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Using two-photon absorption to investigate multilayer semiconductor structures $GaSe_{1-x}S_x$

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Abstract

A new method for studying semiconductor multilayer structures used in the manufacture of optoelectronic devices, based on the analysis of photoluminescence spectra under two-quantum excitation, is described. This method makes it relatively easy in several cases to determine the band gap of individual layers, their thickness, and the degree of doping with impurities.

Keywords: Multilayer; multiphoton; laser; band gap; photoluminescence.

1. Introduction

In the manufacture of optoelectronic devices based on semiconductor layers with a variable bandgap E_g, it is very important to know such characteristics as the composition and, accordingly, the value of E_g, the thickness of the layers, the concentration of deep and fine impurities, which determine the nature of recombination processes and, consequently, the efficiency of the implemented

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structures. Concerning such structures for determining the composition or the value of E_g of individual layers, the most common methods at present are X-ray spectral microanalysis, which makes it possible to perform the most local measurements of the composition, as well as methods based on the detection of photoluminescence of the layers of the structure using layer-by-layer etching, the manufacture of an oblique section or a chip structure followed by single-photon excitation of photoluminescence [1, 2], the energy position of which is associated with the value of E_g. The listed methods have significant drawbacks, which consist in the violation of the integrity of the samples, the requirement for their careful preparation and significant time spent on measurements.

It is known that when semiconductors are exposed to laser radiation with $\hbar\omega < E_g$, due to the smallness of the multiphoton absorption coefficients, it is possible to create a significant concentration of electron-hole pairs in large volumes of matter. The study of recombination radiation arising from this method of creating nonequilibrium carriers is of considerable interest because luminescence in this case comes from the entire volume of the semiconductor and surface phenomena do not affect the observed regularities [3]. In our opinion, this feature of multiphoton absorption can serve as the main method for studying multilayer semiconductor structures. If the investigated multilayer structure is excited by light of sufficiently high intensity with a quantum energy $\hbar\omega$, such that the condition $E_g^{\max}/2 \le \hbar\omega < E_g^{\min}$ is satisfied, where E_g^{min} and E_g^{max} are the minimum and maximum values of the band gap in the sample, respectively, then, in this case, two-photon absorption of the incident light should take place, and due to the smallness of the absorption coefficient, almost uniform excitation of the structure under study occurs. In the case when the layer-by-layer change in the composition is sharp and corresponds to a change in E_g by a value greater than the width of the emission line, fully resolved lines with an energy at the maximum corresponding to E_g of the layer should be observed in the two-photon photoluminescence spectrum. The number of these lines, of course, should be equal to the number of layers in the structure.

Layered semiconductor structures $GaSe_{1-x}S_x$ are convenient objects for conducting this kind of research. The bandgap $GaSe_{1-x}S_x$ depending on the composition (x=0÷0.25) varies in the range ~ (2.020÷2.210 eV) [4-6], therefore the radiation of the 1st harmonic of the Nd:YAG laser ($\hbar\omega$=1.17 eV) should lead to two-photon absorption.

In this work, two-photon photoluminescence in $GaSe_{1-x}S_x$ multilayer semiconductor structures under laser excitation was studied experimentally.

2. Experimental technique

$GaSe_{1-x}S_x$ crystals were obtained by the Bridgman method. Since these crystals have a layered structure, it was possible to obtain very thin single-crystal plates by splitting them off from bulk samples. In this case, mirror optical surfaces were immediately obtained, which did not require special polishing. The multilayer structure was created by fitting onto a [7-10] optical contact. The essence of the method of landing on an optical contact is that two contacting semiconductor materials must have an almost "ideal" surface. In other words, for optical contact, the surface roughness of the contacting materials should not exceed a quarter of the wavelength of light. Of course, not all semiconductors meet these requirements. For this, the multilayer structure fabricated by fitting onto an optical contact must be characterized by the absence or a low concentration of states at the interface between the contacting phases. The $GaSe_{1-x}S_x$ crystals we used are ideal for this purpose. These crystals have a layered structure with a natural mirror surface, and this causes the chemical and adsorption inertness of natural cleavage surfaces. In this case, the samples were not subjected to mechanical and chemical treatment. In the manufacture of a multilayer structure, $GaSe_{1-x}S_x$ crystals with a thickness of 20÷100 μm were used, by simple splitting of a massive sample. Then the freshly chipped surfaces were pressed against each other and they held for several hours under pressure. The surfaces of the contacting samples interlocked due to intramolecular forces arising between them. Thin films of indium selenide (InSe) were used as the substrate. These crystals are also layered and their band gap is E_g=1.30 eV. The lowest layer was GaSe (E_g =2.02 eV), and the remaining layers were located with increasing E_g value, so the recombination radiation arising from two-photon excitation could leave the sample through wide-gap layers without absorption.

