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Abstract

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1. Commonly used gas lasers are excited by a gas discharge. This method of excitation creates a nonequilibrium population of levels, so that the population of the upper level for the transition on which the laser operates considerably exceeds the equilibrium value of the population. In this note we consider a laser scheme in which the atoms in the upper excited state are in thermodynamic equilibrium with the medium. Therefore, in the laser under consideration a thermal method of excitation may be used. In the laser scheme investigated, atoms in the upper excited state are formed as a result of charge exchange of negative ions on positive ions, so that, owing to this process and to the inverse process considered in detail below, thermodynamic equilibrium of the atoms in the upper excited state with the gas is maintained. The density of atoms in the lower of the states considered is considerably less than its equilibrium value because of radiation.

2. The cross section for charge exchange of a negative ion on a positive ion with the formation of atoms in the ground state and in the given excited state, which is obtained on the basis of the Landau-Zener formula, is determined by the relation [1]

$$\sigma = 4\pi R_0^2 I(\eta), \qquad I(\eta) = \int_1^\infty e^{-\eta x}\left(1 - e^{-\eta x}\right)\frac{dx}{x^3}, \tag{1}$$

$$\eta = \frac{\pi\gamma\alpha^2 A^2}{v}\left(\frac{4}{e}\right)^{1/\gamma} e^{-(\alpha+\gamma)R_0}(\alpha R_0)^{2/\alpha}(2l+1)\Gamma^{-1}\left(\frac{1}{\alpha}+l+1\right)\Gamma^{-1}\left(\frac{1}{\alpha}-l\right).$$

Here v is the relative velocity of collision of the nuclei; $\alpha^2/2, \gamma^2/2$ are the binding energies of the electron in the excited atom and in the negative ion; l is the angular momentum of the electron in the excited atom; $R_0 = 2/(\alpha^2 - \gamma^2)$ is the distance between nuclei at which two terms of the quasimolecule intersect, one of which corresponds to the positive and negative ion, the other to the atoms in the ground and in the given excited state. We assume that the weakly bound electron in the negative ion is in an s-state, and the coefficient A characterizes

the asymptotic behavior of its wave function: the radial wave function of the weakly bound electron, normalized to unity, far from the nucleus is equal to $\chi =\sim A/r\sqrt{2\gamma}\,e^{-\gamma r}$, where r is the distance of the electron from the nucleus. Everywhere, unless specially stated, we use the system of atomic units $\hbar = m_{\text{el}} = e^2 = 1$.

The function $I(\eta)$ for small η is equal to η, and for $\eta \gg 1$ is equal to $\eta^{-1}e^{-\eta}$. Because of the exponential dependence of the quantity η on the distance, the charge-exchange cross section of a negative ion on a positive ion depends sharply on the ionization energy of the excited atom thereby formed. Thus, in the case of charge exchange of a negative oxygen ion on a cesium positive ion, the value of η, obtained on the basis of formula (1), for the formation of an excited cesium atom in the state 8^2S (ionization energy 0.88 eV) proves to be $1.6 \cdot 10^{-5}A^2v^{-1}$, while in the case of formation of 8^2S of a rubidium atom

(ionization energy 0.94 eV) $\eta = 7.9 \cdot 10^{-5}A^2v^{-1}$.* For this reason, upon charge exchange of a negative ion on a positive one, excited atoms are formed in a limited number of excited states. For example, in a mixture of oxygen and cesium at a temperature of about 1000°K, as a result of the process under consideration, excited atoms are formed only in the states 8^2S and 4^2F, with the formation of the first state corresponding to a cross section $7 \cdot 10^{-14}A^2\text{cm}^2$, and of the second to $4 \cdot 10^{-15}A^2\text{cm}^2$.

