Linear frequency-dependent optical characteristics of perovskites of the $CsSn[Br_{1-x}I_x]_3$ system

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Abstract: - The optical properties of perovskites of the $CsSn[Br_{1-x}I_x]_3$ system in the spectral frequency range of 3–10 THz were studied by quantum-chemical calculations in the framework of DFT using the Wien2K package, and the frequency dependences of the refractive index, photoconductivity, and absorption coefficient of these nanocrystals were estimated. It has been established that in the indicated frequency range with an increase in the iodine content, the absorbing properties of the $CsSn[Br_{1-x}I_x]_3$ (x=0.25, 0.50, 0.75, 1.00) system increase linearly.

Key-Words: - Wien2K package; FP-LAPW; optical properties; solar energy materials; frequency dependence; terahertz technology; absorption coefficients

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1 Introduction

The use of various nanoscale semiconductor materials in photovoltaic (PV) applications in recent years have led to the creation of highly efficient solar cells [1-3], and hybrid perovskite nanostructures and thin films based on them have found wide application and rapid development due to their unique optoelectronic properties, such as excellent mobility carriers, long diffusion length, high optical gain, and nonlinear response, adjustable and controlled band gap, as well as inexpensive methods and fabrication technologies [2–4]. Many researchers are focusing on their fundamental properties to explore the possibilities of new applications such as lasers, storage devices, and terahertz (THz) detectors [5,6]. Other advantages of devices based on perovskite nanostructures are the ease of fabrication of devices based on them and a wide choice of substrate materials and low cost [7]. Moreover, the high quantum efficiency and stability of perovskite nanostructures under normal conditions and many tunable optoelectronic properties make them ideal for a wide range of electronic and optical applications [8–11].

Another aspect of the problem is that thanks to the achievements of scientists in recent years, perovskites have demonstrated excellent performance for LEDs and the desired photovoltaic efficiency [12]. Day after day, there is a huge demand for ultra-fast tunable devices operating especially at terahertz (THz) frequencies, as they open up huge opportunities for a wide range of applications ranging from image processing, spectroscopy, and wireless communication [12–15]. The existence of self-organizing quantum wells due to the alternating arrangement of organic and inorganic atomic layers in perovskite systems provides an additional relaxation path for photoexcited free carriers to relax back at ultrafast time scales making them ideal candidates for ultrafast active photonic devices [16-18]. In recent years, perovskites have been widely used to achieve unprecedented control over terahertz waves due to their tunable optical properties through structural reconfiguration [19,20]. In addition, the integration of semiconductors with metamaterials offers a unique platform for the dynamic control and management of longwave and terahertz radiation to study the effects of

interfacial coupling in the resulting hetero nanostructures [21-24].

With the development of terahertz time analysis (TDS) technology, it has become possible to measure terahertz optical properties, such as refractive indices and absorption coefficients, and study low-frequency optical phonon resonances [5, 25–27]. The study of the properties of materials using waves of the terahertz (THz) range is associated with the study of the frequency interval that occupies a part of the electromagnetic spectrum between the infrared (IR) and microwave ranges, that is, the terahertz region of electromagnetic frequencies is in the range from 0.3 to 10 THz [28]. In recent years, the absorption and refractive index of inorganic Sn-containing perovskites, including $CsSn(Br, I)_3$ systems, have been experimentally studied [29-31], and various other perovskite structures in the terahertz frequency range from 0.3 to 3 THz [32-35], but data on their properties in the terahertz frequency range of 3–10 THz are still scarce. Therefore, the optical properties of these materials in the terahertz frequency range of electromagnetic radiation require further study. In this article, ab-initio quantum chemical calculations are implemented that investigate the refractive indices, and adsorption properties of the $CsSn[Br_{1-x}I_x]_3$ system since the prediction of the optical properties of materials based on the density functional theory (DFT) has become a revolutionary approach that allows one to conduct a fairly effective search for optical materials even without the experimental formation of materials [34, 36-38].

2 Materials and Methods

The relaxation of the orthorhombic phase (Pnma, 62) of the $CsSn[Br_{1-x}I_x]_3$ system using unit cells of 20 atoms was carried out within the time-dependent density functional theory (DFT) implemented in the Wien2k simulation package [39]. The exchange-correlation effects of electrons were taken into account using the modified Becke-Jones approximation (mBJ) [40] since it was shown in many works that mBJ gives an accurate and experimentally comparable estimate of the bandgap [41-54] compared to other known approximations, such as GGA and LDA. The outer s-, p-, and d-electrons were considered valence electrons. All calculations were carried out, taking the spin-polarized and spin-orbit effects into account. Calculations of optical properties according to the nonstationary density functional theory were carried out according to the schemes proposed in [55,56]. For the wave function in the interstitial region, the plane wave cutoff value Kmax = 7/RMT was chosen. For integration in k-space in the Brillouin zone (BZ), a grid of 1000 k-points was used. The charge convergence was chosen to be 0.0001e during self-consistency cycles. The energy cutoff was chosen to be -6.0 Ry, which determines the separation of the valence and core states.

