INTERACTION BETWEEN A PREPOLARIZED BEAM AND AN ELECTROMAGNETIC FIELD

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We consider the stationary interaction between a prepolarized molecular beam and a high frequency field, with account taken of relaxation. An expression is obtained for the polarization of such a beam, and it is shown that it contains convective, proper, and mixed polarization components that differ from one another in phase. Account is taken of the reaction exerted on the beam by its own field in a closed cavity by neglecting relaxation relative to the small parameter. The singularities of such an interaction are described in a linear approximation, and the role of each of the polarization components is explained. The results can serve as a physical basis for a rigorous nonlinear description of the experimentally observed effects.

A detailed investigation of the phenomena first observed by Higa [1] and by Reder and Bickart [2] enabled Strakhovskiĭ and Tatarenkov to observe experimentally[3] the very interesting effects described in detail in[4]. The experiments reported in these papers recall Ramsay's well-known experiments[5] with separated oscillating fields, except that they made use of active molecular beams at absolute temperature $T^\circ < 0$, that the oscillating field in one of the resonators was self-excited, and that the coupling between the two oscillating fields was not independent of the polarized beam. Individual singularities of the Strakhovskiĭ-Tatarenkov effects were considered by Basov and Oraevskiĭ[6] (see also[7]) under the assumption that the change in the population difference during the time of the travel of the molecules through the second oscillating field can be neglected (see[8]). These phenomena were qualitatively considered also by Wells[9].

It is perfectly obvious that the group of questions outlined above is closely related to the singularities of the interaction between a prepolarized beam and a high-frequency electromagnetic field. It is therefore advantageous to investigate in somewhat greater detail the singularities of such an interaction, without resorting beforehand, where possible, to the limitations used in [5-7,9], and under assumptions that admit of sufficiently simple intuitive physical interpretation of the results. It is apparently sensible to confine oneself to two basic physical problems—the polarization of the prepolarized beam in the specified field, and the reaction of such a beam on the field producing its polarization. The first problem is a feature of interaction in an unbounded space, where the proper radiation field has, by virtue of the radiation principle, the form of diverging waves and its reaction on the beam can be neglected. The second problem is essential in cavities bounded by conducting walls - resonators - where the beam's own radiation field cannot be neglected, since the radiation is also localized in the same bounded region of space.

Consequently, within the framework of the problem posed above, it is perfectly sufficient to consider the influence of an extraneous homogeneous field $E(z, t)$ directed along the beam on the prepolarization of the latter, and then find the field excited by the beam with due allowance for this influence. For a single molecule the first half of such a general problem was first considered in part in [5], but important physical features of the interaction remained, unfortunately, unnoticed and will be obtained further from more general considerations.

Assume that the molecule is acted upon by an electric field

$$E(z,t)=\begin{cases} E_0 e^{i\omega t}, & 0\leqslant z\leqslant L \\ 0, & z<0,\ z>L \end{cases}, \qquad (1)$$

directed along the beam, whose complex polarization [10] in the two-level approximation is

$$P(z,t)=P_0\rho_{12}(z,t)\exp(i\omega_{21}t), \qquad (2)$$

and the difference of the level populations of the component molecules is proportional to

$$D(z,t)=\rho_{11}(z,t)-\rho_{22}(z,t), \qquad (3)$$

where ω_{21} is the transition frequency, and $\rho_{ik}(z, t)$ the density-matrix elements determined from the Klimontovich-Khokhlov equations [11] with account of relaxation.[10] When $z < 0$ the beam is polarized, and therefore we have at the point $z = 0$

$$D(0,t) = D_0, \quad \rho_{12}(0,t) = \rho_{21}^*(0,t) = \rho_0 e^{i\delta t}, \quad \delta = \omega - \omega_{21}. \tag{4}$$

In the general case it is obvious that D_0 and ρ_0 are time-dependent, but if we take this circumstance into account, then the final expressions for $P(z, t)$ and $D(z, t)$ become very cumbersome and require a special detailed analysis. We shall henceforth consider only case (4) of a stationary[11] distribution of the molecules over the levels.

