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Correlation Between Refractive Index and Electronegativity Difference for $A^N B^{8-N}$ Type Binary Semiconductors

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A simple correlation between the high-frequency refractive index and electronegativity difference has been established for large number of $A^N B^{8-N}$ type binary semiconductors. The proposed relation has been applied satisfactorily to evaluate the refractive index of binary semiconductors belonging to groups I–VII, II–VI, III–V and IV–VI. The estimated values of refractive index from present relation are utilized to calculate the electronic polarizability of compounds by applying the Lorentz–Lorentz formula. Our calculated values of refractive index and electronic polarizability are in good agreement with the values evaluated by earlier researchers.

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1. Introduction

The compound semiconductors have potential applications in the fields of electronics, optical, optoelectronic devices and nowadays in nanotechnology and biotechnology [1–5]. Due to technological importance of these materials, the experimental and theoretical study of fundamental properties of such compounds has attracted much attention [6–10]. The optical refractive index is one of the fundamental properties of materials because it is closely related to the electronic polarizability of ions and local field inside the material, which plays an important role in determining the electrical properties of such materials. Therefore, the evaluation of refractive index has been a matter of considerable importance and several investigations have been carried out for this purpose [11–21].

Moss [11] made first attempt to interrelate the refractive index and energy band gap in semiconductors and proposed a relation as

$$E_{\mathrm{g}} n^4 = 95 eV, \tag{1}$$

where n, E_{g} are the optical refractive index and the energy band gap, respectively. This relation was based on the general assumption that all energy levels in a solid are scaled down by a factor of $1/\varepsilon_{\mathrm{eff}}^2$, where $\varepsilon_{\mathrm{eff}}$ is effective dielectric constant. A review article on this subject can be found in Ref. [12]. Ravindra et al. [13] have proposed a linear relation governing the variation of optical refractive index with energy gap between bonding and antibonding states as

$$n = 4.08 - 0.62 E_{\mathrm{g}}. \tag{2}$$

But this relation does not provide reasonably good results for compounds having low values of the refractive index. Finkenrath [14] has widely pointed out the shortcomings of Ravindra's relation at low and high values of refractive index. Gopal [15] has modified Penn model [16] for semiconductors possessing high frequency dielectric constant and proposed a general formula relating refractive index with energy band gap. In order to remove the shortcomings of Moss [11] and Ravindra's relation [13], Reddy et al. [17] have proposed another relation between refractive index and energy gap in the semiconductors. This relation is as follows:

$$n = 3.59 - \log_{\mathrm{e}} E_{\mathrm{g}}. \tag{3}$$

Based on the theory of vibrations, Hervé and Vandamme [18] proposed the following relation for refractive index as:

$$n = \sqrt{1 + \left(\frac{A}{E_{\mathrm{g}} + B}\right)^2}, \tag{4}$$

where $A = 13.6$ eV and $B = 3.4$ eV are constants. Hervé and Vandamme [18] predicted that this relation provides the lowest deviation for III–V, I–VII group semiconductors and chalcopyrite. This model is accurate for most materials usually employed in the optoelectronic device structures and high band gap materials. However, it does not explain the behavior of the IV–VI group semiconductors. Bussmann-Holder [19] has investigated the interplay of polarizability and ionicity in some IV–VI compounds within a diatomic linear chain model. Pan et al. [20] have studied the effect of the electronegativity and bond ionicity on energy band gap (E_{g}) and the refractive index (n) in nitrogen-doped $\mathrm{SnO_2}$ and other doped semiconductors by extending Bussmann-Holder's model. Recently Reddy et al. [21] have proposed an interesting relation between optical refractive index and electronegativity difference for large number of binary and ternary chalcopyrite semiconductors, insulators, oxides and alkali halides. For binary compound semiconductors having low values of the refractive index (1–2), this relation gives better results, but for high value of refractive index (4.0–5.7) the deviation is very high (45.5%). Several models based on energy gap and electronegativity

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difference have been proposed by earlier researchers for calculating the refractive index of materials, but they are mostly applicable to limited number of materials.

In the present work, we propose a simple correlation between optical refractive index and electronegativity difference for A^NB^{8-N} type binary compound semiconductors (including I–VII, II–VI, III–V and IV–VI group compound semiconductors). The results obtained from present approach are found to be consistent with the known values and reported by other authors. The validity of earlier proposed relations [21] have been examined for these compound semiconductors and pronounced deviations between known and evaluated values of refractive index were observed. Employing the Lorentz–Lorentz formula in classical theory, the electronic polarizability of these group semiconductors has also been calculated.

