Bifurcation in Degenerate Four-Wave Mixing in Liquid Suspensions of Microspheres

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We examine phase conjugation in liquid suspensions of microparticles in the saturation limit. If the polarizations of the pump waves are orthogonal, new states should appear discontinuously in a pairwise fashion which are stable at the bifurcation point. These states correspond to different intensities of the conjugate and amplified probe waves. Analysis indicates that this effect should be experimentally accessible in the microwave region utilizing 10-μm microspheres.

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Recently, phase conjugation has attracted considerable interest with numerous publications discussing the fundamental and technological implications of this effect.[1] Here we examine degenerate four-wave mixing in liquid suspensions of microspheres operating in the saturation regime and predict the existence of novel nonlinear behavior. Specifically, if the polarizations of the two pump beams are orthogonal, then in this regime feedback in the four-wave mixing process itself should be sufficient that the active medium will support a collection of different states for the same values of system parameters. These different states are distinguishable from each other by the intensity of the conjugate and exiting probe waves and, on a more microscopic level, correspond to different spatial distributions of microparticles. More precisely, we predict that as the value of κL increases, where κ is the four-wave mixing coefficient and L is the interaction length, the systems exhibit bifurcation in the sense that new states appear in pairs for sufficient increases in κL. Further, these new states arise discontinuously with nonzero values of the conjugate wave; the precise details depend on the specific material characteristics of the active medium. In addition, stability analysis indicates that these new states are stable, at least in the immediate vicinity of the bifurcation point. Finally, we show that bifurcation in degenerate four-wave mixing should occur in general whenever the active medium is operating in the saturation regime with the polarizations of the two pump beams orthogonal.

In previous research, we examined degenerate four-wave mixing in liquid suspensions of microspheres in the presence of weak pump waves.[2] For these media, electrostrictive forces modulate the microparticle density such that two orthogonal spatial gratings are created. Coherent scattering of pump radiation from these two gratings gives rise to the formation of a conjugate wave,[3] as well as to amplification of the probe wave. To describe the nonlinear electrodynamics of such media, one uses the Maxwell equations for the radiation field coupled to the Planck-Nernst equation for the microparticle density.[4] Specifically,

$$\frac{\partial}{\partial t}n(\mathbf{r},t) = D\nabla^2 n(\mathbf{r},t) - \frac{D}{kT}\nabla\cdot[\mathbf{F}(\mathbf{r},t)\,n(\mathbf{r},t)], \qquad (1)$$

$$\mathbf{F}(\mathbf{r},t) = -\tfrac{1}{2}\alpha\nabla\langle E^2(\mathbf{r},t)\rangle_{\mathrm{av}}, \qquad (2)$$

$$\left[\nabla^2 - \frac{1}{v^2}\frac{\partial^2}{\partial t^2}\right]\mathbf{E}(\mathbf{r},t) = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{r},t), \qquad (3)$$

where $n(\mathbf{r},t)$ is the microsphere density, D the diffusion coefficient for microspheres in a viscous liquid, $\mathbf{F}(\mathbf{r},t)$ the electrostrictive force which drives the microparticles, and α the polarizability of a sphere in an electromagnetic field whose wavelength is long compared to the particle size; $\mathbf{E}(\mathbf{r},t)$ denotes the total electromagnetic field irradiating the suspension, v the velocity of light in the medium; and $\mathbf{P}(\mathbf{r},t)$ is the nonlinear polarization of the medium. Equation (3) assumes that losses are negligible, a situation that can be obtained by use of a nonabsorbing suspension whose Rayleigh scattering length is much greater than any propagation lengths that appear in the problem. Finally, the angular brackets in Eq. (2) denote a temporal average.

