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LID sound generated by pulsed excitation in gases

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Abstract. A theory has been developed for radiative collisional generation of sound (RCGS) by single-pulse optical excitation. In a mixture of 'resonant' and buffer gases the light pulse is shown to give rise to the pressure pulse-acoustic signal due to light-induced drift (LID). The dynamics of spatial-temporal characteristics (amplitude, shape and duration) of the acoustic pulse and their relation to the dynamics of the signal spectrum have been investigated. The orders of the quantities characterising the acoustic signal and the electromagnetic radiation stimulating it have been estimated under typical experimental conditions. The calculation may serve as the theoretical basis of a method of investigating the collisional parameters of excited atomic particles by employing pulsed lasers and high-sensitivity optoacoustic spectroscopy techniques.

1. Introduction

In recent years much interest has been drawn to the phenomenon of light-induced drift (LID) of gases, which consists of light stimulation of counterflows of the components of gaseous mixtures (Gelmukhanov and Shalagin 1979, Antsygin *et al* 1979). Under non-stationary conditions the LID effect may result in a radiative collision generation of sound (RCGS) (Gelmukhanov 1983). The acoustic LID signal contains important information on the scattering cross sections of excited atomic particles obtainable by using the highly sensitive technique of optoacoustic spectroscopy (Pao 1977). Earlier (Shalaev and Yakhnin 1984) we showed the advantages of using pulsed lasers to generate LID sound when the pulse duration substantially exceeds the free-path time of the particles τ_f. However most lasers produce light pulses of duration ranging from 10^{-9} to 10^{-12} s (Q-switched and mode-locked lasers). For gas pressures of the order of (1-10) Torr, which is typical of LID experiments (see e.g. Antsygin *et al* 1979, Werij *et al* 1984), these pulses are short in the timescale τ_f.

The present paper develops the theory of RCGS for an arbitrary relation between the light-pulse duration and the free-path time of the particles. Unlike our earlier paper (Shalaev and Yakhnin 1984) studying spectral characteristics of LID sound, this paper concentrates on the analysis of its integral spatial-temporal characteristics. The generated acoustic signal is treated as a pulse of the pressure of the gaseous mixture with the duration, amplitude and shape varying with its propagation. Under certain conditions (see § 4) the acoustic pulse reproduces the form of the inducing light pulse.

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2. Basic equations

Let us consider the action of a light pulse of duration τ_p on a mixture of an absorbing and a buffer gas contained in an acoustic resonator of length l. For the energy spectrum of the absorbing particles we shall adopt the model of two non-degenerate states n and m (n is the ground state). The evolution of the medium is described by the set of kinetic equations

$$\hat{A}f_\mu = S_\mu \qquad \hat{A} = \partial_t + v\partial_r \qquad \mu = 1, \mathrm{B} \tag{2.1}$$

where f_μ and S_μ are the distribution function (dependent on the velocity v, the radius vector r and the time t) and the collision integral of the component μ; subscripts $\mu = 1$ and $\mu = \mathrm{B}$ label quantities referring to the absorbing gas and the buffer, respectively. In the process of interaction with the radiation, some of the absorbing particles go 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, representing S_1 in the form

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

The collision integrals S_φ ($\varphi = m, n, \mathrm{B}$) have the following structure:

$$S_\varphi = \sum_\psi S_{\varphi\psi} \qquad \psi = m, n, \mathrm{B} \tag{2.3}$$

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:

$$\int S_{\varphi\psi}\,\mathrm{d}v = 0 \qquad \int (Q_\varphi S_{\varphi\psi} + Q_\psi S_{\psi\varphi})\,\mathrm{d}v = 0$$
$$Q_\varphi = M_\varphi v,\ M_\varphi v^2/2 + \varepsilon_{\mathrm{in}\,\varphi}. \tag{2.4}$$

Here M_φ and $\varepsilon_{\mathrm{in}\,\varphi}$ are the mass and the internal energy of a particle of type φ ($M_m = M_n = M_1$; $\varepsilon_{\mathrm{in}\,m} = \varepsilon_{\mathrm{in}\,n} \equiv \varepsilon_{\mathrm{in}\,1}$). Using (2.1)-(2.4) we obtain in the usual way (Cherchiniani 1978) a set of balance equations for the mass, momentum and energy of the gaseous medium. We write down these equations in a linearised form, assuming that the medium state is close to equilibrium

$$\partial_t \rho_\mu + \partial_r J_\mu = 0 \tag{2.5a}$$
$$\partial_t J_\mu + \partial_r P_\mu = F_\mu \tag{2.5b}$$
$$\sum_\mu (\rho_\mu^0 \partial_t \varepsilon_\mu + P_\mu^0 \partial_r u_\mu) = 0. \tag{2.6}$$

