Ionization-induced bleaching of a gas

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An analysis is made of the ionization of a gas in the field of a resonant electromagnetic wave and of the propagation of this wave accompanied by such ionization. Formation of a plasma is found to be due to initial associative processes and due to the subsequent effects of electrons which acquire energy by superelastic collisions with excited atoms. The change in the law of propagation is due to a reduction in the density of the absorbing particles and an increase in the probability of quenching of the excited atoms.

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§1. INTRODUCTION

High-power resonant radiation interacting with a medium causes saturation in which the populations of the ground n_1 and excited n_2 states are linked by[1]

$$n_2 \approx g_2 n_1 / g_1, \tag{1}$$

where g are the statistical weights of the levels involved. Bleaching of a gas and emission of radiation of intensity I_0 traveling a distance L is observed in this saturation; the distance can be found from the energy balance[2]

$$L \approx \left(1+\frac{g_1}{g_2}\right)\frac{I_0}{A_{21}n_1(0)}, \tag{2}$$

where A_{21} is the probability of spontaneous decay and $n_1(0)$ is the initial concentration of the atoms.

We shall consider propagation of resonant radiation which is accompanied by the ionization of a gas. A plasma is formed because electrons colliding superelastically with excited atoms acquire an energy which is dissipated subsequently in the excitation, ionization, and heating of the gas. Formation of the primary electrons may be due to, in particular, associative processes. Transition of the gas to the plasma state reduces the number of absorbing particles, but increases the probability of quenching of excited atoms by electron impact. Predominance of the first process results in additional bleaching of the gas, whereas in the case of the latter process a darkening of the gas can be expected.

§2. TRANSPORT EQUATION FOR ELECTRONS AND GAS IONIZATION

An electron in the field of an electromagnetic wave acquires energy as a result of inverse bremsstrahlung or because of the absorption of photons by atoms and subsequent electron quenching of these atoms. The second process predominates in a weak resonant field.[3] Let $n(\varepsilon, t)$ describes the distribution, at a moment t, of the energy of electrons which acquire energy by superelastic collisions of frequency ν_{21} and excite a resonant level at a frequency ν_{12}. The transport equation for electrons is

$$\frac{\partial n(\varepsilon,t)}{\partial t}=\{\nu_{21}(\varepsilon-E)n(\varepsilon-E,t)-\nu_{21}(\varepsilon)n(\varepsilon,t)$$
$$-\nu_{12}(\varepsilon)n(\varepsilon,t)+\nu_{12}(\varepsilon+E)n(\varepsilon+E,t)\}-\frac{\partial}{\partial\varepsilon}\frac{2m}{M}\nu_m\varepsilon n(\varepsilon,t)+Q. \tag{3}$$

The term Q is associated with the ionization and recombination processes; $E=\hbar\omega$ is the photon energy; ν_m is the frequency of elastic collisions; m and M are the masses of an electron and an atom.

We shall solve Eq. (3) using a method suggested by Zel'dovich and Raizer.[4] We shall assume that $\hbar\omega/\varepsilon \ll 1$, and we shall expand in terms of this small parameter. To within terms of the second order of smallness, we find from Eq. (3) that

$$\frac{\partial n(\varepsilon,t)}{\partial t}=-\frac{\partial j}{\partial\varepsilon}+Q,\quad j=-D\frac{\partial n(\varepsilon,t)}{\partial t}+n(\varepsilon,t)(u-u_c), \tag{4}$$

$$D=\frac{1}{2}E^2[\nu_{21}(\varepsilon)+\nu_{12}(\varepsilon)],$$
$$u=E[\nu_{21}(\varepsilon)-\nu_{12}(\varepsilon)]-\frac{dD}{d\varepsilon},\quad u_c=-2m\nu_m\varepsilon/M. \tag{5}$$

We shall assume that resonant radiation saturates a transition in atoms. We shall apply the principle of detailed equilibrium which determines the relationship between the cross sections for the excitation and quencing of atoms by electron impact[5]:

$$g_1\varepsilon\sigma_{21}(\varepsilon)=g_1[\varepsilon+E]\sigma_{12}(\varepsilon+E). \tag{6}$$

