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Acousto-optical four-wave-mixing processes in compressible artificial dielectrics

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Compressible artificial dielectrics, such as bubbly fluids, have sufficiently large acousto-optical coefficients that they can be utilized as nonlinear media for acoustical-optical four-wave-mixing processes in which two of the input beams are sound waves and the other two are electromagnetic waves. We examine frequency shifting and acoustic-beam deflection at visible wavelengths as well as acoustically pumped optical phase conjugation at microwave frequencies in these media.

I. INTRODUCTION

Several years ago, Shiren, Arnold, and Kazaka[1] achieved an acousto-optical analog of optical phase conjugation via degenerate four-wave mixing in glasses and generated a backward-wave phonon echo. These experiments, conducted at 9 GHz, involved the generation of a time-reversed acoustic pulse in response to a pulsed acoustic signal utilizing microwave beams as the pump waves. Their results, which are of interest in themselves, stimulate one to examine the reverse effect, i.e., acoustically pumped optical phase conjugation.[2] For this process the probe and conjugate beams are electromagnetic; but the pump waves are acoustical beams. More generally, we investigate acousto-optical four-wave-mixing (AOFWM) processes, in which we seek to control the electromagnetic beam's phase, frequency, or propagation direction. To date, such processes have not been studied in great detail and this paper examines the feasibility of achieving AOFWM processes in compressible, artifical dielectrics. More precisely, we present a theory of acoustically pumped phase conjugation of microwave radiation as well as beam deflection and frequency shifting of visible light by higher-order acoustic index gratings in compressible artifical dielectrics.

Our motivation for utilizing compressible artifical dielectrics arises from the large acousto-optical coefficients that these media possess. For example, the first-order correction to the optical dielectric constant of a bubbly fluid, $\epsilon_1(\Omega)$, arising from sound waves of frequency Ω is[3]

$$\epsilon_1(\Omega)=-9\epsilon_h f_0[(\epsilon_r-1)/(\epsilon_r+2)](p/3\gamma p_0)R(\Omega)\ , \tag{1.1}$$

where ϵ_h is the dielectric constant of the host fluid, f_0 is the equilibrium volume fraction of bubbles, ϵ_r is the ratio of the dielectric constant of the bubble to ϵ_h, p is the pressure generated by the sound wave in the fluid, γ is the ratio of the specific heats, $R(\Omega)$ is the bubble response function which peaks at $\Omega=\Omega_0$, the resonance frequency and p_0, the ambient pressure. For a compressible, artifical dielectric composed of a 1% volume fraction of 2.5-μm bubbles, irradiated by a sound wave whose pressure is 3 kPa, $\epsilon_1/\epsilon_h=2.5\times10^{-3}$. This is three orders of magnitude greater that its value in the absence of the microbubbles and encourages one to consider higher-order processes with such two component, compressible media. The studies presented here suggest that the acousto-optical coefficients of this class of active media are sufficiently large, that a variety of AOFWM processes should be experimentally accessible. In contrast, we find that the efficiencies for AOFWM processes, in which the signal waves are electromagnetic, are essentially negligible for most solids and liquids due to the fact that they are not sufficiently compressible.

This paper is organized into four sections, of which this, the first, is the Introduction. In Sec. II, the problem of AOFWM processes is formulated and the physics of counterpropagating pump-induced index gratings in optical media is examined. Such index gratings are of paramount importance for achieving AOFWM processes in compressible artifical dielectrics. In Sec. III, we investigate acousto-optical frequency shifting and beam deflection of visible radiation by these second-order acoustically generated index gratings. We also study acoustically pumped microwave phase conjugation in compressible artifical dielectrics. Detailed numerical results are presented and we discuss possible experimental verification of these theories. Finally, in Sec. IV we present our conclusions and to underscore the unique properties of this class of nonlinear media, contrast them to typical liquid and solid-state media.

II. BASIC CONCEPTS AND PROBLEM FORMULATION

A. Counterpropagating pump-induced index gratings

In optical phase conjugation, both static and dynamic index gratings are created by the nonlinear interaction of electromagnetic waves in a material medium. For most nonlinear media, the coupling between the probe wave with either pump beam will generate a number of static index gratings. These gratings are periodic, with spatial periods,[4]

$$\Lambda_\pm=2\pi/|\mathbf{Q}\pm\mathbf{K}|\ , \tag{2.1}$$

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and contain all of the phase information impressed on the probe wave. Here, $\pm\mathbf{K}$ refers to the two counterpropagating pump waves and $\mathbf{Q}$ is the probe wave vector. Phase-conjugate radiation is generated in the following fashion. The $+\mathbf{K}$ pump beam and the probe couple together to create an index grating with spatial periodicity Λ_-. This grating coherently scatters the counterpropagating pump beam to produce a wave that is phase conjugate to the probe. A similar event occurs with the Λ_+ grating.

The counterpropagating pump waves can couple to produce two different types of index gratings: (i) a static grating with a spatial period of $2K$ and (ii) a spatially uniform high-frequency index grating, which oscillates at twice the laser frequency, i.e., 2ω. The static pump grating can lead to modulation effects in phase conjugation which have been discussed elsewhere.[5] For most nonlinear media, the high-frequency, spatially uniform index grating plays no role in phase conjugation. However, for sufficiently fast media, usually those whose nonlinear response is essentially electronic, phase-conjugate radiation can be produced by a process similar to parametric downshifting of the probe wave. Specifically, the probe beam is scattered by this high-frequency index grating and its frequency is downshifted, i.e., $\omega\rightarrow\omega-2\omega=-\omega$. To obtain a greater understanding of the physics of this grating and how it implies the possibility of acoustically pumped microwave phase conjugation, we review phase conjugation in optical Kerr media.

