World Academy of Science, Engineering and Technology
International Journal of Materials and Metallurgical Engineering
Vol:9, No:7, 2015

Investigation about Structural and Optical Properties of Bulk and Thin Film of 1H-CaAlSi by Density Functional Method

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Open Science Index, Materials and Metallurgical Engineering Vol:9, No:7, 2015 publications.waset.org/10006016/pdf

***Abstract*—**Optical properties of bulk and thin film of 1H-CaAlSi for two directions (1,0,0) and (0,0,1) were studied. The calculations are carried out by Density Functional Theory (DFT) method using full potential. GGA approximation was used to calculate exchange-correlation energy. The calculations are performed by WIEN2k package. The results showed that the absorption edge is shifted backward 0.82eV in the thin film than the bulk for both directions. The static values of the real part of dielectric function for four cases were obtained. The static values of the refractive index for four cases are calculated too. The reflectivity graphs have shown an intensive difference between the reflectivity of the thin film and the bulk in the ultraviolet region.

***Keywords*—**1H-CaAlSi, absorption, bulk, optical, thin film.

I. Introduction

SINCE the discovery of 'high-T_c' superconductivity hexagonal layered compounds, nH-CaAlSi has attracted broad interest. In order to understand the nature of these compounds experimental and theoretical investigations have been made extensively [1], [2]. In all these compounds the light elements are arranged in honeycomb layers which are intercalated by alkaline earth atoms. The nH-CaAlSi belongs to a class of ternary alloys, formed by Al and Si atoms sitting on Graphene-like sheets, and alkaline earth atoms (Ca, Sr, Ba) intercalated between them [2], [3].

nH-CaAlSi also has attracted considerable interest, because it exhibits an ultrasoft phonon mode and crystalizes with several stacking variants. Thermodynamic experiments on polycrystalline have revealed unexpected differences between CaAlSi and the isoelectronic and isostructural SrAlSi. Ab-initio calculations and later neutron scattering experiments, have shown that an ultrasoft phonon mode exists in CaAlSi. These ab-initio calculations so far, and analyses of the experimental data based on, have assumed that the stacking of Al-Si planes along the c-axis is either uniform or completely disordered [1], [2].

It is worth to mention that n in nH-CaAlSi, refers to different stacking variants of CaAlSi. According to earlier works there are other variants of CaAlSi in addition to 1H, such as 5H and 6H [2], [3].

In this study we calculated and compared different aspects, especially optical properties of bulk and thin film of 1H-CaAlSi using the full potential density functional based method. The space group, (was used here), by which the atoms are intercalated at their positions is P-6m2 [1].

Since the optical properties is mostly attended here, regardless of what method WIEN2k uses to calculate parameters, for more information, the relations of dielectric function, refractive index and reflectivity depending on the real and the imaginary part of dielectric function are as follow:

$$\varepsilon = \varepsilon_1 + i\varepsilon_2 \tag{1}$$

$$n = \frac{(\varepsilon_1+(\varepsilon_1^2+\varepsilon_2^2)^{1/2})^{1/2}}{\sqrt{2}} \tag{2}$$

$$R = \left| \frac{(\varepsilon)^{1/2} - 1}{(\varepsilon)^{1/2} + 1} \right|^2 \tag{3}$$

where n, R, ε, ε_1 and ε_2 are refractive index, reflectivity, dielectric function, real part of dielectric function and imaginary part of dielectric function, respectively [4], [5]. It is worth to mention that dielectric function plays an important role in description of optical properties of materials.

II. Computational Method

The DFT calculations have been performed using the WIEN2k package. The exchange-correlation effects have been described within the generalized gradient approximation (GGA) using the FP-LAPW.

Fig. 1 shows a conventional cell of 1H-CaAlSi [1]. Earlier works showed that crystal structure of 1H-CaAlSi is a closed pack hexagonal structure with a=4.196(3) Å and c= 4.414(4)Å [1].

After constructing the bulk form of 1H-CaAlSi the lattice parameters obtained by the others [1] were optimized. Fig. 2 shows the variation of primitive unit cell energy vs. c/a. In addition, Fig. 2 shows the optimized value of c/a that corresponds to the minimum value of the primitive unit cell energy is obtained by adding 2.2 % to the assumed c/a. The optimized c and a, that are obtained are 4.479 Å and 4.166 Å, respectively, which are in agreement with the experimental ones [1].

