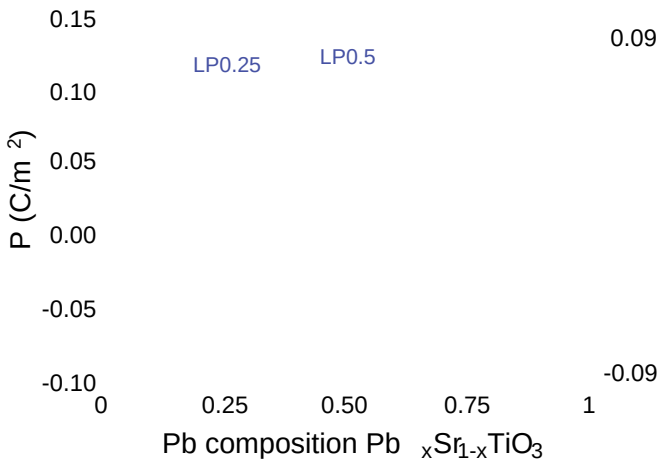

FIG. 1. (Color online) Polarization enhancement of PTO/STO conÞgurations with respect to the concentration weighted average of bulk PTO and STO,$P_{CE}(\ )$, predicted by the cluster expansion approach. The two conÞgurations at breaking points of the convex hull represent the optimal conÞgurations with the largest polarization (LP) at PTO concentrations of 0.25 and 0.5 (named LP0.25 and LP0.5), respectively.

where $J_{ij}, J_{ijk}, \ldots$ represent the effective-cluster interactions (ECIs) for pair, three-body,..., interactions in the chemical system, and the $s_i s_j, s_i s_j s_k, \ldots$ are the multisite cluster functions that form a complete basis set in the conÞguration space. The ECIs can be obtained by Þtting the Þrst-principles calculated results (E) of a set of ordered conÞgurations to Eq. (1). The CE approach has been applied to total energy [9], Curie temperature [11], elastic modulus [12], thermoconductivity [13], and so on. It is applied here to ferroelectric polarization.

Using the data from DFT calculations, an iterative training process was used to Þt the ECIs in Eq. (1) [14]. A good convergence was achieved with only 48 DFT inputs. The obtained $P_{CE}(\ )$ includes 15 pairs, one triplet, three quadruplet, and one quintuplet clusters. The cross-validation score [15], representing the prediction error of $P_{CE}(\ )$, is less than 0.011 C/m^2.

Searching 3 000 000 conÞgurations for identifying the one with the largest polarization, Figure 1 and Fig. S1 [16] show the polarization enhancement $P_{CE}(\ )$ of the $O(3\times 10^6)$ ordered conÞgurations with respect to the concentration weighted average of the bulk PTO and STO. The two breaking points of the convex hull (PTO concentrations of 0.25 and 0.5) represent the conÞgurations of the largest polarization (LP) that exhibit the mostly enhanced values. Their crystal