We suppose that an optical Kerr medium, characterized by a third-order susceptibility $\chi^{(3)}$, is irradiated by coherent light. The dielectric constant $\epsilon(\mathbf{r},t)$ is

$$\epsilon(\mathbf{r},t)=\epsilon_0+4\pi\chi^{(3)}E^2(\mathbf{r},t) \ . \tag{2.2}$$

Here ϵ_0 is the linear dielectric constant of the medium and $\mathbf{E}(\mathbf{r},t)$ is the electric component of the total radiation field, which we write as

$$\begin{aligned}\mathbf{E}(\mathbf{r},t)=&E_0\mathbf{e}_1\cos(\mathbf{K}\cdot\mathbf{r}-\omega t)+E_0\mathbf{e}_2\cos(\mathbf{K}\cdot\mathbf{r}+\omega t)\\&+(E_p/2)\mathbf{e}_p\exp(i\mathbf{Q}\cdot\mathbf{r}-\omega t)+\text{c.c.}\end{aligned} \tag{2.3}$$

If the medium response time $\tau\gg1/\omega$, then Eq. (2.2) is averaged over a time long compared to the optical period, but short compared to the response time.

Focusing on the contribution to the dielectric constant arising from the two pump waves and denoting this by $\delta\epsilon(\mathbf{r},t)$, we have

$$\delta\epsilon(\mathbf{r},t)=2\pi\chi^{(3)}E_0^2(\mathbf{e}_1\cdot\mathbf{e}_2)[\cos(2\mathbf{K}\cdot\mathbf{r})+\cos(2\omega t)] \ . \tag{2.4}$$

In Eq. (2.4), we have assumed that the medium exhibits a broadband response for simplicity. This is not a necessary condition and the derivation below can be generalized to more realistic frequency responses. The nonlinear polarization $\mathbf{P}_{\mathrm{NL},p}(\mathbf{r},t)$ generated by this index grating is

$$\mathbf{P}_{\mathrm{NL},p}(\mathbf{r},t)=\delta\epsilon(\mathbf{r},t)\mathbf{E}(\mathbf{r},t) \ . \tag{2.5}$$

The spatially varying component of this grating, i.e., the $\cos(2\mathbf{K}\cdot\mathbf{r})$ does not directly give rise to a phase-conjugate wave. However, the spatially uniform piece will downshift the probe wave by 2ω. This is equivalent to replacing $t\rightarrow-t$, i.e., time reversal or phase conjugation. In fact, we have

$$\begin{aligned}\mathbf{P}_{\mathrm{NL},p}(\mathbf{r},t)=&-\pi K^2\chi^{(3)}E_0^2(\mathbf{e}_1\cdot\mathbf{e}_2)(E_p/2)\mathbf{e}_p\\&\times\exp[i(\mathbf{Q}\cdot\mathbf{r}+\omega t)]+\text{c.c.}\end{aligned} \tag{2.6}$$

Next, we determine the contribution of this term to the four-wave-mixing coefficient κ. Inserting the expression for the nonlinear polarization into the wave equation for the conjugate wave, making the slowly varying envelope approximation (SVEA) and assuming a steady state, we have

$$d\mathbf{E}_C(z)/dz=-2i\pi^2K\chi^{(3)}E_0^2(\mathbf{e}_1\cdot\mathbf{e}_2)(E_p/2)\mathbf{e}_p \ , \tag{2.7}$$

which implies a contribution to the four-wave-mixing coefficient of

$$\kappa=2\pi K\chi^{(3)}(\mathbf{e}_1\cdot\mathbf{e}_2)E_0^2 \ . \tag{2.8}$$

The only feature of this index grating that is crucial to phase conjugation is the fact that it oscillates at twice the probe frequency. Other electrical quantities that appear in Eq. (2.4) are the pump energy density $U_{\text{pump}}=E_0^2/8\pi$, the scalar product $(e_1\cdot e_2)$ of the pump polarization vectors, and the nonlinear susceptibility $\chi^{(3)}$. It follows that optical phase conjugation should be possible with other types of pump waves, provided the active medium has an analog of $\chi^{(3)}$, denoted by δ, which couples the probe wave to the pump beams to create a nonlinear polarization. Specifically, if the electromagnetic waves are replaced by acoustic waves, the mixing coefficient for acoustically pumped, optical phase conjugation is

$$\kappa=2\pi K\delta A_{\text{pump}}^2 \ . \tag{2.9}$$

B. Acousto-optical interactions

Following standard procedure, we write the energy density U_T as[6]

$$U_T=U_A+U_E+U_{\mathrm{AO}} \ , \tag{2.10}$$

where the first term refers to the acoustic energy, the second to the energy associated with coupling of the medium to any electrical or electromagnetic fields, and the third is the acousto-optical interaction. The different energy densities, in turn, consist of the following:

$$U_A=c^{(4)}A^2/2!+c^{(6)}A^3/3!+c^{(8)}A^4/4!+\cdots \ , \tag{2.11}$$

where A is the acoustic amplitude, usually a tensor, equal to

$$A_{ij}=\left[\frac{\partial u_i}{\partial x_j}+\frac{\partial u_i}{\partial x_j}\right]\bigg/2 \ , \tag{2.12}$$

