Radiative collision generation of sound in gases by single-pulse optical excitation

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The generation of sound in the field of a single light pulse, due to the effect of light induced drift (LID) of gases, is investigated theoretically. The spectral characteristics of the acoustic LID signal and their relation with the parameters of the medium and the radiation are analyzed. The existence of two mechanisms of generation of LID sound is established: selective and diffusion. The restrictions on the medium and radiation are less stringent for the diffusion mechanism than for the selective one. In this sense, the diffusion mechanism is more universal. The calculation may serve as the theoretical basis of a method of determining the transport scattering cross sections of excited atomic particles by using pulsed lasers and the high-sensitivity optical-acoustic spectroscopy technique.

1. INTRODUCTION

In recent years, the interest of investigators has been drawn to the phenomenon of light-induced drift (LID) of gases[1–3], which consists in the stimulation, by the radiation of a traveling light wave, of counter flows of absorbing and buffer gases. In connection with the possibility of a strong macroscopic manifestation, the LID effect can find application in physical investigations and in technology (for example, for the separation of isotopes[1)]). To obtain the maximum drift velocities, certain requirements must be satisfied by the following characteristics of the inducing radiation: spectral width, detuning from resonance, intensity, and pulse-repetition frequency in the case of a pulsed-periodic excitation.[5,6] These requirements are not always easily satisfied, since the number of experimental researches devoted to the LID phenomenon is still relatively small[7–9] (see also the bibliography in Ref. 9). For a more task-oriented application of continuous and pulsed lasers, and to obtain a strong LID effect, information is necessary on the transport collision cross sections of excited atomic particles, information very scanty at the present time.[10,11] Along with this, this information is of interest for the physics of atomic and molecular collisions and spectroscopy. There exists the attractive possibility of obtaining data on the collision cross sections of excited particles when a gas is acted upon by a single radiation pulse. This allows us to expand the experimental possibilities, including in the "arsenal" of sources of light the broad class of single-pulse lasers existing at the present time, and use the methods of frequency tuning developed for them. The basis of the corresponding experimental method can be the effect of radiation-collision generation of sound[2)] in single-pulse optical excitation, an effect due to LID in gases. The present work is devoted to the theoretical analysis of this phenomenon.

2. GENERAL RELATIONS

Let us consider the action of a light wave on a mixture of an absorbing and a buffer gas. For the energy spectrum of the absorbing particles, we limit ourselves to the model of two nondegenerate states m and n (n is the ground state). The evolution of the medium is described by the set of kinetic equations

$$\hat{A}f_\mu = S_\mu, \quad \hat{A} = \partial_t + \mathbf{v}\partial_\mathbf{r}, \quad \mu = 1, b, \tag{2.1}$$

where f_μ and S_μ are the distribution function (dependent on the velocity $\mathbf{v}$ and the radius vector $\mathbf{r}$ with components x, y, z and on the time t) and the collision integral of the component μ; subscripts $\mu = 1$ and $\mu = b$ label quantities referring to absorbing gas and to the buffer, respectively. In the process of interaction with the radiation, part of the absorbing particles goes into the excited state m. We take into consideration the difference of the kinetics of the excited particles from their kinetics in the ground state n, representing S_1 in the form

$$S_1 = S_n + S_m. \tag{2.2}$$

The collision integrals S_φ ($\varphi = n, m, b$) have the following structure:

$$S_\varphi = \sum_\psi S_{\varphi\psi}, \quad \psi = n, m, b, \tag{2.3}$$

where the component $S_{\varphi\psi}$ describes the collisions of particles of type φ with particles of type ψ. From the laws of conservation of the number of particles, momentum, and energy in the collisions, we extract the following properties of the partial collision integrals $S_{\varphi\psi}$ (see, for example, Ref. 13):

$$\int S_{\varphi\psi}\, d\mathbf{v} = 0, \qquad \int (Q_\varphi S_{\varphi\psi} + Q_\psi S_{\psi\varphi})\, d\mathbf{v} = 0,$$
$$Q_\varphi = M_\varphi \mathbf{v}, \quad {}^1/_2 M_\varphi v^2 + \varepsilon_{\text{int}\,\varphi}. \tag{2.4}$$

