

Nonlinearity, optoelectronic properties, and their correlations for some mixed ternary defect chalcopyrites

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Abstract. The refractive index, optical nonlinearity, lowest energy band gap, and other related parameters of some mixed defect ternary chalcopyrites are calculated using Levine's bond charge model and its modification developed by Samanta et al. for multinary and mixed compounds. The dependence of the band gap energy on the average quantum number, molecular weight, and anion displacement parameter is shown for the first time, which will be very useful for designing various optoelectronic and nonlinear laser devices.

Keywords: defect chalcopyrite, mixed crystal, band gap, quantum number, molecular weight, effective charge.

1. Introduction

Ternary chalcopyrites (II-IV-V₂ and I-III-VI₂) [1–3] and their mixed versions [4, 5] have shown considerable technological potential in a variety of applications, e.g., nonlinear laser devices, optoelectronics, solar cells, high-speed transistor technology because such design parameters as the lowest energy band gap, refractive index, thermal expansion coefficient can be tailored to meet specific requirements. In addition, the wide range of optical energy band gaps and carrier mobilities in ABC₂ ternary semiconductors, their ability to produce various solid solutions and to accommodate dopants has made it possible to fabricate technologically important and useful materials.

The chalcopyrite structures represent the ZnS superstructures and are characterised by the presence of two cation sublattices rather than one, by tetragonal distortion and displacement of the anion from the sphalerite-like position, i.e., they have an ideal tetrahedral structure. Not only the original chalcopyrites (II-IV-V₂ and I-III-VI₂) but also the defect chalcopyrites belonging to groups II-III-VI₄ and I-III-IV-VI₄ (particularly the latter) have a significant potential in various technological applications [6, 7] because of their high nonlinearity, intense luminescence, and high photosensitivity. Having the direct band gap, the Cd and Hg thiogallates are very useful for constructing LEDs and lasers while their residual conduc-

tivity helps to develop memory cells. Such materials as HgGa₂S₄, HgGa₂Se₄, CdGa₂S₄ and their combinations, namely, HgGa₂(Te_xSe_{1-x})₄ [8] can be used in nonlinear devices and tunable filters. Because of the trends aimed at expanding the applications of these crystals, we have studied the samples Hg(Ga_xIn_{1-x})₂Se₄, Hg(Ga_xIn_{1-x})₂Te₄, Cd(Ga_xIn_{1-x})₂Se₄, Cd(Ga_xIn_{1-x})₂Te₄, CdIn₂(Se_xTe_{1-x})₄, Zn_xCd_{1-x}Ga₂S₄, Zn_xHg_{1-x}Ga₂S₄ and evaluated for the first time the linear and nonlinear optical properties, lowest energy band gap, and effective charges by comparing the calculation results with available experimental data. This investigation originated because these materials play a key role in characterising the optical properties of the compound through variation of the cationic/anionic concentration, and the end crystals do not always satisfy the stringent device requirement imposed on the devices based on them. In this connection, we have also been able to find out the dependence of the band gap on such some important physical parameters as the average quantum number, molecular weight, and the anion displacement parameter, which is very important and useful for designing devices and understanding the mechanism controlling such processes.

2. Structural features

The defect group crystallises in a partially ordered chalcopyrite structure with a space group I $\bar{4}2d$ (D_{2d}¹²) that is similar to the structure of the A^IB^{III}C₂^{VI} chalcopyrites. The defect chalcopyrite structure A^{II}B₂^{III}C₄^{VI} can be derived by successive substitution from the sphalerite (Zinc blende). Figure 1 shows the obtained defect structure. Here, the unit cell of such a defect compound (HgGa₂Se₄) contains two formula units and the primitive cell has seven atoms. Such a defect ternary compound (A^{II}B^{III}C^{VI}) have four different cations occupying the lattice vacancy, which makes it possible to derive two possible arrangements; i.e., the A and B lattice can be occupied by both (A^{II}-□) and (B^{III}-B^{III}) and by (A^{II}-B^{III}) and (B^{III}-□) if use is made of the formula

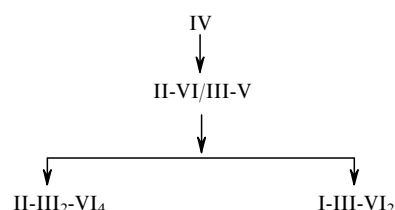


Figure 1. Derivation of defect chalcopyrite structures.

