distribution. The over-all effect of such collisions is difficult to estimate. Previous calculations[16] concerning molecular beams interacting with the exterior cloud, although not strictly applicable to the experiment, have indicated the correction is probably of the order of only a few percent. To emphasize the above considerations, portions of the experimental curves where some doubt exists concerning the magnitude of $F(\epsilon)$ are indicated by broken lines.

Finally, there remains the question of whether or not the discharge electrons are in equilibrium with the electric field at the values of pressure and electrode spacing used in this experiment. While there is probably not time for elastic collisions alone to cause the distribution function to reach equilibrium, inelastic collisions play an important role. The product of electrode spacing and reduced pressure, $p_0\,x=2.2$ Torr cm, is comparable or greater than the $p_0\,x$ for which Chanin and Rork[17] have obtained essentially constant α/p_0 in helium for fixed higher values of E/N. In view of their results, there is strong evidence that the experimental distribution represents an equilibrium situation, especially in the important range $E/N\leq 3\times 10^{-15}$ V cm^2.

ACKNOWLEDGMENT

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[16] I. R. Estermann, O. C. Simpson, and O. Stern, Phys. Rev. **71**, 238 (1947).

[17] Lorne M. Chanin and G. D. Rork, Phys. Rev. **133**, A1005 (1964).

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Quantum Theory of Photon Interaction in a Plasma

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The quantum theory of interaction of electromagnetic waves in a plasma is formulated from two different points of view. The first is to consider scattering of light off light (in the form of laser beams) with the plasma acting as a mediator of the interaction; the second is to consider scattering of one of the light beams off a system consisting of the plasma and the other laser beam. Based on the first viewpoint, the light-light scattering cross sections, both elastic and inelastic, are calculated in the lowest order. By summing over the final states of one of the photons, we obtain, based on the above results, the lowest order cross section of scattering of the other photon from the photon-plasma system. In the presence of a second stimulating laser beam, this cross section is enhanced. When both laser beams are very intense, the lowest order perturbation treatment is inadequate. The second viewpoint is then conveniently adopted to include the plasma-laser beam interaction to all orders. The results are discussed and compared with those in previous treatments. Finally, a simple model is considered. In this model, the plasmon is treated as the quantum of a harmonic oscillator which is linearly coupled to a system of phonons. All the previous results are explicitly verified in this model, which is solved exactly.

I. INTRODUCTION

TREATMENTS of the interaction of light with light in a plasma, both quantum mechanically[1,2] and classically,[3] have been given. However, the results of Refs. 1 and 2 differ from those of Ref. 3. This difference was finally resolved in a brief communication.[4] The present paper gives a systematic account of the subject.[5]

II. GENERAL THEORY OF LIGHT-LIGHT INTERACTION

We consider the interaction of a plasma with photons 1, 2, 3, and 4. The total Hamiltonian is

$$H=H_0+H_1+H_2\,, \tag{1}$$

[1] P. M. Platzman, S. J. Buchsbaum, and N. Tzoar, Phys. Rev. Letters **12**, 573 (1964); also P. M. Platzman and N. Tzoar, Phys. Rev. **136**, A11 (1964).

[2] D. F. Dubois and V. Gilinsky, Phys. Rev. **135**, A995 (1964).

[3] N. M. Kroll, A. Ron, and N. Rostoker, Phys. Rev. Letters **13**, 83 (1964).

[4] H. Cheng and Y. C. Lee, Phys. Rev. Letters **14**, 426 (1965).

[5] After the publication of Ref. 4, a letter by D. F. Dubois appeared [Phys. Rev. Letters **14**, 818 (1965)] the results of which are essentially contained in Ref. 4 and are roughly equivalent to those of Sec. III in the present paper.

where H_0 is the Hamiltonian of the plasma and the free radiation field;

$$H_1=C_{24}(a_{\mathbf{k}_2}{}^\dagger a_{\mathbf{k}_4}\rho_{-\mathbf{k}}+a_{\mathbf{k}_4}{}^\dagger a_{\mathbf{k}_2}\rho_{\mathbf{k}}),\quad \mathbf{k}=\mathbf{k}_4-\mathbf{k}_2, \tag{2}$$

$$H_2=C_{13}a_{\mathbf{k}_3}{}^\dagger a_{\mathbf{k}_1}\rho_{-\mathbf{k}'}+\text{Hermitian conjugate},\quad \mathbf{k}'=\mathbf{k}_1-\mathbf{k}_3, \tag{3}$$

and

$$\rho_{\mathbf{k}}=\frac{1}{\sqrt{V}}\sum_{\mathbf{p}} b_{\mathbf{p}}{}^\dagger b_{\mathbf{p}+\mathbf{k}}, \tag{4}$$

$$C_{ij}=2\pi\hbar e^2(m^2V\omega_i\omega_j)^{-1/2}\hat{e}_i\cdot\hat{e}_j. \tag{5}$$

In above, $a_{\mathbf{k}}$, $a_{\mathbf{k}}{}^\dagger$ and $b_{\mathbf{k}}$, $b_{\mathbf{k}}{}^\dagger$ are the annihilation and creation operators for photons and electrons, respectively; V is the total volume of interaction; m is the electron mass, $\hat{e}_i$ is the unit polarization vector of photon i. The Hamiltonian H_1 represents the interaction of photons 2 and 4 with the plasma, and H_2 represents the interaction of photons 1 and 3 with the plasma. Anticipating that both $|\omega_1-\omega_3|$ and $|\omega_2-\omega_4|$ are close to the plasma resonance frequency we have included only the resonance interactions in Eqs. (2) and (3). In H_1 and H_2, the $\mathbf{A}\cdot\mathbf{j}$ term is neglected.[2,1]

The problem of light-light interaction in a plasma can be solved in two complimentary ways. We may consider either the scattering of photons 1 and 2 into photons 3 and 4, with the plasma acting as a mediator, or alternatively, the scattering of photon 1 into photon 3 by a combined system consisting of the plasma and the other photons. In the former case, H_0 is treated as the unperturbed Hamiltonian and the scattering is caused by H_1+H_2 together. In the latter viewpoint, since the interaction of photons 1 and 3 is given by H_2, therefore H_0+H_1 is treated as the unperturbed Hamiltonian. If, in the former case, all processes contributing to the scattering of photon 1 to photon 3 are summed, we may indeed obtain the result in the latter case. However, it is both interesting and convenient to formulate our problem based on these two viewpoints separately. In fact, the formulas obtained in the second formulation prove useful for taking into account the interaction H_1 to all orders.

Let us take the first viewpoint. The S matrix is given by

$$S_{fi}=\langle\varphi_f n_1'n_2'n_3'n_4'|\times T\left\{\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{\infty}(H_1(t)+\mathrm{H}_2(t))dt\right]\right\}\times|\varphi_i n_1n_2n_3n_4\rangle. \tag{6}$$

In (6), φ_f and φ_i are the initial and final states of the plasma; n_1', n_2', n_3', n_4' and n_1, n_2, n_3, n_4 are the initial and final occupation numbers of photon states 1, 2, 3, and 4, respectively; T is the time-ordering operator, and

$$H_1(t)=\exp\left(\frac{i}{\hbar}H_0t\right)H_1\exp\left(-\frac{i}{\hbar}H_0t\right), \tag{7}$$

$$H_2(t)=\exp\left(\frac{i}{\hbar}H_0t\right)H_2\exp\left(-\frac{i}{\hbar}H_0t\right). \tag{8}$$

The lowest order term in (6) which gives rise to light-light scattering is

$$S_{fi}=\left(\frac{i}{\hbar}\right)^2\int_{-\infty}^{\infty}dt_1\int_{-\infty}^{\infty}dt_2\langle\varphi_f n_1'n_2'n_3'n_4'|\times T(H_1(t_1)H_2(t_2))|\varphi_i n_1n_2n_3n_4\rangle. \tag{9}$$

