Stark-Enhanced Phase Conjugation in Shaped-Microparticle Suspensions

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We examine optical phase conjugation for a shaped-microparticle suspension maintained in a uniform electric field. Theory asserts that the presence of a Stark field decreases the grating response time and enhances the composite's four-wave-mixing coefficient. These changes arise from and reflect the narrow angular range in which the particles are constrained to point, giving rise to reduced rotational times and the appearance of a new coherent scattering mechanism.

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This Letter examines optical phase conjugation in a shaped-microparticle suspension maintained in a uniform, static electric field. These investigations are motivated by two recent experimental studies in which nonlinear optical birefringence[1,2] and optical phase conjugation[3,4] were observed in shaped-microparticle suspensions at microwave (18 GHz) and visible (0.5 μm) wavelengths. Since anisotropy is the mechanism responsible for the nonlinear optical properties of these media,[5] it is logical to inquire how these properties are influenced by an electric field used to orient a majority of the particles along a particular direction. Theory asserts that a strong electric field will vastly alter optical phase conjugation in shaped-particle suspensions. Four novel effects are predicted to occur: (1) A new scattering mechanism appears which contributes to optical phase conjugation, (2) the four-wave-mixing coefficient associated with the orientational grating is measurably altered, (3) the degeneracy between rods and disks is split in the sense that their nonlinear coefficients are no longer equal, and (4) the grating response time is reduced.

These effects arise from and reflect the fact that an intense Stark field will restrict the microparticles to a relatively narrow range of angles which alters the macroscopic properties of the suspension in a number of striking ways. For example, in the presence of a sufficiently intense electric field, grating formation requires that the microparticles rotate through much smaller angles than in its absence, thereby reducing response times in shaped-particle suspensions. Since particle orientation is confined to within a small range of angles about a particular direction (determined by the electric field), their individual dipole moments will tend to be in phase and add coherently. In contrast, if only weak radiation fields are present, the microparticle distribution is nearly isotropic and most of the particles are randomly aligned, giving rise to a much smaller net induced dipole moment than that displayed by a highly oriented suspension. This strong anisotropy in the microparticle orientation gives rise to a new scattering mechanism which can enhance the magnitude of the original effect. Finally, the potential minima for disks and rods differ not only in angle, but also in magnitude, which manifests itself in the medium's electrodynamics when the particle distribution becomes noticeably anisotropic.

Light-scattering experiments[6] find that a Stark field will modify the scattering attenuation coefficient, increasing (decreasing) it for radiation polarized parallel (perpendicular) to the electric field. These findings suggest that the four-wave-mixing coefficient will exhibit the same behavior. Specifically, recent studies[7] of thermal light-scattering noise establish a direct relationship between the scattering attenuation and the optical Kerr coefficients with an increase in one implying a corresponding increase in the other. Thus, we expect that if the radiation fields are polarized parallel (perpendicular) to the Stark field, the four-wave-mixing coefficient will increase (decrease).

A monodisperse suspension of shaped microparticles is maintained in a uniform, static electric field $\mathbf{E}_S$ and irradiated by three degenerate coherent radiation beams [denoted by $\mathbf{E}_R(r,t)$]: two counterpropagating pump waves and a probe beam. These fields polarize the microparticles and induce both static and dynamic dipole moments which interact with these fields, giving rise to two electrostrictive interactions:

$$U(\mathbf{r};\Omega)=U_S(\Omega)+U_R(\mathbf{r};\Omega)=-\tfrac{1}{2}[\mathbf{E}_S\cdot\boldsymbol{\alpha}(0)\cdot\mathbf{E}_S+\langle\mathbf{E}_R(\mathbf{r},t)\cdot\boldsymbol{\alpha}(\omega)\cdot\mathbf{E}_R(\mathbf{r},t)\rangle]. \quad (1)$$

Here $\Omega=(\theta,\phi)$ specifies the particle orientation relative to $\mathbf{E}_S$, ω is the radiation frequency, and $\boldsymbol{\alpha}(\omega)$ is the microparticle polarizability tensor,

$$\boldsymbol{\alpha}(\omega)=\alpha_S(\omega)\hat{\mathbf{I}}+\beta(\omega)\hat{\mathbf{K}}(\Omega)/3. \quad (2)$$

In Eq. (2), $\alpha_S=(\alpha_\parallel+2\alpha_\perp)/3$, $\beta=\alpha_\parallel-\alpha_\perp$, $\hat{\mathbf{I}}$ is the unit matrix, $\hat{\mathbf{K}}(\Omega)$ is the standard orientation matrix, and $\alpha_\parallel$ ($\alpha_\perp$) is the component of the particle polarizability parallel (perpendicular) to the symmetry axis. The angular brackets imply an average over a time long compared to $1/\omega$, but short compared to the suspension's response time.

