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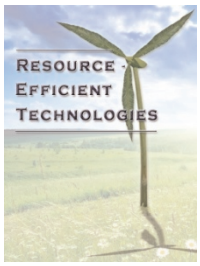

Research Paper

POTENTIALITIES OF FAST ION NEUTRALIZATION AT GRAZING INCIDENCE ANGLES FROM CRYSTAL SURFACES FOR DEVELOPMENT OF NEW GENERATION OF UV LASER SOURCES

P.A. Aleksandrov, S.S. Fanchenko*, S.G. Schemardov

National Research Center Kurchatov Institute,
1 Akademika Kurchatova pl., Moscow, Russian Federation, 123182

Abstract

The paper considers processes of electron capture by fast ions scattered from metal surfaces at grazing incidence angles. It discusses the possibilities of excitation of high atomic levels in neutralization process and laser generation. The Doppler compression effect for ion beams is shown to be rather essential for laser generation in the ion energy range of several tens keV. It is shown that the four-level laser generation scheme is the preferable one. The values of critical currents for laser generation are evaluated. The experimental setup for ion beam neutralization is observed. The experimental results on nitrogen ion beam neutralization on silicon (001) wafers at grazing incident angles are presented. The dependence of the neutralization coefficient on the grazing incidence angle is measured, a good coincidence between peak maximum and Lindhard angles is demonstrated. It is shown that in case of neutralization without special vacuum surface cleaning the neutralization coefficient makes approximately 30 % for ion energy of 40 keV. The one-electron resonant neutralization to nitrogen 2P1/2, 2D3/2, 4S3/2 atomic terms is discussed. The mechanism of resonant coherent excitation of fast nitrogen atoms in front of a Si (001) surface is observed, the resonant energy being estimated about 70 keV.

Key words: ion sources, optics, lasers, Doppler compression, resonant neutralization, Lindhard angle.

1. Introduction

To our knowledge, the idea to use the atomic levels inversion produced in the process of fast ion neutralization from metal surfaces under grazing incidence conditions for two-step laser generation was proposed in 1983 [1], the laser generation scheme being proposed earlier [2].

It was shown that the ion neutralization process could be used to generate a long-lived reservoir of Li $1s2s2p\ ^4P^0_{5/2}$ with enough density of metastables. As a result, after laser-induced transfer to Li $1s2p\ ^{22}P$ a strong lasing to Li $1s^22p\ ^2P^0$ at 20.7 nm should occur.

It was expected that after the reflection of an incoming beam of 70 keV Li^{2+} the yield of neutral Li $1s2s2p\ ^4P^0$ should be about 40 % so that the requirements for laser generation are satisfied. Nevertheless, it was not confirmed experimentally. One of the reasons could be the consideration of only one-electron resonant neutralization neglecting other processes, such as Auger ionization, de-excitation and ground state neutralization, which could strongly influence the inversion of resulting atomic levels.

Firstly, only one-electron resonant neutralization process was considered [3–5]. Later, for He ion beams with energy of 2 keV all Auger processes were considered in details in [6]. The electron capture to atomic orbitals starts from distances comparable with atomic orbital radius, so that the 2s level of

* Corresponding author. National Research Center Kurchatov Institute, 1 Akademika Kurchatova pl., Moscow, Russian Federation, 123182E-mail address: Fanchenko_SS@nrcki.ru

