ISSN 0021-3640, JETP Letters, 2007, Vol. 86, No. 3, pp. 172–177. © Pleiades Publishing, Ltd., 2007.
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Laser-Induced Quantum Adsorption of Atoms on a Surface

A. E. Afanasiev, P. N. Melentiev, and V. I. Balykin

Institute of Spectroscopy, Russian Academy of Sciences, Troitsk, Moscow region, 142190 Russia

Received June 19, 2007

A method of the quantum adsorption of atoms on a surface is proposed and experimentally implemented. The loading of atoms into a surface potential well (adsorption) occurs due to the loss of kinetic energy in the process of the inelastic collision of two laser-excited atoms. This scheme is implemented for Rb atoms adsorbed on the surface of a YAG crystal. The possibility of producing microstructures of arbitrary shape that consist of atoms localized on the dielectric surface is also demonstrated.

PACS numbers: 32.80.Lg, 34.50.Bw, 34.50.Dy, 81.16.Mk

DOI: 10.1134/S0021364007150052

The adsorption of an atom (molecule or cluster) on a surface is a natural process of the trapping of the atom (molecule or cluster) in a surface potential well (van der Waals interaction, formation of a chemical bond). The probability of trapping of the particle on the surface is determined both by the electronic structures of the particle and surface and by the thermodynamics of the collision of particles with the surface. In addition to the general physical interest in the process of the adsorption of particles on the surface [1], the adsorption of particles underlies the modern industry of micro- and nanoelectronics based on the methods of molecular beam epitaxy or gas phase epitaxy [2, 3]. For this reason, the control of elementary processes of the adsorption of particles on the surface is of both fundamental and large applied importance.

At present, one proposal on controlled loading of atoms into the surface potential well is known. It is based on the effect of the photoadsorption of the atom on the surface [4]. This process is similar to the photoassociation of atoms, i.e., the formation of a molecule from two atoms due to the absorption of a photon at the instant of collision of these atoms. The effective light-induced pairing of atoms became possible after the development of the methods of the laser cooling of atoms, which made it possible to significantly increase the phase density of atomic ensembles [5] (the probability of a triple atom–atom–photon collision event is proportional to the atomic phase density $\rho\lambda_B^3$, where ρ is the density of atoms and λ_B is the de Broglie wavelength). The probability of the photoassociation of particles can also be increased if one of the colliding particles is macroscopic. This is the case when the atom collides with the solid surface in the presence of a laser field. The rate of photoassociation of the atom with the surface can be many orders of magnitude higher than the corresponding rate of the photoassociation of two atoms (the ratio is $S/\lambda_B^2 \gg 1$, where S is the area of the surface illuminated by light). Nevertheless, the photoassociation-induced loading of atoms into the surface potential well (their adsorption) still requires ultracold atoms and the efficiency of the process is low according to the theoretical estimates [4].

In this paper, we propose a new mechanism of loading atoms into the surface potential well (i.e., their adsorption) and demonstrate the implementation of this scheme for Rb atoms adsorbed on the surface of a YAG crystal. We also show the possibility of producing micro- and nanostructures of arbitrary shape that consist of atoms localized on the dielectric surface. The proposed mechanism of the loading of atoms into the surface trap is based on the energy-pooling effect, i.e., inelastic collision of two excited atoms followed by the transition of one of them to the ground state and the other one to the highly excited state [6–8]. The defect of the internal energy is compensated by the kinetic energy of the atoms. When the atomic collision occurs inside the surface potential well, an atom can be trapped in this well.