If the beam enters the field (1) from a source that is in thermodynamic equilibrium, then the distributions of the phases of ρ_{ik} is random [12], and ρ_0 [(Eq. (4)] is known only accurate to value of the phase. In the case of interest to us the beam either interacts initially with the extraneous field [5] or is transformed into a state with $T^\circ < 0$ and then participates in the self-oscillating process [1-3]. As shown by Tolman [12], this leads to phase ordering of ρ_0 or, what is the same, to the appearance of polarization. Therefore ρ_0 in (4) is a definite and known complex quantity. Regarding (4) as the boundary conditions for the solution of the Klimontovich-Khokhlov equations for $z \geq 0$, we put

$$D(z,t) = D_1(z) + D_2(z), \tag{5}$$

$$\rho_{12}(z,t) = [R_1(z) + R_2(z) + R_3(z)]\, e^{i\delta t}, \tag{6}$$

where

$$D_1(z) = \sum_{k=1}^{3} \Theta_0(p_k) \left[\frac{\tau D^e}{p_k} (e^{p_k z} - 1) + D_0 e^{p_k z} \right], \tag{7}$$

$$D_2(z) = -2 \sum_{k=1}^{3} \Phi_k(p_k)\, e^{p_k z}; \tag{8}$$

$$R_1(z) = \frac{\Lambda}{2v} \sum_{k=1}^{3} \Theta_0(p_k) \left\{ \frac{\tau D^e}{p_k} \left[\frac{\Gamma_k}{p_k + \varepsilon + i\beta} - \frac{1}{\varepsilon + i\beta} \times (1 - e^{-(\varepsilon + i\beta)z}) \right] + \frac{D_0 \Gamma_k}{p_k + \varepsilon + i\beta} \right\}, \tag{9}$$

$$R_2(z) = -\frac{\alpha}{v} \sum_{k=1}^{3} \Phi_k(p_k) \frac{\Gamma_k}{p_k + \varepsilon + i\beta}, \tag{10}$$

$$R_3(z) = \rho_0 \exp[-(\varepsilon + i\beta) z]. \tag{11}$$

The functions entering in (7)—(11) are defined by

$$\Theta_0(p_k) = [(p_k + \varepsilon)^2 + \beta^2] \left[\prod_{m \neq k} (p_k - p_m) \right]^{-1},$$

$$\Phi_k(p_k) = [\eta_1(\varepsilon + \operatorname{Re} p_k) - \eta_2 \beta + i\eta_1 \operatorname{Im} p_k] \left[v \prod_{m \neq k} (p_k - p_m) \right]^{-1}$$

(the products are taken over all $m = 1, 2, 3$, except $m = k$),

$$\Gamma_k = e^{p_k z} - e^{-(\varepsilon + i\beta)z}, \qquad p_1 = 2F_1 - \mu_0 - \varepsilon,$$
$$p_2 = -F_1 + i\sqrt{3} F_2 - \mu_0 - \varepsilon, \qquad p_3 = p_2^*$$
$$F_1 = {}^3/_2 \mu_0 ({}^1/_3 - \beta^2/\alpha^2) + O(\mu_0^2), \qquad F_2 = {}^1/_3 \sqrt{3}\alpha + O(\mu_0^2),$$

where $O(\mu_0^2)$ are terms of order of μ_0^2 and higher. The parameters of the problem are

$$\Lambda = \frac{2}{i\hbar} p_{12} E_0$$

(p_{12} is the matrix element of the dipole moment),

$$\alpha = \sqrt{\beta^2 + \varkappa^2}, \quad \beta = \delta/v, \quad \varkappa = |\Lambda|/v,$$
$$\eta_1 = \operatorname{Re} \Lambda \rho_0^*, \quad \eta_2 = \operatorname{Im} \Lambda \rho_0^*;$$