We confine ourselves to situations in which all of the radiation fields have the same polarization, $\hat{e}_R$,

$$\mathbf{E}_R(\mathbf{r},t) = \hat{e}_R \sum E_j(\mathbf{r}) \exp[i(\mathbf{K}_j \cdot \mathbf{r} - \omega t)] + \text{c.c.}, \quad (3)$$

where $E_j(\mathbf{r})$ is the complex amplitude of the jth beam, $\mathbf{K}_1 = -\mathbf{K}_2 \equiv \mathbf{K}$, $\mathbf{K}_p = -\mathbf{K}_c \equiv \mathbf{Q}$, $j=(1,2)$ refers to the two counterpropagating pumps of equal intensities, $j=p$ to the probe, and $j=c$ to the conjugate wave.

An examination of Eqs. (1)–(3) reveals that $\mathbf{E}_R(\mathbf{r},t)$ induces microparticle density and orientational gratings whose physical characteristics are controlled by the interference patterns created by the different possible sets of fields. Here, we confine ourselves to situations in which $U_R/kT \ll 1$, but U_S/kT is arbitrary. In steady state, the microparticle density is given by the Boltzmann distribution, approximated via

$$n(r,\Omega) = n_0[1 - U_R(r,\Omega)/kT] \exp[-U_S(\theta)/kT] Z$$
$$\equiv n_0(r,\Omega) + \delta n_c(r,\Omega), \quad (4)$$

with Z the partition function and n_0 the zero-field particle density. The exponential factor tends to confine the particles to a narrow range of angles dictated by $\mathbf{E}_S$. Phase conjugation arises from the term proportional to U_R, i.e., δn_c, which induces two sets of mutually orthogonal index gratings: translational and orientational. The translational gratings, which emanate from the isotropic component of the particle polarizability, consist of a modulation in the density. The orientational gratings, which derive from the anisotropic component, consist of a modulation in the direction that the particles point. Clearly the translational gratings are unaffected by a spatially uniform electric field and we may set $U_S(\theta)/kT = -g_S(\cos^2\theta - 1/3)$ with $g_S = \beta E_S^2/2kT$. However, for orientational gratings, the situation is quite different as the microparticles are aligned by $\mathbf{E}_S$. This Stark-induced alignment alters the structure of the orientational gratings which now consist of a periodic tightening and easing of the particle orientation about a preferred angle determined by $\mathbf{E}_S$.

The nonlinear polarization responsible for optical phase conjugation is given by the orientational average of the induced dipole $\boldsymbol{\alpha}(\omega) \cdot [\mathbf{E}_1(\mathbf{r},t) + \mathbf{E}_2(\mathbf{r},t)]$ over the microparticle distribution $\delta n_c(\mathbf{r},\Omega)$,

$$p^{\mu}_{\text{NL}}(\mathbf{r},t) = P_\mu \exp[i(\mathbf{Q} \cdot \mathbf{r} + \omega t)] + \text{c.c.}, \quad (5a)$$

$$P_\parallel = -4\pi(\omega/c)^2 n_0[\alpha g_1 + (\alpha g_2 + \beta g_1/3) R_1(g_S) + \beta g_2 R_2(g_S)/3], \quad (5b)$$

$$P_\perp = -4\pi(\omega/c)^2 n_0[\alpha g_1 - (\alpha g_2 + \beta g_1/3) R_1(g_S)/2 + \beta g_2 R_2(g_S)/3], \quad (5c)$$