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$A^{II}B_2^{III}\square C_4^{VI}$ where \square denotes a vacancy. This vacancy is an ordered defect and various attempts to fill up the vacancy are unsuccessful, which indicates that the defect is an inherent one. Thus, we may say that the macroscopic properties of ternary defect chalcopyrites are due to the microscopic contribution from the basic structural II-VI, III-VI, III-VI, and \square -VI units connected in an infinite network.

Many compounds with this structure were grown by Hahn and Strick [9]. The unit cell of the defect chalcopyrite has 24 bonds while the normal chalcopyrite has 32 bonds. The valance electron concentration (VEC) for this structure is 4.57 and there are 25% ordered defects in the cation sublattice while in the normal chalcopyrite the VEC is 4, thereby making the defect structure very interesting for studies as they are very difficult to dope, have strong photoconductivity and are resistant to radiation. The latter property is very attractive while working with high-power lasers. In addition, the switching phenomena, memory effects, and partial disorder in these compounds make them attractive for detail studies. In principle, the non-linearity provides a means of thermal contact between different electromagnetic modes. Besides, these structures exhibit some important dependences: for example, their refractive index increases with decreasing energy gap, the energy gap decreases with increasing lattice constant, and nonlinear susceptibilities increase with increasing refractive index, these dependences playing a key role in characterising the performance potential in semiconductors.

3. Theory and method of evaluation

The requirements to materials for nonlinear optical devices are very stringent and initiate the search for new materials, for example, by varying the cationic or anionic concentrations or by using artificial structures based on superlattices and heterostructures. Despite the progress reported in various works, some fundamental parameters such as the lowest energy band gap, ionicity and susceptibilities (linear and nonlinear) are still unknown but very important and necessary for the better understanding of effective charges, cohesive energies, or impurity binding energies.

Both the dielectric constant and the refractive index of a semiconductor material are very important physical parameters from theoretical and applied considerations because their knowledge is highly relevant for the development of many sophisticated semiconductor devices. The optical nonlinearity, on the other hand, is one of the key parameters for assessing the importance of these materials for several frequency mixing devices. In addition, a compound with a high refractive index usually has a higher nonlinearity, thereby stimulating the search for new materials with higher indices.

Using the density matrix formalism and dipole approximation, Shen [10] and Wherret [11] derived the expressions for nonlinear susceptibilities in terms of the perturbation expansion [12]. Out of several approaches [13–16] for evaluating the nonlinearity of materials, Levine's bond charge model [15, 16] appears to be promising while Samanta et al. [17] calculated the optical nonlinearity of some mixed ternary systems and defect crystals [17, 18], with necessary modifications. The linear and nonlinear optical properties of a semiconductor crystal can be assessed by the

proper geometric sum of the corresponding properties of its constituent chemical bonds. Being noncentrosymmetric, the chalcopyrite crystal structure possesses the fundamental property of a nonzero nonlinear optical (NLO) tensor which can take large values. As the defect chalcopyrites are used in some optoelectronic and nonlinear laser devices, it is reasonable to study these types of materials. For a compound of type $A^{II}B_2^{III}\square C_4^{VI}$, the total susceptibility of the compound is given by

$$\chi = \frac{1}{4}(\chi_{AC} + 2\chi_{BC} + \chi_C).$$

The total nonlinearity [or Miller's delta (Δ)] is the sum of individual contributions from different types of bonds and can be written as