The probability of transition of photons 1, 2 to photons 3, 4 is equal to the square of the absolute value of S_{fi} in (9) after summing over the final states and averaging over the initial states of the plasma,

$$P=\bar{\sum_{\varphi_i}}\sum_{\varphi_f}|S_{fi}|^2=\frac{C_{13}{}^2C_{24}{}^2}{\hbar^4}\int_{-\infty}^{\infty}dt_1\int_{-\infty}^{\infty}dt_2\int_{-\infty}^{\infty}dt_3\int_{-\infty}^{\infty}dt_4e^{-i\omega(t_1-t_3)+i\omega'(t_2-t_4)}\times|\langle n_1'n_2'n_3'n_4'|a_3{}^\dagger a_4{}^\dagger a_1a_2|n_1n_2n_3n_4\rangle|^2\langle\{T(\rho_{\mathbf{k}}(t_1)\rho_{-\mathbf{k}'}(t_2))\}^\dagger T(\rho_{\mathbf{k}}(t_3)\rho_{-\mathbf{k}'}(t_4))\rangle, \tag{10}$$

where

$$\omega=\omega_4-\omega_2,\quad \omega'=\omega_1-\omega_3;$$

$\langle\ \rangle$ denotes the ensemble average over the initial plasma states and

$$\rho_{\mathbf{k}}(t)=e^{(i/\hbar)H_0t}\rho_{\mathbf{k}}e^{-(i/\hbar)H_0t}.$$

The term $a_{\mathbf{k}_2}{}^\dagger a_{\mathbf{k}_4}\rho_{\mathbf{k}}$ in H_1 does not contribute since $n_4=0$ in the present case.

If we consider the scattering of photon 1 to photon 3, treating H_2 to the lowest order only but including H_1 to all orders, then (6) is approximated as

$$S_{fi}=\langle\varphi_f n_1'n_2'n_3'n_4'|-\frac{i}{\hbar}\int_{-\infty}^{\infty}T\left\{\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{\infty}H_1(t')dt'\right]H_2(t)\right\}dt\times|\varphi_i n_1n_2n_3n_4\rangle.$$

From the above expression the transition probability of photon 1 to photon 3, after summing over the final states of the plasma and the other photons and averaging over the initial plasma states, is obtained as

$$P=\langle (M^\dagger M)_{n_2n_4,n_2n_4}\rangle, \tag{11}$$

where $O_{n_2n_4,n_2n_4}$ denotes $\langle n_2n_4|O|n_2n_4\rangle$ and where

$$M=\frac{C_{13}}{\hbar}\int_{-\infty}^{\infty}\exp(-i\omega' t_2)\times T\left\{\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{\infty}H_1(t_1)dt_1\right]\rho_{-\mathbf{k}'}(t_2)\right\}dt_2. \tag{12}$$

In (11), the final states of the plasma and those of photon 2 and 4 have been summed and an ensemble average has been taken over the initial plasma states. Equation (11) includes the interaction H_1 to all orders.

If we adopt the second viewpoint, i.e., taking H_0+H_1 as the unperturbed Hamiltonian and H_2 as the interaction, the S matrix is given by

$$S_{fi}=\lim_{\substack{t_0\to-\infty\\ t\to\infty}}\left\langle T\left\{\exp\left[-\frac{i}{\hbar}\int_{t_0}^{t}(H_0+H_1)dt'\right]\right\}\psi_f(t_0)\right|\times\left|T\left\{\exp\left[-\frac{i}{\hbar}\int_{t_0}^{t}Hdt'\right]\right\}\psi_i(t_0)\right\rangle, \tag{13a}$$

where the possible explicit dependence of H_0, H_1 and H_2 on time is implied but not indicated for the sake of clarity. The function $\psi(t_0)$ represents a state of the plasma and the photons at t_0. To understand (13a), we note that

$$T\left\{\exp\left[-\frac{i}{\hbar}\int_{t_0}^{t}Hdt'\right]\right\}|\psi_i(t_0)\rangle$$

is the state at time t which propagates from $|\psi_i(t_0)\rangle$ at time t_0 with the full Hamiltonian $H=H_0+H_1+H_2$. Since H_0+H_1 is now considered as the unperturbed Hamiltonian, the state $|\psi_f(t_0)\rangle$ would become

$$T\left\{\exp\left[-\frac{i}{\hbar}\int_{t_0}^{t}(H_0+H_1)dt'\right]\right\}|\psi_f(t_0)\rangle$$

at time t if the interaction is absent. The transition amplitude induced by the interaction H_2 from state i to state f is therefore given by (13a). Note that if H_2 is zero, the right side of (13a) becomes δ_{fi}, as is expected.

Use will be made of the following operator relation:

$$T\left[\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t}(\mathcal{H}_0+\mathcal{H}')dt'\right)\right]=U(t,0)T\left[\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t}\mathcal{H}_I'(t')dt'\right)\right]U(0,t_0), \tag{13b}$$

where

$$U(t_1,t_2)=\begin{cases}T\left[\exp\left(-\frac{i}{\hbar}\int_{t_2}^{t_1}\mathcal{H}_0dt\right)\right], & \text{if } t_1>t_2\\ \left\{T\left[\exp\left(-\frac{i}{\hbar}\int_{t_1}^{t_2}\mathcal{H}_0dt\right)\right]\right\}^{-1}, & \text{if } t_2>t_1\end{cases} \tag{13c}$$

and

$$\mathcal{H}_I'(t)=U^{-1}(t,0)\mathcal{H}'U(t,0).$$

If we identify $\mathcal{H}_0=H_0+H_1$ and $\mathcal{H}'=H_2$, and keep only the linear term in H_2, (13a) is reduced to

$$S_{fi}=\lim_{\substack{t_0\to-\infty\\ t\to\infty}}-\frac{i}{\hbar}\langle\psi_f^I(t_0)|\times\int_{t_0}^{t}U^{-1}(t',0)H_2U(t',0)dt'|\psi_i^I(t_0)\rangle, \tag{13d}$$

where $U(t_1,t_2)$ is defined as in (13c) with H_0 replaced by H_0+H_1, and where

$$|\psi^I(t_0)\rangle=U(0,t_0)|\psi(t_0)\rangle$$

is the wave function in the interaction representation with H_0+H_1 considered as the unperturbed Hamiltonian. Also, $U^{-1}(t,0)H_2U(t,0)$ is the operator H_2 in the same interaction representation.

If we apply (13b) to (13d) again but this time identifying $\mathcal{H}_0$ with H_0, $\mathcal{H}'$ with H_1, we obtain

$$S_{fi}=\lim_{\substack{t_0\to-\infty\\ t\to\infty}}-\frac{i}{\hbar}\langle e^{(i/\hbar)H_0t_0}\psi_f(t_0)|\int_{t_0}^{t}dt'\left\{T\left(\exp\left[-\frac{i}{\hbar}\int_{t_0}^{t'}H_1(t'')dt''\right]\right)\right\}^{-1}H_2(t')\times T\left(\exp\left[-\frac{i}{\hbar}\int_{t_0}^{t'}H_1(t'')dt''\right]\right)|e^{(i/\hbar)H_0t_0}\psi_i(t_0)\rangle, \tag{13e}$$

where $H_1(t'')$ and $H_2(t'')$ are defined as in Eqs. (7) and (8).