Figure 1a shows the scheme of the lowest levels of the Rb atom. The $5^2P_{3/2}$ level of the Rb atom is populated due to the absorption of 780-nm laser light. The energy of the $5d$ level is close to the $5p + 5p$ asymptotic energy of the Rb_2 molecule and, therefore, can be populated due to the energy-pooling collisions of excited atoms:

$$\mathrm{Rb}(5p) + \mathrm{Rb}(5p) + \Delta E \longrightarrow \mathrm{Rb}(5d) + \mathrm{Rb}(5s), \quad (1)$$

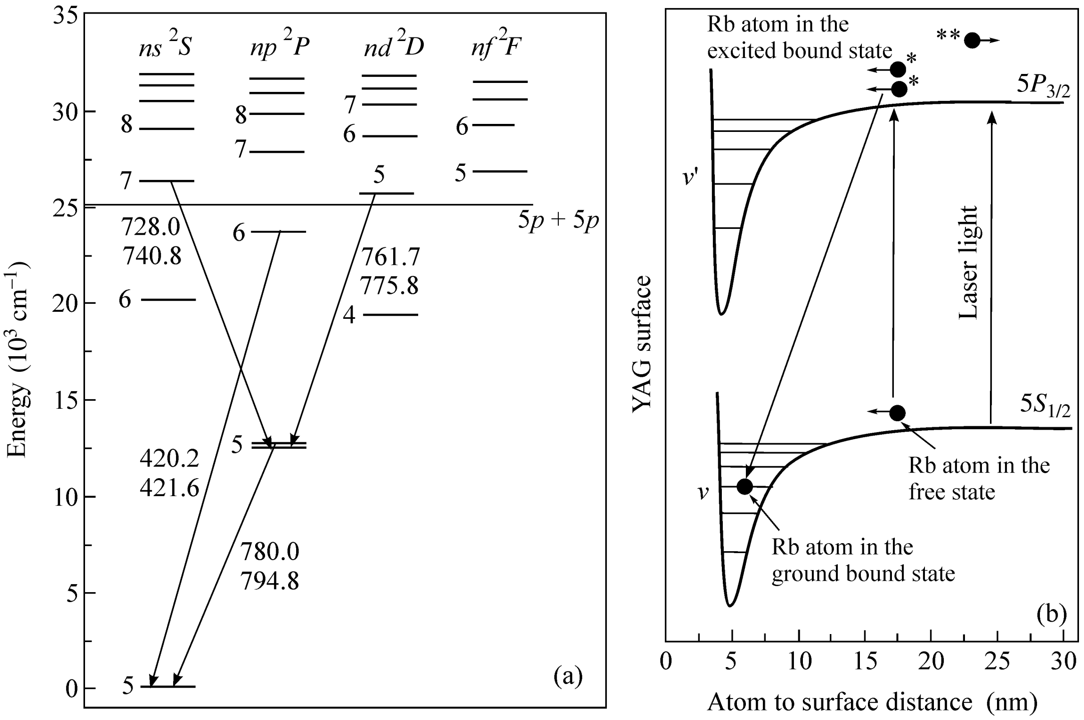

Fig. 1. (a) Scheme of the low-lying levels of the Rb atom. The energy of the $5d$ level is close to the $5p + 5p$ asymptotic energy of the Rb_2 molecule and, therefore, can be populated due to the collision of excited atoms. (b) The loss of the kinetic energy in the atomic collision can lead to the localization of an atom in the surface potential.

where $\Delta E = 93$ K is the difference between the total kinetic energies before and after collision (energy defect). As a result of collision (1) of two atoms, one atom undergoes transition to the ground state and the other atom, to the $5d$ excited state. The energy defect is compensated by the energy of the translational atomic motion; i.e., the kinetic energy of two atoms after collision event (1) decreases by 93 K. The energy-pooling process is easily identified by observing blue fluorescence appearing due to the transition of the atom from the $5d$ state to the $6p$ state, from which the atom decays into the ground state with the emission of 420.2- and 421.6-nm photons [6–8]. If the collision of two excited atoms occurs near the bottom of the surface potential well, the loss of kinetic energy in the atomic collision can lead to the localization of an atom in the surface potential, i.e., to its adsorption, as schematically shown in Fig. 1b.

In this study, the adsorption of the Rb atom near the surface of the YAG crystal is investigated. The atomic surface potential is taken as the sum of the long-range attractive van der Waals potential and the short-range repulsive potential [9]

$$V(z) = V_0 \exp[a\psi(\mathbf{R}) - z], \qquad (2)$$

where $\psi(R) = \psi(G_0, x, y)$ is a function describing the surface corrugation, G_0 is the reciprocal lattice vector of the surface, and x and y are the coordinates in the surface plane.

