Using isotopic effect in nanostructures

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Abstract. This article is devoted to creating nanostructures based on the isotopic effect in solids. It is based in the renormalization of the band-gap value in case of changes in nuclear interaction, and has been experimentally proven in a broad range of semiconductors and dielectrics. In this paper we describe the isotopic method of opening the band-gap of the graphene and its implementation.
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Introduction

Currently, the most vital task is the searching for new materials that can improve the speed of operation of electronic and optoelectronic devices in order to increase the speed of transmitting and processing information in computing and telecommunication systems.

Operation speed of microprocessors, optical modulators and other technical devices depends on dimensions of the element base, electronic properties of the materials used, and manufacturing technologies. This requires transition of the element base to semiconductor nanostructures and nanotechnologies.

Obtaining nanostructures at the atomic level (carbon nano-tubes, fullerenes, graphene) can be considered a major breakthrough in creating new materials and technologies of their manufacturing. Further development in this direction requires deep study of structure properties and use of smaller dimensions. Such a useful property of the substances is the isotope effect [1-5]. A century has passed since the discovery of isotopes (chemical elements with the same electric charge, but with different number of neutrons and nucleus mass). This notion has become fundamental in nuclear, atomic and molecular physics. Within the last fifty years, the isotope effect has become one of the most powerful methods of studying the properties and the structure of the environment [6-9]. More and more research studies in solid-state physics are being performed with use of radioactive isotopes. Successful use of radioactive elements is known in medicine and geochronology. The leading role of physics of isotopes is known in studying the nature of nuclear interactions and reconstructing the process of nucleogenesis in the Universe.

Main part

Isotope effect

Modern scientific ideas show that the nuclear forces that interact between nucleons (neutrons and protons) include four types of interaction: strong intra-nuclear, electromagnetic, weak short-range, weak gravitational. Nuclear forces are charge-independent, i.e., interaction between neutrons or protons is analogous to interaction of neutrons and protons among themselves. The most direct method of changing nuclear forces is changing nucleons in the nucleus. So, changing the number of neutrons in the nucleus with a constant number of protons is the essence of the isotope effect [10].

In case of isotopic substitution, not only the phonon spectrum changes, but also the electron-phonon interaction, as well. Within recent decades, the detailed studies of the isotope effect has been performed on a large number of semiconductors and dielectrics [10,11,12].

Experimental evidence of the isotope effect existence is the Raman scattering spectrum. Ideal objects are semiconductor crystals with diamond-like structure ($C, Si, Ge, \alpha - Sn$). This is promoted by the fact that today there exist high-quality crystals grown from isotopically highly enriched material. As it is well known, materials with the diamond structure are characterized by triply degenerated states of phonons in the G-point of the Brillouin zone. These phonons are active in the Raman spectrum [6]. Figure 1 shows dependence of the shape and position of first-order line of optical phonons in a germanium crystal in various isotopic composition at the temperature of liquid nitrogen [8]. Position of the center of scattering lines is inversely proportional to the square root of reduced mass of an elementary cell. This means that a change in the isotopic composition (increasing the number of neutrons in the nucleus) leads to decreasing

energy and frequency of phonons. Another proof of existence of the isotope effect is renormalization of the band gap energy by adding a neutron in case of the diamond. So, in case of full replacement of C^{12} with isotope C^{13}, an increase in the band gap width occurs in the diamond by $14,7\text{MeV}$ [13].