2. Theory and calculation

The optical electronegativity is a key parameter to understand the nature of chemical bonding and other important parameters. Duffy [22, 23] has correlated the energy gap and optical electronegativity difference for various binary systems as

$$E_g = 3.72\left(\Delta\chi^*\right), \tag{5}$$

where $\Delta\chi^* = [\Delta\chi^*_{\text{anion}} - \Delta\chi^*_{\text{cation}}]$ and $\Delta\chi^*_{\text{anion}}$ and $\Delta\chi^*_{\text{cation}}$ are optical electronegativites of anion and cation, respectively. Using Duffy relation, Eq. (5), Reddy et al. [21] have modified Hervé and Vandamme relation [18] and proposed an interesting relationship between refractive index, energy gap and optical electronegativity difference as

$$n = \sqrt{1 + \left(\frac{A}{3.72\Delta\chi^* + B}\right)^2}. \tag{6}$$

In addition, they have also modified Moss [11] and Ravindra's relation [13] which are given as follows:

$$n = \left[\frac{25.54}{\Delta\chi^*}\right]^{1/4} \tag{7}$$

and

$$n = 4.08 - 2.3064\left(\Delta\chi^*\right). \tag{8}$$

Singh et al. [24] have correlated electronic dielectric constant and average energy gap as

$$\varepsilon_\infty = 1 + bE_g^S, \tag{9}$$

where ε_∞ is optical dielectric constant, b and S are an ion characteristic constant and family characteristic constant, respectively. Using Eq. (5), above relation (9) can be written as

$$\varepsilon_\infty = 1 + b\left(3.72\Delta\chi^*\right)^S. \tag{10}$$

Since optical refractive index is related to dielectric constant as $n^2 = \varepsilon_\infty$, Eq. (10) can be rewritten as

$$\frac{n^2 - 1}{b} = \left(3.72\Delta\chi^*\right)^S. \tag{11}$$

$\Delta\chi^{*S} = \frac{n^2-1}{c}$, where $c = b3.72^S$ is a constant.

$$\Delta\chi^* = c^{-1/S} n^{2/S}\left[1 - \frac{1}{n^2}\right]^{1/S},$$

$$\Delta\chi^* = c^{-1/S} n^{2/S}\left[1 - \frac{1}{nS^2} + \ldots\right],$$

using binomial expansion up-to the second term since $\frac{1}{n^2} \ll 1$,

$$\Delta\chi^* = c^{-1/S} n^{2/S} - \frac{1}{Sc^{1/S}} n^{-2\left(1-\frac{1}{S}\right)},$$

$$\Delta\chi^* = ln^m + l'n^{m'}$$

where $l = c^{-1/S}$, $l' = \frac{-1}{Sc^{1/S}}$ and $m = \frac{2}{S}$, $m' = -2\left(1 - \frac{1}{S}\right)$. Employing values of constants, the contribution due to second term, $l'n^{m'}$ is found negligible and hence,

$$\Delta\chi^* = ln^m,$$

$$n = \frac{1}{l^{1/m}}\Delta\chi^{*1/m}, \tag{12}$$

$$n = k\Delta\chi^{*\gamma}, \tag{13}$$

which is our proposed relation between refractive index and electronegativity difference. Here k and γ ($\gamma = -0.32$) are constants, which have been obtained by optimizing known values of refractive index (n) and electronegativity difference ($\Delta\chi^*$) data in regression software. In present work, the necessary optical electronegativity difference data were taken from Refs. [25, 26]. Using Eq. (13), we have calculated the refractive indices for binary compound semiconductors belonging to groups I–VII, II–VI, III–V and IV–VI, and presented in Table I. The proposed relation is found to hold well with a constant value of k for each group of compounds such as I–VII ($k = 2.0$), II–VI ($k = 2.4$), III–V ($k = 2.38$), and IV–VI ($k = 4.0$) compounds. The evaluated values show a systematic trend and are consistent with the available data reported so far, which proves the validity of the present approach. The value of the constant k is considered to be the characteristics of each group of semiconductors. It is not unique for all groups of semiconductor, in contrast with the unique values as predicted by other authors. In Table I a comparison has also been made between our calculated values with the Reddy modified relations Eqs. (6), (7) and (8).

