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Effect of Temperature on Dielectric Constant of Silicon Using FHI-aims

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Abstract

The phonopy program package and its FHI-aims interface (Phonopy-FHI-aims) code was used to perform the simulation of Silicon (Si) in this work. Parameter optimization, determination of the best exchange functional and k-grid convergence test were carried out for Si. The effect of temperature on the dielectric tensor and dielectric constant of Si was determined using the quasi-harmonic approximation to account for the anharmonic effects in the determination of the lattice expansion. The shifts in the dielectric constant of Silicon due to lattice expansion for a temperature range of 0 to 1000 K were investigated. The real components of the linear dielectric tensor and the dielectric constants decrease with increase in temperature; the changes observed were not significant because there is just a different of 0.4 between dielectric constants at 0 K and 1000 K. Discontinuities in the dielectric constants were observed between the 200 K and 250 K and 450 K and 500 K. The dielectric constant of Si at room temperature (300 K) is 11.1 for [100], [010], and [001] crystallographic directions.

Keywords: Silicon (Si), Dielectric tensor, FHI-aims, Dielectric constant, linear-thermal-expansion

Introduction

Quantum mechanical first-principles calculations in condensed matter physics have greatly expanded when phonon calculations became routine in the last two decades. [1]Quantum mechanical approaches provide a deep understanding of the properties of many-body systems such as mechanical, electrical, and optical properties. Among the available theoretical approaches, the density-functional theory (DFT) has become overwhelmingly popular. Its success greatly relies on the existence of efficient computer numerical codes. In these numerical codes, the input parameters can be adjusted.

The overall principles of DFT are based on Hohenberg–Kohn's theorems [2]. DFT has a strong versatility especially in the description of the ground-state properties of semiconductors and metals. An increase in computing power has afforded further capabilities in the system's size that DFT methods can handle. DFT calculations for solids provide not only electronic structure information but also give energy and stress to the system as well as the force on each atom.

Minimization of residual forces and optimization of stress tensors give rise to equilibrium crystal structures. When an atom in a crystal is displaced from its equilibrium position, the forces on all atoms in the crystal increase. Analysis of the forces associated with a systematic set of displacements provides a series of phonon frequencies[1]. Silicon is a chemical element with the symbol Si and atomic number 14, a hard and brittle crystalline solid with a blue-gray metallic luster, it is a tetravalent metalloid. It is a member of group 14 in the periodic table. Silicon is a solid at room temperature, with a melting point of 1,414°C [3]. . Like germanium, silicon is rather strong, very brittle, and prone to chipping. Silicon, like carbon and germanium, crystallizes in a diamond cubic crystal structure with lattice constant of 5.4307 Å. The popularity of silicon increased with the advent of the integrated circuit (IC). ICs are commonly built on a substrate and interconnected to perform desired functions. In most cases, silicon is chosen as the basic semiconductor substrate because of its many technological advantages. As of 2016, about 88% of the ICs in the market are metal-oxide (SiO_2)-semiconductor-based [4]. Silicon substrates are widely used for the optical study of transparent materials such as oxides, fluorides, etc., because of the large difference between the indices of refraction of the film and the substrate. [5]. At 300 K, silicon has a dielectric constant of 11.9[6].

The dielectric tensor ϵ_r consists of a real part that represents the storage and an imaginary part that represents the loss[7]. The dielectric constant is obtained from the response of the material to an external electric field; it depends on the frequency of the applied electric field and is described by a tensor. The dielectric constant also called relative permittivity is the value of the real part of the dielectric tensor at frequency equals zero[8] i.e. $Re[\epsilon_r(\omega = 0)]$. Dielectric constant determines the magnitude of the coulomb interaction between electron-hole pairs and charge carriers as well as any fixed ionic charges in the lattice; high dielectric constants are required for high efficiency solar cells. The permittivity expresses the ability of a material to polarize in response to an applied field. It is the ratio of the permittivity of the dielectric to the permittivity of a vacuum. Physically it means the greater the polarization developed by material in an applied field of given strength, the greater the dielectric constant will be. Both dielectrics with a low and

high dielectric constant are essential in electronic industries. A low dielectric constant is required basically as insulators. They are known as passivation materials [9].