$$\Delta = \sum [F\Delta(E_h(x)) + F\Delta(C(x))].$$

According to Levine's bond charge model, the expression for Miller's delta for $A^{II}B_2^{III}\square C_4^{VI}$ crystals can be written in the form

$$\begin{aligned} \Delta_{pqr}(x) = & F\{ \Delta_{pqr}^{II-VI}(C(x)) + \Delta_{pqr}^{II-VI}(E_h(x)) \\ & + 2[\Delta_{pqr}^{III-VI}(C(x)) + \Delta_{pqr}^{III-VI}(E_h(x))] \}, \end{aligned}$$

where

$$F\Delta(C(x)) = 0.288b(Z_\alpha + Z_\beta)[\chi_r^1(x)]^2 C^1(x) V_r^1(x)$$

$$\times \frac{\exp[-k_s r_0(x)]}{E_g^2(x) d^2(x) \chi_{comp}^3 q(x) / e};$$

$$F\Delta(E_h(x)) = 0.232f_e(x) \frac{[\chi_r^1(x)]^2 \rho(x) V_r^1(x)}{d(x) \chi_{comp}^3 q(x) / e}.$$

The quantities F , Δ , $E_h(x)$ and $C(x)$ refer to a particular bond and the other quantities are borrowed from [15, 16] and [17, 18], where the method of evaluation of various parameters was also discussed. While the refractive index of a compound may be found using the expression $\epsilon = 1 + 4\pi\chi_{comp}$, the bond length of atoms in mixed crystals is calculated using the relation first suggested by Van-Vechten and Bergstrasser [19], which makes it possible to find the nonlinearity of the mixed crystal.

Again the presence of defects inherent in the structure makes these compounds attractive for various investigations aimed at examining the effect of the defect on the band gap, nonlinearity, ionicity, and effective charges. The ionicity of a compound is a key factor, which accounts for the trend from semiconducting to insulating behaviour and is responsible for phase transitions from a fourfold to sixfold coordinated structure. Besides, the ionicity concept is also very useful for the complex bonding and mixed semiconducting crystal structure where the material properties are composition dependent. The knowledge of the chemical bond is very important not only in understanding the complex relationship between the crystallographic structure, composition but also in assessing the physical and chemical properties of solids. The estimation of a chemical bond between two atoms with different electro-negativities allows the transfer of electrons between them

and gives rise to the appearance of effective charges [20]. The information on the effective charge of defect chalcopyrites and correlation among many dielectric parameters is needed to investigate the dielectric properties of these compounds and to classify the crystal structures.

In this connection, we have evaluated the effective charges of these mixed defect chalcopyrites for both amalgamated and persistence types using the approach of Hubner [21] and Hubner and Unger [22] but modifying it accordingly for the mixed crystals. Because these types of mixed crystals contain either two types of cations or anions, the probability of finding the valence electron of the unit cell of the mixed crystal changes. For the II-III₂-VI₄ compounds we have thirty two valence electrons $[32(1 - f_i)/2]$ at II and III (2 atoms) and one vacancy $[32(1 + f_c)/2]$ as the four VI atoms. The expressions for the effective charges for these types of compounds are given by

$$q_{\text{II}} = 4f_i - 3.12, \quad q_{\text{III}} = 4f_i - 2.12, \quad q_{\text{vac}} = 4f_i - 2.88,$$

$$q_{\text{VI}} = 2 - 4f_i.$$

In this connection we have evaluated the lowest energy band gap E_g of these mixed crystals from the dependence obtained for E_g on the molecular weight M and the average quantum number \bar{n} of some defect chalcopyrites. The derived results have been compared with the available experimental data. We have obtained for the first time the dependence of E_g on the anion displacement parameter u for this type of compound because it gives valuable information for designing a new optoelectronic device.

