

*POLARIZATION OF A BEAM OF MOLECULES IN AN ALTERNATING FIELD
WITH VARIABLE AMPLITUDE AND PHASE*

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Equations are derived which describe the behavior of a beam of molecules under the action of an alternating field with variable amplitude and phase; these equations are solved for various cases. An exact solution is obtained for the case in which the frequency of the applied field, the amplitude of which is varying, is the same as the frequency of the molecular transition. Approximate solutions are obtained for the cases in which the amplitude and frequency of the field vary slowly or rapidly during the time of flight through the cavity.

THE operation of the molecular oscillator is based on the interaction between molecules which move through a cavity and the electric field in the cavity.¹ As a result of this interaction the molecules make a transition from an upper energy state to a lower energy state, thereby giving energy to the cavity. The state of the molecular beam which interacts with the electric field in the cavity can be described by means of a polarization function. In general the relations between this polarization and the electric field in the cavity is extremely complicated. At the present time this relation has been determined only for the case in which the beam of molecules interacts with an alternating electric field whose frequency and amplitude remain constant in time. However, in analyzing all possible transient processes in a molecular oscillator and in analyzing the stability of the steady-state oscillations, it is necessary to determine the polarization of a beam which is subject to an alternating field of variable amplitude and phase (frequency).

The present paper is devoted to a theoretical study of the polarization of the beam of molecules under these conditions.

**1. FORMULATION OF THE PROBLEM AND DERIVATION
OF THE BASIC EQUATION**

The state of a monochromatic beam of molecules, acted on by an electric field $E(t)$ is determined completely by the molecular density matrix $C_{jk}(x, t)$. The dependence of the matrix elements on the coordinate x and the time t is determined by the following system of partial differential equations:²

$$\frac{\partial D}{\partial t} + v \frac{\partial D}{\partial x} = -\frac{2p}{i\hbar} [e^{i\omega_0 t} C_{12} - e^{-i\omega_0 t} C_{21}] E(t), \quad \frac{\partial C_{12}}{\partial t} + v \frac{\partial C_{12}}{\partial x} = -\frac{p}{i\hbar} e^{-i\omega_0 t} D E(t), \quad \frac{\partial C_{21}}{\partial t} + v \frac{\partial C_{21}}{\partial x} = \frac{p}{i\hbar} e^{i\omega_0 t} D E(t). \quad (1.1)$$

Here $D(x, t)$ is a function which characterizes the molecular distribution over energy states: $D = C_{22} - C_{11}$, v is the velocity of the molecules, p is the dipole moment of the molecules, and ω_0 is the frequency of the molecular transition.

The polarization of the beam is related to the elements of the density matrix by the expression

$$P(x, t) = p [e^{i\omega_0 t} C_{12} + e^{-i\omega_0 t} C_{21}]. \quad (1.2)$$

As is apparent from Eq. (1.2), the polarization of the beam is intimately related with the molecular transitions from one energy state to another due to the action of the cavity field. At the entrance to the cavity, where the molecules have not as yet made any energy transitions,

$$C_{12} = C_{21} = 0 \text{ for } x = 0. \quad (1.3)$$

We shall assume, moreover, that the number of active molecules which enter the resonator cavity is such that

$$D = 1 \text{ for } x = 0. \quad (1.4)$$

The further development is limited to the case in which the variation of the electric field in time is more or less sinusoidal, so that

$$E(t) = E_0(t) \cos[\omega_0 t + \varphi(t)], \quad (1.5)$$

where the field amplitude $E_0(t)$ and phase $\varphi(t)$ are slowly-varying functions of time as compared with the period $T = 2\pi/\omega_0$. Under these conditions the polarization of the beam can be written in the form of a sum of real and reactive parts:

$$P(x, t) = P_1(x, t) \sin[\omega_0 t + \varphi] + P_2(x, t) \cos[\omega_0 t + \varphi]. \quad (1.6)$$

The amplitudes of the real and reactive parts, P_1 and P_2 , are slowly-varying functions of time. If, in the right halves of Eqs. (1.1) we neglect terms which vary rapidly in time (frequency $2\omega_0$) and introduce new variables defined by the relations

$$\xi = x - vt; \quad \eta = x + vt, \quad (1.7)$$

the system of equations in (1.1) assumes the form

$$\frac{\partial D}{\partial \eta} = \frac{E_0 p}{2i\hbar v} [e^{i\varphi} C_{21} - e^{-i\varphi} C_{12}], \quad \frac{\partial C_{12}}{\partial \eta} = -\frac{p}{4i\hbar v} e^{i\varphi} E_0 D, \quad \frac{\partial C_{21}}{\partial \eta} = \frac{p}{4i\hbar v} e^{-i\varphi} E_0 D. \quad (1.8)$$

