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KINETIC THEORY OF OPTICAL MASER

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ABSTRACT

The kinetic theoretic study of the interaction between a coherent light wave and the polarized molecular beam at the near free molecular flow region is presented. A theory corresponding to the weak signal model is developed and the method of harmonic analysis is incorporated to study the nature of the generation of higher harmonics caused by non-linear interaction. Calculations of the first seven harmonics including D.C. components, $\pm w$, $\pm 2w$ and $\pm 3w$ and the time variation of the amplitude of electric field intensity, governed by the integro-differential equations are presented. The theory is applied to the interaction of a linearly polarized wave, at near resonant frequency, and a beam whose distribution function is near Maxwellian. The condition for the self-sustained oscillation of the maser is obtained in terms of the kinetic parameters and the electric conductivity of the beam. The linear and non-linear susceptibilities are obtained as a function of the plasma dispersion function. The dispersion curves and the absorption coefficients, calculated at two different densities, reveal that the reduction in the active medium density leads to the reduction in the frequency interval for the self-sustained oscillation and also the reduction in the coefficient of negative absorption.

1. INTRODUCTION

Recent success in producing or amplifying the coherent light beam in laser or maser technology has stimulated a great deal of theoretical and experimental interest in the problem of the interaction between matter and light. The theories of the optical maser, and other related subjects, have been continuously refined in response to the ever increasing degree of accuracy achieved in the experimental measurement.

Nevertheless, the existing theoretical studies are primarily concerned with homogeneous media, thus completely or partially ignoring the molecular interaction at the kinetic level. Some authors introduce certain statistical factors to account for the collective behavior of the molecular beam, yet such a phenomenological treatment renders no accurate information for the maser operating at certain critical stages where the interaction between molecules begins to inhibit maser action.

Experimental evidence seems to indicate that the practical maser beam is of an intermediate nature between free molecular and viscous flow, in which the effect of intermolecular collisions and molecular acceleration could alter the nature of the light and matter interaction. Further experiments reveal the ultimate collapse of the maser oscillation at higher pressure, presumably caused by the enhanced intermolecular collisions, indicating a coupling between light and matter at the kinetic level for which conventional theory is powerless. Unfortunately, no kinetic theoretical study of the maser has been explored, and many experimental results are greatly in need of theoretical support.

The purpose of the present paper is to develop a kinetic theory of the maser at near free molecular flow. In particular, our questions are concerned with (i) the response of the near free molecular maser beam to an electric field at near resonant frequencies (ii) the condition for the self-sustained maser oscillation, (iii) the harmonic generation of light, and (iv) the susceptibility of the near free molecular maser beam. The hydrodynamic approach which requires independent treatment will not be included in the present paper.

In the present study the molecular beam is assumed to obey the classical Boltzmann equation in which the collision integral is approximated by Krook's statistical model. The response of the dipole moment of a molecule is calculated from quantum theory.

In section II, the Maxwell, Schrödinger, and Krook equations are recapitulated, and the perturbation scheme for near free molecular flow is discussed. In section III, the harmonic analysis is used to solve the system of perturbed coupled equation. The Maxwell Equations are deduced in the form of an integro-differential equation. The analysis is carried out exclusively by a matrix formalism which provides a particularly convenient way of investigating harmonic generation due to non-linear interaction. The method of the calculation of the first seven harmonics, namely the d.c. component, and $\pm w$, $\pm 2w$, $\pm 3w$ generated by the interaction between maser and linearly polarized wave at frequency $+w$ is presented.

The application of the theory is demonstrated in section IV, where the condition for the self sustained oscillation, the case of stimulated emission, and the corresponding inverse dispersion curves are presented. The response of the polarization and the lowest order non-linear susceptibility are also discussed.

II MATHEMATICAL FORMULATION

We consider a molecular beam consisting of, say, ammonia molecules whose distribution function is $f(x,u,t)$. The molecules flow in a one-dimensional resonant cavity where they interact with the coherent light wave at near resonant frequency.

The distribution function satisfies the Boltzmann equation with the collision integral approximated by Krook's statistical model

$$\frac{\partial f}{\partial t} + u\frac{\partial f}{\partial x} + \frac{F_x}{m}\frac{\partial f}{\partial u} = \frac{1}{\tau_c}(F_m - f) \tag{1}$$

F_m is the local Maxwellian distribution function, and τ_c is the characteristic time for collision, which will be assumed to be constant for the sake of simplicity. The force F_x is the axial force acting on a molecule and is given as[1]

$$F_x = \frac{\partial}{\partial x}(V) = \frac{\partial}{\partial x}(pE) \tag{2}$$

p is the dipole moment, and E is the electric field intensity.

The Maxwell equation may be combined and written in the form of the one-dimensional wave equation

$$-\frac{\partial^2 E}{\partial x^2} + \mu_o\sigma\frac{\partial E}{\partial t} + \mu_o\epsilon_o\frac{\partial^2 E}{\partial t^2} = -\mu_o\frac{\partial^2 P}{\partial t^2} \tag{3}$$

where $P(x,t)$ is the macroscopic polarization density defined by

$$P(x,t) = \int_{-\infty}^{\infty} f(x,u,t)\ p(x,u,t)du \tag{4}$$

The dipole moment $p(x,u,t)$ is to be calculated from the quantum theory.

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1 W. K. H. Panofsky and M. Phillips, Classical Electricity and Magnetism (Addison-Wesley Publishing Company, Inc. Mass. 1956)

For matter interacting with the electric field, the system is described by a wave function $\tilde{\psi}$ satisfying Schrödinger Equation.

$$i\hbar \frac{\partial \tilde{\psi}}{\partial t} = (H_o - \mu E)\tilde{\psi} \tag{5}$$

where H_o is the Hamiltonian of the non-interacting system and μE is the interaction potential.

It will be assumed that the molecule consists of two energy levels W_a and W_b, and that the wave function $\tilde{\psi}$ is expressed by a linear combination of eigenfunctions $\tilde{\psi}_a$ and $\tilde{\psi}_b$ of the two stationary states

$$\tilde{\psi} = a(x,t)\tilde{\psi}_a + b(x,t)\tilde{\psi}_b \tag{6}$$

The coefficients $a(x,t)$, $b(x,t)$ are to be determined from the following equations

$$\frac{\partial a}{\partial t} = \frac{i}{h}(-W_a a + \mu_{ab} Eb) \tag{7a}$$

$$\frac{\partial b}{\partial t} = \frac{i}{h}(-W_b b + \mu^*_{ab} Ea) \tag{7b}$$

where μ_{ab} is the matrix element of the dipole moment defined as

$$\mu_{ab} = \mu^*_{ba} = \int_v \tilde{\psi}^*_a \mu \tilde{\psi}_b dV \tag{8}$$

The integration is taken over the configuration space of the molecule. Note $\mu_{aa} = \mu_{bb} = 0$, if the energy levels are not degenerate. The molecular dipole moment is given by

$$p = \int_v \tilde{\psi}^* \mu \tilde{\psi} dV = \mu_{ab} ab^* + \mu_{ba} a^* b \tag{9}$$

The dipole, moment p, can be calculated from Eqs. (7a), (7b), (8) and (9) according to the time dependent perturbation theory provided the electric field is known. However, it is also possible,[2] and as a matter of fact more convenient, to deal with the following system of equations, which are derived from Eqs. (7a), (7b), (8) and (9)

$$\left(\frac{\partial}{\partial t} + u\frac{\partial}{\partial x}\right)^2 p(x,u,t) + w_{ab}^2 p(x,u,t) = -2w_{ab}h^{-1}\mu_{ab}^2 E(x,t)R(x,u,t) \qquad (10)$$

$$\frac{\partial R(x,u,t)}{\partial t} + u\frac{\partial R(x,u,t)}{\partial x} = 2h^{-1}\omega_{ab}^{-1}E(x,t)\left(\frac{\partial P(x,u,t)}{\partial t} + u\frac{\partial p(x,u,t)}{\partial x}\right) \qquad (11)$$

where $R(x,u,t) = a(x,u,t)^2 - b(x,u,t)^2$ (12a)

and $\omega_{ab} = h^{-1}(w_a - w_b)$ (12b)

Eq. (10) represents the response of the dipole moment to the electric field whereas Eq. (11) governs the change of the probability for a molecule to occupy the active state. Note that the time derivative $\frac{\partial}{\partial t}$, originally appearing in Eqs. (5), (7a), and (7b) are replaced by total time derivative $\frac{\partial}{\partial t} + u\frac{\partial}{\partial x}$.[2] The system of Eqs. (1), (3), (4), (10), (11) and other related auxiliary equations are non-linear coupled equations. By virtue of the near free molecular assumption, we consider a perturbative scheme corresponding to a small deviation of the distribution function from the initial function. In as much as the acceleration term is proportional to the interaction potential pE, which would be regarded as a perturbation quantity, it is natural to treat the acceleration as a small quantity.

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2 A.N. Oraevskii, "A Theoretical Study of the Frequency Stability of Maser", in "Soviet Maser Research" Ed. by D.V. Skobeltsyn Transactions of P.N. Lebedev Physics Institute; Vol. XXI. (1963) [English Translation Consultant Bureau, N.Y. 1964]

III EXPANSION IN HIGHER HARMONICS

(A) Free molecular approximation:

According to previous discussion, the following type of solutions will be assumed

$$f(x,u,t) = f_o(x,u,t) + \frac{1}{\tau_c} f_1^{\dagger}(x,u,t) + f_1(x,u,t) + \ldots \tag{13a}$$

$$p(x,u,t) = p_o(x,u,t) + \frac{1}{\tau_c} p_1^{\dagger}(x,u,t) + p_1(x,u,t) + \ldots \tag{13b}$$

$$R(x,u,t) = R_o(x,u,t) + \frac{1}{\tau_c} R_1^{\dagger}(x,u,t) + R_1(x,u,t) + \ldots \tag{13c}$$

$$E(x,t) = E_o(x,t) + \frac{1}{\tau_c} E_1^{\dagger}(x,t) + E_1(x,t) + \ldots \tag{13d}$$

$$P(x,t) = P_o(x,t) + \frac{1}{\tau_c} P_1^{\dagger}(x,t) + P_1(x,t) + \ldots \tag{13e}$$

To zeroth order free molecular approximation, we have

$$\frac{\partial f_o}{\partial t} + u\frac{\partial f_o}{\partial x} = 0 \tag{14}$$

$$-\frac{\partial^2 E_o}{\partial x^2} + \mu_o \sigma \frac{\partial E_o}{\partial t} + \mu_o \epsilon_o \frac{\partial^2 E_o}{\partial t^2} = -\mu_o \frac{\partial^2 P_o}{\partial t^2} \tag{15}$$

$$P_o(x,t) = \int_{-\infty}^{\infty} p_o(x,u,t)\ f_o(x,u,t)du \tag{16}$$

$$\left(\frac{\partial}{\partial t} + u\frac{\partial}{\partial x}\right)^2 p_o + \omega_{ab}^2 p_o = \gamma\ E_o R_o \tag{17}$$

$$\frac{\partial R_o}{\partial t} + u\frac{\partial R_o}{\partial x} = \beta\ E_o\left(\frac{\partial p_o}{\partial t} + u\frac{\partial p_o}{\partial x}\right) \tag{18}$$

where $\gamma = -2\omega_{ab} h^{-1} \mu_{ab}^2$, $\beta = 2h^{-1}\omega_{ab}^{-1}$.

One important fact in the maser oscillation is the harmonic generation by non-linear interaction, characterized by non-linear coupling terms appearing in Eqs. (17) and (18). Thus when lineary polarized light wave, such as $A\exp i(kx-wt) + A^{*}\exp[-i(kx+wt)]$, shines on the maser beam, certain higher harmonics will also be created. Anticipating such a non-linear response, we assume the following type of solution

$$p_o(x,t,u) = \sum_{n=-\infty}^{\infty} \eta_o^{(n)}(t,u)\exp in(kx-wt) \tag{19a}$$

$$R_o(x,t,u) = \sum_{n=-\infty}^{\infty} \zeta_o^{(n)}(t,u)\exp in(kx-wt) \tag{19b}$$

$$E_o(x,t) = \sum_{n=-\infty}^{\infty} \xi_o^{(n)}(t)\exp in(kx-wt) \tag{19c}$$

$$P_o(x,t) = \sum_{n=-\infty}^{\infty} \tau_o^{(n)}(t)\exp in(kx-wt) \tag{19d}$$

Two further remarks concerned with the solutions (19a)-(19d) are in order. Firstly, the non-linear nature of the interaction between matter and light inevitably changes the amplitude of the oscillation as time increases.