The electromagnetic field can be written conveniently as

$$\mathbf{E}(\mathbf{r},t) = \tfrac{1}{2}\textstyle\sum_j \epsilon_j(\mathbf{r},t)\hat{\mathbf{e}}_j \times \exp[i(\omega t - \mathbf{k}_j\cdot\mathbf{r}_j - \phi_j)] + \text{c.c.}, \qquad (4)$$

where $j=(1,2)$ refers to the pump waves, $j=p$, the probe field, and $j=c$, the conjugate wave. Here

$\epsilon_j(\mathbf{r},t)$ is the complex amplitude of the jth field, $\hat{\mathbf{e}}_j$ and $\mathbf{k}_j$ the corresponding polarization and wave vector. For our purposes, we shall take the pump amplitudes to be equal and undepleted so that $\epsilon_1 = \epsilon_2 \equiv E_0$, and if $\hat{\mathbf{e}}_p$ is taken to be parallel to $\hat{\mathbf{e}}_1$, then $\hat{\mathbf{e}}_c$ will be parallel to $\hat{\mathbf{e}}_2$, since $\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}_2^* = 0$. For the situations considered here, we anticipate pump powers many orders of magnitude greater than either the exiting probe or the conjugate wave. Thus the pump waves will transfer only a small fraction of their power to either the probe or the conjugate wave and thus can be treated within the nondepleted pump approximation. We take $\mathbf{k}_1 = -\mathbf{k}_2 \equiv \mathbf{K}$, $\mathbf{k}_p = -\mathbf{k}_c \equiv \mathbf{Q}$, and note that for this choice of pump polarization, only one microparticle grating will be created, i.e., the one whose spacing $\Lambda = 2\pi/|(\mathbf{K}-\mathbf{Q})| \equiv 2\pi/|\mathbf{q}|$. For convenience, we scale the probe and conjugate waves to the initial probe field $\epsilon_p(0)$, i.e., $\epsilon_c(\mathbf{r},t) \equiv \epsilon_p(0) u(\mathbf{r},t)$ and $\epsilon_p^*(\mathbf{r},t) = \epsilon_p(0) v(\mathbf{r},t)$. Finally, we set $\phi_1 = \phi_2 = 0$ and $\phi_p = -\phi_c \equiv \Phi$, the common phase of the conjugate and probe waves.

Inserting Eq. (4) into Eqs. (1)–(3), and decomposing the microsphere density into various grating orders m_l,

$$n(\mathbf{r},t) = \left(\frac{N}{V}\right) \sum_{l=-\infty}^{\infty} m_l(t) \exp[il(\mathbf{q}\cdot\mathbf{r} - \Phi + \theta)], \quad (5)$$

with N/V the microsphere density in the absence of external fields, we have

$$\dot{m}_l + ilm_l\dot{\theta} = -\frac{l^2}{\tau_D} m_l + \frac{l}{\tau_D} \beta A (m_{l-1} - m_{l+1}), \quad (6)$$

where $m_l = m_{-l}^*$ if n is to be real, $\tau_D = (Dq^2)^{-1}$ is the time needed for a microparticle[5] to diffuse a distance $1/q$, $\beta \equiv \alpha E_0 \epsilon_p(0)/4kT$ is the saturation parameter, and $(u+v) \equiv -A \exp[i\theta]$. We can define a saturation field E_s by $\beta = E\epsilon_p(0)/E_s^2$, so that $E_s \equiv 2(kT/\alpha)^{1/2}$. Typical values at room temperatures for 10-μm spheres are on the order of 10^{-2} V/cm, corresponding to powers of 0.1 W/cm^2. Since the response time of the microparticles is viscous limited, the radiation field will follow the motion of the microspheres adiabatically. Thus, within the slowly varying phase and amplitude approximation,[2]

$$\frac{\partial}{\partial z}[A \exp(i\theta)] = \frac{i}{2} \kappa L m_1 \exp(i\theta), \quad (7)$$

where z is a dimensionless coordinate in the direction of propagation which is scaled to the interaction length L, and $\kappa \equiv \pi Q \alpha (N/V) \alpha E^2/kT$. Note that the conjugate and probe waves are only directly coupled to the first-order grating. Furthermore, phase mismatch assures that only negligible radiation will be scattered from the higher-order gratings.