with u_i the displacement and x_j the position. A_{ij} is dimensionless and equal to the acoustical displacement of the material divided by a characteristic length. For phonons, A_{ij} is on the order of a typical lattice displacement divided by the atomic separation, usually 10^{-5} or less. The coefficients $c^{(i)}$ are constants with dimensions of $\mathrm{N/m^2}$ or $\mathrm{J/m^3}$. The coupling to the electromagnetic field is

$$U_E=\chi^{(1)}E^2/2!+\chi^{(2)}E^3/3!+\chi^{(3)}E^4/4!+\cdots , \quad (2.13)$$

where E is the electric component of the radiation field and the coefficients $\chi^{(i)}$ are the different nonlinear susceptibilities. Finally, the acousto-optical interaction is

$$U_{AO}=\alpha AE+\beta EA^2+\delta E^2A^2+\cdots . \quad (2.14)$$

For most material media,[6] β is on the order of 1–10 N/m^2 and δ is about equal to $\beta\chi^{(3)}/\chi^{(2)}$ implying values of order 10^{-2} or less.

C. Nonlinear acousto-optical coefficients for compressible, artifical dielectrics

Suspensions of compressible microparticles should have large acousto-optical coefficients and are logical media for AOFWM processes. At visible wavelengths, an aqueous suspension of bubbles whose dimensions are much less than the wavelength of the incident light can be utilized. At microwave frequencies, nonaqueous suspensions of compressible microparticles must be used to ensure that the incident microwave radiation will propagate in the host fluid. Finally, an aerosol of plant cells might be suitable for infrared wavelengths.

We first evaluate the coefficient for AOFWM processes in a bubbly medium assuming that the host fluid is completely passive to both acoustic and electromagnetic radiation. The expression for the dielectric constant of a two-component medium is[6]

$$\epsilon=\epsilon_h\{1+3[(\epsilon_r-1)/(\epsilon_r+2)]f_0\} . \quad (2.15)$$

An acoustic wave will give rise to both volume pulsations and shape deformations. If the microbubble is small compared to the acoustic wavelength, then only its size will be altered and the acoustic amplitude A is

$$A=\delta r/r_0 . \quad (2.16)$$

Here r_0 is the equilibrium bubble radius and δr is the change in bubble size due to the applied sound waves. To evaluate the dielectric constant when the suspension is driven by sound waves, the equilibrium volume fraction in Eq. (2.15) must be is replaced by its instantaneous value. Thus $f_0\to f(\mathbf{r},t)=4\pi NR^3(\mathbf{r},t)/3$ with N the microbubble density and $R(\mathbf{r},t)$ the instantaneous bubble radius (of the microbubble located at the point $\mathbf{r}$) which oscillates in time due to the impinging sound waves. The third-order susceptibility can be extracted from the expression above by noting that

$$R(r,t)=r_0+\delta r(r,t) , \quad (2.17)$$

and identifying the coefficient of the term proportional to δr^2,

$$\gamma=9\epsilon_h[(\epsilon_r-1)/(\epsilon_r+2)]f_0/4\pi . \quad (2.18)$$

For an aqueous suspension of microbubbles, with a volume fraction of 1%, $\delta=6\times10^{-4}$ at visible wavelengths. In the microwave region of the spectrum, a nonaqueous suspension of microbubbles with the same volume fraction has $\delta=2.87\times10^{-3}$. Thus, if the bubble size changes by 10%, AOFWM will change the dielectric constant by 1.7×10^{-4} at visible wavelengths and 3.6×10^{-4} at microwave frequencies.

III. FOUR-WAVE-MIXING ACOUSTO-OPTICAL PROCESSES

In Sec. III A frequency shifting of visible laser light in a bubbly medium is studied. In Sec. III B we examine beam deflection and in Sec. III C acoustically pumped optical phase conjugation of microwave radiation in the same medium is discussed. Finally, Sec. III D is concerned with possible experimental confirmation of these theories.

An expression for the radial pulsations of a microbubble driven by a sound wave of frequency Ω and wave vector $\mathbf{K}$ is required. The sound wave generates a sinusoidal pressure of amplitude p

$$p(r,t)=p\cos(\mathbf{K}\cdot\mathbf{r}-\Omega t) , \quad (3.1)$$

which tends to compress and expand the microbubbles. To simplify matters, the microbubbles are assumed to respond linearly to the acoustic waves, so that the equation of motion for $\delta r(t)$ is[8]

$$\frac{d^2\delta r}{dt^2}+(\Omega_0/Q)\frac{d\delta r}{dt}+\Omega_0^2\delta r=-(p/\rho r_0)\cos(\mathbf{K}\cdot\mathbf{r}-\Omega t) , \quad (3.2)$$

with Q the bubble Q. The microbubble's resonance frequency is equal to the speed of sound divided by the bubble size

$$\Omega_0=(3\gamma p_0/\rho r_0^2)^{1/2} . \quad (3.3)$$

Solving Eq. (3.2) for the fractional change in the bubble size, we find

$$\delta r(r,t)/r_0=-(p/3\gamma p_0)R(\Omega)\cos(\mathbf{K}\cdot\mathbf{r}-\Omega t+\phi) , \quad (3.4a)$$

where the response function $R(\Omega)$ and the phase lag ϕ between the microbubble pulsations and the acoustic pressure wave are

$$R(\Omega)=\Omega_0^2/[(\Omega^2-\Omega_0^2)^2+(\Omega_0\Omega/Q)^2]^{1/2} , \quad (3.4b)$$

$$\sin\phi=(\Omega_0\Omega)/\{Q[(\Omega^2-\Omega_0^2)^2+(\Omega_0\Omega/Q)^2]^{1/2}\} . \quad (3.4c)$$

If instead of a bubble a hollow flexible shell of thickness $h\ll r_0$ is used, the resonance frequency Ω_S is[9]

$$\Omega_S=(3\rho_A r_0/\rho_S h)^{1/2}\Omega_0\ll\Omega_0 , \quad (3.4d)$$

where ρ_S (ρ_A) is the shell (air) density.

