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Research paper

Mechanical and thermal properties versus effective cubic lattice constant in Cu_2-II-IV-VI_4 quaternary compounds

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Abstract

The present work aims to study the dependence of the bulk modulus B and the Debye temperature θ_D with the effective cubic lattice constant a_{eff} of some Cu_2-II-IV-VI_4 compounds. We are also studied the correlation between the bulk modulus B, the Debye temperature θ_D, the microhardness H and the melting point T_m.
The fits of the data of the bulk modulus B and the Debye temperature θ_D versus the effective cubic lattice constant a_{eff} show that B of Cu_2-II-IV-VI_4 semiconducting materials decreases almost linearly with increase of the effective cubic lattice constant a_{eff}, while that of Debye Temperature θ_D decreases exponentially with a rising of the effective cubic lattice constant a_{eff}. The coefficients of the correlation were found at around -0.78 for the bulk modulus B, and at around -0.94 for the Debye temperature, respectively.
For the bulk modulus B, the best fit was obtained using the following expression: $B = - 596.52\ a_{eff} + 393.4$, where B is expressed in GPa, and a_{eff} in nm, while that of θ_D is: $\theta_D = 165.46 + 3.8 \exp(-57.2\ a_{eff})$, respectively. The average error on the estimation of B was found at around 10%, while that on the estimation of θ_D is only around 4.5%, respectively. Our expressions perhaps used with high accurate to predict the bulk modulus B and the Debye temperature θ_D of other quaternary Cu_2-II-IV-VI_4 semiconducting materials.

Keywords: *Cu_2-II-IV-VI_4 Semiconductors; Least-Squares Fit; Thermal Properties; Effective Cubic Lattice Constant; Microhardness*

1. Introduction

Functional materials play an important role in developing new technologies due to their interesting physical properties [1–12]. Several works [13–20] investigated the correlation between the different properties of functional semiconducting materials. Daoud [13] has established simplified expressions for calculating Debye temperature and melting point of II-VI and III-V compounds; while in other work [14], He investigated the correlation between Debye temperature and lattice thermal conductivity in II-VI and III-V semiconductors.
Because of their applications in the fabrication of low cost solar cells and their large magneto-optical effects which are observed when II elements are paramagnetic atoms, I_2-II-IV-VI_4 (II = Zn, Cd, Hg, Mn, Fe or Co, IV = Si, Ge, Sn or Pb and VI = S, Se or Te) quaternary semiconducting materials, Quintero et al. [16] used X-ray powder diffraction measurements, at 300 K, and differential thermal analysis (DTA) to investigate the thermal properties of several polycrystalline samples of Cu_2-II-IV-S4(Se4) magnetic materials.
Adachi [17] studied the correlation between several physical properties of tetragonal and orthorhombic Cu_2-II-IV-VI_4 compounds and their molecular weight; while very recently, Daoud and co-authors [18] investigated the correlation between the melting point T_m and the microhardness H of tetragonal and orthorhombic Cu_2-II-IV-VI_4 (II = Zn, Cd, Hg; IV = Si, Ge, Sn) compounds. They also use their model to predict the microhardness H of Cu_2ZnSnS_4 (CZTS) compound.
The physics of the photovoltaic effect including the spectral irradiance of solar radiation and the influence of the Earth's atmosphere on it, and several other interesting physical properties of copper zinc tin sulfide-based thin-film solar cells were presented by Ito [19]. Additionally, He explored the electronic and optical properties of the kesterite and stannite phases of Cu_2ZnSnS_4 and $Cu_2ZnSnSe_4$ (CZTSe) by means of first-principles modeling.
Chalapathy et al. [21] elaborate Cu_2ZnSnS_4 thin films by sulfurization of Cu/ZnSn/Cu precursorin sulfur atmosphere. The CZTS films obtained exhibited the Kesterite phase; and under standard AM 1.5 conditions, they found that the solar cell fabricated with the CZTS material showed a conversion efficiency of ~ 4.59% for 0.44 cm^2.
In the present work, we studied firstly the correlation between the bulk modulus B, the Debye temperature θ_D and the effective cubic lattice constant a_{eff} of tetragonal and orthorhombic Cu_2-II-IV-VI_4 (II = Zn, Cd, Hg; IV = Si, Ge, Sn) compounds, then the correlation between the bulk modulus B and the melting point T_m; and the Debye temperature θ_D and the microhardness H of these materials.