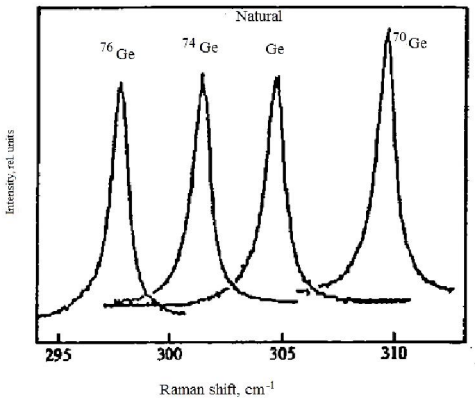

Fig. 1. Intensity of optical phonons scattering in germanium crystals

Semiconductor graphene

The most promising material for creating nanostructures is graphene, being a single-atom layer of carbon. Graphene is a semi-metal where conduction and valence bands touch at the K[th] point of the Brillouin zone [14-19]. In order to create element base for electronic devices using graphene, this material should become a semiconductor. Scientists have already proposed carbon nano-tubes and graphene nano-bands that have semiconductor properties. Another way to open the band gap in graphene is straining samples. Study of Raman spectra with applying uniaxial stress to graphene samples showed that in this case opening of band gap energies can occur in the K[th] point of the Brillouin zone. As shown in work [20], in such objects, 2D and G bands in the combinational spectra of light scattering experience red shift by the amount of 27.8 and $14.2\ cm^{-1}$, respectively, with the load equal to 1 % of the respective maximum for inter-atomic bonds. Changes in frequency of scattered light are caused by stretching inter-atomic bonds in carbon. In their calculations, the authors showed that these shifts correspond to opening band gap of $300MeV$. Results of calculation and experiment with stressed samples made by the authors [20] and presented in Figure 2 show good agreement. Thus, the method of uniaxial stress can be very successful in creating semiconductor graphene.

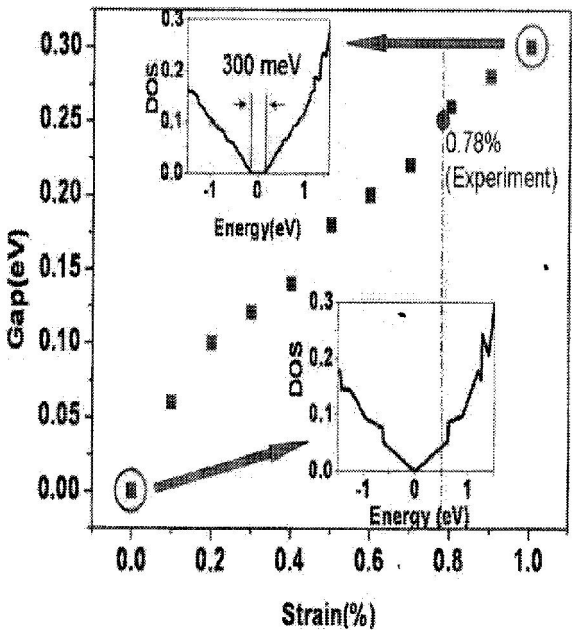

Fig. 2. Opening band gap energies of a stressed graphene sample.

Dots - experiment, solid lines - theoretical calculation (data from work [21])

Another effective method of obtaining semiconductor graphene could become the method of isotopic renormalization of elementary excitation energy in solids. The first experimental study of isotope effect influence on graphene thermal conductivity was reported by the authors of work [21]. Thermal conductivity K at temperature $320K$ of isotopically pure graphene $C^{12}(0,01C^{13})$ was slightly higher than $4000W/mK$, which is approximately two times larger than that in diamond [9]. Thermal conductivity increases twice for samples consisting of $50\%C^{12}$ and $50\%C^{13}$. Of interest are Raman scattering spectra of graphene samples obtained at room temperature (Fig. 3), as set out in work [21]. As in the case of isotopically mixed diamond [9, 22], the frequency of an optical phonon in the center of the Brillouin zone varies as $\omega \approx M^{-\frac{1}{2}}$, thereby making the shift of scattering line approximately proportional to $(12/13)^{-\frac{1}{2}}$ towards the lower frequencies, as concentration of the isotope C^{13} increases. Experimental variance of optical phonon frequencies in graphene (from $99.2\%\,C^{13}$ to $0.01\%\,C^{13}$) marked in Fig. 3 by the lower maximum is $64cm^{-1}$, which is in good agreement with theory

[21]. It is important to note that a similar shift for the optical phonon of the diamond (with sp^3 - bond) is equal to $52{,}3cm^{-1}$ [22], which is also consistent with the above noted mass ratio of isotopes. Considering a softer bond in graphene (sp^2 instead of sp^3 in diamond) in work [23], it was estimated that the isotope effect will lead to creation of semiconductor graphene, i.e., to obtaining a band gap of several hundreds of MeV.

