

As the absolute intensities of RAMAN lines due to the symmetrical $\text{C}\equiv\text{N}$, $\text{C}-\text{C}$ and $\text{C}\equiv\text{C}$ stretching vibrations depend on the derivatives of the polarizabilities of the respective bonds, such calculations have been made here from the above derived equation using the internuclear distances, delta-function strengths, BOHR radius, etc. given earlier for carbon subnitride. The calculated values of the polarizability derivatives in \AA^2 for the $\text{C}\equiv\text{N}$, $\text{C}-\text{C}$ and $\text{C}\equiv\text{C}$ bonds are 2.347, 1.227 and 2.411, respectively. These values compare well with those measured experimentally in other related systems having similar chemical bonds. They are 2.61\AA^2 for the $\text{C}\equiv\text{N}$ bond in acetonitrile⁴⁸,

1.37\AA^2 for the $\text{C}-\text{C}$ bond in ethane⁴⁹ and 2.92\AA^2 for the $\text{C}\equiv\text{C}$ bond in acetylene⁴⁹. The bond region electrons alone are involved in the calculation of polarizability derivatives and the nonbond region electrons do not have any influence on the polarizability derivative. As in the case of polarizability, polarizability derivatives also increase with the increase in the bond order. From the results of the present investigation it is seen that the derivatives of polarizabilities are in general transferable from one molecular system to another with similar chemical bonds with nearly identical internuclear distances.

⁴⁸ G. W. CHANTRY and R. A. PLANE, *J. Chem. Phys.* **35**, 1027 [1961].

⁴⁹ T. YOSHINO and H. J. BERNSTEIN, *Spectrochim. Acta* **14**, 127 [1959].

Light Mixing and the Generation of the Second Harmonic in a Plasma in an External Magnetic Field¹

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The theory of light scattering in a plasma is extended by including an external electric field (e.g. the field of a laser beam) in calculating the density fluctuations. It is shown that in the presence of a time constant homogeneous magnetic field there arise density fluctuations with the frequency and the wave number of the external electric field. Expansions of the general expressions are obtained for the case that the frequency is large compared to the electron gyrofrequency. The special case that the external electric field is a transverse wave is discussed in detail.

The light of a second beam may be scattered by these forced density fluctuations. The scattered light has the sum and the difference frequency of the two light beams, i.e. light mixing occurs. In the framework of this theory the effect occurs only if the two beams are parallel. — If one considers the scattering of the same beam that forces the density fluctuations, the scattered light is the second harmonic.

In the past few years several papers have been published dealing with the problem of the scattering of electromagnetic waves in a plasma. The radiation energy $dI_2(\omega_2, \mathbf{k}_2)$ with the frequency ω_2 and the wavevector \mathbf{k}_2 that is scattered per second into a given solid angle do is given by

$$dI_2(\omega_2, \mathbf{k}_2) = \lim_{T \rightarrow \infty} \frac{1}{T} J_1(\omega_1, \mathbf{k}_1) \langle |n(\mathbf{k}, \omega)|^2 \rangle \frac{\Delta\omega_2}{2\pi} \sigma_e do \quad (1)$$

where $J_1(\omega_1, \mathbf{k}_1)$ is the primary intensity with the frequency ω_1 , and the wave vector \mathbf{k}_1 , σ_e is the scattering crosssection for a single electron and T is

the duration of observation. The ensemble average is denoted by $\langle \rangle$ and $n(\mathbf{k}, \omega)$ is the FOURIER transform of the electron density, where \mathbf{k} and ω are given by

$$\mathbf{k}_2 = \mathbf{k}_1 + \mathbf{k} \quad \text{and} \quad \omega_2 = \omega_1 + \omega \quad (2a)$$

$$\text{or} \quad \mathbf{k}_2 = \mathbf{k}_1 - \mathbf{k} \quad \text{and} \quad \omega_2 = \omega_1 - \omega. \quad (2b)$$

These conditions mean that in general the \mathbf{k} -vectors must form a triangle.

From eq. (1) and (2) follows that the spectral distribution of the scattered radiation represents the spectral distribution of the electron density fluctuations, if the primary radiation is monochromatic. By this the problem of calculating the spectrum of the scattered radiation is reduced to the problem of calculating the density fluctuations. This problem has

¹ The basic ideas of this paper were reported in Proc. VIth Intern. Conf. Ionization Phenomena in Gases, Vol. III, p. 189, Paris 1963.



been treated in the case of thermal fluctuations by several authors (e. g. SALPETER² and HAGFORS³).

The aim of the present paper is to discuss first the possibility of forcing density fluctuations by an external electric field (e. g. the field of a laser beam) and then secondly to discuss the scattering of light by these fluctuations.

The problem of calculating the forced density fluctuations is treated with the aid of the linearized VLASOV equation. It can be readily shown that within this approximation for an initially uniform plasma density fluctuations can only be forced by the electric field of a transvers wave, when a magnetic field is present in the plasma and so oriented that a longitudinal component of motion is acquired by electrons oscillating in the external electric field (see Fig. 1).

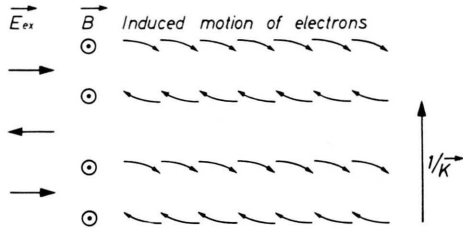


Fig. 1. Qualitative explanation of induced density fluctuations.