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He ion is filled at first. This is followed by the processes of Auger de-excitation to the ground state and resonant ionization with the following Auger neutralization to the ground state. As a result of these kinetic processes for 2 keV ions, practically all reflected neutral atoms occur in the ground state. No inversion is produced in this case and the laser generation is impossible. The problem of Auger de-excitation mechanism in the process of emission of optical radiation from He particles scattered at a monocrystalline surface at grazing incidence in the visible spectra range was considered in [7]. To change this situation we propose to use He ions with energies of several tens keV, for which (due to higher velocities) the de-excitation of the initially excited level will not be completed during the scattering process. The experiments for hydrogen atoms neutralization on Al surfaces with 25 keV energy showed that the population of hydrogen levels depends on n^{-6}, where n is the principle quantum number [8]. In this case the excited levels survive though no inversion is produced. The Auger neutralization and de-excitation rates strongly depend on the electron level structure, so one can expect that they will be lower in case of He ions due to the unique energy level intervals in He. Neglecting the splitting of 2S and 2P levels, the main yield of neutral He atoms should be in the following states: 2^1S, 2^3S, 2^1P, 2^3P, with intensities proportional due to spin and orbital multiplicities to 1/16; 3/16, 3/16 and 9/16, accordingly. The metastable state 2^1S could be used for a two-step laser generation as in case of Li-atom discussed above, while 2^1P state could be used for the generation of vacuum UV radiation at 58.4 nm within a one-step process.

2 Theory

The criterion for laser generation is as follows [9]:

$$\frac{L\lambda^2 n^-}{8\pi\tau_0 Д\upsilon} \sim 10, \quad (1)$$

where L – cavity length; λ – wavelength; n^- – media inversion density; τ_0 – radiation life time; $\Delta\upsilon$ – bandwidth.

The bandwidth for fast ion beams presents a particular interest [10]. The velocity distributions of ions can be changed through acceleration in electric fields thus contributing to direct reduction of velocity profiles. The distribution of initial energy is conserved during acceleration so the Doppler compression may be described as follows:

$$Д\upsilon = \frac{\upsilon_0 ДE_T}{mvc} = \frac{Д\upsilon_D \mathrm{v}_T}{v}, \quad (2)$$

where $\Delta\upsilon_D$ – common temperature bandwidth with regard to the Doppler broadening value; v – ion velocity after acceleration; v_T – thermal velocity; c – light velocity; υ_0 – center of the frequency bandwidth. At 10 keV, the velocity distribution v_T/v in the profile is about $1.5\cdot10^{-3}$, so that for such and larger energies only homogeneous radiation broadening should be considered. In that case, the laser generation criterion is rewritten as follows:

$$\frac{L\lambda^2 n^-}{2\pi} \sim 10. \quad (3)$$

Estimating L as $\tau_0 v_i$ and n^- as $0.1\cdot j/ev_i$, where j – current density; e – electron charge, we obtain a relatively simple criterion for critical current density for laser generation:

$$j_c \sim \frac{600e}{\lambda^2\tau_0}. \quad (4)$$

This estimation for current density is rather evident from dimension criteria though the right value of the coefficient may differ several times. For He main 2P-1S transition the critical current density makes $j_c \sim 6\cdot10^3$ A/cm^2. For other transitions the radiation lifetime could be much greater (for 10P-1S transition with 50.7 nm wavelength this time is 113 times greater and for metastable 2S-1S transition the lifetime is about 0.02 s).

3 Experimental design

The above estimation confirms that the one-step laser generation is almost impossible and only two-step process with excitation of metastable levels with further laser induced generation is possible for available current densities. For the case of metastable excited state 2S the cavity length is limited by experimental setup since for high-energy atoms their path before irradiation exceeds kilometers. As a result, the critical current densities depend on L value. Using mirrors, the path could be enlarged many times thus lowering the critical current densities by several orders down to some tenths of A/cm^2, being rather realistic assessment. However, this case causes the problem of inversion. If the initial population of 2P levels is 3/4 due to orbital multiplicity, then at 0.6 ns the 2P levels will be depopulated to the ground state due to radiative transition and no inversion will occur for the second stage of laser generation. But due to energy-level splitting of 0.6 eV the ratio of populations of 2S and 2P levels could be different, which shall be confirmed experimentally. Due to uncertainties of all rates of Auger neutralization to the ground state and Auger de-excitation process it is rather difficult to make direct calculations.

Hence, we are going to check neutralization of He ions with the energies in the range of 20–80 keV on Mo single crystal in order to obtain the populations of excited levels using ILU-4/17 ion source (Fig.1).

