THE CONDITIONS OF STATIONARITY OF THE ABSORPTION COEFFICIENT UNDER OPTICAL PULSE PROPAGATION THROUGH A DOUBLE–LEVEL MEDIUM

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Propagation of a Gaussian pulse through a double–level medium is considered. Based on the system of Bloch–Maxwell equations the approaches that result in Bouguer's law with the absorption coefficient independent of time are analyzed.

In the most methods of remote laser sounding of the atmosphere the propagation of optical pulse from a laser source to a distant atmospheric volume is analyzed at the first stage and then the propagation of radiation reemitted by the volume towards the receiving aperture at the second. In accordance with this, for calculations of the energy losses of sounding and reemitted pulses the square (for unshifted frequencies) or the product (for shifted frequencies) of the atmospheric transmission defined by empirical Bouguer's law are involved in equation of sounding. This is practically always valid for transmission of reemitted pulse because of its small power. As to the sounding pulse propagation, disagreements with Bouguer's law are possible when the pulse frequency occurs within a molecular absorption line. The best–known example of such a disagreement is the absorption saturation effect.[1] In order to determine conditions of the Bouguer's law applicability let us use the Maxwell–Bloch equations describing within the framework of a semiclassical approach the interaction of sounding pulse with a two–level system modeling the molecular absorption.

Let the subscript b indicates the low energy level and the subscript a does the upper one, $\omega_0 = (\mathrm{E}_a - \mathrm{E}_b)/\hbar$ is the transition frequency. Dynamics of a two–level system interacting with the electric field E can be described by the system of the Bloch equations for the density matrix elements[2]

$$\dot{\rho}_{aa} = \frac{n_a - \rho_{aa}}{T_1} + i\frac{\mu_{ab}}{\hbar}E(z',\, t)(\rho_{ba} - \rho_{ab})\;; \tag{1}$$

$$\dot{\rho}_{bb} = \frac{n_b - \rho_{bb}}{T_1} + i\frac{\mu_{ab}}{\hbar}E(z',\, t)(\rho_{ab} - \rho_{ba})\;; \tag{2}$$

$$\dot{\rho}_{ab} = (-\,i\omega_0 - 1/T_2)\rho_{ab} + i\frac{\mu_{ab}}{\hbar}E(z',\, t)(\rho_{bb} - \rho_{aa})\;, \tag{3}$$

$$\rho_{ba} = \rho^*_{ab}\;.$$

Here, $\rho_{aa}(z',\, t)$ and $\rho_{bb}(z',\, t)$ are the populations of the a and b levels at the point z' for molecules having the velocity component ν_z, μ_{ab} is the matrix element of the electric dipole moment, T_1 and T_2 are the longitudinal and transverse relaxation times, n_a and n_b describe the equilibrium values of the populations and are given by

$$n_a = \frac{N_a\,\mathrm{e}^{-\nu_z^2/\bar{u}^2}}{\bar{u}\sqrt{\pi}}\;, \tag{4}$$

where N_a is the total number density of molecules at the level a, $\bar{u}$ is the average velocity of a molecule.

The field $E(z,\, t)$ is calculated from the Maxwell equation

$$-\frac{\partial^2 E}{\partial z^2} + \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2}\;. \tag{5}$$

The medium polarizability P is given by

$$P(z,\, t) = \int\limits_{-\infty}^{+\infty} \bar{\mu}(z,\, \nu_z,\, t)\,\mathrm{d}\nu_z\;; \tag{6}$$

$$\bar{\mu}(z,\, \nu_z,\, t) = \mu_{ab}\left[\rho_{ba}(z,\, \nu_z,\, t) + \rho_{ab}(z,\, \nu_z,\, t)\right]\;. \tag{7}$$

It should be noted that the field $E(z',\, t)$ in Eqs. (1)–(3), in accordance with Ref. 2, is defined in a coordinate system connected with a molecule, while Eq. (5) defines the field in a laboratory system of coordinates. Connection between the fields in these two systems is performed with the nonrelativistic ($\nu_z/c \ll 1$) transformation

$$z' = z - \nu_z\, t\;;$$

$$\Omega' = \Omega - k\,\nu_z\;; \tag{8}$$

$$k' = k\;,\;\; t' = t\;.$$

In practice, the calculations should be done as follows. First, one sets some approach to the field $E(z',\, t)$ and then the corresponding approach, for example, for $\rho_{ab}(z',\, \Omega',\, t)$ is found from the system of equations (1)–(3). After that transformation (8) is carried out, the result is substituted into Eqs. (5)–(7), and the subsequent iteration of $E(z,\, t)$ is found.