If we substitute (3) into (13d), and choose

$$e^{(i/\hbar)H_0t_0}|\psi_i(t_0)\rangle=|\varphi_in_1n_2n_3n_4\rangle,\quad e^{(i/\hbar)H_0t_0}|\psi_f(t_0)\rangle=|\varphi_fn_1'n_2'n_3'n_4'\rangle,$$

which are eigenstates of H_0, then (13a) becomes

$$S_{fi}=-(i/\hbar)C_{13}\langle n_1'n_3'|a_3^\dagger a_1|n_1n_3\rangle\langle\varphi_fn_2'n_4'|N_{-\mathbf{k}'-\omega'}|\varphi_in_2n_4\rangle, \tag{14}$$

where

$$N_{-\mathbf{k}',-\omega'}=\int_{-\infty}^{\infty}e^{-i\omega' t}\left\{T\left(\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t}H_1(t')dt'\right]\right)\right\}^{-1}\rho_{-\mathbf{k}'}(t)T\left(\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t}H_1(t')dt'\right]\right)dt$$

or

$$N_{-\mathbf{k}',-\omega'}=\int_{-\infty}^{\infty}\left(\rho_{-\mathbf{k}'}(t)+\frac{i}{\hbar}\int_{-\infty}^{t}[H_1(t_1),\rho_{-\mathbf{k}'}(t)]dt_1+\left(\frac{i}{\hbar}\right)^2\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2[H_1(t_2),[H_1(t_1),\rho_{-\mathbf{k}'}(t)]]+\cdots\right)e^{-i\omega' t}dt. \quad (15)$$

Just like the amplitude M in (12), S_{fi} in (15) is linear in H_2 but includes H_1 to all orders. Notice that[6] the expectation value of $N_{-\mathbf{k}'-\omega'}$ is the Fourier transform of the electron density of the plasma under the influence of laser beams 2 and 4. The total probability of scattering of photon 1 to photon 3 from the combined system consisting of the plasma and the laser beams 2 and 4 is obtained by summing $|S_{fi}|^2$ over the final states of the plasma and the laser beams 2 and 4. Upon averaging over the initial plasma states, the total probability is derived from (14) as

$$P=(C_{13}^2/\hbar^2)\langle(N_{-\mathbf{k}'-\omega'}{}^\dagger N_{-\mathbf{k}'-\omega'})_{n_2n_4,n_2n_4}\rangle. \quad (16)$$

Equation (16) gives of course the same result as (11). This fact can be concluded from the relation below:

$$M=\frac{C_{13}}{\hbar}\mathrm{T}\left\{\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{\infty}H_1(t_1)dt_1\right]\right\}N_{-\mathbf{k}',-\omega'}. \quad (17)$$

The differential cross section of scattering of photon 1 to photon 3 is obtained from (16) by multiplying it with the density of states

$$\frac{V}{(2\pi)^3}\int d^3k$$

and dividing it by the flux c/V and the total interaction time τ.

$$\frac{d^2\sigma}{d\omega d\Omega}=\frac{V}{\tau}\frac{e^4}{m^2}\frac{(\hat{e}_i\cdot\hat{e}_3)^2}{2\pi c^4}\frac{\omega_3}{\omega_1}\times\langle(N_{-\mathbf{k}',-\omega'}{}^\dagger N_{-\mathbf{k}',-\omega'})_{n_2n_4,n_2n_4}\rangle. \quad (18)$$

Equation (18) is the quantum analog of the classical formula[7]

$$\frac{d^2\sigma}{d\omega d\Omega}\sim\langle|N_{-\mathbf{k},\omega}|^2\rangle.$$

It is actually valid for the scattering of a photon from any system (provided that the $\mathbf{A}\cdot\mathbf{j}$ term in the interaction is neglected[2]), if $N_{-\mathbf{k}',-\omega'}$ is defined as in (15), i.e.,

$$N_{-\mathbf{k}',-\omega'}=\int_{-\infty}^{\infty}e^{(i/\hbar)H_st}\rho_{-\mathbf{k}'}e^{-(i/\hbar)H_st}e^{-i\omega' t}dt', \quad (19)$$

where H_s is the Hamiltonian of this system. If $H_s=H_{s0}+H_s'$ where H_{s0} is the unperturbed Hamiltonian and H_s' is the interaction, then (19) can be reduced as before to (15) with $H_{s0}=H_0$, $H_s'=H_1$.

Equation (16) can be generalized in the following way. Let us consider the scattering of a system a from a system b, with the total Hamiltonian

$$H=H_a+H_b+H',$$
$$H'=\textstyle\sum_m A_m{}^\dagger B_m,$$

where H_a+H_b is the unperturbed Hamiltonian and H' is the interaction between systems a and b, and where $A_m{}^\dagger$, B_m are operators of systems a and b, respectively. The probability in the lowest order of H' of transition of system a from state $|i_a\rangle$ to state $|f_a\rangle$, after summing over the final states and averaging over the initial states of b, is given by

$$P=\frac{1}{\hbar^2}\sum_{m,n,i_b}\langle i_a|A_m|f_a\rangle\langle f_a|A_n{}^\dagger|i_a\rangle\times\int_{-\infty}^{\infty}dt\int_{-\infty}^{\infty}dt'e^{-(i/\hbar)(E_{af}-E_{ai})(t-t')}\times\langle i_b|B_m{}^\dagger(t)B_n(t')|i_b\rangle\sigma_{i_bi_b}, \quad (20a)$$

where σ is the density matrix for system b at time $t=-\infty$; E_{af} and E_{ai} are the energies of states $|f_a\rangle$ and $|i_a\rangle$, respectively;

$$B_m(t)=e^{(i/\hbar)H_bt}B_me^{-(i/\hbar)H_bt},$$

and $|i_b\rangle$ is the wave function of system b at time $t=-\infty$ in the interaction representation with H_b considered as the unperturbed Hamiltonian.

If σ is diagonal with respect to the eigenstates $|i_b\rangle$ of H_b, then (20a) is reduced to

$$\text{Transition Rate}=\frac{1}{\hbar^2}\sum_{m,n}\langle i_a|A_m|f_a\rangle\langle f_a|A_n{}^\dagger|i_a\rangle\times\int_{-\infty}^{\infty}dte^{-i\omega t}\langle B_m{}^\dagger(t)B_n(0)\rangle, \quad (20b)$$

where

$$\langle B_m{}^\dagger(t)B_n(0)\rangle\equiv\mathrm{Tr}(B_m{}^\dagger(t)B_n(0)\sigma),$$

and

$$\hbar\omega=E_{af}-E_{ai}.$$

However, if $H_b=H_{b0}+H_b'$, where H_b' is a perturbing potential and the density matrix σ of the system b at time $t=-\infty$ is diagonal with respect to the eigenfunc-

[6] H. Cheng (to be published).

[7] See, for example, M. N. Rosenbluth and N. Rostoker, Phys. Fluids 5, 776 (1962).

tions of H_{b0}, then (20a) can be reduced by a procedure similar to that used in deriving (14) to the following form:

$$P=\frac{1}{\hbar^2}\sum_{m,n}\langle i_a|A_m|f_a\rangle\langle f_a|A_n^\dagger|i_a\rangle\langle B_m^\dagger(\omega)B_n(\omega)\rangle, \quad (20c)$$

where

$$B_n(\omega)=\int_{-\infty}^{\infty}dte^{i\omega t}\times\left\{T\left(\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t}H_b'(t')dt'\right]\right)\right\}^{-1}B_n^I(t)\times T\left(\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t}H_b'(t')dt'\right]\right),$$

with

$$B_n^I(t)=e^{(i/\hbar)H_{b0}t}B_ne^{-(i/\hbar)H_{b0}t},$$

and

$$H_b'(t)=e^{(i/\hbar)H_{b0}t}H_b'e^{-(i/\hbar)H_{b0}t}.$$

III. LIGHT-LIGHT SCATTERING AND OPTICAL MIXING IN THE LOWEST ORDER

In order to evaluate explicitly the transition probability of photons 1 and 2 into 3 and 4 from (10), we need to evaluate expressions like

$$\langle\rho_1(t_1)\rho_2(t_2)\rho_3(t_3)\rho_4(t_4)\rangle.$$

We shall make the following approximation:

$$\langle\rho_1(t_1)\rho_2(t_2)\rho_3(t_3)\rho_4(t_4)\rangle=\langle\rho_1(t_1)\rho_2(t_2)\rangle\langle\rho_3(t_3)\rho_4(t_4)\rangle+\langle\rho_1(t_1)\rho_3(t_3)\rangle\langle\rho_2(t_2)\rho_4(t_4)\rangle+\langle\rho_1(t_1)\rho_4(t_4)\rangle\langle\rho_2(t_2)\rho_3(t_3)\rangle. \quad (21)$$

In doing so we have included only two-particle correlation. In other words, only "bubble" diagrams are taken into account.[8] This approximation is consistent with the random-phase approximation.