The technical characteristics of ILU-4/17 experimental setup are the following: energies in the range of 20–80 keV, available ions: H, He, Si, O, N. It is shown that the four-level laser generation scheme is much more preferable than the three-level one in case of fast ion neutralization at grazing incidence angles from metal surfaces and could be implemented using the experimental setup similar to ILU-4/17. Due to technical problems with He ion beam the test experiments were performed for neutralization of nitrogen ion beam using ILU-4/17. Though the neutralization of He ions on Mo surface is more interesting for optical applications, the main aspects of the process could be checked by neutralizing the nitrogen ions on silicon surface (nitrogen beam is more stable than the helium one and the silicon wafer surface is much smoother than Mo single crystal one).

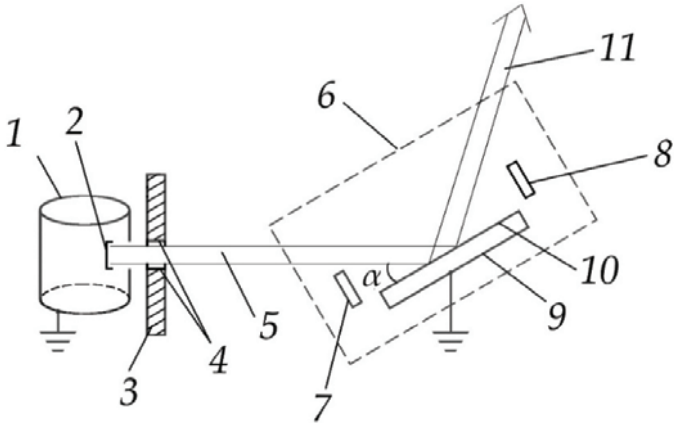

Fig. 1. *ILU-4/17 experimental setup: 1 – gas cell; 2 – slits; 3 – ion optical system; 4 – aperture; 5 – grazing incidence ion beam; 6 – laser resonator; 7, 8 – mirrors, 9 – neutralization region; 10 – crystal surface; 11 – excited neutrals*

The importance of the neutralization process at grazing incidence angles is that at angles of incidence less than the critical angle (Lindhard angle) the ion interacts with the entire atomic plane and not with a separate atom in the lattice, and thus practically does not sputter the surface. At angles greater than the Lindhard angle, the ion interacts with the nearest plane surrounding the lattice and substantially sputters it.

The Lindhard angle is determined by a well-known equation [11]:

$$\theta_L = \sqrt{\frac{E_c}{E_0}}, \tag{5}$$

where E_0 – ion energy; E_c – critical energy determined by the following equation:

$$E_c = 2\pi n_s a e^2 Z_1 Z_2, \tag{6}$$

where n_s – surface atomic density; e – electron charge; Z_1, Z_2 – incident and lattice ion charge numbers; a – effective screening radius determined in the simplest Thomas-Fermi approximation by the equation:

$$a_{TF} = \frac{0{,}8854 a_0}{\sqrt[3]{(Z_1^{1/2} + Z_2^{1/2})^2}}, \tag{7}$$

where a_0 – Bohr radius.

The experiments on neutralization of ion beams interacting with metal surface were conducted using the ILU-4/17 accelerator. A special circuit was installed in the receiving chamber of the installation to measure the degree of neutralization of the ion beam. The ion beam leaves the ion source, passes through the magnetic separator and hits the diaphragm. Further, it interacts with the surface of a sample attached to the rotating holder and hits the receiving device that measures the degree of neutralization of the ion beam, as shown in Fig. 2.

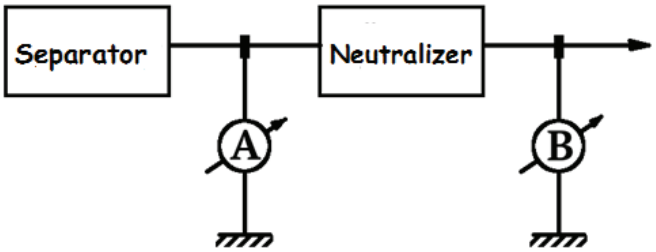

Fig. 2. *Diagram of the device for measuring the neutral and ionic components of the atomic beam (A and B – current values after the separator and the neutralizer, respectively)*

