



FIG. 3. Profiles of  $n$  and  $T_e$  when the initial current distribution is flat:  $j_z = j_{zc}(1 - r^6/a^6)$ ; or peaked:  $j_z = j_{zc}(1 - r^2/a^2)^3$  (Princeton code).

typically 10–20 times higher. This is partly because of the difference in  $T_e$  profile, and partly because of a resistivity enhancement factor of 3–6, apparently accounted for by high- $Z$  impurities.<sup>8,9</sup>

A large, fundamental discrepancy is found in the particle and energy confinement times. For the case of Fig. 1 at 60 msec, we have  $\tau_p = \int_0^a nr dr / (nrv)_a = 2.9$  sec,  $\tau_{Ee'} = 1.5 \int_0^a nT_e r dr / (rQ_e)_a = 0.88$  sec, and  $\tau_{Ei'} = 1.5 \int_0^a nT_i r dr / (rQ_i)_a = 0.90$  sec. These exceed the typical experimental values by almost 2 orders of magnitude. Part of the discrepancy is explained classically by the enhanced effective  $Z$ ; a sizable anomaly remains, especially for particle transport.

In conclusion, we note that the discrepancies be-

tween neoclassical theory and experiment, previously recognized, have become even more striking as a result of the present calculations. The inclusion of trapped-particle pinching has raised the theoretical  $\tau_p$ ; and the proper rounding of the banana/plateau transition has raised  $\tau_{Ee'}$  and  $\tau_{Ei'}$ —especially the latter. The present determination of the neoclassical  $T_e$  and  $n$  profile has also established a marked profile anomaly.

\*Work supported by the U. S. Atomic Energy Commission under Contracts No. AT(11-1)-3073 at Princeton and No. GP-3458 at Texas.

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## Excitation of Plasma Waves by Two Laser Beams\*

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(Received 29 June 1972)

We analyze the effects of (i) the nonlinearity of the large-amplitude plasma wave, and (ii) the inhomogeneity of the plasma, on the excitation of the plasma wave by beating two laser beams.

Recently, there has been considerable interest in the nonlinear excitation of plasma waves by beating two electromagnetic waves,<sup>1–3</sup> both as a plasma heating mechanism for laboratory fusion devices, where the frequencies of presently available high-power lasers are too great to interact with typical confined plasmas, and as a means for studying and controlling the ionosphere.

Here we wish to report the analysis of the following two important effects on this process: (1) the nonlinear behavior of the large-amplitude plasma wave; (2) the effect of an inhomogeneous plasma. First we study the growth and saturation of the large-amplitude plasma wave in a cold homogeneous collisionless plasma due to the beating of two laser beams with frequencies much above the

plasma frequency  $\omega_p$ , taking into account the modulation of the Lorentz force by the large-amplitude plasma wave as well as the temporal variation of its phase. A novel parametric instability is found when the beat frequency is twice the plasma frequency as a result of the modulation of the Lorentz force by the large-amplitude plasma wave. Relativistic effects tend to destroy the phase locking, providing a saturation mechanism for the large-amplitude wave. For simplicity,

consider two parallel propagating electromagnetic waves,  $E^{(1)} \sin(k_1 x - \omega_1 t)$  and  $E^{(2)} \sin(k_2 x - \omega_2 t)$  with polarization electric field along the  $y$  axis and  $\omega_1, \omega_2 \gg \omega_p$ . We assume

$$\alpha_j \equiv eE^{(j)}/m\omega_j c \ll 1, \quad j=1, 2, \quad (1)$$

realistic for most of the present experimental situations. In a cold plasma, the fluid element responds to the electric field of one laser wave with a velocity

$$\dot{y} \equiv \dot{\xi}_y = -(e/2m\omega) \{E^{(1)} \exp[i(k_1 x - \omega_1 t)] + \text{c.c.}\}, \quad (2)$$

where the  $\vec{v} \times \vec{B}$  force is neglected since  $v/c \ll 1$ , and the self-consistent electrostatic field is neglected because of the high frequency of the laser ( $\omega \gg \omega_p$ ). The ion motion is also neglected. Let  $\xi_x(x_0, t)$  be the displacement of the charged fluid element along  $x$  from the equilibrium position  $x_0$ , so that the instantaneous position of the fluid element is

$$x(t) = x_0 + \xi_x(x_0, t). \quad (3)$$

The equation of motion for  $\xi_x$  at the beat frequency is

$$d^2 \xi_x / dt^2 + \omega_p^2 \xi_x = (e/mc) (\dot{\xi}_y^{(1)} B_z^{(2)} + \dot{\xi}_y^{(2)} B_z^{(1)}). \quad (4)$$