For counterpropagating pump waves, the induced nonlinear index grating is

$$\delta\epsilon(r,t)=0.5f_0\epsilon_h[(\epsilon_r-1)/(\epsilon_r+2)][pR(\Omega)/\gamma p_0]^2 \times[\cos2(\Omega t+\phi)+\cos2\mathbf{K}\cdot\mathbf{r}] . \quad (3.5)$$

The first grating oscillates at twice the acoustic frequency and gives rise to frequency shifting if $\Omega\ll\omega$, the optical frequency. If $\Omega=\omega$, then this grating will backscatter the optical beam to form its phase conjugate. The second grating is static in nature; however, it is spatially periodic and will deflect the light beam without shifting its frequency. This feature discriminates the AOFWM process from any second-order diffracted light arising from the standard acousto-optical scattering, i.e., the effect of first-order changes in the composite's dielectric constant. On resonance, $\delta\epsilon(\mathbf{r},t)$ attains its maximum value

$$\delta\epsilon(r,t)=0.5f_0\epsilon_h[(\epsilon_r-1)/(\epsilon_r+2)](pQ/\gamma p_0)^2 \times(\cos 2\mathbf{K}\cdot\mathbf{r}-\cos 2\Omega_0 t)\ . \qquad (3.6)$$

A. Optical frequency shifting by counterpropagating acoustic beams

The physical situation is depicted in Fig. 1. Two counterpropagating sound waves drive a bubbly fluid to form the index grating described by Eq. (3.5). Phase mismatch can be utilized to isolate the effects of these two gratings on a probing laser beam. In particular, the phase mismatch associated with the static grating can be maximized by employing a configuration in which the light beam propagates parallel to one of the sound waves. For this configuration the phase mismatch parameter is KL (L is the optical path length) and for 100-MHz sound waves with a 1-cm path length, KL exceeds 10^4. Thus beam deflection by the static grating can be ignored and we may examine frequency shifting due to the oscillating grating in isolation.

The electric field amplitude of the radiation field obeys the wave equation

$$\left[\nabla^2-\frac{1\partial^2}{v^2\partial t^2}\right]E(r,t)=-\frac{\epsilon_2\partial^2}{2c^2\partial t^2}[E(r,t)\cos 2\Omega t]\ . \qquad (3.7)$$

The oscillating grating will up- and downshift the incident radiation in multiples of 2Ω. Spectrally decomposing $E(\mathbf{r},t)$ via

$$E(r,t)=\sum_{m=-\infty}^{\infty}A_m\exp[i(\mathbf{k}\cdot\mathbf{r}-\omega_m t)]\ , \qquad (3.8)$$

with $\omega_m=\omega+2m\Omega$, inserting this expression into the wave equation and making the SVEA, we find that the amplitudes A_m obey the differential-difference equations

$$\left[\left[\frac{2m\Omega}{v}\right]^2+\frac{4mk\Omega}{v}-2i\mathbf{k}\cdot\nabla\right]A_m = -\frac{\epsilon_2\omega_m^2}{2c^2}(A_{m-1}+A_{m+1})\ . \qquad (3.9)$$

For sound waves, $\Omega\ll\omega$, and it is clear that phase mismatch is negligible. In particular, for this process the phase mismatch parameter is $\Phi=\Omega L/v$, which is on the order of 0.1 for a 1-cm pathlength, Ar^+-ion laser light, and $\Omega=10^9$ cycles/s. Accordingly, Eq. (3.9) reduces to

$$-2i\mathbf{k}\cdot\nabla A_m=-\frac{\epsilon_2\omega_m^2}{2c^2}(A_{m-1}+A_{m+1})\ . \qquad (3.10)$$

Equation (3.10) is a Bessel function recurrence relation,[10] provided ω_m is replaced by ω, which is justified for all reasonable values of m. Thus, if the optical path length is L, then

$$A_m=(-i)^m J_m(\epsilon_2 kL/2\epsilon_h)\ . \qquad (3.11a)$$

The intensity of the mth shifted beam $I_m(\Omega)$ is

$$I_m(\Omega)=I_0J_m^2\left[f_0\left[\frac{\epsilon_r-1}{\epsilon_r+2}\right]\left[\frac{pR(\Omega)}{2\gamma p_0}\right]^2 kL\right]\ , \qquad (3.11b)$$

where I_0 is the initial laser intensity. Figure 2 depicts the frequency dependence of the intensity for the $m=0$, ±1, and ±2 components for the case of an aqueous suspension containing 1% volume fraction of 0.25-μm bubbles. The suspension is irradiated by an Ar^+-ion laser beam as well as two counterpropagating acoustic beams, each with an intensity of 1 W/m^2. The microbubbles have a Q of 10. An examination of this figure reveals the follow-

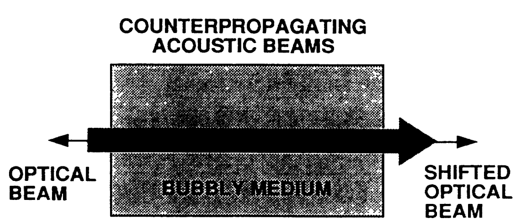

FIG. 1. Parallel beam configuration for frequency shifting of optical beams by a uniform, high-frequency index grating created by two counterpropagating sound waves.