D^e is the equilibrium value of $D(z, t)$, and v is the average molecule velocity in the beam. The quantities ϵ and τ are inverse to the relaxation distances and are expressed in terms of the relaxation times $T_{1,2}$ [10]:

$$\tau = (vT_1)^{-1}, \quad \varepsilon = (vT_2)^{-1},$$

and μ_0 denotes their difference:

$$\mu_0 = (\tau - \varepsilon)/3.$$

It is easy to verify, by direct substitution in the Klimontovich-Khokhlov [11] equations with relaxation taken into account [10], that expressions (5) and (6) are solutions of these equations when the μ_0 are sufficiently small, and satisfy the boundary conditions (4). In the limiting case as $\tau_1 \epsilon_1 / \mu_0 \to 0$ we get from (9)—(11)

$$R_1(z) = \frac{\Lambda}{2\alpha v} D_0 \left[\sin \alpha z - i \frac{\beta}{\alpha} (1 - \cos \alpha z) \right], \tag{9a}$$

$$R_2(z) = -\frac{\eta_1 \Lambda}{v^2 \varkappa^2} \left[i \frac{\beta}{\alpha} \sin \alpha z - \cos \alpha z + e^{-i\beta z} \right] - \frac{\eta_2 \Lambda}{v^2 \alpha^2} (1 - e^{-i\beta z}) - \frac{\beta}{\alpha} \frac{\eta_2 \Lambda}{v^2 \alpha^2} \left[i \frac{\beta}{\alpha} \cos \alpha z + \sin \alpha z - i \frac{\beta}{\alpha} e^{-i\beta z} \right], \tag{10a}$$

$$R_3(z) = \rho_0 e^{-i\beta z}. \tag{11a}$$

By virtue of the neglect of the relaxation processes, expressions (5) and (6) satisfy in this limiting case not only the conditions (4), but also directly the initial Klimontovich-Khokhlov equations [11], and

when $\rho_0 = 0$ they go over, apart from the difference in notation, into the solutions obtained in that paper. Formulas (9a)—(11a) can also be obtained from the solutions of [5] by changing over in the latter to a description of the moving molecules in the laboratory frame, and then carry out a suitable averaging. With such an operation, the form of formulas (9a)—(11a) is not obvious beforehand, and it is therefore appropriate to emphasize once more that they have been derived as a particular case of the more general solutions (9)—(11), and are thus free of the limitations assumed in [5], i.e., they are suitable, according to (2) and (6), for a direct description of the polarization in molecular beams.

Substituting (5) and (6) in (2), we conclude that the polarization of a prepolarized beam in an extraneous field breaks up into three terms. Each of them has a clear-cut physical meaning which does not depend on the relaxation processes (compare (9)—(11) with (9a)—(11a)).

We shall call the component (2), which is proportional to R_1, the proper polarization. It is produced in the beam under the influence of the field (1), vanishes when $E_0 = 0$, and does not depend on ρ_0 [Eq. (4)]. The component proportional to R_3 is, in turn, independent of D_0 [Eq. (4)] and of the field (1), and is carried by the moving molecules of the beam. R_3 vanishes with ρ_0, i.e., with the vanishing of the prepolarization. Finally, that part of the polarization which is proportional to R_2 is likewise independent of E_0, but depends on ρ_0 and E_0, i.e., it appears as the result of the interaction between the prepolarization and the field (1); we shall therefore call it mixed polarization.

Such a breakdown is physically advantageous also because when (9a)—(11a) are expanded in powers of E_0 the convective polarization (which does not depend at all on (1)) appears in the zeroth order, then the proper polarization appears in first order, and only in the second and higher orders do the proper and mixed polarizations again appear. It is therefore clear that when $\rho_0 \neq 0$ the linear part of the interaction is determined by the convective and proper polarization, while the nonlinear part depends both on the proper and on the mixed polarization.

Consequently, the prepolarization ($\rho_0 \neq 0$) leads not only to the appearance of convective polarization, but also to a substantial change in the nonlinear part of the interaction, owing to the presence of mixed polarization. If we note in addition that each polarization component is characterized by a magnitude and a phase shift, then it becomes obvious that in cavities bounded by metallic surfaces (resonators), when the proper field of the beam radiation can no longer be neglected, the prepolarization can greatly change the character of the interaction between the beam and the field.

Let us assume that a prepolarized beam enters such a cavity and the natural modes of the cavity are known. We shall seek the field (1), assuming the polarization (2) to be known, in the form $E(z, t) = x(t)E(z)$, where $E(z)$ is the one cavity mode coinciding with the direction of the beam molecules. From Maxwell's equations we have

$$\ddot{x} + 2\frac{\omega_s}{Q}\dot{x} + \omega_0^2 x = -\frac{4\pi}{N}\int_V \frac{d^2\mathbf{P}}{dt^2}\mathbf{E}(z)\,dV,$$

$$N = \int_V |E(z)|^2\,dV, \qquad (12)$$

where V is the volume of the cavity, $\omega_0 = \omega_S(1 + Q^{-2})^{1/2}$ (the Q is assumed to be large enough to put henceforth $\omega_0 \approx \omega_S$).