With the approximation (21), we have

$$\langle\{T(\rho_{\mathbf{k}}(t_1)\rho_{-\mathbf{k}'}(t_2))\}^\dagger T(\rho_{\mathbf{k}}(t_3)\rho_{-\mathbf{k}'}(t_4))\rangle=\langle\{T(\rho_{\mathbf{k}}(t_1)\rho_{-\mathbf{k}'}(t_2))\}^\dagger\rangle\langle T(\rho_{\mathbf{k}}(t_3)\rho_{-\mathbf{k}'}(t_4))\rangle+\langle\rho_{\mathbf{k}}^\dagger(t_1)\rho_{-\mathbf{k}'}(t_4)\rangle\langle\rho_{-\mathbf{k}'}^\dagger(t_2)\rho_{\mathbf{k}}(t_3)\rangle+\langle\rho_{\mathbf{k}}^\dagger(t_1)\rho_{\mathbf{k}}(t_3)\rangle\langle\rho_{-\mathbf{k}'}^\dagger(t_2)\rho_{-\mathbf{k}'}(t_4)\rangle. \quad (22)$$

Substituting (22) into (10), performing the time integrations and dividing (10) by the total interaction time τ, we obtain the transition rate for light-light scattering as

$$\text{Transition Rate}=\frac{(2\pi)^4e^8}{m^4V^2}\cdot\frac{(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2}{\omega_1\omega_2\omega_3\omega_4}[\delta_{\mathbf{k},\mathbf{k}'}2\pi\delta(\omega-\omega')|S_T(\mathbf{k}',\omega)|^2+\delta_{\mathbf{k},-\mathbf{k}'}2\pi\delta(\omega+\omega')|S(\mathbf{k}',\omega')|^2]. \quad (23)$$

Accordingly, the differential cross section is given by

$$\frac{d^2\sigma}{d\Omega_3d\omega_3}=\frac{1}{c^4}\frac{\omega_3}{\omega_1\omega_2}\frac{(2\pi)^2e^8}{m^4}(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2\left[\frac{\delta(\omega_1+\omega_2-\omega_3-|\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3|c)}{(\omega_1+\omega_2-\omega_3)}|S_T(\mathbf{k}_1-\mathbf{k}_3,\omega_1-\omega_3)|^2+\frac{\delta(-\omega_1+\omega_2+\omega_3-|\mathbf{k}_2+\mathbf{k}_3-\mathbf{k}_1|c)}{(\omega_2+\omega_3-\omega_1)}|S(\mathbf{k}_1-\mathbf{k}_3,\omega_1-\omega_3)|^2\right]. \quad (24)$$

Integrating over ω_3, the elastic[1] and inelastic differential cross sections are given by

$$\frac{d\sigma_{\text{elas}}}{d\Omega_3}=\frac{1}{c^4}\frac{1}{\omega_1\omega_2}\frac{(2\pi)^2e^8}{m^4}(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2\frac{\omega_3^2|S_T(\mathbf{k}_1-\mathbf{k}_3,\omega_1-\omega_3)|^2}{\omega_3(\omega_1+\omega_2)-(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{k}_3c^2}, \quad (25)$$

with

$$\omega_3=\omega_1+\omega_2-|\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3|c,$$

and

$$\frac{d\sigma_{\text{inelas}}}{d\Omega_3}=\frac{1}{c^4}\frac{1}{\omega_1\omega_2}\frac{(2\pi)^2e^8}{m^4}\frac{(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2\omega_3^2|S(\mathbf{k}_1-\mathbf{k}_3,\omega_1-\omega_3)|^2}{|\omega_3(\omega_2-\omega_1)-(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{k}_3c^2|}, \quad (26)$$

with

$$\omega_3=\omega_1-\omega_2+|\mathbf{k}_2+\mathbf{k}_3-\mathbf{k}_1|c.$$

The first and the second terms in (23) and (24) come from the first and the second terms in (22). The third term in (22), corresponding to two independent incoherent scatterings, is neglected.[1,2] In (23), τ is the total time of interaction, and

$$S_T(\mathbf{k},\omega)\equiv\int_{-\infty}^{\infty}dte^{i\omega t}\langle T(\rho_{\mathbf{k}}(t)\rho_{-\mathbf{k}}(0))\rangle, \quad (27)$$

$$S(\mathbf{k},\omega)\equiv\int_{-\infty}^{\infty}dte^{i\omega t}\langle\rho_{\mathbf{k}}(t)\rho_{-\mathbf{k}}(0)\rangle. \quad (28)$$

[8] M. Gell-Mann and K. A. Brueckner, Phys. Rev. **106**, 364 (1957).

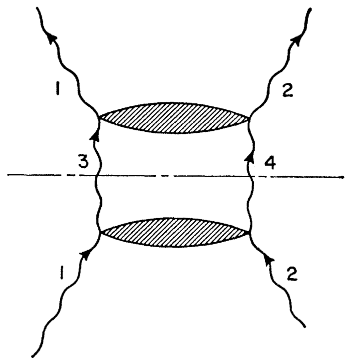

FIG. 1. Elastic contribution to the light-light scattering cross section. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

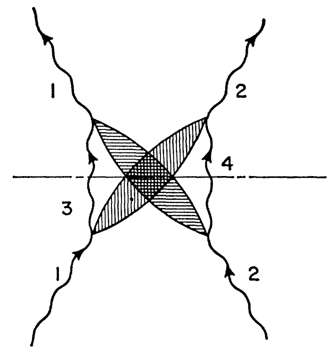

FIG. 2. Inelastic contribution to the light-light scattering cross section. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

In deriving (23), we have made use of the relations

$$\rho_{\mathbf{k}}^\dagger(t)=\rho_{-\mathbf{k}}(t)\,, \tag{29}$$

and

$$\langle\rho_{\mathbf{k}}(t_1)\rho_{-\mathbf{k}'}(t_2)\rangle=\delta_{\mathbf{k},\mathbf{k}'}\langle\rho_{\mathbf{k}}(t_1)\rho_{-\mathbf{k}}(t_2)\rangle\,. \tag{30}$$

The correlation functions $S_T(\mathbf{k},\omega)$ and $S(\mathbf{k},\omega)$ can be expressed in terms of the longitudinal plasma dielectric function $\epsilon(\mathbf{k},\omega)$ in the well-known way:

$$S(\mathbf{k},\omega)=\frac{-2\hbar}{1-e^{-\beta\omega\hbar}}\frac{\mathbf{k}^2}{4\pi e^2}\,\mathrm{Im}\left(\frac{1}{\epsilon(\mathbf{k},\omega)}\right) \tag{31}$$

and

$$S_T(\mathbf{k},\omega)=\frac{\mathbf{k}^2\hbar}{4\pi e^2}\left[-\coth\frac{\beta\hbar\omega}{2}\,\mathrm{Im}\left(\frac{1}{\epsilon(\mathbf{k},\omega)}\right)+i\,\mathrm{Re}\left(\frac{1}{\epsilon(\mathbf{k},\omega)}-1\right)\right]. \tag{32}$$

To help understand the nature of the approximation made in (22), we express the left-hand side of (22) by "diagrams." Take the first term in the right-hand side of (22) as an example. The factor $\rho_{\mathbf{k}}(t_3)$ and $\rho_{-\mathbf{k}'}(t_4)$ correspond to the creation of electron-hole pairs at t_3 and t_4 respectively, and are represented in Fig. 1 by the two lower vertices. Upon taking the statistical average of $T(\rho_{\mathbf{k}}(t_3)\rho_{-\mathbf{k}'}(t_4))$, only those diagrams in which the two electron-hole pairs are connected remain. Thus the lower half of Fig. 1 corresponds to the factor $\langle T(\rho_{\mathbf{k}}(t_3)\rho_{-\mathbf{k}'}(t_4))\rangle$. Similarly, the upper half of Fig. 1 corresponds to the factor $\langle\{T(\rho_{\mathbf{k}}(t_1)\rho_{-\mathbf{k}'}(t_2)\}^\dagger\rangle$. From (30) it is seen that this term represents the contribution from elastic light-light scattering; this is reflected by the fact that the center line (the dash-dot line) does not cut through any electron-hole pairs, which means that the final state of the plasma is the same as its initial state. Similarly, Figs. 2 and 3 correspond to the second and the third terms on the right side of (22). This means that the approximation made in (22) takes into account the diagrams in Figs. 1, 2, and 3.

