DYNAMICS OF PHOTOIONIZATION OF BARIUM ATOMS BY SOLAR RADIATION

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Results of numerical simulation of the process of a plane layer of optically thin and thick Ba *vapor photoionization by solar radiation are presented. A multilevel model of the* Ba *atom has been constructed which allows one to describe dynamics of vapor photoionization with an account of kinetics of population of singlet* $^1D_2$ *and triplet* $^3D_{1,2,3}$ *states. Ionization of dense vapor occurs inhomogeneously along the spatial variable due to the effect of the incident radiation absorption by the layer, and the effective time of the layer ionization increases in comparison with an optically thin vapor. Two modes of the vapor clearing up for radiation with* $\lambda = 5536.48$ Å *have also been found, the first one is related to populating of metastable states, and the second to the gas photoionization.*

Injection of barium vapor into the upper layers of the atmosphere is used to create artificial ion clouds (AIC) to make diagnostics of the environment. A cloud of neutral atoms is formed in the process of gas–dynamic separation of reaction products (vapor), which is further ionized under the effect of solar radiation.[1–4] In these papers it was noted a decisive role of atoms in the $^{1,3}D$ metastable states in the dynamics of the AIC formation, which is associated with the fact that the photoionization rate from the $^{1,3}D$ levels considerably exceeds the photoionization rate from the ground state of barium atoms. Such a strong difference is determined by the shape of solar radiation spectrum.[8,9] In the cases, when a sufficiently large amont of substance is injected, there can occur optically thick, electrically neutral clouds at frequencies corresponding to resonance transitions of barium atoms.[5,6] Therefore, to give a correct description of the photoionization process of dense formations in the upper atmosphere it is necessary to take into account the radiation transfer on resonance lines of atoms.

Thus, study and simulation of a stepwise process of ionization of barium atoms by sunlight in the theoretical aspect is of interest for determination of particular channels of ionization, and at the same time is of practical importance for forecasting the dynamics of the AIC formations when planning experiments. In addition, the obtained values of populations of excited states of atoms and ions are important for calculation of spectral line intensities and interpretation of the available experimental data on the AIC glow.

The aim of the work is to determine channels and time of ionizations for a plane layer of optically thin and thick barium vapor.

OPTICALLY THIN MEDIA

The dynamics of optically thin vapor ionization is described by a system of balance equations for a population of the levels which are included into the barium–atom model. The system of equations allows one to take into account the processes of excitation, quenching, ionization of atoms under the action of a broad–band radiation, and the spontaneous decay. Before the irradiation all atoms are assumed to be in the ground state, their concentration known and equal to N_0. The rates of the photoexcitation and spontaneous decay have been calculated using data on the forces of oscillators[7] and on the solar radiation spectrum.[8,9] The estimations of the rate values and photoionization cross sections have been made by the semiempirical technique described in Ref. 10. Note that thus obtained values of the photoionization cross sections for metastable ^3D levels are in a good agreement with experimental data.[11]

For the simplest three–level model, involving the ground, metastable, and optically associated with them excited states, the system of balance equations has an analytical solution, if the population $N_3(t)$ of the excited level adiabatically follows the change in populations of the ground $N_1(t)$ and metastable $N_2(t)$ states. Such an approximation (quasistationarity mode) gives that $dN_3/dt \approx 0$ and its employment in our case is reasonable because the characteristic photoionization time is much longer than the photoexcitation and spontaneous decay time

$$\frac{1}{F_j} \gg \frac{1}{B_{ij}} \gg \frac{1}{A_{ji}} , \qquad (1)$$

where F_j is the rate of atom photoionization from the jth level, B_{ij} and A_{ji} are the rates of photoexcitation and spontaneous decay for the transition $i \to j$.