Here

$$(\rho, J, P, \varepsilon)_\mu = \left\{ M \int f\,\mathrm{d}v \left[1, v, \frac{V^2}{3}, \frac{1}{\rho}\left(\frac{V^2}{2} + \frac{\varepsilon_{\mathrm{in}}}{M}\right)\right]\right\}_\mu \tag{2.7}$$
$$(u, V)_\mu = (J/\rho, v - u)_\mu \tag{2.8}$$
$$F_{1,\mathrm{B}} = \pm M_1 \int v(S_{n\mathrm{B}} + S_{m\mathrm{B}})\,\mathrm{d}v. \tag{2.9}$$

† No quantum transitions m–n are assumed to occur in the collisions. This also accounts for the fact that in the quantities $\varepsilon_{\mathrm{in}\,m,n}$ in (2.4) the part of energy related with 'the degree of freedom, interacting with the radiation' is neglected.

Expressions (2.7) and (2.8) should be understood as the equality of column matrices: for example, $\rho_\mu = M_\mu \int f_\mu \, d\boldsymbol{v}$ and so on. The quantities $(\rho, \boldsymbol{J}, P, \varepsilon)_\mu$ are, respectively, the mass density, the mass flux density, the pressure and the internal energy per unit mass of the component μ (ρ_μ^0 and P_μ^0 are the equilibrium values of the quantities ρ_μ and P_μ); $(\boldsymbol{u}, \boldsymbol{V}, \boldsymbol{F})_\mu$ are, respectively, the macroscopic and thermal velocities of the component μ and the density of the 'friction' force acting on this component from the other one. Equations (2.5) are the equations of continuity and momentum balance of the components; (2.6) is the energy balance equation for the gas mixture as a whole. Equations (2.5) and (2.6) were obtained with neglect of the non-diagonal elements of pressure tensors and heat flux vectors. In hydrodynamics these quantities are responsible for the dissipative processes to be taken into account below in discussing the problem of absorption of the acoustic signal.

Furthermore, we shall treat the mixture components as ideal gases and make use of the typical relations

$$(P, \partial_t \varepsilon)_\mu = (kT\rho/M, \, c_v \partial_t T/M)_\mu \tag{2.10}$$

where k is Boltzmann's constant; T and $c_{v\mu}$ are the temperature and the heat capacity at constant volume for a single particle of the component μ†. Then using (2.5), (2.6) and assuming the heat capacity of the absorbing and buffer gases to be identical ($c_{v1} = c_{vB} \equiv c_v$) the following equation for the total pressure of the gas mixture $P = P_1 + P_B$ can be derived:

$$\hat{W}P = -c^2 \frac{n_1^0 n_B^0}{n^0} \Delta M \partial_{rt}(\boldsymbol{u}_1 - \boldsymbol{u}_B). \tag{2.11a}$$

Here

$$\hat{W} = \partial_t^2 - c^2 \partial_r^2 \qquad c^2 = (c_p/c_v)(P^0/\rho^0) \qquad c_p = c_v + k$$
$$(P^0, \rho^0, n^0) = \sum_\mu (P^0, \rho^0, n^0)_\mu \qquad n_\mu^0 = \rho_\mu^0 / M_\mu \qquad \Delta M = M_B - M_1. \tag{2.11b}$$

Expression (2.11a) is an inhomogeneous wave equation (c is the sound velocity). A similar equation was obtained by Gel'mukhanov *et al* (1985) for the case when the absorbing and buffer gases are monatomic. In (2.11b) c_p is the heat capacity at constant pressure per particle; P^0, ρ^0 and n^0 are the equilibrium values of the total pressure, density and concentration of the gas mixture; n_μ is the equilibrium concentration of the component μ.

We shall limit ourselves to the case in which the absorbing gas is a small admixture to the buffer:

$$n_1^0 \ll n_B^0 \qquad \rho_1^0 \ll \rho_B^0. \tag{2.12}$$

Motion of the mixture components in the LID process is due to their mutual repulsion (force $\boldsymbol{F}_\mu$ in (2.5b)). Hence for $\rho_1^0 \ll \rho_B^0$ the buffer gas velocity is small and equation (2.11) under conditions (2.12) may be written down as

$$\hat{W}P = -c^2 \frac{\Delta M}{M_1} \partial_{rt} \boldsymbol{J}_1 \qquad c^2 = (c_p/c_v)(kT^0/M_B). \tag{2.13}$$

† The relation from the second term on the RHS of (2.10) is valid for slow development of non-equilibrium in the timescale of the energy exchange between the translational and internal degrees of freedom of the particles.