In this case the boundary conditions given in (1.3) and (1.4) assume the form

$$D = 1, \quad C_{12} = C_{21} = 0 \quad \text{for } \eta = -\xi. \quad (1.9)$$

The values of the elements of the density matrix C_{12} and C_{21} and consequently the polarization of the beam $P(x, t)$, in accordance with the second and third equations in (1.8), can be expressed in terms of the function D as follows:

$$C_{12} = -\frac{p}{4i\hbar v} \int_{-\xi}^{\eta} e^{i\varphi(\zeta)} E_0(\zeta) D(\zeta) d\zeta, \quad C_{21} = \frac{p}{4i\hbar v} \int_{-\xi}^{\eta} e^{-i\varphi(\zeta)} E_0(\zeta) D(\zeta) d\zeta. \quad (1.10)$$

Substituting (1.10) in (1.2) and, in accordance with (1.6), dividing the polarization into real and reactive parts, we obtain the expressions

$$P_1 = -\frac{p^2}{2\hbar v} \int_{-\xi}^{\eta} \cos[\varphi(\zeta) - \varphi(\eta)] E_0(\zeta) D(\zeta) d\zeta, \quad P_2 = -\frac{p^2}{2\hbar v} \int_{-\xi}^{\eta} \sin[\varphi(\zeta) - \varphi(\eta)] E_0(\zeta) D(\zeta) d\zeta. \quad (1.11)$$

Whence it is easy to show that the system given in (1.8) is equivalent to the following system of equations:

$$\frac{\partial D}{\partial \eta} = \frac{1}{2\hbar v} E_0(\eta) P_1, \quad \frac{\partial P_1}{\partial \eta} = \varphi'(\eta) P_2 - \frac{p^2}{2\hbar v} E_0(\eta) D, \quad \frac{\partial P_2}{\partial \eta} = -1 \varphi'(\eta) P_1 \quad (1.12)$$

with the boundary conditions at $x = 0$

$$D = D_0, \quad P_1 = P_2 = 0. \quad (1.13)$$

The function D is described by an integro-differential equation which can be obtained by substituting (1.10) in the first equation of (1.8):

$$\frac{\partial D}{\partial \eta} = -\gamma^2 E_0(\eta) \int_{-\xi}^{\eta} E_0(\zeta) \cos[\varphi(\eta) - \varphi(\zeta)] D(\zeta) d\zeta; \quad (1.14)$$

here $\gamma = p/2\hbar v$.

On the other hand, the function D obeys the differential equation

$$\frac{d^2}{d\eta^2} \left[\frac{1}{E_0(\eta)} \frac{\partial D}{\partial \eta} \right] - \frac{\varphi''(\eta)}{\varphi'(\eta)} \frac{d}{d\eta} \left[\frac{1}{E_0(\eta)} \frac{\partial D}{\partial \eta} \right] + \gamma^2 \frac{d}{d\eta} [E_0(\eta) D(\eta)] + \frac{\varphi'^2(\eta)}{E_0(\eta)} \frac{\partial D}{\partial \eta} - \gamma^2 \frac{\varphi''(\eta)}{\varphi'(\eta)} E_0(\eta) D(\eta) = 0 \quad (1.15)$$

with the boundary conditions

$$D = 1, \quad D' = 0, \quad D'' = -\gamma^2 E_0^2(-\xi) \quad \text{for } \eta = -\xi, \quad (1.16)$$

which can be obtained starting from the system in (1.12) for the functions P_1 and P_2 . The equivalence of (1.14) and (1.15) is easily demonstrated. Thus, the determination of the polarization of the beam of molecules being acted on by an electric field such as that given in (1.5) leads to the solution of the differential equation in (1.15). This equation is not amenable to solution in the general case in which both the amplitude and phase of the field vary in time. However, Eq. (1.15) can be solved if certain limitations are imposed on the amplitude and phase of the field. The remainder of this paper is devoted to such solutions.

