A GAS LASER BASED ON MOLECULAR IONS

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Abstract

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B. M. SMIRNOV

A GAS LASER BASED ON MOLECULAR IONS

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1. Of practical importance is the creation of a laser that generates short-wavelength photons. In the present work a scheme is proposed for such a laser, operating on transitions between the ground state and a resonantly excited state of an inert-gas atom.

Let us consider a mixture of inert gases at not very low pressures, such that the ions formed in such a gas are molecular. The mechanism of recombination of molecular ions and electrons has been discussed in sufficient detail in the literature (^{1}) and consists of the following. An electron is captured by a molecular ion, so that the electronic state of the molecule formed, at the given internuclear separation, is autoionizing. The nuclei then fly apart, and in doing so reach internuclear separations at which the term of the molecular state under consideration intersects the term of the molecular ion, i.e., the boundary of the continuous spectrum. At larger internuclear separations the electronic state of the molecule under consideration ceases to be autoionizing, and if it has not had time to decay into an electron and an ion before the intersection separation has been reached, then the molecule dissociates into two atoms (a molecule and an atom), with one of the atoms formed being in an excited state.

Since the interaction potential of an excited atom with an atom is smaller than the interaction potential of an ion with an atom, the binding energy of the valence electron in the excited atom formed upon recombination of an electron and a molecular ion only slightly exceeds the dissociation energy of the molecular ion. The dissociation energy of the molecular ion is much smaller than the ionization potential of the atoms. Thus, recombination of an electron and a molecular ion leads to the formation of excited atoms that are in certain excited states. The number of these states is very limited, which is connected with the small number of autoionizing states of the molecule into which the electron can be captured. One of these states may be chosen as the upper level of the laser.

2. Let us determine the operating conditions of the laser under consideration. Let the density of atoms in the upper level of the laser transition be N_b, and in the lower level of the laser transition N_0, with the transition to the lower state occurring as a result of radiation whose frequency is $1/\tau$. Atoms in the lower state are destroyed as a result of reactions with gas particles, the frequency of such transitions being ν. Under these conditions the balance equation for atoms in the lower state has the form

$$dN_0/dt = N_b/\tau - N_0\nu.$$

Hence, in the stationary case we obtain the condition for laser operation ($N_b > N_0$), assuming that the statistical weights of the atoms in the upper and lower states are equal to unity:

$$\nu\tau > 1. \tag{1}$$

Thus, the rate at which the lower state enters the chemical reaction must exceed the probability per unit time of radiation from the upper state.

3. In our opinion, an important advantage of the type of laser under consideration is that it makes it possible to create an inverted population at highly excited levels and to generate ultraviolet radiation. We shall consider one variant of the proposed laser, which operates on transitions between the ground and resonantly excited states of an atom. For example, an inert gas can be used as impurity atoms. Then the energy of the generated photons will exceed 10 eV, so that such a laser is of great practical interest.

Table 1

Molecular ion	He_2^+	Ne_2^+	Ar_2^+	Kr_2^+	Xe_2^+
Dissociation energy, eV	2.24 (2)	1.4 (3)	1.1 (4)	1.13 (5)	1.0 (5)
Ionization potential of the atom, eV	24.59	21.56	15.76	14.0	12.1

Table 1 gives the dissociation energies of molecular ions of inert gases and, for comparison, the ionization potentials of the corresponding atoms. From it one may conclude that recombination of electrons and molecular ions produces strongly excited atoms. In order to determine the probability of formation of a given excited state, a detailed spectroscopic analysis of the given plasma would have to be carried out. In this respect some information has been obtained for helium plasma.

It has been shown (^{6}, ^{7}) that in a decaying helium plasma the density of excited states with ionization energy 0.3–1.5 eV is proportional to the square of the electron density, i.e., these excited states are formed as a result of recombination of an electron and a molecular ion. Under several other conditions recombination of an electron and a molecular ion leads mainly to the appearance of an excited helium atom in the 3^3P state (ionization energy 1.47 eV). The data concerning plasmas of other inert gases (^{3}, ^{9}) are much less complete, but they indicate the important role of the recombination mechanism under consideration in the distribution of excited atoms over states. Because of the lack of information we cannot determine which resonantly excited state of the atom corresponds to the upper level of the laser transition; a rough estimate gives that the ionization energy of this atom is of the order of the dissociation energy of the molecular ion.

The density of resonantly excited atoms is much smaller than the density of atoms in the ground state. Therefore generation can be obtained only on the wing of a line.* In this connection it should be taken into account that the atoms formed as a result of dissociative recombination possess kinetic energy of the order of the dissociation energy of the molecular ion. This follows from the recombination mechanism and is confirmed experimentally (for example, in the work of Connor and Biondi (^{3}) for neon plasma). Atoms in the ground state correspond to a Maxwellian velocity distribution function. Therefore a necessary condition for the creation of an inverted population, corresponding to atomic velocities equal to the mean velocities of the separating atoms, is

$$\bar{\varepsilon}/T > \ln N_0/N_b, \tag{2}$$

where $\bar{\varepsilon}$ is the mean energy of the separating atoms; T is the gas temperature; N_0, N_b are the densities of atoms in the ground and resonantly excited states, respectively. We shall show that in the real case this condition is fully satisfied. Let us determine the density of excited atoms of an inert gas from the balance equation

$$N_e N_i \alpha - N_0 N_b \langle v\sigma_{\text{ion}}\rangle = 0,$$

where a realistic value for the recombination coefficient α, leading to the formation of an atom in the given excited state, is $\alpha \sim 10^{-8}$ cm^3/s, the collision velocity of atoms is $v \sim 10^5$ cm/s, and the cross section for associative ionization σ_{ion} in collisions of atoms in the ground and excited states is $\sim 10^{-15}$ cm^2. Let us take $N_0 \sim 10^{16}$ cm^{-3} and the density of charged particles in the plasma $N_e = N_i \sim 10^{11}$ cm^{-3}. Then we obtain $N_b \sim 10^8$ cm^{-3}, i.e., condition (2) gives $\varepsilon/T > 20$. Since the mean energy of the separating atoms is $\varepsilon \sim 1$ eV, we

