DETECTION OF LAYERS OF METASTABLE ATOMS IN THE UPPER ATMOSPHERE AND IN SPACE

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Received April 25, 1996

The possibility is considered of detecting metastable copper atoms with the use of Cu–vapor laser radiation at 510.6 *and* 578.2 nm *wavelengths. The fluorescent re-emission channel at* 324.7 *and* 327.4 nm *allows the sought information about the concentration of metastable copper atoms to be obtained. Our calculations show that for the layer of copper metastable atoms* 10 km *thick and number density of* 1 atom/cm^3 *at the altitude of* 90 km *their detection is possible with the use of a laser with medium performance characteristics both from the ground and from a spacecraft flying at an altitude of* 300 km.

INTRODUCTION

Typical gaseous constituents of the upper atmosphere and near space are atoms of chemical elements in the ground, metastable and ionized states. They are of crucial importance for physico-chemical reactions and radiative properties of the upper atmosphere.[1]

However, the problem on the mechanism of bringing a number of atoms, including metals, into the upper atmosphere is so far open to discussions. In particular, both space and the Earth are considered as possible sources of atoms of metals. In the first case, the sources are meteors, overwhelming part of which is destroyed, when entering the atmosphere, at altitudes of 60–100 km. Spectral analysis of meteors composition shows the presence of atoms and ions of such elements as H, O_2, Na, Mg, As, Ca, Cr, Mn, Fe, Ni. Some meteorites contain Al, S, Au.[7–9] In the second case, it is assumed that the presence of atoms and ions in the upper atmosphere is due to not only meteors, but also their vertical transport from the ground. Similar element composition of aerosol and air near ground and in the upper atmosphere as well as correlation between the Na content in the upper atmosphere and that over the ocean surface (some of the data borrowed from Ref. 1 and illustrating this point are presented in Tables I–III) provides qualitative evidences in support of the terrestrial origin of metal layers. As is seen from Tables I–III, the list of metals present in the upper atmosphere is rather wide.

TABLE I. Aerosol composition in the layer adjacent to the surface of the Baltic Sea and the Atlantic Ocean (ng/m^3).

Element	Na	Zn	Cu	Mn	Fe	Cr	Li
Baltics	200–700	20–30	10–20	4–8	10	1–2	1–4
Atlantics	170–800	90–130	5–13	4–26	5–100	5–16	–

TABLE II. Chemical composition of air in different regions (ng/m^3).

Element	As	Cd	Hg	Pb	Zn	Fe
Continent	0.02–0.15	0.02–30	0.02	0.2–400	2–70	9–5100
Seas	0.01–0.8	0.003–0.5	0.02–5	0.05–64	0.04–56	3–1000
Antarctica	0.0007–0.04	0.018	–	0.2–1.2	0.02–0.06	0.5–1.2

TABLE III. Anthropogenic emissions in some European countries (ton/year).

Element	As	Cd	Cu	Pb	Zn	Fe
Total	3560	1155	9390	89100	57400	5100

It should be noted that presence, density, altitude profile, and other parameters of normal and excited states of atoms and ions of minor constituents are practically not studied (except for sodium and potassium[2,3]).

In this paper we consider one simple scheme for detection of metastable atoms in the upper atmosphere from the ground and from space and conclude that lidars built using metal-vapor lasers allow one to detect and study the layers of metastable atoms of a number

0235-6880/96/08 730-04 $02.00

of chemical elements even at a very low number density about 1÷10 atom/cm^3.

BASIC CONCEPT OF THE METHOD

When considering different versions of observations of metastable atoms of elements rare in the upper atmosphere, it is worthwhile to make use of the opportunity to apply lasers at vapors of these elements operating at transitions from resonance to metastable states. The conditions of laser excitation are close to the conditions in the upper atmosphere that ensures the very close agreement between the profiles of emission and absorption by particles and thus high efficiency of interaction.

The idea of constructing a lidar with lasers at vapors of chemical elements is based on the possibility to excite, with high efficiency, the anti-Stokes fluorescence in the layers of metastable atoms. The excitation energy, accumulated by atomic and ion particles, is very high – $E_{ex} = 1 \div 2$ eV. This results in the anti-Stokes shift of fluorescence by 10^4 cm^{-1}. For example, the energy of metastable states of potassium ion is 13711 and 13650 cm^{-1}. Under the exposure of the potassium layer, containing ions in metastable states, to IR radiation with wavelengths 854 and 855 nm, anti-Stokes violet fluorescence arises at wavelengths 397 and 393 nm. This example is typical – sensing in the infrared excites the fluorescence in the ultraviolet. In this case it is not very difficult to separate, by spectral methods, return signals for sensing the layers under study.