The calculated refractive index from Eq. (13) has been utilized to evaluate the electronic polarizability (α_e) by applying the Lorentz–Lorentz formula [31] as

$$\alpha_e = \frac{n^2 - 1}{n^2 + 2}\frac{M}{d}3.95 \times 10^{-25}\ \text{cm}^3, \tag{14}$$

where M and d are molecular weight and density of compounds, respectively. The present calculated and known values of electronic polarizability have been presented in Table II. A comparison has also been made between our calculated values and reported by earlier researchers in the same table. Our calculated values of α_e are in closer agreement with the known [27, 28, 32] one and earlier reported values [13, 15, 17, 29].

TABLE I

Refractive index (n) of binary compounds.

Compounds	$\Delta\chi^*$ [25, 26]	Known values [13, 27–30]	Present work Eq. (13)	Calc. by Reddy et al. [21]		
				Hervé Eq. (6)	Moss Eq. (7)	Ravindra Eq. (8)
I–VII						
LiF	3.0	1.39	1.41	1.37	1.71	NA
LiCl	2.0	1.66	1.60	1.60	1.89	NA
LiBr	1.8	1.78	1.66	1.68	1.94	NA
LiI	1.5	1.95	1.76	1.81	2.03	0.62
NaF	3.1	1.34	1.39	1.35	1.69	NA
NaCl	2.1	1.54	1.58	1.51*	1.82*	NA
NaBr	1.9	1.64	1.63	1.58*	1.88*	NA
NaI	1.6	1.77	1.72	1.76	2.00	0.39
KF	3.2	1.36	1.38	1.34	1.68	NA
KCl	2.2	1.49	1.55	1.52*	1.83*	NA
KBr	2.0	1.56	1.60	1.58*	1.87*	NA
KI	1.7	1.68	1.69	1.72	1.97	0.159
RbF	3.2	1.40	1.38	1.34	1.68	NA
RbCl	2.2	1.49	1.55	1.54*	1.85*	NA
RbBr	2.0	1.55	1.60	1.58*	1.88*	NA
RbI	1.7	1.65	1.69	1.72	1.97	0.159
CsF	3.3	1.48	1.36	1.32	1.67	NA
CsCl	2.3	1.61	1.53	1.56*	1.86*	NA
CsBr	2.1	1.67	1.58	1.60*	1.89*	NA
CsI	1.8	1.79	1.66	1.72*	1.97*	0.19*
CuF	2.1	1.58	1.58	1.57	1.87	NA
CuCl	1.1	1.93	1.94	2.26*	2.32*	2.03*
CuBr	0.9	2.10	2.07	2.38*	2.39*	2.28*
CuI	0.6	2.35	2.36	2.37*	2.38*	2.26*
AgF	2.0	1.70	1.60	1.60	1.89	NA
AgCl	1.0	2.00	2.00	2.16	2.25	1.77
AgBr	0.9	2.15	2.07	2.25	2.31	2.00
AgI	0.6	2.22	2.36	2.61	2.55	2.70
II–VI						
MgO	1.93	1.63	1.94	1.63*	1.91*	NA
MgS	1.27	2.26	2.22	1.75	1.99	0.339
MgSe	1.20	2.43	2.26	1.54	1.85	NA
MgTe	0.79	2.65	2.59	1.48	1.80	NA
CaO	2.50	1.82	1.79	1.73*	1.98*	0.21*
CaS	1.50	2.12	2.11	2.49	2.46	2.48
CaSe	1.40	2.26	2.16	2.21	2.28	1.91
SrO	2.50	1.80	1.79	1.70*	1.96*	0.06*
SrS	1.50	2.10	2.11	2.10	2.21	1.63
SrSe	1.40	2.21	2.16	2.01	2.15	1.34
SrTe	1.10	2.41	2.33	1.78	2.01	0.482
BaO	2.55	1.98	1.78	1.87*	2.07*	0.87*
BaS	1.60	2.15	2.06	1.78	2.01	0.48
BaSe	1.50	2.27	2.11	1.62	1.90	NA
ZnO	1.90	2.00	1.95	2.16*	2.25*	1.79*
ZnS	0.90	2.39	2.48	2.20*	2.28*	1.89*
ZnSe	0.80	2.43	2.58	2.49*	2.47*	2.49*
ZnTe	0.50	2.70	3.00	2.61*	2.55*	2.68*
CdO	0.67	2.68	2.73	2.52*	2.48*	2.53*
CdS	0.90	2.38	2.48	2.55*	2.51*	2.60*
CdSe	0.86	2.44	2.52	2.85*	2.74*	3.03*
CdTe	0.38	3.23	3.27	2.99*	2.85*	3.19*
III–V						
AlN	1.43	2.20	2.12	2.14*	2.24*	1.73*
AlP	0.60	2.75	2.80	2.35*	2.37*	2.23*
AlAs	0.50	2.92	2.97	2.65*	2.58*	2.75*
AlSb	0.40	3.19	3.19	2.90*	2.78*	3.09*
GaN	1.40	2.24	2.14	2.28*	2.33*	2.07*
GaP	0.50	2.90	2.97	2.61*	2.55*	2.70*
GaAs	0.37	3.30	3.27	3.04*	2.90*	3.25*
GaSb	0.24	3.75	3.76	3.38*	3.29*	3.58*
InN	1.26	2.35	2.21	1.96	2.12	1.17
InP	0.41	3.10	3.17	3.08*	2.94*	3.30*
InAs	0.30	3.51	3.50	3.76*	4.04*	3.86*
InSb	0.23	3.96	3.81	3.93*	4.80*	3.97*
IV–VI						
SnS	0.70	4.42	4.48	2.48	2.46	2.47
SnSe	0.60	4.79	4.71	2.61	2.55	2.70
PbS	0.70	4.15	4.48	2.48	2.46	2.47
PbSe	0.60	4.78	4.71	3.82*	4.27*	3.90*
PbTe	0.30	5.73	5.88	3.79*	4.16*	3.88*
Average dev. [%]			3.53	10.28	14.57	31.37