2. APPLIED FIELD WITH CONSTANT PHASE

In the case in which the frequency of the electric field acting on the beam of molecules is equal to the frequency of the molecular transition but the amplitude E_0 varies in some arbitrary manner the solution of Eq. (1.15) can be found by quadratures. In Eq. (1.15) we set $\varphi = \text{const} = 0$. Then the equation assumes the form

$$\frac{d^2}{d\eta^2} \left[\frac{1}{E_0(\eta)} \frac{\partial D}{\partial \eta} \right] + \gamma^2 \frac{d}{d\eta} [E_0(\eta) D(\eta)] = 0. \quad (2.1)$$

Integrating Eq. (2.1) once and setting the constant of integration equal to zero, we have

$$\frac{d}{d\eta} \left[\frac{1}{E_0(\eta)} \frac{\partial D}{\partial \eta} \right] + \gamma^2 E_0(\eta) D(\eta) = 0. \quad (2.2)$$

As can be shown by direct substitution, the general solution for this equation is given by the expression

$$D = A \cos \left[\gamma \int_{-\xi}^{\eta} E_0(\zeta) d\zeta \right] + B \sin \left[\gamma \int_{-\xi}^{\eta} E_0(\zeta) d\zeta \right]. \quad (2.3)$$

Here A and B are constants of integration. The boundary conditions in (1.8) are satisfied if $A = 1$ and $B = 0$.

In accordance with (1.12), in the constant-phase case the reactive part of the polarization P_2 vanishes while the real part becomes

$$P_1 = 2h\nu \frac{1}{E_0} \frac{\partial D}{\partial \eta} = -p \sin \left[\gamma \int_{-\xi}^{\eta} E_0(\zeta) d\zeta \right]. \quad (2.4)$$

If we now transform back to the variables x and t , the argument of the sine function in (2.4) assumes the form

$$2v\gamma \int_0^{x/v} E_0(t-t') dt'. \quad (2.5)$$

An examination of this expression shows that the time variations of the field amplitude are averaged over an interval x/v which is of the order of magnitude of the molecular time of flight across the cavity. This situation is the basis for the "inertia non-linearity" of a molecular oscillator where the time constant of the "inertia" is equal to the time of flight across the cavity. It also follows from Eq. (2.5) that the quasi-static expression for the function D

$$D = \cos 2\gamma Ex,$$

as well as the corresponding expression for the polarization of the beam

$$P(x, t) = -p \sin \omega_0 t \sin 2\gamma Ex,$$

can be used in the case in which the amplitude of the field varies very slowly,

$$\tau E' / E \ll 1.$$

It will be shown below that these results can be extended to the general case in which both the amplitude and phase of the field vary although Eq. (1.15) cannot then be solved by quadratures.

3. APPLIED FIELD WITH SLOWLY VARYING AMPLITUDE AND PHASE

In the case in which the amplitude E_0 and frequency φ' vary slowly as compared with the time interval τ , it is possible to find an asymptotic solution for Eq. (1.15); the method is similar to that used to find the quasi-classical approximations for the Ψ -function in quantum mechanics³ or the geometric-optic methods used in the analysis of the propagation of electromagnetic waves.⁴ For this purpose we write the solution of Eq. (1.15) in the form

$$D(\eta) = e^{i\Phi(\eta)}. \quad (3.1)$$

The equation which describes the function $\Phi(\eta)$ is of the form:

$$-i\Phi'^3 - 3\Phi'\Phi'' + i\Phi''' - \left[2\frac{E'}{E} + \frac{\varphi''}{\varphi'}\right][i\Phi'' - \Phi'^2] + [\gamma^2 E^2 + \varphi'^2 + 2\left(\frac{E'}{E}\right)^2 - \frac{E''}{E} + \frac{\varphi'''}{\varphi'}]i\Phi' + \gamma^2 E^2 \left[\frac{E'}{E} - \frac{\varphi''}{\varphi'}\right] = 0. \quad (3.2)$$

In the case in which E and φ' are constant in time, the function D is a periodic function of η , with frequency²

$$\Omega = \sqrt{\gamma^2 E^2 + \varphi'^2}. \quad (3.3)$$

If the amplitude and phase of the field do not vary significantly over a period of the function D , we can introduce a large parameter λ , which characterizes the rate of change of the function D as compared with the variations of the functions E and φ' :

$$\gamma E \sim \lambda, \quad \varphi' \sim \lambda, \quad E'/E \sim 1, \quad \varphi''/\varphi' \sim 1. \quad (3.4)$$