Thus by investigating the physical factors contributing to the time wise variation of the electric field and the dipole moment, one could adequately control the maser oscillation by proper adjustment of those physical parameters.

Secondly, the harmonic solutions (19a)-(19d) contain both right and left running wave. For example, if $\eta_o^{(n)}(t,u) = A(t,u)\exp 2inwt$, where A(t) is non-periodic function of time, then p_o has a non vanishing left running wave at the nth harmonic. The detailed investigation of these two points will be presented in section IV.

As already mentioned we shall employ matrix formalism in the ensuing analysis.

We will write, for example, the molecular dipole moment in the following matrix form.

$$p_o(x,t,u) = (\exp N\psi)\eta_o(t,u), \tag{20a}$$

where

$$\psi = i(kx-wt) \tag{20b}$$

p_o is an column vector $(p_o^{(n)}, p_o^{(n-1)}, p_o^{(1)}, p_o^{(-1)}, p_o^{(-n)}$, and N is a diagonal matrix defined as

$$N = \begin{vmatrix} n & 0 & . & . & . & . & & . \\ 0 & n-1 & . & . & . & . & & . \\ . & 0 & . & . & . & . & & . \\ . & . & . & 1 & 0 & 0 & 0 & . \\ . & . & . & 0 & 0 & 0 & 0 & . \\ . & . & . & 0 & 0 & -1 & 0 & 0 \\ & & & & & & -(n-1) & \\ 0 & 0 & & 0 & 0 & 0 & 0 & -n \end{vmatrix} \tag{20c}$$

On substituting Eq. (20a) and other similar expressions for E_o and R_o into Eqs. (17) and (18) we obtain

$$I\ddot{\eta}_o + 2\varphi N\dot{\eta}_o + (\varphi^2 N^2 + w_{ab}^2 I)\eta_o = \gamma\zeta_o^{(o)}\xi_o^{(o)} + \gamma\,\mathcal{E}_o\zeta_o \tag{21a}$$

$$I\dot{\zeta}_o + \varphi N\zeta_o = \beta(\mathcal{E}_o\eta_o + \varphi\mathcal{E}_o N\eta_o) \tag{21b}$$

where $\varphi = i(ku-w)$, I is an unit matrix, dot ( ) denotes time differentiation.

In general $\mathcal{E}_o$ is a (2n+1)x(2n+1), non-diagonal matrix. The appearance of these off-diagonal elements, evidently implying non-linear interaction, is thus responsible for the higher harmonic generation.

In the present analysis we shall limit out calculation to the first seven harmonics, namely the d.c. component and harmonics at $\pm w$, $\pm 2w$, $\pm 3w$. Under such circumstances, $\mathcal{E}_o(t)$ reads

$$\mathcal{E}_o(t) = \begin{vmatrix} \xi_o^{(o)} & \xi_o^{(1)} & \xi_o^{(2)} & \xi_o^{(3)} & 0 & 0 & 0 \\ \xi_o^{(-1)} & \xi_o^{(o)} & \xi_o^{(1)} & \xi_o^{(2)} & \xi_o^{(3)} & 0 & 0 \\ \xi_o^{(-2)} & \xi_o^{(-1)} & \xi_o^{(o)} & \xi_o^{(1)} & \xi_o^{(2)} & \xi_o^{(3)} & 0 \\ \xi_o^{(-3)} & \xi_o^{(-2)} & \xi_o^{(-1)} & \xi_o^{(o)} & \xi_o^{(1)} & \xi_o^{(2)} & \xi_o^{(3)} \\ 0 & \xi_o^{(-3)} & \xi_o^{(-2)} & \xi_o^{(-1)} & \xi_o^{(o)} & \xi_o^{(1)} & \xi_o^{(2)} \\ 0 & 0 & \xi_o^{(-3)} & \xi_o^{(-2)} & \xi_o^{(-1)} & \xi_o^{(o)} & \xi_o^{(1)} \\ 0 & 0 & 0 & \xi_o^{(-3)} & \xi_o^{(-2)} & \xi_o^{(-1)} & \xi_o^{(o)} \end{vmatrix} \tag{21c}$$

The matrix $\mathcal{E}_o(t)$, for the case of more harmonics than presently considered, can be readily constructed.

Equations (21a) and (21b) are coupled non-linear equations. This coupling is resolved under the circumstances prevailing in the practical maser amplifier, that is the case of $\beta \ll 1$. This corresponds to slow changes in the probability matrix ζ_o, which we shall assume in the following analysis.

We write

$$p_o(x,t,u) = \sum_{\ell=0}^{\infty} \beta^{\ell} (\exp \psi N) \eta_{o,\ell}(t,u) \tag{22a}$$

$$R_o(x,t,u) = \sum_{\ell=0}^{\infty} \beta^{\ell}(\exp\psi N)\zeta_{o,\ell}(t,u) \tag{22b}$$

$$E_o(x,t) = \sum_{\ell=0}^{\infty} \beta^{\ell}(\exp\psi N)\xi_{o,\ell}(t,u) \tag{22c}$$

$$P_o(x,t) = \sum_{\ell=0}^{\infty} \beta^{\ell}(\exp\psi N)\tau_{o,\ell}(t,u) \tag{22d}$$

Substituting Eqs. (22a)-(22c) into (21a) and (21b) we obtain for the ℓ-th approximation

$$\ddot{\eta}_{o,\ell} + 2\varphi N\dot{\eta}_{o,\ell} + (\varphi^2 N^2 + w_{ab}^2 I)\eta_{o,\ell} = \gamma\zeta_o^{(o)}\xi_{o,\ell} + \gamma\sum_{\ell'}^{\ell-1} \mathcal{E}_{o,\ell'}\zeta_{o,\ell-\ell'} \tag{23a}$$

$$\dot{\zeta}_{o,\ell} + \varphi N\zeta_{o,\ell} = \sum_{\ell'}^{\ell-1}\left\{\mathcal{E}_{o,\ell'}\left(\dot{\eta}_{o,\ell-\ell'-1} + \varphi N\eta_{o,\ell-\ell'-1}\right)\right\} \tag{23b}$$

$\mathcal{E}_{o,\ell'}$ is the matrix obtained by replacing $\xi_o^{(j)}$ in Eq. (21c) by $\xi_{o,\ell}^{(j)}$. Since the right hand side of Eq. (23b) contains terms of the lower order perturbative quantities, the particular solution is formally given as

$$\zeta_{o,\ell}(t,u) = \sum_{\ell'}^{\ell-1} \int^{t} \exp[-\varphi N(t-t')]\left\{\mathcal{E}_{o,\ell'}(t')\left[\dot{\eta}_{o,\ell-\ell'-1}(t',u) + \right.\right.$$

$$\left.\left. + \varphi N\eta_{o,\ell-\ell'-1}(t',u)\right]\right\}dt' \tag{24a}$$

The amplitude of the dipole moment is calculated from (23a) in terms of the electric intensity matrix $\xi_{o,\ell}$,

$$\eta_{o,\ell}(t,u) = \int^{t} L(t-t',u)\Big\{\gamma\zeta_o^{(o)}\xi_{o,\ell}(t') +$$

$$+ \gamma\sum_{\ell'=0}^{\ell-1} \boldsymbol{\xi}_{o,\ell'}(t')\zeta_{o,\ell-\ell'}(t',u)\Big\}dt' \qquad (24b)$$

where $L(t-t',u)$ is a diagonal matrix given by

$$L(t-t',u) = \frac{i}{2\omega_{ab}}\Big\{\exp(\varphi N - i\omega_{ab}I)(t-t') - \exp(\varphi N + i\omega_{ab}I)(t-t')\Big\} \qquad (24c)$$

The electric intensity, which appears in the integrand of Eq. (24b), will be calculated from the Maxwell equation (15). To do so, we first calculate the macroscopic polarization matrix by multiplying Eq. (24b) by the distribution function $f_o(u)$ and then integrating with respect to u.

$$P_{o,\ell}(x,t) = (\exp\psi N)\gamma\zeta_o^{(o)} \iint_{-\infty}^{t\infty} f_o(u)L(t-t',u)\xi_{o,\ell}(t')dudt' +$$

$$+ (\exp\psi N)q_{o,\ell}(t) \qquad (25a)$$

where

$$q_{o,\ell}(t) = \gamma\sum_{\ell'}\iint_{-\infty}^{t\infty} f_o(u)L(t-t',u)\boldsymbol{\xi}_{o,\ell'}(t')\zeta_{o,\ell-\ell'}(t',u)dudt' \qquad (25b)$$

Substituting $E_{o,\ell}(x,t)$ and $P_{o,\ell}(x,t)$ in Maxwell's equation (15), the following integro-differential equation for $\xi_{o,\ell}(t)$ is obtained.

$$\ddot{\xi}_{o,\ell}(t) + \frac{1}{\mu_o\epsilon_o}(\mu_o\sigma I - 2i\mu_o\epsilon_o\omega N)\dot{\xi}_{o,\ell}(t) + \frac{1}{\mu_o\epsilon_o}[(k^2-\omega^2)N^2 - i\mu_o\sigma\omega N +$$

$$+ \mu_o H(o)]\xi_{o,\ell}(t) = \int^{t} K(t-t')\xi_{o,\ell}(t')dt' + \Pi_{o,\ell}(t) \qquad (26a)$$

where the kernel $K(t-t')$, $\Pi_{o,\ell}(t)$, and $H(o)$ are given by

$$K(t-t') = \frac{-i\gamma\zeta_o^{(o)}}{2\epsilon_o\omega_{ab}}\int_{-\infty}^{\infty}\Big\{[(\varphi N-i\omega_{ab}I)^2-2i\omega N(\varphi N-i\omega_{ab}I)+\omega^2N^2]\exp(\varphi N-i\omega_{ab}I)(t-t')$$

$$-[(\varphi N+i\omega_{ab}I)^2-2i\omega N(\varphi N+i\omega_{ab}I)+\omega^2N]\exp(\varphi N+i\omega_{ab}I)(t-t')\Big\}f_o(u)du \qquad (26b)$$

$$\Pi_{o,\ell}(t) = -\frac{1}{\epsilon_o}[q_{o,\ell}(t)-2i\omega Nq_{o,\ell}(t)-\omega^2N^2q_{o,\ell}(t)] \qquad (26c)$$

$$H(o) = \lim_{t'\to t}\gamma\zeta_o^{(o)}\int_{-\infty}^{\infty}f_o(u)\frac{\partial L}{\partial t}(t-t',u)du = \gamma\zeta_o^{(o)}\int_{-\infty}^{\infty}f_o(u)Idu \qquad (26d)$$

The integral expression that appears in the right hand side of Eq. (26a) is a convolution of K and $\xi_{o,\ell}$. Hence the method of Laplace transform appears to be most convenient.

Let

$$\xi_{o,\ell}(s) = \int_o^{\infty}\xi_{o,\ell}(t)e^{-st}dt \qquad (27a)$$

The application of the Laplace transform to Eq. (26a) gives

$$\xi_{o,\ell}(s) = \Big\{s^2I+\frac{1}{\mu_o\epsilon_o}[(\mu_o\sigma I-2i\mu_o\epsilon_o\omega N)s + (k^2-\omega^2)N^2-i\mu_o\sigma\omega N +$$

$$+\mu_oH(o)-K(s)\Big\}^{-1}x\Big\{\Pi_{o,\ell}(s)+\xi_{o,\ell}(o)+\Big[\frac{1}{\mu_o\epsilon_o}(\mu_o\sigma I-2i\mu_o\epsilon_o\omega N)+sI\Big]\xi_{o,\ell}(o)\Big\}$$

(27b)

The inverse transform of (27b) yields the amplitude of the oscillation of the electric field vector

$$\xi_{o,\ell}(t) = \frac{1}{2\pi i}\int_c e^{st}[\mathcal{L}\xi_{o,\ell}(s)]ds \qquad (27c)$$

The dipole moments $\eta_{o,\ell}$, and macroscopic polarization are subsequently calculated from Eqs. (24b) and (25a).