Quantities with an asterisk* are taken from Ref. [21],
those without are from present calculations; NA — not applicable.

TABLE II

Electronic polarizability (α_e) of binary compounds.

Compounds	Mol. weight	Density [g/cm^3] [27, 33]	Electronic polarizability [10^{-24} cm^3]					
			Known values [27, 28, 32]	Present work Eq. (10)	Ravindra et al. [13]	Gopal et al. [15]	Reddy et al. [17]	Kumar et al. [29]
LiF	25.937	2.64	0.89	0.96			1.14	1.03
LiCl	42.392	2.07	2.98	2.78			2.55	2.43
LiBr	86.843	3.46	4.12	3.65			3.51	3.27
LiI	133.843	3.49	6.15	6.21			6.95	6.01
NaF	41.988	2.56	1.15	1.54			1.81	1.89
NaCl	58.443	2.17	3.24	3.53			3.47	3.49
NaBr	102.894	3.20	4.38	4.51			4.48	4.42
NaI	149.894	3.67	6.41	6.38			6.97	6.46
KF	58.10	2.48	1.99	2.14			2.28	2.63
KCl	74.555	1.98	4.08	4.77			4.69	4.90
KBr	119.006	2.75	5.22	5.86			5.74	5.92
KI	166.006	3.13	7.25	7.99			8.47	8.13
RbF	104.468	3.56	2.54	2.68			2.96	3.39
RbCl	120.923	2.80	4.63	5.47			5.40	5.75
RbBr	165.374	3.35	5.77	6.69			6.51	6.79
RbI	212.374	3.55	7.80	9.01			9.57	9.24
CsF	151.903	4.12	3.60	3.25			3.85	4.37
CsCl	168.358	3.99	5.69	5.17			5.44	5.69
CsBr	212.809	4.44	6.83	6.28			6.46	6.46
CsI	259.809	4.51	8.86	8.37			8.97	8.78
CuF	82.54	4.23	2.57	2.56				
CuCl	98.99	4.14	4.50	4.53	4.82	5.75	5.77	
CuBr	143.45	4.98	6.05	5.94	6.58	7.27	7.27	
CuI	190.44	5.62	8.05	8.06	8.43	8.93	8.88	
AgF	126.87	5.85	3.31	2.94				
AgCl	143.32	5.56	5.09	5.09	5.60	6.36	6.39	
AgBr	187.78	6.47	6.27	5.99	6.81	7.42	7.39	
AgI	234.77	6.00	8.76	9.31	9.29	10.07	9.97	
MgS	56.38	2.84	4.53	4.45				
MgSe	103.27	4.21	6.00	5.61				
MgTe	151.91	4.54	8.82	8.66				
CaO	56.08	3.32	2.90	2.83				
CaS	72.14	2.50	6.13	6.09				
CaSe	119.04	3.57	7.62	7.22				
SrO	103.62	4.70	3.72	3.69	2.73	4.29	4.84	
SrS	119.68	3.70	6.80	6.83	4.01	7.03	7.10	
SrSe	166.58	4.38	8.47	8.24				
SrTe	215.22	4.83	10.84	10.5				
BaO	153.34	5.72	5.22	4.44				
BaS	169.40	4.25	8.61	8.20				
BaSe	216.30	5.02	9.88	9.10				
ZnO	81.37	5.61	2.86	2.78	2.43	3.31	3.34	
ZnS	97.43	3.98	5.91	6.12	4.46	5.72	5.76	
ZnSe	144.33	5.42	6.53	6.87	6.65	7.04	7.00	
ZnTe	192.97	6.34	8.14	8.74	8.09	8.38	8.29	
CdS	144.46	4.82	7.20	7.49	7.78	8.10	8.04	
CdSe	191.36	5.51	8.10	8.79	9.52	9.67	9.58	
AlN	40.99	3.26	2.79	2.68	1.98	2.84	2.86	
AlP	57.96	2.85	5.51	5.59	4.55	5.09	5.09	
AlAs	101.9	3.81	7.55	7.64	7.23	7.45	7.38	
AlSb	148.73	4.22	10.49	10.5	10.3	10.47	10.36	
GaN	83.73	6.10	3.10	2.94	2.83	3.33	3.34	
GaP	100.69	4.13	6.85	6.96	6.52	6.73	6.66	
GaAs	144.64	5.32	8.24	8.20	8.16	8.31	8.23	
GaSb	191.47	5.62	10.94	11.0	10.73	11.05	11.00	
InN	128.83	6.88	4.45	4.17	5.19	5.31	5.26	
InP	145.79	4.79	8.92	9.02	9.22	9.39	9.29	
InAs	189.74	5.66	10.47	10.5	10.89	11.35	11.53	
InSb	236.57	5.78	13.42	13.2	13.44	14.09	14.56	
SnS	150.75	5.22	9.82	9.86				
SnSe	197.65	6.18	11.11	11.1				
PbS	239.26	7.61	10.48	10.7	10.18	10.60	10.73	
PbSe	286.15	8.15	12.19	12.1	11.45	11.97	12.16	
PbTe	334.79	8.16	14.81	14.9	13.36	13.95	14.21	
Average dev. [%]				5.38	10.1	9.64	11.63	16.51