As a matter of fact, this procedure is very simple because transformation (8) keeps complete phase of the field, so $\Omega' t - kz' = \Omega t - kz$.

Let us represent $E(z',\, t)$, $\rho_{ab}(z',\, t)$, and $P(z,\, t)$ in the forms

$$E(z',\, t) = \frac{1}{2}\,E_0(z',\, t)\,\exp[i(\Omega' t - kz')] +$$

$$+\,\frac{1}{2}\,E^*_0(z',\, t)\,\exp[-\,i(\Omega' t - kz')]\;; \tag{9}$$

$$\rho_{ab}(z',\, t) = \tilde{\mathrm{r}}_{ab}(z',\, t)\,\exp[-\,i(\Omega' t - kz')]\;; \tag{10}$$

$$P(z,t) = \frac{1}{2}P_0(z,t)\exp[i(\Omega t - kz')] + \frac{1}{2}P^*_0(z,t)\exp[-i(\Omega t - z)]; \tag{11}$$

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$$P_0(z,\ t) = 2\mu_{ab} \int\limits_{-\infty}^{+\infty} \tilde{\rho}_{ba}(z' \to z - v_z t,\ \Omega' \to \Omega - k v_z,\ t) dv_z. \qquad (12)$$

For the envelopes $E_0(z',\ t)$ and $_{ab}(z',\ t)$ by substituting Eqs. (9) and (10) into Eqs. (1)–(3) and using the rotational wave approximation one obtains the following equations:

$$\dot{\tilde{\rho}}_{ba} = (-i\Delta - 1/T_2)\, \tilde{\rho}_{ba} - i \frac{\mu_{ab}}{2\hbar} E_0\, n\ ; \qquad (13)$$

$$\dot{n} = \frac{n_0 - n}{T_1} + i \frac{\mu_{ab}}{\hbar} \left(E_0 \tilde{\rho}_{ab} - E_0^* \tilde{\rho}_{ba}\right)\ ; \qquad (14)$$

$$\Delta = \Omega' - \omega_0\ ;\quad n = \rho_{bb} - \rho_{aa}\ ;\quad n_0 = n_b - n_a\ .$$

The shortened Maxwell equations for the envelopes can be written in the form

$$\frac{\partial E_0}{\partial z} + \frac{1}{c} \frac{\partial E_0}{\partial t} = 2\pi\, i\, k\, P_0\ , \qquad (15)$$

where P_0 is defined by formula (12).

If the real and imaginary parts of E_0 and P_0 are separated out

$$E_0 = \mathrm{E}^0 + \mathrm{E}_c + i\, \mathrm{E}_s\ , \qquad (16)$$

$$P_0 = P_c + i\, \mathrm{P}_s \qquad (17)$$

it can be shown that Eqs. (15) are valid if the following unequalities:

$$\left|\frac{\partial^2 \mathrm{E}_c}{\partial z^2}\right| \ll 2\, k \left|\frac{\partial \mathrm{E}_s}{\partial z}\right|\ ;\quad \left|\frac{\partial^2 P_c}{\partial t^2}\right| \ll 2\Omega \left|\frac{\partial P_s}{\partial t}\right|\ ;$$

$$\left|\frac{\partial^2 \mathrm{E}_s}{\partial t^2}\right| \ll 2\Omega \left|\frac{\partial \mathrm{E}_c}{\partial t}\right|\ ;\quad \left|\frac{\partial P_s}{\partial t}\right| \ll \frac{\Omega}{2} \left|P_c\right| \qquad (18)$$

hold.

The same unequalities should be added to the foregoing but with the substitutions

$$P_c \leftrightarrow P_s\ ,\quad \mathrm{E}_c \leftrightarrow \mathrm{E}_s\ .$$

Below we shall consider propagation of a Gaussian optical pulse with the envelope

$$\mathrm{E}^0(z,\ t) = \Pi \exp\left[-\frac{4(t - z/c)^2}{\tau_\mathrm{p}^2}\right]$$

through a medium occupying the space to the positive direction of the z axis.

