hydrostatic pressure (up to 20 GPa). We present here a detailed comparison of the experimental data with DFT computations of the phonon modes both in the cubic and in the three distorted phases. We show that the phonon modes computed in the distorted phases do not match our experimental results, ruling out the scenario of nanoscale structural distortions. Moreover, the high energy Raman phonon modes seem well described by the overtone density of states computed by DFT in the cubic structure and we provide a thorough analysis of possible assignments to second-order processes. The atomic displacements related to these assignments are also discussed.

Fig. 1. (a) Experimental Raman spectra at low temperature (4K) in different light polarization configuration showing selection rules and (b) overtone density of states in the cubic phase, for each atomic specie, computed by DFT

[1] Akbarzadeh et al., Phys. Rev. B, 72, 205104 (2005)
[2] Amoroso et al., Phys. Rev. B, 97, 174108 (2018)
[3] Chemarin et al., J. Sol. State. Chem., 149, 298 (2000)
[4] Xin et al., CrystEngComm, 21, 502-512 (2019)

Summary

Topic

1. Dynamics of solids by crystallography

Primary authors: TOULOUSE, Constance (Physics and Material Science Research Unit, University of Luxembourg,); Dr AMOROSO, Danila (Theoretical Materials Physics, Quantum Materials Center (Q-MAT), Complex and Entangled Systems from Atoms to Materials (CESAM), University of Liège); Dr XIN, Cong (Materials Research and Technology Department, Luxembourg Institute of Science and Technology); Dr VEBER, Philippe (Institut Lumière-Matière (ILM) - UMR 5306, Université Claude Bernard Lyon 1); Prof. MAGLIONE, Mario (Institut de Chimie de la Matière Condensée de Bordeaux (ICMCB)); Prof. GHOSEZ, Philippe (Theoretical Materials Physics, Quantum Materials Center (Q-MAT), Complex and Entangled Systems from Atoms to Materials (CESAM), University of Liège); Prof. KREISEL, Jens (Physics and Material Science Research Unit, University of Luxembourg); Dr GUENNOU, Mael (Physics and Material Science Research Unit, University of Luxembourg)

Presenter: TOULOUSE, Constance (Physics and Material Science Research Unit, University of Luxembourg,)

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