3. Results and discussions

In the present paper, we have presented an empirical relation relating refractive index with the electronegativity difference between constituent atoms of binary compound semiconductor. The computed refractive index from proposed relation for I–VII, II–VI, III–V and IV–VI groups binary semiconductor agree well with the recently reported [18] and known values [13, 27–30]. The behavior of different compounds within a group of compounds has been found almost similar, but significantly different from the compounds of the other groups. It is interesting to note that all groups of binary compound exhibit linear variation of electronegativity difference with the optical refractive index. Our calculated refractive index from proposed relation hardly deviates 0.27% to 11% and the maximum discrepancy for refractive index is 19.0% (MgO) from known values. In addition, the maximum deviation for low values of n (1–2) has been found to 10.2% in comparison to earlier modified Eqs. (6), (7) and (8) which give 17.1%, 26.1%, and 96.7%, respectively. Similarly, for high values of n (4.0–5.7) it has been found to be 7.95% in comparison to recently modified Eqs. (6), (7) and (8) which give 45.5%, 46.8% and 44.1%, respectively. This indicates that our proposed relation, Eq. (13) makes reasonably good predictions for both low and high values of refractive index. The low values of the average percentage deviation (5.38%) in electronic polarizabilities for all studied binary compound semiconductors indicate the correctness of the estimated refractive indices in the present work.

4. Conclusion

From the above results and discussions obtained by using the present approach, we conclude that the proposed relation between optical refractive index and electronegativity difference holds well for large number of A^NB^{8-N} type binary semiconductors and gives better results in comparison to earlier proposed relations. The reasonable agreement between our calculated and previously known values of α_e indicates that the proposed relation is useful and more accurate for estimating the optical refractive index. The minimum average percentage deviation (3.53%) for refractive index obtained in present study indicates the soundness of present approach. It is natural to say that present approach can easily be extended to the other complex compounds for which work is in progress and will be appearing in the forthcoming papers.

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