Here M_φ and $\varepsilon_{\text{int}\,\varphi}$ are the mass and internal energy of a particle of type φ ($M_m = M_n \equiv M_1$). We shall assume that no quantum transitions m-n occur in the collisions. We can then eliminate the transition energy E_{mn} from $\varepsilon_{\text{int}\,m}$ in (2.3) and assume that $\varepsilon_{\text{int}\,m} = \varepsilon_{\text{int}\,n} = \varepsilon_{\text{int}\,1}$. Using (2.1)–(2.4) we obtain in the usual way[14] a set of hydrodynamic equations:

$$(\partial_t \rho + \partial_\mathbf{r} \mathbf{J})_\mu = 0, \tag{2.5}$$

$$\left[\partial_t J_\alpha + \sum_\beta \partial_\beta (u_\alpha u_\beta \rho + P_{\alpha\beta})\right]_\mu = F_{\mu\alpha}, \quad \alpha, \beta = x, y, z, \tag{2.6}$$

$$\sum_{\mu\alpha\beta} [\partial_t(\rho E) + \partial_\beta (J_\beta E + q_\beta + u_\alpha P_{\alpha\beta})]_\mu = 0, \quad E_\mu = \frac{u_\mu^2}{2} + \varepsilon_\mu, \tag{2.7}$$

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$$(\rho, \mathbf{J}, P_{\alpha\beta}, \varepsilon, \mathbf{q})_\mu = \left\{ M \int f\, d\mathbf{v} \left[1, \mathbf{v}, V_\alpha V_\beta, \frac{1}{\rho}\left(\frac{V^2}{2}+\frac{\varepsilon_{\text{int}}}{M}\right), \left(\frac{V^2}{2}+\frac{\varepsilon_{\text{int}}}{M}\right)\mathbf{V} \right] \right\}_\mu, \tag{2.8}$$

$$(\mathbf{u}, \mathbf{V})_\mu = (\mathbf{J}/\rho,\ \mathbf{v}-\mathbf{u})_\mu, \tag{2.9}$$

$$\mathbf{F}_{1,b} = \pm M_1 \int \mathbf{v}(S_{nb}+S_{mb})\, d\mathbf{v}. \tag{2.10}$$

The expressions (2.8) and (2.9) can be understood as the equality of column matrices: for example $p, \rho_\mu = M_\mu \int f_\mu d\mathbf{v}$ and so on. The quantities $(\rho, \mathbf{J}, P_{\alpha\beta}, \varepsilon, \mathbf{q})$ are respectively the mass density and mass flux density, the pressure tensor, the thermodynamic internal energy per unit mass, and heat flow of component μ; $(\mathbf{u}, \mathbf{V}, \mathbf{F})_\mu$ are the macroscopic and thermal velocities of the component μ and the "friction" force density acting on this component. Equations (2.5) and (2.6) are the equations of continuity and momentum balance of the components, and (2.7) is the energy balance equation for the gas mixture as a whole.

We use next the model of an ideal liquid:[14]

$$P_{\alpha\beta\mu} = P_\mu \delta_{\alpha\beta}, \quad \delta_{\alpha\beta} = \begin{cases} 1, & \alpha=\beta \\ 0, & \alpha\neq\beta \end{cases}, \quad \mathbf{q}_\mu = 0. \tag{2.11}$$

We shall also assume that the macroscopic quantities $(\rho, \mathbf{J}, P, \varepsilon)_\mu$ depart slightly from their equilibrium values $(\rho^0, 0, P^0, \varepsilon^0)$ and introduce into consideration their nonequilibrium parts:

$$(\rho', \mathbf{J}, P', \varepsilon')_\mu = (\rho, \mathbf{J}, P, \varepsilon)_\mu - (\rho^0, 0, P^0, \varepsilon^0)_\mu. \tag{2.12}$$

