

Theory of the Stimulated Raman Effect in Plasmas

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The stimulated Raman effect in gaseous plasmas is investigated by means of a coupled-mode analysis. A macroscopic representation for the electron density fluctuations is employed, and is shown to reduce to previous results on the optical pumping of plasmas. The Raman threshold power levels calculated are currently achievable in giant-pulse ruby lasers, but the available laser pulse durations are too brief to allow observable amounts of Raman radiation to build up from the thermal noise background, in the case of ordinary plasmas. However, the effect may possibly be observable in the laser-produced type of plasma.

I. INTRODUCTION

THE stimulated Raman effect in liquids was first observed by Woodbury and co-workers.^{1,2} They noticed that a substantial amount of the coherent light leaving the optical cavity of a giant-pulse ruby laser was down-shifted in frequency by an amount equal to the vibrational frequency characteristic of the liquid in the Kerr shutter. Subsequently, Hellwarth^{3,4} worked out the corresponding theory of stimulated emission of light in Raman-active materials. For further details, the reader is referred to the recent book by Bloembergen,⁵ and the recent review articles by Loudon⁶ and Zubov *et al.*⁷

The quantized molecular-vibrational waves correspond to optical phonons, as do the electron oscillations in a gaseous plasma; hence it is natural to look for the stimulated Raman effect in plasmas, as well as liquids and solids.⁸ However, the nonlinear optical effects in a plasma are expected from elementary considerations⁹ to be weaker than the analogous effects in a liquid by a factor of 10^{-7} . Since the Woodbury experiments^{1,2} indicate a threshold laser intensity exceeding 1 MW/cm², one would estimate the threshold in plasmas to exceed 10^7 MW/cm², a figure which lies only slightly outside the present state of the art.¹⁰ Consequently, it seems worthwhile to do a more careful calculation.

In the present paper a classical, coupled-mode analysis is performed to determine the Raman laser threshold in gaseous plasmas governed by Maxwell-

Boltzmann statistics. Bloembergen⁵ has already demonstrated the close connection between the classical, coupled-mode approach of parametric-amplifier theory and the quantum-mechanical treatment of Hellwarth^{3,4}; furthermore, since Raman-scattering matrix-element calculations tend to involve a large number of intermediate virtual states, it seems prudent to work along classical lines.¹¹ The analogous question of the stimulated Brillouin effect associated with ion-acoustic oscillations (acoustical phonons) in plasmas shall not be examined here, in view of the experiments of Chiao, Townes, and Stoicheff¹⁰ indicating thresholds in solids many orders of magnitude higher than the Raman thresholds.

2. OPTICAL PUMPING OF ELECTRON DENSITY FLUCTUATIONS

Consider a fully ionized plasma with discrete electrons of charge density $-\rho_0 + \rho(\mathbf{x}, t)$ and current density $\mathbf{j}(\mathbf{x}, t)$, immersed in a uniform neutralizing background of positive ions having charge density ρ_0 . Close to Maxwellian equilibrium the electron fluctuations, in the presence of a self-consistent electric field \mathbf{E} , are governed by the macroscopic equations

$$\partial\rho/\partial t + \nabla \cdot \mathbf{j} = 0, \quad (1)$$

$$\partial\mathbf{j}/\partial t + 3v_0^2 \nabla\rho - (e/m)(\rho_0 + \rho)\mathbf{E} = -\nu_c \mathbf{j}, \quad (2)$$

$$\nabla \cdot \mathbf{E} = 4\pi\rho, \quad (3)$$

where the thermal speed is given by $v_0^2 = KT/m$, and ν_c is a phenomenological collision frequency for ion-electron encounters (it is well known that electron-electron collisions tend to contribute negligibly to electron-wave damping). As shown in the Appendix, strong electromagnetic fields contribute nonlinear electric currents of the form $\mathbf{j}_{NL} = \boldsymbol{\sigma}_{NL} : \mathbf{E}\mathbf{E}$, which in turn act as driving sources for the density fluctuations; thus the nonlinear form of Eq. (1) becomes

$$\partial\rho/\partial t + \nabla \cdot \mathbf{j} = -\nabla \cdot (\boldsymbol{\sigma}_{NL} : \mathbf{E}\mathbf{E}). \quad (4)$$

¹ E. J. Woodbury and W. K. Ng, Proc. IRE **50**, 2367 (1962).