At altitudes about 100 km the atmospheric density is 10 cm^{-3} and temperature is 200 K (Ref. 1). Under these conditions, the time of intermolecular collisions is 10^{-3} s. Since the radiative lifetime of atomic resonance states is about 10^{-8} s, then the quenching processes can be neglected and the quantum yield of fluorescence can be believed equal to unity.

Let us assume, for example, that there is the layer of metastable copper atoms. As is seen from Tables I–III, copper is the medium-abundance element in both meteor and terrestrial sources. For copper atoms both exciting (at 510.6 and 578.2 nm) and fluorescence radiation (at 324.7 and 327.4 nm) is only slightly absorbed by atmospheric gases. For sensing the copper layers, we have the most high-power and easily available copper-vapor laser.[4] Figure 1 shows the diagram of lower energy levels of a copper atom. If initially some copper atoms are in the metastable states $^2D_{3/2,\,5/2}$, then the exposure to Cu-vapor laser radiation (excitation channel) will stimulate the transition of metastable atoms into the resonance states $^2P^{\,0}_{1/2,\,3/2}$ and then they will transit to the ground $^2S_{1/2}$ state with the emission (fluorescence channel).

Let us estimate the lidar signals for a hypothetical layer of metastable copper atoms. Remind, for example, that the layer of sodium atoms with thickness about l = 10 km is at the altitude of 92 km and has the density $N_{Na} = 10^2$–10^3 atom/cm^3 (Refs. 1–3). As is seen from Tables I–III, the layer of copper atoms should be a little bit lower and have lower density. Assume, similarly to the Na layer, that the layer of copper atoms is at an altitude H=90 km and has thickness l = 10 km and density of atoms in metastable states $N_{Cu} = 1 \div 10$ atom/cm^3. Optical thickness of such a layer for the exciting radiation (510.6 and 578.2 nm) is $\gamma = 10^{-7}$ and for fluorescence radiation (324.7 and 327.4 nm) $\gamma_f = \sigma_f\ N_{Cu}\ l = 10^{-6}$ due to a greater cross section σ_f. The absorption cross sections for sensing and fluorescence radiation were calculated for Cu from the data on transition probabilities[10] and were found to be equal to 10^{-13} and 10^{-12} cm^2, respectively, assuming Doppler line profiles. The cross sections are taken approximately, as only order of magnitude, because the real profiles differ from the Doppler ones due to isotopic splitting.[11] The density of metastable atoms is taken the same as for atoms in the ground state.

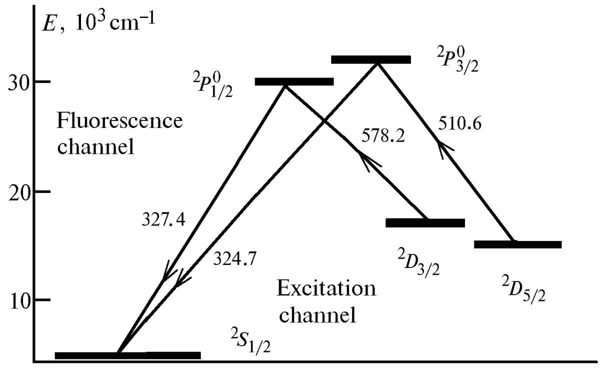

FIG. 1. The scheme of excitation of anti-Stokes fluorescence in the layer of copper atoms containing metastablle particle.

As is seen from the estimates presented, the layers of atoms turn out to be optically transparent, therefore the problem of remote detection and study of such layers is purely the lidar problem.

TECHNIQUE FOR RETURN SIGNAL CALCULATION AND THE RESULTS OBTAINED

To estimate the laser radiation attenuation in the atmosphere, one has to use the lidar equation for fluorescence.[2,3] We calculated the signals for two versions of lidar installation: on the ground and aboard a spacecraft. In the former case the sensing path passes through the whole atmospheric column from 0 to 100 km, while in the second case it passes only through the atmospheric layer 100–80 km. In our calculations we took the performance parameters of the lidar system as follows:

receiving telescope diameter, m	1
mean output power, W	10
radiation wavelengths (Cu-vapor laser), nm	510.6 and 578.2
laser pulse repetition frequency, kHz	10
laser pulse duration, ns	30