We transform the energy balance equation. Using (2.11), the relations that are characteristic for an ideal gas[15]:

$$(P, \partial_t \varepsilon)_\mu = (kT\rho/M,\ c_v \partial_t T/M)_\mu,$$

where k is Boltzmann's constant, T_μ and $c_{v\mu}$ are the temperature and heat capacity at constant volume for a single particle of the component μ and the equation of continuity (2.5), we can obtain a linearized variant of Eq. (2.7) in the following form:

$$\partial_t \sum_\mu \left[\frac{1}{M}(\rho^0 c_v T' - kT^0 \rho') \right]_\mu = 0, \quad T_\mu' = T_\mu - T^0, \tag{2.13}$$

where T_0 is the equilibrium temperature of the gas mixture. We neglect the difference in the temperatures of the absorbing and buffer components, i.e., we assume that

$$T_1' = T_b' \equiv T'. \tag{2.14}$$

Ths approximation is valid, since the time of exchange of energy between the components amounts to several times the free path time τ_{fr}, whereas the characteristic times of the hydrodynamic processes greatly exceed τ_{fr}.[3)] The linearized equations of state for the components of the mixture, with account of (2.14), can be written down in the following general form:

$$P_\mu' = \frac{k}{M_\mu}(T^0 \rho_\mu' + \rho_\mu^0 T'). \tag{2.15}$$

We eliminate the nonequilibrium part of the temperature from (2.15) with the help of (2.13) and (2.14). Then the equations of state for the absorption component and the gas mixture as a whole will have the form

$$P_1' = \frac{kT^0}{M_1}\left[\rho_1' + (\tilde{\gamma}-1)\frac{\rho_1^0}{\rho_b^0 + M_b\rho_1^0/M_1}\left(\rho_2' + \frac{\Delta M}{M_1}\rho_1'\right)\right],$$

$$\tilde{\gamma} = \sum_\mu \left(\frac{c_p \rho^0}{M}\right)_\mu \Big/ \sum_\mu \left(\frac{c_v \rho_0}{M}\right)_\mu, \tag{2.16}$$

$$\rho_2' = \rho_1' + \rho_b', \quad \Delta M = M_b - M_1,$$

$$P_2' = \frac{\tilde{\gamma} kT^0}{M_b}\left(\rho_2' + \frac{\Delta M}{M_1}\rho_1'\right), \quad P_2' = P_1' + P_b'. \tag{2.17}$$

Here $c_{p\mu} = c_{v\mu} + k$ is the heat capacity at constant pressure of component μ per particle.

For simplicity, we limit ourselves to the case in which the absorbing gas is a small admixture to the buffer:

$$\rho_1^0 \ll \rho_b^0, \quad \rho_1^0/M_1 \ll \rho_b^0/M_b. \tag{2.18}$$

The motion of the absorbing and buffer gases is due, as can be seen from (2.6), to their mutual repulsion. Therefore, at $\rho_1^0 \ll \rho_b^0$ the inequality $|\mathbf{u}_b| \ll |\mathbf{u}_1|$, $|\mathbf{u}_m|$ is satisfied ($\mathbf{u}_m$ is the macroscopic velocity of the excited component of the absorbing gas) and the following expression is valid for the force $\mathbf{F} \equiv F_1$ in linear approximation:

$$F = \tilde{\nu}\mathbf{j}_m - \nu_n \mathbf{J}_1, \quad \tilde{\nu} = \nu_n - \nu_m. \tag{2.19}$$

Here ν_j $(j = n, m)$ is the diffusion frequency of collisions of the absorbing particle in quantum state j with the particles of the buffer gas, and $\mathbf{j}_m$ is the mass flow density of the excited component of the absorbing gas.

Using Eqs. (2.5) and (2.6) in the linearized variant and taking account of (2.11) and (2.19), and also transforming (2.16) and (2.17) in correspondence with the conditions (2.18), we can obtain the following set of equations:

$$\partial_t \rho_{1,2}' + \partial_r J_{1,2} = 0, \quad \mathbf{J}_2 = \mathbf{J}_1 + \mathbf{J}_b, \tag{2.20}$$

$$(\partial_t + \nu_n)\mathbf{J}_1 + \partial_r P_1' = \tilde{\nu}\mathbf{j}_m, \quad \partial_t \mathbf{J}_2 + \partial_r P_2' = 0, \tag{2.21}$$

$$P_1' = \frac{v_0^2}{2}\rho_1', \quad P_2' = c_0^2\left(\rho_2' + \frac{\Delta M}{M_1}\rho_1'\right),$$

$$v_0^2 = \frac{2kT^0}{M_1}, \quad c_0^2 = \frac{\gamma kT^0}{M_b}, \quad \gamma = \frac{c_{pb}}{c_{vb}}. \tag{2.22}$$