T^0 is the equilibrium temperature of the medium. The density of the absorbing gas flow, determining the right part of (2.13), can be found with the help of (2.5*b*) for $\mu = 1$. This equation contains the 'friction' force density acting upon the absorbing gas from the buffer. By definition (2.9), the force $\boldsymbol{F}_1$ consists of two parts: $\boldsymbol{F}_1 = \boldsymbol{F}_m + \boldsymbol{F}_n$ related with the scattering of excited and unexcited absorbing particles on the buffer-gas particles. In the linear approximation $\boldsymbol{F}_\alpha \sim \boldsymbol{u}_B - \boldsymbol{u}_\alpha$ ($\alpha = m, n$; $\boldsymbol{u}_\alpha$ is the macroscopic velocity of the α component of the absorbing gas). However under conditions (2.12) the buffer gas moves slowly so that

$$\boldsymbol{F}_1 = -\nu_n \boldsymbol{j}_n - \nu_m \boldsymbol{j}_m = \tilde{\nu} \boldsymbol{j}_m - \nu_n \boldsymbol{J}_1 \qquad \tilde{\nu} = \nu_n - \nu_m \tag{2.14}$$

where ν_α is the diffusion frequency of collisions of the absorbing particle in quantum state α with the buffer ones; $\boldsymbol{j}_\alpha$ is the mass flow density of the 'resonant' gas α component. A qualitative description of the absorbing gas evolution as a whole under conditions (2.12) may appear as follows. Light-induced drift of this component causes its inhomogeneous distribution over the space (the gas is drawn towards one of the ends of the resonator) which results in the diffusion flow counterdirected with respect to the initial one. If the light-pulse duration is essentially shorter than the time for establishing LID diffusion equilibrium (for such equilibrium $\boldsymbol{J}_1 = 0$), the term $\partial_t P_1$ in (2.5*b*), responsible for the diffusion of the absorbing gas, may be neglected. Then taking into account equation (2.14) we obtain, for $\boldsymbol{J}_1$, the expression

$$\boldsymbol{J}_1 = \tilde{\nu} \int_0^\infty \boldsymbol{j}_m(t-\tau)\, \mathrm{e}^{-\nu_n \tau}\, \mathrm{d}\tau. \tag{2.15}$$

This formula acquires a certain meaning if the value $\boldsymbol{j}_m$ is known. The next section is devoted to calculation of this value.

3. Motion of the excited component

Let the energy interval between the states m and n of the absorbing particles be equal to $\hbar\omega_{mn}$ and let the excited state decay to the ground state at the rate Γ_m. Then the behaviour of the excited component of the absorbing gas in the radiation field with frequency ω, wavevector $\boldsymbol{k}$ and photon flux density I ($I = \tilde{I}/\hbar\omega$, where $\tilde{I}$ is the radiation intensity) is described by the following kinetic equation:

$$(\hat{A} + \Gamma_m) f_m = \sigma I (f_1 - 2f_m) + S_m$$
$$\sigma = \frac{4\pi |\boldsymbol{d}|^2 \omega}{c_L \hbar} \frac{\Gamma}{\Gamma^2 + (\Omega - \boldsymbol{k}\boldsymbol{v})^2}. \tag{3.1}$$

Here f_m is the distribution function of the excited particles; σ is the cross section for the transition m–n taking into account the Doppler shift $\boldsymbol{k}\boldsymbol{v}$ of the radiation frequency; $\boldsymbol{d}$ and Γ are the dipole moment and the homogeneous halfwidth of the transition m–n; $\Omega = \omega - \omega_{mn}$ is the resonance defect for the immobile particle; c_L is the light velocity.

Under conditions (2.12) the kinetics of the excited component is determined primarily by the scattering of the absorbing particles on the buffer ones. In this connection, in (3.1) the substitution $S_m \to S_{mB}$ is valid. Our further assumption will be that the atoms are weakly excited by the radiation $f_m \ll f_1$. This, together with the assumption that the medium state is close to equlibrium, allows the following approximation on the right-hand side of (3.1):

$$f_1 - 2f_m \simeq f_1 \simeq \rho_1^0 W_1(v)/M_1. \tag{3.2}$$

Here

$$W_1(v) = (\pi^{1/2} v_0)^{-3} \exp[-(v/v_0)^2] \qquad (v_0^2 = 2kT^0/M_1)$$

is the Maxwellian velocity distribution for the absorbing gas. Now applying to (3.1) the operations $M_1 \int \dots \mathrm{d}v$ and $M_1 \int v \dots \mathrm{d}v$ we have the following linearised equations of mass and momentum balance of the excited component:

$$(\partial_t + \Gamma_m)\rho_m + \partial_r j_m = M_1 \lambda I \tag{3.3}$$

$$(\partial_t + \Gamma_m + \nu_m) j_m + \partial_r P_m = a v_0 M_1 \lambda I. \tag{3.4}$$