I. Forced Density Fluctuations

We consider a given volume V containing a homogeneous plasma consisting of n_0 electrons (the average density being $\bar{n} = n_0/V$) and $N_0 = n_0/Z$ positive Ions of atomic charge Z . It is assumed that the distribution function for the electrons may be written as

$$f(\mathbf{v}, \mathbf{r}, t) = f_0(\mathbf{v}) + f_1(\mathbf{v}, \mathbf{r}, t) \quad (3)$$

where $f_0(\mathbf{v})$ is independent of space and time. Under the assumption

$$|f_1(\mathbf{v}, \mathbf{r}, t)| \ll f_0(\mathbf{v}) \quad (4)$$

we can treat the problem by means of the linearized VLASOV equation.

$$\frac{\partial f_1(\mathbf{v}, \mathbf{r}, t)}{\partial t} + \mathbf{v} \cdot \frac{\partial f_1(\mathbf{v}, \mathbf{r}, t)}{\partial \mathbf{r}} - \frac{e}{m} \mathbf{E}(\mathbf{r}, t) \cdot \frac{\partial f_0(\mathbf{v})}{\partial \mathbf{v}} - \frac{e}{m} \frac{\mathbf{v} \times \mathbf{B}}{c} \cdot \frac{\partial f_1(\mathbf{v}, \mathbf{r}, t)}{\partial \mathbf{v}} = 0. \quad (5)$$

A corresponding relation holds for the distribution function of the ions $F(\mathbf{v})$. We split the electric field

into two parts

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_{\text{in}}(\mathbf{r}, t) + \mathbf{E}_{\text{ex}}(\mathbf{r}, t) \quad (6)$$

where $\mathbf{E}_{\text{ex}}(\mathbf{r}, t)$ is the external electric field to force the density fluctuations. The internal field $\mathbf{E}_{\text{in}}(\mathbf{r}, t)$ is due to the space charges and may be determined from the Poisson equation:

$$\frac{\partial^2 \Phi(\mathbf{r}, t)}{\partial r^2} = -4\pi e [ZN(\mathbf{r}, t) - n(\mathbf{r}, t)], \quad (7)$$

$$\mathbf{E}_{\text{in}}(\mathbf{r}, t) = -\nabla \Phi(\mathbf{r}, t) / \partial \mathbf{r}. \quad (8)$$

The densities $n(\mathbf{r}, t)$ and $N(\mathbf{r}, t)$ are connected with the distribution functions by

$$n(\mathbf{r}, t) = \int_{-\infty}^{+\infty} f(\mathbf{v}, \mathbf{r}, t) d^3v, \quad (9a)$$

$$N(\mathbf{r}, t) = \int_{-\infty}^{+\infty} F(\mathbf{v}, \mathbf{r}, t) d^3v. \quad (9b)$$

We now assume the magnetic field \mathbf{B} to be homogeneous and constant in time. In order to determine the density fluctuations from the preceding equations we perform a FOURIER transformation in space and a LAPLACE transformation in time:

$$f_1(\mathbf{v}, \mathbf{r}, t) = \sum_{\mathbf{k}} f_1(\mathbf{v}, \mathbf{k}, t) \exp\{-i\mathbf{k} \cdot \mathbf{r}\}, \quad (10)$$

$$f_1(\mathbf{v}, \mathbf{k}, s) = \int_0^{\infty} f_1(\mathbf{v}, \mathbf{k}, t) \exp(-st) dt. \quad (11)$$

In eq. (10) it is assumed that the boundary conditions on V are periodic. In the case $V \rightarrow \infty$ the FOURIER sum (10) becomes a FOURIER integral.

From eq. (5) follows by these transformations:

$$[s - i\mathbf{k} \cdot \mathbf{v}] f_1(\mathbf{v}, \mathbf{k}, s) - \frac{e}{m} \mathbf{E}(\mathbf{k}, s) \cdot \frac{\partial f_0(\mathbf{v})}{\partial \mathbf{v}} + \frac{e}{m c} \mathbf{B} \left(\mathbf{v} \times \frac{\partial f_1(\mathbf{v}, \mathbf{k}, s)}{\partial \mathbf{v}} \right) = f^1(\mathbf{v}, \mathbf{k}) \quad (12)$$

$$\text{with } f^1(\mathbf{v}, \mathbf{k}) = f_1(\mathbf{v}, \mathbf{k}, t) \text{ at } t = 0. \quad (13)$$

In order to solve the differential equation (12) we introduce cylindrical coordinates in the \mathbf{v} -space, with the direction of \mathbf{B} as axis and the plane $\varphi = 0$ identical with the plane defined by \mathbf{k} and \mathbf{B} . If we denote the component of \mathbf{v} parallel to \mathbf{B} by u and the component perpendicular by w , we have

$$v_x = w \cos \varphi; \quad v_y = w \sin \varphi; \quad v_z = u. \quad (14)$$

In this coordinate system eq. (12) may easily be solved, and following exactly the procedure outlined by HAGFORS³ one obtains for the FOURIER transforms of the electron and ion density:

² E. E. SALPETER, Phys. Rev. **120**, 1528 [1960].

