Active Optomechanical Media for Nonlinear Microwave Processes

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Abstract—**Theory asserts that three-dimensional arrays of electrically small particles are excellent media for wave-mixing processes at microwave and millimeter wavelengths. As a specific example, spheres that are free to move on a stack of flat, transparent surfaces and that interact with incident radiation are examined. Electrostrictive forces move the spheres in such a way as to form density index gratings that can be used for controlling the propagation characteristics of coherent radiation. Phase conjugation in this medium is also examined.**

In this letter, we examine the optical and dynamical properties of three-dimensional arrays of electrically small particles that are free to move under the action of electromagnetic forces. We refer to such materials as active optomechanical media and show that they are eminently suitable for such wave-mixing processes as phase conjugation [1] at microwave and millimeter wavelengths. The four wave-mixing coefficients and optical response times for generating phase conjugate radiation are determined for an optomechanical medium composed of a three-dimensional array of spheres that are free to roll on flat, transparent planes to form density index gratings.

Optomechanical media constitute a special class of artificial Kerr media [2] whose properties appear to differ substantially from those of microparticle suspensions [3], [4]. Theory asserts that optomechanical media have much faster response times and larger nonlinear wave-mixing coefficients than those of comparable microparticle suspensions. However, optomechanical media can be understood as the limiting case of a microparticle suspension operating in the fully saturated regime.

For simpicity, we consider a three-dimensional array of identical spheres of radius r_0, free to roll on a stack of flat, transparent planes and irradiated by three coherent beams whose wavelengths are much greater than r_0. The volume fraction of spheres is $f = 4\pi r_0^3 n_0/3$, n_0 is the sphere density and the sphere's polarizability is α. Let $\boldsymbol{E}(\boldsymbol{r}, t)$ be the electric field of the incident radiation fields

$$\boldsymbol{E}(\boldsymbol{r}, t) = \sum_{m=1,2,p,pc} \boldsymbol{e}_m E_m \exp\left[i(\boldsymbol{K}_m \cdot \boldsymbol{r} - \omega_m t)\right] + cc \tag{1}$$

where $\boldsymbol{e}_m$, E_m, $\boldsymbol{K}_m$ and ω_m are the unit polarization vector, complex amplitude, propagation vector and frequency of the mth beam. Here, $m = 1$ (2) corresponds to the pump beam propagating to the right (left), $m = p$ refers to the probe wave and $m = pc$ corresponds to the wave that is phase conjugate to the probe. For standard phase conjugation, $\boldsymbol{K}_1 = -\boldsymbol{K}_2 \equiv \boldsymbol{K}$; $\boldsymbol{K}_p = -\boldsymbol{K}_{pc} \equiv \boldsymbol{Q}$ and all of the frequencies are degenerate, i.e., $\omega_m \equiv \omega$.

In the presence of radiation, each sphere acquires an induced electric dipole moment $\boldsymbol{p}(\boldsymbol{r}, t) = \alpha \boldsymbol{E}(\boldsymbol{r}, t)$ that, in turn, couples to the incident radiation via an electrostrictive coupling $U(\boldsymbol{r}, t) = -\frac{1}{2}\alpha\langle E^2(\boldsymbol{r}, t)\rangle$. Here, the angular brackets imply a time average that is long compared to the optical period, but short compared to the medium response time. The total polarization vector $\boldsymbol{P}(\boldsymbol{r}, t) = \alpha\rho(\boldsymbol{r}, t)\boldsymbol{E}(\boldsymbol{r}, t)$, where $\rho(\boldsymbol{r}, t)$ is the number density of spheres.

Suppose all of the beams are linearly polarized, with the pump waves orthogonally polarized with respect to one another and the probe beam polarized parallel to the $m = 1$ pump wave. Then $U(\boldsymbol{r}) = -\frac{1}{2}\alpha E_p E_1 \cos[(\boldsymbol{K} - \boldsymbol{Q}) \cdot \boldsymbol{r}]$, which gives rise to an electrostrictive force $\boldsymbol{F}(\boldsymbol{r}, t) = -\nabla U(\boldsymbol{r}, t)$ that moves the spheres to the points on the planes, denoted by (x_j, y_l) where $U(\boldsymbol{r}, t)$ is minimum. The z points are set by the plane positions, z_p, and the equilibrium sphere density can be approximated as

$$\rho(\boldsymbol{r}, t) = N \sum_j \sum_l \sum_p \delta(x - x_j)\delta(y - y_l)\delta(z - z_p), \tag{2}$$

where N is the total number of spheres.