The quantum adsorption of Rb atoms is experimentally implemented with the use of a sapphire cell with a vapor of Rb atoms. The windows of the cell are made of a YAG crystal. The temperature of the cell can be varied from room temperature to 240°C. The temperature of the cell windows is higher than the cell temperature by 30°C. In such a cell heating regime, the cell windows are free of Rb atoms. Laser radiation is tuned to resonance with the $5^2S_{1/2}$–$5^2P_{3/2}$ transition and a laser beam passing through the cell is perpendicular to the cell windows. The laser beam diameter is varied between 0.5 and 2 mm and the maximum power of laser radiation is equal to 70 mW. The energy-pooling process is identified by detecting the blue fluorescence of Rb atoms ($\lambda = 420.2$ and 421.6 nm) from the laser beam region in the cell. Blue fluorescence is detected through a monochromator by means of a photodiode.

The laser-induced adsorption of Rb atoms on the surface is accompanied by the appearance of a thin surface layer of atoms on a cell window at the point of the intersection of the laser beam with the cell window. At present, two main methods are used for the diagnostics of the dynamics of the adsorption of atoms on the surface. The first method is based on the diffraction of fast electrons [10]. The second method is the scanning tunneling microscopy of the surface [11]. In view of the

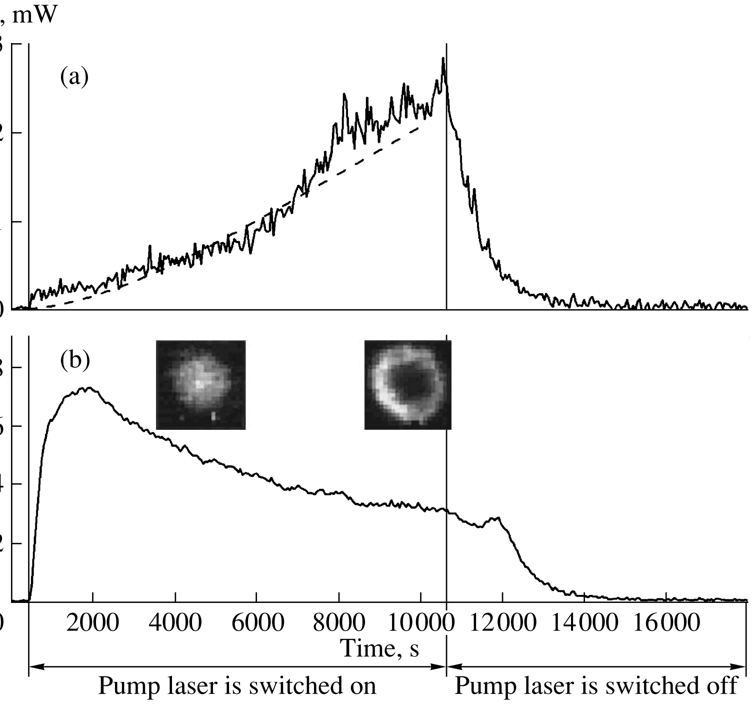

Fig. 2. Process of the adsorption of atoms and the subsequent growth of a monolayer on the surface vs. the time of the irradiation of a cell with Rb vapor: (a) the slow-adsorption regime (laser-radiation intensity $I = 150$ mW/cm^2) and (b) the fast-adsorption regime (laser-radiation intensity $I = 1.26$ W/cm^2).

difficulty of the application of both these methods in our experiment, we use another approach to the diagnostics of the adsorption of atoms on the basis of the scattering of incoherent light by atoms adsorbed on the surface. The cell window with the adsorbed atoms is illuminated by a light beam from an incandescent lamp and radiation scattered by the atoms is focused by a lens on a CCD matrix. This technique makes it possible both to measure the integral signal scattered by atoms and to determine the shape and structure of the spot of the atoms adsorbed on the surface.