At a zero of $\mathrm{Re}\epsilon(\mathbf{k},\omega)$, a resonance in the scattering occurs. The first term in the right side of (23) corresponds to elastic scattering with the plasma state unchanged. The second term in the right-hand side of (23) corresponds to inelastic scattering, each photon receiving an energy transfer ω and momentum transfer $\mathbf{k}$ from the plasma. These two terms are of the same order of magnitude at resonance.

If we consider the scattering of photon 1 to photon 3, and sum over the final states of the photons 2 and 4, then the transition probability is expressed by (11). To obtain P to the order e^8, we need to calculate M up to the order e^6. Write

$$M=M_1+M_2+M_3\,, \tag{33}$$

then

$$P=\langle(M_1^\dagger M_1)_{n_2n_4,n_2n_4}\rangle+\langle(M_2^\dagger M_2+M_1^\dagger M_3+M_3^\dagger M_1)_{n_2n_4,n_2n_4}\rangle\,, \tag{34}$$

where M_1, M_2, and M_3 are terms proportional to e^2, e^4, and e^6, respectively, in the expansion of (12). The Feynman diagrams for M_1, M_2, M_3 are illustrated in Figs. 4, 5(a), 5(b), 6(a), and 6(b). Note that they are Feynman diagrams written for M itself and not for ensemble average value like those in Figs. 1, 2, and 3. Since the expectation value $(M^\dagger M)_{n_2n_4,n_2n_4}$ with respect to the initial photon states 2 and 4 is taken, only those products in $M^\dagger M$ which have the same number of $a_4^\dagger(a_2^\dagger)$ and $a_4(a_2)$ contribute. The term $M_1^\dagger M_1$ is illustrated in Fig. 7 and is given by

$$\langle(M_1^\dagger M_1)_{n_2n_4,n_2n_4}\rangle=(C_{13}^2/\hbar^2)S(\mathbf{k}',\omega')\tau\,, \tag{35}$$

where τ is the total interaction time. The contributions of $\langle(M_{2\mathrm{a}}^\dagger M_{2\mathrm{a}})_{n_2n_4,n_2n_4}\rangle$ and $\langle(M_{2\mathrm{b}}^\dagger M_{2\mathrm{b}})_{n_2n_4,n_2n_4}\rangle$ are

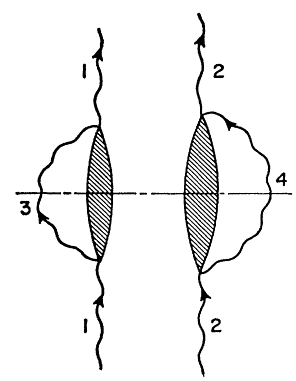

FIG. 3. Double incoherent scattering. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

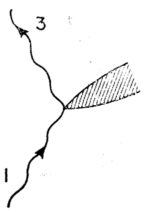

FIG. 4. Lowest order Feynman diagram depicting single incoherent scattering.

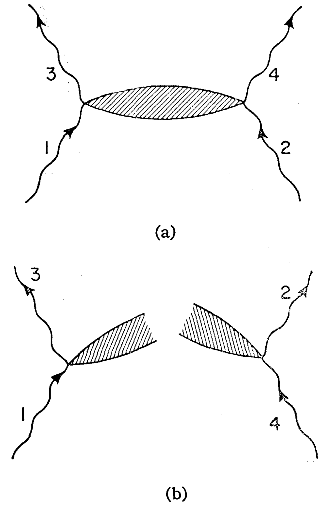

FIG. 5(a) Feynman diagram depicting elastic light-light scattering in the lowest order. The plasma state is unchanged. (b) Feynman diagram depicting inelastic light-light scattering in the lowest order. Two plasmons are created.

illustrated in Figs. 1 and 8, respectively, and are given by

$$\langle (M_{2a}{}^\dagger M_{2a})_{n_2n_4,n_2n_4}\rangle = (C_{13}{}^2C_{24}{}^2/\hbar^4) \times n_2n_4\tau\,|S_T(\mathbf{k}',\omega')|^2\delta_{\mathbf{k}',\mathbf{k}}2\pi\delta(\omega-\omega'), \tag{36}$$

and

$$\langle (M_{2b}{}^\dagger M_{2b})_{n_2n_4,n_2n_4}\rangle = (C_{13}{}^2C_{24}{}^2/\hbar^4) \times n_2n_4\tau S^2(\mathbf{k},\omega)\delta_{\mathbf{k}',\mathbf{k}}2\pi\delta(\omega-\omega'). \tag{37}$$

In obtaining (36) and (37), uses have been made of the relations

$$S_T(\mathbf{k},\omega)=S_T(-\mathbf{k},-\omega), \tag{38}$$

$$S^*(\mathbf{k},\omega)=S(\mathbf{k},\omega). \tag{39}$$

The contributions of

$$\langle (M_1{}^\dagger M_{3a})_{n_2n_4,n_2n_4}\rangle,\quad \langle (M_1{}^\dagger M_{3b})_{n_2n_4,n_2n_4}\rangle,$$
$$\langle (M_{3a}{}^\dagger M_1)_{n_2n_4,n_2n_4}\rangle,\quad \langle (M_{3b}{}^\dagger M_1)_{n_2n_4,n_2n_4}\rangle$$

are illustrated in Figs. 9, 10, 11, and 12, respectively. They are given by

$$\langle (M_1{}^\dagger M_{3a})_{n_2n_4,n_2n_4}\rangle+\langle (M_1{}^\dagger M_{3b})_{n_2n_4,n_2n_4}\rangle = \frac{C_{13}{}^2C_{24}{}^2n_2n_4}{\hbar^4}\tau S(\mathbf{k},\omega)S_T(\mathbf{k},\omega)\delta_{\mathbf{k}',\mathbf{k}}2\pi\delta(\omega-\omega'), \tag{40}$$

and

$$\langle (M_{3a}{}^\dagger M_1)_{n_2n_4,n_2n_4}\rangle+\langle (M_{3b}{}^\dagger M_1)_{n_2n_4,n_2n_4}\rangle = \frac{C_{13}{}^2C_{24}{}^2n_2n_4}{\hbar^4}\tau S_T{}^*(\mathbf{k},\omega)S(\mathbf{k},\omega)\delta_{\mathbf{k},\mathbf{k}'}2\pi\delta(\omega-\omega'). \tag{41}$$

From (34), (35), (36), (37), (40), and (41), we obtain

$$\frac{P}{\tau}=\frac{(2\pi)^2e^4}{m^2V}\frac{(\hat{e}_1\cdot\hat{e}_3)^2}{\omega_1\omega_3}S(\mathbf{k}',\omega')+\frac{(2\pi)^4e^8}{m^4V^2}\frac{(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2}{\omega_1\omega_2\omega_3\omega_4}n_2n_4\delta_{\mathbf{k}'\mathbf{k}}2\pi\delta(\omega-\omega')\,|S(\mathbf{k},\omega)+S_T(\mathbf{k},\omega)|^2. \tag{42}$$

Expressing $S(\mathbf{k},\omega)$ and $S_T(\mathbf{k},\omega)$ in terms of $\epsilon(\mathbf{k},\omega)$ by use of (31) and (32), we get

$$\frac{P}{\tau}=\frac{\pi e^2}{m^2V}\frac{(\hat{e}_1\cdot\hat{e}_3)^2}{\omega_1\omega_3}\frac{-2\hbar}{1-e^{-\beta\omega\hbar}}\mathbf{k}^2\,\mathrm{Im}\left(\frac{1}{\epsilon(\mathbf{k},\omega)}\right)+\frac{\pi^2e^4}{m^4V^2}\frac{(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2}{\omega_1\omega_2\omega_3\omega_4}n_2n_4\hbar^2\mathbf{k}^4\delta_{\mathbf{k}',\mathbf{k}}2\pi\delta(\omega-\omega')\left|\frac{1-\epsilon(\mathbf{k},\omega)}{\epsilon(\mathbf{k},\omega)}\right|^2. \tag{43}$$

