Optical Properties of Large-Bandgap PbI_2 and $Sb_xBi_{1-x}I_3$ Compounds

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Abstract

The optical absorption and the dielectric functions of PbI2 and $Sb_xBi_{1-x}I_3$ have been investigated experimentally by means of absorption measurements and spectroscopic ellipsometry respectively, and theoretically by a full-potential linear muffin-tin-orbital method. These materials have been recognized as very promising detector materials with large technological applicabilities. The calculated band-gap energies, optical absorption and the dielectric functions were found to be in a very good agreement with the experimental results. The electron-energy-loss functions are also obtained for both experiment and theory for comparison. The band-gap energy as a function of temperature, for PbI_2 has also been measured by optical absorption. The temperature dependence has been fitted by a new relation, and a discussion of this fitting is given.

Introduction

Lead iodide (PbI_2) and antimony-triiodide ($Sb_xBi_{1-x}I_3$) compounds have large bandgap energies and are therefore very promissing materials for future room temperature devices [1,2]. Both materials have hexagonal structures, were grown by the Bridgman method, characterized by transmission spectroscopy and spectroscopic ellipsometry (PbI_2) as well as be the temperature dependence of the measured bandgap energy, Eg, (PbI_2). Theoretically they were investigated by a full-potential linear muffin-tin-orbital method (FPLMTO) [1]. The electron-energy-loss (EEL) function which is a function of the dielectric functions is also obtained.

Experimental Setup

Figure 1 shows the experimental setup.

Results and Discussions

The imaginary part of the dielectric function, $\varepsilon_2(\omega) \equiv \mathrm{Im}\,\varepsilon(\mathbf{q}=0,\omega)$, has been obtained using the joint density-of-states and the optical matrix overlap [1]. It is written as

$$\varepsilon_2^{ij}(\omega) = \frac{4\pi^2 e^2}{\Omega m^2 \omega^2} \sum_{\mathbf{k}nn'\sigma} \langle \mathbf{k}n\sigma | p_i | \mathbf{k}n'\sigma \rangle \langle \mathbf{k}n'\sigma | p_j | \mathbf{k}n\sigma \rangle \times f_{\mathbf{k}n}(1 - f_{\mathbf{k}n'}) \delta(e_{\mathbf{k}n'} - e_{\mathbf{k}n} - \hbar\omega) \quad (1)$$

In Eq. (1), *e* is the electron charge, *m* its mass, Ω is the crystal volume and f_{kn} is the Fermi distribution. Moreover, $|\mathbf{k}n\sigma\rangle$ is the crystal wave function corresponding to the nth eigenvalue with crystal momentum **k** and spin σ.

The real part of the dielectric function, $\varepsilon_1(\omega)$, was calculated from the Kramers-Kronig dispersion relatios. The electron-loss-energy function (EEL) is obtained from the relation

$$ELF = - Im \{ 1 / [\varepsilon_1(\omega) + i \varepsilon_2(\omega)] \} \quad (2)$$

The measured temperature dependence of the band-gap energy, Eg, is extracted from a more accurately relation compared to that of Varshni [1]

$$Eg(T) = Eg(0) - S \langle h\omega/2\pi \rangle [\coth (\langle h\omega/2\pi \rangle / 2K_BT) - 1] \quad (3)$$

Where S is a dimensionless coupling constant, and $\langle h\omega/2\pi \rangle$ is an average phonon energy. For PbI_2 we obtain Eg(0)=2.485eV , S=3.60, and $\langle h\omega/2\pi \rangle$ = 13.1 meV. For absorption measurement Eg(10K)=2.484 eV. At room temperature we obtain Eg(300K)=2.345eV from absorption measurement and Eq(3) as well.. For photoacoustic and transmission spectroscopies at 300K ,we obtain the values Eg(300K)=2.320 eV and 2.319 eV respectively.

The electron-energy-loss (EEL) is shown in Fig.2 with the dielectric functions obtained from spectroscopic ellipsometry and in Fig. 3 from our calculations. From Figures 2 and 3 we notice onsets of absorption around 2.4 eV (corresponding to Eg) and around 3.0 and 3.5 eV. In order to have a more reliable identification of the plasma peak, further investigation of the optical transitions at higher photon energy is necessary.

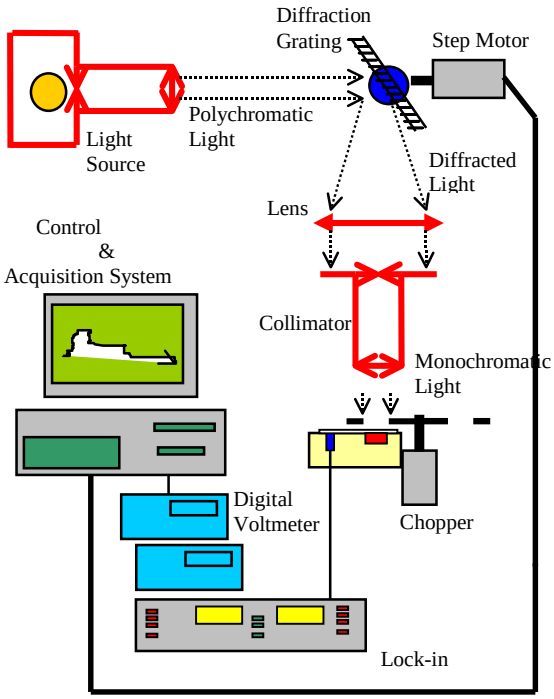

Fig. 1

Figure 4 gives both Eg (0 K) and Eg (300K) from absorption measurement and the fitting equation 3. Figure 5 gives Eg (Theory) ≅ Eg (Exp.) around 2.12 eV.