The process of the adsorption of atoms and the subsequent growth of a monolayer on the surface includes several stages: (i) adsorption of individual atoms, (ii) diffusion of adsorbed atoms on the surface, (iii) nucleation of the growth centers of atomic islands, (iv) growth of the islands, (v) coalescence of the islands, and (vi) appearance of the monolayer [12]. The sensitivity of our method for detecting atoms on the surface from light scattering makes it possible to observe the effects of the growth of the islands and the coalescence of the islands into the monolayer.

Figure 2 illustrates the process of the adsorption of atoms and subsequent growth of a monolayer on the surface with the time of the irradiation of the cell with Rb vapor. The vertical axis shows the integral intensity of light scattered by Rb atoms adsorbed on the surface and the horizontal axis is the time of the illumination of the cell by laser radiation. Figure 2a corresponds to the slow-adsorption regime and shows change in the number of adsorbed atoms at a relatively low rate of trapping atoms in the surface potential. Such a regime is implemented with the use of a low laser radiation intensity of $I = 150$ mW/cm^2. The initial rising section of the curve corresponds to the first four stages of the growth of the monolayer from the adsorption of individual atoms to the growth of the islands. When laser radiation is terminated at time $t = 10^4$ s, the signal begins to decrease due to the process of the thermal desorption of atoms from the surface with a typical (temperature-dependent) time of $t_\mathrm{d} = 10^3$ s.

Figure 2b corresponds to the fast-adsorption regime implemented at a laser-radiation intensity of $I =$ 1.26 W/cm^2. The intensity of light scattered by the adsorbed atoms is saturated in a time of $t_\mathrm{s} = 2 \times 10^3$ s. With subsequent irradiation of the cell of a vapor of Rb atoms by laser radiation, the scattered signal decreases because the process of the adsorption of atoms passes to the stage of the coalescence of the islands into the monolayer, which reflects light as a mirror rather than scattering it.

The image of the spot of the adsorbed Rb atoms on the surface consists of separate scattering points. The left inset in Fig. 2 shows a CCD image of the spot of the adsorbed Rb atoms at the initial adsorption stage. The spot is formed by separate scattering centers, where the maximum of scattering centers corresponds to the center of the laser spot at which the radiation intensity and, correspondingly, adsorption rate are maximal. The right inset in Fig. 2 shows an image of the adsorbed atoms at the later stage of the adsorption of atoms and the growth of the monolayer, when individual islands overlap and the atomic monolayer is formed at the spot center. The monolayer reflects atoms as a mirror and, as a result, a hole appears in the image of the spot of adsorbed atoms due to the decrease in the light scattering intensity.

To calculate the adsorption of atoms, we use a simple model of the rate equations describing the adsorption of atoms at the initial stage when the degree of covering of the surface by atoms is $Q \ll 1$ [13]. The rate of the adsorption of Rb atoms on the surface is determined by the rate of the energy-pooling process [6–8]:

$$F = \frac{1}{2}Kn_{5p}^2\Delta R, \tag{3}$$

where K is the rate of this process, n_{5p} is the density of Rb atoms in the excited $5p$ state, and ΔR is the characteristic size of the surface potential in the direction perpendicular to the surface. The intensity of light scattered by the adsorbed atoms is calculated with the use of the model of light scattering on metal particles that are small as compared to the wavelength. The corresponding scattering cross section is given by the expression [14]

$$d\sigma = \frac{\omega^4}{2c^4}V^2|\alpha|^2(1+\cos^2\vartheta)do. \tag{4}$$

Expression (3) for the rate of the adsorption of atoms on the surface and expression (4) for the cross section of light scattering by adsorbed atoms are used in the rate equations. The dotted line in Fig. 2a shows the atom adsorption rate calculated from the rate equations. As seen, the general behavior of the calculated dependence is in good agreement with the experimental curve.