4. Results and discussions

The results of our calculations of the optical nonlinearity, refractive index, the results of calculations of the same parameters with Moss's formula as well as the available experimental data are shown in the Table 1. Table 2 presents the results of calculations for the band gap and effective charges. Considering the nonlinearity, we found that when one of the component of the mixed crystal with a lower atomic number is replaced by a corresponding element with a higher atomic number, the nonlinearity increases and this is true both for amalgamated (cationic)

Table 1. Calculated values of the nonlinearity and refractive index, experimental data from paper [23], and the refractive index estimated using Moss's formula for various compounds.

Compound	Value of x	Nonlinearity/ 10^{-6} esu	Refractive index		
			Calculated	Using Moss's formula	Experimental
Hg(Ga _{x} In _{$1-x$}) ₂ Se ₄	0	2.38	2.60	2.66	
	0.25	2.27	2.60	2.63	
	0.50	2.20	2.61	2.58	
	0.75	2.16	2.61	2.55	
	1.00	2.19	2.62	2.51	
Hg(Ga _{x} In _{$1-x$}) ₂ Te ₄	0	2.75	2.83	3.13	2.92
	0.25	2.28	2.83	3.08	
	0.50	2.24	2.84	2.99	
	0.75	2.19	2.84	2.95	
	1.00	2.43	2.85	2.86	
Cd(Ga _{x} In _{$1-x$}) ₂ Se ₄	0	2.12	2.44	2.56	
	0.25	2.09	2.44	2.55	
	0.50	2.08	2.43	2.52	
	0.75	2.08	2.43	2.50	
	1.00	2.08	2.42	2.47	2.52
Cd(Ga _{x} In _{$1-x$}) ₂ Te ₄	0	2.39	2.69	3.00	2.55
	0.25	2.36	2.69	2.91	
	0.50	2.24	2.70	2.86	
	0.75	2.17	2.70	2.82	
	1.00	2.18	2.71	2.75	
CdIn ₂ (Se _{x} Te _{$1-x$}) ₄	0	2.39	2.69	2.86	2.55
	0.25	2.31	2.63	2.77	
	0.50	2.23	2.56	2.69	
	0.75	2.16	2.50	2.64	
	1.00	2.08	2.44	2.56	
Zn _{x} Cd _{$1-x$} Ga ₂ S ₄	0	1.95	2.24	2.29	2.30
	0.25	1.81	2.23	2.29	
	0.50	1.64	2.23	2.27	
	0.75	1.60	2.23	2.25	
	1.00	1.41	2.22	2.27	
Zn _{x} Hg _{$1-x$} Ga ₂ S ₄	0	2.04	2.28	2.27	2.36
	0.25	1.91	2.26	2.26	
	0.50	1.72	2.25	2.25	
	0.75	1.70	2.24	2.24	
	1.00	1.41	2.23	2.22	

Table 2. Values of the band gap and effective charge calculated in this paper and in other experiments for various compounds.