³ T. HAGFORS, J. Geophys. Res. **66**, 1699 [1961].

$$n(\mathbf{k}, s) = \frac{[Y_e(\mathbf{k}, s) - S_e(\mathbf{k}, s)][1 - R_i(\mathbf{k}, s)] - R_e(\mathbf{k}, s)[Z Y_i(\mathbf{k}, s) - S_i(\mathbf{k}, s)]}{1 - R_e(\mathbf{k}, s) - R_i(\mathbf{k}, s)}, \quad (15)$$

$$N(\mathbf{k}, s) = \frac{[Y_i(\mathbf{k}, s) - Z^{-1} S_i(\mathbf{k}, s)][1 - R_e(\mathbf{k}, s)] - Z^{-1} R_i(\mathbf{k}, s)[Y_e(\mathbf{k}, s) - S_e(\mathbf{k}, s)]}{1 - R_e(\mathbf{k}, s) - R_i(\mathbf{k}, s)} \quad (16)$$

where the functions R, S, Y are defined by

$$R_e(\mathbf{k}, s) = - \int_{-\infty}^{+\infty} d^3 v \int_{-\infty}^{\varphi} \frac{i 4 \pi e c \mathbf{k}}{k^2 B} \frac{\partial f_0(\mathbf{v}')}{\partial \mathbf{v}'} A_e d\varphi', \quad (17 a)$$

$$R_i(\mathbf{k}, s) = - \int_{-\infty}^{+\infty} d^3 v \int_{\varphi}^{+\infty} \frac{i 4 \pi Z e c \mathbf{k}}{k^2 B} \frac{\partial F_0(\mathbf{v}')}{\partial \mathbf{v}'} A_i d\varphi', \quad (17 b)$$

$$S_e(\mathbf{k}, s) = - \int_{-\infty}^{+\infty} d^3 v \int_{-\infty}^{\varphi} \frac{c}{B} \frac{\partial f_0(\mathbf{v}')}{\partial \mathbf{v}'} \mathbf{E}_{ex}(\mathbf{k}, s) A_e d\varphi', \quad (18 a)$$

$$S_i(\mathbf{k}, s) = + \int_{-\infty}^{+\infty} d^3 v \int_{\varphi}^{+\infty} \frac{c}{B} \frac{\partial F_0(\mathbf{v}')}{\partial \mathbf{v}'} \mathbf{E}_{ex}(\mathbf{k}, s) A_i d\varphi', \quad (18 b)$$

$$Y_e(\mathbf{k}, s) = + \int_{-\infty}^{+\infty} d^3 v \int_{-\infty}^{\varphi} \frac{1}{\Omega_e} f^1(\mathbf{v}', \mathbf{k}) A_e d\varphi', \quad (19 a)$$

$$Y_i(\mathbf{k}, s) = + \int_{-\infty}^{+\infty} d^3 v \int_{\varphi}^{+\infty} \frac{1}{\Omega_i} F^1(\mathbf{v}', \mathbf{k}) A_i d\varphi' \quad (19 b)$$

$$\text{with } A_e = \exp \left\{ - \frac{1}{\Omega_e} [(s - i k u \cos \psi)(\varphi - \varphi') - i k w \sin \psi (\sin \varphi - \sin \varphi')] \right\}, \quad (20 a)$$

$$A_i = \exp \left\{ \frac{1}{\Omega_i} [(s - i k u \cos \psi)(\varphi - \varphi') - i k w \sin \psi (\sin \varphi - \sin \varphi')] \right\}. \quad (20 b)$$

Ω_e and Ω_i are the electron and the ion gyrofrequency, ψ is the angle between \mathbf{k} and \mathbf{B} and \mathbf{v}' is obtained from eq. (14) by replacing φ by φ' . The functions R, S and Y are discussed in the appendix.

From the scattering formula (1) we see that the spectrum of the scattered light is essentially determined by the quantity

$$Q(\mathbf{k}, \omega) = \lim_{T \rightarrow \infty} \frac{1}{2 \pi T} \langle |n(\mathbf{k}, \omega)|^2 \rangle \quad (21)$$

where the average is taken over the initial values. In order to connect the quantity $n(\mathbf{k}, s)$ of eq. (15) with $Q(\mathbf{k}, \omega)$, as defined in (21), we put

$$s = \gamma + i \omega \quad (22) \quad \text{with} \quad \gamma = 1/2 T. \quad (23)$$

By introducing the damping γ we take into account that the LAPLACE integral (11) is extended over an infinite time interval, while the duration of observation T — and by that the total measured energy of the scattered radiation — is finite. The damping constant γ is chosen such that for a stationary plasma

$$\int_0^T |n(\mathbf{k}, t)|^2 dt \approx \int_0^\infty |n(\mathbf{k}, t)|^2 \exp(-2 \gamma t) dt. \quad (24)$$

$$\text{From eq. (21) — (23) follows} \quad Q(\mathbf{k}, \omega) = \lim_{\gamma \rightarrow 0} \frac{\gamma}{\pi} \langle |n(\mathbf{k}, s)|^2 \rangle. \quad (25)$$

This quantity can be calculated from eq. (15) — (20). From eq. (15) follows:

$$Q(\mathbf{k}, \omega) = \lim_{\gamma \rightarrow 0} \frac{\gamma}{\pi} \frac{\langle |Y_e(\mathbf{k}, s) - S_e(\mathbf{k}, s)|^2 \rangle |1 - R_i(\mathbf{k}, s)|^2 + \langle |Z Y_i(\mathbf{k}, s) - S_i(\mathbf{k}, s)|^2 \rangle |R_e(\mathbf{k}, s)|^2}{|1 - R_e(\mathbf{k}, s) - R_i(\mathbf{k}, s)|^2}. \quad (26)$$