To demonstrate bifurcation, we first extract the steady-state solutions of Eqs. (5)–(7). If all time derivatives are set equal to zero, Eq. (6) reduces to the indicial equation for the Bessel functions of imaginary argument. In particular, the normalized microparticle density for the grating of order l is[6]

$$m_l = \left(\frac{I_l(2\beta A)}{I_0(2\beta A)}\right), \quad (8)$$

and one can show from Eq. (5) that $n(r)$ is given by the Maxwell-Boltzmann distribution for a collection of microparticles in an electrostrictive potential $U(r) = \alpha E \epsilon_p(0) A \cos(\mathbf{q}\cdot\mathbf{r} - \Phi + \theta)$. Next, an examination of Eq. (7) reveals that if m_1 is given by Eq. (8), A is independent of position, and $\theta(z) = \theta_0 + 2\kappa L F(2\beta A) z$, where $F(x) \equiv I_1(x)/xI_0(x)$ and θ_0 is a constant. If we introduce the standard boundary conditions,[7] the A satisfies the transcendental equation

$$A = \sec\{\kappa L[2F(2\beta A)]\}. \quad (9)$$

The efficiency for generating conjugate waves is $\eta = \tan^2\{\kappa L[2F(2\beta A)]\}$, and the probe amplification is $\xi = \sec^2\{\kappa L[2F(2\beta A)]\}$, which are analogous to the weak-field result, except that the four-wave mixing coefficient κ is replaced by its saturated value,

$$\kappa_s = \kappa \frac{I_1(2\beta A)}{\beta A I_0(2\beta A)}. \quad (10)$$

Note that different values of A correspond to different microparticle densities through Eq. (8).

Before presenting numerical solutions of Eq. (9), we can obtain some insight into bifurcation by considering the limit $\beta A \gg 1$, where Eq. (9) reduces to $B = \sigma \cos B$, with $B = \sigma/A$ and $\sigma = (\kappa L/\beta)$. Clearly, this exhibits multiple solutions so long as $\sigma \gg 1$. The larger the value of σ, the greater are the number of allowed solutions. This feature of bifurcation is depicted in Fig. 1, which displays A, the normalized probe beam exiting from the sample, as a function of κL for $\beta = 0.1$. An examination of this figure reveals that as κL increases, new solutions appear discontinuously in pairs. Initially, these solutions exhibit a strong dependence on κL, but this saturates rapidly to a linear dependence. Note, too, that the phases of each pair of new solutions differ by π from the previous set. Since $\xi = A^2$ and $\eta = A^2 - 1$, the saturated probe amplification and conjugate-wave efficiency all eventually exhibit a quadratic dependence on κL. Further examination of Fig. 1 reveals that bifurcation occurs at $\kappa L = 2.2$, 5.2, 9.2, 11.5, etc., corresponding to $\sigma = 22$, 52, 92, 115, etc. To illuminate the role of the saturation parameter, we have depicted the quantity $A' \equiv A\beta$ in Fig. 2. An examination of this figure reveals that as β increases, i.e., as σ decreases, solutions are lost, as implied by the discussion above. Note that as long as the various solutions exist, they depend on $1/\beta$. To understand why, we note that since A' is vir-