Then, we obtain

$$\nu_{21}(\varepsilon)=[(\varepsilon+E)/\varepsilon]^{1/2}\nu_{12}(\varepsilon+E). \tag{7}$$

Applying Eq. (7) and expanding in Eq. (5) in terms of the small parameter, we find that

$$D=E^2\nu_{21}(\varepsilon),\quad u=D(\varepsilon)/2\varepsilon. \tag{8}$$

We shall now discuss a situation in which the main channel for the electron energy losses is in the form of elastic collisions. Then, the solution of Eq. (4) under steady-state conditions is

$$n(\varepsilon)=c\varepsilon^{1/2}\exp\left\{-\frac{(2m)^{1/2}mg_1}{ME^2g_2n_1(0)}\int_0^\varepsilon\frac{\nu_m\varepsilon^{1/2}}{\sigma_{21}(\varepsilon)}d\varepsilon\right\}, \tag{9}$$

where c is the normalization constant. If the integrand is independent of the electron energy and equal to c', we obtain the Maxwellian energy distribution of electrons with a temperature

$$T_e=ME^2g_2n_1(0)/g_1(2m)^{1/2}mc'k, \tag{10}$$

where k is the Boltzmann constant. If the electron energy is expended almost entirely in heating the gas, it becomes weakly ionized, so that we shall consider the opposite situation when the the energy losses are due to the excitation and ionization of the gas.

We shall consider a model of a three-level atom with ground, resonant, and ionized states. Bearing in mind that the time for the establishment of the electron energy distribution is much less than the ionization time, we shall represent the function $n(\varepsilon, t)$ in the form

$$n(\varepsilon, t)=n(\varepsilon)n(t) \tag{11}$$

and we shall subject $n(\varepsilon)$ to the normalization conditions

$$\int_0^\infty n(\varepsilon)d\varepsilon=1. \tag{12}$$

Then, $n(t)$ describes the density of electrons and ions since the creation of doubly charged ions will be ignored. We shall consider a gas of low densty and assume that the loss of charged particles occurs inside the volume under consideration because of radiative recombination with a coefficient α. Consequently, we find that

$$Q=-\alpha n(\varepsilon)n^2(t). \tag{13}$$

The process of ionization of a gas at a frequency ν_i will be allowed for by postulating an intense "sink"[4] in the energy range $\varepsilon=I_1$, where I_1 is slightly greater than the ionization potential of an atom which is initially in an excited state. Bearing in mind that the population of the resonance level is high, we shall ignore direct ionization. We can now describe the ionization process by

$$\frac{dn(t)}{dt}=\nu_i n(t)-\alpha n^2(t). \tag{14}$$

The energy spectrum of electrons subject to Eqs. (3), (11), and (14) is found from

$$\nu_i n(\varepsilon)+\frac{d}{d\varepsilon}\left(-D\frac{dn(\varepsilon)}{d\varepsilon}+un(\varepsilon)\right)=0 \tag{15}$$

and the relevant boundary conditions

$$n(I_1)=0, \tag{16}$$

$$j(0)=2j(I_1), \tag{17}$$

which describes the presence of a concentrated sink and creation of slow electrons associated with the ionization process.

We shall model the excitation cross section of a resonant level by

$$\sigma_{12}(\varepsilon)=\begin{cases}0, & \varepsilon\leqslant E\\ c_1(\varepsilon-E)^{1/2}, & \varepsilon>E.\end{cases} \tag{18}$$

We then obtain from Eq. (8)

$$D=E^2n_1c_1(2/m)^{1/2}\varepsilon, \quad u=E^2n_1c_1/(2m)^{1/2}. \tag{19}$$

In this case the solution of Eq. (15) has the form

$$n(\varepsilon)=c_2\,\mathrm{sh}[2.18(1-(\varepsilon/I_1)^{1/2})], \tag{20}$$

$$\nu_i=Sn_2, \quad S\approx1.2c_1\left(\frac{2}{m}\right)^{1/2}E\frac{E}{I_1}. \tag{21}$$

The constant c_2 is found from the normalization condition (12).