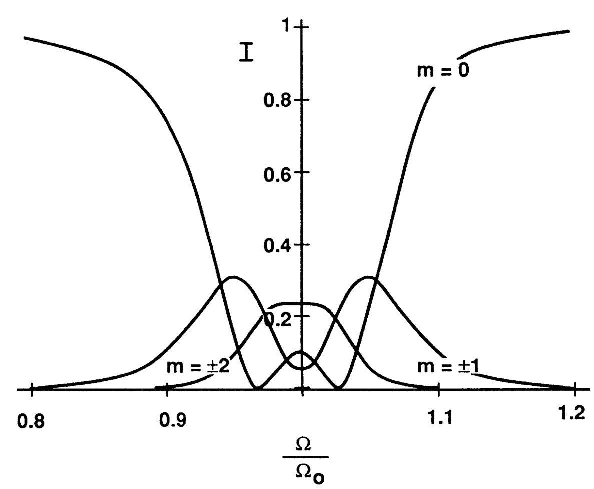

FIG. 2. Intensities of the unshifted and first two shifted components for acoustic intensities of 1 W/m^2, 1% volume fraction of 0.25-μm bubbles and Ar^+-ion laser light.

ing: off-resonance, i.e., $|\Omega-\Omega_0|/\Omega_0>0.15$ very little radiation is shifted. As the acoustic frequency is tuned to resonance ($\Omega_0=8\times10^7$), significant radiation is up- and downshifted into the $\omega\pm2\Omega$ modes. Further tuning towards line center gives rise to higher efficiency for frequency conversion to these modes and significant amounts of the second sidebands, i.e., $\omega_2=\omega\pm4\Omega$ appear. Further in towards line center, the first sideband intensity decreases and the second sideband begins to dominate the line spectra. The fundamental mode vanishes altogether at $0.96\Omega_0$. At line center, the second sideband dominates the spectrum.

B. Optical beam deflection by counterpropagating acoustic beams

Next, we examine beam deflection by the static index grating. To isolate the effects of this grating from the one studied above, we again resort to phase mismatch. The phase mismatch parameter for the high-frequency grating is $\Phi=\Omega L/v$, which must be on the order of 10 to ensure that this grating can be ignored. For $\Omega=10^9$ cycles/s, Φ will exceed 10 if $L>200$ cm.

The wave equation for the electromagnetic wave is

$$\left[\nabla^2-\frac{1}{v^2}\frac{\partial^2}{\partial t^2}\right]E(r,t)=-\frac{\epsilon_2}{2\epsilon_0}k^2E(r,t)\cos(2\mathbf{K}\cdot\mathbf{r}) , \tag{3.12}$$

which tends to deflect light at angles θ_m, given by

$$\tan\theta_m=2mK/k . \tag{3.13}$$

The electric field can be conveniently decomposed into a series of deflected components A_m via

$$E(r,t)=\sum_{m=-\infty}^{\infty}A_m\exp[i(k_mr-\omega t)] , \tag{3.14}$$

where the amplitudes obey the differential-difference equation

$$4mK(k\cos\Psi+mK)A_m-2i(\mathbf{k}+2m\mathbf{K})\cdot\nabla A_m=-\frac{\epsilon_2k^2}{2\epsilon_0}(A_{m-1}+A_{m+2}) . \tag{3.15}$$

In deriving Eq. (3.15), we have made the SVEA and Ψ is the angle between $\mathbf{K}$ and $\mathbf{k}$. For this case, phase mismatch is a serious issue. In particular, the mismatch parameter for deflected light of order m is $2mKL(K/k)$. For visible light and $\Omega=10^9$ cycles/s, $K/k<0.03$, KL is on the order of 10^4 or more, so the angle Ψ must be adjusted. If we set $\Psi=\pi/2\pm K/k$, then only the $m=-1$ mode or the $m=+1$ mode will appear. Thus we must employ a configuration in which the acoustic and optical beams are nearly orthogonal to one another, as depicted in Fig. 3. For either case, the intensity of the deflected beam at L is

$$I_{\pm1}(L)=I_0\sin^2\left[f_0\left[\frac{\epsilon_r-1}{\epsilon_r+2}\right]\left[\frac{pR(\Omega)}{2\gamma p_0}\right]^2kL\right] , \tag{3.16a}$$

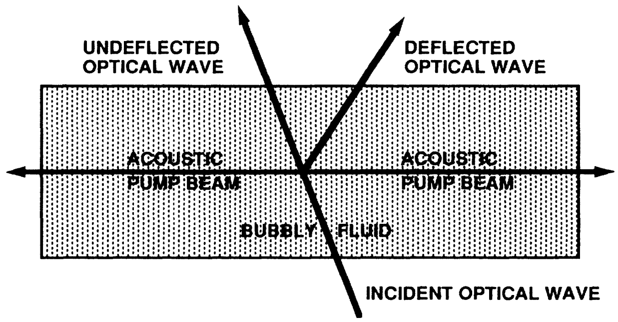

FIG. 3. Beam geometry for optical deflection via the static, spatially periodic grating generated by two counterpropagating pump waves.

and the undeflected beam intensity is

$$I_0(L)=I_0\cos^2\left[f_0\left[\frac{\epsilon_r-1}{\epsilon_r+2}\right]\left[\frac{pR(\Omega)}{2\gamma p_0}\right]^2kL\right] . \tag{3.16b}$$

Figure 4 depicts the intensity of the deflected and undeflected waves as a function of the acoustic wave frequency.