The differential cross section for scattering of photon 1 to 3 *per unit volume of interaction is given as*

$$\frac{d^2\sigma}{d\omega_3d\Omega_3}=\frac{e^2(\hat{e}_1\cdot\hat{e}_3)^2}{4\pi^2c^4m^2}\frac{\hbar\mathbf{k}^2}{1-e^{-\beta\omega\hbar}}\frac{\omega_3}{\omega_1}\,\mathrm{Im}\left(\frac{1}{\epsilon(\mathbf{k},\omega)}\right)+\frac{e^4\pi^2}{c^4m^4}\frac{(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2}{\omega_1\omega_2\omega_4}\omega_3\frac{n_2n_4}{V^2}\hbar^2\mathbf{k}^4\delta(\mathbf{k}'-\mathbf{k})2\pi\delta(\omega-\omega')\left|\frac{1-\epsilon(\mathbf{k},\omega)}{\epsilon(\mathbf{k},\omega)}\right|^2. \tag{44}$$

Integrating (44) with respect to ω_3 gives

$$\frac{d\sigma}{d\Omega_3}=\frac{e^2(\hat{e}_1\cdot\hat{e}_3)^2}{4\pi^2c^4m^2}\int_0^\infty d\omega_3\frac{\hbar(\mathbf{k}_3-\mathbf{k}_1)^2}{1-e^{-\beta\hbar(\omega_3-\omega_1)}}\frac{\omega_3}{\omega_1}\,\mathrm{Im}\left(\frac{1}{\epsilon(\mathbf{k}_3-\mathbf{k}_1,\,\omega_3-\omega_1)}\right)$$
$$+\frac{2e^4\pi^3}{c^4m^4}\frac{(\hat{e}_1\cdot\hat{e}_3)^2(\hat{e}_2\cdot\hat{e}_4)^2}{\omega_1\omega_2\omega_4}(\omega_1+\omega_2-\omega_4)\frac{n_2n_4}{V^2}\hbar^2\mathbf{k}^4\delta^3(\mathbf{k}'-\mathbf{k})\left|\frac{1-\epsilon(\mathbf{k},\omega)}{\epsilon(\mathbf{k},\omega)}\right|^2, \tag{45}$$

where the condition

$$\omega_1+\omega_2-\omega_4=c\,|\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_4|$$

has to be satisfied. Since the roles of beams 2 and 4 can be interchanged, there is also a term for $d\sigma/d\Omega_3$ exactly the same as the right-hand side of (45) with $\omega,\mathbf{k}$ replaced by $-\omega,-\mathbf{k}$.

IV. OPTICAL MIXING OF STRONG BEAMS IN THE PLASMA

In this section we shall be concerned with the scattering from the combined system consisting of the plasma and laser beams 2 and 4. Our treatment will be based on the second viewpoint. This is especially motivated by the considerations below.

If the numbers of photons in beams 2 and 4 are n_2 and n_4, respectively, the probability of light-light scattering in the lowest order is equal to the product of $n_2 n_4$ and the right-hand side of (23). Comparing the expression so obtained with (42), we find that, at resonance when $\text{Re}\,\epsilon(\mathbf{k},\omega)=0$, the elastic term in the former is equal to the product of $\tanh^2(\beta\omega\hbar/2)$ (which is less than one) and the e^8 term in the latter, provided that (32) is taken into account. Now, (42) is the transition probability of photon 1 to photon 3 including contributions from all final states of the other photon. Since the contribution from any final state should be positive, it may appear surprising that the total probability of transition to all channels is smaller than that to one of the channels, i.e., the elastic light-light scattering channel.

To answer this we first note that in (42), we have included the channel of incoherent scattering up to the order e^8, in addition to the channels of light-light scattering. If the first term (proportional to e^4) in (42) is much larger than its second term (proportional to e^8), so that the perturbation expansion is justified, then the total probability given by (42) is indeed greater than the probability to the light-light scattering channels alone. If, however, the beam intensities n_2 and n_4 are so strong[1] that in (42) the e^8 term dominates over the e^4 term, the probability of transition to the incoherent scattering channel (Fig. 7 +Fig. 9+Fig. 10+Fig. 11+Fig. 12) becomes negative, if terms of orders higher than e^8 are not included. This clearly points out that the higher order terms are important. Neglect of the higher order terms is then unjustified.

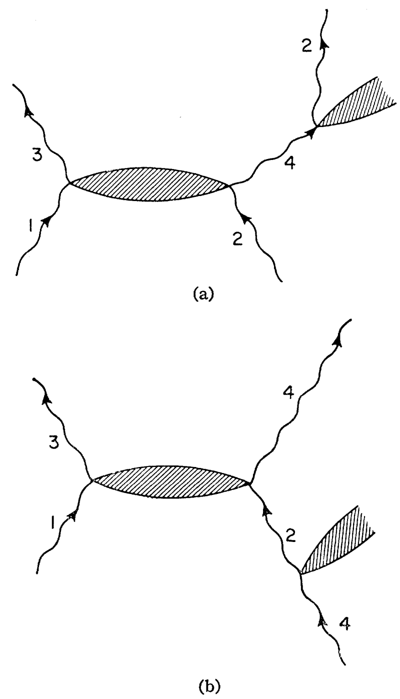

FIG. 6(a) A higher order diagram of incoherent scattering. (b) Another higher order diagram of incoherent scattering.

The fact that in the e^8 order the total probability becomes smaller than the probability to a certain channel does not mean that the same is true to all orders. This only makes it imperative to take all higher order terms into account in the interesting cases when the lowest order light-light scattering probability is greater than the lowest order incoherent scattering probability.

It appears difficult to apply (11) and (12) to calculate P to all orders. We found that (16) and (15) are especially convenient for this purpose.

When the beams 2 and 4 are strong, we may treat them as classical fields and disregard the commutation relations governing the photon operators. Thus, in (25), the photon fields can be taken out of the multiple commutators which now involve just the electron-density operators ρ:

$$N_{-\mathbf{k}',-\omega'}=\int_{-\infty}^{\infty}\rho_{-\mathbf{k}'}(t)e^{-i\omega' t}dt+\frac{i}{\hbar}C_{24}(n_2n_4)^{1/2}\int_{-\infty}^{\infty}dte^{-i\omega' t}\int_{-\infty}^{t}dt_1[e^{-i\omega t_1}\rho_{-\mathbf{k}}(t_1)+e^{i\omega t_1}\rho_{\mathbf{k}}(t_1),\,\rho_{-\mathbf{k}'}(t)]+\left(\frac{i}{\hbar}C_{24}(n_2n_4)^{1/2}\right)^2$$
$$\times\int_{-\infty}^{\infty}dte^{-i\omega' t}\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2[e^{-i\omega t_2}\rho_{-\mathbf{k}}(t_2)+e^{i\omega t_2}\rho_{\mathbf{k}}(t_2),\,[e^{-i\omega t_1}\rho_{-\mathbf{k}}(t_1)+e^{i\omega t_1}\rho_{\mathbf{k}}(t_1),\,\rho_{-\mathbf{k}'}(t)]]+\cdots. \quad (46)$$

Notice that the expansion parameter is now $C_{24}(n_2n_4)^{1/2}$. To evaluate $\langle N_{-\mathbf{k}',-\omega'}{}^{\dagger}N_{-\mathbf{k}',-\omega'}\rangle$, one again only takes care of two particle correlation as in (21). In other words, one breaks up the statistical average of a product of many ρ's into products of the statistical average of pairs of ρ's in the following manner:

$$\langle\rho_{\mathbf{k}_1}\rho_{\mathbf{k}_2}\cdots\rho_{\mathbf{k}_{2n-1}}\rho_{\mathbf{k}_{2n}}\rangle=\langle\rho_{\mathbf{k}_1}\rho_{\mathbf{k}_2}\rangle\cdots\langle\rho_{\mathbf{k}_{2n-1}}\rho_{\mathbf{k}_{2n}}\rangle+\text{all other possible permutations.} \quad (47)$$

Also,

$$\langle\rho_{\mathbf{k}_1}\rho_{\mathbf{k}_2}\cdots\rho_{\mathbf{k}_{2n+1}}\rangle=0\,, \quad (48)$$

since

$$\langle\rho_{\mathbf{k}_i}\rangle=0\,,\quad\text{for}\quad\mathbf{k}_i\neq0$$

and since all the $\mathbf{k}_i$'s involved in (46) are not equal to zero.