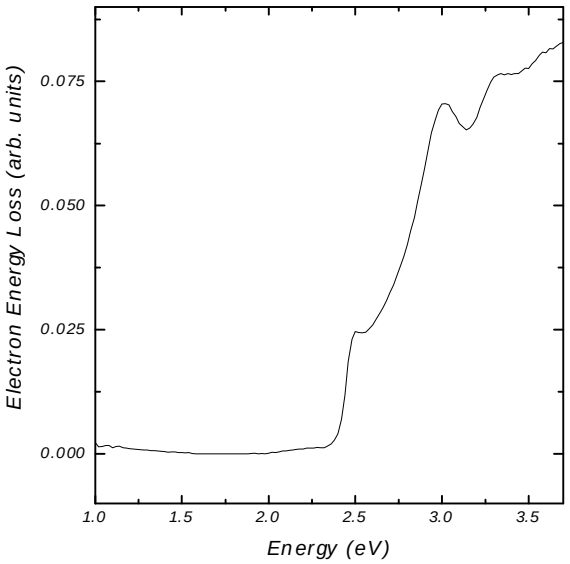

Fig. 2 - EEL of PbI_2 Experiment

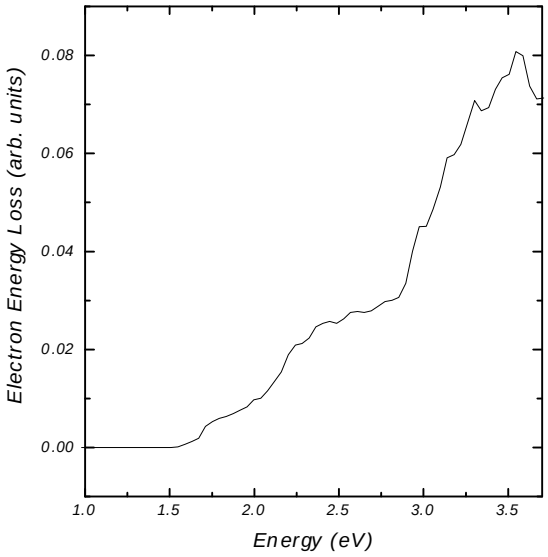

Fig. 3 - EEL of PbI_2 Theory

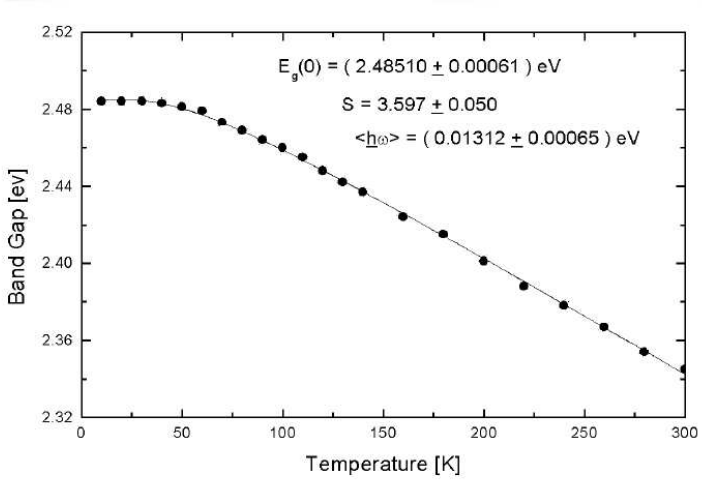

Fig. 4 - Measured variation of Eg (T) for PbI_2 , as full circles. Curve shows the best fit to the experimental results obtained with the use of Eq.(3)

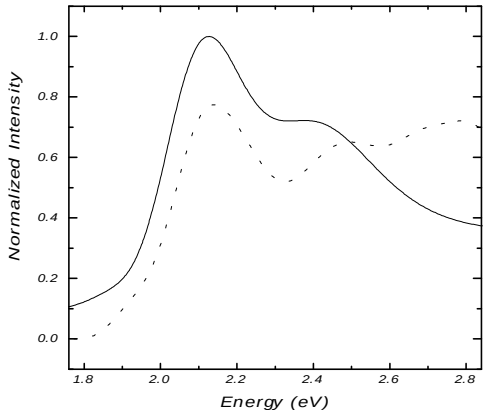

Fig. 5 - Absorption for $SbBiI_3$ (x = 1). Full line (Exp), dotted line (Theory)

Conclusions

We have investigated the optical properties of PbI_2 and $Sb_xBi_{1-x}I_3$. The experimental and theoretical results present a reasonable agreement between them.

References

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Acknowledgements

This work was financially supported in part by the Brazilian National Research Council (CNPq), Swedish Research Council (VR), Danish Natural Science Research Council ,the Carlsberg Foundation and FAPESB (Bahia).