3. Let us consider a laser operating on a transition between two levels, such that the state of the upper level corresponds to a large formation cross section as a result of charge exchange of a negative ion on a positive one, whereas for the state of the lower level this process may be neglected. We shall assume that the rate at which the upper level is populated as a result of charge exchange is sufficiently large, so that the population of the upper level, like the density of free electrons, is in thermodynamic equilibrium with the gas. Let the density of states in the upper excited state be N_1, and in the lower one N_2, so that in the stationary case

$$dN_1/dt = 0 = N_+N_-\langle v\sigma\ \rangle - N_1N_a\langle v\sigma\ \rangle - \frac{N_1}{\tau_1} - N_1N_e\langle v\sigma_e\rangle, \qquad (2a)$$

$$dN_2/dt = 0 = -N_2/\tau_2 + N_1N_e\langle v\sigma_e\rangle. \qquad (2\ )$$

Here N_e, N_a, N_+, N_- are the densities of electrons, atoms, positive and negative ions; $1/\tau_1, 1/\tau_2$ are the probabilities of transition from the given level as a result of radiation; $\langle v\sigma\ \rangle, \langle v\sigma\ \rangle$ are the particle-distribution-averaged products of the relative collision velocity and the cross section for formation of an excited atom 1 as a result of charge exchange or its destruction; $\langle v\sigma_e\rangle$ is the corresponding quantity for transition from state 1 to state 2 as a result of collision with a free electron (we neglect the reverse transition). In addition, in equations (2) we have neglected transitions between neighboring excited states due to collisions

with atoms, since such transitions are adiabatically unlikely. Since the atom in excited state 1 is in equilibrium with the gas, the first two terms in (2a) are equal, and each of them is much larger than the third term, i.e.

$$N_a\langle v\sigma \quad \rangle \gg 1/\tau_1. \tag{3a}$$

On the other hand, since the population of level 2 is smaller than that of level 1, then for an optically thin gas layer

$$N_e\langle v\sigma_e\rangle \ll 1/\tau_2. \tag{3 }$$

Let us determine what these conditions correspond to if the gas consists of a mixture of oxygen and cesium at a temperature of 1000°K, and the laser operates on the transition between the cesium levels $8^2S_{1/2}-7^2P_{3/2}$. In this case the first condition ($\sigma \quad \sim 10^{-13}$ cm^2, $1/\tau_1 = 5\cdot 10^6$ sec^{-1}) gives the oxygen molecule density $N_a \gg 10^{15}$ cm^{-3}, and the second ($\sigma_e \sim 10^{-13}$ cm^2, $1/\tau_2 = 7\cdot 10^7$ sec^{-1}) gives $N_e \ll 5\cdot 10^{13}$ cm^{-3}. Thus, conditions (3a) and (3) require that the gas be weakly ionized. These conditions correspond to the rate of formation of state 1 by charge exchange being greater than as a result of recombination of free electrons. In addition, because of the low density of positive ions, the characteristic time of destruction of negative ions as a result of charge exchange on positive ions considerably exceeds the characteristic time of destruction of negative ions by collisions with atoms and electrons. Therefore thermodynamic equilib–

* In these calculations the electron affinity energy of the oxygen molecule was taken to be 0.44 eV ([2]).

The method of obtaining this value for the electron affinity energy of O_2 seems to us more convincing than the other results ([3–5]), which contradict it.

···of negative ions with the gas is not disturbed by charge exchange on positive ions even in the case where the excited atoms thereby formed are not in equilibrium with the gas because of radiation.

4. Criteria (3) make it possible to implement the laser scheme considered in two ways. In the first of them, the positive and negative ions belong to one and the same kind of particle. An example of such a laser is a laser operating on the sodium transitions $6^2S_{1/2}$—5^2P, $6^2S_{1/2}$—4^2P. Since the electron binding energy in the negative sodium ion is 0.35 eV, the value $A = 1.7$ ([6]), and the cross section for formation of the $6^2S_{1/2}$ state at a temperature of 1000° K is $\sim 1.2\cdot 10^{-14}$ cm^2.