The initial and boundary conditions are

$$E(z,\ -\infty) = 0\ ;$$

$$E(0,\ t) = \Pi \exp\left(-\frac{4\, t^2}{\tau_\mathrm{p}^2}\right) \cos\Omega\, t\ .$$

Here τ_p is the pulse duration measured at the level e^{-1}.

Let us introduce the dimensionless variables

$$\tau = \frac{t - z/c}{\tau_\mathrm{p}}\ ;\quad x = z/c\tau_\mathrm{p}$$

and the dimensionless functions

$$N = n/n_0\ ; \qquad R = \tilde{\rho}_{ba}/n_0\ ; \qquad Z = E_0/\Pi\ . \qquad (19)$$

In this set of the variables the system of equations (13)–(15) takes the form

$$\frac{\partial R}{\partial t} + (i\lambda_1 + \lambda_2)\, R = -i \frac{\lambda_3}{2} Z N\ ;$$

$$\frac{\partial N}{\partial t} + \lambda_4 (N - 1) = i\lambda_3 (Z R^* - Z^* R)\ ;$$

$$\frac{\partial Z}{\partial x} = -i\lambda_5 \int\limits_{-\infty}^{+\infty} \exp\left(-v_z^2/\overline{u}^2\right) R\, dv_z \qquad (20)$$

with the dimensionless parameters

$$\lambda_1 = \Delta\tau_\mathrm{p}\ ,\quad \lambda_2 = \tau_\mathrm{p}/T_2\ ,\quad \lambda_3 = \Omega_R\, \tau_\mathrm{p}\ ,$$

$$\lambda_4 = \tau_\mathrm{p}/T_1\ ,\quad \lambda_5 = \frac{4\pi\, k\, c\, \tau_\mathrm{p}\, \mu_{ab} (N_b - N_a)}{\overline{u}\sqrt{\pi}\, \mathrm{P}} \qquad (21)$$

and under the initial and boundary conditions

$$\tau = -\infty:\ N_0 = 1\ ,\ R_0 = 0\ ,\ Z_0 = 0\ ,\ x = 0:\ Z_0 = \exp(-4\tau^2)\ .$$

In Eq. (21) the value Ω_R is the Rabi frequency defined as

$$\Omega_R = \frac{\mu_{ab}\, \mathrm{P}}{\hbar}\ .$$

As follows from Eq. (20), at small values of λ_3 the optical pulse shows only negligible influence on a system of two–level molecules. Let us make the assumption that

$$\lambda_3 \ll 1 \qquad (22)$$

and write the sought–for solution in the form

$$R = \sum_{m=0}^{\infty} \lambda_3^m R_m\ ;\quad N = \sum_{n=0}^{\infty} \lambda_3^n N_n\ ;\quad Z = \sum_{k=0}^{\infty} \lambda_3^k Z_k\ . \qquad (23)$$

Condition (22) by virtue of Eqs. (21) is reduced to the limitation on the pulse duration

$$\tau_\mathrm{p} \ll \Omega_R^{-1} = \hbar/\ \mu_{ab}\, \Pi. \qquad (24)$$

Substitution of Eqs. (23) into the system of equations (20) gives the solutions in the first approach with respect to λ_3

$$R = \lambda_3 R_1\ ,\quad N = 1\ ,\quad Z = Z_0 + \lambda_3 Z_1\ ;$$

$$R_1 = -\frac{i}{2} \exp[-(i\lambda_1 + \lambda_2)\tau] \int\limits_{-\infty}^{\infty} \exp[-4y^2 + (i\lambda_1 + \lambda_2)\, y]\, dy\ ;$$

$$Z_1 = -\frac{\sqrt{\pi}}{2}\, \overline{u}\, \lambda_5 Z_0 \times$$

$$\times \int\limits_0^{\infty} \exp\left[-\left(4 + \frac{k^2 u^2 \tau_\mathrm{p}^2}{4}\right) y^2 - (\lambda_2 - 8\tau + i\Delta_1\tau_\mathrm{p}) y\right] dyx\ ;$$

$$\Delta_1 = \Omega - \omega_0\ . \qquad (25)$$

From Eqs. (19) and (26) by passing from the dimensionless coordinate x to z one obtains,

$$E_0(z,\ \tau) = \Pi \exp(-4\tau^2)(1 - Kz)\ ; \quad (26)$$

$$K = \frac{\sqrt{\pi}\,u}{2c\tau_p}\lambda_3\lambda_5 \int\limits_0^\infty \exp\left[-\left(4 + \frac{k^2\overline{u}^2\tau_p^2}{4}\right)y^2 - (\lambda_2 - 8\tau + i\Delta_1\tau_p)\,y\right]\mathrm{d}y = K_1 - iK_2$$