² G. Eckhardt, R. W. Hellwarth, F. J. McClung, S. E. Schwarz, D. Weiner, and E. J. Woodbury, Phys. Rev. Letters **9**, 455 (1962).

³ R. W. Hellwarth, Phys. Rev. **130**, 1850 (1963).

⁴ R. W. Hellwarth, Appl. Optics **2**, 847 (1963).

⁵ N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965).

⁶ R. Loudon, Advan. Phys. **13**, 423 (1964).

⁷ V. A. Zubov, M. M. Sushchinskii, and I. K. Shuvalov, Usp. Fiz. Nauk **83**, 197 (1964) [English transl.: Soviet Phys.—Usp. **7**, 419 (1964)].

⁸ The theory of ordinary Raman scattering in a solid-state plasma was first investigated by I. I. Sobel'man and E. L. Feinberg, Zh. Eksperim. i Teor. Fiz. **34**, 494 (1958) [English transl.: Soviet Phys.—JETP **7**, 339 (1958)].

⁹ See the free-electron calculations presented in the Appendix of the present paper.

¹⁰ In the stimulated Brillouin scattering measurements of R. Chiao, C. H. Townes, and B. Stoicheff, Phys. Rev. Letters **12**, 592 (1964), power levels up to 10^6 MW/cm² were obtained from focused, giant pulse ruby lasers.

¹¹ Examples of the pitfalls involved in quantum derivations of related nonlinear plasma phenomena are given by H. Cheng and Y. C. Lee, Phys. Rev. Letters **14**, 426 (1965); and D. F. DuBois, *ibid.* **14**, 818 (1965).

Upon combining Eqs. (2) through (4) one gets the plasma wave equation

$$\frac{\partial^2 \rho}{\partial t^2} + \nu_e \frac{\partial \rho}{\partial t} + \omega_p^2 \rho - 3\nu_e^2 \nabla^2 \rho + (4\pi e/m)\rho^2 + (e/m)\nabla \rho \cdot \mathbf{E} = -(\partial/\partial t)\nabla \cdot (\boldsymbol{\sigma}_{\text{NL}}: \mathbf{E}\mathbf{E}), \quad (5)$$

where $\omega_p^2 = 4\pi e\rho_0/m$.

It is of interest to pause for a moment in order to examine the properties of this equation. The first four terms describe a simple harmonic plasma wave $\exp(i\mathbf{k}\cdot\mathbf{x} - i\omega t)$ with the usual Bohm-Gross dispersion relation

$$\omega^2 = \omega_p^2 + 3k^2\nu_e^2 - i\omega\nu_e. \quad (6)$$

The fifth term corresponds to second harmonic generation in that waves of form $\exp(-i\omega_p t)$ give rise to waves of form $\exp(-2i\omega_p t)$. For simplicity, this term shall be neglected in the ensuing analysis.

The sixth term of Eq. (5) refers to the coupling of transverse electromagnetic waves to longitudinal density fluctuations in the presence of a density gradient. As an example, consider the perturbation problem where $\rho = \rho_0 + \rho_1$, with $|\rho_1(\mathbf{x}, t)| \ll |\rho_0(\mathbf{x})|$, under the influence of a uniform, external electric field $\mathbf{E} \exp(-i\omega t)$. If the second harmonic and nonlinear-optical terms are temporarily ignored one can write down the wave equation

$$\left[\frac{\partial^2}{\partial t^2} + \nu_e \frac{\partial}{\partial t} + \omega_p^2 - 3\nu_e^2 \nabla^2 \right] \rho^{(1)} = -(e/m)\nabla \rho^{(0)} \cdot \mathbf{E} \exp(-i\omega t), \quad (7)$$

which has the Fourier transform solution

$$\rho^{(1)}(\mathbf{k}, \omega) = (ie(\mathbf{k} \cdot \mathbf{E})/m\omega^2 \epsilon(k, \omega))\rho^{(0)}(\mathbf{k}), \quad (8)$$

where the longitudinal dielectric constant ϵ is given by

$$\epsilon(\mathbf{k}, \omega) = 1 - (\omega_p^2/\omega^2)(1 + 3\nu_e^2 k^2/\omega_p^2) + i\nu_e/\omega. \quad (9)$$

Assuming that the unperturbed density $\rho^{(0)}$ corresponds to a perfectly random distribution of N charges, one gets, near the plasma resonance $\omega^2 = \omega_p^2 + 3k^2\nu_e^2$, the fluctuation spectrum

$$\langle |\rho^{(1)}(k, \omega)|^2 \rangle \approx N e^2 (\mathbf{k} \cdot \mathbf{E})^2 / m^2 \omega^4 (\nu_e/\omega)^2, \quad (10)$$

a result derived by Berk¹² in his kinetic-theory treatment of optical pumping near the plasma frequency ω_p . This mechanism will also be neglected in the ensuing analysis, although it could be included in a more complete analysis dealing with an inhomogeneous or bounded plasma.