We also investigate the efficiency of the quantum adsorption as a function of the laser radiation frequency by varying the frequency across the D_2 adsorption line of a vapor of Rb atoms in the range $\Delta\nu = 24$ GHz. The laser radiation frequency is controlled by observing the absorption spectrum in an additional cell with a vapor of Rb atoms as shown in Fig. 3a. The intensity of 420-nm (blue) fluorescence corresponding to the $6p$–$5s$ transition in the Rb atoms in the cell under investigation is measured simultaneously with variation of the frequency. This dependence is shown in Fig. 3b. The characteristic feature of the frequency dependence of the blue fluorescence is the decrease in the fluorescence intensity at resonance frequencies. The suppression of the fluorescence signal and, correspondingly, the efficiency of the energy-pooling process are explained by the deexcitation of atoms from the $5p$ level due to the collisions with the wall. It is known [15] that the penetration depth of laser radiation into the region of high-density atomic vapor depends strongly on the radiation frequency. Laser radiation with frequency near resonance penetrates only to a depth of about light wavelength λ and the excited atomic state is deexcited due to collisions with the wall in a characteristic time of $t = \lambda/V \sim 1$ ns, which is an order of magnitude smaller than the time of the spontaneous emission from the Rb atom (~28 ns). At large detunings, laser radiation penetrates deeper and produces a significantly higher density of excited atoms near the surface of the cell window.

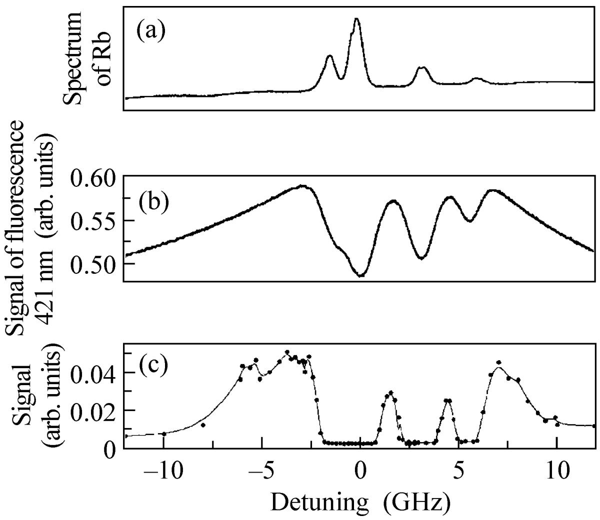

Fig. 3. (a) The absorption spectrum of Rb atoms in the vapor reference glass cell at room temperature. (b) The laser-frequency dependence of the intensity of 420-nm (blue) fluorescence corresponding to the $6p$–$5s$ transition in the Rb atoms in the sapphire cell under investigation. (c) The laser-frequency dependence of the signal of light scattered by Rb atoms adsorbed on the YAG surface. This signal characterizes the efficiency of the loading of atoms into the surface potential.

Figure 3c shows the signal of light scattered by Rb atoms adsorbed on the surface as a function of the laser frequency. This dependence characterizes the efficiency of the loading of atoms into the surface potential. Measurements are carried out at a cell temperature of 215°C. This temperature is chosen so as to ensure both the sufficiently fast creation of the atomic structure on the surface (a characteristic time of ~2 min) and the sufficiently fast destruction (a characteristic structure lifetime of ~10 min) of the structure due to thermal desorption after the removal of laser radiation. The measurement procedure is as follows: (i) the laser radiation frequency is set at the D_2 absorption line of Rb atoms, (ii) the spot of Rb atoms adsorbed on the surface is created and the number of adsorbed atoms is measured from the scattered signal, and (iii) laser radiation is switched off and the atomic spot is destroyed due to