Using the simple Debye model, Jean-Mistral et al. reported that the dielectric constant falls as the temperature increases at a low frequency of 0.1 Hz[10]. However, Sommer-Larsen et al. showed that the dielectric constant increases as a function of temperature up to −20 °C and then decreases to a minimum value at 40 °C at 0.01 Hz[11]. For compounds with lower dielectric constants, dielectric constant increases with increasing temperature, whereas for compounds with high dielectric constant it decreases with increasing temperature[12]. There are several discontinuities in the dielectric constant as temperature changes. Dielectric constant changes suddenly at points where lattice structure and composition differs, these changes depend on the exact two points involved. This is due to structural change when there is phase change and, the dielectric constant is strongly dependent on the structure[13]
In an ideal harmonic system, which is fully determined by the dynamical matrix $\boldsymbol{D}(\boldsymbol{q})$, its Hamiltonian does not depend on the volume, this implies that the harmonic Hamiltonian is independent of the lattice parameters, and as a consequence of this, the lattice expansion coefficient $\alpha(T)$vanishes [14].

$$\alpha(T) = \frac{1}{a}\left(\frac{\partial a}{\partial T}\right)_P \qquad (1)$$

To determine the temperature dependence of the dielectric tensor, there is a need to determine the lattice expansion. The quasi-harmonic approximation is used to account for the anharmonic effects in the determination of the lattice expansion[15]. The usage of the quasi-harmonic approximation requires the determination of how the phonons, i.e., the vibrational band structures and the associated free energies, change with the volume[16]. In this work, the effect of temperature on dielectric tensor ϵ_r and dielectric constant ϵ of diamond phase Silicon was studied using phonopy-FHI-aims program package.

Methodology

In this work ab-initio calculation in the framework of density functional theory, as implemented in the FHI-aims package was performed[17], FHI-aims requires exactly two input files; control.in and geometry.in located in the same directory from which the FHI-aims binary is invoked. To start FHI-aims, no further input should be needed [18]. The generalized gradient approximation GGA with the blyp parameterization was employed for the evaluation of the exchange-correlation energy. The following FHI-aims configuration parameters; *occupation type (Gaussian or Fermi), charge mix param, initial moment, and n max pulay* were optimized; the optimization process involves the determination of the value of each configuration parameters that minimize the energy of the structure.

In the process of determining how the associated free energies change with the volume of materials; the optimal lattice constant of the materials were determined by finding the minimum of the total energy $E_{DFT}(V)$ and $F^{ha}(T,V)$ by using Birch-Murnaghan's equation of state[19]. Though, in the canonical ensemble, the relevant thermodynamic potential that needs to be minimized is the free energy $F(T,V)$ which is given by:

$$F(T,V) = E_{DFT}(V) + F^{ha}(T,V) \qquad (2)$$

To account for the volume dependence of $F(T,V)$, the total energy, $E_{DFT}(V)$ and vibrational free energy, $F^{ha}(T,V)$ were calculated for a series of lattice constants. Equation (2) was then evaluated and minimized using Birch-Murnaghan's equation of states, the phonopy program package and its FHI-aims interface (phonopy-FHI-aims) is used. A FHI-aims python script (*Compute_ZPE_and_lattice_expansion.py*)[16] is used to perform the above procedure, this script requires the following inputs; equilibrium lattice constant of elemental Si (obtained from FHI-aims calculations), the temperature range (0 to 1000 *K*), and Si geometry information. The script gives two output files; ***T_a0_alpha*** containing temperature, the lattice constant and the lattice expansion coefficient, and ***ZPE*** containing the equilibrium lattice constant computed with and without zero-point energy (ZPE).

To obtain the linear dielectric tensor in FHI-aims the tag *compute dielectric* was added in control.in the file. It calculates and outputs the component of the imaginary and real part of the inter-band and intra-band contribution to the linear dielectric tensor. To obtain the linear dielectric tensor for Si for different temperatures; a second FHI-aims python script (*Compute_dielectric_tensor_at_different_volumes.py*) is used. The output file ***T_a0_alpha*** from the previous calculation is used as the input file for the second FHI-aims python script. The linear dielectric tensor was determined in [100], [010], and [001] cubic directions for the temperature range of 0 to 1000 *K* in a step of 50 *K*.

Results and Discussion

The lattice constant determined diamond phase Silicon is given in Table 1 along with reported values. The lattice constants are given in the Angstrom (Å) unit.

Table 1: Lattice Constant in Angstrom (Å)

This work			Reported
From Series of Single point calculations	From phonopy without ZPE	From phonopy with ZPE	
5.4200	5.4486	5.4577	5.4307

The lattice constants obtained from series of single point calculations, phonopy without ZPE and phonopy with ZPE gave values that have a difference of $-0.12, +0.33$ and $+0.50\%$ frespectively with experimental reported values. Thus, the lattice constants obtained from series of single point calculations gave good approximation of lattice constant of diamond phase Silicon. The graph of real components of linear dielectric tensor against frequency in Electron-Volts for cubic directions [100], [010], and [001] are shown in Figures 1, 2 and 3. From the graphs, it can be seen that the real components of the linear dielectric tensor are temperature-dependent

