**Guiding Principles for Enhancing Piezoelectricity in Wurtzite Materials: First-Principles Calculations**

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Recently, piezoelectric wurtzite materials such as ZnO and GaN have received a lot of attention as piezotronics device materials. The wurtzite materials, especially AlN, have another advantage of applicability in high-temperatures such as sensors in automobile engines, because of the good thermodynamic stability of their noncentrosymmetric crystal structures even at high temperatures. However, the piezoelectric constants of wurtzite-type materials are generally much smaller than those of the perovskite-type materials such as Pb(Zr*x*Ti1-*x*)O3 by a few orders. It remains a challenge to explore better piezoelectric wurtzite materials. Among the wurtzite materials, the highest piezoelectricity has been experimentally discovered for Sc*x*Al1-*x*N (about 25 pC/N for *x* ~ 0.5). Novel low-cost materials, which are superior to Sc*x*Al1-*x*N, have not been synthesized yet as there are no clear and general materials-design criteria practically usable for enhancing the piezoelectricity of wurtzite materials.

In this study, we calculate longitudinal piezoelectric constants (*e*33) of more than a dozen binary wurtzite materials, which are listed in the crystal structure databases, by the first-principles methods, and we study relations between the piezoelectric constants and several material parameters using the statistical-learning methods [1]. The results show that the wurtzite materials with high *e*33 generally have small lattice constant ratios (*c*/*a*) almost independent of constituent elements, and approximately expressed as *e*33 ∝*c*/*a* - (*c*/*a*)0 with the ideal lattice constant ratio (*c*/*a*)0. We find that this simple relation also holds for highly-piezoelectric ternary materials such as the calculated *e*33 values of Sc*x*Al1-*x*N [2]. Therefore, this material-design criterion can be applicable to the case in doped ternary materials. Based on the insight above, we have conducted a computational search for highly-piezoelectric wurtzite materials by identifying materials with smaller *c*/*a*. Effects of in-plane strain on piezoelectricity of AlN, element-combination effects in Li*X* (*X* = halogen elements), and element-doping effects into ZnO are examined. The result shows that the piezoelectricity of ZnO can be significantly enhanced by partial substitutions of Zn with Ca. Though the calculated value of *e*33 of Ca*x*Zn1-*x*O is still smaller than that of Sc*x*Al1-*x*N, we expect that Ca*x*Zn1-*x*O is at a definite advantage in materials cost and natural resource in abundance of constituent elements.

1. H. Momida and T. Oguchi, Appl. Phys. Express **11**, 041201 (2018).
2. H. Momida, A. Teshigahara, and T. Oguchi, AIP Adv. **6**, 065006 (2016).