(B) Effect of molecular collisions:

The molecules inter collide during their motion, causing the distribution function, macroscopic polarization and the electric field intensity to change accordingly. Assuming the collision frequency is independent of fluid properties, the first order approximation for the distribution function at steady state is governed by the following equation

$$u\frac{\partial f_1^+}{\partial x} = F_o(u) - f_o(u) \tag{28}$$

where F_o is the Maxwellian distribution function defined by the zeroth approximation. f_1^+ is given by single quadrature as

$$f_1^+(x,u) = \int^x \frac{1}{u}\Big(F_o(u)-f_o(u)\Big)dx = \frac{x}{u}\Big(F_o(u)-f_o(u)\Big) \tag{29}$$

The first order perturbation equations are

$$-\frac{\partial^2 E_1^+}{\partial x^2} + \mu_o\sigma\frac{\partial E^+}{\partial t} + \mu_o\epsilon_o\frac{\partial^2 E_1^+}{\partial t^2} = -\mu_o\frac{\partial^2 P_1^+}{\partial t^2} \tag{30a}$$

$$P_1^+(x,t) = \int_{-\infty}^{\infty} [p_1^+(x,u,t)f_o(u) + p_o(x,u,t)f_1^+(u,x)]du \tag{30b}$$

$$\left(\frac{\partial}{\partial t} + u\frac{\partial}{\partial x}\right)^2 p_1^+ + w_{ab}^2 p_1^+ = \gamma(E_o R_1^+ + E_1^+ R_o) \tag{30c}$$

$$\frac{\partial R_1}{\partial t} + u\frac{\partial R_1}{\partial x} = \beta\left\{E_o\left(\frac{\partial p_1^+}{\partial t} + u\frac{\partial p_1^+}{\partial x}\right) + E_1^+\left(\frac{\partial p_o}{\partial t} + u\frac{\partial p_o}{\partial x}\right)\right\} \tag{30d}$$

Assume a harmonic solution in matrix form.

$$P_1^+(x,t,u) = \sum_{\ell} \beta^{\ell} (\exp\psi N)\left\{x\tilde{\eta}_{1,\ell}(t,u) + \tilde{\tilde{\eta}}_{1,\ell}(t,u)\right\} \tag{31a}$$

$$R_1^+(x,t,u) = \sum_{\ell} \beta^{\ell} (\exp\psi N)\left\{x\tilde{\zeta}_{1,\ell}(t,u) + \tilde{\tilde{\zeta}}_{1,\ell}(t,u)\right\} \tag{31b}$$

$$E_1^+(x,t) = \sum_{\ell} \beta^{\ell} (\exp\psi N)\left\{x\tilde{\xi}_{1,\ell}(t) + \tilde{\tilde{\xi}}_{1,\ell}(t)\right\} \tag{31c}$$

$$P_1^+(x,t) = \sum_{\ell} \beta^{\ell} (\exp\psi N)\left\{x\tilde{\tau}_{1,\ell}(t) + \tilde{\tilde{\tau}}_{1,\ell}(t)\right\} \tag{31d}$$

After substituting Eqs. (31a), (31b) into (30c) and (30d) and equating the sum of the terms containing x and x^o to be zero respectively the following equations result

$$\ddot{\eta}(t,u) + 2\varphi N\dot{\eta}(t,u) + (\varphi^2 N^2 + w_{ab}^2 I)\eta(t,u) = \gamma\zeta_o^{(o)}\xi(t) + J(t,u) \tag{32a}$$

$$\dot{\zeta}(t,u) + \varphi N\zeta(t,u) = S(t,u) \tag{32b}$$

Eqs. (32a) and (32b) are applicable for the calculation of $\tilde{\eta}_{1,\ell}$, $\tilde{\tilde{\eta}}_{1,\ell}$, and $\tilde{\zeta}_{1,\ell}$, $\tilde{\tilde{\zeta}}_{2,\ell}$ respectively. The proper inhomogeneous term J in the calculation of $\tilde{\eta}_{1,\ell}$ and $\tilde{\tilde{\eta}}_{1,\ell}$, and s for $\tilde{\zeta}_{1,\ell}$ and $\tilde{\tilde{\zeta}}_{2,\ell}$ is given in appendix A. The solution for $\tilde{\eta}_{1,\ell}$, $\tilde{\tilde{\eta}}_{1,\ell}$, $\tilde{\zeta}_{1,\ell}$ and $\tilde{\tilde{\zeta}}_{1,\ell}$ are also given in the appendix A.

The total macroscopic polarization within this approximation is calculated to be (Appendix A)

$$P_{1,\ell}^+(x,t) = \exp\psi N\int_0^t \int_{-\infty}^{\infty} f_o(u)L(t-t',u)\left\{\gamma\zeta_o^{(o)}\left(x\tilde{\xi}_{1,\ell}(t') + \tilde{\tilde{\xi}}_{1,\ell}(t')\right) + \right.$$

$$\left. + \frac{x}{u}\left(F_o(u) - f_o(u)\right)\eta_{o,\ell}(t',u) - 2u\varphi N\tilde{\eta}_{1,\ell}(t',u)\right\}du\,dt' + (\exp\psi N)(x\tilde{q}_{1,\ell} + \tilde{\tilde{q}}_{1,\ell}) \tag{33}$$

By substituting $P^{+}_{1,\ell}(x,t)$ into Maxwell's equation, the following integro-differential equation yields

$$\ddot{\xi}(t) + \frac{1}{\mu_o\epsilon_o}(\mu_o\sigma I - 2i\mu_o\epsilon_o\omega N)\dot{\xi} + \frac{1}{\mu_o\epsilon_o}\left\{(k^2-\omega^2)N^2 - i\mu_o\sigma\omega N + \mu_o H(o)\right\}\xi$$

$$= \int_o^t K(t-t';u)\xi(t')dt' + \Pi + W \qquad (34)$$

This is the same type of equation previously obtained in Eq. (26a) except a minor change in the inhomogeneous term. The inhomogeneous term Π appearing in q. (34) and (26a) is of the quantum electric nature, whereas W is caused by the change in the distribution due to the intermolecular collision.

Eq. (34) is used in the calculation of $\tilde{\xi}_{1,\ell}$ and $\tilde{\tilde{\xi}}_{1,\ell}$. The corresponding inhomogeneous terms are given in appendix A.

The integro-differential equation (34) is again solved by the Laplace transform as was previously done.

The dipole moment $\tilde{\eta}_{1,\ell}$, $\tilde{\tilde{\eta}}_{1,\ell}$, and macroscopic polarization $\tilde{\tau}_{1,\ell}$ and $\tilde{\tilde{\tau}}_{1,\ell}$ are calculated from eqs. (33d), (33f) and (34a) in appendix A.

(C) Effect of molecular acceleration:

The dipole acceleration create the direct interaction between radiation and the molecules at kinetic level.

The system of equations are

$$\frac{\partial f_1}{\partial t} + u\frac{\partial f_1}{\partial x} = -\frac{1}{m}\frac{\partial v_o}{\partial x}\frac{\partial f_o}{\partial u} \qquad (35a)$$

where $v_o(x,t)$ is given by

$$v_o(x,t) = \sum_n\sum_{n'}\sum_{\ell}\beta^{\ell}(\exp n\psi)\left(\xi^{(n')}_{o,\ell'}\ \eta^{(n-n')}_{o,\ell-\ell'}\right) \qquad (35b)$$

$$-\frac{\partial^2 E_1}{\partial x^2} + \mu_o \sigma \frac{\partial E_1}{\partial t} + \mu_o \epsilon_o \frac{\partial^2 E_1}{\partial t^2} = -\mu_o \frac{\partial^2 P_1}{\partial t^2} \tag{35c}$$

$$P_1(x,t) = \int_{-\infty}^{\infty} \left\{ p_1(x,u,t) f_o(u) + p_o(x,u,t) f_1(x,u,t) \right\} du \tag{35d}$$

$$\left(\frac{\partial}{\partial t} + u\frac{\partial}{\partial x}\right)^2 p_1 + w_{ab}^2 p_1 = \gamma(E_o R_1 + E_1 R_o) \tag{35e}$$

$$\frac{\partial R_1}{\partial t} + u\frac{\partial R_1}{\partial x} = \beta\left\{ E_o\left(\frac{\partial p_1}{\partial t} + u\frac{\partial p_1}{\partial x}\right) + E_1\left(\frac{\partial p_o}{\partial t} + u\frac{\partial p_o}{\partial x}\right)\right\} \tag{35f}$$

The following harmonic solutions are assumed,

$$f_1(x,t,u) = \sum_{\ell} \beta^{\ell} (\exp\psi N) g_{1,\ell}(t,u) \tag{36a}$$

$$p_1(x,t,u) = \sum_{\ell} \beta^{\ell} (\exp\psi N) \eta_{1,\ell}(t,u) \tag{36b}$$

$$R_1(x,t,u) = \sum_{\ell} \beta^{\ell} (\exp\psi N) \zeta_{1,\ell}(t,u) \tag{36c}$$

$$E_1(x,t) = \sum_{\ell} \beta^{\ell} (\exp\psi N) \xi_{1,\ell}(t) \tag{36d}$$

$$P_1(x,t) = \sum_{\ell} \beta^{\ell} (\exp\psi N) \tau_{1,\ell}(t) \tag{36e}$$

Substituting (36a) into (35a) results in the following equation

$$\dot{g}_{1,\ell}(t,u) + \varphi N g_{1,\ell}(t,u) = -\frac{ik}{m}\frac{\partial f_o}{\partial u} \sum_{\ell'=0}^{\ell} \mathcal{E}_{o,\ell'}(t) \eta_{o,\ell-\ell'}(t,u) \tag{37}$$

$g_{1,\ell}(t,u)$ is calculated to be

$$g_{1,\ell}(t,u) = -\frac{ik}{m}\frac{\partial f_o}{\partial u}\sum_{\ell'=0}^{\ell}\int_o^t \left\{\exp\left(-\varphi N(t-t')\right)\right\} N\,\mathcal{E}_{o,\ell'}(t')\eta_{o,\ell-\ell'}(t',u)dt' \tag{38}$$

$\eta_{1,\ell}(t,u)$, and $\zeta_{1,\ell}(t,u)$ are governed by the equations similar to (32a) and (32b) respectively, except that the proper inhomogeneous functions J and S are substituted in those two equations.

The macroscopic polarization is affected by the changes in the dipole moment and the distribution function.

$$P_{1,\ell}(x,t) = (\exp\psi N)\int_o^t\int_{-\infty}^{\infty} f_o(u)L(t-t';u)\gamma\zeta_o^{(o)}\xi_{1,\ell}(t')dudt' +$$

$$+ (\exp\psi N)q_{1,\ell}(t) + (\exp\psi N)\sum_{\ell'}^{\ell}\int_{-\infty}^{\infty}\mathcal{F}_{1,\ell'}(t,u)\eta_{o,\ell-\ell'}(t,u)du \tag{39}$$

$\mathcal{F}_{1,\ell}(t,u)$ is the matrix given as that of $\mathcal{E}_o(t)$, except that each element $\xi_o^{(j)}$ that appears in Eq. (21c) is replaced by $f_{1,\ell}^{(j)}$ (t,u). With proper modification of the inhomogeneous terms in Eqs. (32a), (32b), (33) and (34), the amplitude $\xi_{1,\ell}(t)$, the dipole moment, and the macroscopic polarization can be similarly calculated.

The system of the perturbed equations preserve a property that (-n)th harmonic of any physical quantity is the complex conjugate of nth harmonic, provided this relation is satisfied initially.

To prove the property mentioned above, we shall use the mathematical induction.

Preparatory remarks about notation are due. In Eqs. (19a)-(19d), the superscripts such as $\eta_o^{(n)}$, $\xi_o^{(-n)}$ designate the nth and -nth harmonic. The appearance of negative indices is awkward if not prohibited. Hence for this section only, we shall use the index inside the bracket such as

$$\eta_{o,\ell}^{(n)} \to \eta_{o,\ell'}^{(m)}, \qquad m=1,2\ldots 7 \qquad (40)$$

The index in the bracket designates the component in the order of decreasing harmonics starting from the highest harmonic; i.e.
$\eta_{o,\ell}^{(3)} = \eta_{o,\ell'}^{(1)},\ \eta_{o,\ell'}^{(2)} = \eta_{o,\ell'}^{(2)},\ \eta_{o,\ell}^{(1)} = \eta_{o,\ell}^{(3)} \ldots$
$\eta_{o,\ell}^{(-3)} = \eta_{o,\ell}^{(7)}$.