The evolution of the electron density subject to allowance for strong ionization is found from Eq. (14) and is of the form

$$n(t)=n_e(0)\exp[n_1(0)S't]\left\{1-\frac{n_e(0)}{n_1(0)}\left(1+\frac{\alpha}{S'}\right)[1-\exp(n_1(0)S't)]\right\}^{-1}, \tag{22}$$

$$S'=g_2S/(g_1+g_2), \tag{23}$$

where $n_e(0)$ is the initial electron density. The steady-state value of the electron density is

$$n(\infty)=n_1(0)S'/(S'+\alpha), \tag{24}$$

and the condition for the conservation of the number of particles is

$$n(\infty)+n_1+n_2=n_1(0). \tag{25}$$

We shall apply this approach to atoms with a distributed system of energy levels (Li, Na, K, Rb, Cs, Mg, Ca, Ba, etc.). We shall obtain specific estimates for the Cs vapor excited by radiation of the $\lambda=894.3$ nm wavelength, which is characterized by $E/I_1\approx0.3$. Employing the experimental results of Refs. 6 and 7, we obtain $\alpha\approx4\times10^{-10}$ cm^3/sec and $S\approx4\times10^{-7}$ cm^3/sec. Primary electrons are formed by the process of associative ionization, whose cross section is 6×10^{-18} cm^2 (Ref. 8). The predominant role of the electron impact in the gas ionization begins from $n_e(0)\approx10^{-6}n_1(0)$. The steady-state electron density is $n(\infty)\approx0.999n_1(0)$. This possibility of ionization of the gas has already been pointed out in Ref. 9. We shall consider only qualitatively the atoms with a system of levels concentrated near the ionization potential. The ionization frequency can be estimated from

$$\nu_i\approx\frac{1}{I_1}\frac{\Delta\varepsilon}{\Delta t}, \tag{26}$$

$$\frac{\Delta\varepsilon}{\Delta t}\approx E[\nu_{21}(I_1-E)-\nu_{12}(I_1)]. \tag{27}$$

Using Eqs. (7) and (19), we obtain from Eq. (26) the expression

$$\nu_i\approx(2/m)^{1/2}c_1E. \tag{28}$$

This expression is similar to Eq. (21), because in this case we have $E\sim I_1$ and its physical meaning is that all the energy acquired by an electron is lost in the excitation and ionization of the gas.

§3. PROPAGATION OF RADIATION

Ionization of a gas reduces the number of atoms and increases the probability of quenching of the excited state. This alters the character of the propagation of radiation. We shall write down the radiative transfer equation for the steady-state conditions:

$$\frac{dI}{dx}=I(\sigma_{21}'n_2-\sigma_{12}'n_1), \tag{29}$$

where I is the radiation intensity (photons·cm^{-2}·sec^{-1}) and σ_{12}' and σ_{21}' are the absorption and stimulated emission cross sections. The populations of the excited and normal states are related by the following expression which allows for collisional and radiative processes:

$$\frac{n_2}{n_1}=\frac{n(\infty)K_{12}+\sigma_{12}'I}{\sigma_{12}'I+n(\infty)K_{21}+A_{21}} \tag{30}$$

$$K_{ij}=\int_0^\infty \nu_{ij}(\varepsilon)n(\varepsilon)d\varepsilon. \tag{31}$$

In writing down Eq. (30) an allowance has been made for the fact that in the case of cesium the probability of ionization obeys $S\ll K_{21}$, i.e., it is less than the probability of superelastic collisions.