Here

$$(\rho, j, P)_m = M_1 \int f_m [1, v, \tfrac{1}{3}(v - j_m/\rho_m)^2] \, \mathrm{d}v$$

$$\lambda = \sigma_0 \rho_1^0 / M_1 \qquad \sigma_0 = \sigma|_{v=0} \qquad a = k v_0 \Omega / (\Omega^2 + \Gamma^2).$$

ρ_m and P_m are the density and the pressure of the excited component; λ is the coefficient of light absorption; parameter a (or rather its modulus) indicates the degree of selectivity of the optical excitation of the particles in their motion velocities. In deriving (3.3) and (3.4) we assumed that

$$\Gamma \text{ or } |\Omega| \gg |k| v_0. \tag{3.5}$$

In other words, the velocity selectivity of the excitation was assumed to be weak.

The quantity $\partial_r P_m$ in (3.4) may be linearised with respect to the density ρ_m and the temperature T_m of the excited component by using the equation of state in general form $P_m = k T_m \rho_m / M_1$. Since the equilibrium value ρ_m is $\rho_m = 0$, the linearisation yields $\partial_r P_m = (kT^0/M_1)\partial_r \rho_m$. On this account and omitting also the term $\partial_r j_m$ in (3.3), the following solution for (3.3), (3.4) can be obtained:

$$\rho_m = M_1 \lambda \int_0^\infty I(t-\tau) \, \mathrm{e}^{-\Gamma_m \tau} \, \mathrm{d}\tau \tag{3.6}$$

$$j_m = v_0 M_1 \lambda \int_0^\infty \left(a + \frac{v_0}{2\nu_m} (1 - \mathrm{e}^{-\nu_m \tau}) \partial_r \right) I(t-\tau) \, \mathrm{e}^{-(\Gamma_m + \nu_m)\tau} \, \mathrm{d}\tau \tag{3.7}$$

that satisfies the 'equilibrium' initial conditions $\rho_m(t = -\infty)$, $j_m(t = -\infty) = 0$. Now, using (3.6) and (3.7) it can easily be shown that the neglect of the term $\partial_r j_m$ is valid provided

$$v_0 l_p \Gamma_m^{-1} \leqslant 1 \tag{3.8}$$

(l_p is the scale of spatial inhomogeneity of the photon flux density) and the requirement $f_m \ll f_1$ now has a concrete meaning: the inequality $f_m \ll f_1$ within the framework of (3.5) is realised for

$$\sigma_0 I_0(t=0) \tau_p \ll 1 \qquad \text{for} \qquad \tau_p \equiv I_0^{-1}(t=0) \int_0^\infty I_0(t) \, \mathrm{d}t \ll \Gamma_m^{-1} \tag{3.9}$$

or

$$\kappa \equiv \sigma_0 I_0(t=0)/\Gamma_m \ll 1 \qquad \text{for} \qquad \tau_p \geqslant \Gamma_m^{-1} \tag{3.10}$$

where $I_0(t)$ is the photon flux density at the medium entrance; the time moment $t=0$ corresponds to the maximum of the value I_0; τ_p is the duration of the light pulse; κ is the maximum value of the saturation parameter for the quantum transition m–n.

The two terms in (3.7) reflect the existence of two mechanisms of generation of a directed motion of excited particles. The first of these (the component proportional to a) is related to the selectivity of the optical excitation of particles by velocity. The second mechanism is connected with the diffusion of particles in state m that results from the spatial inhomogeneity of the radiation intensity. The corresponding component is proportional to $\partial_r I$.

Substitution of (3.7) into (2.15) yields the following expression:

$$J_1 = \tilde{\nu} v_0 M_1 \lambda \int_0^\infty \left[\frac{e^{-\nu_1 \tau} - 1}{\nu_1} \frac{v_0}{2\nu_m} \partial_r - \frac{e^{-\nu_2 \tau} - 1}{\nu_2} \left(a + \frac{v_0 \partial_r}{2\nu_m} \right) \right] \times I(t-\tau)\, e^{-\nu_n \tau}\, d\tau \qquad \nu_1 = \Gamma_m - \nu_n \qquad \nu_2 = \nu_1 + \nu_m. \tag{3.11}$$

Expression (3.11) describes the motion of the absorbing gas as a whole: light-induced drift. According to the general regularities for this phenomenon, it develops with the difference in collisional characteristics of the absorbing particles in the ground and excited states ($\tilde{\nu} = \nu_n - \nu_m$).