If the (-n)th harmonic of $\eta_{o,\ell'}$ is the complex conjugate of nth harmonic of $\eta_{o,\ell'}$, then we have

$$\eta_{o,\ell'}(n) = \eta^{*}_{o,\ell'}(8-n) \qquad n=1,2,\ldots 7 \qquad (41)$$

The n mth element of the matrix $\mathcal{E}_{o,\ell}$ Eq. (21c) is similarly written as

$$\mathcal{E}_{o,\ell}(n\ m)\ldots \qquad \begin{matrix} n=1,2\ldots 7 \\ m=1,2\ldots 7 \end{matrix} \qquad (42)$$

The first index represents the raw, and the second index the column. Inspection of the matrix $\mathcal{E}_{o}(t)$ reveals that if $\xi_{o,\ell'}(-n) = \xi_{o,\ell'}(n)^{*}$ for $\ell' < \ell$ then

$$\mathcal{E}_{o,\ell'}(n\ m) = \mathcal{E}^{*}_{o,\ell'}(8-n\ 8-m) \qquad (43)$$

We shall first prove that $\zeta_{o,\ell}(-n) = \zeta_{o,\ell}(n)^{*}$ if $\xi_{o,\ell'}(-n) = \xi_{o,\ell'}(n)^{*}$ and $\eta_{o,\ell'}(-n) = \eta_{o,\ell'}(n)^{*}$ for $\ell' < \ell$.

From Eq. (24a) we have

$$\zeta_{o,\ell}(-n) = \sum_{\ell'}^{\ell-1} \int^{t} \exp[+\omega n(t-t')]\Big\{\mathcal{E}_{o,\ell'}(8-n,m)\dot{\eta}_{o,\ell-\ell'-1}(m) -$$

$$- \omega n\mathcal{E}_{o,\ell'}(8-n,m)\eta_{o,\ell-\ell'-1}(m)\Big\}dt' = \sum_{\ell'}^{\ell-1} \int^{t} \Big\{\exp^{*}[-\omega n(t-t')]\Big\}\Big\{\mathcal{E}^{*}_{o,\ell'}(n, 8-m)\dot{\eta}^{*}_{o,\ell-\ell'-1}(8-m) +$$

$$+ (\omega n)^{*}\mathcal{E}^{*}_{o,\ell'}(n, 8-m)\eta^{*}_{o,\ell-\ell'-1}(8-m)\Big\}dt'$$

by virtue of (41) and (43). Since 8-m appears as a dummy index, we replace it by m' to have

$$\zeta_{o,\ell}(-n) = \sum_{\ell'}^{\ell-1} \int^{t} \left\{\exp[-\varphi n(t-t')]^{*}\right\}\left\{\boldsymbol{\xi}^{*}_{o,\ell'}(n,m')\eta^{*}_{o,\ell-\ell'-1}(m') + \right.$$

$$\left. + (\varphi n)^{*}\boldsymbol{\xi}^{*}_{o,\ell'}(nm')\eta^{*}_{o,\ell-\ell'-1}(m')\right\}dt' = \zeta_{o,\ell}(n)^{*} \quad (44)$$

The proof for $\xi_{o,\ell}(-n) = \xi_{o,\ell}(n)^{*}$, is accomplished by proving that $\mathcal{L}\xi_{o,\ell}(-n) = \mathcal{L}\xi^{*}_{o,\ell}(n)$. We first prove the following relations

$$L(nn) = L^{*}(8\text{-}n, 8\text{-}n) \quad (45)$$

$$K(nn) = K^{*}(8\text{-}n, 8\text{-}n) \quad (46)$$

$$q_{o,\ell}(-n) = q_{o,\ell}(8\text{-}n) = \gamma\sum_{\ell'}\iint_{-\infty}^{t\infty} f_o(u)L(8\text{-}n,8\text{-}n)\boldsymbol{\xi}_{o,\ell'}(8\text{-}n,m)\zeta_{o,\ell-\ell'}(m)du\,dt$$

$$= \gamma\sum_{\ell'}\iint_{-\infty}^{t\infty} f_o(u)L^{*}(nn)\boldsymbol{\xi}^{*}_{o,\ell'}(n\ 8\text{-}m)\zeta^{*}_{o,\ell-\ell'}(8\text{-}m)du\,dt = q^{(n)*}_{o,\ell} \quad (47a)$$

$$\Pi_{o,\ell'}(-n) = \Pi_{o,\ell}(8\text{-}n) = -\frac{1}{\epsilon_o}[q_{o,\ell'}(8\text{-}n) + 2i\omega n\ q_{o,\ell'}(8\text{-}n) +$$

$$- \omega^2 n^2 q_{o,\ell'}(8\text{-}n) = \Pi^{*}_{o,\ell}(n) \quad (47b)$$

From the above four relations together with the initial conditions

$$\xi_{o,\ell}(-n, t=0) = \xi^{*}_{o,\ell}(n, t=0) \quad (48a)$$

$$\dot{\xi}_{o,\ell}(-n, t=0) = \dot{\xi}^{*}_{o,\ell}(n, t=0) \quad (48b)$$

it is immediately apparent from Eq. (27b) that $\xi_{o,\ell}(-n) = \xi^{*}_{o,\ell}(n)$. The proof of $\eta_{o,\ell}(-n) = \eta_{o,\ell}(n)^{*}$, $\tau_{o,\ell}(-n) = \tau_{o,\ell}(n)^{*}$ are also easily made by similar procedure.

When $\ell = 0$ $\eta_{o,o}^{(-n)} = \eta_{o,o}^{(n)*}$, $\xi_{o,o}^{(-n)} = \xi_{o,o}^{(n)*}$, $\zeta_{o,o}^{(-n)} = \zeta_{o,o}^{(n)*} = 0$,

except for $\zeta_{o,o}(o)$ = const, hence by mathematical induction we obtain the required property for all ℓ.

The cases of the higher order perturbation can be proven similarly and hence will not be carried out further.

IV SELF-EXCITATION, STIMULATED EMISSION AND NON-LINEAR SUSCEPTIBILITY

To illustrate the method developed, we consider a linearly polarized wave $E^{(1)}_{o,o}(1) = \xi_{o,o} \exp i(kx-wt) + \xi^*_{o,o} \exp -i(kx-wt)$, at near resonant frequency, incident on a molecular beam whose distribution function is Maxwellian.

The amplitude $\xi_{o,o}$, and its time derivative $\dot{\xi}_{o,o}$ are initially prescribed. As is pointed out previously, the amplitude of the oscillation changes as a result of matter-light interaction.

From the point of view of energy conservation, the emitted radiant energy must be greater than the energy loss which arises as a result of finite conductivity, or often referred to as cavity loss.

To provide quantitative criterion, we define the maser self-excitation to be the oscillation in which the amplitude of electric intensity dipole moment and macroscopic polarization do not decay as time increases.

It is often advantageous to investigate the index of refraction (n) of a maser beam. The curves of n^2-1 versus frequency is referred to as dispersion curves. In the quantum theory the negative dispersion which was unheard of in classical theory could take place. The negative dispersion arises as a result of transition of a molecule from a higher energy level to a lower one, i.e. stimulated emission. In the later part of this section the curves of dispersion, and the negative dispersion are studied at various kinetic parameters.

We also calculate the non-linear susceptibility to the lowest order.

The calculation is straight forward, nevertheless, it is extremely labourous. Hence our illustrative calculation is limited within the rarefied gas assumption.

We first write the response of the dipole moment and the macroscopic polarization at the same frequency as the incident electric field.

From Eqs. (24b) and (24c) we obtain

$$\eta^{(1)}_{o,o}(t,u) = \frac{i\gamma\zeta_o(o)}{2w_{ab}} \int_o^t e^{iku(t-t')}[e^{-i(w+w_{ab})(t-t')} - e^{-i(w-w_{ab})(t-t')}]\xi^{(1)}_{o,o}(t')dt' \tag{49a}$$

$$\tau_{o,o}(1)(t,u) = \frac{i\gamma\zeta_o(o)}{2w_{ab}} \int_o^t\int_{-\infty}^{\infty} Ae^{-bu^2+iku(t-t')}[e^{-i(w+w_{ab})(t-t')} - e^{-i(w-w_{ab})(t-t')}]\xi_{o,o}(1)(t')dudt' \tag{49b}$$

where $f_o(u)$ is replaced by the Maxwellian distribution $Ae^{-b(u-u)^2}$. U is the mean velocity of the molecular beam.

The Maxwell's integro-differential equation is now given explicitly by

$$\ddot{\xi}^{(1)}_{o,o}(t) + (\sigma/\epsilon_o - 2iw)\dot{\xi}^{(1)}_{o,o}(t) + [\Omega^2 - w^2 + \gamma\zeta_o(o)\sqrt{\pi}\, A/b\epsilon_o + - i\sigma w/\epsilon_o]\xi^{(1)}_{o,o}(t) = \int_o^t K(t-t')\xi^{(1)}_{o,o}(t')dt' \tag{50a}$$

where $\Omega = k^2/\mu_o\epsilon_o$, w is in general a number close to Ω, i.e. $\Omega \simeq w$.

Kernel $K(t-t')$ is calculated to be

$$K(t-t') = \frac{-i\gamma\zeta_o^{(o)}\sqrt{\pi}A}{2\epsilon_o b w_{ab}}\left\{\left[\frac{k^4}{4b^2}(t-t')^2 - i\frac{k^2}{b}(\theta_1 - w)(t-t') - (\theta - w)^2 - \frac{k^2}{2b}\right]e^{i\theta_1(t-t')}\right.$$

$$\left. - \left[\frac{k^4}{4b^2}(t-t')^2 - i\frac{k^2}{b}(\theta_2 - w)(t-t') - (\theta_2 - w)^2 - \frac{k^2}{2b}\right]e^{i\theta_2(t-t')}\right\}e^{-\frac{k^2}{4b}(t-t')^2} \tag{50b}$$

with

$$\theta_1 = kU - (w+w_{ab}) \tag{50c}$$

$$\theta_2 = kU - (w-w_{ab}) \tag{50d}$$

The solution of (50a) is given, as was shown in (21c), by

$$\xi_{o,o}(t) = \frac{1}{2\pi i}(P)\int_{c-i\infty}^{c+i\infty} \frac{[\xi_{o,o}^{(1)}(o) + (\sigma/\epsilon_o - 2iw + s)\xi_{o,o}^{(1)}(o)]e^{st}ds}{s^2 + (\sigma/\epsilon_o - 2iw)s + (\Omega^2 - w^2 - i\sigma w/\epsilon_o + \gamma\zeta_o(o)\sqrt{\pi}\,A/b\epsilon_o) - \tilde{K}(s)} \tag{50e}$$

The exact evaluation of the inverse transform (50e) is extremely complicated by the appearance of $\tilde{K}(s)$. Nevertheless, the integral shall be evaluated based on the following reasoning.

When both conductivity and the polarization parameter are negligibly small, Maxwell's wave equation admit two solutions, undistorted right running and left running waves. The existence of two wave solutions in Eq. (50e) are readily demonstrated by equating $\sigma, \zeta_o(o)$ and $\tilde{K}(s)$ to be zero. The integrand, with $\sigma, \zeta_o(\sigma)$, $K(s)$ all zero, has two simple poles, one located at $s = 0$, giving rise to a right running wave, the other at $s = 2iw$, a left running wave.

For a finite, but small conductivity and polarization parameter, the amplitudes of the right, and left running waves changes.

In order to be able to examine the time wise variation of amplitude, which would give us the quantitative criterion of the self-sustained oscillation, we shall assume both the conductivity and polarization parameter to be of the same order small quantity. This assumption permits us to assume the following type of series solution in order to find the location of two poles.

$$s = s_o + s' + \ldots \tag{51}$$

Substitute (51) in the algebraic equation appearing in the denominator of the integrand of Eq. (50e), equating the terms of zeroth, and first order to be zero yields

$$s_o^2 - 2iws_o + \Omega^2 - w^2 = 0 \tag{52a}$$

$$2(s_o - iw)s' = \widetilde{K}(s_o) - \gamma\zeta_o(o)\sqrt{\pi}\ A/b\epsilon_o - \sigma(s_o - iw)/\epsilon_o \tag{52b}$$

Hence

$$s_o(\pm) = i(w \mp \Omega) \tag{52c}$$

$$s'(\pm) = -\frac{\sigma}{2\epsilon_o} + \frac{K\left(s_o(\pm)\right) - \gamma\zeta_o(o)\sqrt{\pi}\ A/b\epsilon_o}{2\left(s_o(\pm) - iw\right)} \tag{52d}$$

Thus the denominator of the integrand of Eq. (50e) is replaced by

$$\left\{s - [s_o(+) + s'(+)]\right\}\left\{s - [s_o(-) + s'(-)]\right\} \tag{53a}$$

The electric field intensity is then calculated to be

$$E_{o,o}^{(1)}(x,t) \simeq \frac{\left[1 + i\left(\sigma/\epsilon_o + s'(t)\right)/2w\right]\xi_{o,o}^{(1)}(o) + i\dot{\xi}_{o,o}^{(1)}(o)/2w}{1 + i[s'(+) + s'(-)]/2w}\ e^{i(kx - wt) + i(w - \Omega)t + s'(+)t}$$

$$- i\frac{[\sigma/\epsilon_o + s'(-)]\xi_{o,o}^{(1)}(o) + \xi_{o,o}^{(1)}(o)}{2w\left\{1 + i[s'(+) + s'(-)]/2w\right\}}\ e^{i(kx \mp wt) + i(w + \Omega)t + s'(-)t} \tag{53b}$$

Since $w \simeq \Omega$, the first term on the right hand side of Eq. (53b) represents right running wave whereas the second term is the left running wave.

