

New optimal schemes for the laser photoionization technology for control and cleaning the semiconductor substances and preparing the films with pure composition at the atomic level and the schematic diagram of a new corresponding equipment are presented. The computer modeling of the optimal scheme for preparing the films with pure composition with a possibility to create 3D heterostructural superlattices (layers of  $\text{Ga}_{1-x}\text{Al}_x\text{As}$ and GaAs) is carried out.

### 1. Introduction

The development of efficient methods for obtaining the especially pure substances or their control and cleaning from admixtures (cf. [1, 2]) is considered as one of the actual problems of the modern technology of production of semiconductors and other materials. In particular, we mean the methods to control the contents of Al, B, admixtures in Ge, and the other third group acceptor elements admixtures at a level of  $10^{-8} - 10^{-10}\%$ . In some cases, the traditional analytic methods [1] or their modifications can provide a similar sensitivity. However, its sensitivity is limited by a level of  $10^{-7}\%$ . Selective photophysics methods [2–9] allow developing a basically new approach to the technologies of obtaining the pure substances at the atomic level.

The basis for its successful realization is, first, carrying out the optimal multistep photoionization schemes for different elements and, second, the availability of enough efficient lasers with high average power in the UV and visible ranges. The standard laser photoionization scheme can be realized with the use of processes of the multistep excitation and ionization of atoms by a laser pulse. The scheme of selective ionization of atoms, based on the selective resonance excitation of atoms by laser radiation into states near the ionization boundary and the further photoionization of excited states by additional laser radiation, has been first proposed and realized by Letokhov *et al.* (cf. [2]). This scheme is of great interest for the laser separation of isotopes and nuclear isomers. However, a significant disadvantage of the two-step selective ionization of atoms by the laser radiation method is a great difference between the cross-sections of resonant excitation  $\sigma_{\text{exc}}$ and photoionization  $\sigma_{\text{ion}}([\sigma_{\text{exc}}/\sigma_{\text{ion}}]>10^4\div10^8)$ . This requires to use a very intensive laser radiation for the ionization of excited atoms.

The situation is significantly simplified for autoionization resonances in atomic spectra, but the detailed data on characteristics of these levels are often absent. Here, the main problems are related to difficulties of the theoretical study and calculation of the autoionization resonance characteristics. In [10, 111], we proposed new optimal schemes for the laser photoionization method of separating the heavy isotopes and the nuclear isomers and executed the appropriate computer-based modeling. They are based on the selective laser excitation of isotope atoms into Rydberg states and the mechanism of the further dc electric field ionization. To carry out the modeling of parameters of the optimal scheme of the sensing of U and Tm isotopes (nuclei), we have used new highly efficient quantum models of elementary atomic processes, the optimal laser action method, and the density matrix formalism (cf. [9–21]).

In this paper, we develop new optimal schemes for the laser photoionization technology for the control over and the cleaning of semiconductor substances and the preparation of films with pure composition at the atomic level and present the schematic diagram of a new

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Fig. 1. Scheme for preparing the films with superpure composition during the sedimentation of ions of the A<sup>+</sup> kind which are obtained by the optimal method of selective photoionization of A atoms in a beam containing the other atoms: 1 – source of the atomic beam; 2 – vacuum chamber; 3 – collector of non-selective ions; 4– diaphragm; 5 – laser ray for the first-step excitation; 6 – laser ray for the second-step excitation into Rydberg states and the ionization by an external electric field; 7 – deflecting electrodes; 8– sublayer; 9 – cold sublayers to freeze atoms; 10 – laser ray to vaporize the substance

corresponding equipment. We have carried out the computer modeling of optimal schemes of the laser photoionization method, in particular, a scheme of the creation of 3D heterostructural superlattices (layers of  $\text{Ga}_{1-x}\text{Al}_x\text{As}$  with width 10 Å and GaAs of 60 Å). New models of optimal realization of the first-step excitation and the further ionization of  $\text{Ga}^+$  ions into Rydberg states by an electric field are used and optimal parameters of the photoionization process are detertmined. Preliminary results for Al and others ions have been presented earlier [12, 14].

# 2. Method of Laser Phoionization of Atoms. Schematic Diagram of a New Equipment

In the classic scheme, the laser excitation of admixture atoms is realized in several steps: atoms are resonantly excited by laser radiation, and then the photoionization of excited atoms is realized (cf. [2–8]). In this case, the photoionization process is characterized by relatively low cross-sections  $\sigma_{ion} = 10^{-17} - 10^{-18} \text{ cm}^2$ , and one must use the powerful laser radiation on the ionization step. An alternative mechanism is the transition of atoms into Rydberg states and the further ionization by an electric field. As a result, the requirements to the energy of an ionizing pulse are decreased by several orders. A possible device for preparing the films with pure composition by means of the two-step selective ionization of atoms was proposed by Letokhov [2]. Such a scheme was not experimentally verified. However, it is obvious that the two-step laser ionization scheme is not optimal. The main feature and the innovation of our scheme is related to using a pulse of the electric field on the last ionization step [8–13]. Such an optimal scheme can be used as a basis for devices aimed at preparing the films with superpure composition during the sedimentation of ions of the  $A^+$  kind which are obtained by the optimal method of selective photoionization of A-kind atoms in a beam containing also the other atoms. This point is a key one for our work. In Fig. 1, we present a possible scheme for preparing the films with super pure composition during the sedimentation of ions. In fact, our scheme generalizes the known Letokhov's scheme [2] but is significantly more efficient.