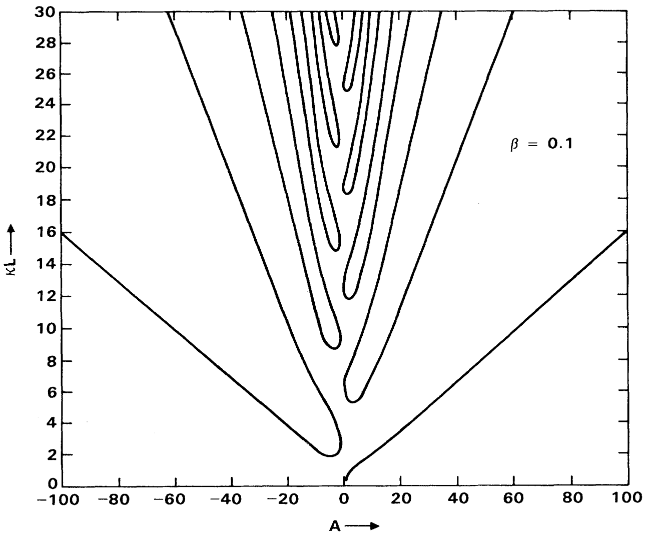

FIG. 1. Bifurcation of A as a function of κL for $\beta = 0.1$ in a lossless medium.

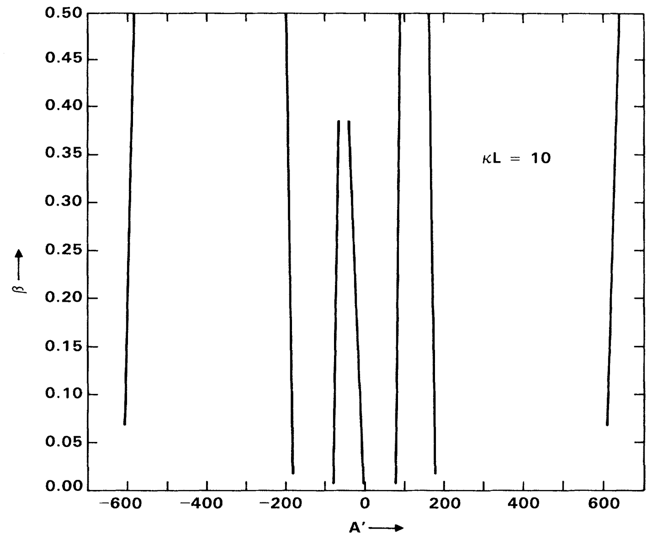

FIG. 2. Bifurcation of $A\beta$ as a function of β for $\kappa L = 10$ in a lossless medium.

tually independent of β, A varies as $1/\epsilon_p(0)$. Since the probe wave that exits from the system has $\epsilon_p(L) = \epsilon_p(0)A$, it follows that in the saturation region the emitted probe wave is independent of the incident probe power.

As a specific numerical example, we consider a liquid suspension of nonabsorbing microspheres 10 μm in radius and irradiated by 1-mm radiation. For an interaction path length of 20 cm and a filling factor of 10^{-4}, $\kappa L = 1.23\delta$, where δ is the pump power in watts per square centimeter. Thus pump power densities of several watts per square centimeter should reveal the first five or six bifurcation points. By setting the input probe power at 1 mW/cm^2, we find $\beta \simeq 0.1$ so that the conditions of Fig. 1 are achieved. Note that the exiting probe and conjugate wave power are always much smaller than the pump power (i.e., several tens of milliwatts at most) so that the pump fields will not be significantly depleted. The Rayleigh scattering length for this system is over 3 km.

Finally, we note that self-focusing[8] should not occur for the situations considered here because there are no electrostrictive forces between the microparticles and just the pump waves. Specifically, self-focusing for this situation requires $(P_0P_p)^{1/2} \sim 1$ W, where P_0 (P_p) is the pump (probe) power. For the problem of interest here, the probe power is on the order of 1 mW, and the pump powers are about 10 W , so that $(P_pP_p)^{1/2} \sim 0.1$ W, which is too small by an order of magnitude.