C. Acoustically pumped microwave phase conjugation

We consider a nonaqueous suspension of microbubbles irradiated by two counterpropagating acoustic pump beams and a microwave probe beam which is orthogonal to the pumps, as depicted in Fig. 5. This configuration is chosen to minimize acoustic losses and is discussed in Sec. III D 2. To simplify matters we neglect any gratings created between the acoustic pump waves and the electromagnetic probe beam. The analysis of Sec. II gives the following value for the four-wave-mixing coefficient κ_A:

$$\kappa_A(\Omega)=2\pi Kf_0\epsilon_h[(\epsilon_r-1)/(\epsilon_r+2)][pR(\Omega)/\gamma p_0]^2 . \tag{3.17}$$

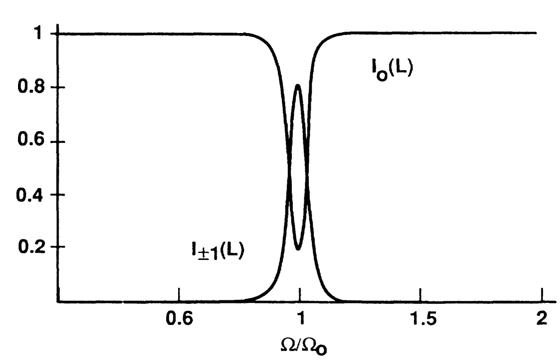

FIG. 4. Intensities of various components of the deflected light by the second-order acoustical gratings.

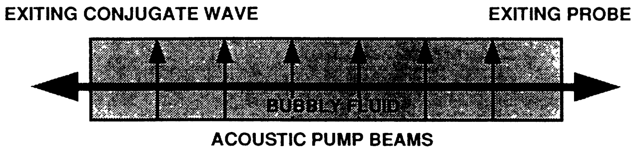

FIG. 5. Beam configuration for acousto-optical phase conjugation in a bubbly medium.

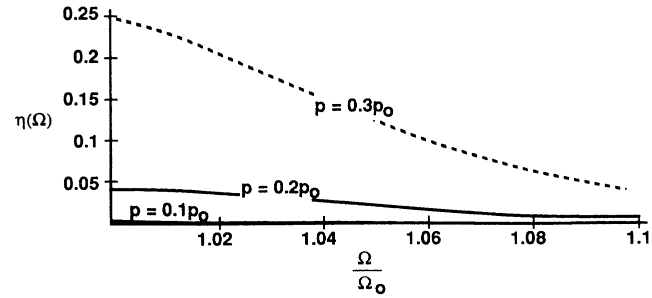

FIG. 7. Efficiency for generating phase conjugate radiation using a 1% volume fraction of microbubbles.

Note the sensitive dependence on the frequency since $\kappa_A(\Omega)$ declines as $(\Omega^2-\Omega_0^2)^{-2}$ far from resonance, so that the efficiency decreases as $(\Omega^2-\Omega_0^2)^{-4}$ in the small signal regime. As a specific numerical example, consider the situation in which $\Omega \gg \Omega_0$ where, with λ the radiative wavelength,

$$\kappa_A(\Omega)=(\pi f_0/4\lambda)(p/\gamma p_0)^2(\Omega_0/\Omega)^4 \, . \tag{3.18}$$

For $p/p_0=0.2$, $\Omega_0/\Omega=0.1$, and a volume fraction of 20%, the four-wave-mixing coefficient is on the order of 10^{-8} for 10-cm radiation. If $\Omega_0/\Omega=0.75$, and the other parameters have the same values, $\kappa_A=2.5\times10^{-4}$; then, $\kappa_A L=0.1$ for optical path lengths on the order of 400 cm. These values of p/p_0 require very mild acoustic pressures, approximately 20 W/m^2. 10-cm radiation corresponds to a few gigahertz; which implies microbubble sizes on the order of several hundred angstroms.

On resonance, the four-wave-mixing coefficient is

$$\kappa_A(\Omega)=2\pi K f_0 \epsilon_h[(\epsilon_r-1)/(\epsilon_r+2)](pQ/3\gamma p_0)^2 \, , \tag{3.19}$$

where we have used the expression for the resonance frequency of a microbubble. For a bubble Q of 10, 500-MHz radiation, a volume fraction of 20% and an acoustic pressure of 20 W/cm^2, the four-wave-mixing coefficient achieves a value of 0.03, so that optical path lengths on the order of several meters should be sufficient to readily observe the effect. Note that for resonant systems acoustic absorption will be a problem.

Figure 6 depicts the frequency dependence of the four-wave-mixing coefficient for a nonaqueous suspension of consisting of a 1% volume fraction of microbubbles. The bubble Q was chosen to be 5, with $\epsilon_r=0.5$ and $\epsilon_h=2$. The suspension is irradiated by 2.4-GHz microwave and acoustic radiation. The frequency dependence of the four-wave-mixing coefficient is exhibited for three different values of p/p_0: 0.1, 0.2, and 0.3. The corresponding efficiency for generating phase-conjugate radiation is depicted in Fig. 7 for an optical path length of two meters. An examination of Fig. 7 reveals that the efficiency can exceed several percent if the acoustic pressures are on the order of 20 kPa. Long optical path lengths can be obtained if the acoustic and microwave beams propagate parallel to one another and the suspension is maintained in a waveguide.

