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Molecular Dynamics Studies of Structure, Dynamics and Dielectric Response in a Relaxor Ferroelectric

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Since the first synthesis of the classic $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) material in 1961, relaxor ferroelectrics have been the subject of ongoing experimental and theoretical investigation due to their fundamental scientific interest and their importance in technological applications. We use atomistic molecular dynamics simulations to study relaxor behavior in the 0.75PMN-0.25PT material. Even for a fairly small simulation size of 1000 atoms, the system exhibits frequency dispersion and deviation from the Curie-Weiss law typical of relaxor materials. Analysis of the time autocorrelation functions for individual atoms allows us to identify the Nb atoms with a high concentration of neighboring Ti atoms as the nucleation sites for the relaxor behavior. This is due to the higher coupling between the cation displacements induced by the presence of overbonded oxygen atoms. We also analyze local structure and dynamics in PMN-PT using instantaneous, time-averaged and frequency resolved pair distribution functions (PDF). We find that dynamic Pb and Ti off-centering is present even in the paraelectric phase, below T_b the rate of growth of local Pb off-centering increases, followed by the freezing in of the local displacement direction at an intermediate temperature T_c and a transition to a ferroelectric-like phase at T_f. Thus there is a sequence of four phases, PE, dynamic relaxor, mixed dynamic and frozen phase, and the non-ergodic frozen relaxor phase. We identify the average instantaneous local cation off-centering as the order parameter for the dynamic relaxor phase, and the time-averaged local cation off-centering as the order parameter for the two lower-temperatures relaxor phases. Examination of the dynamic PDF data reveals the shape and the range of correlation between the cation displacements. We also show that the relaxor phase is characterized by the appearance of strong nearest-neighbor correlation between the off-center displacements along the Cartesian directions.