This is an exact nonlinear equation for  $\xi_x$  in a cold plasma so long as the charge sheets do not cross over,<sup>4</sup> because the electrostatic field is then simply obtained from Gauss's law:  $E = 4\pi n_0 e \xi_x$ . We assume that the solution to Eq. (4) is of the form

$$\xi_x(x_0, t) = A(t) \sin[k_0 x_0 - \omega_p t + \varphi(t)], \quad (5)$$

where the amplitude  $A(t)$  is small compared with  $k_0^{-1}$ , and both  $A(t)$  and the phase  $\varphi(t)$  are slowly varying compared with  $\omega_p$ . In the absence of the electromagnetic wave, Eq. (5) with  $A$  and  $\varphi$  constant is an exact solution for the large-amplitude wave in a cold plasma provided  $Ak_0 \ll 1$ , the condition for no crossing of the charge sheets. The nonlinearity in the interaction between the electromagnetic waves and the plasma through the Lorentz force then causes the amplitude  $A$  and phase  $\varphi$  of the plasma wave to change in time, if the matching conditions in the frequency and wave number are met. The rate of change of  $A$  and  $\varphi$ , however, is slow compared with  $\omega_p$  if  $(eE/m\omega c)^2 \ll 1$ . Substituting Eq. (5) into the right-hand side of Eq. (4) and using the Bessel identity

$$\exp(i\alpha\theta) = \sum_{l=-\infty}^{\infty} J_l(\alpha) \exp(i\theta),$$

we can rewrite Eq. (4) as

$$\ddot{\xi}_x + \omega_p^2 \xi_x = -\left(\frac{e}{m}\right)^2 \frac{E_1 E_2^* \Delta k}{4\omega_1 \omega_2} \left(\frac{1}{i} \sum_l J_l(\Delta k A) \exp\{i[(\Delta k + lk_0)x_0 - (\Delta\omega + l\omega_p)t + l\varphi]\} + \text{c.c.}\right). \quad (6)$$

where  $\Delta k = k_1 - k_2$ ,  $\Delta\omega = \omega_1 - \omega_2 = \Delta k c$ . We consider first the case in which  $\Delta k = k_0$ ,  $\Delta\omega = \omega_p$ . Substituting Eq. (5) into Eq. (6) and performing the average over space  $x_0$  and fast time variation  $\sim \omega_p^{-1}$ , we obtain the equation for  $A(t)$  and  $\varphi(t)$  on the slow time scale, keeping only the leading terms in  $(\dot{A}/A)/\omega_p$  and  $(\dot{\varphi}/\varphi)/\omega_p$ ,

$$\dot{A} = \lambda \sin\varphi, \quad (7)$$

$$A\dot{\varphi} = \lambda \cos\varphi, \quad (8)$$

where  $\lambda = \alpha_1 \alpha_2 \omega_p / 4k_0$ , and  $\alpha_j$  is defined in Eq. (1), and we have set  $J_0 = 1$ , since  $k_0 A \ll 1$ , and  $\omega_p$

$= ck_0$ . From Eq. (8) it is clear that the plasma oscillation phase locks very rapidly with the beat wave with stationary phase  $\varphi = \frac{1}{2}\pi$ , since  $\dot{\varphi}$  is positive for  $-\frac{1}{2}\pi < \varphi < \frac{1}{2}\pi$  and negative for  $\frac{1}{2}\pi < \varphi < \frac{3}{2}\pi$ . The stationary phase is reached when  $\cos\varphi = 0$  and  $\dot{A} > 0$ . Thus, the large-amplitude plasma wave grows linearly in time,

$$A(t) = A(0) + \frac{1}{4} \alpha_1 \alpha_2 (\omega_p / k_0) t. \quad (9)$$

In this approximation the wave would grow until  $k_0 A \approx 1$ ,  $\dot{\xi}_x \approx c$ . Later we therefore discuss rela-

tivistic corrections.