Then, the RK_{max} and the number of K points were optimized. The unit cell energy corresponding to the some different either RK_{max} or number of K points are shown in the

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separate pictures in Figs. 3 and 4, respectively. As it is shown, optimized RK_{max} and number of K points are 7 and 160, respectively. In the case of number of K points, in order to be more accurate, we carried out our calculations with 320 K points.

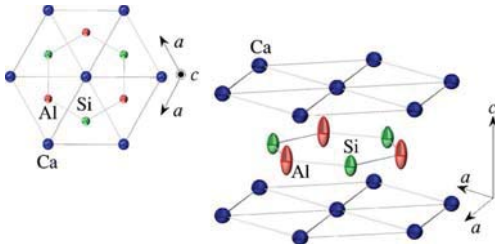

Fig. 1 A conventional unit cell of 1H-CaAlSi [1]

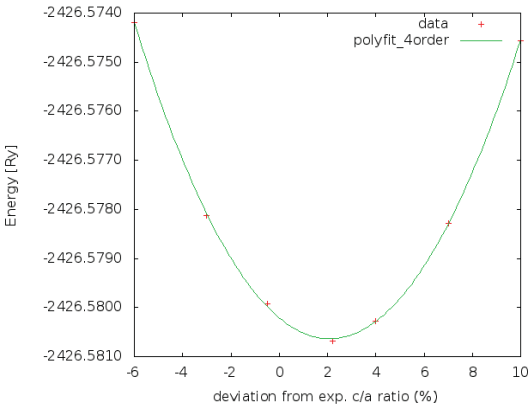

Fig. 2 The horizontal axis denotes that how much percent we have subtracted from or added to supposed c/a. The vertical axis denotes unit cell energy per c/a

The calculations for the thin film of 1H-CaAlSi were proceeded as follow: The supercell used to form the thin film is shown in Fig. 5. This supercell height is the thickness of the thin film as well. One surface of the thin film is ended up to Ca atoms and the other surface is ended up to Al and Si atoms. The RK_{max} was considered the same as that of the bulk and the number of K points was considered 52. Finally, the primitive unit cell of the thin film was relaxed.

Since the number of K points must be big enough to calculate optical properties [6], thus for this purpose the number of K points was considered 10000 for the bulk and 1500 for the thin film.

III. Results and Discussion

A. The Imaginary Part of Dielectric Function

The imaginary part of dielectric function helps effectively to predict absorbing properties of material. The imaginary part of dielectric function versus incident photon energy in (1,0,0) and (0,0,1) directions for bulk and thin film of 1H-CaAlSi were calculated and are shown in Figs. 6 (a) and (b), respectively. As Fig. 6 shows, it is seen that the absorption in both bulk and thin film of 1H-CaAlSi is intensively anisotropic.