In this case, the function Φ in Eq. (3.1) can be expanded in powers of λ

$$\Phi = \lambda\Phi_0(\eta) + \Phi_1(\eta) + \frac{1}{\lambda}\Phi_2(\eta) + \dots \quad (3.5)$$

Substituting (3.4) in Eq. (3.1), expanding the left half in powers of λ , taking account of (3.4), and equating terms with the same power of λ , we have

$$\lambda^3) \quad -\Phi_0'^3 + (\gamma^2 E^2 + \varphi'^2)\Phi_0' = 0, \quad (3.6)$$

$$\lambda^2) -3\Phi_0'\Phi_0'' - 3i\Phi_0'^2\Phi_1' + 2\frac{E'}{E}\Phi_0'^2 + \frac{\varphi''}{\varphi'}\Phi_0'^2 + i\gamma^2 E^2\Phi_1' + i\varphi'^2\Phi_1' - \gamma^2 E^2\frac{\varphi''}{\varphi'} + \gamma^2 E^2\frac{E'}{E} = 0. \quad (3.7)$$

The solutions of these equations are of the form:

$$\Phi_{01}' = 0; \quad \Phi_{02,3}' = \pm \sqrt{\gamma^2 E^2 + \varphi'^2}, \quad \Phi_{11} = -i \ln \frac{\varphi'}{\sqrt{\gamma^2 E^2 + \varphi'^2}}; \quad \Phi_{12,3} = -i \ln \frac{\gamma E}{\sqrt{\gamma^2 E^2 + \varphi'^2}}. \quad (3.8)$$

Substituting these solutions in (3.1) and taking account of the boundary conditions (1.16) we can obtain an approximate expression for the function D in the case in which the amplitude and frequency of the field which acts on the beam are slowly varying

$$D(x,t) = \frac{\gamma^2 E(t) E \left(t - \frac{x}{v}\right) \cos \int_{t-x/v}^t 2v \left(\gamma^2 E^2(t_1) + \frac{\varphi'^2(t_1)}{4v^2}\right)^{1/2} dt_1 + \frac{\varphi'(t)\varphi'(t-x/v)}{4v^2}}{[\gamma^2 E^2(t) - \varphi'^2(t)/4v^2]^{1/2} [\gamma^2 E^2(t-x/v) - \varphi'^2(t-x/v)/4v^2]^{1/2}}. \quad (3.9)$$

This expression, which applies when

$$\tau\varphi''/\sqrt{\gamma^2 E^2 + \varphi'^2} \ll 1, \quad \tau\gamma E'/\sqrt{\gamma^2 E^2 + \varphi'^2} \ll 1, \quad (3.10)$$

indicates that the beam tends to average the effect of the field with variable amplitude and frequency during the time in which the molecules move through the cavity; this effect, however, is small because the changes are slow. Substituting Eq. (3.9) in Eq. (1.12) it is easy to determine the polarization of the beam.

4. APPLIED FIELD WITH RAPIDLY VARYING AMPLITUDE AND FREQUENCY

Equation (1.15) can also be solved approximately when there are very rapid periodic changes in the amplitude and frequency of the field which acts on the beam, i.e., changes fast as compared with the time

of flight of the molecules through the cavity. Let

$$E_0 = E_1 + e(\lambda, t); \quad \varphi' = \varphi_1' + \chi(\lambda, t), \quad (4.1)$$

where E_1 and $\varphi_1' = \delta/2v$ are constants ($E_1 \neq 0$, $\delta = \omega - \omega_0$), which characterize the mean amplitude and frequency deviation of the field while $e(t)$ and $\chi(t)$ are periodic functions of time whose periods are smaller by a factor of λ than the molecular time of flight through the cavity ($\lambda \gg 1$).

In solving Eq. (1.15) for $D(\eta)$ in this case we can separate terms which vary rapidly with a change in the argument from those which vary slowly:

$$D(\eta) = D_0(\eta) + \frac{1}{\lambda} D_1(\eta) + \dots + \frac{1}{\lambda} d_0(\lambda, \eta) + \frac{1}{\lambda^2} d_2(\lambda, \eta) + \dots \quad (4.2)$$

We substitute (4.1) and (4.2) in Eq. (1.14) and collect terms in the same power of the parameter λ . In the zeroth approximation we have

$$D'_0 + d'_0 = -\gamma^2 E_1^2 \int_{-\xi}^{\eta} \cos \delta(\eta - \zeta) D_0(\zeta) d\zeta - \gamma^2 E_1 e(\lambda, \eta) \int_{-\xi}^{\eta} \cos \delta(\eta - \zeta) D_0(\zeta) d\zeta. \quad (4.3)$$