Next we consider the case where  $\Delta\omega = 2\omega_p$ ,  $\Delta k = 2k_0$ . Again performing the spatial and fast time average on Eq. (6), we obtain

$$\dot{A}_2 = -\frac{1}{2}\alpha_1\alpha_2\omega_p A_2 \sin 2\varphi = -2\lambda k_0 A \sin 2\varphi, \quad (10)$$

$$\dot{\varphi} = -\frac{1}{2}\alpha_1\alpha_2\omega_p \cos 2\varphi = -2\lambda k_0 A \cos 2\varphi. \quad (11)$$

Thus, the plasma wave and the beat again tend to phase lock. At the stationary phase  $\varphi = -\frac{1}{4}\pi$ , the amplitude of the plasma wave is exponentially growing,  $A_2(t) = A_0 \exp(\gamma t)$ , with growth rate  $\gamma = \alpha_1\alpha_2\omega_p$ . By one  $e$ -folding of this instability with  $\Delta\omega = 2\omega_p$ , the plasma wave growth given by Eq. (9) for the case  $\Delta\omega = \omega_p$  has long since saturated. Thus, the second-harmonic case gives somewhat weaker coupling. This parametric instability is in fact a four-wave process in which the large-amplitude plasma wave, modulating the Lorentz force due to the beat wave at  $2\omega_p$ , renders the plasma wave itself unstable.

As the amplitude increases, the relativistic effect on the frequency mismatch becomes important, leading eventually to phase unlocking and saturation of the amplitude. With relativistic corrections for  $\xi/c \ll 1$ , i.e., letting  $\xi \rightarrow (d/dt)\xi(1 + \frac{1}{2}\xi^2)$ , Eqs. (7) and (8) become, for the case  $\Delta\omega = \omega_p$ ,

$$\dot{a} = \sin\varphi, \quad (12)$$

$$a\dot{\varphi} = \cos\varphi + \delta a^3, \quad (13)$$

where  $a = A/\lambda$ ,  $\delta = \frac{3}{16}(\omega_p^3/c^2)\lambda^2$ . We can integrate the above equations to obtain

$$\dot{a}^2 = 1 - \frac{1}{16}(\delta a^3)^2, \quad (14)$$

for the case  $a(0) \ll 1$ . Thus, the wave growth is stopped when  $\delta a^3 = 4$ , and the amplitude saturates at

$$Ak_0 = (\frac{1}{16}\alpha_1\alpha_2)^{1/3} \ll 1, \quad (15)$$

where  $\omega_p = k_0 c$  is used. We may remark that, under the assumption  $\alpha \ll 1$ , the harmonic excita-

tion of plasma waves at  $2\omega_p$  etc. can be neglected since the amplitude of the harmonic is limited by  $A_2 k_0 \approx \alpha_1\alpha_2$ , much less than Eq. (15). Because of this relatively low level of saturation, wave breaking as a thermalization process does not occur. Without breaking or other dissipative mechanisms, the amplitude would oscillate and the motion is reversible. Thus, collisions or other parametric processes are necessary for thermalization of wave energy. Landau damping and other thermal effects are not important in a homogeneous plasma, as the plasma wave generated by two parallel propagating electromagnetic (em) waves has a phase velocity equal to the velocity of light. On the other hand, the plasma wave generated by two opposing em waves has a phase velocity  $\omega_p/k_0 = c(\omega_1 - \omega_2)/(\omega_1 + \omega_2)$ , which can be comparable to the electron thermal speed  $v_e$  if  $\omega_p/\omega_1$  is of the order of  $v_e/c$ . In this case, the electron Landau damping of the plasma wave can provide the dissipation. For  $\omega_p/k_0 v_e \gg 1$  (weak Landau damping), a similar analysis for the large-amplitude wave can also be carried out for the case of opposing laser beams. The growth rate of the plasma wave is found to be larger than that given in Eq. (9) by a factor of  $(\omega_1 + \omega_2)/\omega_p$ .