As a radiation source, we used a pulsed Nd:YAG laser with built-in generators of the 2-nd and 3-rd harmonics, designed to generate radiation with a wavelength of 1064, 532, and 335 nm, and a pulsed liquid laser with a wavelength tuning range in the range (594÷643) nm and (568÷605) nm. Pulse duration ~10 ns, repetition rate 20 Hz. The pulse power was 12 MW/cm^2. The radiation intensity was varied using calibrated neutral light filters. The optical absorption and luminescence spectra of $GaSe_{1-x}S_x$ crystals were studied using an automatic M833 double dispersion monochromator (spectral resolution ~0.024 nm at a wavelength of 600 nm), computer-controlled and a detector that records radiation in the wavelength range of 350-2000 nm. The experimental technique is similar to that described in [11, 12].

3. Experimental results and their discussion

We have studied a large number of samples based on solid solutions $GaSe_{1-x}S_x$ (x=0÷0.25) with different number of layers (from two to five). As mentioned above, the multilayer structure was created by fitting onto an optical contact. In the event that the implementation of areas of close contact of a multilayer structure is a consequence of partial displacement of the air gap or "envelopment" of clusters of adsorbed atoms under the action of the weight of the contacting semiconductors, it seems appropriate to study the effect of pressure on the photoconversion parameters of the optical contact.

Figure 1 shows the current-voltage characteristics of a structure consisting of two layers of InSe and GaSe at various applied external pressures. These layers were subjected to mechanical pressure directed perpendicular to the interface plane (along the c axis of symmetry of the InSe and GaSe crystals). The pressure varied in the range from 5 to 500 kg/cm^2. It can be seen that with an increase in pressure, the direct branch of the I–V characteristic improves, but starting from ~ 500 kg/cm^2, the characteristic deteriorates. Therefore, in the future, only those multilayer structures that are under pressure not exceeding 500 kg/cm^2 were studied.

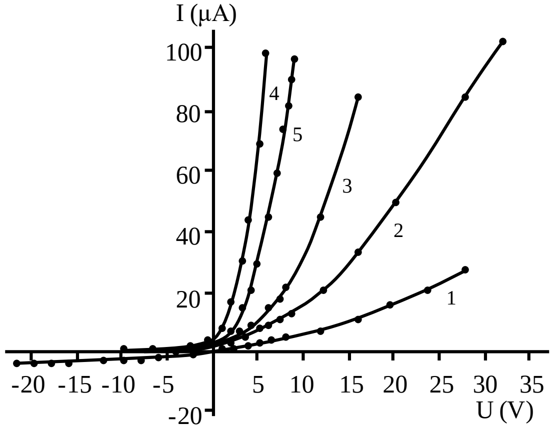

Fig. 1. Current-voltage characteristics of the InSe/GaSe heterojunction at various applied external pressures P (kg/cm^2): 1-50, 2-100; 3-300; 4–500; 5–600.

Figure 2 shows the photoluminescence spectra of the five-layer $GaSe_{1-x}S_x$ structure upon two-photon excitation by the first harmonic of the Nd:YAG laser (ħω=1.17 eV). As can be seen from the figure, the number of lines in the photoluminescence spectra coincides with the number of layers in the structure. To reveal the nature of the observed emission lines, we studied the transmission spectra of individual layers in the region of the fundamental absorption edge (Fig. 3).

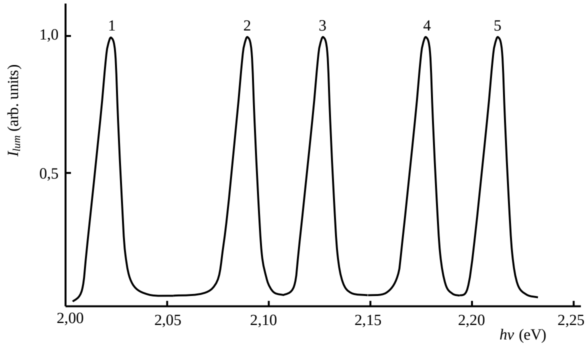

Fig. 2. Photoluminescence spectra of the $GaSe_{1-x}S_x$ multilayer structure, with different x values: 1-0; 2=0.5; 3=0.1; 4=0.2; 5=0.25.