Compound	Value of x	Band gap E_g/eV		Effective charge			
		Calculation	Experiment	$-q_{II}$	q_{III}	q_{VI}	q_{vac}
$Hg(Ga_xIn_{1-x})_2Se_4$	0	1.55	1.58 [24]	0.64	0.36	0.48	1.84
	0.2	1.60	1.69 [24]	0.68	0.32	0.44	1.80
	0.4	1.70	1.79 [24]	0.72	0.28	0.40	1.76
	0.6	1.80	1.89 [24]	0.76	0.24	0.36	1.72
	0.8	1.85	1.99 [24]	0.80	0.20	0.32	1.68
	1.0	1.95	2.08 [24]	0.84	0.16	0.28	1.64
$Hg(Ga_xIn_{1-x})_2Te_4$	0	0.80	0.78 [25]	0.52	0.48	0.60	1.92
	0.2	0.85		0.60	0.40	0.52	1.88
	0.4	0.90		0.68	0.32	0.44	1.88
	0.6	1.00		0.76	0.24	0.36	1.72
	0.8	1.05		0.84	0.16	0.28	1.64
	1.0	1.15		0.92	0.08	0.20	1.56
$Cd(Ga_xIn_{1-x})_2Se_4$	0	1.76	1.81 [26]	0.32	0.68	0.80	2.16
	0.2	1.80		0.44	0.56	0.68	2.04
	0.4	1.90		0.56	0.44	0.56	1.92
	0.6	2.00		0.68	0.32	0.44	1.80
	0.8	2.05		0.80	0.20	0.32	1.68
	1.0	2.15	2.27 [27]	0.92	0.08	0.20	1.56
$Cd(Ga_xIn_{1-x})_2Te_4$	0	0.95	1.15 [23]	0.64	0.36	0.48	1.84
	0.2	1.05		0.73	0.27	0.39	1.75
	0.4	1.10		0.82	0.18	0.30	1.66
	0.6	1.20		0.90	0.06	0.22	1.57
	0.8	1.25		0.99	0.01	0.13	1.49
	1.0	1.35		0.08	0.01	0.04	1.40
$CdIn_2(Se_xTe_{1-x})_4$	0	0.95	1.15 [23], 1.27 [28]	0.64	0.32	0.48	1.84
	0.2	1.10	1.25 [28]	0.57	0.42	0.54	1.90
	0.4	1.25	1.28 [28]	0.51	0.49	0.61	1.97
	0.6	1.45	0.35 [28]	0.45	0.55	0.67	2.03
	0.8	1.60	1.40 [28]	0.38	0.62	0.74	2.09
	1.0	1.73	1.81 [26], 178 [28]	0.32	0.68	0.80	2.16
$Zn_xCd_{1-x}Ga_2S_4$	0	2.90	2.92 [6]	0.88	0.12	0.28	1.76
	0.2	2.95					
	0.4	3.00					
	0.6	3.05					
	0.8	3.10					
	1.0	3.18	3.18 [6]	0.88	0.12	0.24	1.60
$Zn_xHg_{1-x}Ga_2S_4$	0	2.79	2.79 [6]	0.80	0.20	0.32	1.68
	0.2	2.80		0.82	0.18	0.30	1.64
	0.4	2.90		0.83	0.17	0.29	1.65
	0.6	2.95		0.85	0.15	0.27	1.63
	0.8	3.05		0.86	0.13	0.26	1.62
	1.0	3.15	3.18 [6]	0.88	0.12	0.24	1.60

and persistence types (anionic) of compounds. In addition, the nonlinearity increases if Cd is replaced by Hg and Se by Te, confirming that the contribution from a stronger ionic bond is much greater than from a weaker one. Analysis of the results shows that the $Hg(Ga_xIn_{1-x})_2Se_4$ crystal might be a suitable nonlinear material for the visible range, while the $Cd(Ga_xIn_{1-x})_2Se_4$ and $Cd(Ga_xIn_{1-x})_2Te_4$ crystals – in the NIR region, and the $Hg(Ga_xIn_{1-x})_2Te_4$ and $CdIn_2(Se_xTe_{1-x})_4$ crystals – in the FIR region of the spectrum having sufficient nonlinearity for the device design. Currently, good quality crystals for mid-IR generation through frequency down-conversion are limited and their commercial availability [29] is also low so this study may give some impetus for finding new materials. Analysis of the

optical nonlinearity shows that bowing is more pronounced in the former than in the latter case and it is possible to set the desired nonlinearity either by varying the cationic/anionic concentration or both.