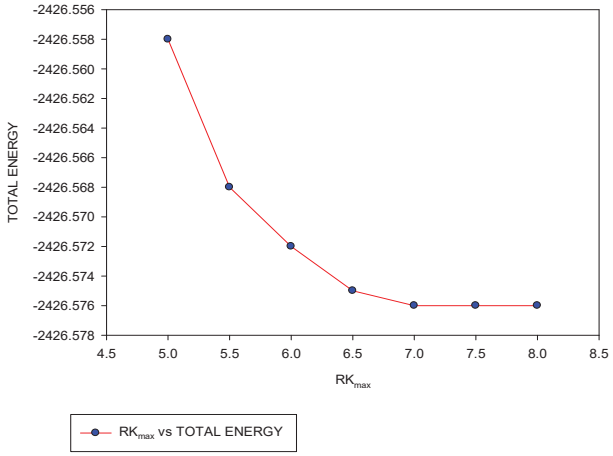

Fig. 3 Variation of the primitive unit cell energy with RK_{max}

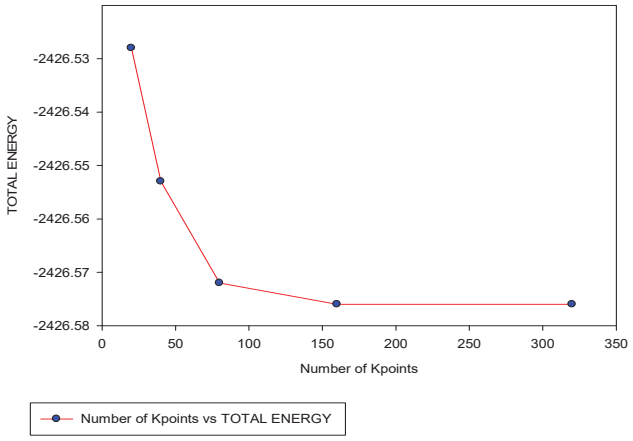

Fig. 4 Variation of the primitive unit cell energy with number of K points

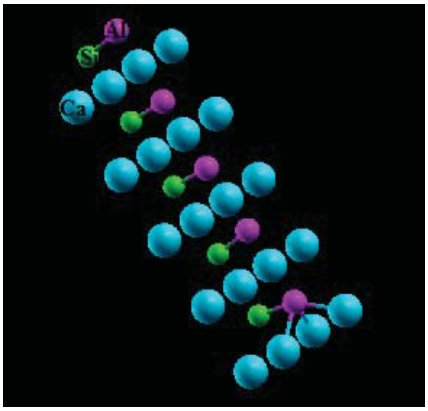

Fig. 5 The supercell used to form the thin film. The supercell height is the thickness of the thin film

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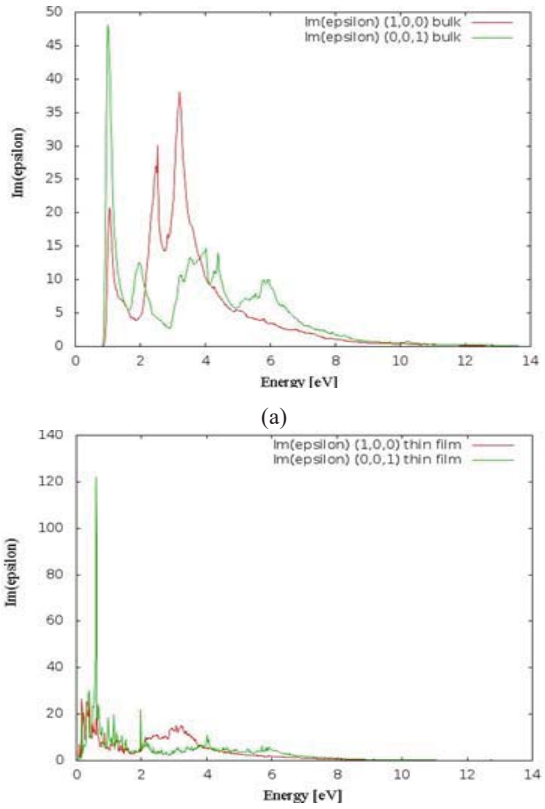

Fig. 6 Imaginary part of dielectric function of 1H-CaAlSi versus incident photon energy for the directions (1,0,0) and (0,0,1) in (a) the bulk and (b) the thin film

In the bulk for both directions the absorption begins at 0.87eV and ends at 13.61eV but in the thin film for both directions the absorption begins at 0.05eV and ends at 13.61eV. The absorption edge is shifted backward 0.82eV in the thin film than the bulk for both directions. Therefore, the noticeable point is that the thin film is excellent absorbent for the photons in the range of (0.05eV, 0.87eV) comparing with the bulk, while the amount of absorption is equal to zero in this range in the bulk for both directions. Thus, for photons in the range of (0.05eV, 0.87eV) of energy, the thin film of 1H-CaAlSi can be used as a shield against this range of waves while the absorption of the bulk in this range is equal to zero (Figs. 7(a) and 7(b)).

In addition, as Fig. 7 shows, the range in which there is absorption, can say that the absorption of bulk of 1H-CaAlSi is more than that of thin film of 1H-CaAlSi for both directions except the point 1.97eV for (0,0,1) direction at which the absorption of the thin film is mainly more than that of the bulk.