Now we turn to the effects of plasma inhomogeneity. Here the waves are generated in the region where the local plasma frequency  $\omega_p(x)$  is close to the beat frequency  $\Delta\omega$  of the two em waves. As the plasma wave propagates into the region of lower density, its wave vector  $k(x)$  increases and phase velocity decreases as a result of the dispersion relation  $\omega^2 = \omega_p^2 + 3k^2 v_e^2$ , leading to its eventual absorption by Landau damping in the region where the phase velocity is comparable to the thermal speed. In the following analysis, we assume small-amplitude plasma waves. The equation of motion of the electron fluid in the presence of two em waves  $E_1 \sin(k_1 x - \omega_1 t)$  and  $E_2 \sin(k_2 x - \omega_2 t)$  with  $\omega_1, \omega_2 \gg \omega_p$  and  $E_1, E_2$  along the  $y$  axis is

$$nm(\partial\vec{v}/\partial t + \vec{v} \cdot \nabla\vec{v}) = -\nabla p + ne\vec{E} + ne(\vec{E} + \vec{v} \times \vec{B}/c), \quad (16)$$

where  $p$  is the electron pressure and  $\vec{E}$  is the electrostatic field. The velocity due to the high-frequency em wave is given by Eq. (1). The linearized Fourier-component equation of motion for  $\omega_0 = \Delta\omega$  is

$$i\omega_0 n_0 m v_x = -\gamma v_e^2 m \frac{\partial n_1}{\partial x} + n_0 e E_x - \frac{i e^2 n_0}{4 m} \frac{\Delta k E_1 E_2^*}{\omega_1 \omega_2} \exp(-i \Delta k x), \quad (17)$$

where  $\Delta k = k_1 - k_2$ ,  $\omega = \Delta\omega = \omega_1 - \omega_2$ , and  $\gamma = 3$  for the adiabatic equation of state:  $p n^{-\gamma} = \text{const}$ . The Poisson equation is  $\partial E/\partial x = -4\pi n_0 e v_x$ . Thus, we have the following equation for the electrostatic field:

$$\partial^2 E/\partial x^2 - \{[\omega_p^2(x) - \omega_0^2]/3v_e^2\} E = S \exp(-i \Delta k x), \quad (18)$$

where

$$S = -\frac{i}{12} \frac{e}{m} \frac{\omega_p^2(x) \Delta k}{\omega_1 \omega_2 v_e^2} E_1 E_2^*.$$

For a linear density profile,  $\omega_p^2(x) = \omega_0^2(1+x/L)$  with  $\omega_0 = \Delta\omega$ , Eq. (18) becomes

$$\partial^2 E / \partial x^2 - \alpha x E = S \exp(-i \Delta k x), \quad (19)$$

where  $\alpha = \omega_p^2 / 3v_e^2 L$ . The boundary conditions are such that the solution is spatially damped at  $x \rightarrow \infty$  and oscillatory and outgoing at  $x \rightarrow -\infty$ . The homogeneous equation is the well-known Airy equation, whose solution has the following integral representation:

$$E_{\pm} = \int_{C_{\pm}} dp \exp(px - p^3/3\alpha), \quad (20)$$

where  $E_+$  is the solution damped at  $x \rightarrow +\infty$ , and  $E_-$  is the outgoing-wave solution at  $x \rightarrow -\infty$ . The contour  $C_+$  runs from  $-i\infty$  to  $+i\infty$ .  $C_-$  runs from  $+\infty$  to  $+i\infty$ . The saddle points are  $p = \pm(\alpha x)^{1/2}$ , on the real axis for  $x > 0$  and on the imaginary axis for  $x < 0$ . The asymptotic solutions can then be obtained from Eq. (20) by integrating along the path of steepest descent,

$$E_+(+\infty) = (\pi)^{1/2} (\alpha/x)^{1/4} \exp(-\frac{2}{3}\alpha^{1/2} x^{3/2}), \quad x \rightarrow \infty, \quad (21)$$

$$E_-(-\infty) = (\pi)^{1/2} (\alpha/|x|)^{1/4} \exp(-\frac{2}{3}i\alpha^{1/2}|x|^{3/2} - i\pi/4), \quad x \rightarrow -\infty. \quad (22)$$