Under such assumptions the dynamics of atom populations $N_1(t)$, $N_2(t)$, and $N_3(t)$ and electron concentration $N_e(t)$ is determined by the rates of photoprocesses and has the form

$$N_1(t) = N_0\{q_1[\exp(-\lambda_2 t) - \exp(-\lambda_1 t)] + \exp(-\lambda_1 t)\} , \qquad (2)$$

$$N_2(t) = N_0\{q_2[\exp(-\lambda_1 t) - \exp(-\lambda_2 t)]\} , \qquad (3)$$

$$N_3(t) = \frac{B_{13}}{\alpha_0} N_1(t) + \frac{B_{23}}{\alpha_0} N_2(t) , \qquad (4)$$

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$$N_e(t) = N_0 - p_1 N_1(t) - p_2 N_2(t), \tag{5}$$

$$\begin{cases} q_1=\dfrac{\alpha_{11}(\lambda_1-\alpha_{11})}{\Delta};\ q_2=\dfrac{(\lambda_1-\alpha_{11})(\lambda_2-\alpha_{11})}{\Delta}; \\ p_1 = 1 + B_{13}/\alpha_0;\ p_2 = 1 + B_{23}/\alpha_0; \\ \lambda_1 = \dfrac{(\alpha_{11}+\alpha_{22})}{2}(1+\delta);\ \lambda_2 = \dfrac{(\alpha_{11}+\alpha_{22})}{2}(1-\delta); \\ \Delta = \alpha_{12}(\lambda_1-\lambda_2);\ \delta = \left[1-\dfrac{4(\alpha_{11}\cdot\alpha_{22}-\alpha_{12}\cdot\alpha_{21})}{(\alpha_{11}+\alpha_{22})^2}\right]^{1/2}; \\ \alpha_1 = F_3 + B_{32} + A_{32};\ \alpha_2 = F_3 + B_{31} + A_{31}; \\ \alpha_0 = \alpha_1 + B_{31} + A_{31}; \\ \alpha_{11} = F_1 + B_{12} + B_{13}\cdot\dfrac{\alpha_1}{\alpha_0};\ \alpha_{21} = B_{12} + \dfrac{B_{13}}{\alpha_0}(B_{32}+A_{32}); \\ \alpha_{12} = B_{21} + A_{21} + \dfrac{B_{23}}{\alpha_0}\cdot(B_{31}+A_{31}); \\ \alpha_{22} = F_2 + B_{21} + A_{21} + \dfrac{\alpha_2}{\alpha_0}\cdot B_{23}. \end{cases} \tag{6}$$

A theoretical estimation of the characteristic photoionization time τ_{ph} can be obtained, using the formula

$$\tilde{N}_e(t) = N_0\left[1 - \exp\left(-\frac{t}{\tau_{ph}}\right)\right] \tag{7}$$

for the approximation of time dependence of the electron concentration $N_e(t)$, which is set by Eq. (5). Numerical value of τ_{ph} is calculated from the condition of functional minimum

$$R(\tau_{ph}) = \frac{1}{N_0^2}\int_0^\infty \left[N_e(t) - \tilde{N}_e(t)\right]^2 dt = R_0 + \tau_{ph}\times$$

$$\times\left\{\frac{1}{2} + \frac{2(p_2q_2 - p_1q_1)}{(\lambda_2\tau_{ph}+1)^2} + \frac{2[p_2q_2 - p_1(1-q_1)]}{(\lambda_1\tau_{ph}+1)^2}\right\}, \tag{8}$$

describing the value of the approximation rms error. The functional minimum (8) is reached at the values of τ_{ph} which satisfy the equation

$$\frac{1}{4} - (p_2q_2 - p_1q_1)\frac{(\lambda_2\tau_{ph}-1)}{(\lambda_2\tau_{ph}+1)^3} - \left[p_2q_2 - p_1(1-q_1)\right]\times$$

$$\times\frac{(\lambda_1\tau_{ph}-1)}{(\lambda_1\tau_{ph}+1)^3} = 0. \tag{9}$$

Given in Table I are the values of the characteristic ionization time τ_{ph} and minimum values of functional (8) corresponding to the ionization of atoms according to a three–level scheme from different metastable states.

TABLE I. The values of characteristic times of barium photoionization τ_{ph} corresponding to different metastable states found using a three–level model of atom.