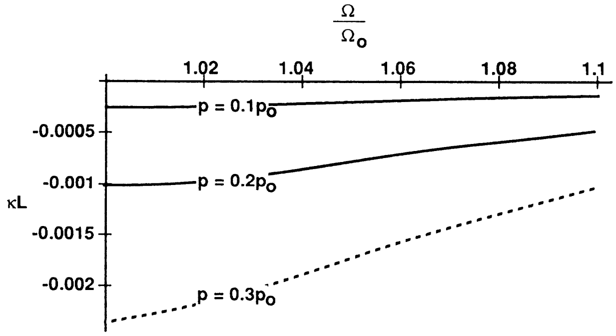

FIG. 6. Frequency dependence of the four-wave-mixing coefficient for acoustically pumped optical phase conjugation.

D. Experimental considerations

The visible and microwave experiments are markedly different and it is convenient to examine them separately.

1. Frequency shifting and deflection of visible light

Maintaining aqueous suspensions of submicron sized bubbles presents no fundamental problem and techniques to produce and maintain these suspensions are well known. The principal difficulty, which we believe will beset these experiments, is scattering. The scattering length L_S for light propagating through a microbubble suspension is

$$L_S^{-1}=f_0(kr_0)^4\epsilon_h^2[(\epsilon_r-1)/(\epsilon_r+2)]^2/r_0 \, , \tag{3.20}$$

and we require that the scattering length be large compared to the optical path length and that L_S^{-1} be small compared to the shifting parameter. Thus we require

$$L_S \gg L \, , \tag{3.21a}$$

$$f_0\epsilon_h[(\epsilon_r-1)/(\epsilon_r+2)][pR(\Omega)/p_0]^2 kL_S \gg 1 \, . \tag{3.21b}$$

For water, $\epsilon_h=1.69$, so that $\epsilon_r=0.59$ and the second condition places the following limitation on the experimental parameters:

$$pR(\Omega)/p_0 \gg 1.45(kr_0)^{1.5} \, . \tag{3.22}$$

If the acoustic pressure is on the order of 10 kPa, then $R(\Omega)$ must exceed $15(kr_0)^{1.5}$. If we use the He-Ne line at 6328 Å, then we require $r_0 < 3100$ Å, if we wish to examine resonant conditions for a bubble Q of 10. Off resonance, the suspension will have to be driven harder to compensate for the decrease in $R(\Omega)$ if frequency shifting is to dominate scattering. The scattering length for a suspension which consists of a 10^{-4} volume fraction of 3000-Å microbubbles is 2.5 cm. Finally, we note that when $\Omega = \Omega_0$ the microbubbles will resonantly absorb and scatter acoustic radiation. The formulas for phase shifting and beam deflection are still valid as the acoustic pump waves are counterpropagating and the product of the two pressure waves has equal intensity throughout the medium. More precisely, $p^2 \rightarrow p^2 e^{-2\alpha L}$, where α is the acoustic attenuation coefficient. The acoustic attenuation length is on the order of 1 mm, which should not rule out experimental investigation.

2. *Acoustically pumped optical phase conjugation*

For microwave experiments it is necessary to use nonaqueous suspensions of microparticles. Such suspensions must be stabilized by a surfactant, but well researched techniques are available. For the acoustically pumped microwave experiments, scattering of electromagnetic radiation is not an issue. However, acoustic losses will prevent any resonant experiments. This difficulty can be appreciated by noting that the attenuation length due to acoustic absorption is

$$L_A = r_0 / f_0 ,$$

and the bubble size must be on the order of 100 Å for resonant excitation at 1-cm wavelengths. Reasonable acoustic attenuation lengths would require volume fractions on the order of 10^{-7}, which reduces the efficiency to undetectable values unless either very long optical pathlengths or high microwave probe powers are employed. Instead, there are two possible alternatives: $\Omega \ll \Omega_0$ or $\Omega \gg \Omega_0$. At low frequencies, the acoustic attenuation length will increase by a factor of $(\Omega/\Omega_0)^4$ and should be suitable. At high frequencies, a smaller volume fraction of microbubbles can be employed, but the shorter wavelength might ensure sufficiently large efficiencies. We examine these two limits below.

As a specific numerical example, consider a microbubble suspension consisting of 25-Å microbubbles irradiated by ultrasonic waves with $\Omega = 10^9$ cycles/s and 100-cm microwave radiation. If the volume fraction is 10^{-4}, then the attenuation length will be 25 cm and $kL = 0.01$ if $L = 3000$ cm. The required acoustic pressure is 30 kPa. Next, we consider high frequencies. Here, we irradiate large bubbles, on the order of 1 μm, with acoustic waves such that $\Omega = 10^{11}$ cycles/s. Although the microbubbles are no longer resonant with the incident sound wave, the attenuation length is still on the order of r_0/f_0. Thus attenuation lengths on the order of 1 cm are possible if the volume fraction of microbubbles is 10^{-4}. However, much higher acoustic pressures are required to achieve meaningful changes in the bubble amplitude and detectable phase conjugate powers. In particular, $R(\Omega) = (\Omega_0/\Omega)^2$ if $\Omega \gg \Omega_0$. This requires acoustic powers in excess of 10^4 W/cm^2 and probably confines one to the low-frequency situation.

IV. CONCLUSIONS

In this paper we have developed a theory of acousto-optical four-wave-mixing processes in which control of the electromagnetic wave's phase, frequency, or propagation direction is sought. Prior research into such higher-order acousto-optical effects focused on processes that utilized electromagnetic waves as the pump beams and control was achieved of the acoustic wave's phase. These processes did not require specialized media, for two reasons: (i) electromagnetic intensities are much greater than the corresponding quantity that will appear in the processes of interest to us, i.e., the quantity A, and (ii) the acoustic wavelength sets the spatial scale over which events occurred. Due to the slow group velocity of sound waves in all media, the acoustical wavelengths are very much shorter than the corresponding optical wavelengths at a given frequency. This ensured that the various four-wave-mixing coefficients that control the efficiency of these acousto-optical processes are reasonably large so long as the coupling constant g is not too small. This situation does not prevail for processes which seek to effect the propagation characteristics of electromagnetic beams, especially at low frequencies. These issues motivate us to search for specialized media which are endowned with either very large acousto-optical coefficients or are very compressible.