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3 Results and Discussion

According to the results presented in Figure 1, it can be seen that the photoconductivity of these systems increases slowly and linearly with an increase in the iodine concentration in the system, as well as with an increase in the frequency of terahertz waves. However, for the refractive index of these nanocrystals, the decrease is very small (less than 3%) since the wave frequencies increase, and the value of n in the (3-10) terahertz range remains almost unchanged (Figure 2), which is in very good agreement with results given in [57,58].

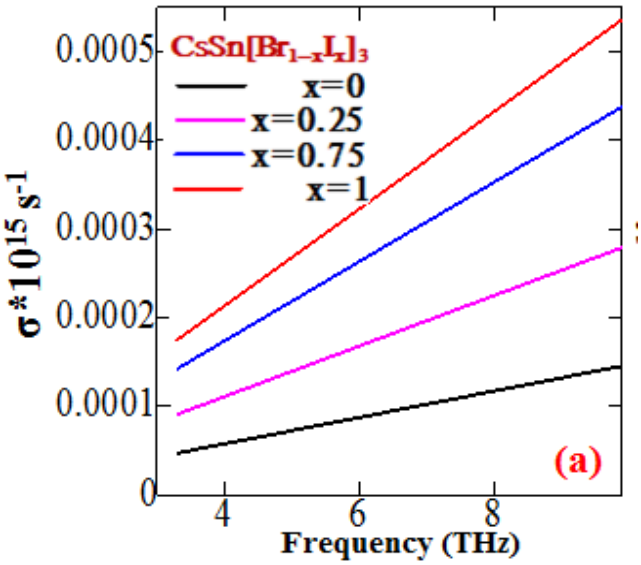

Figure 1. Frequency dependence of photoconductivity of the $CsSn[Br_{1-x}I_x]_3$ system.

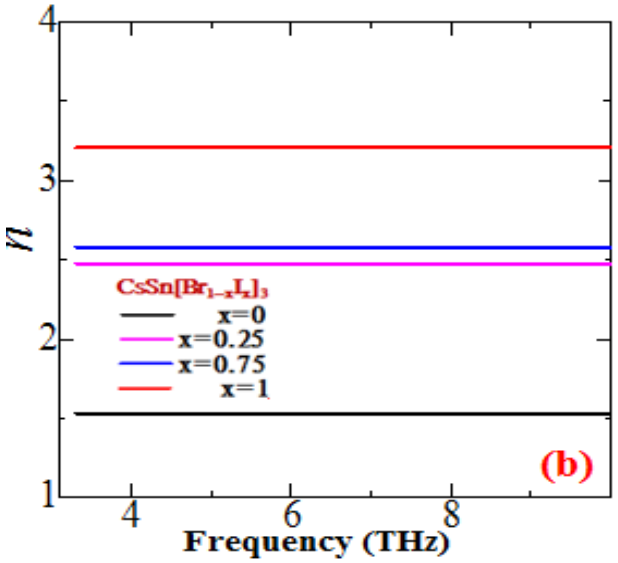

Figure 2. Frequency dependence of refractive index of the $CsSn[Br_{1-x}I_x]_3$ system.

The graphs shown in Figure 3 shows that the absorption coefficient of the samples of the $CsSn[Br_{1-x}I_x]_3$ system increases linearly with an increase in the frequency of terahertz waves.

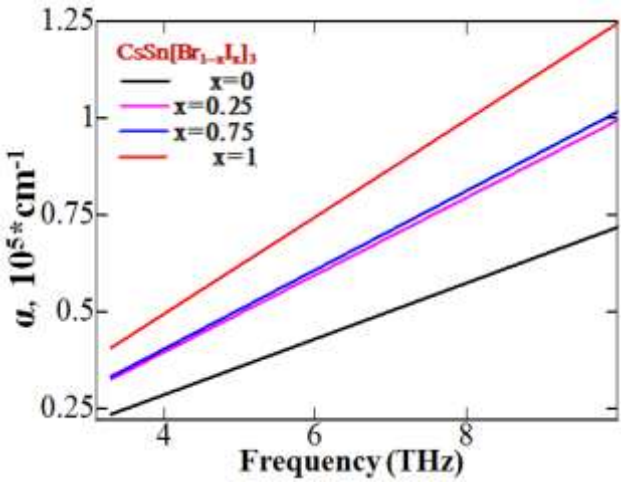

Figure 3. Frequency dependence of absorption coefficients $CsSn[Br_{1-x}I_x]_3$ system.

Slowly varying refractive indices, as well as high coefficients of absorption and photoconductivity of the members of the $CsSn[Br_{1-x}I_x]_3$ families in the terahertz range, make it possible to propose them as promising candidate materials for solar cells and other optoelectronic devices [59-61].

4 Conclusions

The frequency dependences of the optical properties of the $CsSn[Br_{1-x}I_x]_3$ system in the terahertz frequency range from 3 to 10 THz are characterized by the DFT method. The absorption spectra, photoconductivity, and refractive indices of these materials were measured and compared. The change in the refractive index of the studied materials was less than 3% in the frequency range from 3 to 10 THz, but the absorbing properties of these materials showed a very sharp frequency-dependent dependence. The results obtained in this study suggest that the studied materials are good candidates for solar cells operating with terahertz electromagnetic radiation, integrated circuits, and ultra-low-loss terahertz waveguides.

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The authors declare no conflict of interest.

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