Resonance transition, λ, Å	Metastable level	Time τ_{ph}, sec	Value of functional
$^1S_0 \to {}^1P_1$ 5535.48	$^1D_2^*$	207.94	– 0.0579
$^1S_0 \to {}^3P_1$ 7911.34	$^3D_1$	450.00	– 0.0240
$^1S_0 \to {}^3P_2$ 7911.34	$^3D_2$	773.55	$-3.373\cdot10^{-4}$
$^1S_0 \to {}^3P_1$ 3889.33	$^3D_1$	444.35	– 0.0607
$^1S_0 \to {}^3P_1$ 3889.33	$^3D_2$	338.36	– 0.1363
$^1S_0 \to {}^1P_1$ 3501.11	$^1D_2$	369.36	– 0.0557
$^1S_0 \to {}^1P_1$ 3071.58	$^1D_2$	511.09	– 0.0318
$^1S_0 \to {}^1P_1$ 5535.48	$^1D_2$	160.05	– 0.1158

*The metastable level $^1D_2$ decays at a rate of 2 s^{-1}.

The analysis of results shows, that the least time τ_{ph} of 207.9 s is obtained for the case, in which the ionization occurs from the $^1D_2$ level which is populated through the excited state $^1P_1$ by radiation with the wavelength 5535.48 Å. Based on this fact, one may evidently assume that it is this stepwise process that makes the main contribution into the ionization of barium atoms. However, the value τ_{ph} obtained for it considerably differs from the experimentally determined characteristic time of ionization within the interval of from 20 to 30 sec.[4] This may be caused by approximation used in the description of the atom ionization process by radiation in the three level model, since it does not take into account the contributions of other states into ionization. The computations have shown that the total contribution into the atom ionization from $^1D_2$ level from other excited states, except for $^1P_1$, is small and practically does not decrease the τ_{ph} value.

The effect of the triplet group of metastable levels $^1D_{1,2,3}$ on the photoionization kinetics has been taken into account within the frameworks of the multi level model of barium atom that accounts for the ground and metastable $^1D_2$ and $^3D_{1,2,3}$ states, and, in addition, those excited states which optically couple the ground and metastable levels or the metastable states $^1D_2$ and $^3D_{1,2,3}$. As a result one obtain a twelve–level model of barium atom. The corresponding system of balance equations relative to the population has been solved by numerical methods.[10]

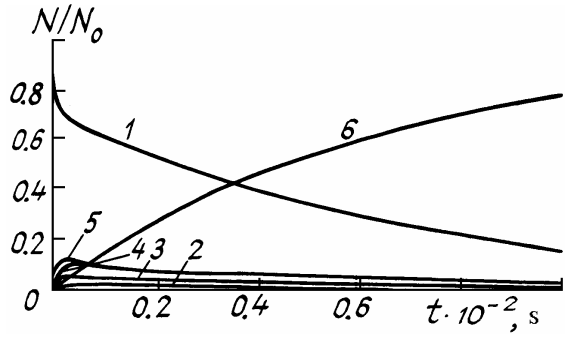

FIG. 1. Dinamics of a number density of ions (6) and Ba *atoms in the states:* 1) $^1S_0$, 2) $^3D_1$, 3) $^3D_2$, 4) $^3D_3$, and 5) $^1D_2$.

Temporal behaviors of the electron number density, populations of the ground, and metastable ($^3D_1$ – (*2*), $^3D_2$ – (*3*), $^3D_3$ – (*4*), and $^1D_2$ – (*5*)) states are shown in Fig. 1. As can be seen at the beginning of the ionization during the time interval shorter 1 s there occurs a redistribution of atoms between the ground and metastable levels under the action of photoexcitation and spontaneous decay. Then a slow ionization with the characteristic time $\tau_{ph} \approx 67$ s of metastable atoms by radiation takes place. In addition, it has been revealed that coupling between metastable states $^1D_2$ and $^3D_{1,2,3}$ through intermediate excited states plays an important role in barium ionization kinetics, since at its absence the value of the characteristic time τ_{ph} increases up to 127 seconds.

OPTICALLY THICK MEDIA

A description of the photoionization of dense barium clouds should take into account the radiation transfer processes on resonance lines of atoms, since it can lead to a spatial inhomogeneity of the AIC. In addition, the radiation transfer can lead to formation of a spatial distribution of internal sources of radiation (excited atoms), which finally determine the dynamics of the neutral and ion clouds glow under the effect of external radiation.

In this section we discuss the main results of a numerical simulation of the process of photoionization of the plane–parallel dense layer of barium vapors one side of which is irradiated by solar radiation at right angle of incidence.

