Journal of ELECTRONIC MATERIALS, Vol. 42, No. 6, 2013
DOI: 10.1007/s11664-013-2492-4
© 2013 TMS

First-Principles Study of Electronic, Elastic, and Lattice Vibrational Properties of *Pbnm* Orthorhombic $SrHfO_3$

A. YANGTHAISONG[1,2]

1.—Computational Materials and Device Physics Group, Department of Physics, Faculty of Science, Ubon Ratchathani University, Ubon Ratchathani 34190, Thailand. 2.—e-mail: a.yangthaisong@gmail.com

Through first-principles pseudopotential calculations based on density functional theory, the electronic structure and lattice vibrational properties of *Pbnm* orthorhombic $SrHfO_3$ were investigated in the framework of standard functional approximation and density functional perturbation theory, respectively. The calculated equilibrium lattice constants of *Pbnm* orthorhombic $SrHfO_3$ are in good agreement with available experimental and theoretical results. The results show that *Pbnm* orthorhombic $SrHfO_3$ is an insulator with a direct band gap of 3.9 eV and 4.0 eV within the calculations using local density approximation (LDA) and generalized gradient approximation (GGA), respectively. Use of the screened exchange local density approximation (sX-LDA) as a functional in a successive band calculation has also been performed. The band gap is predicted to be 6.7 eV within sX-LDA, somewhat higher than the gap values of 6.1 ± 0.1 eV and 6.5 eV obtained from recent x-ray photoelectron spectroscopy. The phonon dispersion curves of *Pbnm* orthorhombic $SrHfO_3$ were also calculated. All-positive phonon frequencies were observed in the whole Brillouin zone, indicating stability of the *Pbnm* orthorhombic $SrHfO_3$ structure. In addition, the infrared-active and Raman-active vibrational modes of $SrHfO_3$ were calculated and compared with available theoretical and experimental investigations.

Key words: Orthorhombic $SrHfO_3$, first principles, screened exchange, Raman modes

INTRODUCTION

Perovskite alkaline-metal hafnate compounds such as $BaHfO_3$, $CaHfO_3$, and $SrHfO_3$ have attracted increasing interest for various optical and electronic applications in recent years.[1–4] Among these oxides, $SrHfO_3$ is a promising candidate high-κ dielectric oxide for the next generation of complementary metal–oxide–semiconductor (CMOS) technology.[5,6] The perovskite-structure $SrHfO_3$ has high permittivity of 35 and band gap of 6.1 eV to 6.5 eV,[7,8] providing sufficient valance- and conduction-band offsets to the Si layer for metal–oxide–semiconductor field-effect transistor (MOSFET) operation. In fact, recent investigations suggest that the perovskite compound $SrHfO_3$ can be considered a potential gate dielectric to fabricate *n*- and *p*-MOSFETs with equivalent oxide thickness below 1 nm.[4,5] On the other hand, $SrTiO_3$, a compound with composition and lattice structure similar to $SrHfO_3$, has dielectric constant of 150 to 300 at room temperature while its optical band gap is as low as 3.2 eV.[9] Besides, the conduction-band offset of $SrTiO_3$/Si is about −0.14 eV.[10] Thus, it is unfavorable for use as a high-κ gate dielectric. To this end, composite materials in which both high κ and wide band gap of the parent materials can be tailored have attracted more attention since they could form ideal high-κ dielectrics. Recently, $SrHf_{0.67}Ti_{0.33}O_3$ high-κ films deposited on Si by pulsed laser deposition were reported with dielectric constant of

(Received September 10, 2012; accepted January 10, 2013; published online March 2, 2013)