As noted above, the hydrodynamic processes are slow in the time scale of free path times. In this connection, we can neglect the term $\partial_t \mathbf{J}_1$ in comparison with $\nu_n \mathbf{J}_1$. In this approximation, the following equations are obtained from (2.20)–(2.22):

$$(\partial_t - D\partial_r^2)\mathbf{J}_1 = \frac{\tilde{\nu}}{\nu_n}\partial_t \mathbf{j}_m, \quad D = \frac{v_0^2}{2\nu_n}, \tag{2.23}$$

$$(\partial_t^2 - c_0^2 \partial_r^2)\mathbf{J}_2 = \frac{\Delta M}{M_1} c_0^2 \partial_r^2 \mathbf{J}_1. \tag{2.24}$$

Equation (2.23) is the inhomogeneous diffusion equation (D is the diffusion coefficient), while (2.24) is the inhomogeneous wave equation (c_0 is the sound velocity). For solution of the set (2.23) and (2.24), we must calculate the quantity $\mathbf{J}_m$, to which the next section of the paper is devoted.

3. MOTION OF THE EXCITED COMPONENT

Let the energy interval between the states m and n of the absorbing particles be equal to $\hbar\omega$, and let the excited state decay to the ground state at the rate Γ_m. Then the behavior

of the excited component of the absorbing gas in the field of radiation with frequency ω, wave vector $\mathbf{k}$, and photon flux density I ($I = \tilde{I}/\hbar\omega$, where $\tilde{I}$ is the intensity of the radiation) is described by the following kinetic equation:

$$(\hat{A}+\Gamma_m) f_m = \sigma I (f_1 - 2f_m) + S_m,$$
$$\sigma = \frac{4\pi |\mathbf{d}|^2 \omega}{c\hbar} \frac{\Gamma}{\Gamma^2 + (\Omega - \mathbf{kv})^2}. \tag{3.1}$$

Here f_m is the distribution function of the excited particles, σ is the cross section for the transition $m - n$ with account of the Doppler shift $\mathbf{k}\cdot\mathbf{v}$ of the radiation frequency; $\mathbf{d}$ and Γ are the dipole moment and the halfwidth of the transitions $m - n$; $\Omega = \omega - \omega_{mn}$ is the resonance defect for the immobile particle; c is the velocity of light. Applying to (3.1) the operations $M_1 \int d\mathbf{v}$ and $m_1 \int \mathbf{v} d\mathbf{v}$, we can obtain the linearized equation of the hydrodynamics of the excited component:

$$(\partial_t + \Gamma_m)\rho_m + \partial_r \mathbf{j}_m = \sigma_0 \rho_1^0 I, \tag{3.2}$$
$$(\partial_t + \Gamma_m + \nu_m)\mathbf{j}_m + {}^1/_2 v_0^2 \partial_r \rho_m = (\mathbf{k}/|\mathbf{k}|) a v_0 \sigma_0 \rho_1^0 I, \tag{3.3}$$

where

$$\rho_m = M_1 \int f_m \, d\mathbf{v}, \quad \sigma_0 = \sigma(\mathbf{v}=0), \quad a = \frac{\Omega |\mathbf{k}| v_0}{\Gamma^2 + \Omega^2}.$$

In the derivation of (3.2) and (3.3), we have used the first of the equations of state (2.22) and assumed satisfaction of the conditions

$$\Gamma \text{ or } |\Omega| \gg |\mathbf{k}| v_0, \quad \rho_m \ll \rho_1^0, \tag{3.4}$$

which mean that the excitation of the particles by radiation is weak and weakly selective in the velocity. In the case in which the characteristic time scale τ of change of the photon flux density exceeds the relaxation time Γ_m^{-1}, i.e.,

$$\tau \gg \Gamma_m^{-1}, \tag{3.5}$$

we can neglect the terms $\partial_t \rho_m$ and $\partial_t \mathbf{j}_m$ in Eqs. (3.2) and (3.3). Omitting also the term $\partial_r \mathbf{j}_m$ in (3.2), we can obtain the following solution for $\mathbf{j}_m$:

$$\mathbf{j}_m = \frac{\mathbf{k}}{|\mathbf{k}|} a v_0 \frac{\Gamma_m}{\Gamma_m + \nu_m} \rho_m - D_m \partial_r \rho_m,$$
$$\rho_m = \sigma_0 I \rho_1^0 / \Gamma_m, \quad D_m = v_0^2 / 2(\Gamma_m + \nu_m). \tag{3.6}$$