The mathematical model of the process under consideration was constructed based on the following assumptions:

1) reemission of absorbed photons by atoms is described in the approximation of a complete frequency redistribution;[12,13]

2) contours of the emission and absorption lines have a Doppler shape; and,

3) scattering of photons by atoms is isotropical (spherical scattering phase function).

Then in the system of balance equations for a multi level atom, in contrast to an optically thin layer, there appear terms, describing the contribution in to the photoexcitation processes coming from the direct solar radiation penetrated to a given point of the layer from outside, and also there will appear terms which are determined only by the intensity of radiation emitted by excited atoms of the medium. The equations should take into account the total over the volume contribution to photoexcitation rates and, therefore, for populations of the considered atomic states we obtain a system of integro–differential equations. Similar terms appear in the integral equation which allows one to find a distribution of excited atoms over the layer under the effect of external radiation and/or an electron impact in a stationary case for a two–level atom.[14,15,16] The dynamics of direct solar radiation intensity is described by a stationary transfer equation. The derivative of radiation intensity with respect to time is assumed to be equal to zero because the characteristic time of radiation propagation in the absorbing medium is too short compared to the characteristic time of atomic photoexcitation. This circumstance explains the fact that spatial distribution of intensity reaches its steady state long before the moment, when physical properties of the absorbing medium that govern the process of radiation propagation noticeably change.

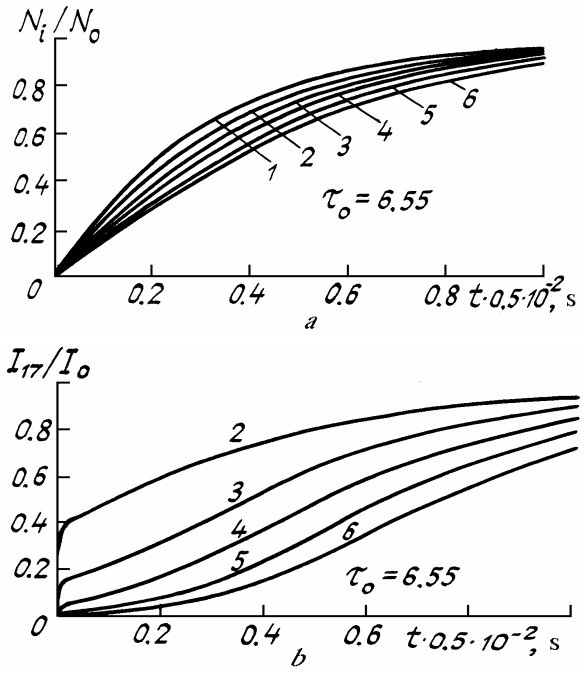

FIG. 2. Time behavior of ion density (a) and radiation intensity at central frequency of the Doppler contour for the resonance line with $\lambda = 5538.48$ Å *(b) at different points of the medium x/L: 1)* 0.0, *2)* 0.2, *3)* 0.4, *4)* 0.6, *5)* 0.8, *and 6)* 1.0.

Thus obtained a system of equations is too cumbersome and therefore we omit it. This system can be reduced to a system of ordinary differential equations (ODE) for populations of levels at the assigned set of points throughout the layer with the appropriate choice of optimal cubature formulas for integrating with respect to angular, frequency, and spatial variables. The initial value problem for the system of the ODE was solved by numerical methods.[17,18]

The results of computations have shown, that the photoionization dynamics occurs inhomogeneously within the layer (Fig. 2*a*). This is caused by the fact that the photoexcitation rate at the resonance transition decreases with depth, because the radiation intensity decreases due to absorption. Consequently the characteristic photoionization time for optically thick media depends on the spatial variable x.

Let the temporal behavior of the electron concentration $N_e(x_i, t)$ at a given point of the medium x_i be approximated as follows

$$\tilde{N}_e(x_i, t) = N_0\left\{1 - \exp\left[-\frac{t}{\tau_{ph}(x_i)}\right]\right\}, \quad (10)$$

then from the condition of minimum of the rms error of approximation described by the functional

$$\hat{J}[\tau_{ph}(x_i)] = \frac{1}{N_0^2}\sum_{j=1}^{M}\left[\tilde{N}_e(x_i, t_j) - N_e(x_i, t_j)\right]^2, \quad (11)$$