2. Theory, results and discussion

At normal temperature and pressure, most of the Cu_2–II–IV–VI_4 semiconducting compounds crystallize in the tetragonal or orthorhombic structures [17], [19]. The unit cell geometry of tetragonal phase was assigned as: $a = b \neq c$ (lattice constants), and $\alpha = \beta = \gamma = 90°$ (lattice angles), while in orthorhombic structure, the unit cell geometry was assigned as: $a \neq b \neq c$, and $\alpha = \beta = \gamma = 90°$ [15]. In the tetragonal phase, the effective cubic lattice constant a_{eff} is calculated, using the lattice parameters a and c as follow: $a_{eff} = (a^2c/2)^{1/3}$ [17].
We compile in Table 1 the effective cubic lattice constant a_{eff} [17], the melting points T_m[17], the microhardness H [17], the bulk modulus B [17], [22 - 24], and the Debye temperature [16], [17], [22], [23] reported for a number of Cu_2-II-IV-VI_4 compounds. It must be noted that the data of the bulk modulus B taken from the Ref.[24] are theoretical, and they are approximately reproduced from the histogram of the figure 9 reported in the work of Hasan et al. [24]

Table 1: Effective Cubic Lattice Constant a_{eff}, Melting Point T_m, Bulk Modulus B, Debye Temperature θ_D, and Microhardness H of Some Cu_2-II-IV-VI_4 Semiconducting Compounds[17],[a] Ref [22],[b] Ref [23], [c] Ref [16], [d] Ref [24], Values with * are Theoretical

Materials	Structure-type	a_{eff}(nm)	T_m(K)	B_m(GPa)	H (GPa)	θ_D(K)
Cu_2ZnSiS_4	Orthorhombic	0.5265	1396	81 [a] *	3.4	490 [a] *
$Cu_2ZnSiSe_4$	Orthorhombic	0.5539	1246	61 [b]*	2.8	282 [b] *
Cu_2ZnGeS_4	Tetragonal	0.5313	1377	72[d]*	3.4	-
Cu_2ZnSnS_4	Tetragonal	0.5428	1377	86	-	302
$Cu_2ZnSnSe_4$	Tetragonal	0.5682	1259	70	-	195[c]
Cu_2CdSiS_4	Orthorhombic	0.5364	1289	64 [d]*	2.5	-
Cu_2CdGeS_4	Orthorhombic	0.5419	1288	70 [d]*	-	267 [c]
$Cu_2CdGeSe_4$	Tetragonal	0.5674	1107	56 [d]*	1.9	198 [c]
Cu_2CdSnS_4	Tetragonal	0.5530	1190	62 [d]*	2.2	-
$Cu_2CdSnSe_4$	Tetragonal	0.5786	1054	34 [d]*	1.5	182 [c]
$Cu_2HgSnSe_4$	Tetragonal	0.5678	985	55 [d]*	1.4	166 [c]
$Cu_2HgGeSe_4$	Tetragonal	0.5787	1027	51 [d]*	1.5	-

We examine the correlation between the bulk modulus B and the effective cubic lattice constant a_{eff} of different Cu_2-II-IV-VI_4 compounds, so the data of B and a_{eff} reported in Table 1are traced in Figure 1. We can observe that B decreases with the increase of a_{eff}. The linear fit was given as follow: $B = -596.52a_{eff} + 393.4$, where B is expressed in GPa, and a_{eff} in nm. The significance of the regression is given as the probability p of the null hypothesis $p < 2.81 \times 10^{-3}$, while the coefficient of the correlation was found at around -0.78. In Table 2, we report the calculated bulk modulus B of different Cu_2-II-IV-VI_4 semiconducting compounds using our model ($B = -596.52a_{eff} + 393.4$). We also calculated the error on B, the average error on the estimation of B was found at around 10%.