It is important to point out here that the order of magnitude of the right running wave is

$$\xi^{(1)}_{o,o}(o) \ \exp\mathrm{Re}\ s'(+) \tag{53c}$$

and that of the left running wave is

$$\frac{\sigma}{w}\xi^{(1)}_{o,o}(o) \ \exp\mathrm{Re}\ s'(-) \tag{53d}$$

Evidently, aside from the exponential factor, the amplitude of the left running wave is of the first order small quantity as is physically expected.

We now look into the time wise variation of the amplitude of the right running wave, or the condition of maser self-sustained oscillation.

For the amplitude of the electric intensity not to decay exponentially, $\mathrm{Re}s'(+)$, and $\mathrm{Re}s'(-)$ must not be negative numbers. Accordingly from Eqs. (52c), and (52d), we set the condition of self-sustained oscillation to be

$$\mathrm{Re}s'(+) = -\frac{\sigma_o}{2\epsilon} - \frac{\widetilde{K}_i\big(s_o(+)\big)}{2w} \geq 0 \quad \text{for right running wave} \tag{54a}$$

$$\mathrm{Re}s'(-) = -\frac{\sigma}{2\epsilon_o} + \frac{\widetilde{K}_i\big(s_o(-)\big)}{2\Omega} \geq 0 \quad \text{for left running wave} \tag{54b}$$

where $\widetilde{K}_i(s_o)$ is the imaginary part of $\widetilde{K}(s_o)$.

We shall investigate these two conditions by examining the approximate value of $\widetilde{K}_i\big(s_o(+)\big)$ and $\widetilde{K}_i\big(s_o(-)\big)$.

The Laplace transform of K(t-t′), Eq. (50b) is found to be

$$\tilde{K}(s) = \frac{\gamma \xi_o(o)\sqrt{\pi}\ A}{2\sqrt{2b}\ k w_{ab}\epsilon_o}\left\{e^{\frac{b(s^2-\theta_1^2)}{2k^2}}\left[e^{i\left(\frac{\theta_1 bs}{k^2}+\frac{\pi}{4}\right)}\left(a_3 D_{-3}(z_1^+)+a_2 D_{-2}(z_1^+)+a_1 D_{-1}(z_1^+)\right)\right.\right.$$

$$\left.+ e^{-i\left(\frac{\theta_1 bs}{k^2}+\frac{\pi}{4}\right)}\left(a_3 D_{-3}(z_1^-)+a_2 D_{-2}(z_1^-)+a_1 D_{-1}(z_1^-)\right)\right] - e^{\frac{b(s^2-\theta_2^2)}{2k^2}}\left[e^{i\left(\frac{\theta_2 bs}{k^2}+\frac{\pi}{4}\right)}\right.$$

$$\left.\left.\left(b_3 D_{-3}(z_2^+)+b_2 D_{-2}(z_2^+)+b_1 D_{-1}(z_2^+)\right) + e^{-i\left(\frac{\theta_2 bs}{k^2}+\frac{\pi}{4}\right)}\left(b_3 D_{-3}(z_2^-)+b_2 D_{-2}(z_2^-)+b_1 D_{-1}(z_2^-)\right)\right]\right\} \tag{55a}$$

where

$$a_3 = b_3 = k^2\Gamma(3)/2b \tag{55b}$$

$$a_2 = -i\sqrt{2}\ \Gamma(2)k(\theta_1 - w)/\sqrt{b} \qquad b_2 = -i\sqrt{2}\ \Gamma(2)k(\theta_2 - w)/\sqrt{b} \tag{55c}$$

$$a_1 = -[(\theta_1 - w)^2 + k^2/2b] \qquad b_1 = -[(\theta_2 - w)^2 + k^2/2b] \tag{55d}$$

$$z_1^{\pm} = \sqrt{2b}\ (s \pm i\theta_1)/k \qquad z_2^{\pm} = \sqrt{2b}\ (s \pm i\theta_2)/k$$

where $D_p(z)$ is the parabolic cylinder function, related with Whittaker's function as follows

$$D_p(z) = 2^{\frac{1}{4}+\frac{p}{2}} W_{\frac{1}{4}+\frac{p}{2},\ -\frac{1}{4}}\left(\frac{z^2}{2}\right) \tag{55f}$$

Now in order to calculate $\tilde{K}_i\left(s_o(+)\right) = \tilde{K}_i(o)$, the value of $\lim_{s_o \to o} D_p(z_1^{\pm})$, $\lim_{s_o \to o} D_p(z_2^{\pm})$, must be evaluated.

It is evident that

$$\lim_{s_o \to o} D_p(z_1^{\pm}) = D_p\left\{\pm i\sqrt{2b}\ [U-(w+w_{ab})/k]\right\} \tag{55g}$$

$$\lim_{s_o \to o} D_p(z_2^{\pm}) = D_p\left\{\pm i\sqrt{2b}\ [U-(w-w_{ab})/k]\right\} \tag{55h}$$

Remembering b is the factor appearing in Maxwellian distribution, $\sqrt{b}$ is the quantity proportional to the thermal speed a_s of the medium, hence the order of magnitude of the arguments of (55g) and (55h) are

$$\lim_{s_o \to o} z_1^{\pm} = \pm i\sqrt{2b}\,[u-(w+w_{ab})/k] = \mp iO(c/a_s) \qquad (56a)$$

$$\lim_{s_o \to o} z_2^{\pm} = \pm i\sqrt{2b}\,[u-(w-w_{ab})/k] = \pm iO[(U-\Delta c)/a_s] \qquad (56b)$$

In a more refined approximation, it can be shown that the argument of z_1^+ and z_1^- lies in the sector with hatched lines as shown in fig. 1a and fig. 1b respectively.

Since $O(c/a_s) \gg 1$, the asymptotic approximation for $D_p(z_1^{\pm})$, is given as follows[3]

$$\lim_{s_o \to o} D_p(z_1^{\pm}) \simeq e^{-\frac{b}{2}[U-(w+w_{ab})/k]^2} \left\{2b[U-(w+w_{ab})/k]\right\}^p \left\{1-\frac{p(p-1)}{2(z_1^{\pm})^2} + O(z_1^{\pm})^{-4} + \ldots\right\}$$

$$-\frac{\sqrt{2\pi}}{\Gamma(-p)} e^{\mp ip\pi} e^{\frac{b}{2}[U-(w+w_{ab})/k]^2} \left\{2b[U-(w+w_{ab})/k]\right\}^{-p-1} \left\{1+\frac{(p+1)(p+2)}{2(z_1^{\pm})^2} + O(z_1^{\pm})^{-4} + \ldots\right\} \qquad (57a)$$

The expression of $D_p(z_2^{\pm})$ differ from that of $D_p(z_1^{\pm})$ when the absolute magnitude of $z_2^{\pm}$ is smaller than unity, i.e. $z_2^{\pm} = (U-\Delta c)/a_s \ll 1$.

$$\lim_{s_o \to o} D_p(z_2^{\pm}) \simeq 2^{\frac{p}{2}} \sqrt{\pi} \left\{\Gamma^{-1}\left(\frac{1-p}{2}\right)\left[1-\left(p+\frac{1}{2}\right)\frac{(z_2^{\pm})^2}{2!} + \right] - \Gamma^{-1}\left(-\frac{p}{2}\right)\left[z_2^{\pm} + \left(p+\frac{1}{2}\right)\frac{(z_2^{\pm})^3}{3!} + \ldots\right]\right\} \qquad (57b)$$

3 I.S. Gradshteyn, I.M. Ryzhik, Table of Integrals Series and Product (Academic Press, New York and London 1965) pp. 1064-1066.

It is important to observe the following fact. By substituting Eqs. (57a) and (57b) into (55a), the terms associated with a_3, a_2, a_1, etc. are multiplied by the number which are of the order of

$e^{-(c/a_s)^2}(a_s/c)^3 \ll 1$, $e^{-(c/a_s)^2}(a_s/c)^2 \ll 1$, and $e^{-(c/a_s)^2}(a_s/c) \ll 1$

whereas the term associated with b_3, b_2, b_1, etc. are multiplied by the number of the order of $e^{-[(U-\Delta c)/a_s]^2} \approx 0(1)$ provided $U-\Delta c \approx 0(a_s)$.

It is relevant to point out that those first three smaller terms which appeared with a_1, a_2, a_3 are associated with $\theta_1 = kU - (w+w_{ab})$, and the later three terms with $\theta_2 = kU - (w-w_{ab})$. As we shall see later θ_1 occurs in the so called non-resonant term whereas θ_2 appear in the resonant term. We conclude that $k_i\left(s_o(+)\right)$ which appeared in Eq. (54a) primarily depends on the resonant term.

Thus neglecting non-resonant parts in (55a), we obtain

$$\tilde{K}\left(s_o(+)\right) \simeq -\frac{\gamma\zeta_o^{(o)}\pi A}{2\sqrt{b}\,w_{ab}k\epsilon_o}e^{-\frac{b}{2}[U-(w-w_{ab})/k]^2}\left\{\frac{b_3}{2}+\frac{b_2}{\sqrt{2}\,\Gamma(3/2)}+b_1+0(z_2^+)+0(z_2^-)+\ldots\right\} \tag{58}$$

Substituting Eqs. (58) into (52d) yield

$$s'(+) = -\frac{\sigma}{2\epsilon_o} + Be^{-\frac{b}{2}[U-(w-w_{ab})/k]^2} - \frac{i\sqrt{\pi}A\zeta_o(o)u_{ab}^2 w_{ab}}{2\sqrt{2}\,h\epsilon_o\sqrt{b}\,w}\left\{\frac{w_{ab}}{k}e^{-\frac{b}{2}[U-(w-w_{ab})/k]^2}-\sqrt{\frac{2}{b}}\right\} \tag{59a}$$

where
$$B = \frac{\sqrt{2\pi}\,A\zeta_o(o)u_{ab}^2 w_{ab}k}{k\epsilon_o bw} \tag{59b}$$

And for the self-sustained oscillation, i.e. $\mathrm{Re}\left(s'(+)\right) > 0$, the following equality must be satisfied

$$\frac{2N\zeta_o(o)\mu_{ab}^2 w_{ab}k}{\pi hw}\sqrt{\frac{m}{k_bT}}\,e^{-\frac{m}{4k_bT}[U-(w-w_{ab})/k]^2} \geq \sigma \tag{60}$$

where we replaced A by $N\left(\frac{m}{2\pi k_b T}\right)^{\frac{3}{2}}$ and b by $\frac{m}{2k_b T}$, respectively, k_b is the Boltzmann's constant.

Note that since $\sigma > 0$, the condition of self-sustained oscillation (60) requires that $\zeta_o(o) > 0$. This implies that more molecules should be in higher energy level than in lower one. Eq. (60) thus give the threshold active molecule's density for a sustained oscillation for a given conductivity.