The measuring device action is based on the principle of secondary electron emission in the interaction of ion beams with materials, in particular with metals. A certain amount of secondary electrons will be emitted from the metal plate-bench measuring the incident beam, which is determined by the secondary emission coefficient ɣ [12]. This coefficient depends on mass and energy of the incident ion (atom), the angle of incidence, the material of the frame, etc. However, in the absence of other side factors with fixed implantation parameters the coefficient is constant for a given ion-frame pair. Ion accelerators for measuring ion current widely use benchmarks in a weak transverse magnetic field due to their simplicity and reliability. It is known that charged particles in a transverse magnetic field move along circular trajectories. For a constant magnetic field B = 0.1 T the radius of such trajectory is about 0.1 mm. Therefore, all the secondary electrons emitted from the surface of the frame located in this magnetic field will return back, and the frame will only fix the ion component of the incident beam. In the absence of a magnetic field, the ions and atoms incident on the frame will cause the secondary electron emission, and the frame current will make a sum of the ion current and the current of the emitted electrons. Since both ions and neutral atoms with one energy lead to the emission of

the same number of electrons, measurements of the beam current inside and outside the magnetic field make it possible to determine the fraction of the neutral component in the beam. Thus, the current without a magnetic field is presented as follows:

$$I_w = I_i + e_i + e_n. \quad (8)$$

where I_i is the current ion beam component; e_i – secondary electron current due to ions; e_n – secondary electron current due to neutral atoms.

Taking into account the same secondary electron coefficient ɣ the equation (8) may be rewritten as follows:

$$I_w = I_i (1+\gamma) + \gamma I_n. \quad (9)$$

where I_n – neutral atom beam component. Evidently,

$$I_m = I_i. \quad (10)$$

Measuring I_w and I_m, one can easily obtain I_i and I_n values from equations (9), (10) (the measured experimental value of ɣ is 0.83).

For the beam analysis, a receiving device was constructed, which allowed to move the measuring frame in a vacuum and translocate it into the transverse field of permanent magnets.

Test experiments on neutralization of the ion beam were carried out for 40 keV nitrogen ions at a current value of 80 μA. A single-crystal silicon (100) wafer was used as a sample. According to equation (5), the Lindhard angles for 40 keV nitrogen ion reflection from silicon (100), (111), and (110) surfaces are 2.6°, 2.8° and 3.1°, respectively.