The remaining plasma wave equation may be expressed as

$$\left[\frac{\partial^2}{\partial t^2} + \nu_e \frac{\partial}{\partial t} + \omega_p^2 - 3\nu_e^2 \nabla^2 \right] \rho = -(\partial/\partial t)\nabla \cdot (\boldsymbol{\sigma}_{\text{NL}}: \mathbf{E}_2 \mathbf{E}_1^*) \quad (11)$$

for the case of two electromagnetic waves $\mathbf{E} = \mathbf{E}_1 + \mathbf{E}_2$, where $\mathbf{E}_1 \sim \exp(i\mathbf{k}_1 \cdot \mathbf{x} - i\omega_1 t)$ and $\mathbf{E}_2 \sim \exp(i\mathbf{k}_2 \cdot \mathbf{x} - i\omega_2 t)$. Suppose the frequencies and wave vectors are perfectly matched (corresponding to the conservation of momen-

tum and energy between optical phonons and photons):

$$\mathbf{k} = \mathbf{k}_2 - \mathbf{k}_1, \quad \omega = \omega_2 - \omega_1. \quad (12)$$

If the two electromagnetic waves have the same polarization and propagate parallel to the plasma waves, so that the momentum-matching relation becomes scalar: $k = k_2 - k_1$, it follows from Eq. (11) that the optically-pumped electron density fluctuations are given by

$$\rho(k, \omega) = k \sigma_{\text{NL}} E_1^* E_2 / \omega \epsilon(k, \omega), \quad (13)$$

where the nonlinear conductivity is obtained from the free-electron theory presented in the Appendix, namely,

$$\sigma_{\text{NL}}(\omega = \omega_2 - \omega_1) = \rho_0 e^2 / m^2 c \omega_1 \omega_2, \quad (14)$$

and the dielectric constant ϵ is found from Eq. (9). Near the plasma resonance, $\omega = \omega_p = [\omega_p^2 + 3k^2\nu_e^2]^{1/2}$, the density fluctuation spectrum becomes (letting $n = \rho/e$)

$$\langle |n(k, \omega_p)|^2 \rangle / n_0 = k^4 E_1^2 E_2^2 / 256 \pi^2 n_0 m^2 \omega_1^2 \omega_2^2 (\nu_e/\omega_p)^2, \quad (15)$$

which agrees with Eq. (6) of the work of Kroll, Ron, and Rostoker,¹³ who investigated the optical pumping of plasma fluctuations by two tuned laser beams (i.e., $\omega_2 - \omega_1 = \omega_p$) from a kinetic-theory viewpoint. Hence, the present macroscopic model turns out to be equivalent to a Vlasov-equation treatment.

3. AMPLIFICATION OF RAMAN-SCATTERED LIGHT WAVES

In the previous section it was shown that electron density fluctuations can be driven by two light waves tuned to the frequency ω_p of the fluctuations. Now, as has been pointed out by Rosen,¹⁴ the presence of density fluctuations constitutes a periodically varying dielectric which can scatter incident electromagnetic waves parametrically. As a result, the scattered light waves suffer a frequency shift of amount ω_p . The scattered waves can now interact with the incident waves to further drive the density fluctuations, and in this manner optical energy can regeneratively produce an amplified Raman light wave. Such a process is sometimes referred to as Raman laser action in analogy to the stimulated emission mechanism taking place within the ruby laser light source itself.

A. Brillouin Wave Equation

To begin the analysis, one may consider the plasma as a medium with the transverse dielectric constant

$$\epsilon_T(k, \omega) = 1 - \omega_p^2/\omega^2 + i\omega_p^2 \nu_e/\omega^3, \quad (16)$$

the parametric generalization of which is

$$\epsilon_T(k, \omega) = \epsilon_0 + \epsilon_1 \rho + i\epsilon_2, \quad (17)$$

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

¹⁴ P. Rosen, Phys. Fluids 3, 416 (1960).