As noted in Sec. II, the acousto-optical analog of the third-order susceptibility γ is on the order of 10^{-2}–10^{-4}. Furthermore, the pump amplitude A is on the order of the acoustical displacement of the material divided by a characteristic length. For most liquids and solids this is on the order of 10^{-5} or less. The mixing coefficient for any four-wave-mixing processes of interest to us is generally of the form

$$4\kappa = 2\pi k \delta A^2 ,$$

and will be of order 10^{-7} for visible light. To achieve values of kL on the order of unity would require optical path lengths of 10^7 cm. One scheme to overcome this difficulty is based on using a material which is highly compressible as the active media. As noted in Ref. 3 and in the Introduction, microbubbles are very compressible and it is quite easy to achieve fractional changes on the order of 1–10%, even with very low acoustic powers. Such large values of A lead to kL on the order of unity for optical path lengths of 1 cm or less at visible wavelengths. In the microwave region of the spectrum, much longer optical path lengths are required. However, theory indicates that such processes are at least possible on the laboratory scale.

In this paper we examined three different types of acousto-optical processes: frequency shifting and beam deflection of visible light as well as acoustically pumped optical phase conjugation at microwave wavelengths. The first two effects have well-defined first-order analogs. However, they have a number of interesting differences that are worth noting. In first order, frequency shifting and beam deflection are intimately tied together due to the fact that both processes arise from the same acoustically generated index grating. However, for AOFWM processes frequency shifting and beam deflection can arise from two different index gratings. Specifically, two counterpropagating, degenerate acoustic beams will induce two different types of index gratings in a material medium: a spatially uniform, high-frequency grating which oscillates at twice the acoustic frequency and a static grating whose spatial periodicity is twice the pump wave vector. The first grating leads to frequency shifting without beam deflection while the second gives rise to beam deflection without shifting the frequency. By an appropriate choice of beam configurations it is possible to separate these two gratings and examine them in isolation. Since these process simultaneously involve two sound waves, the frequency shift and beam deflection occur in multiples of $\pm 2\Omega$ and $\pm 2K/k$, which differ significantly from first-order effects. Furthermore, the mixing coefficient is proportional to the square of the acoustic pressure and these features can be utilized as a convenient means to separate AOFWM effects from first-order processes.

Acoustically pumped optical phase conjugation has not, to our knowledge, been proposed elsewhere. This is an interesting process as it involves the direct conversion of phonons into photons of the same frequency which is unique in acousto-optical processes. It is very unfortunate that it cannot be studied at resonant excitation, since experimental investigation would be far more convenient. Another difficulty lies in the nature of the microbubbles themselves in that they cannot support large oscillations of sufficiently high frequency to carry out microwave phase conjugation at useful wavelengths. In particular, it would be useful to have compressible artifical dielectrics whose resonance frequencies are on the order of 180 GHz. With such a system it would be possible to drive it nonresonantly at 18 GHz, which is a convenient wavelength for experiments. It might be possible to achieve such high frequencies with slightly more rigid media, such as plant cells or some other structure.

To conclude, we note that the highly compressible nature of microbubble suspensions makes them ideal for study of AOFWM processes at relatively low acoustic powers. If resonant behavior is too lossy, with respect to acoustic absorption, it is still quite possible to examine these processes off resonance.

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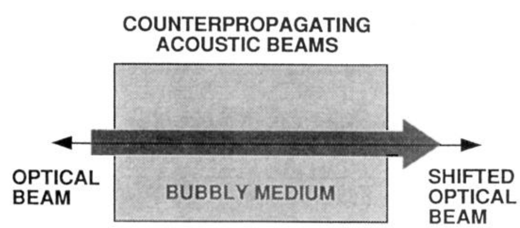

FIG. 1. Parallel beam configuration for frequency shifting of optical beams by a uniform, high-frequency index grating created by two counterpropagating sound waves.

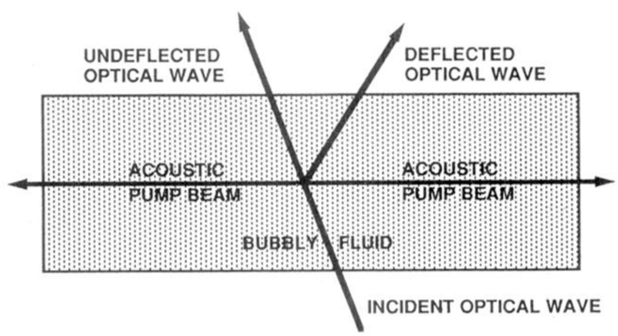

FIG. 3. Beam geometry for optical deflection via the static, spatially periodic grating generated by two counterpropagating pump waves.

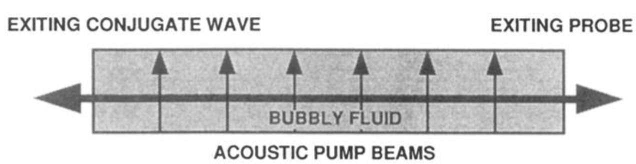

FIG. 5. Beam configuration for acousto-optical phase conjugation in a bubbly medium.