In the case of weak optical excitation ($f_m \ll f_1$) the interaction of light with atoms has a linear character and the process of photoabsorption obeys Bouguer's law. In particular, the photon flux density of the light wave travelling in the positive z direction is described by the following expression:

$$I(r, t) = I_0(t - z/c_L)\, e^{-\lambda z}$$
$$I_0(t - z/c) = I(x, y, z = 0, t - z/c_L). \tag{3.12}$$

If $l/c_L \ll \nu_\alpha^{-1}$, the delay of the light pulse described by the term $-z/c_L$ in (3.12) will not affect the development of the LID process. in this case for the value I in (3.11) the expression

$$I(r, t) = I_0(t)\, e^{-\lambda z} \tag{3.13}$$

may be used.

4. Sound generation

To analyse the problem of light-induced non-equilibrium of the total pressure of the gas mixture we make use of (2.13) in which the value J_1 is given by (3.11) and (3.13). We recall that the gas mixture is assumed to be located in an acoustical resonator (the coordinates of the ends of the resonator are $z = 0$, l). We also assume that the field intensity of the light pulse propagating in the positive direction of the z axis is homogeneous in the xy plane, i.e. $I_0(t)$ in (3.13) depends only on the time. Under these conditions the problem becomes one dimensional and, in our further consideration, instead of the vector J_1 we shall deal with its projection J_1 on the z axis. The formulation of the problem corresponds to the boundary and initial conditions

$$J_1,\ \partial_z P'|_{z=0,l} = 0 \qquad \partial_t P'|_{t=-\infty} = 0 \qquad P' \equiv P - P^0 \tag{4.1}$$

which express the absence of particle fluxes through the ends of the resonator and correspond to an equilibrium initial state of the gas mixture. The Fourier method

yields the following solution for the boundary problem:

$$P' = \sum_{s=1}^{\infty} P_s \cos k_s z \qquad k_s = \pi s/l \tag{4.2}$$

$$P_s = \frac{\Delta M}{M} c\Delta_s \int_{-\infty}^{t} J_{1s}(t') \cos \Delta_s (t-t')\, dt' \qquad \Delta_s = k_s c \tag{4.3}$$

$$J_{1s} = \frac{2}{l}\int_0^l J_1 \sin k_s z\, dz. \tag{4.4}$$

The solution simplifies for

$$t \gg \tau_p, \nu_n^{-1}, \Gamma_m^{-1}$$

i.e. at the stage when the transient processes, resulting in LID sound, have been completed. In particular, substituting (3.11) and (3.12) into (4.3) and (4.4) for small s, such that

$$\Delta_s \ll \nu_n, \Gamma_m$$

for the given time interval, we find

$$P_s \cos k_s z = \tfrac{1}{2}\tilde{\gamma} P_1^0 \chi_s \operatorname{Re} \int_{-\infty}^{\infty} i(t')\{\exp[i\Delta_s(T^- - t')] + \exp[i\Delta_s(T^+ - t')]\}\, dt'. \tag{4.5a}$$

Here

$$\tilde{\gamma} = \frac{\tilde{\nu}}{\nu_n}\Delta M\left(\frac{2\gamma}{M_1 M_B}\right)^{1/2}\left(\frac{a\Gamma_m}{\Gamma_m+\nu_m} + \frac{D_m\lambda}{v_0}\right)\kappa \qquad D_m = \frac{v_0^2}{2(\Gamma_m+\nu_m)}$$

$$\chi_s = 2\pi\Delta_1 \frac{\Delta_s^2}{\Delta^2+\Delta_s^2}[1-(-1)^s e^{-\pi\Delta/\Delta_1}] \tag{4.5b}$$

$$\Delta = \lambda c \qquad i(t) = I_0(t)/I_0(0) \qquad T^{\mp} = t \mp z/c.$$

Parameter a is the projection of the a vector on the axis z; D_m is the diffusion coefficient for excited absorbing particles. Formulae (4.5a, b) describe the standing sound waves (Δ_s and k_s are the frequencies and the wavenumbers) corresponding to the low-frequency modes of the acoustical resonator.