Making use of (48), we obtain from (46)

$$\langle N_{-\mathbf{k}',-\omega'}{}^\dagger N_{-\mathbf{k}',-\omega'}\rangle=\int_{-\infty}^{\infty}dt\int_{-\infty}^{\infty}dt'\langle\rho_{-\mathbf{k}'}{}^\dagger(t')\rho_{-\mathbf{k}'}(t)\rangle e^{i\omega'(t'-t)}+\left(\frac{C_{24}(n_2n_4)^{1/2}}{\hbar}\right)^2\int_{-\infty}^{\infty}dt'e^{i\omega't'}\int_{-\infty}^{\infty}dte^{-i\omega't}$$
$$\times\int_{-\infty}^{t'}dt_1'\int_{-\infty}^{t}dt_1\langle[\rho_{-\mathbf{k}'}{}^\dagger(t'),\,e^{i\omega t_1'}\rho_{-\mathbf{k}}{}^\dagger(t_1')+e^{-i\omega t_1'}\rho_{\mathbf{k}}{}^\dagger(t_1')][e^{-i\omega t_1}\rho_{-\mathbf{k}}(t_1)+e^{i\omega t_1}\rho_{\mathbf{k}}(t_1),\,\rho_{-\mathbf{k}'}(t)]\rangle+\cdots. \quad (49)$$

Substitution of the first term in the right-hand side of (49) into (16) gives rise to the first term in (42). To evaluate the rest of the terms in the right side of (49), we first make use of (47) to obtain

$$\langle\rho_1\rho_2[\rho_3\rho_4]\rangle=\langle\rho_2\rho_2\rangle\langle[\rho_3,\rho_4]\rangle. \quad (50)$$

Note that in (50), there is no pairing of any ρ inside the commutator with any ρ outside of the commutator, since all such pairings give vanishing result. For example, if ρ_3 is paired with ρ_1, then after the pairing is made, the relative order of ρ_3 to ρ_4 in the commutator becomes irrelevant, and therefore the two terms in the commutator $[\rho_3,\rho_4]$ cancel each other. This argument can be extended to give

$$\langle\rho_1\rho_2\cdots\rho_n[\rho_{n+1},[\rho_{n+2},\cdots[\rho_{m+n-1},\rho_{n+m}]\cdots]]\rangle$$
$$=\langle\rho_1\rho_2\cdots\rho_n\rangle\langle[\rho_{n+1},[\rho_{n+2},\cdots[\rho_{n+m-1},\rho_{n+m}]\cdots]]\rangle. \quad (51)$$

One then has

$$\langle[\rho_1,[\rho_2\cdots[\rho_{n-1}]\rho_n]\cdots]]$$
$$\times[\rho_{n+1},[\rho_{n+2},\cdots[\rho_{n+m-1},\rho_{n+m}]\cdots]]\rangle$$
$$=\langle[\rho_1,[\rho_2\cdots[\rho_{n-1},\rho_n]\cdots]]\rangle$$
$$\times\langle[\rho_{n+1},[\rho_{n+2},\cdots[\rho_{n+m-1},\rho_{n+m}]\cdots]]\rangle. \quad (52)$$

Furthermore, by similar arguments, one has

$$\langle[\rho_1,[\rho_2\cdots[\rho_{n-1},\rho_n]\cdots]]\rangle=0 \quad \text{when } n>2. \quad (53)$$

Making use of (51), (52), and (53), one finds that the only nonvanishing terms in the right side of (49) are the two lowest order terms explicitly given there. Thus, we have

$$\langle N_{-\mathbf{k}',-\omega}{}^\dagger N_{-\mathbf{k}',-\omega'}\rangle=\langle\rho_{-\mathbf{k}',-\omega'}{}^\dagger\rho_{-\mathbf{k}',-\omega'}\rangle$$
$$+\frac{C_{24}{}^2n_2n_4}{\hbar^2}\left|\int_{-\infty}^{\infty}dte^{-i\omega't}\int_{-\infty}^{t}dt_1\right.$$
$$\left.\times\langle[\mathrm{e}^{-i\omega t_1}\rho_{-\mathbf{k}}(t_1)+e^{i\omega t_1}\rho_{\mathbf{k}}(t_1),\,\rho_{-\mathbf{k}'}(t)]\rangle\right|^2, \quad (54)$$

where

$$\rho_{-\mathbf{k}',-\omega'}=\int_{-\infty}^{\infty}dt\rho_{-\mathbf{k}'}(t)\mathrm{e}^{-i\omega't}. \quad (55)$$

Equation (54), together with (16), gives exactly the same result as (42) or (43). This was not anticipated since (42) or (43) includes H_1 only in the lowest orders, while (54) includes H_1 to all orders. The agreement of these two calculations is due to the exact cancellation of all terms of orders higher than e^8. For example, in the approximation (47), Eq. (46) leads to

$$\langle N_{-\mathbf{k}',-\omega'}\rangle+\frac{C_{24}(n_2n_4)^{1/2}}{\hbar}\int_{-\infty}^{\infty}dte^{-i\omega't}\int_{-\infty}^{t}dt_1$$
$$\times\langle[e^{-i\omega t_1}\rho_{-\mathbf{k}}(t_1)+e^{i\omega t_1}\rho_{\mathbf{k}}(t_1),\,\rho_{-\mathbf{k}'}(t)]\rangle, \quad (56)$$

which means that after statistical average is taken, only the second term of the perturbation expansion in (46) remains. Equation (54) can be rewritten as

$$\langle N_{-\mathbf{k}',-\omega'}{}^\dagger N_{-\mathbf{k}',-\omega'}\rangle=\langle\rho_{-\mathbf{k}',-\omega'}{}^\dagger\rho_{-\mathbf{k}',-\omega'}\rangle$$
$$+\langle N_{-\mathbf{k}',-\omega'}{}^\dagger\rangle\langle N_{-\mathbf{k}',-\omega'}\rangle. \quad (57)$$

Equation (57) is the formula implicitly used in Ref. 3 in which $\langle N_{-\mathbf{k}',-\omega'}\rangle$ is calculated only to the lowest order. Our investigation explains why (54) and (16) agree with the result in Ref. 3. The agreement, however, can not be expected *a priori*. In fact, as was pointed out in the beginning of this section, the lowest order perturbation gives rise to a negative probability for scattering into the incoherent channel, when the beams 2 and 4 are sufficiently strong. Our discussion on the cancellation of higher order terms means that the higher order correction to the probability of incoherent scattering is positive for strong beams. This in turn is exactly cancelled by the negative higher order corrections to other processes. The probability of transition to any channel is actually positive.

V. A SIMPLE MODEL

In the calculation of the probability of scattering of photon 1 to photon 3 by the combined system consisting of the plasma and the light beams 2 and 4 it was not anticipated that we could obtain $\langle N_{-\mathbf{k}'-\omega}{}^\dagger N_{-\mathbf{k}',-\omega'}\rangle$, accurate to all orders in H_1, by the use of Eq. (57) in which $\langle N_{-\mathbf{k}',-\omega'}\rangle$ is calculated to the lowest order in H_1.

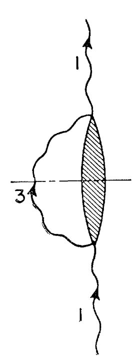

FIG. 7. Contribution of $\langle M_1{}^\dagger M_1\rangle$, i.e., single incoherent scattering. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors $S_{fi}{}^*$ and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

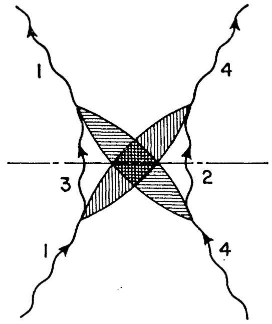

FIG. 8. Inelastic contribution to light-light scattering. This is the same as Fig. 2 with the photons 2 and 4 interchanged. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

Our proof in Sec. IV that all higher order terms in H_1 do not contribute to $\langle N_{-\mathbf{k}',-\omega'}{}^\dagger N_{-\mathbf{k}',-\omega'}\rangle$ was based on the approximations (47) and (48). In the present section we shall demonstrate that, in a simple model, Eq. (57) is an exact relation in which $\langle N_{-\mathbf{k}',-\omega'}\rangle$ is indeed proportional to H_1.