The attenuation was calculated for the aerosol atmosphere within the model of mid-latitude summer.[5] At the wavelengths 510.6 and 578.2 nm we took into account the absorption by O_3 and NO_2 molecules, while at the fluorescence wavelengths (wavelengths of information) 324.7 and 327.4 the account was taken of the absorption by molecules O_3, NO_2, and SO_2. The absorption coefficients for the above gases were borrowed from Ref. 6. The return signals from the whole layer can be calculated based on the following relationship for the fluorescence channel[2,3]:

$$E = E_0 K\, S \exp(-\gamma_1) \exp(-\gamma_2) \exp(-\gamma_3)\, \sigma_f\, N_{\mathrm{Cu}} / (4\pi H^2),$$

where E_0 is the number of photons in the laser pulse; $\gamma_1 = \int \alpha_1(x)\, dx$ is the layer optical thickness for the exciting radiation; $\gamma_2 = \int \alpha_2(x)\, dx$ is the atmospheric optical depth for the fluorescence channel; $\gamma_3 = \sigma N l$ is the optical thickness of the whole layer of Cu vapors for the exciting radiation; $\alpha_1 = \alpha_{a1} + \alpha_{m1} + \alpha_{g1}$ is the atmospheric extinction at the wavelength of exciting radiation due to aerosol (a), molecular scattering (m), and gas absorption (g); $\alpha_2 = \alpha_{a2} + \alpha_{m2} + \alpha_{g2}$ is the atmospheric extinction at the wavelength of fluorescent channel; S is the area of receiving mirror; K is the instrumental constant; H is the height of the layer sounded. It should be noted that if the quantum yield of fluorescence equals unity, then the fluorescence cross section σ_f is equal to the absorption cross section.

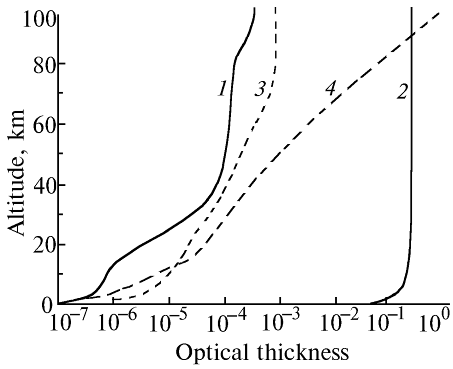

FIG. 2. Optical thickness of the molecular atmosphere at the wavelengths corresponding to excitation and fluorescence wavelengths for the case of ground-based lidar: optical thickness due to aerosol (1) and molecular extinction (2) at the excitation wavelengths; optical thickness due to aerosol (3) and molecular extinction (4) for the fluorescence channel (dashed curves).

Figure 2 shows the altitude dependences of the optical depth of aerosol and gaseous constituents of the atmosphere for excitation and fluorescence channel. As is seen from the figure, the optical thickness for exciting radiation is $\gamma = 0.2$ (curve *2*) and for fluorescence channel $\gamma = 1$ (curve *4*). For $H = 90$ km the signal sought is $E = 2.5 \cdot 10^{-21} \cdot E_0$ photons per pulse. For the Cu-vapor laser pulse repetition frequency of 10 kHz, $E_0 = 10^{16}$ photons, the speed of signal photons arrival is $v \approx 1\ s^{-1}$, that is generally taken as a good signal in comparison to other problems of sensing the upper atmosphere.

TABLE IV. Some of chemical elements, metastable states of whose atoms and ions can be detected using the fluorescence method.

No.	Element	Excitation wavelength, μm	Fluorescence wavelength, μm	Ground-based	Spaceborne
		Atoms			
1	Mn	0.515	0.222	–	+
2	Cu	0.511; 0.578	0.397; 0.393	+	+
3	Sr	6.456	0.408	–	+
4	Ba	1.500	0.553	–	+
5	Au	0.628	0.267	–	+
6	Pb	0.723	0.283	–	+
7	Bi	0.472	0.307	+	+
8	Tm	0.590	0.389	+	+
9	Ca	5.546	0.422	–	+
10	Eu	1.760	0.450	–	+
		Ions			
1	Ca	0.854; 0.855	0.397	+	+
2	Sr	1.033; 1.092	0.408	–	+
3	Ba	0.614	0.455	+	+
4	Eu	1.002	0.372	–	+

Along with the metastable copper atoms, this method is applicable to detection of other elements (see Table IV). For a part of them, sensing can be done from the ground, if the absorption by atmospheric gases is small, or from a spacecraft. In the case, when the atmosphere is not transparent for exciting and (or) fluorescence channels, it is worthwhile to conduct the measurement from space.

Thus, the method of detecting the layers of metastable atoms and ions in the upper atmosphere and near space with the use of ground-based and spaceborne lidars with the laser active media of vapors of chemical elements as radiation sources can be a good instrument for detection and study of these layers if they occur.

ACKNOWLEDGMENTS

The authors thank V.S. Shamanaev for useful advice and discussions.

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