Next, we examine the stability of these solutions in the vicinity of the bifurcation points by linearizing Eqs. (6) and (7) about their steady-state values:

$$m_l(z,t) = \frac{I_l(2\beta A_0)}{I_0(2\beta A_0)} + \mu_l(z,t) + i\nu_l(z,t), \qquad (11a)$$

$$\theta(z,t) = 2\kappa L F(2\beta A_0) z + \psi(z,t), \qquad (11b)$$

$$A(z,t) = A_0 + \alpha(z,t). \qquad (11c)$$

Here A_0 is any of the allowed solutions of Eq. (9) in the vicinity of the bifurcation point; μ_l, ν_l, ψ, and α are infinitesimal quantities which are first-order fluctuations of the microparticle density and electromagnetic fields away from steady state. Inserting Eqs. (11) into the coupled Maxwell and Planck-Nernst equations yields

$$\tau_D \dot{\mu}_l = l^2 \mu_l + l\beta A_0(\mu_{l-1} - \mu_{l+1}) + l^2 \left[\frac{I_l(2\beta A_0)}{I_0(2\beta A_0)} \right] \frac{\alpha(z,t)}{A_0}, \qquad (12a)$$

$$\tau_D \dot{\nu}_l = l^2 \nu_l + l\beta A_0(\nu_{l-1} - \nu_{l+1}), \qquad (12b)$$

$$\frac{\partial \alpha}{\partial z} = -\frac{1}{2}\kappa L \nu_1, \qquad (12c)$$

$$\frac{\kappa L}{2A_0} \left[\frac{I_1(2\beta A_0)}{I_0(2\beta A_0)} \right] + A_0 \frac{\partial \psi}{\partial z} = \frac{1}{2}\kappa L \mu_1. \qquad (12d)$$

In the vicinity of the bifurcation point, $\beta A_0 << 1$ and

Eqs. (12) reduce to

$$\tau_D \dot{\mu}_l(p,t) + l^2 \mu_l(p,t) = il^2 \frac{I_l(2\beta A_0)}{I_0(2\beta A_0)} \nu_1(p,t), \quad (13a)$$

$$\tau_D \dot{\nu}_l(p,t) + l^2 \nu_l(p,t) = l \frac{I_l(2\beta A_0)}{I_0(2\beta A_0)} \left[i \left(\frac{\kappa L}{2A_0 p} \right) \dot{\mu}_1 + \left(\frac{\kappa L}{2A_0 p} \right)^2 \dot{\nu}_1 \right] \tau_D, \quad (13b)$$

where p is the Fourier transform of the dimensionless spatial coordinate z. We may test for stability by assuming that μ_l, ν_l vary with time as $e^{-\epsilon t}$ so that the solution in question is stable if $\epsilon > 0$. Carrying this out, we find that $\epsilon(p)$ has two solutions:

$$\epsilon_{\pm}(p) = \frac{1}{\tau_D} \left(1 \pm \frac{p_0}{(p^2 + p_0^2)^{1/2}} \right), \quad (14)$$

where $p_0 \equiv \kappa L/2A_0$. Thus, in the vicinity of the bifurcation point, all solutions are stable. However, the smaller p, i.e., the longer the wavelength of the fluctuation, the less stable is the solution. This feature is quite reasonable since the longer the wavelength of the fluctuation, the greater are the numbers of microparticles involved; hence the longer the time needed to reestablish equilibrium. Note that the most stable solutions occur for the smallest values of the nonlinear parameter and the largest values of the exiting probe wave, i.e., the most stable solutions are those which occur first.

Finally, we demonstrate that bifurcation is not limited, at least in principle, to just liquid suspensions of microspheres. This can be done most readily by observing that when losses are negligible, typical nonlinear susceptibilities scale as $(E_s/E)^2 \simeq E_s^2/E_0 \epsilon_p(0) A$ for $E >> E_s$, the saturation field. However, this is precisely how suspensions of microspheres behave in the large-field limit; in particular see Eq. (10), where the calculations regarding the appearance of bifurcation solutions for suspensions should apply to other active media.

[1]See *Optical Phase Conjugators,* edited by R. A. Fisher (Academic, New York, 1983), and references therein.

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