In performing the ensemble average in eq. (26) one averages in Y over the initial values of the fluctuations and in S over the phase of the external electric field $\mathbf{E}_{\text{ex}}(\mathbf{k}, t)$ at the time $t = 0$. Because these two quantities are obviously not correlated with each other the following relations are valid:

$$\langle |Y_e(\mathbf{k}, s) - S_e(\mathbf{k}, s)|^2 \rangle = \langle |Y_e(\mathbf{k}, s)|^2 \rangle + \langle |S_e(\mathbf{k}, s)|^2 \rangle, \quad (27)$$

$$\langle |Y_i(\mathbf{k}, s) - S_i(\mathbf{k}, s)|^2 \rangle = \langle |Y_i(\mathbf{k}, s)|^2 \rangle + \langle |S_i(\mathbf{k}, s)|^2 \rangle. \quad (28)$$

From these relations follows that we can split $Q(\mathbf{k}, \omega)$ into two parts:

$$Q(\mathbf{k}, \omega) = Q_{\text{th}}(\mathbf{k}, \omega) + Q_{\text{f}}(\mathbf{k}, \omega) \quad (29)$$

where Q_{th} corresponds to the thermal density fluctuations as treated by HAGFORS³, while Q_{f} represents the forced density fluctuations. We have that

$$Q_{\text{f}}(\mathbf{k}, \omega) = \lim_{\gamma \rightarrow 0} \frac{\gamma}{\pi} \frac{\langle |S_e(\mathbf{k}, \omega)|^2 \rangle |1 - R_i(\mathbf{k}, s)|^2 + \langle |S_i(\mathbf{k}, s)|^2 \rangle |R_e(\mathbf{k}, s)|^2}{|1 - R_e(\mathbf{k}, s) - R_i(\mathbf{k}, s)|^2}. \quad (30)$$

Eq. (26) with (27) and (28) is our general result. It implies no assumption as to the transverse or longitudinal character of the external electric field \mathbf{E}_{ex} , but does assume that when \mathbf{E}_{ex} is the electric field of an electromagnetic wave, the influence of the magnetic field of this wave may be neglected.

In the following we shall treat in more details the special case that the external electric field \mathbf{E}_{ex} is transvers with $\omega \gg \omega_p$, Ω_e , ω_p being the plasma frequency and Ω_e being the electron gyrofrequency. In this case the problem is greatly simplified.

At first we note that under these assumptions one term on the right hand of eq. (26) may always be neglected. Either is $\omega/k \ll c$ and consequently $Q_{\text{f}}(\mathbf{k}, \omega) = 0$ because $\mathbf{E}_{\text{ex}}(\mathbf{k}, \omega) = 0$, or $\omega/k \approx c$, in which case $Q_{\text{th}}(\mathbf{k}, \omega)$ may be neglected. This results from the fact that the phase velocity of the thermal fluctuations is coupled to the particle velocities and also from the expansion of Q_{th} and Q_{f} according to (A 17) – (A 18). Since we are interested in the forced density fluctuations, we choose ω and \mathbf{k} such, that they correspond to the forcing field. As pointed out, this implies

$$Q(\mathbf{k}, \omega) = Q_{\text{f}}(\mathbf{k}, \omega). \quad (31)$$

Furthermore from the fact that $\omega/k \approx c$ it follows that:

$$|R_e(\mathbf{k}, \omega)|, \quad |R_i(\mathbf{k}, \omega)| \ll 1 \quad (32)$$

and by this eq. (27) reduces to

$$Q(\mathbf{k}, \omega) = \lim_{\gamma \rightarrow 0} \frac{\gamma}{\pi} \langle |S_e(\mathbf{k}, \omega)|^2 \rangle. \quad (33)$$

With the approximation (A 24) this becomes:

$$Q(\mathbf{k}, \omega) = \lim_{\gamma \rightarrow 0} \frac{\gamma}{\pi} \bar{n}^2 \langle |E_{\text{ex}}(\mathbf{k}, \omega)|^2 \rangle B^2 \frac{1}{\omega^4} \left(\frac{e}{m c} \right)^4 \cdot \sin^2 \psi \sin^2 \chi_1 \sin^2 \chi_2. \quad (34)$$

The angles ψ , χ_1 , and χ_2 denote the directions of \mathbf{k} and \mathbf{E}_{ex} relative to \mathbf{B} (see appendix). Because of the assumption that \mathbf{E}_{ex} is a transvers wave ($\mathbf{E}_{\text{ex}} \perp \mathbf{k}$), the three angles must fulfill the condition (A 22). This implies that only two of these angles can be chosen independently.

II. Light Mixing and the Generation of the Second Harmonic

In the preceding section it was shown that density fluctuations may be forced by an electromagnetic wave of a given frequency ω_{f} and wave vector \mathbf{k}_{f} . The light of a second beam may now be scattered by these fluctuations according to eq. (1). According to eq. (2) the scattered light has the sum and the difference frequency of the two beams — i. e. light mixing occurs. If we neglect the influence of the dispersion, i. e. if we put the index of refraction to unity ($\omega_p \ll \omega_1, \omega_2, \omega_{\text{f}}$), the following relation holds:

$$\frac{\omega_1}{k_1} = \frac{\omega_2}{k_2} = \frac{\omega_{\text{f}}}{k_{\text{f}}} = c. \quad (35)$$

From the conditions (2) and (35) it follows that the light mixing effect occurs only if the two light beams are parallel⁴.

It should be remarked that the scattering formula (1) is valid for both coherent and incoherent density fluctuations⁵. This implies that in the case $\omega_1 = \omega_{\text{f}}$ eq. (1) describes the generation of the second harmonic ($\omega_2 = 2\omega_1$).

