Laser effects at resonant optical excitation of aluminum vapor

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Different laser effects were obtained at atom transitions to the ground state in the visible and the near infrared ranges, when exciting Al vapor by UV resonant laser fields.

At present, a series of papers are available, devoted to interesting spectroscopic effects accompanying the propagation of resonant or quasi-resonant laser radiation in vapor of chemical elements. Vapors of chemical elements are generated, as a rule, through their thermal evaporation in the atmosphere of inert gases.

If the gas density is much larger than the vapor density, the vapor drift to cold windows of cells is hampered. Under these conditions, there exists a possibility of constructing extended cells and considerable vapor densities of difficult evaporable elements. Interesting optical effects appear, when laser radiation passes through extended cells filled with vapor. For example, at optical excitation of one component of the resonant doublet of atoms of alkali elements, superluminescence was observed at the second component.^{1–3}

An asymmetry of the process was observed at resonant optical excitation of the copper atom: laser radiation resonant to the long-wave component of the resonant doublet, excited the both resonant levels; however, only one level was excited at the same excitation of the doublet short-wave component.⁴

Interesting phenomena accompany the propagation of quasi-resonant radiation in vapor of chemical elements as well. Ouasi-resonances between laser radiation and resonant transitions in atoms at 0.1- 10 cm^{-1} detuning are of frequent occurrence. For example, the excimer laser radiation is quasi-resonant to atomic transitions of barium, ytterbium, thulium, lead. Transmission of such radiation through a vapor column of the above-mentioned elements is accompanied by the effective Raman scattering.⁵ Quasi-resonant laser radiation also stimulates stimulates superluminescence at atom transitions not connected by evident processes of excitation transmission from a chosen resonant state.⁶ At a quasi-resonant two-photon excitation of an extended vapor column, the optical ion excitation due to shifted spectrum of an atom can be observed.⁷ The ionization process is so effective and selective that it results in superluminescence at resonance lines and at r-mtransitions in spectrum of ions.⁷

This paper describes laser effects in aluminum vapor at optical excitation of Al atom resonant states.

Figure 1 shows the structure of terms and the Al atomic spectrum corresponding to this structure. The spectrum contains the resonance doublet, resonance triplet, and doublet in the near infrared range. Consider peculiarities of the aluminum atom.



Fig. 1. Structure of terms of Al atom.

One of the peculiarities of Al lower terms is connected with the fact that the short-wave component of the resonance triplet at $\lambda_{Al} = 308.16$ nm is quasi-resonant to the XeCl-laser radiation at $\lambda_{las} = = 308.22$ nm (tuning out of ~6 cm⁻¹).

Another peculiarity is that the metastable state ${}^{2}P^{0}_{3/2}$ is located close to the ground state (the energy

defect relative to the ground state is 112 cm^{-1}). Three levels ${}^{4}P$ in the energy region 30 000 cm⁻¹ are also metastable states, because optical transitions ${}^{4}P - {}^{2}S$ are forbidden in evenness and multiplicity. The Al atom can be considered as an interesting medium of r-m-lasers provided there exists a possibility of obtaining Al atoms in one of sublevels of the ground state. Optical transition from the resonant state ${}^{2}D_{3/2}$ to the metastable ${}^{2}P^{0}_{3/2}$ is resolved but the ratio between statistical weights of levels is not optimal.⁸

The next peculiarity of the Al atom structure is that there exist ${}^{2}D_{3/2,5/2}$ states of the configuration $3s^24p$ close to (in the energy scales) resonant states ${}^{2}P_{1/2,3/2}$. They are connected by optical transitions with the singlet level ${}^{2}S_{1/2}$ (doublet $\lambda = 1312$ nm and 1315 nm in Fig. 1). Laser effects were observed in gas-discharge plasma at such infrared transitions in the structure of the first excited states of atoms having no evident channels of selective electron excitation of upper working states. This fact was first observed when analyzing the inversion mechanisms in the Yb atomic spectrum.^{9,10} The same laser effects were observed at infrared transitions of strontium¹¹ and thulium^{10,12} atoms as well. These observations point to the existence of a sufficiently common inversion mechanism of the first excited atomic states in the pulse electric discharges. At present, two concepts exist on possible mechanism of pumping the first excited but non-resonant atom states in the discharge plasma. One of them¹³ assumes photodissociation in a discharge of molecular components of the working mixture under the action of natural resonance radiation of atoms¹⁴; another concept means the excitation transmission from the resonance state to neighboring ones in super-elastic electron shocks in the discharge plasma.¹⁴

Thus, the above-mentioned analysis has shown that spectroscopic investigations of laser optical excitation of resonant states of the Al atom are useful for understanding phenomena taking place in metal vapor located both in the resonance radiation field and in the discharge plasma.