Using (3.6), we can easily show that the neglect of the term $\partial_r \mathbf{j}_m$ in (3.2) is valid in the case

$$D_m / \Gamma_m l_{ph} \ll 1, \tag{3.7}$$

as is indeed assumed. In (3.7) l_{ph} is the scale of spatial inhomogeneity of the photon current density.

The two components in (3.6) reflect the existence of two mechanisms of generation of direct motion of the excited particles. The first of these (the component proportional to a) is associated with the selectivity of the optical excitation of particles by velocity. The quantity av_0 is the mean rate of excitation of the particle before a Maxwellizing collision, while the factor $\Gamma_m/(\Gamma_m + v_m)$ determines the fraction of the lifetime of the excited particle in the translationally nonequilibrium state $(\Gamma_m + \nu_m)^{-1}$ of the total decay time Γ_m^{-1} of the excited state. The second mechanism is connected with the diffusion of particles in state m, which takes place as a consequence of the spatial inhomogeneity of the intensity of the exciting radiation. The factor $v_0^2/2(\Gamma_m + \nu_m)$ is the diffusion coefficient of particles in the state m. Its difference from the ordinary diffusion coefficient $v_0^2/2\nu$ (ν is the collision frequency) is due to the fact that the decay of state m leads to additional "braking" of the flux of the excited particles.

Within the framework of the conditions (3.4), the interaction of light with atoms takes on a linear character and the process of photo-absorption obeys Bouguer's law. In particular, the photon flux density of the light wave traveling in the positive z direction is described by the following expression:

$$I(\mathbf{r}, t) = I_0(t - z/c) e^{-\lambda z},$$
$$I_0(t - z/c) = I(x, y, z=0, t - z/c), \quad \lambda = \sigma_0 \rho_1^0 / M_1. \tag{3.8}$$

Upon satisfaction of the condition (3.5), the quantity τc appreciably exceeds the length of the absorbing media usually used in experiments. Taking this into account, (3.8) can be rewritten in the form

$$I = I_0(t) e^{-\lambda z} \tag{3.9}$$

with accuracy up to terms proportional to the small factor $z/\tau c$.

4. SOUND GENERATION

We now analyze the problem of light-induced disequilibrium of the total pressure of the gas mixture in the case in which the light wave is a homogeneous pulse of length τ with a smooth time envelope. This can be accomplished on the basis of the solution of the set of equations (2.23), (2.24), using next the equations of continuity (2.20) and the second of the equations of state (2.22).

We turn to the system (2.23), (2.24), where the quantity $\mathbf{j}_m$ is determined by the expressions (3.6) and (3.9). We shall solve it by assuming that the mixture of gases is located in an acoustical resonator of length l (the coordinates of the ends of the resonator are $z = 0, l$). We shall also assume that the photon flux density is homogenous in the xy plane, i.e., $I_0(t)$ in (3.9) depends only on the time. Under these conditions, the problem becomes one-dimensional. We consider next the projections j_m and $J_{1,2}$ of the vectors $\mathbf{j}_m$ and $\mathbf{J}_{1,2}$ on the z axis. The formulation of the problem corresponds to the boundary and initial conditions

$$j_m, \; J_{1,2}|_{z=0,\,l} = 0, \quad J_{1,2}, \quad \partial_t J_2|_{t=-\infty} = 0, \tag{4.1}$$

which expresses the absence of particle fluxes through the ends of the resonator and corresponds to an equilibrium initial state of the gas mixture. The solution of Eq. (2.23) satisfying the conditions (4.1) has the form