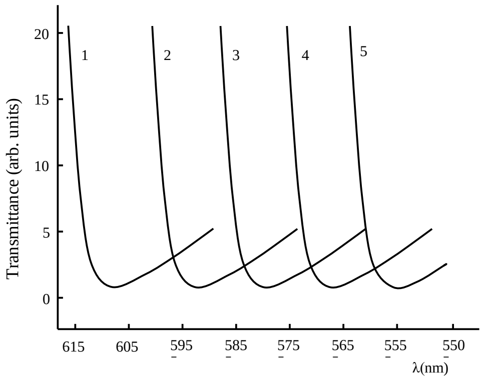

Fig. 3. Transmission spectra of individual layers of $GaSe_{1-x}S_x$ solid solutions in the region of the fundamental absorption edge with different x values: 1-0; 2=0.5; 3=0.1; 4=0.2; 5=0.25.

A comparison of Figures 2 and 3 shows a good agreement between the lines observed in the transmission spectra and in the luminescence spectra. In addition, we also measured the photoluminescence spectra of individual layers under the action of laser radiation with two-photon excitation. The photoluminescence of the individual layers belonging to each composition x coincides completely with the given luminescence line in the case of a multilayer structure. Thus, based on the studies performed, it can be argued that the band gap of each composition in the $GaSe_{1-x}S_x$ multilayer structure is 2.020 eV, 2.084 eV, 2.127 eV, 2.175 eV and 2.210

eV, respectively, for different x values: 1-0; 2=0.5; 3=0.1; 4=0.2; 5=0.25. It should also be noted that the band gap values found by us for $GaSe_{1-x}S_x$ solid solutions are in satisfactory agreement with the literature data [4-6].

As can be seen from Fig. 2, the radiative recombination intensities are practically the same throughout the entire volume of the multilayer structure, which allows us to assume that the line intensity in the luminescence spectra is proportional to the layer thickness. In this case, the ratio of the line intensities in the luminescence spectrum can be used to judge the relative thicknesses of the layers in the structure, and knowing its total thickness, one can determine the absolute values of the layer thicknesses. According to our definition, the total thickness of the layers turned out to be ~500 µm, which allows us to state that the thickness of each layer in the multilayer structure is equal to ~100 µm.

Along with this, we also determined the thickness of individual layers used in multilayer structures. Figure 4 shows the transmission spectrum of one of the used GaSe layers as an example. As can be seen from the figure, when light passes through plane-parallel layers, the thickness of which is commensurate with the wavelength of light, interference fringes appear. With a known refractive index (n~3 for GaSe), based on the wavelengths λ_m and λ_{m-1} corresponding to neighboring extreme in the transmission spectrum, the layer thickness can be determined [13]:

$$d = \frac{\lambda_m \lambda_{m-1}}{2n(\lambda_{m-1} - \lambda_m)}$$

Estimates show that the thickness of an individual GaSe layer is approximately equal to 100 µm, which is in satisfactory agreement with the values found from the luminescence spectrum of the multilayer structure.

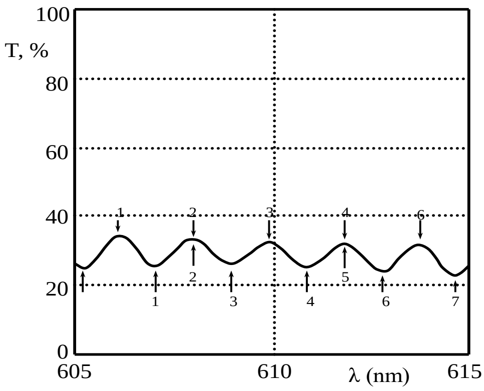

Fig. 4. Transmission spectrum of the GaSe layer. The thickness of the GaSe layer is d=100 µm.

4. Conclusion

A multilayer $GaSe_{1-x}S_x$ semiconductor structure was created by fitting onto an optical contact. The photoluminescence spectra of $GaSe_{1-x}S_x$ (x=0÷0.25) were experimentally studied under two-photon excitation by the first harmonic of the Nd:YAG laser (ħω=1.17 eV). The observed emission lines with energies of 2.02 eV, 2.084 eV, 2.127 eV, 2.175 eV, and 2.210 eV are associated with the band gap of the individual layers that make up the multilayer structures. The thickness of individual layers was determined from the intensity of radiative recombination of each layer and was found to be ~100 µm. It is shown that the two-photon absorption method proposed in this paper for studying a multilayer semiconductor structure can find several applications in the manufacture of optoelectronic devices and can also be useful in studying the influence of various technological factors on a multilayer structure.

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