The dipole moment is calculated by substituting Eq. (53b) into (49a) and multiply by $e^{i(kx-wt)}$

$$
\begin{aligned}
P^{(1)}_{o,\sigma}(x,t) \simeq \frac{i\gamma\zeta_o(o)}{2w_{ab}}\Bigg\{ &- \frac{\left(1+i\left(\sigma/\epsilon_o+s'(t)\right)/2w\right)\xi^{(1)}_{o,o}(o) + i\dot{\xi}^{(1)}_{o,o}(o)/2w}{1+i\left(s'(+)-s'(-)\right)/2w} \\
&\left[\frac{1}{i\left(ku-(w-w_{ab})\right)-s'(+)} - \frac{1}{i\left(ku-(w+w_{ab})\right)-s'(+)}\right] e^{i(kx-wt)+i(w-\Omega)t+s'(+)t} + \\
&+ i\frac{\left(\sigma/\epsilon_o+s'(-)\right)\xi^{(1)}_{o,o}(o) + i\dot{\xi}^{(1)}_{o,o}(o)}{2w\left[1+i\left(s'(+)-s'(-)\right)/2w\right]}\left[\frac{1}{i[ku-(w-w_{ab})-(w+\Omega)]-s'(-)} - \right. \\
&\left. - \frac{1}{i[ku-(w+w_{ab})-(w+\Omega)]-s'(-)}\right] e^{i(kx-wt)+i(w+\Omega)t+s'(-)t}
\end{aligned}
$$

The first two terms are the right running waves, and the second two terms are the left running waves, the ratio of the amplitude of the left running wave to that of the right running wave is $\sigma/2\epsilon_o w$, which is small compared with unity. There are two terms appearing in the right running waves. The first of which is the resonant term and the second one is the non-resonant term. Two terms in the right running waves are all non-resonant.

In calculating macroscopic polarization, we take account of the right running wave resonant term only. The other three non resonant parts are multiplied by the factor of exp - $(c/a_s)^2 << 11$, and is properly neglected.

$$P^{(1)}_{o,o}(x,t) = \frac{\gamma\zeta_o(o)AE^{(1)}_{o,o}(o)}{2w_{ab}k}\left\{\exp i \;(kx-wt)+i(w-\Omega)t+s'(+)t\right\}\int_{-\infty}^{\infty}\frac{e^{-b(u-U)^2}du}{u-\left(\frac{w-w_{ab}}{k}\right)+i\frac{s'(+)}{k}}$$

$$= \frac{\gamma\zeta_o(o)AE^{(1)}_{o,o}(x,t)}{2kw_{ab}}\; Z\left[\sqrt{b}\left(\frac{w-w_{ab}}{k} - U - i\frac{s'(+)}{k}\right)\right] \tag{61a}$$

where $Z(\tau)$ is called the plasma dispersion function whose numerical value is tabulated.[4]

The linear susceptibility $\varkappa^{(1)}_{o,o}$ is defined as

$$P^{(1)}_{o,o}(x,t) = \varkappa^{(1)}_{o,o}\; E^{(1)}_{o,o}(x,t) \tag{61b}$$

Comparing Eqs. (61a) and (61b), we obtain

$$\varkappa^{(1)}_{o,o} = \frac{\gamma\zeta_o(o)\sqrt{\pi}\;N}{2w_{ab}k}\left(\frac{m}{2\pi k_b T}\right)^{\frac{3}{2}} Z\left[\sqrt{b}\left(\frac{w-w_{ab}}{k} - U - i\frac{s'(+)}{k}\right)\right] \tag{61c}$$

$\varkappa^{(1)}_{o,o}$ may also be written in terms of complex index of refraction n

$$\varkappa^{(1)}_{o,o} = \frac{1}{4\pi}\left[(n-ik)^2-1\right] \tag{61d}$$

where n is the index of refraction, k is the extinction coefficient. The physical meaning of the real and imaginary part of the susceptibility is clear.

$$n-1 \simeq 2\pi \;\mathrm{Re}\; \varkappa^{(1)}_{o,o} \tag{61e}$$

$$k \simeq 2\pi \;\mathrm{Im}\; \varkappa^{(1)}_{o,o} \tag{61f}$$

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4 B.D. Fried and S.C. Conte, The Plasma Dispersion Function, The Hilbert Transform of The Gaussian. (Academic Press, New York and London 1961).

To show the nature of oscillation we plot the real part of $s'(+)$ versus frequency for two different numerical values of parameter B. This is shown in fig. 2 in which the calculation has been made for $U = \sqrt{2/b}$, $\sigma/2\epsilon_o = 0.1$, with $B = 0.2$, and $B = 1$. Observe that as B decreases the frequency band for sustained oscillation is reduced. This is primarily attributed to the reduction in the number density of active molecules. Note also that when w differs greatly from the characteristic frequency, the population of molecules is negligibly small. The oscillation is practically damped.

In fig. 3, we present the curves of dispersion and absorptions at the operating conditions previously described. A marked difference is observed in the nature of the absorption at $B = 1$, and 0.2. For example the medium exhibits negative absorption at all frequency for $B = 0.2$. For larger B, say $B = 1.0$, a small positive absorption is observed at $\frac{\sqrt{2}}{kU} \left| w-w_{ab} \right| > 1.0$. The peak of the positive absorption is expected to increase as the parameter B increases. The dispersion curve has a rather sharp peak for larger B compared with smaller B.

It has been proved previously that

$$\xi^{(-n)}_{m,\ell}(t) = \xi^{(n)*}_{m,\ell}(t) \tag{62a}$$

$$\tau^{(-n)}_{m,\ell}(t) = \tau^{(n)*}_{m,\ell}(t) \tag{62b}$$

provided these relations are compatible with the initial conditions. The validity of the above relations will be assumed for the sake of simplifying the ensuing analysis. The nature of the oscillation and the linear susceptibility are practically the same as those of the oscillation at frequency w_o and thus will not be reproduced here.

The fundamental harmonics at the next perturbative analysis will be presented in the subsequent section.

We first calculate $\zeta_{o,1}^{(2)}$ from Eq. (24a). The result is

$$\zeta_{o,1}^{(2)}(t,u) \cong \int e^{-2\varphi(t-t')} \xi_{o,o}^{(1)}(t')[\eta_{o,o}^{(1)}(t,u) + m\eta_{o,o}^{(1)}(t,u)]dt'$$

$$\simeq \frac{\gamma\zeta_o(o)}{4w_{ab}k} \frac{\xi_{o,o}^{(1)^2}(o)e^{2s'(+)t}}{\left\{u-[(w-w_{ab})-is'(+)]/k\right\}} \tag{63}$$

Here only the resonant part of the right running wave is taken in the calculation.

Substituting Eq. (63) into Eq. (24b), the amplitude of the dipole moment is given by

$$\eta_{o,o}^{(1)}(t,u) \simeq \frac{i\gamma\zeta_o^{(1)}}{2w_{ab}} \int^{t} \left\{e^{i[kU-(w+w_{ab})](t-t')} - e^{i[kU-(w-w_{ab})](t-t')}\right\} \xi_{o,1}^{(1)}(t')dt'$$

$$- i\frac{\gamma^2\zeta_o^{(o)}}{8kw_{ab}^2} \frac{\left(\xi_{o,o}^{(1)}\xi_{o,o}^{(1)*}\right)\xi_{o,o}^{(1)}}{\left(s'(+)+s'^{*}(+)\right)} \left\{\frac{e^{[2s'(+)+s'(+)^{*}]t}}{u-[(w-w_{ab})-is'(+)]/k} - \right.$$

$$\left. - \frac{e^{[2s'(+)+s'*(+)]t}}{u-[w-w_{ab}-i(2s'(+)+s'*(+1)]/k}\right\} \tag{64}$$

The last two terms are the near resonant part induced by the combined oscillations of the electric field and the probability of the molecules in the active state.

The macroscopic polarization is obtained by multiplying Eq. (64) by the distribution function $f_o(u)$ and then integrating the resulting equation with u to yield

$$P^{(1)}_{o,1}(x,t) \simeq \frac{i\gamma\zeta_o(o)\sqrt{\pi}A}{2w_{ab}b}\, e^{i(kx-wt)} \int^{t} \left\{ e^{-i(w+w_{ab})(t-t')} - \right.$$

$$\left. - e^{-i(w-w_{ab})(t-t')} \right\} e^{ikU(t-t') - \frac{k^2}{4b}(t-t')^2}\, \xi^{(1)}_{o,1}(t')dt'$$

$$- \frac{i\sqrt{\pi}\gamma^2\zeta_o^{(o)}}{8kw^2_{ab}} \frac{\left(\xi^{(1)}_{o,o}(t)\xi^{(-1)}_{o,o}(t)\right)\xi^{(1)}_{o,o}(t)}{s'(+) + s'^*(+)} (\Delta Z) e^{[2s'(+)+s'^*(+)]t+i(kx-wt)} \tag{65a}$$

$$\text{where } \Delta Z = Z\left[\sqrt{b}\left(\frac{w-w_{ab}}{k} - U - i\frac{s'(+)}{k}\right)\right] - Z\left[\sqrt{b}\left(\frac{w-w_{ab}}{k} - U - i\frac{2s'(+)+s'(+)^*}{k}\right)\right] \tag{65b}$$

Assuming $\xi^{(1)}_{o,1}(o) = \xi^{(1)}_{o,1}(o) = 0$, and taking the resonant part of the right running wave, the electric field is calculated from the integro-differential equation as follows:

$$E^{(1)}_{o,1}(x,t) \simeq \frac{i\sqrt{\pi}\gamma^2\zeta_o^{(o)}}{8k\epsilon w^2_{ab}} \frac{\left(\xi^{(1)}_{o,o}\xi^{(1)*}_{o,o}\right)\xi^{(1)}_{o,o}}{s'(+) + s'(+)^*} A(\Delta Z)[2s'(+)+s'^*(+)-iw]^2$$

$$\left\{ \frac{e^{[2s'(+) + s'(+)^*]t}}{[s'(+)+s'^*(+)][2s'(+)+s'(+)^*-s_o(-)-s'(-)]} + \frac{e^{s'(+)t}}{[s'(+)+s'^*(+)][s_o(-)+s'(-)-s'(+)]} \right.$$

$$\left. - \frac{e^{[s_o(-) + s'(-)]t}}{[s_o(-)+s'(-)-s'(+)][2s'(+)+s'^*(+)-s_o(-)-s'(-)]} \right\} e^{i(kx-wt)} \tag{66}$$

where ΔZ is given by (65b).

Note that there are three distinct components for $\xi^{(1)}_{o,1}(t)$. The first component increases exponentially as exp $3[\mathrm{Re}\,s'(+)]t$. The other two terms increase as exp $[\mathrm{Re}\,s'(+)]t$, and are multiplied by the factor of $(s'(+)w)^{-1}$ respectively. The first two components are right running waves whereas the third component is the left running wave.

The dipole moment and macroscopic polarization are calculated by taking only the leading component of $E_{o,1}(1)(x,t)$ given in Eq. (66). The result is

$$\eta_{o,1}(1)(t,u) \cong i\frac{\sqrt{\pi}\gamma^3\zeta_o^{(o)^2}}{16\epsilon k^2 w^3_{ab}} \frac{\left(\xi^{(1)}_{o,o}\xi^{(1)*}_{o,o}\right)\xi^{(1)}_{o,o}A(\Delta Z)(2s'(+)+s'*(+)-iw)^2}{[s'(+)+s'(+)*]^2[2s'(+)+s'*(+)-s_o(-)-s'(-)]}$$

$$\frac{e^{[2s'(+)+s'*(+)]t}}{u-(w-w_{ab})/k+i[2s'(+)+s'*(+)]/k}$$

$$-\frac{i\gamma^2\zeta_o^{(o)}}{8kw^2_{ab}} \frac{\left(\xi^{(1)}_{o,o}\xi^{(1)*}_{o,o}\right)\xi^{(1)}_{o,o}}{s'(+)+s'*(+)}\left\{\frac{e^{[2s'(+)+s'(+)*]t}}{u-(w-w_{ab})/k+is'(+)/k}\right.$$

$$\left.-\frac{e^{[2s'(+)+s'(+)*]t}}{u-(w-w_{ab})/k+i[2s'(+)+s'*(+)]/k}\right\} \tag{67}$$

$$P^{(1)}_{o,1}(t,x) \cong i\frac{\pi\gamma^3\zeta_o^{(o)^2}}{16\epsilon k^2 w^3_{ab}} \frac{\left(\xi^{(1)}_{o,o}\xi^{(1)*}_{o,o}\right)\xi^{(1)}_{o,o}A^2(\Delta Z)[2s'(+)+s'*(+)-iw]^2}{[s'(+)+s'*(+)]^2[2s'(+)+s'*(+)-s_o(-)-s'(-)]}$$

$$Z\left\{\sqrt{b}[w-w_{ab})/k-i[2s'(+)+s'*(+)]/k-U]\right\}e^{[2s'(+)+s'*(-)]t}$$

$$-\frac{i\gamma^2\zeta_o^{(o)}\sqrt{\pi}}{8kw^2_{ab}} \frac{\left(\xi^{(1)}_{o,o}\xi^{(1)*}_{o,o}\right)\xi^{(1)}_{o,o}}{s'(+)+s'*(+)}A(\Delta Z)e^{[2s'(+)+s'(+)*]t+i(kx-wt)} \tag{68}$$

or

$$P^{(1)}_{o,1}(x,t) \cong \varkappa^{(1)}_{o,1}\left(\xi^{(1)}_{o,o}(t)\xi^{(1)*}_{o,o}(t)\right)\xi^{(1)}_{o,o}(t)e^{i(kx-wt)} \tag{69}$$

where $\varkappa^{(1)}_{o,1}$ is the non-linear susceptibility.