$$J_1 = \sum_{s=1}^{\infty} J_{1s} \sin k_s z, \quad k_s = \frac{\pi s}{l},$$
$$J_{1s} = \frac{\tilde{\nu}}{\nu_n} v_0 \rho_1^0 \delta \left\{ i(t) - D k_s^2 \int_{-\infty}^{t} \exp[-Dk_s^2 (t - t')] i(t') \, dt' \right\} \chi_s, \tag{4.2}$$
$$\delta = \varkappa \left(\frac{a\Gamma_m}{\Gamma_m + \nu_m} + \frac{D_m \lambda}{v_0} \right), \quad \varkappa = \frac{\sigma_0 I_0(0)}{\Gamma_m}, \quad i(t) = \frac{I_0(t)}{I_0(0)},$$

$$\chi_s=\frac{2}{l}\frac{k_s}{\lambda^2+k_s^2}[1-(-1)^s e^{-\lambda l}].$$

The motion of the absorbing gas as a whole can be written qualitatively in the following fashion. The light-induced flux that arises because of the difference in the frequencies ν_m and ν_n leads to an inhomogeneous distribution of the absorbing gas in space (the gas is drawn towards one of the ends of the resonator). As a consequence, a diffusion flux is produced in the opposite direction. Two components in the expression for J_{1s} are connected with the presence of these two fluxes. Using (4.2), we find the following solution of the boundary-value problem (2.24), (4.1):

$$J_2=\sum_{s=1}^{\infty} J_{2s}\sin k_s z,$$

$$J_{2s}=\frac{\Delta M}{M_1}\frac{\bar{v}}{\nu_n}v_0\rho_1^0\delta\chi_s\int_{-\infty}^{t} i(t')\{\Delta_s\sin\Delta_s(t-t') \tag{4.3}$$

$$-Dk_s^2[\exp[-Dk_s^2(t-t')]-\cos\Delta_s(t-t')]\}\,dt',\qquad \Delta_s=c_0k_s.$$

Substituting (4.2) and (4.3) in (2.22) and finding $\rho'_{1,2}$, we obtain next with the help of (2.22) the following expression for P_2':

$$P_2'=\sum_{s=1}^{\infty}P_{2s}\cos k_s z,$$

$$P_{2s}=\xi P_1^0\chi_s\Delta_s\int_{-\infty}^{t} i(t')\left[\cos\Delta_s(t-t')-\frac{Dk_s^2}{\Delta_s}\sin\Delta_s(t-t')\right]dt', \tag{4.4}$$

$$\xi=\Delta M\left(\frac{2\gamma}{M_1M_b}\right)^{1/2}\frac{\bar{v}}{\nu_n}\delta.$$

In what follows, we shall be interested in the low-frequency modes of the resonator, i.e., we shall consider such values of s at which

$$Dk_s^2/\Delta_s\ll 1. \tag{4.5}$$

For times $t\gg\tau$, the upper limit of integration in the formula for P_{2s} can be replaced by ∞.[4] We then have in the approximation (4.5):

$$P_{2s}=\xi P_1^0\chi_s\Delta_s i_s\cos(\Delta_s t+\varphi_s);$$

$$i_s=(A_s^2+B_s^2)^{1/2},\quad \operatorname{ctg}\varphi_s=-\frac{A_s}{B_s},$$

$$\begin{Bmatrix}A_s\\B_s\end{Bmatrix}=\begin{Bmatrix}\mathrm{Re}\\\mathrm{Im}\end{Bmatrix}\int_{-\infty}^{\infty} i(t')\exp(i\Delta_s t')\,dt'. \tag{4.6}$$

The contributions $P_{2s}\cos k_s z$ to the nonequilibrium part of the total pressure describe at $t\ll\tau$ standing sound waves with frequencies Δ_s and wave numbers k_s.