Free acoustical oscillations die out due to the dissipative processes associated with the viscosity and thermal conductivity of the medium. In addition, sound absorption is caused by the walls near which there are large gradients of the macroscopic velocity and the temperature. For waves of small amplitude an exponential damping law is valid (Landau and Lifshitz 1954). In this connection, the factor

$$\exp(-\beta_s t) \tag{4.6}$$

(β_s is the absorption coefficient of the sound wave with frequency Δ_s), taking into account all the dissipative processes mentioned above, should be introduced on the right-hand side of relation (4.5). The contributions from these processes to β_s have been calculated by Landau and Lifshitz (1954). Here, as an illustration, we shall give the expression for β_s disregarding the influence of the resonator walls:

$$\beta_s = \Delta_s^2/\nu \qquad \nu = 2\rho_B^0 c^2\left[\tfrac{4}{3}\eta + \eta' + \zeta\left(\frac{1}{c_v} - \frac{1}{c_p}\right)\right] \tag{4.7}$$

where η, η' and ζ are the coefficients of viscosity, second viscosity and thermal conductivity of the buffer gas†. Estimates obtained with the use of elementary gas-kinetic formulae for η, η' and ζ (Lifshiftz and Pitaevsky 1979) show that the parameter ν is of the same order of magnitude as the collision frequency in a pure buffer gas. This accounts for the interest in the low-frequency modes of the acoustical resonator.

Along with the mode structure, the spectral integral time-spatial characteristics of LID sound are an important consideration. The case in which only several resonator modes are excited readily yields a numerical analysis. When a great number of modes is generated, further analytical investigation can be carried out on the basis of substituting the integral for the sum (4.2). Let us consider in detail the sum (4.2) taking account of the modifications related to the sound absorption‡. We illustrate the analysis by the case when the relaxation parameters ν_n, Γ_m and ν are the same order of magnitude: $\nu_n \sim \Gamma_m \sim \nu$. We denote them as ν_R. Next we note that (4.7) determining the sound absorption coefficients β_s is valid, as well as (4.5*a*, *b*), only for low-frequency modes of the resonator such that $\Delta_s \ll \nu_R$. Assuming however its approximate validity also for $\Delta_s \sim \nu_R$ we would come to the conclusion that only low-frequency modes are 'long-lived' and form the acoustical signal at times

$$t \gg \tau_p, \nu_R^{-1}. \tag{4.8}$$

Within this time interval the properties of the acoustic LID signal can be studied on the basis of calculating the following sum of the Fourier series:

$$P' = \tilde{\gamma} P_1^0 (\Sigma^+ + \Sigma^-)$$

$$\Sigma^{\pm} = \tfrac{1}{2} \sum_{s=1}^{\infty} \exp(-\Delta_s^2 t/\nu) \chi_s \operatorname{Re} \int_{-\infty}^{\infty} i(t') \exp[i\Delta_s (T^{\pm} - t')] \, dt'. \tag{4.9}$$

Within the framework of (4.8) the low-frequency terms in (4.9) describe the corresponding standing sound waves while the high-frequency terms are exponentially small. If we denote the sum (4.9) as

$$\sum_{s=1}^{\infty} F(\Delta_s)$$

the condition when this sum may be approximately substituted by the integral

$$(1/\Delta_s) \int_0^{\infty} F(\Delta_s) \, d\Delta_s$$

can be written in the form

$$\Delta_1 \left| \frac{d}{d\Delta_s} F(\Delta_s) \right| \ll |F(\Delta_s)| \tag{4.10}$$

in the region of variation of s giving the main contribution to the sum. In other words, the function $F(\Delta_s)$ should be slow in the scale of intermode frequency interval Δ_1. It can be shown that the requirement (4.10) is satisfied for the terms in the sum (4.9) provided

(1) the spectral width of the signal

$$\delta \equiv \min(\tau_p^{-1}, \delta_1) \gg \Delta_1 \tag{4.11}$$

† Since the 'resonant' gas, according to the assumption (2.12), is a small admixture to the buffer, the intensity of sound absorption is determined primarily by the properties of the buffer component.