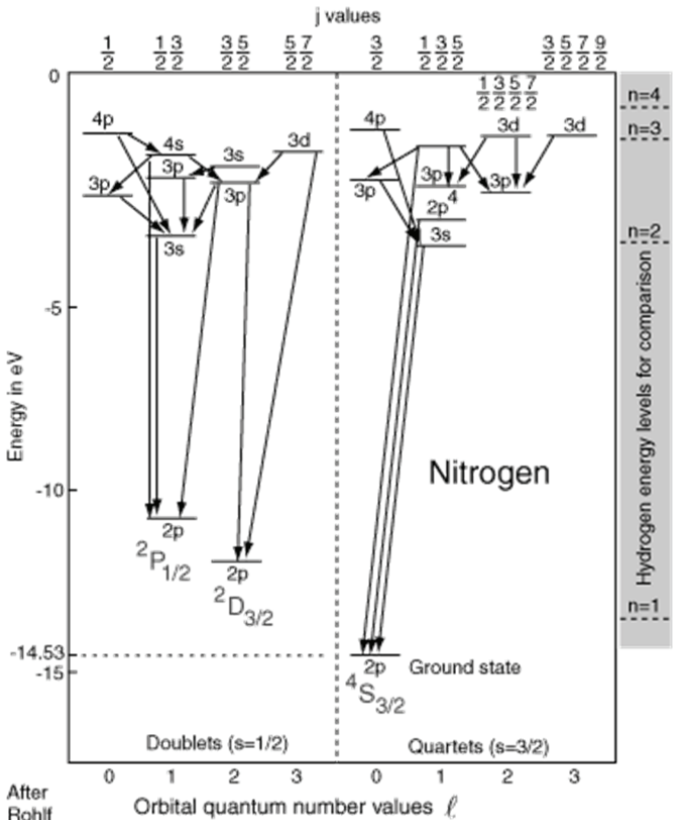

Fig. 3. *Nitrogen energy levels [13]*

Preliminary the silicon crystal surface was cleaned with alcohol, while the special ion beam cleaning was not performed. For better understanding, one should take into account the structure of nitrogen energy levels and silicon energy bands. The energy levels of nitrogen atom are shown in Fig. 3 [13]. However, positioned near the surface the levels shift due to image forces. It is known that for a neutral atom the repulsive interaction between the electron and the image charge of the core is stronger than the attraction to its own image, which is illustrated in Fig. 4 for three lowest nitrogen energy levels. The value of the electron affinity in silicon is 4.1 eV, the band gap value – 1.1 eV and the width of the valence band – 8 eV, meaning that only three lowest energy levels $^2P_{1/2}$, $^2D_{3/2}$, $^4S_{3/2}$ in the nitrogen atom may interact in the resonant one-electron neutralization.

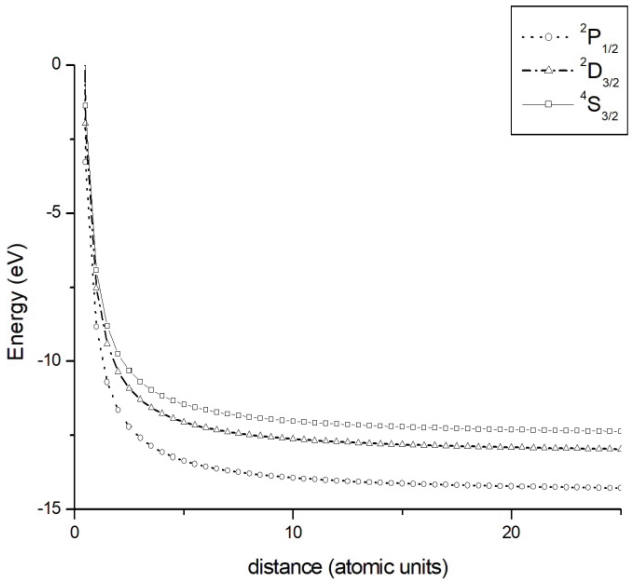

Fig. 4. *Dependence of the nitrogen three first energy levels on the distance from silicon wafer*

The averaged results of neutralization experiments are shown in Fig. 5.

Fig. 5. *Angular dependence of neutralization coefficient*

4 Discussion

In fact, for metals with a smooth clean surface the neutralization coefficient falls within the range of 95–99 %, while for Si it makes approximately 30 %. Even in n-doped silicon the number of free electrons is many orders less than in a metal. Besides, there is always a thin oxide film on the top of the silicon wafer preventing tunneling, so it is rather reasonable that in our case the neutralization is by times weaker than for metals. As mentioned above, there is a very low probability of exciting high energy levels of nitrogen by resonant one-electron tunneling from silicon due to level and band structure, but another dielectric mechanism is possible for excitation of higher levels [14]. It was shown, the resonant coherent excitation of fast hydrogen atoms in front of a LiF (001) surface exists at resonance energies of 5 keV [15]. Considering the fact that the resonant energy is proportional to the ion mass, one can expect that for nitrogen ions with the energy of about 70 keV it is possible to get the excitation of the higher energy levels and detect the corresponding UV irradiation. The test experiments are soon expected using the ILU-4/17 facility.

5 Conclusions

It is shown that the resonant one-electron neutralization is rather effective in case of neutralization at grazing incidence angles on silicon wafer surface and could be used as the first step in high atomic level excitation provided the conditions of resonant coherent excitation are also fulfilled. The same is highly likely for helium beam neutralization but should be proved experimentally. This work is the first step to study the potential of ion beam neutralization for optical applications and should be continued.

6 Acknowledgements

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