Bearing in mind Eqs. (25) and (30), we find that under saturation conditions the radiative transfer equation (29) can be written in the form

$$\frac{dI}{dx}=-\frac{(n_1(0)-n(\infty))}{(1+g_1/g_2)}\left[A_{21}+n(\infty)\left(K_{21}-\frac{g_1}{g_2}K_{12}\right)\right]. \qquad (32)$$

The first factor on the right-hand side of Eq. (32) represents the reduction in the number of atoms due to ionization, and the second term describes the effective probability of quenching of the excited state. If $n(\infty)=0$, Eq. (30) describes the usual bleaching of a medium over a distance given by Eq. (2). The coefficient

$$R=\left(1-\frac{n(\infty)}{n_1(0)}\right)\left(1+\beta\frac{n(\infty)}{n_1(0)}\right),\quad \beta=\frac{n_1(0)\,(g_2K_{21}-g_1K_{12})}{g_2A_{21}} \qquad (33)$$

allows for the change in the absorption law and the new bleaching length L' is given by

$$L'=L/R. \qquad (34)$$

For

$$R<1,\ \ L'>L \qquad (35)$$

we can expect additional bleaching of a gas. If the opposite inequalities are obeyed, the medium becomes darker. The condition for the ionization bleaching (35) has a clear physical meaning:

$$\frac{n_1(0)\,(g_2K_{21}-g_1K_{12})}{g_2A_{21}}<\frac{S'+\alpha}{\alpha}=\frac{n_1(0)}{n_1(0)-n(\infty)}. \qquad (36)$$

In the case of additional bleaching the increase in the probability of the quenching collisions should be more than compensated by the reduction in the number of absorbing particles. The ionization state has a large statistical weight and is in a sense metastable since the probability of its decay governed by the radiative recombination is low. Consequently, in this state the particles coalesce and this reduces the absorption of the resonant radiation. In the case of cesium with $n_1(0)=10^{13}$ cm^{-3} we have $R\approx 10^{-3}$.

We shall now consider the transient process. During the first stage the radiation excites the gas and equalizes the populations of the linked levels. Then, associative processes create primary electrons whose energy spectrum is established rapidly and has a quasi-steady form throughout the whole process. Gas is ionized and if $R\leqslant 1$ the ionization occurs throughout the process, which is followed by additional bleaching and further propagation of the radiation. The velocity of propagation of the ionization-bleached zone can be estimated from

$$v\sim L/\tau,\quad \tau\approx[S'(n_1(0))]^{-1}\ln[n_1(0)/n_e(0)], \qquad (37)$$

where τ is the characteristic ionization time. The process of ionization is governed by electron impact because the initial associative processes take much less time. If $I_0=7.5\times10^{21}$ photons·cm^{-2}·sec^{-1}, $n_1(0)=10^{13}$ cm^{-3}, and other parameters are as given above, we find that $v\sim 3\times10^6$ cm/sec.

In the situation when $\beta\leqslant 1$ we find that $R\leqslant 1$ throughout the ionization process and the medium becomes bleached. If $\beta<1$, the medium first darkens to the value

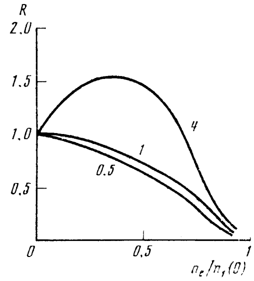

FIG. 1.

$$n_e(t)=\frac{\beta-1}{2\beta}n_1(0), \qquad (38)$$

when R reaches its maximum value

$$R=(\beta+1)^2/4\beta, \qquad (39)$$

which is followed by a fall of R to

$$R^*=\alpha[\alpha+(\beta+1)S']/(S'+\alpha)^2, \qquad (40)$$

and if $R^*<1$, the medium is ionization-bleached, Figure 1 shows the behavior of R for different values of β.

We must point out once again that the possibility of ionization bleaching demonstrated in this paper is due to a change in the aggregate state of the medium during illumination. This situation must be allowed for in investigations of the interaction of resonant radiation with the gases, particularly under steady-state conditions. A similar effect appears also when the loss of charged particles is due to diffusion processes. A three-level model of an atom used in our analysis is subject only to quantitative restrictions, because in real situations a multistage process has not only an undesirable influence resulting from additional radiative losses but it also increases the ionization coefficient.

Calculations carried out in the approximation of the Maxwellian distribution of the electron energy also demonstrate the possibility of appearance of the ionization bleaching in the absence of saturation. This effect reduces the energy losses in the process of propagation and can be used for more effective transfer of the radiation energy.

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