‡ We shall be concerned with the case when the sound is absorbed mainly in the resonator volume and shall use the relation (4.7) for the coefficients β_s.

where $\delta_1 = (\nu/t)^{1/2}$ and

(2) $$\{\Delta_1 T^{\mp}/2\pi\} \ll 1. \tag{4.12}$$

Here the symbol { } denotes the fractional part of the number. Thus under conditions (4.11) and (4.12) from (4.9) it follows that

$$P' = \tilde{\gamma} P_1^0 (I^+ + I^-) \tag{4.13}$$

$$I^{\mp} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\Delta_s^2}{\Delta^2 + \Delta_s^2} \exp[-(\Delta_s/\delta_1)^2]\, d\Delta_s \int_{-\infty}^{x} i(t') \exp[i\Delta_s(T^{\mp} - t')]\, dt'.$$

Below, we give the expressions for the integrals $I^{\mp}$ corresponding to several specific cases:

I: $\Delta \ll \tau_p^{-1}, \delta_1$

(a) $\tau_p^{-1} \ll \delta_1$ $$I^{\mp} = i(T^{\mp}) \tag{4.14}$$

(b) $\tau_p^{-1} \gg \delta_1$ $$I^{\pm} = \frac{1}{2\pi^{1/2}} \tau_p \delta_1 \exp[-(T^{\mp}\delta_1/2)^2] \tag{4.15}$$

II: $\tau_p^{-1} \ll \Delta, \delta_1$ $$I^{\mp} = -(d^2/\Delta^2\, d^2 T^{\mp}) i(T^{\mp}) \tag{4.16}$$

III: $\delta_1 \ll \tau_p^{-1}, \Delta$ $$I^{\mp} = \frac{1}{4\pi^{1/2}} \frac{\tau_p \delta_1^3}{\Delta^2} \left(1 - \frac{T^{\mp 2}\delta_1^2}{2}\right) \exp[-(T^{\mp}\delta_1/2)^2]. \tag{4.17}$$

As is easily seen from (4.14)-(4.17), the functions $I^{\mp}$ describe the pressure pulse propagating in the $\pm$ direction of the z axis. In regimes I(a) and (II) the pulse profile is determined by the envelope of the inducing light pulse and is time independent. The amplitude of the signal also stays constant. For sufficiently small τ_p (regimes I(b) and III) the profile of the pressure pulse is described by the Gaussian curve (I(b)) or by its second derivative (III), independently of the shape of the light-pulse envelope. The pulse duration grows during the propagation and its amplitude decreases. This is accounted for by the specific spectral dynamics of the signal. According to (4.7), the high-frequency modes are absorbed more intensely than the low-frequency ones. This leads to the spectrum narrowing and, as a consequence, to the increased pressure pulse duration. The decreased amplitude is a trivial consequence of the sound absorption.

The analysis of (4.14)-(4.17) predicts the maximum absolute values of the integrals $I^{\mp}$ (such that $|I^{\pm}| \sim 1$) for the case when

$$\Delta \leqslant \tau_p^{-1} \leqslant \delta_1. \tag{4.18}$$

The factor $\tilde{\gamma} P_1^0$, determining, together with $I^{\mp}$, the amplitude of the acoustic signal, depends on the light absorption coefficient λ, and consequently on $\Delta (\Delta = \lambda c)$. The factor consists of two terms, one being linear and the other quadratic over $\lambda (\kappa P_1^0 \sim \lambda$ is taken into account). These terms correspond to the two mechanisms of RCGS: selective and diffusive (Shalaev and Yakhin 1984). For large λ (regimes II and III) $I^{\mp} \sim \lambda^{-2}$, and for small λ $I^{\mp}$ is independent of λ. Thus for the selective mechanism the optimum region of variation of the Δ, τ_p and δ_1 parameters is given by the relation

$$\Delta \sim \tau_p \sim \delta_1 \tag{4.19}$$

and for the diffusive mechanism

$$\tau_p^{-1} \sim \delta_1 \leqslant \Delta. \tag{4.20}$$

We note that the diffusive mechanism requires a non-zero gradient of the photon flux density only (light absorption or transverse inhomogeneity of the beam). Hence on this basis the generation of LID sound is still possible when the selective mechanism 'does not work': large resonance detuning, large homogeneous transition width or broad spectrum of the inducing radiation.