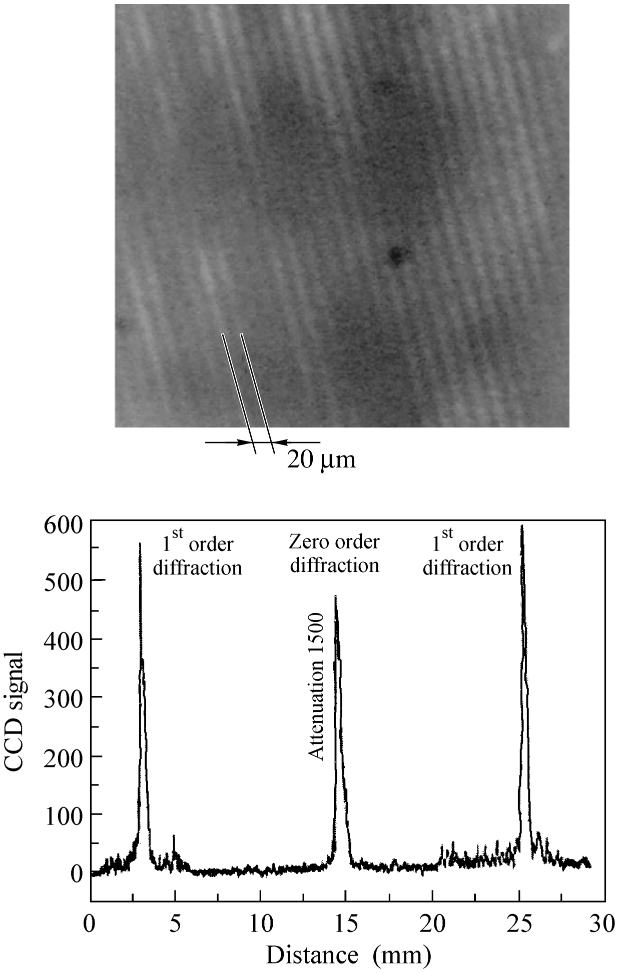

Fig. 4. Amplitude diffraction grating consisting of Rb atoms adsorbed on the surface and the diffraction signal of laser radiation from the atom diffraction grating.

thermal desorption. Then, the measurement is repeated at a different laser frequency, etc. The resulting dependence is shown in Fig. 3c. The comparison of the spectral dependence of the adsorption of atoms (see Fig. 3c) with the spectral dependence of the energy-pooling collisions (see Fig. 3b) shows that the behaviors of both processes are identical. This identity is evidence that the energy-pooling collisions are responsible for the loading of atoms into the surface potential wall.

Quantum adsorption opens the possibility of creating atomic micro- and nanostructures with a given geometry on the dielectric surface. Figure 4 illustrates such a possibility using as an example the creation of the grating of Rb atoms absorbed on the surface. In order to create such an atomic grating, the spatial profile of the laser beam is modulated by its reflection from a Fresnel mirror. The irradiation of the cell window by such a spatially modulated laser beam produces the interference pattern of the maxima and minima of the intensity on the cell window. Atoms are adsorbed on the surface in the intensity maximum regions, whereas the intensity minimum regions are free of atoms. Thus, a regular system of atomic strips is produced on the surface of the cell window. The grating period is equal to 20 μm.

The system of atomic strips is an amplitude diffraction grating for a probe laser beam incident on it. Therefore, the laser beam reflected from the created atomic structure undergoes diffraction. Figure 4 shows the spatial distribution of the intensity of the laser beam reflected from the system of atomic strips on the surface of the cell window. The distribution is recorded with the CCD. The efficiency of the atomic diffraction grating is low. For this reason, the zero-order diffraction signal is reduced by a factor of 1500 by means of neutral filters for simultaneous detection of three diffraction orders. The first diffraction maxima, as well as the zero-order maximum, are seen in Fig. 4.

Note that the use of laser radiation with nanometer spatial dimensions [16, 17] opens the possibility of creating atomic nanostructures of arbitrary shape on the surface by means of quantum adsorption.

Thus, the mechanism of laser-controlled quantum adsorption of Rb atoms on the surface of a YAG crystal that is based on the energy-pooling process has been investigated in this study. The possibility of producing microstructures of arbitrary shape with the use of quantum adsorption has been demonstrated.

This work was supported by the Russian Foundation for Basic Research (project nos. 06-02-16301-a, 06-08-01299-a, and 05-02-16370-a). We are grateful to Profs. V.G. Minogin and V.S. Letokhov for stimulating discussion of the study and results and to the group headed by Prof. O. Sarkisyan for manufacturing the cell with Rb vapor.

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Translated by R. Tyapaev