we can calculate the characteristic photoionization time $\tau_{ph}(x_i)$. Here t_i is the moment at which the value of electron concentration $N_e(x_i, t)$ is found from the numerical solution of the initial problem, M is the number of such moments. For the characteristic ionization time of the layer as a whole (with the optical thickness τ_0) we take the value $\tau_{ph}(\tau_0)$, which is obtained by averaging the function $\tau_{ph}(x)$ over the spatial variable and calculated by formula

$$\tau_{ph}(\tau_0) = \frac{1}{L}\int_0^L \tau_{ph}(x)dx \approx \frac{1}{(n-1)} \times$$

$$\times \left\{\frac{1}{2}\left[t_{ph}(x_1) + t_{ph}(x_n)\right] + \sum_{l=2}^{n-1} t_{ph}(x_l)\right\}, \quad (12)$$

which is a consequence of using the trapezoid rule in calculating the integral (L is the layer thickness, n is the number of nodes along the spatial variable). The ionization time dependence $\tau_{ph}(\tau_0)$ on the optical thickness of the layer τ_0 is shown in Fig. 3. It can be seen that the ionization process in a thick layer decelerates compared to that in an optically thin layer, and more rapidly with increasing optical thickness. Thus, for example, for the layer with $\tau_0 = 13.1$ the ionization time increases by 49.2%.

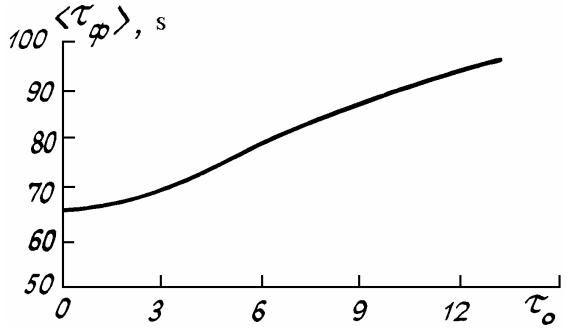

FIG. 3. Dependence of the average ionization time on optical thickness of the layer.

One should also note that a spatial inhomogeneity is also observed in the distribution of atoms in all states. The dynamics of the radiation intensity at the central frequency of a Doppler contour at different points of the absorbing layer is depicted in Fig. 2*b*. Intensity growth is noted at all frequencies of the line absorption contour as ionization by radiation develops. Two qualitatively different modes of intensity change are distinguished: the first with the temporal interval 1 s, which we call "quick", and the second one with a slow growth dynamics — "slow". The "quick" mode of the process of decreasing the density of atoms in the ground state under the effect of photoexcitation, which yields quick redistribution of atoms among the ground and metastable levels. The second mode is maintained by decreasing the number of atoms involved in the photoionization process. As it develops, clearing up of the absorbing layer occurs, what is explicitly seen from Fig. 4, where the frequency dependence of the outgoing radiation intensity at different stages of ionization is given. Thus the two modes of the radiation intensity change are qualitatively quite different of the medium clearing up.

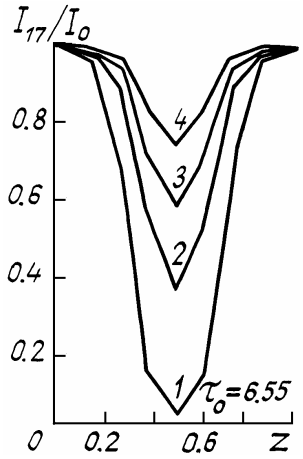

FIG. 4. Frequency dependence of the intensity of the outgoing from the layer radiation at the moment t: 1) 40 s*, 2)* 80 s*), 3)* 160 s*, and 4)* 200 s ($z = (\nu - \nu_0)/\Delta\nu_D - 3.19$).

In conclusion we should like to note that a photoionization analysis of barium vapor in the case of optically thin layer allows one to construct a multilevel atomic model, to describe qualitatively ionization, and to obtain a qualitative estimation of the characteristic ionization time. The numerical simulation of the photoionization for dense formations using the multilevel atomic model has shown that the process of ionization of the layer as a whole decelerates in comparison with that in optically thin media and occurs inhomogeneously in space. Two different modes of the absorbing layer clearing up are observed.

The spatial ionization inhomogeneity must affect the layer glow dynamics but this is the subject for different investigations which the authors intend to take part in.

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