5. Discussion of results; estimates

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

We now estimate, using (4.13), the orders of quantities characterising the acoustic signal and the electromagnetic radiation inducing it for the case of atomic gases. We shall start out from the values of the parameters that are typical of atomic transitions: $\omega_{mn} \sim 10^{15}\,\mathrm{s}^{-1}$, $|d| \sim 1D$, $\Gamma_m \sim 10^7\,\mathrm{s}^{-1}$, $|k|v_0 \sim 10^{10}\,\mathrm{s}^{-1}$. We carry out the estimates for $|\Omega| \sim 10^{11}\,\mathrm{s}^{-1}$, $P_1^0 \sim 1$ Torr ($\rho_1^0/M_1 \sim 10^{16}\,\mathrm{cm}^{-3}$), $P_B^0 \sim 10$ Torr, $M_1 \sim M_B$, $l = 10$ cm. The values $\nu_{m,n} \sim 10^7\,\mathrm{s}^{-1}$ and $\Gamma \sim 10^9\,\mathrm{s}^{-1}$ correspond to a buffer-gas pressure of 10 Torr. Under the conditions discussed $\lambda \sim 1\,\mathrm{cm}^{-1}$, i.e. $\Delta = \lambda c \sim 10^5\,\mathrm{s}^{-1}$. Suppose a microphone (receiving the acoustic signal) is located at the exit (with respect to the laser beam) end of the resonator. Then for the pressure pulse in the microphone region the parameter $\delta_1 = (\nu/t)^{1/2}$ is of the order of $(\nu c/l) \sim 10^5\,\mathrm{s}^{-1}$ ($\nu \sim \nu_{m,n} \sim 10^7\,\mathrm{s}^{-1}$ since $M_1 \sim M_B$). The optimum light-pulse duration τ_{popt} amounts in this case, according to (4.19) and (4.20), to 10^{-5}: $\tau_{\mathrm{popt}} \sim 10^{-5}c$. For $\tau_p \gg \Gamma_m^{-1}$ the photon flux density is bounded from above in virtue of the assumption (3.9). For the chosen parameter values the light intensities

$$\tilde{I} = \hbar\omega I \sim 1\,\mathrm{kW\,cm}^{-2}\,(\omega \sim \omega_{mn})$$

correspond to $\kappa \sim 10^{-1}$.

Using (4.13) and the chosen values of the quantities characterising the gas mixture and the inducing radiation, the amplitude of the pressure pulse is estimated to be $|P'| \sim |\tilde{\nu}/\nu_n| 10^{-2}$ Torr.

According to the data from Walzer *et al* (1979) for the microphone sensitivity, by recording a sound wave of such an amplitude the factor $|(\sigma_n - \sigma_m)/\sigma_n|$ (σ_α is the transport cross section of the collisions between the absorbing atom in the quantum state α and the buffer-gas particles; $\nu_\alpha \sim \sigma_\alpha$) can be determined up to the values $|(\sigma_n - \sigma_m)/\sigma_n| \sim 10^{-5}$.

The estimates have been carried out for the case of weak and weakly velocity-selective optical excitation of particles (relations (3.9) and (3.5), respectively) and that is why the factor $\tilde{\gamma}$ in (4.13) is small: in our case $\tilde{\gamma} \sim 10^{-2}$. The simplifying assumptions (3.5) and (3.9) are of no principal importance and moreover they are not optimum for the manifestation of LID effect (Popov *et al* 1981), in particular for the generation of LID sound. As seen from formulae (4.13) and (4.5*b*), for a stronger (up to $\kappa \sim 1$ at $\tau_p \geqslant \Gamma_m^{-1}$, or $\sigma_0 I_0(0)\tau_p \sim 1$ at $\tau_p \ll \Gamma_m^{-1}$) and more selective ($|\alpha| \to 1$) optical excitation the situation is possible when $|P'| \sim |\tilde{\nu}/\nu_n| P_1^0$.

The widely used lasers generating light pulses in nano- and picosecond ranges of duration show the promise of radiative collision sound generation. Let, for example, the duration of the light pulse be 1 ns and the quantities characterising the gas mixture

have the same values as above. In this case $\tau_p^{-1} \gg \delta_1 \sim \Delta$ and the estimates can be obtained from (4.15) or (4.17) assuming that $\delta_1 \sim \Delta$. Since $\tau_p \ll \Gamma_m^{-1}$, the population of the excited state of the 'resonant' particles, according to (3.6), is determined by the parameter $\tau_p I_0(0)\sigma_0$. Assuming that $\tau_p I_0(0)\sigma_0 \sim 10^{-1}$, we have $I \sim 10^2$ kW cm^{-2}; $|P'| \sim |\tilde{\nu}/\nu_n| 10^{-4}$ Torr. The quantity $\tilde{\gamma}$ is small since the factor δ_1/Γ_m is also small: $\delta_1/\Gamma_m \sim 10^{-2}$ (but not $\tau_p\delta_1 \sim 10^{-4}$). Further shortening of the duration and the constant energy of the light pulse does not affect the properties of the pressure pulse. The radiation intensity, increased by an order of magnitude, would result in a proportional enhancement of the acoustic signal.

The results obtained can also find application in connection with the recent rise in the interest in laser sources of sound. The phenomenon of RCGS, combined with the developed technique of optoacoustic spectroscopy of molecules, can apparently be used in the spectroscopy of atoms and for analysis of the composition of atomic gas mixtures.

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