Let us consider a plasmon as the quantum of a harmonic oscillator, which is linearly coupled to a system of phonons. This coupling gives rise to a shift and a width of the plasma frequency. When such a system is put under the influence of photon fields 1, 2, 3, and 4, the total Hamiltonian is given by

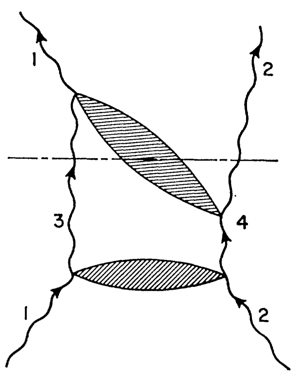

FIG. 9. Contribution of $\langle M_1{}^\dagger M_{3a}\rangle$. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

$$H=H_0+H_1+H_2, \quad (58)$$

$$H_0=\omega_0 a^\dagger a+\sum_j \omega_j b_j{}^\dagger b_j+\sum_j k_j{}^* a b_j{}^\dagger$$
$$+\sum_j k_j a^\dagger b_j+\text{free radiation fields}, \quad (59)$$

$$H_1=e_{24}a^\dagger+e_{24}{}^*a, \quad (60)$$

$$H_2=e_{13}a^\dagger+e_{13}{}^*a. \quad (61)$$

In the Hamiltonians, a, $a^\dagger$ are the annihilation and creation operators for the plasmon; b_j, $b_j{}^\dagger$ are the annihilation and creation operators for the phonons. They obey the usual boson commutation relations. The Hamiltonians H_1 and H_2 of Eqs. (60) and (61), which represent, respectively, the interaction of the plasmon with photons 2, 4 and photons 1, 3, correspond to the Hamiltonians defined in Eqs. (2) and (3). When the photon fields are quantized, $e_{ij}=C_{ij}A_i{}^\dagger A_j$, where A, $A^\dagger$ represent the photon annihilation and creation operators. In the limit of strong beams, the radiation fields 2, 4 can be treated as classical fields and e_{24} becomes $e_{24}(t)$, which is just a c number depending on time explicitly.

When we consider the scattering of photon 1 to photon 3 by the combined system described by H_0+H_1 due to the interaction H_2, the relevant quantity to be calculated is $\mathrm{Tr}[a^\dagger(t)a(t')e^{-\beta H_0}]$, where $a(t)$ evolves in time according to the Hamiltonian H_0+H_1 [see Eqs. (16) and (15)]. To calculate this quantity, we first

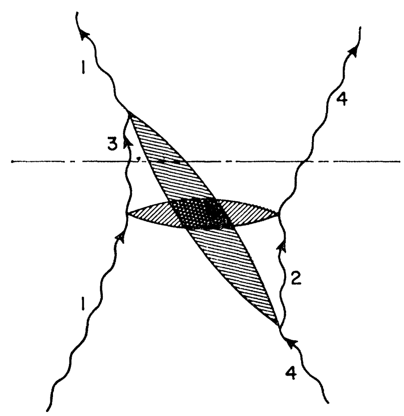

FIG. 10. Contribution of $\langle M_1{}^\dagger M_{3b}\rangle$. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

solve for $a(t)$. The equations of motion are

$$i\frac{da(t)}{dt}=[a,H_0+H_1]=\omega_0 a(t)$$
$$+\sum_j k_j b_j(t)+e_{24}(t), \quad (62)$$

$$i(db_j(t)/dt)=[b,H_0+H_1]=\omega_j b_j(t)+k_j{}^\dagger a(t), \quad (63)$$

with the initial conditions $a(t_0)=a_0$, $b_j(t_0)=b_{j0}$, where t_0 is the time when e_{24} is turned on, a_0, b_{j0} are operators

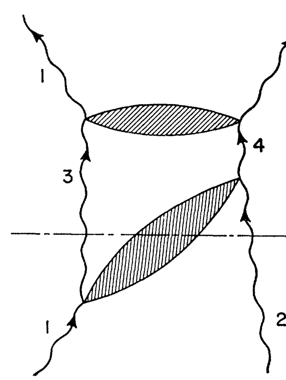

FIG. 11. Contribution of $\langle M_{3a}{}^\dagger M_1\rangle$. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors S_{fi}^* and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

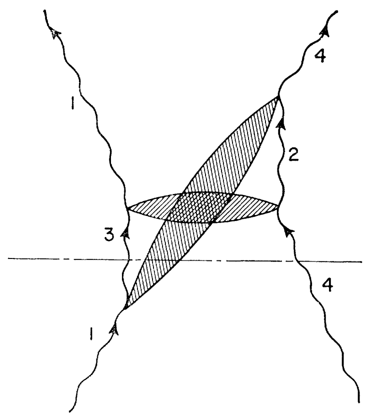

FIG. 12. Contribution of $\langle M_{3b}{}^{\dagger} M_1 \rangle$. The center line (dash-dot line) separates the upper half from the lower half which correspond to the factors $S_{fi}{}^*$ and S_{fi} in the transition probability, respectively. These diagrams should be distinguished from the Feynman diagrams.

which have evolved from the "bare" operators a, b_j, respectively, at the infinite past. Here, the radiation fields of light beams 2 and 4 are treated as classical fields. By Laplace's transform, it is straightforward to solve the coupled Eqs. (62) and (63) for $a(t)$. Thus,

$$a(t)=\alpha_t a_0+\sum_j \beta_{jt} b_{j0}+\gamma_t\,, \tag{64}$$

where

$$\alpha_t=\frac{1}{2\pi i}\int_{-i\infty}^{i\infty} dp e^{p(t-t_0)}\left[p+i\omega_0+\sum_j \frac{|k_j|^2}{p+i\omega_j}\right]^{-1},$$

$$\beta_{jt}=-\frac{k_j}{2\pi}\int_{-i\infty}^{i\infty} dp e^{p(t-t_0)} \times\left[(p+i\omega_j)\left(p+i\omega_0+\sum_l \frac{|k_l|^2}{p+i\omega_l}\right)\right]^{-1},$$

$$\gamma_t=-\frac{1}{2\pi}\int_{-\infty}^{\infty} dp e^{p(t-t_0)}\tilde{e}_{24}(p)\left[p+i\omega_0+\sum_j \frac{|k_j|^2}{p+i\omega_j}\right]^{-1},$$

and

$$\tilde{e}_{24}(p)=\int_{t_0}^{\infty} dt e_{24}(t) e^{-p(t-t_0)}\,.$$

From above, we see that γ_t is proportional to the strength of the coupling in H_1 of Eq. (60). In the absence of the light beams 2 and 4, according to Eq. (64), $a(t)$ becomes

$$a^{(0)}(\mathrm{t})=\alpha_t a_0+\sum_j \beta_{jt} b_{j0}\,, \tag{65}$$

i.e., $a^{(0)}(t)$ is a linear combination of a_0 and b_{j0} which is as expected intuitively.

From Eqs. (64) and (65) it is now obvious that

$$\langle a^{\dagger}(t)a(t')\rangle=\langle a^{(0)\dagger}(t)a^{(0)}(t')\rangle+\langle a^{\dagger}(t)\rangle\langle a(t')\rangle\,, \tag{66}$$

where

$$\langle\varphi\rangle=\mathrm{Tr}\{e^{-\beta H_0}\varphi\}/\mathrm{Tr}e^{-\beta H_0}\,,$$

by noting that $\langle a_0\rangle=\langle b_{j0}\rangle=0$, $\langle a(t')\rangle=\gamma_{t'}$ and $\langle a^{\dagger}(t)\rangle=\gamma_t{}^*$. Therefore, in this simple model, we have succeeded in showing that Eq. (66), which is analogous to Eq. (57), is an exact relation in which $\langle a^{\dagger}(t)\rangle$ and $\langle a(t')\rangle$ are of the lowest order in H_1.

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