⁴ This is a correction to the earlier statement that the difference frequency were generated in the case of anti-parallel beams¹.

⁵ Cf. e. g. W. H. KEGEL, Report IPP 6/21 [1964].

If we assume that the electromagnetic wave forcing the density fluctuations is plane and monochromatic, then in the limit $V \rightarrow \infty$ the FOURIER transform of the electron density is a δ -function in $\omega - \mathbf{k}$ -space. — This indicates that light will be scattered in one direction only. Because of this δ -function it seems more reasonable to ask for the *total* scattered energy than for the differential. From eq. (1) with (21) follows

$$I_2 = J_1 \int_{-\infty}^{+\infty} d\omega \oint d\mathbf{o} Q(\mathbf{k}, \omega) \sigma_e \quad (36)$$

where J_1 is assumed to be perfectly monochromatic.

In order to evaluate the expression (36) we assume the plasma volume to have the form of a rectangular solid with dimensions a , b and L along the axes of the coordinate system, which is assumed to be oriented such that the wave vector \mathbf{k}_t of the electric field, forcing the density fluctuations, possesses only a z -component:

$$\begin{aligned} \mathbf{E}_{\text{ex}}(\mathbf{r}, t) &= \mathbf{E}_{\text{ex}}^0 \exp \{ -i(\mathbf{k}_t \mathbf{r} - \omega_t t - \eta) \} \\ &= \mathbf{E}_{\text{ex}}^0 \exp \{ -i(k_t z - \omega_t t - \eta) \}. \end{aligned} \quad (37)$$

η being the phase of \mathbf{E}_{ex} at $t=0$, $\mathbf{r}=0$.

With these assumptions the FOURIER transform of the forcing field becomes:

$$\mathbf{E}_{\text{ex}}(\mathbf{k}, s) = \mathbf{E}_{\text{ex}}^0 \frac{\exp(i\eta)}{(i\omega_t - s)} \frac{\sin(k_x \frac{1}{2}a)}{k_x} \frac{\sin(k_y \frac{1}{2}b)}{k_y} \frac{\sin\{(k_z - k_t) \frac{1}{2}L\}}{(k_z - k_t)}. \quad (38)$$

Placing this expression in eq. (34) we obtain:

$$Q(\mathbf{k}, \omega) = \left(\lim_{\gamma \rightarrow 0} \frac{\gamma}{\pi} \frac{1}{|i\omega_t - s|^2} \right) \bar{n}^2 E_{\text{ex}}^{02} B^2 \frac{1}{\omega^4} \left(\frac{e}{mc} \right)^4 \sin^2 \psi \sin^2 \chi_1 \sin^2 \chi_2 \frac{\sin^2(k_x \frac{1}{2}a)}{k_x^2} \frac{\sin^2(k_y \frac{1}{2}b)}{k_y^2} \frac{\sin^2\{(k_z - k_t) \frac{1}{2}L\}}{(k_z - k_t)^2}. \quad (39)$$

The ω -integration of eq. (36) and the limiting process of eq. (39) yields the factor 1 from the first bracket in (39). Let

$$A = \bar{n}^2 E_{\text{ex}}^{02} B^2 \frac{1}{\omega_t^4} \left(\frac{e}{mc} \right)^4 \sigma_e \sin^2 \psi \sin^2 \chi_1 \sin^2 \chi_2. \quad (40)$$

There follows from eq. (36):

$$I_2 = J_1 A \oint d\mathbf{o} \frac{\sin^2(k_x \frac{1}{2}a)}{k_x^2} \frac{\sin^2(k_y \frac{1}{2}b)}{k_y^2} \frac{\sin^2\{(k_z - k_t) \frac{1}{2}L\}}{(k_z - k_t)^2}. \quad (41)$$

To simplify the integrand of (41) we make the approximation

$$\frac{\sin^2(k_x \frac{1}{2}a)}{k_x^2} = \frac{a^2}{4} \exp \left\{ -\frac{a^2}{4\pi} k_x^2 \right\}. \quad (42)$$

This approximation is such, that the integral over the left side of eq. (42) gives the same value as the integral over the function on the right side. For $k_x=0$ both functions have the same value. Similar approximations apply for the other factors in eq. (41). These approximations effectively eliminate all details in the diffraction pattern arising from the assumed finite dimensions of the plasma.

We now express \mathbf{k} by means of \mathbf{k}_2 and \mathbf{k}_1 according eq. (2). If we denote by \mathbf{e} the unit vector in the direction of \mathbf{k}_2 , we have

$$\mathbf{k}_2 = k_2 \mathbf{e} = \mathbf{k}_1 \pm \mathbf{k}. \quad (43)$$

Introducing polar coordinates so that \mathbf{e} has the components

$$e_x = \sin \vartheta \cos \varphi; \quad e_y = \sin \vartheta \sin \varphi; \quad e_z = \cos \vartheta \quad (44)$$

we obtain from eqs. (41–44) and the assumption \mathbf{k}_1 parallel to \mathbf{k}_t the relation:

$$\begin{aligned} I_2 = J_1 A \int_0^{2\pi} d\varphi \int_0^\pi d\vartheta \sin \vartheta \frac{a^2 b^2 L^2}{64} \exp \left\{ -\frac{1}{4\pi} (a^2 k_2^2 \sin^2 \vartheta \cos^2 \varphi \right. \\ \left. + b^2 k_2^2 \sin^2 \vartheta \sin^2 \varphi + L^2 [k_2 \cos \vartheta - (k_1 \pm k_t)]^2 \right\}. \end{aligned} \quad (45)$$