An interesting point in the Fig. 7 (b) is the amount of imaginary part of dielectric function which is too high in comparison with the other points. The absorption in the range of 0.61eV to 0.63eV in the thin film for (0,0,1) direction is about 120 while the maximum amount for the others are about 50.

Since the relative maximums in the imaginary part of dielectric function have much importance, therefore the regions with main absorption are represented in Table I.

TABLE I
REGIONS OF ENERGY WITH MAIN ABSORPTION

Region	Bulk x	Bulk z	Thin film x	Thin film z
In the vicinity of: (eV)	1.05, 2.53, 3.21	1.01	----------	0.61
In the range of: (eV)	----------	----------	(2,4), (0,0.87)	----------

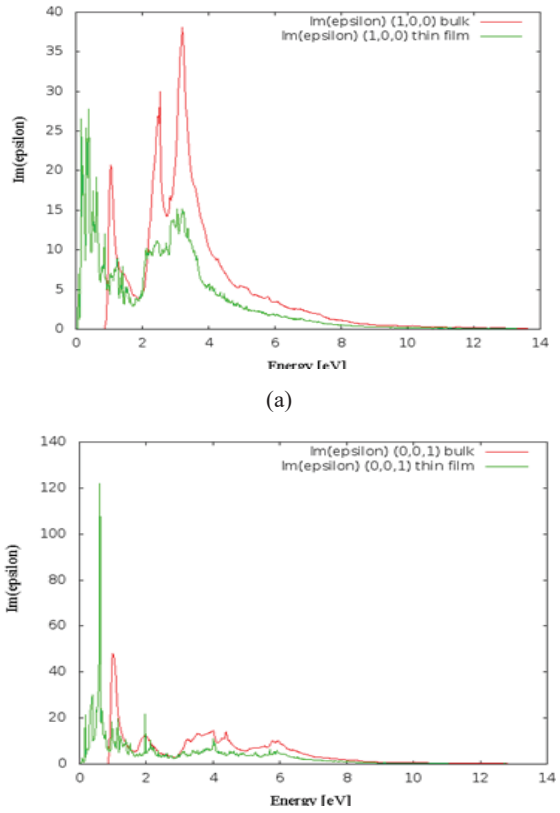

Fig. 7 Imaginary part of dielectric function of 1H-CaAlSi versus incident photon energy in the bulk and the thin film for (a) (1,0,0) direction and (b) (0,0,1) direction

As it is seen in the thin film graphs, there are a lot of oscillations in it which it is probably due to of existence of a lot of states being extremely near to each other in the band structure especially in conduction band. The band structure of thin film of 1H-CaAlSi obtained in present work is shown in Fig. 8.

B. The Real Part of Dielectric Function

Fig. 9 shows the real part of dielectric function versus incident photon energy in the bulk and the thin film of 1H-CaAlSi for directions (1,0,0) and (0,0,1). As it is seen, the real part of the dielectric function is intensively anisotropic either in the bulk or in the thin film.

The static value of ε_1, in the thin film for (1,0,0) direction 25.8592 and for (0,0,1) direction 26.9040 is obtained, while in the bulk for (1,0,0) direction 15.4711 and for (0,0,1) direction

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16.6439 is obtained. As observed, for both directions the proportion of the static value of ε_1 of the thin film to that of the bulk is approximately $\frac{5}{3}$. As Fig. 9 shows, too weak anisotropy of static value of ε_1 (in the both cases) is also obvious.