The widespread method to fabricate complex heterostructures of the superlattice type is the method of molecular epitaxy. At the same time, it is worth to note that it allows creating only 1D superlattices (the wellknown example is layers  $Ga_{1-x}Al_xas$  of 10 Å in width and GaAs of 60 Å in width with the full number of layers of about 100. The 3D heterostructural superlattices can be created by means of the electromagnetic focusing and deflecting systems with the use of photoionic beams of Ga<sup>+</sup>, Al<sup>+</sup>, and As<sup>+</sup>.

As was first indicated in [2] (see also [9, 14]), the opportunity of a spatial control of the sedimentation of ions is of great importance for promising semiconductor atomic technologies of materials. But the key point is the creation of an efficient technology (profitable from technological and commercial viewpoints).

# 3. Computer Modeling of Optimal Schemes within the Laser Photoionization Method

Let us consider the laser photoionization method to obtain Ga<sup>+</sup> ions following [14] and an optimal scheme for preparing the films with pure composition. The scheme of atomic transitions is as follows:

$$4p^2P_{3/2} \rightarrow (\lambda_1 = 417.2 \text{ nm}) \rightarrow 5s^2S_{1/2} \rightarrow$$

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Fig. 2. Results of the modeling of the Ga separation process from a Ga-X mixture by the photoionization method ( $\delta$ +dashed – laser pulse optimal form; curves  $x_1$  and  $x_2$  correspond to the populations of the ground and excited states);  $\tau$  – dimensionless time

$$\rightarrow (\lambda_1 = 420 \div 440 \text{ nm}) \rightarrow np^2 P_{1/2}, \quad n = 14 \div 70.$$
 (1)

The modeling of optimal parameters for the last process, i.e. the ionization of Rydberg states by electric field, can be carried out on the basis of methods developed in [4, 9–12, 18–21]. In Fig. 2, the results of both a numerical modeling of the separation of Ga atoms from a mixture on the basis of the photoionization method and the solution of the corresponding system of differential equations [9, 13, 14] are presented. The following definitions are used:  $\delta$ +dashed line corresponds to the optimal form of a laser pulse, and curves Iand 2 correspond to the populations of the ground and excited states of Ga. The  $\delta$ -pulse provides the maximally possible level of excitation (the excitation degree is about ~0.25; in the experiment [2] with rectangular pulses, this degree was ~ 0.1).

In order that the parasite processes such as spontaneous relaxation, resonant excitation transfer, and resonant recharging cannot change the achieved excitation level during a little time, the last step of the process is the ionization of excited atoms by an electric-field pulse [9] (the field strength is 8.8 kV/cm). To get a high level of the optimality, the electric field has to be switched on during the time interval which is less than the radiative decay time of excited states. In Fig. 3, we present the calculated dependence of the ionization velocity for highly excited atoms of Ga on the electric field strength for states with quantum numbers  $n = 10 - 16, m = 0, n_2 = n - 1$  [14]. The dashed line corresponds to the radiative decay rate.



Fig. 3. Calculated dependence of the rate of ionization for highly excited atoms of Ga on the electric field strength for states with quantum numbers n = 10 - 16, m = 0,  $n_2 = n - 1$ 

From the physical viewpoint, the decay of Ga atoms and ions in highly excited states demonstrates the properties of the hydrogen-like systems at the qualitative level. But, there is a quite significant quantitative difference. We have found that the ionization rate for states with n > 14 is more than the radiative decay rate in an electric field with strength E less than 15 kV/cm. Our estimate for the Ga atom ionization cross-section is  $1.5 \times 10^{-13}$  cm<sup>2</sup> that is higher than the corresponding cross-section of the ionization by a laser pulse in the two-step photoionization scheme [2] ( $\sim 10^{-17}$  cm<sup>2</sup>).

Using a  $\delta$ -pulse provides a quick ionization, but the ionization yield will be less than 100% because of the sticking on intermediate levels. The experimentally obtained dependence of the critical ionization field strength E on the effective quantum number  $n^*$ is usually approximated by the simple theoretical dependence  $E_{\rm cr} = (2n^*)^{-4}$ . Our calculation showed that this is a very approximate estimate, and only the consistent quantum calculation (cf. [4, 9]) provides the excellent agreement with experiment. In any case, the laser photoionization scheme with ionization by an electric field (with the optimal collection of energetic and radiative parameters: pulse form, duration, energies of laser and electric field pulses, *etc.*) could provide the significantly higher yield and the efficiency of the whole process than the other known schemes [2].

#### 4. Conclusions

So, we have presented a new optimal scheme for the laser photoionization technology for the control over and the cleaning of semiconductor substances and for the

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preparation of films with pure composition at the atomic level.

We have considered a new optimal equipment scheme for preparing the films with superpure composition during the sedimentation of ions of the  $A^+$ kind which are obtained by the optimal method of laser and electric field photoionizations of A-kind atoms in a beam containing also the other atoms. The computer modeling of the optimal scheme for preparing the pure films with a possibility to create 3D heterostructural superlattices (layers of  $Ga_{1-x}Al_xAs$  and GaAs) is carried out.

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#### НОВІ ОПТИМАЛЬНІ СХЕМИ ЛАЗЕРНО-ФОТОІОНІЗАЦІЙНОГО МЕТОДУ ОЧИЩЕННЯ НАПІВПРОВІДНИКОВИХ РЕЧОВИН ТА ВИГОТОВЛЕННЯ ПЛІВОК ОСОБЛИВО ЧИСТОГО СКЛАДУ НА АТОМНОМУ РІВНІ

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Резюме

Розроблено нові схеми лазерно-фотоіонізаційної технології контролю та очищення напівпровідникових матеріалів і виготовлення плівок особливо чистого складу на атомному рівні. Наведено нову схему відповідної установки. Виконано комп'ютерне моделювання оптимальної схеми виготовлення плівок особливо чистого складу на прикладі 3D гетероструктурних суперґраток (шари  $Ga_{1-x}Al_xAs$  та GaAs).