¹² H. Berk, Phys. Fluids 7, 917 (1964).

where $\epsilon_0 = 1 - \omega_p^2/\omega^2$, $\epsilon_1 = -4\pi e/m\omega^2$, and $\epsilon_2 = \omega_p^2 v_c/\omega^3$. This provides the additional coupling between electromagnetic waves and density fluctuations, and is entirely consistent with the preceding discussion. The Brillouin wave equation¹⁴ for propagation through such a medium is

$$\nabla^2 \mathbf{E} + \nabla \left(\frac{\nabla \epsilon_T}{\epsilon_T} \cdot \mathbf{E} \right) = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} (\epsilon_T \mathbf{E}), \quad (18)$$

in which the Maxwell curl equations have been combined with the divergence equation in the absence of free charge:

$$\nabla \cdot (\epsilon_T \mathbf{E}) = \nabla \epsilon_T \cdot \mathbf{E} + \epsilon_T \nabla \cdot \mathbf{E} = 0.$$

To keep things simple, as at the end of the last section, assume that all light waves have their polarizations parallel to each other and perpendicular to that of the longitudinal plasma waves. This causes the second term of Eq. (18) to drop out; however, the plasma waves and light waves remain coupled parametrically by the right-hand term. For the case of strong laser fields and forward scattering, one obtains from Eq. (18) the Raman-scattered wave equation

$$[\nabla^2 + (k_s^0)^2 + i(\omega_s^2/c^2)\epsilon_2]E_s(\omega_s) = -(\omega_s^2/c^2)\epsilon_1 E_L(\omega_L)\rho^*(\omega_p), \quad (19)$$

in which the incident laser wave is taken as

$$E_L \sim \exp(i\mathbf{k}_L \cdot \mathbf{x} - i\omega_L t),$$

the Raman wave is taken as $E_s \sim \exp(i\mathbf{k}_s \cdot \mathbf{x} - i\omega_s t)$, and the linear wave vector is $k_s^0 = \omega_s [\epsilon(\omega_s)]^{1/2}/c$. Once again perfect momentum matching has been assumed, namely,

$$k_s = k_L - k_p, \quad \omega_s = \omega_L - \omega_p. \quad (20)$$

As a consistency check, notice that since for optical frequencies and laboratory plasmas $\omega_L, \omega_s \gg \omega_p$, one may write Eq. (20) in the form

$$k_p = k_s - k_L \approx (\epsilon_0^{1/2}/c)(\omega_s - \omega_L) \approx (1/c)\omega_p. \quad (21)$$

In other words, the three coupled waves propagate in unison: $\omega_p/k_p = \omega_s/k_s = \omega_L/k_L = c$. Now, Eq. (21) also implies that

$$k_p \lambda_D = v_0/c \ll 1, \quad (22)$$

where $\lambda_D = v_0/\omega_p$ is the plasma Debye length. But it is well known from the theory of plasma oscillations that $k_p \lambda_D \ll 1$ is consistent with the Bohm-Gross dispersion relation, Eq. (6). Using the same type of reasoning one may show that backward scattering is kinematically impossible, because this requires plasma wavelengths comparable to optical wavelengths.

B. Coupled-Mode Analysis

The plasma wave equation (11) and the Brillouin electromagnetic wave equation (19) may now be solved by means of the coupled-mode analysis described in Bloembergen's book.⁵ Fourier transformation yields

the homogeneous algebraic equations in E_s^* and ρ

$$[-k_s^2 + (k_s^0)^2 - 2ik_s^0 \alpha_s]E_s^* - (1/c^2)\omega_s^2 |\epsilon_1| E_L^* \rho = 0, \quad (23a)$$

$$-(\omega_p k_p / 3v_0^2) \sigma_{NL} E_L E_s^* + [-k_p^2 + (k_p^0)^2 + 2ik_p^0 \alpha_p] \rho = 0, \quad (23b)$$

where the optical attenuation coefficient is $\alpha_s = \omega_p^2 v_c / 2\omega_s k_s^0 c^2$, the plasma-wave attenuation coefficient is $\alpha_p = \omega_p v_c / 6v_0^2 k_p^0$, and the linear-plasma-wave propagation vector is given by $(k_s^0)^2 = (\omega_p^2 - \omega_p^2) / 3v_0^2$. In laboratory plasmas the optical attenuation is very small, whereas the plasma wave attenuation is very large. For example, in a pinch discharge with $n_0 = 10^{16} \text{ cm}^{-3}$ and $T = 10^5 \text{ }^\circ\text{K}$ it turns out for ruby light ($\sim 7000 \text{ \AA}$) that $\alpha_s/k_s^0 \sim 10^{-13}$, $\alpha_p/k_p^0 \sim 10^2$, and $\alpha_s/\alpha_p \sim 10^{-12}$. Consequently the coefficient of ρ in Eq. (23b) may be approximated by $2ik_p^0 \alpha_p$.