We have evaluated the band gap E_g at various concentrations x from its dependence on the molecular weight M and the average quantum number \bar{n} (Fig. 2). Then, the results were compared with the available experimental data listed in Table 2. The agreement is quite encouraging. Analysis of the dependence $E_g(x)$ shows that bowing is more pronounced [30, 31] when varying the cationic concentration (see Fig. 3), and is virtually absent (i.e., the dependence is linear) when varying the anionic concentration (see Fig. 4), thus confirming the earlier observations

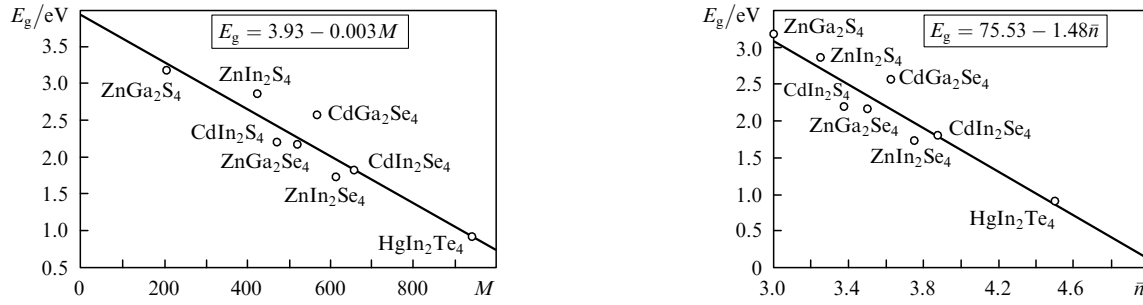


Figure 2. Dependences of E_g on the molecular weight M and average quantum number \bar{n} for some defect chalcopyrites.

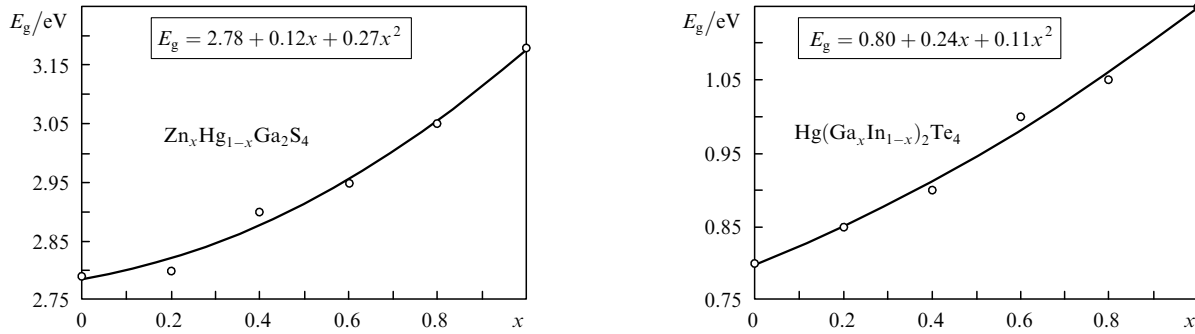


Figure 3. Dependences $E_g(x)$ at different cationic concentrations.

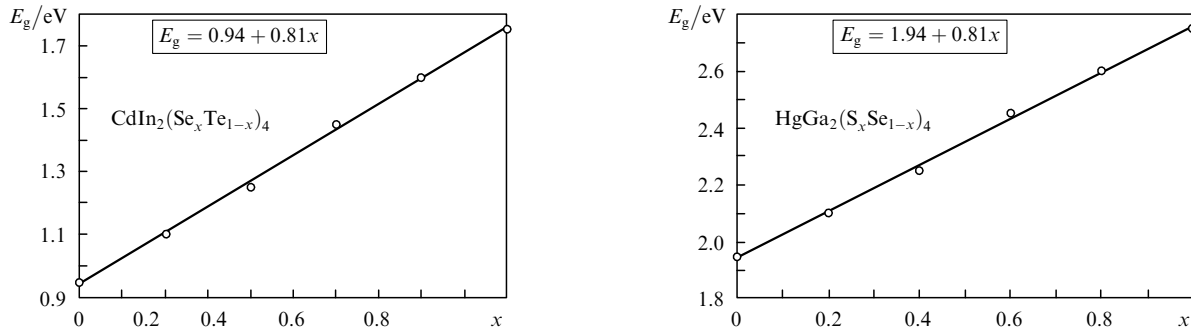


Figure 4. Dependences $E_g(x)$ at different anionic concentrations.