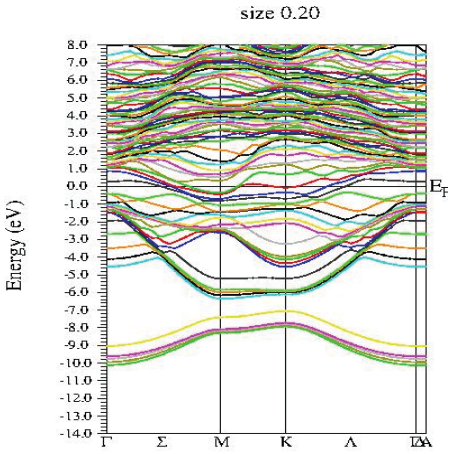
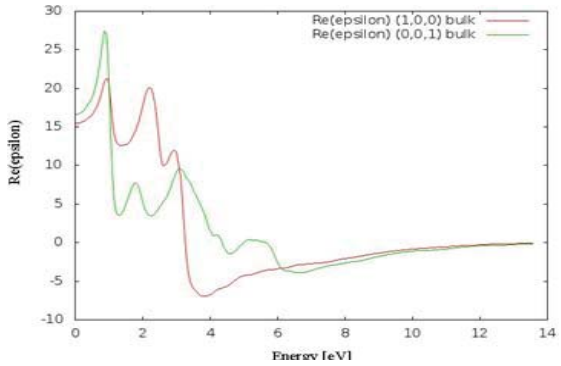
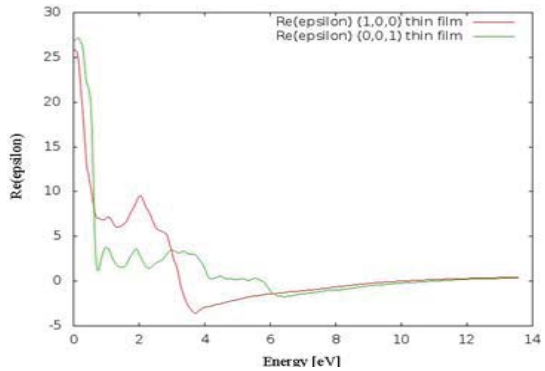


Fig. 8 The band structure of thin film of 1H-CaAlSi, obtained in present work



(a)

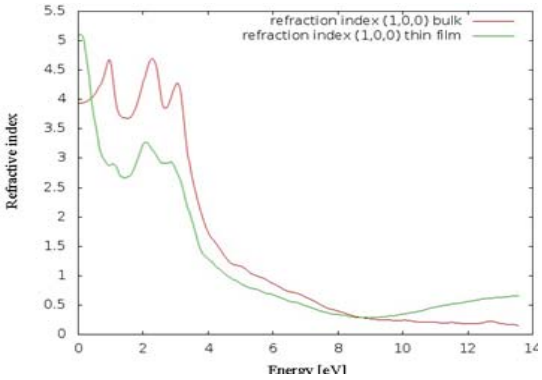


(b)

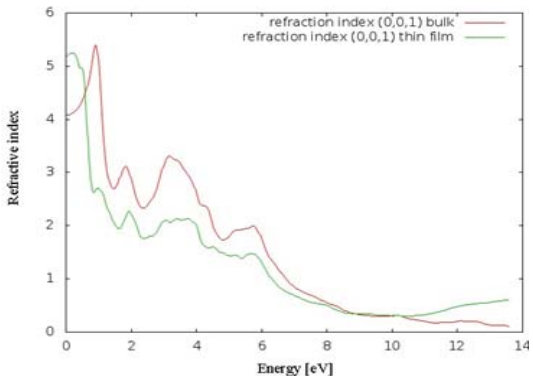
Fig. 9 Real part of dielectric function of 1H-CaAlSi versus incident photon energy for the directions (1,0,0) and (0,0,1) in (a) the bulk and (b) the thin film (Lorentzian broadening with gamma= 0.100000 eV. Im (epsilon) shifted by 0.0000 eV. No intraband contributions added)

C. The Refractive Index

Figs. 10 (a) and (b), compare refractive index of the bulk and the thin film for both (1,0,0) and (0,0,1) directions. The obtained static value of the bulk of 1H-CaAlSi is 3.9 for (1,0,0) direction and 4.1 for (0,0,1) direction and the obtained static value of thin film of 1H-CaAlSi is 5.1 for (1,0,0) direction and 5.2 for (0,0,1) direction.