The resulting secular determinant may be written down in terms of $k_s = k_s^0 + \Delta k$, where Δk is the nonlinear correction to the scattered-wave propagation vector. From the quadratic formula one obtains the nontrivial root

$$\Delta k = i\alpha_s - i\omega_p \omega_s^2 |\epsilon_1| |E_L|^2 \sigma_{NL} / 12k_s^0 v_0^2 c^2 \alpha_p. \quad (24)$$

Positive gain corresponds to the case where the nonlinear growth exceeds the optical losses, or when $\Delta k < 0$. Writing Eq. (24) in more convenient form, one arrives at the Raman laser action threshold condition¹⁵

$$\frac{1}{2} (n\lambda_D)^3 (\omega_p v_0 / \omega_L) I / nmc^3 \geq 1, \quad (25)$$

in which the approximate relation $v_0/\omega_p \sim (n\lambda_D)^{-1}$ has been used, and where the average laser power flux has been defined as $I = c |E_L|^2 / 2\pi$.

To get an idea of the magnitude of the threshold power involved for ruby light ($\sim 7000 \text{ \AA}$), one may evaluate Eq. (25) for a few gaseous plasmas:

- (a) Pinch discharge: $n = 10^{16} \text{ cm}^{-3}$, $T = 10^5 \text{ }^\circ\text{K}$,
 $I = 10^7 \text{ MW/cm}^2$,
- (b) Q machine: $n = 10^{11} \text{ cm}^{-3}$, $T = 10^3 \text{ }^\circ\text{K}$,
 $I = 10^5 \text{ MW/cm}^2$,
- (c) Ionosphere: $n = 10^6 \text{ cm}^{-3}$, $T = 10^3 \text{ }^\circ\text{K}$,
 $I = 10 \text{ kW/cm}^2$.

The results are discussed further in the final section of this paper.

Before concluding the present section it is of interest to compare the classical calculation of stimulated gain Eq. (24) with the quantum-mechanical result. In a quite recent article¹⁶ Goldman and DuBois have

¹⁵ This result is not altered substantially by slight momentum mismatches, provided that $\Delta k = k_L - k_p \ll k_p$ holds. For pinch discharges this amounts to $\Delta k \ll 10 \text{ cm}^{-1}$, which does not seem a particularly stringent matching condition.

¹⁶ M. V. Goldman and D. F. DuBois, Phys. Fluids 8, 1404 (1965). This article appeared after the present work was submitted for publication.

extended the work of Hellwarth^{3,4} to the case of a classical plasma in the following way. The gain per centimeter of scattered light along the incident beam is

$$g_s = (nIc^2/kT)(\omega_{pi}/\omega_L^3)(2\pi)^3 d^2\sigma/d\omega d\Omega,$$

where $d^2\sigma/d\omega d\Omega$ is the differential cross section for the Raman scattering process. If one utilizes the incoherent scattering cross section for thermally excited electron density fluctuations, namely,

$$d^2\sigma/d\omega d\Omega = (\pi)^{-1}(e^2/mc^2)^2(k_p\lambda_D)^2(\nu_e)^{-1},$$

one finds for the stimulated gain, upon use of Eq. (22), the expression

$$g_s/k_L = \frac{1}{2}(\omega_{pi}/\omega_L)^5(\omega_L/\nu_e)I/nmc^3,$$

in agreement with our Eq. (24) for the case of small α_s .

4. DISCUSSION OF TECHNICAL LIMITATIONS

It has just been shown that positive gain can be achieved in a gaseous plasma for laser power levels of not unreasonable magnitude; however, it is known⁷ that Raman cross sections are proportional to the optical absorption coefficient α_s , as may be verified from Eq. (24). Now, $\alpha_s \sim (\omega_p/\omega_L)^3$, which decreases rapidly with decreasing electron density so that it may be expected that lower power thresholds are associated with quite small magnitudes for net gain. Ordinarily, if such a weak process were contained in a Fabry-Perot optical cavity, the low gain per traversal of the plasma would be compensated by the large number of traversals suffered by a light wave before it managed to penetrate the highly reflecting walls. For the present situation, an optical cavity of extremely high Q would be needed. Unfortunately, the Q is always spoiled by the finite duration (~ 20 nsec) of the pulse from a ruby laser operating in the giant pulse mode.