[32–34]. Considering the effective charges one can see that the defect plays a vital role in the charge neutrality and the structures under study do possess the positive effective charge. The analysis of the results shows that for the compound containing Zn, the change in the effective charge q_{VI} is very small when S is replaced by Se but is more pronounced for compounds containing Cd with Se replaced by Te, while for compounds with Hg the change is also small. Because the ionicity of the mixed compound $Zn_xCd_{1-x}Ga_2S_4$ remains almost the same when x changes, the effective charges do not vary as well, and hence are not shown in Tables 1, 2.

Table 1 shows the evaluated refractive indices of the compounds together with the experimental values and the values obtained using Moss’s formula. The agreement is quite encouraging reflecting the fact that Moss’s formula for binary compounds holds good also for multinary compounds. Note in this connection that the both defect compounds $CdGa_2S_4$ and $HgGa_2S_4$ have a refractive index crossing (i.e., an isotropy point) where the crystal changes its sign of birefringence, thus allowing one to construct a narrow band tunable optical filter (NBTOF). For example,

$ZnGa_2S_4$ has no refractive index crossing where replacement of Zn by Cd through the proper choice of x makes it possible to construct a NBTOF, very important for technological applications.

We have been able to obtain for the first time the dependence of the lowest energy band gap E_g on the anion displacement parameter u for some defect chalcopyrites (Fig. 5). The anion displacement is a key parameter because it controls the balance between the covalent and ionic contributions into the band gap and causing the band gap to increase as this parameter increases, thereby showing the importance of this correlation for the development of new materials for technological applications. An attempt to obtain a correlation between the tetragonal distortion and the square of the electronegativity difference for these defect chalcopyrites containing Hg and Cd, as was done by Noolandi [35] separately for Zn and Cd compounds belonging to II-IV- V_2 chalcopyrites, is far from encouraging. This may be explained by the fact that the II-IV- V_2 compounds have two different types of cations, while the defect chalcopyrites have four types if the vacancy is incorporated. The presence of more cations compared to

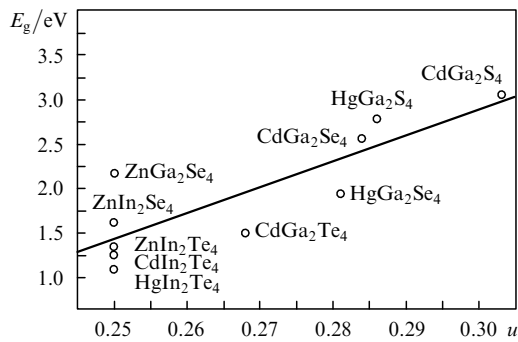


Figure 5. Calculated dependence of E_g on the anion displacement parameter u .

simple II-IV-V₂ compounds makes the situation complicated and results in the absence of such correlation, pointing to the fact that the defect plays a crucial role in this case. Thus, we can conclude that Noolandi's correlation holds only for simple chalcopyrite compounds and not for chalcopyrites containing more than two cations.

The high photoconductivity and problem of doping in these compounds provides evidence for large number of states in the gap. This is valid for all defect compounds; therefore, we can conclude that these states are vacancy related. While the dielectric constant and the refractive index are averaged macroscopic values, the effective charge characterises each ion in a crystal because it is well known that no single effective charge can be defined in a crystal as it varies from phenomena to phenomena.

5. Conclusions

The wide range of physical properties found in defect ternary chalcopyrites makes them promising for using in different devices. The study of various properties of the compounds and improved control of defects during the crystal growth, the development of new devices using ternary and quaternary defect compounds will be attractive and promising for future technological applications.

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