Separating rapidly-varying and slowly-varying terms in this expression we obtain

$$D'_0 = \gamma^2 E_1^2 \int_{-\xi}^{\eta} \cos \delta(\eta - \zeta) D_0(\zeta) d\zeta, \quad (4.4)$$

$$d'_0 = (e/E_1) D'_0. \quad (4.5)$$

The solution of Eq. (4.4) indicates that in the zeroth approximation rapid variations in the amplitude and frequency of the field which acts on the beam have no effect on the distribution of the molecules over states—the field is averaged. The rapid time variations in the molecular distribution over states are small in absolute magnitude and are determined, in accordance with (4.5), by the change in the field amplitude. The rapid changes in the frequency of the field affect the function $D(\eta)$ only to second order in $1/\lambda$.

In a similar fashion we can extend the calculations to the higher approximations. By this means it can be shown that the slowly-varying part $D_1(\eta) = 0$.

Hence, with accuracy up to terms of order $1/\lambda$ inclusive, for a field which is characterized by rapid changes in amplitude and frequency the solution of Eq. (1.15) is given by the relation:

$$D(\eta) = D_0(\eta) + \frac{1}{\lambda} d_0(\lambda, \eta). \quad (4.6)$$

The polarization of the beam, given by the expression

$$P = -p\gamma \int_{-\xi}^{\eta} \sin[\omega_0 t + \varphi(\zeta)] E_1(\zeta) D(\zeta) d\zeta, \quad (4.7)$$

can now be found easily. With accuracy to first order this quantity is

$$P = P_s + P_f, \quad (4.8)$$

where P_s is the part of the polarization which varies slowly with time

$$P_s = -p\gamma \frac{1}{\Omega^2} \left\{ \Omega \sin 2\Omega x [E_1 \sin \omega t + \overline{e\chi} \cos \omega t] + \delta(1 - \cos 2\Omega x) [E_1 \cos \omega t + \overline{e\chi} \sin \omega t] \right\}. \quad (4.9)$$

Here $\overline{e\chi}$ is the mean value of the product of the amplitude deviation and the frequency deviation.

The rapidly varying part of the polarization is given by the expression:

$$P_f = -\frac{p^2}{h} D_0(x) \int_{t-x/v}^t e(t') dt'. \quad (4.10)$$

In conclusion we may note that the relations which have been obtained for the polarization of a beam of molecules are rather simple and can be used directly in studying quasi-periodic modes in

the molecular oscillator; an investigation of this kind, however, is beyond the scope of the present paper.

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CONTRIBUTION TO THE THEORY OF TRANSITION RADIATION

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The transition radiation and the Cerenkov radiation which are produced when a charged particle moves successively through two media which differ in their dielectric and magnetic properties are considered. The cases in which the particle moves from vacuum into the medium and from the medium into vacuum are considered in detail.

THE transition radiation which is produced when a particle moves from a medium characterized by a given dielectric constant into another whose dielectric constant differs from the first was first considered by Ginzburg and Frank¹ (see also Refs. 2-4). In the present paper we consider the radiation fields which are produced in the general case for media which differ in both their dielectric and magnetic properties; certain particular cases are analyzed.

1. GENERAL CASE

We consider the field associated with a particle which has a velocity \mathbf{v} and moves from one medium into another. The first medium will be characterized by the macroscopic constants ϵ_1 and μ_1 (the dielectric constant and magnetic permeability); the second medium is characterized by ϵ_2 and μ_2 . We shall assume that the energy lost by the particle per unit length of path is negligibly small compared with its kinetic energy. Under these conditions the field associated with the particle is given by Maxwell's equations

$$\begin{aligned} \operatorname{curl} \mathbf{H} &= \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t} + \frac{4\pi}{c} \mathbf{v} e \delta(\mathbf{r} - \mathbf{v}t), \quad \operatorname{curl} \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}, \\ \operatorname{div} \mathbf{B} &= 0 \quad \operatorname{div} \mathbf{D} = 4\pi e \delta(\mathbf{r} - \mathbf{v}t). \end{aligned} \quad (1)$$

It will be assumed that the particle moves along the z -axis from $-\infty$ to $+\infty$ and that the interface between the two media is the plane $z = 0$ through which the particle moves at $t = 0$. We resolve the field and currents in triple Fourier integrals:⁵