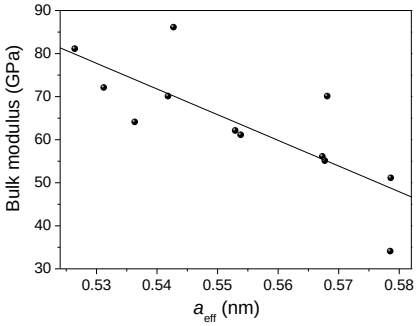

Fig. 1: Bulk Modulus B Versus the Effective Cubic Lattice Constant a_{eff} for Some Cu_2-II-IV-VI_4 Compounds.

Several thermal properties of different materials with different crystallographic structures have been successfully determined from the cohesive energy and elastic constants [25-30]. The bulk modulus B and the melting point T_m data of Cu_2-II-IV-VI_4 compounds listed in Table 1are plotted in Figure 2. We can observe that B increases with the increase of the melting point T_m. The linear fit was given as follows: $B = 0.084\ T_m - 38.12$, where B is expressed in GPa, and T_m in K. The significance of the regression is given as the probability p of the null hypothesis $p < 3.58 \times 10^{-4}$, and a squared correlation coefficient $R^2 = 0.86$.

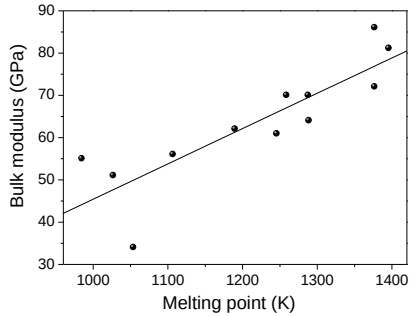

Fig. 2: Bulk Modulus B versus Melting Point T_m for Some Cu_2-II-IV-VI_4 Semiconducting Compounds.

In Table 2, we report the calculated bulk modulus B of Cu_2-II-IV-VI_4 compounds using $B = 0.084T_m - 38.12$ model, along the results obtained from our first model ($B = -596.52a_{eff} + 393.4$). We also calculated the errors on B, the average errors on the estimation of B were found at around 9.75% from our second model ($B = 0.084T_m - 38.12$) and 10% from our first model ($B = -596.52a_{eff} + 393.4$), respectively. Our expressions related B, a_{eff} and T_m can be used to predict the bulk modulus B of other Cu_2-II-IV-VI_4 compounds.

The Debye temperature θ_D and the effective cubic lattice constant a_{eff} data of some Cu_2-II-IV-VI_4 reported in Table 1 are plotted in Figure 3. We can observe clearly that θ_D decreases not linearly with the increase of the effective cubic lattice constant a_{eff}. The best exponential fit with an expression having three adjustable parameters of θ_D (expressed in K) versus a_{eff} (expressed in nm) was given as follows: $\theta_D = 165.46 + 3.8 \exp(-57.2\ a_{eff})$. The significance of the regression is given with a coefficient of the correlation of around -0.94.

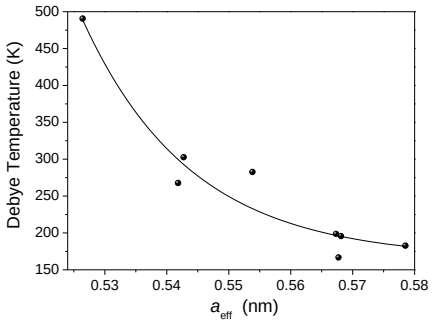

Fig. 3: Debye Temperature θ_D Versus the Effective Cubic Lattice Constant a_{eff} for Some Cu_2-II-IV-VI_4 Compounds.

We also calculated the error on the Debye Temperature θ_D, the average error on θ_D was found at around 4.5%. Our expression related θ_D and a_{eff} can be used to predict the Debye Temperature θ_D of other Cu_2-II-IV-VI_4 semiconducting compounds.
The Debye Temperature θ_D and the microhardness H data listed in Table 1are plotted in Figure 4. We observe that Debye Temperature θ_D rises also not linearly with increase of the microhardness H. The polynomial fit was given as follows: $\theta_D = - 416.5 + 879.8\ H - 434.4\ H^2 + 74.7\ H^3$, where θ_D is expressed in K, and H in GPa. The significance of the regression is given with a coefficient of the correlation of around 0.999, and the probability p of the null hypothesis $p < 0.0244$. Our expression related θ_D and H was used to predict the Debye Temperature θ_D of Cu_2ZnGeS_4, Cu_2CdSiS_4 and Cu_2CdSnS_4 compounds. The results were found: 489 K for Cu_2ZnGeS_4, 235 K for Cu_2CdSiS_4, and 212 K for Cu_2CdSnS_4, respectively. To the best of our knowledge, there are no data available in the literature on θ_D for Cu_2ZnGeS_4, Cu_2CdSiS_4, and Cu_2CdSnS_4 materials.