Table 1

Examples of impurities to molecular oxygen that make it possible to realize a laser on a negative ion, if the electron affinity to oxygen is 0.44 eV

Impurity	Transition on which gain is achieved	Photon wavelength for this transition, Å	Impurity	Transition on which gain is achieved	Photon wavelength for this transition, Å
Li	4^2D—3^2P	17551.6	Zn	5^1D_2—5^1P_1	
Li	4^2F—3^2D	18697.0	Zn	5^1D_2—4^1P_1	4629.81
Li	4^2S—3^2P	24467.0	Zn	4^3F—$4^3D_{1,2,3}$	16483.7; 16490.3; 16498.6
Na	5^2S—4^2P		Cd	6^1D_2—6^1P_1	
Na	4^2D—4^2F		Cd	6^1D_2—5^1P_1	4662.35
Na	4^2F—3^2D	18459.5	Cd	4^3F—$5^3D_{1,2,3}$	16401.5; 16433.8; 16482.2
K	6^2S—$5^2P_{1/2}$, $5^2P_{3/2}$		Hg	5^1F_3—6^1D_2	16920.2
K	4^2F—$5^2P_{1/2}$, $5^2P_{3/2}$		Hg	7^1D_2—7^1P_1	
K	4^2F—3^2D	15165.8	Hg	7^1D_2—6^1P_1	4347.49
Rb	7^2S—6^2P		Hg	5^3F—$6^3D_{1,2,3}$	16942.0; 17109.9; 17072.4
Rb	5^2D—6^2P		Al	4^2F—3^2D	11255.0
Rb	4^2F—4^2D	13444.0	Al	5^2P—5^2S	
Mg	4^1D_2—3^1P_0	5528.46	Al	$5^2P_{1/2}$—$4^2S_{1/2}$	6698.73
Mg	5^3P—3^3D		Al	$5^2P_{3/2}$—$4^2S_{1/2}$	6696.06
Ca	6^3P—4^3D		Tl	5^2F—$6^2D_{5/2}$	16340.3
Ba	7^3D—7^3P		Tl	5^2F—$6^2D_{3/2}$	16123.0
			Tl	8^2P—8^2S	
			Tl	$8^2P_{3/2}$—$7^2S_{1/2}$	6549.84
			Tl	$8^2P_{1/2}$—$7^2S_{1/2}$	6713.80

The greatest number of possibilities corresponds to the second case, in which the positive and negative ions belong to different kinds of atoms. The laser considered above, operating on a mixture of oxygen and cesium, belongs to this case. Table 1 gives a further series of transitions on which a gas laser can operate if its working substance is oxygen and an impurity of other atoms. It is assumed that the emitted line is not absorbed in oxygen.

A gas laser operating according to the scheme considered can be excited not by a discharge, as is usually used, but also by a thermal method*. This may provide a number of technical advantages. Thus, impurity atoms can be obtained by evaporation from a heated surface. In addition, the high temperature of the gas makes it possible to obtain a mixture of oxygen with other···

* It should be noted that the cross section for formation of an excited atom in the case considered is one or two orders of magnitude greater than the atomic

cross section. Therefore excited atoms are formed even in a mixture of gases (for example, cesium and oxygen) that enter into a chemical reaction. This is connected with the fact that the cross section of the chemical reaction is considerably smaller than the cross section for formation of excited atoms.

substances. Under normal conditions, however, many of the impurity atoms listed in Table 1 enter into a chemical reaction with oxygen, forming oxides.

The laser scheme considered is practically applicable if the electron affinity energy of the molecules of the working gas amounts to several tenths of an electron volt. Unfortunately, at present the value of the electron affinity for such atoms and molecules (and, all the more, the value of A) is not known with sufficient reliability. Therefore there is no guarantee that the laser operating mechanism investigated will be realized precisely on the transitions indicated in Table 1.

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Note: Figure translations are in progress. See original paper for figures.