V. CONCLUDING REMARKS

The interaction between light and a molecular beam is studied within the rarefied gas approximation. The theory is free from the use of phenomenological statistical factor in the calculation of macroscopic polarization by introducing the distribution function governed by the classical Boltzmann equation with the collision integral replaced by Krooks model. It is possible to consider the quantum Boltzmann equation, nevertheless the quantum effect is expected to be small at the density range where the maser operates.

It is shown that the intermolecular collision generates higher harmonics which are in general distorted. The degree of distortion of the wave depends on the nature of the collision integral employed, but the qualitative behavior is believed to be similar to what is obtained in the present study.

The general reasonableness of the results obtained, such as the curves of dispersion and the extinction coefficient, strongly suggest the possibility of useful extension of the method to study other limiting case, namely the hydrodynamic model.

The method developed here is presumably applicable in non-linear optics where the optical properties of the medium are believed to be affected by the kinetic behavior of the medium.

APPENDIX A

The equations governing $\tilde{\eta}_{1,\ell}$ $\tilde{\zeta}_{1,\ell}$ $\tilde{\tilde{\eta}}_{1,\ell}$ and $\tilde{\tilde{\zeta}}_{1,\ell}$ are calculated to be

$$\ddot{\tilde{\eta}}_{1,\ell}+2\varphi N\dot{\tilde{\eta}}_{1,\ell}+(\varphi^2 N^2+w_{ab}^2 I)\tilde{\eta}_{1,\ell}=\gamma\zeta_o^{(o)}\ \tilde{\xi}_{1,\ell}+\gamma\sum_{\ell'}\left[\mathcal{E}_{o,\ell'}\tilde{\zeta}_{1,\ell-\ell'}+\mathcal{E}_{1,\ell'}\zeta_{o,\ell-\ell'}\right] \tag{A1}$$

$$\dot{\tilde{\zeta}}_{1,\ell}+\varphi N\tilde{\zeta}_{1,\ell}=\sum_{\ell'}\mathcal{E}_{o,\ell'}\left(\dot{\tilde{\eta}}_{1,\ell-\ell'-1}+\varphi N\tilde{\eta}_{1,\ell-\ell'-1}\right)+\sum_{\ell'}\mathcal{E}_{1,\ell'}\left(\dot{\eta}_{o,\ell-\ell'-1}+\varphi N\eta_{o,\ell-\ell'-1}\right) \tag{A2}$$

The inhomogeneous terms J and s appearing in (32a) and (32b) are easily identified by comparing Eqs. (32a) with (A1), (32b) with (A2) respectively.

$$\ddot{\tilde{\tilde{\eta}}}_{1,\ell}+2\varphi N\dot{\tilde{\tilde{\eta}}}_{1,\ell}+(\varphi^2 N^2+w_{ab}^2 I)\tilde{\tilde{\eta}}_{1,\ell} = \gamma\zeta_o^{(o)}\ \tilde{\tilde{\xi}}_{1,\ell}-2w\tilde{\eta}_{1,\ell}-2u\varphi N\tilde{\eta}_{1,\ell}$$
$$+\gamma\sum_{\ell'}\left(\mathcal{E}_{o,\ell'}\tilde{\tilde{\zeta}}_{1,\ell-\ell'}+\mathcal{E}_{1,\ell'}\zeta_{o,\ell-\ell'}\right) \tag{A3}$$

$$\dot{\tilde{\tilde{\zeta}}}_{1,\ell}+\varphi N\tilde{\tilde{\zeta}}_{1,\ell} = -u\tilde{\zeta}_{1,\ell}+\sum_{\ell'}\Big\{\mathcal{E}_{o,\ell'}\left(\dot{\tilde{\tilde{\eta}}}_{1,\ell-\ell'-1}+\varphi N\tilde{\tilde{\eta}}_{1,\ell-\ell'-1}+u\tilde{\eta}_{1,\ell-\ell'-1}\right)$$
$$+\mathcal{E}_{1,\ell'}\left(\dot{\eta}_{o,\ell-\ell'-1}+\varphi N\eta_{o,\ell-\ell'-1}\right)\Big\} \tag{A4}$$

The solutions for $\tilde{\zeta}_{1,\ell}$ and $\tilde{\eta}_{1,\ell}$ are given by

$$\tilde{\zeta}_{1,\ell}(t,u)=\sum_{\ell'}\int_o^t\Big\{\exp-\varphi(t-t')N\Big\}\Big\{\mathcal{E}_{o,\ell'}(t')\left(\eta_{1,\ell-\ell'-1}(t'u)+\varphi N\eta_{1,\ell-\ell'-1}(t'u)\right)$$
$$+\tilde{\tilde{\mathcal{E}}}_{1,\ell'}(t')\left(\dot{\eta}_{o,\ell-\ell'-1}(t',u)+\varphi N\eta_{o,\ell-\ell'-1}(t',u)\right)\Big\}dt' \tag{A5}$$

$$\tilde{\eta}_{1,\ell'}(t,u) = \int_0^t L(t-t',u)\Big\{\gamma\zeta_0^{(o)}\,\tilde{\xi}_{1,\ell}(t') +$$

$$+\gamma\sum_{\ell'}^{\ell-1}\Big(\mathcal{E}_{o,\ell'}(t')\tilde{\zeta}_{1,\ell-\ell'}(t',u)\tilde{\mathcal{E}}_{1,\ell'}(t')\zeta_{o,\ell-\ell'}(t',u)\Big)dt' \qquad \text{(A6)}$$

Similarly $\tilde{\tilde{\zeta}}_{1,\ell}$ and $\tilde{\tilde{\eta}}_{1,\ell}$ are given by

$$\tilde{\tilde{\zeta}}_{1,\ell'}(t,u) = \int_0^t\Big\{\exp-\varphi(t-t')N\Big\}\Big\{-u\tilde{\zeta}_{1,\ell}+\sum_{\ell'}\mathcal{E}_{o,\ell'}\Big(\dot{\tilde{\tilde{\eta}}}_{1,\ell-\ell'-1}(t',u) +$$

$$+\varphi N\tilde{\tilde{\eta}}_{1,\ell-\ell'-1}(t',u)\Big)+\tilde{\tilde{\mathcal{E}}}_{1,\ell'}(t')\Big(\eta_{o,\ell-\ell'-1}(t',u)+\varphi N\eta_{o,\ell-\ell'-1}(t',u)\Big)\Big\}dt'$$

$$\text{(A7)}$$

$$\tilde{\tilde{\eta}}_{1,\ell}(t,u) = \int_0^t L(t-t',u)\Big\{\gamma\zeta_0^{(o)}\,\tilde{\tilde{\xi}}_{1,\ell}(t')-2u\tilde{\eta}_{1,\ell}(t',u)-2u\varphi N\tilde{\eta}_{1,\ell}(t',u)$$

$$+\gamma\sum_{\ell'}\Big(\mathcal{E}_{o,\ell'}(t')\tilde{\tilde{\zeta}}_{1,\ell-\ell'}(t',u)+\tilde{\tilde{\mathcal{E}}}_{1,\ell'}(t')\zeta_{o,\ell-\ell'}(t',u)\Big)\Big\}dt' \qquad \text{(A8)}$$

The total macroscopic polarization within this approximation is calculated to be

$$P^{+}_{1,\ell}(x,t) = x\exp\psi N\int_{0}^{t}\int_{-\infty}^{\infty} f_o(u)L(t-t',u)\Big\{\gamma\zeta_o^{(o)}\,\tilde{\xi}_{1,\ell}(t')+$$

$$+\frac{1}{u}\Big(F_o(u)-f_o(u)\Big)\eta_{o,\ell}(t',u)\Big\}du\,dt'+x(\exp\psi N)\tilde{q}_{1,\ell}(t) +$$

$$+\exp\psi N\int_{0}^{t}\int_{-\infty}^{\infty} f_o(u)L(t-t',u)\Big\{\gamma\zeta_o^{(o)}\,\tilde{\tilde{\xi}}_{1,\ell}(t')-2u\tilde{\tilde{\eta}}_{1,\ell}(t',u)$$

$$-2u\varphi N\tilde{\eta}_{1,\ell}(t',u)\Big\}du\,dt' + (\exp\psi N)\tilde{\tilde{q}}_{1,\ell}(t) \qquad \text{(A9)}$$

where

$$\tilde{q}_{1,\ell}(t) = \gamma\sum_{\ell'} \int_0^t\int_{-\infty}^{\infty} f_o(u)L(t-t',u)\Big\{\mathcal{E}_{o,\ell'}(t')\tilde{\zeta}_{1,\ell-\ell'}(t',u) +$$

$$+ \tilde{\mathcal{E}}_{1,\ell'}(t')\zeta_{o,\ell-\ell'}(t',u)\Big\}du\,dt' \qquad \text{(A10)}$$

$\tilde{\tilde{q}}_{1,\ell}(t)$ is given similarly as the expression for $\tilde{q}_{1,\ell}$ except that $\tilde{\zeta}_{1,\ell}$ and $\tilde{\mathcal{E}}_{1,\ell}(t)$ appearing in Eq. (A10) are replaced by $\tilde{\tilde{\zeta}}_{1,\ell}$ and $\tilde{\tilde{\mathcal{E}}}_{1,\ell}$ respectively.

An integro-differential equations for $\tilde{\xi}_{1,\ell}(t)$ is Eq. (34)

$$\ddot{\tilde{\xi}}_{1,\ell}(t) + \frac{1}{\mu_o\epsilon_o}(\mu_o\sigma I - 2i\mu_o\epsilon_o wN)\dot{\tilde{\xi}}_{1,\ell}(t) + \frac{1}{\mu_o\epsilon_o}\Big\{(k^2-w^2)N^2 - i\mu_o\sigma wN + \mu_o H(o)\Big\}\tilde{\xi}_{1,\ell}(t)$$

$$= \int_0^t K(t-t',u)\tilde{\xi}_{1,\ell}(t')dt' + \tilde{\Pi}_{1,\ell}(t) + \tilde{w}_{1,\ell}(t) \qquad \text{(A11)}$$

where

$$\tilde{w}_{1,\ell}(t) = -\frac{1}{\epsilon_o}\Big\{\int_{-\infty}^{\infty} \frac{1}{u}\Big(F_o(u)-f_o(u)\Big)\Big[\dot{\eta}_{o,\ell}(t,u)+2iwN\eta_{o,\ell}(t,u)\Big]du$$

$$-w^2N^2\int_0^t\int_{-\infty}^{\infty} \frac{1}{u}\Big(F_o(u)-f_o(u)\Big)\eta_{o,\ell}(t',u)\Big\}dt'du \qquad \text{(A12)}$$

$\tilde{\Pi}_{1,\ell}(t)$ is given similarly to that of $\Pi_{o,\ell}(t)$, except that $q_{o,\ell}(t)$ and their derivatives appearing in (26c) are replaced by $\tilde{q}_{1,\ell}(t)$ etc.

$\tilde{\tilde{\xi}}_{1,\ell}$ is governed by Eq. (34) with inhomogeneous terms $\tilde{\tilde{\Pi}}_{1,\ell}$ and $\tilde{\tilde{w}}_{1,\ell}$. $\tilde{\tilde{\Pi}}_{1,\ell}$ is obtained from Eq. (26c) with $q_{o,\ell}$ replaced by $\tilde{\tilde{q}}_{1,\ell}$.