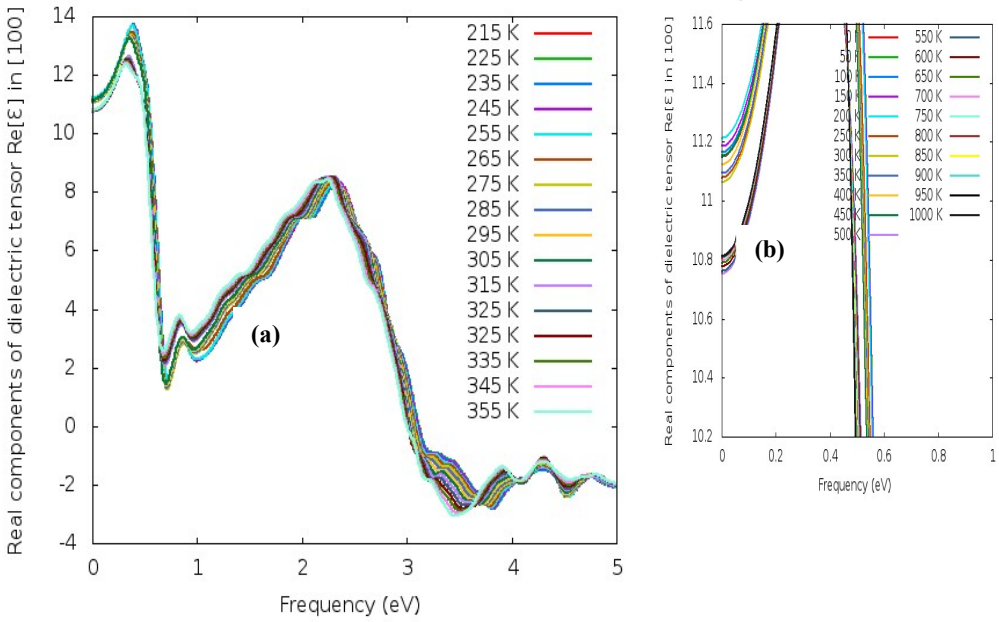

Figure 1: (a) Real components of dielectric tensor against frequency (eV) in [100] direction for the temperature range of 0 to 1000K in a step of 50 K. (b) with emphasis at Re [εx (ω = 0)]

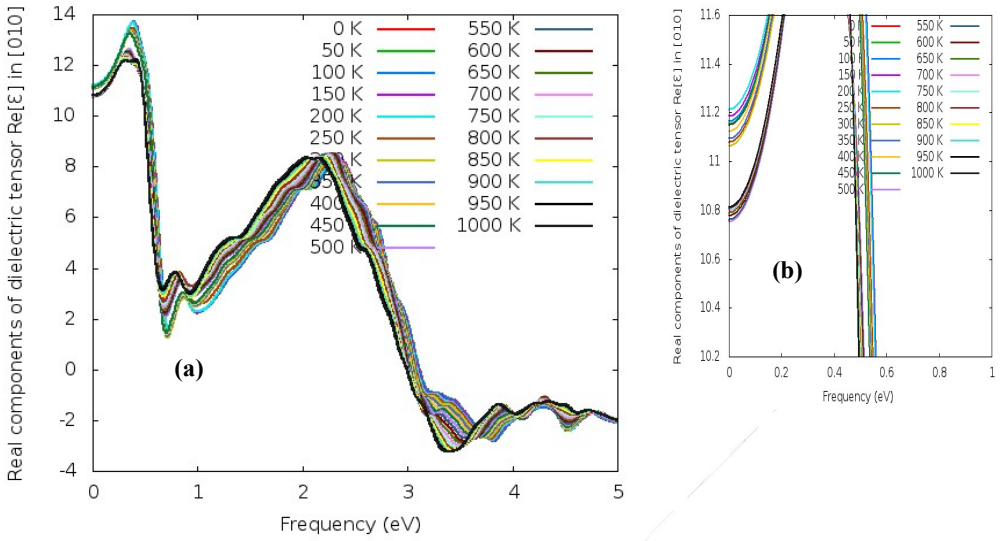

Figure 2: (a) Real components of dielectric tensor against frequency in [010] direction for the temperature range of 0 to 1000 K in a step of 50 K. (b) with emphasis at Re [εy (ω = 0)]