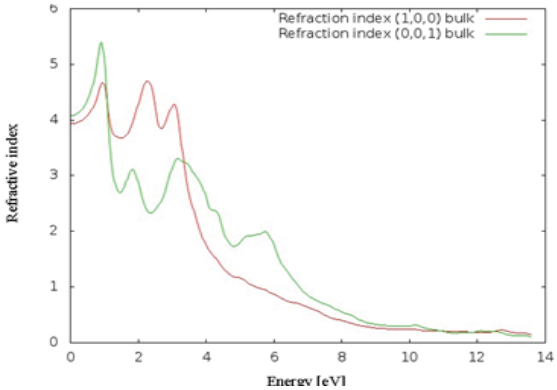


(a)

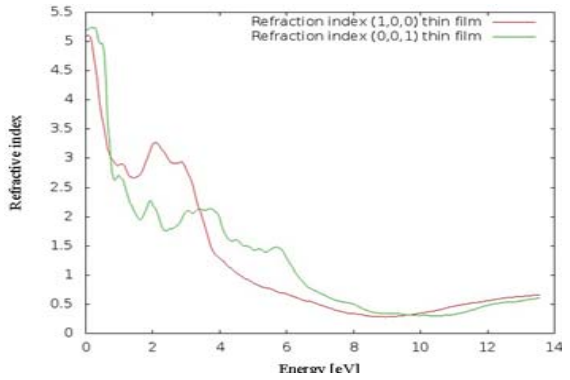


(b)

Fig. 10 Rerfractive index of 1H-CaAlSi versus incident photon energy in the bulk and the thin film for (a) (1,0,0) direction and (b) (0,0,1) direction (Lorentzian broadening with gamma= 0.100000 eV. Im(epsilon) shifted by 0.0000 eV. No intraband contributions added)



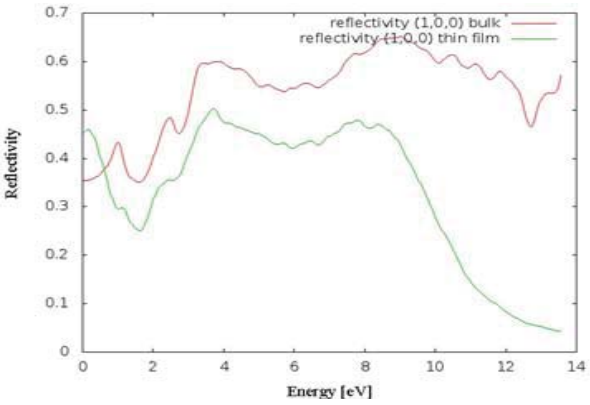
(a)



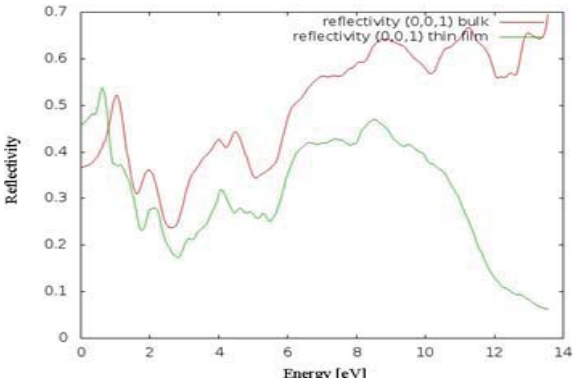
(b)

Fig. 11 Refractive index of 1H-CaAlSi versus incident photon energy for the directions (1,0,0) and (0,0,1) in (a) the bulk and (b) the thin film (Lorentzian broadening with gamma= 0.100000 eV. Im(epsilon) shifted by 0.0000 eV. No intraband contributions added)

Fig. 11 shows anisotropy of 1H-CaAlSi in both the bulk and the thin film. As it is seen the refractive index of 1H-CaAlSi, either in the bulk or in the thin film is totally anisotropic.



(a)



(b)

Fig. 12 Reflectivity of 1H-CaAlSi versus incident photon energy in the bulk and the thin film for (a) (1,0,0) direction and (b) (0,0,1) direction (Lorentzian broadening with gamma= 0.100000 eV. Im(epsilon) shifted by 0.0000 eV. No intraband contributions added)

D.The Reflectivity

Figs. 12 (a) and (b) show photons reflection from the bulk and the thin film for the (1,0,0) and (0,0,1) directions. The intensive difference between the reflectivity of the thin film and that of the bulk in the ultra violet region is observable and in this region the reflectivity of the thin film has been too close to zero. Thus, considering this condition with absorption status (Fig. 6) in the ultra violet region, it can be said that the transition of the photons in the ultra violet region takes place much better in the thin film than the bulk.

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