Let the prepolarization $\mathbf{P}$ be either the result of a self-oscillation process [11] or the result of the action of an extraneous field [5] interacting with an identical mode in another similar cavity of length L_2. The distance between the two cavities is L_1. Then $\mathbf{P}$ in Eq. (12) contains only components coinciding in direction with $\mathbf{E}(z)$. Substituting (6) and (9a)—(11a) in (2) and (12) we obtain, in the case of sufficiently thin beams, an equation for the stationary amplitude of the forced oscillations at the frequency ω, in the form

$$\ddot{x} + \omega^2 x = -(\omega_0^2 - \omega^2)x - 2\frac{\omega_0}{\omega Q}\dot{x} + \mu\omega^2\langle R\rangle e^{i\omega t}, \qquad (13)$$

where

$$\mu = \frac{4\pi}{N}V_n E_0 P_0, \qquad \langle R\rangle = \frac{1}{L}\int_0^L R(z)\,dz,$$

$E_0 = E(z)$ is the homogeneous field and V_n the volume occupied by the beam.

To determine $\langle R\rangle$ we assume that the conditions in the auxiliary cavity correspond exactly to the Klimonotvich-Khokhlov conditions [11] ($\rho_0 = 0$ and $D_0 = D_2 \neq 0$). Then, taking account, with the aid of (9a), of the polarization at the exit from the first cavity and its behavior (11a) in the gap between the cavities, we put $x = ae^{i\omega t}$, after expanding $\langle R\rangle$ in powers of a:

$$\langle R\rangle = F_{00} + aF_{01} + a^2F_{02} + O(a^3), \qquad (14)$$

where

$$F_{00} = A\exp\{i[\xi + \Psi - \beta(L_1 + L/2)]\}, \qquad (15)$$

$$F_{01} = B\exp\{i[\xi + \Psi_1]\}, \qquad (16)$$

with $\xi = \pi$ for $T^\circ > 0$ and $\xi = 0$ for $T^\circ < 0$,

$$F_{02} = \frac{|\Lambda_0|^2}{2\delta^2}\{i[(\beta L u_0 - v_0) + i(\beta L - \beta L v_0 - 2u_0)]\,\mathrm{Im}\,\rho_0^* e^{i\varphi} + [\beta L(1 - v_0) - i(\beta L u_0 - v_0)]\,\mathrm{Re}\,\rho_0^* e^{i\varphi}\}\, e^{-i\varphi},$$

$$\delta > |a||\Lambda_0|, \quad F_{02} = \left(i\frac{|\Lambda_0|^2}{6v^2}\,\mathrm{Im}\,\rho_0^* e^{i\varphi}\right) e^{-i\varphi}, \quad \delta = 0, \quad (17)$$

and the quantities entering in these expressions are defined by

$$|\Lambda| = |a||\Lambda_0|, \quad a = |a| e^{i\varphi},$$

$$u_0 = \frac{1 - \cos\beta L}{\beta L}, \quad v_0 = 1 - \frac{\sin\beta L}{\beta L},$$

$$A = |D_2|\frac{\varkappa_2^2}{\alpha_2^2}\sin\frac{\alpha_2 L_2}{2}\left(\beta^2\varkappa_2^{-2} + \cos^2\frac{\alpha_2 L_2}{2}\right)^{1/2}\frac{\sin(\beta L/2)}{\beta L/2},$$

$$B = \frac{v|\Lambda_0|}{2\delta^2 L}\frac{|D_2|}{\alpha_2^2}(\beta^2 + \varkappa_2^2\cos\alpha_2 L_2)[2(1 - \cos\beta L - \beta L\sin\beta L) + \beta^2 L^2]^{1/2},$$

$$\mathrm{tg}\,\Psi = \frac{\alpha_2}{\beta}\,\mathrm{ctg}\frac{\alpha_2 L_2}{2}, \quad \mathrm{tg}\,\Psi_1 = \frac{u_0}{v_0}.$$