We now analyze the problem of the spectral distribution of the acoustic signal, using (4.6). In the case $\lambda l\ll 1$ (weak light absorption), the factor χ_s in (4.6) reaches large values at odd s. It is maximal and of the order of unity for the fundamental mode (if $\lambda l\ll 1$) or for certain lower-frequency odd modes (if $\lambda l\sim 1$). In the limit $\lambda l\gg 1$ or for certain lower-frequency odd modes (if $\lambda l\sim 1$). In the limit $\lambda l\gg 1$ (strong light absorption) the quantity χ_s as a function of k, constitutes dispersion contour, with widths $2\sqrt{3\lambda}$ of the positive and negative branches (at half-height). Since $s>0$, we are interested in the positive branch, which reaches a maximum $(\lambda l)^{-1}\ll 1$ at $k_s\sim\lambda$. For all $s\sim\lambda/k_1$ the factor χ_s is also $\sim(\lambda l)^{-1}$. The quantity $\Delta_s l_s$, which together with χ_s determines the dependence of the amplitude of the waves on the number of the mode of the resonator, reaches maximum values of the order of unity at $\tau\sim 1/\Delta_s$.[5]

Thus, in the case of weak absorption of radiation, the optimal is pulse duration $\tau_{\mathrm{opt}}\sim\Delta_1^{-1}$. The most effectively excited state is the fundamental mode of the acoustical resonator ($\lambda l\ll 1$) or a small number of the lowest-frequency odd modes ($\lambda l\sim 1$). If strong light absorption takes place, then $\tau_{\mathrm{opt}}\sim(\lambda c_0)^{-1}$. Correspondingly, the frequency of the most efficiently generated mode and the width of the spectrum of the acoustic signal are of the order of λc_0.

In connection with the interpretation of Eq. (3.6), we have noted the existence of two mechanisms of the generation of a flow of excited particles: selective and diffusive. In the analysis of the problem of the acoustic signal, they come forward as two mechanisms of radiative-collisional generation of sound. For the development of a diffusion flux of excited particles, the necessary and sufficient condition is a nonzero gradient of the intensity of the induced radiation (spatially inhomogeneous radiation). Thus, on this basis, generation of sound is possible under conditions in which the selectivity of the excitation with respect to velocity is lacking, viz, a large departure from resonance, a large homogeneous transition width, and broad radiation spectrum. We note that the spatial inhomogeneity of the excitation of particles can be due not only to absorption of radiation but also to the transverse inhomogeneity of the light beam. In the latter case, cylindrical standing waves can also arise in the case of axial symmetry of the beam and of the acoustical resonator.

Free acoustical oscillations die out as a result of momentum and energy exchange between different sections of the gas (viscosity and thermal conductivity). The presence of walls leads to additional absorption because of the large gradients of the macroscopic velocity and of the temperature near them. For waves of small amplitude, an exponential damping law is valid.[17] Under this condition, the factor

$$\exp(-\beta_s t) \tag{4.7}$$

should be introduced on the right side of the relations (4.6) (β is the absorption coefficient of sound waves with frequency Δ_s). This takes into account the dissipation processes noted above. The contributions to the quantity β_s from these processes were calculated in Ref. 17. Here we shall give the expression for β_s without account of the effect of the resonator walls[17,18]:

$$\beta_s=\frac{\Delta_s^2}{2\rho_b^0 c_0^2}\left[\frac{4}{3}\eta+\eta'+\zeta\left(\frac{1}{c_{vb}}-\frac{1}{c_{pb}}\right)\right]. \tag{4.8}$$

In (4.8), η, η^1 and ζ are the coefficients of viscosity, second viscosity, and thermal conductivity of the buffer gas (for monatomic gases, $\eta'=0$).[19] Estimates with the use of elementary gas-kinetic formulas for η, η' and ζ show that $\beta_s\sim\Delta_s^2/\nu$, where ν is the collision frequency of particles with one another. This is why the low-frequency modes of the acoustical resonator are of interest.

5. DISCUSSION OF RESULTS. ESTIMATES

The calculations carried out in the present section can provide the theoretical basis of a method for determining the transport collision cross sections of excited atoms and molecules from the experimental data on the acoustical signal due to the LID effect in the field of a single light pulse.