The phase conjugate wave is produced via diffraction of the $m = 2$ pump beam by the first-order index grating: $\cos[(\boldsymbol{K} - \boldsymbol{Q}) \cdot \boldsymbol{r}]$. Thus, we spectrally decompose $\rho(\boldsymbol{r}, t)$ into grating components

$$\rho(\boldsymbol{r}, t) = \sum_{k=0}^{\infty} A_k \cos\left[k(\boldsymbol{K} - \boldsymbol{Q}) \cdot \boldsymbol{r}\right]. \tag{3}$$

For simplicity, all of the propagation vectors are assumed to lie in the xy plane and the x axis is defined by the grating wave vector $\boldsymbol{K} - \boldsymbol{Q}$. Once steady-state is established, the spheres will be located at the points $x_j = j\pi/K\,|\cos(\gamma/2)|$, $j = 0, \pm 1, \pm 2, \cdots$ with γ the angle between $\boldsymbol{K}$ and $\boldsymbol{Q}$. The depth of the different translational gratings is set by $A_k = n_0(1 + \delta_{k0})$ which is independent of beam power. This statement is valid provided the electrostrictive energy per sphere is large compared to $\frac{1}{2}k_B T$, the thermal energy per sphere. Accordingly, unless the incident beam intensities are very low the medium is saturated.

Manuscript received August 8, 1991.
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IEEE Log Number 9104371.

1051-8207/91$01.00

The polarization vector that gives rise to phase conjugation, $\boldsymbol{P}_{pc}(\boldsymbol{r}, t) = n_0 \alpha E_2 \boldsymbol{e}_2 \exp[i(\boldsymbol{Q} \cdot \boldsymbol{r} + \omega t)] + \text{cc}$. To determine the phase conjugate reflectively, we insert $\boldsymbol{P}_{pc}(\boldsymbol{r}, t)$ into the Maxwell equations, make the slowly varying envelope approximation (SVEA) and solve for the conjugate wave. In the nondepleted pump approximation, the intensity of the exiting conjugate wave, $I_{pc} = I_2[\kappa L]^2$, where $\kappa L = 2\pi K L \alpha n_0$ with I_2 the intensity of the $m = 2$ pump beam. For metallic particles, $\alpha = r_0^3$, $\kappa L = 6\pi f L/\lambda$ and the four-wave mixing coefficient is independent of pump power as one would expect for a nonlinear medium operating in the saturation regime.

In addition to generating the phase conjugate beam, the medium will also amplify the initial probe wave as it propagates through the optical index grating created by the E_{pc} and the $m = 1$ pump beam. The depth of this grating is sufficiently deep throughout the bulk of the nonlinear medium such that the polarization vector that gives rise to probe beam amplification can be approximated as $\boldsymbol{P}_p(\boldsymbol{r}, t) = n_0 \alpha E_1 \boldsymbol{e}_1 \exp[i(\boldsymbol{Q} \cdot \boldsymbol{r} - \omega t)] + cc$. The intensity of the exiting probe beam is $I_p(L) = I_p(0) + 2\kappa L[I_1 I_p(0)]^{1/2} + [\kappa L]^2 I_1$.

If the volume fraction of spheres is 5×10^{-4} and $L/\lambda = 30$, then $\kappa L = 0.28$. If the beam intensities are $I_1 = I_2 = 1$ W/cm^2 and $I_p = 1$ μW/cm^2, then the phase conjugate beam intensity will be 8% of the pump beam or 80 mW/cm^2. The intensity of the exiting probe beam will be 80.5 mW/cm^2. In general, we require $L/\lambda \approx 30$, and volume fractions on the order of 5×10^{-4} are required. For 100 μm sized spheres, this implies a sphere density of 125 particles/cm^3. For 30 GHz radiation the required system size is on the order of 30 cm; implying $N = 3.4 \times 10^6$. For 94 GHz radiation, the required device size is on the order of three centimeters, which implies $N = 1.25 \times 10^5$.

These results can also be extracted from a statistical mechanical analysis using the Maxwell–Boltzmann distribution for the sphere density and taking the limit $U/k_B T \to \infty$

$$\rho(\boldsymbol{r}, t) = n_0 \frac{\exp\left[-\dfrac{U(\boldsymbol{r}, t)}{k_B T}\right]}{I_0(g)} = n_0 \left(1 + 2 \sum_{k=1}^{\infty} \frac{I_k(g)}{I_0(g)} \cos[k(\boldsymbol{K} - \boldsymbol{Q}) \cdot \boldsymbol{r}]\right). \quad (4)$$

Here, $I_k(z)$ is the kth-order modified Bessel function of argument z, $g \equiv 4\pi\alpha(I_1 I_p)^{1/2}/ck_B T$, where I_1 and I_p are the intensities of the $m = 1$ pump and probe beams. In the limit $g \to \infty$, $I_k(g)/I_0(g) \to 1$, in agreement with A_k in (3). For 100-μm metallic spheres at room temperatures, $g \approx 10^5 (I_1 I_p)^{1/2}$, and for $I_1 = 1$ W/cm^2 and $I_p = 10$ nW/cm^2, $g \approx 10$ and a mechanical description is adequate. For smaller spheres, say on the order of 10 μm, the intensity of the probe beam would have to be increased to 10 mW/cm^2 for the present mechanical description to be valid.