for the slow amplitude of the field $E_0(z,\ \tau)$.
From Eqs. (16) and (26) it follows that

$$E_c = -\Pi\exp(-4\tau^2)K_1 z\ ; \quad E_s = \Pi\exp(-4\tau^2)K_2 z\ ; \quad (27)$$

$$K_1 = \frac{\sqrt{\pi}\,u}{2c\tau_p}\lambda_3\lambda_5 \times \int\limits_0^\infty \exp\left[-\left(4 + \frac{k^2\overline{u}^2\tau_p^2}{4}\right)y^2 - (\lambda_2 - 8\tau)\,y\right]\cos\Delta_1\tau_p\,y\mathrm{d}y\ ;$$

$$K_2 = \frac{\sqrt{\pi}\,u}{2c\tau_p}\lambda_3\lambda_5 \times \int\limits_0^\infty \exp\left[-\left(4 + \frac{k^2\overline{u}^2\tau_p^2}{4}\right)y^2 - (\lambda_2 - 8\tau)\,y\right]\sin\Delta_1\tau_p\,y\mathrm{d}y\ .$$

Let us calculate the optical pulse intensity, using the relation

$$J = \frac{c}{4\pi}\frac{1}{2T}\int\limits_{-T}^{+T}|E|^2\mathrm{d}t = \frac{c}{8\pi}\left[(\mathrm{E}^0 + \mathrm{E}_c)^2 + \mathrm{E}_s^2\right]. \quad (28)$$

By substituting relations (27) into relation (28) and taking only linear, with respect to J_0, terms one obtains

$$J = J_0(1 - 2K_1 z)\ , \quad (29)$$

where $J_0 = \frac{c\Pi^2}{8\pi}\exp(-8\tau^2)$ is the intensity of a Gaussian pulse incident on a medium.

For the case of a weak absorption considered here Bouguer's law takes the form

$$J = J_0\exp(-\chi z) \simeq (1 - \chi z)\ . \quad (30)$$

From comparison of Eqs. (29) and (30) for the absorption coefficient, one obtains

$$\chi = \frac{\sqrt{\pi}\,u}{c\tau_p}\lambda_3\lambda_5 \times \int\limits_0^\infty \exp\left[-\left(4 + \frac{k^2\overline{u}^2\tau_p^2}{4}\right)y^2 - (\lambda_2 - 8\tau)\,y\right]\cos\Delta_1\tau_p\,y\mathrm{d}y\ . \quad (31)$$

By changing the variables $\left(4 + \frac{k^2\overline{u}^2\tau_p^2}{4}\right)y = \xi$ and substituting the explicit relations for λ_3 and λ_5 from Eq. (21) one finds that

$$\chi = \frac{8\pi^2\Omega\mu_{ab}^2(N_b - N_a)\tau_p}{hc\sqrt{4 + \frac{k^2\overline{u}^2\tau_p^2}{4}}} \times \int\limits_0^\infty \exp\left(-\xi^2 - \frac{\lambda_2 - 8\tau}{\sqrt{4 + \frac{k^2\overline{u}^2\tau_p^2}{4}}}\,\xi\right)\cos\frac{\Delta_1\tau_p\xi}{\sqrt{4 + \frac{k^2\overline{u}^2\tau_p^2}{4}}}\,\mathrm{d}\xi. \quad (32)$$

Let us now take into account the fact that in derivation of the Bloch equations (1)–(3) (see Ref. 2) it was assumed that the field $E(z,\ t)$ is linearly polarized along the x axis. That means that $E(z,\ t) = E_x(z,\ t)$ and $\mu = \mu_x$. We can write for the matrix element of the dipole moment that

$$\mu_{ab} = (\mu_x)_{ab} = e\cdot\boldsymbol{\mu}_{ab} = |\mu_{ab}|\cos\theta\ ,$$

where e is the unit vector along the x axis, $\boldsymbol{\mu}$ is the vector of a dipole moment. Since molecules in a gas have random orientations, the value $(\mu_x)_{ab}^2$ in Eq. (32) should be averaged over the angle θ that gives[1]

$$(\mu_x)_{ab}^2 = \frac{1}{3}|\mu_{ab}|^2\ .$$

The structure of the absorption coefficient in the form of Eq. (32) allows one to draw a conclusion about nonstationarity of the absorption process for any limited pulse durations. The limit of Eq. (32) as $\tau_p \to \infty$ gives

$$\chi_\infty = \frac{8\pi^2\Omega|\mu_{ab}|^2(N_b - N_a)}{3hc}\frac{2}{k\overline{u}}\int\limits_0^\infty \exp\left(-\xi^2 - \frac{2T_2^{-1}}{k\overline{u}}\,\xi\right) \times \cos\frac{2(\Omega - \omega_0)}{k\overline{u}}\,\xi\mathrm{d}\xi\ . \quad (33)$$