We now limit our treatment to the case of the second harmonic generation $k_2 = 2k_f$. We assume also that $a k_f \gg 1$. Then the integral in eq. (45) can easily be evaluated in the special case that the considered plasma volume V is a cube ($a = b = L$). For this case eq. (45) yields (with $\omega_f/k_f = c$):

$$\begin{aligned} I_2 &= J_1 A \frac{\pi^2}{a^2 k_f^2} \frac{V^2}{64} \\ &= J_1 E_{\text{ex}}^2 B^2 \frac{\bar{n}^2 V^2}{a^2 \omega^6} \\ &\quad \cdot \sin^2 \psi \sin^2 \chi_1 \sin^2 \chi_2 \cdot 8.82 \cdot 10^{24} \end{aligned} \quad (46)$$

where all quantities are to be taken in GAUSSIAN units.

In the more general case $a = b \neq L$ but retaining the remaining previous assumptions we obtain from (45):

$$\begin{aligned} I_2 &= J_1 A \frac{V^2}{64 k_f \sqrt{L^2 - a^2}} \frac{\pi^2}{\pi} \exp \left\{ \frac{k_f^2}{\pi} \cdot \frac{a^4}{L^2 - a^2} \right\} \\ &\quad \cdot \left[\Phi \left(\frac{k_f}{\pi} \frac{a^2 - 2L^2}{\sqrt{L^2 - a^2}} \right) - \Phi \left(-\frac{k_f}{\pi} \frac{a^2}{\sqrt{L^2 - a^2}} \right) \right] \end{aligned} \quad (47)$$

where Φ denotes the error integral. An asymptotic expansion for large arguments yields again the result (46), where a^2 now is the cross-section of the plasma volume through which the light beam passes.

III. The Influence of the Dispersion

The situation becomes more complicated upon consideration of the influence of the dispersion. The dispersion relation

$$c^2 k^2 = \omega^2 - \omega_p^2 \quad (48)$$

(valid if ω is large compared to the electron gyro-frequency) implies that the condition (2) cannot be fulfilled for the case of light mixing. This situation is similar to one arising in the case of harmonic generation in crystals, where the intensity of the generated radiation is a periodic function of the crystal thickness, because interference causes an essential part of the electric field to cancel. Noting that according to (48) the generation of the waves in the case of light mixing is not in phase with the propagation, we conclude that the intensity calculated from (1), (36) or (46) is (under the assumption of parallel beams) to be multiplied by the correction factor

$$\frac{\sin^2(\Delta k \frac{1}{2} L)}{(\Delta k \frac{1}{2} L)^2}. \quad (49)$$

⁶ N. M. KROLL, A. RON, and N. ROSTOKER, Phys. Rev. Letters 13, 83 [1964].

L being the path length of the light beams through the plasma and

$$\Delta k = k_1 \pm k_f - \frac{\omega_1 \pm \omega_f}{c_p(\omega_1 \pm \omega_f)}, \quad (50)$$

where $c_p(\omega)$ is the phase velocity in the plasma of a light wave with frequency ω . From the eqs. (46), (48), (49) and (50) we have that I_2 is a periodic function of L . Should all considered frequencies be large compared to the plasma frequency ω_p , then the maximum of this function is independent of the average electron density \bar{n} .

In the special case of the generation of the second harmonic it follows from (48) and (50) that

$$\Delta k = \frac{2\omega_1}{c} \left(\sqrt{1 - \frac{\omega_p^2}{\omega_1^2}} - \sqrt{1 - \frac{\omega_p^2}{4\omega_1^2}} \right). \quad (51)$$

With $\omega_p \ll \omega_1$ this reduces to

$$\Delta k = -\frac{3}{4} \frac{\omega_p^2}{c \omega_1}. \quad (52)$$

Note that for the light of a ruby laser ($\omega_1 = 2.7 \cdot 10^{15} \text{ sec}^{-1}$) and an average electron density $\bar{n} = 7 \cdot 10^{16} \text{ cm}^3$ one calculates from (52) $\Delta k/2 \approx 1 \text{ cm}^{-1}$.

In the case that Δk or L goes to zero the factor (49) becomes unity. This shows that the influence of the dispersion may be neglected in the case of small dimensions.

IV. Discussion

In this paper the problem of light mixing in a plasma is treated in a semi-linear fashion since the density fluctuations are determined from the linearized VLASOV equation, and nonlinear coupling arises only through eq. (1). This approach is a straight forward extension of the theory of light scattering from thermal fluctuations in a magnetic field³. But because of this linearization our theory does not describe effects such as the resonance, occurring when the difference frequency of the two interacting light beams is the plasma frequency^{6,7}. There is also no interaction between the thermal and the forced density fluctuations in the framework of the presented theory. When nonlinear equations are used throughout, this interaction leads to incoherent nonlinear scattering⁸.

⁷ A. SALAT, Z. Naturforschg. 20 a, 689 [1965].

⁸ A. SALAT, private communication.