Putting further $n_0 = \omega^{-2}(\omega_0^2 - \omega^2)$ and $m_0 = 2\omega_0(\omega Q)^{-1}$, we get from (13) and (14) in the linear approximation

$$a_0 = |a| e^{i\varphi_0} = \mu F_{00}(n_0 + i m_0 - \mu F_{01})^{-1}, \quad (18)$$

where, according to (15) and (16),

$$|a_0| = \mu A[(n_0 + \mu B\cos\Psi_1)^2 + (m_0 + \mu B\sin\Psi_1)^2]^{-1/2},$$

$$\varphi_0 = \xi + \Psi - \Psi_a - \beta\left(L_1 + \frac{L}{2}\right),$$

$$\mathrm{tg}\,\Psi_a = \frac{m_0 + \mu B\sin\Psi_1}{n_0 + \mu B\cos\Psi_1}$$

($B > 0$ if $T^\circ > 0$ and $B < 0$ if $T^\circ < 0$). In this approximation, the field is determined entirely by the convective and the proper polarizations.

Inasmuch as F_{02} is proportional to $L^2|\Lambda|^2 v^{-2} \ll 1$ when $\delta = 0$, we shall estimate the influence of the mixed and the nonlinear part of the proper polarization by the small-perturbation method. Putting $a = a_0 + \epsilon_0$ and neglecting terms of order $|\epsilon_0|^2$ and higher in (13), we obtain

$$\varepsilon_0 = |\varepsilon_0| e^{i\varphi} = \mu a_0^2 F_{02}(n_0 + i m_0 - \mu F_{01} - 2a_0 F_{02})^{-1}, \quad (19)$$

where according to (16) and (17) we have for $\delta = 0$

$$|\varepsilon_0| = \mu\frac{\mu^2|\Lambda_0|^2 A^2 L^2}{12v^2}\sin 2\Psi_a\cos\Psi_a\cos\left(\varphi - \varphi_0 + \frac{\pi}{2}\right),$$

$$\mathrm{tg}\left(\varphi - \varphi_0 + \frac{\pi}{2}\right) = \mathrm{tg}\,\Psi_a + \frac{\mu^2 A^2|\Lambda_0|^2 L^2}{6v^2(n_0 + \mu B\cos\Psi_1)^2}\sin 2\Psi_a.$$

However, when the field amplitudes determined by (18) and (19) are sufficiently small, they can be regarded as extraneous self-consistent fields, if the spectrum of the proper modes of the cavity is so sparse that it is reasonable to neglect the neighboring oscillations. It becomes possible in this manner, in an approximation where μ is small, to observe the characteristic singularities of the interaction with the field of a closed cavity.

When $a \to 0$ we have $\langle R\rangle \to F_{00}$ [Eq. (14)], and therefore if $n_0, m_0 \gg \mu B$ [Eq. (18)] then the interaction occurs only with the proper field of the beam radiation, which is determined entirely by the convective polarization. If $n_0, m_0 \sim \mu B$, then the proper polarization is significant. When $T^\circ > 0$ [5], this polarization only increases the absorption (18) in the beam, since $B > 0$. To the contrary, when $T^\circ < 0$ [1-3] we have $B < 0$ and the growth of the field amplitude (18) is limited by the terms of order $O(a^2)$ [Eq. (14)], i.e., by the proper and mixed polarizations. Consequently when $T^\circ < 0$, other conditions being equal, the nonlinear character of the polarization is more important than when $T^\circ > 0$.

Expression (19) provides an estimate of the region where the nonlinearity has little influence. It turns out here, as can be readily verified, that the effects observed in [1-3] are described in the linear approximation by Eq. (18). When $T^\circ < 0$, however, as already emphasized above, the interaction is essentially nonlinear, and therefore such an approximate description cannot explain satisfactorily the experimental observations in all respects.

Nonetheless, it is quite evident that the singularities of the polarization of a prepolarized beam are quite sufficient for the development of a rigorous nonlinear theory of interaction in a cavity. The solution of such a problem is bound to be very cumbersome, and its investigation is therefore beyond the scope of the present paper.

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