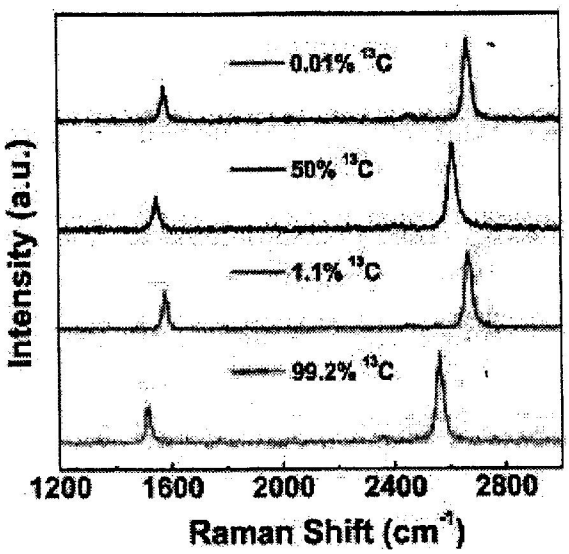

Fig. 3. Raman scattering spectra in graphene with different concentrations of the ^{13}C isotope at room temperature (data from work [21]).

Thus, by changing the strong (intra-nuclear) interaction, namely, by adding one neutron, a renormalization of the electron band gap energy and band gap opening occurs, which is equivalent to creating semiconductor graphene. The most promising method for manufacturing isotopic graphene can be the method of neutron transmutational doping. It is based on industrial technologies of neutron exposure that has been well-developed during several decades. Thermal neutrons and the reaction of their absorption by the target nuclei may be of practical interest for graphene samples exposure. Modern nuclear technologies make it possible to obtain collimated neutron fluxes that have sufficiently high intensity ($10^{17}\frac{neutrons}{cm^2 s}$), which will ensure necessary performance and high quality of graphene nanostructures [1].

Conclusion

The isotopic composition of initial chemical elements greatly affects physical properties of the final material and optoelectronic characteristics of various devices. Besides the effects with thermal conductivity of diamond described above, the following facts can be mentioned. Silica purification of heavy isotopes improves operating speed of microprocessors, reduces kilometric attenuation of optical waveguides, and improves efficiency of photo detectors, optical modulators, and semiconductor lasers [1]. The main reason for this phenomenon is significant reduction of the number of channels of electron scattering within the crystalline grid, which reduces optical losses and increases mobility of charge carriers. Therefore, the use of the isotope effect for creating nanostructures of silicon, germanium, graphite, and other semiconductors will make it possible to design new high-quality materials with preset characteristics [1]. As is well known, constants of crystalline grids of isotopes of the same material are virtually the same. Therefore, one can obtain "ideal heterojunctions" in quantum structures, e.g., by combining layers of different isotopes of the same chemical element. Purity of initial material, together with modern technologies (thermal neutron irradiation, molecular-beam epitaxy, etc.) make it possible to accomplish a technological breakthrough in manufacturing element base for electronic and optoelectronic devices.

Conclusions

1. Increasing operating speed of electronic and optoelectronic devices is related to improving the element base: reducing size and searching for new materials.
2. To obtain new materials and create nanostructures on their basis, isotope effect can be used, based on physical difference between substances from various isotopes of the same chemical element, namely, the constant of electron-phonon interaction, band gap width, refraction index, etc.
3. The most promising material for the element base of electronic devices is graphene. In order to use it, the technology of obtaining semiconductor graphene (opening band gap) is required.
4. As it was noted in work [20], the band gap can be opened by mechanically stressing the sample accompanied by changing optical phonons frequency (the red shift of the $2D, G$ bands).
5. Similar changes in the optical phonon modes were observed in case of changing the isotopic composition of graphene [21]. This fact makes it possible to relate changes in crystalline grid

of graphene that occur as a result of increasing concentration of the C^{13} isotope to band gap opening.

6. As is well known, one can increase the percentage of the heavier isotope in the sample using the reaction of thermal neutron absorption, which underlies the industrial process for manufacturing microelectronic devices (neutron transmutation doping).

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