ANNOTATION

Dissertations for the degree of Doctor of Philosophy (PhD)
6D060400 - Physics

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Features of photoinduced electronic processes in nanostructures based on graphene oxide

Topic relevance. Carbon is one of the most common chemical elements. Carbon materials which include graphite, diamonds, fullerenes, carbon nanotubes and graphene have been well known for a long time.

Graphene and its derivatives are currently being actively studied and used in the development of devices for optoelectronics, photovoltaics, and photocatalysis. Graphene with surface oxygen-containing groups is called graphene oxide. Graphene oxide and its modifications, in contrast to graphene, are a more convenient material for researchers, since it is easy to obtain and use for practical purposes.

Research in the use of graphene structures has recently led to a significant increase in publications in the field of synthesis and study of luminescent carbon and graphene dots. Compared to traditional semiconductor quantum dots and organic dyes, photoluminescent carbon-based quantum dots have high stability in aqueous solutions, chemical inertness, photostability, biocompatibility, and low toxicity.

Various authors have shown that the optical properties of graphene dots depend both on their structure and composition. For example, that the band gap of graphene dots affects the position of their luminescence maximum on the wavelength scale. This parameter can be modified by changing the oxidation state of graphene, as well as surface-edge states. In addition, it was demonstrated that during the chemical synthesis of graphene quantum dots, it is also possible to control the quantum yield of their luminescence by reducing carboxyl and epoxy groups on the surface of graphene oxide, which are centers of nonradiative recombination of electron-hole pairs. The authors of the work demonstrated that the doping of graphene dots with nitrogen atoms leads to a hypsochromic shift of the photoluminescence spectrum, while a decrease in defectiveness and the presence of functional groups lead to a bathochromic effect.

Synthesis of graphene dots is possible in two ways - "bottom-up" and "top-down". The first approach is based on the growth of a suitable precursor, such as small molecules of aromatic hydrocarbons or polymers, into nanosized graphene dots by reactions carried out by metal catalysis methods, or by hydrothermal or microwave synthesis, etc. The top-down approach refers to the direct splitting of bulk carbon materials, such as carbon black or graphite, into nanosized quantum dots, either by liquid stratification or by electron beam lithography. Also, this approach can be implemented using the laser ablation method. This excludes the use of additional reagents, which is especially important for luminescent objects, such as carbon and graphene quantum dots.

The luminescent ability of graphene dots can be increased using the phenomenon of localized plasmon resonance (LPR) of metal nanoparticles (NPs). The study of photophysical processes in luminophores and molecular ensembles passing near the surface of plasmonic nanoparticles is one of the topical problems of modern optical spectroscopy and luminescence. It is known that molecules placed near the surface of metal nanoparticles are subjected to the action of local electromagnetic fields. In this case, depending on the distance between nanoparticles and molecules, the rates of intramolecular transitions either increase or decrease. At present, works devoted to the interaction of graphene dots with plasmonic nanoparticles are just beginning to appear. The plasmonic effect can be used to enhance the luminescence of graphene dots. This will open up opportunities for their effective use in areas such as security inks, bioimaging, antibacterial and disinfection systems, heavy metal ion sensors, photovoltaic devices, and LEDs.

The dissertation aim is to study the features of photoinduced electronic processes in nanostructures based on graphene oxide and its derivatives.

The objects of research are nanostructures based on graphene oxide (SLGO), reduced graphene oxide (rGO), and nitrogen-doped graphene oxide (NGO), as well as plasmonic nanoparticles (NPs) of silver and gold.

The scientific novelty consists of the following:

1. The relationship between the conditions of obtaining, structural-morphological and optical properties of the obtained graphene nanostructures is established. A technique for the synthesis of graphene dots based on graphene oxide and its derivatives by laser ablation has been developed;

2. A systematic study of the influence of metal nanoparticles on photoprocesses in graphene nanostructures under changing conditions of their interaction has been carried out;

3. The effect of silver nanoparticles on the optoelectronic properties of graphene oxide films deposited by airbrushing has been studied. It has been shown that the resulting GO+Ag nanoparticles exhibit photocurrent values, photosensitivity, and detection ability comparable to those obtained by other groups for pure graphene.

The structure and scope of the dissertation. The structure of the dissertation work is determined by the tasks set and consists of an introduction, 4 sections, a conclusion, a bibliography. It is presented on 99 pages of typewritten text, illustrated by 50 figures, 16tables, contains a list of cited literature from 303 titles.

The main results include the following:

1. Structural and spectral-luminescent properties of nanodots based on graphene oxide and its derivatives are determined by the conditions of their production.

2. The quenching of fast fluorescence and the enhancement of the long-term glow of graphene dots near metal nanoparticles are due to the Förstrer energy transfer and the plasmon effect.

3. The doping of graphene oxide films with silver NPs leads to an increase in the photocurrent values and an improvement in their sensitivity and detecting ability.

Scientific and practical significance of the work: The results obtained can be used to control the optical properties of graphene nanostructures of various compositions. The results obtained can be used to create optical nanomaterials, photovoltaics, molecular electronics, and photocatalysis.

The data obtained show that the laser ablation method can be successfully used to obtain graphene dots with desired properties. The luminescent ability of graphene structures can be increased both by using the optimal conditions for their preparation and by using the plasmon effect of metal NPs.

Approbation of the work and publications. The main results of the work were reported and discussed at the following international conferences: 12th international scientific conference “Chaos and structures in nonlinear systems. Theory and Experiment" (Pavlodar, 2022); International Scientific Conference "Chemical Physics of Molecules and Polyfunctional Materials" (Orenburg, 2022); XV International Scientific Conference "Physics of the Solid State" (Astana, 2022).

Publications. Based on the results of the dissertation, 9 publications were published: 3 articles in journals included in the Thomson Reuters and Scopus database (1 article in Carbon Letters IF-4.5, Q2; 1 article in Materials Research Express, IF - 2.025, Q4; 1 article in Materials Today: Proceedings, Percentile, Scopus 38); 3 articles in journals recommended by the Committee for Control in the Sphere of Science and Higher Education of the Ministry of Education and Science of the Republic of Kazakhstan, and 3 publications in the materials of international conferences.