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Appendix

The functions R and Y defined by (17) and (19) were treated by HAGFORS³ and BERNSTEIN⁹. Under

the assumption that the undisturbed distribution functions $f_0(\mathbf{v})$ and $F_0(\mathbf{v})$ are MAXWELL distributions it has been shown, that the functions R can be written as

$$R_e(\mathbf{k}, s) = - \frac{4 \pi \bar{n} e^2}{\kappa T_e k^2} \left[1 - \frac{s}{\Omega_e} g_e(\mathbf{k}, s) \right], \quad (\text{A } 1)$$

$$R_i(\mathbf{k}, s) = - \frac{4 \pi \bar{n} Z e^2}{\kappa T_i k^2} \left[1 - \frac{s}{\Omega_i} g_i(\mathbf{k}, s) \right] \quad (\text{A } 2)$$

where T_e and T_i are the electron and ion temperature, Ω_e and Ω_i the electron and ion gyrofrequency and g_e and g_i are GORDOJEV integrals defined by

$$g_e(\mathbf{k}, s) = \int_0^\infty dx \exp \left\{ - \frac{s x}{\Omega_e} - [\sin^2 \psi (1 - \cos x) + \frac{1}{2} x^2 \cos^2 \psi] \frac{\kappa T_e k^2}{m \Omega_e^2} \right\}, \quad (\text{A } 3)$$

$$g_i(\mathbf{k}, s) = \int_0^\infty dx \exp \left\{ - \frac{s x}{\Omega_i} - [\sin^2 \psi (1 - \cos x) + \frac{1}{2} x^2 \cos^2 \psi] \frac{\kappa T_i k^2}{m \Omega_i^2} \right\}, \quad (\text{A } 4)$$

ψ being the angle between \mathbf{k} and \mathbf{B} .

In order to determine the quantity $\gamma \langle |Y_e(\mathbf{k}, s)|^2 \rangle$ we follow the method used by SALPETER² by introducing at this point the effect of particle discreteness by writing

$$f^1(\mathbf{r}, \mathbf{v}) = \sum_{j=1}^n \delta[\mathbf{r} - \mathbf{r}_j(t=0)] \cdot \delta[\mathbf{v} - \mathbf{v}_j(t=0)]. \quad (\text{A } 5)$$

Using this in eq. (19) one deduces after some calculation⁵:

$$\gamma \langle |Y_e(\mathbf{k}, s)|^2 \rangle = \frac{n_0}{\Omega_e} \Re g_e(\mathbf{k}, s). \quad (\text{A } 6)$$

In the limit $\gamma \rightarrow 0$ this becomes

$$\lim_{\gamma \rightarrow 0} \gamma \langle |Y_e(\mathbf{k}, s)|^2 \rangle = \frac{V \kappa T_e k^2}{4 \pi \omega e^2} \Im R_e(\mathbf{k}, \omega). \quad (\text{A } 7)$$

The corresponding relations for the ions are

$$\gamma \langle |Y_i(\mathbf{k}, s)|^2 \rangle = \frac{N_0}{\Omega_i} \Re g_i(\mathbf{k}, s), \quad (\text{A } 8)$$

$$\lim_{\gamma \rightarrow 0} \gamma \langle |Y_i(\mathbf{k}, s)|^2 \rangle = \frac{V \kappa T_i k^2}{4 \pi \omega Z^2 e^2} \Im R_i(\mathbf{k}, \omega). \quad (\text{A } 9)$$

These relations also follow, but more readily, as shown by HAGFORS³, if one additionally assumes that the spatial density fluctuations are independent of the velocities of the particles.

In order to determine the functions S from (18) we consider the vector \mathbf{E}_{ex} in cylindrical coordinates, with the axis parallel to \mathbf{B} . We denote by χ_1 , the angle between \mathbf{E}_{ex} and \mathbf{B} and by χ_2 the angle between the component of \mathbf{E}_{ex} perpendicular to \mathbf{B} and the plane defined by \mathbf{B} and \mathbf{k} . In this coordinate system \mathbf{E}_{ex} has the components:

$$E_{\text{ex}, x} = E_{\text{ex}} \sin \chi_1 \cos \chi_2, \quad E_{\text{ex}, y} = E_{\text{ex}} \sin \chi_1 \sin \chi_2, \quad E_{\text{ex}, z} = E_{\text{ex}} \cos \chi_1. \quad (\text{A } 10)$$

This representation of \mathbf{E}_{ex} corresponds to that of \mathbf{v} in (14). It follows that:

$$\mathbf{E}_{\text{ex}} \cdot \mathbf{v}' = E_{\text{ex}} [w \cos \varphi' \sin \chi_1 \cos \chi_2 + w \sin \varphi' \sin \chi_1 \sin \chi_2 + u \cos \chi_2]. \quad (\text{A } 11)$$

⁹ I. B. BERNSTEIN, Phys. Rev. **109**, 10 [1958].

In order to obtain expressions for S similar to that for R we rewrite (A 11) :

$$\mathbf{E}_{\text{ex}} \cdot \mathbf{v}' = E_{\text{ex}} \frac{\sin \chi_1 \cos \chi_2}{k \sin \psi} (k w \sin \psi \cos \varphi' + k u \cos \psi) + E_{\text{ex}} u \left(\cos \chi_1 - \frac{\cos \psi \sin \chi_1 \cos \chi_2}{\sin \psi} \right) + E_{\text{ex}} w \sin \varphi' \sin \chi_1 \sin \chi_2. \quad (\text{A } 12)$$

Correspondingly we split $S_e(\mathbf{k}, s)$ into three terms:

$$S_e(\mathbf{k}, s) = S_{eI}(\mathbf{k}, s) + S_{eII}(\mathbf{k}, s) + S_{eIII}(\mathbf{k}, s). \quad (\text{A } 13)$$

