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Title:Broadband dielectric response of Ba(Zr,Ti)O-3 ceramics: From incipient via relaxor and diffuse up to classical ferroelectric behavior
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Abstract:Dielectric responses of ceramics from the lead-free isovalent BaZrO3-BaTiO3 (BZT) system were investigated from Hz frequencies up to the infrared in a broad temperature range, 10-700 K. Pure BaZrO3 is a displacive weak-incipient ferroelectric with a simple cubic perovskite structure down to low temperatures, whose dielectric response is fully determined by polar phonons, the lowest-frequency one being of the Last type, unlike BaTiO3, where it is of the Slater type. BaZr0.4Ti0.6O3 is a relaxor ferroelectric whose dielectric anomaly is caused by a strong, overdamped excitation, which softens from the THz down to MHz range according to the Arrhenius law and merges into a constant-loss background at low temperatures. Such a reponse is similar to lead-containing and heterovalent relaxors, but unlike them, the lowest-frequency TO1 polar phonon does not soften appreciably. In the case of BaZr0.2Ti0.8O3 we have investigated the dynamic response connected with a diffuse ferroelectric phase transition. The main dielectric anomaly is again due to similar overdamped THz-microwave excitation, which, however, softens only to the GHz range near the transition temperature and below it merges with a near-constant-loss background. The picture of polar nanoregions in BZT differs from that in heterovalent relaxors, because they are pinned to the regions of the off-centered Ti4+ ions, which are frozen in our temperature range. Therefore we assign the soft relaxations to hopping of the off-centered Ti4+ ions. This is compared with the behavior of pure BaTiO3 ceramics, in which the hopping of the off-centered Ti4+ ions also substantially contributes to the phase transition dynamics. Unlike BaTiO3, the dynamic instability, which is responsible for the diffuse ferroelectric and relaxor behavior in BZT, is fully due to the hopping dynamics of the off-centered Ti4+ ions rather than due to soft phonons, and therefore the diffuse transition is of the order-disorder type.
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