The solution of the inhomogeneous equation can then be constructed in terms of the solutions of the homogeneous equation  $E_+$  for  $x > 0$  and  $E_-$  for  $x < 0$  by the method of Green's functions,

$$E(x) = E_+(x) \int_{-\infty}^x dx' \frac{E_-(x') S(x')}{W} + E_-(x) \int_x^{\infty} dx' \frac{E_+(x') S(x')}{W}, \quad (23)$$

where  $W = E_+'E_- - E_- 'E_+ = 2\pi\alpha$  is the Wronskian. The outgoing wave at  $x \rightarrow -\infty$  is then

$$E(-\infty) = E_- \int_{-\infty}^{\infty} dx' \frac{E_+(x') S(x')}{2\pi\alpha} = \frac{E_- S}{\alpha} \exp\left[\frac{i(\Delta k)^3}{3\alpha}\right]. \quad (24)$$

With the group velocity given by  $v_g = \partial\omega/\partial k = 3kv_e^2/\omega_p$ , and the energy density of the plasma wave given by  $2|E|^2/4\pi$ , where the factor of 2 accounts for the kinetic energy of the oscillation, we obtain the outgoing energy flux of the plasma wave,

$$F_{\text{out}} = v_g \frac{|E_{-\infty}|^2}{2\pi} = \frac{1}{32} \frac{L}{\omega_0} \left(\frac{\omega_0^2}{\omega_1 \omega_2}\right)^2 \left(\frac{e}{mc}\right)^2 E_1^2 E_2^2 \approx \frac{\pi^2}{2} \left(\frac{e}{mc^2}\right)^2 \left(\frac{\omega_0}{\omega_L}\right)^4 \frac{L}{\omega_0} |F_{\text{in}}|^2, \quad (25)$$

for  $E_1 \approx E_2$ ,  $\omega_L = \omega_1 \approx \omega_2$ , where  $F_{\text{in}} = c(E_1^2 + E_2^2)/8\pi \approx c|E|^2/4\pi$  is the incident energy flux. The outgoing energy flux would be absorbed by the plasma through Landau damping.

For opposing laser beams only the factor  $\Delta k$  is changed in our calculations from  $(\omega_1 - \omega_2)/c$  to  $(\omega_1 + \omega_2)/c$ , and the outgoing energy flux is hence enhanced over that given in Eq. (25) by a factor  $4(\omega_L/\omega_p)^2$ . In this case for 10- $\mu\text{m}$  ( $\text{CO}_2$ ) lasers and plasma density  $n = 10^{16}$ , we find

$$F_{\text{out}}/F_{\text{in}} = 10^{-14} F_{\text{in}} L,$$

with  $L$  in centimeters and  $F_{\text{in}}$  in watts per square centimeter. Since  $\text{CO}_2$  lasers of powers  $10^{13}$  W/cm<sup>2</sup> are presently available, the method appears within the realm of practicality, although at much higher power levels our linear approximation will not apply.

In the case of two parallel lasers incident on a parabolic density profile,  $\omega_p^2(x) = \omega_0^2(1-x^2/L^2)$  with  $\Delta\omega = \omega_0$ , the outgoing flux is enhanced over that in Eq. (25) by a factor  $c/v_e$ , and the absorption length is reduced by the same factor.

It has recently been pointed out<sup>5</sup> that in a three-wave process of the type considered here, the maximum energy which can be extracted is limited to  $\omega_p/\omega_1$  by action conservation. The authors of Ref. 5 then consider cascading to higher and lower frequencies,  $\omega_1 + \omega_p$ ,  $\omega_1 - 2\omega_p$ , etc., to increase the absorption efficiency. It is interesting to note that the laser powers which they find necessary for efficient cascading are comparable to those which we find necessary in the presence of the typical plasma inhomogeneities.

