First-principles Study on Atomic Structure and Stability of Ferroelectric Domains in Lithium Niobate

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Lithium niobate ($LiNbO_3$, LN) is widely used as ferroelectric materials with pyroelectric, piezoelectric, electro-optic and photoelastic properties.[1] The performance of LN is intensely affected by domain structures and domain walls in the crystal.[2-3] In LN, only the 180° domain wall is formed along the [0001] axis due to its crystal structure of *R*3*c*. For decades, a lot of studies have been conducted to understand the structure, stability and motion of domain walls in LN.[4-5] However, they are still not fully understood because it is hard to experimentally observe the domain walls at the atomic level. In this study, therefore, first-principles calculations were performed to investigate the atomic structure and stability of the domain walls in LN.

In this study, the 180° domain walls along $(11\bar{2}0)$ and $(1\bar{1}00)$ in LN was investigated. Computational models including the domain walls were produced by arranging a domain inversion region in supercells. The calculations were performed using the Vienna Ab Initio Simulation Package (VASP), and the Projected Augmented Wave (PAW) method with Generalized Gradient Approximation (GGA) was employed.[6] The energy cutoff for the plane waves was 500 eV. Structure optimizations were performed to all the ions until the residual forces on the ions were less than 0.01 eV/Å.

Figure 1 (a) and (b) show the atomic structures of the $(11\bar{2}0)$ and $(1\bar{1}00)$ domain walls, respectively, which are obtained through the structure optimization in the case that the size of the arranged domain inversion region is the half of the supercell. Owing to the periodic boundary condition, the supercells contain two domain walls. It is noted that the atomic structures of two domain walls are geometrically equivalent in the model of the $(11\bar{2}0)$ domain wall whereas those are not in the $(1\bar{1}00)$ domain wall model. It was found that the $(11\bar{2}0)$ domain wall has an excess energy of 170 mJ/m^2 and the $(1\bar{1}00)$ one does 190 mJ/m^2 on average. It can be said that the $(11\bar{2}0)$ domain wall is more stable than the $(1\bar{1}00)$ domain wall. This is in agreement with the experimental report that the $(11\bar{2}0)$ domain wall is preferentially formed in domain inversion.[5]

In order to investigate stability of the domain inversion regions, a size of one domain inversion region was varied in the same supercells, and the atomic structures were optimized. It was found that stability of the domain inversion region depends on the size. It is interesting that the arranged domain inversion regions often disappeared and a single domain was formed by the structure optimizations if the size was smaller than the half of the supercell. This suggests that the ferroelectric domains in LN have a critical size to be stably formed.

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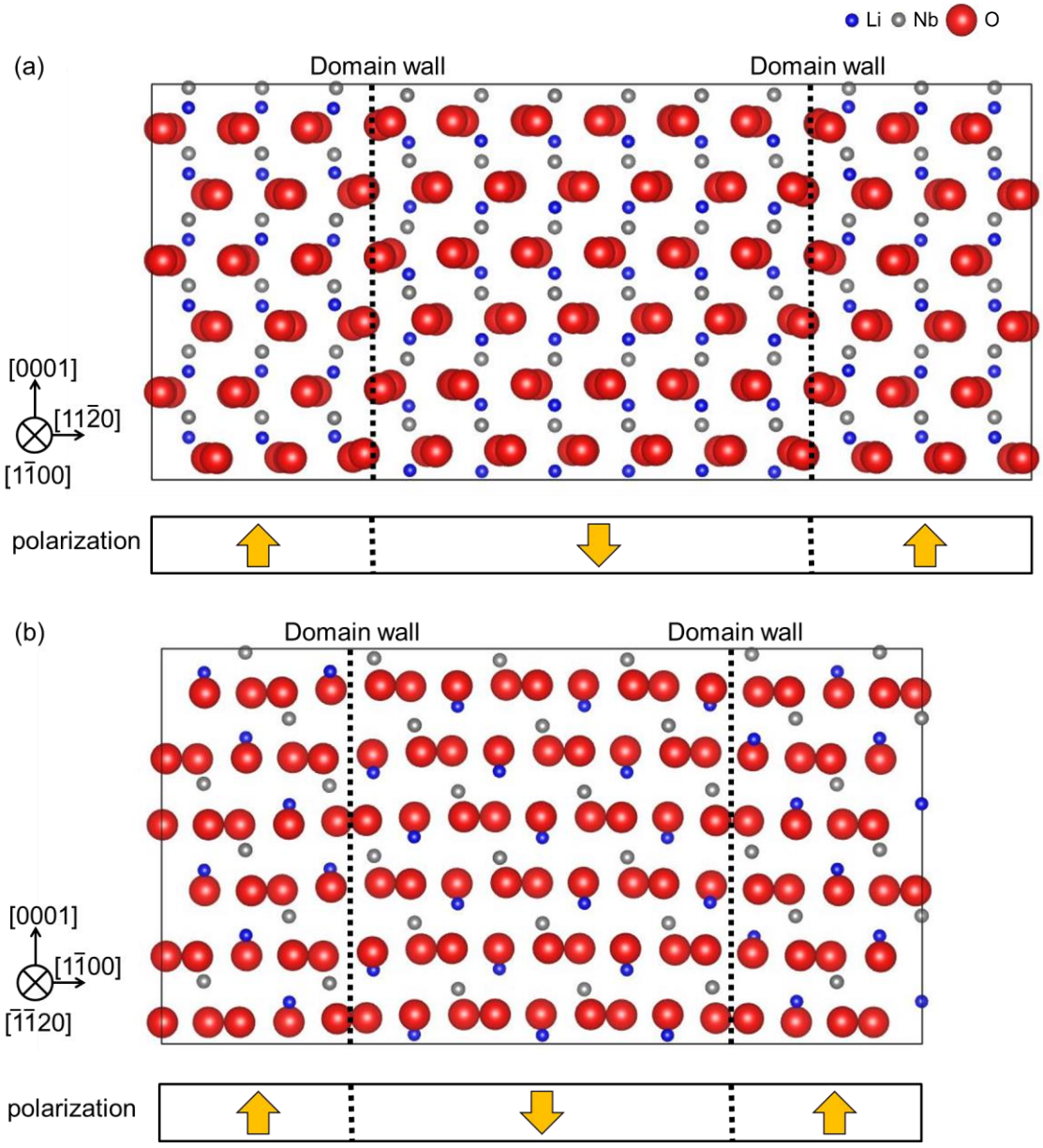

FIG 1. Schematic illustrations of (a) the (11$\bar{2}$0) domain wall, (b) the (1$\bar{1}$00) domain wall. Equivalent two domain walls are present in (a), while different two domain walls are formed in (b). The yellow allows indicate the polarization direction.