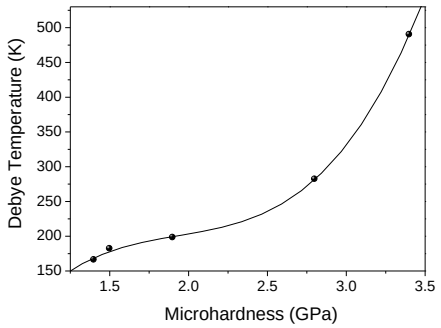

Fig. 4: Debye Temperature θ_D versus the Microhardness H for Some Cu_2-II-IV-VI_4 Semiconducting Compounds.

Table 2: Calculated Bulk Modulus B (GPa) and Debye Temperature θ_D(K) and the Errors on B and on θ_D for Some Cu_2-II-IV-VI_4 Semiconductors

Material	B_{Calc}*	Error (%)*on B	B_{Calc}**	Error (%)on B**	θ_{DCalc} ***	Error (%) on θ_D***	θ_{DCalc}****	Error (%) on θ_D****
Cu_2ZnSiS_4	79	2.06	79	2.29	492	0.38	489	0.17
$Cu_2ZnSiSe_4$	63	3.26	67	9.09	234	17.14	281	0.33
Cu_2ZnGeS_4	76	6.21	78	7.71	414	-	489	-
Cu_2ZnSnS_4	70	19.06	78	9.83	294	2.63	-	-
$Cu_2ZnSnSe_4$	54	22.20	68	3.38	196	0.30	-	-
Cu_2CdSiS_4	73	14.73	70	9.62	351	-	235	-
Cu_2CdGeS_4	70	0.21	70	0.10	301	12.67	-	-
$Cu_2CdGeSe_4$	55	1.90	55	2.02	197	0.51	199	0.66
Cu_2CdSnS_4	64	2.46	62	0.26	237	-	212	-
$Cu_2CdSnSe_4$	48	41.92	50	48.28	182	0.05	178	2.25
$Cu_2HgSnSe_4$	55	0.55	45	18.87	196	18.24	169	1.67
$Cu_2HgGeSe_4$	48	5.50	48	5.59	182	-	178	0.17

* Using $B = - 596.52 a_{eff} + 393.4$, ** Using $B = 0.084 T_m - 38.12$, *** Using $\theta_D = 165.46 + 3.8 \exp(-57.2\ a_{eff})$, **** Using $\theta_D = - 416.5 + 879.8\ H - 434.4\ H^2 + 74.7\ H^3$

3. Conclusion

Using the data reported in the literature, we investigate the dependence between the bulk modulus B, the Debye temperature θ_D and the effective cubic lattice constant a_{eff} of Cu_2-II-IV-VI_4 (II = Zn, Cd, Hg; IV = Si, Ge, Sn) compounds. We found that the bulk modulus B decreases almost linearly with the increase of the effective cubic lattice constant a_{eff} as follow: $B = - 596.52 a_{eff} + 393.4$, where B is in GPa, and a_{eff} in nm. We found also that the bulk modulus B increases almost linearly with the increase of the melting point T_m as follow: $B = 0.084 T_m - 38.12$. The average error on the estimation of B was found at around 10%.
We studied also the dependence between the Debye temperature θ_D and the effective cubic lattice constant a_{eff} of our materials of interest. The Debye temperature θ_D decreases exponentially with the increase of the effective cubic lattice constant a_{eff} as follow: $\theta_D = 165.46 + 3.8 \exp(-57.2\ a_{eff})$. The average errors on the estimation of θ_D was only around 4.5% from $\theta_D = 165.46 + 3.8 \exp(-57.2\ a_{eff})$ model, and around 1% from $\theta_D = - 416.5 + 879.8\ H - 434.4\ H^2 + 74.7\ H^3$ model, respectively.

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