First-Principles Calculation Software for Dielectric Response Study of High-k Materials

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1. Introduction

The CMOS transistor has a great advantage that its performance can be improved by reducing its size. However, ironically, we are going to have a serious problem, because of the CMOS size reduction itself. It is the gate leakage current problem that would be serious in CMOSs of the next generation. To overcome this problem, use of high-k materials as a CMOS gate insulator has been proposed. But at present, no high-k material in practical use has been reported yet. This is due to that the development of high-k materials by means of the experimental try and error approach is difficult. A theoretical approach seems to be rather necessary.

We have developed new first-principles calculation programs, which enable to investigate the dielectric response of high-k materials at the atomistic level. The new programs can calculate the dielectric function of materials that originates from both the electronic and lattice vibration of the materials. The programs are especially suitable for study of high-k materials, which have the significant contribution of lattice vibration to the material dielectric constant [1].

2. Feature of our programs

Our program consists of two major parts (Fig.1): Epsilon calculating electronic dielectric constant $\varepsilon_{elec}$, and Berry-Phonon calculating lattice vibrational dielectric constant $\varepsilon_{vib}$. Both Epsilon and Berry-Phonon use Phase, a density functional (DFT) pseudopotential band calculation program, and Ekcal, a utility program of Phase. The norm conserving as well ultra soft pseudopotential[2] can be used in these programs.

Epsilon calculates imaginary part of $\varepsilon_{elec}$, $\text{Im}(\varepsilon_{elec})$, using Eq.(1), and obtains real part of $\varepsilon_{elec}$, $\text{Re}(\varepsilon_{elec})$, by making Kramers-Kronig Transformation of $\text{Im}(\varepsilon_{elec})$.

$$\text{Im}(\varepsilon_{elec}(\omega)) = \frac{2\varepsilon^2}{\varepsilon_0} \sum_{k,c,v} \left| \langle \psi_k^v | u | \psi_k^c \rangle \right|^2 \delta(E_{k}^c - E_{k}^v - \hbar \omega)$$  (1)

Here, $\omega$ is photon energy, $u$ is photon polarization vector, $\psi_k^v$ and $\psi_k^c$ are conduction and valence band orbitals with wave vector $k$, respectively, and $E_k^c$ and $E_k^v$ are their orbital energies, respectively.

Berry-phonon performs lattice vibrational analysis and calculates Born effective charge $Z^*$ of atoms in material by making the Berry phase calculation [3]. Berry-Phonon obtains the real part of $\varepsilon_{vib}$, $\text{Re}(\varepsilon_{vib})$, of materials by using Eqs. (2) and (3).

$$\bar{Z}_{i\alpha} = \sum_{i\beta} Z_{i\alpha\beta} \xi_{i\beta} \sqrt{m_i}$$  (2)

$$\varepsilon_{vib,\alpha\beta} = \frac{4\pi}{\Omega} \sum_{\lambda} \bar{Z}_{i\alpha\beta} Z_{i\lambda}^{*} \omega_{\lambda}$$  (3)

$\xi_{i\beta}$ is $\lambda$-th normal mode eigenvector of lattice vibration, $\omega_{\lambda}$ and $Z_{i\lambda}^{*}$ are the frequency and effective charge of the mode, respectively. $m_i$ is mass of $i$-th atom and $\Omega$ is unit cell volume. Total dielectric function $\varepsilon_{total}$ is given as $\varepsilon_{elec} + \varepsilon_{vib}$. Phase, Epsilon, and Berry-Phonon are public domain programs. Phase can be downloaded from our web site[4], and Epsilon and Berry-Phonon are going to be uploaded to the web site in near future.

3. Calculation of HfO$_2$ dielectric function

We calculated the electronic structure of crystal HfO$_2$ in the cubic phase by using Phase and obtained the dielectric function of the crystal by using Epsilon and Berry-Phonon. HfO$_2$ was chosen as a calculation example, because it is one of most promising high-k materials. The crystal structure of cubic HfO$_2$ is shown in Fig.2. The electronic structure of cubic HfO$_2$ was calculated by using Phase and Ekcal. The ultrasoft pseudopotentials for Hf and O atoms used in the calculation were generated by CIAO, our pseudopotential generation software [4]. The non-local DFT method in the generalized gradient approximation (GGA), was used. The calculation used a set of k-points with a mesh parameter of 4x4x4 and a plane wave cut-off of 36 Rydberg. Calculated band structure of the crystal is shown in Fig.3, along with the crystal Brillouin zone.

Calculated dielectric functions of cubic HfO$_2$ are shown in Figs. 4(a) and (b). Fig. 4(a) shows $\varepsilon_{total}$ ($=\varepsilon_{elec} + \varepsilon_{vib}$) of the crystal in radio frequency (RF) region, and Fig. 4(b) shows $\varepsilon_{elec}$. Note that the calculation gives the frequency dispersion of $\varepsilon_{total}$ in the RF region including CMOS operation frequencies. Fig. 6(a) shows that at frequencies lower than 0.01 eV (ca. 2.4 THz), the frequency dispersion is negligible, and $\varepsilon_{total}$ is about 31 ($\varepsilon_{elec}$=7 and $\varepsilon_{vib}$=24). The $\varepsilon_{total}$ value is in the experimental range of $\varepsilon$ of HfO$_2$(0=25-40)[5], and the $\varepsilon_{vib}$ value also is in the experimentally estimated range ($\varepsilon_{vib}$=21-36). The results indicated that HfO$_2$ is a promising
high-k material, showing large enough dielectric responses and small dielectric dispersion at CMOS operation frequencies.

4. Conclusions

We have developed new first-principle programs calculating dielectric response of insulator material. Our new programs are concluded to be effective for studying the dielectric response of high-k materials.

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References


Fig. 1 Our calculation system: Phase+Ekcal, Epsilon, and Berry-Phonon work in trinity and calculate dielectric function of material. Z means effective charge (see text).

Fig. 2 Crystal structure of cubic HfO₂.

Fig. 3 Brillouin zone (a) and calculated electronic band structure of cubic HfO₂ (b).

Fig. 4 Calculated dielectric function of cubic HfO₂: (a) total dielectric function ε_{total}; (b) electronic dielectric function ε_{elec}.