We acknowledge helpful conversations with J. M. Dawson and F. W. Perkins on this subject.

\*Work partially supported by the U. S. Atomic Energy Commission under Contract No. AT(11-1)-3237.

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## Thermodynamic Fluctuations in a Reacting System—Measurement by Fluorescence Correlation Spectroscopy

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(Received 10 July 1972)

The temporal correlations of thermodynamic concentration fluctuations have been measured in a chemically reactive system at equilibrium by observing fluctuations of the fluorescence of a reaction product. The experiment yields the chemical rate constants and diffusion coefficients and shows the coupling among them. Data are reported for binding of ethidium bromide to DNA.

The time correlations of thermodynamic concentration fluctuations in reactive multicomponent systems at equilibrium are determined by the kinetics of chemical reactions and diffusion processes. Purely diffusive fluctuations have been measured with great success by quasielastic light scattering<sup>1</sup> and extension to reaction kinetics has prompted several experiments<sup>2</sup> and attracted considerable theoretical attention.<sup>3</sup> However, it now appears that homogeneous chemical kinetics are not amenable to scattering studies because the dielectric-constant changes that reveal the fluctuations are usually too small. In contrast, optical absorbance coefficients and fluorescent quantum yields frequently display large changes. Hence we chose to observe intrinsic concentration fluctuations using a fluorescent indicator.

We report here direct observations of fluctuations about thermodynamic equilibrium in a reactive multicomponent system of biophysical interest. We have studied the reversible binding to DNA of ethidium bromide (EtBr), a dye that inhibits nucleic acid synthesis.<sup>4</sup> The complex of dye and DNA is strongly fluorescent<sup>5</sup>; thus fluctuations in the number of molecules of the complex in a small volume defined by a beam of exciting radiation are indicated by fluctuations of the total fluorescent power.<sup>6</sup>

The principal chemical reaction between the DNA (*A*) and the EtBr (*B*) to form the fluorescent complex (*C*) is supposed to be a single-step bimolecular process with rate constants  $k_f$  and  $k_b$  (in fact the system is more complex, as will be

seen):



The equilibrium constant is defined in terms of the equilibrium concentrations  $\bar{C}_A$ ,  $\bar{C}_B$ , and  $\bar{C}_C$  as  $K = k_f/k_b = \bar{C}_C/\bar{C}_A\bar{C}_B$ . Thermodynamic concentration fluctuations decay via the chemical reaction as  $\exp(-Rt)$ , where the inverse relaxation time  $R$  is<sup>7</sup>

$$R = k_f(\bar{C}_A + \bar{C}_B) + k_b. \quad (2)$$

Since diffusion provides alternative relaxation paths, the correct description of the fluctuation spectrum requires a set of coupled differential equations which for ideal solutions assume the form

$$\delta \delta C_j / \partial t = D_j \nabla^2 \delta C_j + \sum_k T_{jk} \delta C_k, \quad (3)$$

where  $\delta C_j \equiv \delta C_j(\vec{r}, t)$  are the local concentration fluctuations of the three reactants of Eq. (1); the  $D_j$  are corresponding diffusion coefficients, and the  $T_{jk}$  are the elements of the matrix of linear chemical interaction coefficients implied by Eq. (1).<sup>3</sup>

In our experiment, fluctuations of the concentration  $C_C$  of the fluorescent complex cause the fluctuations in a photodetector current. The autocorrelation function of the photocurrent fluctuation  $\delta i(t) = i(t) - \langle i(t) \rangle$ , with  $|\tau|$  written  $\tau$ , is  $G_i(\tau) = \langle \delta i(t + \tau) \delta i(t) \rangle$ . Neglecting shot noise for simplicity, the photocurrent due to fluorescence induced by the exciting radiation  $I(\vec{r})$  is

$$i(t) = g \epsilon Q \int_{-\infty}^{\infty} I(\vec{r}) C_C(\vec{r}, t) d^3r, \quad (4)$$