The experiments were conducted using a setup consisting of a tunable laser, a cell with Al vapor and a system of vapor radiation registration (Fig. 2).



Fig. 2. Schematic representation of the experimental setup: excimer laser 1; dye laser 2; frequency doubler 3; filters 4; long-focus lenses 5, 7; cell with Al vapor 6; mono-chromator 8; system of photoreceivers 9; analog system of signal processing 10.

Cells of two types were used for preparation of Al vapor. In one of the cells the pieces of metal Al were placed into a ceramic tube made of pure aluminum oxide, with an internal diameter of 1 cm and an active part length of 20 cm. As a heat insulator, the zirconium dioxide powder was used, located between the ceramic tube and external cell sheath being a quartz tube of 6.2 cm in diameter. The active area was heated to a necessary temperature by a molybdenum spiral, wound around the ceramic tube, to which the alternating current was applied. Such a construction allows heating active area up to 1500°C. A cell of this type is traditional for metal vapor laser technology.

Its major disadvantages are the presence of the insulator powder, which at high temperatures has a chemical effect on the molybdenum wire (the molybdenum wire temperature is always higher than the temperature inside the ceramic tube), and a longtime outgassing. The former limits the cell service time, the latter results in uncontrolled changes in gas composition inside the cell. The uncontrolled changes in the gas composition can also result in gaseous products arising from chemical interaction of molybdenum with the insulator powder.

A cell of the second type does not contain the zirconium dioxide powder. A system of heat screens is used in it as the heat insulator. Such a system consists of three thin-wall cylinders made of molybdenum foil and located coaxially with the ceramic tube and external quartz sheath. The cylinder diameters are 24, 42, and 54 cm, respectively. The length of all three cylinders is equal to the ceramic tube length (40 cm). Such a construction allows an existence of a uniform heating zone of 18 cm long in the central part of the cell. The increased energy consumption because of high radiation losses is a disadvantage of this construction.

The experiment involved studies of the spectrum of the resonance and quasi-resonance UV radiation scattered by Al vapor and its transformation at varying densities of vapor and buffer gas. Observations were conducted along the cell axis toward the exciting radiation.

As a rule, four lines of the superradiant emittance are observed in the scattered radiation spectrum. Two of them lay in the near infrared range at transitions shown in Fig. 1 and two are in the dark-blue range at transitions to the atom ground state. In the visible range, the superluminous emittance was determined visually using the directional diagram of the scattered radiation. For infrared lines the presence of superluminous emittance regime was determined from augmentation of the signal intensity, when placing a dielectric mirror to the 1.3 μ m range between filter 4 and lens 5.

Studies of intensity distribution over the scattered signal spectrum have revealed a connection of the intensity with the exciting radiation spectrum and the buffer gas density. Figure 3 shows the symmetric behavior of intensities of the visible and infrared lines in pairs at variations of the buffer gas (helium) density.

The pair symmetry implies that at vapor excitation by radiation at $\lambda = 308$ nm the dependence of the

396-nm intensity on the helium density is described by curve 1 (Fig. 3a), and of the 394-nm intensity – by curve 2, while at vapor excitation by 309-nm radiation the inverse intensity distribution is observed for scattered radiation, i.e., the dependences in Fig. 3a change places. The pair symmetry in behavior of intensities at growing helium density is typical of infrared lines of superradiant emittance as well. In particular, the vapor excitation by the radiation at $\lambda = 308$ nm results in excitation of superradiant emittance predominantly at $\lambda = 1312$ nm (Fig. 3*b*, curve 1) as compared to $\lambda = 1315$ nm (Fig. 3b, curve 2); at a change of the exciting radiation wavelength - to predominant superradiant emittance excitation at $\lambda = 1315$ nm, i.e., curves 1 and 2 change places.



Fig. 3. Dependence of line intensities of aluminum vapor superradiant emittance on the helium density.

Dependences in Fig. 3 indicate that the buffer gas participates in the superradiant emittance at Al lines. Probably, one of mechanisms of such a participation is connected with the energy transmission from pump-excited states ${}^{2}P_{1/2,3/2}$ to the levels ${}^{2}D_{3/2,5/2}$ (energy defect is 513 cm⁻¹). It should be also taken into account that there is a common level ${}^{2}S$ for all four observed lines, i.e., both the cascade pumping and the transition competition take place.

To reveal the role of cascade pumping in the emergence of superradiant emittance at resonance transitions, experiments were conducted with the IR cavity modulation at $\lambda = 1315$ and 1312 nm. These experiments have shown that the cascade pumping

is not a basic excitation process for $\lambda = 394$ and 396 nm lines. For superradiant emittance at these lines, an individual pumping process should take place. We believe that the dissociation of the vapor molecular component can be such process, as is shown in Fig. 4.



Fig. 4. Diagram of participation of molecular and atomic vapor components in formation of the scattered radiation spectrum.

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