We now estimate, by using Eq. (4.6), the orders of the quantities characterizing the acoustic signal and the electromagnetic radiation stimulating it for the case of atomic gases. We shall start out from the values of the parameters that are characteristic for atomic transitions: $\omega_{mn} \sim 10^{15}$ sec^{-1}, $|\mathbf{d}| \sim 1D$, $\Gamma_m \sim 10^7$ sec^{-1}, $|\mathbf{k}|v^0 \sim 10^{10}$ sec^{-1}. We carry out the estimates for the case $|\Omega| \sim 10^{11}$ sec^{-1}, $P_1^0 \sim 1$ Torr ($\rho_1^0/M_1 \sim 10^{16}$ cm^{-3}), $P_b^0 \sim 10$ Torr, $M_1 \sim M_b$, $l \sim 1$ cm. The values $\nu_{m,n} \sim 10^7$ sec^{-1}, $\Gamma \sim 10^9$ sec^{-1} correspond to a buffer gas pressure of 10 Torr. Under the considered conditions $\lambda l \sim 1$, i.e., there is a tendency for the effecient generation of the lowest frequency modes of the resonator and, in particular, for the basic mode with frequency $\Delta_1 \sim 10^5$ sec^1. Thus, the optimal duration of the light pulse amounts to $\tau \sim 10^{-5}$ sec. The photon flux density is bounded from above, by virtue of the second of the assumptions (3.4) [see also (3.6)], to values such that $\sigma_0 I_0(0)/\Gamma_m \sim 10^{-1}$. This corresponds to $\hbar\omega I_0(0) \sim 10^3$ W/cm^2. The estimate from (4.6), together with the use of the values of the quantities characterizing the gas mixture and the inducing radiation given above, shows that the amplitude of the oscillations of the pressure with frequency Δ_1 reaches values of the order of $|\tilde{\nu}/\nu_n| \cdot 10^{-2}$ Torr at the ends of the resonator. According to the data of Ref. 20 for the sensitivity of microphones, by recording a sound wave of such an amplitude one can determine the factor $|(\sigma_n - \sigma_m)/\sigma_n|$ ($\sigma_{m,n}$ is the transport collision cross section of the absorbing atom in quantum states m, n; $\nu_{m,n} \propto \sigma_{m,n}$) down to values of 10^{-5}. For the determination of the sign of the difference $\sigma_m - \sigma_m$ the phase of the wave must also be recorded.

The high sensitivity of sound receivers[20,21] allows us to choose experimental conditions by aiming not only at the maximum of the effect but also at the possibility of comparing the experimental results with the simplest theoretical formulas. Thus, at not too high pressures of the absorbing and buffer gases (such that $\lambda l \ll 1$, $\lambda v_0 \ll \Gamma_m |a|$, $\nu_m \ll \Gamma_m$ and pulse durations $\tau \ll \Delta_1^{-1}$ the expression (4.6) for the fundamental mode of the acoustic resonator takes the form

$$P_{21} = \frac{4}{\pi} \Delta M \left(\frac{2\gamma}{M_1 M_b} \right)^{1/2} \frac{\tilde{\nu}}{\nu_n} \frac{\sigma_0 \varepsilon_i \Delta_1}{\Gamma_m} a P_1^0 \cos \Delta_1 t. \qquad (5.1)$$

Here

$$\varepsilon_i = \int_{-\infty}^{\infty} I_0(t')\, dt'$$

is the number of photons landing on a unit area of the transverse cross section of the resonator. Thus, the amplitude of the sound wave is determined in this case only by the energy of the pulse and does not depend on the time envelope of the intensity of the radiation.

The results can find application also in connection with the interest arising recently in laser sources of sound.[22] The phenomenon of radiative-collision generation of sound in connection with the well-developed techniques of optical-acoustical spectroscopy of molecules[21] can apparently be used in the spectroscopy of atoms and for analysis of the composition of atomic gas mixtures.

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[1)] The separation of the components of the mixture $^{13}CH_3F + {}^{12}CH_3F$ in the field of a continuous CO_2 laser on the basis of the phenomenon of LID in gases was reported in Ref. 4.

[2)] The possibility of sound generation in gases by LID in a of continuous-radiation, field whose characteristics undergo modulation has been shown by Gel'mukhanov.[12]

[3)] We note that the hydrodynamic description that we have assumed is valid only in the case in which the characteristic time scale τ of change of the intensity of the light wave exceeds τ_{fr}, i.e., $\tau \gg \tau_{fr}$, as is indeed assumed.

[4)] It is assumed that the photon flux density reaches a maximum at the instant of time $t = 0$.

[5)] Similar optimal conditions are well known in mechanics in connection with the problem of the maximum energy transfer upon excitation of vibrations of a linear oscillator[16].

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