where $\parallel$ ($\perp$) implies that the radiation fields are polarized parallel (perpendicular) to the Stark field, $g_1 = \alpha E_p E_1/2kT$, and $g_2 = \beta E_p E_1/2kT$. The functions $R_1(g_S)$ and $R_2(g_S)$ are angular integrals of the microparticle polarizability over δn_c. The first term on the right-hand side of Eq. (5a) is a nonlinear polarization consisting of a translational grating of isotropic dipoles and is unaffected by $\mathbf{E}_S$. The next two terms arise from a new coherent scattering mechanism which is due to the anisotropic orientational distribution induced by the Stark field. The term $\alpha g_2 R_1(g_S)$ originates from an orientational grating of particles, whose induced dipole moment arises from the interaction of the radiation fields with the isotropic component of the particle polarizability tensor. The second term, i.e., $\beta g_1 R_1(g_S)/3$, consists of a translational grating of microparticles, whose induced dipole moment is due to the interaction of the anisotropic component of the particle polarizability tensor with the radiation. On physical grounds, one expects these contributions to vanish as they depend upon a unique direction and in fact $R_1(g_S) \to 4g_S/45$ as $g_S \to 0$. In the opposite limit, i.e., $g_S \to \pm\infty$, $R_1(g_S) \to \frac{2}{3} - 1/g_S$ for rods and $R_1(g_S) \to -\frac{1}{3} + 0.5/g_S$ for disks. The difference in both sign and magnitude of the induced dipole moment, as characterized by $R_1(g_S)$ reflects the orientation of the microparticle relative to the Stark field. Specifically, rods (disks) tend to orient parallel (perpendicular) to $\mathbf{E}_S$ and $\langle\cos^2\theta\rangle \to 1$ (0) in this limit. Note that these two contributions, although equal in magnitude, exhibit quite different optical response times as one requires an orientational grating while the other demands a translational grating. Finally, analogs of these terms appear in light-scattering experiments in which the shaped-particle suspension is maintained in a Stark field.[6]

The last term in the nonlinear polarization, $\beta g_2 R_2 \times (g_S)/3$, consists of an orientational grating of shaped microparticles which is always present. In the limit that the electric field vanishes, $R_2(g_S) \to \frac{4}{45} + 16g_S/945$. As the field is made more intense, $R_2(g_S) \to \frac{4}{9} - 1.33/g_S$ for rods and $R_2(g_S) \to \frac{1}{9} - 0.332/g_S$ for disks: The orientational gratings are now composed of microparticles which deviate slightly from the direction preferred by the Stark field.

Inserting the nonlinear polarization into the Maxwell equations and making the slowly varying envelope approximation, we find for steady state the usual expression for the intensity of the emitted conjugate wave.[8] If the radiation is polarized parallel to $\mathbf{E}_S$, the four-wave-mixing coefficient is given by

$$\kappa_\parallel = 16\pi^2 K n_0[\alpha^2/kT + \alpha\beta R_1(g_S)/3kT + \beta^2 R_2(g_S)/9kT] I/c\varepsilon_h, \quad (6a)$$

whereas if $\hat{e}_R$ is perpendicular to $\mathbf{E}_S$,

$$\kappa_\perp = 16\pi^2 K n_0[\alpha^2/kT - \alpha\beta R_1(g_S)/6kT + \beta^2 R_2(g_S)/9kT] I/c\varepsilon_h, \quad (6b)$$

with I the pump intensity and ε_h the dielectric constant of the host fluid. The different response times of the orientational and translational gratings can serve as a

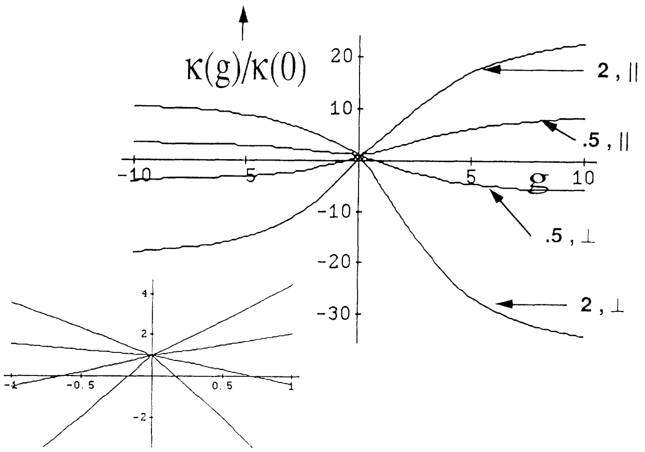

FIG. 1. $\kappa_0^{(\mu)}(g_S)/\kappa_0$ vs g_S for $\alpha/\beta=0.5$ and $\alpha/\beta=2$ for both polarization cases.