* Such lasers, operating on the wing of a spectral line, have been discussed more than once in the literature (^{10}).

can create a population inversion at room temperatures, and it is within our capabilities also to lower the gas temperature.

Thus, we are able to create a population inversion on the resonance lines of atoms for certain atomic velocities. A resonantly excited atom is stopped as a result of transfer of excitation after its collision with an identical atom in the ground state. The cross section of this process is equal to (11)

$$\sigma_{\text{per}} = 2.26\pi|(D_x)_{0b}|^2/\hbar v,$$

where $(D_x)_{0b}$ is the matrix element of the dipole moment of the atom, taken between the ground and resonantly excited atomic states. The process of excitation transfer creates fast atoms in the ground state, which can lead to the destruction of the population inversion. Therefore, the operating condition for the type of laser under consideration is

$$\tau v_{\text{per}} < 1, \tag{3}$$

where $1/\tau$ is the radiation frequency, and $v_{\text{per}} = N_0 \cdot 2.26\pi|(D_x)_{0b}|^2/\hbar$ is the excitation-transfer frequency. Condition (3) gives, for the density of atoms in the ground state,

$$N_0\lambda^3 < 1, \tag{4}$$

where $2\pi\lambda$ is the wavelength of the emitted photon.

Let us take condition (1) into account. The frequency ν corresponds to the slowing down of fast atoms in the ground state that were formed as a result of de-excitation of excited atoms. These atoms are slowed as a result of elastic collisions with gas atoms, and the cross section of an elastic collision σ_{el}, which is of the order of the atomic cross section, is much smaller than the excitation-transfer cross section. It follows from this that conditions (1) and (3) prove incompatible if we want to obtain a population inversion in a one-component gas. A population inversion is possible only in a two-component system, with the inversion being created on impurity atoms. According to conditions (1) and (3), the densities of atoms of the main gas N_g and of impurity atoms N_0 must be in the ratio

$$N_g/N_0 > \langle v\sigma_{\text{per}}\rangle/\langle v\sigma_{\text{el}}\rangle, \tag{5}$$

where the averaging is over the Maxwellian distribution of atoms.

If, as the amplifying medium, one chooses an inert gas consisting of atoms A with an admixture of atoms B, then in such a system molecular ions of the type A_2^+, AB^+, or B_2^+ can be formed, and, by changing the conditions, we can achieve predominance of ions of a definite kind. To create a population inversion it is more convenient to use recombination of the molecular ion AB^+ than of the ion B_2^+. Indeed, upon recombination of an electron and the molecular ion B_2^+, two fast atoms with identical kinetic energy are formed, one of which is in the excited state and the other in the ground state; i.e., the state of one of them corresponds to the upper laser level, and the state of the other to the lower one. It is possible that this still will not preclude the possibility of creating a population inversion, since the law governing the slowing of atoms in the ground and excited states is different. However, the circumstance indicated significantly complicates the creation of an invers-

population as a result of recombination of B_2^+ ions. In the recombination of AB^+ ions, no such difficulty arises.

It is possible that a laser of this type is better suited for operation in a pulsed regime, since it is then easier to maintain a low gas temperature, which is necessary for the formation of molecular ions, and it is also possible to vary the mean electron energy more freely, i.e., to obtain more varied conditions. In the steady-state regime, the mean electron energy can be reduced by adding a molecular gas.

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CITED LITERATURE

[1] D. Bates, A. Dalgarno, in *Atomic and Molecular Processes*, ed. by Bates, Moscow, 1964, p. 224.
[2] P. N. Reagan, S. C. Brown, Phys. Rev., **132**, 304 (1963).
[3] T. R. Connor, M. A. Biondi, Phys. Rev., **140**, 778 (1965).
[4] W. Kaul, R. Taubert, Zs. Naturforsch., **17a**, 88 (1962).
[5] J. A. R. Samson, R. B. Cairns, J. Opt. Soc. Am., **56**, 1140 (1966).
[6] M. A. Biondi, T. Holstein, Phys. Rev., **82**, 962 (1951).
[7] W. A. Rogers, M. A. Biondi, Phys. Rev., **134A**, 1215 (1965).
[8] W. Kaul, P. Seyfield, R. Taubert, Zs. Naturforsch., **18a**, 432 (1963).
[9] F. W. Lampe, G. G. Hess, J. Am. Chem. Soc., **86**, 2952 (1964).
[10] G. Gould, Proc. III Intern. Congress on Quant. Electr., **1**, 1965, p. 457; M. L. Sosinskii, E. N. Morozov, Optics and Spectroscopy, **18**, 510 (1966).
[11] T. Watanabe, Phys. Rev., **138A**, 1573 (1965); **140A**, 135 (1965).

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