Since the expression S_{eI} has the same structure as R_e , we have:

$$S_{eI}(\mathbf{k}, s) = i \frac{e}{\kappa T_e} \bar{n} E_{\text{ex}}(\mathbf{k}, s) \left(1 - \frac{s}{\Omega_e} g_e(\mathbf{k}, s) \right) \frac{\sin \chi_1 \cos \chi_2}{k \sin \psi}. \quad (\text{A } 14)$$

The second term yields:

$$S_{eII}(\mathbf{k}, s) = i \bar{n} \frac{k e E_{\text{ex}}(\mathbf{k}, s)}{m \Omega_e^2} \cos \psi \left(\cos \chi_1 - \frac{\cos \psi \sin \chi_1 \cos \chi_2}{\sin \psi} \right) \cdot \int dx x \exp \left\{ -\frac{s}{\Omega_e} x - [\sin^2 \psi (1 - \cos x) + \frac{1}{2} x^2 \cos^2 \psi] \frac{\kappa T_e k^2}{m \Omega_e^2} \right\} \quad (\text{A } 15)$$

and the third term in (A 13) may be reduced to a onedimensional integral by a procedure similar to that used for the treatment of R_e . Then one obtains:

$$S_{eIII}(\mathbf{k}, s) = -i \bar{n} \frac{k e E_{\text{ex}}(\mathbf{k}, s)}{m \Omega_e^2} \sin \psi \sin \chi_1 \sin \chi_2 \cdot \int_0^\infty dx (1 - \cos x) \exp \left\{ -\frac{s}{\Omega_e} x - [\sin^2 \psi (1 - \cos x) + \frac{1}{2} x^2 \cos^2 \psi] \frac{\kappa T_e k^2}{m \Omega_e^2} \right\}. \quad (\text{A } 16)$$

Similar expressions are valid for $S_i(\mathbf{k}, s)$.

(A 13) with (A 14) to (A 16) is the general form of our result. To this point we made no assumptions as to the transvers character of the external electric field, nor was a value assumed for the ratio of its frequency to the electron gyrofrequency.

Let us now make an expansion of S_e for $\omega \gg \Omega_e$. For this purpose we write

$$s = \gamma + i\omega \quad \text{and} \quad \exp(ix) = \cos x + i \sin x$$

and expand the three terms of S_e according to the formulae

$$\int_0^\infty \sin ax f(x) dx = \frac{f(0)}{a} - \frac{f'(0)}{a^3} + \frac{f^{IV}(0)}{a^5} - \dots, \quad (\text{A } 17)$$

$$\int_0^\infty \cos ax f(x) dx = -\frac{f'(0)}{a^2} + \frac{f'''(0)}{a^4} - \frac{f^{VI}(0)}{a^6} + \dots \quad (\text{A } 18)$$

which are obtained by continued partial integration. The expansions are valid for all functions $f(x)$, for which all derivatives exist and go to zero for $x \rightarrow \infty$. In the special case of the function S_e (a being ω/Ω_e) the series converge rapidly, if $\omega \gg \Omega_e$. From eqs. (A 14) to (A 16) it follows in the limit $\gamma \rightarrow 0$, if all terms of higher order in B are neglected, that

$$S_{eI}(\mathbf{k}, s) = -i \bar{n} \frac{k e E_{\text{ex}}(\mathbf{k}, s)}{m \omega^2} \frac{\sin \chi_1 \cos \chi_2}{\sin \psi}, \quad (\text{A } 19)$$

$$S_{eII}(\mathbf{k}, s) = -i \bar{n} \frac{k e E_{\text{ex}}(\mathbf{k}, s)}{m \omega^2} \cos \psi \left(\cos \chi_1 - \frac{\cos \psi \sin \chi_1 \sin \chi_2}{\sin \psi} \right), \quad (\text{A } 20)$$

$$S_{eIII}(\mathbf{k}, s) = -\bar{n} \frac{\Omega_e k e E_{\text{ex}}(\mathbf{k}, s)}{m \omega^3} \sin \psi \sin \chi_1 \sin \chi_2. \quad (\text{A } 21)$$

In the special case that the external electric field is a transvers wave ($\mathbf{E}_{\text{ex}} \mathbf{k} = 0$) the angles ψ , χ_1 and χ_2 satisfy the condition

$$\cos \chi_1 \cos \psi = -\sin \chi_1 \cos \chi_2 \sin \psi. \quad (\text{A } 22)$$

From this condition follows that with the approximations (A 19) and (A 20) the sum of S_{eI} and S_{eII} vanishes, i. e.:

$$S_e(\mathbf{k}, \omega) = S_{eIII}(\mathbf{k}, \omega). \quad (\text{A } 23)$$

In this case only the component of \mathbf{E}_{ex} that is perpendicular to \mathbf{B} contributes to S_e .

If we finally assume the index of refraction to be unity, i. e. $\omega/k = c$, we have¹⁰:

$$S_e(\mathbf{k}, \omega) = -\bar{n} \frac{\Omega_e e E_{\text{ex}}(\mathbf{k}, \omega)}{m c \omega^2} \sin \psi \sin \chi_1 \sin \chi_2 = -\bar{n} E_{\text{ex}}(\mathbf{k}, \omega) B \cdot \frac{1}{\omega^2} \left(\frac{e}{m c} \right)^2 \sin \psi \sin \chi_1 \sin \chi_2. \quad (\text{A } 24)$$

¹⁰ This is a correction of the result given in ref. ¹, which differs from (A 24) by a numerical factor.