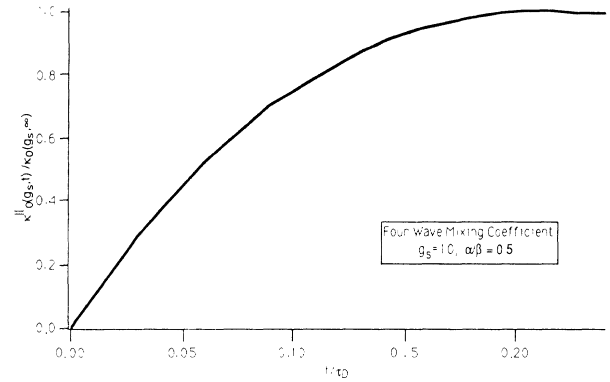

FIG. 2. Transient evolution of the four-wave-mixing coefficient due to the orientational gratings for the case $g_S=10$, $\hat{\mathbf{e}}_R$ parallel to $\mathbf{E}_S$, and $\alpha/\beta=0.5$.

convenient means to isolate the rotational mechanism. For example, at 18 GHz it takes so long for the particles to diffuse a grating spacing that the translational grating is never formed.[2] At visible wavelengths, the translational grating can be suppressed by using an optical chopper.[4] The dramatic influence of a Stark field on optical phase conjugation is illustrated by the ratio $\kappa_0^{(\mu)}(g_S)/\kappa_0(0)$, where $\kappa_0^{\mu}(g_S)$ is the contribution of the orientational grating and μ refers to the polarization of the radiation fields. Figure 1 depicts this ratio for $\alpha/\beta=0.5$ and $\alpha/\beta=2$ vs g_S. The limiting value for rods is $\kappa_0^{\parallel}(g_S)/\kappa_0(0)\to 5+45\alpha/4\beta$ while for disks $\kappa_0^{\parallel}(g_S)/\kappa_0(0)\to \frac{5}{4}-45\alpha/8\beta$. For the suspensions used in Ref. 1, the value of $\alpha/\beta\simeq 0.5$ so that the four-wave-mixing coefficient should increase by a factor of 10.625, which can be achieved with a field strength of 300 V/cm. For the suspension used in Ref. 2, $\alpha/\beta\simeq 2$, so that the four-wave-mixing coefficient should increase by a factor of 27.5 for an electric field strength of 3 kV/cm. Note that weak Stark fields decrease the four-wave-mixing coefficient if $\hat{\mathbf{e}}_R$ is perpendicular to $\mathbf{E}_S$.

Next, we focus on the transient behavior of the suspension for situations in which the microparticle orientation is initially in equilibrium with the Stark field and the radiation fields are switched on at $t=0$. On physical grounds, we anticipate that the orientational response time will decrease substantially since the microparticles need to rotate through much smaller angles to achieve a new equilibrium state. The transient dynamics of the orientational distribution $n(\Omega,t)$ is governed by the Planck-Nernst equation,[5]

$$\partial n(\Omega,t)/\partial t=-\Theta_0\{L^2 n(\Omega,t)-L[\Gamma n(\Omega,t)/kT]\}, \quad (7)$$

where L is the angular momentum operator, Θ_0 is the rotational diffusion coefficient, and $\Gamma=-LU(r,\Omega)$ is the electrostrictive torque exerted by all the fields on the particles. Figure 2 depicts the evolution to steady state of $\kappa_0^{\parallel}(g_S;t)/\kappa_0^{\parallel}(g_S;\infty)$ for the case $g_S=10$, with the suspension initially in equilibrium with the Stark field and $\alpha/\beta=0.5$. Note that the four-wave-mixing coefficient evolves on a time scale set by $3\tau_D/g_S$, where τ_D is the orientational diffusion time. For the suspension employed in Ref. 1, the response time would be reduced by a factor of 10 to a few seconds with a field strength of 300 V/cm, whereas that used in Ref. 2 would be reduced to a few hundred μs with a Stark field of 2 kV/cm.

In summary, by confining the particles to a narrow angular range, the medium's nonlinear response to coherent radiation can be measurably enhanced while its response time is reduced. Other approaches for achieving a highly anisotropic microparticle orientation are by intense, polarized coherent radiation and by flowing the suspension in a cell, without turbulence. Similar effects should also occur in self-focusing, the optical Kerr effect, and coherent beam combination[9] which will be examined elsewhere.

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