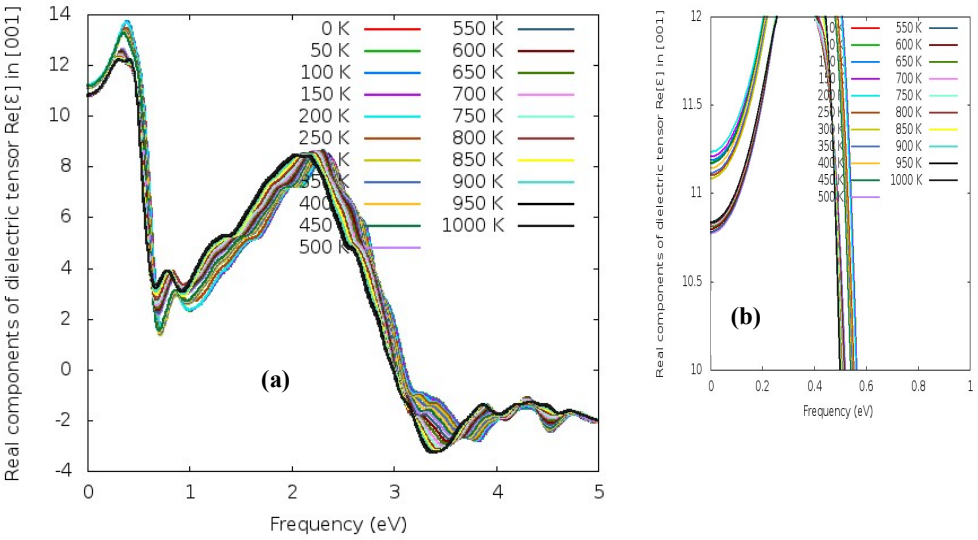

Figure 3: (a) Real components of dielectric tensor against frequency in [001] direction for the temperature range of 0 to 1000K in a step of 50 K. (b) with emphasis at Re [εz (ω = 0)]

The dielectric constants (relative permittivity) which is the value of the real part of the linear dielectric tensor at frequency equals zero (i.e., $Re[\epsilon(\omega = 0)]$) in each of the three directions for the temperature range of 0 to 1000 K were obtained from Figures 1, 2 and 3. The dielectric constants obtained were plotted against temperature (K) in [100], [010] and [001] directions as shown in Figure 4.

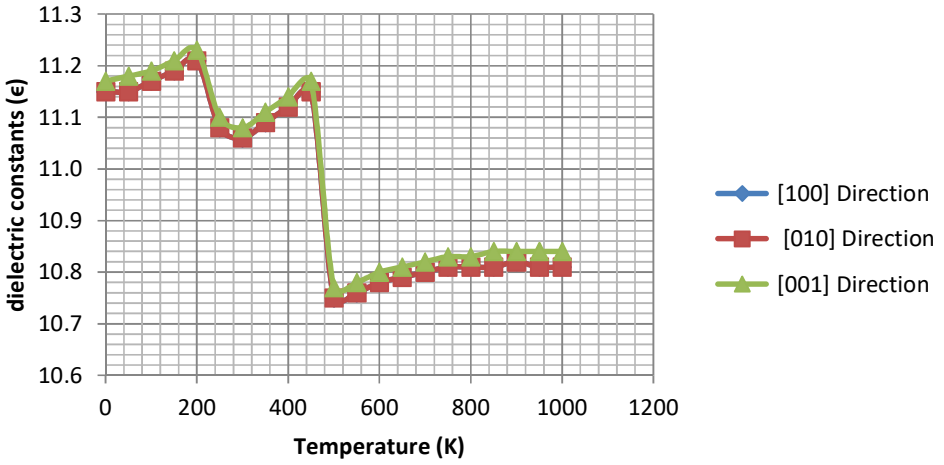

Figure 4: Graph of Dielectric constant against Temperature (K) in [100], [010] and [001] directions

The dielectric constants obtained in all three directions for all the temperatures have very close values, but for [100] and [010] the dielectric constants obtained are the same, this implies that the dielectric property of these materials is isotropy in nature. The highest value of dielectric constants was obtained at 200 K while the lowest value was obtained at 500 K for all three (3) directions. It is observed from figure 4 that there are sharp changes in the dielectric constants between the 200 K and 250 K, and 450 K and 500 K, these sharp changes indicate change in the geometry buildup of the material, as the dielectric constant is strongly dependent on the structure of materials [13]. The dielectric constant of Si at 300 K is 11.1 for all the three directions, this translate to an underestimation of 6.7 %, from the reported value of 11.9 at 300 K [6]. On a general note the real components of the linear dielectric tensor and the dielectric constants decrease with increase in temperature, this is due to the effect of heat on orientational polarization. The changes observed were not significant because there is just a different of 0.4 between dielectric constants at 0 K and 1000 K.

Conclusion

In this work, optimization of geometric construction and configuration parameters were carried out for silicon. Phonopy-FHI-aims was used to investigate the effect of temperature on the linear dielectric tensor of Diamond phase Si. The results obtained were in agreement with already established results and findings, indicating that Phonopy-FHI-aims is a useful tool in determining the effect of temperature on dielectric constant. The similarities of the dielectric constants obtained in the three directions imply that the dielectric property of Si is isotropic. The discontinuities observed indicate a change in the geometry build-up of Si.

Declaration of conflicting interests

The authors declared no potential conflicts of interest.

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