To illustrate these difficulties, consider the problem of Raman laser-action buildup from the thermal background of electron density fluctuations. Simple statistical arguments, based upon the fact that the noise power is $\frac{1}{2}cKT$ times the number of longitudinal modes which contribute to scattering into a cone of small apex angle in the forward direction, lead one to estimate required gains of around 30 nepers for build-up from thermal noise to megawatt levels. During a 20-nsec giant pulse the laser beam covers a path length l of 600 cm; hence for a pinch discharge the gain expression

$$\Delta\kappa l \sim \alpha_s l I (\text{MW/cm}^2)/10^7 \sim 30 \quad (26)$$

indicates a required power level of $I \sim 10^{14}$ MW/cm². Such enormous powers lie far outside the present state of laser technology.

To observe the stimulated Raman effect in a plasma one would have to employ dense, thermonuclear, laser-produced plasmas such as have been envisaged by

Dawson,¹⁷ and Meyerand and Haught.¹⁸ If the ordered kinetic energy of a dense, laser-heated droplet of plasma could be converted into random kinetic energy, perhaps by expansion against a strong magnetic field, the parameters $n = 10^{20}$ cm⁻³ and $T = 10^8$ °K might conceivably be produced. The corresponding pump power required to cause buildup from thermal noise would become $I \sim 10^8$ MW/cm², a figure only two orders of magnitude higher than currently achieved power levels.¹⁰

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APPENDIX: NONLINEAR CONDUCTIVITY OF A FREE-ELECTRON GAS

The nonlinear optics of a gas of noninteracting electrons has been treated by Bloembergen⁵ by means of the equation of motion

$$m\dot{\mathbf{v}} = e\mathbf{E} + (e/c)\mathbf{v} \times \mathbf{B}, \quad (\text{A1})$$

where, for a plane-polarized electromagnetic wave, one has $\mathbf{E} = (E, 0, 0)$, $\mathbf{B} = (0, E, 0)$. The corresponding scalar equations are

$$m\ddot{x} = eE - (e/c)\dot{z}E, \quad (\text{A2})$$

$$m\ddot{y} = 0, \quad (\text{A3})$$

$$m\ddot{z} = (e/c)\dot{x}E. \quad (\text{A4})$$

Assuming harmonic time dependence and using successive approximations, one arrives at the nonlinear electric current for two harmonic light waves $\mathbf{E} = \mathbf{E}(\omega_1) + \mathbf{E}(\omega_2)$:

$$\begin{aligned} j_x(\omega_p) &= ne\dot{z}(\omega_p = \omega_2 - \omega_1) \\ &= (ne^3/m^2c)(1/\omega_p\omega_1 - 1/\omega_p\omega_2)E_2E_1^* \\ &= (ne^3/m^2c\omega_1\omega_2)E_2E_1^*. \end{aligned} \quad (\text{A5})$$

Hence, the nonlinear conductivity is

$$\sigma_{\text{NL}}(\omega_p = \omega_2 - \omega_1) = ne^3/m^2c\omega_1\omega_2, \quad (\text{A6})$$

which was used in Secs. 2 and 3.

Now, from Eq. (A2) the linear current is $j_x(\omega) = ine^2E(\omega)/m\omega$, so that the ratio of nonlinear to linear effects is

$$j_{\text{NL}}/j_L \sim eE/mc\omega \sim E/E^*. \quad (\text{A7})$$

For ruby light the successive-approximation scheme breaks down whenever $E \sim E^* = 3 \times 10^{11}$ V/cm. The corresponding critical electric field strength for solids is 10^8 V/cm; accordingly, one is tempted to estimate that the plasma nonlinear coupling is weaker than that of solids by a factor of 3×10^{-4} in electric field strength and by a factor 10^{-7} in power level.

¹⁷ J. M. Dawson, Phys. Fluids 7, 981 (1964).

¹⁸ R. G. Meyerand and A. F. Haught, Phys. Rev. Letters 13, 7 (1964).