Next, we determine the medium response time. If the spheres were immersed in a viscous fluid, their dynamics would be described by the Planck–Nernst equation. However, as they are sited in air, the Planck–Nernst equation is inappropriate and their motion is determined by classical mechanics. In particular, the correct description is that of a rolling sphere in contact with a flat surface and subject to an electrostrictive force, a reaction force and friction from the flat surface and air. The equation of motion for a given sphere are

$$\frac{dV}{dt} = -\frac{5}{7m}\left(\nabla U(\boldsymbol{r}, t) + \mu_R m g \frac{V}{V}\right) - \frac{52\pi}{7m} \eta a V + \Gamma(t), \quad (5)$$

where m is the sphere's mass, μ_R the coefficient of rolling friction, η the dynamical viscosity of air ($1.8 \times 10^{-4} p$), V the sphere velocity and $\Gamma(t)$ is the fluctuating Langevin force that is associated with the dissipation due to air friction. The first term on the rhs of (5) arises from the electrostrictive force, the second is due to rolling friction, while the third term is a consequence of air friction. Typically, μ_R is negligible, i.e., $\mu_R \approx 0.1\%$.

The acceleration that a Copper sphere experiences is independent of its size and for one Watt beams is 0.18 cm/s^2, i.e., ≈ 180 μgravities. The total force on a 100 μm sized Copper ($\rho_m = 8.96$ g/cm^3) sphere is on the order of 6.76 μdynes. The equation of motion for an individual microsphere rolling on a perfectly flat plane ($\mu_R = 0$) under the action of the forces previously discussed is

$$\ddot{\xi} + 2\beta\dot{\xi} + \omega_0^2 \sin\xi = N(t) \quad (6)$$

Here, $\omega_0 \equiv (2\pi/\Lambda)(15[I_p I_1]^{1/2}/7\rho_m c)^{1/2}$, $\beta = 39\eta/14\rho_m r_0^2$, $\xi \equiv (\boldsymbol{K} - \boldsymbol{Q}) \cdot \boldsymbol{r}$, the grating spacing $\Lambda = |\boldsymbol{K} - \boldsymbol{Q}|/2\pi$, ρ_{m} is the sphere's mass density and $N(t) = (\boldsymbol{K} - \boldsymbol{Q}) \cdot \Gamma(t)$. In particular $\langle N(t)\rangle = 0$ and $\langle N(t)N(t')\rangle = 2kT\beta\delta(t - t')/m$. For situations of interest to us, the motion is dominated by the electrostrictive force and dissipation arising from friction with the air.

Examination of (6) reveals two regimes of interest: 1) an oscillatory regime and 2) an overdamped regime. The particular regime the medium is in depends on the relative sizes of ω_0 and β. In the oscillatory regime, where $\omega_0 \gg \beta$, the time it takes for the medium to achieve steady-state (τ_R) is β^{-1}, while ω_0 is the resonant frequency. Note that in the absence of dissipation, i.e., $\beta = 0$, the sphere's never achieve steady-state and if they were initially randomly positioned, a coherent density grating will never form.

In the heavily overdamped regime where $\omega_0 \ll \beta$, $\tau_R^{-1} = \omega_0^2/2\beta \equiv (2\pi/\Lambda)^2[5\alpha E_1 E_p/104\pi\eta r_0]$. In this regime, (7) can be solved analytically

$$\sin[\xi(t)] = \sin[\xi(0)] \cdot \frac{(1 - \cos[\xi(0)])e^{-t/\tau_R}}{\sin^2[\xi(0)] + [(1 - \cos[\xi(0)])e^{-t/\tau_R}]^2}. \quad (7)$$

An examination of (7) shows that the particle rolls from its initial position $\xi(0)$ into its equilibrium position on a time scale set by τ_R. Note that the inverse response time in (7) can be written as $1/\tau_R \approx D'(2\pi/\Lambda)^2(U/k_B T)$, with $D' = 5k_B T/104\pi\eta r_0$ the translational diffusion coefficient for a sphere moving in a medium with a viscosity η. This should be contrasted with a suspension in the strong field regime, where the medium response time [5], [6] $\tau_R^{-1} = DK^2(U/k_BT)$, with $D = 6\pi k_BT/r_0\eta$ being the translational diffusion coefficient for a sphere in a viscous fluid. At 94 GHz, with 2 W/cm^2 rms beam intensities and 100 μm Copper spheres, $\tau_R = 4.5$s. For 18 GHz radiation and the same beam powers, the medium response time will be on the order of 120s. The optical response time for the formation of a translational grating in a carbon fiber microparticle suspension [1] at these wavelengths and particle sizes is on the order of several hours to a few days. Finally, the medium response time scales inversely with air pressure through the dynamic viscosity of the air. If the phase conjugator is enclosed and the air pressure is reduced to 10 torr; the grating formation times will be reduced by a factor of 0.0132 to 0.06s and 1.6s, respectively.

To summarize, we have examined the nonlinear optical and dynamical characteristics of a new class of materials that can be utilized as the active media for such wave mixing processes as phase conjugation at microwave and millimeter wavelengths. The specific materials considered are composed of three-dimensional arrays of spheres that are free to roll on transparent, flat planes. Grating formation times are typically on the order of a second and in the saturated regime the emitted phase conjugate power is on the order of a few percent of the pump power. Future studies will focus on using anisotropic media and will consider other nonlinear processes.

Acknowledgment

The authors would like to acknowledge useful and interesting discussions with G. Freeman, R. McGraw, and H. Fetterman.

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