$\tilde{\tilde{w}}_{1,\ell}$ is given by

$$\tilde{\tilde{w}}_{1,\ell}(t) = -\frac{2}{\epsilon_o}\Big\{\int_0^t\int_{-\infty}^{\infty} uf_o(u)L(t-t',u)\Big(\dot{\tilde{\eta}}_{1,\ell}(t',u)+t\varphi N\tilde{\eta}_{1,\ell}(t',u)\Big)du\,dt'$$

$$+\int_{-\infty}^{\infty} uf_o(u)\frac{\partial L}{\partial t}(o,u)\Big(\dot{\tilde{\eta}}_{1,\ell}(t,u)+t\varphi N\tilde{\eta}_{1,\ell}(t,u)\Big)du+2iwN\int_0^t\int_{-\infty}^{\infty} uf_o(u)\frac{\partial L}{\partial t}(t-t',u)$$

$$\Big(\dot{\tilde{\eta}}_{1,\ell}(t',u)+t\varphi N\tilde{\eta}_{1,\ell}(t',u)\Big)du\,dt'+w^2N^2\int_0^t\int_{-\infty}^{\infty} uf_o(u)L(t-t',u)\Big(\dot{\tilde{\eta}}_{1,\ell}(t',u)+t\varphi N\tilde{\eta}_{1,\ell}(t',u)\Big)du\,dt' \tag{A13}$$

The integro-differential equation (34) is solved by Laplace transform. $\tilde{\xi}_{1,\ell}(t)$, for example, is given by Eq. (27c) where we replace $\mathcal{L}\,\xi_{o,\ell}$ by $\mathcal{L}\tilde{\xi}_{1,\ell}(s)$ which is given by

$$\Big\{s^2 I+\frac{1}{\mu_o\epsilon_o}\Big[(\mu_o\sigma I-2i\mu_o\epsilon_o wN)s+(k^2-w^2)N^2-i\mu_o\sigma wN+u_o\dot{H}(o)-\mathcal{L}K(s)\Big]\Big\}^{-1}$$

$$\Big\{\mathcal{L}\tilde{\Pi}_{1,\ell}(s)+\mathcal{L}\tilde{w}_{1,\ell}(s)+\dot{\tilde{\xi}}_{1,\ell}(o)+\Big[\frac{1}{\mu_o\epsilon_o}(\mu_o\sigma I-2i\mu_o\epsilon_o wN)+sI\Big]\tilde{\xi}_{1,\ell}(o)\Big\} \tag{A14}$$

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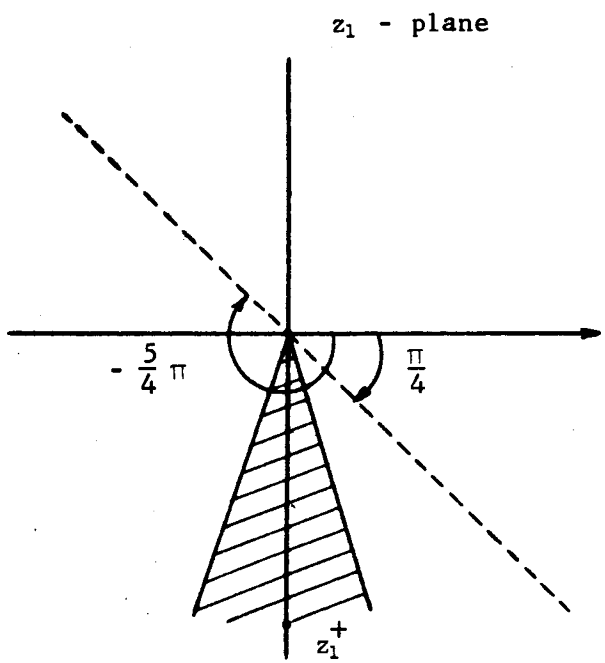

Fig. 1a - ARGUMENT OF z_1^+

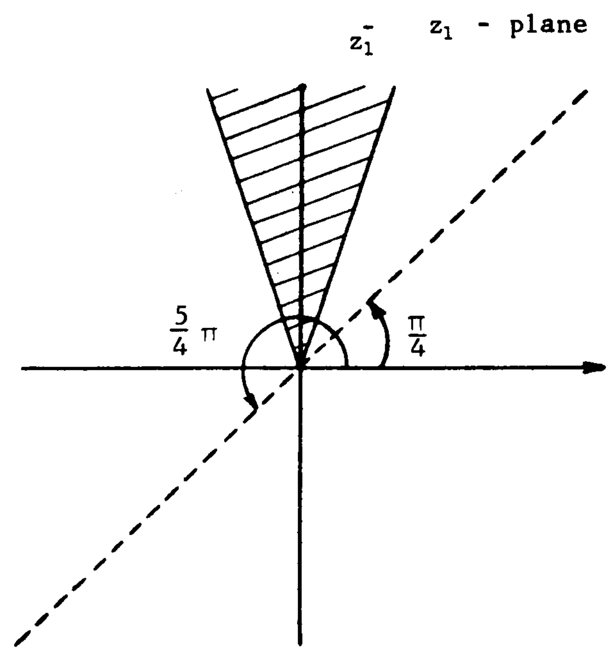

Fig. 1b - ARGUMENT OF z_1^-

Fig. 1

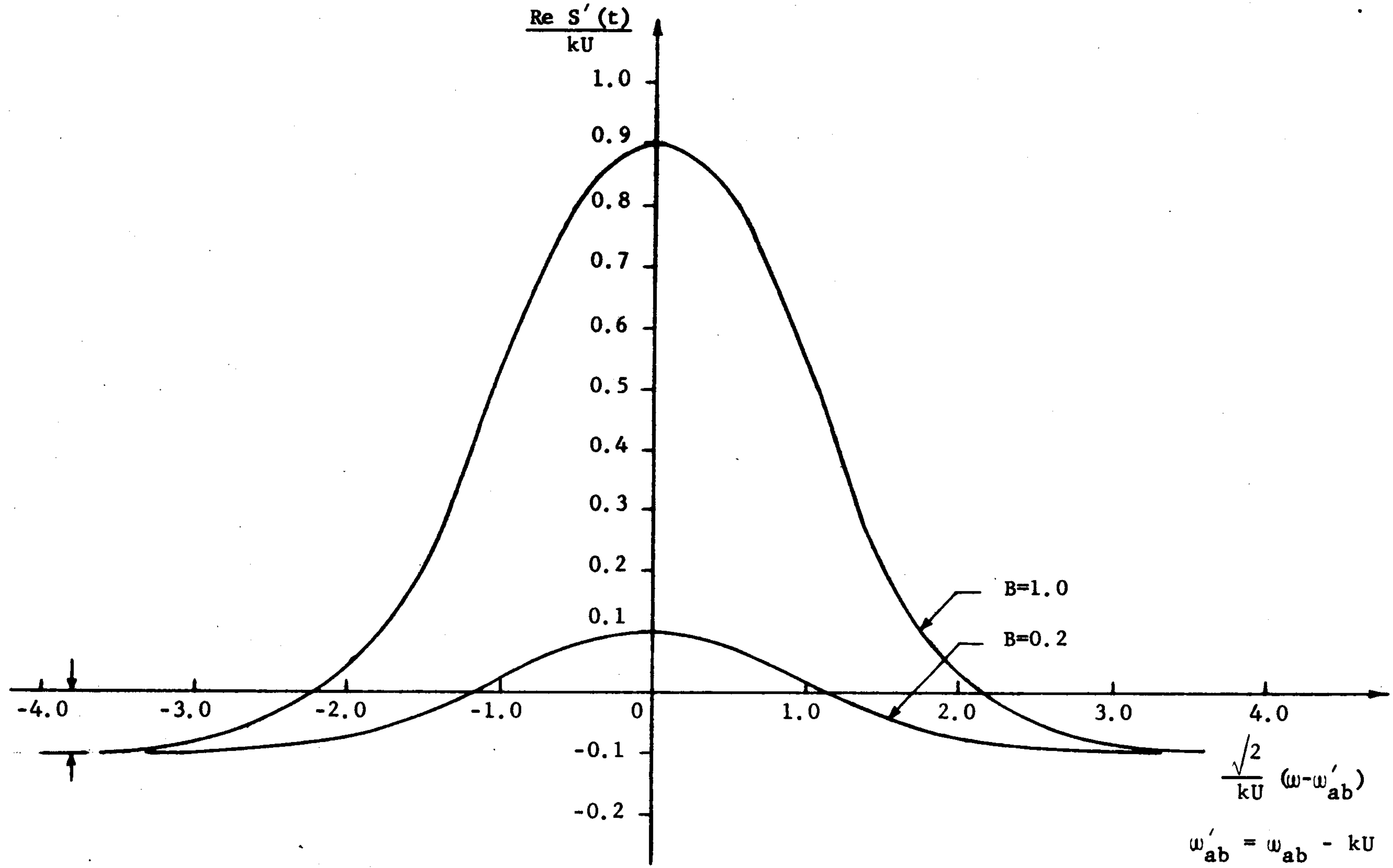

Fig. 2 CURVE FOR SUSTAINED AND DAMPED OSCILLATION

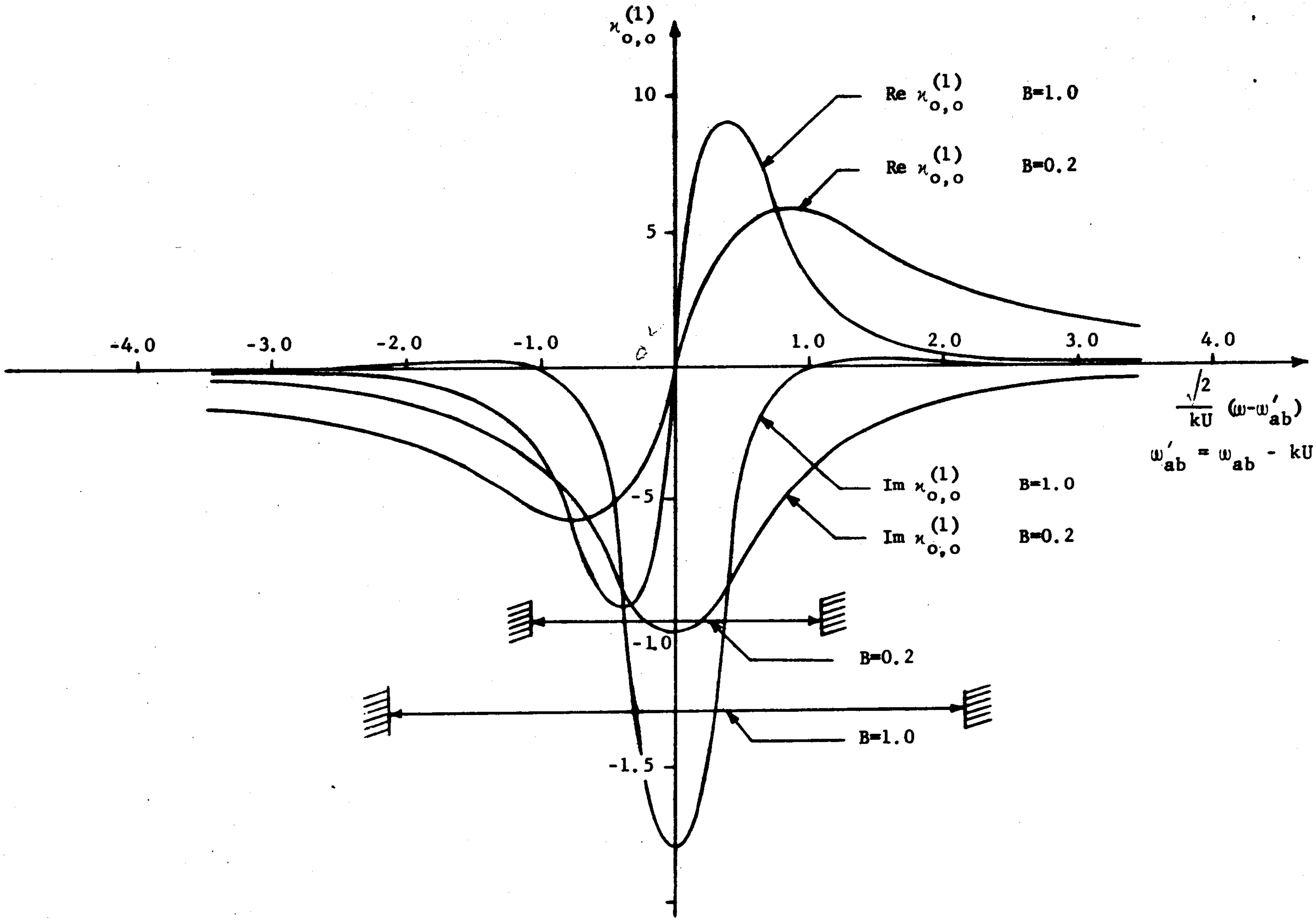

Fig. 3 DISPERSION CURVE FOR MAXWELLIAN DISTRIBUTION