This relation corresponds to the Voight profile.[3] In a limiting case of $T_2^{-1}/k\overline{u} \gg 1$ from Eq. (33) one obtains the dispersion contour

$$\chi_L = \frac{S}{\pi}\frac{\gamma_L}{(\Omega - \omega_0)^2 + \gamma_L^2} \quad (34)$$

with the line intensity

$$S = \frac{8\pi^3}{3hc}\omega_0|\mu_{ab}|^2(N_b - N_a) \quad (35)$$

and halfwidth

$$\gamma_L = \frac{1}{2\pi T_2}\ . \quad (36)$$

In so doing in Eq. (35) the substitution $\Omega \to \omega_0$ was made, permitting to refer S to as the fundamental characteristic of any transition. Really, this does not distort the frequency dependence due to narrow spectral lines.

The other limiting case, when $T_2^{-1}/ku \ll 1$ in Eq. (33), corresponds to the Doppler line shape

$$\chi_D = \frac{2S}{\sqrt{\pi}\gamma_D} \exp\left[- \frac{(\Omega - \omega_0)^2}{\gamma_D^2} \right], \qquad (37)$$

where S is defined by formula (35),

$$\gamma_D = k\overline{u} = \frac{\Omega}{c}\,\overline{u} \simeq \frac{\omega_0}{c}\,\overline{u}\ ,\quad \overline{u} = 2\left(\frac{2\ln 2\, k_B T}{M}\right)^{1/2},$$

k_B is the Boltzmann constant and M is the molecular mass, so

$$\gamma_D = \omega_0 \left(\frac{2\ln 2\, k_B T}{M}\right)^{1/2} . \qquad (38)$$

The condition $\tau_p \to \infty$ is too indefinite for estimates. As follows from Eq. (32) the absorption can be considered stationary in the following cases:

(a) $k\overline{u}\tau_p \ll 1$; $\tau_p/T_2 \gg 1$.

This case corresponds to a negligibly small Doppler width $\gamma_D = ku$ provided that $\tau_p \gg T_2$ and

(b) $k\overline{u}\tau_p \gg 1$.

In the case of $\tau_p \gg \dfrac{1}{k\overline{u}}$ and the line shape defined by the relation $T_2^{-1}/k\overline{u}$.

In the case of nonstationary absorption we have

$\tau_p \ll \dfrac{1}{k\overline{u}}$ and $\tau_p \ll \dfrac{1}{T_2}$.

In the latter case of extremely short pulse durations one should verify fulfilment of conditions (18) by substituting there, for example, relations (27).

In conclusion, let us present some estimates for a concrete laser system.[4] Radiation of a ruby laser at the wavelength 634.383 nm coincides with the center of a water vapor absorption line with $\mu_{ab} = 3.8\cdot10^{-22}$ CGSE. The pulse of 1 J energy and duration $2\cdot10^{-8}$ s collimated by a telescope to the value of divergence $2\cdot10^{-4}$ rad provides the value of $\Omega_R \sim 6\cdot10^6\ \text{s}^{-1}$ in a volume at 1 km from the lidar. Therefore, $\lambda_3 = 0.12$, and the process of absorption can be considered linear. For tropospheric conditions H_2O line at 694.383 nm has the Doppler width $k\overline{u} \sim 1.2\cdot10^9\ \text{s}^{-1}$ and $k\overline{u}\ \tau_\pi = 24 \gg 1$. The transverse relaxation time T_2 can be defined from the linewidth ($\sim 0.1\ \text{cm}^{-1}$) and is equal to $5\cdot10^{-11}$ s. Then, $T_2^{-1}/k\overline{u} = 17 \gg 1$ and the condition of stationarity of the propagation process corresponding to the above–mentioned point b is fulfilled.

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